

PRODUCING ELEMENTS OF MASS NUMBER 137 AND 141 BY DEUTERIUM PERMEATION ON MULTI-LAYERED Pd SAMPLES WITH Cs DEPOSITION

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Abstract: An elemental analysis of Pd samples after deuterium permeation experiment was performed using TOF-SIMS. The TOF-SIMS has provided the marked count peaks at mass numbers 135 and 137 in spectra after deuterium permeation at 70 °C, only when the multilayered Pd sample with a small amount of Cs was used. The substance with mass number 137 could be ^{137}La , ^{137}Ba or ^{137}Cs . They were produced during deuterium permeation by some nuclear transmutation occurring on/in the uppermost of multi-layered Pd sample. The single and five couples of Pd/CaO thin films on Pd foil might contribute to induce production of an element with mass number 137. This would imply a transmutation of 4 mass number increasing before ^{141}Pr production.

Keywords: Pd-deuteride, Deuterium permeation, Transmutation, TOF-SIMS, Surface analysis

1. Introduction

Nuclear transmutation from a selected element into another one at near room temperature is of interest to give important information for constructing a standard model of low energy nuclear reaction. Among several experimental methods for the transmutation, the gas permeation method is one of the promising methods. Applying this method to Pd film complexes, Iwamura et al. reported a low energy nuclear transmutation from Cs into Pr and from Sr into Mo¹⁾. They used five couples of Pd/CaO thin films on a basal bulk Pd foil in their experiment. Kitamura et al. have investigated the transmutation from Sr into Mo using another permeation method and have revealed that the transmutation takes place between the thin film and the base Pd foils.²⁾

We have taken advantage of these gas permeation method to perform the transmutation from Cs into Pr using single couple of Pd/CaO thin films instead of the five couples. We have not observed marked amount of Pr but detected a small amount of substance with mass 137 using TOF-SIMS^{3, 4)}. Next, we have performed several reference experiments to search for the formation of the substance of mass number 137. The substance with mass 137 has not observed for

these reference experiments; the series of test gave a strong suggestion that the substance of mass 137 formed during the D₂ gas permeation through the multi-layered Pd sample was a newly produced element ^{137}La , ^{137}Ba or ^{137}Cs .

It might be possible that the result in almost no production of Pr in our experiment was due to the low number of the couple of Pd/CaO thin films. Thus, in this present investigation, we have compared the result for samples with single couple with that for five couples of Pd/CaO films after the deuterium permeation experiment.

2. Experimental

The experimental method and setup to investigate the transmutation of Cs into other elements of larger mass number are basically the same as before.^{3, 4)} The multi-layered sample consisted of single or five couples of CaO and Pd thin films on the basal Pd foil of 0.1 × 12.5 × 12.5 mm in size. The CaO and Pd thin films were formed in this order on the Pd foil by Ar ion beam sputtering. After forming the thin films, small amount of Cs was deposited on the multi-layered Pd sample by an electrochemical method. Each thickness of

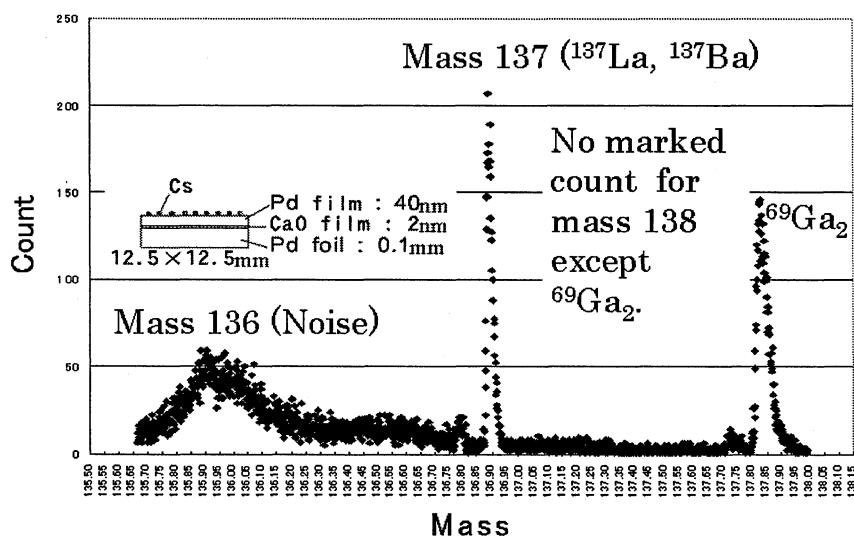


Fig. 1 TOF-SIMS spectrum around mass number 137 for a multi-layered Pd sample with single couple of Pd/CaO thin films after deuterium permeation.

