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Research Article

What is needed in LENR/FPE studies?

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Abstract

ICCF16 marks nearly 22 years of research into the phenomenon first called "cold fusion". This new field has expanded in breadth to the point that numerous acronyms compete to describe overlapping effects. Two of these are: LENR (Low Energy Nuclear Reactions) and its subset FPE (the Fleischmann Pons Effect). Research to elucidate the basic processes and shed light on mechanisms has proceeded effectively continuously since March 1989 at SRI and elsewhere, and diligently and as "time and funds" have allowed at numerous other accomplished institutions worldwide. It is now clear to a well-informed but relatively small group of scientifically interested individuals that the deuterium-palladium systems conceals a heat source with energy exceeding by several orders of magnitude mechanical, lattice storage or chemical energy effects. Evidences of possible dd fusion products have been widely and numerously observed, at least partially ratifying the original speculative designation of "cold fusion". The effect, however, is apparently not limited to deuterium as a fuel, to helium as a product or to palladium as a matrix, hence the classification of the field as Condensed Matter Nuclear Science (CMNS) to differentiate these "new" effects from those observed in the rarified environments of particle and plasma physics. The materials-related issues associated with the so-called irreproducibility of CMNS effects presents the greatest barrier to the advancement and acceptance of the field. Precisely because we are dealing with condensed matter, micro-structural and micro-impurity control is difficult and expensive, requiring specialized equipment, control and skill. Problems of similar scale (and origin) have been faced and overcome in the past in the development of technologies having similar significance for mankind. Two (of many) examples are: the development of solid-state semiconductors; implementation of conventional nuclear power (and weapons). Absent another Bell Labs or Manhattan Project how should the CMNS field best proceed? Several overlapping options will be discussed. It is well within the capability of "mainstream" scientists and engineers in academia, and in national and private laboratories to resolve the materials issues and the questions of whether or not specific products are or are not present. Without funding they will not be encouraged to do so; without (a higher degree of) acceptance, adequate funding will not be made available. This paper will address various options and strategies to surmount this logical dilemma.

Keywords: Calorimetry, Cold fusion, Deuterium, Electrochemistry, Excess heat, Helium, Palladium

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1. Introduction

The broadening of definition from “cold fusion” to “condensed matter nuclear science” reflects a marked expansion of avenues of research, interest and opportunity. This ICCF conference series has heavily focused on deuterium/palladium studies with a large emphasis on electrochemical studies as originally taught by Fleischmann and Pons [1]. This paper reflects that emphasis as it relies on experimental evidence generated at SRI in the years 1989–2011, almost exclusively in Pd/D studies of both electrochemical and gas loading systems. At the outset, however, it is worth noting that the work of Piantelli, Focardi and Rossi [2,3] in nickel/natural hydrogen, gas-loading systems has recently risen to interesting prominence. If demonstrated either by commercial application or by scientific acclaim as the result of publication, this “light” branch offers substantial advantage over more expensive routes employing palladium and deuterium, particularly if these alternate paths avoid electrochemical processes that are more challenging and potentially hazardous.

The process of selecting effective pathways forward remains the same whichever the route (or whatever, since others are imaginable). We are concerned with answers to the following questions:

1. What is the present state of knowledge?
    - What do we think we know?
    - Why do we think we know it?
2. How did we arrive at this position?
3. Why do doubts remain in the broader scientific community?
4. How do we propose to make progress?
5. What are the effective ways to proceed?

Salient questions on experimental procedure and observation emerged in 1989 and 1990 that were addressed in the experiments that followed. Here we pose some of the more relevant assertion and briefly address answers to them.

1.1. The experiments cannot be reproduced.

With the advantage of hindsight, Questions 1 and 2 above can be addressed effectively by answering Question 3. After 15 International Conferences in the ICCF series alone, thousands of published papers detailing nuclear products and heat effects consistent with nuclear but not chemical energies, why does doubt persist regarding the existence of condensed matter nuclear effects? What is the basis for this confusion?

We have previously addressed the issue and importance of replication in CMNS studies [4] and what Martin Fleischmann has referred to as “pathological criticism” [5]. However, the importance of “replication” and “reproducibility” (or its lack) in the criticisms of “cold fusion” cannot be overstated. First, it is worth noting that the key test in science is consistency, not identicality, but where do claims that CMNS experiments and results are not reproducible come from? Were these claims ever sensibly and scientifically correct? Are they correct now?

