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**Japan CF Research Society** The Thermal & Electric Energy Technology Inc. Foundation

Edited by Akira Kitamura

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#### PREFACE

This is the Proceedings of the 11th Meeting of Japan CF-Research Society (JCF12), which was held at Kobe University on December 17-18, 2011. In this meeting were given 3 review articles and 14 oral presentations, out of which 13 papers were submitted for the proceedings to the editorial board. They have been peer reviewed by the referees, and revised for the publication as the proceedings. As a result, 6 papers concerning the experiment and 7 papers concerning the theory were accepted for publication.

We publish the proceedings on our web-site http://jcfrs.org, as well as the abstracts of all the presentations including the review articles. For almost all meetings from and after JCF4, we published electronic versions of Proceedings in addition to their printed versions. However, in view of efficiency and effectiveness in distributing information, we have decided to discontinue the printed version. Only the electronic version will be published hereafter. Needless to say, any responses from the world scientists will be welcomed.

Last year, we had the unprecedented nuclear disaster caused by the unexpectedly titanic Tsunami triggered by the Tohoku Earthquake. This made us reconsider more and more carefully the utilization of nuclear power plants, which almost put away the  $CO_2$  problem from our mind. However, we have to keep in mind that the nuclear energy could be a very efficient source of energy with zero emission of the greenhouse gas and other harmful oxides, if ionizing radiation is effectively suppressed.

We, JCF members, have been making research of the cold fusion that has a potential ability to establish a small-scale, radiationless nuclear reactor, and hopefully to shorten half-lives of radioactive wastes by nuclear transmutation. We believe that our approach is one of the most promising way to realize an environmentally-sound nuclear system.

Finally, we would like to thank all the participants, and also the people who have collaborated in organizing this meeting. In particular, we would appreciate the financial support by the co-host, The Thermal & Electric Energy Technology Inc. Foundation.

Editor-in-Chief Akira Kitamura, Kobe Univ., and Technova Inc. June 2012

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# Gas-phase hydrogen isotope absorption/adsorption characteristics of a Ni-based sample

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**Abstract**: Using a nano-composite sample of Ni-Cu-Zr oxide, it has been confirmed that nickel does not make observable absorption of hydrogen at room temperature. Preliminary results show that absorption of both D and H is possible at a temperature above 373 K, and the loading ratios reach about 1.0 at 525 K. As for the heat balance, the endothermic tendency is observed both in D and H runs below 500 K, above which only D-gas run gave the exothermic tendency. At 523K, while the deuterium run remains endothermic, the protium run showed the exothermic tendency with a specific output energy reaching the anomalously large value of about 300 eV/atom-Ni. If this turns out to be a true exothermic phenomenon, it is suggested strongly that a nuclear process should participate in the phenomenon.

Keywords: nano-composite sample, hydrogen isotopes, absorption, Ni·Cu, exothermic,

#### 1. Introduction

Usually nickel doesn't absorb hydrogen at room temperature. In the case of our  $Ni \cdot ZrO_2$  (NZ) sample nano-fabricated by Santoku Corporation, we haven't observed any absorption of hydrogen accompanied by exothermic temperature change [1,2]. Hydrogen absorption characteristics of the PNZ2B sample with a small fraction of Pd, Pd<sub>1</sub>Ni<sub>7</sub> nano-particles dispersed in a ZrO<sub>2</sub> supporter, supplied by B. Ahern has shown a dramatic change, and the amount of absorbed hydrogen was very large [3,4,5]. This result gives us a great hint that significant improvement of absorption characteristics would be expected by adding a small amount of foreign atoms to the NZ sample.

Recently, Rossi and Focardi (see their Internet blog J. Nuclear Physics) made a demonstration to appeal large and persistent heat generation on the order of kW from a Ni-based alloy powder sample absorbing protium (H)–gas at elevated temperatures over 573 K. Brian Ahern infers that their sample would be a mixture of Ni and Cu. In the present study, we used a ternary compound of Cu·Ni·Zr mixed oxide ("CNZ") nano-fabricated by Santoku Corporation as a sample for hydrogen isotope absorption experiments. Preliminary results implying anomalously long-lasting endothermic/exothermic characteristics are shown below.

#### 2. Experimental apparatus and preparatory absorption runs

Physical properties of the sample are tabulated in Table 1. The experimental setup is shown schematically in Fig. 1. The system is composed of two identical chambers,

	Cu	Ni	Zr
Specific surface area [m <sup>2</sup> /g]		45.3	
Composition [%]	7.9	56.1	36.0
Average grain size of oxide [nm]	6.8	24.5	

Table 1. Charactristics of the CNZ sample.



Fig. 1. Functional view of the system  $A_1 \cdot A_2$ . Water cooling of the reaction chamber with a flow rate of 6 m $\ell$ /min was done only in the #1 run at room temperature, while the pipe was emptied of the cooling water in the #5 runs at elevated temperatures.

each of which contains a reaction chamber connected to an independent gas supply system. Three thermocouples per each chamber are set up to provide isothermal calorimetry data, at a middle point of the outer surface of the reaction chamber, a bottom point and a point near the flange of the outer vacuum chamber for thermal isolation. Two outer vacuum chambers of the twin system were cooled by water kept at a regulated temperature of  $20 \pm 0.1$  °C. The laboratory was air-conditioned to keep room temperature at  $20 \pm 0.1$  °C. As blank runs of isothermal calorimetry, He-4 gas charging runs instead of H(D)-gas runs were done to take correction data for time-evolutions of the temperature and the pressure in the reaction chamber. To elevate temperature, ohmic heating was used. Time variation of input powers were routinely registered and used for correction in calorimetry calculations. Any fluctuation of the temperature of the reaction chamber not coincident with intentional change in the heater input power is regarded as a systematic error, and was evaluated to be  $\pm 0.2$  W. An example of such fluctuations is seen in Fig. 3 shown later. And the amount of absorbed hydrogen atoms in each reaction chamber can be calculated momentarily by measuring pressures of both the reaction

chamber and the reservoir tank.

Ten-gram aliquots of the as-received CNZ sample set up in each reaction chamber have been baked at 440 K for 2 hours in vacuum, and subjected to D<sub>2</sub> and H<sub>2</sub> absorption runs (#1 runs) first at room temperature. Figure 2 shows typical variation of the thermal output power data,  $W_D(t)$  and  $W_H(t)$ , pressure data in the reaction chambers,  $P_D(t)$  and  $P_H(t)$ , in units on the left vertical axis, and the time-dependent loading ratios,  $L_D(t)$  and  $L_H(t)$  in units on the right vertical axis, in the runs D-CNZ1 and H-CNZ2, respectively. Table 2 shows time-integrated parameters in these runs; the specific output energy  $E_{D(H)}$ is the time-integrated output power per 1.0-g of nickel, and the loading ratios D(H)/Pd shown are the finally (in the end of run) attained values of  $L_{D(H)}(t)$ .



Fig. 2. Results of D<sub>2</sub>/H<sub>2</sub> absorption runs for Cu·Ni·Zr oxide compounds at 300K; the run numbers are D-CNZ1#1 and H-CNZ2#1.

		U U	1	1	
Run	Duration	Output energy	Specific output energy	Averaged output power	Loading ratio
number	[h]	<i>E</i> [kJ]	$E_{\rm D(H)}$ [eV/atom-Ni]	$W_{\rm D(H)}$ [W]	D/Ni, H/Ni
D-CNZ1#1	8.33	0.00	0.00	0.00	0.06
H-CNZ1#1	8.33	0.13	0.04	0.00	0.09

Table 2. Time-integrated parameters in the #1 runs at room temperature.

These runs at room temperature, D-CNZ1#1 and H-CNZ2#1, have given small, effectively negligible, D(H) absorption rate and thermal power evolution. This fact makes us confirm that nickel even in nano-powder has negligible effect on the hydrogen absorption nor heat evolution at room temperature.

However, activation of the H(D)-gas absorption reaction is expected at higher temperature. Therefore, the experiments were also conducted at elevated temperatures up to 573 K with use of the heater input power of up to 105 W.

In the high temperature runs, since the conventional water mass flow calorimetry cannot be used, the thermal output power was measured by the change in the temperature of the reaction chamber in comparison with a blank run using inert gas instead of hydrogen isotopes. In order to make influence of environmental temperature change as small as possible, only the outer vacuum vessel was cooled with constant temperature water (regulated within 0.1 °C variation) and the experimental room was air-conditioned to keep temperature change within 0.1 °C variation. In order to obtain a calibration curve for the relation between the temperature and the colorific power, the temperature of the reaction chamber was measured for the heater input powers of 0 W, 15 W, 27 W, 43.5 W, 70 W, and 105 W. The results are shown in Table 3. Linear interpolation using the derivatives,  $\Delta T / \Delta W$  values, is applied for an arbitrary value of

In most in series in [XX7]	Т	[K]	$\Delta T$	$\Delta W$
Input power [w]	$A_1$	A <sub>2</sub>	A <sub>1</sub>	$A_2$
0.0	294.2	294.2		
15.0	393.3	388.2	6.61	6.27
27.0	435.9	429.3	3.55	3.43
43.5	480.7	472.1	2.72	2.59
70.0	524.5	523.2	1.65	1.93
105.0	559.2	558.0	0.99	0.99

Table 3. Relation between the temperature of the reaction chamber and the heater input power for each subsystem,  $A_1$  and  $A_2$ . The differential value  $\Delta T/\Delta W$  is the slope of the line connecting the neighboring points.

the temperature other than the temperatures measured in the calibration.

When a gas is introduced through the cold trap (for impurity removal) into the reaction chamber maintained at a specified elevated temperature, the temperature falls off due to the effect of cooling. Therefore, blank runs in which <sup>4</sup>He gas was introduced into the reaction chamber under the same experimental conditions as in the cases of the hydrogen runs were performed. An example of the blank run using <sup>4</sup>He operated at 70 W is shown in Fig. 3. The fall of temperature, with the difference in the heat capacity of



Fig. 3. Variation of temperature and pressure in the blank runs with <sup>4</sup>He gas, at an input power of 70 W by the heater.

He and D(H)<sub>2</sub> taken into account, is used for correction in the data processing to obtain the thermal output power  $W_{D(H)}(t)$ .

The time-dependent hydrogen loading ratio  $L_{D(H)}(t)$  is calculated from the difference in the total amount of the hydrogen gas indicated as the pressure data at the reservoir cylinder and the reaction chamber for the A<sub>1</sub> or A<sub>2</sub> subsystem, before and after introduction of the gas. Just after the introduction of the cooling gas, an apparent reduction in the loading ratio is caused by the expansion of the introduced cooling gas. The blank runs described above are used also for this purpose of corrections in the loading ratios, as well as the correction for the thermal power data. An example of the apparent loading ratio  $L_{D(H)}(t)$  in the blank run using <sup>4</sup>He at 70 W is shown in Fig. 4. This apparent reduction in the loading ratio, with the difference in the heat capacity of He and D(H)<sub>2</sub> taken into account, is used in the data processing to calculate the time-dependent loading ratio  $L_{D(t)}(t)$ . The effect of the difference in the heat capacity on the value of  $L_{D(H)}(t)$  is 30 % at most.



Fig. 4. Pressure and apparent loading ratio in the blank run with <sup>4</sup>He gas at an input power of 70 W from the heater.

#### 3. Hydrogen absorption runs at elevated temperatures

The hydrogen gas absorption runs were conducted for the CNZ1(2) samples at temperatures of 373 K, 473 K and 523 K. After the #1 runs the CNZ1(2) samples in the A<sub>1</sub> and A<sub>2</sub> chambers were baked at 440 K for 3 hours in vacuum, and then subjected to the D<sub>2</sub> (H<sub>2</sub>) absorption runs at 373K; D(H)-CNZ1(2)\_373K\_1. The baking-absorption run cycle was repeated four times with additional three runs at 473 K and 523 K; D(H)-CNZ1(2)#5\_373K\_1, D(H)-CNZ1(2)#5\_473K\_1, D(H)-CNZ1(2)#5\_525K\_1 and D(H)-CNZ1(2)#5\_525K\_2. The time evolution data of the thermal output power  $W_{D(H)}(t)$ , the pressure in the reaction chamber  $P_{D(H)}(t)$  and the loading ratio  $L_{D(H)}(t)$ 

calculated from the pressures are shown in Fig. 5-(a) through (d), and Table 4 shows the time-integrated parameters of these runs, D(H)-CNZ1(2)#5\_373K\_1 through D(H)-CNZ1(2)#5\_523K\_2.



Fig. 5. Variation of the thermal output power  $W_{D(H)}(t)$ , the pressure  $P_{D(H)}(t)$  and the time-dependent loading ratio  $L_{D(H)}(t)$  in the runs (a) D(H)-CNZ1(2)#5\_373K\_1, (b) D(H)-CNZ1(2)#5\_473K\_1, (c) D(H)-CNZ1(2)#5\_523K\_1 and (d) D(H)-CNZ1(2)#5\_523K\_2.

Dun number	Duration	Amount of	Specific output	Averaged output	D.Ni
Kull liulildel	[h]	Ni	energy	power	or H/Ni
D-CNZ1#5_373K_1	7.0	2.0	-1.06	-0.06	0.14
H-CNZ2#5_373K_1	7.0	2.0	-3.18	-0.19	0.10
D-CNZ1#5_373K_2	21.2	2.0	-6.58	-0.13	0.06
H-CNZ2#5_373K_2	21.2	2.0	-7.26	-0.14	0.06
D-CNZ1#5_473K_1	20.2	2.0	-2.86	-0.06	1.04
H-CNZ2#5_473K_1	20.2	2.0	-4.17	-0.09	0.97
D-CNZ1#5_523K_1	47.4	2.0	33.0	0.29	0.79
H-CNZ2#5_523K_1	47.4	2.0	38.7	0.34	0.92
D-CNZ1#5_523K_2	101.2	2.0	-103	-0.23	0.18
H-CNZ2#5_523K_2	191.5	2.0	278	0.61	0.21

Table 4. Time-integrated parameters in the runs D(H)-CNZ1(2)#5\_373K\_1, D(H)-CNZ1(2)#5\_473K\_1, D(H)-CNZ1(2)#5\_523K\_1 and D(H)-CNZ1(2)#5\_523K\_2...

Absorption of hydrogen was observed by heating-up a CNZ sample to 373K. However, it showed an endothermic behavior as a whole. In the D(H)-CNZ1(2)  $#5_473K_1$  runs the loading ratio increased sharply reaching to about 1.0. Although the thermal output power became positive slightly in the early stage of the run, it showed an endothermic tendency as a whole. At 523 K the loading ratio of about 1.0 was reproduced, and the prolonged exothermic tendency continued for 47 hours. Although it is only a small change compared with the input heater power of 69 W, the output energy is 19.3 (29.6) eV/atom-Ni, respectively for the D(H)-run, when the power is time-integrated up to 2000 min. This is a very big generation of heat.

In order to check the reproducibility of the large loading ratio and the prolonged exothermic tendency shown in D(H)-CNZ1(2)#5\_523K\_1, the last run (523 K\_2) was done at the same temperature. The Loading ratios in the repeated runs reached only to 0.24 and 0.25 for D and H, respectively, and the thermal output power data showed quite different behavior. While the power  $W_{\rm H}(t)$  for protium increased up to 2 W,  $W_{\rm D}(t)$  for deuterium turned back even to negative. While the time-integrated power  $E_{\rm H}$  (the output energy for protium) was calculated to be  $2.8 \times 10^2$  eV/atom-Ni, that for deuterium  $E_{\rm D}$  was negative (-1.0×10<sup>2</sup> eV).

It seems that the repeated runs D(H)-CNZ1(2)#5\_523K\_2 should be regarded as extensions of the D(H)-CNZ1(2)#5\_523K\_1 runs, since the baking process seems to have had little effect on the initial conditions of the D(H)-CNZ1(2)#5\_523K\_2 runs. This is because the baking process done at the temperature of 340 K is much lower than the operating temperature of 523 K in the D(H)-CNZ1(2)#5\_523K\_1 and D(H)-CNZ1(2)#5\_523K\_2 runs. If this is the case, and if no consideration related to the baking process is necessary, the overall loading ratios are summed up to D(H)/Ni = 1.1 (1.2), while the output energies are summed up to  $E_{D(H)} = -1.4 \times 10^2 (3.1 \times 10^2)$ eV/atom-Ni for the operating period of 2.4×10<sup>2</sup> h.

As concluding figures for the hydrogen absorption runs using the Cu-Ni-Zr mixed oxide sample, Fig. 9 and Fig. 10 summarize the integrated behavior of the specific output energy and the loading ratio as a histogram. If this reflects a true aspect of the phenomenon, the exothermic reaction is only for protium and has a threshold temperature between 200 K and 250 K, as is said by other researchers (B. Ahern, F. Piantelli, private communications). Moreover, the value of the specific output energy  $E_{\rm H}$  reaching about 300 eV/atom-Ni is too high to be explained only by known chemical processes.

(In the later experiments after JCF12, we have found that the D-gas data made catch-up to the H-gas exothermic thermal power level in the later time interval of



several weeks of long run-time. We will report it separately to the present report.)

Fig. 6. Variation of the specific output energy E in the runs D(H)-CNZ1(2)#1 through D(H)-CNZ1(2)#5\_523K\_2.



D(H)-CNZ1(2)#1 through  $D(H)-CNZ1(2)#5_523K_2$ .

## 4. Summary

It has been confirmed that nickel nano-powder does not make observable absorption of hydrogen and generation of heat at room temperature. Absorption of both D and H became possible at elevated temperatures above 300 K, and the loading ratios D(H)/Ni reached about 1.0 at 523 K.

As for the heat balance, the endothermic tendency was observed both in D and H runs below 500 K, above which only H had the exothermic tendency. At 523K, while the deuterium run remained endothermic, the protium run showed the exothermic tendency with a specific output energy reaching about 300 eV/atom-Ni which is anomalously high in view of the known chemical reactions.

If this is a true exothermic phenomenon, it cannot be a simple chemical reaction caused by re-arrangement of orbital electrons for chemical compounds, and it is suggested strongly that some nuclear process participates in the phenomenon. It is necessary to advance extended experiments further to check the reproducibility of this phenomenon with D(H) isotope effects and to understand the underlying mechanisms.

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# Silica-included Pd nano-powders repeatedly available for heat production by hydrogen isotope absorption

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**Abstract** To prevent agglomeration of nano-particles of Pd under repeated use for heat generation by gas-phase hydrogen isotope absorption, we have used silica-included Pd nano-particles. Similar results were repeatedly obtained in the repeated runs after repeated oxidization-hydridation cycles, which means that the sample structure prevents the clumping together causing deterioration of the absorption/adsorption characteristics.

keywords: Pd nano-particles, nano-porous-silica, inclusion, hydrogen isotope absorption, adsorption

# 1. Introduction

It is well known that Pd nanoparticles clump together to become agglomerated, when simple nanoparticle samples of Pd are subjected to repeated use for gas-phase hydrogen isotope absorption experiments. This causes substantial decrease in absorbed amount of hydrogen isotopes and induced heat release.

It has been reported that large heat release was observed under hydrogen isotope absorption by Pd nano-powders included in the basket structure of zeolite [1]. Purpose of inclusion in the basket structure is to prevent agglomeration of nano-particles of Pd under repeated use. In the present study we have used nano-porous-silica-included Pd nanoparticles manufactured by Admatechs Co., Ltd. instead of zeolite-included one to study effect of such structure on absorption and heat release characteristics. We call this sample "PSII".

## 2. Experimental apparatus and procedure

The PSII sample is composed of nano-particles of Pd and nano-porous-silica with a mean size of 4 µm. The size of Pd ranges from about 3 nm to about 150 nm, with 90 % being 3 – 10 nm.

We have used a twin absorption system consisting of two equivalent chambers for hydrogen isotope gas absorption/adsorption experiments. Figure 1 shows a schematic of one-part of the twin system. The samples are put in the reaction chamber, and the outer thermal insulation chamber is evacuated for during hydrogen isotope absorption/adsorption. Sheath heaters with resistance of 37.9  $\Omega$  and 53.8  $\Omega$  wound around the reaction chambers in the  $A_1$  and  $A_2$  systems, respectively, are used for sample baking, and also for sample heating in the cases of forced deoxidization and forced oxidization. Alumel-chromel thermo-couples are used to measure temperatures.

For calorimetry the coolant water is maintained constant ( $\pm 0.1$  °C) at near-room temperature with a chiller, and the flow rate is controlled with a digital coolant transmitter at a rate of 12 cc/min, which recovers heat with an efficiency of 90.0  $\pm$ 1.7 %. Calorific power is calculated from temperature difference between the exit and the entrance of water-coolant. There is a delay in the response of the temperature difference due to the indicial response with a time constant of 2.2 minutes. The calibrated

conversion factor is 0.929 W/K.

Figure 2 shows the experimental procedure. The run number expresses the conditions of the sample used in the run, using the letters defined in Table 1. The asreceived sample is baked at 440 K for 2 hours in vacuum, and subjected to the  $D_2$  (H<sub>2</sub>) absorption run (#1 run). The sample is reused either without any treatment (A or B run) or after the specified treatment; forced deoxidization (#2 run) or forced oxidization (#3 run) or passive oxidization (#4 run).

The reservoir tank is filled with  $D_2$  (H<sub>2</sub>) gas at a pressure of 0.3 MPa, typically, before an absorption run starts. The reaction chamber and the outer chamber are evacuated, and the coolant water is run. When the chambers attain constant temperature, the  $D_2$  (or H<sub>2</sub>) gas is run with a flow rate adjusted and regulated with a "Super Needle" valve.

In the case of forced deoxidization, the sample is heated after filling the reaction chamber with the hydrogen isotope gas at a pressure of 0.3 MPa, and kept at 570 K for 24 hours. On the other hand, in the case of forced oxidization, the sample is treated in the similar manner using oxygen gas at a pressure of 0.3 MPa kept at 570 K.



Tab	le	I. Nomei	nclature	e for	the	run	number.

#1	absorption run using a virgin sample
#2	absorption run after forced de-oxidization
#3	absorption run after forced oxidization
#4	absorption run after forced oxidization
#nA	absorption run following #n without baking
#nB	absorption run following #nA without baking
	desorption run by evacuation aftyer the
#n(1N)d	absorption run #n(N)

Fig. 1. Schematic of one-part of the twin system.



Fig.2. Flow-chart of the experimental procedure.

#### 3. Results and discussion

Figure 3 shows typical variation of the heat output,  $W_D(t)$  and  $W_H(t)$ , pressure in the reaction chamber,  $P_D(t)$  and  $P_H(t)$ , and the loading ratio,  $L_D(t)$  and  $L_H(t)$ , in the hydrogen isotope absorption run for as-received samples, D-PSII3 and H-PSII4, respectively. The D(H)-PSII3(4)#1 runs have extraordinary large loading ratios  $L_{D(H)}(t)$ which reach D(H)/Pd $\approx$  3.6 (3.5) at the end of the exothermic phase. This phase is thought to be dominated by chemical process, and is called the first phase in the present work. The specific heat release, is also very large;  $E_1 \approx 2.7$  (2.4) eV/atom-Pd for D(H). The values of the loading ratio are more than 4 times larger than the maximum value of 0.85 ever observed for loading bulk Pd metal, while the values of the specific heat release evaluated as the energy per one D (H) atom absorption give 0.75 (0.69) eV/atom-D(H), which is also about 4 times larger than the bulk-Pd metal value of 0.2 eV/atom-H, and even a factor of 1.5 (1.4) larger when compared with the surface adsorption energy of 0.5 eV/atom-H for bulk-Pd metal.



Fig. 3. Evolution of heat-power  $W_{D(H)}$ , pressure  $P_{D(H)}$  in the reaction chamber after introduction of D<sub>2</sub> (or H<sub>2</sub>) gas and the time-dependent loading ratio  $L_{D(H)}$  in runs D-PSII3#1 and H-PSII4#1.



Fig. 4. The run after forced deoxidization (D-PSII1#2 and H-PSII2#2)

After finishing the #1 run, we made forced deoxidization at 573 K. Figure 4 shows the #2 absorption runs using these samples. The values of D(H)/Pd and  $E_1$  were

0.95 (1.02) and 0.21 eV/atom-Pd. The loading ratio is slightly larger than that for the bulk Pd, and the values of the released heat give 0.22 (0.20) eV/atom-D(H) which are nearly equal to the bulk value. It appears that the deoxidized samples have fewer adsorption sites, and/or very small adsorption energy. If the former is the case, a higher temperature and/or a longer period of heating are/is necessary in the deoxidizing process for purging the D(H) atoms in the adsorption sites on the PSII sample surface.

