

ISOTOPIC CHANGES OF THE REACTION PRODUCTS INDUCED BY CATHODIC ELECTROLYSIS IN Pd

Tadahiko Mizuno¹, Tadayoshi Ohmori², Michio Enyo³

ABSTRACT

It was confirmed by several analytic methods that reaction products with mass numbers ranging from 6 to 220 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 of many elements ranging from hydrogen to lead. Isotopic distributions for the produced elements were radically different from the natural ones.

INTRODUCTION

Since the announcement that, successfully a large amount of heat generation had occurred by simple electrolysis in a heavy water solution using a Pd electrode, nuclear reactions in a solid electrode at ordinary temperatures have been reported by many experimenters. Especially, the phenomena attracts much attention because of the generation of energy. However, this phenomenon is still not well accepted among researchers because of poor reproducibility and control and moreover the lack of the knowledge for the mechanism. What is urgently needed now is to obtain precise and quantitative relationships between potential nuclear reactions and their corresponding reaction products.

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 claims could be upheld if the reaction mechanism consisted of proper theories. However, there is no proof that the conventional mechanism explains the observed reactions.

It is difficult to detect the emission of radiation and radioisotope if the mechanism is different from the standard 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. For example, natural chromium is 4.3% Cr50, 84% Cr52, 9.5% Cr53 and 2.4% Cr54. But the chromium found in the cathode was 14% Cr50, 51% Cr52, 2.4 % Cr53 and 11 % Cr54. Natural isotopic distribution varies by less than 0.003% for chromium. We represent that the reaction mechanism

¹ Dept. Nucl. Engr., Facul. Engr., Hokkaido Univ., Kita-ku, North 13, West 8, Sapporo, 060 JAPAN, Tel:81-11-706-6689, Fax:81-11-706-7835, E-mail: mizuno@hune.hokudai.ac.jp

² Catalysis Research Ctr., Hokkaido Univ., Kita-ku, North 11, West 10, Sapporo, 060, JAPAN, Tel:81-11-706-2620, Fax:81-11-709-4748

³ Hakodate Nat. Col. Technol., Tokura-cho 14-1, Hakodate, 042, JAPAN, Tel:81-138-59-1376, Fax:81-138-59-4410, E-mail:presiden@cc.hakodate-ct.ac.jp

was completely different from the accepted 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].

EXPERIMENTAL

Palladium rods used were of high purity (99.97% min.) supplied by Tanaka Noble Metals, Ltd. Impurities in the sample were as follows; B: 110, Si: 10, Ca: 9, Cr: 10, Cu: 6, Ti: 5, Ag: 44, Mg: 1, Pt: 20 and Au: 23 ppm, respectively. Nothing more was detected by atomic absorption photospectroscopy. The Pd rod was evacuated in a 5×10^{-5} Torr of vacuum at 473 K for 6×10^5 sec to begin. Heavy water was supplied by Showa Denko, Ltd. It is 99.75% pure and includes 0.077 micro Ci/dm³ of tritium. The heavy water was purified once in a quartz glass distiller. Reagent grade lithium hydroxide was obtained from Merck, Ltd. Impurities in the reagent were specified as follows; Li₂CO₃: 2% max, Cl: 0.05%, Pb: 20, Ca: 200, Fe: 20, K: 200 and Na: 200 ppm and the other impurities were under detectable limit by the atomic absorption analysis. The anode and recombiner catalyst were, respectively, a high purity (99.99%) Pt plate and a Pt mesh. The Pt metal is specified to contain impurities as follows; Rh: 18 ppm; Si, Cr and Pd: 2 ppm; Au, Ag, B, Ca, Cu and Fe: less than 1 ppm. Other impurities were under the limits of detection.

Electrolysis was performed in a closed cell made of stainless steel. The cell has another inner cell made with 1 mm thick Teflon; the height and diameter are 20 cm and 7 cm, the volume is 770 cm³. The details have been described elsewhere [2]. Before the electrolysis experiment, electrolyte was pre-electrolyzed with other Pt mesh electrodes at 1 A and 150° C for 6×10^5 s (7 days) in the cell with the upper cover closed. After, the Pt electrode was removed and the Pd rod sample was attached at the terminal part. Electrolysis experiments were performed at a current density of 0.2 A/cm², total current of 6.6 A ($33 \text{ cm}^2 \times 0.2 \text{ A/cm}^2$) at 105°C for 2.76×10^6 s (32 days). After the electrolysis rod was washed by Mill Q water (Mill Q-lab; Japan Millipore Ltd.) and covered by a Teflon tube. The analysis samples were prepared by cutting the rod into 1 cm lengths and again cut into two half-moon-shaped masses by a diamond cutter. The sample electrodes were supplied to element detection after removing 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). EDX measurements were done by 20 keV of electron irradiation and varying the scanning area; the energy spectra were measured by Silicon Li drift detector. The range of analysis energy was zero to 20 keV; the interesting range was divided into 1024 channels of energy width. The resolution power of energy was 150 eV at 5.9 keV, practically that was 200 eV. Peaks were calibrated using high purity of C, Al, Si, Ti, Cr, Mn, Fe, Co, Zn, Sr, Nb, Mo, Pd, Ag, Sn, Ce, Hf, W, Pt, Au and Pb. AES analyses were performed to obtain the depth distribution of the elements by ANELVA AAS-200; the ion irradiation energy and the current are 3 keV and 2.5 A respectively. EPMA-8705 of Simazu LTD. was used to obtain the elements distribution on the samples. The IMA-3 of Hitachi Co. analyzed for SIMS measurement; O₂⁺ ions were irradiated on the sample as a spot diameter of 100 micro meters square having primary energy of 12 keV and 100 nano-Amperes of ion current. Resolution for the mass was $m/e=10,000$. Mass numbers were calibrated by high purity metals of Li, B, C, Al, Si, Ca, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, Sr, Nb, Mo, Pd, Ag, Cd, Sn, Ce, Hf, W, Os, Pt, Au and Pb; no isotopic changes existed in the calibration measurements over the natural deviations.

RESULTS

EPMA analyses were done first to obtain the elements' distributions on the sample surface. There were no detectable depositions on the Pd before electrolysis. And there also no detectable depositions on the surface in case of the current density kept under 0.2 A/cm². Moreover, no deposition was seen if the current density was kept up to 0.2 A/cm² with no change. 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 present and differed more than 3 times at the surface place compared with

C, Cl, Si, Mn, Co, Ni, Zn and Sn which existed rather uniformly. These changeable elements were also fluctuating with sample lot as 3 times deviations. It means that uncontrollable factors such as surface conditions are of main importance for the reaction.

Depth distributions of the several elements were decided for one micro meter by the AES measurement. Carbon and Oxygen atoms occupied 40 atomic percentages respectively and Cr, Fe, Ca and Cd elements were detected as few atomic percentages; Oxygen concentration once increased in the bulk layer about 65% and gradually decreased with depth; carbon atom concentration gradually decreased with depth but other elements kept almost uniform.

