

OBSERVATION OF NUCLEAR TRANSMUTATION REACTIONS INDUCED BY D₂ GAS PERMEATION THROUGH Pd COMPLEXES

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We have been studying low-energy nuclear transmutations induced by D₂ gas permeation through Pd complexes (Pd/CaO/Pd). We presented experimental results at ICCF9 and ICCF10. In this paper, we report recent progress. Transmutations of Ba into Sm were observed in two cases: with natural Ba on Pd complex samples (a definite result), and with mass 137-enriched Ba (probable). In these experiments, the atomic mass increase was 12 and atomic number increase was six. One of our experimental apparatuses was carried to SPring-8, which is the world's largest synchrotron radiation facility, located at Hyogo prefecture in Japan. Pr was confirmed several times by XRF at SPring-8. Some experiments were done to explore physical structure of the CaO layer. According to a D⁺ ion beam bombardment experiment performed at Tohoku University, the deuterium density of our Pd complex is one order larger than normal Pd. When we replaced CaO with MgO, we did not obtain any positive results. These results shed light on the role of the CaO layer in the Pd complex.

1. Introduction

Anomalous low-energy nuclear transmutation reactions, such as the transmutation of Cs into Pr, have been observed on the Pd complexes, which are composed of Pd and CaO thin film and a Pd substrate. The reactions occur when the Pd complexes are subjected to D₂ gas permeation.¹⁻⁴

Figure 1 shows a schematic of our experimental method. This method can be characterized by the permeation of D₂ gas through the Pd complex and the addition of an element that is specifically targeted to be transmuted.

Permeation of deuterium is performed by exposing one side of the Pd complex to D_2 gas while maintaining the other side under vacuum conditions. On the D_2 gas side, dissociative absorption causes the D_2 molecules to separate into D atoms, which diffuse through the Pd metal toward the vacuum side where they emerge from the Pd metal, combine, and are released as D_2 gas.

The second feature is the addition of an element targeted to be transmuted. Our sample is a Pd complex composed of bulk Pd on the bottom, alternating CaO and Pd layers, and a Pd thin film on top. After fabricating a Pd complex, Cs, Ba, Sr, or some other element is deposited on the surface of the top thin Pd layer. We can observe transmutation of the added Cs or Ba. In other words, with this composition, we can provide a deuterium flux through the Pd complex on which a target element is placed as a target to be transmuted. We perform in situ elemental analyzes of the given elements after D_2 gas permeation, by evacuating the D_2 chamber and using the built in X-ray photoemission spectroscopy (XPS) unit.

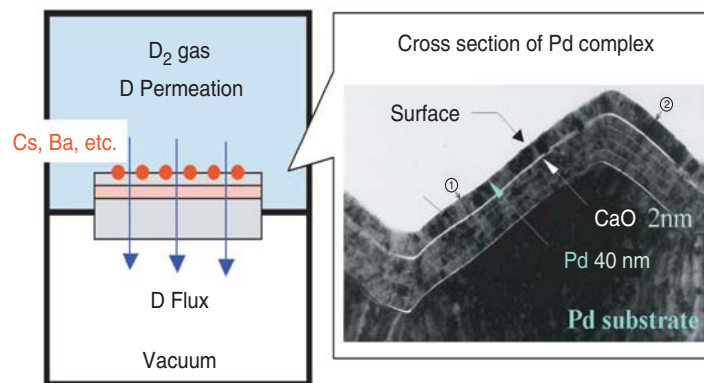


Figure 1. Schematic of the present method.

Let us briefly summarize the experimental results presented so far. The main experimental results are as follows:¹⁻⁴

- (1) Transmutation reactions of Cs into Pr were observed by D_2 gas permeation for about 1 week through Pd complexes. The D_2 gas pressure was about 1 atm and the temperature of the Pd complex was nearly 70°C .
- (2) Transmutation of Cs into Pr was demonstrated in more than 60 cases, with reproducibility close to 100%.
- (3) Transmutation of Sr into Mo was observed three times after D_2 gas permeation for 2 weeks. The isotopic composition of all detected Mo was different from the natural isotopic abundance of Mo.
- (4) The Pr was crosschecked by various methods such as XPS, TOF-SIMS (time of flight secondary ion mass spectrometry), XANES (X-ray absorption near

edge structure), XRF, and ICP-MS (inductively coupled plasma mass spectrometry).