CaO and Pd films formed were 2 and 18 nm, respectively. Only the thickness of uppermost Pd film was 40-50 nm. No deuterium gas was loaded to the samples before deuterium permeation experiment.

The chamber was usually filled with N₂ gas under non-experiment condition.

Just before the permeation experiment, the Pd samples were set into the sample holder in an air environment and it was placed at the vacuum chamber.

Then, the chamber was filled with deuterium gas at a pressure 0.1 or 0.2 MPa; the thin Pd film side of multi-layered Pd sample with small amount of Cs was exposed to D₂ gas. The other side of sample was evacuated by a turbo molecular pump. The deuterium permeated from the chamber through the Pd sample to the evacuated side by the pressure gradient for about 2-4 weeks. A heater was employed to keep the temperature of the chamber at 70°C during the experiment.

After the permeation experiment, the heater is turned off and the chamber was filled with N₂ gas, then the sample ("Permeation sample") was taken out from the holder. The sample surface of gas-filled side was analyzed by Time-of-flight secondary ion mass spectroscopy (TOF-SIMS)

(ULVAC-PHI: TFS-2100).

The primary ion in TOF-SIMS was Ga⁺ and we measured at least three randomly selected areas of 40×40 micron square. The spectra were obtained before and after a sputter cleaning of the uppermost surfaces of samples by the Ga⁺ for 5-10 sec. In order to take into account the contamination from the environment, we prepared the control sample ("Control sample") without flowing the deuterium gas, which was prepared by the same procedure for the permeation samples.

3. Result and Discussion

TOF-SIMS spectrum of mass number range 136-138 after sputter cleaning for a sample with single couple of Pd/CaO thin films is presented in Fig. 1. The schematic view of cross-section of sample is shown in this figure. An anomalous peak is seen at mass number 137. Another anomalous peak was also observed at mass number 135 for the same measured area, even though the count intensity was lower than that of the mass 137. These peaks were not observed for control samples. Considering the difference between the measured mass of substance with mass 135 and that of ¹³⁵Ba, the substance seems to be molecule CsD. The peak observed at mass 136 could be due to

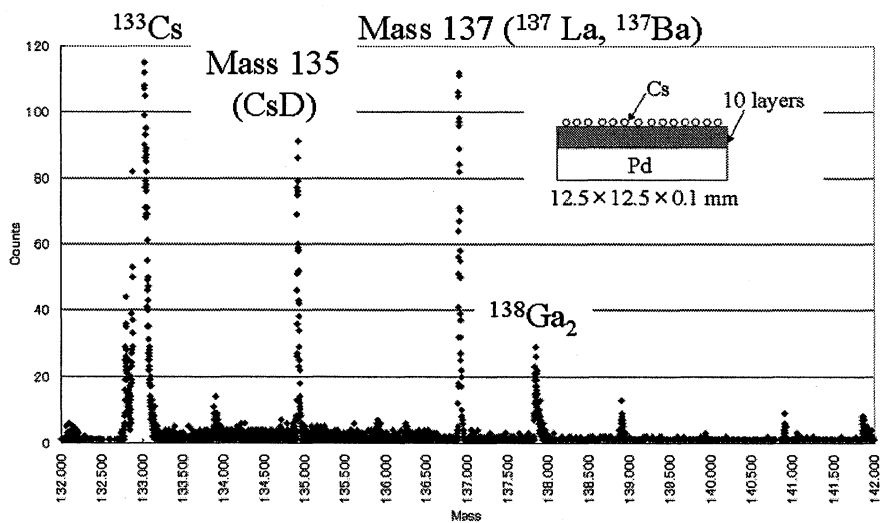


Fig. 2 TOF-SIMS spectrum of mass number range 132-142 for a multi-layered Pd sample with five couples of Pd/CaO thin films after deuterium permeation.

