

Condensed matter nuclear reaction products observed in Pd/D co-deposition experiments

P. A. Mosier-Boss^{1,*}, L. P. Forsley², F. E. Gordon³, D. Letts⁴, D. Cravens⁵, M. H. Miles⁶, M. Swartz⁷, J. Dash⁸, F. Tanzella⁹, P. Hagelstein¹⁰, M. McKubre⁹ and J. Bao⁹

¹9112 Fermi Ave., San Diego, CA, USA

²JWK Corp., 5101B Backlick Road, Annandale, VA 22003, USA

³SPAWAR Systems Center Pacific, San Diego CA, USA

⁴12015 Ladrado Lane, Austin, TX 78727, USA

⁵Cloudcroft, NM 88317, USA

⁶University of La Verne, La Verne, CA 91750, USA

⁷JET Energy, Inc., Wellesley, MA 02481, USA

⁸Portland State University, Portland, OR 97207, USA

⁹SRI International, Menlo Park, CA 94025, USA

¹⁰Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, MA, USA

Pd/D co-deposition has been used by a number of researchers to explore the condensed matter nuclear reactions occurring within the palladium lattice by generating highly loaded layers of lattice over the cathode. Reaction products that have been observed include heat, transmutation, tritium, energetic charged particles and neutrons. The results of these experiments are discussed here.

Keywords: Energetic particles, excess heat, Pd/D codeposition, transmutation.

Introduction

In the typical Pd/D co-deposition experiment, the cathode and anode are immersed in a solution of palladium chloride and lithium chloride in deuterated water. The cathode is comprised of either palladium or a metal that does not absorb hydrogen isotopes. The difference between these cathodes is shown in [Figure S1 \(see Supplementary Information online\)](#). Palladium is then electrochemically reduced onto the surface of the cathode in the presence of evolving deuterium gas. In the co-deposition process, an ever-expanding electrode surface is created that assures the existence of non-steady-state conditions and local high loading (D/Pd ratio ≥ 1), as shown by cyclic voltammetry^{1,2} and galvanostatic pulsing³ experiments. More importantly, Pd/D co-deposition eliminates long charging times and prevents lattice cracking. The resultant Pd deposit has a uniform ‘cauliflower’-like structure consisting of aggregates of spherical micro-globules and has a large surface area. In this article, results of Pd/D co-deposition experiments conducted by several researchers are discussed.

*For correspondence. (e-mail: pboss@san.rr.com)

Results and discussion

One of the earliest Pd/D co-deposition experiments⁴ involved measuring the temperature of the cathode and the solution during the course of the reaction. It was found that the temperature of the cathode was 2–4°C hotter than the solution indicating that the heat source was the cathode and not Joule heating. This was further confirmed by real-time infrared imaging of the cathode⁵. Not only did the infrared imaging show that the cathode was hotter than the solution, the steepness of the temperature gradients indicated that the heat sources were located in close proximity to the electrode–solution contact surface.

Miles was the first to report on calorimetric measurements of Pd/D co-deposition⁶. In addition to palladium chloride, Miles’ electrolyte also contained ND_4Cl and ND_4OD , but no $LiCl$. He used an open, isoperibolic, Dewar calorimetry cell that was tall enough to keep the Pd deposit from reaching the gas–liquid interface, thereby preventing D_2 and O_2 recombination from occurring. Miles conducted three co-deposition experiments using the Dewar cells. The results, summarized in Figure 1 *a*, show that all three experiments produced excess heat. The amount of excess heat produced by co-deposition was comparable to that obtained using bulk Pd cathodes. Additional electrochemical studies showed that the excess heat was not due to any shuttle reactions⁷. Periodically in these experiments, calibration heating pulses were applied that cause an increase in cell temperature. When no excess power was present or when the excess power remained constant, the cell temperature relaxed back to the expected baseline upon termination of the heating pulse. The positive feedback effect (Figure 1 *b*) is observed when the cell temperature did not relax back to the original baseline upon cessation of this added thermal input. While the cell temperature increased, the cell potential at constant current

decreased. This positive feedback effect is the simplest and clearest indication of excess power.

