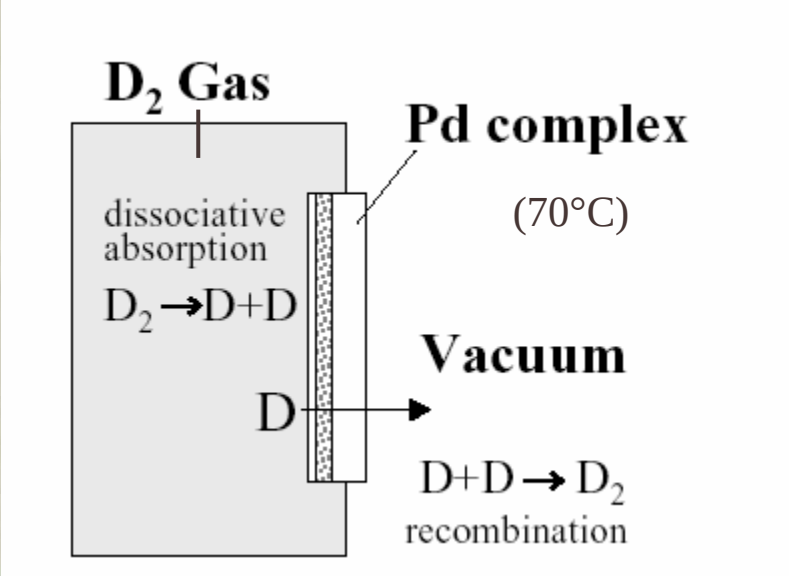
Evaluation of the Claim of Transmutation of Cesium to Praseodymium with the Mitsubishi Heavy Industries (MHI) Structure – Part 1

Kenneth Grabowski, David Kidwell, Catalina Cetina, and Carmine Carosella
Naval Research Laboratory
Washington, DC 20375

ICCF-15

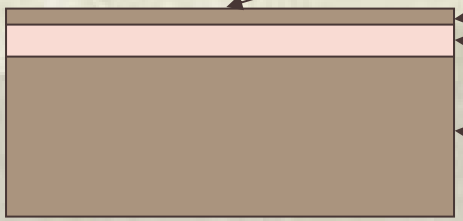
Summary of MHI Claims

- By permeating Deuterium though a Pd complex foil, various elemental transmutations can be made to happen
  - Reported transmutations:
    - $^{88}Sr \rightarrow {}^{96}Mo$ Addition of 4D
    - $^{133}Cs \rightarrow {}^{141}Pr$ Addition of 4D
    - $^{137}Ba \rightarrow {}^{149}Sm$ Addition of 6D



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

Pd complex used



25 mm x 25 mm

Thin Cs seed layer on surface

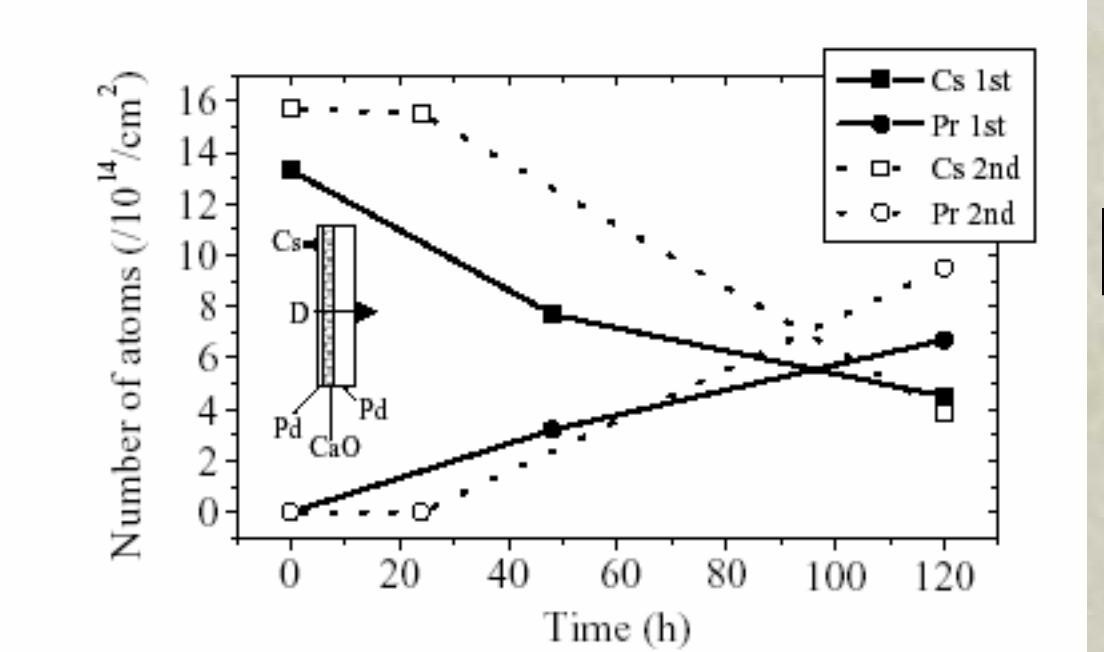
40 nm - Pd Cap Layer
100 nm total - 5 x (2 nm CaO / 18 nm Pd)

100 μm - Bulk Pd

QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

Cross section TEM of Pd Complex,
Y. IWAMURA, et al., Proc. ICCF-10, Cambridge MA 2003

Apparent transmutation of Cs into Pr observed by XPS



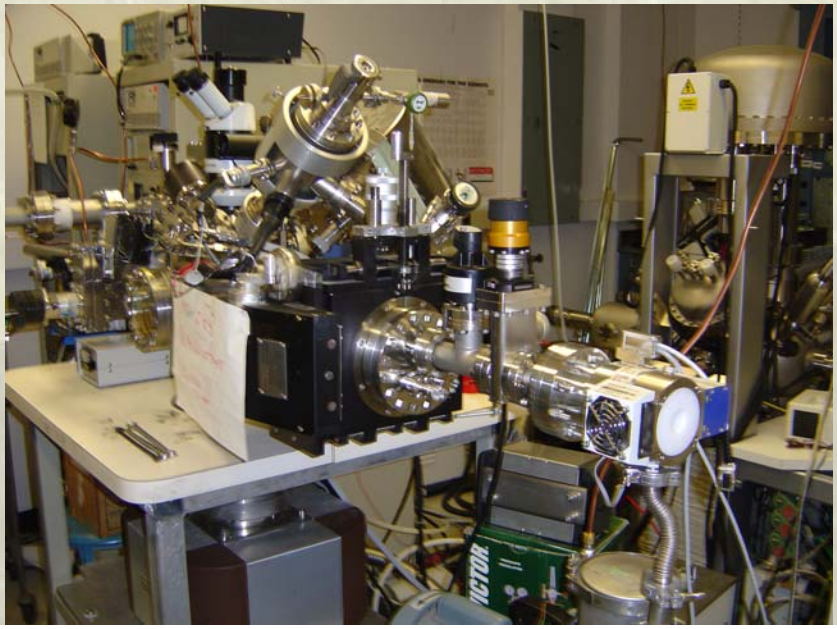
$1x10^{15}$ Pr = 235 ng

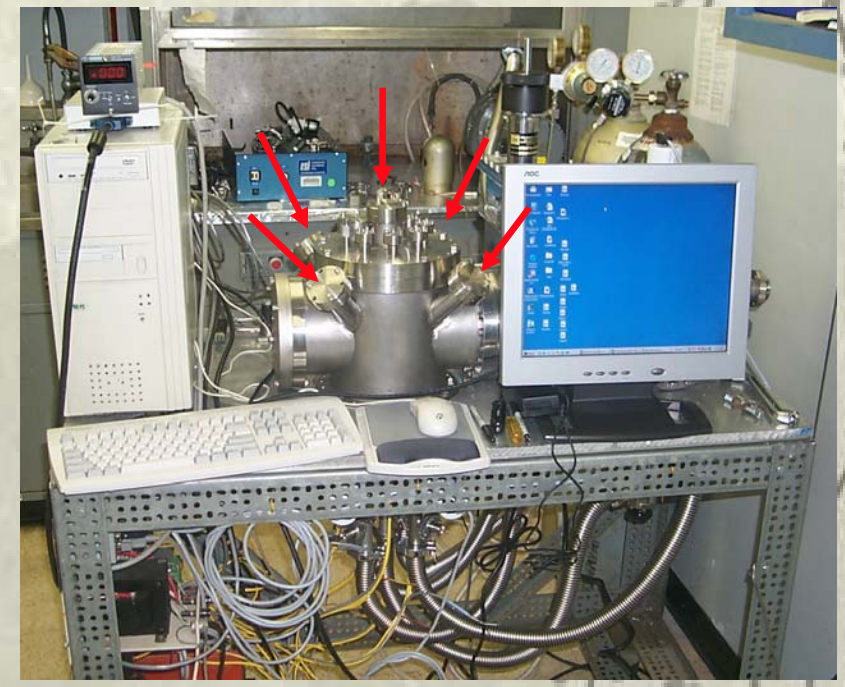
Elemental Analysis of Pd Complexes: Effects of D_2 Gas Permeation
Y. Iwamura , M. Sakano and T. Itoh
Jpn. J. Appl. Phys. Vol. 41 (2002) pp. 4642–4650

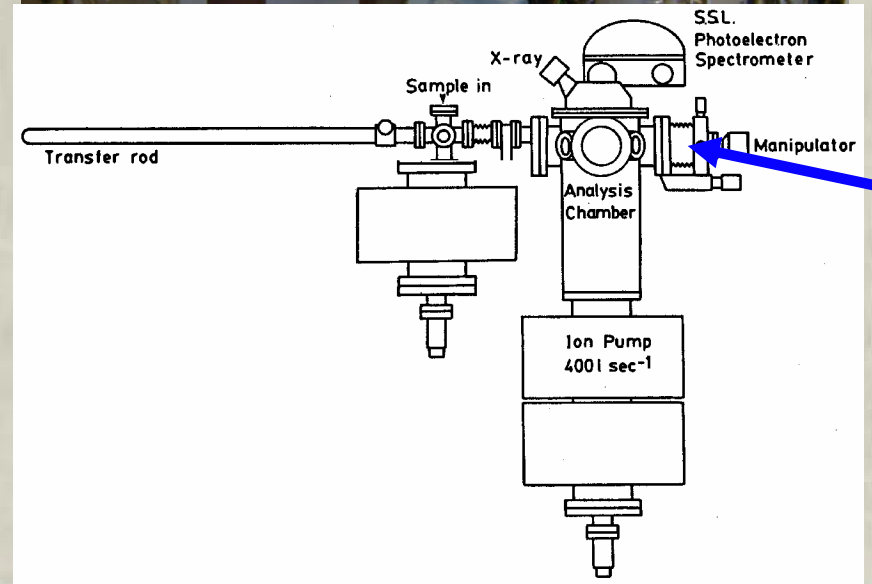
NRL Research Effort

- Collaborated with MHI to verify Pr present on their permeated samples
  - Initially with Accelerator Mass Spectrometry
  - Eventually with ICP-MS
  - NRL convinced Pr was present on some samples
- NRL attempted to independently reproduce result
  - Modified XPS instrument to enable permeation of Pd sample
  - Built 5-sample chamber with line-of-site view of sample
  - Developed Pd-complex fabrication capability
  - Pd complex structures would not permeate sufficient D
  - Unsuccessful at producing Pr
- Therefore, performed joint research with MHI

