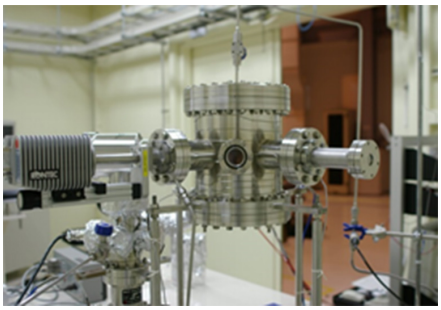
Transmutation Reaction Induced by Deuterium Permeation Through Nanostructured Multi-layer Thin Film



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The new method of nuclear transmutation is a simple method of nuclear transmutation that uses Mitsubishi Heavy Industries, Ltd.'s (MHI) original nanostructure multi-layer reactional film (hereinafter, reactional film) to transmute elements at low energy cost. So far, transmutation from cesium (Cs) to praseodymium (Pr), from barium (Ba) to samarium (Sm), from strontium (Sr) to molybdenum (Mo), etc., has been observed. If this technology is established, it is expected to contribute to society in the field of detoxification treatment of radioactive waste including the transmutation of radioactive cesium into a harmless nonradioactive element in the future.

1. Introduction

In the previous article[(1)], we reported that a new phenomenon was discovered that when a certain element is added to an original reactional film composed of palladium (Pd) and calcium oxide (CaO) and then deuterium (D_2) gas permeates the complex, the added element transmutes into a different element. Since then, MHI has been obtaining experimental data which support the existence of this phenomenon and increasing the yield obtained by the transmutation reaction. At the same time, several experiments to reproduce the experimental results using MHI's method were conducted mainly by domestic research organizations, and the transmutation from Cs to Pr has been confirmed[(1)(2)]. Recently, Toyota Central R&D Labs., Inc. also reported success in the reproduction of the phenomenon using MHI's method[(3)]. This phenomenon has been increasingly attracting attention every year.

Normally, large-scale equipment such as a nuclear reactor or an accelerator was needed to induce nuclear transmutation. Accordingly, a method of nuclear transmutation simply by inducing D_2 gas permeation is expected to prove to be a great technical and social contribution. This article describes an outline of this phenomenon and the latest research results.

2. Nuclear transmutation by D_2 gas permeation

MHI has observed the new phenomenon of nuclear transmutation by the following simple method. As shown in **Figure 1**, on a reactional film with a nanostructure where Pd and CaO are alternately laminated, the element (Cs, Ba, for example) to be transmuted is added, the side of the reactional film on which the element was added is filled with D_2 gas, the other side is evacuated with a pump, thereby permeating the reactional film with D_2 gas, so that the elements added on the reactional film decrease with the passing of time, and elements that did not exist before are detected.

The reactional film required for nuclear transmutation is prepared by the following procedure (**Figure 2**). Pd substrate (25 mm x 25 mm x 0.1 mm) is subjected to ultrasonic washing with acetone. After that, it is heated at 900°C under vacuum ($<10^{-5}$Pa), for 10 hours. The Pd substrate is cooled to room temperature, and it is subjected to etching by aqua regia (an orange-red liquid

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obtained by mixing concentrated hydrochloric acid and concentrated nitric acid in a volume ratio of 3:1) to remove impurities. Then, by the sputtering method, CaO (2 nm thick) and Pd (20 nm thick) are alternately deposited in 5 layers. (However, the Pd layer of the outermost surface is 40 nm thick). Next, by electrodeposition or ion implantation, the element to be transmuted (Cs, for example) is added on the reactional film.

