



Research Article

A Review on Nuclear Products Generated During Low-Energy Nuclear Reactions (LENR)

Pamela A. Mosier-Boss *

Research Laboratory of Electronics, MIT, Cambridge, MA 02139, USA

Abstract

Given the response to the Fleischmann–Pons news conference in 1989, it became clear to many researchers in the field that excess heat was not convincing enough evidence to prove that nuclear processes were occurring inside a metal lattice. Skeptics attributed the excess heat to recombination of deuterium and oxygen gases and/or poor calorimetry, despite the fact that control experiments showed that this was not the case. Consequently, a number of researchers redirected their efforts from measuring heat to looking for nuclear products such as neutrons, charged particles, X- and gamma rays, and transmutation. The results of these efforts are discussed in this communication.

© 2012 ISCMNS. All rights reserved.

Keywords: Charged particles, Helium-4, Neutrons, Transmutation, X- and gamma rays

1. Introduction

When Stanley Pons and Martin Fleischmann held their press conference on March 23, 1989, the only evidence they had that nuclear processes were occurring inside the palladium lattice was excess heat. Their electrochemical cells were producing more heat than could be accounted for by chemical means. The lack of collaborating nuclear products, particularly neutrons, outraged many scientists. But that did not stop many scientists world wide from going into their laboratories to replicate the Fleischmann–Pons experiment and to look for evidence that the effect had a nuclear origin. Many of these efforts failed and the reasons for the failures are now understood. They include improper cell configurations causing inhomogeneous deuterium loading of the palladium, cracks in the palladium rods used in the experiments which impeded deuterium loading, and the lack of recognition that there was an incubation time needed to load the palladium rods with deuterium. In each of these instances, the experimental conditions used failed to achieve the high deuterium loadings and flux inside the lattice needed to initiate the effect. Since the Fleischmann–Pons announcement, continuing research, using a wide variety of techniques from electrolysis, gas loading, to glow discharge, has yielded evidence that supports the nuclear nature of the effect. Nuclear products that have been observed include tritium, neutrons, charged particles, gamma and X-rays, transmutation, as well as helium-4. A brief discussion

*E-mail: pboss@san.rr.com

of the experiments performed and their results follows. It is by no means complete. Yet it does illustrate the number of techniques and approaches that have been taken to measure LENR nuclear products. The fact that these different approaches yield similar results strengthens the validity of those results.

2. Neutrons and Tritium

In the March 23, 1989 press conference, Pons claimed that they had “established a sustained nuclear fusion reaction by means which are considerably simpler than traditional techniques.” He further claimed that the deuterium is driven inside a metal rod to such an extent that fusion between the deuterons occurs. Because of these claims, researchers conducted experiments looking for DD fusion products.

In thermonuclear fusion, the DD fusion reactions are:



In thermonuclear fusion, the branching ratio for reactions (1) and (2), n/T , is 1. That is, 50% of DD fusion follows reaction (1) and 50% follows reaction (2). Reaction (3) occurs 10^{-6} of the time and is negligible. If deuteron fusion reactions were occurring inside the palladium lattice and if those reactions were similar to what was observed in plasma fusion, expected products would include tritium and neutrons. Because of these expectations, early investigators focused on detecting tritium and 2.45 MeV neutrons.

When researchers at India’s Bhabha Atomic Research Center (BARC) heard the Fleischmann–Pons announcement, they began to conduct electrolysis experiments using a Milton-Roy electrolytic cell, Fig. 1(a) [1]. The cathode area was 300 cm^2 and the cells could operate up to 100 A current. During electrolysis, the cell was monitored using two different kinds of neutron detectors – BF_3 counters embedded in paraffin blocks for thermal neutron detection and proton recoil plastic scintillator counters for fast neutron detection. The background was monitored using ${}^3\text{He}$ counters embedded in paraffin. At the same time they recorded cell current, voltage, and temperature, and they collected samples for tritium analysis. Figure 1(b) shows the results of the BF_3 and proton recoil neutron detectors during the first run in April of 1989. Clearly the detectors track one another. As the current was slowly increased to 100 A, the cell overheated which caused the trip circuit to turn off cell power. This was followed by a burst of neutrons that was approximately two orders of magnitude larger than background levels over a 2 min interval. Of the 11 cells, six saw a neutron signal within 9 h of operation, one within 24 h, and two showed a neutron signal after 2 weeks. The neutron emissions were observed to stop after continued electrolysis. It was also observed that high D/Pd loadings were not required to obtain a neutron emission. But more importantly, it was observed that, although neutrons and tritium were produced simultaneously, the branching ratio (n/T) was not 1 as observed in thermonuclear fusion. For the Pd/D system, the n/T ratio was measured to be $\sim 10^{-7}$.

Iwamura et al. [2] of Mitsubishi Heavy Industries gas-loaded palladium foils with deuterium to a D/Pd loading of 0.66. Once loaded, the deuterated foils were sealed with either a thin aluminum or gold film. The samples were then placed in a vacuum chamber and set on a heater. The chamber was equipped with two ${}^3\text{He}$ neutron detectors, a NaI scintillator detector for gamma-ray spectroscopy, and a high-resolution quadrupole mass spectrometer for gas analysis. The samples were heated to 400 K causing the release of deuterium. During deuterium gas desorption from the samples, they observed the emission of neutrons and tritium as well as charged particles. No gamma rays were detected using these palladium structures.

A consortium of scientists from the Russian Academy of Sciences, Hokkaido University, New Hydrogen Energy Laboratory in Sapporo Japan, and Osaka University conducted experiments using Au/Pd/PdO heterostructures [3].

