Research Article

The Fleischmann–Pons Effect: Reactions and Processes

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Abstract

A selected group of experimental evidence indicates that the Pd/D–D_2O system can be put in its nuclear active state. This is done by negatively polarizing the system which (i) starts the process of self-organization, i.e. development of coherent processes involving protons/deuterons and lattice defects to produce the pre-nuclear active state and (ii) creates conditions for the electron capture by proton/deuteron reaction to occur. The low energy neutrons transform the pre-nuclear active state into an active state, i.e. display of features such as hot spots, transmutation and particle emission which, in turn, yields information on participating reactions and processes.

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1. Introduction

Shortly after the ICCF–2 meeting Fleischmann was asked by the Royal Society to give an account of the status of research in cold fusion. In his address he stated that *In the development of any area of research (and especially in one likely to arouse controversy) it is desirable to achieve first of all a qualitative demonstration of the phenomena invoked in the explanation of the observations. It is the qualitative demonstrations which are unambiguous: the quantitative analyses of the experimental results can be the subject of debate but if these quantitative analyses stand in opposition to the qualitative demonstration then these methods of analysis must be judged to be incorrect* [1]. Two of such phenomena are observed in operating cells Pd/D_2O,Li^+,OD^-/Pt employing massive Pd cathodes namely (i) excess enthalpy generation and (ii) time separating complete saturation and the onset of thermal activities, the incubation time. The first was examined in great detail [2,3]. It is the second that can be explained only through the participation of processes that put the system in its pre-nuclear active state.

Fleischmann et al. [4], noted that, in this time period, nuclear reactions in a host lattice are affected by coherent processes, and that ... *there are appropriate thermodynamic conditions for the formation of large large clusters of hydrogen nuclei or of regions of the lattice containing ordered arrays of hydrogen nuclei at high H/Pd ratios*, resulting

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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*. The formation of clusters of deuterons suggests that excess enthalpy generation is localized and can be displayed by infrared photography [5].

The thermodynamic arguments may be extended by the teachings of the non-equilibrium thermodynamics. To start, an equilibrium is defined as a state generated by the balance between operating forces. Mathematically it is expressed by a minimum of the free energy (thermodynamic interpretation) or by the equality of forward and reverse velocities (kinetic interpretation), As the departure from equilibrium is increased, the system becomes unstable and evolves to form new structures exhibiting coherent behavior [4]. The system undergoes "self-organization", the process of formation of new structures, which is complex [6]. It is quite accurately described by an interplay of kinetic and thermodynamic quantities. The interaction between them takes the form of a struggle – some are eliminated others are formed, i.e. there exists a state of dynamic equilibrium.

1.1. Scope and order of presentation

A single observation, that of the incubation time, has led to conclusions which, in turn, opened a way to explore the meaning and content of other characteristic features such as hot spots, transmutation and particle emission. Here, we limit our remarks to behavior of cells employing cathodes prepared by *an interesting new variant of the electrochemical method* [1], which eliminates the incubation time. In our interpretation we employ terminology that is common in chemical research. However, to remove ambiguity we define what is meant by certain terms (cf. Sections 2.1–2.5).

In what follows, we argue that a lot can be learned about the nuclear events when three key observations (i) hot spots, (ii) transmutations and (iii) particle emission are interpreted using the concept of self-organization and accepting that the electron capture by a proton/deuteron starts a set of nuclear reactions.

2. Background Information

A general observation: The system is not in equilibrium, consequently, methods of non-equilibrium thermodynamics are an indispensable tool to examine the nature of the F-P effect. Furthermore, the mini-explosions recorded as a function of time, yield information from which one can speculate on the kinetics of formation of domains and their stability. To avoid miss-interpretation we define (i) molecule, (ii) chemical reaction, (iii) interphase, (iv) self-organization and (v) co-deposition. To prevent misunderstanding, terminology common in chemical research is employed.

2.1. Molecule, aggregate, cluster and domain

A molecule is defined as an assembly of two or more atoms bound together to form a structure with sufficient stability to consider it as an identifiable specie [6]. As used here, an aggregate is an assembly of molecules, a cluster is an assembly of aggregates held together by forces operating in chemical systems. A domain denotes a volume containing one or more clusters interacting with lattice defects.

