# Pd Complex

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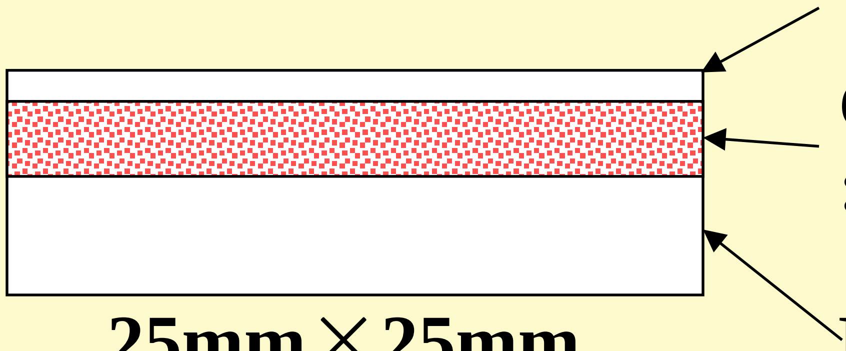
## Pd complex containing CaO




25mm×25mm

# Transmutation of Cs into Pr

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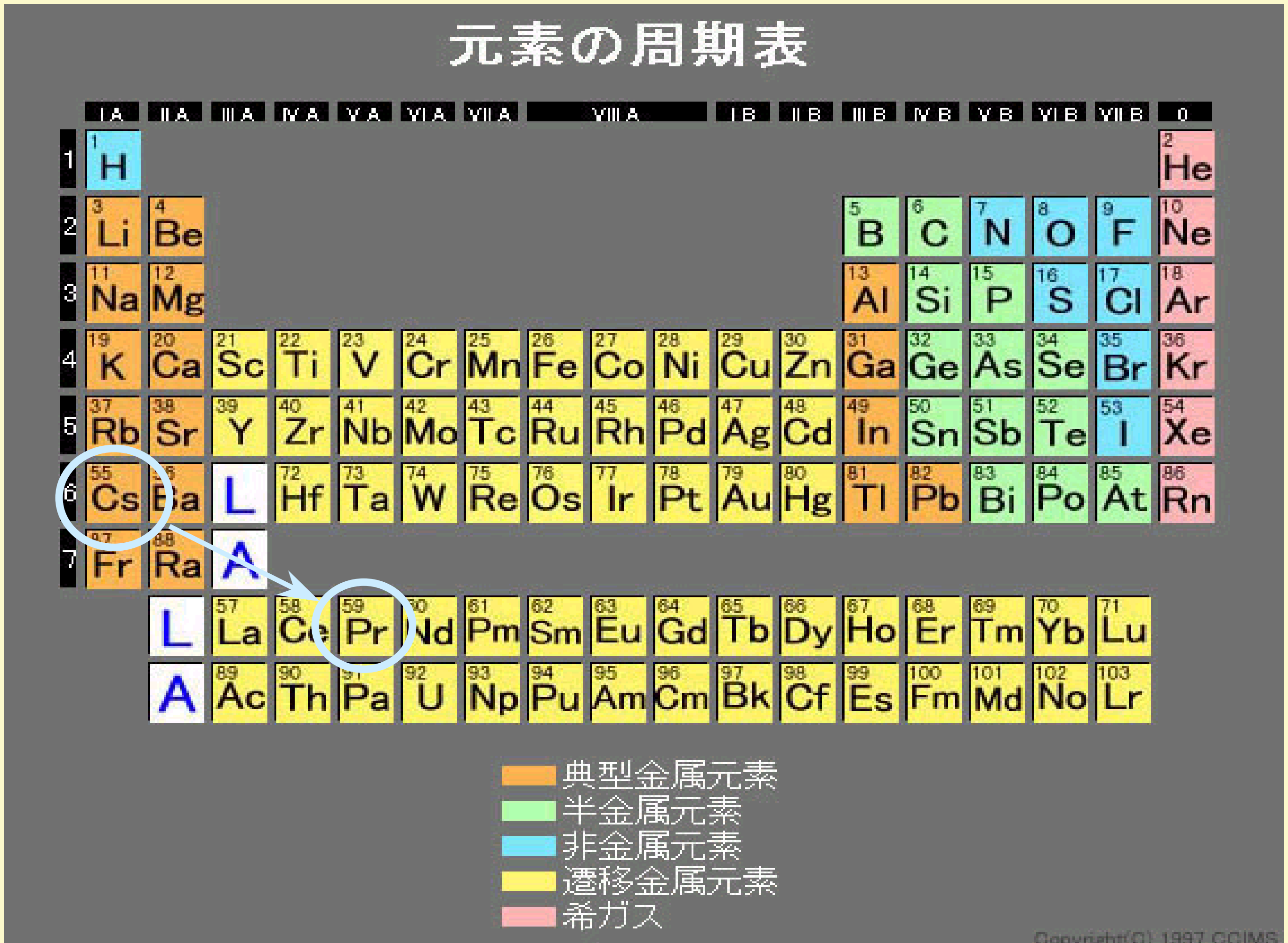


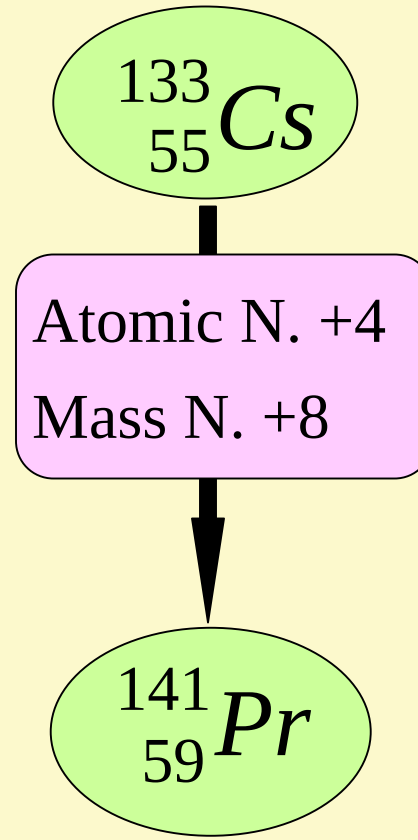

# Transmutation of Sr into Mo

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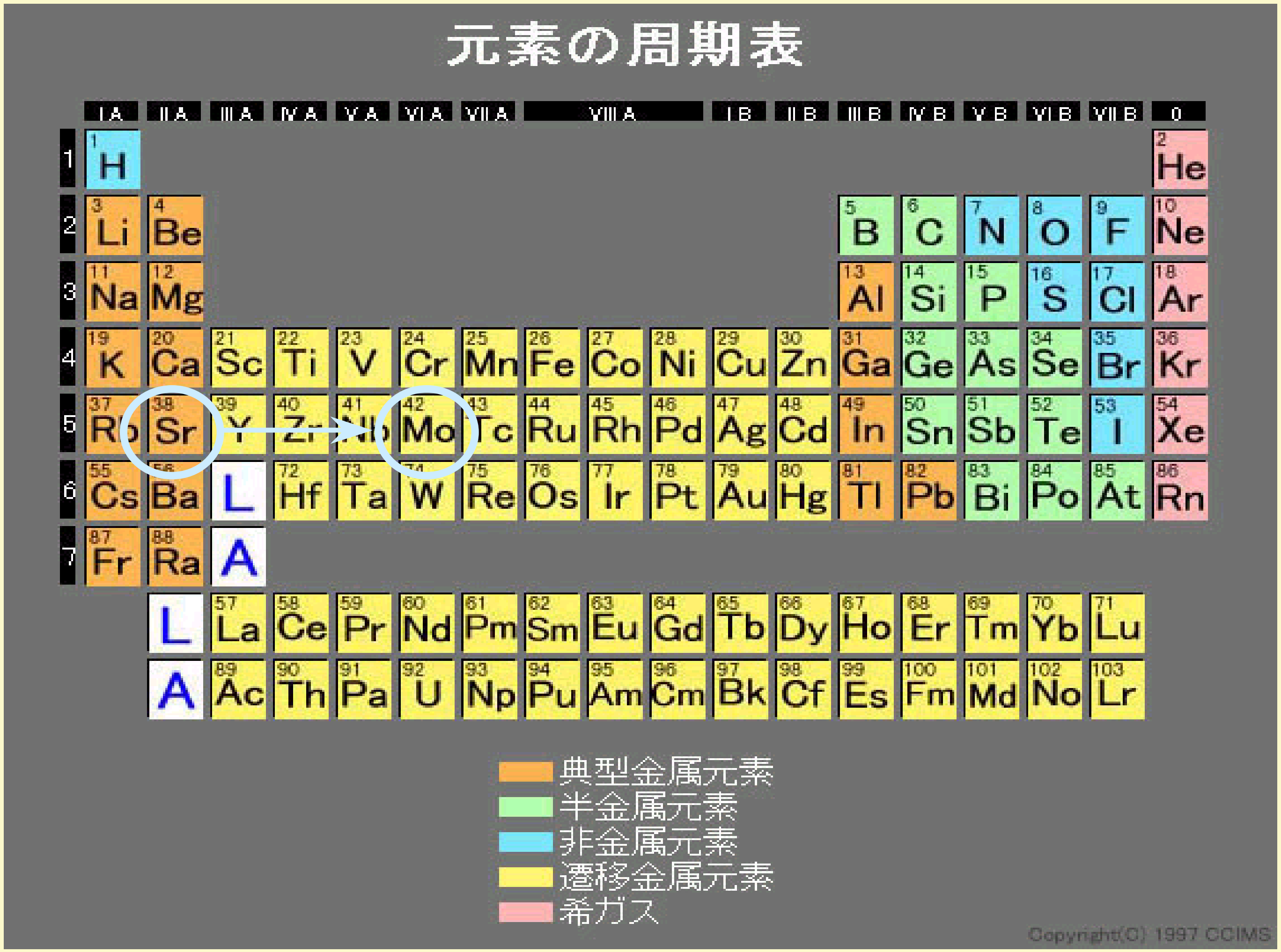


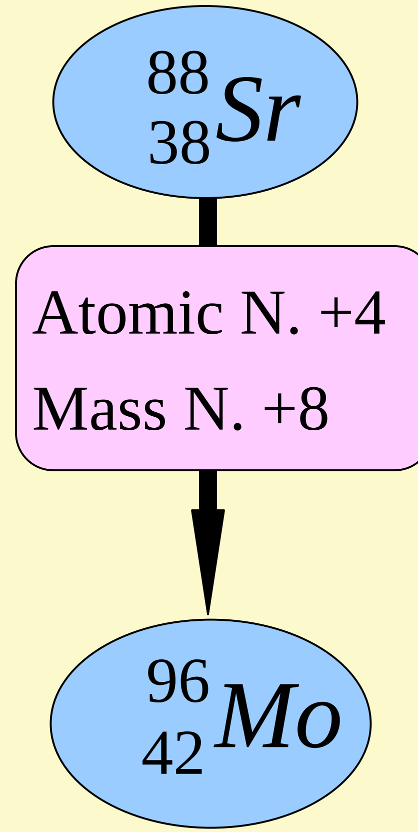

# Transmutation of Ba into Sm

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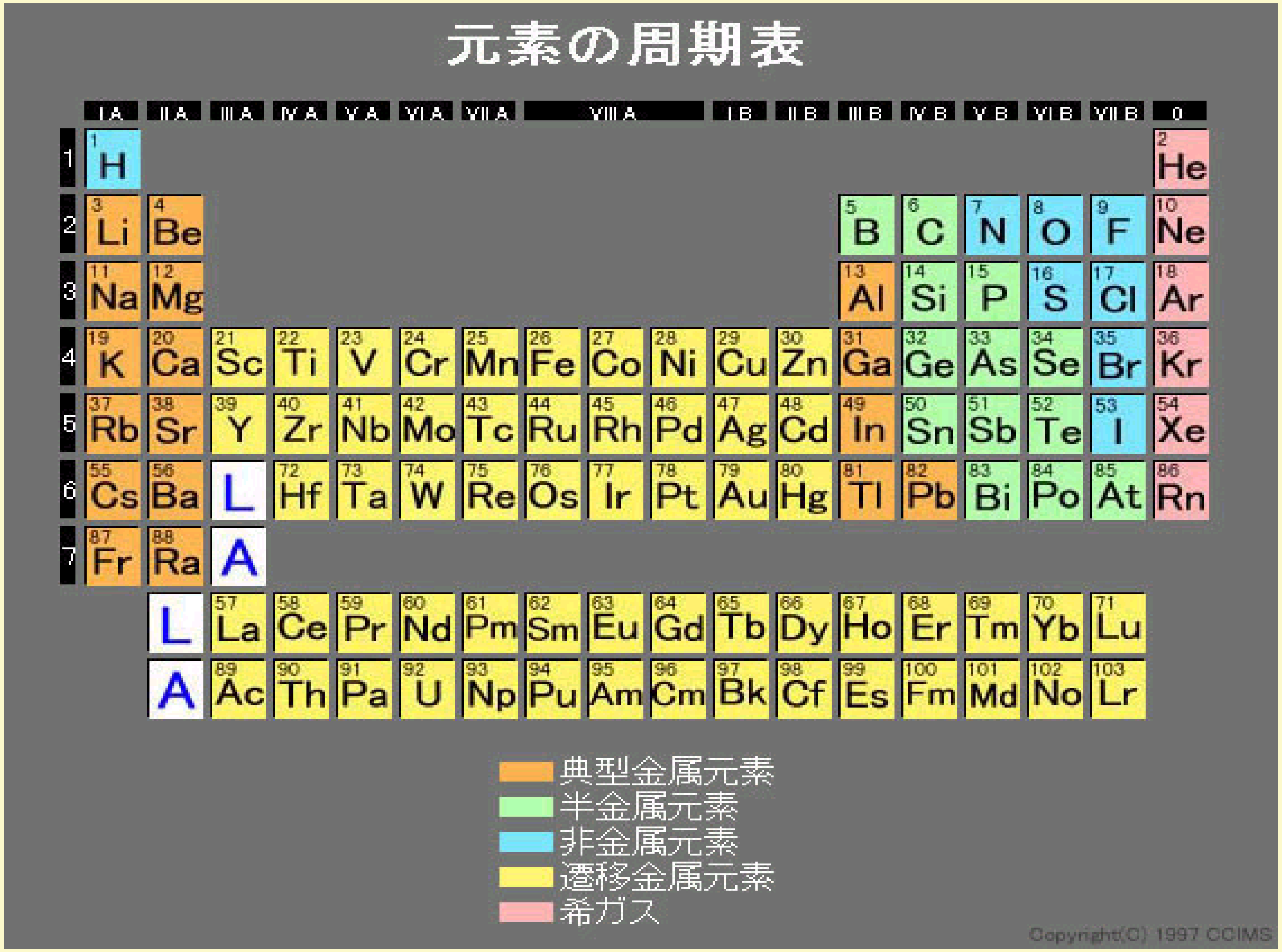
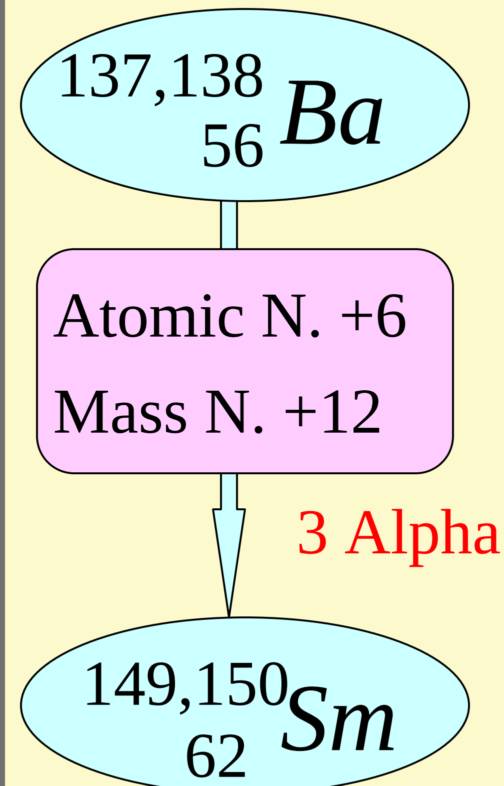


元素の周期表

| | 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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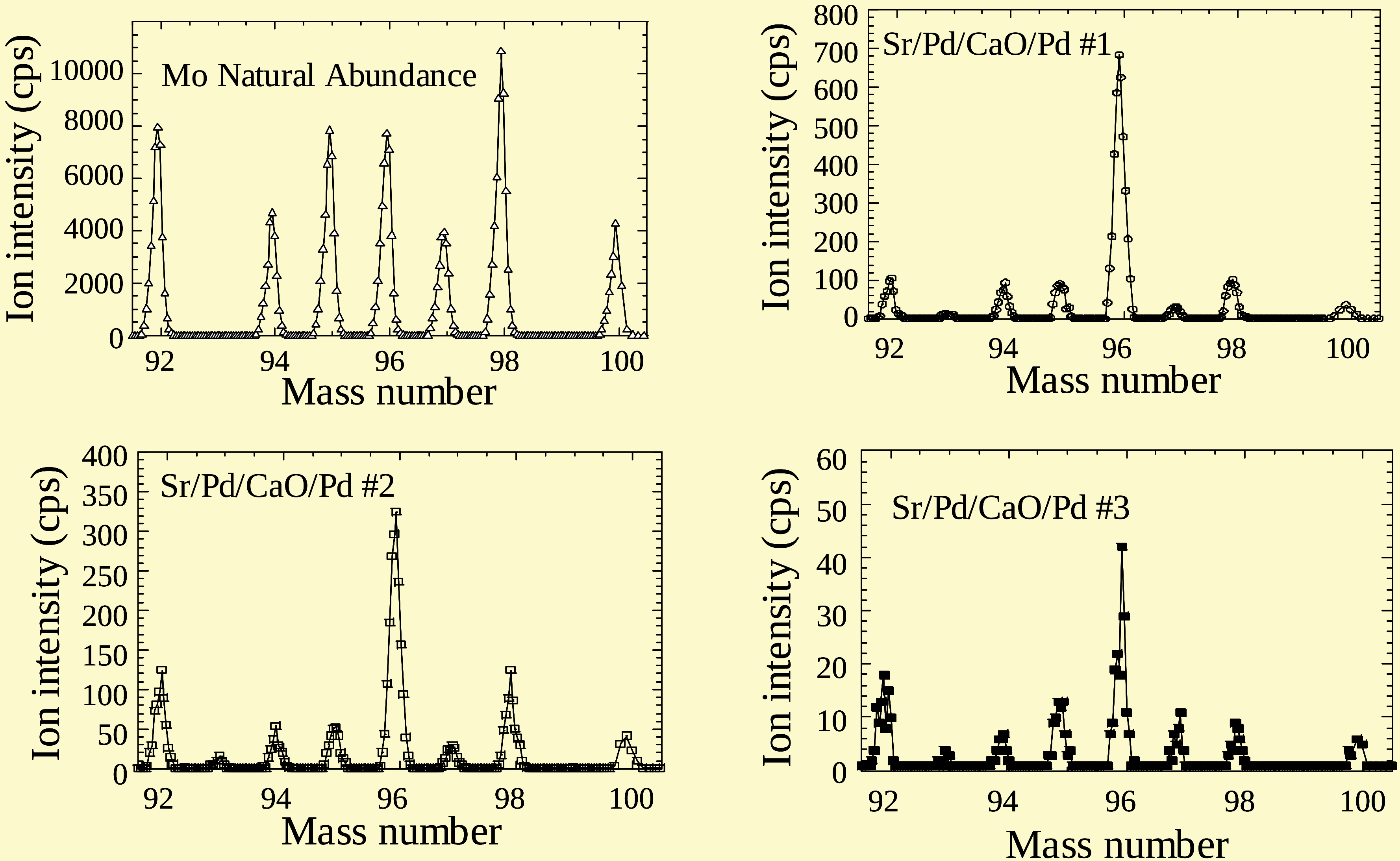

# Evaluation of $D_2$ Gas Flow Rate

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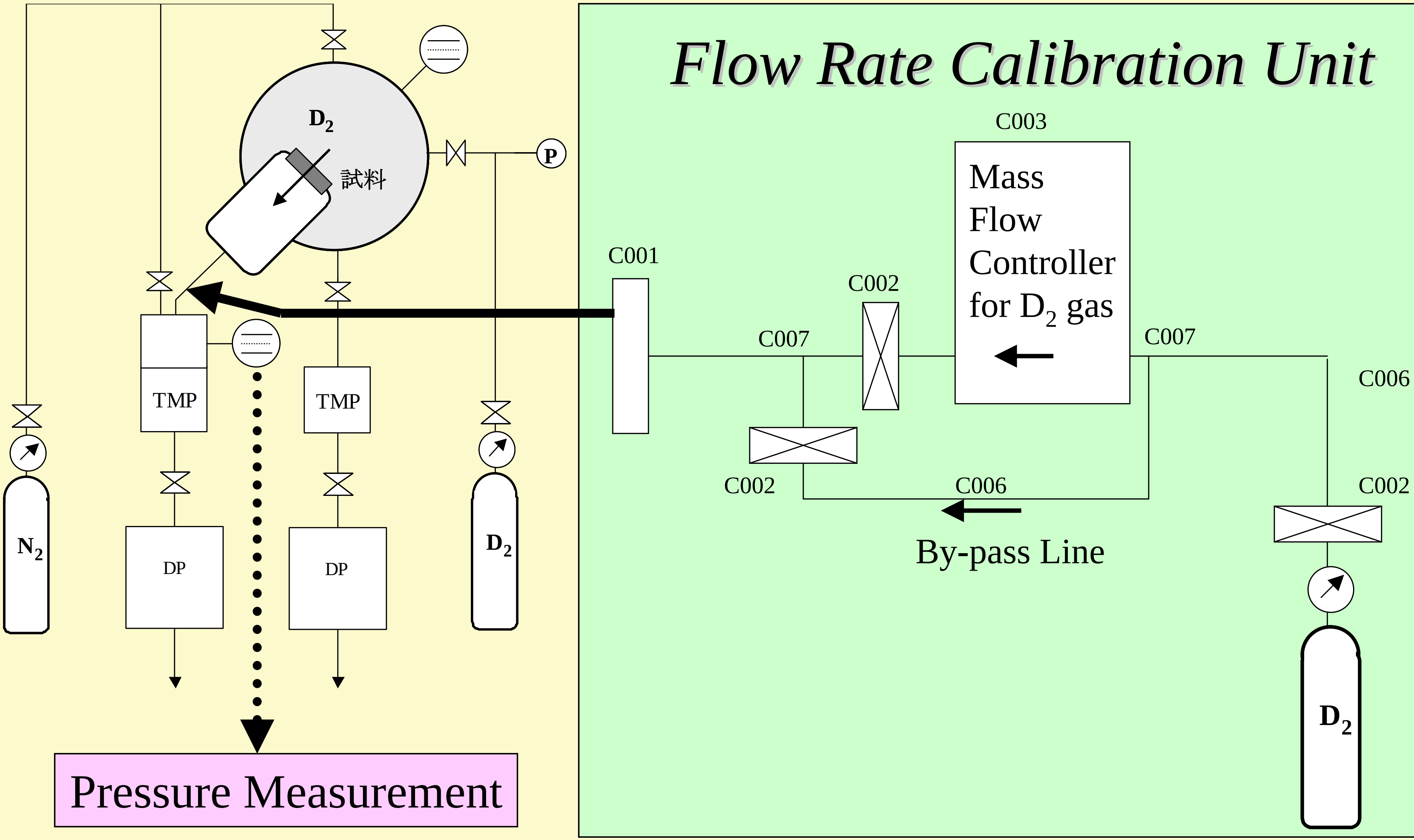

