# Proceedings of the 17<sup>th</sup> Meeting of Japan CF Research Society JCF17

March 19-20, 2017 National Institute of Technology, Tokyo College

Japan CF-Research Society

**Edited by Shinya Narita** 

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#### PREFACE

This is the proceedings of the 17<sup>th</sup> Meeting of Japan CF-Research Society (JCF17), which was held at National Institute of Technology, Tokyo College on March 19 - 20, 2017. In this meeting, 16 presentations were given and 8 papers were submitted to the editorial board. They have been peer reviewed by the referees, and revised for the publication as the proceedings. In addition, an author was permitted to make contribution as "A note without reviewing".

For all meetings, JCF1 through JCF16, we published the Proceedings. For the meetings after JCF4, we published electronic versions of the proceedings on our web-site http://jcfrs.org/proc\_jcf.html in addition to their printed versions. In view of low efficiency and low effectiveness in distributing information, we decided to discontinue the printed version for the meetings, JCF12. Only the electronic versions have been published thereafter. Needless to say, any responses from the world scientists will be welcomed.

Finally, we would like to thank all the participants and the people who have collaborated in organizing this meeting.

Editor-in-Chief Shinya Narita, Iwate University October 2017

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# Heat evolution from silica-supported nano-composite samples under exposure to hydrogen isotope gas

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*Abstract* Hydrogen isotope absorption characteristics of nanoparticles supported by silica, Pd/SiO<sub>2</sub> ("PSf1") and CuNi<sub>10</sub>/SiO<sub>2</sub> ("CNS3"), have been examined. Large absorption energy  $(1.3\pm0.3 \text{ eV/Pd})$  with large apparent loading ratio ( $L_M = 2.6$ ) was observed in the initial phase of the D-PSf1#1 run with D<sub>2</sub> at room temperature, which could be ascribed to reduction of PdO and hydrogen absorption by Pd nanoparticles. To reduce the NiO in the CNS3 sample, heating up to around 200 °C was necessary. The excess heat was observed in the elevated temperature phases of the runs with CNS3, while no excess heat was observed with PSf1. Taking also into account the experimental results obtained previously for other samples, we can conclude that the excess heat is observed only in the elevated temperature phases of the runs with binary nanocomposite samples, but not with single-element nanoparticles. In the H-CNS3#2 run, the excess heat amounts to 200 eV/Ni or more than 0.9 keV/H without detectable dose rate of hard radiations, which cannot be explained by any chemical process.

*Index Terms* – Silica supported nanoparticles, Cu·Ni/SiO<sub>2</sub>, Pd/SiO<sub>2</sub>, hydrogen gas absorption, excess power, 200 eV/Ni, 0.9 keV/H.

#### I. INTRODUCTION

There have been increasing interests in experiments of hydrogen-gas charged nickel-based nano-composite samples for excess power generation, owing to higher availability of nickel than palladium. A Ni·Cu·Mn alloy thin wire, for example, has been examined extensively by Celani et al. <sup>[1]</sup>. In addition, a number of entrepreneurs are publicizing their own "products" of nano-fabricated samples on web sites with undisclosed details, and therefore with little scientific corroboration <sup>[e.g., 2-3]</sup>. Among them, replication experiments of the Rossi-type reactors have been performed by several researchers <sup>[4-7]</sup>, which seemingly appears to show unignorable reproducibility of the Rossi method. However, little is known about the accuracy of the calorimetry and the mechanism of the claimed anomalously large energy production.

As reviewed in ref. [8], the 8 year-long (2008-2015) series of study on anomalous heat effects by interaction of metal nanoparticles and D(H)-gas under the collaboration of Technova Inc. and Kobe University has become the basis for the present collaborative research of a new  $CO_2$  free energy source. This new project on New Metal-Hydrogen Energy was started on October 2015 under the collaboration of six Japanese organizations, one of which the individual author of the present paper belongs to. The first report of the results by the project was done in ICCF20<sup>[9]</sup>.

In the present work, hydrogen isotope absorption by nickel-based silica-supported nanocomposite samples has been examined as the collaborative work using the experimental apparatus installed at Kobe University <sup>[8 - 13]</sup>. The system has a reaction chamber containing the sample with a capacity of 500 cc, and a flow-calorimetry system capable of working at elevated temperatures up to 300 °C with use of a liquid hydrocarbon coolant. The samples tested so far include silica-included Pd nanoparticles ("PSf1") fabricated in Kyushu University and mesoporous-silica-supported CuNi<sub>10</sub> nanoparticles ("CNS3") synthesized in Kobe University, whose D(H)-absorption/heat-generation characteristics are discussed in the present paper.

#### II. EXPERIMENTAL PROCEDURE AND SAMPLES

The PSf1 sample consists of Pd nanoparticles embedded in silica balls with diameter of several tens of nm. The detailed description of this sample will be published elsewhere. The CNS3 sample consists of CuNi<sub>10</sub> nanoparticles supported by the mesoporous silica (mp-silica) PC700G fabricated by Admatechs Co. Ltd. The sample was synthesized from a solution of nickel chloride and copper chloride containing the mp-silica powder as a suspended material to adsorb Ni and Cu in mesoscopic pores. After filtration, the mp-silica was annealed at 800 °C for 3 hrs. Each sample containing 8.4-g-Pd (PSf1) or 1.2-g-Cu and 11.4-g-Ni (CNS3) occupied the 500-cc volume of the reaction chamber (RC) without any filler.

The specifications of the samples are tabulated in Table 1. Those of the similar sample CNS2<sup>[11,12]</sup> are also shown in the figure for comparison. The particle size was visually observed in STEM-EDS photos. In the case of the CNS samples, the diameter of the Ni and Cu particles are in the range of  $5 \sim 100$  nm. It seems that the larger diameter particles are sticking on the outer surface of the mp-silica blocks. The Ni and Cu atoms appear to be included in the same pores of the mp-silica, or contained in the larger diameter particles with a density ratio approximately equal to the mixing ratio.

	Silica-coated Pd nanoparticle; PSf1		Mesoporous-silica- supported CuNi nanoparticle; CNS3		Mesoporous-silica- supported CuNi; CNS2 (for reference)		
Particle size	20.8 nm (average)		10 ~ 100 nm		5 ~ 20 nm		
Fabricated at;	Kyush	u Univ.	Technova-Kobe U.		Technova-Kobe U.		
Tested on;	Septov.	t. – N 2016 Nov. 2016 – Feb. 2017 Aug. –		Nov. 2016 – Feb. 2017		Oct. 2014	
	Mass (g)	Number of moles	Mass (g)	Number of moles	Mass (g)	Number of moles	
Amount	115.37		150.50		160.33		
0	1.27	0.079	3.40	0.21	3.76	0.24	
Ni			11.36	0.19	12.07	0.21	
Cu			1.22	0.02	1.89	0.03	
Pd	8.42	0.079					
Zr							
SiO <sub>2</sub>	105.68	1.76	134.52	2.24	146.38	2.44	

Table 1. Specifications of the samples tested.



Fig. 1. Schematic of C<sub>1</sub> absorption system equipped with oil-flowcalorimeter system with flow-rate-monitors and dual heaters.

A schematic of the absorption-calorimetry system C<sub>1</sub> is shown in Fig. 1. Refer to the references [9, 11 - 13] for detailed description of the system. Calibration of the flow calorimetry with a flow rate of 20 cc/min was performed using the bare mp-silica powder. The heat conversion coefficient from the power to the oil-outlet temperature  $T_{C2}$  at TC2,  $dT_{C2}/dW = 1.65$  °C/W or 1.16 °C/W, was obtained at room temperature (R.T.) or in the temperature range from 200 – 300 °C, respectively. The heat recovery rate, 0.88 – 0.79 in the same temperature range, was calculated by

$$R_{\rm h} = F^{\alpha} \cdot \rho \cdot C \cdot (T_{\rm C2} - T_{\rm C6}) / (W_1 + W_2), \tag{1}$$

where *F*,  $\rho$  and *C* are the flow rate, the mass density and the specific heat capacity, respectively, of the coolant BarrelTherm-400 (BT400), Matsumura Oil Co. Ltd., and  $W_1$  and  $W_2$  the power of the outer sheath heater (#1) and the inner cartridge heater (#2), respectively. The parameter  $\alpha$  is determined empirically as follows. A correction factor for the flow rate fluctuation  $\Delta F$  (= *F* – *F*<sub>0</sub>) to be subtracted from *T*<sub>C2</sub> is derived from eq. (1);

$$\Delta T_{C2} = (dT_{C2}/dF) \cdot \Delta F = (-\Delta F/F) \cdot (W_1 + W_2) \cdot (dT_{C2}/dW) \cdot \alpha.$$
<sup>(2)</sup>

The correction is applied to  $T_{C2}$  for some samples or gas species to determine  $\alpha$ , so that the corrected temperature is not unreasonable, not giving negative excess temperature, or giving null excess for the Ar filling run, in the flow rate range of  $0.825 \le F/F_0 \le 1$  for  $F_0 = 20$  cc/min;

$$\alpha = 1.9 \times 10^{-2} \cdot \exp[4.0 \cdot (F/F_0)]. \tag{3}$$

The calibration run also serves as a control run giving reference temperatures, the flow rate of BT400 and the heater power for foreground runs using the silica-supported samples. Comparing the temperatures in the foreground and background runs, the excess power will be calculated using the heat conversion coefficient mentioned above.

#### III. RESULTS AND DISCUSSION

#### (1) PSf1 SAMPLE

Deuterium (D) absorption runs, D-PSf1#1, #2, #3 and #4, were performed after vacuum baking (#0) for more than 30 hours at RTD and TC2 temperatures of 200 - 300 °C with the heater power of  $(W_1+W_2) = (69+20) \sim (124+30)$  W and with the BT400 flow rate of 20 cc/min. The temperature history in the D-PSf#0 through #4 is shown in Fig. 2(a). Each time the heater power was varied, the phase number is advanced; #1-1 for D<sub>2</sub> introduction with the heater power of (0+0) W, #1-2 for (20+10) W, #1-3 for (30+20) W, and so on. At the end of each run, the heated sample was outgassed ("OG" phase) by evacuating the RC, and the run number is advanced for the succeeding run started with filling of the fresh D<sub>2</sub> gas.

In fig. 2(b) are shown the pressures at the RC and at the storage tank (ST),  $P_r$  and  $P_s$ , respectively, and the apparent loading ratio  $L_M = (D/M)$  or (H/M), *i.e.*, the number of hydrogen isotope atoms lost from the gas phase relative to the number of metal atoms (Pd in PSf1 runs or Ni in CNS3 runs). The loading ratio is calculated from the values of  $P_r$  and  $P_s$ , and volumes of the RC and the ST with a correction for the temperature based on the Boyle-Charles' law using the averaged temperature of four RTD's.



Fig. 2. Temperature (a) and D loading (b) history in D-PSf1#0 through #4 runs.

It should be noted that the apparent loading ratio  $L_M$  is very high in the low temperature phases of the #1 run (#1-1 through #1-4, where RTD and TC2 temperatures are below 200 °C), while it is modest in the #2 run and later. It is inferred that the PdO molecules in the virgin sample are reduced by D<sub>2</sub> introduction in the #1-1 phase, and the D<sub>2</sub>O molecules are exhausted in the evacuation process in the #1-10 OG phase. The  $L_M$  in the #2 - #4 runs is the proper loading ratio reflecting the proper temperature dependence, and is very reproducible.

(1-1) Absorption and heat evolution at R.T.

The initial bursts of heat are observed on the RTD and TC traces at the beginning of the #1-1 phase at R.T. Figure 3 shows the thermal power calculated from the temperature evolution in the #1-1 phase with the conversion factor mentioned above.



Fig. 3. Thermal power and deuterium loading ratio in the room temperature phase of the virgin sample run, D-PSf1#1-1.

The hump at TC2 is time-integrated to calculate an emerging energy per an absorbent atom,

$$E_{\rm a} = \int_0^t W_{\rm a} \,\mathrm{d}t\,,\tag{2}$$

where  $W_a$  is the power per an adsorbent atom, Pd in the present case. The energy  $E_a$  is calculated to be  $1.3 \pm 0.3$  eV/atom-Pd. This is rather large in view of the hydrogen absorption energy of about 0.2 eV/atom-Pd for bulk crystalline Pd. The energy  $E_a$  is divided by  $L_M$  to obtain the specific energy per D atom adsorbed/absorbed or lost from the gas phase,  $\eta_{av} \equiv E_a/L_M$ , averaged over the #1-1 phase. Similarly,  $E_a$  and  $\eta_{av}$  are calculated for #*n*-1, where *n* is the integer indicating the run number. Those are summarized in Fig. 4 together with  $L_M$ .



Fig. 4. Comparison of loading ratio and specific output energies in room temperature phases of D-PSf1#*n*-1 runs.

The reproducibilities of not only  $L_M$  but also  $E_a$  and  $\eta_{av}$  are rather good for the runs #n-1 (n > 1). This means that the reduction of PdO was completed in #1-1, and that the values of  $L_M$ ,  $E_a$  and  $\eta_{av}$  in #n-1 (n > 1) are the intrinsic ones for the sample. As one of the nanoparticle characteristics, the intrinsic  $L_M \sim 1.6$  is larger than that for the bulk Pd  $\sim 0.8$ , while  $\eta_{av}$  is nearly equal to that ( $\sim 0.2 \text{ eV/a-D}$ ) for the bulk Pd.

Now the large values in #1-1 are examined to check whether any reaction is involved other than reduction of PdO and adsorption/absorption. If we assume that  $(100 \cdot y)\%$  of Pd atoms were oxidized in the virgin sample, we have the following set of energy equations;

$$(PdO + D_2 \rightarrow Pd + D_2O + 1.77 \text{ eV/atom-Pd})\cdot y, \qquad (3)$$

$$Pd + (\mathbf{x}/2)D_2 \rightarrow PdD_{\mathbf{x}} + 0.19 \cdot \mathbf{x} \text{ eV/atom-Pd.}$$
(4)

Doing addition of these two, we obtain

$$\mathbf{y} \cdot \mathrm{PdO} + \{(2 \cdot \mathbf{y} + \mathbf{x})/2\} \mathrm{D}_2 \rightarrow \mathrm{PdD}_{\mathbf{x}} + \mathbf{y} \cdot \mathrm{D}_2 \mathrm{O} + (1.77 \cdot \mathbf{y} + 0.19 \cdot \mathbf{x}) \text{ eV/atom-Pd.}$$
(5)

The amount of the product D<sub>2</sub>O is y.79 mmol, since we have 79 mmol of Pd. Almost all of them turn into the liquid phase, since only 0.8 mmol is necessary to fill the free space of 450 cc of the RC at the saturated H<sub>2</sub>O vapor pressure of 4 kPa. Then the apparent D/Pd =  $(2 \cdot y + x) = 2.6$ . And if x = 1.6 observed in the #n-1 ( $n \ge 1$ ) phases apply also to the #1-1 phase, then y = 0.5. The reaction energy reduces to  $E_a = (1.77 \cdot y + 0.19 \cdot x)$ = 1.2 eV/a-Pd, and therefore  $\eta_{av} = 0.46$  eV/a-D. These are in fairly good agreement with the observed values of  $E_a$  and  $\eta_{av}$ . Thus, by assuming that the Pd was partially oxidized with a fraction of 0.5 in the virgin sample, the observed energies could be reasonably accounted for without assuming unconventional reactions including nuclear one.

#### (1-2) Heat evolution at E.T.

Next, we discuss the oil-outlet temperature  $T_{C2}$  in the elevated temperature phases in the D-PSf1 runs,  $T_{C2}(PSf1)$  (the red line in Fig. 5), in comparison with  $T_{C2}(SiO_2)$  in the calibration / control run using the mp-silica (the black line in Fig. 5). As is seen,  $T_{C2}(PSf1)$  is higher or lower than  $T_{C2}(SiO_2)$  even by more than 10 °C in some elevated temperature phases. This is largely due to fluctuation of the flow rate. When we apply the correction for the flow rate fluctuation according to eq. (2), the difference between them is reduced to below 2 ~ 3 °C.

The difference is converted to power by dividing it by  $dT_{C2}/dW$ , and shown in Fig. 6. We conclude that the PSf1 sample containing single-element nanoparticles shows no anomaly in the elevated temperature range up to 300 °C at TC2, if we admit a systematic error of  $\pm 2.3$  W, which is  $\pm 1.2\%$  of the input power of 204 W.



Fig. 5. Excess temperature in elevated temperature phases of D-PSf1#1 through #4 runs.



Fig. 6. Excess power in elevated temperature phases of D-PSf1#1 through #4 runs.

#### (2) CNS3 SAMPLE

Protium absorption runs, H-CNS3#1 and #2, were performed similarly. The temperatures at TC2 and RTD1 ~ RTD4 are shown in Fig. 7(a), and the H loading ratio  $L_{\rm M}$  together with the pressures at ST and RC are shown in Fig. 7(b). In contrast to PSf1 sample, little absorption is observed in the R.T. phases, #1-1 and #2-1, while substantial consumption of H<sub>2</sub> is observed in the elevated temperature phases.



Fig. 7. Temperature (a) and H loading (b) history in H-CNS3#1 and #2 runs.

The temperature evolution in the #1-1 phase at R.T. is shown in Fig. 8. The apparent loading ratio  $L_{\rm M}$  is small, and the initial burst of heat is not substantial, in accordance with the established fact that both Ni and Cu particles do not absorb much hydrogen isotopes at R.T. in contrast to Pd. The first-phase energy evaluated from  $T_{\rm C2}$ ,  $E_{\rm a} = (1.6\pm0.8)\times10^{-2}$  eV/atom-M, is more than one-order-of-magnitude smaller than that in the D-PSf1#1-1 phase. However, the resultant specific absorption energy is not so small;  $\eta_{\rm av} = (6.5\pm3.2)\times10^{-1}$  eV/atom-H.

The apparent loading ratio  $L_{\rm M}$  increases up to about 0.8 at elevated temperatures above ~ 200 °C in #1-2 and later phases. However, it does not have large values any more in the #2 run. Similar tendency had been observed also for the PNZ3 and CNZ5 samples <sup>[9]</sup>. These phenomena are consistent with a view that the main reaction consuming H<sub>2</sub> is deoxidation occurring at elevated temperatures;

$$\begin{split} \text{NiO} + D_2(\text{H}_2) &\to \text{Ni} + D_2\text{O}(\text{H}_2\text{O}) + 0.176 \ (0.033) \ \text{eV/atom-Ni}, \\ \text{CuO} + D_2(\text{H}_2) &\to \text{Cu} + D_2\text{O}(\text{H}_2\text{O}) + 1.00 \ (0.86) \ \text{eV/atom-Cu}. \end{split}$$

The very small value of  $L_{\rm M}$  at R.T. in the #1-1 phase is simply a result of Arrhenius law for absorption with an activation energy of around 0.04 eV as inferred by the temperature dependence of the absorption <sup>[9]</sup>.

Since heating by external power is necessary to induce the reaction (6) and (7) thoroughly, it is very difficult to distinguish the reaction energy from the heater power. Therefore it is difficult to make similar discussion to that for the PSf1 sample using eqs. (3) - (5).



Fig. 8. Thermal power and protium loading ratio in room temperature phase of H-CNS3#1-1.

Next, we discuss the excess power in the elevated temperature phases in the H-CNS3 runs. The temperature difference was converted to the excess power, after correction for the input heater power and the flow rate of BT400 as described above, and shown in Fig. 9. When we take into account the systematic error decided above, we see that in the CNS3 powder some anomalous effect is induced to generate excess power in some phases with the elevated temperature of about 200 to 300 °C at TC2.



Fig. 9. Excess power in elevated temperature phases of H-CNS3#1 and #2 runs.

The phase-averaged sorption energy  $\eta_{av,i}$  (closed circles) and the integrated excess energy  $E_a$  (closed squares) in the elevated temperature phases are also plotted in Fig. 9. It should be noted that  $\eta_{av,i}$  is defined as  $E_a$  divided by the absolute value of  $\Delta L_M$ , the increment of adsorbed/absorbed hydrogen atoms in the relevant phase;

$$\eta_{\text{av};i} \equiv \frac{\int_0^T W_{\text{a}} \, \mathrm{d}t}{\left|\Delta L_{\text{M}}(T_i)\right|},\tag{8}$$

The absolute value is taken to keep  $\eta_{av,i}$  positive under desorption. This is because we assume that the exothermic event could occur along with hydrogen isotope displacement under both absorption and desorption. The value of  $\eta_{av,i}$  approaches 5 keV/H, and the integrated output energy  $E_a$  exceeds 200 eV/Ni. However, the definition of  $\eta_{av,i}$  is rather problematic, since the real number of the hydrogen atoms getting in and out of the surfaces of the nanoparticles is not always represented by  $|\Delta L_M|$  in the denominator of eq. (8). Even if we divide  $E_a$  by the total amount of H absorbed in each run,  $L_M$ , to evaluate the integrated output energy per an H atom participating in the absorption, the energy is far beyond the value explainable by any chemical reaction;  $E_a/L_M = 0.9$  keV/H in the H-CNS3#2 run. The large values of the excess energy suggest the nuclear origin of the excess heat.

The excess power is observed, but with marginal amplitude in view of the systematic error of  $\pm 2.3$  W. We compare the present result with that for the CNS2 sample having the similar composition, CuNi<sub>7</sub>/mp-silica <sup>[11,12]</sup>. The result is reproduced in Fig. 10. The excess power exceeding 10 W for the CNS2 is much larger than for the CNS3 discussed above. The phase-averaged sorption energy  $\eta_{av,i} \sim 20$  keV/a-H and the integrated excess energy  $E_a = 700$  eV/a-Ni are accordingly much larger than for the CNS3. Further examination is necessary to conclude that this difference is caused by the difference in the composition.



Fig. 10. Excess power in elevated temperature phases of H-CNS2#1 through #4 runs <sup>[11,12]</sup> reprinted for reference.

Finally, the result of measurements of  $\gamma$ -ray counting rate and neutron dose rate is shown in Fig. 11. There are some periods with high neutron dose rate. However, they agree with the periods when the accelerator in the next room was operated in the neutron emitting mode. We conclude that any hard radiation does not accompany the excess heat at least of the order of several W.



Fig. 11. Radiations and temperatures at TC2 and RTD4 in H-CNS3 runs.

# IV. SUMMARY AND CONCLUDING REMARKS

The results of the absorption/heat measurements are summarized as follows in comparison with those obtained previously for other samples:

- (1) It is not impossible to ascribe hydrogen absorption and heat evolution in the initial phase at R.T. to reduction of PdO and hydrogen absorption by Pd nanoparticles in the PSf1 sample.
- (2) Heating up to around 200 °C is necessary to reduce the NiO in the CNS3 sample.
- (3) In the E.T. phases, the excess heat is observed in the runs with binary nanocomposite samples (CNS3 in the present work), while no excess heat is observed with single-element nanoparticles (PSf1 in the present work).
- (4) In the CNS3#2 run, the excess heat amounts to 200 eV/Ni (20 MJ/mol) or more than 0.9 keV/H (90 MJ/mol) without detectable dose rate of hard radiations, which cannot be explained by any chemical process.

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# Anomalous Heat Generation Experiments Using Metal Nanocomposites and Hydrogen Isotope Gas

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# Abstract

We built a new experimental system to replicate and measure precisely the anomalous heat generation experiments reported by A. Kitamura and A. Takahashi et al [1]-[3]. A liquid hydrocarbon coolant enables us to use the flow-calorimetry method at temperatures higher than 373K. This system is based on the paper [1], but improved in regard to the number of temperature measurement points and some functions. Four experiments were performed up to now. Experiments using PNZ4s ( $Pd_{0.044}Ni_{0.31}Zr_{0.65}$ ) sample with D<sub>2</sub>, CNZ5s ( $Cu_{0.044}Ni_{0.31}Zr_{0.65}$ ) with H<sub>2</sub>, PSn1 (Pd in SiO<sub>2</sub>) with D<sub>2</sub> and CNS3s ( $Cu_{0.09}Ni_{0.91}$  in SiO<sub>2</sub>) with H<sub>2</sub>/D<sub>2</sub> were performed. Anomalous excess heat generations were observed for all the samples at elevated temperature (150°C-350°C), except for the Pd nanoparticles embedded in mesoporous SiO<sub>2</sub> (PSn1). Integrated excess heat reached more than several MJ/mol-H(D) which could not be explained by any known chemical process. Coincident burst-like increase events of the pressure of reaction chamber and gas temperature, which suggested sudden energy releases in the reaction chamber, were observed many times for an experiment using the CNZ5s sample. Qualitative reproducibility between Kobe and Tohoku experiments was good. It is possible to note that the work described in [1] was successfully replicated.

## **1. Introduction**

Recently, anomalous excess heat generation observed in nickel-based nano-composite metal with H<sub>2</sub> gas at elevated temperature has been paid much attention. The following two facts are main reasons; 1) Ni and hydrogen are abundant in the world, 2) Elevated temperature is convenient to get electricity from the anomalous excess heat in the view of energy conversion efficiency. For example, F. Celani has been extensively investigated anomalous heat from Ni·Cu·Mn alloy thin wire [4]-[5]. Also some experiments similar to Rossi was performed by A.G. Parkhomov [6].

Akira Kitamura and Akito Takahashi team of Technova Inc. and Kobe University have been studying on anomalous heat effects by interaction of metal nanoparticles and hydrogen isotope gas for long time [1]-[3]. Based on their results, a new research project started on October 2015 under the collaboration of six Japanese organizations; Technova Inc., Nissan Motor Co., Ltd., Kyushu University, Kobe University, Nagoya University and Tohoku University.

Objective of the collaborative research is to clarify the existence of the anomalous heat generation phenomena and contribute to the setup of a new national project by obtaining guiding principles on how to control it. For the purpose, anomalous heat experiments at Kobe and Tohoku

Universities and sample preparations and analyses at Nissan, Kyushu and Kobe Universities have been performed. Replication experiments were performed at Tohoku University, at first, using high quality heat measurement system similar to the apparatus at Kobe University.

The Research Center for Electron Photon Science of Tohoku University and CLEAN PLANET Inc. established collaborative research division - Condensed Matter Nuclear Reaction Division in 2015 [7]. Research on anomalous heat generation started at the CMNRD (Condensed Matter Nuclear Reaction Division) although Iwamura and Itoh have been engaged in the study on transmutation reactions observed in nano-structured Pd/CaO multilayer thin film induced by D<sub>2</sub> gas permeation [8]-[13] for years at Mitsubishi Heavy Industry. Replication efforts have been made on two types of experiments as a first step at CMNRD in Tohoku University; one is the present collaborative work[1]-[3], and the other is the experiment using nano Pd/Ni fabricated by glow discharge with D<sub>2</sub> gas by T. Mizuno [14]-[15].

In this paper, the collaborative research results on the experiments performed at Tohoku University are described.

# 2. Experimental

## 2.1 Experimental Apparatus

Schematic of our experimental apparatus is shown in Figure 1. As described, it is basically based on the paper [1] and improved in some points. Main features of this system is as follows.

- 1) Oil mass flow-calorimetry at high temperature
- 2) Sufficient number of measurement points
- 3) Resistant to outer-temperature fluctuation

The reaction chamber (RC) that contains Nickel-based binary nanocomposites and hydrogen isotope gas is located in the center of Fig.1. Heat generation from the RC is estimated by the Mass-Flow-Calorimetry, which is widely used in this field as an accurate measurement method. The coolant flows into the RC from the bottom and heated by the RC. Heat generation from the RC is calculated by the temperature deference between the inlet and the outlet of coolant. The inlet oil temperature (Tin) is measured by three independent thermocouples and the outlet oil temperature (Tout) is also measured by three thermocouples. The system is intended to measure accurately heat generation at elevated temperature above 373K. A liquid hydrocarbon coolant enables us to use the flow-calorimetry method at temperatures higher than 373K. The coolant oil is an aromatic hydrocarbon (Barreltherm-400 (BT400); Matsumura Oil Co. Ltd), which can work up to 600K. The coolant is driven by a digital liquid tubing pump with flow rate from 14.0-14.8 cm<sup>3</sup>/min in this work.

 $H_2$  or  $D_2$  gas is fed from a reservoir through a super needle flow regulator to the Nickelbased binary nanocomposite sample in the RC. Pressures in the RC and the reservoir are continuously monitored. Temperature distribution in the RC is measured by 4 RTDs (Resistant Thermal Detectors) and temperatures along the oil coolant pipe and the stainless-steel pipe for gas introduction are monitored by thermocouples. A lot of temperature measurement points would enable us to judge whether an observed excess anomalous heat is real or not, although heat recovery of this system rate becomes low. Outer chamber is kept at constant temperature by cooling water from the thermobath (23°C±0.1°C). All the apparatus is in a thermostatic chamber controlled at 23°C±0.1°C to avoid the influence by the outside temperature fluctuation.

A 1kW sheath heater (W1) are spirally wound on the outer surface of the RC and a 200

W cartridge heater (W2) is located at the central axis of the RC to heat up the sample in the RC. The power to the heater is fed from a finely regulated DC power supply. The input electrical power for every heater is continuously monitored by two independent voltage and ampere meters to avoid mistakes on the input power estimation.

The coolant oil goes to the oil reservoir located outside of the thermostatic chamber and is cooled down to the ambient temperature by a cooling bath. And it goes back to the tubing pump through a water bath kept at a temperature of  $23^{\circ}C\pm0.1^{\circ}C$ .



Figure 1. Experimental Set-up

# 2.2 Sample Preparation

Procedures for sample preparation for PNZ4s and CNZ5s are the same as in [1]. At first, amorphous mixture of metal elements (Pd, Ni, Zr and Cu) were prepared by the melt spinning method at a Sendai company. By annealing the mixture in air at a temperature of 723K for 60 hr at Kobe University, preferential oxidation of Zr to ZrO<sub>2</sub> was expected with a consequent formation of binary-nanoparticles of Pd/Ni or Cu/Ni embedded in it. The sample compositions for the present paper is shown in Table 1. Atomic ratios of Pd:Ni or Cu:Ni were chosen to be 1:7 based on the Technova-Kobe works so far [1]-[3]. ZrO<sub>2</sub> beads of 1,300g weight were inserted into the RC as fillers in the case of PNZ4s and CNZ5s.

The fabricated materials at Kobe University were separated into two samples. One sample was loaded and tested at Tohoku University in Sendai City and the other at Kobe University in Kobe City, in order to compare the observed experimental results at the two places. The samples CNZ5s and PNZ4s have the same physical or chemical property as CNZ5 and PNZ4, respectively.

"Mesoporous silica" is a mesoporous form of silica and is commercially available. One of the authors fabricated the PSn1 sample using TMPS-4R (Taiyokagaku MesoPorous Silica- 4nm pore size). Pd nanoparticles under 4nm diameter were put into the mesoporous silica and fabricated at Nagoya University. PSn1 is composed of Pd, PdO and SiO<sub>2</sub>. Total weight of PSn1 is 114.6g as

shown in the Table 1. In this case, no filler was used because density of mesoporous silica is low compared to that of  $ZrO_2$ .

The CNS3s sample consists of  $CuNi_{10}$  nanoparticles supported by the mesoporous silica made by Admatechs Co. Ltd. The sample was synthesized from a solution of nickel chloride and copper chloride containing the mesoporous silica powder as a suspended material to adsorb Ni and Cu in mesoscopic pores at Kobe University. After filtration, the mesoporous silica was annealed at 800 °C for 3 hrs.

Sample	Fabricated at	Sample Weight			Mass(g)			
name		(g)	Cu	Pd	Ni	Ο	Zr	SiO2
PNZ4s*	Kobe U.	109.4		6.0	23.0	4.5	75.9	
CNZ5s*	Kobe U.	130.0	3.4		22.0	32.2	72.4	
PSn1	Nagoya U.	112.4		7.5		1.1		103.8
CNS3s	Kobe U.	150.5	1.2		11.4	3.4		134.5

Table 1. Sample composition.

\* ZrO2 beads of 1,300g weight were inserted into the Reaction Chamber as fillers in the case of PNZ4s&CNZ5s.

# 2.3 Experimental Procedure

Calibration or blank runs to know the heat recovery rate of the experimental apparatus were performed using zirconia beads in the RC before or/and after foreground runs. Diameter of the beads was 1mm and weight was 1300g for PNZ4s and CNZ5s experiments. In the case of PSn1 and CNS3s experiments, blank experiments were performed using mesoporous silica without Pd or CuNi.

At the beginning of a PNZ4s or CNZ5s run, a sample with the zirconia beads was put into the RC. As for PSn1 or CNS3s run, only a sample was inserted in the RC. The RC was evacuated by a turbo molecular pump and then heated up to 200-300°C to remove H<sub>2</sub>O or other impurity gas such as hydrocarbon gas. After the baking, the RC is cooled down to room temperature. About 1.0MPa H<sub>2</sub> or D<sub>2</sub> gas were stored in the reservoir chamber in advance, then H<sub>2</sub> or D<sub>22</sub> gas was introduced into the RC by opening the super needle valve as shown in Fig.1. A 1L reservoir chamber for H<sub>2</sub> gas storage and a 2L chamber for D<sub>2</sub> gas were used.

In the case of PNZ4s and PSn1 samples,  $D_2$  gas absorption and heat generation was observed at room temperature due to the presence of Pd. After that, we applied electric power to the heaters located at inside and outer-surface of the RC to increase sample temperature. Data of temperatures, pressures, voltages, currents and a flow rates was logged during experiments. Based on the data, we estimated H or D absorption rate and excess heat generation from the samples.

# 3. Results and Discussion

3.1.Heat Analysis and its Error Estimation

Heat analysis of this system is based on the equation:

$$\eta Q = F_R \cdot \rho(T_{ave}) \cdot C(T_{ave}) \cdot (T_{out} - T_{in}) \tag{1}$$

where  $\eta$  is the heat recover rate, Q is the heat release rate,  $F_R$  is oil flow rate,  $\rho(T_{ave})$  is the oil density as a function of temperature,  $C(T_{ave})$  is heat capacity,  $T_{out}$  and  $T_{in}$  is the outlet and inlet temperatures of the coolant oil, respectively. Physical data of  $\rho(T)$  and C(T) of the coolant oil are already known. As temperature dependence of  $\rho(T)$  and C(T) is linear, we can postulate that  $T_{ave}$  is equal to  $(T_{out}+T_{in})/2$ .

Q is expressed as

$$Q = W_1 + W_2 + H_{EX} \tag{2}$$

where  $W_1$ ,  $W_2$  and  $H_{EX}$  are the input power of heater 1, the input power of heater 2 and the excess heat power from the RC.

Based on these equations,  $\eta$  is determined as a function of  $(W_1+W_2)$  by a blank run because Q, F<sub>R</sub>,  $\rho(T_{ave})$ ,  $C(T_{ave})$  and  $(T_{out}-T_{in})$  are obtained by experimental data. H<sub>EX</sub> is calculated by a foreground run using the determined  $\eta$ .

We simplify equations (1) and (2) for experimental error estimation.

$$H_{EX} = \frac{F_R \cdot \rho \cdot C}{\eta} \Delta T - W;$$
  
$$\Delta T = T_{out} - T_{in}, \quad W = W_1 + W_2.$$

Considering that experimental variables are  $F_R$ ,  $\Delta T$  and W, we can assume that error range of the calculated excess heat is the sum of fluctuations of oil flow rate, temperature difference and input electrical power.

$$\delta(H_{EX}) \approx \left|\delta(F_R)\right| \frac{\rho \cdot C \cdot \Delta T}{\eta} + \left|\delta(\Delta T)\right| \frac{F_R \cdot \rho \cdot C}{\eta} + \left|\delta(W)\right| \tag{3}$$

Actual experimental data shows that largest contribution to the error of  $H_{EX}$  is of the  $F_R$  term and W is most stable.

#### 3.2.PNZ4s

The first experiment was done using the PNZ4s sample at Tohoku University after a blank run. Figure 2 shows the temperature change at 4 points in the RC during absorption of D into PNZ4s at room temperature. Rapid temperature rises from room temperature could be seen depending on the positions of RTDs when  $D_2$  gas was introduced into the RC. RTD1, RTD2, RTD3 and RTD4 are located successively from the bottom as shown in Fig.2. It could be supposed that PNZ4s sample local density was highest around the RTD2 and RTD3, considering that the weight ratio of PNZ4s to zirconia beads is less than 0.1 and sample powder would be localized. This might be the reason why peak temperatures are larger at RTD2 and RTD3.



Figure 2. Temperature Change at 4 Points of the RC during absorption of D into PNZ4s at Room Temperature

Figure 3 shows excess heat generation from PNZ4s with  $D_2$  gas. The peak power induced by  $D_2$  gas absorption at room temperature amounted to 10W but it did not last for long time. On the contrary, excess heat power was not so high at elevated temperatures but it continued more than 100h. The peak ratios of excess heat to input power were about 4% and 5% for 80W and 134W input, respectively. Integrated excess energies were 0.65MJ for 80W input and 1.73MJ for 134W input. Therefore the amount of the released excess energy from the RC was about 2.4MJ.



Figure 3. Excess Heat Generation for PNZ4s

According to the pressure measurement at the RC and the reservoir for  $D_2$  gas, total D atom absorbed into PNZ4s was 1.73mol. Although it is very difficult to postulate that every absorbed D atoms produced any additional energy after the saturation of D-loading, average released energy at elevated temperatures per initial total D-loading is calculated as 14.9eV/D, which is the least estimation of specific energy by one deuterium reaction that might happen. As the long lasting excess heat was observed after the D-loading was saturated, real portion of deuterons contributed to the heat generation is regarded to be very small (several order of magnitude [1]-[3] and consequently estimated specific energy might be very large). It is therefore very hard to explain that the generated excess heat was caused by some chemical reactions, as energy generation from known chemical reactions cannot exceed 10eV/D

#### 3.3.CNZ5s

Comparison of temperatures detected by RTDs and E1 between a blank run and an experiment with CNZ5s is shown in Fig.4 when 134W heater power was provide with the RC. In this experiment, no input was applied by W2 for all the time as the W2 heater did not work due to disconnection-troubles. The thermocouple E1 is located on the top of the RC. Temperature increases of RTD3, RTD4 and E1 from the blank experiment data were observed for the CNZ5s experiment, although there was no difference of RTD1 and RTD2 temperatures compared to those of blank runs. It seems that excess energy generated at higher temperature than 220°C based on these observed experimental results.



**Figure 4. Comparison of Temperatures Detected by RTDs and E1** 

Pressure of the RC (Pr) and a gas temperature (E2) at the near point of the top of RC are plotted in Fig.5 Sudden fluctuations of Pr and E2 could be seen during the CNS5s experiment, though no fluctuation could be seen during the blank run and the baking (time-zone: 0-150 hr) as shown in Fig.5.



## Figure 5 Fluctuations of Pressure of the RC (Pr) and a E2 Gas Temp. (E2) during a CNZ5s Experiment

Coincident increase events of Pr and E2 at random timing were observed as shown in Fig.6. The pressure increase was about 0.02 or 0.03 MPa and the temperature increase ranged from 1°C to 5°C. These were significant values and not explained by an outer noise as the other measurement data were stable and only Pr and E2 changed coincidently.



Figure 6. Coincident increase of Pressure of the RC (Pr) and a E2 Gas Temp. (E2)

If we closed up Pr and E2 further, we can make sure that Pr and E2 increased simultaneously as shown in Fig.7. Time interval for the plotted points for Pr and E2 was 10sec. Excess energy seemed to be generated in the upper region of the RC as demonstrated in Fig.4. And these simultaneous increases of Pr and E2 might be due to the generation of high temperature hydrogen gas. Although it is very difficult to describe what is happening at upper side of the RC, coincident temperature and pressure rises suggest burst-like energy releases from the RC.



Figure 7. Simultaneous increase of Pr and E2 and the hypothesis for it

Excess heat for CNZ5s based on the equations (1) and (2) is illustrated in Fig.8. About 4-5W and 2-4W were released during 80W and 134W heater power inputs, respectively. These excess powers are significantly larger than the estimated errors based on the equation (3). Sudden increase of excess power around 240h seemed to be induced by the increase of oil flow rate, but we have no accurate explanation for it.





All the integrated excess power is 1.86 MJ according to the Fig.8. And estimated H atoms absorbed into CNZ5s during the experiment is 0.29mol based on the pressure measurement at the RC and the reservoir for  $H_2$  gas. Excess energy production is calculated as 67.8eV/H, although it is very difficult to assume that every absorbed H atoms produced any energy same as D atoms.

Also it is very difficult to explain that the generated excess heat was caused by some chemical reactions, as 67.8eV/H energy generation is larger than the usual chemically released energy. This anomalous excess energy generation, in addition to the observed coincident pressure and temperature rises of the RC, might suggest that some condensed matter nuclear reactions occurred in the RC.

3.4. Comparison between Experimental Results at Tohoku University and those at Kobe University

Table 2 summarized experimental results obtained at Tohoku University and those at Kobe University. As previously described, the samples CNZ5s and PNZ4s have the same physical or chemical property as CNZ5 and PNZ4, respectively. The released energies at room temperature from PNZ4s and PNZ4 were 0.57eV/D and 0.56eV/D, respectively. These values are very similar. If we look at the released excess power from CNZ5s and CNZ5, they are close to each other. These results shows that we successfully replicated experimental results on excess heat at least qualitatively, although quantitative reproducibility is not enough.

# Table 2. Comparison between Experimental Results at Tohoku University and those atKobe University

Sample	Composition	Tested at	Released Energy at room temp.	Released Excess Power at elevated temp.	RC temp.
PNZ4s	Pd <sub>0.044</sub> Ni <sub>0.31</sub> Zr <sub>0.65</sub>	Tohoku University	0.57eV/D	2W to 6W	160-300
PNZ4		Kobe University	0.56eV/D	Not estimated malfunction	l due to
CNZ5s		Tohoku University		2W to 5W	150-250
CNZ5	Cu0.0441N10.31ZI0.65	Kobe University		2W to 8W	200-350

# 3.5. PSn1

PSn1 is composed of nano-sized Pd under 4nm and mesoporous silica as described before. Figure 9 shows comparison of the delta T (oil temperature difference between inlet and outlet) and input power (W) between PSn1 and blank run. The blank run is performed with mesoporous silica without Pd nanoparticles. No significant deference of delta T between PSn1 and blank runs were observed if the input power was the same. It means that no anomalous excess heat was confirmed in the case of PSn1.

According to the experimental results at Kobe University, PSf1 sample, which is composed of nano-Pd only, did not give any excess heat [16]. It seems that we cannot observe any anomalous excess heat from nano-sized Pd at elevated temperature.



Figure 9. Excess Heat Evaluation; No Excess Heat

## 3.6. CNS3s

Figure 11 shows excess heat generation from CNS3s sample. Excess heat generation was observed when  $H_2$  gas was fed, although no excess heat was detected in the case of  $D_2$  gas experiment. It seems that higher temperature enhanced excess heat power, but it is difficult to state a solid conclusion. All the integrated excess power is 4.3 MJ and estimated H atoms absorbed into CNS3s during the experiment is 0.40mol. Average excess energy production per H is calculated as 10.7MJ/H-mol (111eV/H).

Summary of experimental results are shown in Table 3.



Figure 11. Excess Heat from CNS3s

Samples	Gas	Temp.	Results	
PNZ4s (PdNi <sub>7</sub> /ZrO <sub>2</sub> )	D	160-300°C	1) Excess Heat 4-5W, Integrated Heat > 15eV/D(1.4MJ/mol-D)	
CNZ5s (CuNi <sub>7</sub> /ZrO <sub>2</sub> )	Н	160-250°C	<ol> <li>Excess Heat 2-5W, Integrated Heat &gt;68eV/H (6.5MJ/mol-H)</li> <li>Coincident increase events of the pressure of the reaction chamber and gat temperature</li> </ol>	
PSn1 (Pd/meso-Si) D 200-300°C		200-300°C	No Excess Heat at elevated temp.	
CNS3s (CuNi <sub>10</sub> /SiO <sub>2</sub> )	H(D)	150-300°C	<ol> <li>In the case of H, Excess heat 2-4W, Integrated Heat&gt;110eV/H (10.7MJ/mol- H)</li> <li>No excess Heat in the case of D</li> </ol>	

 Table 3.
 Summary of Experimental Results

# 4. Concluding Remarks

A new experimental system to replicate the anomalous heat generation using Nickelbased binary nanocomposites and  $D_2/H_2$  gas was introduced at Tohoku University. Four experiments were performed up to now. Anomalous excess heat generations were observed for all the samples at elevated temperature except for the Pd nanoparticles embedded in mesoporous SiO2 (PSn1). Integrated excess heat reached more than several MJ/mol-H(D) which could not be explained by any known chemical process. Experimental results of the present work qualitatively agreed with those of the similar experiment performed at Kobe University. Coincident burst-like increase events of the pressure of reaction chamber and gas temperature, which suggested sudden energy releases in the reaction chamber, were observed many times for an experiment using the Cu-Ni-Zr (CNZ5s) sample.

## Acknowledgements

The authors would like to thank Mr. Y. Shibasaki (Tohoku U.) for his support for establishing experimental system. The authors would like to appreciate Technova Inc. for the corporation on building our experimental apparatus. We also acknowledge CLEAN PLANET Inc. for its significant contributions.

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# Characterization of deuterium diffusion in multilayered metal samples

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#### ABSTRACT

We performed deuterium desorption experiments using multilayered Pd–Ni, Pd–Ag, Pd–Ti, and Pd–Zr complex samples with fine-structured surfaces and simultaneously investigated the deuterium diffusion and thermal behavior of the sample. We observed a short-period temperature fluctuation that lasted 2–4 h. In addition, for the Pd–Ti and Pd–Zr samples, a sudden temperature increase occurred during the desorption experiment. These phenomena are thought to result from a property of the complex sample structure, that is, Pd coated with a thin metal membrane, which causes a unique type of deuterium diffusion.

#### 1. Introduction

In a previous study, we observed anomalous heat evolution as well as radiative tracks during deuterium diffusion in multilayered complex samples such as Pd-Au, Pd-Pd, Pd–CaO, and Pd–Ni [1–3]. In addition, anomalous heat evolution has been observed in a deuterium desorption experiment with binary (ternary) nanoparticles such as Pd-Ni-ZrO [4]. The experimentally observed phenomena could be attributed to a specific characteristic of deuterium diffusion in metal complexes as well as the fine structure of the sample. Hence, it is significant to investigate deuterium diffusion behavior systematically in various types of metal complexes to clarify the anomalous phenomena. We have been performing deuterium desorption experiments using multilayered samples prepared by depositing thin metal films on a Pd foil, such as Pd-Ni, Pd-Ag, Pd-Ti, Pd-Zr, and Pd-Ni-Zr, and have investigated the deuterium diffusion associated with the thermal behavior and the correlation between the two. In the experiment, samples with a fine-structured interface between two metals were also tested to investigate the dependence of the deuterium desorption behavior on the surface condition. We have observed unexpected temperature behavior. For the samples with a fine-structured interface, we observed a short-period temperature fluctuation that lasted 2–4 h at the beginning of the desorption experiment. In addition, in some experiments with Pd-Ti samples, a sudden temperature increase was observed 1-2 h after a DC

current was applied. Some of the results have been presented at JCF15 and JCF16 [5,6]. In this paper, we present the updated results of our ongoing study.

# 2. Heat of Solution of Deuterium

The thermal diffusion behavior of deuterium should depend on the heat of deuterium dissolution. Metals are generally classified as exothermic or endothermic absorbers of hydrogen. Ni, Ag, and Au are classified as endothermic absorbers. If deuterium diffusion in the interface region of binary metals significantly determines their thermal dynamics, we may expect similar temperature behavior for Pd–Ag and Pd–Ni binary metals. On the other hand, Pd, Ti, and Zr are classified as exothermic absorbers. Therefore, a Pd–Ti sample may show different temperature behavior during deuterium desorption. The heats of dissolution of Pd, Zr, Ti, Ni, and Ag for deuterium are shown in Table 1 [7].

Metal	Heat of solution		
	(KJ/IIIOI)		
Pd	-10		
Zr	-63		
Ti	-53		
Ni	16		
Ag	68		

Table 1 Heat of solution for deuterium.

#### 3. Experiment

Multilayered samples were fabricated by depositing a thin metal membrane by  $Ar^+$ ion beam sputtering onto the surface of the Pd foil substrate. The size of the Pd foil used in this study was 10 mm × 10 mm × 0.1 mm. The thickness of the membrane was ~100 nm. We also fabricated samples with fine structure at the interface of the binary metal. These fine structures were obtained by etching the Pd membrane using an Ar ion beam. Figure 1 shows the surface morphology before and after etching as analyzed by atomic force microscopy (AFM). The number of trials for each condition is shown in Table 2.



Fig. 1 Surface morphology before (left) and after (right) etching as observed by AFM.

Sample type	Membrane material	Fine-structured interface	Number of runs
i	NI:	No	20
ii	INI	Yes	17
iii	<b>A</b> -	No	11
iv	Ag	Yes	6
v	T:	No	17
vi	11	Yes	9
vii	7.	No	13
viii	Σľ	Yes	11

Table 2 Sample conditions and number of runs in this study.

To load deuterium to the samples, the fabricated multilayered samples were exposed to deuterium gases at 5 atm for ~24 h. The weight of the samples was measured before and after loading, and the loading ratio (D/Pd) was calculated from the weight difference. Figure 2 shows the loading ratio D/Pd and its average for each sample. For reference, we performed deuterium desorption experiments with a simple Pd foil and a Pd foil with fine structure. The average loading ratio was found to be 0.65–0.70 for most samples, and no significant difference in D/Pd values of each sample was seen. Note that the loading ratio shown in the figure is the value averaged over the entire sample. Thus, it is concluded that D/Pd is not affected by the interface structure of the binary metal or by the type of deposited metal film.



Fig. 2 Average D/Pd ratio of each sample structure.

After each sample was weighed to measure the D/Pd ratio, it was placed in a chamber that can be evacuated by a turbomolecular pump ( $\sim 10^{-4}$  Pa). In the chamber, the sample was heated by supplying electric current to stimulate deuterium out-diffusion from the sample. In the desorption test, the sample temperature and the pressure in the chamber were monitored continuously for  $\sim 24$  h. A thermocouple and ionization gauge were used for these measurements. The current and bias applied to the sample were also monitored during the experiment.

#### 4. Results and Discussion

The temperature behaviors during desorption could be classified into three patterns, which are described below and have already been reported at JCF16. We show some of the same results here again, as well as updated or new results, and then discuss an underlying process explaining the observed results.

#### Pattern I

Figure 3 shows the time dependences of the sample temperature, pressure in the chamber, and applied voltage for the Pd–Ni sample, which demonstrate the distinctive behaviors of Pattern I. After a DC current was applied to the sample, the surface temperature increased owing to Joule heating, and deuterium desorption began. Because

the sample resistance is decreased by desorption of deuterium, the applied voltage decreases. Thus, the sample temperature gradually decreased. In addition, deuterium desorption from Pd is an endothermic reaction, and this is another reason for the temperature decrease. After the desorption process terminated, the sample temperature became constant except for an accidental fluctuation. This Pattern I behavior was observed most frequently for the Pd–Ni, Pd–Ag, and Pd–Ti samples, but not at all for Pd–Zr. Although this pattern itself can be explained reasonably well by well-known processes, it is not clear why it was not observed for Pd–Zr.



Fig. 3 Temperature and chamber pressure (left) and temperature and applied voltage (right) for sample type (i) (Pattern I).



Fig. 4 Temperature and chamber pressure (left) and temperature and applied voltage (right) for sample type (ii) (Pattern II).
### Pattern II

Figure 4 shows typical Pattern II behavior. It shows the short-period fluctuation in temperature that lasted 2–4 h at the beginning of the desorption experiment. Pattern II was observed several times for the samples with a fine-structured interface.

