Yamaguchi, T., et al. *Investigation of Nuclear Transmutation Using Multilayered CaO/X/Pd Samples Under Deuterium Permeation*. in *ICCF-14 International Conference on Condensed Matter Nuclear Science*. 2008. Washington, DC.

Investigation of Nuclear Transmutation Using Multilayered CaO/X/Pd Samples Under Deuterium Permeation

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Abstract

We constructed an experimental system in which accelerator analyses by PIXE, ERDA, NRA and RBS can be made *in situ* under deuterium (D) gas permeation through multilayered CaO/*X*/Pd samples to induce nuclear transmutation in the element *X*. Furthermore, to examine the effect of flow direction, we made a D gas permeation system separated from the accelerator beam-line chamber for *ex-situ* accelerator analysis. We report the results of recent experiments in this paper.

1. Introduction

It has been claimed that the forced permeation of D through Cs(Sr)-deposited multilayered Pd/$(CaO/Pd)_5$ samples induces nuclear transmutations from ^{133}Cs(^{88}Sr) to ^{141}Pr(^{96}Mo) [1]. To confirm the nuclear transmutation and to study the mechanism, we constructed an experimental system in which accelerator analyses by PIXE, ERDA, NRA and RBS can be made *in situ* under D gas permeation through the samples. Furthermore, we have done experiments using a separate D gas permeation system and an *ex-situ* accelerator analysis.

In our previous work [2], we used XPS to characterize the CaO/Sr/Pd sample. The XPS method has a high sensitivity of detection limit 7×10^{12} atoms/cm^2, but it is applicable only for the near-surface (< 3 nm) region, and is necessarily destructive when measurement of the depth distribution over tens of nm is required. Therefore we have used PIXE to measure nondestructively the time-dependent concentration of the elements in the sample *in situ*, at the cost of increasing the sensitivity limit to 1×10^{14} atoms/cm^2.

2. Experimental apparatus and procedure

The major system is an *in-situ* system shown in Fig. 1. The film was mounted on a vacuum flange with an O-ring seal, and the rear surface was exposed to D gas at a pressure of 0.1 MPa typically. The sample temperature was kept at 70°C during D permeation. We performed *in situ* analyses to characterize the sample before, during and after D permeation; we used the 3-MeV proton PIXE for elemental analysis, the 2.5-MeV ^{3}He NRA or 4-MeV ^{4}He ERDA for D distribution analysis.

The other system is a D-absorption apparatus with an *ex-situ* accelerator analysis system, shown in Fig. 2. In the analysis, we intended to make a large incident angle (85°) measurement to increase PIXE sensitivity. First, we made samples and did PIXE analysis. Next, the samples were mounted on a vacuum flange and the rear surfaces were exposed to D_2 gas at a pressure of 0.1 MPa similarly to the *in-situ* system. Finally, after D permeation we did PIXE analysis again. In *ex-situ* experiments, we skipped NRA and ERDA for D distribution analysis.