# Identification of Pr by TOF-SIMS(2)

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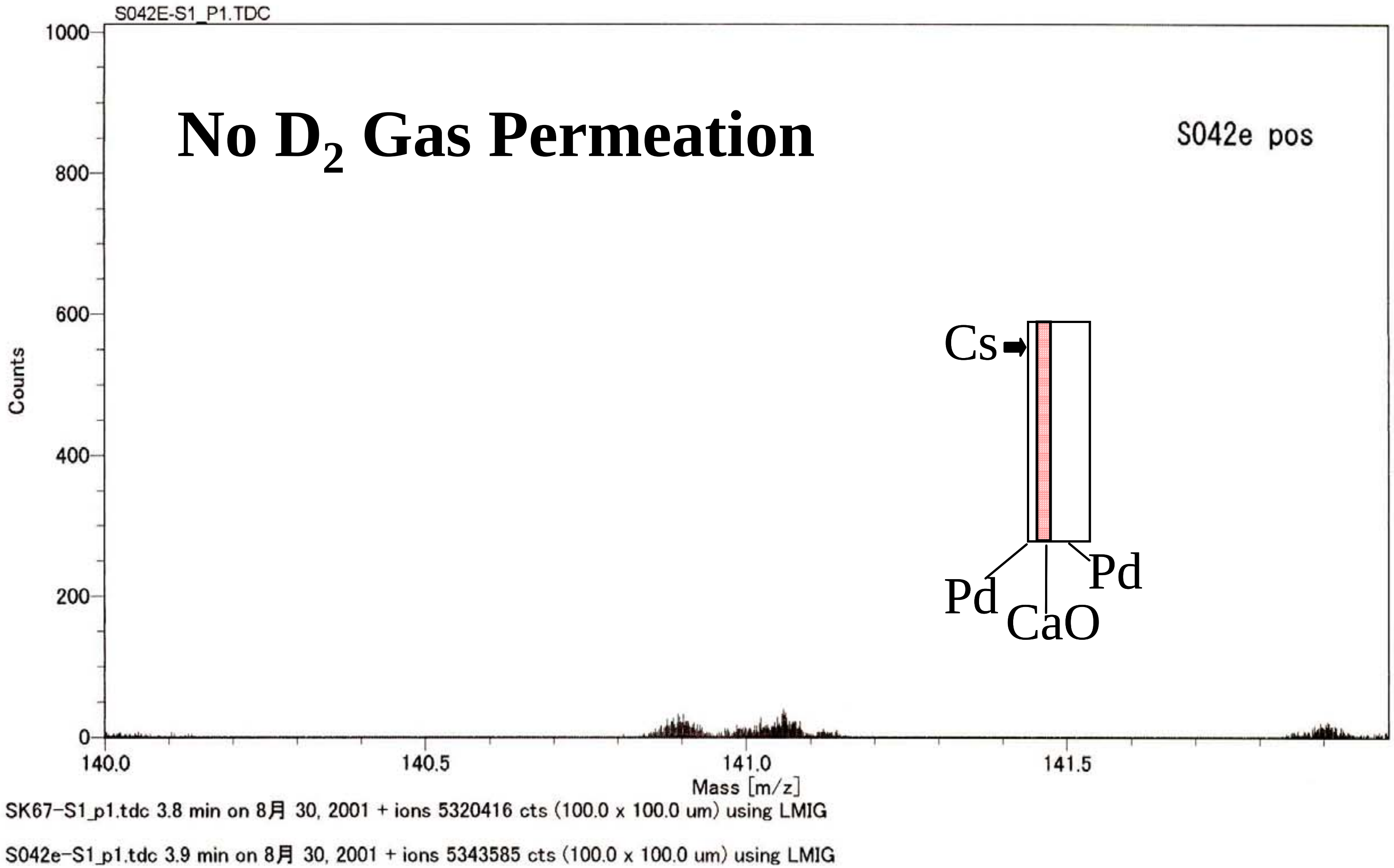


**TOF-SIMS device (TRIFT™ Ⅱ;ULVAC-PHI)**

# Cs添加Pd多層膜重水素透過後

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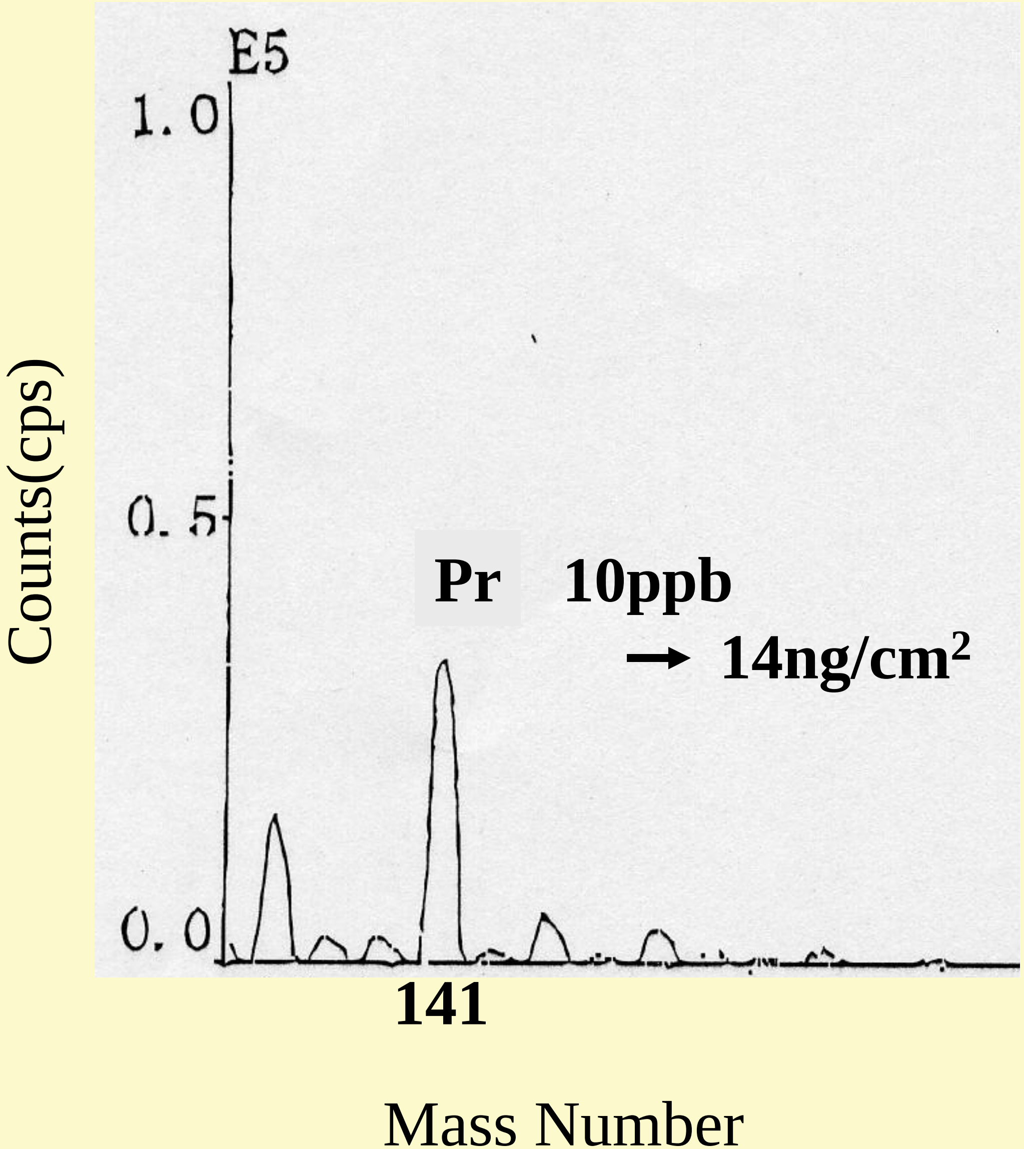


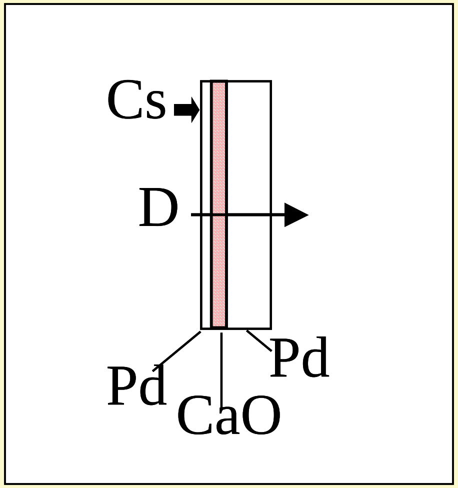


→ Prを検出、定量

# Transmutation of Tm into Ta

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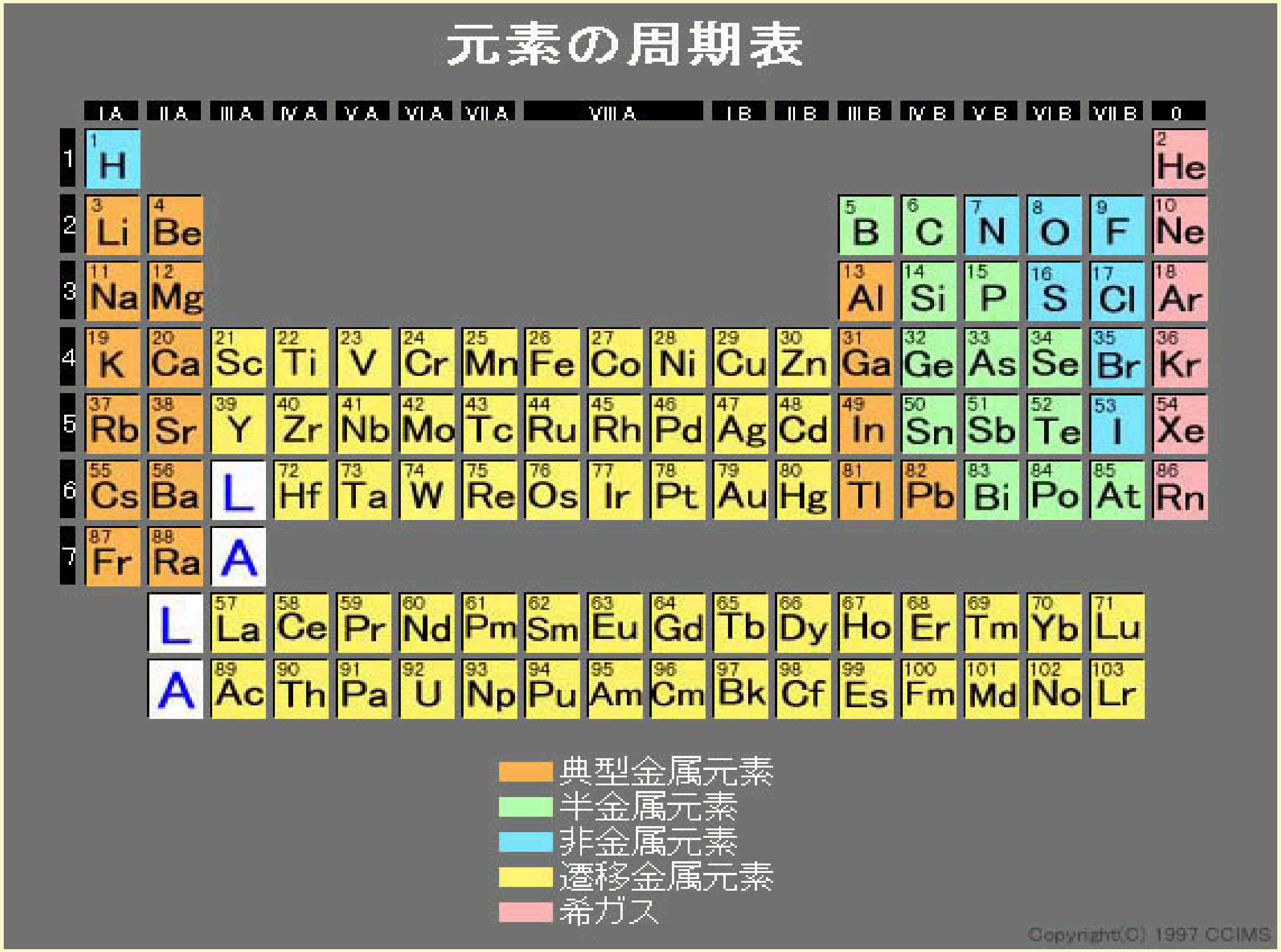
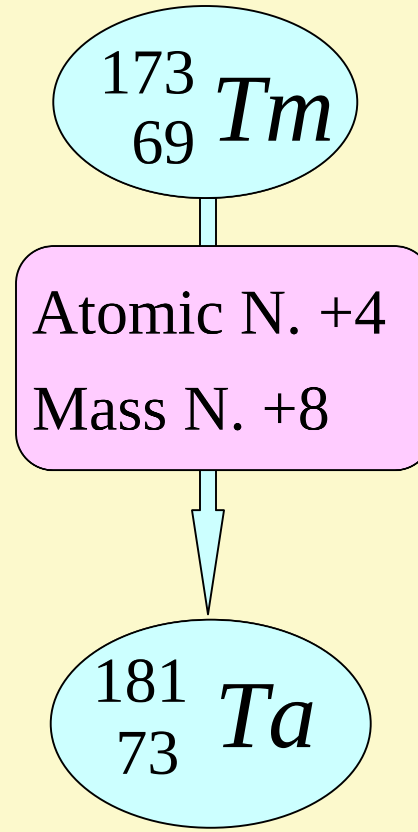
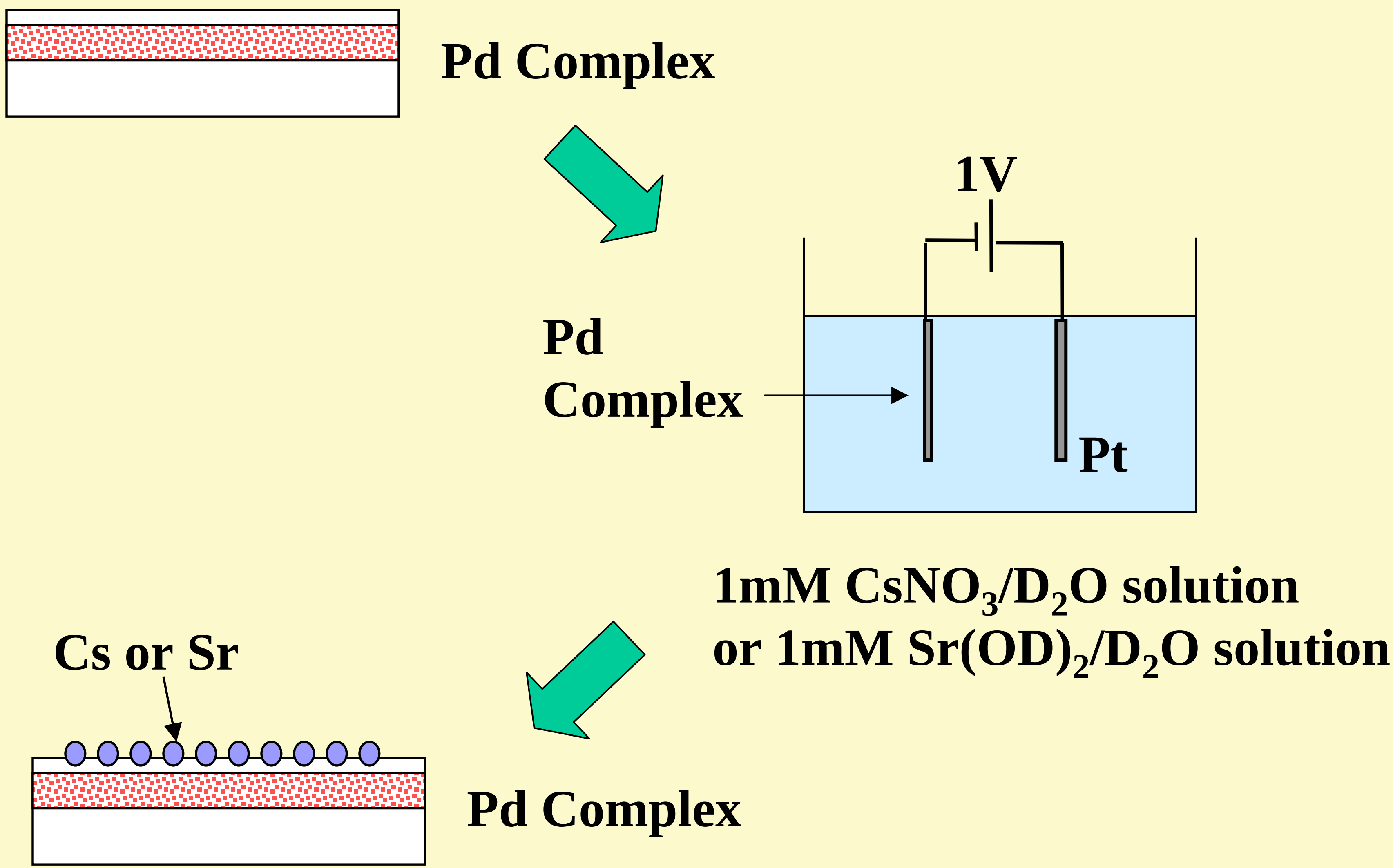


# Element Addition (1: Electrolyte Addition)





# Element Addition
# (2: Ion Implantation)

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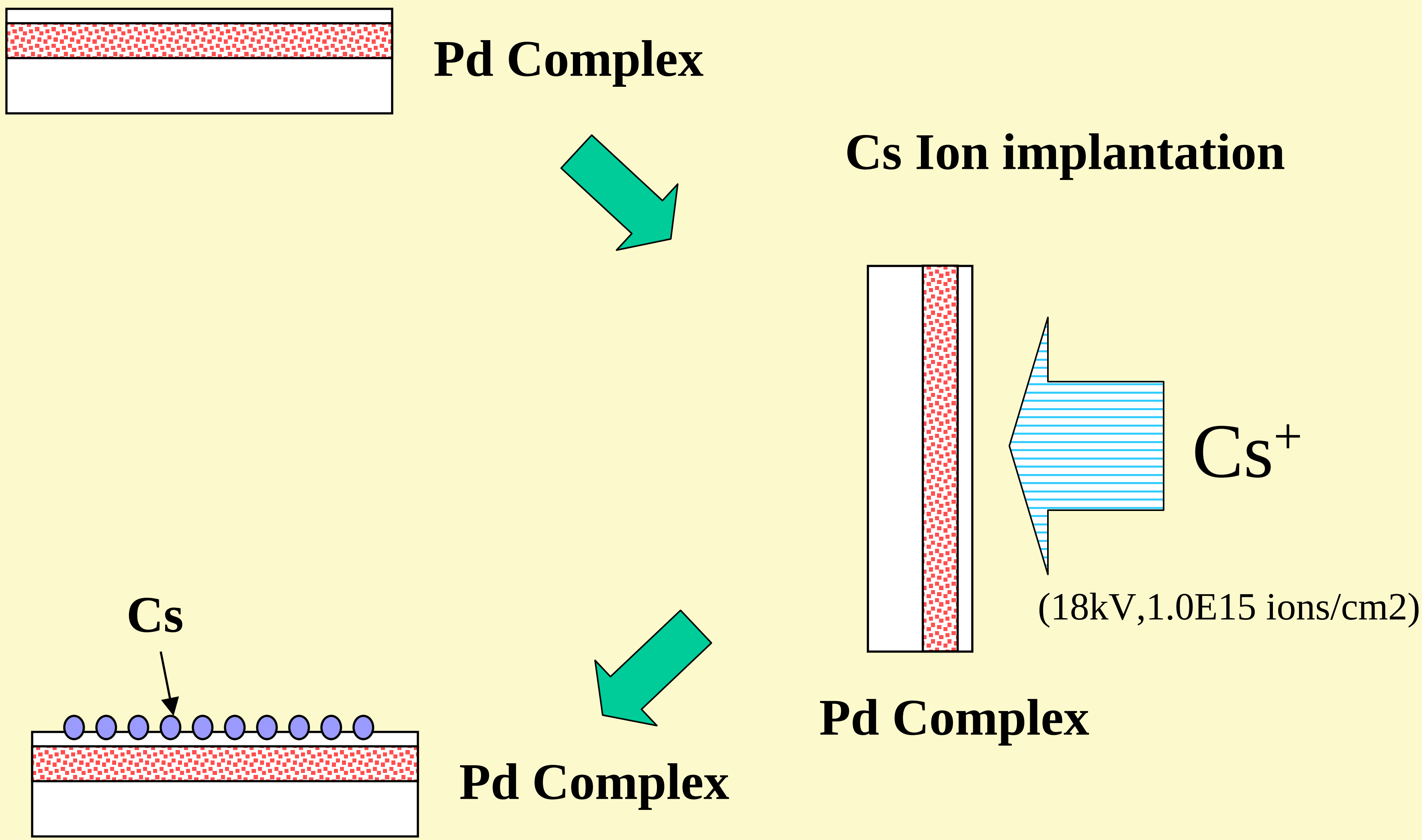

# Identification of Pr by XANES

XANES(X-ray Absorption Near Edge Structure)

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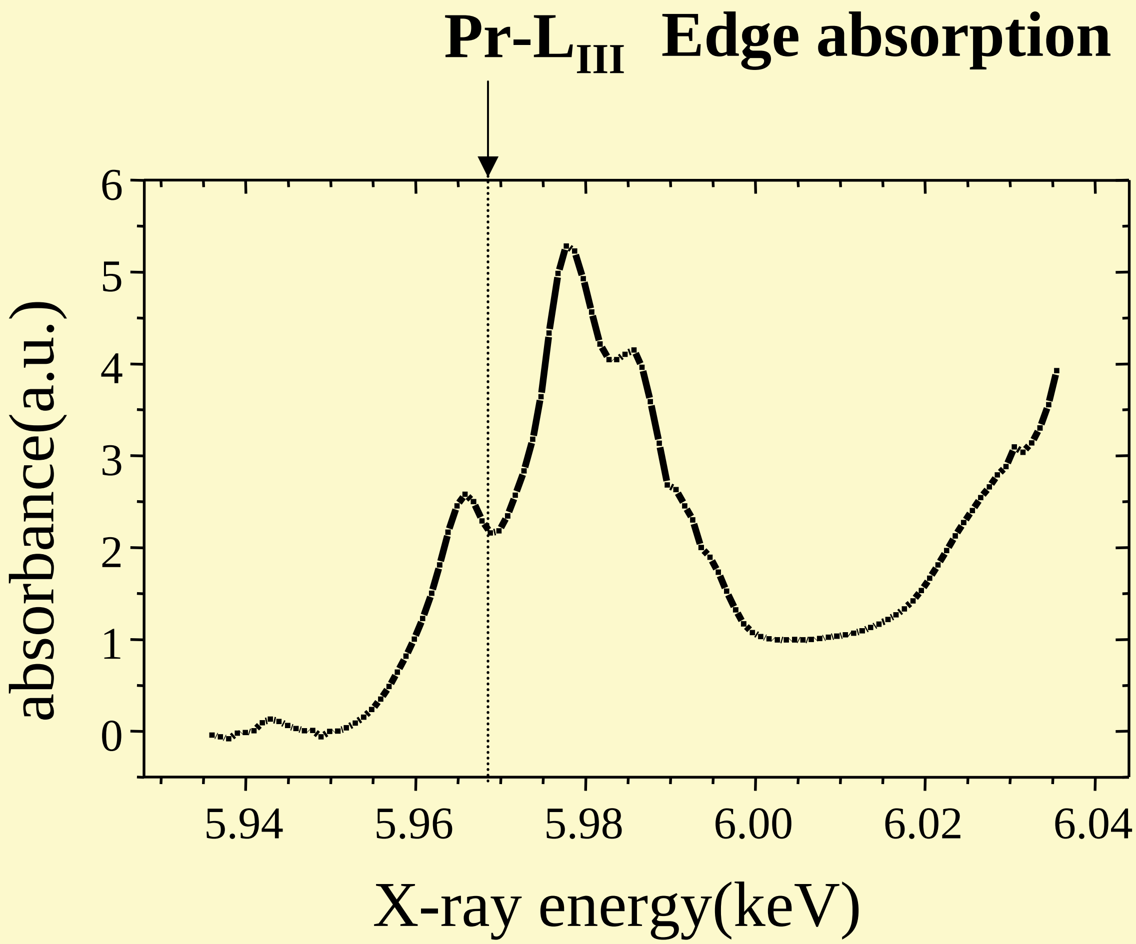
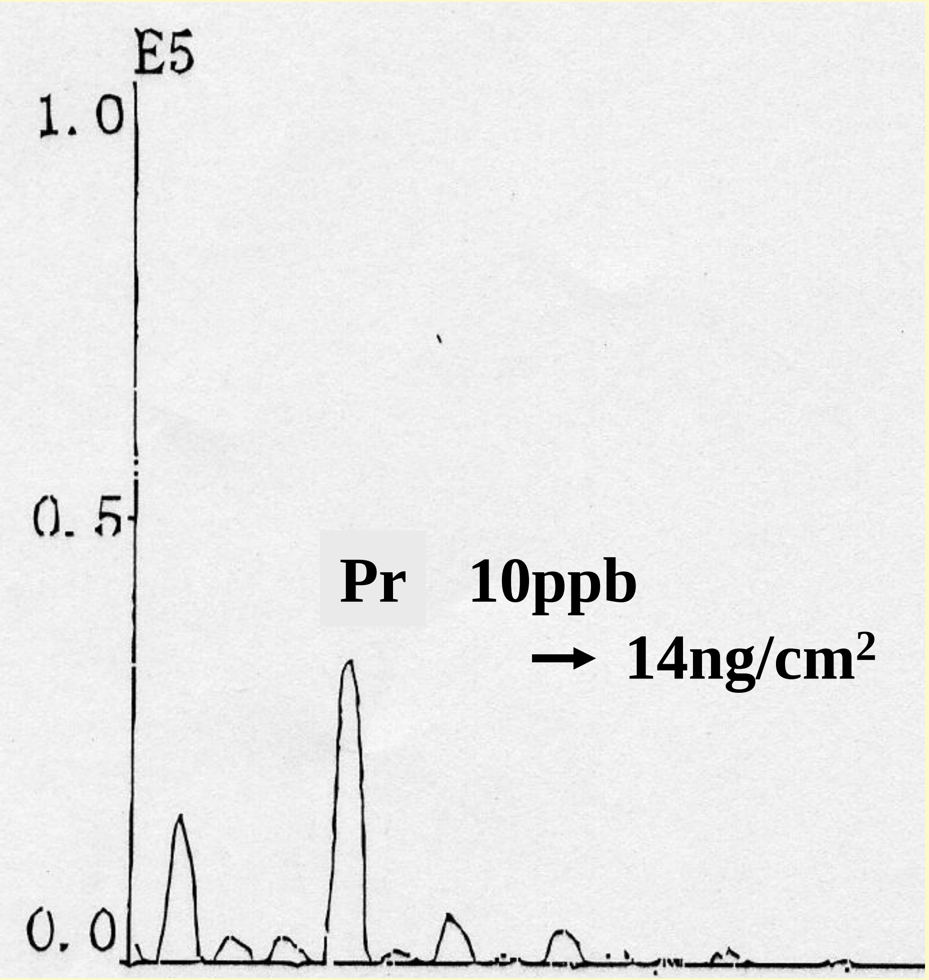



