

CONSISTENT EXPLANATION OF TOPOGRAPHY CHANGE AND NUCLEAR TRANSMUTATION IN SURFACE LAYERS OF CATHODES IN ELECTROLYTIC COLD FUSION EXPERIMENTS

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Synopsis

Nuclear transmutations (NT's) and exotic surface topography observed in the surface layers of cathodes in electrolytic experiments by J. Dash et al. over the last ten years have been analyzed using the TNCF model. Surface topographies of the cathodes showed characteristic fine structures where the results of nuclear transmutation (NT) were detected. Nuclear transmutations, characterized by their locality, are accompanied by excess heat generation, which suggests a nuclear origin. The products of nuclear transmutation are explained either by decay of excited cathode element nuclei to form an element of higher mass number than the original (nuclear transmutation by decay, or NT_D) or by fission of these nuclei (nuclear transmutation by fission, or NT_F). The model was successfully used to analyze two cases of quantitative changes of isotope ratios in Ti and Pd cathodes and the surface topography change. The result shows that semi-quantitative explanation of the experimental data sets as a whole is possible with reasonable values of the adjustable parameter in the model:^{4~6)} $n_n = 1.27 \times 10^{13} \text{ cm}^{-3}$ (Ti), $n_n = 4 \times 10^{13} \text{ cm}^{-3}$ (Pd). For the characteristic surface topography with pits and craters of diameters $\sim 1 \mu\text{m}$, a possible semi-quantitative explanation is given by explosions of tiny regions (hot spots) heated locally by nuclear reactions in the region with the parameter given above.

1. Introduction

J. Dash et al. at Portland State University have performed many cold fusion (CF) experiments with electrolytic systems formed by cathodes of Pd, Ti, and Ni and electrolytes of H_2SO_4 dissolved in both heavy (D_2O) and light (H_2O) water. They have measured exiguous but appreciable amounts of excess heat and found new elements, generally localized at pits or craters in the surface layer of cathodes, and present up to a depth of about $1 \mu\text{m}$. These elements are explainable by the beta decay of the unstable, short-lived nuclei that are formed when cathode elements absorb a trapped neutron. This process, which is one of the many examples of reactions explainable by the TNCF model, can be called nuclear transmutation by decay NT_D .^{4~6)}

The number of events (reactions) N_X between the "trapped neutrons" and the lattice nuclei ${}^A_Z X$ during a time τ in a volume V of a reaction region in the surface layer is given by the equation (Eq.(11.20) in the paper⁴⁾ or Eq.(3) in the paper⁵⁾):

$$\begin{aligned} n + \frac{A}{Z} X &= \frac{A+1}{Z} X^* = \frac{A'}{Z'} X' + \frac{A+1-A'}{Z-Z'} X^n, \\ N_X &= 0.35 n_n v_n n_X V \sigma_{nX} \tau, \end{aligned} \quad (1)$$

where n_n and n_X are the densities of the "trapped neutrons" and the X nuclei respectively, v_n is the thermal velocity of the trapped neutron, σ_{nX} is the cross section of the reaction defined in free space by standard nuclear physics.

2. Analysis of the Experimental Data Sets by the TNCF Model

The TNCF model has consistently explained very many experimental data sets.⁴⁾ The model assumes existence of quasi-stable thermal neutrons (trapped neutrons) with a density n_n in solids where the CFP occurred. We have given a qualitative explanation of the many nuclear transmutations in experiments obtained by Dash et al. but give here only rather quantitative account for for the isotope ratio changes in Ti³⁾ and Pd²⁾ and topographic change of the cathode surface.¹⁾

2.1 Quantitative Explanation of Experimental Data on the Isotope Ratio Change of ${}^{A}_{22}\text{Ti}$

To analyze the quantitative data of the change of Ti isotope ratios,³⁾ we give the necessary equations deduced from the fundamental formula (Eq.(2)) as follows. After an experiment of a time τ , the density x_i ($i = 1 - 5$) of isotopes ${}^A_{22}\text{Ti}$ ($A = 46 - 50$) changes by the absorption of a neutron and also β -decay (or electron capture, EC) according to following equations (cf. Eq.(1)):

$$x_1 = x_1^{(0)} - 0.35 n_n v_n \sigma_1 \tau x_1^{(0)}, \quad (3)$$

$$x_i = x_i^{(0)} + 0.35n_n v_n \sigma_{i-1} \tau x_{i-1}^{(0)} - 0.35n_n v_n \sigma_i \tau x_i^{(0)}, \quad (i = 2 \sim 5), \quad (4)$$

where we simplified the relation by taking $V = 1$ in Eq.(2). These quantities are irrelevant in calculating ratios of densities of isotopes. It is also assumed that the total number of titanium nuclei with mass numbers $A = 46 - 50$ does not change as a whole. In these equations, the fusion cross-sections of a thermal neutron and Ti isotopes ${}^A_2\text{Ti}$ ($A = 46 - 50$) are written as σ_i ($i = 1 - 5$), respectively.