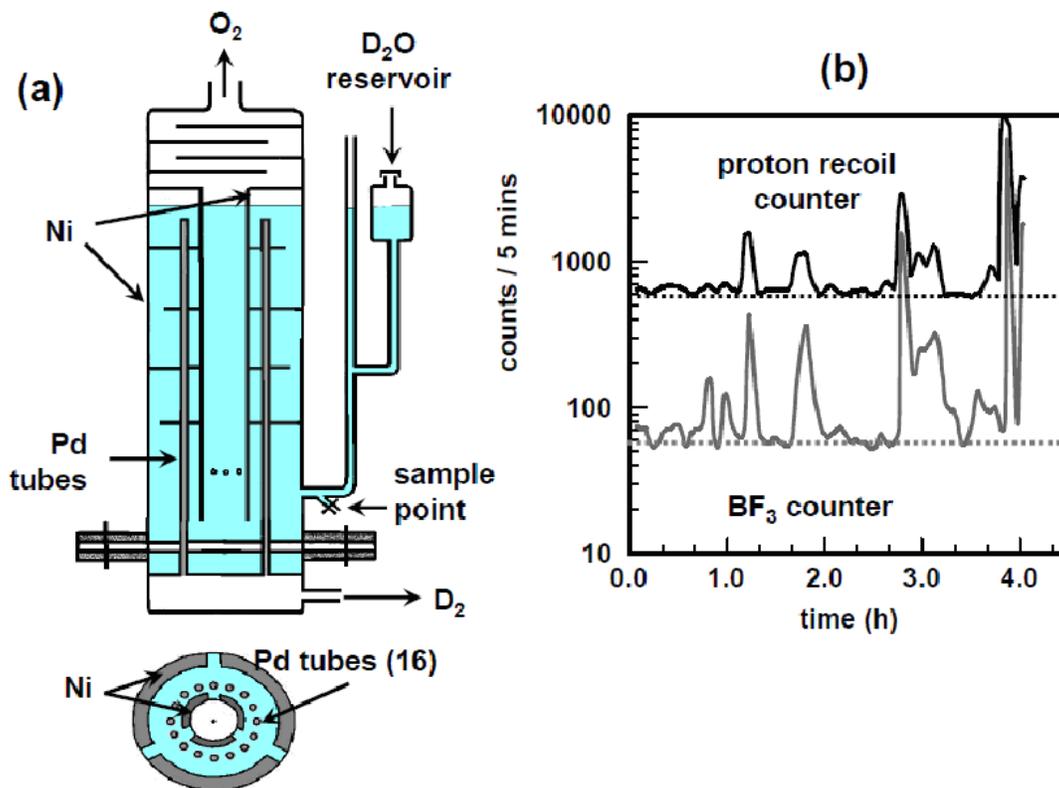


Figure 1. (a) Schematic of a Milton-Roy electrolytic cell used in the BARC experiments. AgPd tubes are 200 mm in height and 3 mm in diameter. (b) Neutron counts measured as a function of time using a proton recoil counter (top) and a BF_3 counter (bottom). Dotted lines indicate background count rates for both detectors. Reproduced with permission from Srinivasen

These heterostructures possess high deuterium loading capacity, high mobility of deuterium at the Pd–PdO interface, and a screening potential, U_S , of 600 eV. After electrochemically loading the Au/Pd/PdO heterostructures with deuterium, they took the samples out of the electrolyte and monitored the exothermic desorption of deuterium using a detection system based upon a liquid scintillator that responds to neutrons and gamma rays, a silicon surface barrier detector that detects energetic particles, and CR-39 detectors that responds to both charged particles (alphas, protons, and tritons) and neutrons. Using this array of detectors, they saw charged particles with energies between 1 and 3 MeV and neutrons between 2.4 and 2.8 MeV. They concluded that the energies of the charged particles and neutrons were consistent with those resulting from DD fusion reactions, (1) and (2). However, they were not able to determine the branching ratios of those reactions.

Other researchers who have measured tritium in electrolytic cells include Ed Storms [4] of Los Alamos National Laboratories, John Bockris [5] of Texas A&M State College, and Szpak et al. [6] of SPAWAR Systems Center Pacific. Storms measured tritium, in both the gas and liquid phases, during electrolysis using palladium and palladium alloys with lithium, carbon, sulfur, boron, and beryllium. In these studies, excess tritium was observed in $\sim 10\%$ of the cells. No excess tritium was measured for the Pd alloys, which were prepared by arc-melting Pd powder with the other element under an argon atmosphere. Storms also looked at the effect of various surface treatments on the production of

tritium. He showed that palladium heated in dihydrogen sulfide to form a sulfide layer did not produce excess tritium while some cells using palladium heated in paraffin vapor did show excess tritium. Of the solution additives examined (thiourea, wheat flour, iron, silver, mercury, uranium-238, lithium sulfide, arsenic trioxide, and silver sulfide), only wheat flour and arsenic trioxide sometimes gave excess tritium.

John Bockris [5] also measured tritium in both gas and liquid phases. Out of 58 experiments using bulk palladium cathodes, 18 showed tritium production. He showed that the tritium production occurred in bursts and that the tritium did not correlate, either temporally or quantitatively, with the heat production. These experiments showed that the measured tritium was less than 0.1% of that needed to produce the observed heat, assuming DD reaction (1) is the heat producing channel.

Szpak et al. [6] used the palladium/deuterium co-deposition process in their experiments. Instead of bulk palladium electrodes, they started with a solution of palladium ions. When a current is applied, palladium plates out onto a cathode in the presence of evolving deuterium gas. Measurements were made of both the gas and the liquid. Out of ten experiments, six gave excess tritium. The tritium production was observed to occur in bursts. John Bockris replicated the co-deposition process and saw excess tritium in six out of nine cells. While the co-deposition process produced tritium more reliably than bulk palladium, the amount of tritium produced using the co-deposition process was orders of magnitude less than what had been observed for those bulk palladium cathodes that did produce tritium.

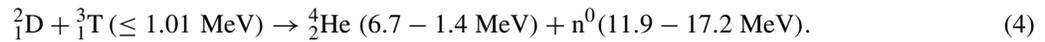
Claytor et al. [7], of Los Alamos National Laboratories, have shown tritium production from a low-voltage deuterium discharge on palladium and other metals. In their procedure, a wire is oriented perpendicular to and a few millimeters above a circular plate. Deuterium gas is added to the chamber and the wire is then pulsed negatively with currents between 2 and 5 A (voltages varied between 1500 and 2500 V) to create a light blue plasma (D^{2+}) with areas of pink (D^{3+} or D^+). Tritium concentration in the gas was monitored continuously using a Femtotech tritium gauge. At the end of the experiment, the tritium concentration was verified by collecting the gases, running them through a recombination chamber to convert the tritiated deuterium gas into tritiated heavy water. The tritium content of the resultant liquid was then determined using a liquid scintillator. The real-time measurements showed no tritium production when platinum wires were used in both hydrogen and deuterium gas discharges. No tritium was observed for the palladium/hydrogen gas system. However the palladium/deuterium gas system produced tritium at rates of 0.1–0.2 nCi/h (background is 0.002–0.01 nCi/h). It was found that the tritium output depended upon the temperature, pressure, and current applied to the cells. It was also sensitive to the purity and metallurgical condition of the palladium used in the experiments.

Neutrons have been measured by Mizuno et al. [8] of Hokkaido University, Francesco Scaramuzzi [9,10] of ENEA in Italy, Roussetski [11] of the P.N. Lebedev Physical Institute in Russia, and Mosier-Boss et al. [12,13]. In their experiments, Mizuno et al. [8] used three ^3He neutron detectors placed 50 cm above and apart from the cell. To reduce noise, the detectors were covered with an electromagnetic shield. Neutron emissions were detected by the coincidence and the anti-coincidence method with one detector that was covered with a cadmium film. The palladium wire cathodes used in these experiments were 3 cm long and 1 mm in diameter. After first loading the palladium wires in a heavy water electrolyte for 3 h, they were placed in a light water electrolyte and the cell voltage was increased from 30 to 85 V. With this voltage change, seven out of ten cells gave bursts of neutrons that lasted between 2.6 and 200 s. Total neutron counts during these bursts ranged from 25,800 to 1,573,800.

Shortly after the 1989 Fleischmann–Pons announcement, Scaramuzzi et al. [9,10] began to conduct deuterium gas-loading experiments of titanium shavings. Like palladium, titanium also absorbs hydrogen isotopes. A BF_3 neutron detector was used to monitor the gas loading. After immersion in a liquid nitrogen Dewar for a short period of time, the stainless steel chamber containing the deuteriated titanium was taken out of the Dewar to warm up and to create non-equilibrium conditions. Two events of neutron bursts were observed. The first event lasted for forty hours. However, the reproducibility of these experiments was very poor. In 2003, Keeney et al. [14] reported on similar gas loading experiments using titanium foil. They used ^3He neutron detectors. The reproducibility of their experiments was 40%. They also looked at electrolytic loading of the titanium foils. Although neutron emissions were observed,

the reproducibility was much less than that observed for the gas loading experiments.

