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Development of Energy Production Systems from Heat Produced in Deuterated Metals

Volume 1

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Technical Report

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Energy Production Processes in Deuterated Metals

Volume 1

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EPRI Project Manager
T.O. Passell

(See last page for authors)

EPRI 3412 Hillview Avenue, Palo Alto, CA 94304, PO Box 10412, Palo Alto, CA 94303, U.S.A. 800.313.3774 or 650.855.2000, www.epri.com

The mass flow calorimeters, because of their size, complexity and the need for environmental isolation, are not provided with on-line nuclear detection capability. Instead, three integrating monitors were provided for experiment M4. These monitors had the capability of detecting integrated:

X-rays
neutrons
helium.

The capability to detect x-rays, integrated over the experiment duration, was provided by placing Kodak dental x-ray films, size 00, outside of the 1 mm PTFE liner around the electrolyte chamber. At the termination of the experiment, after 1840 hours (> 2.5 months) of operation, it was found that water damage had rendered the radiation film badges unreadable. We are therefore not able to draw conclusions regarding the production of X-rays or other penetrating radiation, and its association to excess heat production.

The capability to detect integrated neutron flux, as well as x- and gamma rays, was provided by placing commercial (Radiation Detection Company, Sunnyvale, CA) LiF Thermoluminescent Dosimeters (TLD's) inside quartz tubes in the electrolyte. Four TLD's were placed inside two quartz tubes. Two other TLD's were kept as blanks in a lead storage container during the experiment. These blanks were developed along with the TLD's from the cell. The results from the cells M1, M3 as well as the blank samples were all less than 30 mRem as shown in Appendix 2. The configurations for cells M2 and M4 were such that no TLD's could be placed in the cell. It is obvious from these results that no neutron flux was detected by this method. In fact, the output measured in these cells was always somewhat less than that measured in the blanks because the duration of time spent underwater reduced the exposure to ambient neutrons to below that seen by the blanks in a 2" thick lead cabinet.

In an attempt to measure rates of helium production, the gaseous contents of cell M4 were sampled four times during the experiment, and subjected to analysis for ^4He. These analyses were performed by the U.S. Bureau of Mines at Amarillo, Texas. The sample times and results are presented in Table 3-7.

Table 3-7
Summary of Helium Analysis

Sample	Duration	Date	Time	ppm
1	669.4h	8/16/94	15:07	1.556
2	810.2h	8/22/94	11:55	1.661
3	1172.7h	9/06/94	14:30	0.340

4	1407.7h	9/16/94	09:30	2.077

Excess power was first observed in M4 during the third current ramp at ~ 530 hours (8/10/94 19:44). At this time the current density i = 475 mA cm^{-2} and the loading D/Pd = 0.88. Excess power continued with some variability, reaching a maximum of 375 mW (~ 2% of P_{in}), and terminated abruptly at ~ 668h. At this time the current density i = 987 mA cm-2 and the loading D/Pd = 0.86. The energy integrated from the excess power in this period was 82.45 kJ or 9.27 MJ/mole of Pd.

A sample of gas was taken almost immediately following termination of P_{xs} (Sample 1, at 669 h) and found to contain 1.556 ± 0.007 ppm of ^{4}He. A second sample was taken 5.9 days later (Sample 2, at 810.2 h) and found to contain 1.661 ± 0.009 ppm of ^{4}He.

Sample 1. If ^{4}He is produced in the manner suggested by Miles and Bush via the reaction

$$D + D \rightarrow {}^4He + 22.4\ MeV$$

then from 82.45 kJ we expect

$$\Delta ppm = \frac{\delta \text{ atoms x } 10^6 \text{ x } 22400 \text{ cm}^3\text{/mole (at STP)}}{\text{N x V}}$$

where N = Avagrodo's constant = 6.022×10^{23} atoms/mol

V = Volume of cell plus manifold ≈ 250 cm^3

$$\Delta \text{ atoms} = \frac{82.45 \text{ x } 10^3 \text{ J}}{\left(22.4 \text{ x } 10^6 \text{ eV/atom}\right)\left(1.6 \text{ x } 10^{-19} \text{ J/eV}\right)}$$

$$= 2.30 \times 10^{16}$$

thus

$$\Delta ppm = 3.42 \text{ ppm}$$

Given an (assumed) starting concentration of [^{4}He] = 0.34 ppm (the value in the starting D_2 gas - see subsequent discussion of samples 3 and 4), then the "expected" concentration of ^{4}He is

2000 authors reported 62% with no published mathematical explanation.

$$ppm_{expected} = 3.42 + 0.34 = 3.76 \text{ ppm}$$

In sample 1, only 41% of this amount was found.

