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NUCLEAR PRODUCTS AND THEIR TIME DEPENDENCE INDUCED BY CONTINUOUS DIFFUSION OF DEUTERIUM THROUGH MULTI-LAYER PALLADIUM CONTAINING LOW WORK FUNCTION MATERIAL

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ABSTRACT

Two kinds of experimental methods have been designed to induce nuclear reactions in the Pd-D system. One is the D₂ gas diffusion method, and the other is the electrolysis diffusion method. A common feature of the methods is to cause continuous diffusion of deuterium through a multi-layer Pd that contains low work function material (CaO, TiC, YzOs, etc.). Time dependence of nuclear products (Mg, Si, S, F, Al) were observed by the D₂ gas diffusion method, in which the products were analyzed WITHOUT taking the multi-layer Pd out of the apparatus. The time dependence of the products was reproduced qualitatively. The ³³S/³²S ratio of the products was one order larger than that of natural abundance. Fe isotope ratio anomaly of the multi-layer Pd obtained by the electrolysis diffusion method was confirmed by SIMS and TOF-SIMS. Si powder products detected after electrolysis amounted to 0.057g, and its isotopic composition was anomalous.

1. INTRODUCTION

Nuclear reactions observed in the low energy deuteron and metal system have been investigated intensively. However, the nature of the phenomena is still unclear. Our experimental results so far lead us to assume that necessary conditions to induce nuclear reactions in solids are as follows; (i) existence of a low work function material near the Pd surface, (ii) enough diffusion flux of deuterium, (iii) high D/Pd on the Pd surface.

To meet with the assumptions, we have contrived two kinds of experimental methods characterized by “multi-layer Pd” and “continuous diffusion of D” as shown in Fig. 1. The multi-layer Pd consists of Pd thin film, low work function material and Pd bulk. It corresponds to the assumption (i). Enough diffusion flux of deuterium, assumption (ii), is satisfied with the composition shown in the lower figure. Deuterium atoms are provided with D electrolyte or D₂ gas and released from the vacuum side. They are controllable by the applied electric current or D₂ gas pressure. As for the assumption (iii), enough D/Pd ratio on the Pd thin film is considered to be attained by our methods.

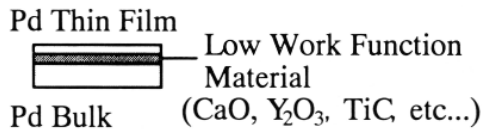
2. Experimental

Figure 2 show the cross sectional view of the D_2 gas diffusion apparatus. The feature of this method is that it can analyze the surface of a Pd sample by XPS (X-ray Photoelectron Spectroscopy) WITHOUT taking it out of the apparatus. Therefore it is possible to avoid contamination onto the Pd sample from outer environment. This newly developed apparatus can provide the data of time dependence of detected nuclear products.

The apparatus consists of two vacuum chambers, an X-ray gun and an electrostatic analyzer for XPS, a mass spectrometer and a Ge semiconductor detector. One chamber is filled with D_2 gas, and the other chamber is evacuated by a turbo molecular pump. These two chambers are divided by a multi-layer Pd composed of Pd thin film (400 angstrom), low work function layer (typically CaO; 1000 angstrom) and Pd sheet (25mm \times 25mm \times 0.1mm).

The experimental procedure is as follows. First, the surface of a multi-layer Pd in the vacuum chamber is analyzed by XPS to confirm that the surface of the Pd sample is clean. Next, D_2 gas is allowed into a chamber and deuterium atoms diffuse from the D_2 side chamber to the vacuum side chamber. At this moment a nuclear reaction occurs on the multi-layer Pd containing low work function material. After certain period (from 2 days to 1 week) of deuterium diffusion through the Pd sample, the D_2 side chamber is evacuated and the surface of the Pd sample is analyzed by XPS in the chamber. New elements, which did not exist on the Pd sample at the beginning of the experiment, can be detected. To obtain a time dependence analysis of the products, the process is repeated 3 to 4 times.

(1) Multi-layer Pd



(2) Continuous Diffuion of

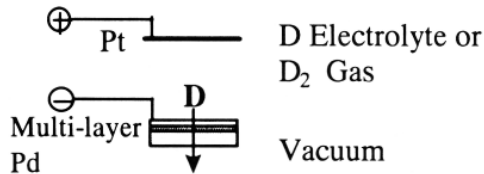


Fig. 1. Features of the Present Method

Experimental results using the electrolysis type of apparatus were presented at ICCF-7[1]. Schematic view of the electrolysis diffusion method is shown in Fig. 3. Details of the apparatus are given in references [1] and [2]. The feature of this apparatus is that much larger reaction rate is possible because D/Pd on the Pd surface is larger than that of D_2 gas diffusion method. We can estimate excess heat and radiation with this type of apparatus, although element and mass analysis of the Pd sample become possible after the end of an experiment.