Figure 5 shows the time differential of the temperature (dT/dt). A set of continuous increases and instantaneous decreases in temperature occurred periodically. We did not observe significant variation of the pressure inside the chamber during the continuous temperature fluctuation, so a significant quantity of deuterium atoms was not thought to be desorbed from the sample. It is possible that deuterium diffusion from Pd to the membrane and from the membrane to Pd occurred frequently in this period and that endothermic and exothermic phenomena associated with the heat of solution repeatedly occurred owing to deuterium transport between the two metals as shown in Fig.6. In addition, the deuterium diffusion velocity in Pd is faster than that in Ni or Ag [6]. Thus, deuterium diffusion from Pd to Ni or Ag might occur more rapidly than that from Ni or Ag to Pd, which might explain the rapid temperature decrease and the gentle increase shown in Fig. 5. Especially for Pd–Ni, the phenomenon was observed several times for the samples with fine structure in the interface region. Therefore, such microstructure might enhance deuterium diffusion between the two metals.

### Pattern III

Figure 7 shows typical Pattern III behavior for the Pd–Ti sample, where a sudden temperature increase was observed 1–2 h after the DC current was applied. Pattern III was observed in 3 out of 26 runs for the Pd–Ti sample. Figure 8 shows the time variation of the applied voltage and the pressure in the chamber at the beginning of the experiment.

We found that the voltage increased gradually and decreased rapidly after reaching its peak value. Because the DC current is constant for the Ti membrane side of the sample (see Fig. 9), the applied voltage depends on the resistance of the Ti membrane, that is, the deuterium content of the Ti membrane. Therefore, an increase in the voltage suggests that the density of the deuterium in the Ti membrane becomes higher. Then, explosive gas release from the sample causes a steep temperature drop concurrently with the pressure rise because Ti is an exothermic absorber of deuterium. The voltage and pressure behavior shown in Fig. 9 might be explained by this model.



Fig. 5 Temperature variation for dT/dt at each second in Pattern II.



Fig. 6 Schematic view of deuterium diffusion in the pattern II.



Fig. 7 Temperature and chamber pressure (left) and temperature and applied voltage (right) for Pd–Ti (sample type v).



Fig. 8 Time dependence of the voltage and pressure in the chamber.



Fig. 9 Schematic view of current supply to Pd-Ti sample.



Fig. 10 Temperature and applied voltage for Pd-Zr (sample type vii).



Fig. 11 Time dependence of the voltage and pressure in the chamber.

Moreover, this phenomenon was observed for the Pd–Ti sample without fine structure on the surface. This may suggest that the fine structure promotes deuterium diffusion, and high deuterium density is not realized in the Ti membrane.

Similar behavior was observed for Pd–Zr samples both with and without fine structure at the interface, although a sudden temperature increase was observed earlier in the desorption process, as shown in Fig. 10. Figure 11 shows the time variation of the applied voltage and the pressure in the chamber at the beginning of the experiment. This was observed in 19 out of 24 runs for the Pd–Zr sample. This phenomenon is induced much more frequently in Pd–Zr than in Pd–Ni. If the behavior originates in the process mentioned above, we could suppose that deuterium is easy to move from Pd to Zr and that the Zr membrane has the capacity to store a certain quantity of deuterium, although the diffusion coefficients of deuterium in Ti and Zr do not differ greatly [8,9].

Figures 12 and 13 show SEM images of the Ti and Zr membrane surfaces, respectively, after the desorption experiment. The surface of the sample in which we observed a sudden temperature (or voltage) increase is compared with that of the sample without a temperature (voltage) increase. We clearly observed the grain boundaries the samples in which a sudden temperature increase occurred. The surface morphology resembles that of a sample exposed to hydrogen, resulting from hydrogen embrittlement [10]. It may indicate that much deuterium was desorbed from the membrane surface. Although we have examined just a typical sample for each, this would be an evidence that deuterium diffused from Pd to Ni or Zr and was desorbed from the membrane side in conjunction with the temperature changes. Thus, we may suppose that the

combination of a bulk Pd foil and an exothermic metal membrane may bring the direction of deuterium diffusion from the Pd foil to the membrane.



Fig. 12 Scanning electron microscopy (SEM) image of Ti membrane of Pd–Ni samples for which sudden temperature increase was not observed (left) and was observed (right).



Fig. 13 SEM image of Zr membrane of Pd–Zr samples in which sudden temperature increase was not observed (left) and was observed (right).

Sample	Pd–Ni		Pd–Ag		Pd–Ti		Pd–Zr	
Pattern	i	ii	iii	iv	v	vi	vii	viii
Ι	20	11	11	5	13	9	0	0
II	0	6	0	1	1	0	2	2
III	0	0	0	0	3	0	11	9

Table 3 Number of runs classified into each pattern.

In Table 3, we summarize the number of runs for each pattern.

### 5. Summary

We performed deuterium desorption testing of multilayered metal samples of Pd–Ni, Pd–Ag, Pd–Ti, and Pd–Zr complexes and investigated the deuterium diffusion associated with the thermal behavior. We observed unusual temperature and deuterium diffusion behavior. For example, we observed a short-period temperature fluctuation that lasted for 2–4 h at the beginning of the desorption experiment (Pattern II). This was frequently observed for the Pd–Ni sample with fine structure at the interface. Another distinctive phenomenon was a sudden temperature increase occurring 1–2 h after a DC current was applied. This behavior was often observed in the Pd–Zr sample. We established a model of deuterium diffusion inside the sample that explains the phenomena observed to date reasonably well, although it is still very speculative. If we can determine the correlation between the properties of each metal with regard to deuterium diffusion and the observed thermal behavior in a further systematic study, the origin of the phenomena might be clarified.

Moreover, we should consider the poor reproducibility of such phenomena. Even for the same sample type, the temperature behavior is not always the same. This is probably the result of nonuniformity of the microstructure of the membrane or its interface with the Pd foil. Microstructural analysis of the samples may help us identify the mechanism of unexpected thermal or diffusion behavior.

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### Microscopic structural change of Pd rod during repeated cathodic and anodic electrolysis in glycerinphosphoric acid and during long-term electrolysis in 0.1 *M* Li OD

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Abstract: Long-term electrolysis for well annealed thick Pd rod (9.0 mm diameter) in 0.1 *M* LiOD was performed. Microscopic observation of the postelectrolysis Pd electrode showed the characteristic surface morphology of slip bands and doubly cross slip, which indicated that the limited areas of Pd electrode were subjected to high stress and high strain over  $\varepsilon$  0.6.

The physico-chemical properties of hydrogen dissolved in Pd have been studied by *in situ* potentiometric, resistance and dilatometric measurements during repeated hydrogen absorption/desorption electrolysis in glycerin and phosphoric acid. During the first C mode, the potential changed obeying Nernst's equation (the  $\alpha$  phase), and held a constant value (the  $\alpha + \beta$  two phase coexistence), and exhibited the transition from the  $\beta$  phase to the  $\beta$ + voids coexistence region. Concurrently with the potential change, the dilation exhibited a gradual increase, from whose slope the apparent molar volume was obtained as 0.64 (the  $\alpha$  phase), 0.40 (the  $\alpha + \beta$  phase) cm<sup>3</sup>/mol. The resistance increased in proportion to the H/Pd ratio and kept constant at 1.7 to 1.8 over  $R_{tr}$ . For over the  $\beta$  phase (> $\beta_{min}$ ) the potential shifted with an increase in dilation, which suggests nonequlibrium PdH<sub>2-x</sub> precipitation followed by conversion to the  $\beta$  phase and void formation.

During the repeated C mode, the onset of the  $\beta$  phase ( $\beta_{\min}$ ) increased as the repetition number increased and the apparent molar volume of the  $\alpha + \beta$  phase coexistence showed that absorption proceeded inhomogeneously, in contrast to the first C mode. According to the suggestions of two literatures, in the  $\alpha + \beta$ phase coexistence region, there evolved nonequilibrium  $\alpha$ ' phase and/or amorphous accompanied with high density of defects or vacancies. The properties of Pd-hydrogen system were further characterized by combining with the well-established knowledge of hydrogen embrittlement.

*Keywords*: cold fusion, deuterium, electrolytic hydrogen absorption, Pd,  $\alpha$  to  $\beta$  transformation, resistance, dilation, electrode potential, void, vacancy, trap site, hydrogen embrittlement

### **1** Introduction

During long-term electrolysis for well annealed thick Pd rod (9.0 mm diameter) in 0.1 M LiOD, vortex pattern was observed <sup>1-2)</sup>. The morphology of the postelectrolysis electrodes revealed two long faults without any cracks on the surface. N-cycle model was proposed <sup>3-4)</sup> to understand the phenomena related with Cold fusion (CF); where the vortex pattern was modeled as the vortex threads moving under beneath the surface. The trace of the vortex pattern was simulated using computer simulation. There were vortex threads realized as the continuous flow of hypothetical particles mass (HPM) from a vessel to a neighboring one in the Scavenger process <sup>5)</sup>. So far, we have succeeded in obtaining, though not precise, a vortex pattern and their cascade using numerical simulation <sup>6)</sup>. In N-cycle model it was predicted that deuterium (hydrogen) occluded in the vessel was encountered by occasional absorption and compression, however the solid-state properties of the vessel and the

mechanism of compression or tensile stress evolution were still ambiguous. Thus, we have attempted to elucidate the elemental phenomenon of the electrochemical H absorption, i.e., subsequent phase changes, lattice expansion, microstructural changes, related resistance increase, etc., under well-defined electrolysis conditions <sup>7</sup>). Since during H absorption followed by desorption mechanical stress accumulates accompanied with evolution of miscellaneous structural defects, it is demonstrated that *in situ* measurements of the electrode potential and solid-state properties are useful for evaluating the influence of absorption/desorption repetitions on solid-state properties of Pd <sup>7</sup>).

Alternatively, Pd-hydrogen (-deuterium) system has been the subject of research in the field of solid-state physics and material science. For example, research themes for engineering problems involve wide areas: hydrogen embrittlement, membrane of hydrogen purification apparatus, hydrogen storage materials, hydrogen sensor, etc. From the viewpoint of theory and experimental measurement methods, recent developments of metal-hydrogen research have been remarkable.

It is significant to reconsider our study from a different point of view for analyzing the hydrogen induced structural change. Hence, we focus recent researches: *in situ* Synchrotron and Neutron Diffraction experiments and material scientific analysis for clarifying structural changes of metals concerned with hydrogen embrittlement. We also pay enough attention to dissimilarities of material preparation (pretreatment and hydration cycling) methods, intermetallic compound and pure metal, *in situ* and *ex situ* measurements, and phenomena with nano and more large scale.

In this study, compared with the morphology of deformed sample of usual tensile test, the surface morphology of the postelectrolysis Pd electrode after long-term electrolysis has been investigated with respect to the applied mechanical condition.

*In situ* measurements of potential, dilation and resistance were made for analyzing the microstructural changes induced by H absorption in a Pd electrode. Based on the analyses obtained, microstructural changes of hydrogen absorbed Pd was thoroughly elucidated by combining with the knowledge of hydrogen embrittlement.

### 2 **Experimental**

# 2.1 Long-term evolution of deuterium on thick rod Pd electrode in 0.1 *M* LiOD

Cold fusion experiments at ambient temperatures had been conducted by electrolysis of heavy water containing 0.1 *M* LiOD on a Pd electrode. The electrolysis cell (content is about 130 ml) was made out of transparent quartz, which had a water jacket. The temperature of the cell was controlled at 40 and 50 °C by Coolnics (Komatsu-Yamato Co., Ltd.). The electrode potential was referred to dynamic type ( $\alpha + \beta$ )-PdD reference electrode. The dimension of a thick Pd rod was 0.9 cm diameter x 5.3 cm long. Using a potentiostat (or current supply) a constant cathodic current was applied to the Pd electrode on which an evolution of deuterium gas was occurred. By continuing a state where deuterium is strongly absorbed in the Pd electrode, heat generation and emissions of neutrons was observed. We successfully performed non-intermittent electrolysis for six months.

The experimental apparatus and procedures are:

(1) Cast rod Pd electrode (2) Pretreatment of Pd electrode (3) Preparatory gas phase absorption of  $D_2$ (D/Pd = 0.36) (4) Increase in electrolysis current density in a form of stepwise (5) Temperature cycling.

The other experimental details: sample pretreatment, experimental apparatus and electrolysis conditions are described elsewhere <sup>1-2)</sup>.

During long-term electrolysis the



Fig. 1 C mode pattern of current density-time.

electrode diameter (at three positions in height; top, middle and bottom) was measured after interruption of electrolysis and dismantling the electrode from the cell. Then the electrode was carefully re-installed. This procedure was repeated 4 times, where the duration periods ranged two months or less. It is noted that in this study, long-term electrolysis was conducted in a deuterium solution, and the experimental data of Pd-D system is considered to be same as Pd-H system; as long as the theme is concerned with material items.

# 2.2 *In situ* potentiometric, resistance and dilatometric measurements of Pd electrode during repeated electrochemical hydrogen absorption

The absorption and desorption of H in Pd rod electrodes (0.8- and 2-mm diameters, 50mm length, 99.95% purity) was performed by applying galvanostatic cathodic and anodic pulse currents, current densities,  $< 2 \times 10^{-3}$  A/cm<sup>2</sup>. The electrolyte was composed of glycerin and phosphoric acid (2:1 in volume ratio). The temperature was kept constant at 40  $\pm$  0.5 °C using an isothermal bath. The electrolytic cell and apparatus for *in situ* measurement of potential, dilation and resistance are described elsewhere <sup>7</sup>). As received Pd was pretreated by our conventional experimental procedures.

Electrolysis was performed using galvanostatic trained pulse currents (abbreviated as C mode), as shown in Fig. 1. After each galvanostatic discharge for a fixed number of hours, the potential, dilation and resistance of Pd were measured. Then the potential reached steady state; equilibrium between the H atmosphere and H activity inside the Pd was believed to be attained. Hydrogen concentration (the H/Pd ratio: x) is given by the summation of each increment of the concentration converted by the charges (current multiplied by time of electrolysis), in accordance with Faraday's law. As shown in Fig. 1, in the C mode, a set of trained pulse currents (absorption) and desorption was repeated until the repetition number was attained.

It is known that interaction between dissolved atoms and surrounding materials often bring about lattice strain in materials. The apparent partial molar volume  $[d(\Delta l/l_0)/dx]$ (abbreviated as apparent molar volume) was calculated from  $3V_M$  ( $V_M$  the molar volume of the metal) multiplied by the slope of the dilation as a function of H/Pd ratio.

### **3** Results and Discussion

## **3.1 Microstructure of a thick Pd electrode during deuterium evolution in 0.1** *M* LiOD



Fig. 2 Schematic illustration of Cup and corn type fracture and tensile test apparatus. (load M, cross sectional area A, initial length  $l_0$  and incremental dilation  $\Delta I$ )

Figure 2 shows usual tensile test apparatus to evaluate mechanical properties of metal, e.g., ultimate tensile strength, yield stress, etc. When a rod is set to the apparatus and tensile load *M* is increased, the initial length  $l_0$  starts to elongate. In actual stress strain curve every point is drawn on the graph with stress  $\sigma$  on the longitudinal axis and strain  $\varepsilon$  on the lateral one. From obtained data stress is calculated as  $\sigma = M/A$  and strain as  $\varepsilon = \Delta l/l_0$  where M, A,  $\Delta l$  and  $l_0$  denote load, cross sectional area, incremental dilation and initial length, respectively. Figure 3 shows the schematic stress strain curve of single crystal f.c.c metal which exhibits 3 categorized stages; especially one stage appears characteristic appearance shown in the right insets <sup>8</sup>.

In the case of polycrystalline the transition of elastic deformation to plastic one is gradual behavior while for f.c.c single crystal (stress strain curve in Fig. 3) the curve exhibits distinct 3 stages: 1 easy glide, 2 work hardening and 3 doubly cross slip. These distinguished

stages reflect individual microscopic solid-state change due to deformation. As seen in Fig. 3, the stress suddenly increases and reach steady elongation showing low stress level in the whole stress strain curve. In this stage dislocation on the main slip plane works resulting gathered fine slip lines in parallel until the  $\varepsilon$  reaches about 0.1. At 2nd stage coarsening of the former slip lines occurs and dislocation loops accumulate to form subgrain boundary. Over  $\varepsilon$  0.6, i.e., in 3rd stage, slip bands and doubly cross slip appear on the surface. The doubly slip is attributable to corporative main and conjugated slips, which implies the occurrence of crystal bending and rotation. In actual tensile test the sample tends to rotate and bend at the slip planes to accommodate applied stress reduction.

In the insets of Fig. 3, typical surface molphlogies of deformed sample shows the feature of doubly cross slip and slip bands which treatment condition is indicated in the capion. Apparently, the molphlogy of doubly cross slip is very characteristic and at that stage, the sample is assured to be subjected to high strained condition.

With regard to experimental results, long-term electrolysis (for six months without any interruption of electrolysis) was conducted and the results obtained from microscopic observation of the postelectrolysis Pd electrode surface is summerlized as

- no surface crack, two faults, marked blisters or slip bands like a feather pattern to arrange each other in two arrays
- electrode interior: a row of defects, which inner surface was covered by differently colored layer and consisted of rounded humps like a nodule
- another surface molphlogy: doubly cross slip in the limited surface areas



Fig. 3 Schematic stress strain curve of single crystal f.c.c metal showing 3 stages. The upper right inset shows surface morphology of doubly cross slip and lower right that of slip bands  $^{8)}$ .



Fig. 4 SEM of a Pd electrode surface after long-term electrolysis in 0.1 *M* LiOD showing slip bands/blister (a) and doubly cross slip (b).

• many holes with increased population density approaching the fault lines

Figure 4 shows the molphlogies of slip bands and doubly cross slip. These two molphlogies are identified as same as those linked to the 3rd stage in the f.c.c single crystal stress strain curve. Hence, the Pd electrode could be locally experienced under high stress/strain condition. Since the Pd electrode was well annealled as pretreatment, and thereby crystal grain growth was throughly occurred resulting 4 big columnar crystals, the material could behave like a single crystal on the limited narrow areas. Then it is inferred that  $\varepsilon$  of the corresponding area is over 0.6 (this value is estimated from Cu; close to well annealed Pd because of both the similar modulus of elasticity).

On the other hand, the diameter of the postelectloysis electrode (designating whole Pd electrode's dimension) was measured at three positions in height, together with the microscopic observation. The dilations of the Pd electrode exhibited  $\varepsilon$  0.007 at first run and approached asymptotically to 0.0078 - 0.0083 during 2nd-4th run. These values are almost 10<sup>2</sup> times lower than that estimated above, which implys that the area subjected to high stress exhibiting high strain is characterized as 'local' because considerablly low strain was observed on the whole electrode.

### **3.2 Solid-state of Pd-hydrogen: crystal structure and electronic properties** *Pd-hydrogen lattice structure*

When a sample is negatively electrolyzed, it first becomes a solid solution in a lower concentration range; hydrogens occupy interstitial sites: octahedral and/or tetrahedral space in the f.c.c palladium lattice in both  $\alpha$ - and  $\beta$ -hydride phases. Although many in situ and ex situ x-ray measurements established the existence of hydrogen in the octahedral site, the tetrahedral site occupation long been a subject of debate in the literature <sup>9</sup>.

Theoretical calculation reveals that hydrogen occupation of octahedral sites is energetically favorable with the pure phases:  $x \le 0.008$  ( $\alpha$ -hydride) and  $x \ge 0.607$  ( $\beta$ -hydride) and with both phases coexisting: at intermediate values of  $x^{10}$ . Moreover, providing that hydrogen exists stably at the position between two atoms, which site meets geometrical criterion for size limitation of the H-occupied site, radius > 0.4 Å, thus, tetrahedral site could absorb considerably smaller radius atom than 0.4 Å (PdH<sub>0.6</sub>)<sup>11</sup>. Even though some researchers had insisted that the occupation of tetrahedral sites was reasonable as the results of solid-state experiments.

Neutron diffraction measurements have experimentally and theoretically identified the tetrahedral occupancy <sup>13</sup>. Therefore, in this study tetrahedral occupation has taken into account as the concurrence of a new state of hydrogen with  $x \ge 0.08$ , aside from such occurrence assuming complete occupation of the lattices with x > 1.

### *Electronic structures of Pd and Pd-hydrides* (Appendix 1)

The electrical resistance increases in nearly proportional to the hydrogen concentration x with the limited concentration range (see Fig. 8(b) of Chapter **3.3**). This shows significant change of electronic state of Pd-hydrogen system by hydrogenation. There has been some effort to study the electronic state of Pd and Pd-hydrogen systems. At first we describe the influence of hydrogen absorption on the electronic state of Pd, where Pd atomhydrogen interactions are considered in terms of the metal-hydrogen bonding band, the *d*-band shift and the shift of Fermi energy:  $E_F^{14}$ . Figure 5 pictorially represents DOS for

transition metals with f.c.c lattice structure together with  $E_F$ . For Pd (also Pt)  $E_F$  is located above the peak of most high 4*d*-bands. Since the  $E_F$  levels reach the top of the 4*d*-band, the vacant remaining 4*d*-band over  $E_F$  plays a role of *d*-hole at W and X points in 1st Brillouin zone. Thus, the charge carrier density: *n* of Pd is attributable to both the electron number density:  $n_e$  and the positive hole number density:  $n_h$ . The value of n is calculated as  $n_e + n_h = 0.6$  (per one electron). This means that Pd and Pt exhibit rather small conduction carrier density than Ag and Au, where  $n \approx 1$  because of their filled *d*-band and resulting *s-p* -band at  $E_F$ .

Figure 6 shows the DOS for Pd-hydrides (b), which indicates that the  $E_{\rm F}$  locates over the top of *d*-band. Major change from the DOS for Pd (a) is the shift of  $E_{\rm F}$  to higher electron energies and fixed at 5*s*-*d*-band <sup>15</sup>. In Fig. 6(b) there appeared additional electronic states related to



Fig. 5 Pictorial DOS curves for f.c.c transition metals. The arrows labelled as positive hole and electron are for Pd and Pt <sup>12</sup>).

the Hydrogen ions far below the  $E_F$ : at -1.0 ~ 2.0 eV. This is attributed to the bonding state originated from 5s and 4d state of Pd and 1s state of H where it can hold 2 electrons. However, the total electron number filled up to  $E_F$  is same as that of pure Pd: i.e., 10, and absorbed hydrogen donates partially 1s electron to the higher s-p band, resulting hydrogen ionized to H<sup>+</sup>. As a result, the electron donation by hydrogen reduces N(E) at  $E_F[N(E)$ : density of electronic states at the E]; the resistance of Pd-H increases as hydrogen concentration increases. On the other hand, Figure 7 shows the partial DOS of s-like character about o-site for PdH, which exhibits electron existence surrounding hydrogen site. Hydrogen (positively charged proton, in this case) inherently has been screened by conduction electron through the host metal: Pd 4d band, and the electron moves toward the lower bonding band, resulting weakening of screening effect. Then the conduction electron scattering with hydrogen increases, linked to the resistance increase.



Fig. 6 DOS of states of one spin for Pd and PdH. (a) for Pd; (b) for PdH.

### **3.3 Microstructural change of Pd** rod during repeated cathodic and anodic electrolysis in glycerinphosphoric acid

### First absorption of H in Pd (x < 0.8) in the C mode

Figure 8(a) shows the potential and dilation changes, and Fig. 8(b) shows plots of the resistance and apparent partial molar volume  $[d(\Delta l/l_0)/dx]$  changes as a function of x under the first C mode. In Figs. 8(a) and



Fig. 7 DOS of states of one spin of *s*-like character about the H site for PdH.

8(b) the concentration  $\alpha_{\text{max}}$  is the limit of the  $\alpha$  phase,  $\beta_{\text{min}}$  is the limit of the  $\alpha + \beta$ phase coexistence,  $\beta_{\text{tr}}$  is the transition from the  $\beta$  phase to  $\beta + \text{PdH}_{2-x}$ ,  $V_{\text{min}}$  is the onset of the  $\beta$ + void coexistence region, and  $R_{tr}$  is the transition from increasing resistance to a damped one. Figures 8(a) and 8(b) also show  $\alpha$  single,  $\alpha + \beta$  phase coexistence,  $\beta$  single and  $\beta$ + void coexistence regions. Shown below is the description focusing the coincidence among the potential, resistance, dilation and apparent molar volume.

As shown in Fig. 8(a), the potential decreases with increasing x (H/Pd ratio) within the single  $\alpha$  phase and reaches a constant value corresponding to the  $\alpha + \beta$  coexistence (two-phase coexistence region). Meanwhile, the resistance (see Fig. 8(b)) monotonically increases showing a slight deviation from exact proportionality to the x (see Chapter **3.2**). Although the resistance curve is not shown at very low concentrations, resistance rises steeply, and the slope of the resistance as a function of x changes to a small value at the onset of the  $\alpha + \beta$  phase coexistence. For x > 0.55 the slope in the resistance curve begins to decrease and levels off at the point of inflection ( $R_{tr}$ ). Furthermore, it approaches a limiting value of 1.8 for x > 0.80. This behavior appeared only in the first absorption of the C mode. The samples subjected repeated H absorption and desorption exhibited almost the same behavior up to the  $\beta$  phase, but above  $R_{tr}$  they flatten at 1.7 (see Fig. 9(b)). The difference in the resistance behavior above  $R_{tr}$  is attributed to the contribution of unrecovered absorbed H. Thus during the first absorption and desorption, a small volume of H remained trapped near the surface, whose interstitial sites are already occupied, do not, therefore, contribute to the resistance change <sup>16</sup>.

The values of apparent molar volume obtained from Fig. 8(b) are 1.64 cm<sup>3</sup>/mol for the  $\alpha$  single phase (this value was obtained from the dilation ( $\Delta l/l_0$ ) vs. x plot at x < 0.10) and 0.4 cm<sup>3</sup>/mol for the  $\alpha + \beta$  phase coexistence. For two-phase coexistence the values do not vary with increasing x, where the absorption presumably occurs in a homogeneous structure free from voids and cracks, in contrast to the repeated C mode, as described below. The potential in Fig. 8(a) shows Nernstian behavior ( $\alpha$  single phase), the constant value of the  $\alpha + \beta$  phase coexistence, the transition toward less noble direction ( $\beta_{\min} \sim V_{\min}$ ), and the next constant value of the  $\beta$ + void coexistence, where apparent molar volume follows two

constant values corresponding to the  $\alpha$  and  $\alpha + \beta$  phases coexistence (1.64 and 0.4 cm<sup>3</sup>/mol). At  $x < \beta_{\min}$  the coincidence between the phases defined by the potential ranges and the apparent molar volume of corresponding phases seems well enough.

In Fig. 8(b) a sharp rise of apparent molar volume to ~ 1.1 at x  $\beta_{tr}$  followed by a drop and subsequent slow decrease are shown at x >  $\beta_{min}$ . The peak of the apparent molar volume corresponds to the abrupt increase (at  $\beta_{min}$ ) and to the progressive damping in dilation. This peak is at the transition from apparent volume recovery region to the next state (designated as  $\beta_{tr}$ ). In Figs. 8(a) and 8(b) the onset of the potential shift  $\beta_{min}$  and that of the  $\beta$ + void



Fig. 8 The C mode potential, dilation, resistance, and apparent molar volume as a function x of the first C absorption at 40°C. (a) Potential ( $\Box$ ) and dilation ( $\blacksquare$ ) as a function of x; (b) resistance ( $\triangle$ ) and apparent molar volume ( $\diamondsuit$ ) as a function of x.

coexistence region  $V_{\min}$  coincide the onset and the end of the peak response of the dilation. Therefore, the less-noble potential shift ranging from -0.2 to -0.28 V is intimately related to anomalous H absorption, resulting in the sharp rise of the apparent molar volume. In other words, the potential shifts accompanied with the sharp rise of apparent molar volume, concurrently above  $R_{tr}$ , the resistance increases asymptotically toward 1.8 ( $\beta_{tr}$ ).

At  $x > \beta_{\min}$  the strange behavior of apparent molar volume accompanied with the potential shift is explained as

If the H absorption proceeds, like the first C mode, the potential might decreases in accordance with Nernst's equation and concurrently the apparent molar volume recover 1.64 cm<sup>3</sup>/mol (absorption in octahedral sites of Pd). However, the experimental data did not show ideal behavior; hence, the following new interpretation has drawn. Above  $\beta_{\min}$  predominant absorption of the octahedral occupation is disturbed by a precipitation of PdH<sub>2-x</sub> ( $0 < x \le 1$ ) due to the occupation of tetrahedral sites. Then through  $\beta_{tr}$ , the increased pressure (corresponding to decreased potential) allows the unstable precipitates to decompose to PdH<sub>1-x</sub> and H gas. On this occasion H evolution in the matrix causes void formation. Above  $R_{tr}$  (between  $\beta_{tr}$  and  $V_{min}$ ) the apparent molar volume drops concurrently with the transition from PdH<sub>2-x</sub> precipitation to absorption in voids when the resistance curve levels off (see Fig. 8(b)). Here, resistance did not respond to void formation because the size of void was larger than the mean free path of conduction electrons, which means that the resistance was not sensitive to the existence of voids. Such a new phase different from the  $\beta$  phase is proposed by Mizuno et al. <sup>17)</sup> and Storms <sup>18)</sup>.

### Repeated absorption and desorption of H in Pd (x<0.8) in the repeated C mode

The same absorption and desorption of H was repeated in the C mode (0.8 mm diameter, 50mm long, 7000s) where the electric charge of absorption was more than that required to attain x of 1.0.

Figures 9(a) and 9(b) show the absorption behavior of the 2nd and 3rd repetitions in the C mode. Comparing the values of the limiting  $\alpha + \beta$  phase coexistence ( $\beta_{\min} 2$ ,  $\beta_{\min} 3$ ) to that of the first C mode shows that these increase, in contrast to the first C mode, with an increasing number of repetitions. Furthermore, in the later stages of the  $\alpha + \beta$  phase coexistence, the dilation seems to be intermittent, showing abrupt drops in the apparent molar volume (see Fig. 9(b)), whereas the potential and resistance behave similarly to those described previously. Figure 9(b) shows the resistance and apparent molar volume of the 3rd repetition of the repeated C mode together with that of the first C mode (open square symbols). In the enlarged the  $\alpha + \beta$  phase coexistence region:  $\Delta(\beta_{\min})$ , the apparent molar volume decreased considerably, and the resistance increased with an increase in the *x*. The point at which the absorption-dilation correlation is temporarily broken off is termed the 'breathing mode'. It is interestingly noted that the breathing mode: it looks like absorption accompanied with irregular desorption, is occurred only when the sample experienced the desorption process.

During the repeated C mode, in the  $\alpha + \beta$  phase coexistence region, the H absorption proceeds inhomogeneously, showing an apparent molar volume of 0.6 to 0.8 cm<sup>3</sup>/mol and a subsequent lower apparent molar volume of 0.32 to 0.08 cm<sup>3</sup>/mol. The latter lower molar



Fig. 9 The C mode potential, dilation, resistance, and apparent molar volume as a function x of the 2nd and 3rd absorption at 40°C. (a) potential of 2nd ( $\Box$ ) and 3rd ( $\triangle$ ) absorptions, dilation of 2nd ( $\blacksquare$ ) and 3rd ( $\blacktriangle$ ) absorptions; (b) resistance ( $\blacklozenge$ ) and apparent molar volume of 3rd ( $\blacksquare$ ) and first ( $\Box$ ) absorptions.  $\Delta(\beta_{\min})$  denotes ( $\beta_{\min}$ ) incremental charge between 2nd and 3rd repetition.

volume suggests that H was absorbed in the high-density defect areas surrounding voids, which were introduced by the preceding absorption/desorption repetitions. The areas of high-defect density are characterized by the lower apparent molar volume, 0.32 to 0.08 cm<sup>3</sup>/mol, whose volumetric ratio tends to increase with above an increase in the number of repetitions. As a result, above  $\beta_{min}$  the volumetric ratio of voids (3rd repetition) was 1.6%, assuming the corresponding dilation is purely due to void formation.

During the C mode, *in situ* measurement of these variables was necessary for a comprehensive understanding of the microstructural changes due to H absorption/desorption. *Effect of number of repetitions on void formation in the repeated C mode with intervening anodic oxidation reaction* 

The values of  $\beta_{\min}$  and  $R_{tr}$  increased with repeated absorption and desorption where the incremental charge in the *x* corresponds to the charges for these voids reaching the equivalent H pressure. The resistance curves level off 1.7 at  $R_{tr}$ , and the potentials exhibit two constant



Fig. 10 Effect of number of repetitions on characteristic values of  $\beta_{\min}$  and  $R_{tr}$  at 40°C.

values corresponding to the  $\alpha + \beta$  phase and the  $\beta$ + void coexistence regions. Figure 10 shows the dependence of the values of  $\beta_{\min}$  and  $R_{tr}$  on the number of repetitions. These values tend to approach saturation, which does not yet appear in the figure. Since the first absorption does not require the corresponding charge, the incremental value from each repetition is equal to that of the summed absorption in the voids as hydrogen gas and high-density defect areas. Then the volumetric ratio required for voids is evaluated to be  $\approx$  9, assuming the equivalent H pressure 0.05 x 10<sup>5</sup> Pa, while those obtained from the dilation as a function of *x* curves range from 1.6 to 2.4%. Hence, the incremental amount of the *x* is attributable to the charge by absorption in high-defect density areas having lower apparent molar volume.

# 3.4 Microstructural analysis for the $\alpha' + \beta'$ phase coexistence region from *in situ* small punch test and the knowledge of hydrogen embrittlement

The aforementioned result as well as the first C mode elucidated that above  $\beta_{\min}$ , the microstructural change was void +  $\beta$  coexistence phase formation through the transient precipitation of PdH<sub>2-x</sub> (0 < x  $\leq$  1). In this chapter, in the  $\alpha + \beta$  phase coexistence region: less than x  $\beta_{\min}$ , the microstructural change could be accounted for by combining with the well-established knowledges of hydrogen embrittlement.

Figures 11(a), (b) and (c) show a typical PCT diagram indicating Inflection point:  $C(DB_{inf})$  (a); the in situ measurements of potentials of 2nd and 3rd repetitions and 3rd apparent molar volume (b), and the evolution of various phases and void during the repeated C mode (c), as a function of x. In the figures the concentration ranges for two characteristic phase regions based on the 3rd repetition curves are shown as red brackets for the  $(\alpha' + \beta)$  phase coexistence: x  $\alpha_{max} \sim \beta_{min}$  and ( $\beta$  + void) coexistence: x  $\beta_{min} \sim V_{min}$  for latter discussion.

Matsumoto et al.<sup>19)</sup> reported that Ductile-to-Brittle Transition Hydrogen Concentration (DBTC) for Nb, V and their alloys was useful for the analysis of hydrogen embrittlement using the *in situ* small punch (SP) testing apparatus equipped with a gas-flow system. It was found that the fracture morphology of the group 5 metals exhibited ductile manner with an increase of hydrogen pressure, while it changed to brittle one over the threshold hydrogen concentration (DBTC). The systematic study of DBTC showed that that value was close to the concentrations of inflection point of given metals' PCT diagrams. In Figs. 11(a) and (b), the inflection point (abbreviated as C(DB<sub>inf</sub>)) is indicated schematically in a given PCT diagram and the *in situ* measured potential and apparent molar volume plots are also shown for comparison. Applying the (DBTC) criterion to the repeated C mode, inflection point of Pd-hydrogen is conjugated to the midpoint *x* of the plateau for the  $\alpha'+\beta$  phase coexistence region (see C(DB)<sub>inf</sub>) in Fig. 11(b)).

In Fig. 11(b), during hydrogen absorption at the low concentration the single  $\alpha$  phase



Fig. 11 Schematics of a typical PCT diagram indicating Inflection point:  $C(DB_{inf})$  (a); the in situ measurements of potential of 2nd (double dotted line) and 3rd (dotted line) repetitions and 3rd apparent molar volume (solid line) (b), and the evolution of various phases and voids during the C mode (c) as a function of x.  $\Delta(\beta_{min})$  denotes ( $\beta_{min}$ ) incremental charge between 2nd and 3rd repetition.

holds up to  $x \ \alpha_{\text{max}}$ , and subsequently in the two-phase region, the volumetric ratio of the  $\beta$  phase ( $x \ \beta_{\text{min}}$ ) increases attaining  $x \text{ C(DB}_{\text{inf}}$ ). On the phase transformation hydrogen moves from *o*-site to other sites resulting the  $\beta$  phase growth. If we assume nonequilibrium reaction (due to delayed H diffusion), the new phase evolved is characterized as 'with excess hydrogen':  $\alpha$ ' denoted in Fig. 11 (c). Hence, it is predicted that, the formed  $\alpha' + \beta$  phase might exhibit brittleness when the x exceeds C(DB)<sub>inf</sub>.

In the review of hydrogen embrittlement of steel, Nagumo <sup>20)</sup> suggests that the embrittlement is related with crystal instability; hence, the phenomenon occurs, e.g., on melting or reduced elasticity. Thermodynamically, such changes of materials are caused by exceeding some criterion of free energy due to the increase of vacancy or crystal defect density. It is also discussed about necessary conditions for amorphization of intermetallic compound by H absorption.

Combining above two literatures, over x C(DB)<sub>inf</sub>, in the  $\alpha' + \beta$  phase coexistence region, there might be evolved nonequlibrium  $\alpha'$  phase containing excess hydrogen and/or amorphous phase accompanied with high density of defects or vacancies. By repeating the discussion, the formation of the  $\alpha' + \beta$  phase coexistence occurs exhibiting brittleness; i.e., from the (DBTC) criterion, and the  $\alpha'$  phase means nonequlibrium phase. From Nagumo's suggestion, since the  $\alpha' + \beta$  phase coexistence exhibits brittleness, this matrix has the property of crystal instability, which leads to the matrix with high density of defects or vacancies.

In addition, this new phase somewhat holds lattice mismatch resulting surrounded by strain field. This strain field might play a role of stabilization of hydrogen absorbed.

As discussed above, in Fig. 8(b) the apparent molar volume of the first absorption shows that the absorption occurs in a homogeneous structure free from voids and cracks. While the apparent molar volume of 3rd absorption (see Fig. 11(b)) increases until almost midpoint x of the plateau for the  $\alpha' + \beta$  phase coexistence region; followed by a decrease from 0.8 to 0.6 and lower hump from 0.32 to 0.08. Therefore, during the latter half the coexistence region, the decrease of apparent molar volume implies the evolution of nonequilibrium  $\alpha'$  phase, amorphous phase and areas with high density of defects or vacancies. This result is consistent with the analysis of the full width at half-maximum [FWHM] of the 220 reflection measured by *in situ* neutron diffraction, indicating that deuterium absorption introduces a significant strain in the hydride Pd <sup>21</sup>.

Let us think of the (DBTC) that is the phenomenon occurred with some transition from one state to another one. Hence, an evolution of brittle fracture necessitates the gradual change of material linked to the evolution of macroscopic material degradation. On this point, the above nonequilibrium  $\alpha$ ' phase containing excess hydrogen and/or amorphous phase might start at x  $\alpha_{max}$  (see Fig. 11 (C)). It is noted that the amorphous matrix probably alters to core possessing higher H content surrounded by circumference. The matrix might show reduced elasticity resulting brittle fracture.

Coincidence of two hydrogen states in the repeated C mode with the characteristic hydrogen states defined by defects induced by the interaction of hydrogen and applied stress

H content		← low	$(\beta_{\min})$ high $\rightarrow$		
Two phase	Туре	$(\alpha' + \beta)$ phase coexistence	(void $+\beta$ ) coexistence phase		
coexistence during	Absorpti	Dislocation, vacancy or vacancy	Void + high-density defect		
C mode (This	on site	cluster; Brittle	areas		
study)					
	Corres- pondenc	Î	Î		
	e	↓ ↓	Į,		
	Туре	Peak 1	Peak 2		
Two H desorption		Weak trap site (diffusible)	Strong trap site (non		
peaks in TDA profile	Trap site		diffusible)		
		Brittle (H-induced crystal			
		defect)			

Table 1 Two hydrogen desorption peaks in TDA profile and two phase coexistence during the C mode. The correspondence between two experimental results is indicated as arrows.

The study of hydrogen states by Thermal desorption analysis (TDA) combined with SIMS etc. elucidated that two desorption peaks of hydrogen were observed; a peak (termed as peak 1) at low temperature (100°C or < 200°C) and another one (peak 2) at more than 300 ~ 500 °C in a H desorption profile. Further analysis for hydrogen states clarified that peak 1 diffusible, i.e., spontaneous desorption around 30°C; peak 2 non diffusible at ambient temperature. The peak 1 is characterized as weak trap site, in other words, trapped in the shallow trap potential; the peak 2 strong trap site; in the deep trap potential. Table 1 summarizes two H desorption peaks (peak 1 and peak 2) and our two (the  $\alpha' + \beta$  phase coexistence) and (void +  $\beta$  coexistence phase) where the correspondence of two H peaks and our two phase coexistence states is evaluated. Noting that peak 1 considerably affects H embrittlement susceptibility, while peak 2 exhibits no effect on the embrittlement <sup>24</sup>.

Here we are interested in determining which corresponds to above mentioned two-phase coexistence regions, peak 1 or peak 2. For this issue it is important to know the experimental results shown as

Application of elastic stress on hydrogenated steel enhances hydrogen embrittlement, which is attributable to the formation of hydrogen-induced lattice defects. Under this condition, hydrogen is diffusible and trapped with dislocations and vacancies or vacancy clusters. Furthermore the repetitions of elastic stress applications affect the brittleness due to an increase of hydrogen-induced lattice defects<sup>23</sup>.

In Fig. 11(b) during the repeated C mode, there appeared two characteristic ( $\alpha' + \beta$ ) phase and (void +  $\beta$ ) coexistence regions, where the former phase transformation proceeds accompanied with high density of defects or vacancies. Such change of materials might exhibit brittleness. Furthermore the repetition of the C mode shows the enlargement of the  $\alpha$ + $\beta$  phase coexistence region and the incremental charge in the x (see Fig. 10) corresponds to the charges for these voids' saturation and that by absorption in high-defect density areas.

Thus, in the ( $\alpha' + \beta$ ) phase coexistence of our data, the brittleness, the enlargement of

the corresponding region during the C mode repetitions, and hydrogen absorption accompanied with high density of defects or vacancies, those of which are coincident with hydrogen state corresponds to peak 1; diffusible and trapped in dislocations and vacancy or vacancy clusters.

On the other hand, taking into account consequence in the hydrogen absorption process (see Table 1), the latter (void +  $\beta$ ) coexistence region might correspond to the hydrogen state of non diffusible (peak 2). This peak is attributable to hydrogen strongly trapped by dislocations in the ferrite/cementite(precipitate) boundary. If we accept void as a phase, absorption of hydrogen in high-defect density areas might coincidence with the situation of peak 2. Thus, high-defect density areas around voids correspond to dislocations and vacancies



Fig. 12 Effect of NH<sub>4</sub>SCN content on H content. Total (dashed line), peak 1 (dotted line) and peak 2 (solid line) are determined from TDA profile. Lateral axis indicates that the relative level of H absorbed corresponds to the NH<sub>4</sub>SCN content used for H charging <sup>24</sup>.

surrounding precipitates/matrix. These results are schematically illustrated below in Fig. 14. *Microscopic mechanism of the breathing mode during hydrogen absorption/desorption* 

Figure 12 shows the schematic representation of H content during H absorption where the values of peak 1 and peak 2 are obtained from the summation of released H at the corresponding temperature ranges. In Fig. 12, Doshida et al. <sup>24)</sup> reported that hydrogen absorption proceeded superiorly in peak 2 (strong trap site), after its saturation, in peak 1 (weak trap site). This means that during the 2nd repetition, in early stage, peak 2; i.e., voids' saturation and absorption in high-defect density areas proceed superiorly, soon after the charging of peak 1 becomes dominant. Thus, in the ( $\alpha' + \beta$ ) phase coexistence two kinds of different processes concurrently occur, and the process proceeds via not cascade reactions but two parallel reactions, each of which occurs superiorly depending on x.

The concept of above shown might be helpful to understand the phenomenon of 'breathing mode' of the apparent molar volume described above.

The breathing mode is seen in the relationship of resistance and dilation as a function of x, where x is limited within the  $\alpha$  and the  $\alpha + \beta$  phase coexistence regions (see Figs. 9(a) and (b)). As shown in Figs. 8(a) and (b), during the first C mode, the resistance increases in nearly proportional to the hydrogen concentration x and similarly the dilation increases corresponding to x. For apparent molar volume the value is calculated from the slope of the dilation vs. x plot, and hence, it exhibits constant in the  $\alpha$  and the  $\alpha + \beta$  phase coexistence regions. As shown in Fig. 9(b), during the repeated C mode the 3rd resistance and apparent molar volume do not exhibit such linear dependence on x, where the absorption-dilation correlation is temporarily broken off. As schematically shown in Figs.14(b), apparent molar volume changes irregularly (see red bracket:  $(\alpha' + \beta)$ ) and experimental reproducibility is lacking. The author has been suffered by no coincidence between the dilation and resistance

behavior, especially less than x  $\beta_{\min}$ . It is inferred that microstructural changes during desorption might play a role on such phenomenon. To analyze the real phenomenon occurred on the postelectrolysis electrode, the concept of parallel processes will be considered.

# **3.5** Summary of microstructural changes by taking into consideration of desorption process

Following the above Doshida's result, it is found that hydrogen desorption at first proceeds from the weak trap site followed by from the strong trap site, in contrast to the absorption. Although with respect to desorption process we are not fully understood; what happens in microstructural change during the repeated C mode, the schematic of sequential change of the total charges (defined as the summation of the enlarged (void +  $\beta$ ), and the  $\alpha$  and the  $\alpha' + \beta$  phase coexistence region) can be derived, as shown in Fig. 13. Then during the first C mode H absorption proceeds without void introduced by the preceding run, and the charges of the enlarged (void +  $\beta$ ) coexistence region become decreased as the repetition number increases. It can be also seen that the total charge of the repeated C mode approaches saturation.

Figures 14(a) and (b) show the schematics of microstructural changes (shown in the insets) and the in situ measurements of potential and apparent molar volume of 1st (a); and those of 3rd repetitions (b); as a function of x. In Figs.14(a) during the first H absorption, the inset is the microstructure without void in the  $\alpha + \beta$  phase coexistence region, and that with small volume of voids in (void +  $\beta$ ) coexistence region. In Figs. 14(b) the microstructures show the H absorption of matrix with small volume of voids and large volume of voids in the corresponding two regions. Furthermore, we consider the repeated C mode absorption followed by desorption, where hydrogen adsorbed once is desorbed electrochemically. For the diffusible hydrogen, the adsorbates was trapped in the field of tensile stress, which exists



Fig. 13 Effect of number of repetitions on the total charge of H absorption, which is composed of enlarged (void +  $\beta$ ), and  $\alpha$  single and  $(\alpha'+\beta)$  coexistence regions.



Figs. 14(a) and (b) Schematics of the in situ measurements of potential (dotted line) and apparent molar volume (solid line) of 1st (a); and those of potential (dotted line) and apparent molar volume (solid line) of 3rd repetitions (b); as a function of x.  $\Delta(\beta_{\min})$  denotes ( $\beta_{\min}$ ) incremental charge between 2nd and 3rd repetition. For clarification, the longitudinal axis of Figs. 14(b) has been here reduced.

in a weak trap potential. On the occasion of desorption of repeated absorption/desorption cycles such hydrogen easily diffuse out of matrix, subsequently, defects introduced during former absorption are remained. This microscopic matrix change is not shown in Figs. 14(a) and (b).

Next our approach is addressed to the followings,

- Comprehensive understanding of resistance and dilation behavior
- Estimation of stable micro void, with respect to hydrostatic pressure and hydrogen content
- Microstructural change during desorption.

### 4 Conclusion

We summarize our study as follows:

- 1. Long-term electrolysis for well annealed thick Pd rod (9.0 mm diameter) in 0.1 M LiOD was performed. Microscopic observation of the postelectrolysis Pd electrode showed the surface morphology of slip bands and doubly cross slip. These two characteristic morphology are identified as same as those appeared on the sample surface during tensile test. It is shown that the limited areas of Pd electrode was subjected to high stress and high strain over  $\varepsilon$  0.6.
- 2. In situ potentiometric, resistance and dilatometric measurements of Pd electrode were conducted to elucidate the elemental steps of electrochemical hydrogen absorption. During the first C mode, the structural phase changes occurred where the  $\alpha$  phase, the ( $\alpha$ + $\beta$ ) phase coexistence, fine PdH<sub>2-x</sub> precipitates (void formation) and finally the ( $\beta$ + void) evolved.
- 3. During the repeated C mode the enlarged the  $(\alpha+\beta)$  phase coexistence region was measured, where the charges of the corresponding region exhibited an increase with an increase in the number of repetitions.
- 4. During the repeated C mode, the absorption proceeds via the  $\alpha$  single phase, the ( $\alpha' +\beta$ ) phase coexistence region and (void +  $\beta$ ) coexistence region; the former involves nonequlibrium  $\alpha'$  phase and/or amorphous phase accompanied with high density of defects or vacancies.
- 5. According to the suggestions with hydrogen embrittlement, the findings of this study as follows:

Correspondence between our C mode data and TDA analysis revealed that the matrix in the  $(\alpha' + \beta)$  phase coexistence region might exhibit brittleness, and was coincident with hydrogen state corresponds to peak 1; diffusible and trapped in dislocations and vacancy or vacancy clusters.

It is important to consider that during hydrogen absorption/desorption two parallel reactions proceed, each of which occurs superiorly depending on x.

6. These phase transformations and nonequilibrium reactions were reasonably evaluated by the comprehensive understanding of potential, dilation and resistance behaviors.

### Acknowledgements

The author would like to express sincere thanks to Mr. T. Ooi, Mr. A. Nakajima and Professor I. Ohno of Tokyo Institute of Technology for co-operative work. He thanks S. Numata for the preparation of conference fee.

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### Appendix 1

The electrical resistivity ( $\rho$ ) is given as

 $\rho = \mathrm{m}/n \mathrm{e}^2 \tau$ 

(1)

where e is elementary electric charge, m is electron mass, n is number density, and  $\tau$  is mean free time. In the case of metal the resistivity is mainly determined by  $\tau$  because *n* do not change much among metals. Since  $\tau$  is defined as mean free time, during which electron freely move in one collision to next one under applied electric field, therefore, it is influenced by N(E) around  $E_F$ .

# Progress of density functional methods in LENR and their problems

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### Abstract

The properties of hydrogen-metal systems strongly depend on the charge distribution around hydrogen. Therefore density functional calculations are powerful method to discuss them. Recently, we calculate the states of deuteron in solids based on the method of Y.E.Kim as a problem of charged bose particles in metals. In this study, we show some results reported in ICCF20 and discuss the problems for the convergence aspects of the iterative calculations.

(keywords; density functional theory, bose particle, low energy nuclear reaction)

### 1 Introduction

The well known way to describe many-body problems are Hartree Fock method using Slater determinant. However, it is so hard to perform numerical calculations. On the other hand, the density functional formalism, which is developed by Hohenberg, Kohn and Sham at first [1,2], is easy to do calculations. They obtained self-consistent equations including exchange and correlation effects. These equations are very useful because we can treat the many-electron system as if it were one body problem. For example, metal hydrogen systems can be treated by regarding the hydrogen as external potential to the electron gas.

In our previous works [3–5], we discussed the quantum distribution of hydrogen or deuteron by using the method of Y.E.Kim et al.[6,7]. Our equations are written in the density functional styles. In order to obtain the numerical solutions to them, we introduced doubly connected flow chart, which is drown in Fig.1 for the case of two hydrogen atoms in metal. As shown in Fig.1, it is an iterative calculation including Schrödinger and Poisson equations. The results of our previous works were obtained by using this flow chart.

