Pd Complex

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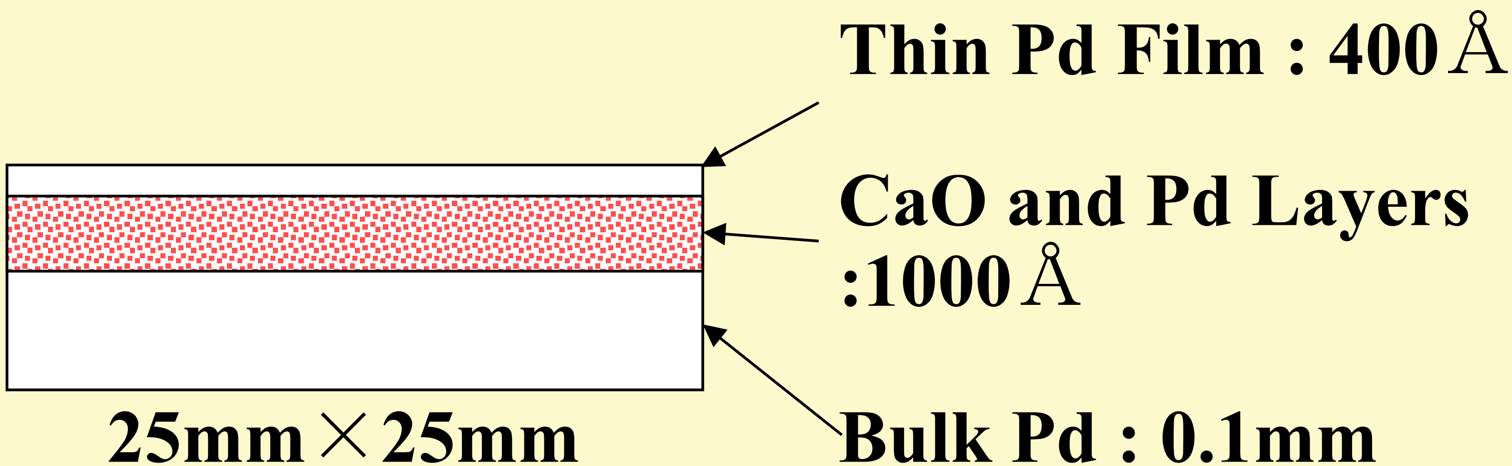
Pd complex containing CaO



Transmutation of Cs into Pr

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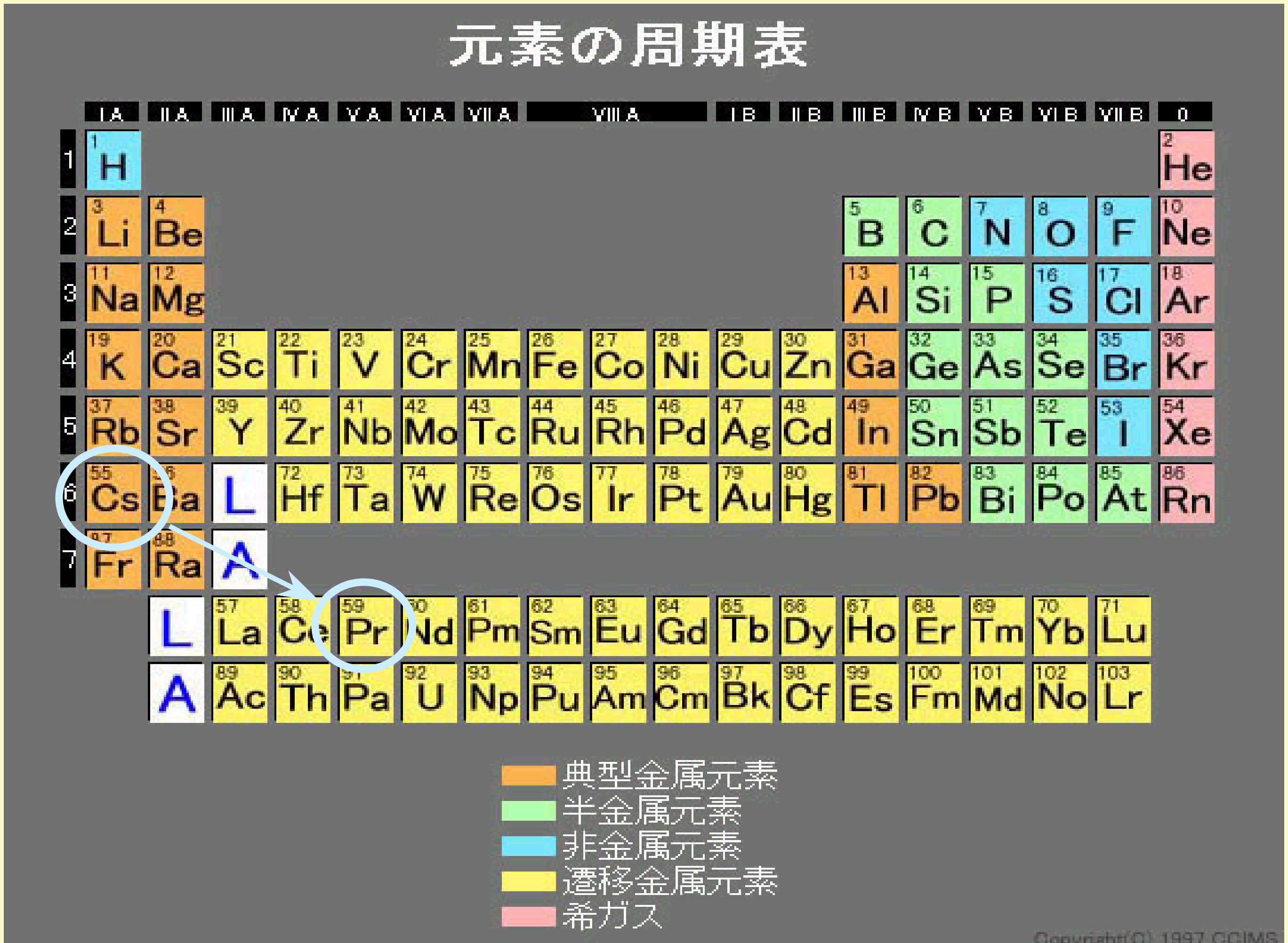

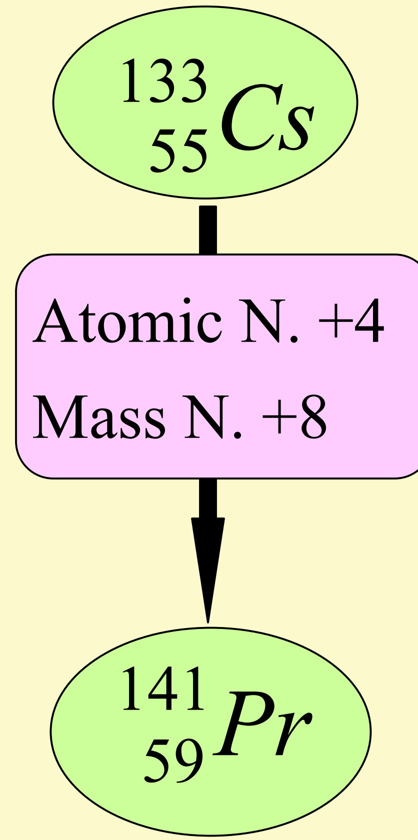

Transmutation of Sr into Mo

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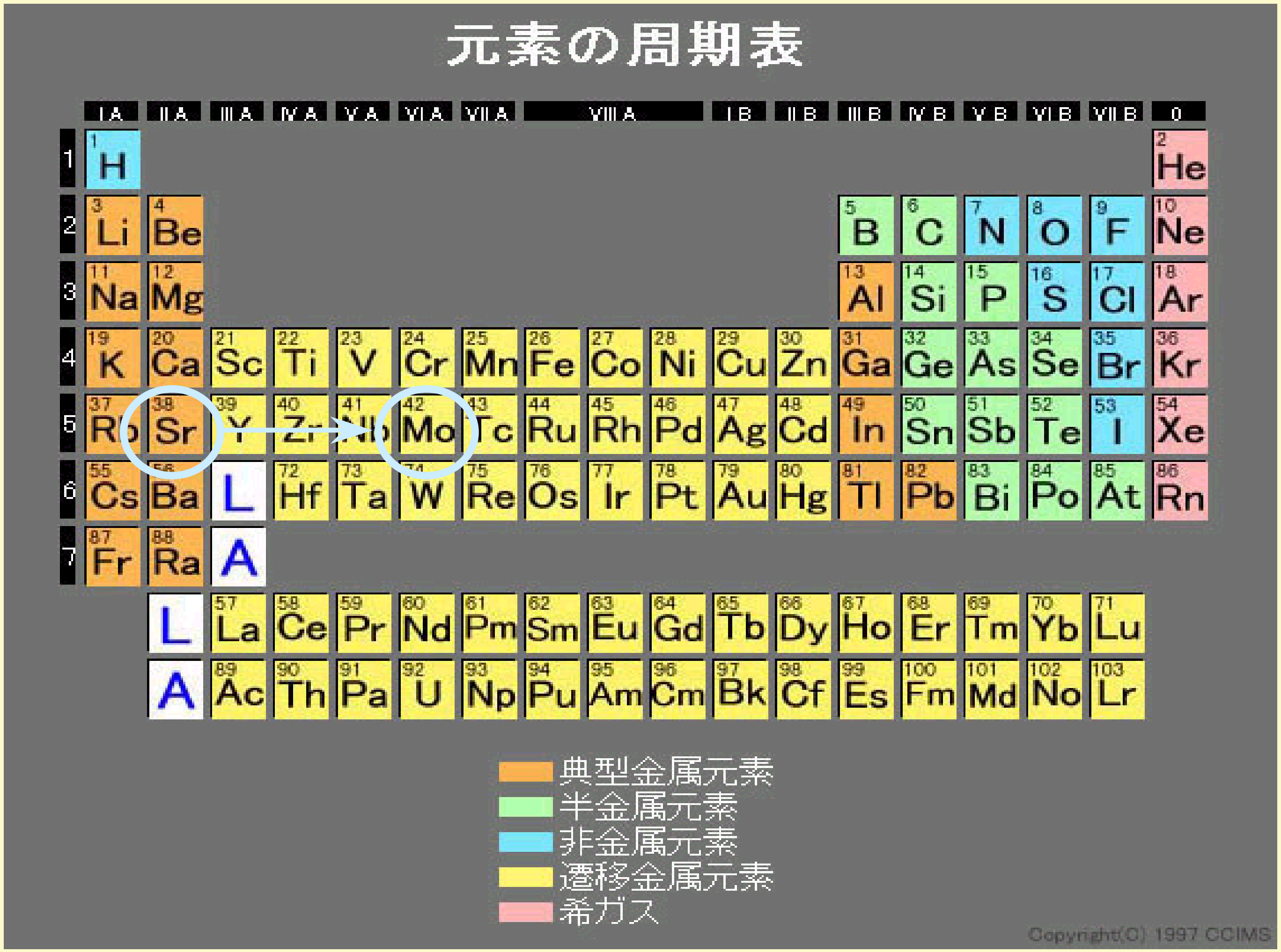

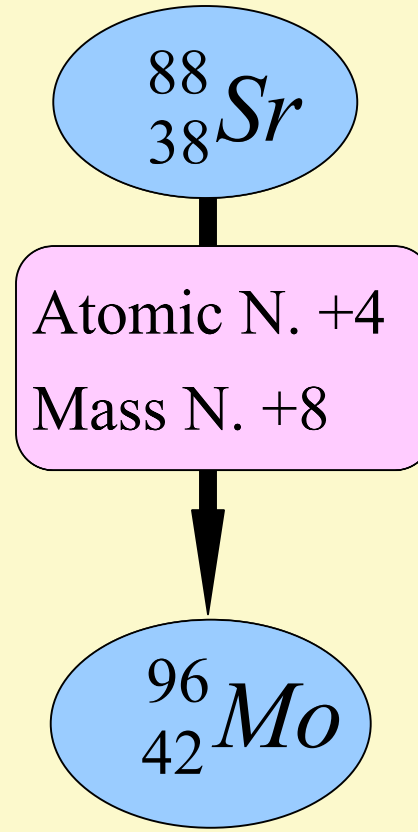

Transmutation of Ba into Sm

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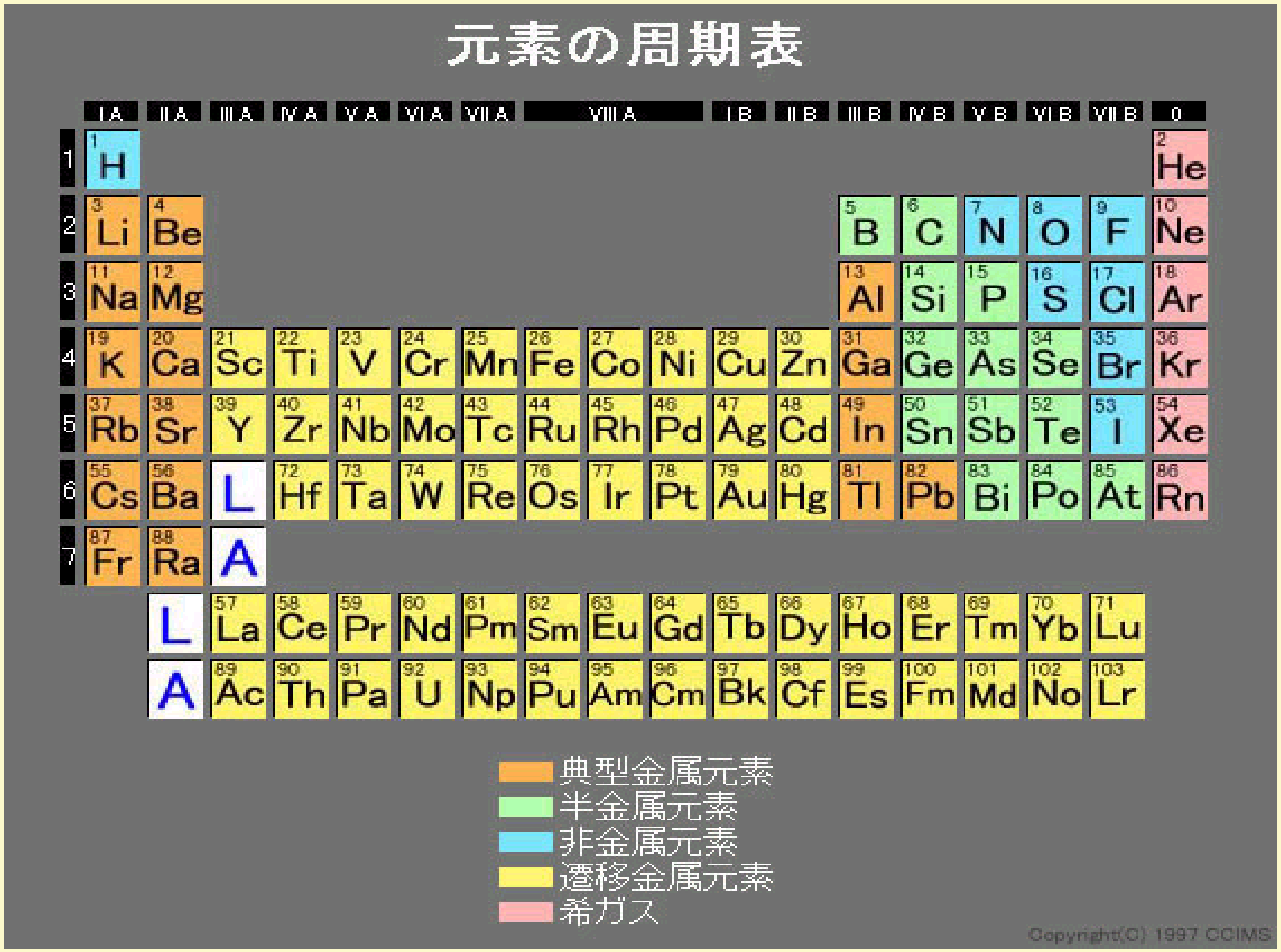
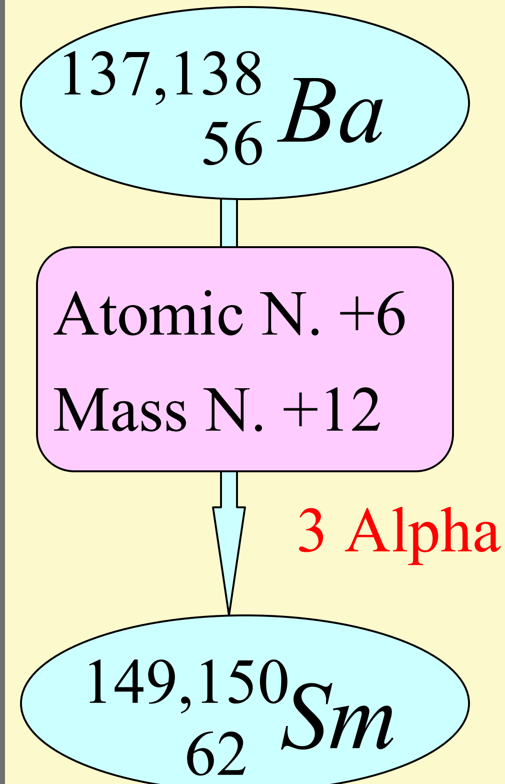


元素の周期表

	IA	IIA	IIIA	IVA	VA	VIA	VIIA	VIIIA			IB	IIB	IIIB	IVB	VB	VIB	VIIB	0
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	L	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	A															
		L	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	
		A	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr	

典型金属元素
半金属元素
非金属元素
遷移金属元素
希ガス



Isotopic Anomaly of the Detected Mo

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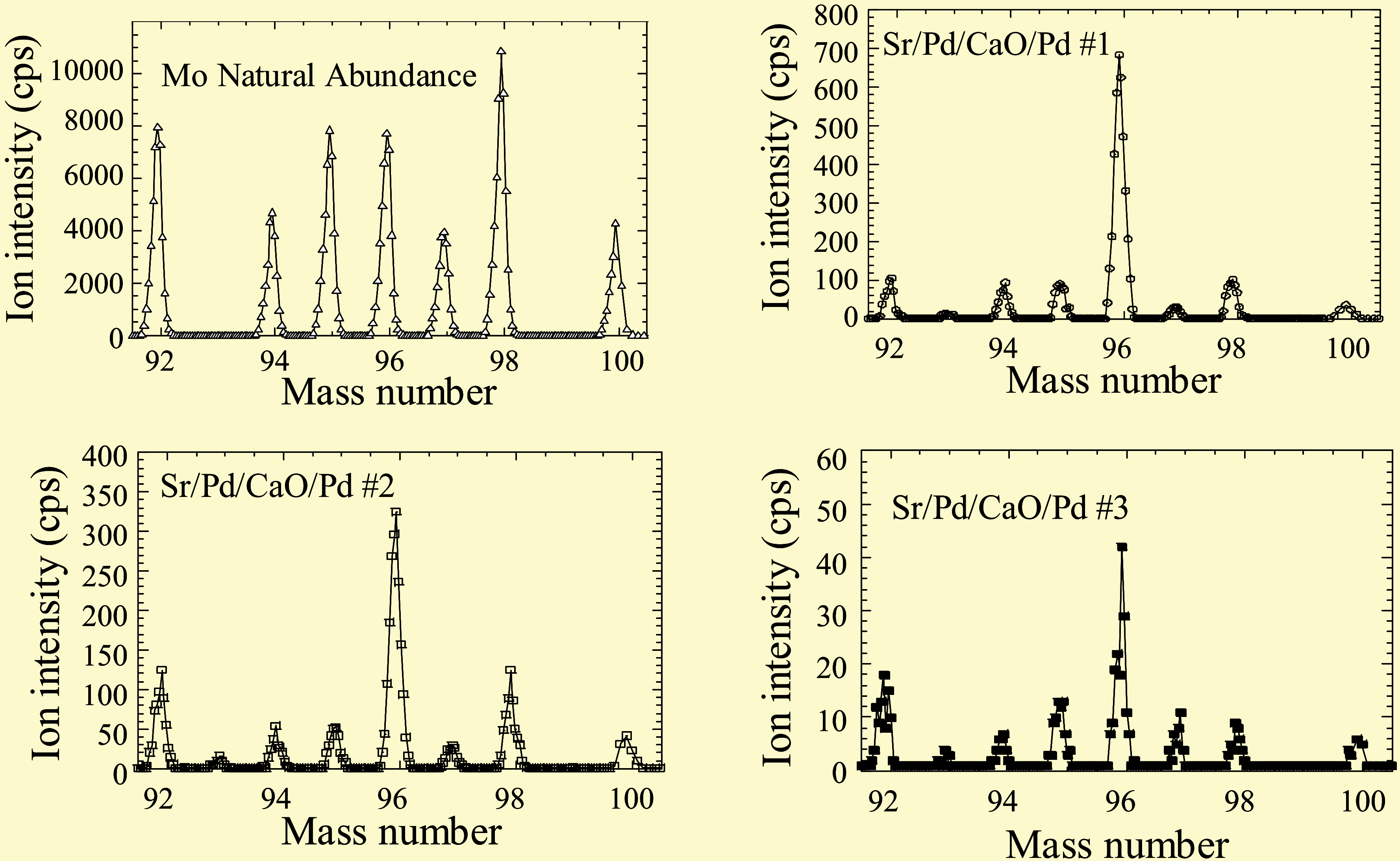

Evaluation of D_2 Gas Flow Rate

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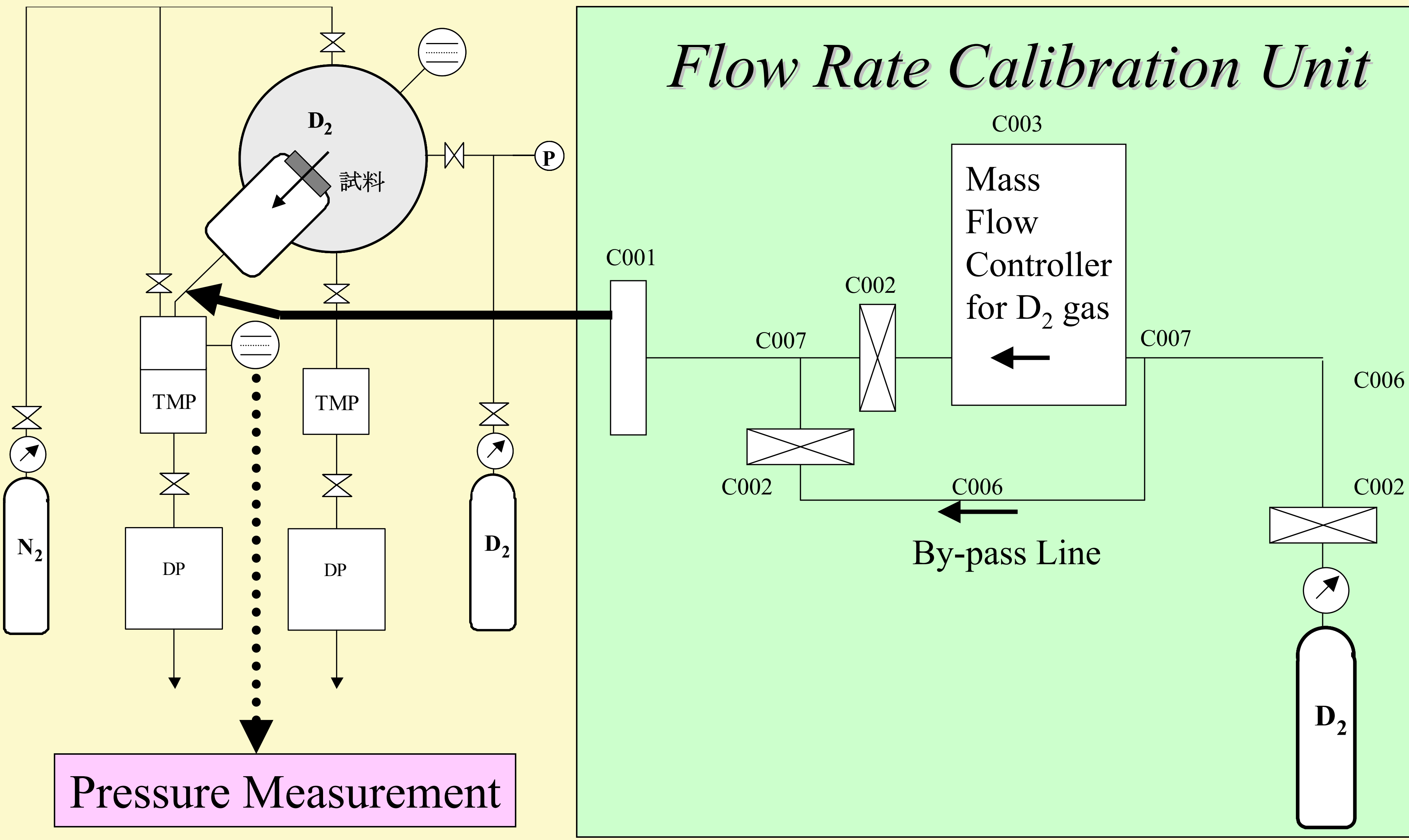