- (5) Based on an analysis of the depth profile of the Pr, a very thin surface region up to 100 seemed to be active transmutation zone.
- (6) Experimental results suggested that the conversion rate from Cs into Pr, which is the ratio of detected Pr to Cs, was positively correlated with deuterium flux through Pd complex.

In this paper, we describe the following recent progress and results:

- (1) transmutations of Ba into Sm,
- (2) confirmation of Pr by X-ray fluorescence (XRF) spectrometry at SPring-8,
- (3) some experiments relating to the role of CaO layer in the Pd complex.

2. Experimental

The experimental method and setup are basically the same as before.¹⁻⁴ Therefore, we shall omit a detailed description, and describe only the changed and improved points of the experiment.

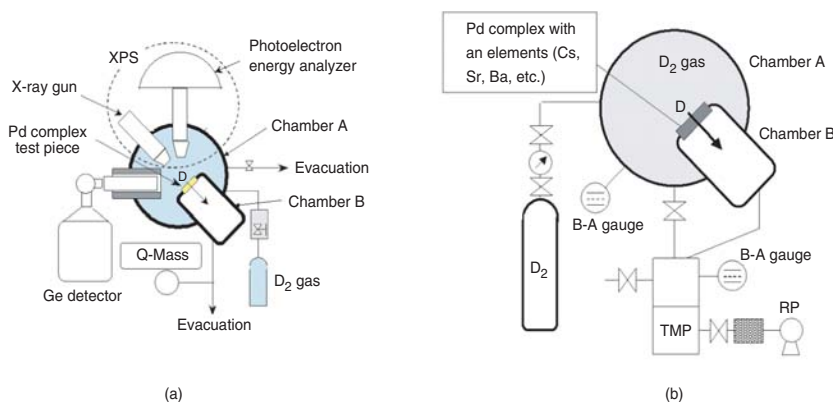


Figure 2. Schematic view of experimental setup: (a) semi-in-situ measurement apparatus with XPS and (b) ex situ measurement apparatus without XPS.

Originally, we used an experimental apparatus with XPS (X-ray photoelectron spectrometry) shown in Fig. 2a. Elemental changes on Pd complexes were measured by XPS without taking them out of the vacuum chamber, to prevent contamination from outside of the chamber. It is necessary to evacuate the D_2 gas during XPS measurement, because XPS does not work when the gas is present. This does affect the environment in the chamber; so strictly speaking, this should not be called an “in situ” method, although the Pd complexes are kept at the same position during experiments. It might be called a “semi-in-situ” method.

We introduced an additional permeation apparatus without XPS shown in Fig. 2b. Using this apparatus, only external (“ex situ”) measurements are possible, with instruments such as ICP-MS, SIMS, and XPS.

Figure 3 shows the experimental setup for in situ measurement at SPring-8. We developed this apparatus and carried to SPring-8, which is the world’s largest synchrotron radiation facility, located in the west part of Japan. This setup enables us to observe elemental changes during D₂ gas permeation by XRF.

In the case of SPring-8 experiments, Cs was deposited by the ion beam implantation method (voltage: 5 kV, dose: $(2.5\text{--}5) \times 10^{14}/\text{cm}^2$). We permeated D₂ gas through a Pd complex with Cs for 10–14 days. D₂ gas pressure is about 170 kPa and the temperature was 70°C. XRF was performed during D₂ permeation in situ at the beginning and the end of the experimental runs.

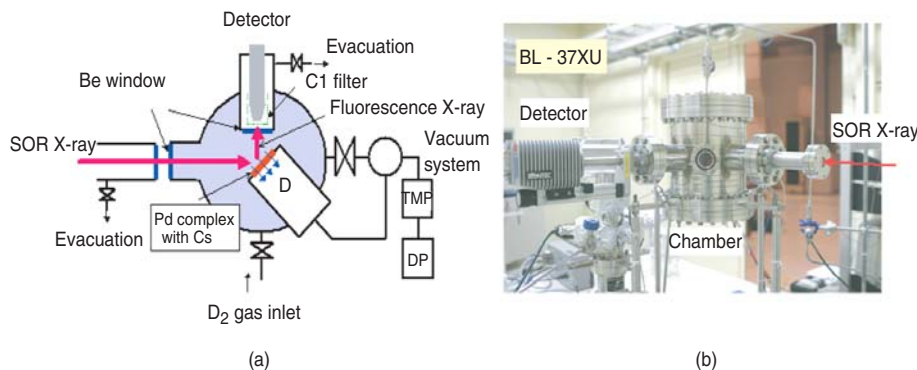


Figure 3. Experimental set-up for the in situ measurement located at the SPring-8 synchrotron laboratory, BL-37XU beamline. (a) schematic of in situ measurement apparatus and (b) photograph of the equipment we brought to SPring-8.

