Recent Res. Devel. Electroanal. Chem., 2 (2000): 35-46 ISBN : 81– 86846 – 94 – 8

Electroanalytical chemistry in cold fusion phenomenon

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ABSTRACT

Fundamental characteristics of the cold fusion phenomenon (CFP) are explained from the present point of view based on the pile of complex experimental facts obtained in these eleven years after its discovery. Surface nature of the reactions and qualitative reproducibility, two of remarkable characteristics of CFP are explained by a model (TNCF model) with a single adjustable parameter. The Premises assumed in the model indicate importance of atomic processes on the surface of electrodes to realize CFP which should be investigated by electroanalytical chemistry.

1. INTRODUCTION

The cold fusion phenomenon (CFP), or more precisely "Nuclear Reactions and Accompanied Phenomena in Solids including High Density Hydrogen Isotopes", was discovered in 1989 by M. Fleischmann et al.[1] in electrochemical experiments with Pd cathode and an electrolyte LiOD in heavy water D_2O where they expected the fusion of two deuterons occurs by the presumed high pressure of deuterium occluded in the Pd cathode as high as 10^{27} atm. This presumption has induced long lasting confusion in the

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interpretation of the experimental results among both supporters and critiques.

The observed phenomenon had included such sporadic events as the excess heat generation and tritium and neutron productions in several experimental sets with null results in many other sets.

After more than 11 years from the discovery, it is recognized by serious researches that CFP is a rather complex phenomenon including the excess heat, transmuted nuclei, tritium, helium-4, neutron, gamma ray productions and occurring in electrolytic, gas contact, gas discharge and other systems where are solids with high density hydrogen isotopes (not only deuterium D but also protium H).

The electrolytic system is, however, used most often in CF experiments with positive results. Investigation of experimental results shows the cause of this fact and also characteristics of CFP. A unified and systematic explanation of CFP has been given using a model[2] proposed by the author with a single parameter and quantum mechanical explanation of Premises used in the model has been investigated progressively.[3~ 5] It becomes clear that electroanalytical chemistry will give fundamental knowledge related with realization of CFP.

2. EXPERIMENTAL FACTS SHOWING SURFACE NATURE OF THE COLD FUSION PHENOMENON (CFP)

From the present point of view formed with abundant experimental data accumulated in these more than eleven years after its discovery[1] of CFP in 1989, the presumption in the first stage of d - d fusion in PdD_x ($x \simeq 0.6 \sim 1.0$) was false as a principal cause of CFP even if it occurs in special situations. The imbalance of products, the excess heat, neutron, helium-3, tritium, proton, helium-4 and gamma ray inexplicable by the presumed reactions has been insoluble riddles causing unfruitful dispute between supporters and critiques.

In addition to this apparent imbalance of the products contradicting to the presumed reactions, there has been discovered a new decisive clue about nature of nuclear reactions and position where they occur which have shown difficulty of its explanation by the d - d fusion reaction.

In the CFP, such events as nuclear transmutation and helium-4 production have specified the position where occur the events. The direct information has been given by localized distribution of the transmuted nuclei around speckled regions in the surface layer of cathodes in electrolytic experiments[6~ 9] and of cathodes in gas discharge experiments.[10,11] The fact that helium-4 atoms observed in the surface layer of cathodes[12,13] are little in its amount compared with those observed in gas phase[14,15] shows also, somewhat indirectly, surface nature of reactions generating helium-4.

It is interesting to notice that there are two kinds of the nuclear transmutation (NT) observed in experiments; one is so-called "NT by decay" (NT_D) which is explained by decay of a nucleus formed from pre-existed nucleus absorbing a neutron. Another is so-called "NT by fission" (NT_F) which is explained by fission of a nucleus formed from pre-existed nucleus absorbing several neutron.

Other evidences showing surface nature of nuclear reactions causing CFP are distribution changes of minor components in cathode where evolved the excess heat.[16,17]

It is necessary to give a little detailed explanation of the surface nature of reactions to depart completely from the false presumption of the d - d fusion reactions as a principal cause of CFP and to resolve the confusion induced by it in the research community. In the following explanation of several of typical experimental data, discrimination of NT_D and NT_F is not given without exceptional cases because it is apparent from the species of the product nuclei.

Experiment by J.O'M. Bockris et al.[6]

Bockris and his collaborator performed an experiment[6] in which hydrogen was electrolyzed from water in contact with a palladium electrode (size and shape not described). The electrolyte is not described in the paper.

To indicate whether a new nucleus was formed by means of transmutation or whether it came from the solution as an impurity, they utilized Inductively Coupled Plasma method (ICP) to analyze the materials in the solution. They analyzed the surface in two ways. In the first case, they used X-ray photoelectron spectroscopy (XPS) which gives rise to materials which are only 30 Å from the surface. Then, they also used electron dispersive analysis (EDA) to analyze deeper, i.e. 1 μm.

The concentration and depth of impurities were measured by these methods as a function of electrol-

ysis time to identify their origin. It was found that two different sets of impurities could be observed after 3 weeks, one set within 50Å of the surface and another set different chemical species, about 1 μm inside the cathode metal (Pd).

