北海道大学工学部研究報告
第176号（平成8年）

Bulletin of the Faculty of Engineering
Hokkaido University, No. 176 (1996)

Radioactive Analysis of Palladium Used for an Explosive Cold Fusion Experiment

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(Received October 27, 1995)

Abstract

A radioactive analysis of palladium metals that were used for an explosive cold fusion experiment was performed. The samples were irradiated by thermal neutrons at the graphite column of the Kyoto University Reactor (KUR). The production of ^{115}In and ^{104}Ru was clearly observed. The production of the elements is discussed based on The Nattoh Model, in which the nuclear transmutation of the palladium element was proposed.

Keywords

cold fusion, radioactive anlalysis, KUR, nuclear transmutation, production of In and Ru, The Nattoh Model

1. Introduction

The nuclear transmutation was discussed earlier in The Nattoh Model (1, 2) and was proposed by one of the authors (T. M) related to cold fusion phenomena (3). The model proposed three processes for the nuclear transmutation: (1) sequential many-body fusion reactions of hydrogen isotopes, (2) capturing of electrons or hydrogen isotopes by a host element, and (3) multi-body fission reaction induced by a multiple neutron. All three processes simultaneously take place during cold fusion experiments. The distribution of the produced elements depends on the process. The first process sequentially produces light elements, and the second process generates heavy elements near the host element.

An explosive cold fusion experiment was carried out with a palladium metal rod in a previous experiment (4), in which light elements such as Na, Mg, Ti and Fe and heavy elements of Ru and In were found in the metal by an analysis with a scanning electron microscope (SEM) and an energy dispersive X-ray spectroscopy (EDX). A portion of those elements was asigned to be not impurities but to be nuclearly transmuted during the electrolysis. The nuclear transmutation was suggested to be closely related to the cold fusion phenomena (2).

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This paper describes a radioactive analysis (RA) of the palladium metal that was used in the previous experiment. The RA method was applied to confirm the production of the heavy elements through the second process. The palladium samples were irradiated at the Kyoto University Reactor (KUR). The RA method was a sensitive analysis so that provided an alternative method to the SEM-EDX method.

2. Experiment

An experiment of explosive cold fusion was carried out in the previous paper (4), in which hydrogen isotopes were moved through a palladium metal rod to violently burn hydrogen fuel. The experiment was made by electrolyzing heavy water mixed with 3 % NaCl. After charging a sufficient amount of hydrogen into the palladium rod for 19 days, a special procedure of moving hydrogen through the metal was made to induce a small scale explosion several hours later. The detailed result was described elsewhere (4).

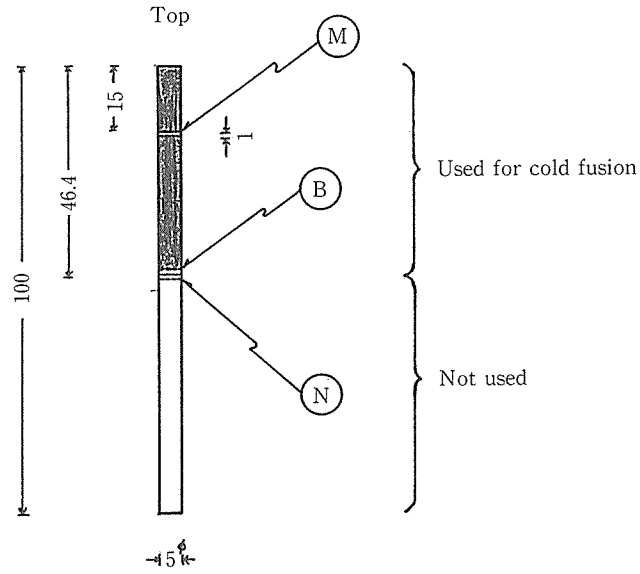

Fig. 1; Sample discs cut from Pd rod

In this paper, the palladium rod used in the previous experiment was analyzed by the RA method. Figure 1 shows the preparation of three disc-like samples (about 1 mm thick) of palladium, that were cut away from the palladium rod (purity 99.95 %, 5 mmϕ x 100 mm). A half upper portion of the palladium rod was used for the electrolysis experiment of cold fusion and the other lower portion was left for the measurement of the background. The samples of M and B were cut at the center and the bottom of the used rod, respectively, and N was the top of the reference rod. In order to avoid impurity problems as much as possible, careful treatments were carried out. The adjacent samples of B and N were selected because of reducing the impurity problems. All the samples were simultaneously irradiated