3. Results and Discussion

Let us describe Ba transmutation experiments. Natural Ba was deposited on some samples using the electrochemical method, in a 10 mM Ba(OH)₂ solution. On other samples, a special form of Ba with enriched ¹³⁷Ba was deposited, in a 7.3 mM Ba(NO₃)₂ solution. Applied voltage was 1 V and deposition time was 10 s, the same as the Cs and Sr experiments.

Figure 4 shows XPS spectra for a Pd complex after deuterium permeation lasting 2 weeks, starting with natural Ba. The full XPS spectrum is shown in Fig. 4a. Peaks for Ba and Sm 3d and Sm 4d can be seen. Figure 4b shows the Sm 3d spectra. Measurements were performed twice to test measurement reproducibility. Clear Sm spectra were obtained in both measurements.

In order to investigate the Sm isotopic distribution, we analyzed the detected Sm by SIMS. The natural abundance of Sm and the SIMS spectrum of the Sm

found on the Pd complex are shown in Fig. 5a, b, respectively. Natural Sm has seven isotopes; the most abundant is ^{152}Sm . The Sm spectrum on the complex was quite different. This clearly was Sm, as shown by the XPS spectra. Figure 5b shows data from a Pd complex with natural Ba on the surface after D_2 permeation (marked “Used”), and for a complex that has not been permeated (“Unused”).

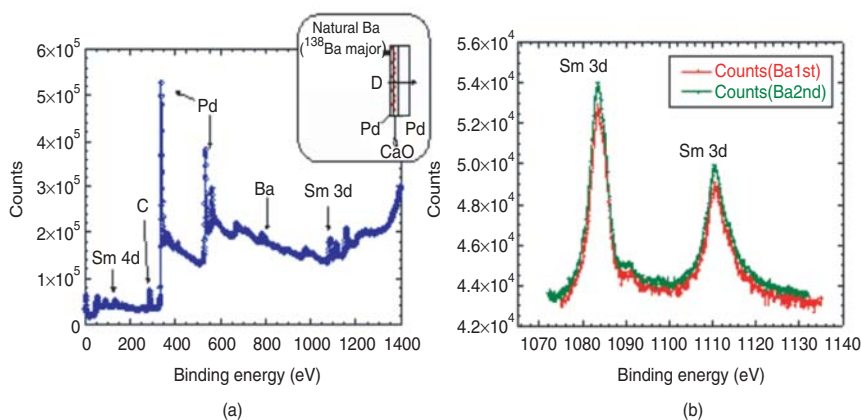


Figure 4. Detected Sm XPS spectra after D_2 gas permeation through a Pd complex deposited with natural Ba; (a) full XPS spectrum and (b) XPS spectra for Sm 3d. (Measurements were performed twice to confirm measurement reproducibility.)

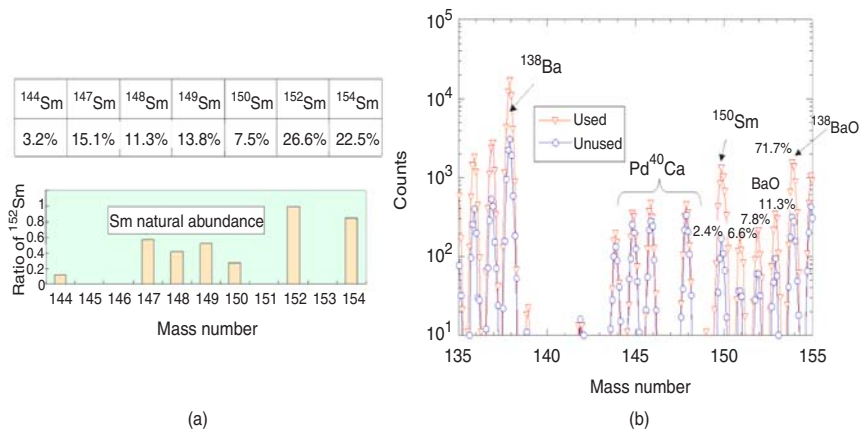


Figure 5. SIMS spectrum for detected Sm; (a) natural abundance of Sm and (b) SIMS spectra of detected Sm (“Used” indicates a Pd complex with natural Ba after D_2 permeation, and “Unused” indicates a Pd complex with natural Ba that has not been permeated with D_2).