Several elements were detected in the Pd electrode by the EDX method; the measurements were taken to determine the rough concentration for the elements because in the SIMS measurement the mass peaks have a possibility to contain other molecular peaks. Fig. 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 clearly observed. The EDX analyses were repeated on various places of the sample; the deviation for the EDX counts sometimes reached to 10 times in various places. This change was depended on the scanning area. Deviation was 10 times for 10 micro meter square scanning areas but it decreased 2 times for 100 micro meters square. It can be suggested that the reactions had occurred in quite restricted places.

The amounts for the evolved elements were finally estimated as detected by SIMS measurement; a typical result obtained for the Pd surface electrolyzed by 0.2 A/cm² of current density is shown in Fig. 2. Mass number for 1 to 210 were detected and most of the masses have more than several hundred count numbers; it can be estimated for the counting errors were within few percentages. However, while there were only 1, 12, 16, 102, 104, 105, 106, 108 and 110 of mass number peaks existed in the before-processed Pd sample. 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 by the 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 each mass. Fig. 3 shows the factor when the O₂⁺ ion was irradiated for the analysis. 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 above mentioned procedures. The ratios were shown in Fig. 4 normalized with the total mass set as one. Typical counts by EDX and SIMS ranged from 10² to 10⁶ 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 numbers: 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 numbers 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 ratios can be considered that the many gas atoms may be released and successively contribute to the counting rate from the spot place heated up by ion bombardment.

AES and SIMS measurements were also made after bombardment by Ar⁺ and O₂⁻ ions, thus removing surface layers, but the element concentrations at 1 micro meter below the electrode surface were decreased to 60%, 10% and 1% at the surface at 5 microns depth and 10 microns depth. Many holes and cracks having 1 to 10 microns of opening size were observed in the bulk layer; The same elements, having almost the same concentration, were also found at the inner surface. The SIMS analysis showed other elements; As, Ga, Sb, Te, I, Hf, Re, Ir, Br and Xe. These elements, except Xe, were difficult to detect 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 such gas atoms easily escape during 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 Fig. 4 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.

No peak except Pd has been observed after pre-electrolysis on the Pd surface. Pt and 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 1 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 plotted in Fig.5. Here, the total impurities except for Ca are less than the deposition amounts; especially, Mn, Ti, Cd and Hg were not present as an impurity.

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. 6 for an example; the concentrations except Cr52 decreased exponentially with depth but Cr52 has a peak at 0.5 microns and they showed large shift in isotopic ratios: all the ratios are shown with the ratios of Cr52 which exist in natural most abundant. The reasons that we represented such the existence ratios for isotopes instead to show the results calculated by the ratios to the total existence were to avoid misunderstanding to some change of the isotopic distribution probably caused from mixing with others elements; for this chromium case the mixing may be raised with the existence of iron atom if there are Cr and Fe, may be Cr54 and Fe54 peaks overlapped together even if we employ such the high resolution SIMS measurement of $m/e=10000$ still the mass difference between Cr54 and Fe54 is only 0.0000135. That means it is impossible to divide their two isotopes by the SIMS measurement having the resolution power. 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 3 microns depth position. These isotopic distribution changes occurred mainly within the layer of most outer surface in 1 microns and their ratios approached normal values toward the inner bulk layer.

In Fig. 7, 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 3 microns depth in the figure.

In Fig. 8, Lead was seen. The Pb fluctuations are larger than Cr and Pd because their counting rates were one or two orders less. Their concentrations are represented with the ratio to Pb208 which exists in natural as most abundant. Their atomic concentrations show constant except 208 which increased with depth. The isotopic distributions for Pb206 and 207 are lower value than the natural ones that values are shown at 3 microns depth Figure 8.

Neutron intensity and the energy measurements were carried out simultaneously, in parallel. The neutron evolution rate was sporadic and weak, as previously reported [3], with levels of ~ 0.4 counts per second. No gases such as He, O_2 and Ar were detected during electrolysis experiment. Excess heat generation was less reproducible, but was detected and the value was the order of 10^7 joules [4].

DISCUSSION

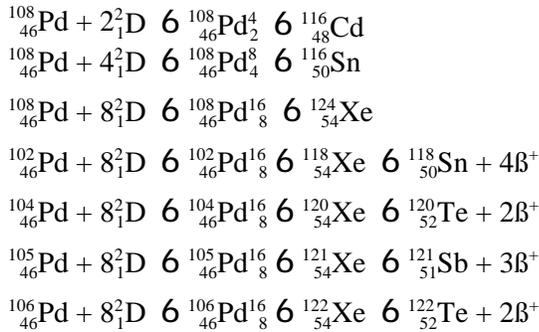
Essentially the same phenomenon was confirmed five times with high reproducibility at high cathodic current density, above 0.2 A/cm². Current density ranged from 0.2 to 0.6 A/cm². Different isotope distributions were obtained, depending on the current density. This will be described more fully in an upcoming paper. 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. Hence, 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, D₂O, Pd, Pt, K, Na, Ca, B, C, Ag and Fe may have provided the starting material for the nuclear reactions.

The palladium surface became rough and porous after several weeks of electrolysis, probably due to hydrogen attack. The current may have increased in such roughened surface areas, which would in turn cause a larger reaction and a higher concentration of the reaction products. Enyo [5-7] reported that the effective hydrogen pressure at the hydrogen evolving electrode depends upon the hydrogen atom recombination process which follows the water discharge process; the total hydrogen overpotential O_t is represented by addition with two of components by $O_t = O_1 + O_2$; where O_1 and O_2 are arisen from discharge and recombination process. The O_1 is a slow step compared with O_2 of Tafel step for the normal palladium surface. But the claim can be held at a completely flat and uniform surface. The values of O_1 and O_2 are only estimated as averaged over the whole surface. The division of the overpotential between these two steps may be important. It was suggested that at Pd electrodes in alkaline solution the effective hydrogen pressure may rise as high as 10⁶ atm [8,9]. There may be further increases at local points on a heterogeneous surface. One may even speculate that the hydrogen isotope nucleus sometimes closely approaches the medium nucleus [10]. An estimate by Nernst Equation indicates that 5 x 10⁴ atm of pressure may be realized at 140 mV of overpotential at the electrolysis current density of 0.2 A/cm² on a flat palladium surface [11]. Furthermore, the pressure distribution depends on the roughness of the surface, because local current density and the partition of overpotential components may vary with roughness.

There are several reports [12-14] of evolution of elements by electrolysis. However, only a few [15,16] demonstrated shifts in isotopic abundance. For a gold electrode [17], which also generated various elements by cathodic electrolysis, different isotopic distributions were seen. In this case also, the reaction sites were unevenly distributed on the surface. Typically, the active points may have occupied about 10⁻⁶ cm² areas and numbered 10⁴ to 10⁵ per cm² at the surface. Thus, the current is likely to be concentrated at localized points 10 to 100 times higher than average. Moreover, the rate control may be changed at the parts by the condition of electrode surface, the recombination process becomes slow step compared with the discharge process; the over potential caused from the recombination process can be a considerable part of the over potential. Total cell voltage reaches sometimes more than 1 Volt at 0.2A/cm² of cathodic current density in alkaline solution. The major overpotential may be occupied by the discharge process at a flat and uniform part of the Pd surface, however, at the top of the Pd part where are rough surfaces, the major part of the overpotential may be occupied by recombination process. Then, the overpotential due to recombination process becomes more than 0.3V if the process occupied 2 times bigger than the usual if the current density reached more than 1V. We can conclude the statistical hydrogen pressure exceeds more than 10¹⁰ atm at the parts of Pd electrode.

