



Research Article

Increase of Reaction Products in Deuterium Permeation-induced Transmutation

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Abstract

Low-energy nuclear transmutations have been observed in the nano-sized Pd complexes, which are composed of Pd and CaO thin film and Pd substrate, induced by D₂ gas permeation. In order to increase the transmutation products, an electrochemical method was applied to increase deuterium density near the surface of the nano-structured Pd multilayer film. Transmutation products were successfully increased by this approach. Laser irradiation method was also applied to make surface Plasmon on the Pd multilayer, however, the effect of laser irradiation was not so prominent.

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Keywords: Deuterium permeation-induced transmutation, Increase of reaction products, Nano-structured Pd multilayer thin film, Transmutation

1. Introduction

Transmutation reactions in nano-structured material have been observed in nano-structured Pd multilayer thin film which is composed of Pd and CaO thin film and Pd substrate, induced by D₂ gas permeation [1–5]. Experimental data that indicates the presence of transmutation have been accumulated and experimental conditions for inducing low-energy transmutation reactions are gradually becoming clear, although systematic experimental study is still insufficient. Replication experiments have been performed by some researchers and similar results have been obtained [6–8]. Potential applications would be expected as an innovative nuclear transmutation method of radioactive waste and a new energy source.

Figure 1 shows schematic of our experimental method. Our approach can be characterized by the permeation of D₂ gas through the nano-structured Pd complex and the addition of an element that is specifically targeted to be transmuted. Permeation of deuterium is attained by exposing one side of the Pd multilayer thin film to D₂ gas while maintaining the other side under vacuum conditions. The surface of the plate was covered by layers of CaO and Pd, which were obtained by five times alternately sputtering 2-nm-thick CaO and 20-nm-thick Pd layers. Then a 40-nm-thick Pd layer was sputtered on the surface of the CaO and Pd layers. These processes were performed by the Ar ion beam sputtering

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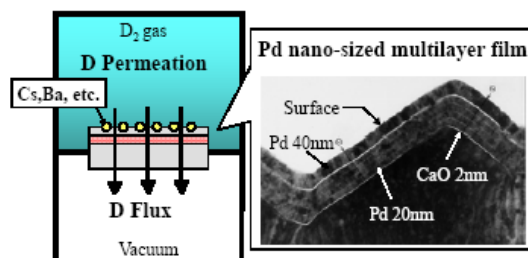


Figure 1. Experimental method for permeation-induced nuclear transmutation.

method. After fabricating a Pd complex, Cs was deposited on the surface of the thin Pd layer. After fabricating a Pd complex, Cs, Ba, or other element is deposited on the surface of the top thin Pd layer. The added elements can be transmuted.

Reactions observed so far in our group are shown in Table 1. Based on these experimental results, alkali elements seem to be transmutable by our method. In other words, chemically active elements that can easily emit electrons might be transmutable. And the obtained experimental results so far suggest that a certain rule seems to exist. We can notice that 2d, 4d or 6d look like reacting with deposited elements. Multi-body reactions like 2d, 4d and 6d require sufficient number of d. Therefore, we can see that sufficient deuterium density would be important to induce transmutation reactions.

Table 2 shows correlation between intermediate material in Pd multilayer film and transmutation results. If we replaced CaO with MgO, we did not obtain any positive transmutation products; we could not observe any transmutation reactions. It means that MgO cannot work instead of CaO. Three cases out of the three experiments using MgO show no Pr by ICP-MS measurements, although D₂ gas Flow rates were enough (2–3 sccm) in all cases. However, if we replaced CaO with Y₂O₃, we could observe transmutation reactions from Cs to Pr. Y₂O₃ works like CaO. Work functions for MgO, Y₂O₃ and CaO are shown in the Table 2. Although it is difficult to make conclusive results, the existence of low work function of intermediate material might have some effects to induce transmutation.

The permeation-induced transmutation technology would be expected as an innovative nuclear transmutation method for radioactive waste and a new energy source. However, it is necessary to increase the amount of transmutation products for commercialization.

The author is now assuming that the following two conditions are important to increase up transmutation products based on the experimental results.

- (1) Local deuterium density is sufficiently high.

Table 1. Typical reactions observed so far.