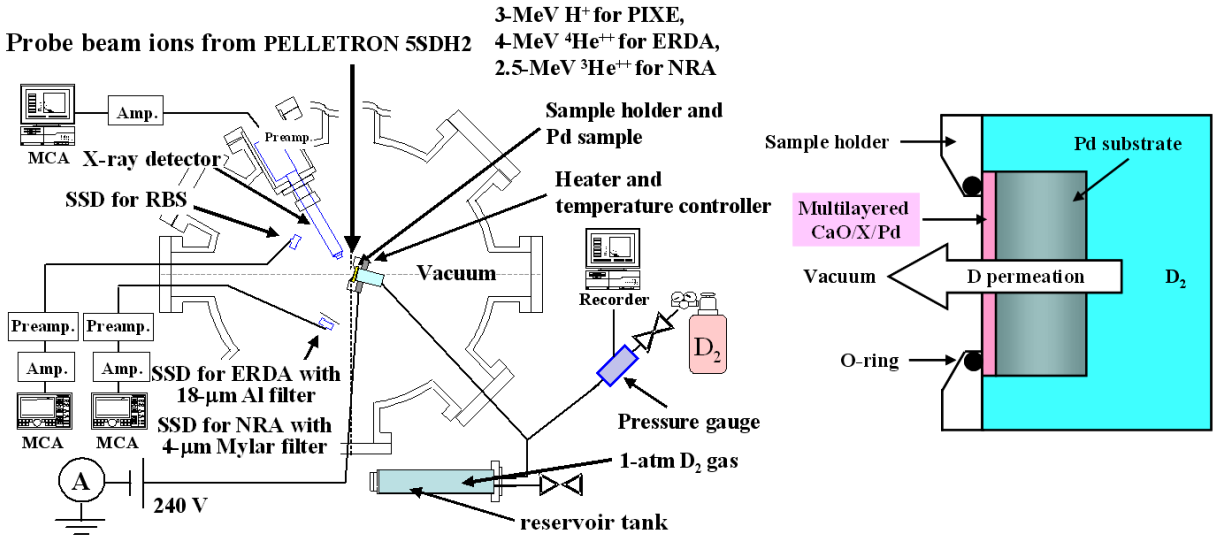

Figure 1. ***In-situ*** **system.**

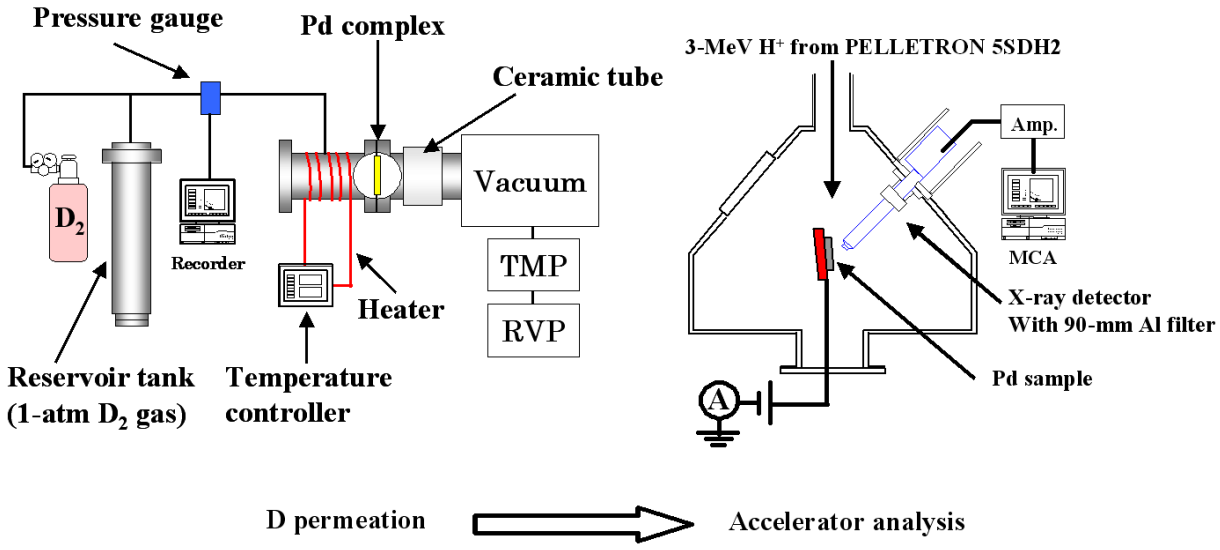

Figure 2. ***Ex-situ*** **system.**

3. *in-situ* experiments

3-1. D permeation through monolayer samples

A 0.1 mm thick Pd bulk plate was annealed for 3 to 10 hours at 570 - 1170 K, and then immersed in aqua regia/H_2O or aqua regia/D_2O for 100 seconds. Sr atoms were deposited on one side of the Pd bulk surface using electrochemical deposition, by carefully placing the Pd bulk

plate on the surface of the 10-mM $Sr(NO_3)_2/D_2O$ solution. Finally, a CaO layer with a thickness of 2 - 8 nm was deposited on the Sr/Pd samples.

The results of Run 9 are shown in Table 1. We observed some increase in molybdenum areal density. Run 9 is discussed in more detail below.

Table 1. Temporal variation of the areal densities of detected element during the permeation process in Run 9.

Run 9	**0.05 sccm**	
Vacuum/CaO/Sr/Pd/D_2		
D fluence [10^{21} cm^{-2}]	**Areal density [10^{15} cm^{-2}]**	
	Sr	**Mo**
0	**2.5 ± 0.3**	**0**
18.8	**2.3 ± 0.3**	**1.8 ± 0.8**
21.6	**2.1 ± 0.4**	**1.7 ± 1.1**
31.1	**2.1 ± 0.3**	**2.1 ± 0.9**
33.7	**2.7 ± 0.3**	**1.8 ± 0.9**
34.9	**2.3 ± 0.3**	**1.1 ± 0.9**
40.9	**1.9 ± 0.3**	**2.5 ± 0.9**
43.9	**2.4 ± 0.2**	**1.1 ± 0.7**

The PIXE spectra are shown in Fig. 3. An increase (2.5×10^{15} cm^{-2}) in Mo areal density and a slight decrease (0.6×10^{15} cm^{-2}) in Sr areal density were observed until after the 2160 hour permeation run. Typical examples of ERDA spectra obtained at 1650 hours after initiation of D permeation are shown in Fig. 4. We notice small humps at the recoil particle energy of 1.3 MeV, below which recoil hydrogen atoms (H) are detected. Depth profiles of the D and H densities deduced from them are shown in Fig. 5. The D density at 1650 hours of 3.9×10^{22} cm^{-3} averaged over the surface region up to depths of 0.5 μm corresponds to the mean composition of $PdD_{0.58}$.

