Cold Fusion 1.01:

Introduction to excess heat in the Fleischmann-Pons experiment

Peter Hagelstein Mitchell Swartz

Jan. 22-29, 2013

Warning

Working in this field at this time can destroy your career.

Being interested may be damaging to your personal and professional life.

Why a new course?

•Need understanding to develop technology
•Need to understand the problem to evaluate claims/technology
•Beginning of need for trained workers in the field

But
•Area controversial
•Many issues not settled
•Lack of acceptance of effects by scientific community

Objectives of this IAP course

•Review excess energy in the Fleischmann-Pons experiment
•Consider why it is important
•Separate problem into known science separate from new science
•Look at experimental issues connected with known science
•Clarify aspects of experiment connected with new science
•Review ideas, models and results from one theoretical approach
•Examine experimental results that shed light on new physics
•Implications for condensed matter and nuclear physics

Introduction

From Wikipedia (2012)

Cold fusion, also called **low-energy nuclear reactions** (LENR), is a type of relatively low temperature nuclear reaction reported to have occurred by some experimenters, but which others have not been able to reproduce. Both the experimental results and the hypothesis are disputed.

From Wikipedia (2013)

Cold fusion is a hypothetical type of nuclear reaction that would occur at, or near, room temperature, compared with temperatures in the millions of degrees that is required for "hot" fusion. It was proposed to explain reports of anomalously high energy generation under certain specific laboratory conditions. It has been rejected by the mainstream scientific community because the original experimental results could not be replicated consistently and reliably, and because there is no accepted theoretical model of cold fusion.

Excess heat (Wikipedia, 2012)

In experiments such as those run by Fleischmann and Pons, a cell operating steadily at one temperature transitions to operating at a higher temperature with no increase in applied current.[22] If higher temperatures were real, and not experimental artifact, the energy balance would show an unaccounted term. **In the Fleischmann and Pons experiments, the rate of inferred excess heat generation was in the range of 10-20% of total input**, though this could not be reliably replicated by most researchers.[120]:3 Unable to produce excess heat, possibly as a result of being unable to achieve high deuterium loading, most researchers declared that heat production was not a real effect and ceased working on the experiments.[84]

Excess heat (Wikipedia, 2013)

…In experiments such as those run by Fleischmann and Pons, a cell operating steadily at one temperature transitions to operating at a higher temperature with no increase in applied current.[25] If higher temperatures were real, and not experimental artifact, the energy balance would show an unaccounted term. In the Fleischmann and Pons experiments, the rate of inferred excess heat generation was in the range of 10–20% of total input, though this could not be reliably replicated by most researchers.[127] Researcher Nathan Lewis discovered that the excess heat in Fleischmann and Pons's original paper was not measured, but estimated from measurements that didn't have any excess heat.[128]

K. L. Shanahan, *J. Env. Mon.* **12** 1756 (2010)

Many positive results have been reported with very high signal to noise ratios as normally computed, and therein resides the problem.

…it would seem that cold fusion calorimetry is currently near or at its limits of accuracy and precision.

Take away message

•If you can't trust Wikipedia, who can you trust?
•Everyone already "knows about" cold fusion
•Everyone "knows" that Fleischmann and Pons experiment cannot be reproduced
•Cold fusion as a field has been discredited
•Working on cold fusion at this time can destroy your career
•Being interested in cold fusion may be problematic

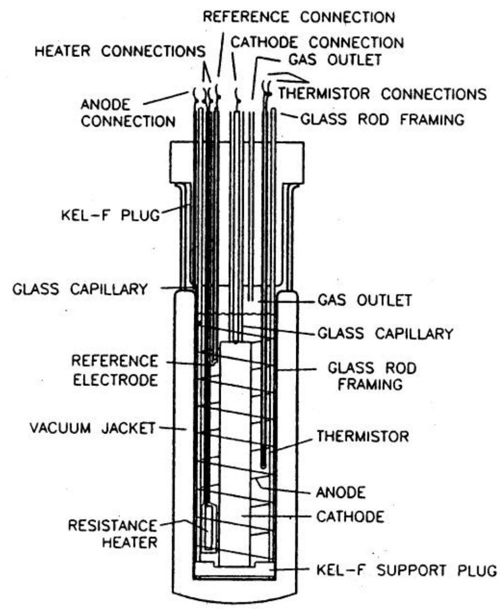
•Which is why it needs to be better understood

Fleischmann-Pons Experiment

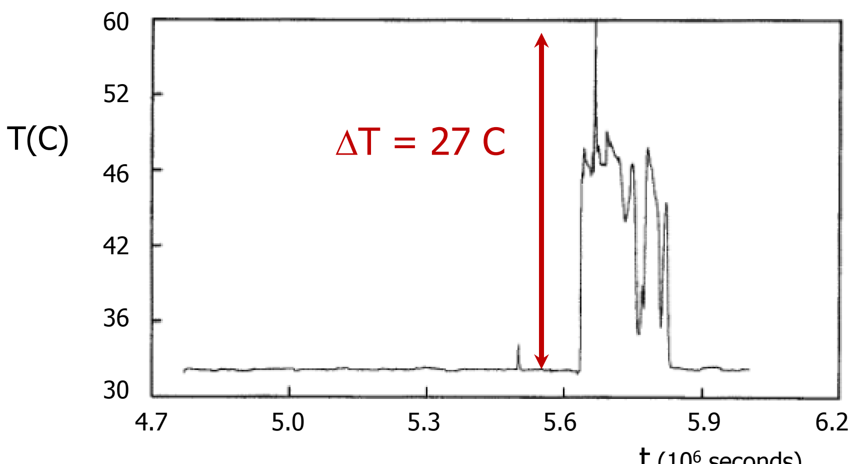
Fleischmann and Pons

Electrochemical cell

The cell

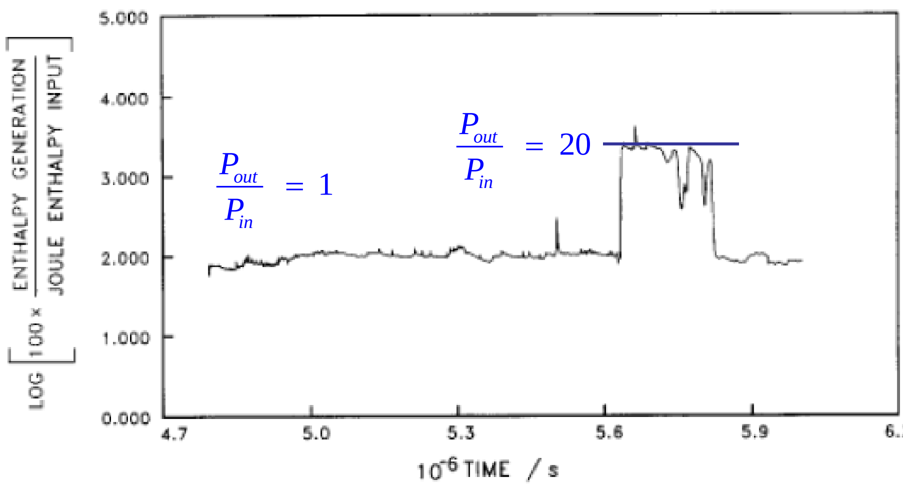


Observation of a heat burst

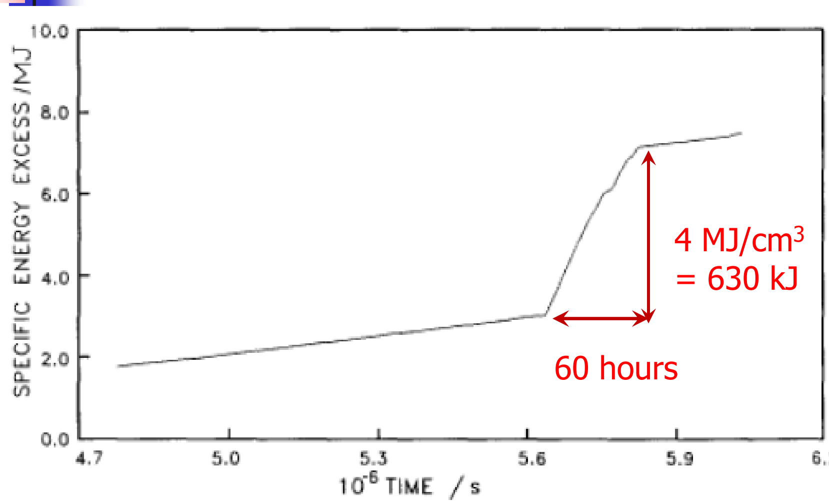


Fleischman, Pons, et al, *J Electroanal Chem* **287**, 293 (1990)

Excess power



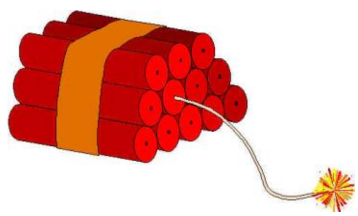
Integrated energy



Thinking about the energy

$$\frac{0.63\ \text{MJ}}{60\ \text{hr}} = 2.9\ \text{Watts}$$

We would only get 1.2 kJ from detonating an equivalent volume (0.157 cc) as the Pd cathode of TNT



Effect not chemical

No commensurate chemical reaction products observed:

In the cell are

Electrolyte: D_2O + 0.1 M LiOD

Cathode: Pd

Anode: Pt

Reference electrode: Pd

Fleischmann conjecture:

Energy produced is of nuclear origin.

Perhaps deuteron-deuteron fusion reactions of some new kind.

