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Correlation between Behavior of Deuterium in Palladium and Occurrence of Nuclear Reactions Observed by Simultaneous Measurement of Excess Heat and Nuclear Products

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Abstract

We developed a new type of experimental apparatus for simultaneous measurement of excess heat and nuclear products with intent to induce continuous nuclear reactions in D_2-Pd system. It consists of two parts: an electrochemical cell for calorimetry, and a vacuum chamber for nuclear measurement. Deuterium atoms flow continuously from the electrochemical side to the vacuum side through a palladium plate, and we might expect that nuclear reactions last for a long term in the system, since it is considered that diffusion process of deuterium atoms in palladium is important to induce nuclear reactions.

Our experimental results suggest that absorption and desorption behavior of deuterium was greatly influenced by unspecified factors (impurity or metallurgical conditions of palladium). X-ray emission lasting for long term and excess heat generation was observed, however, correlation between excess heat and the x-rays is not clear under our experimental condition.

1. Introduction

We previously reported that diffusion process of deuterium, in addition to high D/Pd ratio, is an important factor for causing nuclear reactions by the method of gas release experiments in vacuum[1,2]. Based on these experimental results, we developed a new experimental apparatus for simultaneous measurement of excess heat and nuclear products.

The basic idea of our apparatus is that we might make continuous nuclear reactions occur in the D_2-Pd system by making continuous flow of deuterium under *certain appropriate conditions.* Although high D/Pd ratio is one of the *certain appropriate conditions,* the other necessary conditions still remain unclear at present. Therefore it might be possible to clarify the other *appropriate conditions* by making continuous flow of deuterium in palladium under high D/Pd conditions; for this is considered the most probable way to induce nuclear reactions according to our experimental results[1-3].

In this paper, we describe our experimental apparatus and the results on simultaneous measurement of excess heat and nuclear products.

2. Experimental Method

Fig. 1 shows a cross sectional view of the experimental apparatus. An electrolyte of 1 M $LiOD/D_2O$ and a vacuum chamber are separated by a palladium plate. Deuterium atoms are loaded by electrochemical potential into one side of the palladium sample and released from the other side.

Excess heat is estimated by the flow calorimetry method. The electrolyte side of the apparatus consists of a palladium plate cathode (25 × 25 × 1 mm; 99.9% Tanaka Kikinzoku Kogyo K.K.), a circular shaped anode of platinum mesh (diameter > 1 mm), a recombiner and a cooling pipe for measurement of excess heat generation. Two thermocouples measure the inlet and outlet temperature of the water. Solution, gas, recombiner and environmental temperatures are measured and consistency between these temperatures is always checked.

