Tritium and Helium Measurements by Bockris et al. Analyzed Using the TNCF Model

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Synopsis

Experimental data of tritium and helium production in Pd by Bockris et al. are analyzed using the TNCF model proposed by us. The parameter n_n of the model is determined as 9.4 x 10^6 cm^{-3} with the observed maximum amount of tritium production of 3.8 x 10^7 s^{-1} cm^{-2} observed in the electrolyte (and 1.9 x 10^8 s^{-1} cm^{-2} observed in gas). The value n_n determined in this analysis is in the lowest range determined hitherto in various cold fusion systems because of, perhaps, the small value of the surface-to-volume ratio S/V of the sample; $S/V = 5.3$.

1. Introduction

In the almost 10 years after its discovery[1] in 1989, the cold fusion phenomenon has extended its range of events from excess heat and neutron production to helium generation and nuclear transmutation using materials from Pd and deuterium to compounds and protium systems. There still remains, however, some researchers persisting with the deuterium-in-solid system and $d - d$ fusion reactions, and seeking ways to explain the cold fusion phenomenon, though it seems difficult to explain its appearance by ordinary reactions in a vacuum.

We have proposed a model[2-5] where thermal neutrons, which are abundant in the environment[6] play catalytic roles in the cold fusion phenomenon to consistently explain the various events, including sometimes quantitative relations. In our point of view, those events from the excess heat generation to nuclear transmutation in materials from transition metals to proton conductors should be and can be treated using the

same physics, i.e. solid state nuclear physics.

In this paper, we take up a work[7] by J.O'M. Bockris et al. which was presented at ICCF3, held in Nagoya, Japan in October 1992, to give a consistent explanation of tritium and helium generation in Pd cathodes observed there.

2. Experimental Result

From various experimental data obtained by Bockris et al.[7], we introduce only those related with tritium and helium in this paper so we can analyze them using the TNCF model.

Electrolytic experiments with Pd rod cathodes of 1.5 cm x 1.0 cmø (S= 6.28 cm^2, V = 1.18 cm^3 and S/V = 5.3) and electrolytic solution 0.1 M LiOD + D_2O were performed with a DC power supply. A liquid scintillation counter was used for tritium measurements. The helium analysis was done on the Pd samples, which had shown tritium production, by Rockwell International, Los Angels, California for ^{3}He and ^{4}He ,where the samples packed in solid CO_2 were sent.

The usual procedure for the initiation of tritium production was to charge the Pd electrode cathodically, at ~ 0.05 V (vs. RHE) after the anodic pre-treatment. In this process, there was no increase in tritium activity. The increase of applied electric potential in cathodic direction triggered the reaction and a steady increase in tritium activity was observed. The initial, almost linear, relationship between tritium activity and time, was obtained which was common for two cells, A and B, run in parallel. The maintenance of a continuous increase in tritium activity, triggered by small increases in applied potential, had not been recorded previously, though tritium activity in a burst-like manner has been observed.

The tritium production rate reached its maximum of 3.8 x 10^7 t/s·cm^2 (in electrolyte) after 327 hours of electrolysis. (This value corresponded to total tritium production rate of 2.3 x 10^8 t/s·cm^2 due to the ratio 5 of tritium in gas and electrolyte given in their Table[3]. The reaction was quenched at 406 hours, when D_2O was added. Tritium production restarted again at 471 hours of electrolysis with no potential increase. After the addition of heavy water tritium production ceased for two days. The reaction was quenched for the second time after 406 hours of electrolysis and the decrease in t activity was observed due to dilution. An incubation period of 65 hours was needed for the reaction to restart again.

The average values of total tritium production rate observed in the gas and in the electrolyte in three weeks (504 h) were 4.2 x 10^8 and 8.7 x 10^7 t/s, respectively, and therefore, total production rate was 5.1 x 10^8 t/s from the cathode.

A linear relation between over-potential and the rate of tritium production was obtained with a value of the slope

$$4.5\,(F/2{\cdot}2.203{\cdot}RT - 8.2).$$

Tritium contamination of Pd virgin material was examined with negative results.

