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NUCLEAR PRODUCTS ASSOCIATED WITH THE PONS AND FLEISCHMANN EFFECT; HELIUM COMMENSURATE TO HEAT GENERATION, CALORIMETRY AND RADIATION.

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Abstract: The nature of the nuclear phenomena associated with the Pons and Fleischmann effect remains largely unexplored. The phenomena are reproducible, but the processes lack controllability. The circumstances of the electrolysis experiments do not produce the same nuclear product distribution as that expected during hot plasma D + D fusion experiments.

From our earliest qualitative heat versus helium nuclear products analyses[1] to our more recent quantitative helium analyses; the utmost care has been exercised with respect to the scientific rigor of our work. The correlation between the production of helium and the generation of excess heat has been reproduced in different laboratories and under different experimental protocols. Preliminary results are shown in Fig. 1. The quantitative correlation between the amount of energy generated and the helium produced is at the level that is expected for a high energy nuclear reaction, such as fusion. These results are underwritten by extensive ^{3}He:^{4}He:^{20}Ne control experiments.

Calorimetric quality is the foundation of this work. In our early work, isoperibolic calorimetry was used successfully. In our later work as depicted in Fig. 1, high performance Calvet calorimetry is used. This is the most rigorous method of calorimetry known, amounting to an integrated measurement of the total thermal flux. Electrolysis off-gas production rates were measured to determine the Coulombic efficiency of the electrolysis. Atmospheric helium contamination was precluded by use of all-metal sampling flasks and all-metal gas collection equipment with helium leak-tight Cajon VCR metal seals.

Figure 1. D + D --> ^{4}He + 24 MeV is the most energetic reaction known. The heat *versus* helium analysis can be used to attempt to identify the nuclear reaction pathway by comparing the quantity of helium produced to the amount of energy generated. Thus, the most energetic reaction known would generate 24 MeV/^{4}He, as depicted by the line in the plot. Likewise, the 4 MeV/^{4}He line is included to add perspective. These results were obtained with all-metal apparatus shown to be capable of eliminating atmospheric contamination.
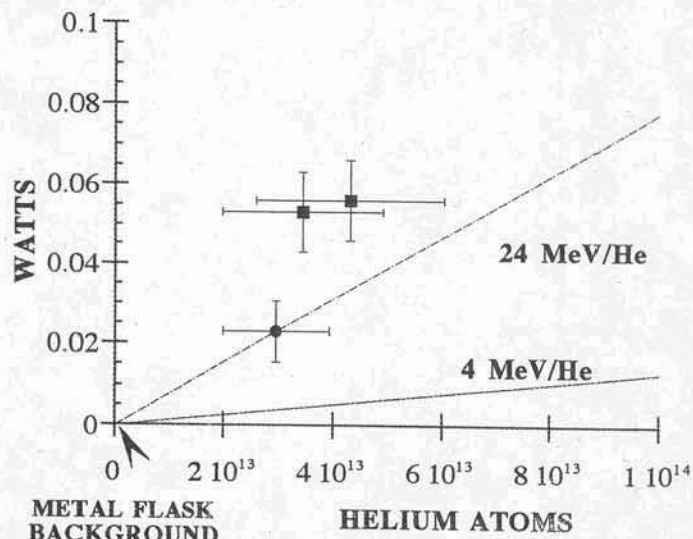

Radiation monitoring suggested the presence of a weak source of high energy γ-radiation. The weakness of the source tended to confound the analysis because of the statistics of the minimum detectable activity associated with various radiation detectors. The cathodes used in these experiments were palladium electroplated on gold-flashed copper. No calorimetry was associated with these radiation experiments.

The ultimate goal of this effort is to identify explicitly which nuclear reactions result in the Pons and Fleischmann effect.