The impurities measured by XPS (Pt, Si, Zn), which spread no further than a few tens of Å inside the Pd, are originated in the solution by means of electrochemical deposition or adsorption on the electrode surface and subsequent diffusion into the electrode.

On the other hand, the maximum intensity of the signal, observed by EDA, arose from a greater depth than that of XPS. In fact, for Pd, for a 20 keV X-ray beam, the escape depth maximizes at 2 μm inside the Pd and the signals found both after one hour's electrolysis and then after three weeks' electrolysis clearly show that the origin is not outside but inside of the cathode. It is seen that new nuclei have appeared at this depth in the presence of hydrogen at high fugacity.

The concentration of the new elements (Mg, Al, Cl, K, Ca, Ti, Fe, Cu, Zn, and so on) is in the range 1 $\sim$ 10 atomic %. These elements have no relationship to the impurities in the solution.

Experiment by T. Ohmori et al.[7]

A group in Hokkaido University, Japan have been working with the light water system. T. Ohmori et al. observed NT, i.e. increases of $^{57}_{26}$Fe and $^{54}_{26}$Fe, generation of C and S, in light water electrolysis with electrodes Au and Pd and electrolytes Na_2SO_4, K_2CO_3 and KOH in H_2O.

In the electrolysis with a gold electrode and an electrolyte Na_2SO_4 (or K_2SO_4, K_2CO_3, KOH) in H_2O for a week, a notable amount of iron atoms in the range of 1.0×10^{16} to 1.8×10^{17} atoms/cm^2 are detected at surface together with the generation of a certain amount of excess energy. The isotopic abundance of iron atoms, measured at the top surface of a gold electrode, are 6.5, 77.5 and 14.5 % for $^{54}_{26}$Fe, $^{56}_{26}$Fe and $^{57}_{26}$Fe and are obviously different from the natural abundance of 5.84, 91.68 and 2.17 %, respectively. The content of $^{57}_{26}$Fe tends to increase up to 25% in the more inner layers of the electrode.

Experiment by G. Miley et al.[8]

The electrolytic experiments with cathodes of the packed-bed cell (about 1000 microspheres (ms's)) in a electrolyte 1M $LiSO_4$ + H_2O were performed with nickel, palladium and Pd-Ni multilayer cathodes and titanium electrodes. Voltages across the bed were held at about 2 $\sim$ 3 V, with several mA of current, giving an electrical input power of approximately 0.06 W. Inlet-outlet thermocouples provided a measure of the temperature increase of the flowing electrolyte. Positive but often very small increases in temperature across the cell ranging from 0.1 to 4 °C were observed in all cases.

We take up here only one data set, Run #11, where Pd thin-film of 200 μm was on the polystyrene microsphere. The volume and mass of the film was 7.05×10^{-7} cm^3 and 8.46×10^{-6} g, respectively. The number of Pd atoms in the film was 4.76×10^{16}.

The analysis of the sample microspheres by NAA (Neutron Activation Analysis) after an experiment lasted 211 hours showed appearance of elements with yields shown in Table 1.

Experiment by J. Dash et al.[9]

There were two kinds of the Pd cathode used in the experiment by Dash et al.[9] The Pd cathodes were (A) a cold-rolled 0.35 mm-thick polycrystalline sheet and (B) a 5.5×10^{-2} mm thick foil. The anodes were Pt foils of 3×10^{-2} mm thickness in both cases. The Pd cathodes which produced the excess heat were first cleaned ultrasonically in deionized water and then examined with a scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS). We explain here only the experiment A.

The cathodes used in the previous study lasted about 400 h of comparative heat measurements were examined for the nuclear transmutation (NT). This

Table 1: Elements detected by NAA and their amounts per ms (microsphere) (Miley et al.[8])

Element	Al	Cu	V	Ni	Fe	Co	Cr	Zn	Ag
Yield ($\times 10^{-3}$ μg/ms)	233	277	4.06	388	153	2.18	60.4	806	70.6
No. of atoms($\times 10^{13}$/ms)	520	262	4.80	398	165	223	699	742	39.4

study involved electrolysis from two cells in series with the same electrolyte (0.06 mol fraction H_2SO_4) one in H_2O and another in D_2O, and both containing above mentioned Pd cathode and Pt anode.

After about 400 h of electrolysis, the original smooth, shiny surfaces of the cathodes changed to a dull, corrugated topography. Dark spots were present on the lower ends of both cathodes where the shape changes were greatest. EDS spectra of both Pd cathodes in light water (H_2O) and heavy water (D_2O) showed an appreciable amount of Pt and Au.

a) Pd cathode in H_2O. Several regions on the concave side of the Pd cathode, which appeared darker and rougher with more contrast, gave EDS spectra which showed an appreciable amount of Pt and Au, in addition to Pd. It was likely that Pt was plated from the electrolyte where it was due to slow dissolution of the Pt anode.

b) Pd cathode in D_2O. Similar analysis of the bottom of the heavy-water Pd cathode on the concave side also revealed Au in localized regions. The concentration of Au on the heavy-water cathode (e.g. 6 %) appeared to be greater than on the light-water cathode (e.g. 3 %).