In 2000, Roussetski [11] prepared PdO/Pd/PdO and PdO/Pd/Au samples that were electrochemically deuterated. After electrolysis, each sample was placed on CR-39, a solid state nuclear track detector (SSNTD), and the sample was heated to $\sim 50^\circ\text{C}$ to stimulate desorption of deuterium. This process was repeated a number of times. Analysis of the detectors showed tracks consistent with DD reaction products as well as triple tracks that are diagnostic of the carbon shattering reaction, $^{12}\text{C}(n,n')3\alpha$, caused by $a \geq 9.6$ MeV neutron. In plasma fusion, one potential source of these neutrons is the DT thermonuclear fusion reaction:



However, Roussetski found that the mean DT neutron flux emitted from the deuterated samples was higher than the yield of tritons from DD fusion reaction (1), suggesting that there was either another source of fast tritons or fast neutrons. This was the first evidence suggesting that both primary and secondary fusion reactions were occurring in the palladium/deuteride system.

Mosier-Boss et al. [12,13] have been monitoring their palladium/deuterium co-deposition experiments using CR-39. Using these detectors, they have observed tracks on both the front and the backsides of the detectors. The only particles that can traverse through 1 mm thick CR-39 detectors are ≥ 40 MeV alphas, ≥ 10 MeV protons, or neutrons. The size distribution of the tracks on the backside of the CR-39 detectors was consistent with the energy of protons and neutrons observed in DD fusion reactions (1) and (2), respectively, and of neutrons that are observed in DT fusion reaction (4). They also observed triple tracks [12] in the CR-39 detectors, like those shown in Fig. 2(a). In Fig. 2(a), the right hand images, which are an overlay of an image taken with the microscope optics focused on the surface of the detector and another taken with the optics focused on the bottom of the track, clearly show three particles breaking away from a center point. Very few of these triple tracks, usually between five and ten, are observed in the CR-39 detectors used in the co-deposition experiments. These triple tracks have been observed on both the front and backsides of the detectors. Figure 2 compares DT fusion generated triple tracks, with the palladium/deuteride generated triple tracks. It can be seen that the features of the palladium/deuteride generated triple tracks, Fig. 2(a), are indistinguishable from those of DT fusion generated triple tracks, Fig. 2(b).

3. Charged Particles

If DD fusion reactions were occurring inside the palladium lattice as conjectured by Pons and Fleischmann in their March 23, 1989 press conference, these reactions should produce charged particles in addition to neutrons. Techniques used to measure these charged particles include silicon surface barrier detectors, solid state nuclear track detectors such as CR-39, and scintillating detectors.

Shortly after the Fleischmann–Pons announcement, Ed Cecil [15] of the Colorado School of Mines implanted deuterium in a $6 \mu\text{m}$ thick palladium film. When an electric current was passed through the thin film, 5 MeV charged particles were detected using a silicon surface barrier detector. The same experiments using thin deuterated titanium films gave similar results. George Chambers [15] of NRL bombarded titanium foils with 350 eV deuterons and detected 4.9 MeV charged particles. Of 13 experimental runs, four produced charged particles and five did not. In four of the null experiments, the titanium foil delaminated from the base thereby voiding the run. Iida et al. [16] of Osaka University also conducted deuteron implantation experiments on titanium foils. When titanium foils with an aluminum oxide layer were bombarded with 243 eV deuterons, peaks in the 5–12 MeV range were observed. These peaks were attributed to alpha particles resulting from multibody fusion reactions as proposed by Takahashi, also of Osaka University. In 2003, Keeney et al. [17] reported on gas loading experiments on titanium foils. Using a PMT/dual scintillator system, they were able to detect 2.6 MeV protons.

In 1990, Li et al. [18] of Tsinghua University in China were the first to use CR-39 SSNTDs to detect charged particle emission in palladium. In these experiments, palladium foil was in direct contact with the CR-39 detector. The

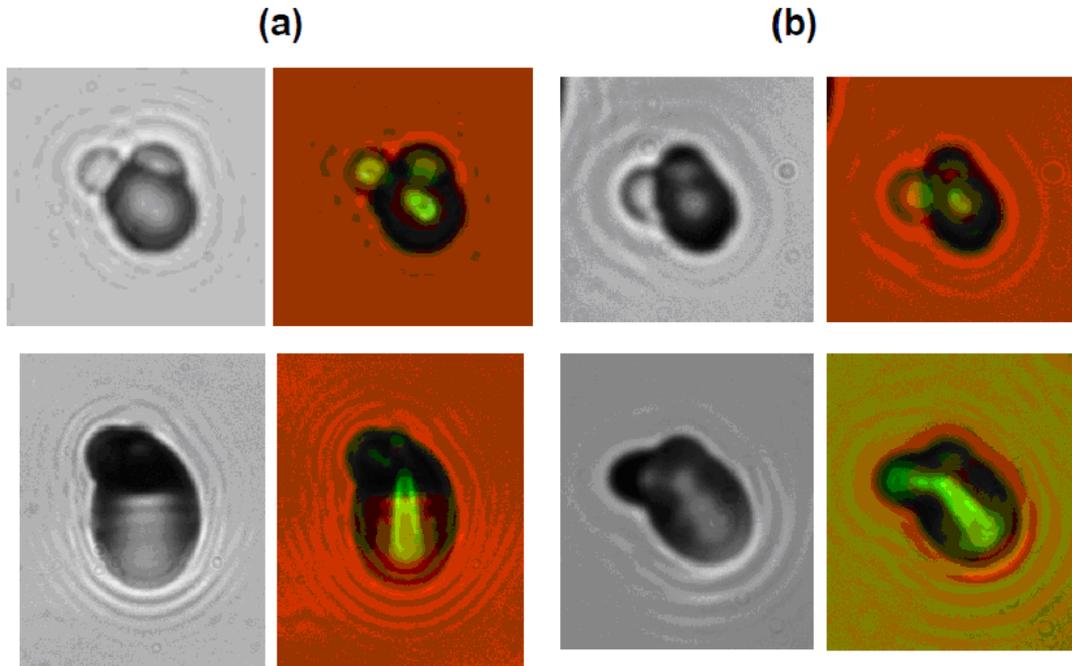


Figure 2. Photomicrographs of (a) Pd/D co-deposition generated and (b) DT neutron generated triple tracks. In (a) and (b), the left-hand images were taken with the microscope optics focused on the surface of the detectors. The right-hand images are an overlay of two images taken at two different focusing depths (surface and bottom of the tracks). Reproduced with permission from Mosier-Boss.

palladium was then gas loaded with either hydrogen or deuterium. No tracks were observed for the hydrogen loading experiments. However, a large number of tracks in the CR-39 were obtained as a result of the palladium/deuteride experiment. It was found that when palladium was cleaned in aqua regia, no tracks were obtained. Li et al. concluded that the aqua regia treatment poisoned the surface of the palladium. From these results Li et al. concluded that the observed nuclear effects were related to some surface phenomenon.

