

Research Article

On the Mechanism of Tritium Production in Electrochemical Cells

Stanislaw Szpak^{*,†} and Frank Gordon^{‡,§}

SPAWAR Center San Diego, CA, USA

Abstract

An electron capture reaction $e^- + d^+ \rightarrow 2n$ followed by deuteron to triton transmutation, the $n + d^+ \rightarrow t^+$ reaction, is judged to be the prime reaction in polarized Pd/D–D₂O system. Supporting evidence for the proposed mechanism is interpreted and discussed in terms arising from the content and meaning of chemical and nuclear reactions occurring in condensed matter.

© 2014 ISCMNS. All rights reserved. ISSN 2227-3123

Keywords: Co-deposition, Polarized Pd/D–D₂O system, Tritium production

1. Introduction

The substitution of cathodes prepared by the Pd+D co-deposition for massive palladium when applied correctly assured a 100% reproducibility in excess power generation. It was expected that they will reproducibly produce tritium as well. Indeed, this was the case. Samples of electrolyte from ten (10) cells were sent to Stanford Research Institute (Dr McKubre) and to University of Utah (Prof. Pons) for analysis. All showed marked increase in tritium concentration. In what follows, we examine the arguments leading to the proposed model of tritium production. These arguments are based on evidence resulting from the work done in the laboratory at the SPAWAR Center. The purpose of this communication is not to review the published literature, but to emphasize the connection between the event, e.g. $e^- + d^+ \rightarrow 2n$ reaction and the removal and transport of tritium from the reaction space to the environment in which they occur. To avoid misunderstanding, we define the content and meaning of the background material using terminology common in reporting results of chemical research.

2. Background Material

Fleischmann et al. [1], noted that nuclear reactions in a host lattice are affected by coherent processes and that *there are appropriate thermodynamic conditions for the formation of large clusters of hydrogen nuclei or of regions of the*

*Current address: 3498 Conrad Ave, San Diego, CA 02117, USA.

†Retired.

‡E-mail: fgordon@san.rr.com

§Retired.

lattice containing ordered arrays of hydrogen nuclei at high H/Pd ratios, resulting in the formation of clusters of deuterons dispersed in palladium lattice that would lead to the formation of ordered domains having high D/Pd ratios. These quotes augmented by, and derived from the thermodynamic structure of electrochemistry[2], guided the order in which the material is presented.

2.1. Chemical reaction

A chemical reaction is usually described by $aA + bB = cC + Q$, where $Q > 0$ denotes an exothermic and $Q < 0$ an endothermic reaction. This representation of a chemical reaction indicates that a mole of A react with b mole of B to yield c mole of C and d mole of D. Characteristic features of any equation, whether chemical or not, are its form and content. Here as written, only the initial and final states are not specified, i.e. the system consists of unbounded particles in the sense that there is a continuous range of possible energies. Thermodynamic considerations specify conditions for the reaction to occur.

2.2. Electron capture: a chemical reaction

In the Landau and Lifshitz treatise [3] one reads *It is not difficult to write down the thermodynamic conditions which govern the “chemical equilibrium” of the nuclear reaction, which may be symbolically written as $A_Z + e^- \rightarrow A_{Z-1} + \nu$ where A_Z denotes nucleus of atomic weight A and charge Z, e^- an electron and ν a neutrino. The neutrinos are not retained by matter and leave the body, i.e. this nuclear reaction may be represented by equations which correspond exactly to those employed in chemical reactions.*

2.3. Reaction space

Chemical reactions and/or processes do not occur in ideal conditions. Here, the reaction space, a three-dimensional entity, its location, structure and dynamics, and time when the reaction takes place, are discussed.

2.3.1. Location

One procedure to locate the reaction space for nuclear events is to electrolyze heavy water and record emission of particles. This can be done by placing a co-deposited Pd + D film on the CR-39 chip [4]. If the Pd + D film is co-deposited onto an open metallic substrate, e.g. a screen or a single wire and if the emission of particles occurs, then they are detected only along the electrode edge, as illustrated in Fig. 1. Evidently, the nuclear reaction is located very close to the contact surface. The bright line along the peripheries of a single eyelet is an overlap of hundreds of impingement tracks.

2.4. The interphase: its structure

The interphase is an element of the cathode structure where the concepts and definitions must be clearly presented and it is here, where the complex interplay involving kinetic and thermodynamic considerations must be considered.

