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Research Article

The Case for Deuteron Stripping with Metal Nuclei as the Source of the Fleischmann–Pons Excess Heat Effect

Thomas O. Passell*

TOP Consulting, P.O. Box 336, Palo Alto, CA 94302-0336, USA

Abstract

Evidence is cited from the research literature on metals containing absorbed deuterium supporting the hypothesis that the excess heat episodes observed over the past 25 years are the result of exothermic deuteron stripping reactions with atomic nuclei of the absorbing metal. The deuteron stripping reaction is one in which the neutron half of the mass 2 deuteron is captured by an atomic nucleus while the proton half of the deuteron is ejected, repelled by the coulomb field of the positively charged metal nucleus. This hypothesis provides a plausible explanation why so little external radiation accompanies the episodes of excess heat first observed by Fleischmann and Pons [1]. The reaction products from stable isotopes of the host metal are a proton with energies up to 9.2 MeV energy and a recoiling nucleus with energies of ∼100–to 600 keV. These two reaction products are retained near their birthplace because their range in solids is less than ∼100 μm. The emitted proton is energetic enough to produce by (p,n), (p,α), (p,T), and (p,X-ray), reactions with host metal nuclei and their light-element impurities, the small number of neutrons, alpha particles, tritium atoms (T), and X-rays, occasionally observed associated with deuterated Ti and Pd. The PIXE process (proton induced X-ray emission) is expected in which numerous K, L, and M X-rays of the absorbing metal are produced. For metals with thicknesses of >1 mm the vast majority of such X-rays do not escape the metal. In experiments with foils of the host metal sufficiently thin, low levels of charged particles (mostly protons) have been observed. Some of the observed protons were at energies larger than 3.0 MeV, the largest possible energy of protons from the fusion of two deuterons. Widely observed He^4 and tritium are known products of the deuteron stripping reaction with Li^6, which is a major constituent of electrolytes and a minor impurity in most metals. In any case, researchers have observed small but definite indicators of nuclear reactions other than d+d fusion in deuterated metals at temperatures not significantly above ambient.

Keywords: Deuterium, Energy, Metals, Nuclear, Oppenheimer–Phillips

1. Introduction

One of the most robust sets of observations over the past 25 years following the initial claims of excess heat in March, 1989, have been many episodes of excess heat in palladium and titanium cathodes polarized in lithium deuteroxide or potassium carbonate electrolytes [1]. Yet the nature of the implied nuclear reaction source of that excess heat

*E-mail: top94302@gmail.com

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has remained unresolved. The most persuasive reason behind the assertion that the heat source is of nuclear origin is its sheer amount, many times greater than any conceivable chemical or metallurgical process in so small a mass of materials. The most common reasons for doubting a nuclear source of the excess heat has been the absence of experimental proof of the predicted amount of the product(s) of any hypothesized nuclear reaction. If the deuteron stripping reaction turns out to be the primary source of the excess heat episodes then it will be understandable why expected levels of the nuclear reaction products have gone undetected all these years. Subtle metal isotope abundance shifts and protons are difficult to identify in such experiments.

This paper presents evidence for the deuteron stripping reaction as the possible primary source of the observed excess heat episodes. The evidence comes from a few of the hundreds of papers published in the proceedings of 18 international conferences on this subject since 1989. Most of the entire set of papers are available as downloadable items from the web site lenr-canr.org [1].

This reaction type was first studied by Oppenheimer and Phillips in 1935 [2]. In a series of deuteron bombardments in the Berkeley cyclotron their colleagues observed products of the (d,p) reaction with target metal nuclei significantly greater than products of all the other deuteron reactions with the same nuclei at the same energy. They theorized that the (d,p) reaction probability should be much larger than the other reactions at a given deuteron energy because the (d,p) reaction did not require the entire deuteron to enter the target nucleus. The neutron end of the cigar-shaped deuteron had no coulomb repulsion to overcome and might allow an interaction in which the neutron end of the deuteron was captured by the target nucleus while the proton was physically still outside the main portion of the target's repulsive coulomb field. Thus the proton can be stripped away by the strength of the repulsion by the positively charged metal nucleus – readily overcoming the proton's weak binding energy of 2.25 MeV to its neutron companion; hence the name, the deuteron stripping reaction.

