Isotopic changes of elements caused by various conditions of electrolysis

Tadahiko Mizuno

Department of Engineering, Hokkaido University

Hideo Kozima

Cold Fusion Research Laboratory

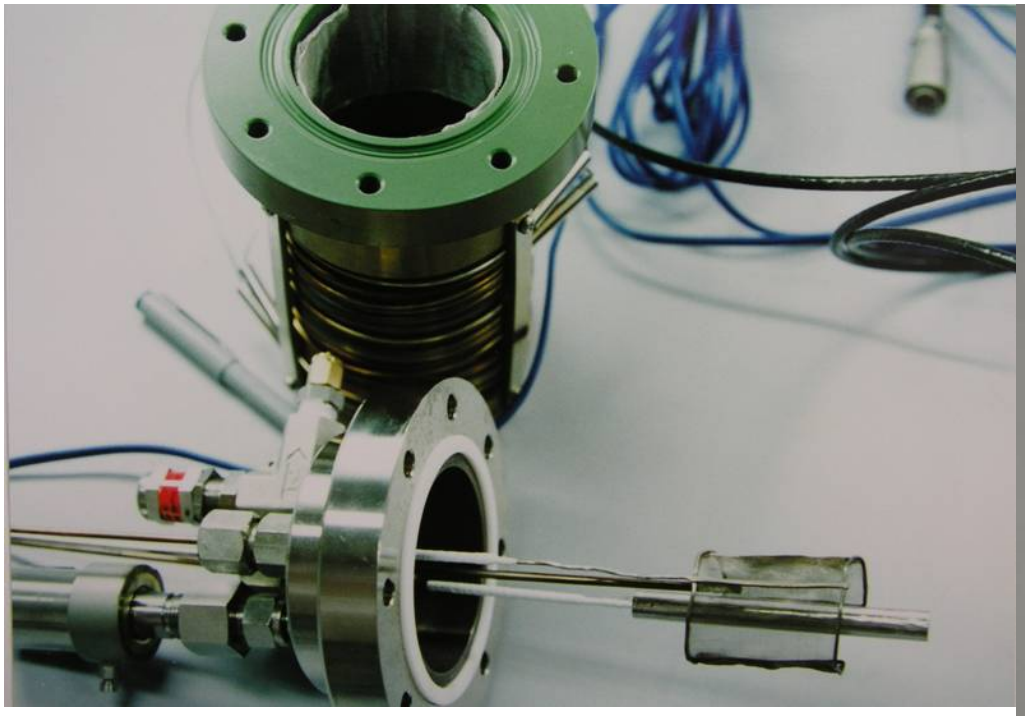
The appearance of elements on palladium electrodes after electrolysis in various conditions were confirmed by several analytic methods. Mass numbers as high as 208 corresponding to elements ranging from hydrogen to lead were found, and the isotopic distributions of many of these elements were radically different from the naturally occurring ones. Changes in element distribution and in their isotopic abundances took place during electrolysis in both heavy and light water, whether or not excess energy was generated. If the transmutation mechanism can be understood, it may then be possible to control the reaction, and perhaps produce macroscopic quantities of rare elements by this method. In the distant future, industrial scale production of rare elements might become possible, and this would help alleviate material shortages worldwide.

Contents

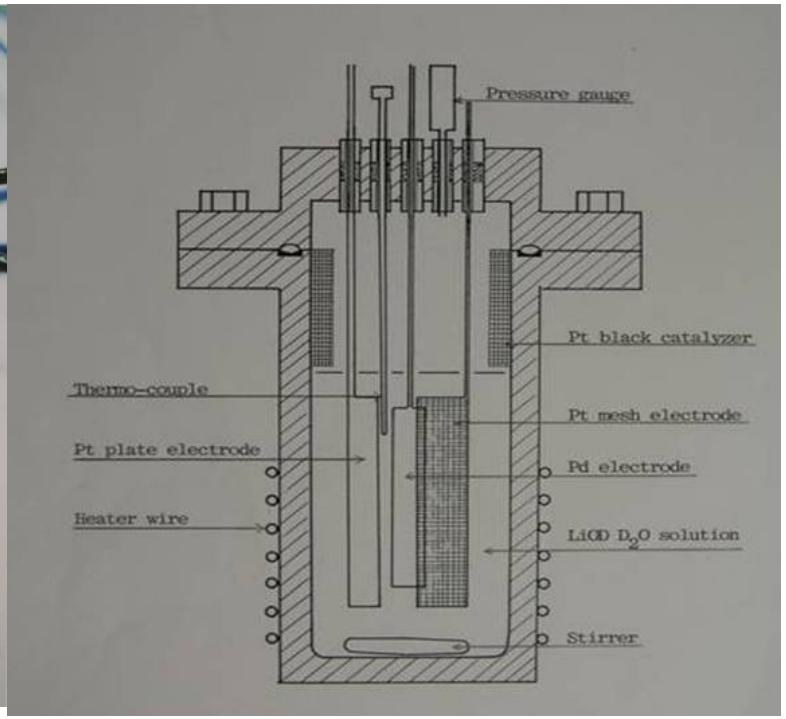
- Low energy nuclear reaction
- Various condition: 1cm diameter, 10cm length Pd rod, 0.2A/cm^2, electrolyzed in 0.5M- LiOH（LiOD) solution, 100℃, 500-800 hours.
- Excess heat
- No excess heat
- Heavy water
- Light water
- Element distribution

This work presents evidence that a nuclear reaction takes place during electrolysis that produces isotopically changed elements on the cathode surface. These elements are generated by a mechanism that does not induce any detectable radiation. The anomalous isotopic abundances of these elements show that they do not come from contamination. We suggest that the operative nuclear mechanism is completely different from any known nuclear reaction.

Photo and sketch of the closed cell



Cell and cover

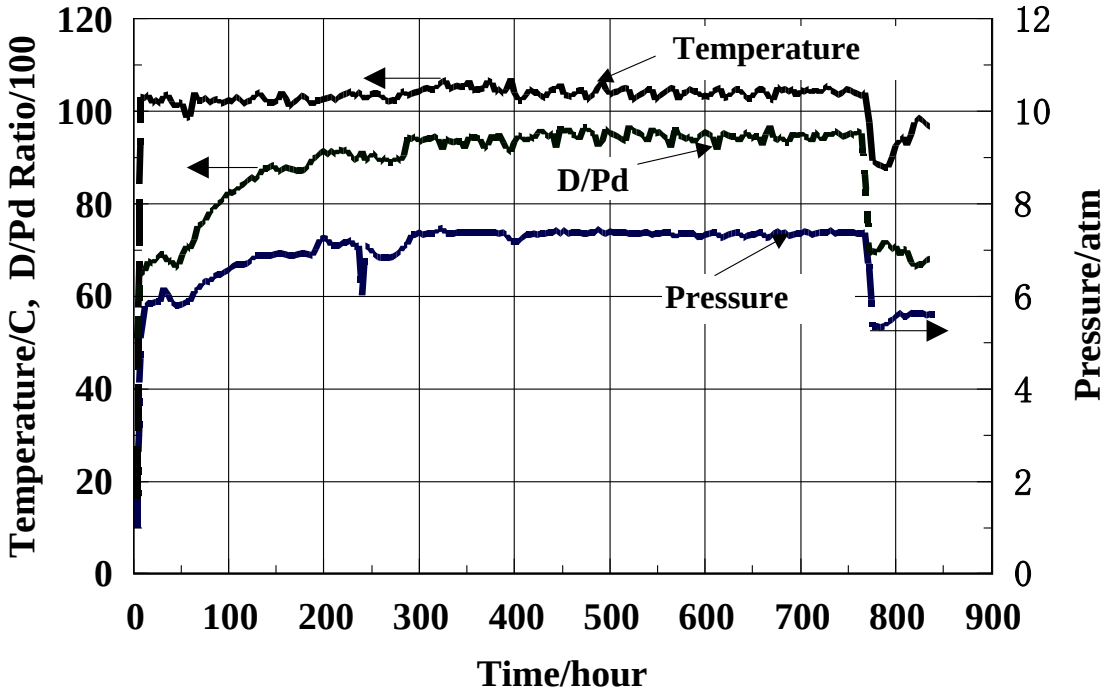


Cross sectional sketch

All electrolysis was performed in a closed cell made from a stainless steel cylinder. The cell has an inner Teflon cell made with 1-mm thick wall and 1 cm thick upper cap; the inner height and diameter are 20 cm and 7 cm, the volume is 770 cm^3. Before electrolysis electrolyte were pre-electrolyzed using another Pt mesh electrode. After that the Pt electrode was removed and the palladium rod sample was connected to the electrical terminal. Electrolysis experiments were performed with the current density of 0.2 A/cm^2 for 20 days at 105 ℃.

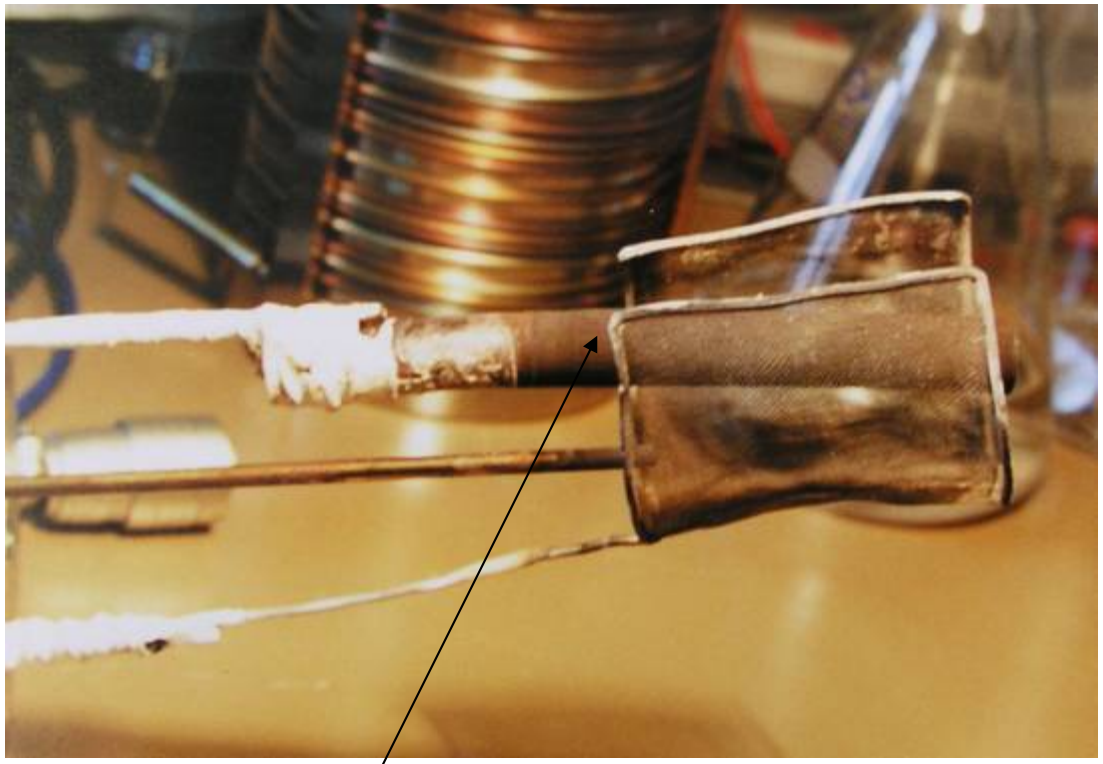
Changes of Temperature, D/Pd ratio and pressure;

1cm diameter, 10cm length Pd rod, 0.2A/cm^2, electrolyzed in 0.5M LiOD solution



The figure shows a typical time variations of current, temperature and D/Pd ratio during electrolysis in heavy water solution. In the experiment shown here, some excess heat was generated after electrolysis, as indicated by the temperature rise of the electrolyte after the electrolysis was stopped at 766 hours. The phenomenon is very anomalous since the temperature would be expected to decrease after shutting off the input power. All electrolysis was carried out at 0.2 A/cm^2 or a total current of 6.6 A for 32 days at around 100° C.

