

Low Energy Nuclear Reaction Investigations at the Naval Research Laboratory

Off the chart!

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Applying the Scientific Method to Understanding Anomalous Heat Effects: Opportunities and Challenges

18th International Conference on Condensed Matter Nuclear Science – Columbia, Missouri
July 22-26, 2013



The Prime Directive



Simplified steps in scientific method:

1. Make some observation
2. Make-up some hypothesis **refuting**
3. Collect a lot of data ~~supporting~~ that hypothesis
4. Make a few presentations and write a few papers
5. Win a Nobel prize or make lots of money

In science, the Prime Directive is to
prove yourself wrong!

Logically, something only can be falsified

“There is no surer way to screw-up an experiment
than to be certain of its outcome”

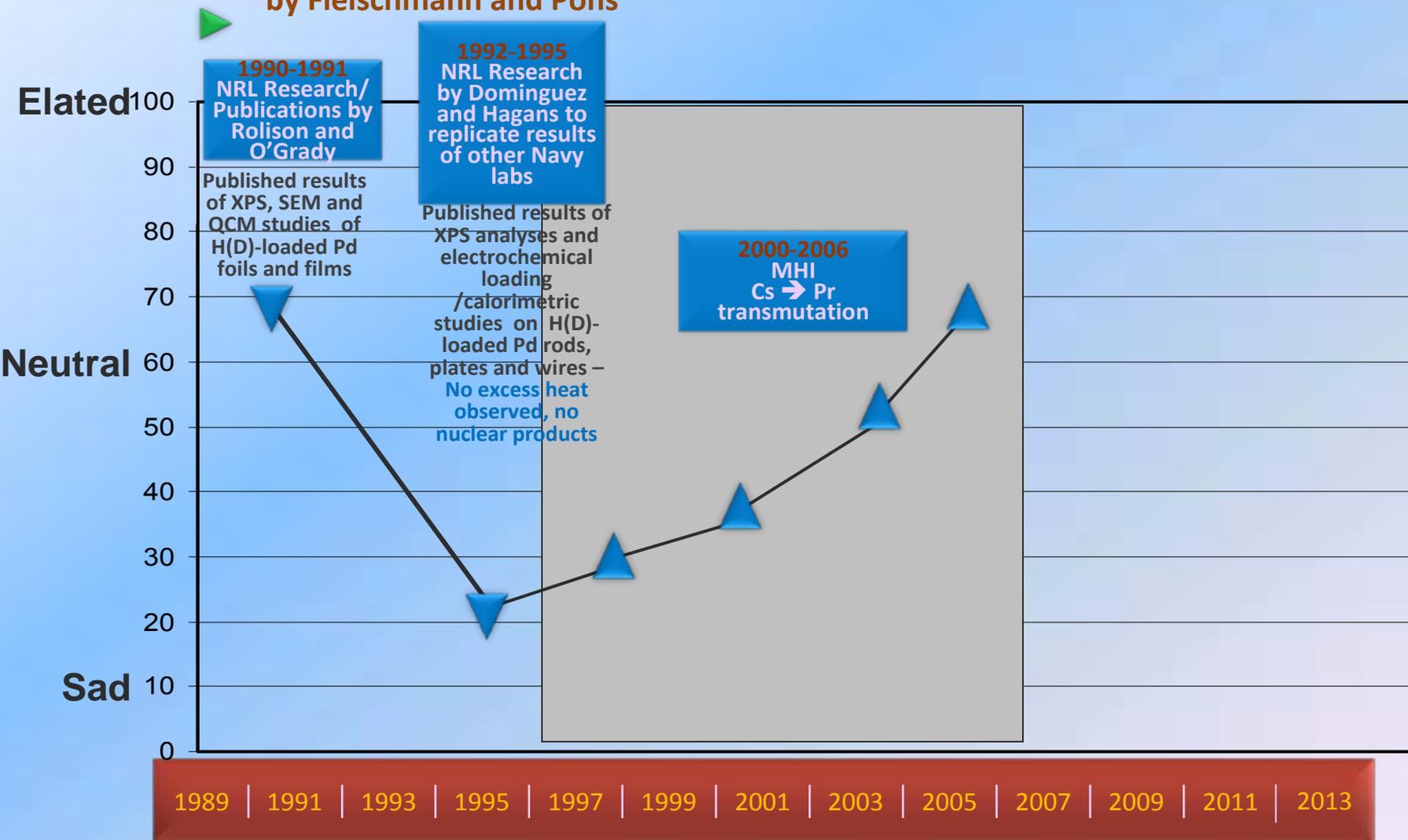
Stuart Firestein – **Ignorance: How it Drives Science**

What Has NRL Observed?



Chart of Happiness

Announcement/Publication
by Fleischmann and Pons



***Prima-Facie* Evidence that LENR Occurs**

Something non-conventional is happening



1. Radiation on demand

2. Transmutation
 - Production of a radioactive isotope
 - Isotopic pattern change
 - Production of element not originally present

3. Definitive demonstration of excess energy
 - Self-sustained energy production beyond known chemistry

Radiation on Demand



Short answer is
no definitive signal

Nal Scintillation Detectors



Advantages:

- The most sensitive over a broad range of energies
- Rugged and non-cooled
- Provide some spectral information – but not highly resolved

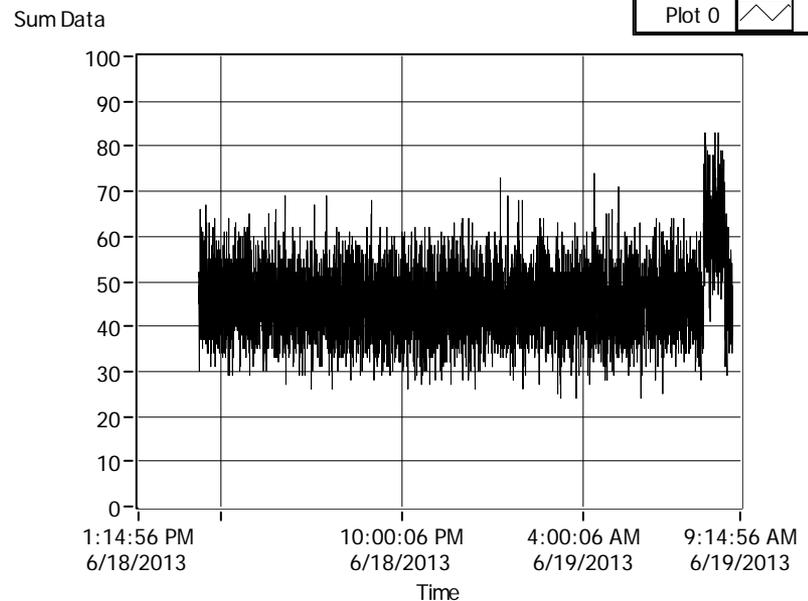
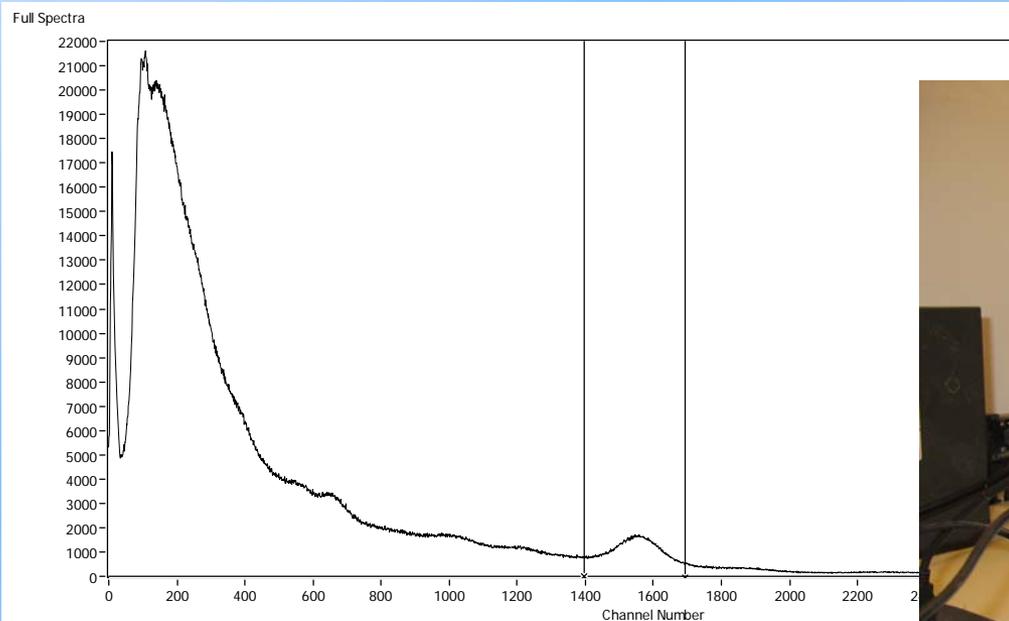


Nal Scintillation Detectors

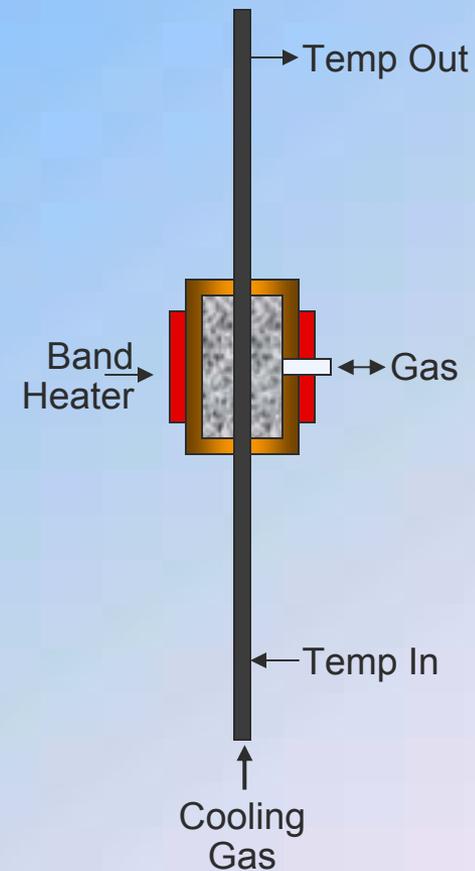
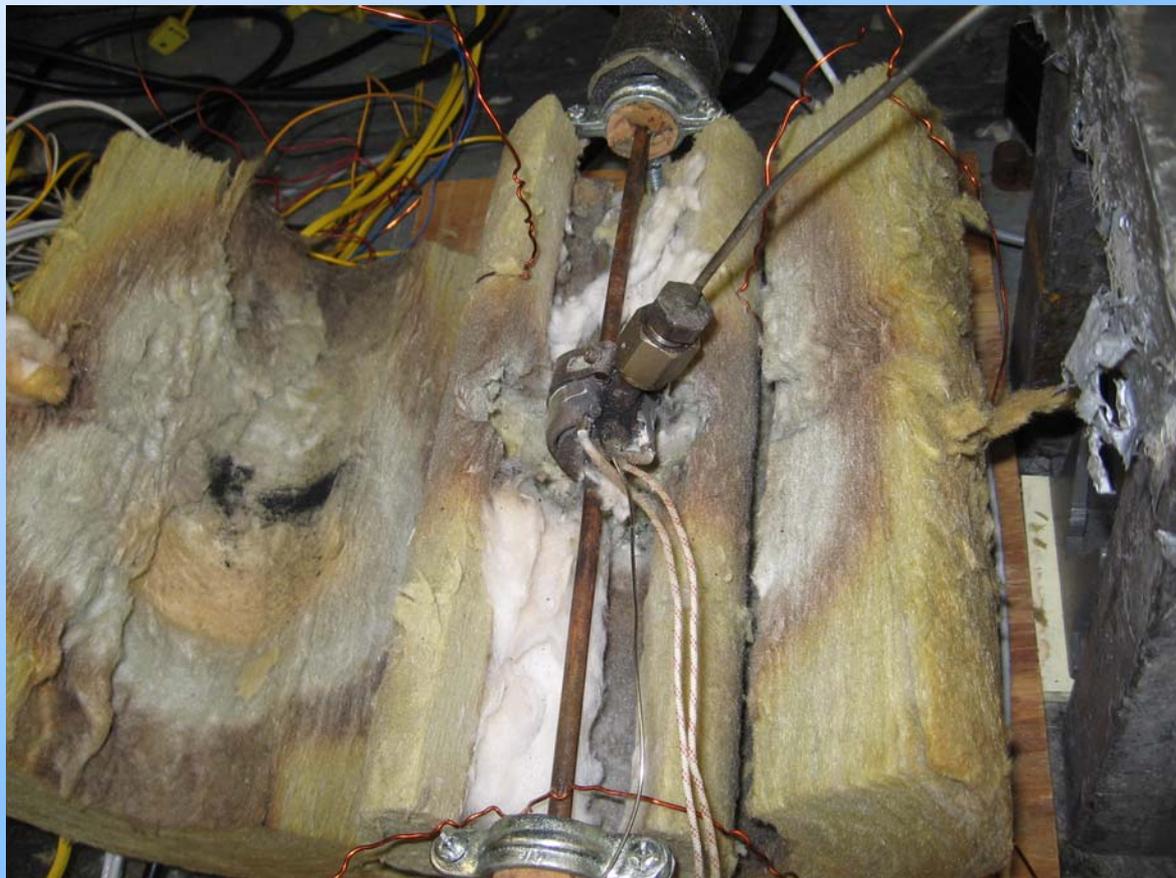


Advantages:

- The most sensitive over a broad range of energies
- Rugged and non-cooled
- Provide some spectral information – but not highly resolved



“Rossi-Like” Device



Yan Kurochov, “Method and Apparatus for Energy Generation”, WO 98/15986

➡ Theory requires large thermal gradient

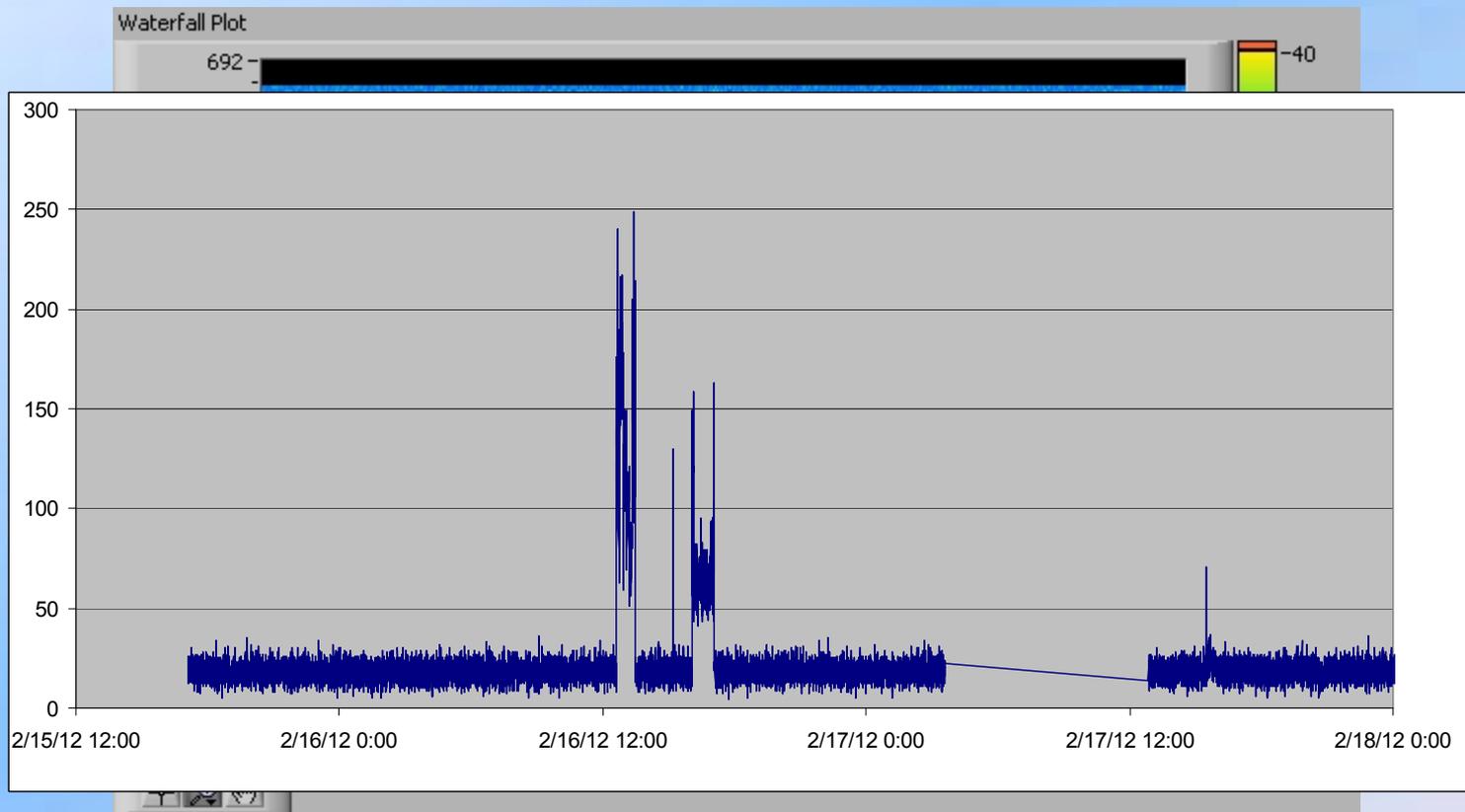
Rossi-Like Device





Radiation as Measured by a NaI Detector

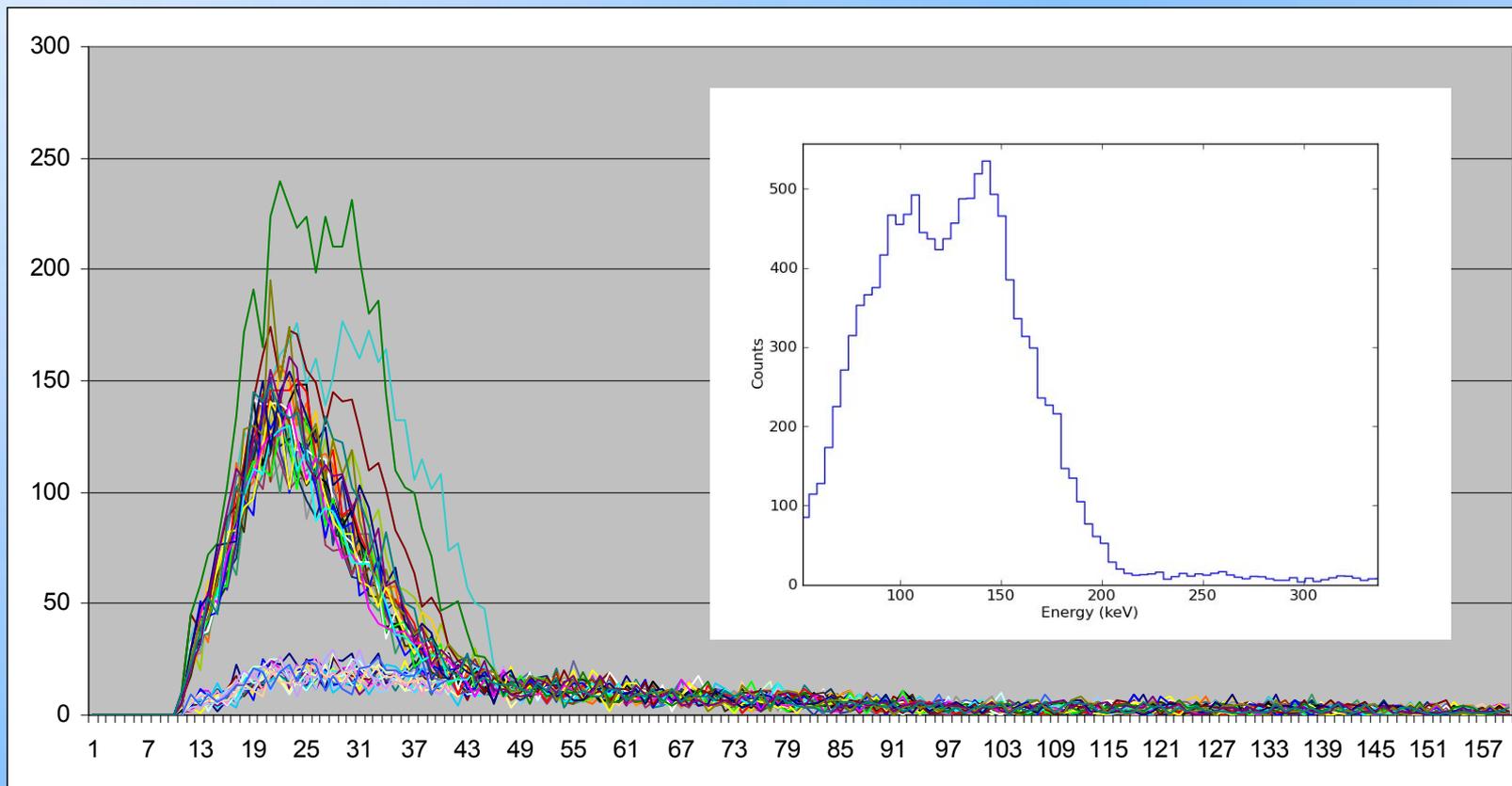
Les Case material



- Clear indication of three radiation events
 - 12:30, 16:07, 15:38 next day



Radiation as Measured by a NaI Detector Spectra

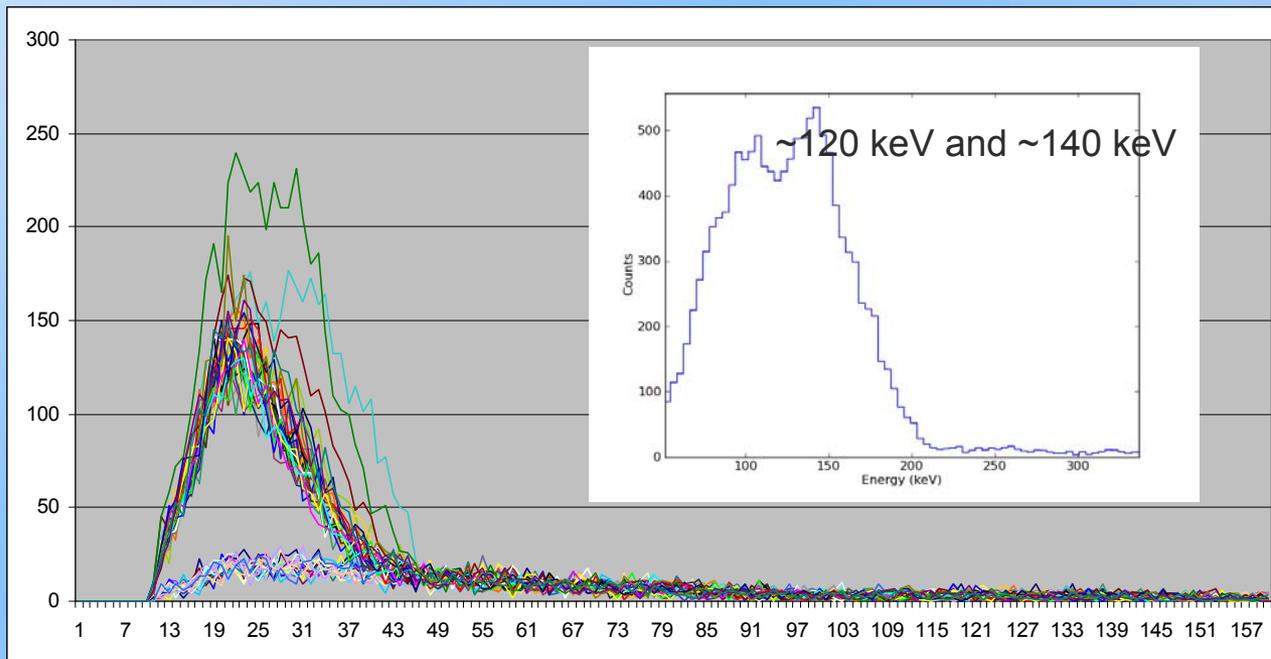


- Spectra substantially above background
 - Looks like two peaks at ~120 keV and ~140 keV