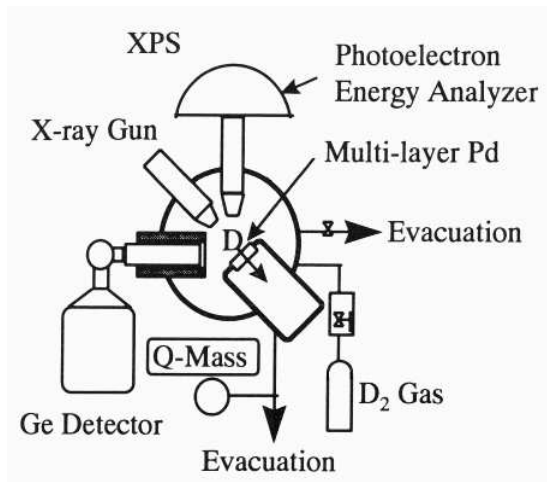


Fig. 2. D₂ Gas Diffusion Method

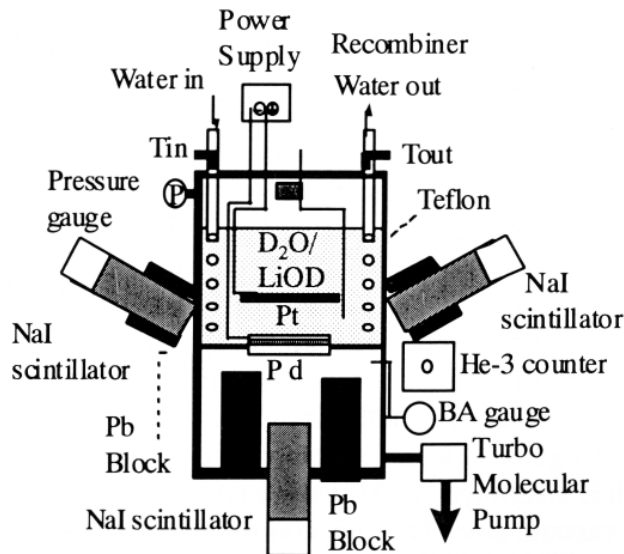


Fig. 3. Electrolysis Diffusion Method

3. RESULTS AND DISCUSSION

Experimental results by the D₂ diffusion method are shown in Fig. 4-6. We tried three kinds of materials; normal Pd, multi-layer Pd (Pd/CaO/Pd) and Li doped multi-layer Pd (Pd,Li/CaO/Pd).

Figure 4 shows the time dependence of C on the normal Pd (Pd only) sample which existed as an impurity at the beginning of the experiment. C usually exist on the surface of a Pd sample unless we remove it. Therefore, C and Pd are detected by XPS on the normal Pd sample. The numbers of C atoms detected by XPS did not change for two cases (No. 1, 2), although the initial C and experiment time were different each other. No time dependence of C was observed if Pd-only samples were used.

