Observation of Nuclear Transmutation Reactions induced by D_2 Gas Permeation through Pd Complexes

Yasuhiro Iwamura[1], Takehiko Itoh[1], Mitsuru Sakano[1], Noriko Yamazaki[1], Shizuma Kuribayashi[1], Yasuko Terada[2] and Testuya Ishikawa[3] and Jirohta Kasagi[4]

[1]Advanced Technology Research Center, Mitsubishi Heavy Industries, Ltd.
[2]Japan Synchrotron Radiation Research Institute
[3]Coherent X-ray Optics Laboratory, SPring-8/RIKEN
[4]Laboratory for Nuclear Science, Tohoku University

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

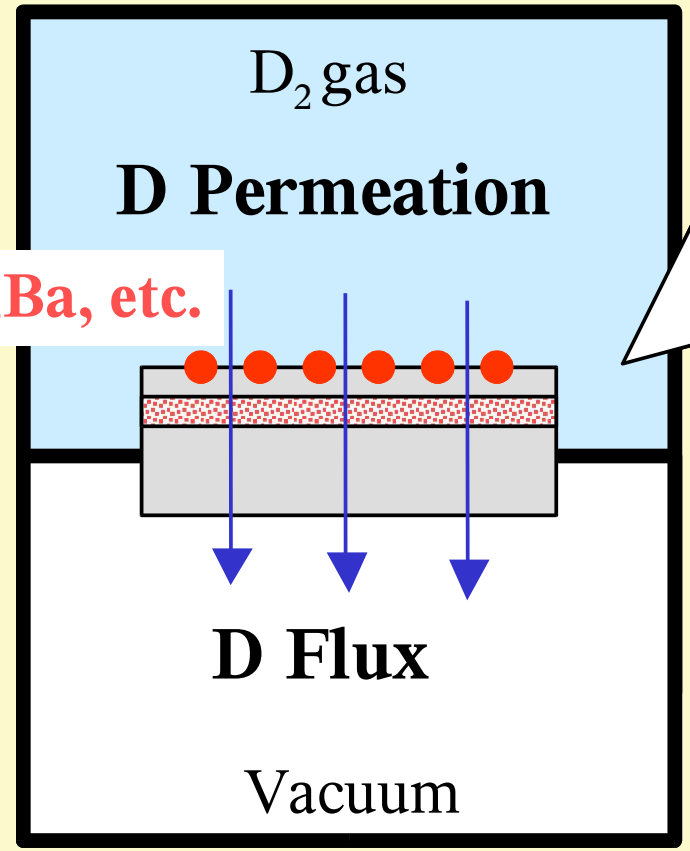
ICCF-11, Marseilles, France, October31-November5, 2004

Contents

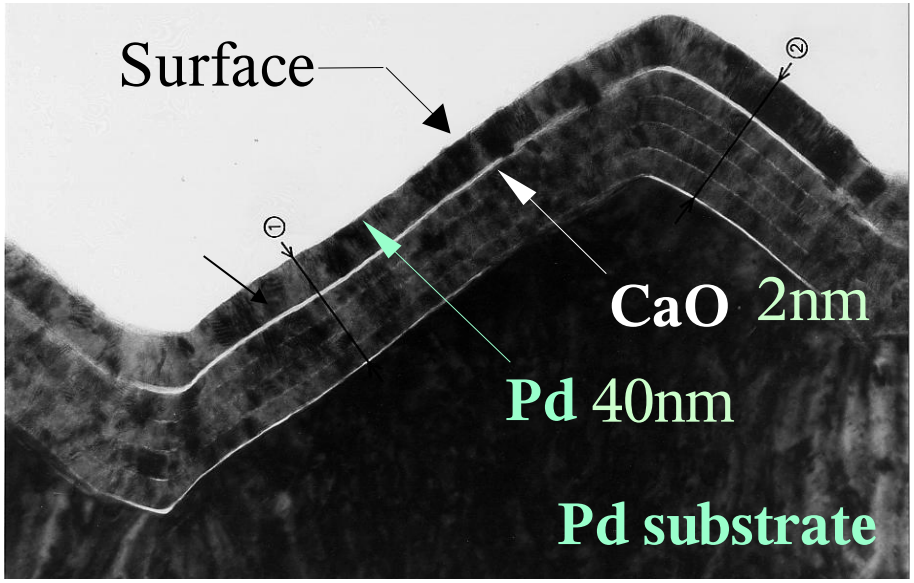
Features of the Present Method

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

D_2 gas permeation through the Pd complex



Cross Section of Pd Complex



Fabrication of Pd Complex

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

Washing a Palladium Sample with Acetone

↓

900° C 10H Annealing under Vacuum Condition (< 10^{-7} Torr)

↓

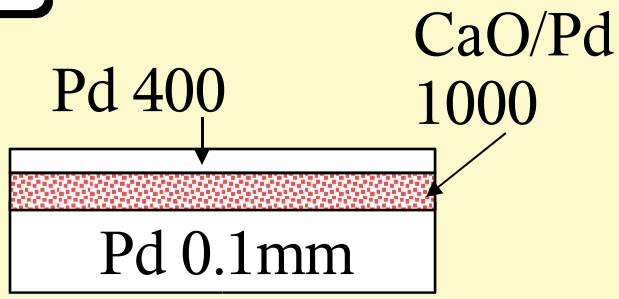
Washing the Sample with Aqua Regia (100sec)

↓

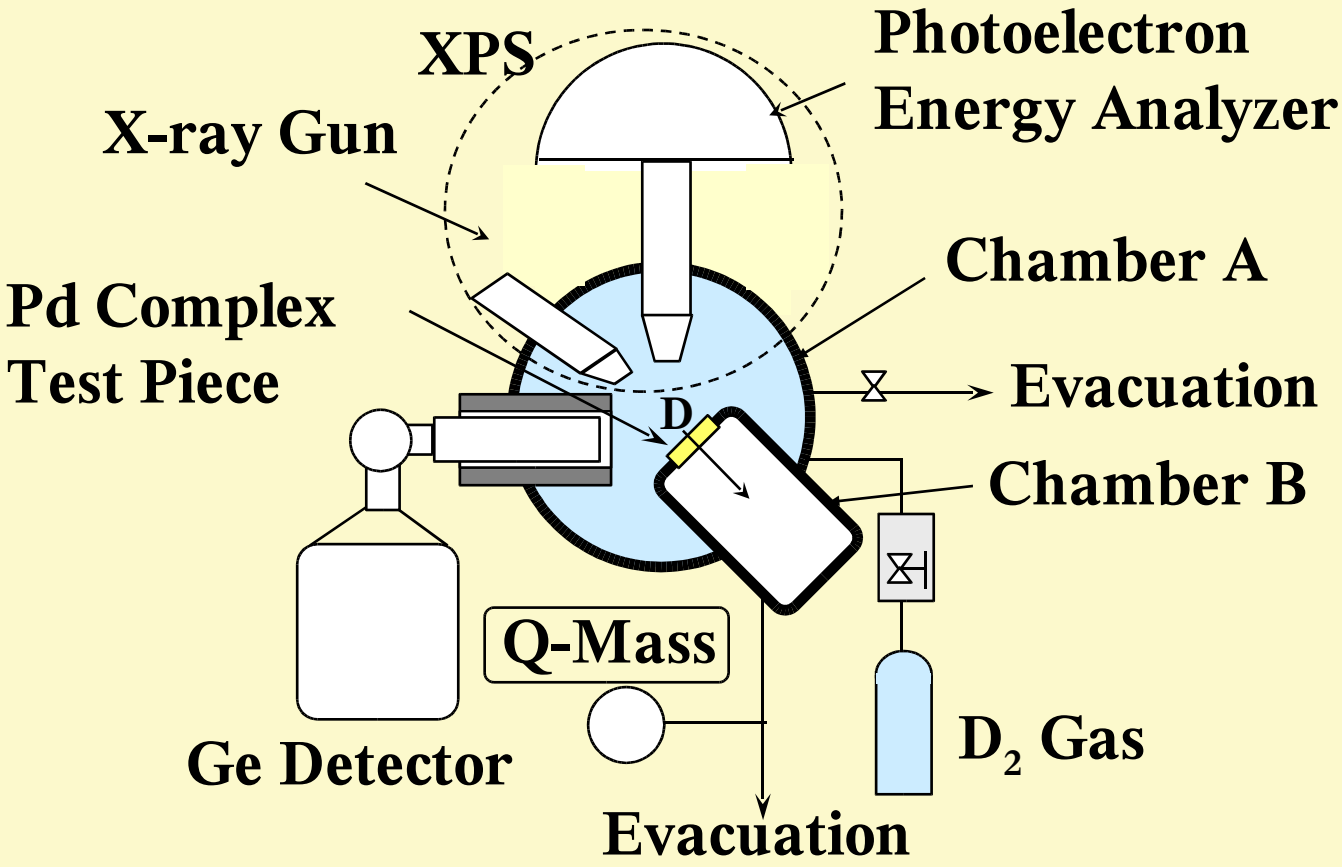
5 times Alternatingly Sputtering of CaO (20) and Pd (180)

↓

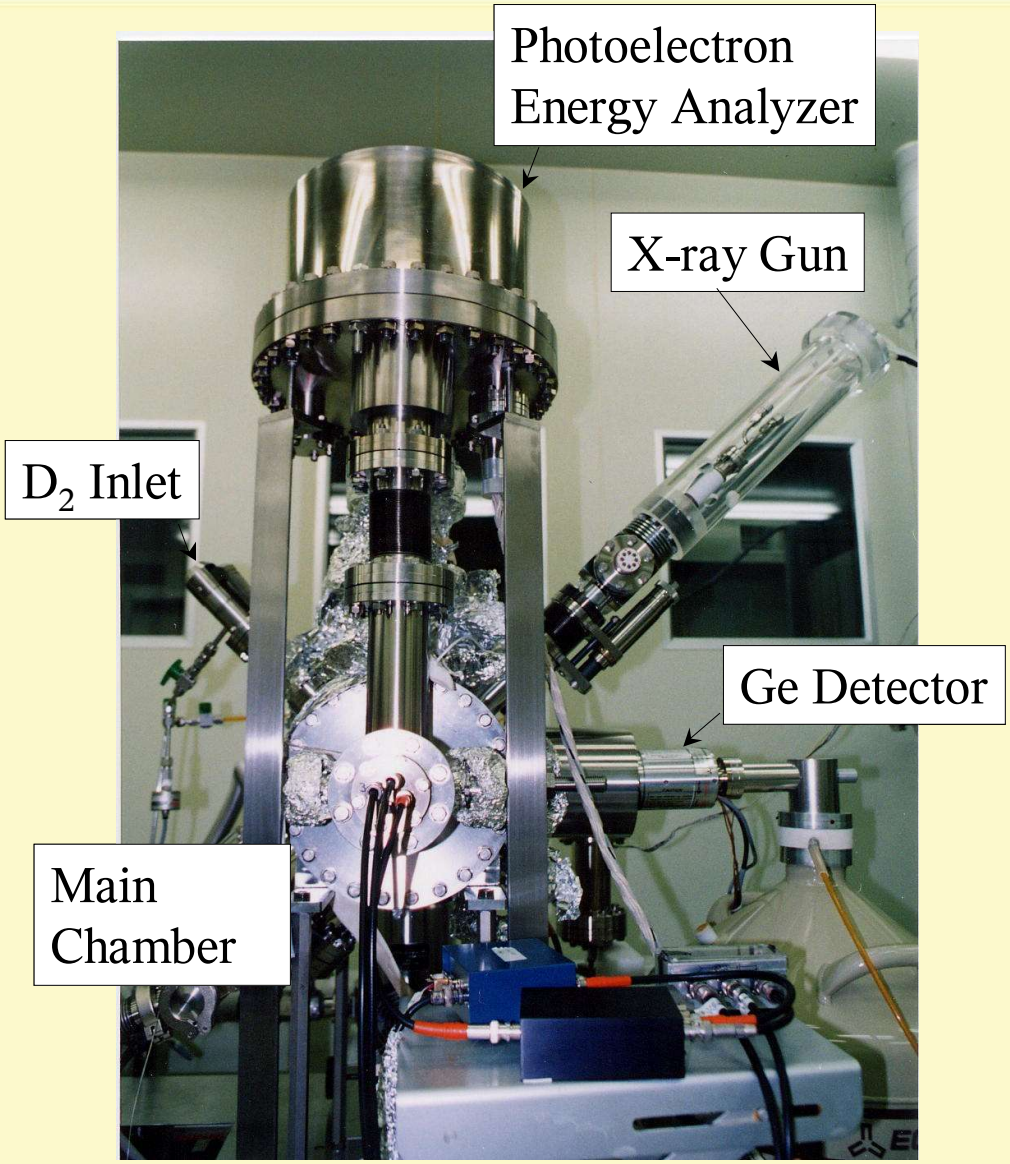
Ion Beam Sputtering of Pd only (400)