Identification of Pr by TOF-SIMS(2)

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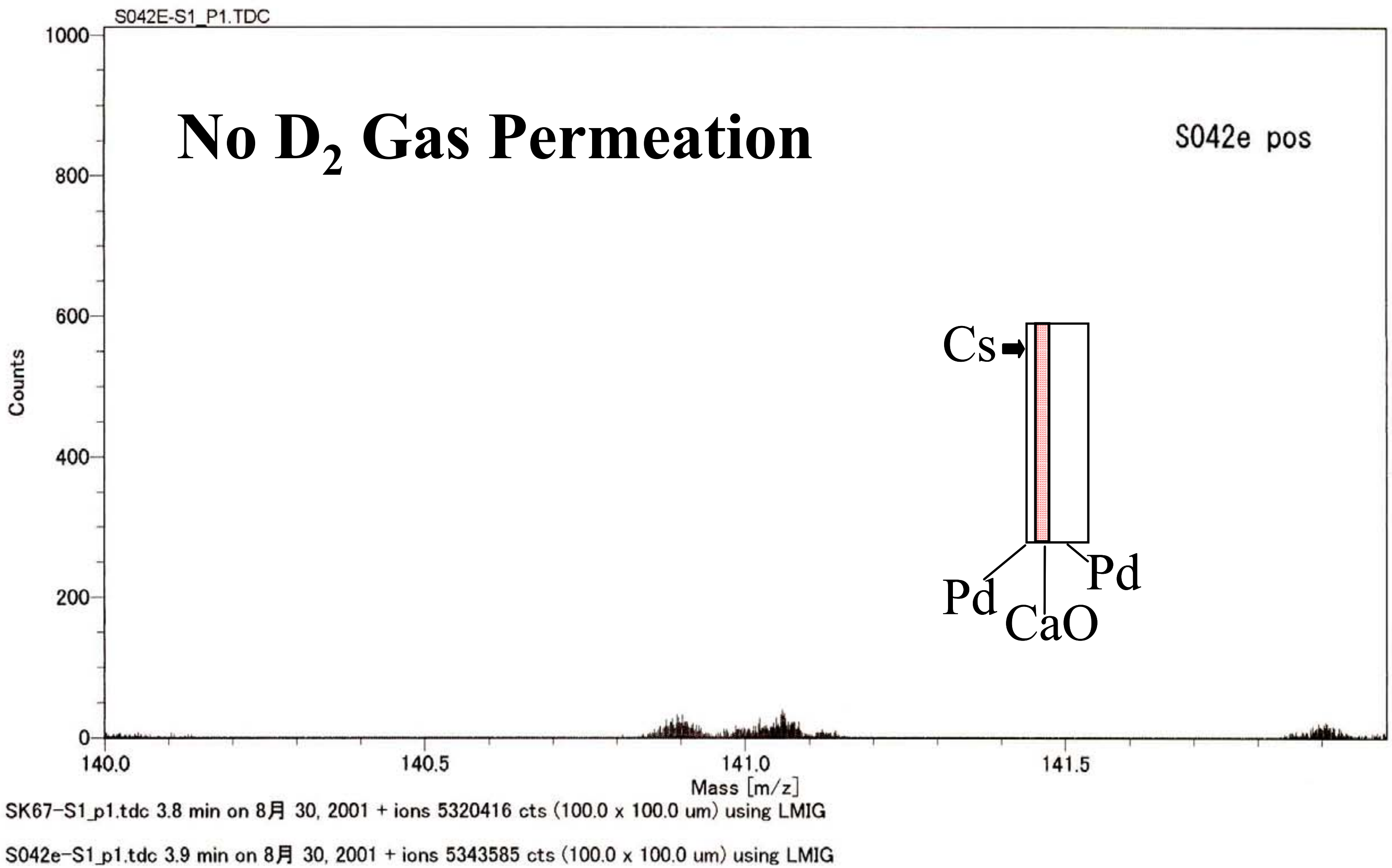
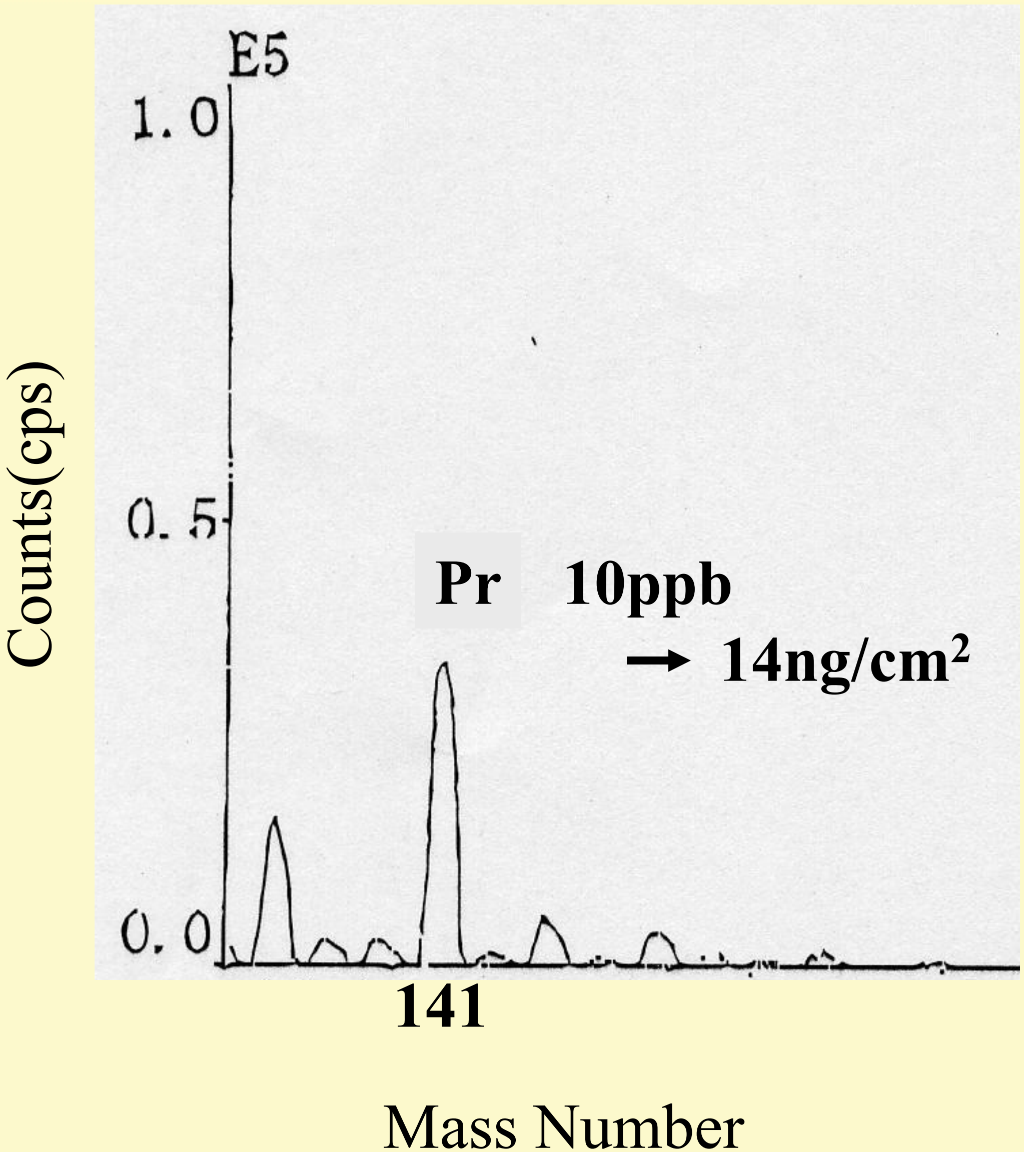
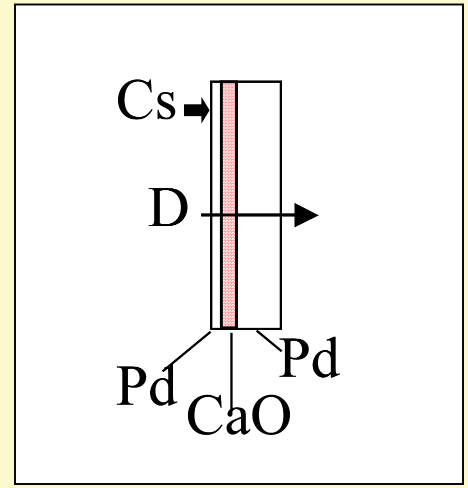


TOF-SIMS device (TRIFT™ Ⅱ;ULVAC-PHI)

Cs添加Pd多層膜重水素透過後





→ Prを検出、定量

Transmutation of Tm into Ta

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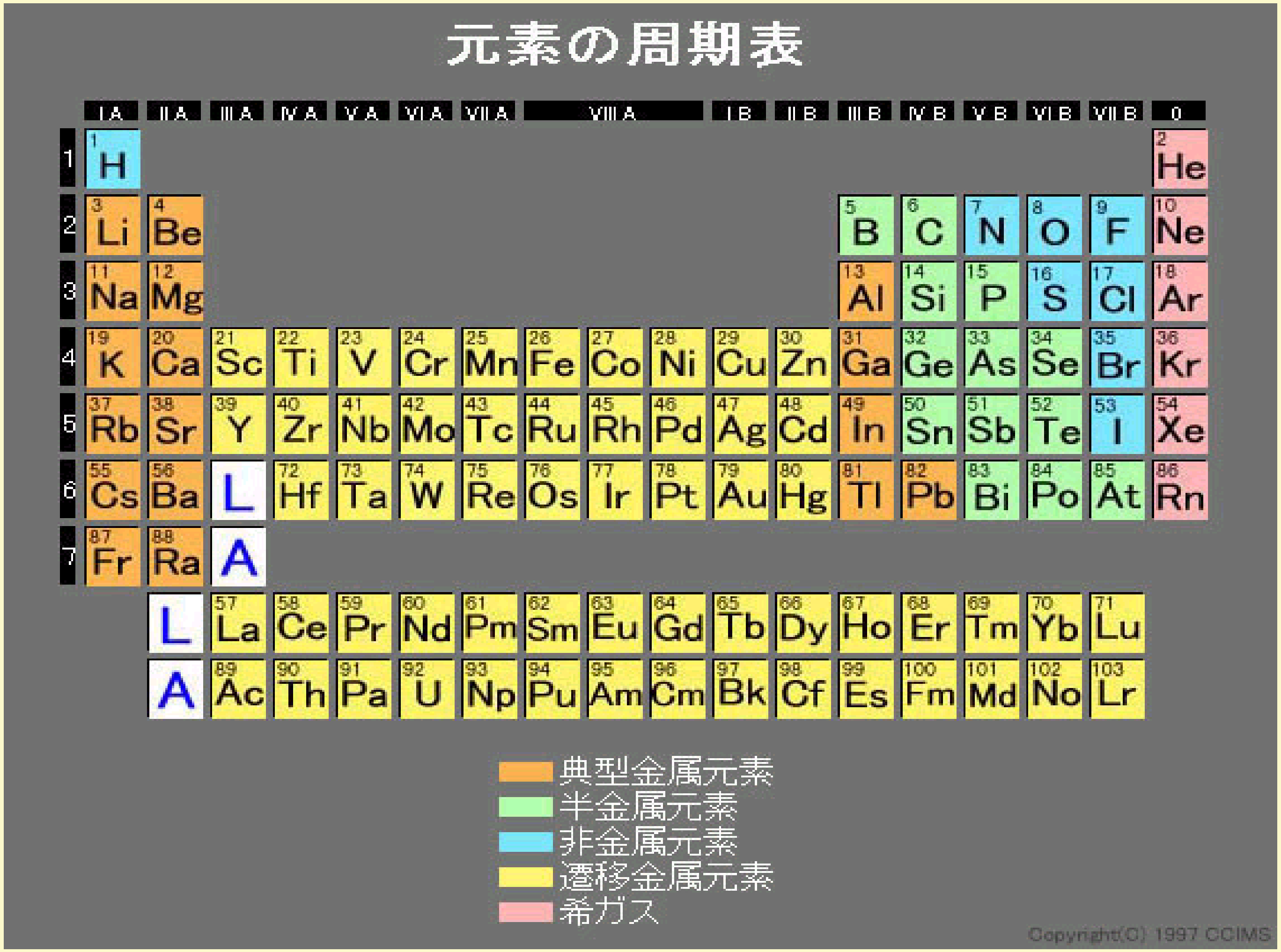
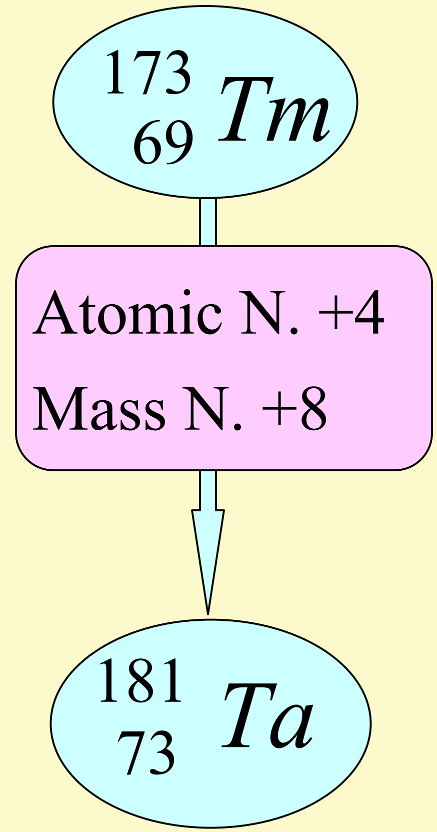
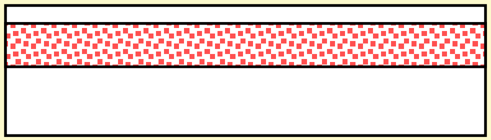
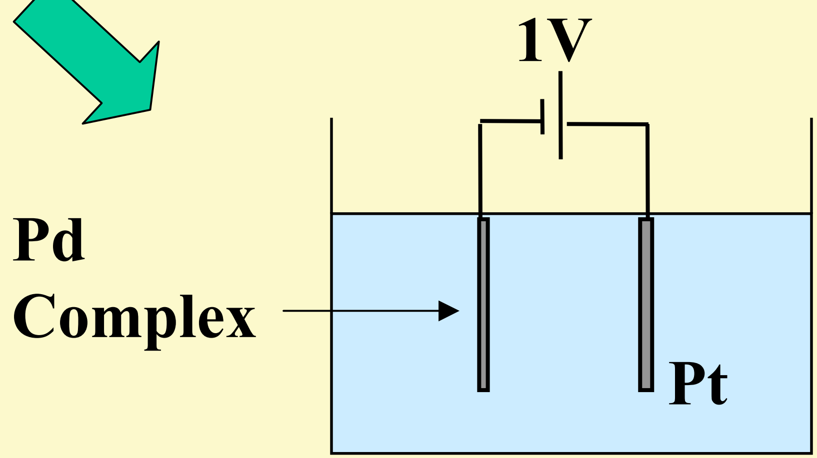




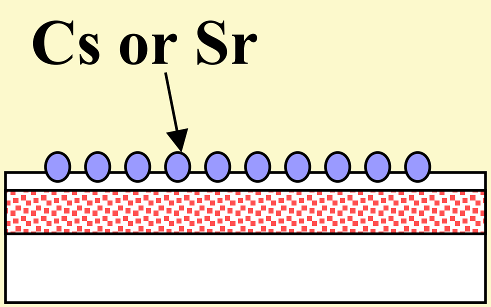
Element Addition (1: Electrolyte Addition)



Pd Complex

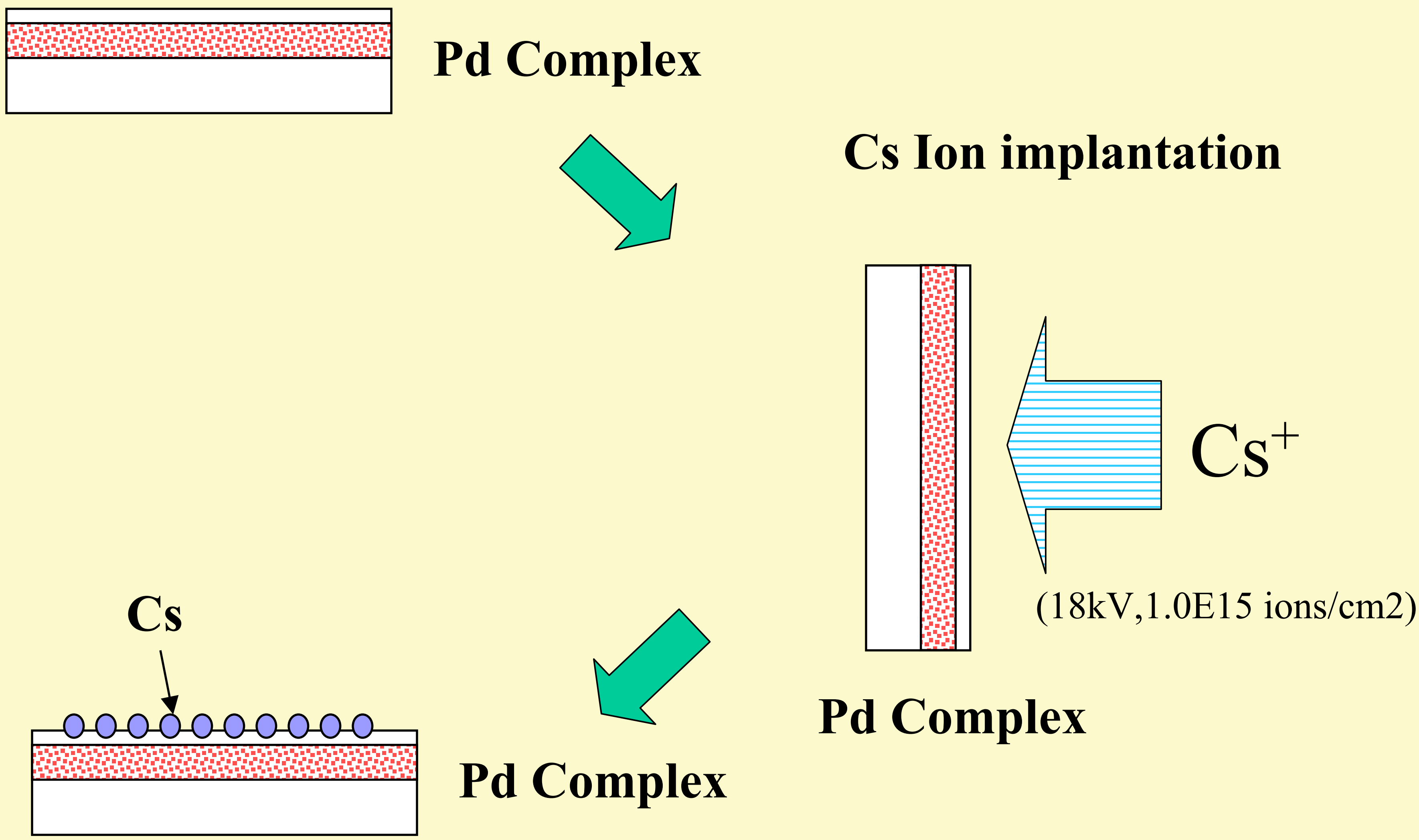


1mM $CsNO_3/D_2O$ solution
or 1mM $Sr(OD)_2/D_2O$ solution



Pd Complex

Element Addition
(2: Ion Implantation)



Identification of Pr by XANES

XANES(X-ray Absorption Near Edge Structure)

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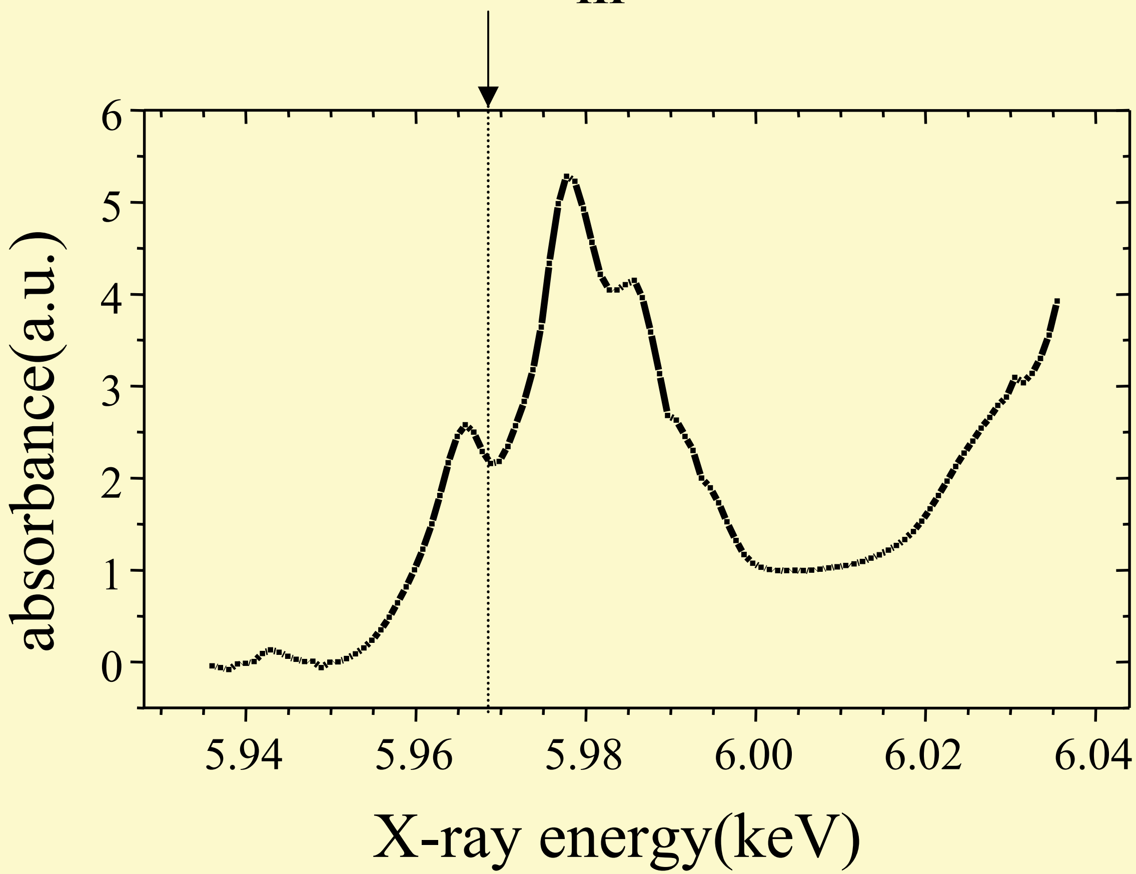
Pr-L_{III} Edge absorption



