ELEMENTAL ANALYSIS OF PALLADIUM ELECTRODES
AFTER PD/PD LIGHT WATER CRITICAL ELECTROLYSIS

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Elemental analyses of palladium electrodes were conducted after a new type of light water electrolysis at the optimum condition and system to induce nuclear reaction, that is referred to as Pd/Pd light water critical electrolysis. At the same time the conjecture of nuclear transmutation process becomes easier through this experiment, because detected elements are strictly restricted within the alternative of impurities or transmutation products. We consider that elements detected from cathode palladium only, namely iron, titanium, chromium, and so on, are transmutation products. Furthermore countless Ohmori-type palladium craters were observed for the first time as an indirect evidence of nuclear reactions occurred at the electrode surface.

1 Introduction

Since Ohmori et al. reported nuclear transmutation reactions with anomalous isotopic yields in his light water electrolysis system [1-4], many researchers still have claimed various kinds of low energy nuclear reactions. At the same time he observed continuous excess heat as results and countless craters as an evidence of nuclear reactions. A chain of his results in light water system deserves close attention that strongly points out the existence of condensed matter nuclear reactions.

It is, however, still difficult to replicate that reaction on normal electrolysis condition. Through our previous experiments and the other reports [5-8], we conjecture that the electrolysis system and electrical parameters are very important factors to induce nuclear
reactions. We have investigated an optimum electrolysis system and condition and describe details in this paper.

Not only the transmutation products but also surface damages are essential to prove the nuclear reaction. Figure 1 shows an example of damages on a gold electrode reported by Ohmori et al [9]. Although many researchers have reported nuclear transmutation, as far as I know, there has been no case that mentions the Ohmori-type craters. If the nuclear reactions were occurred on the electrode surface, the surface damages like Ohmori-type craters would be found after the experiment as an indirect evidence of reactions.

![Figure 1. Surface damage as an indirect evidence of nuclear reactions occurred at the electrode surface. These craters were observed on gold electrode after normal electrolysis, with current density of 0.5A/cm² for 30days in 0.5M Na₂CO₃ solution [9].](image)

2 Plasma Electrolysis

2.1. Time Variations

It is widely recognized that plasma electrolysis can produce a large amount of excess heat, and nuclear transmutation products. When a temperature of electrode exceeds the boiling temperature of electrolyte due to intense polarization, a thin vapor layer is generated at the electrode / electrolyte interface in which high electric field ionize vapor molecules to generate the plasma state. Formation of this vapor film is the first key factor to achieve the plasma electrolysis condition.

Time variations of cell voltage and current during the plasma electrolysis are shown in Figure 2. In this case the axis of ordinates means not only current but also current density because the electrode surface of cathode is ca. 1 cm². All electrolysis period is divided into 6 regions for the sake of convenience, namely conventional region, critical region, breakdown point, transitional region, partial plasma region, full plasma region.

The electrical current increases with voltage applied before the breakdown point at which the sheath of vapor film is generated. Then current drops down through the
transitional region because the electrode and the electrolyte cannot touch each other directly owing to the vapor layer.

If cell voltage is sufficiently high, some atoms or molecules in that gas phase could be ionized. Therefore current doesn’t become zero and the electrode temperature can keep high due to the good electro conductivity and the less thermal loss of the gas phase. From this region, light emission is occurred, whose color depends on the electrolyte solution. Light emission area is being expanded with increasing the cell voltage. Finally, the glow fills all surface of the electrode and the intention of light becomes strong during region-6. We defined these two regions, region-5 and 6, as plasma region.

![Figure 2. Time variations of cell voltage and current during plasma electrolysis with 1.5mm diameter tungsten cathode in 0.2mol/dm³ K₂CO₃ solution.](image)

2.2. VI Relation

A voltage-current relation of plasma electrolysis is shown in Figure 3, which is converted from Figure 2. Thus axis of ordinates also means current density. If the current density increased over approximately 2.5 A/cm², the type of electrolysis could change to the plasma where the current is almost constant. This critical value also supports the experimental result reported by Ohmori et al [10]. It is necessary to select an experimental condition, current and voltage, from this VI curve.