If the preference for stripping relative to production of a compound nucleus is extrapolated from the heavy elements at $\sim$5-MeV deuteron bombarding energies down to near thermal (sub-eV) energies, the preference for stripping to compound nucleus formation is in the range of millions to one.

2. Logic of the Evidence for (d,p) Stripping Reactions in Deuterated Pd and Ti

The logic of the evidence is as follows: (A) An extensive study at the University of Rome showed evidence for the emission of gamma rays at an energy of $\sim$89 keV/photon from a deuterated palladium cathode during a 150-hour episode of credibly measured excess heat [3]; (B) Interpretation of the $\sim$89 keV gamma rays as plausible evidence for the production of the radioactive isotope Pd^{109}; (C) Assumption that the production of Pd^{109} is plausible evidence for the deuteron stripping reaction on stable isotope Pd^{108}; (D) Calculation of the total excess heat represented by the heat from the Pd^{108} deuteron stripping reaction (3.87 MeV/reaction) and the heat of the radioactive beta decay of Pd^{109} to Ag^{109}; (E) Finding the total energy represented by the above calculation to be a significant fraction of the total excess heat actually measured during 150 of the 1000 h experiment; (3) (F) finding evidence of small changes in isotopic abundance among three of the six isotopes in palladium that had produced excess heat and (G) reported observation of protons with energies greater than the maximum possible from d+d fusion (3.0 MeV) emitted from thin foils of deuterated titanium during changes in sample temperature. (see Kieth et al. ICCF10 proceedings within [1])

3. Isotopic Abundance Ratio Changes among three of the six Pd Isotopes from Cathodes That Had Produced Excess Heat

A significant indication of the direct involvement of the Pd metal atomic nuclei in reactions producing excess heat is the change in the ratio of the abundance of three of the six Pd isotopes relative to one another after excess heat episodes – compared to those same ratios measured in the virgin Pd material.

Due to the generosity of Professor Arata and his colleague Dr. Zhang, at the University of Osaka, four samples of palladium powder were obtained from their experiments which had produced excess heat [4]. Three were from the sealed interiors of hollow palladium cylindrical cathodes each of which had been observed to produce excess heat of $\sim$ 60 MJ during a six-month period of electrolysis. The fourth was virgin material from the same batch of powdered Pd. Passell and George reported the results of neutron activation analysis (NAA) on all four samples from which one can derive abundance ratio changes for Pd^{110}, Pd^{102}, and Pd^{108} [5]. These three Pd isotopes are the only ones that produce gamma-ray-emitting radioactive isotopes following neutron capture in NAA.

The result was the observation of an increase in the ratio of Pd^{110} to $Pd^{102}$and Pd^{108}, respectively, relative to those of the virgin material. These isotopic ratio changes indicate that these Pd isotopes must have been differently affected by whatever nuclear reaction between deuterons and each of them caused the expression of excess heat. One such difference is the likelihood of different probabilities of the (d,p) reactions on each separate Pd isotope, the Q's of which vary from 2.88 MeV for Pd^{110}, 3.87 MeV for Pd^{108}, to 5.54 MeV for Pd^{102}. The amounts of the ratio changes ranged from +8(0.8)% for 110/108 to +24(11)% for 110/102. If the cause of the ratio changes were a heat-producing (d,p) reaction that differed among the Pd isotopes, the changes are of the right order of magnitude to explain the amount of excess heat reported by Arata and Zhang [4]. Of course all three isotopes could be sources of heat, not just 108 and 102 Also the remaining stable Pd isotopes with masses of 104, 105, and 106, not amenable to NAA ratio measurements, could also be producing heat by deuteron stripping reactions.