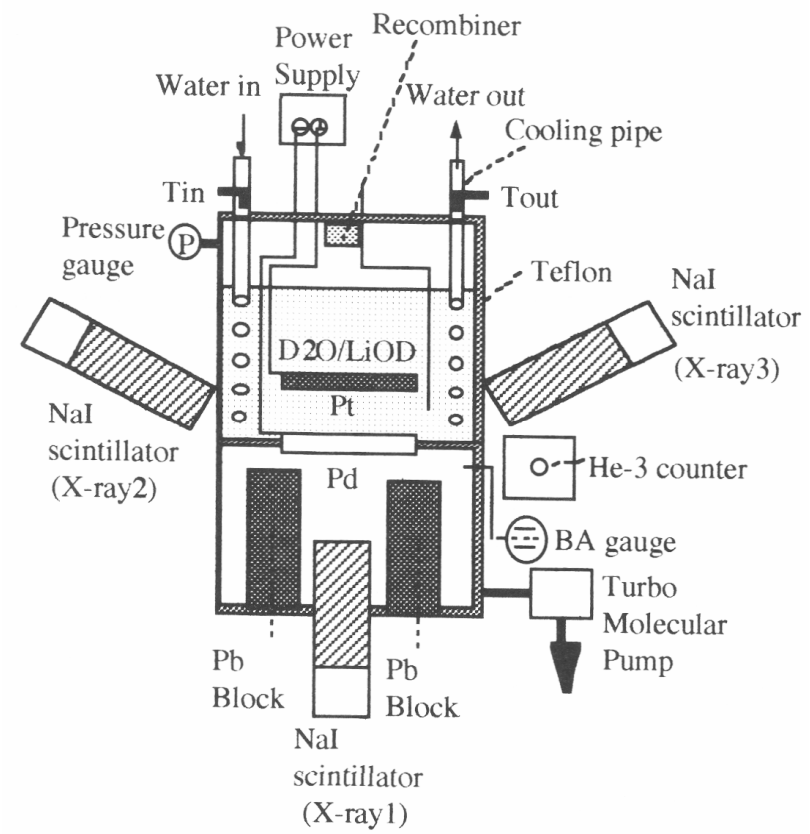

Fig. 1. Experimental Apparatus

The apparatus is equipped with three NaI scintillation counters (Bicron: 1.5XM1/2B) for x-ray spectroscopy and a He-3 neutron detector (Reuter-Stokes: RS-0806-207). The X-ray 1 detector located in the vacuum chamber is surrounded by a lead cylinder (thickness: 2.5 cm) to reduce background x-rays. X-ray spectroscopy and counting system consists of preamplifiers (EG&G Ortec: 276), amplifier and single channel analyzers (EG&G Ortec : 590A), counters (EG&G Ortec: 997) and multi-channel analyzers (SEIKO EG&G : MCA4100, 4200). As to neutron counting, we use a preamplifier (EG&G Ortec : 142PC), an amplifier and single channel analyzer (EG&G Orec: 590A) and a counter (EG&G Ortec : 997).

Pressures of the upper part of electrolyte and in the vacuum chamber are monitored by a pressure gauge and a BA gauge, respectively. D/Pd is calculated by the obtained pressures as will be described. All the experimental data is acquired by personal computers.

The apparatus and measuring systems are located in a clean-room where temperature and humidity are always controlled at constant levels (23°C ±1°C, 40% ±5%). Furthermore, all these electric devices are supplied by an isolated power source to prevent electric noise from the outside of the clean room.

The sample preparation procedure is as follows. Palladium plates were washed with acetone and annealed under vacuum condition (<10^{-7} torr) at 900°C for 10 hours. The samples were cooled down to room temperature in furnace and washed with aqua regia to remove impurity on the surface of the palladium samples. After that, some samples were covered with MgO or Al by ion beam sputtering, some samples were electroplated by copper or platinum, and the others had no films. The aim of the surface modification is to reduce the rate of deuterium gas release from the vacuum side of the palladium.

3. Data Analysis

D/Pd is estimated by the following procedure. Equation of state for the upper part of the electrolyte side is expressed as:

$$p_{O_2} V_{cell} = N_{O_2} \kappa T_{cell} \tag{1}$$

where *cell* denotes upper part of the electrolyte side and κ is the Boltzmann constant. The number of deuterium atoms absorbed into the palladium is given by

$$N_{D_{in}} = 2N_{D_2} = 4N_{O_2} = \frac{4V_{cell} p_{O_2}}{\kappa T_{cell}} . \tag{2}$$

As the vacuum chamber is evacuated by the pumping speed of *S*, an equation of mass balance is obtained as

$$\frac{d}{dt}\left(N_{vac} \kappa T_{vac}\right) = V_{vac} \dot{p}_{vac} + p_{vac} S . \tag{3}$$

where the denotation of *vac* means the vacuum side. Almost all of the gas in the vacuum chamber is composed of D_2 gas, so the time derivative of the number of deuterium atoms is given by

$$\dot{N}_{D_{out}} = 2 \frac{V_{vac} \dot{p}_{vac} + p_{vac} S}{\kappa T_{vac}} . \tag{4}$$

Combining equation (2) and (4), we obtain the following equation:

$$D/Pd = \frac{M_{Pd}}{\omega_{Pd} N_{AB}} \left[\frac{4V_{cell} p_{O_2}}{\kappa T_{cell}} - 2\int_{t_0}^{t} \frac{V_{vac} \dot{p}_{vac} + p_{vac} S}{\kappa T_{vac}} dt \right], \tag{5}$$

where N_{AB}, M_{Pd}, ω_{Pd} and T_0 denote Avogadro number, mass number of palladium, mass of the palladium sample and the time that D_2 gas begins to release out, respectively.