The $Y_1Ba_2Cu_3O_{7-8}$ (YBCO) high temperature super conductor has the ability to absorb hydrogen. Pellets or powders of $Y_1Ba_2Cu_3O_{7-8}$ were placed in contact with CR-39 detectors. The pellets/powders were then gas loaded with deuterium. After a one to two days exposure to the deuterated $Y_1Ba_2Cu_3O_{7-8}$ pellets/powders, the CR-39 detectors were etched. Microscopic examination of the detectors showed tracks with a density of $\sim 3 \times 10^5$ tracks cm^{-2} [19].

As discussed *vide supra*, Lipson et al. [3] used CR-39 detectors to detect charged particles emitted from the deuterated Au/Pd/PdO heterostructures. In these experiments, the heterostructures were electrochemically loaded with deuterium. Once loaded, the heterostructures were placed in contact with the CR-39 detectors and the temperature was cycled to induce desorption of deuterium. Tracks consistent with 2.5–3.0 MeV protons and 0.5–1.5 MeV tritons were observed in the CR-39 detectors.

In 2002, Oriani and Fisher [20] were the first to report the use of CR-39 detectors in an electrolysis experiment. They placed the detectors above and below the Pd foil cathode so as to not impede uniform loading of the cathode with deuterium. Because charged particles cannot travel far in an aqueous medium (10 μm of water slow alpha particles by 2 MeV), this is not the optimum geometry to detect charged particles. During each run, control detectors were immersed in bottled electrolyte solution. Track densities ranged between 59 and 541 tracks cm^{-2} for the control detectors and

156–3760 tracks cm^{-2} for detectors used in active cells. They concluded that the reactions responsible for the particles causing the tracks did not occur at the distant cathode but most likely occurred in the electrolyte very close to the plastic surface. Their more recent experiments involving sequential etching of the detectors show additional tracks deeper inside the plastic. The tracks could either be due to recoils from neutrons or from ≥ 10 MeV protons.

Also in 2002, Lipson et al. [21] at the University of Illinois began to conduct in situ electrolysis experiments in which the CR-39 detectors were in direct contact with palladium foils. They observed tracks due to charged particles in the detectors and concluded that the cathode was the source of those particles. Using Cu spacers between the cathode and the detector, they were able to identify the particles as being 11–16 MeV alphas and ~ 1.7 MeV protons.

Mosier-Boss et al. [22,23] have used CR-39 detectors in their palladium/deuterium co-deposition experiments. Track density was highest where the cathode had been in contact with the detector indicating that the source of the tracks is the cathode. The distribution of tracks along the cathode was inhomogeneous indicating that some Pd sites were more active than others. Control experiments showed that the tracks were not due to radioactive contamination of the cell components; nor were they due to impingement of the deuterium gas bubbles on the surface of the detector; nor were they the result of chemical attack by D_2 , O_2 , or Cl_2 gases; nor were they due to the metal dendrites of the palladium deposit piercing into the plastic. Spacer experiments indicated that the charged particles formed had energies on the order of 1–3 MeV.

Karabut et al. [24] of the Scientific Industrial Association LUTCH in Russia have conducted glow discharge experiments using palladium in deuterium gas. They used both silicon surface barrier and CR-39 detectors in these experiments. The silicon surface barrier detectors measured charged particles up to 18 MeV in energy. Tracks were observed in the CR-39 detectors. The measured track density was on the order of 2×10^5 tracks cm^{-2} .

4. Gamma/X-rays

The first evidence of gamma/X-ray emission is shown in Fig. 3(a). At the end of a deuterium plasma loading experiment, Srinivasan [25] of BARC placed the titanium rod on a sheet of Polaroid paper. Figure 3(a) is the positive image of the Polaroid negative. Dark areas are regions of greater activity. Srinivasan attributed the observed image to be due to X-rays from tritium generated in the titanium rod. This same electrode was repeatedly autoradiographed over a one-year period, revealing the same pattern.

Knowing of the BARC results, McKubre [26] of SRI took a palladium cathode that had produced excess heat and placed it between two layers of Polaroid film for 12 days. The negative image is shown in Fig. 3(b). Fogging of the film was observed indicative of some type of ionizing radiation. The points of light with diffuse halo exposure suggest that some of the radiation may be coming from point sources within the metal and being scattered by the lattice structure.

At the end of a palladium/deuterium co-deposition experiments on a silver disk, Szpak [22] placed a plastic sheet over the cathode and a piece of photographic film on top of that. After a week, the film was developed and the positive image is shown in Fig. 3c. The one aspect that all three images have in common is that the emission of X-rays is inhomogeneous and that some sites are more active than others. Measurements of co-deposition experiments using high purity germanium (HPGe) and lithium-doped silicon detectors showed that the emission of radiation was sporadic and of limited duration [27]. They also showed that the energy distribution was broad with the occasional emergence of peaks near 20 and 8–12 keV.

Miles [28] of China Lake placed dental film around his calorimeter cells using bulk palladium cathodes. He also observed fogging of the film. The film closest to the cathode showed the greatest exposure. Karabut et al. [24] also reported on the emission of 200 keV gamma rays in their glow discharge experiments.

Violante et al. [29] of ENEA and SRI conducted electrolysis experiments using thin films of copper and/or nickel or palladium. The experiments were monitored using a HPGe detector optimized for the 2–22 keV energy range. The copper/palladium cathodes gave signals significantly above background in the energy range where the palladium X-ray

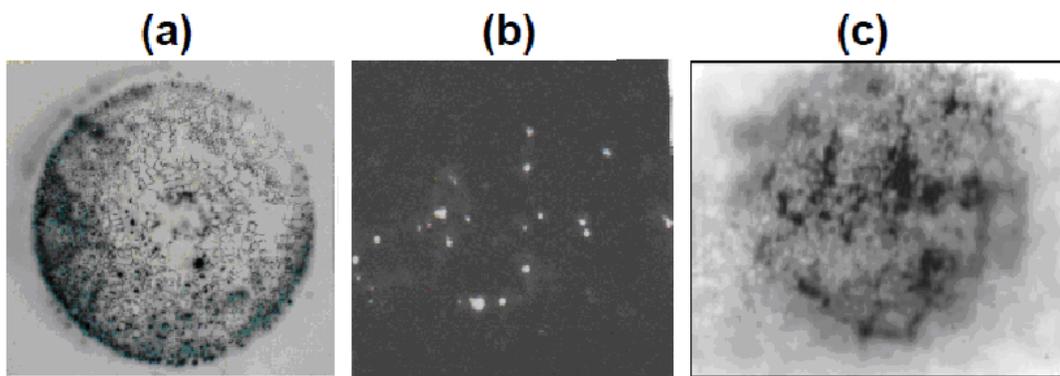


Figure 3. Autoradiographs of cathodes used in LENR experiments. These autoradiographs were obtained by placing photographic film in contact with the cathode over a period of 7–12 days. In the positive images, dark areas show greater activity while the reverse is true for negative images. (a) Positive image of a titanium rod that has been plasma loaded with deuterium. Reproduced with permission from Srinivasan. (b) Negative image of a bulk palladium cathode that had produced excess heat for SRI. Reproduced with permission from McKubre. (c) Positive image of a cathode used in a palladium/deuterium co-deposition experiment. A 100 μm thick plastic sheet separated the cathode from the photographic film. Reproduced with permission from Mosier-Boss.

lines occur. For the copper/nickel cathodes, X-ray emissions and a change in the copper-63/copper-65 isotopic ratios were observed after the deuterium-loaded cathodes had been irradiated with a HeNe laser for 3 h.