Elements		Assumed reactions
Cs	4d	$^{133}_{55}\text{Cs} \xrightarrow{4d(2\alpha)} ^{141}_{59}\text{Pr}$
Ba	6d	$^{138}_{56}\text{Ba} \xrightarrow{6d(3\alpha)} ^{150}_{62}\text{Sm}$, $^{137}_{56}\text{Ba} \xrightarrow{6d(3\alpha)} ^{149}_{62}\text{Sm}$
W	4d or 2d	$^{182}_{74}\text{W} \xrightarrow{4d(2\alpha)} ^{190}_{78}\text{Pt}$, $^{186}_{74}\text{W} \xrightarrow{2d(\alpha)} ^{190}_{76}\text{Os}$

Table 2. Correlation between intermediate material in Pd multilayer film and transmutation results.

Intermediate material	Work function (eV)	Results for analysis after permeation
CaO	1.2	Pr detected > 100 cases
Y ₂ O ₃	2.2	Pr detected > 10 cases
MgO	3.3	No Pr (3 cases)

(2) Electron rich state is important.

According to these assumptions, we tried to increase the amount of transmutation products by the increase of deuteron density and the excitation of surface Plasmon on the Pd multilayer.

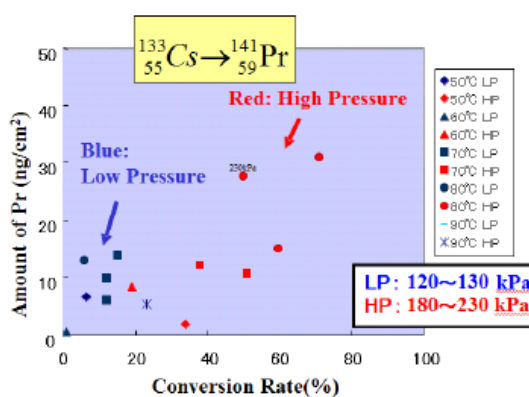
2. Increase of Deuteron Density Using an Electrochemical Method

Figure 2 shows the transmutation products dependence on D₂ gas pressure in the case of Cs transmutation into Pr. Conversion rate means how much portion of initial Cs was transmuted into Pr. Amount of Pr is expressed as the mass of Pr divided by the permeated surface area. Typical permeated surface area is about 1.0 cm².

It is possible to see that the amount of Pr and conversion rate increase as the D₂ gas pressure increase. It is reasonable that multi-body reactions observed in the permeation-induced transmutation require sufficient number of deuterium. Therefore we need high D₂ gas pressure since it gives much deuterium near the surface.

An electrochemical method is applied to increase deuterium surface density as shown in Fig. 3. A photo of our apparatus is shown also in Fig. 3. If we apply this method, we can provide very high deuterium density by controlling applied voltage between the Pd/CaO multilayer thin film and the anode made of Platinum. Simple pressurization is, off course, one of the solutions which give high deuterium density, however, it needs relatively large-scale apparatus. Therefore, we choose this electrochemical method for giving high deuterium density to the Pd surface, taking into the consideration of future commercialization.

An example of experimental results is shown in Figs. 4 and 5. A 0.1 M CsNO₃–D₂O solution was used for this experiment. Purity for both NO₃ and D₂O is more than 99.9%. Mass distributions by Secondary Ion Mass Spectrometry

**Figure 2.** Pr Dependence on D₂ gas pressure.

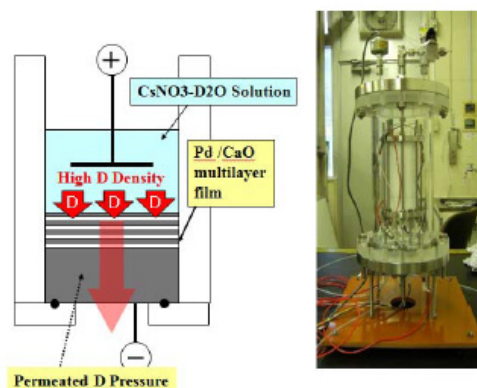


Figure 3. Electrochemical deuterium permeation apparatus aiming the increase of deuterium density.

(SIMS) are plotted in Fig. 4 and mass distributions by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for the same sample: E006 are shown in Fig. 5.

We can postulate that nuclear transmutation reactions occur at the permeated part of the Pd multilayer sample (center), however, no reactions occur at the part where no D permeated (corner). In advance, Cs ion implantation (20 kV , $10^{16} \text{ ions/cm}^2$) were applied to Pd multilayer.