Photo of the cathode and anode after electrolyzed

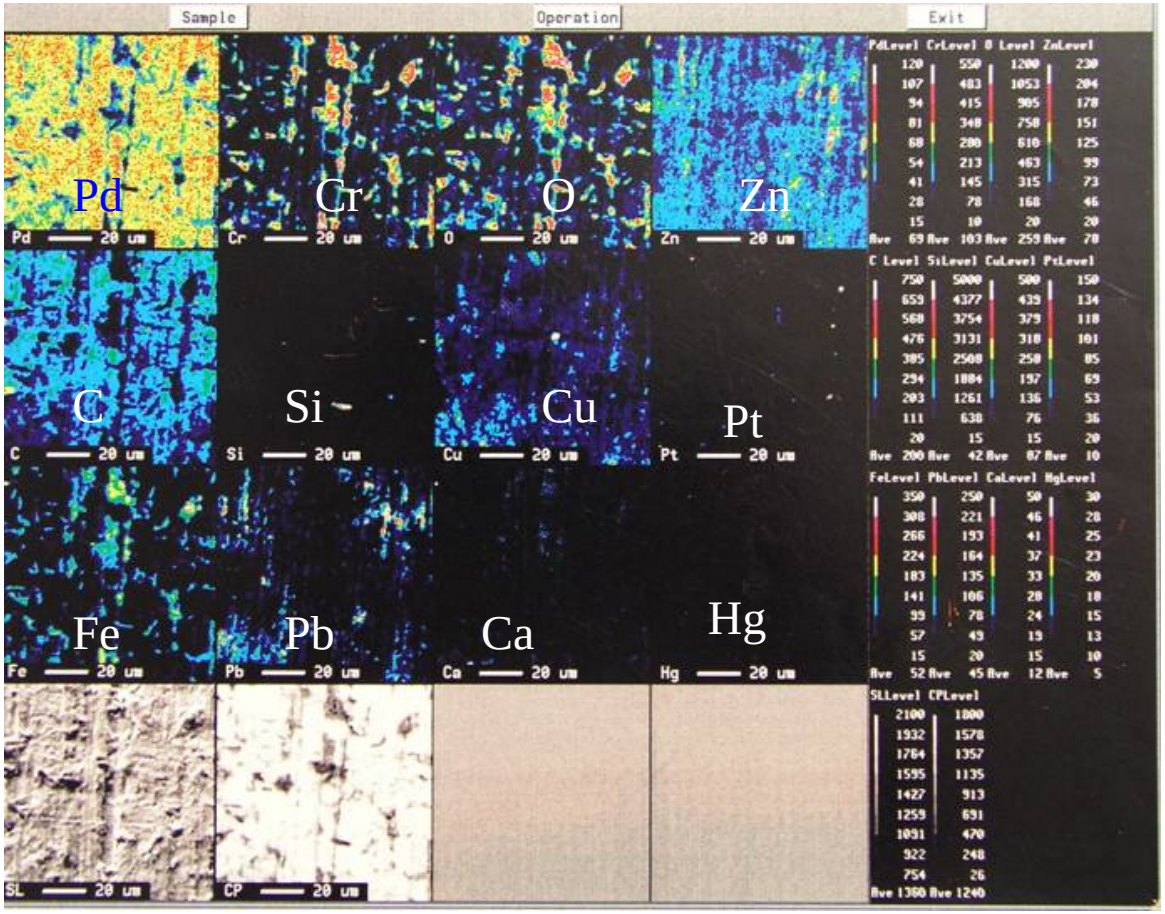


Many elements were deposited on the electrode surface in an irregular distribution. The concentrations varied depending on the parameters of the electrolysis. The elements that have been detected on the samples were Ca, Ti, Cr, Fe, Ni, Cu, Zn, Pb and so on. The amount of each element varied by the sample lots. This may mean that some factors such as surface conditions play an important role in the reaction.

Photo of SEM, EDX and ICP mass analyzer

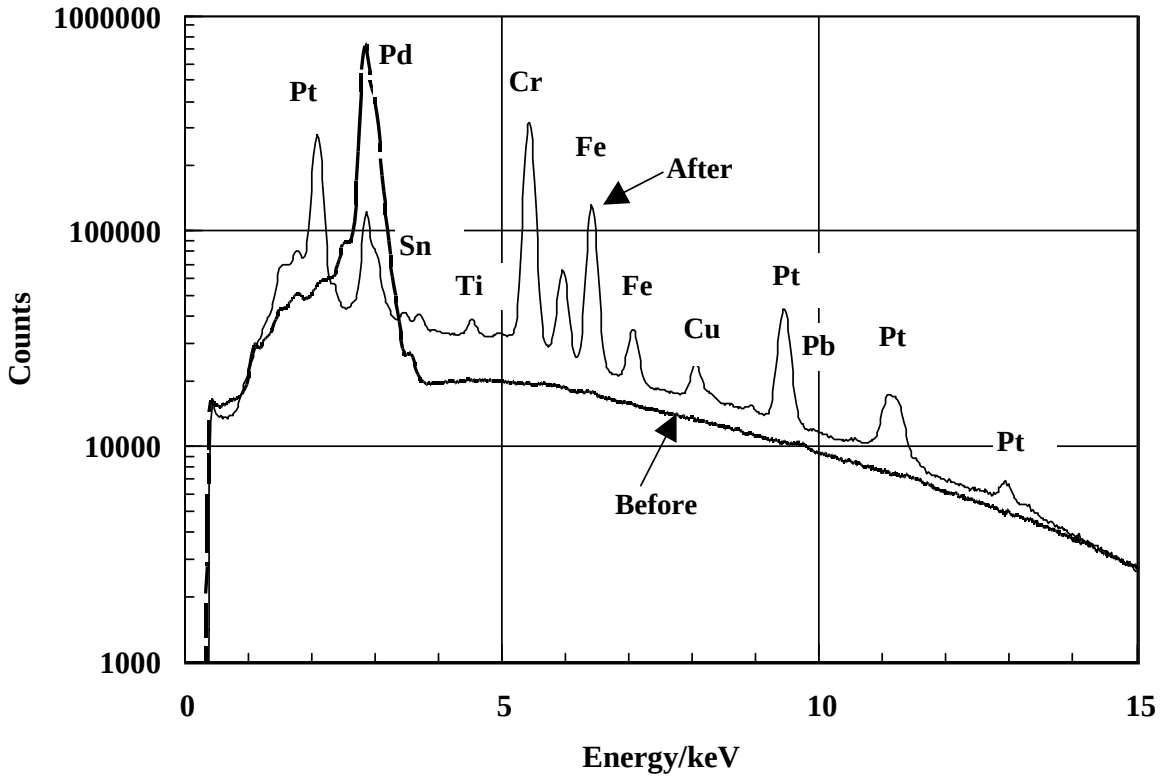
Several elements were also detected in the palladium sample by the EDX and ICP method; the measurements were taken to determine the rough level of concentration of the elements because mass peaks in the SIMS measurement can include signals from other molecular peaks.

EPMA mapping of sample Pd ; excess heat, Pd, Cr, O, Zn, C, Si, Cu, Pt, Fe, Pb, Ca and Hg



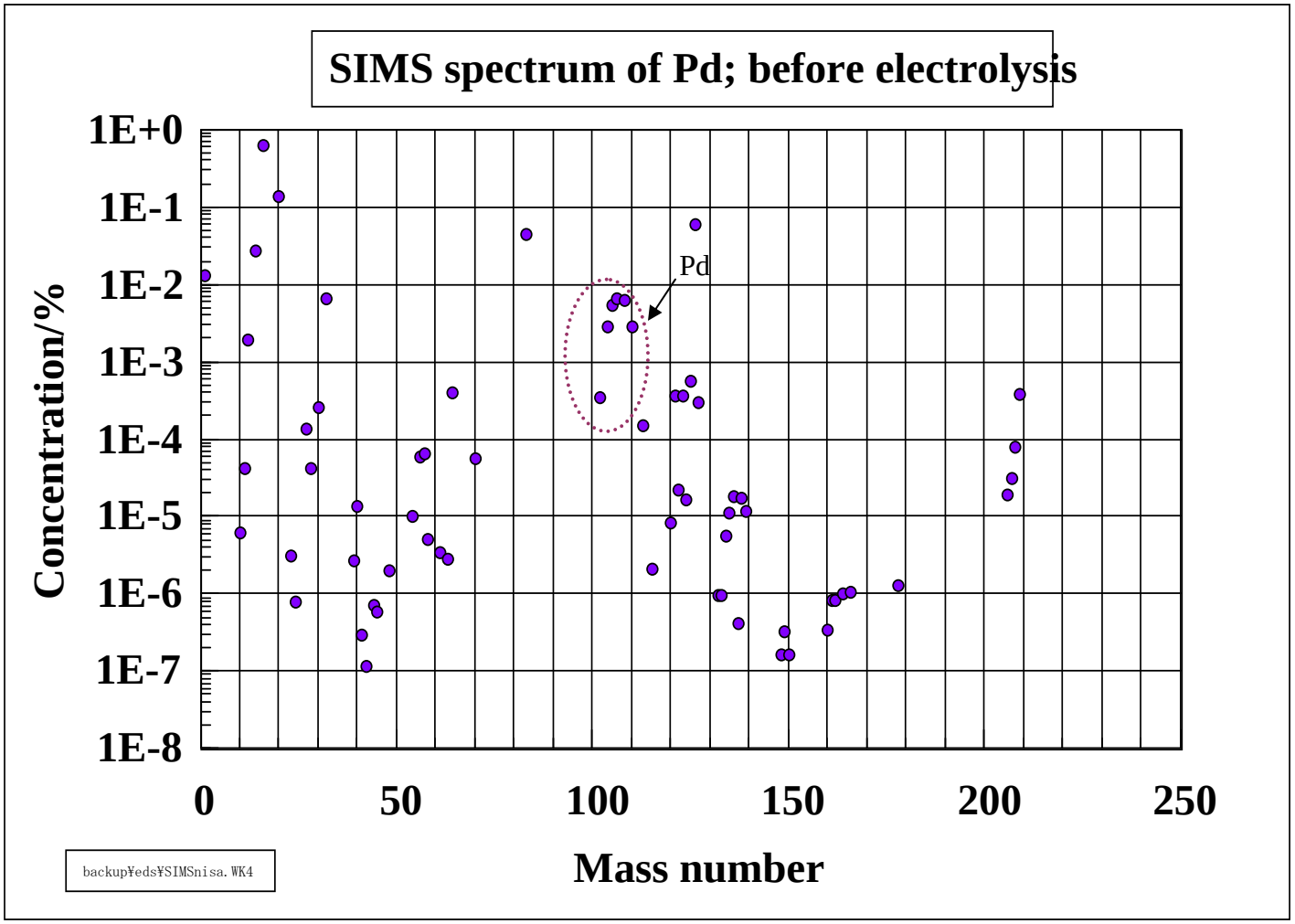
This photo shows the element mapping for the Pd surface after electrolysis by the EPMA measurement. We can see typical deposition of Cr, Zn, Cu, Fe, Pb and others. The concentration for the elements is shown by the color that changes from white to blue. Especially, in the case, strong existence of Cr, Zn and Fe are very clear.