A large part of the apocrypha of irreproducibility was established by two highly influential early papers, authored by major institutions, Caltech [6] and MIT [7]. At the time in late 1989 and early 1990 when these papers were published, the importance and relative difficulty of loading deuterium (D, rather than protium, H) cathodically into palladium was recognized as an issue but had not been addressed fully. In the first Annual Conference on Cold Fusion [1] in early 1990 the SRI group made the comment that *“none of the “cold fusion” electrolysis experiments described to date contain any means of determining the D/Pd content in situ. Yet this ratio may be a crucial difference between those experiments that have produced a Fleischmann–Pons effect and those that have not”*. By making systematic study of D/Pd loading in situ, and its effect on Fleischmann–Pons excess heat product, it subsequently became clear that this was a threshold phenomenon [2], with a failure to load being a sufficient reason for the “negative” (more correctly null) results reported from Caltech [6] and MIT [7].

The experimental program at SRI was designed to pursue the hypothesis that *"an unexpected source of heat can be observed in the D/Pd System when Deuterium is loaded electrochemically into the Palladium Lattice, to a sufficient degree"*. This was the fundamental (and potentially transformational) claim of Fleischmann and Pons [1]. It was also clear from many decades of work with the electrochemical D/Pd system that no anomalous heat was observed in the well-studied region of low loading. Looking a little further ahead to a possible nuclear hypothesis one might also have predicted that the concentration of the most likely reactant (D) could play a role in the magnitude of the effect. It took several years of work at SRI [8–11] and elsewhere [12,13] to develop strong evidence corroborating the claim of Fleischmann and Pons [1] and supporting the hypothesis that deuterium plays a crucial and threshold effect in producing nuclear-level excess heat in the electrochemical deuterium-palladium system.

Figure 1 presents as a histogram an accumulation of positive excess heat results obtained at SRI (Menlo Park, US) and ENEA (Frascati, Italy) versus the maximum D/Pd loading obtained by the cathode. Also plotted are the null results (no measured thermal anomaly) obtained at Caltech [6] and MIT [7], versus the "expected" loading of their calorimetric cathodes. Neither Caltech nor MIT measured loading in situ, and instead compared the loading of calorimetric cathodes to values measured by weighing "similar cathodes" in a similar electrochemical environment [6,7].

With the advantage of hindsight we can make three observations:

1. The D/Pd loading of the population of successful excess heat producing cathodes at SRI and ENEA is clearly different from that of the cathodes unsuccessfully operated at Caltech and MIT.
2. Variability in loading is such that unless loading is poor (i.e. close to the equilibrium value with the 1 atmosphere of D2 bathing the cathode under conditions of vigorous electrolysis where D/Pd ~ 0.7) then measurements of

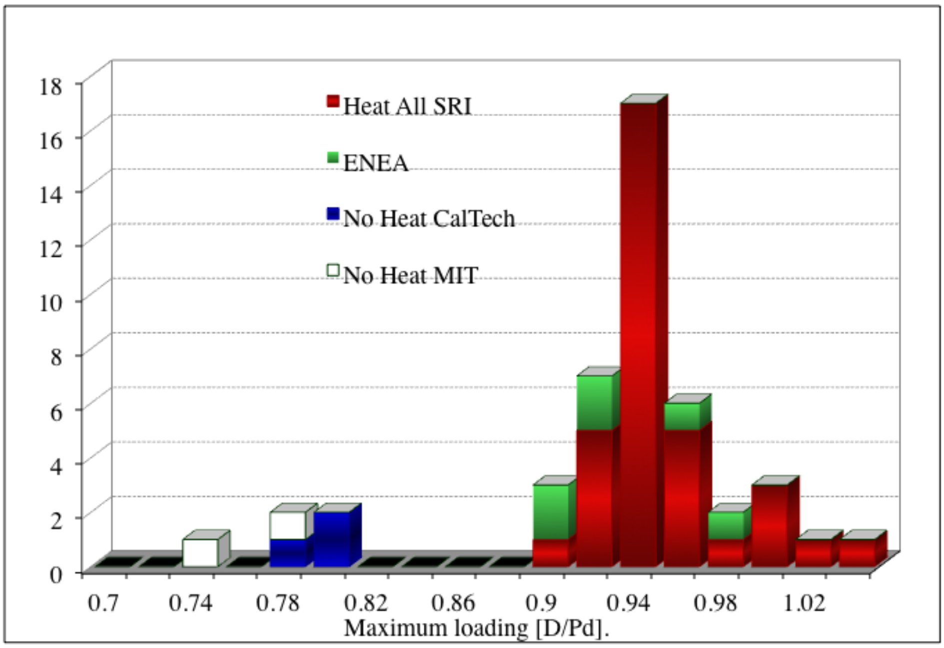

Figure 1. Histogram of excess heat results obtained at SRI, ENEA Frascati, Caltech [6] and MIT [7].

