

Search for Nuclear Reaction Products in Gas Phase Experiments – Deuterium Permeation and Absorption –

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Abstract. Employing both *in situ* and *ex situ* accelerator analyses, we have attempted to replicate the Iwamura-type nuclear transmutation of Sr to Mo under deuterium permeation through a variety of multilayered CaO/Sr/Pd samples. Apparently positive results have been obtained in 8 of 14 runs, although the identification of Mo peaks in the PIXE analysis is not definite. It is implied that sputtering loss of the atoms could be responsible for the observed tendency that the areal density of Sr decreases in most cases, while there are modest increases in Mo. In addition to the accelerator analyses, γ -ray detection has been tried for samples implanted with W atoms in expectation of transmutation from ¹⁸³W to radioactive ¹⁹¹Pt.

In another series of experiments, we examined heat and ⁴He generation by deuterium absorption in nano-sized Pd powders, as reported by Arata and Zhang. In order to determine the cause of the large isotope effects observed, nuclear ash including charged particles, neutrons and gamma rays was examined.

1. Introduction

Iwamura *et al.* claimed [1] that nuclear transmutation occurs during forced permeation of deuterium (D) through multi-layered films of X/CaO/Pd, with X being the element to be transmuted, *e.g.*, ¹³³Cs transmuted to ¹⁴¹Pr, ⁸⁸Sr to ⁹⁶Mo, etc. We have attempted to replicate the transmutation using a modified exposure system to enable *in situ* characterization of the sample, which was installed at the end of a beam line of a tandem electrostatic accelerator 5SDH2. In addition to this system with a reversed flow direction, a stand-alone D permeation system was used to examine the phenomenon by *ex situ* PIXE analysis. The results have been presented in ref. [2], and are briefly summarized in the present paper.

Arata and Zhang recently reported [3] that highly pure D₂ gas charging of Pd nano-powders in the form of Pd-ZrO₂ nano-composite induced significantly higher temperatures inside the reactor vessel than on the outside wall for more than 50 hours, while runs with H₂ gas showed almost no temperature difference. To verify that the excess heat originated in a nuclear process, a QMS was employed to show the existence of ⁴He as nuclear ash in the vessel and in the powder after the charging. The charging system is a sophisticated, yet simplified, version of the previous-generation DS reactor [4]. Replication experiments using systems similar to the DS reactor with Pd-black seem to be successful [5,6].

However, few reports on replication experiments producing heat and ⁴He with the new configuration have been published in spite of the extreme importance of the phenomenon. It is crucial to confirm the phenomenon of heat and ⁴He generation with fully quantitative reliability.

We have initiated our own replication experiments to confirm the phenomenon and to investigate the underlying physics. We have reported the first results of deuterium/hydrogen absorption and accompanying heat generation in ref. [7-11], where an anomalously large absorption energy and isotope effects were found. In the latter half of the present paper, a description of our efforts to detect possible nuclear reaction products is given.

2. Deuterium permeation experiments

2.1 Summary of PIXE analysis

We have tried to replicate the nuclear transmutation of Sr ($2 - 50 \times 10^{15} \text{ cm}^{-2}$) to Mo under deuterium permeation through a variety of multilayered samples; (v)/M_n/CaO/Sr/Pd/(D₂), (v)/Pd/M_n/CaO/Sr/Pd/(D₂), (v)/Sr/Pd/CaO/Pd/(D₂), and (v)/Pd/CaO/Sr/Pd/(D₂), where (v) stands for vacuum and “M” for a layer of CaO(18nm)/Sr/Pd(22nm). The D flow rate was 0.03 – 0.6 sccm, and the fluence was 0.1 - $2 \times 10^{23} \text{ cm}^{-2}$. The deuterium loading reached 0.86 for the sample (v)/Pd/M₉/CaO/Sr/Pd/(D₂), and the areal densities of Mo were 0 – $4.7 \times 10^{15} \text{ cm}^{-2}$. Apparently positive results were obtained in 8 of 14 runs, although the identification of Mo peaks in the PIXE analysis was not definite.