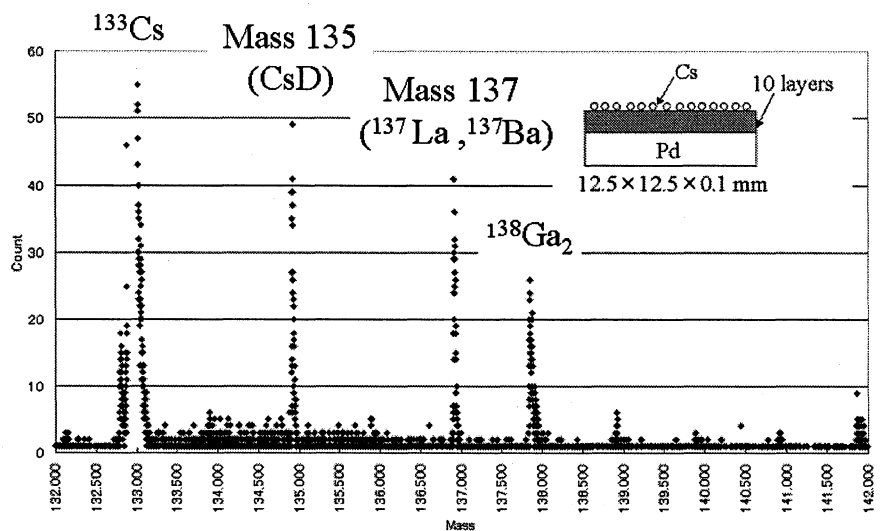


Fig. 3 TOF-SIMS spectrum of mass number range 132-142 for a multi-layered Pd sample with five couples of Pd/CaO thin films after deuterium permeation.

nose signal relating to the TOF-SIMS characteristics.

Similar spectrum for a multi-layered Pd-base sample with five couples of Pd/CaO thin films is shown in Fig. 2 and 3. Similar peaks as that for single couple of Pd/CaO thin films are seen at mass 137 in these figures. The peak intensity at mass 137 is higher than that at mass 135 in Fig. 2. However, the peak intensity at mass 137 in Fig. 3 is

slightly lower than that of mass 135; such lower peak at mass 137 was sometimes observed. This indicates that the formation process for the substances of mass 135 is independent of that of mass 137.

Table 1 gives the candidates of the substances of mass 137. The peak position of Cs on TOF-SIMS spectrum was the standard against which we have measured the peak position of substance of mass number 137.

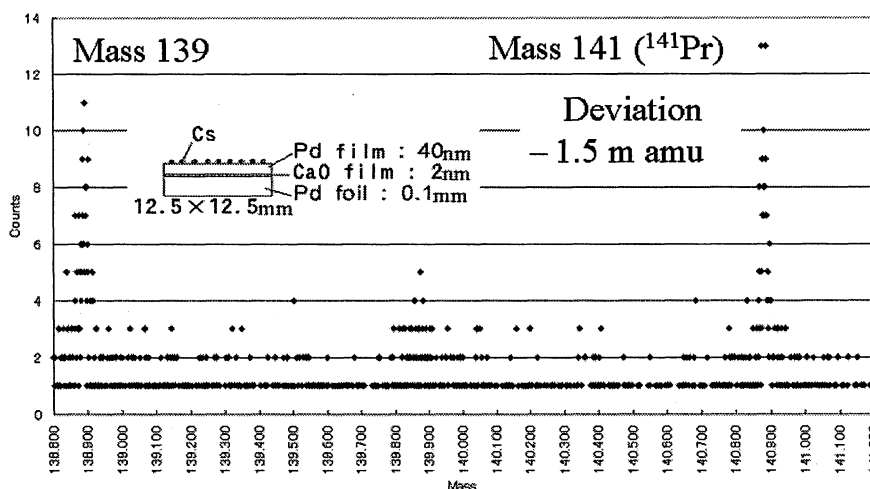


Fig. 4 TOF-SIMS spectrum of mass number range 139-141 for a multi-layered Pd sample with five couples of Pd/CaO thin films after deuterium permeation.

The peak deviation in the Table 1 is defined as the difference between the measured mass number and each real mass number of substance, that is, 136.9065-(each real mass number).

Table 1. Candidates of the substances with mass 137

	Mass (amu)	Peak deviation (m amu)
CsD ₂	136.9336	-27.1
¹⁰⁶ PdOCH ₃	136.9219	-15.4
¹³⁷ Ba contaminant		
¹³⁷ La	136.9065	0.0
¹³⁷ Ba	136.9058	0.7
¹³⁷ Cs	136.9071	-0.6

As mentioned above, the count intensity observed at mass 137 was usually higher than that observed at mass 135. This is the characteristics commonly observed in almost all the samples and almost all the measured areas. In general, amount of CsD formed by chemical process during the deuterium permeation experiment is thought to be much more than that of CsD₂. Accordingly, the substance corresponding to mass 137 cannot be CsD₂. Furthermore, the absolute value of peak deviation -27.1 m amu for CsD₂ in the

Table 1 is enough larger than that of ± 3 m amu, which is the rough value of the error range. This also ascribes that the substance of mass 137 must not be CsD₂.