Forced oxidization of the samples was performed by keeping them in the O<sub>2</sub> atmosphere at a temperature of 570 K for 30 hours. The degree of oxidization,  $x \equiv PdO/Pd$ ), is calculated from the pressure difference between the beginning (0.3 MPa) and the end of the exposure to O<sub>2</sub> in the closed system. The values of x ranged from 0.69 to 0.79. Figure 5 shows the traces of the parameters in the #3 absorption runs using the oxidized samples. The oxidized samples has almost recovered the large values of  $E_1$  and D(H)/Pd.



Next, the samples were left outside the chamber for 24 hours to allow natural oxidization in the ambient air, and subjected to the #4 absorption runs. Then the samples were subjected to forced oxidization again, followed by #3\_2 absorption runs, which were also followed by passive oxidization runs twice, #4\_2 and #4\_3.

Figures 6 and 7 show the parameters  $E_1$  and D(H)/Pd for runs #1 through #4\_3 as histograms. In the PSII#1 run the released heat and the loading ratio were both considerably large in comparison with the bulk Pd as described above. The loading

ratios of the A, B and #2 runs were about 1.0 and slightly larger than that of the bulk Pd, while the released heat remained the bulk value. In the two runs after the forced oxidation of the samples, #3 and #3\_2, the released heat and the loading ratio were largely recovered. However, the passive oxidization was not effective for substantial recovery of the large values of loading ratio and the released heat.

These results show that the PSII sample can be repeatedly used with little deterioration. The PSII sample serves our purpose.

Next, we discuss time resolved parameters. The time-resolved specific sorption energy, or differential heat of hydrogen uptake,  $\eta_{D(H)}$ , is defined as the output energy per one hydrogen isotope atom absorbed/adsorbed [2];

$$\eta(t) = \frac{\int_{t}^{t+\Delta t} W_{\text{true}}(t) dt}{L(t+\Delta t) - L(t)}$$
(1)

The time-variation of  $\eta_{D(H)}$  values is shown in Fig. 8 together with the heat-power evolution in the D-PSII1 and H-PSII2 runs.



Fig. 8. Evolution of heat, W, and the time-dependent absorption energy,  $\eta$ , for D<sub>2</sub> gas and H<sub>2</sub> gas (D-PSII1 and H-PSII2)

Qualitative time-variation of  $\eta_D/\eta_H$  values is similar to that for the PZ sample [2]. In the #1 run the first phase is clearly divided into two sub-phases, 1a and 1b. In the 1b-phase both  $\eta_D$  and  $\eta_H$  values are nearly equal to those for the bulk Pd expressed as the chain lines, while they are extraordinarily large in the 1a-phase. However, the #2 run loses the 1a-phase. And in the #3 absorption run of the oxidized sample, the 1a-phase with the large values of  $\eta_{D(H)} \approx 1.0 \text{ eV}/\text{atom-D(H)}$  is recovered.

As for the isotope effect, the value of  $\eta_D/\eta_H$  sometimes exceeds unity especially

in the beginning of the 1a-phase of the #1 runs, and also in the 1a-phase of the #3 run with a maximum of about 1.3, as is seen in the third graph. In addition, the value of  $\eta_D/\eta_H$  sometimes differs from unity also in the 1b-phase. However,  $\eta_D/\eta_H$  is erroneous in the 1b-phase, since the change in the loading ratio, the denominator of eq. (1), is sometimes very small in this phase. It is thought that  $\eta_D/\eta_H$  exceeding about 1.1 can hardly be explained by chemical isotope effect.

We have changed the extent of oxidation to examine effect of oxygen on hydrogen isotope absorption/adsorption. Table 2 together with Figs. 9 and 10 shows the released heat  $E_1$  and the loading ratio D(H)/Pd as a function of the oxidizing fraction x. Here it should be noted that any decrease in the pressure is expressed as D(H)/Pd.

-			
A1	Oxidizing fraction, x	<i>E</i> <sub>1</sub> [eV/atom-Pd]	D/Pd
#3_2	0.06	0.31	1.07
#3_4	0.29	0.67	1.42
#3	0.46	1.02	1.83
#3_3	0.69	1.39	2.20
A2	Oxidizing fraction, x	<i>E</i> <sub>1</sub> [eV/atom-Pd]	H/Pd
#3_2	0.08	0.29	1.10
#3_4	0.34	0.66	1.50
#3	0.50	0.94	1.78
#3 3	0.79	1.24	2.08

Table 2. Dependence of the released heat and the loading ratio on the oxidizing fraction.





Fig.9. Relation between the specific output energy  $E_1$  and the oxidizing fraction for the PSII samples.

Fig.10. Relation between loading ratio and the oxidizing fraction for the PSII samples.

The lines in the figures show the least-square fittings of the relevant dependences, where x is the fraction of Pd atoms oxidized as defined above, and y is either the specific output energy  $E_1$  or the ratio of number of D(H) atoms for hydridation to that of host Pd atoms, D(H)/Pd. We see that D(H)/Pd linearly increases with x, while  $E_1$  increases with x, having a saturation tendency.

We assume here that oxygen pickup reaction from PdO and the hydridation

reaction proceed in the following manner;

$$Pd + \frac{1}{2}O_{2} \rightarrow PdO + Q_{0}$$

$$D(H)_{2} + \frac{1}{2}O_{2} \rightarrow D(H)_{2}O + Q_{WD(H)}$$

$$(2)$$

$$(3)$$

where the oxide formation energy  $Q_0 = 0.886$ eV for the bulky Pd, and the water formation energy  $Q_{wD(H)} = 2.654$  (2.511) eV. Summing up these formulae, we have the following as the oxygen pickup reaction;

$$PdO + D(H)_2 \rightarrow Pd + D(H)_2O + Q_{red}; \quad Q_{red} = Q_{wD(H)} - Q_0$$
(4)

For the bulky Pd the oxygen pickup reaction energy  $Q_{\text{redD(H)}} = 1.768 (1.625)$  eV. If hydridation follows the oxygen pickup, we have the following for the total output energy  $E_1$ ;

$$PdO_{x} + \left(x' + \frac{y}{2}\right)D(H)_{2} = PdO_{x-x'} + x'D(H)_{2}O + PdD(H)_{y} + E_{1};$$
(5)

$$E_{1} = x' Q_{\text{redD}(H)} + y Q_{D(H)}.$$
 (6)

If we assume that the reaction proceeds in the above manner, the hydridation energy  $Q_{D(H)}$ , can be calculated from eq.(6) using experimental values of  $E_1$ , x' and ytogether with an assumed value of  $Q_{redD(H)}$ . In our case of the PSII samples, the hydridation energy  $Q_{D(H)}$  is calculated to be negative, when perfect deoxidization of PdO<sub>x</sub> to form  $xD(H)_2O$ , *i.e.*, x' = x, and the above value of  $Q_{redD(H)}$  are assumed. We have to assume either/both  $x' \ll x$  or/and  $Q_{redD(H)} \ll 1.768$  (1.625) eV ( $Q_O \gg$ 0.886eV).

The loading ratio for the case with x = 0,  $[D(H)/Pd]_0$ , is nearly equal to that for the case with the de-oxidized sample. The slope of the fitting line for the loading ratio in Fig. 10 should be equal to or greater than 2x'/x, even if the increase in D(H)/Pd over  $[D(H)/Pd]_0$  is thoroughly ascribed to the pickup reaction (4). Then we have to conclude that  $x'/x \le 0.93(0.71)$  for D(H), or  $x'/x \le 0.87(0.63)$  for D(H) if condensation of water is taken into account. It seems that the difference is too large as a chemical isotope effect. In the following paragraph we see that an approximation  $x'/x \ll 1$  could be reasonable.

A consequence of approximation that x'/x is negligible gives that  $Q_{D(H)} \approx 0.64(0.60) \text{ eV/atom-D(H)}$ , which is substantially larger than the bulk absorption energy and even larger than the adsorption energy. This is considered to be due to a size effect of a nanoparticle. In view of this consequence, an assumption that  $Q_0 \gg 0.886\text{eV}$  could also be probable as a result of the size effect of the nanoparticle. Anyway, it is hard to assume that the increase in the released heat and the loading ratio is ascribed completely to the oxygen pickup reaction and condensation of the water.

Now we discuss possibility of the approximation  $x'/x \ll 1$ . Figure 11 and 12 show the time-resolved relation between the loading ratio  $L_{D(H)}(t)$  and the pressure  $P_{D(H)}(t)$  for the PSII and the PNZ2B samples. The latter is a sample of nano-particles of Pd·Ni compound dispersed in ZrO<sub>2</sub> prepared by the melt-spinning method. The hydrogen absorption characteristics of the PNZ2B sample in detail will be published elsewhere. Here we refer only to the  $P_{D(H)}(t)-L_{D(H)}(t)$  diagram. After hydrogen isotope gas introduction, pressure increases without significant increase in  $L_{D(H)}(t)$  up to about 50 kPa, at which the pressure abruptly decreases down to 7 – 8 kPa, making a pressure hump. This pressure hump is considered to be typical of supersaturation followed by condensation of the  $D(H)_2O$  vapor into liquid with a vapor pressure of about 4 kPa at room temperature.

The pressure hump is not observed in the PSII sample runs. After  $L_{D(H)}(t)$  increases above 1.0,  $P_{D(H)}$  rises above 10(4) kPa with a large isotope effect, which is never related to oxygen pickup reaction, but is common to bulk absorption [2,5]. The isotope effect could be explained by difference in velocity leading to difference in probability of tunneling through the periodic potential barriers between the lattice atoms. It is therefore inferred that D<sub>2</sub>O(H<sub>2</sub>O) formation is negligible for the present PSII sample.



Fig. 11. Relation between the loading ratio and the pressure in the D(H)-PSII1(2)#3runs.

Fig. 12. Relation between the loading ratio and the pressure for the PNZ2B samples.

## 5. conclusion

The PSII samples have shown distinguishing absorption characteristics summarized in the following;

- (1) The as-received PSII samples showed very large loading ratio reaching 3.5 and absorption energy exceeding 2.5 eV/atom-Pd, both being much larger than other Pd nano-particle samples ever used [2][3][4].
- (2) Subsequent runs without baking and runs after forced de-oxidization showed almost the same characteristics;  $D(H)/Pd \approx 1.0$ , and  $E_1 \approx 0.2 0.3$  eV/atom-Pd. These are slightly larger than or equal to those of bulk Pd. However, forced oxidization recovered large values of D(H)/Pd exceeding 2.2 and  $E_1$  exceeding 1.3 eV.
- (3) Silica-inclusion of nano-particles of Pd is effective to make the sample reusable to the hydrogen isotope absorption/adsorption.
- (4) The isotope effect in the released heat in the first phase is rather large.
- (5) It has been inferred that contribution of the reaction of oxygen pickup from PdO to the observed loading ratio and the first phase output energy is small. This means that both loading ratio D(H)/Pd and the hydridation energy  $Q_{D(H)}$  are rather large compared with bulky Pd samples.

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# Investigation of nuclear phenomena in deuterium adsorption to Pd film T. Kamihira\*, Maziah. N. B. Ishak, S. Narita, H. Yamada Department of Electrical Engineering and Computer Science, Iwate University Morioka, Iwate, 020-8551 \*t2311013@iwate-u.ac.jp

#### Abstract

We have performed deuterium sorption/desorption experiment initiated by the temperature control using a thin Pd film deposited onto a Ni foil. During the sorption/desorption runs, the pressure in the chamber and the sample temperature were measured and we searched for the anomalies in the behaviors attributable to the nuclear reaction. As the result, possible charged particle emission was observed in some runs. We have also characterized the surface of the Pd film to find out the condition to induce a nuclear phenomenon.

Keywords: Absorption/desorption of deuterium, Thin-layered metal, Pd/Ni, CR-39

#### 1. Introduction

In the research of the nuclear reaction at low temperature, observations of nuclear phenomena such as charged particle emission and anomalous exothermic reaction in deuterium loading/unloading process with thin-layered metal samples have been reported [1,2]. Besides, it has been widely recognized that the low energy nuclear reaction (LENR) can be a candidate for the observed anomalies in the deuterium sorption/desorption of the nano-particles [3,4]. These results suggest that the specific properties of the fine-structured metal play important roles for the appearance of the anomalies and it is important to understand the deuterium behavior in such samples for clarifying the mechanism of the LENR. In this study, we performed deuterium sorption/desorption experiments for the Pd/Ni multi-layered sample, and investigated the nuclear phenomenon. Moreover, we analyzed the sample surface to specify the condition for triggering a nuclear reaction.

#### 2. Experiment

#### 2.1 Sample preparation

The thin-layered Pd/Ni sample was prepared by the following procedure. The Ni foil (10 mm x 10 mm x 0.1 mm) was subjected to washing by acetone followed by annealing at 900  $^{\circ}$ C for 8 hours. After annealing, the surface contaminants were removed by aqua regia. Then, the Pd layer

with the thickness of  $\sim 100$  nm was formed on the Ni foil by the radio frequency sputtering. The fabricated sample structure is shown in Fig. 1.



Fig.1 sample structure Pd/Ni

## 2.2 Absorption/desorption experiment

A stainless (SUS304 and SUS316) chamber with a volume of 0.64 L was used for the experiment. It can be evacuated by a rotary pump (ULVAC; G-20D) and a diffusion pump (ULVAC; DPF-50). The sample temperature and the pressure in the chamber were measured by a K-type thermocouple and a vacuum gauge (ULVAC; CCMT-10D), and these values were recorded by a logger.

After placing the fabricated sample into the chamber, it was evacuated to  $< 10^{-1}$  Pa. Then, the deuterium gas was supplied until inside pressure became 500 Pa. Afterwards, the chamber was cooled by ice and heated by a ribbon heater repeatedly. We expected that the deuterium atoms can be given the dynamic motion by the temperature change, resulting in sorption and desorption of deuterium. Each cooling and heating step continued for 15 minutes, and typically two cycles operation was done as a run. In the present study, we performed 20 runs in total. We also performed the experiment with a bare Ni foil sample to identify the anomalous behavior of the temperature and the pressure.

A pair of CR-39 track detectors ( $10 \text{mm} \times 10 \text{mm}$ ) was put into the chamber to observe the particle emission. The CR-39 is a kind of plastic detector and the track pit made along the trajectory of a charged particle appears after chemical etching. In this experiment, the CR-39 plastics were etched with 5 mol/l NaOH solutions at 70 °C for 8 hours. After etching, we surveyed the surface of the CR-39 with the digital microscope to observe the pit tracks. In principle, we can obtain the information on the particle energy from the track size. Two pieces of CR-39 were placed in the chamber. One piece called "foreground (FG)" was set just facing the sample. The other piece called "background (BG)" was placed so as not to be affected by the charged particle tracks emitted from the sample for the background track evaluation.



Fig.2 Experimental apparatus

We also characterized the Pd film sample by the atomic force microscope (AFM) and the scanning electron microscope (SEM) to clarify the conditions for triggering the nuclear reaction.

# 3. Results and Discussions3.1 Temperature and Pressure Measurements



Fig.3 Time variation of the sample temperature and the pressure in the chamber for the experiment with Pd/Ni sample



Fig.4 Time variation of the sample temperature and the pressure in the chamber for the experiment with Ni foil sample

Figure 3 and 4 show the time resolved behaviors of the sample temperature and the pressure in the chamber in the experiment with Pd/Ni and Ni foil samples, respectively.

The sample temperature got to 0-5 °C in the cooling stage, and it increased gradually to ~40 °C at the heating stage. The pressure change was correlated with the temperature change, and it was well explained by the equation of gas state. As long as these macroscopic parameters are compared, no anomalous behaviors were found which may indicate the occurrence of a nuclear reaction.

## 3.2 Charged particle measurement

Figure 5 shows the number of tracks observed on FG and BG pieces.



Fig.5 Number of tracks on CR-39 for FG and BG pieces.

The mean value and the standard deviation of the BG distribution were 6.0 /cm<sup>2</sup> and 2.9 /cm<sup>2</sup>, respectively. Considering these values on the basis of the 3- $\sigma$  criterion, we

regard the events with the number of tracks above 14 /cm<sup>2</sup> as anomalous ones. In 20 total runs, there were 2 runs with the number of tracks above that. We found the number of tracks of 22 /cm<sup>2</sup> and 15 /cm<sup>2</sup> in each run. We mention these runs "run-1" (N=22 /cm<sup>2</sup>) and "run-2" (N=15 /cm<sup>2</sup>), hereafter.



Fig.6 Track-diameter distribution

Figure 6 shows distribution of the diameter of the FG tracks observed in "run-1" and "run-2" compared with that of the BG tracks observed in 20 total runs. The mean diameters of the FG tracks and the BG tracks were 8.3  $\mu$ m and 9.4  $\mu$ m, respectively. No clear difference was observed between these two distributions. Figure 7 and 8 shows the track-diameter distribution for "run-1" and "run-2" compared with that for the background tracks.



Fig.7 Track-diameter distribution in the run-1



Fig.8 Track-diameter distribution in the run-2

The mean diameters of the tracks observed in "run-1" and "runt-2" were 6.2  $\mu$ m and 6.8  $\mu$ m, respectively. These values were apparently smaller than the mean diameter of the BG tracks. The tracks with smaller diameter in "run-1" and "run-2" were possibly due to charged particles.

# **3.2 Surface Analysis**

Also, we analyzed the sample surface.



Fig.10 Typical AFM images of the sample surface before the experiment



Fig.11 Typical AFM images of the sample surface after the experiment.

Figure 10 and 11 show the AFM images of the sample surface before and after the experiment. We analyzed several areas on the surfaces and two typical images are

shown in each. Many grains and protrusion were found in every sample, and the surface condition was different on every spot analyzed even on the sample. No significant change was found in the surface condition of the sample after exposing to deuterium, including the sample used in "run-1" and "run-2", that is, the runs in which a large number of tracks was observed on CR-39.



Fig.12 Typical SEM images of the sample surface before the experiment.



Fig.13 Typical SEM images of the sample surface after the experiment.

Figure 12 and 13 show the SEM images of the sample before and after the experiment. The grain boundaries were found and they got narrower after the experiment. This change might be due to deuterium sorption. As for the runs in which anomalous distribution of the track diameter were observed, *i.e.* "run-1" and "run-2", no distinctive conditions were found in the SEM image.

# 4. Conclusion

In this study, we have performed absorption/desorption of deuterium using a Pd/Ni thin-layered sample. Then, we investigated the charged particle emission possibly caused by some anomalous exothermic nuclear reaction. We have observed the events with large number of tracks on CR-39 in 2 out of 20 total runs, which indicated the charged particle emission. We have analyzed the sample surface by the AFM and the SEM to specify conditions associated with the anomalous events. However, we have not found any specific states as of now. It is necessary to make extended measurements of

the particle tracks as well as systematic surface characterization to clarify the mechanism of the LENR.

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# Deuterium desorption test using Ni/Pd multi-layered sample

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#### ABSTRACT

We performed the deuterium desorption test using Ni/Pd multi-layered sample and investigated the nuclear phenomena induced. Unexpected heat evolution on the sample was often observed in the deuterium desorption process. The phenomena might be attributed to a unique property of the deuterium diffusion in Ni-Pd complex system. We estimated the heat balance considering the possible exothermic and endothermic processes in the deuterium diffusion, and we gave a limit for the quantity of the heat contributed by a nuclear reaction. We also observed the considerably large number of tracks on the CR-39 which can be the evidence of the charged particle emission.

Keywords: Pd/Ni multi-layered sample, heat of deuterium dissolution

#### 1 Introduction

In the past study, we have investigated the low energy nuclear reaction in the deuterium desorption from the various types of the multi-layered film sample such as Au/Pd/MnO, Pd/Au, Pd/CaO, Au/Pd/CaO, Pd/Pd, and we observed the events possibly associated with the nuclear reactions [1-3]. However, the origin of the phenomena or the condition for inducing the events has not been understood. Then, the systematic study with various conditions is still necessary. Ni is one of the interesting materials in the study of the condensed matter nuclear reaction. The events indicating the occurrence of the nuclear reaction have been often observed in deuterium/hydrogen absorbing and desorbing processes with the Ni or the Ni-Pd complex [4,5]. It is supposed that a specific property of Ni may induce the low energy nuclear reaction effectively.

In terms of hydrogen/deuterium dissolution, Pd, Ti and V have negative enthalpy, while Ni, Pt and Ag have positive one [6]. A hydrogen storing alloy is made of these two types of metals, in principle. If the complex of thin layers with these two types of metal has a similar property to an alloy in the interface region, a high deuterium density is expected in that region. As the result, a nuclear reaction may be induced. In this study, we performed the deuterium desorption test using Ni/Pd multi-layered sample and investigated the possibility of the occurrence of a nuclear reaction.

## 2 Experiment

The multi-layered Ni/Pd sample was fabricated by depositing Ni 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 x 10 mm x 0.5 mm and 10 mm x 10 mm x 0.1 mm. The thickness of Ni membrane was ~100 nm. In this paper, the multi-layered samples with Pd foil with thickness of 0.5 mm and 0.1 mm are respectively mentioned *NiPd05* and *NiPd01*.

For loading deuterium to the sample, the fabricated multi-layered sample was exposed to 5 atm deuterium gases for ~24 hours. The weight of the sample was measured before and after loading, and the loading ratio (D/Pd) was calculated from the weight difference. After weighing the sample to measure the D/Pd ratio, it was put into the chamber which can be evacuated by a TMP (~10<sup>-4</sup> Pa). In the chamber, the sample was heated by supplying electric current to stimulate the deuterium out-diffusion from the sample. In the desorption test, the sample temperature and the pressure in the chamber were monitored continuously for ~24 hours. The thermo-couple and the ionization gauge were used for their measurements. The current and the bias applied to the sample were also registered during the experiment.

The CR-39 track detector (BARYOTRAK; Nagase Landauer Ltd.) was used for charged particle detection. The detector is made of allyl-diglycol-carbonate (ADC) with thickness of ~0.9 mm. A particle irradiated to the CR-39 deposits a part of the energy. As the result, chemical composition of the materials is changed along the particle trajectory and the latent tracks are generated inside. If we make etching process for the detector in the chemical reagent, a track pit appears because the etching rate for the latent tracks is larger than other regions. In this study, the etching conditions were 5 N NaOH solutions at 70 °C for 8 hours. The 4 pieces of CR-39 were placed surrounding the multi-layered sample. The layout of the CR-39 in the chamber is shown in Fig. 1. For evaluating the background tracks on the CR-39, we performed experiments with the same condition as the desorption test but without setting the multi-layered sample. (These runs are mentioned "*blank runs*", hereafter).



Figure 1. The layout of CR-39 in the vacuum chamber in the desorption experiment.

## 3 Results and Discussion

Figure 2 shows the D/Pd ratios of *NiPd05* and *NiPd01* samples comparing with those for other type of multi-layered samples measured in our past experiments [1-3]. Their mean values are summarized in Table 1. The D/Pd ratio for the Ni/Pd sample looks comparable to that for other type of multi-layered sample. Note that the loading ratio shown in the figure is the value averaged over the sample entirely. The distribution of the deuterium density is possibly different for each sample. As for the difference of the thickness of the Pd substrate, no clear dependency was found.



Figure 2. D/Pd ratio for the multi-layered samples. ("MHI-type" expresses the Pd/CaO multi-layered complex having the same structure of the sample used in the deuterium permeation experiment by MHI-group [7])

Sample type	NiPd05	NiPd03	Bulk Pd	Au/Pd	CaO/Pd	Au/Pd/CaO	MHI-type
D/Pd	0.68	0.66	0.61	0.64	0.70	0.71	0.68

Table 1. The mean D/Pd for each multi-layered sample.

Figure 3 shows the time resolved behavior of the sample temperature and the pressure in the chamber in the deuterium desorption experiment. The temperature increased just after applying DC current. Then, it showed unstable behavior. Especially, the significant temperature increases were observed several times during the experiment. In some events, the pressure in the chamber and the applied electric power simultaneously changed. Such unexpected temperature increase was observed 14 out of 18 runs with *NiPd05* and 4 out of 13 runs with *NiPd01*, respectively. We have observed such anomalous heat evolution in our previous experiments with some types of multi-layered sample [1,3]. However, the phenomena were much more often observed in the present study with Ni/Pd sample.

