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# Observation of Nuclear Products under Vacuum Condition from Deuterated Palladium with High Loading Ratio

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## Abstract

Gas release experiments with a method of heating highly deuterated palladium metals (D/Pd=0.7 ~ 0.83) in a vacuum chamber to induce anomalous nuclear effects have been performed. Neutron emission and X-ray emission were observed in some cases, and DT gas breeding with high reproducibility. DT gas breeding was correlative with D/Pd and degassing rate of deuterium gas. It shows that anomalous nuclear effects are related to D/Pd and diffusion process of deuterium atoms in palladium metals.

## 1. Introduction

Much research has been performed and many papers have indicated that nuclear reactions occur in deuterated palladium since the announcement of "cold fusion" phenomena by Fleischmann and Pons[1]. We previously reported performance of gas release experiments[2] in ICCF-4 and indicated neutron emissions and tritium productions from gas loading deuterated palladium samples (D/Pd≤0.66). We assume that key factors to induce the anomalous nuclear effects are as follows. First point is high D/Pd ratio. This factor is widely recognized as an important factor to cause nuclear effects. Second point is the diffusion process of deuterium atoms in palladium metals. In order to clarify the effects of these factors, we performed gas released experiments using electrochemical loading samples with high D/Pd ratio. Figure 1 demonstrates an outline

of the experiments. It consists of combined two parts. First part is the gas release experiments presented in ICCF-4. The aim of these experiments is to induce anomalous nuclear effects by the diffusion of deuterium atoms by heating deuterated palladium in a vacuum chamber. Second part is the sample preparation of deuterated palladium with high D/Pd ratio by electrochemical loading. Combining these two parts, we performed gas release experiments under the vacuum condition using deuterated palladium that has high loading ratio.

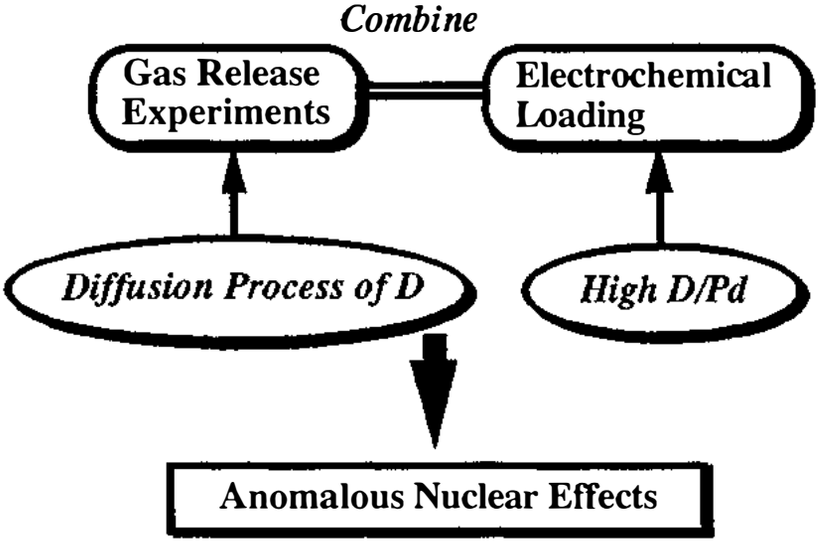


Figure 1 Outline of experiments

## 2. Experiment

An experimental procedure is as follows.

(1) Pre-treatment of a palladium sample.
(2) Deuterium loading
(3) Surface modification by Cu coating.
(4) Calculation of D/Pd ratio by measuring mass of the sample.
(5) Set the sample into the vacuum chamber.
(6) Evacuating the chamber (~$10^{-6}$Torr)
(7) Begin to measure neutron, gamma-ray, charged panicle, X-ray, pressure in the chamber, temperature of the heater and start up the quadrupole mass spectrometer.
(8) Heating the sample up to 393K
(9) Observation
(10) Heater off

Figure 2 shows an experimental device and procedure. A preparation of a deuterated palladium sample with high loading ratio was as follows. We heated a palladium rod (ø3 × 25mm: Tanaka Kikinzoku Kogyo K.K.) washed with acetone in a supersonic cleaning device in the air at 1173K for 2 hours to degrease from its surface. After that, we quenched the sample in pure water and set it in a $D_2O$-LiOD electrolysis cell. The cell was operated at constant current (0.1A) for 24 to 48 hours, and loading ratio of the sample reached about D/Pd=0.8. We electroplated the samples with Cu in $Cu_2SO_4$ electrolysis to reduce the rate of deuterium gas release and to maintain high deuterium loading ratio.

