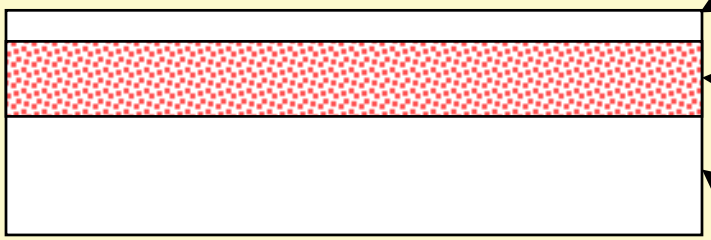
Pd Complex

Pd complex containing CaO



Thin Pd Film : 400Å

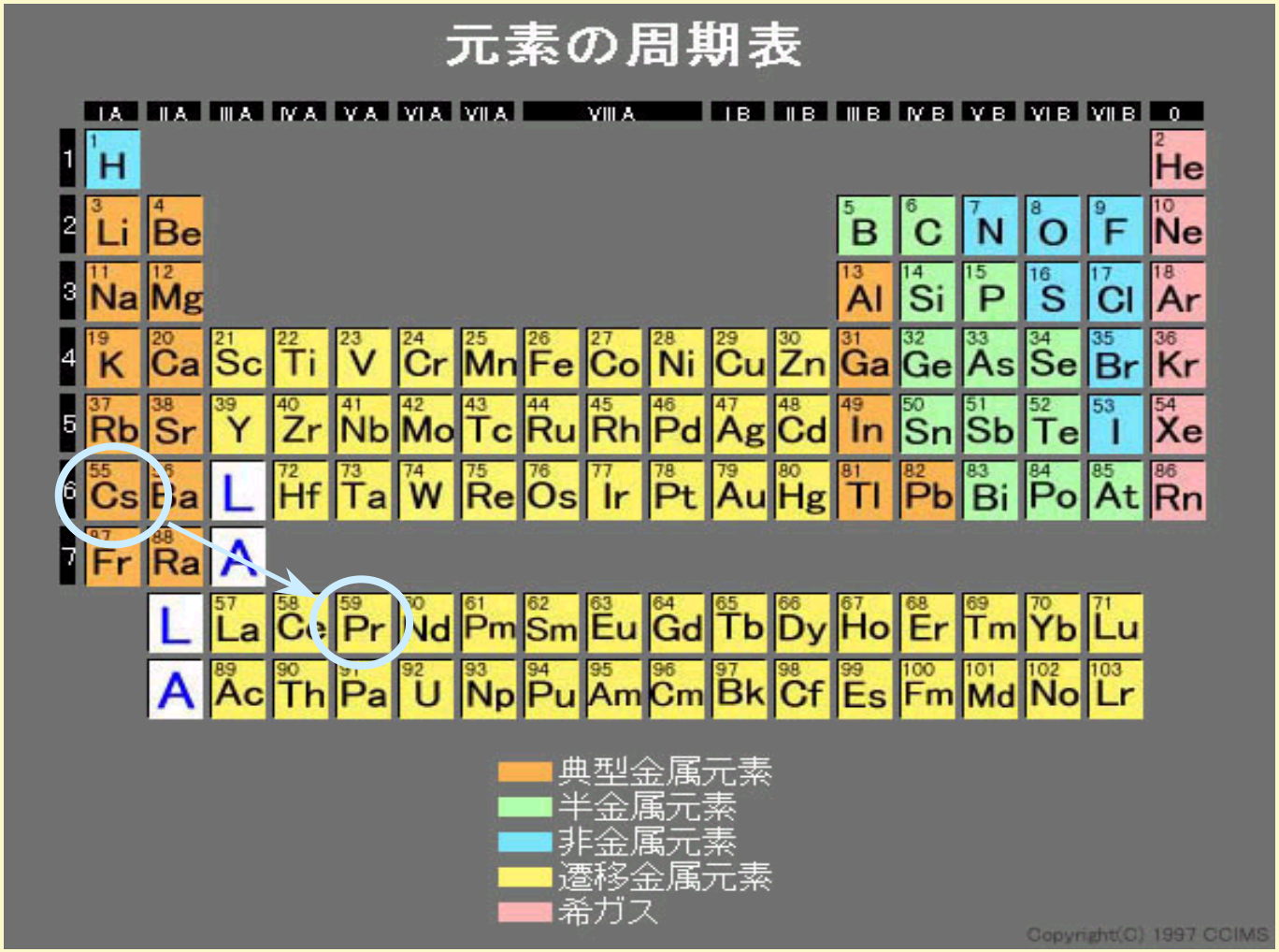
CaO and Pd Layers
:1000Å

25mm×25mm

Bulk Pd : 0.1mm

Transmutation of Cs into Pr

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



$^{133}_{55}Cs$

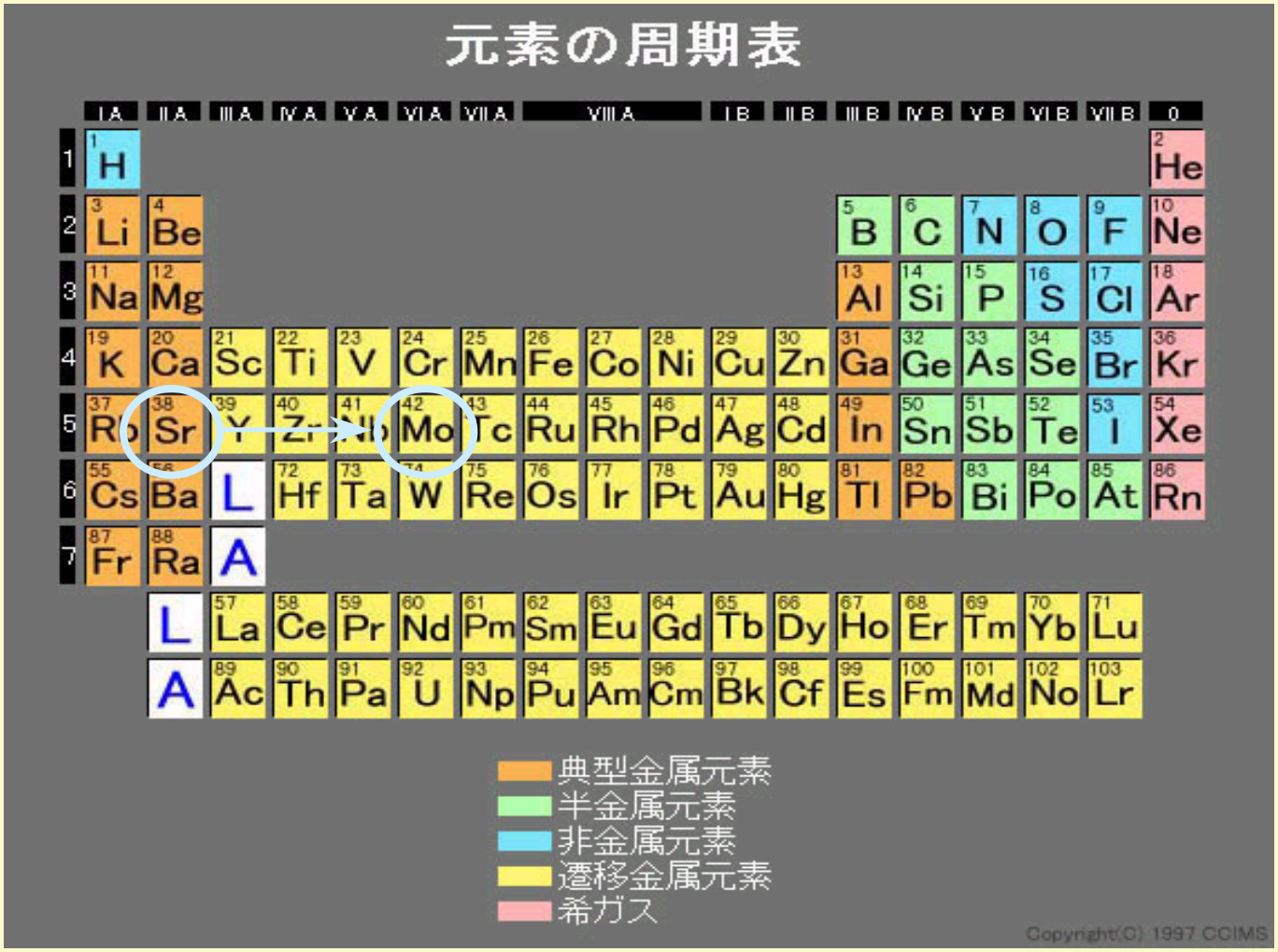
Atomic N. +4

Mass N. +8

$^{141}_{59}Pr$

Transmutation of Sr into Mo

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



$^{88}_{38}Sr$

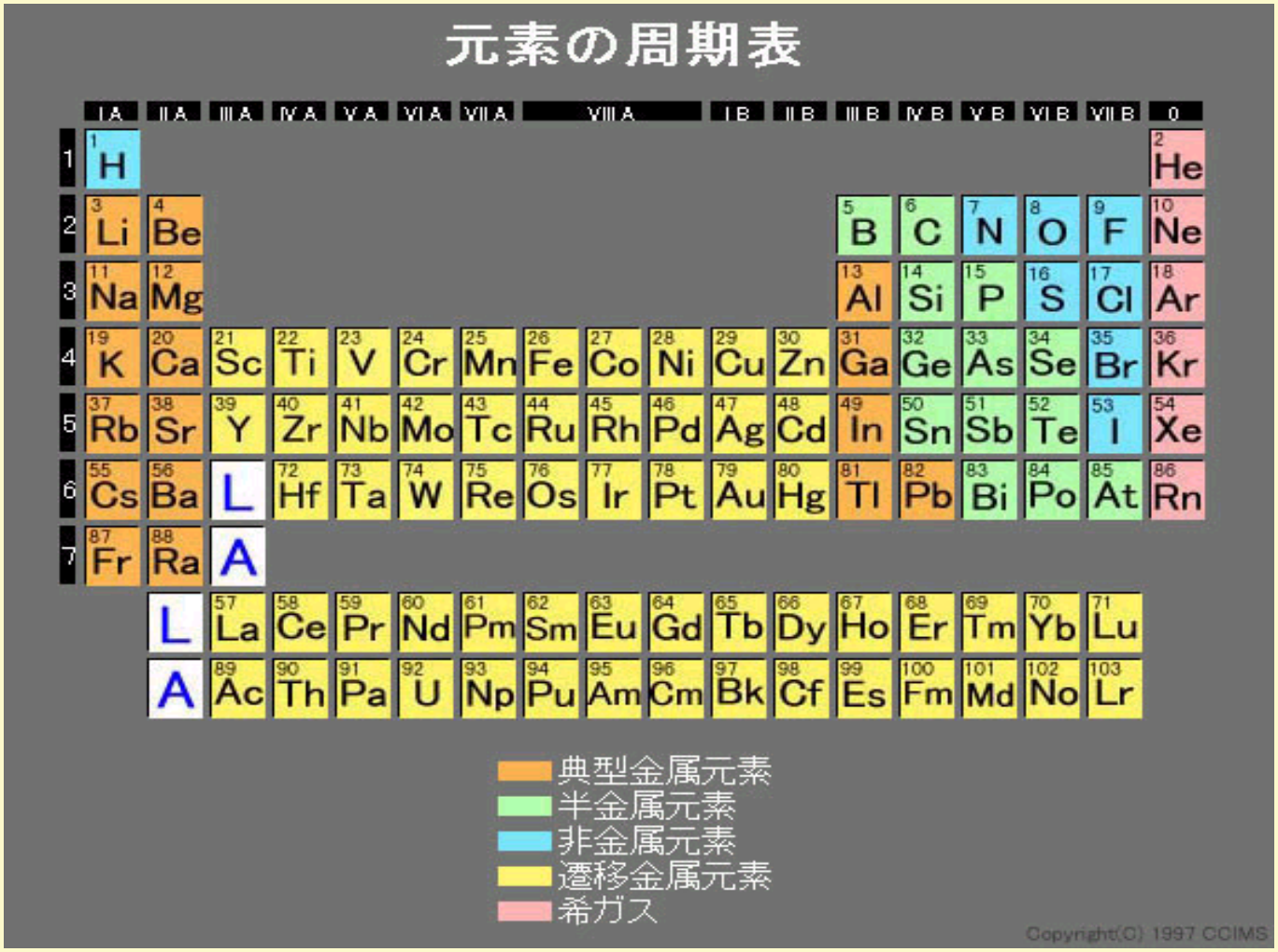
Atomic N. +4

Mass N. +8

$^{96}_{42}Mo$

Transmutation of Ba into Sm

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



$^{137,138}_{56}Ba$

Atomic N. +6

Mass N. +12

3 Alpha

$^{149,150}_{62}Sm$

Isotopic Anomaly of the detected Mo

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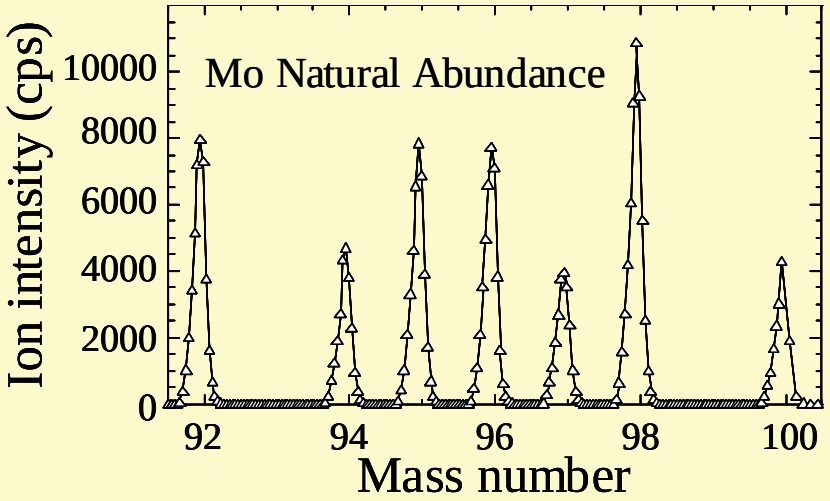
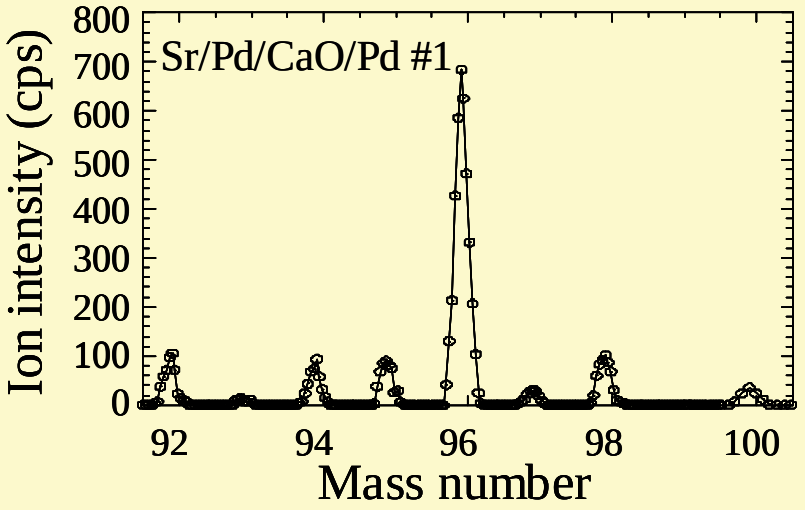
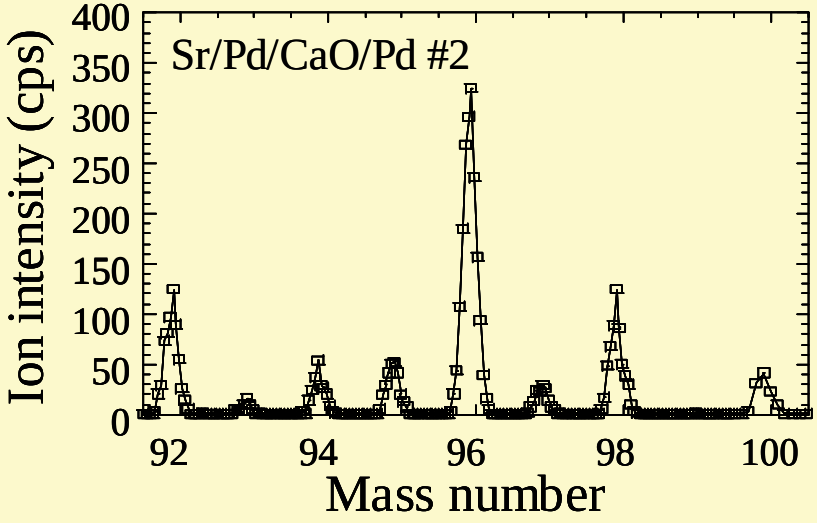
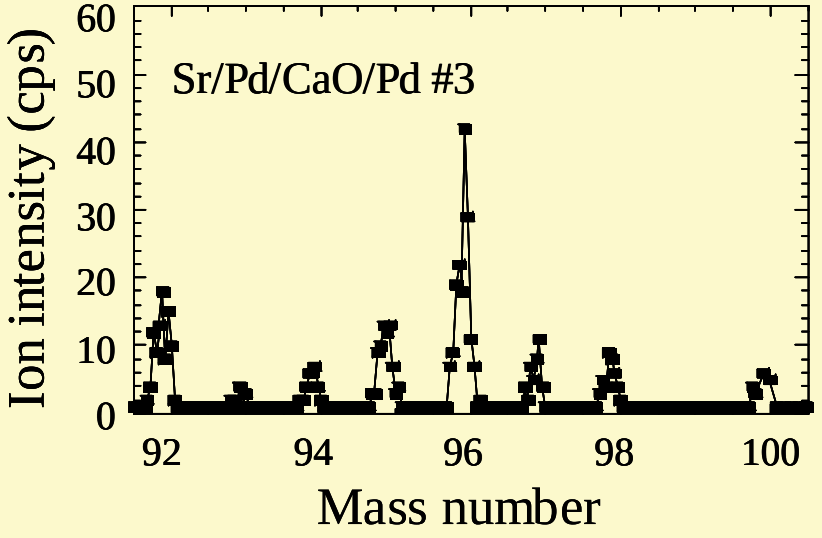
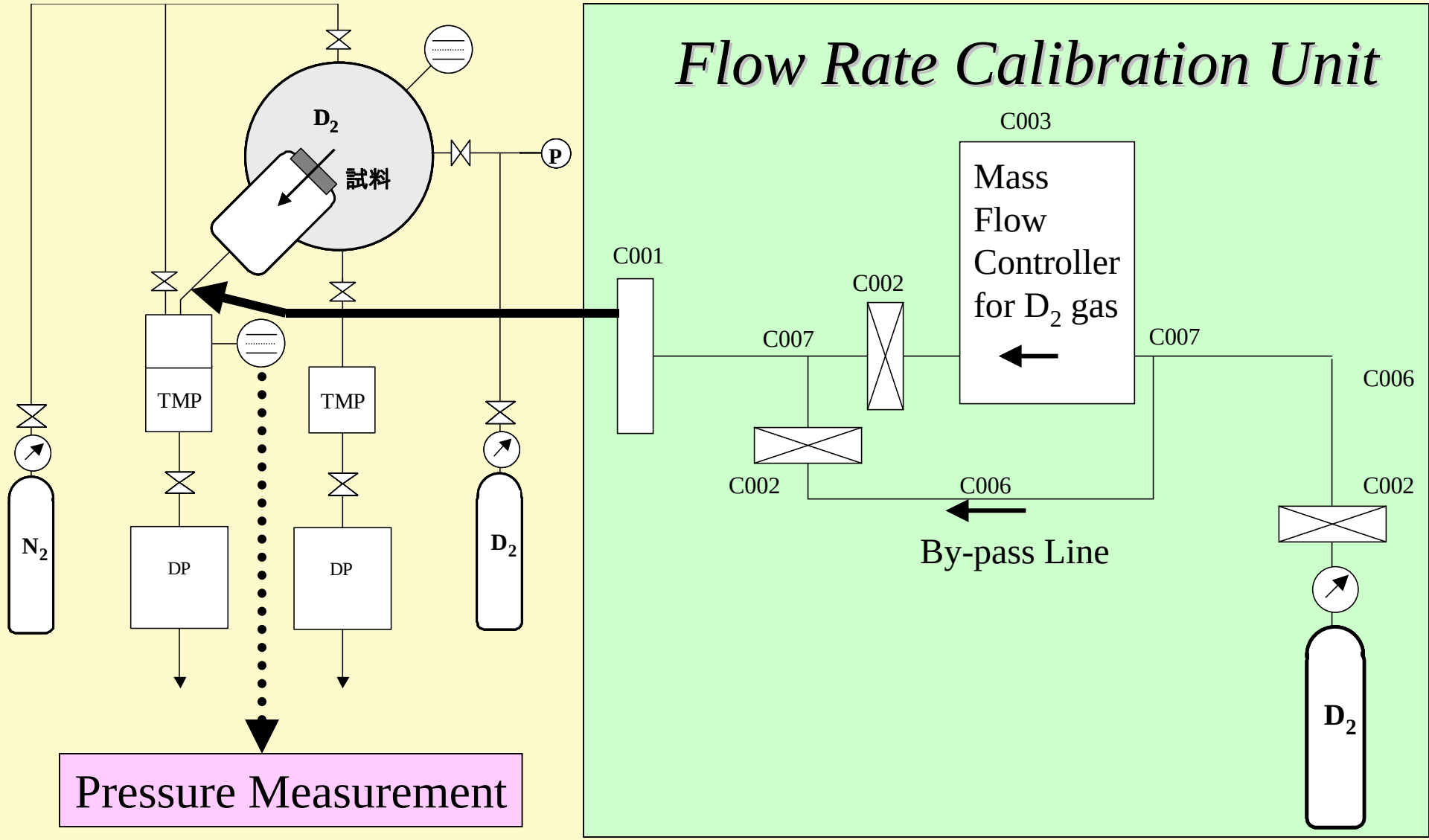