Now the heat balance for the observed temperature increase is estimated quantitatively considering the following processes; 1) Joule heating due to fluctuation of the electric power applied, 2) Deuterium dissolution into Pd, 3) Deuterium diffusion from Pd layer into Ni layer and other process including a nuclear reaction. Here, the heat values for each process are expressed as  $H_1$ ,  $H_2$  and  $H_3$ , respectively. The heat  $H_{obs}$  derived from the observed temperature increase  $\Delta T$  can be written as

$$H_{obs} = mC\Delta T = H_1 - H_2 + H_3 \tag{1}$$

where *m* is the weight of Pd foil and *C* is the specific heat of Pd.  $H_1$  can be estimated by the relationship between the change of the input power and the sample temperature. We have performed the calibration test to obtain it in advance.  $H_2$  can be calculated by the amount of deuterium desorbed simultaneously with the temperature increase, which can be obtained by the change of the pressure monitored. Although the heat of the deuterium dissolution depends on the amount of deuterium remained in the metal in practice, we assumed that it is a constant value, that is, -10 kJ/mol, in this estimation. Then, we can derive  $H_3$ . Figure 4 is a part of Fig. 3, showing the first 10 hours of the experiment. For each event with the temperature increase (1)-(6), the calculated values of  $H_3$  are shown in Table 2.  $H_3$  was found to be almost zero for every event, anyhow. Assuming that  $H_3$ includes the contribution from a nuclear reaction, the heat should be comparable to that



generated in the process of deuterium diffusion from the Pd foil to the Ni layer.

Figure 3. Time dependence of the sample temperature, the pressure in the chamber, and the bias applied to the sample.



Figure 4. Time dependence of the sample temperature, the pressure in the chamber, and the bias applied to the sample in the first 10 hours of the experiment.
	(1)	(2)	(3)	(4)	(5)	(6)
$H_3$ [J]	-1.8	-1.9	-1.0	0.5	0.6	1.3

Table 2. Heat balance at each point with significant increase of temperature

Figure 5 shows the number of tracks recorded on the CR-39 in the desorption experiments and the *blank runs*. The events with a large number of tracks were found in the runs with *NiPd05* rather than *NiPd01*. However, even in the *blank runs*, numerous tracks were sometimes observed. Figure 6 shows the number of tracks and the mean diameter for the runs with *NiPd05* and the *blank runs*. These distributions look quite similar and no specific events considered as the evidence of charged particle emission were observed.

The sensitivity of the CR-39 in the particle detection definitely depends on the environments where it is used. Especially, it has been reported that the sensitivity can degrade by using the detector under the vacuum condition [8]. When the charged particle enters the CR-39, the molecular chain around the trajectory is broken. Then, the latent track is formed by coupling of such molecular defects and the dissolved oxygen. In case of using the detector in the vacuum, the dissolved oxygen is easily diffused to outside of the detector. Thus, the efficiency to form the latent track decreases. In terms of formation of the false tracks, we should consider the surface damage and the defects in the constituent molecules, which can appear as tracks by the etching [9]. It varies from lot to lot and batch to batch in the same lot. Additionally, the defects can be generated during the experiment. For example, the molecular defect can be caused by the diffusion of the inner gases of the detector. It might be possible that a marked number of tracks observed in blank runs were formed by such gas out-diffusion process since the CR-39 was placed in the highly evacuated chamber in our experiment. Besides, the formation of the latent tracks and the etching rate can be affected by various conditions such as the environmental temperature, aging effect in the long-term storage, and so on [10,11]. Therefore, we should carefully consider these effects when we use the CR-39 to identify the rare event.



Figure 5. Number of tracks on CR-39 in runs with NiPd05 (left) and NiPd01 (middle), and in blank



Figure 6. Number of tracks vs. mean diameter for "Type-A runs" (left) and "blank runs".

#### 4 Summary

We have performed deuterium desorption experiment with the multi-layered Ni/Pd sample. We have frequently observed unexpected increase of the sample temperature in the deuterium desorption process. It was possibly due to the unique properties of Ni-Pd system. The heat balance by the possible processes was preliminarily estimated at each temperature change. As the result, the heat contributed by a nuclear reaction, if it occurred, was found to be comparable to the heat of the deuterium dissolution to the Ni layer. In charged particle detection by the CR-39, marked number of tracks was sometimes observed although we have not clarified the origin of the tracks. We still have to study the uncertainties of the tracks on CR-39 as well as trying other detection methods to confirm the events as the nuclear origin.

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# Change in Isotopic Ratio of Li by Light Water Electrolysis

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#### Abstract

A light water electrolysis of  $Li_2SO_4$  solution with constant current density of 3 mA/cm<sup>2</sup> for 2 weeks using several metal cathodes including Au was performed. The cathode surface after the electrolysis and that of the control sample were analyzed by TOF-SIMS. The isotopic ratio of Li on the cathode metals Co, Ni and Pd as well as that on the control sample fairly agrees with that of natural one. To the contrary, a marked change in the isotopic ratio of Li was sometimes observed only on the Au cathode after the electrolysis. The largest change in the occupation ratio of <sup>6</sup>Li (<sup>6</sup>Li/<sup>6</sup>Li+<sup>7</sup>Li) was 0.114 which is remarkably larger than the natural value of 0.076.

#### 1. Introduction

The low energy nuclear reaction (LENR) in deuteride system has been widely studied using several experimental methods for the last two decades. Further, similar studies in hydrogen system also have presented the positive results for LENR<sup>1, 2)</sup>. Among several experimental methods for the reaction, electrolysis method is one of the attractive methods. Ohmori et al. <sup>3, 4)</sup> have studied using this method and have reported the production of several elements on the cathodes after the DC light water electrolysis.

The difference in the results between the deuterium and hydrogen systems would be interesting to construct the model commonly acceptable for the mechanism of LENR. Such difference would be expected to observe in a change in the isotopic ratio of elements after the electrolysis. The change in the isotopic ratio of any elements is considered to be an obvious evidence of LENR. For recognizing the change in isotopic ratio of any element by mass spectroscopy, there exists a difficulty in distinguishing one element from other molecules. This ascribed to that a larger value of atomic mass unit (u) of an element is usually closely those of other molecules, when the mass number of the element equals to that of other molecules. However, in case of counting number of substance with mass number 6, only element <sup>6</sup>Li is counted as mass number 6 by time-of-flight secondary ion mass spectroscopy (TOF-SIMS); there is no overlapping <sup>6</sup>Li with other elements at mass number 6. Accordingly, we have been interested in the change in the isotopic ratio of Li rather than these of other elements. In this present study, we performed DC light water electrolysis of Li<sub>2</sub>SO<sub>4</sub> solution and investigated the change in the isotopic ratio of Li using TOF-SIMS.

## 2. Experimental

The electrolysis was carried out in a cell shown schematically in left side of Fig. 1. It consists of a cylindrical vessel made of PTFE with volume capacity of  $\sim 100$  cm<sup>3</sup> and a plastic stopper holding the test electrodes. The top of the stopper was covered by a small plastic cap with loose contact, which permits the escape of the gas produced inside of the cell by the electrolysis. A Pt foil of 0.1x5x10 mm in size was employed as the anode. Metals Au, Co, Ni and Pd were employed as the cathode. These metals were supplied from Nilaco Corporation in Japan. The Au and Co cathodes of 0.1x5x10 mm in size were bulk foil as received from the supplier. The Ni and Pd film cathodes of 150 nm thickness were deposited on Al<sub>2</sub>O<sub>3</sub> foil of 0.5x10x10 mm in size by vacuum deposition and sputtering methods, respectively. Another Au foil of 0.1x5x10 mm in size, which surface was scraped with a cleaned edge of a scissor made of ceramics, was also used as the cathode. Both electrodes were suspended by  $\phi 0.5$  mm lead wires made of same metal used for each electrode. The upper portions of the both electrodes were sheathed by heat-shrinkable FEP tube surrounded with TFE. The gap spacing was ~10 mm and the electrolyte solution used was 0.001 M Li<sub>2</sub>SO<sub>4</sub> which was prepared from Merck superpure grade chemicals. The electrolysis was conducted for 2 weeks by a constant current density of 3 mA/cm<sup>2</sup>. The current for the electrolysis was supplied by a constant-current power supply and no water was added during the electrolysis.

In order to investigate the change in the composition of elements on the cathode surface, we prepared a control sample using a control cell. The cell, shown in right side of Fig. 1, was a same cylindrical vessel as that used for the electrolysis, but without electrodes. The control sample was immersed into the same solution for the same period of 2 weeks without electrolysis. The cathode surface after the electrolysis and that of the control sample were analyzed by TOF–SIMS. The primary ion in TOF-SIMS was Ga<sup>+</sup> and measured area was  $40x40 \ \mu\text{m}^2$ . Three randomly selected areas on the surface were analyzed. The spectrometry for the sample surface was also performed after 10 s sputter cleaning with the Ga<sup>+</sup> ion. This sputtering can remove several atomic layers of the upper most surface of the samples.



Control sample

Fig. 1 Experimental test cell (left), control test cell (right).

#### 3. Result

The occupation ratio of <sup>6</sup>Li out of all the Li isotopes is defined as Eq. (1), where  $\Sigma^{6}$ Li and  $\Sigma^{7}$ Li are the total count of <sup>6</sup>Li and <sup>7</sup>Li, respectively.

$$\frac{\sum_{i=1}^{6} \text{Li}}{\sum_{i=1}^{7} \text{Li} + \sum_{i=1}^{6} \text{Li}} \times 100 \quad [\%]$$
(1)

The u ranges of counting <sup>6</sup>Li and <sup>7</sup>Li in the TOF-SIMS spectrum are indicated in Fig. 2. The relation between the occupation ratio of <sup>6</sup>Li and the total count of <sup>7</sup>Li for cathode metals Co, Pd and Ni are shown in Figs. 3, 4 and 5, respectively. The occupation ratios obtained from each measured area in the electrolysis and control samples are provided as red and blue dots in the figures, respectively. No difference of plot distribution in the relation between the occupation ratio and count number of <sup>7</sup>Li is seen for the control samples of Co, Pd, Ni and Au. Therefore, all the data for the control samples of Co, Pd, Ni and Au were compiled to use commonly for the data of the control experiments in Figs. 3-7. The red horizontal line in each figure indicates 7.59% of the natural occupation ratio of <sup>6</sup>Li.



Fig. 2 TOF-SIMS spectrum around mass number range 6-7.

When the number of produced ion of an element increases exceedingly, the counted number of the ion in TOF-SIMS becomes less than the produced one. This characteristic is seen even at the small total count of <sup>7</sup>Li in these figures. Beside, the occupation ratio of <sup>6</sup>Li increases with the total count of <sup>7</sup>Li. However, it was found in the figures that such count error is relatively small; the ratios for the control samples are less than 9.0% in the region of total count of <sup>7</sup>Li less than  $1.4 \times 10^5$ .

No apparent change in the occupation ratio of <sup>6</sup>Li is seen in this region of these figures between electrolysis and control samples. Namely, the occupation ratio of <sup>6</sup>Li obtained from the cathode foil Co, Ni and Pd as well as the control sample looks to fairly agree with that of natural one in the region of total count of <sup>7</sup>Li less than  $1.4 \times 10^5$ .



Fig. 3 The relation between the occupation ratio of <sup>6</sup>Li and the total count of <sup>7</sup>Li for Co cathode. The red horizontal line in figure indicates 7.59% of the natural occupation ratio of <sup>6</sup>Li.



Fig. 4 The relation between the occupation ratio of <sup>6</sup>Li and the total count of <sup>7</sup>Li for Pd cathode. The red horizontal line in figure indicates 7.59% of the natural occupation ratio of <sup>6</sup>Li.



Fig. 5 The relation between the occupation ratio of <sup>6</sup>Li and the total count of <sup>7</sup>Li for Ni cathode. The red horizontal line in figure indicates 7.59% of the natural occupation ratio of <sup>6</sup>Li.

In contrast, a marked change in the occupation ratio of <sup>6</sup>Li was sometimes observed only on the Au foil cathode after the electrolysis. Fig. 6 shows the relation between the occupation ratio of <sup>6</sup>Li and the total count of <sup>7</sup>Li for the Au foil, which presents two increased values of the occupation ratios in the region of total count of <sup>7</sup>Li less than  $1.4\times10^5$ . In addition, there exists another such large ratio of ~10 at the count of ~  $2.0\times10^5$ . The red horizontal line in the figure indicates 7.59% of the natural occupation ratio of <sup>6</sup>Li. The largest change in the occupation ratio of <sup>6</sup>Li, shown as the pointed end of the red arrow in Fig. 6, was 11.4% which was remarkably larger than the natural value of 7.6%. The sample and the area providing the ratio of 11.4% is designated as "RED ARROW". The three ratios including the largest one, which were obtained from the same electrolysis sample, are given with red circles in the figure. The three correspoding ratios for the control sample are given with green squares.

This remarkably large ratio of 11.4% is compared with the ratios obtained from the corresponding control sample, as precisely shown in Table 1. Similarly, the remarkably large ratio is compared with the ratios obtained from other randomly selected two areas in the same electrolysis sample, as shown in Table 2. It should be noticed that the 5 values of the total count of <sup>7</sup>Li, obtained from the corresponding control sample and 2 other areas on the electrolysis sample, are commonly less than  $1.4 \times 10^5$ . Thus, the count error of the 5 values for the total count of <sup>6</sup>Li is considered to be small. It is found in the last columns of the two tables that these compared 5 occupation ratios of <sup>6</sup>Li equal fairly to the natural one. This could imply that no LENR

occurred in the 5 areas corresponding to the 5 normal occupation ratios of  ${}^{6}Li$ . Further, the result shown in Table 2 suggests that the LENR took place at a small part of surface area but did not over the all surface of the Au foil cathode.



- Fig. 6 The relation between the occupation ratio of <sup>6</sup>Li and the total count of <sup>7</sup>Li for Au cathode. The red horizontal line in figure indicates 7.59% of the natural occupation ratio of <sup>6</sup>Li.
- Table 1Total count of <sup>6</sup>Li, <sup>7</sup>Li and the occupation ratio of <sup>6</sup>Li for "RED ARROW",<br/>compared with those obtained from the corresponding control sample.

Area	Total count of <sup>7</sup> Li	Total count of <sup>6</sup> Li	The occupation ratio of <sup>6</sup> Li(%)
RED ARROW	60866	7846	11.4
1 in control	40751	3788	8.50
2 in control	10816	905	7.72
3 in control	33018	3246	8.95

Table 2Total count of <sup>6</sup>Li, <sup>7</sup>Li and the occupation ratio of <sup>6</sup>Li for "RED ARROW",<br/>compared with those obtained from other two randomly selected areas.

Area	Total count of <sup>7</sup> Li	Total count of <sup>6</sup> Li	The occupation ratio of $^{6}\text{Li}(\%)$
RED ARROW	60866	7846	11.4
Area 1	2571	200	7.22
Area 2	10353	962	8.50

Similar relation for the scraped Au foil was shown Fig. 7. The red horizontal line in the figure indicates 7.59% of the natural occupation ratio of <sup>6</sup>Li. Of particular interest is that several red dots obtained from the samples of electrolysis experiments are positioned above those obtained from the control ones over the whole total count range of <sup>7</sup>Li. This suggests that the anomalous change in the occupation ratio took place at any total count of <sup>7</sup>Li. The surface inhomogeneous on the Au foil cathode caused by the scrapping might stimulate the LENR. Ohmori et al. have reported the considerable amount of newly produced elements after the light water electrolysis, using the Au cathode scraped with a cleaned glass fragment edge<sup>3, 4)</sup>.



Fig. 7 The relation between the occupation ratio of <sup>6</sup>Li and the total count of <sup>7</sup>Li for scraped Au cathode. The red horizontal line in figure indicates 7.59% of the natural occupation ratio of <sup>6</sup>Li.

# 4. Conclusion

The occupation ratio of <sup>6</sup>Li was observed to increase with the total count of <sup>7</sup>Li by the count error of TOF-SIMS for all the cathodes tested and all the control samples. It was found that the count error for the occupation ratio of <sup>6</sup>Li was relatively small in the region of total count of <sup>7</sup>Li less than  $1.4 \times 10^5$ . The occupation ratio of <sup>6</sup>Li for the cathode metals Co, Ni and Pd as well as for the control sample fairly agrees with that of natural one. To the contrary, a marked change in the occupation ratio of <sup>6</sup>Li was sometimes observed only on the Au foil cathode after the electrolysis. When the scraped Au foil cathode was used, the similar anomaly larger occupation ratio of <sup>6</sup>Li was observed over the whole total count range of <sup>7</sup>Li.

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# Numerous etch pits on CR-39 produced by light and heavy water electrolysis

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#### Abstract

We performed the electrolysis of Li<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O, Na<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O, Li<sub>2</sub>SO<sub>4</sub>/D<sub>2</sub>O and LiOH/D<sub>2</sub>O solutions under DC current range 20-160 mA for time range 20min-168 h using cathodes of a 5µm thick Ni film and a 2.5µm thick Au film. A CR-39 detector of  $30 \times 30$ mm in size is set in close contact with the rear surface of the metal film cathode to detect an energetic charged particle. Anomalous increasing in number density of etch pit is sometimes observed on the chip for the electrolysis experiment. These results suggest that a low energy nuclear reaction occurs and an energetic charged particle is generated at the Ni film during the electrolysis.

#### 1. Introduction

The plastic track detector has become a popular method to detect energetic charged particles in low energy nuclear reaction (LENR) studies especially in electrolysis experiments. In these studies, the evidence of the reaction is in the form of nuclear damage trails made visible by etching of the plastic chips. Lipson et al.<sup>1)</sup> and Oriani et al.<sup>2)</sup> performed light and heavy water electrolysis using the plastic track detector CR-39 and have reported the generation of charged particle emission during the electrolysis. However, there still exist technical complexities in using plastic detector in electrolysis experiment. In the previous studies, there have been a thin layer of electrolyte and a solid film between the cathode electrode and the plastic detector. The construction could cause a considerable decrease in the energy of the charged particle emitted from the cathode.

In this present study, a chip of the plastic track detector CR-39 is positioned just under the metal film cathode to avoid such energy decrease  $^{3,4)}$ ; a CR-39 chip of 30x30 mm in size is set in close contact with the rear surface of the cathode film. The simplicity of this construction is expected to detect energetic charged particles produced on cathode during electrolysis with relatively high efficiency.

#### 2. Experimental

The test cell made of Polyoxymethylene is shown in Fig. 1. The anode is  $\phi 0.5$  mm Pt wire. The upper portion of it is sheathed by heat-shrinkable PTFE tube and the lower part is formed spiral. The cathode is a 5 µm thick Ni film and a 2.5 µm thick Au film. The lower end of the spiral plane is parallel to the cathode surface. The distance between the lower end of the anode and the cathode is ~10 mm. The arrangement of the cathode and the chip of the track

detector CR-39 is shown in the small circle of Fig. 1(a). A CR-39 chip of 30x30 mm in size is set in close contact with the rear surface of the cathode film. Both the metal film and the CR-39 chip are clamped together on the disc forming the bottom cap of the cell with an O-ring seal. This construction is able to avoid chemical attack on the CR-39 chip by ions generated in the electrolyte and keep the ideal distance between the cathode and the detector chip.



Fig. 1 Test cell for the electrolysis, (a) vertical cross section, (b) the view of the component of the cell (left) and its assembled (right).





The electrolyte solution for the heavy water electrolysis was  $0.1M \text{ Li}_2\text{SO}_4/\text{D}_2\text{O}$  and  $0.1M \text{ LiOH}/\text{D}_2\text{O}$ . That for the light water electrolysis is  $0.1M \text{ Li}_2\text{SO}_4/\text{H}_2\text{O}$  and  $0.1M \text{ Na}_2\text{SO}_4/\text{H}_2\text{O}$ . The volume of the electrolyte solution in the test cell is ~6 ml. The electrolysis was conducted for 200 min and 168 h under DC current range 3-160 mA at voltage range 3-30 V. After the electrolysis experiment, the cell assembly is immediately disassembled to remove the CR-39 chip and the metal film used.

A control experiment is performed without electrolysis. The CR-39 chip used in control experiment is carefully handled in exactly the same way as those used in the electrolysis experiments. The exposure time of CR-39 for the control experiments is the same as that for the electrolysis experiment.

The CR-39 chip is etched in 6N NaOH solution for 7 h at 70 °C immediately after each experiment. Then, the surface of the chip is observed to count the number, to measure the

diameter and to take photograph of the etch pit by a digital microscope system. The measurement area 10x10 mm is the center of the chip surface as shown in Fig. 2.

Total 11 types of electrolysis experiments were performed for H<sub>2</sub>O and D<sub>2</sub>O solutions and all the electrolysis conditions are compiled into Table 1. There were 4 DC application patterns in the 11 types of electrolysis. The patterns are designated as "20mA", "20mA/20min-R", "20mA/24h-R" and "3-160mA-S", as indicated in the Table 1. The "20mA" means that the application current is constant DC 20 mA for one week as shown in Fig. 3. The "20mA/20min-R" consisted of 8 cycles as shown in Fig. 4; the application current was DC 20 mA with negative cathode for 15 min, followed by DC 20 mA with positive cathode for 5 min in each cycle. Thus, the total electrolysis time is 200 min. The "20mA/24h-R" consisted of 7 cycles as shown in Fig. 5; the application current is DC 20 mA with negative cathode for 23 h and 55 min, followed by DC 20mA with positive cathode for 5 min in each cycle. The total electrolysis time is 168 h. The "3-160mA-S" means that DC is changed from 3 to 160 mA in stepwise every 24 h as shown in Fig. 6.

Solvent	Electrolyte	mol/l	Cathode	Current pattern	Total time of electrolysis	Type of electrolysis
D <sub>2</sub> O	Li <sub>2</sub> SO <sub>4</sub>	0.1	Ni	20mA	168h	E1
		0.1	Ni	20mA/20min-R	200min	E2
		0.1	Ni	20mA/24h-R	168h	E3
		0.1	Ni	3-160mA-S	168h	E4
	LiOH	0.1	Ni	20mA	168h	E5
		0.1	Ni	3-160mA-S	168h	E6
H <sub>2</sub> O	Li <sub>2</sub> SO <sub>4</sub>	0.1	Ni	3-160mA-S	168h	E7
		0.1	Au	3-160mA-S	168h	E8
	Na <sub>2</sub> SO <sub>4</sub>	0.1	Ni	3-160mA-S	168h	E9
		0.1	Ni	20mA/24h-R	168h	E10
		0.1	Au	20mA/24h-R	168h	E11

Table 1Electrolysis condition.



Fig. 3 Current pattern "20mA".



Fig. 4 Current pattern "20mA/20min-R".



Fig. 5 Current pattern "20mA/24h-R". One cycle of the pattern is shown in the right side.



Fig. 6 Current pattern "3-160mA-S".

- 3. Result and discussion
- 3.1 Control experiment

The relation between total number and the diameter of the etch pit for control experiment is shown in Fig. 7. The total number of pits for each diameter bin in the figure is the sum of them over 30 runs. The total number of etch pit over all the bins is 134, from which 4.5/ chip as the relatively small average number of etch pit. No marked peak is fairly observed in the distribution.



Fig. 7 Distribution of etch pit diameter collected from 30 runs of control experiment.

#### 3.2 Electrolysis of $D_2O$ solution

The result of most of the experiment in 11 electrolysis types including for the H<sub>2</sub>O solutions revealed no apparent increase in number of etch pits as well as no apparent difference in the distribution of etch pit diameter between the electrolysis and control experiments. Fig. 8 shows such typically relation between total number and the diameter of the etch pit with constant DC 20 mA application for 168 h, which electrolysis type was "E1" in Table 1. The total number of pits for each diameter in this figure is the sum of them over 5 runs. In contrast, a marked difference in the distribution of etch pit diameter between electrolysis and control experiment was observed in the electrolysis types "E3" and "E5". Fig. 9 shows the result obtained from "E5" using LiOH/D<sub>2</sub>O solution and presents an apparent difference in the distribution between the electrolysis and control experiments even though the current pattern "20mA" was same as that of "E1". It was observed that the number density of the etch pit was larger than that in the outer area of O-ring. There exists no peak in the distribution for the control experiment, while a peak at ~10 µm is seen in the distribution of the electrolysis experiment. However, it should be pointed out that the difference in the distribution between the electrolysis and control experiment will not always appear in each run, even though the electrolysis was performed under the same experimental condition. A typical photomicrograph of the surface of the CR-39 chip used in one of the experiment of "E5" is given in Fig.11. Total 10 number of etch pits are seen in the figure, corresponding to an area 500x700 µm in one of the CR-39 chips.