under thermal neutrons at the graphite column of KUR (5), in which the thermal neutron flux was about 2.6×10^{10} n/cm^2.sec. The neutron spectrum at the irradiation field was sufficiently thermalized so that only the reaction of (n, gamma) contributed to activate the samples. The neutron-induced gamma rays were detected by a Ge(Li) detector.

3. Radioactive Analysis

The activity of the neutron-induced gamma rays is related to the number density of a parent isotope as follows,

$$C(E_k : T_m) = fN_0\phi \qquad (1)$$

where
- C : counting rate of activity
- E_k : k-th energy of gamma ray
- T_m : measuring time
- f : modification factor.
- N_0 : number density of parent isotope
- ϕ : thermal neutron flux

The modification factor, f, can be given as follows,

$$f = \varepsilon(E_k)\,\sigma_k\delta(1-e^{-\lambda T_e})\frac{1}{\lambda}e^{-\lambda(T_m-T_e)}(1-e^{-\lambda\Delta T}) \qquad (2)$$

where
- $\varepsilon(E_k)$: detector efficiency
- γ_k : gamma-ray intensity
- σ : cross section of (n, gamma)
- λ : decay constant
- T_e : irradiation period
- ΔT : measuring period

The detector efficiency, $\varepsilon(E_k)$, was measured by using a standard source of ^{233}U, located at the same position as the samples. Numerical values derived from Ref. 6 were used for γ, σ and λ.

4. Results

Figure 2 shows a comparison of the energy spectra of the neutron-induced gamma rays, that were measured immediately after the irradiation. Figures 3 and 4 show the similar spectra that were measured two hours later and after sufficient time elapsed, respectively. Several peaks due to the palladium isotopes (^{109}Pd, ^{111}Pd and ^{111m}Pd) were seen for all the samples of M, B and N with many peaks due to impurities.

Three peaks due to ^{116m}In (1097, 1294 and 2112 keV) was obviously observed for M and B, but not for N, as shown in Figs. 3 and 4. After sufficiently time elapsed, on the other

