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Edited by Shinya Narita

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PREFACE

This is the proceedings of the 15th Meeting of Japan CF-Research Society (JCF15), which was held at Hokkaido Citizens Activities Promotion Center KADERU 2.7, Sapporo, Japan on November 1 -2, 2014. In this meeting, 9 presentations were given and 7 papers were submitted to the editorial board. They have been peer reviewed by the referees, and revised for the publication as the proceedings. As a result, 3 papers concerning the experiment and 4 papers concerning the theory were accepted for publication.

For all meetings, JCF1 through JCF13, we published the Proceedings. For the meetings after JCF4, published of we electronic versions the Proceedings on our web-site http://jcfrs.org/proc_jcf.html in addition to their printed versions. In view of low efficiency and low effectiveness in distributing information, we decided to discontinue the printed version for the meetings, JCF12. Only the electronic versions have been, and are to be published thereafter. Needless to say, any responses from the world scientists will be welcomed.

Recently, a new research laboratory has been established in cooperation with Tohoku University and Clean Planet Inc. This is a very good news for Japanese society. The laboratory is now expected to be a center of research of condensed matter nuclear science (CMNS) in Japan. However, there is a still serious problem that the number of active researchers is decreasing. We, Japanese society, should consider how we secure human resources, especially encourage young people to participate in the CMNS research.

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

Editor-in-Chief Shinya Narita, Iwate University June 2015

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Comparison of some Ni-based nano-composite samples with respect to excess heat evolution under exposure to hydrogen isotope gases

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Abstract – Three kinds of Ni-based metal-oxide nano-composite samples, NiO/Ni (NN), $Pd_{0.016}Ni_{0.070}/SiO_2$ (PNSII) and $Cu_{0.011}Ni_{0.077}/SiO_2$ (CNS2), were subjected to hydrogen absorption runs at various temperatures up to 350 °C. At room temperature (RT) only the PNSII sample absorbed substantial amount of hydrogen with positive heat evolution. The virgin PNSII sample at RT showed evolution of heat much larger than the energies produced by oxygen pickup reactions and the bulk absorption / adsorption, while in the #2 and succeeding runs the PNSII showed nearly equivalent absorption with the loading ratio and the heat evolution characteristic of the bulk Pd. In elevated temperature runs both of the PNSII and the CNS2, some phases recorded the specific excess power reaching 8 W/g-Pd and 1 W/g-Ni with integrated excess energy of 3.8 keV/Pd (6.5 keV/atom-H) and 0.6 keV/Ni (10 keV/atom-H), respectively. These excess heat values were observed after the saturation of initial H(D)-absorption to metal and implied non-chemical energy source.

Index Terms – Cu·Ni/SiO₂ oxide nano-composite, hydrogen gas absorption, oil-flow calorimetry, excess energy, 0.6 keV/atom-Ni.

I. INTRODUCTION

The authors have been studying phenomena of anomalous heat evolution from hydrogen-isotope-loaded nano-composite metal-oxide samples at room temperature (RT) using a twin absorption system, A_1A_2 [1, 2] and at elevated temperatures using scaled-up system C_1 with a ten-times larger reaction chamber (RC) equipped with a flow calorimeter using an oil coolant with a boiling point of 390 °C [3, 4].

The samples tested in the A₁A₂ system were mainly Pd-based and Ni-based nanocomposites supported by or mixed with zirconia; Pd/ZrO₂ ("PZ"), Pd_{0.08}Ni_{0.35}/ZrO₂ ("PNZ"), Pd_{0.04}Ni_{0.29}/ZrO₂ ("PNZ2B"), Ni/ZrO₂ ("NZ"), Cu_{0.081}Ni_{0.36}/ZrO₂ ("CNZ") and Cu_{0.21}Ni_{0.21}/ZrO₂ ("CNZIV"), and have shown effectiveness of metal-oxide nanocomposites for production of anomalously large hydrogen uptake and associated heat. These experiments were also effective to make an inference that surface adatoms of the minor component have a catalytic effect to enable hydrogen absorption of the host crystalline bulk and release of enhanced anomalous heat. The A₁A₂ system was also used for samples Pd/SiO₂ ("PS") and Pd_{0.011}Ni_{0.062}/SiO₂ ("PNS") to reveal effectiveness of meso-porous silica (mp-silica) for a supporter of nano-composite particles.

The purpose of the fabrication of the C₁ system was to make precise measurements for Ni-based samples at elevated temperatures using the oil flow calorimetry. The results of the absorption runs with $Cu_{0.02}Ni_{0.083}/SiO_2$ (CNS) and $Cu_{0.076}Ni_{0.36}/ZrO_2$ (CNZ4) are described in ref's. [3] and [4], respectively, to show some excess heat characteristics yielding 30 - 100 eV/atom-Ni and 5 W/g-Ni, respectively.

To confirm the interesting characteristics of the CNS sample, a sample with similar composition, $Cu_{0.011}Ni_{0.077}/SiO_2$ (CNS2), containing much larger amount (12 g) of Ni

was synthesized in our laboratory using mp-silica powder supplied by Admatechs Co. Ltd., and hydrogen absorption runs were performed at various temperatures from RT up to 350 °C. The performances are discussed in comparison with the results of the NiO/Ni-Al₂O₃ (NN) and Pd_{0.016}Ni_{0.070}/SiO₂-Al₂O₃ (PNSII) samples. The former NN sample had been recommended as a good sample for excess heat production by Dr. B. Ahern [5], which has to be reproduced in the present work. The latter PNSII sample was reproduced with a similar composition to confirm the interesting characteristics of the PNS sample that the energy levels of hydrogen absorption sites are deeper than 1.0 eV, and that the sample has another absorption site with rather high barrier energy requiring rather high temperature to load it.

II. DESCRIPTION OF THE SYSTEM

A schematic of the experimental system C_1 is shown in Fig. 1. For detailed description of the system, refer to the previous papers [3, 4]. Here are described only the results of calibration of the flow calorimetry with a flow rate of 10 cc/min. The calibration run also serves as a control run for foreground runs using Ni-based samples.



Fig. 1. Schematic of the C₁ absorption system after modification of the flow-ratemonitor and addition of the 200-W cartridge heater inside the reaction chamber (RC).

We have to know the heat conversion coefficient (dT/dW) and the heat recovery rate (R_h) beforehand using a dummy powder. We used the same kind of powder of alumina (sample ID; Al₂O₃3) with an average size of 60 µm [6] as that previously used in the calibration runs for the system with the flow rate of 20 cc/min [4]. From the temperature

history during the calibration run operated at a variety of combination of heater input powers of $(W_1+W_2) = (0+26)$ W to (69+78) W with a coolant flow rate of 10 cc/min, plots of temperature at TC2, T_{C2} , versus the total input power (W_1+W_2) are made as shown in Fig. 2. Here, W_1 denotes the outer (#1) heater power, and W_2 does the inner (#2) cartridge heater power. The plots are grouped according to the values of (a) W_1 , or (b) W_2 .

The temperatures in each group can be fitted by a linear function. The slope gives the mean coefficient of power-to-temperature conversion, dT/dW, for each group. Neglecting the cases of $W_1+W_2 < 30$ W, the average slope in (a) is 1.33 °C/W, and that in (b) is 1.64 °C/W. The former is smaller than the latter, which means that the #2 heater power is transmitted to the oil-outlet temperature (monitored by TC2) less effectively than the #1 heater power. This is because heat loss from the bottom flange of the RC due to radiation and conduction via connecting leads for the RTD's and the cartridge heater is not negligible at all for the rather slow oil flow of 10 cc/min. However, we adopted this slow flow rate in order to monitor the flow rate accurately on-line by the oil-droplet counting monitor. In the foreground measurements described in the following sections, the measured temperature excess, if any, will be divided by the conversion coefficient dT/dW = 2.7 °C/W or 1.33 °C/W to give the excess power observed in RT runs or elevated temperature runs, respectively. The latter value of dT/dW is very reasonable, since in most cases of the elevated temperature runs the excess power appears in the temperature range 200 °C < T_{C2} < 300 °C with 50 W $\leq W_1$ \leq 94 W, and since the reaction heat source would be located nearer to the #2 heater than the #1.



Fig. 2. Calibration run using H-filled Al₂O₃ powder, Al₂O₃3, which serves also as a control run with a coolant flow rate of 10 cc/min; H-Al₂O₃3. Temperatures at TC2 are plotted as a function of the input power, $W_1 + W_2$. They are grouped according to the values of (a) W_1 , or (b) W_2 .

The heat recovery rate is calculated as $R_{\rm h} = F \cdot \rho \cdot C \cdot (T_{\rm C2} - T_{\rm C6})/(W_1 + W_2)$, where F, ρ and C are the flow rate, the mass density and the specific heat capacity, respectively, of the coolant BT400. The mean value of $R_{\rm h} = 0.8$ in the cases of the input power

mentioned above will be used in calculation of correction factor for fluctuating flow rate of BT400.

III. HYDROGEN ABSORPTION RUN FOR THE NN SAMPLE

First, the absorption runs using the "QSI-Nano Nickel" powder, QuantumSphere, Inc. [7], are described. The particles of the powder have a core (Ni) - shell (NiO) structure with diameter ranging from 10 to 25 nm. We call this sample the NN sample. The thickness of the NiO layer is smaller than 2.5 nm [7]. We used 31-gram of the NN powder, which contains 27 gram (0.46 mol) of Ni, with molar ratio of O/Ni ranging from 0.8 to 0.4. Since it occupies only 150 cc, we filled the 500-cc volume of the RC with the same kind of alumina powder as that used for the calibration run, and mixed two kinds of the powder homogeneously.

XRD measurements showed the spectra shown in Fig. 3. The spectrum for the sample before use (B) without mixing with the alumina has two peaks of Ni, while peaks of NiO are absent. It seems that the layer on the (10-25)-nm-diam. particles has no crystalline planes with a specific orientation, or is amorphous. In the spectrum for the sample after use for H-NN/Al₂O₃#1 through #3 (A), we find no peaks originating in elements / compounds other than Ni and alumina.



Fig. 3. XRD spectra from NN-B (before use) and NN-A (after use).

Protium (H) and deuterium (D) absorption runs, H-NN/Al₂O₃#1, H-NN/Al₂O₃#2, and D-NN/Al₂O₃#3, were performed after vacuum baking for 120 hours at temperatures up to 270 °C with the heater power of $(W_1+W_2) = (69+20)$ W. The temperature history in the H-NN/Al₂O₃#1 run is shown in Fig. 4, as an example. Each time the heater power was varied, the phase number is advanced; #1-1 for (0+0) W, #1-2 for (20+10) W, #1-3 for (30+20) W, and so on.



Fig. 4. Temperature history in the H-NN/Al₂O₃#1 run.

In the figure also shown are the pressures at the RC and at the ST, P_r and P_s , respectively, and (H/Ni), the number of H atoms lost from the gas phase relative to the number of Ni atoms, which is calculated from the values of P_r and P_s , and volumes of the RC and the ST with a correction for the temperature based on the Boyle-Charles' law. As we see in Fig.4, (H/Ni) saturates at ~0.25, which means that the system has no gas leak from the ST and/or the RC. We discuss the value of (H/Ni) later in this section.

The most important variable among the temperatures at TC1 through TC6 and RTD1 through RTD4 is that at TC2, monitoring the oil-outlet temperature, while the oil-inlet temperature being monitored by TC6 was always kept near RT. The TC2 temperatures in the H-NN/Al₂O₃ runs are compared with that in the calibration / control run, H-Al₂O₃3, in Fig. 5. As is seen, we find no meaningful increase in TC2 temperature in all NN/Al₂O₃ runs. We conclude that in the NN powder is induced no anomalous effect, or no excess heat in the temperature range from RT to 300 °C.

In Fig. 5 the values of (H/Ni) are plotted again. In addition to absorption / adsorption (collectively called 'sorption'), the variable (H/Ni) could involve hydrogen consumption due to oxygen pickup reaction,

$$NiO + H_2 \rightarrow Ni + H_2O + 0.033 \text{ eV},$$
 (1H)

if the H_2O molecules are in the liquid phase or in the solid phase due to adsorption. If strong adsorption sites happen to be created on the surface of the NN particles, the latter would be possible, and the H_2O molecules would remain stuck on the surface throughout the run before the succeeding baking-absorption runs.



Fig. 5. Comparison of the temperature at TC2 for NN-sample runs with that TC2(Al₂O₃) in the calibration / control run, H-Al₂O₃3, to see nonexistence of excess temperature in the H(D)-NN/Al₂O₃ runs. Heater powers (W_1, W_2) in W are: <u>#0</u>; vacuum baking;; <u>#1</u>-1; (0,0) H₂ fill, -2;(20,10), -3;(30,20), -4;(50,30), -5-6;(69,40), -7; (94,40), -8;(94,57), -9;evacuation-baking (0,0)~(69,20), -10;(0,0);; <u>#2</u>-1;(0,0) H₂ fill, -2;(20,10), -3;(30,20), -4;(50,30), -5;(69,40), -6; (94,40), -7:(94,57), -8: (69,20) evac.-baking, -9:(0,0);; <u>#3</u>-1;(0,0) H₂ fill, -2;(20,10), -3;(30,20), -4:(50,30), -7:(94,57), -8:(69,20) evac.-baking, -9:(0,0); <u>#3</u>-1;(0,0) H₂ fill, -2;(20,10), -3;(30,20), -4:(50,30), -7:(94,57), -8:(69,20) evac.-baking.

On the other hand, the former, *i.e.*, condensation into liquid phase, is really possible in the present system. The sample has 0.23 mol of NiO on the average. If all of NiO molecules are deoxidized, the same amount of H₂O is produced, consuming 0.23 mol of H₂, *i.e.*, (H/Ni) ~ 1.0. The H₂O gas would take on a pressure of 1.2 MPa in the part downstream of the SuperNeedle valve including the RC with a free volume of 420 cc, if it can remain in the gas phase. However, since the vapor pressure of water at RT is 2.3 kPa, almost all of the H₂O molecules will be condensed in the tube outside the RC at RT. The liquid water will be vaporized and removed by subsequent evacuation prior to the next run.

The real value of (H/Ni) observed at saturation is 0.25. Then about 3/4 of NiO could be remaining in the subsurface region of the NN particles. This could be gradually deoxidized during the #2 and #3 runs, which could be the reason for the small values of (H/Ni) in the phases #2-2, #2-3, #3-2 and #3-3. However, there is also a possibility that the NN particles are really absorbing a little amount of hydrogen with very small sorption energy in the phases at elevated temperatures from 100 to 150 °C. It is hard to determine each contribution to the observed values of (H/Ni). In any case it is worth noting that the reaction (sorption / deoxidation) takes place with only a little frequency on the NiO/Ni particles at RT.

Also it is worth while to discuss a small burst of heat observed at the beginning of the #1-1 phase just after opening the SuperNeedle valve, which is shown in Fig.6. The #1-1 phase was initiated during the cooling phase #0-7 after the vacuum baking at the #0-6 phase. However, the gas injection was done without waiting for the equilibrium in the temperature distribution inside the RC; the equilibrium distribution was realized several hours after the initiation of the #1-1 phase, and all the temperatures at RTD1 through RTD4 became almost the same. Recall that the RTD1, RTD2, RTD3 and RTD4 are located at distances of 30 mm, 60 mm, 90 mm and 120 mm, respectively, from the bottom. Before the gas introduction, the upper portion of the powder above around RTD2 had higher temperature, and therefore larger (excess) energy stored, than after the initiation. It could be possible therefore that the burst was formed by fast release of the stored (excess) energy to the wall of the RC due to increased thermal conduction and convection under the H₂-gas fill. However, this hypothesis requires an assumption that there was a portion with a temperature higher than that of RTD4, *i.e.*, 35 °C. Moreover, a reason should be given for nonexistence of the similar burst in the #2-1 and #3-1 initiated similarly.

Another explanation of the initial burst could be that a real exothermic reaction was taking place inhomogeneously accompanying a consumption of hydrogen, (H/Ni) = 0.014. If this is the case, the excess energy calculated from the excess power integrated over the hump is 0.29 kJ using the temperature-to-power conversion coefficient dT/dW = 2.7 °C/W for the case of null heater power. Since it is not known how much sample atoms and/or hydrogen atoms are involved in the reaction, the specific energy is discussed only on the average over the whole sample. The averaged energy is then 0.0064 eV/atom-Ni, or 0.47 eV/atom-H, although the kind of the reaction is left unidentified.



Fig. 6. A small burst of heat at the beginning of the absorption run H-NN/Al₂O₃#1-1 at RT.

IV. HYDROGEN ABSORPTION RUN FOR THE PNSII SAMPLE

The PNSII sample has a composition of Pd_{0.016}Ni_{0.070}/SiO₂. Since 50-g of the PNSII sample containing 1.4-g (0.013 mol) of Pd and 3.5-g (0.060 mol) of Ni occupied only 170 cc, the sample was mixed homogeneously with 270-g of Al₂O₃ to fill the RC similarly to the NN sample described in the preceding section. An XRD spectrum of the sample before use is shown in Fig. 7. We find peaks of PdO and NiO accompanied by no peaks of Pd and Ni, which shows that almost all Pd and Ni atoms are oxidized. In addition, peaks at diffraction angles of 40.0 deg. and 46.5 deg. are observed, but are left unattributed. These could be peaks of some compounds of Pd and Ni, although the amount could be only a fraction.



Fig. 7. XRD spectrum from the PNSII sample compared with that for the CNS sample.

Similarly to the NN sample, three cycles of [baking - H₂(D₂) filling - heater power up-and-down] were done; D-PNSII/Al₂O₃#1-1 through #1-8, D-PNSII/Al₂O₃#2-1 through #2-8, and H-PNSII/Al₂O₃#3-1 through #3-8. The phases #*n*-1 (where n = 1, 2, 3) were started with filling the RC with D₂ or H₂ gas at RT. Figure 8 shows the temperature evolution in the #*n*-1 phases. As shown in the figure, humps with FWHM ~ 6 - 12 min. are formed on the RTD traces at the beginning of the #*n*-1 phases at RT, which caused delayed humps on the traces of the TC's with much larger time constant. In contrast to the case of the H-NN/Al₂O₃#1-1 phase, the humps are observed on all the RTD's, suggesting almost uniform evolution of heat inside the RC. In the figure, the number of D(H) atoms lost from the gas phase is expressed as the ratio to that of Pd atoms, (D(H)/Pd). If it is expressed as the ratio to the number of metal atoms 'M', where 'M' stands for both Pd and Ni, it is reduced by a factor of 5.5; *e.g.*, the very large saturation value of (D/Pd) ~ 3.2 in the #1-1 phase is reduced approximately to (D/M) ~ 0.58.



Fig. 8. D(H) absorption runs D(H)-PNSII/Al₂O₃#*n*-1 showing exothermic sorption at RT.

The humps at TC2 in the #n-1 phase are time-integrated to calculate an emerging energy per absorbent atom,

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

where W_a is the power per unit adsorbent atom. The energy E_a of about 3.8 kJ/0.013mol-Pd, *e.g.* in the #1-1 phase, corresponds to 3.0 eV/atom-Pd, or 0.54 eV/atom-M. One of the most remarkable fact is that the values of (D/Pd) ~ 3.2 and the energy in the hump $E_a \sim 3.0$ eV/atom-Pd in the #1-1 phase are much larger than those in #2-1 and #3-1 phases. It is rather easy to understand the values in the latter two, (D(H)/Pd) ~ 1 and $E_a \sim 0.2$ eV/atom-Pd, which are a little larger than or nearly equal to the values characteristic of bulk Pd absorption. In this case it has to be admitted that the Pd particles are separated from the Ni particles, and do not exhibit, or have lost nanoparticle characteristics in the #2 through #3 runs.

If we assume that (D/Pd) ~ 1.0 and $E_a \sim 0.2 \text{ eV/atom-Pd}$ also hold for sorption by Pd particles in the #1-1 phase, we have large differences between the observed ones, *i.e.*,

 $(D/Pd)_{ex} \sim 2.2$ and $E_{a,ex} \sim 2.8$ eV/atom-Pd, or excess D₂ lost from the gas phase is 0.029 mol and excess energy is 3.6 kJ.

One of the possible mechanisms for the excess loss and the associated heat evolution in the #1-1 phase is deoxidation of the oxides, NiO and PdO;

$$NiO + D_2(H_2) \rightarrow Ni + D_2O(H_2O) + 0.176(0.033) eV/atom-Ni,$$
 (1D(H))

$$PdO + D_2(H_2) \rightarrow Pd + D_2O(H_2O) + 1.77(1.63) eV/atom-Pd.$$
 (3D(H))

If 0.013 mol of PdO and 0.06 mol of NiO had been completely reduced, energy of 3.4 kJ would have been produced in compensation for D_2 consumption of 0.073 mol which is more than twice as large as the excess loss of 0.029 mol mentioned above. The loss of 0.029 mol can produce 2.6 kJ at most by the reactions (1D) and (3D) using the oxides available in the present sample. Another 1.0 kJ (or 0.7 kJ, if deoxidation of NiO (1D) is neglected in view of the result of the preceding section.) in the heat evolution in the #1-1 phase should then be accounted for by another unknown mechanism.

Also plotted in the traces in Fig. 8 is the time-dependent sorption energy, $\eta(t)$ [eV/atom-D]; the specific excess energy per unit absorbent atom in each time interval Δt (= 1 min. in the present study), $W_{a,ex} \times \Delta t$, divided by the increment of $L \equiv (D/Pd)$;

$$\eta(t) = \frac{W_{\text{a,ex}}(t) \cdot \Delta t}{L(t) - L(t - \Delta t)}.$$
(4)

The value of $\eta(t)$ is about 1 eV/atom-D at around the peak of the heat burst observed in the RTD's in the D-PNSII/Al₂O₃#1 run, *e.g.*, and increases sharply to reach the local maximum of about 60 eV/atom-D just after the peak of the burst. The value is almost one order of magnitude greater than that explainable by a chemical reaction. Although the value and the delay of the peak could have an important meaning, the discussion on this point should be made cautiously with possible delay in the signals taken into account. Not only the RTD signal might have some delay due to thermal diffusion from the surrounding area, but also the pressure just inside the RC might have some delay relative to that indicated by the gauge, which could be caused by finite conductance of the filter located downstream of the gauge. Evaluations of these effects are not possible at the moment.

After the maximum, $\eta(t)$ decreases, and turns to negative after L(t) reaches saturation. In this phase the $\eta(t)$ values have less importance in the present run, since the variation of L(t) after the saturation appears to be caused by fluctuation of the noisy signal. In this sense, the maximum value of $\eta(t=11:48) = 160$ should not be taken too seriously.

It is plausible, nevertheless, that the heat burst in the #1-1 phase is enhanced by some mechanism. At least, it is inferred that (i) the energy of metal hydride formation or sorption is anomalously large, or some unknown reactions are taking place, only in the #1-1 phase when O atoms are existing in the sample, or (ii) the sample contained O atoms other than those of PdO and NiO, and D₂O formation energy contributes to the observed excess heat in the #1-1 phase;

$$O + D_2(H_2) \rightarrow D_2O(H_2O) + 1.3(1.2) \text{ eV/atom-D(H)}.$$
 (5D(H))

It is necessary to know the amount of water molecules produced in the runs in order to factor out the amount of D atoms consumed for the water formation reactions and to know the number of absorbed / adsorbed atoms from the measured value of (D/M). It is therefore difficult to conclude the enhancement of the metal hydride formation energy, or the sorption energy, only from the present data. In the following, we discuss the excess energy mainly in the steady state, or the "saturation phase" in the elevated temperature phases.

Figure 9 shows the TC2 temperatures compared with those at the same input power in the control run using Al_2O_3 . Since the duration of the phase does not coincide with each other, the control temperatures are the saturation ones in each phase. In the figure the (D(H)/Pd) is also plotted. The large value of (D/Pd) in the #1-1 phase is considered to be due not only to sorption but also to deoxidation as discussed above, while those in the phases #2-1 and #3-1 at RT is considered to be due solely to sorption.



