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Evidence of nuclear reactions in the Pd lattice

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Abstract An operating Pd//D₂O, Li⁺, Cl⁻//Pt cell, placed in an external electrostatic field, yielded unexpected results, viz. (i) Morphological changes in the form of discrete sites exhibiting molten-like features, i.e. features that require substantial energy expenditure. (ii) Presence of elements (Al, Mg, Ca, Si, Zn, . . .) that could not be extracted from cell components and deposited on discrete sites. The cell design and the experimental protocol assuring reproducibility is described in detail.

Introduction

Low temperature nuclear reactions were first reported nearly a century ago. In a brief communication, Oliphant et al. (1934) disclosed that by bombardment of perdeutero inorganic compounds, e.g. (ND₄)₂SO₄, by deuterons produced tritium and hydrogen. One year later, commenting on the Oliphant et al. communication, Dee (1935) stated: “. . . this no doubt being the result of transmutations effected by slower diplons which have lost energy by collision in the target.” This view was not challenged for the next 50 years. The situation has changed dramatically with the Fleischmann and Pons announcement (23 March 1989) that an operating cell Pd//D₂O, LiOD//Pt produced excess enthalpy in the amount that could not be accounted by any known chemical or electrochemical process.

The presence of light elements (e.g. He, T) was expected in view of the suggested D + D reaction path. The lack of direct correspondence between the amount of nuclear ash and the excess power suggested that other, as yet, unidentified nuclear process(es) could be responsible. To test this suggestion, a search for heavier elements was initiated. The first attempt was by Rolison and O’Grady (1991) who through surface analysis, done after a prolonged elec-

trolysis of Pd electrodes, showed the surface to contain a number of heavy elements. The search for the low temperature transmutations continued. A review of this effort was provided, among others, by Miley and Patterson (1996), Bockris and Mallove (1999) and Miley and Shrotra (2003). In most cases, the experimental methodology was different from that associated with the Fleischmann and Pons experimental protocol. The closest, in approach, is the work of Klopfenstein and Dash (1998) who found a number of new elements when heavy water was electrolysed on Ti electrodes. However, their work differed from the finding of this communication in that here: (i) co-deposited Pd/D electrodes were used and (ii) the cell was placed in an external electrostatic field.

Materials and methods

Cell construction

An electrochemical cell, Fig. 1a, consisted of (i) a rectangular vessel (2 cm × 2 cm × 8 cm) made of clear plastic (acrylic) with copper plates attached to parallel walls; (ii) negative electrode (cathode) – an Au foil 1.5 cm in length 0.5 cm wide and a Pt wire, all anchored to a polyethylene base, as illustrated in Fig. 1b; (iii) positive electrode (anode) – a Pt screen to allow for the escape of gaseous reaction products; (iv) an electrolyte – 20 mL of the solution having composition 0.03 M PdCl₂ + 0.3 M LiCl in D₂O. The cell assembly was connected to a galvanostat, the copper plates to a regulated high voltage source. The cathode assembly is placed parallel to the electric field.

Cell operation

The experimental protocol covers three time periods: (i) preparation of the Pd electrode, (ii) “stabilization” of the system and (iii) exposure to an external field after increasing the current to put the system in far from equilibrium condition.

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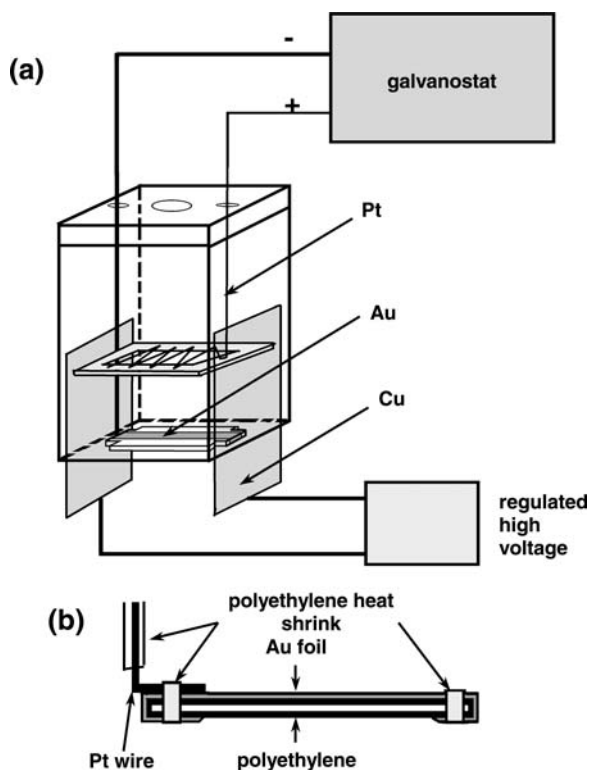