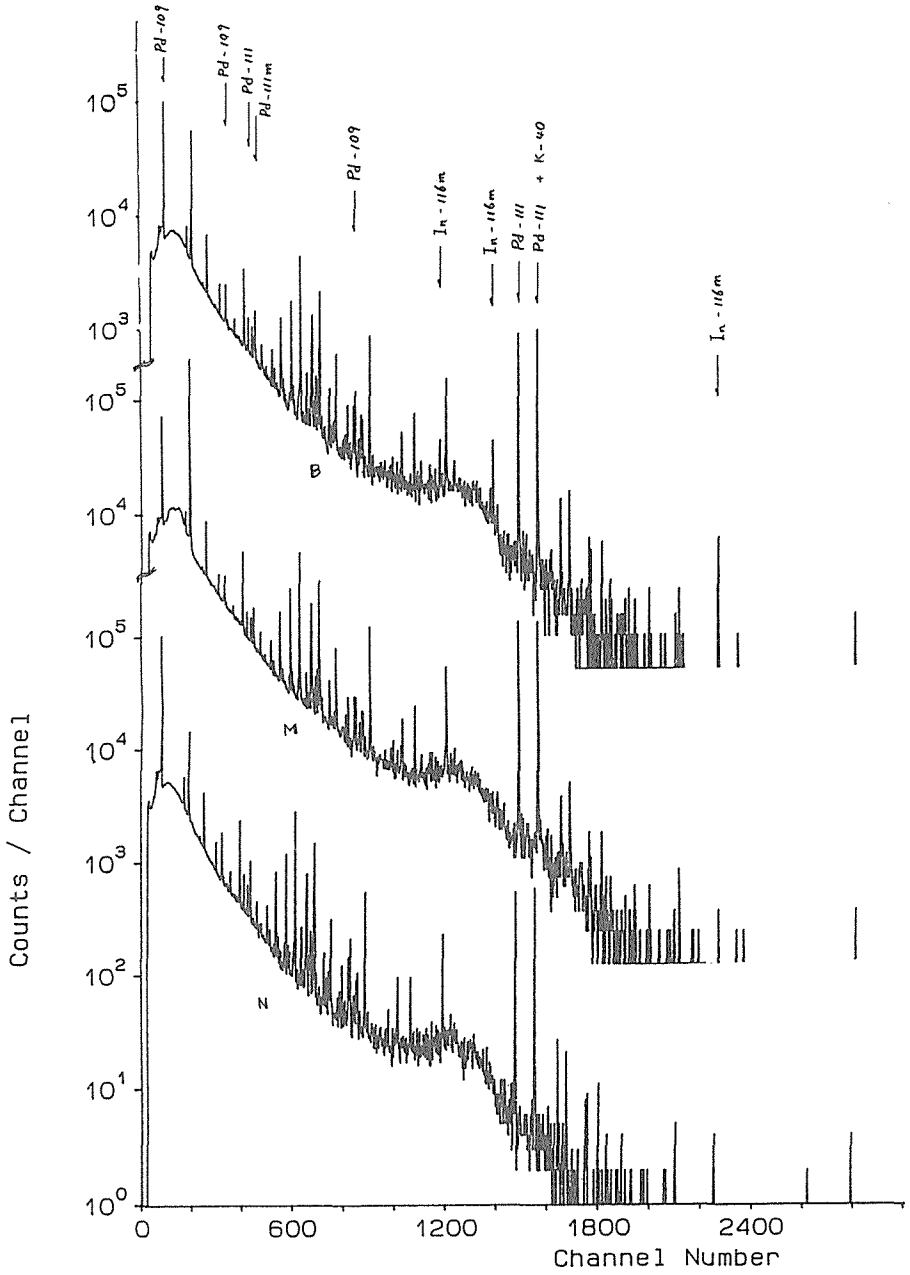

Fig. 2: Gamma ray energy spectrum immediately after irradiation
M: middle in used Pd
B: bottom of used Pd
N: top of non-used Pd