Next, we describe the excess heat analysis. An assumption in this analysis is that we consider only steady state and dissipated heat from the apparatus is estimated by Pd/H_2O system.

The heat balance equation of the apparatus is expressed as

$$P_{ex} + P_{in} - P_{out} - P_{diss} - \Delta\dot{H}_{gas} + \Delta\dot{H}_{abs} - \Delta\dot{H}_{des} = 0\,, \tag{6}$$

where *ex, in, out, diss, gas, abs* and *des* mean excess, input, output, dissipation, gas release, sorption and desorption. The terms of gas release, absorption and desorption of deuterium are gligible if we use typical experimental parameters. Therefore excess heat is calculated by

$$P_{ex} = \dot{m}_w c_w (T_{out} - T_{in}) - IV + \int_A h(T_{sol} - T_{room})dA \tag{7}$$

where IV is the input power, m_w and c_w are mass of the water and specific heat, h is the heat transfer coefficient and A means the whole surface of the apparatus.

4. Results and Discussion

Fig. 2 shows an example of D/Pd analysis. In this case, the sample has no surface film. Since the deuterium atoms absorbed on the surface of the electrolyte side of the palladium do not reach the opposite surface, deuterium gas is not released at the early period of the experiment. The D/Pd ratio increases gradually and attains to 0.8 at the time of 1×10^5 sec. This result indicates that D/Pd ratio reaches about 0.8 even for the palladium sample without surface barrier on the vacuum side.

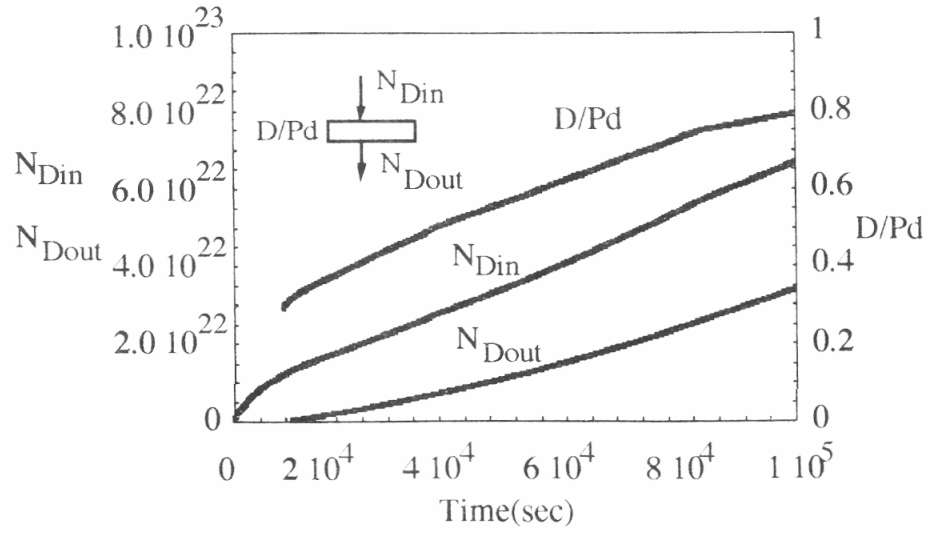

Fig. 2. D/Pd Analysis

The variation of deuterium behavior depending on palladium samples is shown in Fig. 3. The "Pressure(electrolyte)" lines indicate the pressure of the upper side of the electrolyte side which corresponds to the quantity of deuterium going into palladium, and the "Pressure(vacuum)" lines indicate the vacuum side proportional to time derivatives of the number of released deuterium atoms.

These samples, EV29 and EV34, are prepared by the same procedure; they are derived from the same lot, and the method of annealing and etching is all the same. However, it is easily seen

that that the absorption and desorption of deuterium is entirely different. This suggests that absorption and desorption behavior of deuterium is greatly influenced by unspecified factors; metallurgical conditions such as impurity and defects in palladium.