BL-9A Line, KEK, Tsukuba, Japan

Identification of Pr by ICP-MS

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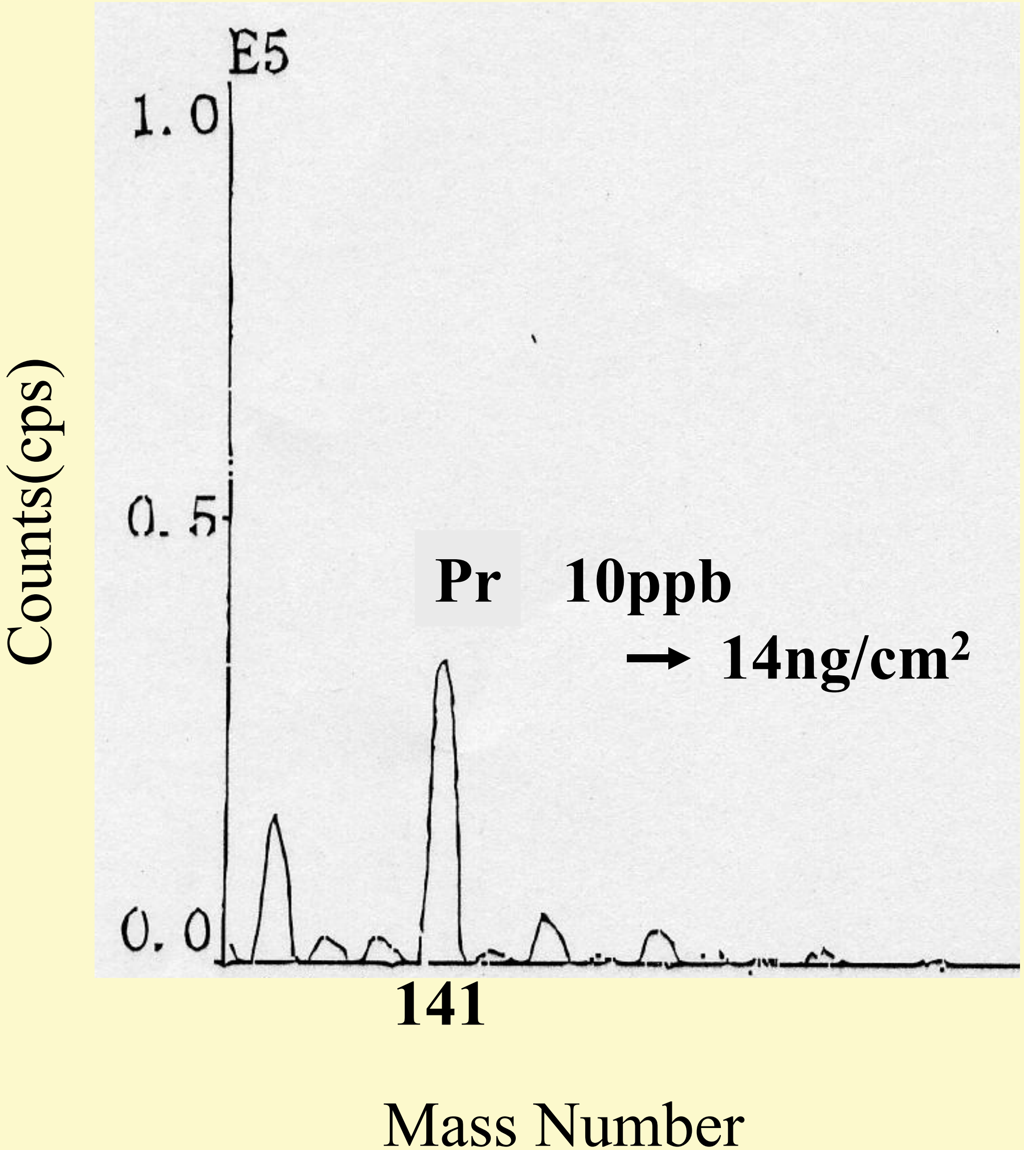

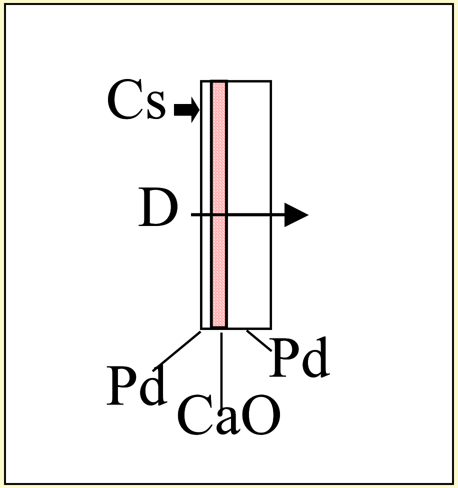

→ <u>Quantitative Analysis</u>

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)

↓

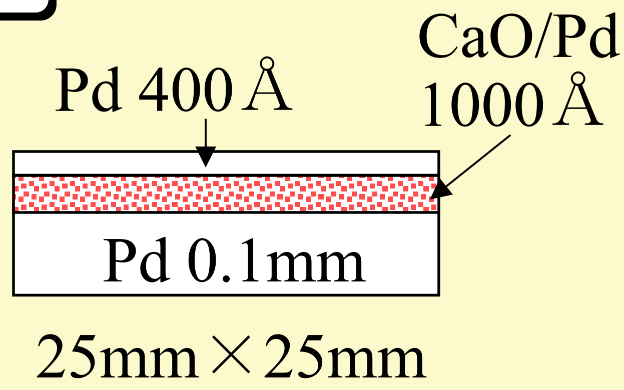
Washing the Sample with Aqua Regia (100sec)

↓

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

↓

Ion Beam Sputtering of Pd only (400Å)



Procedure of an Experiment

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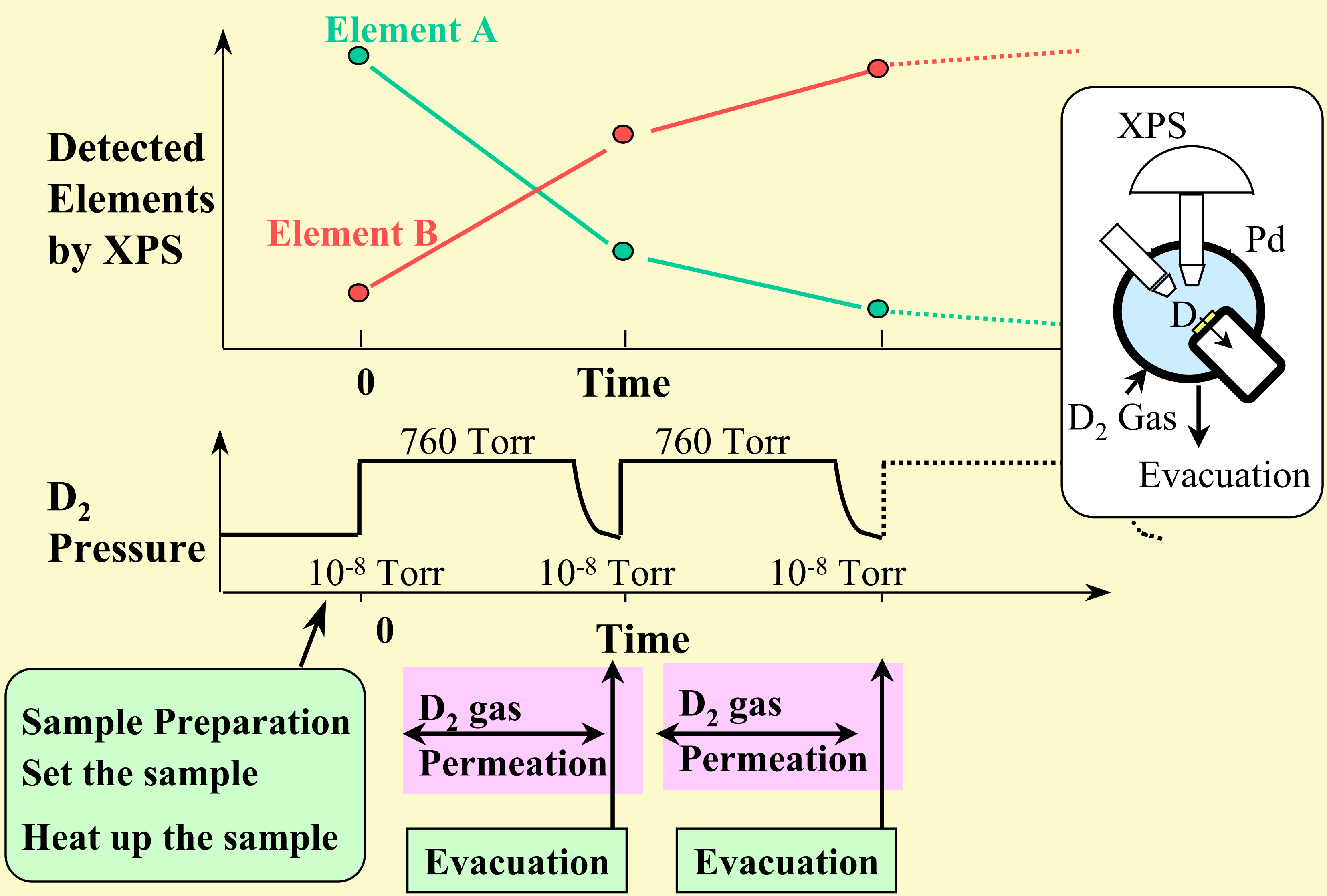

Transmutation of Sr into Mo

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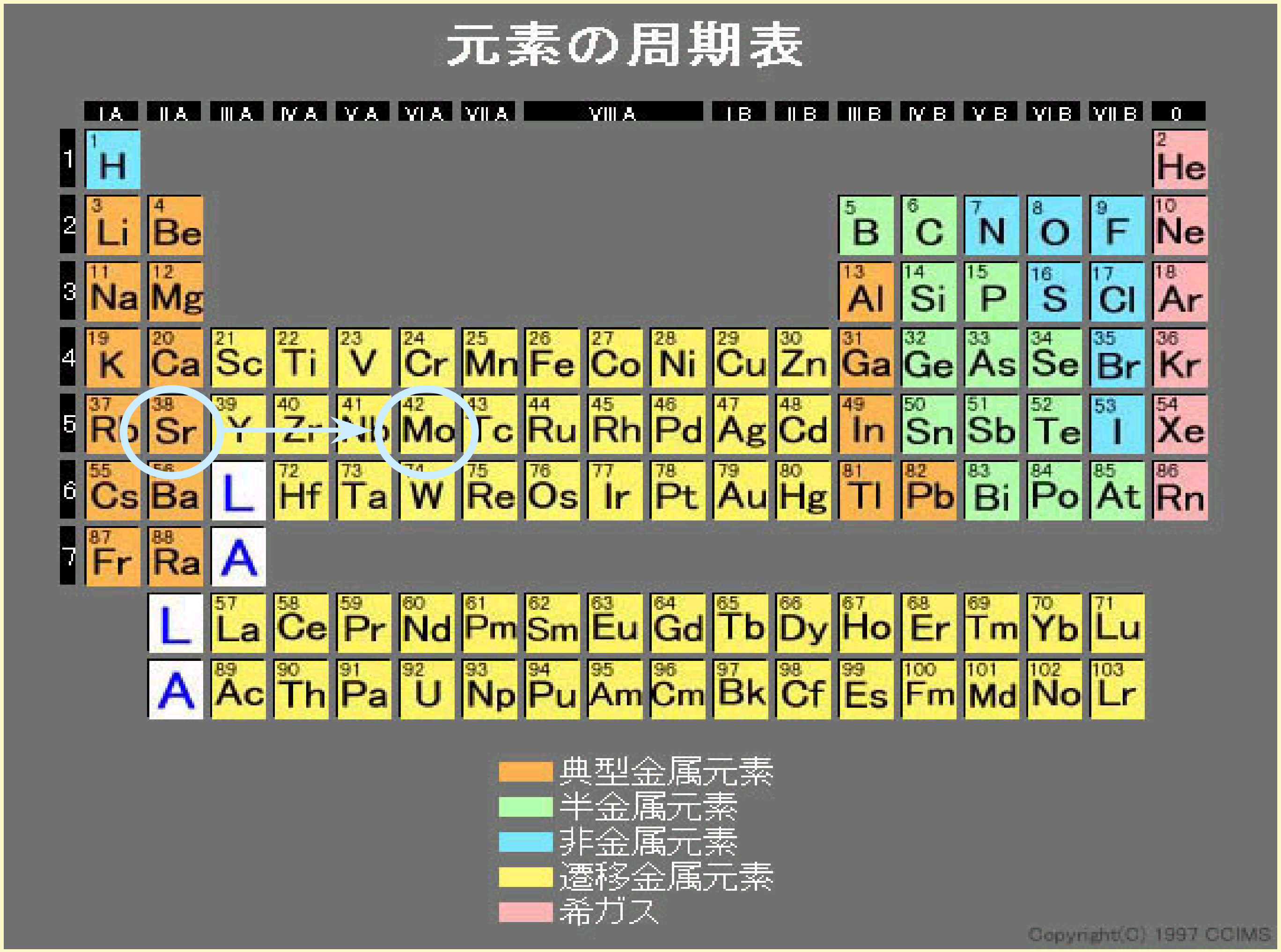

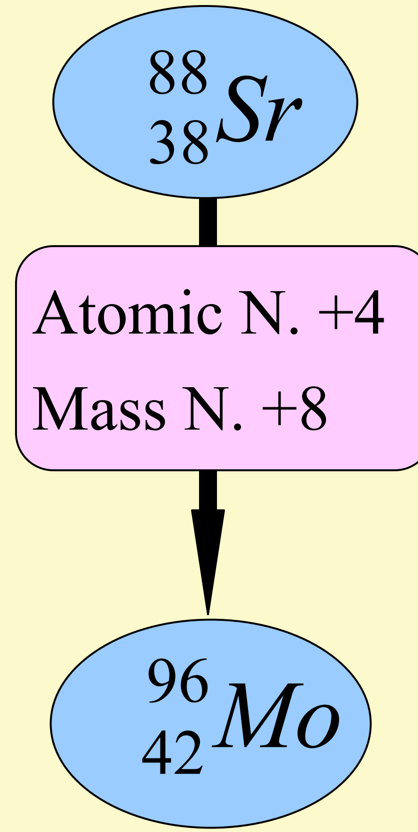

Excess Energy and Q-value

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

$$ Q \approx 50.5MeV $$

$$ \Downarrow $$

$$ EXH = 800J $$

$$ \Downarrow $$

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

Experimental results

$$ \left(\begin{array}{l} \mathrm{Pr} \approx 10^{14}\, atoms \\ reaction\ time \approx 100h \end{array} \right) $$

→**Undetectable in our experimental setup**

Depth Profile of Cs and Pr
: Ion Implantation

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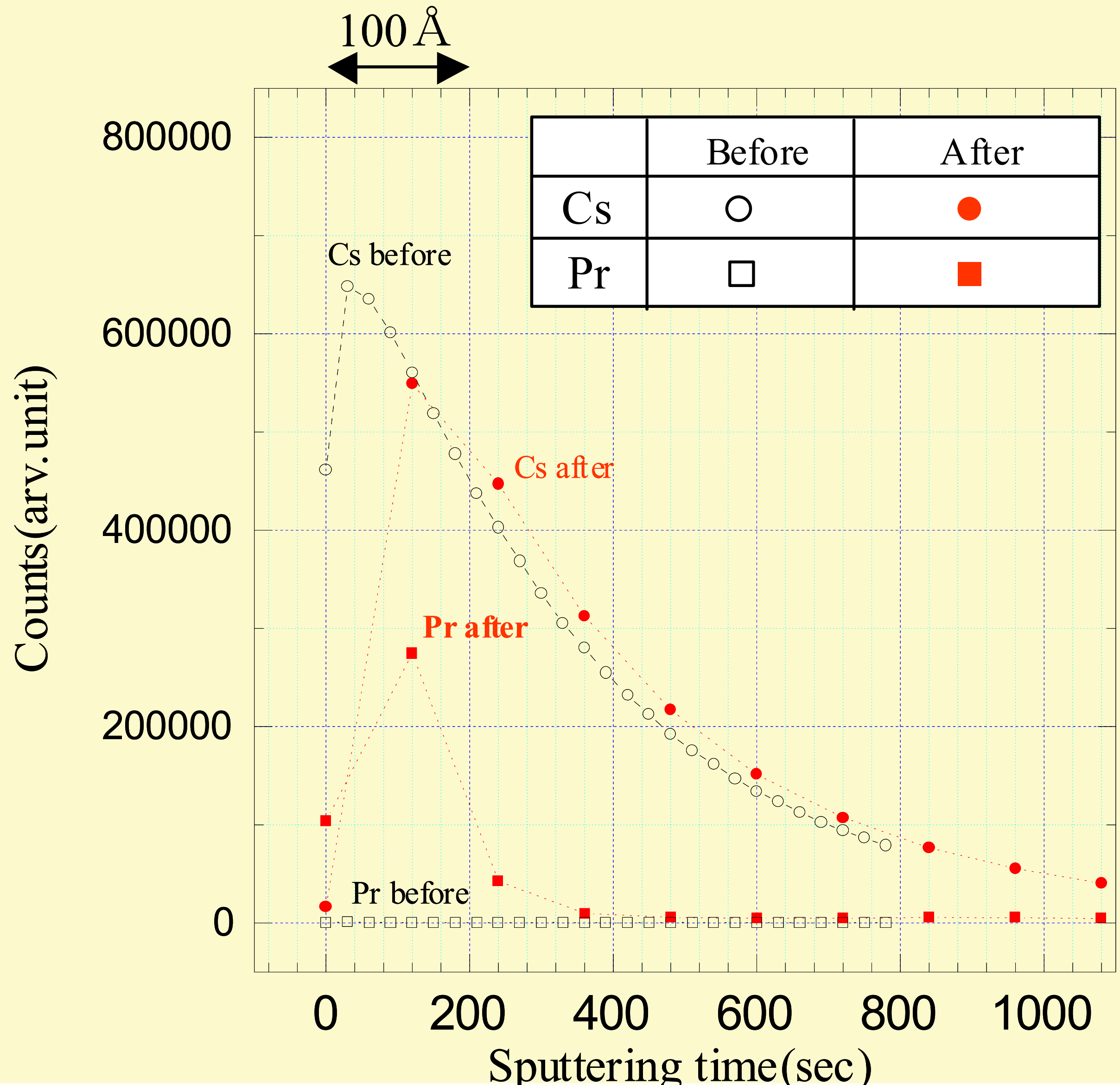
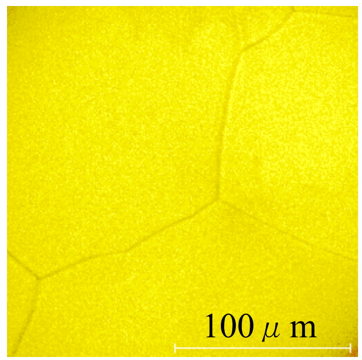
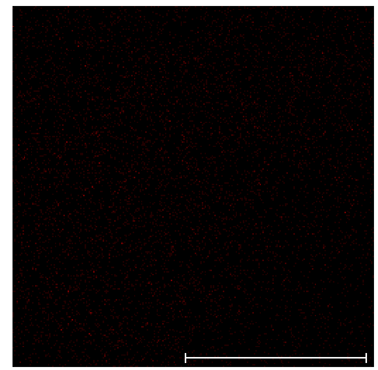
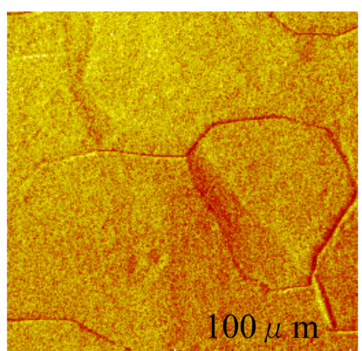
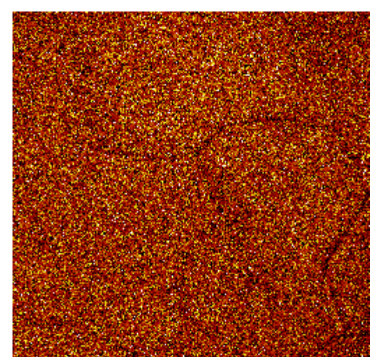
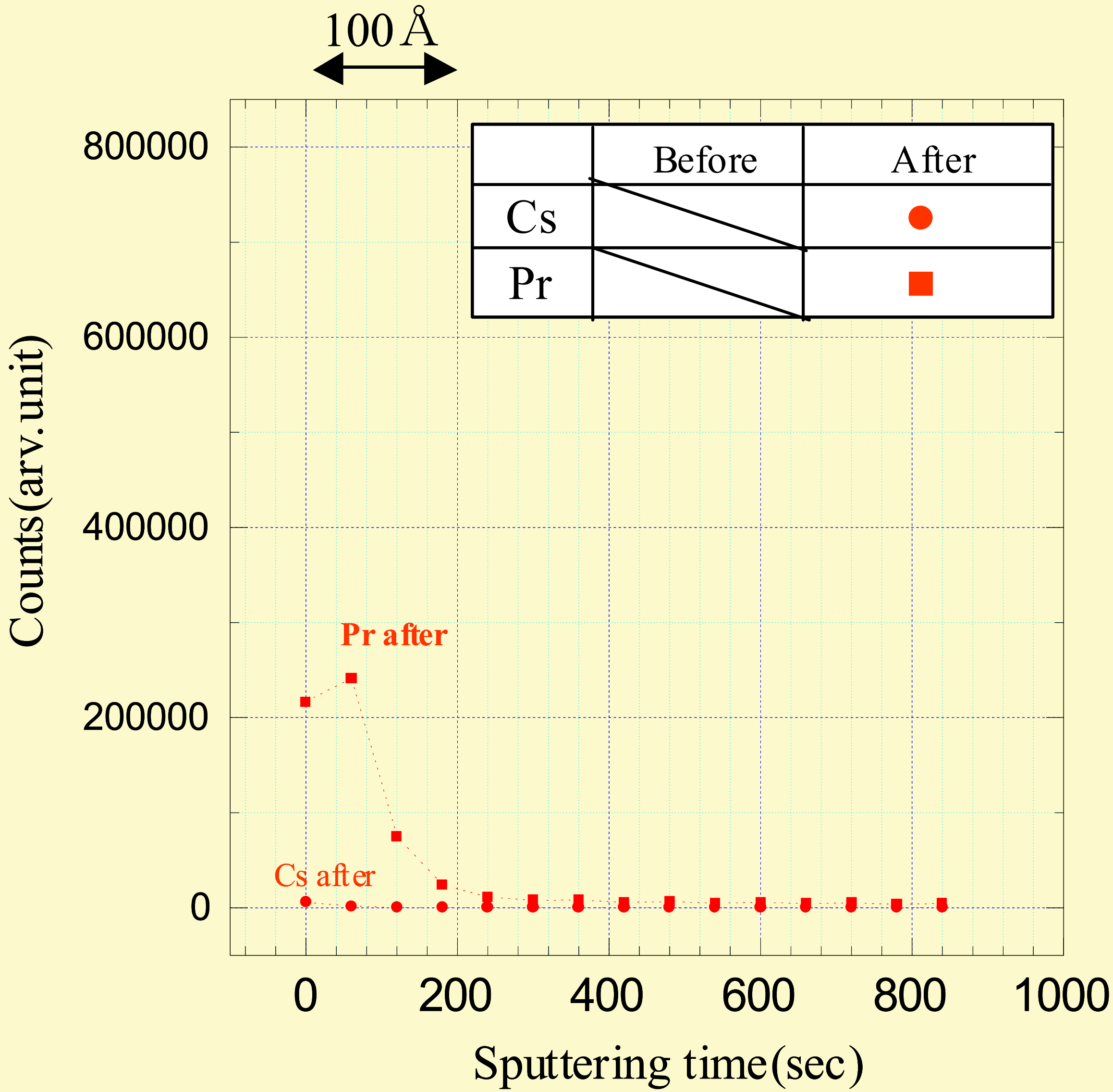


Surface Distribution of Cs and Pr : Ion Implantation

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

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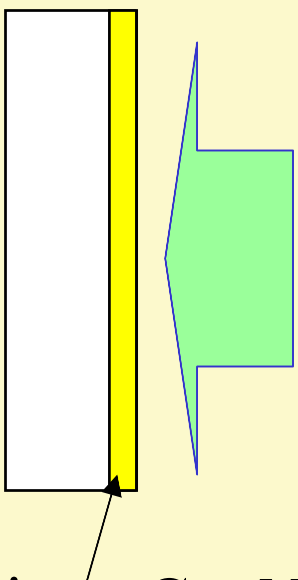
D Permeation(D Flux)

⇕

Ultra low D beam



D flux; ϕ

Given Cs; N_{Cs}

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

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

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

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

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

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

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

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

Separation of the Products and Contaminants(1)

1.Rare Earth Element : Pr

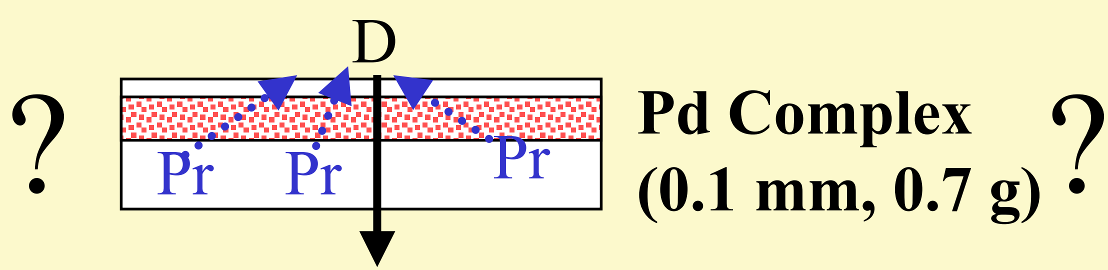
a) D_2 gas :The purity > 99.6% most of the impurity is H_2～0.4% N_2, D_2O, O_2, CO_2, CO, HC<10 ppm

b) A Pd complex deposited with Cs
Pr < detection limit (0.01 ppt) by ICP-Mass

If all of the Pr at 0.01ppt distributed in the Pd test piece gathered in the analyzed area,

Max contamination Pr～0.1ng

→Less than the detected Pr(10ng～100ng)!