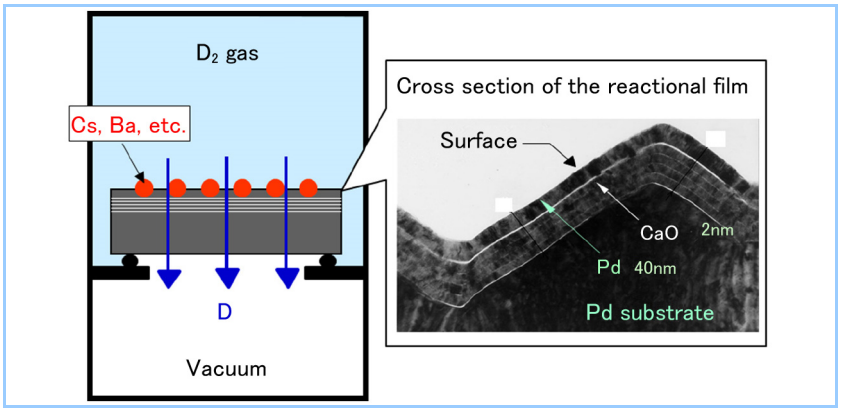

Figure 1 Method of transmutation by D_2 gas permeation

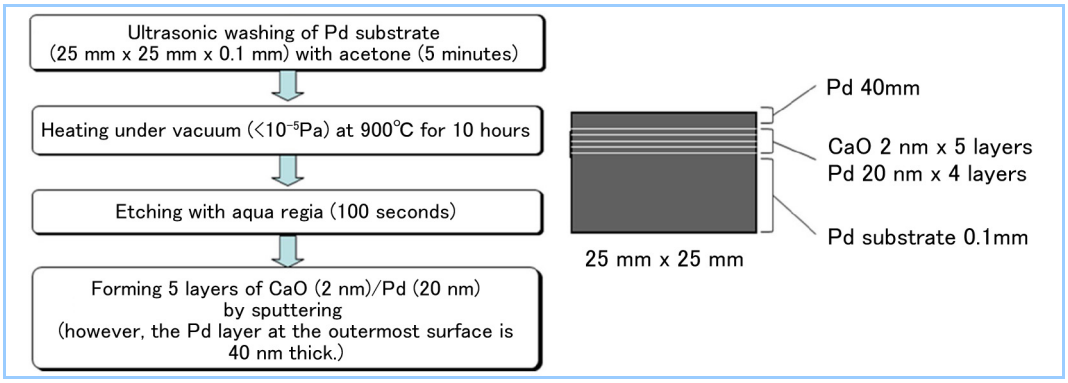

Figure 2 Preparation of the reactional film

When the prepared reactional film was put inside a vacuum chamber and permeated with D_2 gas for about 100 hours, it was observed using X-ray Photoelectron Spectrometry (XPS) placed in the chamber such that Cs was decreased with the passing of time and Pr was increased in turn. As a cross check, the existence of Pr has also been observed by Time of Flight Secondary Ion Mass Spectrometry (TOF-SIMS) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). (For the details of these analysis results, see the previous reports[(1)(5)].)

In this way, by using the method of producing a reaction within a vacuum chamber using D_2 gas (hereinafter, the gas permeation method), impurities from outside of the reaction system can be substantially reduced and products from the reaction can be analyzed with high accuracy. Accordingly, using this gas permeation method, we conducted research mainly about the clarification of the transmutation phenomenon. As an example, the results of the in-situ measurement experiment of the transmutation reaction conducted at the large synchrotron radiation facility SPring-8 (RIKEN) are presented. In this experiment, in order to obtain data which supports transformation from Cs to Pr, the synchrotron radiation of SPring-8 is used to conduct an in-situ measurement experiment of the transmutation process by X-ray fluorescence (XRF). **Figure 3** is a schematic diagram of the test device. X-rays (5.97 keV) by synchrotron radiation are introduced into the reaction chamber through Be window and irradiated to the reactional film. The intensity of the radiated X-rays are 10^{12}-10^{13} photons/sec, and Cs-L line and Pr-L line were determined using a detector. With this silicon drift detector, the in situ measurement was conducted by XRF from the beginning to the end of the D_2 gas permeation. XRF spectra obtained before and after D_2 gas permeation are shown in **Figure 4**. The measurement was implemented in the area shown in the figure. By D_2 gas permeation, the Cs peaks decreased and the Pr peaks increased. Transmutation from Cs to Pr was successfully measured in situ. However, the increase rate of Cs and the decrease rate of Pr are distributed on the reactional film surface, and it is estimated that the transmutation phenomenon occurs at certain local sections on the reactional film.