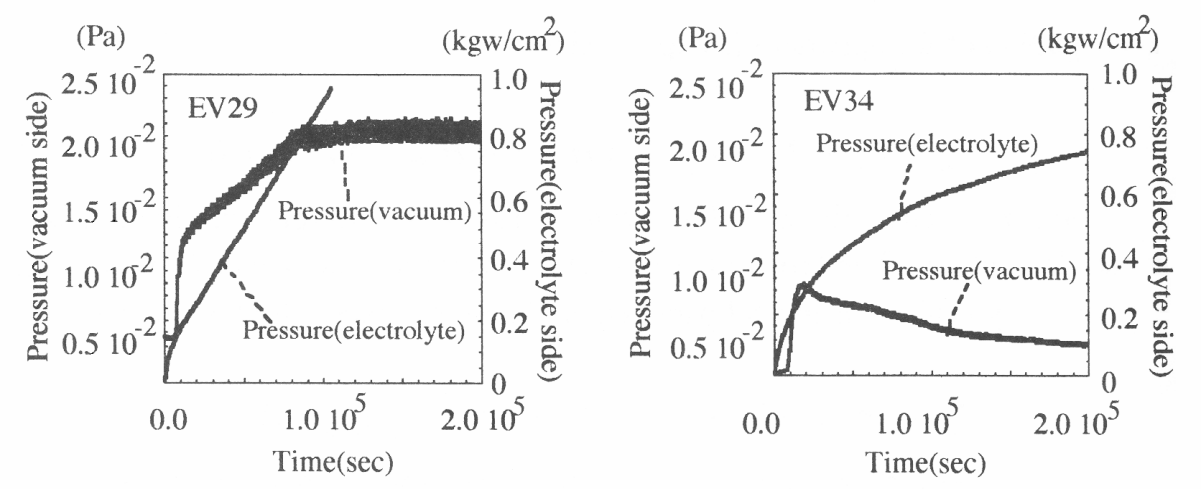

Fig. 3. Variation of Absorption and Desorption of Deuterium

An example of long duration x-ray emission and its energy spectrum is shown in Fig. 4. This sample has Al thin film (~400 A). Upper figure indicates time variation of x-ray 1 (see Fig. 1) located in the vacuum chamber. X-ray count rate reaches over 10 times larger than that of background. We can see that x-ray emission lasts for a long time; more than 1 day. It was a successful example in that we were able to make a long-term continuous nuclear reactions occur in the Pd-D_2 system.

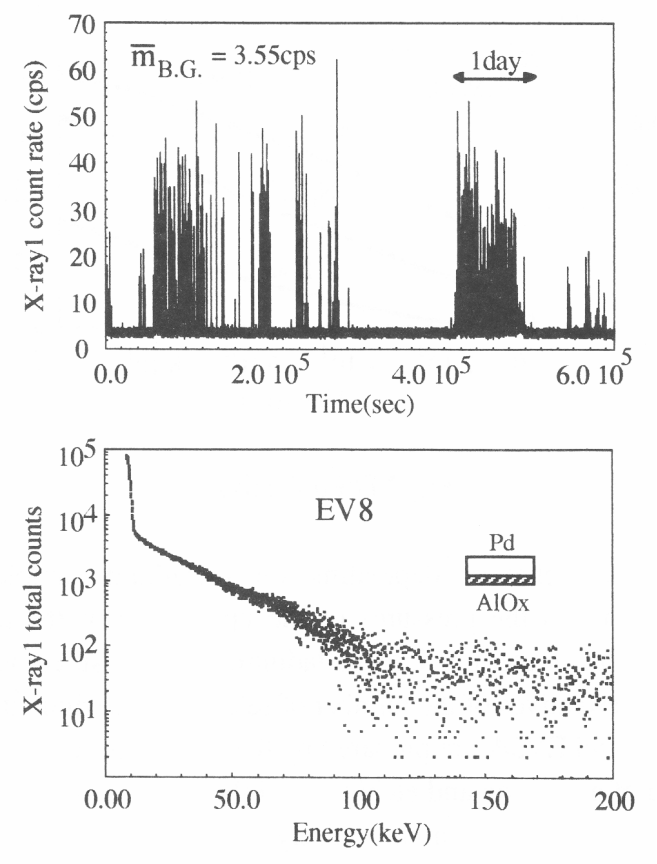

Fig. 4. Long-term X-ray Emission and its energy spectrum

The energy spectrum of the x-ray emission for about a week is plotted in the lower figure. X-ray energy distributes continuously as shown in the figure. However, at present, the authors cannot explain consistently why such an energy spectrum was obtained. It is an important topic for future work.