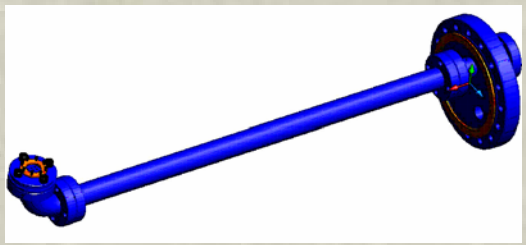
Modified XPS and 5-Sample Chambers







Modified Sample Stage to Accept Reaction Cell



Experimental Plan for Joint Research with MHI

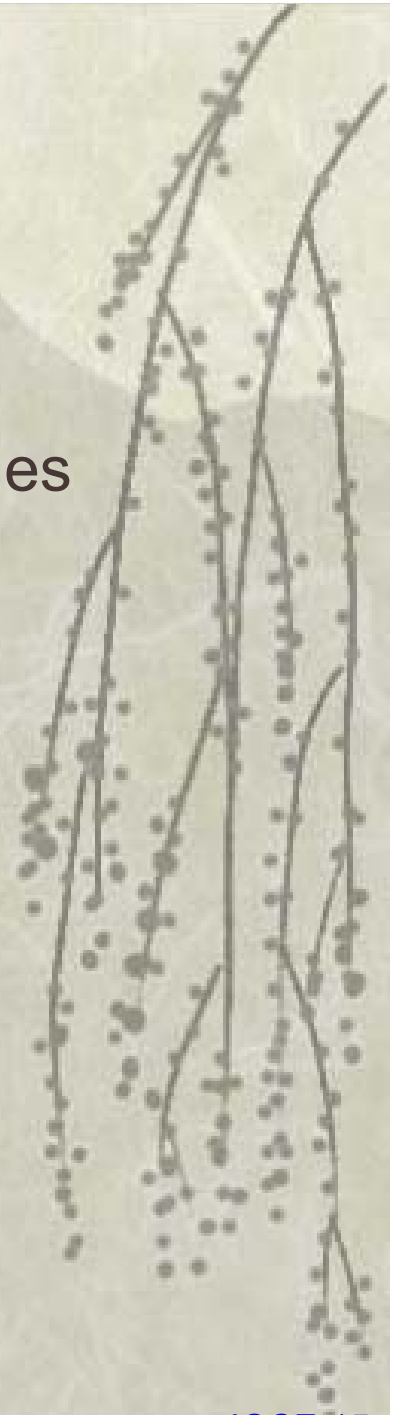
- Bulk analysis of Pd
  - Key decision point
  - Sufficient Pr to explain MHI result?
  - NO! < 10 ppb/wt, i.e., < 7 ng total
- Sample production at MHI
- NRL observers at MHI
- Analysis at MHI and NRL

NRL Analysis (Pr in components)

Group		Cs solution	Pd foil	Comment
MHI components		1 of 1	4 of 2	Executed of Planned
			MHI 106 (< 0.6 ng/cm^2)	GDMS by NRL
			MHI 107 (<0.6 ng/cm^2)	GDMS by NRL
		< 8 pg per 20 mL	MHI 106 (42 pg/cm^2)	Bulk ICP-MS by NRL
			MHI 107 (53 pg/cm^2)	Bulk ICP-MS by NRL
			MHI 157 (<3 pg/cm^2)	Surface ICP-MS by NRL
			MHI 158 (<3 pg/cm^2)	Surface ICP-MS by NRL
Chisai	Cs Complex, Non-Permeated Blank	Non-Cs Complex, Permeated Blank	Cs Complex, Permeated	Comment
	0 of 3	0 of 3	3 of 3	Executed of Planned
			MHI 113 (<4 pg/cm^2)	Surface ICP-MS by NRL
			MHI 113 (36 pg/cm^2)	Bulk ICP-MS by NRL
			MHI 119 (10 pg/cm^2)	Surface ICP-MS by NRL
			MHI 119 (30 pg/cm^2)	Bulk ICP-MS by NRL
			MHI 124 (<3 pg/cm^2)	Surface ICP-MS by NRL
			MHI 124 (42 pg/cm^2)	Bulk ICP-MS by NRL

Observer's Tasks

- Monitor all production steps of selected samples
  - Starting materials and their cleaning
  - Annealing foils
  - Etching foils
  - Deposition of multilayer film
  - Deposition of Cs
  - Permeation processing
  - Storing and shipping

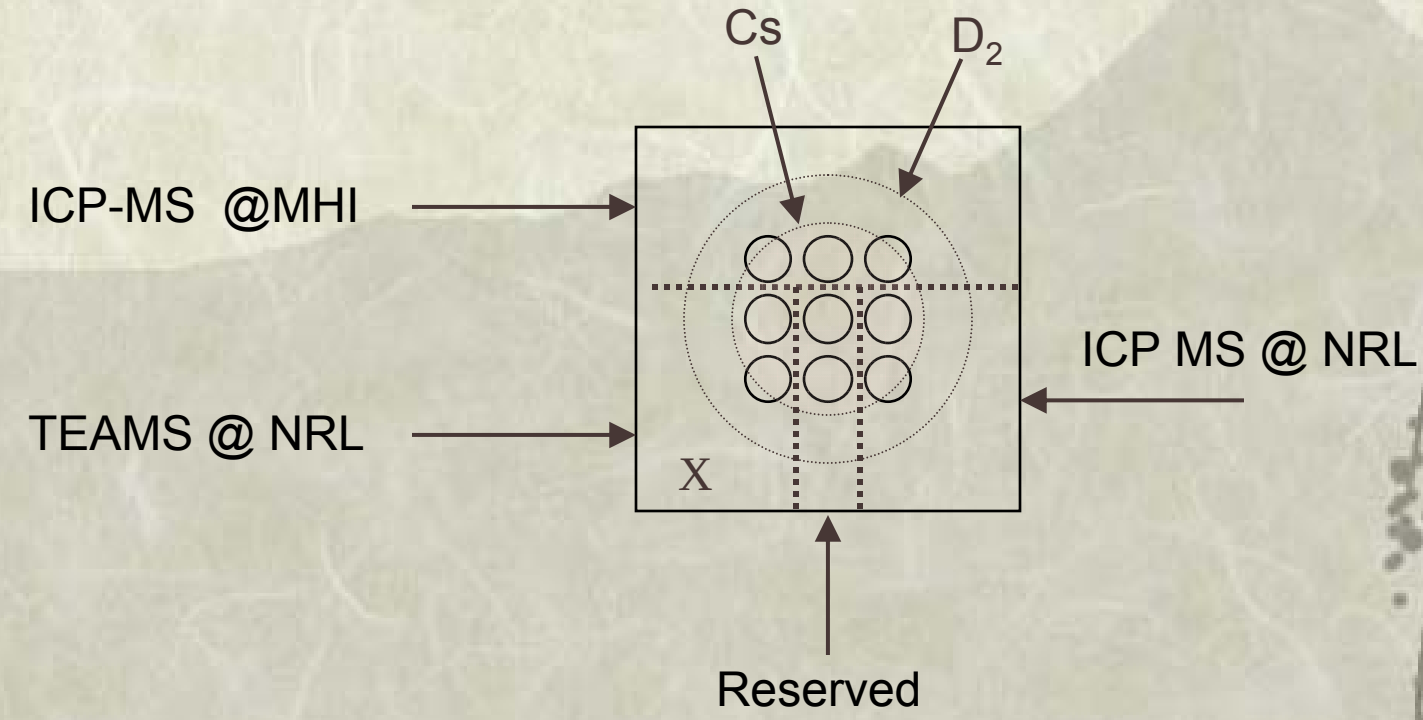


Observer's Tasks (2)

- Record observations without disrupting tasks
  - Video recording of key steps
  - Audio recording of instructions
  - Notations of events
- Review observations
  - Amongst NRL staff
  - With MHI staff, using translator for key discussions
- Report back to US
  - Frequently, digitally and verbally
  - In summary documentation

Foil - Analysis Layout

(typical)



Note: Lower 2/3 Sent to NRL for Analysis

Foil Specimens Planned

		Sample	Cs blank	Perm. blank
	Pd Foil, as Manufactured	**Cs Complex, Permeated**	**Cs Complex, Non-Permeated**	**Non-Cs Complex, Permeated**
XPS Chamber		3	3	3
Chisai Chamber		3	3	3
Total	2	6	6	6
Demonstrates:	Pr Not in Pd Foil	Pr Production	Initial Structure Free of Pr	No Bulk Redistribution Not From D_2

MHI samples & MHI analysis

Cs blank Perm. blank Sample

Group	Cs Complex, Non-Permeated Blank	Non-Cs Complex, Permeated Blank	Cs Complex, Permeated	Comment
XPS	3 of 3	3 of 3	3 of 3	Executed of Planned
	MHI 127 (0 ng/cm^2)	MHI 123 (0 ng/cm^2)	MHI 118 (13.4/20 ng/cm^2)	Surface ICP-MS by MHI
	MHI139 (0 ng/cm^2)	MHI 135 (0 ng/cm^2)	MHI 131 (7.8/9.2 ng/cm^2)	Surface ICP-MS by MHI
	MHI 151 (0 ng/cm^2)	MHI 147 (0 ng/cm^2)	MHI 143 (13/26 ng/cm^2)	Surface ICP-MS by MHI
Chisai	1 of 3	1 of 3	4 of 3	Executed of Planned
	MHI 101 (0 ng/cm^2)	MHI 109 (0 ng/cm^2)	MHI 103 (0 ng/cm^2)	Surface ICP-MS by MHI
			MHI 113 (0 ng/cm^2)	Surface ICP-MS by MHI
			MHI 119 (0 ng/cm^2)	Surface ICP-MS by MHI
			MHI 124 (0 ng/cm^2)	Surface ICP-MS by MHI
				Unknown Problem On Hold
Total	4 of 6	4 of 6	7 of 6	Executed of Planned