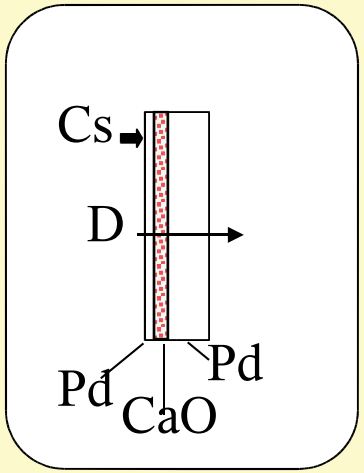
Schematic View of the Experimental Apparatus

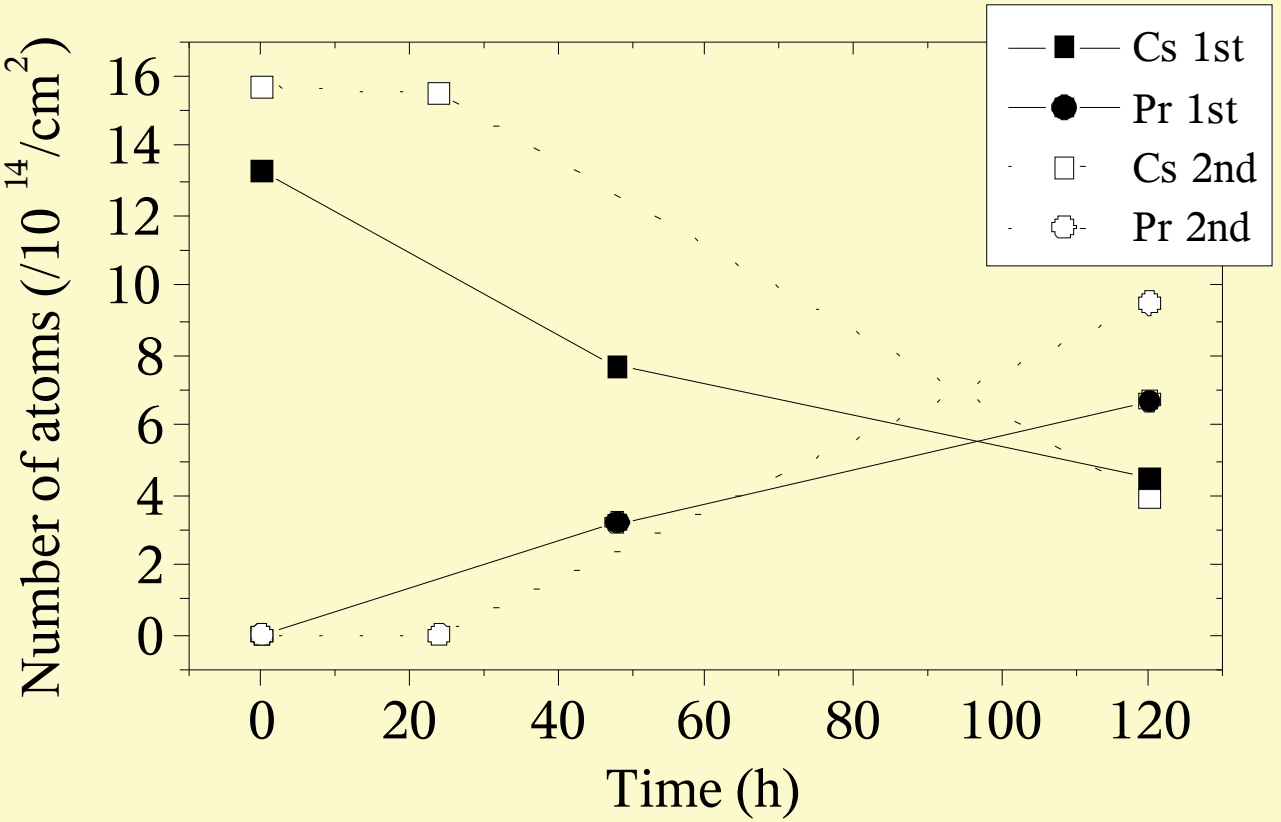


Photograph of the Experimental Setup

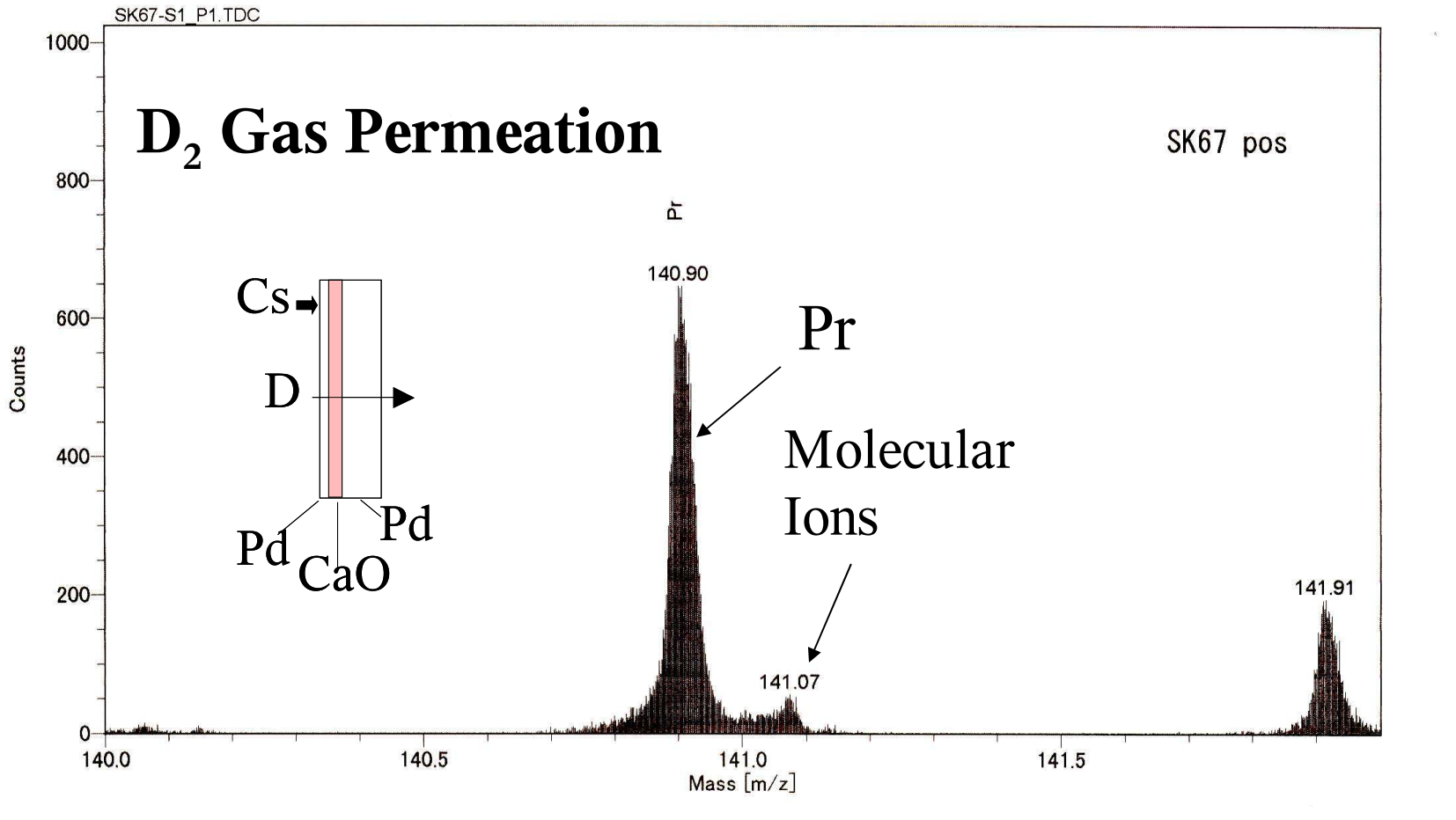


Decrease of Cs and Emergence of Pr



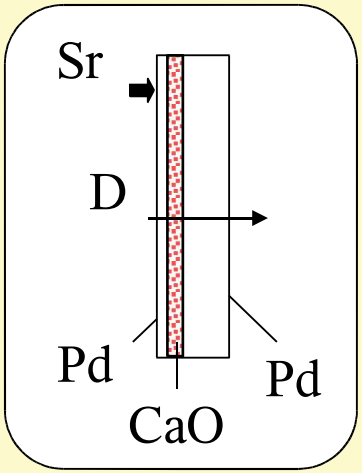


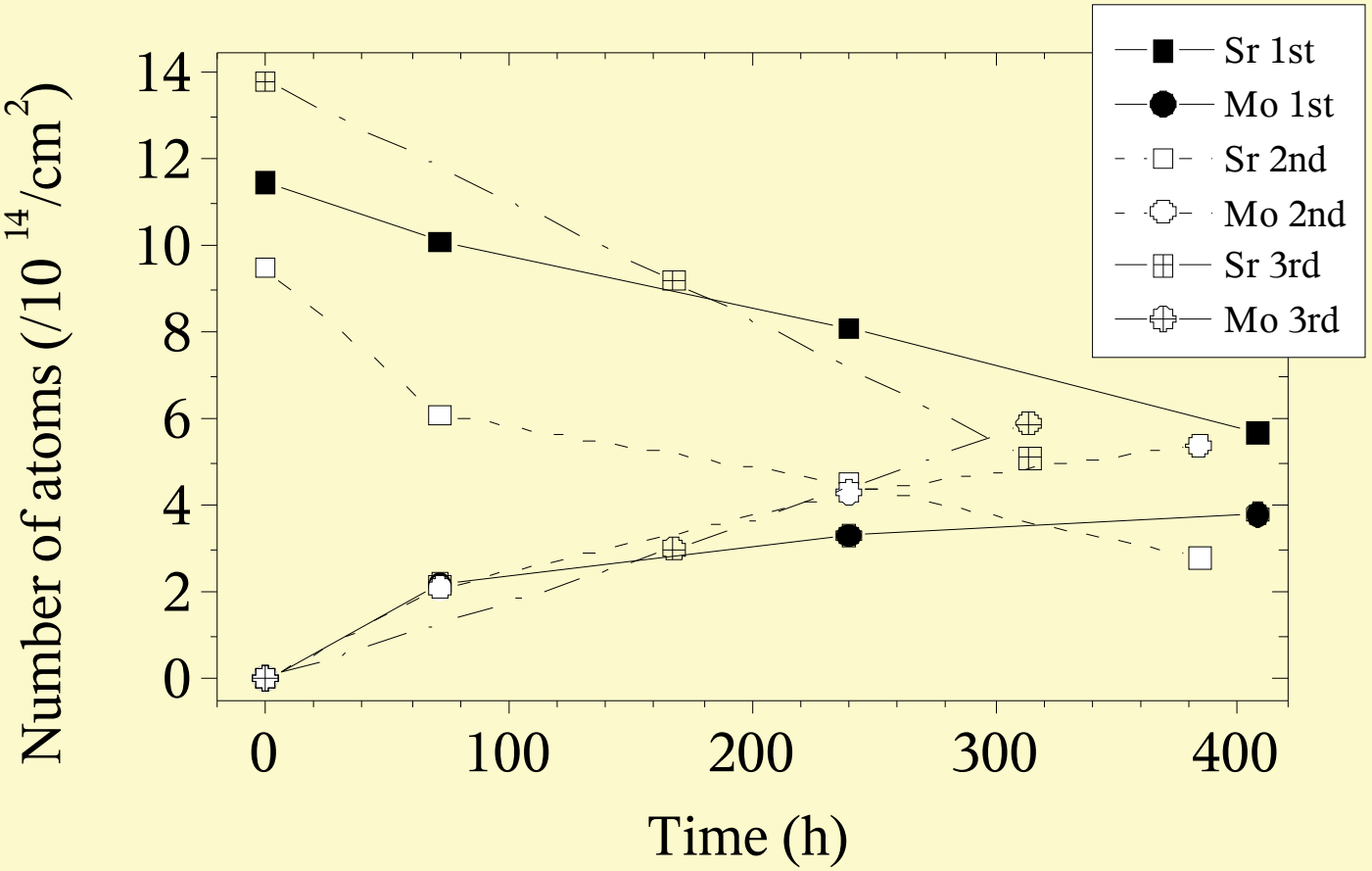
Identification of Pr by TOF-SIMS



TOF-SIMS device (TRIFT™ ;ULVAC-PHI)