Radiation as Measured by a NaI Detector

Explanation



- Someone using a source nearby? – no

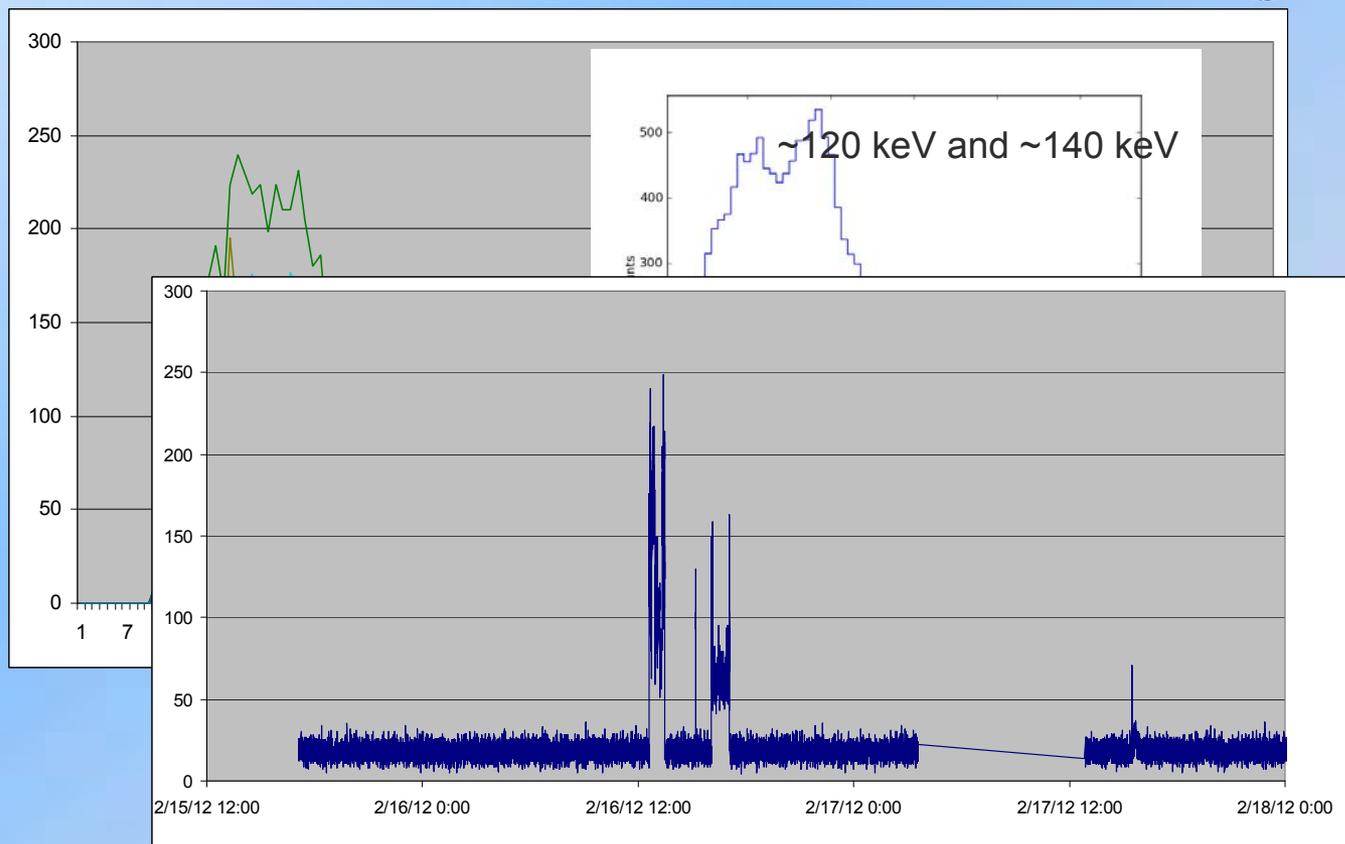
Something not right:

- System running for several days before radiation appeared
 - Nothing being done to change conditions
- Radiation never seen before
 - Conditions not correct?



Radiation as Measured by a NaI Detector

Explanation



- ^{99m}Tc
- $T_{1/2}$ 6 h
- Gamma at 140.3 keV

Conclusions

Radiation on Demand



- Evidence for radiation weak
- For NRL, Chart of Happiness for radiation on demand is:

Off the chart!

Transmutation

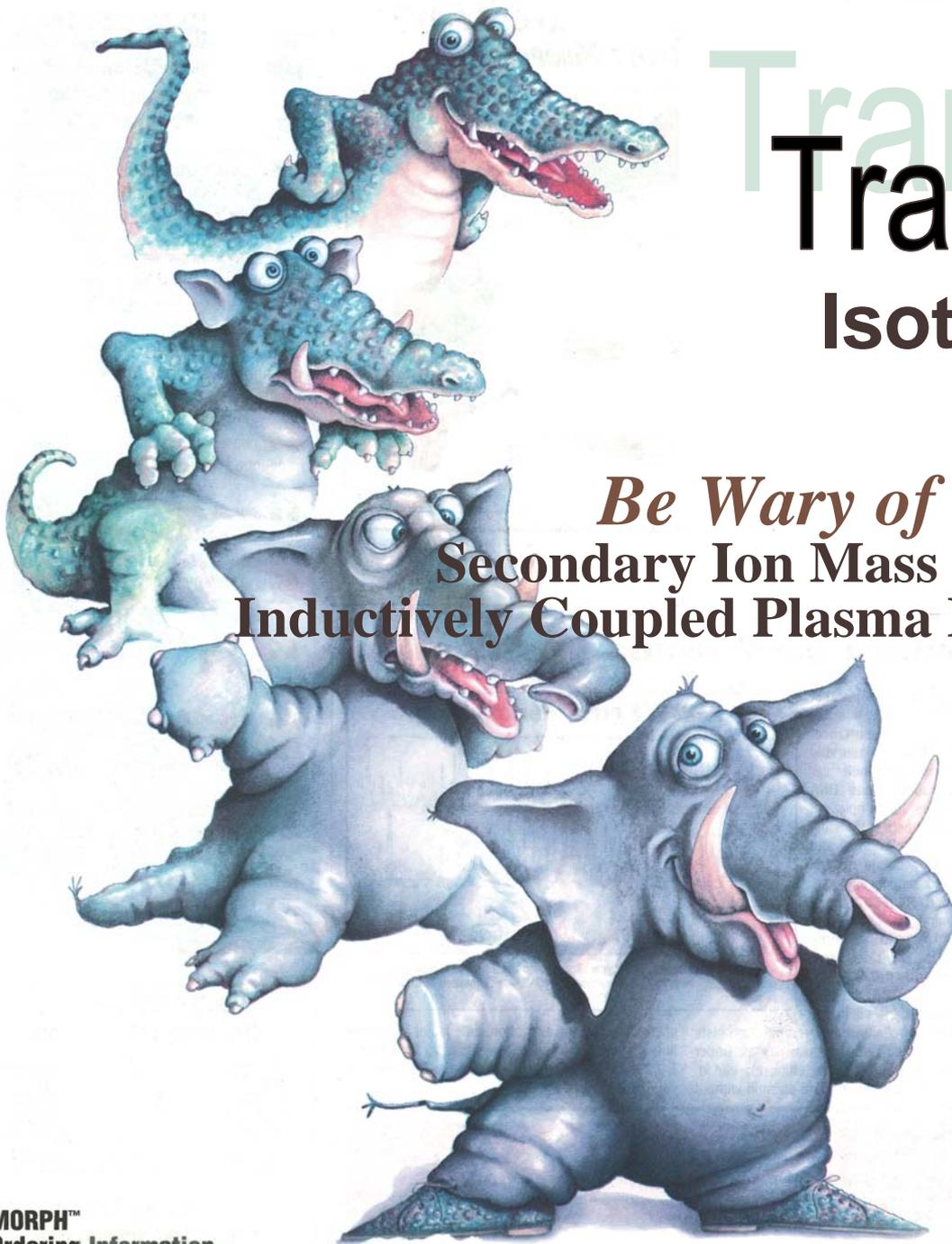
Isotopic Anomalies

Be Wary of Cluster Ions

Secondary Ion Mass Spectrometry (SIMS)

Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

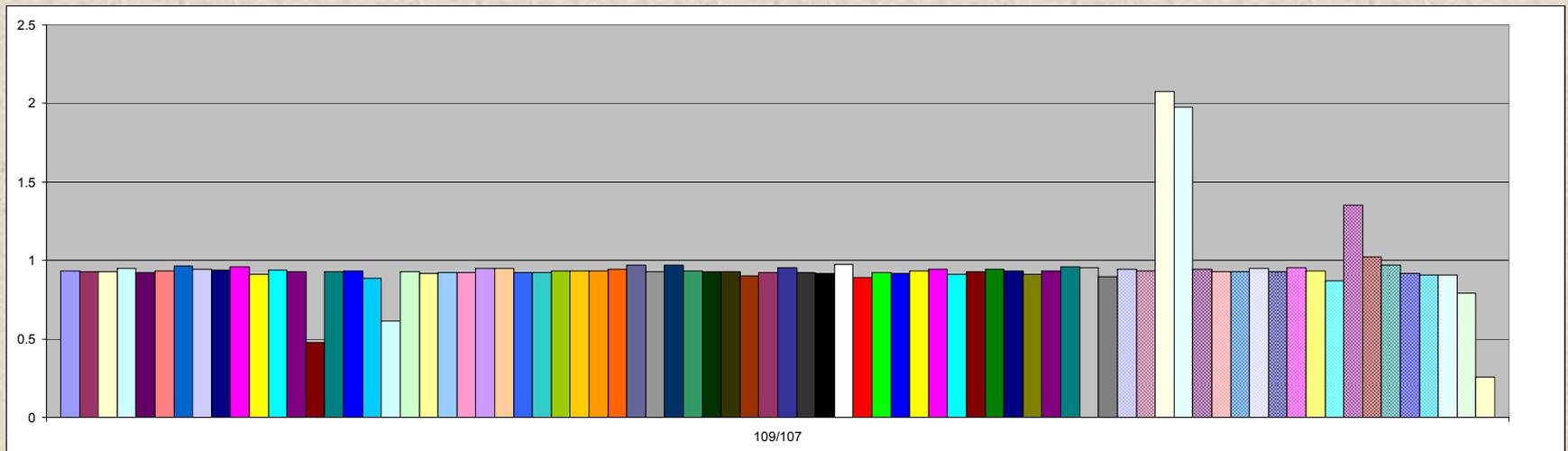
Short answer is
no definitive result



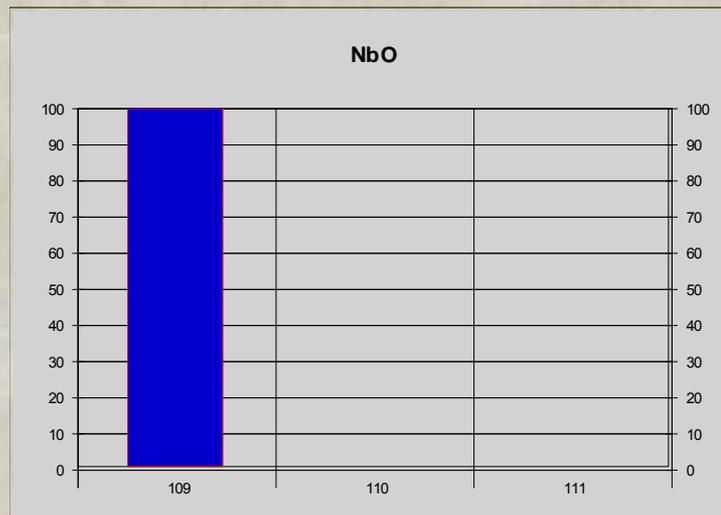
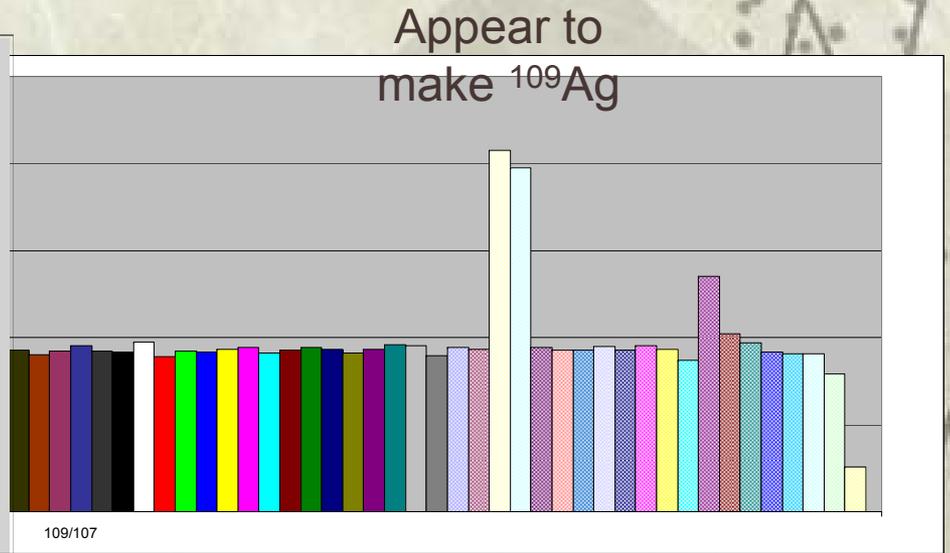
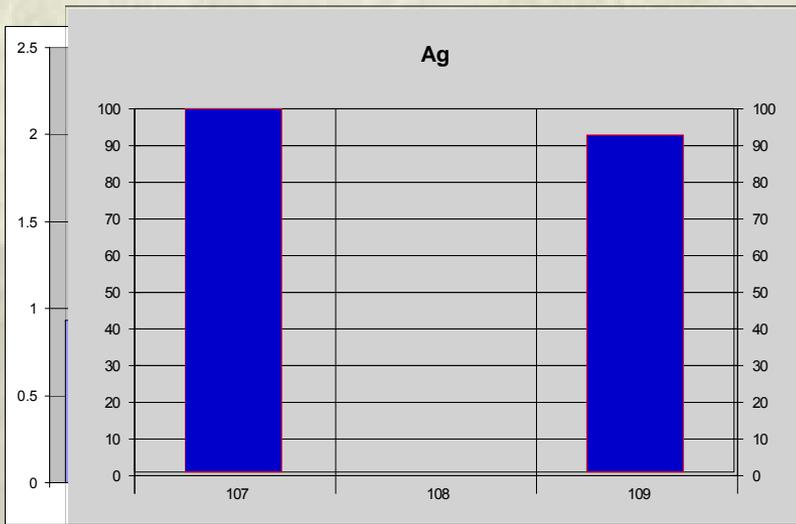
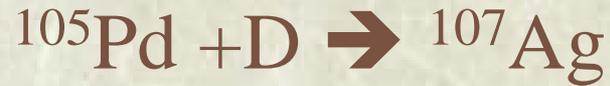
Speculation



- ❖ Silver alleged to be created in some heat producing reactions
 - Two isotopes ^{107}Ag and ^{109}Ag
- ❖ BUT Silver is a trace contaminant in palladium
- ❖ How does this ratio vary across samples by ICP-MS?
 - How does ICP-MS Work?

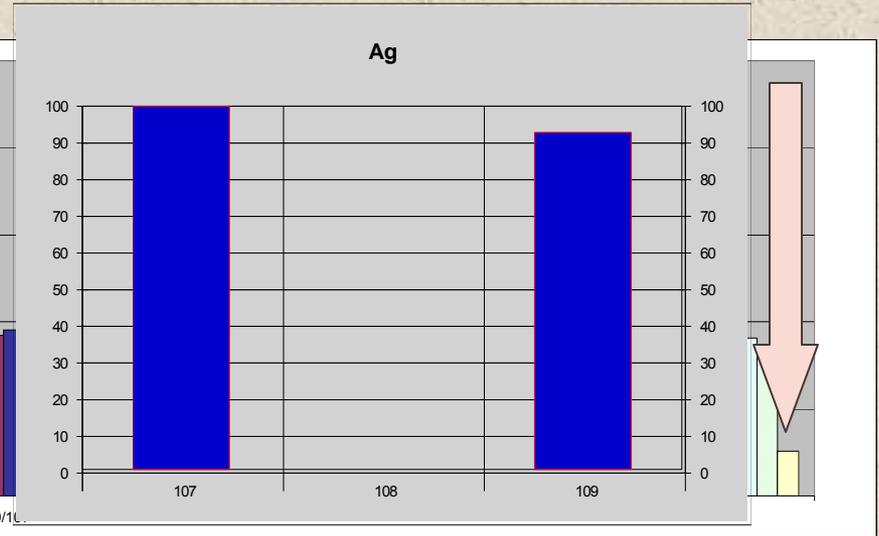
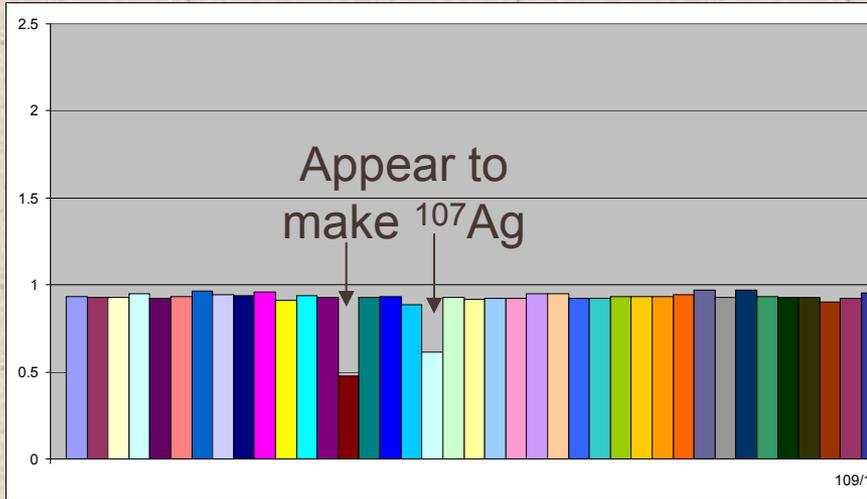


Speculation

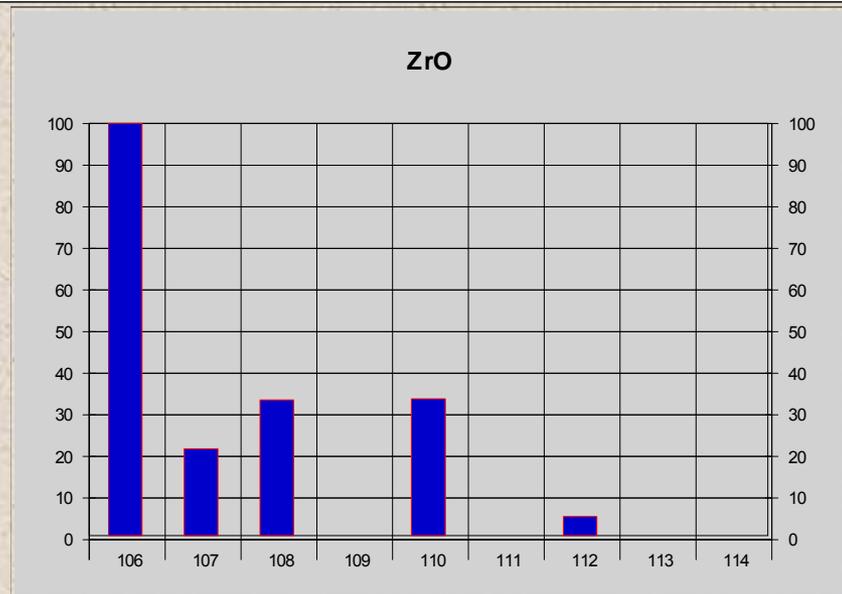


- ❖ Niobium present
- ❖ m/z 109 Likely NbO interferent
- ❖ Good Thing because nuclear chemistry unclear

Speculation

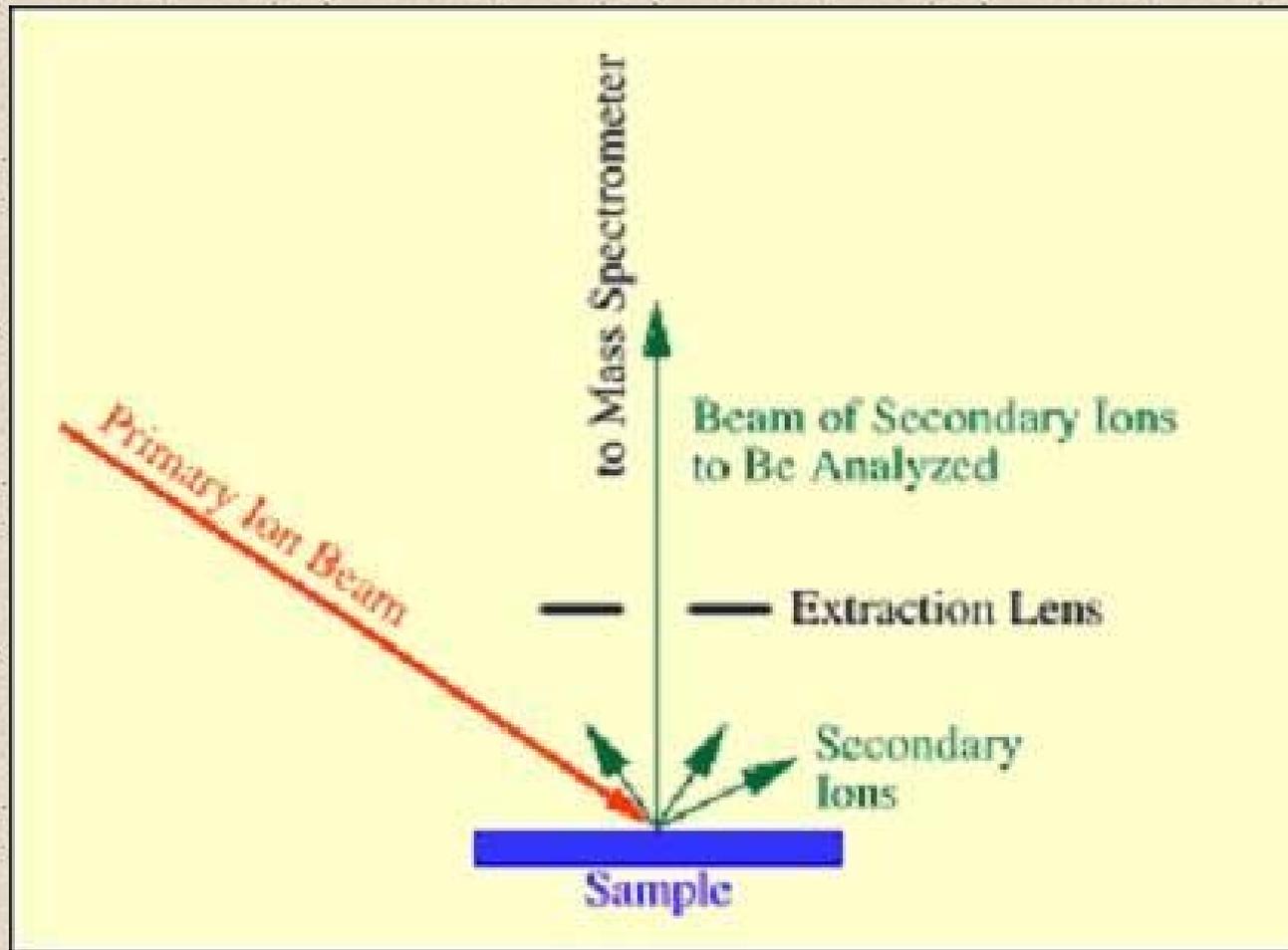


- ❖ Zirconium present
- ❖ m/z 107 Likely ZrO interferent



Be Wary of Cluster Ions

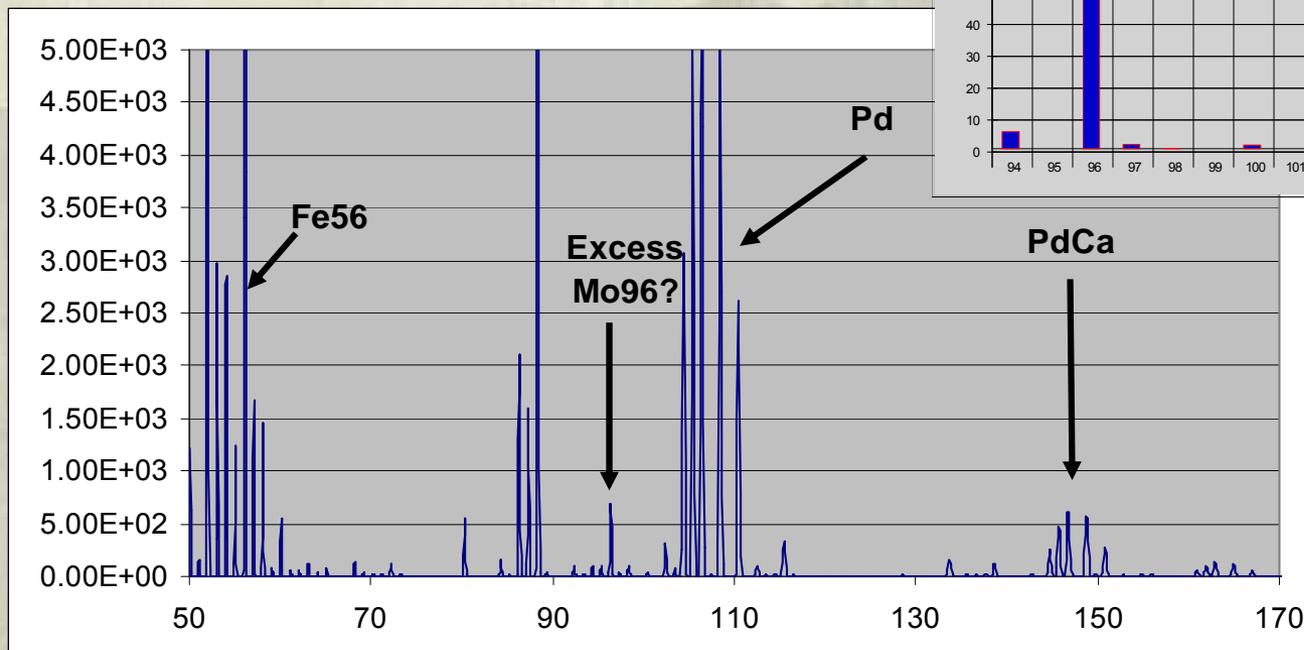
Secondary Ion Mass Spectrometry (SIMS)



Be Wary of Cluster Ions

Secondary Ion Mass Spectrometry (SIMS)

❖ Example: was ^{96}Mo due to a cluster ion?