Experiment by I. Savvatimova et al.[10]

The researchers in the Institute LUTCH in Podolsk near Moscow have been working in the glow discharge experiments with D_2 and other gases and with cathodes of Pd and other transition metals (the cathode was with thickness of 100 μm). They measured the excess heat, NT (nuclear transmutation) of various isotopes and elements in the surface layer of the multi-layer cathodes. After the discharge of 4 hours, the sample was sent to SIMS (Secondary Ion Mass Spectrometry) and was analyzed its isotope composition there about 3 months later

Here we take up only one data set of an increase of $^{107}_{47}$Ag from 20 to 5000 ppm in the glow discharge with D_2 gas and Pd cathode. The transmuted nuclei of Ag were observed in several localized regions on the surface of the cathode.

Experiment by H. Yamada et al.[11]

To confirm the cold fusion phenomenon under glow discharge condition, a point-to-plane electrode configuration in slightly pressurized (2 atm) deuterium gas for highly non-uniform electric field was employed by Yamada et al.[11] of Iwate University, Japan. A neutron burst took place in 2 runs out of total 37 runs.

Using an optical microscope, black deposit was observed covering the tip surfaces of two positive Pd electrodes which was used in the runs with neutron bursts. X-ray photo-electron spectroscopy (XPS) have revealed the black deposit to be carbon, mixed with palladium on the surface of palladium point electrode. The total amount of carbon impurity in the Pd electrode and in environment deuterium gas did not account for the large amount of carbon on the tip surface of electrode.

Experiment by J.R. Morrey et al.[12]

One of the earliest examples of helium detection was the data by Morrey et al.[12] The Pd sample supplied by Pons was distributed to six laboratories with dummies to blind check the existence of helium in them, the presupposed d - d reaction in volume of the sample suggested it. The result was not decisive to show consistency of the observed amounts of helium and the excess heat (announced by Pons). In their analysis, it was assumed the occurrence of the reaction (13) below and existence of whole generated helium in the cathode. (If the responsible reaction occurred in volume, the generated helium was expected to remain in the sample.)

They could not prove that the minimal excess heating in one of the rods (rod 5) reported by Fleischmann and Pons could be attributed to the formation of ^{4}He on the assumption that the ^{4}He had been generated by the reaction (13).

Though this result[12] was sometimes supposed to show a decisive disproof against the cold fusion, we have given a consistent explanation of amounts of the excess heat and ^{4}He assuming the observed amount of ^{4}He in the sample is 3 % of the whole generated in the surface layer.[2,18]

Experiment by C-C. Chien et al.[13]

Detection of tritium, ^{3}He and ^{4}He had been done simultaneously by Bockris' group in Texas A&M University in an electrolytic system Pd/D/Li.[13] They have given the first reliable observation of simultaneous tritium and helium-4 production and absence of ^{3}He in this system.

The cathode was a Pd cylinder with a size 1 cmϕ and 1.6 cm long, the anode was a Pt wire 0.5 mmϕ and 100 cm, electrolyte is 0.1M LiOD in D_2O in a

cell with a volume of 80 cm^3. The distance between the working and counter electrodes was 2 mm and yielded an electrolyte resistance of about 10 *Omega*. The electrolyte liquid was checked for H_2O and determined its content in the liquid as 1.6% after three days and 9.8% after 22 days of electrolysis. Electrolysis was took place in periods of up to 750 h.

Remarkable data explained here are those of helium.

It was concluded that ^3He was not detected while tritium and ^4He were detected several times well above the background level.

Local distribution of ^4He is observed in the Pd cathode within 1 mm from surface. Excess ^4He was observed in 9 out of the 10 electrolyzed Pd samples cut out from electrodes which produced tritium. The amounts ranged from 0.4 to 166.8×10^{11} atoms/g for samples near the surface of the cathode and $0.4 \sim 2.5 \times 10^{11}$ atoms/g for those away from the surface.

The analysis by the TNCF model has shown the amounts of tritium and helium-4 detected in this experiment are consistent if the amount of ^4He detected in the sample is few % of the total generated in the surface region.[19] This is consistent with the assumption made in the analysis of the data by Morrey et al.[12] and also with the known behavior of helium in solids.

Experiment by M.H. Miles et al.[14]

Third data showing the generation of helium-4 in the Pd/D/Li electrolytic system was obtained by Miles et al.[14] analyzing the gas from the system. The amount of ^4He in terms of the excess heat was $10^{11} \sim 10^{12}$ helium atoms per 1 J.

The amount of the excess heat per second (watt) plotted against the amount of ^4He in the gas per second shows that the excess heat is about 1.5 times more than the expected value from the reaction (13) below if the energy 23.8 MeV of the photon is all thermalized in the system even if the reaction has unrealistically small branching ratio of 10^{-7}. They also had shown using X ray film and Geiger counter that radiation was observed only when the excess heat was generated.

Experiment by E. Botta et al.[15]

The Trino group in Italy performed measurements of the ^4He content in samples with high loading ratio D/Pd up to 0.7. A successful measurement[15] of ^4He was reported at ICCF6 (October, 1996) and the succeeding report was given at ICCF7 (April, 1998) with addition of a little more data on the sample composition, the former of them is taken up in this explanation.