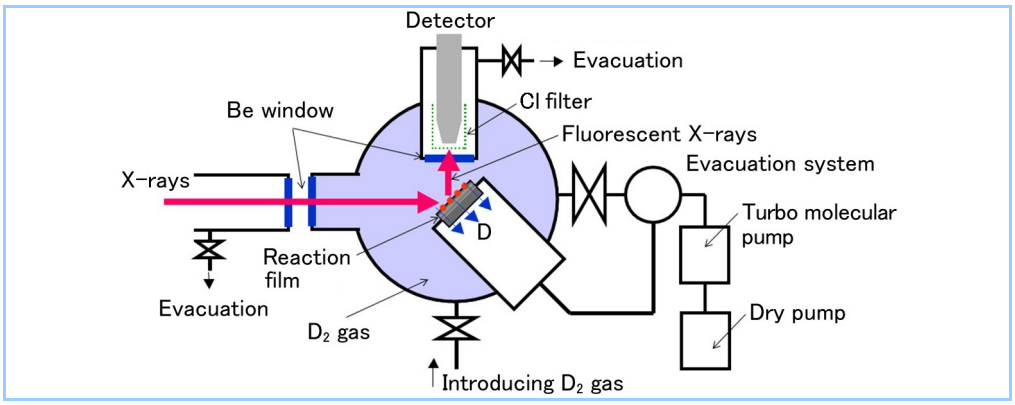

Figure 3 Experimental device used for in situ measurement in SPring-8

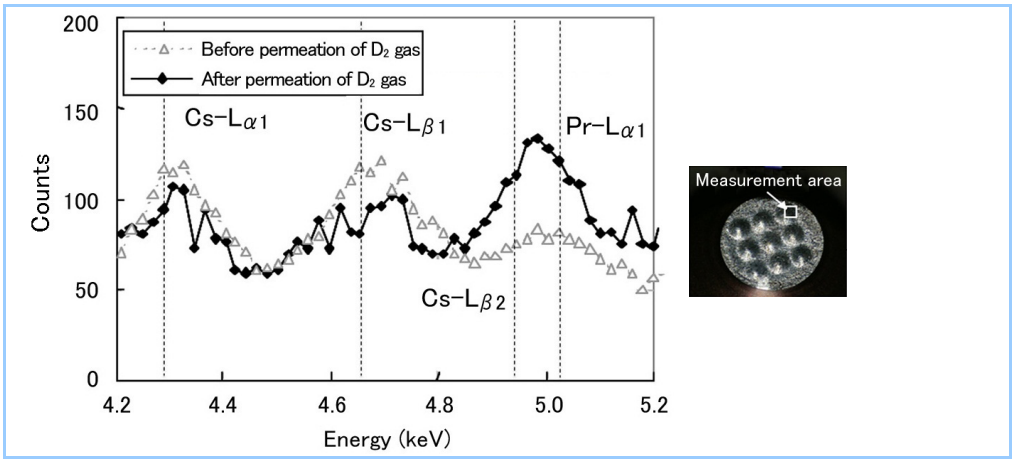

Figure 4 Results of the in situ measurement for transmutation from Cs to Pr by XRF

3. Characteristics of transmutation phenomenon

Next, in order to clarify where the reaction occurs on the reactional film, an analysis in the depth direction was carried out by XPS and the results are shown in **Figure 5**. Cs is added to the surface of the reactional film by ion implantation. On the reactional film before D_2 gas is permeated, Cs gradually decreases in the depth direction from the surface, and Pr is not detected, which is not shown in the figure. On the other hand, after D_2 gas permeation, Cs decreases up to the point of 10 nm from the surface, and Pr is detected in turn. Therefore, it is considered that under the present experimental conditions, the transmutation phenomenon occurs in the area of about 10 nm or less under the surface.