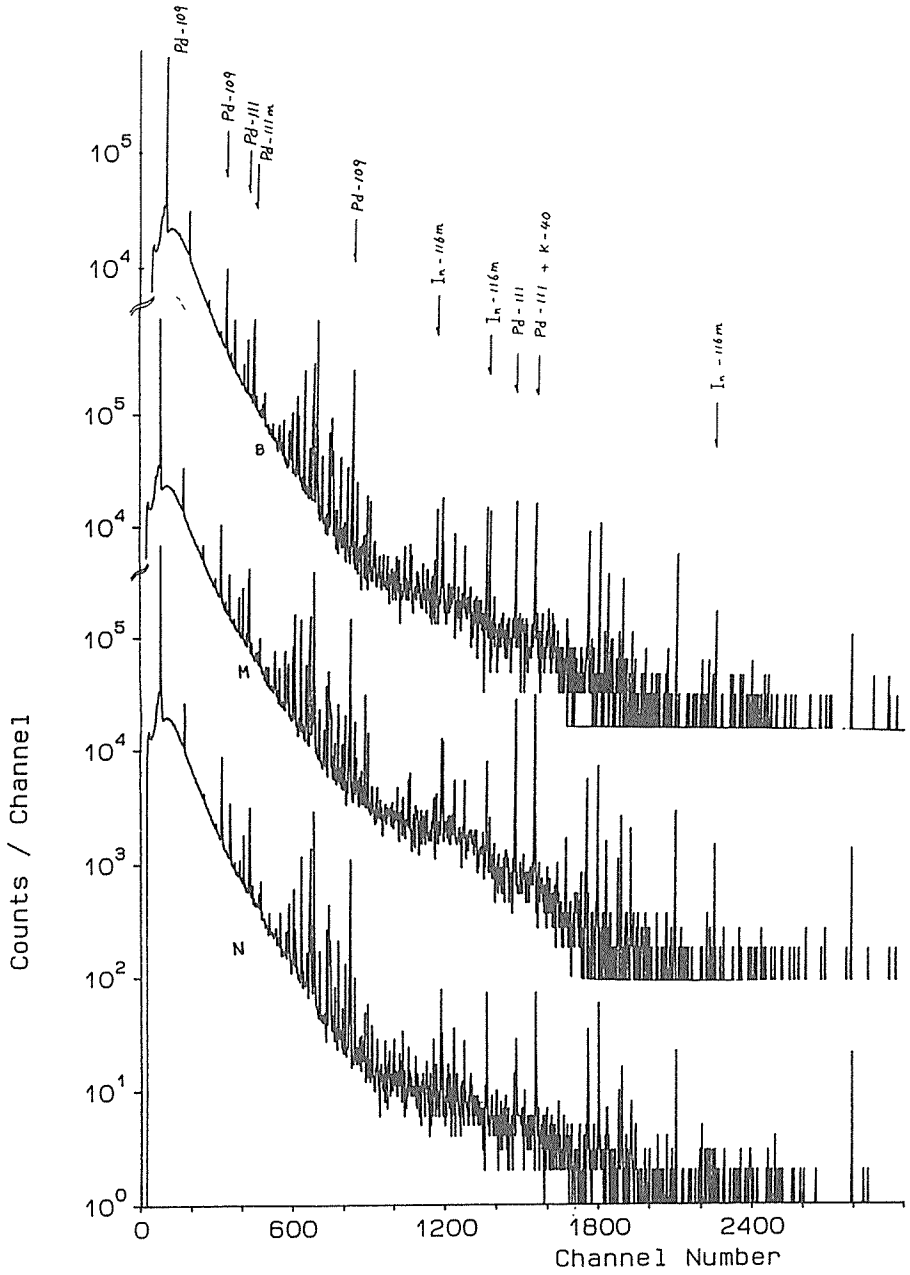

Fig. 3: Gamma ray energy spectrum at 2 hrs
M: middle in used Pd
B: bottom of used Pd
N: top of non-used Pd