Separation of the Products and Contaminants(2)

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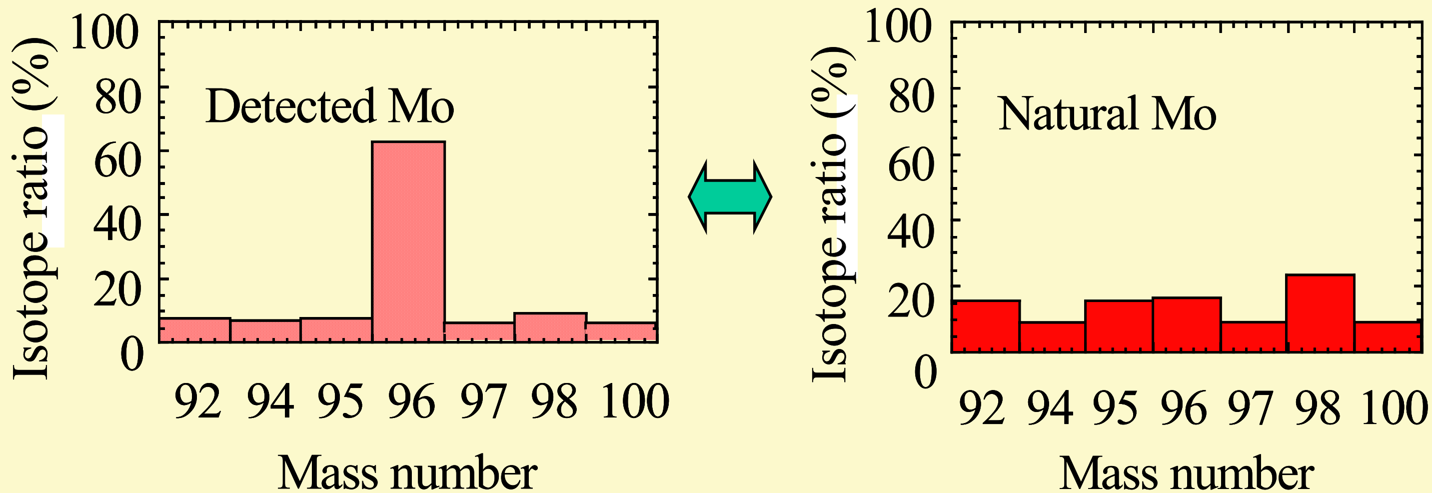
2. Anomalous Isotopic Composition: Mo



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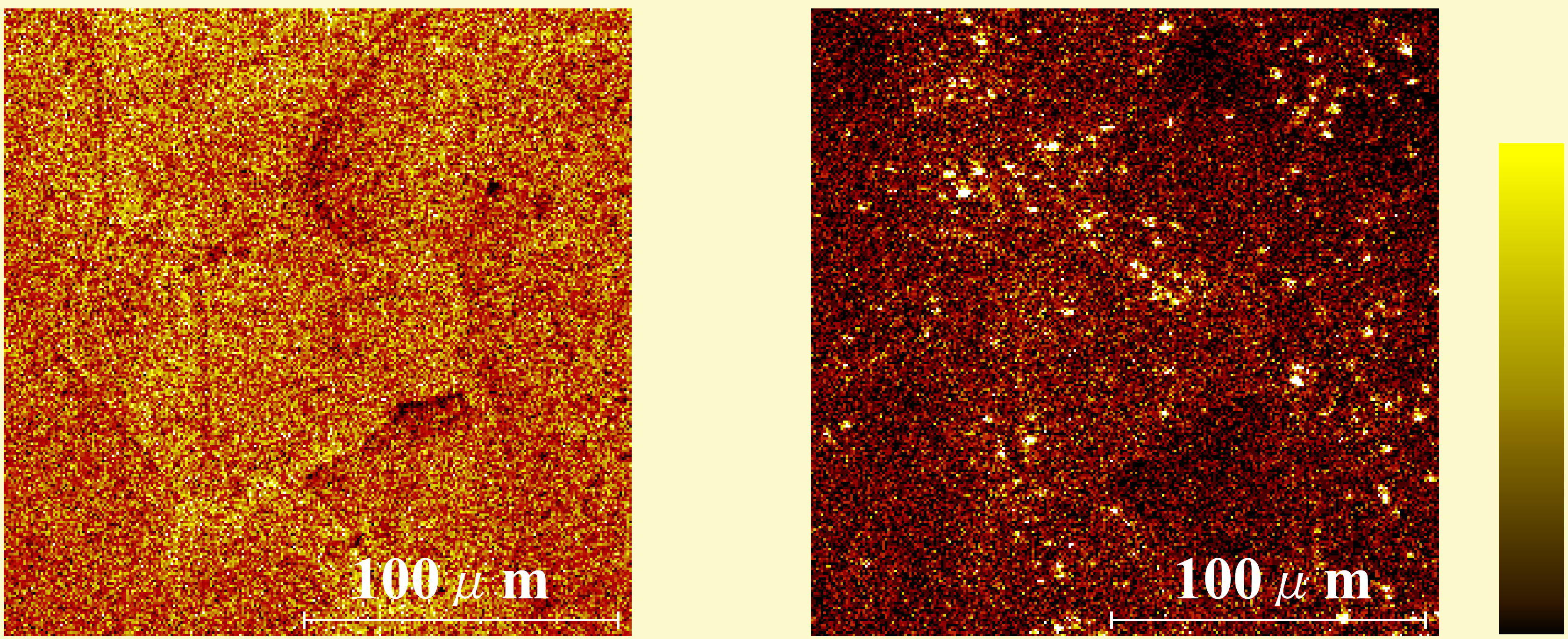
Sample:Cs is added using electrochemical method
Analysis:Tof-SIMS(ULVAC fai)

Cs

Pr



Results from the depth profile and the surface distribution analyses

1. **Transmutation occurs in the thin surface region up to 100 angstrom.**
   →Correlated with D/Pd?
   →Important to surface analysis

2. **The surface distribution of Pr basically seems to be uniform. There is no correlation between Pr and grain boundaries. However, in the case of electrolyte addition, slight non uniformity of Pr was found.**
   →Migration of Pr ?
   →Due to non uniformity of Cs addition?

実験装置概観

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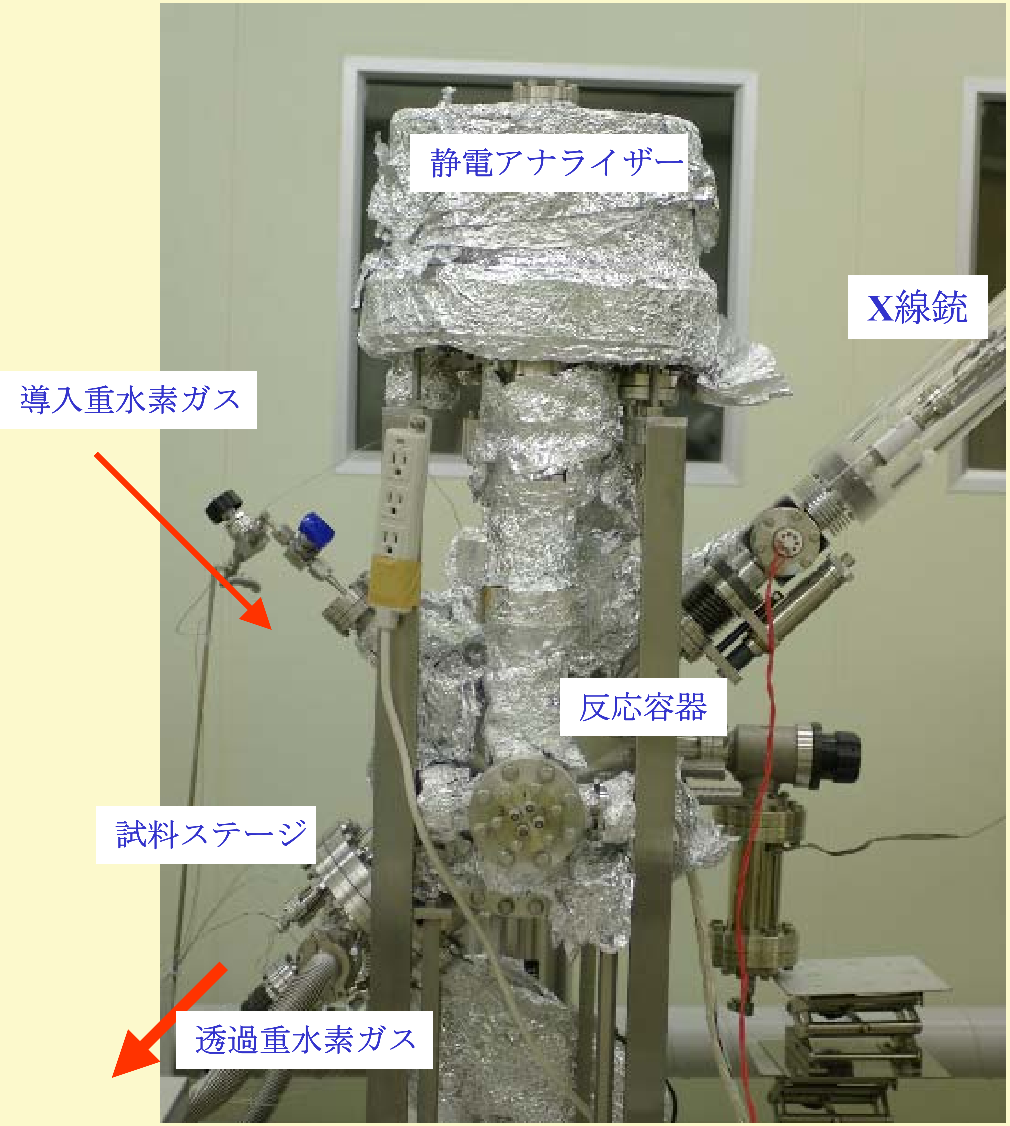

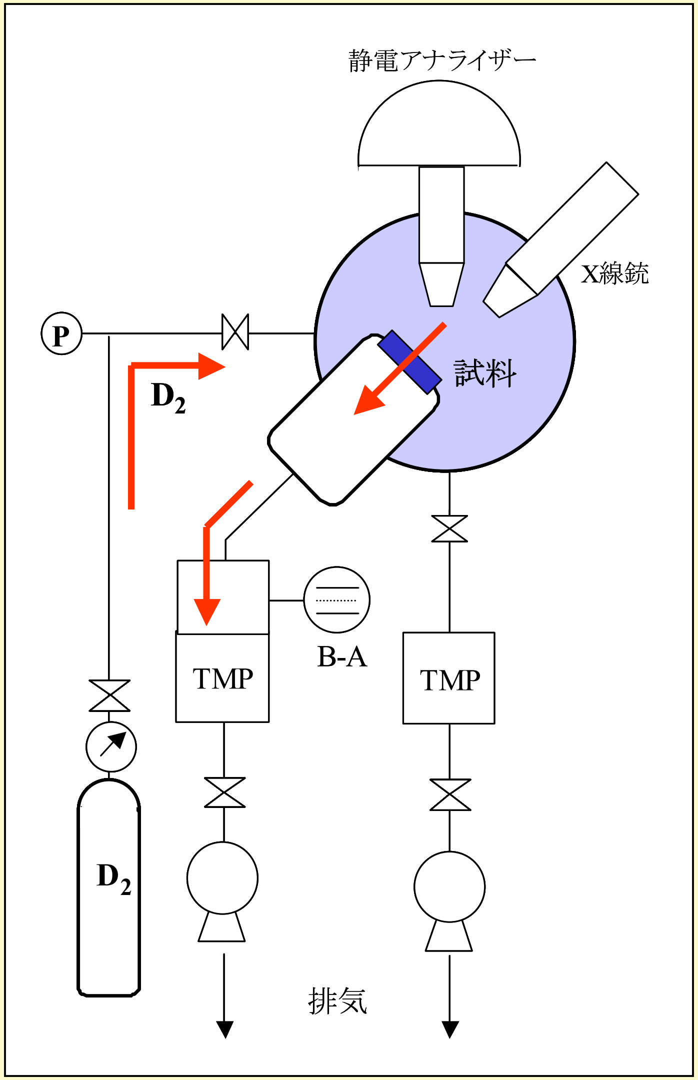

試料ステージ概観

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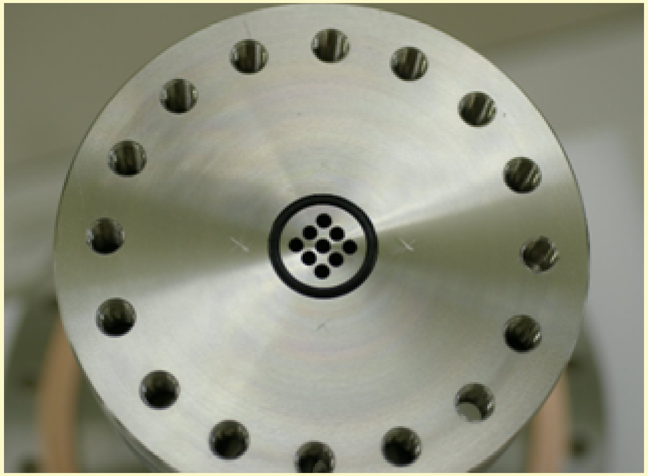
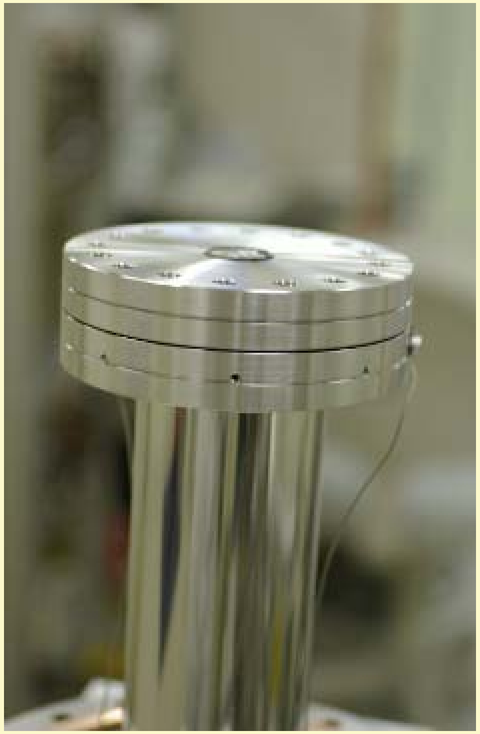
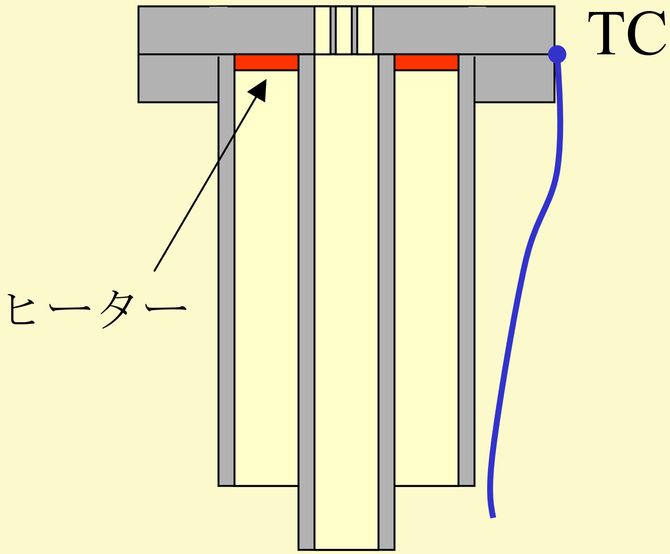
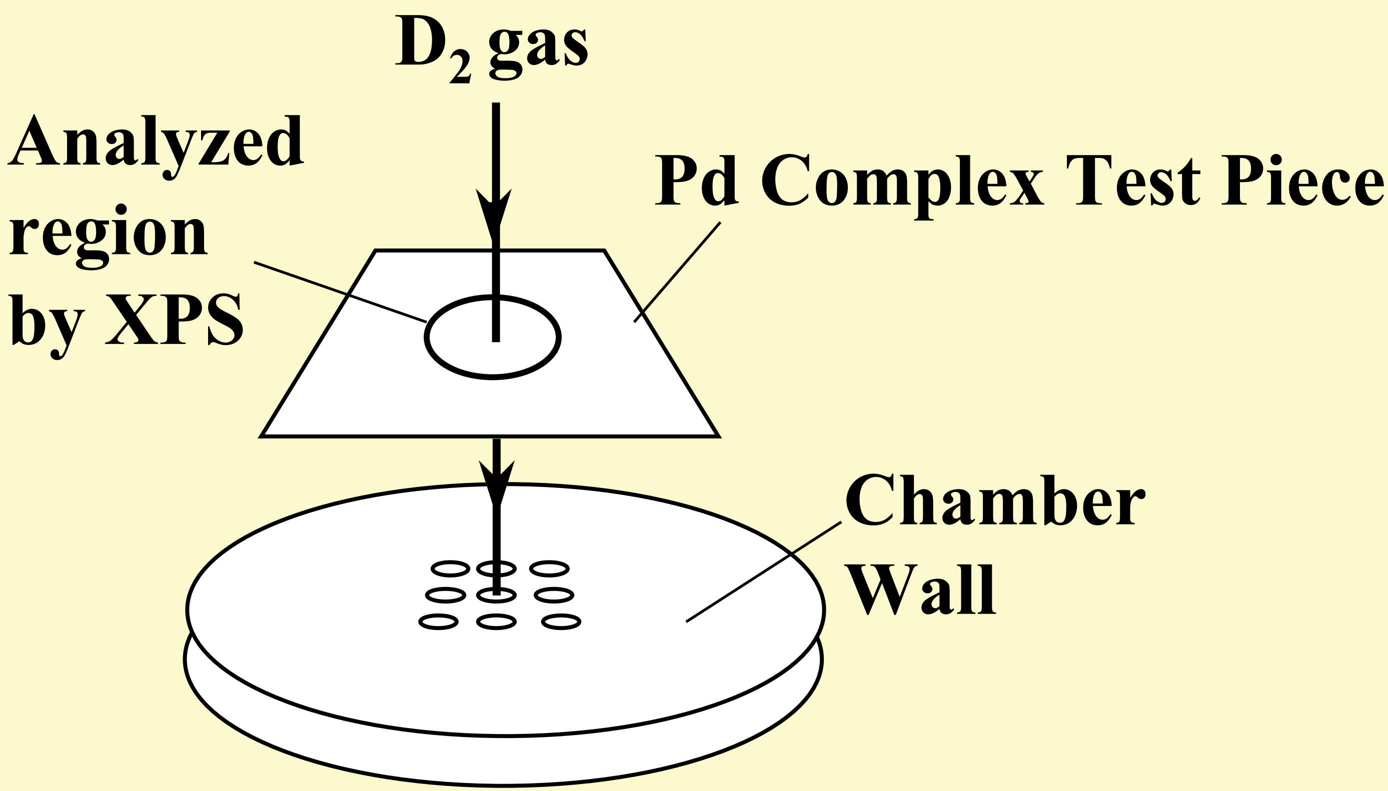






重水素ガスの透過経路


D_2 gas
Analyzed region by XPS
Pd Complex Test Piece
Chamber Wall

Identification of Pr by TOF-SIMS(2)

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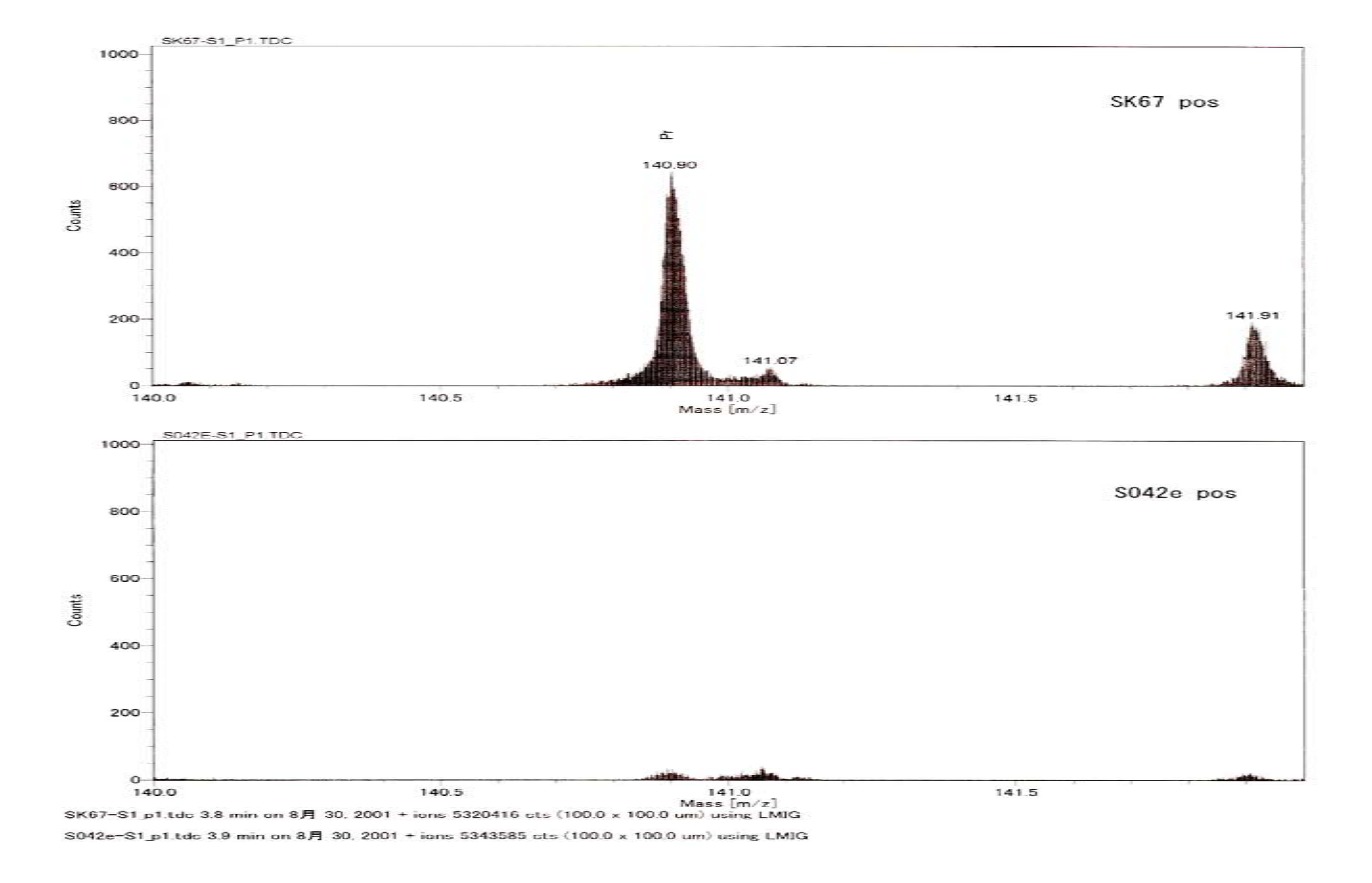

Quantitative Analysis of Pr by ICP-MS

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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

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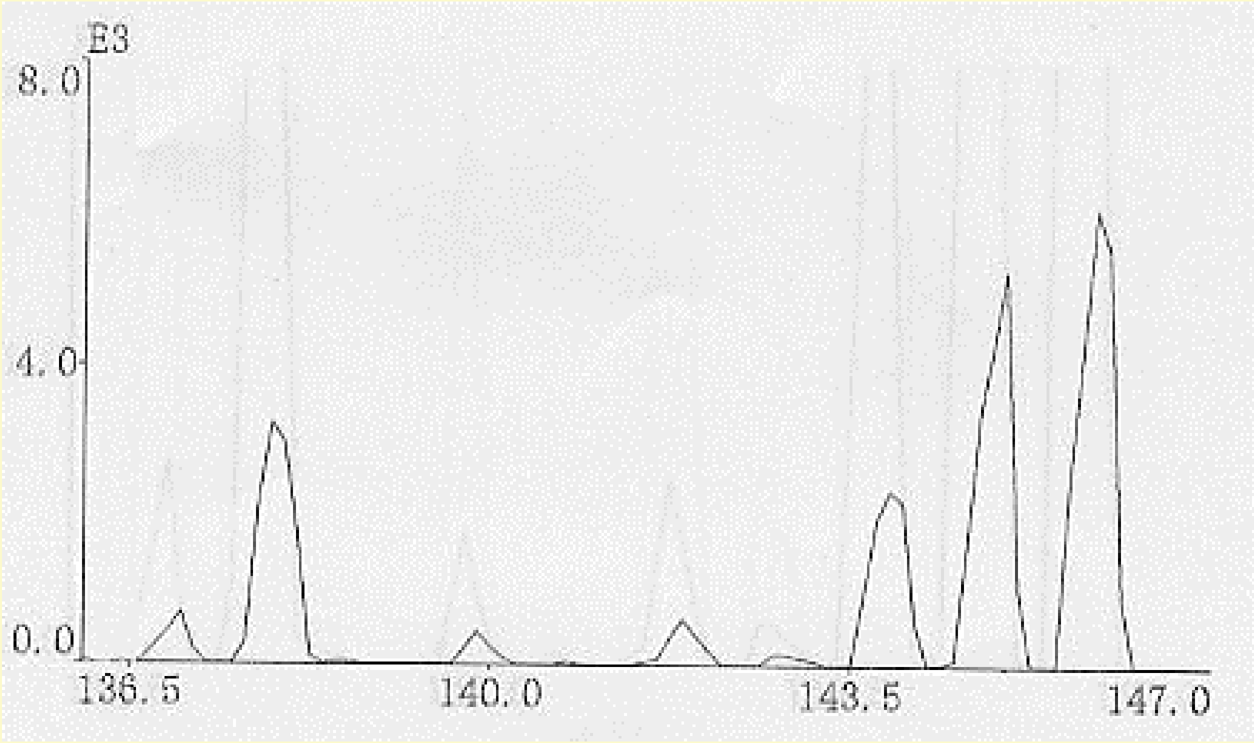

