Recent Advances in Deuterium Permeation Induced Transmutation Experiments using Nano-Structured Pd/CaO/Pd Multilayer Thin Film

Y.Iwamura , S.Tsuruga and T.Itoh

Advanced Technology Research Center,
Mitsubishi Heavy Industries, ltd., Japan

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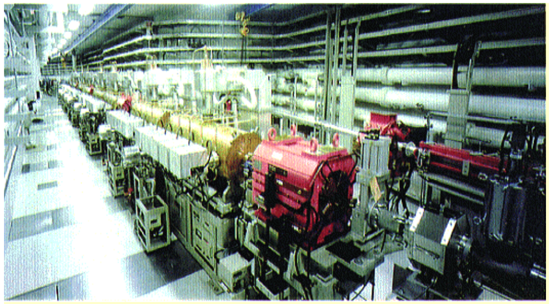
1. Introduction

Merits of Our Transmutation Method

Conventional Transmutation

Requires a large apparatus such as an accelerator and a nuclear reactor

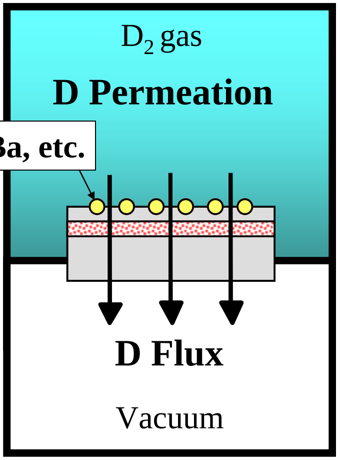


Permeation Induced Transmutation

Nuclear Transmutation can be induced only by deuterium permeation through our original nano-structured Pd multilayer film

Compact

Low Energy

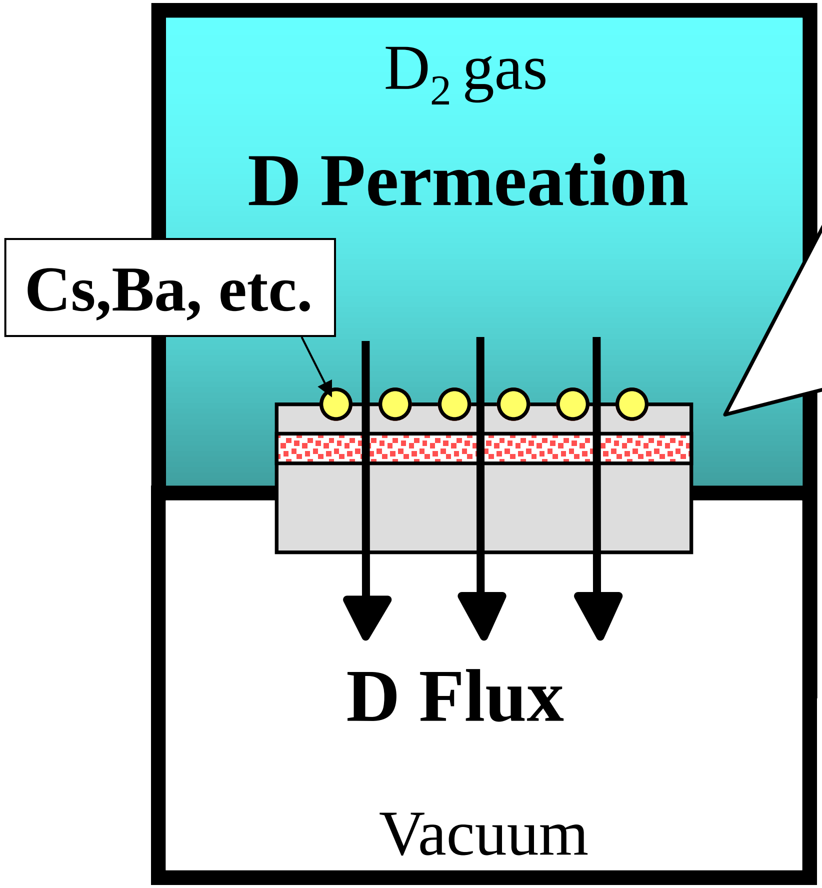


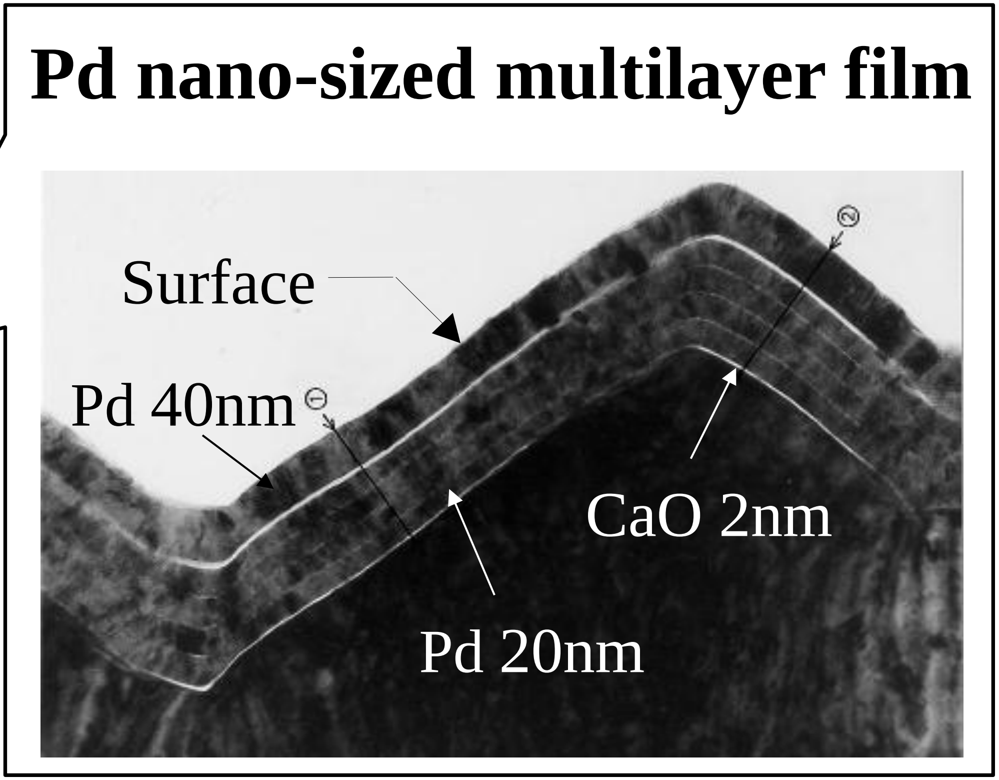
60-80 C
~1atm

Y. Iwamura et.al, J. Appl. Phys. 41, 4642-4648 (2002)

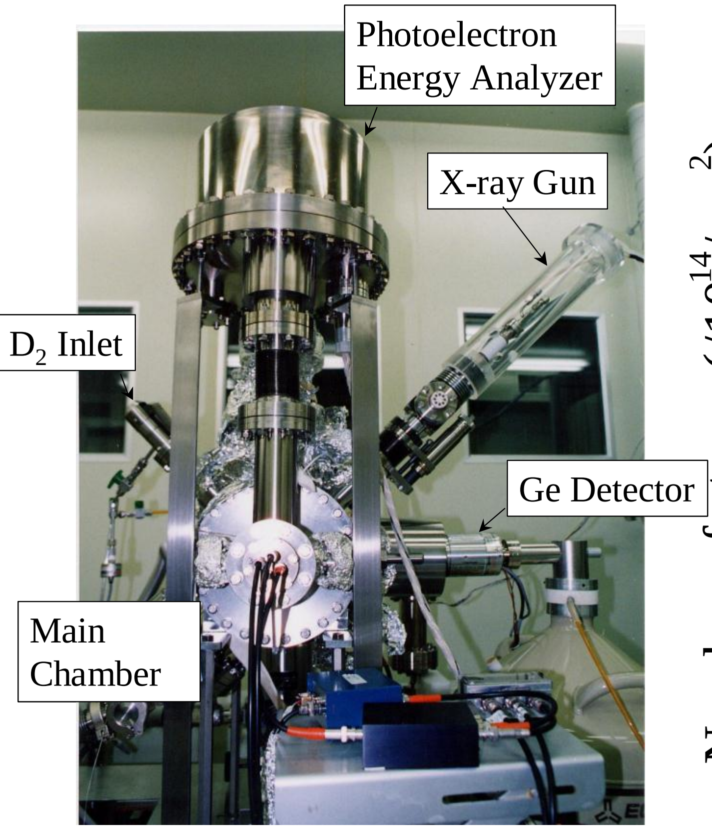
Features of the Mitsubishi Method

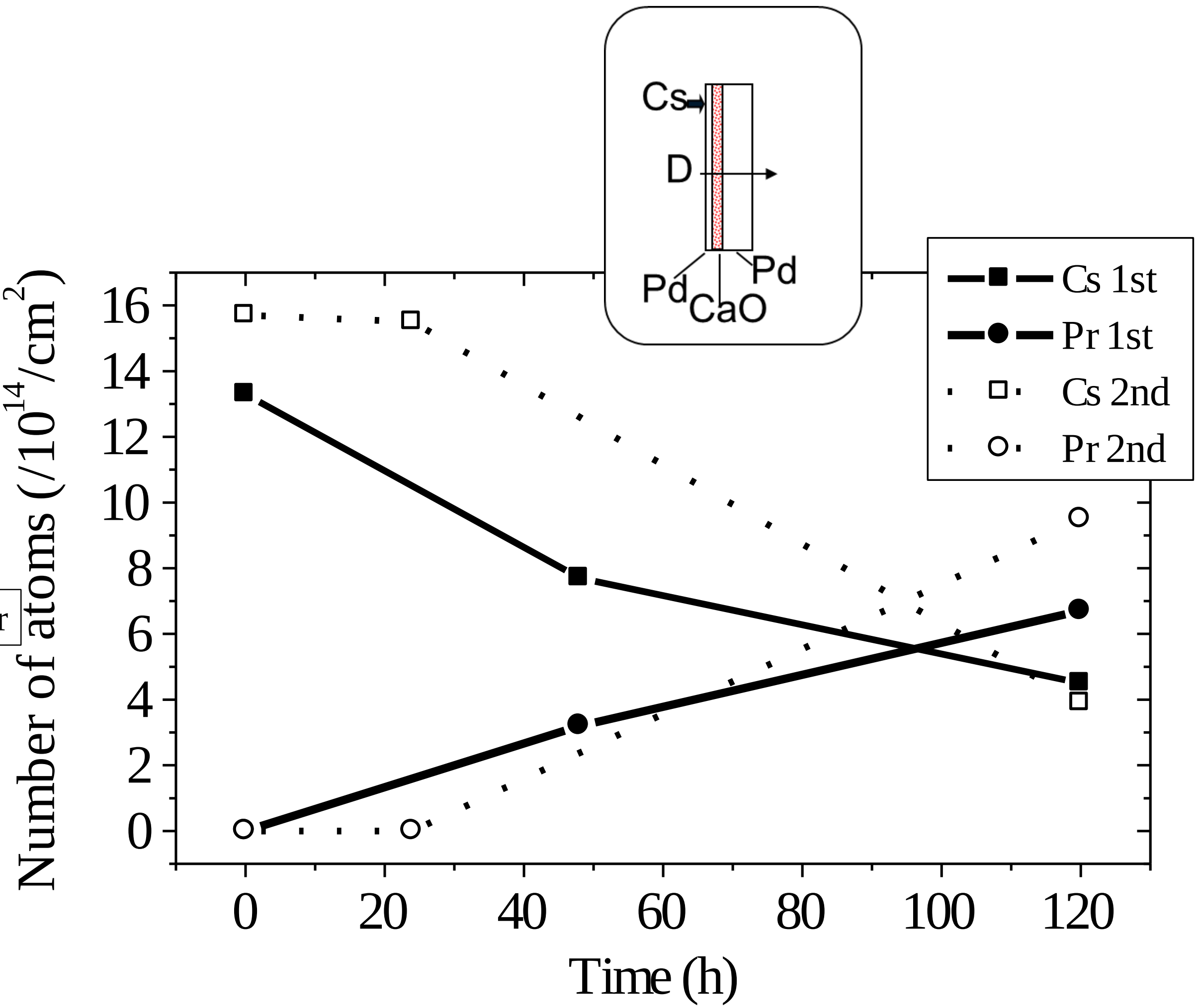
D_2 gas permeation through nano-structured Pd complex





Transmutation of Cs into Pr





Reactions observed so far in MHI

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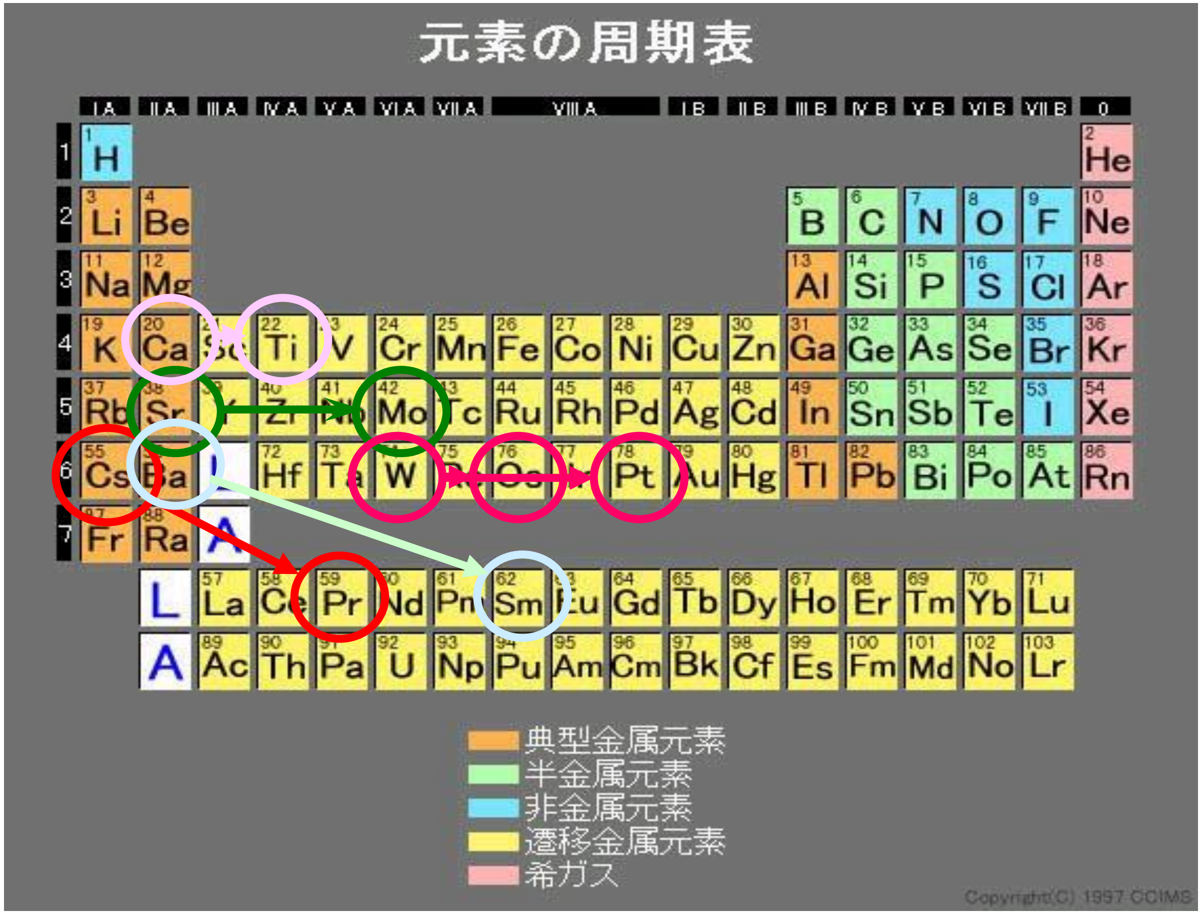


$$ {}^{133}_{55}Cs \xrightarrow{4d(2\alpha)} {}^{141}_{59}Pr $$

$$ {}^{88}_{38}Sr \xrightarrow{4d(2\alpha)} {}^{96}_{42}Mo $$

$$ {}^{138}_{56}Ba \xrightarrow{6d(3\alpha)} {}^{150}_{62}Sm $$

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

$$ {}^{44}_{20}Ca \xrightarrow{2d(\alpha)} {}^{48}_{22}Ti $$

$$ {}^{184}_{74}W \xrightarrow{2d(\alpha)} {}^{188}_{76}Os $$

$$ {}^{182}_{74}W \xrightarrow{4d(2\alpha)} {}^{190}_{78}Pt $$

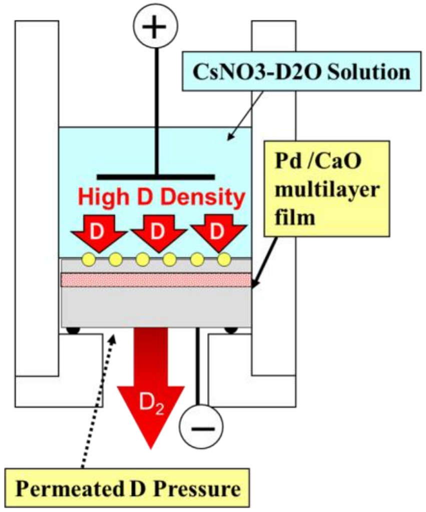
1)Alkali metals; Electron Emitter

2)2d, 4d, 6d; α capture reactions

Key factors in Permeation Experiments

Key factors based on experimental results

- ICCF18 -



Hypothesis

Local Deuteron Density

High Density

Electronic Structure

Electron Rich

→

Electrochemical Permeation
(since ICCF17)

