Transmutation

ISOTOPIC DISTRIBUTION FOR THE ELEMENTS EVOLVED IN PALLADIUM CATHODE AFTER ELECTROLYSIS IN D_2O SOLUTION

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Abstract

It was confirmed by several analytic methods that reaction products with mass number ranging from 1 to 208 are deposited on palladium cathodes subjected to electrolysis in a heavy water solution at high pressure, high temperature, and high current density for one month. These masses were composed with many elements ranged from hydrogen and lead. Isotopic distributions for the produced elements were radically different from the natural ones.

1. Introduction

Many claimed that if nuclear reactions have been induced by electrochemical reaction occurring in solid electrodes, there must be clear evidence such as the evolution of radioisotopes and radiation. Moreover, the evolution rates of the reaction products should be quantitatively explained in terms of the proper nuclear reaction mechanisms. But such the claim could be held if the reaction mechanism had been consisted by proper theories. However, there is no proof that the conventional mechanism holds the reactions. It is difficult to detect the emission of the radiation and radioisotope if the mechanism was different from the proper ones. In this work, evidence which indicates the occurrence of some nuclear reactions is presented, in the form of isotopic changed elements in and on the cathode surface. These products have been obtained with a mechanism which had not

induced any detectable radiations. The anomalous isotopic distribution of these elements shows they do not come from contamination. We represent that the reaction mechanism was completely different from the proper nuclei formation process. However, we attempt to explain the process which produced these anomalous products by the mechanism within the framework of the proper theory(1).

2. Experimental

The experimental details for the sample, cell and electrolysis conditions have been described elsewhere(2). The sample electrodes were supplied to element detection after pull off the Teflon coat, washed by the Mill Q water for energy dispersive X-ray spectroscopy (EDX), Auger electron spectroscopy (AES), secondary ion mass spectroscopy (SIMS) and electron probe microanalyzer (EPMA).

3. Results

Many elements have deposited on the surface and distributed irregularly; these concentrations were changed with samples. The elements that have commonly been detected were C, O, S, Cl, Si, Ca, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, Mo, Pd, Sn, Pt, Hg and Pb for all the samples. The amounts for Ca, Ti, Cr, Cu, Pt, Hg and Pb were abundantly existing and differed more than 3 times at the surface place compared with C, Cl, Si, Mn, Co, Ni, Zn and Sn which were existing rather uniformly. These changeable elements were also fluctuating with sample lot as 3 times deviations. It meaning that the uncontrollable factors such as surface conditions are the main importance for the reaction.

Several elements were detected in the Pd electrode by the EDX method; the measurements were taken to know the rough concentration for the elements because in the SIMS measurement the mass peaks have a possibility to contain other molecular peaks. Figure 1 shows typical results before and after electrolysis. Several peaks of Pt, Cr and Fe are clearly seen; these amounts were comparably with Pd bulk peak. And less amounts for Sn, Ti, Cu and Pb elements were also

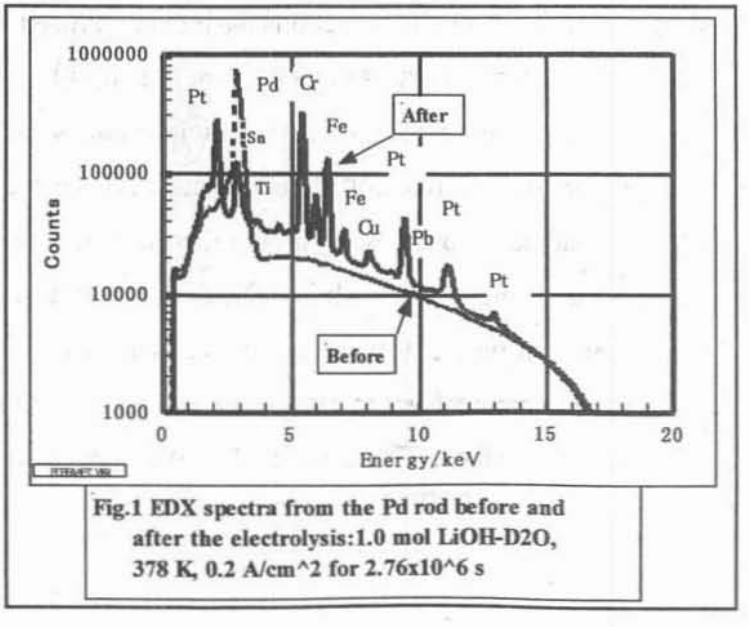

Fig.1 EDX spectra from the Pd rod before and after the electrolysis:1.0 mol LiOH-D2O, 378 K, 0.2 A/cm^2 for 2.76x10^6 s

clearly observed. The EDX analyses were repeated on various place of the sample; the deviation for the EDX counts sometimes reached to 10 times with the place. This change was depending on the scanning area. The amounts for the evolved elements were finally estimated by SIMS measurement. The EDX, AES and EPMA methods were complimentary used to correspond a mass Spectra to a certain atom and decide their isotopic distributions. The procedure is described as follows: (1); the mass number were decided in the first from light mass number. (2); the mass number were adjusted with the EDX and AES spectra. (3) the large count number of mass peaks was confirmed to existence of their molecular ion and oxide ion peaks. (4) the final mass spectra were estimated by multiplying the factors of counting correction to the original count of mass. The factor shows very high and low value at the inert gases and alkali metals respectively.

