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EXPERIMENTAL OBSERVATION OF THE NEW ELEMENTS PRODUCTION IN THE DEUTERATED AND/OR HYDRIDE PALLADIUM ELECTRODES, EXPOSED TO THE LOW ENERGY DC GLOW DISCHARGE.

A.Arapi, R.Ito, N.Sato, M.Itagaki, S.Narita, H.Yamada
Faculty of Engineering, Iwate University, Morioka 020-8551, Japan

ABSTRACT

Elemental and isotopic structures of the palladium cathode before and after experiment under DC glow discharge were investigated by time of flight secondary ion mass spectrometry (TOF-SIMS). Production of new elements with various atomic masses and impurities increase were surveyed. By analyzing obtained results, it was found that beryllium and nickel for deuterium-palladium system, and lithium, nickel and barium for hydrogen-palladium system, were generated during glow discharge experiments.

1. INTRODUCTION

The possibility of inducing nuclear reactions at low temperature in solid-state materials has been widely investigated by DC glow discharge experiments for several years. Previously during this experiment low energy gamma emissions with 70-110 keV and increase of the impurities were reported1). Excess heat, impurities nuclides yield and high energy radiation have been observed in experiments with high current glow discharge.2) Savvatimova et al.3) have reported production of different elements with atomic mass lighter and heavier than Pd isotopes during glow discharge using deuterium (D) or hydrogen (H) gas. Mizuno et al.4,5) also have demonstrated the production of elements, during electrolysis, with mass number range from 1 to 208 with isotopic distribution quite different from the natural ones.

Because of irreproducibility of the detecting radiation during experiment our attention was concentrated on the element analysis that seemed to be reproducible and give an important result in understanding the phenomena. High mass resolution of the TOF-SIMS allowed separating of the most of atomic ions from ions fragmentation at the same nominal masses and reliable peak assignments were made due to the accuracy of the mass determination. For this reason each sample’s composition was analyzed before gas absorption and after discharge experiment by TOF-SIMS analysis method. In this kind of experiment, the evidences that indicated the occurrence of some nuclear reaction were given in the form of appearance of new elements that were not introduced at all on the Pd samples before gas absorption as well as significantly increase of the impurities.

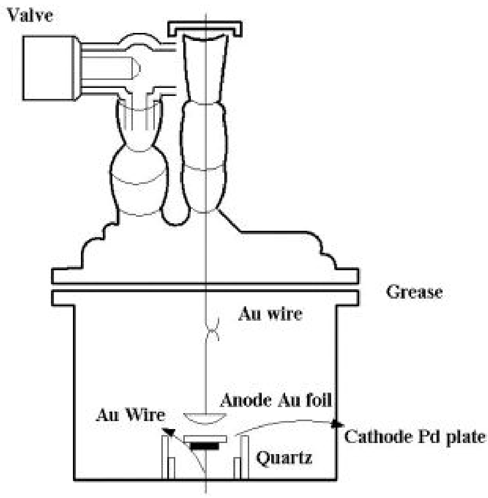

Fig.1 Pyrex glass cell

2. EXPERIMENTAL SETUP.

The experiments were carried out with the aim of determining possible new elements production from Pd plate, subjected to DC glow discharge in deuterium or hydrogen working gas. Many efforts were made to avoid any possible contamination during preparation of the samples as well as during discharge experiment. The experiments were carried out with glow discharge at 60 min, according to the following procedure. At the beginning each Pd sample (10x10x0.1mm in size, 99.95% pure) was washed with aqua regia for 100s and then its composition was analyzed by TOF-SIMS. This analysis allowed us to know the elemental composition of the Pd cathode before gas absorption and used it as a control result. Absorption of deuteron or proton in the Pd electrodes was performed by applying gas-loading method, which could involve fewer impurities than other ones. The samples were put into the loading chamber. The chamber was evacuated at 10-3Torr and then filled with deuterium or hydrogen until 5 and 10atm, respectively for 48h. The loading ratio was determined by measuring mass change of the samples. After that the samples were taken out from the chamber, they were placed in a discharge test cell.