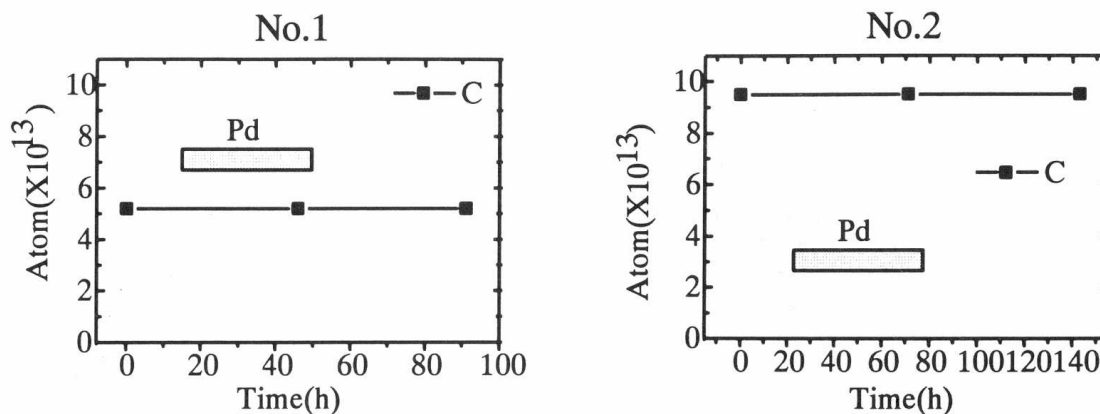


Fig. 4. Time Dependence of C (impurity) detected on the Pd only samples

Multi-layer Pd gave us entirely different results. Time dependence of C, Mg, S, Si on the multi-layer Pd (Pd/CaO/Pd) is shown in Fig. 5. First, we will explain the result of experiment No. 3. There were no elements except C and Pd at the beginning of the experiment. No Mg, Si and S existed on the sample. Mg, Si and S peaks emerged and the C peak decreased after 42 hours of deuterium diffusion through the multi-layer Pd. At 116 hours, S and Si increased and Mg decreased. As for No. 4 experiment, amount of C was slightly larger than that of No. 3. Mg, Si and S were detected again after 24 hours. After that, Mg decreased, Si and S increased, and C decreased monotonically as shown in the figure on the right. These results indicated that the behavior of C, Mg, S, Si were reproduced qualitatively.

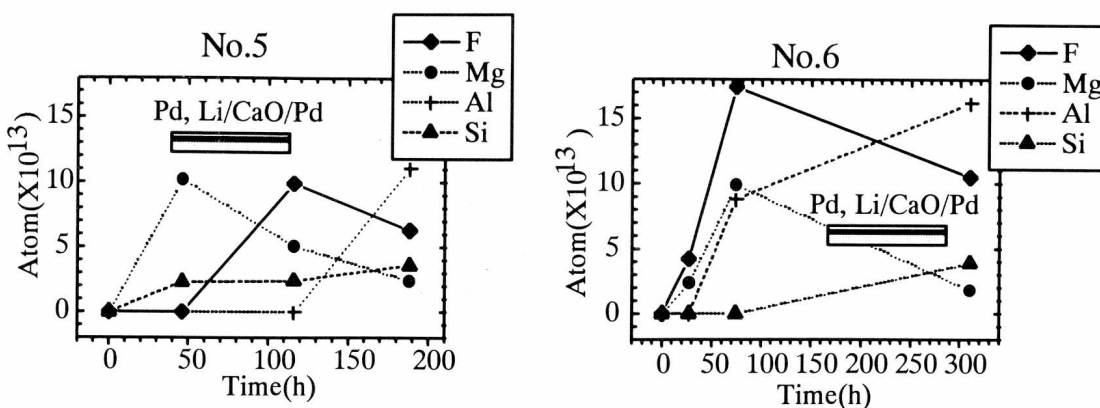


Fig. 5. Time Dependence of C, Mg, Si, S detected on the Pd/CaO/Pd samples

Examination on isotope ratio of the detected elements was carried out. Table 1 shows comparison between multi-layer Pd and normal Pd on isotopic abundance of S. Samples No. 1 and No. 4 were analyzed by SIMS (Secondary Ion Mass Spectroscopy) after the experiments. S on the No. 1 sample was detected by SIMS, although no S was detected by XPS. Since the sensitivity for S of SIMS is higher than that of XPS, S on the No. 4 sample was observed by both XPS and SIMS. As for ^{36}S , no effective counts were obtained because abundance of ^{36}S is very small.

Although secondary ion intensity of the No. 4 was larger than that of the No. 3, $^{34}\text{S}/^{32}\text{S}$ was almost equal to each other. $^{34}\text{S}/^{32}\text{S}$ is nearly equal to the natural abundance. On the other hand, $^{33}\text{S}/^{32}\text{S}$ of the Pd/CaO/Pd (No. 4) was one order larger than that of Pd only (No. 1). $^{33}\text{S}/^{32}\text{S}$ of the No. 1 was nearly equal to the natural abundance. These results shown in Table 1 indicate that isotopic abundance of S on the multi-layer Pd (No. 4) was anomalous and S on the normal Pd seemed natural.

