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Research Article

On the Path Leading To The Fleischmann–Pons Effect

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Abstract

Processes leading to the excess enthalpy production, the Fleischmann–Pons effect, are identified. The thermodynamic treatment [1] is extended to include self-organization. Discussion is limited to cells employing cathodes prepared by the co-deposition process.
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1. Introduction

The Pd//D_2O,LiOD//Pt electrochemical cell, operating in the deuterium evolution substance producing mode generates more energy than it consumes, the Fleischmann–Pons (F–P) effect. This excess energy is attributed to nuclear reaction(s). When employing the F–P protocol, the nuclear activity starts several days after the completion of charging. It is assumed that within this time period the polarized Pd/D–D_2O system is put into such a state that when triggered by a stimulus, which may be either internal or external to the system, initiates the nuclear activity. At first there was no interest in the examination of the incubation period, even though events that occur within this time period are critical and may lead to better understanding of the nature of the F–P effect. An extension of Fleischmann et al. [1] thermodynamic treatment by including self-organization led me to believe that a satisfactory model of the pre-nuclear active state can be constructed.

2. Background Information

A brief summary of selected experimental work is presented to provide support for the interpretation of the conclusion derived from the thermodynamic considerations. The supporting material covers (i) the Pd+D co-deposition and (ii) localized heat sources (hot spots).

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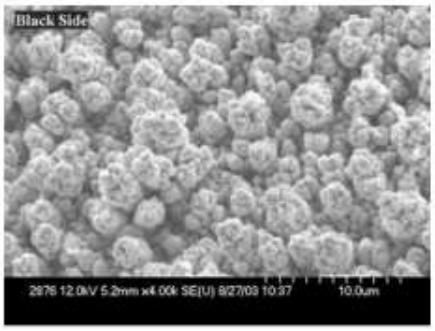

Figure 1. SEM of the Pd/D co-deposited material.

2.1. Co-deposition

One of the conditions for the occurrence of nuclear activity is the high D/Pd atomic ratio. The needed high D/Pd atomic ratio is assured if the working electrode is prepared by Pd+D co-deposition, a process in which the Pd^{2+} ions are electro-deposited in the presence of evolving deuterium. Electrodes prepared by co-deposition represent a convenient research tool because (i) they retain all features of "massive" electrodes, e.g. positive feedback and life-after-death [1], (ii) the D/Pd atomic ratios exceeding unity are obtained within seconds, (iii) the intensity of the F–P effect is higher and (v) if applied correctly then the reproducibility is 100% [2].

Figure 1 shows the structure of the Pd+D deposit employed in experimental work presented here. It is noted that the cell performance depends to a degree on the deposit morphology which, in turn, is governed by the the rates of the Pd^{2+} and D^{+}ions reduction.

2.2. Hot spots

Hot spots represent fast reactions which release thermal energy in very short time. That is to say, they resemble mini-explosions. Qualitatively, the localized heat sources lead to the following conclusions: (a) transition from non-reactive to reactive state occurs at discrete sites, (b) heat producing reactions are fast, (c) a large number of reacting particles is confined in a small volume. Quantitatively, the information derived from hot spots is as follows: (i) formation of domains having the volume of a sphere having radius of a few hundred Angstroms, in which 10^9 fast reactions occur [3], (ii) the energy released is on the order of 0.1 kJ/g and must have been occurred on at time scale of less than 10 ns involving milligrams of total mass [4].

3. Processes Leading to the F–P Effect

The content of the quote *"It should be entirely general line of attack in the study of all types of systems and processes to begin with a thermodynamic analysis and to exhaust the possibilities of thermodynamic reasoning before introducing models and assumptions of a mechanical or molecular nature."* [5] is most certainly applicable to the study of the nature of the F–P effect. It was this advice that led to the extension of the Fleischmann et al. [1] treatment by adding the concept of self-organization and including processes and/or reactions that are necessary in arriving at the identity of the pre-nuclear active state.

3.1. Self-organization

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 lattice containing ordered arrays of hydrogen nuclei at high H/Pd ratios*, resulting in the ... *formation of clusters of deuterons dispersed in the palladium lattice that would lead to the formation of ordered domains having high D/Pd ratios*. The appropriate conditions can be expressed in terms of self-organization which means that that there exists a volume of element within the system having dimensions much larger than the characteristic molecular dimensions but smaller than the total volume of the system. Within this volume fluctuations behave coherently thus modifying its microscopic behavior. At far from equilibrium, new structures, involving coherent behavior are formed and can be maintained only through a sufficient flow of energy [6].

An operating Pd/D//D_2O LiCl//Pt represents a system that satisfies the conditions necessary for self-organization, namely (i) system not in equilibrium, (ii) energy flow is assured, (iii) volume element is identified (hot spot). It is the volume element, located in the reaction space, where coherent processes take place and where the number of participating species can be estimated.

3.2. Charge transfer kinetics

Regardless of the cathode structure, (solid or Pd+D co-deposited), when the Pd//D_2O, LiCl//Pt cell operates in its substance producing mode, the cathode charge transfer reaction is

$$D_2\,O + e^- \rightarrow D_{ads} + OD^- \tag{1}$$

followed by deuterium absorption

$$D_{ads} \rightarrow D_{lattice}. \tag{2}$$

As written, Eq. (1) represents unbounded particles. But in the real world, particles interact with environment which, in turn, leads to identification of a number of statements that are relevant to the understanding the F–P effect. Using thermodynamic arguments, Fleischmann [7] concluded that:

(i) The expression $\Delta(\Phi_m - \Phi_s)F$ is responsible for (a) compression of D^+ into the lattice, (b) formation of clusters of D^+ which will be initiated at special sites, e.g. possibly at octahedral sites which would distort so that these sites would then be described as as parts of dislocation loops.
(ii) $D^+_{lattice}$ is viewed as an high density, low ion temperature plasma existing in an high electron concentration.
(iii) Electron density in the clusters is highly asymmetric.