More clear difference in distributions is given in Fig.10 for the current pattern "20mA/24h-R", corresponding to "E3". The highest peak is seen in the distribution for the electrolysis experiment at 8  $\mu$ m, which is near the value of that observed in Fig.9. The average density of the pit in each CR-39 chip for "E3" is 42/cm<sup>2</sup>. Of particular interest is the fact that total 30 pits were observed in a small area 1x1 mm in one of the CR-39 chips; the density is extremely large value of 3.0x10<sup>3</sup>/cm<sup>2</sup>.



Fig. 8 Distribution of etch pit diameter collected from 5 runs of electrolysis experiment "E1" and the corresponding 5 control experiments.



Fig. 9 Distribution of etch pit diameter collected from 5 runs of electrolysis experiment "E5" and the corresponding 5 control experiments.



Fig. 10 Distribution of etch pit diameter collected from 5 runs of electrolysis experiment "E3" and the corresponding 5 control experiments.



Fig. 11 Photomicrograph of a surface of the CR-39 chip obtained from the electrolysis "E5", providing a group of pits.

#### 3.3 Electrolysis of H<sub>2</sub>O solution

The anomalous distribution was observed for H<sub>2</sub>O solution as well as D<sub>2</sub>O solution. A marked difference in the total number of etch pit and in the distribution and between electrolysis and control experiments was observed in one out of 5 electrolysis types for H<sub>2</sub>O solution. Fig.12 provides the relation between total number and the diameter of the etch pit for electrolysis and control experiments using Li<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O solution under the current pattern "3-160mA-S", corresponding to "E7". The diameter ~7  $\mu$ m at the peak in the electrolysis distribution is near the value of the peaks observed in Fig.9 and 10 for the D<sub>2</sub>O solutions. Accordingly, most of the etch pit with diameter range 7-10  $\mu$ m seems to be produced by the electrolysis and to be independent on the kind of solution. The common key factors to increase number of the anomalous etch pit for the both solutions might be Ni film cathode, Li in the electrolyte solution and the long electrolysis time of 168 h.



Fig. 12 Distribution of etch pit diameter collected from 3 runs of electrolysis experiment "E7" and the corresponding 3 control experiments.



Fig. 13 Distribution of etch pit diameter collected from 5 runs of electrolysis experiment "E9" and the corresponding 5 control experiments.

## 3.4 Fluctuation of the number of etch pit produced in the control CR-39 chip

Total number of etch pit for each electrolysis type and that for the corresponding control experiment are compiled into Table 2. It should be pointed out that almost same number of etch pit was sometimes observed in outer area of O-ring as that in inner area of the chip after the electrolysis for the electrolysis types "E4", "E8" and "E9". Fig.13 shows typically the distribution obtained from the electrolysis type "E9", where the total number of etch pit in outer area reaches ~2/3 of that in the inner area. Similar characteristic was observed in the CR-39 chips used in the electrolysis type "E8", where the control chip gave the large number of total 91 etch pits as shown in Table 2. It was found that those chips used for "E4", "E8" and "E9" were obtained from the same lot of CR-39 sheet. Consequently, the relatively large fluctuation of the number of etch pit observed in the control chip is attributed to other phenomena than nuclear one. As the increase in number of etch pit is observed even for the control experiment, it seems that the CR-39 chip was sometimes damaged before the electrolysis and control experiments to have a considerable number of defects.

Table 2Total number of etch pit compiled for each type of experiment. The total number of<br/>etch pit in inner region of O-ring roughly equals to that in the outer region for<br/>electrolysis type E4, E8 and E9. These numbers are given in the red cells.

Solution Cathode		Current	Total	Total Number of etch pit		Total	
		pattern	electrolysis period	Electrolysis	Control	number of run	Lot
		20mA	168h	40 (E1)	31	5	a
Li <sub>2</sub> SO <sub>4</sub> / D <sub>2</sub> O	Ni	20mA/ 20min-R	200min	27 (E2)	20	5	b
		20mA/ 24h-R	168h	210 (E3)	35	5	b
		3-160mA-S	168h	56 (E4)	21	5	с
LiOH/	Ni	20mA	168h	79 (E5)	19	5	a
D <sub>2</sub> O	D <sub>2</sub> O	3-160mA-S	168h	51 (E6)	14	4	с
Li <sub>2</sub> SO <sub>4</sub> /	Ni	3-160mA-S	168h	201 (E7)	9	3	b
H <sub>2</sub> O	Au	3-160mA-S	168h	105 (E8)	91	5	С
Na <sub>2</sub> SO <sub>4</sub> / H <sub>2</sub> O	Ni	3~160mA-S	168h	66 (E9)	27	5	С
	Ni	20mA/ 24h-R	168h	27 (E10)	26	4	С
	Au	20mA/ 24h-R	168h	26 (E11)	32	4	С

# 4. Conclusion

Anomalous increase in number of etch pit was observed in 2 out of 6 and one out of 5 electrolysis experiments for  $D_2O$  and  $H_2O$  electrolyte solutions, respectively. The result has provided a possibility of LENR occurring on the Ni film cathode during the electrolysis. The common factors to increase number of the anomalous etch pit in the CR-39 chip might be Ni film cathode, Li in the electrolyte solution and the long electrolysis time. All the results might show a characteristic of LENR in the electrolysis that the reaction dose not always take place in every experiment but does occasionally under the same experimental condition.

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# Search for advanced simulation model of cascade vortices under

# beneath the electrode surface

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**Abstract**: In the cold fusion experiment, there observed vortex patterns on a thick Pd electrode surface during long-term electrolysis in 0.1M LiOD. To understand the peculiar phenomenon, we have proposed N-cycle model, which is composed of four sequential processes including the cold fusion (CF) reaction. In this study, a numerical simulation for the analysis of the vortex patterns was performed to elucidate the relation between the vortex formation and CF. Supposing that the hypothetical particles mass evolved due to CF reaction energy, two numerical simulations of the motion of the hypothetical particles mass were performed: Cellular automata (CA) and discretization method. Results of both simulations have shown generation of a vortex or a cascade of vortices, which were identical to those obtained experimentally. However, there still exist differences between the experimental and theoretical patterns in scale and form. Then, it motivates us to build a more sophisticated model. On the other hand, Miesch et al. performed the numerical simulation of the convection structure in the solar convection zone, where the appearance of vortex was deduced from the helioseismic data. The numerical simulation showed that the strong downflow was characterized by the helicity reversal near the base of the convection zone. Reviewing the numerical calculation method and the restricted conditions inspired us the important term of helicity change under the conservation, relaxation and transportation across a boundary.

Keyword: Computational fluid dynamics, Pd, Nuclear reaction cycle model, vortex, numerical simulation, cold fusion

## **1** Introduction

During long-term electrolysis for well annealed thick Pd rod (9.0 mm  $\phi$ ) in 0.1M LiOD, vortex pattern was observed<sup>1-2)</sup>. The morphology of the postelectrolysis electrodes revealed the two long faults without any cracks on the surface. Since the formation of this peculiar pattern of vortices can be highly plausible to the result of the Cold Fusion (CF) reaction, the precise mechanism of vortices formation must be elucidated in relation with the solid-state phenomenon accompanied with long-term evolution of deuterium in the Pd cathode in 0.1M LiOD. So far, an in-situ measurement of the solid -state properties of dilation, resistance and electrode potential revealed that the thick Pd electrode was composed of the core structure enveloped by the sub-surface layer. The latter exhibits non-equilibrium deuterium absorption/desorption reaction. Then, N-reaction cycle model proposed<sup>3-4</sup>) is composed of four sequential processes: in-taking and compression — triggering (the CF reaction) — scavenger. There the last process: scavenger shows the traces of vortex on the electrode surface as a consequence of the process continuation. In the last papers<sup>5-6)</sup>, the vortex pattern was successfully obtained in 2D space analyzing the dynamics of the hypothetical particles mass by a numerical simulation method: Lattice gas cellular automata (LGCA) numerical

simulation. Next we performed the numerical simulation using discretization method for 3D R dynamics of the hypothetical  $\frac{\Gamma}{\Gamma}$ particles mass (HPM) in the layer<sup>7)</sup>. However, sub-surface there still remained ambiguous in 2 the vortex and CF reaction relation. Under such а circumstance, we note that the rules physical which were established in natural phenomena might help us to make a model

lun No.	Current, mAcm <sup>-2</sup>	Pretreatment	
l st	0.05-40 40-500	Cast, 800°C anneal (10 <sup>-6</sup> Acid treatment	Torr)
nd	40	Polishing, Acid Evacuation, $D_2$ gas charge	treatment, e
Brd	40	Evacuation, Polishing treatment	g, Acid
lth	40	Evacuation, Polishing treatment	g, Acid

**Table 1** Experimental conditions of Exp.1.

incorporating the effect of magnetic field and matrix inhomogeneity. In this study, firstly, the experimental results of the morphology: vortex or vortex thread is presented in relation to preliminary PC simulations so far, and secondary to improve those PC simulations hydrodynamic dynamo action in the solar convection zone is briefly mentioned in terms of the dynamics coupled with the magnetic fields.

## 2 Experimental results of vortex for N-reaction cycle model

As shown in **Table 1** the electrolysis for deuterium absorption was conducted as follows; the electrode was removed from the cell and carefully re-installed four times during which the diameter of the electrode was measured at three positions (top, middle and bottom). During 1st run the dilation at the bottom end shows 7 % while those of 2nd - 4th runs the values at these positions approached asymptotically to 7.8 - 8.3 %.



**Figure 1** Vortex appeared on Pd electrode surface after long-term electrolysis in 0.1M LiOD (a), Duplicate of SEM picture (b).

Figure 1 shows a significant morphology of a thick rod Pd electrode observed on the surface after long-term electrolysis in 0.1M LiOD <sup>1-2, 8</sup>). It is not the substance adhered on the surface, but is a material on which the pattern was deeply impressed in a shape of a ditch. This is the morphology which formation mechanism will be elucidated in the present study. Alternately, we have investigated the microscopic structural change of Pd at absorption/desorption of deuterium by electrolysis as a fundamental study of deuterium absorption behavior. Although a precise description is not shown in this paper, structural change of discrimination of the sub-surface and the bulk was developed by the deformation during prolonged deuterium absorption<sup>9-10</sup>. The above result with respect to the structural change of Pd must be useful in the elucidation of intake of reactant followed by compression of N-cycle model (see Ref10)).

# **3 3D turbulent structure of the solar convection zone**

It is known that the terrestrial earth phenomena (e.g., magnetic storm) are influenced by the intensity of solar flare magnetic and re-connection, whereas an electromagnetic fluid motion the solar. in especially the convection zone is closely related to changes in the activities. large-scale For phenomena sun spots exhibit the fluctuations as a function of date and latitude. This



**Figure 2** The instantaneous snapshot of the radial velocity on horizontal surface located in the upper convection zone; the equator is indicated by a dashed line. The right shows a blowup of a  $50^{\circ}$  x  $60^{\circ}$  segment in latitude and longitude, as indicated by solid lines (M. Miesch et al.: APJ **532**, 593(2000)).

phenomenon was interpreted through the solar outer layer dynamo action. The so-called solar dynamo action involves the generation of magnetic fields by the turbulence in the deep convection zone <sup>11</sup>. (See the interaction between magnetic field and fluid velocity Appendix 1.) The above macroscopic simulation has advanced to more consistent and sophisticated simulation due to recent PC performance improvement.

On the other hand, the solar convection zone is highly turbulent giving characteristic Reynolds number of order of  $10^{12}$  and then more realistic approach of mean-field approximation is amenable to the substantial solar convection hydrodynamics. There have been reported the progressive magnetohydrodynamics (MHD) simulations, e.g., anelastic approximation. Recently, 3D non-linear simulation

of the spherical convection cell possessing turbulent nature and an axial rotation explored the strong downward turbulent convection into a base of the convection  $zone^{12}$  (see Appendix 2). The simulation model and parameters (global scale solar convection) were adopted to be adequately resolved for convection penetration. Figure 2 shows the structure of convection in the turbulent solution where the downflow network (blue) surrounded by the upflows (red to yellow). As shown in the snapshot, narrower downflow lanes surrounded by upflows indicates a downflow ring associated with a vortex sheath. Considering the net non-linear energy and momentum transport within the convection zone the downflow is contributed to an efficient poleward momentum transport in the base. Furthermore, comparing the radial velocity of the upper and that in the overshoot region (the base) it was demonstrated that the strong, vortical downflows and plumes intermit at the middle and link to upflows. Figure 3 (b) shows the schematic of the downflows linked to the upflow where solid lines and arrows indicate the sense of the flows and the dotted line the radial direction for comparison. The coherent and midlatitude downflow is more prominent because of the strong, plumelike downflows in turbulent case (high Rossby number). In Fig. 3 (a), by Coriolis force both turbulent and laminar produced spinup downflowing, which was rendered at the stable boundary or density stratification. Hence, the helicity reversal (see Appendix 3) is seen near the base of the convection zone.



# 4 Mechanism of vortices formation in Pd electrodes deduced from N-cycle model

First of all, it is remarked that vortex appearances are caused by the CF reaction, during which the mobile hypothetical particles mass (HPM) assumed in the previous paper<sup>9)</sup> could trail through the sub-surface layer resulting in the experimentally obtained vortex (Fig. 1). So far, we have claimed that N-cycle model could comprehensively explain the phenomena occurring on Pd during long-term electrolysis in 0.1M LiOD, where the model is composed of four sequential processes: in-taking and compression – triggering (the CF reaction) – scavenger. In due course of N-reaction cycle process there were shown alternative two mechanisms of HPM motions in the scavenger. One is ① of Fig. 4 (same as previous report<sup>13)</sup>): to electrode surface and another ②: to neighboring reaction vessel. In the upper of Fig. 4① it shows the locus of the occasional particle flow on the Pd surface, which was identical to the observed vortex pattern. By comparing the vortex pattern with the experimentally obtained one the axis of the motion leaned due to the interface's magnetic field<sup>13</sup>. The lower of Fig. 4② shows the continuous flow of HPM from a vessel to a neighboring one and many



vortices are evolved behind obstacles and a cascade of vortices is seen. Here, assuming that a vortex pattern is attributed to a locus of a vortex-thread motion, it has been suspected that single HPM also has the vortex pattern at the electrode surface. Furthermore, a reasonable inference leads us to conclude that the vortex occurred occasionally, while the cascade vortices: vortex-thread moves under beneath the electrode surface. This view might not be inconsistent with the irregularity of the sub-surface layer under an annealing at 1100 C<sup>o 14)</sup>.

On this point, we have drawn the concept of equilibrium and non-equilibrium states in Fig. 5 to solve the problem. The structure of 3D sub-surface layer and the transportation of momentum and mass are schematically shown, where the explosive energy is transferred from the reaction site. There appeared to move HPM through the thick piping. The vortex threads themselves (see also Fig.  $4^{\circ}$ ) may enable to move instead of HPM. However, there exists ambiguous on how large scale of vortices evolved. Then it necessitates to present accurate model with respect to energy and mass transfer, i.e., dynamics in the sub-surface layer. Momentum and mass transportation in the sub-surface might be accomplished in the following ways; it holds forward transportation rate and backward one equal implying an equilibrium state, and otherwise net transportation rate with either direction continues implying a non-equilibrium state. In the latter case, as shown in Fig. 5, the net flow encounters by Rayleigh–Taylor's instability resulting in an evolution of vortices.



**Figure 5** Motion of HPM, transportation phenomenon and evolved cascade vortices in the sub-surface layer.



**Figure 6** LGCA simulation result of vortices evolved behind plates on 4640x1152 grids at Time step=100000. This work is directed toward the advanced LGCA simulation model: the cascade of the vortices (vortex-thread).

Hereafter, we describe our results of two numerical simulations: Cellular automata (CA) and discretization method, both of which generated a vortex or a cascade of vortex. As shown in Fig. 6, the CA simulation result indicates the stationary flow creates two vortices behind two flat plates (shown by arrows in Fig. 6). Next approach is addressed to n-cascade of vortices for dynamics of vortex-threads. Also, simulation domain was a



**Figure 7** Time evolution of streamlines of the particles mass ejected at the inner surface toward the electrode surface in the sub-surface layer. Inset (a) shows four particles possessing initial velocity  $v_0$  on the quadrupoles of the ejection disk and that of (b) shows reached particles possessing velocity v.

simple 2D-rectangular, which will be advanced to 3D-domain possessing curved boundaries and branching. The latter simulation is numerical calculation to solve Maxwell's equation and fluid equation, and subsequently electromagnetic interaction is involved in HPM transfer calculation. As shown in Fig. 7, a field, though is not precise, an ejected HPM (Inset a) exhibited a helical stream line showing a vortex on the electrode surface (Inset b). Thus, under the influence of 3D magnetic structure arrayed HPM moves in the sub-surface layer resulting in a peculiar pattern at the surface. Hence, to substantiate more detailed motions of HPM in the sub-surface layer, we necessitate precise geometry and microstructure involving precipitates and /or voids.

# 5 Model of HPM motion in the sub-surface layer deduced from sun's investigation

As the average flow structure of the solar convection zone mentioned above is elucidated in conjunction with the observation by the solar seismology. Although the obtained convection pattern was large scale, the nature of the strong downflows was adequately clarified in the simulation. Based on the investigations of the sun's HMD

	Characteristics of the sun	Characteristics deduced from	
	Characteristics of the sun	N-cycle model	
		Cylinder (large scale):	
	Thin rotating spherical shell	core/sub-surface layer/electrolyte	
	(plasma); size radius 700000km	(ionic plasma); size 9-20mm $\phi$ ,	
Structure (size)	$(10^2 \text{ times larger than the earth})$	length 43mm	
	Several zones including the	Microstructure: precipitates	
	convection zone	and/or cluster of voids; size	
		several 10 μm	
Content	Hydrogen (75%), He (25%)	HPM, deuterium, Pd, electrolyte	
	Global: Toroidal 1-10kG		
Magnatia field	Local: Poloidal (Pole) several G,	Global: 55G	
Magnetic neiu	sun spot 10 <sup>3</sup> G (base of convection	Local: not yet known	
	10 <sup>4</sup> G)		
Magnetic polarity change	22 year (periodic)		
Course of Monarchie C		Supplied current flow in the	
Source of Magnetic flux	Base of the convection zone	sub-surface layer	

**Table 2** The characteristics of the sun and CF experiments

and our CF experiments, we could draw the following deduction.

Table 2 shows a comparison of the characteristics between the sun and CF

experiments. The difference of the scale is insignificant; however, it is not yet known that CF experimental system possesses the dynamics like the sun's axisymmentic rotation. At this stage, we prefer the description of following items.

- HPM (hypothetical particles mass): its physical nature will be apparent in the next issue.
- Re-generation or modification of magnetic field through a dynamo process: preliminary calculation was done using ANSYS<sup>15</sup>.
- If we accept much more developed idea, the gas phase (babble) or electrolyte over the sub-surface layer probably corresponds to the arch filaments (emerging magnetic flux) closely related with the dynamics of the convection zone.

## 6 Conclusion

We described comprehensive model to analyze the phenomena occurring during CF experimental conductance. To reproduce the peculiar vortex pattern, we performed two numerical simulations: cellular automata and the discretization method. Although both methods succeeded in obtaining a vortex pattern and a cascade of vortices, the differences between the experimental and theoretical patterns became apparent. Meanwhile we noted that the magnetic configuration under beneath the surface layer plays an important role in the simulation of the motion of HPM. To inspire new idea we investigated the numerical simulation methods applied to the convective flow in the solar convection zone. As a result, the helicity change is found to be an important item to simulate the dynamics of HPM in the sub-surface layer. Furthermore, the magnetic structure can be calculated through the MHD dynamo process.

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#### Appendix 1

In kinetic dynamo theory of electronically conducting fluids the induction or creation of magnetic field is described by the induction of magnetic field:

$$\begin{split} &\frac{\partial B}{\partial t} = \nabla \times \left( v \times B \right) + \eta \, \nabla^2 B \\ &B \text{ magnetic field, } v \text{ plasuma velocity} \\ &\eta = \frac{1}{\sigma \, \mu_0{}^{_{\mathrm{M}}}} \quad resistance \ (magnetic \ difusivity) \\ &\sigma \ electric \ conductivity, \ \ \mu_0{}^{_{\mathrm{M}}} \ permeability \ of \ vaccum \\ &Also, mean - \ field \ dynamo \ mod \ el \ is \ used. \end{split}$$

The set of MHD equation is a combination of the Navier-Stokes equation and Maxwell's equations. For magnetic field of fluid plasma MHD equation consists of the basic equation of motion, Ampere's low (neglecting displacement current) and a temperature evolution equation. Also, fluid is almost neutral and convective current due to flowing ions is negligible.

## Appendix 2

The helioseismic investigation by Solar Oscillation Investigation-Michelson Doppler Image (SOI/MDI) experiments revealed internal dynamics of the convection zone. The following web site shows the zonal convective flows and temperature change in close accord with the upper convection zone.

http://soi.stanford.edu/papers/One.year/figure\_09.gif .

 $\rightarrow$ 

#### Appendix 3

- Helicity is :fluid helicity. It is a degree of movement of the solid which rotates around the advance axis of fluid.
- Rotation sees from the object front, and it is a negative value, if it is a clockwise rotation and is a positive value and a counterclockwise rotation.

 $\rightarrow$ 

 $H = \int u \bullet (\nabla \times u) d^3 r$ 

- The example of the above figure, number of rotations / advance distance (a mark is direction of rotation)
- Similarly, it is magnetic helicity. The involved degree is expressed, a twist of a line of magnetic force or when being involved.  $H = \begin{bmatrix} A & B & d^{3}r \end{bmatrix}$

# Theoretical Study of Nuclear Reactions in Solids using Bose-Einstein Condensation Model

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#### Abstract

In our previous work on Bose-Einstein condensation (BEC) approach to the theoretical interpretation of cold fusion [1], we estimated the transition temperature of BEC in palladium deuteride. It was based on the Kim's work [2] by using equivalent linear two-body method to the many-body problems of charged bosons trapped in an ion trap. In the recent work of Kim et al.[3], they have expanded their theory in order to explain the Rossi's experiment [4]. In this study, Kim's theory is verified and a possible algorism for the numerical calculations are proposed.