Element spectrum by EDX of Pd sample; excess heat



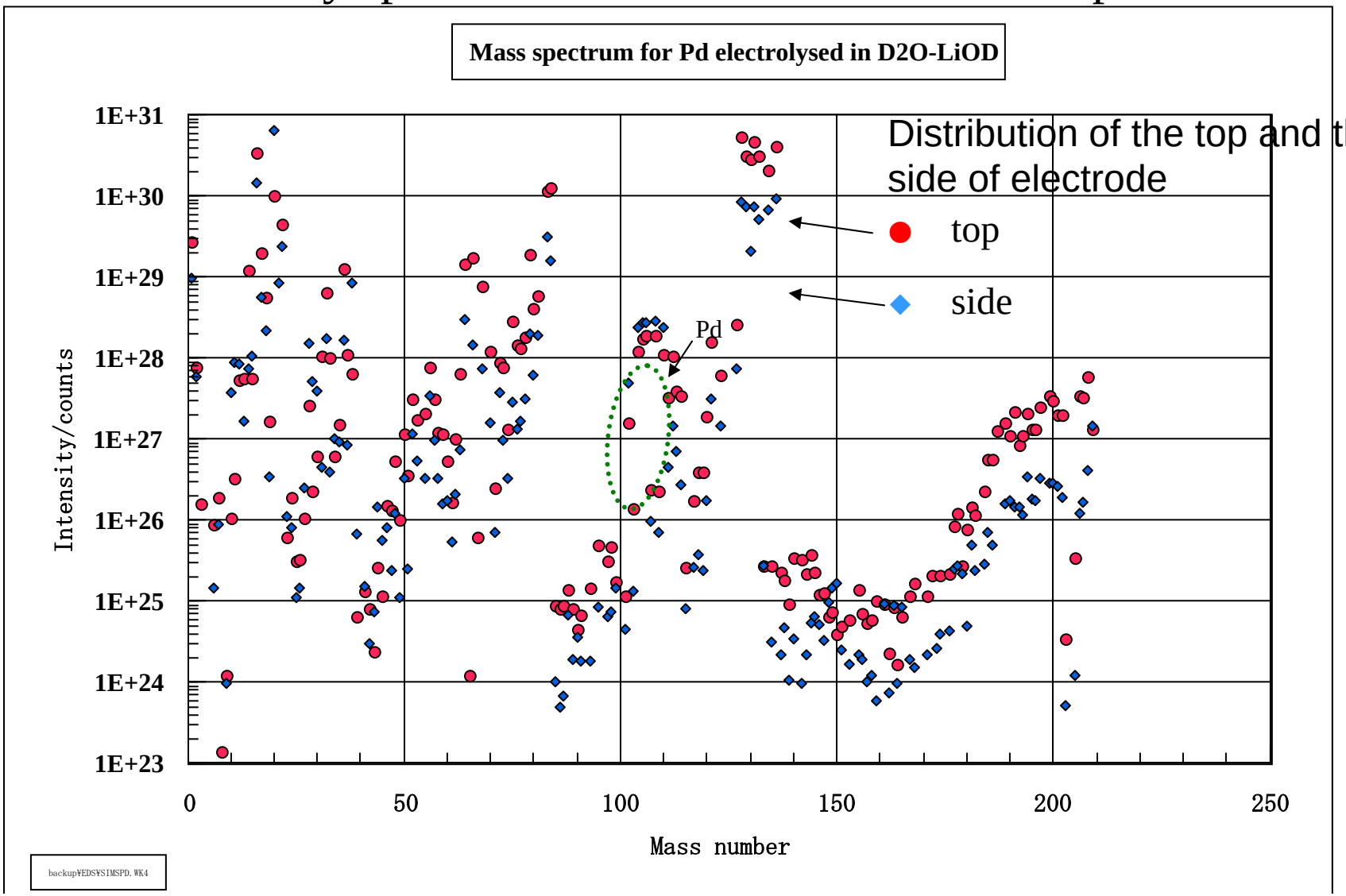
This figure shows a typical EDX spectrum for a sample that was electrolyzed in heavy water solution and evolved some excess heat (1.2 × 10^7 J) after electrolysis (1 micro meter thickness). It shows data from before and after electrolysis. Several peaks of Pt, Cr and Fe are clearly seen; these amounts were comparable with the Pd bulk peak. Smaller amounts of Sn, Ti, Cu and Pb are also clearly observed. The EDX analyses were repeated at various locations on the sample surface; the EDX counts sometimes varied by as much as a factor of ten depending on the location.

Mass spectrum of Pd



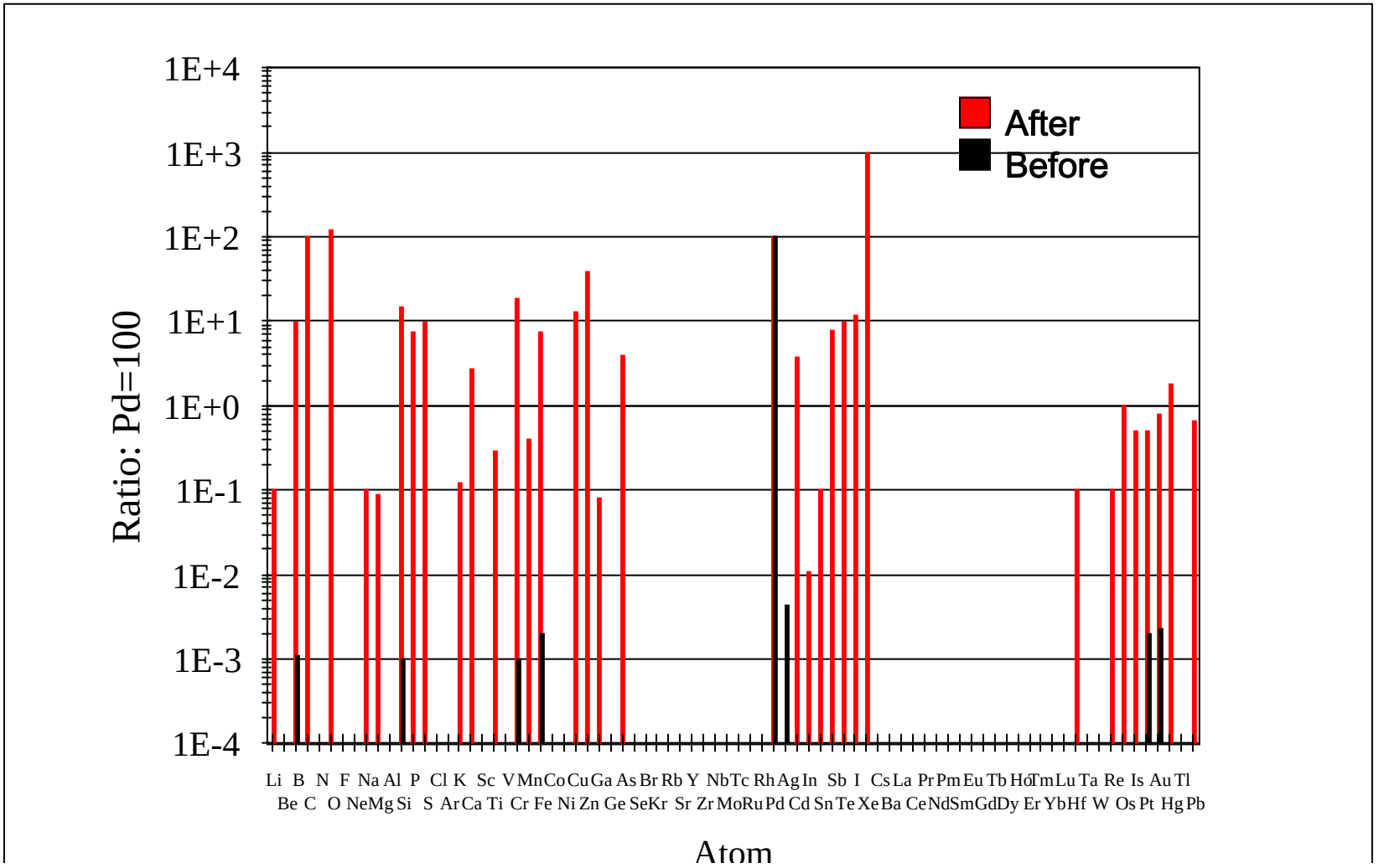
This graph shows the mass spectrum of SIMS measurement for the Pd sample surface (1 micro meter thickness) before electrolysis. Here, the mass concentration is normalized by all AIMS count number.

Intensity spectrum of SIMS count for Pd sample



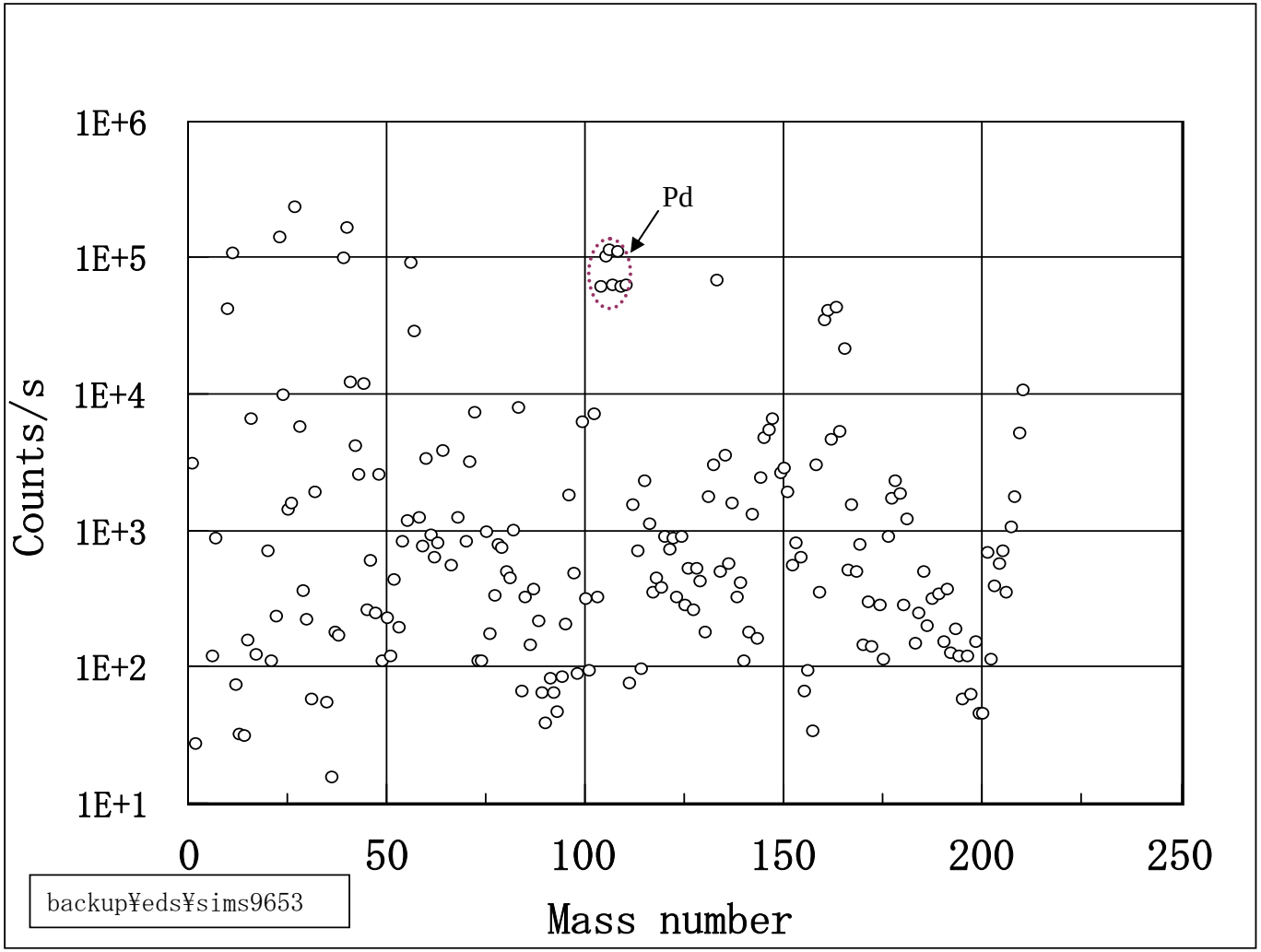
This graph shows the mass spectrum of SIMS measurement for the Pd sample surface (1 micro meter thickness) generated excess heat after electrolysis. Two of area for the sample surface were analyzed and you can see these spectra are similar. Here, the mass concentration is normalized by all AIMS count number. The spectrum has very characteristic element of Xenon. This is the inert gas and not proper element but the intensity existence is very high in the Pd electrode.

Element profile of Pd electrode before and after electrolysis



This figure shows the relative concentration for the elements before and the after electrolysis (1 micro meter thickness). I the case, the Pd electrode generated some excess heat. The concentration for the element other than the Pd were five or six order smaller than that of Pd. However, these element increased almost same as the Pd for Cr, Zn, Xe and Cu.