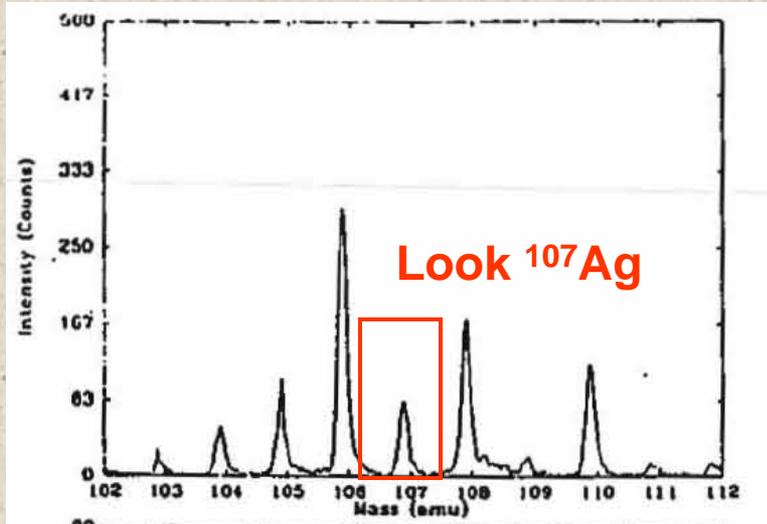
Speculation was $^{88}\text{Sr} + 4\text{D} \rightarrow ^{96}\text{Mo}$

❖ Observe Pd+40 (likely Ca), why not $^{56}\text{Fe} + ^{40}\text{Ca} \rightarrow \text{mass } 96$

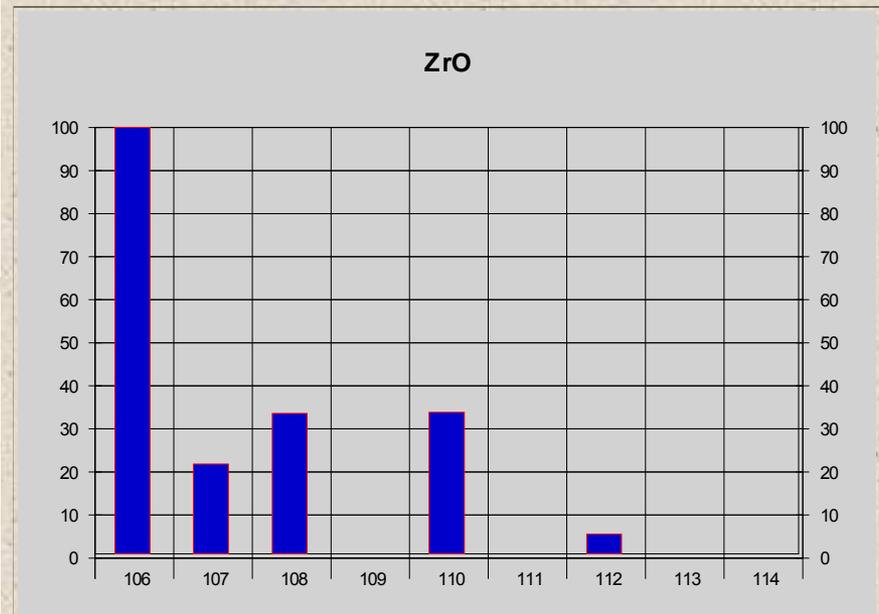
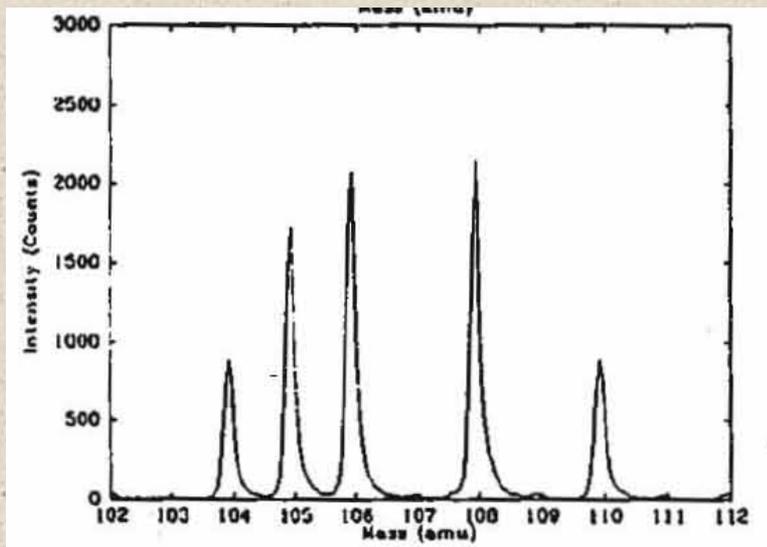
Be Wary of Cluster Ions

Secondary Ion Mass Spectrometry (SIMS)

Data from: D.R. Rolison and W.E. O'Grady, "Mass/charge anomalies in Pd after electrochemical loading with deuterium", EPRI-NSF Workshop on Anomalous Effects in Deuterated Metals, Oct 16-18, 1989.



- ❖ Observe large ¹⁰⁶Pd enhancement
- ❖ Zirconium present
- ❖ *m/z* 106 Likely ZrO interferent



Conclusions for **Transmutation based on isotopic patterns**

- ❖ Molecular interferences will confuse isotopic patterns
 - SIMS be very wary
 - ICP-MS better but not perfect
- ❖ For NRL, Chart of Happiness for Transmutation based on isotopic patterns is:

Off the chart!

Transmutation

*Evaluation of the Claim of
Transmutation of Cesium to
Praseodymium with Mitsubishi
Heavy Industries (MHI)*



Summary of MHI Claims

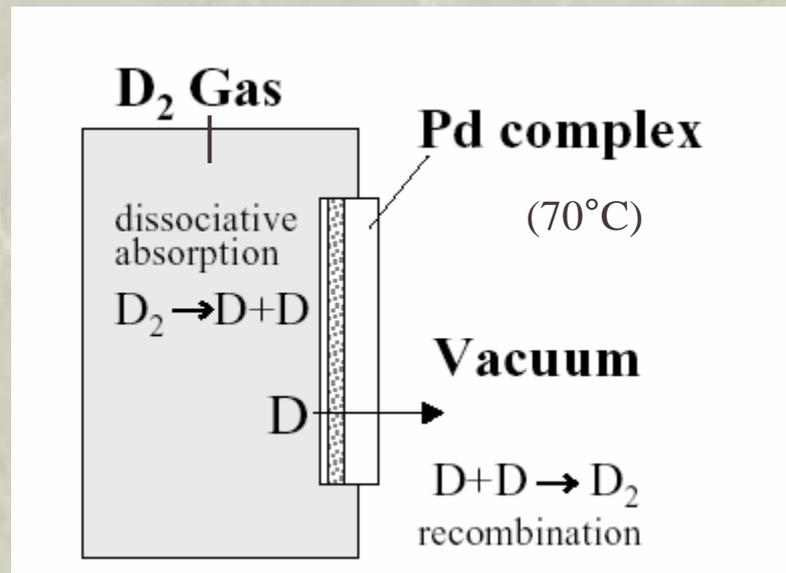
- ❖ By permeating Deuterium through a Pd complex foil, various elemental transmutations can be made to happen

– Reported transmutations:

• $^{88}\text{Sr} \rightarrow ^{96}\text{Mo}$ Addition of 4D

• $^{133}\text{Cs} \rightarrow ^{141}\text{Pr}$ Addition of 4D

• $^{137}\text{Ba} \rightarrow ^{149}\text{Sm}$ Addition of 6D

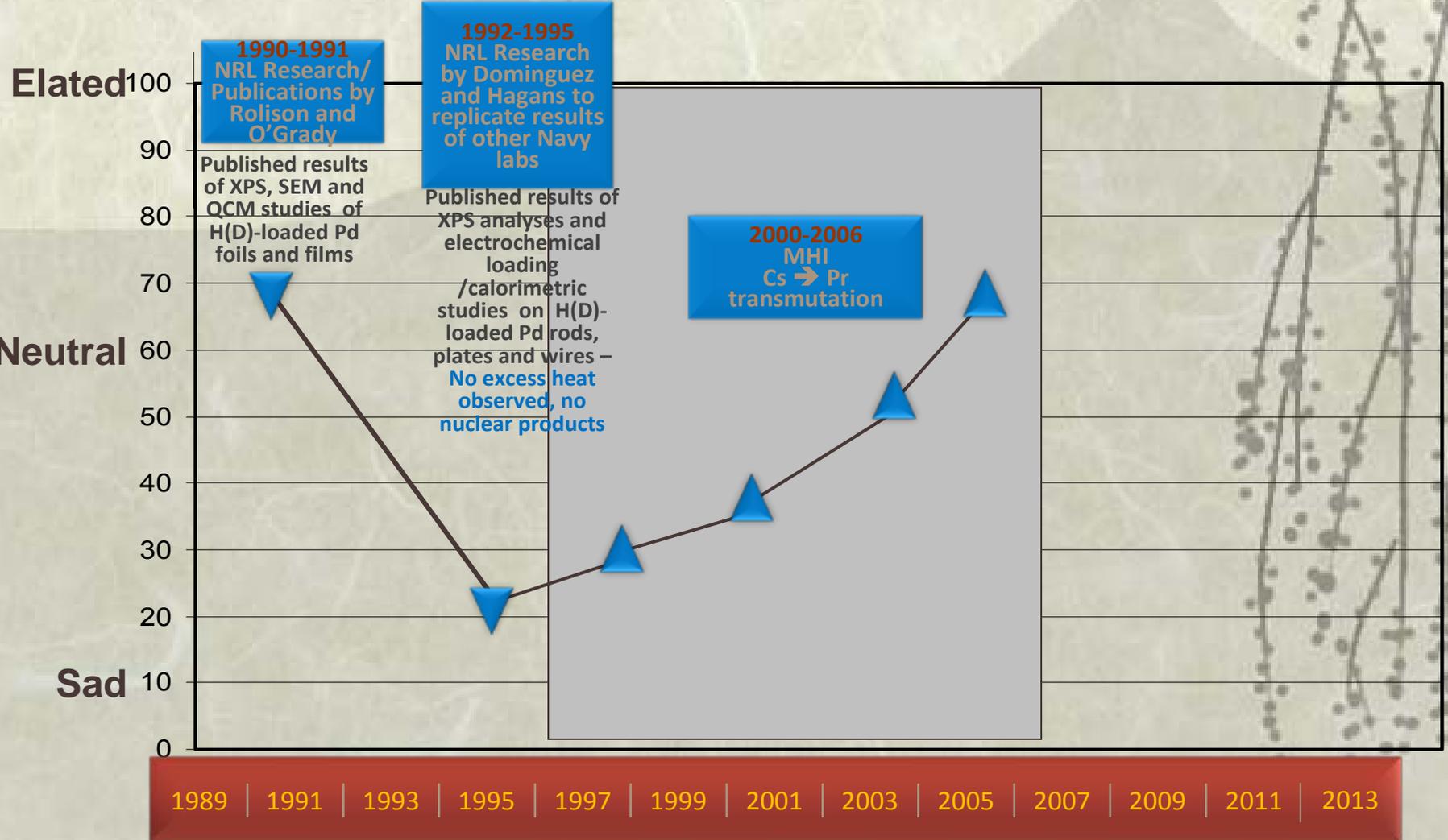


Y. Iwamura, M. Sakano and T. Itoh, "Elemental Analysis of Pd Complexes: Effects of D_2 Gas Permeation", *Jpn. J. Appl. Phys.*, **41** (2002) 4642–4650.

What Has NRL Observed?

Chart of Happiness

Announcement/Publication
by Fleischmann and Pons

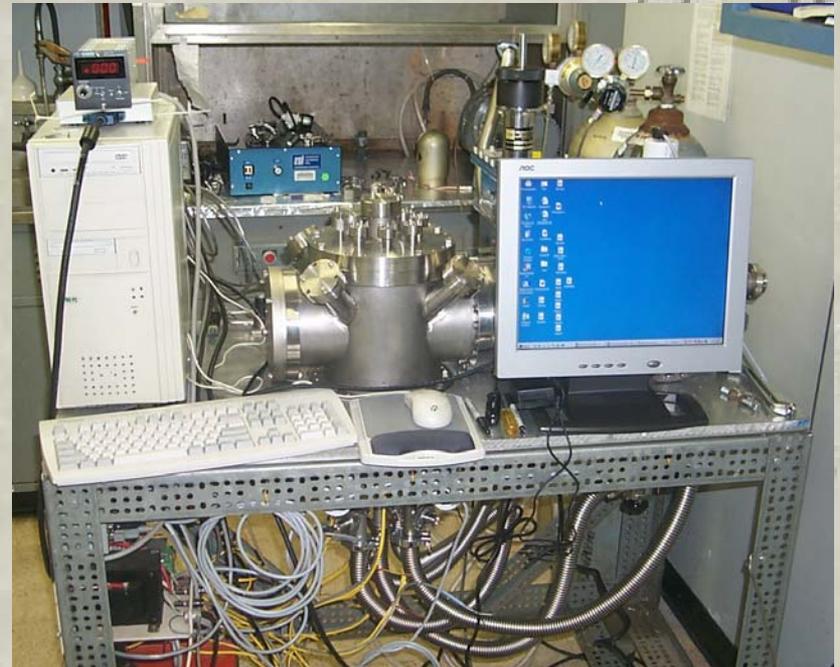
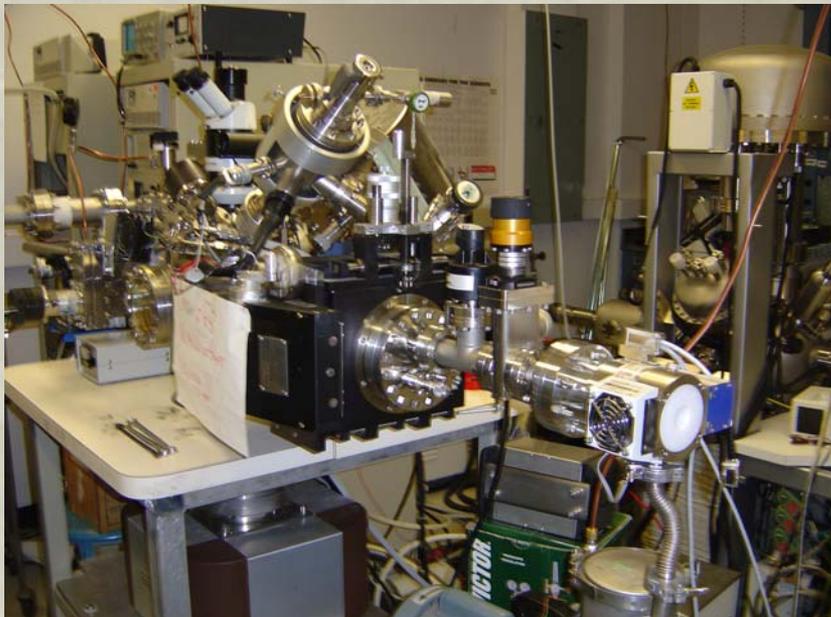


NRL Research Effort

- ❖ Collaborated with MHI to verify Pr present on their permeated samples
 - Initially with Accelerator Mass Spectrometry
 - Eventually with ICP-MS at part per quadrillion levels (10^{-15})
 - NRL convinced Pr was present on some samples

Modified XPS and 5-Sample Chambers @ NRL

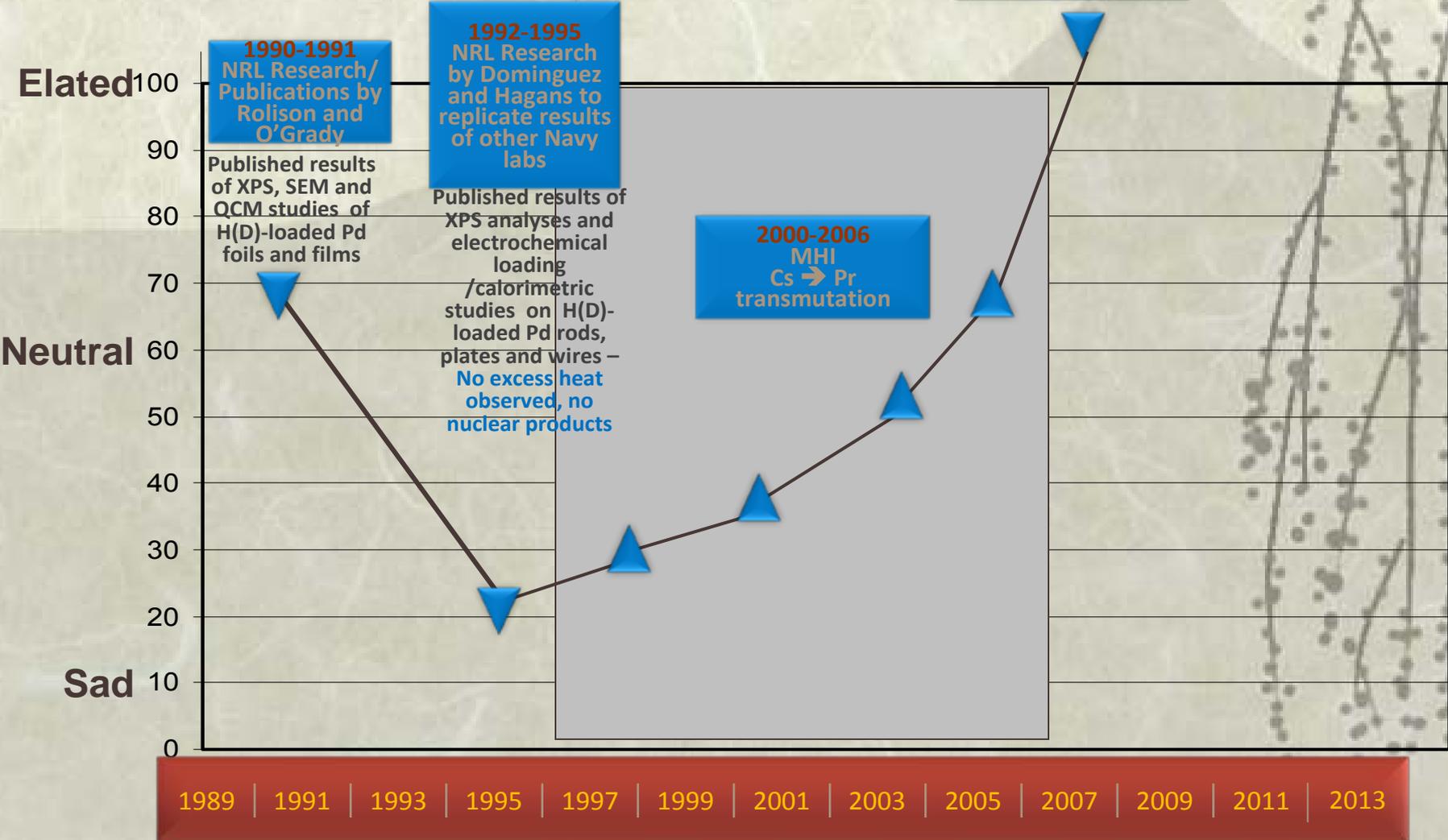
- ❖ NRL attempted to independently reproduce result
 - Unsuccessful at producing Pr
- ❖ Therefore, performed joint research with MHI



What Has NRL Observed?