#### 1. Introduction

Recently, Y.E.Kim et al. tried to explain the results of Rossi's experiment by applying the ELTB (equivalent linear two-body) method to the bosons trapped to the harmonic potential [3]. Here, Kim's theory is briefly introduced. They considered a mixture of different two species of positive charged bosons. In this study, we verified Kim's method and considered how to perform the numerical calculation. The Hamiltonian for this many-particle system is written as

$$H = H_1 + H_2$$
 , (1)

where  $H_n$  is the Hamiltonian for specie n and defined as

$$H_n = \sum_{i=1}^{M_n} \left\{ -\frac{\hbar^2}{2m_n} \nabla_{ni}^2 + \frac{1}{2} m_n \omega_n \mathbf{r}_{ni}^2 + Z_n e^2 \int d\mathbf{r}' \frac{Z_1 N_1(\mathbf{r}') + Z_2 N_2(\mathbf{r}')}{|\mathbf{r}_{ni} - \mathbf{r}'|} \right\} .$$
(2)

In this equation,  $\mathbf{r}_{ni}$  means the position of the i-th particle in specie n.  $Z_n$  and  $N_n$  mean charge and number density of specie n, respectively.  $M_n$  is the number of the particle in specie n. The Schrödinger equation for these system can be written as

$$H\Psi = E\Psi \quad , \tag{3}$$

where many-body wave function  $\Psi$  is separated into the product of two functions as

$$\Psi(\mathbf{r}_{11}, \mathbf{r}_{12}, \cdots \mathbf{r}_{1M_1}; \ \mathbf{r}_{21}, \mathbf{r}_{21}, \mathbf{r}_{21}, \cdots \mathbf{r}_{2M_2}) = \Phi_1(\mathbf{r}_{11}, \mathbf{r}_{12}, \cdots \mathbf{r}_{1M_1}) \Phi_2(\mathbf{r}_{21}, \mathbf{r}_{21}, \cdots \mathbf{r}_{2M_2}) ,$$
(4)

where the function  $\Phi_n$  depends on the positions of the particles in species n. Using this, the variational form is written as

$$E = \frac{\langle \Psi | H | \Psi \rangle}{\langle \Psi | \Psi \rangle} = \sum_{n=1}^{2} \frac{\langle \Phi_n | H_n | \Phi_n \rangle}{\langle \Phi_n | \Phi_n \rangle} = \sum_{n=1}^{2} \varepsilon_n .$$
(5)

It is well known that the bound state of this system can be obtained by minimizing the variational form with respect to the trial function. Minimizing E with respect to  $\Phi_n^*$ , we obtain

$$\delta E_n = \frac{\langle \delta \Phi_n | (H_n - \varepsilon_n) | \Phi_n \rangle}{\langle \Phi_n | \Phi_n \rangle} = 0 , \qquad (6)$$

and this gives equations for each case as

$$H_n \Phi_n = \varepsilon_n \Phi_n \qquad (n = 1, 2) . \tag{7}$$

The expanded form of  $\Phi_n$  by hyperspherical harmonics  $U_{k\nu}(\Omega)$  is written as

$$\Phi_n = \rho_n^{-(3M_n - 1)/2} \sum_{k\nu} \phi_{nk\mu}(\rho_n) U_{k\nu}(\Omega_n) , \qquad (8)$$

where  $\Omega_n$  is a set of  $(3N_n - 1)$  angles and  $\rho_n$  is defined as

$$\rho_n = \left(\sum_{i=1}^{M_n} \mathbf{r}_{ni}^2\right)^{1/2} \ . \tag{9}$$

The hyperspherical harmonics is a generalized spherical harmonics for n-dimensional space and satisfies

$$\int d\Omega_n U_{k\nu}^*(\Omega_n) U_{k'\nu'}(\Omega_n) = \delta_{kk'} \delta_{\nu\nu'} .$$
(10)

Using the expanded form we obtain

$$\sum_{i=1}^{N_n} \nabla_{ni}^2 \Phi_n = \rho_n^{-(3N_n-2)/2} \sum_{k\nu} \left\{ \frac{d^2}{d\rho_n^2} - \frac{L_{nk}(L_{nk}+1)}{\rho_n^2} \right\} \phi_{nk\mu}(\rho_n) U_{k\nu}(\Omega_n) , \qquad (11)$$

where  $L_{nk}$  is defined as

$$L_{nk} = k + \frac{3M_n - 3}{2} . (12)$$

Multiplying  $U_{00}^*$  from left side of Schrödinger eq.(7) and integrating over all solid angles, we obtain

$$\int d\Omega_n U_{00}^*(\Omega_n) (H_n - \varepsilon_n) \Phi_n = 0 .$$
(13)

This gives separated two equations as

$$h_n \phi_{n00} = \varepsilon_n \phi_{n00} \qquad (n = 1, 2) , \qquad (14)$$

where the Hamiltonian  $h_n$  is written as

$$h_n = -\frac{\hbar^2}{2m_n} \left\{ \frac{d^2}{d\rho_n^2} - \frac{L_{nk}(L_{nk}+1)}{\rho_n^2} \right\} + \frac{1}{2}m_n\omega_n^2\rho_n^2 + V_n(\rho_n) .$$
(15)

The potential  $V_n(\rho_n)$  in this Hamiltonian is calculated by

$$V_n(\rho_n) = \frac{1}{\int d\mathbf{R}_n \int d\Omega_n} \int d\mathbf{R}_n \int d\Omega_n \sum_{i=1}^{M_n} Z_n e^2 \int d\mathbf{r}' \frac{Z_1 N_1(\mathbf{r}') + Z_2 N_2(\mathbf{r}')}{|\mathbf{r}_{ni} - \mathbf{r}'|} , \quad (16)$$

where the gravity center  $\mathbf{R}_n$  is defined as

$$\mathbf{R}_n = \frac{1}{M_n} \sum_{i=1}^{M_n} \mathbf{r}_{ni} \ . \tag{17}$$

In deriving eqs.(14-15), spherical approximation is used. It is effective around the bottom of the harmonic potential.

#### 2. Possible algorism

Here, a possible algorism for the numerical calculation is shown. Firstly, we should consider how to calculate the potential term. Using eq.(16), the term is rewritten as

$$V_n(\rho_n) = \frac{M_n}{2\pi\Gamma\left(\frac{3}{2}\right)} \frac{\Gamma\left(\frac{3M_n}{2}\right)}{\Gamma\left(\frac{3(M_n-1)}{2}\right)} \frac{1}{\rho_n^3} \int_0^{\rho_n} dr r^2 \left(1 - \frac{r^2}{\rho_n^2}\right)^{(3M_n-5)/2} I(r) , \qquad (18)$$

where the function I(r) is defined as

$$I(r) = \frac{16\pi^2}{r} \left\{ \int_0^r dr' r'^2 q(r') + r \int_r^\infty dr' r' q(r') \right\} .$$
(19)

In this equation, the function q(r) means total charge density including two species of charged bosons, which is written as

$$q(r) = Z_1 e N_1(r) + Z_2 e N_2(r) . (20)$$

Seeing eqs.(18-20), we can easily understand that the potential  $V_n$  depend on the densities  $N_1$  and  $N_2$ . This means that eq.(14) for one specie links to the other through the potential term. Adding to this difficulty, the density  $N_n$  should be calculated from the wave function  $\Phi_n$ . However, the wave function is the solution of eq.(14) whose Hamiltonian  $h_n$  depends on the densities.

Secondly, we should consider how to overcome the difficulties described above. In this case, the iterative process is used for the numerical calculations. For example, if the initial potentials are assumed, then we can calculate the first wave functions. After the first step, we can obtain the new density and new potential. Then the new wave function is obtained as a numerical solution of eq.(14) and so on. If the iteration converges in enough accuracy, they are self-consistent solutions.

A possible algorism proposed in this paper is illustrated in Fig.1.



Fig.1 Iterative process to calculate the state of the many-particle system including two species of charged bosons following the Kim's theory

One of the most appropriate candidate to use for the convergence criterion in the iterative process is the eigenvalues in eq.(14), because they are not functions of  $\rho_n$  but constant values.

## 3. Results and Discussions

We used ground state wave function of the harmonic potential to make initial densities  $N_1$  and  $N_2$ . However, eigenvalues  $\varepsilon_1$  and  $\varepsilon_2$  were not converged in the iterative process. In this case, the maximum points of the initial densities  $N_1$  and  $N_2$  were overlapped at the bottom of the harmonic potential. This means that the positive charge density becomes excessively large at the bottom of the harmonic potential. We estimate that the excess charge causes the divergence.

In order to avoid the large overlap, an additional condition should be included in

the calculations. If the wave functions for n = 1 and 2 are orthogonalized with each other, excess charge at the bottom of the potential can be avoided. Remembering the shapes of the ground state and the first excited state wave functions for the harmonic potential, we can easily understand it.

## 4. Conclusions

We have tried to calculate the many-body wave function for the system including two species of charged bosons by using Kim's theory [3]. We have shown that the iterative calculation should be used. However, starting from the ground state particle densities for both species, the convergence in the iterative process cannot be obtained. We have pointed out that the orthogonality condition should be included in the theory. In near future, we will do the iterative calculation with the orthogonality condition.

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# **Kinetic Reaction Energy of Cold Fusion**

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Some essential consequences of the TSC (tetrahedral symmetric condensate) theory on underlying quantum mechanical physics of nuclear fusion reactions in condensed matter are discussed. To attain enhanced cold fusion rate in condensed matter at room temperature, mean kinetic energy of mutual (relative) d-d motion in dynamically trapped 4D-cluster should be elevated as same as the case of hot plasma fusion (typically 10 keV).

The mutually high kinetic energy is attained in a deep dynamic (time-dependent) trapping potential of deuteron-cluster in condensed matter. This is required to satisfy the Heisenberg Uncertainty Principle (HUP) of quantum mechanics. Trapped D-cluster in very microscopic domain (ca. 20 fm size) behaves like a gas-molecule motion at room temperature outside the trapping potential, so that "high temperature" of gross condensed matter is NOT necessarily required.

Procedures for quantitative fusion rate estimations by Fermi's golden rule are explained in combination of the multi-body strong interaction based on the one-pion-exchange potential (OPEP) and the Coulomb barrier penetration in the QM-Langevin equation analysis. Time-dependent total energy of multi-body system settles in local minimum under the constraint of HUP. Using the variational method, mean kinetic energy (expectation value) of d-d pair trapped in the TSC potential was time-dependently estimated with the heavy mass electronic quasi-particle expansion theory method, which also gave mean kinetic energies for D<sub>2</sub> molecule and muonic d-d molecule for the comparison purpose.

Keywords: Kinetic reaction energy, hot fusion, cold fusion, D-cluster, confinement in microscopic space, TSC theory, variational method

#### 1. Introduction

The review of TSC (tetrahedral symmetric condensate) theory for condensed matter nuclear effects including cold fusion has been recently published [1] in the open access electronic journal of JCMNS. Some detailed explanation of TSC theory as shown by quantum mechanical (QM) Langevin equation was described in two papers in ACS NET books [2, 3]. By reconstructing key results obtained by two papers [2, 3], the physical principle of cold fusion is explained in this paper in comparison with the principles of hot plasma fusion and the muon-catalyzed d-d fusion. We conclude that the principles of nuclear fusion reactions are analogous in each others in the view of reaction kinetic energy for mutual d-d motion in the confined space, although hot fusion requires very large scale devices as magnetic torus bin (as ITER), while cold fusion will be attained in an extremely small space of dynamic cluster trapping potential as TSC-minimum state.

The essence of physical principle is as follows: The hot plasma DD (DT) fusion is attained by the random collision process of 10-100keV relative kinetic energy of d-d (d-t) interactions under the plasma confinement by very large scale magnetic bin as ITER machine. The cold fusion is expected to happen by relatively high (as high as 10keV) reaction kinetic energy of mutual d-d
motion of D-cluster as a TBEC (transitory Bose-Einstein condensate) state as dynamically confined in very microscopic trapping potential spaces (as small as 20 fm diameter sphere) of condensed matter. It is the 4D/TSC-minimum state, as shown in the present paper (already implicitly shown in references 2 and 3). Hot plasma fusion will realize "long-time-lasting" (or hopefully steady) high reaction rates of random two-body d-d (or d-t) reactions which produce high intensity neutrons as well as charged particles, while the cold fusion by 4D/TSC will take place in a very short time-interval (about  $2x10^{-20}$  s) of the final stage of 4D/TSC condensation motion under the dynamic ordering/constraint conditions of metal-deuterium condensed matter systems to generate predominantly 4D simultaneous multi-body fusion with <sup>4</sup>He dominant products with negligibly small amount of neutron and gamma-ray emissions. Two body d-d reactions are negligible in cold fusion.

## 2. Fusion Reaction Rates

Fusion rate of hot plasma two-body d-d reaction is defined by Eq.(1), as well known.

$$< \text{Macroscopic Fusion Rate} > = < N_d (E_k)^2 v \sigma_{dd} (E_k) >$$
(1)

Here,  $N_d(E_k)$  is the deuteron density with Maxwell-Boltzmann energy distribution with relative kinetic energy  $E_k$ . The relative velocity of colliding d-d pair is v. The  $\sigma_{dd}(E_k)$  is the d-d fusion cross section as given by using the astrophysical S factor:

$$\sigma_{dd} = (S(E_k)/E_k)exp(-\Gamma_{dd})$$
<sup>(2)</sup>

 $\Gamma_{dd}$  is the Gamow factor as given by the WKB approximation. As the cross section is steeply increasing function as  $E_k$  increases (up to about 1 MeV for d-d reaction), macroscopic fusion rate of hot plasma with 10-100 keV temperature has a Gamow-Teller peak at  $E_0 >>$  plasma temperature, by statistically averaged value of reaction rate over energy (written as bracket of Eq.(1)). So the mean kinetic reaction energy of hot fusion is larger than averaged mutual kinetic energy of deuterons, for plasma conditions under research with MCF and ICF devices.

In contrast, microscopic fusion rate per D-cluster is defined by Eq.(3) (see Ref.1, 2, 3, 4) based on Fermi's first golden rule [4].

$$\lambda_{nd} = \frac{2}{\hbar} \langle W \rangle P_{nd}(r_0) = 3.04 \times 10^{21} P_{nd}(r_0) \langle W \rangle$$
(3)

Here,  $\langle W \rangle$  is the averaged value (with inter-nuclear wave-functions, initial and final state, weight) of imaginary part of nuclear optical potential for strong nuclear interaction of multi-body deuteron cluster simultaneous fusion reaction, and  $P_{nd}(r_0)$  is the barrier (Coulomb barrier) penetration probability (equivalent to the squared outer-nuclear wave-function weight within the strong interaction domain, as given by the Born-Oppenheimer approximation) of D-cluster to the interaction surface (r0) of the fusion strong interaction domain (about 5fm for d-d fusion).  $\langle W \rangle$  values for D-cluster (n=2, 3, 4, 6) are empirically estimated by extrapolation of S-values for known reactions (p-d, d-d and d-t) to higher *PEF* (pion exchange force of fusion) values of simultaneous D-cluster fusion, using the PEF definition by the one-pion exchange potential (OPEP) [4],

$$V_{OPEP} (x) = v_0 \cdot (\vec{\tau}_1 \cdot \vec{\tau}_2) \left\{ \vec{\sigma}_1 \cdot \vec{\sigma}_2 + (1 + \frac{3}{x} + \frac{3}{x^2}) S_{12} \right\} \frac{\exp(-x)}{x}$$
(4)

with  $x = \frac{m_{\pi}c}{\hbar}r = \frac{r}{1.43}[fm]$  and 1.43 fm is the Compton wave length of charged pion, and spin (sigma-vector) and tensor (*S*<sub>12</sub>) terms in bracket. The Yukawa potential is given by exp(-x)/x. The product of isospin operator  $\vec{\tau_1} \cdot \vec{\tau_2}$  has eigenvalue -3 for n-p nucleon interaction, and OPEP becomes attractive (fusion reaction case), while it does +1 for n-n or p-p nucleon interaction to become repulsive. *PEF* = *I* is defined as relative unit of attractive force as,

$$\left\langle OnePEF \right\rangle = -\frac{\partial \left\langle V_{OPEP}(x) \right\rangle_{\tau,\sigma}}{\partial r} = -\frac{1}{1.43} \frac{\partial \left\langle V_{OPEP}(x) \right\rangle_{\tau,\sigma}}{\partial x}$$
(5)

And we define PEF = 0 for the repulsive n-n and p-p nucleon interactions.



## D-D Fusion: Strong Force vs. Coulomb Force

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Fig.1: Yukawa-type strong force for d-d fusion (PEF = 2), compared with repulsive Coulomb force between deuterons

For the n-p nucleon interaction, mean Yukawa force is obtained as,

$$\langle YukawaForc \ e \rangle = -\frac{197.32}{(R-r_0)} (\frac{1}{R-r_0} + \frac{1}{1.41}) \exp(-(R-r_0)/1.41)$$
 (6)

Yukawa force for d-d fusion reaction is drawn in Fig.1, in comparison with Coulomb repulsive force.

 $\langle W \rangle$  values extrapolated for 4D fusion (*PEF=12*) is shown in Table-1 together with that for d-d (*PEF=2*) reaction and other key values.

Molecule	Rdd=Rgs (pm)	Pnd; B-Factor	< <i>W</i> >(MeV)	$\lambda 2d$ (f/s)	$\lambda$ 4d (f/s)			
D2	74.1	$1.0 \times 10^{-85}$	0.008	2.4x10 <sup>-66</sup>				
dde*(2,2)	21.8	$1.3 \times 10^{-46}$	0.008	3.16x10 <sup>-27</sup>				
$\mu$ dd	0.805	1.0x10 <sup>-9</sup>	0.008	$2.4 \times 10^{10}$				
4D/TSC-min	0.021	1.98x10 <sup>-3</sup>	62		$3.7 \times 10^{20}$			

Table-1: Key physical values for cold fusions by molecules and 4D/TSC [2, 3]

Attention: fusion rates are for assumed steady states of molecules and clusters, and their life-times should be taken into account for actual fusion rates for dynamic states.

To calculate barrier factors (time-dependent) for the 4D/TSC condensation motion, analyses by the QM-Langevin equation were successful [2, 3]. No quantitative theories have been proposed for predicting drastically enhanced barrier penetration probabilities for two body d-d fusion, in D<sub>2</sub> type, in condensed matter at room temperature, except for the case of muonic dd molecule (the muon catalyzed fusion). The TSC theory predicts the TBEC (transitory Bose-Einstein condensate) state for 4D-cluster condensation motion, under the condition that 4 electron wave-functions and 4 deuteron wave-functions may orthogonally couple to make a Platonic symmetry in 3 dimensional domain with the ordering/constraint condition of metal deuteride lattice or surface defect sites. Once a 4D/TSC (t=0) state forms in sub-nano-meter scale site, it may condense very rapidly in a fs  $(1.4 \times 10^{-15} \text{ seconds by Langevin equation analysis})$  to be very small charge neutral entity (about 20) fm diameter) to cause 100% 4D simultaneous fusion in a very short  $(2x10^{-20} \text{ s})$  time-interval of its final stage of condensation motion [2]. We have defined the cluster fusion rate as fusion event per one 4D/TSC generation, by time-integration of varying barrier factors. Using the HMEQPET (heavy mass electronic quasi-particle expansion theory) method [2, 3], one-to-one conversion is possible from a condensation-time of TSC to a mutual d-d distance  $R_{dd}$  of HMEQPET molecule dde\*(m,2) with continuous equivalent mass of EQP(electronic quasi-particle). This was a pure mathematical trick, not proposing heavy electron mass Cooper pair, to implement numerical calculation for time-dependent barrier factors of TSC. As the Gamow integral for barrier penetration is easily calculated for a HMEQPET dde(m,2) virtual molecule, we have obtained [2] the convertible relation between condensation time,  $R_{dd}$  and barrier factor as shown in Table-2.

As shown in Table-1, muonic dd molecule has a very large microscopic fusion rate as  $2.4 \times 10^{10}$  f/s. This very large fusion rate results in a 100% d-d fusion in about 100ps. The life time of muon is 2,200ps. Therefore, liberated muon by a d-d fusion of ddµ molecule may be trapped to make new ddµ molecule making the second d-d fusion. Thus, several chain reactions of d-d fusion may happen within the muon life time. In the case of 4D/TSC-min, 100% 4D fusion may take place in  $2 \times 10^{-20}$  s because of very large "steady state fusion rate"  $3.7 \times 10^{20}$  f/s. The end state of 4D/TSC condensation, as calculated by the Langevin equation analysis (Fi.g.2), may realize such condition.

## 3. Kinetic Reaction Energy of Cold Fusion

To calculate mean relative kinetic energy of d-d pair in dde\* type molecule, which may include  $D_2$  molecule, ddµ molecule and arbitrary virtual dde\*(m, 2) EQPET molecules, we have applied the QM variational method as shown in Fig.3.

We have assumed that local energy minimum state settles in TSC system at any time of its condensation motion, and applied the QM variational method using Gaussian type wave-function. Corresponding QM Langevin equation and TSC trapping potential (time dependent) are shown in Eq.(7) and Eq.(8). The numerical results of 4D/TSC condensation motion is drawn in Fig.2. By knowing the one-to-one relation between TSC condensation time and  $R_{dd}$  d-d distance, we have made the comparison table (Table-3) for relating  $R_{dd}$  values and mean kinetic energy values using the results of the variational calculations.

$$6m_{d} \frac{d^{2}\langle R_{dd} \rangle}{dt^{2}} = -\frac{11.85}{\langle R_{dd} \rangle^{2}} - 6\frac{\partial V_{s}(\langle R_{dd} \rangle; m, Z)}{\partial \langle R_{dd} \rangle} + 6.6 \left\langle \frac{(R'-R_{dd})^{2}}{R_{dd}} \right\rangle$$
(7)

$$V_{tsc}(R':R_{dd}(t)) = -\frac{11.85}{R_{dd}(t)} + 6V_s(R_{dd}(t);m,Z) + 2.2\frac{|R'-R_{dd}(t)|^3}{[R_{dd}(t)]^4}$$
(8)

Table-2: Time dependent barrier factors  $P_{nd}$  and d-d distances,  $R_{dd}$  calculated by Langevin equation and HMEQPET method

C E	One-to-one rela Barrier factor, w	tion holds between th hich can be approxim	ne d-d distance an nately given by the	d the time-depende HMEQPET model	ent I.
	Elapsed Time (fs)	R <sub>dd</sub> (pm)	P <sub>2d</sub> : 2D barrier facotor	P <sub>4d</sub> : 4D barrier factor	
	0	74.1 (D <sub>2</sub> molecule)	1.00E-85	1.00E-170	
	1.259	21.8 (dde*(2,2); Cooper pair	1.30E-46	1.69E-92	
	1.342	10.3	2.16E-32	4.67E-64	
	1.3805	4.12	9.38E-21	8.79E-41	
	1.3920	2.06	6.89E-15	4.75E-29	
	1.3970	1.03	9.69E-11	9.40E-21	
	1.39805	0.805 (muon-dd molecule)	1.00E-9	1.00E-18	
	1.39960	0.412	9.40E-7	2.16E-13	
	1.40027	0.206	3.35E-5	1.12E-9	
	1.40047	0.103	1.43E-3	2.05E-6	
	1.40062	0.0412	1.05E-2	1.12E-4	
	1.40070	0.0206 (TSC-min)	4.44E-2	1.98E-3	

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Fig.2: Mean d-d distance and mean deuteron kinetic energy of 4D/TSC cluster as a function of condensation time, calculated by QM Langevin equation [2, 3].



Fig.3: Variational method for estimating eigenvalues of dde\* molecules; ground state d-d distance, mean kinetic energy, etc.

ACS2010	Mean Relative Kinetic Energy								
HMEQPET molecule parameters									
Molecule	b0 (pm)	Rmin (pm)	Vs-min (keV)	Ed-d (keV)	Rgs (pm)	Egs (keV)			
D2	22	70	-0.03782	0.00268	76.69	-0.03514			
dde*(2,2)	4.5	19.3	-0.1804	0.01013	21.82	-0.17027			
dde*(5,2)	1.9	7.6	-0.4509	0.0208	8.72	-0.43007			
dde*(10,2)	0.90	3.8	-0.9019	0.0418	4.36	-0.86012			
dde*(20,2)	0.45	1.9	-1.8039	0.0837	2.18	-1.7202			
dde*(50,2)	0.18	0.76	-4.5097	0.2094	0.873	-4.3003			
dde*(100,2)	0.09	0.38	-9.0194	0.4196	0.436	-8.5998			
dde*(200,2)	0.045	0.19	-18.039	0.843	0.218	-17.196			
dde*(500,2)	0.018	0.076	-45.097	2.135	0.0873	-42.968			
dde*(1000,2)	0.009	0.038	-90.194	4.336	0.0436	-85.858			
dde*(2000,2)	0.0045	0.019	-180.39	8.984	0.0218	-171.406			

Table-3: Relation between ground state d-d distance  $R_{dd} = R_{gs}$  and mean kinetic energy of d-d pair  $E_{d-d}$ 

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Table-4: Mean Relative Kinetic Energies (MKE) of d-d pair in clusters

Cluster	Potential	$R_{dd} = R_{gs}$ (pm)	MKE (keV)	$E_{gs}$ (keV)
$D_2$	$V_{s2}(1,1)$	76.7	0.0027	-0.0351
ddµ	$V_{s1}(208,1)$	0.8	0.18	-3.005
4D/TSC-min	V <sub>tsc</sub>	0.025	13.7	-130.4

Used trapping potential for dde\* EQPET molecule is defined in Ref.2 with its referred papers. The trapping potential for ddµ molecule can be replaced with that for dde\*(50, 2) in Table-3.

We have done the variational calculations to find minimum values of Hamiltonian by varying

 $\sigma$ -values and  $R_{gs}$ -values of Gaussian wave-function of d-d pair using preliminarily calculated

trapping potentials,  $V_{s2}(1,1)$ ,  $V_{s1}(208,1)$ ,  $V_{tsc}$ , respectively for D<sub>2</sub> molecule, muonic dd molecule and 4D/TSC-min cluster. We have obtained ground state eigenvalues for  $R_{gs}$ ,  $E_{gs}$  and mean kinetic energy (*MKE*) of d-d pair, as shown in Table-4. *MKE* values thus obtained are comparable to those of EQPET molecules (Table-3) with similarly close  $R_{gs}$  values, respectively.