**BL-9A Line, KEK, Tsukuba, Japan**

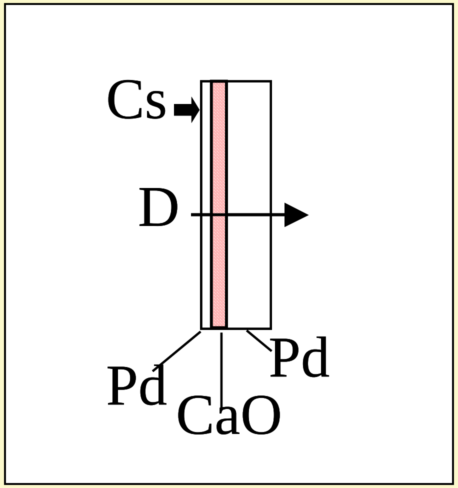
# Identification of Pr by ICP-MS






Counts(cps)

Mass Number




→ 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)

↓

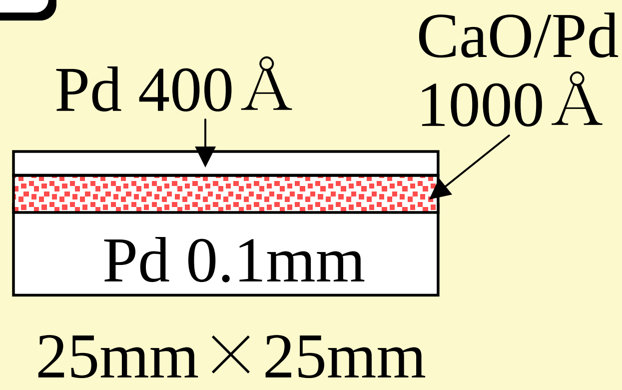
Washing the Sample with Aqua Regia (100sec)

↓

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

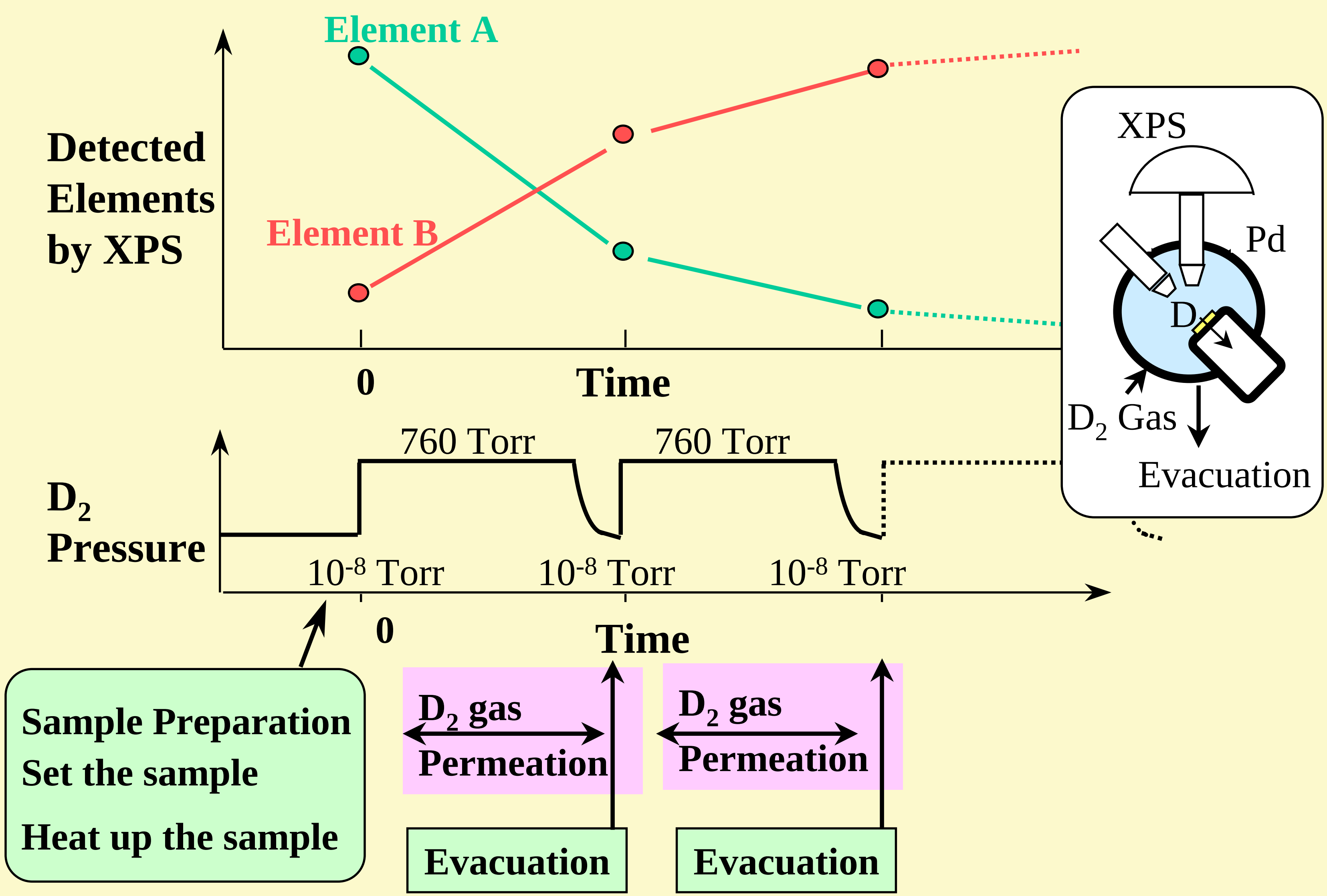
↓

Ion Beam Sputtering of Pd only (400 Å)



# Procedure of an Experiment





# Transmutation of Sr into Mo

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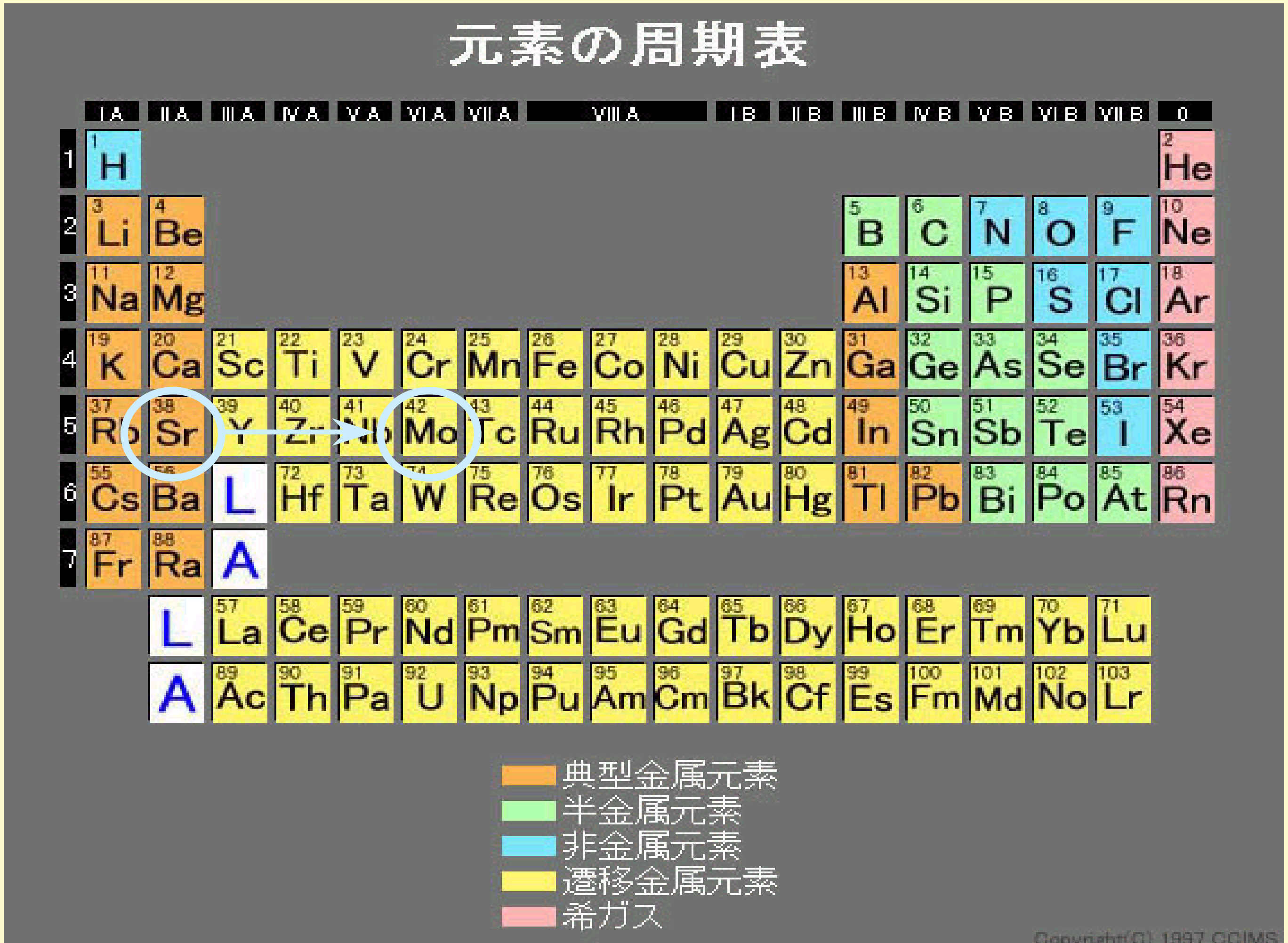


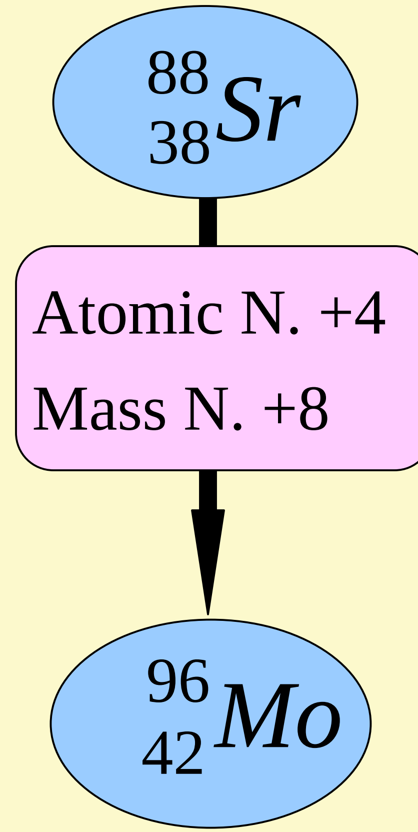

# Excess Energy and Q-value



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

$$Q \approx 50.5MeV$$

↓

$$EXH = 800J$$

↓

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

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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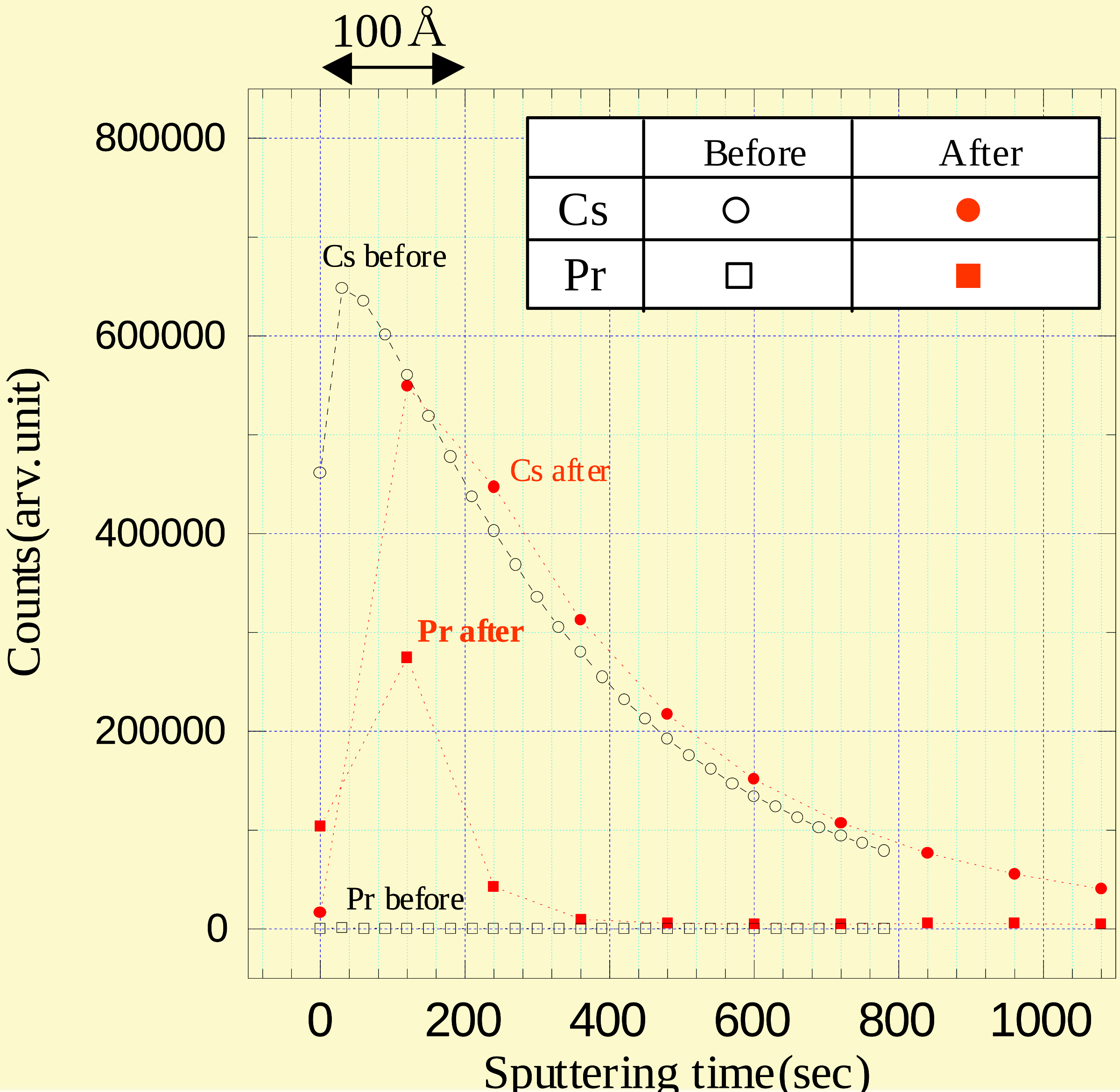

# Surface Distribution of Cs and Pr : Ion Implantation

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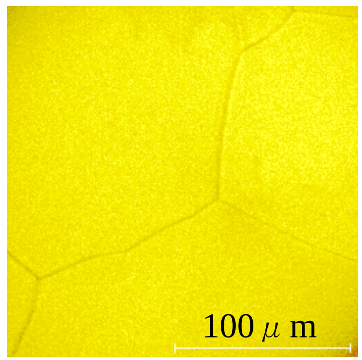
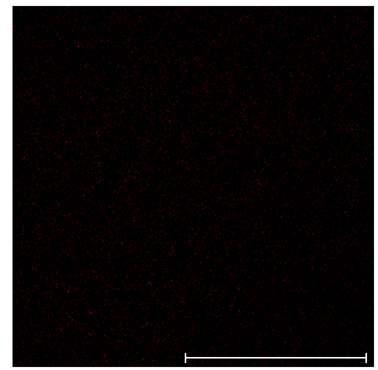
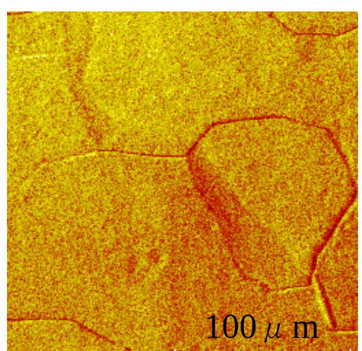
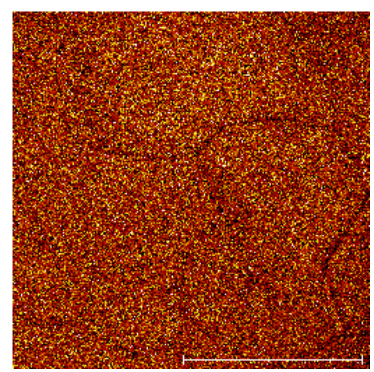
**Sample:$Cs^{+}$ Ion Implantation**
**Analysis:Tof-SIMS(ULVAC fai)**

| | Cs | Pr |
|---|---|---|
| before |  |  |
| after |  |  |

# Depth Profile of Cs and Pr
# : Electrolyte Addition

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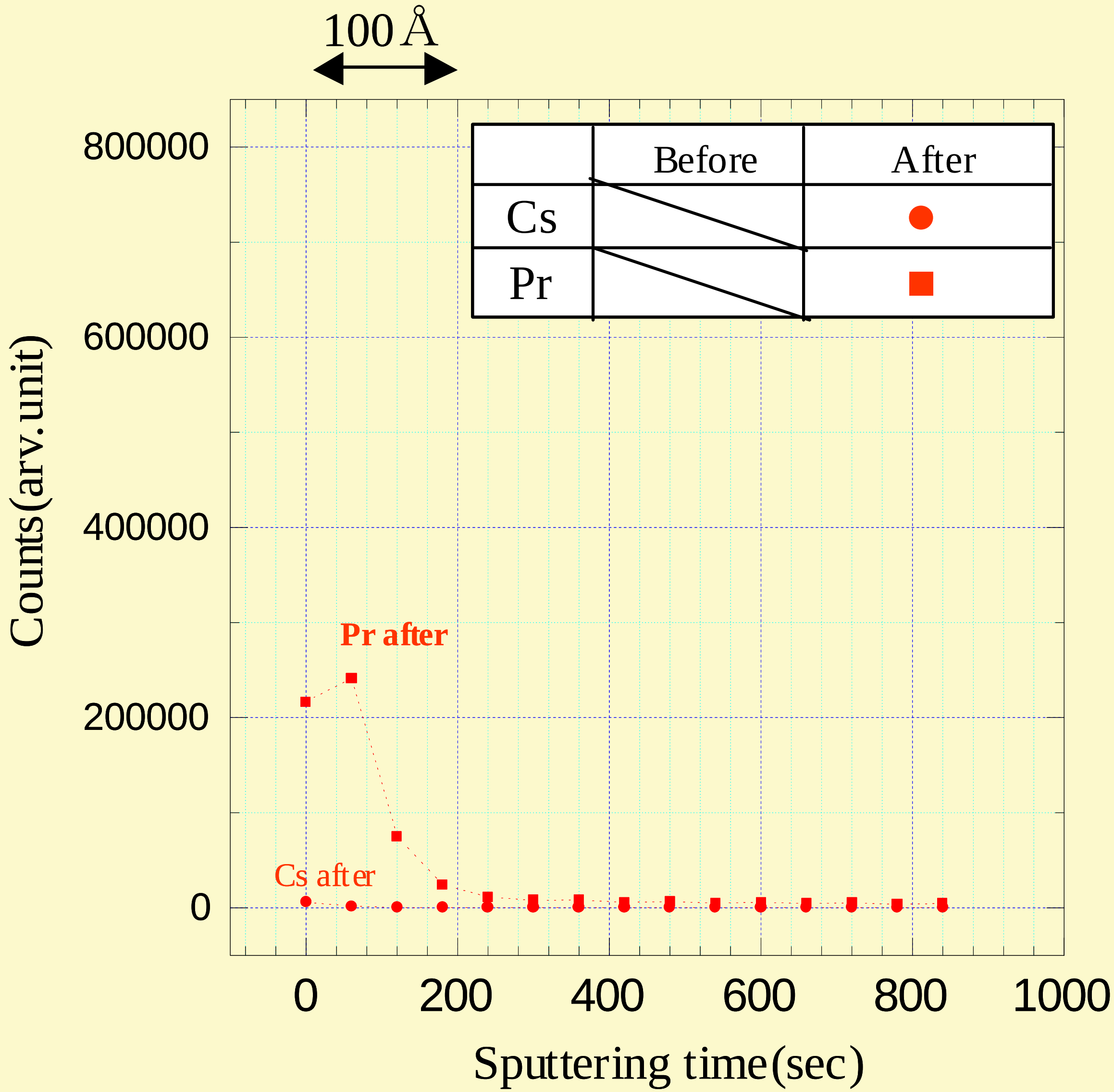


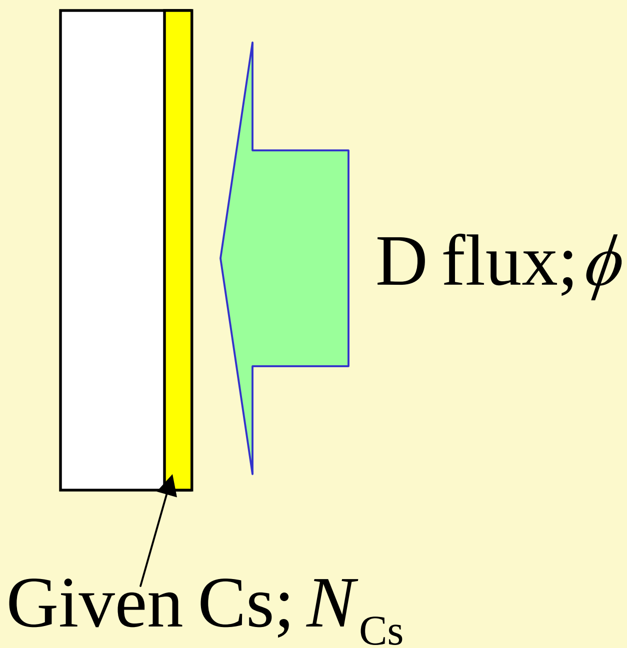
# Ultra Low Energy Beam Model



**D Permeation(D Flux)**

⇕

**Ultra low D beam**




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

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

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

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

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

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

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

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

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

# Separation of the Products and Contaminants(1)

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## 1.Rare Earth Element : Pr

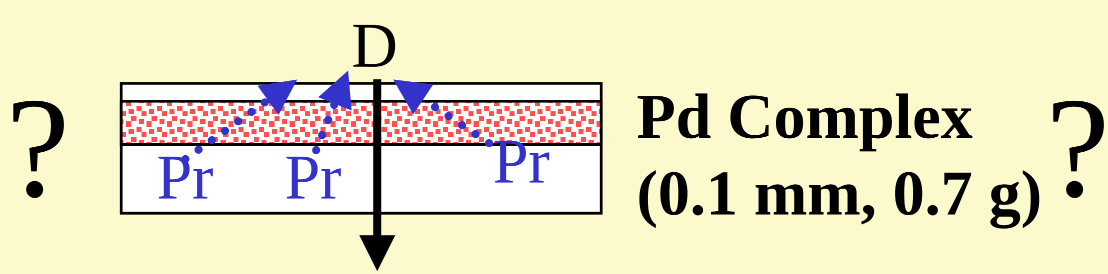
a) $D_2$ gas :The purity > 99.6% most of the impurity is $H_2$～0.4% $N_2$, $D_2O$, $O_2$, $CO_2$, CO, HC<10 ppm

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

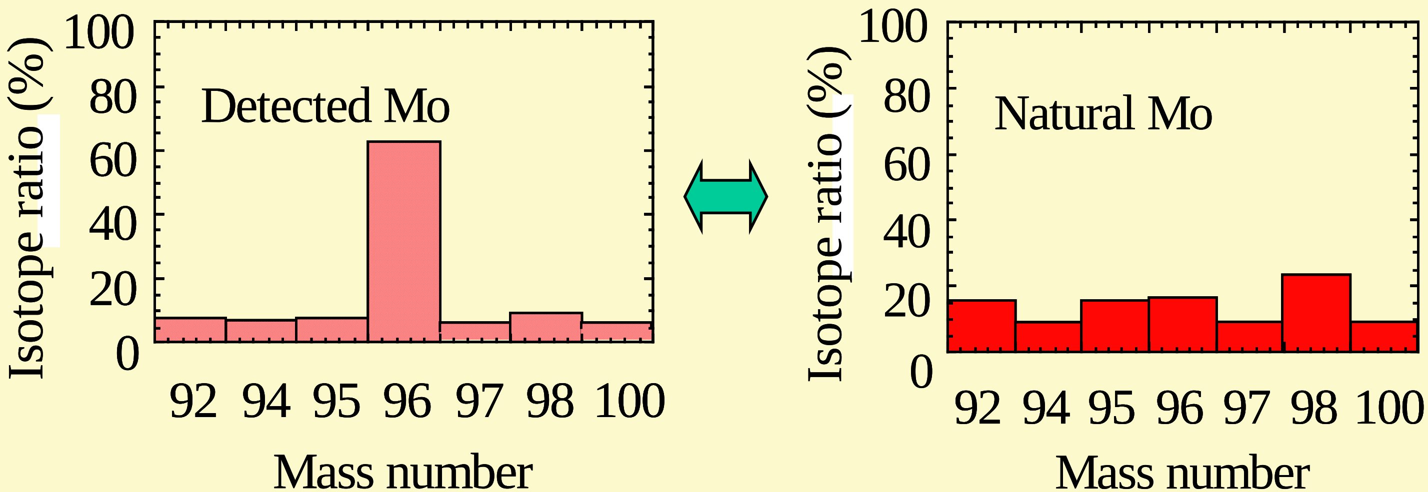
→Less than the detected Pr(10ng～100ng)!