A tendency has been observed, however, for the areal density of Sr to decrease in most cases, while there are modest increases in that of Mo. Figure 1 shows the areal densities of Sr and Mo atoms as a function of time, or equivalently the deuterium fluence. In the former the density values are normalized to the initial values to show that almost all samples have decreasing density of Sr, while the absolute values are plotted in the latter. Although some samples have contaminant Mo atoms inherently, most samples show increasing density of Mo.

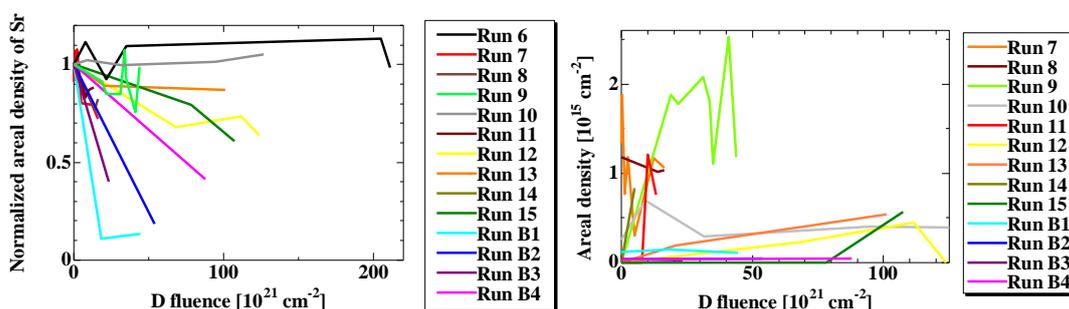


Fig. 1. - Temporal variation of areal densities of Sr (left) and Mo (right).

These results imply that sputtering loss of the atoms by 3-MeV protons and/or 4-MeV ⁴He used for PIXE and ERDA analyses could be responsible for these changes. Although a simulation program ACAT has shown that the contribution of sputtering to the decrease would be 2 orders of magnitude smaller than the decrease observed, the sputtering yield for deposited atoms weakly bonded to the bulk atoms should be much larger than the bulk atoms. This could account for the decrease in areal densities of Sr and Mo, which could result in the apparently smaller transmutation yield.

2.2 Radioactivity measurement

If the transmutation product is a radioisotope, detection will be much easier. If we assume the regularity of transformation that the atomic number and the mass number increase by 4 and 8, respectively, we can expect, for example, a production of ¹⁹¹Pt (emitting 0.538-MeV γ rays with a half life of 2.86 d) from ¹⁸³W. We prepared samples with a structure (V)/Pd/CaO/W/Pd/(D₂). The thickness of the first Pd layer and the second CaO layer were 38 nm and 2 nm, respectively. The tungsten atoms were introduced into the samples by implantation with 380-keV W⁺⁺ ions accelerated by the tandem Pelletron 5SDH2, and were expected to be distributed at a depth of 40 nm with a range straggling of 16 nm.

Gamma ray measurements were performed with an NaI(Tl) scintillation probe during permeation, and an HPGe detector after finishing the permeation. Unfortunately, we were statistically unable to find the 0.538-MeV γ ray peak in the spectra. This means that the areal density of ¹⁹¹Pt was below 10^7 cm^{-2} .

3. Absorption experiments

3.1 Experimental apparatus

Among possible nuclear reaction products, neutrons and γ rays are detectable outside the gas absorption chamber. During the absorption runs using the A₁-A₂ system, these were simultaneously monitored with a BF₃ neutron dose rate meter and a scintillation probe, respectively [7-9]. The radioactivity of some samples after the absorption runs was also measured using an imaging plate. However, we have not succeeded in finding meaningful signals in these measurements.