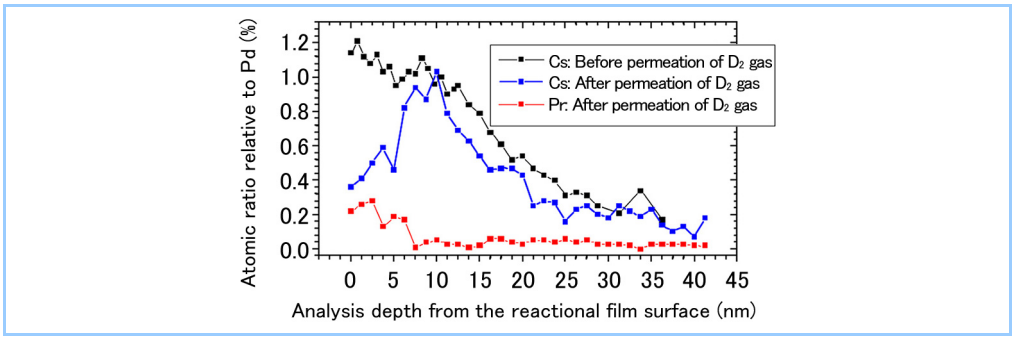

Figure 5 Results of analysis of Cs and Pr in the depth direction by XPS
The analysis depth of 0 nm from the reactional film surface corresponds to the reactional film surface.

In the transmutation experiments using an element other than Cs, the phenomenon which is assumed to be transmutation from Ba to Sm and from Sr to Mo shown in **Table 1** has been observed. It is estimated that 6 deuterium is involved in the reaction of transmutation from Ba to Sm and 4 D in the transmutation from Sr to Mo. In many cases, the transmutation has been observed in elements with a low electronegativity such as alkali metal and alkali earth metal, and there is a possibility that chemical properties affect this phenomenon.

Table 1 Example of transmutation phenomenon observed

Element	Assumed reaction					
Cs	$^{133}_{55}Cs$	→	$^{141}_{59}Pr$			
Ba	$^{137}_{56}Ba$	→	$^{149}_{62}Sm$	$^{138}_{56}Ba$	→	$^{150}_{62}Sm$
Sr	$^{88}_{38}Sr$	→	$^{96}_{42}Mo$			

4. Transmutation by electrochemical method

As described above, we have conducted research with the emphasis placed on obtaining experimental data to explicate the transmutation phenomenon by the gas permeation method. In recent years, we have been conducting research for the purpose of increasing the reaction yield with an eye toward commercialization. From the research results so far, it is assumed that the deuterium density near the reactional film surface is one of the key parameters in the increase of the reaction yield. Based on this assumption, we used the method for increasing the deuterium density near the reactional film surface by electrolysis (electrochemical method) to increase the reaction yield. **Figure 6** shows a comparison between the conventional gas permeation method and the electrochemical method. By applying a voltage exceeding the threshold value at which deuterium oxide (D_2O) is decomposed between the cathode, which is the reactional film, and the opposing anode (platinum), D_2O is electrolyzed to generate D, which permeated the reactional film. After electrolysis was continued for about 100 hours, the products were analyzed by XPS or ICP-MS. The reactional film used in the electrochemical method is the same as that used in the conventional gas permeation method. **Figure 7** shows the XPS analysis results of the reactional film surfaces before and after electrolysis. Before electrolysis, the peak was not observed, but after electrolysis, the peak which is assumed to be Pr was observed. As is the case with the gas permeation method, it is assumed that transmutation from Cs to Pr occurs in the electrochemical method as well.

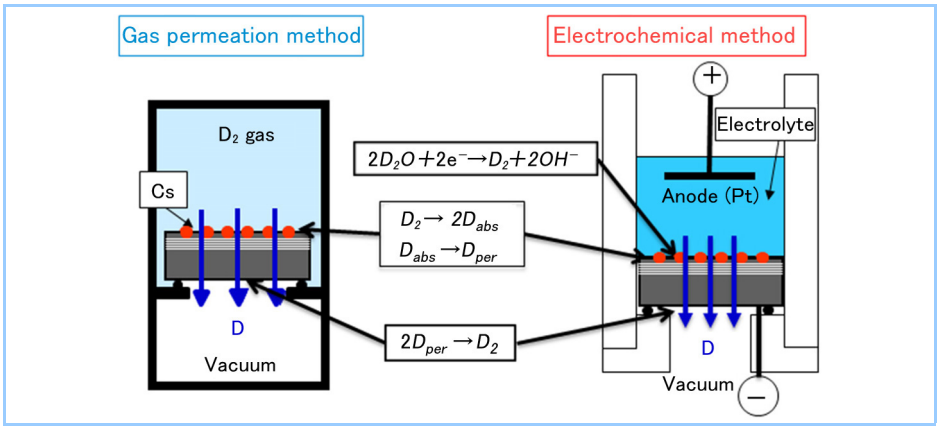

Figure 6 Comparison between the conventional gas permeation method and the electrochemical method
The solution obtained by dissolving cesium nitrate ($CsNO_3$) in D_2O was used as an electrolyte.