SIMS counts are plotted in a logarithmic scale in Fig. 4(a). SIMS analysis was performed by O^{2+} ions and the analyzed area was circle in $60 \mu\text{m}$ diameter. Mass 133 corresponds to Cs and SIMS counts for mass 133 of E006-corner are larger than those of E006-center as shown in Fig. 4(a). It means that ^{133}Cs decreased at the center of E006. On the contrary, SIMS counts around mass 140 of E006-center are larger than those of E006-corner. It means that elements or molecular compounds that have mass number around 140 increased at the deuterium permeated point where SIMS analysis was performed. As a reference, SIMS mass distributions for no Cs implantation sample are plotted for both center and corner. The increase of SIMS counts around mass 140 cannot be seen as for the no implantation sample.

The magnified mass spectra from mass 130 to mass 150 is shown in Fig. 4(b) where SIMS counts are plotted in a linear scale. It can be seen that SIMS counts around mass 140 are greatly increased only for the E006-center.

We performed ICP-MS analysis to confirm the obtained results by SIMS analysis. Basically, the mass information obtained by SIMS is at the analyzed small point. In contrast, the information obtained by ICP-MS is from the sample surface because mass distribution was analyzed for the HNO_3 solution that contains the surface of Pd multilayer thin film. In the ICP-MS analysis, we cut a permeated Pd multilayer sample into 1/4 size and put it into 68% high purity HNO_3 solution and analyzed the HNO_3 solution that contained the surface part of it.

Figure 5 (a) shows ICP-MS results for E006 and the no Cs implantation and D permeated sample. ICP-MS counts around mass 140 of E006 are larger than those of no Cs implantation sample. It means that elements or molecular compounds that have mass number around 140 increased on the dissolved surface by HNO_3 . The magnified mass spectra from mass 130 to mass 150 are shown in Fig. 5(b) where ICP-MS counts are plotted in a linear scale. It would be possible to say that Figs. 4 and 5 are similar around mass 140, although the difference of signals between E006 and no Cs implantation is smaller for Fig. 5. It is reasonable because SIMS data is taken from the point where transmutation reactions were supposed to occur, however, ICP-MS results contained corner part where no D was permeated and therefore no transmutation reactions were supposed to occur.

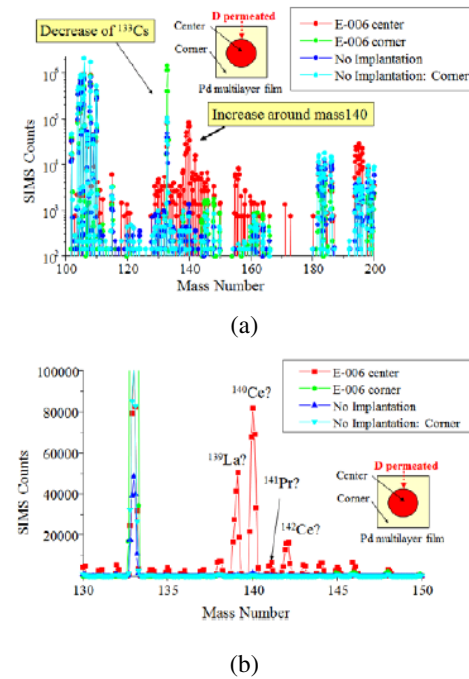


Figure 4. Comparison of SIMS mass spectra between E006 and No Cs implantation sample. (a) Wide SIMS Spectra in a logarithmic scale. (b) Magnified SIMS Spectra from mass 130 to mass 150 in a linear scale.

Next we examined the formations of compound species for mass 139 and 140 that large increases were observed. Possible compounds for mass 139 and 140 are shown in Table 3. If we carefully examine and take natural abundances for each element into consideration, mass 139 and 140 cannot be explained by these compounds. Therefore, mass 139 and 140 would be ^{139}La and ^{140}Ce , respectively. Off course, it is preferable to make cross check by the other analysis methods that have other measurement principle, for example, XRF or XPS. It is the next step work to confirm these results by the other methods.

Table 3. Possible compounds for mass 139 and 140.