Fig. 1 (a) Electrochemical cell (b) Assembly of the Au cathode

1. The Pd/D films are prepared by the co-deposition process in which palladium and deuterium are simultaneously deposited onto substrates that do not absorb deuterium, e.g., Au, Cu, at sufficiently high-negative potentials from an electrolyte containing Pd salt dissolved in D_2O (Szpak et al. 1994). The appropriate cell current profile depends on the surface area and the composition and amount of electrolyte in the cell. For the present case, the cell current profile was as follows: 1 mA for the first 24 h followed by 3.0 mA for a period necessary to reduce all Pd^{2+} ions (i.e., when the solution becomes colorless).
2. The “stabilization” period refers to the time needed to assure uniform distribution of the D/Pd atomic ratio throughout the electrode volume. This is done by raising the cell current to a value yielding a visible D_2 gas evolution, for the present case 30–50 mA, and maintaining it for 2–3 h.
3. The activation of an external electric field is done by connecting the copper plates to the regulated high-voltage source. In the present experiment, the potential difference of 6000 V was maintained for at least 48 h. The far from equilibrium condition is realized by increasing the cell current to 100 mA or higher.
4. The Pd/D structure is examined by scanning electron microscopy (SEM) using an instrument equipped with an energy-dispersive X-ray analysis system (EDX).

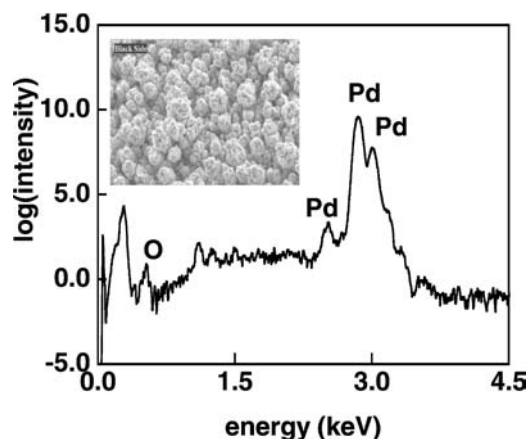


Fig. 2 The EDX of the Pd/D film deposited in the absence of an electric field. Insert: SEM of the film

Results

The structure of the co-deposited Pd/D material, after electrolysis of D_2O in the absence of an external field, including the EDX analysis of its surface, is illustrated in Fig. 2. The electrode structure consists of globules 3–7 μm in diameter arranged in short columns. Each of the individual globule is an aggregate of much smaller, almost spherical units, having a diameter in the sub-micron range. This “cauliflower-like” structure is uniform throughout the electrode. The analysis by EDX results in Pd –95.17, O –4.83 wt%, i.e. it shows only elements originally present in the cell.

If, upon the completion of the co-deposition, an external field is activated, then morphological changes occur. Among the various structural forms, we find morphologies with molten-like features, Figs. 3a–c, the development of which requires high-energy expenditure. The question that arises is: Are these structural changes due to the action of the electrostatic field alone or is additional energy required to produce the features? One such energy source is that of a nuclear origin. If the distinct features are the result of a nuclear reaction, then their chemical composition should reflect it. Indeed, compelling evidence is provided by the EDX analysis of selected spots from various runs illustrated in Figs. 3a–c. The analysis of the boulder-like form, Fig. 3a(ii), shows the presence of Al-21.72, O-71.48, Pd-6.80 wt% while the area adjacent to it Pd-95.17, O-4.83 wt%, Fig. 3a(iii). In the second example, Fig. 3b (i), the “crater-like” form, we find in the center Al-21.87, Mg-0.46, O-71.11 and Pd-6.75 wt%, Fig. 3b (ii), and in the periphery Al-21.27, Mg-4.33, O-68.19 and Pd-6.21 wt%, Fig. 3b (iii). In the third example, Fig. 3c, the results are: O-45.49, Mg-0.43, Al-0.50, Si-1.71, Cl-1.54, Ca-19.80, Zn-1.57 Pd-9.10, Au-19.86 wt%.