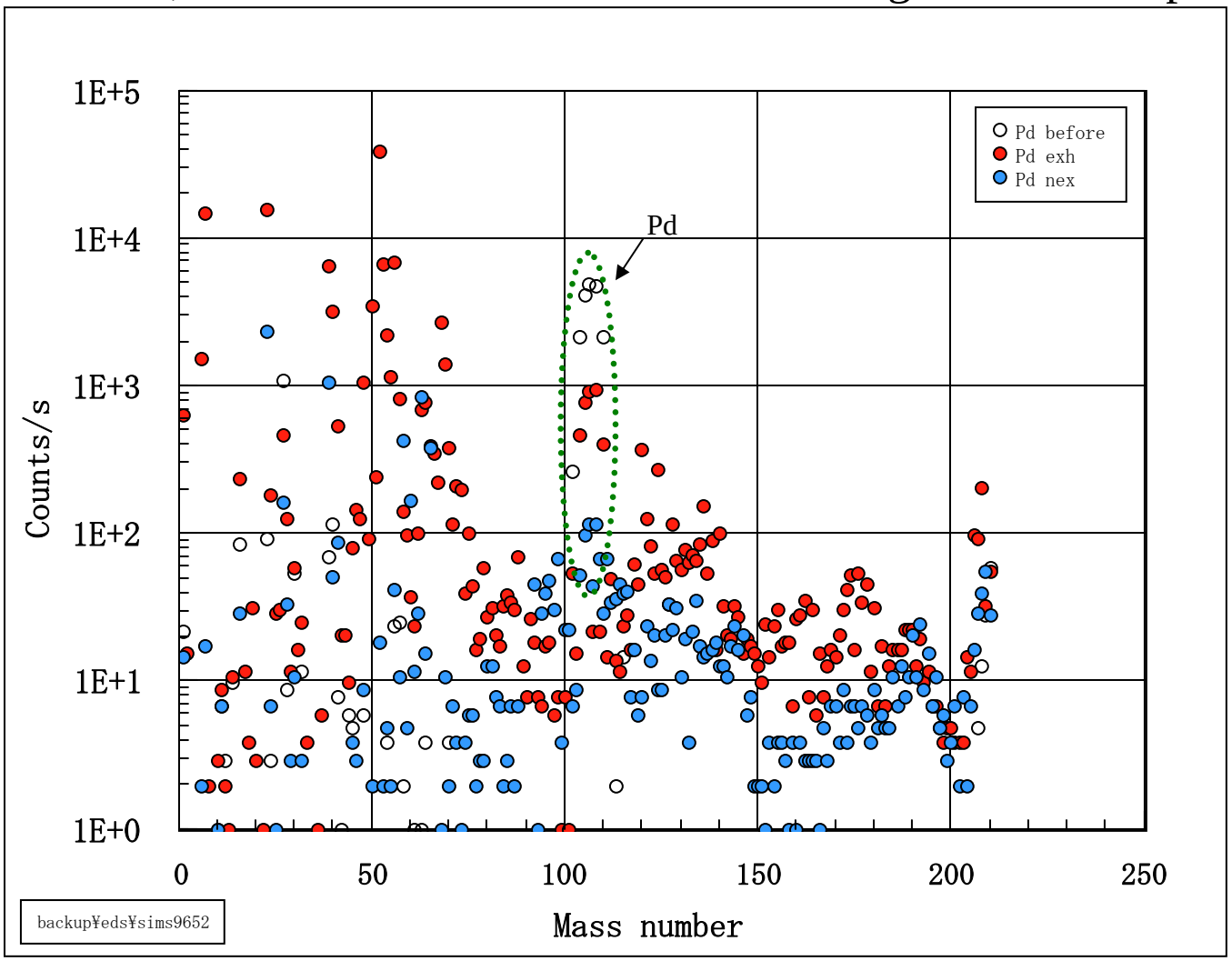
SIMS count spectrum for no excess heat evolved Pd



This graph shows the mass spectrum of SIMS measurement for the Pd sample surface (1 micro meter thickness) not generated any excess heat by electrolysis. It seems rather simple compared by the before case that the Pd generated excess heat. Most different point with the excess heat is there were no Xenon, Cr, Fe, Zn and Cu.

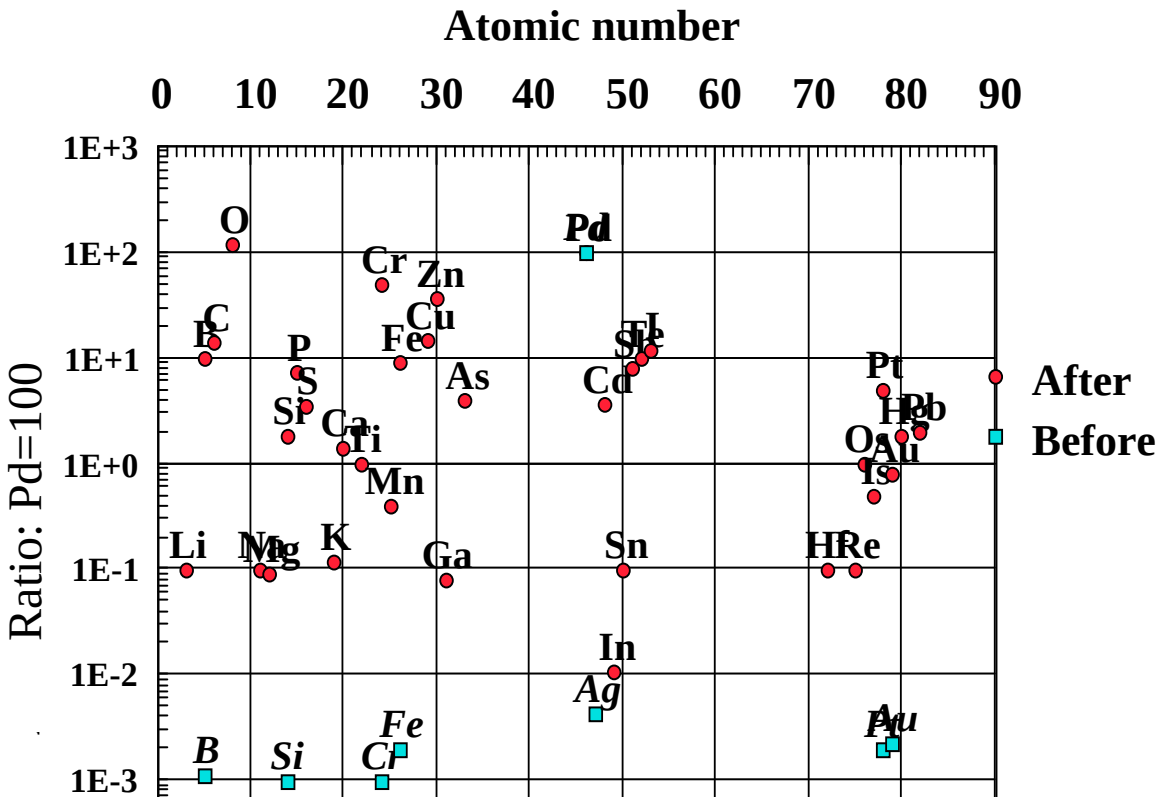
SIMS count spectrum for various mass for Pd samples

○:before, ●excess heat and ●no excess heat generated samples



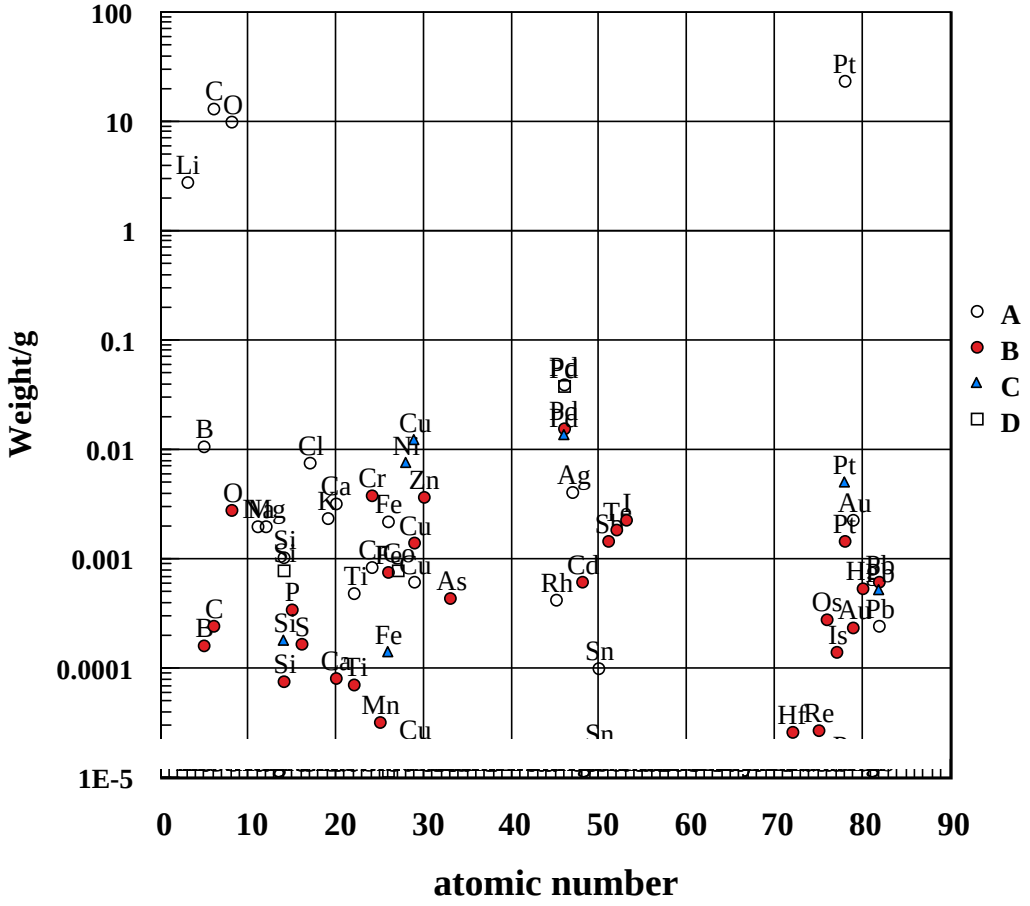
This graph shows the mass spectrum of SIMS count for the Pd sample surface (1 micro meter thickness) before and after generated excess heat and no excess heat after electrolysis. These spectra are very different from each others.

Element concentration of Pd surface: Atomic ratio with Pd



This shows the typical concentration in 10 micrometer layer of the sample. Two values are shown, for before and after electrolysis that produced excess heat. Impurities before the experiment were as follows, where the Pd concentration is set as 100, all other concentration for the element were presented compared with the Pd value. Based on this, it seems that the detected elements are distributed in atomic numbers close to those of the impurity elements that originally existed in the cell. It should be stressed that the total amount of deposited elements on the palladium is much higher than the total impurity in the electrolyte and palladium samples, except for Ca, Cl, Ti and Hg. Note especially that measurable levels of Zn impurities did not exist in the cell before electrolysis.

Weight distributions for elements after various experiments



(A,○): Sample of the before electrolysis, this includes the solute, anode material and total impurity in Pd, Pt and electrolyte in the cell.