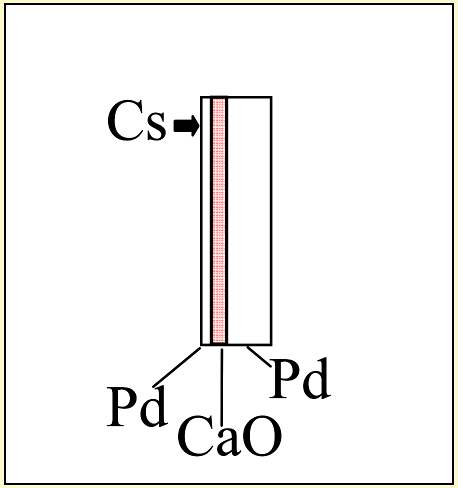

→ Prは検出されず。

F.G.Data(1)

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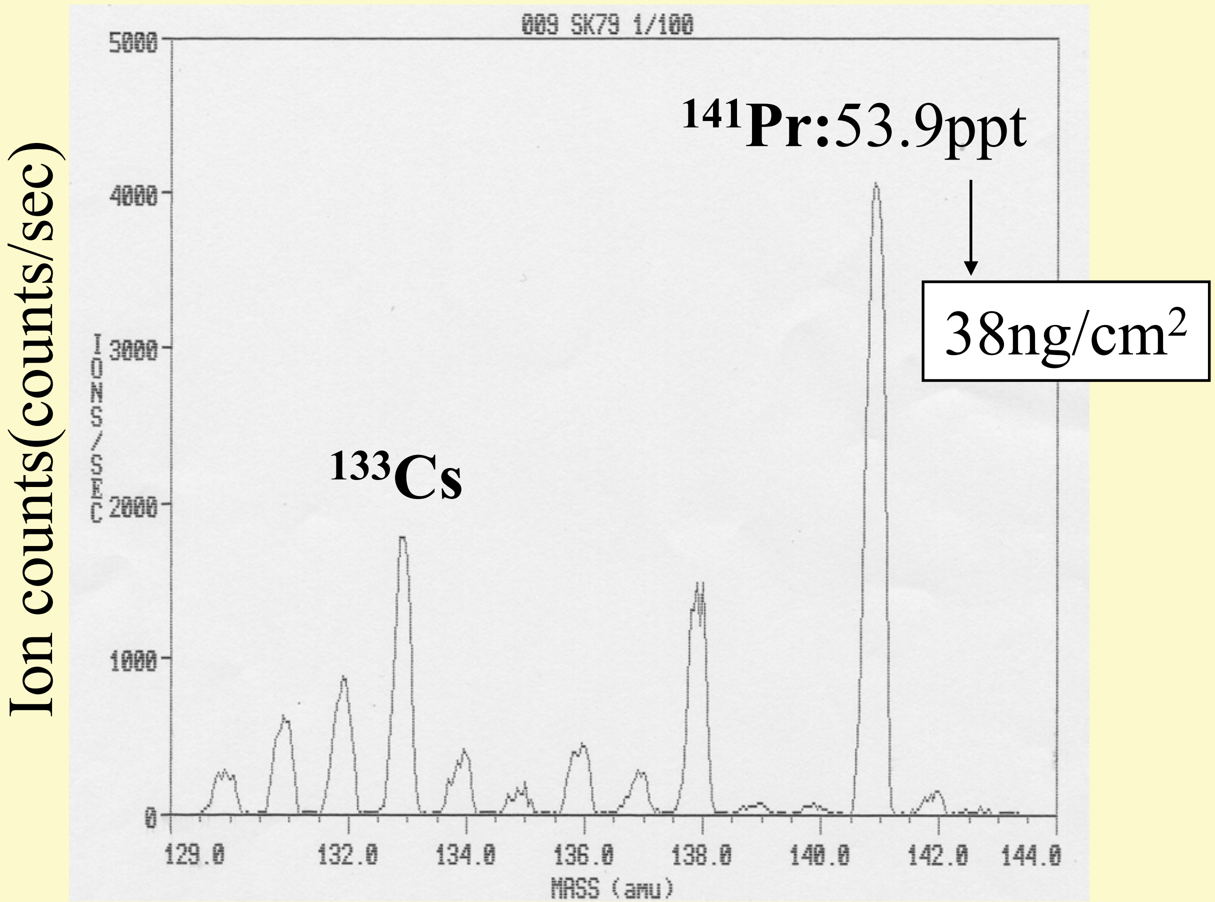
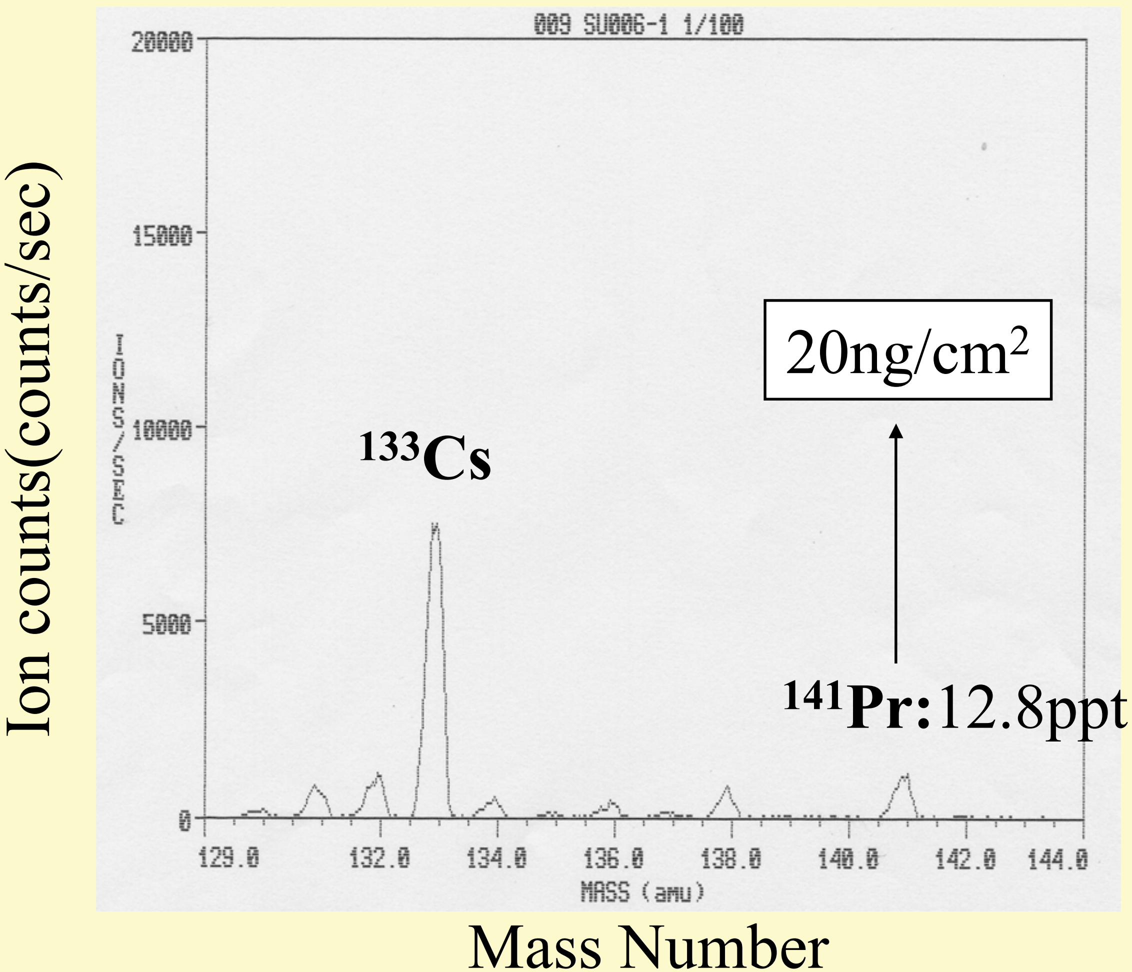


Mass Number

F.G.Data(2)


009 SU006-1 1/100
20000
15000
10000
5000
0
IONS/SEC
129.0
132.0
134.0
136.0
138.0
140.0
142.0
144.0
MASS (amu)
^{133}Cs
20ng/cm^2
^{141}Pr:12.8ppt
Ion counts(counts/sec)
Mass Number

Analysis of the Standard Solution

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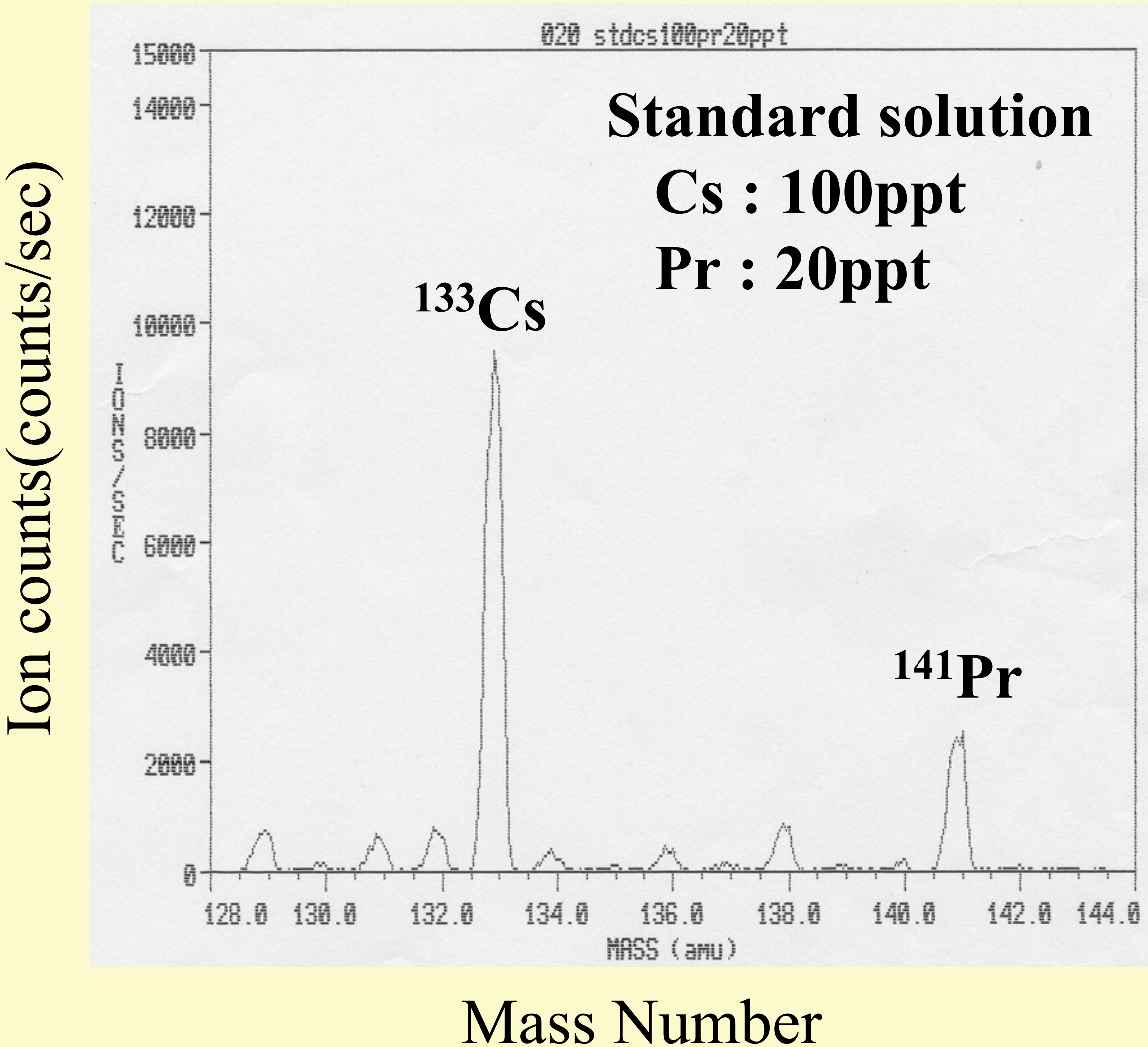
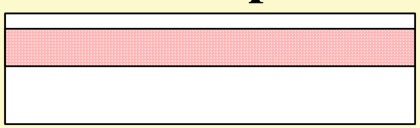
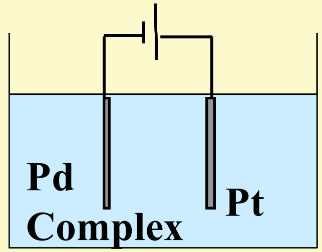
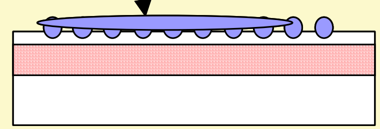
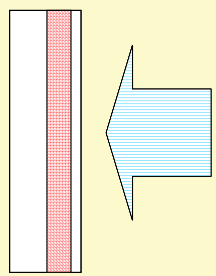
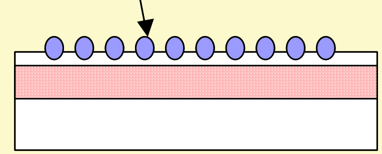
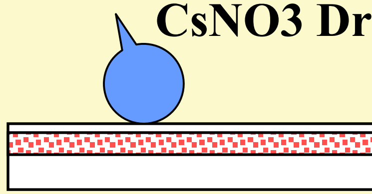
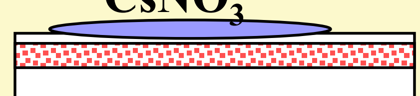


Transmutation Dependence on the Element Addition Method

Pd Complex 	**1V** **Pd Complex** **Pt**  **1mM $CsNO_3/D_2O$ solution**	**Cs + CsNO3** 	Positive
	 Cs^+ **Pd Complex**	**Cs** 	Positive
	CsNO3 Drop 	**$CsNO_3$** 	Negative

Conjecture on D distribution in the Pd Complex

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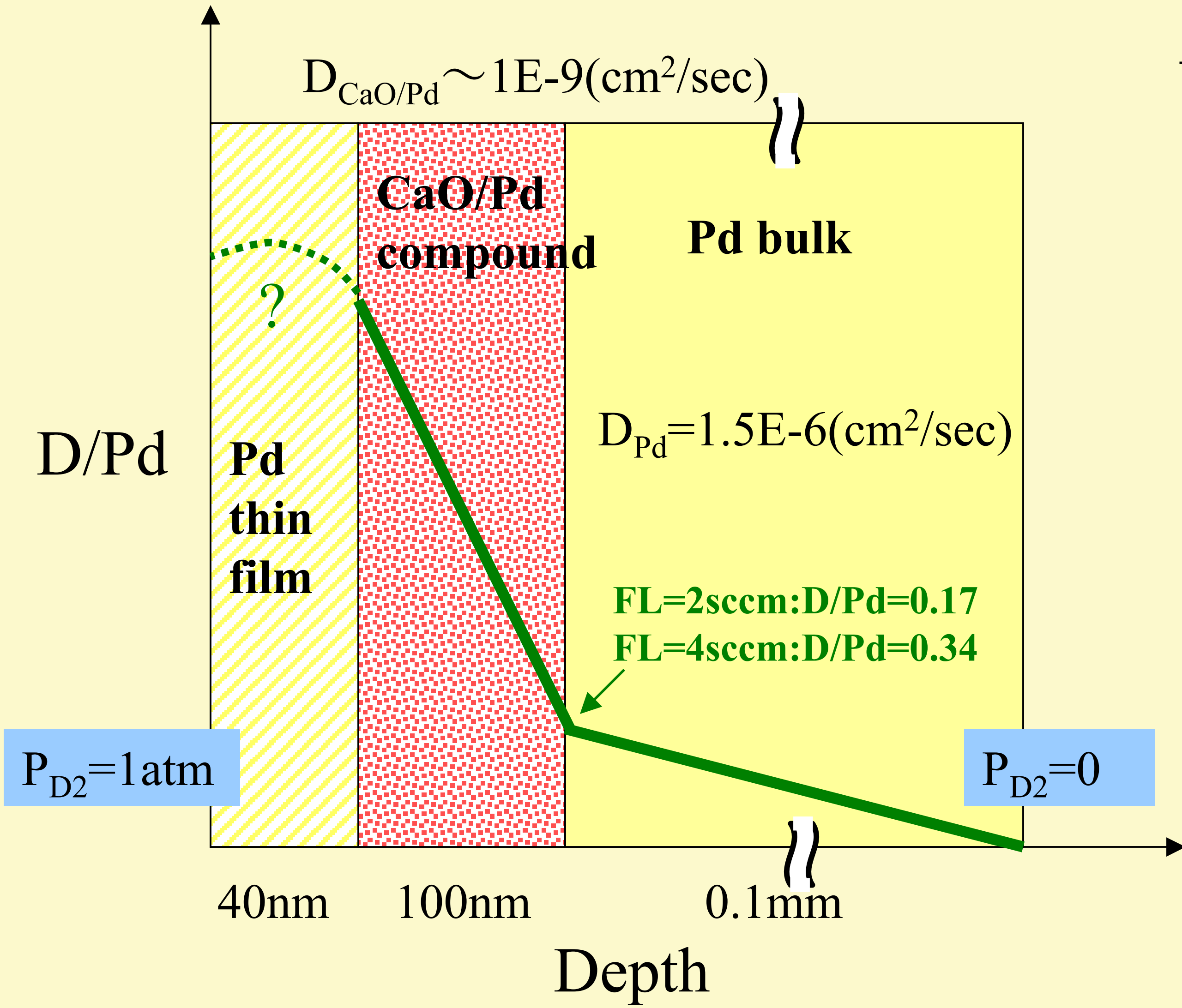


$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} \qquad \frac{\partial}{\partial t} = 0$$

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

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

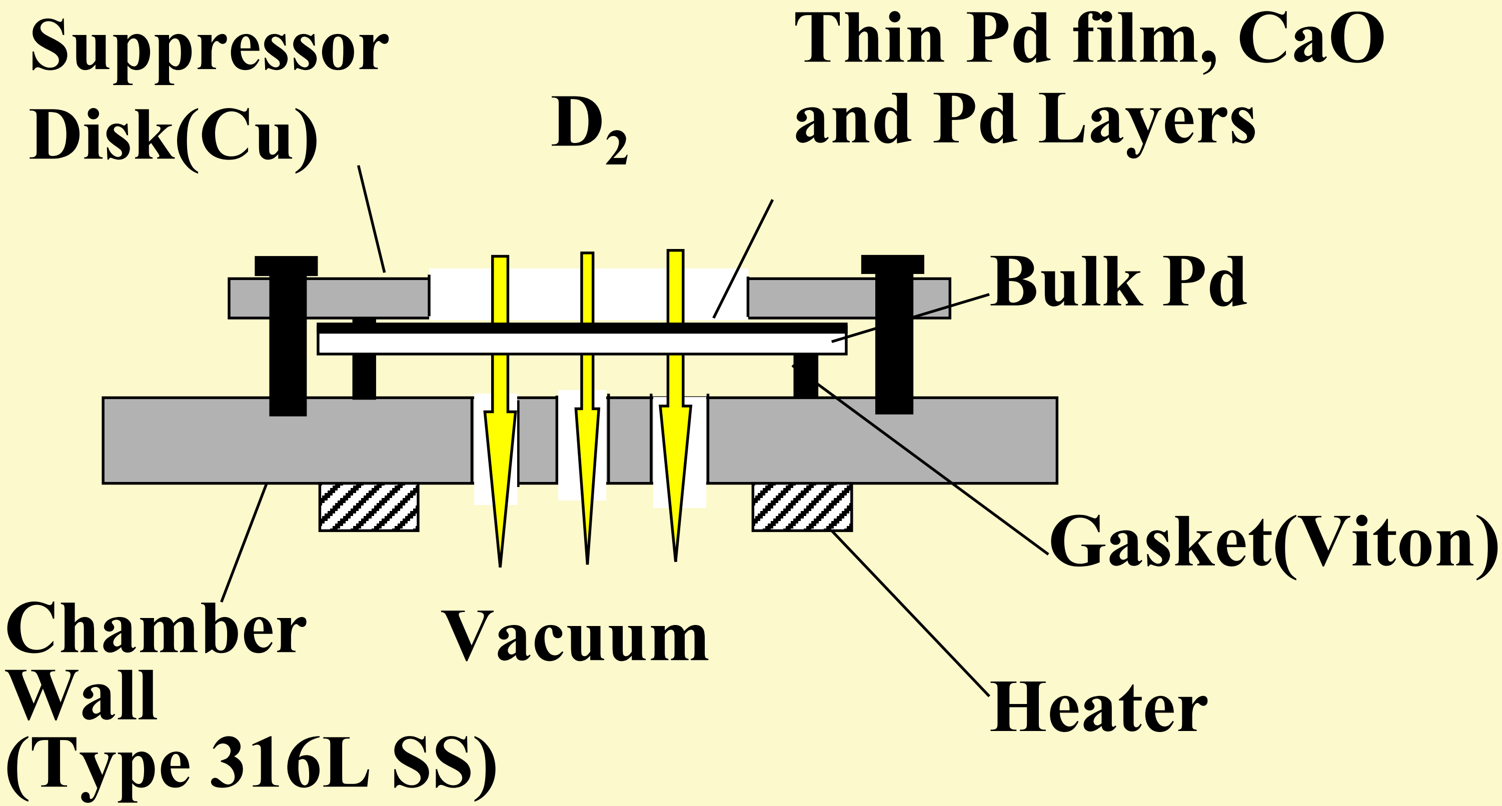
$$= -A \cdot D_{Pd} \cdot \left.\frac{\partial C}{\partial x}\right|_{Pd}$$

