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# **Replication Of MHI Transmutation Experiment By D<sub>2</sub> Gas Permeation Through Pd Complex**

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Unusual nuclear transmutation reactions have been reported by Mitsubishi Heavy Industries (MHI). In their experiment,  $D_2$  gas permeates through a Pd complexes, which consists of a thin Pd layer, alternating CaO and Pd layers and bulk Pd.<sup>1</sup> When they used sample Pd complexes with additional Cs on the surface, Pr emerged on the surface while Cs decreased after the sample was subjected to  $D_2$  gas permeation at 343 K and 1 atm for about one week. The elemental analysis was performed by X-ray photoelectron spectroscopy (XPS). This phenomenon was reproduced qualitatively in the present replication experiment.

We performed D-permeation experiments similar to the MHI's experiment<sup>1</sup> three times, and we confirmed the production of Pr. Pd complex samples were provided to us by MHI. The surface was electrolytically cleaned to remove hydrocarbons before depositing Cs.  $D_2$  gas was permeated through the Pd complexes at 343 K and 1 atm for about 5 days. Inductively Coupled Plasma Mass Spectrometry (ICP-MS) was performed to analyze the existence of the elements (Cs and Pr) and the mass distribution. The results showed the existence of Pr. And we also confirmed the existence of Pr by using fast Neutron Activation Analysis (NAA) in Fusion Neutronics Source (FNS) of Japan Atomic Energy Research Institute (JAERI).

As a result, we confirmed that the nuclear transmutation reaction, from <sup>133</sup>Cs to <sup>141</sup>Pr, was occurred. This transmutation suggests that the mass numbers and atomic numbers increase 8 and 4, respectively. The model of multi-body resonance fusion of deuterons proposed by A. Takahashi<sup>2</sup> can explain this mass-8-and-charge-4 increased transmutation.

# 1 Introduction

We received a report from Iwamura's group that unusual nuclear reactions were caused by permeating  $D_2$  gas through Pd complexes with added Cs or Sr on the surface<sup>1</sup>. The condition was that  $D_2$  gas flow was over 1 sccm, the temperature was 343 K,  $D_2$  gas pressure was 1 atm and the flow time was 1 week. (Note: the unit "sccm" is a flow rate of one cc per a minute in standard conditions.)

In contrast, permeating  $H_2$  gas instead of  $D_2$  gas showed no decreasing of Cs and no generation of Pr. The elemental analysis was done by XPS.

Following the MHI procedure exactly, we performed  $D_2$  gas permeation experiments through Pd complexes and analyzed the production of Pr. The following is the first report of this replication experiment.

#### 2 Experiments

#### 2.1 Samples

Pd complex samples were composed of a Pd thin film, alternating CaO and Pd layers and bulk substratum Pd as shown in Fig. 1. The surface of the substratum Pd plate was first covered by alternative layers of CaO and Pd

(1000Å). Then a 400-Å-thick Pd layer was sputtered on the surface of the alternative CaO and Pd layers. These processes were performed by Ar ion beam sputtering. After forming a Pd complex, Cs was deposited on the surface of the thin Pd layer. Cs atom was deposited by applying a weak electric field to 1 mM CsNO<sub>3</sub> solution. A 1 V negative voltage was applied to the Pd complex for 10 seconds.

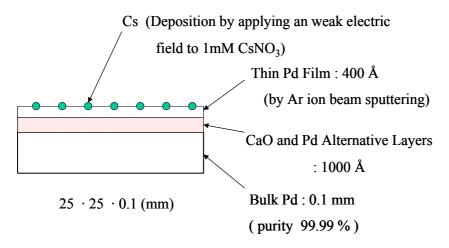


Figure 1. Sample fabricated by MHI

To remove surface hydrocarbon contamination from on the sample, we performed following pre-cleaning operation before putting Cs on the sample. The sample and a Pt plate were applied with an electric field to 1mM HNO<sub>3</sub>. Then the bubble was formed on the surface of the Cs deposit side. And Teflon was used to set up the sample and the Pt plate.

# 2.2 Experimental equipments

Chamber A was filled with 1 atm  $D_2$  gas, and Chamber B was evacuated by a turbo-molecular pump (TMP) as shown in Fig. 2, so that the front surface of a Pd complex sample was in 1 atm  $D_2$  gas and the back surface was on vacuum.  $D_2$  gas was supplied at 1 atm on the Cs side of the sample so that deuterium atoms permeate to the backside of the sample in Chamber B. The sample was normally heated to 70 degree Celsius during these experiments.