# Separation of the Products and Contaminants(2)



## 2. Anomalous Isotopic Composition: Mo




**Detection of Pr and Mo cannot be explain by contamination.**

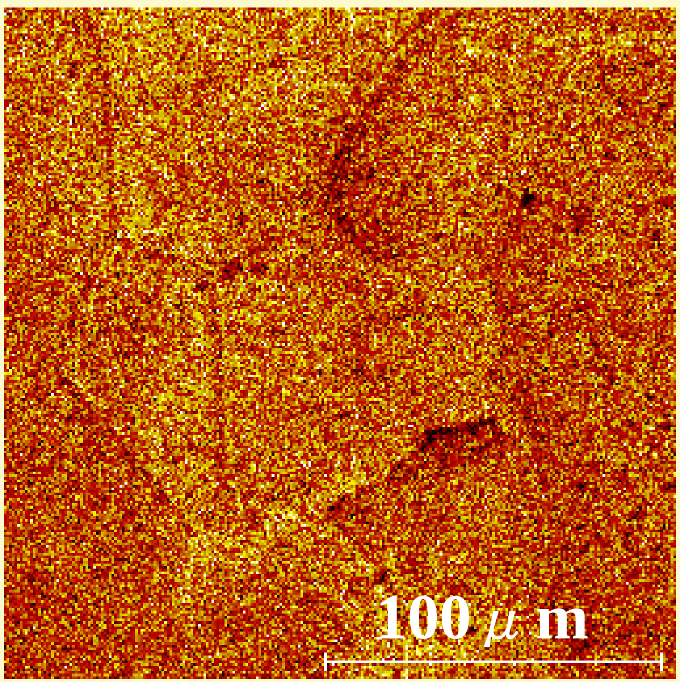
**Pr and Mo are the products of nuclear transmutation reactions.**

# Surface Distribution of Cs and Pr : Electrolyte Addition

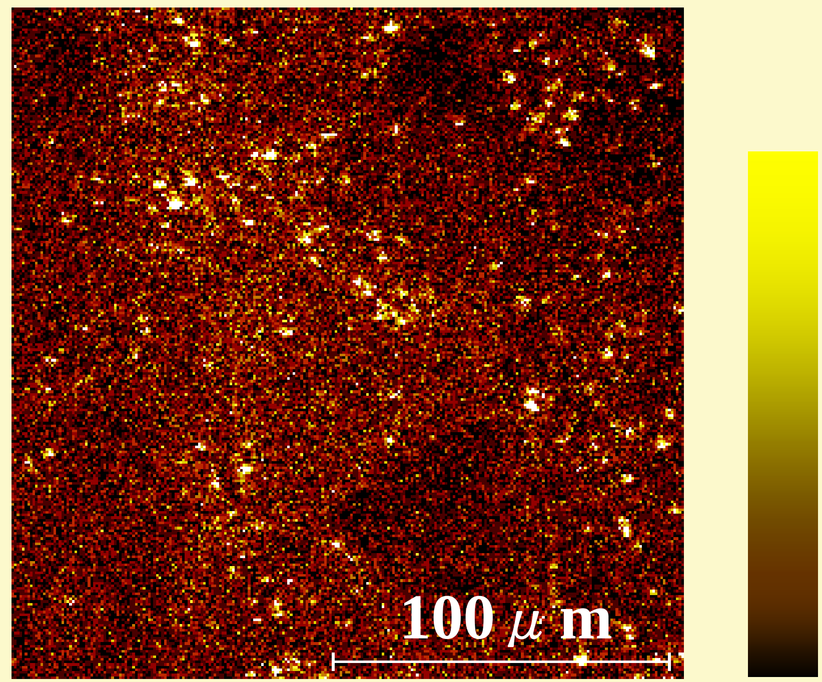


**Sample:Cs is added using electrochemical method**
**Analysis:Tof-SIMS(ULVAC fai)**

## 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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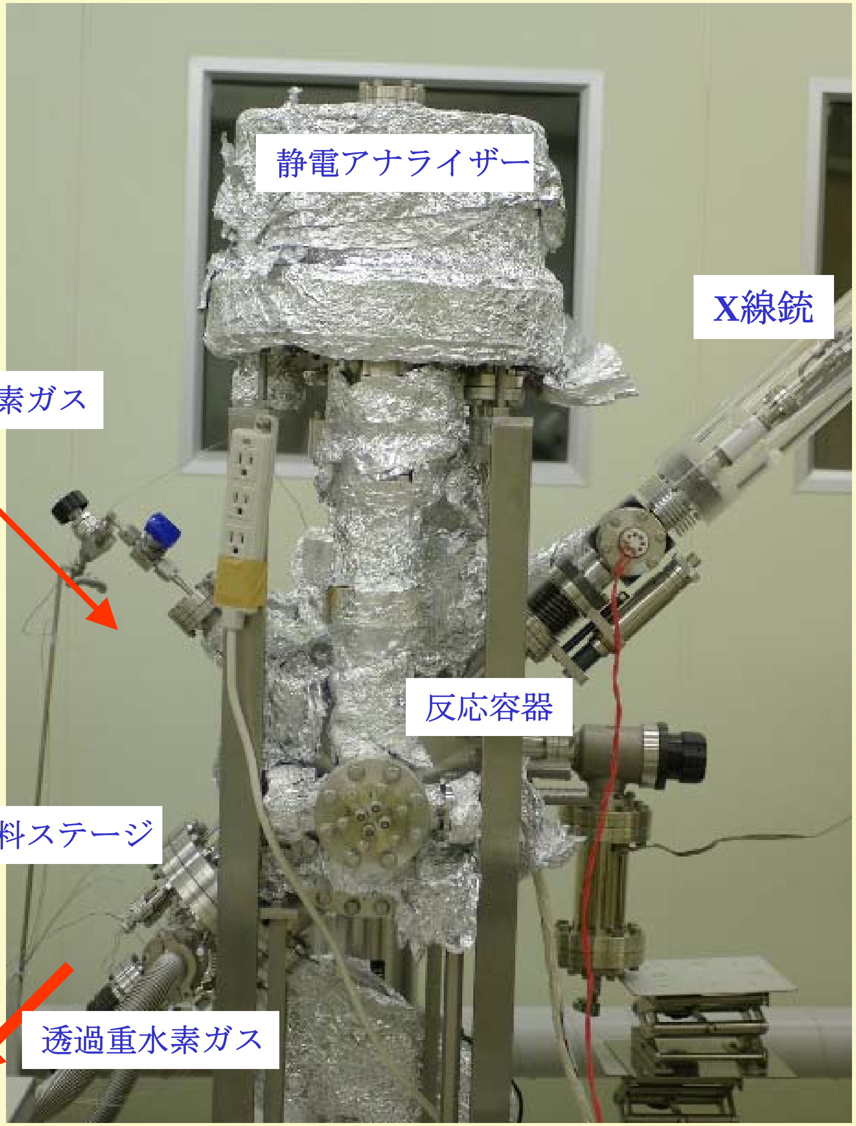
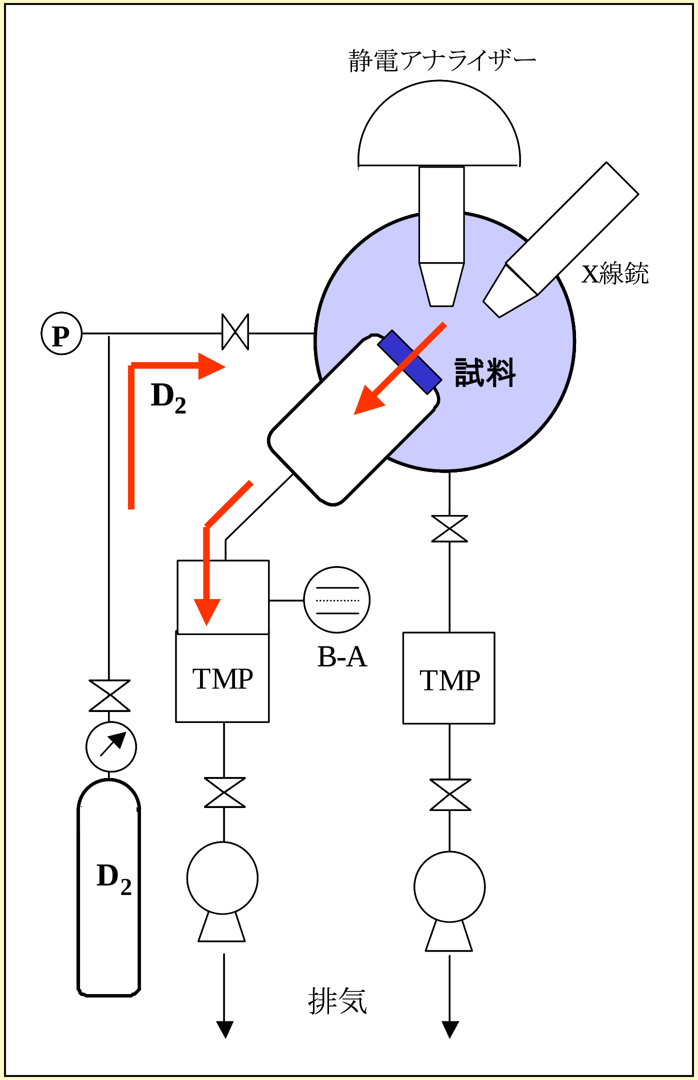
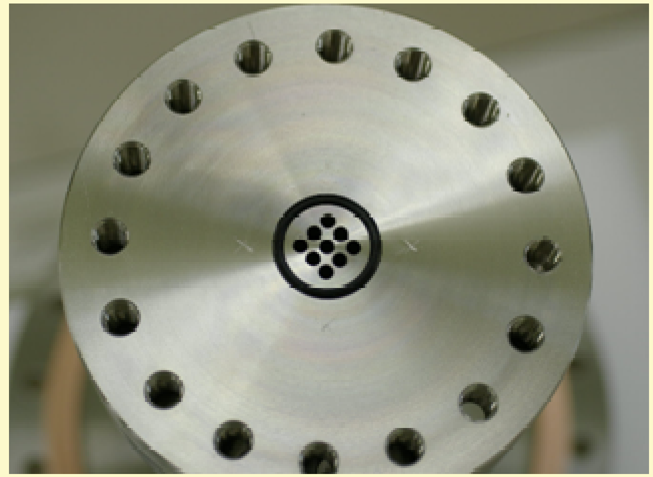
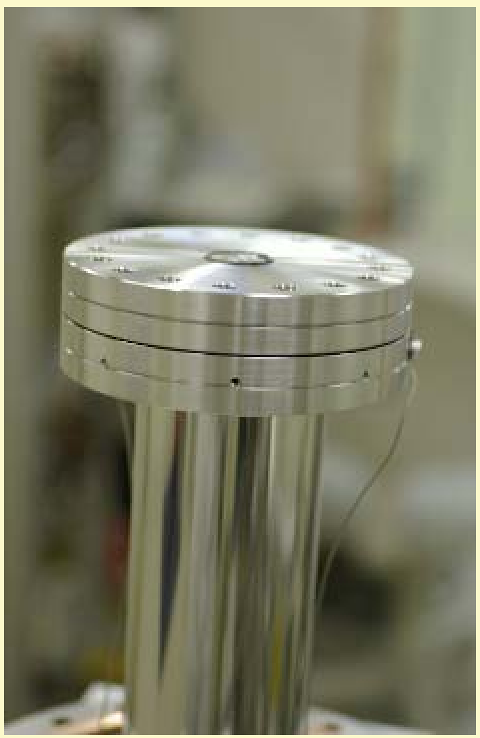


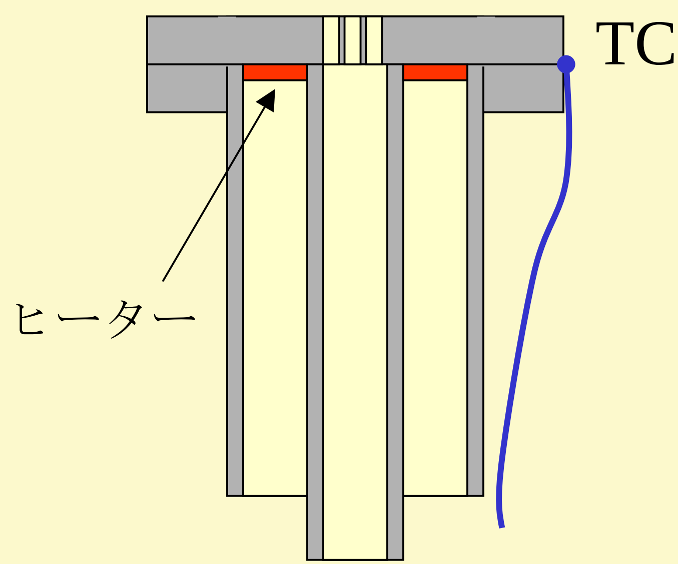



# 試料ステージ概観









# 重水素ガスの透過経路

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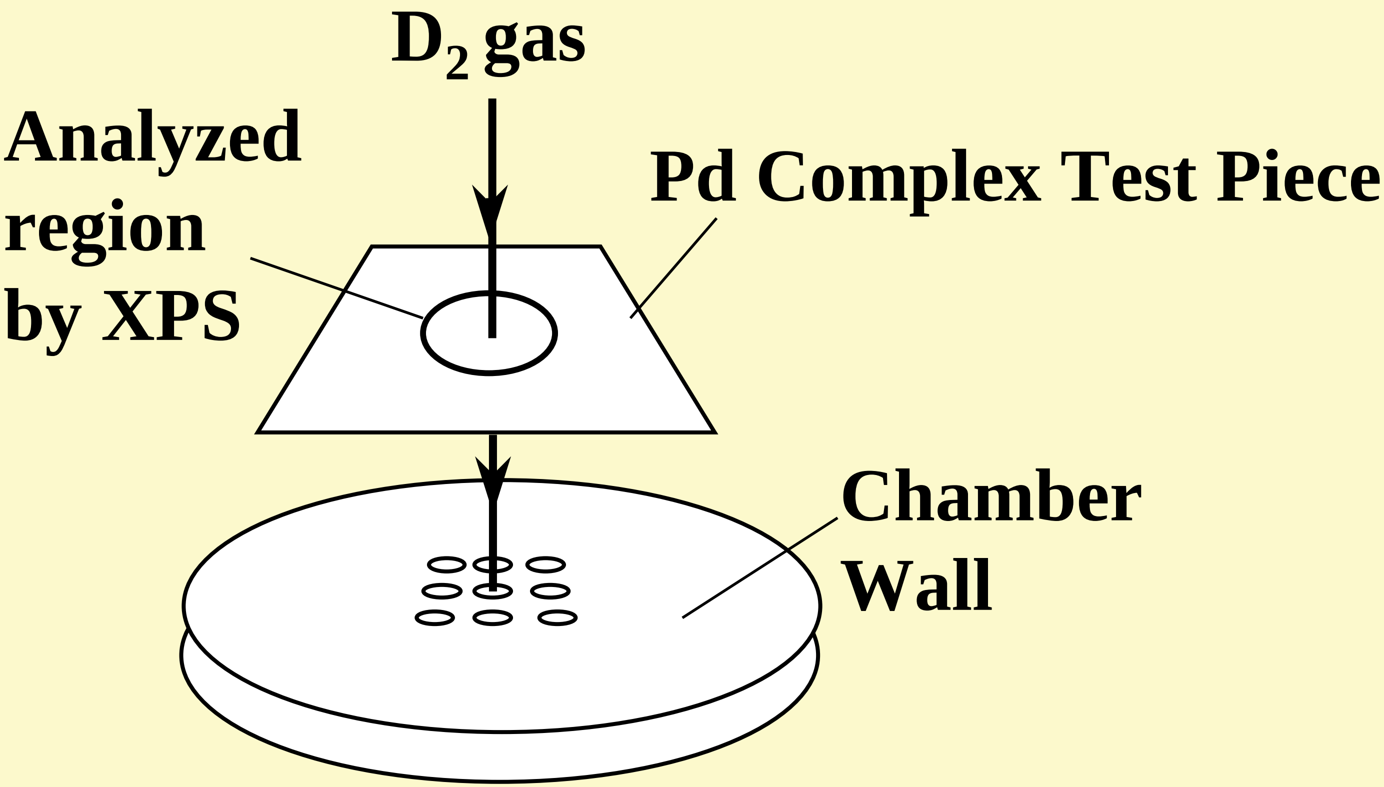

# Identification of Pr by TOF-SIMS(2)

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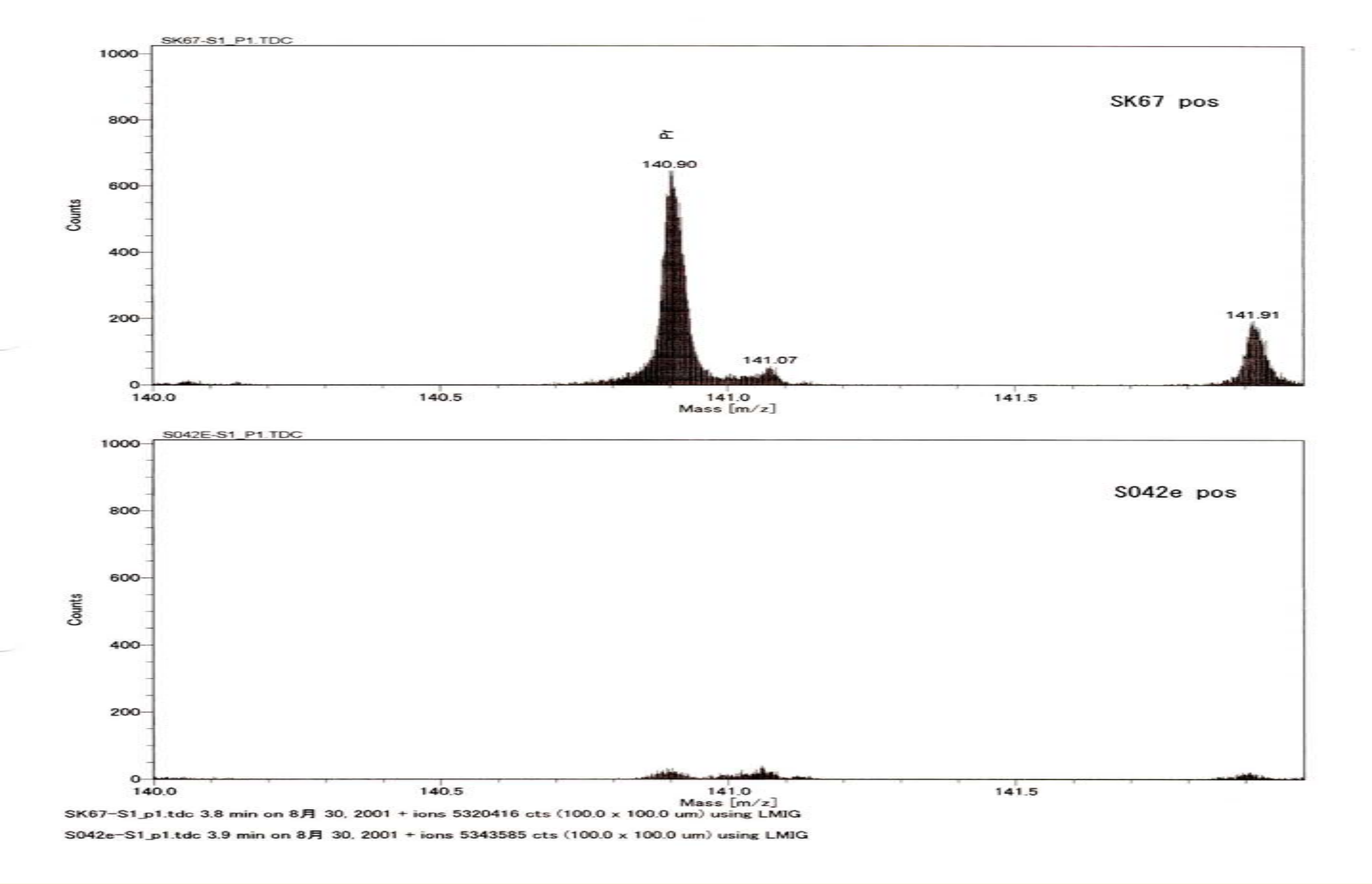