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

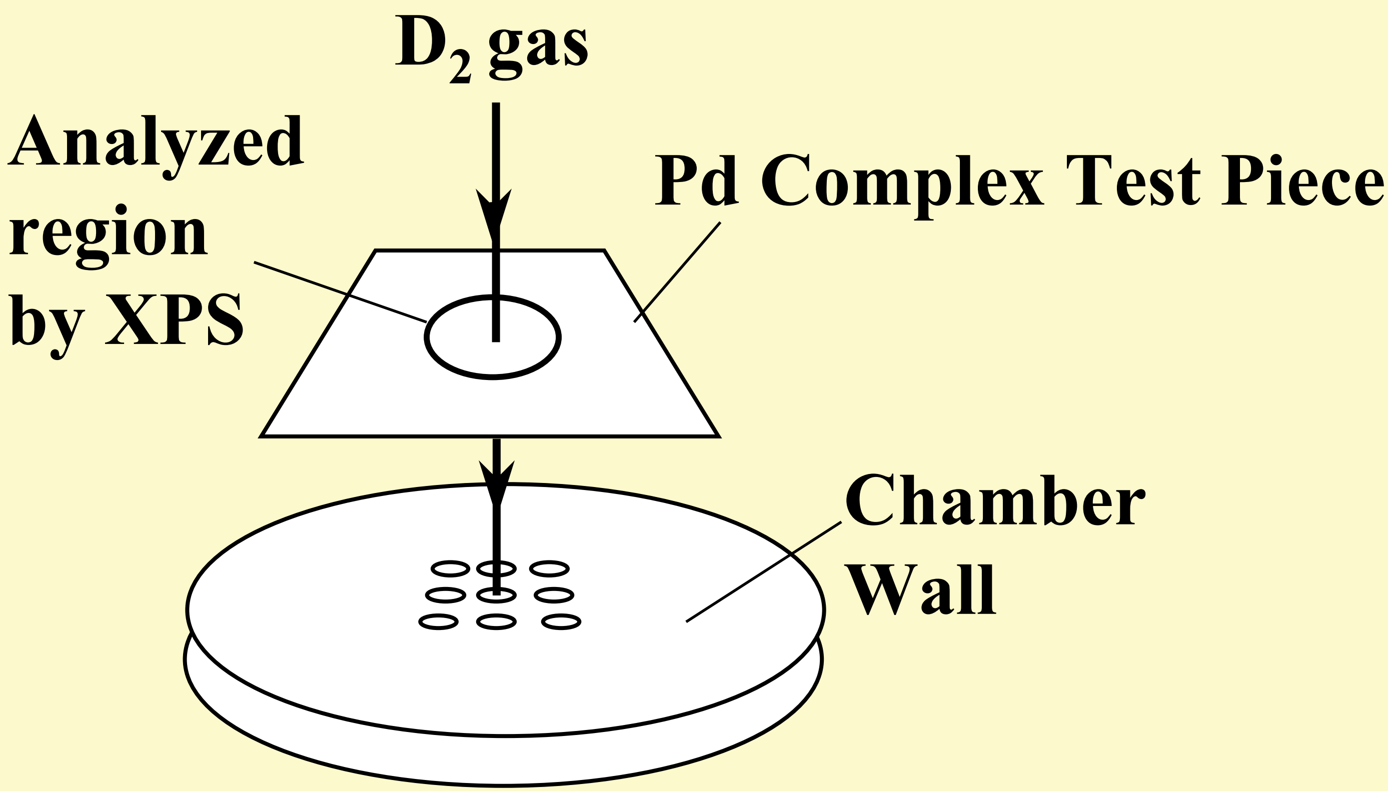
$$\Downarrow$$

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

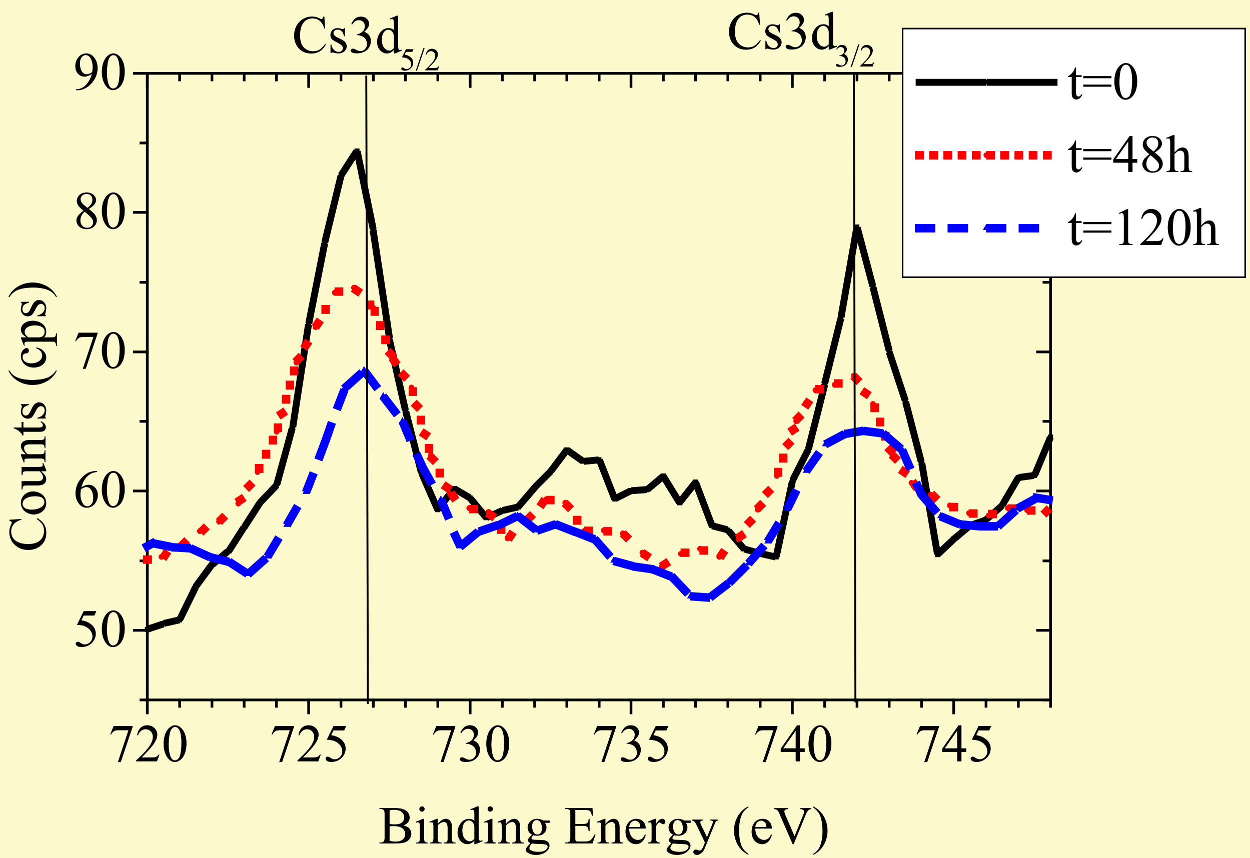
Cross Sectional View



Passage of D_2 Gas



Change of XPS Spectrum of Cs



Change of XPS Spectrum of Pr

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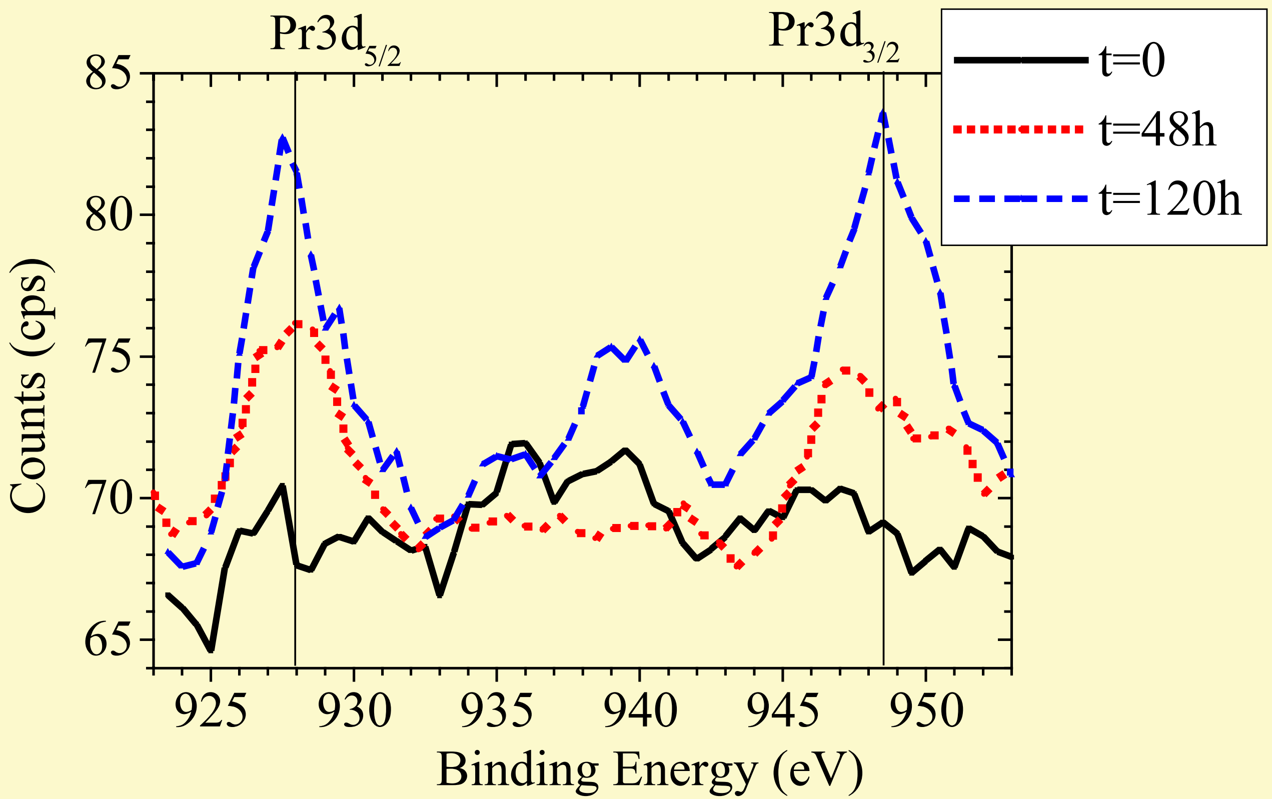

Time Variation in number of Cs and Pr atoms in the case of D_2 Permeation through thin film and bulk Pd with added Cs

MITSUBISHI HEAVY INDUSTRIES, LTD.
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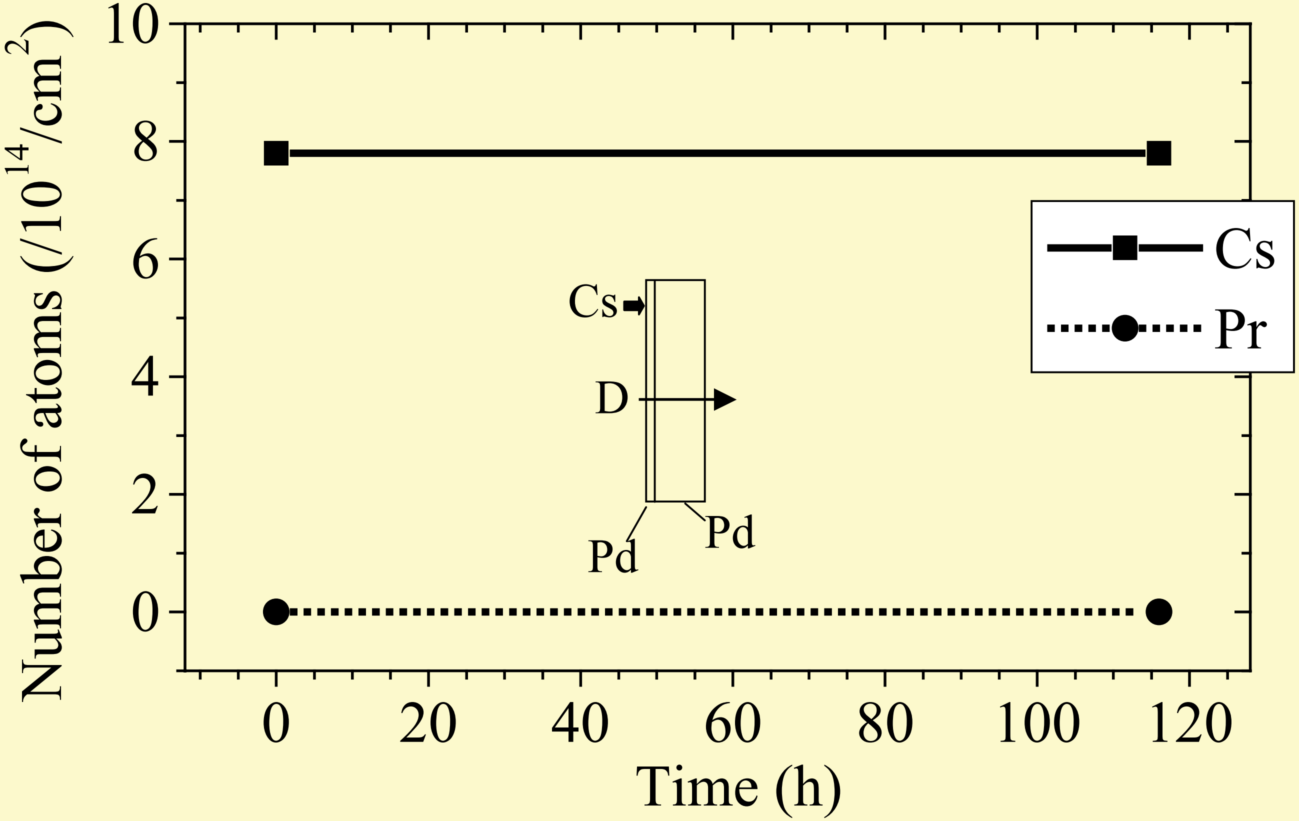

XPS Spectrum of Cs (No CaO)

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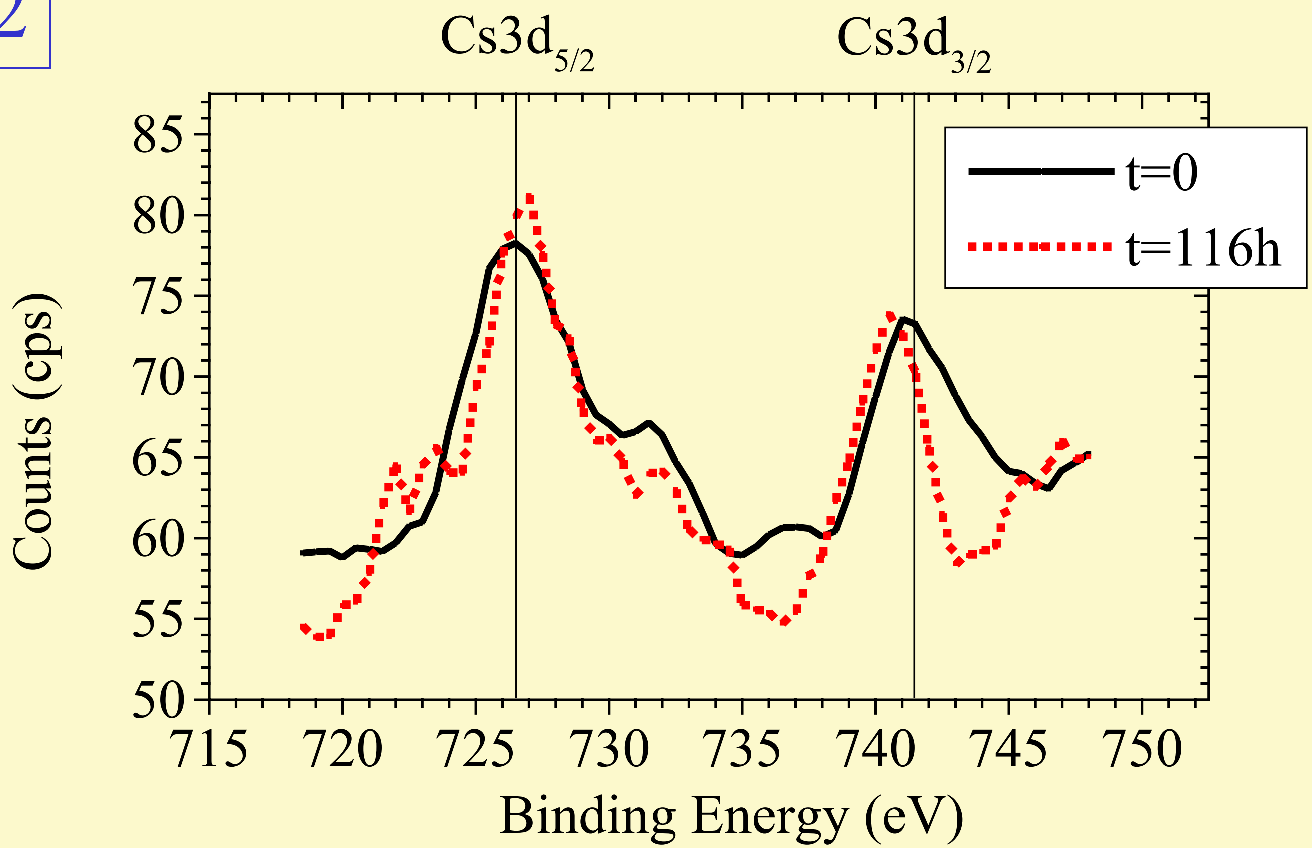
Pd/Pd/D2



XPS Spectrum of Pr (No CaO)

MITSUBISHI HEAVY INDUSTRIES, LTD.
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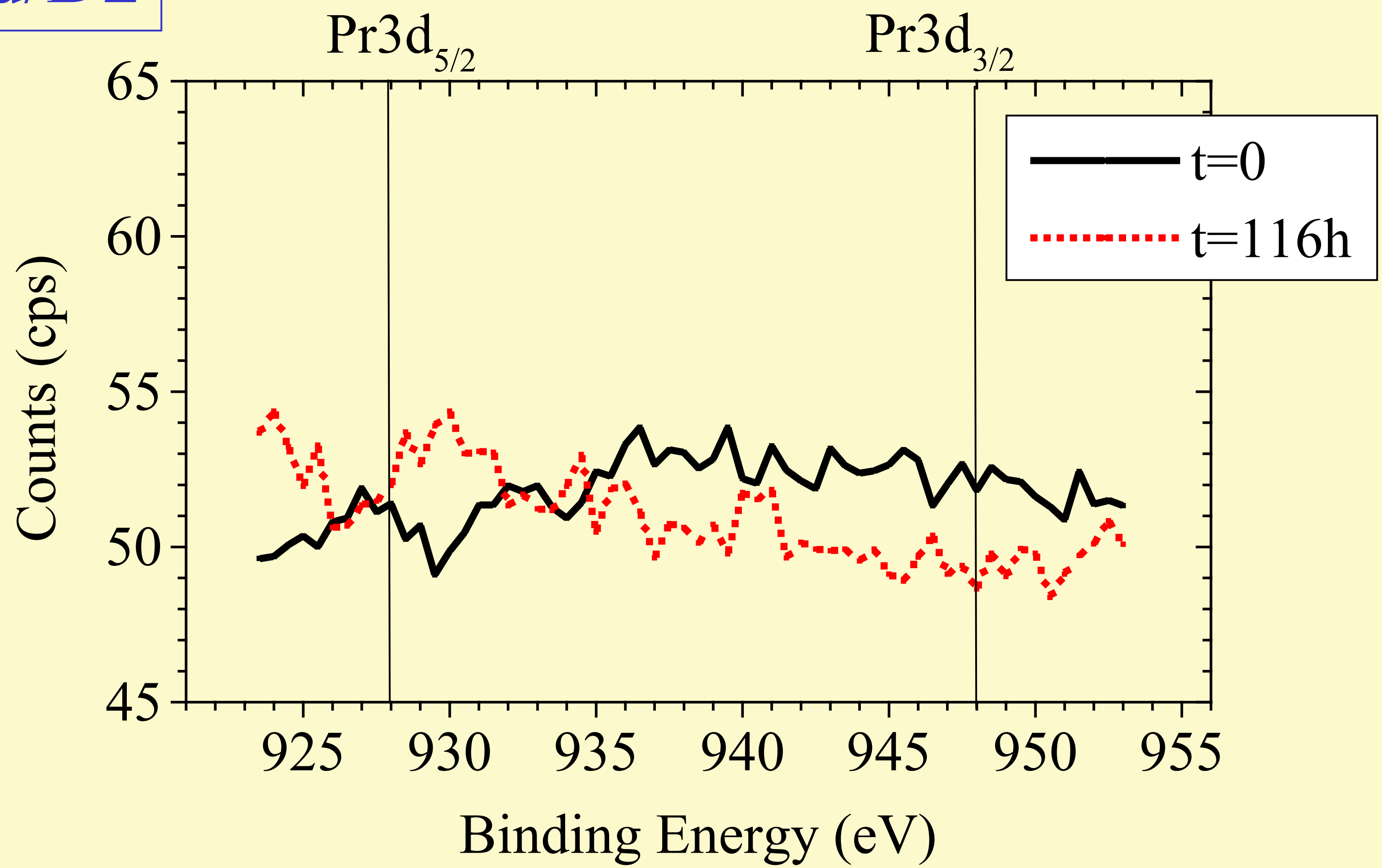
Pd/Pd/D2



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

MITSUBISHI HEAVY INDUSTRIES, LTD.
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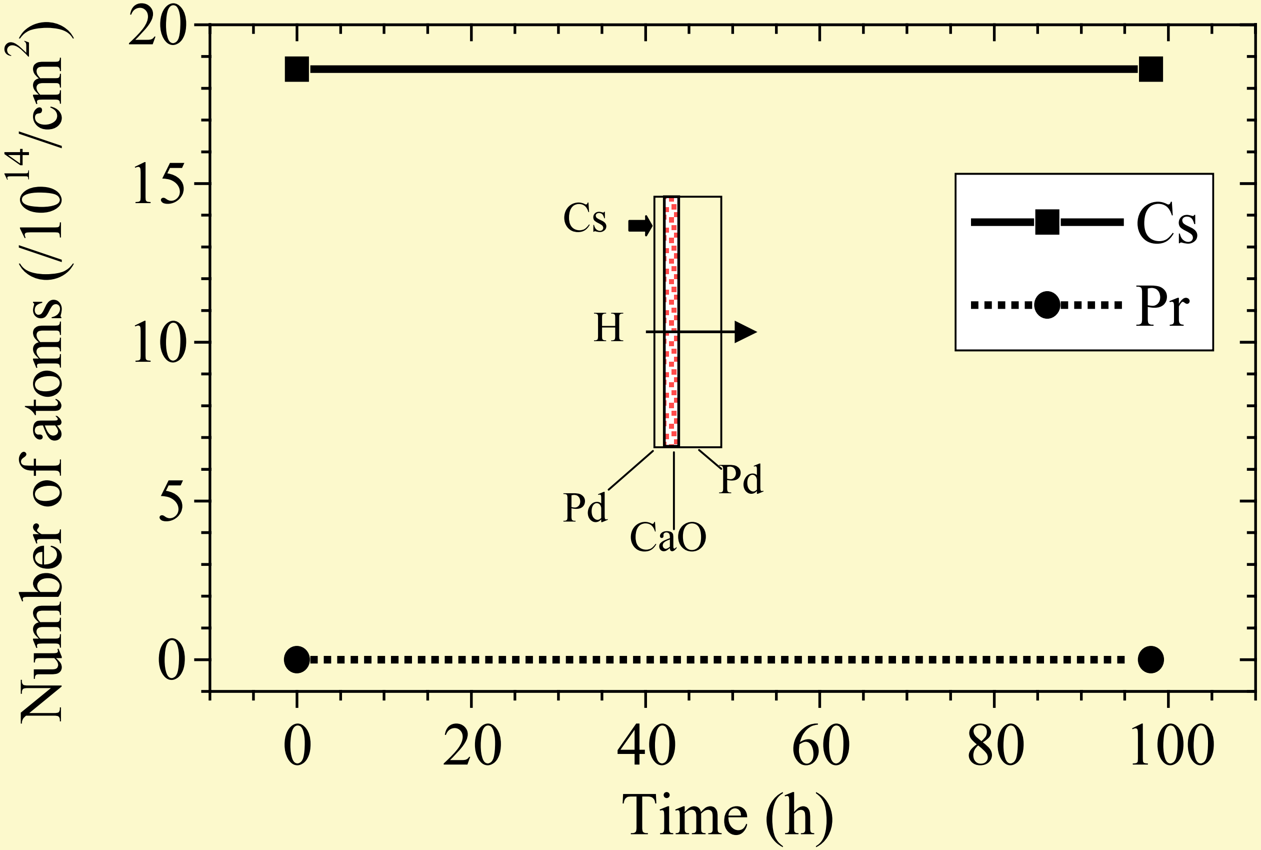
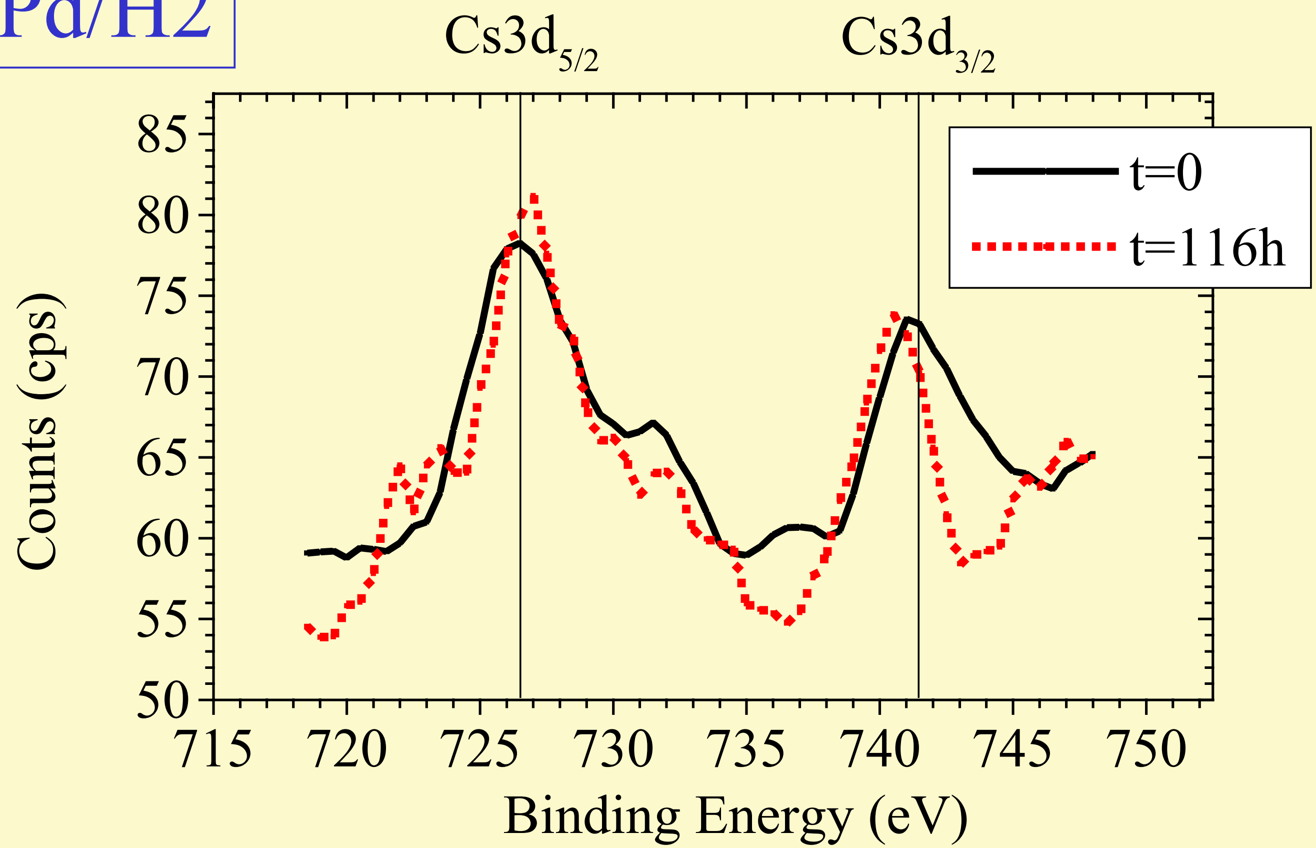