Meas/Exp: 1,556/3.76 = 41%

Sample 2. The gas sampled at 669h (Sample 1) had 1.556 ppm ^4He. The volume of this sample, reduced the system pressure by 0.73 Atm., from 0.69 to - 0.04 Atm. gauge. Using gas from the D_2 source, the system pressure was increased by 0.59 Atm, to 0.55 Atm. gauge.

4He samples measured individually, not cummulatively as shown in 2004 paper

Given a system volume of 250 cm^3, and a helium content of 0.34 ppm in the make-up D_2 gas (subsequently verified), we can calculate the expected value of ^4He in Sample 2.

$$ppm_{expected} = \frac{0.96 \text{ Atm. x } 1.556 + 0.59 \text{ Atm. x } 0.34}{1.55 \text{ Atm.}}$$

$$= 1.13 \text{ ppm}$$

2000 authors Reported 69% with no published mathematical explanation.

Sample 2 contained 1.66 ppm; 0.53 ppm more than "expected".

Meas/Exp: 1.66/1.13 = 147%
Exp/Meas: 1.13/1.66 = 68%

Discussion of Samples 1 and 2. Sample 1 was lower in ^4He, than predicted by the Miles Bush mechanism, and sample 2 was high. Two opposed hypotheses are offered:

2000/2004 authors offered only one hypothesis

1. Helium is not sourced with P_{xs} by the mechanism of reaction [1], and the ^4He measured originates by air in-leakage or by poor sampling procedures.

2. Reaction [1] is relevant, the integral power excess is measured accurately, but the release of ^4He to the gas phase is subject to an appreciable delay.

These hypotheses are discussed below with reference to the analyses of Samples 3 and 4.

Sample 3. Sample 3 was measured after extensively flushing the (operating) calorimeter with D_2 gas. This sample reflects any residual ^4He in the cell, the ^4He level in the D_2 purge gas, and any in-leakage of ambient air due to poor sampling technique. The value of 0.34 ± 0.01 ppm is consistent with samples previously taken by B. Bush of other D_2 gas cylinders, suggesting:

a. The gas in the cell was adequately purged

b. The sampling effectively excludes room air.

Sample 4. Sample 4 was measured 9.79 days (8.46 x 10^5 s) after Sample 3. During this time the cathode was ramped from 0.1 to 3.1A at 25 mA/hour, held at 3.1A for ~ 2 days, Explained in 2000/2004 as "heating the cathode"

All omitted in 2000/2004 papers

and subjected to current oscillations 3.1/-0.001A with a 4 minute period twice for a total of ~ 2 days. The cell was also subjected to a "mini-boiloff", with the mass flow stopped for 76 minutes; during this time the cell electrolyte temperature rose to 57°C (from 45°C)

In the period between samples there were 5 instances of rapid loading or de-loading (large $\delta x/\delta t$) and the cathode attained a maximum loading of D/Pd = 0.918.

(Loading flux & high loading = prerequisites to P_{xs})

Excess power was not noted during the period between samples. Under steady state conditions, $P_{xs} = 0 + 20/-50$ mW. Several features of the calorimetric balance should, however, be noted as unusual.

(But calorimetry Disabled)

i. Because of the temperature step and current steps, the calorimeter was at significant remove from its steady state for long periods of time (10-20% of the between sample period).

ii. The thermal baseline was not well established. Prior to the ramp, the calorimeter was 10-20 mW above thermal balance, while at the end of the ramp the calorimeter was 40-50 mW below thermal balance even with the non-steady state correction applied.

iii. During the two periods of current oscillations the calorimeter was apparently endothermic, by as much as 100 mW.