X rays and charged particles have a mean free path or a range too small to allow measurements outside the chamber wall. An experimental system shown schematically in Fig. 2 was prepared for charged particle measurements independently of the A₁-A₂ heat measurement system. A solid state detector, a Si surface barrier detector (SSBD) or an ion implanted Si detector (IISD), is located inside the reaction chamber containing a sample holder on which several tens of milligrams of the Pd-Zr oxide complex samples are mounted. A sheath heater is wound around the holder for sample baking, and the holder temperature is measured with an alumel-chromel thermocouple attached on the side of the holder. During the sample baking a retractable shield plate is inserted between the sample and the SSBD/IISD to avoid its deterioration by heating. Energy calibration of the SSBD/IISD is done with use of an ²⁴¹Am checking source placed on the retractable shield. During the calibration and before deuterium absorption runs, the chamber is evacuated with a TMP-diaphragm pumping kit. The deuterium gas is introduced into the reaction chamber through a liquid nitrogen cold trap and a “SuperNEEDLE” valve which enables precise control of the gas flow rate mechanically.

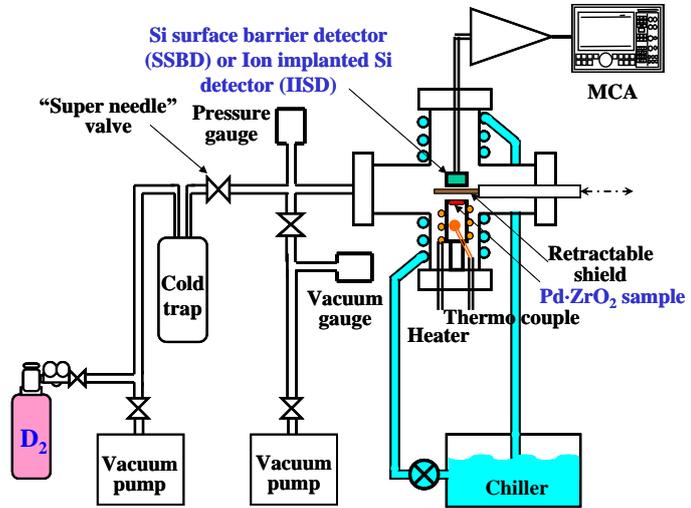


Fig. 2. - Schematic of the system B for particle measurement.

3.2 Examples of the measurement

In the initial stage of this study, we used an SSBD with a depletion layer thickness of 200 μm and the used sample PZ1. The energy spectra recorded during a 5-day run D-PZ1#3B through D-PZ1#4B and evolution of the counting are shown in Fig. 3(a) and (b), respectively. Here, the run number following “#” is advanced for every run after each gasfill-evacuation cycle, while the number preceding “#” is advanced for every new piece of the sample PZ (Pd-ZrO₂).

We see several counts in the energy range from 2 to 4 MeV and a few counts at 7 - 9 MeV. Although these could be caused by charged particles, we cannot rule out the possibility that these are due to electronic noise induced by mechanical shock or oscillation. It is shown in Fig.3(b) that these counting increased during phases of pressure change. Although it is fascinating to imagine that some nuclear events are occurring when