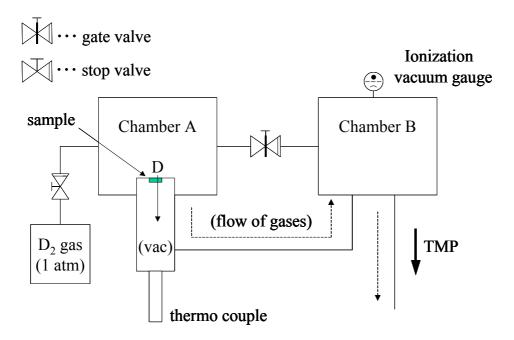


Figure 2. Experimental equipments

# 2.3 $D_2$ gas permeation experiments

# 2.3.1 The first run

Using the experimental system shown in Fig. 2, we have performed three experimental runs of  $D_2$  gas permeation through Pd complexes. We changed the temperature stepwise to 70, 60 and 80 degree Celsius and permeated  $D_2$  gas for about 120 h in the first run.

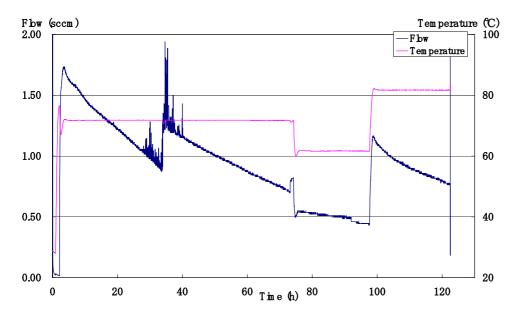


Figure 3. Relation between deuterium flow rate and temperature for the first run

We show the result in Fig. 3. The flow rate exceeded 1.5 sccm initially, decreased slowly, and then fell below 1.0 sccm, which is considered the critical value for transmutation, according to the MHI group. The flow rate increased as the temperature was stepped up, and exceeded 1 sccm again, but it soon declined again. We observed unnatural fluctuations in the flow rate between 20 h and 40 h. We assume that electrical miscontact of the ionization vacuum gauge caused this.

#### 2.3.2 The second run

In the second run, the flow rate was below 1 sccm from the beginning, so we raised the temperature from 70 to 80 degrees Celsius and permeated  $D_2$  gas for about 200 h in this experiment, as shown in Fig. 4. But the flow was still below 1 sccm. We consider the likely reason is that moisture from the atmosphere came inside the chamber and formed on the sample surface, which prevented  $D_2$  gas from permeating through the sample efficiently. The data gap near 30 h occurred when we stopped the vacuum equipment because of a shortage of  $N_2$  gas.

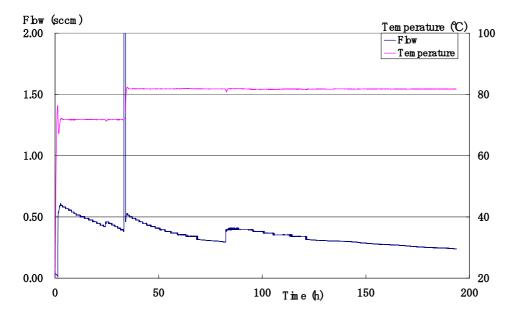


Figure 4. Relation between deuterium flow rate and temperature for the second run

#### 2.3.3 The third run

We kept the temperature constant at 70 degree Celsius and permeated  $D_2$  gas for about 120 h in the third run. To keep the sample clean, and the chamber free of moisture from the atmosphere, we first baked out the chamber, and then  $N_2$  gas was fed and evacuated before setting up the sample in the chamber. As the result, flow rate was improved to over 2 sccm at first. But because we did not have enough remaining  $D_2$  gas, the pressure in the chamber filled with  $D_2$  gas could not be maintained at 1 atm and the flow rate decreased gradually. The detailed flow rate data before 40 h was lost because of a problem in the data acquisition system, so the curve is approximated with data from 0 h and 20 h taken manually from the ionization vacuum gauge, as shown in Fig. 5.

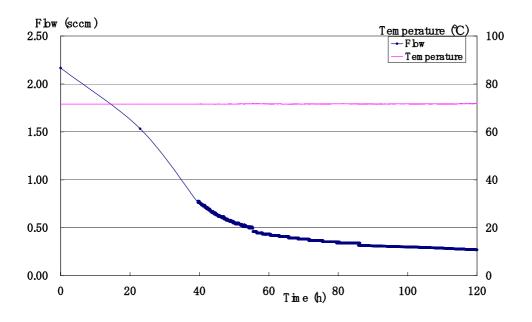


Figure 5. Relation between deuterium flow rate and temperature for the third run

## **3** Results of Element Analysis

## 3.1 Analysis by ICP-MS

The elements on the surface of the sample were analyzed by ICP-MS in the MHI laboratory. The result revealed the production of Pr atoms, although amounts were different with each run, as shown in Table 1. But a correlation exists between the amounts of Pr produced and the maximum flow rates.

	Pr (ng)	Cs (ng)	max flow (sccm)	average (sccm)	minimum(sccm)
1st run	18	180	173	0.93	043
2nd run	51	141	0.61	0.35	0.29
3rd run	36	330	217	0.76	0.27

Table 1. Results analyzed by ICP-MS in the MHI

# 3.2 Analysis by NAA

We also analyzed the Pd complex sample by Neutron Activation Analysis (NAA) using 14 MeV neutrons in Fusion Neutronics Source (FNS) of Japan Atomic Energy Research Institute (JAERI), for the sample that was permeated  $D_2$  gas in MHI and analyzed by XPS in MHI<sup>1</sup>.