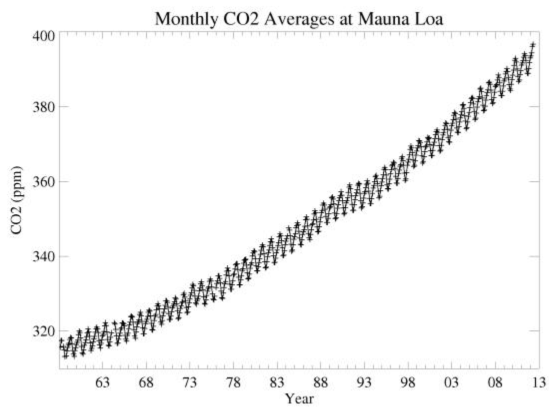
But without commensurate nuclear radiation

Take away message

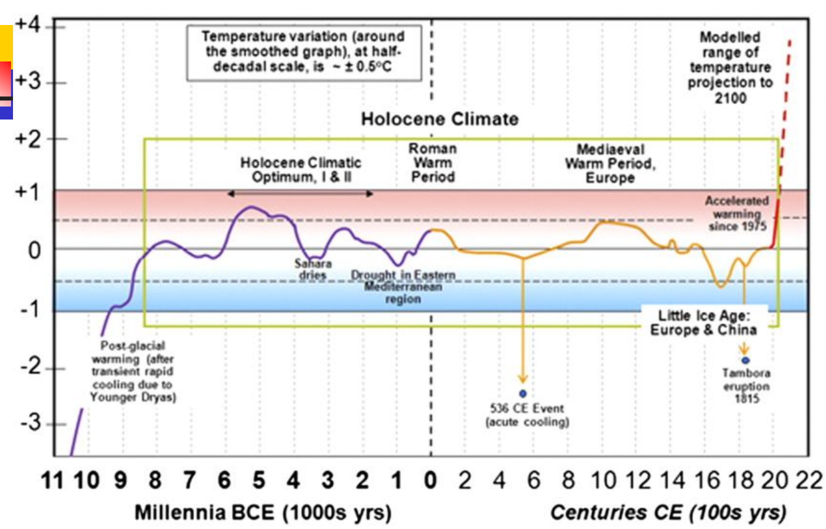
- Large amount of energy observed
- No commensurate chemical reaction products
- No commensurate nuclear radiation
- Fleischmann conjecture that effect is due to a new kind of nuclear process

Implications

disc.sci.gsfc.nasa.gov/featured-items/airs_acos_co2_satellite



Variations in northern hemisphere temperature, °C (relative to mean temperature during 1960–1980), averaged from multiple sources published since 2007.



McMichael A J PNAS 2012;109:4730-4737

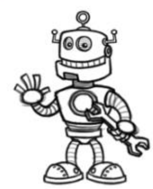
PNAS

If true, what would it mean?

- Clean nuclear energy
- Alternative to oil, coal, natural gas
- No greenhouse gas
- Lots of deuterium available
- Clean water
- Good energy/weight ratio
- Big impact on robotics
- Changes space travel options







DoE ERAB Report

The excitement stems mainly from the claims of heat production by nuclear fusion in these experiments, and the implications of these claims on future energy supply. The attribution of heat production to fusion arises from the presence of deuterium, D, an isotope of hydrogen widely abundant in nature. The known fusion reactions in hydrogen isotopes are shown in Table 1.1. All of these nuclear reactions produce millions of times more energy per reaction than do chemical reactions. **A simple way to harness this energy would be an extremely important discovery.**

…the harnessing of fusion energy for commercial use has been an elusive dream for many decades. The Fleischmann-Pons claim of cold nuclear fusion gave the world the promise of the century, namely, the promise of a virtually limitless supply of a cheap, safe and environmentally clean nuclear energy. **If true, this would be an extraordinary accomplishment.**

John Huizenga, *Cold Fusion: Fiasco of the Century*

Take away message

If excess heat in the Fleischmann-Pons experiment is real then it is important

Seeking confirmation

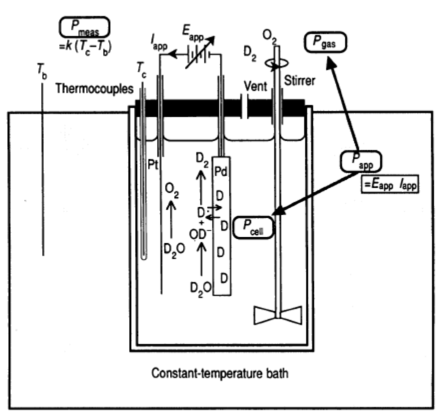
Should we believe it?

We need experimental confirmation before we are sure that it is right!

Within the next few weeks, experiments will surely show whether cold nuclear fusion is taking place; if so, it will teach us much besides humility … **Large heat release from fusion at room temperature would be a multi-dimensional revolution. I bet against its confirmation.**

R. Garwin

CalTech experiment



G. M. Miskelly, M. J. Hebem, A. Kumar, R. M. Penner, M. J. Sailor, N. S. Lewis, *Science* **246** 793 (1989).

No excess power!

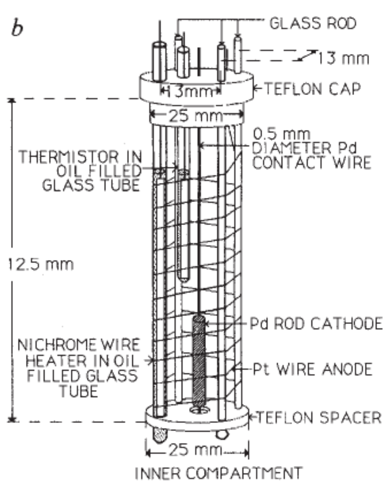
Table 2. Representative calorimetry data for H_2O and D_2O electrolysis cells. The total power was calculated as the resistor power plus the electrolysis power. The measured temperatures are ±0.04°C; bath temperature = 27.000° ± 0.005°C. The error bars quoted were based on 2σ values for random errors in the multimeters and temperature measurement devices. The comparison between H_2O and D_2O gives an estimate of the magnitude of any systematic errors in the calorimeter.

Time (hours)	Current density (mA/cm^2)	Electrolysis power (W)	Resistor power (W)	Total power (W)	Temperature (°C)
Drawn and machined Pd rod (0.21 to 0.22 by 2.1 cm), 0.1M LiOH/H_2O					
34.5	59	0.174 ± 0.002*	0.118 ± 0.001	0.292 ± 0.003	42.62
46.5	86	0.292 ± 0.003*	0	0.292 ± 0.003	42.68
67.5	59	0.175 ± 0.002*	0.119 ± 0.001	0.294 ± 0.003	39.82
77.5	86	0.297 ± 0.003*	0	0.297 ± 0.003	39.92
Drawn Pd rod (0.22 by 2.4 cm), 0.1M LiOD/D_2O					
44.0	66	0.253 ± 0.003†	0.250 ± 0.002	0.502 ± 0.005	49.36
46.0	97	0.457 ± 0.003†	0.044	0.501 ± 0.003	49.39
72.0	97	0.473 ± 0.003†	0	0.473 ± 0.003	45.99
90.0	66	0.277 ± 0.003†	0.204 ± 0.002	0.481 ± 0.005	46.01

*Electrolysis power = (E − 1.48 V) I. †Electrolysis power = (E − 1.54 V) I.

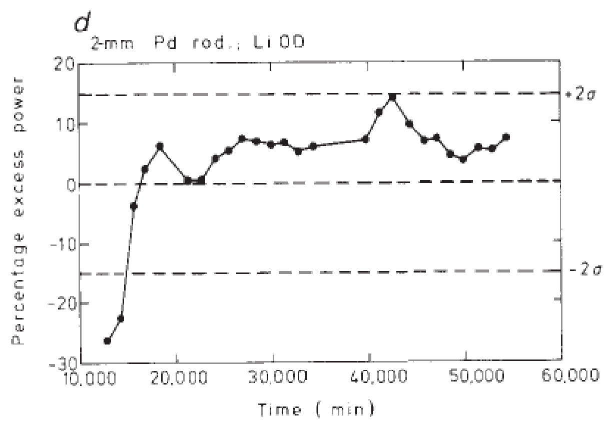
Harwell experiment

b



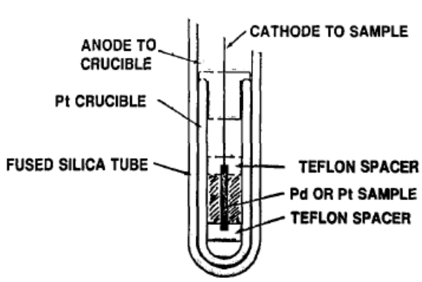
D. E. Williams, D. J. S. Findlay, D. H. Craston, M. R. Sene, M. Bailey, S. Croft, B. J. Hooton, C. P. Jones, A. R. J. Kucernak, J. A. Mason, and R. I. Taylor, *Nature* **342** 375 (1989).

No excess power!

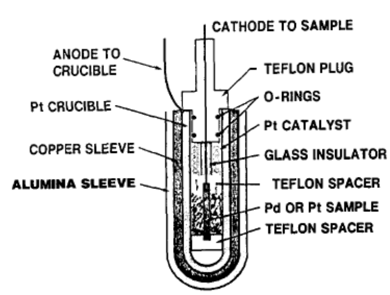


Bell Labs experiment

Open cell



Closed cell



J. W. Fleming, H. H. Law, J. Sapjeta, P. K. Gallagher, and W. F. Mahron, *J. Fusion Energy* **9** 517 (1990).

No excess power!

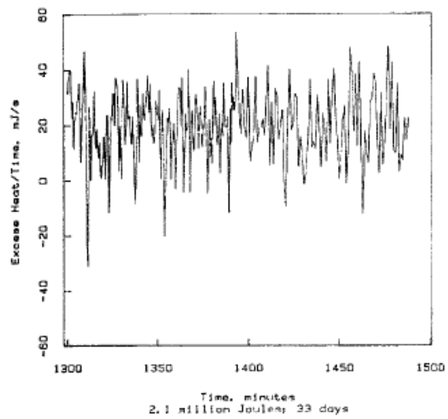
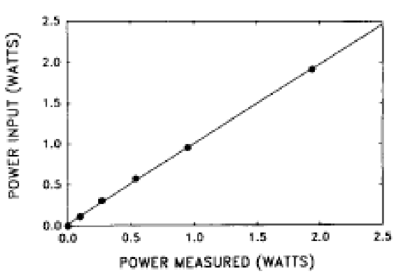
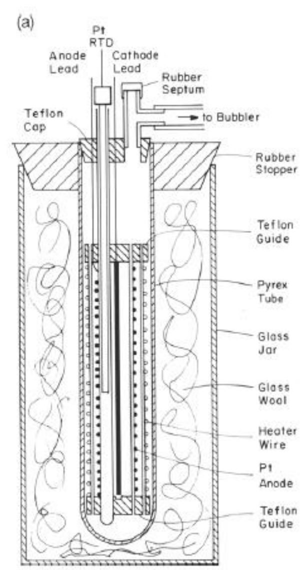


Fig. 11. Excess rate of heat generation vs. time; 1 mm in diameter cast Pd electrode; 1300 mJ/s.