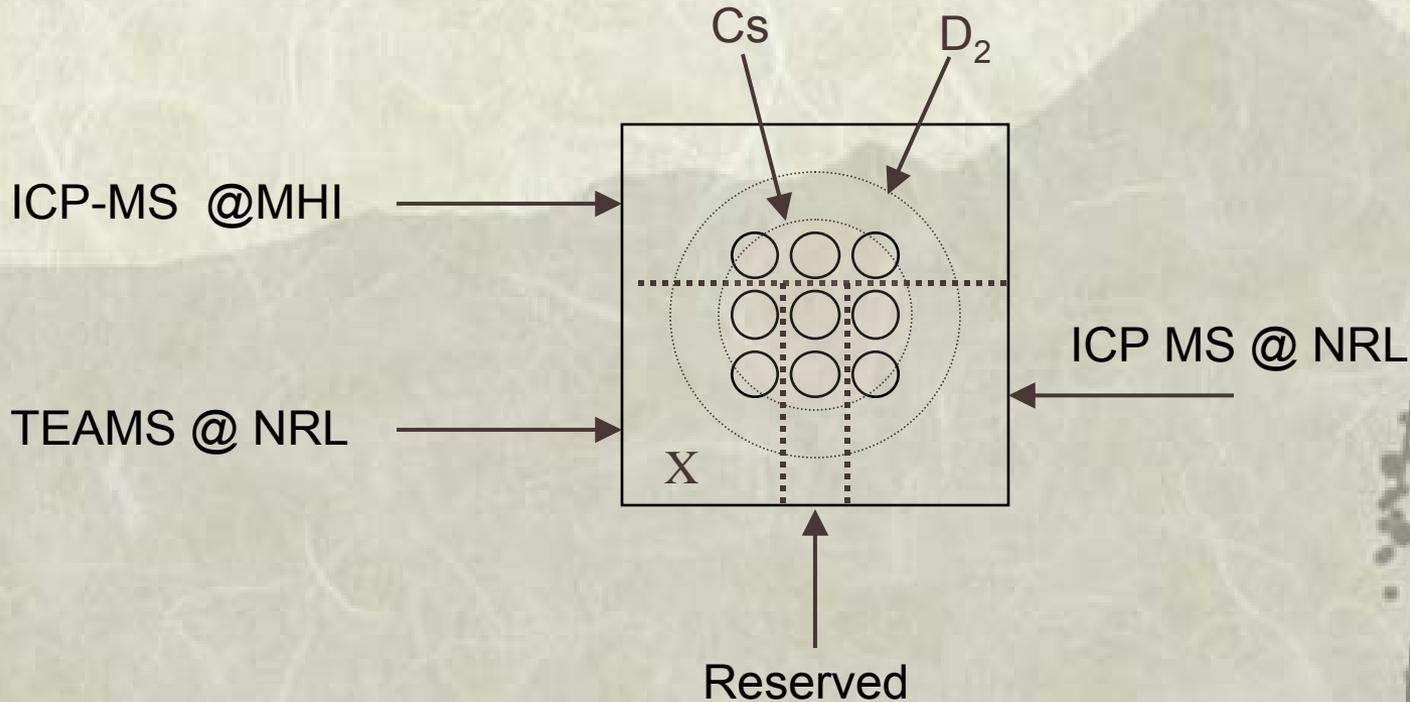
Chart of Happiness

Announcement/Publication
by Fleischmann and Pons



Foil - Analysis Layout

(typical)

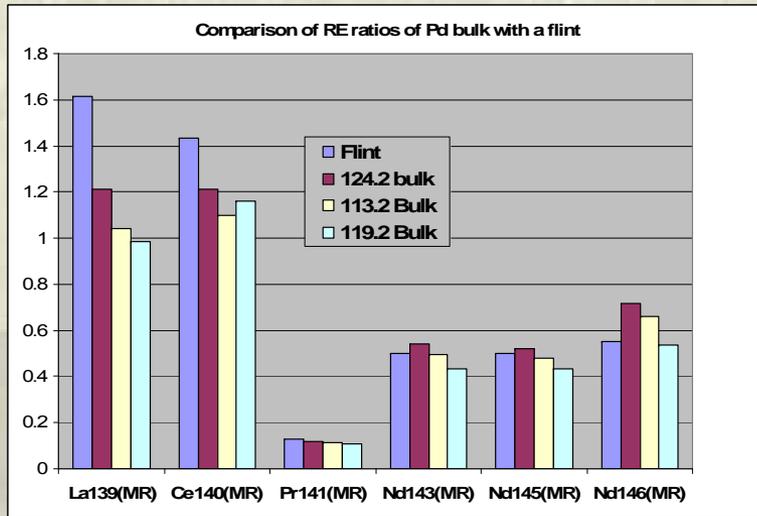


Note: Lower 2/3 Sent to NRL for Analysis

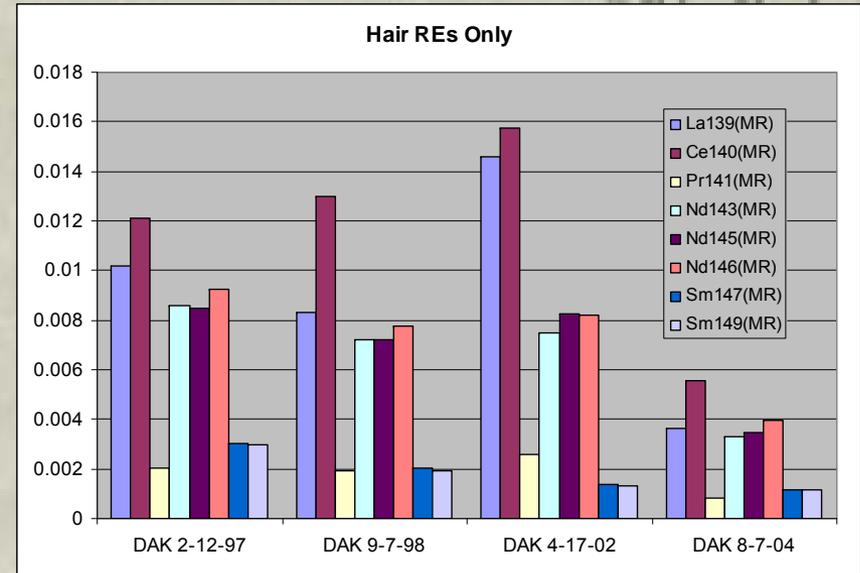
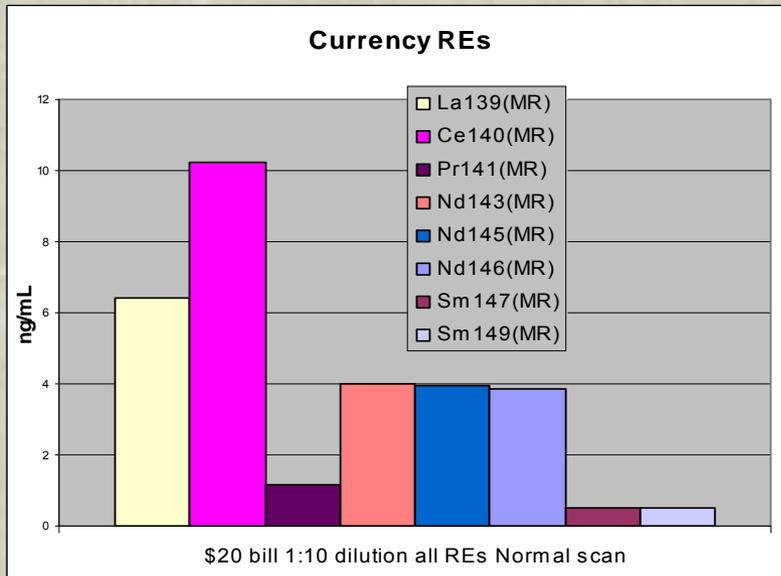
➡ Pr is mono isotopic so what would be the signature?

Rare Earths in the Environment

Rare earths are not so rare

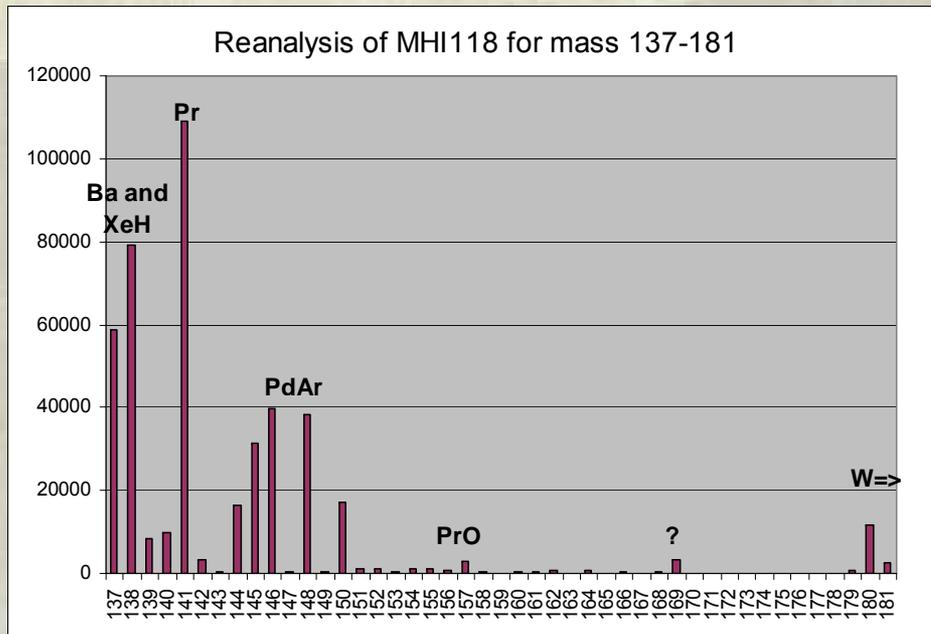


- ❖ Environmental sources of Pr
 - Flints – “Misch Metal”
 - Specialty glazes on ceramics
 - Phosphors in CRTs (tends to be with other REs)



Results - Molecular Interferences or Misch Metal Contamination?

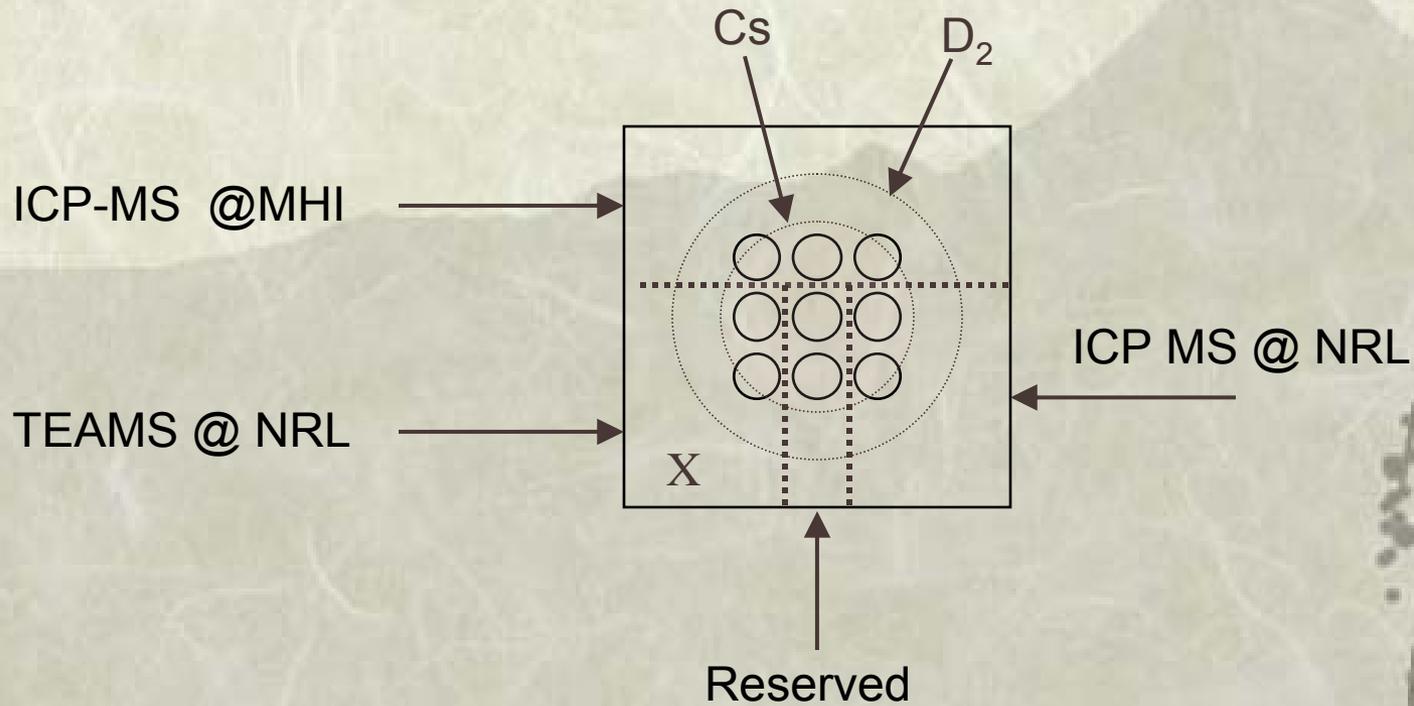
- ❖ 15 extracts & 6 blanks were sent from MHI
- ❖ 18 Confirmed negative for Pr – LOD <0.045 ng/cm
 - Includes 6 controls with the three positives (three sets of three)



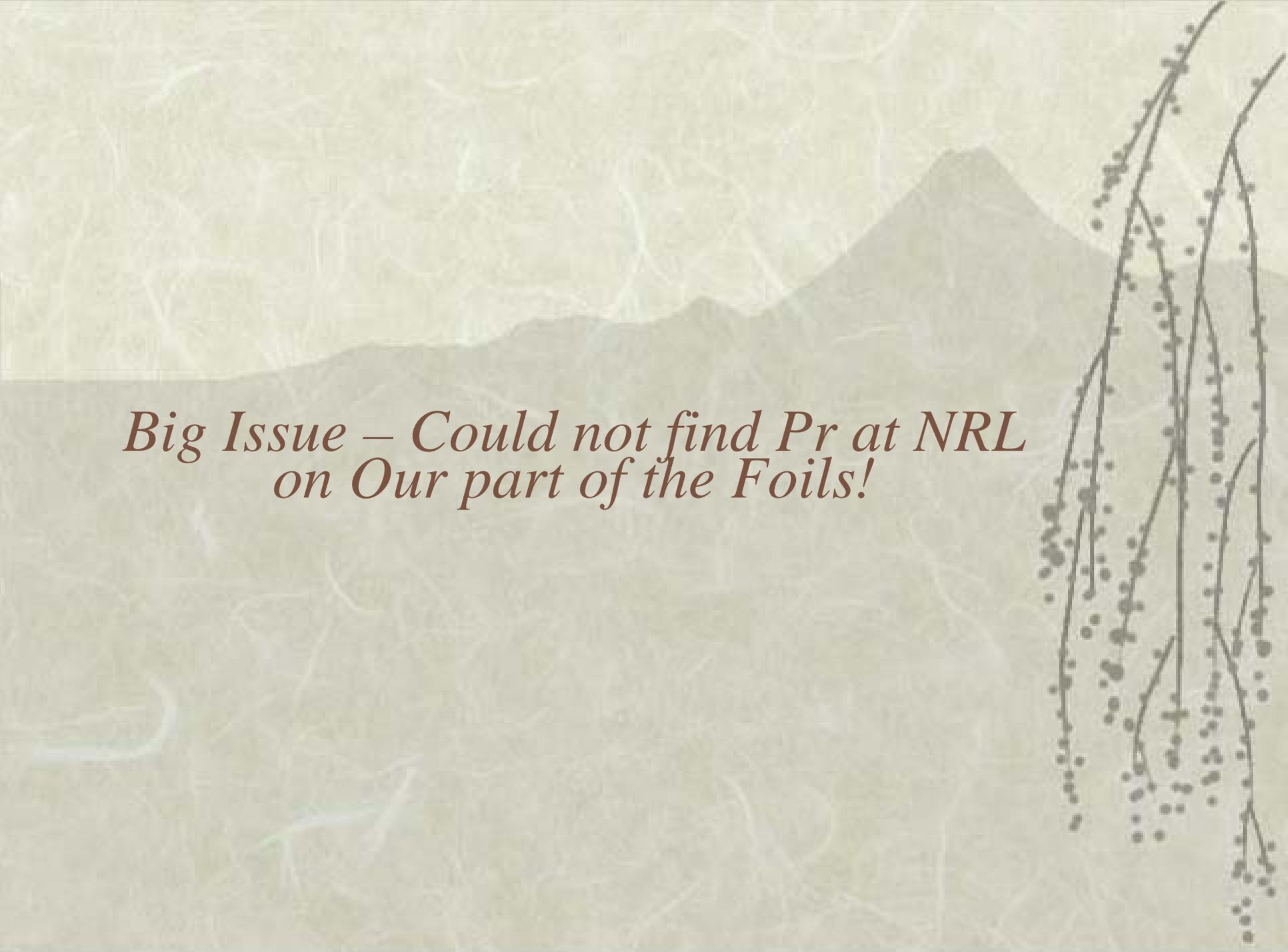
Sample	Toray ng/cm ²	NRL ng/cm ²	% Differen ce
MHI118	20	19	5.00%
MHI131	9.1	9.2	-1.50%
MHI143	25	24	5.54%

Foil - Analysis Layout

(typical)



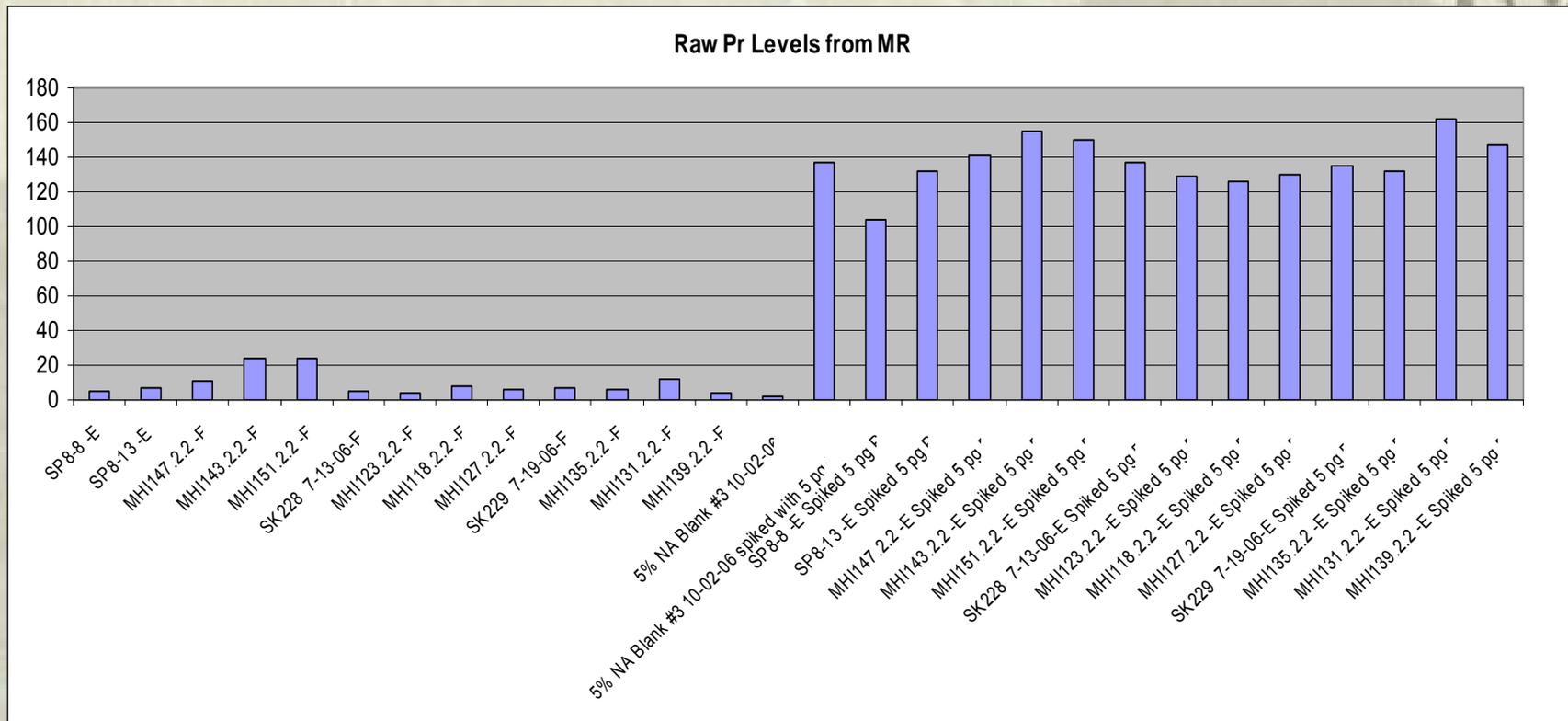
Note: Lower 2/3 Sent to NRL for Analysis

The background features a stylized landscape with a range of mountains in shades of green and brown. On the right side, there is a detailed illustration of a willow tree with long, drooping branches and small, dark leaves. The overall aesthetic is soft and artistic.

*Big Issue – Could not find Pr at NRL
on Our part of the Foils!*

Example Showing That NRL Could Have Observed Pr, If Present

- ❖ One series spiked with equivalent of 170 pg/cm² Pr (~100x lower than expected)



Possible Explanations for failure to find Pr

- ❖ Pr not there – we clearly have the required sensitivity and specificity
 - Mistakes at MHI (blanks mitigate this)
- ❖ Wrong part of foil
 - Systematic issues – statistically improbable
- ❖ Pr migrates into the interior – bulk analysis should find it!
 - Have done bulk analysis on MHI118 and essentially negative
- ❖ Pd catalyzes reverse nuclear reaction with time?
 - Real stretch of science
- ❖ Pr lost in transit or handling
 - Film unstable – Pr should be in boxes – checked and no
- ❖ Not etching enough Pd foil
 - Loss of deuterium with time causes lower Pd etching
 - Travel to MHI to extract a freshly prepared sample

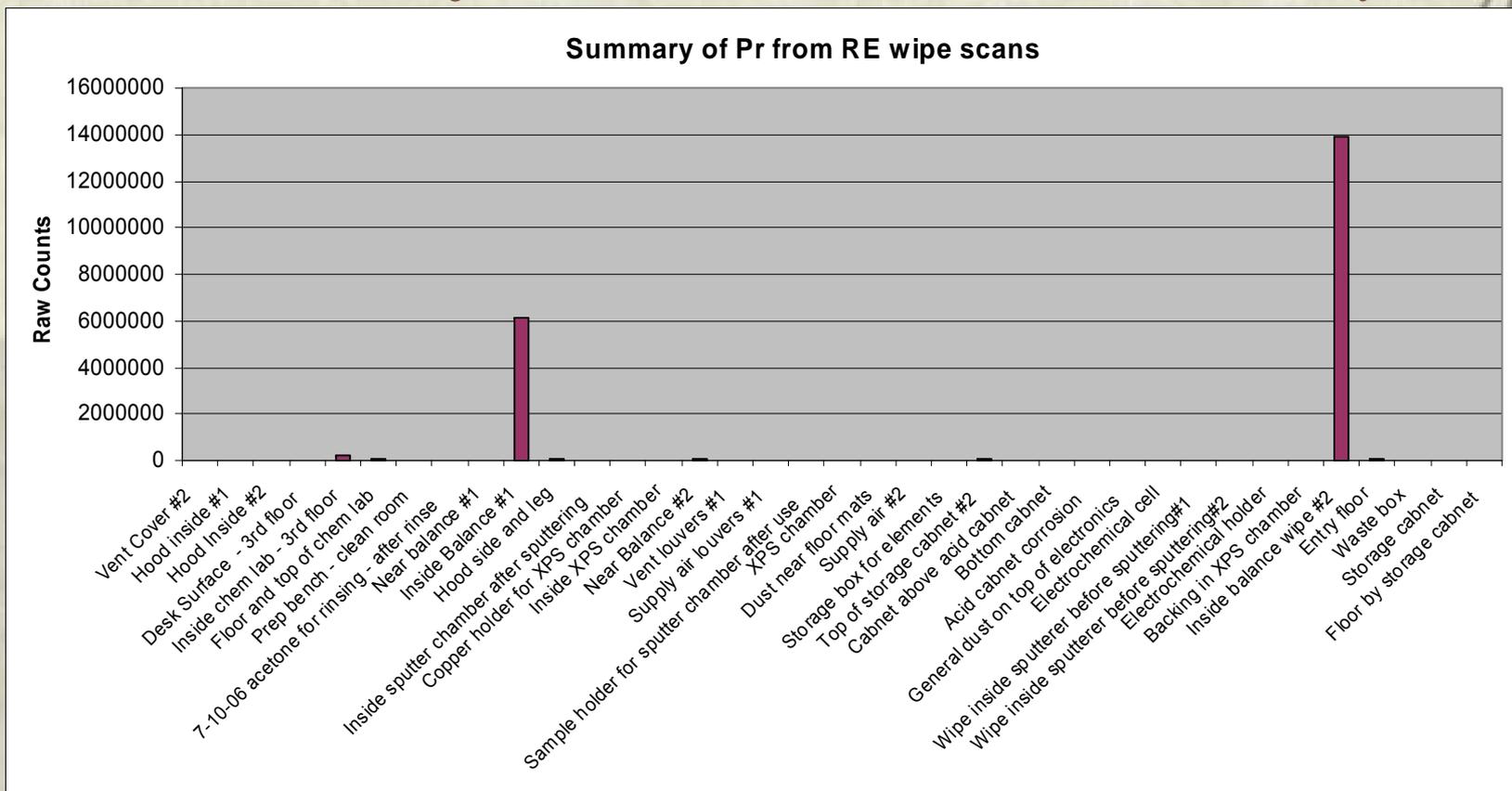
Months worth of work (frustration) later...