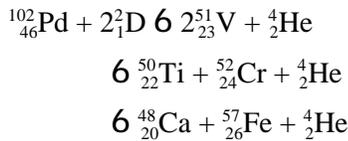
Hydrogen gas and the electrode atom are compressed and changed like a metal solution under the such high pressure. The distance between their atoms were approached and entered through the electron shield. The hydrogen atoms can occupy the orbit according their spin quantum numbers. More electrons take the higher orbit and it occurs some possibility that can induce proton and neutrons into enter heavy nucleus under the above mentioned conditions [18]; two, four, eight and ten hydrogen and deuterium atoms can enter at the moment in

the heavy nucleus. If the reactions start from palladium as the electrode material, fusion and fission may take place simultaneously. Several possible reactions might be considered:



Here, left-hand sides in the equations are the reactants, right-hand sides are the products and the middle are the intermediate states that are unstable due to containing excess protons and neutrons; the bottom subscript at the right-hand side of the intermediates represents the proton number and the top superscript represents the total nuclear number which is connected with nucleus as the hollow atom. It can be assumed that these particles will stay in a stable orbit according to their quantum spin number as the same concept with electron orbits. The connecting nuclei are caught into medium nuclei by the force of high external pressure.

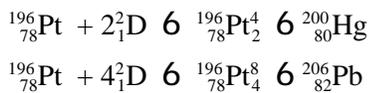
Other reactions have to be assumed because many light elements were observed. They may be as follows:



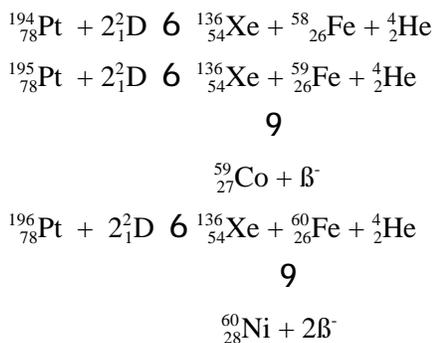
Generally, one can write the fission reactions as follows:



In the case of platinum deposited from the aug. electrode to the Pd electrode surface, some reactions may be involved, such as:



They may induce fission reactions as follows:



Atomic numbers of 20, 28, 50 and 82 are called magic numbers. Here, Xe136 nuclei are abnormally increased, and hence the number of neutrons changes to magic number 82, and especially stable nuclei are selectively produced. In this way, the isotopic distributions of the products after electrolysis can be explained in terms of the difference of binding energy of the nuclei.

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.

CONCLUSION

Anomalous evolution of various elements in palladium electrodes was confirmed after high current density cathodic electrolysis under high temperature and pressure for a long time. The following conclusions were drawn:

(1) The mass number of the evolved elements was distributed roughly in four groups: less than 20, 20 to 28, 46 to 54 and 72 to 82, with the amounts, respectively, 10 %, >50 %, 10 % and <5% compared to palladium.

(2) These evolved elements were found on the surface, and also in the bulk of the electrode in amounts 10 to 100 times smaller.

(3) **Many evolved elements accumulated in holes and cracks on the electrode which formed during electrolysis.**

(4) Some of the evolved elements have isotopic distributions drastically different from natural ones, especially for Cr, Fe, Cu, Zn, Br, Xe, Pd, Cd, Hf, Re, Pt, Ir and Hg. The layer showed large change of isotopic concentration in 1 micro meter of the Pd surface.

(5) Gaseous Xe was noteworthy because it was so abundant. Xenon is particularly unlikely to be a contaminant because metals do not absorb noble gases, and because the cathode was degassed in a vacuum at 473°K for 20 hours.

(6) The elements in the bulk layer changed in concentration with depth and showed shifts of isotopic distribution.

(7) Light elements such as O, C, Ca, Na, Mg and Al showed small isotopic shifts.

(8) Ni and Co were also confirmed but their isotopic distribution could not be measured because their SIMS peaks overlapped with those of other elements and isotopes.

(9) The isotopic distributions of Pd and Pt were also shifted.

(10) We conclude that nuclear reactions must have occurred during the electrochemical process.

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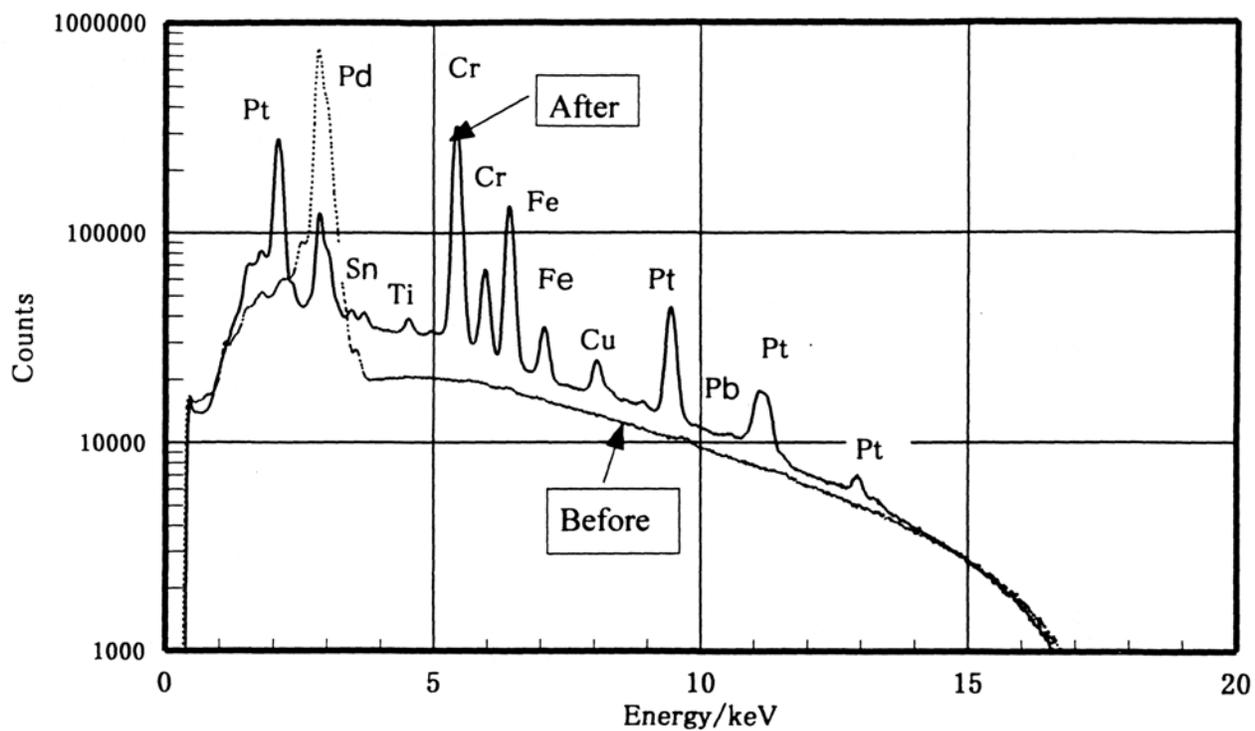


Fig. 1. EDX spectra from the Pd rod before and after the electrolysis.