Let us consider these spectra in Fig. 5b, using Table 1 that examines the effects of molecular ions. The ^{138}Ba signal for unused and used samples does not match.

Table 1. Examination of molecular ions.

Pd	Pd ⁴⁰ Ca	Ba	Ba ¹⁶ O
102 (1%)	142	130 (0.1%)	146
104 (11%)	144	132 (0.1%)	148
105 (22%)	145	134 (2.4%)	150
106 (27%)	146	135 (6.6%)	151
108 (26%)	148	136 (7.8%)	152
110 (12%)	150	137 (11.3%)	153
		138 (71.7%)	154

We assume this is because the Ba deposition is not uniform. Pd⁴⁰Ca molecular ion peaks are the same in the unused and used samples. Both ¹¹⁰Pd (12%)⁴⁰Ca, and ¹³⁴Ba(2.4%)¹⁶O are candidates for mass 150, however their signals should be lower than ¹⁰⁶Pd(27%)⁴⁰Ca and ¹³⁸Ba(71.7%)¹⁶O. The SIMS data shows that mass 150 for used sample cannot be explained by ¹¹⁰Pd⁴⁰Ca and ¹³⁴Ba¹⁶O.

Next, we consider the effect of the ¹²C ion. Mass 150 can be created by combining ¹³⁸Ba and ¹²C. If ¹³⁸Ba¹²C is created, then ¹³⁷Ba¹²C (mass 149) and ¹³⁶Ba¹²C (mass 148) should appear. However, we do not observe any increase of mass 149 and 148 in the used sample. This indicates that BaC molecular ions have no effect on SIMS spectra. Therefore the large mass 150 in the used sample cannot be explained by ¹³⁸Ba¹²C formation.

Based on these above SIMS considerations and XPS results, it seems clear that mass 150 in the used sample is derived from Sm. This strongly suggests that ¹⁵⁰Sm exists on the Pd complex after D₂ gas permeation.

In the case of the mass 137-enriched Ba sample, we could not obtain a clear XPS spectrum. However, we obtained SIMS data that showed an increase in mass 149. Figure 6 shows two SIMS spectra for Pd complexes with 137 enriched Ba after D₂ permeation. An increase in mass 149 of about one order of magnitude was observed for both experiments 1 and 2. Table 1 shows that mass 149 cannot be created by Pd⁴⁰Ca and Ba¹⁶O. ¹³⁷Ba¹²C also cannot be the cause of the increase, for the same reason. These facts imply that ¹⁴⁹Sm exists on the Pd complex, when we consider that Sm spectra were obtained by XPS using natural Ba. ¹⁴⁹Sm was probably detected after D₂ permeation through the Pd complex.

Figure 7 shows the mass correlation between the starting and final elements on the complex surface. If we put ¹³⁸Ba on the complex we obtain ¹⁵⁰Sm. And if we put ¹³⁷Ba on the Pd complex, we obtain ¹⁴⁹Sm, assuming the mass 149 increase of the SIMS spectra are caused by Sm. Observed transmutation reactions of Ba into Sm belong to the category of mass 12 (atomic number 6) increase reactions. Nuclear transmutation induced by our experimental method have also produced mass 8 (atomic number 4) increases, such as Cs → Pr, and Sr → Mo.