The ratios of atomic number for the mass were finally estimated with the procedures. The ratios were shown in fig.2 normalized with the total mass set as one. Typical counts by EDX and SIMS ranged from 10^2 to 10^6 and were 10 to 100 times higher than the background counts. Thus, the presence of Ca, Ti, Cr, Mn, Fe, Co, Cu, Zn, Cd, Sn, Pt and Pb was clearly confirmed. These elements are mostly grouped in four ranges of mass number:lightest elements under 50 mass number,light elements from 50 to 80;middle elements 100 to 140; and heavy elements from 180 to 208. The ratio of the mass number from 102 to 110 which correspond with Pd atom is under 1% of total even if it was bulk substance; the large ratio of existence for Oxygen and Xenon pull down their values. The reason for these high ratio can be considered that the many gas atoms may be released and succeeding contributes to the counting rate from the spot place heated up by ion bombardment.

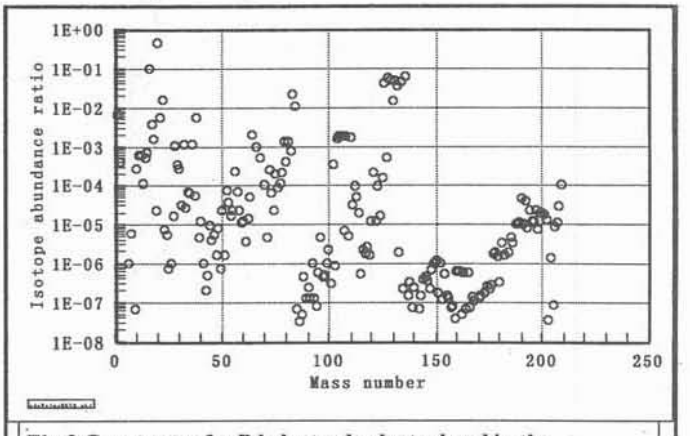

Fig.2 Count rates for Pd electrode electrolyzed in the heavy water solution observed by SIMS measurement.

The SIMS analysis showed other elements; As, Ga, Sb, Te, I, Hf, Re, Ir, Br and Xe. These elements, except Xe, were difficultly detected by AES and EDX because the peaks were very close and sometime overlapped with others and these were lower than the detection limits by the measurements. Xe atoms are naturally difficult to detect by EDX method because the such the gas atoms are easily escaped by ion bombardment from the spot area. The SIMS count numbers ranged from 10^3 to 10^6 where the background counts were as low as ~10, so we have confidence in these results. In Figure 2 we show the peak intensities normalized with the total peak. The intensity of Xe was 10 times larger than Pd; it may be that the gas was released by bombarding with O_2^- ions which

caused a temperature rise at the sample.

We have no peak except Pd been observed after pre-electrolysis Pd surfaces. Pt a Pd concentrations in the electrolyte after experiment were 10 and 15 ppm respectively by atomic absorption measurement and no other elements excepts for Li was observed. Total amount for the elements existed in one micro meter depth of Pd surface were calculated as follows; C: 0.37, S: 0.67, Ca: 0.55, Ti: 0.86, Cr: 7.0, Mn: 0.005, Fe: 0.17, Co: 0.0057, Ni: 0.0157, Cu: 0.026, Zn: 0.80, Mo: 0.005, Pd: 4.77, Cd: 0.105, Sn: 0.069, Pt: 0.025, Hg: 0.0375 and Pb: 0.021 atomic percentage. (The total deposited elements on the Pd and the calculated summation for the impurity in the electrolyte and Pd samples are less one and mere order than depositions.) Here, the total impurities except Ca are less than the deposition amounts; especially, Mn, Ti, Cd and Hg were not existing as the impurity.

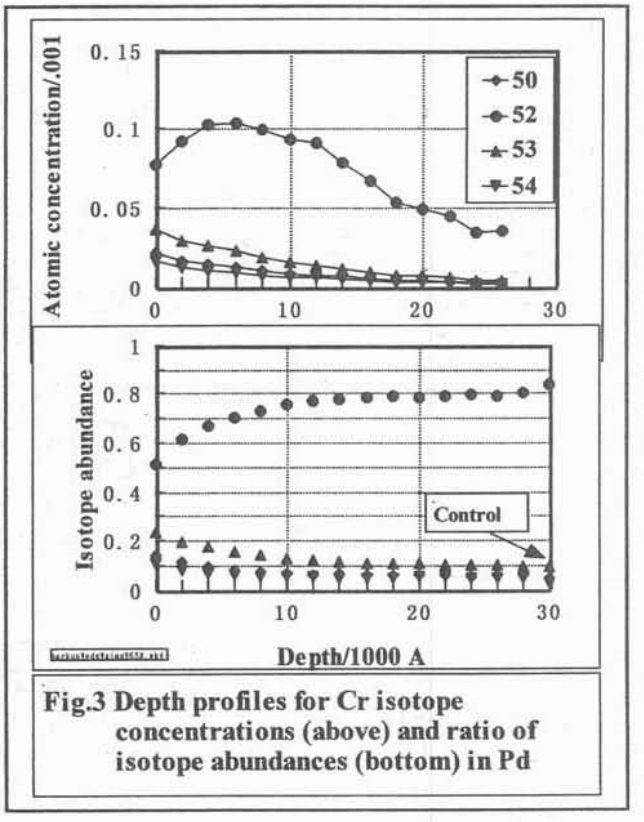

Fig.3 Depth profiles for Cr isotope concentrations (above) and ratio of isotope abundances (bottom) in Pd