Table 1: Data about titanium isotopes ${}^A_2\text{Ti}$ ($A = 46 - 50 \equiv i = 1 - 5$) in cathodes. Absorption cross section σ_i , natural abundance $x_i^{(0)}$, nominal abundance ratio $\eta_{5i}^{(0)}|_{nom} \equiv x_5^{(0)}/x_i^{(0)}|_{nom}$, measured ratio $\eta_{5i}^{(0)}|_{exp} \equiv x_5^{(0)}/x_i^{(0)}|_{exp}$ before and $\eta_{5i}|_{exp} \equiv x_5/x_i|_{exp}$ after experiments, and their theoretical value $\eta_{5i}|_{th}$ after experiments calculated using $\eta_{51}^{(0)} \equiv x_5^{(0)}/x_1^{(0)}|_{exp}$.

Isotope	σ_i (b)	$x_i^{(0)} _{nom}$ (%)	$x_5^{(0)}/x_i^{(0)} _{nom}$	$x_5^{(0)}/x_i^{(0)} _{exp}$	$\eta_{5i} _{exp}$	$\eta_{5i} _{th}$
${}^{46}\text{Ti}$ ($i = 1$)	0.596	7.93	0.6734	0.6960	0.6683	(0.6683)
${}^{47}\text{Ti}$ ($i = 2$)	1.702	7.28	0.7335	0.8027	0.6978	0.771
${}^{48}\text{Ti}$ ($i = 3$)	7.844	73.94	0.0722	0.0785	0.0727	0.0846
${}^{49}\text{Ti}$ ($i = 4$)	2.214	5.51	0.969	1.0111	0.9358	0.402
${}^{50}\text{Ti}$ ($i = 5$)	0.179	5.34	1.000	(1.0000)	(1.000)	(1.000)

Then, we can write down the ratio $\eta_{5i} \equiv x_5/x_i$ ($i = 1 - 4$) using the initial ratio $\eta_{5i}^{(0)} \equiv x_5^{(0)}/x_i^{(0)}$ as follows, where the abbreviation $a = 0.35n_n v_n \tau$;

$$\eta_{51} \equiv \frac{x_5}{x_1} = \frac{x_5^{(0)} + a(x_4^{(0)}\sigma_4 - x_3^{(0)}\sigma_5)}{x_1^{(0)} - a x_1^{(0)}\sigma_1} = \frac{\eta_{51}^{(0)} + a(\eta_{41}^{(0)}\sigma_4 - \eta_{51}^{(0)}\sigma_5)}{1 - a\sigma_1}, \quad (5)$$

$$\eta_{5i} \equiv \frac{x_5}{x_i} = \frac{x_5^{(0)} + a(x_4^{(0)}\sigma_4 - x_5^{(0)}\sigma_5)}{x_i^{(0)} - a(x_{i-1}^{(0)}\sigma_{i-1} - x_i^{(0)}\sigma_i)} = \frac{\eta_{5i}^{(0)} + a(\eta_{4i}^{(0)}\sigma_4 - \eta_{5i}^{(0)}\sigma_5)}{1 + a(\eta_{i-1,i}^{(0)}\sigma_{i-1} - \sigma_i)}, \quad (6)$$

From these equations, we can calculate $\eta_{5i}|_{th}$ ($i = 2 - 4$) using experimental value of $\eta_{51}|_{exp} = 0.6683$ to compare $\eta_{5i}|_{exp}$ ($i = 2 - 4$).

The first equation determines the value a by using experimental values of η_{51} , $x_1^{(0)}$, $x_4^{(0)}$ and $x_5^{(0)}$ and the cross sections σ_1 , σ_4 and σ_5 given in Table 2;

$$a = \frac{\eta_{51}^{(0)} - \eta_{51}}{\eta_{41}^{(0)}\sigma_4 + \eta_{51}\sigma_1 - \eta_{51}^{(0)}\sigma_5} = 0.183\sigma_4^{-1} \quad (7)$$