Letts⁸ designed a Seebeck calorimeter with an output power variation of ± 0.01 W, a precision within 1%, and a sensitivity of 5 mW. This calorimeter was used in a series of Pd/D co-deposition experiments that were conducted with a recombiner and in the presence of a 675 G magnetic field at the cathode. The cell contained a gold-plated copper cathode inside a Pt anode coil as well as Ti cathode placed outside of the Pt anode coil. Because Pd does not plate out on Ti, the latter is an ideal inert cathode that can be used for calibration purposes. Consequently, both the inert Ti cathode and the gold plated Cu cathode/Pt anode assembly were placed in the PdCl₂/LiCl electrolyte at the same time. The calorimeter and cell were designed to permit changing or repositioning of the electrodes and removal/replacement of the electrolyte while the experiment was running. Figure S2 (see Supplementary Information online) shows the output of this experiment⁹. Up to point 800, the inert Ti cathode was electrolysed at 3 W outside the anode coil. During this time the cell was in power balance to within 1%, i.e. no excess heat production was observed. At point 800,

electrolysis was diverted to the gold-plated copper cathode inside the Pt anode coil. Pd/D co-deposition was then done on the gold-plated copper cathode. An excess power signal of 250 mW appeared quickly and cell temperature increased at the same time by 4–5°C. The excess power signal and cell temperature remained elevated, producing ~7 kJ of excess energy. At point 1100, a pump was activated to remove the deuterium-based electrolyte and replace it with hydrogen-based electrolyte of identical volume and concentrations of PdCl₂ and LiCl used at the start of the D₂O experiment. As seen in Figure S2 (see Supplementary Information online), the excess power signal declined over a 9 h period after replacing D₂O with H₂O, which demonstrated an isotopic dependence. Letts also discovered that geometry made a difference whether or not excess heat was observed. In Pd/D co-deposition experiments done with the gold-plated copper cathode placed outside the Pt coil, no excess heat generation was observed. This demonstrates the need for good symmetry between the anode and cathode in experiments.

Cravens and Letts¹⁰ explored using chemical additives to trigger excess heat in Pd/D co-deposition. In these experiments, a 0.636 cm diameter copper tube was used as the cathode. The copper tube was sealed by crimping. Gold was plated on the tube and then masked with Teflon tape leaving a 0.6 cm wide, unmasked region available for co-deposition. A thermistor was inserted inside the tube. The cathode was first plated with palladium for use in a series of laser stimulation trials using 660 nm excitation. As shown in Figure S3 (see Supplementary Information online), no significant excess thermal output was observed. Then about 10 drops of a separate plating material was used that contained additional metal ions. The plating additive was derived from Pd, Rh, Ce, La and U salts in acidified D₂O. U and Rh increase the absorption of D within the palladium black. La and Ce mischmetal were added to allow for spin exchanges to aid spin conservation terms for wave functions in the D + D → He nuclear channel. The addition of Ce also increases the diffusion rate of deuterium within Pd. The addition of U and La created a convoluted surface. It is worth noting that the rationale for the addition of U (natural abundance) and Ce mischmetal was to supply metals with a non-zero quadrupole. Such materials are required for coupling of the nuclear spins to the phonon states, as is known within the area of nuclear acoustics resonance. This is thought to provide pathways for both spin polarization of the deuterium and for thermal energy release. The results of the deposition of the additive components are shown in Figures S3 and S4 (see Supplementary Information online). The excess reached approximately 2 W from the small 0.6 cm length of plated area and persisted for a day before the experiment was terminated. Figure S4, supplementary material, shows that the cathode temperature response is faster than the electrolyte. This indicates the source of the heat is the cathode.