The aim of Ba transmutation experiments is to investigate the mechanism of the present transmutation phenomena by the nuclear resonance scattering. ¹⁴⁹Sm is a Mossbauer isotope and its excitation energy is 22.5 keV. If we measure the Mossbauer effect of ¹⁴⁹Sm by synchrotron orbital radiation, we will obtain clear

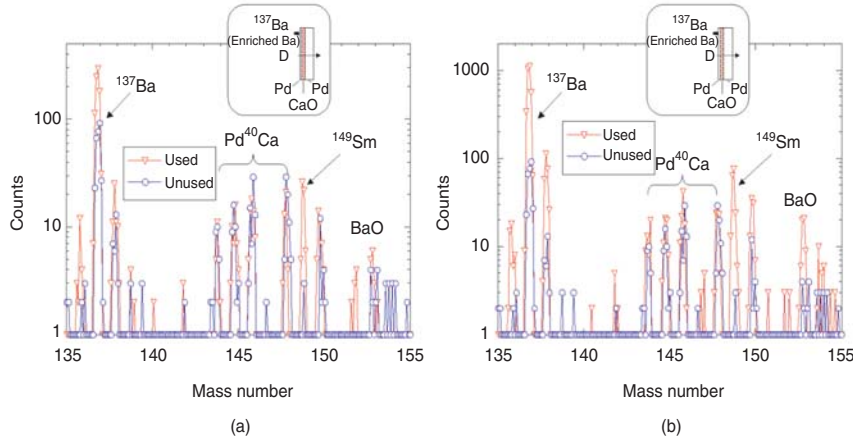


Figure 6. SIMS spectra for Pd complexes with 137 enriched Ba after D_2 gas permeation; (a) spectrum for experiment 1 and (b) spectrum for experiment 2.

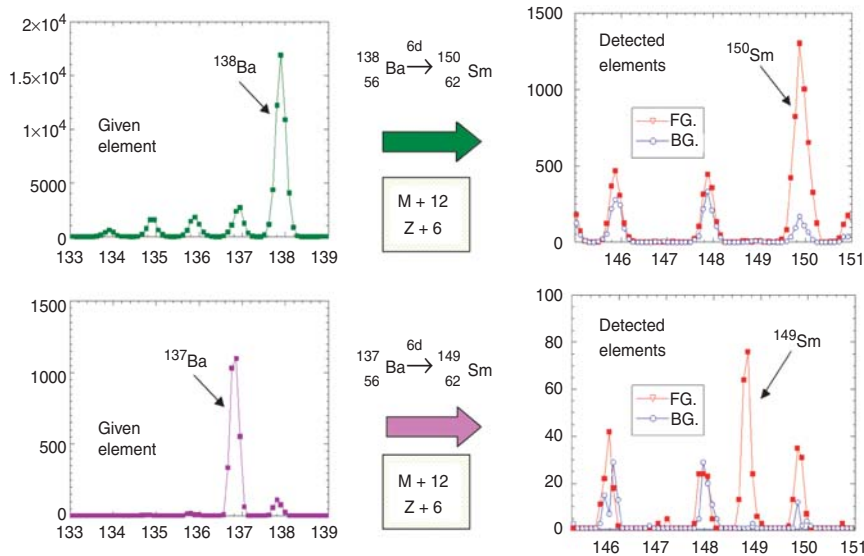


Figure 7. Mass correlation between the elements in the Unused and Used samples.

evidence of generation of ^{149}Sm and the information on the ultra fine structure relating to the electronic state and phonon of the generated ^{149}Sm . The authors are now examining and planning some experiments in collaboration with the researchers of University of Tokyo.

Let us move to the next results: Pr confirmation by XRF and experiments with in situ measurement at SPring-8. Ex situ detection of Pr by XRF using SOR X-ray at SPring-8 is shown in Fig. 8. All the permeation experiments in this figure

8

were performed in XPS apparatus. Data marked “Used 1” and “Used 2” are from samples after D_2 permeation, and “Unused” data is from a sample that did not undergo permeation. The Pr-L lines are clear in both Used samples, while no Pr peak was seen in the Unused sample. Conversion rates from Cs to Pr are high, and Cs-L lines have almost vanished in the two Used samples.

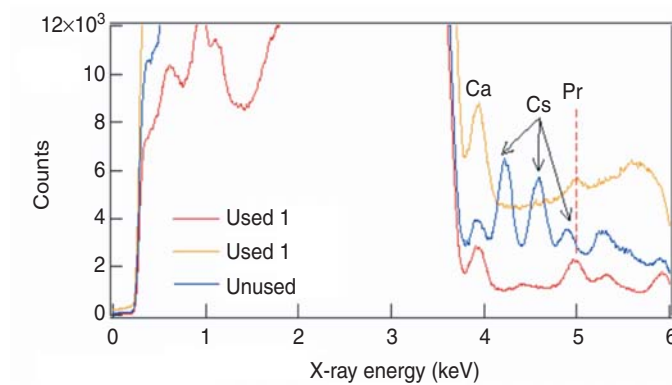


Figure 8. Ex situ detection of Pr by XRF using SOR X-ray at SPring-8, Hyogo, Japan (Used 1, Used 2: data from Samples after D_2 Permeation. Unused: data from a sample before permeation).