# Quantitative Analysis of Pr by ICP-MS



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

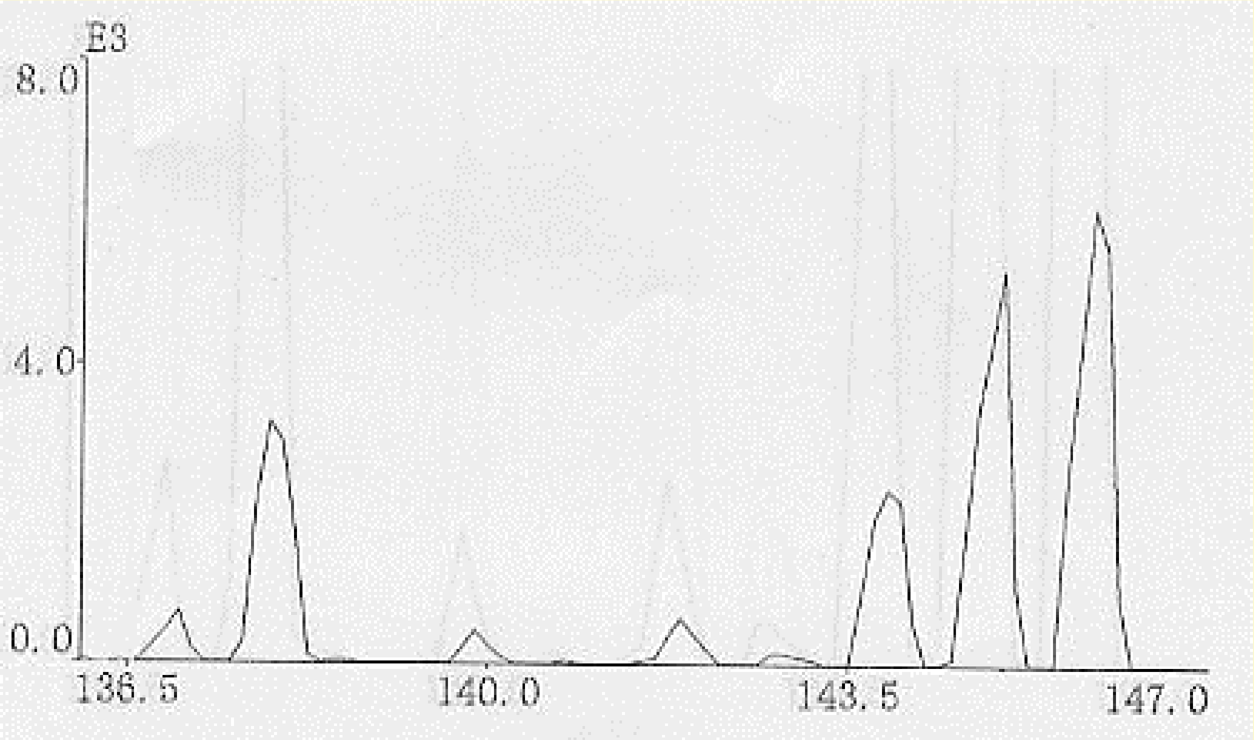


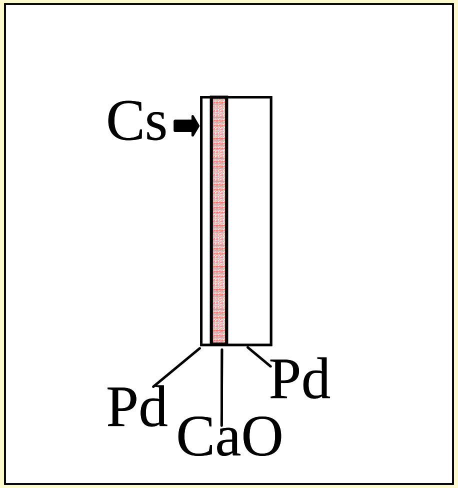
| Pd | Pd(NO) | $PdO_2$ |
|---|---|---|
| 102(1%) | 132 | 134 |
| 104 (11%) | 134 | 136 |
| 105 (22%) | 135 | 137 |
| 106 (27%) | 136 | 138 |
| 108 (26%) | 138 | 140 |
| 110 (12%) | 140 | 142 |

**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)

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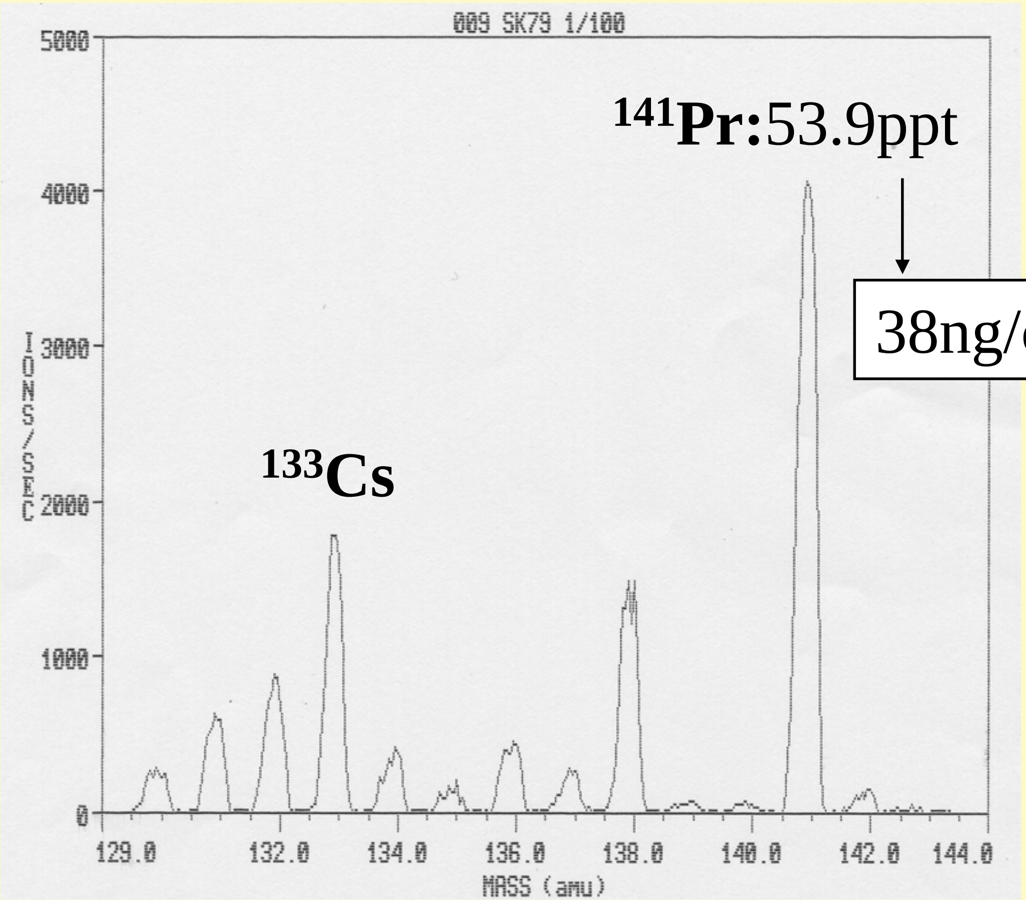



$^{141}$**Pr:**53.9ppt

$38ng/cm^2$

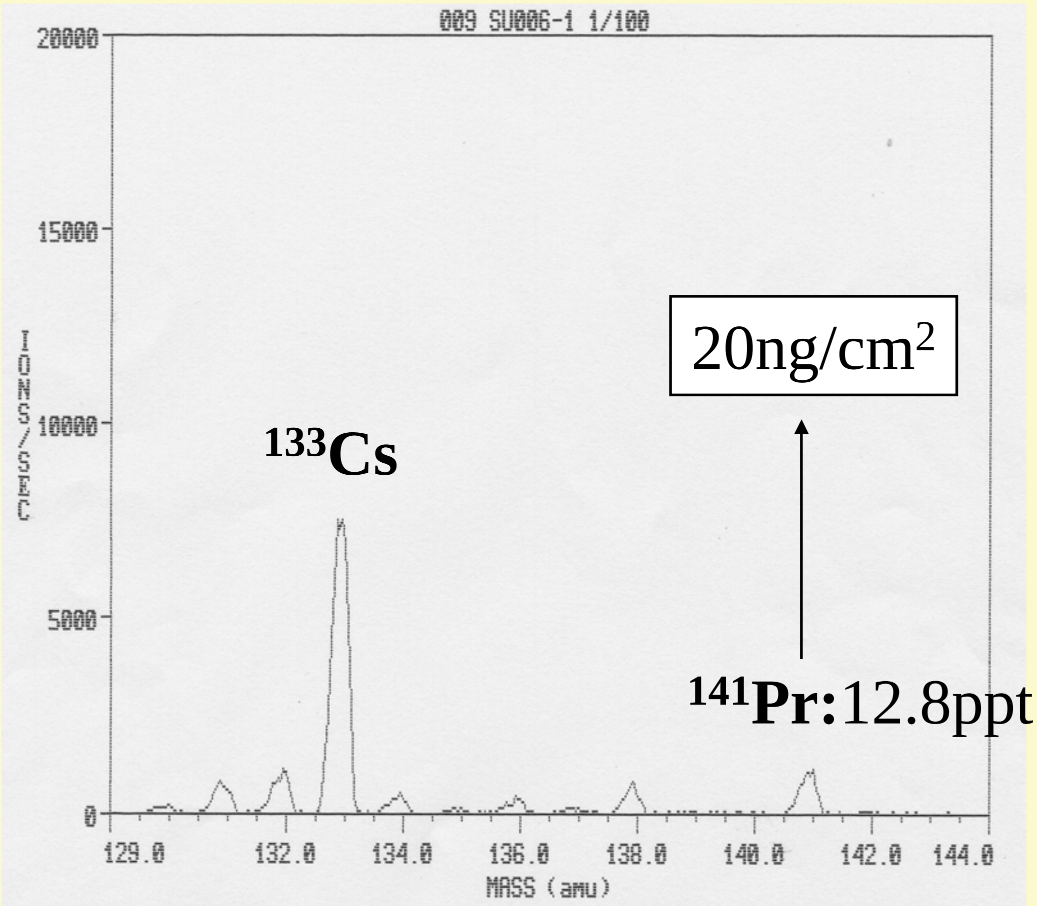
$^{133}$**Cs**

Ion counts(counts/sec)

Mass Number

# F.G.Data(2)






Ion counts(counts/sec)

Mass Number

# Analysis of the Standard Solution

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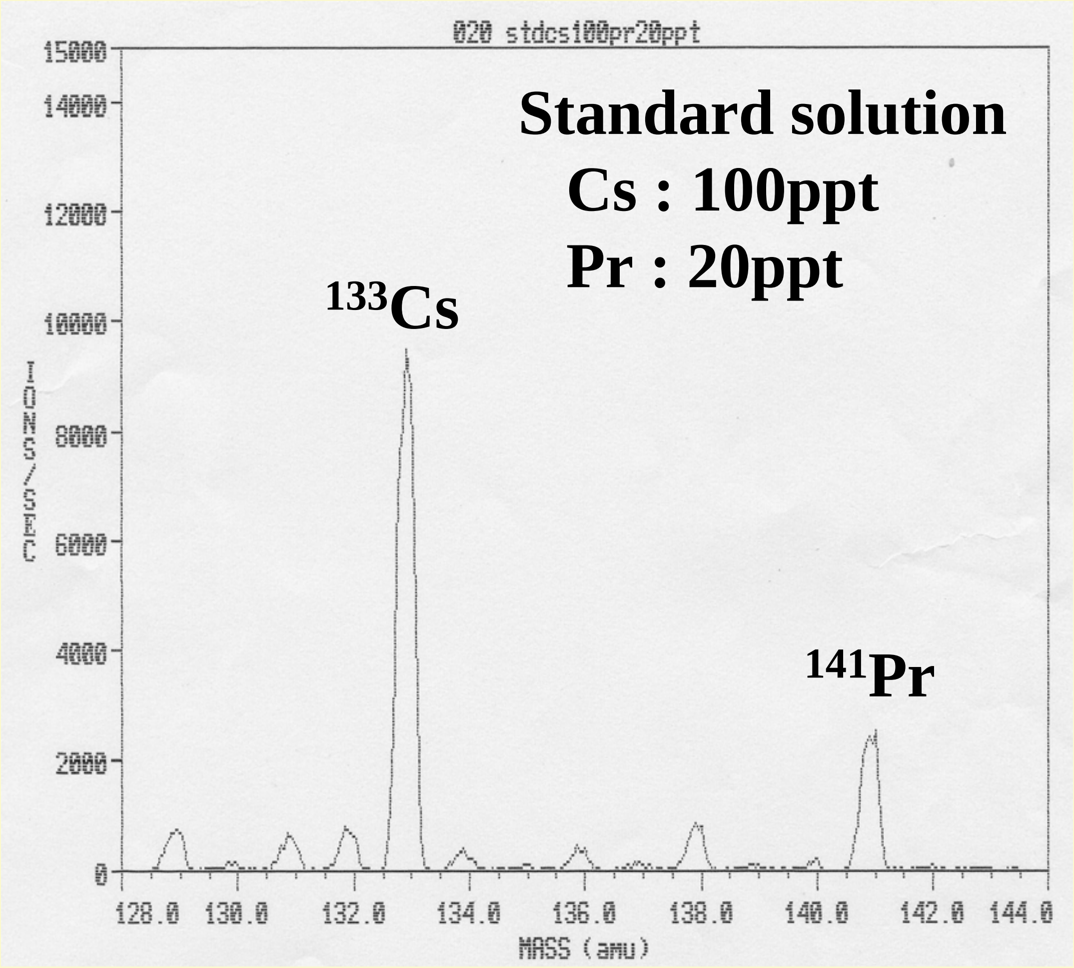
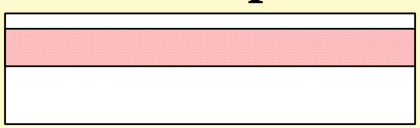
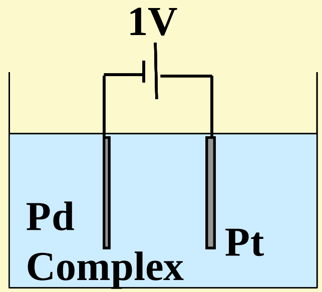
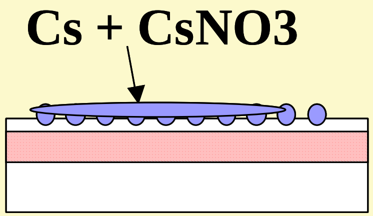
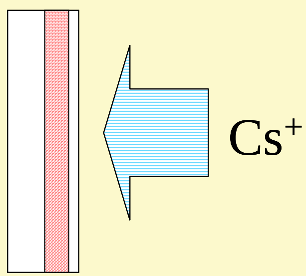
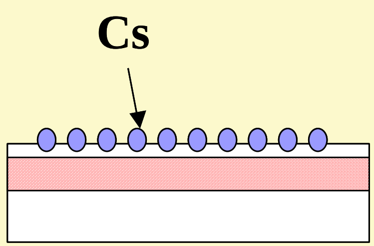
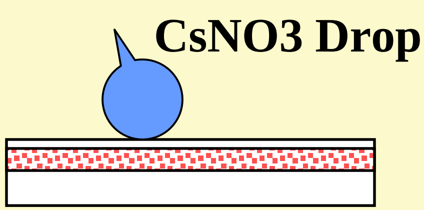
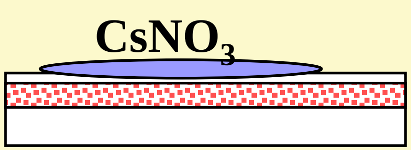
Ion counts(counts/sec)




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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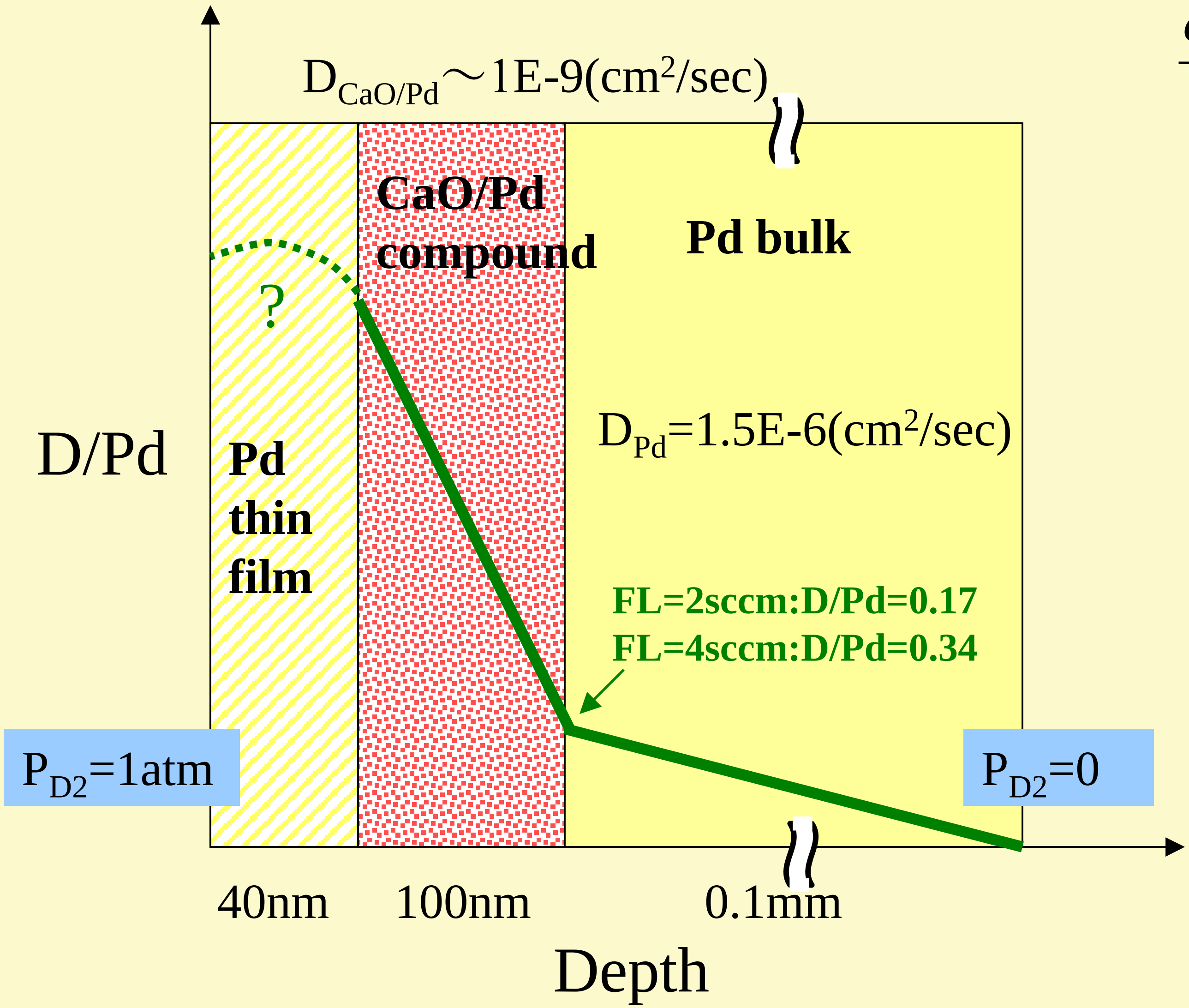


$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} \qquad \frac{\partial}{\partial t} = 0$$

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

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

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

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

⇓

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

# Cross Sectional View

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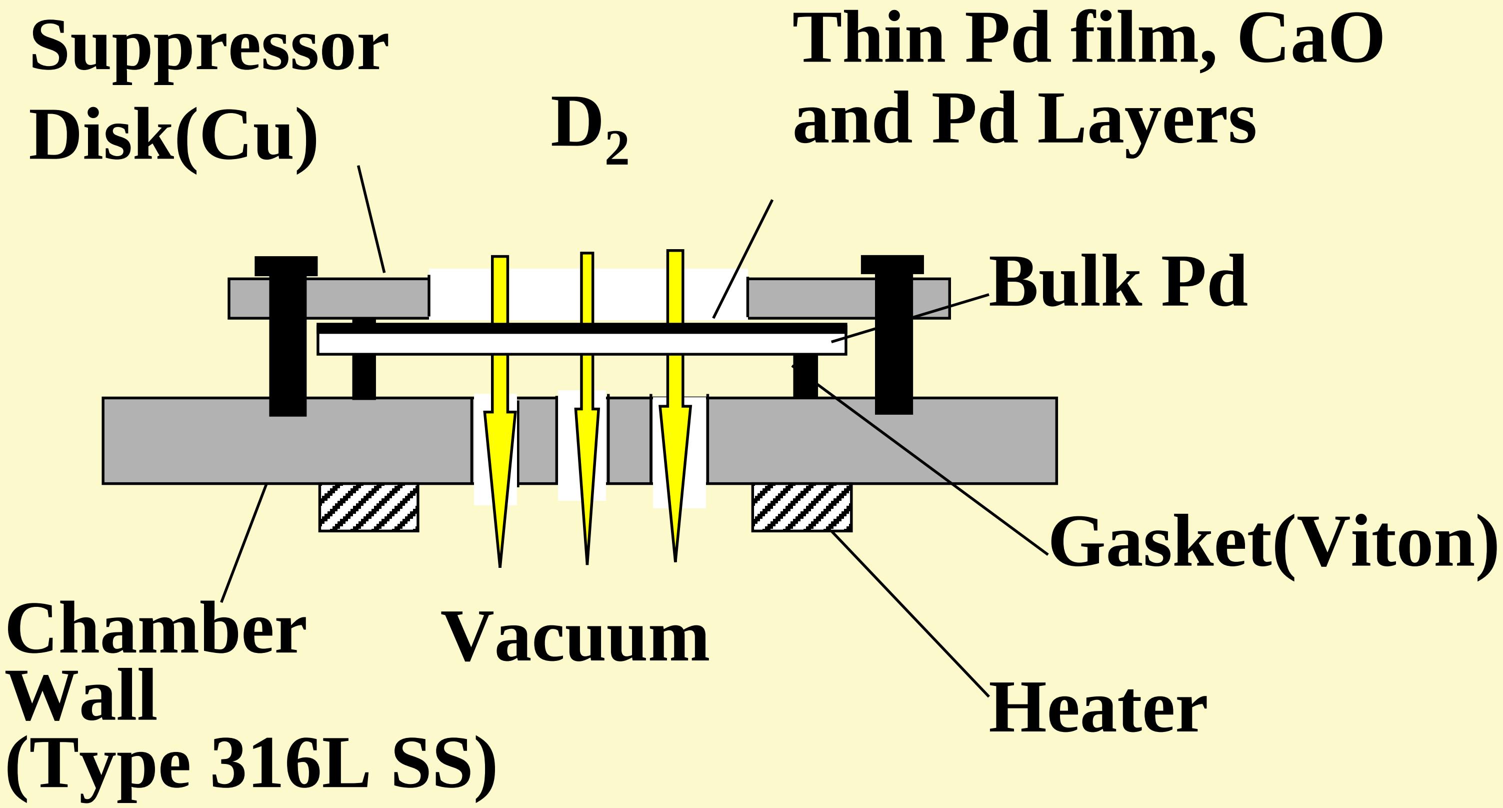
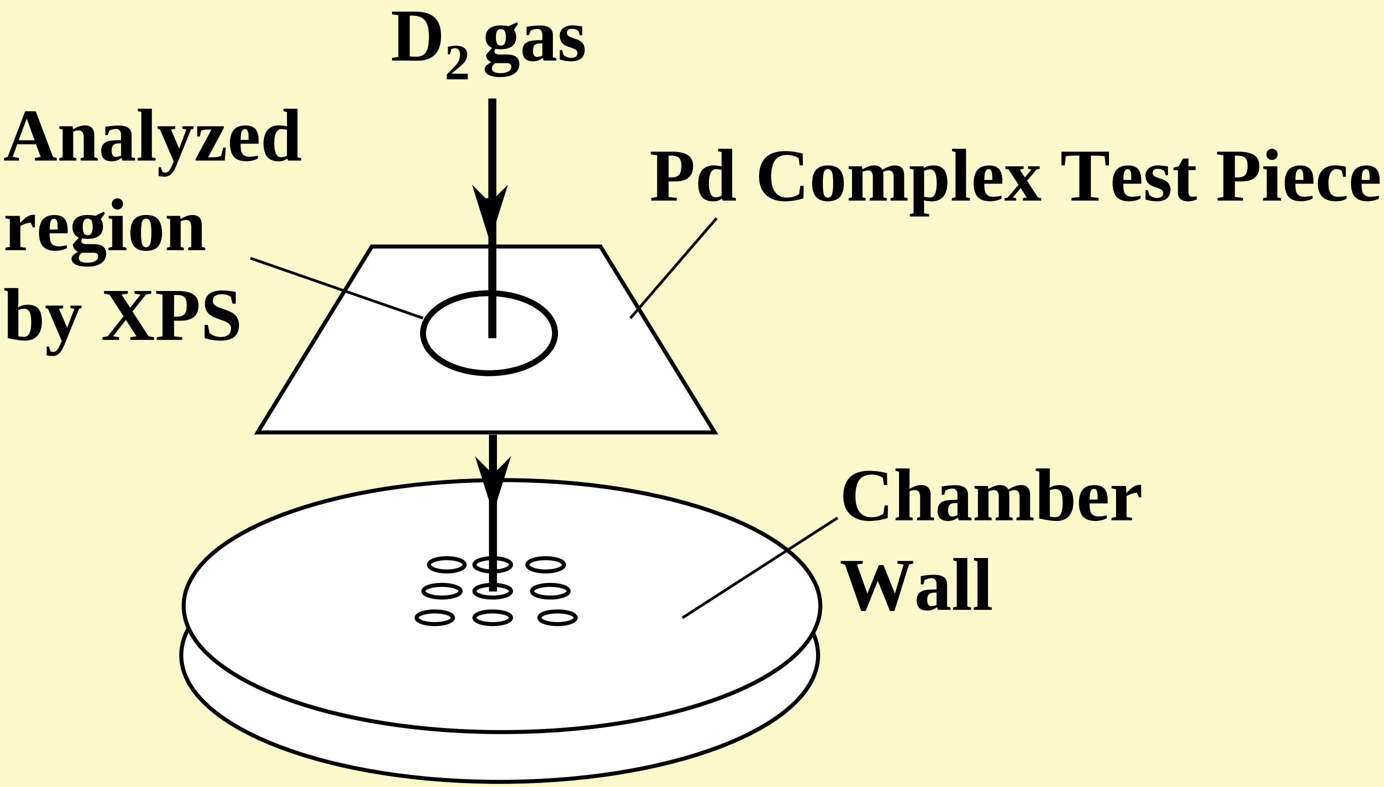