DID NOT ATTEMPT TO CALCULATE 4He % OF REACTION [1]

2000 authors showed values of 84% & 104% with no calculations to support values

Hypothesis 1 The helium sourced between purging at Sample 3 and Sample 4 can be calculated as:

$$\Delta \text{ atoms} = \frac{\Delta \text{ppm}}{10^6}\frac{350\text{cc}}{22400} \quad 6.022 \times 10^{23}$$

$$= 1.17 \times 10^{16}$$

$$\Delta \text{ time} = 9.79 \text{ days} = 8.46 \times 10^5 \text{ s}$$

$$\text{Source} = 1.38 \times 10^{10} \text{ atoms/s}$$

We can imagine that the source of this helium is one of the following:

i. Diffusional in-leakage of ^{4}He contained in room air.

ii. Convective in-leakage of ^{4}He contained in room air, either progressively, or at the time of sampling.

iii. Unobserved production via D + D → ^{4}He (or some other reaction)

iv. Slow release of ^{4}He previously produced or occluded.

i. Diffusion

Diffusional flux, $F = \frac{D\,\Delta C\,A}{l}$ moles s^{-1}

where D = diffusion coefficient

ΔC = concentration gradient

A = available area for in-diffusion

l = effective thickness of diffusing area

We can define a parameter

$$X = \frac{DA}{l} = \frac{F}{\Delta C} \quad cm^3\,s^{-1}$$

Assuming constant and uniform in-diffusion,

$$\overline{\Delta C} = C_{air} - \frac{C_{initial} + C_{final}}{2}$$

$$= C_{air} - \frac{C_{initial} + C_{final}}{2}$$

C_{air} = 5.7 ppm

$C_{initial}$ = 0.34 ppm

C_{final} = 2.077 ppm

$\Rightarrow$ $\overline{\Delta C}$ = 4.49 ppm

= 4.49 ppm

$$F = \frac{(\Delta ppm)\ 250\ cm^3}{\Delta t}$$

$$= \frac{(2.077 - 0.34)\ 250}{8.46 \times 10^5}$$

$$= 5.13 \times 10^{-4}$$

$$X = F/\overline{\Delta C} = 1.14 \times 10^{-4}\ cm^3\ s^{-1}$$

This diffusional rate is large. It represents ~ 0.4 cm^3/day which seems too much. If we ascribe all of this diffusion to the ceramic member holding the electrical feed-throughs ($A \approx 10\ cm^2$, $l \approx 0.2$ cm), then $D_{ceramic} = 2 \times 10^{-6}\ cm^2\ s^{-1}$; this is much too large a number.

ii. Convection

The pressure in the system varied from 0.6 to 1.05 atmospheres *above* ambient in the period between Samples 3 and 4. When corrected for temperature, the pressure was not noted to change at all. There is therefore no reason to suspect convective leakage of gas out of the system, and much less reason to suspect convective in-leakage.

Had in-leakage occurred, we can calculate how much room air (at 5.7 ppm ^4He) would be needed to increase the concentration in 250cc from 0.34 to 2.077 ppm.

$$V_{leak} = \frac{2.077 - 0.34}{5.7} \quad 250$$

$$= 76\ cm^3$$

This value seems implausibly large.

iii. Production

If ^4He were produced by a reaction such as

$$D + D \rightarrow {}^4He + 22.4\ MeV$$

we must ask the question whether or not we should have expected to observe calorimetrically the associated power or energy.

2000/2004 papers contradict this statement because those papers assert that a) they knew as fact that there should not have been Pxs/Exs, b) intentionally took actions to, allegedly, release trapped helium.

From the previous calculation, we need to account for 1.17×10^{16} atoms sourced in 8.46×10^5 s (1.38×10^{10} atoms s^{-1}).