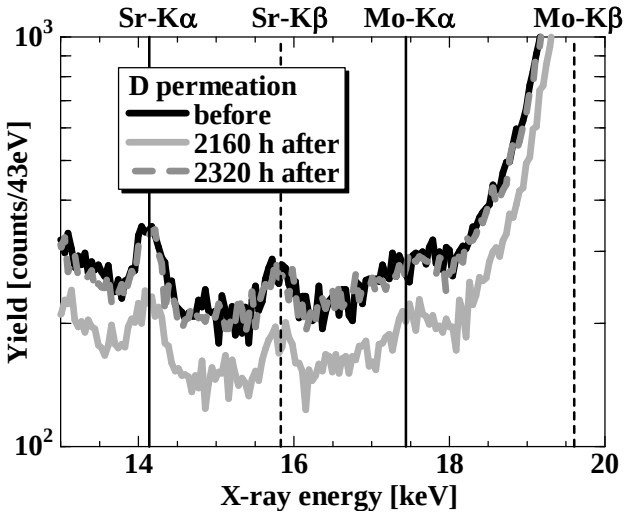

Figure 3. PIXE spectra in Run 9.

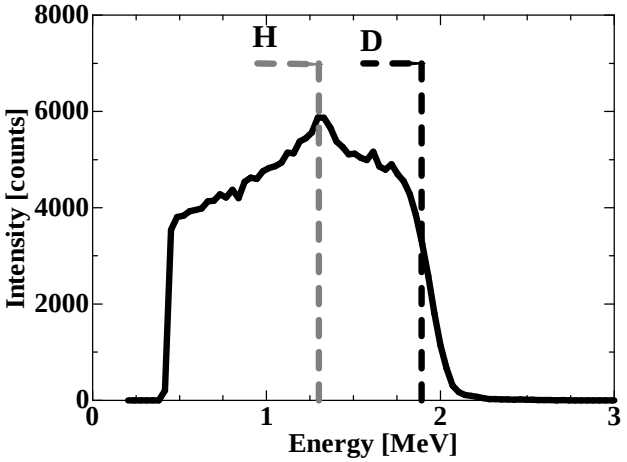

Figure 4. Typical ERDA spectrum.

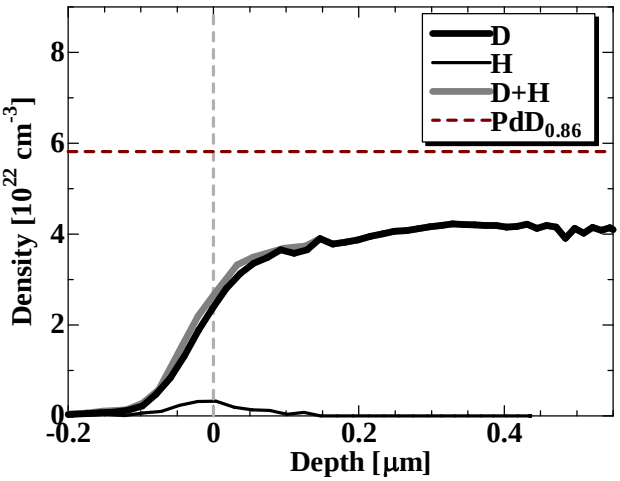

Figure 5. Depth profiles of the D and H densities in run 9.

3-2. D permeation through multilayered samples

Next, we discuss the results of Runs 4, 10 and 11 using multilayered samples prepared for the purpose of increasing the number of the atoms to be transmuted. For the sample of Run 4, a Pd layer with a thickness of 54 nm was deposited on the Sr/Pd surface by RF sputtering. We repeated the process of Sr deposition followed by Pd sputtering nine times to make a $(Sr/Pd)_9/Sr/Pd$ sample. The multilayered samples used in Run 10 and 11 were prepared by means of sputtering deposition of CaO and Pd layers with a thickness of 2 nm and 18 nm, respectively, in addition to electrochemical deposition of Sr. The results of Run 10 and 11 are summarized in Table 2. The PIXE spectra in Run 4 are shown in Fig. 6. In this Run 4, the identification of Mo was satisfactory.