loading made on “fraternal” cathodes are unlikely to be representative.

3. Only with great difficulty, with attention to cathode metallurgical and surface condition, scrupulous avoidance of deleterious impurities and slow and patient electrochemical skill driven primarily by feedback from the measured loading [3] can cathodes be loaded reliably to the minimum value of successful excess heat production shown in Fig. 1.

Obviously these details were unknown in 1989. Although the critical effect of loading might have been anticipated, the only error was over-simplification leading to a rapid rush to negative judgment that was inappropriate and unfortunate in a matter of such import. With a collective sigh of relief (and sometimes sharper tones) a majority in the establishment of physicist and physical chemists returned to what they were doing before 23 March 1989, effectively convinced by the lack of evidential support of the claims of Fleischmann and Pons in the work of Caltech [6] and MIT [7] (and also Harwell in England [14]).

In his definitive study of the Fleischmann Pons heat effect Beaudette [5] draws an excellent distinction between validation and corroboration. Corroboration shows that the initial report was not figmentary, but it does not exclude the possibility of some systematic error in the experiment. That one gets similar results from similar experiments is altogether expected. The argument surrounding the important null results cited above is the implicit claim that these researchers had done similar experiments (electrochemically) but had not obtained similar calorimetric results to Fleischmann and Pons. Leaving aside the (stated) assertion that Fleischmann and Pons were delusional, only two scientific conclusions are possible: either the experiments of Fleischmann and Pons were subject to systematic error, or the two sets of experiments were not similar in one or more detail that was important to the effect.

The Fleischmann–Pons Effect (FPE) has now been validated in numerous laboratories around the world [15,16], in independent experiments with no possibility of systematic overlap with the original experiments. It is also now evident that more than one detail must be attended to before the effect can be observed. One such detail relates to the precise metallurgy of the cathode. Figure 2 plots excess thermal power as a percentage of input electrochemical power for a set of cathodes manufactured at ENEA Frascati from different stock palladium and with subtly varied metallurgical processing [17]. Two sets of experiments were run, all with $\sim 50\,\mu$m thick foil cathodes in similar cells with similar electrochemistry. Calorimetric measurements were made of all of the material lots, either at ENEA, SRI or both; excess power was not observed for those material lots without data points. Two conclusions are clear: materials that did not yield excess power in one laboratory also did not in the other; when excess power was observed in one laboratory it was seen at similar percentage in the other.

The apparent discrepancy for L17 material is worth noting and elaborating, as this touches on another detail of difference between the original Fleischmann and Pons results and those of Caltech and MIT. The FPE has two active Modes referred to by us as “A” and “B”. Mode A has been best studied using bulk wire cathodes and requires high loading ($x^{o} = \mathrm{D/Pd} > \sim 0.9$), long initiation times (100’s of hours at loading), and produces excess power:

$$P_{xs} = M\,(x - x^{\circ})^2\,(i - i^{o})|i_{\mathrm{D}}|, \tag{1}$$

where i is the electrochemical current density and i° the current threshold and $|i_{\mathrm{D}}|$ is the magnitude of the deuterium flux permeating the interface. Although not extensively studied, Mode B excess heat production may have a lower loading threshold, at least when loading is measured as the bulk average value, and appears to be a feature of small dimension electrodes or regions. This mode can appear very early in an experiment and is not strongly or positively influenced by current density. Accordingly Mode B can appear with a large percentage excess power since the denominator (IV) can have small values and still achieve an excess heat effect. The L17 cathodes experienced at SRI showed reproducible excess power in Mode A of 12% and 13%, but 500% excess power at ENEA in Mode B.