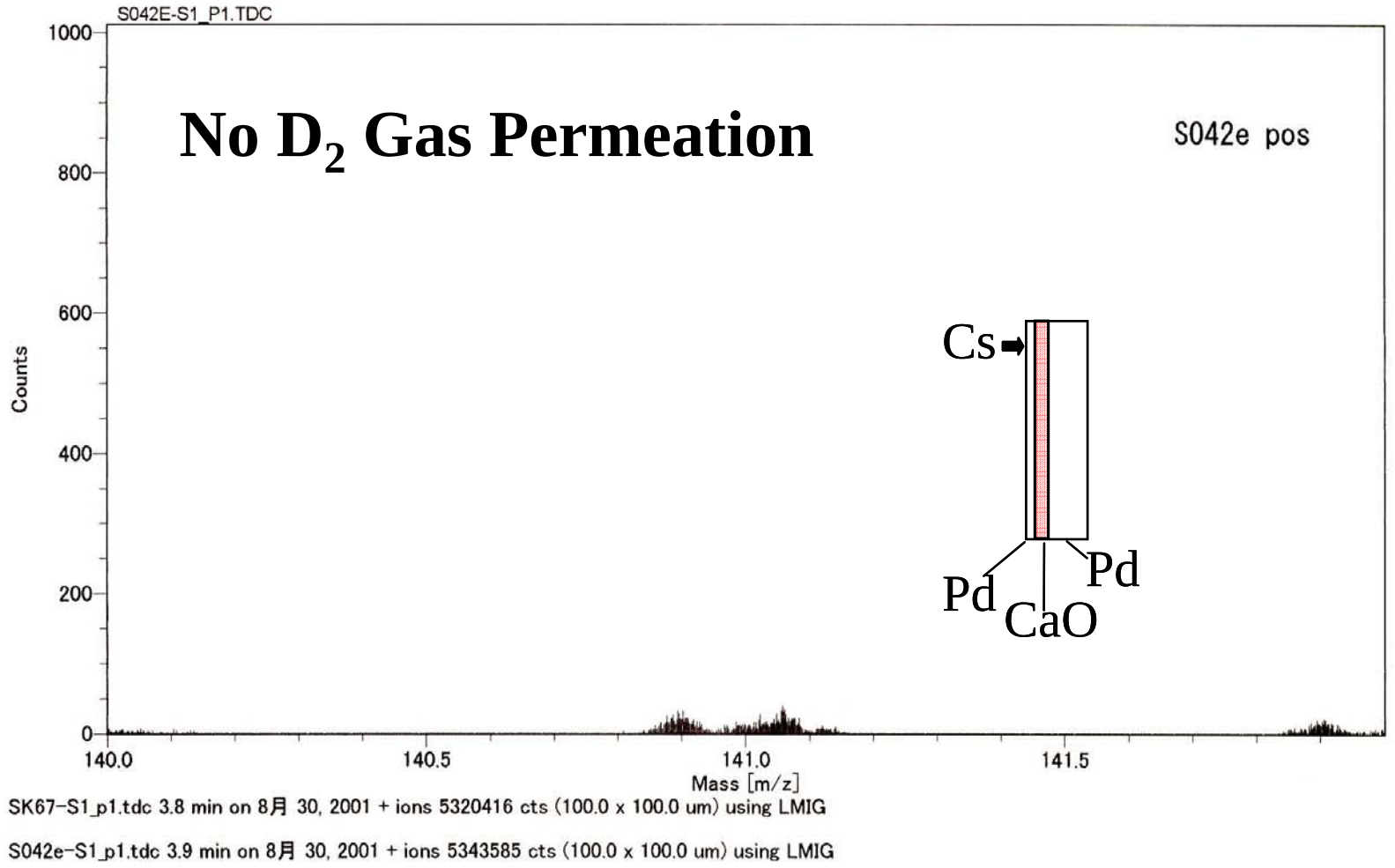


Evaluation of D_2 Gas Flow Rate



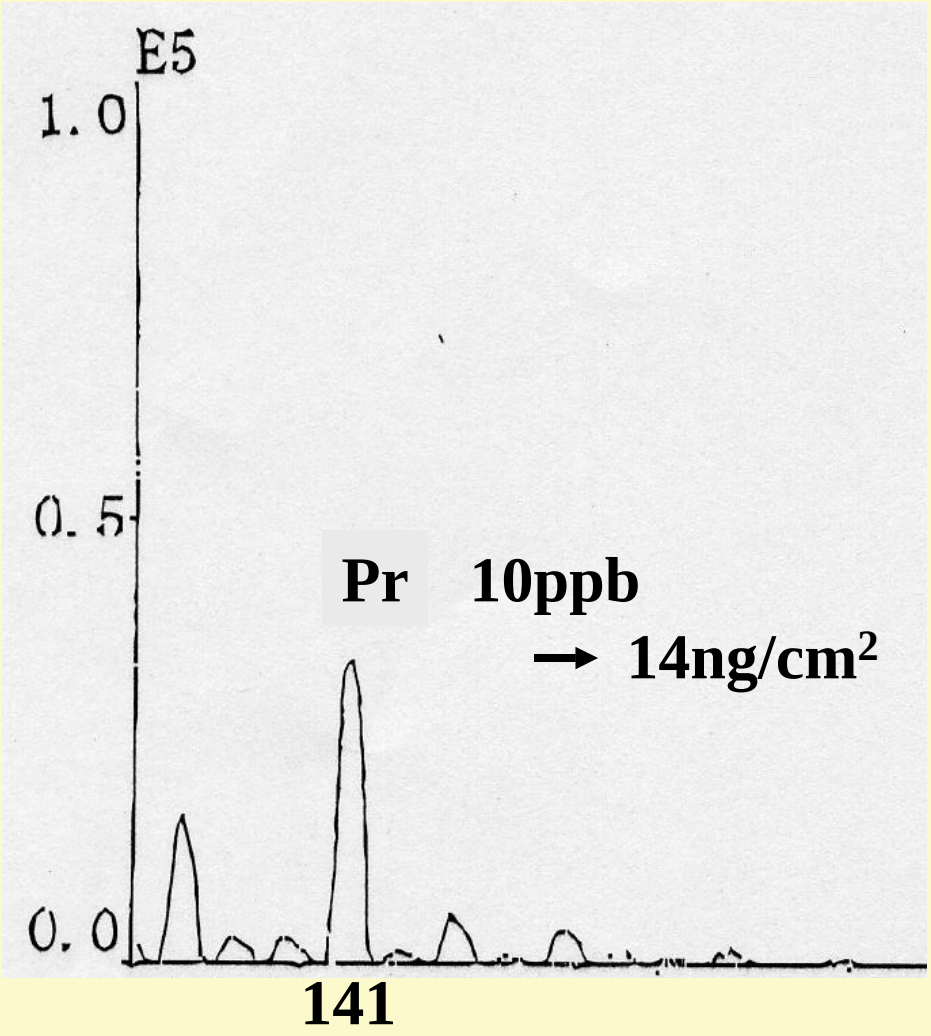
Identification of Pr by TOF-SIMS(2)

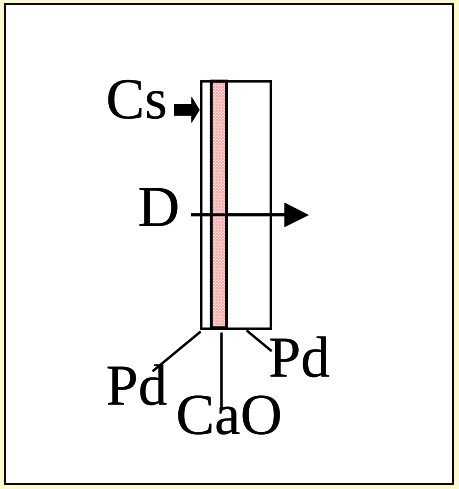
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



TOF-SIMS device (TRIFT™ Ⅱ;ULVAC-PHI)

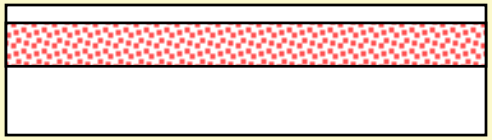
Cs添加Pd多層膜重水素透過後



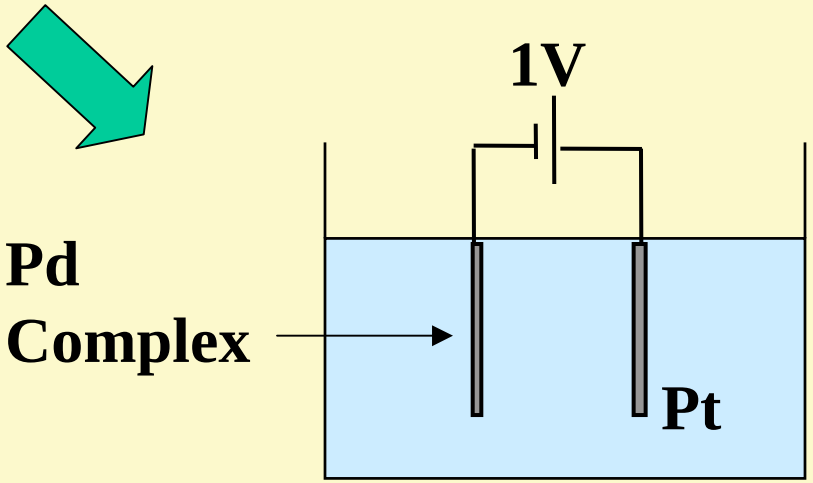


→ Prを検出、定量

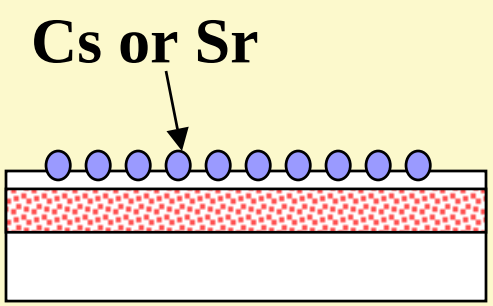
Element Addition
(1: Electrolyte Addition)



Pd Complex

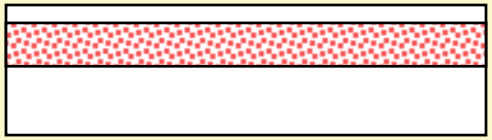


1mM $CsNO_3/D_2O$ solution
or 1mM $Sr(OD)_2/D_2O$ solution



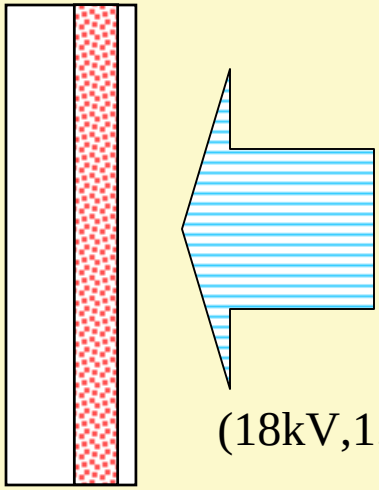
Pd Complex

Element Addition
(2: Ion Implantation)



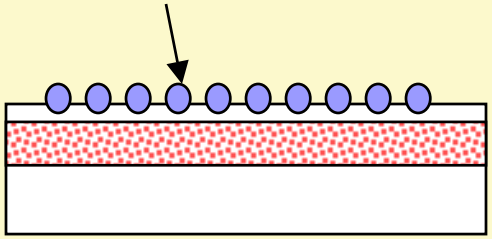
Pd Complex

Cs Ion implantation



(18kV,1.0E15 ions/cm2)

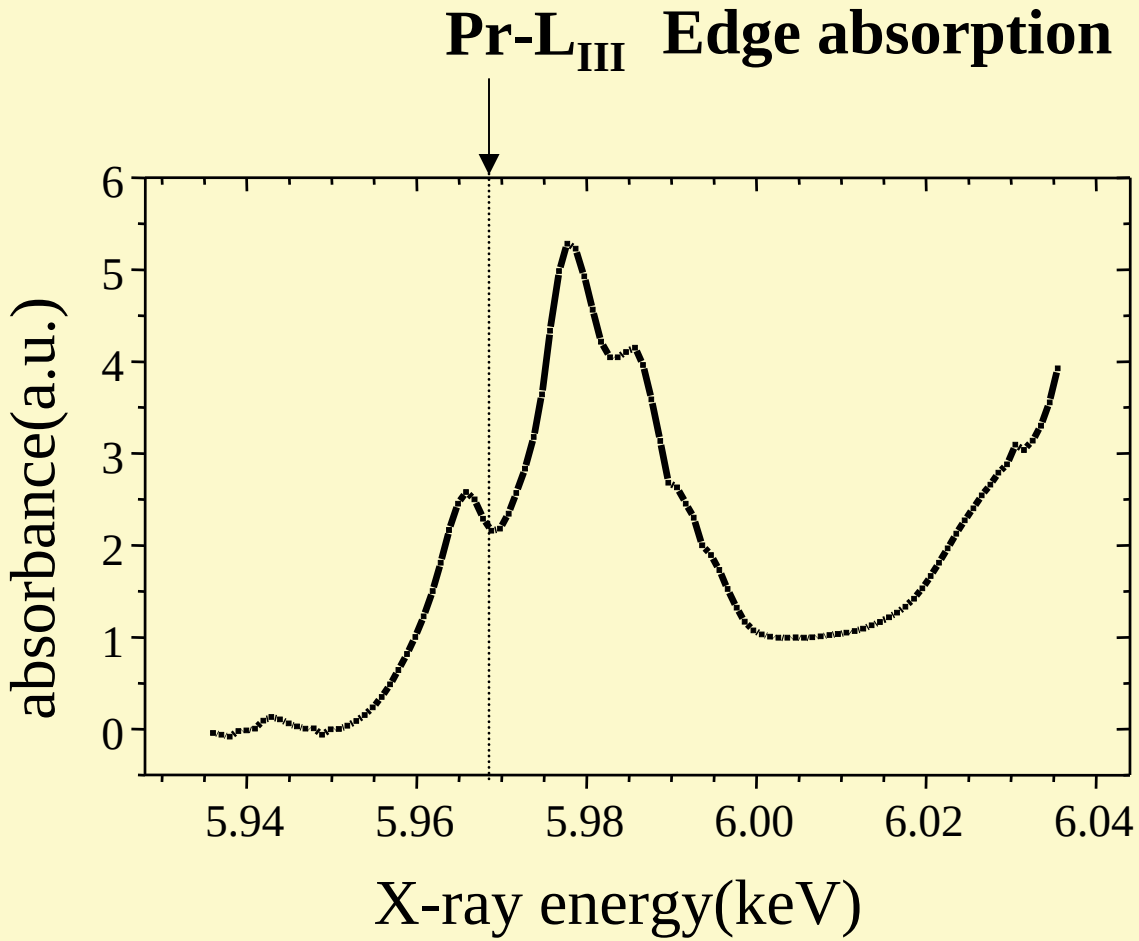
Pd Complex



Pd Complex

Identification of Pr by XANES

XANES(X-ray Absorption Near Edge Structure)



BL-9A Line, KEK, Tsukuba, Japan

Identification of Pr by ICP-MS

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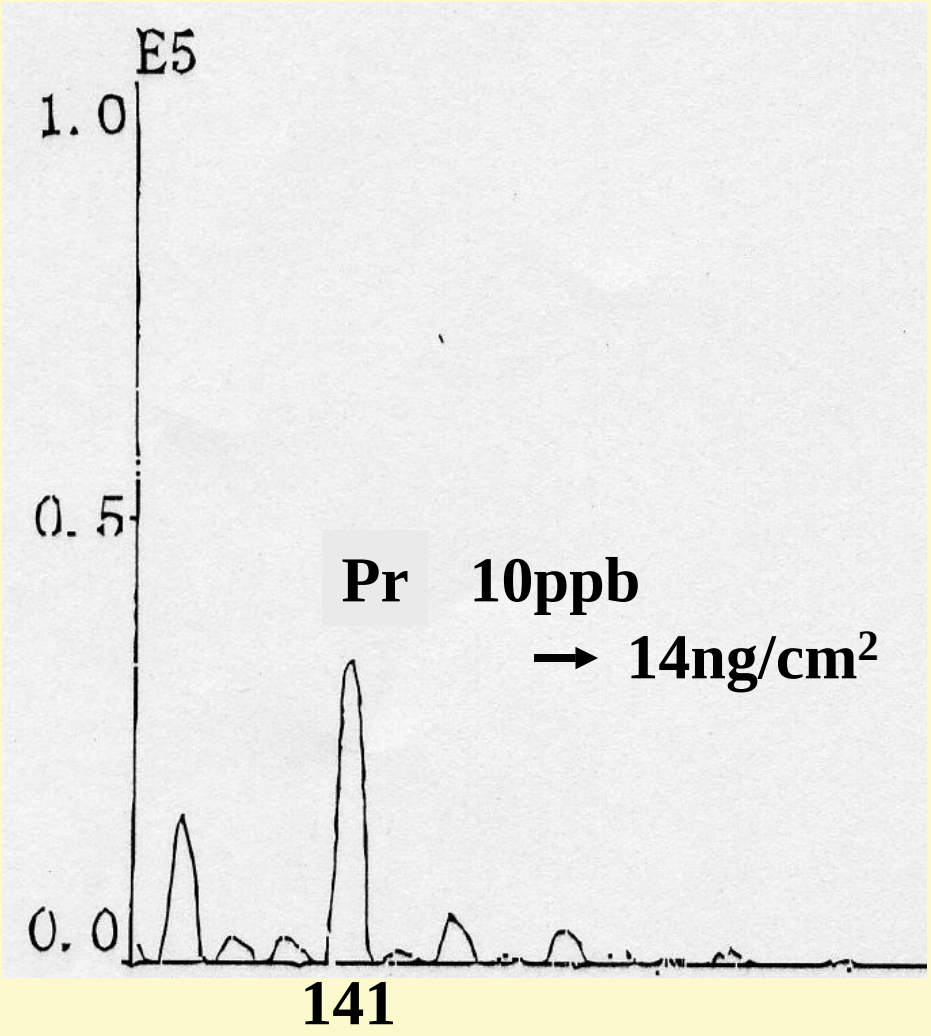
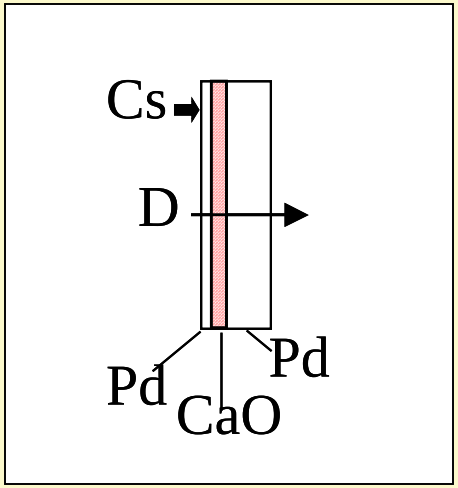
ADVANCED TECHNOLOGY RESEARCH CENTER





→ Quantitative Analysis

Preparation of the Pd Complex

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Washing a Palladium Sample with Acetone

↓

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

↓

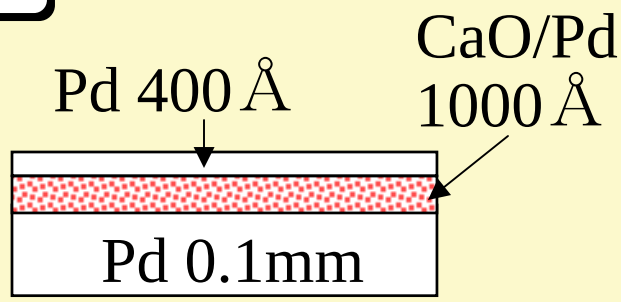
Washing the Sample with Aqua Regia (100sec)

↓

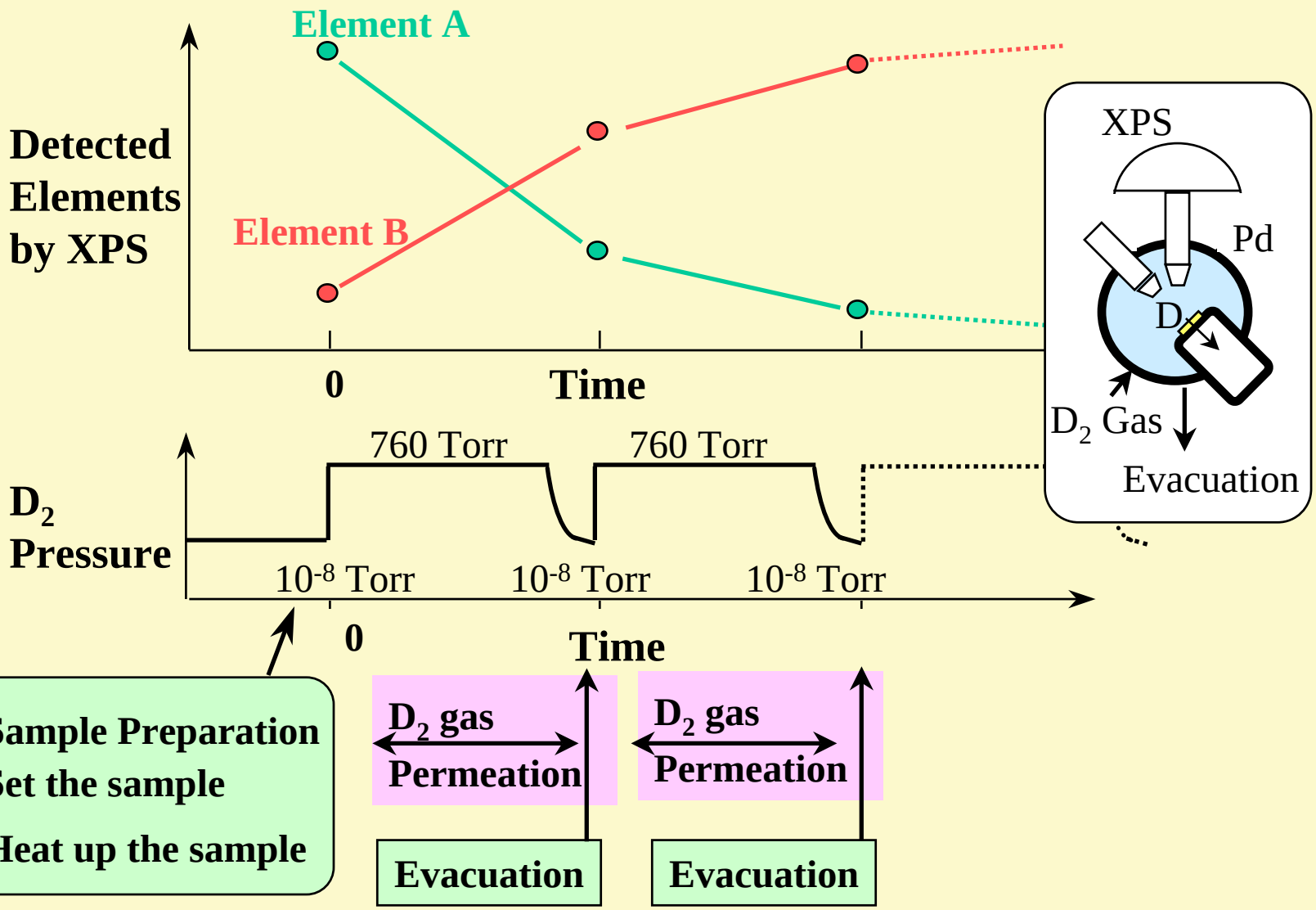
5 times Alternatingly Sputtering of CaO(20 Å) and Pd(180 Å)