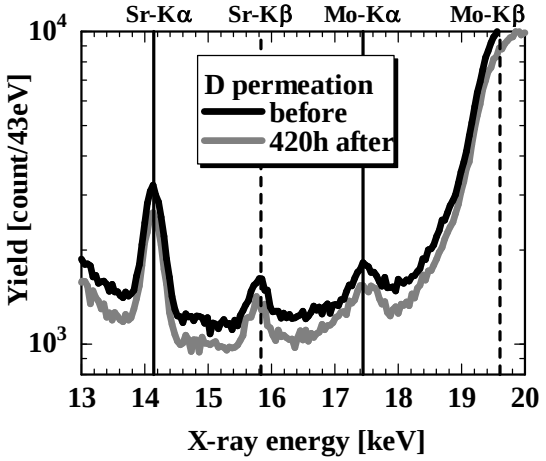

Figure 6. PIXE spectra.

Table 2. Temporal variation of the areal densities of detected element during the permeation process in Run 10 and 11.

Run 10	0.47* sccm		Run 11	0.03 sccm	
Vacuum/$(CaO/Sr/Pd)_9$/CaO/Sr/Pd/D_2			Vacuum/Pd/$(CaO/Sr/Pd)_9$/CaO/Sr/Pd/D_2		
D fluence	Areal density [10^{15} cm^{-2}]		D fluence	Areal density [10^{15} cm^{-2}]	
[10^{21} cm^{-2}]	Sr	Mo	[10^{21} cm^{-2}]	Sr	Mo
0	49.5 ± 0.4	0.2 ± 0.1	0	24.1 ± 0.4	0
9.0	50.5 ± 0.4	0.7 ± 0.3	1.9	24.3 ± 0.4	0
31.6	49.4 ± 0.5	0.3 ± 0.1	7.8	20.1 ± 0.3	0
95.0	50.1 ± 0.6	0.4 ± 0.3	10.2	21.0 ± 0.3	1.2 ± 0.5
126.0	52.0 ± 0.5	0.4 ± 0.2	13.3	21.2 ± 0.3	0.7 ± 0.4

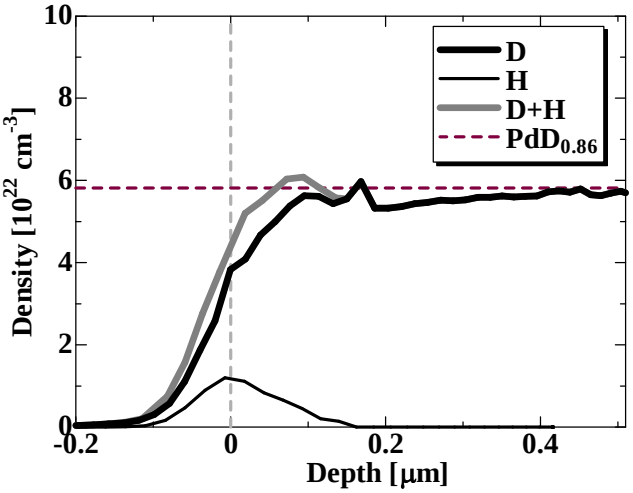

Figure 7. Depth profiles of the D and H densities in Run 10.

In Run 10, we saw hardly any change in the areal densities of Sr and Mo. The ERDA was successfully applied to reveal the D density approaching the saturation value in this sample. The depth profiles of the D and H densities deduced from them are shown in Fig. 7. The D density at 189 hours of 5.8×10^{22} cm^{-3} averaged over the surface region up to the depth of 0.5 μm corresponds to the mean composition of $PdD_{0.86}$.

Finally, in the Run 11, a slight increase in Mo areal density and a decrease in Sr areal density were observed until the end of the 971-hour permeation. The depth profiles of the D and H densities deduced from them are shown in Fig. 8. The D density at 189 hours of 1.5×10^{21} cm^{-3} averaged over the surface region up to the depth of 0.5 μm corresponds to the mean composition of $PdD_{0.02}$. The D density is very small in this sample. There is a rather large amount of contaminant H atoms.

