BRIGHAM YOUNG
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THE GLORY OF GOD
IS INTELLIGENCE

October 11, 1994

Dr. Walter M. Polansky, Director
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Office of Basic Energy Sciences, ER-16
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Washington, DC 20585
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Dear Dr. Polansky,

This morning I received a telephone call from Russ George; we
discussed experimental results obtained by him and Roger
Stringham in which helium-4 and helium-3 have been found in
evidence. I have also discussed these claims with Nate Hoffman
of Rockwell International where the helium was measured, and he
assures me that the helium isotopes are indeed present. Although
the origin of the helium isotopes is unclear, the claims
certainly deserve attention. Accordingly, Russ and I (with the
concurrence of Dr. Stringham) will propose tests to be conducted
with the Stringham apparatus at BYU, to search for charged-
particles, neutrons, gammas and x-rays, for which we have state-
of-the-art detectors. In particular, our neutron detectors
benefit from years of development, benefitted by guidance from
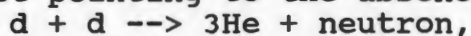John Huizenga, Al Mann, Steve Koonin, Charlie Barnes and others.

As you know, we at BYU are by now quite skeptical of claims of
cold fusion in metals. Our recent paper, "Search for Neutron,
Gamma and X-ray Emissions from Pd/LiOD Electrolytic Cells: A
Null Result," has been accepted for publication in Fusion
Technology and should appear in December. In addition, we
submitted last month two papers to the Journal of Physical
Chemistry: 1) "Examination of claims of Miles et al. in Pons-
Fleischmann type cold fusion experiments", and 2) "Faradaic
efficiencies less than 100% during electrolysis of water can
account for reports of excess heat in 'cold fusion' cells." I
will send copies of these papers in their 'final' form.

With regard to the Stringham experiments, we would express
caution regarding claims of excess heat in a system involving
large power inputs via ultrasound. The calorimetry which we have
seen does not provide compelling evidence for excess heat (EPRI
Proceedings of the Fourth Annual Conference on Cold Fusion); the
researchers would benefit from cooperation from an experienced
calorimetrist like Prof. Lee Hansen of the BYU Chemistry
Department. The fact that the light-water 'controls' show <u>some</u>
excess heat is puzzling to say the least, and the adequacy of
controls is not clear.

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The nuclear-products claims likewise require further scrutiny. How is it that helium-4 and helium-3 are detected in cells which showed no neutron production during activation? Here we are not just pointing to the absence of evidence for reactions such as
    d + d --> 3He + neutron,
but also to the lack of neutrons from secondary reactions, such as photodisintegration of deuterons from gammas which should accompany such reactions: d(gamma,n)p reactions. We have measured such secondary reactions in our deep-underground laboratory facility in Provo Canyon using both gamma and alpha sources, and find that the secondary-neutron production is consistent with calculations. Then why are there no secondary neutrons from the Stringham cells (in experiments conducted at Los Alamos)? So the detection of helium isotopes in these experiments, without concomitant detection of neutrons, is indeed puzzling.

We can look for primary and secondary neutron production with high sensitivity in the Provo Canyon laboratory; our backgrounds are about 1% of those in the best Los Alamos detectors. Indeed, Thomas Claytor and Howard Menlove have come here for high-sensitivity neutron detection in recent years. Charged-particle production can be searched for either via secondary reactions (by adding beryllium foil or salts as a radiator, neutron production by charged particles is greatly enhanced) or by primary reactions using a surface-barrier detector. As usual here, all signals will be digitized using fast-waveform digitizers to facilitate signal analysis and noise rejection.

Gamma-rays, including possible 22-MeV gammas from d + d --> 4He, can be detected using Germanium detectors (two are available here) or NaI detectors (we have a large, 5"-diameter, NaI crystal). We are also equipped to measure x-rays produced via excitation of the metal lattice in which nuclear reactions are purported to occur, using state-of-the-art detectors (SiLi and reversed-biased photodiode detectors).

At present, the Stringham/George results are intriguing yet not compelling. State-of-the-art equipment is needed to ascertain the validity of the claims. To determine whether all the proper controls have been done, one must assess all possible paths for contamination. However, detection of charged-particles having MeV-scale energies would provide unequivocal evidence for nuclear reactions. Hence the suggestion that experiments be undertaken as touched on above.
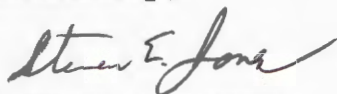
We are willing to undertake such experiments since the likely sources of helium-4 and helium-3 (in particular) have been ruled out already. This approach of combining our detectors with the Stringham/George apparatus is consistent with the recommendations of the DOE/ERAB panel regarding "Cold Fusion Research":

"The Panel is sympathetic toward modest support for carefully focused and cooperative experiments within the present funding

system. ... Cooperative expeiriments are encouraged to resolve some of the claims and counterclaims in calorimetry... If the excess heat is to be attributed to fusion, such a claim should be supported by measurements of fusion products at commensurate levels." (DOE/S-0073, "Cold Fusion Research," November 1989.)

In keeping with these considerations, we are preparing a modest proposal to scutinize the Stringham/George apparatus, in conjunction with these scientists. It should reach your office within a week or so. Your attention to this proposal would be appreciated.

Sincerely,

Steven E. Jones
(for the BYU Laboratory for Fusion Studies)

SEARCH FOR NEUTRON, GAMMA AND X-RAY EMISSIONS FROM Pd/LiOD ELECTROLYTIC CELLS: A NULL RESULT

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Abstract

We have conducted a series of experiments using state-of-the-art neutron, gamma and x-ray detectors to search for evidence for nuclear reactions occurring in Pd/LiOD electrolytic cells. No evidence for primary or secondary emissions from nuclear reactions was obtained in extended experiments.

I. INTRODUCTION

Since the announcement by M. Fleischmann and B. S. Pons of excess heat production via nuclear reactions in Pd/LiOD electrolytic cells[1], we have stressed the need for state-of-the-art detectors to scrutinize these claims. Here we present new results from our most sensitive detectors. We also caution that compelling results can only be obtained with state-of-the-art systems, which we describe.

II. OVERVIEW OF DETECTOR SYSTEMS

Our primary detector for low-level neutron emissions consists of a combination of a large plastic scintillator core with a surrounding bank of sixteen 3He-filled proportional counter tubes (Figure 1), with all signals digitized at 50 Mhz and stored in computer memory. The central plastic scintillator is 35 cm in length and 8.9 cm in diameter. A central cavity of 4.8 cm diameter admits test cells. Fast neutrons from the sample can generate a recoil proton in the plastic generating scintillations (efficiency about 40%) which are viewed by a photomultiplier tube. Then the neutron slows further in polyethylene moderator 28 cm diam. X 30 cm long, and finally may be captured in one of 16 helium-3-filled proportional counter tubes embedded in the moderator (efficiency for 2.5 MeV neutrons is 34%). These tubes are arranged in four quadrants incorporating 4 proportional-counters in each segment.
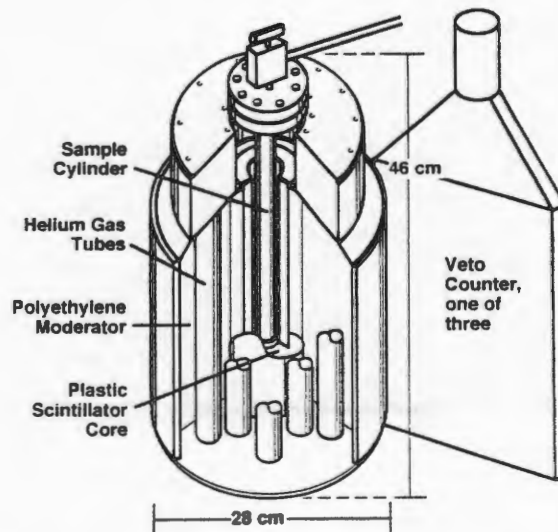

Figure 1. Los-Alamos-type detector with plastic scintillator core detector and cosmic-ray veto paddles (3) added at BYU.

The detector and experiments have the following special features:

1. All signals are digitized using a LeCroy fast-waveform digitizer operating 50 MHz, so that we retain pulse-shape information as well as timing between pulses. Pulse-shape analysis permits excellent noise rejection, along with giving some neutron-energy information (from the prompt plastic scintillator pulse). By rejecting (in software) events having small or no plastic pulses, we strongly discriminate against slow (especially thermal) neutrons. This background-reducing feature is not available to many detectors including those using BF3, 3He and even the Kamiokande detector in Japan[2]. By studying neutron-capture time distributions based on prompt and capture-neutron pulses, we check whether observed distributions agree with those found using a plutonium source.

2. The PC-based data acquisition system records which of the four quadrants of the 3He-type counter showed neutron capture, allowing for checking that the quadrants are hit in equal proportions.

This detector segmentation has, for example, allowed us to throw out apparent large bursts of neutrons

(over 60 "neutrons" in a 320-microsecond window) whose signals unrealistically came from just one quadrant. (Occasionaly two quadrants are involved, due to electronic cross-talk). Even if neutrons are somehow emitted in one or two directions, the polyethylene moderator has the effect of spreading out the neutrons as they slow down, so that detected neutrons of sufficient statistics will necessarily appear in all four quadrants.

We have seen several cases of such large bursts in the past three years of running; but all bursts of over five detected neutrons have proven to be spurious. (see Ref. 3) The large multiplicity "events" are correlated with high-voltage breakdown in the standard electronics of this ^{3}He-type detector. Therefore we retract earlier claims of high-multiplicity time-correlated-neutron events, notably those which appeared to correlate with sample cooling using liquid nitrogen.[4] Compelling data for large neutron bursts require detector segmentation and pulse digitization (allowing signal visualization) as we have done, or other reliable methods of noise elimination.

3. We have added three large cosmic-ray veto counters to show the passage of cosmic rays, which events are rejected off-line. Passive shielding of at least 35 m of rock (12,000 g/cm^2) also greatly reduces cosmic ray-induced events and removes dependence of cosmic-ray rates on fluctuations in atmospheric pressure. After cosmic-ray rejection, the event rate is approximately 0.65 neutron-like singles per hour with an efficiency of 14% for 2.5 MeV neutrons, and 0.07 burst-events per hour with a detection efficiency exceeding 20% (increasing with neutron-burst multiplicity)[5].

4. Two additional highly-sensitive neutron detectors are available in the same deep-underground facility based on a different neutron-capture scheme (capture in lithium-doped glass), to permit checking of any positive results found in the primary detector[5].

III. RESULTS OBTAINED WITH Pd/LiOD ELECTROLYTIC CELLS

A. Experimental Protocols

The neutron data presented below represent 1,054.6 hours (6.3 weeks) of observation of Pd/LiOD cells and backgrounds in our most sensitive neutron detector. Experimental protocols follow those provided by Dr. Thomas Passell[6], for these Pons-Fleischman-type cells, namely:

1. Pd cathodes (6 mm diam. except 4 mm diam rod described in 3 below) were used in a 0.1 M LiOD solution (in D$_2$O). Electrode spacing of the Pd rods relative to Ni-gauze which formed the cylindrical anode is approximately 2 mm, with a septum used to prevent electrical contacts.

2. Three cells were polarized in series at 40 mA from Sept. 24, 1993 to October 25, 1993, then at 80 mA until October 29, 1993.

3. Following a suggestion of Prof. K. Wolf[7], a fourth Pd/LiOD cell was operated at high altitude (8,500') for three weeks at 20mA/cm2, then added in series connection with the other three cells on October 25, 1993.

4. The palladium cathode rods were scraped/sanded approximately every seven days, and replaced in the cells within a period of about fifteen minutes to minimize deuterium loss from the cathodes during the cleaning procedure. We noticed that the cell potential slowly increased over days of (constant-current) operation, then decreased after the cathodes were cleaned, showing that a resistive surface coating had built up during cell operation. We also observed a gradual rise in electrolytic cell temperature, using a platinum-resistance probe, consistent with increased resistance and joule heating as the resistive surface coating developed on each Pd cathode.

5. A 12-hour cooling treatment was applied to the three primary cells on day 17. The fourth cell (described in 2 above) was subjected to diurnal cooling and heating due to its exposure to a mountain environment; the electrolyte was found to be frozen on two occasions.

6. Boron and aluminum (about 0.001 molar) were added to the LiOD electrolyte on the 18th day.[7]

B. Search for Neutron Burst Events.

A neutron burst event is defined as having a hit in the plastic scintillator core followed by two or more signals in the 3He-filled proportional-counter tubes within 320 microseconds. Since the die-away time for neutrons in the outer detector/polyethylene moderator is 55 microseconds, there is a possibility to see multiple distinct neutron hits there. In effect, the outer detector "de-multiplexes" neutrons should an instantaneous burst occur, as first reported by H. Menlove et al.[4]. A burst is then defined as two or more neutrons captured in 3He within 320 microseconds of a start pulse in the plastic

scintillator. The background rate for bursts is (0.07 +- 0.01) n/hr, all from multiplicity = 2 events, established using Pd loaded with hydrogen in 394 hours of separate runs.