Table 1. Comparison between Multi-layer Pd and Normal Pd on Isotopic Abundance of S

Type of Sample	Secondary Ion Intensity (cps)			Isotope Ratio	
	^{32}S	^{33}S	^{34}S	$^{33}\text{S}/^{32}\text{S}$	$^{34}\text{S}/^{32}\text{S}$
Pd/CaO/Pd (No. 4)	1.93×10^4	4.89×10^3	1.05×10^3	0.25	0.054
Pd Only (No. 1)	9.70×10^2	1.0×10	5.0×10	0.010	0.052
Natural Abundance	-	-	-	0.0079	0.044

Let us discuss on the above experimental results on the multi-layer Pd. The first point is the time dependence of C, Mg, Si, S. These elements were detected by XPS without taking the multi-layer Pd out of the vacuum chamber. The surface of the sample was just only exposed with D_2 gas. Therefore it was difficult to add these elements on the surface of the multi-layer Pd, or remove them from it. Especially Mg once increased and decreased. It is very difficult to explain their behaviors by certain contamination processes. The second point is that product S had anomalous isotopic abundance. As shown in Table 1, $^{33}\text{S}/^{32}\text{S}$ is one order larger than natural isotope ratio. If the S were contamination, was such efficient isotope separation possible? According to the points, we can conclude that it is strongly suggested that Mg, Si, S are formed by certain nuclear reactions.

If these elements are nuclear products, they are basically explained by the EINR (Electron-Induced Nuclear Reaction) model[2]. Experimental results enable us to make an interpretation that C was transmuted to Mg, Si, S. The EINR model gives the following explanation. At first deuterium nuclei capture electrons and form di-neutron clusters. Simultaneously, the di-neutron clusters react with C, and produce Mg. After that, Mg reacts with di-neutron clusters again and is transmuted into Si or S.



In the Pd/CaO/Pd experiments, impurity C was transmuted into Mg, Si, S. The next experiments were performed aiming to transmute Li into other elements.

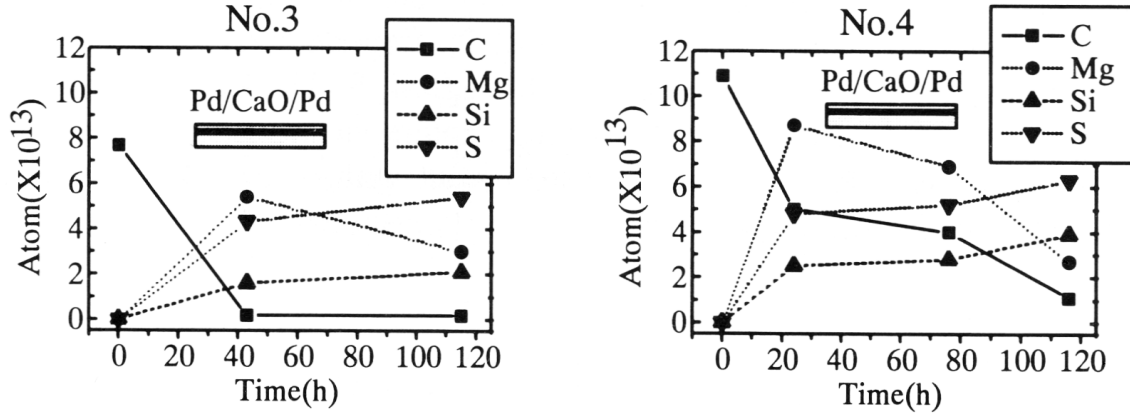


Fig. 6. Time Dependence of F, Mg, Al, Si detected on the Pd, Li/CaO/Pd samples

Experimental results on Li doped multi-layer Pd are shown in Fig. 6. Lithium atoms were doped by the electrolysis of LiOD solution onto the surface of the multi-layer Pd (Pd/CaO/Pd). The new elements F, Al emerged in the both cases, No. 5 and No. 6. F initially increased and then decreased. Al increased monotonically. The behaviors of F and Al were similar to Mg and Si, respectively, though the emergence time of F and Al in the case of No. 5 is different. Mg and Si are considered to have originated from impurity C. We observed F and Al by adding Li on the surface of multi-layer Pd, therefore we assume that F and Al were transmuted from the added Li as shown in the following equation.



At present, the model is theoretically incomplete. However, it is noticeable that the similar explanation is possible between the transmmutations of C and Li.