## 4. Discussions

The *MKE* value 2.7 eV of d-d pair in D<sub>2</sub> molecule corresponds to 32,000°K temperature-equivalent, very high temperature compared with kT = 0.025 eV (300°K) of 0.1MPa D<sub>2</sub> gas at room temperature. An important aspect is that the high *MKE* value of d-d pair inside a D<sub>2</sub>

molecule's trapping potential (some 100pm size well) is adiabatic, namely isolated from the outside thermal motion of  $D_2$  gas, colliding randomly and getting equilibrium at 0.025 eV mean kinetic energy. Therefore, we do not need to heat up  $D_2$  gas to get the 32,000°K temperature-equivalent state inside a  $D_2$  molecule. This state is analogous to the confinement of high temperature (ca. 10 keV) DT(or DD) plasma by huge magnetic bin (ITER machine of tens meters size) or ICF device: The hot fusion plasma is adiabatic to the outside gas energy state of facility operation room where people are working at room temperature (300°K).

The other important aspect of the high *MKE* is that the confinement of d-d pair in microscopic potential well as  $V_{s2}(1,1)$ ,  $V_{s1}(208,1)$  and  $V_{tsc}$  is quantum-mechanically constrained by the Heisenberg Uncertainty Principle (HUP). In other words, de Broglie wave-length of deuteron should become shorter as the space of trapping potential well becomes smaller. The smaller de Broglie wave-length forms, the higher is the expectation value of particle momentum or kinetic energy. This QM constraint is automatically working by the variational method calculation (Fi.g.3). The case of D<sub>2</sub> molecule is shown in Fig.4, and the case of muonic dd molecule is shown in Fig.5.

We know that D<sub>2</sub> molecule at room temperature is stable (infinite life time). The squared Gaussian wave-function of d-d pair should have negligibly small tail outside the about 100 pm wide well of  $V_{s2}(1,1)$  potential. To satisfy this QM condition, *MKE* value should be elevated to be 2.7 eV in the case of D<sub>2</sub> molecule, and 180 eV in the case of muonic dd molecule. In the case of 4D/TSC-minimum at the final stage of condensation motion, *MKE* of d-d pair was calculated [2] to be 13.7 keV with  $R_{dd} = 25$  fm, which can be regarded as an adiabatic state of d-d pair trapped within an adiabatic TSC potential with -130.4 keV depth and approximately as an virtual HMEQPET dde\*(2000,2) state (see Table-3 and Table-4). By the Langevin equation analysis [2, 3], we have estimated that the existing time interval of 4D/TSC-minimum state, as an adiabatic very condensed neutral D-cluster is as short as  $2x10^{-20}$  s, for the 100% 4D fusion will take place in that time interval to break-up the 4D cluster to <sup>4</sup>He and other minor charged particles [1].



Fig.4: Gaussian wave-function and  $V_{s2}(1,1)$  potential of  $D_2$  molecule



#### Gaussian Wave Function and Vs Potential for dd-muon

Fig.5: Gaussian wave-function and  $V_{sl}(208,1)$  potential for muonic dd molecule

Since we predict the event of 100% 4D fusion per one 4D/TSC(t=0) generation, the microscopic nuclear active condition in condensed matter should be as high density as  $10^{12}$  tsc/mol-metal, to meet 10 watt/mol-metal clean heat generation. We have studied some possibilities in our recent works [2, 5, 6] for realizing such 'dissipative' level TSC densities in PdD<sub>x</sub> lattice and nano-particles, as for dynamic solid state and/or surface physics conditions.

Mostly in other theories treating BEC state nuclear fusion [7, 8] and review works [9], they are conceiving that some nuclear reactions in condensed matter are taking place with low energy (as low as 0.025 eV at room temperature, or smaller [10]). However, in the domain of nuclear reactions with strong nuclear force, relative kinetic energy of interacting particles should be so elevated as to satisfy HUP.

### 5. Conclusions

The TSC theory predicts that the physics principle of cold fusion is the transitory confinement of high kinetic energy deuteron cluster in a very deep and short life adiabatic potential. The estimated mean kinetic energy of d-d pair was about 10 keV within a very short time-interval as  $2x10^{-20}$  s with -130 keV deep dynamic potential well. The general principle of cold fusion is the confinement of d-d pair with elevated mean kinetic energy within Coulombic trapping potential well and we showed examples for D<sub>2</sub> molecule and muonic dd molecule as well as 4D/TSC

dynamic condensation as TBEC (transitory Bose-Einstein condensate). In the view of nuclear reaction physics, such principle as given by the TSC theory is quite analogous to the high temperature (as 10 keV for example) DD(DT) plasma confinement by very large device of magnetic bin (ITER) and ICF device. Therefore, the cold fusion stands as the hot fusion does.

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## The Cold Fusion Phenomenon in Hydrogen-graphites

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### Abstract

The cold fusion phenomenon (CFP) observed in solid materials with specific compositions (CF-materials) in open, non-equilibrium conditions has been consistently explained by a model assuming existence of neutrons in the materials. The essential factor of the material where realized the CFP is the interlaced superlattices of a host atom and a hydrogen isotope (protium and/or deuterium); one example of the first is Ni-H and another of the second is Pd-D. An exceptional example is XLPE (cross-linked polyethylene) in which a carbon lattice and protium lattice are interlaced according to its special structure.

In addition to the systems described above, there are several experimental data on the nuclear transmutation in carbon systems. In these experiments, 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 elements found in the experiments exist abundant in nature as we had pointed out in relation to one of the statistical laws in the CFP, the stability effect of nuclear transmutation products, as shown in Fig. 2.11 of our book *The Science of the Cold Fusion Phenomenon* (2003, ISBN 0-080-45110-1)

On the other hand, it is well known that graphite absorbs hydrogen isotopes very much as to be nominated as a reservoir of hydrogen. In our previous papers, the formation of neutron drops in the cf-matter due to the interaction between carbon sublattice and proton sublattice is used for the explanation of nucl0065ar transmutations in XLPE. This mechanism suggests that a similar interaction may exist in the hydrogen-graphite where the carbon sublattice of graphite interacts with the proton sublattice interlaced with the former.

Then, the neutron drops in the cf-matter formed by the interaction between the interlaced sublattices facilitate the generation of new elements in hydrogen-graphite at a condition of room temperature. The generated elements have, as the stability effect of nuclear transmutation shows, similar characteristics to elements generated in celestial conditions. The experimental results obtained in the carbon arcing in water and in air show a possible application of CFP in hydrogen-graphite to eliminate hazardous nuclear waste by nuclear transmutations and to generate excess energy economically.

## 1. Introduction

The cold fusion phenomenon (CFP) discovered more than twenty years ago [1.1, 1.2] is composed of complex features with various fields of its occurrence and various products composed of excess energy exceeding those expected from possible chemical and physical processes and of new elements or nuclides only expected from nuclear reactions.

At first, some of these products observed by the pioneers of this field have been assumed to be results of nuclear fusion reactions in the material composed of transition metals, Ti, Ni, or Pd, and hydrogen isotopes, H and/or D. The phenomenon has been called "cold fusion" irrespective of inconsistency between the expectation based on the assumed fusion reactions between hydrogen isotopes and the experimental results.

As the increase of experimental data sets in various materials with large variety of composition has shown inadequacy of the use of the name "cold fusion" to express this new field, we proposed a name "cold fusion phenomenon (CFP)" keeping the original naming and considering our ignorance about the necessary and sufficient conditions for this phenomenon. We use this name throughout this paper also together with its abbreviated form CFP.

As we know at present [1.3, 1.4], the cold fusion phenomenon (CFP) encompasses a large variety of materials where occurs the CFP (CF materials) and of products only understandable by the occurrence of nuclear reactions (products of nuclear transmutations) accompanying excess energy.

Not considering the transition metal-hydrogen isotope systems, the most extensively investigated CF materials, there is very interesting CF materials composed of carbon and hydrogen isotopes. The one extensively investigated is the cross-linked polyethylene (XLPE) where detected generation of new elements in relation to the appearance of the so-called water trees. The data has been investigated and given a consistent explanation based on our model [1.5].

In addition to the data sets of XLPE, there are several data sets of nuclear transmutations in CF materials composed of carbon (graphite) and hydrogen. Due to the fragility of the material, the detailed investigation of the nuclear transmutation in these materials has been in its infantile stage. In this paper, we will take up experimental data sets obtained in this difficult material for its interesting properties and also its hopeful applications along our line of investigation successfully applied to XLPE and also to other CF materials [1.3, 1.5, 1.6]

## 2. Experimental Facts

Carbon is an element popular in our daily life. It is also used in our technology almost everywhere especially in electronics. Recently, graphite is noticed for its high ability to include a lot of hydrogen isotopes in relation to the demand for hydrogen as a fuel. Another property of graphite eagerly investigated is its use for walls of fusion apparatuses.

Unfortunately, the pioneering works on the graphite as a CF material revealing its ability to generate nuclear transmutation and accompanying excess energy is looked over due perhaps to the fragility of graphite causing poor reproducibility even if we expect a qualitative one for the CFP in general.

We survey experimental data sets of the CFP observed in materials including carbon (graphite) and necessarily hydrogen in this section.

## 2.1Nuclear Products of Carbon Arcs in Water and in Air

The experimental data sets reporting nuclear transmutations in CF materials composed of carbon (graphite) and hydrogen have been started with the arc discharge in water with carbon (C) cathode and C or other metals. In these experiments, excess energy has been out of consideration due to their experimental conditions.

### 2.1.1 R. Sundaresan and J. O'M. Bockris

R. Sundaresan and J. O'M. Bockris [2.1] observed generation of iron (F) by carbon arc in highly purified water in the presence of oxygen. The amount of the generated iron increased with the time of discharge as shown in Fig. 2.1.



Fig. 2.1 Effect of time of electrolysis on the formation of iron (Sundaresan et al. Fig. 2 [2.1])

## 2.1.2 M. Singh, M.D. Saksena, V.S. Dixit and V.B. Kartha,

Singh et al. [2.2] used DC arc between ultrapure graphite electrodes dipped in ultrapure water. The graphite residue collected at the bottom of the water trough was analyzed for iron content to show that the iron content in the graphite residue was fairly high, depending on the duration of the arcing. The experiment was repeated initially six times, and the results showed large variations in iron content [50 to 2000 ppm] in the carbon residue.

The iron in the carbon residue was also analyzed mass spectrometrically for the abundance of its various isotopes, and the results were more or less the same as that of natural iron as shown in Table 2.1. Besides iron (Fe), the presence of other elements like silicon (Si), nickel (Ni), aluminum (Al), and chromium (Cr) was also determined in the carbon residue, and it was found that the variation of their concentrations followed the same pattern as that of iron.

Table 2.1 Mass spectrometric analysis results of the iron recovered from the graphite residue in the carbon arc in water experiments along with the analysis of natural iron (Spec pure) [2.2, Table II].

	. Atom Ratios of Iron Isotopes					
Experiment Number	<sup>54</sup> Fe/ <sup>56</sup> Fe	57Fc/56Fe	58Fe/56Fe			
3	a	$\begin{array}{c} 0.0231 \pm 0.0007 \\ 0.0230 \pm 0.0002 \end{array}$	*			
Natural iron (Spec pure)	0.0636 ± 0.0005		0.00310 ± 0.00004			
4	*	$\begin{array}{c} 0.0230 \pm 0.0002 \\ 0.0233 \pm 0.0002 \end{array}$	*			
Natural iron (Spec pure)	0.0635 ± 0.0005		0.00308 ± 0.00004			
5	$\begin{array}{c} 0.0635 \pm 0.0010 \\ 0.0638 \pm 0.0005 \end{array}$	0.0232 ± 0.0002	a			
Natural iron (Spec pure)		0.0233 ± 0.0002	0.00310 ± 0.00004			

<sup>6</sup>In the sample, because of isobaric interference of <sup>54</sup>Cr<sup>+</sup> and <sup>58</sup>Ni<sup>+</sup>, the respective iron isotope results of <sup>54</sup>Fe/<sup>56</sup>Fe and <sup>58</sup>Fe/<sup>56</sup>Fe could not be reported.

## 2.1.3 I. Ogura, I. Awata, T. Takigawa, K. Nakamura, O. Horibe and T. Koga,

In the experiments by Ogura et al. [2.3], arc discharge was carried out in a water phase (distilled  $H_2O$  was used) with a DC discharge voltage of 25 V (110 V for the case (2) below). From analysis of water used for the carbon arc discharge, increases in quantity of Ca, Fe, and other several elements were recognized. When silicon or metal sticks were used for the counter electrode, the content of Ca was distinctly increased with the former. The amount of the generated element increases with the discharge time.

In the arc discharges, carbon (C) is used as the cathode, and as the anode (1)  $_6$ C, (2)  $_{14}$ Si, (3)  $_{23}$ V, (4)  $_{22}$ Ti, (5)  $_{27}$ Co, or (6)  $_{28}$ Ni is used. Generation of new elements in these cases is (1) Ca, V, Cr, Ni (by PIXE analysis) (2) Remarkable increase of Ca on the

anode, (3) Ca, Cr, Fe, Cu (by PIXE analysis) (4)  $_{23}$ V, (5)  $_{28}$ Ni, and (6)  $_{29}$ Cu. In the cases (4) – (6), it is noticed that the generated elements have one more large proton numbers than the anode metals;  $_{22}$ Ti  $\rightarrow_{23}$ V,  $_{27}$ Co  $\rightarrow_{28}$ Ni, and  $_{28}$ Ni $\rightarrow_{29}$ Cu.

## **2.1.4 T. Hanawa**

Hanawa [2.4] made experiments of carbon arc in (1) pure water and (2) aqueous electrolyte with  $Li_2CO_3 0.05 \text{ mol}/l$  and (3) one with  $Na_2CO_3 0.05 \text{ mol}/l$ . He observed Ca, Cr, Mn, Fe, Co, Ni, Cu, and Zn in cases (1), (2) and (3) in carbon residue. In the cases (2) and (3), there are no Ca, Co and Cu on the electrodes. In the case (3), there are a lot of metals on the anode.

Recently Esko [2.5] made an experiment similar to those introduced above. Non-metallic graphite or silicon powders (scientific grade 99.999% pure) are placed in a pure (99.999%) graphite crucible. The powders are charged with 36 V of direct current through a pure (99.999%) graphite rod. The crucible is connected to the negative pole, the rod to the positive pole of a power pack consisting of three 12 V solar charged batteries.

The powders display apparent magnetic activity following the above treatment. Moreover, magnetic activity remains in the powders six months after treatment, suggesting the effect is permanent. Treated graphite shows the presence of magnetic iron at a level of up to 1.6% by weight. A typical analysis sample (ICP analysis by New Hampshire Materials Laboratory,

August 9, 2007) shows the appearance of metals Si (10.500), Mg (1800), Fe (4700), Cu (4200), Al (7800), Ti (440), S (580) and K (1000 ppm) in treated graphite.

In addition to changes in the composition of the graphite power used in the tests, changes have been noted in the pure graphite rods used in the experiments. In a study conducted in October 2007, a shiny metallic "bubble" appeared on the striking surface of the rod. Upon analysis, the rod was found to contain the following metals Sc (35), Fe (640), Co (160) and Ni (1120 ppm).

## 2.2 Electrolysis with Charcoal Cathode with Light and Heavy Waters

A somewhat different experiment was performed by R. Takahashi [2.6].

## 2.2.1 R. Takahashi

He used charcoal cathode in electrolysis with electrolytes of 0.25N alkali hydroxides (LiOH, NaOH, KOH, RbOH and CsOH) in  $D_2O$ ,  $D_2O + H_2O$ , and  $H_2O$ . The input power was less than 4 W. Colorization of electrolyte liquids is observed with excess heat generation showing synthesis of substance by the electrolysis when  $D_2O$  or

 $D_2O(0.75) + H_2O(0.25)$  is used. Maximum excess heat was observed for the electrolyte of LiOH in  $D_2O(0.75) + H_2O(0.25)$  with an input power 2.5 W. He concluded that charcoal tends to form chemical compounds in the electrolysis and from this reason the fusion between deuterons, the cause of the excess heat, is poor.

## 2.3Characteristics of the Experimental Results

The experimental data obtained in the carbon arc experiments are destined by the statistical nature that is a common characteristic to nuclear reactions such as the statistical law governing the alpha decay of radium  ${}^{226}_{88}$ Ra  $\rightarrow {}^{222}_{86}$ Rn ( $\tau_{1/2} = 1.6 \times 10^3$  y). Therefore, we will look at the data as a total not individually.

## 2.3.1 Elements observed in Carbon Arcing Experiments in Water

(1) Detection of New Elements

The experimental data introduced in Section 2.2 are summarized as follows;

Sundaresan and Bockris; Fe

Singh et al.; Si, Ni, Al, Cr, Mn, Fe (and ratios <sup>54</sup>Fe/<sup>56</sup>Fe, <sup>57</sup>Fe/<sup>56</sup>Fe, <sup>58</sup>Fe/<sup>56</sup>Fe)

Ogura et al.; Ca, V, Cr, Fe, Ni, Cu

Hanawa; Ca, Cr, Mn, Fe, Co, Ni, Cu, Zn

Esko; Si, Mg, Fe, Co, Ni, Cu, Al, Ti, S, K

As a whole, we can list up the generated elements as follows according to their proton numbers with frequency in a parenthesis after the symbol.

 $_{12}$ Mg (1),  $_{13}$ Al (2),  $_{14}$ Si (2),  $_{16}$ S (1),  $_{19}$ K (1),  $_{20}$ Ca (2),  $_{22}$ Ti (1),  $_{24}$ Cr (3),  $_{25}$ Mn (2),  $_{26}$ Fe (5),  $_{27}$ Co (2),  $_{28}$ Ni (4),  $_{29}$ Cu (3),  $_{30}$ Zn (1).

The elements observed more than twice are marked by bold letters.

## (2) Determination of Isotopic Ratios

The isotopic ratios <sup>54</sup>Fe/<sup>56</sup>Fe, <sup>57</sup>Fe/<sup>56</sup>Fe, <sup>58</sup>Fe/<sup>56</sup>Fe detected by Singh et al. are not different from natural ones.

#### (3) Excess heat

The excess heat and unknown elements in the liquid are observed in the electrolytic experiment by R. Takahashi [2.6]. In relation to this experiment with charcoal cathodes, we would like to point out a characteristic of the charcoal. Charcoal has micro-channels built in biologically. This characteristic of the natural regular array of carbon corresponds to the graphite lattice in the carbon arc experiments and has close relation to biological nuclear transmutations as discussed several times by now, e.g. in our paper

[2.8] and book [2.9, Section 10.1].

We have discussed possible formation of regular arrangement of physiological cells to realize nuclear transmutations in biological systems. If the CFP is confirmed in charcoal systems and graphites, possibility of biological nuclear transmutation will have a strong support.

## **2.4 Comparison of the Experimental Data with other Data obtained in the CFP**

The first law of the CFP found by comparison of the data in the CFP [1.3] and the data obtained in the Universe [2.7] says that the more natural abundance of an element is the more product of the element by nuclear transmutation in the CFP as shown in Fig. 2.2.

The peaks of the  $\log_{10}H$  in Fig. 2.2 correspond to the following Z's shown in Table 2.2. It is clear that they coincide with the elements marked by bold letters observed in carbon arcing experiments except  $_{25}Mn$  which is not at a peak but the value is fairly large.

Table 2.2 Peaks of  $log_{10}H$  of elements in the universe [2.7].

Z	13	14	20	24	[25]	26	28	29	30
Element	Al	Si	Ca	Cr	Mn	Fe	Ni	Cu	Zn





Fig. 2.2 Correspondence between the frequency  $N_{ob}$  observing elements in the CFP and the relative abundances  $\log_{10}H$  of elements in the universe [2.7]; (a) (Z = 3 - 38), (b) (Z = 39 - 83) [1.3, Fig. 2.11].

Therefore, this law (the First Law of the CFP) seems applicable to the case of nuclear transmutation products in the above case of carbon arcing and tells us that the nuclear transmutation in the carbon arc is a phenomenon belonging to the same CFP as observed in the transition-metal hydrides and deuterides.

The isotopic ratios of iron nuclides observed by Singh et al. [2.2] are similar to the natural ones and is also an evidence of the replication of the nuclear reactions in the star by the nuclear transmutation in the CFP depicted in the First Law of the CFP, the stability effects of NT in the CFP [1.3, Section 2.5.5].

# **3** Theoretical Approach to the Nuclear Transmutation in Hydrogen-graphites

The experimental data sets overviewed in the preceding section confirm that it is possible to apply our theory successful in the explanation of the CFP in transition-metal hydrides and deuterides to the present case.

Our theory is based on the formation of the neutron band in the CF material where occurs the CFP. The neutron band is formed by the super-nuclear interaction between lattice nuclei X's at different lattice points mediated by hydrogen isotopes Y's (H or D) at interstices when X and Y are on the interlaced sublattices of a superlattice. We examine a possibility of the neutron band formation in hydrogen-graphite using the current knowledge of carbon nuclei and graphite structure.

For the formation of the neutron band, there are three necessary conditions related to

host nuclei, hydrogen isotopes and the superlattice composed of host sublattice and hydrogen sublattice.

## 3.1 Neutron Halo in Carbon Nuclei

Recently, many exotic nuclei with neutron halo of medium and heavy nuclei were discovered such as  ${}^{19}{}_{6}$ C [3.1],  ${}^{22}{}_{6}$ C [3.2], and  ${}^{31}{}_{10}$ Ne [3.3] by RIBF (RI beam factory) experiments.

The neutron halo of these carbon nuclei,  ${}^{19}{}_{6}$ C and  ${}^{22}{}_{6}$ C, is favorable for the realization of the neutron band as is discussed before [1.3, Section 3.7] in cooperation with the extended wave functions of hydrogen isotopes in graphite as shown in Section 3.3.

What kind of structure the nucleus has at the neutron drip line? This question is not solved yet. It is, however, theoretically concluded that there are few halo structures in heavy nuclei at neutron drip line [3.4]. If this conclusion is right, carbon is a favorable nucleus for the host as a CF material even if heavier nuclei that have no stable halo states in free space may become halo states by interaction with hydrogen isotopes in super-lattice. The halo states of carbon nucleus work to realize super-nuclear interaction even if the H or D lattice is not perfect to form the necessary ideal super-lattice supposed for PdD or NiH [1.3, Section 3.7].

## **3.2 Extended Proton Wavefunctions in Graphite**

Unfortunately, there are no explicit data of proton (or deuteron) wavefunctions in graphite crystal due perhaps to the fragility of the lattice and wide spread character of the wavefunctions. There are, however, results of simulations showing rather extended proton wavefunctions in graphene double layer (Graphene sheets stack to form graphite with an interplanar spacing of 0.335 nm) as shown in Fig. 3.1 [3.5].



Fig. 3.1 Probability densities for selected lowest eigenstates of the translational nuclear Hamiltonian. The lowest in-phase (top to bottom: first, second, and fifthe) eigenstates for the double-layer structure are shown (d = 8 Å) [3.5].

## 3.3 Superlattice of Carbon and Hydrogen

The structure of a graphite lattice including hydrogen isotopes is not explicitly shown due perhaps to the fragility and difficulty to obtain an ideal sample. However, there is a lot of data on the lattice structure of  $\text{LiC}_6$  used in the lithium battery [3.6, 3.7]. The lattice structure of  $\text{LiC}_6$  is depicted as shown in Fig. 3.2 [3.6].



Fig. 3.2 Lattice structure of  $LiC_6$  ( $\circ:C$ ,  $\bullet:Li$ ) [3.6]

We can guess the structure of  $HC_x$  by analogy with  $LiC_6$  as follows. The protons may locate at similar interstices as Li atom in  $LiC_6$  with more extended wavefunctions as shown in Fig. 3.1 due to the light mass of proton  ${}^{1}_{1}H$  compared to  ${}^{6}_{3}Li$  or  ${}^{7}_{3}Li$ . Therefore, we can imagine the structure of  $HC_x$  as a superlattice composed of a carbon sublattice and another proton sublattice, the latter may be rather incomplete.

The data obtained by NMR studies support this conjecture as follows [3.8]. The NMR spectrum of  $C^{nano}H_{0.96}$  is well represented by the sum of a Lorentzian and a Gaussian line, indicating two types of hydrogen coordinations. These two components may be ascribed to hydrogen in graphite interlayers and hydrogen chemisorbed at dangling bonds.