Fig. 9. Evolution of the temperature at TC2 for PNS-II sample runs compared with that in the calibration / control run H-Al₂O₃3. <u>#1</u>-1; (0,0) D₂ fill, -2;(10,5), - 3;(20,10), -4;(30,20), -5;(50,30), -6;(69,40), -7; (94,40), -8;(0,0), evac.-baking, (0,0);; <u>#2</u>-1;(0,0) D₂ fill, -2;(10,5), -3;(20,10), -4;(30,20), -5;(50,30), -6;(69,40), -7:(94,40), -8: (69,20) evac.-baking, (0,0);; <u>#3</u>-1;(0,0) H₂ fill, - 3;(20,10), -4;(30,20), -5:(50,30), -6;(69,40), -2:(10,5), -7:(94,40), -8:evac. (0,0).

In addition to the initial increase in (D/Pd) at RT in the phase #1-1, another increase up to (D/Pd) ~ 7.0 is observed in the phases with elevated temperature from 190 to 270 °C, *i.e.*, #1-5 through #1-7. If we assume that the mesoscopic lattice of Ni also acts as the absorbent, (D/M) is reduced to 1.2, which is even much larger than (H/Ni) ~ 0.23 in the H-NN/Al₂O₃ runs. It is probable, therefore, that not only deoxidation of NiO with negligibly small reaction energy (Eq. 1D) but also absorption into Ni lattice are prompted by a catalytic effect of Pd. We also see gradual increases in the elevated temperature phases of #2 and #3, which could be the absorption by the Ni lattice. In most phases the TC2 temperatures coincide with the control ones except for those at #2-7, #3-5 and #3-7, in which the excess temperatures of 14 - 11 °C corresponding to excess power of 11 - 8 W are recorded. These are a little larger than the systematic error of about 10 °C due to error in setting the heater power and uncertainty in the thermal insulation capability which could change when reassembling the system after replacement of the sample. Another uncertainty could be a fluctuation in the flow rate, which will be important in the CNS2 sample runs described in the next section. However, the deviation of the flow rate from the reference one was negligible in the present case of PNSII.

In conclusion, it is possible that the excess temperature is due to the true excess power. The excess power could be the excess heat due to D(H) sorption by Pd nanoparticles or some compounds of Pd and Ni at elevated temperature. The specific excess power per unit absorbent atom, $W_{a,ex} = 7.9 - 5.9$ W/g-Pd or 3.1 - 2.4 W/g-Ni, is integrated over each phase, and divided by the increment of L = (D(H)/Ni) to give the phase-averaged sorption energy, $\eta_{av,i}$ [eV/atom-D(H)], and the total excess energy per unit absorbent atom, $E_{a,ex}$ [eV/atom-Pd(Ni)];

$$\eta_{\mathrm{av},i} = \frac{\int_{t-T_i}^t W_{\mathrm{a,ex}} \mathrm{d}t}{L(t) - L(t-T_i)},\tag{4}$$

$$E_{a,ex} = \int_0^t W_{a,ex} dt , \qquad (2)'$$

where T_i is the period of the phase *i*. We have very large values of $\eta_{av,i} = 2.2 - 6.5$ keV/atom-D(H) and $E_{a,ex} = 3.8$ keV/atom-Pd or 0.84 keV/atom-Ni at the end of the #3 run, when the variation of *L* is very small. These are more than two orders of magnitude larger than the conventional chemical reaction energy, which suggests nuclear origin of the excess heat.

V. HYDROGEN ABSORPTION RUN FOR THE CNS2 SAMPLE

The 160-g CNS2 sample synthesized in our laboratory with a composition of $Cu_{0.011}Ni_{0.077}/SiO_2$, containing 12 g (0.21 mol) of Ni and 1.9 g (0.03 mol) of Cu, occupied the full volume of RC, 500 cc, leaving no void to be filled with Al₂O₃ powder. The sample CNS2 before use showed XRD spectra very similar to that for the CNS sample used earlier [3]. This indicates that the synthesizing procedure was almost successful. Since the CNS2 sample after use has not yet been subjected to the XRD analysis, effect of absorption runs on the crystallographic structure is discussed here with use of the XRD spectra for the CNS sample before and after use.

The XRD spectrum before mixing with Al₂O₃ powder is shown in Fig. 10 as 'CNS-B' together with that after use 'CNS-A'. In the former spectrum we find peaks of CuO and NiO with no peaks of Cu and Ni accompanied, which shows that almost all Cu and Ni atoms are oxidized in the as-received CNS sample. On the other hand, in the latter spectrum we see that almost all oxides are deoxidized to become Cu and Ni in the CNS sample during the hydrogen absorption runs. We have to imagine that the similar deoxidation was taking place in the CNS2 sample during hydrogen absorption runs described in the following.



Fig. 10. XRD spectra of the CNS sample after use, 'CNS-A', compared with that before use, 'CNS-B'. It is clear that almost all oxides were deoxidized after exposure to hydrogen isotope gases.

The hydrogen absorption run H-CNS2#1 was initiated after vacuum baking at temperatures higher than 240 °C similarly to the runs for other samples. The temperature evolution in the runs H-CNS2#1 through #4 (red line) compared with that in the calibration / control run H-Al₂O₃3 (black line) is shown in Fig. 11 together with (H/Ni) (orange line). Here it is assumed that the Cu atoms acting as a catalyst for H absorption of Ni lattice, and do not absorb themselves. We notice that the (H/Ni)

behaviour is very similar to that in the NN/Al₂O₃ runs except for the range of temperature (100 – 250 °C) inducing the initial increase in (H/Ni) in the phases #1-4 through #1-6, and the saturation value of about 0.9 in #1-7 and later. Similarly to the NN/Al₂O₃ runs, it is inferred that the initial increase is mainly due to deoxidation of NiO and CuO. The larger saturation value mentioned above in comparison to the NN/Al₂O₃ sample might be the possible results of catalytic effect of the minor constituent Cu.

During the runs, exceptionally large fluctuation in the flow rate of BT400 measured as the number of drops falling per second was observed. Then a correction to the temperatures in the control runs has been made by adding an extra temperature calculated from fractional decrease in the flow rate multiplied by a product of the input power, the heat recovery rate and a reciprocal of the power-to-temperature conversion coefficient. The corrected control temperatures are plotted as the blue chain line.



Fig. 11. Evolution of the temperature at TC2 in the H-CNS2 runs compared with that in the calibration / control run H-Al₂O₃3. Heater powers (W_1, W_2) in W are: <u>#1</u>-1; (0,0) H₂ fill, -2;(10,5), -3;(20,10), -4;(30,20), -5;(50,30), -6;(69,40), -7; (94,40), -8;(0,0), -9;(69,40), -10;(69,40) flow rate readjusted, -11;(69,20) evac.;; <u>#2</u>-1;(69,40) H₂ fill, -2;(94,40), -3;(69,40), -4;(50,30), -5;(69,40), -6; (69,20) evac.;; <u>#3</u>-1;(69,40) H₂ fill, -2;(94,40), -3;(50,30), -4;evac.;; <u>#4</u>-1; (50,30) H₂ fill (0.095MPa), -2;(69,40), -3;(69,20) evac.

In most phases of a variety of the input power, the measured TC2 temperature agrees with the corrected control temperatures within the systematic error of 10 $^{\circ}$ C. However, during the period from the phase #1-9 through #2-3, substantial increase beyond the error range is observed. After this period, the excess temperature shows a decline below the systematic error to become null in the phase #3-3 and later.

During this period the maximum excess temperature corresponds to the excess power of 10 W, or 0.8 W/g-Ni. The phase-averaged sorption energy, $\eta_{av,i}$, and the total excess energy per unit absorbent atom, $E_{a,ex}$, are calculated and plotted at the end of

each phase in Fig. 11. The maximum values are $\eta_{av,max} = 1.5 \times 10^4 \text{ eV/atom-H}$ and $E_{a,ex} = 3.8 \times 10^2 \text{ eV/atom-Ni}$. The former and the latter are more than three orders of magnitude and two orders of magnitude, respectively, larger than the conventional chemical reaction energy again. It should be emphasized here that the excess heat emerges in the phases with the loading ratio (H/Ni) approaching saturation and changing only slightly. This point again makes us infer that the excess energy has nuclear origin.



Fig. 12. Excess energy per H atom, $\eta_{av,i}$ (•), and the integrated excess per Ni atom, $E_{a,ex}$ (•), both plotted at the end of each phase.

Another feature characteristic of this sample is that the temperature distribution inside the RC is different from that in the control run using the Al_2O_33 sample. A comparison is shown in Fig. 13, where the H-NN/Al_2O_3#2 run is used as a reference instead of the control run, since the H-NN/Al_2O_3#2 run having the similar sequence of the heater power change showed the temperature distributions similar to those in the control run with no anomaly. It is shown in the figure that in the H-CNS2#2-1, #2-3 and #2-5 phases the temperatures at RTD1 and RTD2 located in the lower portion of the RC are higher by about 40 °C than those at RTD3 and RTD4 located in the middle part. On the other hand, the lower portion is cooler than the middle portion in the H-NN/Al_2O_3#2-5 phase.

The reason for this temperature inversion inside the RC could be the excess-heatcausing reaction in the lower part of the RC. The phases H-CNS2#2-1, #2-3 and #2-5 are ones with high values of excess temperature and $\eta_{av,i}$ as seen in Figs. 11 and 12, respectively.

However, the inversion was also observed in almost all phases of the runs from H-CNS2#1 through H-CNS2#4. In the last run H-CNS2#4 which was performed with H₂ gas at lower pressure, $P_r = 0.095$ MPa, the degree of the inversion was higher: The RTD2 temperature in the phase of (69+40) W exceeded 330 °C, while the temperatures at RTD4 and TC2 remained almost unchanged. The tendency that the lower pressure

results in the higher degree of the temperature inversion was also confirmed in the vacuum baking phase with the heater power of (69+20) W. In regard to the temperature distribution in the RC, the lower pressure means lower convection and lower thermal conductivity which reduce heat flow from the heater #2 to the bulk powder. As a result, the temperatures become higher not only on the surface of the heater #2 but also around it including the RTD1 and the RTD2.

The heat conductivity of a powder is a sum of contributions from conduction and convection by the filled gas but also from conduction through the powder material itself in addition to radiation. The conductivity of a powder material depends on the grain size and the bulk conductivity of the material. Both of these parameters are smaller for the mp-silica than for the Al₂O₃ filler; *i.e.*, the mean particle size of the mp-silica is around 1 μ m [8], while that of Al₂O₃ is 60 μ m, and the bulk conductivity of SiO₂ is 10 – 1.3 W/mK, while that of Al₂O₃ is 20 – 36 W/mK [9]. Therefore, the temperatures around the heater #2 in the H-CNS2#2 run are higher than those in the H-NN/Al₂O₃#2 run. This mechanism would account for the temperature distribution at least partly. The temperature inversion cannot be a corroborating evidence for the excess heat.



Fig. 13. Comparison of the temperature distribution in the H-CNS2#2 run with that in the H-NN/Al₂O₃#2 run equivalent to the control run H-Al₂O₃3. Heater powers (*W*₁,*W*₂) in W are: <u>H-CNS2#2</u>-1;(69,40), -2;(94,40), -3;(69,40), -4; (50,30), -5;(69,40): <u>H-NN/Al₂O₃#2</u>-4;(50,30), -5;(69,40), -6;(94,40), -7; (94,57)

Another point regarding the distribution to be discussed is drift of the temperature during the phases. Figure 14 compares the temperature evolutions in the H-CNS2#0-#1 and H-NN/Al₂O₃#0-#1 runs, where the virgin CNS2 sample and the virgin NN/Al₂O₃ sample, respectively, were subjected to baking followed by first exposure to H₂ gas. In

the former the RTD temperatures in the phases H-CNS2#1-5, #1-6 and #1-7 are decreasing after rising up to some values, while in the latter no appreciable decay is observed in the phases with the same heater power, H-NN/Al₂O₃#1-4, #1-5-6 and #1-7. This can be a firm evidence for existence of the temporally decaying excess heat evolution in the CNS2#1 run.

A counterargument might be that the temporally decaying temperatures could be caused by a change in the sample property. As is well known, ceramic particles easily suffer from densification under elevated temperatures. Similar process might proceed in our sample powder with a result that the heat conductivity of the powder increases. If the densification-like phenomenon was taking place, when the CNS2 sample was heated to several hundred °C, the temperatures inside the RC would have been lowered gradually, as discussed in the preceding paragraphs. Although the NN/Al₂O₃ sample showed no temperature drift in the H-NN/Al₂O₃#1-4, #1-5-6 and #1-7 phases, the densification-like process seems to have been completed in the baking phases, H-NN/Al₂O₃#0-5 and #0-6, as is shown in the figure. This hypothesis, however, requires substantial reason for nonexistence of the densification-like phenomenon in the D-PNSII/Al₂O₃ runs.



Fig. 14. Comparison of the temperature distribution in the H-CNS2#0-#1 runs with that in the H-NN/Al₂O₃#0-#1 runs equivalent to the control run H-Al₂O₃3. Heater powers (W_1, W_2) in W are: **H-CNS2#0**-2;(69,20), -3;(0,0), #1-1;(0,0) H₂ fill, -2;(10,5), -3;(20,10), -4;(30,20), -5;(50,30), -6;(69,40), -7;(94,40), -8; (0,0), -9;(69,40): **H-NN/Al₂O₃#0**-5;(69,0), -6;(69,20), -7;(0,0), #1-1;(0,0) H₂ fill; -2;(20,10), -3;(30,20), -4:(50,30), -5;(69,40), -6;(94,57) \rightarrow (69,40), -7; (94,40), -8; (94,40), -8;(94,57), -9;(0,0).

VI. SUMMARY AND CONCLUDING REMARKS

Three kinds of Ni-based nano-composite samples were subjected to hydrogen isotope absorption tests using the calorimeter system working at elevated temperature up to 350 °C. A comparison of their performances is shown in Table 1. The NN sample showed little heat evolution with no anomaly. At RT only the PNSII sample absorbed substantial amount of hydrogen with positive heat evolution. The virgin PNSII sample exposed to D₂ gas at RT showed evolution of heat much larger than the energies from the bulk sorption in addition to oxygen pickup reactions, while in the #2 and later runs the loading ratio and the heat evolution had a little larger than or nearly equal to the values characteristic of the bulk Pd sample. At elevated temperatures both in the PNSII runs and the H-CNS2 runs, the excess powers were observed in time intervals where the (H/M) ratios varied very slowly, while little excess powers were recorded in the phases with rapidly increasing (H/M) in the #1 runs. Therefore, it appears that the anomalous excess heat at elevated temperatures has some origins other than chemical energy of H-absorption/adsorption.

		NN/Al ₂ O ₃	PNSII/Al ₂ O ₃	CNS2	
Transient	Loading ratio (H/M)	0.02	3.2	~0	
phase at RT	Specific power (W/g-M)	0.26	3.9	~0	
in #1 run	Sorption energy (eV/atom-M)	0.17	3.0	~0	
Transient	Loading ratio (H/M)	< 0.02	0.87 - 1.0		
phase at RT	Specific power (W/g-M)	< 0.004	0.35 - 0.43		
in #2 or later	Sorption energy (eV/atom-M)	< 0.003	0.21 - 0.19		
	Loading ratio (H/M) in #1 run	0.23	7.6	0.9	
	Loading ratio (H/M) in #2 or	0.06	20.25	0.12 -	
	later	0.00	2.0 - 2.3	0.08	
Saturation	Max. excess power (W)	~0	11 - 8	10	
phase at	Max. specific excess power	~0	79-59	0.8	
elevated	$W_{\rm a,ex}$ (W/g-M)		1.) = 5.)	0.0	
temp.	Specific excess energy	~0	3.8	0.38	
	$E_{\rm a,ex}$ (keV/atom-M)		5.0	0.50	
	Phase-averaged sorption Energy	- 0	22 65	15	
	$\eta_{\mathrm{av},i}$ (keV/atom-H)	~0	2.2 = 0.3	15	
* 'Loading' in #1 runs includes hydrogen atoms spent for deoxidation of PdO or NiO.					
** 'H' stands for either H or D, and 'M' stands for either Ni or Pd.					

Table 1. Comparison of the samples in regard to excess heat.

Finally, we have to mention that any noticeable change both in the γ -ray and the neutron counting rates was not coincident with the excess power evolution. The range of the specific excess power and the specific excess energy obtained in our laboratory so far are indicated in Fig. 15, the extended Ragone plot originally presented in ref. 10, where the power density vs the energy density obtained by various energy sources are plotted. The sources include the ²³⁸Pu battery and 'eCat' device in addition to conventional ones; gasoline, H₂ fuel cells, Li-ion batteries, advanced flywheels, *etc.* It is obvious that the present device of ours as well as the 'eCat' has the parameters well superior to the conventional energy sources. The energy density of the CNS and CNSII sample runs is as high as 1000 times of gasoline. Thus, the present apparatus could be a

potentially hopeful, distributed high-energy-density source without hard radiations and radioactive wastes.



Fig. 15. Extended Ragone plot. The data from KobeU-Technova group are added to the original [10].

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Deuterium adsorption test using Pd-Ni and Pd-Ag multi-layered samples

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ABSTRACT

We performed deuterium desorption experiments using multi-layered Pd-Ni and Pd-Ag complex samples with fine-structured surfaces and investigated the deuterium diffusion dependence on the surface condition. We observed short-period temperature fluctuations for the samples with fine-structured Pd-Ni or Pd-Ag interfaces. This behavior might be explained by the deuterium diffusion properties in the metal complex.

1. Introduction

We have performed deuterium desorption experiments using various types of multi-layered complex samples such as Pd-Au, Pd-Pd, Pd-CaO, Pd-Ni, and Pd-Ag, and we observed anomalous heat evolution as well as radiative tracks [1-3]. In addition, anomalous heat evolution has also been observed in deuterium desorption experiments with Pd-Ni binary nano-particles [4,5]. In the experiment, the phenomenon could be attributed to the fine structure of the sample as well as a specific property of Ni in deuterium diffusion. Considering the experimental results, we performed deuterium desorption experiments using multi-layered Pd-Ni complex samples and investigated the thermal and deuterium diffusion behavior. In the experiment, samples with a fine-structured surface or interface of two metals were tested to investigate the dependence of the deuterium desorption behaviors on the surface condition.

The thermal behavior in deuterium should depend on the heat of hydrogen/deuterium dissolution. In general, metals are classified into exothermic and endothermic absorbers of hydrogen. Ni as well as Ag, Au, Pt, and Cu are classified as endothermic absorbers. If deuterium diffusion in the interface region of the binary metals has a significant effect on the thermal dynamics, we may expect similar behavior for a binary metal of Pd and an endothermic metal. Thus, we also tested a multi-layered Pd-Ag complex in this study. Table 1 lists the heat of dissolution of Pd, Ni, and Ag for deuterium [6].

Metal	Heat of solution (kJ/molH)
Pd	-10
Ni	16
Ag	68

Table 1. Heat of solution for deuterium.

2. Experiment

Sample preparation

The samples were prepared according with the following procedure. Pd foil (10 mm x 10 mm x 0.1 mm) was annealed at 900 °C for 10 h. After annealing, the surface contaminants were removed using aqua regia. Then, a Ni and Ag layer was deposited onto the sample surface using Ar ion beam sputtering.

In this study, we prepared samples with a fine structure at the surface of the thin film and interface of the binary metal. These fine structures were obtained by etching using an Ar ion beam or aqua regia. Table 2 lists the conditions of the samples used in this study. We tested 8 types of samples (i–viii); the number of runs for each sample is also shown in the table. Figure 1 presents a schematic view of the samples. Figure 2 shows the surface morphology before and after etching analyzed by AFM. In addition, we performed deuterium desorption experiments with a simple Pd foil for comparison.

Deuterium loading

The samples were exposed to 5 atm D_2 gas for 24 h. The weight of the sample was measured before and after loading, and the loading ratio (D/Pd) was calculated from the weight difference. Note that the loading ratio of the complex sample was calculated assuming that it was only composed of Pd.

Deuterium desorption

After loading, the samples were placed into a chamber and fixed by stainless steel clips, as illustrated in Fig 3. A Au needle was attached at the end of the clip, and the other side was connected to a DC power supply outside of the chamber such that DC current was applied to the sample to stimulate the diffusion of deuterium by the Joule heat at the point contact between the Au needle and sample surface. In this desorption experiment, the chamber was evacuated to ~10⁴ Pa by a TMP, and the current was set to 6 A. During the experiment, the surface temperatures of the sample and the inside pressure of the chamber were recorded continuously for 24 h.

Membrane	Etched surface		Number of	Sample type
material	Outer	Pd/membrane	runs	(see Fig. 1)
	surface of	interface		
	membrane			
Ni		_	10	i
	0	_	8	ii
	_	0	4	iii
	0	0	4	iv
Ag	_	_	8	V
	0	_	3	vi
		0	2	vii
	0	0	2	viii

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



Pd	Pd	Pd	Pd
V	vi	vii	viii

Fig. 1 Condition of sample structure.



Fig. 2 Surface morphology the before and the after etching observed by AFM.



Fig. 3 Experimental apparatus of unloading experiment.

3. Results and discussion

Deuterium loading

Figure 4 shows the D/Pd ratios and their averages for each sample. The loading ratios were observed to be 0.65–0.70 for most samples, and no significant difference of D/Pd for each sample was observed. These results suggest that D/Pd could not be affected by the structure of the sample surface and interface of the binary metal or the type of deposited metal film.



Fig. 4 Average value of D/Pd ratio of each sample structure.

Deuterium unloading

The time dependencies of the sample temperature and pressure in the chamber in the experiment for each sample type (i–viii) are shown in Fig. 5–12. We also tested plain Pd foil with and without deuterium loading, and the results are presented in Fig. 13 and 14, respectively. Directly after applying DC current to the sample, the surface temperature increased in every sample, which is supposed to be due to Joule heat. Then, deuterium desorption started such that the sample temperature gradually decreased because the deuterium desorption from Pd is an endothermic reaction. For sample ii, an unexpected temperature increase was observed approximately 2h after applying current. This phenomenon was also observed for sample vi (~7 h after desorption). Although its origin has not been clarified, this phenomenon might be related to the explosive diffusion of deuterium, especially at the interface region between the Pd substrate and metal membrane (Ni or Ag). Further iteration of the experiments is necessary to characterize the phenomenon.

Other interesting behavior is the short-period fluctuation, which lasted 2–4 h in the beginning of the desorption experiment, as observed in Fig, 7 (sample iii), Fig. 8 (sample iv), and Fig. 11 (sample vii). Such phenomena were observed in 1 out of 4 runs for sample iii, 2 out of 4 runs for sample iv, and 1 out of 2 runs for sample vii, and no clear behaviors were observed for the other types of samples.



Fig. 5 Surface temperatures and the inside pressure for sample i.



Fig. 6 Surface temperatures and the inside pressure for sample ii.



Fig. 7 Surface temperatures and the inside pressure for sample iii.



Fig. 8 Surface temperatures and the inside pressure for sample iv.



Fig. 9 Surface temperatures and the inside pressure for sample v.



Fig. 10 Surface temperatures and the inside pressure for sample vi.



Fig. 11 Surface temperatures and the inside pressure for sample vii.



Fig. 12 Surface temperatures and the inside pressure for sample viii.



Fig. 13 Surface temperatures and the inside pressure (Pd foil).



Fig. 14 Surface temperatures and the inside pressure for unloaded Pd foil.

Samples iii, iv, and vii have fine structures at the interface of the Pd surface and Ni or Ag membrane, and the fluctuation in temperature might be attributed to the specific condition of the sample.