We also scrutinize the time spectra of 3He-captured neutrons relative to the start pulse in the plastic scintillator to determine whether the time distribution corresponds to the 55-microsecond die-away time for neutrons in the 3He-portion of the counter, as seen with a plutonium neutron source.

The Pd/LiOD cells described above were polarized for 708.8 hours. During this time, 24 neutron-like burst events were seen, all having multiplicity = 2. (This represents approximately one burst candidate per 30 hours, a very low rate indeed.) Thus, the neutron-like rate for these events was 48/708.8h = (0.07 ± 0.01) n/hr. These numbers are in complete agreement with those found with hydrogen controls discussed above. There was no significant change in rate for neutron-like burst events between background and runs with electrical currents in the Pd/LiOD cells. There is no indication of a neutron burst signal above a very low background.

To complete the scrutiny for burst-like events, we compare time spectra from these Pd/LiOD electrolytic cell runs with those obtained from H2-control runs and from Pu-source runs. Figure 3 displays the time intervals between each start pulse in the plastic scintillator detector and each stop pulse from the 3He-type outer detector.
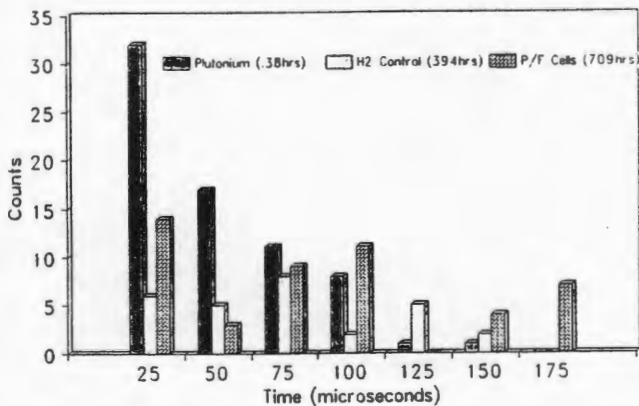

Figure 2. Time intervals between plastic scintillator "start" pulse and ^{3}He-capture "stop" pulse for multiple-neutron ("burst") events. the spectra associated with P/F electrolytic cells and with H_2 controls are consistent and represent background spectra.

The neutrons from the plutonium source follow a

pattern consistent with the calculated 55-microsecond die-away time for neutrons in the counter, but neither the controls nor the Pd/LiOD cells show such a distribution (the latter two spectra are consistent with backgrounds.) We conclude that there is no evidence whatsoever for neutron-burst activity in the electrolytic cells. The upper limit on excess power from nuclear, neutron-generating reactions in the electrolytic cells is at the picowatt level.

C. Total neutron-like count rate

Even though there is no neutron-burst signal, there may still be neutron counts above background which we consider "singles." The background rate for such events has been established as (0.65 ± 0.1) counts/hour using Pd loaded with hydrogen. Figure 3 displays results from each run of the electrolytic cells, showing 1-sigma error bars (statistical only). All of the observed rates are entirely consistent with background levels of 0.65 h^{-1}. This exercise has as its conclusion that no neutrons were seen above very low background levels, in a high-efficiency detector. The most important observation may be that state-of-the-art neutron detectors are now available for studies requiring high-sensitivity instruments.
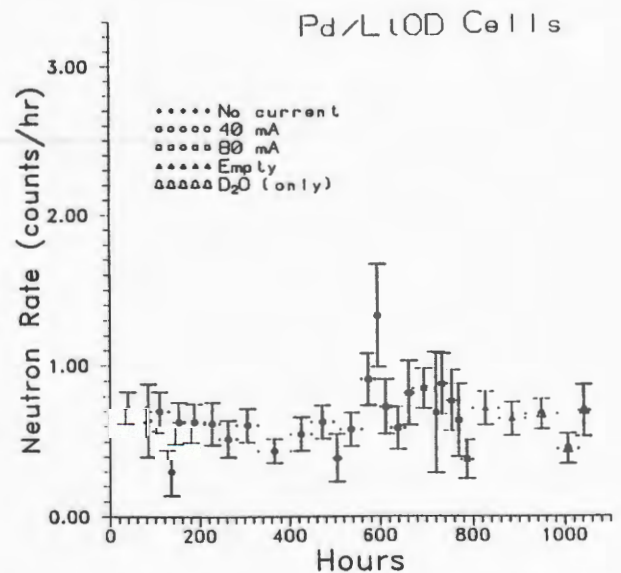

Figure 3. Signal rate in Pd/LiOD cells compared with rates seen for background conditions. Background rates are only 0.7 counts per hour, and there is no evidence of neutron production (above background) in the Pd/LiOD cells.

IV. GAMMA AND X-RAY SPECTROSCOPY

A. Search for gamma rays

Immediately following the neutron search, all palladium rods were taken to Los Alamos for gamma-ray spectroscopic analysis. The purpose of this search was to determine whether radioactive isotopes of palladium, rhodium, ruthenium and silver might have been generated during the electrolytic runs, pursuant to claims of Y. Kucherov and others of such transmutations in deuterium-loaded palladium[7,8]. All four Pd rods were placed in a low-background germanium detector operated by Dr. J. Parker and counted for 75,000 seconds. No gamma lines above background were seen, except for a weak 59.5 keV line which represents americium-241. The americium contamination was traced to the nickel gauze used for anodes. The migration of americium from anode to Pd cathode during operation of the electrolytic cells demonstrates that radioisotopes can be picked up by the cathode originating from either the electrolyte or the anode. Therefore, any claims of nuclear transmutation in such cells must first show that the claimed radioisotopes were not originally present in the electrolyte or the anode. Contamination of the cathode must also be carefully scrutinized.

Further gamma-spectrographic analyses of the palladium cathodes used in experiments at BYU and Kamiokande over the past five years have been undertaken: we found absolutely no evidence for radioisotope formation in any palladium cathodes. Careful scrutiny should therefore be applied to any claims that nuclear reactions produce transmutations in electrolytic cells. In particular, claims that radioisotopes are formed far off the line of nuclear stability should immediately arouse suspicion that materials used in the electrodes or electrolyte could have been contaminated or subjected to irradiation by an energetic particle beam. For example, if palladium-100 is found by gamma spectroscopy, then beam irradiation is likely since negative-Q reactions are implicated.

B. Search for X-rays

We have also followed our own challenge[9] of searching for x-rays as would be expected if nuclear reactions are indeed producing measureable heat in electrolytic cells. Nuclear reactions are characterized by release of MeV-scale energies, hence their importance to power-production schemes. Energy release at the nuclear level implies that secondary x-rays will be produced in the environment of a metal lattice, where only tens of keV are required to generate x-rays. That is, if nuclear reactions are indeed producing heat at the levels claimed (>1 mW), then sufficient x-rays should be produced to be detectable, since x-rays arise from ionizing effects of nuclear products on the materials in which the purported heat develops. Thus, x-ray spectral measurements provide a crucial test for the presence, or absence of heat-generating nuclear reactions.

Characteristic x-rays of Pd (K-alpha of 21.1 keV) or Ni (K-alpha of 7.5 keV) which result from K shell vacancies produced by nuclear products are readily detected. We have searched for such lines using two x-ray spectrometers, a 10mmX10mm silicon detector having high sensitivity down to about 4 keV[9] and a lithium-drifted silicon detector with high sensitivity down to approximately 1 keV. We used a Pd/D2O electrolytic cell in which 25 micron Pd foil formed both cathode and external wall. No x-ray production was seen with this electrolytic cell. Similarly, a search for x-rays was conducted using a Ni/H2O cell in which the Ni cathode was placed against a very thin plastic window. Again, no x-ray production was in evidence in the electrolytic cell. Using a montecarlo calculation to determine the overall x-ray detection efficiency from electrolytic cells[9], we set an upper limit of 10 microwatts of excess power from these cells, from any nuclear reactions which produce secondary x-rays.

In a "search for cold fusion using x-ray detection"[10], M.R. Deakin et al. found, as we have, that no x-rays above background were produced by Pd-LiOD electrolytic cells. An important caveat is provided in that paper: "Room background radiation fluoresces the cathode and Pd K x-rays are therefore present as an artifact of background."[10] Thus, the presence of x-rays alone (including fogging of x-ray film) is insufficient to demonstrate the presence of heat-generating nuclear reactions.

No "cold fusion" experiment anywhere has shown the presence of characteristic secondary x-rays lines (using an x-ray spectrometer) which would characterize fusion or any other nuclear reaction in a metal lattice[11]. There are some experiments that show fogging of x-ray dental film, but such experiments are too crude to provide quantitative information regarding x-ray energies and intensities, and are subject to artifacts.

Thus, we find no compelling evidence to link nuclear reactions to excess-heat production claims. Instead, the lack of significant (primary or secondary) x-rays, gammas and neutrons after five years of searching argues convincingly against claims of excess heat production via nuclear reactions in electrolytic cells (or

equivalent). This conclusion is supported by related experiments at BYU which show up to 750% "excess heat", but which apparent "excess heat" is in fact due to hydrogen-oxygen recombination in the cells coupled with commonly-used (but misleading) analysis techniques for excess-power production in "cold fusion" experiments[12]. Thus, the "excess heat" admits prosaic explanation: it is not nuclear at all.

IV. CONCLUSIONS

In order to find compelling evidence for cold-fusion effects, state-of-the-art calorimeters and nuclear detectors are requisite. Table 1 juxtaposes such systems with other systems which are still more generally in use. It is disquieting that some researchers select open electrolytic cells over closed cells, and excessively long sampling intervals (e.g., 5-minute sampling intervals for input voltage used by Pons and Fleischmann in calculating excess heat over a 10-minute boiling period[13]). Some researchers continue to use x-ray films instead of x-ray spectrometers, helium or tritium gas sampling instead of charged-particle spectrometers, Geiger counters rather than silicon or germanium detectors, and neutron survey meters instead of sensitive neutron detectors as described above. It is time to strongly question claims of cold fusion based on crude techniques and to demand tests at a rigorous scientific-proof level. Compelling evidence requires use of the best instruments available, incorporating fast data-sampling and digitization methods, the use of different detectors whose signals agree quantitatively, and presence of signals well above background levels. A real signal should be capable of scaling, and should not shrink as background levels are reduced.

TABLE 1. COMPARISON OF COLD-FUSION RESEARCH METHODS

It is evident that much of the present confusion surround "cold fusion" stems from the continued use of inadequate detectors. This list juxtaposes crude, better and state-of-the-art systems to promote the quest for compelling data, one way or the other. Use of the best available methods is clearly the path-of-logical science.

Crude (simply add to the confusion)	Better (but not good enough)	State-of-the-art (can provide compelling evidence)
Neutron survey meters BF3	Segmented 3He, Plastic scintillators	Segmented 3He 3He or Li-doped glass *plus* scint. and digitizing
Helium gas detection, Tritium gas	Charged-particle det. (Si surface barrier) (requires thin foil)	Thin dE/dx detectro plus Si spectrometer (particle ID & energy)
X-ray film	X-ray film with energy-filters	X-ray spectrometer (Si,HgI2, CdTe,etc.)
Geiger-Mueller counter	see detectors listed above; Ge detector	
Infrequent I*V(t) sampling (e.g., every 300 s)		Integral I*V(t) correct via frequent, redundant sampling
Open cell calorimetry, no H2/D2 +O2 during experiment	Measure H2/D2 +O2 simultaneous w/heat	Recombiner inside separate calorimeter
Metal of unknown source quality or purity		Alloyed with known purity and properties
D2O of unknown	D2O from known source,	Highly distilled D2O, known H,O source, isotopes
Visual techniques	Computer-logging several probes	Redundant probes with fast data acquisition
Theories which disregard P, E conservation or light-cone constraints (e.g., "heating lattice")	Fractofusion ignoring e- vs. d+ acceleration	???

With these criteria for state-of-the-art detectors, we find that no compelling evidence for neutron, gamma or x-ray production from deuterided materials currently exists in any cold-fusion experiment, including our own. The only verified form of cold nuclear fusion to date is muon-catalyzed fusion. Nevertheless, having an obligation to resolve remaining issues[14], we will continue our search for several more months. We invite those with evidence for neutron production to accept our invitation to test their systems in the deep-underground neutron detection facility in Provo Canyon in order to confirm results. Gamma and x-ray spectrometers are also available on request.