2.4.1. Its structure

When the system is in equilibrium, its structure can be defined in terms of physical properties. When the system is not in equilibrium, then it is often convenient to discuss its structure in terms of occurring processes [2]. Moreover, in a

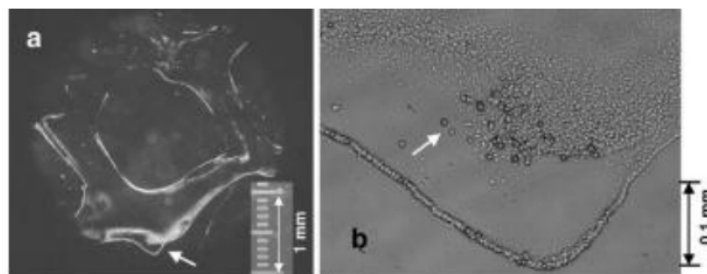


Figure 1. Emission of charged particles from a polarized Pd + D film. (a) Single eyelet, bright line consists of hundreds of impingement tracks along the edges of the screen. (b) Expanded section indicated by an arrow.

number of situations, the interphase represents a region consisting of a number of thin layers that are homogeneous and where, to assure their homogeneity, an average value of a particular variable is taken. The imposition of homogeneity on each layer results in its non-autonomous character which arises from the interaction of molecules in adjacent layers [5]. Consequently, changes in any part cause changes throughout the whole region. An abrupt change in any of the process variables will produce a relaxation spectrum with gradients associated with this variable.

2.4.2. Its dynamics

Random distribution of gas bubbles results in local changes in the charge transfer current density which, in turn, produces changes within the interphase and insures formation of localized gradients. Moreover, any motion of hydrogen within the interphase generates stresses that, in turn, produce dislocation and other types of interaction sites. How deep into the interphase they occur depends on the relaxation time of dominant processes. That it is to say, within the interphase exists a state of dynamic equilibrium which governs the distribution of hydrogen interacting with the palladium lattice which means that some interaction sites are formed, others disappear thus releasing the interacting protons and making them available for the electron capture reaction.

3. A Brief History

Shortly after the discovery of nuclear reaction in a test tube, referred to as *cold fusion*, the tendency was to explain everything including tritium production, as the result of deuteron fusion reactions. Here we present a different mechanism from that due to $d^+ + d^+$ fusion reaction (namely where the excited helium decays into t^+ and p^+) i.e. that arising from electron capture being the first step leading to tritium production which leads to a different interpretation of the observed behavior.

With the discovery of transmutation it was natural to consider a reaction path involving an electron capture by a deuteron. In September 2006, we suggested that the reaction $e^- + d^+ \rightarrow n_2 \rightarrow 2n$, plays significant role in the initiation of the F-P effect [4]^a In this paper we suggested a set of nuclear reactions, where the reactants within the reaction volume are: D^+ , and neutrons generated by the electron capture and where the reaction products are: excess enthalpy, γ -radiation and tritium, the latter being: $n + d^+ \rightarrow {}^3_1H^+ + \nu$, an analog of $e^- + p^+ \rightarrow n$.

^aIn June 2007, we submitted a manuscript "On the evidence for and origin of nuclear activities in polarized Pd/D–D₂O system" to Zeitschrift fuer physikalische Chemie. This paper was sent for review on 14 June 2007 to Mr Martin Radke who advised us to check the review status with the Editor-in-Chief, Prof. Dr Baumgaertel. Our attempts to contact Prof. Baumgaertel were not successful.

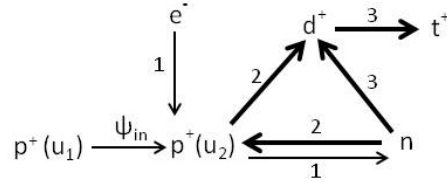


Figure 2. Flow diagram showing reactions and processes in polarized Pd/H–H₂O system exposed to an external magnetic field. ψ_m which reduces the interaction energy with $u_2 < u_1$. Arrow 1 – produces neutrons, arrow 2 – transmutes $p^+ \rightarrow d^+$, arrow 3 transmutes deuteron to triton.