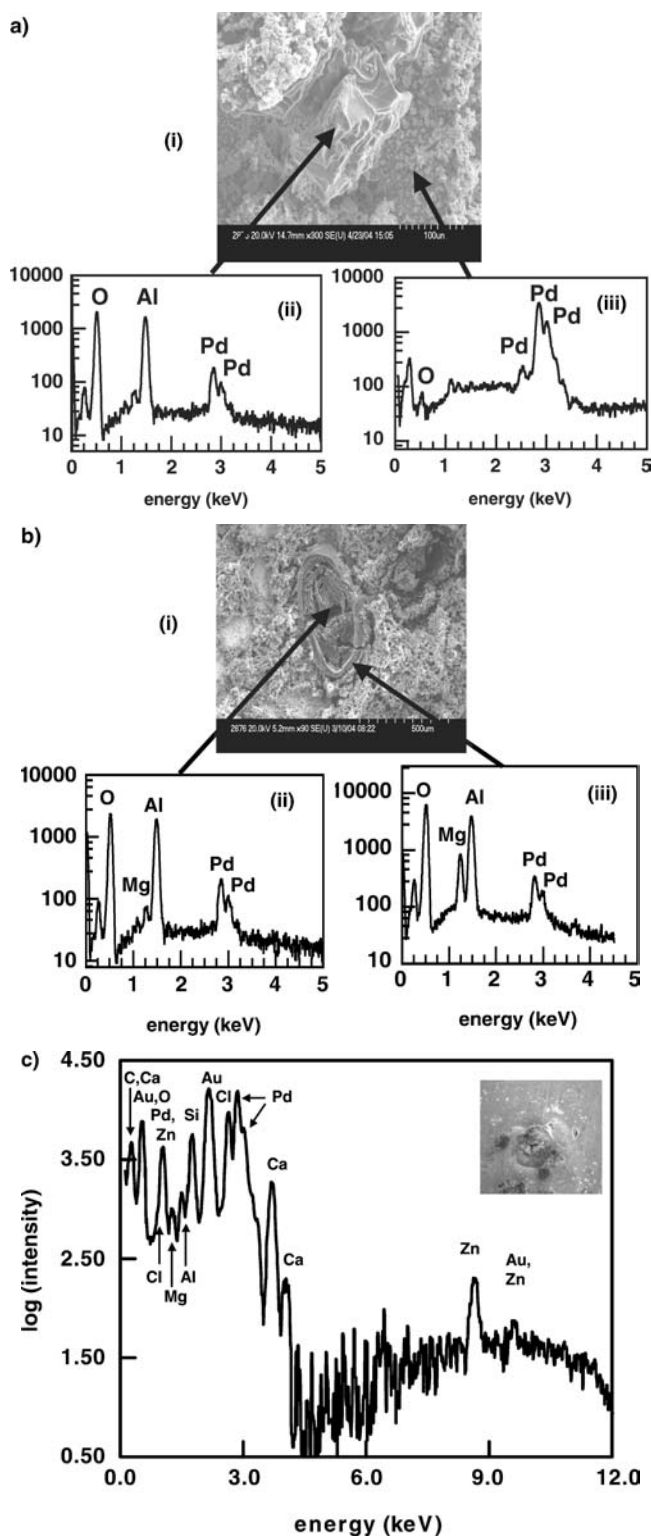


Fig. 3 (a) (i) SEM of "boulder-like" structure; (ii) EDX of "boulder-like" structure; (iii) EDX of area adjacent to it (b).(i) SEM showing a "crater-like" structure (ii) EDX of center of crater (iii) EDX of periphery of crater. (c) EDX of blister-like (insert) structure

The source of "new elements" (Al, Ca, Mg, Si, ...) could be either transmutation or contamination. Arguments against the latter are: (i) Total amount of impurities in 20 mL of electrolyte is about 0.5 mg (based on 95.5% pure reactants with Al, Ca, Mg below detectable limits by standard analytical procedures), i.e. too small to account for the observed concentrations of e.g., Al 21.27%, Fig. 3b(ii), or Ca 19.80%, Fig. 3c.(ii) Elements Al, Ca, Mg cannot be co-deposited under the experimental conditions. However, if they could be co-deposited they would be uniformly distributed throughout the electrode volume and not localized. (iii) The segregation effects associated with the surface transport processes are not likely to occur in the "cauliflower-like" structures. (iv) All cell components were analyzed for the presence of "new elements". None was found. Oxygen, chlorine cannot be associated with a nuclear reaction, rather they arise from reaction with components of the electrolyte immediately after the cessation of cell current flow, i.e. when reducing conditions are removed. The gold peaks in Fig. 3c are due to the substrate onto which the Pd/D was co-deposited. In areas in which the gold peaks were observed, the Pd/D film had separated from the underlying gold substrate.

Discussion

An operating cell is viewed as a system consisting of three subsystems: The electrolyte, the interphase and the bulk Pd/D. The region separating the bulk phases, here, the electrolyte and bulk Pd metal, constitute the interphase. In general, the interphase is an assembly of non-autonomous layers (segments) (Defay et al. 1996). In practice, different representations can be employed. In one representation, the interphase includes the electrical double layer on the solution side of the contact surface and the corresponding feature on the metal side. In another representation, thermodynamic considerations dictate its structure (van Rysselberghe 1996). In the present case, the first representation was used to discuss the shape change and the second in the search for precursors.