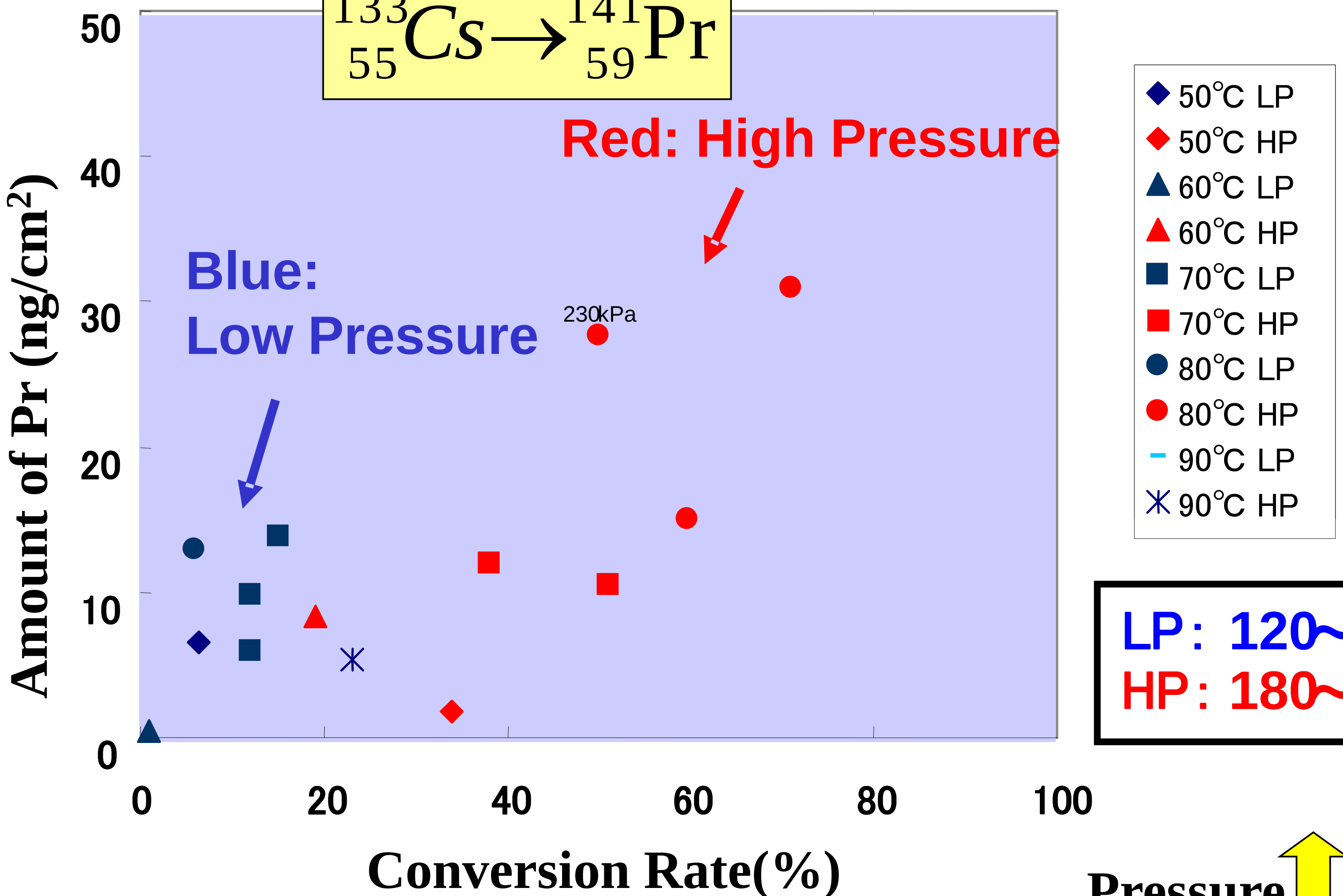
1. Transmutation products **confirmed by XPS**