2.2. Chemical reaction

A chemical reaction is usually described by either (i) $aA + bB \rightarrow cC + dD + Q$ or (ii) $A + a \rightarrow B + b + Q$ where the heat of reaction $Q > 0$ denotes an exothermic and $Q < 0$ an endothermic reaction. As written, only limited information is provided, viz conservation of matter and charge, i.e. the system consists of unbounded particles in the sense that there is a continuous range of possible energies. In reality, the initial state in both cases is controlled by experimental protocol and the final state by energy considerations.
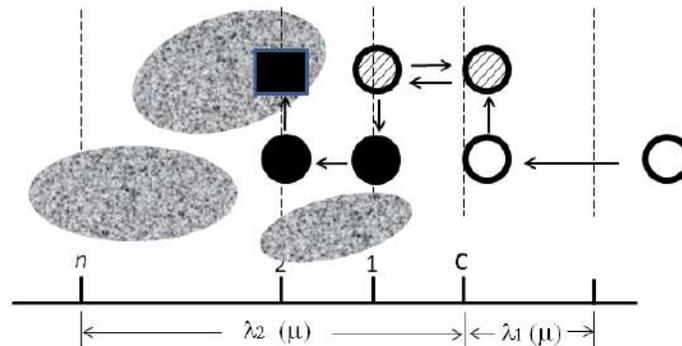
Figure 1. Structure of the interphase. c - contact surface, 1, 2, . . . , n - atomic layers, λ - thickness of the interphase, $\lambda_2(\tau)$ - thickness of the time dependent metal side layer, λ_1 thickness of the solution side layer, τ - relaxation time of the dominant process(es). open circle - H^+ ion. shaded – H in atomic state, solid - proton, square - lattice defect, shaded areas location of clusters.

2.3. Interphase

Chemical reactions rarely occur in an ideal environment, i.e. at constant temperature in time and space and at constant concentration in space. In the present case additional difficulties arise from the structural aspects of the solid phase.

These considerations lead to recognition that to uncover the true mechanism one must consider (i) conditions of an experiment, (ii) problems arising from transport of energy (heat) and matter and (iii) structural factors of the solid phase. Within the metal side of the interphase, hydrogen isotopes are distributed as follows: the sub-surface hydrogen, in the top-most layer of Pd atoms provides link between the adsorbed and dissolved atomic hydrogen, with the latter interacting with the lattice, yielding protons. With adsorption there is associated surface reconstruction but, because the surface processes are faster than bulk processes, only the hydrogen in the top-most Pd layer is responsible for its maintenance [7]. That is to say, within the interphase exists a state of dynamic equilibrium which governs the distribution of hydrogen interacting with the palladium lattice.

2.4. Self-organization

The self-organization denotes a set of processes putting the system in its nuclear active state. These processes are due to the change in the energy stored in the electric double layer, i.e. the change in its value is the driving force that generates the domains containing Pd lattice defects and deuterons. That is to say: Self-organization promotes the formation of a volume element within the system having dimensions much larger than the molecular dimensions but smaller than the volume of the system. It can occur only if the system is able to exchange part of its energy with the outside world [8].

2.5. Co-deposition

Co-deposition refers to production of electrodes by simultaneous reduction of palladium and hydrogen ions.

3. Key Features: Experimental Protocol

Characteristic features selected to identify chemical aspects of the Fleischmann–Pons effect are: (i) thermal effects, (ii) mechanical effects, (iii) transmutation and (iv) particle emission. A detailed protocol is provided only for experiments that have not been described previously.
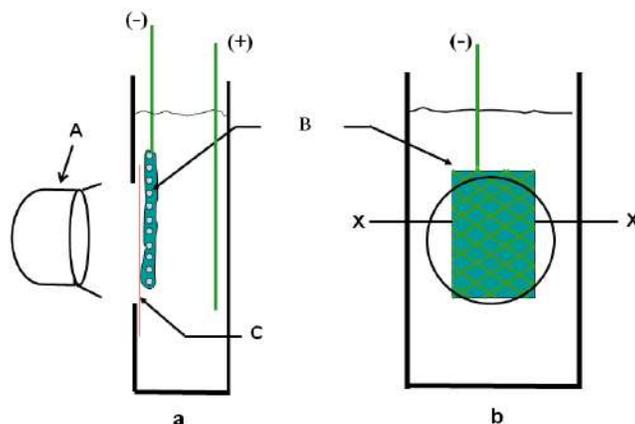
Figure 2.   Experimental set-up for display of thermal activities.

3.1. Thermal effects

3.1.1. *Experimental*

One method to examine thermal behavior of the polarized Pd/D–D$_2$O system is to view the electrode surface using an infra-red camera. Experimentally the IR camera A, Fig. 2(a), views the surface of an active negative electrode B placed next to the thin Mylar sheet C, affixed to the wall of a rectangular cell made of clear plastic. Two conditions must be met for a successful monitoring of the thermal behavior, viz. (i) the amount of the D$_2$O between the electrode surface and the IR camera must be minimal (in order to avoid the attenuation of the signal), and (ii) the electrode surface facing the IR camera must be open (in a sense that the electrode processes are accessible to viewing by the IR camera). These conditions are met by co-depositing the Pd/D film on an open substrate, e.g. on a Ni screen placed in close proximity to the thin wall (made of Mylar). The IR camera can be operated in two modes: (i) to monitor temperature distribution on the electrode surface and (ii) to measure the cell temperature across the cell (i.e. along the *X–X* line), Fig. 2(b).