We precisely investigated the time behavior of the temperature for these samples. Figure 15 shows the time differential of the temperature (dT/dt) as well as the temperature variation for the sample for which we observed the short-period fluctuation in the desorption process. A set of continuous increases and instantaneous decreases in temperature occurred periodically. Similar behavior was observed in every sample for which the short-period temperature variation was observed.



Fig. 15 Temperature variation for every second dT/dt.

Now we consider the following scenario assuming the heat of deuterium absorption/desorption results in the phenomenon. Because we did not observe a significant variation of the inner pressure of the chamber during the continuous temperature fluctuation, as shown in Fig. 16, deuterium atoms were not considered to be desorbed from the sample. Then, we speculate that deuterium diffusion from Pd to the Ni/Ag membrane and from the membrane to Pd frequently occurred in the period and that the heat of solution generated by deuterium back and forth diffusion between the two metals causes repeated endothermic and exothermic phenomena (Fig. 17). In addition, the deuterium diffusion velocity in Pd is faster than that in Ni or Ag [6]. Then, deuterium diffusion from Pd to Ni or Ag might occur more rapidly than that from Ni/Ag to Pd, which might explain the rapid temperature decrease and the gentle increase, as shown in Fig. 15. As mentioned above, the phenomenon was observed only for the sample for which the surface was etched before depositing the Ni/Ag membrane. Therefore, the fine structure of the interface of the metals is considered to be related to the activation of deuterium diffusion in the region. Although this scenario remains speculative, further study may lead to an understanding of the origin of the anomalous heat evolution observed in various loading/unloading experiments with binary or complex metal samples I and may clarify the trigger condition of a condensed matter nuclear reaction.



Fig. 16 Temperature and pressure variation.



Fig. 17 Schematic view of deuterium diffusion in the multi-layered sample with and without an etched interface.

4. Summary

We performed deuterium desorption experiments using multi-layered Pd-Ni and Pd-Ag complex samples with fine-structured surfaces and investigated the deuterium diffusion dependence on the surface condition. We observed a continuous short-period fluctuation in the desorption process in the experiment using the sample with an etched interface between Pd and the Ni or Ag membrane. We speculate that the unique property of deuterium diffusion in the region may be related to the unique temperature behavior.

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Analysis of heat generation by adiabatic calorimeter and matrix calculation for the reaction of fine metal in deuterium gas

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1. Abstract

Heat generation could be caused by the transmutation of hydrogen isotopes with metal nanoparticles as the catalyst. This paper presents a rigorous analysis of the excess heat generation that may arise from this process. Data analysis incorporated several factors such as room temperature variation, input heat, and the work done by the circulation pump. The effect of each of these factors can be computed. The results show the factors of heat flow during various control calibration tests with no reactants in the reactor. When these factors are applied to the excess heat generation tests, they provide clear evidence of excess heat generation.

2. Introduction

After the presentation at MIT colloquium⁽¹⁾, we improved the analysis of data for the excess heat generation test. We changed the test system, which include the position of the thermocouples, the room temperature variable, and the flow-rate and heat from the circulating pump. We added the quantity of the heat escape calculation into the calorimetry. One can calculate the rate of heat escape based on previous data which has been already collected. The gas used, the reactants, thermocouples and other equipment were described previously in reference ⁽¹⁾.

We have modified the old reactor chamber design which ran for up to one month, to its current design which enables data collection on an hourly basis. In the new reactor chamber, we replaced the previous heater with a new high temperature $(1,000^{\circ}C)$ ceramic heater that has a thermocouple built-in.

Moreover, we improved the inside arrangement of the reactor chamber by modifying the shape of the reactor itself and by increasing the amount of reactive metals. We estimated the heat generation by inputting a constant electric power for several hundred seconds. The temperature change was measured from the reactor chamber and the coolant water providing us with the amount of heat loss or gain. Measured and variable factors are the gas pressure and temperature shown as below along with the reactant metals;

- Reactant: pure D₂ gas 20~500 Pa
- Temperature: Heater 300~800 °C
- Ni mesh: 2 sheets of 300 x 300 mm, 46 g
- Pd wire: 0.2 mm , 2.82 g

There are many reports that discuss excess heat generation using Nickel or Nickel-based-alloy with protium gas⁽²⁻⁹⁾.

3. Experiment

3.1 Reaction test equipment, measurement methods and the reactor

Figure 1 shows the configuration of the reactor chamber. It is cross-shaped and the upper part has inlets for heater power, plasma discharge, and a thermocouple. These are connected with an ICF152 flange produced by Ulvac Co.. The bottom of this upper part is connected with an ICF203 flange to the reactor chamber. The details of the flange are shown in fig. 2, and is colored in pale blue.

In the fig. 1, the right side of the reactor has a viewing window connected to the chamber with an ICF152 flange. The viewing window, made of cover glass, is set in an ICF114 flange. In this figure, the left side is connected to the vacuum system and the mass spectrometry system, which has a pressure gauge and the gas inlet. These are attached with an ICF152 flange. This flange is connected with another ICF152 flange to the chamber. The lower part of the main chamber is connected with an ICF152 flange to the bottom part of the chamber. This part has an ICF203 flange and is connected to the vacuum system. The bottom part of the chamber has eight ICF70 flanges. The one flange is connected to the vacuum system and other flanges are for spare use.

In fig. 1 and fig. 2, the connecting gaskets are shown with yellow lines. These gaskets are fixed between the flanges for the purpose of keeping tight connections. They are all thin ring plates, made of SUS304. The flange and gasket seen in these figures have an in-line contact with protruding edges on each flange. Thermal conductivity of these gaskets is less than that of copper. The weight of each part shown is as follows;

- The reactor body chamber: 23 kg
- The top of input part: 2.7 kg
- The window port: 1.56 kg
- Gas inlet part: 4.55 kg
- The bottom part of the chamber: 20.2 kg
- The entire weight: 52.0 kg
- The weight except for the viewing window made of glass material and pipe: 50.5 kg

The weight of all the connection bolts, nuts, and gaskets are not included in the entire weight above.



Figure 1: Schematic view of component configuration of the reactor.



Figure 2: Reactor flange and gasket.

Figure 3 shows the outline drawing of the reactor and the simplified drawing of the arrangement of internal reactor materials. Figure 4 is a photo of the reactor. The entire body is made of SUS316 and the volume in the reactor is 5,530 cm³ with a total weight of 50.5kg. A 1,000 K-type electrical heater covered with alumina ceramic is

placed in the center of the fig. 3. The shape of the heater is a square 30 mm by 30 mm, with a thickness of 2 mm. Reaction material of palladium wire has a diameter of 0.2 mm and a length of 3,000 mm (Pd density is 12.02, the volume is 0.031 cm^3 and the weight is 1.14 g) is wound around the heater like a coil. The coil was wound with 15 full rotations. A R-type thermocouple is built inside the heater.

In addition, there is an electrode for plasma activation as shown on the right side in the fig. 3. The length of the electrode tip is 50 mm with a 3 mm diameter, covered with another Pd wire which is 300 mm in length and 1 mm in diameter (weight is 2.82 g). The reactor has two discharge electrodes. These electrodes are insulated from the reactor body. The polarity of these electrodes can be changed to either positive or negative. We always have the reactor body grounded to the earth.

Gas temperature near the Pd reactant is measured with another thermocouple (drawn as vertical bars in the center of figure 3). The K type thermocouple is covered with thin stainless steel and has a length of 300 mm and a diameter of 1.6 mm, which provides a very quick response. The thermocouple is 1.6 mm in diameter and 500 mm in length and is insulated with three alumina tubes having an outer diameter of 3.2 mm, an inner diameter of 2 mm and a length of 100 mm. The thermocouple, is not affected by the high voltage of the electric field from the plasma discharge.

The surface of the reactor inner wall is covered with two nickel mesh sheets as reactant. It is a 30mm square constructed with wires of 0.15 mm in diameter. The mesh number is 50. The weight of one mesh is 23 g, so the total weight is 46 g. The mesh is set at the center of the reactor, and is brought into contact with the reactor's inner surface to maintain heat conductance.

We have designed a new heat measuring system with a combination of an adiabatic method and the flow calorimetric method so that we can conduct heat generation tests and measure the results every hour. The newly designed heat measuring system has a cooling water pipe that is wound around the outside of the reactor wall with insulation. This provides a constant temperature of the reactor surface wall and the coolant in the dewar container. The entire surface of the reactor is covered with insulating material to maintain the temperature and to prevent the heat from escaping from the reactor body. Heat escapes only from the top and the bottom parts that have the pipe and the electrode terminals with the port. After the test, we cool the reactor by blowing air with the use of a pump into the opening between the reactor and the insulating material.



Figure 3: Arrangement of the internal materials in the reactor chamber.

Figure 4 is a photograph of the bare reactor that has no insulation materials and no cooling tubes. There are no coating materials on it. Parts indicated with yellow lines in fig. 4 indicate the positions where the stainless steel gaskets are placed.



Figure 4: Appearance of the reactor body.

Figure 5 shows a detailed drawing of the insulation and the cooling plastic tubes. The plastic tube is easily wound around the reactor body without any gaps. The arrangement of the outer-heater-tape is shown in fig. 6.



Figure 5: Drawing of the insulation and the cooling plastic tubes.

Figure 6 shows sectional details of the components of the reactor chamber. The difference between this set-up and that of our previous set-up, is the flowing cooling water system on the reactor chamber which allows us to measure heat generation in real time. The purpose for the flowing cooling water system is to uniforms the temperature of the reactor chamber as well as a heat recovery. A magnetic pump produced by Iwaki Co., model MD-6K-N, is used as a circulation pump. The pump has the maximum flow rate of 8,000 cm³/min, maximum head pressure of 0.1MPa, and the output power of the pump is 10 W.

We used a thin film heater tape for the heat calibration of input. It obtains the relationship between input power and the temperature. The heater tape is wrapped against the outside of the reactor wall. The heater is made by Tokyo Giken Co., model FTH-5, 10 mm in width, 5 m in length, has an output of 150 W at 100 V. It provides excellent heat conduction due to its small thickness of 1mm. The heater tape is wound around the outer surface of the reactor chamber as shown in fig. 6 (It is described as "Outer heater" in the figure).



Figure 6: Schematic detailed view of the reactor parts, connections, outer heater tapes and insulation.

Figure 7, 8 and 9, are photos for each part connected to the reactor chamber. We set six ICF 35 flanges on the reactor top as shown in fig. 7. The inlets that are shown at the center and the right side of the flange are for the high voltage terminals. The left side of the center high voltage terminal is the inlet of thermocouple set near the Pd wire electrode for measuring temperature. The right side is the inlet for the R type thermocouple in the heater terminal. The two flanges at the front/bottom are the spare terminals.



Figure 7: Photo of the reactor top; heater and high-voltage inlet.

In the photo of Figure 8, the valve shown at the bottom in the center is connected to the tube of the vacuum pump. The gray colored cylindrical part is MKS

Baratron, type 622 A, for measuring the absolute pressure of the transducer. This part has a temperature correction system. The instrument can measure pressure ranges from 0 to 10 kPa. At the bottom near the Baratron pressure transducer is a valve made by Swagelok Co. The valve is connected to the gas cylinder and it controls the D_2 gas flow into the reactor. A connection valve for the vacuum pump is shown at the bottom center of the photo. The sample gas is led to the mass analyzer through a flexible type connection tube. There are two other valves set as spares.



Figure 8: Side view of the reactor; vacuum and gas inlet pipe.



Figure 9: Photo of the reactor bottom connected with a vacuum pipe and other spare connectors.

3.2 Placement of the measuring instrument

The reactor is shown in the center of fig.10 together with the control system, the exhaust system and the gas analysis equipment. On the left side of the figure are the control and measurement instruments. The left side of the figure 10, from the bottom to the top, shows the input power supply, the power input analyzer (Yokogawa Co., type PZ4000), the high-voltage power supply (Matsusada Co., type HARb-3R200-L, 100V, 3 kV-200 mA), the current waveform analyzer (IWATSU, digital oscilloscope, type DS-8607, 60 MHz), the data logger (Agilent, 34970 A), and the PC for data collection. Neutron measuring instruments are shown above the reactor, which are the pulse measuring device made by Fuji Electric Co., LTD (neutron rem counter type NSN10014), the neutron intensity measuring device made by Aloka Co. (neutron survey meter of type TPES-451S), the γ -ray measuring device made by Aloka Co. (Scintillation survey meter of type TCS161), and the pressure measuring Baratoron, MKS Instruments.

Two K-type thermocouples are set on the reactor wall. These thermocouples are made by Chino Co. and are sheathed thermocouples of 1.6 mm and 3.2 mm in diameter respectively and mounted in two places. One is in the reactor and the second is attached on the wall. One is for measuring the cooling water and the other one is for measuring the room temperature. All the thermocouples are connected with the compensation type wire of 0.5mm in diameter to minimize the error of temperature measurement. The standard deviation at room temperature was ± 0.086 °C, which is equivalent to the energy measurement error of ± 2.3 kJ, as shown later in the heat calculation section. Other different R-type thermocouples are installed inside the ceramic heater. One thermocouple of 2 mm in diameter is mounted on the outside of the reactor wall at 15 cm from the top of the reactor. The second thermocouple of 4 mm in diameter is mounted on the outside of the reactor wall has been wrapped with aluminum foil with a thickness of 5 mm, five times. The thermocouples are attached to the outside.

Cooling water flows in the circulation pipe wound on the outside of the aluminum foil, and the pipe is made of plastic, with a 14 mm outer diameter, 10 mm inner diameter and 16 m in length. The cooling tube wraps around the reactor body, the flange of the viewing window, the flanges of the pipes, the top surface, and the bottom connection outlets. The connection parts include the bolts, the gas pipe, the electrode connecting port, and the 70 mm diameter flange at the bottom are not covered. Furthermore, we used aluminum foil to cover the entire reactor with heat insulating tape.

The outside is covered with aluminum foil. The outermost side, shown in figure 11 and 12, is covered with polyurethane foam, which is 20 mm in thickness.

By the adiabatic treatment, the temperature gradient on the reactor wall was expected to be kept minimum, so the heat and thermal calculations are simplified. Cooling water flows into the insulated dewar container, the temperature is measured inside the container by a thermocouple. Pure water with a volume of 4,000 cm³ is used as cooling water. 100 g of NaHCO₃ reagent is dissolved as a fungicide. Cooling water that passes through the tube wound around the reactor wall enters the bottom of the dewar container. The flow rate of the cooling water is 8,000 cm³/minute. The water in the dewar is thoroughly stirred, therefore the temperature in the dewar is uniform. The change of the temperature in the dewar was confirmed by the thermocouple placed at various locations.

The vacuum system and the quadrupole gas analyzer are illustrated on the right side in fig. 10. The connections are attached to a 152CF flange at the side of the reactor. The data from six temperatures, current, voltage, input power, gas pressure, neutrons emission, gamma rays emission, and change of ion currents from the Q-Mass-Spectrometer are collected with the logger and stored in the PC every 25 seconds. Only power analysis data is gathered in the data logger through the GPIB port. The changes of the gas pressure, temperatures, input power, radiations can be directly observed on the PC screen.



Figure 10: Schematic representation of measuring equipment

Figure 11 shows a picture of the experimental system. The measurement and signal processing system is shown on the left side, and the test equipment is shown on the right side. The equipment on the shelf from top to bottom of the photo includes a pressure measuring device, a waveform analyzer, a high voltage power supply, a power analyzer and a power supply. On the right side back is the reactor which is covered with thermal insulation material, polyurethane foam. In front of the reactor radiation detectors for neutron and gamma ray are placed.

Vacuum exhaust system and the mass analyzer are placed further to the right of the photograph. The D_2 and He gas cylinder can be seen behind the reactor. Data logger and PC are set on a table, as seen on the left side of the photo in fig. 11. The cooling water pipe, circulation pump and dewar container for the adiabatic calorie measurement of the reactor are seen at the right. The connection pipe for cooling water circulation is covered with insulation foam rubber. The lid of the dewar having holes to accommodate the pipes is made of Styrofoam with a thickness of 100 mm.



Figure 11: Adiabatic reactor system with flow calorimeter.

Figure 12 is a photo taken from the back-side and fig. 13 is a photo taken from the right side. The back-side view shows the gas cylinder. A copper pipe that is a connection pipe between the mass analyzer and the reactor can be seen on the side. Vacuum evacuation with a rotary pump was performed at the beginning to evacuate to 10^{-2} Pa, then a turbo-molecular pump of the mass analyzer was used to evacuate further to 10^{-5} Pa.



Figure 12: Back view of the reactor. Figure 13: R

Figure 13: Right view of the Dewar.

3.3 Heat measurement method

3.3.1 Thermal calibration concept

A conceptual scheme of the adiabatic calorimeter system is as shown in fig. 14.



Figure14: Conceptual diagram of calorimetry

The reactor is shown in fig. 14 as the portrait-shaped square in the center of the diagram. The cooling water reservoir is shown as smaller square to the right of that. The actual cooling water reservoir is the dewar vessel. A circulation pump circulates the water from the dewar through plastic pipes wrapped around the outside of the reactor surface.

This schematic shows both the reactor and the dewar vessel enveloped by the same insulation, but they are actually not. Conceptually they may be thought of as being under the same envelope. The cooling water (which includes 100 g of NaHCO₃ fungicide) is pumped from the bottom of the dewar by the circulation pump, and from there it enters the reactor section from the top to the bottom of the plastic cooling water

pipe coiled around the reactor. In this figure, the direction of the flow is shown with the gray arrows. The cooling water flows at a rate of 130 g per second, and the temperature distribution within the dewar is uniform. The reactor, the cooling water dewar, and the cooling water plastic pipe are all thermally insulated with foam plastic.

The circulation pump produces heat when it operates, and its temperature rises. Therefore, the circulation pump is cooled with a sirocco blower. In the figure, this blower is shown as the round figure under the pump. The circulation pump is rated at 12 W.

The temperature notation for each part of the calorimeter conceptual diagram is described as follows. The room temperature is represented by T_1 . The reactor temperature is equal to the cooling water temperature, shown here as T_3 . The circulation pump temperature is T_4 . (The subscript numbers of the temperature notation, 1, 3 and 4 refer to the column number in the Excel spreadsheet in which these temperatures are recorded. Temperature T_2 is the reactor core temperature, which is not shown in this conceptual diagram.)

The electric power input to the reactor core is shown as W_{in} . One hundred percent of this input power converts to heat inside the reactor. Some of the heat is consumed driving the anomalous reaction, but in the end it all emerges from the system. Some of the heat from the reactor heats the reactor wall, and from there it is transferred to the dewar vessel by the circulating cooling water. After the circulation pump has worked for a sufficient amount of time, the temperatures of the reactor vessel, and the cooling water reservoir dewar become uniform.

At this stage the total/integrated heat in the reactor and cooling water reservoir are designated as H_r . Some of the H_r heat dissipates to the outside through the insulation envelope (heat loss W_e in equation (1)). The amount of that heat is proportional to the temperature difference between the reactor and the outside; $(T_3 - T_1)$. This can be described with a simple one-dimensional heat diffusion equation. The thermal flux of the steady-state, W_e (W/cm²) is expressed by the following equation:

$$W_e = D_e (T_3 - T_1)/d - \dots$$
 (1)

where D_e is thermal conductivity (W/cm/deg)of the insulator, T_3 is the coolant and reactor temperatures, T_1 is the temperature of the ambient side of the insulator wall, and d is the thickness of the insulator wall.

The circulation pump also causes the temperature to rise. Some of the heat from the pump rotor passes through the pump walls into the cooling water. This amount is also proportional to the temperature difference between the pump temperature T_4 and the cooling water temperature T_3 : (T_4 - T_3). The amount of heat transfer in a steady-state

condition is similar to that shown in equation (1). The thermal power transferred from the circulation pump is denoted by W_P , and expressed in the steady state by the following equation:

 $W_p = D_P (T_4 - T_3)/d_p$ ------(2)

where D_P is thermal conductivity, d_p is the effective wall thickness between the cooling water and the pump rotor.

Without heat transfer from the pump (W_p) and the loss (W_e) , heat production rate (W_{out}) would equal the electric power input (W_{in}) .

This would be the case if there were no excess heat. However, adiabatic calorimeter heat losses cannot be completely prevented, even when insulated. Heat escapes through the insulating material surrounding the reactor, and from the walls of the circulation pump. Also, when the cooling water temperature rises sufficiently, there is energy loss from the evaporation of the cooling water. In a detailed analysis this loss would have to be taken into account. However the dewar vessel is covered with a thick 100 mm piece of Styrofoam, so evaporation is kept to a minimum and need not be considered in this analysis.

With this system, when

 $H_{out} = \int W_{out} dt = \int (W_r + W_e) dt > \int (W_p + W_{in}) dt$ ------(3) is obtained with $H_r = \int W_r dt$ being the accumulated thermal energy in the reactor system, it can be considered as an evidence that excess heat generation has occurred.

3.3.2 Thermal calibration test

The temperature data from the thermocouples placed at various locations are read into the data collection system at regular intervals. For each thermocouple, at time interval t_x , values of $T_1(t_x)$, $T_3(t_x)$, $T_4(t_x)$ are read, where *t* represents time, t_x is serial number (the spreadsheet row number) with x = 1, 2, 3, ..., n. In these data sets, the data were collected every 24.47 s, and the highest values of m were in the thousands. (See Table 2.)

The following items were computed for each data point: the integrated power into the reactor $(H_{in} = \int W_{in} dt)$, the heat of the reactor body (H_r) , heat losses from the reactor system through the insulation (W_e) , and heat transfer from the circulation pump (W_p) . These values were computed for each data point collected, for each of the thermocouples. The accumulated heat of the reactor (H_r) was obtained with the following equation.

 $H_r = M \times 4.18 \times \sum_1 [T_3(n) - T_3(n-1)]$ (J) ------(4)

where M is the combined thermal mass of the reactor vessel, the dewar vessel inner wall, the cooling water in the dewar and the cooling water pipe, all converted to the thermal mass of the coolant, for convenience. The factor 4.18 is the conversion factor for calories to joules.

The total amount of heat transferred from the reactor vessel to the outside environment (W_e) is expressed by the following equation:

In the same way, heat transfer from the pump is expressed in this equation:

 $H_p \equiv \int W_p \, dt = W_P \times \sum_1 {}^m [T_4(n) - T_3(n)] \, dt, \quad (J) \quad -----(6)$

Here, the sum \sum for equations (4), (5) and (6) can be expressed in the simplified form as follows.

$$H_{r} = 4.18M \times [T_{3}(m) - T_{3}(1)], \text{ (J)} ------(7)$$

$$H_{e} = W_{e} \times [\sum_{1}^{m} T_{3}(n) - \sum_{1}^{m} T_{1}(n)] dt, \text{ (J)} -------(8)$$

$$H_{p} = W_{p} \times [\sum_{1}^{m} T_{4}(n) - \sum_{1}^{m} T_{3}(n)] dt, \text{ (J)} -------(9)$$

By these formulae, the time-integrated thermal energies H_r , H_e and H_p can be obtained. Variables include the room temperature T_1 , the reactor temperature, which is assumed here to be equal to the cooling water temperature T_3 , and the circulation pump temperature T_4 , in three equations (7), (8) and (9). The values for the input energy, the room temperature, the temperature of the circulating pump have to be known when the variables change, to solve the equations.

The thermal calibration tests were conducted with all reactants removed from the reactor, and the gas pressure set at atmospheric pressure (100 Pa). Four types of calibrations are performed. First, (C1) was the calibration with no input power. The second and the third (C2, C3) tests were with fixed power levels. Under these conditions, the equations (7) ~ (9) are solved to obtain the factors. An example of a calibration would be changing input power to various different levels in order to determine the value of various factors. Once these are established, other factors can be evaluated.

Shown here first is a calibration test type (C1) with no input. Figure 15 and 16 show examples of tests with no input power and with 10 W continuous input.