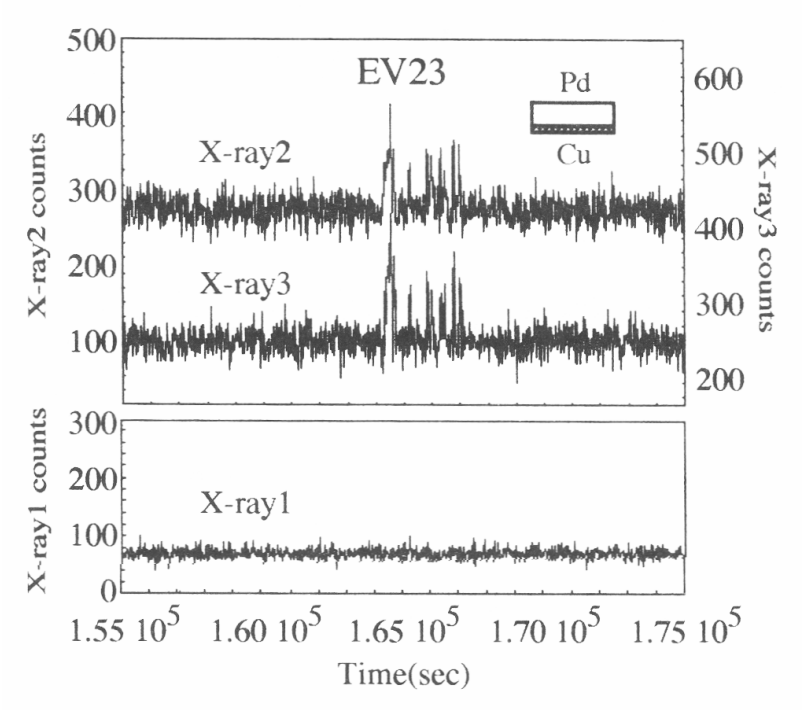

Fig. 5. Simultaneous detection by upper side x-ray detectors

The next example shows simultaneous detection by the upper side of x-ray NaI counters (Fig. 5). Sample EV23 is electroplated with copper. The coincidence of x-ray 2 and x-ray 3 is very good, but on the other hand, counts of x-ray 1 are almost equal to those of background. In 14 out of 40 cases, we observed this kind of x-ray emission. Judging from the experimental results, nuclear reactions must occur on the electrolyte side of the palladium. Since the thickness of palladium is 1 mm, the reduction rate of x-rays through the palladium is larger than that through the D_2O (4 cm).

Figure 6 illustrates the correlation between excess heat and other parameters. In this case (EV39), we use a palladium sample covered with MgO thin film. At low current, no excess heat can be seen. However, after increasing current up to 3 A (current density: 1.5A/cm^2), we observed clear excess heat generation. Input power is about 40 W when current of 3 A is applied, therefore the excess heat we obtain is a few percent of the input power.

A comparison of heat distribution between EV39 (excess heat generation) and EV40 (no excess heat) is shown in Figure 7. In both cases, the other experimental conditions are the same except the palladium sample treatment. MgO thin film is formed on the vacuum side of EV39, and Fe thin film on the electrolyte side of EV40. It is clear that the mean value and deviation of excess heat for EV39 are larger than those of EV40.

Up to now, we observed excess heat generation several times. However, we could not see any relation between excess heat generation and x-ray emission. In some cases, excess heat generation seems to be related with pressure in vacuum chamber; it corresponds to time derivatives of the number of deuterium atoms. As for neutron measurement, a clear emission has not been observed during the experiments using this apparatus. Nevertheless, the authors feel that we must investigate further before we can reach a conclusion regarding the correlation between excess heat and nuclear products.