* Data listed from Takasago/Toray facilities

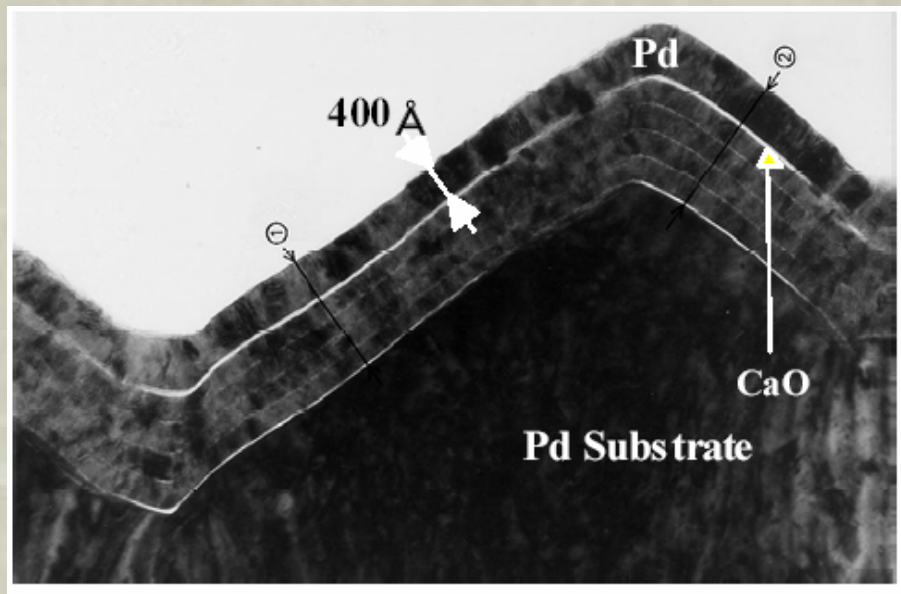
Evaluation of the Claim of Transmutation of Cesium to Praseodymium with the MHI Structure – Part 2

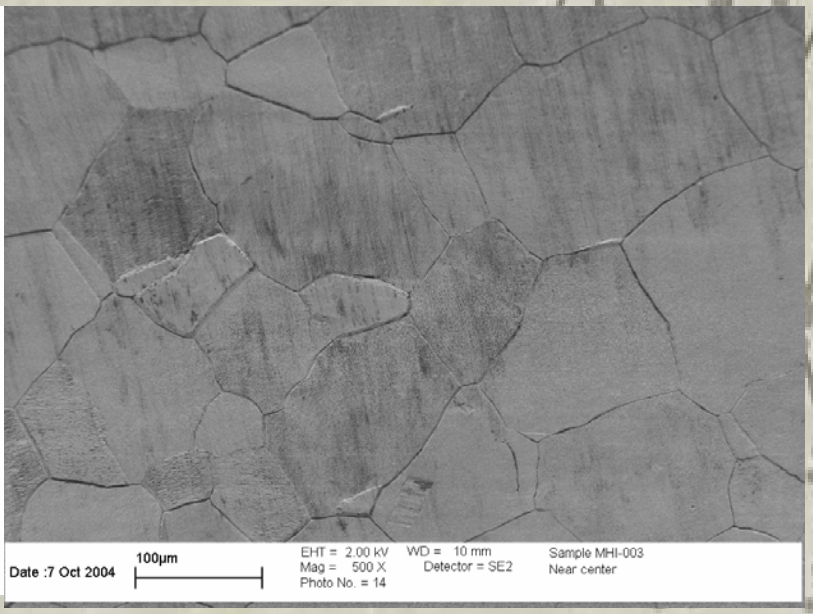
David Kidwell, Kenneth Grabowski, Catalina Cetina, and Carmine Carosella
Naval Research Laboratory
Washington, DC 20375
(202)767-3575
David.Kidwell@nrl.navy.mil

ICCF-15

Summary of MHI Claims

- By permeating Deuterium though a complicated layer various elemental transmutations can be made to happen
  - Reported transmutations:
    - $^{88}Sr \rightarrow {}^{96}Mo$ Addition of 4D
    - $^{133}Cs \rightarrow {}^{141}Pr$ Addition of 4D
    - $^{137}Ba \rightarrow {}^{149}Sm$ Addition of 6D



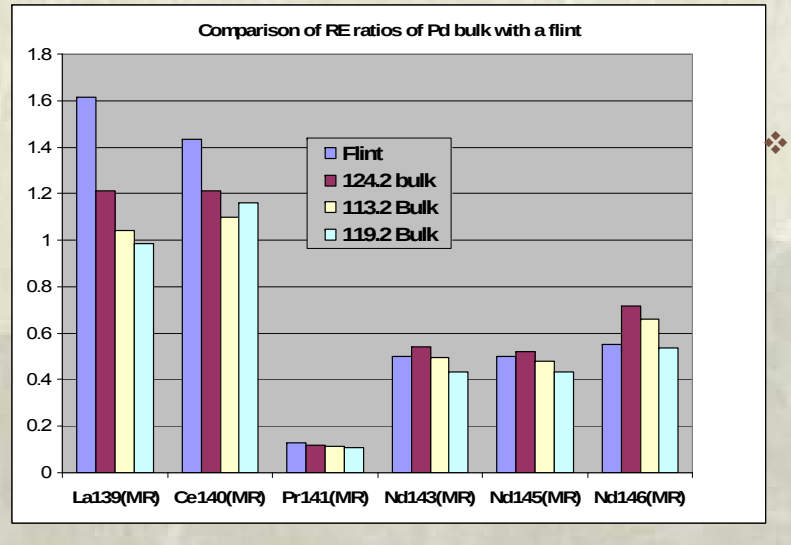


Cross section of Pd complex (Pd/CaO/Pd) observed by TEM.
From: Iwamura, Y., et al. Low Energy Nuclear Transmutation In Condensed Matter Induced By D2 Gas Permeation Through Pd Complexes: Correlation Between Deuterium Flux And Nuclear Products. in Tenth International Conference on Cold Fusion. 2003.

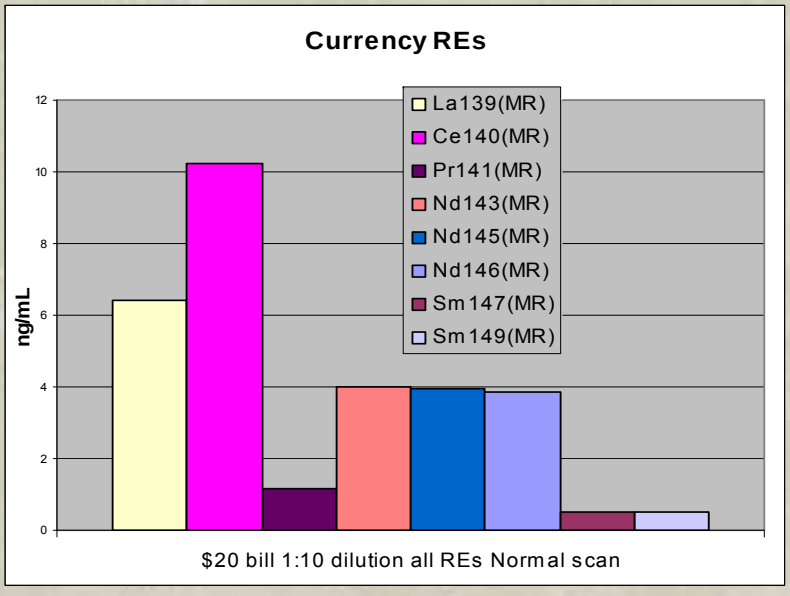
From: C. Carosella - Visual Observations of MHI and NRL Samples