XPS Spectrum of Cs(H_2 Permeation)

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

Pd/CaO/Pd/H2



XPS Spectrum of Pr (H_2 Permeation)

MITSUBISHI HEAVY INDUSTRIES, LTD.
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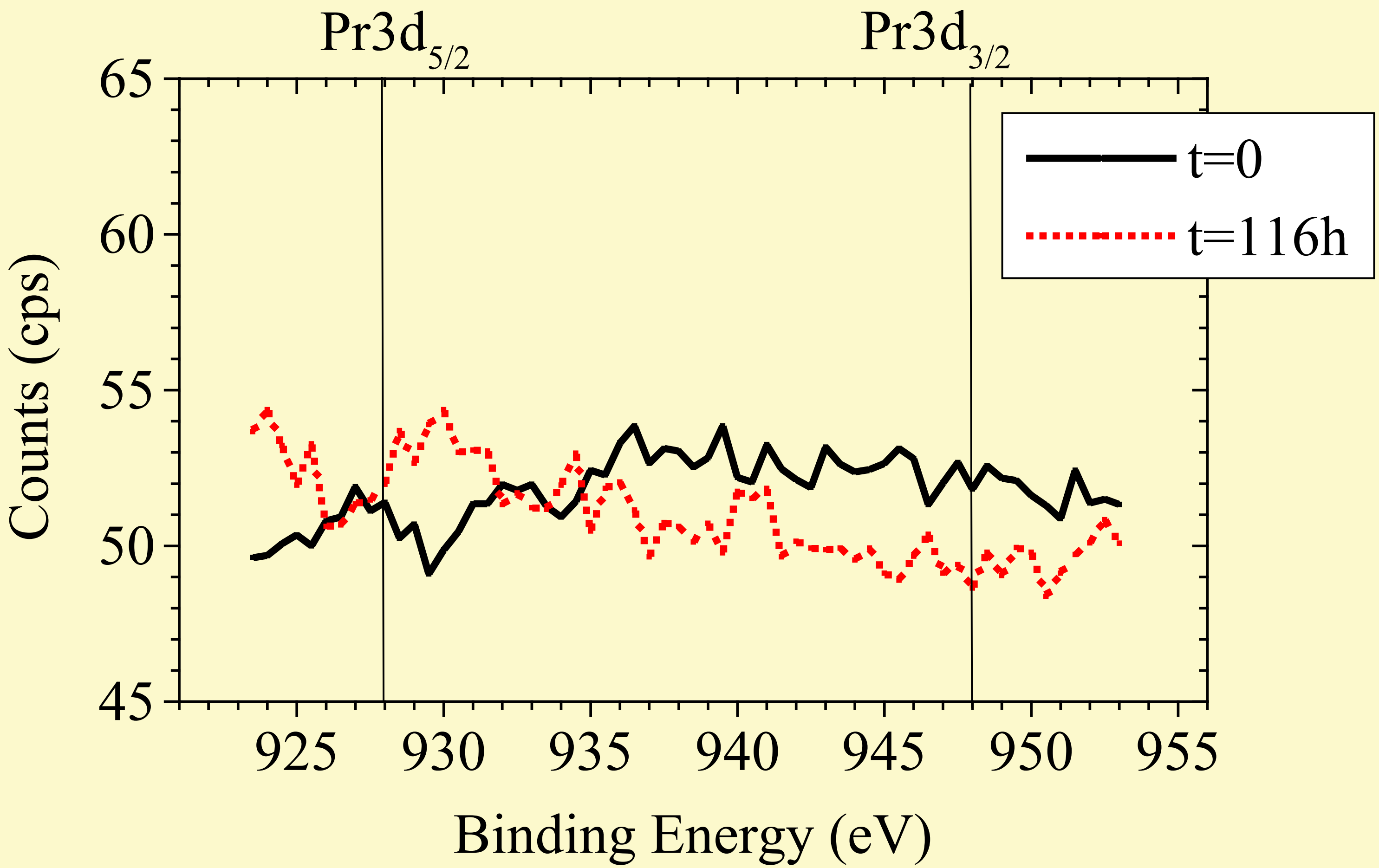
Pd/CaO/Pd/H2



Average Permeation Rate and Products

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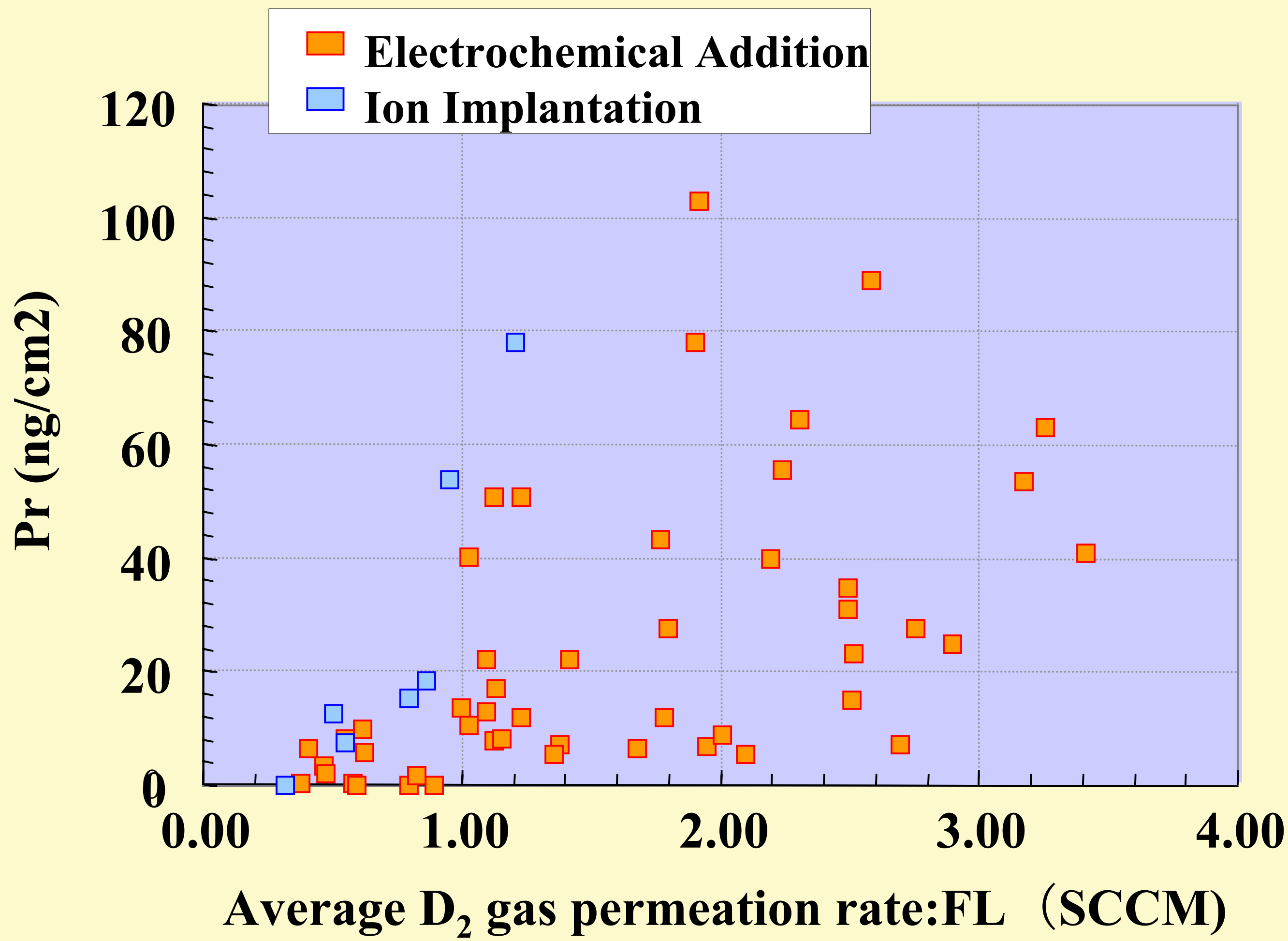
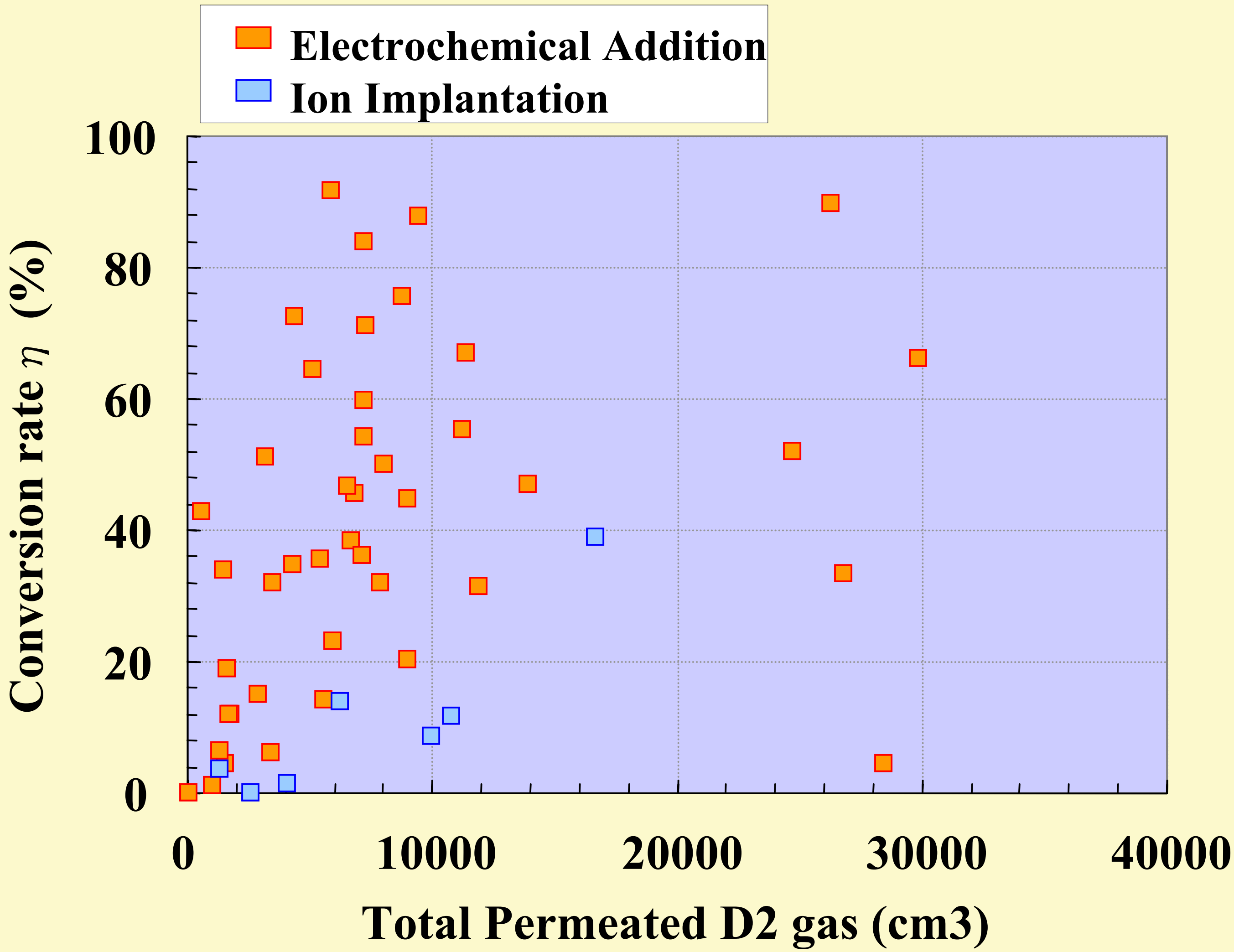


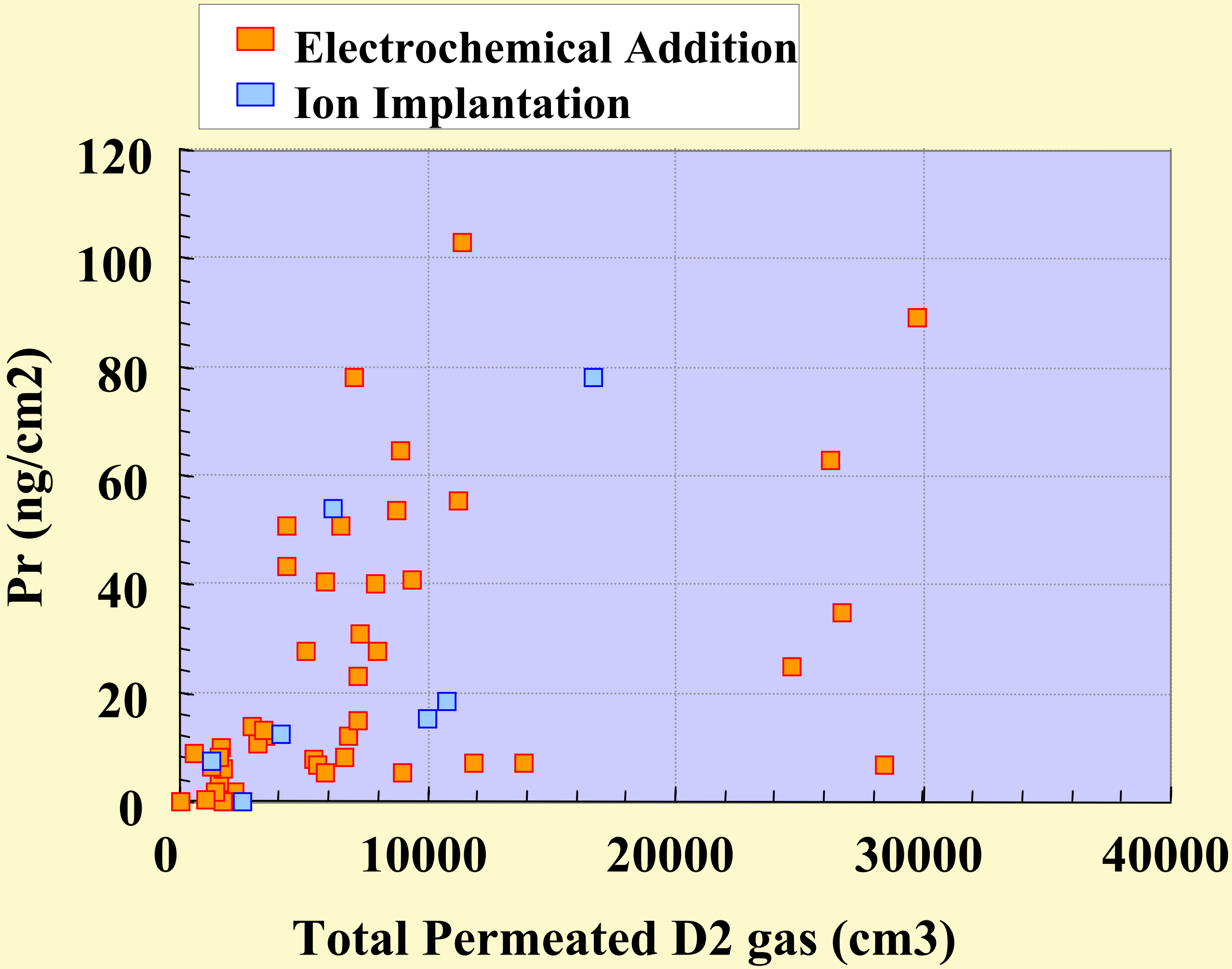
Total Permeated D2 Gas and Conversion Rate

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



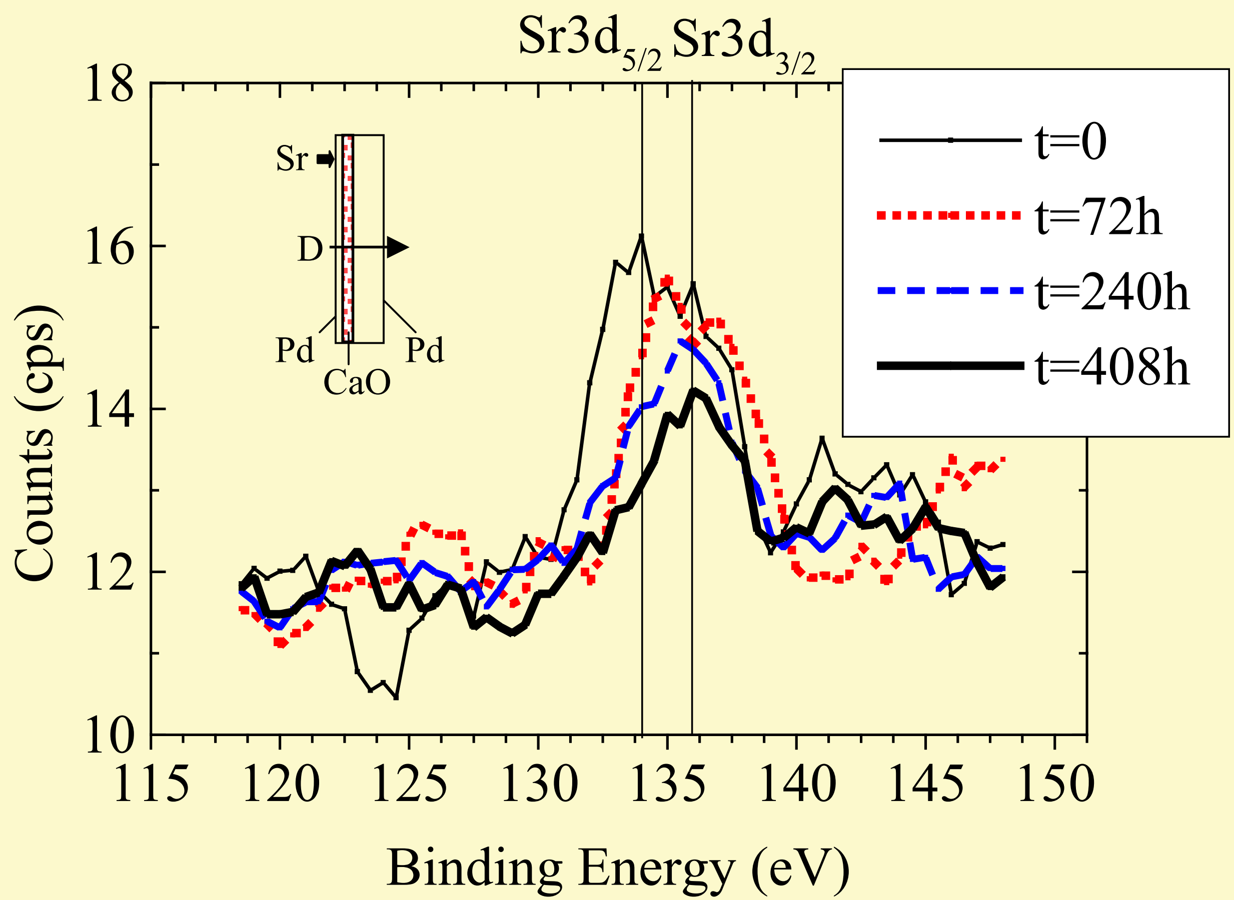
Total Permeated D2 Gas and Products

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



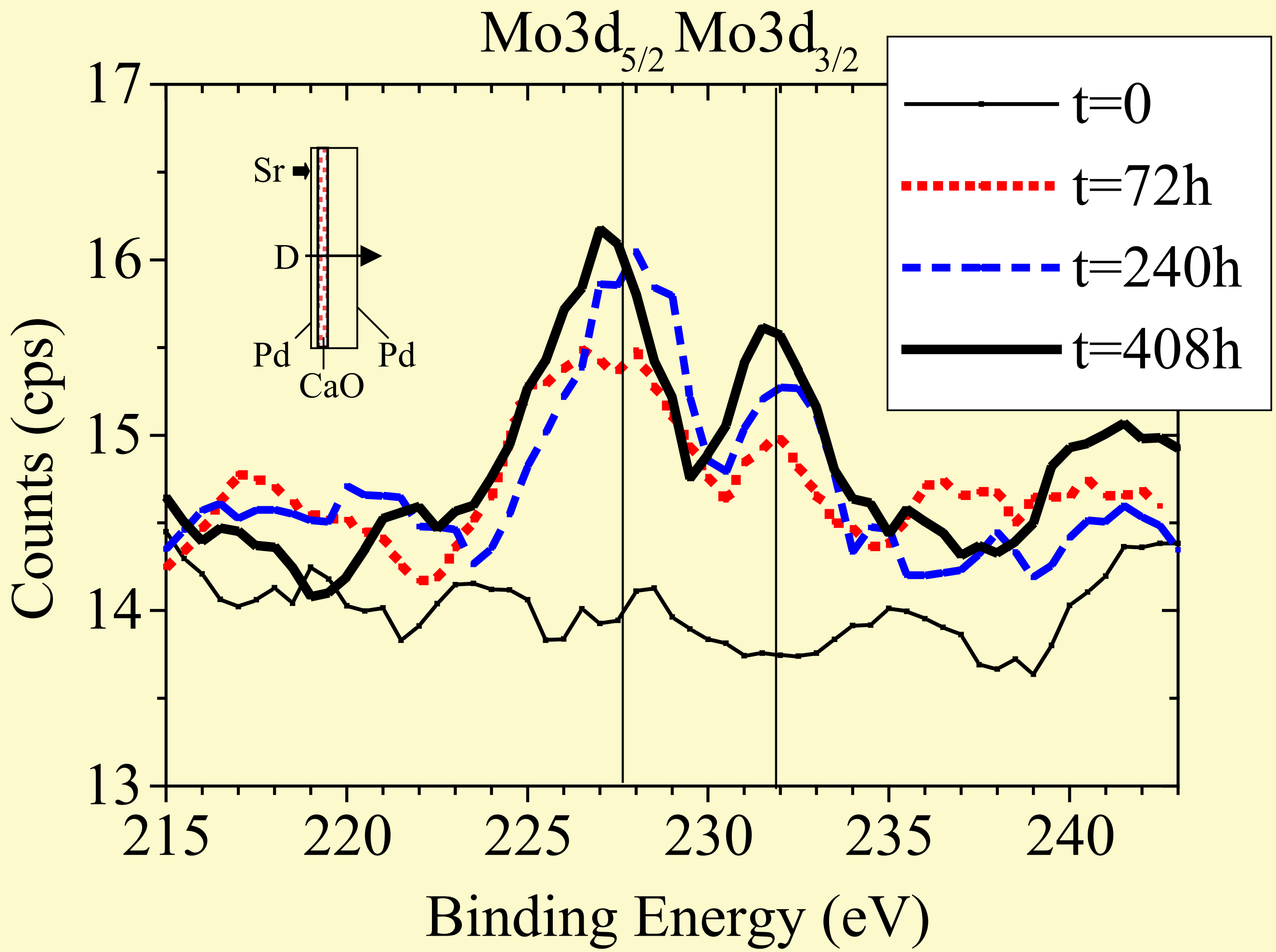
Change of XPS Spectrum of Sr

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



Change of XPS Spectrum of Mo

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



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

MITSUBISHI HEAVY INDUSTRIES, LTD.
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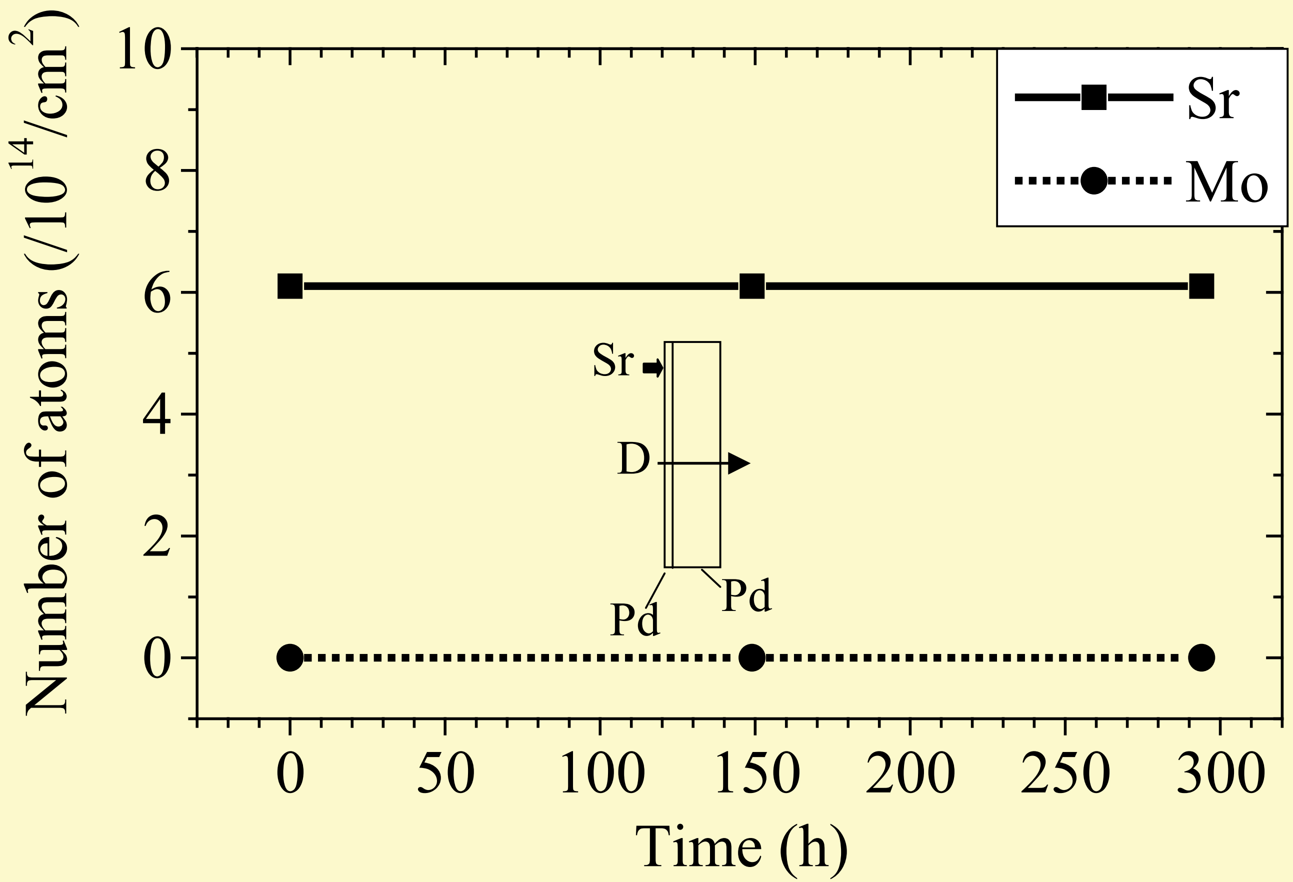