On the retention and diffusion of proton in graphite, there is a data telling us diffusion of protons in graphite lattice along the layer as shown in Fig. 3.3 [3.9]. The characteristics of retention and diffusion of hydrogen in graphite is expressed as follows; "When bombarded with energetic hydrogen ions at room temperature, graphite

is observed to retain all non-reflected hydrogen until a saturation concentration is reached. At room temperature the saturation level is  $\sim 0.4$  H/C. Further implanted hydrogen is rapidly released at virtually the implantation rate. However, some hydrogen atoms may be driven into the bulk of graphite beyond the saturated implantation layer. - - While molecular hydrogen does not react with graphite, the chemical reaction between atomic hydrogen and graphite occurs at thermal hydrogen energies in the absence of radiation induced displacements" [3.9].





 $E_{a}$  = activation energy for hydrogen recombination

Fig. 3.3 Potential energy diagram within the bulk of graphite. Region 1 represents the crystallite lattice, with higher activation energy and slow atom diffusion. Region 2 represents internal surfaces, with lower activation energy and fast atom diffusion. Region 3 represents the release of hydrogen into the gaseous phase (vacuum) as either atoms or molecules depending on the surface temperature. [3.9, Fig. 16]

## **3.4 Super-nuclear Interaction and Neutron Band Formation**

It is possible to have an interaction between nuclei  $X(\mathbf{r}_i)$  and  $X(\mathbf{r}_{i'})$  at different lattice points by super-nuclear interaction mediated by occluded hydrogen isotopes (H or D) at interstices ( $\mathbf{r}_j$ 's) when there is a super lattice of X and H (or D) composed of a sublattice of X and a sublattice of H (or D) [1.3, Sec. 3.7.2].



Fig. 3.4 Super-nuclear interaction between neutrons at a lattice point i and another i' mediated by a proton at an interstice j.

Then, it is possible to form a neutron band by the super-nuclear interaction between lattice neutrons and there appears a high density neutron matter at a boundary region by positive interference of reflected neutron waves [1.3, Sec. 3.7.4].



Fig. 3.5 Formation of neutron drops  ${}^{A}{}_{Z}\Delta$  composed of Z protons and A – Z neutrons at the boundary region where neutron density becomes very high by the coherent augmentation of neutron waves reflected at the boundary [1.3, Sec. 3.7.4].

The speculation given in the previous Sections 3.1, 3.2 and 3.3 gives us a hope to expect formation of a neutron band by this super-nuclear interaction in hydrogen-graphite samples in its optimum situation with a fairly good superlattice structure. If we can expect the formation of a neutron band in a hydrogen-graphite system, there occur nuclear reactions resulting in such a CFP as that observed in the transition-metal deuterides and hydrides.

### 4. Discussion

We can expect two types of nuclear transmutations due to the neutron drop-nuclear interaction at the boundary region.

- (1) Generation of a nucleus according to the stability effect where a rather stable neutron drop  ${}^{A}_{Z}\Delta$  transforms into a nucleus  ${}^{A}_{Z}X$ .  ${}^{A}_{Z}\Delta \rightarrow {}^{A}_{Z}X$ . (1)
- (2) Generation of a nucleus  ${}^{A}{}_{Z}X$  by an absorption of a neutron by a nucleus  ${}^{A-1}{}_{Z}X$  and then a beta-decay of  ${}^{A}{}_{Z}X$  to be  ${}^{A}{}_{Z+1}X'$ , i.e.  $n + {}^{A-1}{}_{Z}X \rightarrow {}^{A}{}_{Z}X \rightarrow {}^{A}{}_{Z+1}X' + e^{-} + v_{e},$  (2)

where  $\underline{v}_e$  is an antiparticle of electron neutrino.

The generation of new elements introduced in Sections 2.1 and 2.3 reveals these two phases of the nuclear transmutation in the CFP. The frequency of detection of new elements corresponding to the First Law (stability effect) and the isotopic ratios of iron the same to the natural ones reflect the above property expressed Eq. (1).

In the experiment by Ogura et al. [2.3], they observed that the generated elements have one more large proton numbers than the anode metals suggesting following nuclear transmutations;  ${}_{22}\text{Ti} \rightarrow {}_{23}\text{V}$ ,  ${}_{27}\text{Co} \rightarrow {}_{28}\text{Ni}$ , and  ${}_{28}\text{Ni} \rightarrow {}_{29}\text{Cu}$ . This data may be explained by the property (2) of the nuclear transmutation in the CFP expressed in Eq. (2). Actually, the nuclear data tell us that  ${}^{49}{}_{22}\text{Ti}$  suffers the transmutation (2) to be  ${}^{50}{}_{23}\text{V}$ ,  ${}^{59}{}_{27}\text{Co}$  to be  ${}^{60}{}_{28}\text{Ni}$ , and  ${}^{62}{}_{28}\text{Ni}$  to be  ${}^{63}{}_{29}\text{Cu}$  by absorption of a neutron and a succeeding beta-decay.

The existence of neutron halo nuclei of carbon  ${}^{19}{}_{6}$ C and  ${}^{22}{}_{6}$ C is favorable for the CFP in hydrogen-graphite and also in XLPE. Recently, many exotic nuclei with neutron halo have been discovered in light and medium nuclei such as  ${}^{19}{}_{6}$ C [3.1],  ${}^{22}{}_{6}$ C [3.2], and  ${}^{31}{}_{10}$ Ne [3.3] by RIBF (RI beam factory) experiments.

The neutron halo of the carbon nuclei  ${}^{19}{}_{6}$ C and  ${}^{22}{}_{6}$ C is favorable for the realization of the neutron band as is discussed before [1.3, Section 3.7] in cooperation with the extended wave functions of hydrogen isotopes in graphite as shown in Section 3.4.

What kind of structure the nucleus has at the neutron drip line? This question is not solved yet. It is, however, theoretically concluded that there are few halo structures in heavy nuclei at neutron drip line [3.4]. If this conclusion is right, carbon is a favorable nucleus for the host as a CF material. The halo states of carbon nucleus work to realize super-nuclear interaction even if the superlattice composed of a carbon sublattice and H or D sublattice is not perfect. However, the fragility of graphite may give influence on the qualitative reproducibility of the CFP to make its degree less than in rather stable transition-metal hydrides and deuterides.

This may be a reason for the CFP in hydrogen-graphite despite fragility of the graphite lattice and in XLPE (crosslinked polyethylene). Furthermore, this logic can be extended to biological molecular structure where are various carbohydrate ( $C_nH_{2m}O_m$ )

which have more or less ordered arrangements of carbon and hydrogen atoms. The data obtained by R. Takahashi [2.6] in a system composed of charcoal and deuterium might be an example of this type. Bionuclear transmutations extensively investigated by Vysotskii et al. [4.1] may essentially be understood by the same mechanism.

In the case of the NiH and PdD superlattice, the situation is a little different. Recent knowledge of the halo nucleus tells us that the formation of exotic nuclei with a large excess neutron number is limited to elements with small proton numbers *Z* in the free space. In CF materials, there is a mechanism to stabilize neutron halo nuclei with a medium *Z* value by the super-nuclear interaction mediated by hydrogen isotopes in the sublattice. Therefore, it is necessary to form a rather perfect superlattice to realize the neutron band in NiH and PdD [1.3, Section 3.7].

It should be noticed the fact that the cathodes in the carbon arc experiments become rather higher temperature as 3000 C. This fact should be closely related to the occurrence of the CFP in a hydrogen-graphite influencing the structure of the superlattice, a necessary condition for the CFP from our point of view. The self-organization of regular stable structure in non-equilibrium, open system is known in the nanotube formation [4.2]. The same mechanism may be participating in the formation of graphene (as seen in Fig. 3.1) at the surface of graphite in the carbon arc experiments.

Carbon is a hopeful material in application of the CFP and investigation of the structure giving rise to the nuclear reactions generating new elements is very important from scientific and engineering point of view.

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## Computer Simulation of Hydrogen States in Pd Metal of Face Centered Cubic Lattice

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**Abstract:** We simulated hydrogen states in palladium metal using a quantum molecular dynamics on a personal computer. Calculations were done with the 2x2x2 super cell of 28 Pd atoms with four vacancies and four H atoms simulations, which were located outside, on or inside the octahedron composed of six Pd atoms around an O site, by changing the inter-atomic distances of the octahedron with or without an impurity atom of H or alkali/alkaline-earth metal on the O site of the octahedron. We obtained the possibility that four H atoms were drawn into the octahedron when an impurity atom like calcium entered into the O site with the expansion of the octahedron and they were left near there when impurity atom went out from the O site with the contraction.

Keywords: computer simulation, quantum molecular dynamics, H states, octahedron, Pd, 4H state

#### 1. Introduction

When nuclear fusion or nuclear transmutation occurs in condensed matter like metals, it will be necessary for two and more hydrogen (H) or deuterium (D) atoms to gather and condense in the narrow space of the host metal<sup>1-3)</sup>. About the processes after H or D atoms gather, there are already some proposals in which they will overcome Coulomb repulsion and condense, and then they will cause nuclear reactions<sup>4,5)</sup>.

Then, in order to examine which kind of conditions cause two and more H to gather in metal, we have simulated before the H states outside, on or inside the tetrahedron in the 1x1x1 unit cell of four metals such as nickel (Ni), palladium (Pd) and platinum (Pt) of face centered cubic lattice using a quantum molecular dynamics on a personal computer<sup>6</sup>. This time, we made the calculations with the assembled eight units 2x2x2 super cell of 28 Pd atoms with four vacancies and four H atoms simulations, which were located outside, on or inside the triangular lattices of the octahedron composed of six Pd atoms changing the inter-atomic distances of it. Imposing the periodic boundary conditions on the super cell, we calculated the total energy, charge density and electronic structure of the host bulk metal of Pd by a computer simulation program within Density Functional Theory based on the local density approximation.

As a result of calculations, we observed the total energy of H states outside the triangular lattices of the octahedron were lower than that on them at first, and then those energies were reversed by changing the inter-atomic distances of the octahedron longer. This would mean that four H atoms located outside the triangular lattices of the octahedron could enter into it when it expanded. But they could not go up together far beyond the triangular lattices of the octahedron because Coulomb repulsion of them became strong.

However, it seemed to be possible that when the inter-atomic distances of the octahedron increased, an impurity atom like Ca entered into the O site drawing four H atoms into the octahedron, and then when those of the octahedron decreased, the impurity atom went out from the O site leaving the four H atoms near there to cause condensations.

## 2. Calculations

Making the calculations under the following conditions, we obtained the total energy, charge density and electronic structure of the simulation system of 28 Pd atoms with four vacancies and four H atoms in the 2x2x2 super cell.

(1) Hardware and Software used for Calculation

We used a personal computer which had 4 core / 8 way CPU and 8 GB main memory. And we used the first principles electronic state calculation program ABINIT which was using plane waves for the basis function and norm preservation type pseudo-potentials<sup>7, 8)</sup>, and we used VESTA for visualization of the electron density of calculation results<sup>9)</sup>.

## (2) Restrictions of the Location of Pd atoms

We applied the following restriction for the location of Pd atoms because of computational complexity and calculation time.

- ① Four vacancies are located at four of the 32 Pd atoms, which are on the four sides of cube circumscribed around the regular octahedron in the 2x2x2 calculation cell.
- ② The locations of Pd atoms outside the octahedron are not altered even when the inter-atomic distances of it are changed.
- (3) Locations of Four H atoms

Under the restriction mentioned above, the quantum electronic state calculation of total energy, charge density and electronic structure was made for the bulk Pd metal system in the following three cases of location of four H atoms outside, on or inside the octahedron changing the inter-atomic distances of it. And each calculation was done when an impurity atom of H or alkali/alkaline-earth metal entered or not entered into the

O site in the octahedron.

- ① Keeping tetrahedral symmetry, four H atoms are located on four of the eight centers of tetrahedrons (T sites) which adjoin to the octahedron respectively.
- ② Four H atoms are located on four of the eight centers of triangular lattices (P points) which compose the boundary planes between four tetrahedrons mentioned above
   ① and the octahedron respectively.
- ③ Four H atoms are located on four of the eight half distance points (O(1/2) points) between four P points mentioned above ② and the O site respectively.
- (4) Relation between T sites, P points, O(1/2) points, O site and Pd atoms

T sites, P points and O(1/2) points where four H atoms located were geometrically related to the positions of O site in the octahedron, Pd atoms which composed it and the tetrahedrons which adjoined to it as Fig. 1 shows. When the lattice constant was supposed to be 1 and the coordinates of two Pd atoms locating on the diagonal of the unit lattice were supposed to be described as (0, 0, 0) and (1, 1, 1) respectively, then the coordinates of representative T site, P point, O(1/2) point and O site were described as (1/4, 1/4, 1/4), (1/3, 1/3, 1/3), (5/12, 5/12, 5/12) and (1/2, 1/2, 1/2) respectively, and the ratio of distances to each position from coordinates (0, 0, 0) was 3: 4: 5: 6.

The four O(1/2) points mentioned above (3) - ③ are significant for this H(D) condensation because the distance of two H atoms next to each other in our calculation system is close to the nuclear distance (74pm) of H molecule, and because the side length of the cube which is circumscribed around the tetrahedron of four H atoms in our calculation system is close to the critical length of Takahashi's TSC (Tetrahedral Symmetric Condensation) Theory which supports H/D-cluster condensation to cause nuclear reactions in condensed matter.



Figure 1 Schematic Relation between T site, P point, O(1/2) point, O site and Pd atoms (The lattice constant is supposed to be 1.)

#### (5) Change of Inter-atomic Distances

We made the quantum electronic state calculation under the deformation of lattice changing the inter-atomic distances of the octahedron composed of six Pd atoms around an O site near the center of 2x2x2 super cell. The deformation  $\triangle a/a$  was varied from -0.1 to 0.1 in 11 steps, where 'a' denoted the side length of unit cell.

Fig. 2 shows the charge density distribution when an impurity Ca atom enters into the O site of Pd lattice of 2x2x2 super cell, four H atoms locate on four of the eight P points of octahedron around the O site, and the deformation is 0.01 that means the inter-atomic distance of the octahedron increased with 0.01 of the lattice constant of Pd.



Figure 2 Charge Density Distribution in Pd lattice of 2x2x2 super cell

It shows that Pd atoms (silver) line up wrapped in the cloud of electrons (yellowish green), Ca atom (blue) is surrounded by six Pd atoms of the octahedron and H atoms (yellow) are located on the center of triangular lattices of the octahedron.

### 3. Results and Discussion

As a result of calculations, the total energy of H states represents gentle slopes with the deformation of the octahedron as shown in Fig. 3. The upper graphs of (A) are the total energy of 28 Pd atoms with four vacancies in the calculation cell when an impurity atom of H or alkali/alkaline-earth metal enters into the O site in the octahedron. And the lower ones are those when four H atoms are added on four of the eight T sites adjoined to the octahedron compared with the upper ones. Lower graphs of (B) are the total energy when four H atoms are added on four of the eight P points in the octahedron, and lower ones of (C) are those when four H atoms are added on four of the eight O(1/2) points in it. To compare with the total energy mentioned above, those in cases where no impurity atom enter into the O site are shown at the first places of upper and lower graphs on (A), (B) and (C) respectively. And upper graphs of (B) and (C) are the same

ones of (A).



Figure 3 Plot of Total Energy versus Deformation of Octahedron,

For Pd28 with Four Vacancies, an Impurity Atom in the O site and

- (A) Four H atoms located on four of the eight T sites outside the octahedron.
- (B) Four H atoms located on four of the eight P points on the octahedron.
- (C) Four H atoms located on four of the eight O(1/2) points inside the octahedron.

In the upper graphs of (A), (B) and (C), the impurity Li, Na or K atom looks to be weakly constrained by the O site than the impurity H atom, and especially it looks that Na or K atom could not stay there when the octahedron contracts. Mg or Ca atom looks to be strongly constrained by the O site than H atom while the contraction of the octahedron is small, and then Mg or Ca atom would deeply enter into it as it expands.

In the lower graphs of added four H atoms of (A), (B) and (C), in cases where no impurity atom enters into the O site, the total energy is large in order of the states as these four H atoms locate on four of the eight T sites, P points and O(1/2) points because of Coulomb repulsion. In cases where the impurity atom enters into the O site, the whole total energy decreases as much as constrained energy of the impurity atom, but the each total energy increases because of Coulomb repulsion between the impurity atom and the four H atoms.

The increase of the total energy in the contraction of the octahedron with the impurity Ca atom looks most noticeable of all. Fig. 4 shows again the total energy when four H atoms locate on four of the eight T sites, P points or O(1/2) points outside, on or inside the octahedron respectively in cases where the impurity atom of H or Ca enters into the O site, comparing those in cases where no impurity atom enters into it.



Figure 4 Plot of Total Energy versus Deformation of Octahedron,

For Pd28 with Four Vacancies, H or Ca in the O site and

- (A) Four H atoms located on four of the eight T sites outside the octahedron.
- (B) Four H atoms located on four of the eight P points on the octahedron.
- (C) Four H atoms located on four of the eight O(1/2) points inside the octahedron.

In cases where no impurity atom enters into the O site, the total energy with four H

atoms located on four of the eight T sites, P points or O(1/2) points closes together in expansion of the octahedron, especially those of T sites and P points become reversed slightly. But it would be difficult for only these changes of total energy to explain that four H atoms enter into the octahedron and go up to the O(1/2) points near the O site to cause condensations and nuclear reactions.

On the other hand, in cases where Ca atom enter into the O site as an impurity atom, although the total energy increases in order of the states as four H atoms locate on four of the eight T sites, P points and O(1/2) points because of Coulomb repulsion, the total energy changes more rapidly from lower to higher than those of the case with no Ca impurity atom with the contraction of the octahedron crossing them. It would mean that Ca atom in the O site could draw four H atoms from the T sites to the P points and more deeply to near the O(1/2) points, and go out from it leaving them to be same energy near there.

#### 4. Summaries

We could simulate more real lattice system of 2x2x2 super cell assembled eight unit cells by a personal computer using the first principles electronic state calculation program. Investigating the change of the total energy of H states, we obtained the possibility that when the inter-atomic distances of the octahedron increase an impurity Ca atom entered into the O site drawing four H atoms into the octahedron, which located on the T site adjoining to the vacancies, next when the inter-atomic distances of the octahedron decrease the impurity Ca atom go out from it leaving the four H atoms near the O site. Like this, it would be possible for two or more H(D) atoms to gather in a narrow space by the help of impurity atom in the bulk metals which are limited in the deformation of lattice. But we have no idea about the detail mechanism of this process.

This time, we did not treat all permeated H atoms in the O sites of Pd metal but only four H atoms located outside, on or inside the octahedron at temperature 0K because of the restriction of computational complexity and calculation time. After this, in first we will simulate similarly the states of four H atoms located outside, on or inside the octahedron treating all H atoms enter into the O sites of Pd metal, and those of the tetrahedron in cases where an impurity atom of H or such as alkali/alkaline-earth metal enters into the T site in it. And next, the author would like to simulate similarly the states of four H atoms located outside, on or inside the octahedron and tetrahedron of cuboctahedron nano-particle of 55 Pd atoms which can deform larger than the bulk metal, and those of icosahedron nano-particle also. It would be necessary to carry out these simulations of H states in other face centered cubic lattice metals such as Ni and Pt, and at the finite temperature considering the phonons of lattice metals and the motions of H(D) and impurity atoms, so that we could obtain much information about the condensation of H(D)-cluster in metals of condensed matter.

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## Three Laws in the Cold Fusion Phenomenon and Their Physical Meaning

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### Abstract

There have been discovered three empirical laws in the CFP; (1) The First Law: the stability effect for nuclear transmutation products, (2) the Second Law; the inverse power dependence of the frequency on the intensity of the excess heat production, and (3) the Third Law: bifurcation of the intensity of events (neutron emission and excess heat production) in time. There are two corollaries of the first law: Corollary 1-1: Production of a nuclide  ${}^{A'}_{Z+1}X'$  from a nuclide  ${}^{A'}_{Z}X$  in the system. Corollary 1-2: Decay time shortening of unstable nuclei in the system. These laws and the necessary conditions for the CFP tell us that the cold fusion phenomenon is a phenomenon belonging to complexity induced by nonlinear interactions between agents in the open and nonequilibrium CF systems as far as we assume a common cause for various events in the CFP, i.e. excess heat production, neutron emission, and nuclear transmutation. The characteristics of the CF materials for the CFP are investigated using our knowledge of the microscopic structure of the CF materials consulting to the complexity in relation to the three laws explained above. A computer simulation is proposed to reproduce an essential feature of the CFP using a simplified model system (a super-lattice) composed of two interlaced sublattices; one sublattice of host nuclei with extended neutron wavefunctions and another of proton/deuterons with non-localized wavefunctions.

#### 1. Introduction

The science of complexity has developed in the last half of 20<sup>th</sup> century to give a rather complete perspective of nature not only inorganic but also organic systems including human beings extending mathematical treatment from the physical science of simple systems developed since the birth of modern science in 16the century to social and human sciences.

Now, our understanding of nature is not confined to the traditional area of natural science described by differential equations but extends to the events determined by nonlinear dynamics without quantitative reproducibility.

As we know from the beginning of the research in the cold fusion phenomenon

(CFP), there is no quantitative reproducibility and this is sometimes used to denounce the value of the investigation of the CFP. This situation is unreasonable if we recollect the fact that the qualitative reproducibility or probabilistic laws are popular in nuclear physics. One of these examples is the  $\alpha$ -decay of <sup>226</sup><sub>88</sub>Ra nucleus; We can not predict when a nucleus <sup>226</sup><sub>88</sub>Ra under investigation will decay to <sup>222</sup><sub>86</sub>Rn by emission of <sup>4</sup><sub>2</sub>He but we know the constant of the decay,  $\tau_{1/2} = 1.6 \times 10^3$  y, which describes a statistical law for temporal variation of the number of nuclei of <sup>226</sup><sub>88</sub>Ra in a system.

It is interesting to find out several empirical laws or regularities between physical quantities observed in the CFP [1.1 – 1.3] from experimental data sets summarized in books [1.4, 1.5]. These laws suggest statistical nature of the events in this field. The laws or regularities can be divided into three; (1) the stability effect for nuclear transmutation products, (2) the inverse power dependence of frequency on intensity of the excess heat production, and (3) the bifurcation of intensity of events (neutron emission and excess heat production) in time. There are two corollaries to the first law: Corollary 1-1: A nuclide  ${}^{A'}_{Z+1}X'$  is generated from a nuclide  ${}^{A'}_{Z}X$  in the system. Corollary 1-2: Shortening of decay time of unstable nuclei.

Recognizing the existence of these laws in this research field, we might be able to take a correct point of view for the science of the cold fusion phenomenon (CFP).

# 2. Three Empirical Laws deduced from Experimental Data and Their Explanation by Nonlinear Dynamics [2.1 – 2.3]

In the vast amount of information we have obtained in these more than 20 years since 1989, we can recognize several regularities or laws between observables in the CFP. The three laws we have figured out are specified as follows [1.1 - 1.3]; (1) First Law, the stability effect for nuclear transmutation products and two corollaries, (2) Second Law, the inverse power dependence of frequency on intensity of the excess heat production, and (3) Third Law, the bifurcation of intensity of events (neutron emission and excess heat production) in time. We give an explanation for them in this section.

# 2.1 The First Law; Stability Effect for Nuclear Transmutation Products ([1.1] Sec. 2.11, [2.1 – 2.3])

If we survey numbers of elements produced by the nuclear transmutation in the CFP, we notice the frequency obtaining an element has a positive correlation with the amount of the element in the universe (e.g. [2.4]). Plotting out (i) the number of experiments where observed an elements X together with (ii) that of the amount in the universe compiled by Suess and Urey [2.4] against its proton number Z, we obtain a diagram

shown in Fig. 2.1.1. The coincidence of the peaks of numbers (i) and (ii), gives the stability effect for nuclear transmutation products [1.1]. We may call this regularity the "stability law" for nuclear transmutation in the CFP.



(b)



Fig. 2.1.1. Correspondence between the frequency  $N_{ob}$  observing elements in the CFP and the relative abundances  $\log_{10}H$  of elements [2.4] in the universe: (a) Z = 3 - 38 and (b) Z = 39 - 83 [1.1].


Fig. 2.1.2. Measured production rate N(Z) for the nuclear transmutation on the atomic number Z for protons in palladium where and exponential decay of the maxima on Z follows a relation of a equation  $N(Z) = N^{\circ} \exp(-Z/7.86)$  and  $N^{\circ} = 3.56 \times 10^{17}$  atoms/cm<sup>3</sup>s ([2.5] Fig. 1).

In addition, the maxima of measured production rate N(Z) in many experimental data sets including the data by Hora et al. [2.5] shown in Fig. 2.1.2 rather agree with the magic numbers with exception of the magic number 20 where a clear minimum of N(Z)was observed in all cases. This coincidence of the maxima of N(Z) and the magic numbers is another example of the stability effect on the nuclear transmutation in the CFP.