In this study, we review previous results in ICCF20 [8] and discuss the problems for the convergence aspect of the iterative calculations. At the end, we discuss the method how to estimate the nuclear reaction rate between two charged bose particles.



Fig.1 The flow chart for the self-consistent calculation. The densities  $n_i$  and the electro-static potentials  $W_i$  are obtained alternatively by using Eq.(1) and Eq.(2). The self-consistencies should be checked on the two loops in this diagram connected in parallel.

### 2 Theory

Here, we briefly introduce Kim's theory [6,7]. In their study, mixture of charged bosons confined in harmonic traps and Bose-Einstein condensation mechanism are considered. For the two species of charged bose particles i (= 1, 2), the Schrödinger equation is written as

$$\left\{-\frac{\hbar^2}{2m_i}\nabla^2 + \frac{1}{2}m_i\omega_i^2r^2 + W_i(\mathbf{r})\right\}\psi_i(\mathbf{r}) = \mu_i\psi_i(\mathbf{r}),\tag{1}$$

where  $W_i(\mathbf{r})$  and  $\mu_i$  mean electro-static and chemical potentials, respectively.

Based on their formalism, we proposed the equation to calculate electro-static potentials. They are described by using the integral form of Poisson's equation, which is written as

$$W_i(\mathbf{r}) = \int d\mathbf{r}' \frac{\exp(-k|\mathbf{r} - \mathbf{r}'|)}{4\pi |\mathbf{r} - \mathbf{r}'|} \left[ \frac{e^2}{\varepsilon_0} Z_i \left\{ Z_j n_j(\mathbf{r}') \right\} + k^2 W_i(\mathbf{r}') \right], \tag{2}$$

where  $Z_i e$ ,  $\varepsilon_0$  and k mean effective charge of species *i*, dielectric constant of vacuum and arbitrary constant, respectively. In Eq.(2),  $W_i(\mathbf{r})$  is written in a form of a density functional formalism, because the integrand depends on the density  $n_j = |\psi_j|^2$ . If we assume the functions  $n_i(\mathbf{r}')$ ,  $n_j(\mathbf{r}')$  and  $W_i(\mathbf{r}')$  in the integrand of the right hand side of Eq.(2) as old quantities,  $W_i(\mathbf{r})$  in the left hand side can be regarded as a new quantity in the recursive calculations. We can easily understand that these procedures include doubly connected equations.

For the numerical calculations, Eq.(1) is translated into non-dimensional form as

$$\left\{-\frac{d^2}{d\xi_i^2} + \xi_i^2 + v_i(\xi_i)\right\} u_i(\xi_i) = \varepsilon_i u_i(\xi_i), \qquad (3)$$

where  $\xi_i = \alpha_i r$ ,  $\alpha_i = \sqrt{m_i \omega_i / \hbar}$ ,  $\varepsilon_i = 2\mu_i / \hbar \omega_i$ ,  $v_i = 2W_i / \hbar \omega_i$  and  $u_i(\xi_i) = \xi_i \psi_i(\xi_i)$ . The initial conditions at  $\xi_i = 0$  for solving Eq.(3) are given by two types of equations. They are written as

$$u_i(0) = 0, \qquad u'_i(0) = 1$$
 (4)

$$u_i(0) = 1, \qquad u'_i(0) = 0.$$
 (5)

The first conditions express the states that have high probability around the origin, while the others expresses the low probability states around the origin.

### 3 Results and Discussions

Here, we consider two examples for the system including one D<sup>+</sup> (i=1) and one <sup>6</sup>Li<sup>+</sup> (i=2). The trapping frequencies for D<sup>+</sup> and <sup>6</sup>Li<sup>+</sup> are 9.59 × 10<sup>17</sup>sec<sup>-1</sup> and  $5.54 \times 10^{17}sec^{-1}$ , respectively. In Fig.2, we show a result whose initial conditions at  $\xi = 0$  for D<sup>+</sup> and <sup>6</sup>Li<sup>+</sup> are Eq.(4) and Eq.(5), respectively. In Fig.3, we show a result whose initial conditions at  $\xi = 0$  for D<sup>+</sup> and <sup>6</sup>Li<sup>+</sup> are Eq.(5) and Eq.(4), respectively. In Fig.4, we show a result for the system including two D<sup>+</sup> 's trapping at a harmonic potential. The initial conditions at  $\xi = 0$  for one D<sup>+</sup> and other D<sup>+</sup> are Eq.(5) and Eq.(4), respectively. In the numerical calculations, electro-static potentials are normalized by  $v_i = 2W_i/\hbar\omega_i$  and distances from the bottoms of the harmonic potentials are normalized by  $\xi_i = \alpha_i r_i$ 

In Fig.2(a), Fig.3(a) and Fig4.(a), the rapid convergences of energy eigenvalues are not seen. These aspects depends on the complexity of the doubly connected flow chart shown in Fig.1. If the number of the particle is more than two, it becomes highly complex chart and the convergence becomes slower. In order to get rapid convergence, we should reconsider the accuracy of the numerical calculations. For example, the method of the numerical integration in Eq.(2) should be updated especially for the large  $|\xi|$  region. As the second option, we should reconsider the method for finding the binding energies. In this study, the solution for large  $|\xi|$  is checked if it matches to the asymptotic solution by using Newton method. In this process, true eigenvalue may be passed for very rapid change.

Seeing Fig.2(d)(e), Fig.3(d)(e) and Fig.4(d)(e), the regions of overlapped density exist in each figure. This means that we can calculate the probabilities of the nuclear reactions between the two particles. In near future, we will do them and report them.



(a) Binding energy versus iteration.



Fig.2 Results for the system including a  $\rm D^+$  (i=1) and  $^6\rm Li^+$  (i=2).



(a) Binding energy versus iteration.



Fig.3 Results for the system including a  $D^+$  (i=1) and  ${}^6Li^+$  (i=2).



(a) Binding energy versus iteration.



Fig.4 Results for the system including two deuteron atoms.

### 4 Conclusions

The density functional formalism naturally includes probability distributions of the particles. This is a merit of this method, because we need distributions of charged particles in order to get nuclear reaction rates. Therefore, we selected it as a tool for the study of LENR. As a result, we found that the rapid convergences could be obtained for high frequency harmonic potentials.

In our calculations, we assumed extremely high frequency harmonic potentials. In this case, deuteron is trapped in narrow space and the overlapping of the wave functions for both particles becomes large. However, the frequencies we used here are too large to be generated in the ion traps [9]. We should do further considerations for such hypothetical high frequencies.

Here, we summarize conclusions as following three points.

- (1) We found a new method how to calculate the quantum distribution of some charged bose particles trapped at a harmonic potential, based on Kim' theory. Our theory is written in the form of the density functional formalism.
- (2) In our method, the convergences were not always good because of the complexity of the flow chart.
- (3) In near future, we will calculate nuclear reaction rate from the probability density of each particle.

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### Model Mechanism of OHMASA-GAS Related to Water Clusters

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Abstract: One of the hydrogen-including oxy-hydrogen gases called OHMASA-GAS (Oxygen-Hydrogen Mixing Atomic Symmetrized Aeration-Gas) is obtained through the electrolysis of water under vibratory agitation by vibration blades and can be stored as the hydrogen (H) and oxygen (O) mixture gas. In recent years, it has been reported that titanium (Ti), tantalum (Ta), tungsten (W) metal or glassy carbon (C) melts and vaporizes instantly at a temperature of about 3000°C or over when it comes into contact with the flame of burning this gas of a temperature range of 600 to 700°C, and various elements are generated under vibratory agitation of the electrolytic solutions with alkali or alkaline-earth metal ions such as magnesium (Mg), calcium (Ca) or cesium (Cs), and so on. In order to investigate these properties and some other related phenomena, small water cage clusters which encapsulated H atoms, and large water tube clusters with Mg atom were simulated on a personal computer by using a computer simulation program MOPAC based on the semi-empirical molecular orbital method. OHMASA-GAS seems to be generated by stable cavitation, that is, small oscillating spherical bubbles with the nuclei of small water cage clusters, which are made by division of the large cylindrical bubbles, encapsulate H atoms produced by the electrolysis of water. The generation of various elements under vibratory agitation related to the generation of OHMASA-GAS seems to be caused by transient cavitation, that is, collapse of a part of the large cylindrical bubble with the nucleus of large water tube cluster formed at the edge face of the vibration blade.

Keywords: computer simulation, water cluster, OHMASA-GAS, generation of various elements

### 1. Introduction

There are some clathrate hydrates of regular dodecahedron (icosamer water cluster  $(H_2O)_{20}$ ) which encapsulate hydrogen (H) atoms and compose solid-state (some kind of ice) surrounded by other polyhedrons in the pressurized water. On the other hand, there are some H-including oxy-hydrogen gases, and one of them is called OHMASA-GAS (Oxygen-Hydrogen Mixing Atomic Symmetrized Aeration-Gas) which is obtained through the electrolysis of water under vibratory agitation by vibration blades. Some interesting properties of this gas have been reported that, for instance, some metals or glassy carbon melts and vaporizes instantly at high temperatures when it comes into contact with the flame of burning this gas <sup>1,2)</sup>. Some
properties of neutral electrolyte and some phenomena of generation of various elements under vibratory agitation related to the generation of this gas have been also reported.

# 1.1 Properties and Phenomena

Those properties of OHMASA-GAS and neutral electrolyte, and those phenomena of generation of various elements related to the generation of OHMASA-GAS are as follows:

- a) Properties of OHMASA-GAS
- OHMASA-GAS is obtained through the electrolysis of water added like potassium hydroxide (KOH) under vibratory agitation by vibration blades with a frequency of about 40 Hz.
- This gas is made of mixing the gas including H from cathodes and oxygen (O) from anodes without separator, and can be stored at a high pressure.
- This gas is stored in stainless bottles without hydrogen embrittlement.
- This gas is liquefied at a temperature of -178.7°C (oxygen: -183°C, hydrogen: -252.6°C), but it is not solidified at a temperature of -255°C (oxygen: -218.4°C, hydrogen: -259°C) even at a pressure of 25 MPa<sup>1, 2)</sup>.
- This gas is composed of the multimer water clusters of number about 10 to 20, the mode of which is around 14 or 15 and the maximum is 27 according to the analyzation by a liquid-ionization tandem mass spectrometry.
- This gas burns in the same way as H gas, but it is safety, for example, without explosion inhaling the flame.
- Titanium (Ti), tantalum (Ta), tungsten (W) metal or glassy carbon (C) melts and vaporizes instantly at a temperature of about 3000°C or over <sup>3)</sup> when it comes into contact with the flame of burning this gas of a temperature range of 600 to 700°C.
- Carbon dioxide (CO<sub>2</sub>) forms new combustible compounds by mixing with this gas.
- Hydrocarbon such as LNG (liquefied natural gas; mainly methane  $CH_4$ ) and propane ( $C_3H_8$ ) improves combustion efficiency by mixing with this gas.

• An early voltage of fuel cell with this gas is about 5% higher than that with only H gas, however it descend at a temperature higher than  $60^{\circ}C^{4}$ .

• When H gas is added to this gas, the liquefaction temperature becomes higher than that of H gas, and then H can be stored safely. Although this H added gas raises the total heat quantity, it needs to supply oxygen or air for the perfect combustion of it, and Ti, Ta, W metal or glassy C cannot melt and vaporize with burning of it, and the voltage of fuel cell with it becomes that with H gas.

b) Properties of Neutral Electrolyte

- Neutral electrolyte is obtained through the electrolysis of water added very small amount of salt (NaCl) under vibratory agitation by vibration blades with a frequency of about 40 Hz.
- The neutral electrolyte has the mild and continuous sterilizing properties.
- c) Phenomena of Generation of Various Elements under Vibratory Agitation
- Light elements like magnesium (Mg) and heavy elements like zinc (Zn) are generated under vibratory agitation of pure water by palladium (Pd) plating vibration blades with a frequency of about 160 Hz.
- Slightly heavy elements like phosphorus (P) are generated under vibratory agitation of magnesium chloride (MgCl<sub>2</sub>) solution by vibration blades with a frequency of about 160 Hz.
- Heavy elements such as nickel (Ni), copper (Cu), silver (Ag), platinum (Pt) and gold (Au) are generated under vibratory agitation of chloride solution such as calcium chloride or cesium chloride (CaCl<sub>2</sub> or CsCl) solution by vibration blades with a frequency of about 160 Hz. (Appendix) In regard to radioactive cesium (Cs), there is a possibility that the radioactively contaminated water, which is leaked from Fukushima Daiichi Nuclear Power Plant, is treated to reduce the radioactivity.

# **1.2 Suppositions of Generation Processes**

OHMASA-GAS and the neutral electrolyte would be generated by stable cavitation <sup>5)</sup>, that is, the small oscillating spherical bubbles with the nuclei of small water cage clusters which are made by division of the large cylindrical bubble and encapsulate H atoms or bond chlorine (Cl) atom produced by the electrolysis of water or NaCl solution, respectively. The generation of various elements under vibratory agitation related to the generation of OHMASA-GAS would be caused by transient cavitation <sup>5)</sup>, that is, the collapse of a part of the large cylindrical bubble with a nucleus of large water tube cluster, which is formed at the edge face of the vibration blade. Therefore, the following unified processes are supposed:

a) Generation of OHMASA-GAS

- Water sheet clusters are formed on the surfaces of vibration blades in the pressurized water under vibratory agitation of solutions like potassium hydroxide (KOH) with a frequency of about 40Hz.
- Large water tube clusters are made from the water sheet clusters which are moved to the edge of surface of the vibration blade by water stream and partly peeled off by change of water pressure under vibration agitation.

- Large cylindrical bubbles are formed with the nuclei of those water tube clusters at the edge face of the vibration blade in a low water pressure.
- When the vibration blade moves in the opposite direction, the cylindrical bubbles separate from the edge faces of the vibration blades in a high water pressure and they are divided into small spherical bubbles with divided nuclei of water cage clusters.
- The similar phenomena occur on the other surface of the vibration blade in the opposite vibration phase.
- When the cylindrical bubbles are divided into the spherical bubbles, the nucleus of the water tube clusters are divided into small decamer, pentadecamer or icosamer base three-dimensional water cage clusters.
- Through the electrolysis of solution, H atoms are produced from cathodes and O atoms are produced from anodes, which enter into the spherical bubbles with nuclei of water cage clusters when the spherical bubbles touch on the cathodes or anodes.
- H atoms are encapsulated in the water cage clusters, and O atoms combine with  $H_2O$  of the other water cage clusters to form hydrogen peroxide ( $H_2O_2$ ).

b) Generation of Neutral Electrolyte

- Through the electrolysis of very thin salt (NaCl) solution replaced to the solutions like potassium hydroxide (KOH) under vibratory agitation (in the above-mentioned "a) Generation of OHMASA-GAS"), small spherical bubbles with nuclei of small water cage clusters are formed and Cl atoms are produced from anodes.
- Cl atoms enter into the spherical bubbles with nuclei of water cage clusters when the spherical bubbles touch on the anodes, and combine with OH to form hypochlorous acid (HClO) which bonds to the water cage cluster.
- c) Generation of Various Elements under Vibratory Agitation
- Water sheet clusters are formed on the surfaces of Pd plating or normal vibration blades in the pressurized water under vibratory agitation of pure water or solutions like magnesium chloride (MgCl<sub>2</sub>) with a frequency of about 160 Hz.
- Large water tube clusters are made from the water sheet clusters which are moved to the edge of surface of the vibration blade by water stream and partly peeled off by change of water pressure under vibration agitation.
- Large cylindrical bubbles are formed with the nuclei of those water tube clusters at the edge face of the vibration blade in a low water pressure.
- When the vibration blade moves in the opposite direction, a part of the cylindrical bubble collapses in a high water pressure.
- · When the part of the water tube cluster is compressed by inflow of surrounding

water molecules, H, O atoms and the alkali, alkaline-earth metal or another metal atom would collide with one another or collide against the edge face of the vibration blade, and becomes a high-temperature and a high-pressure.

- The similar phenomena occur on the other surface of the vibration blade in the opposite vibration phase.
- Both sides of the partly collapsed cylindrical bubble or not collapsed cylindrical bubbles separate from the edge faces of the vibration blades and they are divided into small spherical bubbles with divided nuclei of water cage clusters.
- When the cylindrical bubbles are divided into the spherical bubbles, the nucleus of the water tube clusters are divided into small decamer, pentadecamer or icosamer base three-dimensional water cage clusters.

# 1.3 Investigations in This Study

The above-mentioned each supposed process is very difficult to solve. Therefore, in this study some states which occur on the way of the above-mentioned processes were considered. Recently, the states of decamer base three-dimensional water cage clusters have been studied and reported <sup>6</sup>). Similarly, the states of the water tube clusters and pentadecamer or icosamer base water cage clusters were investigated by using a simulation program MOPAC. The states of one to three H atoms encapsulated in these water cage clusters were also investigated.

The result showed that the large water tube clusters of 25-50mer, the cross section of which is pentamer, exist. These large water tube clusters would be divided into small water cage clusters. Furthermore, the results showed that one to three H atoms are encapsulated in the small water cage clusters of much kind of shapes. H atoms encapsulate in these water cage clusters and O atoms combine with these water cage clusters would compose OHMASA-GAS. The large water tube clusters of 25-50mer were observed to exist with Mg or Cl atom. If some parts of these water tube clusters were strongly adsorbed at the edge faces of Pd plating vibration blades, or if these water tube clusters were made with alkali, alkaline-earth metal or other metal atoms, excess forces would act on a part of the cylindrical bubble easily to collapse. When the part of the cylindrical bubble collapses, the part of nucleus of water tube cluster would be compressed by inflow of surrounding water molecules, and H, O atoms and the alkali, alkaline-earth metal or another metal atom would collide one another or collide against the edge face of the vibration blade to become plasma state of a high-temperature of several thousand degrees and a high-pressure of several giga-pascals. However, we have not yet understood whether or how the compression of H, O atoms and the alkali, alkaline-earth metal or another metal atom of water tube cluster, or material atom of the vibratory blades cause any other reactions.

# 2. Calculations

# 2.1 Hardware and Software Used for Calculation

Calculations were carried out on a personal computer which had 4 cores / 8 ways CPU with 32 GB main memory by using the computer simulation program MOPAC (Molecular Orbital Program Package; Ver.6) based on the MP3 of semi-empirical molecular orbital method<sup>7, 8)</sup>. This program is working on the molecular computation support system program Winmostar<sup>TM 9)</sup> which assists in exchanging elements and coordinates, showing solid figures of water clusters and outputting graphs of oscillation or vibration analysis, and so on.

### 2.2 Calculated Water Clusters

The cubic ice can be formed in freezing confined aqueous systems differently from the hexagonal crystalline ice, and has the diamond structure composed of two face-centered cubic structures displaced from one another by one-quarter of a body diagonal. On the other hand, a lot of cavitation bubbles are formed by the vibration blades in the generation of OHMASA-GAS. Therefore, water sheet clusters seem to be stuck on the surfaces of vibration blades in a pressurized water under vibratory agitation by vibration blades and to be peeled off and rolled up to the large water tube clusters, and then the large cylindrical bubbles seem to be formed with the bubble nuclei of these water tube clusters at the edge faces of the vibration blades. When the large cylindrical bubbles separate from the edge faces of the vibration blades, they would be divided into small spherical bubbles with the nuclei of decamer, pentadecamer or icosamer base three-dimensional water cage clusters as transient cavitations. On the other hand, when a part of the cylindrical bubble with the nucleus of the water tube cluster collapses, a part of the water tube cluster would be compressed as another kind of transient cavitation.

Recently, the decamer base water cage clusters have been investigated <sup>7</sup>); in this study the other kind of water cage clusters and the water tube clusters are considered. It was investigated whether the large water tube clusters of 25-50mer and the small pentadecamer or icosamer base water cage clusters would exist, and whether H atoms attached on or encapsulated in these water cage clusters would be stable by calculating formation energy and bonding energy per water molecule of the water cage clusters, and how these H atoms could oscillate, by using the computer simulation program

MOPAC. In regard to adding CO<sub>2</sub>, hydrocarbon or H gas to OHMASA-GAS, and to the generation of neutral electrolyte, the states of decamer water cluster were simulated. Furthermore, in regard to the generation of various elements under vibratory agitation, the states of large water tube clusters with Mg or Cl atom were simulated, which came from MgCl<sub>2</sub> solution and combined with O atom or OH radical to form MgO or HOCl and bond to the water cage cluster, respectively.

There are so many structures of water cage clusters in even the same number multimer that calculations were carried out for only a few structures of each multimer. Moreover, H atoms relating to the hydrogen bonds of water clusters are known to move rapidly and change places in very short time, but in this study with personal computer this cannot be considered. Using MOPAC we calculated some basic energy of water molecules and water clusters as follows:

- The formation energy of water molecule: -223.5kJ/mol (Experiment: -241.8kJ/mol)
- The covalent energy of water molecule (one O-H): -544.654kJ/mol (Experiment: -492kJ/mol)
- The formation energy and the bonding energy per water molecule of icosamer (dodecahedron) water cage cluster (H<sub>2</sub>O)<sub>20</sub>: -26.3kJ/mol

# 3. Results and Discussion

By calculations of the computer simulation program MOPAC, the bases for some properties of OHMASA-GAS and the generation of neutral electrolyte were observed, and the possibility of the generation of various elements under vibratory agitation related to the generation of OHMASA-GAS was shown. These subjects of calculations are supposed to relate together by the water clusters of bubble nuclei formed by the vibration blades: The properties of OHMASA-GAS would rely on encapsulated H atoms in the small water cage clusters which are divided from the large water tube clusters. The neutral electrolyte would be made from the same small water cage clusters. The generation of various elements under vibratory agitation would be caused by the collapse of a part of the large water tube clusters.

#### 3.1 Formation of Water Tube Clusters

The large water tube clusters were supposed to be formed at the edge faces of the vibration blades under vibratory agitation, and also supposed to become the nuclei of large cylindrical bubbles of transient cavitation. The water tube clusters, the cross section of which was pentamer, were calculated by the PC, while those, the cross section of which was tetramer or hexamer, could not be calculated. The bonding energy

per water molecule of the water tube clusters of 25-50mer which have 5-10 pentamer cross sections was nearly equal to that of the water cage clusters shown in Fig. 3 in "3.2.1 Bonding Energy per Water Molecule of Water Clusters". Therefore, these water tube clusters, which are 2-4 nm long, seem to be formed at the edge faces of the vibration blades. The following shows examples of the water tube clusters, the cross section of which is pentamer (Fig. 1).





Fig. 1-a: (H<sub>2</sub>O)<sub>35</sub> (7 pentamer cross sections) Fig. 1-b: (H<sub>2</sub>O)<sub>50</sub> (10 pentamer cross sections) Red indicates O atoms, yellow indicates H atoms.

# 3.2 OHMASA-GAS

The large cylindrical bubbles of transient cavitation with the nuclei of water tube clusters of pentamer cross section were supposed to be formed at the edge faces of the vibration blades under vibratory agitation, and also supposed to be divided into the small spherical bubbles with divided nuclei of water clusters. When the divided water clusters have two, three or four pentamer cross sections, they would form decamer, pentadecamer or icosamer water cage clusters, respectively (Fig. 2). At that time, if one to several water molecules were removed from or added to them, many kinds of decamer, pentadecamer and icosamer base cage water clusters could be formed.



Fig. 2-a: Decamer [  $(H_2O)_{10}$  ] Fig. 2-b: Pentadecamer [  $(H_2O)_{15}$  ] Fig. 2-c: Icosamer [  $(H_2O)_{20}$  ] Red indicats O atoms, yellow indicates H atoms.

As the model of OHMASA-GAS, one to three H atoms produced by the electrolysis of water were considered to be encapsulated in the decamer, pentadecamer and icosamer base water cage clusters, and one O atom produced by the electrolysis of water was considered to combine with  $H_2O$  to form hydrogen peroxide ( $H_2O_2$ ) which bonded to another decamer, pentadecamer and icosamer base water cage clusters.

# 3.2.1 Bonding Energy per Water Molecule of Water Clusters

Fig. 3 shows the graphs of the bonding energy per water molecule of the decamer, pentadecamer and icosamer base water cage clusters which attach or encapsulate no H atom and one to three H atoms.



Fig. 3 Bonding Energy per Water Molecule of Water Clusters

Gray and black lines indicate the minimum and the maximum absolute values of the bonding energy per water molecule of decamer base water clusters which attach or encapsulate no H atom, respectively. Dark and light purple lines indicate the mean absolute values of the bonding energy per water molecule of pentadecamer and icosamer base water cage clusters which attach or encapsulate no H atom, respectively. Dark green, green and light green lines indicate the mean absolute values of the bonding energy per water and icosamer base water cage clusters which attach or encapsulate the mean absolute values of the bonding energy per water molecule of decamer, pentadecamer and icosamer base water cage clusters which attach or encapsulate one H atom, respectively. Dark red, red and light red lines indicate those which attach or encapsulate two H atoms, respectively. Dark blue, blue and light blue lines indicate those which attach or encapsulate three H atoms of triangle structure, respectively. Dark bluish-green, bluish-green and light bluish-green lines indicate those which attach or encapsulate three H atoms of slightly curved line structure, respectively.

The bonding energy per water molecule of symmetrical decamer and icosamer base water cage clusters looks to be larger (negative absolute value) than that of unsymmetrical pentadecamer base water cage clusters. The water clusters of from monomer to pentamer which exist usually in the ordinary water could attach H atoms on their surface through the electrolysis of water. On the other hand, the hexamer or larger water cage clusters would be formed under vibratory agitation, which could also attach H atoms on their surface and finally encapsulate H atoms in them to compose OHMASA-GAS through the electrolysis of water. The bonding energy per water molecule of decamer and icosamer base water cage clusters which encapsulate H atoms looks to be larger (negative absolute value) than those of decamer and icosamer base water cage clusters which do not encapsulate H atoms.

# 3.2.2 Oscillations of H Atoms Encapsulated in Water Cage Clusters

Vibrational spectra of infrared absorption of decamer, pentadecamer and icosamer base water cage clusters which encapsulate one to three H atoms were calculated. Oscillations of these H atoms appear at the nearly same wavenumbers through the all water cage clusters (Fig. 4). The infrared-wavenumbers absorbed by the oscillation of the center atom of three H atoms decrease from about 3500 /cm to about 2500 /cm when three H atoms are encapsulated in the water cage cluster.



Fig. 4 Infrared-Wavenumber Absorbed by H2, H3T (Triangle Structure) and H3L (Slightly Curved Line Structure) Oscillations

### a) Encapsulated One H Atom

When one H atom is encapsulated in the water cage cluster, no oscillation or vibration state appears in the wavenumber range of 1000 /cm to 4500 /cm except

for vibrations of water molecules at a wavenumber of about 1700 /cm for bending modes and 3800-4000 /cm for stretching modes (Fig. 5).





#### Red indicates O atoms, yellow indicates H atoms.

#### b) Encapsulated Two H Atoms

When two H atoms are encapsulated in the water cage cluster as one H molecule, a weak vibration of the H molecule, two H atoms move against one another, appears at a wavenumber of about 4400 /cm, which is originally infrared inactive (Fig. 6).



Fig. 6-a: (H<sub>2</sub>O)<sub>10</sub>+2H

Fig. 6-b: Spectrum of (H<sub>2</sub>O)<sub>10</sub>+2H

Red indicates O atoms, yellow indicates H atoms.

c) Encapsulated Three H Atoms in Triangle Structure

When three H atoms are encapsulated in a triangle structure in the water cage cluster, which is composed by one H molecule and one H atom, an oscillation of the whole H molecule appears at a wavenumber of about 1000 /cm and another oscillation of the H atom against the H molecule appears at a wavenumber of about

1200 /cm, furthermore a weak vibration of H molecule, two H atoms in the H molecule move against one another, appears at a wavenumber of about 4400 /cm, which is originally infrared inactive (Fig. 7).



Fig. 7-a:  $(H_2O)_{10}+3HT$ Fig. 7-b: Spectrum of  $(H_2O)_{10}+3HT$ (Triangle Structure)Red indicates O atoms, yellow indicates H atoms.

### d) Encapsulated Three H Atoms in Slightly Curved Line Structure

When three H atoms are encapsulated in a slightly curved line structure in the water cage cluster, an oscillation of the center H atom appears at a wavenumber of about 2500 /cm, the strength of which is nearly similar to the H atom stretches of the water molecule, and another oscillation of the both ends H atoms appears at a wavenumber of about 3000 /cm, which is originally infrared inactive (Fig. 8).



Fig. 8-a:  $(H_2O)_{10}+3HL$ Fig. 8-b: Spectrum of  $(H_2O)_{10}+3HL$  (Slightly Curved Line Structure)Red indicates O atoms, yellow indicates H atoms.

# 3.2.3 Melting and Vaporization of Ti, Ta, W Metal or Glassy C

It have been reported that Ti, Ta and W metals, the melting point of which are about 1700, 3000 and 3400°C and the boiling point of which are about 3300, 5500 and

5600°C respectively, and glassy carbon (C), the heat-resistant temperature of which is about 3000°C, melt and vaporize at a temperature of about 3000°C when they come into contact with the flame of burning OHMASA-GAS. These metals easily oxidize under a high temperature moisture, the melting point and the boiling point of which become lower, so that they may be considered to melt and vaporize at a temperature about 3000°C or less. However, the heat-resistant temperature of glassy C is about 3000°C in a vacancy and about 500°C in the air. Since vaporization of glassy C occurs at a temperature of about 3000°C, the hydrogen oxidation (burning) must precede the carbon oxidation and further the metal oxidation in the oxygen-deficient but hydrogen-sufficient atmosphere. On the other hand, when OHMASA-GAS is mixed with carbon dioxide or propane or added by H gas, melting and vaporization of Ti, Ta and W metals and glassy C cannot occur. Therefore, the high temperature on these metals or glassy C is possibly caused in the following processes which depend on the encapsulated one to three H atoms in the water cage clusters.

a) Through Hydroxyl (OH) Radical by One Released H atom

When the water cage clusters which encapsulate one H atom or three H atoms of triangle structure are decomposed into water molecules by ambient heat, one H atom released and would combine with one released O atom to form hydroxyl (OH) radical generating a large amount of heat. And finally OH radical would combine with another O atom to form a water molecule (H<sub>2</sub>O) near the surface of metals or glassy C. On the other hand, when H gas is added to OHMASA-GAS, one H atom encapsulated in the water cage cluster changes to one H molecule of two H atoms which remains continuously encapsulated in the water cage cluster, and then this hydroxyl radical reaction would become difficult to happen.

b) Collision of Oscillating H Atoms

The strong oscillation of the center H atom and the weak oscillation of both sides H atoms of encapsulated three H atoms of slightly curved line structure at a wavenumber of about 2500 /cm and 3000 /cm would be excited with infrared absorption of a wavelengths of about 4.0  $\mu$ m and 3.3  $\mu$ m, respectively. These wavelengths are close to the peak wavelength of infrared radiation from the metals or glassy C heated by burning OHMASA-GAS of a temperature range of 600 to 700°C, which corresponds to a wavelength range of 3.3 to 3.0  $\mu$ m. Then, these three H atoms would resonate with the infrared from the heated metals or glassy C initially, and would oscillate to cause strong collision. Even if the water cage clusters which encapsulate three H atoms of slightly curved line structure are decomposed into water molecules by ambient heat before the above-mentioned

oscillations occur, released three H atoms of slightly curved line structure would oscillate at a wavenumber of about 3500 /cm which corresponds to a wavelength of about  $3.0 \,\mu\text{m}$ . This wavelength is nearly coincide to the peak wavelength of infrared radiation from the metals or glassy C heated by burning OHMASA-GAS of a temperature of about  $700^{\circ}$ C. Then, these three H atoms would resonate with the infrared from the heated metals or glassy C initially. However, we have not yet understood whether or how the collision of H atoms encapsulated in the water cage cluster causes any heat reactions.

Three H atoms encapsulated in the water cage clusters seem to be special state, since they have not been observed in the clusters of other materials such as  $C_{10}H_{16}$ -adamantane,  $C_{60}$ -fullerene and  $Si_8O_{12}H_8$ -silsesquioxane or in the metallic lattice such as Pd, Ni and Cu or the lattice C of diamond <sup>10</sup>.

# 3.2.4 Mixing of Carbon Dioxide

When carbon dioxide (CO<sub>2</sub>) is mixed with OHMASA-GAS, CO<sub>2</sub> combines with one H atom encapsulated in the water cage cluster of OHMASA-GAS to form new combustible compound of hydroxyoxomethanide (or carboxyl group or formic acid anion; CHO<sub>2</sub>) which bonds to the water cage cluster, or combines with one H molecule to change to carbon monoxide (CO) and H<sub>2</sub>O or to form new compound of formic acid (CH<sub>2</sub>O<sub>2</sub>) (Fig. 9). It is considered that CO<sub>2</sub> would be reduced by H atom which is produced through electrolyzing by the power of solar cell to obtain the products of photosynthesis such as CO, CHO<sub>2</sub> and CH<sub>2</sub>O<sub>2</sub>.



Fig. 9-a:  $(H2O)_{10}+CH_2O_2$ Fig. 9-b: Spectrum of  $(H_2O)_{10}+CH_2O_2$ Red indicates O atoms, yellow indicates H atoms, gray indicates C atom.

# 3.2.5 Mixing of Hydrocarbon such as LNG and Propane

When methane (CH4: main composition of LNG) or ethylene (C2H4) is mixed

with OHMASA-GAS,  $CH_4$  or  $C_2H_4$  remains bonding to the water cage clusters (Fig. 10-a).

When propane ( $C_3H_8$ ) is mixed with OHMASA-GAS, one H atom encapsulated in the water cage cluster of OHMASA-GAS takes one H atom from propane ( $C_3H_8$ ) to make H molecule which goes out the water cage cluster and propyl radical ( $C_3H_7$ ) which remains bonding to the water cage cluster (Fig. 10-b). On the other hand, two or three H atoms encapsulated in the water cage cluster do not make any transformation of  $C_3H_8$ , which remains bonding to the water cage cluster (Fig. 10-c).



Fig. 10-a:  $H@(H_2O)_{10}+C_2H_4$  Fig. 10-b:  $H_2@(H_2O)_{10}+C_3H_7$  Fig. 10-c:  $H_2@(H_2O)_{10}+C_3H_8$ Red indicates O atoms, yellow indicates H atoms, gray indicates C atom.

# 3.2.6 Fuel Cell

When JARI (Japan Automobile Research Institute) standard cell was supplied with OHMASA-GAS instead of H gas to generate electricity, an early voltage of fuel cell is about 5% higher than that with H gas, but it descend at a temperature higher than 60°C. It is because that one released H atom would work directly for the fuel cell through the water cage cluster which encapsulate one H atom or three H atoms of triangle structure, but when the water cage clusters are decomposed into water molecules by ambient heat of a temperature higher than 60°C, the released H atom would combine with the released O atom before generating electricity.

Fig. 11-b shows the vibrational spectrum of decamer water cage cluster with O atom of  $H_2O_2$ . The oscillation of O atom appears at a wavenumber of about 1200 /cm in Fig. 11-b, and the oscillation of one H atom against one H molecule of encapsulated three H atoms of triangle structure in the water cage cluster also appears at almost the same wavenumber in Fig. 7-b. The water cage clusters are also possibly decomposed by the oscillation of the H or O atom with infrared absorption of a wavenumber of about 1200 /cm from the surrounding object of a temperature of about 60°C or over, although they are possibly decomposed by the bend vibration of water molecule with

infrared absorption of a wavenumber of about 1700 /cm which corresponds to the peak radiation from surrounding object of a temperature of about 250°C.



Fig. 11-a: (H<sub>2</sub>O)<sub>9</sub>+H<sub>2</sub>O<sub>2</sub> Fig. 11-b: Spectrum of (H<sub>2</sub>O)<sub>9</sub>+H<sub>2</sub>O<sub>2</sub> Red indicates O atoms, yellow indicates H atoms.

# 3.2.7 Adding of Hydrogen Gas

When H gas is added to OHMASA-GAS, the liquefaction temperature becomes higher than that of H gas, and then H gas can be stored more safely. Although OHMASA-GAS which is added H gas raises the total heat quantity, it needs to supply oxygen or air for the perfect combustion. Furthermore, Ti, Ta, W metal or glassy C becomes not to melt and vaporize with burning of it, and the voltage of fuel cell with it becomes that with H gas.

It is because that one H atom of added H molecule would combine with one H atom encapsulated in the water cage cluster to form one H molecule which remains encapsulated in the water cage cluster, or combine similarly with one H atom of three H atoms of triangle structure encapsulated in the water cage cluster but one of two H molecules go out the water cage cluster.

# 3.3 Neutral Electrolyte

Neutral electrolyte is obtained through the electrolysis of water added very small amount of salt (NaCl) under vibratory agitation by vibration blades related to the generation of OHMASA-GAS. Cl atom produced from anode seem to combine with HO of water molecule to form hypochlorous acid (HClO) which bonds to the water cage cluster same as hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) of OHMASA-GAS to give the mild and continuous sterilizing properties. Fig. 12 shows the structure of the decamer water cage cluster with Cl atom of HClO and the vibrational spectrum of it.



Fig. 12-a: (H2O)9+HOC1Fig. 12-b: Spectrum of (H2O)9+HOC1Red indicates O atoms, yellow indicats H atoms, light green indicats Cl atom.

# 3.4 Possibility of Generation of Various Elements under Vibratory Agitation Related to Generation of OHMASA-GAS

If some parts of large water tube clusters were strongly adsorbed at the edge faces of Pd plating vibration blades, or if the large water tube clusters were formed with the alkali, alkaline-earth metal or other metal atoms, excess forces would act on a part of the cylindrical bubble of transient cavitation easily to collapse.

Fig. 13 shows the structure of the large water tube cluster bonded with MgO and the vibrational spectrum of infrared absorption of it, and Fig. 14 shows those bonded with HOCl.

When both side of the part of collapsed cylindrical bubble are separated from the edge faces of the vibration blades, they would be divided into small oscillating spherical bubbles with divided nuclei of water clusters.





Fig. 13-b: Spectrum of (H<sub>2</sub>O)<sub>34</sub>+MgO

Red indicates O atoms, yellow indicates H atoms, light green indicates Mg atom.



Fig. 14-a: (H2O)34+HOClFig. 14-b: Spectrum of (H2O)34+HOClRed indicates O atoms, yellow indicates H atoms, green indicates Cl atom.

When MgO is bonded to the large water tube cluster of pentamer cross section like  $(H_2O)_{10}$ +MgO, it stands apart from the water tube cluster, and a strong dipole oscillation of Mg atom appears at wavenumber of about 1400 /cm and the strong breathing mode vibration of the whole large water tube cluster appears at wavenumber of about 0/cm in the infrared absorption spectrum. On the other hand, when hypochlorous acid (HOCl) of Cl compound is bonded to the large water tube cluster of pentamer cross section, it stands near to the water tube cluster, and a dipole oscillation of Cl atom at a wavenumber of about 1200 /cm and breathing mode vibration of the whole water tube cluster at a wavenumber of about 0 /cm in the infrared absorption spectrum are weak. It seems that a part of the cylindrical bubble surrounding the nucleus of the water tube cluster with MgO collapse by the strong dipole oscillation of Mg atom apart from the water tube cluster and the strong breathing mode vibration of the whole water tube cluster.

When the part of large cylindrical bubble collapses, a part of the nucleus of water tube cluster would be compressed by the inflow of surrounding water molecules, and H, O atoms and the alkali, alkaline-earth metal or another metal atom would collide with one another or against the end face of the vibration blade to become plasma state of a high-temperature of several thousand degrees and a high-pressure of several giga-pascals. However, we have not yet understood whether or how the compression of H, O atoms and the alkali, alkaline-earth or another metal atom of water tube cluster or material atom of the vibratory blades causes any condensation reactions, but the following phenomena which seem to be nuclear transmutation would be possible:

a) Vibratory Agitation of Pure Water

Metal ions such as Mg and Zn have been reported to be generated under vibratory

agitation of pure water by Pd plating vibration blades of thin Ti plates.

For example, if one O atom and eight deuterium (D) atoms could condense, or if four O atoms or one material atom of the vibration blades such as Ti atom and four D atoms condense, the generation of Mg or Zn will occur, respectively. If tritium (T) atoms replaced D atoms, these reaction rates will increase. And if one T atom and four or five H atoms condense, the generation of helium (He) will occur.

b) Vibratory Agitation of Chloride or Sulfide Solution

Heavy elements such as P, Ni, Cu, Ag, Pt and Au have been reported to be generated in accordance with the alkali, alkaline-earth metal or other metal ions under vibratory agitation of alkali, alkaline-earth or metal chloride/sulfide solution by vibration blades, and these reactions occur at a high rate by addition of heavy water. (Appendix)

As an example, if one Mg atom, three D atoms and one H atom, or one Mg atom, one D atom and five H atoms could condense, the generation of P will occur. The generation of more heavy elements would need four O atoms or one material atom of the vibration blades such as Ti atoms in addition to one alkali or alkaline-earth metal atom such as Cs or Ca and H or D atoms. If T atoms replaced H or D atoms, these reaction rates will increase.

In this time, calculation was limited to the case of MgCl<sub>2</sub>, calculations will be carried out for the other cases of CaCl<sub>2</sub> and CsCl. In particular, it is significant that CsCl needs be studied because radioactive Cs is possibly reduced. Furthermore, it needs to be studied whether a part of the cylindrical bubbles with the nuclei of water tube clusters are formed under vibration agitation of water or some solutions by small thin metallic, plastic or organic blades or rods mechanically, electromagnetically or biomechanically, and whether a part of them collapses to cause the generation of various elements.

# 4. Summary

OHMASA-GAS and the neutral electrolyte were investigated to be possibly generated by stable cavitation, that is, small oscillating spherical bubbles with the nuclei of small water cage clusters which are made by division of the large cylindrical bubbles and encapsulate H atoms or bond Cl atom produced by the electrolysis of water or NaCl solution, respectively. Furthermore, the generation of various elements under vibratory agitation related to the generation of OHMASA-GAS was investigated to be possibly caused by transient cavitation, that is, the collapse of a part of large cylindrical bubble with the nucleus of large water tube cluster.

The large water tube clusters, the cross section of which was pentamer, were observed to exist. It was supposed that the large water tube clusters were formed at the edge faces of the vibration blades under vibratory agitation, and became the nuclei of large cylindrical bubbles of transient cavitation, which were divided into the small spherical bubbles of stable cavitation with divided nuclei of small water cage clusters. These small water cage clusters, which encapsulate one to three H atoms and possibly compose OHMASA-GAS, were observed to exist. In regard to the high temperature on the metals or glassy C which come into contact to the flame of burning OHMASA-GAS to melt and vaporize instantly, one H atom released from the water cage clusters would combine with one released O atom to form hydroxyl (OH) radical generating a large amount of heat.

In regard to other properties of OHMASA-GAS, carbon dioxide (CO<sub>2</sub>) mixed with OHMASA-GAS was observed to combine with one H atom encapsulated in the water cage cluster to form new combustible compound, and hydrocarbon such as LNG and propane mixed with OHMASA-GAS was observed to remain bonding to the water cage cluster, and so on. Furthermore, in regard to the neutral electrolyte, Cl atom produced from anode was observed to combine with OH of water molecule to form hypochlorous acid (HClO) which bonds to the water cage cluster to give the mild and continuous sterilizing properties.

On the other hand, the strong dipole oscillation of Mg atom and breathing mode vibration of whole large water tube cluster was observed when MgO was bonded to the large water tube cluster, which would cause excess forces acting on a part of the cylindrical bubble of transient cavitation easily to collapse. When the part of large cylindrical bubble collapsed, the part of the nucleus of large water tube cluster of the large cylindrical bubble would be compressed by the inflow of surrounding water molecules. A further study of how the compression of H, O atoms and the alkali, alkaline-earth metal or another metal atom of water tube cluster or material atom of the vibratory blades cause any other reactions should be conducted. This study would be done together with the study of collision of H atoms in the water cage cluster.

Since OHMASA-GAS or the neutral electrolyte and the generation of various elements under vibratory agitation would be closely related one another by the water cluster, the formation, decomposition and compression of water clusters need to be investigated together, and accurate experiments or analyses and computer simulations about OHMASA-GAS or the neutral electrolyte and the generation of various elements under vibratory agitation will be necessary to be carried out.

# Acknowledgements

The author wishes to thank Japan Techno Co., Ltd. for providing many valuable data about OHMASA-GAS and related phenomena.

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	Analyzation by a ICP (Indu	ctively	Coupl	ed Pla	sma) at	omic e	missior	n spect	romete	۶r
	at Saitama Industrial Teo	chnolog	gy Cent	ter						
	Cesium Chloride (CeCl)									
		Cs	Au	Ag	Pt	Cu	Ni	Zn	W	
		[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	
	before processing	6400	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	
	after 6 hour processing	5450	20	11	0.9	0.7	0.9	< 0.1	0.3	
Cupper Chloride (CuCl) solution with heavy water										
		Cu	Au	Ag	Co	Ni	Zn	Ga		
		[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]		
	before processing	3100	< 0.1	< 0.1	< 0.1	0.3	< 0.1	< 0.1		
	after 6 hour processing	2800	2.5	2.2	< 0.1	2.5	< 0.1	< 0.1		

# Appendix

# Nuclear Transmutations in Critical and Supra-critical Electrolysis with Graphite, Pd, W, Re, Pt and Au Cathodes Analyzed by the TNCF Model

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### Abstract

Nuclear transmutations observed in the surface region of cathodes made of C (graphite), Pd and 5d elements (W, Re and Au) used in normal, critical and supra-critical electrolysis with light water in addition to some data observed with heavy water are analyzed using the trapped neutron catalyzed fusion (TNCF) model. The occurrence of the cold fusion phenomenon in 5d transition-metal electrodes is consistently explained in accordance with the cold fusion phenomenon (CFP) observed in 3d and 4d transition-metal hydrides and deuterides, such as NiH<sub>x</sub> and PdD<sub>x</sub>, at the normal electrolysis. The necessary conditions for the realization of the CFP, formation of super-lattice of host nuclei and protons/deuterons in these hydrogen non-occluding materials (at near room temperature) are realized by the higher temperatures of the material induced by a long electrolysis at normal electrolysis (up to three weeks) or by the critical and supra-critical electrolysis with high current densities. Using the data analyzed in this paper in addition to the data obtained in 3d and 4d transition-metals analyzed hitherto, we could contemplate some characteristics of the CFP.

Key words; nuclear transmutation, cold fusion phenomenon, critical electrolysis, glow discharge, arc discharge, TNCF model,

# 1. Introduction

The cold fusion phenomenon (CFP) composed of various events, including nuclear transmutation, emission of neutrons with energies from 2.5 up to 20 MeV, generation of tritium and large excess heat, has been observed for more than 25 years mainly in 3d and 4d transition-metal hydrides and deuterides such as NiH<sub>x</sub> and PdD<sub>x</sub> [Kozima 1998,

2006] besides in XLPE [Kozima 2016c] and biological cultures [Kozima 2016d]. The experimental data sets with large dispersion in the CFP have been successfully analyzed phenomenologically using a model (the trapped neutron catalyzed fusion (TNCF) model) proposed in 1993 at ICCF4 [Kozima 1994] in consistency with knowledge of modern physics. The basic assumptions of the model, especially the existence of the trapped neutrons in materials (CF materials) where the CFP was observed have been explained using the novel knowledge of solid-state physics and nuclear physics [Kozima 2006, 2016c].

It should be remembered that the idea of the neutron energy band introduced in the process of the justification of the trapped neutrons in the CF materials may have close relations with such physical characteristics of the transition-metal hydrides and deuterides as extraordinary large diffusivity of H and D in them. Anyway, the novel knowledge of the interaction among lattice nuclei mediated by occluded H or D disclosed by the CFP will open a new perspective of solid state-nuclear physics in these materials.

In relation to the CFP, there are several modes of electrolysis to charge hydrogen isotopes into metals; the most popular one is the electrolysis of light and heavy waters with alkaline ions in electrolytic liquids at a relatively low current density (let us call this case the *normal electrolysis*) to charge hydrogen isotopes into CF materials (hydrogen occluding metals) such as Ni, Ti and Pd (e.g. [Fleischmann 1989, McKubre 1991]). We have analyzed mainly these data sets of the normal electrolysis hitherto [Kozima 1998, 2006]. There are other types of electrolysis with higher current densities; one is the arc discharge between carbon (graphite) rods in water (e.g. [Hanawa 2000]) successfully analyzed before [Kozima 2012]. This stage of current flow between two electrolysis explained below.

Another mode of electrolysis left aside until now is "the critical electrolysis" [Ohmori 2016] taken up in this paper.

To make our image of electrolysis clear, we cite here the case of electrolysis with palladium cathodes with varying current density. The critical electrolysis occurs in the case of Pd cathodes as follows:

Let us consider the experiment performed by the current control scheme. When the current density *i* is less than a critical value  $i_0$ , the electrolysis is in a steady state. However, when the current density exceeds  $i_0$ , the electrode potential suddenly becomes unstable. We call the electrolysis at the current density  $i_0$  "the critical electrolysis" ([Ohmori 2002, 2004]) and the electrolysis with a current density  $i \ge i_0$  is called "the

supra-critical electrolysis" in this paper for convenience of discussion. In the supra-critical electrolysis, at 5 - 10 minutes after arriving at the critical condition, the glow discharge occurs (called also as "the plasma electrolysis" [Ohmori 1998c]), and the electrode becomes incandescent.

As soon as the electrolysis reaches at the critical condition, the regulated DC current-control mode of power supply changes instantaneously to the potential-control mode owing to the momentary jump of the electric resistance of the electrode/solution interface.

#### **Classification of Electrolysis according to Cathode Phenomena**

In summary, we can classify the electrolysis according to the current density at the cathodes, i.e. according to the cathode temperature which affects the behavior of H and D at the cathode.

The electrolysis is classified in three types according to the current density *i*;

(1) Normal electrolysis; current density  $i \leq \sim 1 \text{ A/ cm}^2$ ,

(2) Critical electrolysis; current density in between 1 and 5 A/ cm<sup>2</sup>; ~ 1 A/ cm<sup>2</sup>  $\leq i \leq \sim 5$  A/ cm<sup>2</sup>,

and

(3) Supra-critical electrolysis; current density i > 5 A/cm<sup>2</sup>.

In the supra-critical electrolysis, there occurs glow or arc discharge.

We can cite several examples of these electrolysis from the history of the CF research as follows;

Normal electrolysis; [Fleischmann 1989]  $i < 512 \text{ mA/cm}^2$ , [McKubre 1991] (typically  $i = 300-600 \text{ mA/cm}^2$ , but up to 6400 mA/cm<sup>2</sup>), [Silver 1993] ( $i = 250 \text{ mA/cm}^2$ ) [Dash 1994] (H<sub>2</sub>O + H<sub>2</sub>SO<sub>4</sub>,  $i = 670 \text{ mA/cm}^2$ ), [Ohmori 1998b] ( $i = 100 \text{ mA/cm}^2$ ).

Critical electrolysis; [Ohmori 1998, 2002, 2004, 2016] *i* ~ 3.13 – 3.75 A/cm<sup>2</sup>,

[Ohmori 2002, 2004 (Pd)]

**Supra-critical electrolysis;** [Ohmori 1999, Mizuno 1998a, Hanawa 2000] glow discharge or plasma electrolysis and arc discharge at higher voltages (up to 240 V)., [Ohmori 1999] (W)  $i \sim 2.5$ A/0.5cm<sup>2</sup> = 5 A/cm<sup>2</sup>. [Hanawa 2000] graphite rods electrodes  $\phi = 6$  mm, I = 30 A,  $i \sim 100$  A/cm<sup>2</sup>, V = 20 V and  $Q_{in} = 600$  W).