Prelas et al. [30] of the University of Missouri used a Maxwellian plasma and sub-atmospheric deuterium gas to load palladium metal. The experiments were monitored using both a BF_3 neutron detector as well as a sodium iodide (NaI) gamma-ray detector. In these experiments, it was observed that an increase in the neutron counts and gamma-ray readings occurred whenever the deuterium plasma was formed. The NaI detector showed a mysterious, broad gamma-ray peak at 8.1 MeV.

5. Transmutation

Kevin Wolf [31] of Texas A&M State College was electrolyzing three palladium cathodes simultaneously. The experiment was monitored using both a neutron and a gamma ray detector. During the electrolysis, the neutron counts increased from a background of 24 counts/h up to 150 counts/h, then leveled off to 100 counts/h over a 21 h period. During that time interval, the gamma ray counts stayed at background levels. After 21 h, the neutron counts dropped back to background, but the gamma ray counts increased. Upon termination of the experiment, it was found that all three cathodes had become mildly radioactive. Gamma ray spectra of the cathodes obtained using a germanium detector showed the presence of isotopes of rhodium, silver, and ruthenium that had not been present prior to the experiment.

John Bockris [32] analyzed his bulk palladium cathodes before and after his electrolysis experiments. The analysis of the cathode was done at 1 μm in depth. He observed the production of magnesium, silver, silicon, calcium, titanium, iron, copper, zinc, and platinum. Similar results were reported by Mizuno [33]. In addition to the elements Bockris had observed, Mizuno also detected chromium. He also showed that the new elements accumulated in holes and cracks on the bulk palladium cathode which formed during electrolysis. Also some of the new elements had isotopic distributions different from the natural ones, with the largest deviations found within the outermost 1 μm layer of the cathode. Using thin palladium films, Miley et al. [34] of the University of Illinois observed significant shifts in isotope ratios from natural abundance of a number of elements.

John Dash [35] of Portland State University conducted electrolysis experiments using titanium cathodes. Excess

heat was produced at the rate of about 1.2 W during electrolysis of heavy water. Analysis of the electrodes before and after electrolysis with a scanning electron microscope (SEM) and an energy dispersive spectrometer (EDS) revealed that new surface topographical features with concentrations of unexpected elements (V, Cr, Fe, Ni, and Zn) formed during electrolysis. He also did palladium/deuterium co-deposition experiments on bulk palladium foils. These experiments showed that co-deposition enhanced the production of excess enthalpy. SEM-EDS analysis of the cathodes showed the presence of silver.

Szpak et al. [36] performed palladium/deuterium co-deposition experiments on gold foil in the presence of either an external electric or magnetic field. Upon termination of the experiments, the cathodes were subjected to SEM analysis. Fractal structures, craters, and micro-volcano like features were observed in the SEMs. These features are suggestive of localized melting of the palladium deposit. Elemental analysis of these features using EDS showed the presence of new elements such as silicon, aluminum, calcium, magnesium, iron, zinc, chromium, and nickel. The distribution of these new elements on the surface of the cathodes was not uniform indicating that their presence could not be the result of contamination.

A consortium of Italian scientists did hydrogen and deuterium gas loading of palladium samples [37–39]. Once loaded, these samples were irradiated with either a HeNe or a 308 nm excimer laser. SEM analysis of the palladium showed the formation of small craters and caverns. Associated with these craters were the following new elements: aluminum, silicon, sulfur, chlorine, iron, cadmium, platinum, chromium, magnesium, titanium, and zinc.

Glow discharge experiments done by Karabut [40] and others also yielded new elements. The new elements obtained are the same as those obtained in the electrolysis experiments described above.

Iwamura et al. [41] observed transmutation that was induced by deuterium gas permeation through palladium complexes. A schematic of their experimental set-up is shown in Fig. 4(a). Figure 4(b) shows a schematic and an SEM of one of their palladium complexes. When the source metal is cesium, in situ X-ray photoelectron spectroscopy (XPS) measurements showed a decrease in the cesium peaks with a simultaneous emergence of peaks due to praseodymium as deuterium permeated through the sample. This was confirmed by ex situ X-ray fluorescence (XRF) measurements of the samples. When the source metal was strontium, gas permeation studies showed a decrease in strontium with a simultaneous increase in molybdenum. Secondary ion mass spectrometry (SIMS) analysis showed significant changes in the isotopic ratios of the molybdenum, as shown in Fig. 4(c).

6. Helium-4

In 1991, Miles et al. [42] showed that helium-4 production temporally correlated with the excess heat production. In these experiments, the gases evolved during electrolysis were collected and were sent away for analysis using a high resolution mass spectrometer. Helium-4 was observed in those samples collected when the cell was giving off excess heat.

In 1991, Bockris et al. [43] terminated an electrolysis experiment in which the palladium cathode was producing tritium. The cathode was removed from the cell and immersed in liquid nitrogen for a week to prevent helium from permeating out of the cathode. The cathode was then cut up into smaller pieces, packed in dry ice, and sent away for analysis by high resolution mass spectrometry. To determine the helium-4 content, the sample was melted in a resistance-heated tungsten-wire crucible in the mass spectrometer's high temperature vacuum furnace. Excess helium-4 was observed in nine out of the ten pieces of this tritium-producing cathode. The amount of helium-4 in these nine pieces varied between 1.7 and 166.8 billion helium-4 atoms. Background samples contain 0.5 billion helium-4 atoms. The majority of the helium-4 was found in the near-surface regions of the cathode and not within the bulk. No helium-4 was observed above background in the non-electrolyzed palladium from the same virgin stock or in that platinum anode material. Because earlier helium-4 claims suffered from the criticism of possible permeation through the glass cells, this was the first substantiated report of the production of helium-4.