Possible compounds for mass 140		
^{138}Ba (71.7%) D	^{133}Cs (100%) ^7Li (92.4%)	^{110}Pd ^{30}Si (3.1%)
^{106}Pd ^{34}Si (4.3%)	^{109}Ag (48.1%) ^{31}P (100%)	^{104}Pd ^{36}Ar (0.33%)
^{102}Pd ^{38}Ar (0.06%)	^{110}Pd ^{28}Si (92.3%) D	^{108}Pd ^{30}Si (3.1%) D
^{105}Pd ^{33}Si (0.8%) D	^{102}Pd ^{36}Si (0.02%) D	^{102}Pd ^{36}Ar (0.3%) D
Possible compounds for mass 139		
^{137}Ba (11.2%) D	^{133}Cs (100%) ^6Li (7.6%)	^{110}Pd ^{29}Si (4.7%)
^{106}Pd ^{33}Si (0.8%)	^{104}Pd ^{35}Cl (75.8%)	^{102}Pd ^{37}Cl (24.2%)
^{110}Pd ^{27}Al (100%) D	^{106}Pd ^{31}P (100%) D	^{105}Pd ^{32}S (94.9%) D
^{104}Pd ^{33}Si (0.8%) D	^{105}Pd ^{32}Si (94.9%) D	^{102}Pd ^{35}Cl (75.8%) D

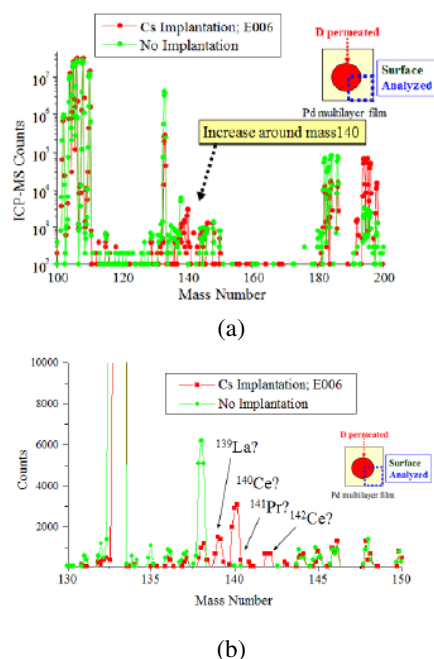


Figure 5. Comparison of ICP-MS Mass Spectra between E006 and No Cs Implantation sample. (a) Wide ICP-MS Spectra in a logarithmic scale. (b) Magnified SIMS Spectra from mass 130 to mass 150 in a linear scale.

According to our experimental results using D_2 gas permeation, we usually observed transmutation of Cs into Pr, in other words, mass 133 decreased and mass 141 increased. In this new apparatus, however, mass 139, 140, 141 and 142 increased while mass 133 decreased. The differences between D_2 gas permeation and electrochemical D permeation can be attributed to the effective deuterium density and/or source of deuterium.

Let us consider about contamination due to the $\text{CsNO}_3\text{--D}_2\text{O}$ solution. If we analyzed the $\text{CsNO}_3\text{--D}_2\text{O}$ solution, we could not see any mass numbers of 139, 140, 141, 142 (La, Ce, Pr). Furthermore, we make comparison between E006 and the D permeated sample. The D permeated sample was contacted with $\text{CsNO}_3\text{--D}_2\text{O}$ solution and almost the same current was applied. The other experimental conditions except Cs implantation were all the same. However, the only permeated part of E006 has different mass distribution as shown in Figs. 4 and 5. So it is very difficult to assume that obtained mass difference between D_2 gas permeation and electrochemical D permeation are derived from the contamination of $\text{CsNO}_3\text{--D}_2\text{O}$ solution. Therefore, we might consider the effective deuterium density make effects on the transmutation reaction pass. Anyway, we should make more experiments using the other measurement methods and improve the reliability of our experiments.

Let us move onto the next point. We describe an example of gamma-ray detection during an electrochemical D permeation experiment. In Fig. 6, applied voltage and corresponding current between Pd multilayer thin film and Pt anode, pressure at the exit side of Pd multilayer cathode are plotted as a function of time. Deuterium permeation rate is proportional to the pressure described here. At the beginning of the experiment, applied voltage was 3 V and the current was low. If we increased the voltage up to 4 V, the current increased and permeated D increased correspondingly, as shown in Fig. 6. If we increased the voltage up to 4.5 V, current amounted to 70 mA and deuterium permeation