# Some characteristics of electrodes in these electrolysis conditions;

(1) In the "Normal Electrolysis," the cathode temperature is in equilibrium with the surrounding electrolyte, and does not become higher than 100 °C (or the boiling point of the electrolytic liquid).

- (2) In the "Critical Electrolysis," the cathode is heated by the proton or deuteron current absorbed by the cathode (Especially, in the case of Pd, the process is exothermic.).
- (3) In the "Supra-critical Electrolysis," the process becomes glow or arc discharge in the liquid, and the cathode is heated more than that in the critical electrolysis up to the boiling point of the electrolytic liquid. In the arc, the anode is also heated very high up to about 3000 K [Hanawa 2000].

In this paper, we take up mainly experimental data sets obtained with the critical and supra-critical electrolysis by Ohmori et al. and also related data sets to them to discuss the cold fusion phenomenon from the unified point of view using the TNCF model based on the solid-state nuclear physics of neutrons in CF materials.

In Section 2, we explain experimental data sets obtained in experimental systems with cathodes of graphite, Pd and 5d transition-metals. In Section 3, we summarize the essence of the TNCF model. In Section 4, we give analysis of the experimental data sets explained in Section 2 by the TNCF model.

In Section 5, we explain the meaning of the phenomenological model based on the experimental data.

In Appendices, we give a summary of physical properties of hydrogen isotopes in metals in Appendix A and the super p-p interaction between protons in interstices in solid state physics, a counterpart of the super-nuclear interaction, in Appendix B.

# 2. Experimental Data Sets on the Nuclear Transmutation in Cathodes at the Normal, Critical and Supra-critical Electrolysis with Graphite, Pd, W, Re and Au Cathodes

The cold fusion phenomenon (CFP) has been investigated mainly in transition-metal hydrides and deuterides since the discovery of this phenomenon in a Pd/D<sub>2</sub>O+LiOD/Pt system by Fleischmann et al. in 1989 [Fleischmann 1989]. There have been investigated the CFP not only in deuterium systems (e.g. PdD<sub>x</sub> and TiD<sub>x</sub>) but also in protium systems such as NiH<sub>x</sub> and PdH<sub>x</sub> in several years after the discovery (cf. e.g. [Kozima 1998, 2006]). Thus, the CF materials where the CFP has been observed increased from the transition-metal deuterides to the transition-metal hydrides in a few years after the announcement of the CFP by Fleischmann et al.

One of the astonishing discoveries of the CFP was that there are observed nuclear transmutations, production of heavy elements, especially a large amount of iron in an unexpected CF material, graphite [Sundaresan 1994, Singh 1994, Hanawa 2000].

Another remarkable data on nuclear transmutation and excess heat have been observed in 5d transition-metal hydrides by Ohmori et al. after 1996 which are the main subject of the analysis in this paper. The experimental data of these works are tabulated in Table 2.1, and discussed further in the following subsections.

Table 2.1 Nuclear transmutations observed in the surface region of cathodes at the critical and supra-critical electrolysis with Graphite, W, Re, Pt and Au cathodes

Cathode/Electrolytic	Electrolysis;	Current density	Transmuted
Liquid/Anode	Normal (N),	at cathode,	nuclei
	Critical (C),	(duration of	(References)
	Supra-critical	experiment)	
	(SC)		
Graphite/H <sub>2</sub> O/Graphite	SC(Arc discharge	$100 \text{ A/cm}^2$ ,	Fe + Cr, Mn, Co,
	in H <sub>2</sub> O)		Ni, Cu and Zn (1)
Pd/D <sub>2</sub> O +K <sub>2</sub> CO <sub>3</sub> /Pt	Ν	$0.2 - 0.8 \text{ A/cm}^2$ ,	Cr, Fe, Cu, Pb (5)
$Pd/H_2O + Na_2SO_4/Pt$	С	$3.1 - 3.8 \text{ A/cm}^2$	Pd, Au, Hg, Pb(6)
W/H2O+Na2SO4+ Na2CO3+K2CO3+/Pt	SC	5 A/cm <sup>2</sup> ,	Cr, Fe, Ni, Re(2,3)
Re/H2O+Na2SO4+	SC (glow	$3 \text{ A/cm}^2$ ?	Fe, Cu, Zn (3)
K <sub>2</sub> SO <sub>4</sub> +K <sub>2</sub> CO <sub>3</sub> +/Pt	discharge in H <sub>2</sub> O)		
Au/H2O+Na2SO4+	Ν	$0.2 \sim 1.5 \text{ A/cm}^2$ ,	F, Si, Fe, Pt, Hg
K2SO4+K2CO3/Pt		(7 – 30 days)	(4)

*References:* (1) [Hanawa 2000], (2) [Ohmori 1999, 2000b], (3) [Ohmori 2000a], (4) [Ohmori 1996a, 1996b, 1996c, 1997a, 1997b, 1998a], (5) [Mizuno 1998, Ohmori 1998b], (6) [Ohmori 1996a, 1998b, 2002, 2004]. *Detailed explanations are given below in this section.* 

The working conditions for "critical" and "supra-critical" electrolysis for the CFP in these 5d-transition metals are explained in the beginning of Sec. 2.1.

# **Classification of the nuclear transmutations**

The nuclear transmutations (NTs) observed in the CFP could be classified by the mechanism to generate new elements according to the TNCF model. The NTs are divided into two types, NT of the first kind (NT I) and that of the second kind (NT II). NT of the first kind (NT I) is characterized by the reaction to generate new nuclides where participates only a single neutron, and we have been able to explain experimental data sets of this type quantitatively as shown in our papers and books [Kozima 1998, 2006, 2014a]. On the other hand, the NT of the second kind (NT II) needs neutron drops,  ${}^{A}{}_{Z}\Delta$  composed of *Z* protons and (*A* – *Z*) neutrons (cf. Sec. 3.1), to explain the data showing large changes of the proton *Z* and nucleon *A* numbers from pre-existed nuclides.

The existence of trapped neutrons in CF materials was an a priori assumption in the early stage of the TNCF model, but has been later explained by the formation of the CF-matter due to the neutron energy band realized by the super-nuclear interaction between neutrons in lattice nuclei mediated by interstitial protons or deuterons. [Kozima 2006] The CF-matter has now to be classified into type-I and type-II corresponding to the classification of the nuclear transmutation into NT I and NT II as explained in Sections 3 and 4.

# 2.1 Explanations of Data Sets of the Nuclear Transmutation by the Normal, Critical and Supra-critical Electrolysis on Graphite, Palladium and 5d Metal Electrodes

It is interesting to notice that such materials as graphite and 5d metals with d-electrons more than 4 (W, Re, - - -) are different from hydrogen-occluding 3d and 4d metals in their interaction with hydrogen isotopes. Usually at near room temperature, the former metals are inactive to hydrogen isotopes with large positive heat of solutions as shown in Fig. A1 (Appendix A). Therefore, they usually have not been used as CF materials where is observed the cold fusion phenomenon (CFP).

The situation changes very much, if these materials are heated to higher temperatures where hydrogen isotopes tend to be absorbed and occluded. These materials at higher temperatures have similar properties to the 3d- and 4d-transition metal hydrides. The experiments performed mainly by Ohmori et al. have shown that 5d-transition metals exhibit an interesting characteristic of CF materials common to 3d- and 4d-transition metals in terms of the CFP. This characteristic is valuable not only from scientific but also from technological point of view as shown in this paper.

In the experiments with 5d transition metal electrodes, the electric power was supplied by either of the regulated DC current/voltage supplies with 160 V or 240 V

maximum output potential in the case of W/H<sub>2</sub>O/Pt system. When the electrolysis is performed at current-control mode with a definite current of  $i_0$ , the electrolysis is kept normal at voltages lower than a value  $v_c$ . When the voltage exceeds a critical voltage  $v_c$ (critical electrolysis), it becomes uncontrollable, fluctuating up and down. This stage is called the "supra-critical electrolysis." Under this situation the electrolytic solution is heated vigorously, and, as a result, a glow discharge starts (plasma electrolysis). In the case of carbon electrode, arc discharge starts.

# 2.1.1 Hydrogen-graphite system; Graphite/H<sub>2</sub>O/Graphite

In the experiments by Sundaresan et al. [Sundaresan 1994] and by Hanawa [Hanawa 2000], a lot of iron is produced in addition to other elements such as Ca, Si, Cr, Mn, Co, Ni, Cu and Zn when carbon (graphite) is used as electrodes for arcing in water and in air. Furthermore, it is shown that the isotopic ratio of the generated iron is the same to the natural one. These data sets was analyzed by the TNCF model, and given a consistent explanation for the transmuted nuclei as a result of nuclear interactions between trapped neutrons and cathode materials [Kozima 2012].

# 2.1.2 Palladium cathode systems; Pd/D<sub>2</sub>O/Pt and Pd/H<sub>2</sub>O/Pt

Using Pd cathodes with heavy and light waters, Ohmori and his collaborators discovered nuclear transmutations of elements at surface regions of cathodes with normal and supra-critical electrolysis.

# 2.1.2a Pd/D<sub>2</sub>O/Pt

Experiments were performed in the system of Pd/D<sub>2</sub>O+Li<sub>2</sub>CO<sub>3</sub>/Pt with current densities of 0.2 - 0.8 A/cm<sup>2</sup> [Mizuno 1998]. In the surface layer of depth about 10,000 Å, there appeared many new elements especially Ti, Cr, Fe, Cu, Sn and Pb with larger amounts compared with others. Furthermore, the isotopic abundances of chromium  ${}^{A}_{24}$ Cr differed from the natural abundances as shown in Table 2.2;

Table 2.2 Isotopic changes of chromium isotopes  ${}^{A}_{24}$ Cr found in the surface layer of the Pd cathode from natural ones [Mizuno 1998].

A	50	52	53	54
Natural abundance (%)	4.3	84	9.5	2.4
Measured abundance (%)	14	51	2.4	11

This experimental data is discussed in Sec. 4.2.2.

# 2.1.2b Pd/H<sub>2</sub>O/Pt

Ohmori et al. have performed extensive measurements of the CFP in  $Pd/H_2O+Na_2SO_4/Pt$  systems at the supra-critical electrolysis condition for a long period of time [Ohmori 1996a, 1998b, 2002, 2004]. Details of the experiments are given below, and their analysis is given in Sec. 4.2.2.

# 2.1.2b-1 Electrolysis

#### **Electrolysis condition**

When the experiment was performed at a current density of 2 A/cm<sup>2</sup>, the solution temperature reached ca 60-70°C.

When the current density was set at 3 A/cm<sup>2</sup>, the temperature was further raised to ca. 85 °C. Under this condition the electrode potential began to fluctuate up and down like a wave (this stage of electrolysis is termed "supra-critical electrolysis" [Ohmori 2002, 2004]), and after 5-10 minutes there occurred a glow discharge, and the electrode became in an incandescent condition. This state of electrolysis is called "plasma electrolysis." [Ohmori 1998c]

When the electrolysis became plasma electrolysis condition, the electrolytic solution started to boil vigorously. As soon as the electrode reached plasma electrolysis condition, the regulated DC current-potential supply mode changed instantaneously from the *current-control mode* to the *potential-control mode* owing to the momentary jump of the electric resistance of the electrode/solution interface.

### 2.1.2b-2 Result

# (a) Isotope distribution

Isotopic distribution of Pd on the surface of the eight kinds of electrodes after the supra-critical electrolysis is shown in a figure [Ohmori 2016 (Fig.4)]. Without exception the isotopic abundance of the palladium electrodes deviates markedly from its natural one for all of the electrodes. In this case, there are following characteristics in common: (i) Content of <sup>108</sup>Pd exceeds significantly its natural isotopic abundance for all of the electrodes: (ii) Content of <sup>110</sup>Pd exceeds its natural isotopic abundance in most cases, (iii) In contrast, content of other lighter palladium isotopes are mostly below their natural isotopic abundance. Consequently, it appears that the content of heavier palladium isotopes tends to increase, whereas, the content of lighter ones tends to decrease.

#### (b) Foreign elements

Foreign elements with larger atomic numbers by 2 - 4, Cd and Sn, were detected

after the supra-electrolysis of Pd electrode [Ohmori 2016 (Fig.7)]. The isotopic distribution of Cd is very different from its natural isotopic abundance. In this case, <sup>114</sup>Cd (main isotopic component in natural Cd) was not contained in Cd detected after the electrolysis [Ohmori 2016 (Fig. 12)]. Unfortunately, isotopic distribution of Sn could not be analyzed because of overlapping of its SIMS signals with those of other materials.

Foreign elements Pt and Au with larger atomic numbers, other than Cd and Sn, were also detected. These elements have different isotopic distributions from their natural ones [Ohmori 2016 (Fig.9)]. In this electrolysis system Pt net was used as a counter electrode. So the Pt was considered to be produced as a result of the electrodeposition of Pt dissolved from the counter electrode. The production of Au may result from the Pt similarly to the production of Cd or Sn from Pd.

# 2.1.3 Tungsten cathode system; W/H<sub>2</sub>O+Na<sub>2</sub>SO<sub>4</sub>+Na<sub>2</sub>CO<sub>3</sub>+K<sub>2</sub>CO<sub>3</sub>/Pt

[Ohmori 1998c, 1999, 2000a, 2000b]

Information of the electrode surface after the plasma electrolysis was obtained only when W and Re electrodes with high melting point were used.

A typical example of the W electrode after the plasma electrolysis is shown in the figure [Ohmori 2016 (Fig.22)]. Foreign elements Fe, Cr, Re, Pb were detected in this case. However, the departure of isotopic abundance of Fe and Cr in this case was not so remarkable. Notable deviation of isotopic abundance is observed for Re and Pb [Ohmori 2016 (Fig. 22 and Table 7)].

# 2.1.4 Rhenium cathode system; Re/H<sub>2</sub>O+Na<sub>2</sub>SO<sub>4</sub>+K<sub>2</sub>SO<sub>4</sub>+K<sub>2</sub>CO<sub>3</sub>/Pt

[Ohmori 2000a]

The rhenium electrode was a square foil (0.8 cm<sup>2</sup> apparent area, 0.02 cm thick). The counter electrode was a platinum net (1 cm  $\times$  10 cm, 80 mesh). The surface of the working electrode was scraped elaborately with a cleaned glass fragment to make a crystal lattice strain. The electrolysis was performed in the light water solutions of K<sub>2</sub>CO<sub>3</sub>, KNO<sub>3</sub>, Na<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, NaNO<sub>3</sub>, NaClO<sub>4</sub>, Rb<sub>2</sub>CO<sub>3</sub>, Ba(ClO<sub>4</sub>)<sub>2</sub>, and Ba(NO<sub>3</sub>)<sub>2</sub>.

The condition of the glow discharge can be accomplished as follows; first, pre-electrolysis was conducted galvanostatically at a current density of 2  $A/cm^2$  for 1 - 1.5 hours by which hydrogen was loaded in the electrode. Then the current density was increased step-wisely for ca. 30 minutes. When the current density exceeds ca. 3  $A/cm^2$ , the applied potential turned labile, and,

after the passage of several minutes, the electrode incandesced, and emitted red-violet glow.

When the experimental condition reaches glow discharge, the electric power source operating in the galvanostatic mode switches automatically to the potentiostatic mode owing to the rapid increase in the electric resistance at the electrode/solution interface. As a result, the input potential jumped up to 160 V, and the current density dropped down to 0.8 - 1 A/cm<sup>2</sup>, and thereafter gradually decreased, as a whole, down to ca. 0.5 A/cm<sup>2</sup>.

After the supra-critical electrolysis with a glow discharge, Fe, Cu and trace amount of Zn or only Zn was detected in the Re electrodes at surface layers of widths about 100 Å. The data of transmutation observed in the Re electrodes are discussed in Sec. 4.

# 2.1.5 Gold cathode system; Au/H<sub>2</sub>O+Na<sub>2</sub>SO<sub>4</sub>+K<sub>2</sub>SO<sub>4</sub>+K<sub>2</sub>CO<sub>3</sub>/Pt [Ohmori 1997b]

Experiments of the normal electrolysis on the hydrogen non-occluding gold cathodes have been performed on Au cathodes with approximate dimensions of 1.0, 2.5 and 5.0 cm<sup>2</sup> × 0.1 mm in electrolytic liquids (H<sub>2</sub>O + Na<sub>2</sub>SO<sub>4</sub> (K<sub>2</sub>SO<sub>4</sub>, K<sub>2</sub>CO<sub>3</sub>)) for fairly long time-duration of 7 – 30 days with electrolytic current densities of 0.1 – 1.0 A/cm<sup>2</sup>. The experiments with Au cathode for fairly long times (up to 30 days) and higher current densities up to 1 A/cm<sup>2</sup> in normal electrolysis scheme have revealed several interesting features of the CFP in electrolytic systems as discussed in Sec. 4.

Details of experimental data in this system are introduced in Sec. 2.3

# 2.2 Critical Electrolysis with Palladium Cathode-Light Water system;

Pd/D2O+K2CO3/Pt and Pd/H2O+ Na2SO4/Pt

Palladium, a hydrogen occluding metal, is the first metal used for the CF material due to its large content of hydrogen at near room temperature (e.g. [Fleischmann 1989, McKubre 1993]). The characteristics of hydrogen isotope-palladium system are peculiar at its inverse-mass dependence of diffusivity and other properties of protium and deuterium. Below about 200 °C, deuteron is more diffusive than proton in Pd. As a result, the Pd-D system is appropriate to measure the cold fusion phenomenon (CFP) than the Pd-H system, even if protium is occluded in palladium (cf. Fig. A3 in Appendix). Above about 200 °C, the diffusivity of proton and deuteron becomes normal, i.e. the former has larger diffusivity than the latter. This is the reason that the critical electrolysis for the Pd + H system is appropriate for the CF research as successfully

performed by Ohmori et al. [Ohmori 1998b, 2002, 2004].

#### **Isotope distribution**

Isotopic distribution of Pd on the surface of the eight kinds of electrodes after the supra-electrolysis is shown in the figure [Ohmori 2016 (Fig.4)]. Without exception the isotopic abundance of the palladium electrodes deviates markedly from its natural one for all of the electrodes. In this, there are some common characteristics in the following: (i) Content of <sup>108</sup>Pd exceeds significantly its natural isotopic abundance for all of the electrodes: (ii) Content of <sup>110</sup>Pd exceeds its natural isotopic abundance in most cases, (iii) In contrast, content of other lighter palladium isotopes are mostly below their natural isotopic abundance. Consequently, it appears that the content of heavier palladium isotopes tends to increase, whereas, the content of lighter ones tends to decrease.

#### **Foreign elements**

Foreign elements with atomic numbers 2-4 larger, Cd and Sn, were detected after the supra-electrolysis of Pd electrode [Ohmori 2016 (Fig.7)]. The isotopic distribution of Cd is very different from its natural isotopic abundance. In this case, <sup>114</sup>Cd (main isotopic component in natural Cd) was not contained in Cd detected after the electrolysis [Ohmori 2016(Fig.12)]. Isotopic distribution of Sn could not be analyzed because of overlapping of its SIMS signals with those of other materials)

Foreign elements with larger atomic numbers other than Cd and Sn, Pt and Au, were also detected. These elements have different isotopic distributions from their natural ones [Ohmori 2016 (Fig.9)]. In this electrolysis system, Pt net was used as a counter electrode. So the Pt was considered to be produced as a result of the electrodeposition of Pt dissolved from the counter electrode. The production of Au may result from the Pt like the production of Cd or Sn from Pd.

# 2.3 Normal Electrolysis with Au Cathode-Light Water System;

 $Au/H_2O + Na_2SO_4 + Na_2CO_3 + K_2CO_3/Pt$ 

# 2.3 (a) Experiment

The gold electrodes used were of Au sheets (1.0, 2.5 and 5.0 cm<sup>2</sup> apparent surface area). Electrolysis was carried out in 0.5M Na<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, and K<sub>2</sub>CO<sub>3</sub> solutions at current densities ranging from 0.1 to 0.5 A/cm<sup>2</sup>.

# 2.3 (b) Precipitates

In the normal electrolysis (0.1 A/cm<sup>2</sup>) no electrolysis deposits were observed. However,

fine black porous precipitates began to appear at the bottom of the electrolytic cell when electrolyzed at current densities of > 0.2 A/cm<sup>2</sup>. In most cases the precipitates began to appear within 2-3 days after starting the electrolysis, and then gradually increased. The amount of the precipitates was in the range from 0.1 to 1 mg, and seemed to be different, depending on the slight difference in the manner of the surface pretreatment of the electrode. The scanning microscope (SEM) image of the precipitate is shown in [Ohmori 1997b (Fig.14)]. To all appearance, the precipitates look like fine black powder, but, in fact, they have a number of very fine porosities ranging from a few nm to  $\mu$  m order as shown in (b, c).

The EPMA images are shown in [Ohmori 1997b (Fig.21)]. As seen from this figure the main component of the precipitates is gold. However other kinds of elements, such as Pt, Hg, Os, Hf, Fe and Si, are also included. Therefore, we can consider that these precipitates are those erupted from the volcano-like structure formed on the gold electrode as will be mentioned below.

# 2.3 (c) Surface topography of the electrode

There have been observed many examples of topographical changes of cathode surfaces in experiments with normal and in supra-critical electrolysis [Silver 1993, Kozima 1998 (Appendix C7)]. The surface topography of electrodes observed in the experiments performed by Ohmori et al. [Ohmori 2016 (Secs, 3-2, 3-3, 4-2)] is discussed briefly in Sec. 4.3.2.

# **2.3 (d) Isotopic distribution of product elements**

The isotopic distribution of the major product elements reveals significant deviation from their natural isotopic abundance. The details will be described below.

# 2.3 (d-1) Elements on/in the electrodes at low current densities

Under the normal electrolysis at current densities > 0.2A/cm<sup>2</sup>, no detectable precipitates appeared throughout the electrolysis. In this case iron atoms were detected exclusively at the part of the electrode close to the surface of the electrode. Figure 9 and 10 [Ohmori 1997b (Figs. 9 and 10)] show the AES spectra of the gold electrode after the electrolysis after 7 days of the electrolysis in Na<sub>2</sub>SO<sub>4</sub> and K<sub>2</sub>CO<sub>3</sub> at a current density of 0.1A/cm<sup>2</sup>. On the spectra of the uppermost layer iron and oxygen signals are observed other than gold signals. On carrying out A<sup>+</sup> ion bombardment, the iron and oxygen signals decline, and disappear after several minutes. The distribution profile of iron atoms calculated from the results in Fig.9 is shown in Fig.12 [Ohmori 1997b (Fig. 12)].

# 2.3 (d-2) Elements on/in the electrodes at high current densities

A considerable amount of Hg is observed on and in the electrode after the normal electrolysis at higher current densities ~ 0.5 A/cm<sup>2</sup>. Figure 17 [Ohmori 1997b (Fig.17)] shows EPMA mapping images of Au, Hg, Fe and Si on the gold electrode after the electrolysis in Na<sub>2</sub>CO<sub>3</sub> at a current density of 0.5 A/cm<sup>2</sup>. Judging from this figure the amount of mercury on the electrode surface is comparable with gold of the electrode material.

The isotopic abundance of the mercury is also different from its natural isotopic abundance [Ohmori 1997b (Fig. 28)]. The content of lighter mercury isotopes, <sup>198</sup>Hg, <sup>200</sup>Hg increase, while, the content of heavier ones, <sup>201</sup>Hg, <sup>202</sup>Hg, decrease. Especially the maximum component, <sup>202</sup>Hg, in the natural world decreases down to several percent.

# **2.3 (d-3)** Elements on and in the precipitate produced in the electrolysis at high current densities

The elements with anomalous isotopic distribution were also detected in the precipitant. As a typical example, we show the isotopic distribution of mercury against the scan number of SIMS [Ohmori 1997b (Fig.34)]. The content of <sup>200</sup>Hg is much larger than its natural isotopic abundance also in this case although the isotopic distribution of mercury of the precipitant shows rather complicated as compared with that of Au electrode itself [Ohmori 1997b (Fig. 28)]. Perhaps such an abnormality is attributed to the structural feature of the precipitant. [Ohmori 1996a, 1996b, 1996c, 1997a, 1997b, 1998a]

# 2.4 Summary of the experimental data sets on the normal, critical and supra-critical electrolysis with 5d transition-metal cathodes

There are more interesting features of the CFP than the nuclear transmutations analyzed in this paper. We explain some of them in addition to the summary of the nuclear transmutations explained above.

# **2.4.1** Nuclear Transmutations in the 5d transition-metal cathodes

It was necessary to perform electrolysis in critical and supra-critical conditions when the electrodes were 5d transition metals. Or it was necessary to use very long times of electrolysis. These conditions are discussed in Section 4.

# 2.4.2 Surface topography

There have been observed many evidences of nuclear reactions at surface regions of electrodes of CF materials. One of the most impressive evidence of nuclear reactions producing huge amount of excess energy has been the topography change of cathode surface showing such high temperatures above the melting points  $T_{\rm m}$  of electrode metals as Ni ( $T_{\rm m} = 1455$  °C) and Pd ( $T_{\rm m} = 1554.9$  °C) in 3d and 4d transition metals (e.g. [Kozima 2006 (Fig. 2.3) and Appendix C7]).

Also, there have been observed topography changes of the cathode surface of 5d transition metals in the experiments where were observed nuclear transmutations; the melting points of these 5d transition metals are very high, i.e.  $T_{\rm m} = 3387 \,^{\circ}\text{C}$  (W),  $T_{\rm m} = 3180 \,^{\circ}\text{C}$  (Re),  $T_{\rm m} = 1772 \,^{\circ}\text{C}$  (Pt) and  $T_{\rm m} = 1064 \,^{\circ}\text{C}$  (Au). Some examples of them are briefly discussed below.

#### Gold cathode

The surface of the electrode after the electrolysis at a current density >  $0.2 \text{ A/cm}^2$  exhibits very anomalous structure. Figure 2.1 [Ohmori 1997b (Fig.40)] shows typical SEM images of the electrode surface before and after the electrolysis for 30 days at a current density of  $0.5 \text{ A/cm}^2$ .

Here (a) shows the electrode surface before the electrolysis. To our surprise a number of craters with various sizes are developed (Figures d and e). Each crater has a deep hole reminding us of a volcano. The size of the largest one reaches ca. 20 µm diameter and 30 µm height (b, c). From the appearance of these craters one can imagine that some explosion took place during the electrolysis, and as a result of this, such excellent craters were completed (f). The outside wall of the craters consists of fine porous substance which reminds us of the structure of the precipitates mentioned above. The inside of the craters at least consists of three kinds of layers as judged from the inspection of the picture, That is, while at the bottom part the wall is made of relatively large irregularly formed crystallites of several µm order, at the intermediate part the wall is made of more fine crystallites of several 100 nm order (g). The magnified picture of this layer shows that these crystallites have hexagonal а shape assignable to Au (111) (h). At the outside the wall consists of very fine crystallite of less than 30 nm. These crystallites also look hexagonal, although the outline is not necessarily clear. Hence these crystallites may be regarded as to be produced by the reconstruction of gold electrode material owing to the intense heat evolved.



Fig. 2.1 Typical SEM images of the Au electrode surface before (a) and after (b) – (h) the electrolysis in  $Na_2CO_3$  solution of light water for 30 days at a current density of 0.5 A/cm<sup>2</sup> [Ohmori 1997b (Fig. 40)].

Similar features have been observed in W and Re cathodes as briefly explained below.

#### Tungsten cathode

Surface topography of W electrode after the plasma electrolysis shows a crater-like structure [Ohmori 2016 (Fig.25)]. The EDX spectra were observed at two different spots on the crater-like structure; one is a spot A at the center part, and the other is a spot B near the edge. On the spectrum from spot A, strong signals of Fe and Cr and moderate signal of Ni are observed, from which the content of Fe, Cr and Ni are calculated as 67.4, 16.9 and 7.9 at.%, respectively. This suggests that the nuclear reaction takes place predominantly at the center of the crater.

#### Rhenium cathode

Surface topography of Re electrode after the plasma electrolysis was investigated [Ohmori 2016 (Fig.20)]. A number of rhombus pits of various sizes are aligned with regularity. In addition, fine sharp grooves run straightly in all directions thrusting through the centers and corners of individual pits. The dimension of these pits is mostly in the range from 10 to 20 nm, respectively. On the surface of the electrode K, Zn, Fe and Cu were observed. [Ohmori 2000]

# **3.** Trapped Neutron Catalyzed Fusion (TNCF) Model for the Cold Fusion Phenomenon (CFP)

The effectiveness of the TNCF model have already been shown by analyses of the CFP, especially the nuclear transmutations in such protium and deuterium systems as NiH and PdD using the idea of the neutron drop  ${}^{A}Z\Delta$  [Kozima 2006 (Sec. 2.4.2), 2014a (Sec. 4)]. In this section, we summarize the essence of the TNCF model which will be used in Section 4 to analyze the experimental data obtained in 5d transition metal hydrides and deuterides.

# 3.1 Phenomenological Model with an Adjustable Parameter $n_n$ , the Density of the Trapped Neutrons in a CF Material

Based on the curious experimental data showing occurrence of nuclear reactions in CF materials, we have elaborated a phenomenological model, the trapped neutron catalyzed fusion (TNCF) model, assuming several premises; the most important one of them is the existence of the trapped neutrons with a density  $n_n$  adjustable to an experimental data [Kozima 1994].
The fundamental nuclear reaction of the TNCF model is a reaction between a trapped neutron (*n*) and a foreign or a displaced nucleus X ( ${}^{A}_{Z}$ X) to generate a new nuclide Y ( ${}^{A+1}_{Z}$ X) followed by some processes to generate a new nuclide Y or nuclides Y and Y', or others;

$$n + {}^{A}_{Z}X \rightarrow {}^{A+1}_{Z}X \rightarrow {}^{A+1}_{Z+1}Y + e^{-} + \underline{\nu}_{e}, \text{ (beta decay of } {}^{A+1}_{Z}X)$$

$$\rightarrow {}^{A-3}_{Z-2}Y + {}^{4}_{2}\text{He}, \text{ (alpha decay of } {}^{A+1}_{Z}X)$$

$$(3.1)$$

$$\rightarrow {}^{A-A'+1}Z-Z'+1}Y + {}^{A'}Z'Y', \text{ (fission of } {}^{A+1}ZX \text{ into } Y \text{ and } Y') \quad (3.3)$$

where  $\underline{v}_e$  is the anti-particle of the electron neutrino  $v_e$ .

This type of the nuclear transmutation (NT) is called the nuclear transmutation of the 1<sup>st</sup> kind (NT of type I) in this paper.

The TNCF model has been applied to a few data sets where were observed several observables (e.g. a and b) simultaneously; the quantitative relations between the values of the observables a and b have been explained by the model in a factor of 3 as expressed as follows;

$$(N_{\rm a}/N_{\rm b})_{\rm th} \cong m (N_{\rm a}/N_{\rm b})_{\rm ex}, (m = 3 - 5).$$
 (3.4)

where  $N_a$  and  $N_b$  are the values of observables a and b.

The resulting data of the analyses were tabulated in our books [Kozima 1998 (Tables 11.2 and 11.3), 2006 (Tables 2.2 and 2.3)]

This success of the model has shown that there is something true in the premises of the model reflecting reality in the cold fusion phenomenon (CFP).

# 3.1.1 Extension to the Neutron Drop Model

The success of the model described above encouraged extending the model to explain wonderful data sets of nuclear transmutations with large changes of proton Z and nucleon A numbers. Suggested by the work by Negele et al. [Negele 1973] on the neutron star matter, we proposed concepts of the CF-matter and the neutron drop  ${}^{A}Z\Delta$ composed of Z protons and (A - Z) neutrons in the CF material [Kozima 1998 (Sec. 12.8), 2006 (Sec. 3.7), 2008b]. The CF-matter [Kozima 2006] is a concept to express a state composed of high density neutrons in the CF material similar to the neutron star matter in the case of the neutron star [Negele 1973].

When there are neutron drops in CF-matter formed around surface/boundary regions by the mechanism discussed above, we can use the neutron drop  ${}^{A}_{Z}\Delta$  and a small neutron-proton cluster  ${}^{A}_{Z}\delta$  in the nuclear reactions as a simultaneous feeder of several nucleons to nuclides. The nuclear transmutations (NT's) induced by this mechanism is called the nuclear transmutation of the 2<sup>nd</sup> kind (NT of type II) in this paper, and include following reactions;

$${}^{A}_{Z}\Delta + {}^{A'}_{Z'}X \longrightarrow {}^{A-a}_{Z-z}\Delta + {}^{A'+a}_{Z'+z}X',$$
  
$$\longrightarrow {}^{A-a}_{Z-z}\Delta + {}^{A'+a-a'}_{Z'+z-z'}X'' + {}^{a'}_{z'}X''' \qquad (3.5)$$

$$A'_{Z'}\Delta + A^{+1}_{Z'}X^* \rightarrow A'_{Z'}\Delta^* + A^{+1}_{Z'}X \rightarrow A'_{Z'}\Delta + A^{+1}_{Z'}X + \phi's,$$

$$(3.5)$$

$$(3.6)$$

$$A'_{Z'}\delta + A_{Z}X \rightarrow A^{+}A'_{Z+Z'}X^* \rightarrow A^{+}A'_{Z+Z'}X + \phi's,$$

$$(3.7)$$

$$^{A}_{Z}\Delta \rightarrow ^{A}_{Z}X.$$
 (3.8)

In the reactions (3.6) and (3.7), we have used a symbol  $\phi$ 's to express dissipation of the excess energies in the intermediate neutron drop  ${}^{A'}_{Z'}\Delta^*$  and nuclide  ${}^{A+A'}_{Z+Z'}X^*$  into surrounding CF-matter (and then to the phonons in the lattice of CF materials) instead of photons in the case of nuclear reactions in free space.

The last reaction (3.8) is the transmutation by transformation (NT<sub>T</sub>) as classified according to the characteristics of nuclear reactions after the absorption of a neutron or of a neutron drop in the former papers (e.g. [Kozima 2014a]). In this case, a neutron drop  ${}^{A}z\Delta$  in a CF-matter transforms into a nuclide  ${}^{A}zX$  when it suffered disturbance by a foreign nucleus or displaced nucleus at a surface region or in a volume of the superlattice where the CF-matter exists.

In these nuclear reactions, a neutron drop can participate in them resulting in the nuclear transmutations such as  ${}^{A}_{79}\text{Pd} \rightarrow {}^{A'}_{82}\text{Pb}$  [Ohmori 1998a].

 $^{197}_{79}$ Pd +  $^{A'}_{3\Delta} \rightarrow ^{A}_{82}$ Pb (A = 206, 207, 208, A' = 9, 10, 11).

Table 3.1 Nuclear transmutations in the cold fusion phenomenon (CFP) arranged according to the nuclear processes from the initial to the final stages; *a*, initial, *b*, intermediate, and *c*, final stages. Nuclear transmutations by decay (NT<sub>D</sub>), by absorption (NT<sub>A</sub>), by transformation (NT<sub>T</sub>), and fission (NT<sub>F</sub>), which have been used in the former books and papers (e.g. [Kozima 1998, 2006, 2014a]) are used to describe the final reactions to assist understanding of the complex nature of the NTs in the CFP.  $\underline{\nu}_e$  is the anti-particle of the electron neutrino.

Initial reactants in the	Intermediate	Final reaction	Classification by the
stage <i>a</i> of NT and	nuclide	products in the stage c	final reaction
(its classification)	in the stage b	(common to	in the stage <i>c</i>
		NT I and NT II are	
		omitted)	
$n + A_Z X$ (NT I)	$^{A+1}ZX^*$	$^{A+1}_{Z}$ X' + e <sup>-</sup> + $\underline{v}_{e}$	$NT_D(\beta)$
		$^{A-3}$ <sub>Z-2</sub> X' + $^{4}_{2}$ He	$NT_D(\alpha)$
		$^{A+1}ZX + \phi$ 's	$NT_{A}(\phi)$
$A'_{Z'}\Delta + A_{Z}X$ (NT II)	<i>A+A'</i> Z+Z'X'*	A+A' - a' Z +Z' - z'X'' +	NT <sub>F</sub>
		<i>а'<sub>z'</sub></i> Х'''+ф's	
$A'_{Z'}\Delta$ (NT II)	<sup>A</sup> zX*	$^{A}ZX + \phi$ 's	NT <sub>T</sub>

The scheme of the nuclear transmutations grasped by the TNCF model is tabulated in Table 3.1.

We are able to clarify the nature of the CF-matter in relation to the NTs of the first kind (NT I) and the second kind (NT II). It is shown that the physical meaning of the two types of the CF-matter, type-I and type-II. The former is formed in CF materials when electrolysis is done with relatively low current density, and responsible for NT I, while the latter is formed there when electrolysis is performed with relatively large current densities including critical electrolysis and supra-critical electrolysis (glow and arc discharges), and results in NT II. The type-I CF-matter seems to correspond to the neutron star matter at lower density while the type-II CF-matter to the neutron star matter at higher density as shown by simulation by Suess and Urey [Suess 1956].

## **3.2** Foundation of the TNCF Model

It has been a common sense in physics that the environment in a crystal around an interstitial proton/deuteron does not have strong influence on the two-body interaction between a lattice nucleus and an interstitial proton/deuteron [Kozima 1998 (Sec 12.2d), 2006 (Sec. 3.4.3)]. However, the investigation of the bases of the TNCF model, a part of which was given in the former book [Kozima 1998 (Secs. 12.4 - 12.8)], has given us an idea of the super-nuclear interactions between lattice nuclei mediated by interstitial protons/deuterons which results in formation of neutron energy bands presupposed before [Kozima 1998 (Sec. 12.4)]. The super-nuclear interaction has a similarity in the superexchange interaction between Mn ions mediated by oxygen ions in MnO anti-ferromagnetism (e.g. [Suzuki 2009 (Introduction), Timm 2009 (Sec. 4.3)]). Thus, it was shown that the two neutrons at lattice points far out of the range of nuclear force of about one fermi (=  $10^{-15}$  m) can interact each other by the mediation of wavefunctions of a proton (or a deuteron) at interstitial sites extended to the lattice points where are the neutrons.

Furthermore, the exotic nuclei with large excess of neutron numbers over those of stable nuclei have recently investigated extensively. Existence of these exotic nuclei with extended distribution of excess neutrons has encouraged the investigation of the super-nuclear interaction [Kozima 2006 (Sec. 3.7), 2008b, 2009].

# **3.2.1** The Super-nuclear Interaction between Lattice Nuclei mediated by Interstitial Protons/Deuterons

There is another factor which encouraged the calculation of the super-nuclear interaction; the extension of proton/deuteron wavefunctions in an interstitial sites in fcc transition metals. It is well known that wavefunctions of proton and deuteron in Pd, one of the best investigated transition-metal hydrides, are not localized at the interstitial sites but extended to overlap with neighbouring lattice points.

On the other hand, neutrons in an outermost shell of a lattice nucleus have rather extended wavefunctions, especially when the nucleus is in an exotic state; a state with a large excess number (A - Z) of neutrons over the number Z of protons; (A - Z) >> Z.

Therefore, the extended distribution of the neutrons in an outermost shell of a lattice nucleus (possibly an exotic nucleus) and the non-localized wavefunctions of interstitial protons/deuterons are favorable for the super-nuclear interaction to realize the neutron energy bands [Kozima 2006 (Sec. 3.7), 2008b, 2009].

#### **3.2.2** Formation of the Neutron Energy Bands and CF-Matter

Once the neutron energy bands are formed by lattice neutrons (neutrons in lattice nuclei) with the super-nuclear interaction, the neutrons in the neutron valence band accumulate at boundary/surface regions of the CF material to form a CF-matter with a density around  $10^8 - 10^{12}$  cm<sup>-3</sup> as determined by the analyses of experimental data sets [Kozima 1998 (Tables 11. 2 and 11.3), 2006 (Tables 2.2 and 2.3).

It should be emphasized again here that the neutrons in a neutron energy band do not interact with lattice nuclei such as Pd and Ni in PdD or NiH as described by the Bloch Theorem [Seitz 1940, Kittel 1976]. They interact with foreign nuclei at crystal surfaces or disordered nuclei from lattice points of the CF-material. This is the cause of the localized appearance of the fusion products in the CFP.

This simple fact has been forgotten in researchers in the field of CFP, even if it has been clearly written down in textbooks for instance as follows;

"These solutions are composed of traveling waves, and they can be assembled into wave packets to represent electrons that propagate freely through the potential field of the ion cores." [Kittel 1976 (p. 190)]

#### **3.3 CFP in CF Materials by the CF-Matter Formation**

Thus, from our point of view, it is necessary to have the CF-matter by any means to realize the CFP in a CF material. In order of preparation of the CF material, we can enumerate the necessary conditions for the realization of this state as follows.

#### **Necessary Conditions for and Characteristics of the CFP in CF materials:**

(1) Absorption of protium/deuterium into the material (at least in the surface regions thicker than at least about 200 A = 20 nm) with a density as high as that of the host element [Kozima 2000 (Sec. 4)]. It is said that the necessary average density of the hydrogen isotopes should be larger than a minimum value, given for instance by the composition ratio D/Pd > 0.7 in the case of PdD<sub>x</sub> [McKubre 1993].

(2) Formation of the superlattice composed of a sublattice of the host nuclei at the lattice points and another sublattice of protons/deuterons at the interstitial sites in localized regions of the material. The self-organization, a cooperative process, might be relevant to this superlattice formation [Kozima 2013, 2016b (Sec. 3.8)]. This condition relates closely with a characteristic of the complexity, and is one of the most effective causes for the qualitative reproducibility, i.e. the lack of the quantitative reproducibility. We can expect at best only a qualitative reproducibility in the CFP which is in accordance with the notorious irreproducibility of events in the CFP.

(3) Existence of lattice nuclei with neutrons at evaporation levels (where the exotic nuclei are favorable) [Kozima 2016c]. The neutrons in the outermost shell of a lattice nucleus may be responsible for the super-nuclear interaction explained below.

(4) Existence of non-localized proton/deuteron wavefunctions in the material. It is probable that the higher the diffusivity of protons/deuterons, the larger the extension of their wavefunctions, from experimental data in solid state physics [Kozima 2009]. This is the reason that the CFP is frequently observed in hydrogen occluding transition-metals at near room temperature.

(5) Realization of the super-nuclear interaction between lattice nuclei (mediated by interstitial protons/deuterons) [Kozima 2006 (Sec. 3.7.2), 2016c (Sec. 5.2.3)]. Overlapping of a proton/deuteron wavefunction and a neutron wavefunction results in the attractive interaction of the proton/deuteron and the neutron in the first order, and then in the super-nuclear interaction between two neutrons in different lattice nuclei in the second order approximation.

(6) Formation of the neutron energy bands by the coupled neutrons with the super-nuclear interaction [Kozima 2006 (Sec. 3.7), 2016c], Mediation of the super-nuclear interaction is realized by the interstitial hydrogen isotopes in transition-metal hydrides/deuterides or by the hydrogen atoms of organic molecules in the polyethylenes and biological cultures.

(7) Supply of neutrons into one (valence band) of the formed neutron energy bands [Kozima 1998 (Sec. 8.3), Kozima 2006]. Neutrons may be supplied by excitation from lower energy levels of lattice nuclei or by neutrons from outside absorbed by the CF material.

(8) Accumulation of neutrons at boundary/surface region of the CF material to form the CF-matter [Kozima 2006 (Sec. 3.7.2.3), 2011, 2016b (Sec. 3.5)]. It is recognized that the CF-matter is classified into two kinds; (1) type-I CF-mater where occur nuclear transmutations of the 1<sup>st</sup> kind (participation of a single neutron) only and (2) type-II CF-matter where occur nuclear transmutations of the 2<sup>nd</sup> kind (in addition to that of the 1<sup>st</sup> kind) with participation of a neutron drop  ${}^{A}_{Z}\Delta$  (cf. Sec. 4).

(9) Strong interaction between particles in the CF-matter is responsible for the dissipation of excess energies liberated by nuclear reactions of the  $1^{st}$  and  $2^{nd}$  kinds the results of which are observed as nuclear transmutations. This could be one of the reasons why we do not observe radiations after nuclear reactions that are unavoidable in free space.

(10) Interaction of neutrons in the neutron energy bands with disordered nuclei and with foreign nuclei [Kozima 2016c (Sec. 5.2)]. It needs hardly be said that the neutrons in the neutron energy bands do not interact with lattice nuclei at their regular array as its nature but interact with nuclei in such disordered sites as displaced positions in the lattice and at boundary regions. This is the cause of localization of nuclear reactions at surface/boundary regions which have observed frequently in experiments.

Nuclear transmutations observed hitherto in CF materials have been explained at least qualitatively and quantitatively sometimes by the TNCF model [Kozima 1998, 2006, 2009, 2014a (Sec. 4), 2016c].

A word should be mentioned on the relation of the CF-matter proposed in our model and the neutron star matter investigated in nuclear physics. The neutron star matter has been taken up in the investigation of the neutron star (e.g. [Suess 1956]) and treated in a uniform space without boundary. On the other hand, the CF-matter is formed by neutrons in neutron energy bands made of neutron eigenfunctions in lattice nuclei in a superlattice with irregularities and boundaries by which the arrangement of the superlattice is disturbed. These disturbances on the CF-matter surly cause perturbation on the behavior of neutrons in the CF-matter, and become causes of interactions between the neutron drops and foreign nuclides resulting in the nuclear reactions described in Eqs. (3.4) - (3.8). This scenario is told by phantasy on the basis of the successful explanation of the CFP by the TNCF model, and should be explained quantitatively by mathematics which seems to acquire tremendous works on the computer. We have to wait anyone dares to do this work.

# 4. Analysis of the Data of the Nuclear Transmutations (NTs) by the TNCF Model

The experimental results on the nuclear transmutation in critical and supra-critical electrolysis are analyzed and interpreted using the TNCF model in consistency with other data in the cold fusion phenomenon in 3d and 4d transition-metal hydrides and deuterides charged by the normal electrolysis and gas contact method.

# 4.1 Necessity of critical and supra-critical electrolysis for such CF materials as Graphite and 5d transition-metals

As is well known in the history of CF research in these more than 25 years, such 3d and 4d transition metals as Ti, Ni and Pd used in the CF experiments at the normal electrolysis condition are known as hydrogen occluding metals where hydrogen isotopes are absorbed into the metal, and form alloys with the host element (occluded). Generally speaking, in these alloys, protons/deuterons have high diffusivity, and their wavefunctions are not localized at an interstitial site but extend out to cover the lattice sites where are the nuclei of the host element.

On the other hand, as shown in Appendix A, the CF materials such as graphite and 5d transition-metals do not occlude hydrogen isotopes at near room temperature, and have not been much investigated as the materials in relation to the occlusion of hydrogen isotopes. However, at higher temperatures above several hundred °C, physical properties of these materials become similar to the 3d and 4d transition-metal hydrides/deuterides at near room temperature, as shown partly in Appendix A.

Therefore, the theoretical investigation based on the TNCF model explained in Section 3 predicts that the graphite and 5d transition metals will be CF materials at higher temperatures where they have ability to occlude enough amount of hydrogen isotopes.

Here is an interesting prediction we are able to deduce from the necessary conditions given in Subsection 3.3. The necessary conditions for realization of the CFP in CF materials tell us that it is necessary to form hydrides with a large ratio (e.g. H/W in WH<sub>x</sub> compound with  $x \sim 1$ ) of proton/deuteron to host metal. The Necessary Condition (2) demands a lower limit for the density of protium in the CF material to form a superlattice by a cooperative process. Then, there should be a threshold value of the input power  $\varepsilon_{th}$  (a product  $i_{th} \times V_{th}$  of the current density  $i_{th}$  and voltage  $V_{th}$ ) of electrolysis (e.g. in Au/H<sub>2</sub>O/Pt system) and the threshold duration  $\tau_{th}$  of electrolysis for realization of the CFP.

About the effect of the input power  $\varepsilon$  and the duration of electrolysis  $\tau$  on the

CF-matter formation (Necessary Condition (8)), we will be able to take up the product of the two quantities,  $P \equiv (\varepsilon \times \tau)$ , in a rough estimation. To feed enough amount of protium/deuterium to CF materials which have positive heats of solution (W, Pt and Au as shown in Fig. A1), it is necessary to have a larger  $\varepsilon \times \tau$  value larger than a threshold value  $P_{th} = \varepsilon_{th} \times \tau_{th}$ . Then, the condition (8) is written down as an inequality  $\varepsilon \times \tau > P_{th}$ . The area of  $\varepsilon$  and  $\tau$  satisfying this condition is shown schematically in Fig. 4.1 above the line  $P_{th} = \varepsilon \times \tau$  (with  $\varepsilon \equiv x$ ,  $\tau \equiv y$ , and  $P_{th} = 1$ ).



Fig. 4.1 The region  $\varepsilon \times \tau > P$  is shown schematically in this non-dimensional figure as the area above the dotted curve  $\varepsilon \times \tau = P_{th} = 1$  ( $\varepsilon = x$ ,  $\tau = y$ , and  $P_{th} = 1$ ). In this region, it is possible to satisfy the necessary condition (8) and to realize the CFP according to the TNCF model.

There are no systematic investigations of the behavior of the CF materials about this relation depicted in Fig. 4.1 yet. However, we are able to speculate some quantitative relations on the limiting behavior of the input power  $\varepsilon_{th}$  and the threshold duration  $\tau_{th}$  of electrolysis in Au/H<sub>2</sub>O/Pt systems. The relations of the input cell voltage and the elapsed time are shown in Fig. 4.2 for an experiment with the electrolysis current density of 0.2 A/cm<sup>2</sup>. From these data shown in Fig. 4.2, we may tentatively take  $\varepsilon_{th} \sim 0.2 \times 5$  AV/cm<sup>2</sup> and  $\tau_{th} \sim 3600$  s, and therefore  $P_{th} \sim 3.6 \times 10^3$  AVs/cm<sup>2</sup>.



Fig. 4.2 The temporal dependences of the solution temperature (1) - (3) and the input potential (1') - (2') after starting electrolysis on gold electrodes: In Fig. 4.2 (a), (1) and (1') for H<sub>2</sub>O + Na<sub>2</sub>SO<sub>4</sub>, (2) and (2') for H<sub>2</sub>O + K<sub>2</sub>CO<sub>3</sub>, and the temperature level (3) when P<sub>ex</sub> = 0; In Fig. 4.2 (b), (1) and (1') for D<sub>2</sub>O + K<sub>2</sub>CO<sub>3</sub>, while (2) the temperature level when P<sub>ex</sub> = 0 (after [Ohmori 1997b (Fig. 2)]).

Qualitatively, this prediction is consistent with experimental facts; (1) In the case of graphite, P should be very large as given by the arc discharge, (2) in the case of Au, it is necessary to have large values of  $\tau$  (by long term electrolysis) to make the P large, (3) in the case of W and Pt, it is necessary to have critical and supra-critical electrolysis to have a large value of  $\varepsilon$  to make the P large. In the case of Pd/H<sub>2</sub>O/Pt system, we need a larger  $\varepsilon$  by a longer electrolysis time or by a larger current density than in the case of Pd/D<sub>2</sub>O/Pt system.

This prediction will be best illustrated by the extensive experiment with Au/H<sub>2</sub>O/Pt system performed by Ohmori et al. (cf. Sec. 2.3 and [Ohmori 1997b]).

# 4.2 Analysis of the experimental data sets on the nuclear transmutation in normal, critical and supra-critical electrolysis with graphite, W, Re, Au and Pd cathodes.

The nuclear transmutation (NT) is the most remarkable and astonishing facts we encountered in the experimental results obtained in the CFP, and we have given a unified explanation for the whole data sets on them using the TNCF model [Kozima 1998 (Sections 11.11 and 11.12), 2006 (Sec. 2.5), 2011b, 2014a]. Especially, the abundant production of iron isotopes with similar isotopic abundances to the natural ones suggested the nature of the CF-matter in this case is similar to the environment where the elements were composed in nature as pointed out before [Kozima 2011b].