We have reported that a Pd compound of PdOCH₃ could be formed at the uppermost surface of multi-layered Pd sample after the D₂ Permeation⁴⁾. This means that small amount of PdOCH₃ would be exist on the surface of the sample even after the sputter cleaning. On the basis of Pd natural isotopic abundance, the amount of Pd isotopic compounds formed should be proportional to the abundance values. The natural isotopic abundance of ¹⁰⁵Pd, ¹⁰⁶Pd and ¹⁰⁸Pd are 22, 27 and 26%, respectively. It should have been expected that count intensity at mass 139 by ¹⁰⁸PdOCH₃ was as large as that at mass 137 when the substance of mass 137 was ¹⁰⁶PdOCH₃. In other words, a marked count due to ¹⁰⁸PdOCH₃ should have been seen at mass 139 in Fig. 2 and 3. However, there existed a peak of extremely low count intensity at mass 139. Consequently, the contribution of ¹⁰⁶PdOCH₃ to form the peak at mass number 137 is thought to be negligible. The absolute value of peak deviation -15.4 for ¹⁰⁶PdOCH₃ in Table 1 is enough larger than that of the rough value of the error range. Furthermore, no marked peak appeared at

mass number 136, which corresponds to the mass number of $^{105}\text{PdCOH}_3$. Considering the fact that Pd has large natural isotopic abundance in mass number 105, 106 and 108, the compounds of ^{106}Pd cannot account for the marked count at mass 137 in Fig. 1, 2 and 3. Therefore, molecule $^{106}\text{PdOCH}_3$ is out of the candidate substance.

Similarly, on the basis of Ba natural isotopic abundance, the amount of Ba isotopic compounds formed should be proportional to the abundance values. The natural isotopic abundance of ^{137}Ba and ^{138}Ba are 11 and 72%, respectively. Thus, the element of ^{137}Ba contaminant can be possible candidate only when remarkably large count intensity was observed at mass 138. To the contrary, we have not recognized such large peak at mass 138 as seen in Fig. 1, 2 and 3, where only a peak corresponding to Ga_2 is seen. This implies that the substance of mass 137 must not be ^{137}Ba contaminant.

Thus, the substance of mass number 137 observed only after deuterium permeation and only with Cs deposition is unlikely to be contaminants but would be an elements ^{137}La , ^{137}Ba , or ^{137}Cs . Among these three possible candidates, ^{137}La might be the most likely candidate, because only the transmutation from ^{133}Cs into ^{137}La takes place via an alpha particle formation. There are other unstable ten candidate elements with mass 137, including such as ^{137}Xe . However, these elements have relatively short half life of time range s-h. Thus, we have excluded these ten elements from the possible candidates.

A TOF-SIMS spectrum of mass number range 139-141 after sputter cleaning for the multi-layered Pd sample with single couple of Pd/CaO thin films is shown in Fig. 4. It is interesting to note that the count intensity at mass 141 is often slightly higher than that at mass 139 as seen in Fig. 4, even though both intensities are relatively low. The absolute value of peak deviation -1.5 of the substance with mass 141 is within the that of the rough error range ± 3 m amu. This provides a possibility that the detected substance of mass 141 might be ^{141}Pr . Considering the results of experiment by Iwamura et al.¹⁾, all

the results obtained in this study suggest an important role of alpha cluster in the transmutation process of 4, 8 and 12 mass number increasing.

4. Conclusion

TOF-SIMS was employed for elemental analysis of surface of multi-layered Pd-base samples with single and five couples of Pd/CaO thin films. The TOF-SIMS has provided the marked count peaks at mass 137 in spectra after deuterium permeation at 70°C, only when a small amount of Cs was deposited on the uppermost surface of sample. The result suggests production of an element with mass 137 which would be produced from ^{133}Cs . The substance with the mass 137 could be ^{137}La , ^{137}Ba or ^{137}Cs . They were produced during deuterium permeation by some nuclear transmutation occurring on/in the uppermost of multi-layered Pd sample. The single and five couples of Pd/CaO thin films on Pd foil might contribute to induce production of an element with mass number 137. This would imply a transmutation of 4 mass number increasing before ^{141}Pr production.

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