For reaction [1]

$$Æ_{XS} = (1.38 \times 10^{10})\ (22.4 \times 10^6)\ (1.6 \times 10^{-19})$$

$$= 4.95 \times 10^{-2}\ W$$

$$E_{xs} = (1.17 \times 10^{16})\ (22.4 \times 10^{6})\ (1.6 \times 10^{19})$$

$$= 4.18 \times 10^{4}\ J$$

(Based on measured 4He, Exs should have been 40kJ)

Given the state of the calorimeter and the number of transient events occurring it is possible (but not likely) that there is a baseline error of 50 mW; this reflects < 0.4% of the average input power, which is the nominal accuracy of the calorimetry. It is also possible (but not likely) that 40 kJ of excess heat could have been sourced during one or more of the calorimetric transients, and not seen.

(But there was no Exs measured)

We therefore cannot rule out the possibility that ^4He was sourced, with excess heat, in the method reported by Miles and Bush.

iv. Hideout

Excess power was observed in this calorimeter, and ^4He measured, some 20 days prior to sample 3. Immediately prior to sample 3 the cell was flushed with D_2 at ~ 10 cm^3/min. for ~ 18 hours; in this time the gas in the cell and manifold was presumed to be equilibrated with that in the D_2 bottle.for ~ 18 hours; in this time the gas in the cell and manifold was presumed to be equilibrated with that in the D_2 bottle.

Since the gas flow enters and leaves at the top of the cell, this equilibration is less likely to have taken place with the ^4He contained in the cell electrolyte (130 cm^3), the cathode (0.08 cm^3) or the PTFE parts of the cell (~ 100 cm^3). We do not know what the partition coefficient for ^4He is between D_2 gas, LiOD, PTFE and Pd metal. Nor do we know the effective diffusion coefficient of ^4He in PTFE or the rate at which ^4He sourced within Pd might be expected to leave. Given a Henry's law coefficient of 5-8 ppm for ^4He in D_2O and PTFE, however, there is certainly sufficient storage capacity to source the observed helium even with some removal during purging with D_2 gas. We do need to consider to what extent the cell parts, including the electrolyte, were saturated with helium (equilibrated with 5.7 ppm in the air) at the outset of the experiment.

Hypothesis 2. In attempting to evaluate a ^4He mass balance on the basis of hypothesis 2 (nuclear source), two critical pieces of information are missing: the helium content of the cell immediately before the initiation of excess heat production at 530h, and before purging at 1154h. We can make progress by assuming that, as intended (a, b) or claimed (c):

a. the system is helium leak tight

b. the initial helium content is that of the D_2 gas cylinder (= Sample 3)

c. helium is produced by reaction [1]

Sources of ^4He	*Atoms*
Initial inventory from D_2	3.9 x 10^{15}
Excess power 530-658 h	2.30 x 10^{16}
D_2 top-up 690 h	1.4 x 10^{15}
D_2 top-up 815 h	1.0 x 10^{15}
D_2 fill 1173 h	3.7 x 10^{15}
Sum	*3.30 x 10^{16}*

Sinks of ^4He	*Atoms*
Sample 1 at 669 h	7.6 x 10^{15}
Sample 2 at 810 h	2.2 x 10^{15}
Purged volume before Sample 3	Unknown
System volume at Sample 4	1.40 x 10^{16}
Sum	*2.38 x 10^{16}*

Clearly, if volume purged before Sample 3 contained ≥ 9.2 x 10^{15} atoms (≥ 1.14 ppm), then a mass balance can be achieved. The inequality is employed because we cannot be certain that all ^4He had been released into the gas at the time of Sample 4. This estimated concentration is entirely plausible, but not provable.

In this model, ^4He is created before Sample 1 (presumably in the cathode, by reaction [1]). This helium is not, however, immediately available in the gas phase where it is accessible for sampling. Instead, the helium is slowly released over a period of a month or more. (2000 paper introduces logical contradiction - states 4He is not released on its own.)

(Might - yes, but not tested)

Diffusion within the metal itself, might explain this time-constant. Alternatively, hold-up in the electrolyte or PTFE parts could supply the mechanism of delay. "It is possibly of significance" that the large ^4He concentration in Sample 4, followed the extended period of temperature pulsing and a temperature step in this sample period.