Figure 15: Test for no input power.



Figure 16: Test for 10 W of continuous input power.

The temperature measurements in the reaction system at each thermocouple location during each calibration test are shown in fig. 17 and fig. 18. Figure 17 shows room temperature, the heater temperature, cooling water, circulation pump surface temperature, and the reactor wall temperature for two places. Test time is about 100 ks, we started the test at midday, and stopped the measurement next midday. Room temperature was 21 °C at the beginning of the test, and the circulating pumps were operated at the same time. After 3 ks, we started the test when all the temperatures with the exception of the circulating pump became stable and were constant. Then, at night, room temperature decreased to about 17 °C. There are slight changes in the room temperature.



Figure 17: Thermocouples at each position with no input power.

Figure 17 shows the temperature changes for thermocouples in each position in the system when there is no input power, as shown in fig. 15. The sum-total temperature changes to the reactor system in equation (7) are taken from changes in the cooling water temperature $[T_3(m) - T_3(1)]$, which were measured directly from the Excel spreadsheet at -0.152 °C.

Also, the term $[\sum_{1}^{m}T_{3}(n) - \sum_{1}^{m}T_{1}(n)]$ in equation (8) is used to compute the total heat output of the reactor system, and the sum of the temperature difference is +4,599°C while the input power indicated by the power meter showed 953J. Similarly, based on the term $[\sum_{1}^{m}T_{4}(n) - \sum_{1}^{m}T_{3}(n)]$ in equation (9), the heat produced by the circulation pump in the cooling water is determined by the sum of the temperature difference between the pump surface and the cooling water, which is 20,582°C.



Figure 18: Thermocouple temperature changes in the case of continuous input of 10 W.

Figure 18 shows readings from the thermocouples at each position in the system with 10 W of continuous input power. Unlike the calibration with no input, 10 W of power was begun 100 ks before this graph begins, and the reactor has reached a stable temperature. Room temperature and other temperatures are shown on the left vertical axis. Changes in room temperature produce temperature fluctuations of several degrees Celsius. There is a difference of 1 °C ~ 7 °C between the cooling water and the circulation pump surface, which is almost the same as it was with no input power. However, there is a large difference between the reactor wall temperature and cooling water temperatures, which is different from the test with no input power. The wall is about 5 °C ~13 °C higher than the circulation pump temperature. This difference is caused by the input power.

In a similar manner, the changes of the temperature in the bracket [] in the equations (7) ~ (9) can be obtained from fig.18. The sum of each of the temperature variations are -0.224° C, $31,582^{\circ}$ C and $-9,704^{\circ}$ C, respectively. During this test, the total input energy was 876,946 Joules.

By varying the input energy during calibration in this manner, changes in temperature can be related to the volume of heat produced by the overall reactor system. Conditionally you can derive factors in equations (7) ~ (9) where the total mass is converted to the thermal mass of the coolant, M. Because there are three types of heat transfer rates for W_e and W_p , three calibration points are sufficient. By using more calibration points, we can increase the accuracy of estimates for other factors. For this reason, we have used three points.

The total numerical value in terms of the temperature changes in equation (7), (8), and (9) of H_r , H_e , and H_p were estimated by the three calibration tests. And the following three results were obtained. This is the case of no input power of (C1), and the C2, C3 are the case of 10W of continuous input power. For this purpose, sufficient calibration data we needed was three cases. We increased the number of calibration for more number of factors in order to obtain the other factors. Here, we used three cases.

Sum of numerical centigrade of the temperature change for the term/deg

	Hin	$[T_3(m) - T_3(1)]$	$\left[\sum_{1}^{m}T_{3}(n)-\sum_{1}^{m}T_{1}(n)\right]$	$\left[\sum_{1}^{m}T_{4}(n)-\sum_{1}^{m}T_{3}(n)\right]$
(C1): 0W	953	-0.152	4,599	20,582
(C2): 10W	87,6946	-0.224	31,582	-9,703
(C3):10W	90,1768	-2.001	34,137	-11,421

The term "Input H_{in}/J " is the input in joules from the external electric power supply; H_r is the energy change of the overall reactor system comparing before and after the test; H_e is the total heat from the reactor system to the surroundings; H_P is the heat from the circulation pump. Total output (H_{out}) is the latter three types of values added together. External inputs and outputs are as shown in the following equation. The items in equation (3) left side are shown on the right side of this equation, and both sides of this equation will be equal if there is no excess heat. Therefore,

$$H_{out} = H_r + H_e = H_{in} + H_p$$

The temperature variation obtained with different levels of input energy for each factor in the above equation is obtained, and it can be solved by a matrix equation.

$$\begin{pmatrix} 953\\ 876,946\\ 901,768 \end{pmatrix} = \begin{pmatrix} -0.152 & 4,599 & 20,582\\ -0.224 & 31,582 & -9,703\\ -2.001 & 34,137 & -11,421 \end{pmatrix} \begin{pmatrix} Hr\\ He\\ Hp \end{pmatrix}$$

As a result, the three factors can be computed as follows:

Entire reactor system heat capacity, $M=8,334\times4.18$ J/deg Heat transfer rate, $W_e=-0.940$ J/deg Circulation pump – heat transfer rate to cooling water, $W_p=0.664$ J/deg

The heat capacity of the entire reactor system is 34.84 kJ/deg, which means that with 1 kW of input power it takes about 35 seconds to raise the temperature by 1 °C. If the temperature difference between the reactor and surrounding is 1 °C, that means 0.94 W of heat is flowing from the reactor to surrounding. When the circulation pump is running, a 1 °C difference between the cooling water and the pump indicates that the 0.664 W is flowing from the pump to the water. Typically, when the circulation pump is on, the temperature of the pump is 4.96 °C warmer than room temperature. Therefore, in normal conditions, the pump transfers a few W to the cooling water.

Using these values, we compute the heat flow, as shown in fig. 19 and fig. 20, based on the calibration conditions shown in fig. 15 and fig. 16.



Figure 19: Variation of heat flow at each location with no input power.

Figure 19 shows the case with no input, however because the insulation around the reactor is not perfect, there is some heat transfer from the reactor to or from the surrounding. After the circulation pump was turned on and run for 3 ks, the temperatures of the cooling water and all of the thermocouples in the reactor system have come into equilibrium.

As a result, the pump temperature is 5 $^{\circ}$ C higher than the reactor. After the test begins, when room temperature falls, the temperatures of the cooling water and reactor also fall. This is because several watts of heat are lost from the reactor body and cooling water to surrounding. In other words, the heat in the reactor system is reduced. This change is shown in the light purple line in fig. 19.

Contrary to this, the heat flow from the reactor body and cooling water is positive by a few watts. We can calculate this amount based on other factor, although the results are not shown here. For a number of reasons, there is not enough insulation around the cooling water pipes between the dewar reservoir and the reactor compared to other parts of the system. If we increase the number of calibrations, we can derive this factor from the results.



Figure 20: Heat flows with 10 W of continuous input, for all thermocouple locations.

Figure 20 shows an example of the case of the continuous input of 10W. There is some heat flow because the insulation around the reactor system is not perfect. The temperature of the circulation pump is about 5 C° warmer than the other parts of the system: because in this test the pump was already turned on for 84 ks before the start of data collection. After that when the ambient temperature rose slightly, the cooling water and reactor vessel wall temperature also increased. A few watts of heat flowed out from the reactor and cooling water, as shown in the light purple line in fig. 20. When the ambient temperature fell, the heat in the reactor body and cooling water decreased slightly, at a rate of about $-1 \sim -2$ Watts. Total heat flow was $1 \sim 10$ W because the temperature difference between the reactor body including coolant water and surrounding was large. The heat flow from the circulation pump to the cooling water is as little as $1 \sim 3$ Watt.



Figure 21: Calibration result with no input power.

Figure 21 shows the final heat flow results for a calibration with no input power. This is a summation of the heat from each of the thermocouple locations shown in fig. 19. The black line indicates the input power that was supplied by the electric power supply. And the output is obtained by adding H_r , W_e and W_p in fig. 19. Results at the beginning of the test were affected by large changes in the room temperature. The increase in the output between 60 ks and 70 ks was also due to heat flow from rapid room temperature changes. At 10 ks, when there was a 2 to 3°C temperature change, the input and the output balanced closely. In fig. 21, the overall input was as small as 0.95 kJ and the output was also so small as 0.81 kJ. The ratio of the output to the input is calculated to be 0.85. However, this value has no meaning, because the values for the input and the output were both very small.



Figure 22: The final calibration results for continuous input power of 10 W.

Figure 22 shows the result of a calibration with a continuous input power of 10 W. The output was obtained by adding the heat accumulated in the reactor system (H_r), the heat from the circulation pump (H_p), and the heat loss from the system (H_e). The black line indicates the input that was supplied by the electric power supply. The output is a moving average of 3 ks. The changes at the beginning of the figure are caused by a spreadsheet limitation; i.e., there is no data for the moving average. After that, the output power is stable, because there were no large changes in room temperature. Integrated output in this figure is estimated to be 876 kJ, and the integrated input was 877 kJ. Thus, the output/input ratio is almost 1.00.

3.3.3 Example Test

We performed excess heat experiments, and evaluated them using the results of these calibration tests. The input power was changed in a manner shown in fig. 23. The first and the second input were both 27 W by glow discharge, and the third was 51 W by the electric heater.



Figure 23: Input power change for excess heat generation test.

Figure 24 shows room temperature, heater, reactor wall, cooling water and pump temperatures.



Figure 24: Change in temperature at all thermocouple locations.

Figure 25 shows changes in heat flow and heat losses for the reactor system, the circulation pump heat flows, and heat losses from the pipes connecting the dewar and the reactor vessel.



Figure 25: Heat flows for various components.

Figure 26 shows the input power, the output power, and the output/input ratio for the excess heat generation test. As shown in Table 1, the output power is about twice as high as the input power in this test, hence, the excess heat is confirmed.



Figure 26: Result of excess heat generation.

Table 1 shows the integrated input, the integrated output and the output/input ratio in the three tests. In the tests of 1 and 2, the input power to the reactor was made with the glow discharge. In the test 3, it was made with the internal heater. The input power includes the heat from the circulation pump. In all the tests, the heat input and the output power were integrated from the time when the input was started until 3 ks after the input power was stopped. The first 2 results with the glow discharge show that excess heat was generated. In test number 3, the Out/In ratio was 0.76, which is 24% less than the calibration run. This is because we considered the heat generation only up until 3ks, even though the actual heat generation continued longer than 3ks.

Table1: Calculated input power, output power and the ratio

	Input/J	Output/J	Out/In
1:	22357	44679	2.09
2:	16468	30452	1.85
3:	51685	39385	0.76

In Table 3, all the results from the analysis performed in this manner are shown. It shows: experiment number, the D_2 gas pressure, the in input power, the total input energy(J), the computed output energy(J), and the output/input ratio.

Results shown in this table were obtained by varying gas pressure, the wattage of input power, and the duration of the test. The experiment number in the first column indicates the order in which the tests were performed. It is a serial number. These results show that initially the output was low. As the tests continued, the output increased, and then dropped off again. This is believed to be correlated with the extent to which the material was activated. The activation increases as more tests are performed, at first. Then as the number of test increases, and the duration of the input increases from a few hundred to several thousand seconds, the reactant temperature drops in each test, presumably because impurities accumulate on the reactant surface. When the test is performed with a sufficiently activated reactant, it can be seen that the excess heat exceeding the input is generated.

4. Conclusion

During the tests with the improved reactor, excess heat generation was observed when D_2 gas pressure is high and stable. We observed that when D_2 gas pressure was high, the excess heat was generated frequently. Hydrogen isotopes (light and heavy hydrogen) were the reactants. Hydrogen isotopes reacted when they were on the metal surface (or in the vicinity of the metal surface), which generated the excess heat. In these tests, nano metal particles acted as a catalyst, causing hydrogen to react and generate heat.

Based on the calibration value, these heat generations ranged from 10% to 100% of the input power. Approximately 4g of palladium was used as a catalyst in some tests, and approximately 46g of nickel was used in the other tests. Based on the total energy output with the palladium (4g), power density was approximately 5 watts per gram. With the nickel (46g), power density was approximately 1 watt per gram. These power densities are comparable to that of a nuclear reactor fuel.

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line number	Time/s	T ₁ /°C	T ₂ /°C	T ₃ /°C	T₄/℃	T ₅ /°C	T₀/℃	Input/W
1	5.30	17.069	221.604	27.008	23.341	30.799	30.24	10.446
2	27.98	17.061	221.56	27	23.357	30.806	30.235	10.443
3	52.48	17.108	221.565	27.013	23.431	30.794	30.25	10.421
4	77.09	17.121	221.578	27.005	23.492	30.801	30.243	10.429
5	101.58	17.149	221.596	27.003	23.54	30.791	30.227	10.447
6	125.91	17.159	221.575	27.018	23.535	30.804	30.227	10.396
7	150.41	17.193	221.534	27.01	23.594	30.806	30.245	10.415
8	174.91	17.201	221.478	27.003	23.574	30.804	30.255	10.399
9	199.35	17.229	221.483	27.036	23.645	30.804	30.248	10.43
10	223.74	17.27	221.455	27.036	23.716	30.811	30.232	10.387
11	248.18	17.281	221.447	27.028	23.783	30.809	30.25	10.447
12	272.73	17.335	221.452	27.046	23.821	30.819	30.258	10.379
13	297.34	17.358	221.452	27.039	23.862	30.806	30.235	10.444
14	321.78	17.397	221.447	27.036	23.915	30.801	30.24	10.387
15	346.27	17.438	221.413	27.049	23.941	30.811	30.237	10.435
16	370.50	17.456	221.403	27.054	23.959	30.811	30.253	10.416
17	395.16	17.492	221.46	27.031	24.025	30.819	30.237	10.412
18	419.44	17.513	221.48	27.046	24.033	30.827	30.245	10.419
19	444.04	17.541	221.519	27.041	24.084	30.824	30.273	10.432
20	468.65	17.575	221.519	27.051	24.135	30.824	30.253	10.362
21	493.04	17.601	221.455	27.051	24.219	30.824	30.25	10.433
22	517.53	17.611	221.375	27.064	24.319	30.834	30.245	10.388
23	542.03	17.657	221.372	27.072	24.426	30.824	30.248	10.393
24	566.42	17.688	221.336	27.046	24.502	30.809	30.271	10.406
25	590.86	17.724	221.354	27.069	24.543	30.816	30.245	10.381
26	615.35	17.758	221.28	27.074	24.597	30.829	30.243	0.326
27	639.91	17.794	221.156	27.064	24.648	30.822	30.24	10.452
28	664.35	17.838	221.072	27.074	24.663	30.822	30.263	10.439
29	688.68	17.864	221.102	27.069	24.712	30.804	30.237	10.467
30	713.07	17.892	221.177	27.069	24.778	30.799	30.253	10.433
31	737.84	17.949	221.2	27.072	24.875	30.796	30.255	10.387

Table2: Sample data of excel table.

Test no.	Pressure/Pa	Input/W	Input/J	Output/J	Rate Out/In
1	10	22	22330	29361	1.315
2	15	22	21656	28056	1.296
3	150	22	31328	40288	1.286
4	130	24	22218	30607	1.378
5	10	25	23551	24074	1.022
6	12	25	22632	25129	1.110
7	20	25	21806	25811	1.184
8	110	22	13874	25140	1.812
9	118	23	13580	16630	1.225
10	5	22	15565	19746	1.269
11	2	22	16733	22508	1.345
12	2	22	15378	16245	1.056
13	1	21	17087	22302	1.305
14	1	23	13388	11716	0.875
15	165	22	13626	24029	1.763
16	165	22	13967	20530	1.470
17	155	22	14930	23685	1.586
18	300	22	14155	17708	1.251
19	285	22	13243	24378	1.841
20	380	50	26210	39396	1.503
21	380	52	27374	41921	1.531
22	380	21	11650	26634	2.286
23	380	20	10926	19847	1.816
24	380	50	26701	34379	1.288
25	5	52	26862	45527	1.695
26	5	102	52821	81037	1.534
27	5	52	28021	46465	1.658
28	1	21	12044	20495	1.702
29	1	100	50319	71850	1.428
30	5	52	25389	37958	1.495
31	5	21	10925	19800	1.812
32	6	99	50692	66673	1.315
33	18	23	23397	26669	1.140
34	8	23	23719	27824	1.173
35	69	82	47540	63790	1.342
36	77	22	48730	53619	1.100
37	75	22	89972	96597	1.074
38	74	99	96640	142004	1.469
39	85	80	94325	98550	1.045

Table3: Result of excess heat generation

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Background for Condensed Cluster Fusion

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Abstract: For explaining the experimentally claimed anomalous excess heat phenomena in metal-D(H) systems, the condensed cluster fusion (CCF) theory has been proposed and elaborated[1-8] since 1989. This paper reviews the latest status of CCF theory development. The paper explains the following key aspects: classical mechanics and free particle fusion, fusion rate theory for trapped D(H) particles, strong interaction rate, condensation dynamics of D(H)-clusters, final state interaction and nuclear products, and sites for Platonic D(H) cluster formation on/in condensed matter.

Keywords: condensed cluster fusion, rate theory, condensation dynamics, nuclear products, cluster formation sites

1. Introduction

The theoretical model of condensed cluster fusion (CCF) of deuterons has been proposed and elaborated since 1989 [1-8], for possible underlying key mechanism of so called cold fusion in the dynamic microscopic environment of condensed matter.

The 4D/TSC model is a typical case of CCF for deuterium multi-body interaction under transient ordering process of d-e-d-e-d-e tetrahedral symmetric condensate (TSC). Here d denotes deuteron and e does electron. For making analysis of time-dependent D(H)-cluster condensation motion, the theory of quantum-mechanical (QM) Langevin equation was developed [1, 2] and a PC-based computation code was provided [8]. For studying likely products of final state interactions of the compound excited nucleus ⁸Be*(Ex = 47.6 MeV), a nucleon halo model was applied to speculate the major energy damping process by BOLEP (burst-of-low-energy-photons) and final ash of two 46 keV ⁴He (alpha) particles.

The model has been extended to the weak-strong (WS) force combined fusion of 4H/TSC (p-e-p-e-p-e tetrahedral symmetric condensation) [9]. Here p denotes proton.

Some speculative models for TSC cluster formation sites in/on nano-scale catalytic surface zone or inner lattice-defects have been proposed [3, 10, 11], although quantitative studies are yet to develop. A scenario for computational simulation of dynamic process of many deuterons (or protons) with electrons in modeled sites will be proposed in this

paper.

This paper reviews so far the elaborated works and future problems to see the developing status of CCF theories.

2. Classical Mechanics and Free Particle Fusion

In the main stream nuclear physics, two-body collisional process of free particles is regarded as extremely dominant process for nuclear fusion events. Three-body or more multi-body collision events can be neglected in such random motion of particles, as in the cases of plasma thermo-nuclear fusion and beam-target type particle-condensed matter interactions. Once two-body fusion cross section data are available, we can make reaction rate estimation by using classical mechanics solvers such as Maxwell-Boltzmann particle transport equations with statistical averaging over particle kinetic energy distribution and spatial distribution.



Fig.1: Image of classical mechanics to treat two-body random fusion event

So, we do not use quantum mechanics (QM) explicitly there for estimating reaction rates or power levels of fission reactors and thermo-nuclear fusion reactors. And we are forgetting that fusion cross sections data have been laboriously evaluated by analyzing experimental data with huge quantum mechanical nuclear physics calculations for twobody interactions, as have been done for evaluated nuclear data libraries such as ENDF/B- VI and JENDL-4 (see their contents by internet search).

For studying newly some unknown nuclear reactions, especially by trapped particles with finite lifetime in chemical potential well in condensed matter, including possibly enhanced multi-body nuclear interactions, we need however to start with quantum mechanics of theoretical tools. An image is illustrated in Fig.1. The classical Newtonian motion assumes particle to be point (size zero), as shown in the left figure, so that we have no chance to make collision between two particles because of zero sizes of particles. However, once finite size cross section (by QM study) is given, as shown in the right figure, particles have chance to make collision. By the QM nuclear physics formulas [4], two-body collision cross section becomes proportional to the square of transition matrix. Such two-body random collision cross sections may be approximately used for some cases of cold fusion analysis [4]. However, if the time interval of trapped particles (deuterons or protons, for instance) in relatively negative potential well of condensed matter is much longer than the collision interaction time (typically 1.0E-22 s for twobody collision), we need to use the Fermi's first golden rule for fusion reaction rate estimation [4], as we recall in the following section. The reaction rate formula becomes proportional to single (not square) transition matrix. Because of elongated life time in trapped state, fusion reaction rate is very much enhanced as calculated for muonic d-d pair molecule [7, 8]. The Thomas-Fermi type charge screening estimation formulas derived for free particles becomes no good approximation, and we need to use real trapping potential having negative well for screening and barrier penetration probability calculation [1, 2].

3. Fusion Rate Theory for Trapped D(H) Particles

As we have studied for the case of D(H)-cluster condensation motion [1, 2], meaningful enhancement of fusion rate (barrier factor of larger than 1.0E-20 is of key issue, in the rough view of Avogadro number 6.023E+23 as order of maximum particle density of condensed matter) may happen by the condensation collapse [8] time-dependently. Some combination of deuterons (protons) and electrons for formation of transient cluster goes to a stable ground state and has no dynamic state to enhance fusion reaction rate to 'visible or detectable' magnitude, as we review in the next section. Therefore, we need to treat particle trapping state in time varying potential well of D(H)-cluster.



Fig.2: Time-dependent trapping potential, d-d pair Gaussian wave function for a pseudo-stable adiabatic state for a very small time-step width and strong-interaction fusion domain

An image is illustrated in Fig.2. In the collapsing condensation motion, trapping potential shape changes continuously from right to left to become deeper with narrower width as time elapses. To treat the state in numerical calculation with small time-step, we approximate a state at a time to be an adiabatically pseudo-steady state of d-d pair with heavy-mass quasi-particle e*(m, Z) of electron pairing (the HMEQPET method [1, 2]) and Vs1(m, Z) type potential (namely generalized Heitler-London type molecular trapping potential) are used. Using the variational principle of QM, inter-nuclear distance (d-d or p-p distance) and its ground state mutual kinetic energy (energy eigen-value) are calculated [1, 2].

To apply the Fermi's first golden rule, we add the nuclear optical potential (Vn + iWn type) to a Vs1(m*, Z*) potential, in the flow of methodology, to formulate nuclear fusion rate equation. However, for the sake of simplicity to follow the QM equations, we simply write the complex potential by its real part (V) plus imaginary part (W). And also for simplicity we use one dimensional space variable only in the following, to avoid complexity of mathematical equations. By doing so, essence of derivation for fusion rate does not change.



Fig.3: We need to treat one way process of three steps, the initial state interaction, the intermediate state and the final state interaction for studying a nuclear reaction

We are going to derive the rate formula for the initial state interaction (see Fig.3). Explanation for the final state interaction with nuclear products will be treated later.

The forward and adjoint (backward) QM Schroedinger equations are:

$$i\hbar\frac{\partial\Psi}{\partial t} = \left[-\frac{\hbar^2}{2M}\nabla^2 + V + iW\right]\Psi\tag{1}$$

$$-i\hbar\frac{\partial\Psi^{*}}{\partial t} = \left[-\frac{\hbar^{2}}{2M}\nabla^{2} + V - iW\right]\Psi^{*}$$
⁽²⁾

By multiplying Ψ from the left side to Eq.(1) and Ψ^* from the left side to Eq.(2), we make subtraction to get,

$$i\hbar\frac{\partial\rho}{\partial t} = -\frac{\hbar^2}{2M} \left[\Psi * \nabla^2 \Psi - \Psi \nabla^2 \Psi *\right] + i \left[2W\rho\right] = -i\hbar div\bar{j} + i \left[2W\rho\right]$$
(3)

Here $\rho = \Psi \Psi^*$ is the particle QM density, and we used the QM current density formula [4]. The second term of the right-hand-side of Eq.(3) shows the particle disappearing (by fusion) rate for negative imaginary part W of the nuclear optical potential.