MIT experiment

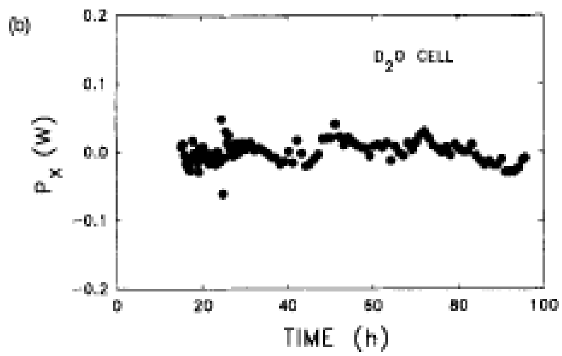
Calibration curve





D. Albagli, R. Ballinger, V. Cammarata, X. Chen, R. M. Crooks, C. Fiore, M. P. J. Gaudreau, I. Hwang, C. K. Li, P. Linsay, S. C. Luckhardt, R. R. Parker, R. D. Petrasso, M. O. Schloh, K. W. Wenzel, and M. S. Wrighton, *J. Fusion Energy* **9** 133 (1990).

No excess power!



On the order of 100 confirmation experiments run in many labs in different countries between 1989-1990 resulting in no excess heat

http://www.dieterbritz.dk/fusweb/biblio.html

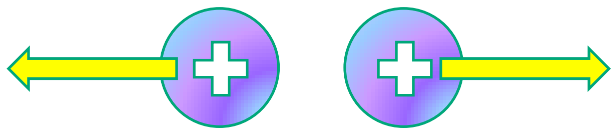
Take away message

- To be sure that excess heat in the Fleischmann-Pons experiment is real, we need confirmations
- A great many experiments were done that gave negative results
- Good scientists from good laboratories were not able to reproduce Fleischmann and Pons results

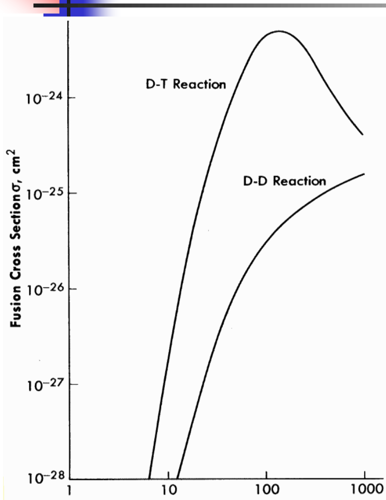
Molecular D_2 Fusion

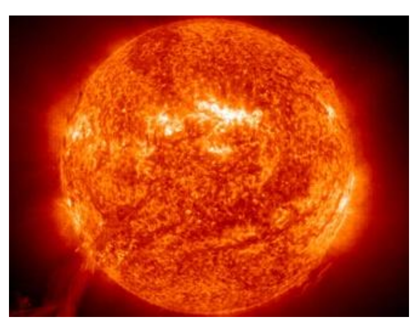
Coulomb repulsion

Deuterons are charged, and Coulomb's law tends to keep them apart

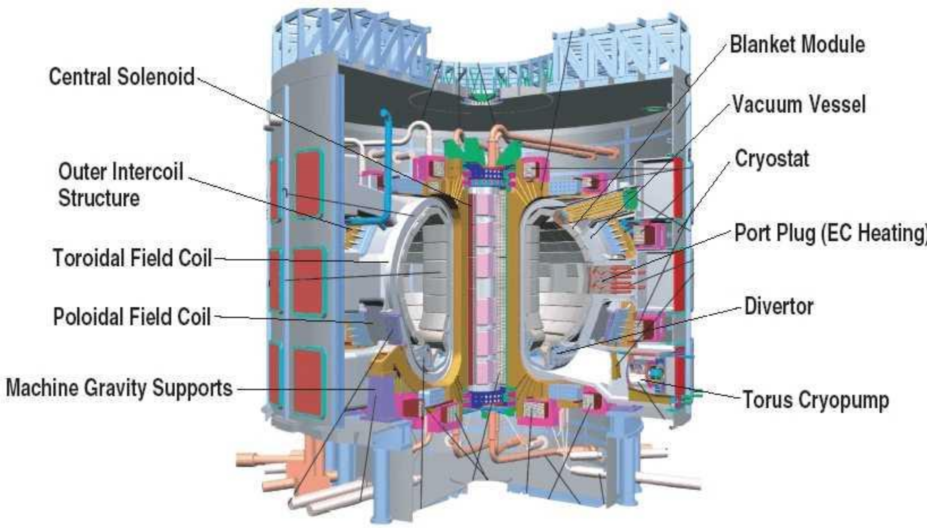


Can use kinetic energy to overcome Coulomb barrier

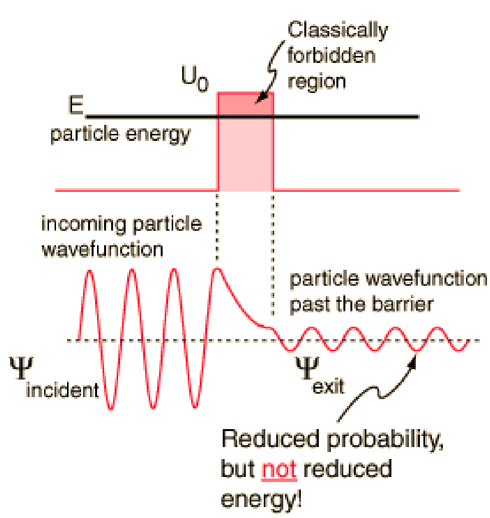




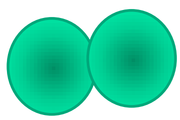
THE ITER-FEAT MACHINE

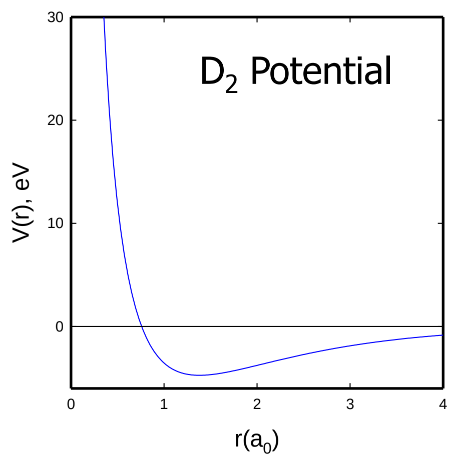


Tunneling in quantum mechanics

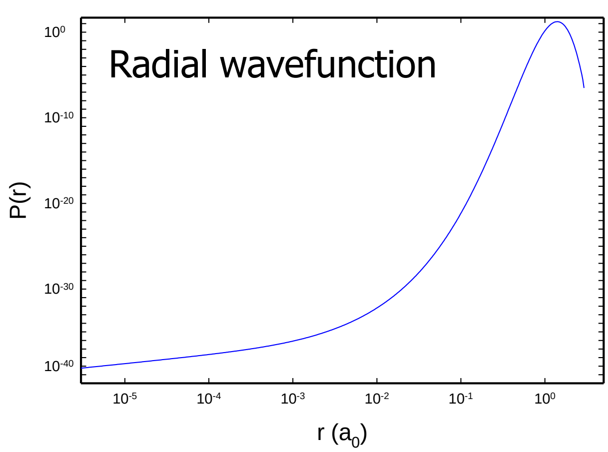


Tunneling in molecular D_2





Quantum calculation



Resulting fusion rates

We would expect deuterons in molecular D_2 to fuse, but it takes a long time. The fusion rate for a D_2 molecule is 3×10^{-64} sec^{-1}.

TABLE 2 Cold-fusion rates in isotopic hydrogen molecules

	$m^*/m_e = 1$	2	5	10
p+p	−64.4	−48.0	−33.2	−25.6
p+d	−55.0	−36.0	−19.0	−10.4
p+t	−57.8	−37.7	−19.7	−10.5
d+d	−63.5	−40.4	−19.8	−9.1
d+t	−68.9	−43.5	−20.9	−9.4

Cold fusion rates are expressed as $\log_{10}$ of the rate in s^{-1}.

Koonin and Nauenberg, *Nature* **339** 690 (1989)

Thinking about D_2 result

$$\gamma = e^{-2G} \frac{v_{nuc}}{v_{mol}} \frac{1}{\tau} = 3 \times 10^{-64}\,\mathrm{sec}^{-1}$$

e^{-2G} : tunneling factor

v_{mol} : relative volume of D_2 molecule

v_{nuc} : relative volume of two deuterons when fusing

τ : time associated with fusion of two localized deuterons

$$\frac{v_{nuc}}{v_{mol}} = 10^{-15} \qquad e^{-2G} = 3 \times 10^{-70} \qquad \tau = 10^{-21}\,\mathrm{sec}$$

Interaction on the nuclear scale is fast

•Once the deuterons get close enough to interact, the fusion reaction happens very fast [$O(10^{-21}$ sec)]

•This is not enough time for light to get to the nearest atom in the lattice ($ct = 3\times10^{10}$ cm/sec x 10^{-21} sec = 3×10^{-11} cm)

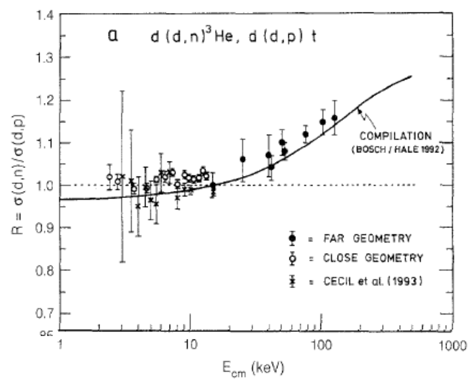
•Arguments have been made that to explain Fleischmann-Pons excess heat, a new pathway must be faster by at least 10 orders of magnitude, which seems impossible kinetically

Primary dd-fusion reactions

Primary deuteron-deuteron fusion channels

$$d + d \rightarrow n(2.45 \text{ MeV}) + {}^{3}\text{He}(0.8 \text{ MeV})$$
$$d + d \rightarrow p(3.0 \text{ MeV}) + t(1.0 \text{ MeV})$$