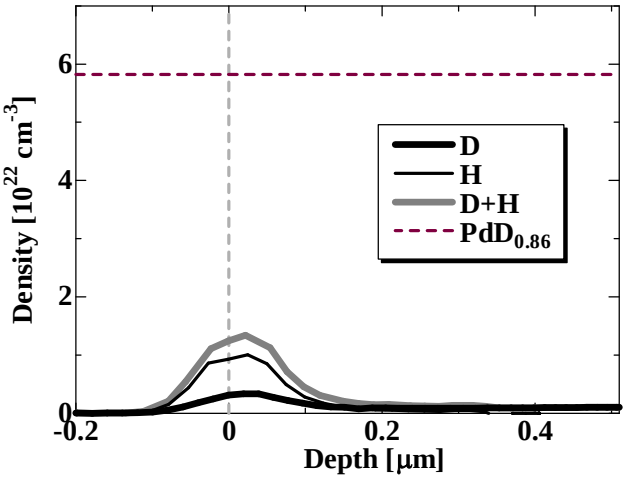

Figure 8. Depth profiles of the D and H densities in Run 11.

3-3. D permeation through multilayered samples (surface version)

We made the monolayer sample again to examine whether a nuclear transmutation was caused at the surface. At first a CaO layer with a thickness of 2 nm was deposited on the Pd substrate. And a Pd layer with a thickness of 18 nm was deposited on the CaO/Pd sample. Finally, Sr atoms were deposited on Pd/CaO/Pd sample.

The results of Run 12 are shown in Table 3. An increase (4.5×10^{14} cm^{-2}) in Mo areal density and a decrease (9.4×10^{14} cm^{-2}) in Sr areal density were observed under D fluence of 111.4×10^{21} cm^{-2} for permeation. However, a null Mo areal density was obtained in the succeeding measurement with the D fluence of 123.7×10^{21} cm^{-2}, which prevents a definite conclusion.

Table 3. Temporal variation of the areal densities of detected element during the permeation process in Run 12

Run12	0.64 sccm	
Vacuum/Sr/Pd/CaO/Pd/D_2		
D fluence [10^{21} cm^{-2}]	Areal density [10^{15} cm^{-2}]	
	Sr	Mo
0	3.5 ± 0.1	0
68.5	2.3 ± 0.1	0.2 ± 0.2
111.4	2.6 ± 0.1	0.5 ± 0.2
123.7	2.2 ± 0.1	0

3-4. Summary of the *in-situ* experiments

Results of Run 9 and Run 12 suggested occurrence of the nuclear transmutation, and Run 11 showed marginal increase in Mo, while the other runs (4, 6, 7, 8, and 10) showed no significant increases in Mo. However, identification of Mo by PIXE was not definite. We need Mo atoms with areal density larger than 3×10^{15} cm^{-2} to make a clear identification by PIXE analysis. In summary, we observed what appear to be positive results in two out of eight cases, but with weak identification.

4. *en-situ* experiments

For Run B1 and B2, we made a CaO/Sr/Pd and a Pd/CaO/Sr/Pd sample. D gas permeates from the palladium substrate side to the multilayer side. The sample used in Run B3 has the same structure as the sample for Run 12, but with a reversed flow direction of D. Results of the PIXE analysis in Run B1 through B3 are shown in Table 4. We could identify Sr with areal density as low as 7.2×10^{13} cm^{-2} by measurements with large incident angle. A decrease in Sr areal density was observed clearly. However, an increase in Mo areal density to compensate the decrease in Sr areal density was not observed.

Table 4. Temporal variation of the areal densities of detected element during the permeation process in Run B1 - 3.

Run B1	0.05 sccm		Run B2	0.07 sccm		Run B3	0.09 sccm	
Vacuum/CaO/Sr/Pd/D_2			Vacuum/CaO/Sr/Pd/D_2			Vacuum/CaO/Sr/Pd/D_2		
Areal density [10^{15} cm^{-2}]			Areal density [10^{14} cm^{-2}]			Areal density [10^{14} cm^{-2}]		
	Sr	Mo		Sr	Mo		Sr	Mo
Before	5.6 ± 0.1	0.1 ± 0.1	Before	8.1 ± 0.1	0.3 ± 0.2	Before	3.4 ± 0.2	0.3 ± 0.2
After	0.7 ± 0.04	0.2 ± 0.1	After	1.5 ± 0.1	0.4 ± 0.2	After	1.4 ± 0.1	0.2 ± 0.1

5. Concluding Remarks

An increase in Mo areal density was seen in deuterium permeation. Apparently positive results in the present transmutation experiments were obtained in about 25% of the runs. However, the identification of Mo resolved peaks was not definite, and needs to be improved.

We have at least two problems to be solved in future research: to improve the sample uniformity, and to increase the D permeation rate.

References

1. Y. Iwamura, M. Sakano and T. Itoh; Jpn. J. Appl. Phys. 41 (2002) 4642-4650.
2. A. Kitamura, et al.; Proc. ICCF12 (World Scientific Publ. Co. Pte. Ltd, 2006) 272-277.