Results of IR viewing the surface of an active cathode are summarized in Fig. 3.

The negatively polarized Pd/D–D$_2$O system shows the development of short lived "hot spots". These thermal activities, illustrated in Fig. 3(a) are observed early during the Pd+D co-deposition and during electrolysis. Temperature
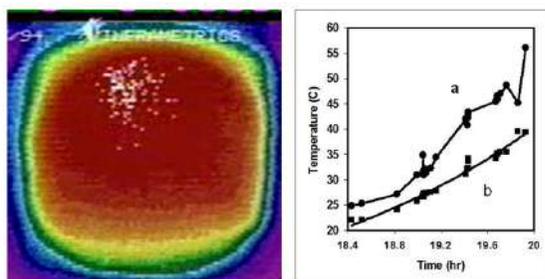

Figure 3.   Thermal activities recorded by an IR camera. (a) hot spots and (b) temperature profile.
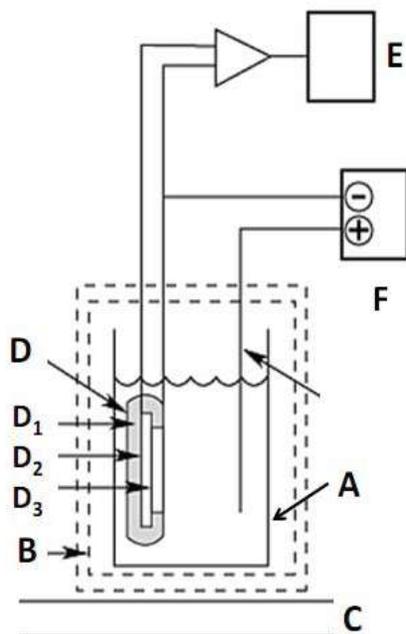
Figure 4. Recording thermal effects by an IR camera 14 Electrochemical cell designed for recording mechanical effects due to hot spots. Fig. 2b 0-0 A – electrochemical cell, B – Faraday case, C –shock absorbing pad, D – cathode assembly, D_1 – insulator, D_2 – piezoelectric disc, D_3 – Pd/D film, F – power source, E – recorder|.

profile recorded across the electrode surface when the IR camera was operated in the second mode is shown in Fig. 3(b). The cell temperature profile was taken periodically during electrolysis. The difference between surface temperature and that of solution increases with time, being initially at ca 2°C and reaching a value as high as 17°C, two hours later.

3.2. Mechanical effects

A fast exothermic reaction causes deformation of the electrode structure and the rise in temperature, thus sending pressure and temperature waves away from the source. Such waves were detected when co-deposited films were placed in contact with a piezoelectric substrate. An experimental arrangement to record their occurrence is shown in Fig. 4.

3.2.1. *Experimental*

Electrochemical cell, **A**, is placed in a Faraday cage, **B** (to prevent external noise) and the whole assembly is placed on a shock absorbing pad, **C**. The key part for the successful display of mini-explosions is the construction of the cathode, **D**. Here, a thin circular slice, in the form of a disk ($r = 1.143 \times 10^{-2}$m, $l = 2.0 \times 10^{-3}$ m of the piezoelectric material (lead–zirconium–titanate) with a conductor, e.g. (Ag) deposited on parallel surfaces was connected to an oscilloscope, **E**, and a power source, **F**, in a manner indicated. The Pd/D film was deposited onto one side a piezoelectric substrate. The characteristic feature of a piezoelectric material is the one-to-one correspondence of direct and reverse effect, e.g. compression develops a potential shift and a shift in the potential produces compression.

Figure 5.   (a) hot spots, (b) System's response to mini-explosions. a – single event, b– burst of events. (c) *Effect of solution temperature.

As a rule, a single event, Fig. 5(a), occurs rarely. A burst of events, Fig. 5(b), is the common occurrence. Here, we see clearly a single spike which, in the negative direction corresponds to the pressure pulse. Using a simple

Figure 6.   Cell design (a) and sampling procedure (b). 1 — cell with graduated wall a, 2 – recombiner. (b) – sampling sequence.

model, e.g. that of a spherical reaction space, one could, from the magnitude of the voltage spike and the Δt, reach some conclusion concerning the position and strength of the heat source which is temperature dependent. This strong temperature dependence is illustrated in Fig. 5, where spike of 30°C are measured in mV while at solution temperature 80°C, the spikes are measured in volts.