In the experiment where observed ^4He in the desorbed gas from the D_2 loaded Pd sample, the sample was a Pd sheet of a size $8 \times 1 \times 1 \cdot 10^{-2}$ cm^3 plated by gold at both ends for a length of 1.5 cm (thickness of gold ~ 15 μm) and clamped there by two Cu electrodes inside the cell with a volume of 166 ± 1 cm^3.

After the vacuum control up to 1×10^{-6} mbar, D_2 gas was introduced in the cell to a final pressure of 2.7 bar and the definite current up to 440 A was applied through the sample for a duration up to 2 hours, at its end the gas analysis was performed by a high resolution mass spectrometer. In a measurement at 117 h from the D_2 gas introduction where the D/Pd ratio was 0.80 ± 0.02 and the mean current 330 A (440 A at maximum) with a duration 0.4 h, an amount of $(5.3 \pm 0.7) \times 10^{18}$ ^4He was observed.

This is a gas contact experiment and shows a different phase of reactions in CFP from that in electrolytic systems.

Experiment by M. Okamoto et al.[16]

M. Okamoto of Tokyo Institute of Technology, Japan had been working with Pd/ D/ Li system. In an experiment, M. Okamoto et al. observed the excess heat and the changes of key and minor elements in the cathode. We take up here, the nuclear transmutation from Al into Si in the surface layer on the cathode in Pd/ D_2O + LiOD system. The change of the density of the elements (up to 80 % for Al) occurred in the surface layer with a thickness of ~ 1 μm.

Experiment by T. Bush et al.[17]

R.T. Bush and R. Eagleton observed the excess heat and the nuclear transmutation of Rb into Sr in $Ni/H_2O + Rb_2CO_3$ system (with Ni sponge cathode). The reaction supposed to occur in the system was that of (5.17) with rubidium isotope $^A_{37}Rb$,

$$n + {}^A_{37}\mathrm{Rb} = {}^{A+1}_{37}\mathrm{Rb}^* = {}^{A+1}_{38}\mathrm{Sr} + e^- + \bar{\nu}_e,$$

in the surface layer of Rb on the Ni cathode. Natural abundance of $^{85}_{37}Rb$ and $^{87}_{37}Rb$ are 72.15 and 27.85 % and decay times of $^{86}_{37}Rb$ and $^{88}_{37}Rb$ are 19.5 d and 17.8

m, respectively.

3. A POSSIBLE EXPLANATION OF THE SURFACE REACTION BY THE TNCF MODEL

The variety or diversity of events in the cold fusion phenomenon (CFP) from the excess heat generation, an effect of statistical average of individual atomic processes, to transmuted nuclei, direct manifestation of the result of individual nuclear process, has revealed a complicated mechanism of CFP not confined to a special reaction like the d - d fusion reaction. It is reasonable, then in these situations, to use a model with some adjustable parameters to disclose science of CFP if it be possible to explain various events of CFP systematically and consistently from a unified point of view.

A model with a single adjustable parameter n_n based on the trapped neutron catalyzed fusion of nuclei in solids including hydrogen isotopes was proposed by the present author[2] seven years ago. The model (TNCF model) suggested by null results in experiments without background neutrons has been successful to give a systematic explanation of the whole data of events in CFP even if there remain some ambiguity and inconsistency accompanied with inevitable errors and ambiguity in experimental data.

Emphasis should be laid on the basis of the model, the null results obtained by big institutions like LLNL and Argonne National Laboratory to check positive results in a condition with extremely low neutron background and enhanced positive results obtained with artificial sources of thermal neutrons.[2]

The TNCF model is based on the following common Premises assumed for explanation of whole events of CFP in all materials.

3.1 PREMISES OF THE TNCF MODEL

The TNCF model is a phenomenological one and the basic premises (assumptions) extracted from experimental data sets are summarized as follows:[2]

Premise 1. We assume a priori existence of the quasi-stable trapped neutron with a density n_n in pertinent solids, to which the neutron is supplied essentially from the ambient neutron at first and then by breeding processes (explained below) in the sample.

This Premise reflects the fact that any experiment done in environment without background neutron has not given a positive result.

The density n_n is an adjustable parameter in the TNCF model which will be determined by an experimental data set using the supplementary premises which will be explained below concerning reactions of the trapped neutron with other particles in the solids. The quasi-stability of the trapped neutron means that the neutron trapped in the crystal does not decay until a strong perturbation destroys the stability while a free neutron decays with a time constant of 887.4 $\pm$ 1.7 s.

Variety of the determined values of n_n for positive results of various events in CFP is considered as due to ability of the sample to trap thermal neutrons determined by atomic processes[18] of hydrogen isotope occlusion in and alkali metal plating on the cathode. The parameter n_n, therefore, distribute from zero (in null result) to a finite maximum value (in optimum positive result).

Premise 2. The trapped neutron in a solid reacts with another nucleus in the surface layer of the solid, where it suffers a strong perturbation, as if they are in vacuum. We express this property by taking the parameter (the instability parameter) ξ, defined in the relation (18) written down below, as $\xi = 1$.

This premise reflects the surface nature of reactions explained in the previous section.

We have to mention here that the instability parameter ξ in the surface layer is not known at all and it can be, as noticed recently, more than one ($1 \leq \xi$) making the determined value of the parameter n_n smaller. This ambiguity is suggested by various anomalous changes of decay character of radioactive isotopes and by unexpected fission products in the surface layer.

