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Research Article

Overview of Pd/D Co-deposition

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Abstract

Pd/D co-deposition has been used by a number of researchers to explore the condensed matter nuclear reactions occurring within the Pd lattice. Reported reaction products include heat, transmutation, tritium, energetic charged particles, neutrons, and gamma/X-ray emissions. An overview of these results are discussed in this communication.
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Keywords: Co-deposition, Energetic particles, Gamma/X-ray emissions, Heat, Transmutation

1. Introduction

Knowing that long incubation times were needed to achieve high D/Pd loadings in bulk Pd cathodes in order to induce heat production, Stanislaw Szpak, an electrochemist at a Navy laboratory in San Diego, began to explore using Pd/D co-deposition to initiate this effect. In this process, working and counter electrodes are immersed in a solution of palladium chloride and lithium chloride in deuterated water (Fig. 1a). Palladium is then electrochemically reduced onto the surface of the working electrode in the presence of evolving deuterium gas. SEM analysis of electrodes prepared by Pd/D co-deposition exhibit highly expanded surfaces consisting of small spherical nodules (Fig. 1b).

Cyclic voltammetry and galvanostatic pulsing experiments indicate that, by using the co-deposition technique, a high degree of deuterium loading (with an atomic ratio D/Pd>1) is obtained within seconds [1–3]. Because an ever expanding electrode surface is created, non-steady state conditions are assured, the cell geometry is simplified because there is no longer a need for a uniform current distribution on the cathode, and long charging times are eliminated. Using variations of Pd/D co-deposition, researchers have reported on observing excess heat, gamma/X-ray emissions, transmutation, as well as the production of tritium and energetic particles.

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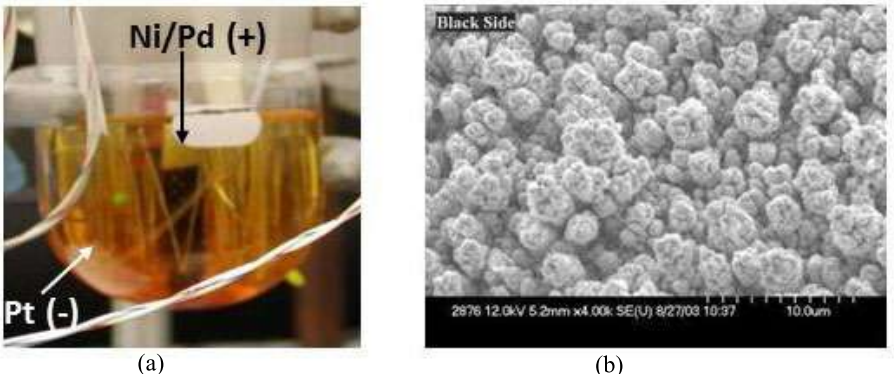

(a) (b)

Figure 1. (a) Photograph of the cell showing the cathode, anode, and plating solution. The original plating solution was $PdCl_2$, LiCl, in D_2O, (b) SEM of an Au/Pd cathode showing the cauliflower structure of the deposit.

2. Results and Discussion

2.1. Heat

Szpak et al. used thermocouples in the cathode and solution [4] and infrared imaging [5] to show that the heat source was the cathode and not Joule heating. Infrared imaging of the cathode showed that heat generation was not continuous but occurred in discrete spots on the Pd deposit. Crater formation and evidence of Pd melting under water have also been observed [5]. Depending upon their geometry and diameter, the melting temperature of Pd nanoparticles occurs between 227–1527°C [6]. Nagle [7] estimated that the energy to create a crater was 1.8×10^{-4} J, which agrees well with the value calculated by Kim [8] (6.5×10^{-4} J), who assumed that craters were formed by vaporization. Since the energy of 1 μJ is equivalent to $6.2 \times 10^5 10$ MeV energy releases, crater formation requires the nearly simultaneous occurrence of many LENR events. This is also inconsistent with the primary hot fusion reactions: D(d,p)T, D(d,n)^{3}He and subsequent D(t,n)α and D(^{3}He,p)α reactions where 2/3 of the 24 MeV released is in fast neutrons. CR-39 neutron rates, as well as results from other LENR researchers, find a dearth of neutrons.

Researchers who have seen evidence of heat production, in Pd/D co-deposition experiments, using calorimetry include Miles [9], Cravens and Letts [10], Letts and Hagelstein [11], DeChairo et al. [12], Swartz and Verner [13], and Dash [14]. Despite the fact that different calorimeters were used as well as different plating solutions, excess heat was measured by these researchers. Miles [9] used the ammonia complex of Pd and an open, isoperibolic calorimeter. He showed that the rates of excess enthalpy generation using electrodes prepared by the Pd/D co-deposition technique were comparable to that obtained when Pd bulk electrodes were used. Miles showed that shuttle reactions were not responsible for the excess heat [15]. Positive feedback [9,10] and heat-after-death effects [4] were also observed with the Pd/D co-deposited electrodes. Letts and Hagelstein [11] used the chloride complex of Pd and a closed, Seebeck calorimeter. They showed that when Pd/D co-deposition was done using low current densities, which improved adherence, no excess heat was produced. However, co-deposition using high current densities did produce excess heat. Like Letts and Hagelstein, DeChairo et al. [12] also used the chloride complex of Pd. They used an open, Seebeck calorimeter. Like Letts and Hagelstein [11], they observed no heat production when Pd/D co-deposition was done using a low current and heat production using high current densities. Swartz and Verner [13] obtained 100,000 J of excess heat doing Pd/D co-deposition in low electrical conductivity D_2O using a spiral wound palladium cathode.