ACKNOWLEDGMENTS

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EXAMINATION OF CLAIMS OF MILES ET AL. IN PONS-FLEISCHMANN TYPE COLD-FUSION EXPERIMENTS

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Abstract

In cold fusion experiments conducted at the Naval Research Laboratory in China Lake, M. H. Miles and co-workers claim to have produced excess heat correlated with helium-4 production, x-rays, and Geiger-counter excitation. However, scrutiny of the claims shows that unreliable calorimetric and nuclear-product detection schemes were used. Moreover, inconsistencies and errors are found in the data and data analysis. The juxtaposition of several poor techniques and inconsistent data does not make a compelling case for cold fusion. We conclude that the evidence for cold fusion from these efforts is far from compelling.

1

This paper scrutinizes claims of excess heat, x-rays, Geiger-counter excitation, and helium-4 production in cold fusion experiments by M.H. Miles and co-workers [1–8]. It is specifically written in response to a request from M. H. Miles: "I hereby challenge Professor Jones to take his allegations regarding my work to a refereed scientific journal. ... I hope this matter can thus be resolved without any further actions." [9]

Miles et al. claim:

"Our electrochemical experiments unambiguously show a direct correlation between the time of generation of excess enthalpy and power and the production of ^{4}He...This correlation in the palladium/D$_2$O system provides strong evidence that nuclear processes are occurring in these electrolytic experiments and that helium is produced.... In summary, nuclear events with ^{4}He as a major product occur during the electrolysis of the Pd/D$_2$O + LiOD system." [2]

These bold claims warrant scrutiny, especially since these are the only published claims for concomitant excess power, helium-4 and x-ray production. Careful examination of the papers shows that unreliable calorimetric and analytical methods were used, incorrect statistical procedures were applied, and there are inconsistencies in the results. Thus, we find these experiments to be ambiguous and the claims of "excess heat" and excess heat/nuclear product correlations to be invalid.

I. CLAIMS OF HELIUM-4 PRODUCTION IN ELECTROLYTIC CELLS

Figure 1 displays data tables published in 1991 [3] and 1993 [6]. Published plots of helium-4 data are shown in Figure 2. Inspection of the tables in Figure 1 shows that the same data are involved. Note that values of ^{4}He atoms/500 mL fall at 10^{12}, 10^{13}, and 10^{14} in Table 1. The lack of values between integer exponents implies a crude estimation procedure. Indeed, the authors state:

"Small peaks near the detection limit of the mass spectrometer are assigned a value of 10^{12} atoms of ^{4}He per 500 ml of effluent gases. Medium peaks were roughly an order of magnitude greater while large peaks were about two orders of magnitude above the detection limit." (Emphasis added.) [3]

A striking change occurs in the same data presented in 1993; the values of ^{4}He atoms/500 mL are tenfold greater and now in putative agreement as to the order of magnitude of the ^{4}He rate required to account for the claimed excess heat rate, i.e. about 5×10^{14} atoms/4440 s [2]. Such an amplification diminishes confidence in the claims. Note also the carelessness of footnoting which leads to an uncertainty in which runs had a current density of 250 mA/cm^2 instead of 200 mA/cm^2 and which are subject to calorimetric error.

2

3

Comparison of Figures 2a and b also shows differences in presentation of the same data. In reference [3] five points are displayed at the "large peak" value whereas in reference [6] only four are shown. In reference [6] two values are plotted each at "medium," "small" and "no peak" positions, whereas in reference [3] there is only one point at the "medium" and four points at the "no peak" position. The plot in reference [3] does not agree with the table from reference [3].

An additional data point showing some He in a N_2 filled control flask appears in the plot from reference [6]. Neither plot shows all of the data points from reference [4] which discloses significant amounts of He in N_2 filled flasks. Two of ten flasks gave "large" amounts of ^{4}He and two gave "small" amounts.

In evaluating the statistical significance of their results, the authors state in 1991:

"Ignoring the helium/heat relationship (Table 1), the simple yes or no detection of helium in 7/7 experiments producing excess heat and the absence of helium in 6/6 experiments not producing excess heat (1 in D_2O, 5 in H_2O) implies a chance probability of only $(1/2)^{13} = 1/8192$ or 0.0122%." [3]

Using the same data set, the authors state in 1993:

"Ignoring the helium-heat relationship (Table 2 in Figure 1), the simple yes or no

detection of helium in 8 out of 8 experiments producing excess power and the absence of helium in 6 out of 6 experiments not producing excess heat (1 in D_2O, 5 in H_2O) implies a chance probability of only $(1/2)^{14} = 1/16384$ or 0.0061%." [6] (Essentially the same wording appears also in ref. [2].)

In addition to obvious discrepancies in the two statements, the helium-heat relationship cannot be ignored. It is crucial to the claims of unambiguous correlation between excess power and ^{4}He production. The data show the obvious problem that runs producing more "excess heat" do not always show more "helium production," so that quantitative agreement between heat and helium generation is lacking. This is a warning that must not be ignored. If ^{4}He is detectible at 10^{13} atoms/500 mL, why can't it be quantitated at a concentration two orders of magnitude higher?

Another way of viewing the ^{4}He data is that 8 of 10 experiments done in 1990 showed ^{4}He to be present while 0 of 5 experiments done in 1991 showed ^{4}He to be present. Recent experiments likewise show little if any ^{4}He. [8] Are they simply getting better at keeping ^{4}He out? To be valid, controls must be interspersed randomly in time with non-controls, or even better, run in parallel. N_2 filled flasks showed diffusion of $3.2x10^{12}$ atoms of ^{4}He/d $- 19x10^{13}$ atoms/60d, sufficient to explain the results. Miles has argued that H_2 (or D_2) prevents diffusion of ^{4}He into the flasks [4]. But studies at Rockwell showed that atmospheric helium enters glass flasks even when hydrogen is present inside the flask [N. Hoffman, private communication].

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Moreover, both arguments of statistical significance cited above leave out run 12/17/90-B which shows more heat than three other supposed heat-producing runs, but which shows no helium-4 at all. Clearly, including this run would dramatically decrease the purported significance of the excess-heat/helium production claims and remove completely the alleged one-to-one correspondence of heat and helium production. There is no apparent justification for including run 12/17/90-A in the calculation in 1993 (thus improving the alleged statistical significance over the 1991 calculation) when run 12/17/90-B, which had the same problem of possible calorimetric error, was excluded.

The statistical significance of the data is meaningless without information on associated uncertainties and if all runs are not treated equally. These steps were clearly not taken. The lack of agreement between the amounts of helium detected and the amount of excess heat generated must be reconciled with the alleged one-to-one correspondence between excess heat and helium-4 production. Finally, the observation of helium-4 in 4 out of 10 nitrogen-filled control flasks [4] must be included for consistent and fair treatment of the data.

Much of the above could be argued to be mistakes in presentation (which we all are prone to), but data purporting to "unambiguously show" [2] a significant new phenomenon such as "cold fusion" must meet a higher standard than is present in Miles' papers.

Contamination from atmospheric helium-4 cannot be strictly ruled out because the concentration of helium-4 detected never exceeds that in the laboratory air. Miles et al. acknowledge that "possible error sources proposed by cold fusion critics include air contamination, helium diffusion into the glass flask, and the escape of helium contained in the palladium rod." [3] Escape of helium contained in the glass must also be considered, an effect which led Paneth and Peters to their erroneous claim of helium production in hydrogen-loaded palladium nearly seventy years ago. [10] Paneth and Peters retracted their claims of helium production in 1927 [10], as did Pons and Fleischmann in May 1989 [11].

With such small helium concentrations observed, a prosaic origin of the helium should be suspected. Helium-4 is prevalent in the laboratory environment, as Miles and co-workers admit: "because of the use of helium-cooled nuclear magnetic resonance instruments and helium-filled glove-boxes in the building, the helium content of the laboratory air can be significantly higher than 5.22 ppm." [4]

In light of these uncertainties, we remain unconvinced that the published data are strong enough to support the conclusion, "our electrochemical experiments unambiguously show a direct correlation between the time of generation of excess enthalpy and power and the production of 4He" [2].

Two very recent papers [6, 12] have attempted to answer some of the above criticisms.

One of these papers [6] now claims an even higher detection limit for ^{4}He of 3.10×10^{13} atoms/500mL in the glass flasks used in previous studies. The other study [12] used 500mL metal flasks to collect electrolysis gases. Five control experiments, four in D_2O + LiOD and one in H_2O + LiOH, all run at 500mA "yield a mean value of 4.4 ± 0.6 ppb" ^{4}He with extreme values at 4.9 and 3.4 ppb. The paper states that "no excess power was measured" for these "controls", but no values for excess powers with attendant error estimates are provided to substantiate this claim and to allow comparison with experiments where a small excess power is claimed. (Thus making the latter non-controls.) Note that designation of an experiment as a "control" is done after the experiment is run, not before. Gases were collected and analyzed for five experiments in D_2O + LiOD electrolyte that produced putative excess heat rates from 30 to 60 mW and excess ^{4}He ranging from 1.0 ± 1.6 to 5.3 ± 1.3 ppb. The four of these experiments producing the highest excess heat rates and excess ^{4}He were run at 400 mA. Only one was run at the same current as the controls, and that run produced both the lowest excess heat rate and excess ^{4}He. Neither result is significantly different from controls. Since the excess heat rate and excess ^{4}He for the other four experiments are not positively correlated, the hypothesis of a correlation between excess heat rate and excess ^{4}He remains unproven. Furthermore, the amount of excess ^{4}He found is less than 1% of that required to account for the excess heat by "cold fusion." Also, as shown later, the error in the excess heat rate measurement is much greater than the 20mW assumed in ref. [12], and therefore none of the excess heat rate data are significantly different from zero. If the excess heat rate is invalidated, then the claimed qualitative correlation

between positive observations of excess heat and excess ^{4}He must also be fortuitous, spurious or covariant. The data treatment by which Miles arrives at the conclusion that the probability of his hypothesis being wrong is only 1/134,217,730 is severely flawed because of the unsubstantiated data selection.

II. CLAIMS OF EXCESS HEAT PRODUCTION

In their critique of experiments by Lewis, Williams, Albagli, Wilson and others [1], Miles et al. challenge "questionable cell calibration procedures" and varying calorimetric cell constants. But the same problems are present in their own experiments.

Miles et al. used the following equation to evaluate excess enthalpy [2]:

$$X = \text{power out/electrolysis power} = K\Delta T / I(E - E^\circ_H) \qquad (1)$$

where K is the calorimetric cell constant, ΔT is the temperature difference, E is the cell voltage, E°_H is the thermoneutral potential and I is the current through the cell. Excess power (enthalpy) is claimed when $X > 1$. Open cells were used with an assumed thermoneutral potential of 1.53V for the D_2O cells and 1.48V for the H_2O controls.

Published results are presented in Figure 1. The highest excess power, 0.46 watts, was observed on 21 October 1990. Since that date nearly five years ago, Miles et al. have not

been able to reach even this low level of excess power [6,8,12]. Moreover, the first 12 days of data for this run have not been published, although the same (partial) data plot has appeared in several publications [2-6] and we have asked for disclosure of all the data. Disclosure of these data is important since X may have been < 1 early on, showing energy storage in the cell, so that the net total energy for the run may be consistent with zero. Figure 6 of ref. [5] shows just such a situation with light water cells where a negative excess heat was observed for the first 12-16 days.

The calorimetric cell "constants" reported in 1990 [5] show significant variations: 0.138 to 0.145, 0.132 to 0.138, 0.133 to 0.137, and 0.135 to 0.141 for the four cells. Moreover, calibrations were done before and after long runs, not during runs. It is not valid to claim an accuracy based on the standard deviation of these values, since at any time the applicable value may be at an extreme. Thus, the spread of about 4.5% is more descriptive of the ability to accurately measure an absolute heat rate at any given time. Also, different thermistors gave calibration constants differing by 5%. Which is the accurate value? If the X value is moved from 1.00 ± 0.05 to 1.05 ± 0.05, 4 of the 8 claims of excess heat would disappear. The results for H_2O (blank) cells given in figure 6 of ref. [5] show positive excursions of X as large as 15% and negative excursions as large as -20%. Why aren't these considered as significant as the smaller excursions observed with D_2O cells? Although the long-term mean of X is different for H_2O and D_2O cells, this difference is meaningless because of fluctuations in the calibration constant. Miles's results simply illustrate the problem of sorting calorimetric errors from real effects in a poorly designed calorimeter. Since large fluctuations in X are found for both H_2O and D_2O runs in the 1990 data [5], the authors should have evaluated K values frequently during the runs in order to verify stability. This was not done.