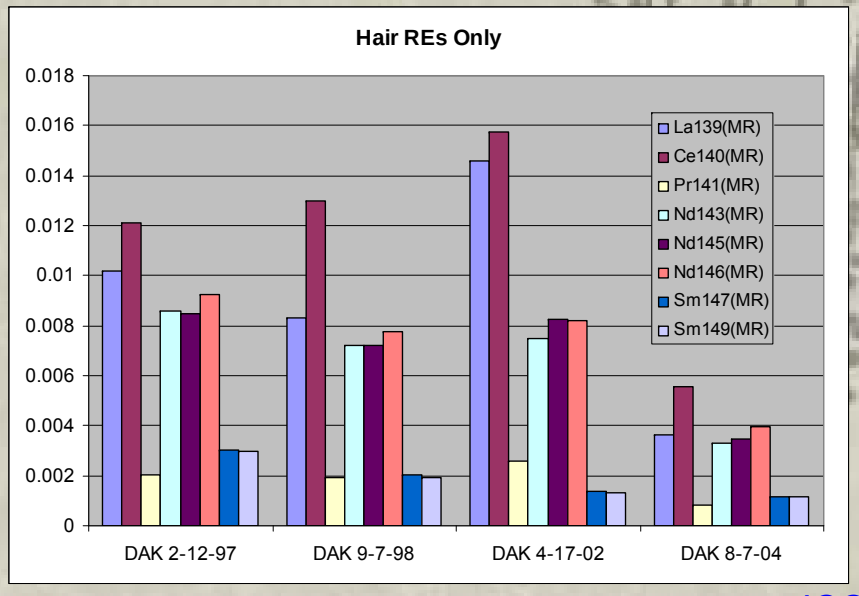
Rare Earths in the Environment

Rare earths are not so rare



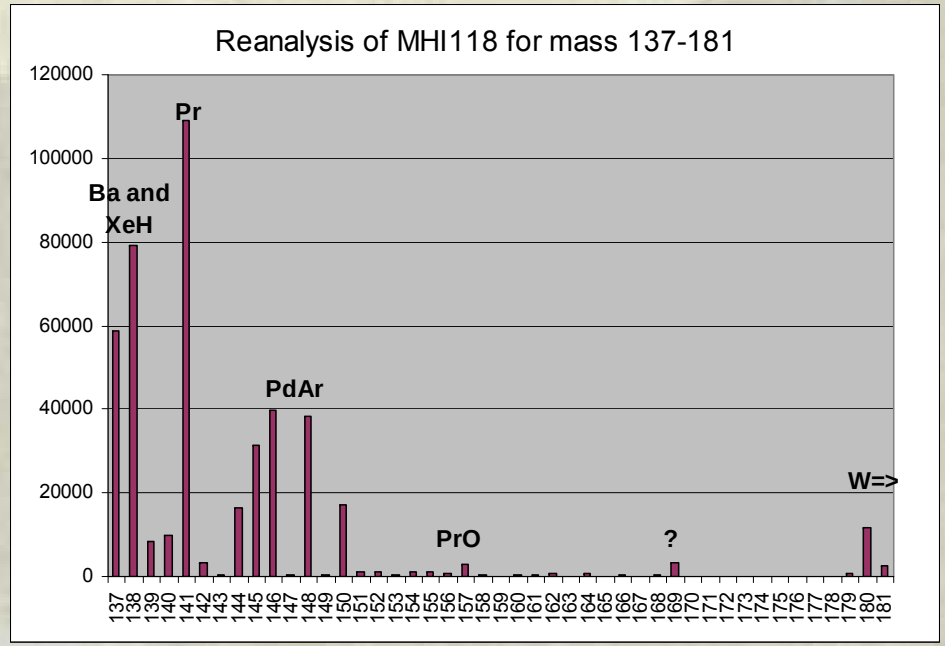
- Environmental sources of Pr
  - Flints – "Misch Metal"
  - Specialty glazes on ceramics
  - Phosphors in CRTs (tends to be with other REs)

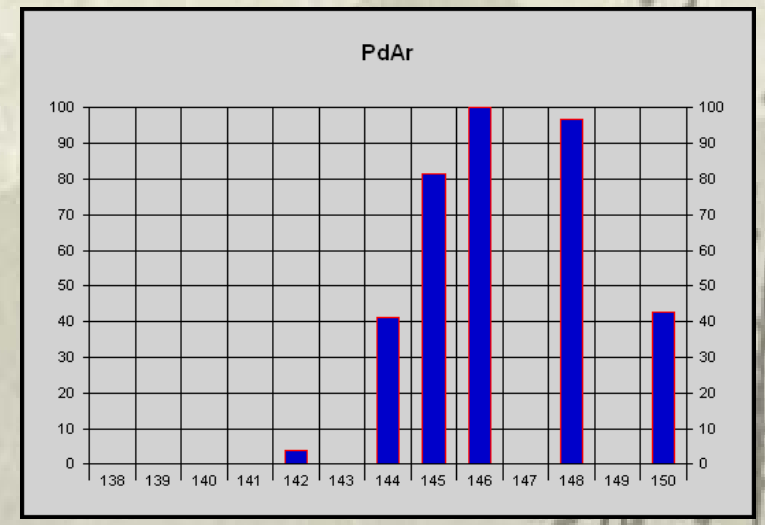


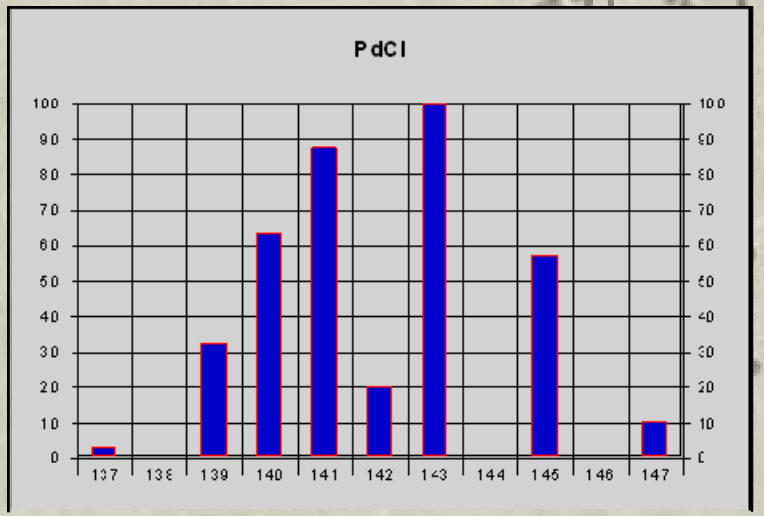


Molecular Interferences or Misch Metal Contamination?
No!