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Introduction: The quantitative nuclear products study of excess heat generation versus ^{4}He production helped garner $750,000.00 of funding for SRI. This data, presented in Fig. 1 was the result of decisive application of the most rigorous methods of calorimetry and helium sample collection known. A collection of available data is shown in Fig. 2:

Figure 2. Quantitative heat *versus* helium analysis, using isoperibolic calorimetry.[1] Note that the data of Fig. 1 overlays; different calorimetric techniques are used but the same type of result is obtained. Helium sequestered by the cathode would result in an apparent increase in the energy per ^{4}He atom. Reaction at the cathode surface is suggested by the ^{4}He being found in the $D_2 + O_2$ off-gas.
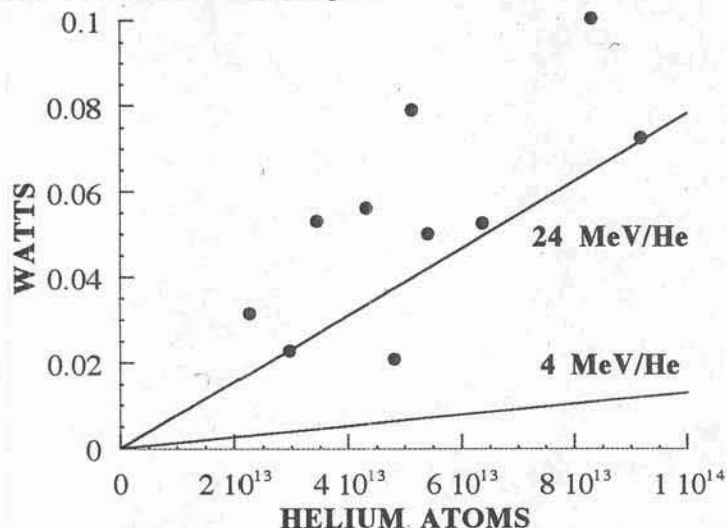
Notice that the data appears to collect around the 24MeV/^{4}He line in Figs. 1 and 2. Nuclear reaction pathways can be identified by comparing the quantity of nuclear product (^{4}He) to the amount of energy generated (~Watts). The 24MeV/^{4}He line in Figs. 1 and 2 correspond to D + D --> ^{4}He + 24MeV, the most energetic reaction known. Theoretically, amount of energy (E) expected for the mass (m) lost in the reaction is given by the equation $E = mc^2$, where c is the speed of light. The mechanism can probably induce more than one nuclear reaction pathway, and so we are beginning the nuclear products analysis of the cathodes, in addition to continuing heat and helium studies.

Experimental: Our early finding of helium production during excess heat generation[1] was validated by control experiments; hence 8 excess heat events correlated to the detection of ^{4}He in the electrolysis off-gas, 6 control experiments produced samples of off-gas containing no detectable helium. The probability that these experiments are due to random causes is $(1/2)^{14}$=0.0061%; i.e. there is a 99.9939% probability that we observed a real phenomenon. The use of isoperibolic calorimetry and glass apparatus resulted in unwarranted criticism. We will now describe more rigorous methods, noting that the conclusions (Fig. 1) are similar to our earlier work (Fig. 2).

Helium does not diffuse through metals, as a practical matter. Thus, a rigorously all metal Seebeck calorimeter and all metal gas collection system was constructed to preclude atmospheric contamination. Our tactic has always been to use a self flushing system, electrolysis off-gas flushes out any contamination in a continuous manner, through D_2O and oil bubblers. The electrolysis energy input is corrected for the $2D_2 + O_2$ gas formation, and the validity of the energy input correction is checked periodically by measuring the $2D_2 + O_2$ gas production rate versus the electrolysis current to ensure that the $2D_2 + O_2$ gas is not recombining in the calorimeter to reform D_2O and thereby increasing the effective electrolysis energy input. The data was normalized to 525mA in Figs. 1 and 2. All metal gas collection flasks are baked out under vacuum and N_2 flushed repititiously, to remove any helium occluded in pores in the metal surface. A schematic of this rigorous system is shown in Fig. 3; use of this system resulted in the Fig. 1 data.

Seebeck calorimetry is, functionally, integrating thermal flux envelope calorimetry. The electrolysis cell is completely enclosed by a thermal flux transducer envelope, so that essentially all heat leaving the cell is measured. The thermal flux transduction envelope consists of series differential thermocouples set across a homogeneous thermally insulative layer (see Fig. 3). The heat flowing through the insulative layer sets up an electrical potential across each differential thermocouple pair; the series sum voltage of all the differential thermocouple pairs represents the total heat flow.