Proton and neutron channels occur with about 50-50 probability in beam experiments



U. Greife, F. Gorris, M. Junker, C. Rolfs, and D. Zahnow*, Z. Phys. A* **351** 107 (1995)

Take away message

- Coulomb repulsion keeps deuterons apart
- Can get fusion at sufficiently high temperature
- Can also tunnel
- Simplest model is molecular D_2
- Easy to analyze, and the fusion rate is very small
- D_2 fusion would lead to energetic p+t and n+^{3}He products

Excess heat effect not consistent with theory

Conventional physics perspective in 1989

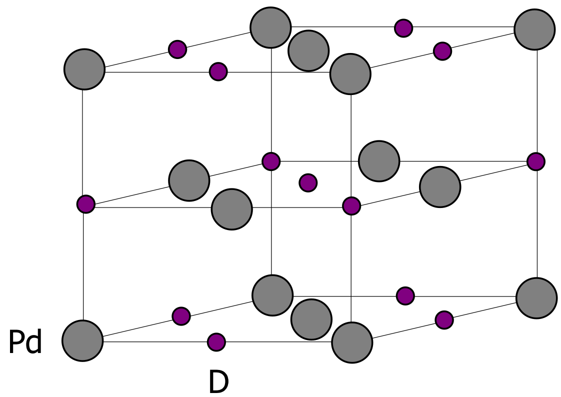
Condensed matter physics

- Born-Oppenheimer separation of electronic and vibrational parts
- Many successes with electron band models
- Many success with phonon dispersion relation models

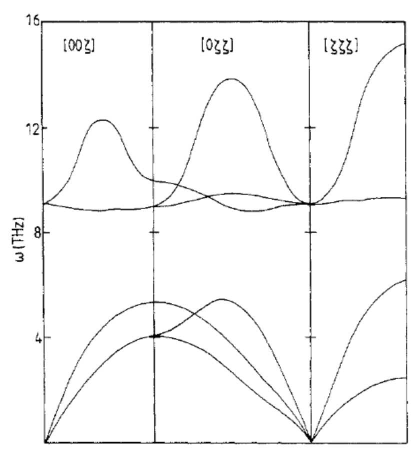
Nuclear physics

- New accurate empirical nucleon-nucleon potentials
- 3,4 nucleon problems solved, now model test problems
- R-matrix methods accurate for few-nucleon reactions
- Early successes with quark models

PdD lattice structure (fcc)

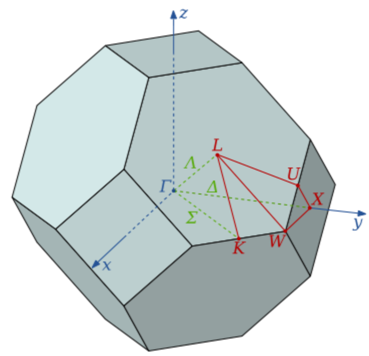


PdD Phonon dispersion curve



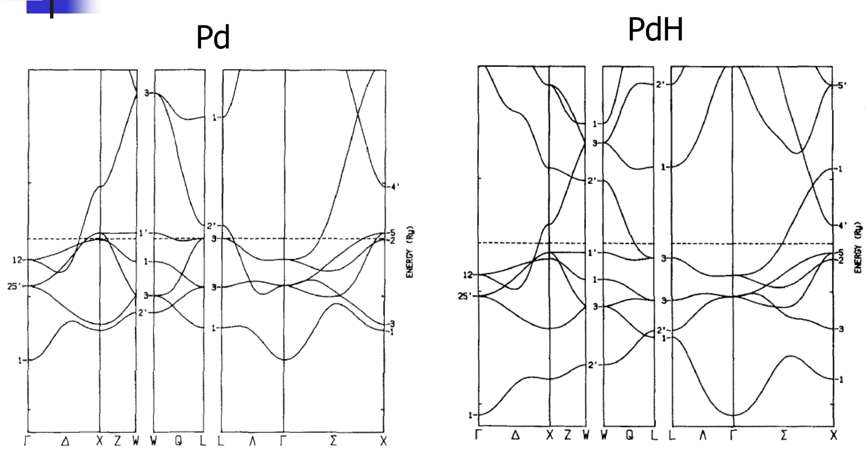
L E Sansores et al
J Phys C **15** 6907 (1982)

fcc Brillouin zone directions



From Wikipedia

Electron bands in Pd, PdH



Klein and Pickett, *J Less Common Metals* **103** 185 (1984)

ARPES measurements in Al

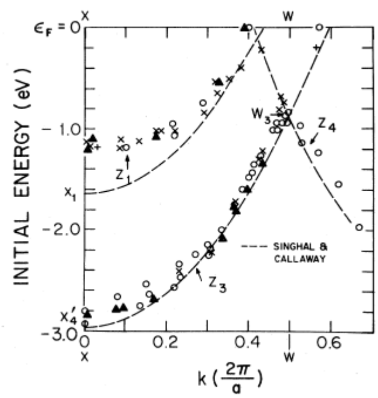


FIG. 10. Comparison of the experimental dispersion from *X* to *W* with theoretical calculations by Singhal and Callaway [Ref. 5(a)].

H J Levinson et al, Phys Rev B **27** 727 (1984)

PdD understood from 1989 condensed matter viewpoint

- •Bonding in Pd due to outer 4d and 5s electrons
- •H or D goes into O-sites
- •Electron density is just right at O-site for H or D occupation
- •Electron density too high for H_2 or D_2 at O-sites
- •D atoms well separated in fcc PdD structure
- •Nothing obvious special about PdD electronic structure or phonon bands

Few-body nuclear physics in the 1980s

$$\hat{H} = -\frac{\hbar^2}{2M}\sum_j \nabla_j^2 + \sum_{j<k}\hat{V}_{jk}$$

Big issue since late 1950s is developing an approximate model for the interaction potential V_{jk} that matches experiments

Argonne 14 model:

$$v_{14,ij} = \sum_{p=1,14}[v_\pi^p(r_{ij}) + v_I^p(r_{ij}) + v_S^p(r_{ij})]O_{ij}^p ,$$

$$O_{ij}^{p=1,14} = 1, \vec{\tau}_i\cdot\vec{\tau}_j, \vec{\sigma}_i\cdot\vec{\sigma}_j, (\vec{\sigma}_i\cdot\vec{\sigma}_j)(\vec{\tau}_i\cdot\vec{\tau}_j), S_{ij}, S_{ij}(\vec{\tau}_i\cdot\vec{\tau}_j), (\vec{L}\cdot\vec{S}), (\vec{L}\cdot\vec{S})(\vec{\tau}_i\cdot\vec{\tau}_j), \vec{L}^2, \vec{L}^2(\vec{\tau}_i\cdot\vec{\tau}_j),$$
$$\vec{L}^2(\vec{\sigma}_i\cdot\vec{\sigma}_j), \vec{L}^2(\vec{\sigma}_i\cdot\vec{\sigma}_j)(\vec{\tau}_i\cdot\vec{\tau}_j), (\vec{L}\cdot\vec{S})^2, (\vec{L}\cdot\vec{S})^2(\vec{\tau}_i\cdot\vec{\tau}_j) .$$

Wiringa et al, *Phys. Rev. C* **29** 1138 (1984)

1980s calculation for the triton

TABLE II. Two-nucleon force results for the RSC and V14 potential models as a function of the number of channels.

		$-E_F$ (MeV)	$\langle r^2\rangle_{ch}^{1/2}(^3He)$ (fm)	$\langle r^2\rangle_{ch}^{1/2}(^3H)$ (fm)
RSC	5	7.02	1.89	1.70
	9	7.21	1.87	1.68
	18	7.23	1.87	1.68
	34	7.35	1.85	1.67
V14	5	7.44	1.86	1.68
	9	7.57	1.84	1.67
	18	7.57	1.84	1.67
	34	7.67	1.83	1.67
Expt.		8.48	1.69(3)	1.51(4)

Old Reid soft-core potential:

Newer Argonne potential:

Chen et al, Phys. Rev. Lett. **55** 374 (1985)

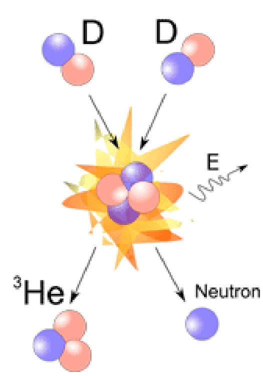
Modern calculations

Table 1. ^{3}H g.s. energies (in MeV), point-proton radii (in fm) and nd scattering lengths (in fm), obtained using the N^{3}LO NN potential [30] with and without the local N^{2}LO NNN interaction [71] with $c_D = 1$ and $c_E = -0.029$, compared to experiment. Calculations performed within the NCSM and/or hyperspherical harmonics (HH) expansion approaches.