There are numerous other problems with the calorimetric measurements. The calorimeters used by Miles are isoperibol (i.e. constant environment) heat conduction calorimeters. The principle of heat measurement in this type of calorimeter is Newton's law of conductive heat transfer, i.e. the rate of heat flow to or from the calorimeter must be directly proportional to the temperature difference between the calorimeter and the surroundings. Measurement of the temperature difference across the heat flow path between the calorimeter and surroundings and proper calibration thus provides a means for measurement of heat flow rate. The design criteria required to obtain accurate results with heat conduction calorimeters were developed more than fifty years ago by Tian, Calvet, and co-workers [13] and have since been refined and polished by many other workers. The most fundamental design criteria is that all heat leak paths between the calorimeter and surroundings must have constant thermal conductance. A second criteria required for accurate operation of the calorimeter is that as large a fraction of the heat as possible must pass through the path on which the temperature difference is measured, and further, this fraction must remain constant. Miles' calorimeters meet neither of these criteria.

Miles et al. (1) describe two calorimeter designs, the first consisting of "a Thermos flask

10

11

(Model 3700) containing the electrochemical cell as well as added insulation," and the second, claimed to be more accurate, "consists of a polyethylene bottle (d = 7.5 cm) fitted with a large glass tube (d = 3.1 cm) and packed with insulation. The electrochemical cell (d = 1.5 cm, L = 15 cm) was positioned within the large glass tube which contains water that serves as a heat-transfer medium. The temperature inside the calorimeter is measured to within ±0.01°C by two thermistors positioned at different levels on the surface of the electrochemical cell." The constant temperature bath around the calorimeters is described only as "B. Braun Thermomix Model 1460 .. set at 27.50°C" with no indication of how stable the temperature is or if there are temperature gradients present in the bath. Thus, we do not know how stable this half of the temperature difference measurement is, nor do we know the stability of the heat conduction paths to the bath. The other half of the temperature difference is measured with the thermistors in the water filled "gap" (Miles' terminology). The use of thermistors, which are self-heated sensors, in an unstirred liquid will give rise to temperature drifts because of temperature gradients in the water, and may lead to occasional large temperature excursions if the temperature inversion in the water (admitted to in an early paper [5]) is discharged by convection.

The heat leak paths from Miles' calorimeters are neither well-defined nor constant with time or cell conditions. Miles et al. (1) recognize part of this problem when they state "the level of the electrolyte exerts a major calorimetric effect .... This effect limits the accuracy to about ±10% in our studies using the Dewar-type cells" and "heat flows out of the top of the cell as well as into the constant-temperature bath." The calorimeter design with the water filled gap is an attempt to ameliorate this problem. However, there are multiple heat leak paths that carry major fractions of the heat transferred in both designs. Furthermore, these paths have greatly different time constants. Miles et al. (1) recognize part of the consequences of this problem in their conclusion that "there is never any steady state for either the cell temperature or the cell voltage," but fail to quantify the consequences in the estimated accuracy of their heat measurements. Accurate heat rate measurements by heat conduction calorimetry can only be made in a steady state unless corrections are made for the time response of the system. Such corrections, which can be very large, can only be made if all significant time constants are known and the system is properly calibrated. Miles et al. estimate a single time constant of 25 to 30 minutes for their system (1,2), but it is clear from the details of the design that there must be several more, some of which must be several hours long; see figure 2 in ref. [1].

There are numerous other potential problems with the calorimetric measurements described by Miles et al. (1-8). For example, in none of these publications is the calibration heater fully described, other than that it is a "20 ohm resistor" (5). There is no indication of any precautions taken to ensure complete delivery of heat from the heater to the calorimeter, to ensure negligible heat generation in the heater leads, or to ensure accurate voltage and current measurements. Details of the calibration procedure are likewise not given. How long was the heater run? What does the calorimeter

response look like during calibration runs? Was the expected heat output from electrolysis ever duplicated with the heater? Thus, the calorimetric measurements of Miles and co-workers are far from compelling in their accuracy.

It is interesting to note the absence of reports of excess heat by workers using commercially available calorimeters with proven designs. Several brands of heat conduction calorimeters suitable for "cold fusion" studies are available. In their calorimeter designs Miles and co-workers choose to ignore more than fifty years of accumulated wisdom concerning how to make accurate heat measurements. It is disturbing to see publication of a paper [1] containing such fundamental errors and misconceptions about a subject as old and fundamental to science as calorimetry in a mainstream journal like J. Phys. Chem.

Another possible source of error in the papers of Miles et al. lies in the tacit assumption embedded in equation (1) that there are no reactions of the deuterium and oxygen gases produced in the cell, i.e., that Faraday efficiency is 100%. In the denominator of equation (1), the term $I\,E^{\circ}_{H}$ represents the electrical power consumed in decomposing the D_2O into D_2 and O_2. In subtracting this power from the input electrical power, it is assumed that there is no recombination of D_2 with O_2 by any mechanism. If recombination occurs in the cell, this provides a prosaic source for excess heat. Miles and co-workers justify the assumption of 100% Faraday efficiency thus:

"There is no evidence for any recombination when a palladium rod cathode is used that is fully immersed in the D2O solution." [9]

In a more recent paper, Miles et al. [1] acknowledge the problem of current efficiency by including a factor in equation (1) to correct for a current efficiency less than 1.

"The current efficiency for D_2O electrolysis (γ) should always be measured to substantiate any claims for excess power." [1]

Their only published claim to have measured current efficiency states,

"Actual measurements of the gas evolution rate by the displacement of water yielded 6.75 ± 0.25 ml/min for cell A and 6.69 ± 0.15 ml/min for cell B. These data add to the substantial evidence that excess enthalpy effects cannot be explained by the recombination of D_2 and O_2 gases within the cell." [2]

But Miles et al. would have had to measure the amount of recombination in each experiment in order to truly ascertain if any "excess heat" were present. Clearly, this needs to be done during the time period when excess heat is claimed, not in separate tests. Furthermore, if the stated error limits on gas evolution rates are one standard deviation, then there is still a high probability of a single measurement having up to 10% of the gases recombined. Recombination is thus a possible explanation for X values

14

15

between 1.0 and 1.1 simply based on the statistical distribution of the measured data.

In another paper Miles purports to have avoided recombination because "both the anode and cathode leads were covered with heat shrinkable Teflon tubing to prevent exposure of the bare metal to the gases in the headspace." [3] But this does not solve the problem. D_2 and O_2 both diffuse rapidly through Teflon, so that recombination could indeed have occurred on the leads in the cell headspace. Moreover, recent experiments at BYU clearly demonstrate that recombination does occur even when both electrodes are fully immersed in the electrolyte. [14] This has been shown for both Ni/H_2O, K_2CO_3/Pt and Pd/D_2O, $LiOD/Pt$ cells. Indeed, we have found up to several hundred percent "excess heat" as calculated by equation 1, which went to zero when recombination was inhibited. By comparison, Miles et al. observed a maximum of 27% excess heat. Thus, apparent excess heat can be obtained when recombination is not carefully excluded during electrolysis runs. [14] Where is the evidence that recombination is not occurring during periods of excess heat generation?

Based on Miles's data, it simply cannot be claimed that excess heat arises from anything other than calorimetric errors and reduced Faraday efficiency.

III. CLAIMS OF NUCLEAR-PRODUCT DETECTION

The nuclear-product detectors used by Miles et al. are described as follows [2]:

"A neutron survey meter (Ludlum model 15) was always kept close to the water bath containing the two electrochemical cells. A Geiger-Mueller (GM) alpha-beta-gamma detector with a thin end window (Ludlum model 44-7) was positioned about 20 cm from the tops of the electrochemical cells and connected to a scaler ratemeter (Ludlum model 2200) and a printer (Casio HR-8A). Dental X-ray film (Kodak ultra-speed, DF-58) placed near the outside of the electrochemical cells was also used to detect any radiation." "Indium (d=0.25 mm, 22 g) and gold (d=0.5mm, 14 g) foils mounted at the surface of the electrochemical cells were used in attempts to detect any sustained neutron emission rates of 10^4 neutrons/s or higher."

None of these detectors provides particle identification or accurate energy information. Use of such crude detectors does not provide the level of scientific proof required to establish "cold fusion." As we shall see, there are also unexplained discrepancies in the data from these instruments.

Miles et al. provide data from their Geiger counter in a recent paper [6]. The mean background rate was stated to be $31,296 \pm 275$ counts per 12-hours, with a maximum of approximately 38,000 counts per 12 hours observed on December 15, 1990. This maximum is calculated to be 27 standard deviations, i.e. $(38,668 - 31,296)/275 = 27$, above background. However, this calculation clearly fails to account for statistical error

16

17

in the single data point at 38,668 counts/12-hrs: it is misleading to consider statistical error in the background rate alone. Moreover, their claim of 27-sigma significance fails to account for large systematic fluctuations expected with a Geiger counter. Thus, the claim in their paper of a 27-sigma effect is clearly inflated.

If the data point of 38,668 counts/12-hr-period is taken at face value, an extremely large total source radiation is implied. A maximum Geiger counter rate of (38,668 – 31,296)/12-hrs., or approximately 600 counts per hour, was reported. The counter was located 20 cm from the electrolytic cells, giving a geometrical acceptance factor of approximately 0.006. Since the neutron detector showed no signal [6], and 20 cm through glass, D_2O-electrolyte and air is too far for betas, alphas or other charged particles, only gammas are left to excite the Geiger counter. The intrinsic efficiency of a Geiger counter for gammas is roughly 0.001. Combining geometric and detection efficiencies with the observed rate, a source rate of 10^8 gammas per hour is found, even without accounting for attenuation. This rate is sufficient to produce counts in the safety monitor (Ludlum model 15) since it is sensitive to gammas at this level, but this detector (Miles states) showed no response at all [6]. Thus, there is a discrepancy between the two counters that casts doubt on the reality of the "signal" in the Geiger counter.

Compelling evidence for gamma-ray emission requires data from a reliable (e.g., germanium) gamma spectrometer, rather than a Geiger counter. Even so, Geiger counter data would be more persuasive if more than one detector had shown signals simultaneously, or if lead and other filters had been placed between detector and cell when an apparent signal was evidenced. But these obvious steps were not used to test the validity of the signal.

Sensitive neutron detectors are available [15], but were not used in the studies by Miles and co-workers. In any case, no significant neutron signals, including secondary neutrons, were found by Miles et al. [2]

In an effort to detect X-rays or other radiations, Miles et al. used dental x-ray film. [2,3] The results are not convincing because artifactual fogging can be caused by mechanical pressure and various chemical vapors. [15] Hence, data from x-ray spectrometers (primarily germanium and SiLi detectors) is considered requisite evidence for x-rays; yet no cold-fusion experiment anywhere has produced an x-ray spectrum showing characteristic (Pd or Ni, etc.) lines. [15] These null results are compelling since any nuclear process occurring in a metal at rates sufficient to produce excess heat must also generate copious x-rays due to excitations in the metal lattice [15]. In 1993, we offered Miles and co-workers the use of one of our sensitive x-ray spectrometers (one of which is readily portable and could be used in his own laboratory). Unfortunately, he did not respond to our offer.

V. THEORETICAL CONSIDERATIONS

18

19

The authors use the following reaction as a "basis for an estimate of helium production" [2]:

$$^2D + ^2D \rightarrow {}^4He + 23.8 \text{ MeV (lattice)},$$
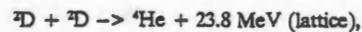
i.e., the 23.8 MeV of energy released in this d-d fusion reaction is assumed to be absorbed by the palladium metal lattice. But there are constraints imposed on transfer of energy to the lattice without formation of energetic particles. Following a nuclear reaction, the energy released can be transferred a distance (R_{max}) limited by the speed of light (c) and the uncertainty principle (ignoring small factors):

$$R_{max} = ct = \hbar c/E = 197 \text{ MeV-fm}/23.8 \text{ MeV} = 10^{-14}\text{m} = 10^{-4}\text{A}.$$

Note that E is of the order of MeV for nuclear reactions, specifically 23.8 MeV for the fusion reaction posited by Miles et al. [3] Since the metal lattice spacing is more than an angstrom, the nuclear energy cannot be transferred to the lattice as hypothesized without violating speed-of-light constraints. Furthermore, conservation of momentum requires that most of the energy be carried by the lighter particle rather than by the lattice, as indeed is observed in the Mossbauer effect.

In muon-catalyzed fusion experiments — the only verified form of cold fusion — d-d fusion produces $t + p$ or $^3He + n$ in nearly equal proportions; ^4He is not detectable (branching ratio $\approx 10^{-6}$). [16] Yet Miles and co-workers claim that ^4He is produced in the absence of detectable tritium, ^3He or neutrons. [2-6] Hence, their claim is inconsistent with the results of experiments involving real, muon-catalyzed cold fusion.