- Two different ICP-MS instruments

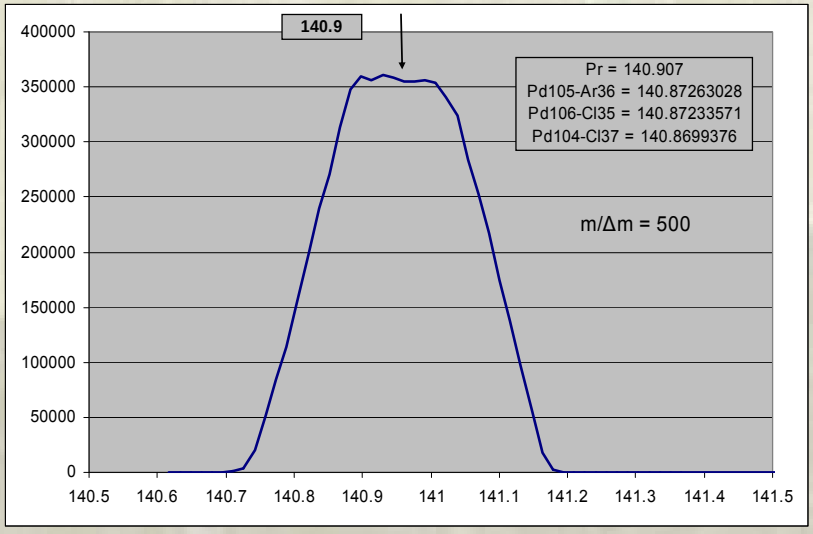




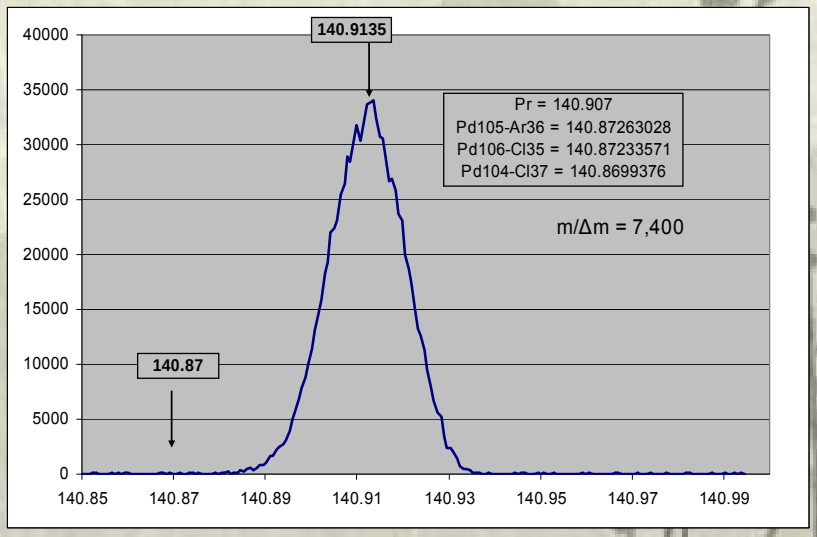


Raw ICP-MS Curves for MHI-006-box

Spectra shown at various resolutions

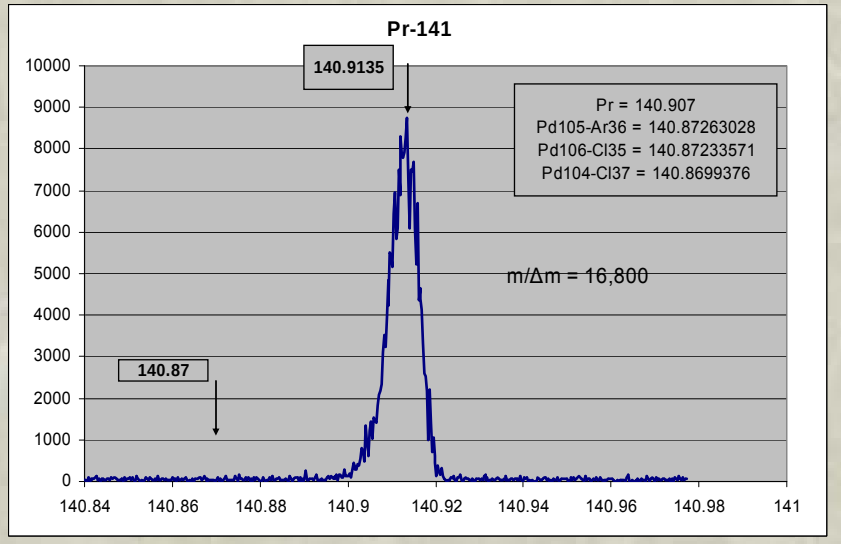


Low - file#Oct20d15LR



Medium - file#Oct20d15MR

High - file#Oct20d15HR



Masses not perfect due to magnet drift

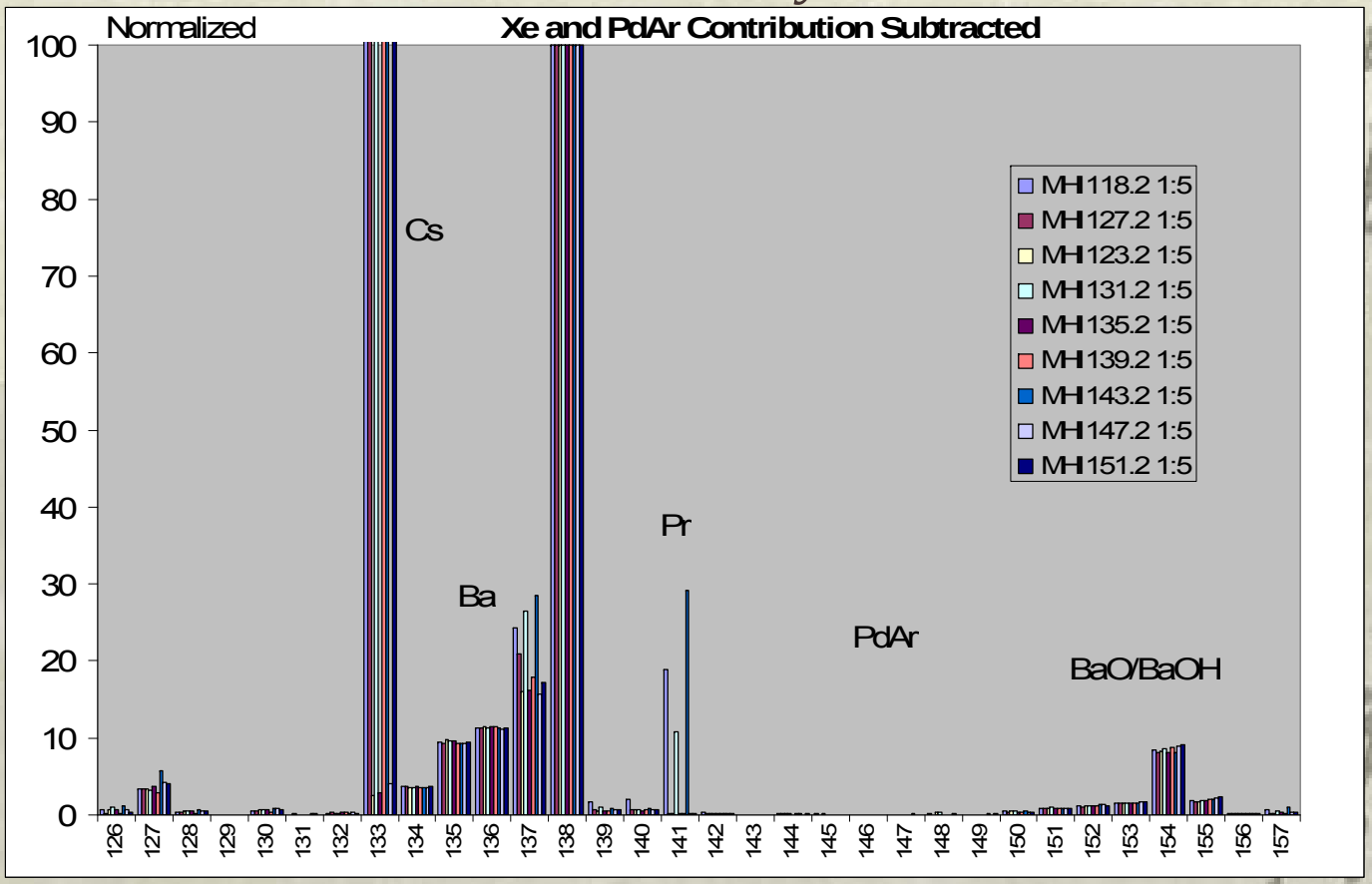
Cross Validation of Pr Levels

- 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)
  - One set from baby chamber – all negative
    - Included a fully permeated sample

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

More Detailed Scan of the Nine Samples

Extracted at MHI – Analyzed at NRL



- Note clean RE levels and high Cs levels

Concentration on Surface from Bulk

No!

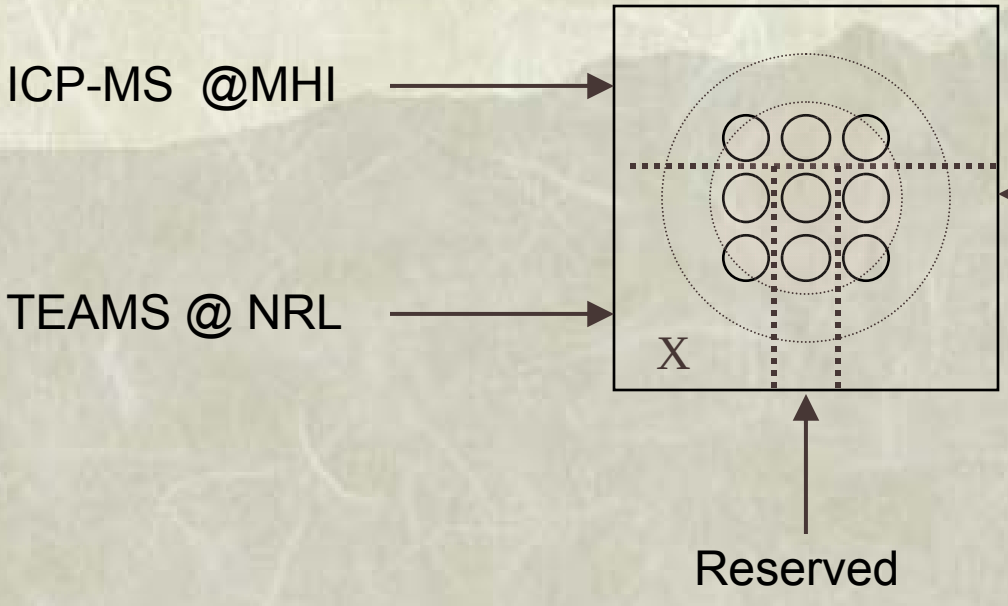
Sample	Pr (ng/cm^2) Raw	Pr (ng/cm^2) Blank subtracted
MHI119.2	0.108	0.030
MHI113.2	0.114	0.036
MHI124.2	0.121	0.042

From UM 9-25-05

- Levels too low to account for Pr in permeated samples
  - Methodology discussed at ICCF-14

Foil - Analysis Layout

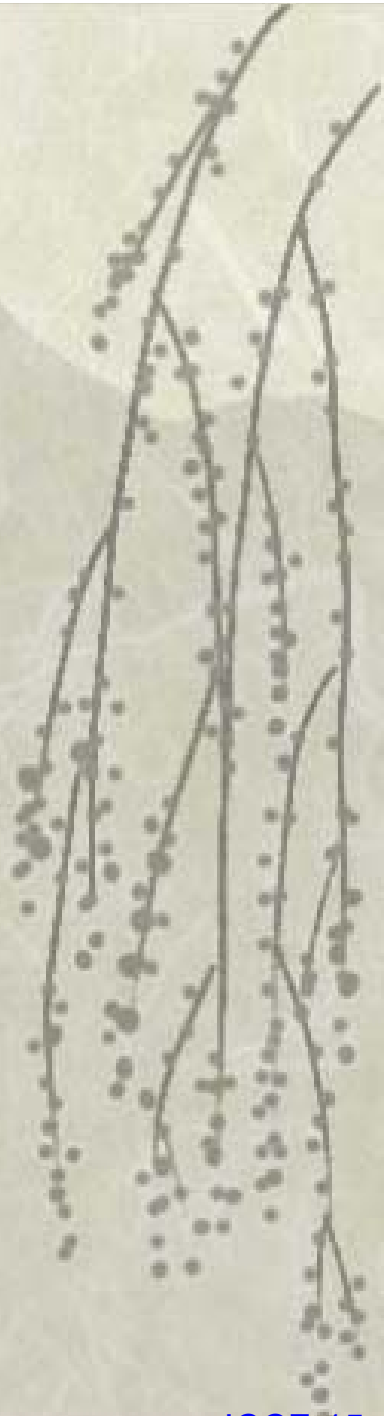
(typical)



ICP MS @ NRL

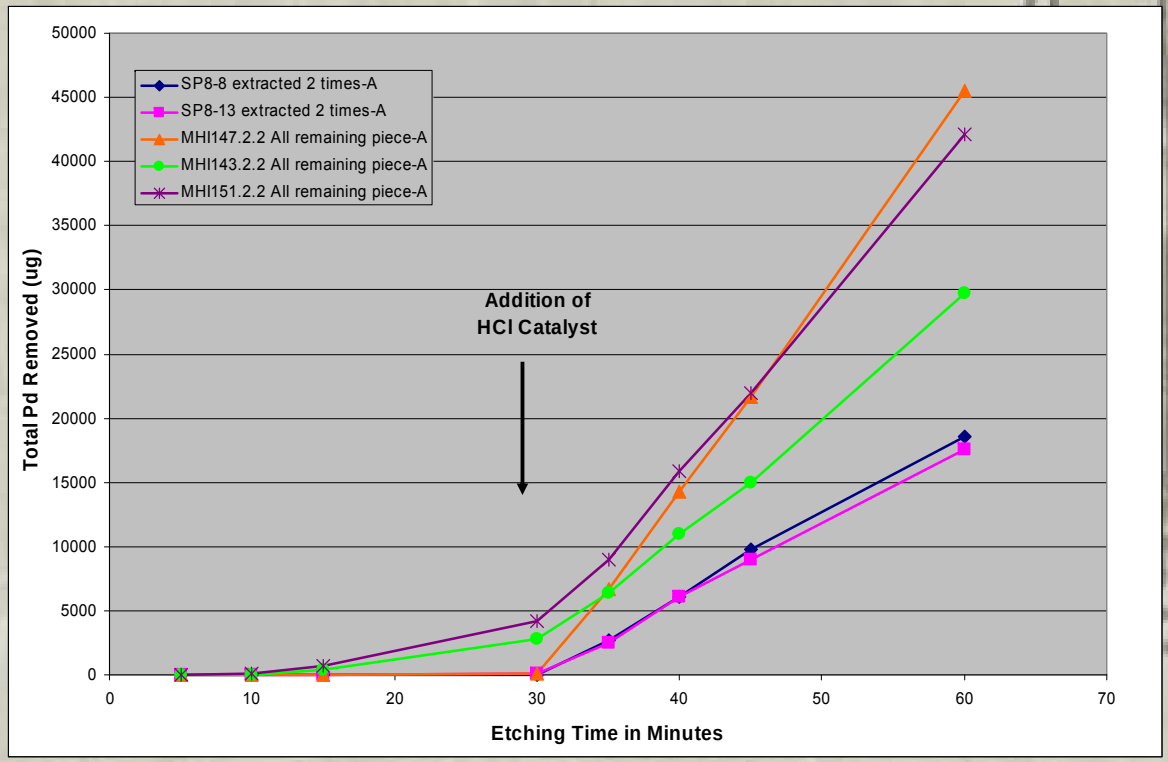
Note: Lower 2/3 Sent to NRL for Analysis

Big Issue – Could not find Pr at NRL on our part of the foils!