- ❖ Traveled to MHI to observe process and extract a freshly prepared sample
- ❖ Participated in extraction using their equipment on a fresh sample
- ❖ Take split sample extracts

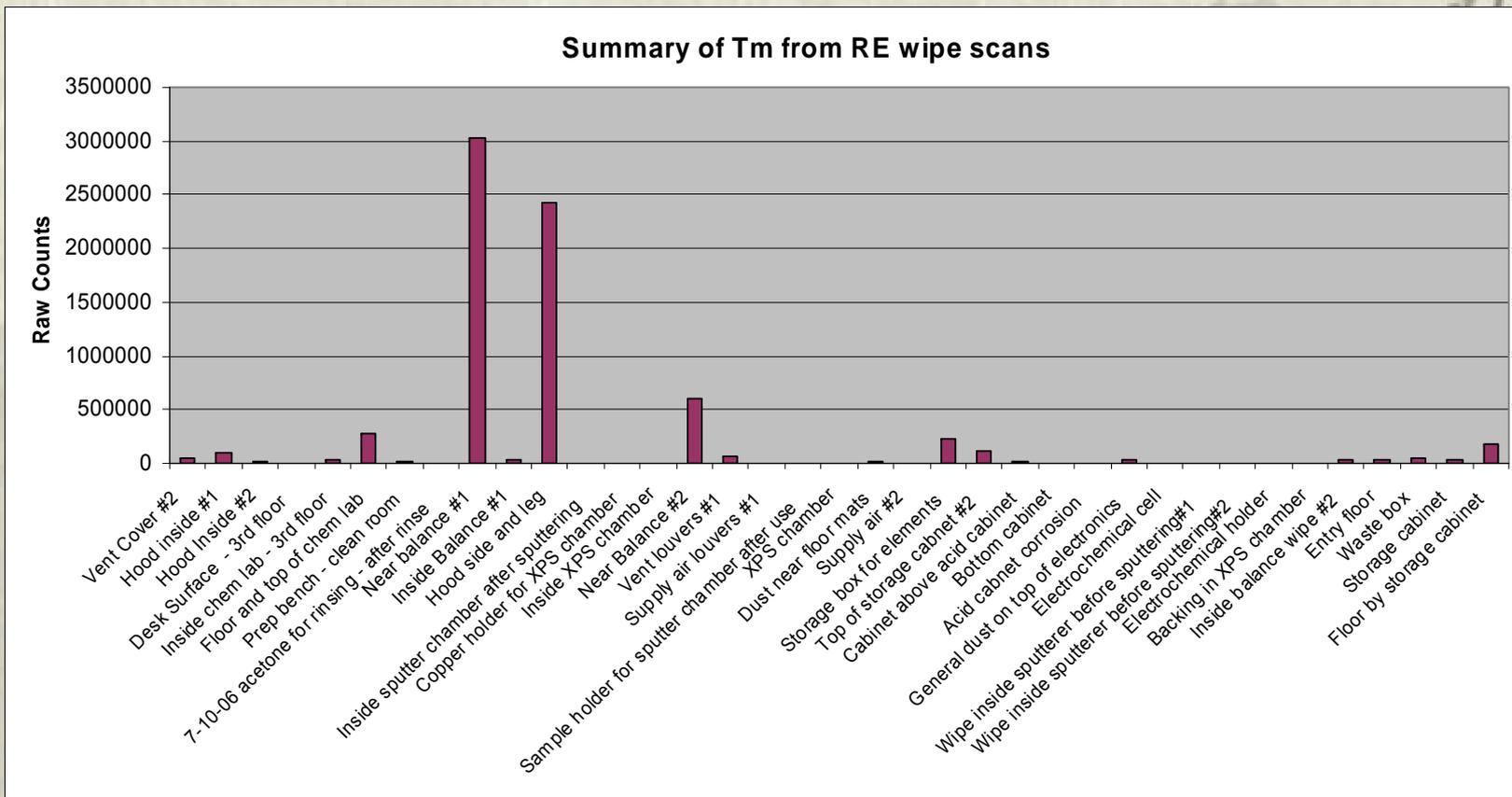
- ❖ Samples extracted at MHI in the presence of NRL personnel were negative at NRL and with a commercial laboratory

On whim, do environmental survey

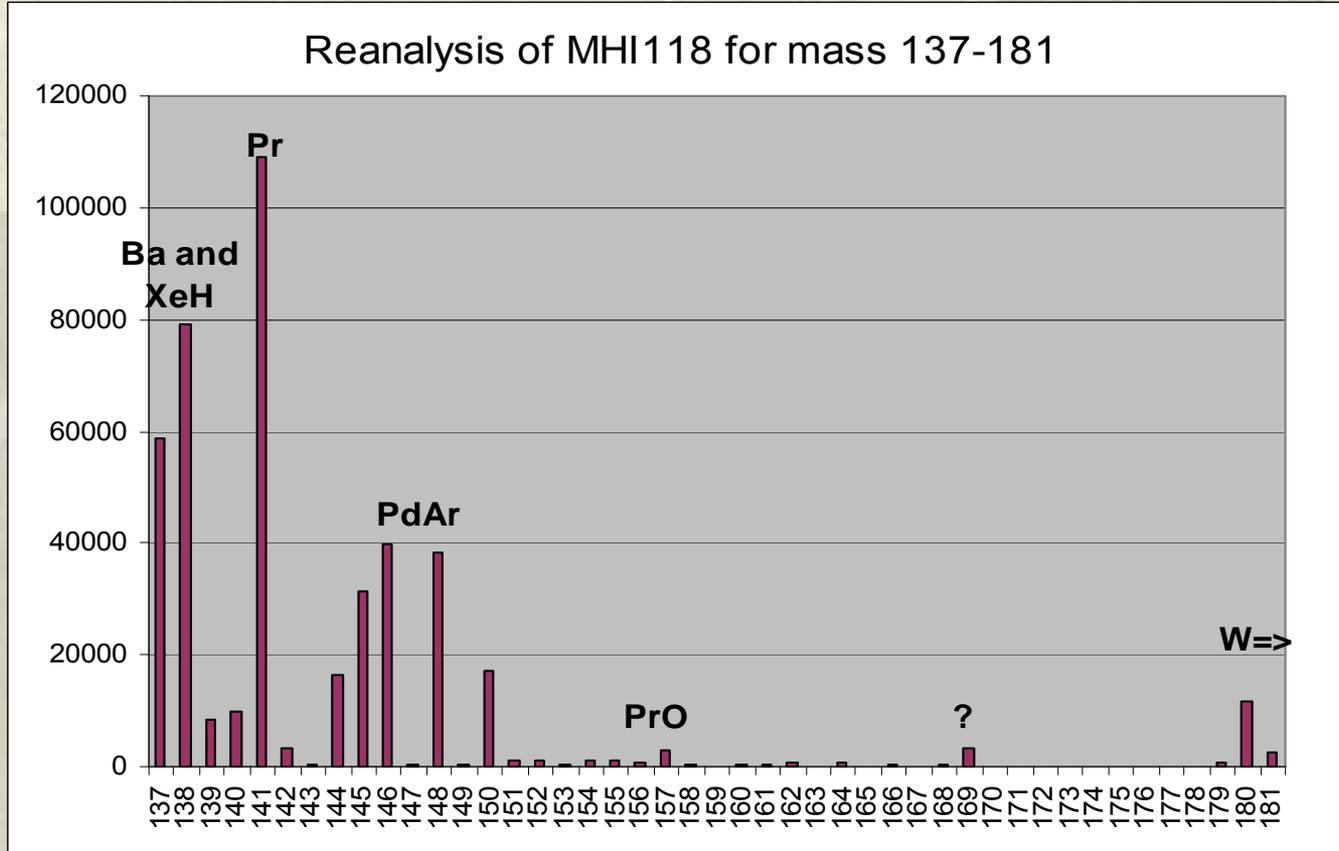
Results from Environmental Survey



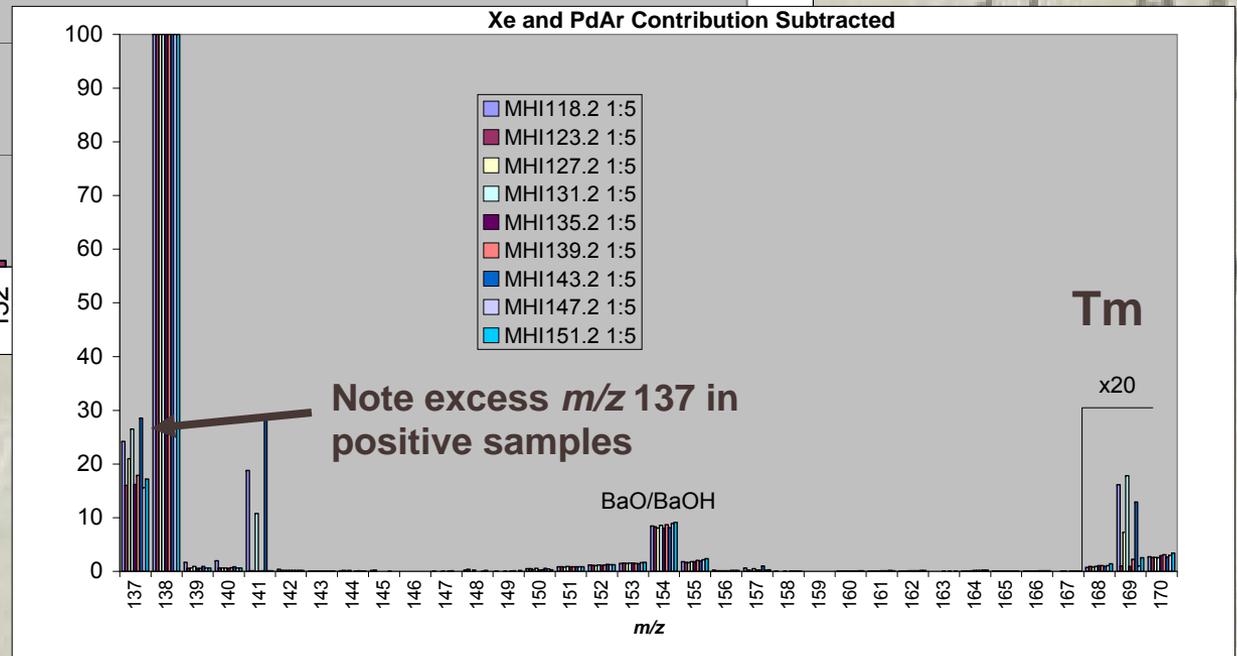
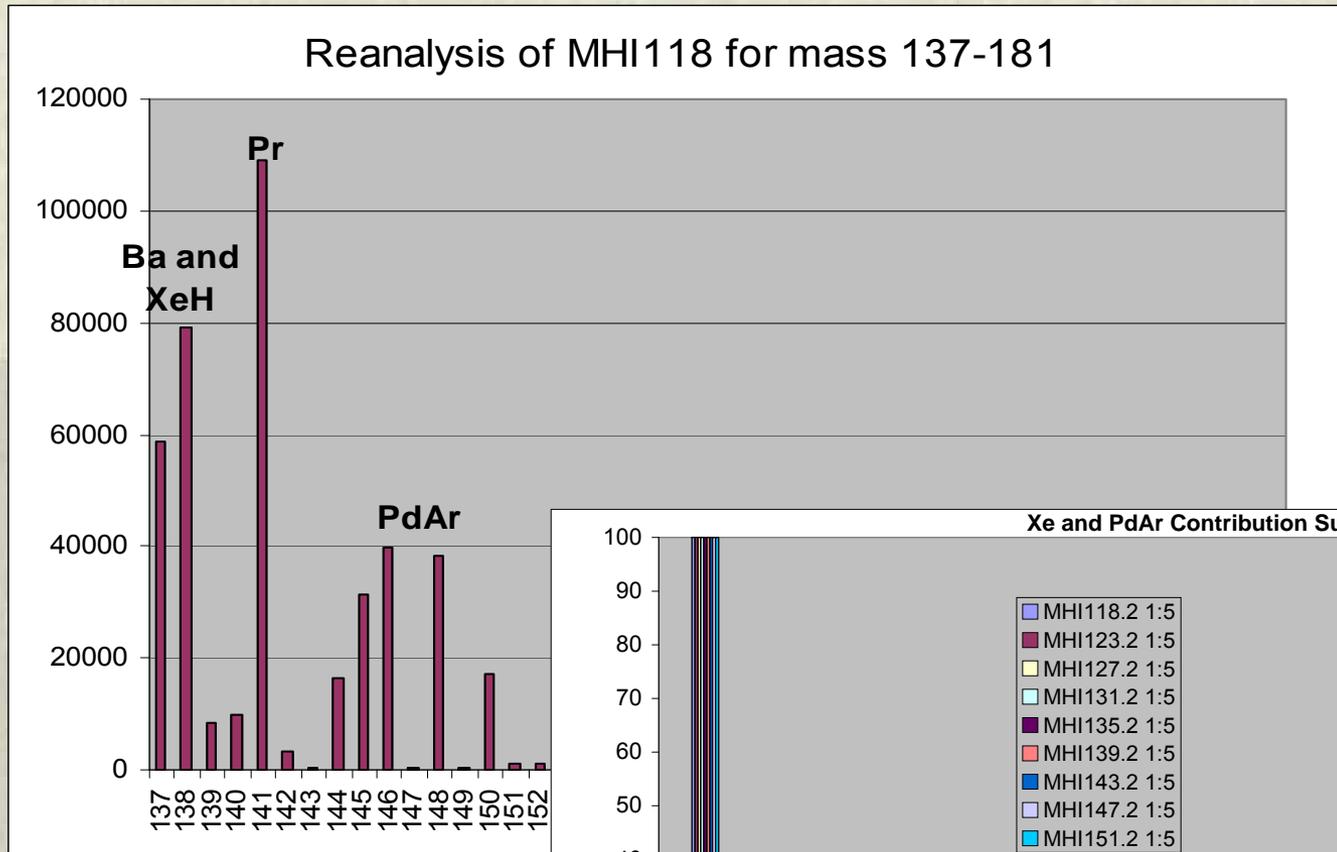
Results from Environmental Survey



Excess ^{137}Ba and Thulium Found in Samples Origin??



Excess ^{137}Ba and Thulium Found in Samples Origin??



Conclusions **for Transmutation**

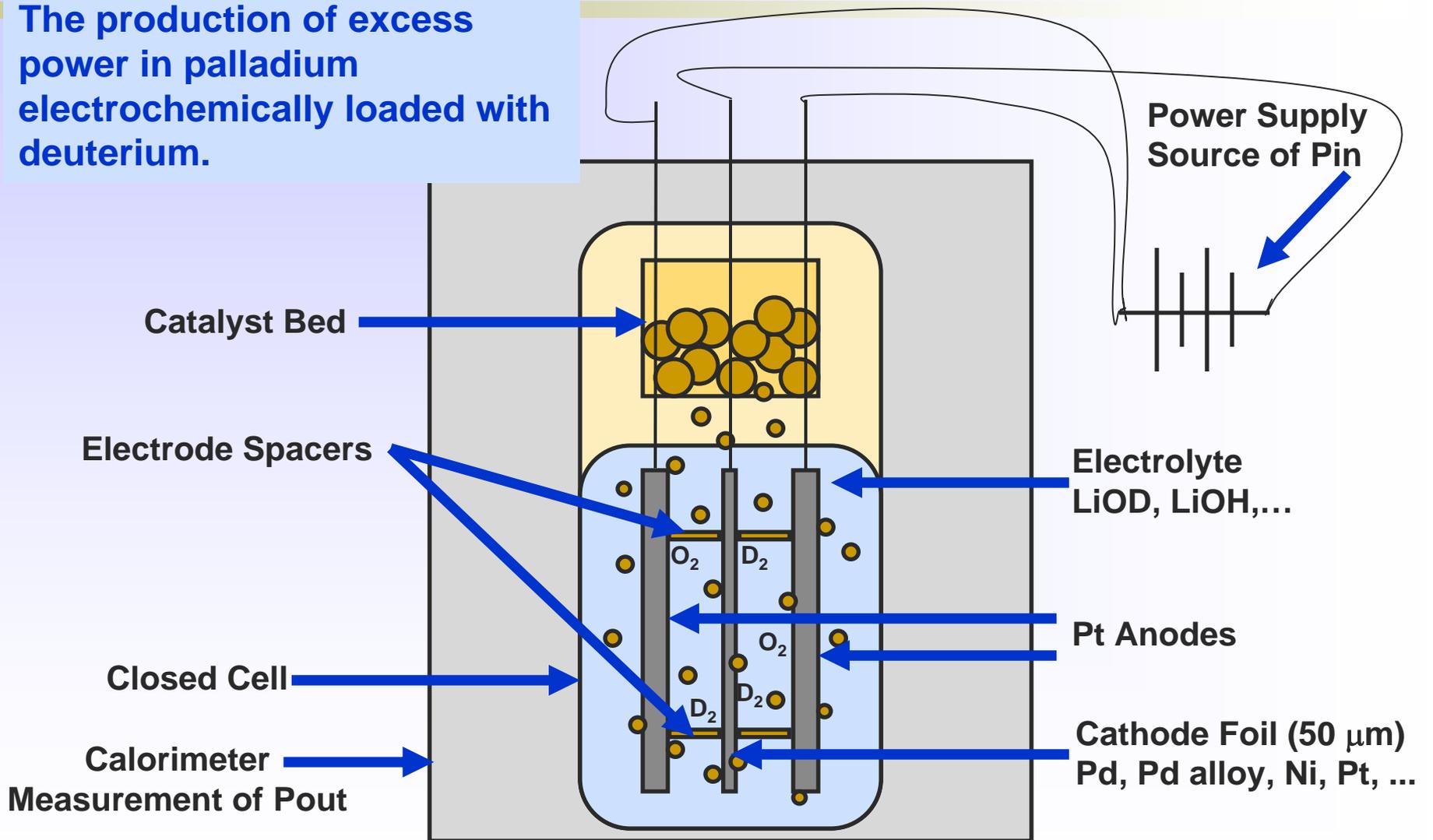
- ❖ Molecular interferences will confuse isotopic patterns
 - SIMS be wary
 - ICP-MS better but not perfect
- ❖ Apply the Prime Directive
- ❖ Evidence for transmutation of Cs to Pr in THIS data set is weak

- ❖ For NRL, Chart of Happiness for Transmutation is:

Off the chart!

Electrochemical Experiments

The production of excess power in palladium electrochemically loaded with deuterium.



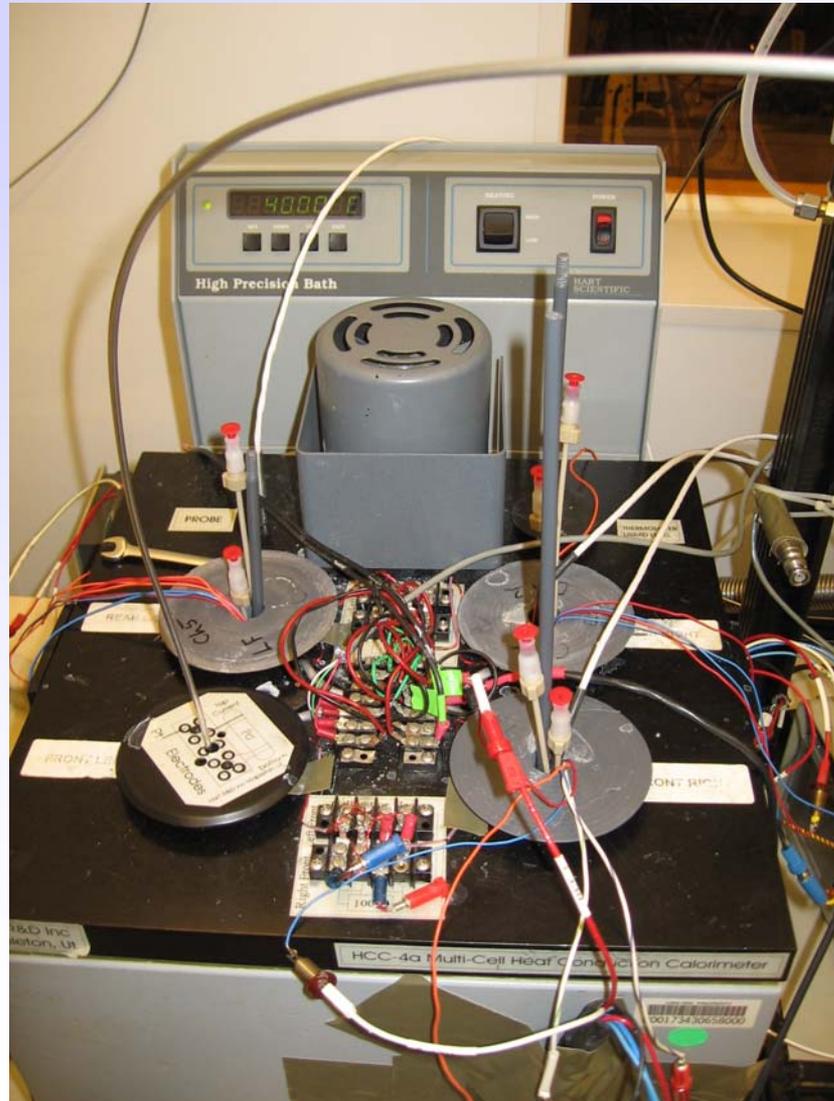
Electrode sandwich configuration based on ENEA / Violante design