		^{3}H		nd	
		$E_{\mathrm{g.s.}}$	$\langle r_p^2 \rangle^{1/2}$	^{2}a	^{4}a
NN	NCSM [71]	−7.852(5)	1.650(5)	–	–
NN	HH [17]	−7.854	1.655	1.100	6.342
NN+NNN	NCSM [71]	−8.473(5)	1.608(5)	–	–
NN+NNN	HH [17]	−8.474	1.611	0.675	6.342
Expt.		−8.482	1.60	–	–
Expt. [74, 75, 76]		–	–	0.65(4)	6.35(2)
Expt. [77, 78]		–	–	0.645(8)	–

P. Navratil et al, *J Phys G Nucl Phys* **36** 083101 (2009).

Deuteron-deuteron fusion to 3+1 final state channels



Good agreement between theory and experiment

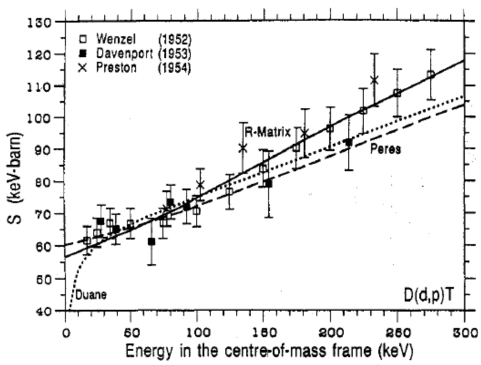


FIG. 12. S-function of the D(d,p)T reaction. The data of Wenzel and Whaling [61] and those of Preston et al. [59] are much higher than the old parametrizations for energies above about 200 keV.

$$\sigma(E_r) = \frac{S(E_r)}{E_r}\exp\left\{-\sqrt{\frac{E_G}{E_r}}\right\}$$

E_r : relative energy

E_G : Gamow tunneling effective energy

Bosch and Hale, *Nuclear Fusion* **32** 611 (1992)

dd-fusion well understood in nuclear physics in 1989

- Nucleon-based models with empirical potentials
- Good description of nuclear structure and reactions
- Few-body nuclear structure problems solved
- Reaction models described d+d fusion very well
- Reaction energy expressed as kinetic energy of p+t, or n+^{3}He

Take away message

•Condensed matter physics and nuclear physics are mature fields

•PdD is a "simple" condensed matter problem

•Deuteron-deuteron fusion is a "simple" nuclear physics problem

•Nothing seems special about PdD that would produce excess heat

•Nothing seems special about deuteron reactions that would help

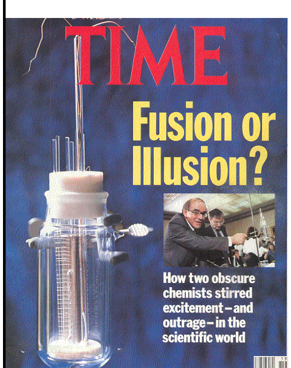
•Conclude that effect is impossible based on these fields

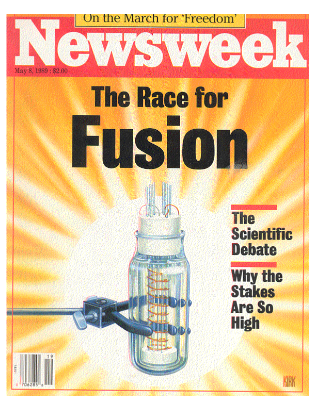
More take away message

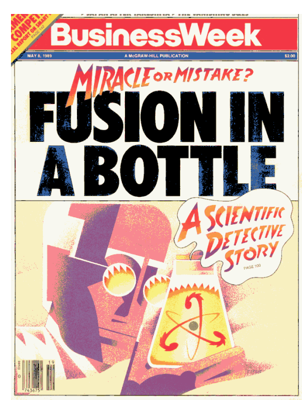
Cold fusion as a real experimental effect would imply that we (collectively) do not understand condensed matter physics and/or nuclear physics in some very fundamental way; that we are missing something very basic in our physical models.

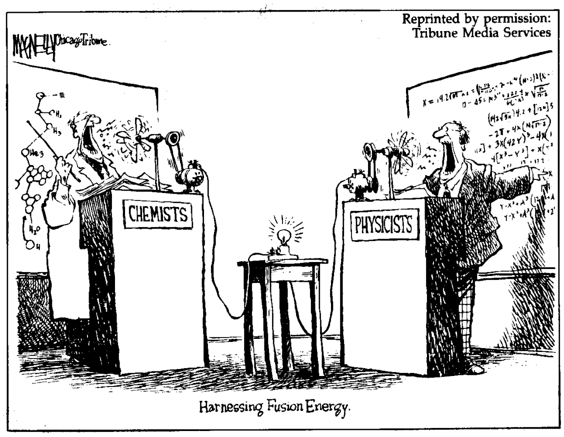
Skepticism

Early May, 1989

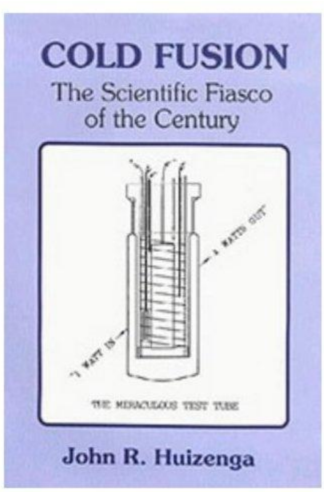








Research and Development Magazine (July 1989)



John Huizenga described three miracles in his book *Cold Fusion, Fiasco of the Century* that would be needed for excess heat in the Fleischmann-Pons experiment to be due to deuteron-deuteron fusion:

Huizenga's three miracles

1. Fusion rate miracle; how can the Coulomb barrier be overcome?

2. Branching ratio miracle; even if two deuterons manage to get together, you would expect reactions to produce n+t and p+^{3}He

3. Concealed product miracle; and if somehow ^{4}He is produced (which normally involves a 24 MeV gamma ray), then to be consistent with experiment the gamma rays have to be absorbed somehow

From *Nature*

"… **the cold fusion fuss is discreditable to the scientific community as a whole.** The reasons are plain. First, it has licensed magic in the particular sense that reports of remarkable phenomena – it could be unicorns next – claim equal credence even when they fly in the face of expectation. Second, by extension, it has shown up in the frailty of collective confidence in theoretical science; why else should so many serious people be bamboozled for so long? …"

Sir John Maddox,
Nature **344** 365 (1990).

The excess heat effect in the Fleischmann-Pons experiment was judged not to be real in 1989 and 1990

Take away message

- Many good scientists could not confirm the excess heat effect
- Nothing in condensed matter physics or nuclear physics to indicate that the effect should occur
- Huizenga's three miracles encapsulate basic theory challenge
- Implication is that experiment result must be wrong

- Or, if it is not wrong, then something broken at fundamental level with condensed matter and nuclear physics

Positive results

But work continued…

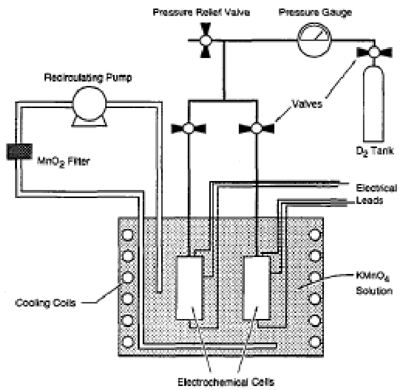
It is almost 23 years after the announcement. Experimental work and theoretical work was pursued on excess heat and other anomalies in many laboratories, and some work continues today.

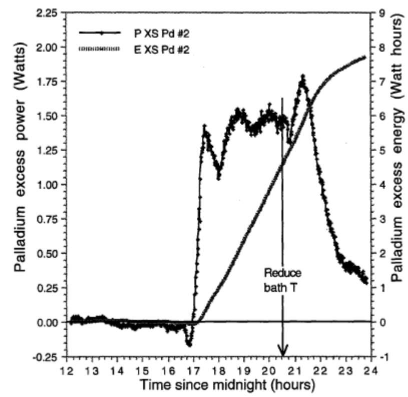
University of Utah
Texas A&M
Stanford (Huggins)
BARC
ENEA Frascati
SRI
SPAWAR
LANL
NRL
Institute of Phys. Chem. and Electrochem., Moscow
Lebedev Institute, Moscow

Tsinghua University, Beijing
Hokkaido University
Osaka University
IMRA Japan
Oak Ridge National Lab
MIT
Portland State
George Washington University
Energetics
JET Energy
UC Berkeley
University of Siena

University of Rome, La Sapienza
University of Milan
National Cold Fusion Institute (Utah)
Luch Institute, Moscow
University of Marseille
University of Torino
University of Missouri (Colmbia)
University of Bologna
U Minnesota (Minneapolis)

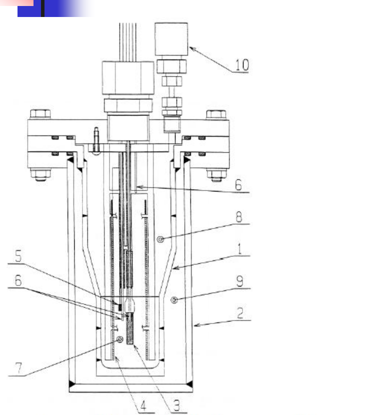
Excess heat at SRI (1989)



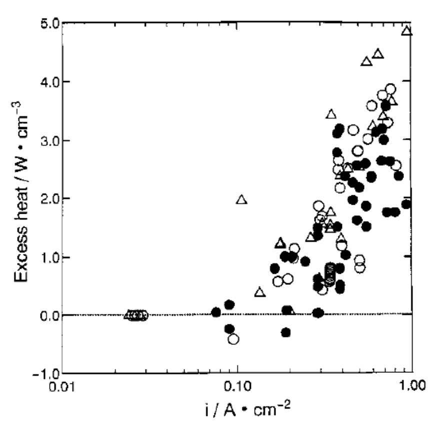


M. C. H. McKubre et al, Development of advanced concepts for nuclear processes in deuterated metals, EPRI Report TR-104195 (1994).