Thus, the explanation of extraordinary data sets obtained in the papers taken up here depends largely on "the stability law" concluded from experimental data as formulated by us [Kozima 2011b], and is necessarily qualitative.

# 4.2.1 Nuclear Transmutations (NT) in the Cold Fusion Phenomenon in General

Explanation of the experimental data sets presented in Sec. 2 is given in this Section using the TNCF model (Sec. 3.1) taking into consideration the facts of occluded H/D in metals given in Sec. 3.2. Especially the data sets obtained in Pd and Au cathodes are discussed in the Section 4.2.2.1 and 4.2.2.2, respectively, for their interesting features among others.

Fundamental nuclear reactions used for the explanation of experimental data are following ones (1) NT of the 1<sup>st</sup> kind (NT I) where participates a single neutron assumed in our original TNCF model and (2) NT of the 2<sup>nd</sup> kind (NT II) where participates a neutron drop assumed in our extended model (cf. Section 3.1);

(1) NT of the 1<sup>st</sup> kind (NT I),  

$$n + {}^{A}_{Z}X \rightarrow {}^{A+1}_{Z}X \rightarrow {}^{A+1}_{Z+1}Y + e^{-} + \underline{\nu}_{e}, \text{ (beta decay of } {}^{A+1}_{Z}X) \qquad (3.1)$$

$$\rightarrow {}^{A-A'+1}_{Z-Z'+1}Y + {}^{4}_{2}\text{He}, \text{ (alpha decay of } {}^{A+1}_{Z}X) \qquad (3.2)$$

$$\rightarrow {}^{A-A'+1}_{Z-Z'+1}Y + {}^{A'}_{Z'}Y', \text{ (fission of } {}^{A+1}_{Z}X \text{ into } Y \text{ and } Y') \qquad (3.3)$$

and

(2) NT of the 2<sup>nd</sup> kind (NT II),  

$${}^{A}_{Z}\Delta + {}^{A'}_{Z'}X \rightarrow {}^{A-a}_{Z-z}\Delta + {}^{A'+a}_{Z'+z}X^{*},$$

$$\to {}^{A-a}{}_{Z-z}\Delta + {}^{A'+a-a'}{}_{Z'+z-z'}X'' + {}^{a'}{}_{z'}X''', \qquad (3.5)$$

$${}^{A'}_{Z'}\Delta + {}^{A+1}_{Z}X^* \to {}^{A'}_{Z'}\Delta^* + {}^{A+1}_{Z}X \to {}^{A'}_{Z'}\Delta + {}^{A+1}_{Z}X + x,$$

$$(3.6)$$

$${}^{A'}_{Z'}\delta + {}^{A}_{Z}X \longrightarrow {}^{A+A'}_{Z+Z'}X^* \longrightarrow {}^{A+A'}_{Z+Z'}X + x.$$

$$(3.7)$$

$${}^{A}_{Z}\Delta \to {}^{A}_{Z}X, \tag{3.8}$$

Where  $\underline{v}_e$  is the anti-electron neutrino.

Then, we can explain the data of nuclear transmutations observed in the 5d transition-metal cathodes using these reactions (3.1) - (3.3) and (3.5) - (3.8).

We give only several examples of explanation in this Section. At first, we give a part of the abundance of elements in universe is tabulated in Table 4.1 [Suess 1956] for convenience of later discussion. In this table,  $log_{10}H$  means the abundance of elements in universe, and Fe is at a marked peak of the  $log_{10}H$  vs. *Z* diagram (cf. [Kozima 2006 (Fig. 2.11)] for the whole diagram).

	0.0					L			
Ζ	13	14	20	24	25	26	28	29	30
Element	Al	Si	Ca	Cr	Mn	Fe	Ni	Cu	Zn

Table 4.1 Peaks of  $log_{10}H$  of elements in the universe [Suess 1956].

## (a) The nuclear transmutation in graphite/H<sub>2</sub>O system at arc discharge

The experimental data sets obtained in graphite-H<sub>2</sub>O system have been analyzed using the TNCF model before [Kozima 2012]. Here, we discuss only a data of isotopic abundance of iron isotopes generated in the graphite-H<sub>2</sub>O system. The isotopic ratios of iron generated in the experiments were the same as that of the natural ones. This correspondence of the products of CF reactions and the primordial production of elements in nature has been interpreted to show an interesting fact of "the stability law" deduced from an overall comparison of CF data with the abundance of elements in universe [Kozima 2011b].

# (b)Data from experiments with W/H<sub>2</sub>O+Na<sub>2</sub>SO<sub>4</sub>/Pt system [Ohmori 2000b]

Here, we take up only the production of  ${}^{A}_{26}$ Fe and  ${}^{A}_{75}$ Re from the data obtained and tabulated in Table 2.1. The *A* and *Z* of nuclides relevant to this system should be noticed at first;  ${}^{A}_{74}$ W (A = 180 - 186),  ${}^{A}_{11}$ Na (A = 23),  ${}^{A}_{78}$ Pt (A = 190 - 198).

# (b1) <sup>56</sup>Fe and <sup>57</sup>Fe with similar isotopic abundances to the natural ones.

This characteristic of isotopic abundances of the product Fe presented in Sec. 2.1.3 is noticed several times before (e.g. (a) above), and suggested the nature of the CF-matter resembling to the environment when elements were formed in nature. The CF-matter comprises neutron drops  ${}^{A}_{Z}\Delta$  with extensive values of A and Z. It is conceivable that the nuclear reaction occurs where occurs a transformation reaction from a neutron drop to a nucleus (3.8) with a large probability for abundant A and Z according to the stability effect;

 ${}^{A}Z\Delta \rightarrow {}^{A}ZX$ 

# (b2) <sup>185</sup>75Re and <sup>187</sup>75Re with different isotopic abundances from the natural ones.

Let us check the natural abundances of these product isotopes first; their natural abundances are 37.07 and 62.93 (with a ratio **0.59**) while the detected abundances are **48.4 and 51.6**, respectively. The ratio of the abundances of the products is 48.4/51.6 = 0.94, higher than the natural case by a factor of 1.6.

The simplest reactions producing  $A'_{75}$ Re from the cathode elements  $A_{74}$ W is the following;

$${}^{A'}_{Z}\Delta + {}^{A}_{74}W \rightarrow {}^{A+A'}_{75}\text{Re} (A + A' = 185, 187)$$
 (4.1)

Let us take tentatively A = 184 and 186 to meet the experimental results A + A' = 185and 187. Then, the neutron drop participating in the reaction (3.8) is  ${}^{1}_{0}\Delta = n$ , and the products reflect the abundances of  ${}^{A}_{74}W$  (A = 184 and 186) which are **30.64 and 28.41**. The ratio of these abundances is **1.08**. This value is compared to the observed value **0.94** closer than the natural value 0.59. This result suggests favorable ability of the TNCF model, if the products  ${}^{A}_{75}Re$  directly reflect the originals  ${}^{A}_{74}W$ . This point is discussed further in Section 4.5.

It should be noticed that there are possibilities generating  ${}^{A}_{75}$ Re directly by transformation of the neutron drop in addition to these reactions (4.1);

 ${}^{A}_{75}\Delta \to {}^{A}_{75}\text{Re} \quad (A = 185, 187)$  (4.2)

#### (b3) <sup>A</sup><sub>82</sub>Pb with different isotopic abundances from the natural ones

There are three isotopes of lead  ${}^{A}_{82}$ Pb (A = 206, 207 and 208) found in the surface layers of W cathode with abundances 38.5, 55.0 and 6.5 compared to the natural once 25, 22 and 52, respectively. A candidate of the reactions producing these lead isotopes from the tungsten isotopes are following;

 ${}^{24}_{8}\Delta + {}^{A}_{74}W \rightarrow {}^{A+24}_{82}Pb \ (A = 182, 183, 184)$  (4.3)

It is interesting to notice a fact appeared here that a neutron drop with a proton number Z of a multiple of 2 and a nucleon number A of a multiple of 4. These kinds of neutron drops are frequently encountered in past in the analyses of experimental data of the nuclear transmutation (e.g. [Kozima 2014a (Sec. 5.2)]).

# (c) Data from experiments with Re/H<sub>2</sub>O+K<sub>2</sub>CO<sub>3</sub>/Pt system [Ohmori 2000a]

The experimental data obtained by Ohmori et al. [Ohmori 2000a] show that there are detected new elements  ${}^{A}_{26}$ Fe and  ${}^{A}_{29}$ Cu or  ${}^{A}_{30}$ Zn in different electrodes. The *A* and *Z* of nuclides relevant to the original system should be noticed at first;  ${}^{A}_{75}$ Re (*A* = 185, 187),  ${}^{A}_{19}$ K (*A* = 39, 40, 41) and  ${}^{A}_{78}$ Pt (*A* = 190 – 198).

The production of Fe is interpreted similarly to the case of (a) graphite and (b) W

cathodes. The productions of Cu and Zn also may be explained by the stability law using the data shown in Table 4.1. The elements Fe, Cu and Zn are at peaks of the  $\log_{10}H$  vs. Z diagram. The accidental appearance of Fe and Cu or Zn in this experiment may reflect the unknown nature of the CF-matter formed at the surface region of Re cathodes.

# 4.2.2 Specific Explanation of the Data Sets obtained in Pd and Au Cathodes

The data sets obtained in the Pd and Au cathodes are given in this section due to their extensive and interesting features showing several characteristics of the CFP in electrolytic systems.

### 4.2.2.1 Pd Cathode

The experimental results on the systems with Pd cathodes are introduced in Sec. 2.1.2. There are two types of experiments with the Pd cathode, one in the system  $Pd/D_2O/Pt$  and another in Pd/H<sub>2</sub>O/Pt system.

#### (a) Pd/D<sub>2</sub>O+Li<sub>2</sub>CO<sub>3</sub>/Pt with current densities of 0.2 – 0.8 A/cm<sup>2</sup> [Mizuno 1998].

In this experiment, the current density was below the limit of the critical electrolysis, but reaches to the limit as the authors call the condition "Strong Electrolysis" in the title of the paper [Mizuno 1998]. This condition of the experiment seems reflected in the experimental results as explained below.

The abundant production of 3d transition-metals may be the result of the stability effect reflecting the log<sub>10</sub>H peaks shown in Table 4.1. This may show the excited or turbulent state of the CF-matter in the Pd surface layers in this experimental condition.

On the contrary to the case 4.2.1a graphite/H<sub>2</sub>O/graphite and 4.2.1b W/H<sub>2</sub>O/Pt systems where was observed generation of iron isotopes  ${}^{A}_{26}$ Fe with the same isotopic abundances to the natural ones, the data of  ${}^{A}_{24}$ Cr had shown difference from the natural abundances. This may be an indication of weaker turbulence of the CF-matter in this Pd/D<sub>2</sub>O+Li<sub>2</sub>CO<sub>3</sub>/Pt system than those in graphite/H<sub>2</sub>O/graphite and W/H<sub>2</sub>O/Pt systems considered in Sections 4.2.1a and 4.2.12b.

# (b) Pd/H<sub>2</sub>O+Na<sub>2</sub>SO<sub>4</sub>/Pt with critical electrolysis at current density of 2 A/cm<sup>2</sup> [Ohmori 1997b, 2016]

The experimental data in this system include many facts related to the nuclear reactions occurring in the surface layers in Pd cathodes at the critical electrolysis. It is certain from discussions given in the above sections that the CF-matter in this case is

rather turbulent than the CF-matter formed in cases of normal electrolysis considered in our papers and books before [Kozima 1998, 2006, 2014a]. We take up two data sets in this system leaving other results common to results in other papers analyzed in other subsections in this Section 4.2.

#### (b1) Isotopic abundances of Pd in surface layers

Experimental result shows that the content of heavier palladium isotopes tends to increase, whereas the content of lighter ones tends to decrease. This qualitative tendency of upwards shift of isotopic abundances of palladium  ${}^{A}_{46}Pd$  is a natural result of following reactions of a neutron and palladium nuclide, but is complicated by the reactions following the formation of intermediate nuclide  ${}^{A+1}_{46}Pd^*$ ;

 $n + {}^{A}_{46}\mathrm{Pd} \to {}^{A+1}_{46}\mathrm{Pd} *.$  (4.5)

### (b2) Isotopic abundances of <sup>A</sup>48Cd generated in surface layers

The generation of cadmium from Pd is most simply explained by the reaction between a neutron drop and a palladium nuclide  ${}^{A}_{46}$ Pd;

 ${}^{A'_2}\Delta + {}^{A}_{46}\text{Pd} \rightarrow {}^{A+A'}_{48}\text{Cd}^* \rightarrow {}^{A+A'}_{48}\text{Cd} + \text{phonons.}$  (4.6)

The intermediate nuclide  ${}^{A+A'}_{48}$ Cd\* is in the CF-matter surrounding it, and is succeeded by reactions expensing excess energy liberated at the second step to the lattice instead of a photon in the case of free space.

Experimental data of isotopic abundance of cadmium  ${}^{A}_{48}$ Cd is tabulated in Table 4.2 using the data given in [Ohmori 2016].

Table 4.2 Isotopic Abundances (%) of  ${}^{A}_{48}$ Cd. Natural abundance  $x_{0A}$  and the average value  $\underline{x}_{A}$  of two values obtained at two points on the sample surface in the experiment [Ohmori 2016 (Fig. 12)] are tabulated according to the atomic number A. The measured abundance  $\underline{x}_{A}$  (%) is read out from the original data given as a histogram [Ohmori (Fig. 12)].

A of Cd isotope <sup>A</sup> 48Cd	110	111	112	113	114	116
Natural abundance $x_{0A}$ (%)	12.39	12.75	24.07	12.26	28.86	7.58
Measured abundance $\underline{x}_{A}$ (%)	0	8	34	56	0	0

Table 4.3 Natural abundances  $x_{0A}$  (%) and absorption cross sections for a thermal neutron (barns) of  ${}^{A}_{46}$ Pd.

A of Pd isotope <sup>A</sup> 46Pd	102	104	105	106	108	110
Natural abundance $x_{0A}$ (%)	0.98	11.0	22.3	27.3	26.7	11.8
Absorption cross-section (barn)	3.36	0.52	20.2	0.30	8.50	0.22

To investigate further details of the reaction (4.6), we tabulate here the isotopic abundances  $x_0$  (%) and absorption cross sections  $\sigma$  of palladium  ${}^{A}_{46}$ Pd for a thermal neutron (barns) in Table 4.3.

The correspondence of the experimental result on Cd (Table 4.2) and the data of Pd in the cathode Pd (Table 4.3) suggests the simplest possible mechanism that the products  ${}^{A}_{48}$ Cd (A = 11, 112, 113) are the results of the absorption of  ${}^{7}_{2}\Delta$  (A' = 7) in the reaction (4.6) by palladium isotopes  ${}^{A}_{46}$ Pd (A = 104, 105, 106). Then, we can deduce an interesting conclusion on the interaction of a neutron drop and a nucleus; the dependence of the probability of the absorption of  ${}^{7}_{2}\Delta$  by  ${}^{A}_{46}$ Pd on A (A = 104, 105, 106) is proportional to 8/11.0 = 0.73, 34/22.3 = 1.53, 56/27.3 = 2.05, respectively. Their ratio is (1: 2.1: 2.8). It is clear that the absorption cross-sections of  ${}^{7}_{2}\Delta$  by  ${}^{A}_{46}$ Pd (A = 104, 105and 106) are independent of the corresponding absorption cross-sections (0.52, 20.2 and 0.30 b) of single thermal neutron tabulated in Table 4.3 with their ratio of (1: 38.8: 0.6).

The supposed participation of the neutron drop  ${}^{7}_{2}\Delta$  is a rare case we met hitherto. The most abundant cases of nuclear transmutations are explained by participation of neutron drops with even values of nucleon numbers A such as  ${}^{4}_{2}\Delta$  and  ${}^{8}_{2}\Delta$ . This may be another exceptional characteristic of a turbulent CF-matter formed in critical electrolysis.

#### 4.2.2.2 Au Cathode

The experimental results introduced in Sec. 2.3 shows that the isotopic abundance of the mercury is different from its natural isotopic abundance [Ohmori 1997b (Fig. 28)]. The content of lighter mercury isotopes, <sup>198</sup><sub>80</sub>Hg, <sup>200</sup><sub>80</sub>Hg increase, while, the content of heavier ones, <sup>201</sup><sub>80</sub>Hg, <sup>202</sup><sub>80</sub>Hg, decrease. Especially the maximum component, <sup>202</sup><sub>80</sub>Hg, in the natural world decreases down to several percent.

The most probable nuclear reactions generating mercury isotope <sup>A</sup><sub>80</sub>Hg from the cathode element, gold with only one isotope <sup>197</sup><sub>79</sub>Au, from our point of view is the following;

$$n + {}^{197}_{79}\text{Au} \rightarrow {}^{198}_{79}\text{Au}^* \rightarrow {}^{198}_{80}\text{Hg} + e^- + \underline{\nu}_e,$$
 (4.7)

with a fairly large neutron absorption cross-section ~ 98.9 barn (if it is applicable to this case). The product nuclide  ${}^{198}{}_{80}$ Hg in the CF material is in the CF-matter surrounding it, and is succeeded by reactions expensing excess energy liberated at the second step to the to the lattice instead of a photon in the case of free space.

$$n + {}^{A}_{80}\text{Hg} \rightarrow {}^{A+1}_{80}\text{Hg}^* \rightarrow {}^{A+1}_{80}\text{Hg} + \text{phonons.}$$

$$(4.8)$$

Here, we cite several data of nuclear physics supposed to be relevant to further

consideration.

Natural abundances of mercury is tabulated in Table 4.4 together with those obtained in the Au electrode after the experiment [Ohmori 1997b (Fig. 28)]. This table shows clearly that the detected mercury nuclei have their origin in the CFP but not in contamination or other artifacts.

Table 4.4 Isotopic Abundances of <sup>A</sup><sub>80</sub>Hg, Natural abundance and data obtained in the experiment [Ohmori 1997b (Fig. 28)]

Nucleon number A	196	198	199	200	201	202	204
Natural abundance $x_{0i}$ (%)	0.15	9.97	16.87	23.10	13.18	29.86	6.87
Experimental abundance $x_i$ (%)		21	15	57	6	5	5

The data of isotopic abundance of product mercury may be tentatively explained using the single neutron reactions (4.7) and (4.8) given above as follows. (It should be remembered that there are other possible reactions induced by neutron drops  ${}^{A}_{Z}\Delta$  with A > 1 and  $Z \ge 0$ ). We may use the cross-sections  $\sigma_{nHg}{}^{(A)}$  of neutron absorption by a nucleus  ${}^{A}_{80}$ Hg given in Table 4.5 and that of  ${}^{197}_{79}$ Au ~ 98.9 barn in these reactions. The first step is naturally the production of  ${}^{198}_{80}$ Hg by the reaction (4.7). The density  $x_0$  (*t*) of gold nucleus  ${}^{197}_{79}$ Au decreases according to the following equation as depicted in Fig. 4.3.

$$x_0(t) = x_0(0) \exp(-c_0 t)$$

where  $c_0$  is a constant proportional to the cross-section  $\sigma_{nAu}$  of neutron absorption by <sup>197</sup><sub>79</sub>Au. This equation is plotted in Fig. 4.3 for illustration;

(4.9)



Fig. 4.3 Decrease of the number of gold nucleus <sup>197</sup><sub>79</sub>Au due to the reaction (4.7).

Then, the next heavier isotope  ${}^{199}{}_{80}$ Hg is produced from  ${}^{198}{}_{80}$ Hg by the Eq. (4.7) from  ${}^{197}{}_{79}$ Au with a rather small cross section  $\sigma_{nHg}{}^{(198)} \sim 0.02$  barn tabulated in Table 4.5. The next process from  ${}^{199}{}_{80}$ Hg to  ${}^{200}{}_{80}$ Hg occurs very fast compared to the former process by the large value of  $\sigma_{nHg}{}^{(199)} \sim 2000$  barn. While the next step from  ${}^{200}{}_{80}$ Hg to  ${}^{201}{}_{80}$ Hg is slower by the smaller cross section of  $\sigma_{nHg}{}^{(200)} < 60$  barn.

1			,		5	5	1 **
Nucleon number A	196	198	199	200	201	202	204
Neutron absorption	(3100)	(0.02)	(2000)	(<60)	(<60)	(4.9)	(0.43)
cross-section $\sigma_{nHg}^{(A)}$							

Table 4.5 Absorption cross-sections  $\sigma_{nHg}^{(A)}$  (in barns) of a neutron by mercury isotopes  $A_{80}$ Hg.

This story gives a qualitative explanation of the accumulation of  ${}^{198}{}_{80}$ Hg and  ${}^{200}{}_{80}$ Hg suggesting also applicability of the TNCF model with the mechanism of single-neutron absorption. To show the process a little more quantitatively, we can calculate the process as follows. This process is simulated by model simultaneous equations for variables  $x_i$  written down as follows with tentative values of  $c_i$ ;  $c_0 \sim c_2 > c_1$ ,  $c_3$ ;

$dx_0(t)/dt = -c_0 x_0(t),$	(4.10)
$dx_i(t)/dt = c_{i-1} x_{i-1}(t) - c_i x_i(t) (i = 1 - 3),$	(4.11)

with initial conditions;

$$x_0(0) = x_0^{(0)}, x_i(0) = 0 \ (i = 1 - 3).$$
 (4.12)

The solutions for  $x_i$  (i = 0 - 3) with constants  $x_0^{(0)} = 10,000$ ,  $c_0 = 100$ ,  $c_1 = 0.01$ ,  $c_2 = 2000$ , and  $c_3 = 60$  are plotted in Fig. 4.4 in semi-logarithmic scale.



Fig. 4.4 Evolution of density  $x_0$  of <sup>197</sup><sub>79</sub>Au and  $x_i$  (i = 1 - 3) of mercury isotopes <sup>*A*</sup><sub>80</sub>Hg (A = 198 - 200, respectively) transmuted by neutron absorption according to the equations (4.7) and (4.8).

The results of the tentative calculation using only a single neutron participation given in Fig. 4.4  $(x_1 > x_3 > x_2)_{\text{th}}$  with parameters taken rather arbitrary duplicate qualitatively the experimental results shown in Table 4.4  $(x_3 > x_1 > x_2)_{\text{ex}}$ , even if the order of  $x_1$  and  $x_3$  is reversed and suggest applicability of the TNCF model in this case again.

#### 4.3 Some Characteristics of the Cathodes with Positive Results

In this section, we want to discuss some minor problems which were not taken up in the above sections. The first is the know-how that is used by experimentalists who performed positive experiments for long years. The second is topographical change of CF materials, another evidence of localization of reactions generating the nuclear transmutation. The third is experimental evidence of development of the CF-matter revealed by the extensive experiments performed by Ohmori et al.

We can boldly speculate on the origin of the two types of CF-matter, type-I and type-II, in relation to the simulation on the neutron star matter [Negele 1973]. Increase of the density  $n_b$  of the neutron star matter results in increase of the number of neutron drops and density of the background neutron-proton medium as shown in Fig. 3.5 of [Kozima 2006a]. It means that the CF-matter is mainly composed of the background neutron-proton medium in the range where  $n_b$  is low (type-I CF-matter) while the number of neutron drops in the CF-matter increases higher where  $n_b$  becomes higher (type-II CF-matter). In the former case, interaction between the CF-matter and a nucleus X may be governed only by n - X interaction, while in the latter case it may be governed mainly by  ${}^{A}Z\Delta - X$  interaction.

Similar situation is replicated in the experiments with 5d transition-metal cathodes (especially with Au cathode) where the increase of electrolyzing current density *i* corresponds to increase of the density  $n_b$  of the neutron star matter. Experimental data shows that increase of the density *i* is accompanied by shift of the nuclear transmutation from NT of the 1<sup>st</sup> kind (NT I) to that of the 2<sup>nd</sup> kind (NT II).

#### 4.3.1 Mechanical treatment – A knowhow to realize the CFP

It is necessary to give a mention on the so-called know-how of CF experiment. There are too many discussions on the reproducibility of CF experiments, and opponents used the lack of quantitative reproducibility to denounce the reality of the CFP.

We have known that there is the know-how in each experimenter to perform their

experiments successfully. From scarce experience of the author, here is introduced know-how used by experimentalists who performed successful experiments for a long year.

"The electrode surface was scraped with a cleaned glass fragment in order to make crystallographically distorted surface." [Ohmori 1998b (p. 36 Experimental)] "One is untreated Pd sheet and the other is cold worked Pd sheet prepared by way of scraping its surface with a cleaned Pyrex-glass fragment." [Ohmori 2004 (p. 36 Introduction)]

"The palladium foil was scratched on the surface, and the area of the scratch was photographed (Fig. 1a)." [Silver 1993 (p. 423 Experimental Methods and Results)] "Experiments have been performed using titanium cathodes with various amounts of deformation by cold rolling, which increased the amount of lattice disorder and stress in the cathodes." [Warner 2000 (p. 161, Experimental Methods and Results)]

# **4.3.2** Topographical change of cathode surface

It is well known that there appears complex surface structures with hills, holes and craters showing melting of the cathode material. A typical figure obtained in an Au cathode is shown in Fig. 2.1 [Ohmori 1997b].

# 4.3.3 Development of the superlattice structure in the CF materials

In Sec. 4.1, we discussed the fulfillment of the necessary condition 2 in the process of electrolysis. The elaborate experiments in the Au/H<sub>2</sub>O/Pt system by Ohmori et al., we can observe another feature of the superlattice formation in the CF material.

In Fig. 4.5, we show the excess heat (W) per unit surface area for Au cathodes; (a) with three kinds of electrolytes Na<sub>2</sub>SO<sub>4</sub>, K<sub>2</sub>SO<sub>4</sub> and K<sub>2</sub>CO<sub>3</sub>, and (b) Excess energy (W) per 5 cm<sup>2</sup> surface area of Au cathode vs. current density (A/cm<sup>2</sup>).



Excess energies on various gold electrodes: (a) Plots of the excess energies against the electrode area, ( $\bigcirc$ ) Na<sub>2</sub>SO<sub>4</sub> ( $\bigtriangleup$ ) K<sub>2</sub>SO<sub>4</sub> ( $\bigcirc$ ) K<sub>2</sub>SO<sub>5</sub> (b) Plots of the excess energies expressed in unit per 5 cm<sup>2</sup> of the electrode against current density. Current density is referred to the true unit area

of the electrode.

Fig. 4.5 (a) Excess energy (W) vs. area of Au cathode and (b) excess energy (W) vs. current density (A/cm<sup>2</sup>) [Ohmori 2000a (Fig. 5)].

In these figures, we see the increase of the superlattice in proportion to the supply of protium (proportional to the current density). We can make an image of superlattice formation in the surface layer of the Au cathode: (1) When protium is supplied to the cathode by electrolysis, protons enter into surface layer of gold at random while its density is low, (2) Increasing the density of proton, there appear islands of superlattice AuH with its total volume proportional to the absorbed protium, (3) Increasing the density of proton further, the volume (or area over the surface) of the superlattice increases by development of individual island or by coalition of islands, (4) Finally, the whole surface of the Au cathode is saturated by the superlattice. The curve in Fig. 4.5 (b) will be saturated in the right with increasing the current density, if the condition of the electrolysis remains the same.

In this point, it should be noticed further the local nature of the superlattice formation as time goes by. Inhomogeneous distribution of islands of the superlattice over the surface, as imagined above, results in the local nature of nuclear reactions in the CFP. As we have discussed often, it is well known in CF research that nuclear reactions in the CFP occur in some localized areas in a surface layer with a width about a few hundred Å [Kozima 2011]. This characteristic is also observed in Au/H<sub>2</sub>O/Pt system as distribution profiles of Fe atoms [Ohmori 1996a] and also as local topographical changes as shown in Fig. 4.3 [Ohmori 1996c].

Here should be noticed another characteristic of the CFP in electrolytic system. The characteristic shown in Fig. 4.4 (a) reveals dependence of the CFP on the electrolyte

used in experiments. As have been discussed before [Kozima 2000], there is an interesting favorable combination of cathode metal and electrolyte such as Pd-Li and Ni-K. This favorable combination of cathode-electrolyte seems purely chemical compatibility, but not well explored yet. A scrutinized investigation of the relation shown in Fig. 4.4 (a) may give a clue for the explanation of this problem.

It should be noticed further that the electrolyte plays an essential role in the CFP. One of the most important examples is the production of helium in  $Pd/D_2O + LiOH/Pt$  systems. The data of helium production observed in the Pd cathode [Morrey 1990] has been analyzed and explained semi-quantitatively in consistent with its excess heat data using the reaction;

 $n + {}^{6}\text{Li} = {}^{4}\text{He} (2.1 \text{ MeV}) + t (2.7 \text{ MeV}),$ 

between a trapped neutron and a lithium nucleus <sup>6</sup>Li absorbed into the Pd cathode from electrolyte [Kozima 1997].

### 4.4 Relation of the CFP with Nucleon Levels in Host Nuclei

As had been discussed in other papers (e.g. [Kozima 2016c]), it is favorable to have neutron orbits of host nuclei at energies near zero for CF materials to realize the super-nuclear interaction mediated by occluded protons/deuterons. The good examples have been seen in Fig. 4.6. We see in Fig. 4.6 that host elements of CF materials effective for the CFP such as Ti, Ni, Pd and C have neutron orbits outermost shell at around the evaporation level, i.e. at around  $E_n \sim 0$  in this figure. This characteristic of these nuclei may have close relation with the appearance of the CFP in such CF materials as TiD<sub>x</sub>, NiH<sub>x</sub>, PdD<sub>x</sub> ( $x \approx 1$ ) and the hydrogen graphite HC<sub>x</sub> (x = 6 - 8) [Kozima 1998, 2006, 2014a].

Similar consideration is applicable to the 5d transition-metal investigated in this paper. The nuclei of host elements investigated in this paper, tungsten  ${}^{A}_{74}W$  (A = 180 - 186), rhenium  ${}^{A}_{75}Re$  (A = 185, 187), platinum  ${}^{A}_{78}W$  (A = 190 - 198), and gold  ${}^{A}_{79}W$  (A = 197) have neutron orbits with energies at around the evaporation level clearly seen in Fig. 4.6. This coincidence may be not accidental but imply deep meaning for the realization of the CFP as discussed hitherto.



Fig. 4.6 Energies of neutron orbits calculated by C.J. Veje [Bohr 1969]

#### 4.5 On the single neutron absorption in the nuclear transmutation

In Sec. 4.2.1 (b3), we have given an explanation of the production of Re isotopes from W cathode using the single neutron absorption followed by beta decay. Similar explanations for the products of nuclear transmutations in the CFP have given rather quantitatively. Two of such examples are given in Appendix C7 of our book [Kozima 2006]. There, the data by Dash et al. [Klopfenstein 1998, Dash 1996] are analyzed by the TNCF model and the experimental results on the isotopic abundances of Ti and Pd are quantitatively explained by the reactions similar to that given in Eq. (3.1).

In these cases, we used the cross-section  $\sigma_{nX}$  of neutron absorption by a nucleus X determined in nuclear physics. In the above explanation of Re production from W in Sec. 4.2.1 (b3), the data have shown the abundance of the original W reflects directly in the abundance of the product Re without knowledge of the absorption cross-section  $\sigma_{nW}$ . We do not know exactly why there is such difference in two cases; (a) Ti and Pd in normal electrolysis and (b) W in supra-critical electrolysis.

One factor we can imagine is the state of the CF-matter in these two cases. In the case (a), there are only nuclear transmutations with sifts of isotope abundance. While, in the case (b), there are more nuclear transmutations, and we need to use the neutron drops to explain them. In the latter case, it seems the CF-matter is rather turbulent and the reaction where a nucleus reacts with a neutron becomes a part of many-particle

reactions including the absorption of a neutron drop (4.3) and transformation of a neuron drop into a nuclide (4.2).

The nuclear transmutation observed in palladium-protium and such hydrogen non-occlusive metals at room temperature as graphite and 5d elements (tungsten, rhenium, gold and platinum) with critical and supra-critical electrolysis are explained by the TNCF model consistently with those observed in 3d and 4d transition-metal hydrides and deuterides with normal electrolysis and also with those observed in XLPE and in biological microbial cultures [Kozima 2016d].

Necessity of critical and supra-critical electrolysis for the CFP in Pd-H system and hydrogen non-occlusive metals seems to heat the samples (CF materials) to higher temperature enough for high proton/deuteron diffusibility and therefore for large extension of wavefunctions of proton/deuteron in the materials ]Kozima 2009]. This fact is consistent with the observation of the CFP in PdD rather than PdH systems in relation to the higher diffusivity of D over H at near room temperatures (cf. Appendix A, Fig. A3).

The effect of the nuclear force between a neutron in a lattice nucleus and an interstitial proton/deuteron resulting in the neutron energy band in the case of the CFP is discussed in Appendix B in relation to the hydrogen diffusion in transition metal hydride.

### 5. Conclusion

The nuclear transmutation observed in CF materials such as palladium-protium and hydrogen non-occlusive metals at room temperature as graphite and 5d elements (W, Re, Au and Pt) with critical and supra-critical electrolysis in electrolytic liquids of light water are explained by the TNCF model consistently with those observed in 3d and 4d transition-metal hydrides and deuterides with normal electrolysis and also with those observed in XLPE and in biological microbial cultures [Kozima 2016d].

Necessity of critical and supra-critical electrolysis for the CFP in Pd-H system and hydrogen non-occlusive metals at around room temperature seems to heat the samples (CF materials) to high temperature enough for high proton/deuteron diffusibility and therefore for large extension of wavefunctions of proton/deuteron in the materials ]Kozima 2009].

The success in analysis of the experimental data obtained in graphite and 5d-transition metals, containing a lot of hydrogen, given in this paper has shown the reality of the nuclear transmutations catalyzed by neutrons in CF materials. It is also suggesting the reality of the neutron energy bands in these CF materials realized by the

super-nuclear interaction mediated by interstitial protons/deuterons proposed by one of the present author (H.K.).

The super-nuclear interaction between lattice nuclei mediated by interstitial protons/deuterons shows in return a possible interaction between interstitial protons/deuterons mediated by lattice nuclei. This interaction may be called the super p-p (super d-d) interaction for convenience of discussion. Details of this interaction and its effects on the solid state physics will be given in Appendix B.

Thus, the extension of occluded proton/deuteron wavefunctions at an interstitial site  $r_j$  until lattice points ( $\mathbf{R}_i$ 's) surrounding  $r_j$  and therefore the nuclear force between the proton/deuteron and a neutron in one of lattice nuclei at  $\mathbf{R}_i$ 's seem to have effect on nuclear physics of the transition metal hydrides/deuterides to induce the CFP on one hand and on solid state physics to induce peculiarity of characteristic diffusivity of hydrogen/deuterium in them on the other.

The effect of the CF-matter on the nuclear physics is characterized by occurrence of nuclear reactions in the transition metal hydrides/deuterides at near room temperature environment which is called by us the cold fusion phenomenon (CFP) to distinguish the phenomenon from other phenomena caused by the nuclear force between particles in the system discussed in solid state-nuclear physics. The nuclear reactions in the CFP are realized by the interaction between disordered nuclei and trapped neutrons (neutrons in neutron bands) and therefore occur only when there is a near ideal superlattice of lattice nuclei and occluded hydrogen/deuterium (with a large density of occluded hydrogen, i.e.  $H/Me \sim 1$ ).

On the other hand, the effect of the super p-p/d-d interaction on the solid state physics seems to occur even when the density of occluded hydrogen/deuterium is not large as in the case of the diffusion of hydrogen in Pd/Ag alloys as shown in Appendix B. This is because of the large density of lattice nuclei that influence the behavior of hydrogen isotopes. We can expect other examples of the effect of the super p-p/d-dinteraction on the hydrogen/deuterium behavior in the solid state physics at higher concentrations. This point is a theme of future investigation.

### Appendix A. Hydrogen isotopes in Metals

[Birnbaum 1972, Sussmann 1972, Springer 1978, Wicke 1978, Voelkl 1978, Puska 1984, Fukai 2005, Kozima 2006 (Sec. 3.6), Griessen 2008]



Fig. A1 Heat of solution of hydrogen in the rows of the periodic system which contains the 3d, 4d and 5d elements. For references see Griessen and Riesterer<sup>15</sup>. [Griessen 2008 (Fig. III.15)]
15. Griessen, R., and T. Riesterer, "Heat of formation models" in Topics in Applied Physics 63 (1988) 219, Springer-Verlag

The data shown in Fig. A1 tells us the high values of heat of solution for the 5d elements (W, Re, Pt and Au) used in the CF experiments introduced in Sec. 2. This thermodynamic data is the cause of high temperature to realize the CFP in them by the critical and supra-critical electrolysis.

Table A1 Heat of solution of hydrogen in the elements of the periodic system [Griessen 2008 (Table III.1)]

Element	Enthalpy of	Enthalpy of
	solution	solution
	(kJ/molH)	(eV/atom)
Не		
Li	-51	-0.528
Be	-2	-0.02
В	-4	-0.041
С		
Ν		
0		
F		
Ne		
Na	2	0.02
Mg	21	0.217
Al	60	0.621
Si	180	1.865
Р		
S		
Cl		
Ar		
K	0	0
Ca	-94	-0.974
Sc	-90	-0.932
Ti	-52	-0.538
V	-30	-0.31
Cr	28	0.29
Mn	1	0.01
Fe	25	0.259
Со	21	0.217
Ni	12	0.124
Cu	46	0.476
Zn	15	0.155
Ga		
Ge	221	2.29
As		
Se	35	0.362
Br		
Kr		
Rb	-54	-0.559
Sr	-61	-0.632
Y	-79	-0.818
Zr	-58	-0.601
Nb	-35	-0.362
Mo	25	0.259
Tc	-12	-0.124
Ru	54	0.559
Rh	27	0.279
Pd	-10	-0.103

Element	Enthalpy of	Enthalpy of
Liement	solution	solution
	(k I/molH)	(eV/atom)
<b>A</b> ~	(KJ/III0IFI)	
Ag	03	0.052
In	-94	-0.974
III Sn	125	1 205
Sh	123	1.295
Те		0.87
I	-04	-0.07
I Ve		
Cs	-56	-0.58
Ba	-88	-0.912
La	-67	-0.694
Ce	-74	-0.766
Pr	-68	-0.704
Nd	-50	-0.518
Pm		
Sm	-70	-0.725
Eu		
Gd	-69	-0.715
Tb	-78	-0.808
Dy	-79	-0.818
Но		
Er		
Tm		
Yb		
Lu	-79	-0.818
Hf	-38	-0.393
Та	-36	-0.373
W	96	0.994
Re		
Os		
Ir	74	0.766
Pt	35	0.362
Au	32	0.331
Hg		
Tl		
Pb	62	0.642
BI		
Po		
At		
Kn En		
Ka A a		
AC		
In Po	-40	-0.414
ra II	7	0.072



Fig. A2 Correlation between the measured heat of formation of binary hydrides and the energy difference  $E_F - E_s$  determined from existing band structure calculations for the host metal [Griessen 2008 (Fig. III.35)]. ( $E_F$ ; Fermi energy and  $E_s$ ; an effective s-band energy defined as the energy where the integrated density of states curve of the host metal equals 1 in the Griessen-Driessen model<sup>39</sup>.) 39. Griessen, R., and A. Driessen, Phys. Rev. **B30** (1984) 4372



Fig. A3 Diffusion of isotopes in palladium [Birnbaum 1972 (Fig. 5)]

H and D diffusion in Pd [Kozima 1998, 2006, 2013a, 2014a, 2016] D has higher diffusibility at room temperature than H while at higher temperatures above 200 °C the situation reverses. This is the reason for the critical electrolysis for Pd-H system to realize the CFP compared to the normal electrolysis for Pd-D system usually performed in CF experiments [Fleischmann 1989, McKubre 1991] It should be remembered that

there have been successful experiments observing the CFP in Pd-H<sub>2</sub>O system at normal electrolysis with a little higher current densities (e.g. [Dash 1994]).

It is interesting to know that Martin Fleischmann noticed this fact shown in Fig. A3 already in 1990 from his point of view as follows;

"Such calculations must also provide answers to other conundrums such as why the diffusion coefficient of D in Pd/D exceeds those of both H in Pd/H and T in Pd/T." [Fleischmann 1990 (p. 347)]

Experiments on the graphite electrodes at arc discharge (e.g. [Hanawa 2000]) was analyzed in our paper [Kozima 2016b].

It is interesting to notice that hydrogen isotopes have low diffusivity, which is the reason the supra-critical electrolysis is necessary as done by Ohmori et al. The data of H and D in Re, Pt, W and Au are given by [Birnbaum 1972, Fukai 2005 (Tables 5.4, 5.10, Figure 5.49)]. H or D have lower diffusivity in Re, Pt and W than in Pd. It means higher temperature is necessary to have the extended wavefunctions of excited states of H and D in these metals. (No data in Au)

### H/D in W, Re and Pt

The vibrational excitation energy of interstitial hydrogen in rhenium (Re) is shown in Table A2 ([Fukai 2005 (Table 5.4)]). The excitation energy of hydrogen in Re ( $\sim$  100, 130 meV at 5 K in ReH<sub>0.09</sub>) is compared with that in Pd  $\sim$  56.0 meV at 15 K in PdH<sub>0.99</sub> and  $\sim$  69.0 meV at 77 K in PdH<sub>0.002</sub> (Table not shown here). The rather high value of the excitation energy in Re tells us the lower diffusivity of hydrogen isotopes in Re than that in Pd which is a typical metal for the high diffusivity of hydrogen.

The binding energy of hydrogen atoms with monovacancies in tungsten (W) is shown in Table A2 ([Fukai 2005 (Table 5.10)]). The large value of the binding energy of proton and vacancy in W (1.43 eV) tells us indirectly that the proton in W is in a rather stable state than that in Pd where the binding energy is lower (0.23, 0.15 eV).

Table A2 Vibrational excitation energies of interstitial hydrogen in metals. The values have been obtained from the observed peaks of inelastic neutron scattering experiments using polycrystalline (powder) specimens. The presence of dispersion in fcc and some hcp metals detracts to some extent from the physical significance of these values. [Fukai 2005 (Table 5.4)]

Structure	Site	$\mathbf{Sample}$	Excitation	n Energy [meV]	Temperature [K]	Reference
bct	0	$VH_{0.5}$	$50.2^{\mathrm{a}}$	223 <sup>b</sup>	80	[5.59]
			56	230	80	[5.72]
			54	220	295	[5.72]
		$VD_{0.5}$	39	164	80	[5.72]
			47		295	[5.72]
2		$VT_{0.5}$	36		295	[5.72]
bcc	т	$\mathrm{Ti}H_{0.14}$	$120(2)^{b}$	$171(2)^{a}$	998	[5.73]
		$VH_{0.012}$	106	$\sim \! 170$	300	[5.58]
		$VH_{0.51}$	113	180	498	[5.74]
		$VD_{0.5}$	82(3)	123(7)	425	[5.75]
		$NbH_{0.03}$	107	163	300	[5.58]
		$NbH_{0.7}$	116.0(7)	167.0(15)	10	[5.76]
		$NbD_{0.85}$	86(1)	120.0(15)	10	[5.76]
		$NbT_{0.7}$	72(1)	101(1)	10	[5.76]
		$\mathrm{TaH}_{0.037}$	114	154	300	[5.58]
		$TaH_{0.08}$	114.0(6)	163.5(8)	300	[5.77]
		$TaH_{0.5}$	121.3(2)	163.4(4)	10	[5.76]
		$\mathrm{TaD}_{0.09}$	84.4(6)	116.0(6)	300	[5.77]
3		$TaD_{0.5}$	88.4(4)	118.7(6)	10	[5.76]
hcp	0	$\mathrm{CrH}_{1.0}$	121		15	[5.53]
		$MnH_{0.86}$	111		100	[5.51]
		$\mathrm{FeH}_{1.0}^{\mathrm{c}}$	105		90	[5.78]
		$\mathrm{CoH}_{0.05}$	118		5	[5.65]
		$CoH_{0.5}$	110		15	[5.65]
		$M \acute{0} H_{1.2}$	113		15	[5.53]
۴	- e1	ReH0.09	100, 130		5	[5.63]
hcp	Т	$\mathrm{ScH}^{\mathrm{d}}_{0.34}$	$103.5(2)^{ m b}$	$147.5(1)^{\rm a}$	8	[5.60]
5		$ScD_{0.34}^d$	79.0(1)	107.3(1)	4	[5.60]
		$YH_{0.18}^d$	100.1(2)	134.2(5)	80	[2.133]
		$\mathrm{YD}_{0.18}^{\mathrm{d}}$	75.8(1)	96.3(1)	80	[2.133]
		$\mathrm{LuH}_{0.19}^{\mathrm{d}}$	102.8(3)	143.5(1)	4	[5.62]
		$\mathrm{LuD}_{0.19}^{\mathrm{d}}$	76.2(1)	101.7(1)	4	[5.62]
		${\rm Ti}H_{0.05}$		141(1)	588	[5.73]
		${\rm Ti}H_{0.07}$	105.5(20)	162.0(25)	599	[5.79]
		$\mathrm{TiD}_{0.09}$		108.4(12)	601	[5.79]
		$\mathrm{ZrH}_{0.05}$		143.1(6)	873	[5.79]
		$\mathrm{Zr}\mathrm{D}_{0.05}$		105.0(10)	765	[5.79]

 Table 5.4. (continued)

<sup>a</sup> doubly degenerate mode
<sup>b</sup> non-degenerate mode
<sup>c</sup> dhcp structure
<sup>d</sup> single-crystal data.

Table A3 Binding energies of hydrogen atoms with monovacancies and bubbles/voids, obtained by various methods; including ion implantation/channeling (I/C), positron annihilation spectroscopy (PAAS), superabundant vacancy formation/thermal desorption spectroscopy (SAV/TDS). All energies are in eV. [Fukai 2005 (Table 5.10)]

Metal	Vacancy	Bubble/Void	Method	Reference
Al	~0.52	≤0.52	I/C	[5.176]
	0.53		PAS	[5.148]
V	0.11		I/C	[5.177]
	0.23		I/C	[5.19]
Cr	0.89, 0.73		SAV/TDS	[4.44]
Fe	0.63,  0.43	0.78	I/C	[5.175]
Ni	0.44, 0.28	0.55	I/C	[5.163]
	0.58		PAS	[5.149]
	0.44		PAS	[5.150]
	0.43,  0.27		SAV/TDS	[5.178]
	0.45,  0.28		SAV/TDS	[5.179]
Cu	0.42,  0.22		I/C	[5.167]
	0.37, 0.23		SAV/TDS	[5.179]
Zr	0.28		I/C	[5.180]
Nb	0.55		PAS	[5.144, 5.145]
Mo	1.03, 0.80	1.15	I/C	[5.176]
	1.4		PAS	[5.149]
Pd	0.23, 0.15	0.31	I/C	[5.153, 5.181]
Ta	0.58		PAS	[5.145]
	0.42	0.53	I/C	[5.182]
W	1.43		I/C	[5.183]

Diffusion data of hydrogen in tungsten at higher temperature from 1100 to 2600 K is given in Table A4 ([Birnbaum 1972]). There is no data of tungsten diffusion at lower temperatures.

The data in Table A2 and A3 tell us that hydrogen is not occluded in tungsten at lower temperatures.

Metal	D <sub>0</sub> (cm <sup>2</sup> /sec)	Q (kcal/mol)	Temperature Range (°K)	Method Ref.
Nb	$2.2 \cdot 10^{-2} \\ 1.8 \cdot 10^{-2} \\ 5.6 \cdot 10^{-2}$	9.37 10.0 19.2	575 - 975 475 - 975 1875 - 2575	Absorption [26] Absorption [27] Desorption [43]
α – Fe	$7.6 \cdot 10^{-4}$ $2.2 \cdot 10^{-3}$ $5.3 \cdot 10^{-3}$ $9.3 \cdot 10^{-4}$ $1.4 \cdot 10^{-3}$ $6.4 \cdot 10^{-4}$ $6.4 \cdot 10^{-4}$ $2.2 \cdot 10^{-3}$	2.3 2.9 3.05 2.7 3.2 1.92 1.33 3.1	675 - 1175 675 - 1175 425 - 1175 475 - 1050 475 - 1050 575 - 1050 283 - 350 283 - 373	Permeability [28] Permeability [29] Desorption [30] Desorption [31] Desorption [32] Desorption [9] Permeability [33] Desorption [34]
Та	$6.1 \cdot 10^{-4} 7.5 \cdot 10^{-2} 1.6 \cdot 10^{-3} 6.5 \cdot 10^{-4}$	3.5 14.4 32.2 3.5	273 - 435 675 - 800 775 - 975 300 - 360	Resistivity [35] Permeability [41] Absorption [42] Lattice Expansion [79]
Мо	7.6 · 10 <sup>-5</sup> 5.9 · 10 <sup>-2</sup> 1.6 · 10 <sup>-1</sup>	8.4 14.7 22.2	525 625 875 1275 1875 2575	Permeability [37] Desorption [38] Desorption [39]
w	4.1 · 10 <sup>-3</sup> 8.1 · 10 <sup>-2</sup> 7.3 · 10 <sup>-4</sup>	9.0 19.8 41.5	1100 - 2400 1900 - 2600 1975 - 2200	Permeability [36] Desorption [42] Thermal Dissociation [40]

Table A4. Hydrogen diffusion data at elevated temperatures – bcc metals. [Birnbaum 1972 (Table 5)].

Change in the lattice location of D atoms implanted in Pt at low temperatures upon subsequent annealing is shown in Fig. A4. [Fukai 2005 (Fig. 5.49)] This data tells us that deuteron in Pt with  $T_D \sim 100$  K is more stable than that in Pd with  $T_D \sim 85$  K. This might be the reason that hydrogen isotopes are diffusive in Pd but not in Pt at around room temperatures.



Fig. A4 Changes in the lattice location of D atoms implanted at low temperatures upon subsequent annealing. The temperatures where interstitials, vacancies and D atoms start migrating are designated, respectively, as  $T_i$ ,  $T_v$ , and  $T_D$ . Lu is an exceptional case where  $T_D$  marks the temperature of the break-up of H-M-H pairs (Sec. 5.6.1). For the geometry of displaced-T (dis-T) and displaced-O (dis-O) configurations, see the text. A transition from solid to dashed line marks the temperature at which D atoms are released from the implanted region. For references, see the text. [Fukai 2005 (Fig. 5.49)]

# Appendix B. Nuclear Interaction between Hydrogen/Deuterium and Lattice Nuclei and Super *p-p* Attractive Interaction in CF Materials

In the analyses of the cold fusion phenomenon (CFP) observed in the transition metal hydrides/deuterides, we have shown that the nuclear force between a proton/deuteron and a neutron in a lattice nucleus resulted in the super-nuclear interaction between neutrons in lattice nuclei, which has given a unified explanation of whole experimental data obtained in protium and deuterium systems altogether. The nuclear force between a proton/deuteron and a neutron in a lattice nucleus has another phase on the behavior of hydrogen/deuterium in these materials.

A proton/deuteron in an interstitial site with extended wavefunction until neighboring lattice points interacts with neutrons in the lattice nuclei at these lattice points. If the proton/deuteron is at an octahedral interstitial site, there are eight neighbors and the interaction may be fairly large; comparable to or stronger than the short-range force with electrostatic nature in some situations.

Increasing the concentration of occluded hydrogen/deuterium, there appears a situation where two protons/deuterons are in adjacent interstitials and they interact with common lattice nuclei surrounding them. Then, there appears the super p-p/d-d interaction between these two protons/deuterons.