Cell Set-up



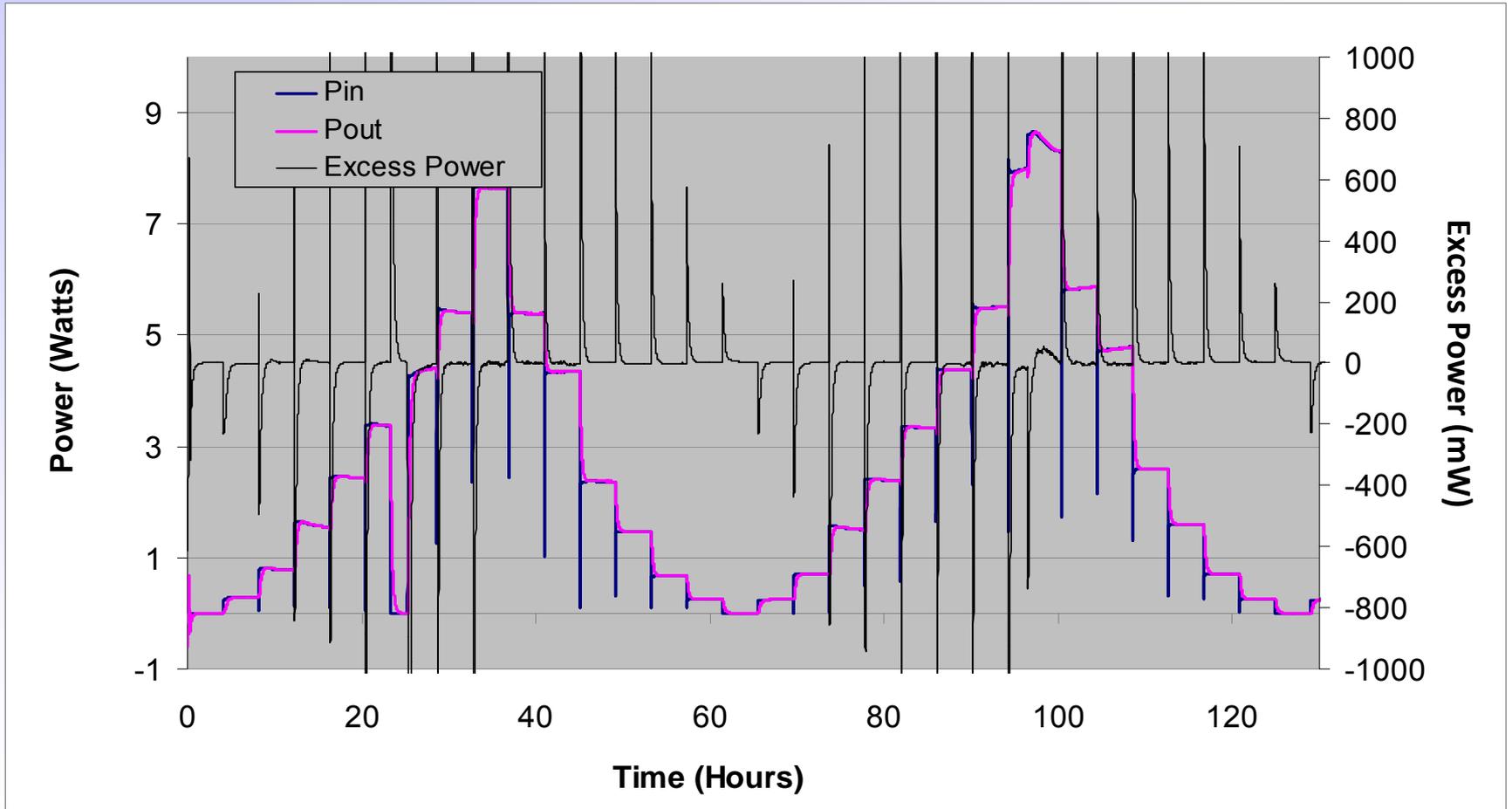
Hart Set-up





You recover the initial energy to heat cell

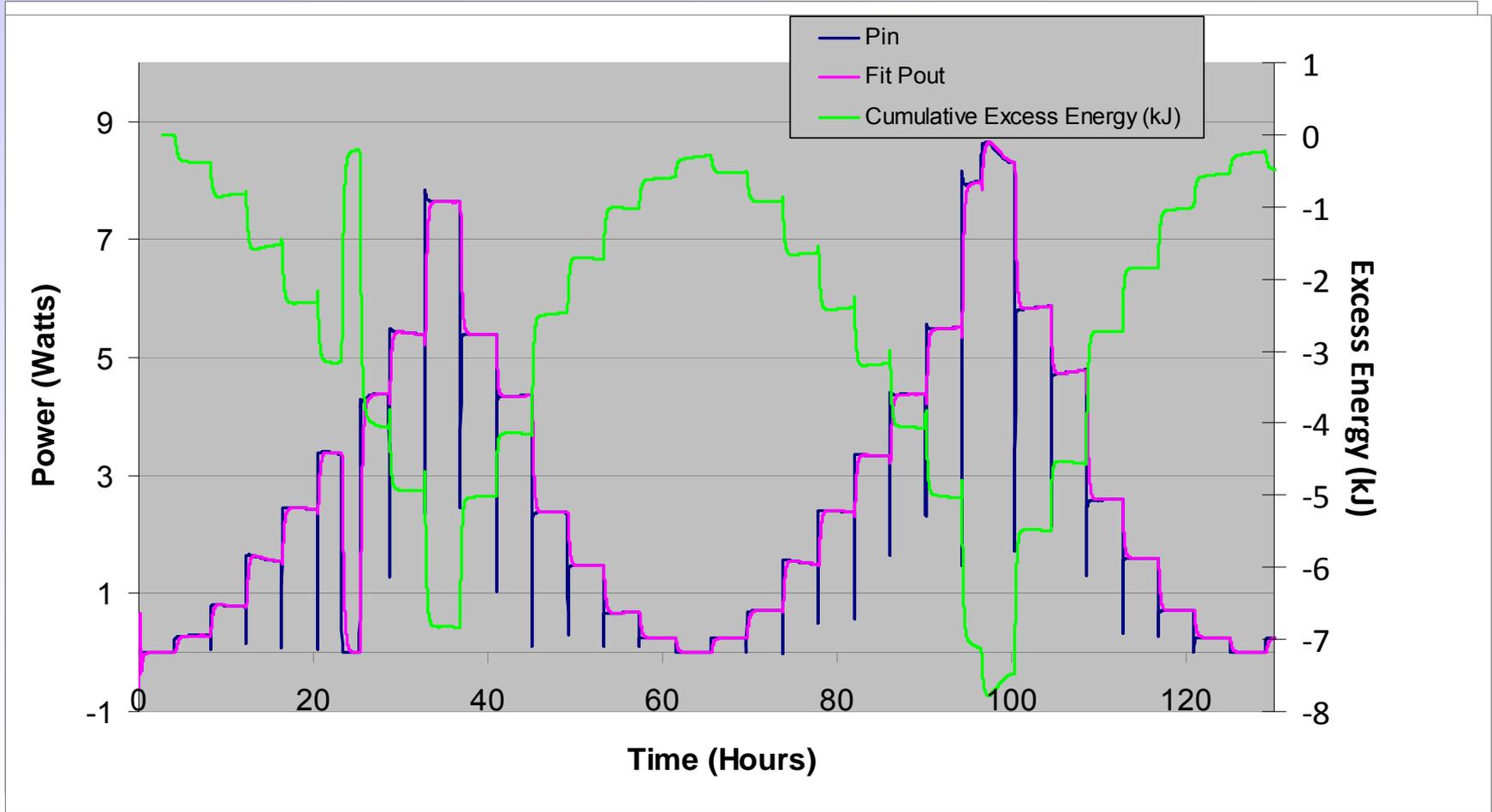
Cathode #OL2 1-29-13 using LiOD and cycled





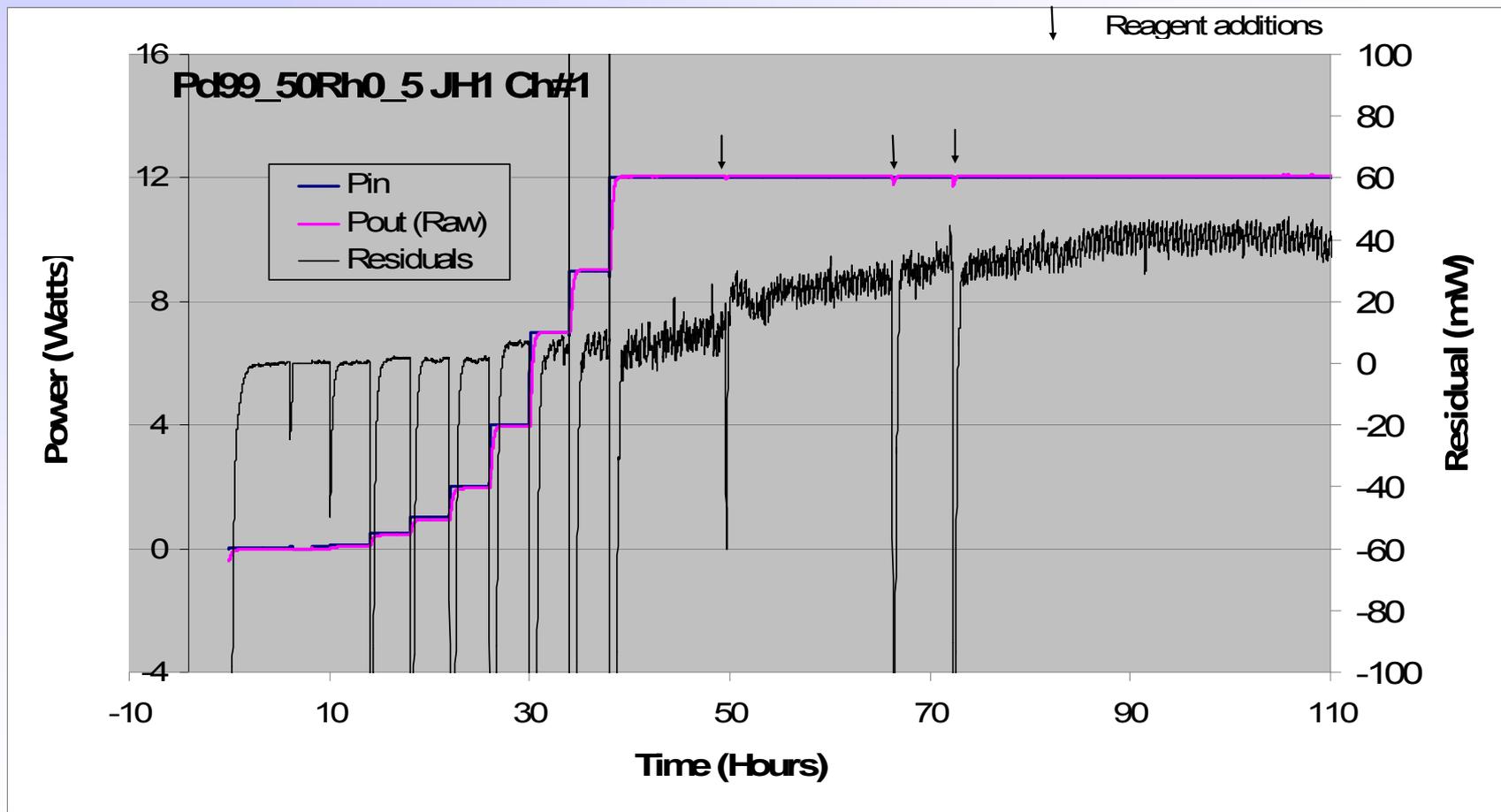
You recover the initial energy to heat cell

Cathode #OL2 1-29-13 using LiOD and cycled



Unbalance after Additions

Adding Raney Nickel often results in ~50 mW extra power

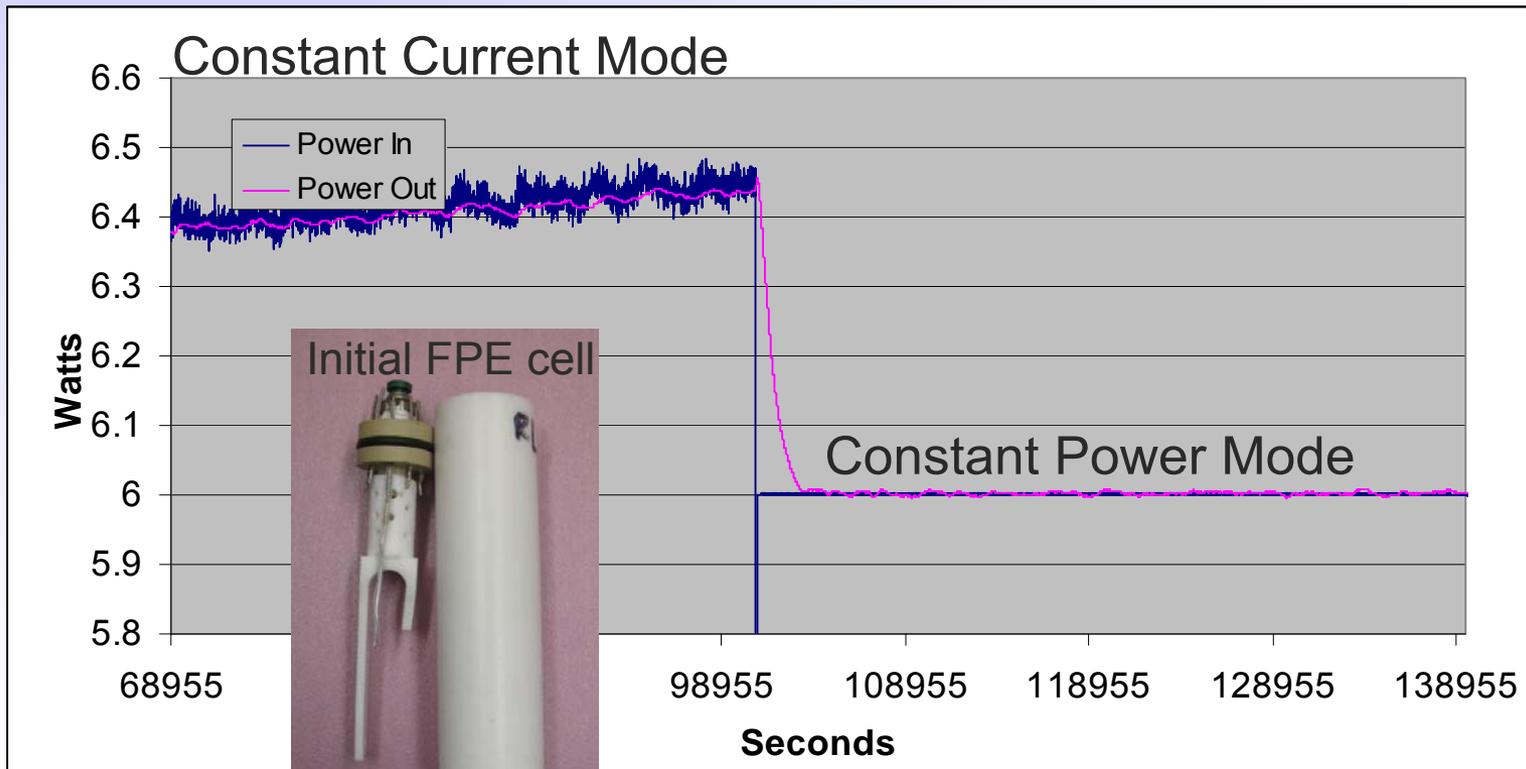


Electrolytic Loading Experiment

Original Fleischmann and Pons Approach



- Many experiments, over 24 months, with consistent results
 - $\text{Power}_{\text{in}} = \text{Power}_{\text{out}}$

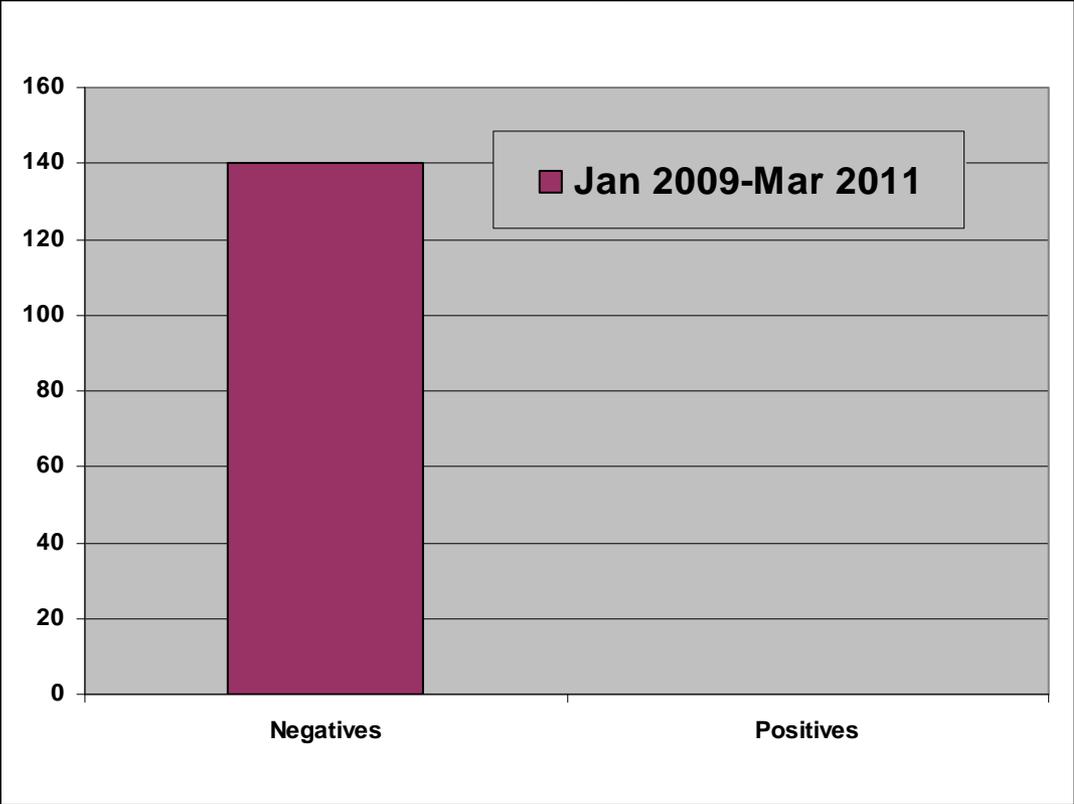


Teflon cell with Teflon internal parts and PEEK top (no capability for chemical additions)

Cathode Materials Investigated



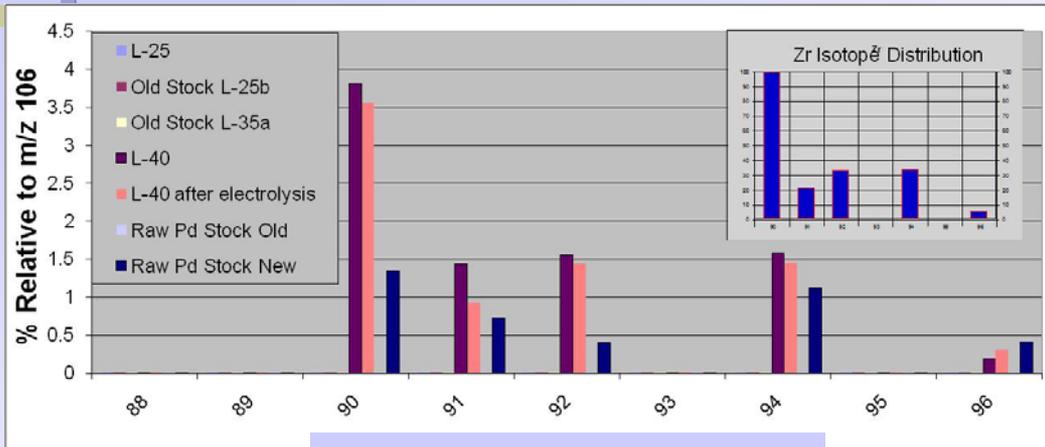
Cathode Material	Number	x/LiOD	Pd 90%/ Rh10%	
Pd/LiOD			1M LiOD, Pt anodes, VSP	61
Goodfellow	22		new calorimeter	18
Platexis	19		BOPs	13
Engelhard	23		G&S Pd	12
				10
				8
				3
				10
				3
				2
				2
Pd/LiOH				4
				2
				2
				2
				1
				1
				1
misc				1
				1
				1
				11
				4
				3
				4
				2
			Ni	7
			Ni/Pd	2
			Ni/Pd/Ni	1
			Nb	3
			Ta	2
			Total	198
			Grand Total	307



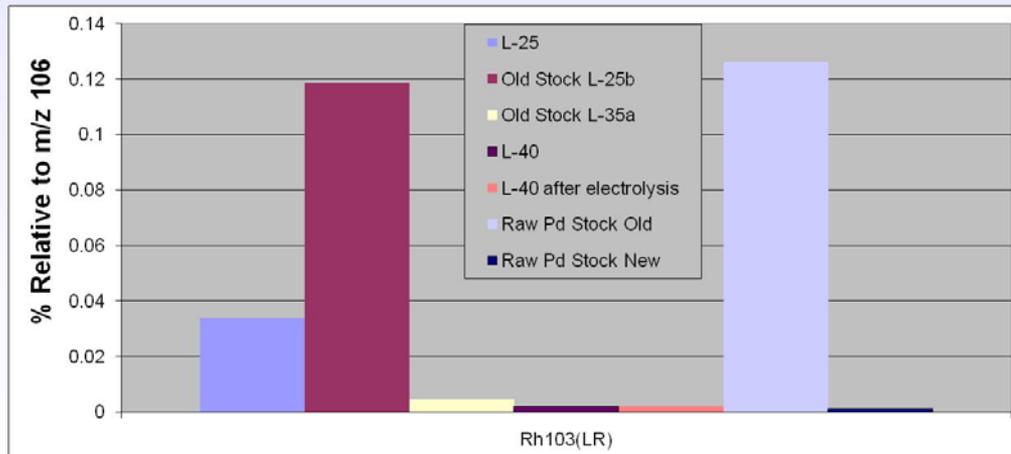
Why focus on Pd 90%/ Rh 10%?

Trace Impurities

Inductively Coupled Mass Spectrometric Analysis



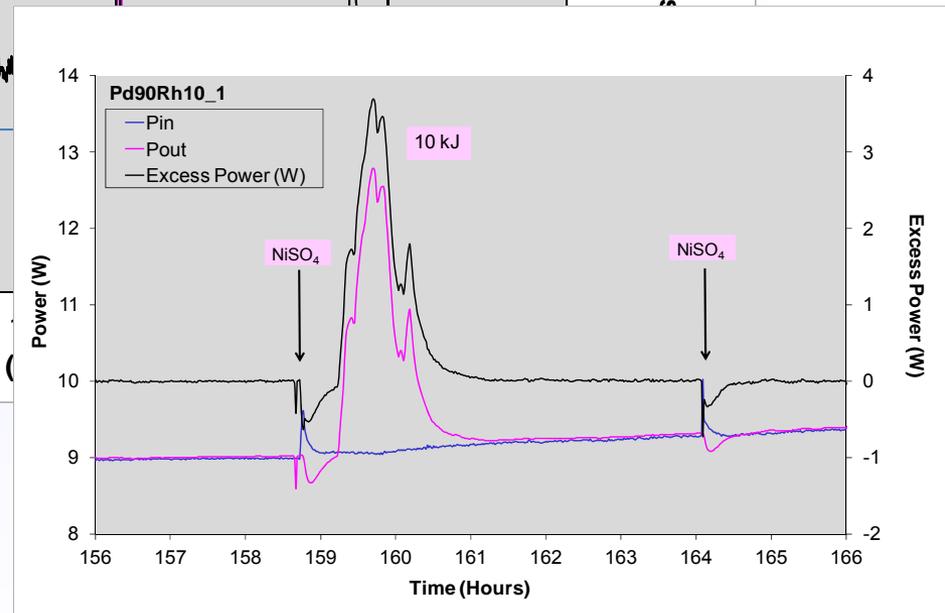
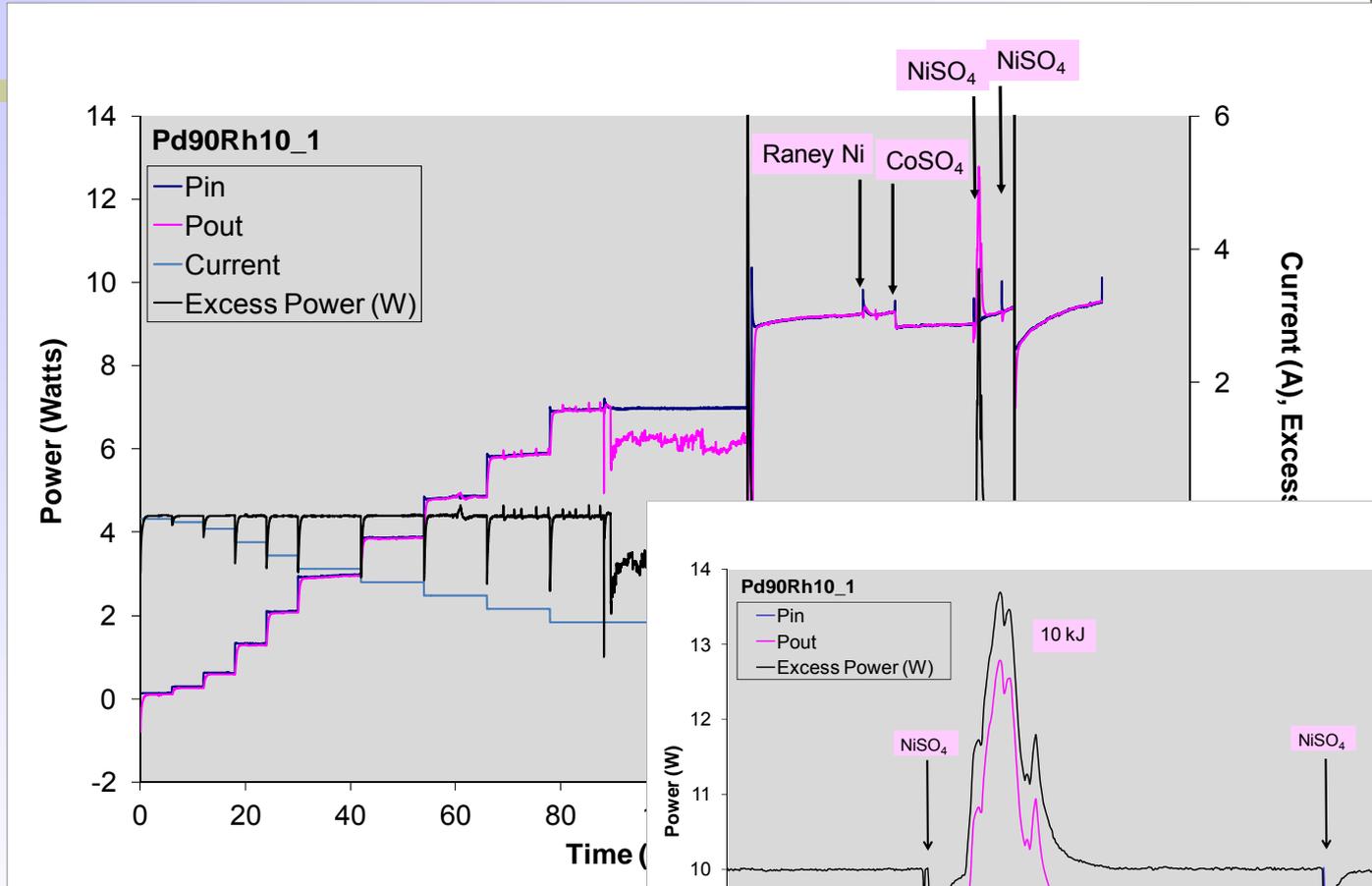
Detection of Zr/Y/Ha
Only in New lots



Detection of Rhodium
Only in Old lots

- Older lots of Palladium, that appeared to produce substantial heat, likely had only ONE source – Engelhard
- ICP-MS analysis shows different impurity profiles than current palladium lots
 - Older lots appear to have recycled Pd from catalytic converters
 - Current lots are much purer in these elements but have zirconium, yttrium, and hafnium present
 - Likely change in crucibles for melting to zirconia
 - Rhodium prices may drive recovery as a separate element
- Are the impurities responsible for the Fleischmann-Pons Effect?

