

SPAWAR Systems Center-Pacific Pd:D Co-Deposition Research: Overview of Refereed LENR Publications

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Abstract

Scientists at the US Navy SPAWAR Systems Center-Pacific (SSC-Pacific), and its predecessors, have had extraordinary success in publishing LENR papers in peer-reviewed journals. This success hasn't come easily and is due to several factors. One key reason for this success was the courage of the SSC-Pacific upper management in allowing scientists to conduct research and publish results in a controversial field. The few journal editors, who had the fortitude to consider our work, also contributed to this success. This contrasts with the majority of their peers who, taking the path of least resistance, ignored our work out of hand and returned manuscripts with, 'the subject matter is not in the purview of the journal'. The reviewers also played a role in the successful publication of LENR-related papers. A multitude of reviewers, many outside the LENR field, had to put aside their biases and look objectively at our data. In turn, the reviewers' relentless concerns forced us to tenaciously address their issues. Ultimately, the SSC-Pacific team published 21 refereed papers in seven journals and a book chapter, spanning 19 years beginning in 1989. This paper is a brief synopsis of those publications.

1. S. Szpak, P.A. Mosier-Boss, and J.J. Smith, "On the Behavior of Pd Deposited in the Presence of Evolving Deuterium," *J. Electroanal. Chem.*, **302** (1991) 255-260

This was a preliminary note introducing the Pd/D co-deposition protocol as an alternative experimental approach to initiate LENR. Temperature measurements using thermocouples placed in the cathode and solution show that the cathode was hotter than the solution. This indicates that the observed heat is not due to Joule heating. A ten fold increase in tritium content in the solution was observed. Experiments were conducted with photographic film in close proximity of the cathode. After development, the film showed a grid pattern due to the Ni screen cathode and was attributable to the emission of soft X-rays.

2. S. Szpak, C.J. Gabriel, and R. J. Nowak, "Electrochemical Charging of Pd Rods," *J. Electroanal. Chem.*, **309** (1991) 273-292

A model was developed to describe the electrochemical charging of palladium rods. This model coupled the interfacial processes with the transport of interstitials in the electrode interior. It was shown that boundary conditions arise from the solution of equations governing the elementary adsorption-desorption and adsorption-absorption steps as well as the symmetry of the electrode.

3. S. Szpak, P.A. Mosier-Boss, S.R. Scharber, and J.J. Smith, "Charging of the Pd/ⁿH System: Role of the Interphase," *J. Electroanal. Chem.*, **337** (1992) 147-163.

Slow scan cyclic voltammetric studies of Au/Pd/ⁿH were conducted to examine the dynamics of transport of electrochemically deuterium/hydrogen across the electrode/electrolyte interphase. It was found that a coupled, two-layer model of the interphase describes the observed behavior as a function of scan rate and electrolyte composition. The effect of chemisorbing species, thiourea, and pH on the transport across the interphase was also investigated.

4. S. Szpak, P.A. Mosier-Boss, C.J. Gabriel, and J.J. Smith, "Absorption of Deuterium in Palladium Rods: Model vs. Experiment," *J. Electroanal. Chem.*, **365** (1992) 275-286.

A model that incorporates variables such as electrochemical rate constants, bulk diffusion coefficient, and charging current has been developed. Such a model can be used to predict the overpotential, surface coverage, and bulk loading of the electrode during charging. The computed time dependence of the bulk loading has been compared with published experimental charging curves. Microscopic examination of a charging Pd cathode using Nomarski optics has shown that, even within a single grain, there are preferred sites of absorption. *In-situ* XRD measurements of the charging Pd cathode shows that deuterium preferentially enters the Pd lattice through the 111 sites. With additional charging, a broadening and a shift to lower 2θ angles was observed which suggested the presence of a supercharged layer.

5. S. Szpak, P.A. Mosier-Boss, R.D. Boss, and J.J. Smith, "Comments on the Analysis of Tritium Content in Electrochemical Cells," *J. Electroanal. Chem.*, **373** (1994) 1-9.

The time dependence of tritium content of an open cell operating galvanostatically with intermittent sampling has been derived and is given by the following expression:

$$f(t) = f(0) \left(\frac{m(0) - r(i)t}{m(0)} \right)^{s-1} + \frac{q}{(S-1)r(i)} \bullet \left\{ 1 - \left[\frac{m(0) - r(i)t}{m(0)} \right]^{s-1} \right\}$$

where f is the tritium mass fraction, m is the mass of the electrolyte phase, $r(i)$ denoted the rate of change associated with the cell current, q is the rate at which tritium is added/removed, and s is the isotopic separation factor. It was concluded that a complete mass balance between the liquid and gas phases was necessary in order to determine that tritium was produced in the Pd/D system.

6. S. Szpak, P.A. Mosier-Boss, and J.J. Smith, "Deuterium Uptake During Pd-D Codeposition," *J. Electroanal. Chem.*, **379** (1994) 121-127.

Deuterium uptake during Pd-D co-deposition was examined using galvanostatic perturbation techniques. The resultant potential relaxation curves exhibit four distinct potential-time intervals where the relaxation process is controlled by the interaction between the transport of deuterium from inside the lattice to the surface to form adsorbed deuterium and the reduction of palladium from solution.