In another theoretical foray, Miles states that:

"For materials at very high pressures, theoretical equations suggest that cations lose their charge at 10^{17} atmospheres and unite with electrons in the plasma with emission of neutrinos. Since this would neutralize the charge of deuterons, the coulombic barrier would collapse and fusion could readily occur." [17]

Miles overlooks major problems with his explanation. (a) A pressure of 10^{17} atmospheres is not reached in palladium in an electrolytic cell, and (b) should an electron capture occur on a deuteron with release of a neutrino as Miles hypothesizes, there are no longer two deuterons present so that d-d fusion obviously cannot occur.

CONCLUSIONS

In response to a request by Miles et al. we have reviewed their published papers purporting to show ^4He, x-ray and excess power production in Pons-Fleischmann-type electrolytic cells. We find the data do not support the conclusion that a nuclear reaction

is the source of the putative excess heat. The claimed correlation between ^{4}He and excess heat is weak to nonexistent and qualitative conclusions are overstated. Putative excess heat observed could be the result of calorimetric errors and recombination of D_2 and O_2 within the cell. Nuclear detection methods are far from adequate and the claimed results are inconsistent. Miles et al. treat within-experiment data as independent observations, thus making it appear that they have much more data than were actually obtained. Far too few control experiments have been done or done properly. Without proper and sufficient controls it is not possible to make the causal inferences Miles et al. have made.

Recent remarks of Miles et al. support our conclusion that excess heat and helium production in Pons-Fleischmann-type cells have a prosaic origin:

"Reproducibility remains a major problem in defining these effects." [6]

"Because helium is present in the atmosphere (5.22 ppm), it is difficult to convince everyone that the ^{4}He measured in the electrolysis gas is a product of a fusion reaction within the cell. It is indeed a very challenging experimental problem to clearly establish the production of ^{4}He from Pd/D_2O electrolysis cells. This situation is compounded by difficulties in obtaining large excess power effects in these experiments." [8]

Clearly, there is no compelling evidence that cold fusion is the source for "excess power"

observed in electrolytic cells. Both the excess power and ^{4}He observed by Miles and co-workers are probably experimental artifacts. The dental X-ray films and Geiger counters likewise are crude detectors and do not provide compelling evidence for nuclear reactions occuring in electrolytic cells. Because many of the criticisms of Miles' work are also applicable to others' claims of excess heat in Pons-Fleischmann type "cold-fusion" experiments, we do not find the data to be compelling proof of any nuclear phenomenon in spite of the number of positive claims for excess heat that have appeared in the literature.

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Figure captions.

Figure 1. Reprinted by permission from [3] and [6]. Juxtaposition of tables published in 1991 [3] and 1993 [6] by Miles et al. showing data from the same experiments. P_{EX} is calculated excess power and X is the ratio of measured calorimetric heat rate to calculated heat rate. Discrepancies are discussed in the text. The tenfold inflation of helium-4 detected from 1991 to 1993 is based on a "revised detection limit of approximately 10^{13} atoms/500 mL." [6] Note that no estimates of statistical or systematic errors are provided.

Figure 2. Reprinted by permission from [6]. Plot showing data points representing amounts of helium measured versus the storage time interval between gas collection and helium analysis. Note that there are no data points which lie between 10^{14} and 10^{15} helium-4 atoms/500 mL. The straight-line fits provided by Miles et al. to these scattered points with the quoted 3-figure precision of the correlation coefficients are a bizarre application of statistics. Note the absence of error bars. The same figure is given in ref. [3] except for addition of 1 point and deletion of 1 point.

26

1991

Table. 1. Helium Production During D_2O Electrolysis.

Sample	P_{EX} (W)	X	^{4}He Atoms/500 mL
12/14/90-A	0.52a	1.20a	10^{14} (large peak)
10/21/90-B	0.46	1.27	10^{14} (large peak)
11/25/90-B	0.36	1.15	10^{14} (large peak)
11/20/90-A	0.24	1.10	10^{13} (medium peak)
11/27/90-A	0.22	1.09	10^{14} (large peak)
10/30/90-B	0.17	1.12	10^{12} (small peak)
10/30/90-A	0.14	1.08	10^{12} (small peak)
10/17/90-A	0.07	1.03	$<10^{12}$ (no peak)
12/17/90-A	0.40b	1.19b	10^{13} (medium peak)
12/17/90-B	0.29b	1.11b	$<10^{12}$ (no peak)

a i = 250 mA/cm^2. All other experiments used i = 200 mA/cm^2.
b Possible calorimetric errors due to low D_2O solution levels. No ^{3}H was detected. Mass spectrometer always at highest sensitivity.

1993

Table 2. Helium Production During D_2O Electrolysis: Revised Detection Limits.

Sample	P_{EX} (W)	X	^{4}He Atoms/500 mL a
12/14/90-A	0.52b	1.20b	10^{15} (large peak)
10/21/90-B	0.46	1.27	10^{15} (large peak
12/17/90-A	0.40b	1.19b	10^{14} (medium peak)
11/25/90-B	0.36	1.15	10^{15} (large peak)
11/20/90-A	0.24	1.10	10^{14} (medium peak)
11/27/90-A	0.22	1.09	10^{15} (large peak)
10/30/90-B	0.17	1.12	10^{13} (small peak)
10/30/90-A	0.14	1.08	10^{13} (small peak)
10/17/90-A	0.07	1.03	$< 10^{13}$ (no peak)
12/17/90-B	0.29b,c	1.11b,c	$< 10^{13}$ (no peak)

a No ^{3}He was detected. Mass spectrometer always at highest sensitivity.
b I = 250 mA/cm^2. All other experiments used I = 200 mA/cm^2.
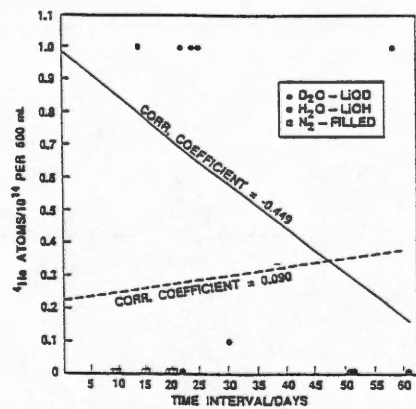c Possible calorimetric errors due to low D_2O solution levels.

Fig. 1.

Fig. 3. Experimental effect of atmospheric helium diffusing into the Pyrex glass flasks. Solid line is for D₂O-LiOD experiments while dashed line is for all experiments.

Fig. 2.

Faradaic Efficiencies Less Than 100% during

Electrolysis of Water Can Account for Reports of Excess

Heat in "Cold Fusion" Cells

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Abstract

The purpose of this study is to evaluate reports of excess heat generation during water electrolysis. Several cells were constructed and operated similarly to those described in the literature. All produced excess heat as defined and calculated in the literature reports, but the production of excess heat could be readily terminated by the introduction of various barriers to the migration of hydrogen and oxygen. Failure to account for reduced Faradaic efficiency (e.g. current that oxidizes H_2 or reduces O_2 instead of H_2O) and systematic calorimetric errors probably account for all the currently available reports of excess heat in both light water Ni/Pt and heavy water Pd/Pt cells. There is no compelling evidence that excess heat is of a nuclear origin in such electrolytic cells.

Introduction

Since Fleischmann and Pons [1] announcement of the generation of excess heat by electrolysis with a Pd cathode in D_2O, many experiments have been done in an attempt to duplicate or even improve upon their results. More recently excess heat has also been observed during electrolysis of light water with K_2CO_3 electrolyte, a Ni cathode and a Pt anode. The purpose of this study is to evaluate these literature reports to determine the source of the reported excess heat.

The excess heat rate in an open cell is given by equation 1,

$$\%\text{Excess heat rate} = 100(q_{meas} - q_{cell})/q_{cell} \qquad (1)$$

where q_{meas} is the heat rate measured in a calorimeter. The value of q_{cell}, assuming 100% Faradaic efficiency, is calculated by equation 2,

$$q_{cell} = I(E_{cell} - E_h) \qquad (2)$$

where I is the cell current, E_{cell} is the total voltage across the cell, and E_h is the thermoneutral potential. E_h is given by equation 3,

$$E_h = \Delta H_f/F \qquad (3)$$

3

4

where ΔH_f is the enthalpy change for formation of water and F is the Faraday constant. Calculations using ΔH_f values from reference 2, gives $E_h = 1.48$ V for light water and 1.53 V for heavy water. The assumption of 100% Faradaic efficiency, required for equation 2 to be valid, means that the electrical current is consumed by the reaction $2H_2O \rightarrow 2H_2 + O_2$ and by no other reaction.

To date, no compelling evidence has been presented for any clear explanation of the origin of the excess heat observed. The excess heat could come from unsuspected chemical reactions, mechanical or electrical work, experimental error, nuclear fusion, or new chemistry. This paper will present the results of experiments showing that reactions of hydrogen and oxygen at the electrodes probably account for many previous observations of excess heat. Other reports of excess heat are shown to result from systematic calorimetric error. Thus, no new physics or chemistry is warranted to explain calorimetric measurements on such cells.

Information in the papers reviewed for this study is insufficient to determine if all of the reports of excess heat can definitely be attributed to the reactions associated with equation 4,

$$2H_2 + O_2 \rightarrow 2H_2O \qquad\qquad (4)$$

but neither do any of the papers give compelling evidence that it cannot be. For example, some papers do not include the cell voltage and/or current, and therefore the total input power cannot be calculated. An excess heat rate greater than the total input power, i.e. IE_{cell}, cannot be attributed to a Faradaic efficiency <100%. Mills et al. [3] make the only clear claim to such an excess heat rate, but the use of pulsed power and uncertainty about calorimetric accuracy complicates interpretation of their work. Neither can Faradaic efficiency explain excess heat generated in a closed cell with the recombiner inside the calorimeter. Bush et al. [4] make claim to observing excess heat this way, but the description of their calorimeter, although recently published, remains incomplete. Most of the papers reviewed have addressed the issue of Faradaic efficiency, and all assumed 100% for various reasons, but without compelling experimental evidence. To provide compelling evidence against reactions of H_2 and O_2 as the source of excess heat, the Faradaic efficiency must be measured *simultaneously* with excess heat.

In considering the possible reactions of H_2 and O_2, Srinivasan et al. [5] suggest, ". . . if the applied voltage is more than 2.96 V, (as in most of our experiments) in order to generate apparent excess power of say 50% the recombination fraction has to be more than

50%. Thus excess heat margins of 50% or more measured in some of our experiments, particularly in the low input power range, cannot be explained away on the basis of recombination effects." This argument is based on the assumption that the Faradaic efficiency cannot be less than 50% under any condition and ignores problems inherent in equation 1 when E_{cell} is near E_h. Noninski [6] argues that nickel is a poor catalyst for recombination, but in fact, nickel, platinum, and palladium are among the best catalysts for the combination of hydrogen and oxygen [7]. Srinivasan et al. [5], Mills et al. [3], Noninski [6], and Miles et al. [8] all measured the Faradaic efficiency of cells similar to those used in their heat measurements, but none measured heat and gas production rates simultaneously. Their evidence against reaction 4 occurring is thus not compelling. After arguing that recombination could not be the source of excess heat in his experiments, Noninski [6] says, "The problem of recombination is a crucial one ..., and it deserves special attention in any further experiments." In an attempt to increase the production of excess heat, Mallove [9] assembled a list of "Protocols for Conducting Light Water Excess Energy Experiments." Many of these protocols enhance the rate of reactions of H_2 and O_2 at the electrodes.

Experimental error also cannot be ruled out as the source of apparent excess heat because most papers do not give enough experimental detail to fully assess the methods used. Most continue to use "freshman-level"[10] home built calorimeters with all their inherent uncertainties rather than switch to proven, commercially available designs. It is often impossible for the reader to detect an error from simply reading a paper even when the author tries very hard to give explicit details. Experimenters have frequently failed to follow normal protocols for accurate calorimetric measurements [11]. For example, Notoya's demonstration at the Third International Conference on Cold Fusion in Nagoya, Japan in 1992 showed a Ni light water cell running approximately 10°C warmer than a resistor-heated control cell with supposedly the same input of thermal power [i.e. power equaled IE_{heater} in the control cell and $I(E_{cell}-E_h)$ in the electrolysis cell with $E_{heater}=E_{cell}-E_h$]. By measurement of the voltage drop in a portion of exposed heater lead wire, it was discovered that a resistance of 2.0 ohms in the heater leads accounted for 36% of the total resistance in the control cell heater circuit. In contrast, power loss was negligible in the leads to the electrolysis cell. Thus, 36% of the total power going to the control cell was being dissipated from the leads into the air, and not into the control cell, making it appear cooler than the electrolysis cell. The temperature

difference in this demonstration did not clearly indicate excess heat, but rather, experimental error. A reader of the resulting publication [12] could not detect this problem however, because the heater leads were not described. None of the publications by Mills et al. [3], Bush et al. [4] or Miles et al. [3] contain details on the design and construction of their calibration heaters. Electrical calibration of calorimeters is notoriously difficult, particularly at high power [13].