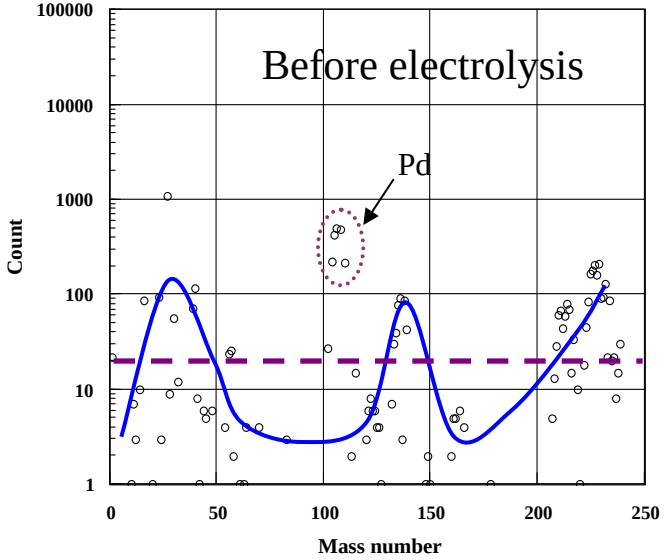
(B,●): Sample of the after electrolysis Pd surface (1 micro meter thickness) in heavy water solution that generated the excess heat,

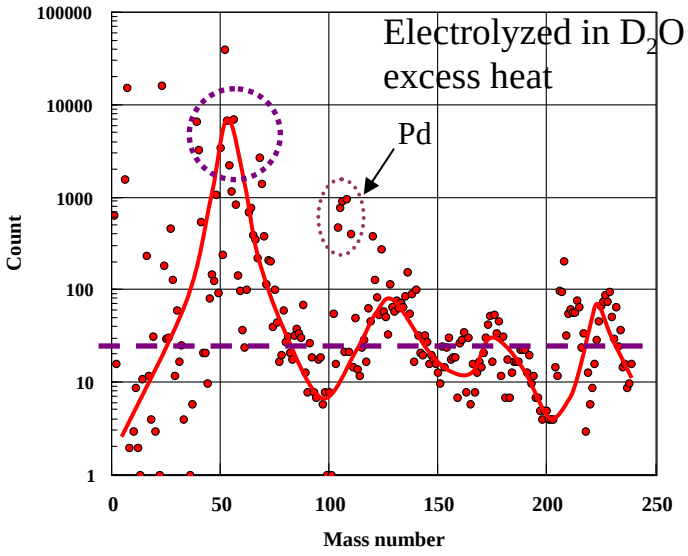
(C,▲): Sample of the after electrolysis Pd surface (1 micro meter thickness) in heavy water solution that have no excess heat,

(D,□): Sample of the after electrolysis Pd surface (1 micro meter thickness) in light water solution with no excess heat.

This shows weight abundance for various samples. These point shows the total material in the electrolyte and electrodes estimated by various methods. These include the anode material (Pt 24 g) and solute materials (Li 2.9 g, C 13.2 g and O 10 g).

Intensity of ion from the SIMS for palladium cathodes; various results



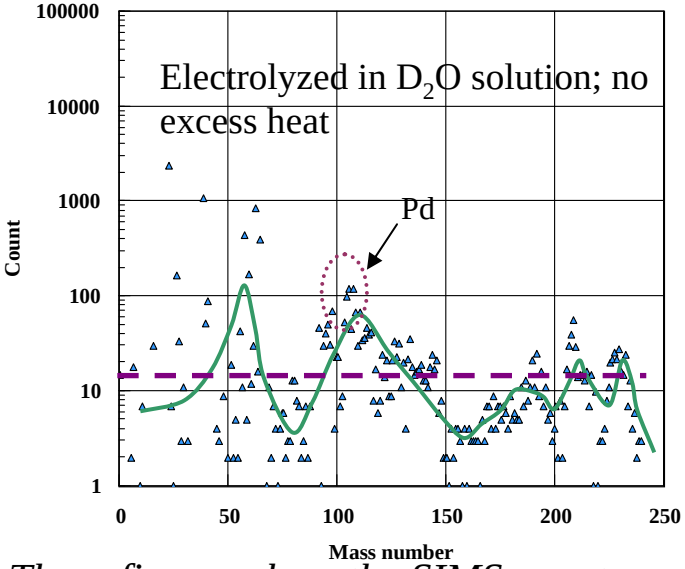


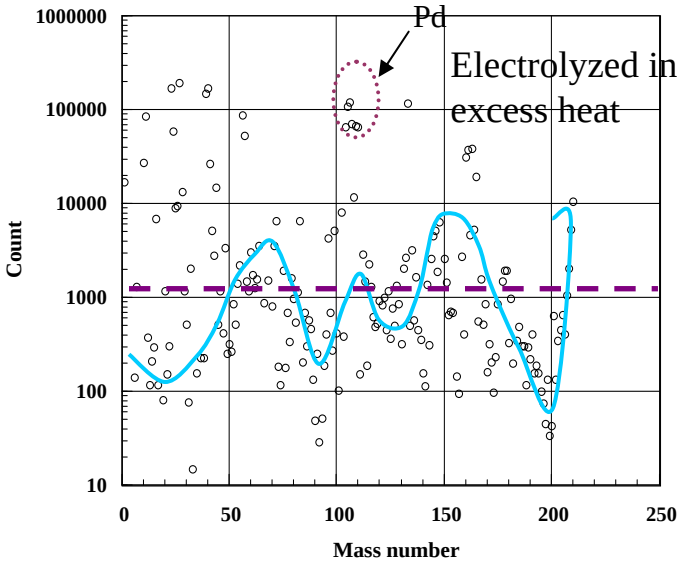
A: before electrolysis,

B: after electrolysis in heavy water when excess heat was generated,

C: after electrolysis in heavy water with no excess heat was generated,

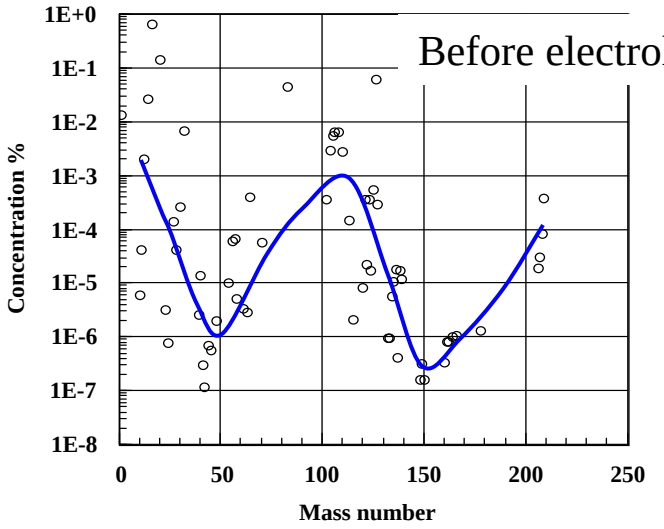
D: after electrolysis in light water with no excess heat.

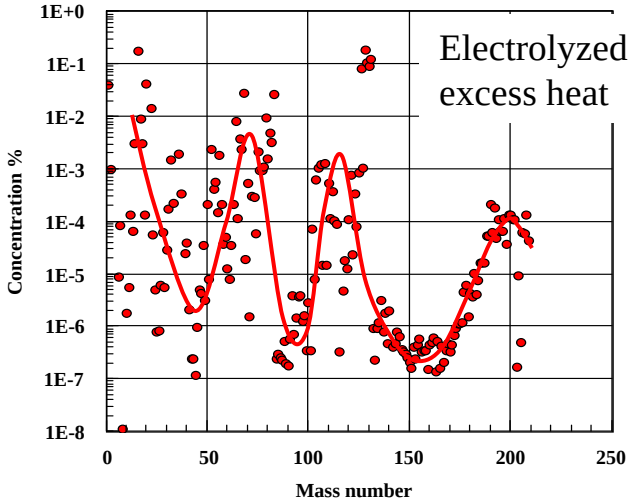


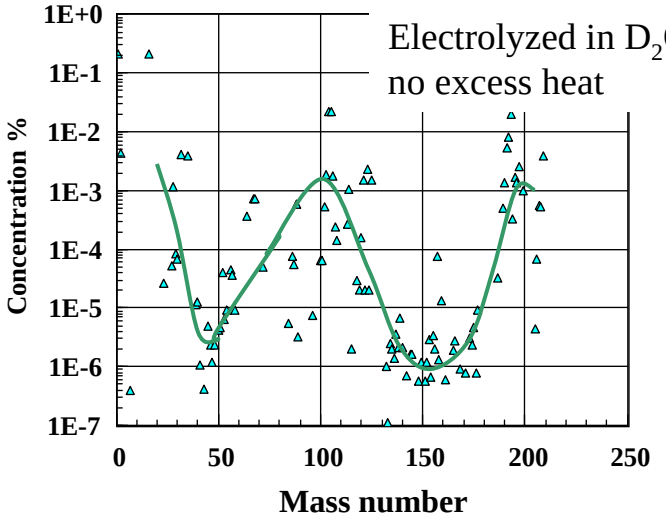


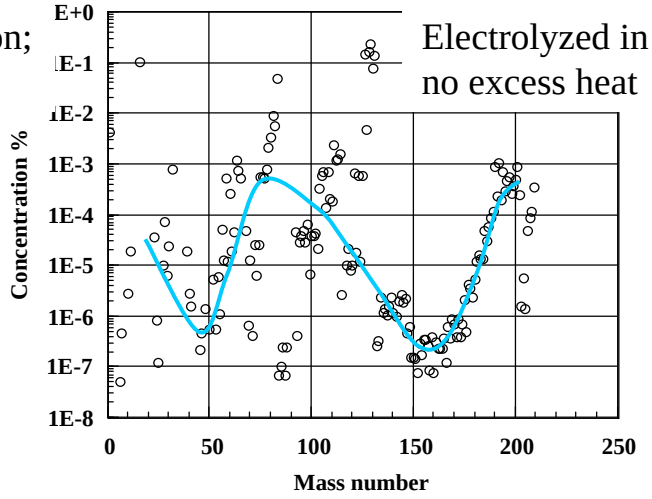
These figures show the SIMS count spectra for palladium samples that were electrolyzed under various conditions. It is very interesting to note the change in the elements according to the electrolysis condition. The change in the elements distribution before and after electrolysis is shown as a ratio of the mass spectra.

Mass distribution obtained from SIMS measurement for palladium cathodes; various condition



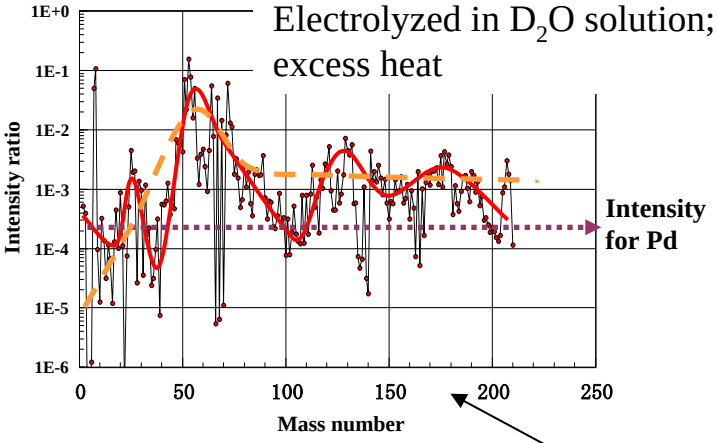


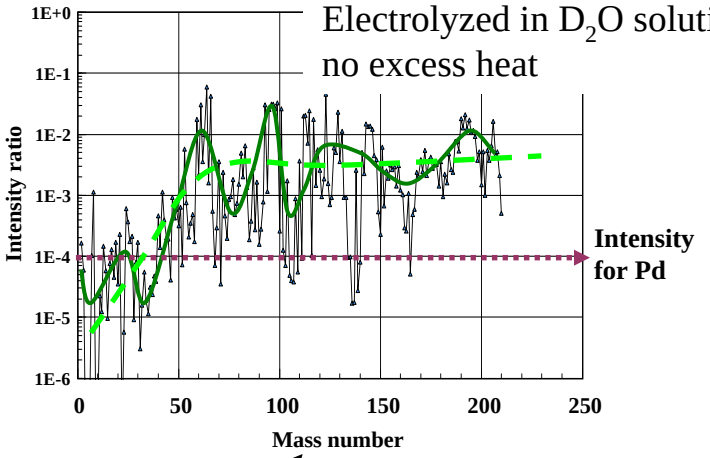


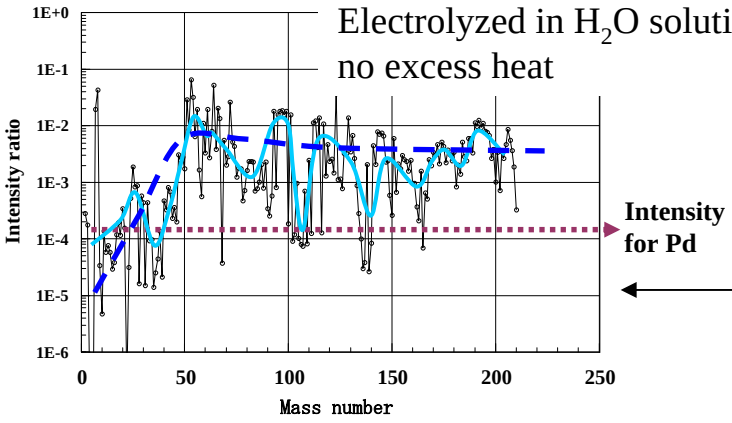


Intensity spectra corrected RSF to the before figure. (A) before electrolysis. (B) after electrolysis in heavy water solution; excess heat generated. (C) after electrolysis in heavy water solution; no excess heat, (D) after electrolysis in light water; no excess heat.

