Observation of Nuclear Reaction in Glow Discharge Experiment Using Deuterated Palladium Electrode

S.Narita*, H.Yamada, A.Arapi, D.Kato, M.Yamamura, M.Itagaki

Department of Electrical and Electronic Engineering, Iwate University

Morioka, Iwate, 020-8551, JAPAN

* narita@iwate-u.ac.jp

ABSTRACT

We performed discharge experiment using Pd deuteride cathode in deuterium atmosphere. Gamma ray emissions in the 80-330keV region were observed during the discharge with certain efficiency. It was assumed that a nuclear reaction took place in the Pd cathode and some short-lived radioisotopes were produced. The low-energy photofission model showed good agreement with the part of our experimental results. Elements on the Pd cathode and their isotopic abundance were analyzed by a time-of-flight secondary ion mass spectroscopy to find the evidence of nuclear reaction.

Keywords : glow(-like) discharge, Pd deuteride, gamma ray, surface analysis, time-of-flight secondary ion mass spectroscopy, low-energy photofission

1. Introduction

We have reported gamma ray emission in the 70-110keV region and the element production on deuterated Pd cathode in DC glow discharge experiment in deuterium atmosphere [1,2]. It has been supposed that a low energy nuclear reaction, producing some elements including radioisotopes, was induced in the experiment. However the reaction efficiency was quite low and the trigger condition for inducing the reaction was not understood. In this study, we attempted to increase the deuterium density in the discharge space, that is, the pressure of atmospheric deuterium gas was changed from ~3Torr to ~1atm expecting the reaction efficiency to be improved. This discharge condition was definitely different from that of the conventional glow discharge, and it was so-called "glow-like" discharge. Moreover, we precisely analyzed the energy of detected gamma rays and nuclear products aiming to specify the reaction occurring during the experiment, considering the existing theoretical model.

2. Experiment

The experiments were carried out by DC glow-like discharge with a deuterated Pd (Pd/D) foil cathode in deuterium atmosphere. The Pd sample used was 10x10x0.1mm^3 in size and >99.95% in purity. At first, the sample was washed with acetone and aqua regia for 100s. Then, it was put in a chamber and loaded with deuterium gas under 10atm pressure for ~48h. The loading ratio was determined by measuring the mass change of the sample, and it was typically 0.6-0.7. After gas loading, the sample was placed in a discharge cell. The cell made of Pyrex glass has a cylindrical shape with an inner diameter of ~12cm and a volume of ~1000cm^3. The thickness of the glass is 5mm. By using Pyrex glass cell instead of metallic one, the contamination to the sample can be minimized and the precise analysis of nuclear products is enabled. The cell consists of two parts connected to each other with silicone grease. It is connected to a vacuum system via a valve on the upper part of the cell so that we can drive out the gaseous impurities in the cell and control the pressure inside the vessel. An Au foil (0.3mm in thickness) hung by an Au wire was used as the anode. The Pd sample was placed as a cathode on an Au stand with a quartz cylinder surrounding it to prevent movement during the discharge. The gap distance between the two electrodes was ~10mm. After closing up the cell, it was evacuated to 10^{-2}-10^{-3}Torr and deuterium gas was supplied until the pressure inside became 1atm. Then, DC voltage was applied to expose the sample to discharge with currents of 2-4mA and voltage of 4000-6000V. The duration time of the discharge was 60min.

A NaI(Tl) scintillation counter (SCIONIX ϕ 25mm x 25mm crystal) was used to detect gamma rays from the experimental system. It was placed perpendicularly ~10mm away from the side wall of the cell. The photons from the scintillation counter were detected by a photomultiplier tube (PMT). The signal from the PMT was amplified and the pulse height distribution was obtained using a multi-channel analyzer (ORTEC ScintiPack), then it was converted to an energy distribution of the gamma rays. The energy calibration for the counter was executed using ^{57}Co and ^{137}Cs, which emit gamma rays with the energies of 122.1keV and 661.7keV respectively. The energy resolutions were estimated to be ~25keV and ~80keV (FWHM) for both energies.

For some samples, the surface was analyzed by time-of-flight secondary ion mass spectroscopy (TOF-SIMS) (ULVAC-PHI:TFS-2100) after the discharge. TOF-SIMS has good sensitivity for detecting a small amount of elements on the surface, and high mass resolution. In addition, TOF-SIMS was capable of analyzing all the elements including their isotopes. However, it is difficult to evaluate the concentration of the elements quantitatively from TOF-SIMS results alone. Therefore, in this study, we only discuss the possibility of new element production qualitatively and anomaly in the isotopic abundance for the elements detected.