Shape change

It is known that (i) a conductor, charged or not, placed in an electric field, cannot remain in a stable equilibrium and a "negative" pressure acts on the surface (Landau and Lifshitz 1960) and (ii) the relation between the surface forces and the bulk response is given by the Gauss' theorem which states that the forces acting on any finite volume in a body can be reduced to forces applied to the surface of that body and vice versa. Consequently, the shape change is associated with the motion due to internal forces derived from the surface forces. The shape change is determined by surface forces whereas the rate of deformation by their magnitude (Szpak et al. 2005).

A system far from equilibrium tends to promote cluster formation which, in turn, produces hot spots that can be

displayed by infra-red (IR) imaging of the electrode surface (Mosier-Boss and Szpak 1999). The presence of “hot spots” indicates that (i) there are regions of specific interactions involving absorbed deuterium and the Pd lattice and (ii) these interactions produce intense heat sources capable of melting palladium. Thus, a model for the transition from a “cauliflower-like” morphology to shapes illustrated in Figs. 3a–c is as follows: (i) a small volume of the Pd is melted by an intense localized heat source, (ii) a blob of molten metal takes on the shape determined by the distribution the surface forces (iii) solidification, due to the heat dissipation, which produces a new and permanent shape. These distinctly different morphologies are the areas where the “new elements” are found.

Precursors

In the reaction $A \rightarrow B + C$, whether chemical or nuclear, the reactant is constrained by the imposed experimental condition while the products by energy considerations. Here, the experimental protocol, by putting the system in far from equilibrium condition, assures (i) domain/cluster formation and (ii) “self-organization”, the latter occurs when the system is open and capable of exchanging parts of its energy with environment (Glandsdorff and Prigogine 1966). Both, formation of active domains and the “self-organization” is supported by experiment (hot spots). The applicable reaction path, for the present case, is: $\dots R \rightarrow A \rightarrow \sum P_i + \sum Q_i$ where dots represent reduction of D^+ and/or D_2O , adsorption, absorption, ionization and cluster formation. $\sum P_i$ refers to the reaction products – the “new elements” and $\sum Q_i$ is the energy produced/consumed. The “self-organization” refers to the production of precursor(s). Just how the external electric field interacts with the system is not quite clear. What is clear is that the interphase is involved and that, through its non-autonomous character, the events at the contact surface are transferred to sub-surface region where they promote the formation of precursors (reactant A) leading to the nuclear events.

The results presented in this communication should be regarded as preliminary since they are limited to one cell

current profile and one value for the strength of the external field. We wish to emphasize that the interpretation offered is the first attempt to explain the anomalous behavior.

As more facts are obtained then, in all probability, a clearer picture will emerge.

References

- Bockris JO'M, Mallove EF (1999) The occurrence of cold nuclear reactions widespread throughout Nature? *Infinite Energy*, 27:29–38
- Dee PI (1935) Some experiments upon Artificial Transmutation using the Cloud-track Method. *Proc Roy Soc A* 148:623–637
- Defay R, Prigogine I, Bellemans A (1966) Surface tension and adsorption. Longmans Green, London, pp 369–389
- Glandsdorff P, Prigogine I (1971) Thermodynamic theory of structure, stability and fluctuations. Wiley-Interscience, London New York Sydney Toronto, pp xxi–xxiii 290
- Klopfenstein MF, Dash J (1998) Thermal imaging during electrolysis of heavy water with a Ti cathode. *Proceedings of the Seventh International Conference on Cold Fusion (ICCF-7)*, pp 98–102
- Landau LD, Lifshitz EM (1960) *Electrodynamics of continuous media*. Pergamon Press Oxford, London New York, pp 3, 312–334
- Miley GH, Patterson JA (1996) Nuclear transmutations in thin film nickel coatings undergoing electrolysis. *J New Energy* 1:5–30
- Miley GH, Shrotra PJ (2003) On transmutations in solids. *Trans Am Nucl Soc* 88:627–635
- Mosier-Boss PA, Szpak S (1999) The Pd/ⁿH system: Transport processes and development of thermal instabilities. *Il Nuovo Cimento* 112A:577–585
- Oliphant ML, Hartreck P Lord Rutherford (1934) Transmutation effect observed with heavy hydrogen. *Nature* 133:413
- Rolison DR, O'Grady WE (1991) Observation of elemental anomalies at the surface of palladium after electrochemical loading of deuterium or hydrogen. *Anal Chem* 63:1697–1702
- van Rysselberghe P (1966) Some aspects of the thermodynamic structure of electrochemistry. In: Bockris JO'M (ed) *Modern aspects of electrochemistry* No 4. Plenum Press, New York, pp 1–46
- Szpak S, Mosier-Boss PA, Smith JJ (1994) Deuterium uptake during Pd-D co-deposition. *J Electroanal Chem* 379:121–127
- Szpak S, Mosier-Boss PA, Young C, Gordon FE (2005) The effect of an external electric field on surface morphology of co-deposited Pd/D films. *J Electroanal Chem* (in press)