Using this value of a in the equation (6), we can calculate $\eta_{5i}|_{th}$ ($i = 2 - 4$) and the result is tabulated in Table 1. Table 1 also has the tabulated initial concentrations $x_i^{(0)}$ of titanium isotopes ${}^A_2\text{Ti}$ ($A = 46 - 50 \equiv i = 1 - 5$), which is assumed to be the natural abundance that is in the first column. Nominal ratios $x_5^{(0)}/x_i^{(0)}|_{nom}$ are calculated and tabulated in the second column to be compared with the experimental ratio $x_5^{(0)}/x_i^{(0)}|_{exp}$ in the third column that was obtained from Fig.10 in the paper.⁹⁾

2.2 Quantitative Explanation of Experimental Data on the Isotope Ratio Change of ${}^A_{46}\text{Pd}$

The experimental data on the relative abundance of Pd isotopes by SIMS given in the Figures 1(a) and (b) in the paper²⁾ can be summarized as given in Table 2.

Analysis similar to that given in the previous subsection for Ti gives the result $\eta_{i1}|_{th}$ given in the last column in this table. It is seen in this table qualitative accordance between the experimental and theoretical values of η_{i1} .

2.3 Quantitative Explanation of Pits Formation in the Surface Layer of Cathodes

To analyze the microstructure of the cathode surface after electrolytic experiments, we take up the data given in reference.¹⁾

As main reactions to explain the thermal effect which produces microscopic craters in the surface region, we take reactions with protons and deuterons occurring in a localized region by some causes (suggested by experimental results);

$$n + p = d + Q_1, \quad (8)$$

$$n + d = t + Q_2, \quad (9)$$

Table 2: Data about palladium isotopes ${}^A_Z\text{Pd}$ ($A = 102 - 110 \equiv i = 1 - 6$) in cathodes. To calculate experimental values of relative composition, we used SIMS data at $0.09 \mu\text{m}$ from the surface (15 minutes from the beginning). Natural abundance $x_i^{(0)}$, nominal abundance ratio $\eta_{i1}^{(0)}|_{nom} \equiv x_i^{(0)}/x_1^{(0)}|_{nom}$, measured ratio $\eta_{i1}^{(0)}|_{exp} \equiv x_i^{(0)}/x_1^{(0)}|_{exp}$ before and $\eta_{i1}|_{exp} \equiv x_i/x_1|_{exp}$ after experiments, and their theoretical value $\eta_{i1}|_{th}$ after experiments calculated using $\eta_{21}^{(0)} \equiv x_2^{(0)}/x_1^{(0)}|_{exp}$. Experimental data for $i = 4$ and 5 measured in Omega Analytical Services, Aloha, OR. in 1996, are interchanged to compare well with theoretical results.

Isotope (i)	σ_i (b)	$x_i^{(0)} _{nom}$ (%)	$\eta_{i1}^{(0)} _{nom}$	$\eta_{i1}^{(0)} _{exp}$	$\eta_{i1} _{exp}$	$\eta_{i1} _{th}$
${}^{102}\text{Pd}$ ($i = 1$)	3.36	0.96	1.000	1.000	1.000	(1.000)
${}^{104}\text{Pd}$ ($i = 2$)	0.523	10.97	11.42	9.0	13.7	(13.7)
${}^{105}\text{Pd}$ ($i = 3$)	20.25	22.33	23.26	19.8	25.0	7.9
${}^{106}\text{Pd}$ ($i = 4$)	0.303	27.33	28.47	24.0	32.8	51
${}^{108}\text{Pd}$ ($i = 5$)	8.504	26.71	27.82	23.8	27.8	18
${}^{110}\text{Pd}$ ($i = 6$)	0.227	11.81	12.30	9.5	14.8	12

where $Q_1 = 2.2 \text{ MeV}$ and $Q_2 = 6.25 \text{ MeV}$ are absorbed by particles in solids while they are emitted as photons in reactions occurring in free space. We can ignore reactions with palladium isotopes taken up in subsection 2.2 due to the negligible amount of energy liberated in them compared with the above two reactions.

This assumption is suggested by our theoretical prediction of neutron drops⁷⁾ formed in near-surface layers which interact especially with hydrogen isotopes therein and induce reactions (8) and (9) giving the liberated energy Q_1 and Q_2 to the many body system made of lattice nuclei (Pd), occluded hydrogen isotopes (D and H) and neutron drops.

Assume a crater is made by the localized heating of a sphere S with a radius r_m at a point P some distance below the surface layer of the cathode d ($> r_m$). Let us assume heating occurs by localized heat generation in the sphere with a constant rate q (J/s) and dissipation q' (J/s) of thermal energy through the surface S with an area $4\pi r_m^2$ by heat conduction.