3.1. Electron capture

The reluctance to accept the $e^- + d^+ \rightarrow 2n$ followed by $n + d^+ \rightarrow t^+$ reaction path for tritium production in the Pd/D–D₂O system was due to the difficulty in how to separate the electron capture reaction from other nuclear events. This difficulty could be removed if we could demonstrate the $e^- + p^+ \rightarrow n$ reaction. To test this premise, we considered an analog, viz. the polarized Pd/H–H₂O system and the $e^- + p^+ \rightarrow n$ reaction. For an electron capture reaction to occur, the proton interaction energy, $u(r)$, must satisfy the condition $\mu(n) - \mu[p^+ + u(r, \phi)] > 0$. Now, it should be possible to produce conditions that would allow the $e^- + p^+ \rightarrow n$ reaction to proceed. Indeed such conditions can be created by placing an operating cell in an external magnetostatic field (0.06T). Through the analysis for the hydrogen isotopes content in cathodes we showed the presence of all hydrogen isotopes in various ratios [6]. This can lead to only one conclusion: $e^- + p^+ \rightarrow n$ reaction did occur and started the transmutation $n + p^+ \rightarrow d^+$ and $n + d^+ \rightarrow t^+$. This set of events is illustrated in Fig. 2.

Evidently the exposure of an operating cell to an external magnetic field reduced the energy u_2 to a value so that the electron capture mechanism can start and that the forces of the proton/lattice interaction are of electronic nature [6].

4. The $n + d^+ \rightarrow t^+$ Reaction

Since all deuterons, located in the reaction volume, interact with the Pd lattice defects to a various degree, consequently their chemical potential is of the form $\mu(d^+)_l = \mu(d^+) + u(r, \phi)$. In this representation, the $u(r, \phi)$ function indicates that a part of the interacting site is incorporated into the deuteron itself, i.e. it represents the degree of overlap which,

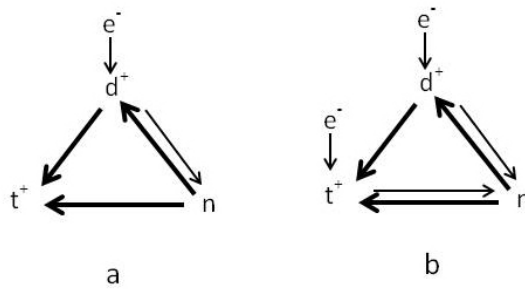


Figure 3. Reaction path generating tritium in polarized Pd/D–D₂O system. (a) Electron capture by deuteron, (b) electron capture by deuteron and triton. (3a) primary reaction, (3b) likely for for high concentration of triton is present in the reaction space.

in turn, determines whether or not the electron capture by deuteron can occur. To force the system into its nuclear active state, the quantity $\mu(n) - \mu[d^+ + u(r, \phi)]$ must be positive.

4.1. Reaction path

The path for the $n + d^+ \rightarrow t^+$ reaction is shown in Fig. 3. The starting point is the electron capture by a deuteron, $e^- + d^+ \rightarrow 2n$, a reaction that can be described by the rules of chemical kinetics. The generated neutrons interact with a deuteron to produce a triton, $n + d^+ \rightarrow t^+$, or they can react with triton to produce helium $n + t^+ \rightarrow {}^4\text{He} + e^-$. This reaction path is illustrated in Fig. 3a.

Because the value of $u(r\phi)$ function for deuteron and triton is approximately the same, it is possible that an electron can also be captured by triton yielding three neutrons, $e^- + t^+ \rightarrow 3n$. This situation, illustrated in Fig. 3b, is likely to occur when concentrations of deuterons and tritons are approximately the same. The proposed reaction path is an example of coupling of chemical processes with those governed by nuclear physics. The chemical processes take place within the confines of the interphase and change its structure which, in turn, affects their response.

5. Tritium Production and Recovery

Tracing of the tritium production and recovery of tritium in polarized Pd/D–D₂O system is illustrated in Fig. 4. It provides two sets of information. The first set shows that the interphase contains three characteristic segments, viz. reaction space and transport zone on the solid side, λ_2 and desorption of tritium from the contact surface on the solution side, λ_1 . If viewed at a given time, t_0 , then it represents the structure of the interphase by identifying the processes and assigning relevant driving forces (chemical potentials). The second set provides information on the relationship between tritium and the environment as it moves across the transport zone, crosses the contact surface and enters the λ_1 segment of the interphase.

Since the processes of production and recovery of tritium are located within the interphase, their behavior depends on and reflects the conditions arising from its dynamic equilibrium. In discussing the processes of interest are (i) the state of two variables namely electrons and deuterons while the recovery involves events occurring in the transport zone and (ii) their response to conditions imposed by the operating cell.

5.1. Tritium production

A cursory examination of Fig. 3 implies that the rate of tritium production is governed by deuteron and neutron concentrations as well as the rate constants for $n + d^+ \rightarrow t^+$ and $n + t^+ \rightarrow {}^4\text{He} + e^-$ reactions. But, the rate of neutron production depends on the rate of electron capture, i.e. on the $e_*^- + d_*^+ \rightarrow 2n$ reaction where binding energies must be specified (cf. Section 2.1). These, expressed in terms of chemical potentials, are (i) for an electron is of the form $\mu(e_*^-) = \mu(e^-) + e^-\phi$ and (ii) for deuteron of the form $\mu(df_*^+) = \mu[d^+ + u(r, \phi)]$. In addition, the electron trajectory must be such that it will collide with a deuteron.

