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The Emergence of a Coherent Explanation for Anomalies Observed in D/Pd and H/Pd Systems; Evidence for ^4He and ^3He Production

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Introduction

Numerous observations have been made of apparent anomalies in carefully performed experimental studies of D/Pd and H/Pd systems. Such anomalies include: prompt emission of electrons and charged particles; unexplained heat in excess of known input sources; the residual presence of light elements (notably ^3H , ^3He and ^4He); the possible occurrence of isotope anomalies in higher mass elements (including the host metal); unusual electrical conductance effects both stable and transient (not discussed in this paper). The features that unify these apparently disparate observations are the common elements of the needed experimental stimuli, and the requirement for extended lattice coherent processes in any obvious explanation.

Presented in this paper are the results of four experiments conducted using palladium structures loaded with hydrogen or deuterium by gas and electrolytic means. The purpose was to evaluate possible evidence for d-d fusion reactions in lattice structures, in the temperature range 25-250°C. Primary evidence sought and evaluated were the appearance of anomalous excess heat accompanied by the production of light isotopes: ^3He and ^4He .

Description

Experiments were performed in four basic geometries:

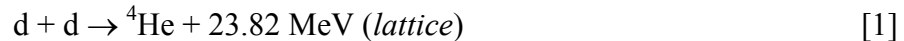
- 1) Open cell electrolysis of D_2O at Pd and Pd-alloy wire cathodes using an accurate integral boundary Seebeck calorimetry.¹ These experiments were designed to replicate earlier observations of Miles et. al.² of the correlated appearance of excess heat and helium in glass cells. The experiments at SRI were performed in rigorously metal sealed cells incorporating active exclusion of ^4He to increase the measurement sensitivity and accuracy.
- 2) Loading of D_2 and H_2 into Pd on carbon supported catalyst using modest gas pressures (1-3 Atm.) and temperatures (170°–250°C). These experiments were designed to test the claim by Case³ to observe excess temperature and increasing ^4He levels under similar conditions. Experiments at SRI were performed in sealed Nupro[®] 50 cc stainless steel vessels connected to a steel manifold. Periodic measurements of ^4He were made by direct connection to an Extrel[®] mass spectrometer capable of resolving the mass-4 peaks of D_2 and ^4He . Using information recorded from temperature sensors placed inside and outside

active and reference gas cells it was possible to obtain heat-flow calorimetric information at times when the catalyst bed temperature rise was significant.

- 3) Closed cell electrolytic loading of D into Pd wire cathodes in a rigorously metal-sealed apparatus using highly accurate mass flow calorimetry.¹ These experiments were intended to replicate earlier results of excess heat measurement at SRI in thermodynamically closed electrolytic loading cells, for which ambient exclusion of ⁴He was not possible.^{4,5}
- 4) Closed cell electrolytic loading of D (and H) into hollow Pd cathodes sealed to contain small dimension Pd-black powders. These experiments were performed to replicate published results by Arata and Zhang in which excess heat, ⁴He and ³He, were found to be associated with the electrolysis of such “double structured” cathodes⁶ in D₂O, but not in H₂O. In experiments performed at SRI accurate mass flow calorimetry⁴ was used to evaluate and compare the heat production of double structured cathodes electrolyzed in D₂O and H₂O in otherwise identical cells. The contents of the cathode void volumes were subjected to retrospective analysis for light isotopes.

Results

Figure 1 presents the results of concurrent excess power and helium measurements performed during open cell electrolysis using two different Pd and Pd-alloy cathodes. In three instances where excess power was measured at statistically significant levels, ⁴He also was found to be conveyed out of the cell in the electrolysis gases (D₂ + O₂). The solid line in Figure 1 plots the regression fit of these data to a line passing through the origin; the dashed line is that expected for ⁴He generation according to the reaction;