Excess heat at IMRA Japan

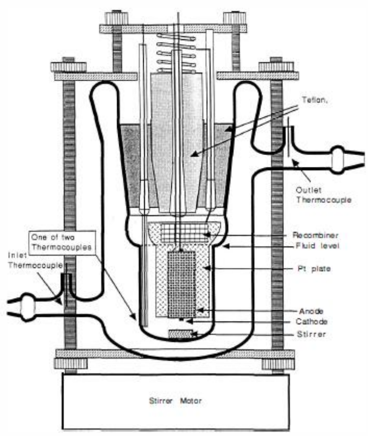


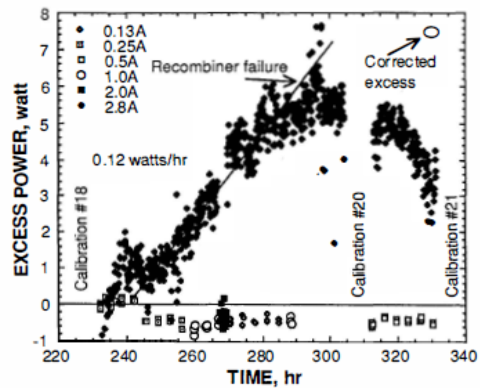
1. Pressure Vessel (SUS316), 2. Vessel (SUS316), 3. Pd Cathode, 4. Fuel Cell Anode
5. RHE (Pt/Pt), 6. Thermocouple, 7. 1 M LiOD, 8. D2 (5-10Kg/Cm2), 9. Air, 10. Pressure Sensor



N. Hasegawa, N. Hayakawa, Y Tsuchida, Y. Yamamoto, and K. Kunimatsu, *Proc.* ICCF4, Vol. I, page 3-1 (1994).

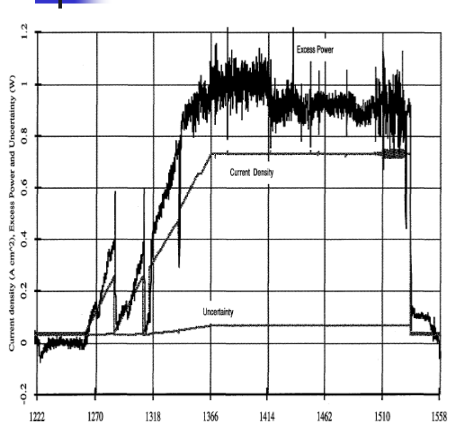
Excess heat at LANL

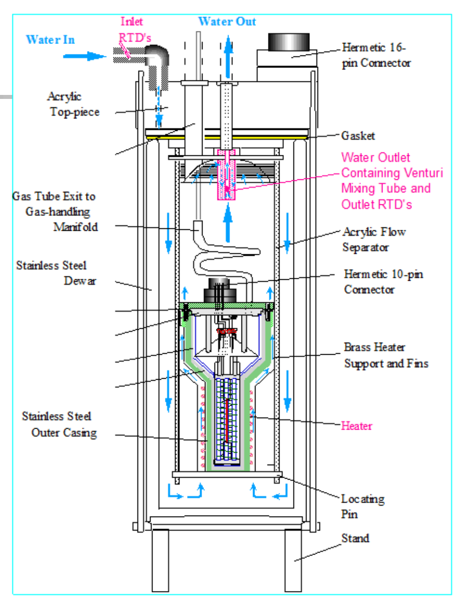




E. Storms, "Measurement of excess heat from a Pons-Fleischmann Cell, Proc ICCF3 p. 21 (1993).

SRI flow calorimeter

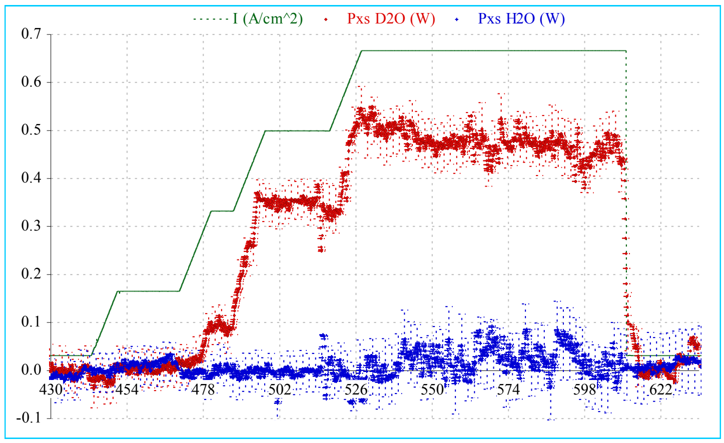




M. C. H. McKubre et al, 1994 EPRI Report

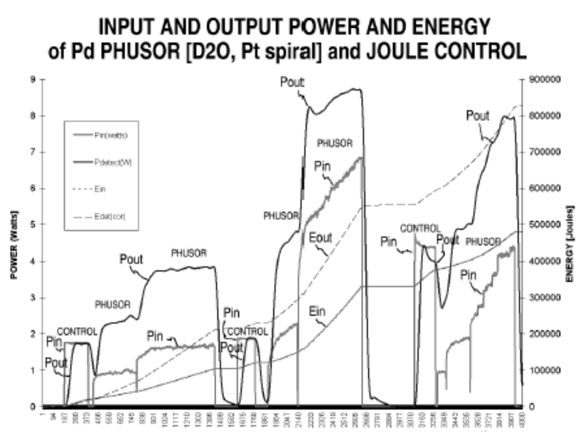
D_2O vs H_2O

P_{xs}(W)



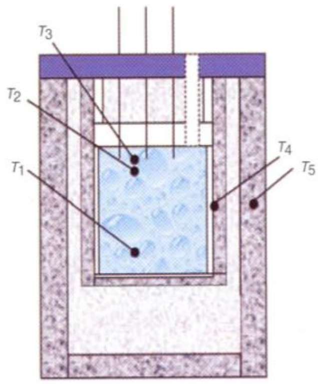
t(hours)

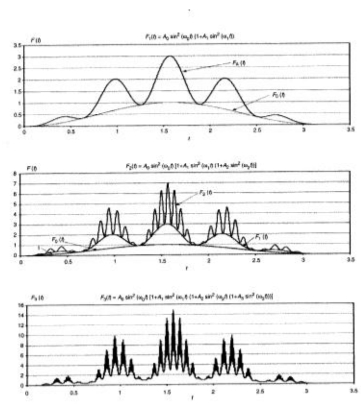
Excess heat at JET



M. R. Swartz, *J. Sci. Expl.* **23** 419 (2009).

Temperatures vs time





I. Dardik et al,
Proc. ICCF11, (2004).

Large energy gain at Energetics, from ICCF11

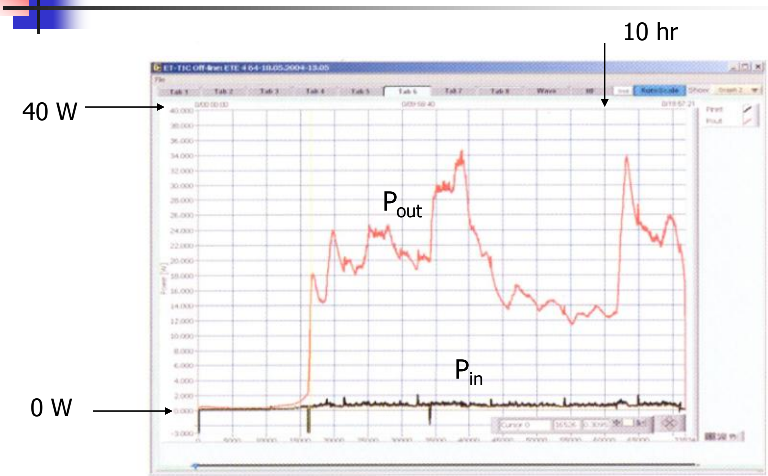


Figure 11. Evolution of net input power (P_{inet}) and output power (P_{out}) in the first loading experiment with foil No. 64.

Letts 2-laser experiment

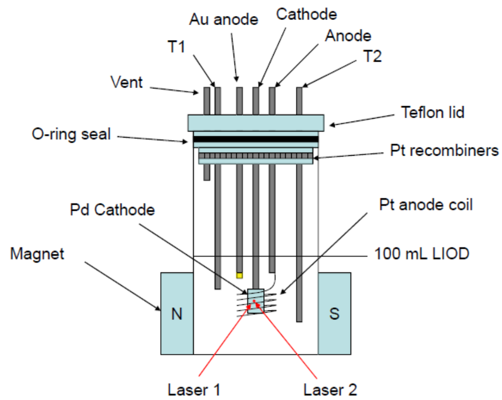
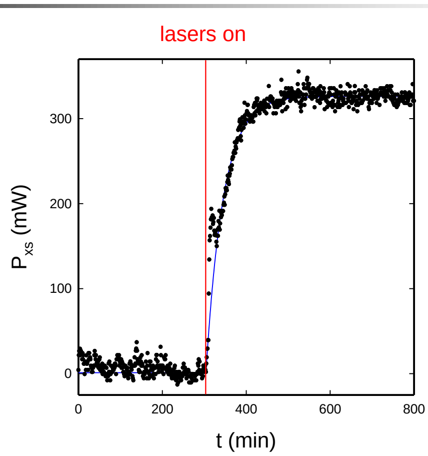


Figure 1. Schematic of the experimental set-up.

D. Letts, D. Cravens, and P.L. Hagelstein, *LENR Sourcebook* Volume 2, ACS: Washington DC. p. 81-93 (2009).

Excess power with 2 lasers

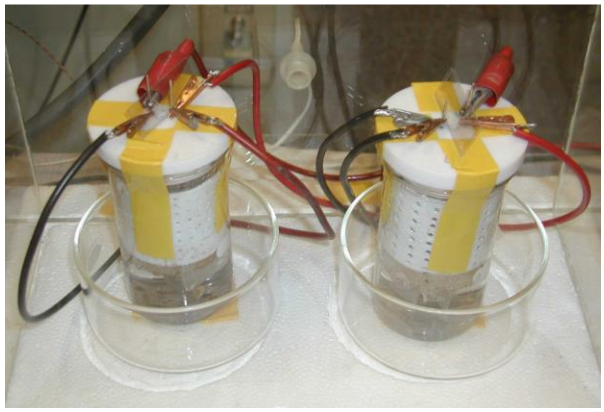


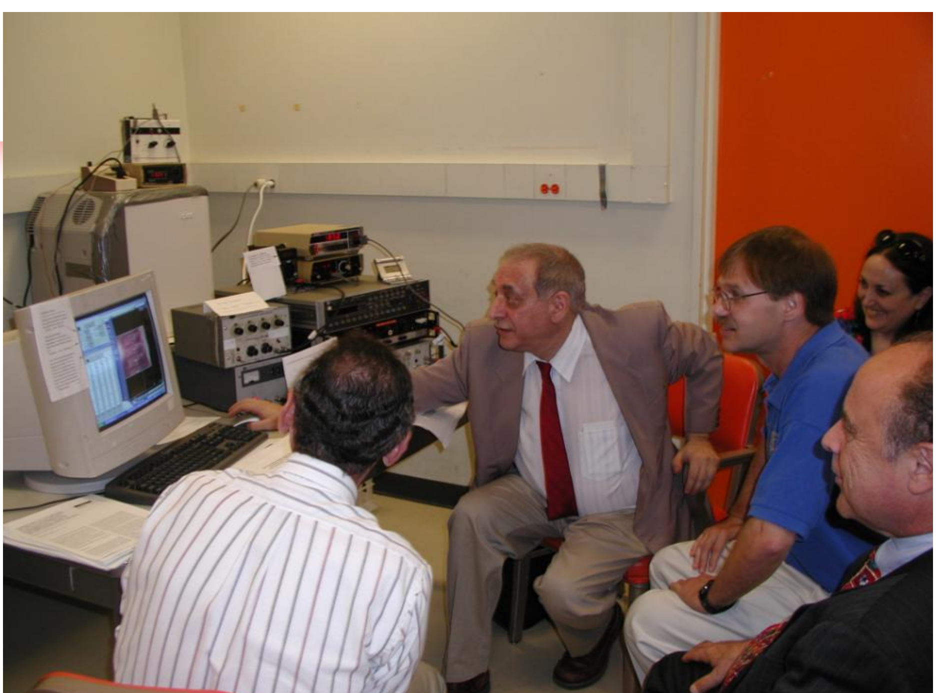
P. Hagelstein, D. Letts and D. Cravens, *J Cond. Mat. Nucl. Sci.* **3** 59 (2010)


