LETTERS TO THE EDITOR

COMMENTS ON "SEARCH FOR ^{3}He AND ^{4}He IN ARATA-STYLE PALLADIUM CATHODES I: A NEGATIVE RESULT" AND "SEARCH FOR ^{3}He AND ^{4}He IN ARATA-STYLE PALLADIUM CATHODES II: EVIDENCE FOR TRITIUM PRODUCTION"

We write in response to two recent papers by Clarke[1] and Clarke et al.[2] The underlying assumption in the experiments described is that the D_2O electrolysis induces reactions, reputed to produce ^{3}He, ^{4}He, and tritium, and occurs in the palladium black filled cavity within the Arata-style cathode. Much prior research on the Pons and Fleischmann[3] style solid palladium cathodes has proven that the heat and helium producing reaction occurs on the surface of the palladium where the deuterium pallide cathode surface is exposed to the driving force of the electrolysis' electrical current. Identifying the location of the induced reaction is of critical importance to future research because detection of the nuclear ash (^{3}He and ^{4}He, usually) requires analysis pertinent to the true reaction location.

We question the implicit assumption that the tritium forming reaction occurred inside the Arata-style cathode; there is, at least, one other viable route to the appearance of tritium in the Arata-style cathode. Tritium can be loaded into palladium electrolytically, like deuterium. Thus, tritium formed at the surface of the cathode would be driven into the core of the cathode, as was observed.[2] Direct experimental evidence that the heat and helium producing reaction occurs only at the surface of the cathode also speaks to this point.

The earliest serious study of nuclear reaction products associated with the calorimetric excess heat generated by the Pons and Fleischmann effect was "Measurements of Helium in Electrolyzed Palladium," by Morrey et al.[4] That extensive collaborative effort found essentially no relevant helium inside the palladium cathodes. The first successful observation of helium produced by the Pons and Fleischmann effect[5] focused on helium in the electrolysis off-gas because of our assumption that the reaction would occur at the surface of the cathode where the electrolysis deposited its electrical energy. During that study, eight electrolysis off-gas samples taken during episodes when excess heat was generated correlated with helium being found each time; during six periods when no heat was generated, no helium was found (the mass spectrometer used being relatively insensitive).[6] The direct relationship between the helium production and excess heat generation study by Bush et al.[5] led to the establishment of the Navy "Program to Understand Deuterated Metals."

Subsequent quantitative helium analysis of the electrolysis off-gas was undertaken by one of us (BB) at SRI and by Miles working independently at China Lake. The amount of calorimetric excess heat generated was correlated with the amount of helium produced, which identified the nuclear reaction pathway explicitly as the $D + D \rightarrow {}^4He + 23.82$ MeV(heat) reaction.[7–9] These findings are reproducible.

The ultimate advantage of helium analysis is expressed in its ability to identify the source of helium by comparing known environmental ratios of ^{3}He:^{4}He:^{20}Ne:^{22}Ne to the ratios found experimentally. The tritium content of the D_2O creates a serious complication as it decays to ^{3}He at perhaps 50 dis/min·ml^{-1} (Ref. 2) depending on the D_2O used. Thus, artificially large ^{3}He:^{4}He ratios will be found if the D_2O is allowed to dwell in contact with the gas to be sampled for a prolonged length of time. Accordingly, gas sampling manifolds were developed to be continuously flushed by the evolving electrolysis off-gas. The manifold system volumes were adjusted so that a system volume of off-gas would be evolved in ~1 h, which also approximated the time response of the calorimeters; thus the helium analysis would correlate with the calorimetric excess heat, in real time. After vigorous nitrogen flushing, each successive system volume of electrolysis off-gas would remove one-half of any residual contamination (e.g., after 14 system volumes of electrolysis off-gas flushing, $(\frac{1}{2})^{14} = 0.0061\%$ of the original possible residual contamination would remain). In the absence of excess heat generation, gas samples valved-off after extensive electrolysis off-gas flushing were demonstrated to contain very low residual atmospheric contamination content, as witnessed by the ^{3}He:^{4}He:^{20}Ne:^{22}Ne ratio analysis.[7]