Premise 3. The trapped neutron reacts with another perturbing nucleus in volume by a reaction rate given in the relation (18) below with a value of the instability parameter $\xi \leq 0.01$ due to its stability in the volume (except in special situations such as at very high temperature as 3000 K).

Following premises on the measured quantities of nuclear products and the excess heat are used to calculate reaction rates, for simplicity:

Premise 4. Product nuclei of a reaction lose all their kinetic energy in the sample except they go out without energy loss.

Premise 5. A nuclear product observed outside of the sample has the same energy as its initial (or original) one.

This means that if an energy spectrum of gamma-ray photon or neutron are observed outside, it reflects directly nuclear reactions in the solid sample. The same is for the distribution of the transmuted nucleus in the sample. Those spectra and the distributions of the transmuted nuclei are *the direct information* of the individual events of the nuclear reaction in the sample.

Premise 6. The amount of the excess heat is the total liberated energy in nuclear reactions dissipated in the sample except that brought out by nuclear products observed outside.

Premise 7. Tritium and helium measured in a system are accepted as all of them generated in the sample.

The amounts of the excess heat, tritium and helium are accumulated quantities reflecting nuclear reactions in the sample indirectly and are *the indirect information* of the individual events.

Premises about structure of the sample are expressed as follows:

Premise 8. In electrolytic experiments, the thickness ℓ of the alkali metal layer on the cathode surface (surface layer) will be taken as $\ell = 1$ μm (though the experimental evidences show that it is $1 \sim 10$ μm).

This is a premise related most closely with the electroanalytical process and will be discussed more in Section 5.

Premise 9. The mean free path or path length ℓ_t of the triton with an energy 2.7 MeV generated by n + ^{6}Li fusion reaction will be taken as $\ell_t = 1$ μm irrespective of material of the solid. Collision and fusion cross sections of the triton with nuclei in the sample will be taken as the same as those in vacuum.

Premise 10. Efficiency of detectors will be assumed as 100 % except otherwise described, i.e. the observed quantities are the same as those generated in the sample and to be observed by the detector in experiments if there are no description of its efficiency.

A premise will be made to calculate the number of events N_Q producing the excess heat Q .

Premise 11. In the calculation of the number of an event (a nuclear reaction) N_Q producing the excess heat Q , the average energy liberated in the reactions is assumed as 5 MeV unless the reaction is identified: N_Q = Excess heat Q (MeV)/ 5 (MeV).

Following relation combines two energy units, the million-electron-volt (MeV) used in nuclear physics and the joule (J) used in calorimetry:

$$1 \text{ MeV} = 1.6 \times 10^{-13} \text{ J}, \quad 1 \text{ J} = 6.25 \times 10^{12} \text{ MeV}.$$

The origin of the trapped neutron can be considered as 1) the ambient background neutrons, the existence of which have been recognized widely in public, and 2) the neutrons breeded in the sample by chain nuclear reactions triggered by reactions of the trapped neutron with perturbing nuclei, proposed in the TNCF model

As fundamental nuclear reactions pertaining with CFP, trigger and breeding reactions explained below are taken up instead of the d - d fusion reaction presupposed at first by many researchers.

3.2 NUCLEAR REACTIONS RELEVANT WITH THE TNCF MODEL

Typical reactions relevant with TNCF model are written down as follows with supplementary explanations. It will be helpful to notice here that the reactions used in the model are ordinary ones established in nuclear physics and do not have any7 special character but those assumed in the Premises.

(1) Trigger reactions.

The trapped thermal neutron can fuse with ^{6}Li nucleus in the surface layer formed on the cathode by electrolysis of D_2O (H_2O) + LiOD (LiOH) with a large cross section $\sim 1 \times 10^3$ b (1 b = 10^{-24} cm^2) (at 300 K):

$$n + {}^6_3\text{Li} = {}^4_2\text{He}\ (2.1 \text{ MeV}) + t\ (2.7 \text{ MeV}). \quad (1)$$

Also, the abundance of the isotope ^{6}Li will be assumed as the natural one, i.e. 7.4 % except otherwise described.

A trapped thermal neutron can fuse effectively with a deuteron in volume or with ^{7}Li nucleus in the surface layer:

$$n + d = t\ (6.98 \text{ keV}) + \gamma\ (6.25 \text{ MeV}), \quad (2)$$

$$n + {}^7_3\text{Li} = {}^8_3\text{Li} = {}^8_4\text{Be} + e^- + \bar{\nu}_e + 13 \text{ MeV}, \quad (3)$$

$${}^8_4\text{Be} = 2\,{}^4_2\text{He} + 3.2 \text{ MeV}. \quad (4)$$

The first reaction for a thermal neutron has a cross section 5.5×10^{-4} b and the second reaction has 4×10^{-2} b which will be used in the calculation.

In the case of solids with protium but deuterium, the following reaction should be taken up in the analysis as the trigger reaction:

$$n + p = d\ (1.33 \text{ keV}) + \gamma\ (2.22 \text{ MeV}). \quad (5)$$

The fusion cross section of this reaction for a thermal neutron is 3.5×10^{-1} b, which is fairly large compared with that of the reaction (2) with a deuteron.