Fig.4: Born-Oppenheimer approximation is adopted to rate calculation. Inter-nuclear wave-function is calculated independently (adiabatically) to electro-magnetic (EM) filed wave function.

The fusion rate formula by the Fermi's first golden rule is then obtained as,

$$< FusionRate >= \frac{2}{\hbar} \left\langle \Psi_f | W(r) | \Psi_i \right\rangle$$
(4)

Here suffixes i and f denotes the initial and final state.

Assuming the adiabatic separation of particle (pair or cluster) wave function into two components of the nuclear force field (very short range force) and the electro-magnetic force field (long force range), we solve two independent Schroedinger equations as illustrated in Fig.4 through Fig.6.

The inter-nuclear fusion rate, the bracket integral of W(r) has been estimated by empirical extrapolation of PEF-value dependence of known two-body reactions as p-d, d-d and d-t fusions, in which PEF value is very roughly counted based on the d-d interaction as illustrated in Fig.7. Here PEF denotes the pion exchange force.


Inter-Nuclear QM Schroedinger Equation:

$$-\frac{\hbar^2}{2m}\nabla^2\Psi_n(r) + [V_{nr}(r) + iW(r)]\Psi_n(r) = E_n\Psi_n(r)$$

Outer-Nuclear QM Schroedinger Equation for Electro-Magnetic Field:

$$-\frac{\hbar^2}{2m}\nabla^2\Psi_c(r)+V_c(r)\Psi_c(r)=E_c\Psi_c(r)$$

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Fig.6: Fusion rate formula for trapped pair (cluster) in condensed matter is written with the product of inter-nuclear fusion rate (nuclear transition matrix element) and Coulomb barrier penetration probability (barrier factor).



Fig.7: Defining a scale of strong fusion interaction force by PEF value



Fig.8: Strong force by one pion exchange potential, cf. Coulomb force Effective <W> values estimated by the empirical extrapolation is listed in Fig.9.

Cluster	<w>(MeV)</w>	$< W > = \langle \psi n f W(R) \psi n i \rangle$
DD	0.008	Inter-nuclear wave functions are governed by the real part of optical potential V(R), having
DT	0.115	Woods-Saxon type well shape that makes inter-nuclear wave functions very
3D	1.93	localized near around V(R). Therefore, <w> becomes approximated by the</w>
4D	62.0	surface sticking at R=R0.

Fig.9: Evaluation procedure of effective transition matrix <W> for two-body and multi-body fusion, estimated by rough extrapolation of known two body reaction values (p-d, d-d and d-t astrophysical S-factors) as a function of effective transition matrix approximation of PEF powered by 5

4. Condensation Dynamics of D(H)-Cluster

The QM Langevin equation has been generalized for treating D(H)-clusters with Platonic symmetry, which means the configuration of orthogonal coupling between deuterons- (or protons-) wave function and electron wave function (or density function) in three dimensional space.

The features of 4D/TSC electron distribution and known electron density distributions for D-atom and D₂ molecule are compared in Fig.10. To derive the QM Langevin equation for a D₂ molecule dynamics calculation, ensemble averaging of QM-observables was done with the Born-Oppenheimer separation of electron-wave function and deuteron-pair wave function, as shown in Fig.11. Used electron wave function is the Heitler-London type well known QM solution (by the variational principle), which is the linear combination of product 1S wave functions according to two deuteron-centers. And the dd pair wave function was approximated with Gaussian distribution [1, 2]. For the case of a d-e-d three body system (D₂+ molecule), electron wave function is the linear combination of 1S wave functions according to two deuteron centers. The QM Langevin equation for the d-e-d system is shown in Fig.12, which includes information of ground state parameters. The Vs1(1,1) trapping potential of this d-e-d three body system has been used for a triangle face of Platonic polyhedron to formulate the generalized QM Langevin equation of polyhedrons of many-particles clusters. The generalized QM Langevin equation is shown in Fig.13.



Fig.10: Comparison of electron density distributions between D(H)-atom, D₂ molecule and 4D/TSC (at t=0)



Fig.11: QM ensemble averaging of an observable is done with the Born-Oppenheimer adiabatic wave

function separation between electron wave function and deuteron pair wave function. Figure is shown for the sample case of D_2 molecule



Fig.12: A QM Langevin equation for a d-e-d three-body system (regarded as a cluster) and key ground state parameters as Rdd, Re, electron kinetic energy



Fig.13: Derivation of generalized QM Langevin equation for D(H)-cluster condensation dynamics

calculation

Cluster	k value (keVpm)	Ne	Nf
2D+: d-e-d	0	1	1
3D+	3.65	3	4
4D/TS	11.85	6	4
6D/RD	18.45	12	8
8D/RD	18.45	12	6
12D/RT	15.51	30	20
20D/RT	19.11	30	12





Fig.14: An example of QM Langevin code calculation [8] for d-e-d cluster. Oscillation with 32 fs period continues eternally. We repeat calculations by changing starting Rdd (t=0) value, and make ensemble averaging of oscillations to get ground state behavior.

Evaluated parameters for Langevin code calculations for various clusters are summarized in Table-1.

In Fig.14, example of calculation is shown for a d-e-d cluster for two oscillation cycles. The minimum approaching distance between two deuterons is about 30 pm, barrier factor for which is about 1.0E-60 (see Table-3) to be negligible for d-d fusion events. By changing starting Rdd (t=0) values, we obtain a number of oscillation curves. Ensemble average of these curves converges to the ground state oscillation trajectory, which has eigen-value of Rdd (gs)=138pm. Maximum mutual kinetic energy of deuterons is about 20 eV for a small time interval in the transient motion.

Similar calculation for d-e-d-e-d five body cluster (D_3 + molecule) is shown in Fig.15. Minimum approaching d-d distance is about 20pm, for which the barrier factor for fusion is on the order of 1.0E-45 to be too small to detect d-d or d-d-d fusion events. Maximum relative kinetic energy of deuteron is about 18 eV and ground state d-d distance is 85pm.



Fig.15: Dynamic behavior (oscillation) of D_3 + molecule (d-e-d-e-d five body cluster), calculated by the QM Langevin code [8]

Most interesting results of similar calculations are of collapsing condensation (one way to collapse), happened for larger clusters as 4D(H)/TSC, 6D(H)/RDC and 8D(H)/RDC. Example of collapse is shown in Fig.16 for the case of 4D/TSC. Here RDC denotes Rohmbic dodecahedron condensation [8]. More detailed feature of near collapse state was shown in Ref. [2] by inverting time axis. In the present calculation, we used Vs1(1,1) trapping potential to get collapsing time 3.61 fs which is larger than 1.4 fs [1, 2] by using the Vs1(2,2) potential assuming bosonized electron pairs on every rectangular face of TSC-configuration.



Fig.16: Calculated results of condensation collapse for 4D/TSC

Summary of condensation motions for various clusters is shown in Table-2. Detail was discussed in Ref. [8].

Summary of Results			
Cluster Type	Collapse? (Rdd-min ≤ 10 fm)	Rdd (gs)	Remarks
d-e-d (2D+)	Ν	138 pm	
d-µ-d	Ν	0.79 pm	DD fusion in 0.1ns
3D+	N	85 pm	
4D/TS	Y		100% 4D fusion
6D/RD	Y		100% 6D fusion
8D/RD	Y		100% 8D fusion or 4D fusion?
12D/RT	Ν	ca. 80 pm	
20D/RT	Y		What kind of fusion?

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Table-2: Summary of condensation/collapse motions for various D(H)-clusters

	Barrier fac Based on Expansion	ctors were calc the Heavy Ma n Theory (HME	ulated time-de ss Electronic G QPET)	pendently ausi-Particle	
Rdd	=R _{gs} (pm)	P2d; 2D Barrier Factor	P _{4d} ; 4D Barrier Factor	QM Tunneli	ng Probability:
0.00 0.04 0.20 0.4 0.80 1.00 2.00 4.10 200 4.11 10.12 21.3 (dd	206 412 03 06 12 05 (μ dd) 3 6 2 3 8 e*(2,2)	4.44E-2 1.06E-2 1.43E-3 3.35E-5 9.40E-7 1.00E-9 9.69E-11 6.89E-15 9.38E-21 2.16E-32 1.30E-46 1.00E-85	1.98E-3 1.12E-4 2.05E-6 1.12E-9 2.16E-13 1.00E-18 9.40E-21 4.75E-29 8.79E-41 4.67E-64 1.69E-92 1.00E-170	$P_{nd}(m,Z) =$ $\Gamma_{dd}(m,Z) = 0.218$	$\exp(-n\Gamma_{dd}(m,Z))$ $= \sqrt{\mu} \int_{r_0}^{r_0(m,Z)} \sqrt{V_z(R;m,Z) - E_d} dR$
74. mol	1 (D2 lecule)				33

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Table-3: Calculated time-dependent barrier factors by HMEQPET code [1, 2]



Fig.17: Estimation of 4D-fusion yield per TSC condensation

When condensation-collapse happens, d-d (or p-p) inter-nuclear distance of cluster becomes very short to be less than 20 fm in its final stage before getting into strong nuclear force range (ca. 5 fm typically for d-d). To calculate fusion rate (simultaneous 4D fusion becomes dominant for 4D/TSC [1, 2]), we need to estimate time-dependent barrier penetration probability. The HMEQPET (heavy mass electronic quasi-particle expansion theory) method [1, 2] is an easy mathematical tool for approximate numerical calculation of Gamov integral. Calculated results by HMEQPET code are shown in Table-3.

In the case of 4D/TSC condensation/collapse, 100% 4D fusion (to go to the ${}^{8}\text{Be*}$ intermediate excited nucleus) was estimated to be happening within about 2.0E-20 s time interval (see Fig.17) in the final stage of condensation-collapse.

5. Final State Interaction and Nucleon Halo Model

It is well established that the final state interaction of two-body d-d fusion happens through the intermediate ${}^{4}\text{He}*(\text{Ex} = 23.8 \text{ MeV})$ excited nucleus which then breaks up to two major branches of n + ${}^{3}\text{He}$ + 3.25 MeV and p + t + 4.02 MeV. The EM (electromagnetic) transition of ${}^{4}\text{He}*(\text{Ex} = 23.8 \text{ MeV})$ to ground state ${}^{4}\text{He}$ happens with very small

branching ratio (ca. 1.0E-7) for lower mutual kinetic energy of d-d fusion less than 1 keV. For the 'cold fusion' by muonic d-d molecule (see Table-2) [8], the mutual d-d kinetic energy is ca. 180 eV and branching ratios for the three out-going channel do not change.



Fig.18: Simplified steps of 4D/TSC condensed cluster fusion

A simplified steps of reaction scheme is copied [3] in Fig.18 for the 4D/TSC condensed cluster fusion.

According to Fig.3, the step-1 to step-2 corresponds to the initial state interaction. The step-3 is the intermediate compound excited state and the step-4 is the final state interaction. We consider that the 4D fusion makes the very highly excited state of ⁸Be, namely ${}^{8}\text{Be}*(\text{Ex} = 47.6 \text{ MeV})$, for which no evaluation studies in main stream nuclear physics are available and we do not know what kind of final state interaction happens (see TUNL library for A=8and ⁸Be energy level scheme • http://www.tunl.duke.edu/nucldata/figures/08figs/08_04_2004.pdf).

We have made a speculative analysis by proposing the nucleon halo model of ⁸Be* [5]. Comparing with inner nucleon clustering (alpha, helion, and triton) model for highly excited states of light nuclei such as ⁸Li, ⁹Li, ¹¹Li, ⁸Be, ⁹Be, ¹¹Be, ¹²Be, ¹²C, etc., with the neutron-state nucleon halo, we speculated that the ⁸Be*(Ex = 47.6 MeV) would be a n-h-h-n halo excited state as illustrated in Fig.19: Here h is helion (p-n-p) inner nuclear cluster, and t (n-p-n) is the triton inner nuclear cluster.



Fig.19: The ⁸Be*(Ex = 47.6 MeV) intermediate compound excited state by the 4D/TSC fusion is modeled as a n-h-h-n neutron halo state.

The n-h-h-n halo state looks similar to the thought n-h-t-n halo state of ⁸Li which has 'very long' (in the view of nuclear physics) life time as 0.838 s and makes beta-decay (weak interaction) only, due to its ground state that prohibits EM transition, to transit to the ${}^{8}\text{Be}*(\text{Ex} = 3.03 \text{ MeV})$ first excited level state that breaks up to two alpha particles. Since the ${}^{8}\text{Be}*(\text{Ex} = 47.6 \text{ MeV})$ is excited state, it has freedom to make EM transitions if life time of the state is so 'long' as that of ⁸Li. And the excited state would be very highly deformed (m=16 spherical harmonics picture was given [5]) nucleus with many modes/nodes of rotation/vibration coupled states, which would generate bosonic coupling (nuclear phonons) of low energy band quanta (so called rotation-vibration band structure of QM energy levels). We speculated several ten-thousands bosonic levels might couple. If so, we may expect an avalanche type multi-photon emission, namely BOLEP (burst of low energy photons), with mean photon energy ca. 1.5 keV. However, we need to consider other minor decay channels via possible discrete energy levels of ⁸Be* by α - α clustering, d-⁶Li clustering, and p-⁷Li clustering. Detail discussions were made in reference [5] comparing with some experimental data: Especially, it was impressive to find the beautiful coincidence of several discrete alpha-energy peaks below 17 MeV between the halo model and the Roussestki experiment [5]. The speculated final state interaction of ${}^{8}\text{Be}*$ (Ex = 47.6 MeV) is copied [5] in Fig.20.



Fig.20: The nucleon-halo model for ⁸Be* speculates this complex final state decay channels. The BOLEP is thought to be major energy damping to the ground state ⁸Be.

Now we switch the halo model for the 4H/TSC WS (weak-strong) fusion process [6, 9]. A simplified steps of reaction is shown in Fig.21. Since there are no strong nuclear force (roughly modeled by PEF; charged pion exchange force) between protons, 4H/TSC condensation collapse does not end at around Rpp = 20 fm (around where 4D/TSC disappears 100% by 4D fusion of strong interaction) and continues to condense.



Fig.21: Simplified steps of 4H/TSC WS fusion process [9]



Fig.22: The weak interaction scheme for electron capture to a proton. Via weak boson W-, inner quark configuration changes to that of neutron. Here u denotes up quark and d does down quark.

It was discussed [5, 9] that the collapse state of 4H/TSC would reach the Pauli-type repulsion domain of ca. 2fm p-p distance of tetrahedron and electron kinetic energy would

reach more than 1 MeV. We speculated the possibility of ca. 3% electron capture to a proton (weak interaction as shown in Fig.22) to generate a neutron. This just born neutron would have to make immediate strong interaction (PEF = 3) with three protons existing within the range of charged pion Compton wave length (1.4 fm) for generating the intermediate excited compound nucleus ${}^{4}\text{Li}*(\text{Ex} = 4.62 \text{ MeV})$. The decay channels of ${}^{4}\text{Li}*(\text{Ex} = 4.62 \text{ MeV})$ excited state is well studied (see TUNL data for ${}^{4}\text{Li}$ energy levels and decays). If we apply the idea of halo state, it would be a h-p halo or p-d-p halo with very weak PEF binding [6] and with very short life (ca. 1.0E-23 s) to break up to p + ${}^{3}\text{He}$ + 7.72 MeV and/or p + p + d + 2.22 MeV promptly.

A view of condensation/collapse of 4H/TSC dynamic motion is shown in Fig.23.



Fig.23: Time-reversed graph of 4H/TSC condensation motion [3], with 1.0 condensation time

One electron may make WI (weak interaction) with a proton due to very short range (2.5am) of W- and no visible multi-body WI is expected. Theoretical approach on very condensed (in several fm p-p distance) 4H/TSC dynamical state getting into relativistic QM state with spin-spin interaction. There electron mean kinetic energy becomes to require relativistic QM treatment. We need to modify QM-Langevin equation or apply Dirac equation for TSC configuration of the multi (8)-particle system.

If the effective life time of 4H/TSC-minimum is on the order of 1 fs or longer, we may expect about 3% or larger ⁴Li* formation per 4H-cluster to see rational excess power level as 100W/mol-Ni for the Ni-H experiment. We need further study for the effective life rime of collapsed state of 4H/TSC to be more accurately estimated.

6. Sites for Platonic D(H) Cluster Formation

We need to estimate finally the macroscopic condensed cluster fusion rate by modeling D(H)-cluster formation sites and formulate computation formulas and schemes for timedependent so many-body problem of deuterons (protons) plus electrons around the sites. A general QM formula is shown in Fig.24.



Fig.24: macroscopic fusion rate formula for condensed cluster fusion

A series of theory has been developed (albeit one-through) for calculating cluster fusion rate per cluster generation as explained in the above-written sections. The remained and most complex (and difficult) problem is the cluster formation rate estimation. To approach it, we have to define sites of condensed cluster formation first. Some models for sites have been proposed [3, 10, 11] by getting hints by nano-metal D(H)-gas loading experiments [10-15]. We are looking for the mesoscopic catalysis effect in near surface zone of mono-atomic metal nano particles of Pd and Ni, binary metal nano-particles of Pd-Ni, Cu-Ni, etc. dispersed in some ceramics support material (zirconia and mesoporous-silica were tried in experiments as shown a general idea in Fig.25 [3]).





AT ICCF17TSC theory 6

Fig.25: Illustration of image for mesoscopic catalyst for CCF fusion



Fig.26: Sub-nano holes (SNH) on surface or inner defects of metal nano-particle may provide sites for CCF

Typically, a three dimensional model of surface SNH (see Fig.27) will be first attacked for simulation calculation of transient 4D(H)/TS (TSC at t=0) generation rate.



Fig.27: A candidate model of site on surface of mesoscopic catalyst for 4D(H)/TS formation



Fig.28 : Speculative pseudo trapping potential of nano-metal particle as mesoscopic catalyst [3, 11]

We need to study <u>the catalytic interaction of H(D)-gas and nano-metal particles if the</u> <u>transient BEC type D(H) Platonic clusters [3] are efficiently formed</u>.

So, the QM mathematics there is much more complicated for solving time-dependent many-body system under mesoscopic D(H) trapping potential like Fig.28. Maybe, we need a <u>step-by-step approach with many divided adiabatic states of dynamic processes</u> as surface H_2 (D₂) adsorption, oscillation in trapped potential in the second modified trapping potential for the next incoming H_2 or D₂ molecule, formation rate of transient H(D)-cluster, competition of its dissociation and cluster condensation, penetration of dissociated H and D into inner trapping sites (O-sites and T-sites, for instance) of metal nanoparticle, non-linear oscillation of trapped H(D) in global mesoscopic potential well (GMPW), transient formation of Platonic cluster by non-linear oscillation, estimation of CCF rates, etc. Application of the electron density functional theory (DFT) for such complicated states will be expected by step-by-step computation trials.

It's challenging problem for finding mathematical solvers, especially with use of timedependent DFT methods.

7. Summary Remarks

For explaining the experimentally claimed anomalous excess heat phenomena in metal-D(H) systems, the condensed cluster fusion (CCF) theory has been proposed. This paper has reviewed the latest status of CCF theory development. In each section, the following key aspects are reviewed and discussed: classical mechanics and free particle fusion, fusion rate theory for trapped D(H) particles, strong interaction rate, condensation dynamics of D(H)-clusters, final state interaction and nuclear products, and sites for Platonic D(H) cluster formation on/in condensed matter.

For studying newly some unknown nuclear reactions aka cold fusion, especially by trapped particles with finite lifetime in chemical potential well in condensed matter, including possibly enhanced multi-body nuclear interactions, we need to start with quantum mechanics of theoretical tools. For the case that the time interval of trapped particles (deuterons or protons, for instance) in relatively negative potential well of condensed matter is much longer than the collision interaction time (typically 1.0E-22 s for two-body collision), we need to use the Fermi's first golden rule for fusion reaction rate estimation. The Thomas-Fermi type charge screening estimation formulas derived for free particles becomes no good approximation, and we need to use real trapping potential having negative well for screening and barrier penetration probability calculation.

To apply the Fermi's first golden rule, we have used the nuclear optical potential (Vn + iWn type) to be added to a Vs1(m*, Z*)-type particle (D or H) trapping potential of the Coulombic field of condensed matter, to formulate nuclear fusion rate equation. The internuclear fusion rate, the bracket integral of W(r) has been estimated by empirical extrapolation of PEF-value dependence of known two-body reactions as p-d, d-d and d-t fusions, in which PEF value is very roughly counted based on the d-d interaction.

The QM Langevin equation has been generalized for treating time-dependent condensation motion of D(H)-clusters with Platonic symmetry, which means the configuration of orthogonal coupling between deuterons- (or protons-) wave function and electron wave function (or density function) in three dimensional space. The QM Langevin equation has been generalized for treating D(H)-clusters with Platonic symmetry, which means the configuration of orthogonal coupling between deuterons- (or protons-) wave function and electron wave function (or density function) in three dimensional space. The Vs1(1,1) trapping potential of the d-e-d three body system has been used for a triangle face of Platonic polyhedron to formulate the generalized QM Langevin equation of polyhedrons of many-particles clusters. The time-dependent minimum approaching distance between two deuterons of the d-e-d three body system was calculated to be about 30 pm, barrier factor for which is about 1.0E-60 to be negligible for d-d fusion events. The ground state oscillation of d-e-d system has eigenvalue of Rdd (gs)=138pm. Maximum mutual kinetic energy of deuterons is about 20 eV for a small time interval in the transient motion. Similar calculation for d-e-d-e-d five body cluster (D₃+ molecule) was done. Minimum approaching d-d distance is about 20pm, for which the barrier factor for fusion is on the order of 1.0E-45 to be too small to detect d-d or d-d-d fusion events. Maximum relative kinetic energy of deuteron is about 18 eV and ground state d-d distance is 85pm. Most interesting results of similar calculations are of collapsing condensation (one way to collapse), happened for larger clusters as 4D(H)/TSC, 6D(H)/RDC and 8D(H)/RDC. When condensation-collapse happens, d-d (or p-p) inter-nuclear distance of cluster becomes very short to be less than 20 fm in its final stage before getting into strong nuclear force range (ca. 5 fm typically for d-d). The HMEQPET (heavy mass electronic quasi-particle expansion theory) method is an easy mathematical tool for approximate numerical calculation of Gamov integral of barrier factor. In the case of 4D/TSC condensation/collapse, 100% 4D fusion (to go to the ⁸Be* intermediate excited nucleus) was estimated to be happening within about 2.0E-20 s time interval in the final stage of condensation-collapse. A nuclear heat-power level of 1 W can be expected by ca. 2.0E11 4D/TSC fusions per second.

We have considered that the 4D fusion makes the very highly excited state of ⁸Be,

namely ⁸Be*(Ex = 47.6 MeV), for which no evaluation studies in main stream nuclear physics are available and we do not know what kind of final state interaction happens. We speculated that the ⁸Be*(Ex = 47.6 MeV) would be an n-h-h-n halo excited state similar to the n-h-t-n halo state of ⁸Li which has long life time as 0.838 s. Since the ⁸Be*(Ex = 47.6 MeV) is excited state, it has freedom to make EM transitions if life time of the state is so 'long' as that of ⁸Li. And the excited state would be very highly deformed (m=16 spherical harmonics picture was given) nucleus with many modes/nodes of rotation/vibration coupled states, which would generate bosonic coupling (nuclear phonons) of low energy band quanta (so called rotation-vibration band structure of QM energy levels). We speculated several ten-thousands bosonic levels might couple. If so, we may expect an avalanche type multi-photon emission, namely BOLEP (burst of low energy photons), with mean photon energy ca. 1.5 keV. The BOLEP process may be main energy damping mechanism of ⁸Be*(Ex = 47.6 MeV) to the ground state ⁸Be(0+) which breaks up to two 46 keV alpha-particles.