Detection of ^{4}He in the cathodes was done by mass spectroscopy using a sample one twentieth of a cathode. The sample was cut into eight specimens and each specimen was investigated for its helium content. All

samples, with and without thermal treatment, showed positive findings of ^{4}He. The amounts of ^{4}He released by the specimens that exceeded the average amount released or desorbed during the analysis of the Rockwell control specimens. Excess of ^{4}He was observed in 9 out of the 10 electrolyzed Pd samples from electrode which produced tritium. In the case of ^{4}He, there was no interference as far as the production of HD artifact was concerned.

The amount of ^{4}He observed in a specimen of a sample from a cathode had spread widely from 0.4 to 166.8 x 10^9, depending on the portion the specimen was cut out from. The total amount of helium from a sample inspected is calculated as a sum of the amounts from eight specimens and is 1.8 x 10^{11} ^{4}He atoms.

Summing up the amounts from all twenty portions of a cathode, assuming their equivalence (20 times that from a sample), we estimate the total helium in the cathode at 3.6 x 10^{11} atoms. No ^{3}He was found, however.

3. Analysis of the Data

Using our recipe in the TNCF model we analyze the data introduced above as follows.

First, we assume the existence of trapped neutrons in the sample PdD_x with a surface layer of Li metal and/or LiPd alloy of a thickness 1 μm (assumed) on its surface. The neutron is quasi-stable and fuses with another nucleus in regions where the neutrons become unstable due to a large disturbance from structural defects (e.g. reflecting surface or impurity nuclei). The rate P_f of the reaction (trigger reaction) is assumed to be given by the same relation as that in vacuum except a factor ξ;

$$P_f = 0.35 n_n v_n n_M V \sigma_{nM} \xi, \qquad (1)$$

where $0.35 n_n v_n$ is the flow density of the neutrons per unit area and time, n_M is the density of the nucleus, V is the volume where the reaction occurs, σ_{nM} is the cross section of the reaction. The factor ξ represents degree of stability of the neutron and is 1 in the region where the neutrons are unstable and is $0 \sim 0.01$ for an impure nucleus in a region where the neutrons are stable.

We consider the above reaction between the trapped neutrons and a nucleus with this probability as a trigger reaction generating energetic particles. The energetic particles generated by the trigger reaction react with particles in the lattice and cause breeding reactions written down below. The rate P_τ per unit time of a reaction between an energetic particle with an energy ε and one of stable nuclei in the solid is given by the similar formula as that in vacuum:

$$P_\tau = N_\varepsilon n_N \sigma_N l, \qquad (2)$$

where N_ε is the number of the particle with an energy ε generated in the sample per unit time, l is the path length of the energetic particle, n_N is the density of the reacting nucleus, σ_N is the cross section of the reaction. The path length l for a charged particle has been taken as 1 μm in the analysis for simplicity.

In the case of the electrolytic system used in the experiments, the relevant trigger and breeding reactions are written down as follows:

$n + {}^6_3\mathrm{Li} = {}^4_2\mathrm{He}$ (2.1 MeV) + t (2.7 MeV), (3)

$n + d = t$ (6.98 keV) + γ (6.25 MeV), (4)

$t\ (\varepsilon) + d = {}^4_2\mathrm{He}$ (3.5 MeV) + n (14.1 MeV) + e, (5)

$n\ (\varepsilon) + d = n\ (\varepsilon') + d\ (\varepsilon'')$, (6)

$d\ (\varepsilon) + d = {}^3_2\mathrm{He}$ (0.82 MeV) + n (2.45 MeV) + ε. (7)

Cross sections σ of the trigger reactions (3) and (4) are given as follows: $\sigma_{n\text{-}Li}$ = 9.4 x 10^2 b, $\sigma_{n\text{-}d}$ = 5.5 x 10^{-4} b, respectively. For the breeding reactions (5) ~ (7) the cross sections are $\sigma_{t\text{-}d}$ = 1.42 x 10^{-1} b (ε_t = 2.7 MeV), $\sigma_{t\text{-}d}$ = 3.0 x 10^{-1} b (ε_t = 6.98 keV), $\sigma_{d\text{-}d}$ = 8.86 x 10^{-3} b (ε_d = 12.5 MeV) and $\sigma_{n\text{-}d}$ = 5.5 x 10^{-1} b (ε_n = 14.1 MeV), respectively. The energy of the deuteron 12.5 MeV, used in the calculation of the reaction (7) for simplicity, is the maximum value obtained in the elastic collision (6).