3.3. Transmutation

In support of conclusions reached by Fleischmann and Pons that the excess enthalpy is of nuclear origin, a search for other manifestation of nuclear activity was initiated. In the SPAWAR laboratory we selected the production of tritium. This was based on the belief that the governing reaction is $d^+ + d^+ \rightarrow He^*$ in which the highly excited He* atom decays into tritium and proton. Somewhat later it was demonstrated that transmutation to other elements is common [9].

3.3.1. *The $d^+ \rightarrow t^+$ transmutation*

The cell with graduated walls to provide check on the volume of electrolyte, was connected to another cell containing a catalyst with sufficiently large surface area to assure complete recombination of evolving gases, Fig. 6(a). Detailed description of sampling procedure, Fig.6(b), and analysis can be found in [10,11].

Two examples of tritium production are illustrated in Fig. 7, viz. the sporadic production, Fig. 7(a) and a massive short time production, Fig. 7(b).

3.3.2. *Transmutation – production of new elements*

The production of new elements in cells using co-deposited cathodes is insignificant. However, if these cells are placed in an external electrostatic or magneto-static field, then the rate of new elements production is markedly enhanced [12]. An external electrostatic field changes conditions at the contact surface and, only via the relaxation mechanisms, extends them into the interphase. An external magneto-static field affects not only the conditions at the interphase (via Lorentz forces) but penetrates the interphase where the gradient forces and, to a lesser degree the Lorentz forces, are
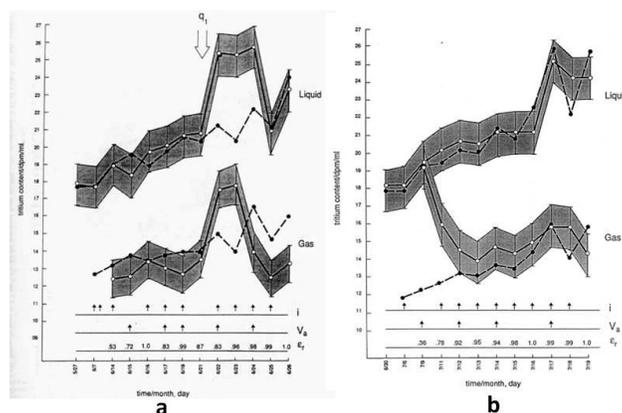

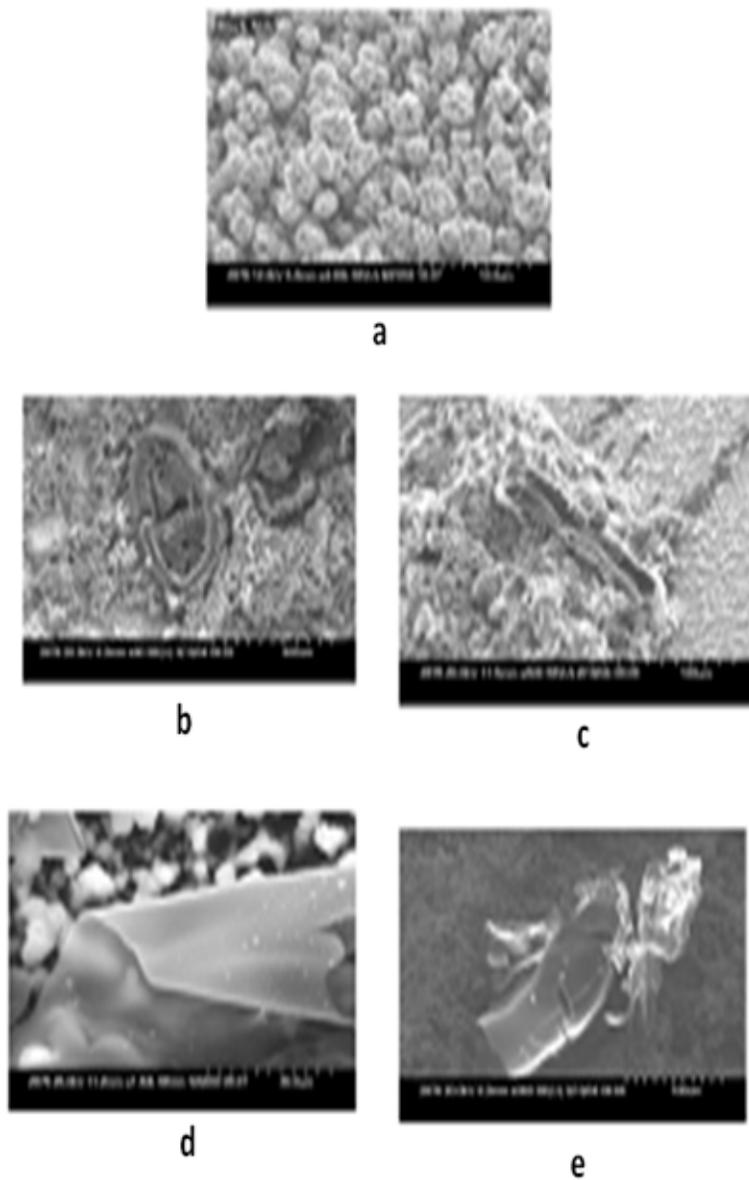**Figure 7.** Experimental and calculated rates of tritium production.