2. Observation of **γ-ray peaks** supposed to be induced by the increase of transmutation products

2. Increase of Transmutation Products induced by the Increase of Deuteron Density

Pr Dependence on D_2 gas pressure

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



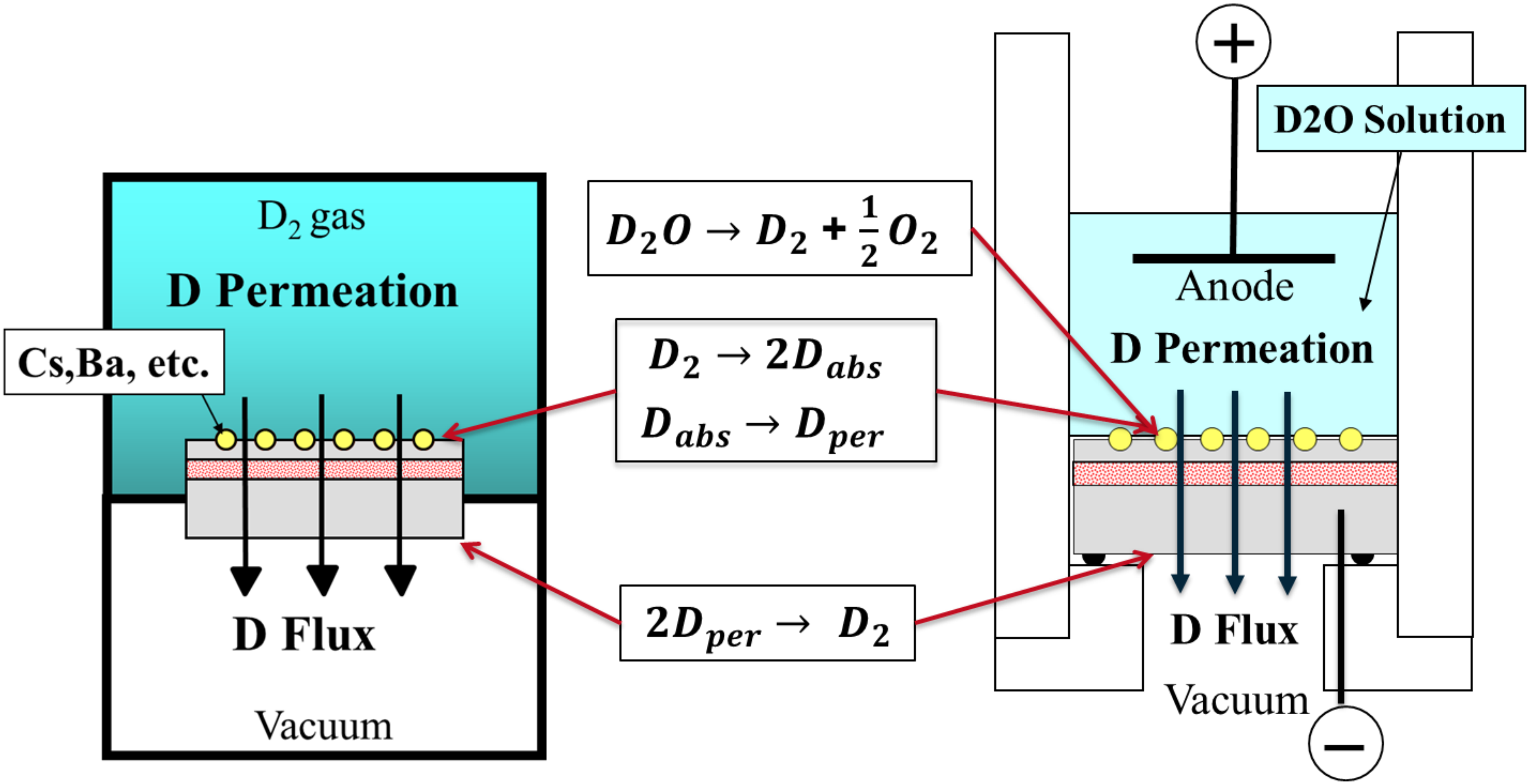
LP : 120～130kPa
HP : 180～230 kPa

Pressure ↑ → Pr ↑

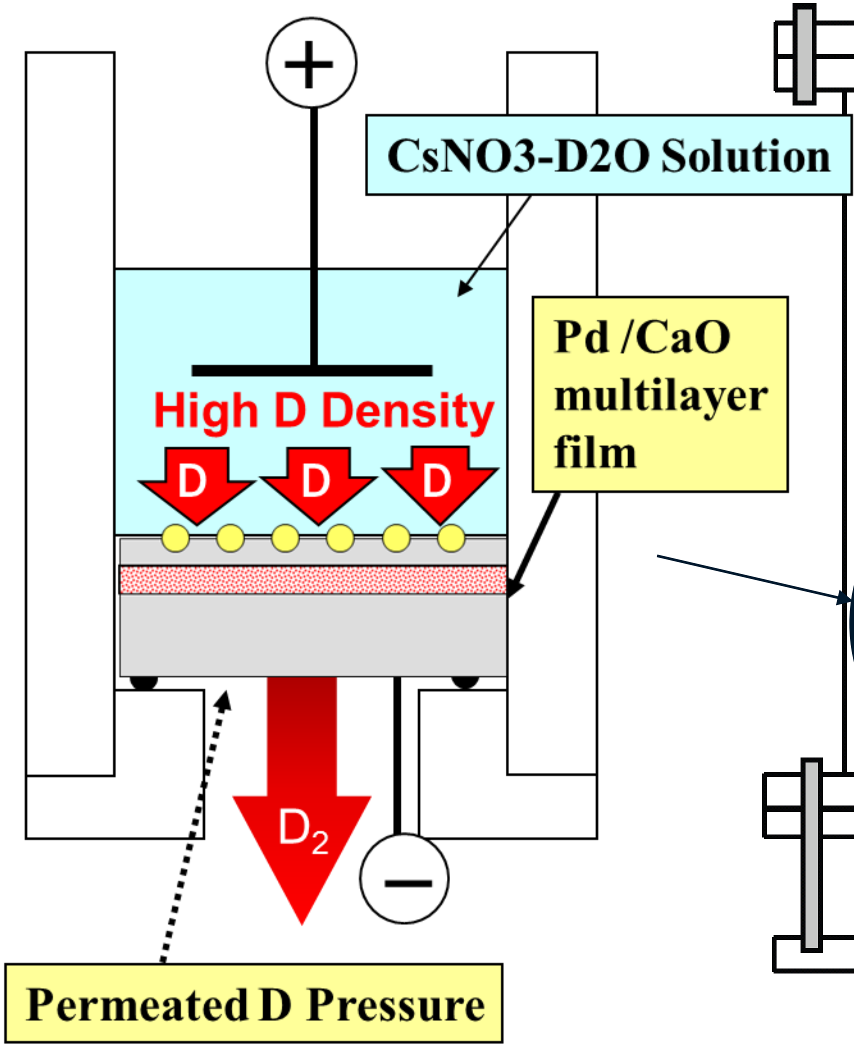
Gas vs. Electrochemical Permeation

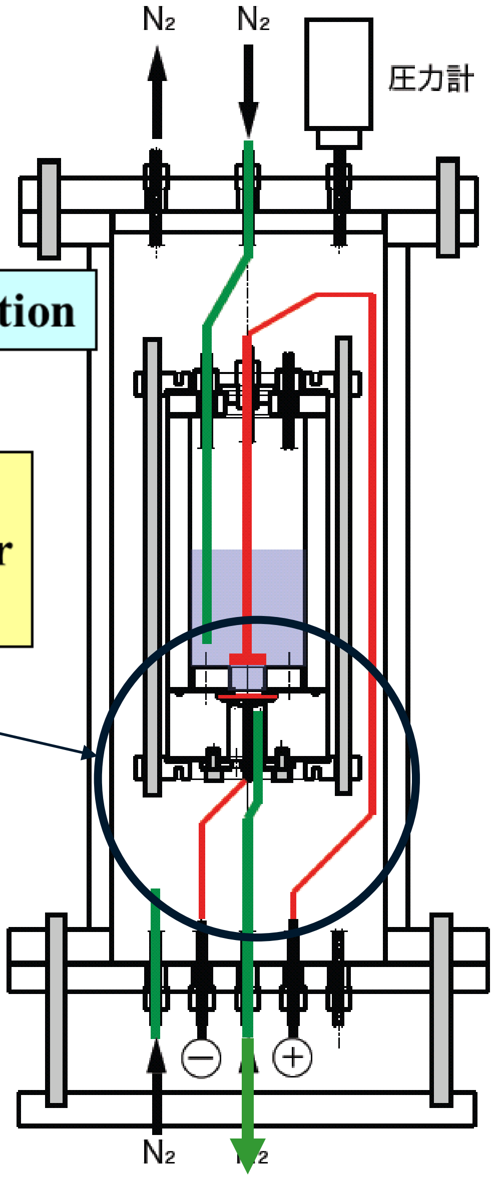
Gas Permeation

Electrochemical Permeation



Experimental Apparatus aiming Increase of D Density





3. Observation of γ-ray peaks supposed to be induced by the increase of transmutation products

Introduce a Gamma-ray Detector

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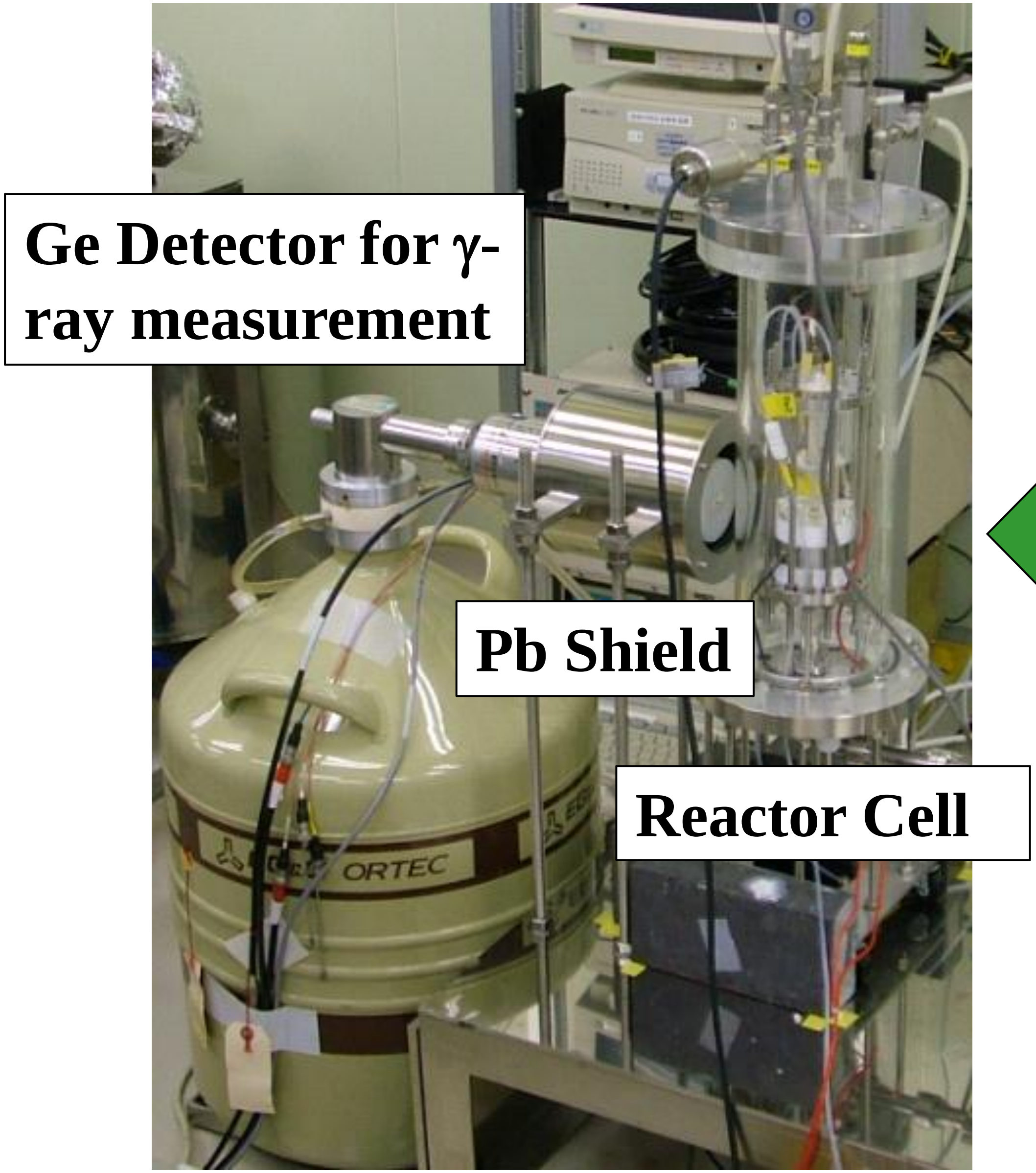
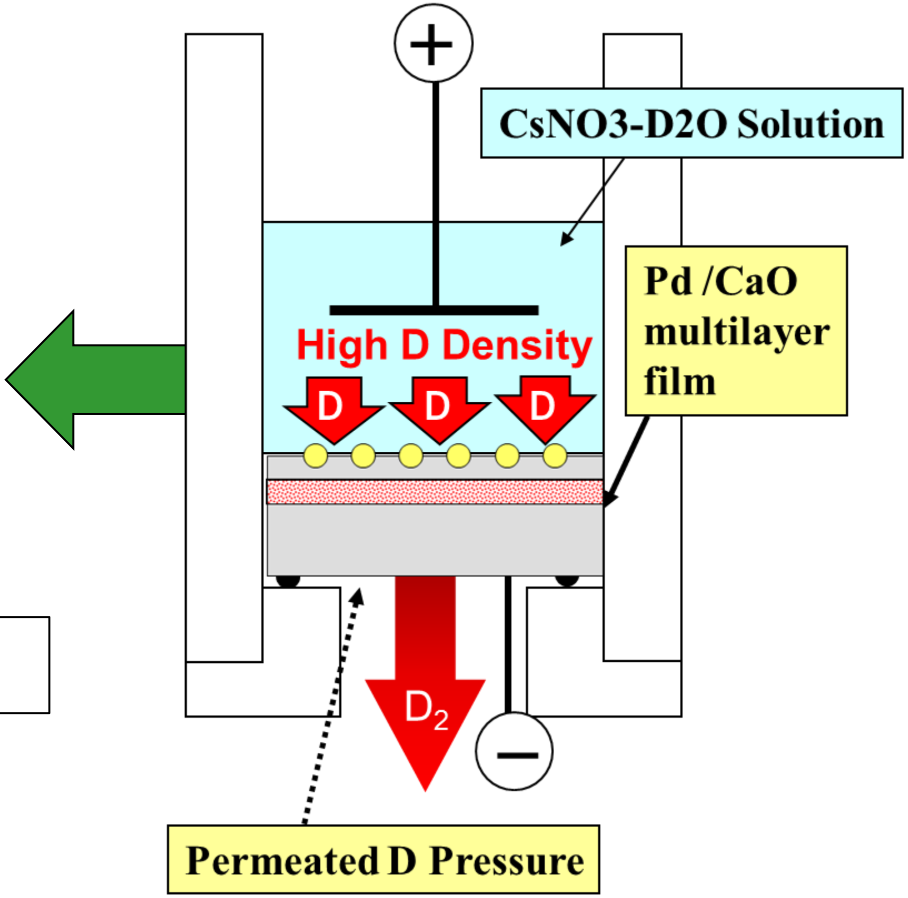
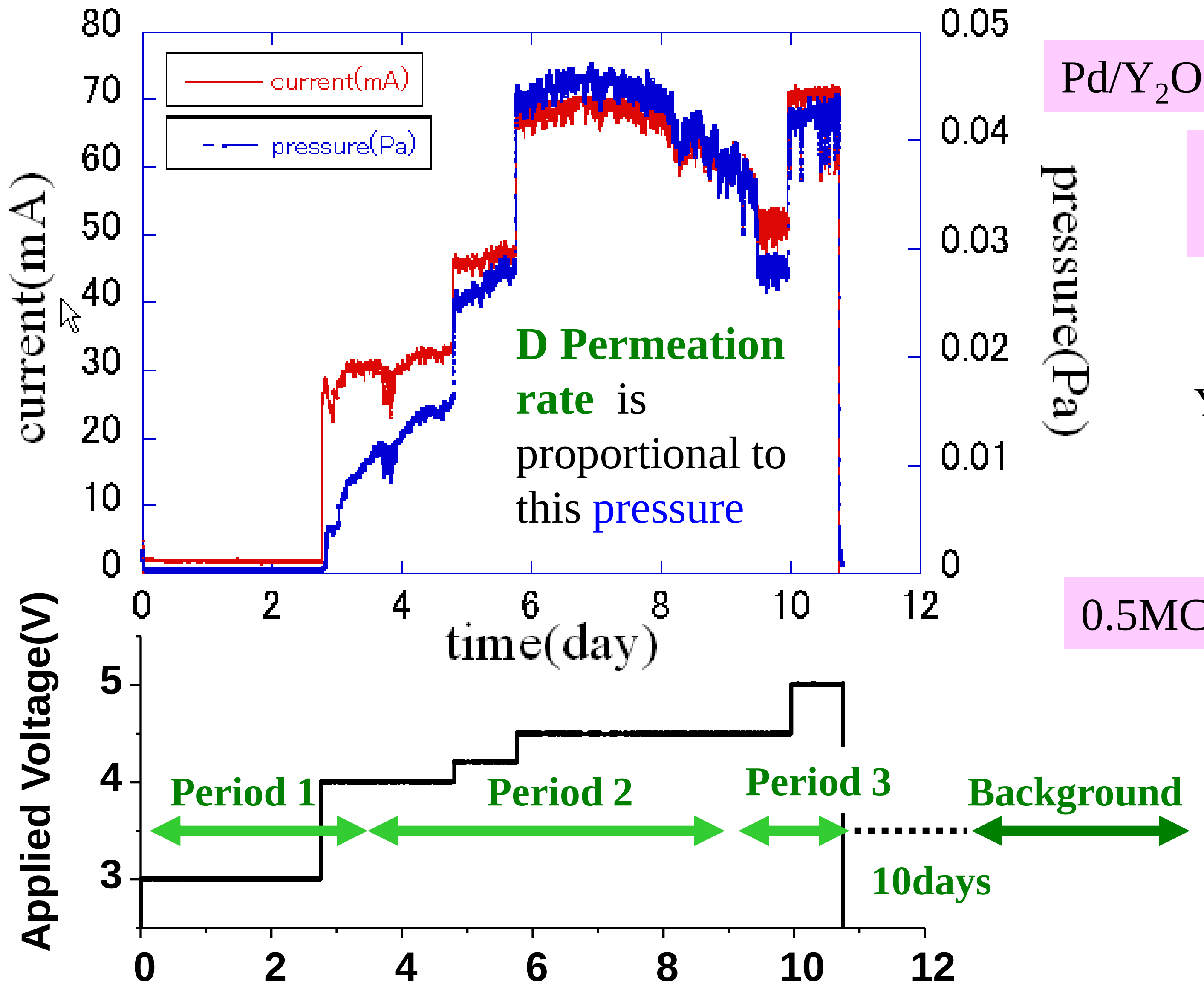


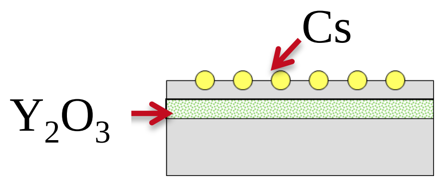


Example of Gamma-Ray Detection; E16



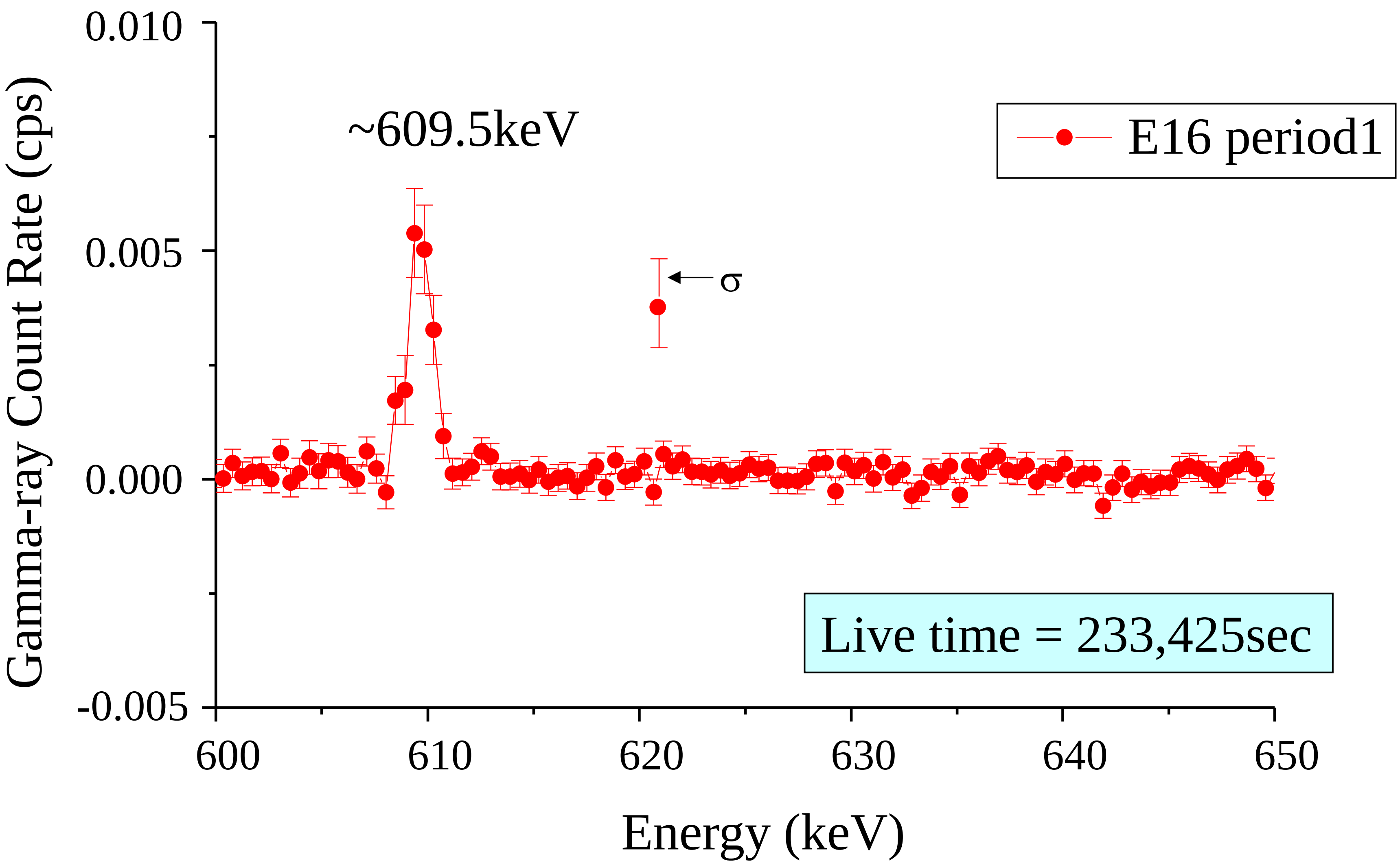
Pd/Y_2O_3/Pd multilayer film

Cs ion implanted
$1\times10^{16}/cm^2$ 20kV

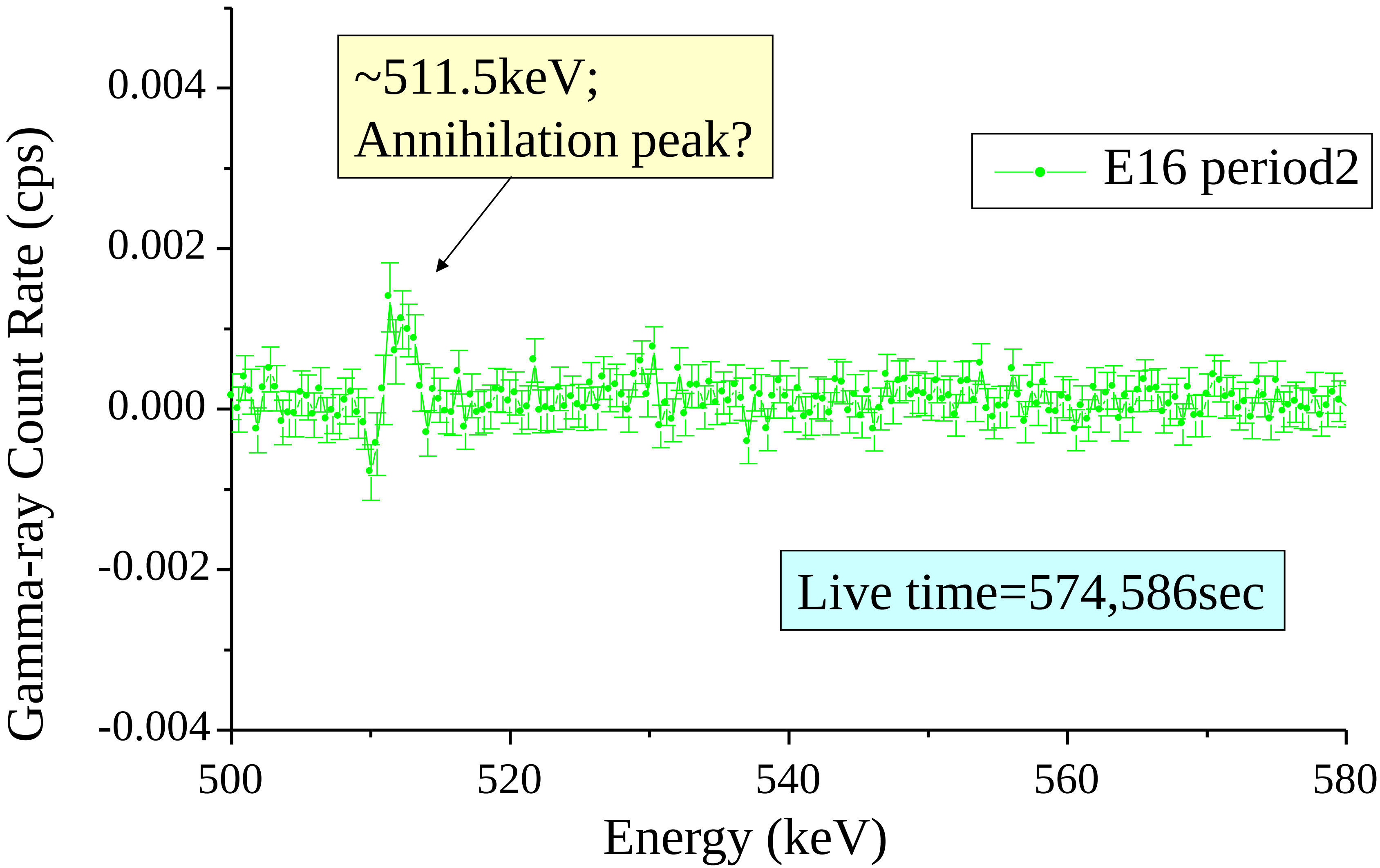


0.5MCsNO$_3$-D$_2$O Solution

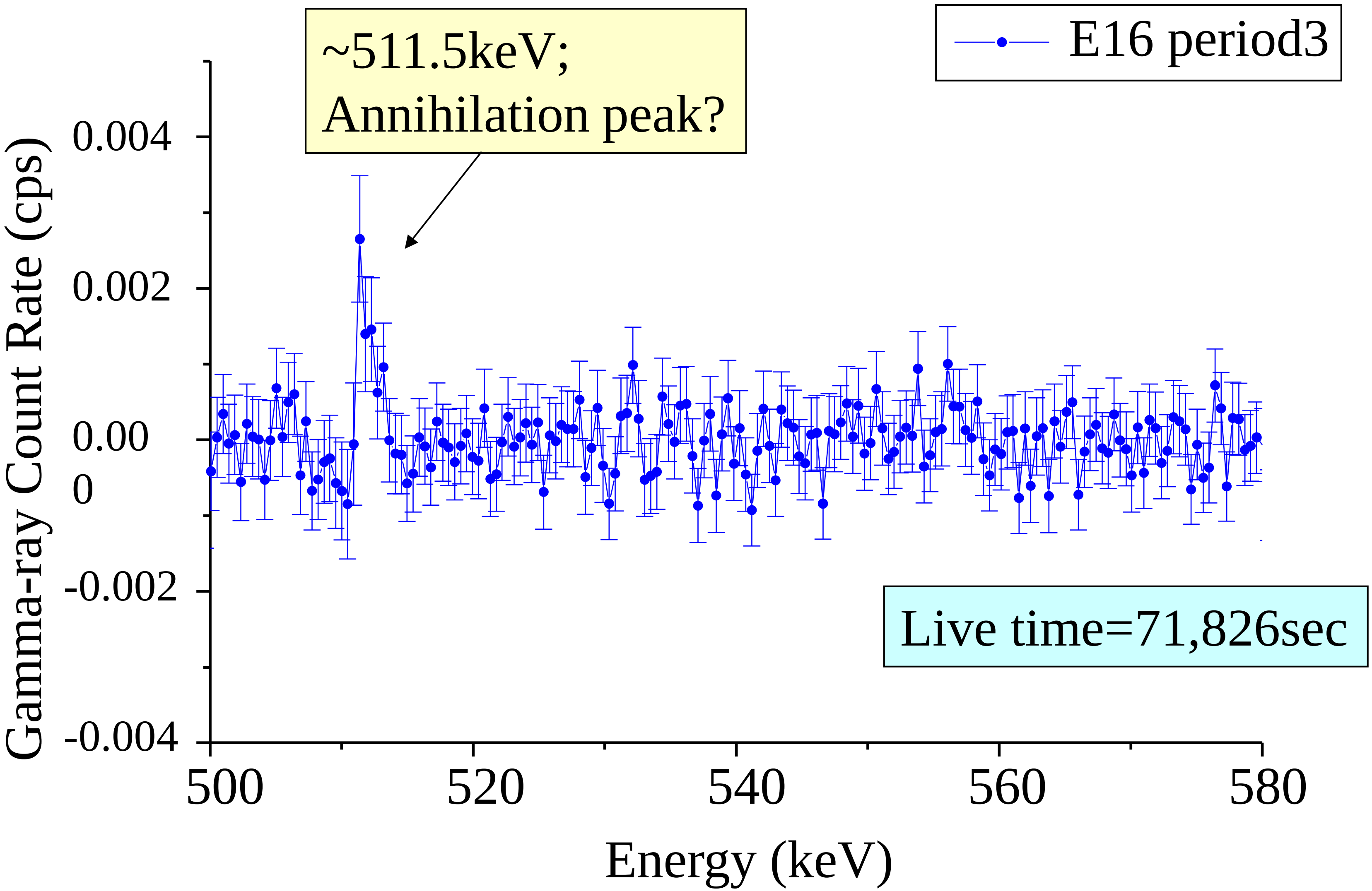
Gamma-ray Measurement (period 1)