We will not attempt to prove that all observations of excess heat are due to either a reduced Faradaic efficiency or experimental error. Evaluating all reports of excess heat in sufficient detail would be an impossible task. Furthermore, the burden of providing adequate evidence that the reactions of H_2 and O_2 are not the source of reported excess heat rests on the experimenters. The results reported in this paper support the hypothesis that reaction 4 can explain most available reports to date of excess heat in both Pd/LiOD(D_2O)/Pt and Ni/K_2CO_3(aq)/Pt cells when running at low current densities. Compelling evidence *against* this hypothesis must be presented before considering new theories that propose other origins for excess heat. We also note that none of the claims that excess heat is of nuclear origin have been confirmed by unequivocal detection of equivalent amounts of nuclear by-products

formed during excess heat production [14].

Experimental

The first two experiments used the cell and circuit shown in figure 1. This cell uses the same materials and current density typical of light water cells where excess heat has been reported. In experiment 1, the cell had no glass tubes (K) surrounding the electrodes, and the mixed gas was vented in a single tube rather than through tubes A and F as shown. Experiment 2 was designed to hinder reactions of hydrogen and oxygen as much as possible. To accomplish this, a glass tube closed at the bottom and open at the top was placed around the Pt anode. The top was open to the electrolyte to allow electrical conductivity but minimal transport of dissolved gases. A second tube open at the bottom enclosed the nickel cathode to conduct evolved oxygen gas to vent-tube F. These tubes, constructed from ordinary borosilicate glass, serve as a barrier to inhibit migration of evolved gases between the electrodes. The tubes around the electrodes were vented through separate tubes, A and F, so the gases could not mix before leaving the calorimeter. Since the increased electrolytic path resulted in increased cell resistance, the voltage was adjusted to keep the current density, and therefore the expected excess heat rate, constant during the

experiment.

Figure 2 shows the cell configuration for experiments 3 and 4. The cell circuitry was the same as that used for experiments 1 and 2. The glass frit at the bottom of the cell was used to bubble dry oxygen or nitrogen through the cell at a constant flow rate. This rate was controlled by a two-stage pressure regulator and a fixed length of glass capillary tubing as a flow restrictor; in experiment 3, 60 cm of 30 μm i.d., in experiment 4, 12 cm of 15 μm i.d.

The cathodes for experiment 1-3 were flat, 1 cm^2 pieces of sintered Ni, and for experiment 4, a 4 cm^2 piece of sintered Ni which was curled in a semicircle around the Pt anode. The sintered Ni used in these experiments was 1 mm thick. Sintered Ni is made by hot pressing a mat of Ni wire or powder. It is commonly used for filtration of gases and liquids, and has a very high surface area compared with Ni foil.

In experiments 1-3, the nickel was stored in an air-tight container and handled with plastic gloves to avoid contamination of the electrode surfaces. In experiment 4 the electrodes were cleaned with a solution of 0.1M HCl in methanol and then rinsed in distilled water.

Two further experiments (beyond 1-4) were done using the cell in figure 2. In experiment 5, the electrolyte was replaced with 0.6M Na_2CO_3. In experiment 6, the Ni electrode was replaced with a 1 cm^2 piece of Pd foil, and the electrolyte was replaced with 0.3M LiOD in heavy water.

Data were collected on a near-continuous basis over a six month period during the performance of these six experiments.

In experiments 3 and 4 with gas flowing through the electrolysis cell, the baseline was established both by (a) measuring the calorimeter output signal as a function of gas flow rate with no electrolysis occurring, and (b) calculating the expected heat rate at different gas flow rates with equation 5.

$$q_{gasflow} = [(r_{gas})(\text{vapor pressure of } H_2O)/RT]\Delta H_{vap} \qquad (5)$$

In equation 5, $q_{gasflow}$ is the heat rate resulting from gas flowing at the volumetric rate r_{gas}, R is the gas constant, T is the Kelvin temperature, and ΔH_{vap} is the enthalpy change for vaporization of water. The vapor pressure over 0.6M K_2CO_3 was assumed to be the same as the vapor pressure over pure water. Vapor pressure data were taken from reference [15] and ΔH_{vap} from reference [2]. The baselines calculated with equation 5 agreed with measured baselines to within $\pm 100\mu$W, as shown in figure 3. The measured baseline was the same for flowing either nitrogen or oxygen.

Apparatus

The cells were powered by a Tenmar model 72-420 dc power supply operated in constant voltage mode. The cell voltage, calorimeter output, and voltage across the calibrated resistor (fig. 1) were recorded with a strip chart recorder and Keithley model 195 digital multimeter for experiments 1 and 2. For the rest of the experiments, data were recorded at 60 sec intervals using the digital multimeter and a computerized data acquisition system.

A Hart Scientific Model 4225, isothermal, differential, heat conduction calorimeter was used to make the heat-rate measurements. It was operated at 25.0°C for experiments 1 and 2, and at 30.4°C for the remainder of the experiments. Both sides of the calorimeter were calibrated electrically with the heaters in each calorimeter cell. The calorimeter cells consist of nickel-plated aluminum blocks with a cylindrical cavity 32mm in diameter and 75mm deep. The calibration heaters are 1kΩ wire-wound, low-temperature-coefficient resistors cemented into holes drilled in the corners of the blocks. Separate pairs of matched leads are used to measure current and voltage and deliver power to the heater. Further details of heater construction and calibration test data are available from the manufacturer, Hart Scientific, PO Box 435, Pleasant Grove, UT 84062. The calorimeter cell and cover completely surround the electrolysis cell, so the distribution of heat generated by the electrolysis cell closely duplicates heat generated by the calibration heater, and thus systematic errors in heat rate measurement are negligible. The baseline of this calorimeter is reproducible to <0.5μW. The absolute accuracy in the configuration used for these experiments with several wires and tubes connected through the cell lid is probably better than 10 μW when no gas is flowing through the electrolysis cell. When gas is flowing through the cell from an external source, fluctuations in gas flow rate cause a 100 μW uncertainty in the baseline, see figure 3. In the experiments described here, the calorimetric time constant is determined by the time constant for heat flow from the sample to the detectors. This time constant was experimentally established to be between 5 and 10 minutes, and therefore short compared to the time period of data gathering.

Results

Table 1 shows the results for experiments 1a and 1b with the cell operating at 0.96 mA and 1.66 mA, respectively. The excess heat rate ($q_{xs} = q_{meas} - q_{cell}$) increased 153 μW with the increase in current, but the

input power (q_{cell}) increased 377 μW.

Figure 4 and table 1 show the results obtained in experiment 2 with the cell shown in figure 1. First, with the glass tubes in place (Table 1-2a), then with the tubes off (Table 1-2b), and again with the tubes on (Table 1-2c). When the tubes were off, the input power was lower, but the heat rate measured by the calorimeter was higher. The increase in heat rate, along with the decrease in input power, gave up to 114% excess heat while the tubes were off. While the tubes were on, the excess heat was only ±17%, which is consistent with no measurable excess heat at the accuracy of this experiment.

In experiment 3a, nitrogen was bubbled through the cell at 1.7 ml min⁻¹ for several hours while measurements were made. Then the gas was changed to oxygen. The results with nitrogen (experiment 3a) and with oxygen flowing at 1.2 ml min⁻¹ (experiment 3b) are given in Table 1. When nitrogen was flowing through the cell, the calculated heat rate was within the uncertainty of the measured heat rate under these conditions i.e. ±100μW. Thus no excess heat was found. The results obtained with O_2 flowing are shown graphically in Figure 5. Initially (at time = 15 min in Fig. 5) the oxygen flow rate was started at 2.0 ml min⁻¹. At this flow rate the cell voltage dropped below 1.48 V resulting in a negative q_{cell}

as shown in Figure 5 at 15<t<30 min. At 30 minutes the O_2 flow rate was decreased to 1.2 ml min⁻¹, and at 100 minutes increased to 2.2 ml min⁻¹ for a short time. Again the cell voltage dropped below 1.48 V. When the flow rate was again decreased to 1.2 ml min⁻¹ (at t=130 to 1100 min, Fig. 5) and left constant, the cell voltage fluctuated between 1.42 V and 1.84 V. The calorimeter output does not show these fluctuations because the calorimeter time constant is too long. The calculated excess heat rate ranged up to 750% when O_2 was flowing through the cell. When the cell voltage equals 1.48 V, note that equation 1 predicts ±infinite excess heat rate if q_{meas} is not exactly zero.

Experiment 4 was done to determine the effect of electrode surface area by replacing the 1 cm² Ni electrode with a 4 cm² Ni electrode cut from the same piece of sintered Ni. Table 1 shows an excess heat rate of 1526 and 1626 μW (65 and 18%) respectively for experiments 4a and 4b with no gases flowing. This result can be compared with that of experiment 2b at the same current density which showed an excess heat rate of 407 μW (103%), see table 1. Thus, the absolute excess heat rate observed increased by nearly a factor of four. Comparing experiments 4a and 4b shows that the absolute excess heat rate increased slightly, if at all (i.e. by 100±100μW), when the current was doubled and the input

15

16

power increased by 6,795 μW. The excess heat rate dropped below 400 μW when the cell was purged with nitrogen to reduce the concentrations of dissolved H_2 and O_2. (See Table 1-4c and 4d.)

With Na_2CO_3 as the electrolyte in the cell, only a small amount of excess heat was observed. A small drop of liquid detergent was then added to the electrolyte, after which the observed excess heat rate increased to the same range as that observed in cells using K_2CO_3 as the electrolyte. Because the measured heat rate showed large fluctuations, no numerical data are given in table 1.

In experiment 6 the Ni electrode was replaced with Pd foil and the electrolyte with 0.3 M LiOD in D_2O. The observed excess heat was about 500 μW (25%) at 0.8 mA/cm^2, see table 1. The excess heat was readily eliminated by flowing N_2 gas through the cell (see figure 6). This experiment was performed with a variety of sizes of Pd foil and wire. Higher percentages of excess heat were obtained at lower current densities while at higher current densities excess heat was completely eliminated.

Notably these results are expressed in μW quantities due to the limited size of the cells that can be accommodated in the calorimeter. It is important to realize that current density is the same magnitude as

17

those in the reviewed literature (except [1,8,16,17]), as is also the percent excess heat.

Discussion

Excess heat was observed with all the cells tested in this study, but the results clearly show it was due solely to reaction of H_2 (or D_2) and O_2 to produce H_2O (or D_2O) in the cell. During electrolysis, the solution around the cathode becomes saturated with hydrogen while the solution around the anode becomes saturated with oxygen. In cells without separators, dissolved oxygen is free to migrate to the cathode and hydrogen can migrate to the anode. If hydrogen is oxidized at the anode and oxygen is reduced at the cathode or if hydrogen and oxygen simultaneously come in contact with a catalytic surface, reaction 4 takes place and the Faradaic efficiency is <100%. Experiment 2 demonstrates that by restricting the migration of gases between electrode surfaces, the production of excess heat can be effectively quenched.

The heat rate from reaction 4 can be obtained from equation 6

$$q_{rec} = I * \Delta_r H * f_{gas} / F \qquad (6)$$

where f_{gas} is the fraction of evolved gas reacted. The

18

value of f_{gas} depends on several factors, including the cathode and anode materials, geometry of the cell, current density, and mixing in the cell. Assuming that 50% of the evolved gas reacts in a typical light water cell ($I = 1$ mA, $E_{cell} = 1.8$ V), equations 1 and 3 predict a measured heat rate 2.3 times the expected heat rate. This is consistent with results reported by Srinivasan et al. [5], Bush [4], Mills et al. [3], and Noninski [6]. Oriani has reported Faradaic efficiencies of 50 to 78% from measured gas flow rates of Ni/K_2CO_3 in light water/Pt cells [18].

One of the most common configurations of light water cells reporting large excess heat rates has a Pt anode wound in a spiral around a Ni cathode. Compared to the configuration of our cells, the spiral configuration reduces the resistance of the cell, lowers E_{cell} for a given current, and increases the chance of reaction of H_2 and O_2. Both the reduced cell voltage and increased rates of H_2 and O_2 reactions increase the excess heat calculated by equation 1. Experiment 3 showed that Faradaic efficiency could be reduced to less than 5% by bubbling oxygen through the cell. The flowing gas thoroughly mixes the cell contents and brings oxygen to the surface of the nickel cathode. This experiment shows that some cell configurations substantially increase the reaction rate of the evolved gases. The rate of reaction

4 and hence the excess heat rate should scale with electrode surface area at any fixed steady-state concentration of dissolved H_2 and O_2 gases. Experiment 4 demonstrated that it does scale as predicted.