Every effort was made in our experiments to eliminate atmospheric contamination and tritium induced contamination of the samples, with the idea that a low ^{3}He:^{4}He ratio would be the characteristic signature of the reaction $D + D \rightarrow {}^4He +$ 23.82 MeV (heat). To our surprise, during excess heat generation the ^{3}He:^{4}He ratio was larger than ever observed before in the electrolysis off-gas. The ^{3}He:^{4}He ratio found was a very close match to that calculated theoretically by Tom Ward (who was then chief scientist of the U.S. Department of Energy's Accelerator Production of Tritium Program), i.e., $^3He/^4He = 2.98 \times 10^{-6}$ (Refs. 8 and 9).

The premise that the cathode is energetically coupled to the fusion reaction is necessary to explain the experimental data because the Pons and Fleischmann effect generates heat without commensurate penetrative nuclear radiation. During electrolysis, a nonconductive film builds up on the cathode surface from species born in solution (e.g., silicates); a large voltage gradient (of up to perhaps 10^7 V/cm) develops across that thin surface film, where the reaction occurs, as witnessed by the helium being found in the electrolysis off-gas. The D + D $\rightarrow$ ^{4}He reaction involves an electromagnetic transition from an activated ^{4}He* intermediate; such transitions typically release their energy over a 10^{-16} to 10^{-21} s time frame.[10] Multiplying the time frame by the speed of light (c) gives an electromagnetic interaction range comparable to atomic lattices. Evidently, electromagnetic forces in the 10^7 V/cm voltage gradient of the electrolysis zone are strong enough to energetically couple the nuclear reaction to the cathode lattice. The voltage gradient does not extend deep into the cathode, so there is nothing to induce reaction in the core of the cathode. Thus, the helium is entrained in the electrolysis off-gas because of the nature of the reaction environment on the cathode surface.

In conclusion, the Pons and Fleischmann effect is a form of nuclear fusion induced by electrolysis at the cathode surface. Quantitative helium analysis of the electrolysis off-gas taken during episodes of excess heat generation identified the nuclear reaction pathway as D + D $\rightarrow$ ^{4}He + 23.82 MeV (heat) (Refs. 7, 8, and 9). Analysis of the ^{3}He:^{4}He:^{20}Ne:^{22}Ne ratios provided a powerful tool for tracing the source of the helium being observed, and for studying the nuclear reaction kinetics.[8,9]

Helium analysis of the cathode metal[1,2] should generally be expected to fail[4] because the Pons and Fleischmann effect occurs at the surface of the cathode so that the helium produced escapes with the electrolysis off-gas.[5–9]

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RESPONSE TO "COMMENTS ON 'SEARCH FOR ^{3}He AND ^{4}He IN ARATA-STYLE PALLADIUM CATHODES I: A NEGATIVE RESULT' AND 'SEARCH FOR ^{3}He AND ^{4}He IN ARATA-STYLE PALLADIUM CATHODES II: EVIDENCE FOR TRITIUM PRODUCTION'"

Before starting our work,[1,2] we were aware that Arata and Zhang had claimed detection[3] of large amounts of ^{3}He and ^{4}He (10^{16} to 10^{17} atoms/mg) in the Pd-black inside hollow Pd cathodes electrolyzed in D_2O. We made no other underlying assumptions, although we were (and still are) extremely skeptical about these and many other claims made since 1989 regarding evidence for so-called "cold fusion."

Our measurements showed that at least 1.8×10^{15} atoms of tritium were produced inside an Arata-style cathode during electrolysis in D_2O (Ref. 2). The conclusion that tritium was produced inside the electrode was not an "implicit assumption" as suggested by Bush and Lagowski,[4] but was based mainly on the following experimental results:

1. Concentrations of ^{3}He in pieces and filings from a section of the Pd electrode[2] showed a negative gradient from the inner to the outer wall. This observation indicates that the source of the tritium parent was inside the electrode and that the ^{3}He gradient was established after cathodic electrolysis

when some of the gaseous tritium (as HT, DT or TT) diffused outward from the electrode interior.