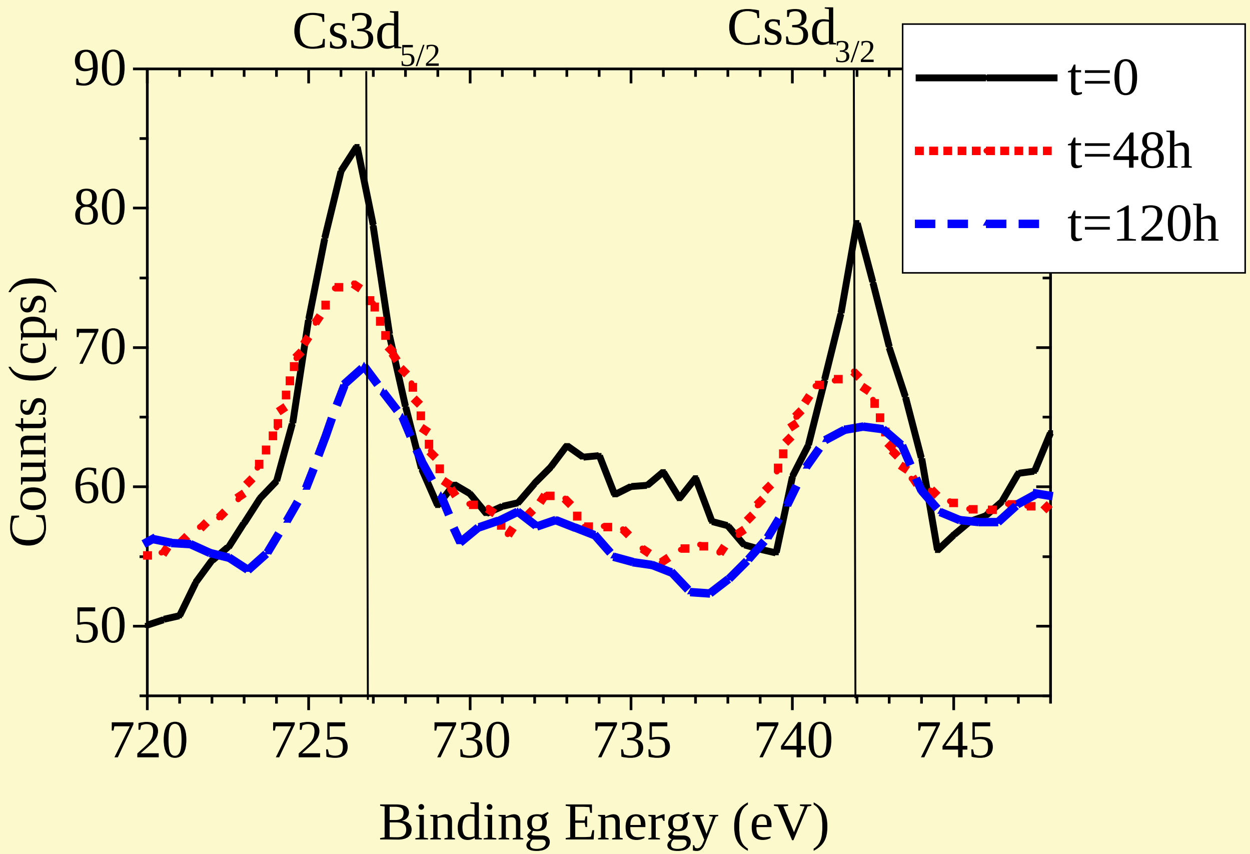
# Passage of $D_2$ Gas





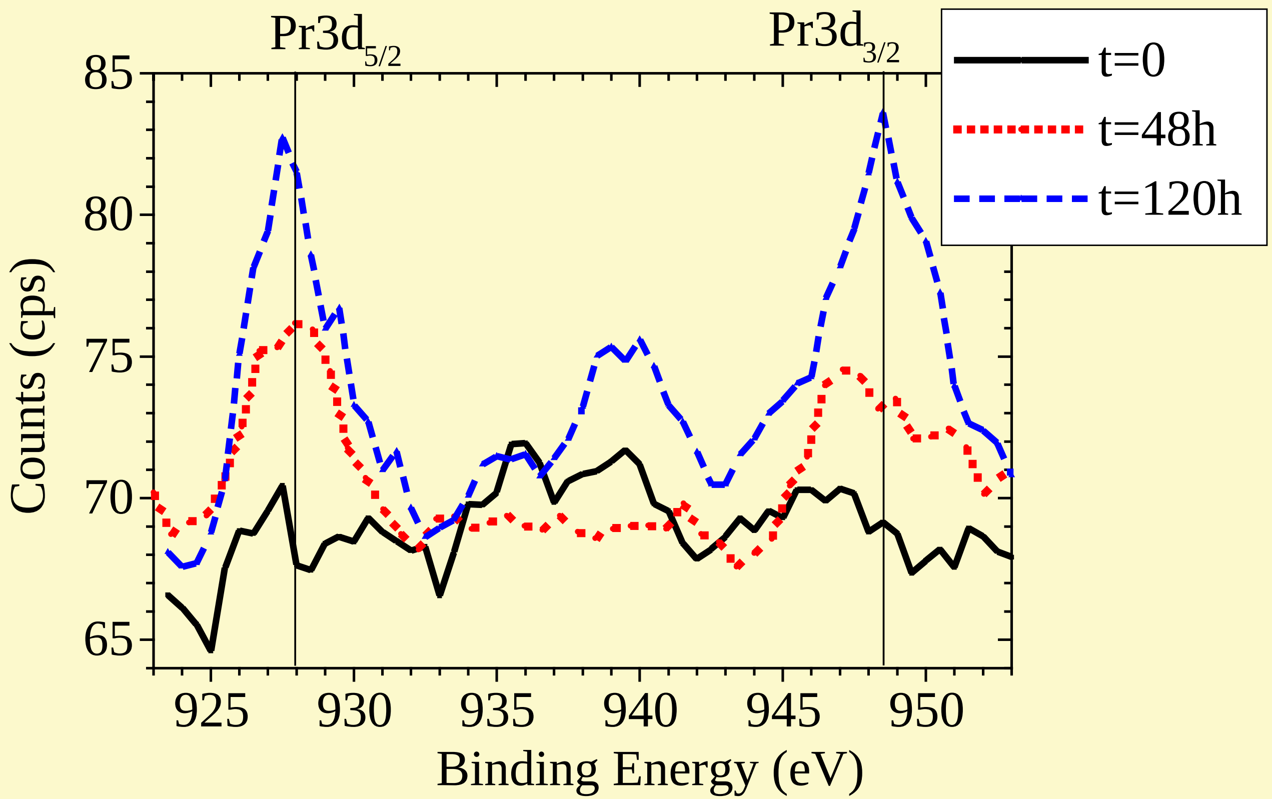
# Change of XPS Spectrum of Cs





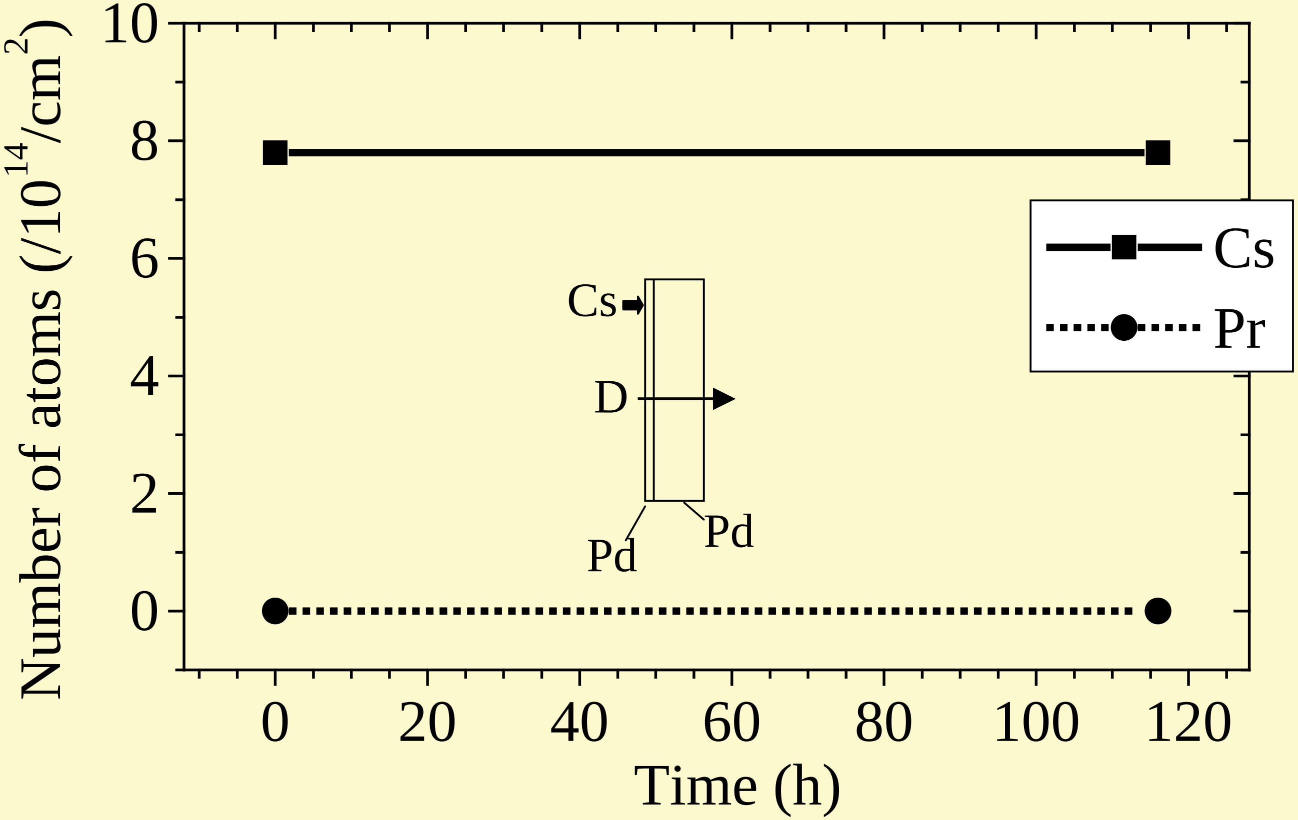
# Change of XPS Spectrum of Pr





# 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

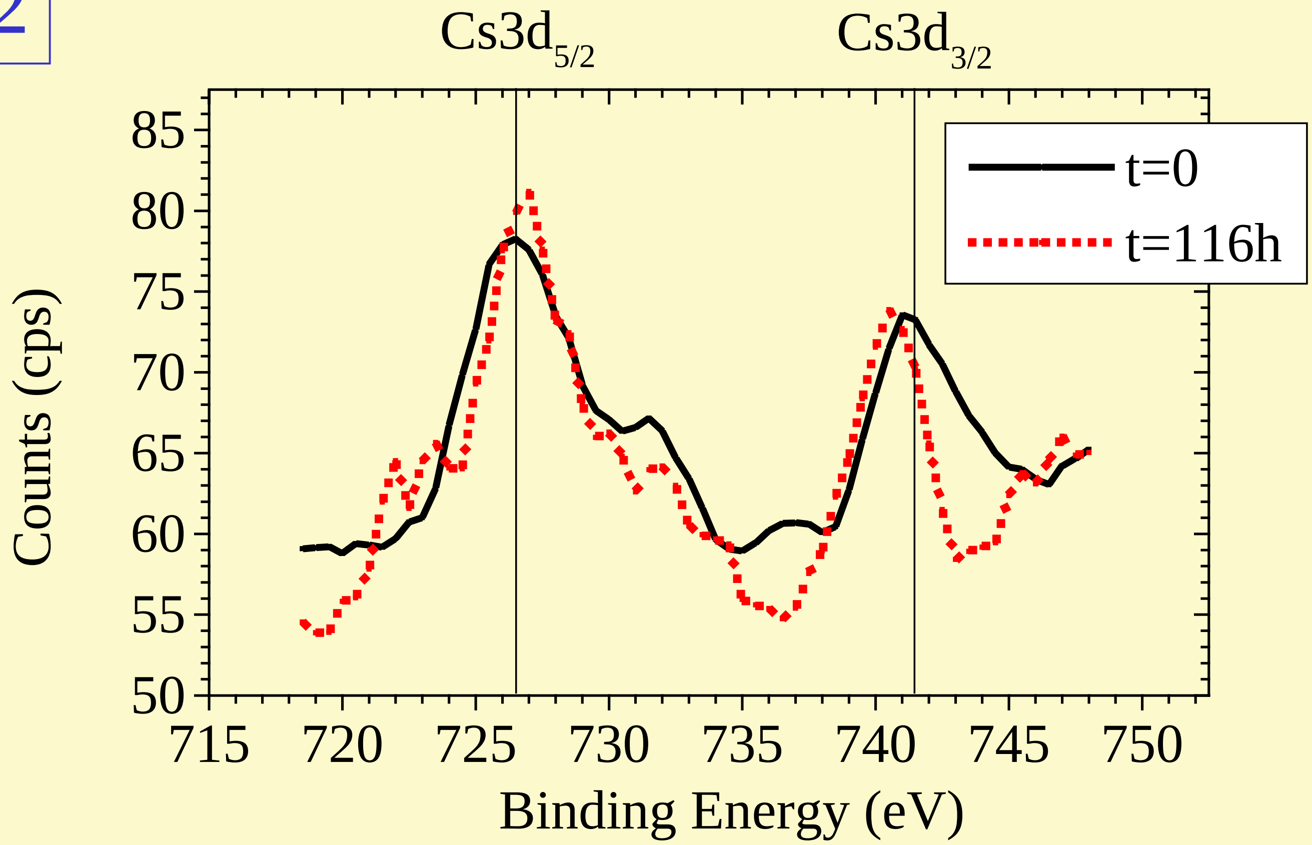
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



# XPS Spectrum of Cs (No CaO)

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

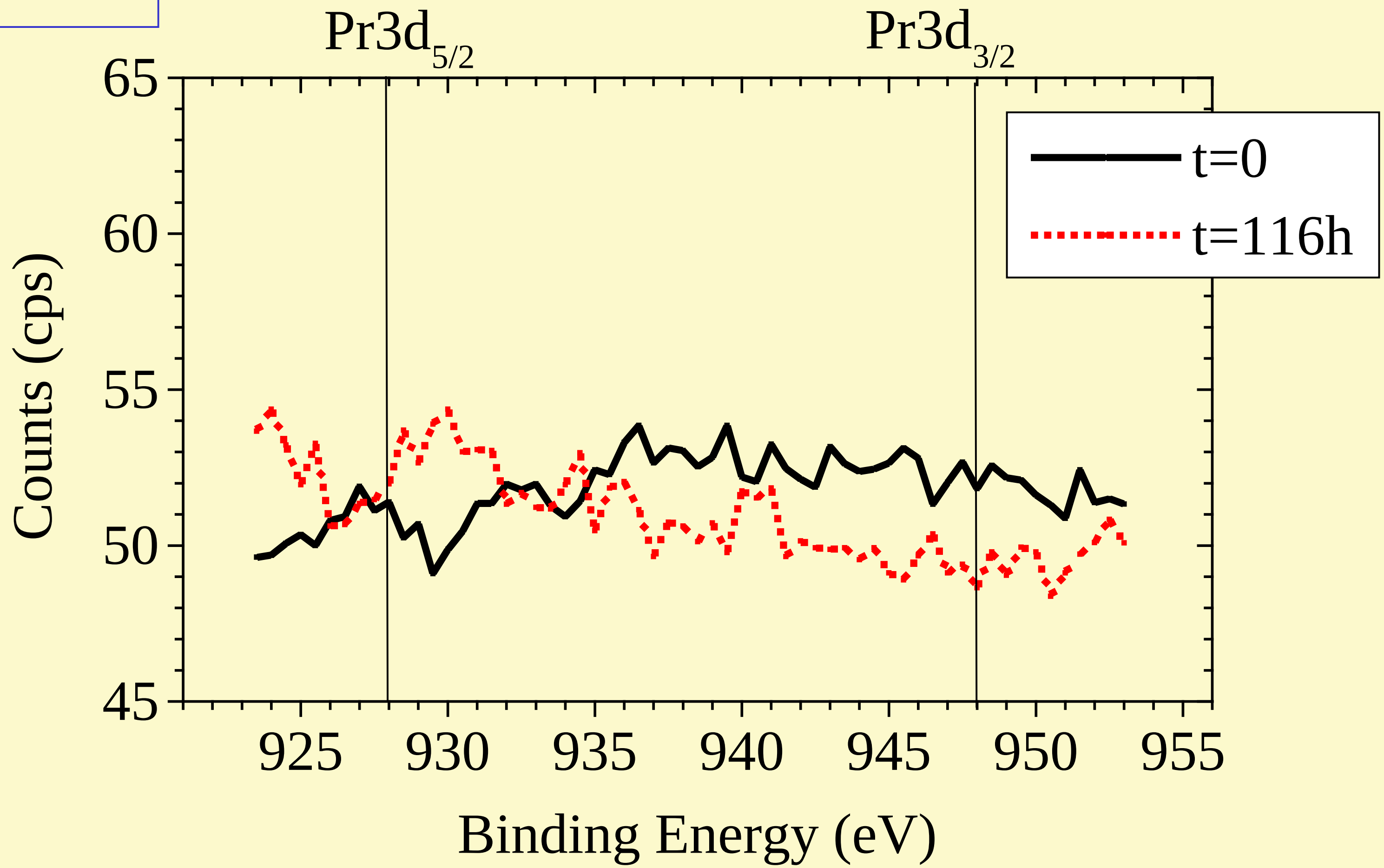
Pd/Pd/D2



# XPS Spectrum of Pr (No CaO)

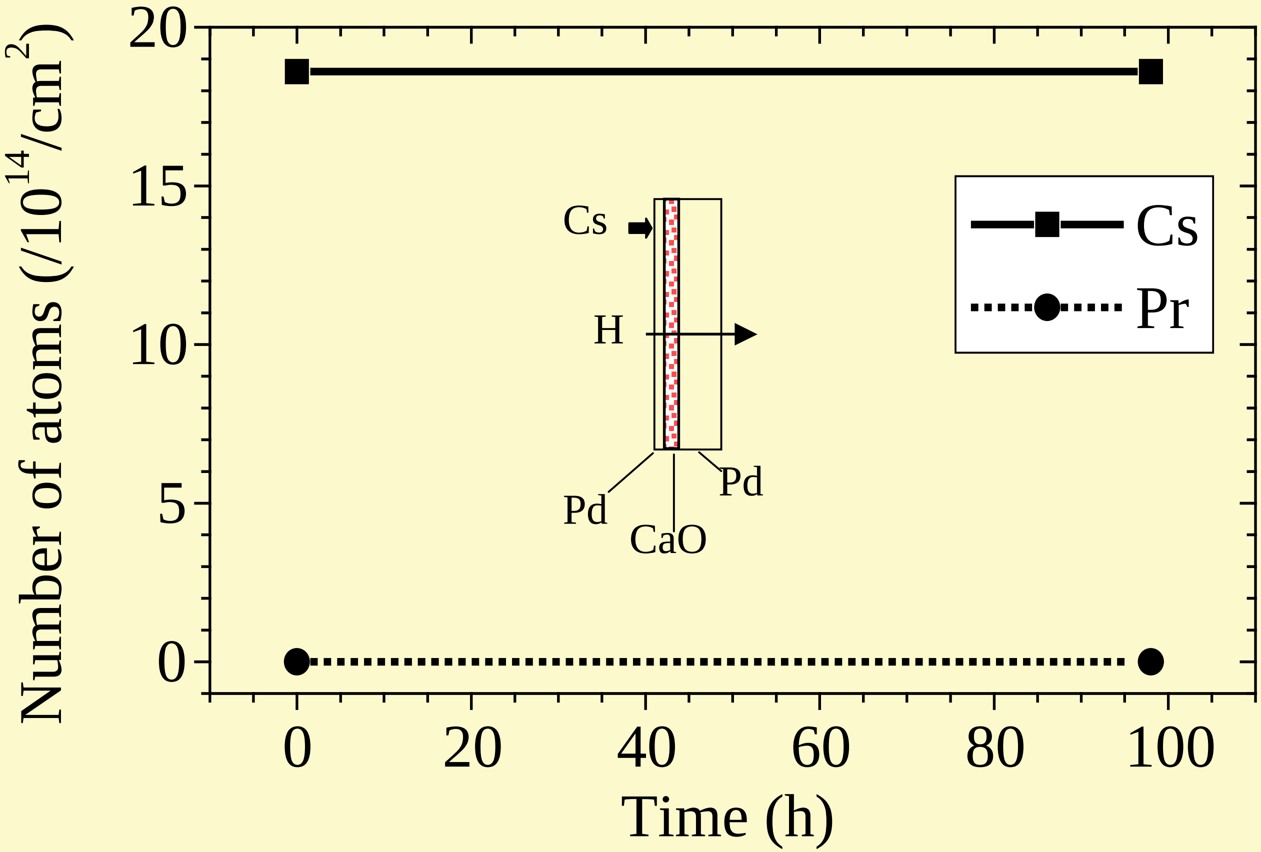


Pd/Pd/D2



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

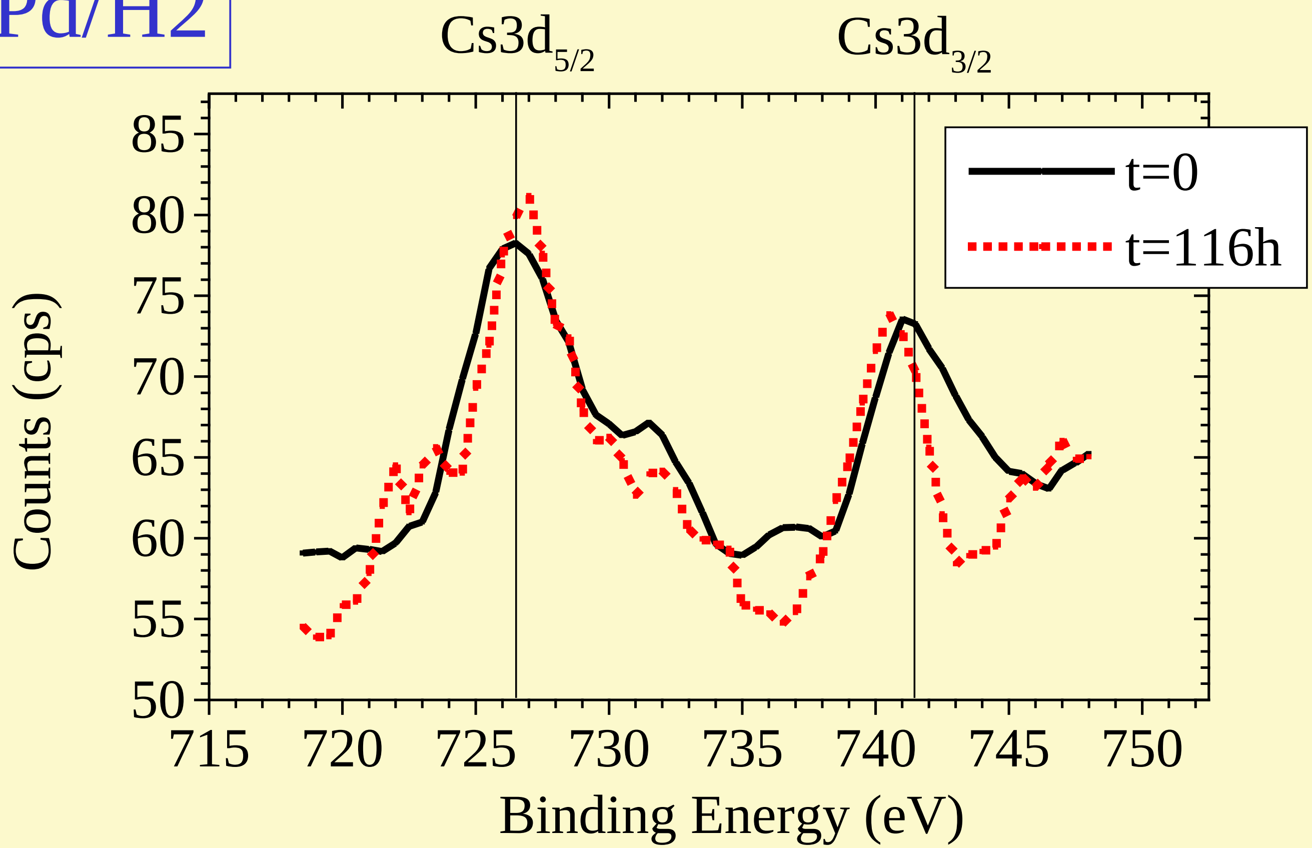
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