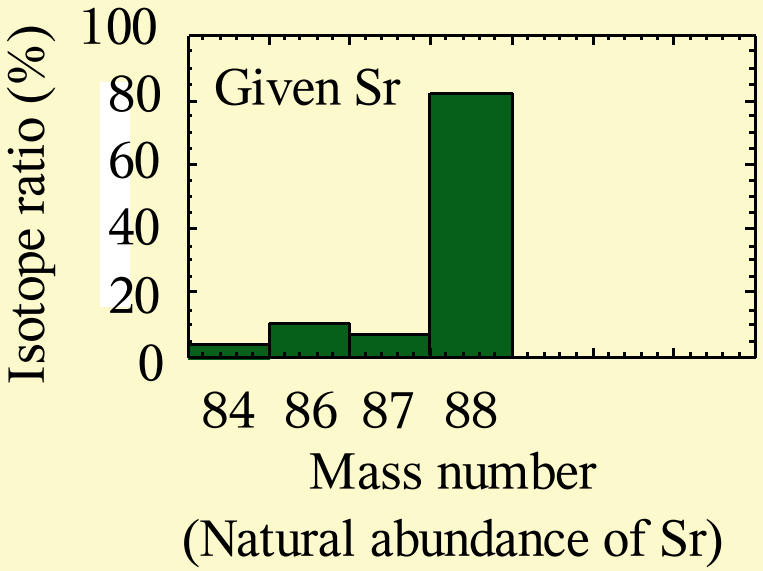
Decrease of Sr and Emergence of Mo

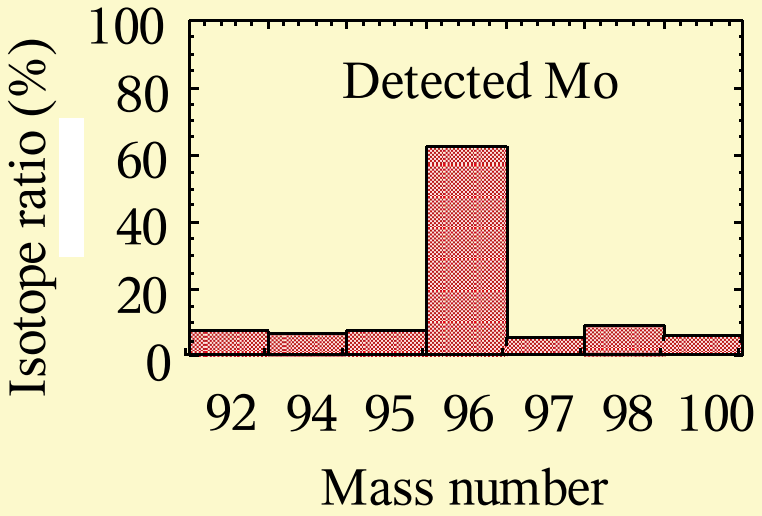




Relation of Isotopic Composition between Sr and Mo

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER





Mass N. +8

$^{88}_{38}Sr \rightarrow ^{96}_{42}Mo$

Atomic N. +4

Mass N. +8

$^{133}_{55}Cs \rightarrow ^{141}_{59}Pr$

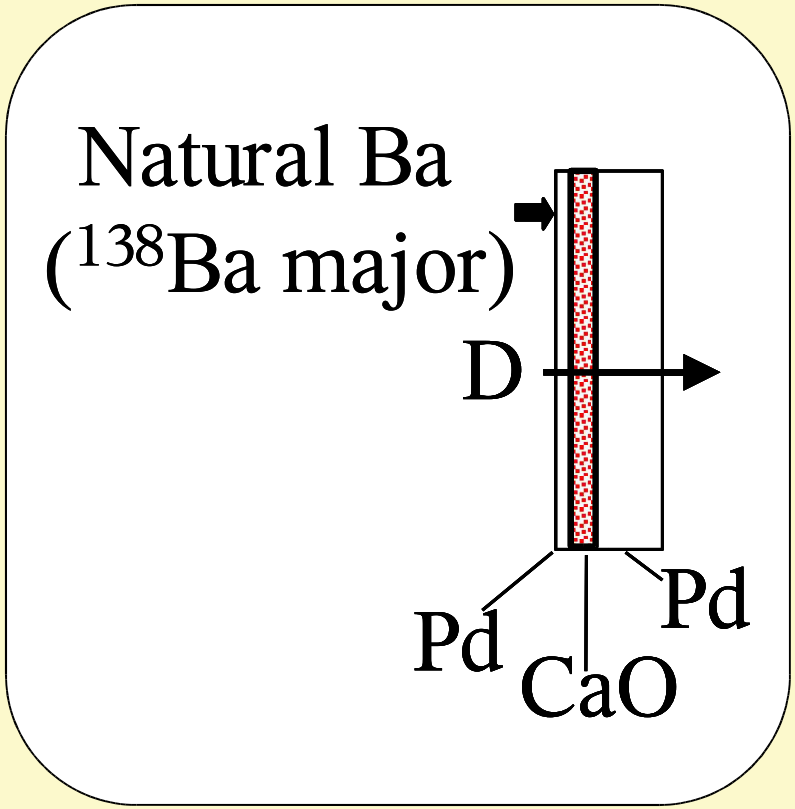
Atomic N. +4

Recent Results Part 1

Transmutation of ^{138}Ba into ^{150}Sm and ^{137}Ba into ^{149}Sm

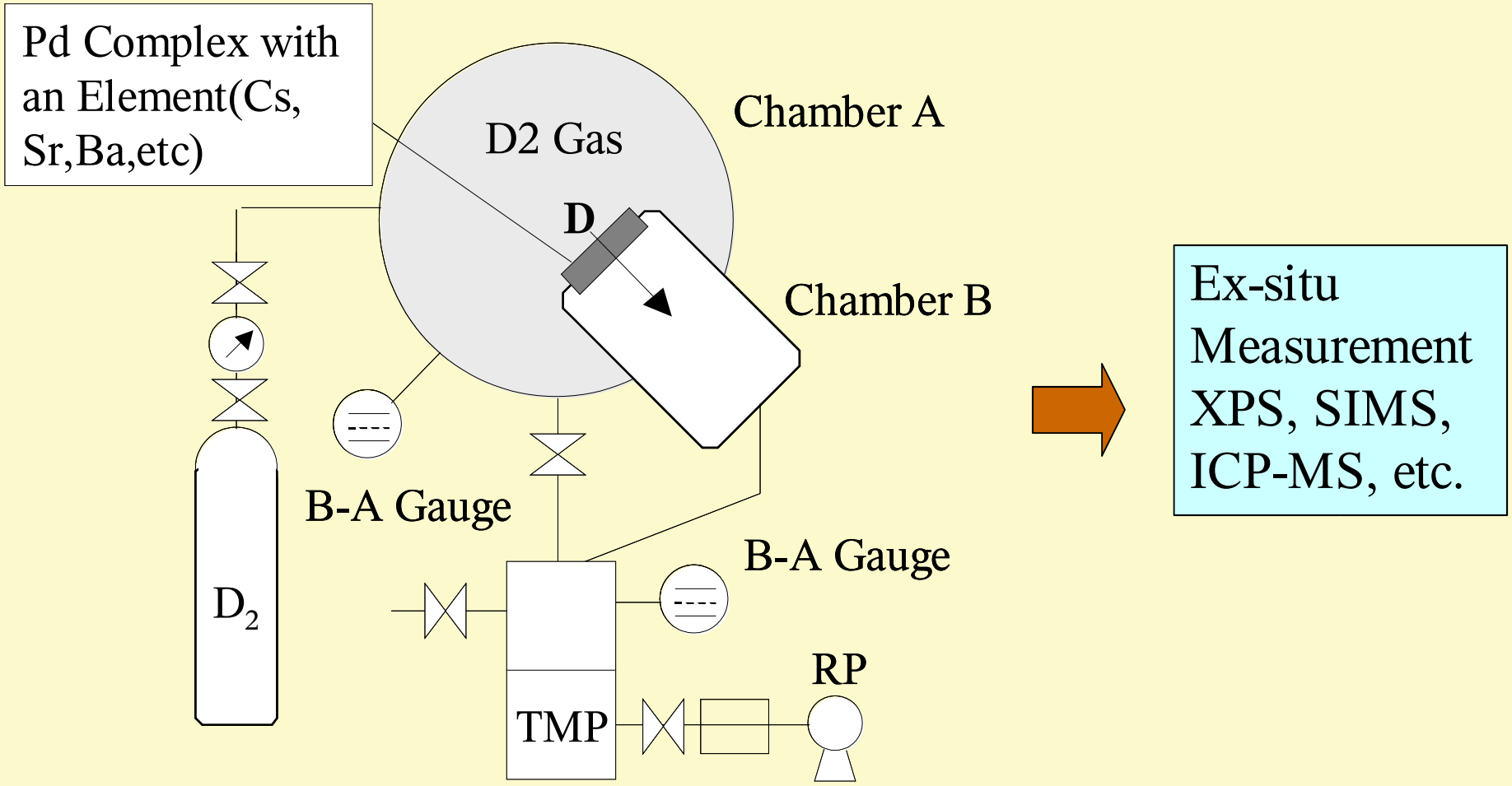
Transmutation of Ba into Sm; Natural Ba

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

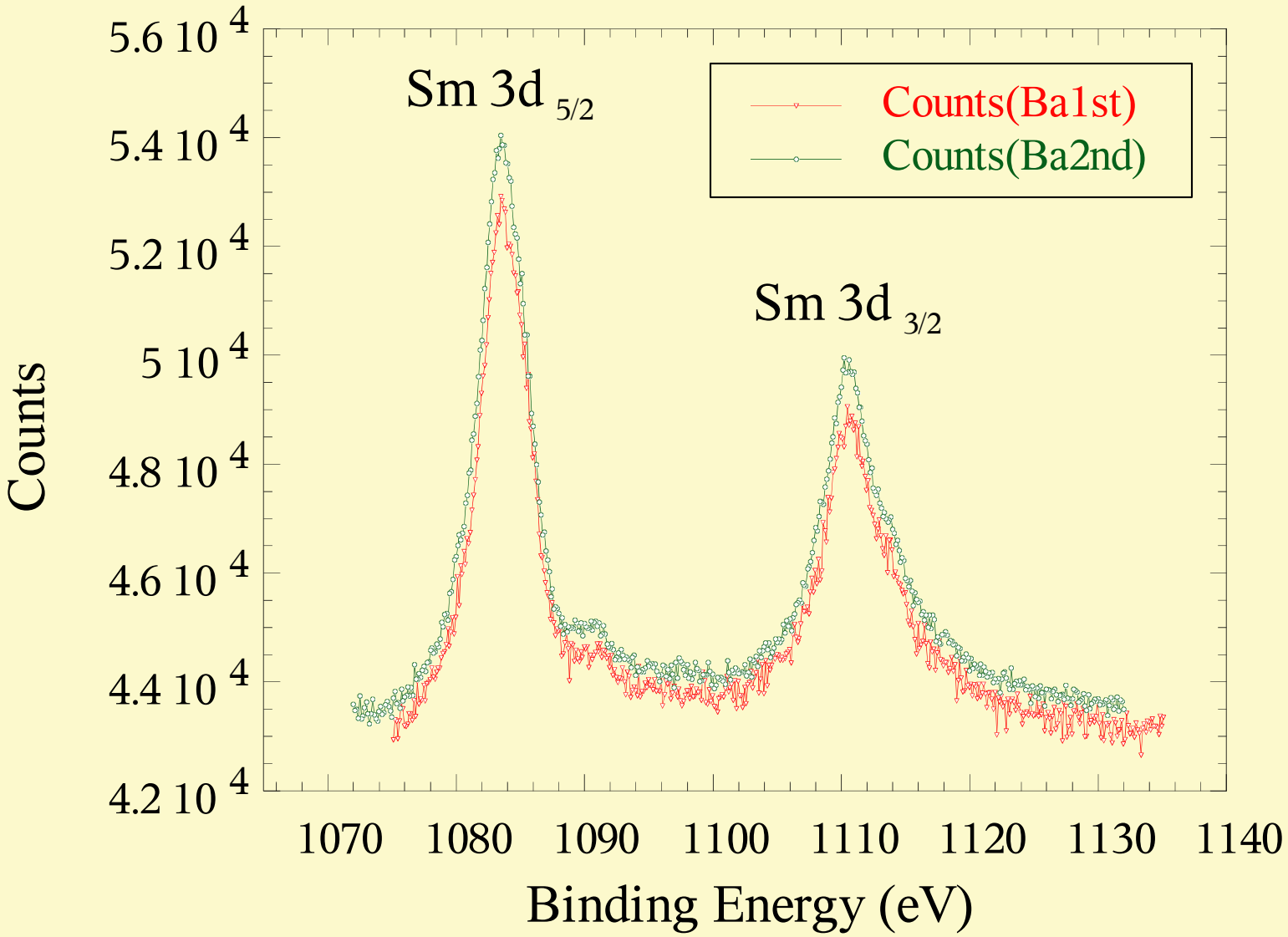


^{150}Sm was detected after D permeation on the Pd complex