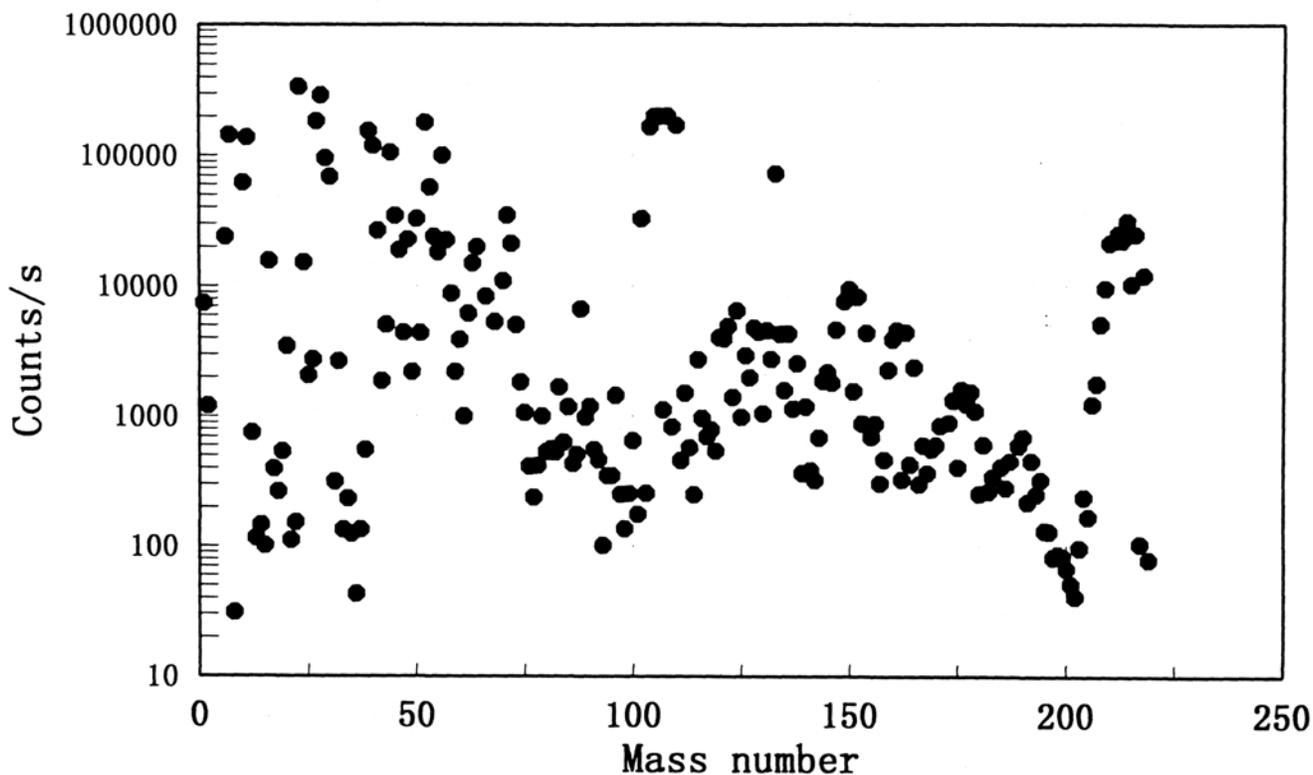


Fig. 2. Count rates for Pd electrode electrolyzed in the heavy water solution by SIMS measurement. (Electrolysis conditions; 1.0 mol LiOH-D₂O, 378 K, 0.2 A/cm² for 2.76 10⁶ s)

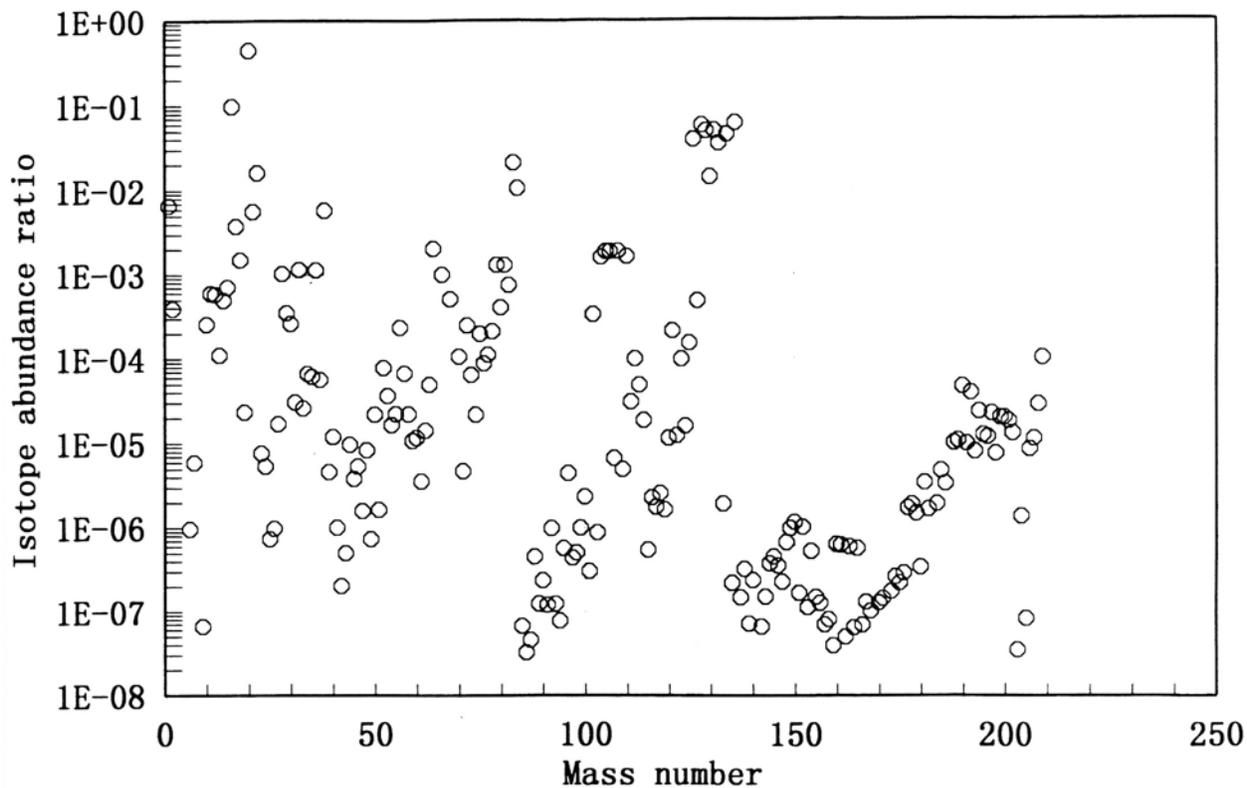


Fig. 4. Estimated mass spectrum for Pd electrode surface.