↓

Ion Beam Sputtering of Pd only (400 Å)



Procedure of an Experiment



Transmutation of Sr into Mo

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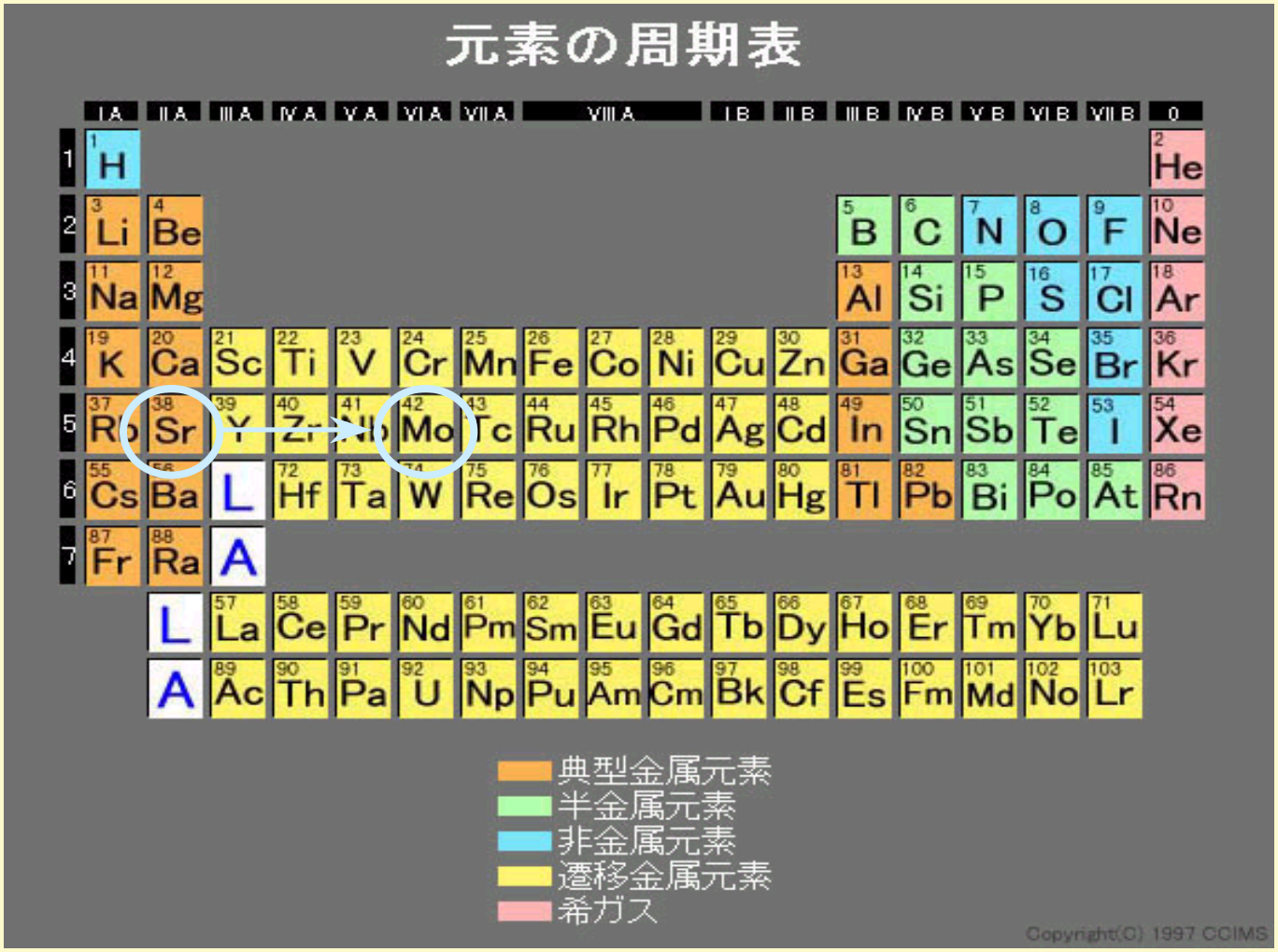
ADVANCED TECHNOLOGY RESEARCH CENTER



$^{88}_{38}Sr$

Atomic N. +4

Mass N. +8

$^{96}_{42}Mo$

Excess Energy and Q-value

$$^{133}_{55}Cs + 4d \rightarrow ^{141}_{59}\mathrm{Pr} + (\gamma, phonon, e^{-}\,?)$$

$$Q \approx 50.5 MeV$$

$$EXH = 800 J$$

$$P_{EXH} \approx 2.2 mW$$

Experimental results

$$\mathrm{Pr} \approx 10^{14}\, atoms$$

$$reaction\ time \approx 100 h$$

→Undetectable in our experimental setup

Depth Profile of Cs and Pr : Ion Implantation

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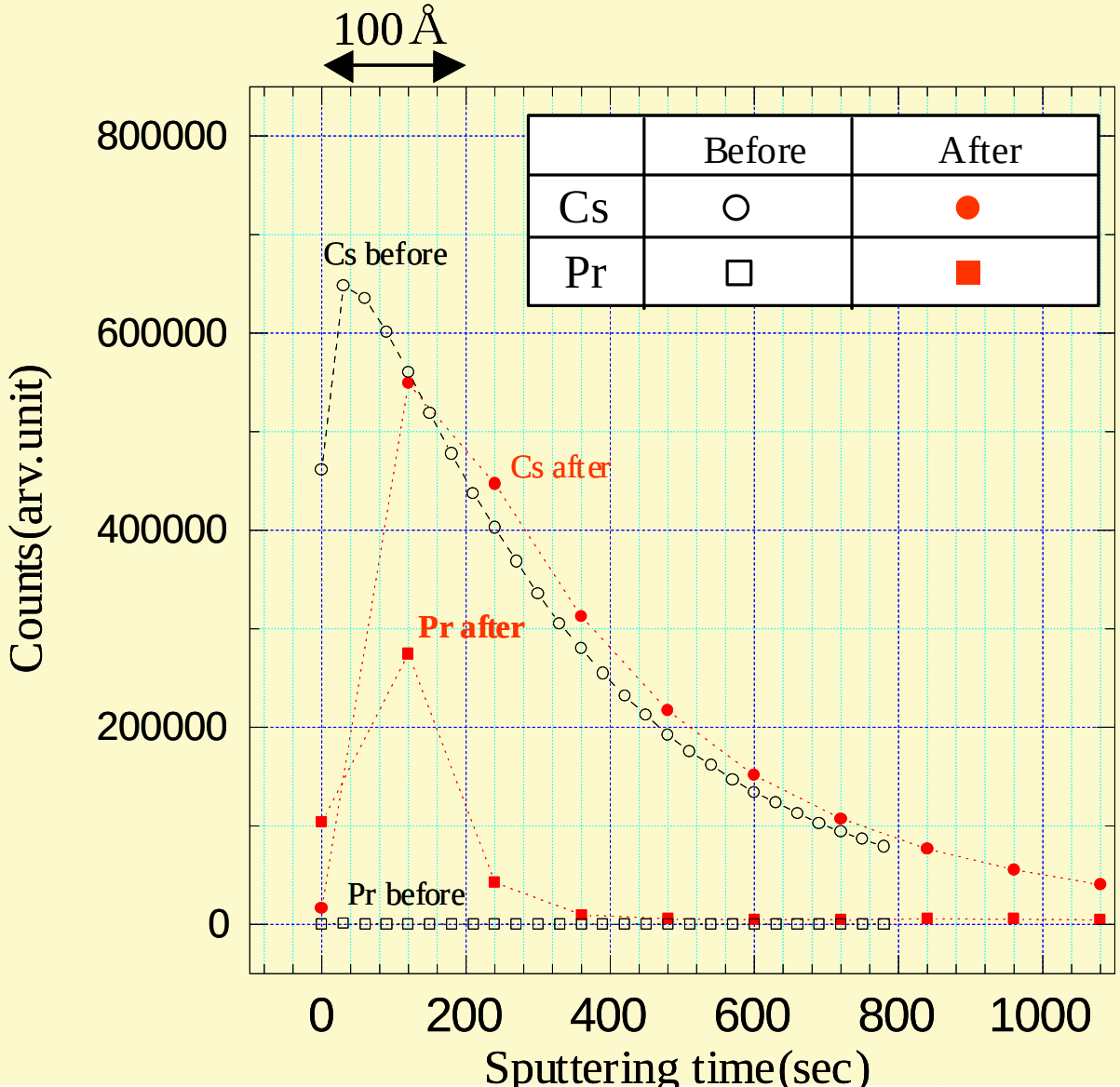
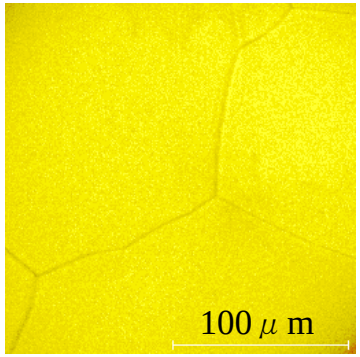
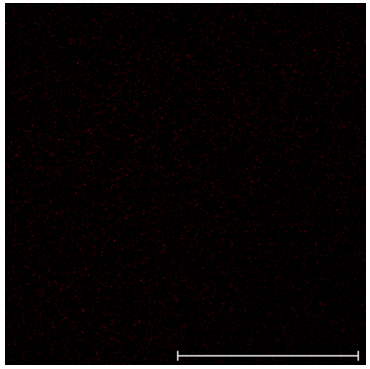
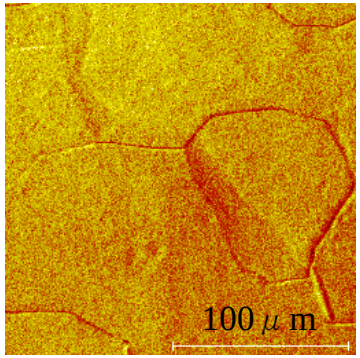
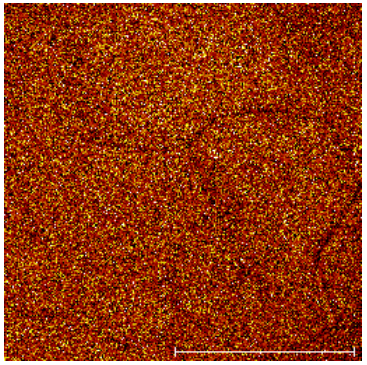


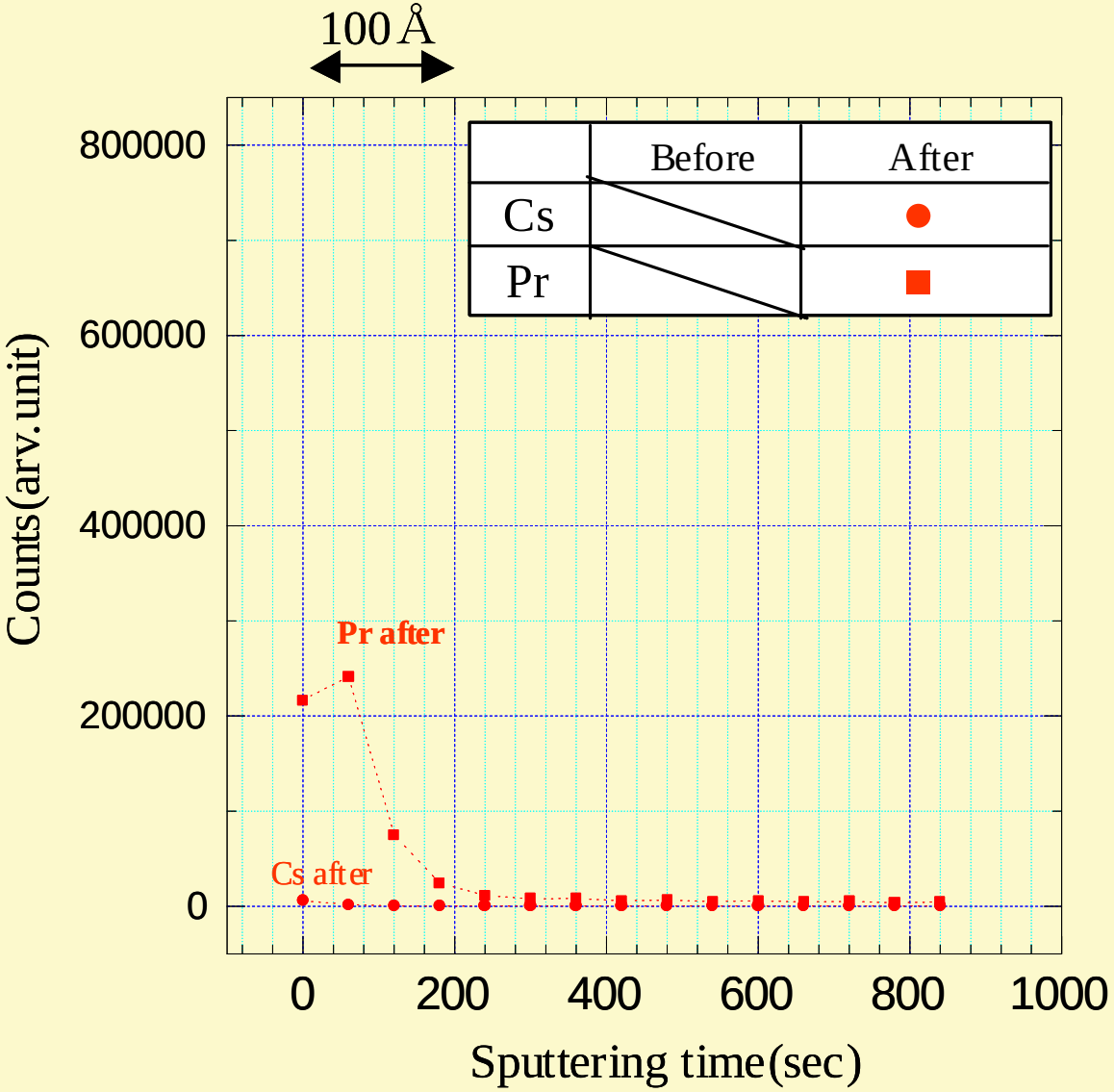
Surface Distribution of Cs and Pr : Ion Implantation

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

Sample:Cs^+ Ion Implantation
Analysis:Tof-SIMS(ULVAC fai)

	Cs	Pr
before		
after		

Depth Profile of Cs and Pr : Electrolyte Addition

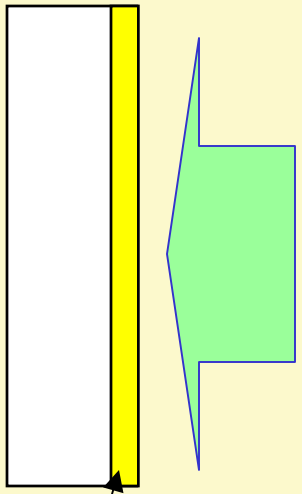


Ultra Low Energy Beam Model

D Permeation(D Flux)

⇕

Ultra low D beam



$$R = \sigma \cdot N_{Cs} \cdot \phi$$

R : reaction rate(event/cm^3/sec)

σ : cross section(cm^2) N_{Cs} : Number of Cs (1/cm^3)

ϕ : Deuteron beam flux(1/cm^2/sec)

$$\eta = \int_t (R / N_{Cs})dt = \int_t (\sigma \cdot \phi)dt = \sigma \int_t \phi \, dt$$

$$\int_t \phi \, dt \approx f \cdot FL \cdot T_{\exp} / S$$

FL : Flow rate(sccm) $T_{\exp}$: Reaction Time(sec)

S : Permeation surface area(cm^2)

$$\therefore \eta \propto FL$$

→ Agree with the experimental results

Rough Estimation of the Cross Section

$$\eta = \sigma \cdot f \cdot FL \cdot T_{\exp} / S$$

$$= \sigma \cdot \frac{2 \times 6 \times 10^{23}}{22.4 \times 10^{3} \times 60} \cdot FL \cdot 100 \times 3600 / 1.0$$

$$= \sigma \left[cm^2 \right] \cdot FL \left[sccm \right] \cdot 3 \times 10^{23} \left[1 / cm^2 / sccm \right]$$

Experimental results $\rightarrow 0.3 = \sigma \cdot 3 \times 10^{23}$

$$\therefore \sigma \approx 1 \times 10^{-24} \left[cm^2 \right] = 1 \left[barn \right]$$

$cf.\ \sigma_c = 27.2\text{barn}$: ^{133}Cs for thermal neutron

Separation of the Products and Contaminants(2)

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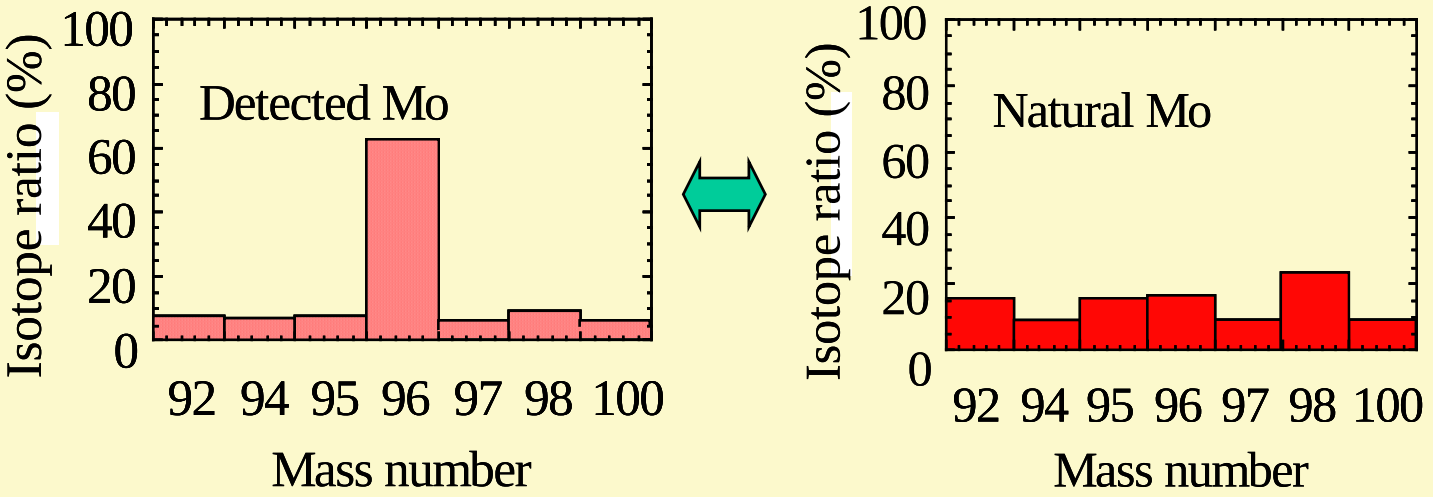
2. Anomalous Isotopic Composition: Mo



Detection of Pr and Mo cannot be explain by contamination.

Pr and Mo are the products of nuclear transmutation reactions.