Seebeck calorimetry differs from Calvet calorimetry in that the insulative layer between

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the differential thermocouple pairs, is a rather good conductor of heat; a high density of thermocouple pairs (about 30 pairs per cm^2) allows high sensitivity. Ease of heat flow to the thermostatic water bath minimizes unmeasured heat leakage, the effect of which is calibrated. Seebeck calorimetry has an enormous dynamic range, our 3X3X9 cm devices can measure from a few milliwatts to tens of watts with approximately $\pm$ 0.3% accuracy. For comparison: isoperibolic calorimetry used in our early work[1] had a dynamic range from about 1 to 4 Watts. Further, it takes about 3 hours for the isoperibolic calorimeters to achieve thermal equilibrium, while it takes only half an hour for the Seebeck.

Figure 3

N$_2$ Flush
N$_2$ Flush
Gas Sample
D$_2$O Calibration Heater
Fluid Level Sensor
Platinum Anode (+)
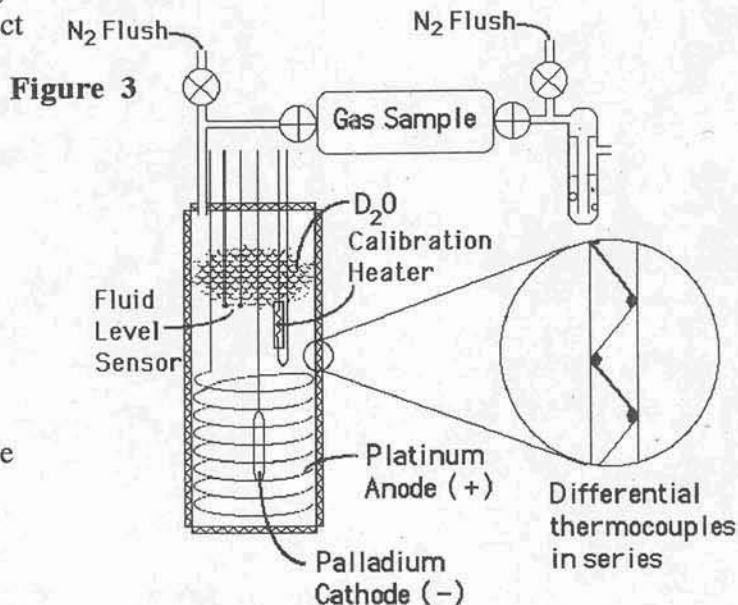Differential thermocouples in series
Palladium Cathode (–)

Seebeck calorimetry is high performance Calvet calorimetry. It combines the benefits of high sensitivity, enormous dynamic range, fast response and convenience of operation. Thermal homogeneity within the calorimeter is irrelevant, because all flux paths are covered by the thermal flux transduction envelope. Seebeck calorimetry is the most rigorous method of calorimetry known (note: Fig. 1).

An attempt was made to replace the Seebeck thermal flux transducers with cheaper Peltier devices.[2] Initially, the calorimeter seemed to give a reasonable response, and then it became more and more non-linear over time. Such an insidious failure appears to have occurred at the Naval Research Lab; and elsewhere.
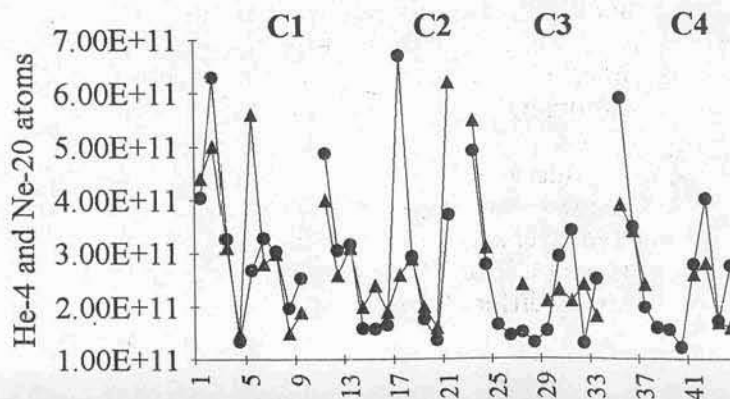
Mass Spectral ^{3}He:^{4}He:^{20}Ne analysis has the advantages of helping identify the helium source and directly probing the branching ratio of the nuclear reaction. Tritium, which decays to ^{3}He can also be analyzed. ^{3}He analysis is blinded by the presence of HD (also mass 3), so all deuterium is removed prior to analysis.