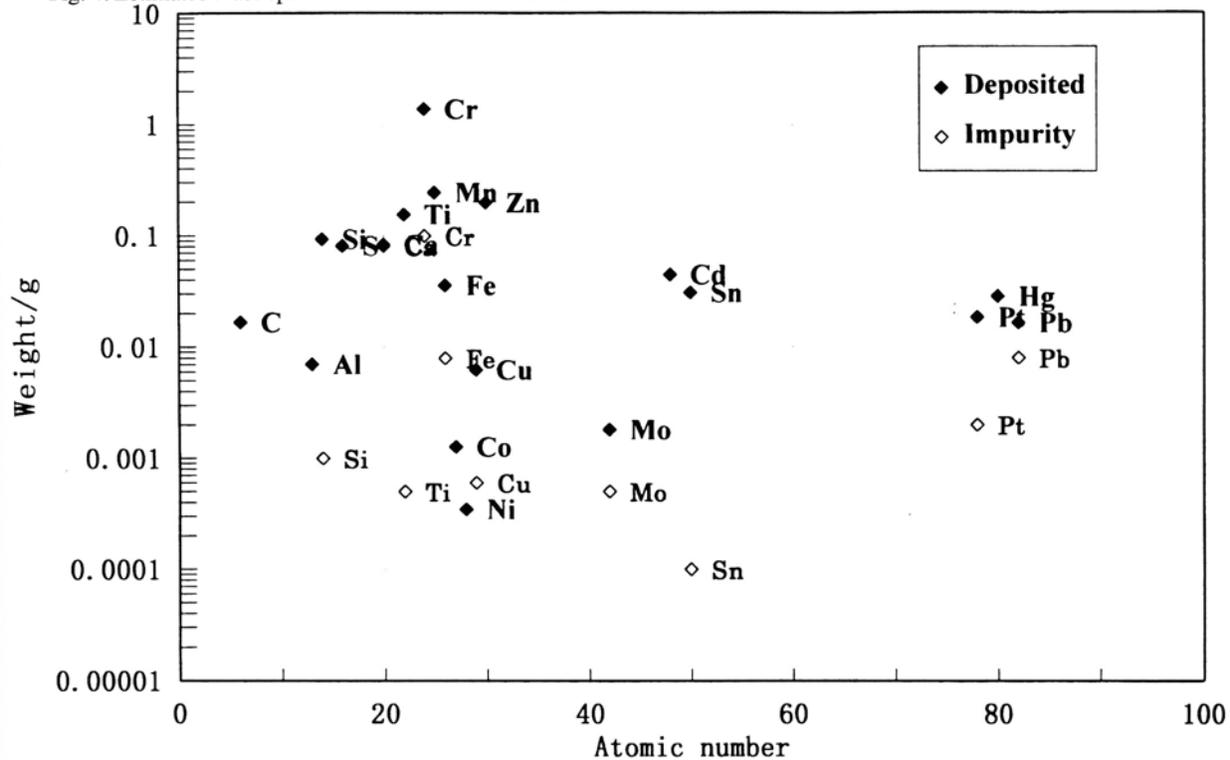


Fig. 5. Total amounts of deposited elements on the Pd and impurities estimated from the sample and solution.

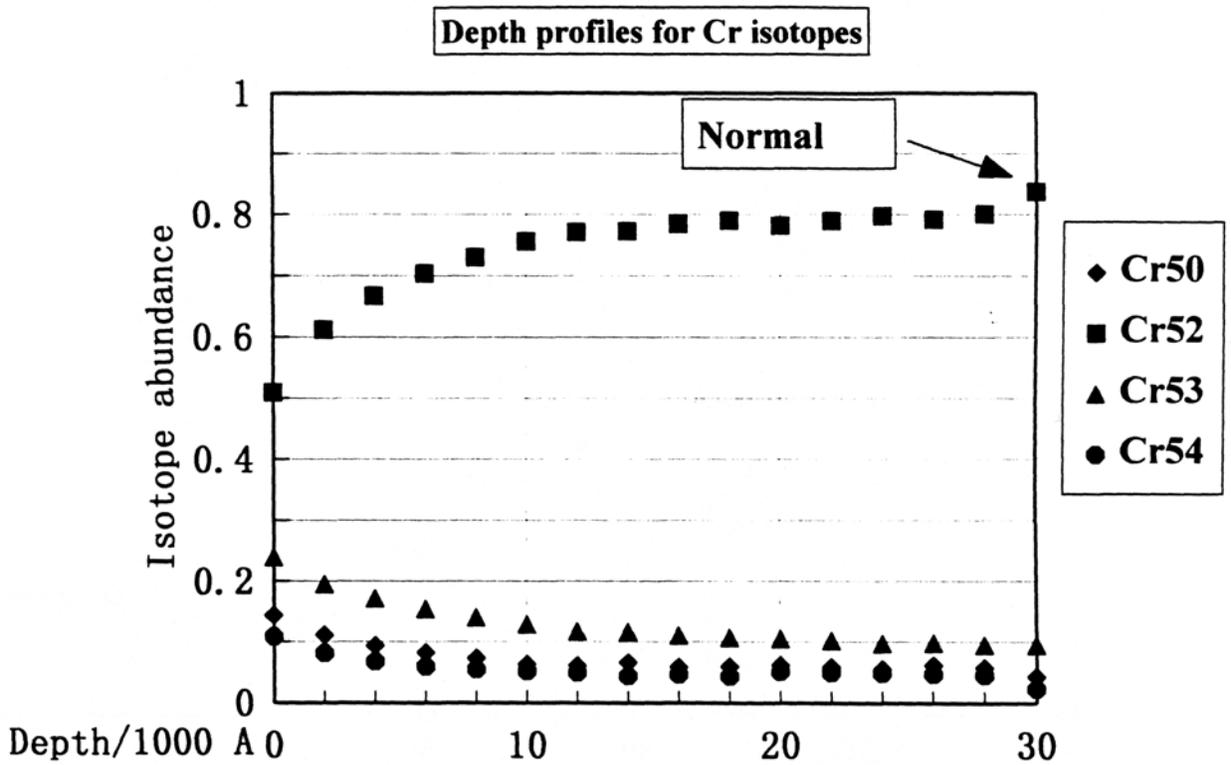
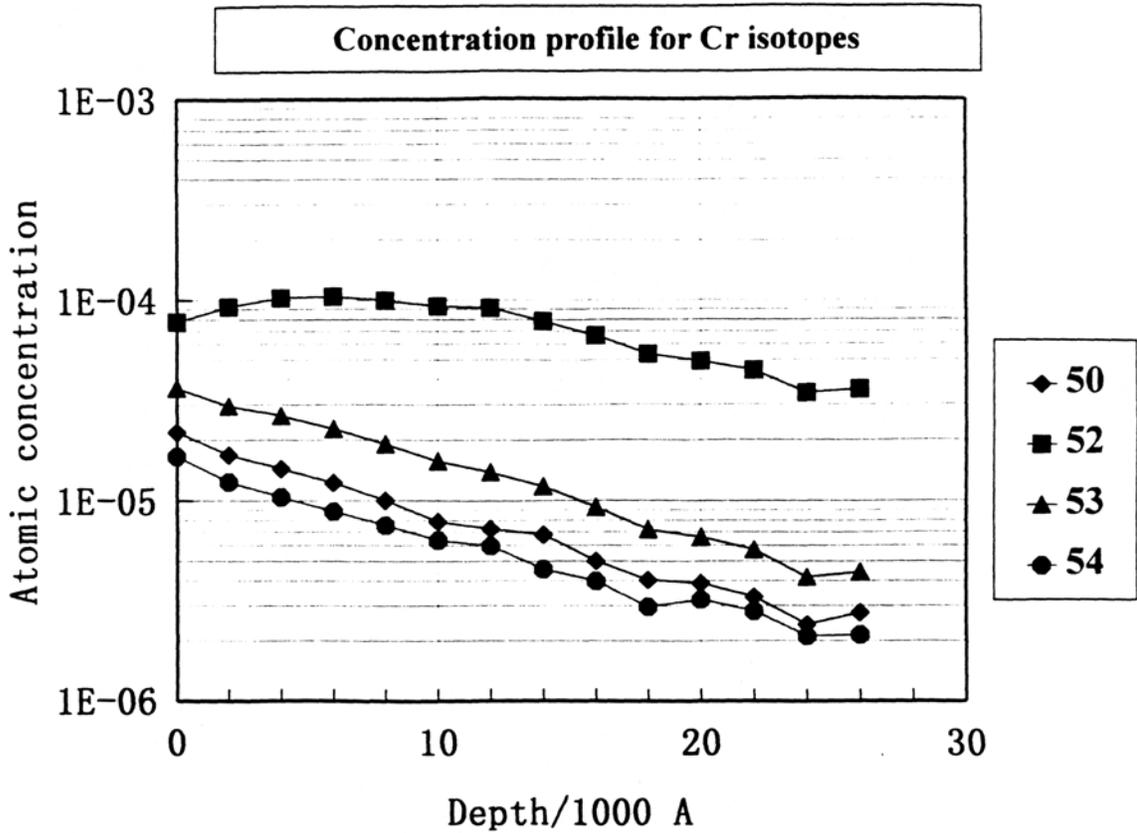