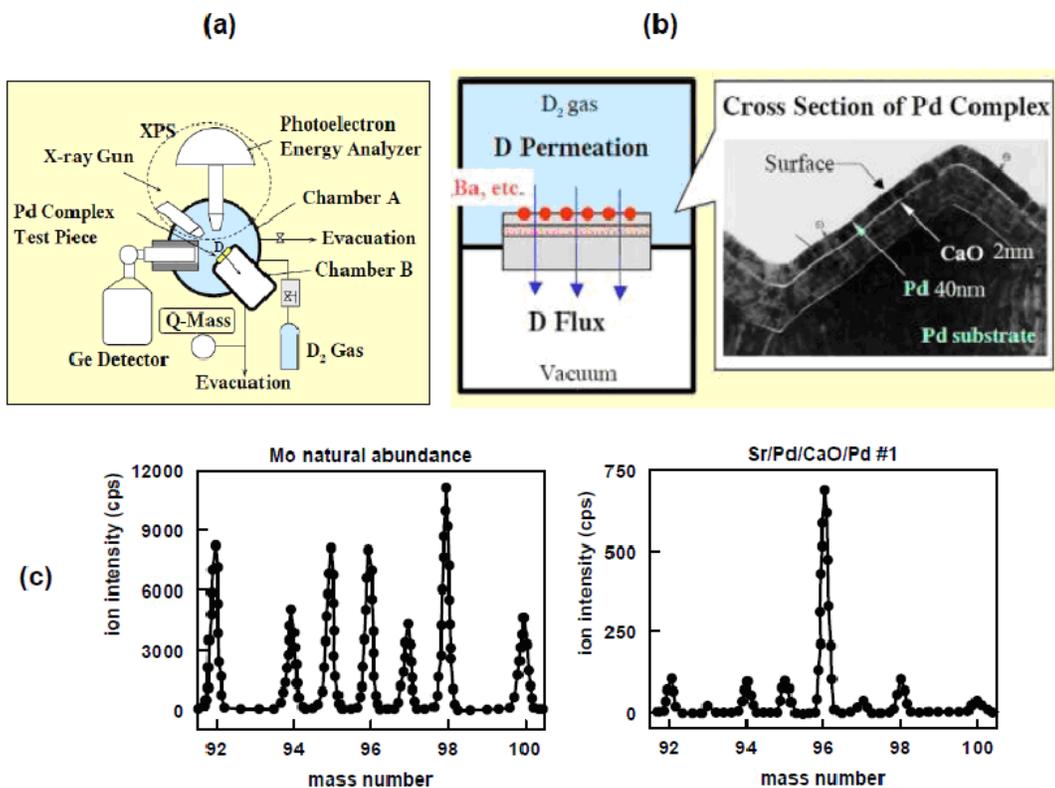


Figure 4. (a) Schematic of the Iwamura deuterium gas permeation experiment. (b) Structure of the palladium complex comprised of M/Pd (400 Å)/CaO and Pd (1000 Å)/Pd (0.1 mm), where M = Cs, Sr, and Ba. (c) Comparison of the natural molybdenum (left) with that observed in the palladium complex (right). Reproduced with permission from Y. Iwamura.

In 1998, McKubre et al. [44] of SRI reported that they had completed a replication of Miles' results. They conducted open cell electrolysis in metal sealed cells incorporating active exclusion of helium-4. The evolving gases were likewise collected in metal vessels and were analyzed by high-resolution mass spectrometry. In three instances when excess power was measured, helium-4 was found in the electrolysis gases. If the helium-4 results from reaction (3), the helium-4 found in the gases constituted only 76% of expectation. However, the results of Bockris et al. [43] showed that helium can be trapped inside the palladium. McKubre et al. [44] concluded that, if helium-4 is produced in association with excess power, it is not released to the gas phase immediately, or completely.

Following the Miles' replication, McKubre et al. [44] conducted electrolysis experiments in a closed cell system using bulk palladium. The cell was leak tight and was designed to allow sampling of the gases in the headspace. During a period of excess power, the gas phase contained only 62% of the helium-4 expected if reaction (3) was the source of the helium-4. The remainder of the helium-4 was retained in the cathode as was determined, in situ, by subjecting the cathode to an extended period (~200 h) of compositional and thermal cycling by varying the current density in both anodic and cathodic directions. The high anodic currents used in this process increased the temperature of the palladium electrode by ~30°C and caused microcracks to form. Both the temperature increase and anodic current drove deuterium out of the lattice thereby quenching the nuclear processes occurring inside. The microcracks formed

in the cathode provided a means for helium-4 to escape. This was the first quantitative correlation between heat and helium-4.

De Ninno et al. [45] of ENEA in Italy sputtered palladium on an inert surface, wound in a ‘bustrophedic’ geometry. The resultant cathode is 100 cm long, 50 μm wide, and 2 μm thick. The cathode was loaded with deuterium electrolytically. The gases evolved during the experiment were collected in a storage vessel. A 6.29% aliquot of this gas was periodically sent to a high resolution mass spectrometer for analysis. Excess heat was measured using a Peltier element that was in good thermal contact with the cathode. Whenever the cathode temperature increased, helium-4 in the gases was measured. As the amount of excess power increased, localized melting of the palladium cathode occurred which caused the experiments to terminate. Because of the extended damage in many separated subsections, it is highly unlikely that the melting of palladium was due to the Joule effect.

In 1998, Gozzi et al. [46] built a calorimetric system, with a recombiner, that allowed online sampling of the residual gases for detection of helium-4 using a high resolution mass spectrometer. Measurements of helium-4 were not performed in a continuous mode. Consequently, a significant volume of the electrolysis gas mixture is lost without being analyzed. Despite that, they noticed that helium-4 in the gas stream temporally correlated with the excess heat. However, the amount of helium was less than the amount predicted if it was produced by reaction (3). At the end of the experiment, the cathode was cut up into smaller segments. Each segment was heated to melting while under high-vacuum pumping. The evolved gases were then analyzed, using the mass spectrometer, for helium-4. Unlike Bockris [43] and McKubre et al. [44], Gozzi et al. [46] did not find any helium-4 inside the palladium cathode. Although helium-4 diffuses slowly through palladium, the presence of micro-cracks in the palladium would cause helium-4 to be released more readily into the atmosphere. It should be noted that, unlike Bockris et al. [43], Gozzi et al. [46] did not immerse the cathode in liquid nitrogen prior to analysis which would have prevented the release of helium-4 through micro-cracks.

In 1998, Lester Case [47] of Fusion Power Inc. had claimed to see excess heat and helium-4 when palladium on a carbon supported catalyst was gas loaded with deuterium. McKubre et al. [44] conducted a series of experiments to verify the Case results. In these experiments, the catalyst was placed in a stainless steel vessel that was connected to a steel manifold. Periodic measurements of helium-4 were made by direct connection to a high resolution mass spectrometer. Hydrogen gas loading showed no excess heat or helium-4. In contrast, both heat and helium-4 were observed to correlate with deuterium gas loading.

Arata and Zhang [48] of Osaka University had reported on obtaining large quantities of excess heat using a ‘double structured’ cathode comprised of a palladium tube filled with palladium black. McKubre et al. [44] replicated the heat results of Arata and Zhang. Post analysis of the cathodes showed the presence of both helium-4 and helium-3.

In 2008, Arata and Zhang [49] reported on heat and helium-4 production as a result of deuterium gas loading of palladium/zirconium oxide nanopowders. No heat or helium-4 were observed in the hydrogen loading experiments. The heat effects have been replicated by Kitamura et al. [50] of Kobe University.

7. Conclusions

Since the announcement by Pons and Fleischmann in 1989, researchers worldwide have conducted experiments to detect nuclear ash to show that nuclear processes are occurring inside the metal lattice. Methods to load hydriding materials, such as Pd, Ti, and YBCO, with deuterium included electrolysis, gas loading, and glow discharge. Both real-time nuclear diagnostics and constantly integrating detectors have been used in these experiments. Regardless of the method or the analytical technique used, the following nuclear emissions have been observed: gamma and X-rays, energetic charged particles and neutrons, and transmutation. These nuclear emissions are consistent with those observed for primary and secondary fusion reactions. Taking all of the data together, there is compelling evidence that nuclear reactions can and do occur inside a metal lattice once the appropriate conditions to initiate those reactions have been

achieved.