Surface Distribution of Cs and Pr : Electrolyte Addition

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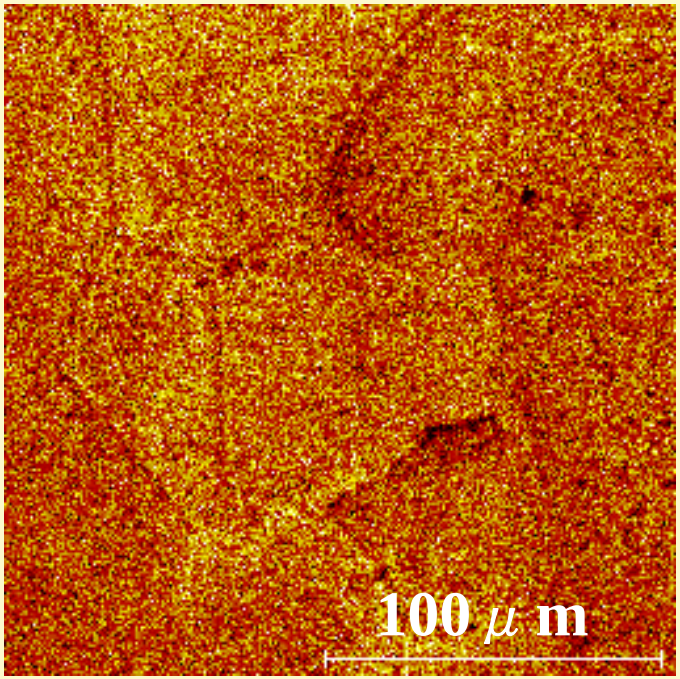
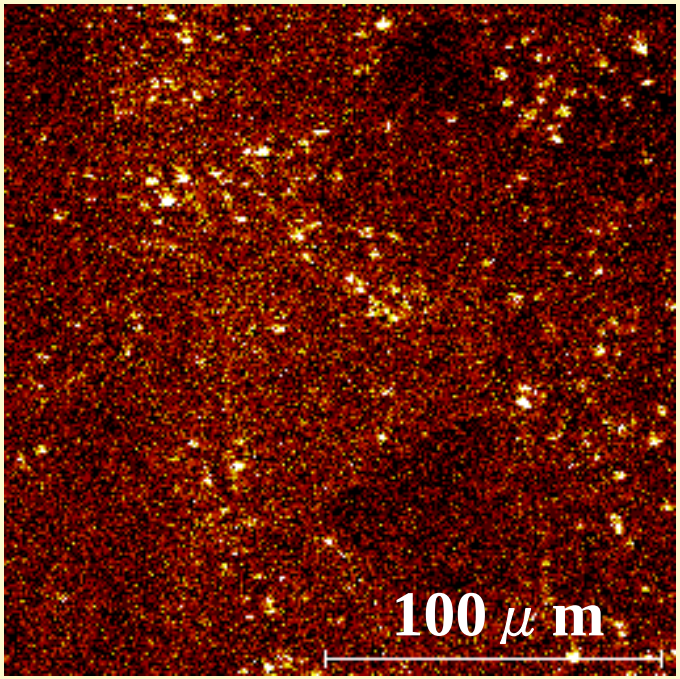
Sample:Cs is added using electrochemical method
Analysis:Tof-SIMS(ULVAC fai)

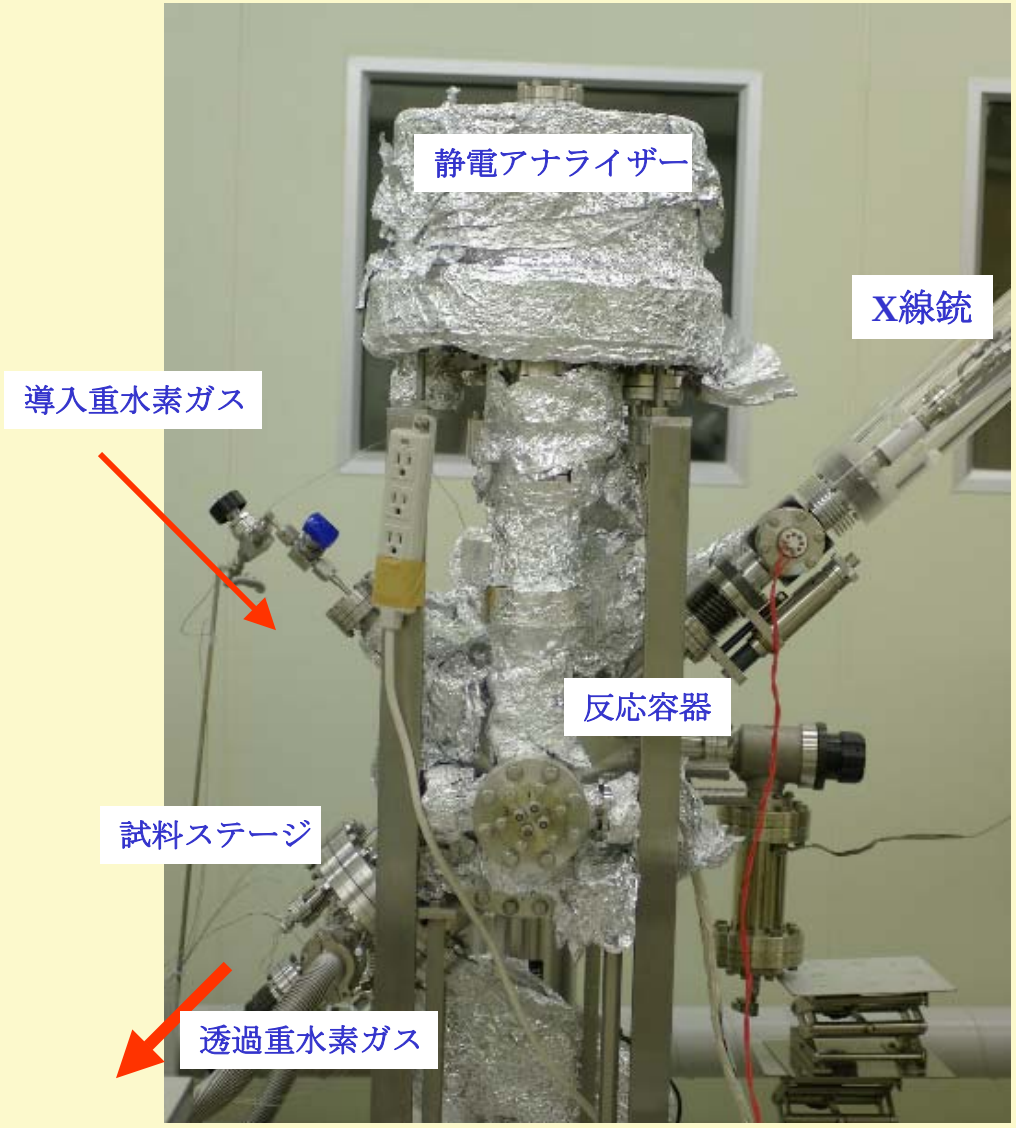
Cs

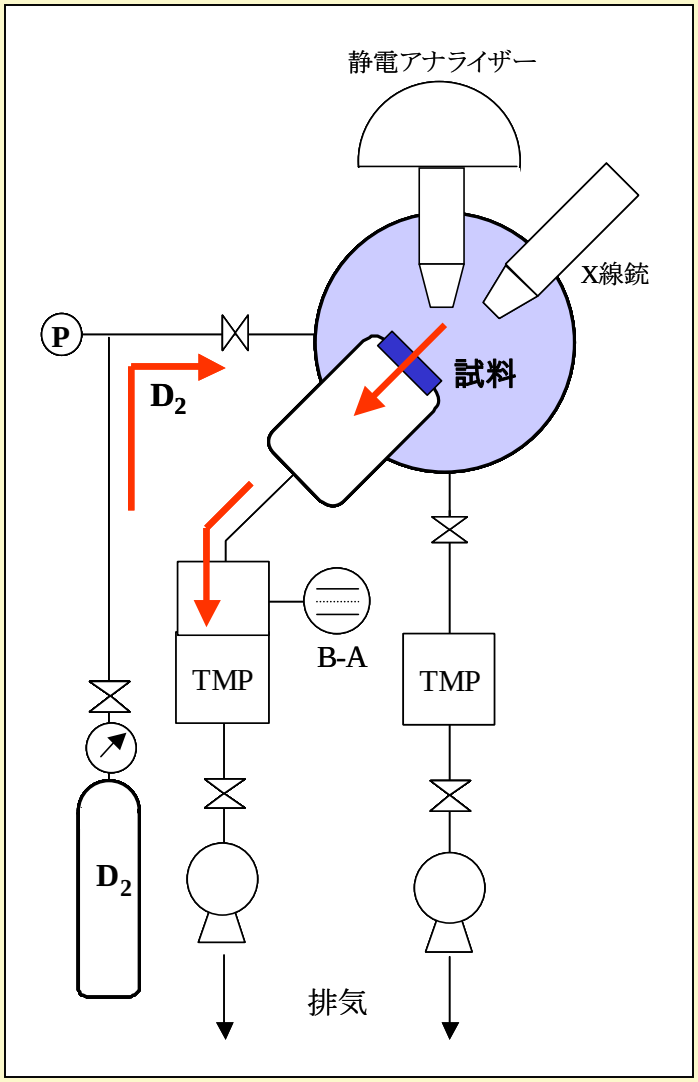


Pr

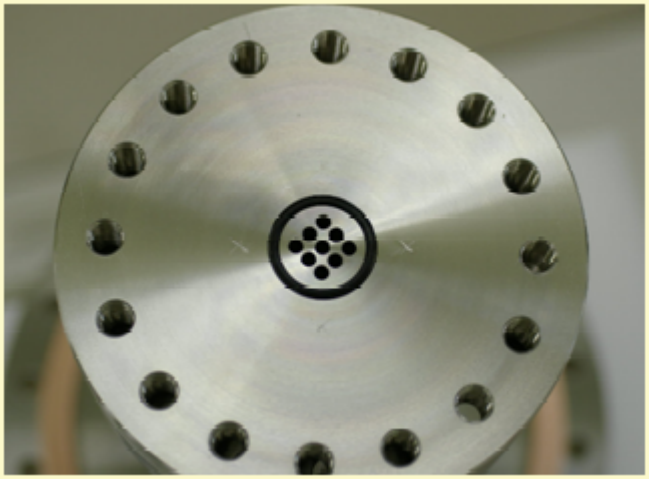


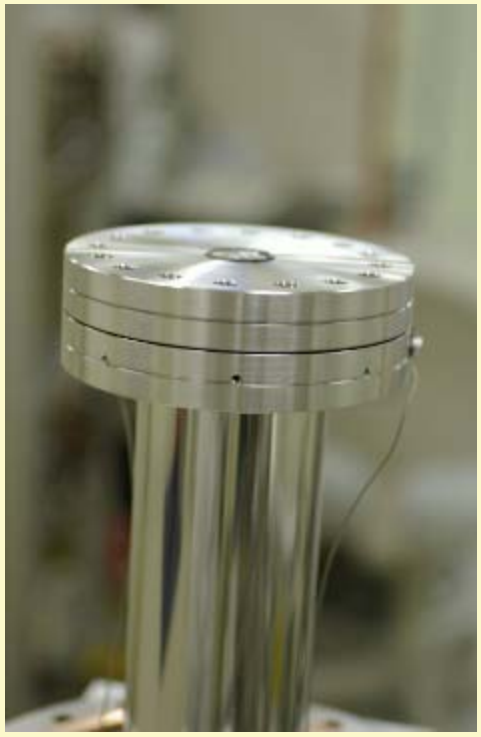
実験装置概観

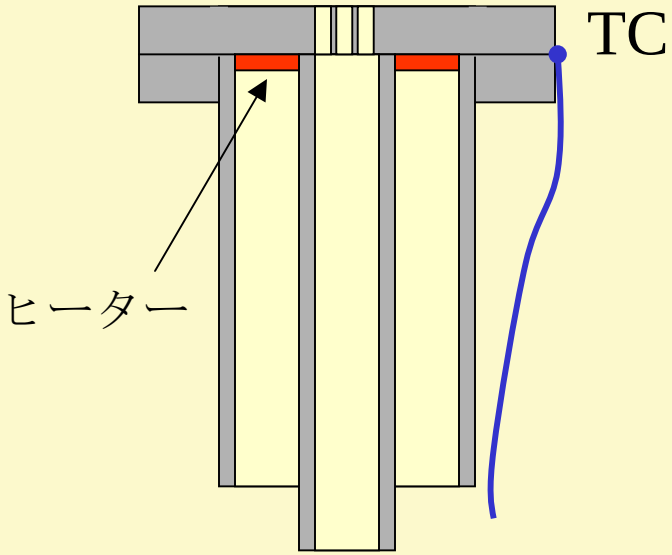

静電アナライザー
X線銃
導入重水素ガス
反応容器
試料ステージ
透過重水素ガス


静電アナライザー
X線銃
P
D2
試料
TMP
B-A
TMP
D2
排気

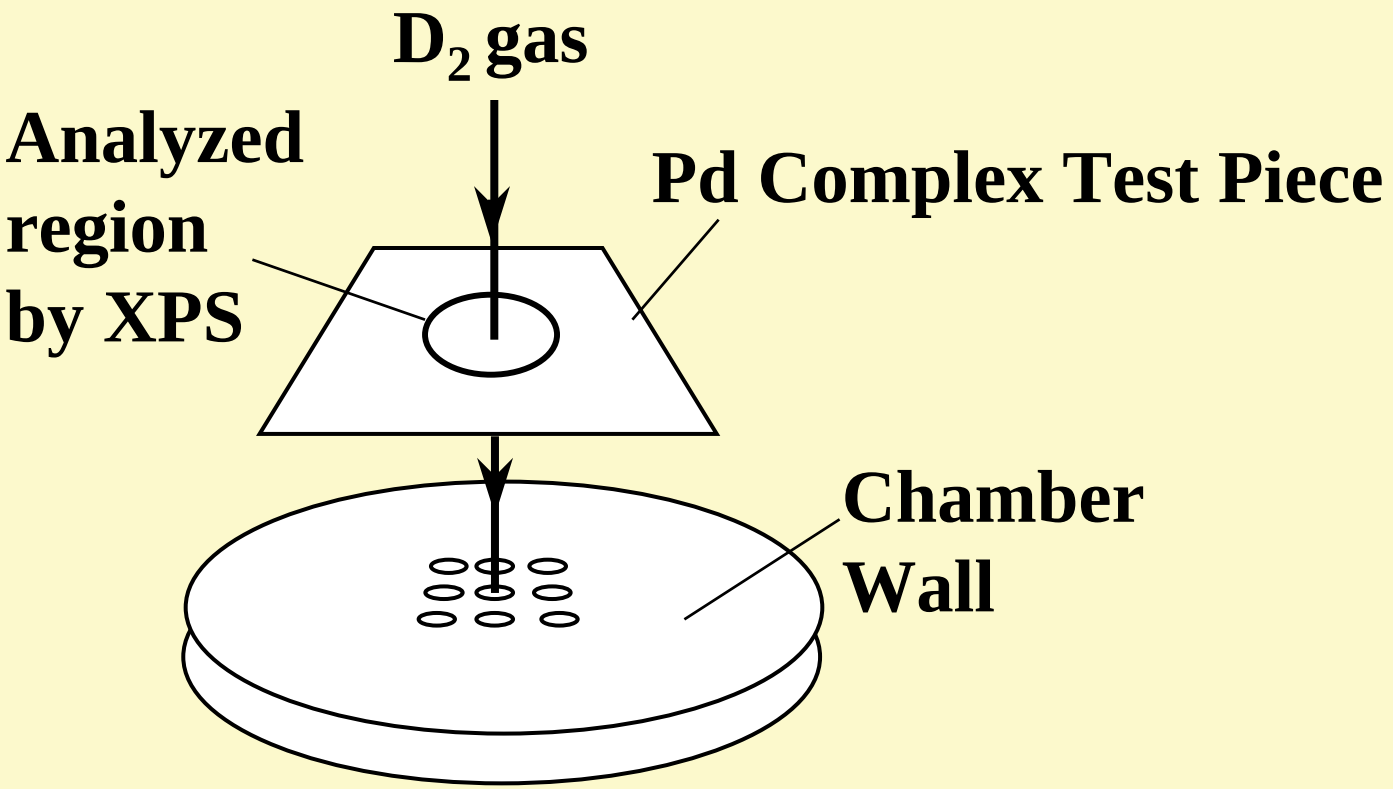
試料ステージ概観



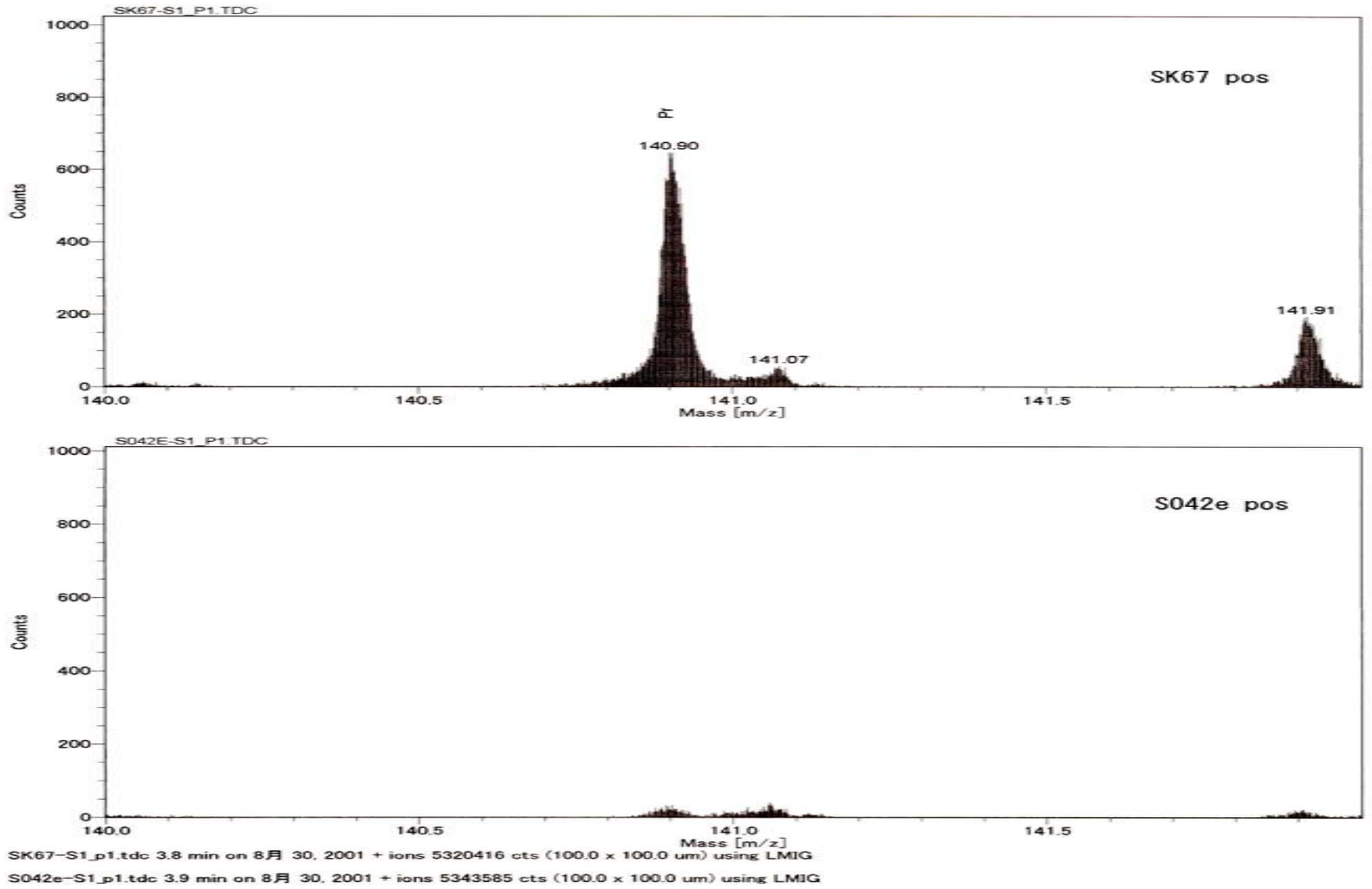




重水素ガスの透過経路


D2 gas
Analyzed region by XPS
Pd Complex Test Piece
Chamber Wall

Identification of Pr by TOF-SIMS(2)



Quantitative Analysis of Pr by ICP-MS

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ADVANCED TECHNOLOGY RESEARCH CENTER

Step1：Solve the surface of the Pd Complex by nitric acid
（The nitric acid is Ultra high purity; impurity Ni,Pb~50ppt

Step2：Quantitative Analysis of the solution by ICP-MS

ICP-MS(Inductively Coupled Plasma Mass Spectrometry）

High Sensitivity: Detection Limit ~ Pr 0.1ng

Necessary to exclude Molecular Ions

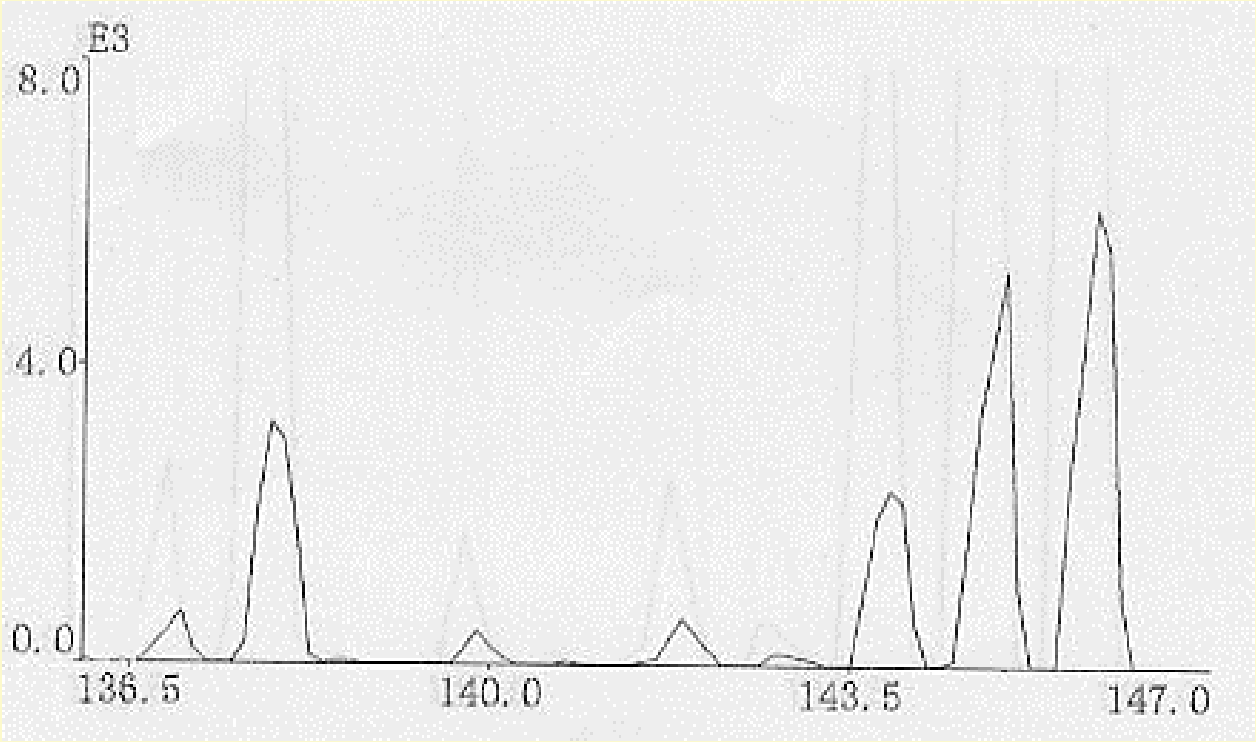
Device：SEIKO Instruments: SPQ9000

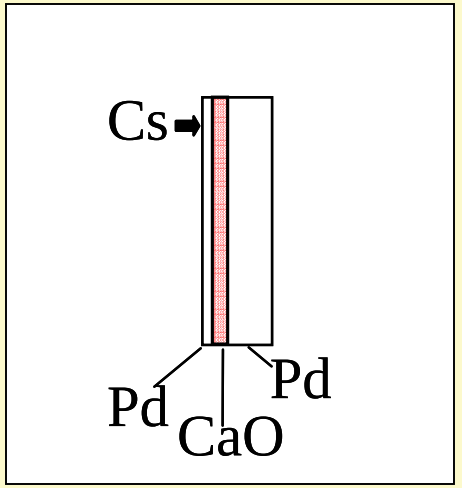
Examination of Molecular Ions

Pd	Pd(NO)	PdO_2
102(1%)	132	134
104 (11%)	134	136
105 (22%)	135	137
106 (27%)	136	138
108 (26%)	138	140
110 (12%)	140	142