Figure 8.   The almost spherical globules, (a) were re-arranged to produce a layered structure, (b) chaotic placement of small thin fragments, (c) a bent large thin plate (foil?) with, what appears to be, branches of wires attached to its surface, (d) and a violent event resembling explosion.

active. Hence, it is not surprising that magnetic field would affect both the Pd/D structure and the reaction products in different way.
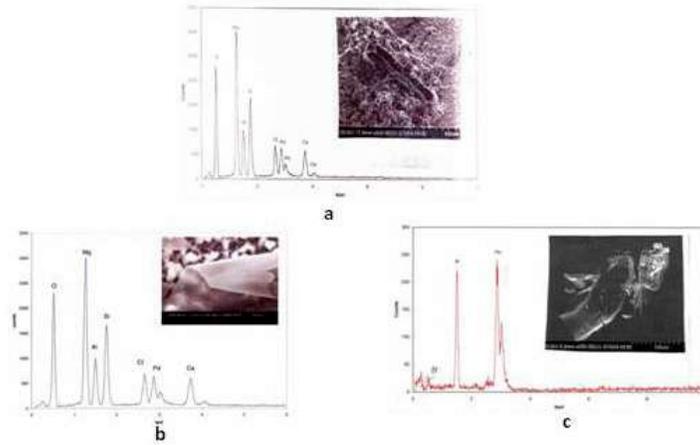
Figure 9.   Selected examples of transmutation and associated morphology in external electric field.

3.3.3. *Electrostatic field*

The set of SEM photographs, assembled in Fig. 8, shows shapes which are difficult to explain, except that their formation would require substantial energy expenditure. Even a cursory examination leads to a conclusion that the energy required to produce such changes is far in excess of that which, under the conditions of experiment, could be extracted from the external field at the required rate and intensity.

The selected examples show that the number of transmutations to new elements varies from a single element, Fig. 9(a), to multiple elements, Fig. 9(b),(c). The number of new elements does not depend on the strength of applied field. Also, there are no recognizable distinct morphological features that would indicate the number or the identity of new element(s). Note that oxygen, chlorine and Pd are not new elements since they are cell components.
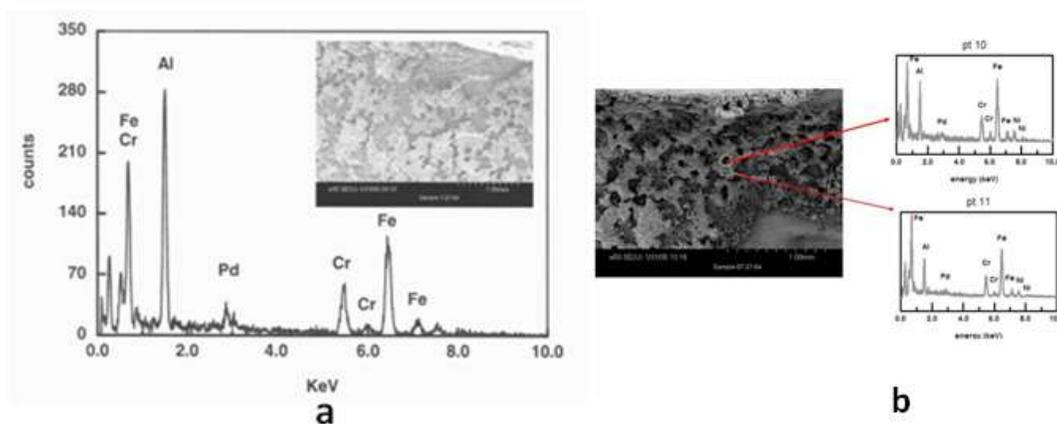

Figure 10.   Selected examples of transmutation and associated morphology in external magnetic field.

3.3.4. *Magnetic field*

The shape change of the co-deposited film when placed in an external magnetic field is somewhat different from that associated with the electric field. The distinct morphological features, e.g. boulders and craters, are replaced by "pancake-like" structures, Fig. 10(a). To emphasize the difference, we selected examples that show production of new elements (transmutation) not seen in electric field. The EDX analysis of selected sites shows production of Fe, Cr in addition to Al, Figs. 10(a), (b). Elements such as Mg, Si were found in other samples. As a rule, the number of new elements found is larger than that observed in an electric field.