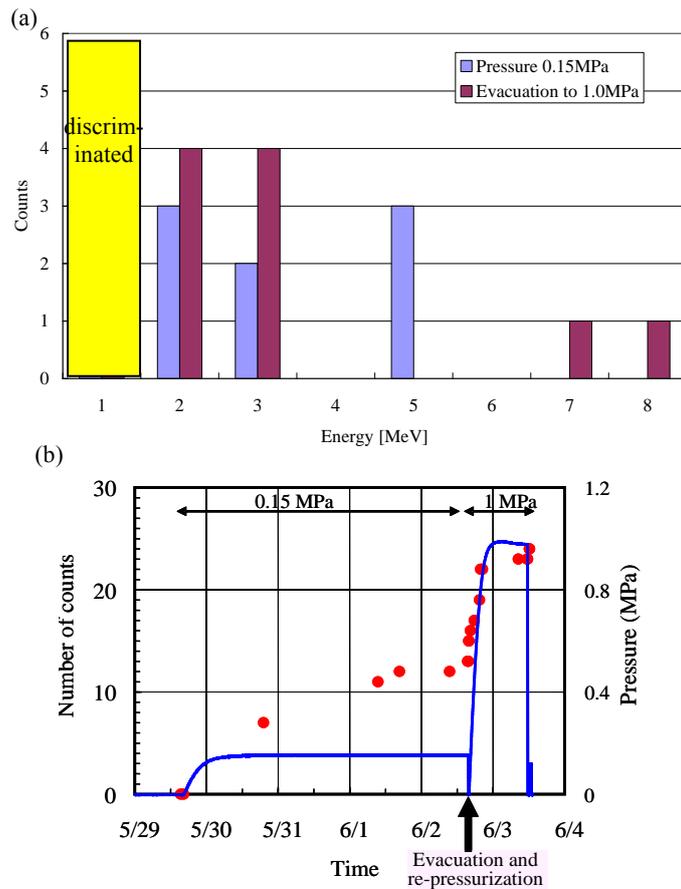


Fig. 3. - (a) Spectra recorded by SSBD with a 200-μm-thick depletion layer, and (b) Evolution of counting in the “1-12 MeV” range of the spectra measured by the SSBD during the run D-PZ1#3B through D-PZ1#4B.

deuterium atoms flow into/out of the Pd lattice, we cannot draw such a conclusion at present.

It is known that any SSBD suffers from deterioration or breakdown when it is used in a hydrogen atmosphere. The output pulse height from the SSBD used in the run D-PZ1#3B through D-PZ1#4B was reduced by a factor of about 30% and the FWHM increased by a factor of about 5. Although the deteriorated characteristics was stabilized after the initial use for several hundred hours, it was better to abandon it in the succeeding runs using virgin samples. We used an IISD with a depletion layer thickness of 500 μm which is known to endure use in a hydrogen atmosphere.

Figure 4(a) and (b) show the energy spectra and evolution of pressure and counting of the IISD and other radiations monitored outside the reaction chamber during the runs D-PZ14#1B (blue), D-PZ15#1B through D-PZ15#2B (red). The broken line in (a) shows the spectrum of ^{241}Am - α particles used for energy calibration of the detectors. We observe several tens of counts in the energy range 2 - 5 MeV in these runs. In (b) the IISD countings are integrated over the runs D-PZ14#1B, D-PZ15#1B-2B, and the baking period. They are reset between the operations.

The integrated IISD counting increased during the baking period between D-PZ14#1B and D-PZ15#1B, which is due largely (85%) to signals lying in the energy range 4.7 - 5.6 MeV. This indicates that the upper surface of the retractable shield plate has a contamination of ^{241}Am . It was found that the active surface of the IISD could see the contamination spot in case of insufficient retraction of the shield plate during the runs. This can account for the increase in the counting also during the runs D-PZ14#1B and D-PZ15#1B-2B.

As shown in Fig. 4(a), the spectra during these runs are a little different from that of the ^{241}Am - α particles, which is also implied by the difference between the counting in the energy range 1 - 8 MeV and that in the range 4.7 - 5.6 MeV. The energy shift of the ^{241}Am - α particles by about 0.8 MeV is consistent with the energy loss of 5.486-MeV α -particles in a 5 mm-MPa hydrogen gas. We see again, however, a tendency that the counting increases during the phase of pressure change. It could be possible that the signals lying in the energy range 2.0 - 4.5 MeV originate in charged particles emitted through some nuclear phenomenon in the sample.

Finally, it is noted that we observed no increases in the signals of neutrons and gamma rays.