No molecular ions interfering Mass 141(Pr) in this system

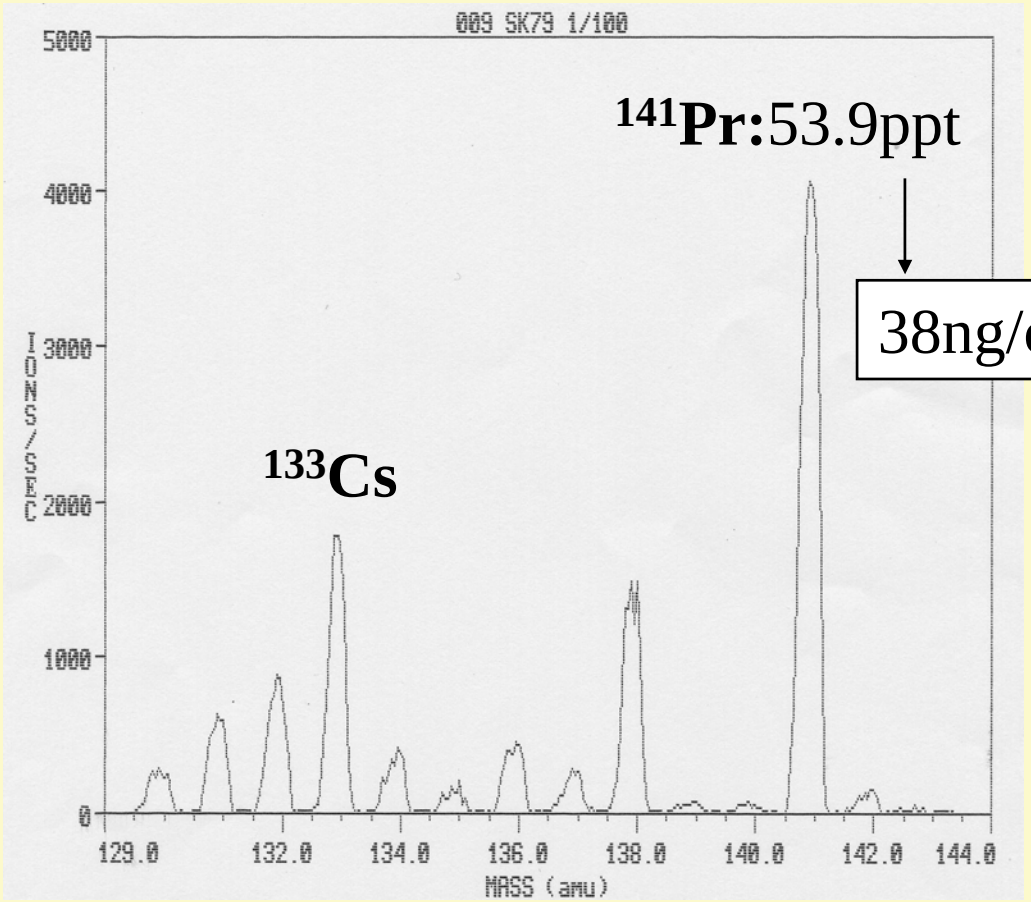
No D_2 Gas Permeation





→ Prは検出されず。

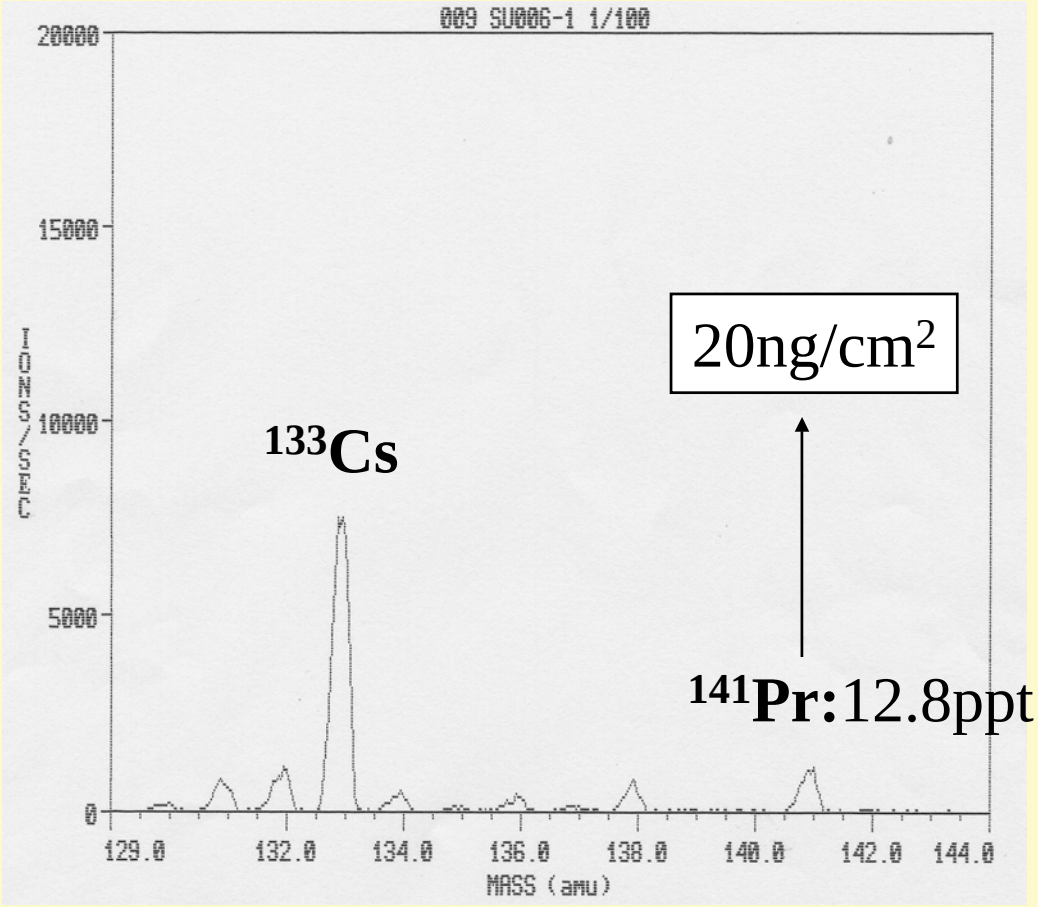
F.G.Data(1)



Ion counts(counts/sec)

Mass Number

F.G.Data(2)

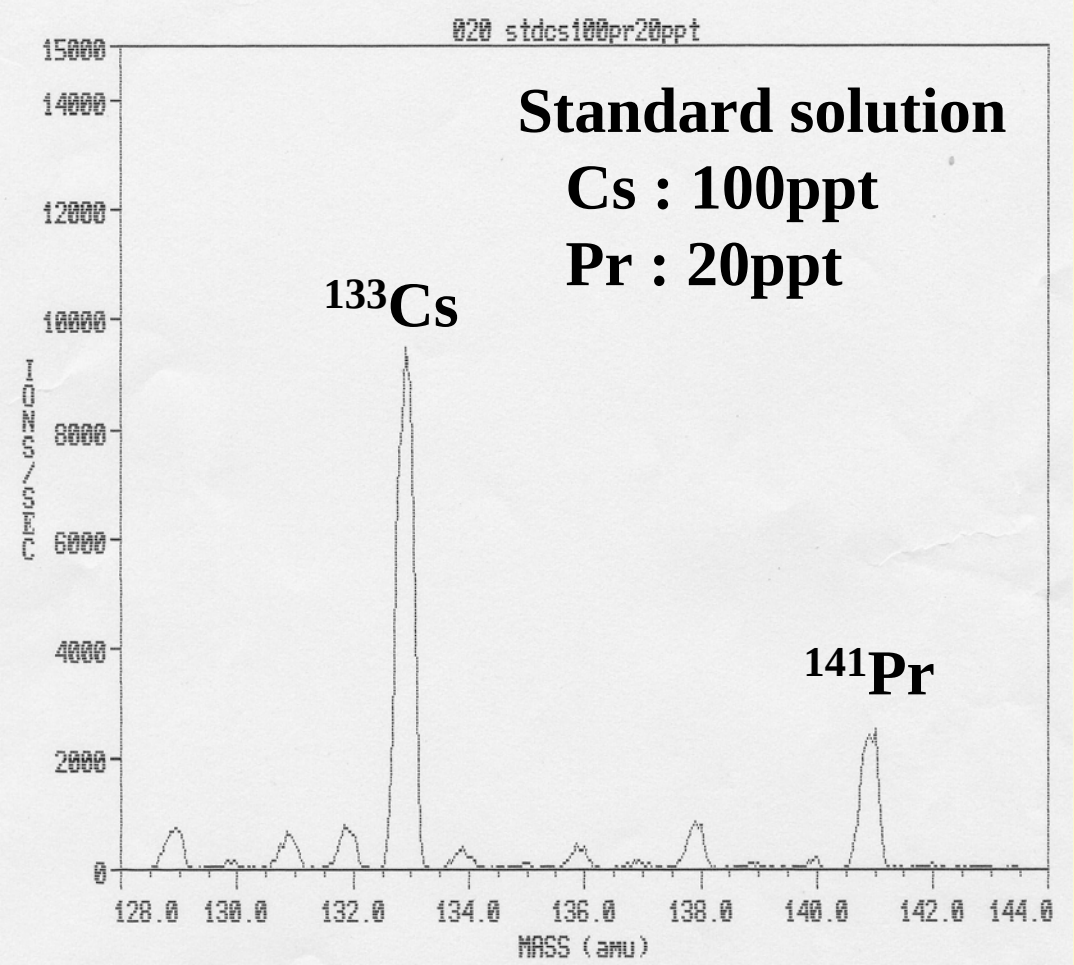


Ion counts(counts/sec)

Mass Number

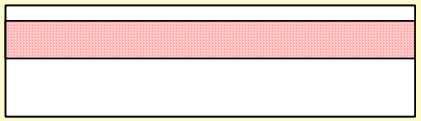
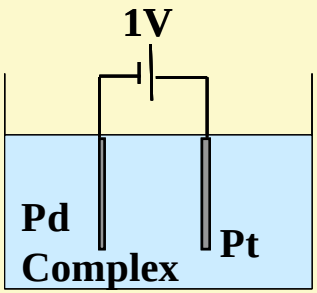
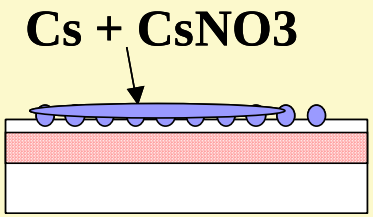
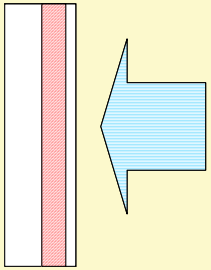
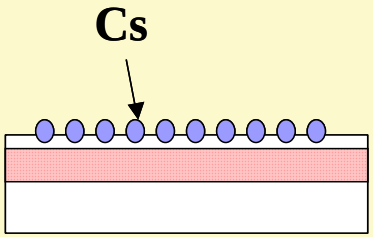
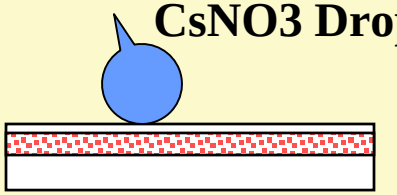
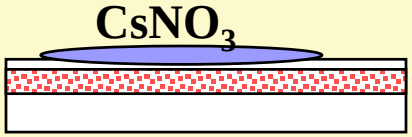
Analysis of the Standard Solution

Ion counts(counts/sec)



Mass Number

Transmutation Dependence on the Element Addition Method

Pd Complex 	 1mM $CsNO_3/D_2O$ solution		Positive
	 Pd Complex		Positive
			Negative

Conjecture on D distribution in the Pd Complex

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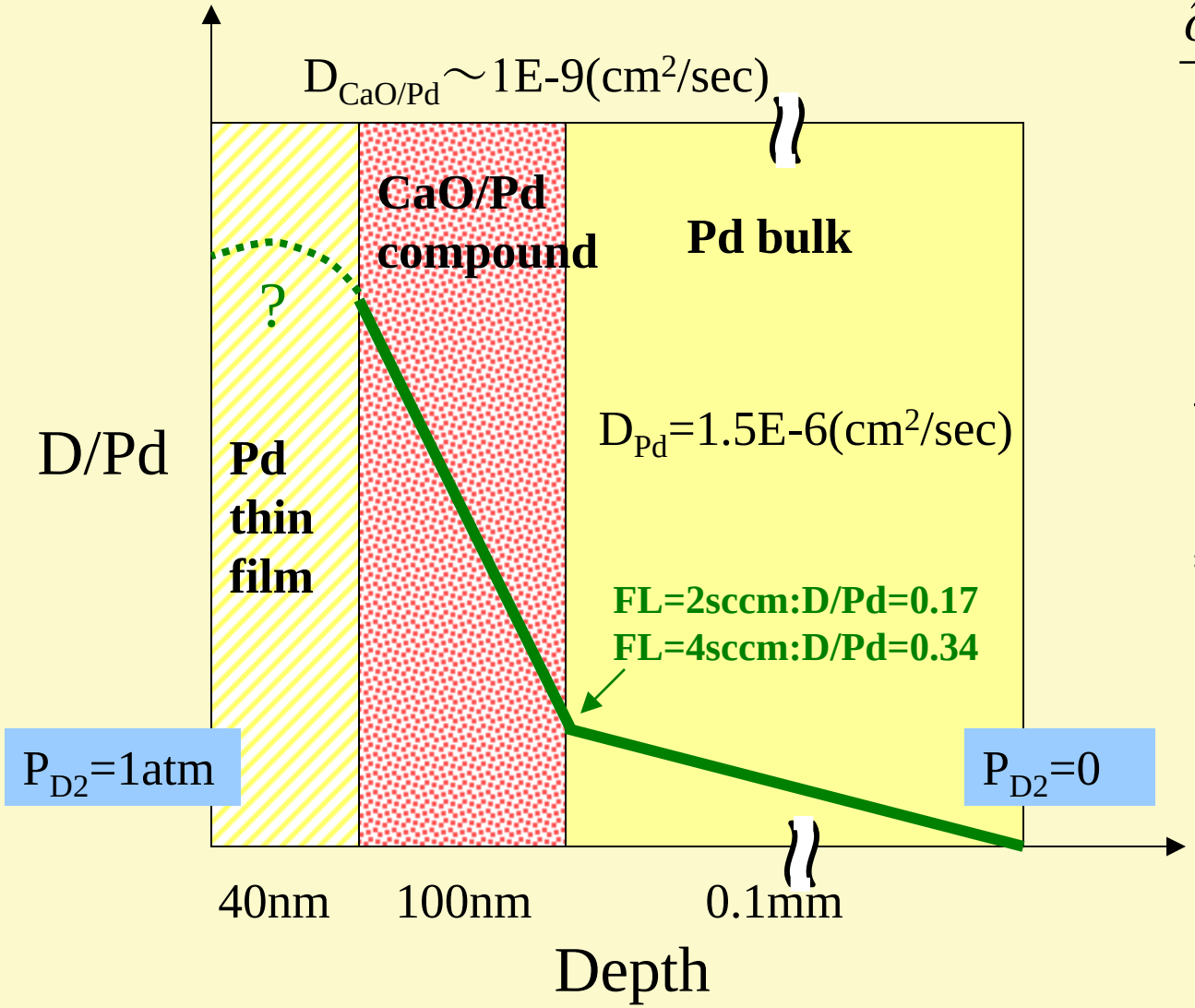


$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} \qquad \frac{\partial}{\partial t} = 0$$

$$Q = A \cdot J = -A \cdot D\frac{\partial C}{\partial x}$$

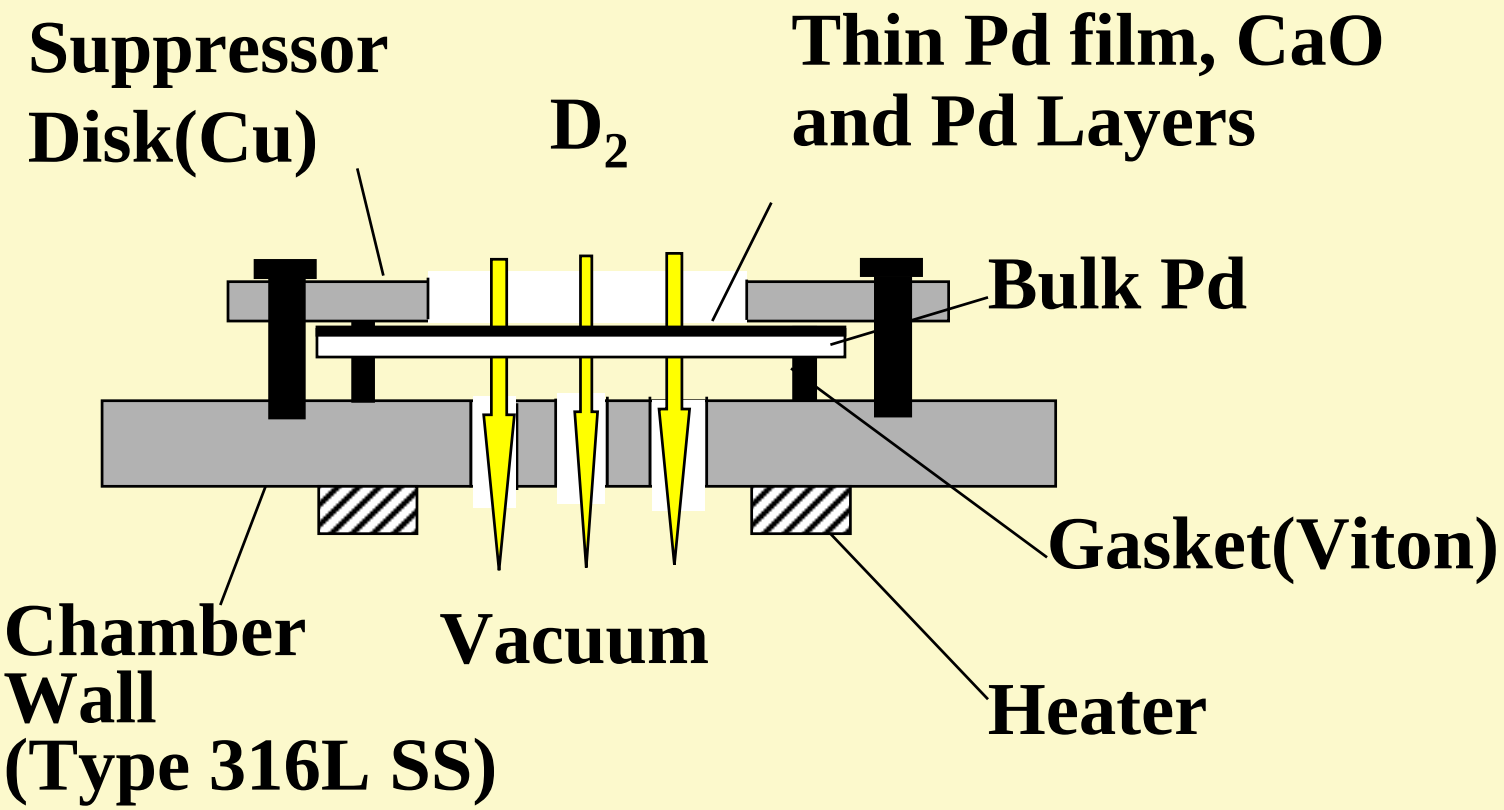
$$-A \cdot D_{CaO/Pd} \cdot \left.\frac{\partial C}{\partial x}\right|_{CaO/Pd}$$

$$= -A \cdot D_{Pd} \cdot \left.\frac{\partial C}{\partial x}\right|_{Pd}$$

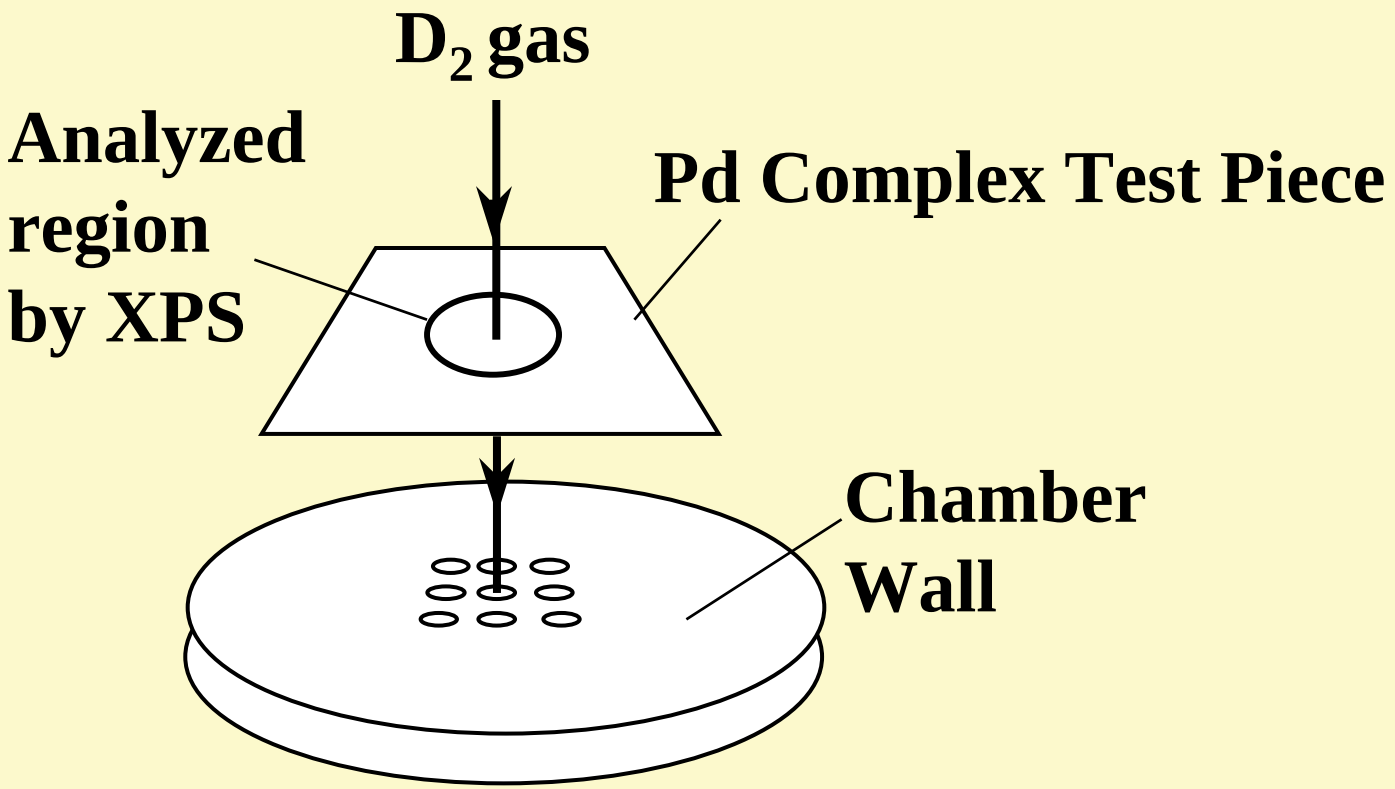
$$D_{CaO/Pd} \le D_{Pd}$$

$$\left.\frac{\partial C}{\partial x}\right|_{CaO/Pd} \ge \left.\frac{\partial C}{\partial x}\right|_{Pd}$$