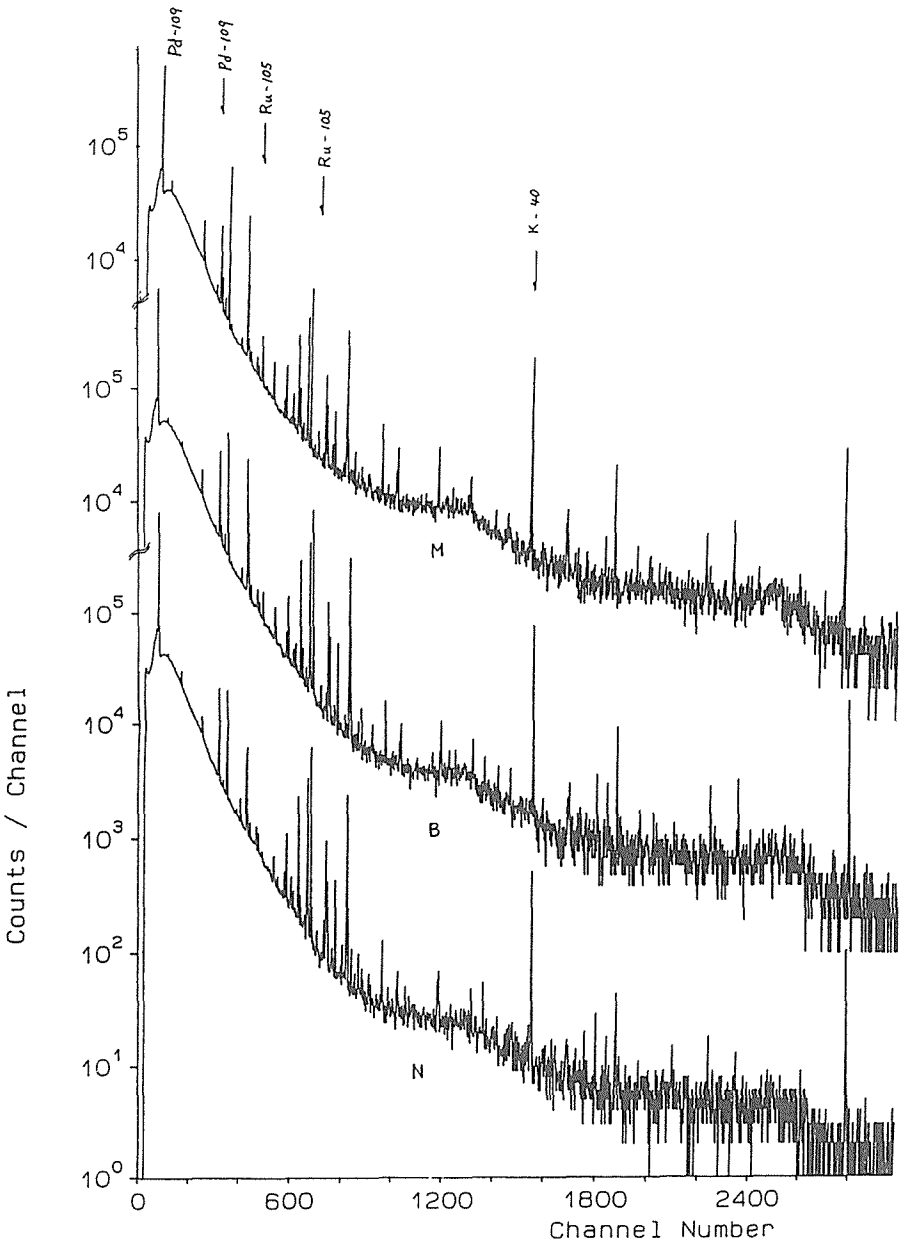

Fig. 4: Gamma ray energy spectrum after sufficient time elapsed
M: middle in used Pd
B: bottom of used Pd
N: top of non-used Pd

hand, two peaks due to ^{105}Ru (675 and 724 keV) appeared. No gamma rays due to ^{116m}In and ^{105}Ru were not observed for the background N that was adjacent to B. Therefore, it is reasonable to consider that those parent isotopes of ^{115}In and ^{104}Ru were not impurities but produced during the electrolysis.

The relative number density was calculated by using Eqs. (1) and (2), and those results are shown in Table 1. The relative number density of ^{108}Pd, deduced from the activity of ^{109}Pd, was in good agreement for the background N with the isotope ratio 26.7 % of the natural palladium. The values of the samples of M and B seemed to somewhat deviate around the natural isotope ratio, but it was difficult to draw a conclusion that the isotope ratio of palladium changed during the cold fusion experiment (7). The relative number density of ^{111}Pd, deduced from the activity of ^{111m}Pd, was also consistent with the natural isotope ratio 11.8 %. However, for the case of ^{111}Pd, the values seemed to be larger for the samples of B and N, but no clear reason was found. Table 1 also shows the relative number density of ^{115}In and ^{104}Ru. Although the number density of ^{115}In was very small, the detection was made possible due to the large cross section. On the other hand, the number density of ^{104}Ru was shown to be large. Since the intensity of gamma rays of ^{105}Ru was not sufficient, the value

Table 1: Relative number density of isotopes

	sotope ratio half-life cross-section	gamma-ray energy (keV)	ralative number density*(%)		
			M	B	N
Pd-109	26.7 13.46h 12.0b	311 781	27.4 26.4	26.4 26.0	26.5 26.5
Pd-111m	11.8% 5.50h 0.020b	390	11.3	10.9	11.1
Pd-111	11.8% 23.4m 0.20b	835	11.2	15.9	14.9
Ru-105	4.44h 0.47b	675	7.91×10^{-2}	1.89×10^{-1}	no
In-116m	54.15m 157.0b	1294	2.02×10^{-5}	1.23×10^{-5}	no

*N/N where N : total number density of Pd

should be considered as the upper limit.