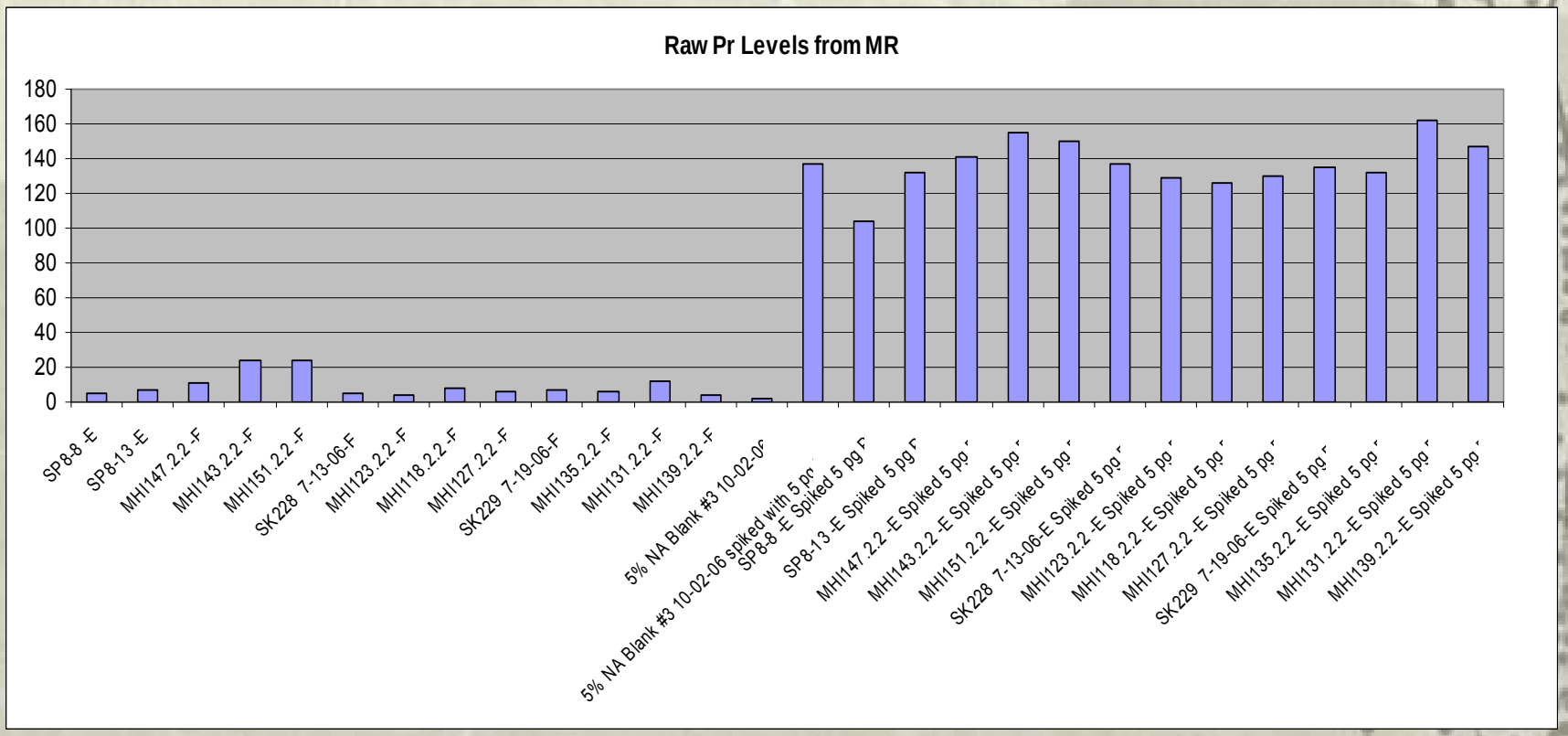
Example Showing That NRL Could Have Observed Pr, If Present

- Controlled etching done on all samples
  - Discussed at ICCF-14
- 8 aliquotes taken and bulk analysis done
  - >100 samples + banks + controls
- No Pr found



Example Showing That NRL Could Have Observed Pr, If Present

- One series spiked with equivalent of 170 pg/cm^2 Pr (~100x lower than expected)



Possible Explanations
for failure to find Pr

- 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
- Pr lost in transit or handling
  - Film unstable – Pr should be in boxes – checked and no
- Pd catalyzes reverse nuclear reaction with time?
  - Real stretch of science
- Pr not there – we clearly have the required sensitivity and specificity

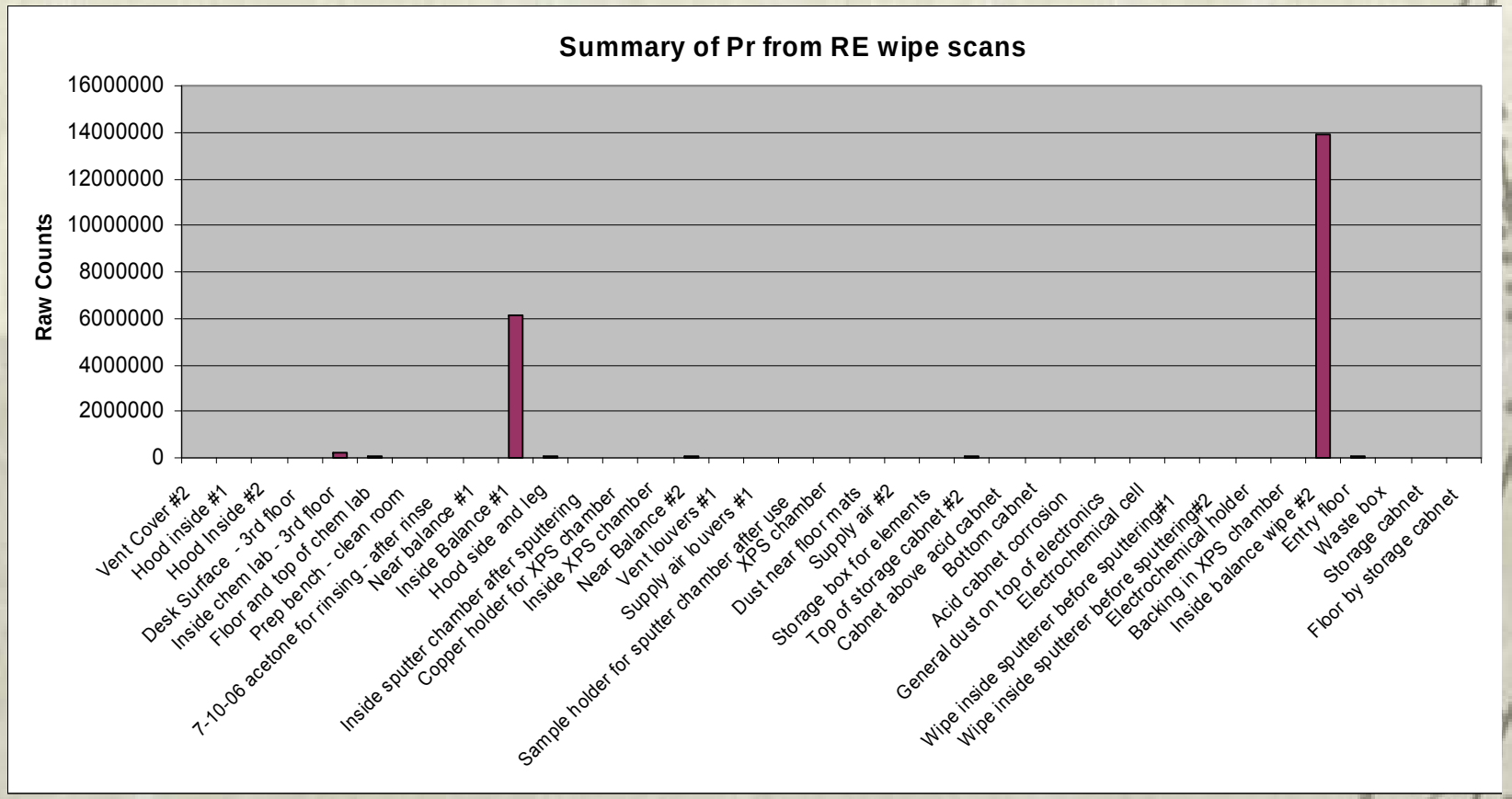
Months worth of work (frustration) later…

- Not etching enough Pd foil
  - Loss of deuterium with time causes lower Pd etching
- 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 negative with a commercial laboratory in Japan