Intensity ratios compared with the distribution before electrolysis







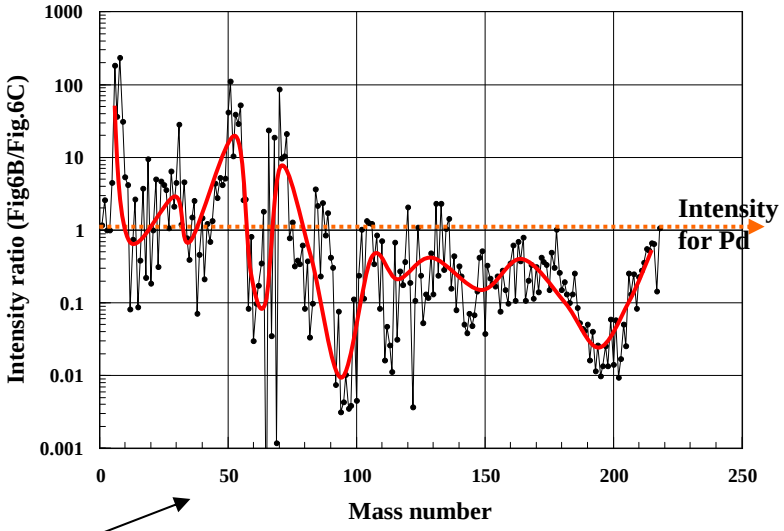
A: after electrolysis in heavy water solution when excess heat was generated.

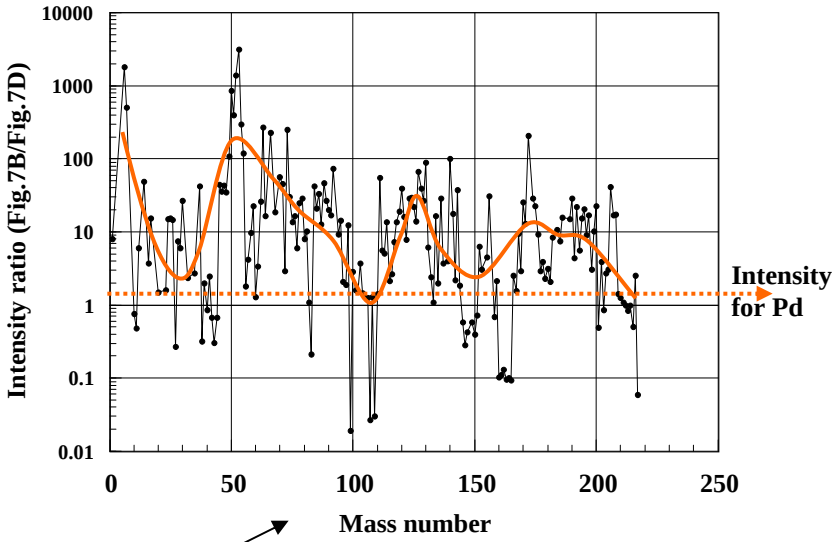
B: after electrolysis in heavy water solution with no excess heat.

C: after electrolysis in light water with no excess heat.

These figures shows the intensity ratio with Pd before electrolysis. We have to remember that the graphs show the ratios normalized by total atom number. Figures (B) and (C) shows similar distribution with the (A), but it is completely different each others. The mass spectrum of (A) has strong mass peak around the Fe and Cr.

Change of ratio of mass distribution between generated element after electrolyzed in heavy and light water solution



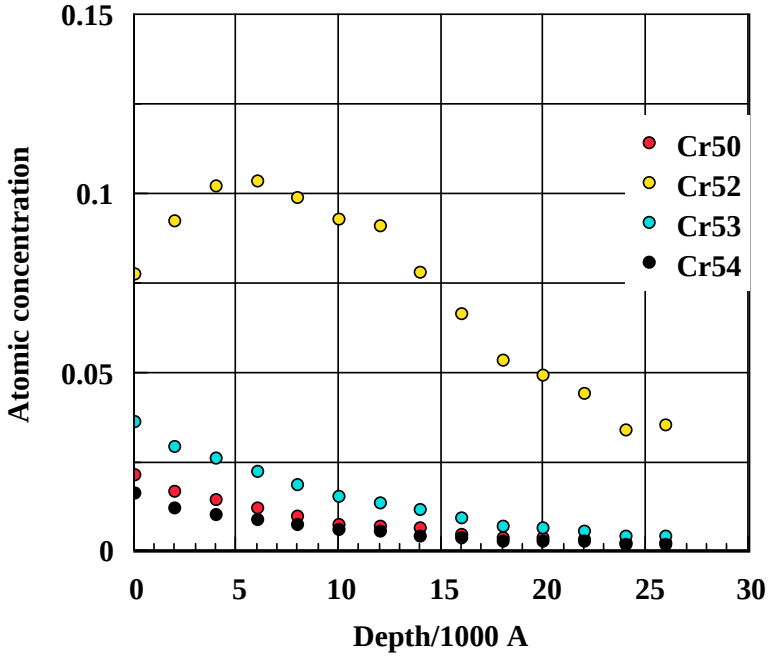


A: Intensity ratios for mass spectra for electrolysis in heavy water with excess heat compared to results with no excess heat.

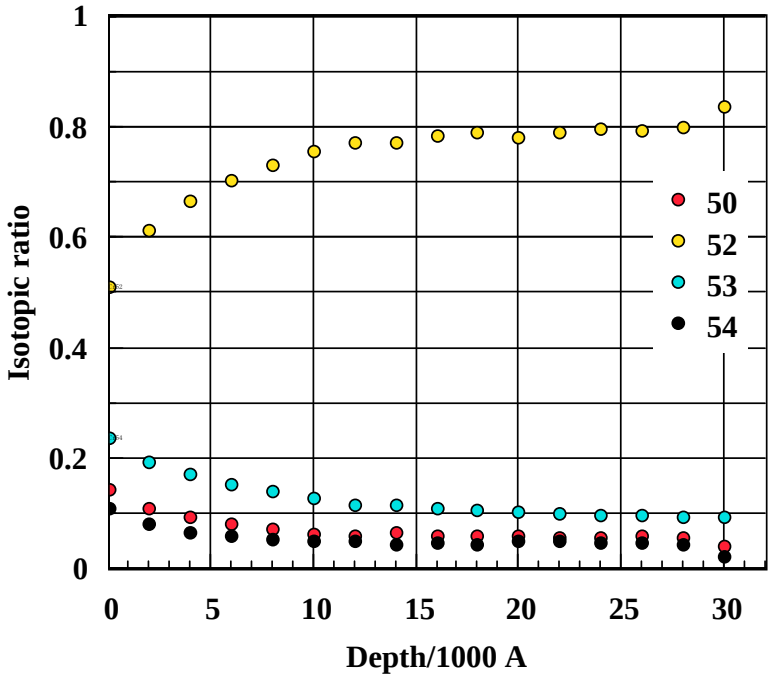
B: Intensity ratio of mass spectra between palladium in heavy water that produced excess heat and in light water that produced no excess heat.

Figure A shows the intensity ratios for mass spectra for a sample that generated excess heat compared to the ratio of a sample that produced no excess heat, both with electrolysis in heavy water. All of the data points were averaged by the total SIMS count that was obtained from surface to one micrometer depth in the palladium. Figure B shows intensity ratio for mass spectra between for a sample that generated heat in heavy water, and a sample electrolyzed in light water. There are large differences in the light elements in these two graphs. Apparently, many light elements are sometimes generated by an unknown nuclear reaction when excess heat is produced.

Cr profile in excess heat Pd rod: concentration and isotopic profiles



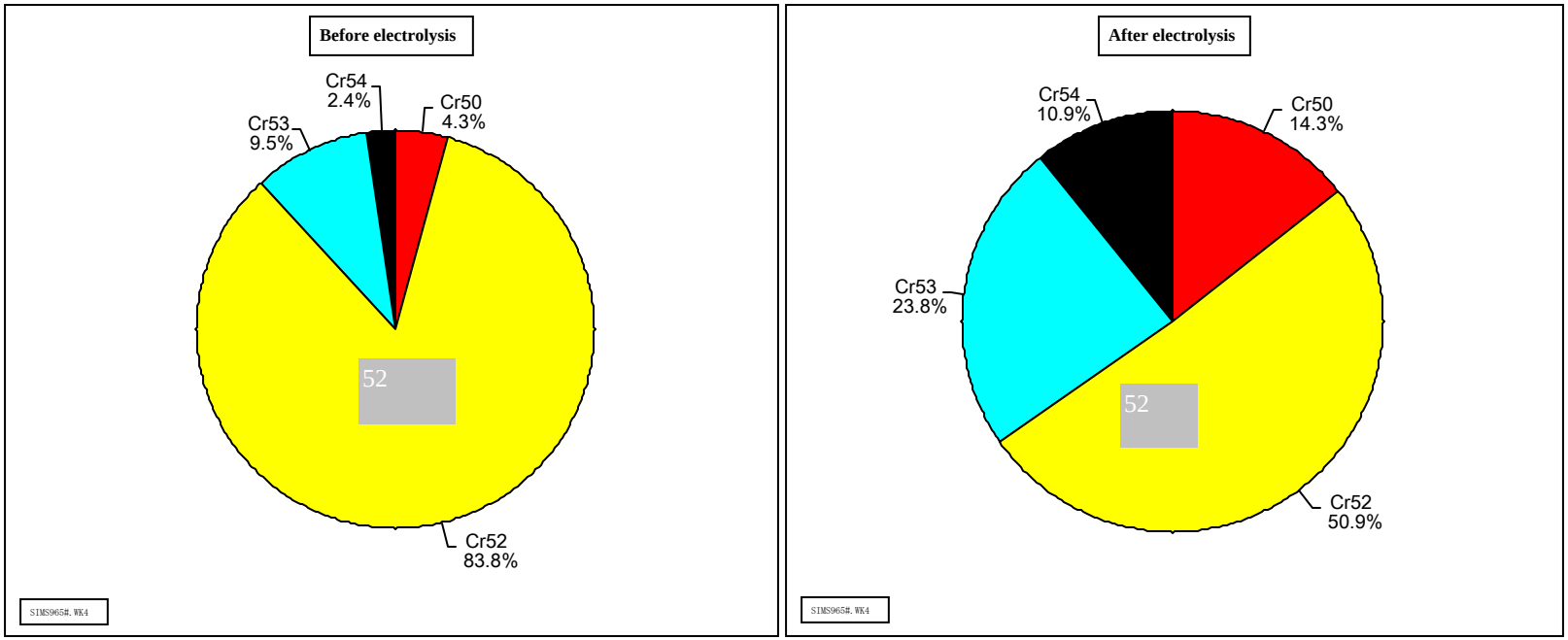
Depth profile for Cr isotopes



Depth distribution for each isotope ratios

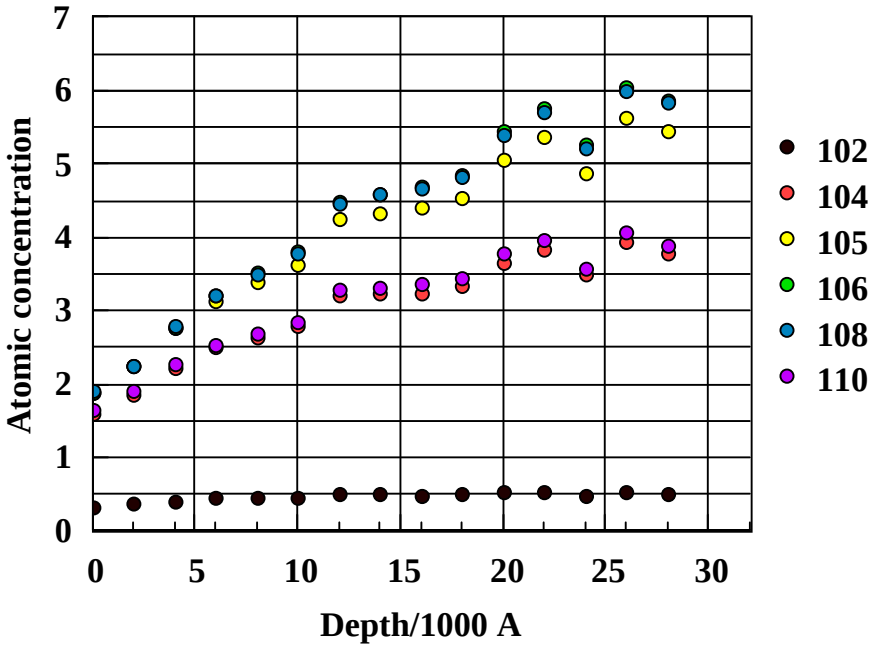
Much Cr was detected on the Pd surface generated excess heat. The quantity of Cr exceeded that of impurities in the electrolysis system. Depth profile of Cr concentration in the sample is shown in left figure. Here, you can see change of each isotopes. And the view of depth distribution for their isotope ratios are seen in right figure. You can recognize very large isotope change at the sample surface.