Rolinson et al. (7) reported that the elements of Rh and Ag were produced during cold fusion. This RA method was not applicable to those elements because the half-life of the neutron-induced radioactive isotopes and the cross section were inadequate.

5. Discussion

The nuclear transmutation of the elements was explained by The Nattoh Model (2). According to the model, an extremely compressed hydrogen-cluster is formed in the metal,

in which electrons are degenerated and interconnected with each other. The special state is called the "itonic" state. During that state, nuclear reaction processes including fusion reactions easily take place. The nuclear transmutation also can be induced in the itonic state through three processes mentioned in Chap. 1.

First, the nuclear transmutation by the electron capture with palladium can be written as follows,

$$^{102}\mathrm{Pd}\ (\ 0.96\ \%) + \mathrm{e} \longrightarrow {}^{102}\mathrm{Rh} \xrightarrow[\mathrm{EC}]{2.9\mathrm{y}} {}^{102}\mathrm{Ru} \tag{3}$$

$$^{104}\mathrm{Pd}\ (10.97\ \%) + \mathrm{e} \longrightarrow {}^{104}\mathrm{Rh} \xrightarrow[\mathrm{EC}]{42.3\mathrm{s}} {}^{104}\mathrm{Ru} \tag{4}$$

$$\beta^- \searrow \quad \mathrm{Pd} \tag{5}$$

$$^{A}\mathrm{Pd} + \mathrm{e} \longrightarrow {}^{A}\mathrm{Rh} \xrightarrow[\beta^-]{} {}^{A}\mathrm{Pd} \tag{6}$$

for A = 105(22.2 %), 106(27.3 %), 108(26.7 %) and 110(11.8 %).

Here, EC and β^- indicate the electron capture and the beta decay, respectively, and the half-life shows a value for the conventional isolated state of the isotope. Since orbital electrons distribute closely to the compound nucleus in the itonic state, the halflives of EC that appear in Eqs. (3) and (4) should drastically be shortened. Furthermore,the beta decay should strongly be surpressed by the same reason so that the schemes of Eqs. (5) and (6) should be ruled out. As resulted, the stable isotopes of ^{104}Ru and ^{102}Ru can be generated by the electron capture. Since the isotope abundance of ^{102}Pd was small and the half-life of ^{102}Ru was fairly long, the production of ^{102}Ru was not recognized here.

Second, palladium can be transmuted by the deuteron capture as follows,

$$^{102}\mathrm{Pd} + \mathrm{d} \longrightarrow {}^{104}\mathrm{Ag} \xrightarrow[\mathrm{EC}/\beta^+]{69.8\mathrm{m}} {}^{104}\mathrm{Pd} \tag{7}$$

$$^{104}\mathrm{Pd} + \mathrm{d} \longrightarrow {}^{106}\mathrm{Ag} \xrightarrow[\mathrm{EC}/\beta^+]{24\mathrm{m}} {}^{106}\mathrm{Pd} \tag{8}$$

$$^{105}\mathrm{Pd} + \mathrm{d} \longrightarrow {}^{107}\mathrm{Ag} \tag{9}$$

$$^{106}\mathrm{Pd} + \mathrm{d} \longrightarrow {}^{108}\mathrm{Ag} \xrightarrow[\mathrm{EC}/\beta^+]{2.39\mathrm{m}} {}^{108}\mathrm{Pd} \tag{10}$$

$$^{108}\mathrm{Pd} + \mathrm{d} \longrightarrow {}^{110}\mathrm{Ag} \xrightarrow[\mathrm{EC}]{24.6\mathrm{s}} {}^{110}\mathrm{Pd} \tag{11}$$

$$^{A}\mathrm{Pd} + \mathrm{d} \longrightarrow {}^{A+2}\mathrm{Ag} \xrightarrow[\beta^-]{} {}^{A+2}\mathrm{Pd} \tag{12}$$

for A = 106, 108 and 110.