3.4. Particle emission

The detection and identification of particles emitted from polarized Pd/D films, when such films were placed in an external field, yields significant information about the chemistry and physics of the nuclear processes occurring within these films.

3.4.1. *Detection of emitted particles*

If CR-39 chips are used to display the particle emission, then an appropriate procedure must be developed that would exclude misinterpretation. One such procedure involves Pd+D co-deposition on an open metallic substrate, e.g. a screen or a single wire, placed in contact with the CR-39 chip. The detection and identification of particles emitted from polarized Pd/D films, when such films were placed in an external field, yields significant information about the chemistry and physics of the nuclear processes occurring within these films. If the emission of particles from the Pd/D film occurs, then they could be detected only along the electrode edge, as illustrated in Fig. 11(a). The bright line along the peripheries of a single eyelet is, in fact, an overlap of hundreds of impingement tracks, as displayed in Fig. 11(b) which represents an expanded area indicated by an arrow. Clearly, the bright line represents a set of impingement tracks next to each other while an arrow points to an area of numerous tracks. The images in Fig. 11(c) show double and triple tracks implying that there are reactions emitting two or three particles having approximately the same mass and energy.

4. Conclusions

Conclusions that are derived from the examination of an observation are either (i) certain, (ii) probable or (iii) speculative and presented in the following order: (I) the starting point, (II) hot spots, (III) transmutation and (IV) particle emission.

4.1. The starting point

The hydrogen isotope, when in palladium lattice, exists in form of protons, deuterons or tritons interacting with the lattice defects and free electrons, i.e. electrons whose chemical potential, when electric field, is $\mu(e^-) = \mu(e^-)_{\phi=0} + e^-\phi$ [14]. When polarized and, in particular, with hydrogen evolution taking place, there is re-arrangement of absorbed deuterium and lattice defects and, if the conditions are right [15], the reaction $e^- + d^+ \rightarrow 2n$ occurs. Upon completion of the self-organization the system is in the pre-nuclear active state and transits into the nuclear active state via reactions involving neutrons. In co-deposited electrodes the self-organization is an integral part of the co-deposition process.
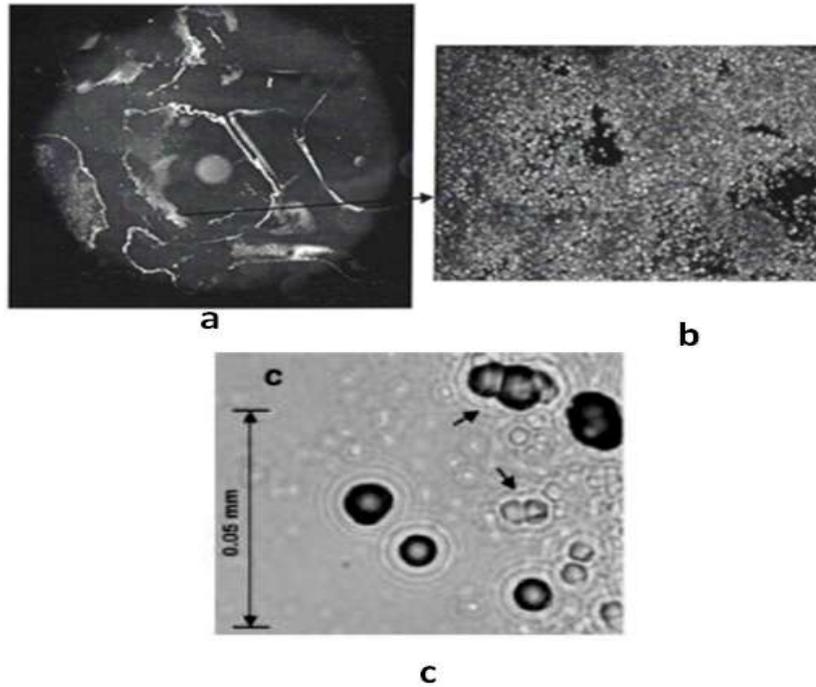
Figure 11.   Impingement tracks generated by particle emission.

4.2. Hot spots

4.2.1. *Certain*

(a) Hot spots represent a situation where a microscopically large, but macroscopically small domains absorb large quantities of deuterium in lattice defects. In order to observe hot spots, it is necessary to confine a large number of reacting particles within a small volume. Thus, there are certain locations where (i) the concentration of reactant(s) is very large, (ii) reactions are fast and (iii) they occur during early stages of co-deposition,
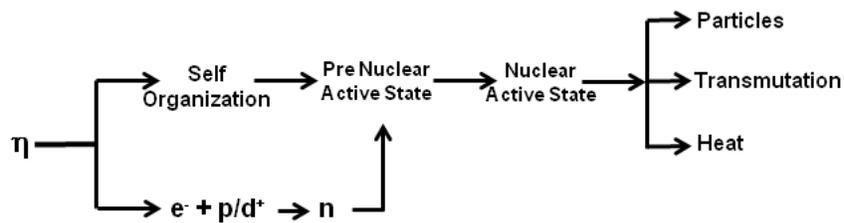

Figure 12.   Sequence of events preceding initiation of nuclear activity.

cf. Fig. 5(a), (b).