A series of theory has been developed (albeit one-through) for calculating cluster fusion rate per cluster generation as explained in Sections 2 through 5. The remained and most complex (and difficult) problem is the cluster formation rate estimation. To approach it, we have to first define sites of condensed cluster formation. Some models for sites have been proposed by getting hints by nano-metal D(H)-gas loading experiments. We are looking for the mesoscopic catalysis effect in near surface zone of mono-atomic metal nano particles of Pd and Ni, binary metal nano-particles of Pd-Ni, Cu-Ni, etc. dispersed in some ceramics support material (zirconia and mesoporous-silica were tried in experiments). The QM mathematics there is much more complicated for solving timedependent many-body system under mesoscopic D(H) trapping potential. Maybe, we need a step-by-step approach with many divided adiabatic states of dynamic processes as surface H₂ (D₂) adsorption, oscillation in trapped potential in the second modified trapping potential for the next incoming H₂ or D₂ molecule, formation rate of transient H(D)-cluster, competition of its dissociation and cluster condensation, penetration of dissociated H and D into inner trapping sites (O-sites and T-sites, for instance) of metal nanoparticle, non-linear oscillation of trapped H(D) in global mesoscopic potential well (GMPW), transient formation of Platonic cluster by non-linear oscillation, estimation of CCF rates, etc.

Further efforts are expected to complete the CCF theory.

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Convergence Aspect of the Self-consistent Calculations for Quantum States of Charged Bose Particles in Solids

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Abstract

In our previous works, we proposed the method to calculate the quantum states of the positively charged two bosons trapped in the ion trap in order to investigate the nuclear reactions in solids. These studies consist of the self-consistent calculations. They are done by obtaining the numerical solutions for the Schrödinger's and the Poisson's equations alternately. However, the selfconsistencies of them were not always adequate. In this study, we have tried higher frequency ion trap potentials and obtained better self-consistencies.

(keywords; Bose-Einstein condensation, nuclear reaction in solids)

1. Introduction

In ICCF17 and JCF14, we proposed the density functional method how to calculate the quantum states of the system including two species of charged bosons in ion traps [1] and showed the results for some calculations [2,3]. They were based on the Kim's theory which explains the mechanism of nuclear reactions in solids using Bose-Einstein condensation (BEC) [4,5].

In this problem, following two factors are important. One is the repulsive interactions between positively charged bose particles confined in ion traps. The other is the quantum mechanical distribution of them. They will determine the overlaps of particles and the nuclear reaction rates. However, the quantum mechanical distributions of interacting many-body are complex problems. In this paper, the methods how to calculate the distributions and some results are shown. Especially, the relation between the convergence performances in the recursive calculations and the shapes of the trapped potentials are discussed.

2. Density functional formalism

The formulas used in our previous works [2,3] are briefly introduced here. At the

beginning, the quantum states of the charged bose particles i (= 1, 2) with mass m_i are described by Schrödinger equation, which is written as

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

where V_i , W_i and μ_i are harmonic, electro-static and chemical potentials, respectively. The chemical potential should be selected to give similar damping solution at large $|\mathbf{r}|$ to those of the pure harmonic problems. The normalized solution of this equation gives the existing probability density of the particle *i* which is written as

$$n_i(\mathbf{r}) = |\psi_i(\mathbf{r})|^2. \tag{2}$$

In Eq.(1), harmonic potentials are electromagnetically induced by ion traps. In real lattices, they correspond to the expressions for the potential wells around the O and T sites. It is written as

$$V_i(\mathbf{r}) = \frac{1}{2}m_i\omega_i^2 r^2,\tag{3}$$

where ω_i means angular frequency factor for the particle *i*. The curvature of the harmonic potential means strength of the repulsive interactions between impurity ions and host metal ions, therefore we can assume

$$\frac{m_i \omega_i^2}{Z_i} = \frac{m_j \omega_j^2}{Z_j}.$$
(4)

On the other hand, the electro-static potentials for particle i with effective charge Z_i are written in the integral form of Poisson's equation as

$$W_i(\mathbf{r}) = \int d\mathbf{r}' \frac{\exp(-k|\mathbf{r} - \mathbf{r}'|)}{4\pi|\mathbf{r} - \mathbf{r}'|} \left[\frac{e^2}{\varepsilon_0} Z_i \left\{ Z_i n_i(\mathbf{r}') + Z_j n_j(\mathbf{r}') \right\} + k^2 W_i(\mathbf{r}') \right], \quad (5)$$

where e and ε_0 mean elementary charge and dielectric constant of vacuume, respectively. We can easily understand that this is a density functional formalism. The parameter k can be chosen arbitrarily, because Eq.(5) is an integral form of the equation which is made by subtracting $k^2 W_i(\mathbf{r})$ from both sides of Poisson's equation. However, we confine it to be a positive real constant in order to give this parameter a role of a damping factor in Eq.(5). In this case, the volume integrations in Eq.(5) over all spaces are easily approximated by the calculations for the finite regions.

If we assume the functions $n_i(\mathbf{r}')$, $n_j(\mathbf{r}')$ and $W_i(\mathbf{r}')$ in the integrand of the right hand side of Eq.(5) as old quantities, $W_i(\mathbf{r})$ in the left hand side can be regarded as a new quantity in the recursive calculations. In this procedure, Eq.(1) and Eq.(5) are doubly connected, because both of the functions $n_i(\mathbf{r}')$ and $n_j(\mathbf{r}')$ are included in Eq.(5) and they are given by the solutions of Eq.(1) for each case.

The flow chart of our calculations for doubly connected problems is shown in our previous paper [3]. The fundamental theories of BEC by Kim et al. are explained in their papers [6,5].

3. Boundary conditions

When we perform numerical calculations, Eq.(1) is translated in the usual manner for the harmonic potential problems into

$$\left\{-\frac{d^2}{d\xi_i^2} + \xi_i^2 + v_i(\xi_i)\right\} u_i(\xi_i) = \varepsilon_i u_i(\xi_i), \tag{6}$$

where $\xi_i = \alpha_i r$, $\alpha_i = \sqrt{m_i \omega_i / \hbar}$, $\varepsilon_i = 2\mu_i / \hbar \omega_i$, $v_i = 2W_i / \hbar \omega_i$ and $u_i(\xi_i) = \xi_i \psi_i(\xi_i)$. In this equation, v_i can be regarded as the perturbation to the harmonic potential problem at large ξ_i . For the case of the unperturbed system, we can write down the ground state solution corresponding to $\varepsilon_i^0 = 3$ as

$$u_i^0(\xi_i) = \xi_i e^{-\xi_i^2/2}.$$
(7)

On the other hand, the perturbed ground state for Eq.(6) is unknown. In particular, the perturbed eigenvalue ε_i is unknown. In this study, we found appropriate ε_i which gave

$$\frac{d}{d\xi_i}\log u_i(\xi_i) = \frac{d}{d\xi_i}\log u_i^o(\xi_i) \tag{8}$$

at the boundary of $\xi_i = \xi_i^{max}$ by using Newton method. This equation is conve-

nient, because $\frac{d}{d\xi_i} \log u_i(\xi_i) = u'_i(\xi_i)/u_i(\xi_i)$ is normalization free. After finding the appropriate eigenvalue, the normalizations of u_i are done. The results also satisfy Eq.(8).

When we solve Eq.(6) numerically, well known Runge-Kutta-Gill method is used. The calculations are started from the origin toward the large ξ . Mathematically, there are two possibilities for the initial conditions at the origin. The first possibility is written as

$$u_i(0) = 1 \tag{9}$$

$$u_i'(0) = 0, (10)$$

where the even function $u_i(\xi)$ is constant near the origin. On the other hand, the second possibility is

$$u_i(0) = 0 \tag{11}$$

$$u_i'(0) = 1, (12)$$

where the odd function $u_i(\xi)$ is proportional to ξ near the origin. The first conditions express the states that have high probability around the origin, while the others expresses the low probability states around the origin. We should choose the initial conditions for each particle from the two possibilities.

4. Results and Discussions

For the simplicity, we tried numerical calculations for the mixture of one D^+ and one ${}^{6}Li^{+}$ in ion traps. In pure harmonic one body problem, the particle exists most probably around the origin. In this problem, D^+ and ${}^{6}Li^{+}$ have same charges and they interact repulsively, so we make following choices. The first initial condition in Eq.(9) and Eq.(10) are adopted for D^+ . The second initial condition in Eq.(11) and Eq.(12) are used for ${}^{6}Li^{+}$. Of course, the reverse choice is possible, however we choose it as an example.

In order to investigate the relation between the magnitudes of the trapping angular frequencies and the convergences of the calculations, we tried three cases. The trapping angular frequencies used in our calculations are listed in Table 1. In each Run, the relation between angular frequencies for D^+ and ${}^{6}Li^+$ are expressed by Eq.(4). In all cases, the recursive calculations were repeated 16 times.

	D^+	⁶ Li ⁺
Run 1	4.79×10^{14}	2.77×10^{14}
Run 2	4.79×10^{16}	2.77×10^{16}
Run 3	9.59×10^{22}	5.54×10^{22}

Table 1. Trapping angular frequencies for D^+ and ${}^{6}Li^+$ [sec⁻¹]

The results for Runs 1, 2 and 3 are plotted in Figs.1-5, 6-10 and 11-15, respectively. For each Run, energy eigenvalue versus iteration, total potentials, probabiliy densities are plotted. Seeing Fig.1, convergences of energy eigenvalues in Run 1 are not adequate. The total potentials plotted in Figs. 2 and 3 are similar with harmonic potentials except for the region around the origin. There are effects of interactions between D^+ and ${}^{6}Li^{+}$ around the origin. The convergences of the probability densities plotted in Figs. 4 and 5 are also not adequate.







Fig. 2. Total potential of D^+ for Run 1



Fig. 4. Probability density of D^+ for Run 1



Fig. 3. Total potential of $^{6}\mathrm{Li^{+}}$ for Run 1



Fig. 5. Probability density of $^{6}\mathrm{Li^{+}}$ for Run 1

Seeing Fig.6, convergences of energy eigenvalues in Run 2 are improved more than Run 1. In this case, trapping angular frequencies are larger than those of Run1. This may be a cause of these results. In Figs. 7 and 8, the effects of interactions between D^+ and ${}^6\text{Li}^+$ around the origin become smaller. The convergences of the probability densities plotted in Figs. 9 and 10 are also improved.







Fig. 7. Total potential of D^+ for Run 2



Fig. 9. Probability density of D^+ for Run 2



Fig. 8. Total potential of $^{6}\mathrm{Li^{+}}$ for Run 2



Fig. 10. Probability density of $^{6}\mathrm{Li^{+}}$ for Run 2

Seeing Fig.11, convergences of energy eigenvalues in Run 3 are extremely improved, because the trapping angular frequencies in Run 3 are much larger than other cases. The total potentials plotted in Figs. 12 and 13 are nearly similar with harmonic potentials in all regions. The effects of interactions between D^+ and ${}^6Li^+$ around the origin become quite small. We can see the adequate convergences of the probability densities plotted in Figs. 14 and 15.



Fig. 11. Energy eigenvalue for Run 3 versus iteration



Fig. 12. Total potential of D^+ for Run 3



Fig. 14. Probability density of D^+ Run 3



Fig. 13. Total potential of ⁶Li⁺ Run 3



Fig. 15. Probability density of $^{6}\mathrm{Li^{+}}$ Run 3

As we know, the frequencies assumed in Run3 belong to the γ -ray region. From the experimental point of view, such situations may be unrealistic. However, the purpose of this study is to investigate the states of charged bosons in crystals through the numerical calculations for ion traps. In near future, we will give the shape of the trapping sites in crystals necessary for the nuclear reactions between charged bosons.

5. Conclusions

In this paper, we used potentials produced by ion traps [1] in place of real lattice potentials, because they are quite similar around the bottoms of the potentials. The merit of using ion traps are variability of the trapping potentials. Tuning the electric and magnetic fields applied to the chamber of the devices, we can change the magnitude of the trapping angular frequencies of the harmonic potentials.

We tried three harmonic potentials listed in Table 1. Seeing the results, we can conclude that the larger the trapping angular frequencies are, the better the convergences are. This is consistent with the intuitive investigation, because the deep potential wells bound particles tightly.

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Computer Simulation of Hydrogen Phonon States

in Palladium Metal

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Abstract: We simulated the hydrogen (H) phonon states in palladium (Pd) metal by using a quantum molecular dynamics on personal computers, in order to examine which conditions gather H or deuterium (D) atoms in condensed matter like a Pd metal to cause nuclear fusion or nuclear transmutation. We calculated the phonon energy and phonon density of states of the bulk Pd metal. We used a computer simulation program based on Density Functional Theory of the local density approximation using a plane-wave basis and pseudo-potentials. These calculations were done on the lattice of conventional 1x1x1 cubic unit cell, being imposed periodic boundary conditions. The lattice was perfect composed of four Pd atoms or defective with a vacancy, and it occluded H atoms on all O sites or all/half T sites with or without an impurity atom. We observed that the Pd metal lattice was nearly structurally stable for the phonon vibration when it occluded H atoms not only on all O sites or all/half T sites but also on those around the vacancy. Especially, even when impurity lithium (Li) atom entered a vacancy or an O site of the H-occluded Pd metal lattice, we could observe that it was structurally stable for the phonon vibration. **Keywords:** computer simulation, quantum molecular dynamics, phonon response, hydrogen, palladium

1 Introduction

A lot of nuclear fusion and nuclear transmutation phenomena have been reported in experiments of hydrogen (H) and deuterium (D) within condensed matter such as metals. When these nuclear reactions occur in condensed matter, it will be necessary for more than one H or D atoms to gather and condense in the narrow space of the condensed matter structure. Then we have simulated the H states in metals of face centered cubic (FCC) lattice such as palladium (Pd), calculating the total energy of H-occluded metals by using a quantum molecular dynamics on personal computers (PC) before. At that time, we have observed that the total energy change on a large scale when the interatomic distances change due to the impurity atoms entered the metal lattice ¹⁾. This time, we investigated the phonon responses of H-occluded Pd metal lattice to see which phonon vibrations would take place, calculating the phonon mode energy and the phonon density of states.

We carried out the phonon response simulation on the smallest 1x1x1 calculation unit sell. We used the perturbation method which showed the phonon vibration caused by the restoring force of the small lattice deformation due to the stress on a part of the metal lattice. We obtained the phonon dispersion curves or the phonon mode energy at Γ , X, M and R points in the reciprocal phonon wave-vector space. It would be recognized that the lattice could be structurally stable for the phonon vibration when the composed atoms take place harmonic oscillations by the proper restoring force of the small lattice deformation, and then we could observe that the each square energy of the individual phonon mode is positive. But when the atoms do not take place harmonic oscillations, the each square energy of the individual phonon mode would be negative and the lattice would be structurally unstable. On that occasion, the occluded H atoms or the entered impurity atoms could not stay there.

We observed that the Pd metal lattice which occluded H atoms on the whole O sites (four in the 1x1x1 unit cell) was nearly structurally stable for the phonon vibration as expected. And the Pd metal lattice which occluded H atoms on the whole T sites (eight in the 1x1x1 unit cell) was nearly structurally stable. Interestingly, it was shown that the Pd metal lattice which occluded four regular tetrahedral H atoms on the T sites in the 1x1x1 unit cell was also nearly structurally stable for the phonon vibration. Especially in these cases, the Pd metal lattice with an impurity lithium (Li) atom which entered the center vacancy of regular tetrahedral H atoms held this structural stability for the phonon vibration. Although this time we carried out the simulation mainly with occluded H atoms, we could reason by analogy the similar results for occluded D atoms, taking account of mass difference between H atom and D atom. Then we expect that the phonon vibration will change to the coherent breathing mode vibration of four regular tetrahedral D atoms when the impurity Li atom hops out from the vacancy, and four D atoms will move collectively to the center of them. Since these four D atoms are weakly recoiled because of the light mass of impurity Li atom, they would squeeze into the condensation to cause the nuclear fusion of Tetrahedral Symmetric Condensate Theory², ³⁾. And it was shown that the Pd metal lattices with an impurity Na, Mg or Ca atom which entered the vacancy were nearly structurally stable for the phonon vibration when they occluded H atoms on the whole O sites. We also expect in these cases that the phonon vibrations will change to the coherent breathing mode vibration of six regular octahedral D atoms when the impurity Ca atom hops out from the vacancy, and the six D atoms will move collectively to the vacancy. Since these six D atoms are strongly recoiled because of the heavy mass of impurity Ca atom, they would squeeze into the

condensation and collide against the other atomic nucleus to cause the nuclear transmutation of Octahedral Symmetric Condensate Theory ^{2, 3)}.

But the calculation cell using this time is too small and the vacancy concentration is too large. So the phonon states of Pd metal lattice with the impurity Li or Ca atom which entered the vacancy might be possibly the ghost states by unreasonable calculation. Then after this, to get more accurate results, the calculations would have to be done with some different vacancy concentrations in larger calculation cell such as 2x2x2 conventional unit cells. Later we would analyze the interatomic force constants and displacements of composed atoms relating to the phonon vibration. And we would also investigate the transition of H phonon states and also D phonon states.

2 Calculations

2.1 Hardware and Software Used for Calculation

We used two personal computers which had 4 cores / 8 ways CPU with 32 GB main memory and 16 GB main memory. And we used the first principles electronic state simulation program ABINIT. This program was based on Density Functional Theory of the local density approximation using plane waves for the basis function and norm preservation type pseudo-potentials with semi core^{4, 5)}. Moreover we used VESTA for visualization of the charge density of calculation results⁶⁾.

2.2 Process of Calculation

We calculated the phonon response of the smallest 1x1x1 unit conventional simple cubic cell by using the perturbation method, being imposed the periodic boundary conditions. This method showed that some phonon vibrations were caused by the restoring force of the small lattice deformation due to the stress on a part of the metal lattice. Calculations were done on the perfect Pd metal lattice or defective one with a vacancy, H atoms which were occluded on O sites or T sites and with or without an impurity atom of Pd lattice atom, H atom and alkali/alkaline-earth metal atom such as Li, Na, K, Mg and Ca.

2.2.1 Calculations with Two Lattice Constants

We could not optimize the lattice arrangement because of computational complexity and calculation time. Then, except a few cases of the Pd metal lattice with a vacancy, phonon response calculations were done with the following two lattice constants which were smaller or larger than those of experiments, expecting the optimum lattice arrangement to be between or near them.

• The first lattice constant is not expanded before the H atoms are occluded or the impurity atom enters.

• The second lattice constant is expanded in order for the total energy of the calculation unit cell to become the lowest while the H atoms are occluded or the impurity atom enters.

2.2.2 Calculations of Phonon Response by Perturbation Method

We calculated the phonon energy, their second derivatives and dynamical matrixes and so on at the 8x8x8 sampling points of reciprocal space by using the cut off energy of about 1 keV. Because the real calculation space was sat as the 1x1x1 conventional simple cubic cell, the reciprocal space was the same cubic cell, and $\Gamma(0, 0, 0)$, X(1/2, 0, 0), M(1/2, 1/2, 0) and R(1/2, 1/2, 1/2) were selected as the special symmetric points in the unit $2\pi/a$ rad where a was lattice constant. On the database of response calculation, the phonon mode energy were estimated at the wave vectors for regular intervals between two reciprocal special symmetric points to get the phonon mode dispersion curves, and the phonon density of states were estimated by summation of phonon mode states number which corresponded to the each phonon mode energy.

2.2.3 Calculations of Phonon Energy at the Special Symmetrical Points

We had to calculate the response calculations at 35 reciprocal space points to get the phonon dispersion curves mentioned above, and we took the considerable long calculation time using the PC (maximum about two weeks). Then we calculated the phonon mode energy only at the special symmetric Γ , X, M and R points in order to obtain the outline of structural stability for the phonon vibration in short calculation time. These results of phonon state calculations were written in tables of appendix.

2.3 Stable or Unstable of Lattice Structure for Phonon Vibration

The dispersion relation of the linear-chain model of a one dimensional diatomic crystal lattice which is based on two atoms with mass M and m respectively is described as below,

$$\omega^{2} = f (1/M + 1/m) \pm f [(1/M + 1/m)^{2} - 4 \sin^{2} (\mathbf{q} \cdot \mathbf{a}) / Mm]^{1/2}$$
(1)

where ω is the angular frequency (phonon energy $E = h / 2\pi \cdot \omega$), **q** is the phonon wave vector, **a** is repeat distance (lattice constant) and f is the spring constant (interatomic force) which is taken to be a harmonic oscillator. There appear two phonon modes; the

lower frequency solution is known as the acoustic mode in which two atoms oscillate in the same phase and the phonon energy equal to zero at Γ point (q = 0), the upper frequency solution is known as the optical mode in which two atoms oscillate in the opposite phase and the phonon energy is larger than zero at Γ point.

When there are n atoms in the three dimensional unit cell, three acoustic phonon modes which energy equal to zero at Γ point appear, and 3n - 3 optical phonon modes which energy are larger than zero at Γ point appear. And the dispersion relation would be represented with the combination of above-mentioned equation (1) for one dimensional diatomic crystal lattice. According to this dispersion equation, the square of the angular frequency ω happen to be negative due to the relation between mass and interatomic force. On that occasion, although the angular frequency ω which is proportional to the phonon energy become imaginary, we would usually describe it as a conventional negative energy in the dispersion relation. Then the negative description of phonon mode energy in the dispersion relation would mean that the crystal lattice is structurally unstable for the phonon vibration due to the improper restoring force of harmonic oscillation in the calculation unit cell. Similarly, in the three dimensional crystal lattice, when the negative description of phonon mode energy is written at Γ point, the calculation unit cell would be structurally unstable for the phonon vibration at the normal temperature and the normal pressure. And when the negative description of phonon mode energy is written at X, M or R point, the calculation unit cell would be structurally unstable for the phonon vibration of each particular direction. In these cases, the occluded H atoms or the entered impurity atom would not stay there.

3. Results

According to Rayleigh's theorems ⁷⁾, it is shown that the higher phonon mode energy increases in the case where the mass of occluded H atom or impurity atom which enters the interstitial site is smaller than those of lattice atoms, or in the case where the interactions between the occluded or impurity atom and the lattice atoms are larger than those of the lattice atoms. And in the opposite cases, the lower phonon mode energy decreases. We obtained nearly same results from the phonon response calculations.

3.1 H atoms Occluded on O sites or T sites

In the perfect metal of FCC lattice such as Pd metal which has no vacancy or other defects, H atoms are usually occluded on O sites at the normal temperature and the normal pressure, and they are occluded on T sites at the increased pressure of H atoms. Then we simulated the phonon states in some cases where the H atoms occluded in the
Pd metal lattice of 1x1x1 unit cell. Fig. 1 shows the phonon dispersion curves of the Pd metal lattice with (A) No occluded H atom, (B) Occluded H atoms on the whole O sites, (C) Occluded four regular tetrahedral H atoms on the T sites and (D) Occluded eight regular hexahedral H atoms on the T sites in the each unit cell. These were calculated with the expanded interatomic distances in order for the total energy of the calculation unit cell to become the lowest while the H atoms were occluded or the impurity atom entered. The outline of the related phonon states are written in Table A1 of Appendix.

Since the perfect Pd metal which occludes no H atom has the phonon mode energy of zero or positive ((A): about 0.01 ~ 0.03 eV), it would be structurally stable for the phonon vibration. When the Pd metal occludes H atoms, the low energy phonon modes which correspond to those of the perfect Pd metal slightly go down, and the high energy phonon modes in which mainly the H atoms vibrate appear above them ((B) ~ (D): about 0.05 ~ 0.15 eV).