This configuration resulted in asymmetric electrolysis on one side of the cathode which caused deuteron flux inside the metal lattice. They did multi-ring thermal spectroscopy with thermal controls and an open system. Dash and Ambadkar [14] used a closed, Seebeck envelope calorimeter. In their experiment, the Pt was the cathode and Pd was the anode. Pd dissolved from the Pd anode and plated out, in the presence of evolving D_2 gas, on the Pt cathode. The observed excess thermal output was 0.93 ± 0.1 W.

2.2. Tritium

Tritium content during Pd/D co-deposition was measured by researchers using the liquid scintillation technique. Bockris et al. [16] measured tritium in gas and liquid phases during electrolysis. They observed bursts of tritium production when low tritiated D_2O was used and a loss of tritium when highly tritiated D_2O was used. Loss of tritium suggests there is a reaction that consumes thermal tritium. These results have been verified by other researchers. Szpak et al. [17] did a similar experiment and also saw bursts of tritium production when low tritiated D_2O was used. Using highly tritiated D_2O, Miles [18] observed a decrease in tritium in the electrolyte upon termination of Pd/D co-deposition experiments. Lee et al. [19] used a closed system to measure changes in the tritium content in Pd/D co-deposition cells. They observed an increase in the tritium content when low tritiated D_2O was used. However, a decrease in tritium was observed when highly tritiated D_2O was used.

2.3. Gamma/X-ray emissions

Using a Si(Li) detector with a Be window, the cathodically polarized Pd/D cathode was observed to emit X-rays with a broad energy distribution and with the occasional emergence of recognizable peaks attributed to the Pd K_α and Pt L lines [20]. The emission of X-rays was sporadic and of limited duration. Fogging of photographic film was observed after exposure to Pd deposited on a Ag disk cathode upon completion of a co-deposition experiment [21]. A thin Mylar sheet separated the cathode from the film. The circular shape of the cathode could be seen on the film. Also the fogging was inhomogeneous indicating that some sites were more active than others.

Miles et al. [22] used a Geiger-Müller (GM) detector with a thin window to monitor Pd/D co-deposition cells. The co-deposition experiments showed high radiation counts within a few hours of beginning the electrolysis. In contrast, solid Pd rods always required one to two weeks before anomalous radiation was detected.

2.4. Transmutation

As discussed *vide supra*, Dash and Ambadkar's Pd/D co-deposition experiment produced an excess power output of 0.93 ± 0.1 W [14]. Upon termination of the experiment, the Pt/Pd electrode was subjected to SEM/EDS analysis. EDS analysis of several spots showed three peaks at 2.84, 2.99, and 3.18 keV that were attributed to the Pd $L\alpha_1$, $L\beta_1$, and $L\beta_2$ X-ray emissions, respectively. For pure Pd, the $L\beta_1/L\alpha_1$ ratio is 0.4. However other spots showed ratios as high as 0.75. This increase in the ratio indicates that another element is contributing to the peak at 2.99 keV. The $L\alpha_1$ peak of Ag occurs at 2.98 keV. Changes in this ratio can be used to quantitate the amount of Ag present. A ratio of 0.75 indicates that this spot contains about 19% Ag. Dash and Ambadkar attributed the formation of Ag to the following reactions:

$$^{108}\text{Pd} + \text{n} \rightarrow {}^{109}\text{Pd} \rightarrow {}^{109}\text{Ag} + \text{e}^- + \text{energy}.$$

Mosier-Boss et al. conducted a Pd/D co-deposition experiment on a Au foil in the presence of an external magnetic field [23]. Upon termination of the experiment. The Au/Pd cathode was subjected to SEM/EDS analysis. In areas

where the deposit exhibited a cauliflower like morphology consisting of aggregates of spherical micro-globules, EDS analysis showed X-ray emissions from Pd. In some areas, the deposit looked sintered. EDS spectra of these areas showed large peaks due to Fe, Cr, Ni, and Al with a very small peak due to Pd. ICP-AES, and ICP-MS analysis of the electrolyte has shown that the presence of these new elements are not due to contamination [12] as the concentration of these new elements on the cathode were several orders of magnitude larger than was found in the electrolyte. The same was true of other cell components. The inhomogeneous distribution of these new elements also supports the notion that the presence of these new elements is not due to contamination. The small size of the Pd X-ray peaks indicates that Pd has been consumed. Since Fe, Cr, and Ni are approximately half the atomic mass of Pd suggests that Pd has been fissioned. The observation of long range alphas ($E_\alpha \geq 12$ MeV) in other Pd/D experiments support fissioning of Pd as these alphas can only form as the result of ternary fission [24–26]. These magnetic field effects were verified by DeChiaro et al. [12], who further showed that the excess heat correlated with the occurrence of these new elements.