The configuration of the test cell is shown in Fig. 1. The test cell was made by Pyrex glass material, in which two parts were jointed together by using silicone grease (Dow Corning) which kept the vacuum at the required pressure, usually ~3Torr. Also two adapters and a valve were jointed to the upper part of the cell. A golden foil (0.03mm in thickness), hanged on the Au wire, was used as anode and rectangular Pd plate as cathode. The cathode was supported in the Au wire and a quartz cylinder surrounding it to avoid any moving of the samples during experiment. The gap distance between electrodes was 10mm and between them a DC was applied. The advantage in using Pyrex glass material comparing with metal one was that there were less possibilities for Pd samples to be contaminated by the sputtering process during glow discharge. The samples were exposed to discharge with DC ~2mA, voltage 600-800 V, pressure ~3Torr and duration time 60min. The gamma ray emitted from the samples was measured by NaI scintillation counter that was set perpendicular to the wall of the cell. After the DC glow discharge the composition of the Pd samples were investigated again by TOF-SIMS.

3. RESULTS AND DISCUSSION

3.1 The composition of the Pd samples before gas absorption.

The composition analysis of the samples was performed at the different areas, 40x40μm for each, on the Pd samples and the composition of them has been revealed to be almost the same over the whole analyzed areas. Each sample was analyzed at least in five randomly selected areas when the data were collected only with sputter cleaning by Ga+ ion beam for 10s (“the sputtering area”) usually for D-Pd system, and in three selected areas when the data were collected without (“uppermost area”) and with sputter cleaning usually for H-Pd system. Note that the selected areas before experiment were different with those analyzed after it. The analysis was performed by considering only counts intensity and/or normalized intensity of the signals but not the quantity. The normalized intensity values were estimated by divided counts intensity of each mass by total counts used to normalize peaks, times 104. In total, 10 samples for D-Pd system and 3 samples for H-Pd system were examined. The obtained data for samples analyzed before gas absorption showed that elements B, Na, Al, Mg, Si, Ca, K and Mn were presented on the Pd metal and for elements Na, Al, Ca and K very high peaks were sometimes observed. Elements Li, Fe, Cr, and Cu were found in a much smaller amount. In this study only elements that did not appear at all from the sample before discharge but appear from that after it as well as those that appeared with remarkable increase in counts after discharge experiments, were considered as real proof of an anomalous phenomenon.

3.2. TOF-SIMS analysis for D-Pd system.

The composition analysis of the Pd electrodes showed that Be and Ni were indeed generated during experiment. Both these elements were produced in two out of ten runs. The control results did not show any signal for these masses at all. Also these elements could not be introduced as impurities transported from environment. Table I presents the counts intensity of Be, Fe, Ni, and Cu in different positions of the sample. The TOF-SIMS data were collected only in the "sputtering zone". The loading ratio was D/Pd=0.68 and the discharge experiment was accomplished under 600V in ~3Torr. In some areas, the signals of the 59Co and 64Zn were observed after discharge. Zn has been claimed as nuclear product also in the Pd-hydride by other studies. 6,7) The atomic masses of Be and Ni are lower than that of the Pd, which could induce us to think of the fission process. We observed the significant increase in the counts for 56Fe and 63Cu even in the Pyrex glass cell (see Table I) that reinforced the possibility of their nuclear origin. Moreover a remarkable increase was observed for Li in three out of ten runs. Ohmori et al.8) has also reported the Fe production by electrolysis in gold electrode.

The other experiment was performed by presenting the palladium samples in spark discharge with deuteron gas. During this experiment different radiation less than 228keV were observed. The so -called "228keV sample" analyzed by TOF-SIMS showed that isotopic distribution of the Mg was different from natural abundance one. Moreover the spark discharge experiment carried out in air did not show any emitted radiation.

Analysis Time	Number of Areas	^{9}Be	^{56}Fe	^{58}Ni	^{63}Cu
Before Gas Loading	G01	0	1208	0	0
	G02	0	302	0	0
	G03	0	521	0	0
	G04	0	0	0	80
	G05	0	385	0	0
	G06	0	19339	0	0
	G07	0	309	0	0
After Discharge	G11	904	34713	1769	4363
	G12	445	4831	2177	0
	G13	1375	26782	5298	0
	G14	1755	46880	0	7726
	G15	4212	34550	0	6944
	G16	925	24848	771	0
	G17	308	15606	0	0
	G18	90	0	0	0
	G19	492	7772	0	0
	G20	185	7519	0	636

TABLE I

Counts intensity of ^{9}Be, ^{56}Fe, ^{58}Ni and ^{63}Cu for all selected areas

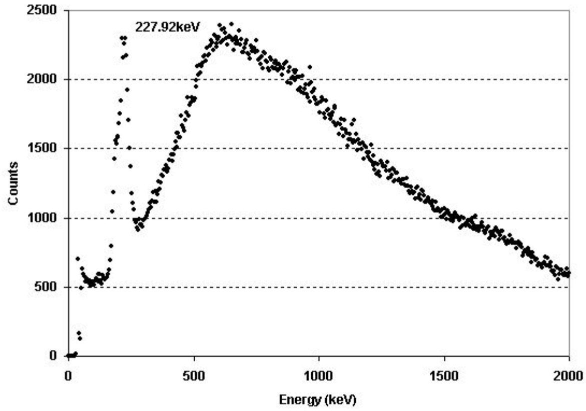

Fig.2
Radiation spectrum after one-hour spark discharge in deuteron atmosphere. (foreground)