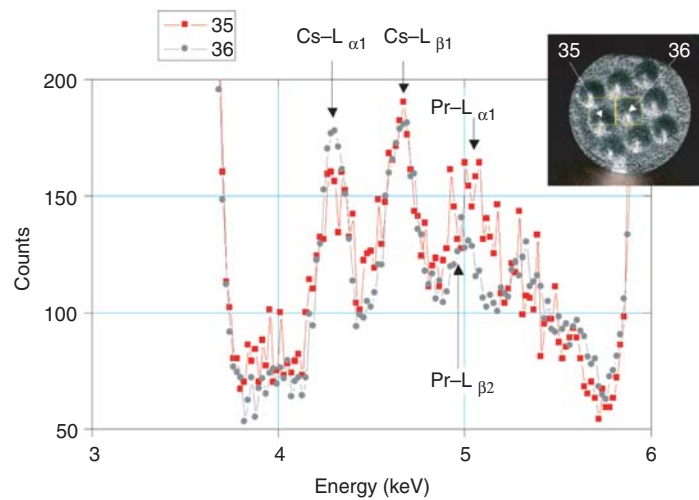


Figure 9. An example of Pr detection by in situ experiments at SPring-8.

The XRF spectrometry was performed using the experimental setup shown in Fig. 3, both during D_2 permeation *in situ* and at the beginning and end of experiments. No Pr was observed in three Used samples.

Pr was detected in three Used samples with normal complexes, whereas no Pr was observed in an experiment without a CaO layer. Figure 9 shows a sample spectrum. Surface distributions of XRF spectra were observed using a 1-mm² X-ray beam. A clear Pr spectrum can be seen at the point 35 shown in Fig. 9. Pr spectra were obtained at some locations, however, no Pr was found in location 36. This indicates uneven distribution of Pr on the complex surface.

The next topic is the role of CaO layer in Pd complex. Depth profiles of Cs and Pr were plotted in Fig. 10. Both TOF-SIMS and XPS analyzes were applied, as shown in Fig. 10a and b. Cs was injected into all the Pd complex samples by the ion implantation method. The relation between the sputtering time and the real depth was estimated in advance using a Pd thin film on Si substrate; thickness of the Pd thin film is known. These measurement shows that a 200s in the case of TOF-SIMS and a 4min sputtering time in the case of XPS correspond to 10nm. Figure 10a was already shown at ICCF10.

Cs and Pr depth profiles for the Pd complex without permeation show normal results in Fig. 10. Cs decreases continuously from the surface and there is no Pr in the background samples. On the other hand, the depth profile estimated by XPS also supports that Cs transmutation reaction into Pr occurs in the near surface region up to 10nm. We can see that there is Cs, which is the same order as given Cs, in the near surface area. Cs depth profiles for the foreground and background samples agree in the deep area. Figure 10 also shows that Cs atoms do not diffuse and migrate with D₂ gas permeation under our experimental conditions. Therefore, it is very difficult to imagine that the detected Pr was a concentrated impurity, and not a transmutation product.

If we could assume that Cs transmutation occurred in the near surface region up to 10nm, direct electronic effect on the region by CaO layer in 40nm depth might be difficult.

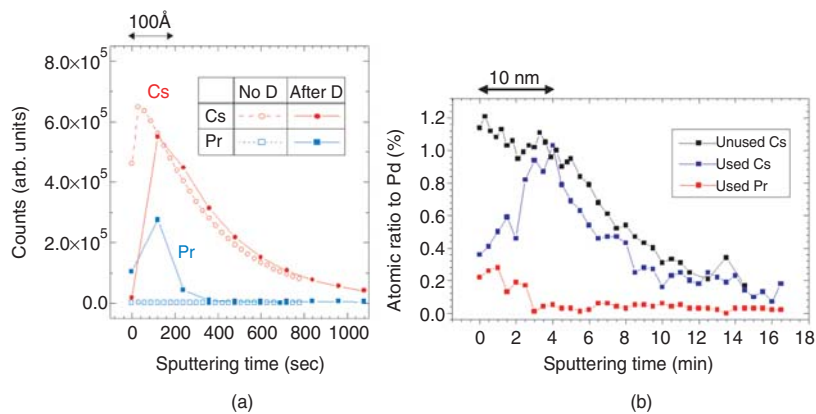


Figure 10. Depth profile of Cs and Pr for samples after D₂ permeation and without permeation; (a) based on TOF-SIMS measurement and (b) based on XPS measurement.