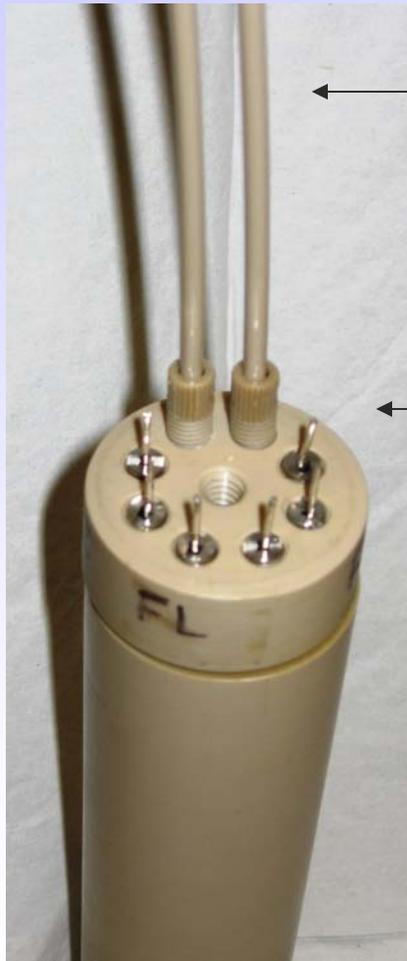
Cathode #1 - Eve



- 10 kJ over 90 minutes
- Event triggered by metal/metal oxide additions into 24 mL electrolyte volume??

- 5-10 mg NiSO₄ →
155 MJ/mole or 1.6 keV/molecule

All PEEK Cell Design

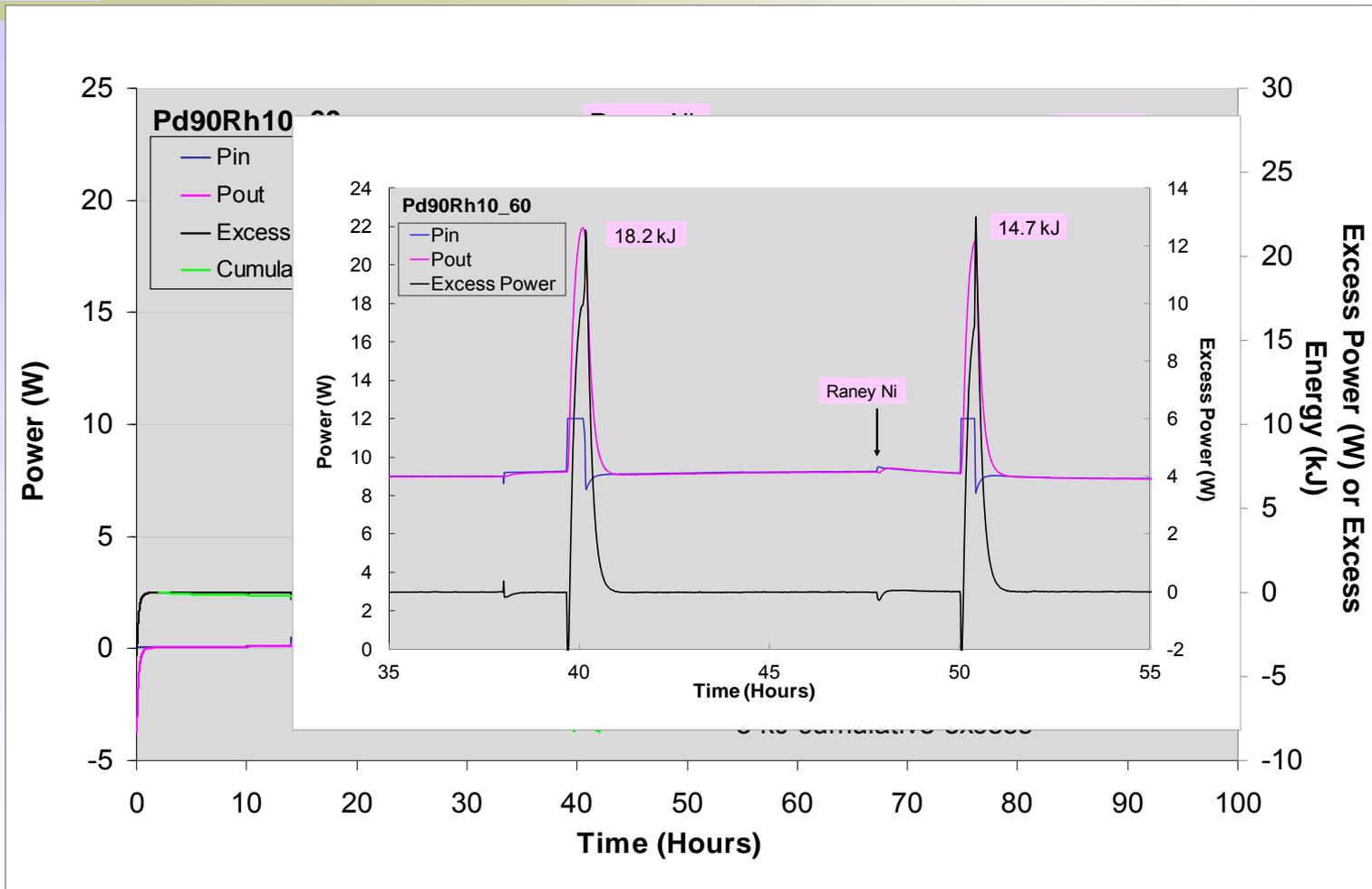


← Tubes for chemical additions and pressure measurements

← Cell top with Platinum feed throughs and High Pressure Liquid Chromatography fittings for sealing



Cathode #60



- Excess energy \gg stored chemical energy
- Excess energy appeared before first addition
 - May have been due to cleanliness of cell

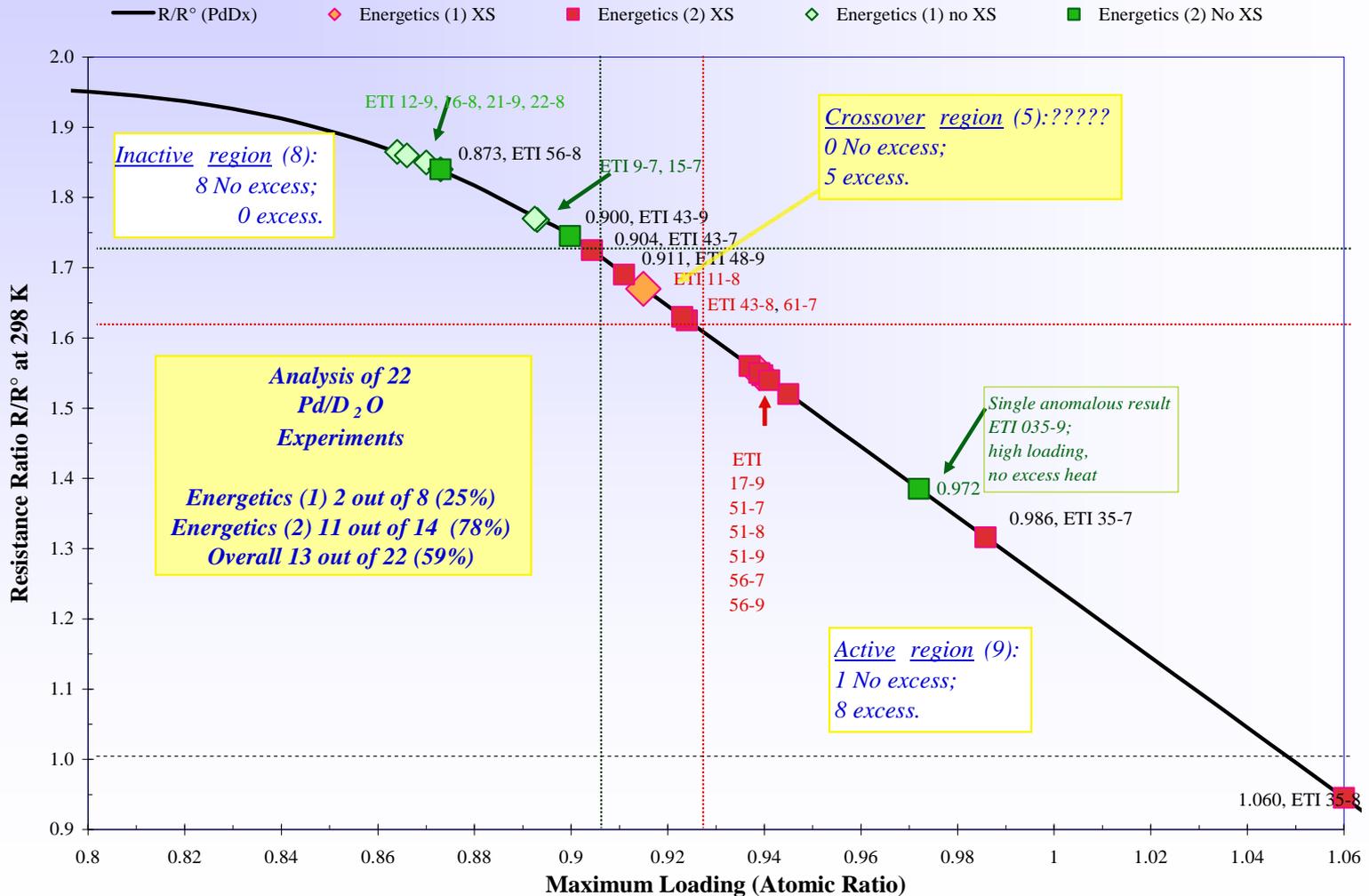
Production of large excess energy in a FPE is Rare!



- Ed Storms
 - 5/90
- SRI
 - 4/26
- NRL
 - 4/61
- Energetics
 - *ca.* 8/800
- F&P
 - 1/?? described

SRI Results

Michael C.H. McKubre and Francis L. Tanzella, "New Physical Effects In Metal Deuterides"
 Contract No. HR0011-05-C-0089

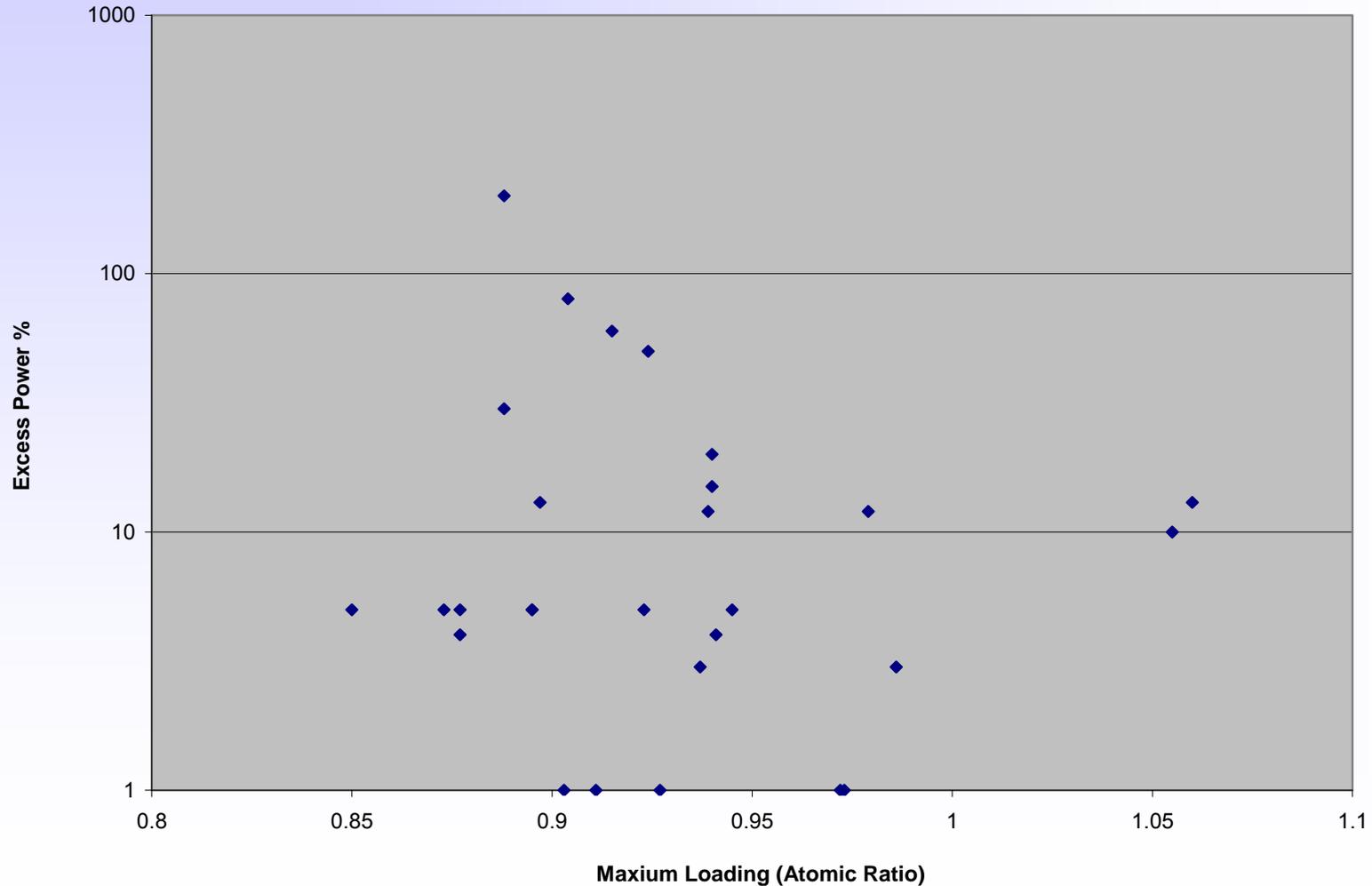


SRI Results

No Apparent Relationship to D Loading



DARPA Phase I Data Only
No Apparent Relationship to D Loading

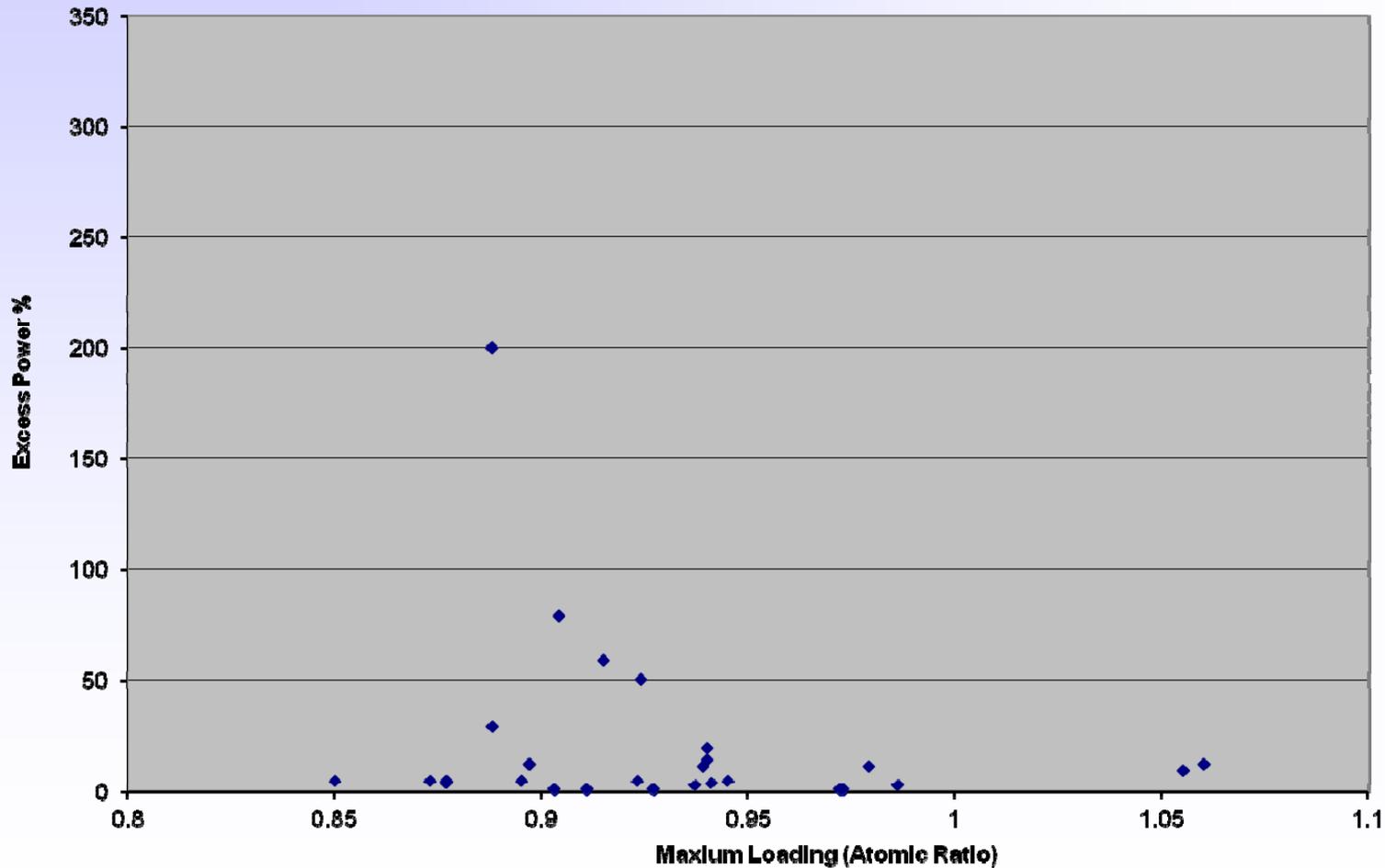


SRI Results

No Apparent Relationship to D Loading



DARPA Phase I Data Only



Instrumental Artifact?

Shorts



- Shorted in a certain way, RF generated
 - Right frequency and similar pattern
 - Very irreproducible – not all shorts show RF
 - Need just the right cell resistance and mode in the VSP

- Shorted in a certain way, the VSP will:
 - Misreport the power into the cell
 - Power nearly sufficient to make observed effects
 - Show no error messages

Conclusions

for heat or excess energy



- NRL's electrochemical shows excess energy that is:
 - Not chemistry
 - But possibly an instrumental artifact
- For NRL, Chart of Happiness for heat is:

At 25%!

Gas Loading

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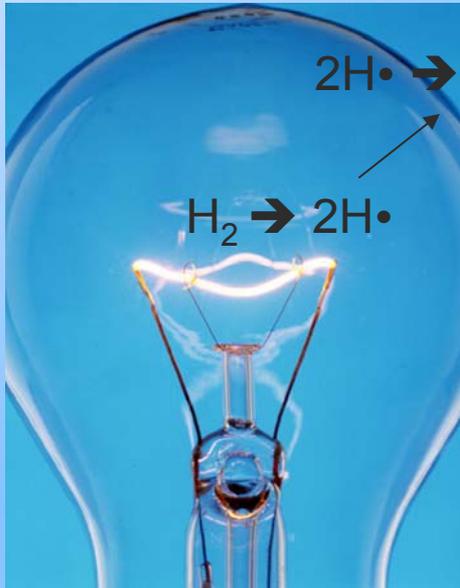
David.Kidwell@nrl.navy.mil



Langmuir



- Worked for GE – wanted better light bulb



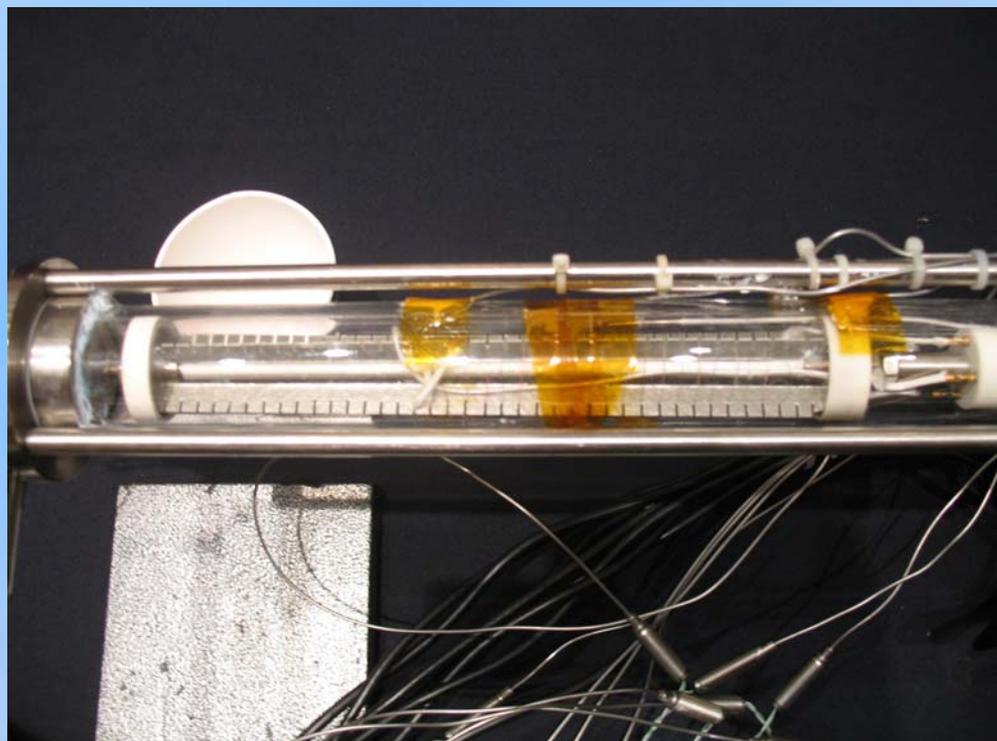
- Tested various gases
- Observed extra energy coming from hydrogen
- *i.e.* surface of bulb much **hotter** with H_2 than any other gas

Eventual explanation:

- Appearance of extra energy – but not really
 - H_2 splits into $\text{H}\cdot$ radicals on hot filament
 - Radical diffuse to cooler surface – cannot recombine in gas phase
 - Radicals recombine at glass surface
 - Transfer lots of energy



Is this happening here?