The sample was introduced into the vacuum chamber and set on a heater located in it. The chamber is equipped with two He-3 neutron detectors (EG&G Ortec: RS-P4-0806-207), two Silicon Surface Barrier Detectors (EG&G Ortec: CU-020-450-300) for charged particle spectroscopy, NaI scintillation counter (Bicron: 2M2/2) for gamma-ray spectroscopy, a CdTe (TOYO MEDIC: CDTE4BE) detector for X-ray spectroscopy and a high resolution quadrupole mass spectrometer (ULVAC: HIRESOM-2SM) for gas analysis. Neutron detectors surrounded with polyethylene modulators are used only for neutron counting. Counting system consists of preamplifiers (EG&G Ortec: 142PC), amplifier and single channel analyzer (EG&G Ortec: 590A) and counters (EG&G Ortec: 996) which are connected to a personal computer by GPIB interface. As for gamma-ray spectroscopy, we use a preamplifier (EG&G Ortec: 276), an amplifier (EG&G Ortec: 575A), and a multi-channel analyzer (SEIKO EG&G: MCA7800). X-ray spectroscopy system consists of a preamplifier (TOYO MEDIC: CT571P), an amplifier (TOYO MEDIC: 571M), and a multi-channel analyzer (SEIKO EG&G: MCA7800). We obtain energy spectra of charged particle using a preamplifier (EG&G Ortec: 142B) and amplifier (EG&G Ortec: 570), and a multi-channel analyzer (SEIKO EG&G: MCA7800).

All these devices are located in a clean-room where temperature and humidity are at constant levels (23°C±1°C,40%±5%) in order to prevent contamination and false counts induced by humidity in the air. Furthermore, we always monitor electrical signals from He-3 counters, charged particle detectors, and CdTe detector using digital oscilloscopes connected to personal computers to confirm that the signals originate from true nuclear events.

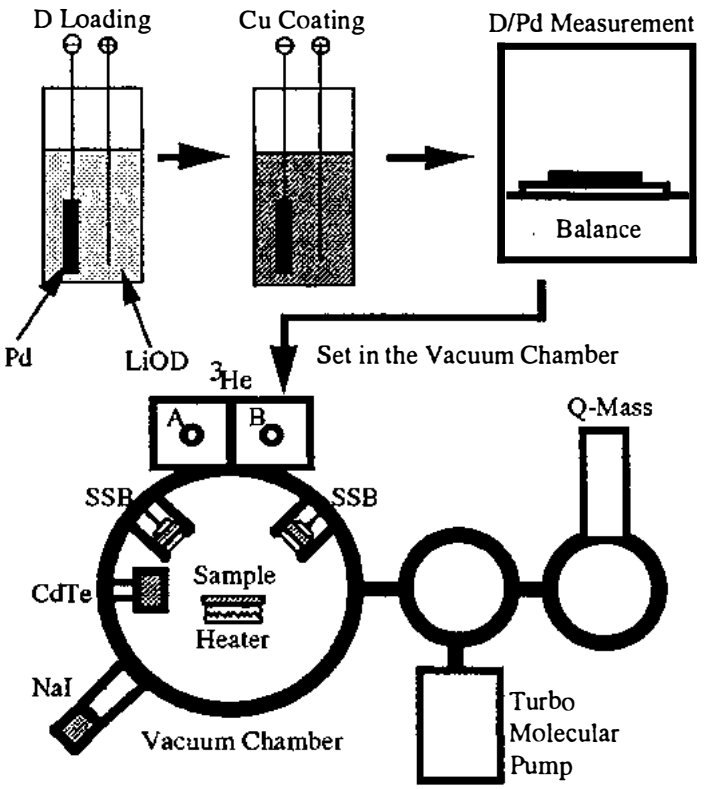


Figure 2 Experimental procedure and device

## 3. Results

Figure 3 shows an example (D/Pd=0.71) of experimental results of neutron emission.