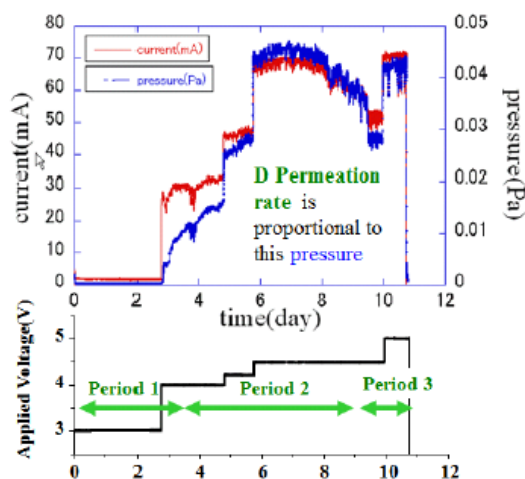


Figure 6. Time variation of applied voltage, current and pressure during an experiment E16.

increased, however, the current and D permeation decreased gradually even though we did not change the applied voltage.

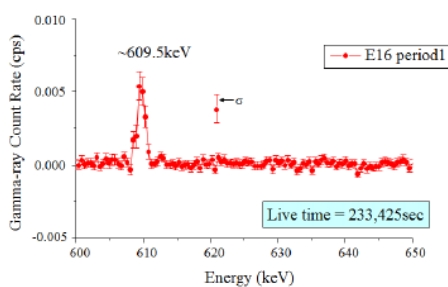
In this experiment, Pd/Y₂O₃/Pd multilayer thin film with Cs implantation and 0.5 M CsNO₃–D₂O solution were used.

Gamma-ray measurements using a Germanium detector were performed during electrochemical D permeation experiments. In this experiment (E16), gamma-ray energy spectra were obtained for three periods shown in Fig. 6. We deduced background energy spectra from obtained gamma-ray spectra during periods 1–3.

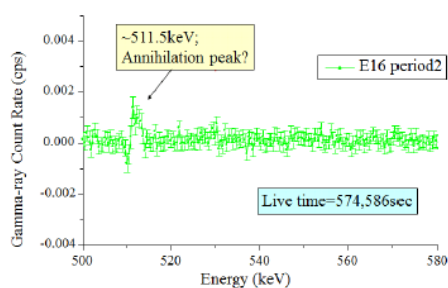
Emitted gamma-ray spectra are plotted in Fig. 7 (a)–(c), which correspond to periods 1–3, respectively. Standard deviations are also plotted for each point in all spectra. During period 1, very low current and low permeation rate, a clear gamma-ray peak around 609 keV exceeding statistic error was detected. During periods 2 and 3, relatively high current and large amount of D permeation, the 609 keV gamma-ray vanished and a peak around 511 keV was detected. There was no 511 keV peak during period 1.

Gamma-rays were not always detected. Energy spectra, in most cases, were exactly the same as background spectra, which were taken several times using the same set-up without D permeation. In the case of experiment E16, as well as a few experiments, gamma-rays exceeding statistical errors were detected. Ge detector and experimental apparatus were fixed during the experiments, off course.

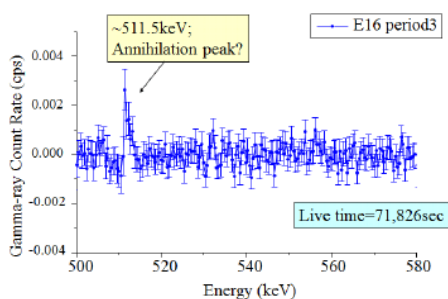
We could not observe clear gamma-ray emissions for D₂ gas permeation experiments for long time. What does it mean that we can observe weak gamma-ray emissions recently from the electrochemical D permeation? The electrochemical D permeation method gives us more transmuted products than D₂ gas permeation method. Detection of gamma-ray might be correlated to the increase of the transmuted products. The 511 keV gamma-ray is closed to annihilation energy and we might have some unstable nuclear species that emit positron. Further study is necessary to identify the source of the gamma-ray emitter. We are now planning to suppress the background gamma-rays in order to obtain better statistic results.



(a)



(b)



(c)

Figure 7. Gamma-ray emissions during an electrochemically D permeation experiment E16. (a) Gamma-ray energy spectrum emitted during period 1. (b) Gamma-ray energy spectrum emitted during period 2. (c) Gamma-ray energy spectrum emitted during period 3.