References

- [1] M. Krishnan, S. Malhotra, D. Gaonkar, M. Srinivasen, S. Sikka, A. Shyam, V. Chitra, T. Iyengar and P. Iyengar, Cold fusion experiments using a commercial Pd–Ni electrolyser, *BARC Studies in Cold Fusion*, P. Iyengar and M. Srinivasen (Eds.), Bhabha Atomic Research Centre, Trombay, Bombay, India, 1989, A1, pp. 1–10.
- [2] Y. Iwamura, T. Itoh and I. Toyoda, Observations of anomalous nuclear effects in D₂–Pd system, *Proc.: Fourth Int. Conf. on Cold Fusion*, Vol. 3, *Nuclear Measurements Papers*, Lahaina, Maui, Hawaii. Dec. 6–9, 1993, T.O. Passell (Ed.), Electric Power Research Institute, Palo Alto, CA, 1994, pp. 12.1–12.9.
- [3] A. Lipson, F. Lyakhov, A. Roussetski, T. Akimoto, N. Asami, R. Shimada, S. Miyashita and A. Takahashi, Evidence for low-intensity D–D reaction as a result of exothermic deuterium desorption from Au/Pd/PdO:D heterostructure, *Fusion Sci. Technol.* **38** (2000) 238–252.
- [4] E. Storms and C.L. Talcott, Electrolytic tritium production, *Fusion Technol.* **17** (1990) 680–695.
- [5] J. Bockris, C.-C. Chien, D. Hodko and Z. Minevski, Tritium and helium production in palladium electrodes and the fugacity of deuterium therein, *Frontiers Sci. Series No. 4, Proc. of the Third Int. Conf. on Cold Fusion*, Nagoya, Japan, October 21–25, Nagoya Japan, H. Ikegami (Ed.), Universal Academy Press, Tokyo, Japan, 1993, p. 231.
- [6] S. Szpak, P. Mosier-Boss, R. Boss and J. Smith, On the behavior of the Pd/D system: evidence for tritium production, *Fusion Sci. Technol.*, **33** (1998) 38–51.
- [7] T. Claytor, J. Jackson and D. Tuggle, Tritium production from a low-voltage deuterium discharge on palladium and other metals, *J. New Energy* **1**(1) (1996) 111–118.
- [8] T. Mizuno, T. Akimoto, T. Ohmori, A. Takahashi, H. Yamada and H. Numata, Neutron evolution from a palladium electrode by alternate absorption treatment of deuterium and hydrogen, *Jpn. J. Appl. Phys.* **40** (2001) L989–L991.
- [9] A. De Ninno, A. Frattolillo, G. Lollobattista, L. Martinis, M. Martone, L. Mori, S. Padua and F. Scaramuzzi, Emission of neutrons as a consequence of titanium–deuterium interaction, *Il Nuovo Cimento, note brevi* **101** (1989) 841.
- [10] A. De Ninno, A. Frattolillo, G. Lollobattista, L. Martinis, M. Martone, L. Mori, S. Padua and F. Scaramuzzi, Evidence of emission of neutrons from a titanium–deuterium system, *Europhys. Lett.* **9** (1989) 221–224.
- [11] A.S. Roussetski, Application of CR-39 plastic track detector for detection of dd and DT-reaction products in cold fusion experiments, *Proc. 8th Int. Conf. on Cold Fusion*, May 21–26, 2000, Lerici (La Spezia), Italy, Italian Physical Society, Bologna, Italy.
- [12] P. Mosier-Boss, S. Szpak, F. Gordon and L. Forsley, Triple tracks in CR-39 as the result of Pd–D co-deposition: evidence of energetic neutrons, *Naturwissenschaften* **96** (2009) 135–142.
- [13] P. Mosier-Boss, S. Szpak, F. Gordon and L. Forsley, Detection of energetic particles and neutrons emitted during pd/d co-deposition, *Low-Energy Nuclear Reactions Sourcebook*, J. Marwan and S. Krivit (Eds.), American Chemical Society, Washington, DC, 2008, p. 311.
- [14] F. Keeney, S. Jones, A. Johnson, P. Hagelstein, G. Hubler, D. Buehler, F. Cecil, M. Scott and J. Ellsworth, Neutron emissions from metal deuterides, *Condensed Matter Nucl. Sci.: Proc Tenth Int. Conf. on Cold Fusion*, Aug. 24–29, 2003, Cambridge, MA, P. Hagelstein and S. Chubb (Eds.), World Scientific, New Jersey, 2006, p. 525.
- [15] C. Beaudette, *Excess Heat: Why Cold Fusion Research Prevailed*, Oak Grove Press, LLC, South Bristol, Maine, 2002, 280–282.
- [16] T. Iida, M. Fukuhara, Sunarno, H. Miyamaru and A. Takahashi, Deuteron fusion experiments with Ti and Pd foils implanted with deuteron beams II, *Proc. Fourth Int. Conf. on Cold Fusion*, Vol. 3, *Nuclear Measurements Papers*, Lahaina, Maui, Hawaii, Dec. 6–9, 1993, T.O. Passell (Ed.), Electric Power Research Institute, Palo Alto, CA, 1994, pp. 13.1–13.8.
- [17] F. Keeney, S. Jones, A. Johnson, P. Hagelstein, G. Hubler, D. Buehler, F. Cecil, M. Scott, J. Ellsworth, Charged-particle emissions from deuterided metals, *Condensed Matter Nucl. Sci., Proc. Tenth Int. Conf. on Cold Fusion*, Aug. 24–29, 2003, Cambridge, MA, P. Hagelstein and S. Chubb (Eds.), World Scientific, New Jersey, 2006, p. 509.
- [18] X. Li, S. Dong, K. Wang, Y. Fang, L. Chang, C. Luo, R. Hu, P. Zhou, D. Mo, Y. Zhu, C. Song, Y. Chen, M. Yao, C. Ren and Q. Chen, The precursor of “cold fusion” phenomenon in deuterium/solid systems. Anomalous nuclear effects in deuterium/solid systems, “AIP Conference Proc. 228”, 1990, Brigham Young Univ., Provo, UT, American Institute of Physics, New York.