Here, the stable isotopes of Pd can be generated through Eqs. (7), (8), (10) and (11).

Therefore, the isotope ratio can change by the deuteron capture.

Two or three deuterons can almost simultaneously absorbed in the itonic state. The nuclear transmutation can be written as follows,
for two deuterons capture,

$$^{105}\mathrm{Pd} + 2\mathrm{d} \longrightarrow {}^{109}\mathrm{Cd} \xrightarrow[\mathrm{EC}]{464\mathrm{d}} {}^{109}\mathrm{Ag} \tag{13}$$

$$^{A}\mathrm{Pd} + 2\mathrm{d} \longrightarrow {}^{A+4}\mathrm{Cd} \tag{14}$$

for A = 102, 104, 106, 108 and 110.

and for three deuterons capture,

$$^{A}\mathrm{Pd} + 3\mathrm{d} \longrightarrow {}^{A+6}\mathrm{In} \xrightarrow{\mathrm{EC}/\beta^{+}} {}^{A+6}\mathrm{Cd} \tag{15}$$

for A = 102, 104, 105, 106, 108 and 110,

$$^{A}\mathrm{Pd} + 3\mathrm{d} \longrightarrow {}^{A+6}\mathrm{In} \xrightarrow{\beta^{-}} {}^{A+6}\mathrm{Sn} \tag{16}$$

for A = 106, 108 and 110.

Here, Eqs. (12) and (16) should be special due to the following beta decay. Unlike the electron capture shown by Eq. (6), the deuterons capture themselves should proceed here, but the itonic state of the induced isotopes should remain for a moment. The special state should slowly decay as the itonic cover fades. As resulted by the deuteron capture, the isotopes of Ag and Cd can be newly generated. However, they were not recognized in this RA analysis due to the small cross section of the neutron capture and/or the inadequately long or short life-time. On the other hand, the stable isotope of ^{115}In that was observed by the RA method cannot be genarated by Eq. (16). Therefore, a complex nuclear transmutation by the deuteron capture can be introduced here as follows,

$$\binom{^{104}\mathrm{Pd}}{^{110}\mathrm{Pd}} + \binom{3\mathrm{d}}{3\mathrm{d}} \longrightarrow 2^{113}\mathrm{In} \tag{17}$$

$$\binom{^{106}\mathrm{Pd}}{^{108}\mathrm{Pd}} + \binom{3\mathrm{d}}{3\mathrm{d}} \longrightarrow 2^{113}\mathrm{In} \tag{18}$$

$$\binom{^{108}\mathrm{Pd}}{^{110}\mathrm{Pd}} + \binom{3\mathrm{d}}{3\mathrm{d}} \longrightarrow 2^{115}\mathrm{In} \tag{19}$$

Here, two stable isotopes of In can be generated by the nuclear transmutation. The isotope of ^{113}In was not detected due to the small cross section of the neutron capture and the inadequately long and short half-lives (^{114}In ; 3.9 b and 72 sec, and ^{114m}In ; 4.4 b and 49.5 day). Therefore, only ^{115}In was detected with the large cross section and the adequate half-life (^{116m}

In : 157 b and 54.15 min).

It might be surprizing that the nuclear transmutation takes place during the low voltage electrolysis. Especially, for the deuterons capture, the deterons have to overcome the Coulombic barrier. The reason why these processes are possible was explained by The Nattoh Model (2). The key point is the formation of the compressed hydrogen clusters, in which the electrons are degenerated and interconnected to each other to narrow the internuclear distance. Recently, many reports began to be published on the nuclear transmutation (3). It is important to remark that no energetic radiations are emitted (2). The exciting energy of the compound nucleus can be transfered to the vibrational energy of the itonic mesh that covers the nucleus to eventually disappear as thermal energy of the metal.

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