XPS Spectrum of Sr (No CaO)

MITSUBISHI HEAVY INDUSTRIES, LTD.
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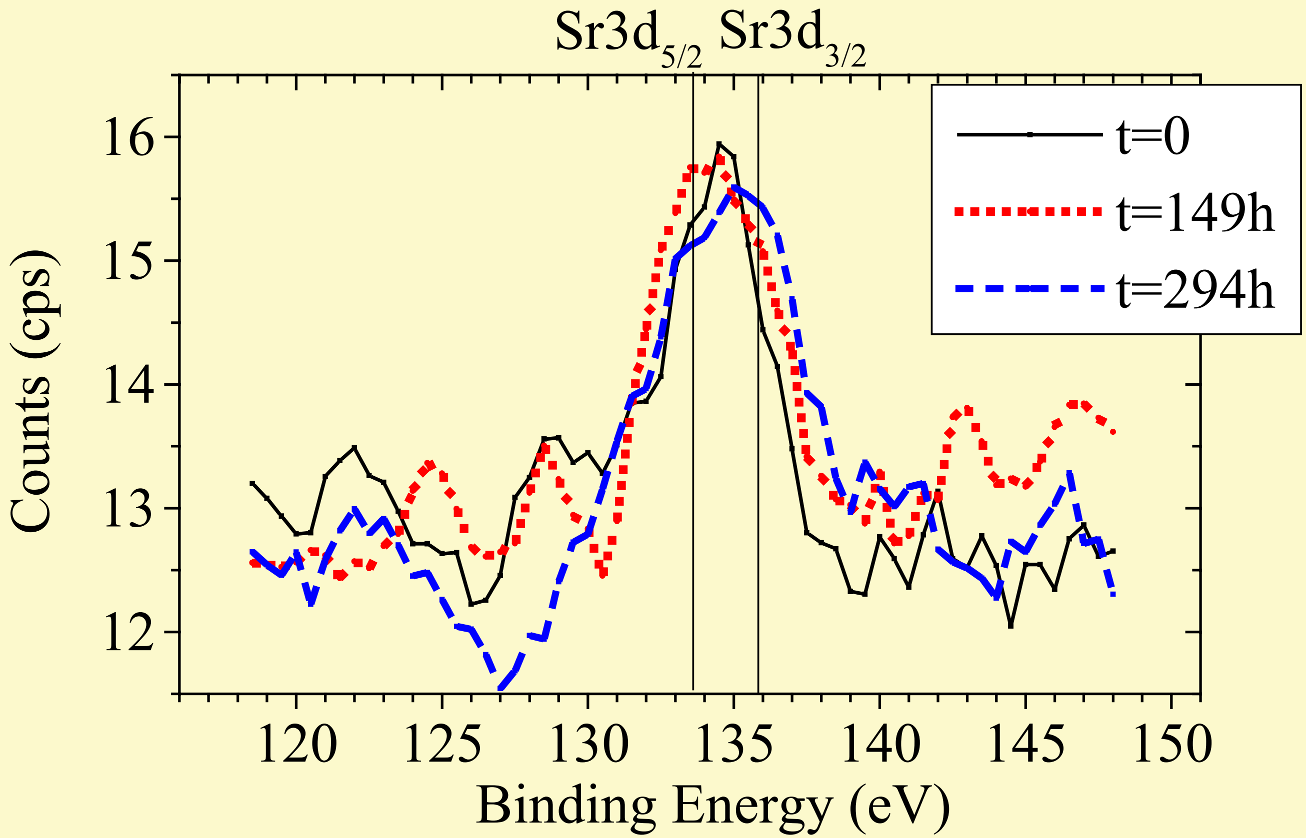
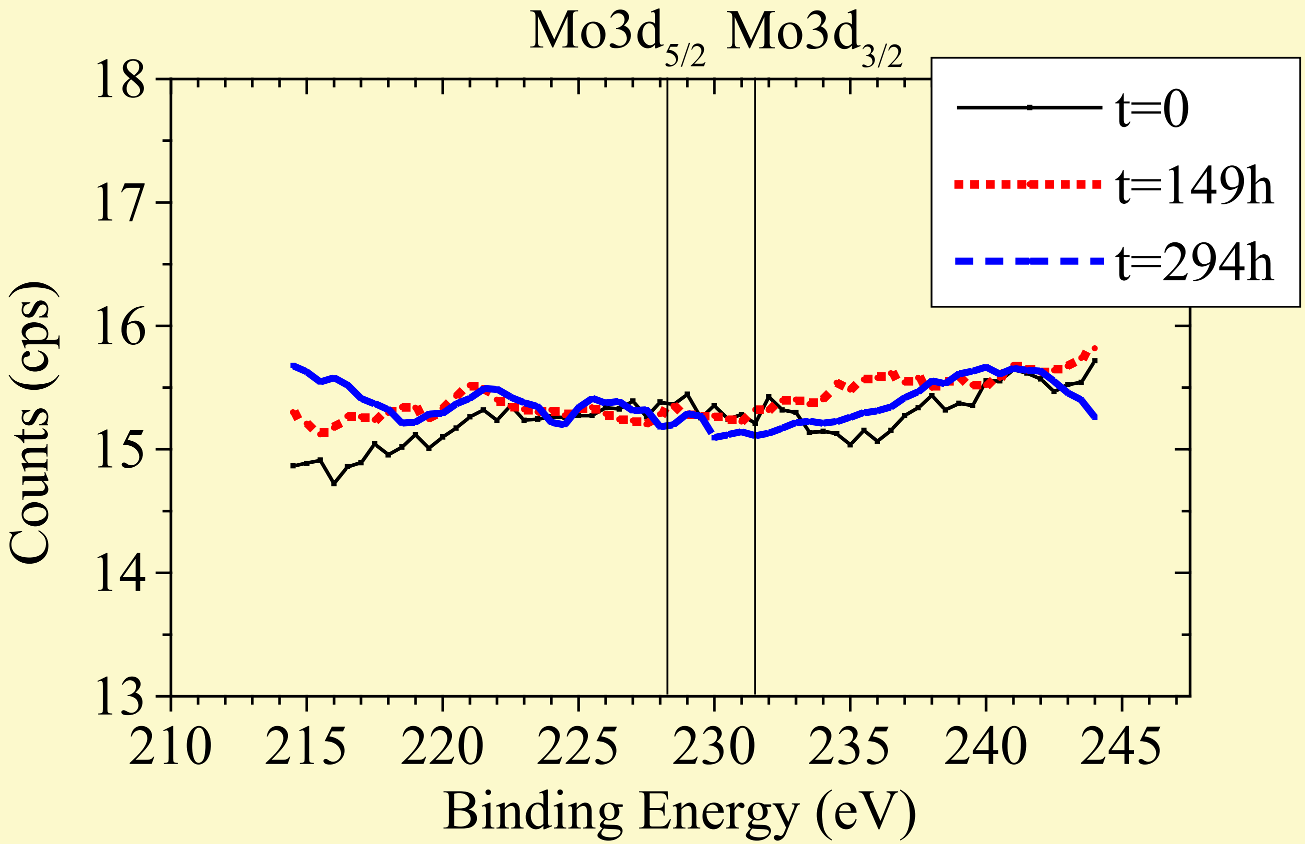
Pd/Pd/D2



XPS Spectrum of Mo (No CaO)

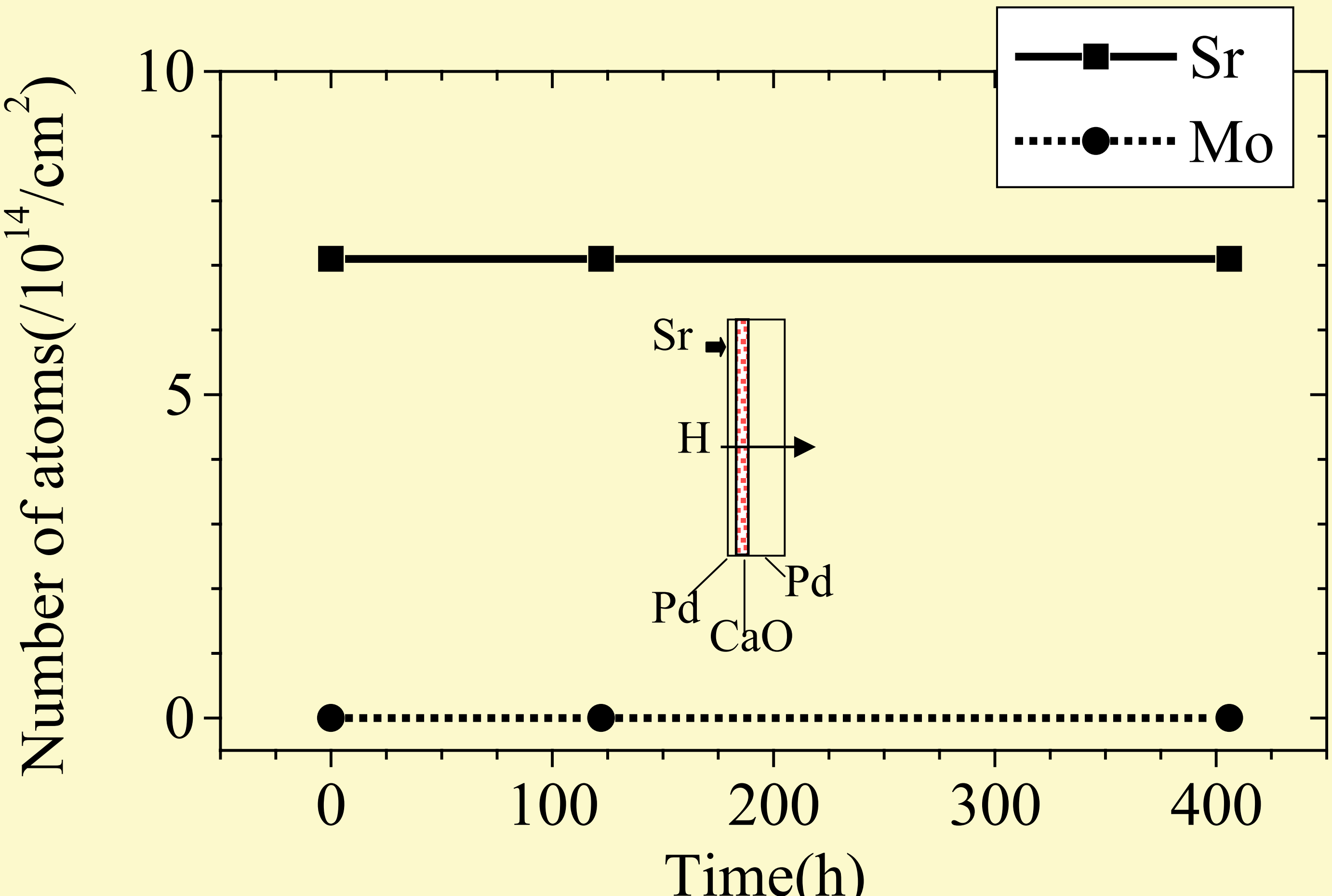
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

Pd/Pd/D2



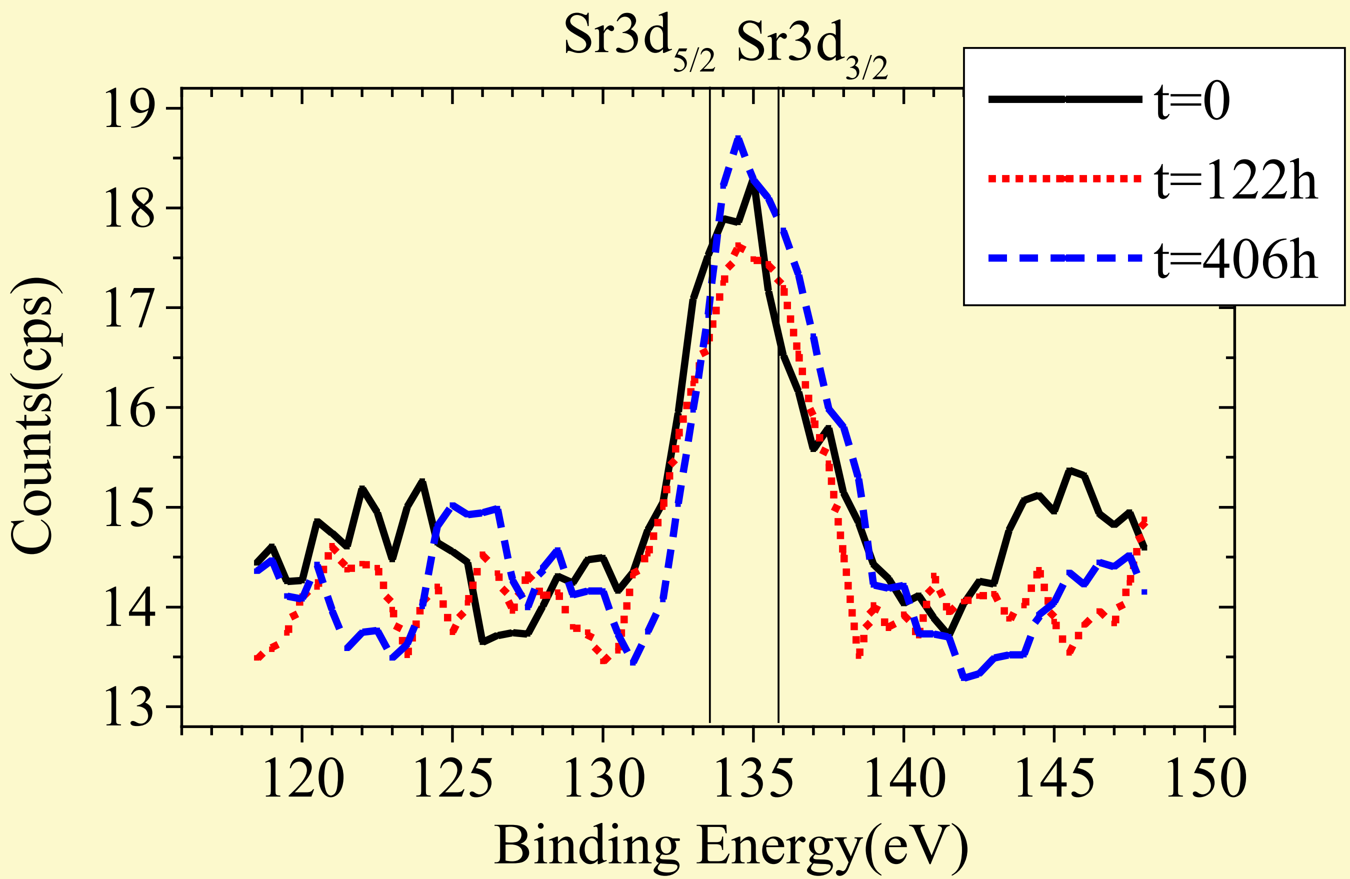
Time Variation in number of Sr and Mo atoms in the case of H_2 Permeation through Pd Complex with added Sr

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



XPS Spectrum of Sr (H_2 Permeation)

Pd/CaO/Pd/H2



XPS Spectrum of Mo (H_2 Permeation)

MITSUBISHI HEAVY INDUSTRIES, LTD.
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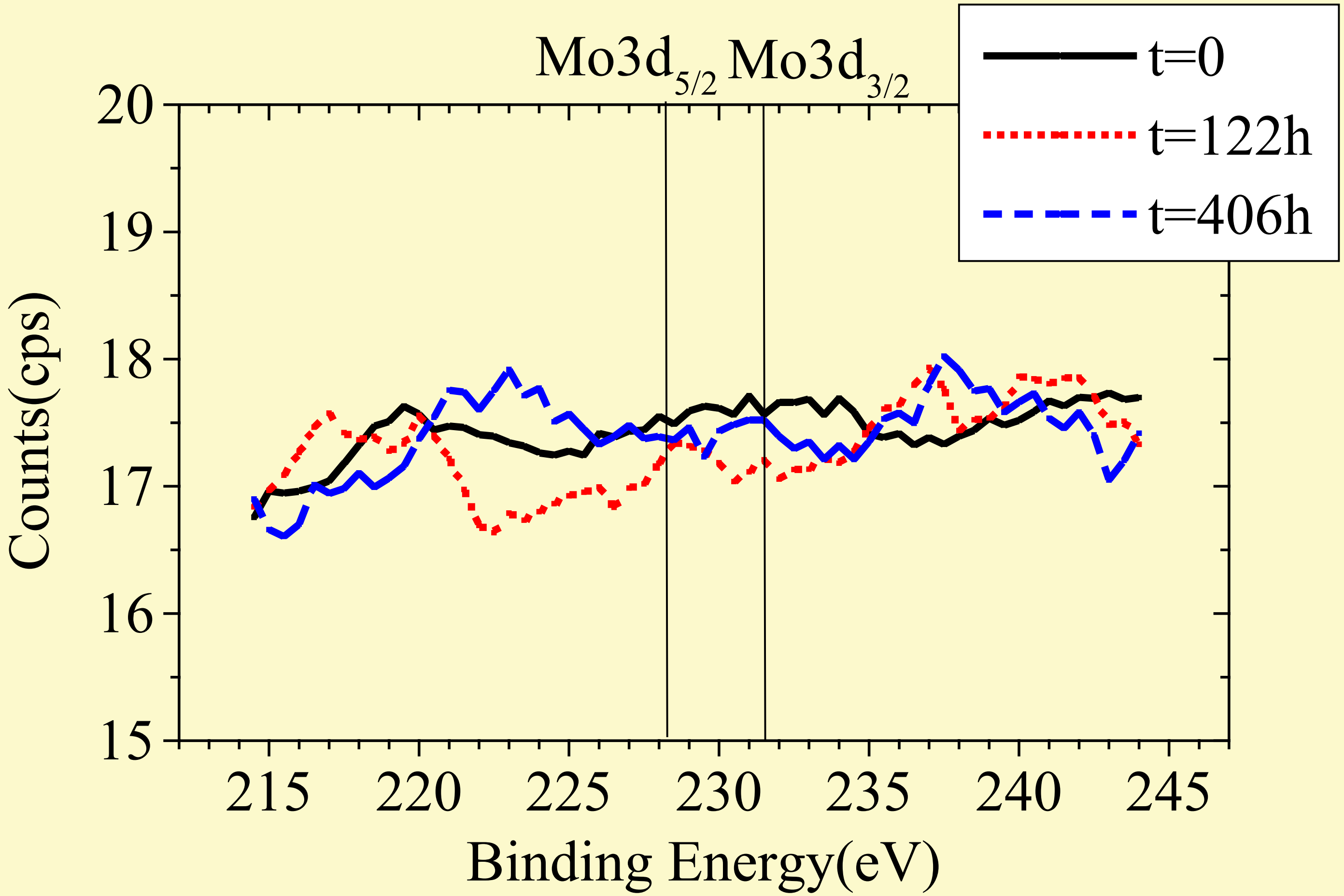
Pd/CaO/Pd/H2



Separation of the Products and Contaminants(3)

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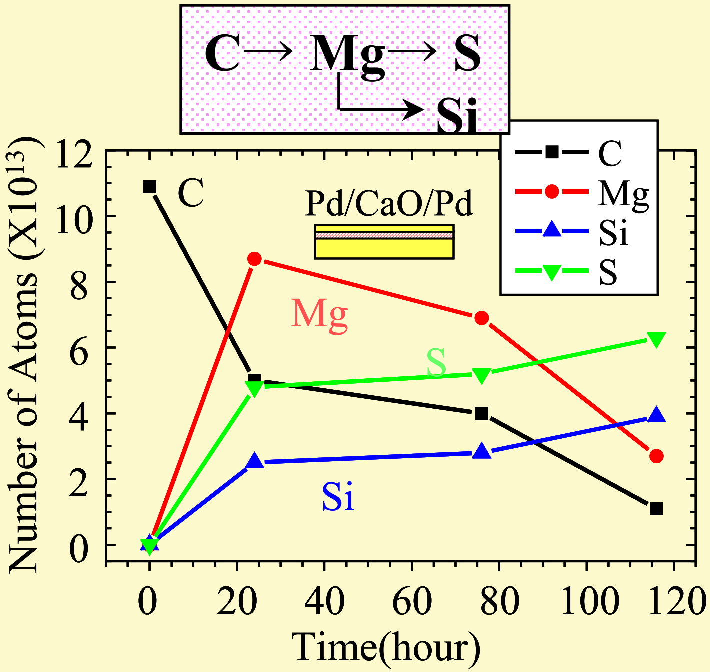
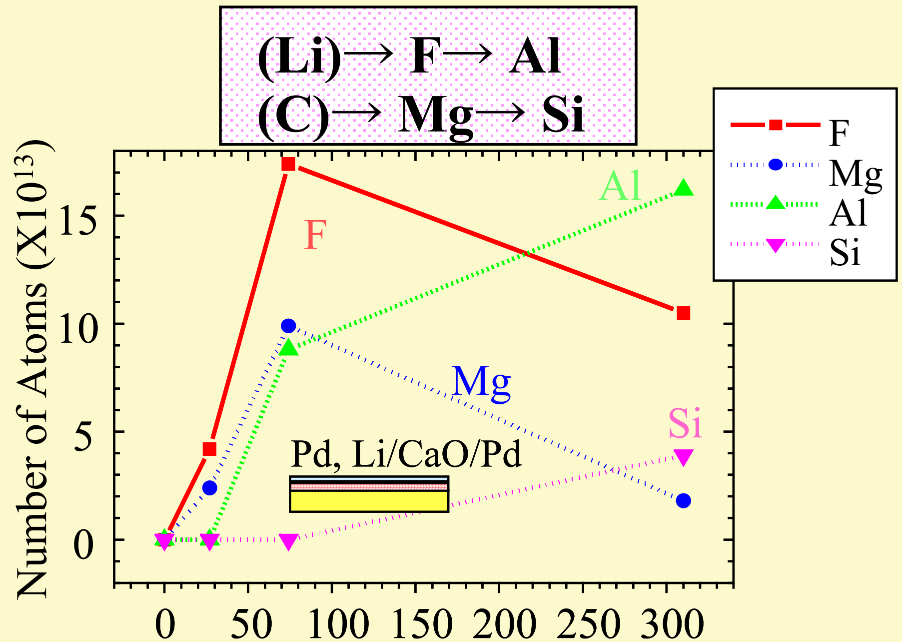
3. Variation of Detected Elements depending on the given Elements

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