If the rate of reaction 4 increases and cell resistance decreases with temperature as expected from kinetic theory, the numerator of equation 1 will increase and the denominator will decrease with increasing temperature, thus causing the calculated percent excess heat to increase substantially with temperature. This may answer the question posed by Fleischmann and Pons [19] in connection with Pd/D_2O cells: "How can it be that the temperature of the cell contents increases whereas the enthalpy input decreases with time?" We expect precisely this relationship if reported excess heat in these cells is due to reaction 4.

An optimum in current density versus percent excess heat was observed by Mills et al. [3] and Noninski [6]. This optimum accounts for the observation in experiments 1 and 4 that the excess heat rate does not scale linearly with current. Once the electrolyte is saturated with hydrogen and oxygen, the observed excess heat rate from their reactions should remain approximately constant. Again, the location of the maximum will depend on details of cell configuration. Also, typical of all our runs, the higher the current density (beyond a certain point)

the lower the calculated percent excess heat, suggesting that purging of the solution near the electrode surface by evolved H_2 is effective in reducing the degree to which O_2 can penetrate to the electrode surface. This study did not experimentally examine the question of the excess heat source in cells operated at high current density, e.g. Fleischmann et al. [1]. Accurate examination of such cells will require a different experimental design than used here. However, we note the lack of compelling evidence for the assumption of 100% Faradaic efficiency or for the accuracy of calorimetric measurements on cells operated at high current density [e.g. 1,8,16,17,etc.].

Excess heat has been observed in $Ni/Na_2CO_3(aq)/Pt$ cells by some workers [4,5], but others have used such cells as a control [3,6]. In agreement with a recent report [20] showing that different electrolytes produce differing bubble sizes in aqueous solution, our experiments show that the difference between Na_2CO_3 and K_2CO_3 as electrolytes probably is due to differences in interfacial properties of the solutions at the electrodes. The H_2 bubbles were smaller when K_2CO_3 was the electrolyte than when Na_2CO_3 was the electrolyte in the same cell. Smaller bubbles allow better mobility of gases in the electrolyte and contact between the electrolyte and the electrode surface, thus allowing more

frequent reaction of dissolved gases. When detergent was added to the Na_2CO_3 electrolyte, the bubbles became much smaller, did not adhere to the electrode, and resulted in about the same rate of apparent excess heat as was observed with the K_2CO_3 electrolyte.

Of all the results reviewed, only Mills et al. [3] claims q_{xs} greater than the input power. Mills' use of several different, low resistance calibration heaters and the paucity of experimental detail suggests calorimetric error, i.e. loss or generation of heat in the heater lead wires, is a likely explanation of these results.

Ni, Pt and Pd are effective catalysts for reaction 4. One measure of catalytic efficacy for this reaction is the exchange current density (η_o). The value of η_o depends on solution composition, temperature, electrode geometry and surface area as well as electrode material [7]. Ni, Pt, and Pd much have higher η_o values than the other metals in their periods when compared under similar conditions.

Another consequence of the high η_o values of Ni, Pt and Pd is a minimal overvoltage required to drive electrolysis. The higher η_o, the lower the overvoltage. The overvoltage is equal to (E_{cell} - 1.23 V) where 1.23 V is the reversible cell potential for light water. If the overvoltage decreases, E_{cell} will approach, and may go below, the thermoneutral potential of 1.48 V as shown in

21

22

experiment 4. The excess heat rate given by equation 1 approaches ±infinity or becomes undefined as E_{cell} approaches 1.48 V. In such cases, very small absolute errors in calorimetry will result in large values of apparent percent excess heat as calculated by equation 1. Thus, some of the extraordinary claims made by Mills et al. [3] and others may be due to the effects of small measurement errors or noise in a cell operating near 1.48 V. If q_{meas} does not equal zero when E_{cell} = 1.48 V, q_{xs} goes to ±infinity. For examples of this condition, see experiments 2 and 3 in reference 3 and Figure 5 in this study. Because time constants of heat conduction calorimeters are typically quite long, oscillating signals like those seen in figure 5 are not present in the calorimetric signal. This mismatch in time constants can result in an incorrect calculation of apparent excess heat. Also, because heat losses from electrical calibration heaters are an ever present problem in calorimetry, positive systematic errors are much more likely in calorimetric measurements than negative errors, thus giving positive, but incorrect, excess heat values. Failure to account for even a small amount of exothermic heat from reaction 4 in a cell operating near 1.48 V can cause an apparent very large excess heat rate. Thus, reports of very large, exothermic excess heat rates on cells with low overvoltage are probably an artifact

arising from a combination of small experimental errors and the way excess heat rate is defined.

Conclusion

This study was designed to determine the sources of excess heat in electrolysis cells operated at low current densities. Pitfalls that can occur with electrical leads, with blockage of electrode surfaces by bubbles, and more importantly, the pitfall of decreased Faradaic efficiency are demonstrated. Less than 100% Faradaic efficiency was shown to occur in both Ni/light water cells with alkali metal carbonate electrolyte and in a Pd/LiOD cell using heavy water. Excess heat was seen in all of these cells unless they were configured or operated in such a way so as to avoid contact between dissolved hydrogen and oxygen and the electrode surfaces.

If compelling evidence for sources of excess heat other than reaction of H_2 (or D_2) and O_2 is to be obtained, Faradaic efficiency must be accurately determined and calorimetric accuracy must be demonstrated while the cell is producing excess heat. In the absence of confirmatory nuclear products, calorimetric measurements must be beyond reproach. Better calorimetric methods are in common usage and compatible instrumentation is commercially available. Because no such studies have been carefully done, there is no

23

24

compelling reason for not adopting the hypothesis that
calorimetric errors or failure to account for reactions
of hydrogen and oxygen during electrolysis of water
account for all reports of excess heat to date. Compared
with other hypotheses [3,4], this hypothesis is much
simpler and requires no changes in well established
scientific principles. Thus, Occams razor places the
burden of proof on those postulating "new science". Such
proof requires adequate experimentation to establish that
a reduced Faradaic efficiency or calorimetric errors
cannot explain the excess heat effect.

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Table 1. Representative results from experiments 1-6.								
Experiment number	$q_{meas}/\mu W$	I/mA	E_{cell}/V	$q_{cell}/\mu W$ $[I(E_{cell}-E_h)]$	$q_{xs}/\mu W$ $[q_{meas}-q_{cell}]$	% Excess	Glass tubes or gas	Cathode size
1a	743	0.96	1.83	336	407	121	none	1 cm^2
1b	1,274	1.66	1.91	713	560	78	none	1 cm^2
2a	732	1.02	2.09	622	110	18	glass tubes	1 cm^2
2b	803	1.04	1.86	395	407	103	none	1 cm^2
2c	656	1.01	2.18	707	-51	none	glass tubes	1 cm^2
3a	278	1.04	1.84	374	-96	none	N_2	1 cm^2
3b	1,609	1.08	1.61	140	1,468	1049	O_2	1 cm^2
4a	3,861	4.17	2.04	2,335	1,526	65	none	4 cm^2
4b	10,756	8.08	2.61	9,130	1,626	18	none	4 cm^2
4c	6,588	5.73	2.62	6,532	55.8	1	N_2	4 cm^2
4d	7,415	5.68	2.72	7,043	372	5	$\tilde{N}_2$	4 cm^2
6a	2,395	1.65	2.70	1,931	464	24	none	1 cm^2
6b	2,071	1.60	2.81	2,048	23	1	N_2	1 cm^2
6c	2,651	1.60	2.81	2,048	603	29	none	1 cm^2

Figure captions

Figure 1 The cell used for experiments 1 and 2. (A) Tube for evolved oxygen, (B) #24 Pt wire, (C) 10kΩ wire-wound potentiometer adjusted to either 2kΩ (experiments 1 - 3) or 5kΩ (experiment 4) and calibrated for voltage measurements to determine current, (D) cell voltage, (E) leads to power supply -- #32 Cu wire, (F) tube for evolved hydrogen, (G) #22 Ni wire, (H) 0.6M K_2CO_3 level, (I) glass vial, (J) 1 cm x 1 cm sintered Ni. Tubes A and F and wires E extend out of the boundaries of the calorimeter.

Figure 2 The cell used in experiments 3 and 4. (A) #24 Pt wire, (B) plastic cap, (C) glass test tube, (D) fine glass frit, (E) tube for exiting gases, (F) tube for incoming gases -- attached to 30 (or 15) μm id glass tubing and gas system, (G) #22 Ni wire, (H) 0.6M K_2CO_3 level, (I) 1 cm x 1 cm sintered Ni. Wires A and G are attached to #32 Cu wire leads and the circuit shown in figure 1. Wires A and G and tubes E and F extend beyond the boundaries of the calorimeter.

Figure 3 Measured and calculated (see equation 1) baselines with dry N_2 or O_2 gas flowing through the cell.

Figure 4 Results of experiment 2.