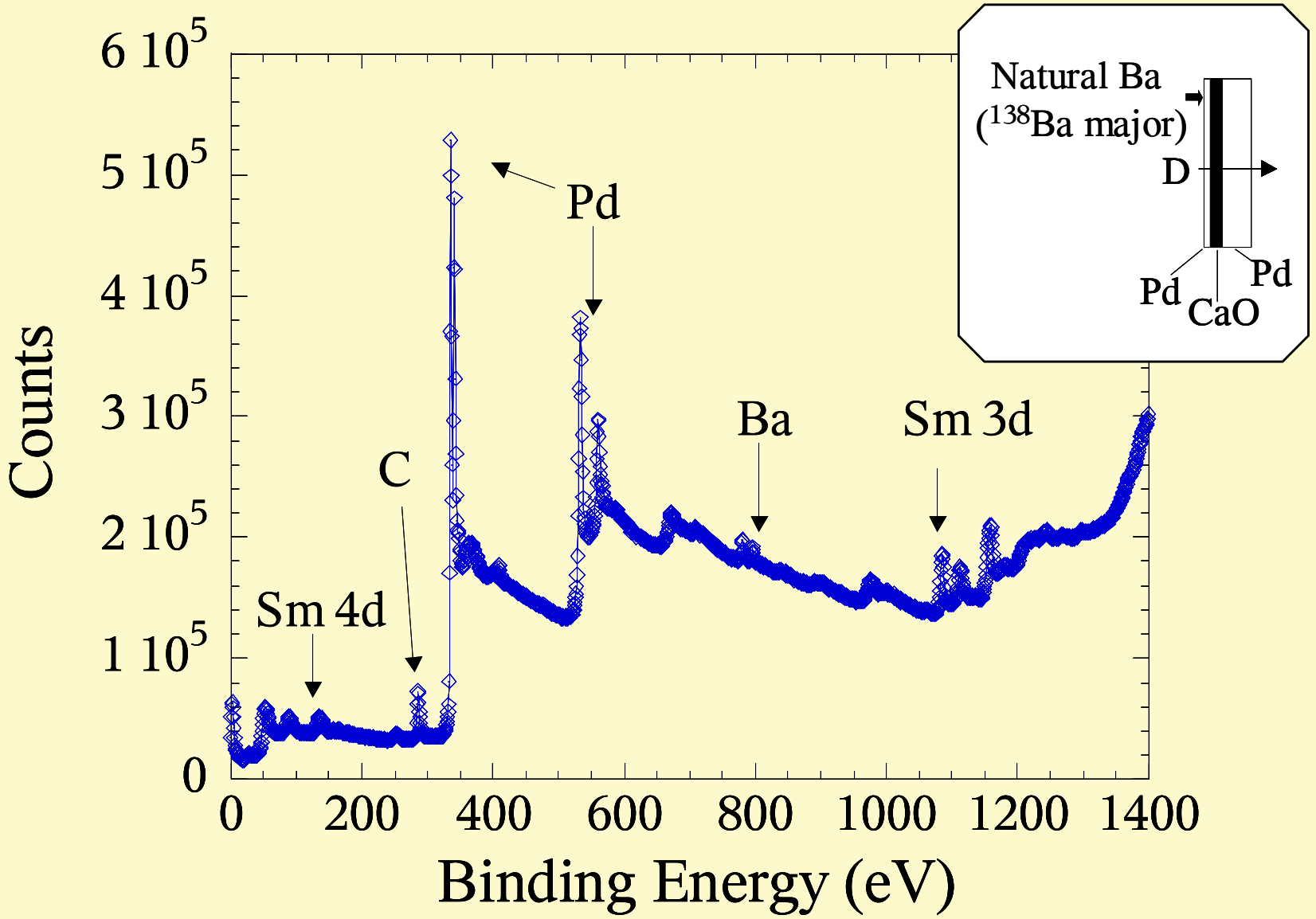
Schematic View of the Ex-situ Measurement Apparatus



XPS Spectra for detected Sm

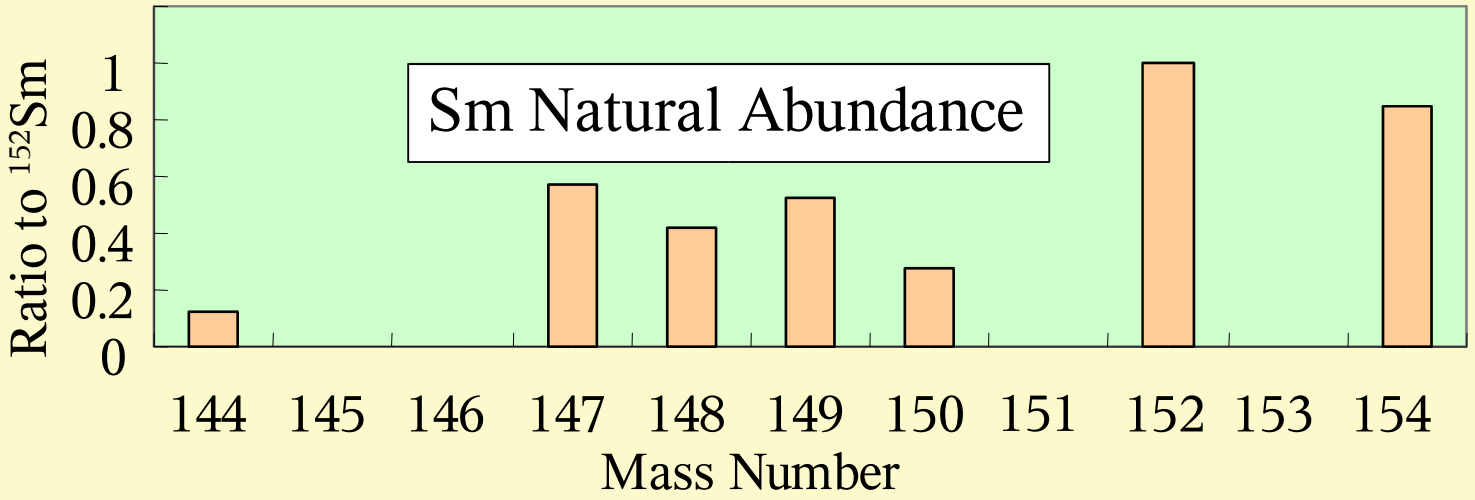


Full Spectrum



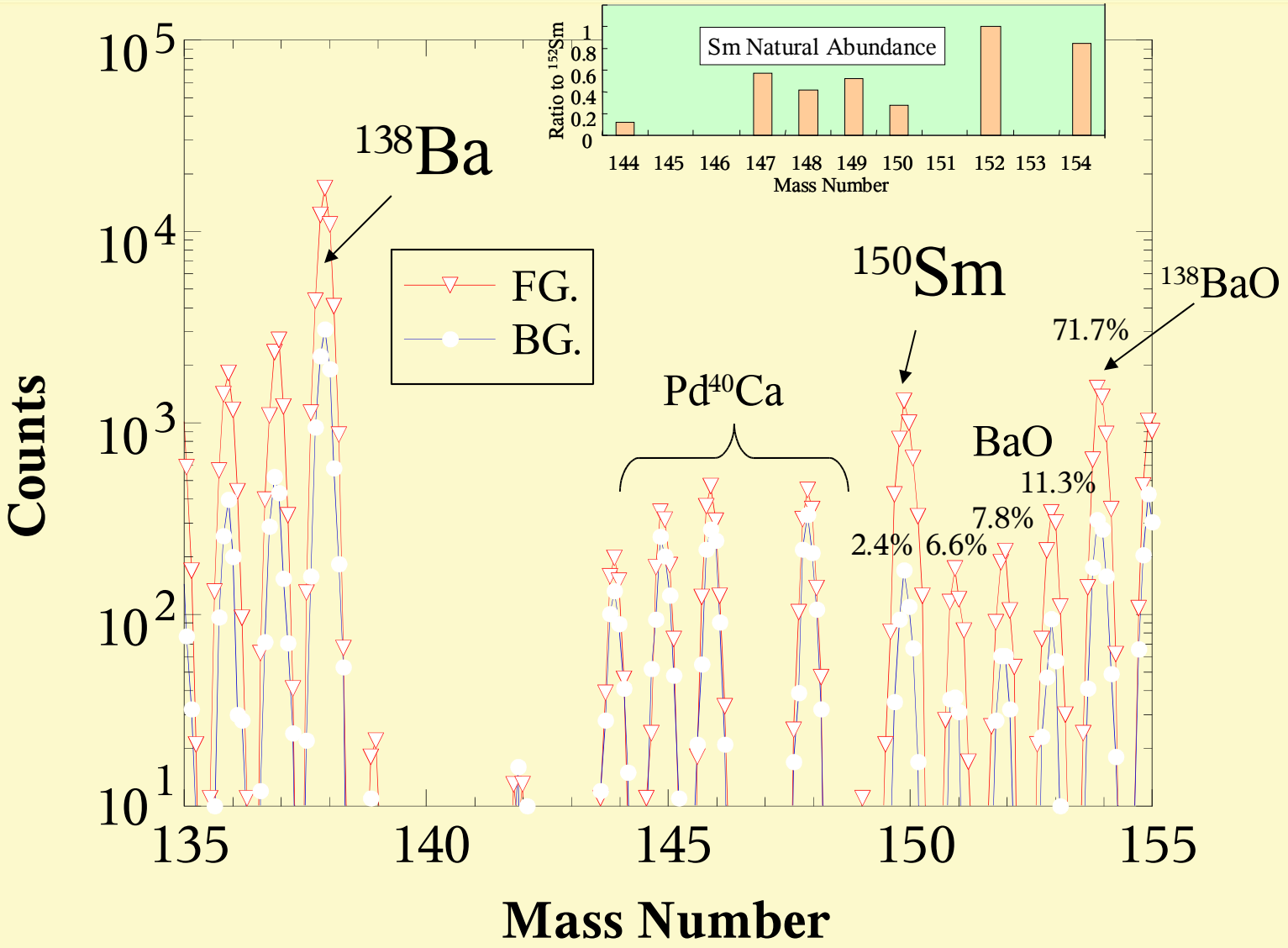
Sm Natural Abundance

^{144}Sm	^{147}Sm	^{148}Sm	^{149}Sm	^{150}Sm	^{152}Sm	^{154}Sm
3.2%	15.1 %	11.3 %	13.8 %	7.5%	26.6 %	22.5 %



SIMS Spectra for Given and Detected Elements

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



Examination of Molecular Ions

Pd	$Pd^{40}Ca$
102(1%)	142
104 (11%)	144
105 (22%)	145
106 (27%)	146
108 (26%)	148
110 (12%)	150

Ba	$Ba^{16}O$
130(0.1%)	146
132(0.1%)	148
134(2.4%)	150
135(6.6%)	151
136(7.8%)	152
137 (11.3%)	153
138(71.7%)	154

No Molecular Ions for 149.