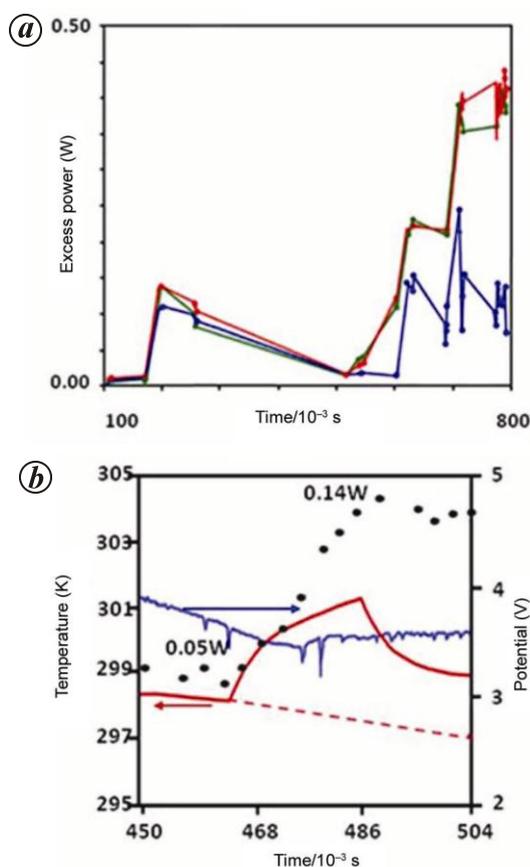


Figure 1. *a*, Excess power measurements in three co-deposition experiments done using an open cell. The Cu cathode used for co-deposition was placed inside a Pt coil anode. *b*, The cell temperature and cell potential curves, during the application of a resistive heater calibration pulse for 6 h. This illustrates the positive feedback effect.

Swartz and Verner^{11,12} prepared spiral-wound palladium cathodes (see [Figure S5 Supplementary Information online](#)). In their experiments, the Pd cathode and Pt anode were immersed in low electrical conductivity D₂O (with no additional electrolyte) that contained 8.2 mM PdCl₂. The spiral cathode system, with its open helical cylindrical geometry, on a high electrical resistance solution, created a unique and unusual electric field distribution that resulted in asymmetric electrolysis on one side of the cathode. This caused deuteron flux inside the metal lattice. Excess heat was measured using a modified dual ring calorimeter with redundant thermometry ([Figure S6 a, see Supplementary Information online](#)), waveform reconstruction, full thermal controls, and time integration ([Figure S6 b, see Supplementary Information online](#)). Figure S6 a shows the input electrical power and thermometry of a Pd co-deposition done on the spiral Pd cathode and an ohmic thermal resistor as control. For the same power input to the ohmic control and the cold fusion component, there is a higher temperature resultant in the latter. Figure S6 b shows the calorimetry of the same experiment. Four curves are shown in Figure S6 b, two of which involve power. It can be seen that the observed power output is much greater for the deuterium-loaded system. Two additional curves on Figure S6 b confirm that the excess heat is not simply stored energy. It can be seen that for the ohmic joule (thermal) control, the integrated energies of the input and output arise in parallel. In contrast, in the deuterium-loaded heavy water system, there is a gap, which increases with time. This shows excess heat of the co-deposition system (more than 100,000 joules excess energy over ~ 15 h compared to the ohmic resistor).

Tanzella *et al.*¹³ loaded and stabilized 50–250 μm diameter PdD_x and PdH_x wires. Deuterium loading of these wires was determined by measuring the resistance of the wires using the four-point probe method. They also did co-deposition on 50–250 μm diameter PdD_x, PdH_x, and Ag wires, which they also stabilized. They then placed these wires in a cryogenic calorimeter and measured the energy released from destructive electrodiffusion of these wires. Generally, the D-loaded metals yielded greater and more reliable excess energy than the H-loaded metals. The co-deposited PdD_x on highly loaded PdD_x wires yielded greater excess energy than the bulk metal hydrides. The co-deposition process generated many point vacancies in the co-deposited layer and these vacancies were stabilized by co-deposited D/H. These vacancies were able to host molecular D₂. These vacancies, along with high loading of D and its flux, are necessary for excess heat generation.