Cr profile in excess heat Pd rod: isotopic profiles for control and after electrolysis at the surface

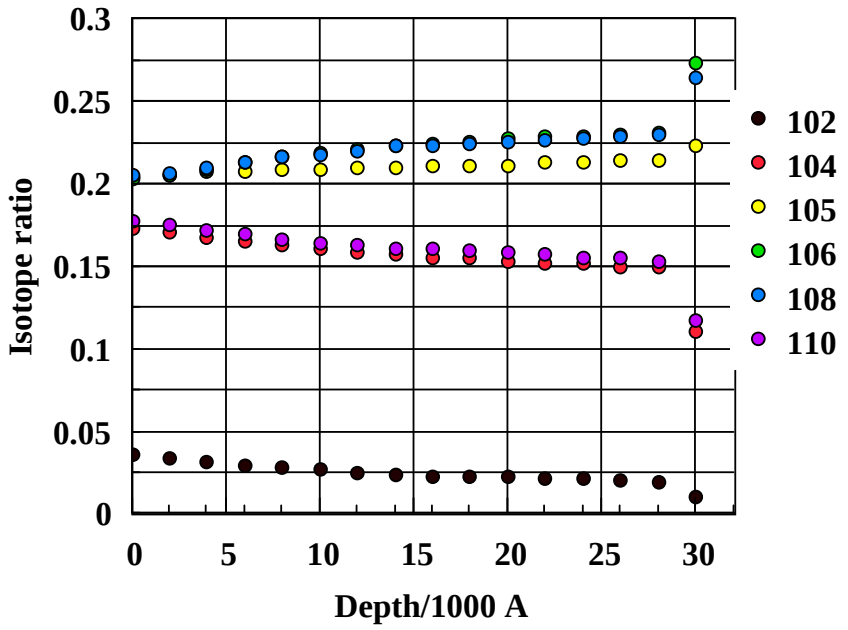


These circle graphs show the isotope abundance for Cr. Left is natural ones and right is the Pd surface that generated excess heat.

Pd profile in excess heat Pd rod: concentration and isotopic profiles

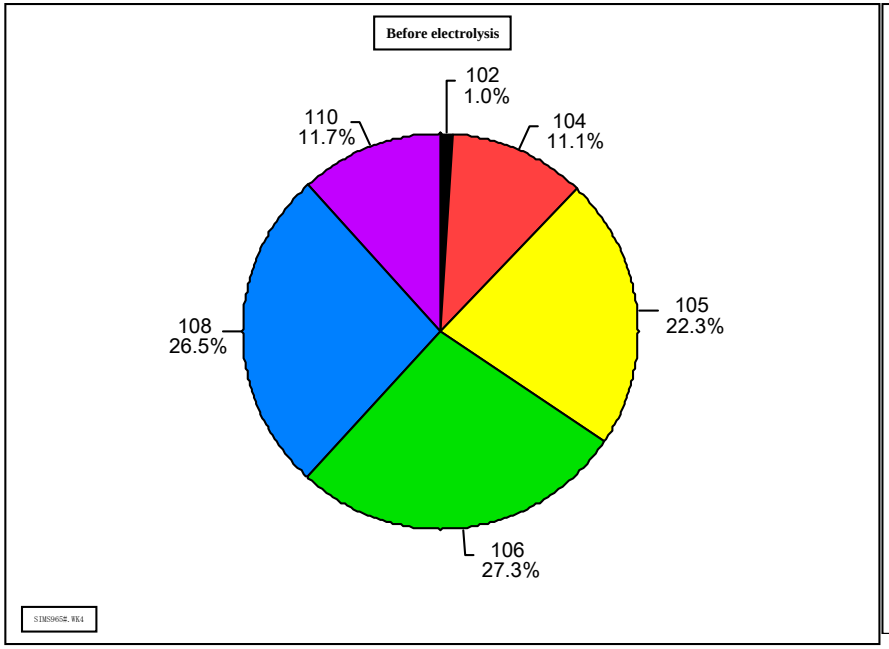


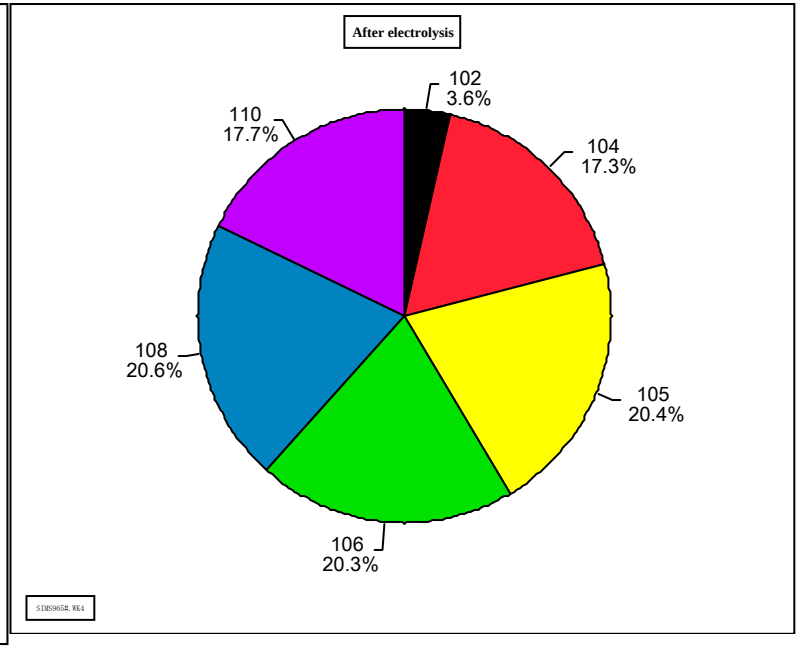
Depth profile for Pd isotopes



Pd shows isotopic change at surface and approaches normal in the Pd

Pd isotopic profiles for before and after the electrolysis at the surface; excess heat generated Pd.



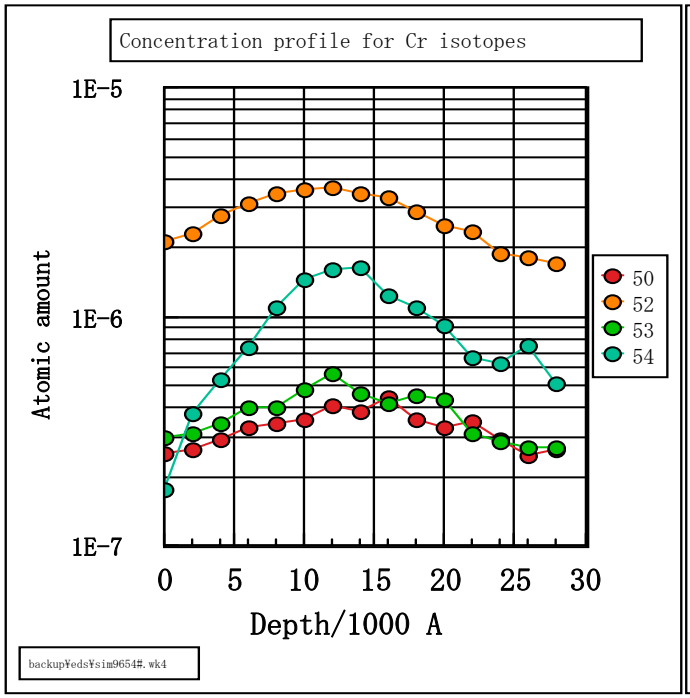


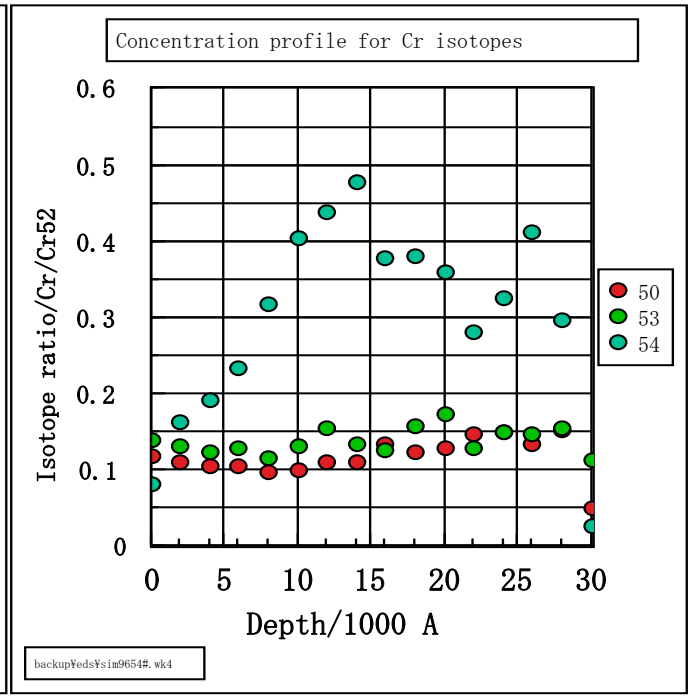
Control

After electrolysis

These circle graphs show the isotope abundance for Pd. Left is natural ones and right shows the Pd surface generated excess heat.