Gamma-ray Measurement (period 2)



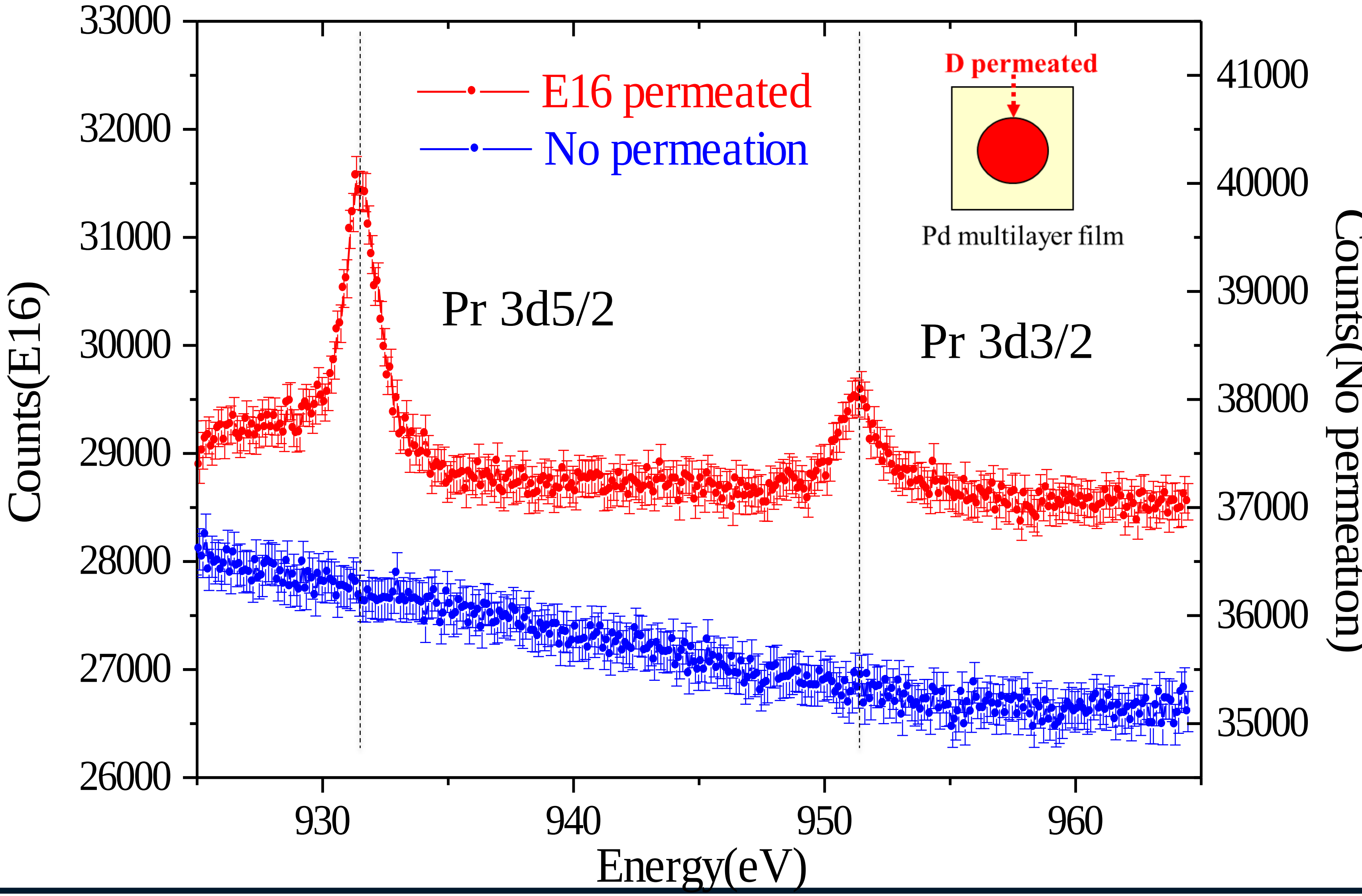
Gamma-ray Measurement (period 3)

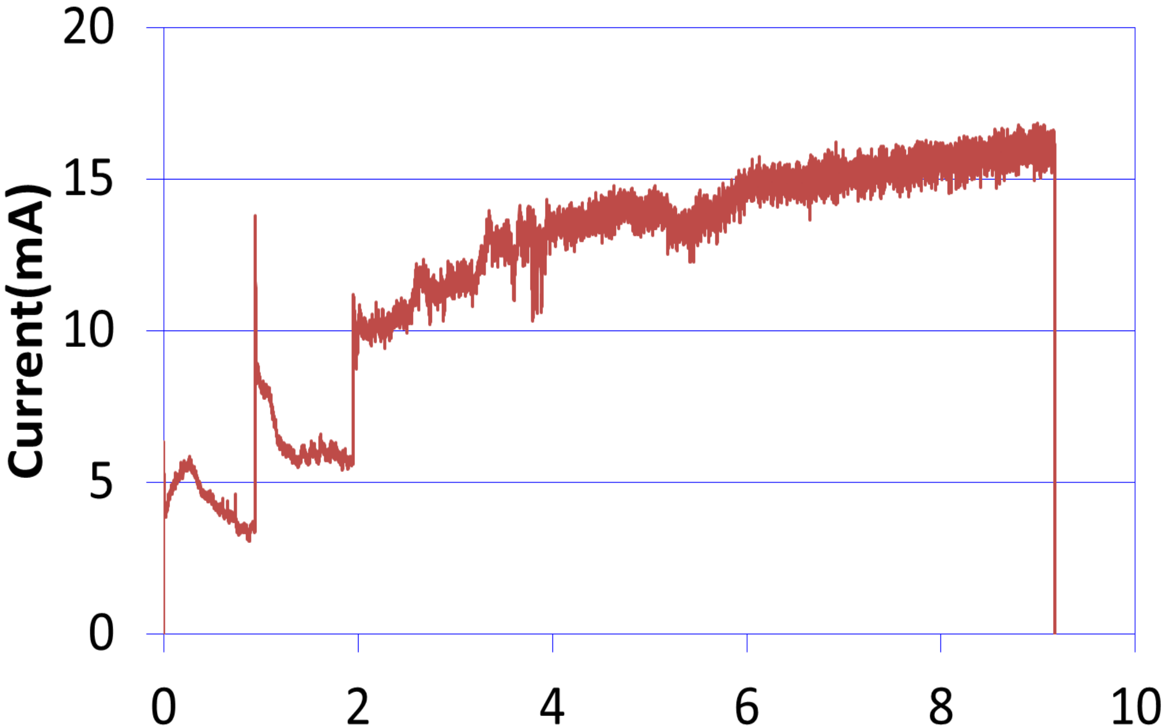


E16 Gamma-ray Measurement Summary

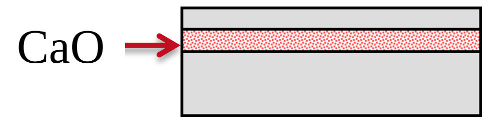
Time	Gamma-ray
Period 1	609.5keV gamma-ray detected No 511keV detected
Period 2	511.5keV gamma-ray detected No 609.5keV detected
Period 3	511.5keV gamma-ray detected No 609.5keV detected