Cold Fusion Research


OMEGA
CELL TEMPERATURES
EXPERIMENTAL CELL
CONTROL CELL
CONTROL CELL VOLTAGE
EFFECT OF ELECTROPLATED Pd ON Pd CATHODE
BEFORE/AFTER

Dash demo



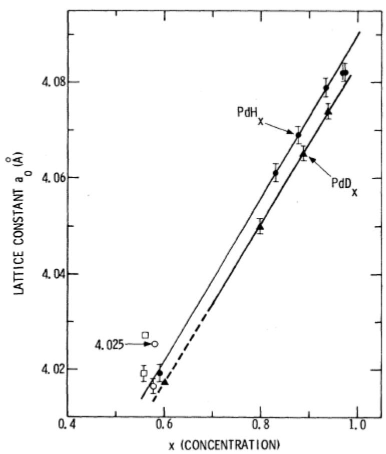


Take away message

•There have been hundreds of positive results in Fleischmann-Pons experiments showing excess heat effect
•Positive results at many labs
•Different kinds of calorimetry used with similar results
•Motivates us to try to understand the experiment better
•Also wonder why so many good labs did not succeed

Lattice expansion

Lattice constant increases with D/Pd loading



a = 3.89 Angstrom for Pd

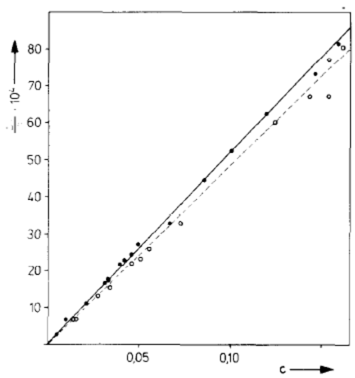
J. E. Schirber and B. Morosin, *Phys. Rev. B* **12** 117 (1975)

Volume change is big effect

$$\frac{V(\mathrm{PdD})}{V(\mathrm{Pd})} = \left(\frac{a(\mathrm{PdD})}{a(\mathrm{Pd})}\right)^3 = \left(\frac{4.08}{3.89}\right)^3 = 1.15$$

This lattice increase effect is very large.

Similar in other metal hydrides



Lattice parameter change $\Delta a/a$ of tantalum versus hydrogen concentration $c = H/Ta$

• Ta/H ∘ Ta/D

H. Pfeiffer and H. *Peisl, Phys. Lett.* **60 A** 363 (1977)

Hydrogen volume in metals

Metal	structure	v_H(Angstrom^3)
V	bcc	2.64
Nb	bcc	3.13
Ta	bcc	2.80
Ni	fcc	2.98
Pd	fcc	2.80

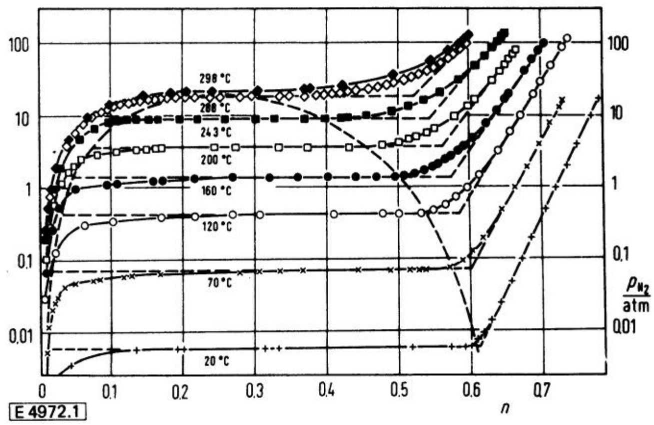
Assumes dilute hydrogen; data from Y Fukai, The hydrogen-metal handbook, Springer-Verlag (1993)

Take away message

- Loading deuterium into Pd stretches the lattice
- The volume increase for PdD is 15%
- We would expect an increase in the elastic energy due to this volume increase

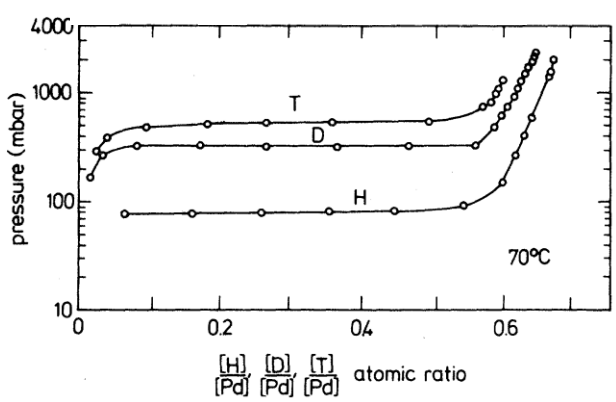
Phase diagram

Phase diagram for PdH



E. Wicke and J. Blaurock, *Ber. Besunges Phys. Chem.* **85** 1091 (1985)

PdH, PdD, and PdT



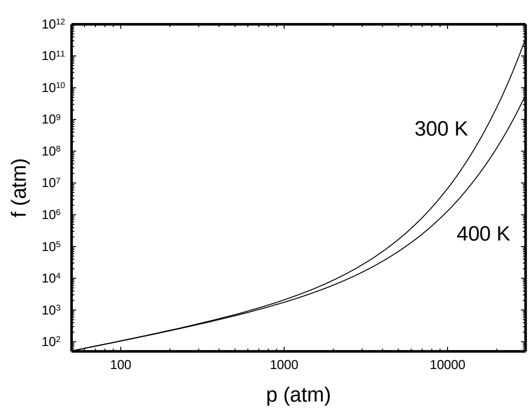
R. Lasser, *Phys. Rev. B* **26** 3517 (1982).

Gas chemical potential relation

$$\mu_D(p) = \mu_0 + \frac{1}{2}k_B T \ln \frac{f(p)}{p_0}$$

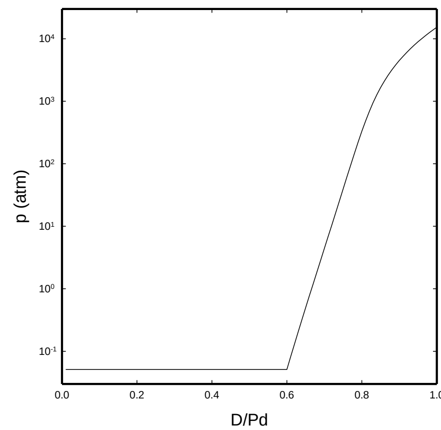
Can use this to get the chemical potential for D in metal from pressure versus loading curve if the fugacity is available

Fugacity

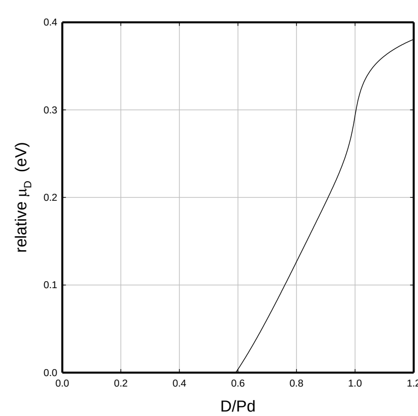


Computed from volume data tabulated by H. Hemmes et al (1986) in Fukai book

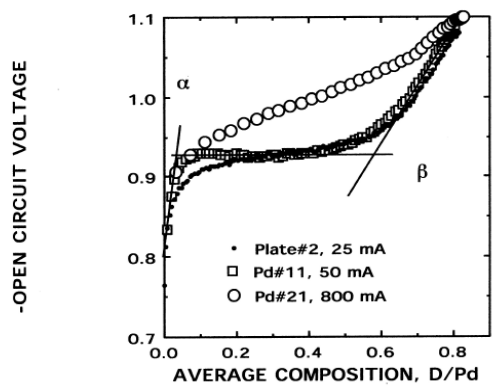
Pressure vs loading



Chemical potential model



Relation to V_{oc}



E. Storms, Proc. ICCF7 p. 356 (1998)

Take away message

- Pd can absorb hydrogen, deuterium and tritium from gas
- The more pressure, the more loading
- Easy to load up to D/Pd = 0.6 near room temperature
- Increasingly hard to load above D/Pd = 0.6
- Notion of H,D chemical potential in connection with H,D loading

Deuterium diffusion in PdD

Diffusion model

Textbook diffusion model:

$$D = D_0 (1-x) e^{-\Delta E / k_B T} \rightarrow 5.5 \times 10^{-7} (1-x) \ \frac{\text{cm}^2}{\text{sec}} \text{ at 300K}$$

barrier factor

prefactor

site occlusion factor

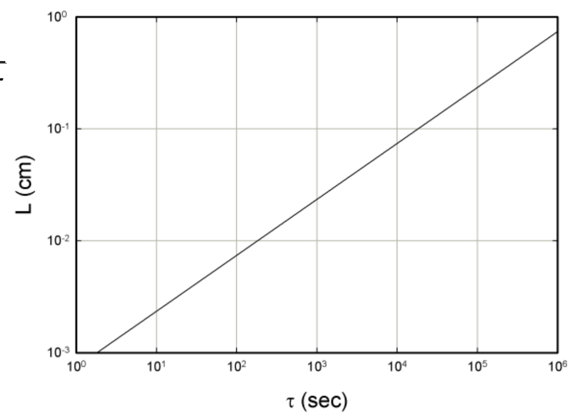
Fukai's book, PdD: D_0 = 1.7 x 10^{-3} cm^2/sec
ΔE = 206 meV