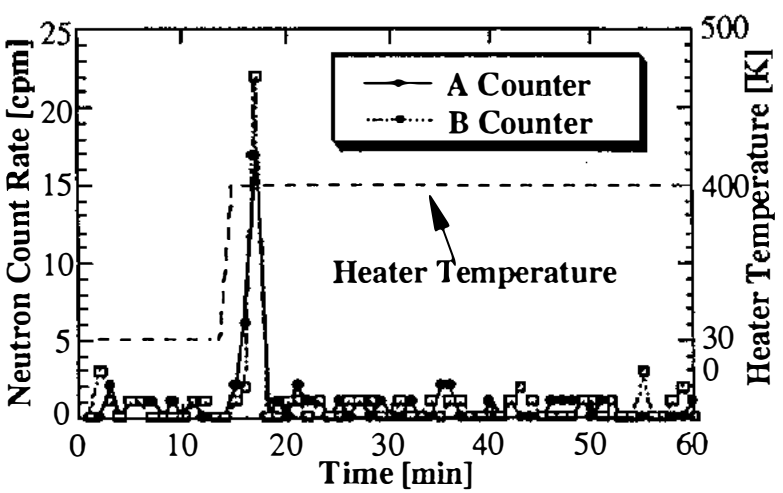


Figure 3 Neutron Emissions

At the beginning of evacuation, heater temperature equals to room temperature (296K), we can see only background neutron counting.With heating the sample up to 393K, a clear and prominent neutron emission peak is detected in both counters. If the neutron of the peak is due to fluctuation of background neutron, the probability is about $10^{-44}$. Therefor, we consider that the sample emits neutron during heating.

Concerning observation of tritium production, we mainly watch behavior of mass number 5 gas by quadrpole mass spectrometer (Q-Mass). Mass number 5 gas detected by Q-Mass consists of DT and $DDH^+$. Regarding $DDH^+$, an electron attack of a filament of Q-Mass leads to formation of ion species $DDH^+$, and partial pressure of $D_2$ and $H_2$ determines the number of $DDH^+$. Figure 4 indicates $DDH^+$ formation in our vacuum chamber. Horizontal axis means total pressure (≈$D_2$ gas pressure) and vertical $DDH^+$ ion current, respectively. We, therefore, define DT gas breeding ratio as the ratio of mass number 5 ion current to $DDH^+$ ion current. If DT gas breeding ratio is larger than 1, it is considered that a sample releases DT gas. With using this breeding ratio, we can demonstrate tritium production detected by Q-Mass.

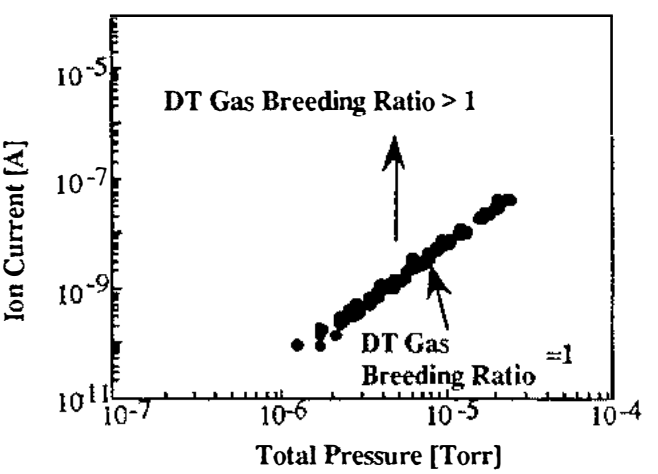


Figure 4 Formation $DDH^+$ in the vacuum chamber

Figure 5 shows time evolution of DT gas breeding ratio. D/Pd ratio of the sample is 0.83. At the beginning of evacuation, DT gas breeding ratio is about 1. However, we see the DT gas breeding ratio increases gradually during evacuation. At this time, it is considered that deuterium atoms in palladium diffuse toward outside, since a deuterium pressure gradient exists between inside and outside of the sample. Simultaneously, we observed X-ray emission as mentioned later. About 26 minutes after evacuating, we start heating up to 393K. Then the sample releases deuterium gas

rapidly, and DT gas breeding ratio increases at the same time. Although we cannot measure DT gas while the total pressure exceeds $3\times10^{-5}$ torr since our mass spectrometer is not able to work, we consider that tritium atoms are produced during gas release by heating.