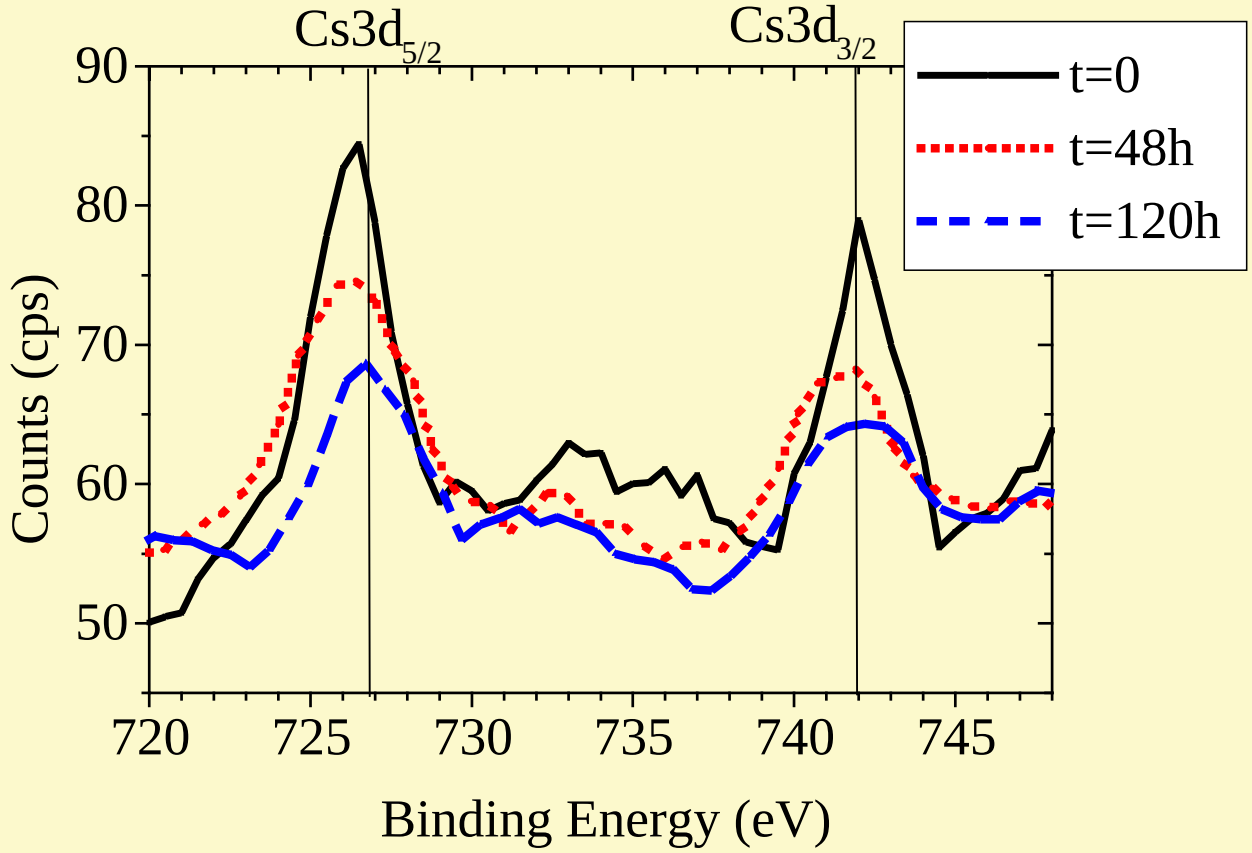
Cross Sectional View



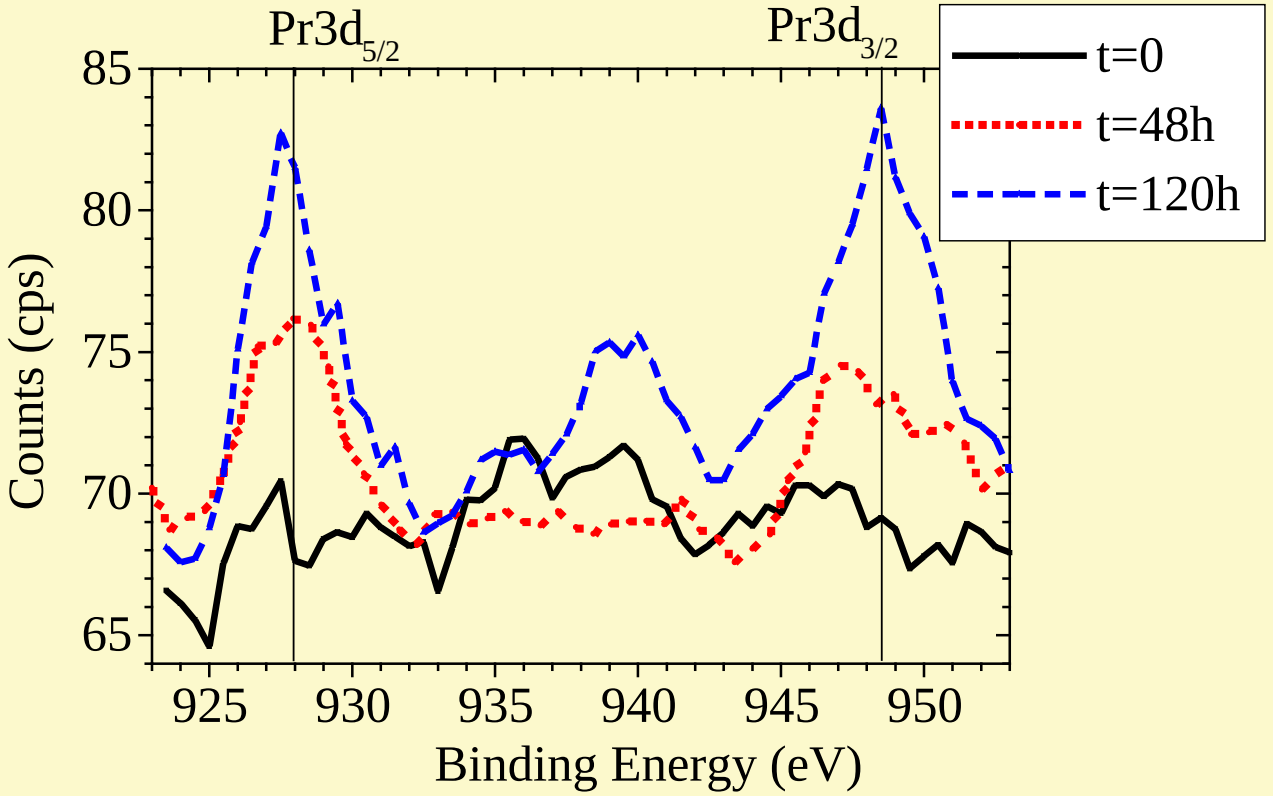
Passage of D_2 Gas



Change of XPS Spectrum of Cs



Change of XPS Spectrum of Pr



Time Variation in number of Cs and Pr atoms in the case of D_2 Permeation through thin film and bulk Pd with added Cs

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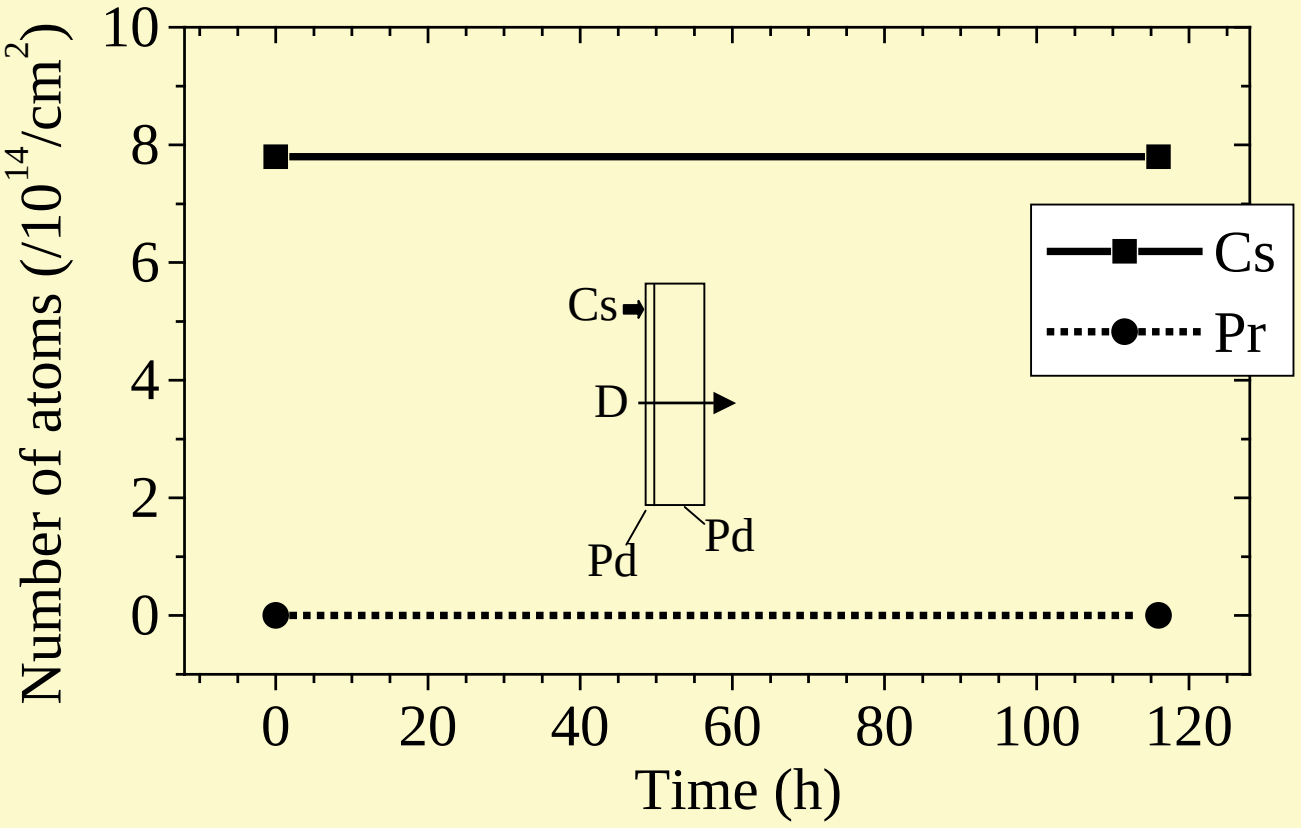
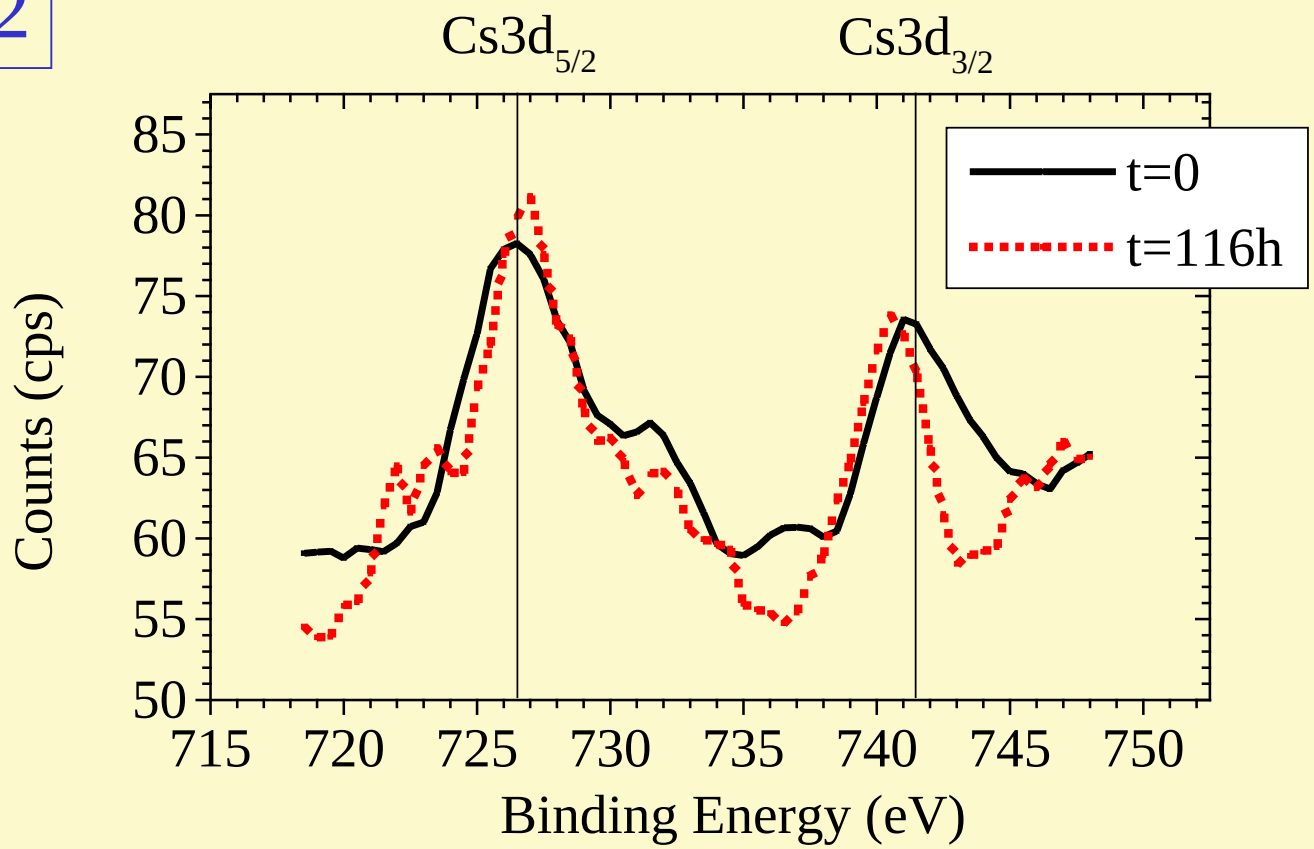


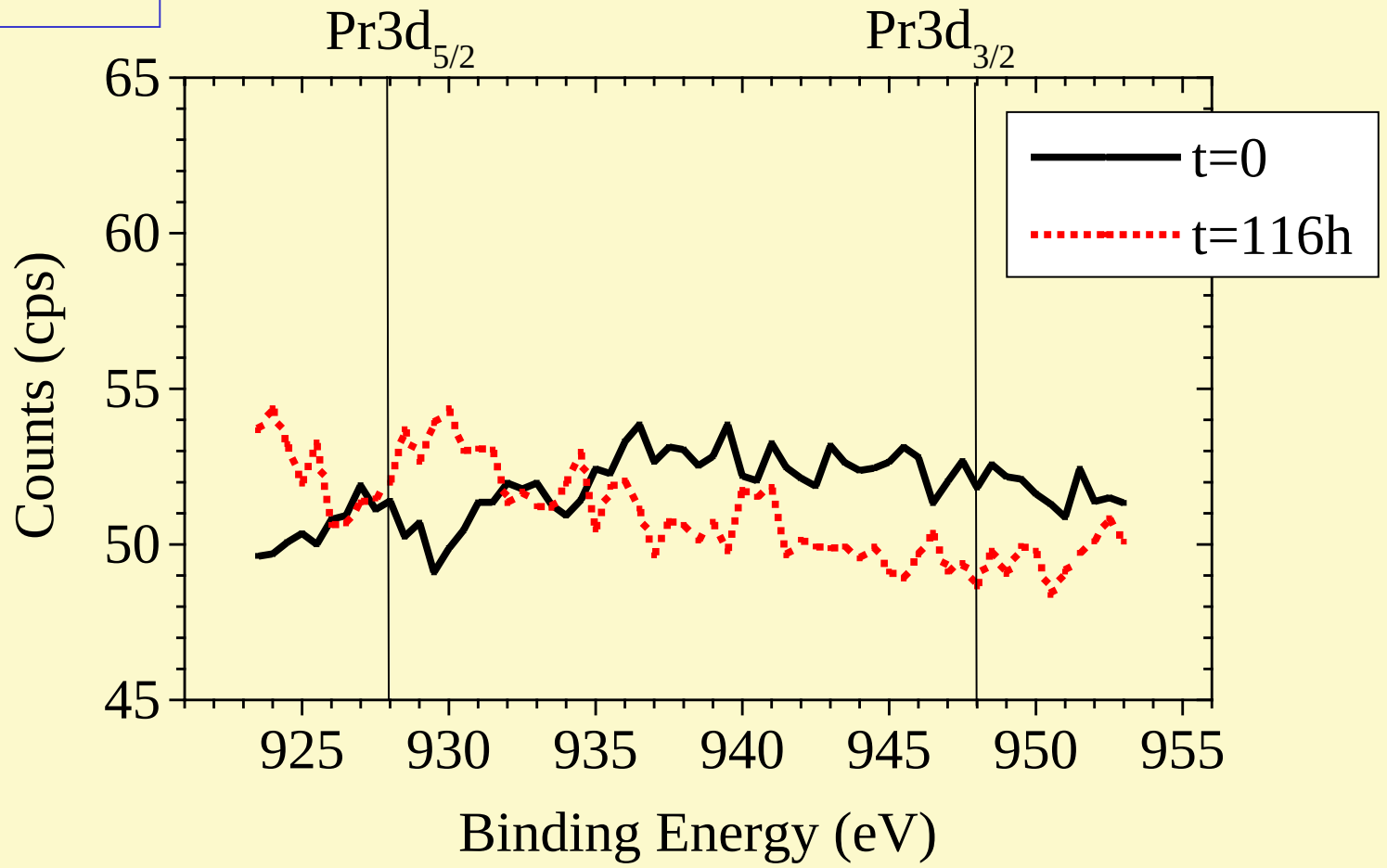
XPS Spectrum of Cs (No CaO)