(b) One characteristic feature is an increase in the piezoelectric sensor response to change in the solution temperature. As the solution temperature of 30°C approaches its boiling point, e.g. at 80°C, there is a significant change in the sensor response, cf. Fig. 5(a), (c). The several orders of magnitude in the sensor response indicates that an increase in solution temperature produces a significant increase in the hot spot's intensity, cf. Fig. 5(c) and Section 3.1.2.

4.2.2. *Probable*

(a) *Origin.* Nuclear reactions are not affected by solution temperatures. Consequently, an increase in the reaction intensity is because (i) domains contain a significantly larger number of active species, or (ii) the rate constants of the participating processes depend strongly on temperature. The reaction mechanism(s) is (are) not known but could be either chain reactions or "collapse" of aggregates as visualized by, e.g. Adamenko [17] who postulated *... evolution of self-organized and self-supported collapse of electronic-nuclear plasma of initial solid-state density under the action of coherent electronic driver up to a state of large non-stationary electronic-molecular clusters with density close to that of nuclear substance.*

(b) *Location.* The temperature difference measured at the front side, cf. Fig. 3(b) and that at the back-side, cf. [16], indicates that the reaction yielding excess power is located close to the electrode/solution contact surface.

4.3. Speculative

(a) *Size.* After viewing the recorded hot spots data Chubb [18] concluded that $10^4 - 10^9$ is the number of single events within a sphere having $r = 100\text{Å}$).

(b) *Safety of operation.* Catastrophic thermal run-a-ways occur very seldom in cells employing either solid or co-deposited cathodes which indicates that the heat source(s) is (are) located near the surface because, if the heat sources were throughout the bulk, then the positive feed-back would cause the temperature to rise exponentially, This model is supported by the increased frequency of thermal-run-a-ways when the cell placed in an external magnetic field, cf. [15].

4.4. Transmutation

4.4.1. *Certain*

(a) *Tritium production.* Tritium production via reaction $\text{n} + \text{d}^+ \to \text{t}^+$.

(b) *Tritium production.* Intermittent production, cf. Fig. 7(a) with occasional "massive" production, cf. Fig. 7(b).

(c) *Location.* Tritium production occurs within the interphase. This conclusion is based on the following: Tritium is absent in the bulk except when Al^{3+} is added to electrolyte which changes the interphase dynamics and effectively slows the desorption process, cf. [10].

(d) *Clusters composition.* Clusters of different composition of aggregates, yielding single or multiple new elements.

4.4.2. *Probable*

(a) *"Massive" production of tritium.* On several occasions tritium was produced at much higher rates, cf Fig.7(b). One possible mechanism is shown in Fig. 13 where electron capture by a deuteron produces two neutrons of which one reacts with deuteron yielding tritium. Assuming that electron capture by triton can occur, i.e. that $\mu(\text{n}) - \frac{1}{3}\mu(\text{t}^+) + \epsilon > 0$, three additional neutrons are produced which, by reacting with deuterons yield tritons.

(b) *Transmutation path*. The multiple new elements suggests that the stable new elements are the result of a series of reactions of the type $n +^A (X) \rightarrow^{A+1} (X) - [p^+, \alpha^{2+}, n, \gamma] \rightarrow^{A_1}_{Z_|} X)$.

4.5. Particle emission

4.5.1. *Certain*

(a) *Location of nuclear reactions*. Nuclear reactions take place within the interphase region, cf. Fig. 11(a).
(b) *Type of emitted particles*. Emission of two or three particles having approximately the same mass and energy, cf. Fig. 11(c).

5. Reformulation

The reported reactions, processes, and conclusions are re-arranged to make them useful to experimenters. Evidently, (i) the bulk of activities, if not all, occur within the inhomogeneous region, the interphase, (ii) they are assembled in groups that identify their principal functions and (iii) they provide rationale for the choice of approach to discussion of the system's behavior.

5.1. Reactions/processes in Group I

Reactions within the first group are: (i) ionization of absorbed deuterium $- D \rightarrow D^+ + e^-$, (ii) production of molecule-ions $D + D^+ \rightarrow D_2^+$ [10,13], (iii) production of hybrid molecule-ion $n + D_2^+ \rightarrow D^+T^+$, (iv) self-organization leading to the formation of clusters of aggregates $Pd^* + nD^+ \rightarrow Pd^* \ldots D_n^+$, (v) production of neutrons $e^- + D^+ \rightarrow 2n$. Reactions (i)–(iv) are typical chemical reactions while (v) is treated as a chemical reaction within the context of nuclear chemistry.