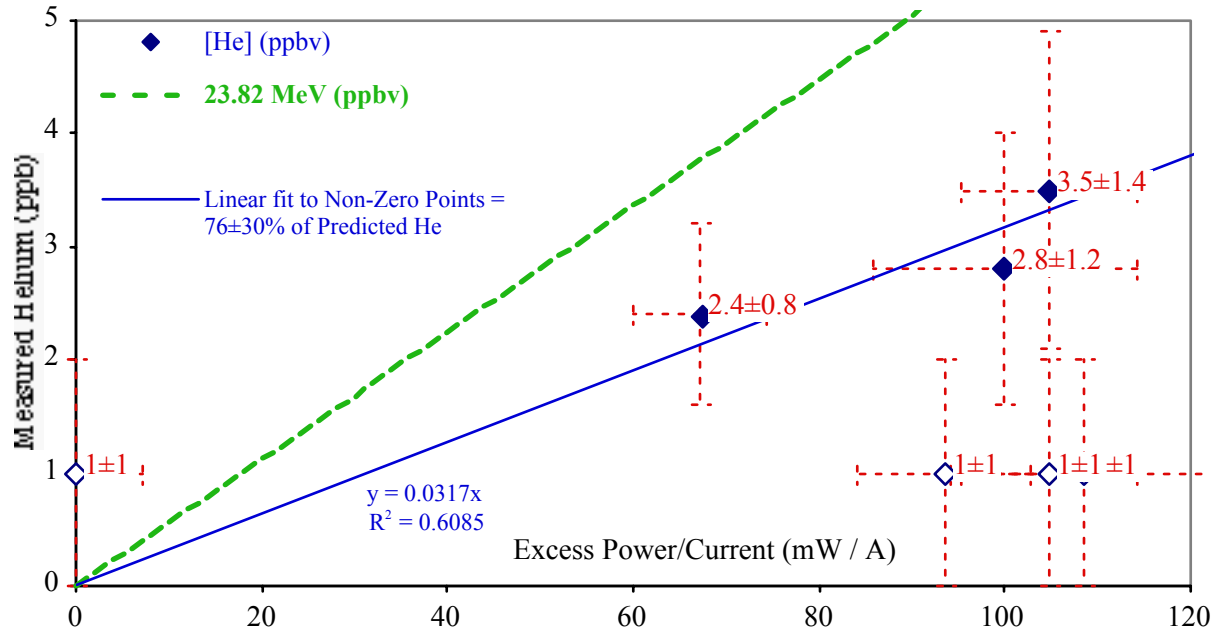


Figure 1 Correlation between rates of Heat and ^4He production in Seebeck Calorimeter.

It is clear from the slopes of these two lines that the observed ^4He constitutes only $76 \pm 30\%$ of the ^4He predicted by equation [1]. A more significant problem in Figure 1 is that three further ^4He samples, taken at times of non-zero excess power (open diamonds), exhibited helium concentrations only at the level of the analytical uncertainty, as did numerous samples taken in the apparent absence of excess power production. Clearly if ^4He is produced in association with excess power, it is not released to the gas phase immediately, or completely.

Experiments in category “2” in which Pd on C catalyst materials were exposed to D_2 and H_2 gases for prolonged periods, exhibited a range of behaviors. Figure 2 summarizes 6 of 16 results obtained in paired cells.