Pd/Pd/D2

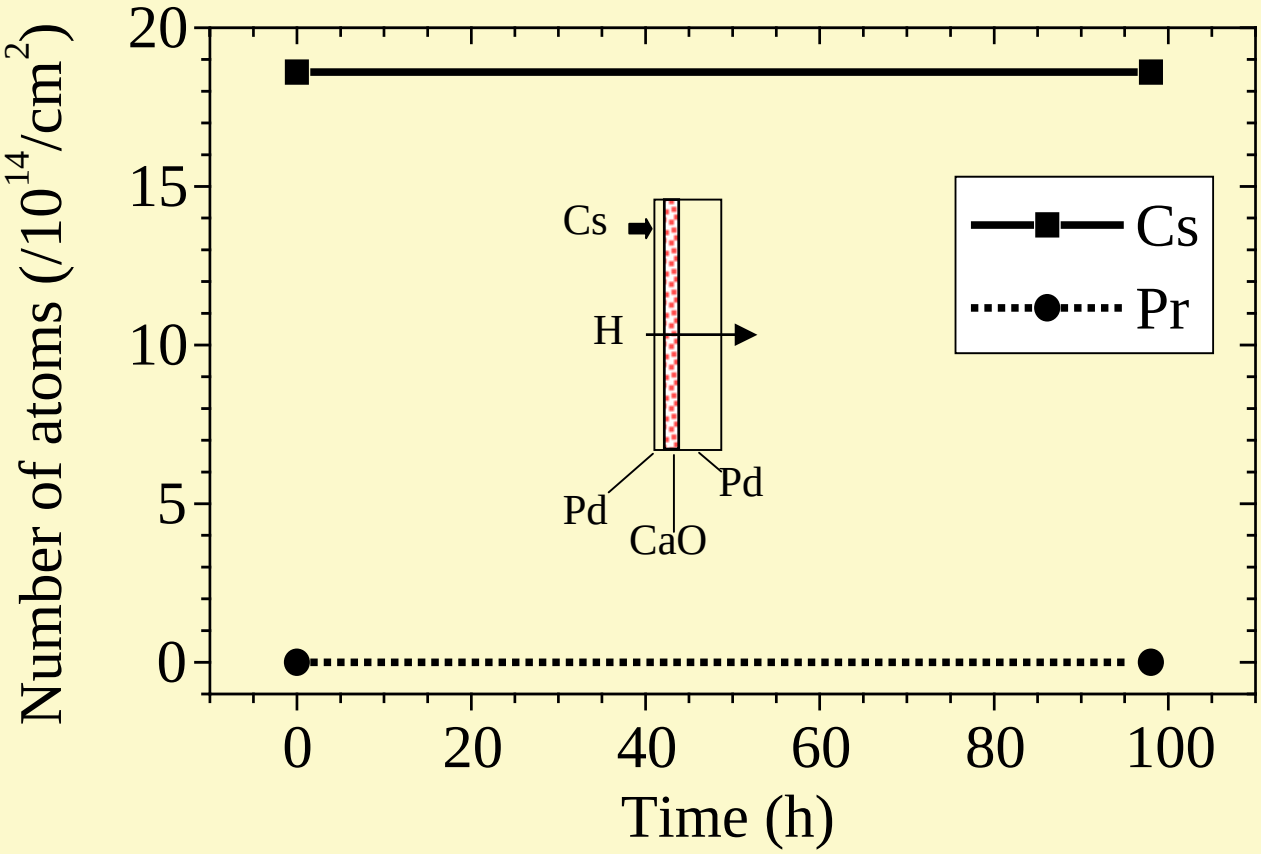


XPS Spectrum of Pr (No CaO)

Pd/Pd/D2

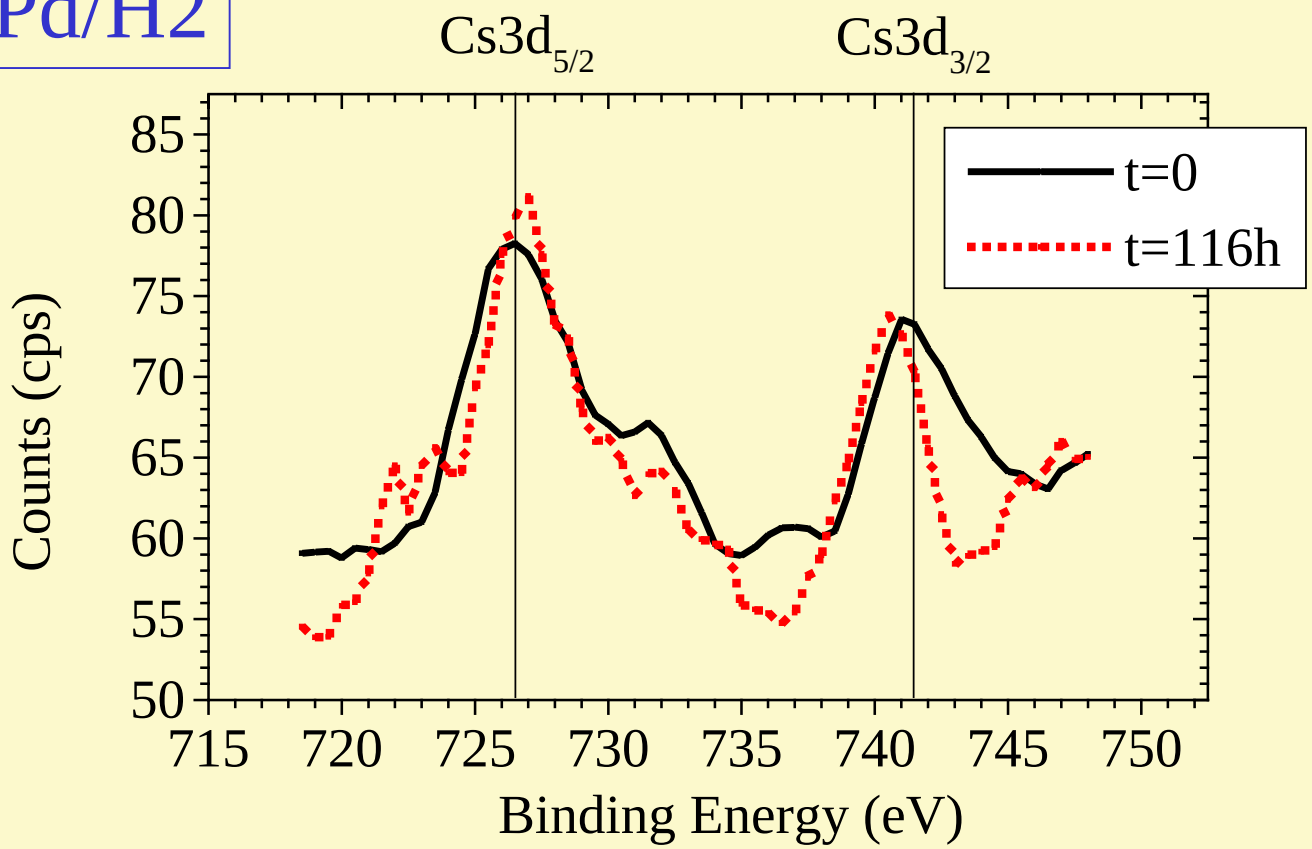


Time Variation in number of Cs and Pr atoms in the case of H_2 Permeation through Pd Complex with added Cs



XPS Spectrum of Cs(H_2 Permeation)

Pd/CaO/Pd/H2



XPS Spectrum of Pr (H_2 Permeation)

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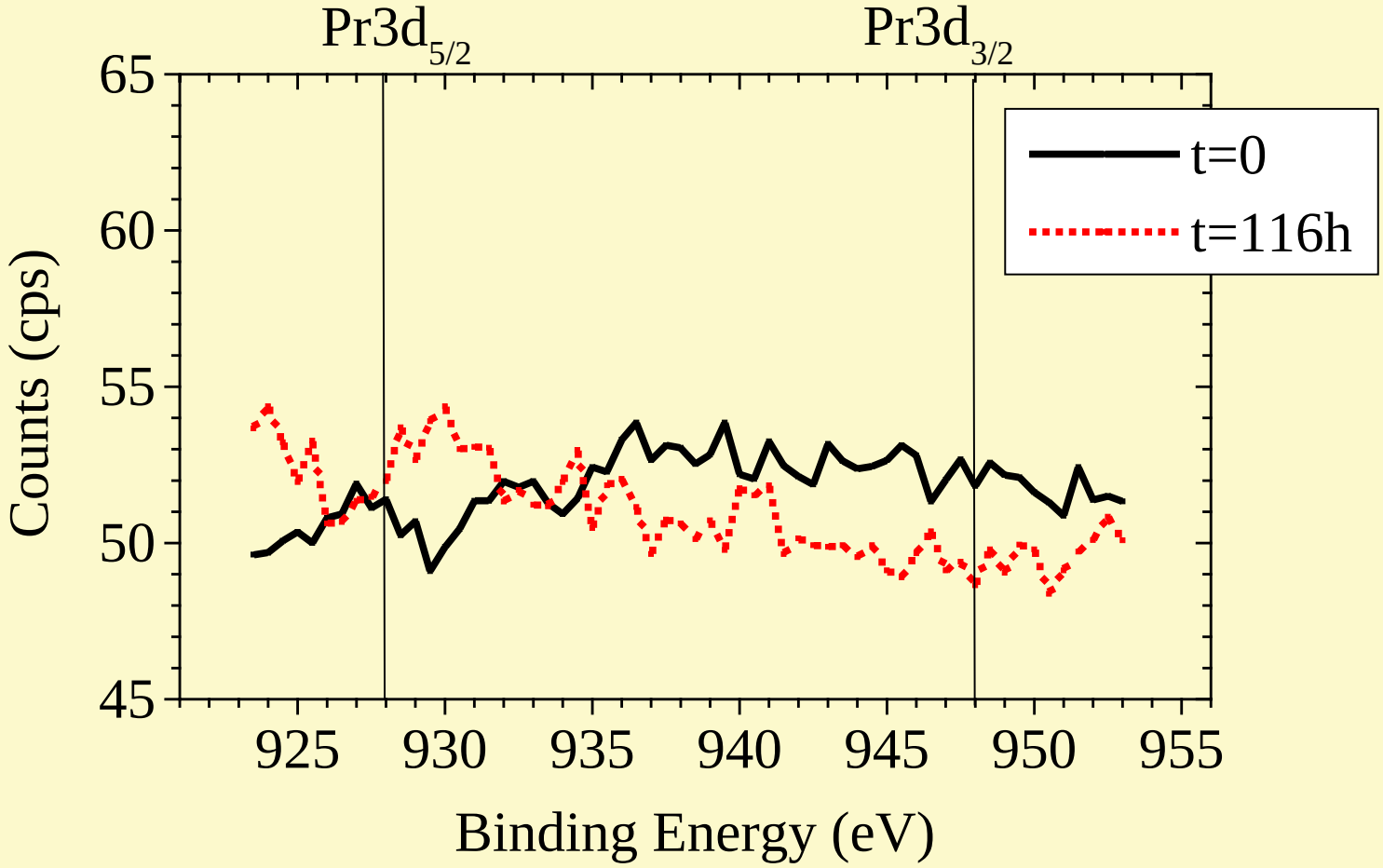
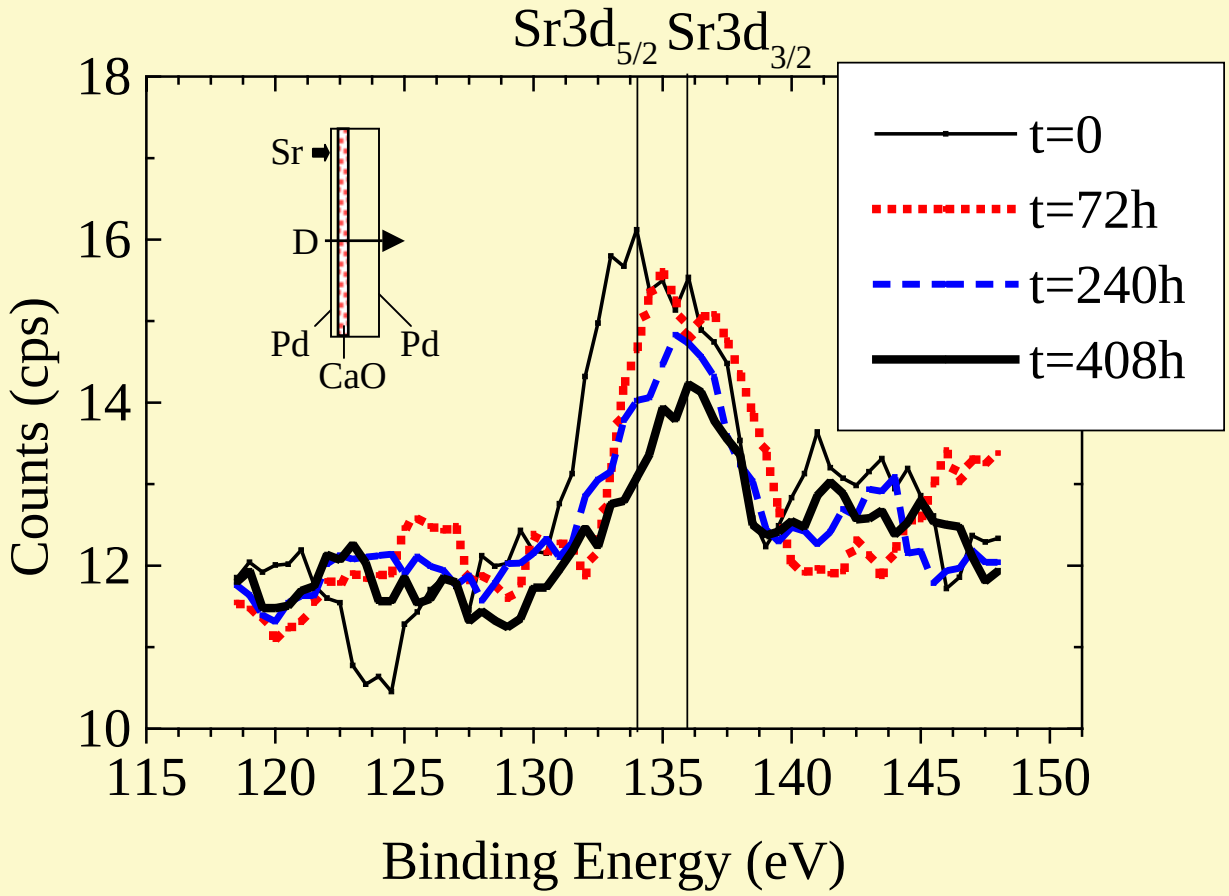
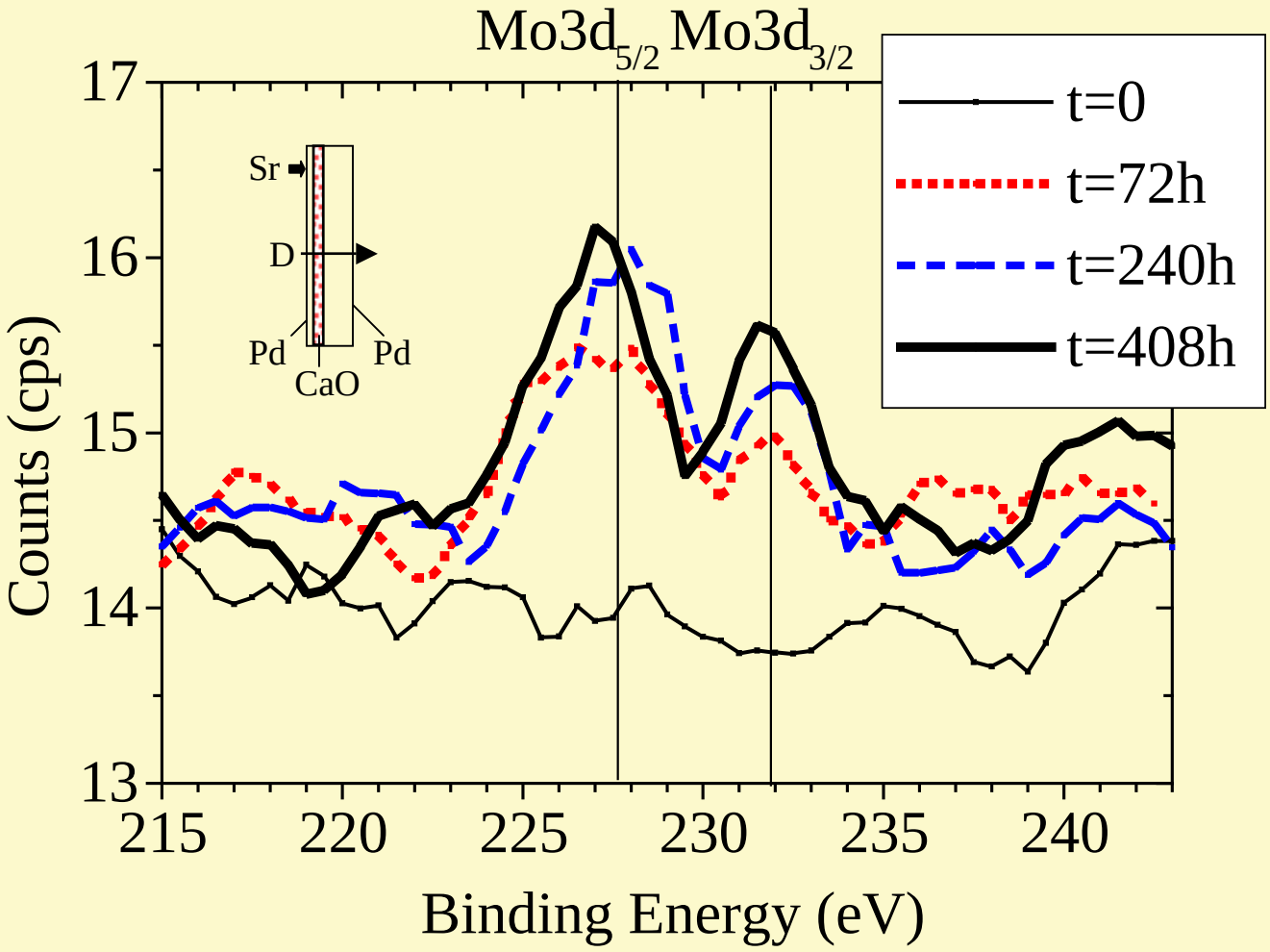
Pd/CaO/Pd/H2



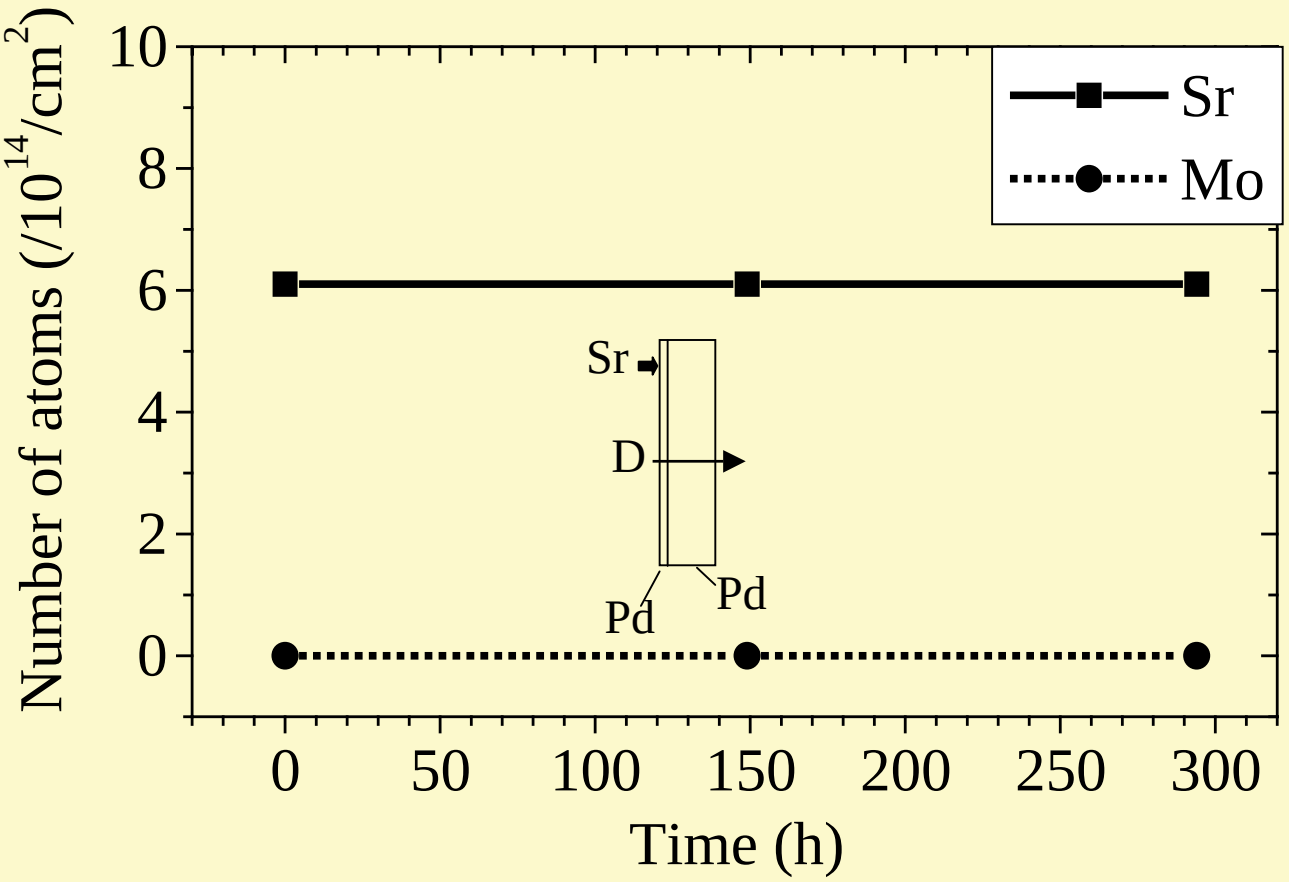
Change of XPS Spectrum of Sr



Change of XPS Spectrum of Mo

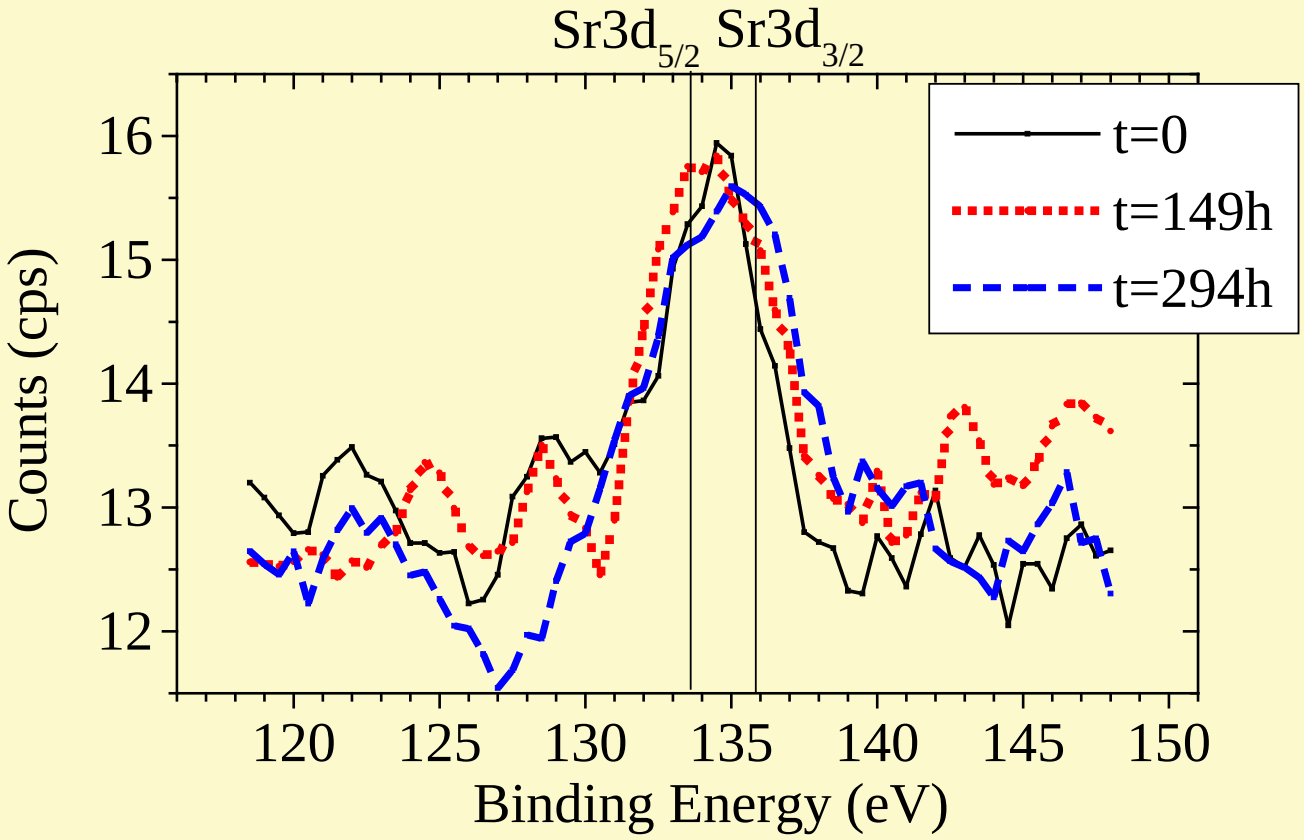


Time Variation in number of Sr and Mo atoms in the case of D_2 Permeation through thin film and bulk Pd with added Sr