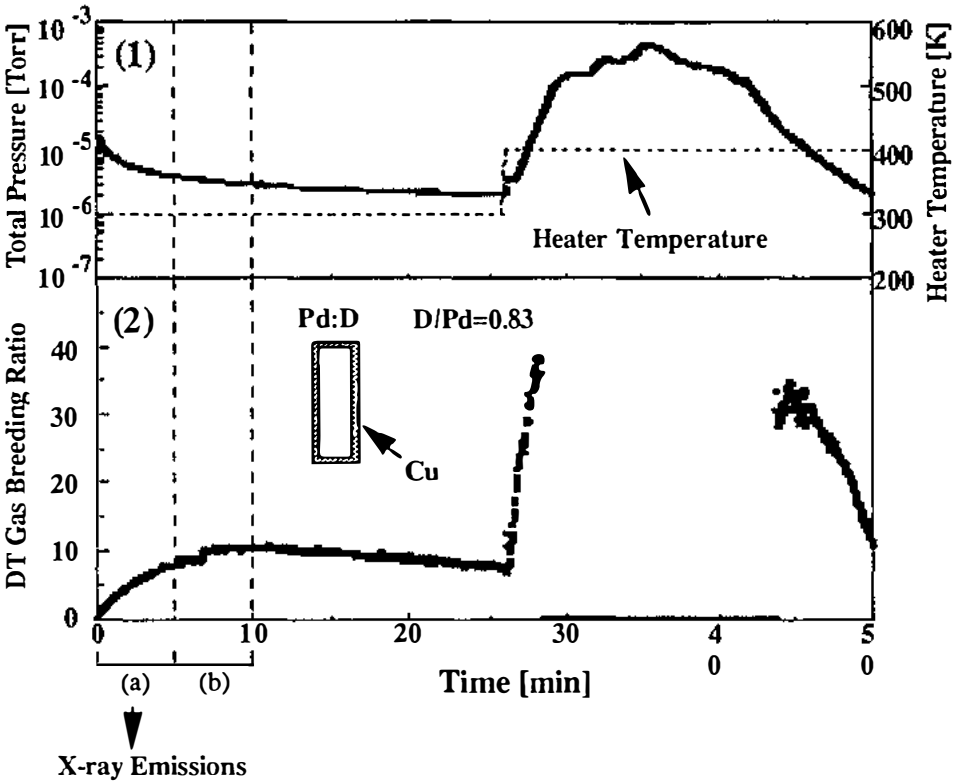


Figure 5 Tritium Production

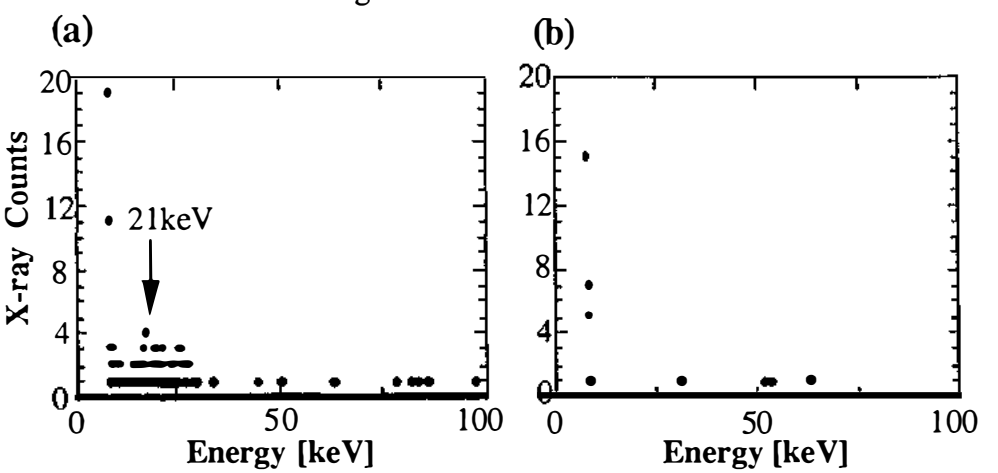


Figure 6 X-ray emission

Figure 6 demonstrates X-ray emission during evacuating. Figure 6-(a) is an energy spectrum for 5 minutes just after the beginning evacuating. This spectrum has a peak around 21keV. Figure 6-(b) is an energy spectrum from 5 to 10 minutes after evacuation. This spectrum has no peak as the spectrum of Figure 6-(a) and the sample radiates no X-ray in this period. Spectra in the other periods are similar to figure 6-(b). We consider that the deuterated palladium sample radiates X-ray that is attributed to K-$\alpha$ characteristic X-ray of palladium.