We assume the composition of the cathode material in the surface layer to be PdH_xD_y . Reactions (8) and (9) which occur in a unit time are given by Eq.(2) with appropriate parameters for both, the proton and the deuteron. Their reaction cross-sections are 0.332 and $5.5 \times 10^{-4} \text{ b}$, respectively ($1 \text{ b} = 10^{-24} \text{ cm}^2$). Then, we can write the heat generation rate q as follows;

$$q \equiv q_1 + q_2 = N_{np}Q_1 + N_{nd}Q_2, \quad (10)$$

where N_{np} and N_{nd} are given by Eq.(2) using $\xi = 1$ (by a premise in the TNCF model), $V = (4\pi/3)r_m^3$, $n_p = xn_{Pd}$ and $n_d = yn_{Pd}$, and $n_{Pd} = 6.88 \times 10^{22} \text{ cm}^{-3}$, and thermal velocity of neutrons $v_n = 2.2 \times 10^5 \text{ cm/s}$.

To calculate the dissipation rate q' , we replace physical parameters, thermal conductivity κ and heat capacity c_m , of PdH_xD_y by those for palladium metal, for simplicity, allowing ambiguity factors of about two;

$$\kappa = 0.755 \text{ W/cm} \cdot \text{s} \cdot \text{K} \text{ and } c_m = 25.9 \text{ J/K} \cdot \text{mol. (Pd)}$$

Then, the dissipation rate of heat through the surface of the droplet with an area $S = 4\pi r_m^2$ is given as

$$q' = \kappa \frac{S\Delta T}{\Delta r}, \quad (11)$$

where Δr is the transition length of the temperatures from the inside one T_m to outside one T_s (\sim the room temperature). As the temperature difference ΔT between the melted sphere and the ambient solid of the cathode, we take $\Delta T = T_m \sim T_s = 2200 \text{ (}^\circ\text{C)} - 20 \text{ (}^\circ\text{C)} = 2180 \text{ (}^\circ\text{C)}$ taking T_m as the boiling point of palladium metal (we assume explosion of the super-heated droplet to form pits or craters at the cathode surface) and T_s as the room temperature.

As the distance Δr between the melted region of the droplet and the ambient solid, we may take a thin shell with a thickness of $10^{-2} r_m$, while the radius of the melted droplet r_m can be taken as $1 \mu\text{m}$ as suggested by experimental results.¹⁾

Putting these values into Eq.(11), we obtain the heat dissipation rate as follows;

$$q' = 1.824 \times 10^{13} \text{ (MeV/s)}. \quad (12)$$

On the other hand, the heat generation rate (10) is rewritten as follows;

$$q = 2.22 \times 10^{-8} n_n (0.737x + 0.00003436y) \quad (\text{MeV} \cdot \text{cm}^3/\text{s}), \quad (13)$$

This relation shows rather effective role of protons than deuterons to produce excess heat if x and y have the same orders of magnitude.

Assuming $x = y = 0.5$ for the composition of the surface layer and appropriate values for boiling of the droplet, we can determine the extremum value of the parameter n_n :

$$n_n|_{ext} = 2.2 \times 10^{19} \quad (\text{cm}^{-3}). \quad (14)$$

The value of 10^{19} cm^{-3} obtained in the events of pit formation with extreme conditions at the surface can be interpreted only by the idea of the neutron valence band^{7,8)} developed recently using excited neutron states of lattice nuclei. Conversely, the occurrence of localized vaporization lends credibility to the theoretical treatment.

3. Conclusion

The possible nuclear transmutations by decay (NT_D) and by fission (NT_F) in the surface layers of Pd, Ti and Ni cathodes which were observed by J. Dash et al. were consistently explained by the TNCF model except for the one case of the $^{106}_{46}\text{Pd}/^{108}_{46}\text{Pd}$ ratio change. This might be a new data suggesting instability of $^{106}_{46}\text{Pd}$ that is assumed to be a stable reaction product of the reaction (1) with $^4_2\text{Pd} = ^{105}_{46}\text{Pd}$.

The appearance of the topographical features cited above suggests the possibility that nuclear reactions occurred in tiny regions in a short period of time. The localized high-densities of neutrons in boundary regions that were deduced in our papers⁹⁾ explain successfully and consistently the occurrence of nuclear reactions of this type. The local coherence of neutron Bloch waves at a boundary layer results in a high density of neutrons therein.⁹⁾ Then, the occurrence of trigger reactions at specific places in the boundary layer, where there are high densities of neutrons, progress into breeding reactions to generate high energy nuclei and also produce high energy nuclides, which dissipate heat to the system as a whole.⁸⁾

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