In yet another variation on co-deposition, Dash and Ambadkar¹⁴ used Pd as the anode and Pt as the cathode. Both electrodes were immersed in a D₂O–H₂SO₄ electrolyte in a cell with a recombiner. When a constant current was applied, Pd dissolved from the Pd anode and co-deposited with hydrogen isotopes on the Pt cathode. After

229 h of electrolysis, the excess thermal power output was 0.93 ± 0.1 W. No excess power was measured in a closed control cell comprised of two Pt electrodes immersed in a H₂O–H₂SO₄ electrolyte. At the end of the experiment, the Pt/Pd cathode was subjected to SEM/EDS analysis. The results are summarized in [Figure S7 \(see Supplementary Information online\)](#). The EDS spectrum, Figure S7 b, of the bright dendritic particles in Figure S7 a, shows the presence of Pd. The ratio of the Pd Lα X-ray peak at 2.84 KeV to the Pd Lβ X-ray peak at 2.99 KeV was 0.45. The ratio of these two peaks in the EDS spectrum (Figure S7 c) obtained for the black area shown in Figure S7 a was 0.71. This increase is caused by the overlap of Ag Lα with Pd Lβ. Since the presence of Ag was not homogeneous over the surface of the cathode, this Ag is not due to contamination.

Additional co-deposition experiments by other research groups have shown evidence of transmutation as well as tritium production and the emission of energetic particles. Szpak *et al.*¹⁵ conducted Pd/D co-deposition experiments in the presence of an external electric field that had a 6% AC ripple which allowed the electric field to couple into the cathode. Upon termination of the experiments, the cathodes were subjected to SEM analysis which showed the presence of craters and other features indicative of localized melting of the Pd deposit ([Figure S8 a–c, see Supplementary Information online](#)). EDX analysis of these features showed the presence of elements such as Ca, Mg, Si, Zn and Al in addition to Pd (Figure S8 d). These new elements are not due to contamination as their distribution over the cathode was not homogeneous.

Using a recombiner in a separate chamber, Bockris *et al.*¹⁶ measured the tritium content, as a function of time, in the liquid and gas phases of co-deposition experiments. The results are summarized in [Figure S9 \(see Supplementary Information online\)](#). Bursts of tritium production were observed in both the gas and liquid phases when low tritiated water was used. However, when highly tritiated water was used, an overall decrease in tritium was observed. Similar results were reported by Lee *et al.*¹⁷ using closed cells and measuring the tritium content of the electrolyte.

Mosier-Boss *et al.*¹⁸ 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 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. Similar damage was observed when a CR-39 detector was wrapped with Cu screen and exposed to a ¹³⁷Cs γ-ray source, suggesting that this damage is due to X-rays. Tracks were observed for co-deposition on Ni screen done in the presence of an external *E/B* field. In contrast, for Pd/D co-deposition done on either Au, Ag or Pt cathodes, tracks were obtained in both the presence

and absence of an external E/B field. These results led to the following composite cathode experiment in which Au was electroplated on half of the Ni screen, as shown in [Figure S10 \(see Supplementary Information online\)](#). This composite cathode was placed in contact with a CR-39 detector and used in a Pd/D co-deposition experiment. At the end of the experiment, the detector was etched and analysed. The results show that no tracks were obtained on the bare Ni half of the cathode. The impression of the Ni screen was observed. However, tracks were observed on the Au-coated Ni screen. Both halves of the cathode experienced the same chemical and electrochemical environment at the same time. If the pitting in CR-39 was due to chemical attack, those reactions would have occurred on both the bare Ni and Au-coated Ni halves of the cathode and both halves would have shown pitting of the CR-39 detector. But this was not observed. Therefore, the tracks observed in the CR-39 detector were not the result of chemical attack.

In addition to tracks due to energetic particles, Mosier-Boss *et al.*¹⁹ also reported triple tracks in CR-39 detectors. These triple tracks are diagnostic of the $^{12}\text{C}(n, n')3\alpha$ carbon break-up reaction due to the reaction of >9.6 MeV neutrons with a carbon atom in the detector. Examples of these triple tracks as well as their corresponding DT neutron generated triple tracks are shown in [Figure S11 \(see Supplementary Information online\)](#). No triple tracks were observed in CR-39 detectors used in control experiments.