4. Appearance of Sc^{46} from Electrolysis of Titanium

Another piece of supporting evidence comes from the electrolysis of titanium metal cathodes by Mengoli and coworkers at the University of Padua [6]. After some twenty days of electrolysis at 95 Degrees C in which several hundred kilojoules of excess heat had been observed, they employed two high-resolution germanium gamma ray detectors in coincidence to discover the appearance of radioactive Sc^{46} in the cathodes. (The two gamma rays emitted by Sc^{46} in its decay to Ti^{46} are in coincidence.) The total number of Sc^{46} isotopes produced were only sufficient to account for an insignificant amount (1×10^{-7} J) of the observed excess heat. Two possible mechanisms for Sc^{46} production are: (1) $Ti^{48}(d,\alpha)Sc^{46}$ and (2) $Sc^{45}(d,p)Sc^{46}$ from possible scandium impurity in the titanium. Both reactions are very unlikely because the first one requires a compound nucleus with the deuteron and the second one the million-fold higher probability stripping reaction with a possible scandium impurity at the PPB to PPM level in titanium. Thus the more probable (d,p) stripping reaction in any or all of the five stable isotopes of titanium (46–50) might well have been present and responsible for the observed excess heat. In effect, the scandium isotope emissions can be considered as the barely measurable tip of an energy iceberg.

5. Discussion

Some highly unusual shielding effects are necessary to allow thermal energy deuterons in metal lattices to undergo stripping reactions. One can speculate that shielding by the electrons in the metal electron conduction band must make possible the close approach of deuterons in the deuterium ion conduction band [7] to the metal atoms' nuclei to allow the stripping reaction to occur. The relatively stringent deuterium flux levels apparently required in Pd and Ti must be necessary before such unexpected nuclear reactions occur. It has been known since 1929 that hydrogen in palladium existed interstitially as positive ions that could undergo electro-diffusion like electrons through a Pd wire, only in the opposite direction [7]. Thus one might plausibly imagine coherent behavior in both the electron and ion conduction bands to be operating to allow close enough deuteron approach to metal nuclei to allow stripping.

If a deuteron stripping reaction is possible for the nucleus of element 46 (palladium) and 22 (titanium), it follows that similar reactions might be found among the $>$20 other metal elements (and their trace interstitial elements such as boron and lithium) known to absorb hydrogen. If such should be the case, a very large matrix of experiments will be required to determine the extent of the generality of such reactions.

Thus stripping reactions on each of some 30% ($\sim$80) of all 281 stable isotopes give many more possibilities of producing useful heat than the one d+d reaction. Their exothermic reaction heats range from 1.1 to 9.4 MeV (with Ti^{47}) vs 23.8 MeV for d+d$\rightarrow$$He^4$; however this is not a significant disadvantage.

Reviewers of this paper have raised some obvious questions about my selection of key parts of the evidence cited above. First, why focus on the appearance of 89 keV photon radiation reported by the Gozzi group at the University of Rome (3).? Clearly there are many possible sources besides Pd^{109} decay of radiation in the energy band they reported (within 1 keV of 89 keV). Viewing the list of 210 known gamma rays from 87.0 to 91.0 keV we note only one of significant percentage in the decay of any Pd radioactive isotope –that of the 88.03 keV gamma from Pd^{109} decay. The same gamma transition is emitted in Cd^{109} decay. So, how do we know that Pd is more probable as the radiation source than Cd? First, the radiation images on the X-ray film are lined up parallel to the cathode of the cell. Second, the cathode of the cell is a bundle of 150 Pd wires, basically 100% Pd. Third, the film images are a series of 69 small spots apparently caused by radiation from deeper layers of the wire bundle that could only exit via gaps between outer layers of Pd wires. Fourth, through detailed analyses of the 69 spots on the film, the Rome researchers could assign a narrow range of energies to the gamma source, since each spot could be optically identified with one of the deeper layers of wires. Gamma rays from these deeper layers of wires were observed to be attenuated by varying thicknesses of Pd wires in the outer layers. Use of the known attenuation coefficients of Pd allowed derivation of the photon energy to the narrow range of 88–90 keV. Of course X-ray film is not the ideal detector of gamma rays so their paper states that the energy of the gamma rays could have energies as high as 150 keV. If one considers the details of Pd^{109} decay, some 3% of the gamma emissions lie at energies of several hundreds of keV

The Gozzi group suggested that Cd^{109} might be the source since it decays to the same Ag^{109m} level as does Pd^{109}. However their supposition of Cd^{109}production by the Pd^{105}+23.8 MeV alpha$\rightarrow$$Cd^{109}$ + gamma is clearly unlikely since no He^4 atoms were found inside the body of the Pd wires in this experiment.