^{4}He:^{20}Ne analysis (Fig. 4) is plotted with respect to 4 separate electrolysis systems. The ^{4}He analysis used in Fig. 1 had a detection threshold of 1 ± 1 parts per billion; the ^{3}He:^{4}He:^{20}Ne analysis now in use (Fig. 4) has a ^{4}He detection threshold in the low parts per trillion range. The ^{3}He:^{4}He isotope ratio associated with the data of Fig. 4 is about twice that expected for air, due to tritium decay. The ^{3}He:^{4}He:^{20}Ne electrolysis off-gas sampling manifold was constructed of metal and glass, sealed with viton o-rings, and is more than good enough to acquire the rigorous data of Fig. 1. Sampling flasks are all metal, in both studies. Hence, the contemporary data (Fig. 4) acts as a massively redundant control, validating our findings depicted in Fig. 1.

Figure 4: Note: multiply 7 X 10^{11}atoms ^{4}He by 10 = 7 X 10^{12} atoms, this corrects for sample flask volume differences. Now compare 7 X 10^{12} atoms with the rigorous helium analysis in Fig. 1. The different permeation rates of ^{4}He and ^{20}Ne accounts for their isotope ratio, ^{20}Ne diffuses slower than ^{4}He.

He(dots) & Ne(triangles), four seperate systems C1- C4

C1   C2   C3   C4

He-4 and Ne-20 atoms

7.00E+11
6.00E+11
5.00E+11
4.00E+11
3.00E+11
2.00E+11
1.00E+11

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A small radiation signal was observed associated with 0.001" thick palladium plated onto gold flashed copper cathodes. We were unable to acquire spectra perhaps owing to broad band scatter.[3] Such broad band scatter might be taken to suggest a bremsstrahlung continuum spectrum[4] arising from "hot electrons" associated with the mechanism of energetic coupling between the nuclear reaction and the metal lattice.

The radiation was traced to the electrolytic cell by systematic displacement of the detector, using the fact that radiant intensity (I) decreases as the square of distance (D) from the source $I \propto 1/D^2$, in Fig. 5.
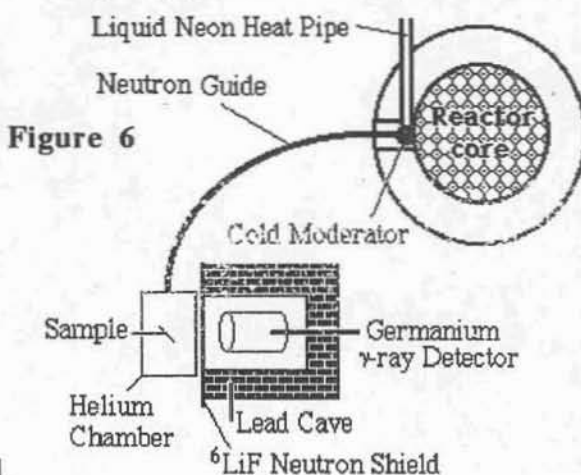
Figure 5: Plots of cell voltage and radiation count rate for a NaI detector and a G-M detector. Count rates are in counts per second average over 0.5 hour per data point.
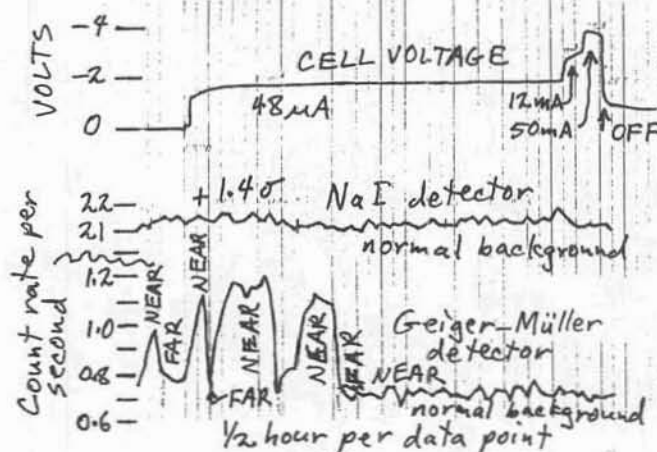
This type of experiment was reproduced many times. Note that the less sensitive detector will detect a smaller radiant intensity than a more sensitive detector, because the less sensitive detector discriminates against background better. For a discussion of the Minimum Detectable Activity with respect to environmental background see: D. A. Gollnick.[5] The background at this location in the lab is well characterized.