Figure 1 Phonon Dispersion Curves of Pd Metal Lattice

(A) No occluded H atom: Pd4

(Four Pd atoms in the calculation unit cell)

- (B) Regular octahedral H atoms on the whole O sites: Pd4H4-O (Four Pd atoms with four occluded H atoms on the O sites)
- (C) Four regular tetrahedral H atoms on the T sites: Pd4+H4-T (Four Pd atoms with four occluded H atoms on the T sites)
- (D) Eight regular hexahedral H atoms on the T sites: Pd4+H8-T (Four Pd atoms with eight occluded H atoms on the T sites)

The phonon modes of the Pd metal lattice with four regular tetrahedral H atoms on the T sites come together more narrow energy width (C) than those of Pd metal lattice with eight regular hexahedral H atoms on the T sites (D), both in which mainly H atoms vibrate. The former have nearly unchanged phonon modes energy at about 0.12 eV. In the perfect Pd metal, since these phonon modes energy are much smaller than the potential barrier between O site and T site (partial free energy: about 1 eV) and these phonon vibration occur randomly as the thermal vibration, it would not be reasonable that four or eight H atoms simultaneously move to gather by superposition of these several phonon modes without any trigger reaction.

3.2 H atoms Occluded on O sites or T sites around a Vacancy

Generally there are some vacancies in the metal (concentration $10^{-5} \sim 10^{-3}$). When H atoms exist in the metal, vacancies seem to increase (concentration $10^{-5} \sim 10^{-2}$) and there are several H atoms around the vacancy, composing vacancy-hydrogen clusters. (In our simulation of this time, we observed that the binding energy for one H atom which was occluded on O site or T site around a vacancy in the Pd metal was slightly larger than that of the perfect Pd metal. And we observed that the binding energy for one H atom was largest when four or six H atoms were occluded.) Then we simulated the phonon states in some cases where a vacancy existed and H atoms were occluded in the Pd metal lattice of 1x1x1 unit cell. Fig. 2 shows the phonon dispersion curves of the Pd metal lattice with (A) No occluded H atom, (B) Occluded H atoms on the whole O sites, (C) Occluded four regular tetrahedral H atoms on the T sites and (D) Occluded eight regular hexahedral H atoms on the T sites in the each unit cell. These were calculated with the expanded interatomic distances in order for the total energy of the calculation unit cell to become the lowest while the H atoms were occluded or the impurity atom entered. The outline of the related phonon states are written in Table A2 of Appendix.





Figure 2 Phonon Dispersion Curves of Pd Metal Lattice with One Vacancy

(A) No occluded H atom: Pd3Vac1

(Three Pd atoms with one Vacancy in the unit cell)

(B) Regular octahedral H atoms on the whole O sites: Pd3Vac1H4-O

(Three Pd atoms with one Vacancy and four occluded H atoms on the O sites)

- (C) Four regular tetrahedral H atoms on the T sites: Pd3Vac1+H4-T (Three Pd atoms with one Vacancy and four occluded H atoms on the T sites)
- (D) Eight regular hexahedral H atoms on the T sites: Pd3Vac1+H8-T (Three Pd atoms with one Vacancy and eight occluded H atoms on the T sites)

Since the Pd metal lattice with a vacancy whichever occludes no H atom or occludes H atoms on the whole O sites has the slightly negative phonon mode energy at M point, it is structurally unstable for this point direction phonon vibration (A), (B). When the Pd metal lattice with a vacancy occludes four regular tetrahedral H atoms on the T sites around the vacancy, high energy phonon modes in which mainly H atoms vibrate separate to three groups and they become higher (C) than those of the perfect Pd metal lattice mentioned above Fig. 1-(C). And separated lower energy phonon modes in which mainly H atoms vibrate has also nearly unchanged narrow phonon modes energy of about 0.1 eV similar to those of the perfect Pd metal lattice Fig. 1-(C), and they would indicate standing waves.

In the vacancy-hydrogen cluster there is no barrier which exists between the T site and O site of the perfect Pd metal lattice. However, in the Pd metal with a vacancy, since these phonon mode vibrations occur randomly as the thermal vibration, it would not also be reasonable that four or eight H atoms simultaneously move to gather by superposition of these several phonon modes without any trigger reaction.

3.3 One Impurity Atom Entered Vacancy, O site or T site

When there are vacancies in the metal lattice, impurity atoms could move and diffuse, staying or going through vacancies, O sites and/or T sites. Then we simulated the phonon states in some cases where a vacancy existed and H atoms were occluded and further a Pd lattice atom, an H atom or an alkali/alkaline earth metal atom such as Li, Na, K, Mg or Ca entered a vacancy, an O site or a T site of the Pd metal lattice of 1x1x1 unit cell. The phonon states of the Pd metal lattice with (A) No occluded H atom, (B) Occluded H atoms on the whole O sites, (C) Occluded regular tetrahedral four H atoms on the T sites and (D) Occluded eight regular hexahedral H atoms on the T sites (or others) of the unit cell were calculated. The outline of the calculated phonon states are written in Table A3 ~ A13 of Appendix.

We observed that generally the Pd metal lattice held the structural stability for the phonon vibration when the impurity atom such as Pd lattice atom or the alkali/alkaline

earth metal atom of Li, Na, K, Mg or Ca entered the vacancy. And we observed that the structural stability was slightly lost when one of these impurity atoms entered the vacancy of the Pd metal lattice with occluded H atoms, but the structural stability was perfectly lost when the impurity H atom entered the vacancy. On the other hand, only when the impurity H or Li atom entered the O site, the Pd metal lattice with three H atoms which occluded on the O sites or with four H atoms which occluded on the T sites in the each unit cell held the structural stability for the phonon vibration. Especially only when the impurity H atom entered the T site, the Pd metal lattice could hold the structural stability for the phonon vibration.

3.3.1 Impurity Li Atom Enters a Vacancy

We observed that the Pd metal lattice nearly held the structural stability for the phonon vibration when an impurity Li atom entered a vacancy which placed at the center of H atoms on O sites (four in the 1x1x1 unit cell) or the center of four regular tetrahedral H atoms of T sites. We expect that these four H atoms will gather, starting from four of six O sites where H atoms usually stayed and ending to four regular tetrahedral T sites, when the impurity Li atom moves from the vacancy to the O site. Table 1 show the related phonon states which are selected from Tables A2, A4, A5 and A13 of Appendix. And Fig. 3 shows one of the charge density distributions of these lattice structures. In Fig. 3, we can see Pd atoms (silver) with H atoms (yellow) on the T sites and Li atom (green) in the vacancy, and they are surrounded by charge (light green and blue).



Figure 3 Charge Density Distributions of Pd3+H4-T+Li1-Vac Silver indicates Pd atoms, yellow H atoms, green Li atom and Light green indicates high charge density and blue indicates low charge density.

In the Table 1 below, we show the phonon states of non-expanded interatomic distances in the upper rows, describing the structure of calculation unit cells, and we show those of expanded interatomic distances in the lower rows, describing the structure of calculation unit cells added "-ex", both in the left column. Furthermore, we show whether the phonon mode energy is zero (0), nearly zero (~0) or negative (-) at the Γ point and positive (+) or negative (-) at the X, M and R points in the middle column, and we show where the minimum (--) or maximum (++) phonon mode energy appears. And we also show the binding energy of the sub-structure which joins with the three Pd metal atoms (Pd3) of 1x1x1 unit cell in the right column, adding the lattice constant used for the calculations. The binding energy was estimated by the subtraction of total energy of Pd3Vac1 from that of the whole joined Pd3 and sub-structure.

			Г	х	М	R	Binding Energy (eV)	
		Pd3Vac1H4_0	0	+		+ ++	-63.69	1
	(a)	Pd3Vac1H4_0-ex	0	+		- ++	-64.58	
\rightarrow	(Li	→ Vacancy) ↓ ↑					\rightarrow (0 sites	\rightarrow H4
↑	(1.)	Pd3H4_O+Li1_Vac	0 ++	+	+	+	-264.5	
I	(D)	Pd3H4_0+Li1_Vac-ex	0 ++	+		-	-264.6	
1		↓ ↑					\rightarrow (0 sites	\rightarrow H4
↑	(\mathbf{r})	Pd3+H4_T+Li1_Vac	0 ++	+		-	-260.2	
I	(C)	Pd3+H4_T+Li1_Vac-ex	~0 ++	+	+	+	-264.0	
1		↓ ↑					\rightarrow (T sites	\rightarrow H4
↑	(.1)	Pd3Vac1+H4_T+Li1_0	0 ++	-		-	-259.7	
I	(a)	Pd3Vac1+H4_T+Li1_O-ex	0	+	+	- ++	-261.0	
←	(Li	$\leftarrow \text{ O site}) \qquad \downarrow \uparrow$					\rightarrow (T sites	\rightarrow H4
	(\mathbf{r})	Pd3Vac1+H4_T	~0 ++	+		-	-61.78	
	(e)	Pd3Vac1+H4_T−ex	0 ++	+	+	+	-64.07	

Table 1 Flow of Phonon States when Impurity Li Atom Moves from Vacancy to O site

ng Energy: Difference of the lotal Energy between "Pd3Vac1+~" and "Pd3Vac1"

Since the phonon mode energy of these three Pd metal lattices with the impurity Li atom in the vacancy and O site ((b) ~ (d)) is all 0 eV at Γ point, these Pd metal lattices would be structurally stable for the phonon vibration at the normal temperature and the

normal pressure. The binding energy of these three structures is so close together that the binding energy fluctuation of impurity Li atom could cover the energy difference among these three states to transfer to each other. And since the phonon mode energy of these three Pd metal lattices becomes negative at M point and R point, these Pd metal lattices would be structurally unstable for the phonon vibration of these point directions. Furthermore, the phonon mode energy of the Pd metal lattice with the impurity Li atom on the O site becomes negative at X point (d). Therefore, the Pd metal lattice of this structure would be the most unstable for the phonon vibration of the three. Then the phonon state would possibly flow from the phonon state (b) to (d).

In one case, the phonon states could flow by way of the second phonon state (c). When some vacancies exist in Pd metal lattice, first of either all H atoms would be occluded on the O sites around the vacancies (a), the following an impurity Li atom would enter the vacancy. Or first an impurity Li atom would enter the vacancy; next H atoms would be occluded on the O sites around the vacancy (a). And then Pd3H4-O+Li1-Vac would be composed (b). After that four H atoms would go to and fro between four of six O sites of Pd3H4-O+Li1-Vac and four of eight T sites of Pd3+H4-T+Li1-Vac around the vacancy due to the vibration of Li atom (c). However in the other case, the phonon state could directly flow from the phonon state (b) to (d) without the second phonon state (c). In both cases, when the impurity Li atom happens to hop out from the vacancy, four H atoms would move from four of six O sites of Pd3H4-O+Li1-Vac (b) to four of eight T sites of Pd3Vac1+H4-T+Li1-O (d).

Fig. 4 shows the phonon dispersion curves of these three structures of the Pd metal lattices with the Li impurity atom entered the vacancy or the O site. These were calculated with the expanded interatomic distances in order for the total energy of the calculation unit cell to become the lowest while the H atoms were occluded or the impurity Li atom entered. The phonon modes in which mainly the impurity Li atom vibrates have narrow unchanged phonon mode energy at 0.04 ~ 0.05eV without H atoms (A), with four H atoms on the O sites (B) and with four regular tetrahedral H atoms on the T sites (C). Similarly, the phonon modes in which mainly H atoms vibrate have nearly unchanged phonon mode energy at about $0.12 \sim 0.15eV$ (B), (C) and (D). These phonon modes of unchanged energy correspond to those of the H-occluded Pd metal lattice without vacancy Fig. 1-(C) or with vacancy Fig. 2-(C), and they would indicate standing waves.







(A) No occluded H atom: Pd3+Li1-Vac

(Three Pd atoms with a Li atom in the Vacancy of the unit cell)

(B) Regular octahedral H atoms on the whole O sites: Pd3H4-O+Li1-Vac

(Three Pd atoms with four H atoms on the O sites and a Li atom in the Vacancy)

(C) Four regular tetrahedral H atoms on the T sites: Pd3+H4-T+Li1-Vac

(Three Pd atoms with four H atoms on the T sites and a Li atom in the Vacancy)

(D) Four regular tetrahedral H atoms on the T sites: Pd3Vac1+H4-T+Li1-O

(Three Pd atoms with four H atoms on the T sites and a Li atom on the O site)

After the phonon states transfer from the state (B) to the state (D) discussed above according to the Table 1, we expect that the phonon vibration will change to the coherent breathing mode vibration of four regular tetrahedral H atoms on the T sites. And further we expect that these four regular tetrahedral H atoms will collectively move to the vacancy or the center of tetrahedron of them when the impurity Li atom hops out from the center vacancy Fig. 2-(C) and Table 1-(e).

Although this time we carried out the simulation mainly with occluded H atoms, we also carried out the simulation with occluded D atoms for several cases of Table 1-(a) ~ (e). And we obtained mostly the phonon mode energy in which mainly D atoms vibrate was reduced about $1/\sqrt{2}$ of the phonon mode energy in which mainly H atoms vibrate. We could reason by analogy the similar results for occluded D atoms, taking account of mass difference between D atom and H atom. Therefore four regular tetrahedral D atoms could move collectively to the center of them to condense tightly without collision against the atomic nucleus in the Pd meal lattice because of the weak recoil of the hopping out of the light mass impurity Li atom. Then four D atoms would

cause the nuclear fusion of Tetrahedral Symmetric Condensation Theory.

3.3.2 Impurity Ca Atom Enters a Vacancy

In addition to the case of impurity Li atom mentioned above, it was shown that the Pd metal lattice could held the structural stability for the phonon vibration when the impurity Na, Mg or Ca atom entered a vacancy of Pd metal lattice with occlude H atoms on the whole O sites (Table 4A of Appendix). Since the phonon modes which mainly H atoms vibrate had nearly unchanged energy, those would be certainly standing waves. We expect that six surrounding H atoms will be drawn and recoiled strongly to collide against the atomic nucleus in the Pd meal lattice when the heavier impurity Ca atom hops out from the center vacancy. Then six H atoms would cause the nuclear transmutation of Octahedral Symmetric Condensation Theory.

3.3.3 Impurity H Atom Enters an O site

Furthermore we observed that the Pd metal lattice with four tetrahedral H atoms on T sites could be structurally stable for the phonon vibration when an impurity H atom entered an O site (Table 9A of Appendix). And only when the impurity H atom entered a T site, the Pd metal lattice could be structurally stable for the phonon vibration (Table 11 of Appendix). Then we expect that these H phonon states will be important in the small size nano-particles of FCC lattice metal which are said to take icosahedral structure composed of tetrahedrons without distinction of O sites and T sites, and only these H-occluded nano-particles will hold structural stability for the phonon vibration.

4. Summaries

We carried out the simulation of H phonon states in the Pd metal lattice of the smallest 1x1x1 unit calculating sell by using the first principles electronic state simulation program on personal computers. And we investigated the structural stability of H-occluded Pd metal lattice for the phonon vibration, examining the phonon mode energy.

We observed that the Pd metal lattice held the structural stability for the phonon vibration when an impurity Li atom entered a vacancy which placed at the center of four regular tetrahedral H atoms of T sites. These four H atoms and Li atom were seemed to vibrate as the standing wave phonon modes. When the Li impurity atom happened to hop out from the vacancy, four H atoms could move collectively to the vacancy which was the center of tetrahedron. Reasoning by analogy to apply the results to D atoms, we expect that four D atoms will move collectively to the center of tetrahedral D atoms to

condense tightly, and they will cause the nuclear fusion of Tetrahedral Symmetric Condensation Theory. And we observed the Pd metal lattice with occluded H atoms on the whole O sites held the structural stability when an impurity Ca atom entered the vacancy. And also the Pd metal lattice with occluded four regular tetrahedral H atoms on the T sites held structural stability when an impurity H atom entered O site. These phonon states seem to be somewhat important for nuclear reactions in the condensed matter.

The calculation on the 1x1x1 unit cell of this time was so vacancy-rich and tight that observed interesting phonon states would be possibly ghosts of unreasonable computer calculation. However on the other hand, these interesting phonon states could seem to really exist if the vacancy-rich domains were located in the small places like near surface or grain boundary of the metal lattice. Anyway, we will carry out the same kind of simulations on 2x2x2 unit cells, varying the concentration of vacancy to make sure the simulation results of this time. Then later we will analyze the interatomic force constants and the displacements of composed atoms for each phonon modes. And we will investigate the transition of H phonon states and also D phonon states and further the phonon interaction with electrons. Although the simulation of the phonon states on 2x2x2 unit cells would be impossible for the PC used this time, desirably a new computer of GPGPU (General-purpose computing on graphics processing units) parallel calculation could do it. Until then we will also simulate the same phonon states in Ni or Cu metal lattice with H and D atoms side by side.

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Appendix

Outline of Structural Stability for Phonon Vibration of Pd Metal Lattice

We calculated the phonon energy only at the special symmetric Γ , X, M and R points in order to obtain the outline of structural stability for the phonon vibration in short calculation time. Calculations were done on the perfect Pd metal lattice or the defective one with a vacancy, with occluded H atoms on O sites or T sites and with or without an impurity atom of Pd lattice atom, H atom and alkali/alkaline-earth metal atom such as Li, Na, K, Mg and Ca.

Since we could not optimize the lattice arrangement because of computational complexity and calculation time, we calculated with the two lattice constants of smaller (non-expanded) and larger (expanded) than those of experiments. For the expanded interatomic distances, calculations were done in order for the total energy of the calculation unit cells to become the lowest while H atoms were occluded or the impurity atom entered. We show the phonon states of non-expanded interatomic distances in the upper rows, describing the lattice structures of calculation unit cells, and we show those of expanded interatomic distances in the lower rows, describing the lattice structures of calculation unit cell added "-ex", both in the left column. Furthermore, we calculated basic Pd4 and Pd3Vac1 lattice structures of calculation unit cell also in order for the total energy to become the lowest similarly to the expanded interatomic distances. We show the phonon states of them in the lower rows, describing the lattice structures of calculation unit cell added "-opt" in the left column. And we show whether the phonon mode energy is zero (0), nearly zero (~0) or negative (-) at the Γ point and positive (+) or negative (-) at the X, M and R points in the middle column, and we also show where the minimum (--) or maximum (++) phonon mode energy appears. And furthermore, we show how much they spread (the effective value of nearly zero was written in ()) in the right column, adding the lattice constants used for the calculation.

The nearly zero (~0) eV phonon mode energy at the Γ point would be calculated exactly 0 eV in more accurate if the cut off energy is larger than about 1 keV which is used in this simulation. (Some phonon states which seem to have negative phonon mode energy all over the Γ , X, M and R points have not been calculated yet.)

	Г	х	м	R	min(l͡) (eV)	~	max (eV)	Lattice Const. (Å)
Pd4-opt	0	+	+	+ ++	0.000	~	0.030	3.862
Pd4H4_0	0	+	+	+ ++	0.000	~	0.124	3.862
Pd4H4_0-ex	~0	+	+	+ ++	(-0.000001)	~	0.091	4.060
Pd4+H2_T	~0++	+	+	+	(-0.000003)	~	0.191	3.862
Pd4+H2_T-ex	~0++	+	+	+	(-0.000003)	~	0.160	4.031
Pd4+H4_T	0 ++	+	+	+	0.000	~	0.198	3.862
Pd4+H4_⊤−ex	~0++	+	+	+	(-0.0000005)	~	0.146	4.160
Pd4+H8_⊤	~0	+ ++	+	+	(-0.000001)	~	0.232	3.862
Pd4+H8_T-ex	~0	+ ++	+	+	(-0.000001)	~	0.137	4.400

Table A1 Perfect Pd Metal Lattice

Table A2 Pd Metal Lattice with a Vacancy

	Г	х	м	R	min(l͡) (eV)	~	max (eV)	Lattice Const. (Å)
Pd3Vac1-opt	0	+		+ ++	(0.000) -0.004	~	0.037	3.714
Pd3Vac1H4_0	0	+		+ ++	(0.000) -0.006	~	0.152	3.714
Pd3Vac1H4_0-ex	~0	+		- ++	(-0.000003) -0.006	~	0.101	3.932
Pd3Vac1+H2_T		+ ++	-	-	-0.011	~	0.241	3.721
Pd3Vac1+H2_T-ex	~0	+	+ ++		(-0.000001) -0.006	~	0.198	3.906
Pd3Vac1+H4_T	~0++	+		-	(-0.0000004) -0.011	~	0.242	3.714
Pd3Vac1+H4_⊤−ex	0 ++	+	+	+	0.000	~	0.172	4.027
Pd3Vac1+H8_T	~0++	-		-	(-0.0009) -0.018	~	0.270	3.714
Pd3Vac1+H8_T-ex	0	+ ++	+	+	0.000	~	0.147	4.268

	Г	х	м	R	min(l͡) (eV)	~	max (eV)	Lattice Const. (Å)
Pd3+Pd1_Vac	0	+	+	+ ++	0.000	~	0.039	3.714
Pd3+Pd1_Vac-ex	0	+	+	+ ++	0.000	~	0.030	(Pd4–opt)
Pd3+H1_Vac	-	-		- ++	-0.122	~	0.038	3.714
Pd3+H1_Vac-ex	-	-		- ++	-0.122	~	0.038	3.708
Pd3+Li1_Vac	0	+ ++	+	+	0.000	~	0.051	3.714
Pd3+Li1_Vac-ex	~0	+ ++	+	+	(-0.000001)	~	0.046	3.791
Pd3+Na1_Vac	0	+	+	+ ++	0.000	~	0.059	3.714
Pd3+Na1_Vac-ex	~0	+	+ ++	+	(-0.000001)	~	0.047	3.895
Pd3+Mg1_Vac	0	+	+	+ ++	0.000	~	0.057	3.714
Pd3+Mg1_Vac-ex	0	+ ++	+	+	0.000	~	0.044	3.883
Pd3+K1_Vac	0	+	+	+ ++	0.000	~	0.045	3.714
Pd3+K1_Vac-ex	0	+	+	+ ++	0.000	~	0.031	4.058
Pd3+Ca1_Vac	0	+	+	+ ++	0.000	~	0.046	3.714
Pd3+Ca1_Vac-ex	~0	+	+	+ ++	(-0.000003)	~	0.031	4.013

Table A3 Pd Metal Lattice with an Impurity Atom Entered a Vacancy

Table A4 Pd Metal Lattice with Occluded H Atoms on the Whole O Sites and an Impurity Atom Entered a Vacancy

	Г	х	м	R	min(l͡) (eV)	~	max (eV)	Lattice Const. (Å)
Pd3H4_0+Pd1_Vac	0	+	+	+ ++	0.000	~	0.112	3.932
Pd3H4_0+Pd1_Vac-ex	~0	+	+	+ ++	(-0.000001)	~	0.091	(Pd4H4−ex)
Pd3H4_0+H1_Vac			-	- ++	-0.132	~	0.099	3.932
Pd3H4_0+H1_Vac-ex	-		-	- ++	-0.132	~	0.098	3.938
Pd3H4_0+Li1_Vac	0 ++	+	+	+	0.000	~	0.128	3.932
	0 ++	Т		_	(0.000)	~	0 1 2 5	2 004
	0 11	т		_	-0.025	~	0.125	5.554
Rd2H4 O+No1 Vee	0 ++	+		_	(0.000)	~	0 1 9 5	3 0 3 3
	0				-0.048		0.105	0.902
Pd3H4_0+Na1_Vac-ex	- ++	-		-	-0.076	~	0.168	4.082
Dd2H4 O+Mg1 Vee	~0++	+	_		(-0.000003)	~	0 1 7 9	3 0 3 3
Fu3H4_O+IVIg1_vac		Т			-0.065		0.176	3.932
Pd3H4_0+Mg1_Vac-ex	- ++	–	—		-0.084	~	0.157	4.089
Pd3H4_0+K1_Vac	- ++	–		-	-0.107	~	0.225	3.932
Pd3H4_0+K1_Vac-ex	- ++	-		-	-0.116	~	0.194	4.230
	0.44	_	_		(0.000)	•	0.226	2 0 2 2
Pu3H4_UTUAI_Vac	0 ++ -	_	-		-0.087	~	0.220	3.932
Pd3H4_0+Ca1_Vac-ex	- ++	-		-	-0.104	~	0.194	4.201