Function: The system is in the state of dynamic equilibrium established through the order–disorder activity at the contact surface. Its principal activity is to provide, via self-organization, reactants that produce excess power and new elements (transmutation). These are essentially chemical processes and could be either exo- or endothermic.

Useful information: Chemical processes are affected by the structure (morphology and formation of mobile lattice defects) of an interphase which, in turn, is affected by the cell current profile and external fields; Methods of non-equilibrium thermodynamics and non-linear chemical kinetics would have to be applied to fully describe the formation of domains associated with production of localized hot spots.
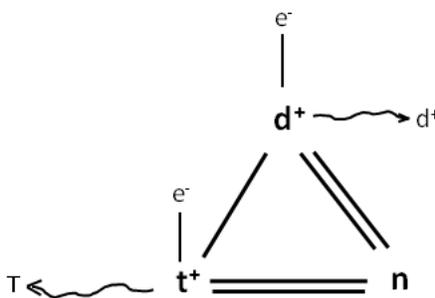

Figure 13.   Coupled reactions leading to tritium production.

5.2. Reaction/processes in Group II

Reactants entering Group II are the products of self-organization. These reactants can interact with electrons (electron capture) or with neutrons. In either case, the dynamic equilibrium of stable aggregates generated by self-organization, is disturbed and the affected aggregates can either collapse or explode, depending on the type of interaction with environment. There is (i) self-organization, (ii) dense plasma corresponding to clusters interacting with the Pd lattice defects, e.g. mobile dislocations, (iii) "of solid-state density" corresponding to the action of the $F\Delta\Phi$ and asymmetric stress field and (iv) "coherent electronic driver...," is equivalent to the formation of new structures on the "super-molecular" level.

Function: The reaction/processes in Group II provide the principal production of excess power via an unspecified set of nuclear processes, except for the electron capture by deuteron.

Useful information: There are many possible theories dealing with the nature of nuclear processes responsible for both production of excess enthalpy, transmutation, radiation and particle emission. So far as we were able to understand, none of the proposed theories meet all of the conditions stated by Chubb [18], namely: *for the model driven research, four conditions must be evaluated: (i) Is it applicable? (ii) Does it violate the second law of thermodynamics? (iii) Is it physically and mathematically complete? (iv) is it reducible to mathematical expressions that are useful to experimenters..*

5.3. Reactions/processes in Group III

While the processes in Groups I and II can be treated in terms of chemical concepts, those in Group III involve system stabilization via the transition from unstable nuclei to stable ones by various decay processes. A very general picture is as follows: The first step, in stage I, is the production of neutrons *via* the electron capture process: $e^- + D^+ \rightarrow 2n$. Stage II is the seat for nuclear reaction of the type $n + \Sigma_Z^A X_i \rightarrow \Sigma_Z^{A+1} X_j$. Here the reaction product, entering stage III, is a set of new unstable nuclei $_Z^{A+1}X_j^*$ which undergo process of "stabilization" by fusion, fission, particle emission and electromagnetic radiation: $\Sigma_Z^{A+1} X_j^* - [p^+, \alpha^{2+}, n, \gamma, X] \rightarrow \Sigma_{Z+1}^{A_1} X_j$.

Function: Production of new elements via the various decay paths leading to the formation of stable elements such as Ca, Al, Si, etc.

Useful information: Although the transition from unstable to stable nuclei is highly exothermic, its contribution to the cell power output is minimal (basis - miss-match, amount of nuclear products and excess enthalpy He + transmutation).

6. Concluding Remarks

Nearly a quarter century ago, a new phenomenon – the room temperature nuclear reaction in a test tube – was disclosed by Fleischmann and Pons. Since this effect was discovered by two professors of chemistry – one would expect that methods and reasoning of chemistry would be helpful in the interpretation and further development of the understanding of this phenomenon. But such has not been the case. The chemical aspects were replaced by topics of interest to physicists. Theories based on specific assumptions elegantly executed, were followed by a search for the predicted behavior. Here, we propose another approach, that advocated by Born [19], who wrote *"My advice to those who wish to learn the art of scientific prophesy is not to rely on abstract reason, but to decipher the secret language of Nature from Nature's documents,the facts of experience."*

In following this advise we relied on two observations, viz. hot spots and production of new elements, and drew conclusions based on chemical reasoning. The conclusions reflect our current understanding of the F–P effect. We conclude this communication with a statement that a lot can be gained by examining the chemical aspects of the polarized Pd/d–D_2O system.

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