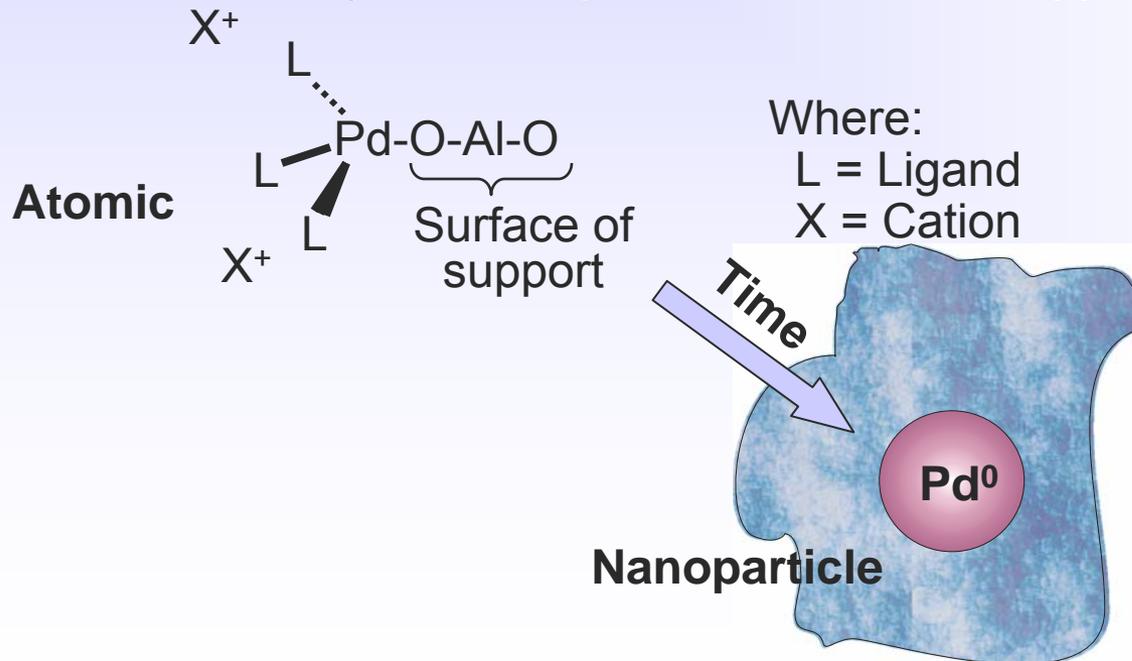
- Not a hot filament
- Is the “catalyst” just a better at making hydrogen radicals?
 - Ni and Pd known to form hydrogen radicals
- Accurate calorimetry will determine

I. Chorkendorff, J.N. Russell, Jr., and J.T. Yates, Jr., “Hydrogen Implantation in Ni(111) - A Study of H₂ Desorption Dynamics from the Bulk”, *Surface Science*, **182** (1987) 375-389.



Concept – Grow Particles *in situ*

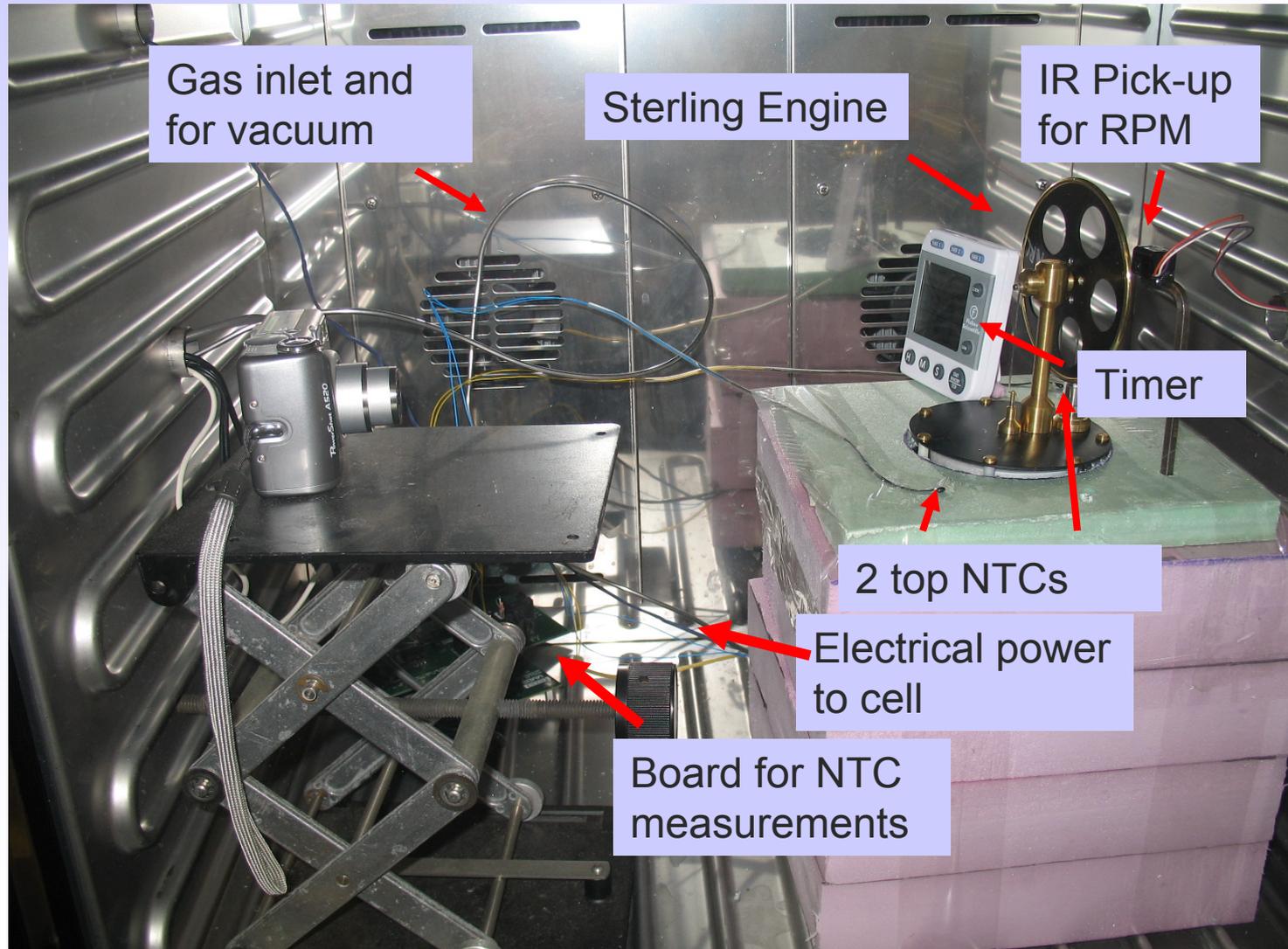
- Use chemistry to modify particle growth
 - Dispersed ions → Nanoparticles
- Requires:
 - Oxide support
 - Moderately chelated palladium bound to support





Useful Work?

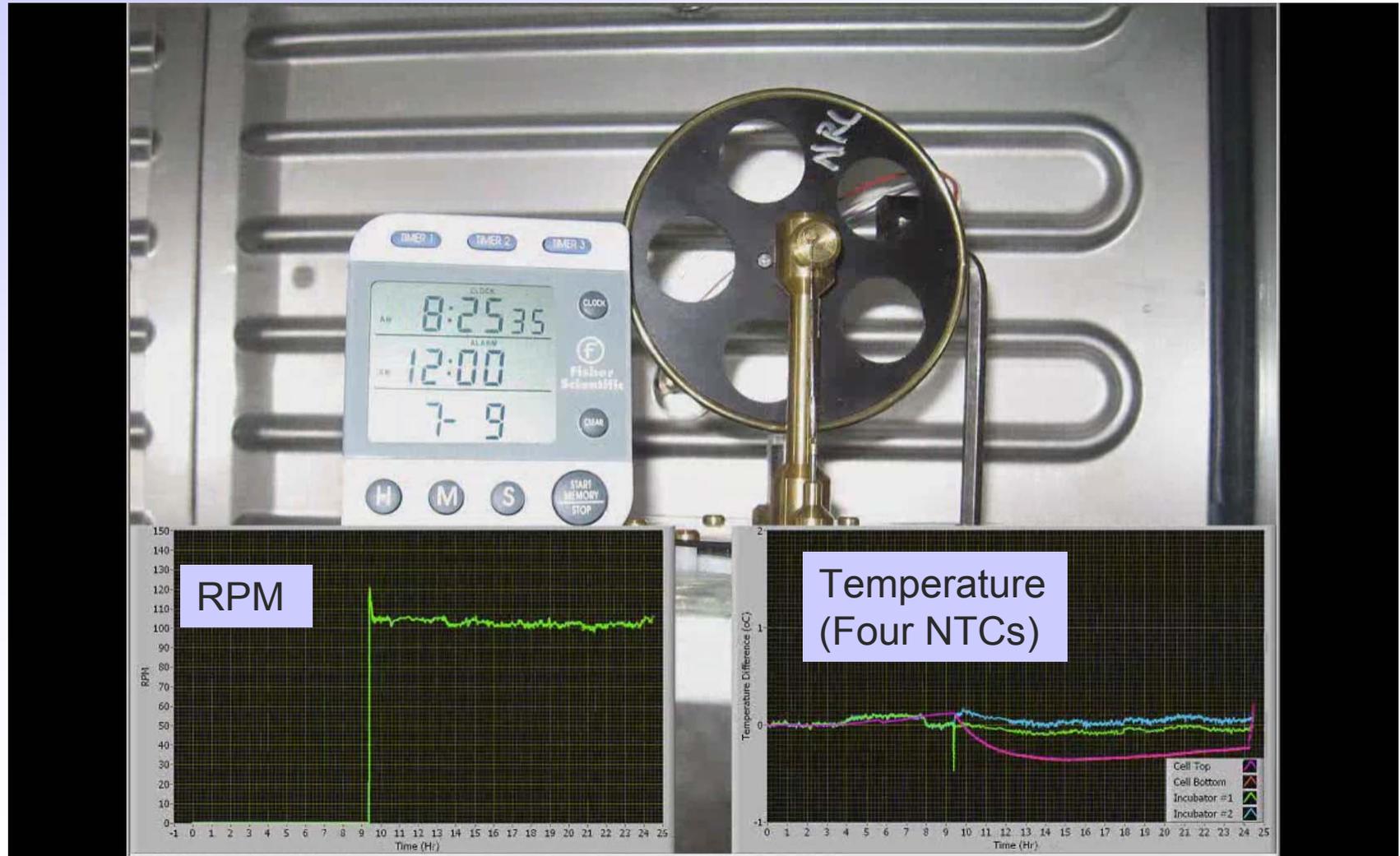
Movie showing D_2/Pd NP partially running a Sterling Engine





Useful Work?

Movie showing D_2/Pd NP partially running a Sterling Engine





Caution Flag



- For system under discussion:

✓ ○ Heat_{Out} >> Heat_{calculated}

✗ ○ H₂ ≈ D₂

(External Control)

- For NRL, Chart of Happiness for gas loading is:

At 50%



Summary - What Has NRL Observed?

@NRL:

- No evidence of nuclear products
 - No X-Rays, neutrons, gammas, or radioactive materials
- No evidence for transmutation
- Evidence of excess heat in electrochemical cells
 - Instrumental artifacts not ruled-out
- Evidence of excess heat in gas loading experiments
 - All possible chemistry not ruled-out

Phenomenon deserves further study

Three greatest gifts

That you can give a curious individual



1. It is possible
2. It is practical
3. It is secret



Wright brothers airplane - December 17, 1903

Curtiss - 1907

Curtiss Aeroplane Company - 1910

Curtiss-Wright Corporation -1929



Acknowledgements



Off the Chart!

Questions

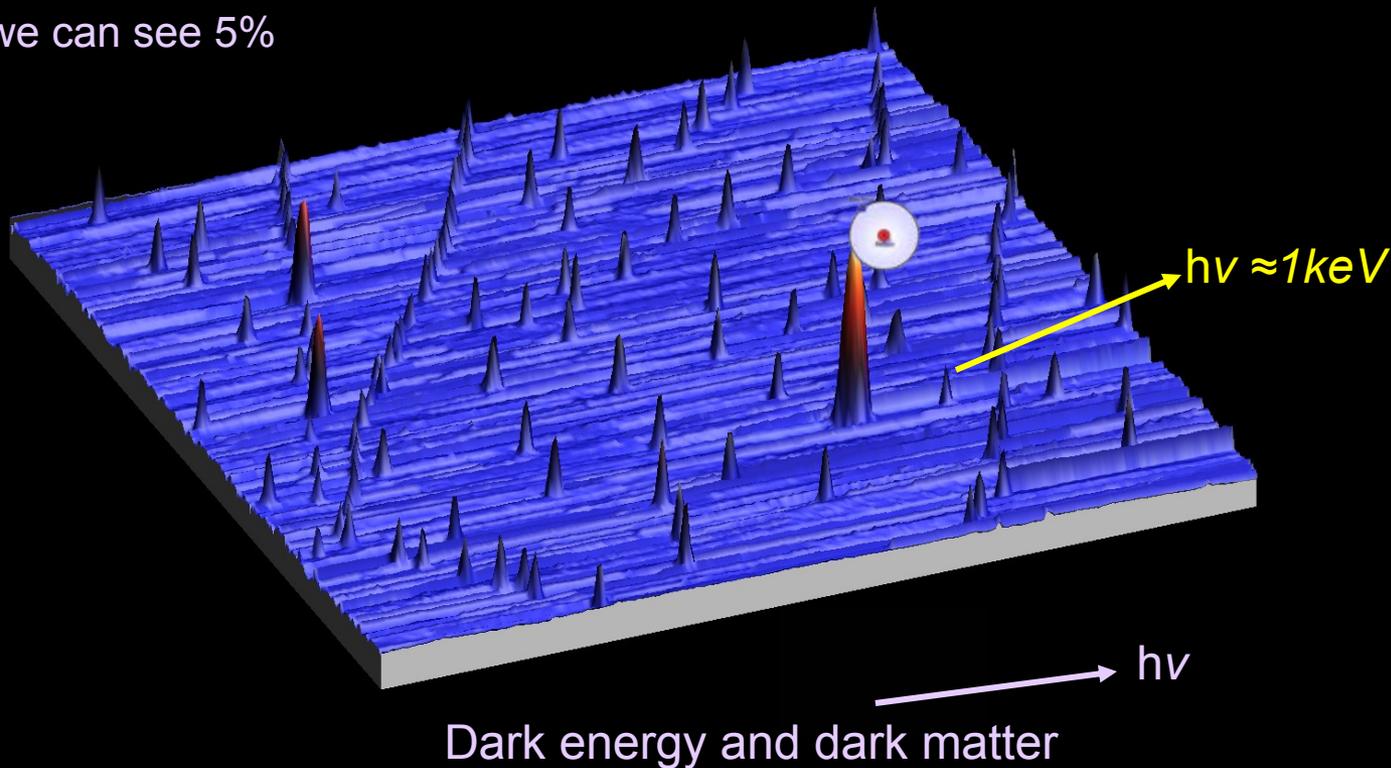
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Theory

Protons being converted into dark matter radiate energy in “normal” and dark energy regimes

- Dark energy 70% of universe
- Dark matter 25% of universe
- Stuff we can see 5%



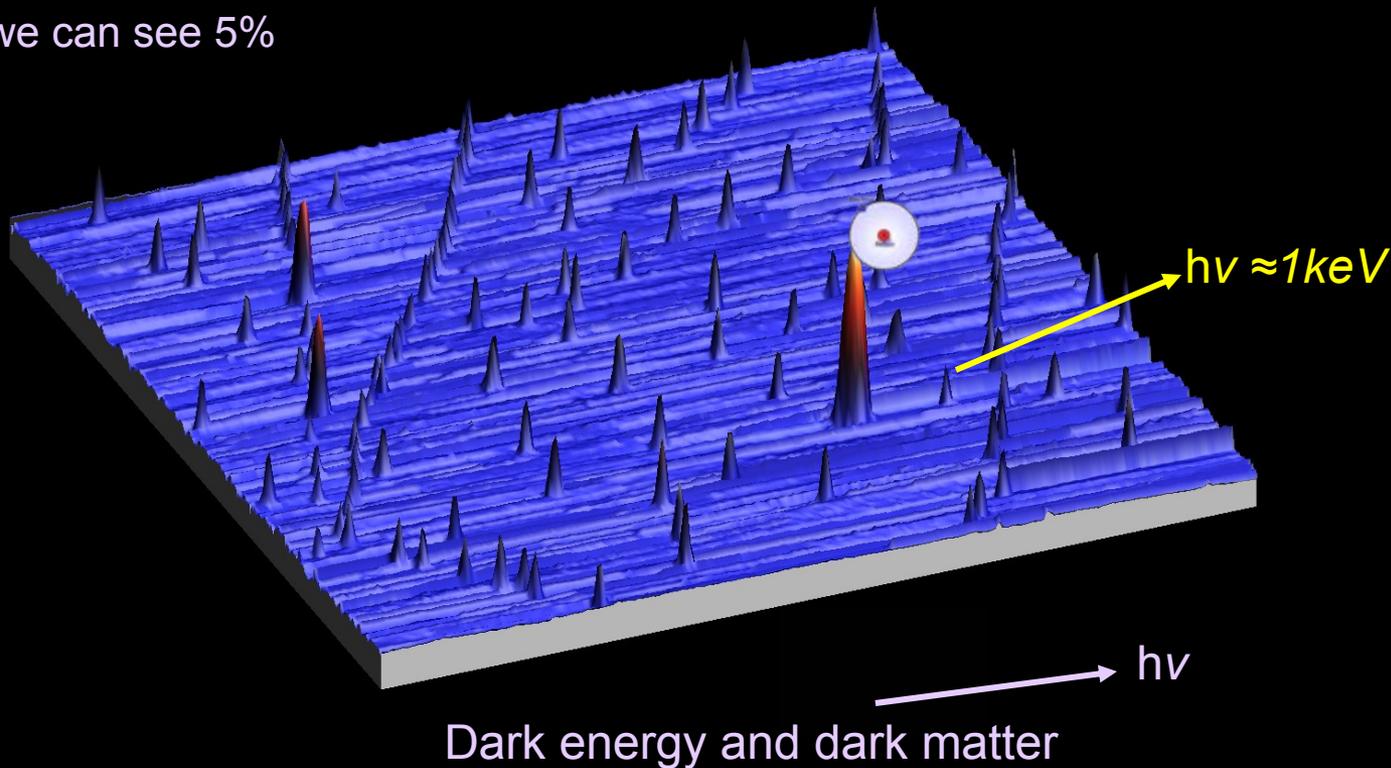
➤ Why 1 keV?

- Difficult energy to detect as non-penetrating and easily absorbed
- In electronic noise
- Film possible but issues with hydrogen radicals



Theory

- Dark energy 70% of universe
- Dark matter 25% of universe
- Stuff we can see 5%



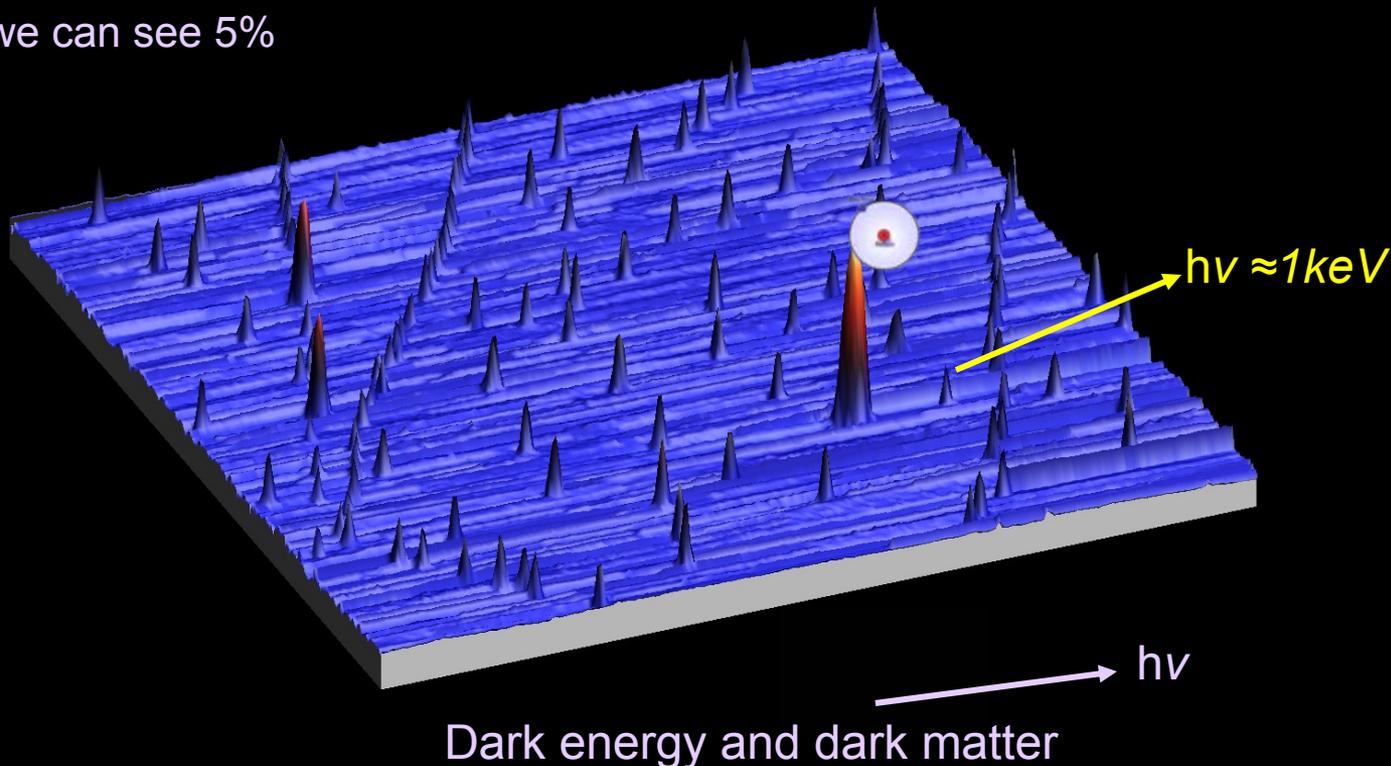
- How many protons?
 - $1\text{g} = 600\text{W}$ for 48 hr or 104 MJ
 - Issue is this low weight loss would look like a leak



Theory

Makes predictions but difficult experiments to confirm

- Dark energy 70% of universe
- Dark matter 25% of universe
- Stuff we can see 5%



- Violates principle of macroscopic reversibility
 - Normal protons can interact with catalyst to make dark matter
 - Dark matter cannot interact with normal matter to reverse process
 - Pumping X-rays onto catalyst could make protons at very low level
 - Difficult to distinguish from normal water background