Fig. 6. Profiles for Chromium isotope concentrations (above) and the depth profiles for the isotopic ratios with Cr52 (bottom) in Pd.

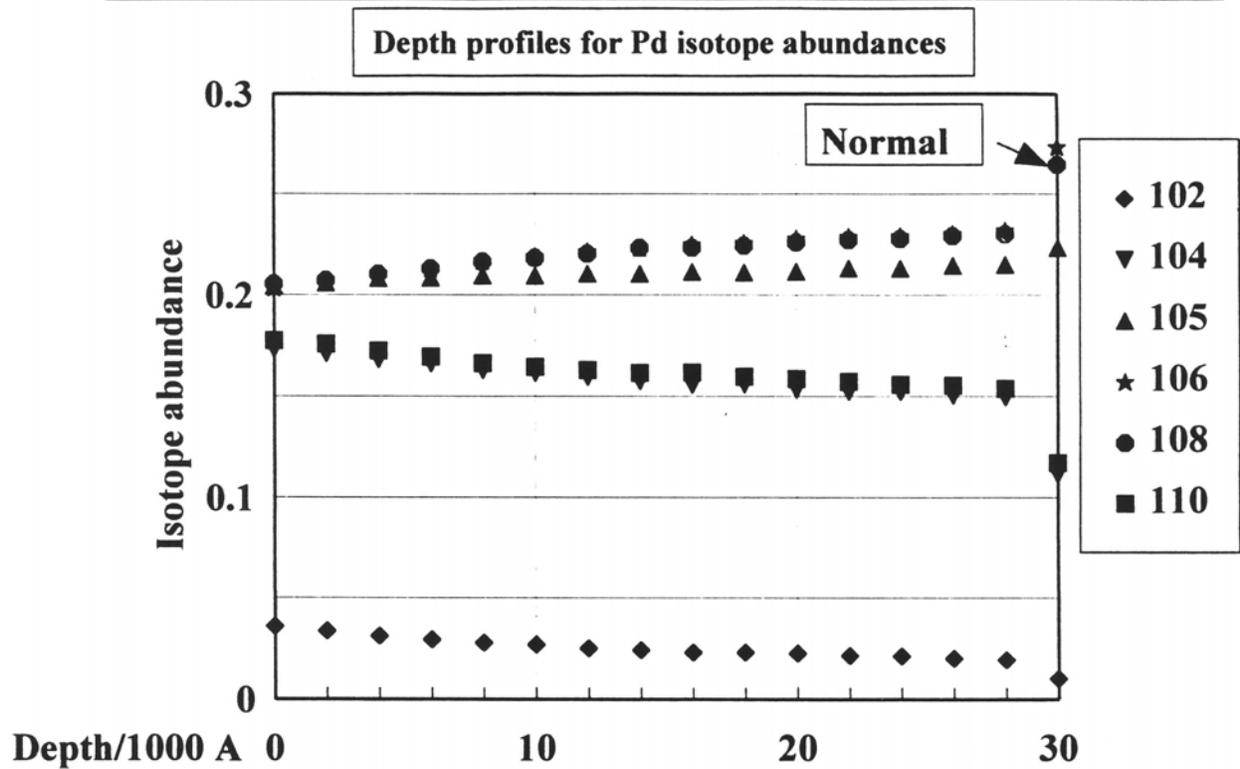
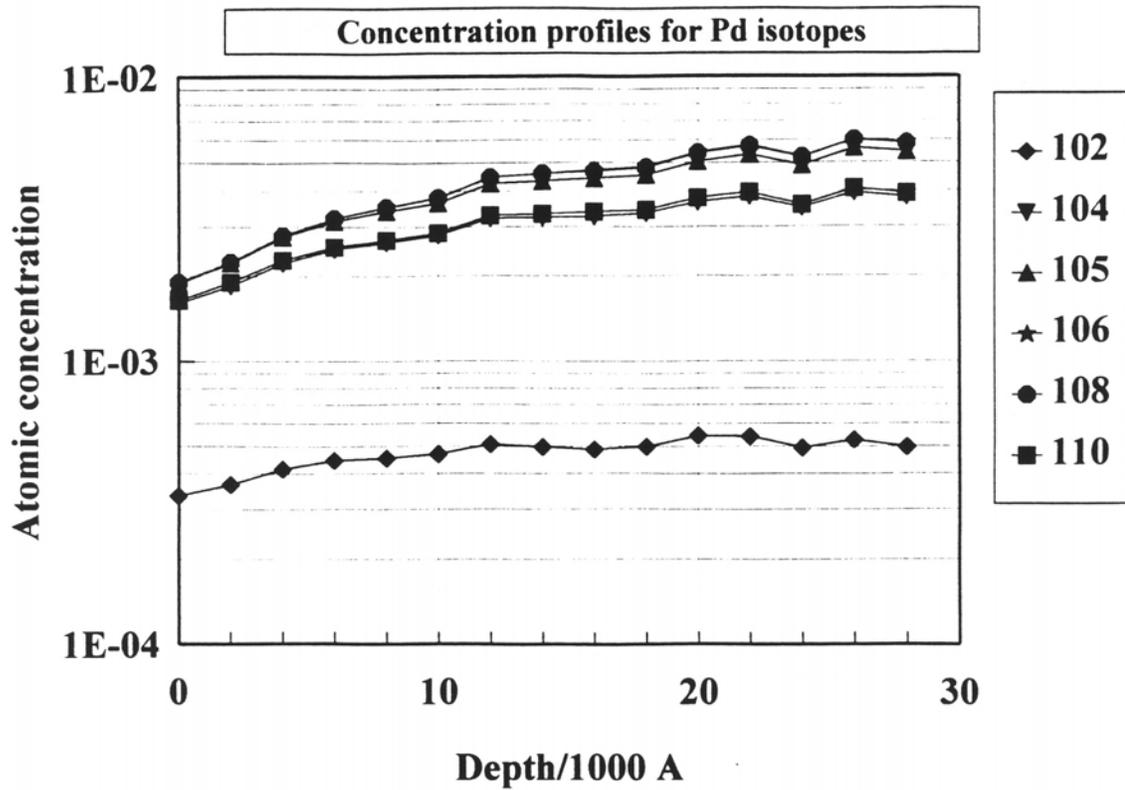


