LOADING, CALORIMETRIC AND NUCLEAR INVESTIGATION OF THE D/Pd SYSTEM

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Hypothesis 1

"There is an unexpected and unexplained source of heat in the D/Pd system, which may be observed when deuterium is loaded electrochemically into the palladium lattice, to a sufficient degree".

Experiments

- Loading \((R/R^o)\)
- Calorimetry

Results

Excess heat and excess temperature observed in mass flow and heat flow calorimeters

Not consistent with known chemistry or with any artifact we have considered

Loading is necessary but not sufficient
- Current (density)
- Temperature
- Initiation (other elements?)
Figure 1  Maximum loading, D/Pd, attained in experiment; determined by R/R°.
Hypothesis 2

"The excess heat originates from an unexpected and unexplained nuclear process"

Experiments

Establish a quantitative and/or temporal correlation between excess heat and nuclear products:

- Gamma and X-ray spectrometer
- Neutron spectrometer
- (Charged particle double telescope)
- Tritium
- Helium
- Isotope shifts

Results

Nuclear process not (presently) confirmed
Separate systems difficult to optimize jointly
Difficult to search for more than one potential product per calorimeter
Attempt to:

- Optimize one system at a time

- Define accurate lower bounds on the production rates in all potential nuclear channels

- Make accurate determination of product energies to aid mechanistic understanding
**Gamma- and X-ray**

Pb passive shield 2' dia., 3' high, 4" wall

NaI Compton suppression annuli, 16" dia., 16" high
- 3" diameter bore
- 5" diameter hole

Ge detectors: mounted coaxially
- 52.5% extra low background, 0.5 mm Be window
  Capability down to 3 keV
  FWHM 1.85 keV @ 1332 keV (Co-60)
- 47.5% low background capsule resolution 2 keV

**Background** 10 keV - 2.5 MeV

Low BG/No Compton suppression ~ 6-7 c/s

Extra low BG/Compton suppression ~ 1 c/s

**Detection Capability /100,000 s**

~ $10^{-3}$ c/s @ 30 keV

~ $10^{-4}$ c/s @ 1300 keV
Neutron

6 active 40x70 cm plastic scintillator
   cosmic ray shields
4" polyethylene passive shield
2 - 5x3" NE-213 liquid scintillators
Pulse shape discrimination between n° and γ
Event-by-event data
1 MeV - 5 MeV
Efficiency ~ 2%

Charged Particles

Triple Si surface barrier detector
Event-by-event mode operation as two δE/E telescopes
with noise rejection

Tritium

Ex-situ electrolyte analysis: 10 pCi/ml sensitivity,
20% Absolute Efficiency
Helium

Extrel # C50
- Ex-situ real time analysis ~ 1 ppm sensitivity
  separate D₂ and He

Bureau of Mines, Helium Field Operations, Texas
- < 1 ppb ⁴He
- < 30 ppt ³He

Isotopes

Mass spectroscopy (SIMS/SALI)
- Depth profiling for solid samples
- Molecular interferences with light elements present
- Sampling problems for highly localized changes

PGAA (Prompt γ Activation Analysis)

CNDP (Cold Neutron Depth Profiling)
Electrochemical Investigation of the Pd-H/D System

EXPERIMENTAL APPROACH

Assess degree of loading (DoL) employing \textit{in-situ} resistance measurements with:

- "Farm" of ten electrolytic cells
- "Movable-Wire" electrode apparatus
  - Batch Differences
  - Anode Selection
  - Surface Modifying Agent
  - Alternating Polarity
  - Annealing Conditions
Reproducibility of Engelhard Pd

Pd Cathode (0.3 x 3 cm); Pt Anode; 1 M LiOD/D2O; Quartz Cell

Electrolysis Time / h

Resistance Ratio, R/Ro
Loading / Stripping Profile

Engelhard (0.3 x 3 cm) Pd Cathode; Pt Anode; 1 M LiOD/D2O; Quartz Cell

Electrolysis Time / h

Resistance Ratio, R/Ro

Cell 1 (A) and V (V)
Effect of Surface Modifying Agent on Loading

![Graph showing the effect of electrolysis time on resistance ratio for different loads.](image)
Effect of Alternating Polarity on Loading

Engelhard Pd Cathode (0.3 x 3 cm); Pt Anode; 1 M LiOD/D2O; Quartz Cell

Electrolysis Time / h

Resistance Ratio, R/R₀

Cell I (A) and V (V)

Lot 3(b)
Lot 2
Effect of Time of Annealing at 850 C on Loading
Engelhard Lot 3 Pd Cathode (0.3 x 3 cm); Pt Anode; 1 M LiOD/D2O; Quartz Cell

Electrolysis Time / h
Resistance Ratio, R/Ro
Cell I (A) and V (V)
CALORIMETRY

Mass Flow: \( I, M \)

Thermodynamically closed

Maintain high accuracy over wide input/output power range

Long term stable operation needed to integrate potential products

Heat Flow

Thermodynamically open

Sensitivity \( (°/W, τ) \)

Flexibility (Geometry, optical detectors)

Redundancy of method

- Isoperibolic: \( T, HF, D, F, G \)
- Seebeck: \( OHF, HH \)
- Lateral: \( LHF \)
LABYRINTH CALORIMETERS

All: 1.0 M LiOD + 200 ppm Al
3mm dia. x 3 cm long Engelhardt Pd
Four Pt wire contacts for $R/R^\circ$

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<th>L2</th>
<th>L3</th>
<th>L4</th>
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"Boil On"
Exp. started 8/3/93 15:43
L3

Exp. started 8/3/93 15:43
90
80
70
TEL - mA/cm² Pxs

Labyrinth de-gassed

Mass Flow Blockage

Exp. started 8/3/93 15:43
3/11/94

5.22
"Heat After Life"
- Step-down from 1642 mA/cm^2

Exp. started 8/3/93 15:43

3/11/94
Exp. started 8/3/93 15:43
3/11/94
"Loss of Resistance"