3. Energetic Particles

Mosier-Boss et al. [27] used CR-39, a solid state nuclear track detector, to measure the emissions of energetic particles in Pd/D co-deposition experiments. It was found that what substrate was used impacted what was observed. When Pd/D co-deposition was done on a Ni screen cathode, in the absence of an external electric/magnetic (E/B) field, no tracks due to energetic particles were measured. Instead the impression of the Ni screen was observed on the CR-39 detector. With time, the area where the Ni screen was in contact with the detector was observed to swell. Both the swelling and the impression of the screen were observed when a detector, covered with a metal screen, was exposed to a Cs-137 γ source. Consequently, the damage observed for the Ni screen cathode in the absence of a field was attributed to γ/X-rays. When either an external *E* (with a 6% AC ripple) or B field was applied in a co-deposition experiment on a Ni screen cathode, tracks were observed in the CR-39 detector. When the cathode substrate was either Ag, Au, or Pt wires, tracks were observed in the detector in both the presence and absence of an external E/B field. A series of control experiments demonstrated that the tracks observed in the CR-39 detectors as a result of Pd/D co-deposition were not due to either chemical or mechanical damage or to radioactive contamination. In addition to tracks due to energetic particles, Mosier-Boss et al. [28,29] also reported seeing triple tracks in CR-39 detectors. These triple tracks are diagnostic of the ^{12}C(n,n')3α carbon breakup reaction due to the reaction of a $\geq$ 9.6 MeV neutron with a carbon atom in the detector and were shown to be indistinguishable from DT neutron generated triple tracks. No triple tracks were observed in CR-39 detectors used in control experiments.

The CR-39 results have been verified by Tanzella et al. [30], NASA Glenn [31], and three groups of UCSD chemical engineering students [32,33]. Tanzella et al. [30] conducted experiments in which the polyethylene covered CR-39 detectors, in contact with the cathode, were immersed in the electrolyte. The polyethylene film covering the detectors was 60 μm thick. Upon termination of the experiment, these detectors underwent microscopic examination, scanning using a TASL (Track Analysis System Ltd.) system, linear energy transfer (LET) analysis of the scanned data, and sequential etching [30,34]. The LET analysis determined that the tracks on the front surface in contact with the cathode were caused by $\geq$ 1.8 MeV protons, $\geq$1.8 MeV alphas, and secondary particles due to recoils from either energetic protons and/or neutrons [34]. On the obverse surface of the detectors, the particles that created the tracks were identified as $\geq$11.8 MeV protons and/or recoils from either energetic protons and/or neutrons. Destructive sequential analysis of the detectors [30] identified tracks due to 3 MeV protons, 12 MeV alphas, and 16 MeV alphas – which was in agreement with the LET spectrum analysis. This analysis also showed 2.2–2.5 MeV neutrons. It was estimated that these neutrons were produced at a rate of 0.3 $\pm$ 0.1 n/s. Tanzella et al. [30] conducted additional experiments in which the CR-39 detector was placed outside the cell. In these experiments, a 6 μm thick Mylar film separated the detector from the cathode. These experiments were conducted in both H_2O and D_2O. The detectors were subjected to destructive sequential etching analysis. No neutrons were detected in the H_2O experiment. However, the

D_2O experiment showed recoil tracks due to 2.2–2.5 MeV neutrons. In this experiment, the rate of neutron generation was 0.6 ± 0.1 n/s.

4. Conclusions

The Pd/D co-deposition experiment has offered great flexibility in experimental design. Different Pd plating solutions have been used as well as different cell configurations (e.g., parallel electrodes or concentric electrodes) working electrode surfaces (Au, Ag, Ni, Cu, or Pt), and electrode geometries (wire, sheet, or screen). Both closed and open system have been used. The Pd/D co-deposition process has been used by several researchers to investigate the phenomenon of condensed mater nuclear reactions within the Pd lattice. Reported reaction products include excess heat, gamma/X-ray emissions, transmutation, as well as the production of tritium and energetic particles. These products indicate that several varieties of nuclear reactions are occurring in the system. These include primary and secondary fusion reactions to produce neutrons, protons, tritium, and ≥ 10 MeV protons and neutrons. There is evidence of transmutation as shown by the production of Ag that can arise from either proton (≥ 10 MeV) or neutron capture by Pd. The observation of long range alpha particles indicate the occurrence of possible ternary and quaternary fission of Pd that is supported by the presence of such elements as Fe, Cr, Ni, and Al with a corresponding decrease in Pd.

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