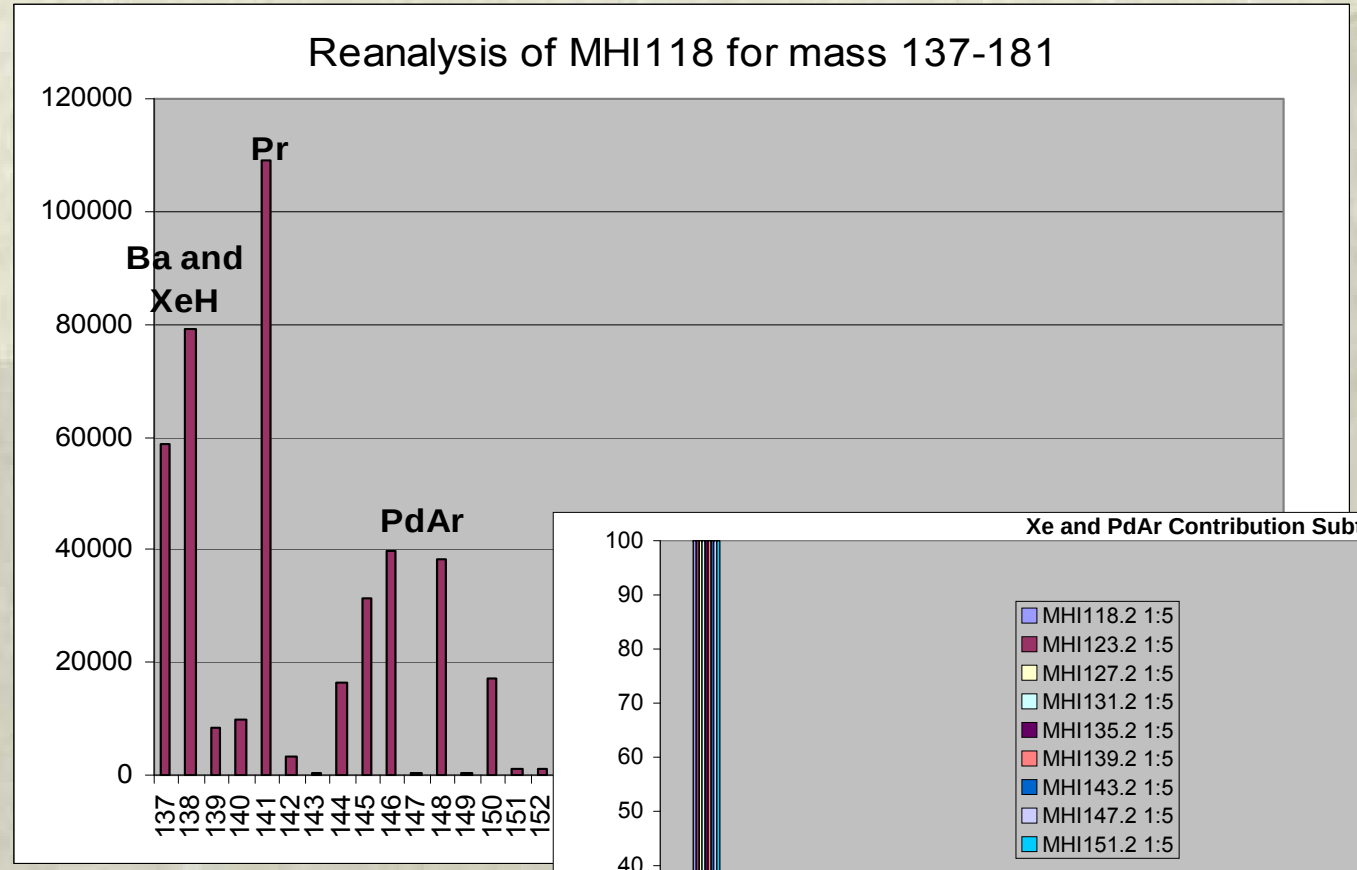
On whim, do environmental survey

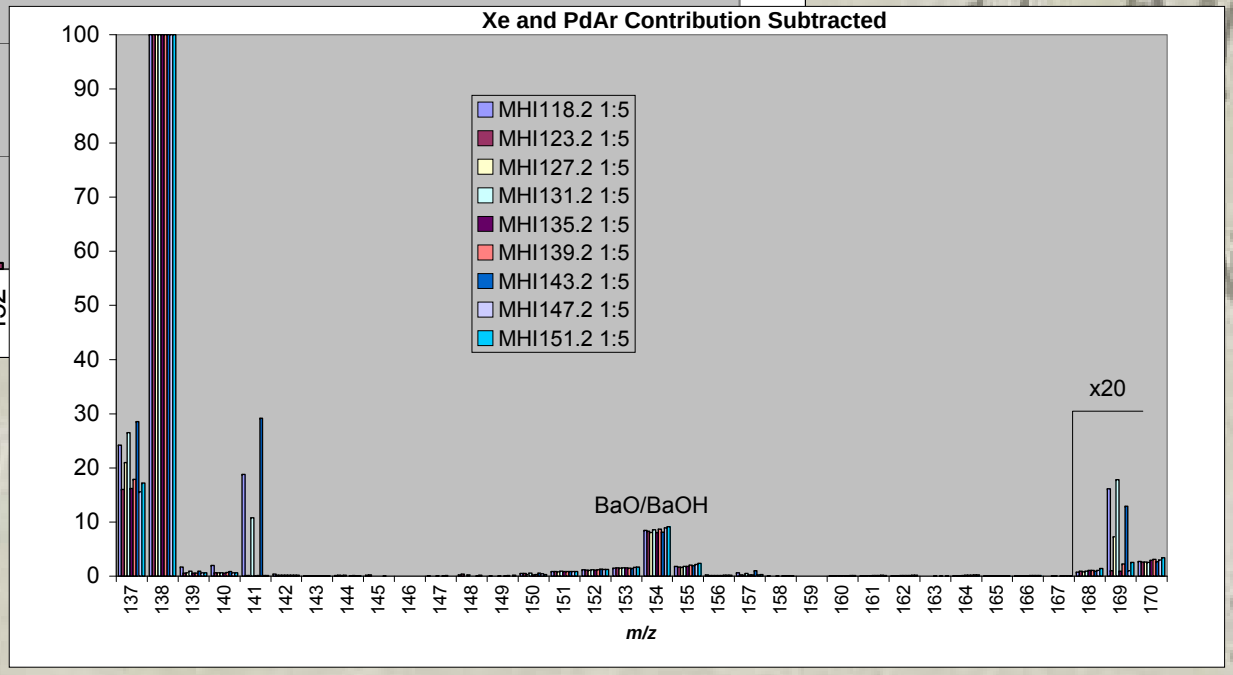
Results from Environmental Survey



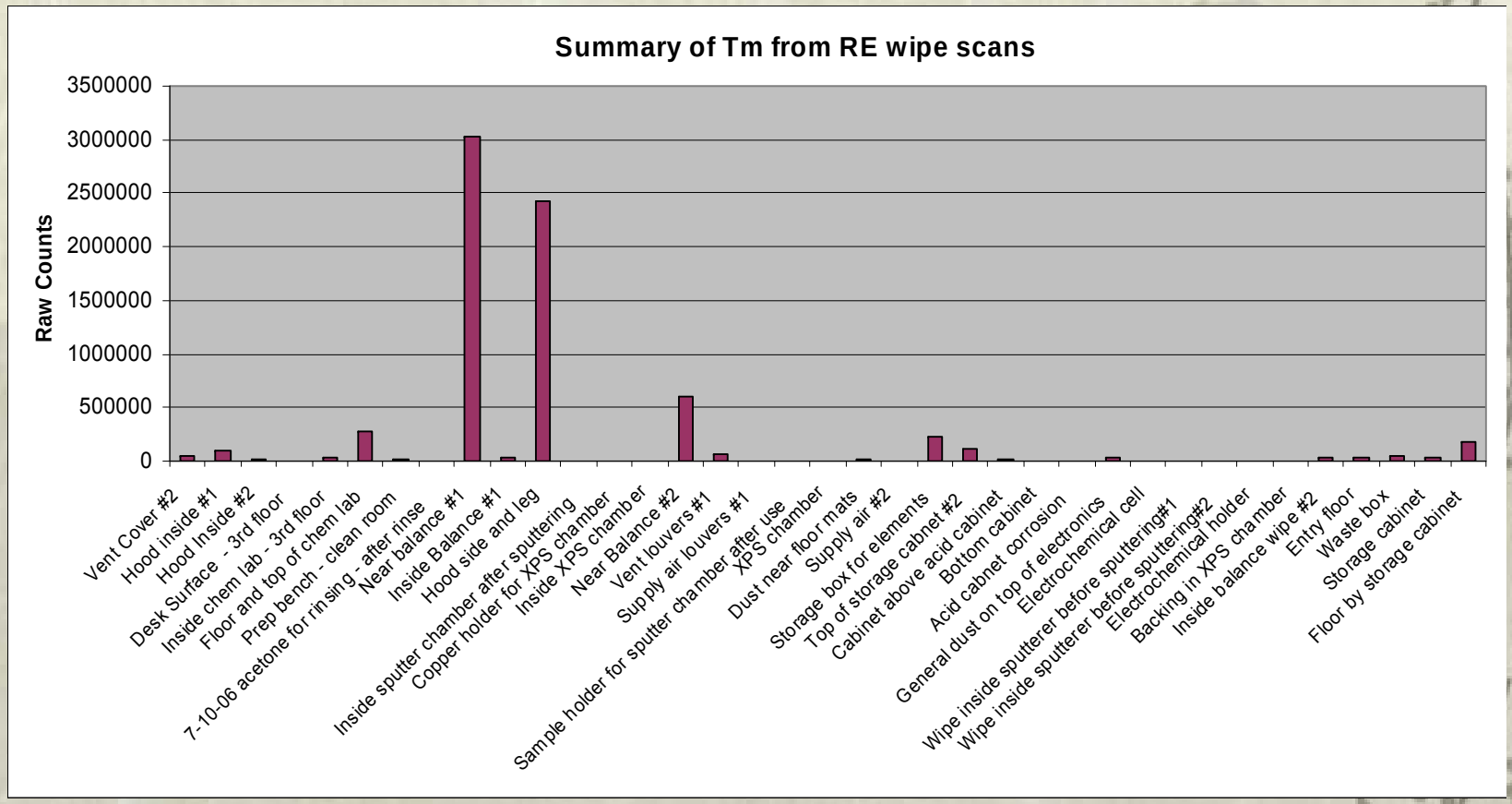
Excess ^{137}Ba and Thulium Found in Samples

Origin??





Results from Environmental Survey



Trace Elements in Samples

Sample	Excess Ba137 present	Total amount of Tm found (LR mode)
SP8-8 -E	No	0.005
SP8-13 -E	No	0.002
MHI147.2.2 -E	No	0.004
MHI143.2.2 -E	**Substantial**	**0.150**
MHI151.2.2 -E	Slight	0.016
SK228 7-13-06-E	No	0.004
MHI123.2.2 -E	No	0.006
MHI118.2.2 -E	**Substantial**	**0.082**
MHI127.2.2 -E	Substantial	0.029
SK229 7-19-06-E	No	0.003
MHI135.2.2 -E	No	0.006
MHI131.2.2 -E	**Substantial**	**0.199**
MHI139.2.2 -E	Substantial	0.008
Blank		0.001

Highlighted lines are the positive samples

Speculation
Explanations of Results

- Two types of contamination occur:
- ICP-MS
  - Extraction at MHI – contaminates the solutions
    - MHI extracts show Pr but NRL extracts do not
    - Individual doing extraction left MHI
      - "Lucky" tweezers??
    - Last samples prepared and extracted in presence of NRL personnel are blank
- *in situ* measurements - XRF/XPS – increase with permeation in a highly localized area
  - Contamination from dust in balance – contaminates the interior of the multi-layer structure with small particles of Pr
    - Blank runs not sensitive enough to see buried Pr
      - Under surface and localized
    - Pr migrates to surface under influence of the Deuterium flux
      - Spreads as migrates
    - Looks like production of Pr!