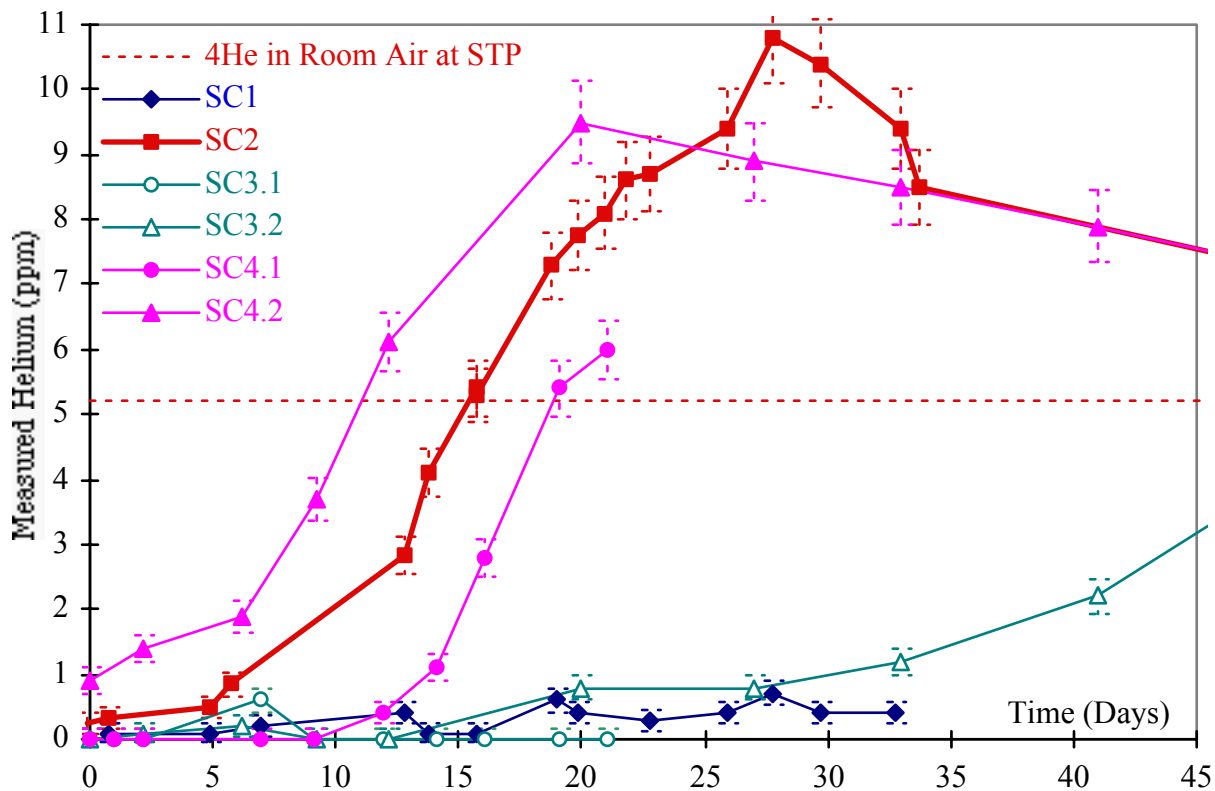


Figure 2 Helium increase in sealed cells containing Pd on C catalyst and D₂ (H₂) gas.

Using direct, on-line, high-resolution mass spectrometric measurement of [⁴He] we observed the following behaviors:

- (i) cells that show no increase of ⁴He over long periods of time (including all cells operated with H₂ instead of D₂);
- (ii) cells that exhibit a slow, approximately exponential increase in [⁴He] with time;
- (iii) cells that display no measurable increase in [⁴He] for a period of several days, followed by a rapid, approximately linear rise in [⁴He] to levels sometimes exceeding that of the ambient background.

Using data from temperature sensors placed in the catalyst and gas phases, and situated in the room, we are able to make heat flow estimates in one of two ways:

- a) a Gradient method, based on the relationship between the temperature difference between catalyst bed and confined gas, and the heater input power;
- b) a Differential method, based on the temperature differences between active and reference catalyst bed sensors, and room temperature, as a function of the relative input heater powers.

The energy estimated in excess of that provided by the heater for these two calorimetric methods is plotted in Figure 3, together with the measured helium concentration during the time of greatest derivative, $\partial[{}^4\text{He}]/\partial t$ in experiment SC2. It is

clear that the appearance of excess heat and the apparent increase in [^4He] are temporally correlated.