Pr detected by XPS from the center of E16 sample

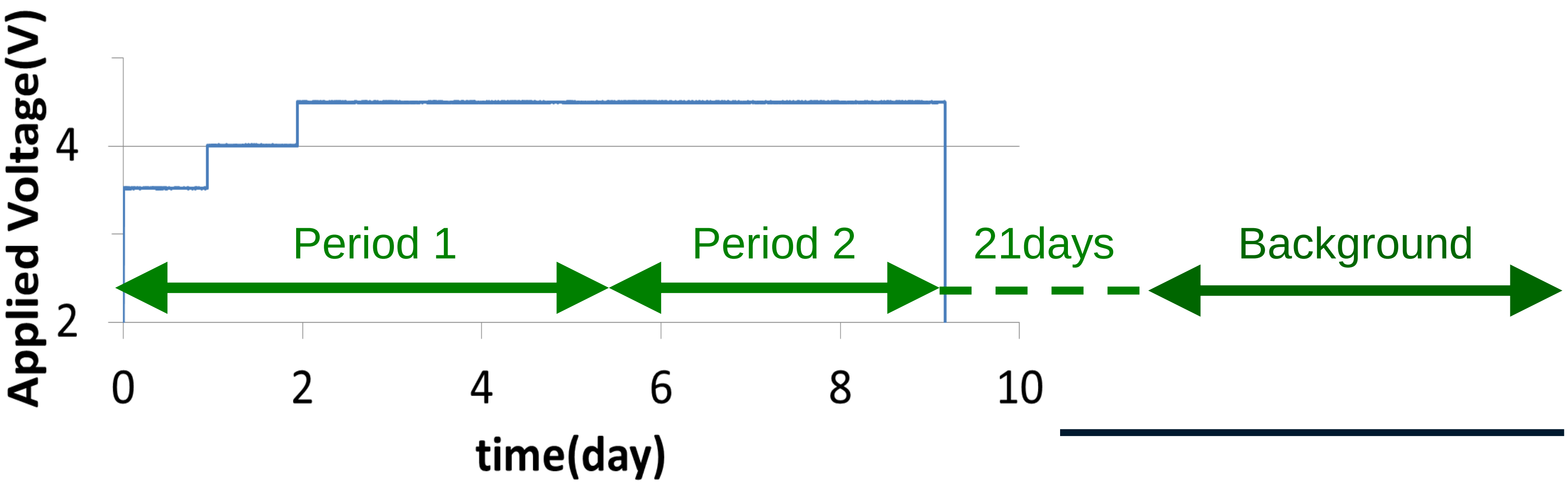




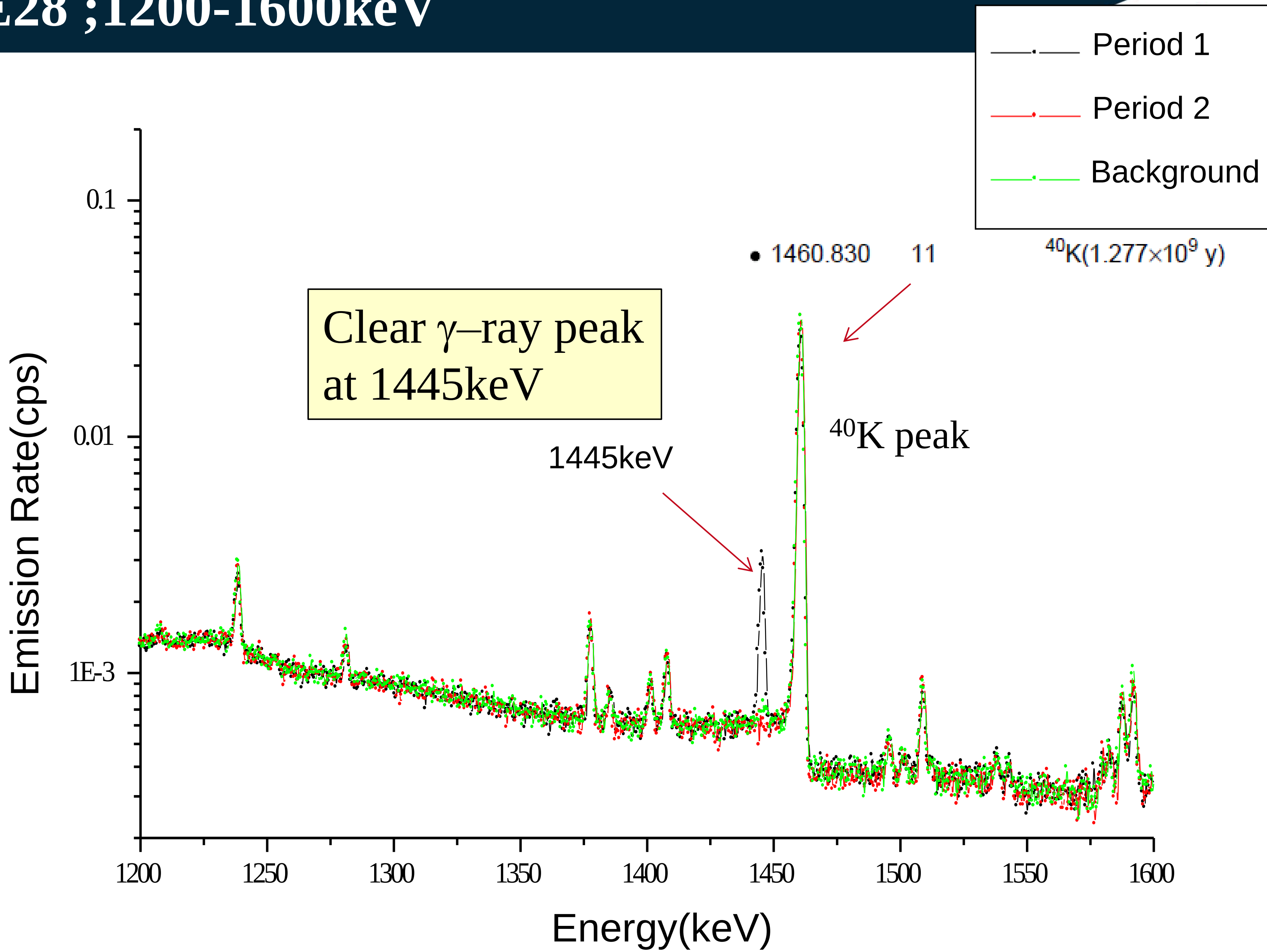
Pd/CaO/Pd multilayer film



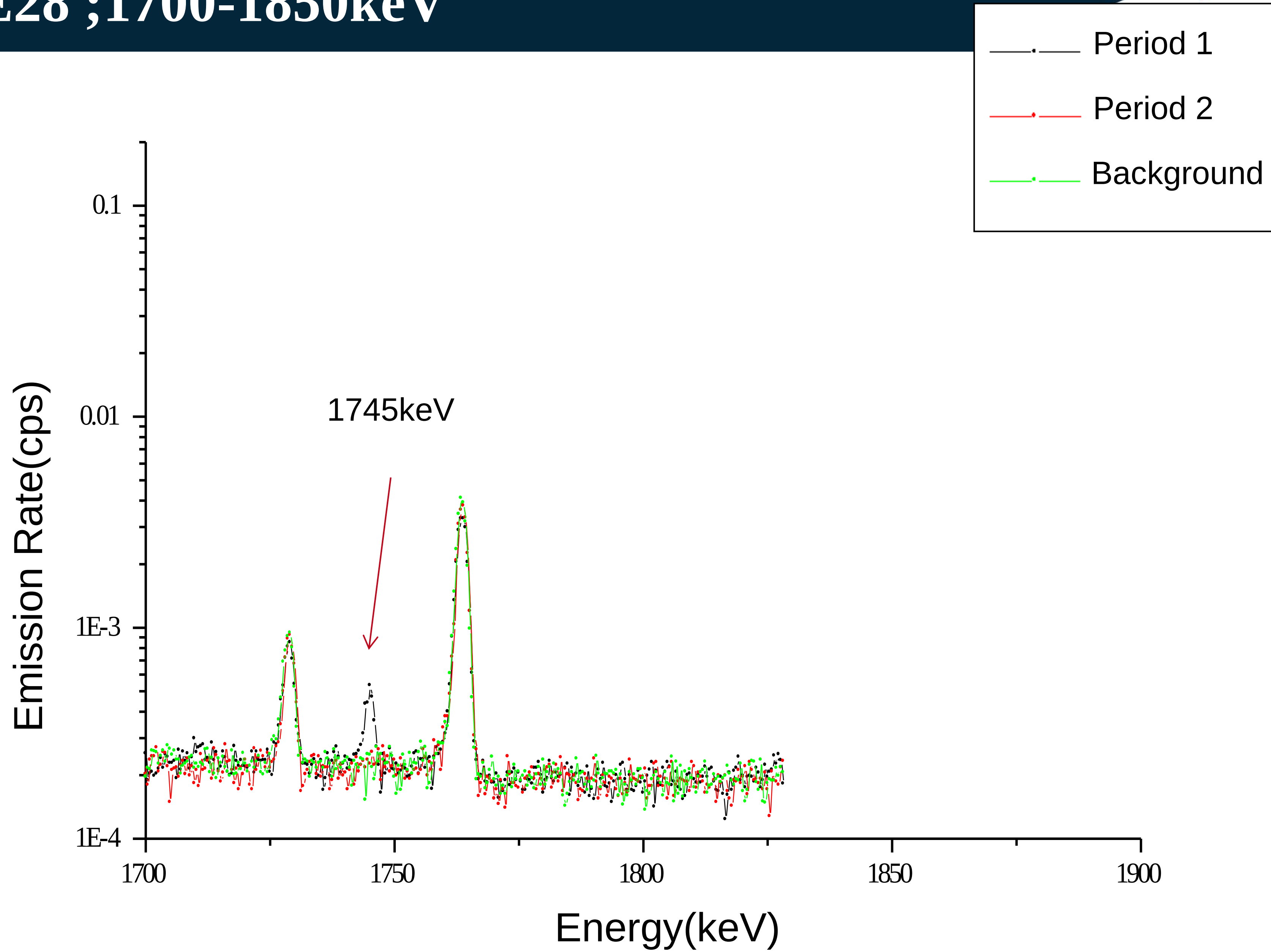
$0.1MCsNO_3$-D_2O Solution



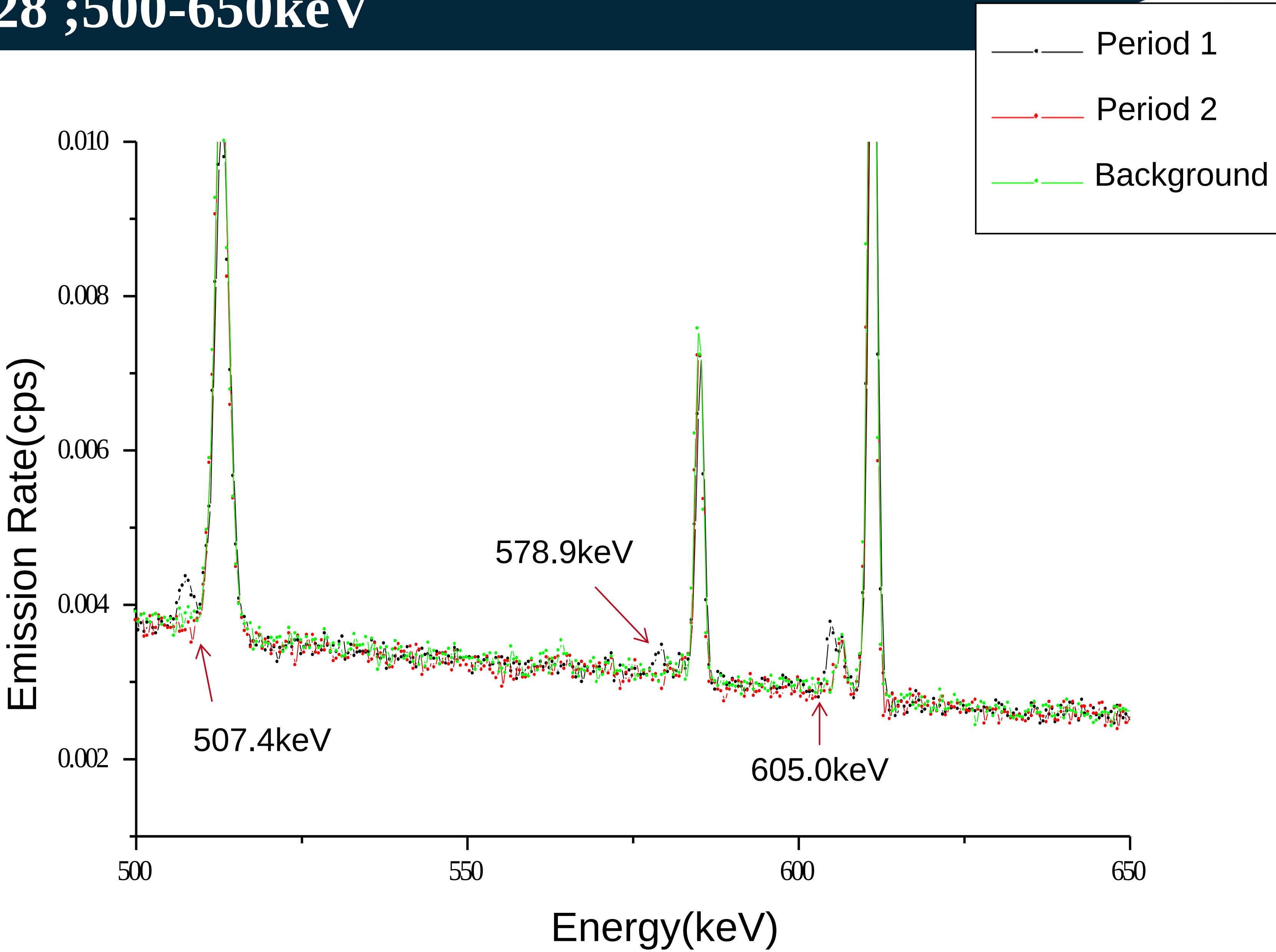
E28 ;1200-1600keV



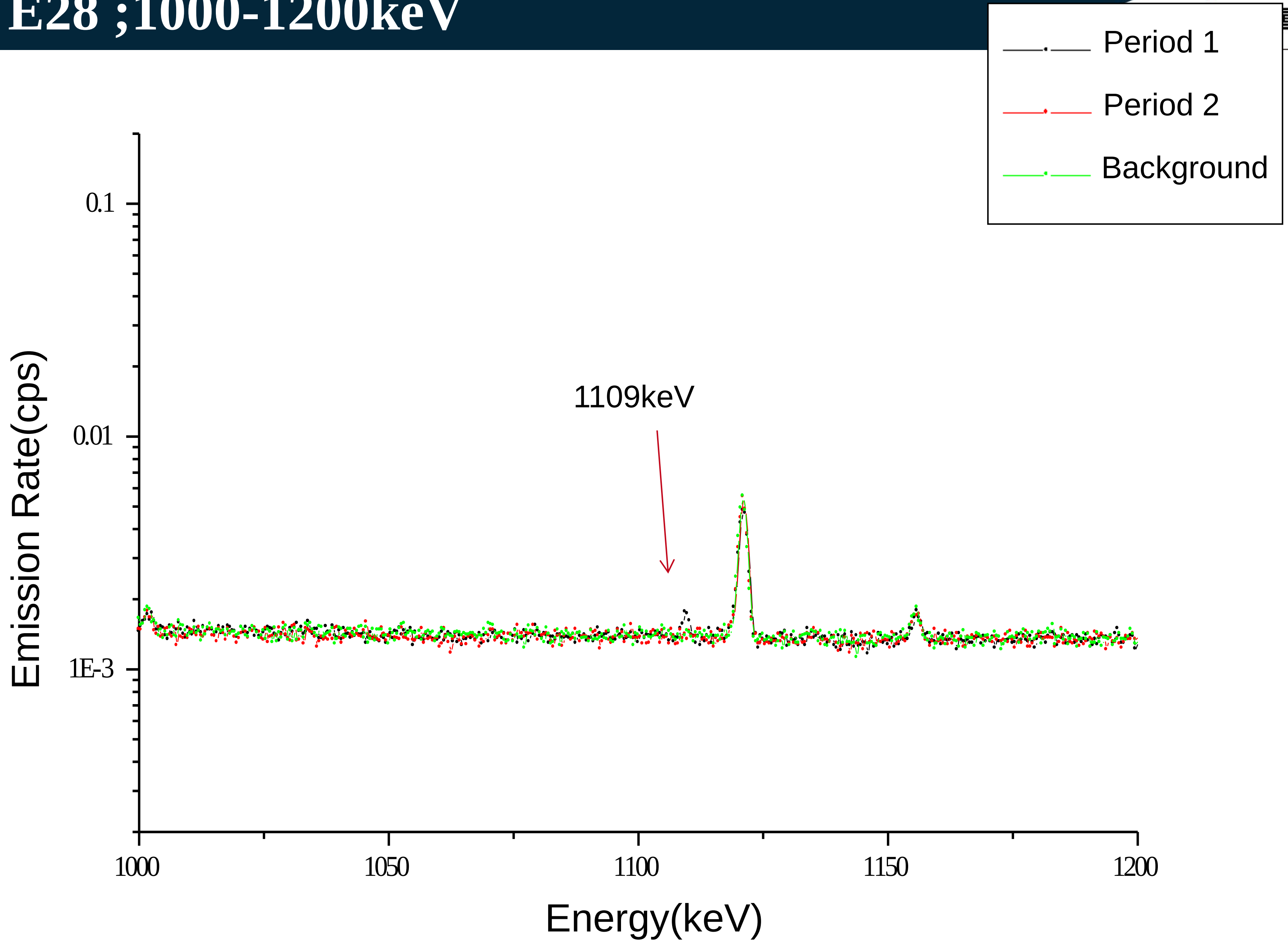
E28 ;1700-1850keV



E28 ;500-650keV



E28 ;1000-1200keV



Discussion on emitted γ-ray during E28 period1

三菱重工

Detected γ-ray energy

Energy(keV)	cps
1445	3.50E-03
1109	1.00E-03
1745	3.00E-04
507.4	5.00E-04
578.9	1.00E-04
605	5.00E-04

Unstable nuclei that emit γ-ray ranging from 1444.5 to 1445.5keV

$E_\gamma(\Delta E)$	$I_\gamma(\Delta I)$	Decay Parent	Associated γ-rays: $E_\gamma(I_\gamma)$
1444.5 *5*		^{144}Cs(1.01 s)	199.326(†100.0), 639.00(†21.2), 758.96(†20.6)
1444.8 *14*	0.13 *4*	^{170}Ta(6.76 m)	100.8(21.0), 221.2(15.7), 860.4(7.39)
1444.86 *16*	†1.3 *4*	^{189}Hg(7.6 m)	320.99(†100), 78.21(†63), 565.42(†48)
1444.90 *17*	0.258 *17*	^{138}I(6.49 s)	588.825(56), 875.23(9.2), 2262.19(3.86)
1444.9 *3*	0.0027 *13*	^{183}Os(13.0 h)	381.768(89.6), 114.463(20.63), 167.844(8.81)
1444.91 *22*	0.25 *3*	^{167}Lu(51.5 m)	29.66(14.4), 239.22(8.6), 213.19(3.6)
1445.0 *1*	0.207 *16*	^{107}Ru(3.75 m)	194.05(9.9), 847.93(5.3), 462.61(3.66)
1445	†2.6	^{107}Sn(2.90 m)	1129.2(†100), 678.5(†100), 1540.6(†30)
1445.0 *2*	0.89 *8*	^{130}La(8.7 m)	357.4(81.0), 550.7(25.9), 908.0(17.0)
1445.04 *25*	0.97 *19*	^{138}Cs(33.41 m)	1435.795(76.3), 462.796(30.7), 1009.78(29.8)
• 1445.058 *39*	0.33 *4*	^{124}Sb(60.20 d)	602.730(97.8), 1690.980(47.3), 722.786(10.76)
• 1445.058 *39*	0.033 *11*	^{124}I(4.18 d)	602.730(60), 1690.980(10.41), 722.786(9.98)
1445.1 *3*	†2.40 *24*	^{120}Cs(64 s)	322.4(†100), 473.5(†30), 553.4(†19.1)
• 1445.10 *30*	0.0358 *18*	^{170}Lu(2.00 d)	84.2551(4.256), 1280.25(3.450), 2041.88(1.434)
• 1445.2 *2*	0.376 *16*	^{146}Eu(4.59 d)	747.2(98), 633.03(43), 634.07(37)
1445.2 *1*	0.087 *16*	^{204}Bi(11.22 h)	899.15(98), 374.72(82), 984.02(59)
1445.3 *1*	0.380 *10*	^{240}Np(7.22 m)	554.60(20.9), 597.40(11.7), 1496.9(1.33)
1445.4 *2*	0.055 *4*	^{151}Nd(12.44 m)	116.80(43.4), 255.68(16.4), 1180.89(14.8)
1445.4 *1*	0.32 *3*	^{234}Pa(6.70 h)	131.30(18), 946.00(13.4), 883.24(9.6)
1445.45 *26*	†0.55 *6*	^{71}Se(4.74 m)	147.50(†211), 1095.26(†43.6), 830.33(†43.2)
1445.5 *3*	3.2 *7*	^{102}Sr(69 ms)	243.80(53), 150.15(18.0), 93.89(13.4)
1445.5 *5*	0.14	^{142}La(91.1 m)	641.285(47), 2397.8(13.3), 2542.7(10.00)

We have not succeed to find a nucleus fit for the observed γ-ray energies.

- **γ-rays from unstable nuclei**
- **γ-rays from excited nuclei**
- **Thermal neutron capture γ-rays**

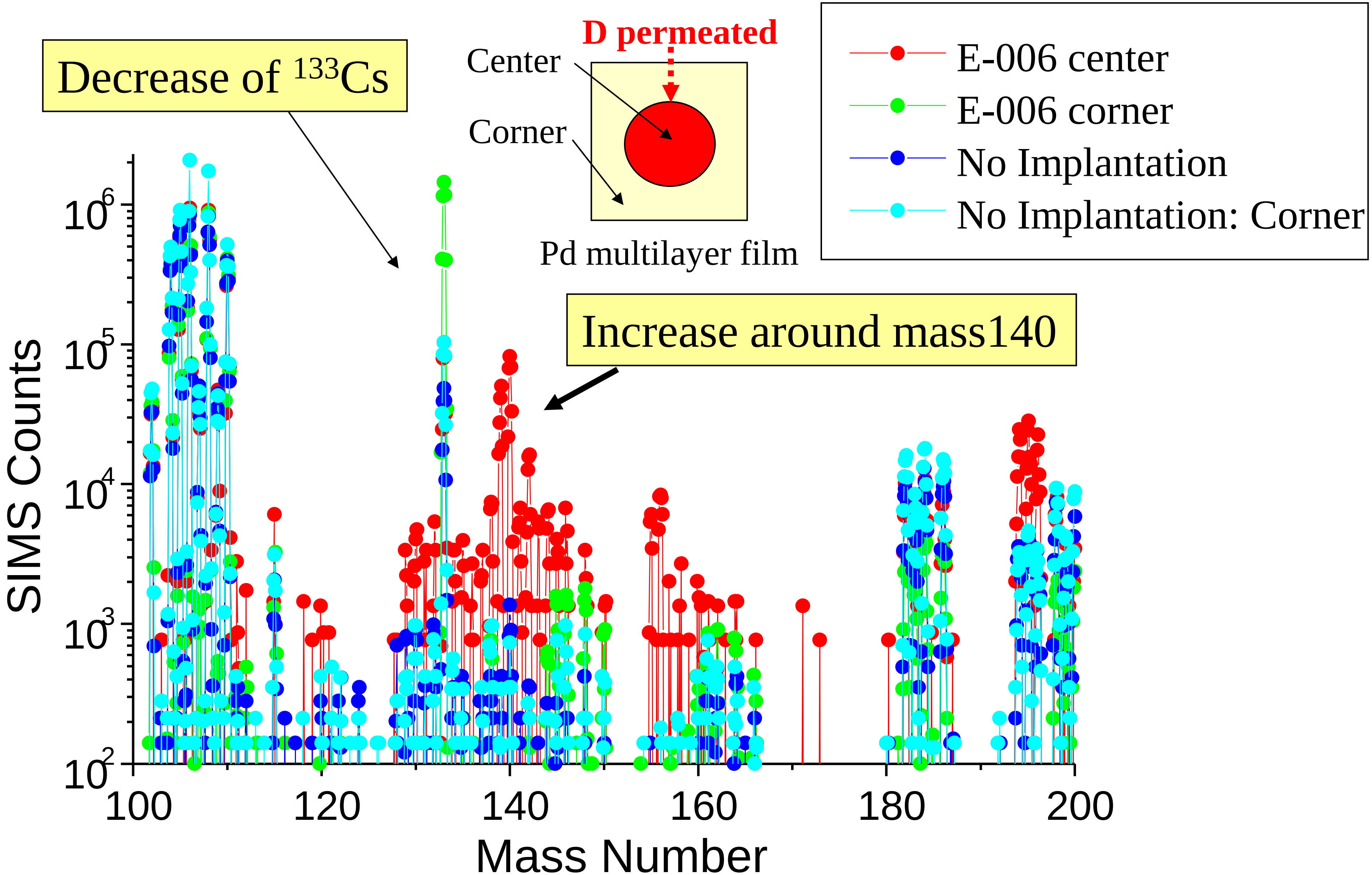
Further Study!