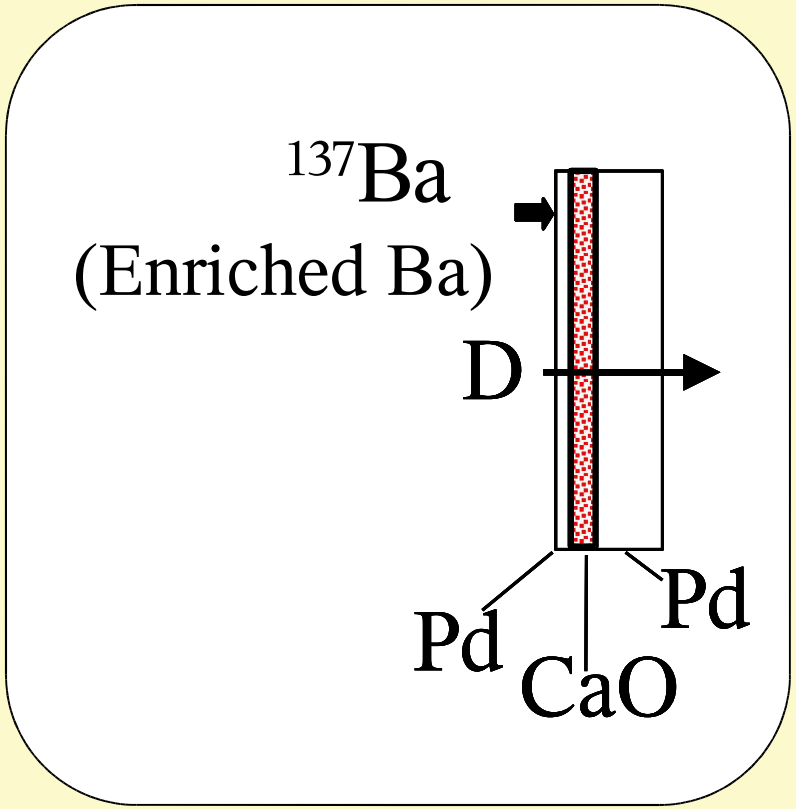
$^{110}Pd(12\%)Ca$ and $^{134}Ba(2.4\%)O$ for mass150, however their effects should be lower than $^{106}Pd(27\%)Ca$ and $^{138}Ba(71.7\%)O$

Transmutation of Natural Ba into Sm

- XPS analysis showed Sm signal.
- SIMS analysis showed the increase of mass 150.
- Natural Sm isotopic distribution did not match with SIMS mass data.
- These facts strongly suggests that ^{150}Sm exists on the Pd complex after D_2 gas permeation.

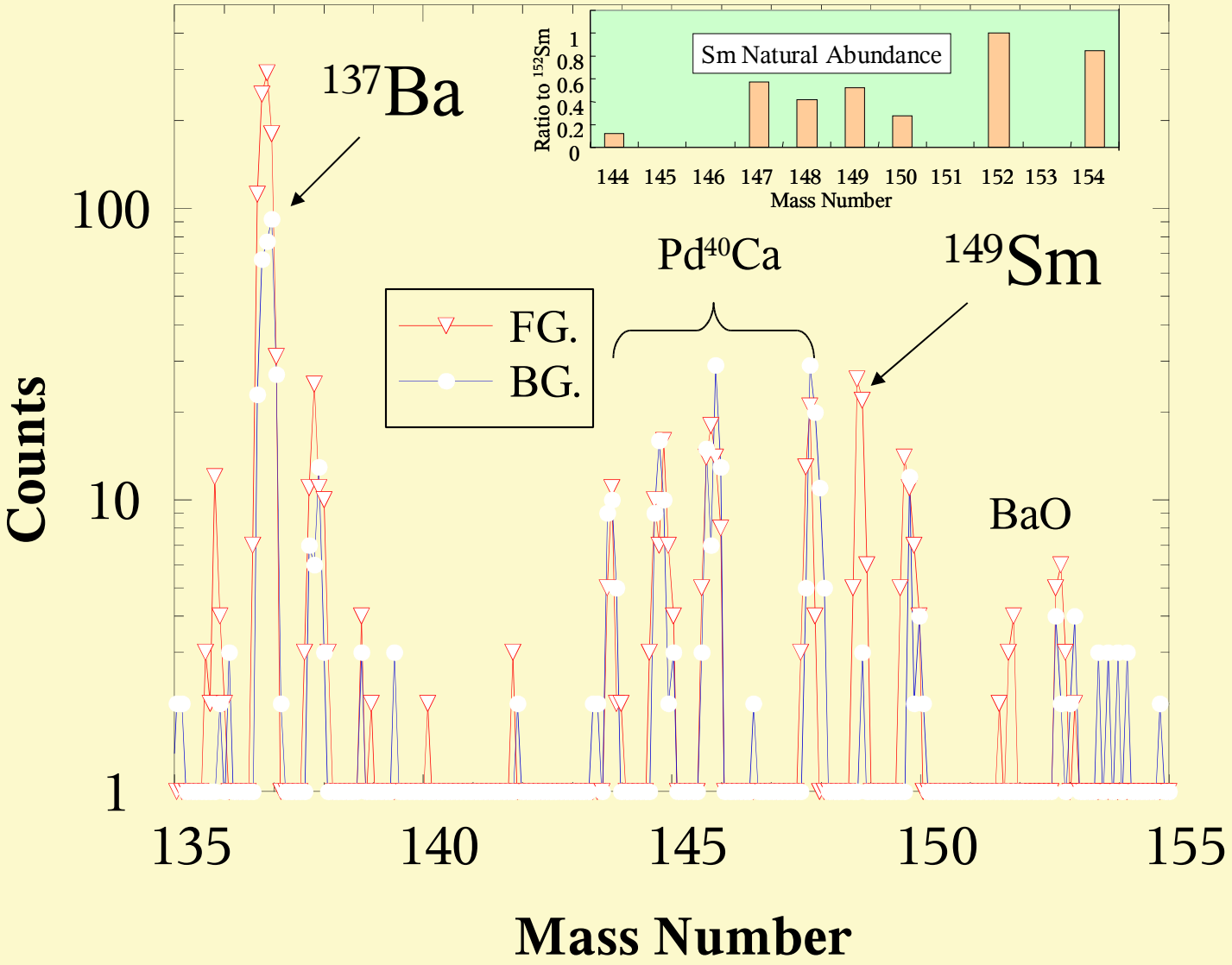
Transmutation of Ba into Sm; mass 137 Enriched Ba

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



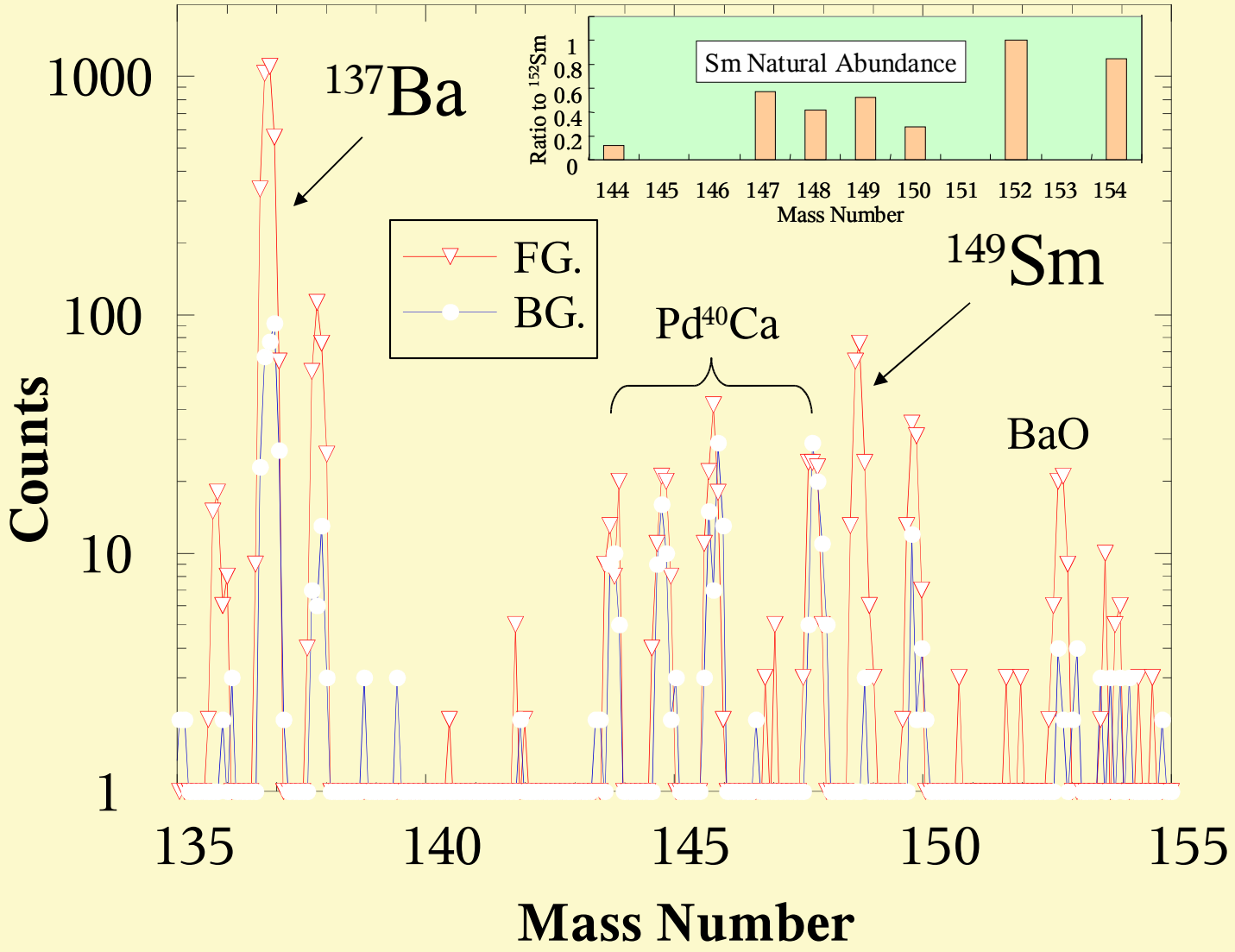
^{149}Sm may have been detected after D permeation on the Pd complex

SIMS Spectra for #1Experiment



SIMS Spectra for #2 Experiment

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

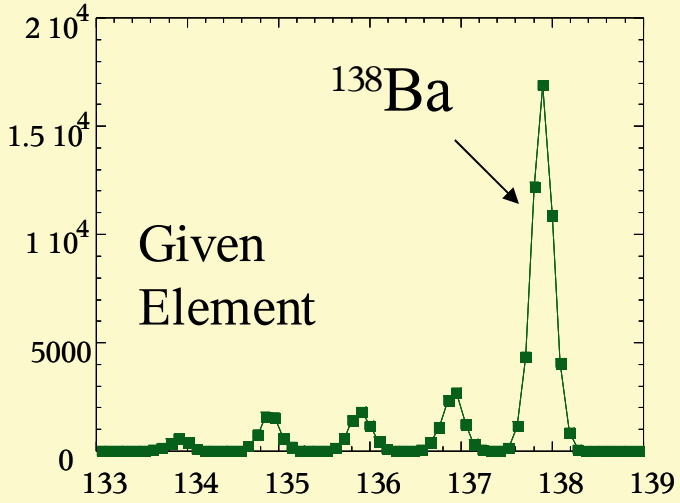


Transmutation of ^{137}Ba into Sm

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

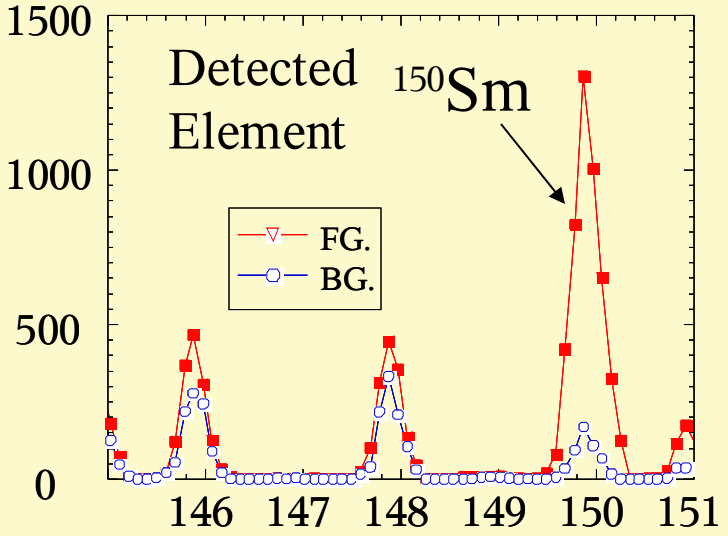
- SIMS analysis showed the increase of mass 149.
- Natural Sm isotopic distribution did not match with SIMS mass data.
- XPS analysis showed very weak Sm spectra. Now we are trying to obtain clear XPS signals.
- These facts suggests that ^{149}Sm exists on the Pd complex if we consider that Sm spectra were obtained by XPS using natural Ba.