![Figure 3. Voltage-current relation of plasma electrolysis.](image)
2.3. Shortcomings

Although the plasma electrolysis can induce a large amount of excess heat with hydrogen and anomalous nuclear transmutation [11, 12], this type of electrolysis has some and fatal shortcomings.

At the boundary between region-5 and 6, that is, at the time when whole area of the electrode surface is covered with the glow, the temperature of surface rises up to 1,400 degree centigrade. Furthermore local temperature exceeds the melting point of the electrode at the sparks like arc discharges happen due to the generation of current flow paths between the electrode and the electrolyte. Therefore the electrode cannot stand over 30 min without being melted or broken. And the electrolyte solution is also pyrolyzed by the high temperature plasma.

The most serious problem is that the rare nuclear transmutation products on the electrode surface would be lost at the same time. Therefore another type of electrolysis after which the electrode cannot be damaged is required to prove the nuclear reactions.

3 Critical Electrolysis

3.1. Optimum Region

Here, we suppose the positive correlation between the excess heat and the current density in normal region, while the excess heat increases with the input voltage in plasma region. This assumption is illustrated in Figure 4 with the VI relation described at Figure 3. In fact some researchers have reported these correlations [13-15].

Since the plasma region is inappropriate, Ohmori et al. specified that region-2 is most favorable region to obtain the excess heat and the transmutation in electrolysis system, and referred to this type of electrolysis as Critical Electrolysis [7, 8]. Although this condition depends on each experimental system, a target current density is approximately 2.5A/cm². Through this type of electrolysis, a long time experiment is possible because the electrode cannot be damaged. Thus transmutation products would be well preserved and continuous excess heat could be obtained.
3.2. Results

We had conducted critical electrolyses with nickel cathodes in 1M Na$_2$CO$_3$ solutions [5]. Figure 5 shows EDX spectrums of nickel electrodes after the experiment through which the current density was kept ca. 2.6A/cm$^2$ for 15 days. The mean voltage applied was 14V and solution temperature raised up to 80 degree centigrade.

Detected strong platinum peaks are probably originated from a counter electrode though the isotopic ratio for this platinum has not been confirmed yet. In general, the platinum anode is less dissolved and electrodeposited in alkaline solution, especially in K$_2$CO$_3$ or Na$_2$CO$_3$ solution. However the experimental result suggests that this unusual reaction is accelerated in a certain condition, and this platinum complicates transmutation processes like photofission [16]. Then that electrodeposited platinum makes an evaluation of process difficult. Therefore the experimental system that the anode material has to be same as cathode is more appropriate to discuss the transmutation simply.

4 Pd/Pd Critical Electrolysis

4.1. Experimental Set-up

Hence Pd/Pd Critical Electrolysis is optimum system and condition to evaluate the transmutation process precisely. In this system, the effect of the counter electrode is avoidable. Therefore detected elements are completely restricted within the alternative of impurities or transmutations. Furthermore, if they are transmutation products, the process becomes simpler.

A schematic view of an experimental set-up is illustrated in Figure 6. The system is required to be very simple, because this has to be cleaned very carefully before an experiment to avoid contamination.
A Teflon (PFA) cell (Flon Industry Co., Ltd.) whose capacitance, diameter and height are 300cc, 80mm and 70mm, respectively is capped with a silicon rubber. Although vapor cannot leak from this system, hydrogen can easily leak from a hole with another silicon rubber to add the Milli-Q water to restore the volume of the electrolyte which decreases due to the decomposition.