Four factors are important in explaining the inability of earlier workers to produce and measure excess heat in electrochemical deuterium–palladium studies:

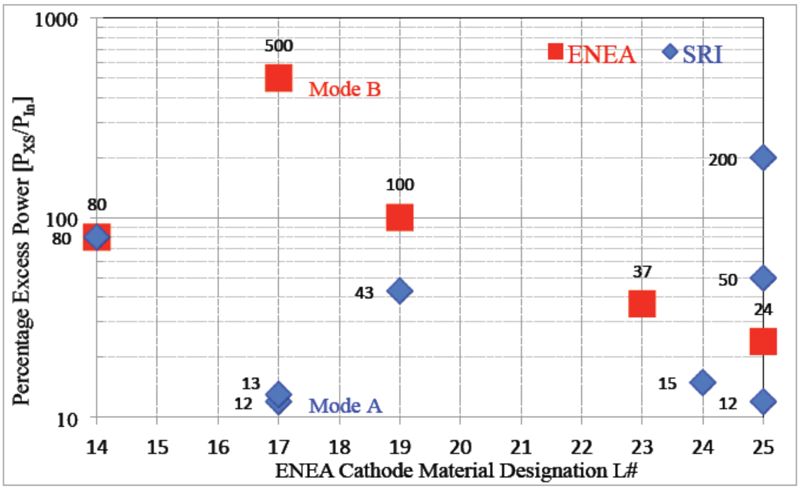

Figure 2. Excess power as a percentage of input power in experiments run at SRI and ENEA, Frascati, for different lots of 50 ìm foil cathodes manufactured at ENEA.

1. As indicated in equation[1] electrodes must be loaded above the loading threshold ($x^{\circ} \sim 0.875$ for Pd wire cathodes). This was very likely not achieved at Caltech and MIT.
2. The current density must be above a significant threshold value also probably not attained in the experiments performed at Caltech and MIT.
3. The condition of the cathode/electrolyte interface must be such that it is facile to deuterium exchange to promote or permit a significant iD flux. The value of this flux is undetermined in the Caltech and MIT experiments because loading was not measured in situ. Nevertheless, because the loading measured in parallel experiments was low it is unlikely that the calorimetric experiments performed at Caltech and MIT achieved a facile surface condition sufficiently free of impurity to promote a high value of i_D.
4. An initiation period orders of magnitude longer than the deuterium diffusional time constant must occur before any excess power is observed in Mode A [4].

This last point is exemplified in Fig. 3 that shows the time of first appearance of excess power for 1–3 mm dia. Pd wire and 50–100 ìm thick Pd foil cathodes for sets of experiments run at SRI and ENEA. For thin foils the earliest appearance of Mode A excess heat was 50 h with one electrode requiring 500 h despite our ability to fully load these cathodes with deuterium in less than 5 h. For the much thicker wires (with cylindrical geometry) Mode A excess heat was first seen at 200 h with one electrode requiring 950 h, although full loading can be achieved in less than 1 day. Clearly some other process is occurring which, if mass transport is involved, has a diffusion coefficient at least 1–2 orders of magnitude less than that of deuterium in the PdD_x lattice.

1.2. The results are inaccurate.

The first place to look when seeking to explain an unpredicted result is error in measurement. For the unexpected results reported by Fleischmann and Pons [1] and for those seeking to verify or refute the FPE, there are two places where such errors might occur. Having first ensured that your experiment faithfully replicates the experimental conditions of the original work (see above) then one must set about measuring accurately the total power that enters the calorimeter, and

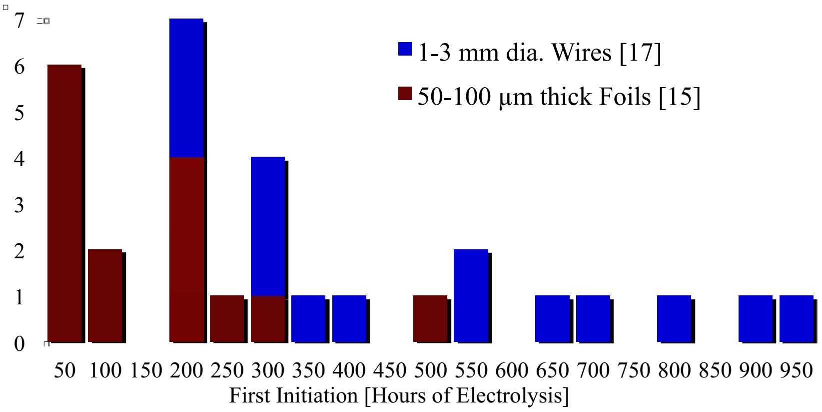

Figure 3. Time of first initiation of excess power for 1–3 mm dia. Pd wire and for 50 ìm Pd foil cathodes in experiments performed at SRI and ENEA, Frascati.

the sum of all heat that results. If more heat is yielded than input then one is measuring excess power, the integral of which is excess energy or heat. If the amount of excess is larger than can be accounted by energy storage or chemical effects then one must look at alternate explanations, potentially nuclear, and seek products (see discussion below).