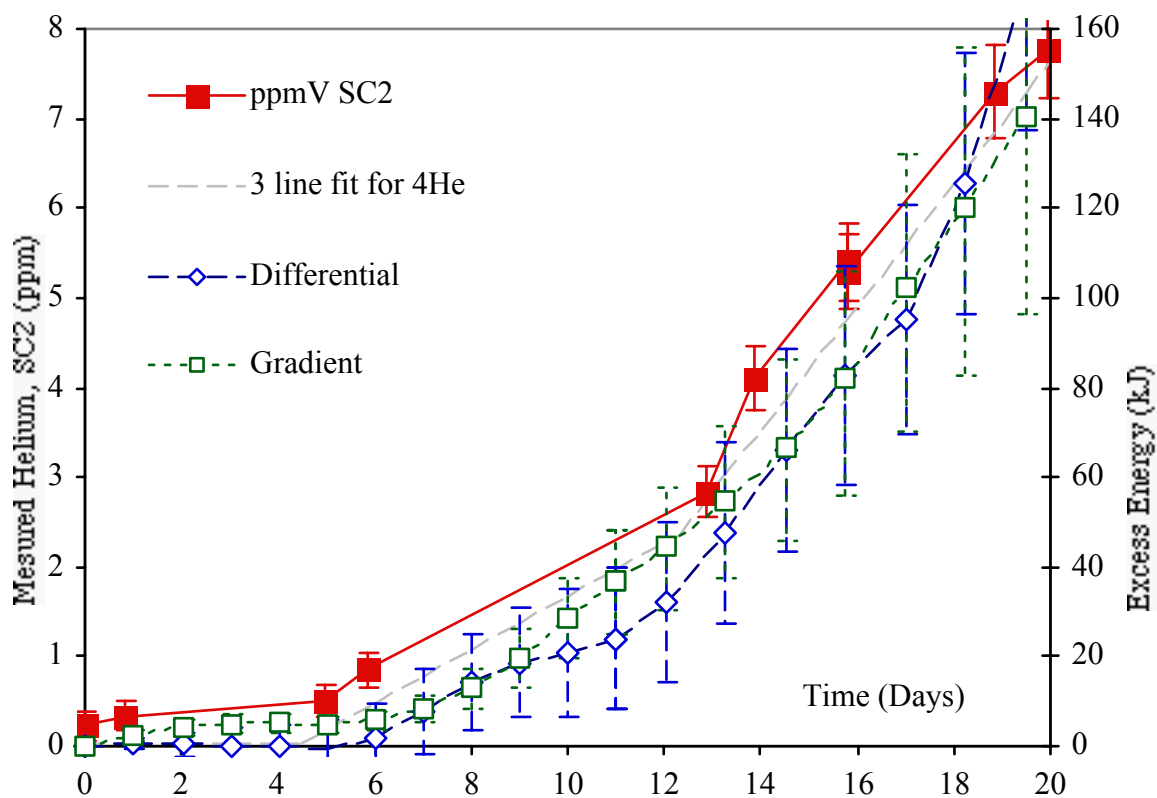


Figure 3 Correlation of Heat and Helium Production in D_2 Gas Cell.

In an attempt to establish a quantitative correlation, Figure 4 plots the two calorimetric estimates of excess heat production interpolated from Figure 3, versus the measured increase in [^4He] (the value plotted in Figure 3 minus the ^4He initially present in the D_2 gas). Regression lines through these data incorporating the origin have slopes: $Q=31 \pm 13$ and 32 ± 13 MeV per ^4He atom, respectively, for the gradient and differential calorimetric methods. Although these Q values include that of reaction [1] within their assigned uncertainties, the mean values for the ^4He presented directly to the gas phase for analysis is only $\sim 75\%$ of that predicted by equation [1].