3.3. Effect of D_2 evolution–production of defects

On smooth surfaces, in the absence of gas evolution, the charge transfer current density is uniformly distributed throughout the electrode surface. The evolving gas changes the cell current distribution from uniform to localized. The development of localized high current density is illustrated using Vogt's model [8] and shown in Fig. 2. In this model, at a distance from the charge transfer surface, the cell current is uni-directional. Close to the surface there is a stagnant layer of gas bubbles of various sizes and "empty" spaces arising from coalescence and growth of gas bubbles. This, in turn, changes the uni-directional current to the multi-dimensional high current density at the "empty" sites and causes the development of stresses which generate vacancies and other lattice defects.

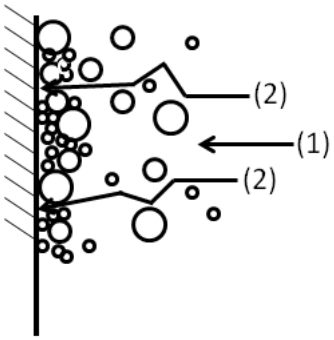

Figure 2. Effect of gas evolution on cell current distribution.

Dislocation loops and other lattice defects, e.g. vacancies, are generated by the action of the evolving deuterium. If dislocation loops can be created at will, then it would be possible to control the cell output.

4. The Path to the Pre-nuclear Active State, the π State

The path leading to the pre-nuclear active state, the π state, involves the following logically connected processes: (i) deuterium gas evolution which causes development of lattice defects (energy required 1–2 eV), (ii) lattice defects attract (draw in) deuterons and electrons (open volume effect), (iii) merging of lattice defects forms domains having high D/Pd atomic ratios (containing plasma). Incidentally, because of the porous structure of the co-deposited Pd/D deposits, cf. Fig. 1, the number of lattice defects would be larger resulting in better performance.

Reiterating, the π state, arises from coherent processes occurring during the incubation period which, for the electrodes prepared by the co-deposition, is few seconds. No information defining the structure of the plasma, except of its asymmetric D^+/e^- ratios, is available The structureless plasma can be modified by placing the operating cell in a external magnetic field.

5. Cell in an External Magnetic Field

Placement of an operating cell in an external magnetic field results in (a) change of the structure of the Pd/D deposit. Figure 3 shows the shape change of the co-deposited film. Evidently, when placed in an external magnetic field, the globules (cf. Fig. 1) are replaced by "pancake-like" structures.

The EDX analysis of selected flattened globules, Fig. 3a, shows the transmutation to new elements, namely Fe, Cr and Al. It is noteworthy that the same new elements were found in two separate runs, Figs 3a and b. Moreover, as illustrated in Fig. 3b, the EDX analysis of two closely placed flattened globules shows that they are identical.

The act of transmutation provides some insight into the nature and composition of the π state. Taking as the starting point the sequence of events which symbolically can be written

$$(!) + \pi - \pi^* \rightarrow \alpha \rightarrow X, \tag{3}$$

where (!) is the triggering mode, π^* is the compressed π state, $\alpha = X^* - [\alpha^{2+}, p^+, \gamma-, X-]$ denotes the nuclear active state and X is the stable element. For the proposed transmutation path to be realistic, two conditions must be

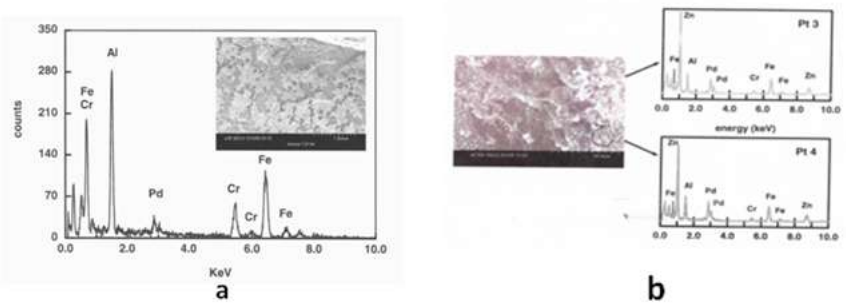

Figure 3. Selected examples of transmutation in an external magnetic field.

met: (i) the density of the molecular structure must be close or equal to the density of a nuclear substance and (ii) the dense state, the $\pi - \pi^*$ transition, must be created instantly, e.g. by either molecular collapse or a fast chain reaction.

6. Concluding Remarks

The formation of the π^* state is due to a complex interaction of kinetic and thermodynamic quantities. The formation of new structures is always the result of an instability which may be due to either internal or external fluctuations to the system, These fluctuations are always followed by the response which may bring the system to its original conditions or may produce a new structure.

The path leading to the formation of the π^* state is based on thermodynamic reasoning and supported by empirical data. Transmutation to three stable elements seem to indicate that in the presence of magnetic field, definite structure may be formed within the compressed plasma.

The nature of the "ignition" of the nuclear event is still unclear. The short-lived, randomly distributed in time and space hot spots favor the internal fluctuations as the source of process(es) leading to the ignition. This observation was taken as the starting point in the mechanism proposed by Dr F. Gordon, namely that the Pd lattice, distorted by dense plasma, responds non-linearly to internal fluctuations thus leading to localized energy spikes [9].

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