	Г	х	м	R	min(l͡) (eV)	~	max (eV)	Lattice Const. (Å)
Pd3+H4_T+Pd1_Vac	0 ++	+	+	+	0.000	~	0.228	3.714
Pd3+H4_T+Pd1_Vac-ex	~0++	+	+	+	(-0.0000005)	~	0.146	(Pd4+H4_T−ex)
Pd3+H4_T+H1_Vac	-	++	-	-	-0.169	~	0.229	3.714
Pd3+H4_T+H1_Vac-ex	-	++	-	-	-0.143	~	0.156	4.058
Pd3+H4_T+Li1_Vac	0 ++	+		-	(0.000) -0.008	~	0.238	3.714
Pd3+H4_T+Li1_Vac-ex	~0++	+	+	+	(-0.000003)	~	0.154	4.111
Pd3+H4_T+Na1_Vac	~0	+ ++	+	+	(-0.000001)	~	0.231	3.714
Pd3+H4_T+Na1_Vac-ex	0 ++	+	+	+	0.000	~	0.172	4.222
Pd3+H4_T+Mg1_Vac	~0		+	+ ++	(-0.000001) -0.002	~	0.238	3.714
Pd3+H4_T+Mg1_Vac-ex	0 ++	+	+	+	0.000	~	0.168	4.223
Pd3+H4_T+K1_Vac		- ++	-	-	-0.028	~	0.250	3.714
Pd3+H4_T+K1_Vac-ex	-	- ++	-		-0.034	~	0.187	4.375
Pd3+H4_T+Ca1_Vac		- ++	-	-	-0.028	~	0.263	3.714
Pd3+H4_T+Ca1_Vac-ex	++	-	-	-	-0.019	~	0.193	4.348

Table A5 Pd Metal Lattice with Occluded Four H Atoms on the T Sites and an Impurity Atom Entered a Vacancy

Table A6 Pd Metal Lattice with Occluded Eight H Atoms on the T Sites and an Impurity Atom Entered a Vacancy

	Г	х	м	R	min (~	max (eV)	Lattice Const. (Å)
Pd3+H8_T+Pd1_Vac	0	- ++	-	_	(0.000) -0.006	~	0.264	3.714
Pd3+H8_T+Pd1_Vac-ex	~0	+ ++	+	+	(-0.000001)	~	0.137	(Pd4+H8_T-ex)
Pd3+H8_T+H1_Vac	++	-	-	-	-0.091	~	0.268	3.714
Pd3+H8_T+H1_Vac-ex	-	- ++		-	-0.098	~	0.139	4.285
Pd3+H8_T+Li1_Vac		- ++	-	-	-0.012	~	0.263	3.714
Pd3+H8_T+Li1_Vac-ex	~0	++	+	+	(-0.0000006)	~	0.133	4.356
Pd3+H8_T+Na1_Vac	-	++	-	-	-0.011	~	0.255	3.714
Pd3+H8_T+Na1_Vac-ex	-	-	++	-	-0.044	~	0.161	4.449
Pd3+H8_T+Mg1_Vac	-	++	-	-	-0.013	~	0.259	3.714
Pd3+H8_T+Mg1_Vac-ex	- ++	-		-	-0.041	~	0.159	4.468
Pd3+H8_T+K1_Vac	-	++	-	-	-0.025	~	0.258	3.714
Pd3+H8_T+K1_Vac-ex	-	- ++		-	-0.076	~	0.173	4.564
Pd3+H8_T+Ca1_Vac	-	- ++	-		-0.026	~	0.269	3.714
Pd3+H8_T+Ca1_Vac-ex	-	++	-	-	-0.062	~	0.175	4.538

	Г	x	м	R	min (∏ (eV)	~	max (eV)	Lattice Const. (Å)
Pd4+Pd1_0	++	-	-	-	-0.044	~	0.115	3.862
Pd4+Pd1_0-ex	++	-	-	-	-0.021	~	0.047	4.665
Pd4+H1_0	0	+	+	+ ++	0.000	~	0.088	3.862
Pd4+H1_0-ex	~0	+	+	+ ++	(-0.0000004)	~	0.075	3.923
Pd4+Li1_0	0 ++	+	+	+	0.000	~	0.107	3.862
Pd4+Li1_0-ex	-	-		- ++	-0.009	~	0.086	4.113
Pd4+Na1_0	- ++		-	-	-0.027	~	0.108	3.862
Pd4+Na1_0-ex		- ++	-	-	-0.020	~	0.047	4.414
Pd4+Mg1_0	- ++		-	-	-0.025	~	0.112	3.862
Pd4+Mg1_0-ex						~		
Pd4+K1_0	-	-	++	-	-0.021	~	0.098	3.862
Pd4+K1_0-ex						~		
Pd4+Ca1_0		-	- ++	-	-0.036	~	0.096	3.862
Pd4+Ca1_0-ex						~		

Table A7 Pd Metal Lattice with an Impurity Atom Entered an O Site

Table A8 Pd Metal Lattice with Occluded H Atoms on the Whole O Sites and an Impurity Atom Entered an O Site

	Г	х	м	R	min(l͡) (eV)	~	max (eV)	Lattice Const. (Å)
Pd4H3_0+Pd1_0	++	-	-	-	-0.036	~	0.101	4.014
Pd4H3_0+Pd1_0-ex	–	-	++	-	-0.085	~	0.050	4.648
Pd4H3_0+H1_0	~0	+	+	++	(-0.000003)	~	0.098	4.014
Pd4H3_0+H1_0-ex	~0	+	+	++	(-0.000001)	~	0.091	(Pd4H4-ex)
Pd4H3_0+Li1_0	~0	+	+	++	(-0.000001)	~	0.098	4.014
Pd4H3_0+Li1_0-ex	++	-	-	-	-0.049	~	0.082	4.206
Pd4H3_0+Na1_0	- ++		-	-	-0.026	~	0.093	4.014
Pd4H3_0+Na1_0-ex		- ++	-	-	-0.071	~	0.072	4.445
Pd4H3_0+Mg1_0	-		-	- ++	-0.024	~	0.098	4.014
Pd4H3_0+Mg1_0-ex	-	- ++		-	-0.075	~	0.070	4.472
Pd4H3_0+K1_0	-	-		- ++	-0.073	~	0.102	4.014
Pd4H3_0+K1_0-ex	- ++	-		-	-0.085	~	0.047	4.632
Pd4H3_0+Ca1_0	-	-	-	++	-0.059	~	0.104	4.014
Pd4H3_0+Ca1_0-ex	- ++	-	-		-0.083	~	0.047	4.624

	Г	x	м	R	min (~	max (eV)	Lattice Const. (Å)
Pd4+H4_T+Pd1_0	-		-	- ++	-0.040	~	0.266	4.060
Pd4+H4_T+Pd1_0-ex	-	-	-	++	-0.036	~	0.063	4.759
Pd4+H4_T+H1_O	~0	- ++	+	+	(-0.00003) -0.009	~	0.206	3.862
Pd4+H4_T+H1_0-ex	-	++	-	-	-0.074	~	0.144	4.210
Pd4+H4_T+Li1_0	~0++	-	-		(-0.0009) -0.010	~	0.217	3.862
Pd4+H4_T+Li1_0-ex	0 ++	+	+	+	0.000	~	0.133	4.350
Pd4+H4_T+Na1_0	- ++		-	-	-0.020	~	0.267	3.862
Pd4+H4_T+Na1_0-ex	++	-	-	-	-0.015	~	0.142	4.574
Pd4+H4_T+Mg1_0	-		- ++	-	-0.023	~	0.264	3.862
Pd4+H4_T+Mg1_O-ex						~		
Pd4+H4_T+K1_0	-		- ++	-	-0.034	~	0.281	3.862
Pd4+H4_T+K1_0-ex						~		
Pd4+H4_T+Ca1_0	-		- ++	-	-0.033	~	0.295	3.862
Pd4+H4_T+Ca1_0-ex						~		

Table A9 Pd Metal Lattice with Occluded Four H Atoms on the T Sites and an Impurity Atom Entered an O Site

Table A10 Pd Metal Lattice with Occluded Eight H Atoms on the T Sites and an Impurity Atom Entered an O Site

	Г	х	м	R	min(Ϧ (eV)	~	max (eV)	Lattice Const. (Å)
Pd4+H8_T+Pd1_0						~		
Pd4+H8_T+Pd1_0-ex						~		
Pd4+H8_T+H1_0	-	- ++	-		-0.131	~	0.236	3.862
Pd4+H8_T+H1_O-ex	-	- ++		-	-0.136	~	0.126	4.466
Pd4+H8_T+Li1_0	-	++	-	-	-0.016	~	0.238	3.862
Pd4+H8_T+Li1_0-ex	-	- ++		-	-0.028	~	0.118	4.567
Pd4+H8_T+Na1_0						~		3.862
Pd4+H8_T+Na1_0-ex						~		
Pd4+H8_T+Mg1_0						~		
Pd4+H8_T+Mg1_0-ex						~		
Pd4+H8_T+K1_0						~		
Pd4+H8_T+K1_O-ex						~		
Pd4+H8_T+Ca1_0						~		
Pd4+H8_T+Ca1_0-ex						~		

	Г	X	М	R	min(Ŋ (eV)	~	max (eV)	Lattice Const. (Å)
Pd4+Pd1_T	-	++	-	-	-0.061	~	0.144	3.862
Pd4+Pd1_T-ex						~		
Pd4+H1_T	0	+ ++	+	+	0.000	~	0.171	3.862
Pd4+H1_T-ex	0	+ ++	+	+	0.000	~	0.154	3.953
Pd4+Li1_T	-	++	-	-	-0.027	~	0.124	3.862
Pd4+Li1_T-ex	-	++	-	-	-0.021	~	0.094	4.224
Pd4+Na1_T	—	++	-	-	-0.043	~	0.134	3.862
Pd4+Na1_T-ex						~		
Pd4+Mg1_T	—	++	-	-	-0.043	~	0.143	3.862
Pd4+Mg1_T-ex						~		
Pd4+K1_T	-	- ++		-	-0.043	~	0.139	3.862
Pd4+K1_T-ex						~		
Pd4+Ca1_T	-	- ++	-		-0.045	~	0.137	3.862
Pd4+Ca1_T−ex						~		

Table A11 Pd Metal Lattice with an Impurity Atom Entered a T Site

Table A12 Pd Metal Lattice with Occluded H Atoms on the Whole O Sites and an Impurity Atom Entered a T Site

	Г	х	м	R	min(Ŋ (eV)	~	max (eV)	Lattice Const. (Å)
Pd4H4_0+Pd1_T						~		4.060
Pd4H4_0+Pd1_T-ex						~		
Pd4H4_0+H1_T	-	-		- ++	-0.033	~	0.148	4.060
Pd4H4_0+H1_T-ex	-	-		- ++	-0.048	~	0.135	4.136
Pd4H4_0+Li1_T	-	—	++	-	-0.031	~	0.135	4.060
Pd4H4_0+Li1_T-ex	-	-	++	-	-0.070	~	0.106	4.138
Pd4H4_0+Na1_T	-	-		- ++	-0.068	~	0.182	4.060
Pd4H4_0+Na1_T-ex						~		
Pd4H4_0+Mg1_T	-	-	-	++	-0.070	~	0.177	4.060
Pd4H4_0+Mg1_T-ex						~		
Pd4H4_0+K1_T	-	-	++	-	-0.129	~	0.204	4.060
Pd4H4_0+K1_T-ex						~		
Pd4H4_0+Ca1_T	—	-	- ++		-0.101	~	0.226	4.060
Pd4H4_0+Ca1_T-ex						~		

Table A13 Pd Metal Lattice with Occluded H Atoms on the O Sites or T Sites and an Impurity Li Atom Entered an O Site

	Г	х	м	R	min ([)	~	max	Lattice
					(ev)		(ev)	Const. (A)
Pd3Vac1H3_O+H1_T+Li1_O	0 ++	+	+	+	0.000	~	0.128	3.932
Pd3Vac1H3_O+H1_T+Li1_O-ex						~		
Pd3Vac1+H4_T+Li1_0	0 ++	-	-	-	(0.000)	~	0.171	4.027
					-0.012			
Pd3Vac1+H4_T+Li1_0-ex	0	+	+	- ++	(0.000)	• •	0.125	4.310
					-0.003	~		

Is Gamma-Less Transmutation Possible? -The Case of Metal plus TSC and BOLEP-

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Abstract: Very speculative modeling and discussions are made for considering a nuclear physics possibility of Iwamura-type (A, Z)-selective transmutation without hard gamma-rays. The BOLEP (burst-of-low-energy-photons) energy damping mechanism from very highly excited intermediate nucleus by the 4D/TSC-min capture to host metal nucleus is basic idea, after the analogy of ⁸Be* nucleon halo model for the BOLEP energy damping to ⁸Be ground state. The very highly excited intermediate nucleus ¹⁴¹Pr*(Ex = 50.49 MeV) by 4D + ¹³³Cs capture reaction may have a two-alpha-halo rotation-vibration deformed state, which would make avalanche energy damping via BOLEP.

Keywords: Gamma-less transmutation, two-alpha halo model, BOLEP, Iwamura transmutation

1. Introduction

The extraordinary claim of discovery of selective transmutation with mass-charge (A, Z) sifting was reported by Iwamura et al [1], by deuterium-gas permeation through Pd/CaO multi-layer film on which surface ¹³³Cs was deposited. The claimed nuclear transmutation is ¹³³Cs to ¹⁴¹Pr, namely (A+8, Z+4) selective sifting, as typical data are shown in Fig.1. Several replication experiments were done to confirm the phenomenon positively [2, 3]. Especially, the latest study [3] by Hioki et al reported convincing data of ¹⁴¹Pr production with anomalously large (relatively in order of magnitude) amount. So the claim of observation looks real.

However, if the observation was by real nuclear transmutation reactions, there remain unresolved puzzles and questions: 1) Why (A, Z) selective sifting happens? 2) Why it happened only for surface-deposited Cs, not for other elements as Pd, Ca and O? 3) What is the role of CaO layer, supposing Pd might has some effect on unknown condensed matter nuclear reactions? 4) Why no intense hard gamma-rays were observed, if nuclear transmutation by capturing 4D or 2-alphas capture to ¹³³Cs? And some other questions

arise. The author focuses on the question 4) in this work to try to find some rational path to damp very highly excited nuclear energy of medium-heavy nucleus to its ground state without emitting hard (high energy) gamma-rays.



Fig.1: Selective transmutation data claimed by Iwamura et al [1]

Is Iwamura 'Transmutation' a real nuclear reaction? It is mystery with unknown theoretical mechanism. A model of selective transmutation of 133 Cs by A-8 & Z-4 increase by 4D/TSC capture was proposed [4] to explain possible nuclear mechanism of the phenomenon. This model answers why the (A, Z)-selective transmutation can happen. However, it could not resolve the 'head-ache issue': why hard gamma-rays were not emitted. In the conventional sense of nuclear physics, such transmutation should damp its 141 Pr* nuclear excitation energy of intermediate compound nucleus by mass defect (50.49 MeV) by some electro-magnetic transition. But how does it go without emitting high energy gamma-rays? Is the BOLEP (burst of low energy photons) from nucleon-halo state [5] as proposed for ⁸Be* after 4D/TDC fusion applicable to the Iwamura transmutation?

M(metal-nucleus) + 4D/TSC fusion may take place with much higher rate than M + 4H/TSC reaction, because of 'very long' life time (a few ms or more) of ⁸Be* cf. a few fs life of 4H/TSC-minimum: ratio is $10^{-3}/10^{-15} = 10^{12}$: and <W> value is much larger too. (This may explain why 'transmutation by H-gas was not observed by Iwamura.)

Damping very high-energy (ca. 50 MeV) excitation of compound nucleus of metal plus 4D/TSC-min (⁸Be*) by the BOLEP mechanism might be a possibility. Very high spin rotation/vibration mode of 2-alpha-halo state of intermediate excited nucleus is a

candidate model. However, the model is very speculative and is yet to study more detailed possible transition channels

2. Brief Summary of 4D/TSC + Metal Interaction

Let us remember the original model [4] briefly. According to the results of QM-Langevin code calculations for cluster condensation [6, 7, 8], 4D/TSC may get into the collapse state with so small size as 20 fm keeping charge-neutrality in several fs after the start of condensation. Such a very small charge-neutral entity may pass through the multi-layer shell of electron cloud of target host metal atom to approach at the core nucleus of metal atom, behaving something like a 'neutron'. An image is shown in Fig.2 for the charge-neutral small entity of 4D/TSC-minimum penetrating through the multi-shell electron cloud of target metal atom which will have the inner-most K-shell electron orbit about 1 pm radius which is much larger than the size ca. 20 fm of 4D/TSC-minimum.



Fig.2: Image of penetration of charge-neutral 4D/TSC-minimum through the electron cloud of target metal atom to approach the core nucleus

Considering the elongated life time of intermediate compound nucleus ⁸Be* by 4D/TSC fusion [5], with status shown in Fig.3 for surrounding left 4 electrons of TSC, supposed 4D/TSC plus metal-nucleus fusion rate may become much larger for 4D/TSC than the 4H/TSC [9] plus metal-nucleus interaction rate. This might explain why Iwamura

observed transmutation dominantly for deuterium gas and non-visibly for protium gas (see Fig.1). In Fig.4, image of fusion strong interaction of 4D/TSC and Cs is shown.



Fig.3: Image of 4D/TSC condensation-collapse and ⁸Be* state with bosonized electrons



Fig.4: Image of strong interaction of ${}^{8}\text{Be*}$ and metal-nucleus, many PEF strings between p-n pion exchanges to induce ${}^{8}\text{Be*}$ + heavier-nucleus fusion event

The fusion rate was estimated by the sudden tall thin barrier approximation (STTBA) [4], as the view graph is copied in Fig.5. Rate estimation is shown in Fig.6.



Fig.5: TSC + metal-nucleus fusion rate is approximately estimated by STTBA



Fig.6: Estimated 4D/TSC + ¹³³Cs fusion rate by STTBA, numerical values are changed after discussion with Iwamura after [4]





Fig.7: Considerable fusion reactions between 4D/TSC-min and ¹³³Cs nucleus

Probable fusion reactions between 4D/TSC-minimum and ¹³³Cs nucleus is shown in Fig.7. Considering the supposed longer life time of ⁸Be* nucleon halo state and its strong PEF coupling, 133 Cs + 4d fusion would be major reaction.

3. Two-alpha Halo Model and BOLEP

By the conventional nuclear physics view, excited states of medium-heavy (A = 50 - 200) nuclei have major EM (electro-magnetic) transition decays emitting hard gammarays. The known energy level scheme of ¹⁴¹Pr is shown in Fig.8, with insertion of very highly excited state by the 4d + ¹³³Cs fusion (capture) reaction. Excitation energy levels Ex < 1657.6 keV are known (evaluated). From excited states, EM transitions (E1, E2, E3, M1, M2, M3) happen in cascade scheme to emit known discrete energy gamma-rays in 0.14 to 1.7 MeV range. Upper energy levels more than 1.7 MeV are not given.

We see the 50.49 MeV level is too far away above that 1.7 MeV known upper-most level. Is the similar EM transitions from that high level a rational nuclear physics mechanism? It might be or might not be. We have no concrete knowledge. The cluster nuclei capture like 4d + Cs generates (by mass defect of strong force rearrangement of intermediate compound nucleus) very highly excited states as Ex = ca. 50 MeV, for other medium heavy nuclei also. This is the new aspect that TSC-induced nuclear reactions may

introduce into the unknown field of nuclear physics, as is the case of ${}^{8}\text{Be}*(\text{Ex} = 47.6 \text{ MeV})$ highly excited low mass nucleus halo state. In general, the main stream nuclear physics study would not have been extended to such very highly excited cases.



Fig.8: Energy-levels and gamma-decay scheme of ¹⁴¹Pr*, after Table of Isotopes 8th edition

So far, we may have a chance to see some new states of energy transition there. When nucleus is highly excited, deformation of nucleus shape becomes larger, deviating more and more from the ground state near spherical shape. The deformation is mathematically drawn by using spherical harmonics pictures (see the case of ⁸Be* [5]). In our special case of initial state interaction between 4D/TSC (⁸Be*) and medium-heavy nucleus as ¹³³ Cs, we may consider the initial condition of ⁸Be* existence may cause the state of deformation of intermediate excited nucleus as ¹⁴¹Pr*, as we imagine by Fig.9. We may imagine rotation of ⁸Be-cluster around core nucleus, combined with vibrational motion between the two. Such rotation-vibration combined deformation will cause the so called band-state of many narrowly gapped discrete energy levels. We may guess the deformation state of Fig.9 to be much lower energy state than the 50.49 MeV state, considering the number of effectively coupling PEF values (8-10) is not large enough to sustain the very highly excited state of 50.49 MeV. Such speculation is possible from analysis of ⁸Be* excited states and possible inner nucleons-clustering (alpha, h and t clusters) as a function of PEF values [5]. So, we need to consider more deformed states.



Fig.9: Image of the initial state strong interaction between ⁸Be* and medium-heavy nucleus



Fig.10: Two-alpha halo state of ¹⁴¹Pr* at highly excited deformation

As shown in Fig.10, two-alpha halo state may be sustaining high excitation energy by the rotation energy of two alpha halos, the rotation-vibration energy of elliptic core and the coupled vibration energy of alphas and core. There may appear so many boson-coupled

discrete energy states, which may make avalanche energy damping by BOLEP (burst-of-low-energy-photons). Probably, more rational image is the dumbbell type deformation of core-cluster as shown in Fig.11. The author considers it most rational configuration to sustain that very high excitation energy of 50.49 MeV for ¹⁴¹Pr*.



Fig.11: Two-alpha halo state with dumbbell deformation of core-cluster

We need rational information of the order of life time for such highly excited/deformed states. Considering very high spin-coupled states of rotation-vibration energy-band with very small gap energies (1-10 keV speculated) as nuclear phonons (bosonic coupling), life time may be much larger than 1 fs which is typical gamma-transition of low level excited states (maybe less than Ex = 10 MeV, comparable to nucleon binding energy). We do not know whether such very highly excited isomer state with 1 ps or longer life time can exist or not. However, we can say it might be possible. Thus we may expect BOLEP energy damping from ¹⁴¹Pr* to ¹⁴¹Pr ground state without emitting hard gamma-rays than BOLEP (soft X-ray range or matter-converted visible light). In competition to BOLEP, we will have to consider fission channel of dumbbell deformation [10] which was analyzed to produce clean (non-radio-active) stable isotopes mostly as ash.

There will be minor decay routes to be trapped intermediate lower excited energy states of ¹⁴¹Pr along with BOLEP. So, the detail decay scheme of ¹⁴¹Pr*(Ex = 50.49 MeV) is complex as drawn in Fig.12.

No concluding remarks should be said because the study is in very speculative stage.

Since the proposed issue of very highly excited states of nuclei from light to heavy mass region is yet to study mechanism of nuclear physics, we will need a long series of experimental and theoretical works to find full consistency between observed results and model predictions. We can say that the consistency study about the proposed nucleon-halo model [5] for the final state interaction of ⁸Be*(Ex=47.6 MeV) born by the 4D/TSC process is the starting stage for the general direction of final state mechanism on such very highly excited nuclei.



Fig.14: Presently modeled energy damping scheme of $^{133}Cs + 4d$ transmutation

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