- [19] S. Jin, F. Zhan and Y. Liu, Deuterium absorbability and anomalous nuclear effect of YBCO high temperature superconductor, *Proc. Fourth Int. Conf. on Cold Fusion*, Vol. 3, *Nuclear Measurements Papers*, Lahaina, Maui, Hawaii, Dec. 6–9, 1993, T.O. Passell (Ed.), Electric Power Research Institute, Palo Alto, CA, 1994, pp. 4.1–4.3.
- [20] R.A. Oriani and J.C. Fisher, Generation of nuclear tracks during electrolysis, *Jpn. J. Appl. Phys. A* **41** (2002) 6180–6183.
- [21] A. Lipson, G. Miley, A. Roussetski, E. Saunin, Phenomenon of an energetic charged particle emission from hydrogen/deuterium loaded metals, *Condensed Matter Nucl. Sci., Proc. Tenth Int. Conf. on Cold Fusion*, Aug. 24–29, 2003, Cambridge, MA, P. Hagelstein and S. Chubb (Eds.), World Scientific, New Jersey, 2006, p. 539.
- [22] P. Mosier-Boss, S. Szpak, F. Gordon and L. Forsley, Use of CR-39 in Pd/D Co-deposition experiments, *Eur. Phys. J. Appl. Phys.* **40** (2007) 293–303.
- [23] P. Mosier-Boss, S. Szpak, F. Gordon and L. Forsley, Characterization of tracks in CR-39 detectors obtained as a result of Pd/D co-deposition, *Eur. Phys. J. Appl. Phys.* **46** (2009) 30901: 1–12.
- [24] A.B. Karabut, Y.R. Kucherov and I.B. Savvatimova, Nuclear product ratio for glow discharge in deuterium, *Phys. Lett. A* **170** (1992) 265.
- [25] M. Srinivasen, Personal communication.
- [26] M. McKubre, *Development of Advanced Concepts for Nuclear Processes in Deuterated Metals*, TR-104195, 1994, Electric Power Research Institute.
- [27] S. Szpak, P. Mosier-Boss and J. Smith, On the behavior of the cathodically polarized Pd/D system: search for emanating radiation, *Phys. Lett. A* **210** (1996) 382–390.
- [28] M. Miles, R. Hollins, B. Bush, J. Lagowski and R. Miles, Correlation of excess power and helium production during D₂O and H₂O electrolysis using palladium cathodes, *J. Electroanal. Chem.* **346** (1993) 99.
- [29] V. Violante, G. Mazzitelli, L. Capobianco, M. McKubre, F. Tanzella, F. Sarto and E. Santoro, Search for nuclear ashes in electrochemical experiments, *Condensed Matter Nucl. Sci., Proc. of the Tenth Int. Conf. on Cold Fusion*, Aug. 24–29, 2003, Cambridge, MA, P. Hagelstein and S. Chubb (Eds.), World Scientific, New Jersey, 2006, 405.
- [30] M. Prelas, F. Boody, W. Gallaher, E. Leal-Quiros, D. Mencin and S. Taylor, Cold fusion experiments using Maxwellian plasmas and sub-atmospheric deuterium gas, *J. Fusion Energy* **9** (1990) 309–313.
- [31] C. Beaudette, *Excess Heat: Why Cold Fusion Research Prevailed*, Oak Grove Press, LLC, South Bristol, Maine, 2002, pp. 264–265.
- [32] J. Bockris and Z. Minevski, Two Zones of “Impurities” observed after prolonged electrolysis of deuterium on palladium, *Infinite Energy Magazine* Nos. 5 and 6 (1995) 67.
- [33] T. Mizuno, T. Ohmori and M. Enyo, Isotopic changes of the reaction products induced by cathodic electrolysis in Pd, *J. New Energy* **1**(3) (1996) 31–47.
- [34] G.H. Miley and J. Shrestha, *Transmutation Reactions and Associated LENR Effects in Solids*, in *Low-Energy Nuclear Reactions Sourcebook*, J. Marwan, and S. Krivit (Eds.), Oxford University Press, 2008, p. 173.
- [35] J. Dash and S. Miguet, Microanalysis of Pd cathodes after electrolysis in aqueous acids. *J. New Energy* **1**(1) (1996) 23–27.
- [36] S. Szpak, P. Mosier-Boss, C. Young and F. Gordon, Evidence of nuclear reactions in the Pd lattice, *Naturwissenschaften* **92** (2005) 394–397.
- [37] A. Lorusso and V. Nassisi, Pd systems loaded with D₂ and H₂ gases and irradiated by laser beams, *Cold Fusion: The History of Research in Italy*, S. Martellucci, A. Rosati, F. Scaramuzzi, V. Violante (Eds.), ENEA, 2009, p. 155.
- [38] A. Battaglia, L. Daddi, S. Focardi, V. Gabbani, V. Montalbano, F. Piantelli, P.G. Sona and S. Nesi, Neutron emission in Ni–H systems, *Nuovo Cimento* **112A** (1999) 921–931.
- [39] E. Campari, S. Focardi, V. Gabbani, V. Montalbano, F. Piantelli and S. Veronesi, Overview of H–Ni systems: old experiments and new setup, *5th Asti Workshop on Anomalies in Hydrogen- /Deuterium-Loaded Metals*, Asti, Italy, 2004.
- [40] A. Karabut, Production of excess heat, impurity elements, and unnatural isotopic ratios in high-current glow discharge experiments, *Condensed Matter Nucl. Sci., Proc. Tenth Int. Conf. on Cold Fusion*, Aug. 24–29, 2003, Cambridge, MA, P. Hagelstein and S. Chubb (Eds.), World Scientific, New Jersey, 2006, p. 99.
- [41] Y. Iwamura, M. Sakano, T. Itoh, Elemental analysis of Pd complexes: effects of D₂ gas permeation, *Jpn. J. Appl. Phys.* **41** (2002) 4642–4650.
- [42] B.F. Bush, J.J. Lagowski, M.H. Miles and G.S. Ostrom, Helium production during the electrolysis of D₂O in cold fusion experiments, *J. Electroanal. Chem.* **304** (1991) 271–278.

- [43] C.-C. Chien, D. Hodko, Z. Minevski, J.O'M. Bockris, On an electrode producing massive quantities of tritium and helium, *J. Electroanal. Chem.* **328** (1992) 189–212.
- [44] M. McKubre, F. Tanzella, P. Tripodi and P. Hagelstein, The emergence of a coherent evaluation for anomalies observed in D/Pd and H/Pd systems: evidence for ^4He and ^3He production, *Proc. 8th Int. Conf. on Cold Fusion*, May 21–26, 2000, Lerici (La Spezia), Italy, Italian Physical Society, Bologna, Italy.
- [45] A. De Ninno, A. Frattolillo, A. Rizzo, E. Del Giudice and G. Preparata, *Experimental Evidence of ^4He Production in a Cold Fusion Experiment*, ENEA, 2002.
- [46] D. Gozzi, F. Cellucci, P.L. Gignini, G. Gigli, M. Tomellini, E. Cisbani, S. Frullani and G.M. Urciuloi, X-ray heat excess and ^4He in the D/Pd system, *J. Electroanal. Chem.* **452** (1998) 251–271.
- [47] C. Beaudette, *Excess Heat: Why Cold Fusion Research Prevailed*, Oak Grove Press, LLC, South Bristol, Maine, 2002, pp. 239–241.
- [48] Y. Arata and Y. Zhang, Solid-state plasma fusion (“cold fusion”), *J. High Temp. Soc.* **23** (1997) 1–56.
- [49] Y. Arata and Y. Zhang, The establishment of solid nuclear fusion reactor, *J. High Temp. Soc.* **34** (2008) 85–93.
- [50] A. Kitamura, T. Nohmi, Y. Sasaki, A. Taniike, A. Takahashi, R. Seto and Y. Fujita, Anomalous effects in charging of Pd powders with high density hydrogen isotopes, *Phys. Lett. A*, **273** (2009) 3109–3112.