3. Results

3.1 Gamma ray measurement

Figure 1 shows the energy distribution measured in the absence of discharge as a background run. For most of runs with discharge using Pd deuteride cathode, quite similar distributions to that for background runs were obtained. However, the anomalous signals in the gamma ray spectra with energies below 500keV sometimes appeared. Such signals were observed in 13 out of 68 runs, ~20% of total runs. The spectra measured in those 13 runs are

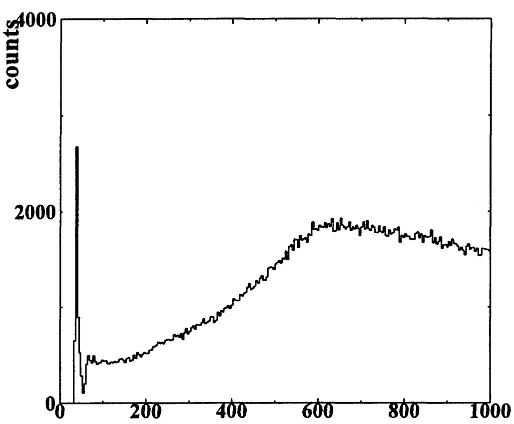

Figure 1: Gamma ray spectrum for the background run.

shown in Figure 2. Between the runs for sample-5 and sample-6, a trouble happened on the electronic system of the detector, then it was fixed and the recalibration for the system was carried out. This is why the height for the dark current signal, i.e. the peak at the lowest energy in each spectrum, was significantly lower for sample-6 through sample-13. Observing the on-line monitor, we noticed that the anomalous peaks in the spectrum were formed just after the supply of the DC power, continuously growing the discharge. If these signals were caused by the electric noise introduced by the discharge or signals continuously generated in the data acquisition system, they should have appeared in all runs. Thus, it was supposed that radioactive sources were produced during the experiment and they emitted the gamma rays. In anomalous events, some spectra have obviously two peaks and other have apparently single peak. However, if we carefully look at such single-like peak and consider the energy resolution of the detector (see above), it may contain at least two monochromatic peaks. So, in this analysis, a combination of two Gaussian functions and a linear function was chosen to be fitted to the observed spectra to determine the energy of the emitted gamma rays precisely. Table 1 shows the mean value and the standard deviation obtained by fitting the two Gaussians. The emitted gamma ray energies distribute in the range 80-330keV.

We also carried out a discharge experiment with hydrated Pd cathode in hydrogen atmosphere in 50 runs, and no anomalous peak in the gamma ray spectrum was observed. This is additional evidence that the anomalous gamma signals in deuterium experiment were not due to a kind of electric noise.

3.2 Surface analysis

For some Pd samples, the surface was analyzed by the TOF-SIMS to search for the symptom of nuclear reaction. As mentioned above, since it is difficult to estimate how much a specified element exists in an analyzed area by TOF-SIMS alone, our analysis was concentrated on checking the existence of some particular elements and the change in their isotopic abundance. In this analysis, we examined 8 samples (4 out of 8 are the samples with which the gamma rays were observed during the discharge) and 5 fresh samples, and compared them. As a result, we did not find clear evidence that any new elements were produced. In our glow discharge experiment (i.e. discharge with deuterium gas at a pressure of ~3Torr), Be had been found to be a candidate for the nuclear reaction product [2], but it was not detected at all. On the sample surface, there are K, Ca, Ni, Cr, Fe and Cu found as impurities. The isotopic abundance for these elements was checked, and no anomaly was found, that is, it was in good agreement with natural one.