# XPS Spectrum of Cs($H_2$ Permeation)

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

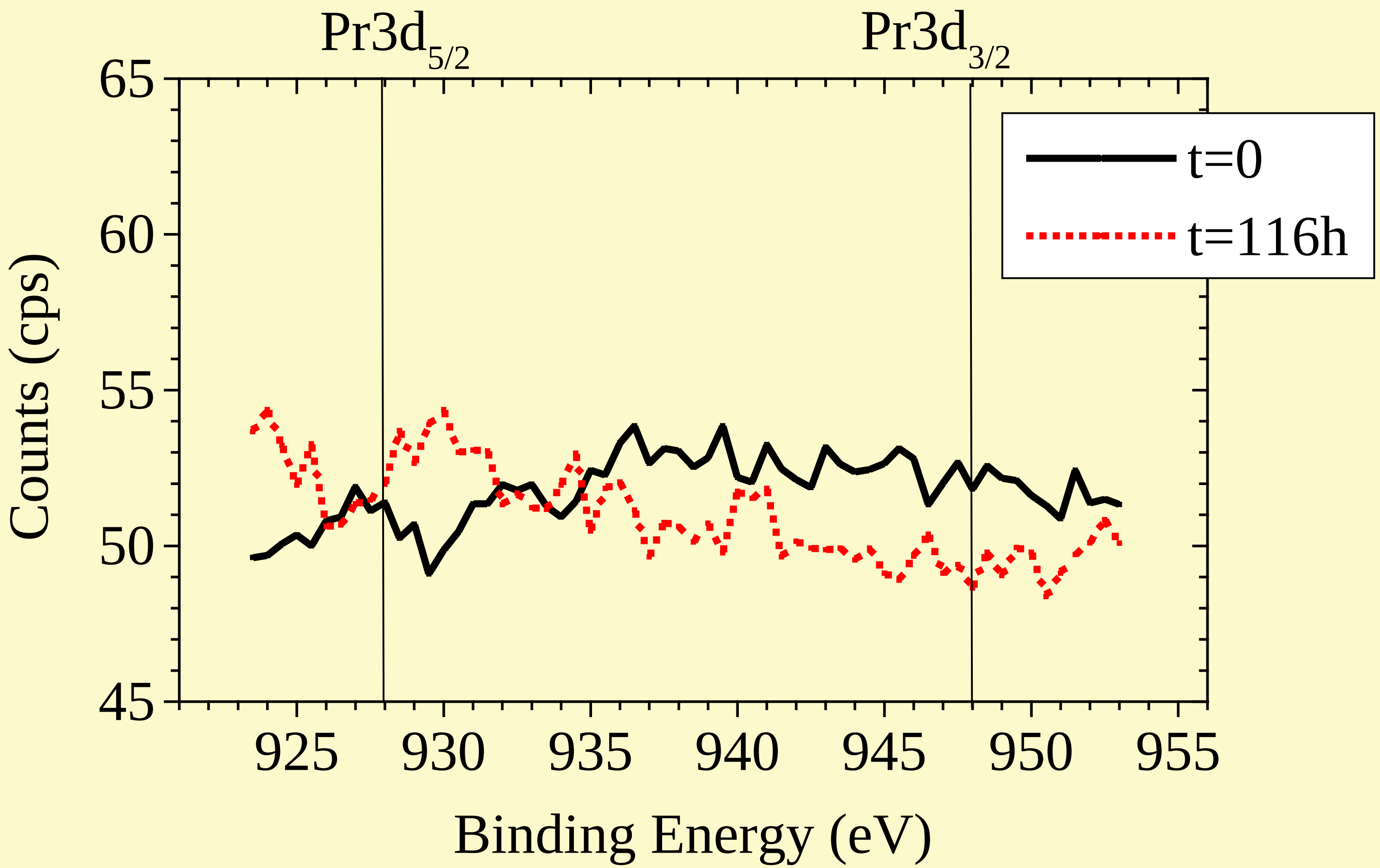
Pd/CaO/Pd/H2



# XPS Spectrum of Pr ($H_2$ Permeation)

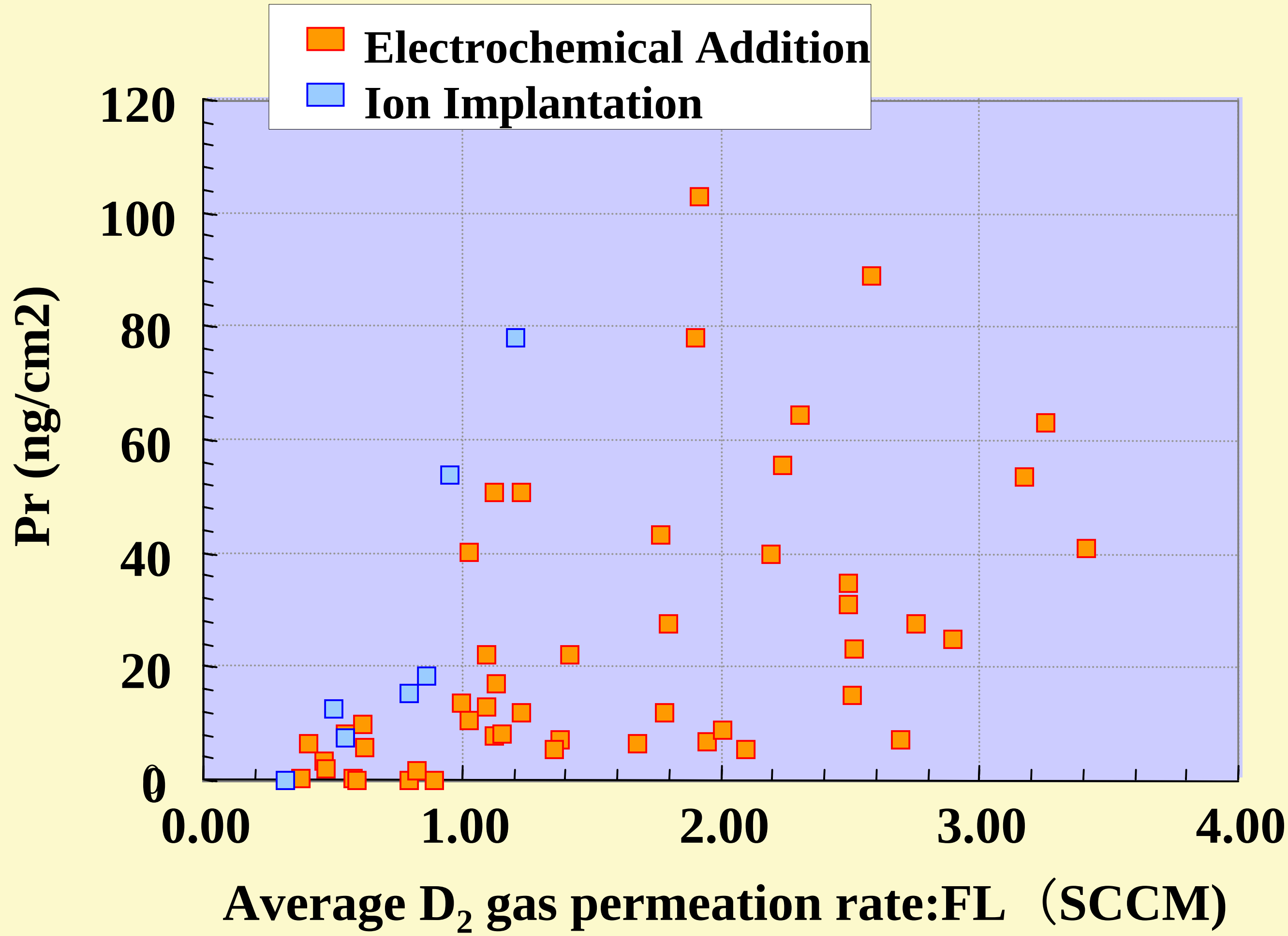
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

Pd/CaO/Pd/H2



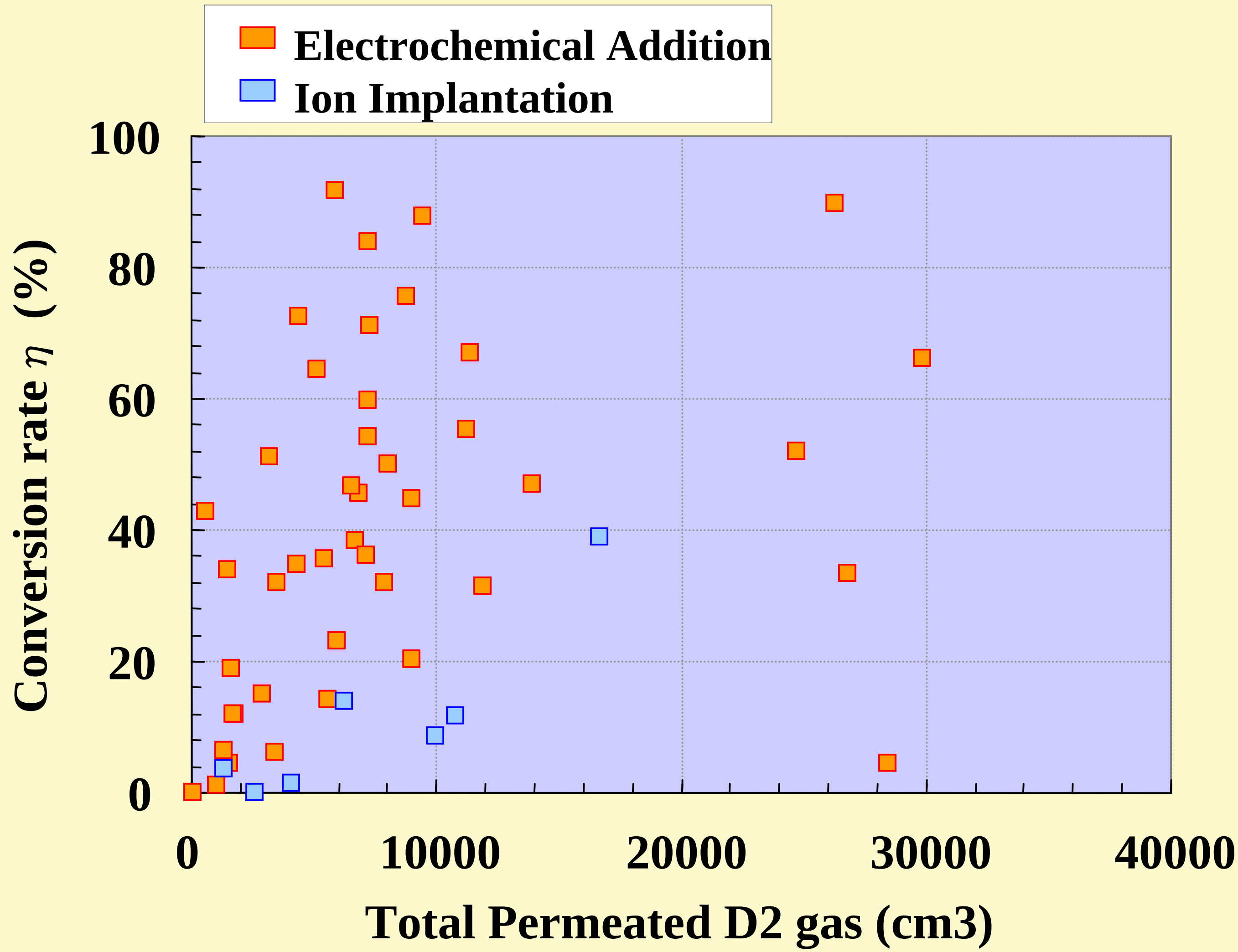
# Average Permeation Rate and Products

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



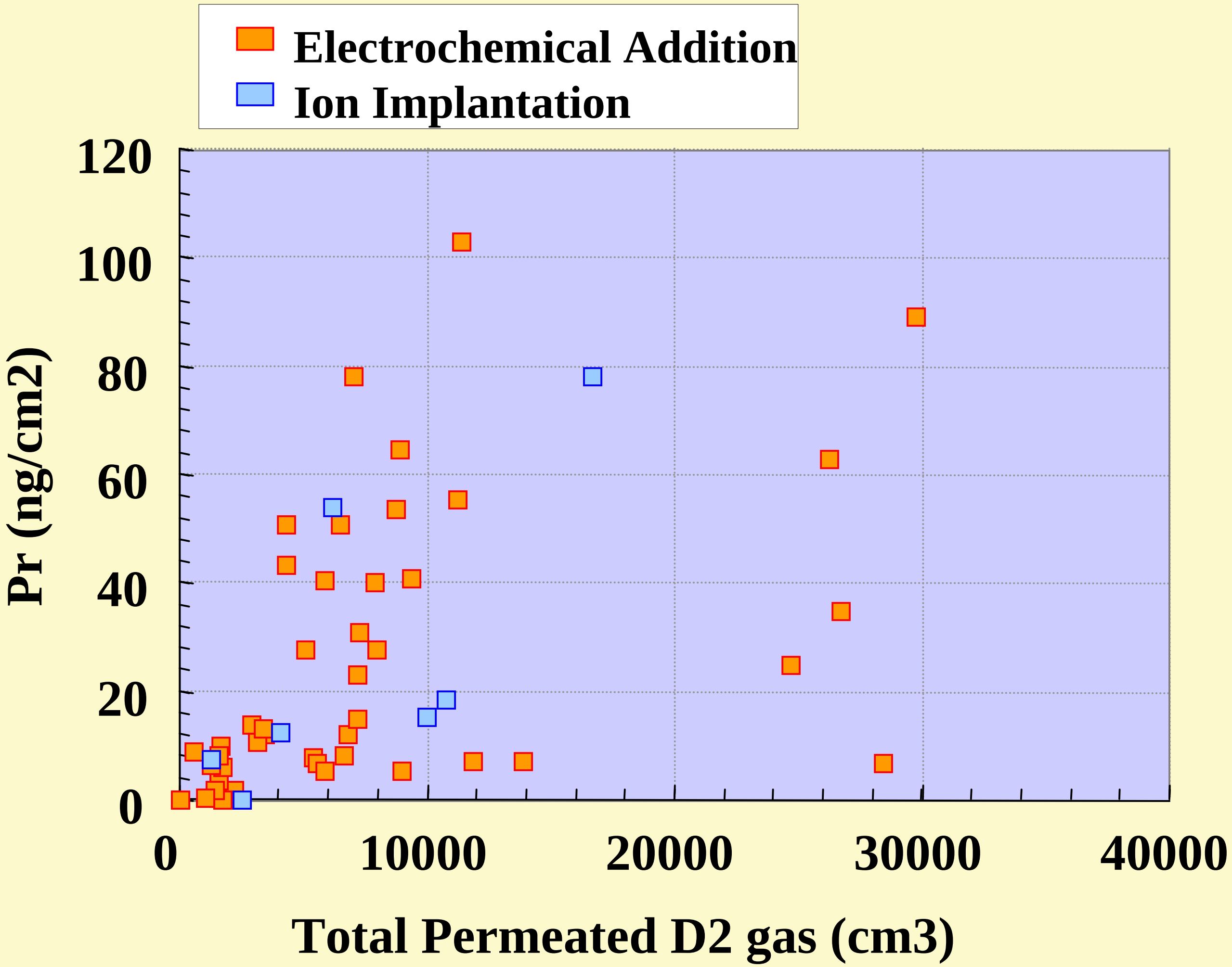
# Total Permeated D2 Gas and Conversion Rate