We can illustrate the super p-p (super d-d) interaction in CF materials schematically using PdH crystal in two dimensions in Fig. B1. Let us take up a proton j at an interstitial site j and another proton k at site k for illustration. We assume that the wavefunctions of these protons at interstitials overlap with lattice nuclei at i and i'. The proton j interacts with the neutrons at i and i' with the nuclear force and the proton k also interacts with these neutrons. Then, the proton j and proton k interact with each other through their interactions with neutrons in lattice nuclei at i and i'. This interaction of two protons j and k mediated by neutrons in lattice nuclei is called the "super p-pinteraction." This situation is illustrated in Fig. B1.



Fig. B1 Schematic illustration of the super p-p interaction between a proton at j and a proton at k mediated by neutrons in lattice nuclei i and i' in two dimensions.

About the strength of the supposed super p-p/d-d interaction compared to that of the electrostatic interactions among surrounding protons/deuterons and lattice nuclei, we have to calculate it from the first principle or estimate it using experimental data related to this interaction. From the experimental data cited below, we can give a rough comparison between the super p-p interaction with the short-range interaction (between nearest neighbors) of two hydrogens in palladium alloys. There are many unsolved problems in solid state physics of transition metal hydrides with known interactions as shown below. The existence of this super p-p/d-d interaction may give a light to solve these problems, if the effect of the former interaction on the behavior of protons/deuterons in solid state physics seems fairly large with the same order of magnitude to that of the latter.

One of excellent reviews on the physics of hydrogen in palladium and palladium alloys has been given by Wicke and Brodowsky [Wicke 1978]. In this review, the fundamental problem of the physics of Pd-H (D) systems is explained as follows:

"Nevertheless, there are quite a number of details in the mechanism of hydrogen diffusion as well as in the behavior of detailed electronic states in this system not yet fully understood. - - -"

"A basic problem still in discussion is which type of model will be most adequate for handling the attractive hydrogen-hydrogen interactions induced by the elastic strains of the host lattice. In one type of model, particular emphasis is laid upon <u>the long-range</u> <u>part</u> of these interactions, and it is suggested that short-range attractions do not occur, i.e., that the short-range interactions are purely repulsive."

"Macroscopic elasticity theory is involved in this model, and mean-field approximations are used; the statistics are rather cumbersome, if the simple Bragg-Williams approximation is not applied. The other type of model is based upon short-range attractive interaction, acting predominantly between nearest neighbors, such that the concept of pair interaction and the statistical method of the quasichemical approximation can be applied. - - The real condition will be somewhere intermediate, as often in nature, between the two simplifying concepts of short-range and of long-range interactions, but the full treatment of the general case is still lacking." [Wicke 1978 (pp. 73 – 74)] (Underlined at citation).

This explanation on the nature of the hydrogen-hydrogen interaction suggests us that the super p-p interaction explained above with an <u>intermediate-range character</u> may give a possibility to explain the physics of hydrogen diffusion in palladium and palladium alloys.

To investigate possible effects of the super p-p/d-d interaction on the physics of transition metal hydrides/deuterides, we take up two examples below.

(1) First example is the hydrogen diffusion in Pd/Ag alloys. Figures B2 and B3 show diffusion coefficients of H in Pd/Ag alloys as functions of hydrogen potential  $\eta$  (proportional to the logarithm of H<sub>2</sub> pressure, which is a function of *n* (=H/Me), in the case of a film penetration experiment) and atomic ratio *n*, respectively.



Fig. B2. Plots of diffusion coefficients of H in Pd/Ag alloys vs. hydrogen potential  $\eta$  (proportional to the logarithm of H<sub>2</sub> pressure) at 30 °C. [Wicke 1978 (Fig. 3.33)]


Fig. B3. Dependence of Fick's (*D*) and Einstein's (*D*\*) diffusion coefficients and of the thermodynamic factor f on atomic ratio H/Me (= n). [Wicke 1978 (Fig. 3.34)]

The data is not fully understood yet as it is explained in their text as follows: "The decrease of  $D^*$  with increasing *n* can be attributed partly to the vacancy factor (1 - n); the residual influence of concentration on  $D^*$  has not been explained so far." [Wicke 1978 (p. 148)]

If we can imagine that there is the attractive super p-p interaction between protons in Pd/Ag alloys, diffusion is disturbed by this interaction in proportion to the density n of the protons in the lattice in accordance with the experimental data.

(2) The second example is the influence of alloy components on the mobility of hydrogen in alloys  $Pd_{1-x}Me_x$  (Me = Ag, Au, Ni, and Pt). The problem is explained by Wicke as follows:

"The influence of alloy components on the mobility of hydrogen in Pd is remarkably small at low additions. <u>Ag, Au, Ni, and Pt up to about 25 at.% leave the diffusion coefficient and the activation energy nearly unchanged.</u> At higher additions the activation energies increase and the diffusion constants decline in a nearly logarithmic fashion to quite low values for alloys with 50 or 60 at.% of solute." [Wicke 1978 (p. 150)] (Underlined at citation).

This interesting behavior of mobility of hydrogen at low concentrations of several minor components (Ag, Au, Ni, and Pt) up to about 25 % may reveal existence of a

force other than the short-range force between nearest neighbors H and Pd or H and Me. Because the super *p*-*p* interaction with an intermediate-range character is not sensitive to the atomic natures of lattice nuclei and also to the small number of minor components, we can expect the same behavior of protons in  $Pd_{1-x}Me_x$  alloys (Me = Ag, Au, Ni, and Pt) for small values of *x* less than 0.25.

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# The Sociology of the Cold Fusion Phenomenon – An Essay –

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#### Abstract

The history of the researches on the cold fusion phenomenon in the modern industrial society is investigated using the framework of the sociology of the science developed in the 20<sup>th</sup> century. The motivation to find out the cold fusion of deuterons in solids and the effort to establish the science of the cold fusion phenomenon (CFP) are deeply interrelated with the flows of economy and science in the history of the latter half of the last century. The foreseen deadlocks of the social development, especially the exhaustion of energy resources, have been a fear for the future of human beings. The controlled thermonuclear fusion (plasma fusion) of deuterons and tritons was taken up as a final solution for this difficulty and had grown up as a big science (or rather a huge science) since the middle of the 20th century. The CFP was taken up as an alternative to the plasma fusion which had been in a stagnant situation after 40 years since its substantial start in the beginning of 1950s. The CFP thus motivated has been destined to be an alternative to the plasma fusion in the sentiment of many researchers in this field and also in the thought of scientists in the established fields of modern science. The situation has given a biased trend in the majority of researchers engaged in the CFP to take up solely the deuterium systems and to disregard the protium systems from CF materials. This trend has given the research of the CFP a biased character and an unsound development. It need scarcely be said that it is necessary to take up whole materials obtained by experiments not only in deuterium but also in protium systems to establish the science of the cold fusion phenomenon. Overviewing the history of the CFP, we point out necessary conditions for the sound development of the science of the CFP. Possible application of the CFP will follow the realization of the science.

**Key words;** Cold Fusion Phenomenon, Sociology of Science, Big Science, Science and Society

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## **Prologue – Science and Military Technique**

"- - - military exigencies have encouraged the growth of one branch of science, especially in seventeenth-century England." [Merton 1975 (p. 204)]

The foci of scientific interest are determined by social forces as well as by the immanent development of science, as R.K. Merton expressed in his paper published in 1935 [Merton 1975 (9. *Interactions of Science and Military Technique*)]. In the social forces, the military techniques are most strong in a short term to influence the science as history has shown. We can cite sentences on this relation from Merton's paper;

"--- Thus it appears that on the average about 10 percent of the research carried on by the foremost scientific body in seventeenth-century England was devoted to some aspect of military technology. To an appreciable extent, then, this one extrascientific concern tended to focus scientific attention upon a given body of scientific problems.

The relations between military demands and the scientific development of the time were primarily of two sorts. The first involved conscious, deliberate efforts of the contemporary scientists directly to solve problems of military technology. This is what is meant by a *direct* relation. The second type is less apparent, for it concerns scientific attention to problems which, although either imposed or emphasized by military needs, seemed to the scientists as of purely scientific interest. This was called the *indirect or derivative* relation." [Merton 1975 (pp. 208 – 209)] (Italicized at citation)

As the technology prevailed over social relations, the science had become an overwhelming factor as a source of wealth and also as an enterprise of a society. In the 19<sup>th</sup> century, Alfred Nobel already established huge wealth by his inventions of explosives used as armaments; "During his life Nobel was issued 355 patents internationally and by his death his business had established more than 90 armaments factories, despite his belief in pacifism." [Hiskey 2011] It is said that A. Nobel was named as "The Merchant of Death" in a French newspaper and had considered how to improve his public image after his death and decided on leaving his enormous fortune to fund a set of prizes named after himself.

The extravagant explosives, the atomic bomb (A-bomb) and hydrogen bomb (Hbomb) developed after 1942, are the two satanic armaments human beings ever obtained. The principle of the A-bomb is the chain reaction of the uranium fission reactions induced by a neutron liberating the nuclear energy contained in uranium nuclei. The atomic power plant based on this principle to generate energy has been developed accompanied by the production of the A-bombs despite of scarce resource of uranium ore and of the unsolved problem how to treat the nuclear ashes.

The principle of H-bomb is the fusion of deuterons occurring in stars. The deuterium (heavy hydrogen) is contained in sea water about 1:5000 of protium (light hydrogen) and is substantially endless. If we can realize the controlled nuclear fusion reactions of deuterium on the Earth, we can get an infinite source of energy. This is the reason the controlled thermonuclear fusion program has started in 1950s worldwide and the program has been pursued for almost 70 years.

Even if A-bomb and H-bomb are satanic, they have duel characters, direct and indirect relations with science as R.K. Merton defined in the sentence cited above. The indirect effect of the study of controlled thermonuclear fusion derived from the H-bomb has induced development of plasma physics which has explained many problems of ionized gases in space and in stars as by-products. The close relation of the controlled thermonuclear fusion with the cold fusion phenomenon is discussed in the following Sections.

## 1. Introduction

"A new scientific truth is not usually presented in a way that convinces its opponents . . .; rather they gradually die off, and a rising generation is familiarized with the truth from the start." (M. Planck, 1948 cited by T.S. Kuhn in [Kuhn 1972 (p. 81)] from his autobiography [Planck 1948]).

The Planck's sayings cited above reflects surely his experience in the development of quantum theory in the beginning of the previous century. The content of the sayings, however, has deep roots in the sociology of science common to the development of the cold fusion phenomenon (CFP) in these about 30 years. The sociological investigation of the history of the CFP is the main theme of this paper.

The history of the controlled thermonuclear fusion (the plasma fusion) research substantially started in 1950s (cf. Appendices B and C) on one hand and that of the cold fusion phenomenon (CFP) started in 1989 on the other show the overwhelming influence of the former on the latter, as shown by the sociology of the science developed in the century.

In this section, we give a brief survey of the energy problem we have confronted at present in Subsection 1.1, of the destined characteristics of science in the present industrial society in Subsection 1.2, and of the motivations of the cold fusion research in Subsection1.3. The terminology of the cold fusion phenomenon is given in Subsection 1.4. Outline of this paper is given in Subsection 1.5.

## **1.1Energy Crisis in Modern Industrial World**

Search for a stable and limitless resource of energy started in the middle of 20<sup>th</sup> century – resulted in the controlled thermonuclear fusion as the most hopeful candidate (e.g. [Kozima 1998 (Chapter 14)]). The investigations of fusion reactors to realize controlled thermonuclear fusion on the Earth started substantially in 1950s. The nuclear fusion reactions supposed to be useful for the above project are written down as follows (cf. Appendix B):

$D + D \rightarrow T (1 \text{ MeV}) + p (3 \text{ MeV}),$	(1-1)
$D + D \rightarrow {}^{3}\text{He} (0.18 \text{ MeV}) + n (2.5 \text{ MeV}),$	(1-2)
$D + D \rightarrow {}^{4}\text{He} (0.08 \text{ MeV}) + \gamma (23.8 \text{ MeV}),$	(1-3)

while the third reaction is essentially ignored due to its too small branching ratio ( $\sim 10^{-7}$ ) compared to those of the first two.

Many textbooks on the plasma physics have taken up the problem of controlled thermonuclear reactions as a motive force for the growth of this new science (e.g. [Chen 1974]).

It will be very helpful to cite the first several pages on the controlled thermonuclear fusion from the Chen's textbook to understand the situation in which new energy sources had been looked after eagerly. The first four pages of *Section 9 Introduction to Controlled Fusion* are cited in Appendix B [Chen 1974].

ITER (International Thermonuclear Experimental Reactor) project started in 1985 as an international project to explore a controlled thermonuclear fusion machine based on the TOKAMAK concept. To know the outline of the project, we cite the *ITER Story* from the ITER homepage in Appendix C [ITER 1985].

# **1.2Science in the Industrial Society – The so-called "Big Science" and the Scientific Spirit**

Science is a human behavior to construct an image of outside world based on the perception a man received in interaction with that world. It is sometimes supposed that the science thus constructed and described by a logical system of the human being is an objective existence accepted by all human beings. It is true as far as the constructed structure of the science is concerned. It is however necessary to remember an important factor of the science at the first stage of its formation. The science is not constructed automatically from materials accepted by sensory organs such as eyes, ears and skin of human beings. The science is essentially an image of the outside world caught by the intuition or instinct of a human being. The image is then manipulated logically and mathematically to make objectively understandable by others. The popularization of science in modern world made the constructed science written in text books familiar and made the subtlety of the creative works obscure and invisible. We have to recollect the trial-and-error stages of scientific endeavors at its initial stage of every science which we are now accepting as established one as a matter of course.

Science and technology are called sometimes "science and technology" (sciencetechnology complex, science-technology conglomerate) all together and supposed to be the same thing. However, they should be considered different thing. Science and technology contain the same knowledge together but differ in which they aim at. The object of the science is just to know about the target and ends its activity when it is obtained. However, the technology does not finish its activity only by knowing the target but uses it for another purpose. And for these purposes, the scientific knowledges are sometimes rearranged, i.e. "A causes B" is transformed into "To get B, find out A." Of course, it is usual to cultivate new techniques for scientific activity but only for the scientific objects. In technology, however, the purpose of the innovation is not only for the new scientific knowledge but also for the technical application. The pile up of the technical endeavor forms a system of technical methodology different from the system of scientific methodology.

In the modern world of technological economy, the former is overwhelming the latter. The scientific spirit is diminishing as well as aesthetic and moral spirits. In the history of the cold fusion phenomenon, we see how the scientific spirit is overwhelmed by the entrepreneur desire and a science is in agony to be well-born in the world and be recognized its true value.

It is an impressive fact that the atomic power generation started after the A-bomb was used in the war in 1945. The research of the controlled thermonuclear fusion started after the H-bomb was exploded in Eniwetok in 1952. It is said sometimes "The war promotes new science and technology." The use of nuclear power as peaceful utilization of an energy source illustrates this saying exactly.

# **1.3 Curiosity for Strange Events and Competition for Financial Supports**

The cold fusion phenomenon is a curious one which cannot be understood by existing knowledge of modern science developed in 20<sup>th</sup> century after a revolutionary opening of quantum physics in 1900. The field of the cold fusion phenomenon (CFP) is known as that occurred in ordinary temperature and pressure, not so high- or low-temperatures, not so high-vacuum or not so high-pressure conditions which have been explored by the end of the last century as a result of pursuance of novel phenomena such as superfluidity, superconductivity, new states of matter. However, there are many, many events wonderful for the eyes accustomed to phenomena observed in the established fields of modern science; the most wonderful one may be the nuclear transmutations in CF materials including transition-metal hydrides and deuterides, hydrogen graphite, cross-linked polyethylene (XLPE) and microbial cultures as explained in our books (e.g. [Kozima 2006]) and papers (e.g. [Kozima 2016a]). The data sets as a whole, especially the three empirical laws found between observables, have shown that the CFP is a phenomenon to be considered as complexity.

In these 25 years after the discovery of a part of the CFP in 1989 by Fleischmann et al. [Fleischmann 1989], there have been piled up many data sets showing various phases of this phenomenon and some of them have been checked by eyes of critical scientists concluding something true are in them in reality [DOE 2004]. Even then, there is no recognition of reality of the CFP in the researchers in this field and also in scientists in established fields in the world. One of the reasons that the former has been losing the point is their persistence in the Fleischmann's hypothesis which is favorable for its

application of the CFP. The most important cause that the latter has lost their reason is not so simple but understandable from a viewpoint of social science of history.

Thomas S. Kuhn explored a new viewpoint in the history of science to understand the changes of scientific entrepreneur in history using a word "paradigm." [Kuhn 1962] According to his terminology used in the history of science, we extend its usage to scientific research activity in process and apply it to the evaluation of the CFP, a part of it was discovered in 1989 and developed by the trial and error method until now, as a process of formation of a new paradigm between traditional nuclear physics and solidstate physics. This viewpoint gives us a new bird's-eye view of the CFP which is useful to understand the situation we are placed in it.

It is possible to say that "A science starts its life when a new fact is (be) recognized to contradict existing paradigms."

In close relation to the characteristic of science in the modern society, the revolution of science characterized as "the paradigm change" had specific nature. We can depict the nature of the CFP as a new paradigm using several sentences from T.S. Kuhn [Kuhn 1962];

"In the absence of a paradigm or some candidate for paradigm, all of the facts that could possibly pertain to the development of a given science are likely to seem equally relevant." [Kuhn 1962 (p. 15)]

The CFP was a product of a scientific mind to search a new possibility of deuteron fusions naturally confined in solids assisted by practical mind to realize an economical energy source. The research field of the CFP has grown up a huge science demanding a paradigm change.

In the history of modern world after 17<sup>th</sup> century, development of a society has been entangled with the development of science and technology and the structure of this development has been a target of researches in social science. The sociology of science had been developed by sociologists who took up the problem in the 17<sup>th</sup> century England as a typical example of the mutual relation between the social development and the development of science (e.g. [Hessen 1931], [Merton 1938]). It is noticed that there are clearly seen strong influences of the sociology developed by Max Weber [Weber 1904] on their works.

It is necessary to have financial supports for the scientific activity. This phase of the science is also analyzed in the sociology of science and the history of the CFP has been

closely wrapped around by competition for funds (cf. Appendix E). The competition for funds shed shadow on the development of the CFP research and recognition of the CFP by the scientific society.

In recent modern states, science is a part of national projects supported and sponsored by a nation. There appear several funding organizations in the story of the history of researches on the cold fusion phenomenon as given in Appendices E and H.

What we have to recognize about the science of the cold fusion phenomenon (CFP) is the situation of the research on the CFP at present after more than a quarter of a century after its discovery in modern science. To do so, it is helpful to learn the sociology of science which analyzes the social existence of science in general developed mainly Merton and others [Merton 1975]. It is the purpose of this paper to show that the science of the CFP has to satisfy several conditions to be recognized in the modern world in view of the sociology of science.

It may be helpful to readers of this paper to give an overview of the external and internal barriers preventing recognition of the CFP by the established scientific society.

The *external barriers* preventing recognition of the CFP as a part of the modern science are related to the following factors in this field; (1) the biased preference of deuteron systems affected by the plasma fusion (thermonuclear fusion) research, (2) neglect of unified perspective of the experimental facts obtained in protium and deuterium systems altogether, (3) lack of recognition that the CFP belongs to an interdisciplinary science between nuclear and solid-state physics and (4) neglect of complexity in formation of the situation appropriate for the CFP in the materials where the CFP occurs (CF materials).

The *internal barriers* preventing recognition of the cold fusion phenomenon are in the society of researchers and in researchers themselves. The most important one of them is the resistance to paradigm change in researchers in this field. As we show in Section 2, the CFP is a phenomenon explained by a new science in the interdisciplinary region between solid state physics and nuclear physics. To establish the science of this phenomenon, it is necessary to have a new paradigm for the experimental data in the CFP which are out of old paradigms. To accept a new paradigm, however, there are mental difficulties in scientists who have lived and therefore trapped in the old paradigms as M. Planck said in the sentence cited at the beginning of this Introduction. This is an internal barrier of researchers preventing development of the science of the CFP investigated in this paper.

## 1.4 Terminology of the Cold Fusion Phenomenon

It is convenient for readers to give a list of terminology characteristic to the cold fusion phenomenon which is a novel science where it is necessary to use some new concepts to describe the relations obtained by experiments..

Following is a list of technical terms characteristic in the field of the cold fusion phenomenon.

CF materials; Materials used in the CF research.

Cold fusion; [ICCF1 1990]

#### Cold fusion phenomena;

"Cold fusion phenomena" used by researchers at early days of investigation (e.g. [Fleischmann 1990]) seems to mean that events observed in the CF materials represent various phases of nuclear reactions between two deuterons. The main reactions had been assumed as those taken up by hot fusionists;

$D + D \rightarrow T (1 \text{ MeV}) + p (3 \text{ MeV}),$	(1-1)
$D + D \rightarrow {}^{3}\text{He} (0.18 \text{ MeV}) + n (2.5 \text{ MeV}),$	(1-2)
$D + D \rightarrow {}^{4}\text{He} (0.08 \text{ MeV}) + \gamma (23.8 \text{ MeV}).$	(1-3)

The complex experimental results piled up in a few years after 1989 obtained in deuterium and protium systems have shown that the nature of the nuclear reactions occurring in the CF materials is not so simple.

## Cold fusion phenomenon; [DOE Workshop, Kozima 1998]

According to the recognition of the complex nature of the nuclear reactions occurring in the CF materials, it has been proposed an idea to take the whole events observed there as a single entity with various faces. The entity has been named "the cold fusion phenomenon" (CFP). The "cold fusion phenomena" used by researchers in early days such as Fleischmann, Li and others correspond to the "events in the CFP" in this terminology. Many examples of the cold fusion phenomenon or the CFP are found in papers and books by H. K. [Kozima 1994, 1998, 2006, 2016a, 2017b]

It should be noticed that the *science of the cold fusion phenomenon* is a part of the science of nuclear interactions in solids which is properly called as the *solid state-nuclear physics, condensed matter nuclear science.*"

## Solid state-nuclear physics or Condensed matter nuclear science.

These two names represent the science or physics of events or phenomena occurring in solids induced by the interactions between nuclei and atoms. The physics of events induced by the interactions between nuclei are called as *nuclear physics* and that of

events induced by the interactions between atoms and molecules are called *solid state physics* or *condensed matter physics*, as well known. The name '*cold fusion phenomenon*' is used to extract 'the events induced by nuclear reactions' from other events without nuclear reactions such as the neutron diffraction by a crystal lattice even if it is induced by the interaction of neutrons and nuclei in solids without nuclear reaction.

## **1.5 Outline of This Paper**

In this paper, as shown in the Contents next to the Abstract, we give a brief survey of the science of the cold fusion phenomenon (the CFP) in Section 2 viewed from a phenomenological point of view using the TNCF model (the trapped neutron catalyzed fusion model), proposed at ICCF4 in 1994 and developed in these more than 20 years. The main structure of the CF research in the modern society had been determined in the first 15 years from 1989 when the first sign of the CFP was discovered, we use mainly materials obtained in this period for discussions in Section 2. Then, the sociology of the cold fusion phenomenon (CFP) is investigated in Section 3 using the framework developed by R.K. Merton [Merton 1975]. Details of materials for the discussions developed in Sections 2 and 3 are given Appendices.

## 2. Discovery of the Cold Fusion Phenomenon

"The man of science must work with method. Science is built up of facts, as a house is built of stones; but an accumulation of facts is no more a science than a heap of stones is a house. Most important of all, the man of science must exhibit foresight." [Poincaré 1902]

## 2.1 Discovery of a New Field of Science in the Post-industrial Society

As explained briefly in Prologue, the social situation on the end of 20<sup>th</sup> century gave an opportunity to recollect an old phantasy dreamed by F. Paneth in 1926 [Paneth 1926, 1928]. The social environment after 60 years, however, changed drastically giving a complex drama to the new discovery explained by G. Taubes extensively but onesidedly in his book published in 1993 [Taubes 1993].

In the course of the short history from the declaration of the discovery in March, 1989 by Fleischmann et al. [Fleischmann 1989] to the condemnation on the end of 1989 by the DOE report [DOE 1989], there appeared characteristics of the paradigm change discussed by T.S. Kuhn in his book *The Structure of Science Revolutions* [Kuhn 1962].

In this Section, we present the characteristic feature of the discovery of the CFP to give the essential problem of the science in the modern society through the history of the CFP. The chronological details of the history are replicated in Appendix D using the sentences from G. Taubes [Taubes 1993].

The discovery of the CFP was surveyed and explained phenomenologically in an author's book published in 1998 for the first time [Kozima 1998]. In this book, the author investigated whole events observed not only in deuterium systems but also in protium systems thus opened a way to the science of this complicated phenomenon characterized by nuclear reactions in CF materials in the interdisciplinary field between solid state physics and nuclear physics.

However, the history of the research on the CFP has been strongly influenced and biased by the researches on the controlled thermonuclear fusion of deuterons pursued earnestly worldwide since 1950s (cf. Appendix C). The history of the controversy between pros and cons to the CFP vividly shows the influence and is depicted briefly in Appendix D.

## 2.2 Science of the Cold Fusion Phenomenon

It is extremely important to realize the difference of facts and truth in daily life and in science. We want to give a short comment on this problem.

## Facts and Truth

First of all, it is necessary to distinguish *truth* from *facts* especially in science. This is a plain common sense but sometimes or very often confused in minds biased by interests other than science.

The brief history of the cold fusion phenomenon depicted in Appendix D shows the reason why the communication between pros and cons has been not fruitful. The key point is that the discussion is confined in the problem if it is possible to induce the d-d fusion reactions in solids at near room temperature without any acceleration mechanisms. The essence of experimental facts has been cast away but details of experiments in contrast to the conventional knowledge of solid state physics and nuclear physics were taken up exaggeratedly. In this sub-section, we take up fundamental problems in the history of the CFP to clarify the essence of the nuclear physics revealed hitherto in the experimental facts of the CFP.

#### 2.2.1 Two ways of the Approach to Unknown Problems and Experimental Facts

It is useful to contrast two useful ways of scientific approaches to investigate unknown problems.

1. Struggle in the Existing Paradigm: The first is the style used to extend the method confirmed its usefulness in a field up to the limit of the field as far as possible. The works by J. Schwinger [Schwinger 1994] and P. Hagelstein et al. [Hagelstein 2004] are typical examples in this group.

2. Look for a New Paradigm outside the Existing Paradigm: The second is contrasted to the first in leaving the existing paradigm and looking for a new one appropriated for the newly found facts. The phenomenological model proposed by us is almost only one example in this group [Kozima 1998, 2006, 1016, 2017a].

The difference in these two styles is based in the evaluation of the facts; the first considers the facts in and the second outside the existing paradigm.

Thus, the experimental facts are definitely important to investigate the CFP and tabulated in Table 2.1. They had been classified into two genres according to the relation to nuclear reactions; direct and indirect as tabulated in Table 3.2 [Kozima 2006 (Table 2.1)].

Mother C, Pd,	Ti, Ni, KCl + LiCl, ReBa <sub>2</sub> Cu <sub>3</sub> O <sub>7</sub> , Na <sub>x</sub> WO <sub>3</sub> , KD <sub>2</sub> PO <sub>4</sub> ,
solids TGS, S	SrCe <sub>a</sub> Y <sub>b</sub> NB <sub>c</sub> O <sub>d</sub> , Hydrogen graphite (HC <sub>6</sub> ), XLPE (CH <sub>2</sub> ) <sub>n</sub> ,
Agents n, d, p,	<sup>6</sup> <sub>3</sub> Li, <sup>10</sup> <sub>3</sub> B, <sup>39</sup> <sub>19</sub> K, <sup>85</sup> <sub>37</sub> Rb, <sup>87</sup> <sub>37</sub> Rb、 (ion beam, O)
<b>Experimental</b> Electro	plysis, Gas discharge, gas contact, (ion beam irradiation)
methods	
Direct Gamm	a ray spectrum $\gamma$ ( $\varepsilon$ ), neutron energy spectrum $n(\varepsilon)$ ,
<b>Evidences</b> Space	distribution of NT products NT( <i>r</i> ),
decrea	ase of decay constants, lowering of fission threshold
energy	
Indirect Excess	s energy $Q$ , number of neutrons $N_n$ , amount of tritium
<b>Evidences</b> atom <i>I</i>	Nt, helium-4 atom N <sub>He4</sub> , NT (NT <sub>D</sub> , NT <sub>F</sub> , NT <sub>A</sub> ), X-ray
spectru	$\operatorname{Im} X(\varepsilon)$
Cumulative NT( <i>r</i> ),	amount of tritium atom $N_{\rm t}$ , helium-4 $N_{\rm He4}$ ,
Observables	
<b>Dissipative</b> Excess	s energy $Q$ , neutron energy spectrum $n(\varepsilon)$ , number of
<b>Observables</b> neutro	ns $N_{\rm r}$ Gamma ray spectrum $\gamma$ (g) X-ray spectrum X(g)

Table 2.1 System and Obtained Evidences of CFP. Mother solids, agents, experimental methods, direct and indirect evidences of the cold fusion phenomenon. Q and NT express excess heat and the nuclear transmutation, respectively. Direct evidences of nuclear reactions in CFP are Energy ( $\varepsilon$ ) and position (r) dependences of reaction products NT( $\varepsilon$ , r), decrease of decay constants of radiative nuclides, decrease of fission threshold energy of compound nuclei [Kozima 2006 (Table 2.1)].

#### 2.2.2 Three Laws in the CFP

It has been found that there exist three empirical laws (or regularities) in experimental facts obtained in the CFP [Kozima 2012]. They are tabulated as follows;

1. The First Law; Stability Effect for Nuclear Transmutation Products

2. The Second Law; Inverse-Power Dependence of Frequency on Intensity of Excess Heat Production

3. The Third Law; Bifurcation of Intensity of Events (Neutron Emission and Excess Heat Production) in Time

These empirical laws suggest that the CFP is a phenomenon classified as complexity investigated in nonlinear dynamics in the several tens of years. The controversial problem of the reproducibility of events in the CFP should be understood by the qualitative reproducibility destined by the complexity at most.

#### 2.2.3 Characteristics of the Cold Fusion Phenomenon (CFP)

It is easy to understand the characteristics of the CFP from the facts tabulated in Table

2.1 and the three laws found phenomenologically.

They are written down as follows with relations to the controversial reproducibility;

- (a) Subtlety or difficulty in realization of the CF state (the state where occurs the cold fusion phenomenon) in CF materials. → irreproducibility
- (b) Fragility of the CF state due to the products (heat and radiation) of nuclear reactions in the CFP. →qualitative reproducibility
- (c) Difficulty of precision measurements of the products of the CFP.  $\rightarrow$  qualitative reproducibility

#### Science of the Cold Fusion Phenomenon (CFP)

We may be able to consider that a first step to the science of the CFP is settled when the facts and the characteristics summarized above are explained at least qualitatively [Kozima 2006, 2016]. The theoretical investigation given by our phenomenological approach has given following *necessary conditions* for the CFP in CF materials (materials where occur nuclear reactions);

## Necessary conditions for the CFP.

(1) Absorption of protium/deuterium into the material (at least in the surface regions thicker than at least about 200 A = 20 nm) with a density as high as that of the host element. It is said that the necessary average density of the hydrogen isotopes should be

larger than a minimum value, given for instance by the relative composition D/Pd > 0.7 in the case of PdD<sub>x</sub> [McKubre 1993].

(2) Formation of the superlattice composed of a sublattice of the host nuclei at the lattice points and another sublattice of protons/deuterons at the interstitial sites (at least in localized regions of the material). The self-organization might be relevant to formation of this superlattice [Kozima 2013].

(3) Existence of lattice nuclei with neutrons at evaporation levels (where the exotic nuclei are favorable) [Kozima 2014 (Sec. 2.7)].

(4) Existence of non-localized proton/deuteron wavefunctions in the material. It is probable the higher the diffusivity of protons/deuterons, the larger the extension of their wavefunctions, from experimental data in solid state physics [Kozima 2009].

(5) Realization of the super-nuclear interaction between lattice nuclei (mediated by interstitial protons/deuterons) [Kozima 2006 (Sec. 3.7.2), 2016 (Section 3.9), 2017a (Sec. 5.2.3)].

(6) Formation of the neutron energy bands by the coupled neutrons with the supernuclear interaction [Kozima 2006, 2017a],

(7) Supply of neutrons in one (valence band) of the formed neutron bands (by excitation from lower energy levels or by supply from outside) [Kozima 1998 (Sec. 8.3), Kozima 2006].

(8) Accumulation of neutrons at boundary/surface region of the CF material to form the CF-matter [Kozima 2006 (Sec. 3.7.2.3)].

(9) Interaction of neutrons in the neutron bands and CF-matter with disordered nuclei [Kozima 2017a (Sec. 5.2)]. The neutrons in the neutron bands do not interact with lattice nuclei as its nature but interact with nuclei in such disordered sites as displaced positions in the lattice and at boundary regions. This is the cause of localization of nuclear reactions at surface/boundary regions which have observed frequently in experiments.

## **2.2.3 The First Stumbling Stone – The Protium System**

As the controversy between pros and cons, partially reproduced in Appendix D, had been fought on the possibility of d-d fusion reactions in CF materials, the cold fusion reactions in protium systems, therefore, have been alienated for a long time irrespective to its importance. This alienation influenced to the biased progress of the investigation of the CFP very much. If the experimental data obtained in deuterium and protium systems are considered equivalently, the investigation of the science of the CFP may have had more profound expansion. The experimental data on protium systems had appeared after 1989 in *Fusion Technology* (*FT*) and *Proceedings of ICCF2*. The numbers of papers on the deuterium and protium systems published in *FT* from 1991 to 1995 are shown in Table 2.1. Individual papers on the protium systems are listed in Appendix F2. G.H. Miley's comments on the Cold Fusion papers in *FT* are cited in Appendix F3 for their importance in the history of CFP research.

FTVol.	16	17	18	19	20	21	22	23	24	25	26	26+	27	Proc. ICCF *
D-system	23	15	27	20	15	11	15	8	13	9	6	65	5	83
H-system	0	1	1	0	0	0	1	2	2	0	2	9	0	29

Table 2.1 Number of papers on deuterium (D) and protium (H) systems appeared in *Fusion Technology* (*FT*) from Vol. 16 (1989) to Vol. 27 (1995) including a special issue Vol. 26, No. 4T-2 for *Proc. ICCF4* (1994) [ICCF4a] expressed as Vol. 26<sup>+</sup>. Individual papers (titles, authors) on protium systems are given in Appendix F2.

\**Proceedings of ICCF4* in 4 volumes [ICCF4b] is published simultaneously with the *Fusion Technology* **26**, **4T-2** (1994) including all papers presented at the Conference. The differences of numbers in the columns Vol. 26<sup>+</sup> and *Proc. ICCF4<sup>\*</sup>* show that papers on H-systems are mainly published in the latter.

After ICCF3 (1993) where four papers on the CFP in protium systems were presented, there appeared gradually papers showing the CFP with protium systems. However, many researchers in this field had adhered to deuterium systems. And therefore, opponents against the CFP concentrated their attack to the experimental data and theoretical works on the deuterium systems neglecting those on protium systems. The situation has been described in Sec. 3.2.1.

As explained in Secs, 3.2.1 and 3.2.2, it is wastes of time and energy to struggle with a phantasy without roots. Taking up only a part of events in the CFP is not a way to the science of the phenomenon. It is necessary to have a common ground to look for scientific explanation of the experimental data as a whole obtained in the CFP: Truth is grounded in facts.

## 2.2.4 The Second Stumbling Stone – The Reproducibility

As described in Section 2.2.2, the experimental facts suggested that the CFP is a phenomenon to be classified in complexity, studied extensively in nonlinear dynamics.

It has been a controversial problem, however, if the experimental results in the CFP are reproducible or not. It is a nonsense question if we notice that there are very many instances where the reproducibility has its meaning only in a statistical sense in the events in quantum physics containing nuclear physics. A simple example of the qualitative reproducibility is the alpha decay of a radioactive nuclide, e.g. radium <sup>226</sup><sub>88</sub>Ra to <sup>222</sup><sub>86</sub>Rn, we have taken up often in our papers and books (e.g. [Kozima 2006 (Sec. 2.14)]). We know a statistical number of decaying nuclei but not which ones will decay next.

Thus, the controversial problem of the reproducibility of events in the CFP is explained as a natural result of complexity in the process of formation of CF materials in the dynamical conditions used in the experiments [Kozima 2006, 2010, 2012, 2016 (Section 3.8)].

# 3. The Sociology of the Cold Fusion Research

"The sociology of knowledge came into being with the signal hypothesis that even truths were to be held socially accountable, were to be related to the historical society in which they emerged." [Merton 1975 (p. 11)]

As we have given a brief survey on the history of researches on the CFP in Section 2, its possible influences and the novel features on the modern world inevitably invited confusion not only in science but also in science policy. After almost 30 years from the discovery of a part of the CFP by Fleischmann et al. [Fleischmann 1989], the situation around the CFP has not changed much. This situation is asking us sociological analyses on it.

We give here an essay to investigate the history of the CFP using the concepts developed by sociologists which are summarized in Appendix A.

## 3.1 Sociology of the Cold Fusion Phenomenon

The Sociology of the Cold Fusion Research (the Research on the Cold Fusion Phenomenon) is a part of the Sociology of Science investigated for about a century by sociologists especially by a group led by R.K. Merton [Merton 1975] (cf. Appendix A).

The general discussion of the sociology of science will give a firm standpoint to analyze the development of the CF research in this society and give a hint to integrate it into modern science as an inevitable part of it. We investigate the sociology of the CFP in this section using the concepts developed by R.K. Merton et al. summarized in Appendix A.

## 3.1.1 The Sociology of Knowledge in 21st Century

The sentences cited in Appendix A1 from Merton's book are mainly for social scientists but applicable also to natural scientists. Especially, the following sentence from the Merton's paper on the sociology of knowledge is suggestive for us [Merton 1975 (p. 11)].

"The 'Copernican revolution' in this area of inquiry consisted in the hypothesis that not only error or illusion or unauthenticated belief but also the discovery of truth was socially (historically) conditioned. - - The sociology of knowledge came into being with the signal hypothesis that even truths were to be held socially accountable, were to be related to the historical society in which they emerged." [Merton 1975 (p. 11)]

We have to notice the social situation in 21<sup>st</sup> century where genuine reality has been overwhelmed by virtual reality in society as especially exhibited in political world. The development of information technology (IT) e.g. such as social network service (SNS), has changed human relations in the society. The Merton's sentence cited above should be considered seriously in consideration of scientific truth.

## 3.1.2 The Sociology of Scientific Knowledge in 21st Century,

The conglutination of technology and science has largely advanced in 20<sup>th</sup> century and appearance of huge industries has changed the character of the science in modern society. It is already noticed by R.K. Merton that the relation of science and its environment changes when the society changes its character (cf. Appendix A2 [Merton 1975 (p. 176)]). Details of the interaction between the cold fusion research and the controlled nuclear fusion casting a shadow on the funding problem have been given in Subsection 3.2 and Appendix D.

The sentences cited in Appendix A2 from Merton's book depict a traditional concept of science extracted from history of science since 16<sup>th</sup> century until the beginning of 20<sup>th</sup> which should be considered obsolete now. The close connection between science and military technique, however, had been noticed early in 1935 [Merton 1975 (Section 9)]. Several sentences are cited below from Merton's paper.

"The foci of scientific interest are determined by social forces as well as by the immanent development of science. We must therefore examine extrascientific influences in order to comprehend more fully the reasons why scientists have applied themselves to one field of investigation rather than another. With the view of tracing such a connection between science and society, I shall indicate the ways in which military exigencies have encouraged the growth of one branch of science, especially in seventeenth century England." [Merton 1975 (p. 204)]

#### 3.1.3 The Normative Structure of Science,

According to the strong connection of science and society and to the huge effects of science on society, science has become an important brick of modern society. Following sentence by Merton explains the situation compactly.

"Incipient and actual attacks upon the integrity of science have led scientists to recognize their dependence on particular types of social structure. Manifestos and pronouncements by associations of scientists are devoted to the relations of science and society. An institution under attack must reexamine its foundations, restate its objectives, seek out its rationale. Crisis invites self-appraisal. Now that they have been confronted with challenges to their way of life, scientists have been jarred into a state of acute self-consciousness: consciousness of self as an integral element of society with corresponding obligations and interests.<sup>1</sup>

1. Since this was written in 1942, it is evident that the explosion at Hiroshima has jarred many more scientists into an awareness of the social consequences of their work." [Merton 1975 (pp. 267 - 268)]

Several explanations on the normative structure of science by Merton are cited in Appendix A3.

#### 3.1.4 The Reward System of Science in 21st Century,

The change of structure of science has given a large alteration in the reward system of science. Especially, the cold fusion phenomenon has not been established its existence in society and therefore many researchers in this field could not find out stable positions for their research. It is necessary to establish the science of the cold fusion phenomenon first to consider the reward system of this field comparable to that in the established fields of science.

Some considerations on the reward system of science discussed by social scientists are given in Appendix A4.

#### 3.1.5 The Process of Evaluation in Science in Modern Industrial Society.

The process of evaluation of works on the CFP is in similar situation to the problem of the reward system due to the unestablished situation of this research field. Several sentences on the process of evaluation in science by Merton are cited in Appendix A5.

There are a few periodicals accepting papers on the cold fusion phenomenon at present as surveyed in Appendix F.

## **3.2 Funding Organizations and Their Influence on the Research**

There have been very many examples showing shortcomings of the Big Science phenomenon in modern society. The episodes described in books by J.R. Huizenga [Huizenga 1992] and G. Taubes [Taubes 1993] had shown typical phases of the shortcomings with many unscientific biased conclusions discarding pearls of truth in the facts.

In relation to the problems considered in Subsections 3.1.4 and 3.1.5, the funding systems in modern technological world are one of fundamentally important organizations in a society. A feature of the competition for funds has appeared in the history of the CFP as described in Appendix E. We cite several sentences from books by G. Taubes, J.R. Huizenga and D.R.O. Morrison below to know the facts. More examples are shown in Appendices E and H.

#### 3.2.1 Taubes describes the background of the competition between U-o-U and BYU

Martin Fleischmann and B. Stanley Pons in the University of Utah (U-o-U) vs. Steven Jones of Brigham Young University

"Once Stan Pons submitted his proposal to his benefactors at the Office, of Naval Research, the cold fusion affair took on an aura of inevitability. Although ONR had a reputation for funding speculative research projects, either Pons or Robert Nowak, his funding officer, decided that ONR was not the right place for cold fusion. Pons told Hugo Rossi that he feared the Department of Defense, of which ONR was a part, might classify cold fusion. They might realize the potential military uses of an invention that, if it wasn't a hydrogen bomb itself, would still produce tritium, a necessary and valuable component of hydrogen bombs.

Since Pons preferred to think of cold fusion as an energy source, not a weapon, he sent the proposal to his friend Jerry Smith, a funding agent in the Department of Energy's physical chemistry program. At one time Smith had been program manager for Pons at ONR, and his name appears as a coauthor on several of Pons's journal articles. Smith, too, felt that cold fusion was inappropriate for his office and suggested that Pons submit his proposal to the Office of Advanced Energy Projects at DOE run by an administrator named Ryszard Gajewski (pronounced Richard Guy-EV-ski)." [Taubes 1993 (Chapter 2, The Competition, p. 19)]

"During his tenure Gajewski had managed to support two projects for six years or longer. The first was an X-ray laser project for hot fusion, which was being pursued by a Princeton physicist.<sup>2</sup> The second was known as muon-catalyzed fusion, the forerunner of what has since been called cold fusion. Muon-catalyzed fusion was one of Gajewski's visions. He equated it to his own background. "Like an immigrant's child Gajewski once wrote of muon-catalyzed fusion, "in order to succeed in a big way we must prove that we have something clearly superior to offer, something the society simply cannot ignore.

The point man for this program was Steven Jones of Brigham Young University. After the announcement on March 23rd, when the press discovered that Jones had also been studying cold fusion and were trying to establish the nature of the relationship between Pons, Fleischmann, and Jones, Gajewski would tell reporters that if he had to choose three people to trust in the world, Jones would be one of the three. Left unsaid, of course, was that neither of the other two would be Pons or Fleischman." [Taubes 1993 (p. 21)]

## 3.2.2 J.R. Huizenga and D.R.O. Morrison also described the situation;

"The First Annual Conference on Cold Fusion (ICCF1), sponsored by the National Cold Fusion Institute, was held in the University Park Hotel, a few yards away from the NCFI, on March 28 – 31, 1990." [Huizenga 1992 (p. 170)]

The National Cold Fusion Institute was established in the University of Utah in March, 1989 by the State of Utah expecting a financial support from the US Government.

"At present the greatest part of the funding for Cold Fusion comes from Japan; IMRA (supported basically by Toyota) has set up labs in France and Japan: and MITI (Ministry of International Trade and Industry, Japan) is starting to devote \$30 million over four years. The question is, why?" [Morrison II (No. 9, General Impressions on ICCF4)]

#### 3.2.3 Distortion of science caused by the funding system

To get financial supports from funding organizations, researchers emphasize the possible applications of the newly found phenomenon rather than engage in scientific researches of the truth behind the facts. In this process, they select useful facts discarding facts seemingly unuseful for the application in their minds.

The CFP in protium systems had been the typical example discarded for a long period due to its irrelevance to the d-d reactions supposed to be valuable for an energy source without much trouble of supply and hazardous waste.

## **3.3 Paradigm Revolution**

It is difficult to leave a world where we are accustomed and living easily without

much care about environmental affairs. However, we have to reconstruct our world to accept new facts contradicting with the established frame of the old world. When it occurs in scientific world, the new world is called a new paradigm by T. Kuhn [Kuhn 1962]. The controversy between pros and cons of the CFP depicted in Appendix D using their sentences is seen as a battle for their paradigms; however their paradigms are very similar confined to the d-d fusion reactions to explain the new facts.

It is necessary to have a new paradigm to give an unified explanation of various facts observed in deuterium and protium systems as introduced in Section 2. We give a brief explanation of the situation explained above.

#### 3.3.1 Neglect of truth in new facts by biased view persistent to old prejudice.

Corresponding to the biased view in the researchers of the CFP, scientists in other relevant research fields, such as nuclear physics and solid-state physics, criticized the efforts by the CF researchers missing in this case the facts showing novel features of solid-state nuclear physics both in protium and deuterium systems too eager to criticize possibilities of the *d*-*d* fusion reactions in solids at near room temperature.

#### 3.3.2 Logics of Opponents to deny the Facts of the CFP

We can trace logics of opponents to deny the facts of the CFP only criticizing the experimental and theoretical demands of CF researchers to show possibilities of the d-d fusion reactions.

### 3.3.2a J.R. Huizenga [Huizenga 1992]

J.R. Huizenga was a co-chairman of the U.S. Department of Energy's Cold Fusion Panel in 1989. After the publication of "DOE Report 1989" (*A Report of the Energy Research Advisory Board to the United States Department of Energy*, DOE/S-0073 DE90 005611 (1989)) [DOE 1989], he published his book on the cold fusion using his knowledge on nuclear physics and materials in the "Report" [Huizenga 1992]. Naturally, he had no motivation to find out truth behind the experimental facts, mixed naturally with correct and incorrect ones.

"The process of fusion of two deuterium atoms was first studied by Ernest Rutherford and his colleagues at the Cavendish Laboratory in Cambridge (*Nature*, **133**, p. 413, 1934; *Proc. Roy. Soc.* **A148**, p. 623, 1934). During the ensuing half century, nuclear fusion of deuterium has been studied intensively and it is now a relatively well understood process. If fusion between deuterium atoms were indeed occurring at room temperature, there is essentially no doubt what the products would be. They would be the same products that are observed for reactions between two low-energy deuterium nuclei, where fusion is known to proceed in three channels:

[Huizenga 1992 (p. 6)].

"If the energy burst were truly nuclear in origin, equivalent amounts of fusion products must have been released. Without this confirming evidence of commensurate yields of fusion products, Fleischmann and Pons' claim did not convince most scientists that a nuclear process had occurred." [Huizenga 1992 (p. 70)]

"These researchers all reported different amounts of excess heat and different criteria for success. In attempts to produce excess heat, Bockris emphasized that there was "considerable ^reproducibility" while Pons stated that he could now "... reproduce it more than 90% of the time" (Salt Lake Tribune, May 10, 1989). Although these two advocates of cold fusion disagreed sharply on the question of reproducibility, they, along with others, claimed success in producing excess heat." [Huizenga 1992 (p. 71)]

"Fleischmann and Pons' gamble to go public with their announcement of watts of 'excess power' from nuclear fusion at room temperature, before they had any solid evidence of fusion products, is the scientific fiasco of the century. The chasm between Fleischmann and Pons' claim of 'excess heat' and the upper limit on the intensities of the associated fusion products continues to be many orders of magnitude! Room temperature nuclear fusion without commensurate amounts of fusion products is a delusion and qualifies as pathological science defined as 'the science of things that aren't so'." [Huizenga 1992 (p. 214)]

## 3.3.2b D.R.O. Morrison [Morrison I, II]

D.R.O. Morrison has enthusiastically pursued the truth behind experimental facts observed by CF researchers and by critics from his point of view confined in the framework restricted only in *d*-*d* fusion reactions. His publications on the CFP have been from 1989 to 2000. He attended all ICCF (the International Conferences on the Cold Fusion) from the first (1990) to 8<sup>th</sup> (2000) held almost annually; the first was held on 28 to 31, March 1990 in University of Utah, USA and the 8<sup>th</sup> on 21 to 26, May 2000

in Lerici (La Spezia), Italy.

Morrison has attacked the experimental data inconsistent with the d-d fusion reactions that have been observed in free space and fully investigated in nuclear physics. We have heard every time his critical questions at ICCFs "That is in contradiction with the common sense of the nuclear physics."

Some examples of his criticism are cited below from his *Cold Fusion News* No. 1 (1989) – 10 (1990) cited as [Morrison I] and *Cod Fusion Updates* No. 1 (1990) – No. 13 (2000) cited as [Morrison II].

"Finally society must make a judgement on this subject that has excited such great interest and raised such wonderful hopes. Although most, though not all, workers in the field realise that cold fusion will never be of any practical use for power production, this is still not the popular perception of it. Most people still hope that maybe something will come of it. There seems to be a tendency amongst some to let the bad news leak out slowly. This is often good politics and PR, but not all scientists feel this way - they want to avoid people getting into false positions because of lack of knowledge of the facts.

There is a second question which is of purely academic interest - does there exist cold fusion at some very low level (a billionth or a trillionth or a billion-billionth of a Watt). Clearly the Fleischmann and Pons level is excluded by the data, but some hope it might occur at the BYU level, but again there are good experiments which find no counts at much lower levels. So one cannot avoid asking the difficult question of whether all the positive results are mistaken. Scientists do not want this to be correct, but can one exclude it?" [Morrison I (No. 14, Conclusions)]

"A major topic is the experiment of Mike Salamon et al. who did not observe any neutrons or gammas when positioned below four of Pons's cells. They have new strong evidence against any fusion product emission at a time when Prof. Pons said one of his cells gave heat. This was published by Nature during the conference." [Morrison I [No. 22 (Summary)]

"From my studies of Pathological Science, it is possible to make some guesses. - - -

Those groups that have found positive results will not suddenly find errors and withdraw their papers. Rather they will publish work enroute but then will stop publishing. They will say they have never found anything wrong with their experiments." [Morrison I (No. 23. What is the Future)]

"Scientists are educated to study and believe experimental results. And when young they do. Their culture makes them very trusting and there results an exceptionally good working atmosphere. However magicians such as Randi say that scientists, especially physicists, are the easiest people to deceive. This is because they virtually never encounter fraud in their work and rarely hear lies.