3. Effect of Laser Irradiation

As I described in Table 2, work function of the intermediate layer seems to be important. Then we assumed that electron rich state was important. In order to make electron rich state near surface of the Pd multilayer thin film, we tried to use laser stimulation method that was reported by Letts [9] to enhance excess heat generation.

If we irradiate the surface of the Pd multilayer, surface Plasmon is excited. So we would have a lot of high electron

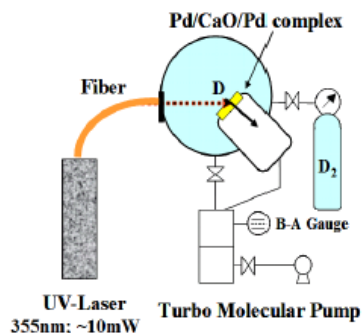


Figure 8. Schematic of the experimental set-up for plasmon excitation by laser irradiation.

density region by the Plasmon excitation. As the Plasmon has pico-second order wave and typical nuclear reaction occurs within the order of femto-second, high electron density region would be kept high during nuclear transmutation reaction proceed. Therefore it might be possible to enhance the rate of nuclear transmutation reaction by laser irradiation.

Figure 8 shows a schematic of the experimental set-up for Plasmon excitation by laser irradiation. A fiber UV-laser that has 355 nm wave length was applied. The multilayer Pd complexes are Pd/CaO/Pd and D₂ gas permeation experiments with laser irradiation were performed 3 times.

An example of experimental results is shown in Fig. 9. SIMS spectra for the permeated with laser irradiation and no permeation samples are plotted. As you can see, ¹³³Cs decreased and ¹⁴¹Pr emerged in a very similar way to the original D₂ gas permeation experiments. According to this result, we cannot find enhancement by laser irradiation.

The effect of laser irradiation on D₂ gas permeation rate is shown in Fig. 10. The laser-irradiated periods are painted with blue color. The D₂ gas permeation rate decreased when the laser was irradiated for all the cases as shown in Fig. 10. The mechanism for this phenomenon is not clear but deuterium absorption process might be influenced by

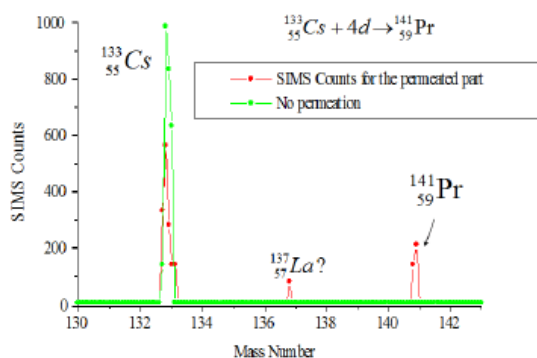


Figure 9. SIMS spectra for the permeated with laser irradiation and No permeation samples.

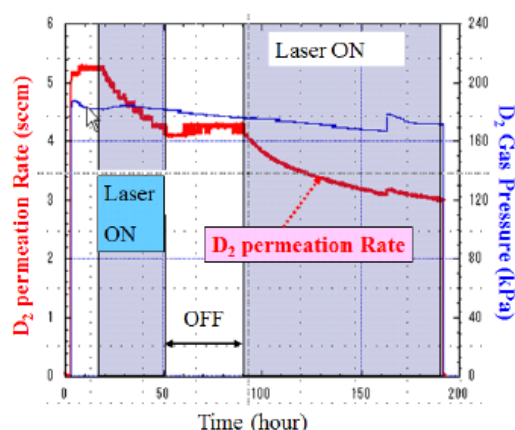


Figure 10. Effect of laser irradiation on D_2 gas permeation rate.

laser irradiation. D_2 gas permeation rate is one of the important factors to induce permeation transmutation reactions [4]; laser irradiation method is not so desirable in the view of the permeation rate.

4. Concluding Remarks

We tried two types of experimental approaches in order to increase transmuted products in the deuterium permeation induced transmutation reactions. One is the electrochemical deuterium permeation method to increase the surface deuterium density near the surface of the nano-structured Pd multilayer film. The other is the laser irradiation method for making electron rich state near the surface. The electrochemical deuterium permeation method gave us increased transmutation products, although the effect of the laser irradiation method was not clear. Many kinds of transmuted products were obtained and gamma-ray emissions were observed by the electrochemical deuterium permeation method. It is necessary to perform more experiments from different viewpoints using the other measurement methods and improve the reliability of our experiments.

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