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



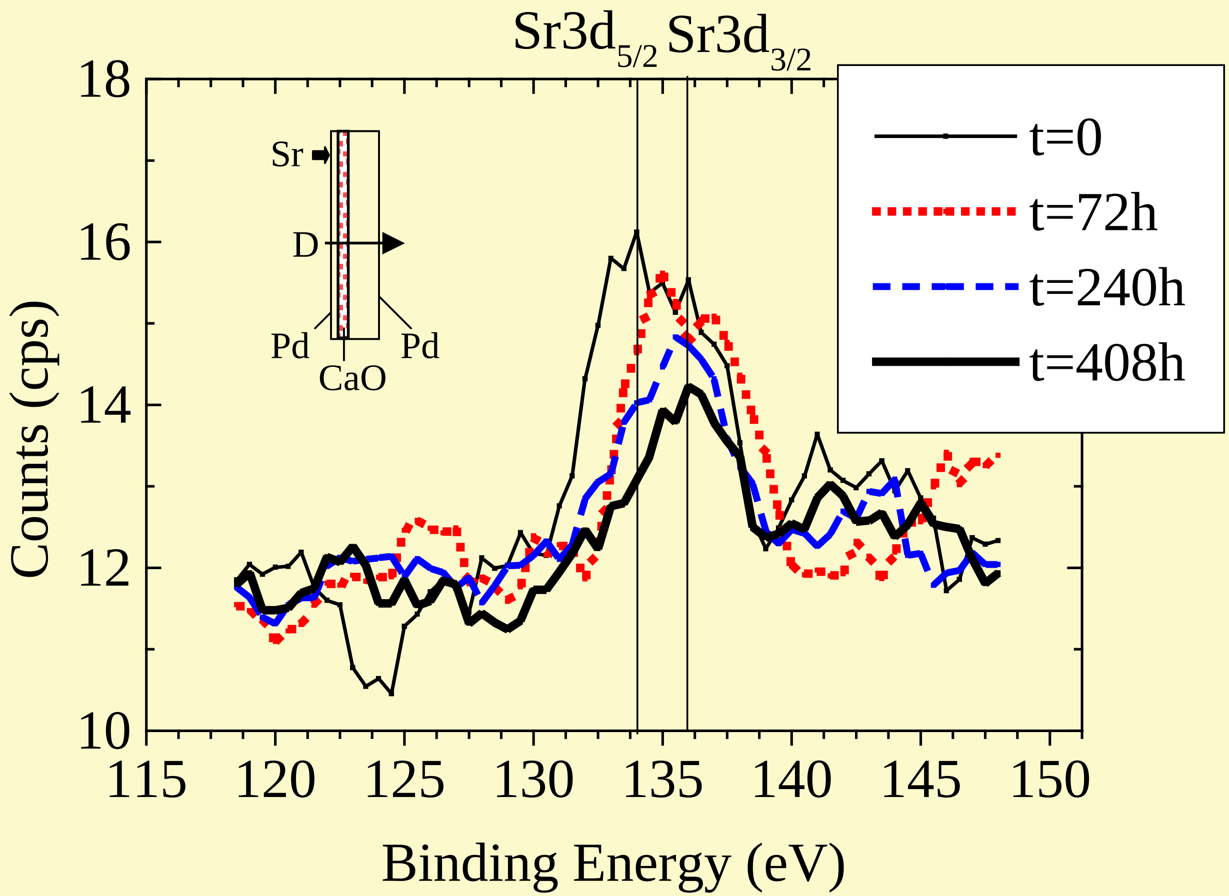
# Total Permeated D2 Gas and Products

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



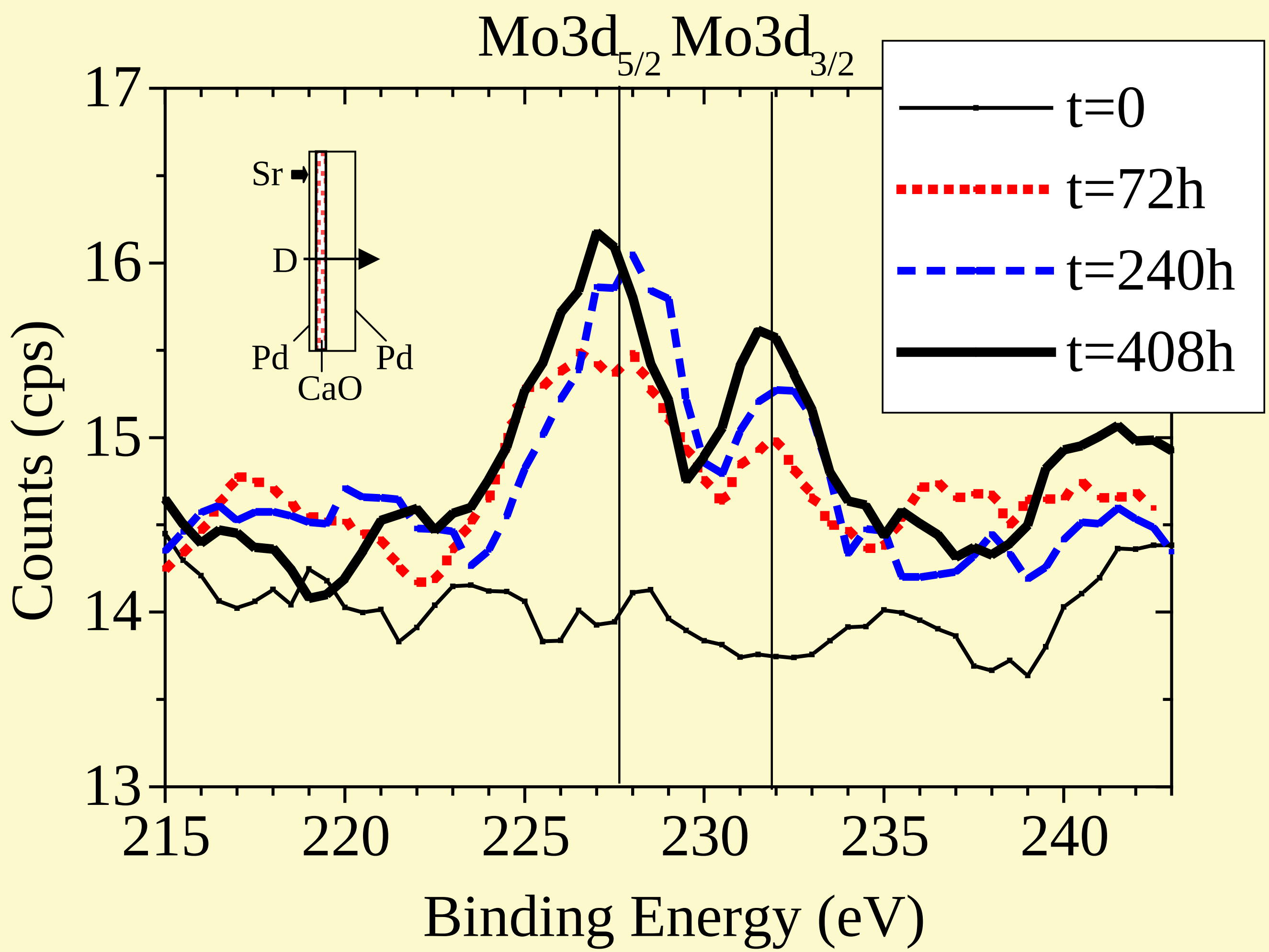
# Change of XPS Spectrum of Sr

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



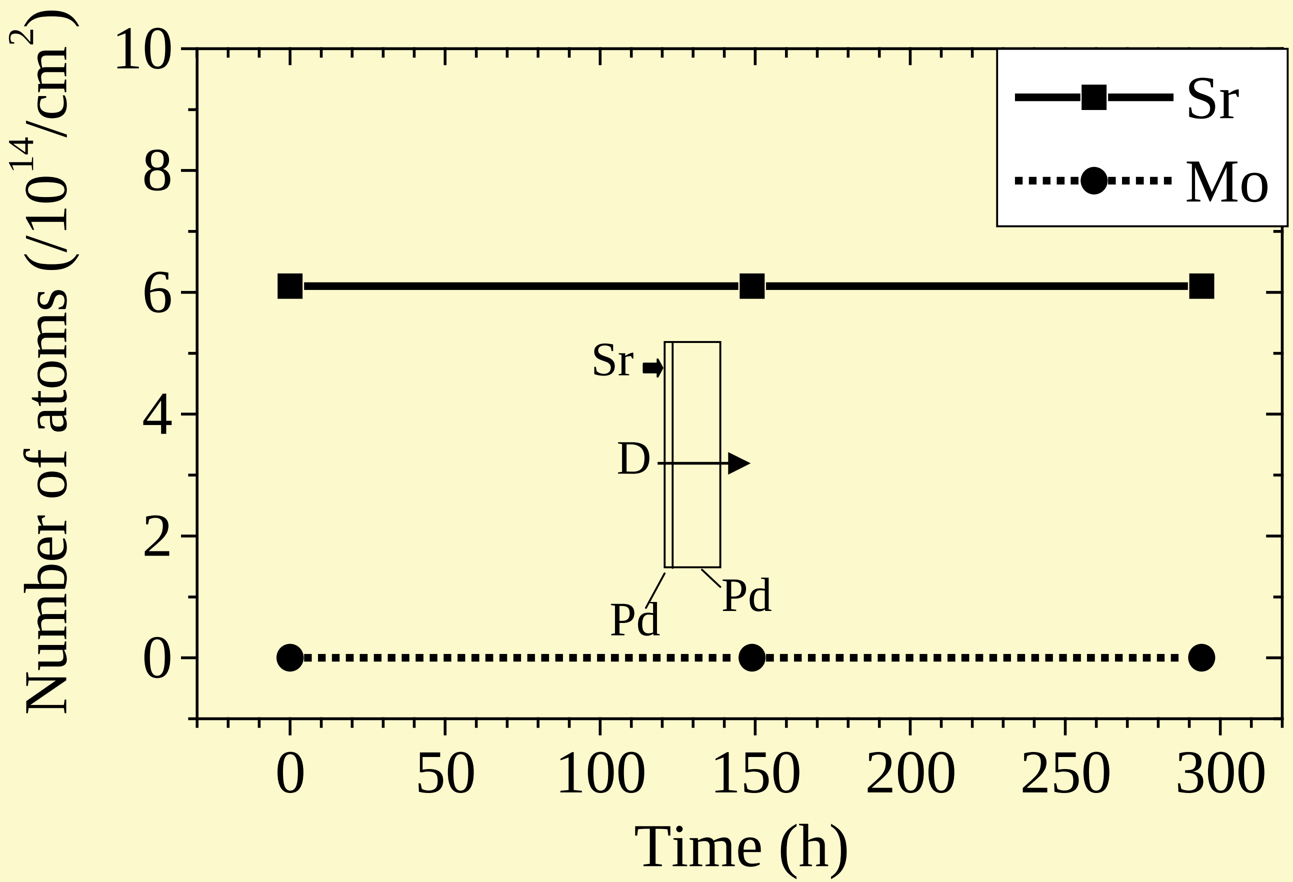
# Change of XPS Spectrum of Mo

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

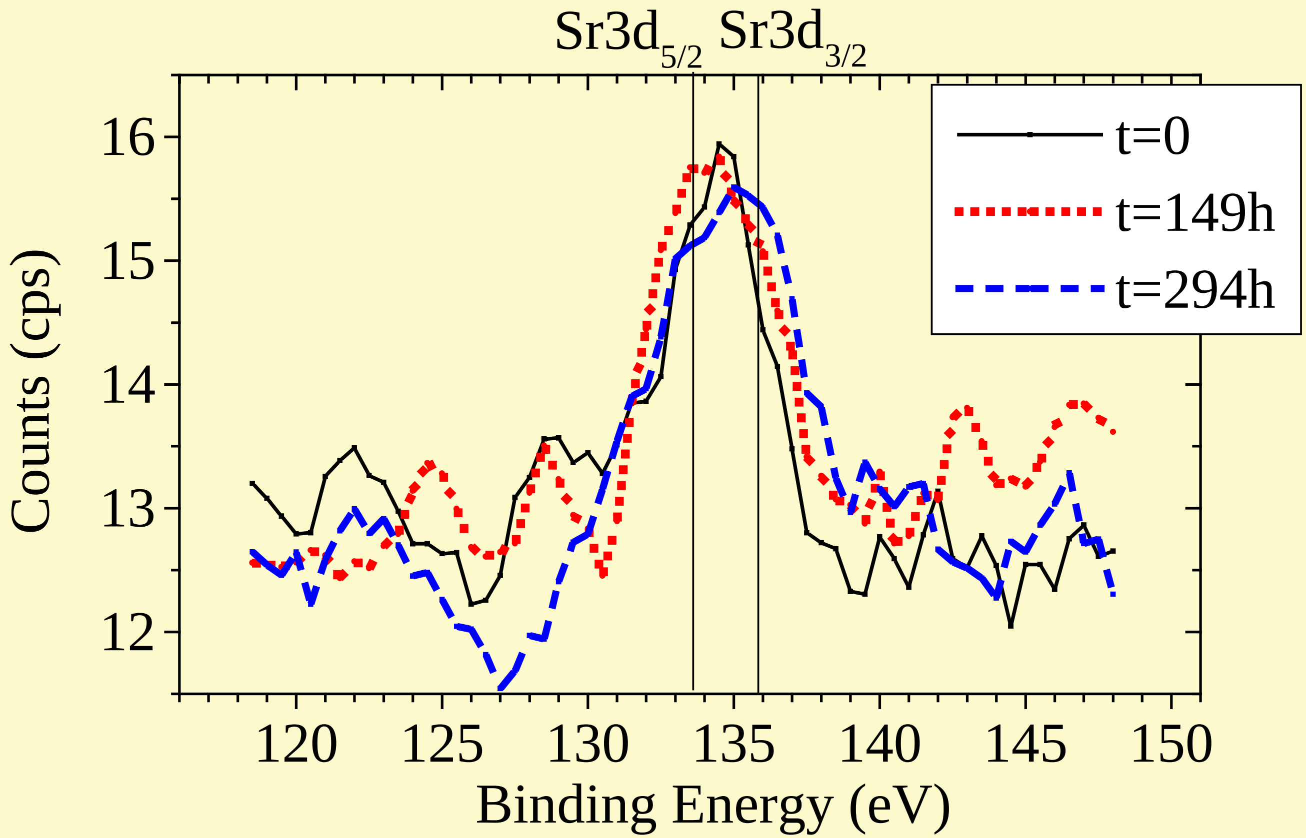
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



# XPS Spectrum of Sr (No CaO)

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

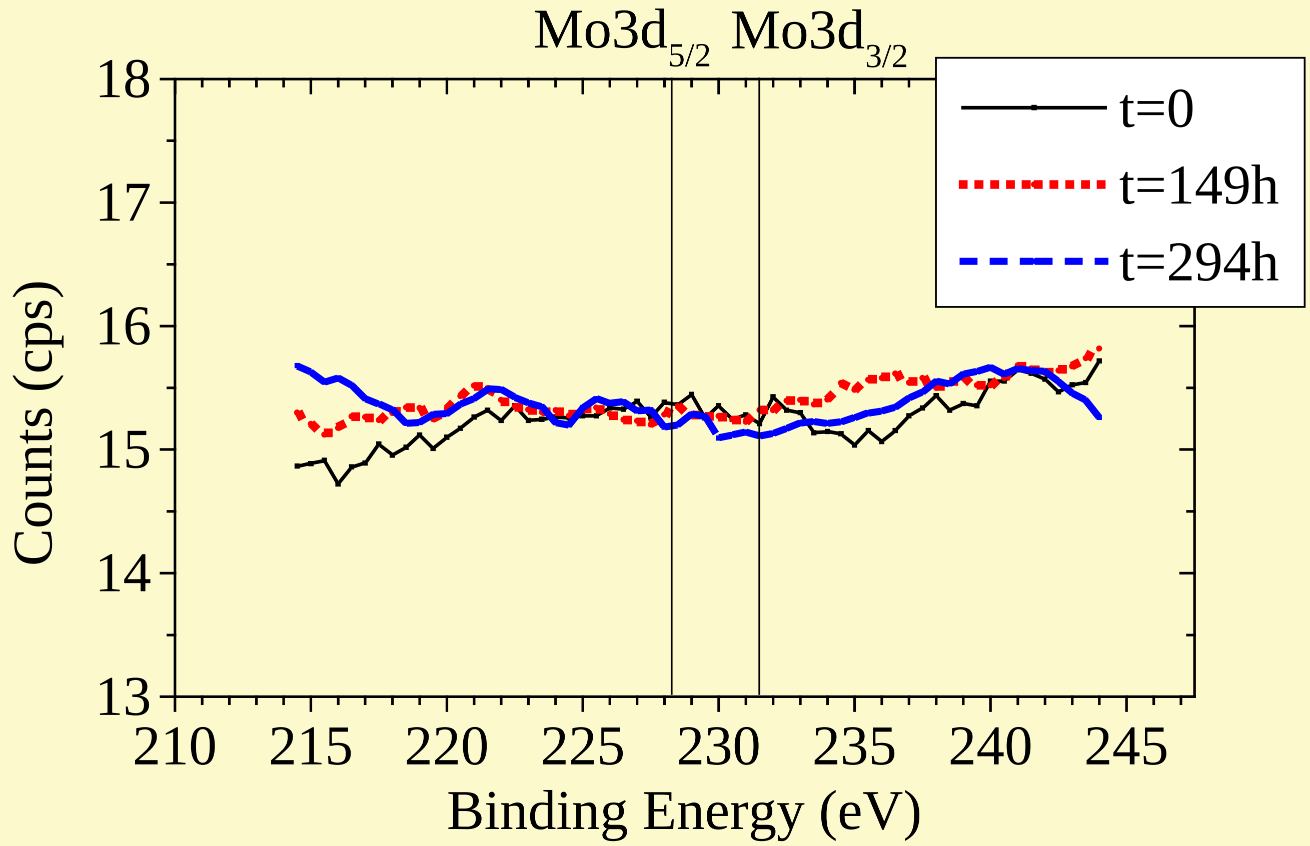
Pd/Pd/D2



# XPS Spectrum of Mo (No CaO)

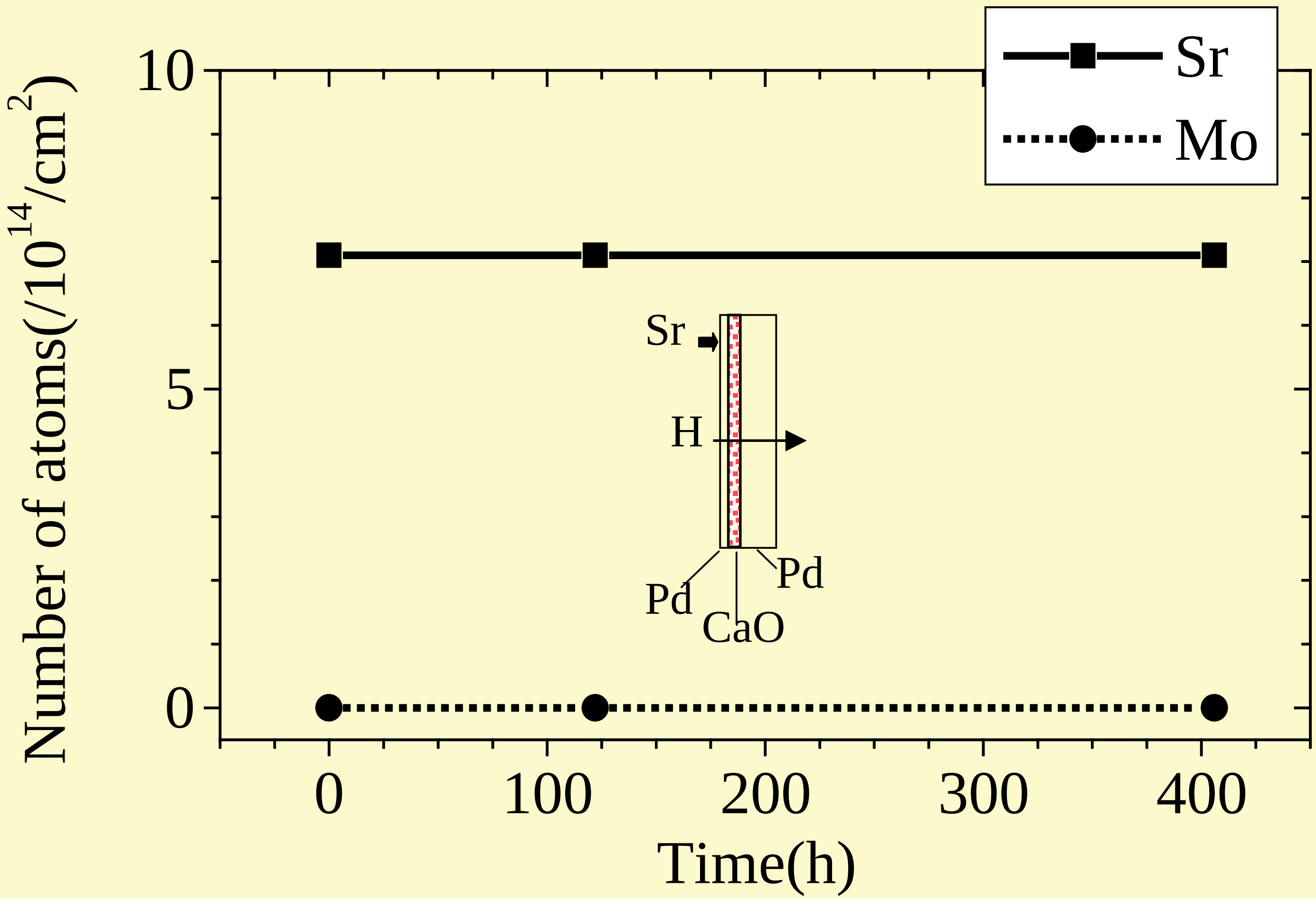
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

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

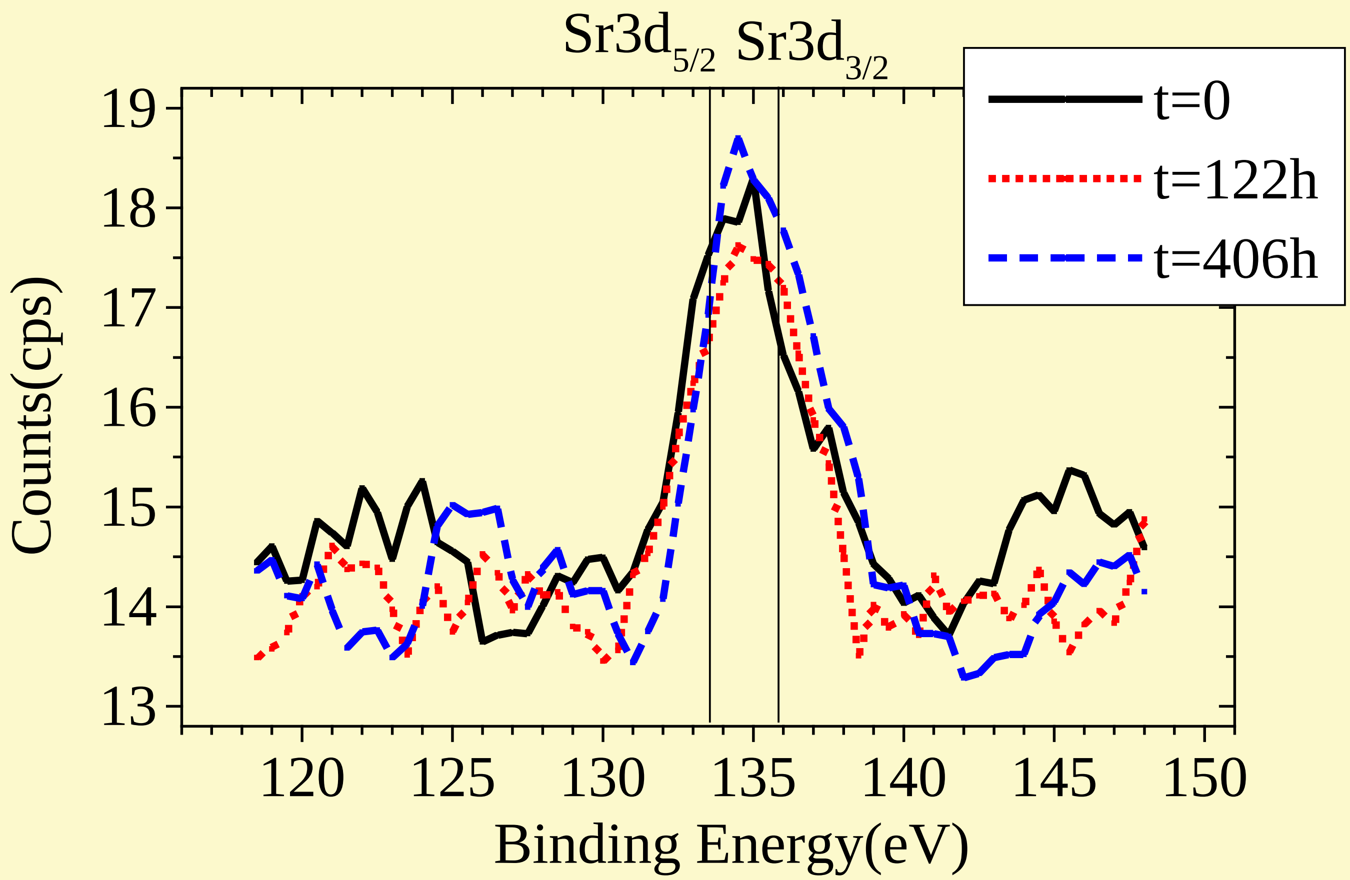
MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER



# XPS Spectrum of Sr ($H_2$ Permeation)

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

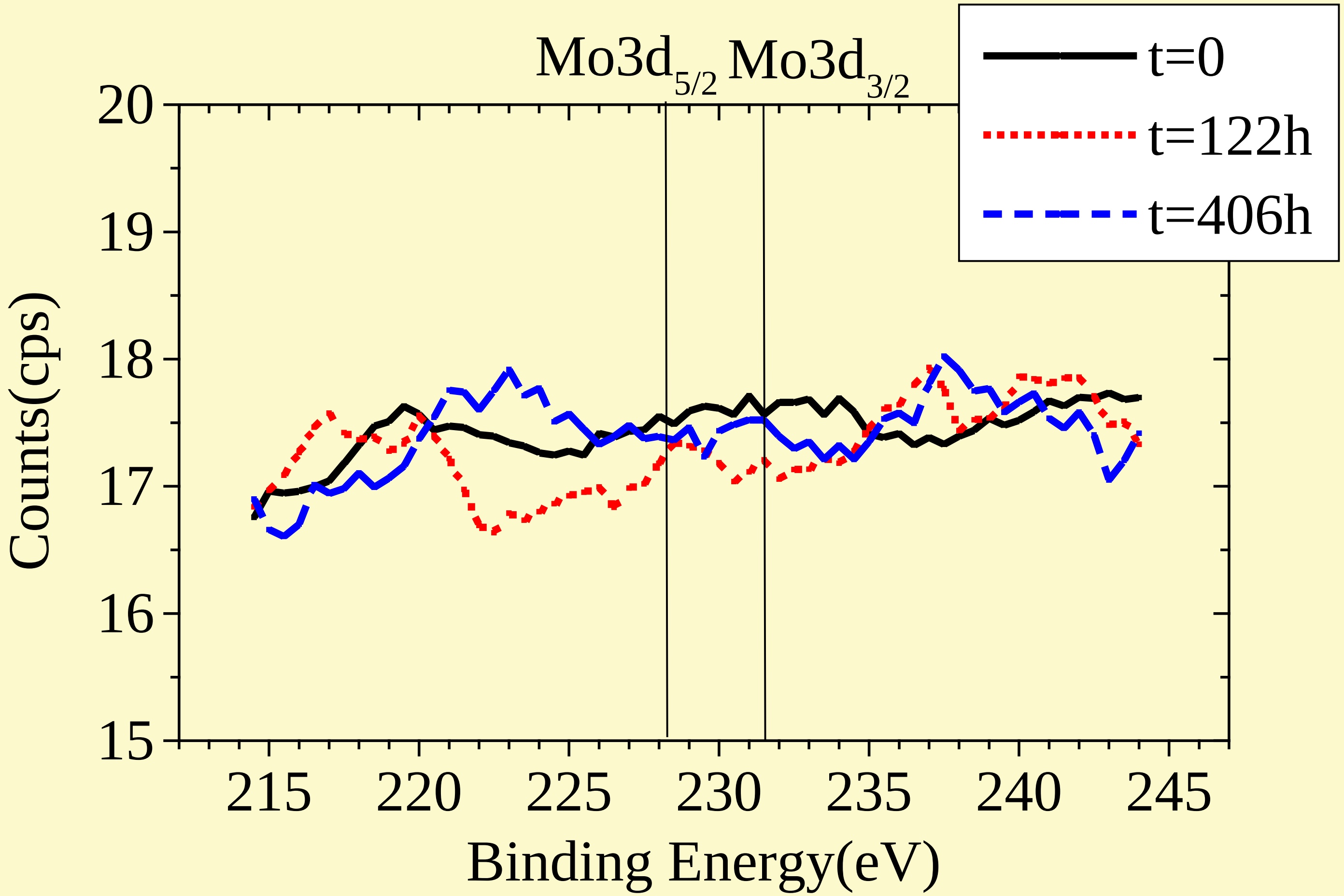
Pd/CaO/Pd/H2



# XPS Spectrum of Mo ($H_2$ Permeation)

MITSUBISHI HEAVY INDUSTRIES, LTD.
ADVANCED TECHNOLOGY RESEARCH CENTER

Pd/CaO/Pd/H2



# Separation of the Products and Contaminants(3)

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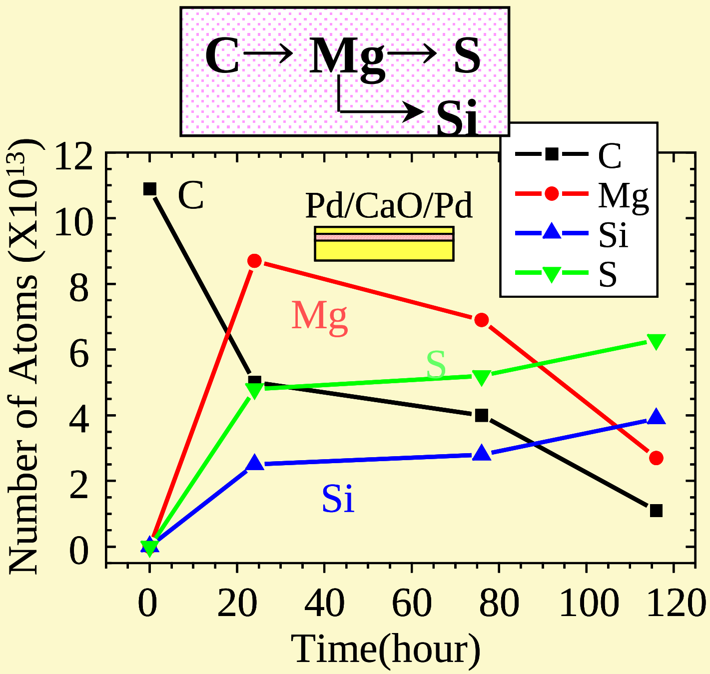
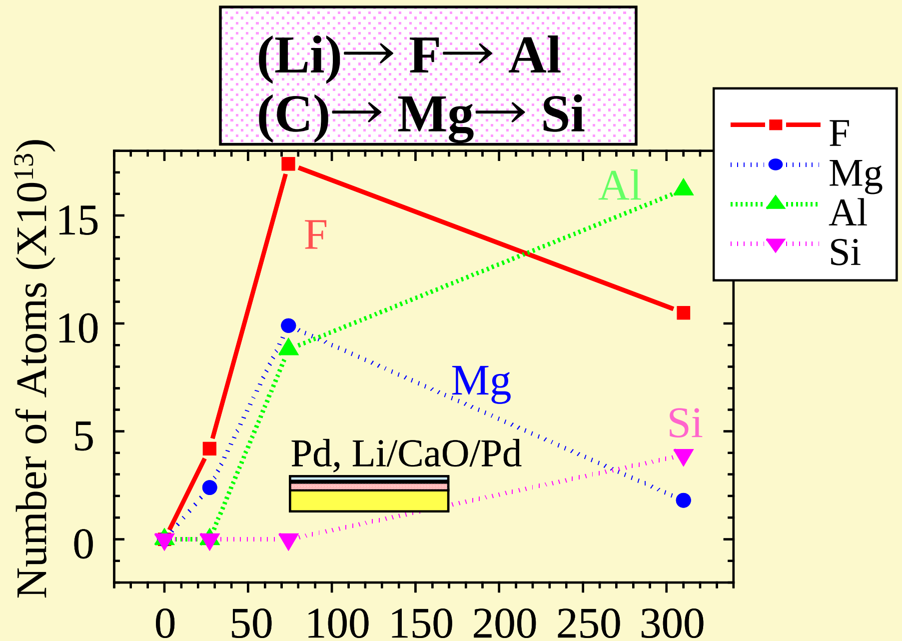
## 3. Variation of Detected Elements depending on the given Elements

**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

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

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

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

# 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**