3.3 Summary of charged particle measurements in gas charging system

We have observed several to several tens of counts in the 1 - 10 MeV range, which appeared to be emitted coincidentally with pressure change. However, we cannot rule out the possibility that they are due to electronic noise or contamination of ^{241}Am . Up to now, we have obtained no firm evidence of charged particle emission. We are planning detection of X-rays as another candidate for the reaction products.

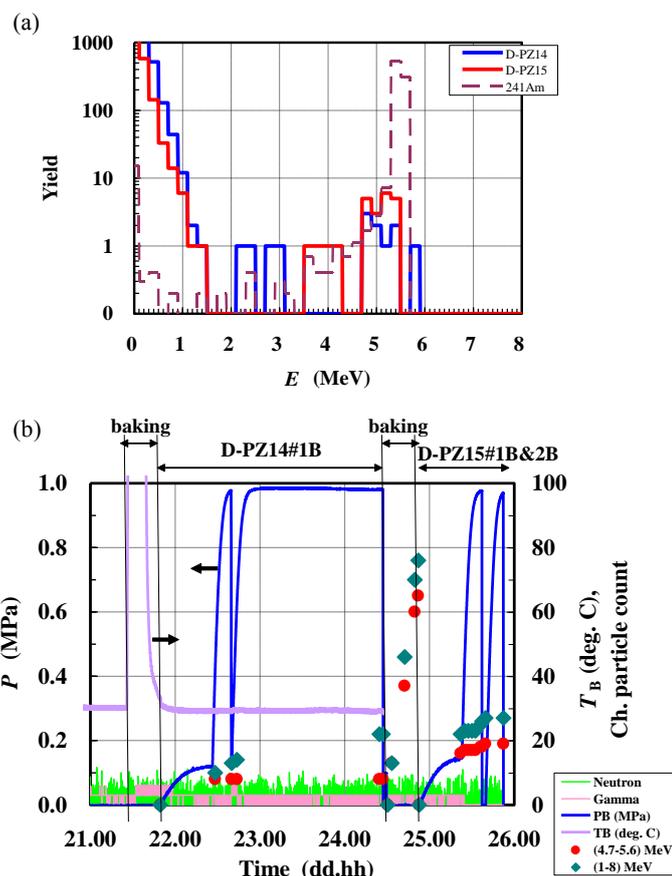


Fig. 4. - (a) Spectra recorded by the IISD with a 500- μm -thick depletion layer, (b) Evolution of pressure and IISD counting and other radiations during the runs D-PZ14#1B through D-PZ15#2B.

4. Concluding Remarks

Employing both *in situ* and *ex situ* accelerator analyses, we have attempted to replicate the Iwamura-type nuclear transmutation of Sr to Mo under deuterium permeation through a variety of multilayered CaO/Sr/Pd samples. Apparently positive results have been obtained in 8 of 14 runs, although the identification of Mo peaks in the PIXE analysis is not definite. It is implied that sputtering loss of the atoms could be responsible for the observed tendency that the areal density of Sr decreases in most cases, while there are modest increases in Mo. Gamma ray measurement for samples implanted with W atoms has also been performed in expectation of transmutation from ^{183}W to radioactive ^{191}Pt , but we have not yet obtained positive results.

In deuterium absorption experiments of Pd-Zr oxide nanopowders, detection of possible nuclear ash including charged particles in addition to neutrons and gamma rays has been examined. We have observed several to several tens of counts in the 1 - 10 MeV range, which appeared to be emitted coincidentally with pressure change. However, we cannot rule out the possibility that they are due to electronic noise or contamination of ^{241}Am . Up to now, we have obtained no firm evidence of charged particle emission.

In both of the above measurements, one of the problems is that the signal-to-noise ratio is rather small. If any nuclear reaction is occurring in the former experiments, it is necessary to find a way to increase the reaction rate not only from a detection point of view but also from a practical application point of view. In the latter, if any nuclear reaction emitting charged particles is responsible for the anomalous heat evolution, it should be possible to detect millions of charged particles. We need further study to look for the conditions suitable for such a reaction and its detection techniques.

5. References

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