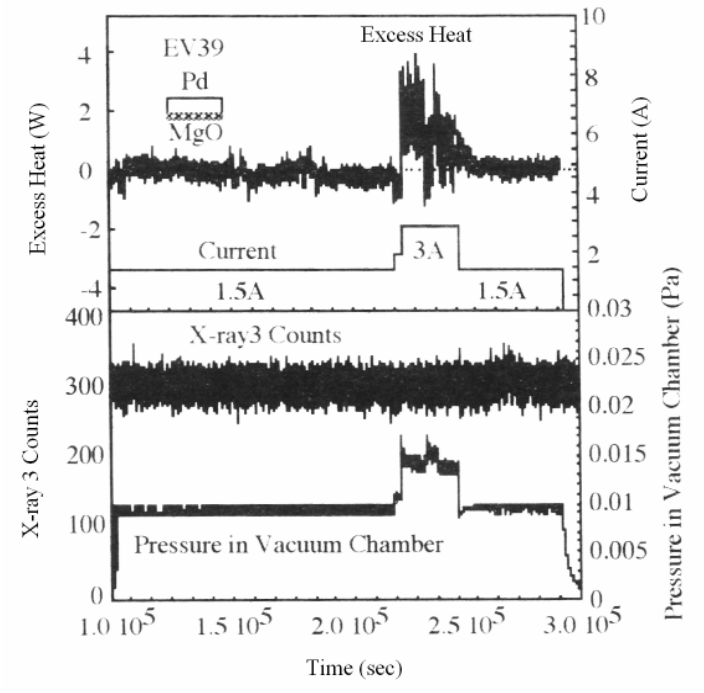

Fig. 6. Correlation between Excess Heat and the other parameters

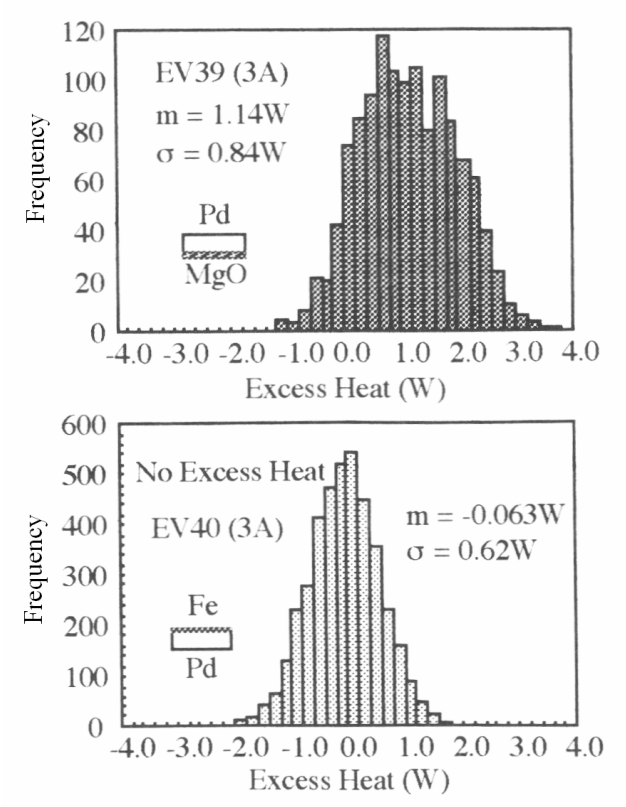

Fig. 7. Excess Heat Distribution

If we assume a few MeV of energy are released by an event, the excess heat we obtained in these experiments corresponds to the order of 10^{12} events/sec. On the other hand, the observed x-ray emissions range from 10^2 to 10^4 events/sec (calibrated with an Am-241 source). Judging from these results, we might postulate that excess heat and x-rays are generated by different nuclear reactions. In our previous paper[3], it was suggested that x-ray and neutron are generated by different reactions. Therefore it seems that we should not exclude the possibility that various nuclear reactions occur and produce various nuclear ashes.

5. Concluding Remarks

A new type of experimental apparatus for simultaneous measurement was developed. Absorption and desorption behavior of deuterium in palladium was greatly changed by unspecified conditions of the palladium. Long-duration X-ray emission, and excess heat generation was observed. The correlation between excess heat and x-rays has not been made clear under our experimental condition. We must investigate further to reach conclusions about the correlation between excess heat and nuclear products.

References

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