Figure 5 Results of experiment 3 with oxygen flowing. The calculated heat rate is equal to $I(E_{cell} - 1.48 V)$.
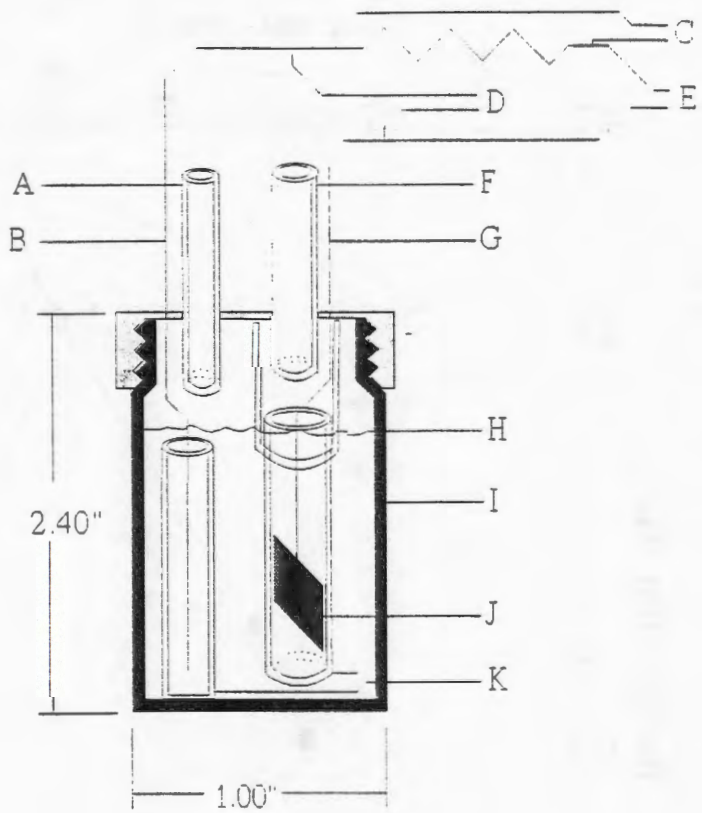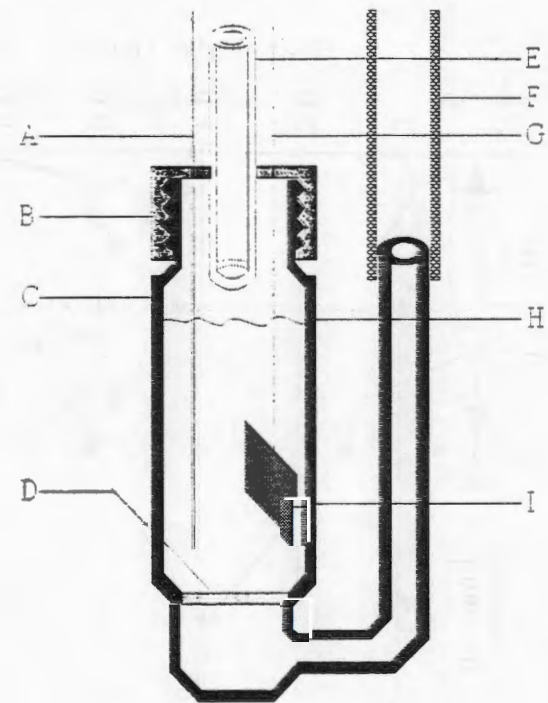
Figure 6 Results of experiment 6 using Pd foil in 0.3M LiOD.
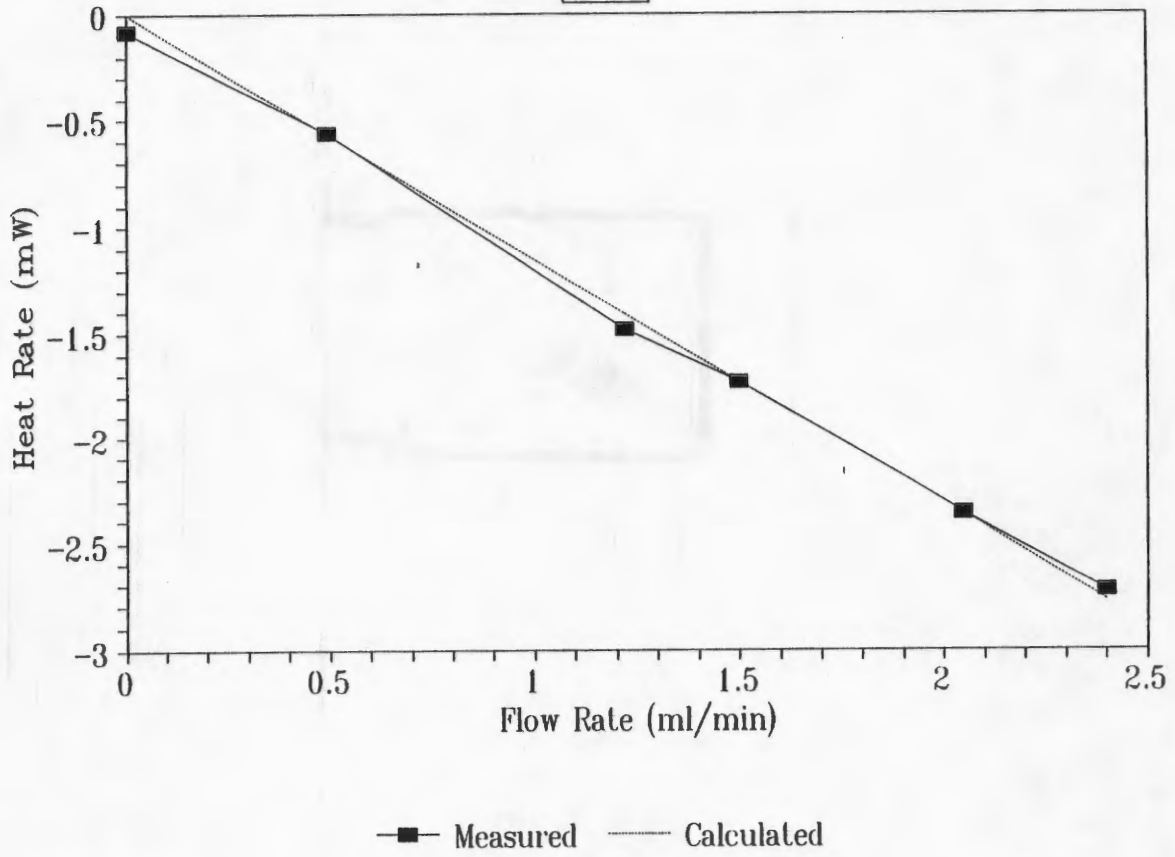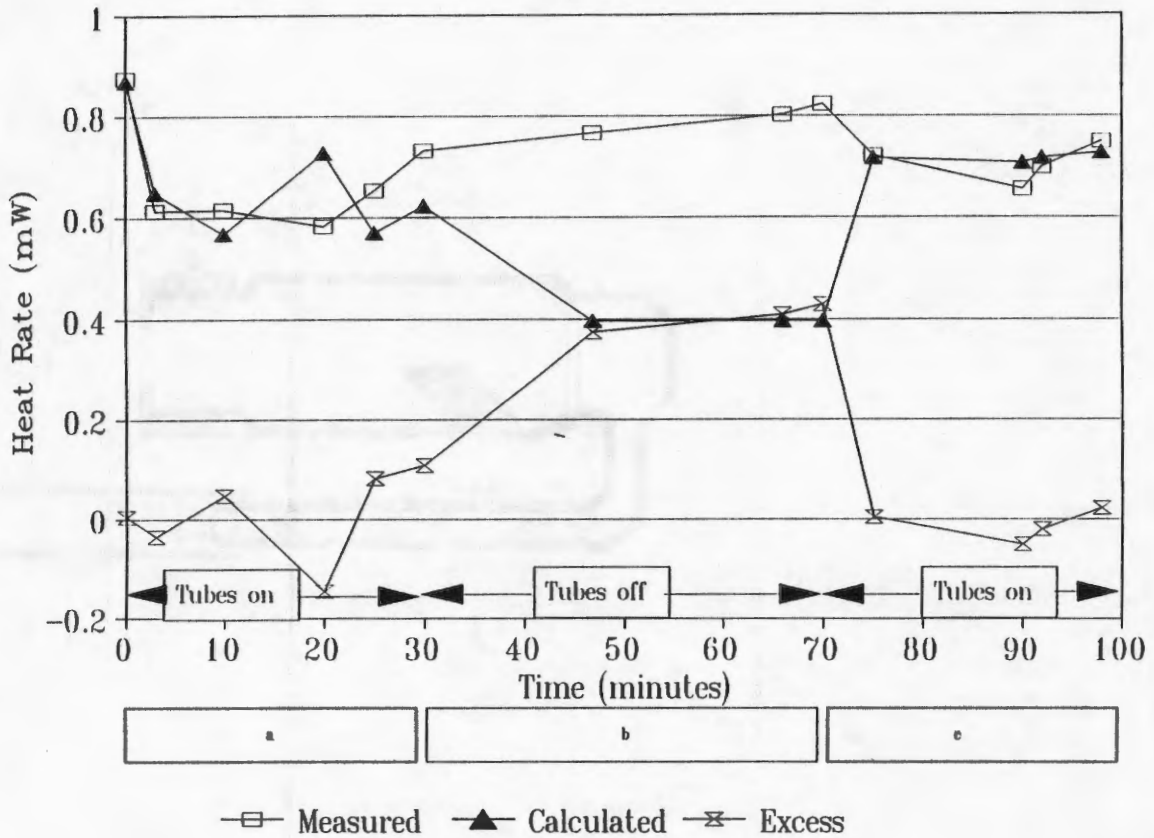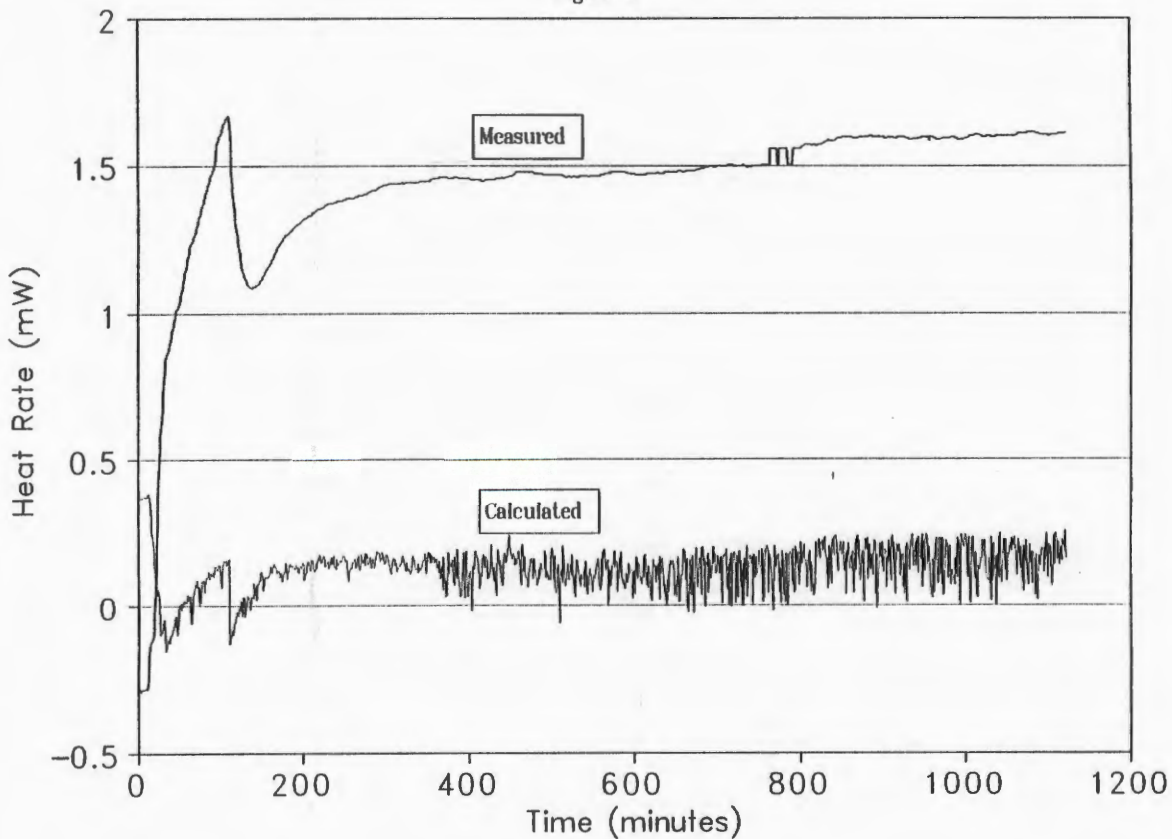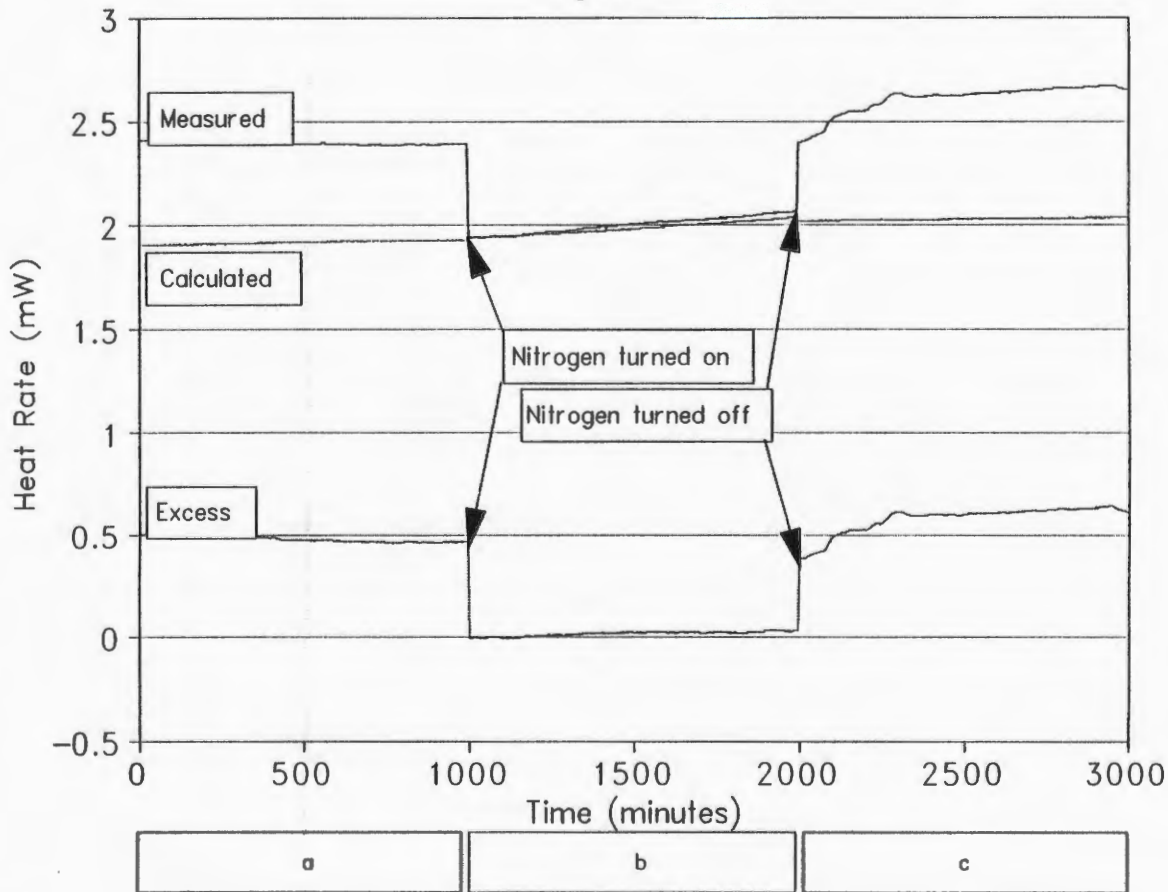
Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6