Large differences in isotopic distributions compared with the natural distributions were observed by the SIMS method for Cr, Cu, Zn, Br, Xe, Pd, Cd, Hf, Re, Pt, Ir, Pb and Hg. The typical concentrations and their ratios for Chromium isotopes are shown in Fig.3 for an example; the concentrations except Cr52 decreased exponentially with depth but Cr52 has a peak at 0.5 micro meters and they showed largely shift in isotopic ratios: However, we can see large deviations for isotopic existence with natural ones, that means, these are higher than Cr52, i.e., Cr52 is less than others, in the figure where the natural isotope existence is plotted at the three micro meter depth positions. These isotopic distribution changes occurred mainly within the layer of most outer surfaces in one micro meter and their ratios approached into normal values toward the inner bulk layer. In Figure 4, for Pd is shown to have large shifts in abundances. Their concentrations are represented with the ratio to Pd106 which exists in natural as most abundant. Their atomic concentration increased with depth. This means the concentration of other deposition relatively decreased. Especially, amounts of Pd104 and 110 are higher value than the natural ones that values are shown at three micro meters depth in the figure.

4. Discussion

It can hardly be imagined that all of the elements found were impurities in electrolyte, electrode or cell. Even if we suppose that all impurities in the system accumulated in the cathode, the amount would be 10 to 100 times smaller than the total amount we detected. Furthermore, it is simple impossible to explain the shifts in the isotopic distribution. Therefore, it must be concluded that some novel reactions occurred, resulting in the reactants which were found abundant in the electrolyte and electrode material. We assume the cathode palladium was the starting material for these reactions, but it is possible that impurities and other cell components such as Li, D_2O, Pd, Pt, K, Na, Ca, B, C, Ag and Fe may have provided the starting material for the nuclear reactions. It must be admitted that these reactions have no solid, detailed theoretical basis yet, but in broad terms this can explain most of the elements which were observed. One may also imagine that as such transmutation reactions were presumably taking place during the electrochemical process, they are likely to be connected with other phenomena such as hydrogen embrittlement and local corrosion.

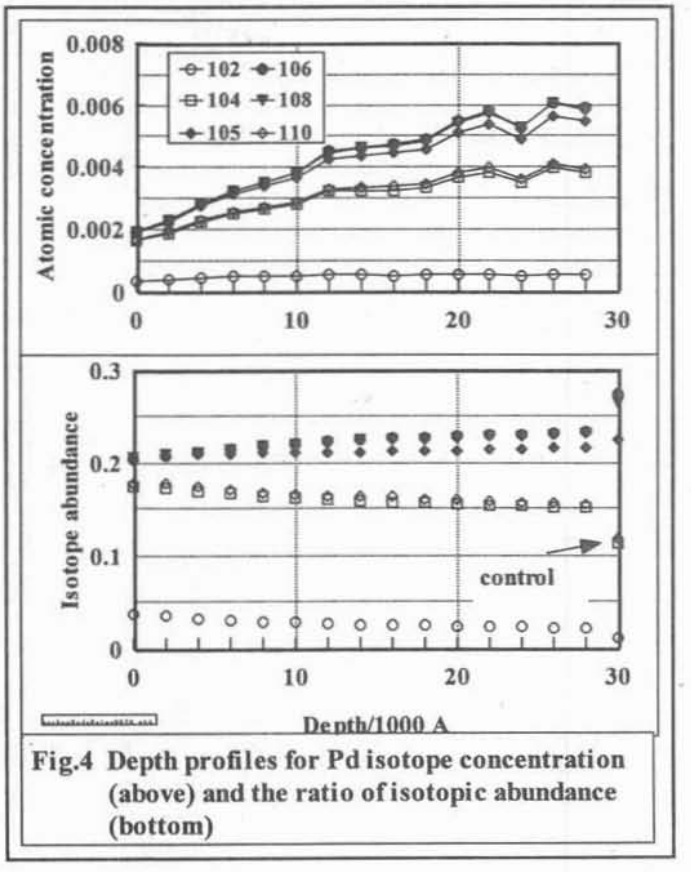

Fig.4 Depth profiles for Pd isotope concentration (above) and the ratio of isotopic abundance (bottom)

5. References

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2. Tadahiko Mizuno, Tadashi Akimoto, Kazuhisa Azumi and Michio Enyo, "Diffusion Rate of Deuterium in Pd during Cathodic Charging", Denki Kagaku 60, no.5, 405 (1992)