This sequence and process of scientific discovery occupied perhaps the first 3 or 4 years of the modern "cold fusion" saga. Many hundreds of papers have been published and presented detailing different modes of calorimetry. These have been well reviewed by Storms [15] and constitute an impressive consensus. Using different approaches, different methods to measure input power, different instrumentation, and different calorimetric methadologies, in different laboratories worldwide, a concurrence of agreement has emerged. With attention paid to the (now) known initiating conditions excess power, energy and heat can be measured in the D/Pd system at levels statistically far in excess of the uncertainties associated with the measurement of electrical (and other) input and system thermal output.

1.3. The source of heat is mundane.

The power of the argument and evidence alluded to in the preceding section is essentially overwhelming. This has caused those, more experienced with measurement (and having read the literature), to look for an alternate explanation for the excess heat. It has been proposed that the excess heat is real but is due to unknown or unaccounted chemical effects or lattice energy storage. This argument can be refuted quantitatively. The heat observed as integrated excess power in many (but not all) FPE experiments exceeds the sum of all possible chemical or mechanical energy storage effects by factors of hundreds or thousands, or more.

The partial molar enthalpy of absorption of hydrogen (deuterium) in palladium varies with the state of loading but is always less than 1 eV per H or D atom added to the lattice [18]. Chemical reaction energies can range up to $\sim$3 eV. The largest continuous excess energy production observed at SRI was slightly greater than 2000 eV/Pd atom (or lattice D atom at $\sim$1:1 loading). Even more impressive numbers were reported by Energetics [19] who measured in a single burst lasting $\sim$17 h a net excess energy of 4.8 keV/Pd atom (with input electrical energy $<$200 eV/Pd). The system exhibited a second burst of 15.1 keV/Pd atom after a period of thermal neutrality (zero excess and no endothermic process), for a total of nearly 20 keV/Pd atom [5].

Energy densities at this level are inconsistent with known chemistry and one must look to the nuclear realm for possible explanations. Although "weak" nuclear reactions are known on the scale of keV, more typically these occur with scale ~1, 10 or even 100 MeV/nucleus. Thus the energies measured in FPE experiments are consistent with known nuclear effects if only a small fraction of he lattice participates simultaneously in the process, but are not consistent with known lattice storage or chemical effects even if all of the lattice participates. Clearly the source of FPE heat is not mundane, and may be sufficiently exotic to be of potential practical interest.

1.4. The nuclear products are missing

At this point we can begin to interrogate the nuclear question directly. The effect reported by Fleischmann and Pons was initially called “cold fusion”, a name already in use for muon catalysis of two-body d–d fusion. Whatever Fleischmann and Pons saw it was clearly not that since the anticipated products of the fusion of two deuterons isolated from all other matter [6] are energetic neutrons and radioactive tritium. That these were not seen in quantitative or temporal correlation with the FPE heat production caused some to question the nuclear hypothesis. This was discussed, apparently with complete absence of chemical intuition, as the “problem of” the missing “nuclear ash” [20].

In a series of papers [21–24] Miles and Bush first communicated information regarding the nuclear process at the core of the FPE, in correlated observation of FPE excess heat and ^{4}He. These authors reported results of excess heat and helium gas analyses performed on evolved gas samples taken from thermodynamically open FPE cells, some of which exhibited excess heat and others did not. Miles and Bush were able to show by statistical analysis of 17 samples that when excess heat was present, so was ^{4}He, and when excess heat was not present neither was ^{4}He. The probability of their results occurring by random chance were computed as 1 in 750,000.