Gozzi et al. also state that the total energy in the 89 keV photons is only $\sim$0.5% of the excess heat measured during the 150 h film exposure, which agrees with the criticism that those gamma rays are not, in themselves, a major source of the excess heat. However, IF they are from Pd^{109} decay, they become the tip of an energy iceberg. First, each of the 88.036 keV gammas in Ag^{109m} decays to the ground state of Ag^{109} accompanied by 25 parallel decays of soft conversion electrons and silver X-rays with the same 88.03 keV energy content. Taking this into account multiplies the 0.5% by 25 times to $\sim$12%. In addition, the beta decay energy of Pd^{109} to Ag^{109m} is about 1.1 MeV. The average energy of electrons in that beta spectrum is about 300 keV. Thus each gamma photon implies the existence of 26 times as many 300 keV beta decay electrons. Thus part of the rest of the energy iceberg brings the total energy into a significant fraction of the observed excess heat about 100 times the energy in the photon itself, i.e. $\sim$50%.

Once we accept the existence of Pd^{109}, we must deal with the energetics of its production. Assuming the production is by Pd^{108}(d,p)Pd^{109}, then each Pd^{109} indicates that the energy associated with each exothermic stripping reaction has occurred, adding its Q of 3.87 MeV per reaction .

In summary, we can associate an energy of 88 keV +25 times 88 keV + 26 times 300 keV + 26 times 3.87 MeV with each 88 keV photon observed. So the energy iceberg is about 118,000 keV relative to only 88 keV per photon or a multiplier of about 1340 times. Using this multiplier on the Gozzi et al estimate of 0.5% of the 2.4 megajoules of excess heat observed during the 150 h film exposure, leads to an impossibly high number of 673% of the excess heat observed. Thus the estimate of the 0.5% by the authors is probably high by about a factor of 10 or more. That estimate is attempting to use analysis of 69 spots on a film to quantify the source, a procedure highly dependent on particular assumptions of uncertain validity.

We can reasonably assume that this extrapolation is the likely reason for the large mismatch in estimated and observed excess heat. If the error is of the order of a factor of ten, then we are left with only 67% of the observed heat explained by deuteron stripping on Pd^{108} alone. How do we explain additional heat sources in this situation?

The most obvious answer is to assume that Pd^{108} was not the only Pd isotope active with the deuteron stripping process. The other likely suspects are those Pd isotopes similar in every way except their exothermic Q values. These would be Pd^{104}, Pd^{106} and Pd^{110}, (Pd^{102} with only 1% abundance is being ignored). Pd^{105} has a larger Q of 7.35 MeV but it exhibits a larger spin change in the stripping reaction than the three zero spin even-even (even numbers of both protons and neutrons) isotopes. Larger spin changes are associated with reduced probability of a given nuclear reaction. The sum of Pd^{104} and Pd^{110} abundances are only ~82% of the abundance of Pd^{108}. Thus we might expect the sum of Pd^{104} and Pd^{110} would double the amount of heat produced by Pd^{108} alone. Adding the Pd^{106} contribution, with its abundance very nearly the same as Pd^{108}, triples the expected heat relative to that calculated from Pd^{108} reaction radiation.

Finally, we note a surprising result from the Univ. of Rome study [3]. Their Pd cathode differs from rod, wire, foil, and mesh shapes used by other researchers over the past 25 years. It is a tightly bound bundle of 150 Pd wires, each with a 250 μm diameter so that dips and gaps occur between the outermost (e.g. seventh, sixth, fifth, etc.) layers of wires; the radiation emitted to darken the X-ray film did not come from that seventh outermost layer of wires but from the sixth, fifth, fourth, etc. concentric layers instead.