Fig. 7. Profiles for Palladium isotope concentrations (above) and the depth profiles for isotopic ratios with Pd106 (bottom) in the sample as compared with natural isotopic distribution.

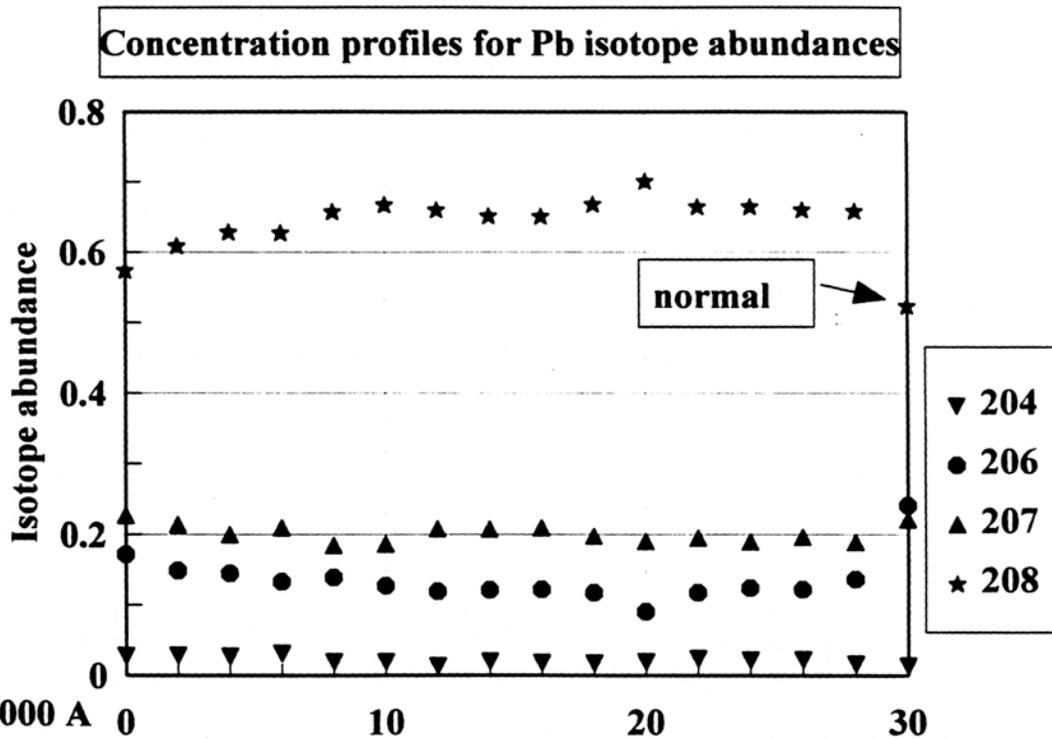
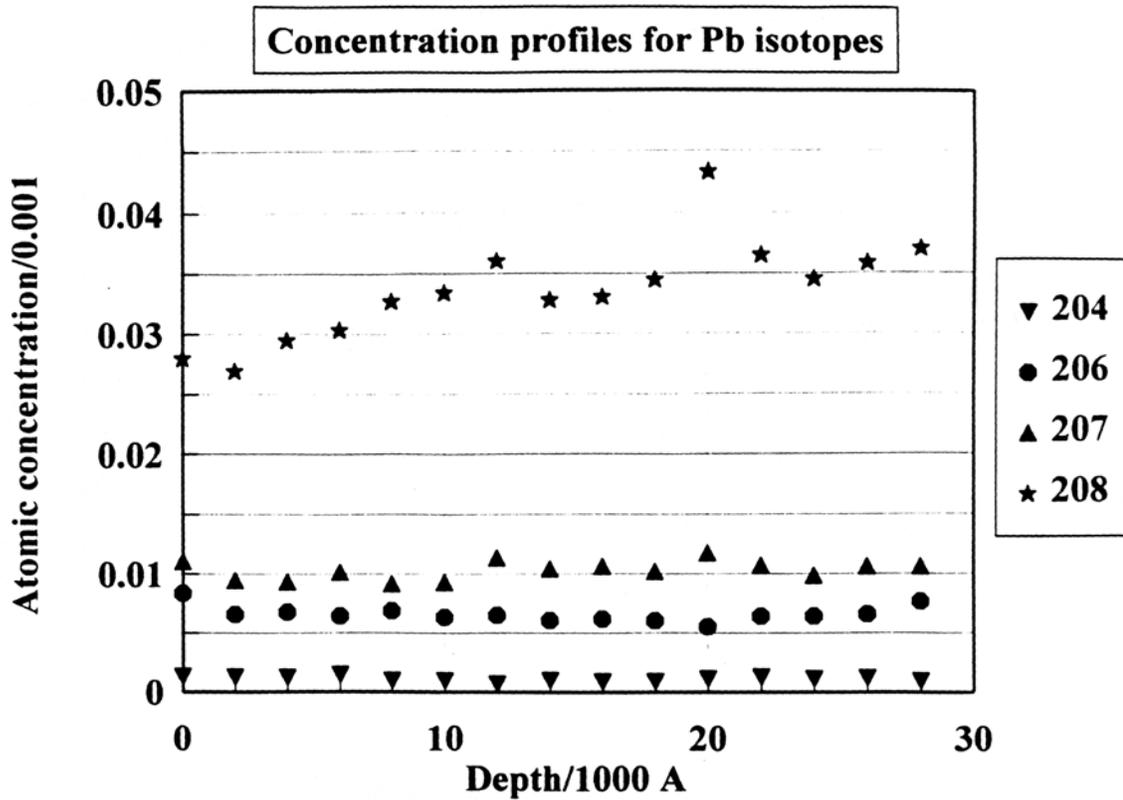


Fig. 8. Profiles for Lead isotope concentration (above) and the depth profiles for isotopic ratios with Pb208 (bottom) in the sample as compared with natural isotopic distribution.