(2) Breeding reactions.

The triton with an energy $\varepsilon = 2.7$ MeV (or 6.98 keV) generated in the reaction (1) (or (2)) can pass through the crystal along the channeling axis on which is an array of occluded deuterons or can proceed a finite distance with a path length ℓ_t ($\simeq 1 \sim 10$ μm) determined by the interaction with charged particles in the crystal. In the process of penetration through a crystal, the triton can react with a deuteron on the path with a length 1 μm (Premise 9):

$$t + d = {}^4_2\mathrm{He}\ (3.5\ \mathrm{MeV}) + n\ (14.1\ \mathrm{MeV}). \quad (6)$$

The cross section of this reaction is $\sigma_{t-d} \sim 1.4 \times 10^{-1}$ b for $\varepsilon = 2.7$ MeV and 3.04×10^{-6} b for 6.98 keV.

It has been a defect in experimental researches not trying to detect higher energy neutrons up to 15 MeV expected to be generated in this reaction (6). In the analysis, we have assumed the path length of 2.7 MeV triton as $\ell_t = 1$ μm.

The neutron with 14.1 MeV generated in the reaction (6) can interact with particles (deuteron d or a nucleus ${}^A_Z\mathrm{M}$) in the crystal, especially with a deuteron elastically giving a large amount of energy to it or inelastically dissociating it. (The energetic neutron, for instance, is expressed as $n(\varepsilon)$ in the following description):

$$n\ (\varepsilon) + d = n'\ (\varepsilon') + d'\ (\varepsilon''), \quad (7)$$

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

$$n\ (\varepsilon) + {}^A_Z\mathrm{M} = {}^{A-1}_{Z}\mathrm{M} + n + n', \quad (9)$$

$$n\ (\varepsilon) + {}^A_Z\mathrm{M} = {}^{A-A'+1}_{Z-Z'}\mathrm{M}' + {}^{A'}_{Z'}\mathrm{M}''. \quad (10)$$

In these reactions, the original high energy neutron loses its energy to be thermalized, or generates another low energy neutron to be trapped in the sample (breeding processes), or generates transmuted nuclei.

The deuteron $d(\varepsilon)$ having an energy up to 12.5 MeV accelerated elastically in the scattering (7) by the neutron $n(\varepsilon)$ with 14.1 MeV can fuse with another deuteron in two modes with a fairly large cross sections of the order of 0.1 b each:

$$d + d = t\ (1.01\ \mathrm{MeV}) + p\ (3.02\ \mathrm{MeV}), \quad (11)$$

$$= {}^3_2\mathrm{He}\ (0.82\ \mathrm{MeV}) + n\ (2.45\ \mathrm{MeV}), \quad (12)$$

Branching ratios of these reactions are, as is well known, 1 : 1. Another possibility is a reaction to produce ${}^4_2$He with small relative probability 10^{-7} compared with the above two:

$$d + d = {}^4_2\mathrm{He}\ (76.0\ \mathrm{keV}) + \gamma\ (23.8\ \mathrm{MeV}). \quad (13)$$

In the case of solids with protium but deuterium, the following breeding reaction between the energetic deuteron $d(\varepsilon)$ and a proton is possible:

$$d + p = {}^3_2\mathrm{He}\ (5.35\ \mathrm{keV}) + \gamma\ (5.49\ \mathrm{MeV}). \quad (14)$$

The following reaction is also probable with the energetic deuteron $d(\varepsilon)$:

$$d + {}^3_2\mathrm{He} = {}^4_2\mathrm{He}\ (3.67\ \mathrm{keV}) + p\ (14.68\ \mathrm{MeV}). \quad (15)$$

Depending on the situation in a cold fusion system, the trapped thermal neutron can induce trigger reactions like the reactions (1) $\sim$ (5) and the generated energetic particles in them can sustain breeding chain reactions (6) $\sim$ (12), (14) and (15) producing a lot of the excess heat and/or the nuclear products.

The photons generated in the reactions (2), (5), **(13) and (14) can induce photo-disintegration of deuterons** and nuclei if they have more energy than the threshold energies of following reactions;

$$\gamma + d = p + n, \quad (16)$$

$$\gamma + {}^A_Z\mathrm{M} = {}^{A-1}_{Z}\mathrm{M} + n. \quad (17)$$

The threshold energy of the reaction (16) is 2.22 MeV. In samples with deuterons, this reaction (16) with a cross section $\sim 2.5 \times 10^{-3}$ b can work effectively as a neutron breeder.

To analyze experimental data in electrolytic systems, we have taken an abundance of ${}^6_3$Li in LiOD as the natural one 7.42 %, an average velocity of the trapped neutron $v_n = 2.2 \times 10^5$ cm/s ($kT \sim 1/40$ eV at $T = 300$ K). Then, we can determine the adjustable parameter (supposed to be the density of the trapped neutron) n_n using a relation between n_n and the number of tritium atom N_t (= number of helium atom N_{He}) generated in the surface layer in a time τ;

$$N_t = N_{He} = 0.35 n_n v_n n_{^6Li} \ell_0 S \sigma_{nLi} \tau \xi, \quad (18)$$

where S is a surface area of the cathode, ℓ is the thickness of the Li surface layer, $\sigma_{nLi} = 10^3$ b, $n_{^6Li} = 3.5 \times 10^{21}$ cm^{-3} and ξ is the instability parameter

of the trapped neutron (which we take as 1 in the surface layer).