The benefits of this cathode structure are two: First, the spots on the X-ray film are distinct and geometrically identifiable in a way to show beyond doubt that the radiation does indeed originate at the cathode; and second, it suggests that deuteron flux may be the significant factor in excess heat production – since interior layers emitting the radiation are more likely to exhale previously absorbed deuterium, covered as they are by gas bubbles at frequent intervals during electrolysis.

6. Conclusions

Evidence presented above is certainly not yet definitive –only suggestive. With so little evidence of nuclear reactions beyond the sheer amount of excess heat all theories have had a difficult time explaining the results of experiments. For example, Gozzi et al. made one of the most thoroughly and expertly executed searches for He^4 expected from a set of episodes of excess heat, yet found far less than predicted [3]. None were found buried in the metal cathode wires. This is evidence that some other process is at work besides the widely assumed reaction d+d→$He^{4\cdot}$.

The protons emitted in the deuteron stripping reactions are energetic enough to excite the first few excited states of nuclei throughout the periodic table. This is a well-known process called coulomb excitation. While these reactions are produced with cross sections only in the range of a few barns, they would be readily observable in cases where excess heat at watt levels is occurring. The first few excited states of most nuclei are well known to high precision and should therefore be a good indicator of the presence of protons from deuteron stripping reactions. Positive identification of this process requires high resolution gamma ray detection.

In addition to coulomb excitation, fast protons can be expected to create neutrons by (p,n) reactions. However such (p,n) reactions with metal atom nuclei are inhibited by the coulomb barrier of the metal atom and secondly by the fact that the reactions with metal nuclei are mostly endothermic by several MeV. Thus most such reactions with metal nuclei are unlikely to be significant. However, such conditions are not so inhibiting for deuterons in the lattice. Protons can elastically collide with lattice deuterons, leading to subsequent d+d fusion reactions by recoiling deuterons. This two stage process drastically reduces the number of final fusion generated neutrons. Still under some assumptions this might lead to observable neutrons outside the experiment.

Since observations of neutrons outside previous experiments have failed to correlate neutron levels with excess heat measurements, more recent excess heat observations have almost never included neutron detection. It would be premature to conclude that the protons from deuteron stripping reactions are ruled out by previous failures to observe external neutrons, since so few first class neutron measurements have been made coupled with first class observations of excess heat.

Small amounts of tritium have been observed associated with both excess heat and pulsed glow discharges. In the cases showing excess heat the tritium levels are of the order of one million times lower than expected if tritium-producing reactions were a significant fraction of those producing excess heat. However, the significance of such observations far outweighs the amounts observed. The unambiguous nature of the detection of tritium shows that nuclear reactions can occur in deuterated palladium, a remarkable proof of the possibility of nuclear reactions in this system (see papers by F.Will et al. and T. Claytor et al. in [1]).

Confirmation of this hypothesis will require finding at least one unequivocal product of the deuteron stripping among elements that absorb deuterium. The search for gamma ray emitters may be more sensitive and less subject to question than the search for excess heat. Moving beyond Ti and Pd to mono-isotopic elements such as scandium, cobalt, terbium, thulium and tantalum might prove fruitful, as each one gives a well-characterized prolific gamma ray emitter of convenient half life (weeks to months) after experiencing a deuteron stripping reaction.

A 1953 thesis by W.B. Hillig gives evidence for hydrogen transport in titanium and tantalum metal in ways similar to such phenomena more thoroughly studied in palladium. Thus we may not be limited to stripping reactions in the expensive metal, palladium [8].

Pursuing the deuteron stripping hypothesis is a significant departure compared with current ones. A large advantage of the deuteron stripping hypothesis is the lack of ambiguity over the mechanism for conversion of energy from excited nuclei to heat. Heating by fast protons stopping in matter is a well established heating phenomena.

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