Cr profile in no excess heat Pd rod: concentration and isotopic profiles



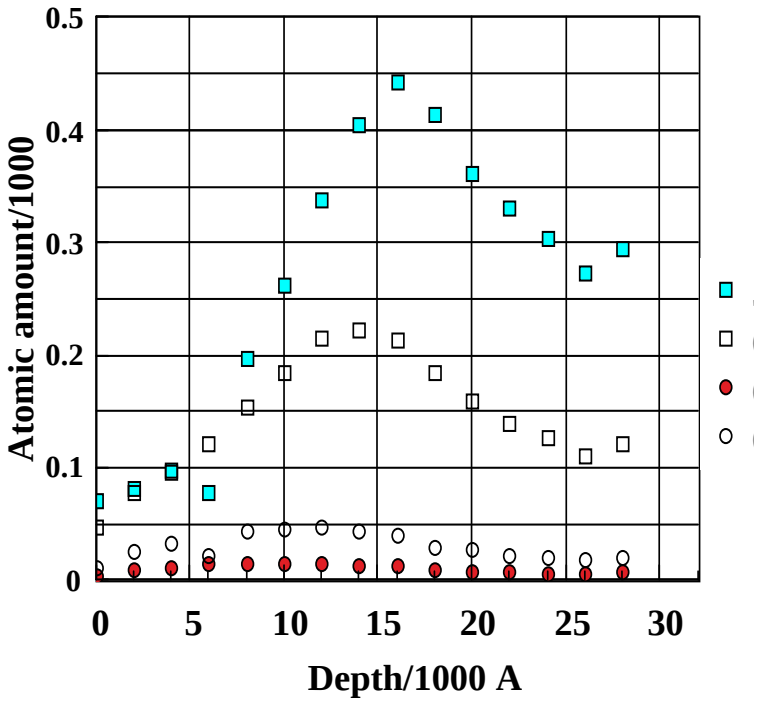


Depth profile for Cr isotopes

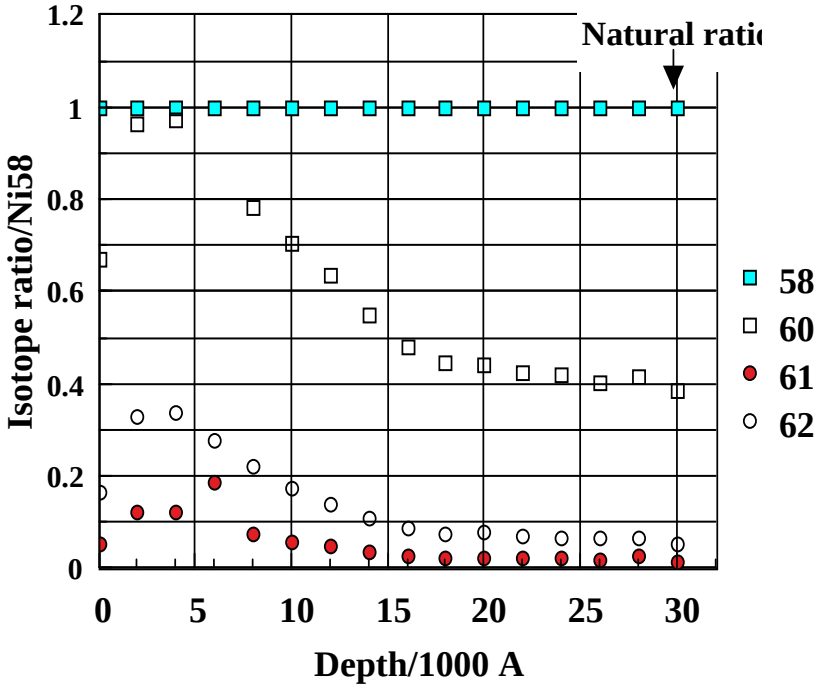
Cr shows large isotopic change in the Pd

Cr50/Cr52=0.0518, Cr53/Cr52=0.114, Cr54/Cr52=0.028

Ni profile in no excess heat Pd rod: concentration and isotopic profiles



A: Concentration profiles

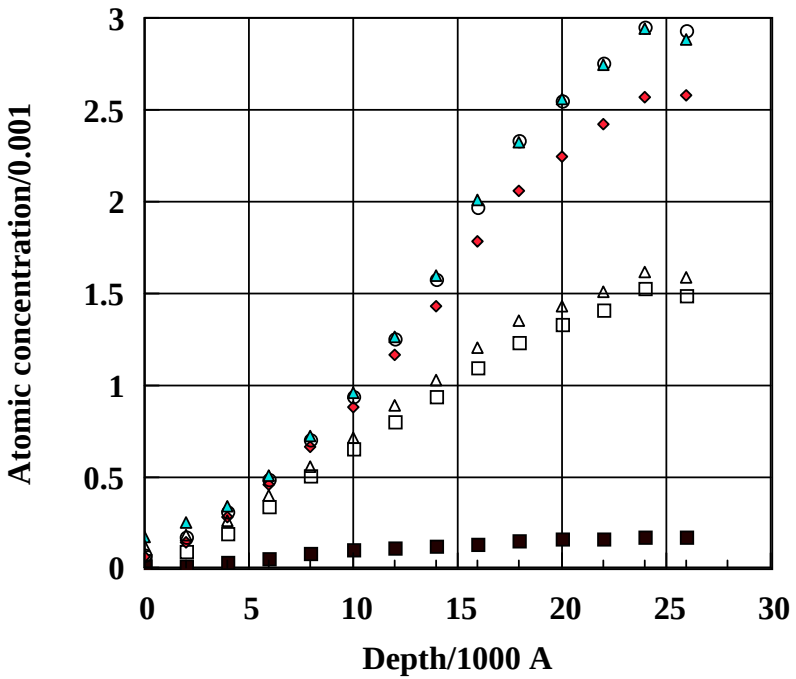


B: Isotope Profiles

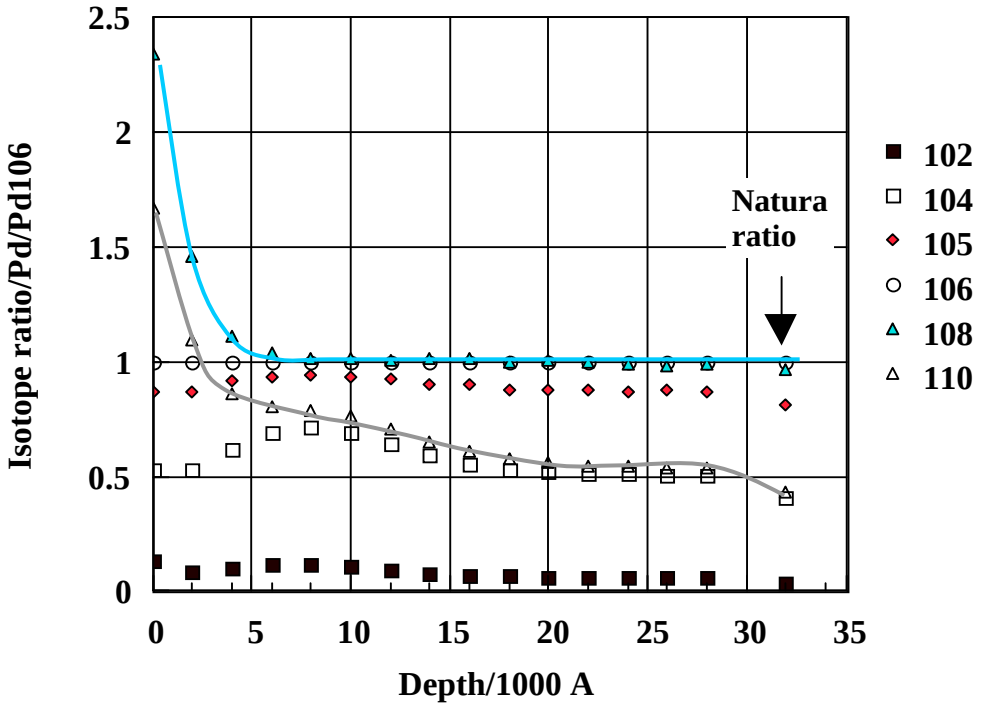
Interference species for Ni isotopes

Ni isotope	Mass number	Natural abundance	Interference species	Mass number	Natural abundance
Ni58	57.93535	68.27%	Fe58	57.93328	0.28%
			$Sn116^{2+}$	57.95087	14.53%
			$Cd116^{2+}$	57.95238	7.49%
Ni60	59.93079	26.1%	$Sn120^{2+}$	59.95110	32.59%
Ni61	59.93106	1.13%	$Sn122^{2+}$	60.95172	4.63%
Ni62	61.92835	3.59	$Sn124^{2+}$	61.95264	5.79%
			$Xe124^{2+}$	61.95329	0.10%
Ni64	63.92797	0.91%	$Zn64^{+}$	63.92915	48.60%
			$Xe128^{2+}$	63.95177	1.91%

Pd isotopic profiles for before and after the electrolysis at the surface; no excess heat (D2O)

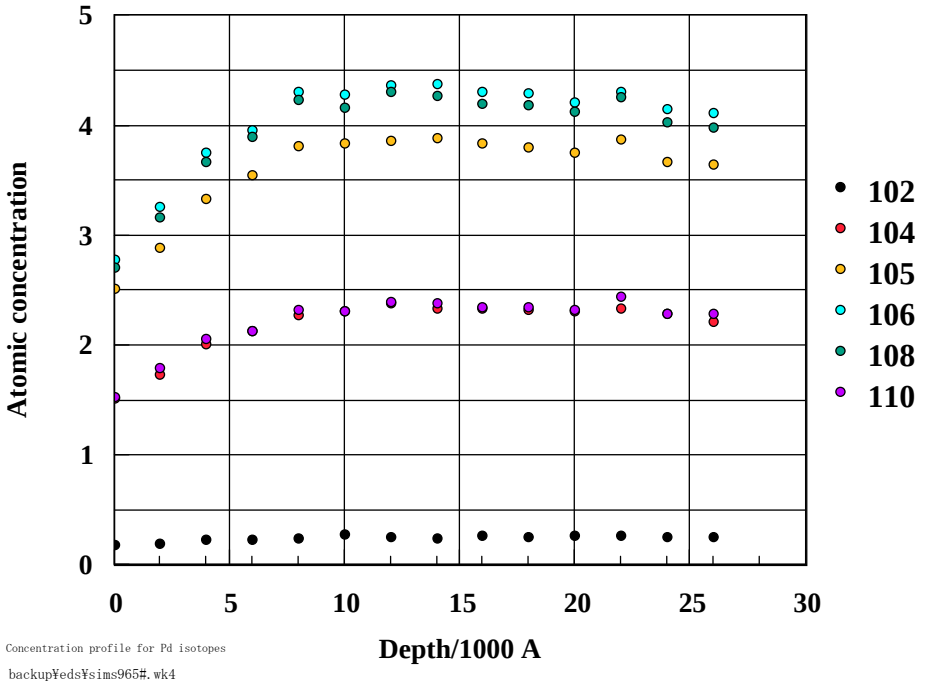


A: Concentration profiles

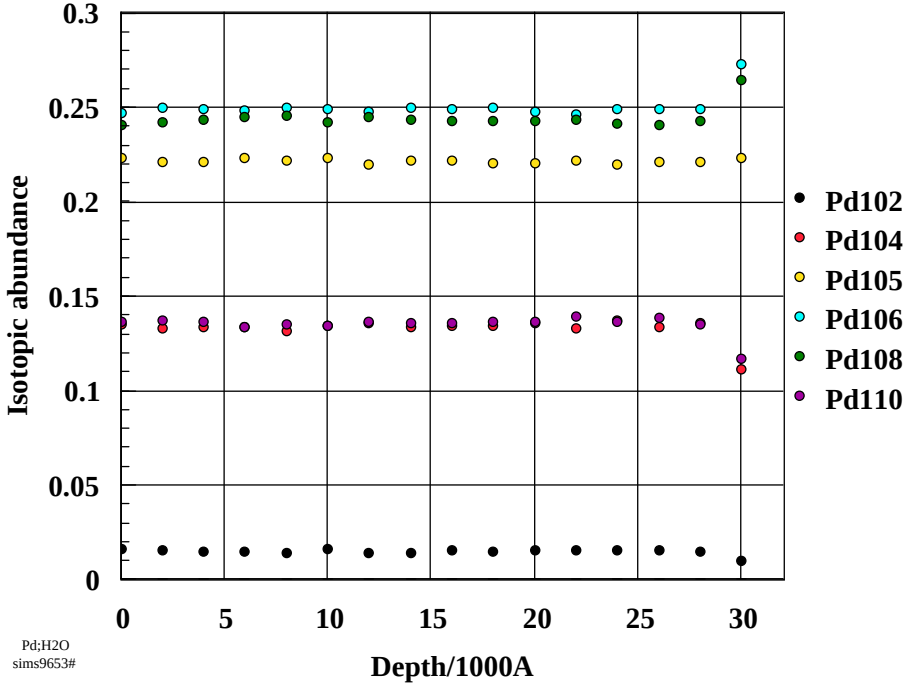


B: Isotope ratio profiles

Pd isotopic profiles for before and after the electrolysis at the surface; (H2O)



A: Concentration profiles



B: Isotope ratio profiles

Results of Pd transmutation study

- Various elements were found in the electrolyte and electrode material.
- Different isotope distributions were obtained, depending on the electrolyte and existence of heat generation.
- We assume that the cathode palladium was the starting material for these reactions, but it is also possible that impurities and other cell components such as Li, D_2O, Pd and Pt may have provided the starting material for the nuclear reactions.
- It is simple impossible to explain the shifts in the isotopic distribution. Hence, it must be concluded that some novel reactions have occurred.