## 4. Discussions

We summarize the data of relationship between loading ratio and DT gas breeding ratio (Figure 7), since tritium production has high reproducibility in our experiment. DT gas breeding ratios are evaluated at $1.0\times10^{-5}$ Torr of total pressure after heating. DT gas breeding ratios tend to increase as D/Pd increase and to ascend rapidly around D/Pd≈0.83. Therefore, it seems that DT gas breeding ratio is related to D/Pd; density of D atoms in palladium.

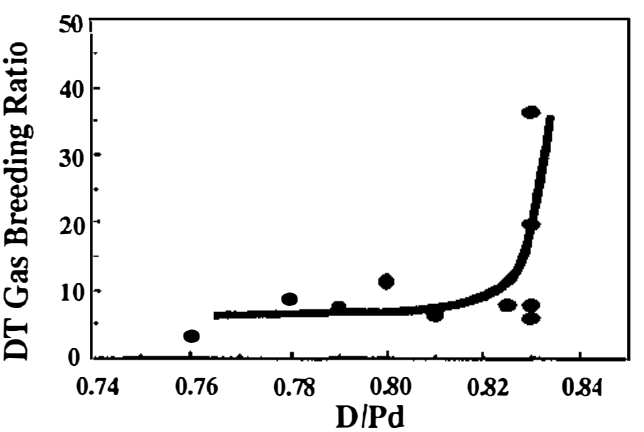


Figure 7 DT gas breeding ratio vs. D/Pd

Figure 8 shows the relation between degassing rate and DT gas breeding ratio. Degassing rate is defined as pressure equation of $Q = V\cdot dp/dt + p\cdot S$. Q; degassing rate, V; chamber volume, p; pressure, and S; the net pumping speed, respectively. The Figure indicates that DT gas breeding ratio increases as the degassing rate increases and saturated at a certain value. Though it is difficult to explain this relationship in detail, at present, degassing rate has a certain correlation with the diffusion process of deuterium

atoms in palladium metals. Therefore, it is probable that tritium production is related to the diffusion process of deuterium atoms in palladium metals.

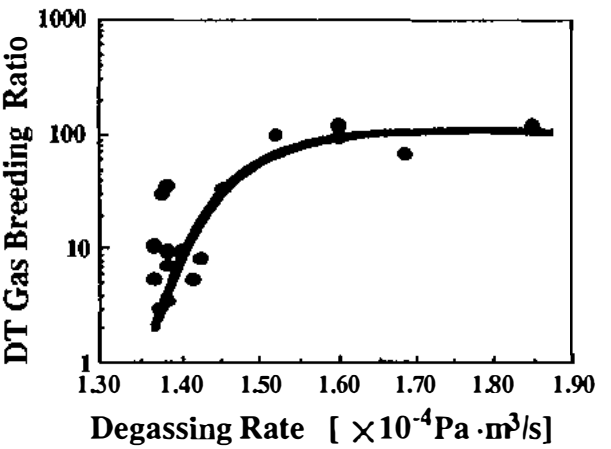


Figure 8 DT gas breeding ratio vs. Degassing rate

## 5. Conclusion

We performed gas release experiments under vacuum condition with high D/Pd samples obtained by electrochemical loading (D/Pd = 0.73 ~ 0.8). We observed neutron emissions and X-ray (about 21 keV) emissions several times, and tritium production with high reproducibility. DT gas breeding is related to D/Pd and degassing rate. It is considered that some of key factors to induce anomalous nuclear effects in Pd-$D_2$ systems are density and diffusion process of D atoms in palladium metals.

## References

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3. Y.Iwamura, N.Gotoh, T.Itoh and I.Toyoda, "Characteristic X-ray and Neutron Emissions from Electrochemically Deuterated Palladium," *Proc. of ICCF-5,* Monte-Carlo, MONACO, April 9-13,1995, to be published