![Figure 6. An experimental set-up for the Pd/Pd critical electrolysis experiments.](image)

In this type of experiment anode and cathode are just same palladium wires (Tanaka Kikinzoku Kogyo) whose purity, diameter, length and surface area are 99.95%, 1.0mm, 15.7mm and ca. 0.5cm², respectively. They were polished up by emery papers (#1500 and #2000), and washed with acetone. After being covered with Teflon (PTFE) tubes, they were located at the both side of the cell symmetrically. Before an experiment, the cell and electrodes were cleaned carefully with nitric acid or sometimes mixed acid (1:1 H₂SO₄+HNO₃), and rinsed with Milli-Q water.

The electrolyte solution of 1M and 200cc was prepared by K₂CO₃ (Kanto Chemical Co., Inc.) whose purity is over 99.95% and Milli-Q water whose specific resistance is over 18.0 MΩ·cm. Isotopic abundant of hydrogen atoms of this Milli-Q water is natural since ordinary water was used. Although it contains D₂O with very low purity, this influence is ignorable.

The cell was placed in a constant temperature chamber (MIR-151, Sanyo Electric Co., Ltd.). The solution temperature was roughly measured by two thermo couples, which located at the center and edge of the cell. Air temperature was also monitored and kept 22.0~24.0 degree centigrade.

The current density was over 2.5A/cm² controlled by a constant current/voltage power supply (GP0250-3R, Takasago Ltd.) in each experiment. The experiments were continued for 7 or 10days, with collecting various data by a data logger (8421-50, Hioki E. E. Co.). During the electrolysis, Milli-Q water was added every 12 hours to restore the volume of electrolyte decreasing due to the decomposition.

4.2. Elemental Analysis

After the experiments both electrodes were observed by SEM (JSM-6500F, JEOL Ltd.) and analyzed by EDX. Figure 7 shows those spectrums of whole area of each electrode. In this case, current density had been 3.2A/cm², for 7days. A thick line means the
cathode palladium spectrum, and a thin line is not one before the experiment but the anode spectrum. The count of the anode spectrum is multiplied 1.18 to be overlapped with the cathode spectrum. Through Pd/Pd electrolysis a comparison between the cathode and the anode is more suitable than that between before and after.

Figure 7. EDX spectrum s of palladium electrodes, cathode and anode, after the Pd/Pd critical electrolysis with current density of 3.2A/cm^2 for 7 days in 1M K_2CO_3 solution. Counts of anode spectrum are multiplied 1.18 to be overlapped with the cathode spectrum.

Bold letter elements iron, titanium, chromium, manganese, nickel were detected from the cathode palladium only, on the other hands, italic letter elements, cupper, zinc and magnesium were detected from both electrodes in this case. Therefore we consider the bold letter elements would be transmutation products. Especially, the iron peaks were detected from all cathode samples after Pd/Pd critical electrolysis.

These are two possibilities of origin for the italic letter elements, transmutations or impurities. If these were transmutation products, at least two processes should exist. The first nuclear reaction should be occurred at the cathode only to produce the bold letter elements, while the second reaction should be occurred at both electrodes to produce the italic letter elements.

Since we have never analyzed the samples by the other methods, we cannot determine the origin of these elements at this moment. If we confirmed the isotopic yields, the origin and transmutation processes of detected elements could be discussed, whether electrodeposited impurities or transmutations. Here all elements, of course, cannot be detected from samples before the experiment.

4.3. Micro Structure

Figure 8 and 9 are SEM photographs of the anode palladium surfaces and the cathode surfaces after the Pd/Pd critical electrolysis, respectively. The anode palladium has some cracks due to the oxidation.

On the cathode surfaces the countless Ohmori-type palladium craters were observed as an indirect evidence of nuclear reactions. These craters, which are located along the surface cracks or grain boundaries, were observed on the cathode only until now. This result means the nuclear reactions are easier occurred around the surface and the cracks.
The maximum width and height of craters are over 10 micrometer. These crater sizes have a positive correlation with the current density. This result supports our prediction that the amount of excess heat and transmutation have a positive correlation with increase of current density. Therefore the critical region is optimum to induce the nuclear reactions as described above.