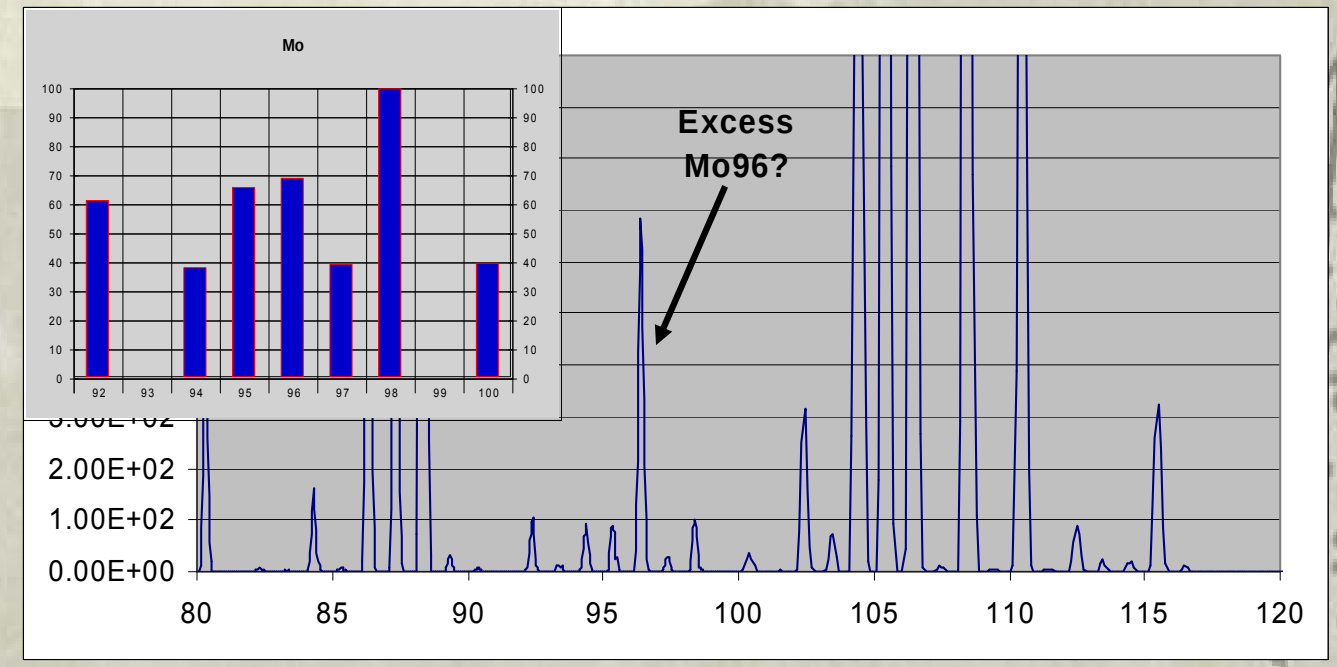
Speculation

Scenario for how this contamination could have occurred

- MHI tried other elements, believed to see ^{133}Cs ➔ ^{141}Pr by *in situ* XPS
  - Cu can be confused for Pr
  - Needed standard of Pr – made one up in the laboratory to confirm XPS or ICP-MS analysis
  - From that time forward, actually was looking at Pr but from what source?
- In initial XPS results, believed to see Sr ➔ Mo.
  - Secondary Ion Mass Spectrometry appeared to confirm ^{88}Sr ➔ ^{96}Mo
  - Observing other transmutations appears to strengthen each observation
    - How strong was the evidence?

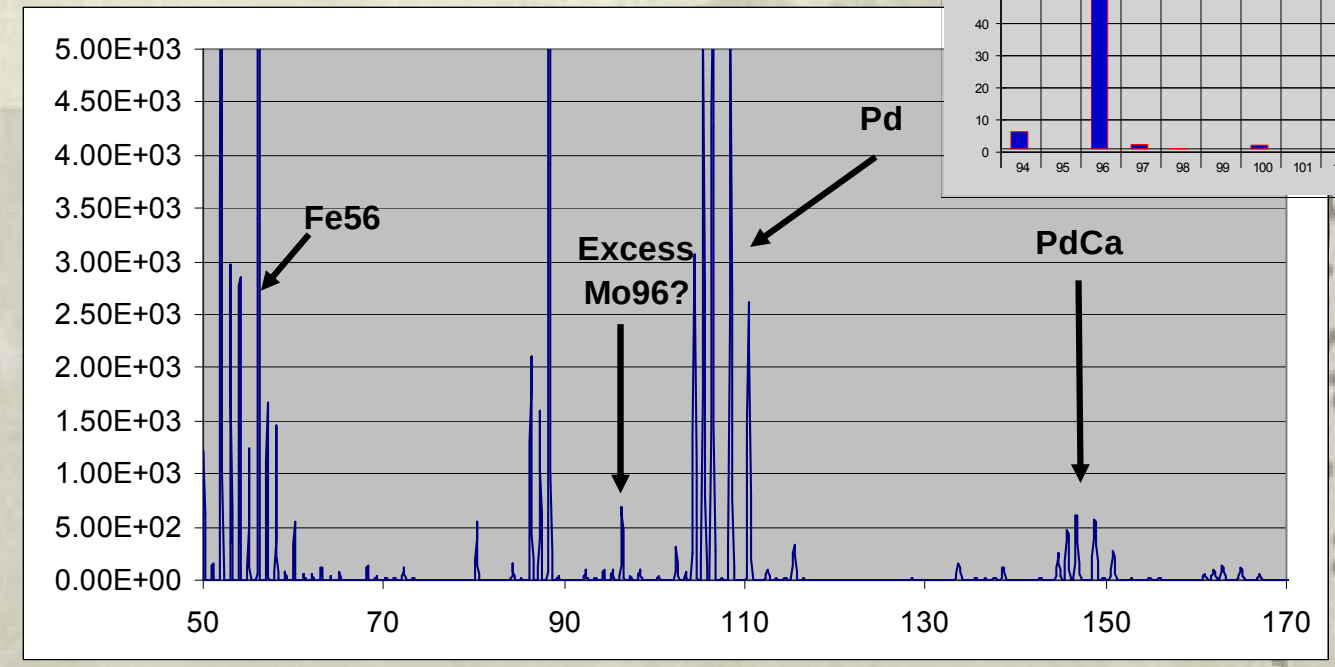
Be Wary of Cluster Ions
Secondary Ion Mass Spectrometry (SIMS)

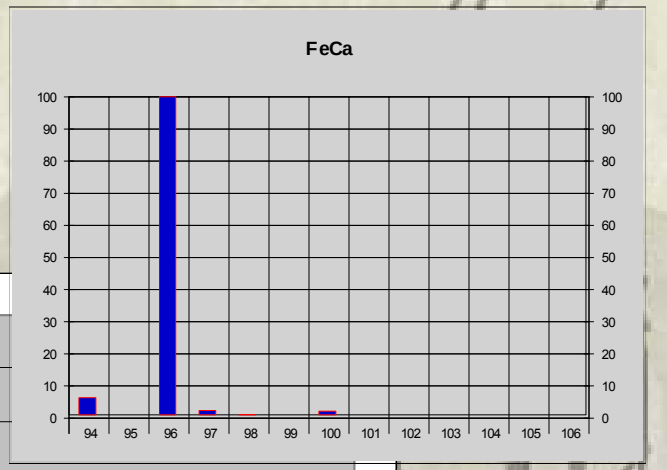
- Example: was ^{96}Mo due to a cluster ion?



Be Wary of Cluster Ions
Secondary Ion Mass Spectrometry (SIMS)

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

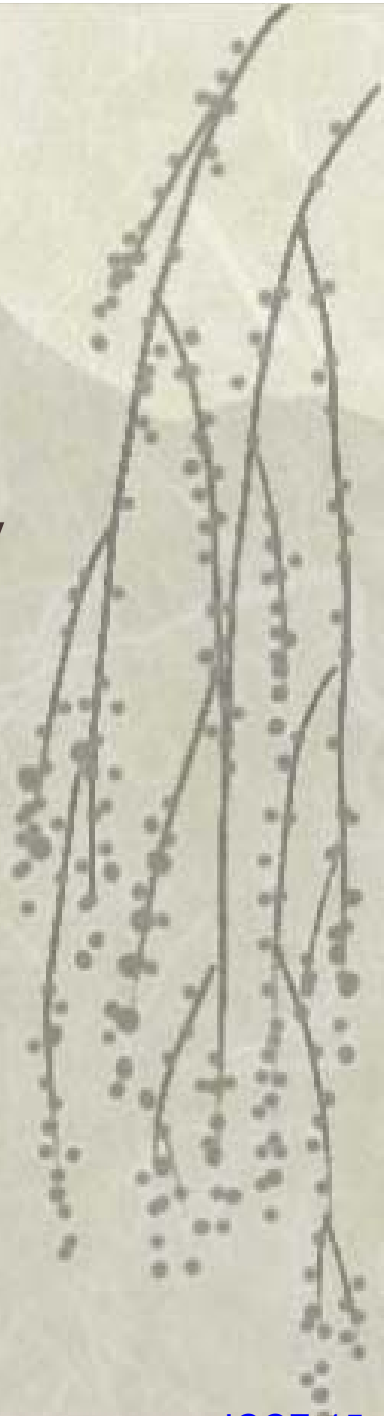




- Observe Pd+40 (likely Ca), why not ^{56}Fe+^{40}Ca ➔ mass 96

Conclusions

- The complex, multi-layer MHI structure was claimed to transmute elements
- Praseodymium is in the samples extracted by MHI at MHI
  - Analyzed by ICP-MS by NRL and a commercial laboratory with good agreement in results
  - Praseodymium in positive samples but not in controls
  - Praseodymium not in bulk palladium
  - Praseodymium signature not consistent with typical contamination from misch metal from the environment



Conclusions

- Praseodymium could NOT be extracted at NRL by NRL from split portions of the samples
- Environmental surveys at MHI by NRL and MHI found Praseodymium in key areas of laboratory
- ^{137}Ba and Thulium also found on positive samples and in laboratory at MHI
  - Not on historic samples prior to this study
  - Both handled previously to this study
  - Found in key areas of laboratory

- Presence of Praseodymium may have other explanations than transmutation of Cs

"We are to admit no more causes of natural things than such as are both true and sufficient to explain their appearances."
- Isaac Newton

Acknowledgements

Funding from DARPA is gratefully acknowledged. The insights provided by Graham Hubler, Michael Melich, Rodney Johnson, Alexander Ehrlich, and David Knies were very helpful. This work could not have been accomplished without the gracious help of Yasuhiro Iwamura, Takehiko Itoh, and Mitsuru Sakano.