This law shows that the stability of a nucleus keeps its nature in the cf-matter composed of high density neutrons in the neutron valence band [1.1, Sec. 2.4.2] and spring out as a nucleus just as in the case of nuclear transformation in stars. This characteristic appears also in somewhat different form in the following corollaries.

# 2.1.1 Corollary 1-1 (to the First Law); A nuclide ${}^{A'}_{Z+1}X'$ is generated from a nuclide ${}^{A}_{Z}X$ in the system.

The First Law explained above tells us that the nuclear transmutations occurring in the stars occur also in the CF materials in an appropriate condition where a nuclear transmutation occurs by transformation as classified before [1.3, Sec. 2.4.5]. However, there are situations where experimental conditions of the CFP reflect in the results of the

nuclear transmutation. The most important of the experimental conditions different from nuclear reactions in free space is the existence of lattice nuclei immersed in the cf-matter [1.1]. The lattice nucleus  ${}^{A}{}_{Z}X$  can absorb one neutron to be a new unstable nucleus  ${}^{A+1}{}_{Z}X^*$  which becomes finally another stable nucleus  ${}^{A-3}{}_{Z-2}X^*$ ,  ${}^{A+1}{}_{Z+1}X^*$  or  ${}^{A+1}{}_{Z}X$  by alpha, beta or gamma decay. This type of nuclear transmutations classified as the nuclear transmutations by decay [1.1, Sec. 2.4.5] can be picked up as a Corollary 1 to the First Law.

There are many examples showing this law in electrolytic and discharge experiments [1.4, Section 11.11]. The clearest example is obtained in electrolytic [2.6] and discharge [2.7] experiments where Pd transmuted into Ag.

Another example obtained in a different system is that in carbon arc in water where used several metal anodes and a carbon cathode [2.8]. In the cases of metal anodes of Ti, Co or Ni, a small amount of elements with atomic numbers increased by one, i.e. V, Ni, or Cu, was observed.

# 2.1.2 Corollary 1-2 (to the First law); Shortening of decay time of unstable nuclei.

The second situation characteristic to the CF material different from the free space is coexistence of the cf-matter and unstable radioactive nuclei in a crystal lattice. In the free space, a radioactive nucleus is governed by a statistical law in its stabilization by some decay processes. In the case of its coexistence with the cf-matter, the interaction between the nucleus and the trapped neutrons give another branch of stabilization giving the surplus energy to the lattice effectively without radiation outward.

There are several examples of drastic shortening of decay time of unstable nuclei including natural radioactive nuclei of uranium  $^{232}_{90}$ <sup>Th</sup>,  $^{235}_{92}$ U and  $^{238}_{92}$ U and of  $^{40}_{19}$ K and  $^{107}_{46}$ Pd supposed to be generated in reactions of the CFP [1.1, Section 2.5.1.1].

The data show very fast nuclear transmutation in the laboratory time scale of following decay reactions (with decay types and times in free space);

 ${}^{107}{}_{46}\text{Pd}^* \rightarrow {}^{107}{}_{47}\text{Ag}, (\beta, \tau_d = 1.3 \times 10^9 \text{ y}) [2.7]$   ${}^{40}{}_{19}\text{K}^* \rightarrow {}^{40}{}_{20}\text{Ca}, (\beta, \tau_d = 6.5 \times 10^6 \text{ y}) [2.9]$   ${}^{232}{}_{90}\text{Th} \rightarrow {}^{228}{}_{88}\text{Ra}, (\alpha, \tau_d = 1.4 \times 10^{10} \text{ y}) [2.10]$   ${}^{235}{}_{92}\text{U}^* \rightarrow {}^{231}{}_{90}\text{Th}^*, (\alpha, \tau_d = 1.0 \times 10^9 \text{ y}) [2.11, 2.12]$   ${}^{238}{}_{92}\text{U}^* \rightarrow {}^{234}{}_{90}\text{Th}^*. (\alpha, \tau_d = 6.5 \times 10^9 \text{ y}) [2.11, 2.12]$ 

2.2 The Second Law; Inverse-Power Dependence of Frequency on Intensity of Excess Heat Production ([1.1] Sec. 2.12, [2.1])

In several experimental data sets, we are able to count numbers  $N_Q$  of an event (excess heat) with a specific amount Q (or an excess power P) and plot them as a function of Q (or P) obtaining  $N_Q$  vs. Q (or P) plot. The first plot was obtained for the data by McKubre et al. [2.13] as shown in Fig. 2.2.1 [1.1]. This plot clearly shows that there is a relation of frequency vs. intensity with an exponent of 1 famous in complexity. This regularity may be called the inverse-power dependence of frequency on intensity of the excess heat production.



Fig. 2.2.1. Inverse power law revealed by excess power generation measured by McKubre et al. [2.13]

Another example of this law is obtained for the data of Kozima et al. [2.14] as depicted in Fig. 2.2.2. In this case, the exponent of the dependence is 2.



Fig. 2.2.2. Distribution of the frequency  $N_p$  (= y) producing excess power  $P_{ex}$  (= x). To depict log-log curve, values of  $N_p$  and  $P_{ex}$  were arbitrarily multiplied by 10<sup>n</sup>. (x = 100 in this figure corresponds to  $P_{ex}$  = 1 W). [2.14]

In addition to these formulations, H. Lietz [2.15] tried to check the inverse-power law using the data accumulated by E. Storms [1.5]. The resulting plot by H. Lietz is given in Fig. 2.2.3 which shows the exponent of 1.0.



Fig. 2.2.3. Distribution of 157 excess heat results summarized by Storms [1.5]. Values have been stored in bins of size 10. The line shows a power-law fit to the binned data with an exponent of 1.0 ( $r^2 = 90\%$ ). (Fig. 3 of [2.15])

Therefore, we may conclude that the excess heat generation in the CFP is governed by a statistical law popular in complexity.

# 2.3 The Third Law; Bifurcation of Intensity of Events (Neutron Emission and Excess Heat Production) in Time

The third law in the CFP is a little subtle statistically compared with the former two.



Fig. 2.3.1. Diagram showing the time evolution of the neutron emission from  $TiD_x$  sample during the run *A* (April 15-16, 1989). The values indicated are integral counts over periods of ten minutes [2.16].

Even if the number of examples is scarce, we have several fortunate data sets of temporal evolution of effects in the CFP. The first one is that of neutron emission from  $TiD_x$  by De Ninno et al. [2.17] published in 1989. The data are shown in Figs. 2.3.1 and 2.3.2.



Fig. 2.3.2. Diagram showing the time evolution of the neutron emission counts (ordinate) during the run B (7-10 April, 1989). The values indicated are integral counts over periods of 10 minutes [2.16].

Another data set is the excess heat generation observed by McKubre et al. [2.13] as shown in Fig. 2.3.3.



Fig. 2.3.3. Variation of Excess Power, Uncertainty and Loading ratio [2.13].

Furthermore, we can cite another example of the temporal evolution of excess heat generation measured by Kozima et al. [2.14] in Fig. 2.3.4.



Fig. 2.3.4. Excess power pulses during a 14 hour period of an experiment (070108) which lasted 12 days as a whole [2.14].

By the nature of events in complexity, we can give only qualitative explanation of experimental result in analogy to the mathematical results of numerical simulations using the logistic difference equation [2.17]. The analogical explanations of the laws observed in the CFP have been given using the nature of an equation of nonlinear dynamics, Feigenbaum's theorem [2.18], in the previous paper [2.1].

We cite hear a bifurcation diagram from J. Gleick's book [2.17] in Fig. 2.3.5. The main figure depicts  $x_{\infty}$  on the ordinate ( $x_{\infty}$  is  $x_n$  at  $n = \infty$ ) vs. the parameter  $\lambda$  on the abscissa of the logistic difference equation (l.d.e.)

$$x_{n+1} = \lambda x_n (1 - x_n), \quad (0 < x_0 < 1)$$
(1)

The inserted figures, a) Steady state, b) Period two, c) Period four, and d) Chaos, depict variations of  $x_n$  with increase of suffix *n* (temporal variation if *n* increases with time) for four values of  $\lambda$ ; a)  $1 < \lambda < 3$ , b)  $3 < \lambda < 3.4$ , c)  $\lambda \approx 3.7$ , d)  $4 < \lambda$ . The region a), b) and d) correspond to "Steady state", "Period two" and "Chaotic region" in the main figure, respectively.

As we have shown in the previous paper [2.3], we may take our parameter  $n_n$  in the TNCF model as  $\lambda$  in the l.d.e. (1)



Fig. 2.3.5. Bifurcation diagrams ([2.17], page 71).

## 3. Physical Meaning of the Three Laws

The characteristics of the CFP have been revealed in the three empirical laws and two corollaries introduced in the previous section. The contents of these regularities exposed in the experimental results have given us important hints to investigate physics of the nuclear reactions occurring in the CF materials resulting in various events in the CFP.

## 3.1 The First Law and the Nature of Nuclear Reactions in the CFP

First of all, the first law and its corollaries disclose the nature of nuclear reactions in the CF materials. The first law tells us that the nuclear reactions in the CFP expressed in the stability effect in the nuclear transmutation are quite similar to those occurring in stars.

However, there is a little modification by the existence of lattice nuclei resulting in the two corollaries. The existence of appropriate nuclides in contact with the cf-matter formed at boundary regions of the CF material [1.1, Sec. 3.7] induces nuclear reactions expressed in the two corollaries. A nucleus  ${}^{A}{}_{Z}X$  which can absorb a neutron to form a new nucleus  ${}^{A+1}{}_{Z}X^*$  and stabilize by a disintegration gives rise to the corollary (1-1). When the nucleus  ${}^{A}{}_{Z}X$  is a radioactive one, the result is drastic acceleration of the decay process of the nucleus resulting in the corollary (1-2).

## 3.2 The Second Law and the Structure of CF-Materials

The inverse-power law is ubiquitous everywhere in nature when the agents of a

system are complex and interacting with nonlinear forces. One of the most familiar examples is the Gutenberg-Richter law of earthquake [3.1, 3.2]. The inverse-power law appears where there is a mechanism to cause an effect triggered by a subtle stimulation caused by any fluctuation of a parameter in the system. The appearance of 1/f fluctuations in every systems composed of many-body agents [3.3] suggest us a close relationship between the inverse-power law and the 1/f fluctuation even the relation is not solved yet.

Therefore, the existence of the second law in the CFP as explained in Sec. 2.2 tells us that the nuclear reaction resulting in the excess energy production, and also in the nuclear transmutations in the CFP, is induced by a subtle fluctuation of any parameter in the system which is essentially uncontrollable.

### **3.3 The Third Law and Complexity**

The similarity between patterns of temporal variation of events in the CFP and those in nonlinear dynamics found by a numerical simulation suggests us existence of similar dynamical processes in CF materials to those assumed in the simulation that gave the bifurcation diagram from the period doubling to the chaos as shown in Fig. 2.3.5.

### 4. Discussion

The fluctuation known as 1/f fluctuation [3.3] is ubiquitous in all world from microscopic to macroscopic scale. Even if its origin is not confirmed yet [4.1], the existence is universal. So, if a phenomenon is caused by a fluctuation, the phenomenon is inherited the nature of the fluctuation and inevitably exhibit its characteristics. The inverse-power law in the CFP explained in Sec. 2.2 may be an evidence of its close relation to the 1/f fluctuation; the self-organization resulting in the formation of cf-matter or the nuclear reactions in the CFP may be triggered by the fluctuation in microscopic system.

Existence of the third law in the CFP explained in Sec. 2.3 suggests directly the statistical nature of the CFP while we do not know the mechanism of nuclear reactions in CF materials. Similarity between the bifurcation in the CFP and the bifurcation behavior exhibited by simulation of nonlinear dynamical systems may be an indirect evidence showing complexity in the CFP.

Once the cf-matter is formed in a CF material, the elementary particles, neutrons, protons and electrons, participating in the nuclear reactions in the CFP seem to behave as if they are in hot plasmas as in the stars if we consider the first law in the CFP as explained in Sec. 3.1. This is an amazing situation because the CF materials are in fairly low temperatures at most  $10^3$  K compared to about  $10^6$  K in the star. This difference

may be related to the existence of lattice nuclei in the CF materials while there is no stable lattice of nuclei in the star. The stable state of cf-matter in the CF material facilitates the same nuclear transmutations as in stars in rather quiet manner than those in the latter.

As we have already discussed [1.1, 4.2], experimental data sets obtained in really various experimental systems [1.4, 1.5] suggest peculiarity of the CFP that asked new quantum mechanical states not noticed until now. The quantum mechanical investigation of the CF material [1.1, 4.2, 4.3] has given evidence or a hint to explain the basis of the TNCF model fairly successful to give a consistent explanation of the experimental results as a whole [1.1, 1.4].

It is desirable to investigate a possibility to form the new state of neutrons suggested by the TNCF model so successful to explain several characteristics of the CFP. The most promising method in this direction will be a computer simulation of the neutron bands in the superlattice made of two sublattices of host nuclei (e.g. Ni or Pd) with simplified but realistic neutron wavefunctions and of protons (or deuterons) with nonlocal wavefunctions.

The Corollary 2 to the First Law may be applicable to the remediation of hazardous nuclear waste in atomic piles.

Investigation of the relation between the three empirical laws discussed in this paper and the science of complexity should be taking up in near future.

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# 4H/TSC Fusion by Simultaneous Weak and Strong Interactions

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The TSC theory is applied for the 4H/TSC condensation motion and possible weak nuclear interaction in the diminished size state (4H/TSC-min) of 4H-cluster. The 4H/TSC cluster may condense to a very small (in 5fm size) charge neutral entity, because the strong interaction - which works to make 100% fusion for 4D/TSC-min of ca. 20 fm size - does not work for the 4H-cluster and therefore the 4H/TSC-min sate may go further into very small size as 5fm. We may expect the kinetic energy of trapped 4 electrons in 4H/TSC-min will reach at around 600 keV (KE) which exceeds the threshold energy (272 keV) of the weak interaction:  $p + e \rightarrow n + v + (KE - 272 keV)$ . Produced neutron shall make promptly (by 100%) multi-body simultaneous strong interaction with 3 protons existing within the Compton wave length (1.4 fm) of charged-pions as interaction force-exchange bosons, to result in the  $3p + n \rightarrow {}^{3}He + p + 7.7 MeV$  reaction. Prediction of fusion rate in Ni-H systems and its secondary nuclear products as neutron (free emission) and gamma-rays is discussed to be very small (10<sup>-13</sup> order of magnitude) level. The primary 4H/TSC WS(weak-strong)-fusion rate is on the order of 10<sup>-7</sup> per 4H/TSC generation, which is much smaller than the 1.0 per 4D/TSC case. Discussion is given about conditions in Ni-H systems for enhancing 4H/TSC WS fusion rates to visible heat-power reaction rates.

Keywords: 4H/TSC condensation motion, weak-strong interaction, 4H/TSC fusion, <sup>3</sup>He, Ni-H system

### 1. Introduction

The TSC (tetrahedral symmetric condensate) theory has been developed for modeling the mechanism of 4D/TSC condensation motion and resulting multi-body D-cluster fusion [1-3]. The essence of 4D/TSC model is shown in a simplified scheme in Fig.1.

The model is applied to protium system 4H/TSC to study if observable nuclear reactions would take place. In the past, we have thought that 4H/TSC would not make fusion reaction, since *PEF* value is zero, namely no attractive strong nuclear interactions exchanging charged pions as force exchanger bosons [1-3]. However, 4H/TSC might condense beyond the size of 4D/TSC-min state (about 20 fm diameter), because break-up of 4H-cluster by strong interaction does not happen. Minimum size of 4H/TSC might come to so small as 5fm in diameter. If it happens so, mean kinetic energy  $E_{ke}$  of electrons in 4H/TSC might exceed the threshold energy of weak interaction, namely the reaction:

$$p + e^{-} + E_{ke} \rightarrow n + \nu + (E_{ke} - 272 keV)$$
(1)

: may take place to produce a neutron as an active hadron to make instantaneous strong interactions with neighboring protons in 4H/TSC-min state to induce the quick cascade strong reaction as,



Fig.1: The condensation and fusion model for 4D/TSC in simplified view on 4 steps



Fig.2: The condensation and weak-strong interaction model of 4H/TSC in simplified view

$$n+3p \rightarrow {}^{4}Li^{*} \rightarrow {}^{3}He+p+7.72 MeV$$
(2)

Problem here is how much reaction rate is expected in experimental CMNS conditions as those for Ni-H system.

#### 2. 4H/TSC Condensation Motion

The condensation velocity of 4H/TSC becomes 1.41 times the 4D/TSC condensation time, in proportional to the square root of mass ratio between deuteron (2.0) and proton (1.0). As shown in Fig.2, 4H/TSC will reach at 20fm diameter range in 1 fs so far. To understand the situation easily, we show Fig.3 for converting the condensation motion from 4D/TSC to 4H/TSC. The 4D/TSC-min state ends at around  $R_{dd} = 20$  fm because of the 100% break-up (by 4D fusion). In the case of 4H/TSC, condensation goes further because of no attractive strong force interaction between 4 protons. A conceivable final size of 4H/TSC-min would be about 5fm as shown in Fig.4. The TSC process may provide much easier high energy electron existence than the W-L model [4]. How long can the 4H/TSC-min state exist as pseudo-stable charge-neutral entity of such small size as 5 fm? We do not know the answer at the moment. However, mean kinetic energy of p-p pair would increase to about 100 keV and associating electron kinetic energy would become about 600 keV trapped within TSC potential of -1.2 MeV depth.

As the neutron producing weak interaction, Eq.(1) will well take place with this  $E_{ke} = 600 \text{ keV}$  which exceeds the threshold energy 272 keV of the electron capture reaction to proton. The analysis shown in the next section provides the rate is  $3 \times 10^{-7}$  n/cluster.



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Fig.3: Converted figure of 4H/TSC condensation, from that of 4D/TSC



Fig.4: Image of 4H/TSC-min state

We remember [1-3] that the 4D fusion happens 100%, namely with the rate 1.0 per 4D-cluster. The rate  $3x10^{-7}$  per cluster for the weak interaction seems too weak to observe it experimentally in the first glance. However, the effective life time of 4H/TSC-min may be much longer than that of 4D/TSC-min. It is one of factors to enhance the weak interaction rate. If 4H/TSC generation rate in condensed matter, e.g. Ni-H system would become isotopic enhancement, compared to 4D/TSC generation rate, we might have additional enhancement factor.

Hydrogen absorption into Ni nano-powder is endothermic at room temperature, but it is reported (Piantelli, for example in private communication) that exothermic absorption happens with intensively at 300°C or higher temperature, selectively for protium (H) gas. We need however further study to reveal the real effect.

## 3. Rapid Cascade Reactions from Weak-to-Strong Interaction

Widom-Larson proposed the production of very low momentum neutron by assumed high energy electrons in coherent harmonic oscillation of metal-H system on surface of condensed matter [4]. He assumes that electron kinetic energy would elevate to 1 MeV. However, such high energy electron generation in rather macroscopic metal-H lattice with many atoms even in coherent motion seems very difficult. If such high KE electron as 1 MeV appears randomly in metal-H system, it should produce very intense (lethal if reaction rate correspond to several watts heat level) bremsstrahlung X-rays to be easily observed: we have never observed such intense X-rays in CMNS experiments. It seems that the presently proposed 4H/TSC-min state may easily generate high KE electrons, as high as 600 keV or more, however within the TSC deep trapping potential adiabatically, which may cause the neutron generation by Eq.(1).

Using Fig.5, we estimated the effective volume of weak interaction on the surface of a proton of 4H/TSC-min state is  $4.5 \times 10^{-3}$  (fm)<sup>3</sup>.



Fig.5: Estimation of effective domain-volume of weak interaction between 600 keV electron orbit (assuming 1s wave function) and weak bosons (W+, W-) on surface of proton core with 1.2fm radius.

$$\Delta V_W = 4\pi R_p^2 \lambda_W = 4\pi \cdot (1.2 fm)^2 \cdot 2.5 \times 10^3 = 4.5 \times 10^2 (fm)^3$$
(3)

Here  $R_p$  is the proton radius and  $\lambda_w$  is the Compton wave-length of weak boson, 2.5 am  $(2.5 \times 10^{-18} \text{ m})$ .

We conceived here that the weak interaction happens within the range of weak boson Compton wave-length, so that the reaction domain is very isolated from other weak interaction domains on other protons of TSC. This means that the multiple neutron (or ployneutron) generation probability is negligible, and single isolated neutron will be born in an event. Reaction rate of this weak interaction is estimated using the procedure as shown in Fig.6. Single weak interaction rate is given by

$$\langle WIrate \rangle = (4\pi/h) \langle W \rangle_{w} \langle \Psi_{e}(r_{w}) \rangle^{2}$$
<sup>(4)</sup>

Here  $\langle W \rangle_W$  is the imaginary part of optical potential for the weak-isospin exchange, and is estimated to be 78 eV, namely very weak.

We remember  $\langle W \rangle$ s value for the p-n strong interaction for d-d reaction was 80 keV, about 1,000 times larger than  $\langle W \rangle_W$ .

The weight of electron wave function in Eq.(4) is calculated by assuming 1s-type wave function with very diminished Bohr-like radius 1.2 fm, to be  $5.9 \times 10^{-5}$  per one orbit electron (600 keV) of 4H/TSC-min state, integrated over the effective weak-interaction volume Eq.(3).



Fig.6: Procedure to estimate weak interaction rate for electron capture into a proton of 4H/TSC-min

Now we come to estimate the rate of strong interaction by just-born neutron by the weak interaction. The procedure of calculation is shown in Fig.7.

Since the strong interaction range of OPEP (or PEF) is 1.4 fm of charged pion Compton wave-length, the just-born neutron should make immediate strong interaction with all of three protons existing within the domain of 1.2 fm radius. The reaction takes place instantaneously within the Heisenberg Uncertainty in space. Thus the n + 3p fusion reaction Eq.(2) will happen with 100% probability because of large  $\langle W \rangle s$  value 0.115 MeV, as large as d-t fusion.

As the first weak interaction to generate neutron and the next cascade strong fusion of n + 3p reaction happens almost simultaneously, total <sup>3</sup>He production rate is  $2.8 \times 10^{-7}$  f/4H-cluster. The minor break-up channel from <sup>4</sup>Li\* to d + 2p + 2.22 MeV may contribute very slightly, because of known level and break-up scheme (TUNL library) as shown in Fig.8.

The 5.7 MeV proton by  ${}^{4}Li^{*}$  break-up may produce secondary neutrons by p +Ni reactions for  ${}^{61}$ Ni,  ${}^{62}$ Ni and  ${}^{64}$ Ni isotopes. And p + Ni reactions for  ${}^{58}$ Ni and  ${}^{60}$ Ni are endothermic with 5.7 MeV proton incidence. However the secondary neutron yield is very small as  $2.5 \times 10^{-13}$  n/p, and almost negligible ( $10^{5}$  neutrons/s per one mega-watt heat level by reactions of n + 3p). 5.79MeV proton will make Ni(p,  $\gamma$ ) reaction with about 100 times the n emission rate, because it happens mainly for  ${}^{58}$ Ni and  ${}^{60}$ Ni of high abundance, but not so biologically dangerous level if usual Pb layer gamma shield is set up to a 1 MW reactor by this 4H/TSC WS fusion reactions (but maybe too much speculation!). Probably, our previous proposal [5] of fission process of compound excited-Ge nucleus after 4p capture to Ni-isotope, during the time-interval of 4H/TSC-minimum state, might be more significant than the direct 4H/TSC WS fusion.



Fig.7: Procedure of reaction rate estimation by strong interaction between just-born neutron and remained 3 protons in the TSC-residual



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Fig.8: Energy levels of <sup>4</sup>Li and its break-up channels

# 4. Conclusions

Simultaneous (very rapid cascade) weak and strong interaction may be predicted in the final stage of 4H/TSC condensation.

About 20 watts (or more)/mol-Ni heat with  ${}^{3}He + p$  and *d* products is predicted (Clean Heat), by assuming long 4H/TSC-min state life (a few fs) and high density 4H/TSC (t=0) formation rate in such system as Ni-H. Heat level strongly depends on TSC generation rate in reaction matter as Ni-H, and is ambiguous to predict the level by the present knowledge.

We need further study in theoretical elaboration and experimental trials to detect predicted nuclear products as <sup>3</sup>He.

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