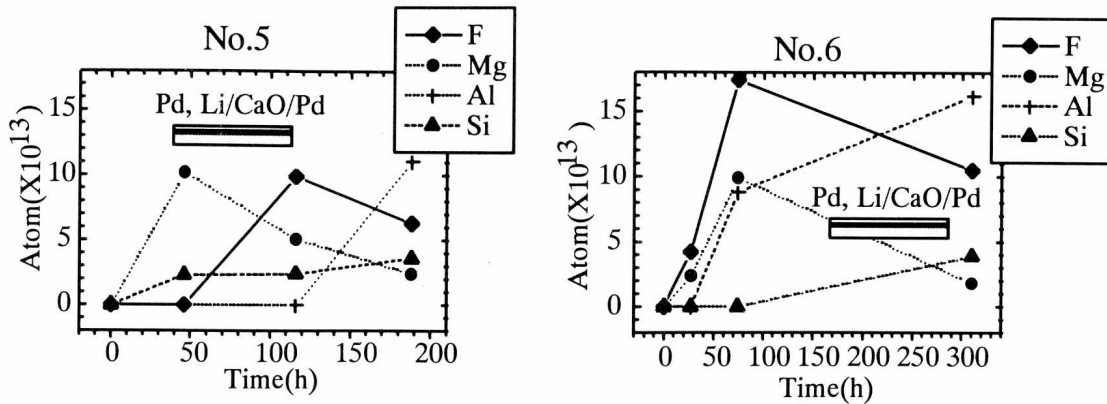


Fig. 7. Fe Isotope Anomaly observed by TOF-SIMS

Next, experimental results obtained by the electrolysis diffusion method are described. Excess heat and nuclear products were observed for almost all the cases we tried using the multi-layer Pd. Isotopic composition of the obtained product were often different from natural abundance. The authors usually estimate the isotopic composition of a

product by SIMS. Figure 7 presents an example in which the Fe isotope ratio anomaly was confirmed by both SIMS and TOF-SIMS (Time of flight SIMS). Better mass resolution can be obtained by TOF-SIMS. According to the SIMS analysis for sample (EV75) in Fig. 7, $^{57}\text{Fe}/^{56}\text{Fe}$ was estimated at 1.8; very high value compared with the natural abundance of 0.0234. TOF-SIMS analysis gave 0.301 for $^{57}\text{Fe}/^{56}\text{Fe}$ as shown in Fig. 7. Anomalous large $^{57}\text{Fe}/^{56}\text{Fe}$ was obtained on a multi-layer sample (EV75) by both SIMS and TOF-SIMS, though $^{57}\text{Fe}/^{56}\text{Fe}$ did not agree with each other. The reason is considered that $^{57}\text{Fe}/^{56}\text{Fe}$ depend on the analyzed position on the sample. Variation of isotope ratio depending on the location of analysis are often observed[1].

As excess heat increased, the amount of nuclear products increased. Figure 8 shows an example of Si powder product. In this case, large excess heat more than input power was obtained. The powder is SiO₂, because Si was oxidized by O₂ in the air after being taken out of the experimental apparatus. The amounts of the Si reach 0.057g.

In order to evaluate Si contamination, we made a list of candidates of contamination source; solution, multi-layer Pd, Pt anode, Ni cooling pipe, Polypropylene and Teflon in the experimental apparatus. The maximum quantity of Si contaminants was estimated at 0.023g, which is smaller than that of the obtained Si powder.

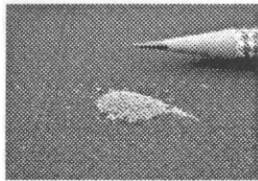


Fig. 8. Detected Si Powder

Table 2. Isotope Ratio of Detected Si Powder

Isotope	Detected Si Powder		Si Standard Solution		Natural Abundance (%)
	Intensity (cps)	Isotope Ratio (%)	Intensity (cps)	Isotope Ratio (%)	
^{28}Si	4,300	96.1	3,256	92.5	92.23
^{29}Si	70	1.6	149	4.3	4.67
^{30}Si	105	2.3	110	3.2	3.10

The isotope ratios of detected Si powder are shown in Table 2. ICP-MS (Inductively Coupled Plasma Mass Spectrometry) was applied to the analysis, the tabulated values were averaged by 5 times measurements. Si of detected Si powder is smaller than Si standard solution as shown in the table. The isotopic composition of Si powder detected in the apparatus after an experiment was different from natural Si abundance.

Judging from the above results, the authors consider that the detected Si powder is composed of nuclear products and Si impurities in the solution. We should confirm this result by performing the experiments with an improved solution that contains smaller amounts of Si.

REFERENCES

- [1] Y. Iwamura, T. Itoh, N. Gotoh, M. Sakano, I. Toyoda and H. Sakata, "Detection of Anomalous Elements, X-ray and Excess Heat Induced by Continues Diffusion of Deuterium Through Multi-Layer Cathode (Pd/CaO/Pd)", Proc. of ICCF-7, Vancouver, Canada, April 19-24, 1997, p. 167
- [2] Y. Iwamura, N. Gotoh, T. Itoh and I. Toyoda, "Detection of Anomalous Elements, X-ray and Excess Heat in a D₂-Pd System and its Interpretation by the Electron-Induced Nuclear Reaction Model", *Fusion Technology*, vol. 33, No. 4, p. 476, 1998