1)Replication experiments

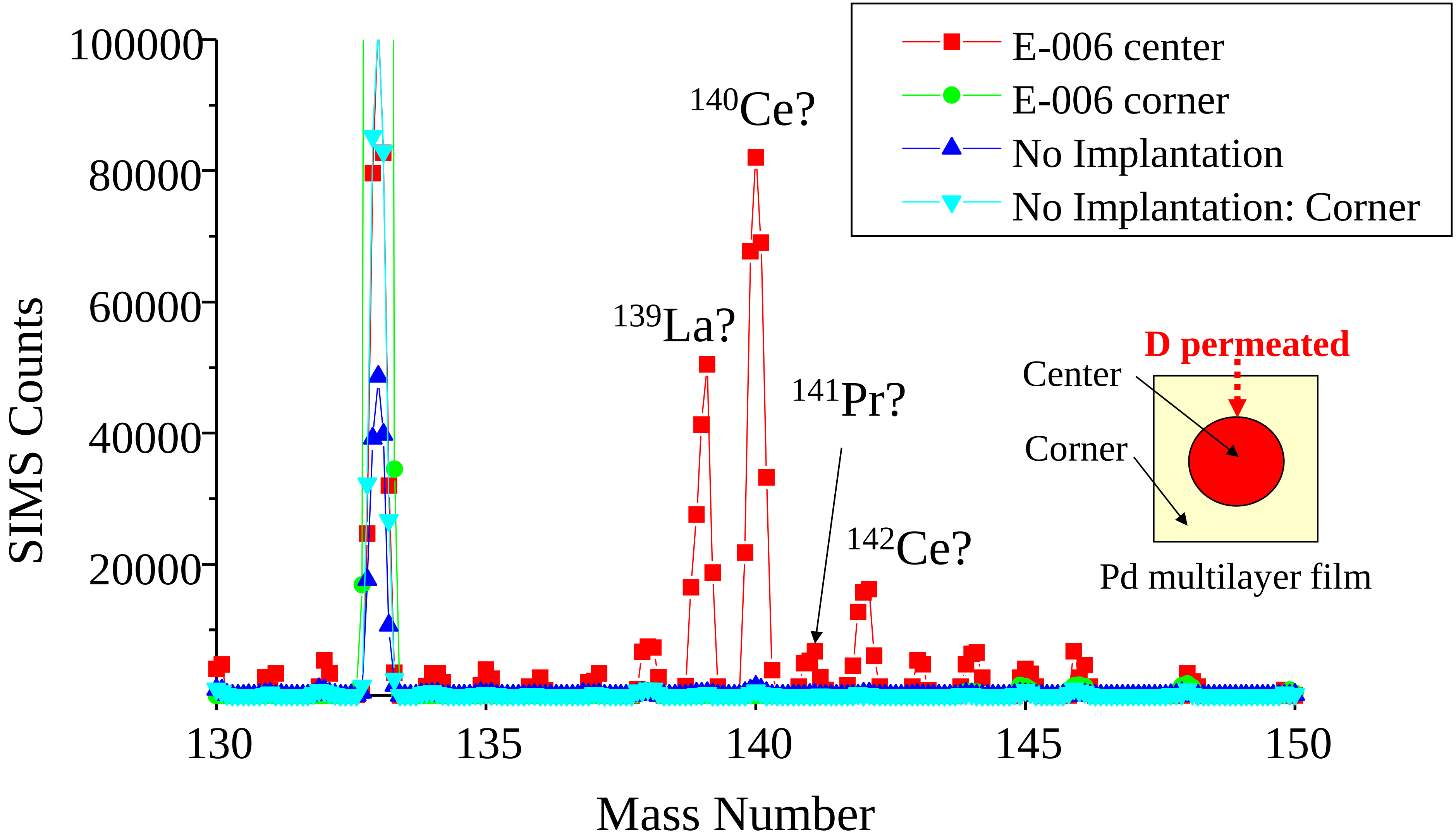
2)Build a physical model

4. Analysis using ICP-MS, SIMS and XPS

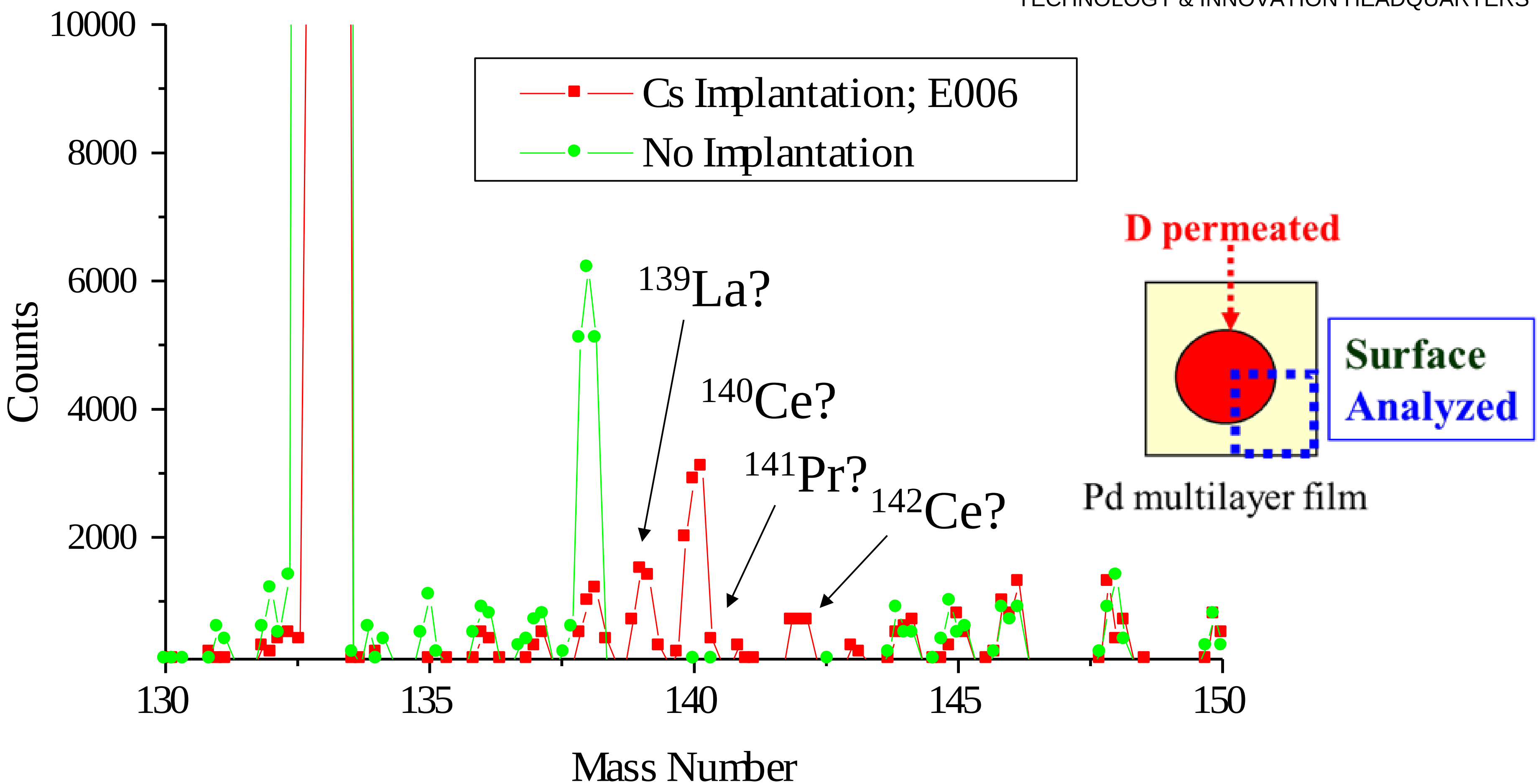
SIMS Analysis; E006 Wide Spectra



SIMS Analysis; E006



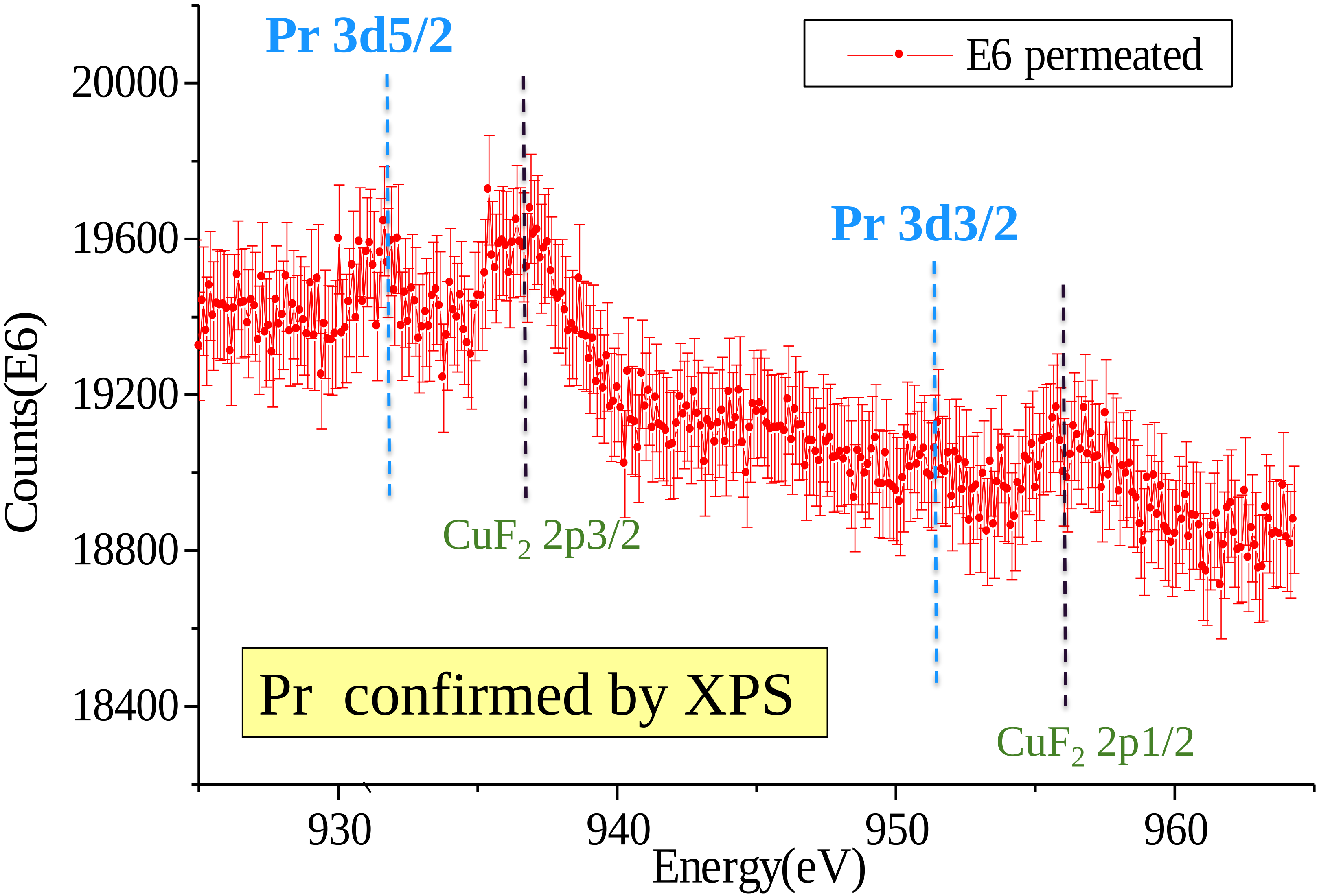
ICP-MS Analysis; E006



SIMS (point) and ICP-MS (all surface) gave similar results

Different Tendency from D_2 gas permeation

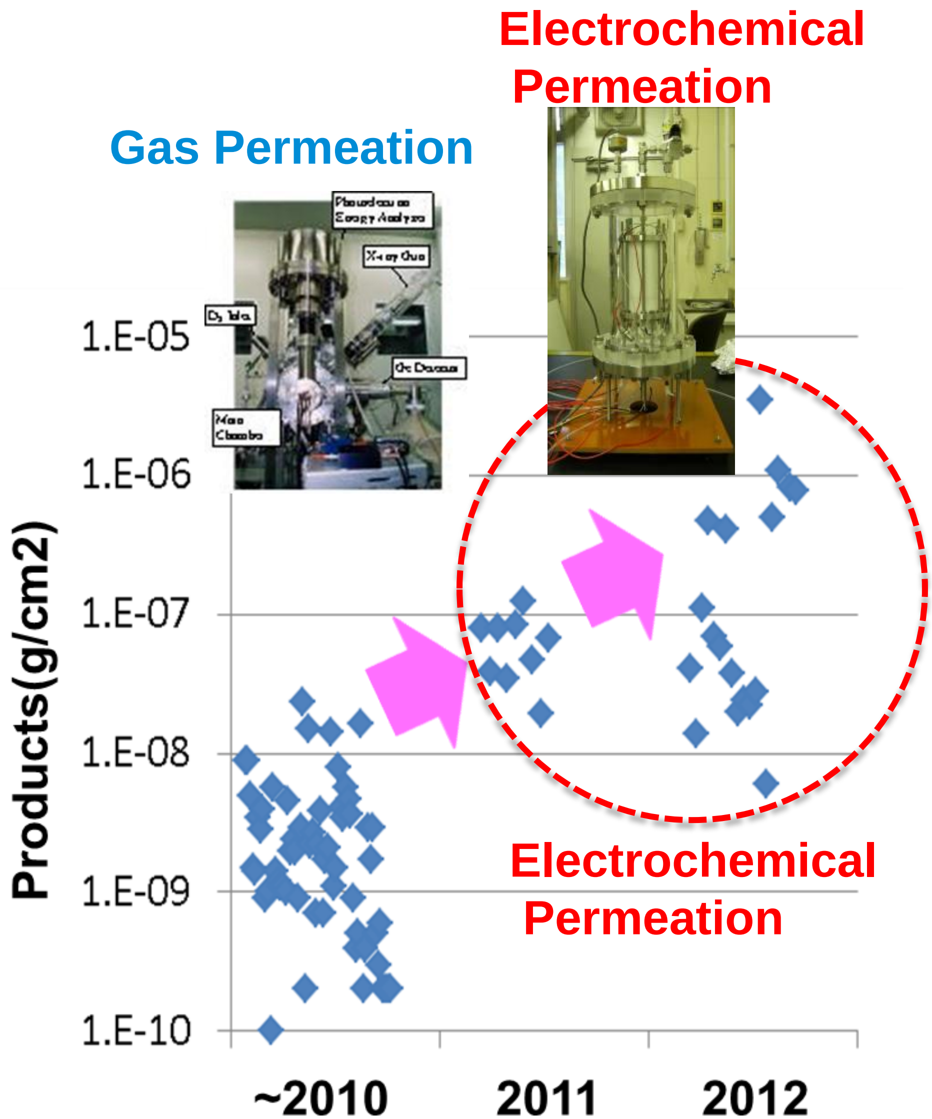
Confirmation of the products by XPS



Increase of Products by Electrochemical Permeation

Applied an electrochemical method to increase deuteron density near the surface of the Pd multilayer film

Transmutation products;
Increased
Gamma-rays
Occasionally detected



Concluding Remarks

1. **Low energy nuclear transmutations from Cs into Pr, Sr into Mo, Ba into Sm and Ca into Ti have been observed in the Pd complexes, which are composed of Pd and CaO thin film and Pd substrate, induced by D_2 gas permeation.**

2. **An electrochemical method was applied to increase the local deuteron density near the surface of the nano-structured Pd multilayer film. Transmutation products were increased up to ~1μg/cm^2 by this approach.**

3. **Statistically significant γ-rays which have clear energy spectra were detected. These emissions were supposed to be caused by the increase of transmutation products. At present, we have limited examples. Further study is necessary.**

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Independently Replicated Transmutation Experiments of Cs into Pr Presented at ICCF17, Aug.12-17, 2012, Deajon, Korea.

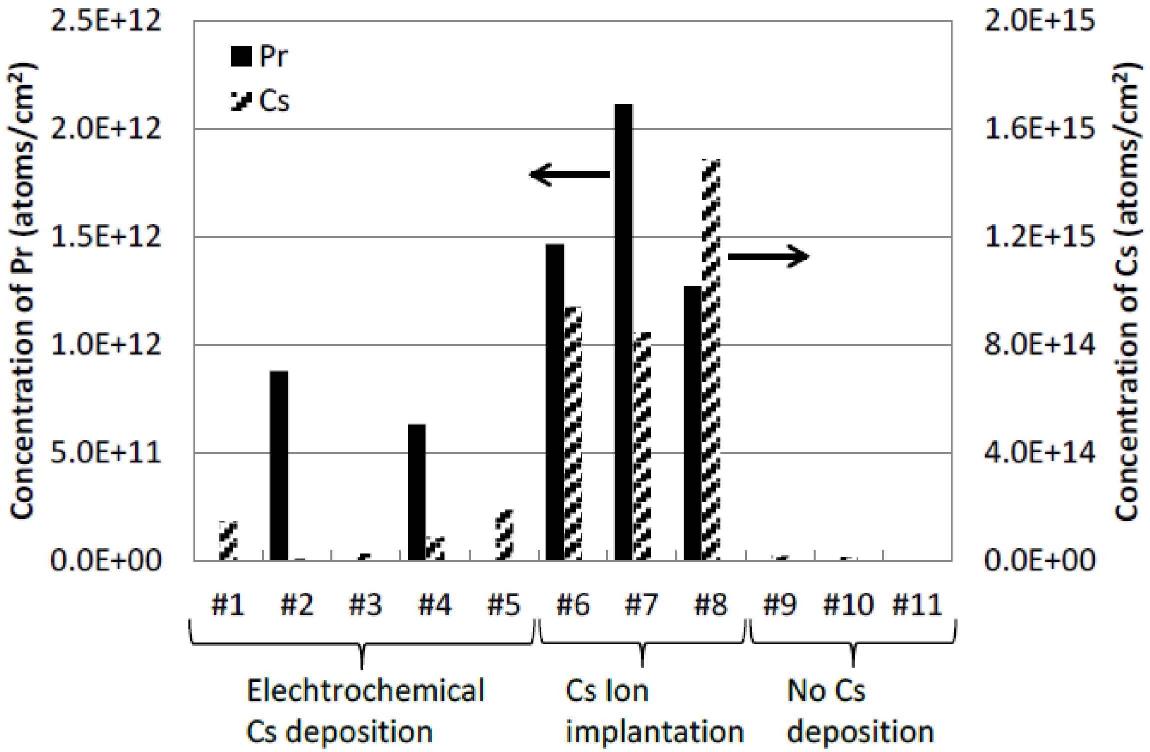
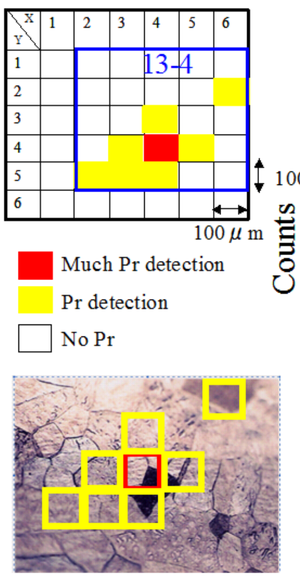


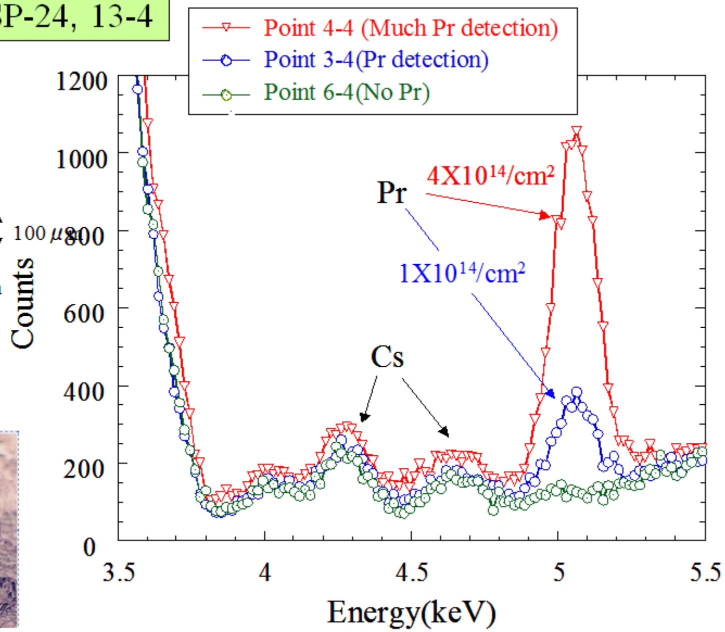
Fig. 4 The detected amounts of Pr and Cs in the samples with D_2 permeation treatments.