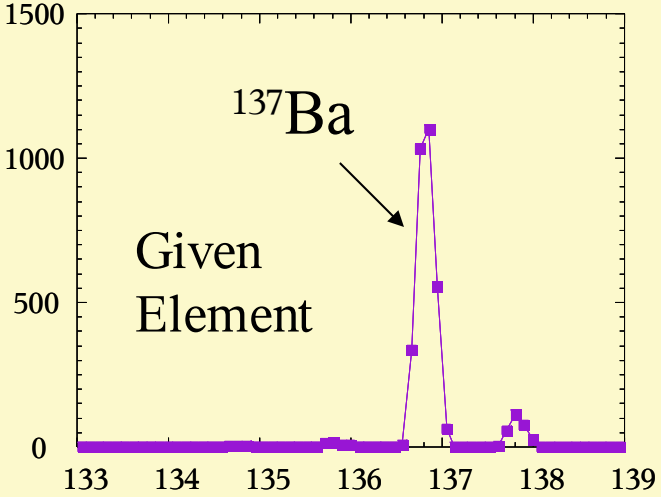
Mass Correlation between Given and Detected Elements



$^{138}_{56}Ba \xrightarrow{6d} ^{150}_{62}Sm$

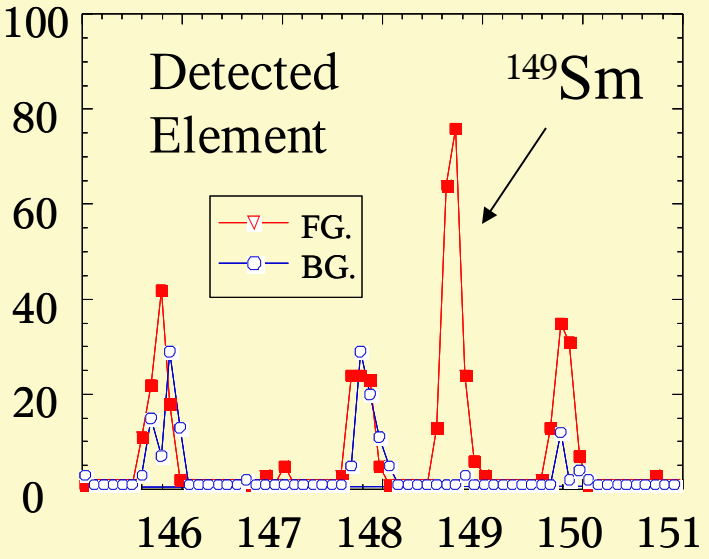
M +12
Z +6





$^{137}_{56}Ba \xrightarrow{6d} ^{149}_{62}Sm$

M +12
Z +6



The Aim of Ba Transmutation Experiments

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

$$^{137}_{56}Ba \xrightarrow{6d(3\alpha)} {}^{149}_{62}Sm^{*}$$

Experimental Results

^{149}Sm is a Mossbauer Isotope

Excitation Energy: 22.5keV

If we measure the Mossbauer effect of ^{149}Sm, we will obtain clear evidence of generation of ^{149}Sm.

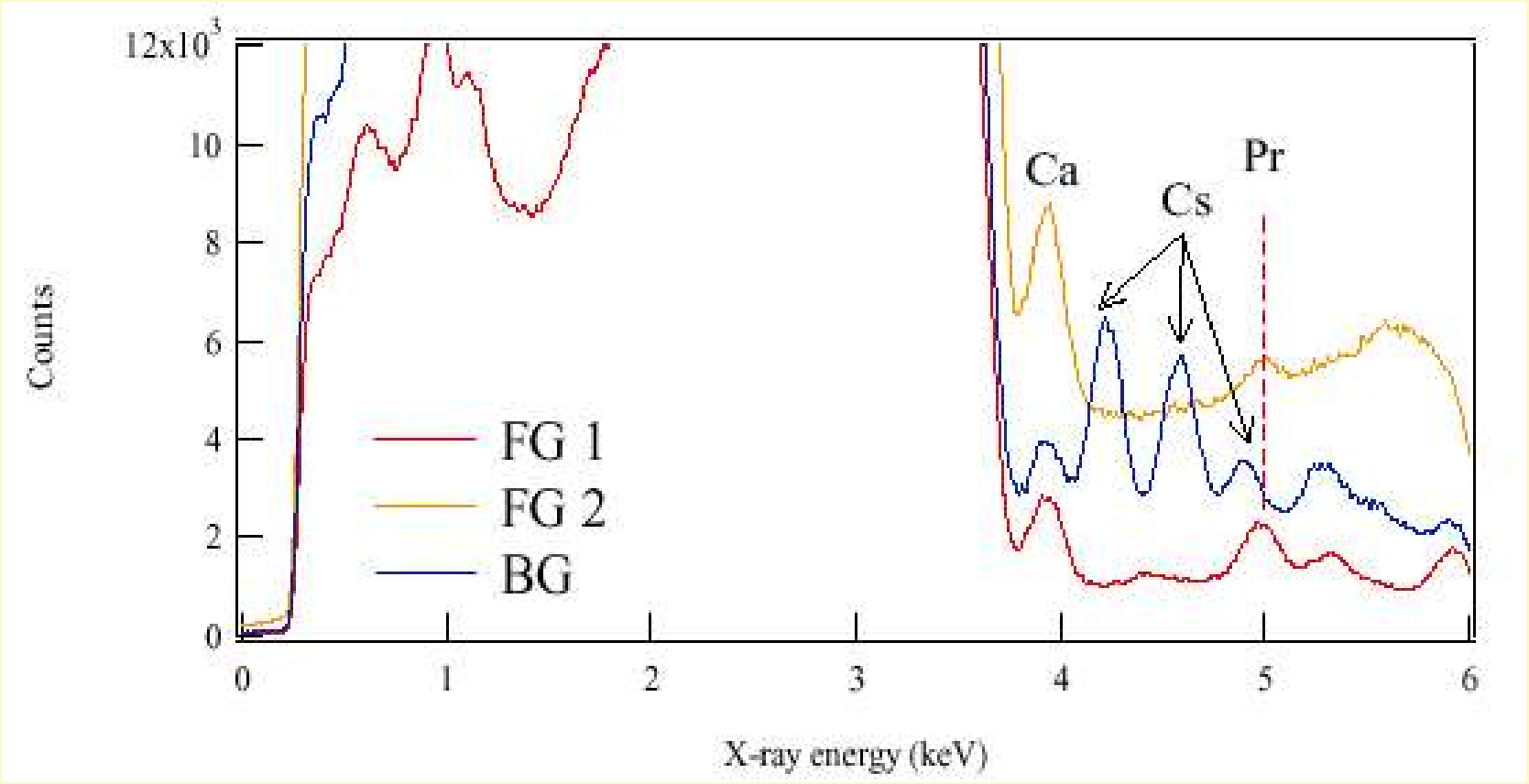
And the information on the ultra fine structure relating to the electronic state and phonon of the generated ^{149}Sm will be obtained.

Recent Results Part 2

Pr Confirmation by XRF and Experiments for in-situ Measurement at SPring-8

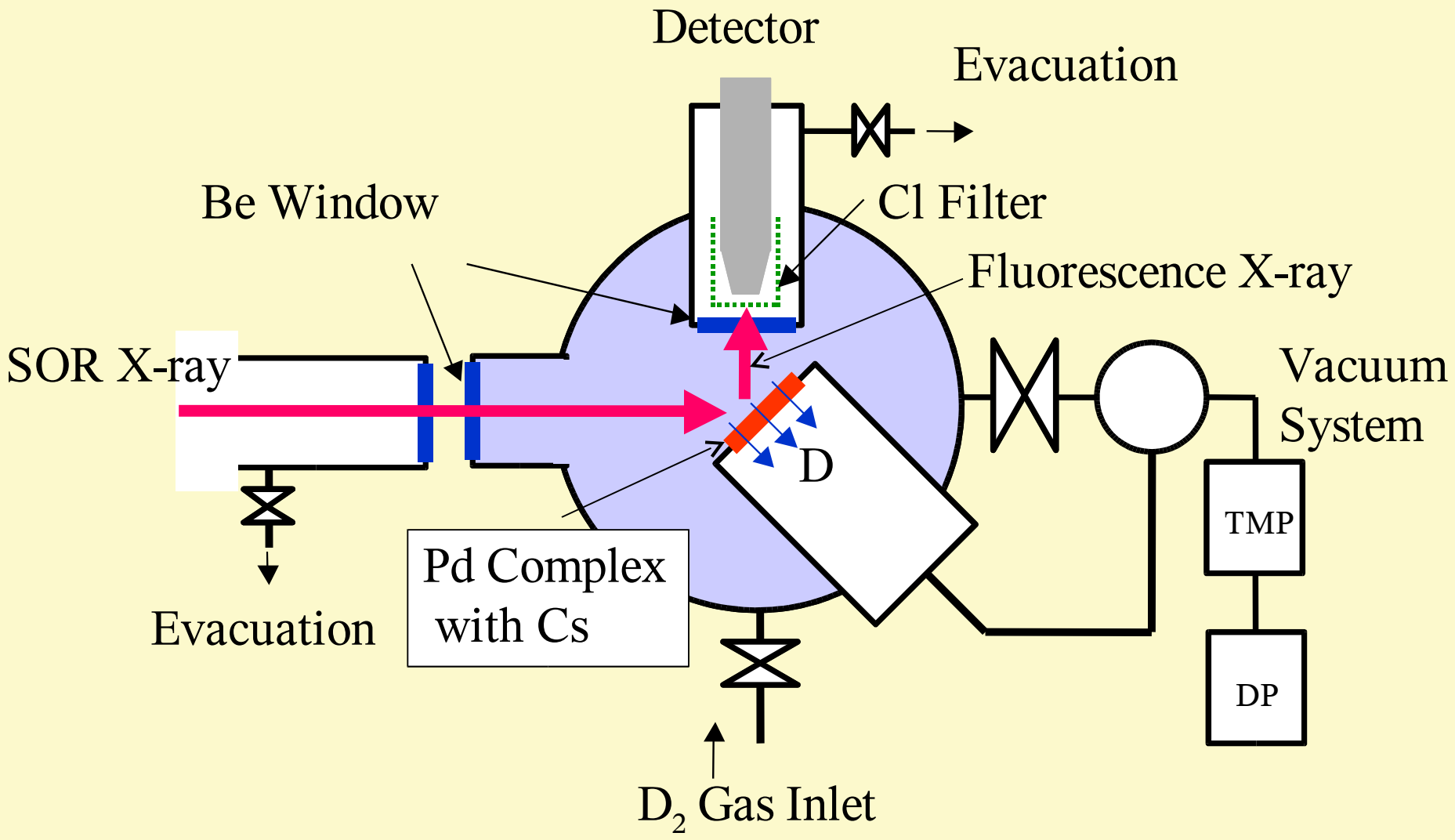
Identification of Pr by X-ray Fluorescence

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



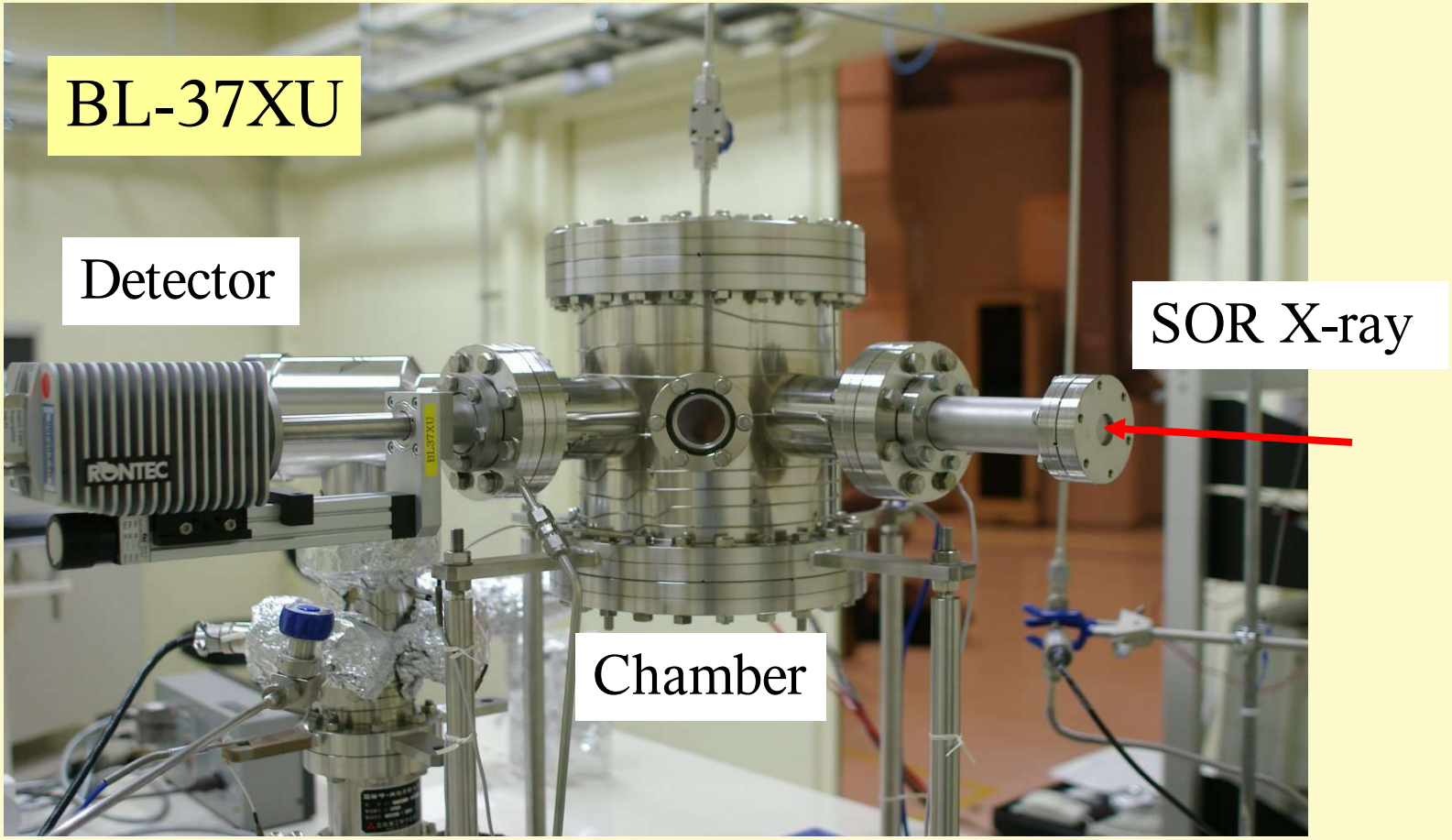
Detection of Pr using SOR X-ray at Spring-8, Harima, Japan
(FG1, FG2: Signals from Samples after D2 Permeation
BG: Signals from the sample before Permeation)