Using the relation (3), which is the dominant reaction generating tritium and ^{4}He in the Pd + LiOD system in our regime, we can determine the parameter n_n from the maximum production rate of tritium 3.8 x 10^7 t/s·cm^2 in electrolyte (and 1.9 x 10^8 t/s·cm^2 in gas) as follows;

$$n_n)_{max} = 9.4 \times 10^6\ \mathrm{cm}^{-3}.$$

This value is in the minimum range of values determined for samples various cold fusion events were observed[4,5]. For an average value of tritium production 8.0 x 10^7/s·cm^2 calculated for 3 weeks (504 h), we obtain an average value of n_n of

$$n_n)_{av} = 3.2 \times 10^6\ \mathrm{cm}^{-3}.$$

Using the maximum amount (1.66 x 10^{11} ^{4}He per cathode) of the observed ^{4}He, we can determine n_n which according to our model should coincide with that determined by tritium generation. The result is:

$$n_n)_{He} = 1.3 \times 10^4\ \mathrm{cm}^{-3}.$$

4. Discussion

The experimental data by Bockris et al.[7] was analyzed in the preceding section and have given the above three values of the parameter n_n using the TNCF model; $n_n)_{max}$ = 9.4 x 10^6, $n_n)_{av}$ = 3.2 x 10^6 and $n_n)_{He}$ = 1.3 x 10^4 cm^{-3}.

This difference in the determined values for the parameter n_n might be considered at first sight showing a failure of the model. This is, however, not the case, considering the facts revealed after ICCF3, where this experimental result was presented. One of typical examples where much ^{4}He was observed is the data by Miles et al.[8], where the helium was detected in the gas from the electrolytic system. Another precision experiment where ^{4}He was detected in the sample was done soon after the first experiment1 by Morrey et al.[9] to check the reality of the phenomenon. This gave an unexpected result as far as the conventional prediction based on the $d - d$ direct reactions in a PdD system. The latter result resulted in a disbelief by skeptics in the reality of the cold fusion phenomenon, and in an effort by many scientists to directly prove the occurence of the $d - d$ reaction in PdD system.

This imbalance of tritium and helium generation had been one of the main riddles between events in the cold fusion phenomenon until our paper[10,11] explained them. The explanation is based on the position in materials where reaction (3) occurs and on the

residence of tritium and helium in the system. Reaction (3) occurs in the surface layer on the cathode using the TNCF model and the generated products t and ^4He go out easily into electrolyte and then into gas. This is why tritium is often observed in liquid and helium less in the cathode. The data obtained by precise measurements[9] was consistently explained assuming only 3 % of the generated helium atoms remained in the Pd cathode.

The occurrence of reactions generating NT noticed later in these several years[12,13] have undoubtedly shown the importance of the surface layer in the cold fusion phenomenon.

So, the above values determined by the average amount of tritium $n_n)_{av}$ and $n_n)_{He}$ should not be compared directly. If we assume the same factor 3% for the amount of helium in the cathode as done in the analysis of the data obtained by Morrey et al.[9], we obtain a following value;

$$n'_n)_{He} = 4.2 \times 10^5 \text{ cm}^{-3}$$

This value is compared with $n_n)_{av} = 3.2 \times 10^6 \text{ cm}^{-3}$. If we consider the ambiguous basis of the calculation 1) of the factor 3 % from analogy of Miles et al. and 2) of average tritium production rate ~ 5×10^8/s calculated from ~ 10^{15} t as a whole (Table 3[7]) in ambiguous 504 h (3 weeks), the coincidence of 3.2×10^6 and 4.2×10^5 cm^{-3} should be taken as fairly good.

The general tendency of a discrepancy between the amount of the excess heat and nuclear products by a factor of one order of magnitude noticed in our analyses[4,5,14,15] by now could be dissolved by the possible occurence of several reactions generating various nuclear products from tritium and helium to NT accompanying the excess heat where only one or two of the products was measured to compare with the excess heat as a whole.

There are many data other than those taken up in this analysis in the paper by Bockris et al.[7] which are interesting from our point of view for the development of the physics of the cold fusion phenomenon and are the theme of our future program.

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