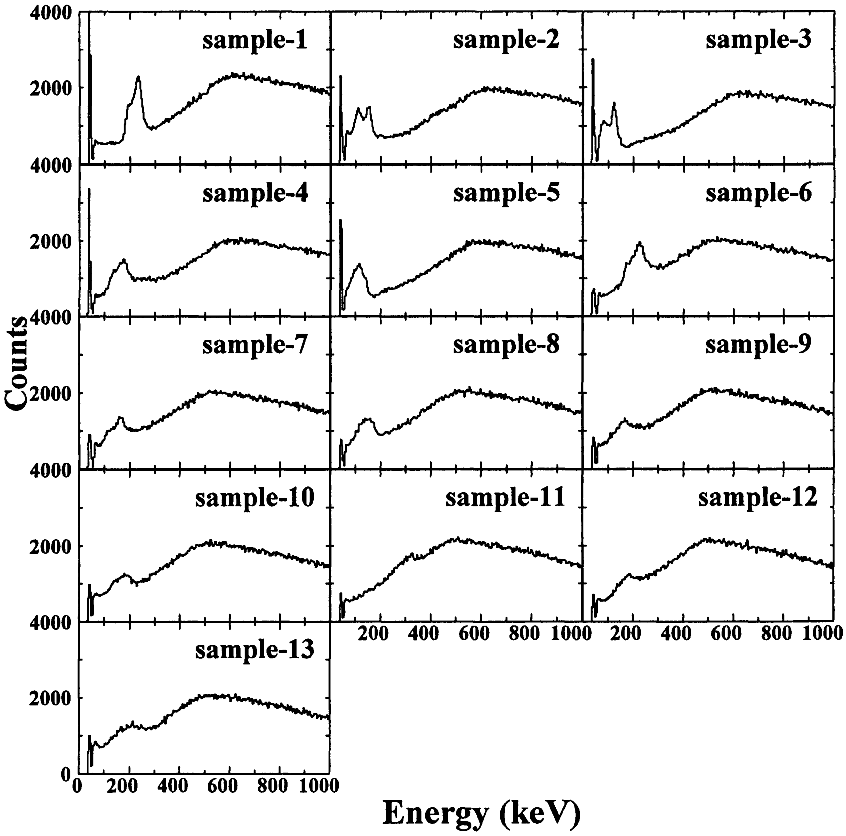

Figure 2: Anomalous signals in gamma ray spectra.

Sample ID	E1 (σ) [keV]	E2 (σ) [keV]
1	193.8 (10.2)	230.2 (16.6)
2	113.3 (15.6)	153.3 (10.6)
3	83.6 (15.2)	124.2 (15.2)
4	134.0 (14.6)	172.2 (17.7)
5	91.0 (25.3)	124.3 (18.5)
6	170.1 (17.0)	219.1 (24.5)
7	117.5 (13.0)	162.6 (19.0)
8	132.1 (23.6)	164.8 (13.1)
9	165.1 (21.5)	215.6 (12.9)
10	137.9 (20.4)	184.5 (20.2)
11	259.0 (22.2)	313.0 (27.6)
12	137.5 (11.2)	179.7 (25.5)
13	153.1 (21.8)	206.6 (29.3)

Table 1: Gamma ray energy from Gaussian fit and the standard deviation.

4. Discussion

Now the trigger condition for inducing the reaction is discussed. Figure 3 shows D/Pd ratio for all samples and the hatched histogram indicates that in the samples with which anomalous gamma ray signals were observed. As is seen in the figure, no clear evidence for the loading ratio dependence was obtained. It has been claimed that there is a strong correlation between the efficiency of the nuclear reaction in the condensed matter and the D/Pd ratio, especially for the case of deuteron fusion [3,4]. Although we have not understood the reaction that occurred in our experiment yet, it is possible that the D/Pd ratio affects the reaction efficiency even in our experiment. If so, the critical threshold of the loading ratio for effectively inducing the reaction may be much higher.

Then, the reaction efficiency can be also related to the discharge condition. In our previous glow discharge experiment with different condition, that is, at a pressure of ~3Torr, we observed similar gamma emissions in 4 out of 105 runs, 4% of total runs. It turns out that the reaction is more likely to take place in glow-like discharge rather than glow one. Therefore, the discharge under a significantly higher density deuterium environment is expected to induce the reaction more effectively. As our current cell is not designed for a discharge in a pressure of greater than 1atm, new type cell will be prepared. Moreover, in order to clarify the trigger conditions for the reaction, other discharge conditions such as current density, distribution of the electric field (shape and arrangement of the electrodes) and so on should be also considered in the future study.

Another interest is what kind of reaction happened during the discharge and how radioactive sources were produced. One of the considerable processes for the formation of radioisotopes in our experimental condition is the low-energy photofission (LEPF) of Pd proposed by Takahashi et al. [5]. They claim that a high peak flux of low-energy photons in a dynamic lattice of Pd deuteride (hydride) can lead to the fission of Pd via a selective channel. The model shows good agreement with the some experimental results on the nuclear products such as Z-distribution, mass distribution and isotopic ratio. According to the model, major fission products are stable isotopes, and there are also reaction channels in which short-lived radioisotopes are produced. Such radioisotopes can emit gamma rays with various energies in their beta decay process. Among them, the isotopes which can emit gamma rays in the 80-330keV range are listed in Table 2. In this table, the radioisotopes, which are produced with relatively lower yield in the LEPF of Pd, are not shown. Because the gamma ray detection was

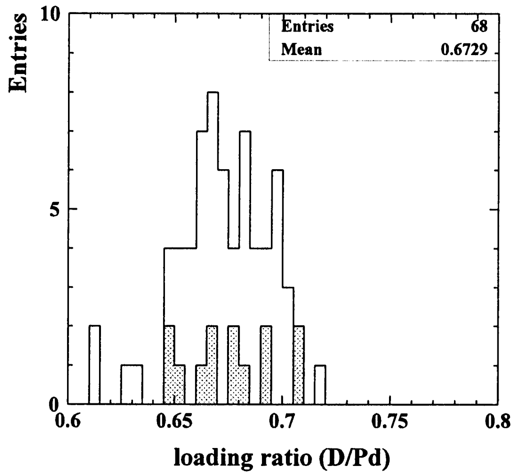

Figure 3: Loading ratio, D/Pd. The hatched histogram is for the sample in which anomalous gamma rays were observed.