Experimental Set-up for *in-situ* Measurement located at SPring-8



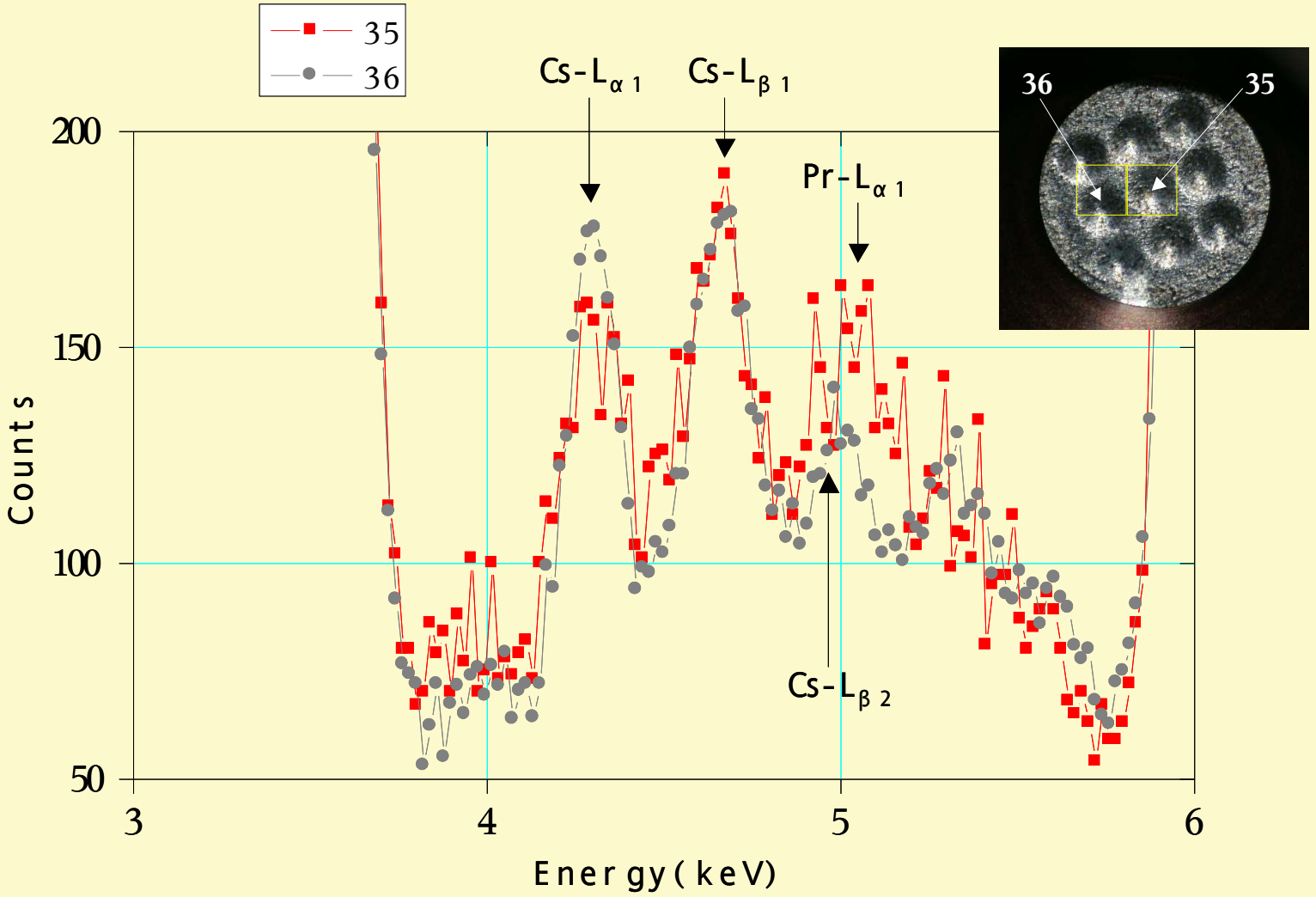
Photograph of the Experimental Set-up

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



An Example of Pr Detection by the Experiments at SPring-8

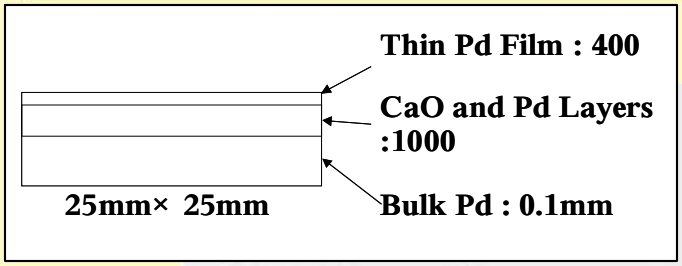
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



Recent Results Part 3

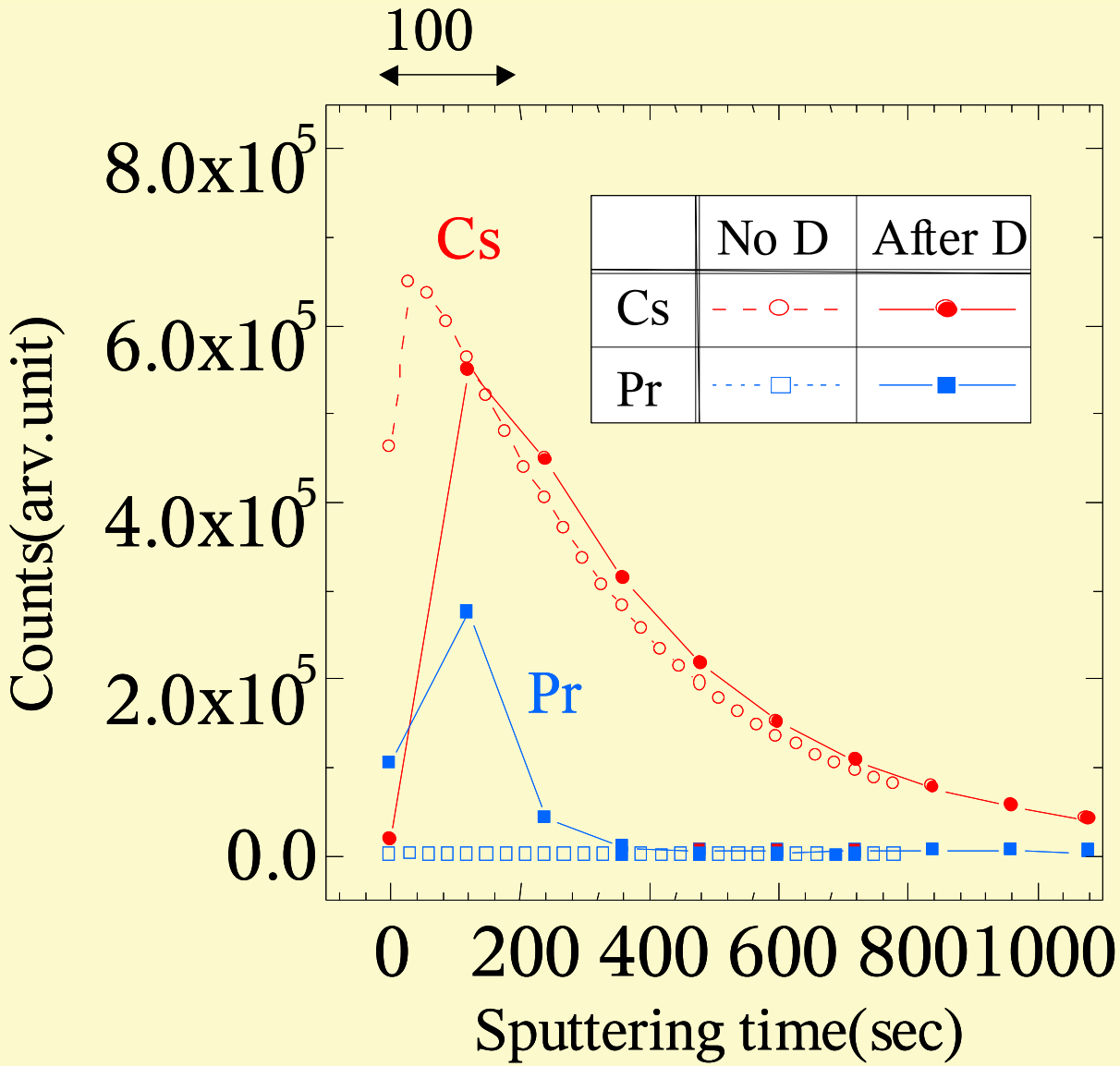
Measurement and Experiments relating to the role of CaO