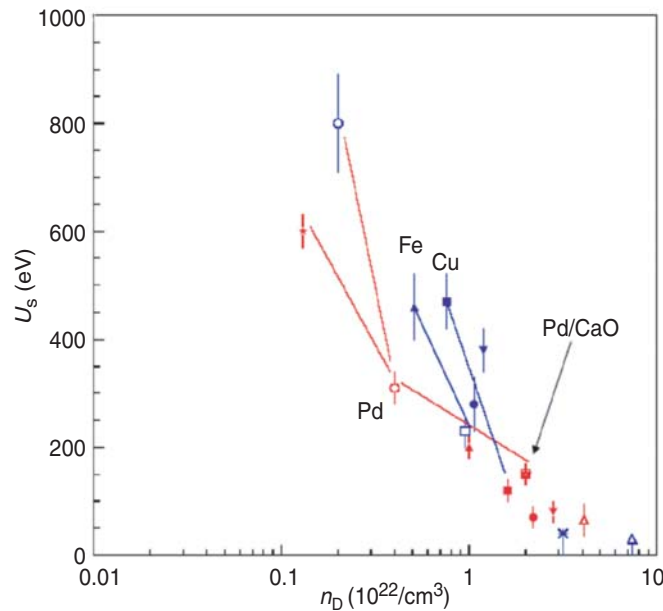


Figure 11. Density measured by D^+ ion bombardment experiment at Tohoku University.

D^+ ion bombardment experiment was performed at Tohoku University using a Pd complex. Low-energy D^+ ion beam from 2.5 to 10 keV irradiates the surface of metal and induce $D(d,p)T$ reactions. Proton yield obtained $\Delta E-E$ counter telescope and its energy dependence enable us to estimate screening potential and deuterium density.⁵ Figure 11 shows that deuterium density of Pd complex(Pd/CaO) is one order larger than normal Pd.

When we replaced CaO with MgO, we did not obtain any positive results. It means that MgO cannot work instead of CaO. Two cases out of two experiments using MgO show no Pr by ICP-MS measurements, although D_2 gas flow rates were enough (2–3 sccm) in both cases. On the other hand, almost every time Pr was detected if we use Pd complex with CaO.

At present the authors do not have definite explanation for the role of the CaO layers. We cannot perfectly exclude out the possibility that CaO layers modified the electronic state of top Pd layer. However, it could be possible to consider that the CaO layers cause the increase of deuterium density according to the result of the bombardment experiment. Anyway, we should make clear how CaO layers work in further studies.

A resonance nuclear reaction would give us precise depth profile deuterium near surface.⁶ Hydrogen depth profiles have already been established and measuring deuterium depth profile technique should be developed. We are planning with the researchers of University of Tokyo to utilize the ^7Li and deuterium reaction.

There is no complete theory that can explain the experimental results without any assumptions, however, some interesting models and theories have been proposed.⁷⁻⁹ The observed transmutation processes must belong to a new category of nuclear reactions in condensed matter. Therefore much more theoretical investigation is necessary.

4. Concluding Remarks

Transmutations of Ba into Sm were observed natural Ba as the starting material on Pd complex samples, and possibly with mass-137 enriched Ba. This indicates that we obtained a mass distribution of Sm depending on the starting isotopic distribution of Ba.

One of our experimental apparatuses was carried to SPring-8 cyclotron facility, where it was used for in situ measurements, and where we obtained some Pr signals by the XRF method.

According to a D^+ ion beam bombardment experiment performed at Tohoku University, deuterium density of our Pd complex indicated one order larger than normal Pd.

Positive replication results were obtained not only in a gaseous environment¹⁰ presented by Prof. A. Takahashi *et al.*, but also in an electrochemical environment¹¹ performed Dr. F. Celani's team. Researchers at the Naval Research Laboratory are now planning a replication of the experiments that produced transmutations of Cs into Pr.

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