7. S. Szpak, P.A. Mosier-Boss, S.R. Scharber, and J.J. Smith, "Cyclic Voltammetry of Pd + D Codeposition," *J. Electroanal. Chem.*, **380** (1995) 1-6.

Processes associated with the Pd + D alloy codeposition were examined by cyclic voltammetry. The dynamics of the interphase region are discussed.

8. S. Szpak, P.A. Mosier-Boss, and J.J. Smith, "On the Behavior of the Cathodically Polarized Pd/D System: Search for Emanating Radiation," *Phys. Lett. A*, **210** (1996) 382-390.

Pd/D co-deposition experiments were conducted inside lead caves while measuring gamma and X-rays, as a function of time, using a HPGe detector with an Al window and a Si(Li) detector with a Be window. The cathodically polarized Pd/D system was observed to emit X-rays with a broad energy distribution and with an occasional emergence of recognizable peaks attributable to the Pd K_{α} and Pt L lines. The emission of X-rays is sporadic and of limited duration.

9. S. Szpak and P.A. Mosier-Boss, "On the Behavior of the Cathodically Polarized Pd/D System: A Response to Vigier's Comments," *Phys. Lett. A*, **221** (1996) 141-143.

Preliminary results of thermal imaging of the Pd/D cathode prepared using the co-deposition technique are presented. Hot spots are observed that appear/disappear chaotically. With time these hot spots merge into larger islands that exhibit oscillatory behavior. SEM images of a Pd/D cathode that had melted during electrolysis are shown.

10. S. Szpak, P.A. Mosier-Boss, R.D. Boss, and J.J. Smith, "On the Behavior of the Pd/D System: Evidence for Tritium Production," *Fus. Technol.*, **33** (1998) 38-51.

In these experiments, the D_2 and O_2 gases were recombined in a separate chamber. The tritium content in the liquid and gas phases were measured daily using a liquid scintillation. The measured data were analyzed using the mass balance expression that was derived earlier. It was observed that tritium production occurred in bursts and sporadically. During a burst, the rate of tritium production was estimated to be 10^3 to 10^4 atoms s^{-1} . Tritium produced during prolonged electrolysis was transported out of the electrode interior by two distinct paths. One path results in enrichment of tritium in both the electrolyte and gas phases. The second results in enhancement only in the gas phase.

11. S. Szpak and P.A. Mosier-Boss, "On the Release of 3H from cathodically polarized Palladium Electrodes," *Fus. Technol.* **34** (1998) 273-278.

The release paths for tritium produced during electrochemical compression of deuterium in a Pd lattice were examined. The results indicate that tritium production requires high D/Pd atomic ratios. This requirement is met if there are no channels reaching the contact surface. The electrogenerated tritium is distributed among the voids and bulk material. Gas evolution promotes a continuous exchange between the 3H atoms residing in the subsurface layer and with those in the adsorbed state. Atoms in the adsorbed state exchange with the molecules of the contacting electrolyte phase or gaseous phase, leading to two distinct transfer paths.

12. P.A. Mosier-Boss and S. Szpak, "The Pd/ 3H System: Transport Processes and Development of Thermal Instabilities," *Nuovo Cimento Soc. Ital. Fis. A*, **112** (1999) 577-587.

The surface temperature distribution of the cathode prepared by Pd/D co-deposition on a Ni screen was measured using an infrared camera. It was observed that, unlike joule heating, excess enthalpy generation occurs in the form of localized events in close proximity to the contact surface. It was also observed that, the higher the electrolyte temperature, the more frequent the events. In the limit, these events overlap to produce oscillating islands.

13. S. Szpak, P.A. Mosier-Boss, and M. H. Miles, "Calorimetry of the Pd + D Codeposition," *Fus. Technol.*, **36** (1999) 234-241.

Calorimetric measurements indicate that the excess enthalpy generated in cells using cathodes prepared by the co-deposition process is, on average, higher than that produced in cells using solid Pd rods. Infrared imaging of the cathodes prepared by Pd/D co-deposition shows that the heat sources are highly localized. The steepness of the temperature gradients indicate that the heat sources are located in close proximity to the electrode-solution contact surface.

14. S. Szpak, P.A. Mosier-Boss, M. H. Miles, and M. Fleischmann, "Thermal Behavior of Polarized Pd/D Electrodes Prepared by Co-deposition," *Thermochimica Acta*, **410** (2004) 101-107.

The thermal behavior of Pd/D electrodes, prepared by the co-deposition technique, was examined using a Dewar-type electrochemical cell calorimeter. Results indicated that excess enthalpy is generated during and after the completion of the co-deposition process. The rates of excess enthalpy generated using the co-deposition technique were higher than those obtained using Pd wires or other forms of Pd electrodes. Positive feedback and heat-after-death effects were observed. The rates of excess power generation were found to increase with an increase in both cell current and cell temperature.