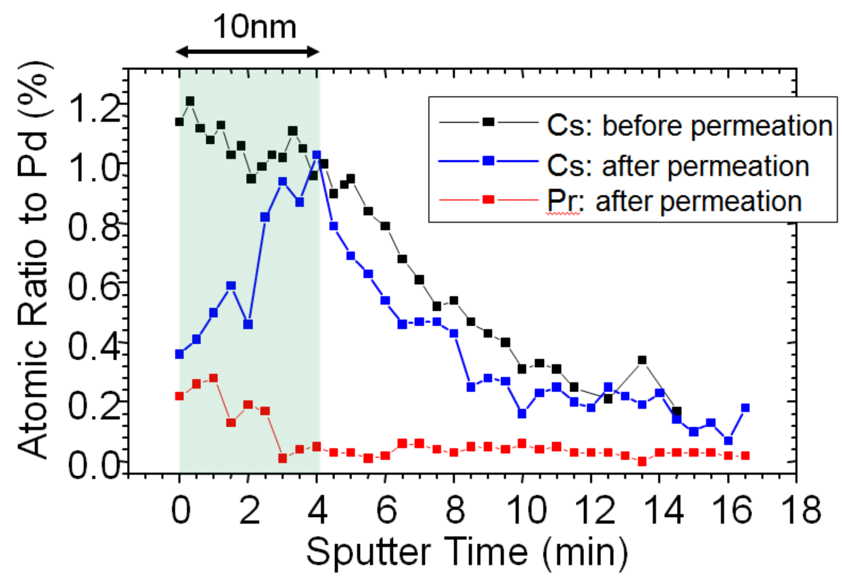
Naoko Takahashi et.al, "Detection of Pr in Cs Ion-Implanted Pd/CaO Multilayer Complexes with and without D2 Gas Permeation", The Preprint of the ICCF-17 Proceedings, August 12~17, 2012 DCC Korea, Daejeon, South Korea

Non Uniformity of Products

100 micron beam;SP-24, 13-4



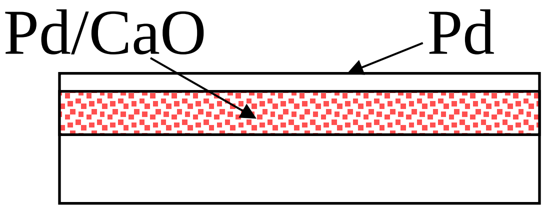




3D Elemental Analysis is Preferable!

Surface and Depth distribution analysis

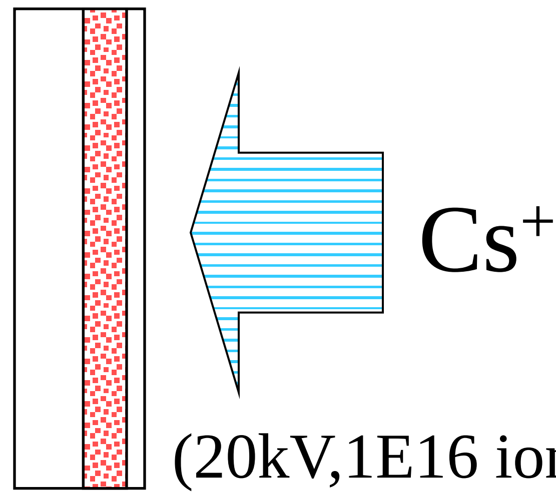
Cs^+ Ion Implantation to Pd/CaO/Pd film



Pd multilayer film

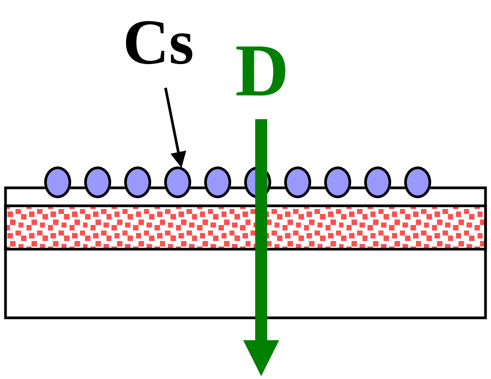
1.Fabrication

2.Cs^+ Ion implantation



(20kV,1E16 ions/cm^2)

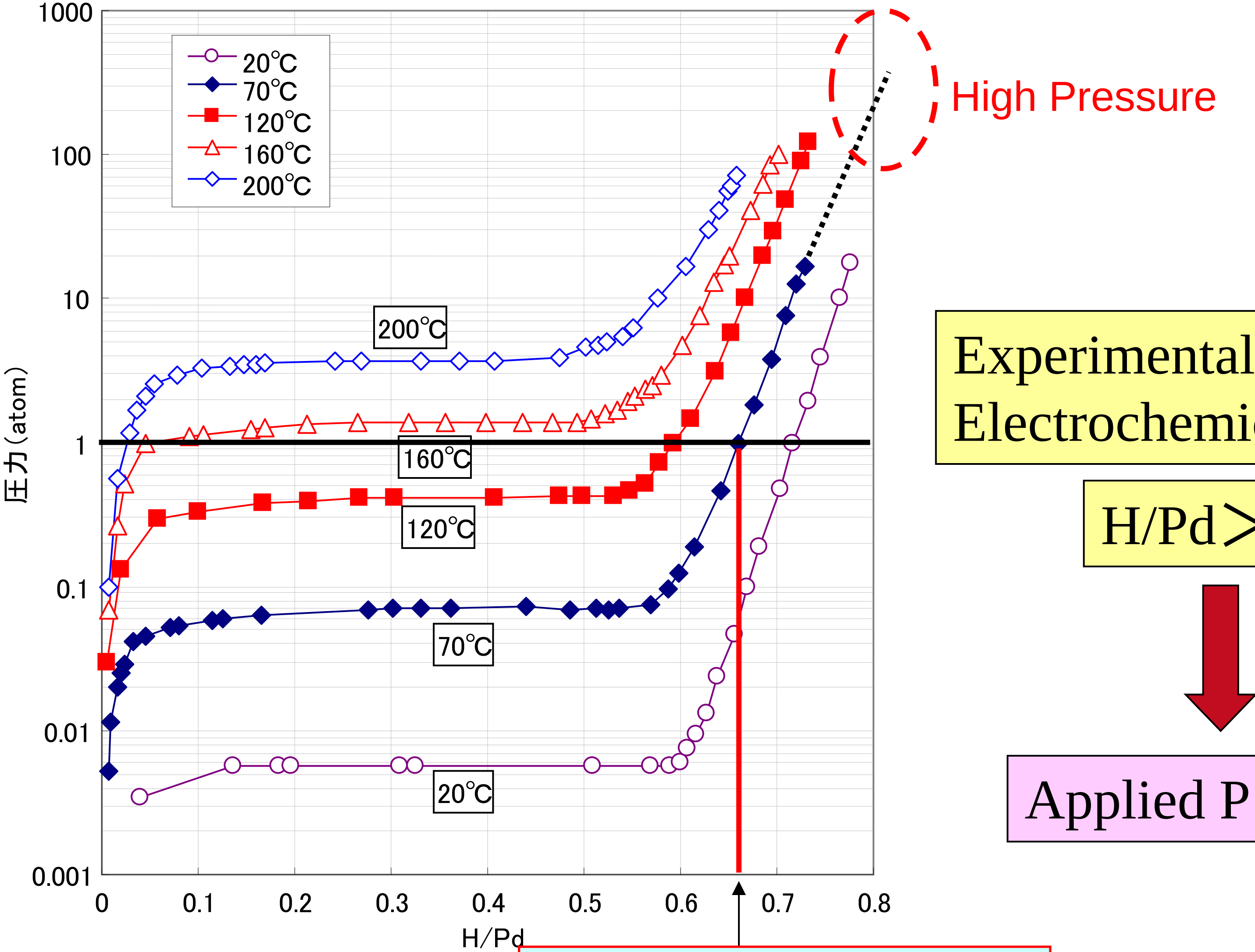
3.D Permeation



1) Give Cs for transmutation

2) Decrease of work function of surface layer

Increase of D Pressure based on Pd-H system



H/Pd=0.66 under atmosphere

Experimental Facts using Electrochemical Method

H/Pd＞0.8

Applied P > 100atm

Consideration on Compound Species

Possible compounds for mass 140

^{138}Ba(71.7%)D	^{133}Cs (100%)^{7}Li (92.4%)	^{110}Pd ^{30}Si(3.1%)
^{106}Pd ^{34}Si(4.3%)	^{109}Ag(48.1%) ^{31}P(100%)	^{104}Pd ^{36}Ar(0.33%)
^{102}Pd ^{38}Ar(0.06%)	^{110}Pd ^{28}Si(92.3%)D	^{108}Pd ^{30}Si(3.1%)D
^{105}Pd ^{33}Si(0.8%)D	^{102}Pd ^{36}Si(0.02%)D	^{102}Pd ^{36}Ar(0.3%)D

Possible compounds for mass 139

^{137}Ba(11.2%)D	^{133}Cs (100%)^{6}Li (7.6%)	^{110}Pd ^{29}Si(4.7%)
^{106}Pd ^{33}Si(0.8%)	^{104}Pd ^{35}Cl(75.8%)	^{102}Pd ^{37}Cl(24.2%)
^{110}Pd ^{27}Al(100%)D	^{106}Pd ^{31}P(100%)D	^{105}Pd ^{32}S (94.9%)D
^{104}Pd ^{33}Si(0.8%)D	^{105}Pd ^{32}Si(94.9%)D	^{102}Pd ^{35}Cl(75.8%)D

Not explained consistently by these compounds

Gamma-ray Estimation

$$n = \left(\frac{N_c}{t_c} - \frac{N_b}{t_b} \right) \pm \left(\frac{\sqrt{N_c}}{t_c} + \frac{\sqrt{N_b}}{t_b} \right)$$

n; γ - ray count rate(cps)

N_c; γ - ray counts

t_c; time for γ - ray measuremnt (sec)

N_b; Backgound γ - ray counts

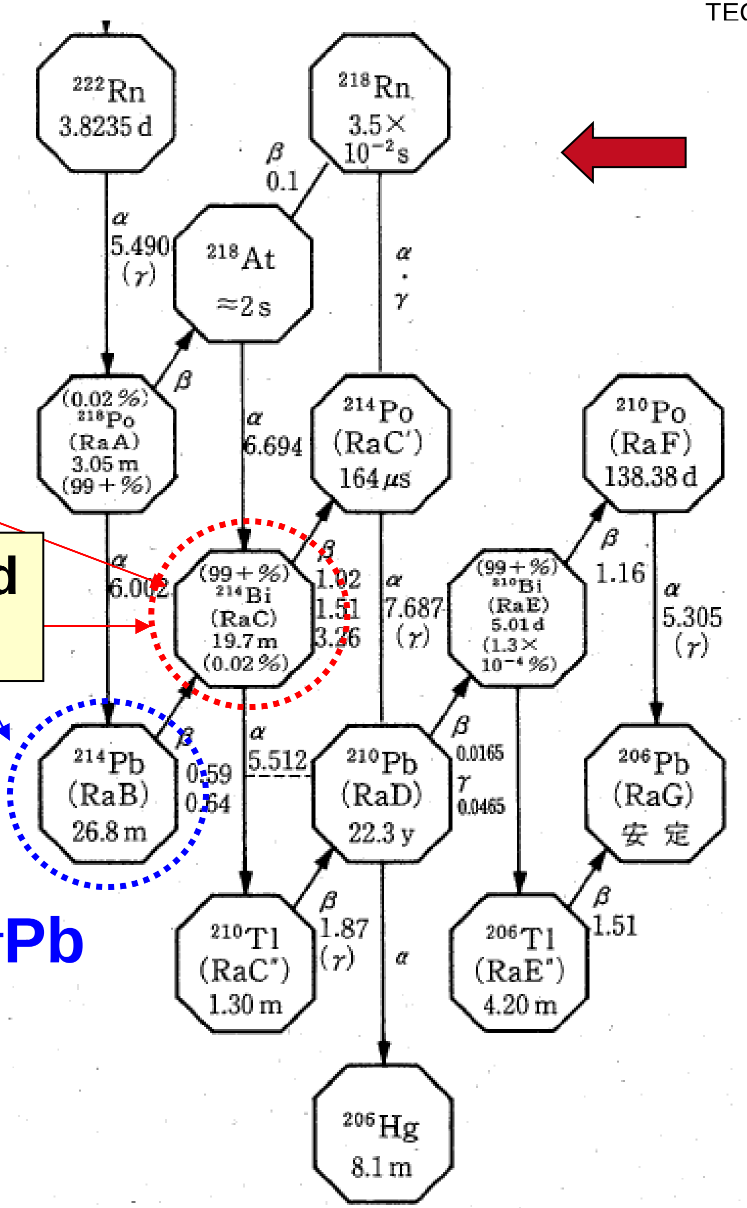
t_b; time for backgound γ - ray measuremnt (sec)

Uranium Series (4n+2)