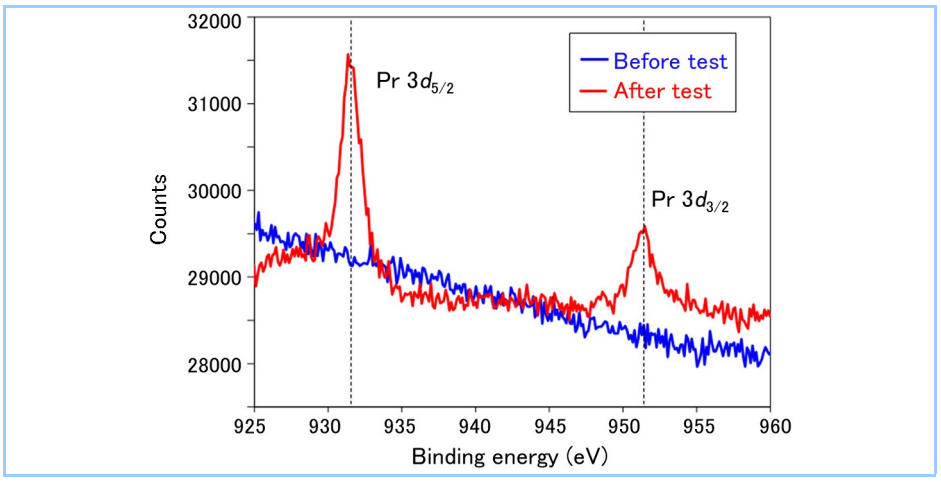

Figure 7 Results of analysis of the reactional film by XPS when the electrochemical method is introduced

In the electrochemical method, the increase of the current density during electrolysis enables the increase of the amount of D generated on the reactional film surface. **Figure 8** shows the results of the reaction yield (yield per 1 cm^2 of reactional film and 1 week of reaction time) obtained by ICP-MS plotted relative to the current density during electrolysis. As the current density increases, the reaction yield tends to increase. In the conventional gas permeation method, the reaction yield was approximately 0.01 μg/cm^2/week. In this electrochemical method, the reaction yield increased by two digits at most[6]. Under the present situation, however, the reaction yield largely fluctuates under experimental conditions. The control of reaction stability is a future issue.

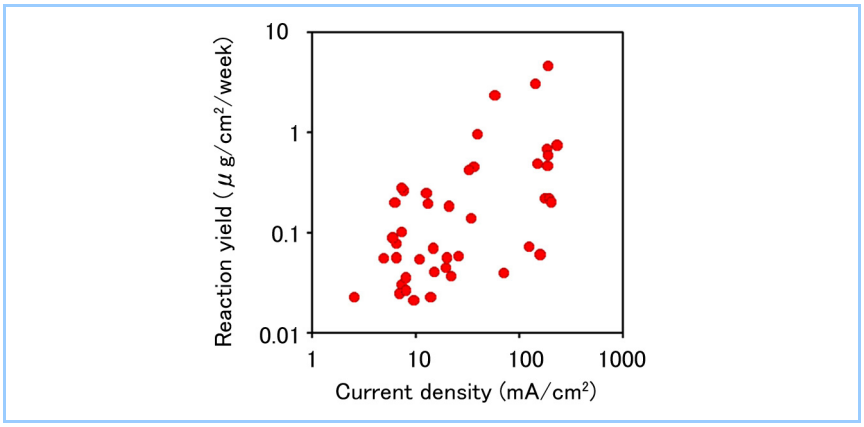

Figure 8 Relationship between the current density and the reaction yield

5. Conclusion

As described above, although this technology has been developing toward commercialization with the increase in the reaction yield, the essential nature of this phenomenon including the principle of the reaction has yet to be clarified. There is a possibility, however, that this technology could become an innovative technology that could be developed for radioactive waste treatment, and we will promote research toward commercialization while continuing to make efforts to elucidate the phenomenon.

We are grateful to Mr. Iwamura, professor by special appointment at Tohoku University and Mr. Ito, visiting associate professor, who have made a significant contribution in the promotion of this research since joining MHI.

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