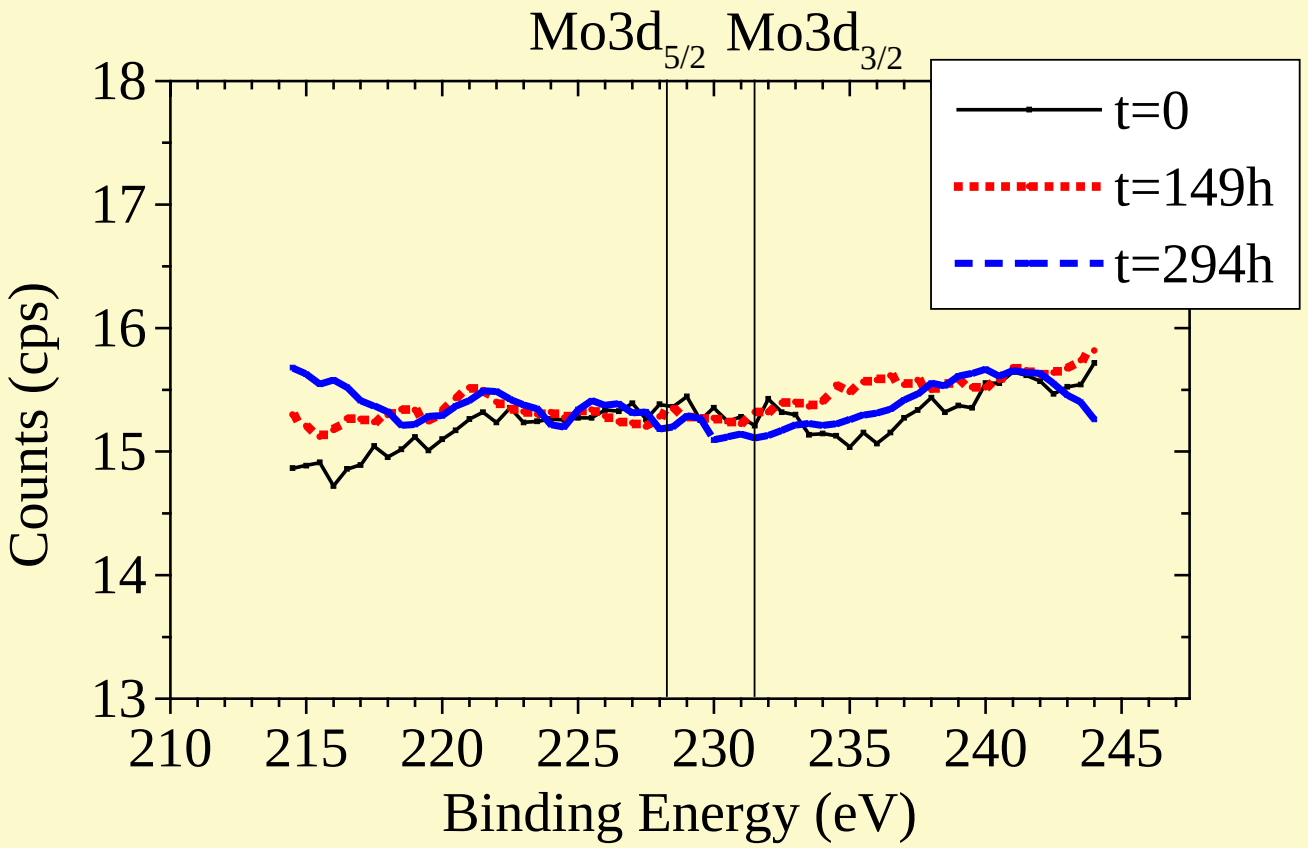
XPS Spectrum of Sr (No CaO)

Pd/Pd/D2

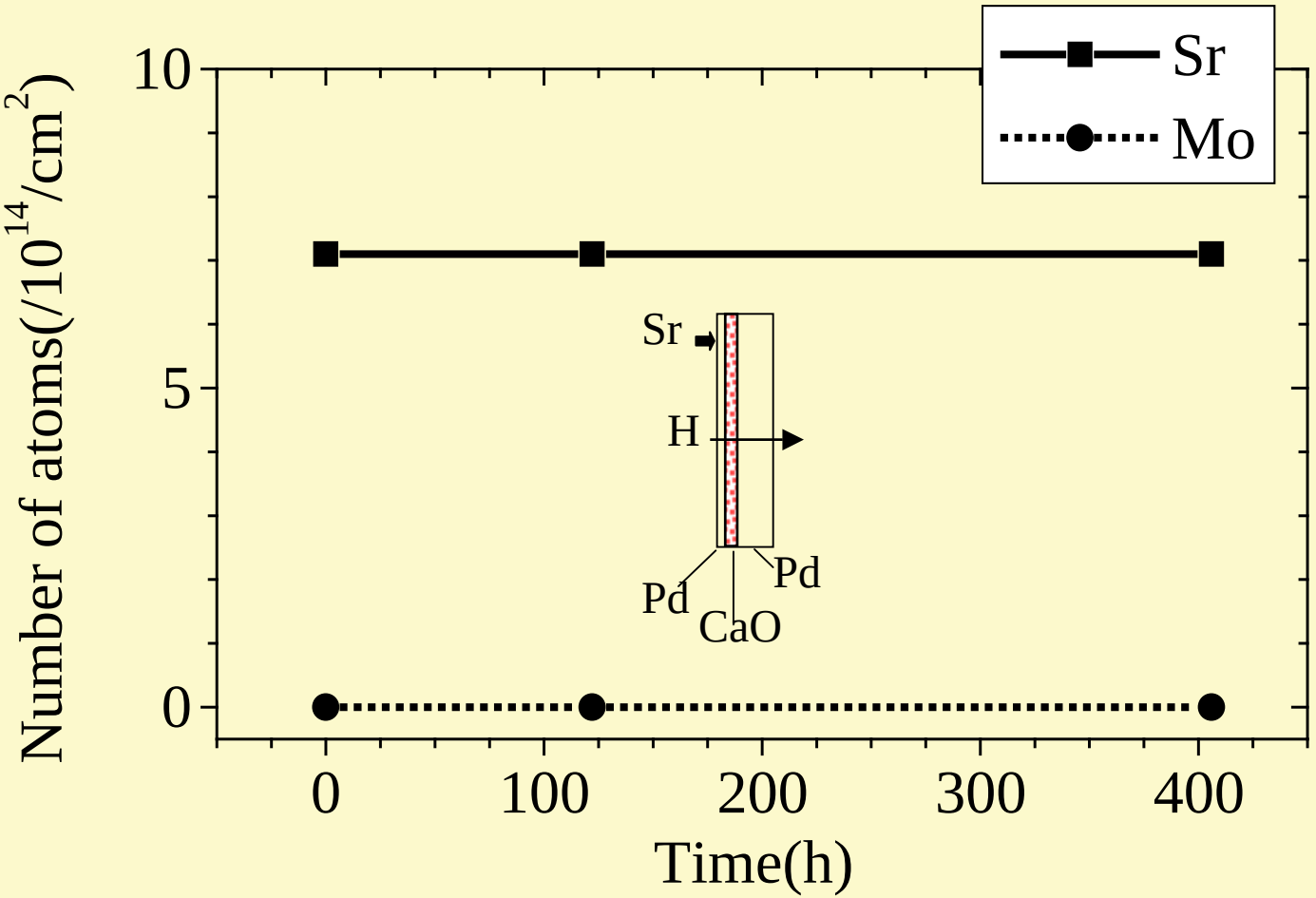


XPS Spectrum of Mo (No CaO)

Pd/Pd/D2



Time Variation in number of Sr and Mo atoms in the case of H_2 Permeation through Pd Complex with added Sr



XPS Spectrum of Sr (H_2 Permeation)

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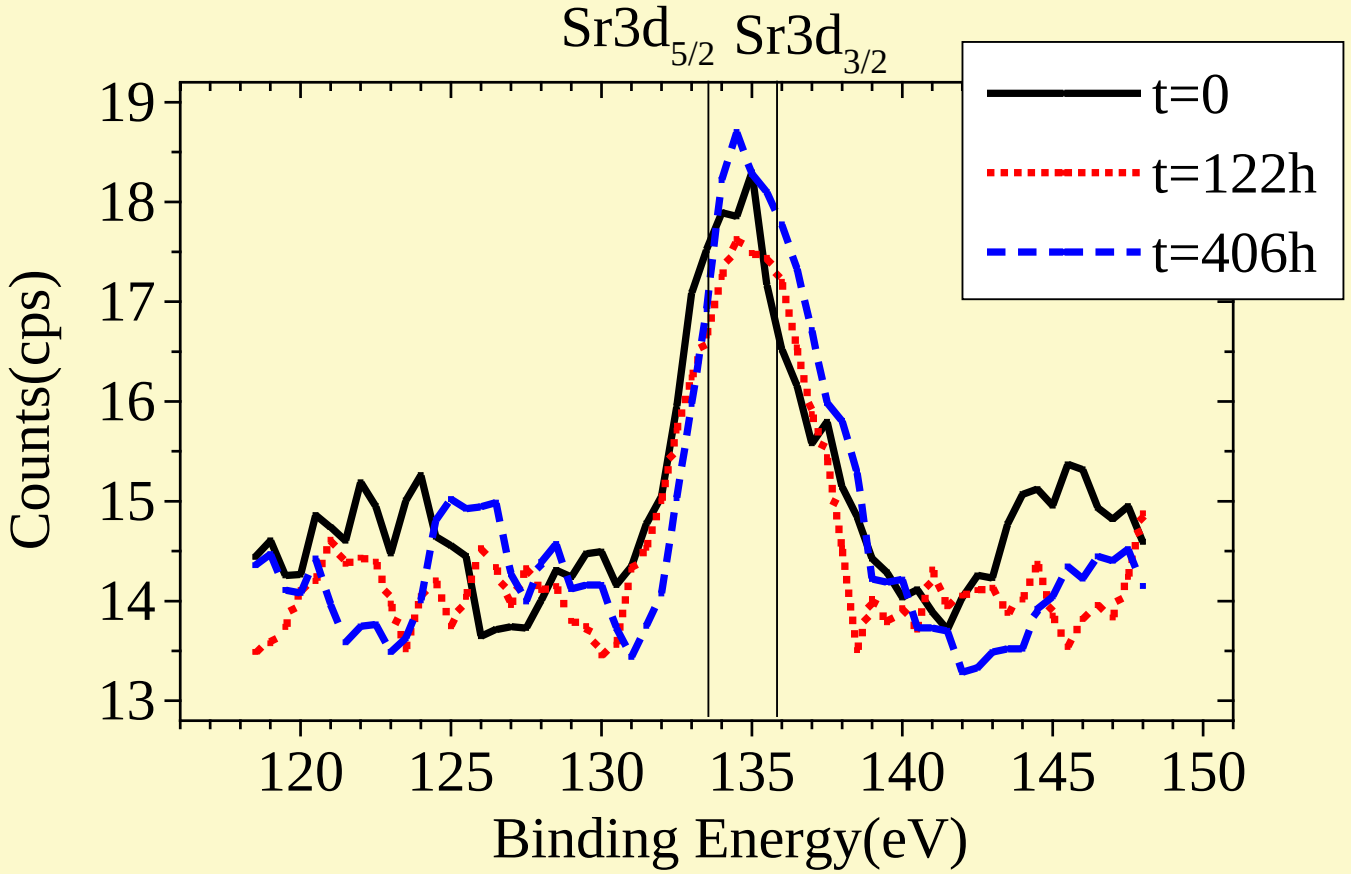
Pd/CaO/Pd/H2



XPS Spectrum of Mo (H_2 Permeation)

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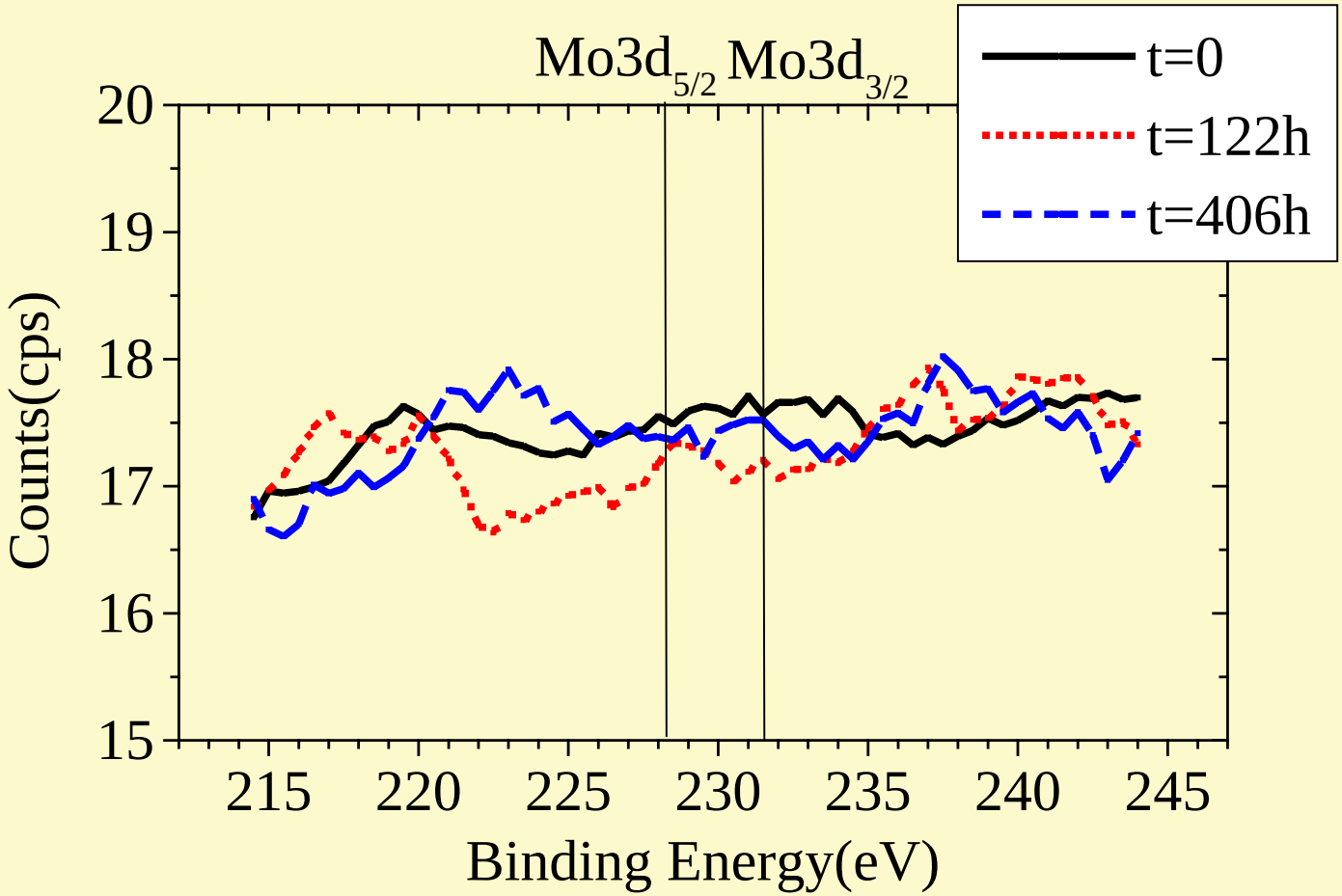
Pd/CaO/Pd/H2



Separation of the Products and Contaminants(3)

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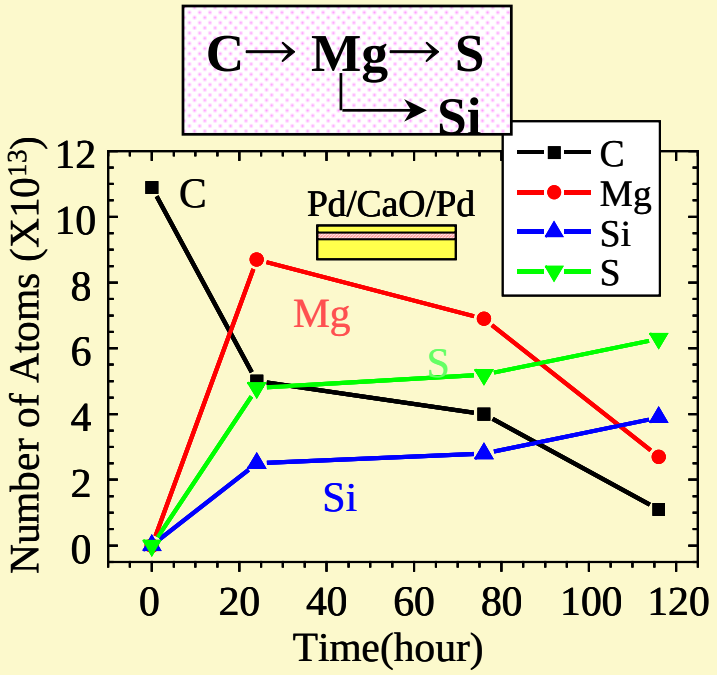
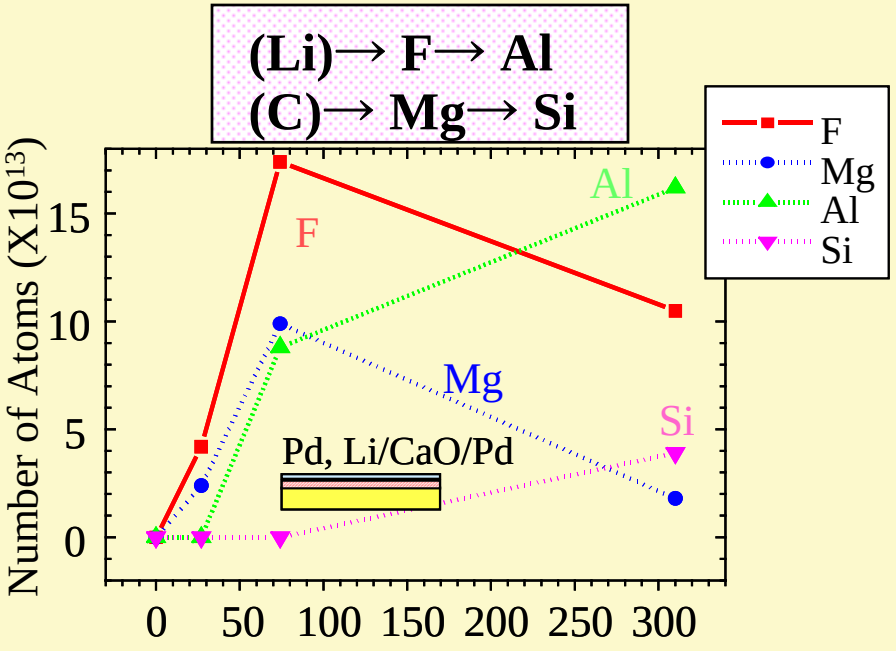
3. Variation of Detected Elements depending on the given Elements

Cs→Pr, Sr→Mo Li→Al, C→S,Si





If the detected elements were contaminants, was it possible that the detected elements changed depending on the given elements?

The other observations

$$^{12}_{6}C \rightarrow ^{24}_{12}Mg \rightarrow ^{28}_{14}Si$$

$$^{12}_{6}C \rightarrow ^{24}_{12}Mg \rightarrow ^{32}_{16}S$$

$$^{7}_{3}Li \rightarrow ^{19}_{9}F \rightarrow ^{23}_{11}Na \rightarrow ^{27}_{13}Al$$

Necessary Conditions

Necessary Conditions to Induce Low Energy Nuclear Reactions

1. **Enough Deuterium Flux**
2. **Sufficient D on the Pd surface**
3. **Existence of a third element except Pd and D**

Experimental Results