Nuclide (half life)	Yield [%]	E_γ [keV] (Release rate [%])
^{50}Ca (87.7s)	1.03	71.6 (52.0)
^{56}Cr (5.9m)	1.50	83.9 (95.3)
^{63}Co (27.4s)	0.57	87.1 (48.7)
^{53}Ti (32.7s)	1.33	100.8 (20.3)
^{57}Mn (87.2s)	1.33	122.1 (13.9)
^{52}Ti (1.7m)	1.50	124.5 (98.6)
^{58}Cr (7.0s)	1.00	126.0 (75.0)
^{53}Ti (32.7s)	1.33	127.6 (46.0)
^{44}Ar (11.9m)	1.06	182.6 (66.0)
^{53}Ti (32.7s)	1.33	228.4 (40.0)
^{50}Ca (13.9s)	1.03	256.9 (98.0)
^{58}Cr (7.0s)	1.00	289.5 (18.8)
^{61}Fe (6.0m)	1.33	297.9 (22.2)
^{51}Ti (32.7s)	1.33	320.1 (93.0)

Table 2: List of major nuclides, which can emit 70-320keV gamma rays [6]. These are predicted to be produced in the LEPF of Pd. The yields were calculated by Dr. M.Ohta (Osaka University)

performed only during the discharge, i.e. 60min, radioisotopes with relatively longer lifetime are also excluded from the list. Some gamma ray energies observed in our experiment look corresponding to the predicted ones in the table. However, some signals, observed in 150~170keV, are not predicted to appear in the model. On the other hand, in Table 2, there are some isotopes emitting gamma rays which we did not observe in the experiment. Actually, on this matter, we should investigate the effective yield of gamma rays, considering the yield of each isotope in photofission process, the release rate of gamma rays, and the energy dependence of the detection efficiency for the gamma rays. In addition, since the model predicts the production of other isotopes which emit gamma rays with higher energy (up to a few MeV), such signals should be surveyed for examining the applicability of the model to the phenomenon observed in the present work.

In order to justify the LEPF model, the origin of low energy photon has yet to be clarified. Takahashi et al. have mentioned that coherent multibody deuteron fusion of deuteron in Pd can be responsible for existence of strong bursts of low-energy photons. If such reaction happens simultaneously, X-rays emission and He production can be observed as the evidence [7]. This is another interest for the future study. Additional consideration is necessary for the results in hydrogen experiment. If high flux low-energy photons are generated around the Pd cathode in the glow(-like) discharge state itself, the LEPF can be induced even in Pd hydride sample, in principle. However, we did not observe any anomalous gamma rays in hydrogen system, as described above. This result suggests that the gamma ray emission is caused by deuteron related reaction. Hence, as far as our experiment is concerned, the low-energy photon burst generated by deuteron fusion can be a candidate for the photon source inducing the LEPF.

In elemental analysis, we did not find any anomalies. If a nuclear reaction took place during the experiment, a symptom such as changes in the isotopic abundance should have been found. The LEPF model predicts that some isotopes of Ca, Cr, Fe, Cu and so on can be produced selectively by the fission of Pd isotopes. It is probable that just a small amount of the elements have been produced with the yield below sensitivity of the TOF-SIMS. For confirming the assumption, we will attempt to make the yield to be enhanced and search out the products carefully with various methods.

5. Summary

We observed anomalous gamma ray signal with 80-330keV in DC glow-like discharge experiment. It suggested that some radioisotopes were produced in the discharge. We found that the efficiency can be improved by changing the discharge condition. The LEPF model is a candidate for the model describing the reactions which may take place in our experimental system and explain the formation of radioisotopes emitting such gamma rays. Some, but not all, of our results agree with the LEPF prediction. Several ideas are considered for future study to understand the trigger condition to induce the reaction and clarify the reaction process.

Acknowledgements

We would like to extend our appreciation to Prof. A. Takahashi and Dr. M. Ohta (Osaka University) for their helpful comments on this work.

References

1. H.Yamada et al., Fusion Technol, 39 (2001) 254.
2. A.Arapi et al., Jpn. J. Appl. Phys., 41 (2002) L1181.
3. M.McKubre, Proc. of 3rd Int. Conf. Cold Fusion, (1993) 5.
4. D.Azzorone et al., Proc. of 8th Int. Conf. Cold Fusion, (2000) 199.
5. A.Takahashi et al., Jpn. J. Appl. Phys., 40 (2001) 7031.
6. R.B.Firestone and V.S.Shirley, "*Table of Isotopes*", (John Wiley and Sons, Inc.) 8th ed., (1996)
7. M.Matsunaka et al., Proc. of JCF4 meeting, (2002) 32.