5.2. Tritium recovery

The process of tritium recovery can be described in two ways (i) tracing its motion from the reaction space to the collection space and (ii) describing changes in the generated triton as it travels through the interphase. In the first case the generated triton, t^+ , transmutes to tritium, ${}^3\text{H}$, which enters the void, ${}^3\text{H}_v$. Since transport of tritium occurs during deuterium evolution, the state of dynamic equilibrium which governs the distribution of lattice defects/voids exists and determines the transport mode. In particular, due to localized forces associated with the dynamic equilibrium, the

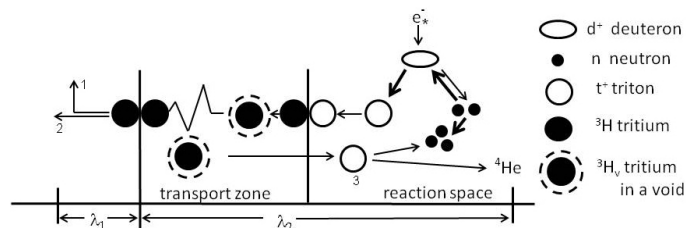


Figure 4. Schematic representation of tritium removal. Path I, line 1 to gas phase, line 2 to liquid phase. Path II, line 3 return to reaction space.

tritium transport occurs via a zig-zag path, proceeds toward the contact surface and enters the absorption layer followed by crossing the contact surface or returning to the reaction space. The empirical evidence suggests that there exists a correlation between the kinetics of deuterium evolution and the structure of the interphase which means that voids capable of retaining tritium, ^3H , are formed. This establishes the link between the kinetics of deuterium evolution and transport of tritium.

The second path is to examine possible reactions involving triton that has returned to the reaction space. The rate of tritium production is controlled by concentration of neutrons which, in turn, is controlled by the electron capture reactions ($e^- + d^+ \rightarrow 2n$ and $e^- + t^+ \rightarrow 3n$). In addition to the $n + d^+ \rightarrow t^+$ reaction, other reactions consume available neutrons. One set of possible reactions: a nucleus X having mass A transmutes to one of mass number $(A + 1)$ expressed by equation $n + {}^A X \rightarrow {}^{A+1} X$, e.g. $n + d^+ \rightarrow t^+$; $n + t^+ \rightarrow {}^4\text{He}$; etc. There are yet other possible reactions that consume neutrons.

The processes illustrated in Fig. 4. show clearly the difference between tritium which is detected and that which is generated. This difference arises from and is affected by the coherent processes occurring within the transport zone and reaction space. It is this coherence that leads to the observed sporadic production of tritium.

6. Conclusions

The proposed model and the presented empirical evidence leads to the following conclusions.

- (1) Nuclear reaction in condensed matter require establishment of a nuclear active state.
- (2) Direct evidence (cf. [6]): The electron capture by a proton starts the set of nuclear reactions via neutron production. Because the binding energy of d^+ is less than for the p^+ specie, there is no need to apply an external magnetic field for the electron capture reaction to occur because the Pd/D–D₂O system is all ready in its nuclear active state where the reaction steps are identical.
- (3) It is necessary to differentiate between what is produced and what is measured. The usually reported rate of tritium generation refers to tritium transported across the interphase region only (transport zone, cf. Fig. 4). If the term sporadic production implies that there are periods of no production, then the proposed mechanism is questionable. But this model accounts well for periods of increased tritium production.

References

- [1] M. Fleischmann, S. Pons and G. Preparata, *Il Nuovo Cimento* **107 A** (1994) 143.
- [2] P. van Rysselberghe, in *Modern Aspects of Electrochemistry*, Vol. IV, Plenum Press, 1966.
- [3] L.D. Landau and E.M. Lifshitz, *Statistical Physics*, Vol. 5, Part 1, Pergamon, Oxford, pp. 318–319.
- [4] S. Szpak, P.A. Mosier-Boss and F.E. Gordon, *Naturwissenschaften* **94** (2007) 511.

- [5] R. Defay and I. Prigogine, *Surface Tension and Absorption*, Longmans, London, 1966.
- [6] S. Szpak and J. Dea, *J. Condensed Matter Nucl. Sci.* **9** (2012) 21.