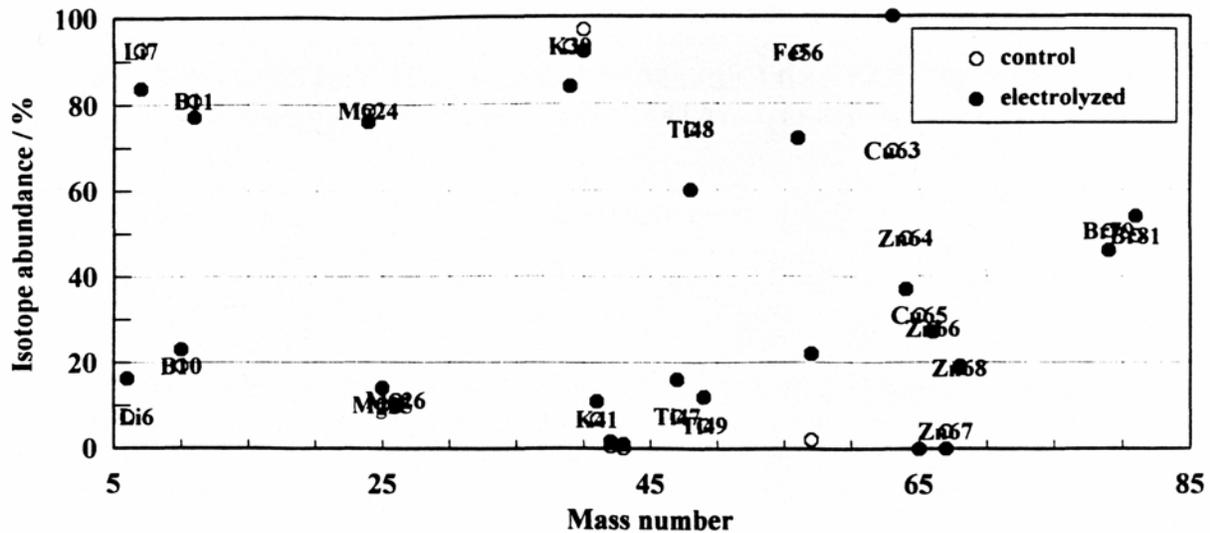


Fig. 9. Changes in isotopic abundance for deposition on Pd

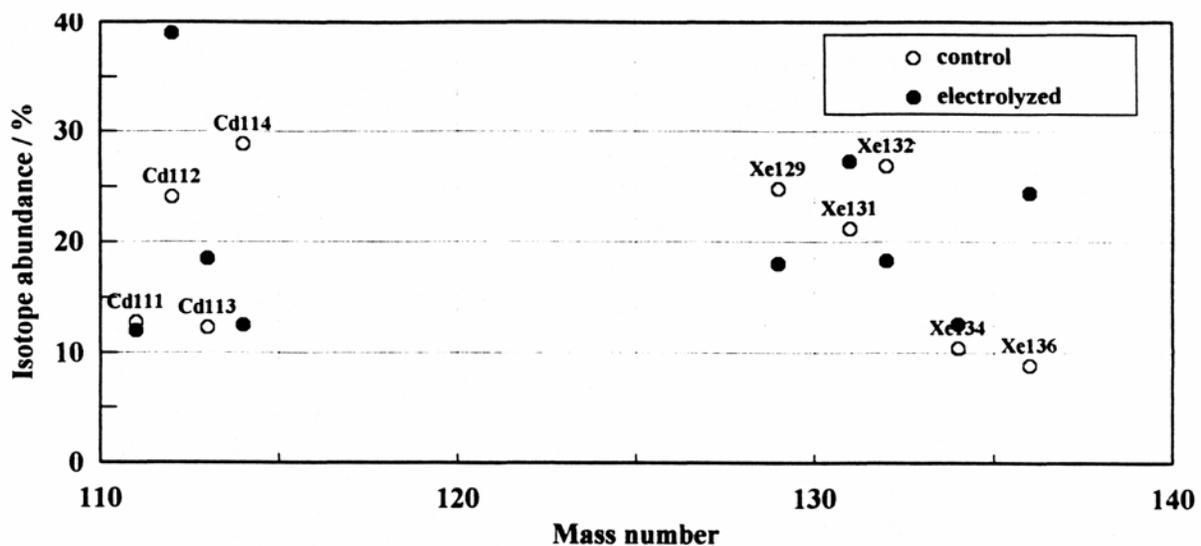


Fig. 10. Changes in isotopic abundance for deposition on Pd

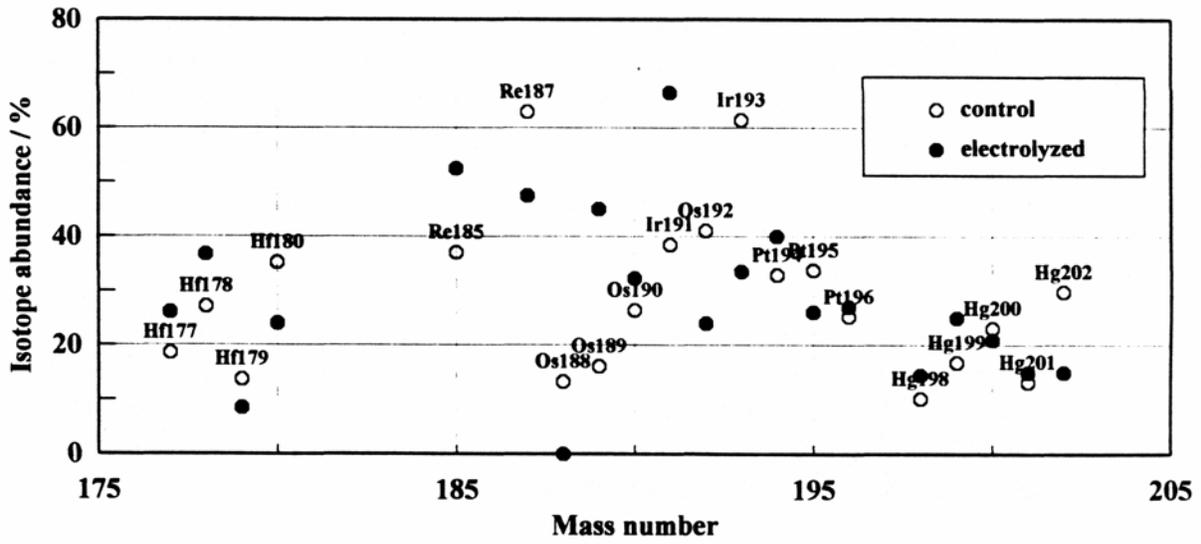


Fig. 11. Changes in isotopic abundance for deposition on Pd