2. The tritium inventory in the 0.5 L of D_2O electrolyte was 1.2×10^{12} atoms before electrolysis began and was 1.6×10^{12} atoms after electrolysis ended.[2] Although a small fraction of tritium originally in the D_2O must have entered the interior of the electrode during electrolysis, it is negligible compared to the tritium found there.

Bush and Lagowski[4] have suggested that the tritium was formed on the outer surface of the Arata-style cathode, and that it was driven into the interior during electrolysis. We considered this possibility, but discarded it because it did not fit the experimental results. The ^{3}He concentration gradient through the electrode wall[2] is not consistent with inward migration of tritium, although this observation alone cannot decide the issue. In our view, the issue is decided by the serious difficulties that arise when tritium-^{3}He residence times are considered. For example, the "fingerprint" of radiogenic ^{3}He deposited near the outer Pd wall during inward tritium migration cannot have exceeded the observed concentration of $\sim 300 \times 10^3$ atoms/mg (Ref. 2). Taking the mean ^{3}He concentration in the wall to be $\sim 150 \times 10^3$ atoms/mg, and a wall mass of 55 g, we calculate that the upper limit for the residence time of 1.8×10^{15} atoms tritium in the wall during postulated inward migration is ~ 0.7 h. This time seems unreasonably short when compared to the cathodic electrolysis time of 90 days. More believable tritium-^{3}He residence times are obtained for tritium diffusing outward from the electrode interior because of the higher ^{3}He concentrations near the inner surface, and the fact that only gaseous tritium, which was $\sim 3\%$ of total estimated tritium when the electrode was punctured[2] was able to diffuse through the electrode wall. In addition, because some gaseous tritium was detected inside the electrode,[2] the observed ^{3}He concentrations in the wall were probably generated during outward migration of only part of the gaseous tritium trapped inside the electrode at the end of cathodic electrolysis.

The last seven paragraphs of Bush and Lagowski's letter,[4] contain several descriptions of their earlier work on helium in electrolysis off-gases. Although we did not examine the electrolyte or off-gases for ^{4}He, measurements on palladium filed from the outer 0.1 mm of the Arata-style electrode indicated an upper limit of 5.5×10^{10} atoms ^{4}He in that layer.[2] During D_2O electrolysis at SRI International, excess heat of 64 MJ was measured for this electrode.[5] If this heat is assumed to come from the reaction $D + D = {}^4He + \gamma + 23.8$ MeV then $\sim 1.7 \times 10^{19}$ atoms of ^{4}He should have been produced. We did not find any significant ^{4}He in the gas from the interior of the electrode, in the Pd-black, or in various pieces of the Pd metal. Thus, the ^{4}He upper limit in the 0.1 mm outer layer indicates that less than ~ 1 part in 300 million of the "expected" ^{4}He recoiled into the electrode.

In a recent letter,[6] we stated our opinion about the helium measurements of Miles et al. At that time we were not aware of the two recent conference papers by Bush et al.[7,8] which were referred to by Bush and Lagowski.[4] We have now examined these papers carefully and have concluded that they contain insufficient new information to convince us that the ^{4}He (and most of the ^{3}He) measured by these workers is not of atmospheric origin. Tritium concentrations in the D_2O and D_2-O_2 electrolysis off-gases, and tritium-^{3}He decay intervals for the off-gases were not given.[8] Thus, we cannot rule out the possibility that observed excess ^{3}He relative to atmospheric ^{3}He/^{4}He, is from decay of tritium contaminant in the D_2O. The observed enrichment in ^{4}He/^{22}Ne (relative to atmospheric ^{4}He/^{22}Ne) could be due to enhanced diffusion of He versus Ne from the atmosphere into the apparatus, or from a remnant of dissolved air in the water bath surrounding the calorimeter used by Bush and Lagowski.

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*We are sorry to inform our readers that Dr. W. Brian Clarke is deceased.