The consequence is that when there are good grounds for suspecting fraud, scientists generally do not know how to deal with it." [Morrison II (No. 1. Problems of Dealing with Fraud)]

"The major problem of Cold Fusion - the disagreement of the amount of heat claimed and the corresponding expected number of nuclear products - still exists. Surveys still indicate other major problems, particularly reproducibility." [Morrison II (No. 5. Summary of Report on ICCF2)]

"These other claims included several groups saying that they observed excess heat with normal water, i.e. light hydrogen - this is in contradiction with Fleischmann and Pons and others who said it happens only with heavy hydrogen (deuterium) and the proof that it is nuclear fusion is that it is NOT observed with light hydrogen." [Morrison II (No. 7. Summary for ICCF3 Report)]

D.R.O. Morrison finally became more tolerant to variety of approaches to the science of the CFP as citing many theoretical works in his review on the ICCF8 but could not recognize the truth hidden behind complex, sometimes contradicting experimental data; one example of the former and his comment showing the latter are cited below from the final *Cold Fusion Updates* **No. 13**, 2000 published after ICCF8.

"12. H. Kozima (abs. 044, 045, 046) Trapped Neutron Catalyzed Fusion, TNCF model. Energy band of neutrons interacts coherently with lattice nuclei.

e.g.

n +  $_{46}$ Pd ---->  $_{13}$ Al +  $_{33}$ As or ---->  $_{26}$ Fe +  $_{20}$ Ca." [Morrison II (No. 13 Appendix 2, Theories at ICCF-8)]

<In this citation, Morrison has shown his correct understanding of the TNCF model as his explanation of the lack of radiation in the nuclear reaction as "*Energy band of neutrons interacts coherently with lattice nuclei*."> (Italicized at citation).

## "11. CONCLUSIONS

I have often looked at experiments which gave results that appeared to violate the laws of Nature which had been established by previous work. Later these experiments turned

out to be false, but I have often found it very difficult to see just where the error was. But the fact that I had not detected the flaw, did not mean that the experiment was correct and that the laws of Nature had been violated.

Rather I feel the same as being at a circus watching a magician. Normally he and I know that the laws of nature are being obeyed but there is a trick which is hard to spot. At trick one, I may spot the trick and am happy that there is no problem with the laws of Nature - similarly with trick number two. But suppose at trick three, I do not see how the magic is performed. The magician may say "I won, I tricked you" and it is left unsaid that the laws of nature have not been violated. But suppose the magician says "You did not see anything wrong with my demonstration, therefore it is true. See, I have supernatural powers. The old laws of Nature have been replaced by new laws". And if I protest, I am told that I have a closed mind, am an establishment figure, and do not face up to the happening performed in front of me. But almost all magicians admit that it is all trickery and the laws of Nature are not threatened.

So if someone comes along and says, "Look - excess heat - do you see anything wrong?", then I feel as if I am at the circus, and although I do not immediately see anything wrong, I am reluctant to give up well-established laws of Nature unless the proof is very strong. Here reports on cold fusion happenings are described, especially in the summary talks by True Believers in cold fusion in their words, and then some clues as to possible explanations are offered. How many Elvis sightings constitute a proof?" [Morrison II (No. 13. Conclusions)].

### **3.3.2c G. Taubes** [Taubes 1993]

G. Taubes wrote his book on the episode of cold fusion using his interviews with over 260 scientists, administrators, and journalists within the limits of his knowledge on physics. He had no intension to find out the truth behind the experimental facts and therefore just denied facts (mixed with correct and incorrect ones) obtained at that time comparing with his knowledge of nuclear reactions in nuclear physics.

"Nuclear reactions generate nuclear radiation. That is the nature of the beast. Even without calculating exactly how much neutron radiation should have been emitted by four watts of fusion power, one indication that Pons and Fleischmann had observed too little was their reasonably robust appearance.<sup>4</sup> The radiation emitted from this level of power generation should have been sufficiently malign for its effects to have been noticeable. In fact, the radiation would have killed the two chemists, not to mention seriously impaired the health of the students working nearby. But it hadn't. So where were the neutrons?

<sup>4</sup> Fleischmann, in fact, admitted at the press conference that they had observed only a billionth as much helium, tritium, and neutrons as they expected, but this hadn't appeared in the press reports." [Taubes 1993 (pp. 117 and 441)]

"He pointed out that the tritium was incompatible with the neutrons, which were incompatible with the heat, and suggested to a panel of what The Deseret News was now calling the fusion fraternity that one or several of these results had to be wrong. Would the people with the wrong results please raise their hands? Kellogg asked. He was ignored." [Taubes 1993 (p. 406)]

"Within six months of the announcement of cold fusion, its public life had deteriorated into a dismaying struggle against reality in which the believers explained the insipid state of their science with all manner of causes, none of which was as simple as the reality itself. Cold fusion—as defined by Stanley Pons and Martin Fleischmann, or Steve Jones, or as modified by John Bockris or Edmund Storms and Carol Talcott, or Bob Huggins-Stanford, or whomever—did not exist. It never had. There was at least as much empirical evidence, if not more, to support the existence of any number of pseudoscientific phenomena, from flying saucers to astrology." [Taubes 1993 (Epilogue pp. 425 - 426)]

"Of all the arguments spun forth in defense of cold fusion, the most often heard was that there must be something to it, otherwise the mainstream scientific community would not have responded so vehemently to the announcement of its discovery. What the champions of cold fusion never seemed to realize, however, or were incapable of acknowledging, was that the vehemence was aimed not at the science of cold fusion but at the method. Positive results in cold fusion were inevitably characterized by sloppy and amateurish experimental techniques. If these experiments, all hopelessly flawed, were given the credibility for which the proponents of cold fusion argued, then science itself would become an empty and meaningless endeavor. Once bad science was accepted as good enough, it could be used to prove the existence of anything, whether it existed or not." [Taubes 1993 (Epilogue p. 426)]

"What cold fusion had proven, nonetheless, was that the nonexistence of a phenomenon is by no means a fatal impediment to continued research. As long as financial support could be found, the research would continue. And that support might always be found so long as the researchers could obtain positive results. In fact, the few researchers still working in the field would have little incentive to acknowledge negative results as valid, because such recognition would only cut off their funds. It promised to be an endless loop." [Taubes 1993 (Epilogue p. 426)]

## **3.4 Miscellaneous**

In this section, we give miscellaneous problems related to the sociology of the cold fusion phenomenon.



3.4.1 Conferences where presented papers on the CFP (cf. Appendix G)

Fig. 4.1 Main journals and conferences where published and presented works on the CFP since 1989 to 2016 (cf. Appendix F).

## 3.4.2 Publication of Works (cf. Appendix F)

Journals which treated the CFP did not last long except a few cases (which are published by organizations) due to the lack of funds, internal competitions within researchers in this field and also by the aging of publishers (cf. Fig. 4.1 and Appendix F).

## 4. Conclusion

"Thus science develops an autonomous corpus of investigation which has its origin in strictly scientific, not utilitarian, considerations. It is these developments (which probably constitute the greater part of science) arising from the relative autonomy of scientific work that seem to have little or no connection with social forces." [Merton 1975 (p. 209)]

The science is a part of social activity and inevitably influenced by the social situation at the time.

In the industrial and capitalistic world at present, the science, especially the big science, is governed by the logic of capitalism which is wholly independent of scientific spirit caused by curiosity and based on facts.

The foundation of the science in a society is closely related to the atmosphere to evaluate reality above all. This is the factor taken up by Merton in *The Sociology of Knowledge* as a premise of *the Sociology of Science* [Merton 1975]. If the atmosphere to evaluate knowledge is lost, it is clear that the science loses its foundation and will wither and die. How changes the atmosphere of a society? It should be a fundamental problem of the sociology of science. We might be in the final stage of a society in which science has prospered for more than 400 years.

It seems that the most important fundamental cause of the confusion in the research of the CFP lasting more than 25 years is the persistence or adherence to the d-d fusion reactions for the nuclear reactions in CF materials. In the persistence to the d-dreactions, there certainly is expectation of financial merits obtained by the application of the phenomenon rather than scientific clarification of the mechanism of the CFP. This biased intention in the scientific research distorted the research work itself and also critics on the work lasting very long without any fruits.

However, as R.K. Merton said in the sentence cited at the top of this Section, the science of the cold fusion phenomenon has been developing steadily revealing a new feature of physics of neutrons in CF materials as a part of solid state-nuclear physics (e.g. [Kozima 2006, 2016, 2017a]).

Application of this phenomenon will be considered apparently when the whole image of the cold fusion phenomenon is brought to light. We are able to expect at least applications of following effects confirmed in the CFP occurring in boundary regions of CF materials; nuclear transmutations of elements, acceleration of decays of radioactive nuclei, and production of large excess energy liberated in nuclear reactions in CF materials.

## **Epilogue – At the Dusk of a Civilization**

"Truth. It's grounded in Facts." – New York Times (April 1, 2017).

It is considered that there had been two periods of fundamental change in the history of science hitherto as mentioned by B. Barnes;

"--- drawing attention to the two most important periods of fundamental change in the history of science – *the cultural revolution of the sixteenth and seventeenth centuries*, and *the social reorganization of the nineteenth*." [Barnes 1972 (Introduction, p. 11)] (Italicized at citation).

We have to add the third period to the two noticed above; *the social revolution of the twenty first century*. Trend of the world seems inclining toward virtual reality from true or genuine reality.

The scientific discovery at the far limits of human knowledge such as uncertainty principle ([Heisenberg 1927]), incompleteness theorems ([Goedel 1931]) and furthermore the complexity (e.g. [Nikolis 1989]) might be influencing the trend in deep bottom of our culture.

The personal inability to interfere the movements of world affairs such as the development of powerful warfare, the global warming, and the population explosion may be causing apathy of people to everything despite of, or rather due to the developments of information technology and biotechnology. As early as the year 1941, E. Fromm noticed the apathy toward reality due to inability to interfere the flow of social affairs [Fromm 1941].

The tendency seems to be prevailing worldwide in the beginning of 21<sup>st</sup> century. The social affairs appeared in recent years on the end of 2010s may be a signal of the apathy in psychological sphere of human being all over the world. The science is a part of human activities belonging to the Reason. In the three major characteristics of human beings, Reason, Feeling and Intention, the Reason may be the last obtained or grown up in our brain. This means the Reason is most insensitive to the change of environment. The changes of social atmosphere appeared in the Feeling and the Intention worldwide in recent years may erode the totality of human beings resulting in such a tragedy we suffered in 1930s in the previous century.

## Acknowledgements

The author would like to express his heartfelt thanks to Dr. Tadayoshi Ohmori\* and
Dr. Hiroshi Yamada<sup>\*\*</sup> for their helps in the preparation of Appendices F - H to find out papers on the CFP published in *Fusion Technology* and also for their encouragement throughout this work.

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## Appendices

Appendix A Sociology of Science

Appendix B Controlled thermonuclear Fusion

- Appendix C ITER (International Thermonuclear Experimental Reactor) Story
- **Appendix D** Controversy between Pros and Cons about *d-d* Fusion Reactions in CF Materials

Appendix E Competition for Funds

- **Appendix F** Main Journals where published papers on the Cold Fusion Phenomenon after 1989.
- **Appendix G** Main International Conferences on the Cold Fusion Phenomenon until 2010
- Appendix H Funding Organizations for the CFP

#### **Appendix A – Sociology of Science**

The Sociology of Science developed by R.K. Merton and others is a part of the Sociology of Knowledge. The works on the Sociology of Science are divided accordingly in five sections as follows [Merton 1973 (Introduction by N.W. Storer)]; (1) The Sociology of Knowledge, (2) The Sociology of Scientific Knowledge, (3) The Normative Structure of Science, (4) The Reward System of Science, and (5) The Process of Evaluation in Science. The each division includes works on science especially in 17<sup>th</sup> century England and in modern Europe and America and exhibit requisite for a science to exist in the society. These divisions show necessary conditions for the CFP to be a branch of science in the modern society. We therefore summarize investigations given in these works in this Appendix A.

"It was at this same time that Merton wrote his dissertation, *Science, Technology,* and Society in Seventeenth-Century England (begun in 1933 and completed two years later)." [Merton 1975 (Storer, p. xv)]

#### A1 Sociology of Knowledge

There are brief characterizations of the sociology of knowledge in the Prefatory Note to Chapter 1.

"--- social scientists have a normative obligation to assert their scholarly values against the short-range and self-interested objectives often found in research requests coming from policy makers. This suggests that the current discovery of such problems and perspectives in "the New Sociology" may rather be a reaffirmation under social conditions now more propitious to such views than before; in fact, the "cryptomnesia" (see the last paper in section 4 of this volume) suffered by successive generations of social scientists on this subject is itself a specific problem for the sociology of knowledge." [Merton 1975 (Storer, p. 6)]

This sentence is suggestive also to natural scientists at present.

"A program for relating philosophical conceptions of the sources of knowledge in society to the empirical investigation of specified problems is sketched out in the explicit "paradigm" laid out early in the essay." [Merton 1975 (Storer, p. 4)]

R.K. Merton formulates the paradigm for the sociology of knowledge in his paper published in 1945 just after the WWII.

"Paradigm for the Sociology of Knowledge [Merton 1975 (pp. 12-13)]

#### 1. Where is the existential basis of mental productions located?

a. social bases: social position, class, generation, occupational role, mode of production, group structures (university, bureaucracy, academies, sects, political parties), "historical situation," interests, society, ethnic affiliation, social mobility, power structure, social processes (competition, conflict, and so on).

b. cultural bases: values, ethos, climate of opinion, Volksgeist, Zeitgeist, type of culture, culture mentality, Weltanschauungen, and so on.

#### 2. What mental productions are being sociologically analyzed?

a. spheres of: moral beliefs, ideologies, ideas, the categories of thought, philosophy, religious beliefs, social norms, positive science, technology, and so on.

b. which aspects are analyzed: their selection (foci of attention), level of abstraction, presuppositions (what is taken as data and what as problematical), conceptual content, models of verification, objectives of intellectual activity, and so on.

#### 3. How are mental productions related to the existential basis?

a. causal or functional relations: determination, cause, correspondence, necessary

condition, conditioning, functional interdependence, interaction, dependence, and so on. b. symbolic or organismic or meaningful relations: consistency, harmony, coherence, unity, congruence, compatibility (and antonyms); expression, realization, symbolic expression, Strukturzusammenhang, structural identities, inner connection, stylistic analogies, logicomeaningful integration, identity of meaning, and so on.

c. ambiguous terms to designate relations: correspondence, reflection, bound up with, in close connection with, and so on.

# 4. Why related? Manifest and latent functions imputed to these existentially conditioned mental productions.

a. to maintain power, promote stability, orientation, exploitation, obscure actual social relationships, provide motivation, canalize behavior, divert criticism, deflect hostility, provide reassurance, control nature, coordinate social relationships, and so on.

5. When do the imputed relations of the existential base and knowledge obtain?

a. historicist theories (confined to particular societies or cultures).

b, general analytical theories." (Boldfaces added at citation)

#### A2 The Sociology of Scientific Knowledge

Scientific knowledge is always a part of knowledge in a society. The sociology of scientific knowledge follows naturally the sociology of knowledge [Merton 1975].

"--- how science comes to develop in the first place. This is followed by the correlative, more specific question: Once science comes culturally and institutionally established, what affects the rate and directions of its development?" (p. 139)

"--- the short-range ebb and flow of scientific attention can be attributed largely to causes internal to science (the sources of more long-range shifts are taken up later in the monograph, with the conclusion that they are much more affected by extrascientific changes). (p. 140)

"--- the autonomy-oriented character of scientific knowledge in its demonstration that aggregates of scientists will frequently pick up a topic because of its applied value and then transform it into a "pure" scientific problem that leads off in directions having less and less relevance to the originating occasion." (p. 141)

In the preface to the reprinted paper, *Science, Technology and Society in 17<sup>th</sup> Century England*, published in 1970, R.K. Merton noticed the historical characteristic of the relation between science and its environment as follows;

"It is now quite evident to me and I hope will become evident to its readers that this inquiry into the interdependence of science and other institutional spheres in seventeenth-century England neither adopts a factor theory nor supposes that the character of interchanges between institutional spheres that occurred in that period is much the same in other cultures and other times. Rather, it states in so many words that the nature and extent of these interchanges differ in various societies, depending on the state of their science and of their institutional systems of economy, politics, religion, military, and so on. This should not come as a strange idea. After all, the relations between science, economy, and government in England of the seventeenth century, when modern science and its technological offshoots were only in their beginning, differ palpably from their relations in the twentieth-century United States or Soviet Union, where science has long been institutionalized, where scientific research requires vast support, and where it has acquired new magnitudes of consequence for technologies of production and destruction. The recent highly publicized discovery of the industrialmilitary-scientific complex only brings to our notice tendencies toward the interdependence of science and other social institutions that have, to a degree, been present all along. That, at least, is the import of the chapters in this monograph that examine the relations of science and technology with economic development and military technique." [Merton 1975 (p. 176)]

#### A3 The Normative Structure of Science

The ethos of science is difficult problem in the conflict of values in modern society. The following sentence written by R.K. Merton in 1942 explains the difficulty briefly. "Science, like any other activity involving social collaboration, is subject to shifting fortunes. Difficult as the notion may appear to those reared in a culture that grants science a prominent if not a commanding place in the scheme of things, it is evident that science is not immune from attack, restraint, and repression. Writing a little while ago, Veblen could observe that the faith of western culture in science was unbounded, unquestioned, unrivaled. The revolt from science which then appeared so improbable as to concern only the timid academician who would ponder all contingencies, however remote, has now been forced upon the attention of scientist and layman alike. Local contagions of anti-intellectualism threaten to become epidemic." [Merton 1975 (p. 268)]

"The ethos of science is that affectively toned complex of values and norms which is held to be binding on the man of science.<sup>2</sup> The norms are expressed in the form of prescriptions, proscriptions, preferences, and permissions. They are legitimatized in terms of institutional values. These imperatives, transmitted by precept and example and reenforced by sanctions are in varying degrees internalized by the scientist, thus fashioning his scientific conscience or, if one prefers the latter-day phrase, his superego. Although the ethos of science has not been codified,<sup>3</sup> it can be inferred from the moral consensus of scientists as expressed in use and wont, in countless writings on the scientific spirit and in moral indignation directed toward contraventions of the ethos." [Merton 1975 (pp. 268 – 269)]

"--- "the sentiments embodied in the ethos of science – characterized by such terms as intellectual honesty, integrity, organized skepticism, disinterestedness, impersonality." (p. 225)

There are, however, some instances that betrayed the normative ethics of science as illustrated by historian in science (e.g. [Broad 1982])

"The antipathy toward the technological products is projected toward science itself." [Merton 1975 (Storer, p. 226)]

"Four sets of institutional imperatives – **universalism, communism, disinterestedness, organized skepticism** – are taken to comprise the ethos of modern science." [Merton 1975 (p. 270)]

We give short explanations for these key concepts below [Merton 1975];

#### A3-1 Universalism

"Universalism finds immediate expression in the canon that truth-claims, whatever their source, are to be subjected to *preestablished impersonal criteria*: consonant with observation and with previously confirmed knowledge." (p. 270)

#### A3-2 Communism

"'Communism,' in the nontechnical and extended sense of common ownership of goods, is a second integral element of the scientific ethos." (p. 273)

#### **A3-3 Disinterestedness**

""Science, as is the case with the professions in general, includes disinterestedness as a basic institutional element." (p. 275)

"It is rather a distinctive pattern of institutional control of a wide range of motives which characterizes the behavior of scientists." (p. 276)

#### A3-4 Organized Skepticism

" --- organized skepticism is variously interrelated with the other elements of the

scientific ethos. It is both a methodological and an institutional mandate." (p. 277) "Conflict becomes accentuated whenever science extends its research to new areas toward which there are institutionalized attitudes or whenever other institutions extend their control over science." (p. 278)

"- - - these conflicts (conflicts over priority) are largely a consequence of the institutional norms of science itself comes closer, I think, to the truth. For, as I shall suggest, it is these norms that exert pressure upon scientists to assert their claims, and this goes far toward explaining the seeming paradox that even those meek and unaggressive men, ordinarily slow to press their own claims in other spheres of life, will often do so in their scientific work." (p. 293)

"Recognition of what one has accomplished is thus largely a motive derived from institutional emphases. Recognition for originality becomes socially validated testimony that one has successfully lived up to the most exacting requirements of one's role as scientist. The self-image of the individual scientist will also depend greatly on the appraisals by his scientific peers of the extent to which he has lived up to this exacting and critically important aspect of his role. As Darwin once phrased it, 'My love of natural science . . . has been much aided by the ambition to be esteemed by my fellow naturalists.'" (p. 293)

#### A4 The Reward System of Science

We cite several sentences related to the Merton's concept on the reward system of science below [Merton 1975];

"--- the heart of the Mertonian paradigm – the powerful juxtaposition of the normative structure of science with its institutionally distinctive reward system – as it provides a simplified but basic model of the structure and dynamics of the scientific community."[Merton 1975 (Storer, p. 281)]

"Not only is the essential character of the rewa4rd system set forth, but in coupling it with the normative system, Merton is able to make sense of the problems which develop out of the incommensurate imperatives of these two components of the institution." (p. 282)

"Several social pathologies of science are pinpointed as the results of specific discontinuities between the normative and the reward systems so that their investigation

may contribute to a cumulating body of knowledge about science." (Storer, p. 282)

#### A5 The Processes of Evaluation in Science

The Chapter 5 titled "*The processes of Evaluation in Science*" of the Merton's Book [Merton 1975] is irrelevant to the CFP at present but is interesting to see the title of papers included in this Chapter (with ordering numbers in front of them):

- 19. Recognition and Excellence; Instructive ambiguities,
- 20. The Matthew Effect in Science,
- 21. Institutionalized Patterns of Evaluation in Science,
- 22. Age, Aging, and Age Structure in Science.

These themes taken up in the above papers will become our problems when the science of the cold fusion phenomenon is established. We cite several sentences from R.K. Merton [Merton 1975] below which are relevant our present researches.

"Matters of role performance (potential and achieved) and of recognition (instrumental and honorific) are central to all the papers, with attention focusing sequentially on the consequences of accumulated honorific recognition, the processes through which such recognition is acquired or denied, and the relevance of age (biological and social) to the preceding topics." [Merton 1975 (Storer, p. 415)]

"--- considers research questions important for the general problem of how excellence is recognized and rewarded or neglected and even penalized in society and now the effectiveness of these processes might be increased." [Merton 1975 (Storer, p. 415)]

"They then analyze the archives of the *Physical Review*, rather than only the distribution of papers actually published in that journal, to determine the kind of recognition received." [Merton 1975 (Storer, p. 416)]

"In science, according to the ideal norm of universalism, it is the quality of work, judged by peers against the current state of the art, that should entirely determine the kind of recognition received." [Merton 1975 (Storer, p. 418)]

#### A5-1 Funding Organization for the CFP

Main funding organizations for researches in the CFP are partially listed in Appendix H. It is seen that very few organizations are responding to the researches in this field due to the venturing nature of these researches.

#### **Appendix B – Controlled Thermonuclear Fusion**

We cite here the Chapter 9 of the Chen's textbook *Introduction to Plasma Physics* [Chen 1974] to show the controlled thermonuclear fusion had supplied the impetus for the growth of plasma physics and had been its important theme.

#### "Chapter 9 Introduction to Controlled Fusion

#### 9.1 The Problem of Controlled Fusion

It is entirely fitting that this book should end with an introduction, since the study of elementary plasma physics leads to an understanding of the complex problem that originally supplied the impetus for the growth of this new science—the problem of controlled thermonuclear reactions. Since the only fuel required in the ultimate fusion reactor is the heavy hydrogen in seawater, the realization of this goal would mean a virtually limitless source of energy (lasting hundreds of millions of years) at virtually zero fuel cost. The enormous impact this would make on our civilization makes controlled fusion the most important scientific challenge man has ever faced." [Chen 1974]

**"9.1.1 Reactions** (numbers of equations are renamed as (A.1) for the original (9.1)) The first generation of fusion reactors will rely on the following reactions:

$$D + T \rightarrow {}^{4}\text{He} (3.5 \text{ MeV}) + n (14.1 \text{ MeV})$$
 (A.1)

$$n + {}^{6}\text{Li} \rightarrow {}^{4}\text{He} (2.1 \text{ MeV}) + \text{T} (2.7 \text{ MeV})$$
 (A.2)

Here, n is a neutron and D and T are atoms of deuterium (<sup>2</sup>H) and tritium (<sup>3</sup>H), respectively. This reaction, as we shall see, has the lowest ignition temperature and lowest confinement requirement of all. However, most of the energy comes out in the form of a 14-MeV neutron and has to be recovered in a heat cycle, whose thermodynamic efficiency is limited to about 40%. Furthermore, the neutron causes damage to the reactor walls.

A second disadvantage of reaction (A-1) is that tritium does not occur naturally and has to be bred via reaction (A-2) in a blanket of lithium surrounding the plasma. Fortunately, <sup>6</sup>Li is an abundant isotope, comprising 7.5% of natural lithium, of which the earth's crust has a copious supply. There is an inexhaustible supply of deuterium, since 0.015% of the hydrogen in sea water is deuterium, and it is easily separated out.

A reactor fueled with deuterium alone would undergo the following reactions, which occur with about equal probability:

 $D + D \rightarrow T (1 \text{ MeV}) + p (3 \text{ MeV})$  (A-3)

 $D + D \rightarrow {}^{3}\text{He}(0.18 \text{ MeV}) + n(2.5 \text{ MeV})$  (A-4)

No tritium breeding is required, and only 34% of the energy appears in the form of neutrons. However, the requirements on the plasma are considerably more difficult to satisfy.

An attractive possibility in a fusion reactor is the use of the charged reaction products to generate electric power directly, thus avoiding the inefficiency of a thermal cycle and minimizing thermal pollution of the environment. If the high ignition temperatures can be achieved, the following reactions, which have only charged-particle products, would be suitable for this purpose:

 $D + {}^{3}\text{He} \rightarrow {}^{4}\text{He} + p + 18.3 \text{ MeV},$   $p + {}^{6}\text{Li} \rightarrow {}^{4}\text{He} + {}^{3}\text{He} + 4.0 \text{ MeV},$   $p + {}^{11}\text{B} \rightarrow 3{}^{4}\text{He} + 8.7 \text{ MeV}.$ [Chen 1974]

#### **"9.1.2 The Necessity for Plasma**

Since ions are positively charged, the Coulomb force of repulsion has to be overcome before the reactions of Section 9.1.1 can occur. Consequently, the nuclei have to be accelerated to considerable energy in order to penetrate the Coulomb barrier. For instance, the cross section  $\sigma$  for the D-T reaction rises sharply as energy is increased up to 50 keV. A peak in  $\sigma$  is reached near 100 keV, and  $\sigma$  decreases gradually at higher energies. A beam of deuterons from an accelerator cannot be used, for it can be shown that if the beam is directed at a target of solid tritium or deuterium, for instance, most of the energy is lost in ionizing and heating the target and in elastic collisions. Colliding beams cannot be made dense enough to give a fusion energy output larger than the energy required for acceleration. The solution is to form a Maxwellian plasma in which the fast particles in the tail of the distribution undergo fusion. Elastic collisions do not change the distribution function if it is Maxwellian, and the energy used to heat the plasma is retained until the particles react or escape from the chamber. This is the reason for the term *thermonuclear* reactions." [Chen 1974]

#### **"9.1.3 Ignition Temperature**

The power produced per cm3 in D-T reactions is

$$P_{\rm r} = n_{\rm D} n_{\rm T} < \sigma v > W$$

(A-6)

(A-5)

where  $\langle \sigma v \rangle$  is averaged over the Maxwellian distribution, and *W* is the 17.6 MeV of energy released in each reaction. To maintain the plasma temperature, this power must exceed that which is lost. Even if the plasma were perfectly confined, there is an

inescapable energy loss due to radiation by the electrons. This radiation, called bremsstrahlung, is emitted when electrons make elastic collisions with the ions and therefore radiate as accelerated charges. The bremsstrahlung power is given by  $P_{\rm b}=5 \times 10^{-31} Z^2 n^2 (kT_{\rm e})_{\rm keV}^{1/2}$  (A-7)

Both  $P_r$  and  $P_b$ , vary as  $n^2$ , but  $P_r$  increases much more rapidly with kT than does  $P_b$ . An ignition temperature can be found by equating  $P_r$  to  $P_b$  and assuming that the product ions have enough time to transfer their energy to the other ions and to the electrons by Coulomb collisions, so that all the temperatures are equal. For the D-T reaction, the ignition temperature is about 4 keV for the D-D reaction, it is about 35 keV. For the high-Z reactions of Eq. (A-5), even higher temperatures would be required." [Chen 1974]

#### **"9.1.4 The Lawson Criterion**

To produce more energy by fusion than is required to heat the plasma and supply the radiation losses imposes a condition on plasma density n and confinement time  $\tau$ , as well as on the temperature. It is assumed that the fusion energy, the bremsstrahlung energy, and the kinetic energy of escaping particles (the escape rate is determined by  $\tau$ ) are all recovered thermally with an efficiency not exceeding 33%. It turns out that n and  $\tau$  occur only in the Product  $n\tau$ . The minimum value of  $n\tau$  required is about  $10^{14}$  cm<sup>-3</sup> sec for D-T and about  $10^{16}$  cm<sup>-3</sup> sec for D-D. This is called the *Lawson criterion*. It is possible in principle to lower these figures by using complex schemes, such as combining beams with Plasmas or by more efficient energy recovery, such as direct conversion to electricity." [Chen 1974]

#### **"9.1.5 Major Problems**

The problems involved in developing a fusion reactor may be divided into three general areas:

- 1. Plasma confinement
- 2. Plasma heating
- 3. Fusion technology .

Confinement has to do with satisfying the Lawson criterion on  $n\tau$ . There are two different approaches: confinement by magnetic fields, with  $n = 10^{15} \text{ cm}^{-3}$  and  $\tau \sim 0.1$  sec, and inertia] confinement as in laser-produced fusion, in which  $n \sim 10^{26} \text{ cm}^{-3}$  and  $\tau \sim 10^{-11}$  sec. Magnetic confinement has received the most attention and is the best understood of the above three areas. Plasma heating is, of course, related to confinement—even a slow heating process would be good enough if the confinement

time were very long. The detailed mechanisms in heating are not yet understood. Fusion technology has to do with the engineering design of a reactor apart from the plasma aspects. The real problems in this field have yet to be faced.

In addition, we should add two subcategories on which considerable progress has been made:

- 1. Plasma diagnostics
- 2. Plasma purity

To measure the parameters of a plasma and what goes on inside it, a large variety of diagnostic methods have been developed. These involve electromagnetic waves, plasma waves, internal probe electrodes, particle beams, and external sensors. Plasma purity is an experimental problem of considerable importance, since the influx of high-Z atoms from the walls causes rapid loss of energy by atomic radiation. There are devices, called divertors, made to isolate a hot plasma from the walls effectively.

A large number of ideas for achieving the plasma conditions for fusion have been tried; but although a few nonstandard methods are still being pursued, the main experimental efforts have narrowed down to the following four approaches:

- 1. Closed systems: toruses
- 2. Open systems: magnetic mirrors
- 3. Theta pinch
- 4. Laser-fusion" [Chen 1974]

"In closed systems, the lines of force are confined within the system, even if they do not close upon themselves. Open magnetic systems work on the mirror effect described in Section 2,3.3. Pinches are plasmas carrying sufficient current to generate their own magnetic fields. The current also serves to heat the plasma. The geometry can be either open or closed. Fusion by laser works on inertial rather than magnetic confinement and, if technically feasible, would obviate the problems of magnetic instabilities." [Chen 1974 (Section 9)]

# Appendix C – ITER (International Thermonuclear Experimental Reactor)Story

To show the situation to overcome the difficulty in the realization of the controlled thermonuclear fusion projects, we cite here the explanation of the agreement to start the ITER (International Thermonuclear Experimental Reactor) project from ITER website [ITER 1985].

#### **"The ITER Story**

The launch of an international effort on fusion: US President Reagan and General Secretary Gorbachev of the Soviet Union at the Geneva Superpower Summit (1985). Nearly 30 years ago, a group of industrial nations agreed on a project to develop a new, cleaner and more sustainable source of energy.

ITER was set in motion at the Geneva Superpower Summit in November 1985, when the idea of a collaborative international project to develop fusion energy for peaceful purposes was proposed by General Secretary Gorbachev of the former Soviet Union to US President Reagan.

One year later, an agreement was reached: the European Union (Euratom), Japan, the Soviet Union and the USA would jointly pursue the design for a large international fusion facility, ITER. Conceptual design work began in 1988, followed by increasingly detailed engineering design phases until the final design for ITER was approved by the Members in 2001.

The People's Republic of China and the Republic of Korea joined the Project in 2003, followed by India in 2005. Selecting a location for ITER was a lengthy procedure that was concluded in 2005, when the ITER Members unanimously agreed on the site proposed by the European Union. The ITER installation would be built near Aix-en-Provence in southern France.

Further negotiations established the ITER Agreement to detail the construction, exploitation and decommissioning phases, as well as the financing, organization and staffing of the ITER Organization." [ITER 1985]

#### **"The ITER Agreement**

The ITER Agreement is signed at the Elysée Palace in Paris on 21 November 2006. In a ceremony hosted by French President Jacques Chirac and the President of the European Commission M. José Manuel Durao Barroso, the ITER Agreement was officially signed at the Elysée Palace in Paris on 21 November 2006 by Ministers from the seven ITER Members; this document established a legal international entity to be responsible for the building, operating, and decommissioning of the Project. Following the ratification of the international treaty by all Members, the ITER Organization was officially established on 24 October 2007. The first teams arrived on site in Saint Paul-lez-Durance, France in late 2005 after the site decision. From that moment until the start of building construction in 2010, staff was increased to approximately 500, the nuclear licensing process was initiated; site preparatory works were carried out, and procurement agencies in each ITER Member (the Domestic Agencies) were established.

Today, over 2,000 people are collaborating at the ITER site in Saint Paul-lez-Durance, France and in China, the European Union, India, Japan, Korea, Russia and the United States to build the ITER Tokamak, the world's most advanced tokamak magnetic confinement fusion experiment." [ITER 1985]

## Appendix D Controversy between Pros and Cons on the Possibility of *d-d* Fusion Reactions in CF Materials

From the point of view looking for a sustainable economic energy source, the cold fusion is an alternative of the controlled thermonuclear fusion of deuterons which have been earnestly investigated all over the world since 1950s. The motivation of the pioneering research by M. Fleischmann and S. Pons was just to realize the d - d fusion reactions in palladium metals charged with deuterium (PdD<sub>x</sub>) by the electrolytic method [Fleischmann 1989, 1990]. There have been severe controversies between pros and cons on the possibility of d - d fusion reactions in transition metal deuterides as briefly reproduced below. From the point of view to establish the science of the cold fusion phenomenon, this controversy is counterproductive and has given negative effects on the investigations of the science of nuclear reactions occurring in the CF materials (materials where occur the cold fusion phenomenon).

# D1 Persistence to the Deuterium System – Influence of Plasma Fusion Research

Proponents and opponents of the cold fusion phenomenon altogether have been trapped in the web of the deuteron-deuteron fusion reactions supposed in the frame-work of the plasma fusion (cf. Appendix B and C).

We cite here representative insistences by proponents, M. Fleischmann, S. Jones, and opposing arguments by opponents, J.R. Huizenga and D.R.O. Morrison.

J.R. Huizenga was the co-chairman of the United States Department of Energy – Energy Research Advisory Board Cold Fusion Panel and also the author of *Cold*  *Fusion: The Scientific Fiasco of the Century* (cf. Appendix D4b and [DOE 1989, Huizenga 1992]).

D.R.O. Morrison was a Professor of the CERN laboratory in Geneva, Switzerland. He published the *Cold Fusion News* (1989-1990) and *Cold Fusion Updates* (1990-2000) electronically and expressed his views on the CFP (cf. [Morrison I, 1990, II]).

# D1a Discovery of a cold fusion reaction in solids at near room temperature by M. Fleischmann et al. by accident [Fleischmann 1989]

#### **"Discussion**

We realise that the results reported here raise more questions than they provide answers, and that much further work is required on this topic. The observation of the generation of neutrons and of tritium from electrochemically compressed  $D^+$  in a Pd cathode is in itself a very surprising result and, evidently, it is necessary reconsider the quantum mechanics of electrons and deuterons in such host lattices. In particular we must ask: is it possible to achieve a fusion rate of  $10^{-19}$  s<sup>-1</sup> for reactions (v) and (vi) for clusters of deuterons (presumably located in the octahedral lattice positions) at typical energies of 1 eV? Experiments on isotopically substituted hydrides of well-defined structures might well answer this question.

The most surprising feature of our results however, is that reactions (v) and (vi) are only a small part of the overall reaction scheme and that the bulk of the energy release is due to an hitherto unknown nuclear process or processes (presumably again due to deuterons)." [Fleischmann 1989 (p. 308)]

# D1b Following the idea expressed by M. Fleischmann, a feature is confirmed by S. Jones et al. [Jones 1989]

#### "Conclusions

The correlation of ideas regarding cold piezonuclear fusion<sup>1</sup> with observations of excess <sup>3</sup>He in metals and in geothermal areas of the Earth led to our experimental studies of fusion in electrochemical cells, which began in May 1986. Our electrolyte compositions evolved from geochemical considerations, and changed as results were observed. The presence of a fusion neutron signal was consistently reproduced, although the rate varied widely. Now that our exploratory searches have disclosed a small piezonuclear fusion effect, it remains to disentangle the factors that influence the fusion rate." [Jones 1989 (p. 740)]

## D1c Examination of the experimental data on the CFP; the facts were denied based on the common sense of physics [DOE 1989]

The Committees in the Department of Energy had been composed of experts in relevant fields to the CFP and their technical opinions should be esteemed. It should, however, be pointed out limitations imposed on them by their duty different from the researchers in this field. Their duty binds them to confine their sight and also their expertise limits their investigation of the data in of the CFP inside their field preventing extension of their sight (cf. [Kozima 2016a (Section 2)]).

The characterization of the *DOE Report* presented in 1989 [DOE 1989] was given in our book [Kozima 1998a (pp. 3 - 7)]. We can cite our conclusion below on the DOE Report 1989;

"Let us point out mistakes in the DOE report.

Conclusion (1) is based on Conclusions (2) ~ (5), and it has no basis if Conclusions (2) ~ (5) are incorrect. The issue of excess heat and fusion products discussed in Conclusion (2) has significance only when D + D reaction is assumed as the main process. This assumption was adopted by the majority of the scientists at that time, including those who discovered cold fusion.

If there is some other mechanism governing the process, this argument is no longer valid. If you are searching for truth, whether one assumption made by a scientist is correct or not has no importance. You should search for the truth based on the fact that the phenomenon did occur. From this point of view, we will show, in Chapters 11 and 12, that it is possible to explain the results of cold fusion experiments without any inconsistency.

Conclusion (3) was based on the fact that the cold fusion phenomenon presented poor reproducibility. However, the reproducibility of a phenomenon is determined by the condition of the entire system, in which the process takes place. Simple analogy from other physical phenomena should not have been used to draw a conclusion. We will also show the reasons for the poor reproducibility and the way to improve it in Chapters 11 and 12.

Conclusion (4) only shows that the interpretations of the discoverers of cold fusion were not appropriate, and it has nothing to do with the truth. It is hard to believe that board members have made such an elementary mistake. It was found later that inside solid, such as Pd or Ti, with a combination of various factors, complex phenomena can occur. There is always such possibility in science. Today, it is quite obvious to everybody. The board members might have forgotten for some reason that natural science is built upon the fact. Conclusion (5) is similar to Conclusion (4). If any new findings had been denied only because they were contradiction with the existing knowledge, there would have been no progress in science and there will not be any progress in the future.

The discussions expressed in the DOE Report remind us Procrustes' bed. As Procrustes used his bed as an absolute standard to measure heights of his captives, the critiques against the cold fusion used d - d reaction as an inevitable standard to judge anomalous events." [Kozima 1998a]

A scientific spirit in the *Report* is sparkling in the following sentence added as a comment (believed to be written by N.F. Ramsey);

"- - - as a result, it is difficult convincingly to resolve all cold fusion claims since, for example, any good experiment that fails to find cold fusion can be discounted as merely not working for unknown reasons. Likewise the failure of a theory to account for cold fusion can be discounted on the grounds that the correct explanation and theory has not been provided. Consequently, with the many contradictory existing claims it is not possible at this time to state categorically that all the claims for cold fusion have been convincingly either proved or disproved - - - ." [DOE 1989]

# D1d Opponents raised their hands in the framework of traditional physics on the same line of DOE giving no insight into truth [Morrison I]

"Now we are in phase two where we do not need more press releases but experiments with good measuring devices and serious controls that would convince other scientists. In working on cold fusion, one is quickly aware of the great knowledge gap between electrochemists and particle physicists and also the different cultures and jargon. An ideal team would include both electrochemists and particle physicists." [Morrison I (No. 6 (April, 1989))]

His failure to confirm the CFP in accordance with his knowledge of nuclear physics may have made him an enthusiastic opponent against the CFP.

## D1e Proponent remained on the same stance as the original one struggling throwing himself on new idea [Fleischmann 1990]

An Overview of Cold Fusion Phenomena by M. Fleischmann;,

"There were (and are) a number of further factors which point to the possibility of inducing nuclear reactions of electrochemically compressed deuterium. The dissolved

D<sup>+</sup> is, in fact, a very high density, low ion temperature plasma existing in a high electron concentration." [Fleischmann 1990 (p. 346)]

" For the Pd-D system the central conundrum, the disparity of the excess enthalpy generation and of the expected nuclear products according to reactions (i) and (ii) however remains unsolved. It is clear that there must be *other nuclear reaction paths* of high cross-section and that these will only be discovered by a careful search for products on the surface and in the bulk of the electrodes (as well as in the solution and gas spaces)." [Fleischmann 1990 (p. 348)] (Italicized at citation).

Martin Fleischmann had considered in the early stages of CF research that "the cold fusion" as he supposed to be was induced essentially by *d-d* fusion reactions but resulting in various "phenomena" tabulated in Table 3.1 as cited below [Fleischmann 1990 (Table 1)]. The words "Cold Fusion Phenomena" used by him in this paper [Fleischmann 1990] show clearly his concept on the cold fusion phenomenon (CFP), in our words, that the CFP is composed not only of phenomena (events in our word) producing the products by ordinary reaction formulae

$D + D \rightarrow T (1 \text{ MeV}) + p (3 \text{ MeV}),$	(D-1)
$D + D \rightarrow {}^{3}\text{He} (0.18 \text{ MeV}) + n (2.5 \text{ MeV}),$	(D-2)
and	
$D + D \rightarrow {}^{4}\text{He} (0.08 \text{ MeV}) + \gamma (23.8 \text{ MeV}),$	(D-3)

but also of unknown reactions between deuterons, other nuclear reaction paths.

At this stage (in the year 1990), he has accepted the experimental results shown in Table D.1 (Table 1 of [Fleischmann 1990]) as "phenomena" induced by d-d fusion reactions not recognizing the vast variety of the phenomena (events) impossible to be explained by the d-d fusion reactions as shown later not only in deuterium systems but also in protium systems which have come out one after another soon from this year [Mills 1991, Noninski 1991, 1992, Bush 1992]. It should be also noticed that at this stage, there are no data of nuclear transmutations generating elements with different proton and nucleon numbers from elements preexisted in the system at all.

Thus, the word "phenomena" used by Fleischmann in this period reflects his concept on the events observed hitherto in the systems composed of transition metals and deuterium as a variety of results of d-d fusion reactions. In this meaning, the "phenomena" should be replaced by a word "events" expressing variety of results of the d-d fusion reactions known in nuclear physics and written down above.

We have proposed to use the name the "cold fusion phenomenon" ("CFP" in short) for the whole events caused by nuclear reactions in CF materials observed in this field including those written in the Table D.1 (Table 1 of [Fleischmann 1990]) keeping the term "cold fusion" used from 1989 and unifying whole events observed hitherto even if they have not fully explained yet.

```
Excess enthalpy
     Bursts in enthalpy
     Tritium
     Bursts in tritium
                                        Bulk,
    Neutrons
                                        Surface.
    Bursts in Neutrons
                                        Special
    X-rays
                                          sites
    Bursts in x-rays
     \gamma-rays
    Bursts in \gamma-rays
    Reaction products
    Theory: coherent phenomena
Table I. The various phenomena which have been reported
            so far in the course of this research.
```

"

Table D1 The various phenomena which have been reported so far in the course of this research by the year of 1990 [Fleischmann 1990 (Table 1)].

# D1f Opponents remained on the conventional stance without insight into truth behind facts [Morrison 1990, II]

"Experimental results on Cold Fusion are reviewed. Most experiments find no effect and the upper limits are appreciably lower than the positive effects claimed in some experiments. It is concluded that (a) there is no excess heat production, (b) the balance of evidence is strongly against fusion products." [Morrison 1990 (Abstract)]

"I am reluctant to give up well-established laws of Nature unless the proof is very strong. Here reports on cold fusion happenings are described, especially in the summary talks by True Believers in cold fusion in their words, and then some clues as to possible explanations are offered. How many Elvis sightings constitute a proof?" [Morrison II (No.13 (July, 2000) Conclusion)]

Here is a typical attitude of unscientific thinking; discard facts which are in contradiction with the "well-established laws of Nature" instead of accepting the facts and to seek clues to reconcile them with the laws of Nature.

#### D2 Fundamental attitude of proponents and oppositions

The discussions between pros and cons have been given mainly whether the d-d fusion reactions have occurred in CF materials, mainly PdD<sub>x</sub> alloys at first stage of the discussion. We have noticed this discussion is unproductive as far as we confine our investigation only in the possibility of the d-d fusion reactions in CF materials not extending our sight out to other reactions resulting into the observed experimental products.

#### D2a Proponents; M. Fleischmann:

Based on the experimental data showing huge excess heat inexplicable by probable chemical reactions in the CF materials, pros or proponents insist that *d*-*d* fusion reactions are occurring in the CF materials (e.g. [Mallove 1991]).

M. Fleischmann had expressed his credo several times in his papers. The first was at ICCF1 (1990) and the recent one was in his paper presented at ICCF7 (1998);

[Fleischmann 1990] M. Fleischmann, "An Overview of Cold Fusion Phenomena," *ICCF1 lecture* (March 31. 1990, Saturday), *Proc. ICCF1*, pp. 344 – 350 (1990).

[Fleischmann 1998a] M. Fleischmann, "Cold Fusion: Past, Present and Future," in "Abstract of Proc. ICCF7."

[Fleischmann 1998b] M. Fleischmann, "Cold Fusion: Past, Present and Future," Proc. ICCF7, pp. 119 – 127 (1998). ENECO Inc., Salt Lake City, Utah, USA.

Even if Fleischmann realized the explanation of the *d*-*d* fusion reactions (D-1) - (D-3) are difficult to occur in CF materials such as  $PdD_x$ , he expected some mechanisms, e.g. quantum field theoretical one to realize *other nuclear reaction paths*. [Fleischmann 1990 (p. 348)]

#### D2b Opponents; Huizenga, Morrison and Taubes

Based on the knowledge of quantum mechanics, solid state physics, and nuclear physics, cons or contras oppose or criticize the idea of *d-d* fusion reactions in solids proposed by the pioneers of the CFP (e.g. [DOE 1989, Morrison 1989, Morrison 1990, Huizenga 1992, Taubes 1993]).

Their logic is, in short, that the facts contradicting to the known laws of physics are not true. This logic is a too simple to be taken sincerely but perhaps is a logic taken to counterbalance the insistence of proponents. Opponents have no desire to find out truth through the facts revealing something behind them.

#### D3 Citations from papers by pros and cons;

In this subsection, we cite several sentences from papers by pros and cons to illustrate their strong persistence to the d - d fusion reactions.

#### D3a. Proponent, Mallove accepts the d - d fusion reactions naively

"It is now reasonably clear that fusion reactions that liberate energy—near but very peculiar relatives of nuclear processes that are the lifeblood of the stars—can occur at room temperature. There is no chance whatever that cold fusion is a mistake." [Mallove 1991 (preface p. x)]

# D3b. Opponents, DOE, J.R. Huizenga, D.R.O. Morrison and G. Taubes deny the d - d fusion reactions in CF materials

"A major fraction of experimenters making calorimetric measurements, either with open or closed cells, using Pd cathodes and  $D_2O$ , report neither excess heat nor fusion products. Others, however, report excess heat production and either no fusion products or fusion products at a level well below that implied by reported heat production. Internal inconsistencies and lack of predictability and reproducibility remain serious concerns." [DOE 1989 (B. Conclusions, Sec. 2)]

"There is no reason to think that these branching ratios (of *d*-*d* fusion reactions, added at citation) would be measurably altered for cold fusion. Therefore, if the Fleischmann-Pons claims of watts of excess heat were due to fusion between deuterium nuclei, large amounts of easily detectable fusion products<sup>2)</sup> would have to be present. In fact, one watt of power from nuclear reactions (1) must have associated with it approximately  $10^{12}$  (one million million) neutrons per second, neutrons being the fusion product most easily detected by direct counting.

2) Even if the branching ratios for cold fusion were very different, large amounts of fusion products would still have to be present." [Huizenga 1992 (p. 7)]

"The major conclusion is that all the measurements of fusion products agree that any possible fusion rate is many orders of magnitude less than would be needed to account for any positive measurement of excess power that is claimed." [Morrison I (No. 14, 4.6 Summary)]

"Within six months of the announcement of cold fusion, its public life had deteriorated into a dismaying struggle against reality in which the believers explained the insipid state of their science with all manner of causes, none of which was as simple as the reality itself. Cold fusion—as defined by Stanley Pons and Martin Fleischmann, or Steve Jones, or as modified by John Bockris or Edmund Storms and Carol Talcott, or Bob Huggins-Stanford, or whomever—did not exist. It never had. There was at least as much empirical evidence, if not more, to support the existence of any number of pseudoscientific phenomena, from flying saucers to astrology." [Taubes 1993 (p. 425)]

#### D4 Remarkable books on the CFP at early stage

There have been several books published by proponents to support and opponents to deny the d-d fusion reactions in CF materials [Mallove 1991, Huizenga 1992]. We cite here several typical sentences from two books by E.F. Mallove and J.R. Huizenga.

## D4a E.F. Mallove; *Fire from Ice – Searching for the Truth Behind the Cold Fusion Furor –* [Mallove 1991]

Mallove introduced the cold fusion phenomenon with too optimistic mind as an entity of real science showing production of excess energy to be used for energy sources in our industrial society based on partial experimental data sets obtained mainly in deuterium systems by the year 1991.

His declaration reflects his hastened conclusion to try justification of the d-d fusion reactions supposed by the frontiers of the CFP;

"It is now reasonably clear that fusion reaction that liberate energy—near but very peculiar relatives of nuclear processes that are the lifeblood of the stars—can occur at room temperature." [Mallove 1991 (p. x)]

"Now that many more facts are available and the furor has quieted down, the story can be told in its delicious and delirious detail. This is an account of the unfolding of a new phenomenon – the scientific process observed." [Mallove 1991 (p. x)]

In this book, Mallove was conscious of some problems involved in science of modern society;

"We will explore the scientific intrigue and infighting that occurred in the cold fusion revolution, which provided much human drama. There were fights to publish and to forestall publication, issues of priority of discovery, funding matters, misinformation and disinformation, rumors that became "fact," questions of academic standing, and even allegations of scientific deceit." [Mallove 1991 (p. xii)]

The problems picked up by him were listed up as follows [Mallove 1991 (p. xii – xiv)]:

\* Spectacular resistance to paradigm shifts in science is alive and well.

\* The majority does not rule in science.

\* It is dangerous and often deceptive to make analogies between one scientific controversy and another.

\* Irving Langmuir's rules for identifying so-called "pathological science" are best retired to the junk heap for prejudice and name calling.