In general, the number of events (reactions) N_{nM} in time τ between the trapped neutron and the lattice nuclei ${}^{A}_{Z}\mathrm{M}$ in a volume of a sample is given by similar relation;

$$N_{nM} = 0.35 n_n v_n n_M V \sigma_{nM} \tau \xi, \qquad (19)$$

where V is the volume of the sample, n_M is the density of the nucleus M, σ_{nM} is the cross section of the reaction and ξ is the instability parameter (which we take as 0.01 for the reaction in volume).

The number of tritium atom determined by the relation (18) is also number of events N_Q generating the excess heat of 4.8 MeV per a reaction (1);

$$N_t = N_Q \equiv Q\,(\mathrm{MeV})/4.8\,(\mathrm{MeV}). \qquad (20)$$

A relation between N_n and N_t in D/Li system is given as follows; when the $n - {}^{6}\mathrm{Li}$ reaction (1) is predominant over the reaction (2) in an electrolytic system with D_2O, a neutron with 14.1 MeV is generated by the reaction (6) giving a relation between N_n and N_t assuming half of the generated triton in (1) contribute the reaction (6),

$$N_n \sim N_t \ell_t n_d \sigma_{t-d}, \qquad (21)$$

where $\ell_t \sim 1\ \mu\mathrm{m}$, $n_d = 6.8 \times 10^{22}\ x\ \mathrm{cm}^{-3}$ (x = D/Pd) and $\sigma_{t-d} \sim 1.4 \times 10^{-1}$ b. For $x = 1$, we obtain a relation

$$N_n/N_t = 9.5 \times 10^{-7} \sim 10^{-6}, \qquad (22)$$

$$\text{or } N_t/N_n = 1.1 \times 10^{6} \sim 10^{6}. \qquad (23)$$

The analyses of experimental data sets of CFP obtained in these more than ten years by the TNCF model have shown that the riddles unsolved by the presumed d - d reaction are apparent ones and explained by the model. The riddles solved are imbalances of N_Q and N_n, N_p and N_t, N_{He3} and N_n, N_{He4} and other products, and also lack of 23.8 MeV gammas in experimental data. The controversial irreproducibility of CFP is explained by the stochastic nature of atomic processes to fulfill the optimum condition for the nuclear reactions in solids and is replaced by the qualitative reproducibility including from null to positive (maximum) results for apparently the same macroscopic conditions imposed.

The experimental data showing surface nature of the nuclear reactions relevant with CFP introduced in Section 2 have been consistently explained by the TNCF model.[2,19]

A new phase of CFP found in recent five years is the nuclear transmutation (NT) in surface regions of samples as explained in Section 2. The successful explanation of NT in addition to the explanation of other events in CFP as descried above has substantiated ability of the TNCF model to give unified and systematic understanding of CFP. Investigation of the physical basis of the TNCF model will show the close relation of CFP and electroanalytical chemistry.

3.3 PHYSICAL BASES OF THE TNCF MODEL

In the process of investigations of theoretical bases for the Premises of the TNCF model, it has become clear that thermal neutrons in a crystal play a key role in nuclear reactions in solids including hydrogen isotopes.[2,3] If a crystal with high density hydrogen isotopes in it has a surface layer of another metal (as assumed in Premise 8), thermal neutrons can be trapped in the crystal (as assumed in Premise 1) and the wave functions of the neutrons are localized at the surface region (which legitimate Premise 2).

An essential nature of the local coherence of the thermal neutron can be explained briefly as follows.

A neutron as a quantum mechanical object has duality of particle and wave; it behaves as a particle in an event where its position or its momentum is determined or as a wave where its phase or its wave length is determined. The former occurs in a nuclear reaction with a nucleus and the latter in neutron diffraction by a crystal, for instance.

If many thermal neutrons are in a crystal with an appropriate lattice structure and constants, they can be treated as waves (neutron Bloch waves) modified by the interaction with nuclei on the lattice points (lattice nuclei) correspondent of the electron Bloch wave familiar in solid state physics.[20]

The neutron Bloch wave happens to be reflected at a boundary of two crystals with different band structures, a cathode and a surface layer of an electrolyte, for instance.[3] In the reflection, there occurs local coherence[3,21] of neutron Bloch waves resulting in very large probability amplitude there if we can remain in the one-body approximation used to deduce the band structure of energy spectrum of neutrons in a crystal.

The local coherence of the neutron Bloch waves

at a crystal boundary makes it inevitable to take into our consideration the mutual strong interaction of neutrons which is working ordinarily in a nucleus making it stable if the neutron number N is not less than the proton number Z of the nucleus ($Z \leq N$). It is possible to contemplate formation of a neutron drop ($Z \ll N$) at the boundary region where is the local coherence of the neutron Bloch waves in a crystal (with sufficient number of hydrogen isotopes[5]) even if such a state has not been observed directly in any experiments in solid state or nuclear physics.