Figure 6 shows the result of Ge gamma-ray spectra by 14 MeV fast neutron irradiation in the FNS. In the first measurement, the sample was irradiated for 11 days, cooled for 25 days and measured for 7 days. And at the second measurement, taken from the same sample after the first measurement, the sample was cooled for 8 days

and measured for more 9 days. We increased the HPGe detector efficiency by setting the sample very close to the detector surface for the second measurement, to improve counting statistics.

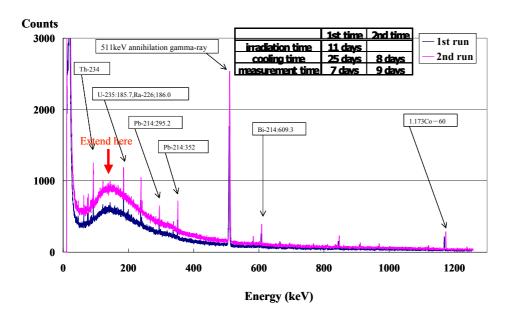


Figure 6. Ge gamma-ray spectra by NAA of 14 MeV neutron irradiation

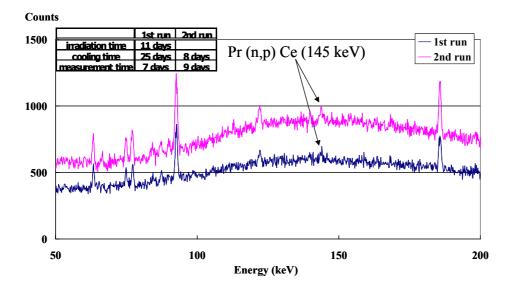


Figure 7. Expanded spectra for detail analysis by NAA of 14 MeV neutron

Figure 7 shows the region near 145 keV which indicates the existence of Pr by the gamma-ray peak at 145 keV of  $^{141}$ Pr(n,p) $^{141}$ Ce reaction. For the second measurement, we increased Ge detector efficiency, so that we compared two (first and second measurement) spectra after the normalization with 511 keV peaks of

background gamma-rays. We could see a decay of 145 keV peaks of <sup>141</sup>Ce from the first measurement to the second measurement, with half-life of about 50 days with large (more than 50 %) error bar obtained by only two data points (32.5 days is the exact half-life). This 145 keV peak with decay convinced us of the existence of <sup>141</sup>Pr in the sample. However the counting statistics for peaks of <sup>141</sup>Ce were not high enough to obtain the exact half-life. We are planning another irradiation to improve the statistics.

#### 4 Discussion and conclusion

After the three runs of experiments permeating  $D_2$  gas through in our laboratory, samples were analyzed by ICP-MS in MHI and we could confirm existence of Pr from three runs, as shown in Table 1.

And we also confirmed the existence of Pr by 14 MeV NAA in FNS as shown in Fig. 7, for the sample which  $D_2$  gas permeation was done by MHI.

In order to explain this phenomenon that Cs changes to Pr, a "theory of 8D nuclear fusion" is considered<sup>2</sup>. This model holds that two high energy Be-8 nuclei (Be\*) are produced by octahedral resonance fusion of eight deuterons in the Pd lattice, and these Be particles may be absorbed by Cs (or Sr) to cause transmutation with mass number 8 and atomic number 4 increased reaction, as shown Fig. 8. Also, these Be-8 nuclei have very short lifetime  $6.7 \cdot 10^{-17}$  s, and they decay to two stable He atoms as soon as they are formed, but we have enough collisions with Cs (or Sr) nuclei within range.

Reaction formula is as follows:

(Primary reaction): 
$$8D \rightarrow {}^{16}O^* \rightarrow {}^{8}Be + {}^{8}Be + 95.2 \text{ (MeV)}$$
  
(Secondary reaction):  ${}^{133}Cs + {}^{8}Be (47.6 \text{ MeV}) \rightarrow {}^{141}Pr^* (50.47 \text{ MeV})$   
or  ${}^{8}Be \rightarrow {}^{4}He + {}^{4}He$ 

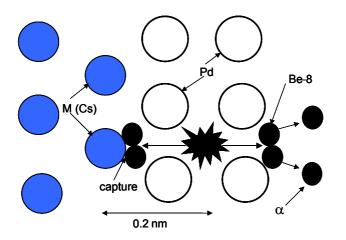


Figure 8. Model of 8D nuclear fusion

If nuclear transmutation is induced by the secondary reactions of 8D fusion, <sup>4</sup>He gas must be produced. So we are planning an experiment to confirm <sup>4</sup>He gas production by using a QMAS system.

#### Reference

- 1. Y. Iwamura et al., Proc. ICCF 9, May 19-23, 2002, Beijing, China, pp.141-146.
- 2. A. Takahashi, Proc. ICCF9, May 19-23, 2002, Beijing, China, pp.343-348.