Miles and Bush demonstrated not only a statistical correlation between the appearance of FPE excess heat and ^{4}He, but also a near-quantitative correlation with the putative net reaction of two deuterons to form one ^{4}He with the release of ~24 MeV of energy inside the calorimeter [7]. Their result was “near-quantitative” in that the average value of measured helium was only $1.4 \pm 7 \times 10^{11}$ ^{4}He s^{-1} W^{-1}, 54% of “expected” value of 2.5×10^{11} ^{4}He s^{-1} W^{-1}. Working at SRI using a more robust and accurate metal-sealed calorimeter Bush later confirmed this result [25], measuring $1.5 \pm 2 \times 10^{11}$ ^{4}He s^{-1} W^{-1} - 58% of “expected” value.

In attempting to define the reaction “*Q*” value to gain insight into the overall reaction process, it is appropriate to ask “where is the missing ^{4}He” or “is ^{4}He missing”? In 1996 Gozzi reported some very striking results [26] in which bursts of excess energy were time-correlated with bursts of ^{4}He observed in the gas stream [8]. When compared one at a time, the number of helium atoms detected per burst was on the order of what might be expected from 23.8 MeV per ^{4}He measured, but varying between 25 and 100% of this amount. If the energy production in these experiments is in fact due to a net reaction consistent with $D + D \rightarrow {}^4He + 23.8$ MeV, then it seems that some of the helium may enter the gas stream and some remain within the metal. More recent work at ENEA Frascati also supports these conclusions.

In two experiments, one performed at SRI [28] and the other at ENEA [17] efforts were made to scavenge near-surface ^{4}He and make it available in the gas phase for analysis. At SRI the experiment was performed in a helium leak-tight, all-metal and metal gasketed calorimeter. Samples were transferred in metal gas sample flasks to be analyzed for ^{4}He by the U.S. Bureau of Mines at Amarillo, Texas. The initial value of ^{4}He was 0.34 ± 0.007 ppmV/V in the D_2 gas used to charge the cell.

Figure 4 traces the history of the cell, M4, from four helium samples taken after excess power was observed as shown in Fig. 5. The upper solid line in Fig. 4 is the expectation for helium concentration presuming: (i) an initial value of 0.34 ppmV/V, and (ii) that ^{4}He is produced in a reaction that delivers 23.8 MeV of thermal energy to the calorimeter. The first gas sample taken shortly following the second heat burst of Figure 5 yielded a value of 1.556 ± 0.007 ppmV/V ^{4}He, which is about 62% of its expected value, and consistent with the earlier observations by Miles et al. [21–24] and also Gozzi and collaborators [26,27]. A second sample taken about six days after the first showed a measurable

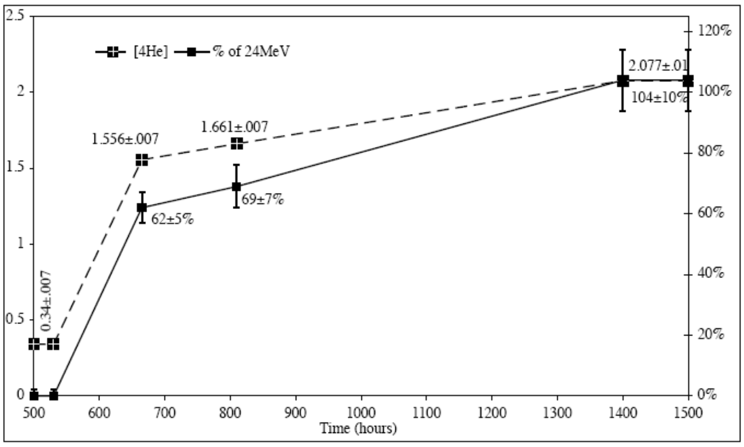

Figure 4. Measured ^{4}He concentration in a metal sealed electrochemical cell and mass flow calorimeter (data from Fig. 5, *left scale*) expressed as a percentage of the ^{4}He expected from D+D $\rightarrow$ ^{4}He $+$ 23.8 MeV heat (*right scale*).

increase in ^{4}He content instead of the decrease that would be expected since, to maintain positive cell pressure, the gas taken for the first sample had been replaced with cylinder D_2 containing a lower level of ^{4}He (0.34 ppmV/V). These findings support earlier observations that helium is released slowly from the palladium after an initial delay.