Y. Fukai, *The Metal-Hydrogen System*, Springer-Verlag (1993).

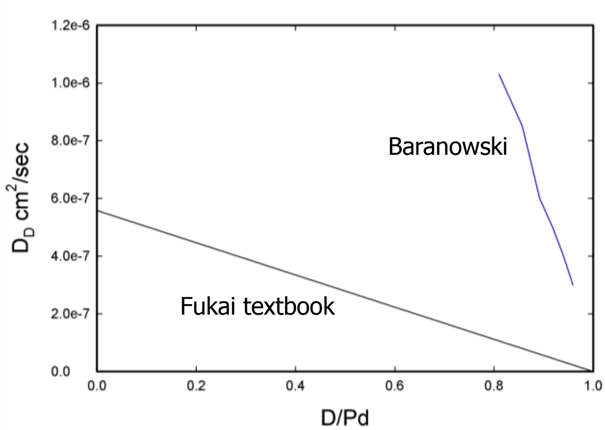
D Diffusion is fast in Pd

At low loading, the diffusion coefficient is $D = 5.5\times10^{-7}\ \dfrac{\text{cm}^2}{\text{sec}}$

$$L = \sqrt{D\tau}$$

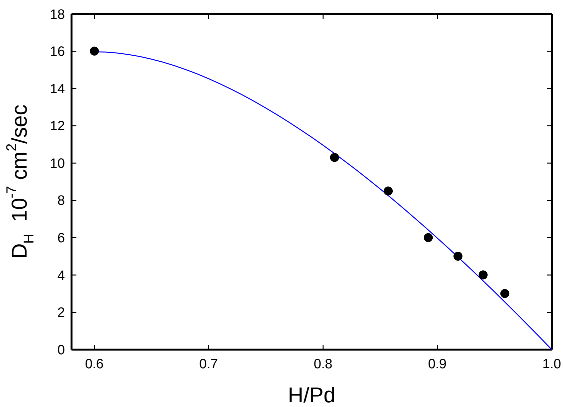


Baranowski expt's

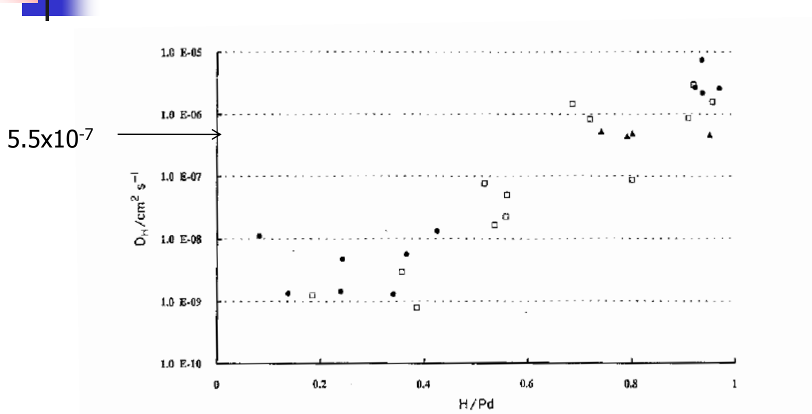


B. Baranowski et al, *J Less Common Metals* **158** 347 (1993)

Fit to data in β phase



Experiment



G. Mengoli and M. Fabrizio, *J Electroanal Chem* **350** 57 (1993)

Onsager Diffusion

Osager's idea…

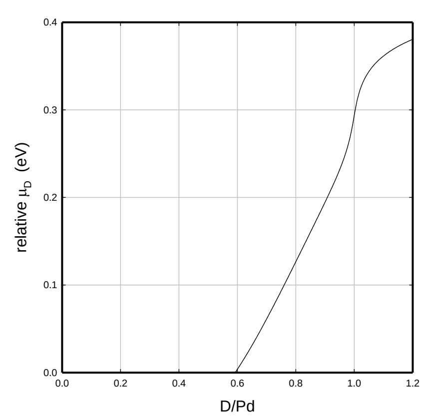
Forces normally come about as gradient of potential

$$F = -\,\mathrm{grad}(\Phi)$$

Then why not the same thing for chemical potential?

$$F_D = -\,B\,\mathrm{grad}(\mu)$$

Chemical potential model



Could use hybrid approach

Diffusion model in α-β region with flat chemical potential:

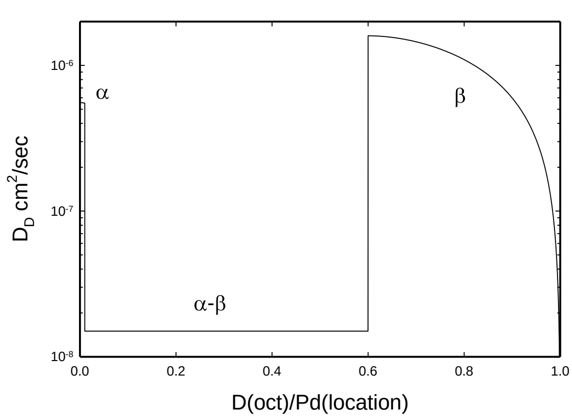
$$\frac{\partial}{\partial t} n_D = \nabla \cdot \left(D \nabla n_D \right)$$

Onsager-type diffusion model for higher loading:

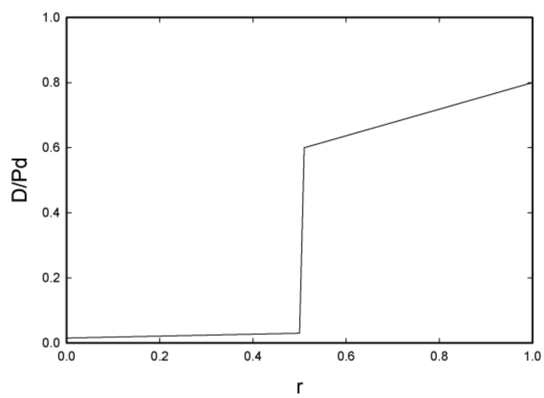
$$\frac{\partial n_D}{\partial t} = \nabla \cdot \left(B n_D \nabla \mu_D \right)$$

Also possible to adopt diffusion model throughout

PdD diffusion model at 300 K

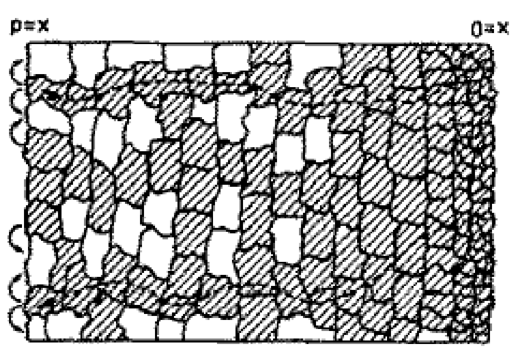


D profile at constant flux



Get discontinuity in loading profile

Grain effects



H. Zuchner and T. Rauf, *J. Less Common Metals* **172-174** 816 (1991)

Take away message

- D diffusion in Pd is “fast”
- But slow in miscibility gap due to flat chemical potential
- Get discontinuity in loading at miscibility gap
- Large grains split into smaller grains during loading
- Grain boundary diffusion important

Resistance ratio

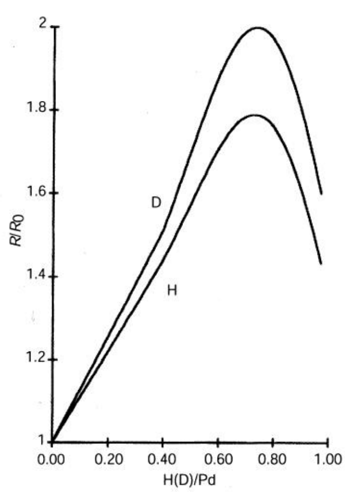
Keeping track of D/Pd ratio

The D/Pd loading ratio was thought to be important by Fleischmann

Several ways to keep track of it:

- Lattice parameter
- Cathode volume
- Cathode weight
- Orphan oxygen
- Electrical resistance

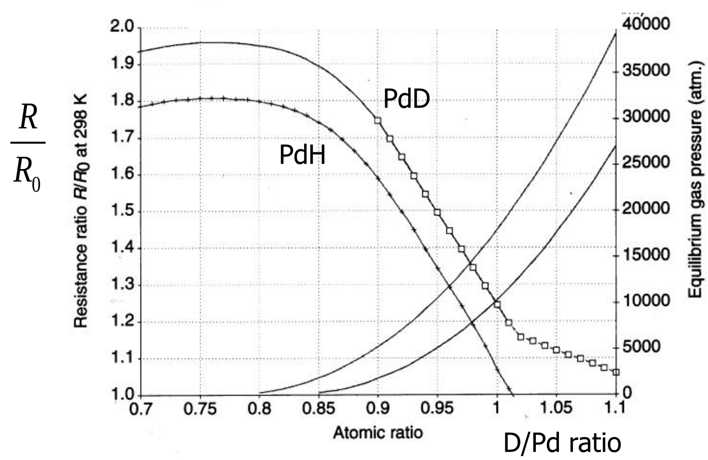
PdH, PdD Resistance ratio



Calibration curve used in the early years

M. McKubre and F. Tanzella, *Proc. ICCF12* (2005).

Updated resistance ratio



M. McKubre and F. Tanzella, *Proc. ICCF12* (2005).

Take away message

- Would like to keep track of D/Pd ratio in the cathode
- Resistance depends strongly on D/Pd ratio
- Provides a convenient way to monitor in real time
- Accurate calibration available
- Older calibration overestimates loading

Day 1 summary

- Excess heat effect in Fleischmann-Pons experiment is controversial
- Inconsistent with nuclear physics
- Inconsistent with condensed matter physics
- Most early confirmation experiments did not see excess heat effect
- Subsequent experiments provide very many positive results
- Begin here to think about Fleischmann-Pons experiment seriously
- Lattice expansion in PdD
- Hard to load deuterium in Pd above D/Pd near 0.80
- Can use resistance ratio to measure loading

Warning

Working in this field at this time can destroy your career.

Being interested may be damaging to your personal and professional life.