\* Ockham's Razor is too easily forgotten. In science, the simplest unifying theory or connection is often most appropriate.

\* Use extreme caution in dismissing experimental results just because theory suggests they are "impossible."

\* The fear that possible scientific error would be ridiculed, or worse, interpreted as fraud, is stultifying.

\* The peer review process by which articles make their way into journals is not infallible.

\* Vested scientific interests are not easily persuaded to share their resources.

In conclusion of his investigation of experimental data on the CFP, E.F. Mallove exclaimed as follows;

"An essential caveat: After reviewing mounting evidence from cold fusion experiments, I am persuaded that it provides a compelling indication that a new kind of nuclear process is at work. I would say that the evidence is overwhelmingly compelling that cold fusion is a real, new nuclear process capable of significant excess power generation." [Mallove 1991 (p. xv)]

# D4b J.R. Huizenga; *The Scientific Fiasco of the Century* —*The Scientific Fiasco of the Century* [Huizenga 1992]

Huizenga discussed whole problem from a point of view that everything is results of the reactions (1a) - (1c) and accused data in contradiction with his assumption. "D + D  $\rightarrow$  [4He]\*  $\rightarrow$ 

<sup>3</sup> He (0.82 MeV) + n (2.45 MeV)	(1a)
T (1.01 MeV) + p (3.02 MeV)	(1b)
<sup>4</sup> He (0.08 MeV) + gamma ray (23.77 MeV)	(1c) "
$[1]_{1} = 1002 (r_{1}(r_{1}))$	

[Huizenga 1992 (p. 6)]

"The reactions (1a) and (1b) have been studied over a range of deuteron kinetic energies down to a few kiloelectron volts (keV) and the cross sections (production rates) for these two reactions have been found experimentally to be nearly equal (to within ten percent). Hence, the fusion of deuterium produces approximately equal yields of 2.45 million-electron-volts (MeV) neutrons (with an accompanying <sup>3</sup>He atom) and 3.02-MeV

protons (with an accompanying tritium atom). This near-equality of the neutron and proton branches (production rates) is expected also on the basis of theoretical arguments. The cross section (production rate) for reaction (1c) is several orders of magnitude lower than reactions (1a) and (1b). This well-established experimental result is consistent with the Bohr model which predicts that the compound nucleus will decay predominantly by particle emission [reactions (1a) and (1b)], as opposed to radiative capture [reaction (1c)], whenever it is energetically possible." [Huizenga 1992 (p. 7)]

"Based on present experimental evidence, the branching ratios for the three reactions shown by Equation (1) appear to be essentially constant at low energies. There is no reason to think that these branching ratios would be measurably altered for cold fusion. Therefore, if the Fleischmann-Pons claims of watts of excess heat were due to fusion between deuterium nuclei, large amounts of easily detectable fusion products<sup>2</sup> would have to be present. In fact, one watt of power from nuclear reactions (1) must have associated with it approximately 10<sup>12</sup> (one million million) neutrons per second, neutrons being the fusion product most easily detected by direct counting." [Huizenga 1992 (p. 7)]

"The secrecy surrounding Fleischmann and Pons' experiments on campus prior to the March 23 press conference caused major problems. The secrecy prevented open debate and discussion. This can be best illustrated by the fact that the University of Utah scientists and administrators did not ever consult members of their own physics department before announcing cold fusion to the world. To have made an announcement, of what appeared at that time to be a major scientific breakthrough in nuclear fusion, without seeking expert opinion from the physicists on campus was a serious intellectual and administrative mistake. When physicists and chemists around the United States learned through the news media about the claims of Fleischmann and Pons, they contacted their physicist and chemist colleagues at the University of Utah. It was with total disbelief that the scientific community outside Utah learned that the University of Utah physicists had been completely unaware of both the impending announcement and the cold fusion research on their own campus! No doubt the rivalry between the University of Utah and Brigham Young University, known to be near publication of its own cold fusion work, closed the debate between the two scientific groups and pressured the University of Utah officials into a premature press conference. Even so, it is hard to understand how university officials could go public on such a major claim, involving nuclear physics at its deepest level, without consulting and involving the physicists on campus. This neglect is even more striking because James Brophy, vice president for research and chief spokesman for the administration, is a physicist by training, and knew of the discrepancy between the expected nuclear products and the findings of Fleischmann and Pons." [Huizenga 1992 (p. 11)]

#### D5 Sociological comments on the conflict between pros and cons

It is interesting to notice that the controversies fought between pros and cons briefly introduced above have been discussed by R.K. Merton in the more general context as follows;

"--- The coexistence of these conflicting perspectives and interpretations within the same society leads to an active and reciprocal *distrust* between groups. Within a context of distrust, one no longer inquires into the content of beliefs and assertions to determine whether they are valid or not, one no longer confronts the assertions with relevant evidence, but introduces an entirely new question: how does it happen that these views are maintained? Thought becomes functionalized; it is interpreted in terms of its psychological or economic or social or racial sources and functions. In general, this type of functionalizing occurs when statements are doubted, when they appear so palpably implausible or absurd or biased that one need no longer examine the evidence for or against the statement but only the grounds for its being asserted at all.8 Such alien statements are "explained by" or "imputed to" special interests, unwitting motives, distorted perspectives, social position, and so on." [Merton 1973 (pp. 8 – 9)]

It is considered that the characteristics of human beings in living things on the Earth are their recognition of Truth, Beauty and Virtue. It seems, however, that they are in our mind in different depth, in the reverse order from the bottom to the surface. The order may be according to the order we obtained them in our history.

#### Appendix E – Competition for Funds

Taubes published a book [Taubes 1993] in which he investigated details of trials to get funds for researches from funding organizations in USA where it is imperative to get a fund to start and continue scientific researches compared with other industrial countries such as Italy, Japan and Russia. The competition for funds has been a cause of deviation of CF research from the ordinary route of progress of science.

A few phases of the competition have been cited below for illustration [Taubes 1993]; "Alan Bewick, an electrochemist at Southampton who was Pons's thesis adviser and Fleischmann's colleague, recalled that Fleischmann had first aired his theory of cold fusion at Southampton back in 1974 or 1975, "before Stan was even heard of." Although Fleischmann kept talking about it, however, he never got around to trying it, nor did he find a colleague at Southampton who considered it worthy enough to pursue. "When he went to Utah [in the early 1980s]," Bewick said, "it gave him the opportunity to actually try out these mad ideas."" [Taubes 1993 (p.15)]

"The operative fact in the release is the propitious 'five-and-a-half' years of work. This sets the origin of cold fusion around October 1983, at least one year before the meltdown and some two years before the earliest date given by Steve Jones's competing experiment at Brigham Young University. It is also a month or so after Pons left the University of Alberta and arrived in Utah and around the time Fleischmann took his early retirement at Southampton and became an honorary research professor. It is, coincidentally or not, the earliest date Utah could establish for cold fusion conception that might avert a legal claim from either Alberta or Southampton." [Taubes 1993 (pp. 16-17)]

"For Pons and Fleischmann, "the meltdown" or "the explosion," as it came to be called, was the pivotal event in their research. It most likely occurred in the autumn of 1984, or the winter of 1985 and represented the dramatic culmination^ Pons and Fleischmann's very first, primitive experiment in cold fusion. They had suspended a solid, one-cubic-centimeter palladium electrode from a palladium wire in a large beaker. The beaker was filled with a cocktail of heavy water and lithium. They charged the contraption by passing a current between the palladium electrode and a second electrode (if their future experiments were any indication) made of platinum." [Taubes 1993 (pp. 3 - 4)]

#### Competition

"In any case, before Jones could compose his own proposal to study piezonuclear fusion, Gajewski sent both him and Rafelski a proposal to review entitled "The Behavior of Electrochemically Compressed Hydrogen and Deuterium," by B. Stanley Pons and Martin Fleischmann. Sometime after Jones had received it, Paul Palmer had the following discussion with him:

"I've got this strange proposal that disturbs me," Jones said. "I don't know what to do about it."

"Oh, what's it about?" Palmer asked.

"Well, it's confidential," said Jones. "I better not tell you. But it could be a possible conflict of interest."

"Well," said Palmer, "who's it from?"

"I wish I hadn't told you," said Jones. "I wish I hadn't said anything about it."

"Well," Palmer said, "I now know what it's about or you wouldn't talk to me about it. It's .about cold .fusion. "

End of conversation. Jones wouldn't say any more." [Taubes 1993 (p. 35)]

#### Preliminary Notes

Press Release on March 23, 1989.

"In the March 23, 1989, press release announcing the discovery of cold fusion, Stan Pons noted that the odds of successfully generating a nuclear fusion reaction in a test tube were a billion to one against. He also said that the experiment, in theory, "made perfectly good scientific sense." Thus he and Martin Fleischmann had initiated their paradoxical exercise in nuclear research "for the fun of it" and to satisfy scientific curiosity. Once they performed the experiment, however, they had "immediate indication that it worked."

When asked at the press conference what this evidence was—"when you discovered that this one chance in a billion comes through," as a reporter phrased the question—Fleischmann told how they had induced what appeared to be a nuclear meltdown in a cube of palladium. "The thing which really triggered the whole thing off fairly early on," Fleischmann said, "was that we realized that you could generate a lot of heat, a lot." [Taubes 1993 (p. 3)]

The hastened publication of the experimental data with words "Preliminary Notes" [Fleischmann 1989] may be obtained in several years as described by Taubes as "*The operative fact in the release is the propitious 'five-and-a-half' years of work*." [Taubes 1993 (p. 16)]

It should be noticed a sentence in the Fleischmann-Pons-Hawkins paper "The most surprising feature of our results however, is that reactions (v) and (vi) are only a small part of the overall reaction scheme and that the bulk of the energy release is due to an hitherto unknown nuclear process or processes (presumably again due to deuterons)." [Fleischmann 1989 (p. 308)]

In this citation, the reactions (v) and (vi) are written as follows;  ${}^{2}_{1}H + {}^{2}_{1}H \rightarrow {}^{3}_{1}H (1.01 \text{ MeV}) + {}^{1}_{1}H (3.02 \text{ MeV})$  (v)  ${}^{2}_{1}H + {}^{2}_{1}H \rightarrow {}^{3}_{2}He (0.82 \text{ MeV}) + n (2.45 \text{ MeV})$  (vi)

Reflecting the sentence in the first paper by Fleischmann et al. cited above, Fleischmann used words "cold fusion phenomena" to express the phenomenon they have discovered by chance guided by the erroneous presumption of d-d fusion reactions in PdDx alloy as the title "An Overview of Cold Fusion Phenomena" given at ICCF1 in 1990. [Fleischmann 1990]

[ICCF1 1990]

**3:45** "An Overview of Cold Fusion Phenomena" M. Fleischmann, University of Utah/NCFI (**March 31**. 1990, Saturday)

# Appendix F – Main Journals where published papers on the Cold Fusion Phenomenon after 1989.

Main periodicals where published papers on the CFP are listed in Section F1. The decision of Dr. G.H. Miley, the Editor of the Fusion Technology (FT) in 1990 was valuable for the development of this field. His comments are cited in Section F3.

#### F1. Journals where published papers on the cold fusion phenomenon.

There are several periodicals where published papers on the CFP. Some periodicals gave a space for the CFP but closed their door soon after the explosion of confusion on the facts and explanation. Some started their lives and terminated in few years due to own affairs. Periodicals published by authorized societies such as Japanese J. of Applied Physics and European J. Physics have accepted papers on the CFP according to their quality and direction in terms of the policies of the periodicals. The course of the periodicals figured out in Fig. E1 shows a phase of our history of the CFP research.



Fig. F1 Periodicals and International Conferences on the Cold Fusion Phenomenon until 2015

## F1-1 Main periodicals on the CFP and their lives.

See Fig. F1 for the pictorial durations of these periodicals. *Cold Fusion, Elemental Energy (Cold Fusion)*, ISSN 1074-5610 (1994 – 1998) *Tables of Contents* of this journal from Vol.1 to Vol 28 are posted at CFRL websites; http://www.geocities.jp/hjrfq930/Papers/paperc/paperc.html *European J. of Physics (II Nuovo Cimento)* ISSN (1994 – ) *Fusion Technology*, ISSN 0748-1896 (1989 – 2001) *Il Nuovo Cimento*, ISSN: 0029-6341 (1989 – ) *Infinite Energy* (1995 – ) *International Journal of Hydrogen Energy*, ISSN 0360-3199, (1989 – ) *Japanese J. of Applied Physics*, ISSN *J. Electroanalytical Chemistry*, ISSN 1572-6657, (1989 – ) *J. New Energy*, ISSN 1086-8259, (1996 – 2001) *Nature* ISSN (1989)

## F1-2 Appendix to F1.

The Cold Fusion Research Laboratory (CFRL), directed by H. Kozima, has been publishing papers on the cold fusion phenomenon (CFP) written by the members of the CFRL as

*Reports of the Cold Fusion Research Laboratory*, (2004 – ).

The papers are posted at CFRL website;

http://www.geocities.jp/hjrfq930/Papers/paperr/paperr.html

Recently, the International Society for Condensed Matter Nuclear Science, has been publishing

*J. Condensed Matter Nuclear Science - Experiments and Methods in Cold Fusion* from 2010. The papers published in the Journal are posted at following website; http://lenr-canr.org/acrobat/ BiberianJPjcondensedv.pdf

# F2. Papers on CFP in protium systems published in *Fusion Technology* (FT) (1991 – 1995)

FT Vol. 20 (1991) No. 1

(TECHNICAL NOTES ON COLD FUSION)

p. 65 Excess Heat Production by the Electrolysis of an Aqueous Potassium Carbonate Electrolyte and the Implications for Cold Fusion / Randell L. Mills, Steven P. Kneizys

## FT Vol. 21 (1992) No. 2

p. 163 Excess Heat During the Electrolysis of a Light Water Solution of  $K_2CO_3$  with a Nickel Cathode / V. C. Noninski (This paper was also presented at ICCF2 as only one paper on the protium system in this Conference).

## FT Vol. 22 (1992) No. 2

p. 301 A Light Water Excess Heat Reaction Suggests That "Cold Fusion" May Be "Alkali-Hydrogen Fusion" / Robert T. Bush

## FT Vol. 24 (1993) No. 2

p. 202 Cold Fusion by Electrolysis in a Light Water-Potassium Carbonate Solution with a Nickel Electrode / Reiko Notoya

## FT Vol. 24 (1993) No. 3

p. 293 Excess Heat Evolution During Electrolysis of H<sub>2</sub>O with Nickel, Gold, Silver, and Tin Cathodes / Tadayoshi Ohmori, Michio Enyo

## FT Vol. 26 (1994) No. 2

p. 179 Tritium Generation and Large Excess Heat Evolution by Electrolysis in Light and Heavy Water-Potassium Carbonate Solutions with Nickel Electrodes /Reiko Notoya, Yohichi Noya, Toshiyuki Ohnishi

## FT Vol. 26 (1994) No. 3

(NUCLEAR REACTIONS IN SOLIDS)

p. 261 Anomalous Reactions During Arcing Between Carbon Rods in Water/R. Sundaresan, J. O'M. Bockris

#### F3. G.H. Miley; Editor's Comments on the Cold Fusion Papers

The journal *Fusion Technology* ISSN: 0748-1896, an international journal of the American Nuclear Society, had a tremendous role on the development of the cold fusion research in its important period for a few years after 1989 when it is very difficult to publish papers on the interdisciplinary field between solid state physics and nuclear physics. The decision of the Editor, G.H. Miley at that time, should be esteemed very high as a symbol of scientific spirit to venture into an unknown field. To commemorate the Editor's courageous decision, we cite here two comments, one the "call for the cold fusion papers" and another the "farewell to the *FT*."

# F3-1 G.H. Miley, "Call for technical notes on cold fusion in *Fusion Technology*," *Fusion Technology* Vol. 16, p. 116 (1989), ISSN 0748-1896.

"In view of the rapid pace in developments related to cold fusion, we intend to run, in upcoming issues of *Fusion Technology* (*FT*), a series of brief "technical notes" on that subject. The technical note section in FT is a standard feature intended for fast publication of important papers on new directions, innovative ideas, and new results. Thus, this section seems to be ideally suited for rapid communication of work in progress in cold fusion.

Technical notes do not have a page limit, but they typically run two to four journal pages (roughly three double-spaced typewritten pages per journal page). A very brief abstract is required; otherwise, the format follows that for standard manuscripts (see "Instructions to Authors" on the inside back cover of the journal). Computer disks with ASCII-format word processing can be accepted per instructions in the July issue of the journal.

Technical notes will receive a review, but this process is set up for a rapid turnaround. Reviewers are instructed to consider technical notes (versus normal manuscripts) as speculative, sometimes incomplete, work that should be judged on the basis of innovation, originality, and importance to fusion power development. Appropriate citations to prior work are also essential.

Deadlines for receipt of technical notes for upcoming issues of FT are as follows:

September issue; May 5 deadline past

November issue: June 30 deadline past

December issue: August 4

January issue: September 5.

Several technical notes have already been received for the September issue so we hope

that this represents a start for articles on cold fusion.

Send manuscripts to George H. Miley, Editor, Fusion Technology, Fusion Studies Laboratory, University of Illinois, 103 S. Goodwin Avenue, Urbana, IL 61801. If sending by Fax, use at (217) 333-2906. Any questions should be addressed to George H. Miley or Chris Stalker at (217) 333-3772."

## F3-2 G.H. Miley, "As an Editor of the Fusion Technology" *Fusion Technology*, 38, iii (November 2000), ISSN 0748-1896. (excerpt)

"It is with deep sadness that I retire in June 2001 as editor of Fusion Technology (FT). Despite the extensive time involvement, I have immensely enjoyed serving as editor. Discussions with authors and reviewers were continuously stimulating, and I always enjoyed a feeling of satisfaction from providing this service to the fusion community and to the American Nuclear Society (ANS). There were, of course, a few downsides, largely concerned with occasional financial struggles, debates over rejected manuscripts, and continued attempts to control paper backlogs that slowly oscillated back and forth from being either too large or too small as circumstances in the fusion community changed."

"Inclusion of papers on "cold fusion" (or anomalous nuclear reactions in solids) in FT has been one of the more controversial decisions I made as editor of FT. Rather than rehash the issues involved, I would simply repeat my view expressed in an early preface that it is the "responsibility of a journal to publish scientific work related to its field of coverage that can pass through peer reviews." Indeed, all papers on this topic in FT have undergone a rigorous peer review. In the early years (1987-1990) following Pons and Fleischmann's original announcement, reviewers ensured that the papers were technically sound but allowed speculations about mechanisms since the field was so new. However, starting in 1990, as the field matured, review standards reverted to the same guidelines as other papers in FT. Further, based on discussions in the FT Editorial Advisory Committee, an additional reviewer from outside the "cold fusion community" was typically added on these manuscripts. Readers who are interested in more detail about events during this period from my point of view as an editor are referred to an article titled "Some Personal Reflections on Scientific Ethics and the Cold Fusion 'Episode''' that I prepared for a fall issue of the Journal on Accountability in Research' Policies and Quality Assurance, Vol. 8, No. 1 (2000)."[Miley 2000a]

[Miley 2000a] G.H. Miley, "As an Editor of the Fusion Technology," *Fusion Technology*, 38, iii (November 2000), ISSN 0748-1896.

## Appendix G – Main International Conferences on the Cold Fusion Phenomenon until 2010

In the early days of the CFP research, there have been scheduled Conferences where were presented papers on the CFP. However, after the publication of the DOE Report in December of 1989, papers on the CFP had been published in limitted periodicals, such as *Fution Technology* and *J. Electroanalytical Chemistry*, and presented at International Conferences scheduled for the CFP. To show the main stream of the International Conferences, a brief data of the main International Conferences on the CFP is given in Fig. E1 in addition to the periodicals in the same period.



Fig. G1 Periodicals and International Conferences on the Cold Fusion Phenomenon until 2015

#### **Appendix H – Funding Organizations for the CFP**

The funding organizations on the cold fusion research have been various depending on the country and time after the discovery in 1989. This is a big theme to be discussed in a large monograph. We give here an incomplete list of organizations which have supported the CFP mainly in the early times of the history of the CF research. The economic system differs from a country to another and the list is given in main countries where have been specific organizations in addition to governmental ones.

There had appeared following funding organizations in the initial period of the

history of researches on the cold fusion phenomenon as we have given in Appendix E; DOE (Department of Energy, USA)

NSF (National Science Foundation)

Then, after 1989, there had been many organizations which supported Conferences on the CFP and Individuals. We cite typical "Organizations sponsored ICCF" (A) and "Organizations supported Individual Researchers" (B) below by countries where researches on the CFP have been performed vividly.

#### H1. USA:

AEP (the Office of Advanced Energy Projects at DOE); (cf. Appendix H5-2).

CFRI (Cold Fusion Research Institute) (A, B); Sponsored ICCF2 (June 29 – July 4, 1991).

DARPA (the Defense Advanced Research Projects Agency); (cf. Appendix H5-3).

**EPRI** (Electric Power Research Institute) (A); Sponsored ICCF2 (June 29 – July 4, 1991) and ICCF4 (Dec. 6 - 9, 1993). EPRI published a complete set of papers submitted to ICCF4 (4<sup>th</sup> International Conference on Cold Fusion held December 6 - 9, 1993, at Lahaina, Maui) as EPRI Report TR-104088, Volumes 1 through 4, which are available through the EPRI Report Distribution Center.

New York Community Trust (B); Supported several researchers for many years.

ONR (Office of Naval Research); (cf. Appendix H5-1).

**SRI** (Stanford Research Institute) (A, B); Many excellent works have been performed and published. One of the excellent works was the paper "Excess Power Observed in Electrochemical Studies of the D/Pd System," *Proc. ICCF3*, pp. 5 – 19 (1993).

Characterization of some organizations is given by G. Taubes in his book [Taubes 1993]. Some sentences have been reproduced in Appendix H5 below which will give more information about the organization than the formal description.

#### H2. JAPAN:

AISHIN SEIKI Co. Ltd. (A); Sponsored ICCF5 (April 9 – 13, 1995).

**IMRA (Europe) (A, B);** a company in the AISIN group; Published Proceedings of ICCF5 (April 9 – 13, 1995).

**JSPS** (Japan Society for the Promotion of Science) (A, B); Sponsored ICCF3 (October 21 - 25, 1992). JSPS distributes funds for researchers in all scientific fields including CFP through "Grants-in-Aid for Scientific Research."

NEDO (New Energy and Industrial Technology Development Organization) in MITI

(Ministry of International Trade and Industry, Japan) (A); Sponsored ICCF6 (October 13 – 18, 1996).

**TECHOVA** (Technova Corporation) (A, B); Sponsored ICCF2 (June 29 – July 4, 1991) and ICCF5 (April 9 – 13, 1995). Also it is supporting several researchers on the CFP.

**TEET** (The Thermal & Electric Energy Technology Inc. Foundation) (A, B); Sponsor of JCF(Japan CF-Research Society) website. Funding researchers in energy-related fields including CFP.

#### H3. ITALY:

**ENEA** (Italian National Agency for New Technologies, Energy and Sustainable Economic Development) (A);

**FIAT;** A sponsor of the ASTI Meeting held in Italy almost bi-annually as seen in Fig. G1 (cf. Appendix G).

"Four years ago Fiat, the automobile group, (and other sponsors) invited a group of nearly 20 international experts in Cold Fusion to a private meeting at Villa Riccardi, near Asti." (W.J.M.F. Collis, "Asti Workshop on Anomalies in Hydrogen / Deuterium Loaded Metals November 27-30, 1997 in Asti, Asti Province, Italy" *Infinite Energy*, December 1997-January 1998 (#17))

Italian Physical Society (A); Published Proceedings of ICCF2 and ICCF8.

#### H4. Other Countries

**ENECO Inc.** (one of the largest producers and suppliers of natural gas, electricity and heat in the Netherlands, serving more than 2 million business and residential customers.); Sponsored ICCF5.

**BARC** (The Bhabha Atomic Research Centre, India), India's premier nuclear research facility based in Trombay, Mumbai, Maharashtra. BARC is a multi-disciplinary research centre with extensive infrastructure for advanced research and development covering the entire spectrum of nuclear science, engineering and related areas; have given many contributions on the CFP from the initial period. The paper "Overview of BARC Studies in Cold Fusion," *Proc. ICCF1* (March 28 – 31, 1990. University of Utah Research Park, Salt Lake City, Utah: National Cold Fusion Institute), pp. 62 – 81 (1990) was a splendid work at this time.

### H5. Characterization of Organizations by G. Taubes.

Following sentences from G. Taubes [Taubes 1993] show us a characteristic situation of scientific researches in a society of the state capitalism (or the state controlled financial capitalism).

#### H5-1 ONR (Office of Naval Research)

"Once Stan Pons submitted his proposal to his benefactors at the Office of Naval **Research**, the cold fusion affair took on an aura of inevitability. Although ONR had a reputation for funding speculative research projects, either Pons or Robert Nowak, his funding officer, decided that ONR was not the right place for cold fusion. Pons told Hugo Rossi that he feared the Department of Defense, of which ONR was a part, might classify cold fusion. They might realize the potential military uses of an invention that, if it wasn't a hydrogen bomb itself, would still produce tritium, a necessary and valuable component of hydrogen bombs." [Taubes 1993 (p. 19)]

### H5-2 AEP (the Office of Advanced Energy Projects at DOE)

"Since Pons preferred to think of cold fusion as an energy source, not a weapon, he sent the proposal to his friend Jerry Smith, a funding agent in the Department of Energy's physical chemistry program. At one time Smith had been program manager for Pons submitted his proposal to the Office of Advanced Energy Projects at DOE, run by an administrator name Ryszard Gajewski (pronounced Richard Guy-EV-ski)." [Taubes 1993 (p. 20)]

"The office had been inaugurated in 1977 to do for the Department of Energy (what the Office of Naval Research and the Defense Advanced Research Projects Agency did for the Pentagon.) The Office of Advanced Energy Projects would support the kind of speculative ideas that might not pass peer review in the conventional programs but if given a glimmer of a chance might pan out—in other words, long shots. In particular, AEP supported exploratory research on power and energy projects that didn't fit into any of the other pigeonholes of the DOE's Basic Energy Sciences Division. The division had a yearly budget of \$10 million and a professional staff of one—Ryszard Gajewski." [Taubes 1993 (p. 20)]

#### H5-3 DARPA (the Defense Advanced Research Projects Agency)

"The office had been inaugurated in 1977 to do for the Department of Energy what the Office of Naval Research and *the Defense Advanced Research Projects Agency did for the Pentagon*. The Office of Advanced Research Projects would support the kind of speculative ideas that might not pass peer review in the conventional programs but if given a glimmer of a chance might pan out – in other words, long shots." [Taubes 1993 (p. 20)]

"Jones had just published a paper with Clinton Van Siclen of the Idaho National Engineering Laboratory on piezonuclear fusion—Jones's term for fusion induced by extraordinary pressure rather than extraordinary high temperatures." [Taubes 1993 (p. 26)]

"And, if he was then planning to use electrolysis, to condense deuterium in a metal and induce fusion, as he would claim later, he never actually wrote down the word electrolysis." [Taubes 1993 (p. 27)]

"On May 13, 1986, Jones submitted his annual progress report on muon-catalyzed fusion to Gajewski and included notes on the piezonuclear fusion ideas. Gajewski, in return, gave Jones the go-ahead to spend a share of his muon-catalyzed fusion money on . piezonuclear fusion experiments." [Taubes 1993 (p. 27)]

"This may have been an unfortunate commentary on the value of the 1989 data, because Czirr, who had been building and using detectors for thirty years, said the 1986 data, were 'nothing worth mentioning' and contained 'no real hint of anything." [Taubes 1993 (p. 29)]

"They could only procure funding in America from Gajewski, and he could only provide it so long as commercial fusion appeared to be close to the next corner. That was the institutional condition of his support." [Taubes 1993 (p. 32)]

"In September 1987, two months after the Scientific American article appeared, Gajewski managed to get Jones two more years of support at \$208,000 per year. And in March 1988, six months after Rafelski began at the University of Arizona,11 Gajewski awarded him over \$975,000 for three years to do "energy related applications of particle theory." It was an extraordinarily large grant for a theorist." [Taubes 1993 (p. 33)]

"In any case, before Jones could compose his own proposal to study piezonuclear fusion, Gajewski sent both him and Rafelski a proposal to review entitled "The Behavior of Electrochemically Compressed Hydrogen and Deuterium," by B. Stanley Pons and Martin Fleischmann." [Taubes 1993 (p. 35)]

"However, when presented with the facts that noting was done on the subject for
twenty-nine days after the meeting and that he had reviewed the Pons-Fleischmann proposal by then, Jones insisted that this level of activity still legitimately meets the definition of "vigorous pursuit." He did not deny that he may have had "impetus" from the Pons-Fleischmann proposal but argued that Pons and Fleischmann had not accused him of "impetus"—they had accused him of stealing ideas wholesale. Jones conceded that perhaps in drafting BYU's official account he should have noted that he had assigned a student to do electrolysis experiments (of the kind Paul Palmer had pursued two years earlier and Pons and Fleischmann were now proposing) only after reading the Utah proposal." [Taubes 1993 (pp. 36 - 37)]

"Whether he did or not was important merely because Pons and Fleischmann believed that Jones only "vigorously" began his research after reading their proposal, and that the fate of billions of dollars, among other things, hinged on whether he did or not. And what Pons and Fleischmann believed, rightly or wrongly, was what led them publicly and emphatically to disclose their invention on March 23, which is to say well before they had gathered sufficient data to support their claim." [Taubes 1993 (p. 37)]

"The abstract that Jones had submitted to the American Physical Society, even with two meager sentences on cold fusion, might constitute a public disclosure once it appeared in print. It was scheduled for the first week of April. That became the deadline for whatever had to be done, which gave the U roughly three weeks. The option of having Pons unveil cold fusion at the American Chemical Society meeting in the second week of April was no longer viable. Jones's APS abstract would appear a week before the meeting." [Taubes 1993 (p. 96)]

"Either way, Fleischmann certainly was the most prescient about the ugliness of the deluge that would follow a news conference. And afterward it was Fleischmann who would lay the entire responsibility for the decision and the subsequent circus on the U administration. "That was the decision of the university," he said. "You can read into that anything you want." Nonetheless, at that point, Pons and Fleischmann still could have put a stop to the affair. They did not." [Taubes 1993 (p. 97)]

"Hawkins had also taken Fleischmann to the airport, where he caught a flight back to England. According to Ron Fawcett, Fleischmann took a revised version of the JEAC paper with him on the plane. Apparently Pons also faxed a copy of the paper to Parsons in Southampton on the twenty-second, because when *JEAC* published the Pons-

Fleischmann paper [Fleischmann 1989], it said that it had been received in "revised form," on March 22. Either way, this conveniently put it one day before *Nature* received the BYU manuscript [Jones 1989]." [Taubes 1993 (Notes p. 440)] ([Fleischmann 1989] and [Jones 1989] in this quotation are added at citation and *JEAC = J. Electroanalytical Chemistry*).

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# Cold fusion by resonance of de Broglie wave

# in Multiple barrier tunnel phenomenon I

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#### Abstract

This paper describes making standardization model for anomalous heat generation and all Low Energy Nuclear Reactions. This paper proposes standardization model due to their effectiveness for understanding a cause of LENR. We propose Cold fusion model by resonance of de Broglie wave in multiple barrier tunnel phenomenon. We concluded that our LENR model are effective for explaining a cause of many phenomenon.

#### Key words

Cold cathode ray, tunnel effect, resonance, soliton, 1/f noise, de Broglie wave, Fourier component, Self-organization, Amplitude modulation, Coulomb Crystal, maser, absorption-line, spectrum, LENR

### 1. Introduction

Many of conventional studies relate to Heat of a chemical reaction and heat of a split of an atomic nucleus. But in this paper, we discuss Flow of the quantum in which potential-wells stand still in line. There is always a common condition in LENR, and there is a flow of the quantum. This study provides resonance and intervention of a flow of quantum, and spatial self-organization. It may be useful for the development of a model of all LENR and useful for reproduction of LENR with stability. Others' experiment is quoted much in this thesis. Let us look at facts from experiment in new light.

#### 2. Force of de Broglie wave and Structure in Electric Discharge

A model of LENR is created from several features. A presence of a flow of the quantum is always common to every LENR. For example there is a flow of deuterium gas in LENR which Y. Iwamura, M. Sakano and T. Itoh reported [1]. An ion has moved in the solution at experiment of M. Fleischmann and S. Pons [2]. Electrons have moved at a vacuum chamber of E . Yamaguchi and H.Sugiura's equipment [3]. Electrons are flowing in the discharge. Electrons have moved in Kamata's equipment [4]. We will now discuss the structure on the flow in detail. First We'll observe electric current discharge. A case of Fujita's and a case of JAXA are quoted in "Chapter2.1"[5,6,7,8].

### 2.1 Potential well and Bamboo screen in Discharge

For example there is an electronic flow in discharge. A model of LENR is considered from this phenomenon. Fujita took a picture of a strange phenomenon in discharge. Fujita released electron for an anode to the top from the bottom vertically in a vacuum tube[Fig.1].

It's important, but it's difficult to get the article for a reader and we can't also obtain a permit from a writer. So I wish and reprint Fujita's rest in peace.

Fujita was death from illness. We contacted some person concerned, but we can't collect yet the experimental report.

When a particle is floating by gravity and Lorentz force of the electric charge, Fujita seems to have planned. Fujita showed a hypothesis in the document, that floating particles is balanced with the electric field and the gravity[5]. We do to call this Fujita's principle or Fujita's hypothesis.

Fujita's condition poured Ar gas of pressure 25Pa into a tube of Pyrex glass of height 60cm by diameter 3.2cm and poured an electric current of 1.8mA into vertical direction. The electron temperature was measured by a probe with 2-4eV and electron densities about  $10^{15}$  m<sup>-3</sup>.Polymethl Metacrylate of 10 µm of diameter has been dropped in the tube.



top and the bottom of the picture direction. Not just flotation, but a particle stood in line every height like a bamboo screen. One of the notable features of phenomenon is file or ranks of particles in the flow. We have the interest in this phenomenon and inspect. Fujita's aim looks successfully.

But we think a scheme isn't successful. For example electrified particles are supposed. If the particles were put in space of the electric field value 0V/m, the particles would spread over the equal density. The most profitable point is restoring force by static electricity for file or ranks of particles. But when it isn't electric field 0v/m, the electric



Fig 1 Fujita's equipment [5]



Fig 2 Fujita's c Picture [5] Elevation view

charge keeps moving to the opposite side. So technology of Static flocking or technology of electrostatic deposition proves that. An electric field isn't 0v/m for Fujita's experiment. But the restoring force to return movement in the location of the particle doesn't exist in Fujita's experiment. More ranks don't stop and a rank moves to an opposite side. Fujita's experiment is different from those. So Fujita's experiment is interesting. We share neither Fujita's view nor other view.

So we'll inspect the workings of the force at the next chapter. Now, we will take a close look at Fujita's experiment.

#### 2.1.1 Contradiction of vertical by gravity

We search for the cause of the phenomenon which stood in line every height like a bamboo screen [Fig 2]. The first one of the force is gravity "Fg" in this experiment. Second of the force is Lorentz force "Fq". Only these force acts on it.

Anything but gravity and Lorentz force doesn't operate on a charged particle. The cause of the action is only gravity and Lorentz force. There are 2 kinds of ingredient in a magnetic field at the same time. One is an ingredient from movement of an electric charge, and another are the surrounding magnetic field. Though a magnetic field isn't given, there are 2 magnetic fields in Fujita's experiment. One is geomagnetic field, and the rest is a magnetic field which involves an electric current.

$$\overrightarrow{Fg} = \overrightarrow{mg}$$
 [eq. 1],  $\overrightarrow{Fq} = q(\overrightarrow{E} + v \times \overrightarrow{B})$  [eq. 2]

Fujita doesn't do the control to make the absolute value of the vector of Fg and Fq equal. Fujita makes no control E and the value of the B. Therefore even the balance of the scalar value of the vertical ingredient isn't also controlled by an experiment at all. When it isn't equivalent in the scalar value of the vertical ingredient at least, Fujita's hypothesis can't explain flotation of a particle. But, gravity is always the vertical direction. Therefore Lorentz force has to be on the same vertical axis. Then a particle shall be observed on the vertical line by Fujita's principle. When asking a floating phenomenon of a particle about the balance from a vector, so it'll be a relation of eq.3.

## $\overrightarrow{Fg} \neq \overrightarrow{Fq}$ [eq. 3]

Therefore it should be the same value in an electric charge, but "Fg" and "Fq" are the just different value. It is enough to prove mathematically that Fujita's hypothesis is false. Fujita's hypothesis makes a charged particle stand in line from balance between the force as Lorentz force and gravity. But Fq isn't always equal to Fg.

A relation of the vector which isn't equal is clear in eq. 3.

But an electric charge particle always stood in line irrespective of E and B by an experiment. Therefore Fujita's logic is a mistake.

When there are plural charged particles, each should be able to observe a parallel vertical line each other. But there is a case by which a particle stands in line radially like picture [Fig.3][6,7,8]. The picture is inconsistent with Fujita's principle.



Fig.3 Particles stand in line radially [8 by Y. Hayashi]

If gravity is the floating cause like Fujita's logic, a line of an electric charge particle will equal vertically. In Fig.3 those electric charge particles stand in line radially. The shape which stands in line radially can't be explained in Fujita's opinion. 2.1.2 Strength of the gravity.

When gravity is weak, a particle can't be caught with Fujita's principle hard. If it's the strong power, the big capture power acts on a particle. If it's the small power, the small capture power acts on a particle. When there is little gravity, so the standstill of a particle isn't continued by a little disorder. But, a particle also stood in line by feeble gravity by the environment of the universe of zero gravity. If a particle stands in line in the satellite on the orbit, it's inconsistent with Fujita's principle [Fig.3][6,7,8].

## 2.1.3 Alternating current

The strength of Lorentz force fluctuates by AC electric field, so a particle can't be stationary. A mean in a period of AC electric field is about DC 0V/m. Therefore a particle doesn't balance with gravity continuously from Lorentz force. But, alternating current corona made a particle stand in line [9,10][Fig.4]. It is inconsistent with Fujita's principle. 2.1.4 The vertical height

When it's a balance of gravity and Lorentz force, a standstill of a particle is predicted by 1 point on the

vertical line. But, a case has a line every height in vertical direction like Toda lattice [5][Fig 2]. So Fujita's principle has contradiction.

## 2.1.5 Toda lattice

The file or ranks of particles on Fig.2 is assumed Toda lattice.

Only file or ranks of quantum just like a crystal is seen in front of de Broglie wave from a cathode. But Toda lattice is found behind de Broglie wave from an anode.



Fig.4 Alternating current [9, 10 by S. Adachi et al.]



Fig.5 Toda lattice

Observation of all phenomenon show that the first important feature of the model is a Toda lattice [Fig.5]. If gravity and Lorentz force are the floating power, a lot of phenomena are inconsistent with logic. So We have to look for the 3rd power except for gravity and Lorentz force.

### 2.2 The 3rd force of de Broglie wave

We consider a floating particle to have been caught by the potential which dented like a well in Fujita's experiment. There is a line of square-well potential in Fujita's experimental chamber. A particle falls in each in a fold, and many particles line up like Toda lattice.

### 2.2.1 Cause of the force

A dent composes material phase boundary and de Broglie wave as an element.

A relation of the force is found from a definition of an effective mass of semiconductor engineering, a definition of the rate of the center of the wave packet of de Broglie wave and a definition of work [eq.4][11,12].

$$\hbar \frac{dk}{dt} = F \quad [\text{eq. 4}]$$

A fluctuation of a spatial de Broglie wave produces the force. A fluctuation of wave number k is force. For example de Broglie wave is maldistributed in space, and a wave number k is fluctuating. 2nd of the feature of the model is that the density of de Broglie wave by which a wave number k fluctuates is partial. Therefore de Broglie wave is forming a line of potential well into a chamber of LENR.

2.2.2 Traverse speed of the quantum and the width of the well

The wavelength is proportional to traverse speed of the quantum. So the width of the well is big by Fujita's vacuum tube, and the form of the line can be observed clearly. But, the electronic traverse speed is slower by an object more than a vacuum. So there are a lot of cases of quite small line. Those can't be observed easily, so we don't notice.

## 2.3 Grown-up of multiple potential well by self-organization

De Broglie wave repeats reflection at a barrier of model. So the same phenomenon happens to a maser and a laser resonator in the model. Reflection is caused by the resonator by which both ends are a mirror by a maser and a laser. A wave is shut in the density different in the respective resonators. So the density of the wave is fixed neither in the location nor at time.

### 2.3.1 The reflection in the well and changing the phase of wave

The phase of the wave changes by reflection. So a change in the phase is big for de Broglie wave which approaches a barrier. Therefore a coherent de Broglie wave occurs to potential well by the same mechanism as a maser. Most de Broglie waves shut in a barrier don't come out from a barrier besides the condition of the tunnel phenomenon. If the size, the phase and the inclination are same and continue smoothly on both sides of the boundary, de Broglie wave seeps beyond a boundary from a tunnel phenomenon. If it's the specific condition, the model who lined up a barrier is same as the nature of the distributed constant circuit to an electronic signal of a square wave. It's also same as the transmission medium to which a soliton is transmitted. But, only the wave penetrated by a tunnel phenomenon can meet the specific condition. If there is never reflection, a line of square-well potential becomes same as a transmission medium distributed constant. 2.3.2 Frequency response of de Broglie wave by square-well potential

A tunnel phenomenon occurs to de Broglie wave by square-well potential. An equation is solved by the condition that a tunnel phenomenon has occurred to square-well potential of the column [Fig.6]. A solution gets one set of the frequency, the amplitude of vibration and the phase of de Broglie wave. One point is drawn to double logarithmic chart from the frequency and the amplitude of vibration. Every time we get one solution, the number of the column of



the square-well potential is added one. And a new equation is solved and a point is drawn. This work is repeated more than 6000 times. Then the feature in inverse proportion to a square of the frequency in an envelope appeared. Nakamura did this sequence [13].

### 2.3.3 Self-organization and growth

In Nakamura's sequence, the number of columns is turning increase of the number of lines of the square-well potential gradually big from the small number. So Nakamura's sequence grew symbolically ideally. The calculation was done and Fig.7 was made .

By the way a bamboo screen was seen as Fujita's vacuum tube. A chamber and a tube of discharge have square-well potential of



the long column in an actual result. So self-organization is growing into discharge. 2.3.4 Line of the square-well potential with the not fixed width

Particles are collected from each variable distance in the origin of the self-organization. Every time it grows, each distance is separated dispersively and the same procedure is observed. The width of the square-well potential was changed to the

random distance even double [14] & [Fig.8].

The envelope which tilted still appeared in a chart. So there is a possibility that self-organization still follows quite far particle. It is inferred from this frequency response that LENR has the special quality.



2.4 Definition of quantum mechanics and frequency response chart

The energy E is related to a product of the frequency and Planck's constant h. E =  $h\nu$  [eq. 5]

Therefor the area a plot of a solution circled in a chart of the preceding section is related in the expectation value of the energy [fig.7]. A quantum mechanics formula of the expectation value of the energy W is indicated in eq.6 [11]. W of a numerical formula is equal to the area. The sinc function shows in a relational expression [eq.7].

$$W = \int_{-\infty}^{\infty} hw_{\varphi} \frac{\sin^2 \pi v}{(\pi v)^2} dv \quad \text{[eq.6], } \operatorname{sinc}(v) = \frac{\sin \pi v}{(\pi v)} \quad \text{[eq.7]}$$

There is a feature of the amplitude of vibration in inverse proportion to the frequency in an envelope for the sinc function. So there is a frequency response of the same feature as Fig.6 and Fig.7 in the definition of the expectation value W [Fig.8]. But when noise is included except for a continuous wave motion like a laser and a maser, the feature of the envelope is hidden. The model with a resonator and a laser and maser was supposed already, so it'll be a characteristic frequency response certainly.

2.4.1 Wave motion of the special feature

Function of y(x) shows the relation which becomes the exponent of x obviously in the inclination of the envelope for double logarithmic charts, but there is a chart of frequency response in the one. We assume that Fourier components of a wave motion signal was drawn in a chart of a frequency response. There are many signals similar to Fig.6 with envelope of double logarithmic chart. For example one shot pulse of rectangular wave Fig.8-a and unit step wave Fig.8-b are the case [Fig.9]. When it's a signal of the same envelope of frequency response chart, it can be expressed uniquely from the numerical sequence picked out from a frequency and those phase.



Fig.9 Form of signal wave

2.4.2 Change in the phase near the barrier of square-well potential

When those reflect de Broglie wave at a barrier laser both, the phase fluctuates certainly. The phase frequently fluctuates intensely inside the resonator naturally. The phase always fluctuates for the wave which can't penetrate through a resonator. Space density is fluctuating by replacement of most waves



Fig.10 Reflection of wave

which can't be penetrated. A fluctuation of a wave number k in eq.4 is accomplished at last, and we guess the force to act on LENR.

But the phase doesn't change and only the wave which could penetrate through a resonator by a tunnel phenomenon becomes smooth continuously in a maser.

#### 2.4.3 Propagation characteristic

For example when a signal goes through a filter, there is a possibility that the feature of the frequency response changes. The square-well potential as file or ranks of particles lets de Broglie wave through just like a filter as it was observed in Fujita's Fig.2. When a wave goes through file or ranks of particles so the shape of the wave changes. The energy is lost by transformation of a wave besides the resonance. A change in the energy also changes an envelope on a frequency response chart. Even if the phase fluctuates, when everything goes around and turns back, it would be synthesized by the original shape again. A signal of the quantum may be a distorted wave after transmission, but it may be one shot of rectangular wave as Fig.9b. An energy flow signal of one quantum always seems to be a figure as Fig. 9b, but it would be different in an element of the amplitude and the width depending on the conditions such as a kind of atom.

### 2.4.4 Spectrum by amplitude modulation

When there is a fine structure in spectrum, the amplitude of spectrum may be modulated by Ultra extreme low band frequency by less than 10Hz from sinc function.

When the summation of the wave motion signal of carrier C and signal-wave S is squared, 2CS by an amplitude-modulated ingredient is included in the product.

 $(C+S)^2 = C^2 + 2CS + S^2$  [eq.8]

The summation and the nonlinear special quality of Fig.7 are operating on de Broglie wave in space of Fig.2 by Fujita's experiment. We insist that all LENR includes this phenomenon in common. Then it's predicted that few absorption-line is carved in a fine structure in plenty in less than several Hz of interval.

### 2.4.5 Frequency response of a unit step-wave and revolution of the phase

If catching the moment when an electric current of DC electric discharge increased, a wave motion of an electric current is a unit step-wave as Fig.9a, we show S. Sugita's experiment for example[15][Fig.11]..

There is a same feature as Fig.7 in an envelope of a frequency response certainly.

$$\mathcal{F}(\text{unit step } f(t)) \propto \frac{1}{\nu} \quad [eq. 9]$$

When an electric current increases, it increases in calorific power naturally. But, there is a possibility that even noise not including the moment when an electric current increased is observed by the same frequency response. But, though a unit step-wave isn't found, the noise which changes and fluctuates by the big amplitude of vibration slowly can be observed by the same frequency response. When reflection is repeated, and the



Fig.11 Unit step-wave [15 by S. Sugita et al.]

phase changes, a unit step-wave will be warped noise or distorted wave. Such electric discharge is possible in a square-well potential line.

2.5 Relation between a differential operator, an integration operator and an exponentiation

When the inclination changes with an envelope of a frequency response, there is meaning of the differential calculus and the integration. When kinetic energy of the quantum is saved at a course, the inclination becomes leveler. If it's reverse, an envelope tilts more than before. Kinetic energy and the mass may be saved at space of electric discharge with a medium transmitted quantum. For example as if the energy is left untransported in square-well potential, and the energy has begun to spring from square-well potential extra in the reverse, we can understand LENR.

2.5.1 Unit step-Wave of springing return from Toda lattice

There is a case which can be regarded as a reverse spout of the energy which accumulated in Toda lattice. For example Toda lattice can be regarded as a distributed constant circuit to an electric current. For example a coaxial cable is a distributed constant circuit. Real equipment is used already [16]. After making them charge it between the conductor of center of axle and the peripheral cylinder conductor of a coaxial cable, when it touches, a unit step-wave electric current can be observed. A corner is sharp and the corrugation is quite better than becoming dull of the current waveform when an air condenser short-circuits. There is also unit step-wave electric current in a scanning tunneling microscope. Therefore we assume from a distributed constant circuit that the energy spouted. The envelope which means a mutual relation between integration and the differential calculus shows in the frequency response on Fig.11 [15].

#### 2.5.2 Exponent of decimal and exponent of not integer

The exponent is observed actually as the small decimal value between -2 to -1 on a phenomenon as exponent of 1/f at fluctuations. But, a random result with the envelope by which the exponent is -2 is expected at W in eq.9. When it's a phenomenon of a coherent maser in particular, only value of -2 is expected to the exponent. Mathematical proof is given by eq.10 to a possibility of this exponent value of decimal according to fractional calculus.

$$(J^lpha f)(x)=rac{1}{\Gamma(lpha)}\int_0^x (x-t)^{lpha-1}f(t)\;dt \qquad [ ext{eq.10}]$$

There is one other thing that is important for LENR. Fluctuation of 1/f is a few condition shy of LENR. LENR involves de Broglie wave penetrated by a tunnel phenomenon certainly.

#### 2.6 Equivalence of the quantum and a soliton.

If the quantum passes an observation point, the time and the quantity would be recorded together. Therefore 1 quantum which propagates in the electric discharge exists in a chart of the time vs. the number as an isolation square wave like a soliton.

A peculiar frequency response shows in the inclination of the envelope from an isolation square wave. Therefore 1 soliton is an aggregate of de Broglie wave which consists of a lot of Fourier components of the different frequency. But, when reflecting, the phase fluctuates, and 1 soliton has the aptitude which becomes a distorted wave.

When a soliton penetrates through a square-well potential barrier, there is still an envelope with the same obvious exponent in a frequency response from the soliton by which Fourier component was divided into a lot of frequencies. If there are decrease and increased of Fourier component, law of the conservation of energy can't be satisfied.

There is no form of the quantum in a distorted wave already, so it's observed as ordinary energy wave. Then the mass of the soliton is exchanged to the calorie for Einstein's principle. After LENR, the previous mass and the number of the previous quantum are different from the after quantity and the previous kind in that. 2.6.1 Round of the phase

The phase of the soliton keeps turning around, and when reaching a round, it'll be the original soliton to which the phase has returned. When a period of those common multiples passes when the phase of each Fourier component returns, a soliton returns in the former state. When a soliton is an intersection of Toda lattice at that time exactly, it's possible to return to the original quantum. At such time, it'll be an emerging substance from some discrepancies.

### 3. Discharge of alternating current

I'll now leave the discharge of direct current and turn to discharge of alternating current. A soliton reflects square-well potential manifoldly and irregularly like Fig.10.

De Broglie wave is reflection and the direction of the progress is turning alternate one after another. A lot of reflection is repeated a short period. The artificial alternating current is low-frequency easily than reverse in the direction of de Broglie wave, and therefore alternating current doesn't contribute to the workings of de Broglie wave.

Because there are no great differences in the condition, LENR also occurs to discharge of alternating current. The evidence is regarded as an experiment of T. Kumazawa et al. and their product [17].

#### 4. Conclusion

We put emphasis of theory on a flow of the quantum and built a model common to LENR. The group of square-well potential which floats in a flow of the quantum and stands in line was found. More its structure was considered about the mutual intervention when the quantum goes through. Concerning of the soliton when being reflection, and de Broglie wave was considered about the quantum. Promotion of LENR by this model is described by the 2nd paper as soon. This model would be useful for improvement of reproducibility and a design of equipment of LENR.

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