After making these measurements, an attempt was made to dislodge near surface ^{4}He by D atom motion, by subjecting the cathode to a period of compositional cycling, while still sealed in the calorimeter. Square and sine wave modulations of varying period and amplitude were imposed on the dc (negative) potential at the Pd electrode in an attempt to flux deuterium atoms through the interface and thus act to dislodge near-surface ad- or absorbed ^{4}He atoms.

At the end of this period, the potential was reversed to withdraw all deuterium atoms from the Pd bulk. No excess heat was observed during the periods of oscillation although calorimetric uncertainties were large due to the strong departures from the steady state that accompanied the pulsing. Gas samples were taken before this procedure, again after purging the cell and refilling with D_2 from the gas bottle with 0.34 ppmV ^{4}He, and once more after cycling. The latter sample exhibited the highest concentration of ^{4}He measured in this cell, specifically 2.077 ± 0.01 ppmV/V. By making a proper mass balance of the helium lost through sampling and purging, and that gained through make-up from the gas bottle, it is possible to assess with defined uncertainty the results of deuterium fluxing in freeing lightly trapped ^{4}He. The final integral mass balance yielded a value of 104 ± 10% of the expected value if the excess power in Fig. 5 is due to a net reaction of the sort $D + D \rightarrow {}^4He + \sim 23.8$ MeV (heat).

2. Conclusions

In this brief review we have attempted to demonstrate that the Fleischmann–Pons Effect (FPE) is a conditioned response of suitably prepared palladium cathodes to prolonged electrolysis in heavy water. The effect occurs when relatively high atomic deuterium loadings are achieved and maintained within a palladium cathode in the presence of suitable lattice

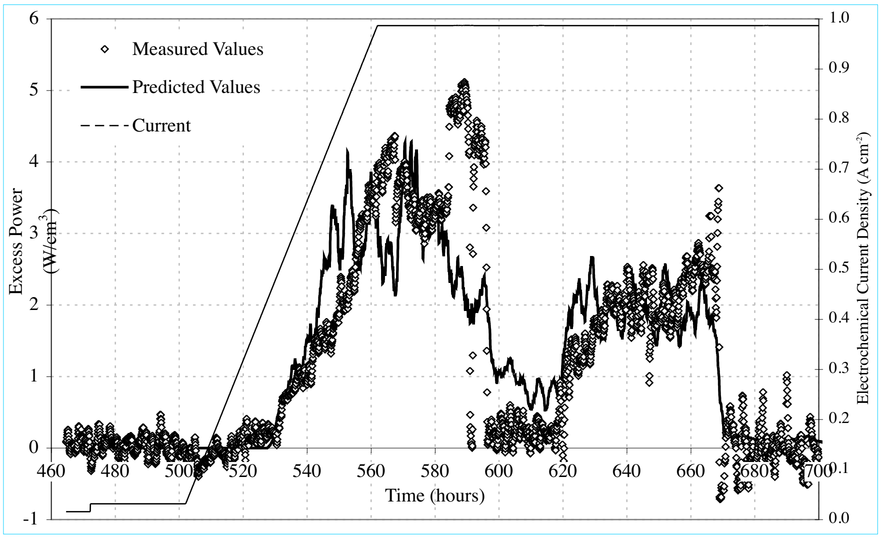

Figure 5. Excess power from a 1 mm × 10 cm Pd wire cathode electrolyzed in 1 M LiOD, in a metal sealed mass flow calorimeter measured and predicted from equation[1] (*left axis*), and electrochemical current density (*right axis*).

stimulation. The reasons for and nature of this stimulation has not been elaborated in this review, although suitable stimuli have been shown to include highly modulated electrochemical driving currents, high axial and interfacial current densities and induced deuterium atom fluxes.

The conditions necessary for the effect were not known or anticipated by early experimenters attempting to replicate the thermal anomalies reported initially by Fleischmann and Pons. Accordingly, these experiments mostly failed to produce the effect, which is now seen in amounts that are large both statistically, thus ruling out measurement error, and in absolute terms. Excess heat is measured in quantities greatly exceeding all known chemical processes and the results are many times in excess of determined errors using several kinds of apparatus. Nuclear products have been sought and found to be associated with the excess heat consistent with an overall reaction $D + D \rightarrow {}^4He + \sim 23.8$ MeV (heat), although a great deal more work is needed to elucidate a detailed reaction mechanism. Although not discussed here, the effect also appears in gas phase experiments employing Pd and D_2, with similar correlation between measured heat and helium production.

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