Conclusions

Several researchers have used Pd/D co-deposition to investigate the phenomenon of condensed matter nuclear reactions within a Pd lattice. The emphasis of many of these investigations has been on heat production. In these particular experiments, excess heat has been measured using different variations of co-deposition as well as different kinds of calorimeters, both open and closed. In addition to heat, other reaction products that have been observed include new elements, tritium, energetic particles and neutrons.

1. Szpak, S. *et al.*, Charging of the Pd/ ^nH system: role of the interphase. *J. Electroanal. Chem.*, 1992, **337**, 147–163.

2. Szpak, S. *et al.*, Cyclic voltammetry of Pd + D codeposition. *J. Electroanal. Chem.*, 1995, **380**, 1–6.
3. Szpak, S., Mosier-Boss, P. A. and Smith, J. J., Deuterium uptake during Pd–D codeposition. *J. Electroanal. Chem.*, 1994, **379**, 121–127.
4. Szpak, S., Mosier-Boss, P. A. and Smith, J. J., On the behavior of Pd deposited in the presence of evolving deuterium. *J. Electroanal. Chem.*, 1991, **302**, 255–260.
5. Mosier-Boss, P. A. *et al.*, Review of twenty years of LENR research using Pd/D co-deposition. *J. Condens. Matter Nucl. Sci.*, 2011, **4**, 173–187.
6. Szpak, S. *et al.*, Thermal behavior of polarized Pd/D electrodes prepared by co-deposition. *Thermochim. Acta*, 2004, **410**, 101–107.
7. Miles, M. H., Investigations of possible shuttle reactions in co-deposition systems. *J. Condens. Matter Nucl. Sci.*, 2012, **8**, 12–22.
8. Letts, D., Codeposition methods: a search for enabling factors. *J. Condens. Matter Nucl. Sci.*, 2011, **4**, 81–92.
9. Letts, D. and Hagelstein, P. L., Modified Szpak protocol for excess heat. *J. Condens. Matter Nucl. Sci.*, 2012, **6**, 44–54.
10. Cravens, D. J. and Letts, D. G., Practical techniques in CF research-triggering methods. In Proceedings of the Tenth International Conference on Condensed Matter Nuclear Science, Cambridge, MA, 2003.
11. Swartz, M. and Verner, G., Excess heat from low electrical conductivity heavy water spiral-wound Pd/D₂O/Pt and Pd/D₂O–PdCl₂/Pt devices. In Proceedings of the Tenth International Conference on Condensed Matter Nuclear Science, Cambridge, MA, 2003.
12. Swartz, M. R., Codeposition of palladium and deuterium. *Fusion Technol.*, 1997, **32**, 126–130.
13. Tanzella, F. *et al.*, Stimulation of PdD_x wires at cryogenic temperatures. In Proceedings of the Sixteenth International Conference on Condensed Matter Nuclear Science, Chennai, India, 2011.
14. Dash, J. and Ambadkar, A., Co-deposition of palladium with hydrogen isotopes. In Proceedings of the Eleventh International Conference on Condensed Matter Nuclear Science, Marseille, France, 2004.
15. Szpak, S. *et al.*, Evidence of nuclear reactions in the Pd lattice. *Naturwissenschaften*, 2005, **92**, 394–397.
16. Bockris, J. O'M. *et al.*, Tritium and helium production in palladium electrodes and the fugacity of deuterium therein. In Proceedings of the Third International Conference on Condensed Matter Nuclear Science, Nagoya, Japan, 1992.
17. Lee, K.-H., Jang, H. and Kim, S.-J., A change of tritium content in D₂O solutions during Pd/D co-deposition. In Proceedings of the Seventeenth International Conference on Condensed Matter Nuclear Science, Daejeon, Korea, 2012.
18. Mosier-Boss, P. A. *et al.*, Use of CR-39 in Pd/D co-deposition experiments. *Eur. Phys. J. Appl. Phys.*, 2007, **40**, 293–303.
19. Mosier-Boss, P. A. *et al.*, Comparison of Pd/D co-deposition and DT neutron generated triple tracks observed in CR-39 detectors. *Eur. Phys. J. Appl. Phys.*, 2010, **51**, 20901.