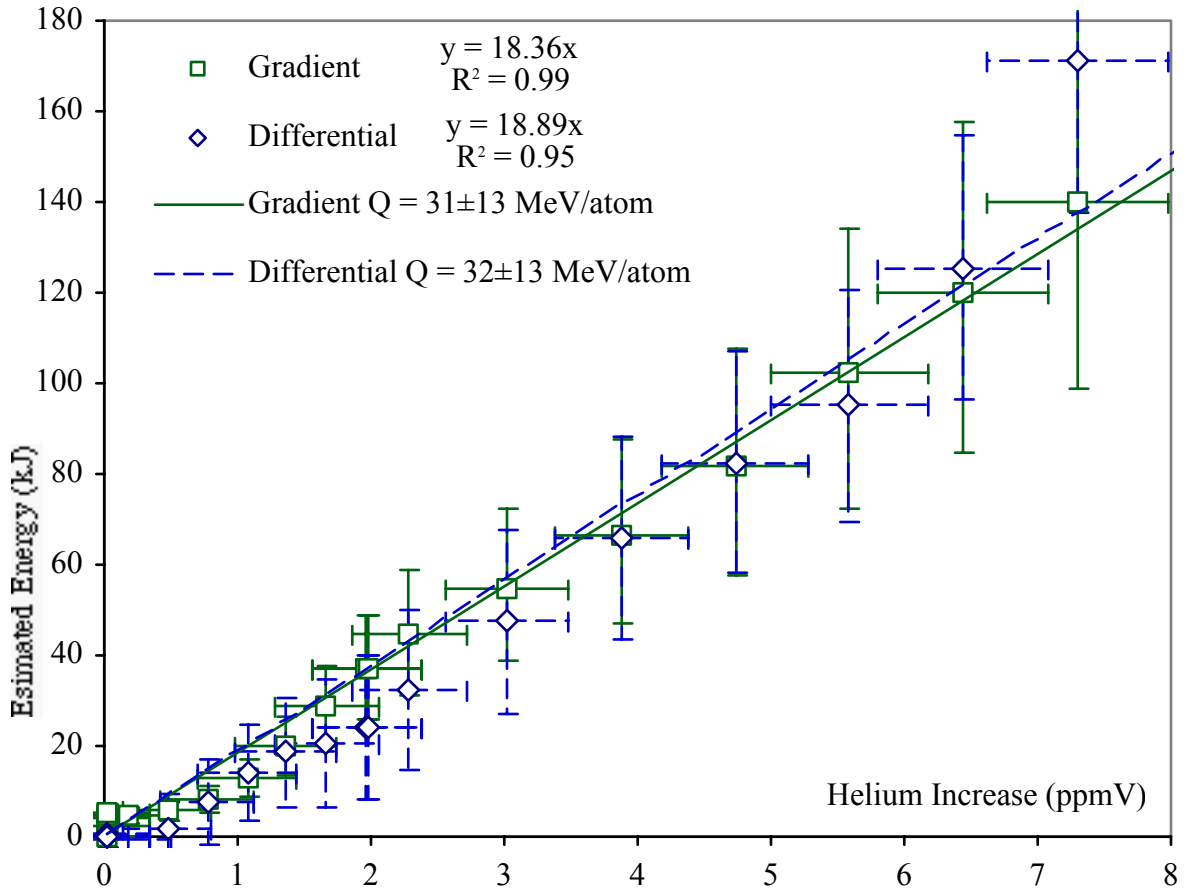


Figure 4 Estimated Energy versus Helium Increase

Clarification of a possible origin for the apparent ^4He deficit in experiments “1” and “2” can be obtained from the results of experiment “3”. Approximately 82 kJ of excess heat was measured in the electrolysis of a 100 mm x 1mm Pd wire cathode in D_2O . This experiment was performed in a rigorously metal sealed and helium leak-tested cell and apparatus provided with the facility to sample the gas in the headspace. When initially analyzed following a period of excess power production, the gas phase contained only 62% of the ^4He expected if reaction [1] were the source of the excess heat. A second sample showed an increase in [^4He] despite the fact that the helium content of the vessel had been diluted with D_2 containing low levels of ^4He , in order to make up the initial gas volume after the first gas sample. Taking these increases as evidence of sequestered ^4He , the cathode was subjected to an extended period (~200 hours) of compositional and temperature cycling by varying the current density in both anodic and cathodic directions.

A mass balance of ^4He was calculated based on two further gas samples: one to determine the helium content of the D_2 gas used initially to fill and refill the sealed metal cell (0.34 ± 0.007 ppmV); the other to measure the final helium concentration in the gas phase after exercising the cathode to release trapped gases (2.08 ± 0.01 ppmV). Taking into account the amounts lost by sampling, and introduced with make-up D_2 , a calculated mass balance for ^4He in the gas phase after compositional and thermal cycling of the

cathode results in a number that is $104 \pm 10\%$ of the number of atoms quantitatively correlated with the observed heat via reaction [1].

The results of experiment “4” generally confirm those published earlier by Arata and Zhang of the appearance of excess heat in the electrolysis of “double structured” palladium cathodes⁶ in D_2O , but not in H_2O . Figure 5 plots the calorimetric results of cathodically exercising two nominally identical cathodes in intentionally similar cells, one in 0.1M LiOD and the other in 0.1 M LiOH. In the same range of input powers, the heavy water cells yields clearly more output heat (plotted as excess power) than does the light water cell.