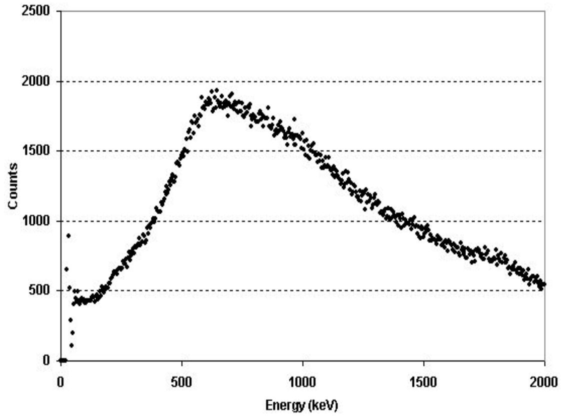

Fig.3
Radiation spectrum without spark discharge (background)

3.3. TOF-SIMS analysis for H-Pd system

The same investigation was performed in the H-Pd by assuming that proton and deuteron might played a similar role in the reaction. The Li, Ni and Ba were detected after experiments for two out of three runs. All Ni and Ba isotopes were presented on the uppermost and sputtering area. Table III shows the variation of the normalized intensity for 7Li, 58Ni and 138Ba in all selected areas. One could easily see the remarkable increase of the Ba from uppermost area to the sputtering one by comparing the values of uppermost to sputtering zone at area number 3, 4 and 5 in Table III. Table IV shows the comparison of the mass deviation between 138Ba, 69Ga2, and possible combination of the Pd and Si isotopes. Since mass deviation of the 138Ba from the signal is much smaller than that of other species, the signal should correspond to 138Ba. Furthermore, high peaks were found for ions fragmentation with masses 147-155 and 157-164 that corresponded to molecular fragmentation of the Ba isotopes with other elements. Note that this group masses distribution ratio was almost similar with natural isotopic distribution of the Ba. This fact showed that Ba was presented in the Pd lattice in very large amount. The appearance of the lighter and heavier masses than Pd isotopes might be caused by a kind of fusion fission reactions3) induced by protons in the Pd.

TABLE III

Counts of ^{7}Li, ^{58}Ni and ^{138}Ba each divided by the total counts used to normalize counts times 10000 for all selected areas

	Number of Area	Normalization Intensity ^{7}Li	Normalization Intensity ^{58}Ni	Normalization Intensity ^{138}Ba	Zone
Before Gas Loading	1	0	0	0	Uppermost
		0	0	0	Sputtering
	2	0	0	0	Uppermost
		0	0	0	Sputtering
After Discharge Experiment	3	21	49	87	Uppermost
		9	2	316	Sputtering
	4	16	24	81	Uppermost
		9	2	315	Sputtering
	5	5	35	130	Uppermost
		6	2	770	Sputtering

TABLE IV

Comparison of the mass deviation between ^{138}Ba, Ga_2, $^{108}Pd^{30}Si$ and $^{110}Pd^{30}Si$

Species	Original Mass9) (amu)	Signal's Mass (amu)	Mass Deviation (Δm (mamu)
^{138}Ba	137.90524	137.9050	0.2
Ga_2	137.85116		53.0
$^{108}Pd^{30}Si$	137.87759		27.4
$^{110}Pd^{28}Si$	137.88207		22.9

4. CONCLUSIONS

The Li, Be, Ni, and Ba were the most obvious elements produced on the Pd cathodes that provided the new evidences of the anomalous nuclear reactions triggered by DC flow in the Pd deuteride and/or Pd hydride. The obtained results showed the generation of Li and Ni for both D-Pd and H-Pd systems,which might support that deuteron and proton played the similar role in the reactions.

The observation of Li, Ni and Ba on the "uppermost area" as well as in the "sputtering area", for H-Pd system, showed that the anomalous nuclear reactions might occur on the surface and/or inside the metal lattice. Moreover the appearance of the elements with atomic masses lighter (Ni, Li) and heavier (Ba) than those of the Pd isotopes from all selected areas for H-Pd system, might suggest a kind of fusion fission reactions.

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