Cathode analysis by Secondary Ion Mass Spec. (SIMS) must fail as a nuclear products analysis, because it does not analyze the bulk, and SIMS is prone to artifacts. Briefly, material from the surface of the cathode is sputtered by an ion beam, the secondary ions from the cathode surface are then analyzed by the mass spectrometer. Unfortunately, the ionization cross section of various elements is vastly different so that elemental quantification is relatively inaccurate. Even the isotopes of a single element suffer from ion fractionation,[6] and in dynamic SIMS the extent of ion fractionation changes with sputter hole depth.[7] Furthermore, it takes a considerable amount of instrumentation time to obtain meaningful precision. Large secondary ion intensities can overwhelm the detector, wildly skewing the isotope ratio observed; this is detector dead time, and has been particularly troublesome with respect to easily ionized elements, like lithium (^{6}Li:^{7}Li). A more insidious dead time effect occurs when spurts of ions overwhelm the detector, and again skew the isotope ratios. Although SIMS and other surface techniques (e.g. EDX and XPS) must fail for decisive nuclear products analysis, much useful information can be gained by their careful application.

Prompt Gamma Activation Analysis (PGAA) using a cold neutron beam is the method of choice; it provides a high sensitivity isotope specific bulk analysis, and is complemented by Neutron Activation Analysis (NAA). The cold neutron beam is essential, because it allows analysis far removed from the reactor cores' gamma ray field Fig. 6.[8] The cold neutrons also have a higher capture cross section, enhancing sensitivity. PGAA with thermal neutrons is futile due to the interference of the reactor cores' gamma ray field.

Figure 6

Liquid Neon Heat Pipe
Neutron Guide
Reactor core
Cold Moderator
Sample
Germanium γ-ray Detector
Helium Chamber
Lead Cave
^{6}LiF Neutron Shield

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Thermal neutrons are moderated by liquid neon cooled 1,3,5-trimethylbenzene before being guided down the beam channel by total internal reflection off the ^{58}Ni coated silica walls, just like fiber optics. The cold neutrons expose the sample housed in a helium filled chamber, captured neutrons cause the sample to emit γ-rays characteristic of the isotope. The germanium γ detector of the spectrometer is housed in a lead cave, sheathed with ^{6}LiF sheets to adsorb scattered neutrons radiationlessly; thus effecting a very low background system.

Cold neutron beam PGAA represents the state-of-the-art. There are only two operational installations known in the USA.

Conclusions: The heat versus helium analysis is reproducible and appears to indicate that the Pons and Fleischmann effect induces the D + D --> ^{4}He + 24MeV reaction pathway (Figs. 1 and 2), by an unknown mechanism. Finding helium in the gas phase suggests that the reaction occurs at the surface of the cathode. Helium born within the cathode would be lost to analysis, and indeed isolated incidents of excess heat generation without helium production have been observed. Future helium analysis will investigate the branching ratio using state-of-the-art ^{3}He:^{4}He:^{20}Ne analysis and Seebeck calorimetry.

Rigor with regard to calorimetry is the experimental foundation in this field. Seebeck calorimetry is used because it is the most fundamentally rigorous method known, amounting to high performance Calvet calorimetry. Further, Seebeck calorimetry provides a complementary method, validating our isoperibolic results.

The solid state physics that allow radiationless reaction remains to be elucidated (See for example Y. Kim). Radiation, though occasionally observable, is not related quantitatively to excess heat. The minuscule amount of radiation intermittently observed suggests an efficient energetic coupling between the nuclear reaction and the metal lattice. The breakdown of the energetic coupling mechanism might result in bremsstrahlung,[4] and hence a continuum spectrum superimposed with characteristic peaks;[3] while efficient energetic coupling results in excess heat.

Isotope specific analysis of the cathodes by PGAA[8] is being pursued in conjunction with NAA and SIMS[6,7] to search for nuclear products in the cathode.

Outlook: A fundamental understanding of the solid state physics that results in the Pons and Fleischmann effect is to be gained through nuclear products analysis. Understanding the mechanisms will allow technological development.

References:
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