If a neutron drop is formed in the boundary region of a crystal, we can expect effects induced by the interaction of the neutron drop and lattice nuclei at the boundary region and some of the effects will be observed as events of CFP. One of such events is NT observed by Miley et al.[8] which can only be explained by a fission of nucleus in the original system after absorption of several neutrons.[3]

Another conclusion of the neutron Bloch wave is stabilization of neutrons against beta decay[2] related with the Premise 1. The interaction of a neutron with lattice nuclei in one-body approximation results in reduction of energy of the neutron which can stabilize the neutron and keep it there quasi-stably. To estimate the stability due to this mechanism, we proposed a concept "the neutron affinity" of a nucleus.[2] If a nucleus has a positive neutron affinity, then a neutron in a lattice made of the nuclei is stable against beta decay.

It is interesting to notice that almost all elements have positive value of the neutron affinity if solids composed of the elements are known to show CFP.

4. ELECTROANALYTICAL CHEMISTRY AND THE COLD FUSION PHENOMENON (CFP)

Electroanalytical chemistry treats phenomena occurring when electrically conductive probes, or electrodes, are in electrical contact with the analyte solution. Equilibrium and rate of an electrode reaction and structure of an electrode/solution boundary, which are closely related with CFP, are some of main themes of electroanalytical chemistry.

In the first experiment in CFP, electrochemistry has been used originally just to feed deuterium to the metal used as a cathode to attain high pressure of the deuterium enough for anticipated d - d fusion reaction by a false presumption. If the problem is just to feed deuterium to a metal to form a metal deuteride, the method is not confined to the electrochemical one but is also possible to use others, e.g. gas contact, gas discharge, and so on. In reality, the electrochemical method is used most frequently in experiments with positive results by an empirical rule. There should be logical reasoning of this empirical rule even if CFP occurs in other systems also.

A characteristic of the electrochemical method different from others is apparently formation of a surface layer of the electrolyte in the solution on the cathode in addition to feeding of hydrogen isotopes into it. Plating or gilding of a metal on the surface of another metal has been one of main subjects of the electroanalytical chemistry.

As is shown in Section 2 by experimental data, principal reactions of CFP occur in surface layer of cathodes in the electrolytic system (and also of solids in gas contact and gas discharge systems). If the successful theoretical analysis by the TNCF model explained in Section 3 reflects physics of nuclear reactions in solids resulting in CFP, the thermal neutron trapped in solids is a key element to realize it.

The surface layer of the cation element on the cathode formed in electrolysis is responsible to the trapping of thermal neutrons as explained in Subsection 3.3. From the investigation of thermal neutrons in solids, following facts became clear: To stabilize thermal neutrons in a solid composed of an element, the neutron affinity of the element should be positive, to trap thermal neutrons in a solid A surrounded by another B, the solid B should have a sufficient thickness enough to prevent tunneling of the neutron everywhere, the structures of neutron bands in A and B should be such that the lowest allowed band in A corresponds energetically to a forbidden band in B, there should be enough hydrogen isotopes around the boundary region between two crystals, and so on.

Thus, cold fusion phenomenon (CFP) is governed sensitively by quality of the surface layer to trap thermal neutrons if they are in environment of the experimental system and results in gigantic effects if an appropriate condition is fulfilled incomparable with atomic effects. Energetically, nuclear reactions realized when necessary conditions are fulfilled produce effects almost 10^6 times larger than those of atomic reactions; the former exaggerate the quality of the electrolytic condition by a factor of about 10^6 to the

latter.

Fulfillment of the necessary condition is a subtle problem in a system depending on various factors related with the cathode, electrolyte, electrolysis conditions etc. governed also by stochastic motion of atoms on and in the cathode and in the electrolytic solution. This should be the real cause of the qualitative reproducibility, sometimes called irreproducibility inappropriately, one of the controversial problems in CFP.

It is also noticed that the species of hydrogen isotopes, i.e. D or H, are not directly related with occurrence of CFP and therefore with the trapping of thermal neutrons from our viewpoint. Deuterium is not the necessary element in CFP. This is the reason why CFP is not confined in the deuterium system but occurs also in the protium system as shown by experimental facts irrespective of expectation of some researchers.

It should be emphasized here that there are preference for combination of a cathode metal (Pd, Ni, Ti, Pt, Au, etc.), an electrolyte (Li, Na, K, or Rb) and a solvent (D_2O or H_2O) to induce CFP. From the present knowledge of experimental data sets with positive results in electrolytic systems, it is possible to conclude that following combinations of a cathode metal; solvent; and electrolyte are preferable for CFP.

Pd, Ti; D_2O; Li,

Ni; H_2O; K, Na, Rb.

The structure, i.e. thickness, homogeneity, microstructures and so on, of the surface layer depends on the condition of electrolysis. It will be an important theme of electrochemistry to study the deposition mechanism of electrolyte on the cathode and microscopic nature of surface layers plated on the cathode by electrolysis in the system used in CF experiments and clarify meaning of the seemingly preferable combination of the cathode, solvent and electrolyte illustrated above. It is also expected finally to find out the necessary and the sufficient conditions to induce CFP for various systems including the above ones in presence of thermal neutrons in ambient.

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