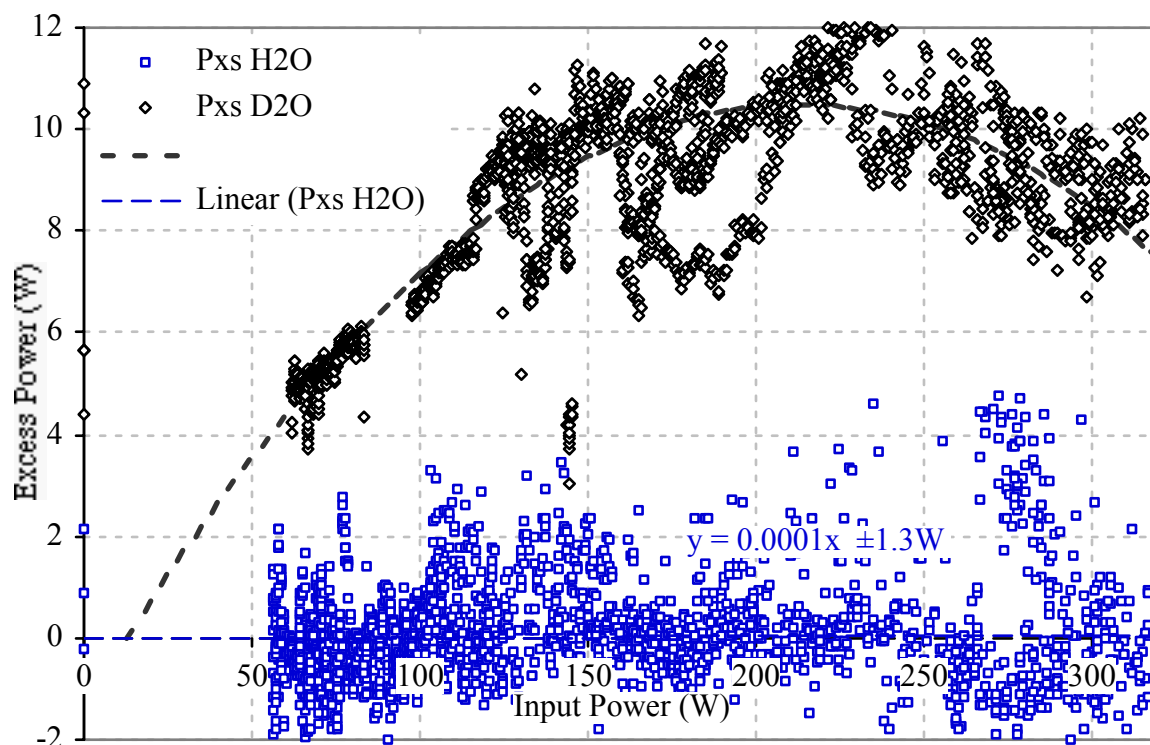


Figure 5 Excess Power versus Input Power measured for Arata-Zhang Double Structured Cathodes using Mass Flow Calorimetry.

The maximum excess power observed in D_2O was $9.9 \pm 1.3\%$ of the measured power input, with the average value being approximately half the maximum. The measured power excess exceeded the experimental uncertainty (1-2%) for a period of ~86 days to produce an integrated energy excess of 64 ± 6 MJ for the D_2O cell. For the H_2O cell in the same period of time the measured energy excess was -1 ± 6 MJ.

At the conclusion of the experiment, both cathodes were removed and placed successively in a sealed chamber where they were punctured mechanically, and the gas contents of the cathode void volumes were extracted for analysis. The Pd black powders also were removed and the Pd metal walls of the hollow cathode were sectioned for analysis. Measurements of 3He and 4He made by volatilizing samples of the metal phases in an ultra high resolution helium isotope mass spectrometer⁷ revealed the presence⁸ of

small amounts of ^4He , but very large amounts of ^3He relative to the natural isotopic abundance ratio (1.38×10^{-6}). This result was observed for the D_2O cathode but not for the H_2O cathode. One set of such results is summarized in Table 1.

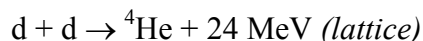
Table 1 Measurements of ^3He and ^4He .

Sample Isotope Error	Palladium - Bulk		Palladium - Black		
	^4He ($\pm 10\%$)	^3He ($\pm 1\%$)	^4He ($\pm 10\%$)	^3He ($\pm 1\%$)	
Measured	7×10^7	1.06×10^6	8×10^7	4.57×10^6	Atoms/mg
Total	7.3×10^{12}	1.1×10^9	9.9×10^{11}	5.5×10^{10}	Atoms
$^4\text{He}/^3\text{He}$	67	1.2×10^4	18	4.0×10^4	R

The numbers in bold represent the factor by which the $^3\text{He}/^4\text{He}$ ratio is enhanced over the natural abundance, and indicate unambiguously a non-natural source for ^3He . The radial distribution of this ^3He (not shown here) clearly demonstrates that a source of this isotope was established within the cathode void volume, and that this source was not present at the outset of the experiment, or in the light water blank.

Conclusions

There exists a strong time correlation between the rates of heat and helium production measured using on-line high-resolution mass spectrometry. In experiments performed using three different metal sealed cells, three different calorimetric methods, by electrochemical and gas loading means and with ^4He analyses made at three different institutional laboratories, there is observed a quantitative or near-quantitative correlation between heat and ^4He production consistent with the reaction:



Evidence for near-surface retention of ^4He in the lattice can be used to accommodate the discrepancy between measured and expected yields of ^4He .

Evidence was obtained for excess heat production in the electrolysis of D_2O using “double structured” cathodes, supporting original claims by Arata and Zhang⁶. Associated with this result, although not quantitatively correlated to it, is the production of a significant amount of ^3He within the cathode void volume, suggesting the presence of a second nuclear reaction. The origins of this ^3He and its theoretical implications are being explored further.

Acknowledgment

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