15. S. Szpak, P.A. Mosier-Boss, C. Young, and F.E. Gordon, "The Effect of an External Electric Field on Surface Morphology of Co-deposited Pd/D Films," *J. Electroanal. Chem.*, **580** (2005) 284-290.

After plating out the Pd on a Au foil, the cell current was increased and an external electric field was applied across the cell. The experiment was terminated after 48 h. The cell was disassembled and the cathode was subjected to analysis using an SEM. In the absence of an external electric field, the Pd deposit exhibits a cauliflower structure. After exposure to an external electric field, significant changes in the morphology of the Pd/D deposit were observed. Fractal features were observed as well as dendritic growths, rods, wires, and craters. Considerable work is needed to account for the variety of shapes. The process of shape change is driven by energy transferred from the electrostatic field and directed by the field.

16. S. Szpak, P.A. Mosier-Boss, C. Young, and F.E. Gordon, "Evidence of Nuclear Reactions in the Pd Lattice," *Naturwissenschaften*, **92** (2005) 394-397.

When a cathode prepared by Pd/D co-deposition is subjected to an external electrostatic field, SEM analysis of the deposit shows discrete sites exhibiting molten-like features. Such features require substantial energy expenditure in order to form. EDX analysis of these features shows the presence of new elements (Al, Mg, Ca, Si, Zn,...) that could not be extracted from cell components.

17. S. Szpak, P.A. Mosier-Boss, and F.E. Gordon, "Further Evidence of Nuclear Reactions in the Pd/D Lattice: Emission of Charged Particles," *Naturwissenschaften*, **94** (2007) 511-514.

CR-39 is a solid state nuclear track detector that is used to detect energetic particles such as alphas, protons, tritons, and helium-3. Pd/D co-deposition was done, in the presence of an external electric or magnetic field, with the cathode in direct contact with a CR-39 detector. Tracks on the CR-39 detector were observed where the cathode was in contact with the plastic indicating that the source of the tracks is the cathode. The features of these tracks (optical contrast, shape, and bright spot in the center of the pit) are consistent with those observed for pits in CR-39 that are of a nuclear origin. The emission of the energetic particles is sporadic and occurs in bursts.

18. P.A. Mosier-Boss, S. Szpak, F.E. Gordon, and L.P.G. Forsley, "Use of CR-39 in Pd/D Co-deposition Experiments," *Eur. Phys. J. Appl. Phys.*, **40** (2007) 293-303.

A series of control experiments were conducted. It was shown that the tracks observed in CR-39 detectors subjected to Pd/D co-deposition were not due to radioactive contamination of the cell components. No tracks were observed when Cu was electrochemically plated on the surface of the CR-39 detectors. This indicates that the pits cannot be attributed to chemical attack of the surface of the CR-39 by either D₂, O₂, or Cl₂ present in the electrolyte. Nor can the pits be attributed to the metal dendrites piercing into the surface of the detectors. Additional experiments showed that LiCl is not essential for the production of pits and that the density of pits significantly decreases when light water is substituted for D₂O. Quantitative analysis using an automated scanner shows that there are three populations of tracks (0.1-0.5 μm, 0.9-4.0 μm, and 4.1-12 μm) and that the pits can be either perfectly circular or elliptical in shape.

19. P.A. Mosier-Boss, S. Szpak, F.E. Gordon, and L.P.G. Forsley, "Detection of Energetic Particles and Neutrons Emitted during Pd:D Co-deposition," *Low Energy Nuclear Reactions Source Book*, American Chemical Society, Chapter 14, pp 311-334. (2008).

Co-deposition procedures and control experiments specifically identified the conditions under which nuclear particles were observed, and ruled out chemical means of mimicking nuclear tracks. The nuclear tracks are quantitatively examined and are consistent with neutron knock-ons. Triple tracks are presented as evidence of ¹²C(n,n')3α indicative of DT fusion.

20. P.A. Mosier-Boss, S. Szpak, F.E. Gordon, and L.P.G. Forsley, "Triple Tracks in CR-39 as the Result of Pd/D Co-deposition: Evidence of Energetic Neutrons," *Naturwissenschaften*, in press. (2008).

Triple tracks have been observed in CR-39 detectors used in Pd/D co-deposition experiments. Microscopic examination of the bottom of the triple track pit shows that the three lobes of the track are splitting apart from a center point. The presence of three α-particle tracks outgoing from a single point is diagnostic of the ¹²C(n,n')3α carbon break up reaction and is easily differentiated from other neutron interactions occurring within the CR-39 detector. The presence of triple tracks suggests that DT reactions that produce ≥9.6 MeV neutrons are occurring inside the Pd lattice.

21. P.A. Mosier-Boss, S. Szpak, F.E. Gordon, and L.P.G. Forsley, "Use of CR-39 in Pd/D Co-deposition Experiments: A Response to Kowalski," *Eur. Phys. J. Appl. Phys.*, **41** (2008) in press.

Earlier we reported that the pits generated in CR-39 detectors during Pd/D co-deposition experiments are consistent with those observed for pits that are of a nuclear origin. Recently, that interpretation has been challenged. In this communication, additional experimental data and further analysis of our earlier results are provided that support our original conclusions.