TEM Photograph of the Pd Complex

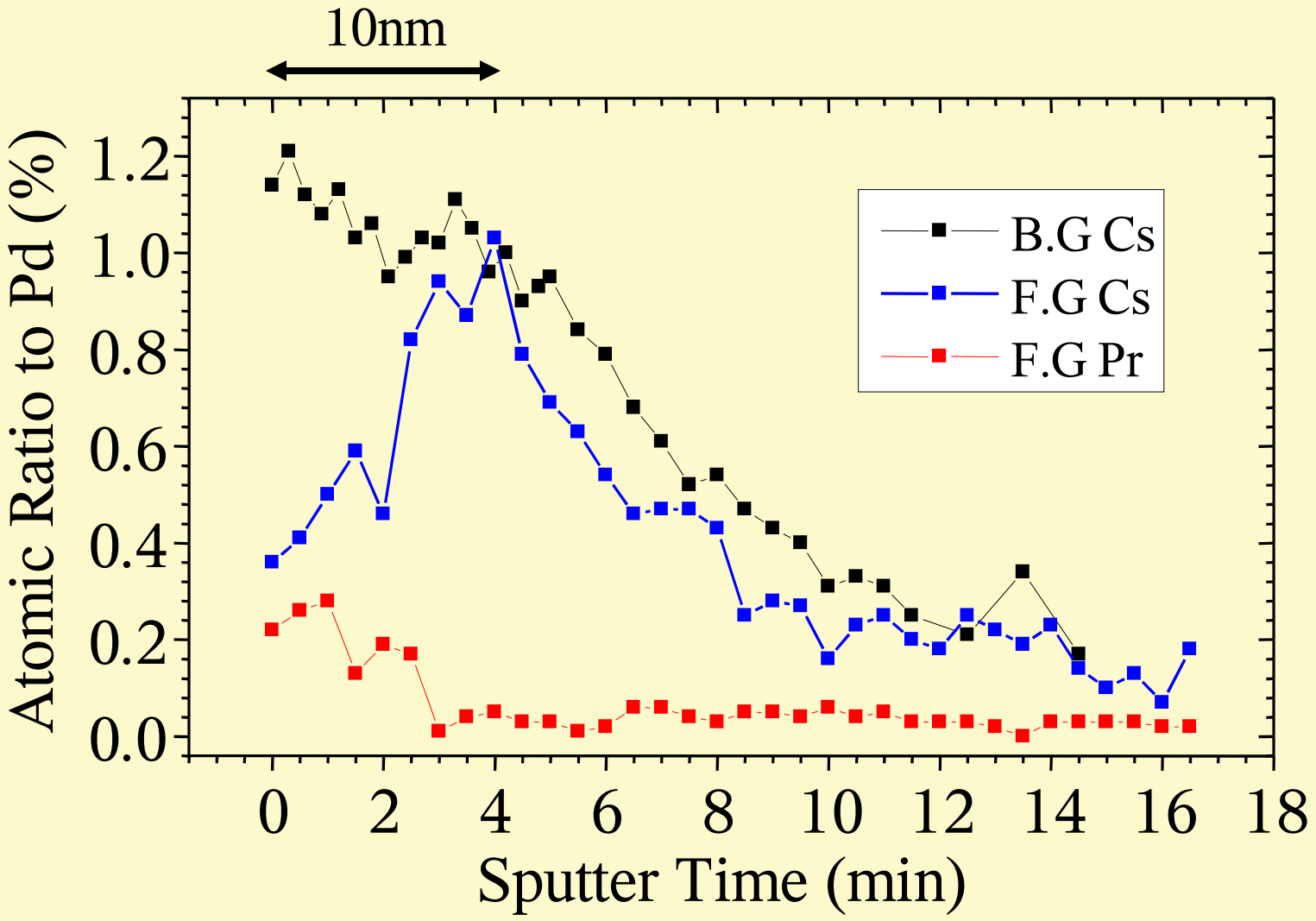


Depth Profile of Cs and Pr by TOF-SIMS

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

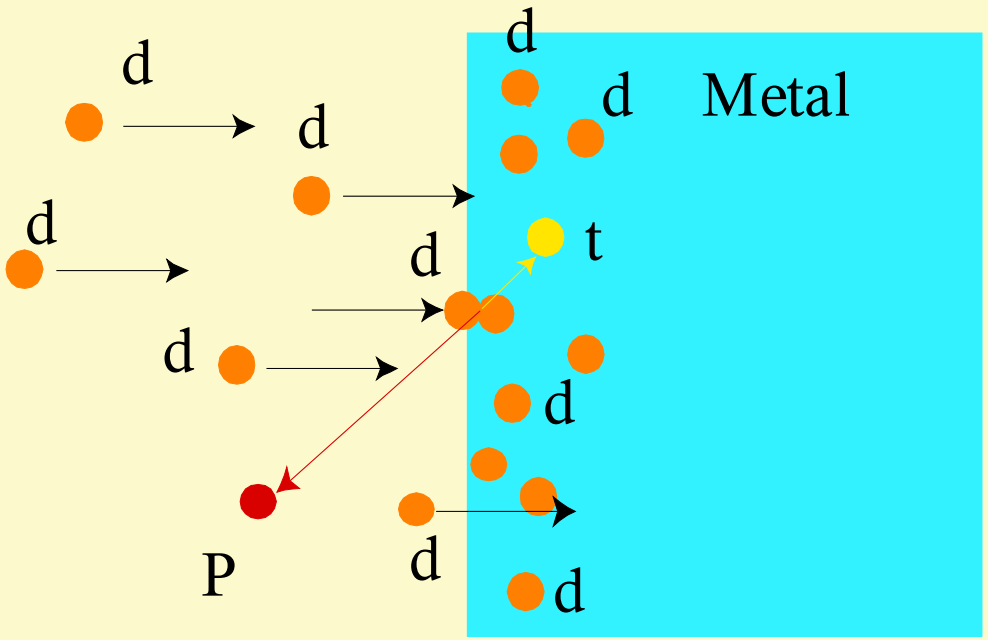


Depth Profile of Cs and Pr by XPS(1)

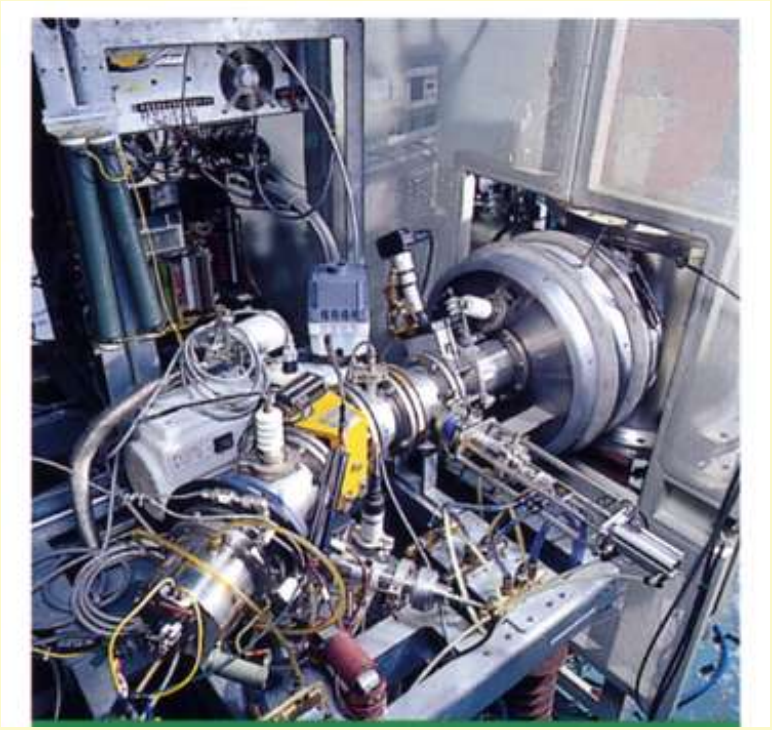


D^+ Ion Bombardment Experiment Performed at Tohoku Univ.

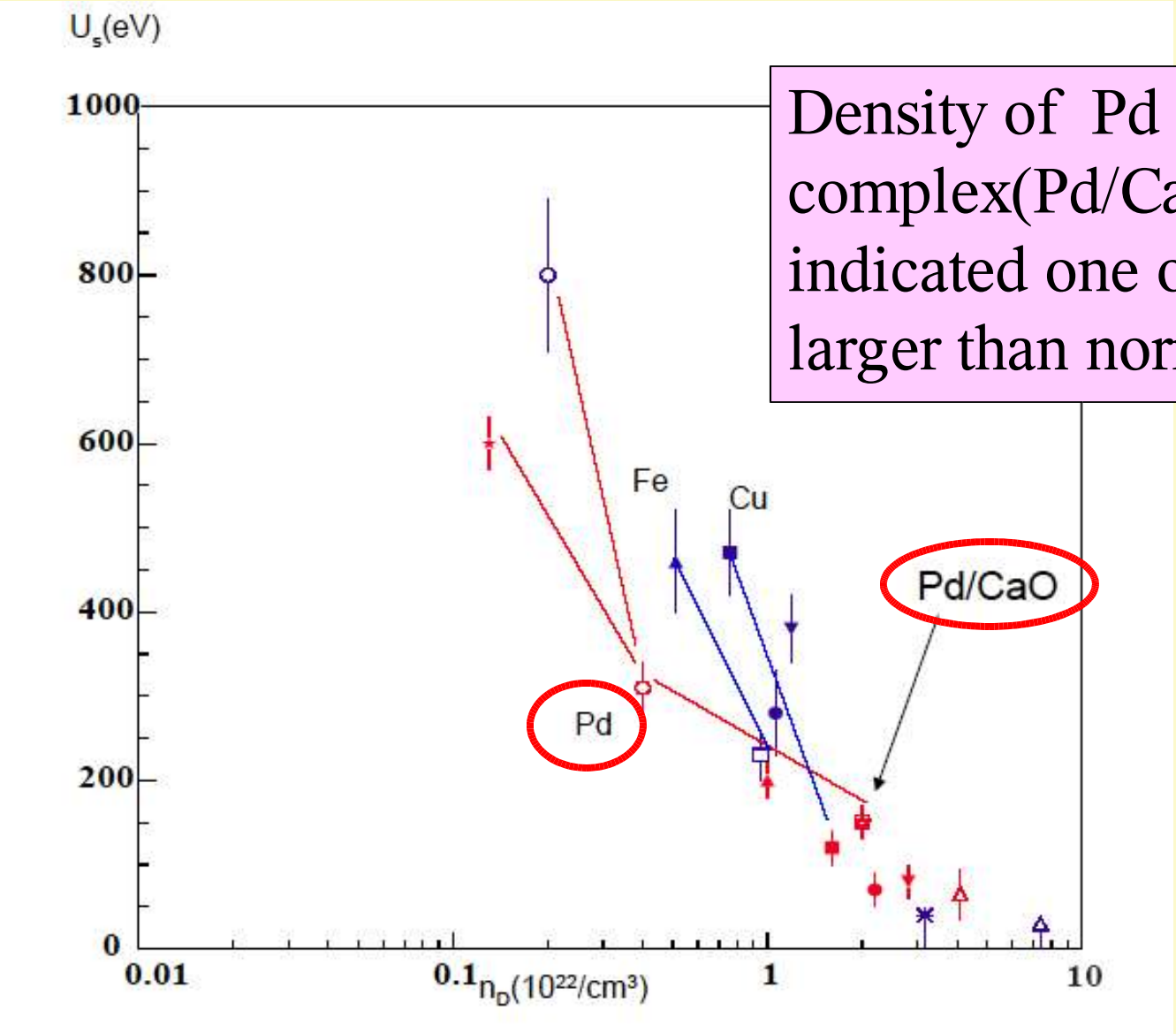
D+ Ion beam bombardment on metal target



Experimental Apparatus

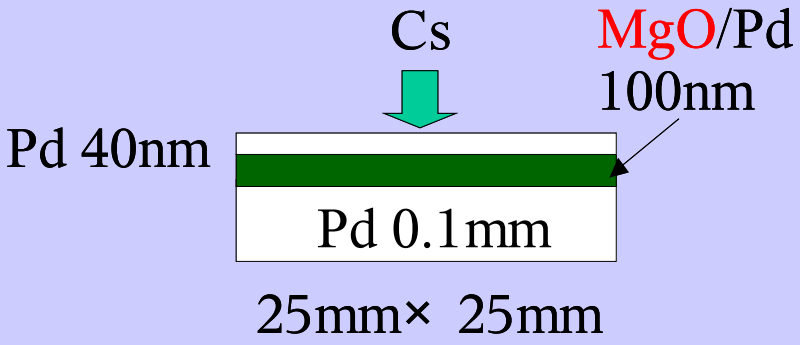


Deuterium Density measured by D^+ Ion Bombardment Experiment



Density of Pd complex(Pd/CaO) indicated one order larger than normal Pd

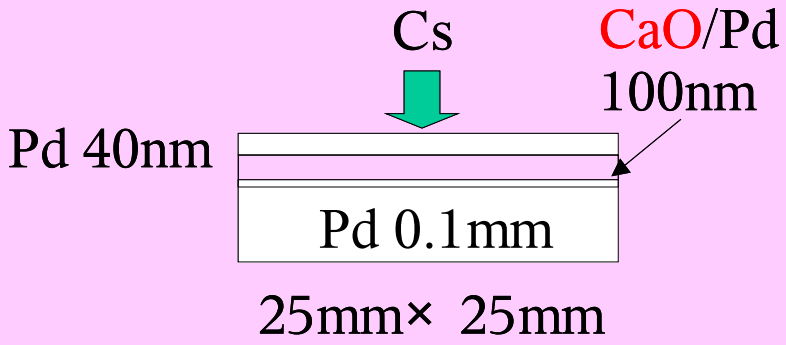
MgO cannot work instead of CaO



No Pr; Two cases out of two experiments.

ICP-MS measurements show no Pr(<0.01ng).

D_2 gas Flow rate enough(2-3sccm)



Almost every time Pr were detected.

More than 60 cases.

Consideration on the Role of CaO

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

- Increase of Deuterium Density?
- Modify the Electronic State of Surface Pd?

Depth Profile Measurement of D
By a Resonance Nuclear Reaction

$$^{7}_{3}Li(^{2}_{1}d,\gamma)^{9}_{4}Be$$

Concluding Remarks

1. **Transmutations of Ba into Sm were observed both when natural Ba was applied to the Pd complex samples, and when mass-137-enriched Ba (monoisotopic Ba) was applied. The mass distribution of Sm that we obtained depended on the starting isotopic distribution of Ba.**

2. **One of our experimental apparatus was carried to SPring-8 to perform an in-situ measurement. We obtained some Pr signals by the X-ray Fluorescence method.**

3. **According to a D^+ ion beam bombardment experiment performed at Tohoku University, the deuterium density of our Pd complex indicated one order larger than normal Pd.**