Figure 8. SEM photographs of the anode palladium surfaces after Pd/Pd critical electrolysis with current density of 3.6A/cm$^2$, for 10days in 1M K$_2$CO$_3$ solution. Magnifying power and scale standard line are shown at the bottom of each photograph.

Figure 9. SEM photographs of Ohmori-type palladium craters on the cathode surfaces after Pd/Pd critical electrolysis with current density of 3.6A/cm$^2$, for 10days in 1M K$_2$CO$_3$ solution. Magnifying power and scale
standard line are shown at the bottom of each photograph. The craters whose maximum size is over 10 micrometer are located along the surface cracks.

4.4. Discussion

To our knowledge, this is the first report of the Ohmori-type craters observed on the palladium electrodes. The characteristics of these palladium craters are similar to the gold craters first reported by Ohmori et al [3]. Although 10 years past after the report, the formation mechanism is still unknown.

Kamada et al. reported anomalous heat evolution and surface melting of deuteron implanted aluminum foil upon electron bombardment [17]. They estimated that the amount of anomalous heat exceeds the total amount of any kind of chemical reactions and, therefore, concluded a novel nuclear reaction had been occurred in the deuterated aluminum. If palladium electrode were melted due to this type of nuclear reaction, the palladium gas should be erupted. Then they should be cooled down in the solution and be recrystallized like the crater.

Numata et al. found vortex patterns on well annealed thick palladium electrodes after a long term heavy water electrolysis [18], and simulated a magnetic interaction of hypothetical particles to elucidate an evolution mechanism of vortexes [19]. They showed the vortex can be formed at the electrode/electrolyte interface by FEM method. If charged palladium particles dissolved from the anode were caught in a vortex which is generated at the electrode/electrolyte interface, they might be accumulated to the cylindrical shape like the vortex to form the craters.

Mizuno et al. described a large explosion occurred during a normal light water electrolysis [20, 21]. They roughly estimated an energy balance and considered it as not a simple explosion but a large excess heat. If micro explosions were occurred on the electrode surface, the craters could be generated.

It is still difficult to explain the mechanism by these theories. Although they may be impurities electrodeposited uniformly, we claim that the crater is an indirect evidence of nuclear reaction occurred at the electrode surface.

5 Conclusion

Pd/Pd light water critical electrolyses were performed as the optimum condition and system to induce nuclear reactions. In this study the shape of both electrodes was just same and the current density was over 2.5A/cm², for 7 or 10days, in 1M K₂CO₃ solution whose temperature was up to 70–90 degrees centigrade.

After the experiment, the obvious transmutation products namely iron, titanium, chromium, manganese, nickel were detected by EDX. In particular, the iron peaks are very strong and detected from all cathode samples. The anomalous isotopic yield of detected iron should be expected since the anomaly has already claimed by many researchers [3, 4, 22]. The elements detected from the both electrodes, namely, cupper, zinc and magnesium, however, cannot determine their origin at this moment. If they were transmutation products, at least two mechanisms exist.
The analysis of isotopic yields for all elements detected by SIMS must be indispensable to prove nuclear reactions. Quantitative analysis of the detected elements, distribution especially around craters, and isotopic yields are under considerations. Moreover, nuclear radiation detection could give important information for process evaluation. Precise heat measurement is also required. The estimation and reduction of impurities is most important.

Although the isotopic yields have not confirmed yet in this study, Ohmori et al. have already reported anomalous isotopic distribution of palladium with excess heat in their Pd/Pt critical electrolysis system [7, 8]. Furthermore the indirect evidence of nuclear reactions, namely Ohmori-type palladium craters were observed for the first time. Since the transmutation products and the surface damages have been found, Pd/Pd critical electrolysis is optimum to induce condensed matter nuclear reactions.

References