Start from Uranium-238

^{214}Bi

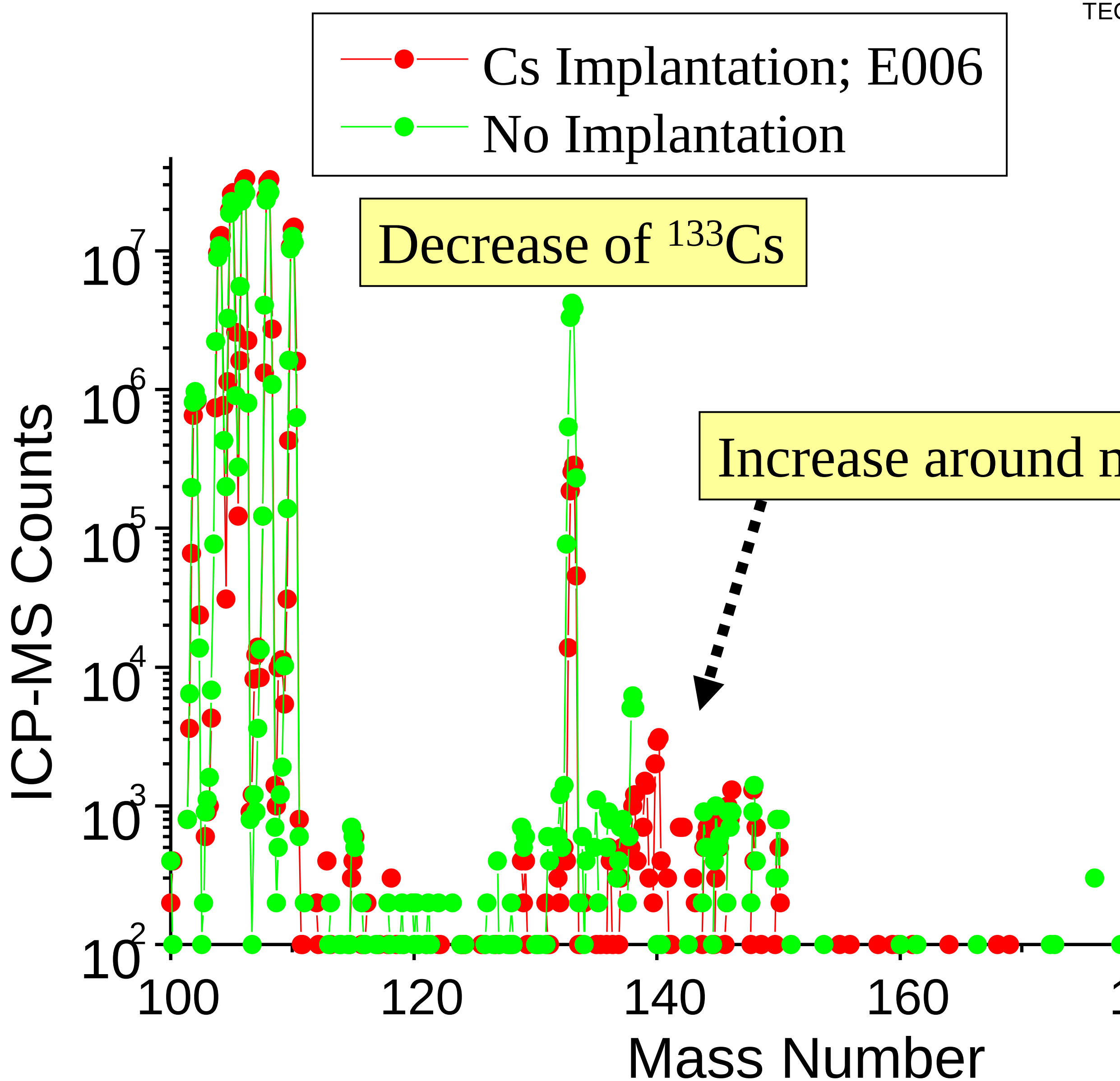
Seems to be detected in E16 period 1

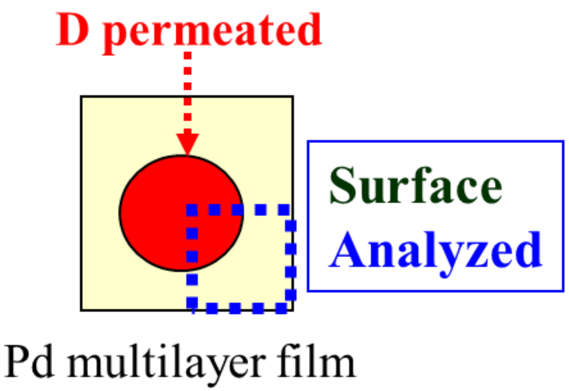


^{214}Pb

ICP-MS Analysis; E006 Wide Spectra

TECHNOLOGY & INNOVATION HEADQUARTERS





Effect of Intermediate Layer

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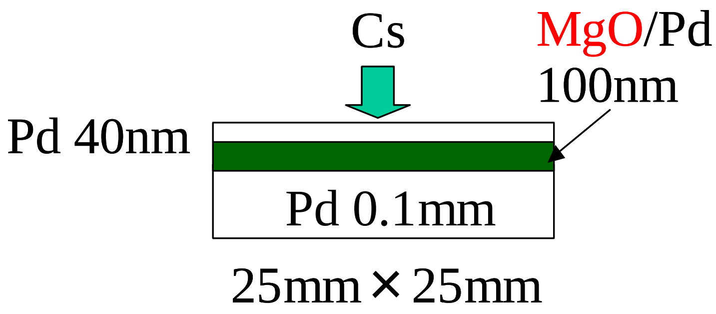
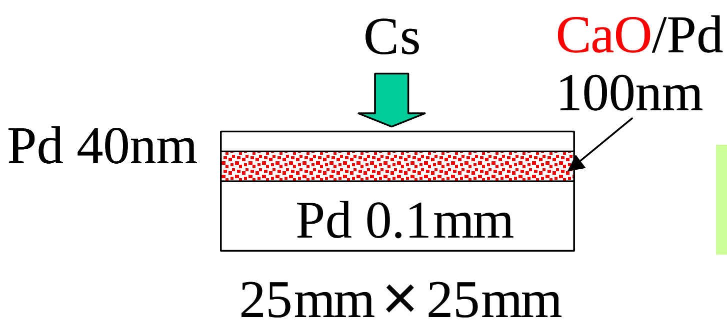


Work Function

MgO;3.3eV → NO Pr detected

3 cases



CaO;1.6eV → Pr detected !

Many cases

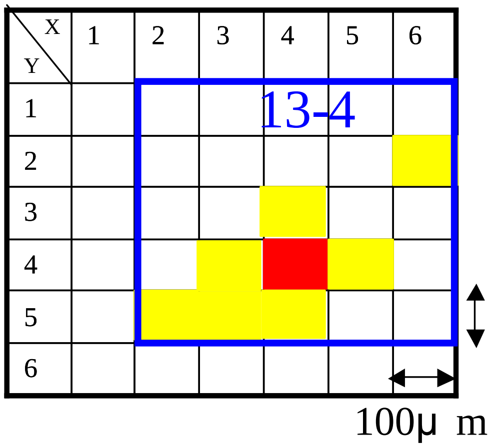
Y_2O_3;2.2eV → Pr detected !

Many cases

Work function of the intermediate layer seems to be important. → ***Assumption* :Electron rich state is important**

Detection of Localized Pr

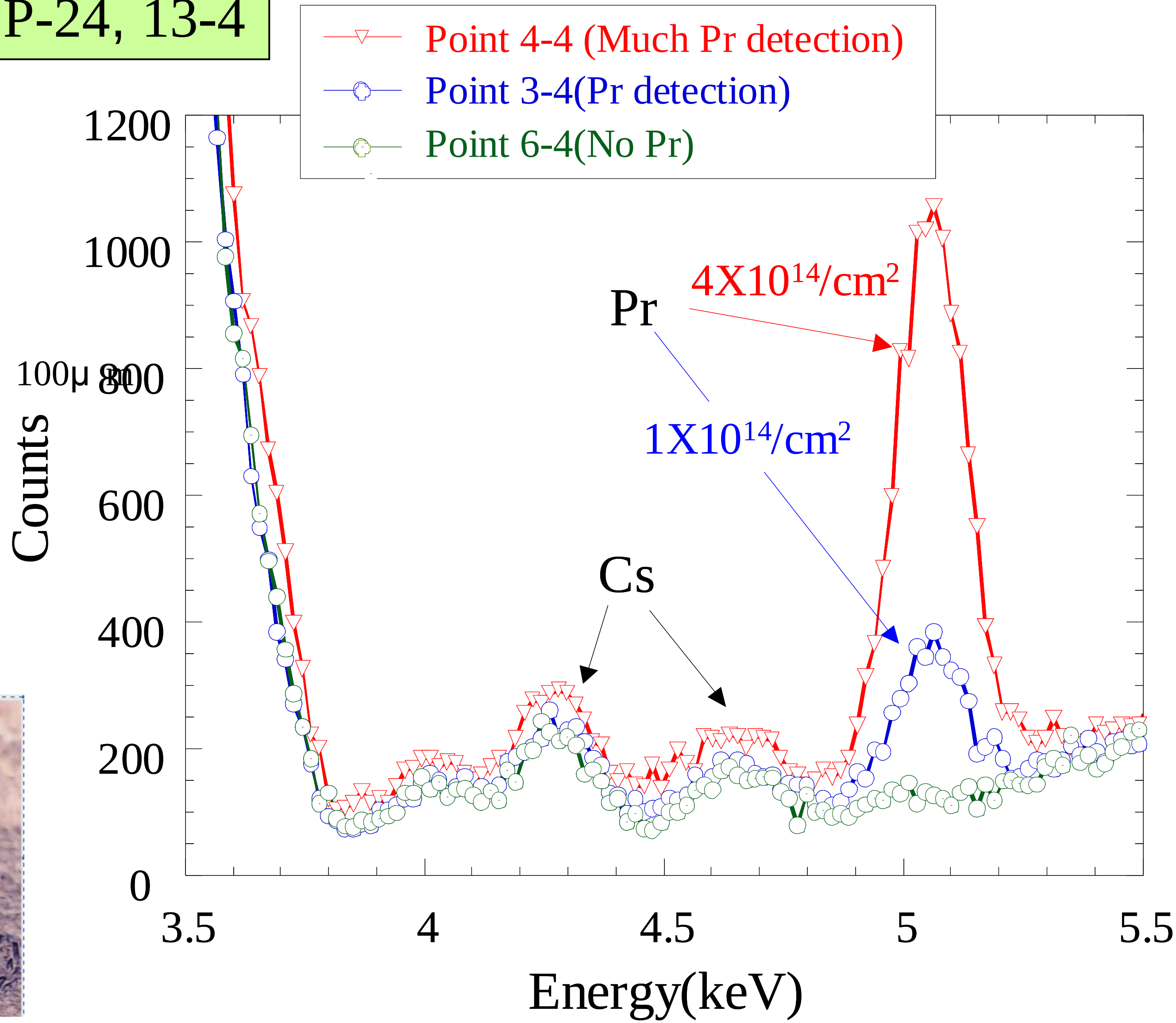
100 micron beam;SP-24, 13-4



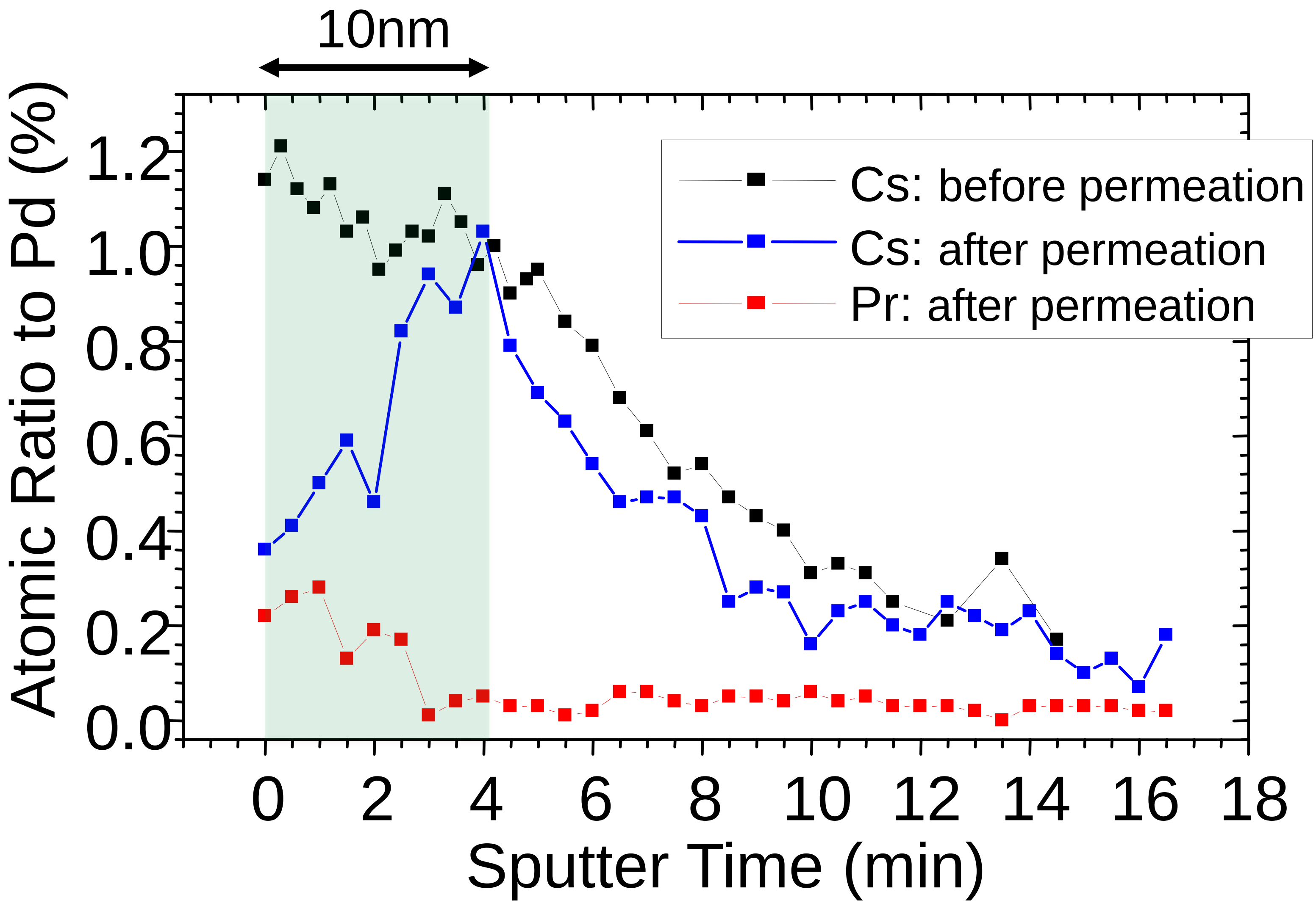
Much Pr detection

Pr detection

No Pr



Depth Profile of Cs and Pr by XPS



Fabrication of Nano-structured Pd Multilayer Film

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Washing a Palladium Sample with Acetone

↓

900°C 10H Annealing under Vacuum Condition (< 10^{-6} Torr)

↓

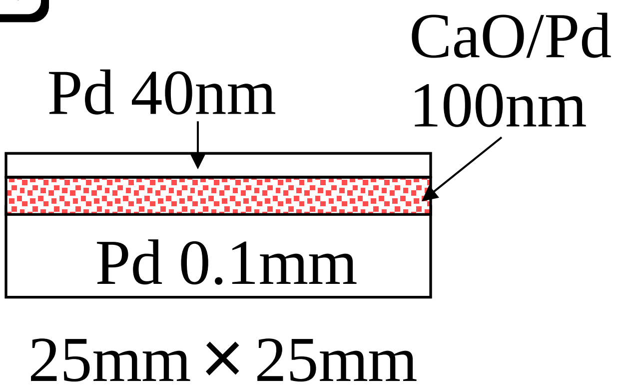
Washing the Sample with Aqua Regia (100sec)

↓

5 times Alternatingly Sputtering of CaO (2nm) and Pd (18nm)

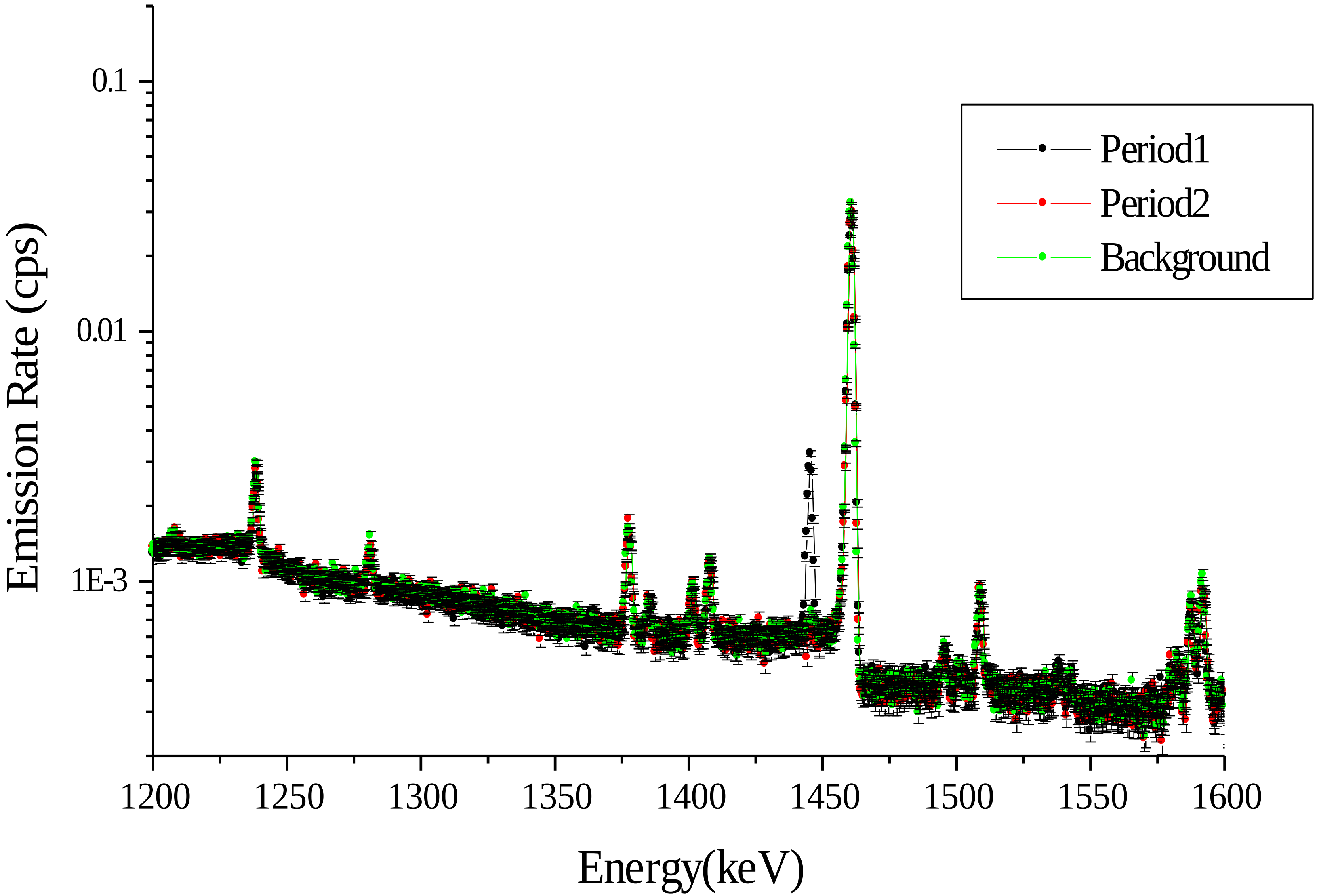
↓

Ion Beam Sputtering of Pd only (40nm)



E28 ;1200-1600keV with sigma

三菱重工



E28 ;1700-1850keV with sigma