Conclusions

("Possibly of significance" in 1998 turned into 2000 paper "cell was subjected to thermal & mechanical... to release trapped helium.)

1. We cannot rule out the possibility that ^4He was sourced during the period between samples 3 and 4, or that the measured helium represents a hold-over from helium previously dissolved in D_2O or PTFE.

2. In the event of delayed release, a satisfactory mass balance can be obtained for ^4He on the assumption that

    a. the system is helium leak tight, and

1. The 1998 authors did not know where the sample 4 came from.
2. They did not test to find out its source.

b. the helium is sourced by reaction [1].

3. Convective in-leakage during cell operation or sampling seems a very unlikely source of the measured ^4He, and diffusional in-leakage, while possible, would be very hard to account for quantitatively.

They did not test.

4. The possibility of ^4He hide-out and slow emergence into the gas phase must be tested by experiment. This applies to both the ^4He thought to be produced by reaction [1] and to an initial inventory of ^4He in the LiOD and PTFE, due to equilibration with the ambient.

2000/2004 authors asserted that 4He hide-out was fact, and was used by 1998 authors to "release trapped helium."

5. Definitive statements will be difficult to make about ^4He production in this or future experiments unless or until it is measured at several times the ambient background level.

2000/2004 authors made definitive statements.

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Abstract

EPRI sponsored an experimental program to investigate the idea that heat, and possibly nuclear products, could be created electrolytically in palladium lattices. Observations using high precision mass flow calorimetry revealed that excess heat could be produced in electrochemical cells with palladium cathodes and a heavy water electrolyte in a more or less reproducible manner, when a number of criteria were satisfied. This excess heat generated is far too large to be a chemical or metallurgical transformation. By inference, a nuclear reaction of some as yet undetermined nature is the hypothesized heat source. This report details the observation of excess powers documented in calorimetry experiments.

Background

Palladium (Pd) cathodes electrochemically charged with deuterium (D) to unusually high D/Pd ratios have exhibited episodes of heat in excess of measured electrical inputs. While investigators have not yet definitively observed nuclear reaction products commensurate with the excess heat, they have detected suggestive evidence of nuclear reactions in the form of helium-4 ((4)He), in the cell vapor space in a few cases.

Objective

To measure, optimize, and control the excess heat produced in highly deuterated Pd cathodes; to measure any signatures of possible nuclear reactions associated with the production of the excess heat.

Approach

The project team designed electrochemical cells within totally closed, precision flow calorimeters equipped with catalytic recombiners of the electrochemically produced D and oxygen gases. These systems were sensitive to excess heat episodes in the range above 50 mW, during inputs ranging from 1-45 W. Approximately 38 separate cells/calorimeters operated for periods of several days to several weeks each, with one cell operating nearly 3 mo. Separately, the team operated 107 open cells to test various procedures for attaining the high cathode D/Pd atomic ratios, believed to be a key condition for obtaining excess heat. They measured loading every few minutes by monitoring the electrical resistance of the cathode relative to its value in the pure metal, which has a known functional dependence with respect to D/Pd ratios. They accomplished loading with a combination of initial low cathode current densities of about 20-50 mA/cm(2), followed by current ramps up to about 1.0 A/cm(2). Current reversals to deload or "strip" the cathodes of D and clean the surface by temporarily making it an anode resulted in high loadings. Lithium deuteroxide was almost always the electrolyte at a concentration of 1.0 mol/l, with occasional additions of 100-200 ppm of aluminum, boron, silicon, or copper.

Results

CITATIONS

This report was prepared by

SRI International
333 Ravenswood Ave.
Menlo Park, CA 94025-3493.

Lockheed Martin Company
3251 Hanover St.
Palo Alto, CA 94304-1191

Principal Investigators

M.C.H. McKubre
S. Crouch-Baker
A. Hauser
N. Jevtic
S. I. Smedley
F. L. Tanzella
M. Williams
S. Wing

Non-SRI Contributors

B. Bush
F. McMohon
M. Srinivasan
A. Wark
D. Warren

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