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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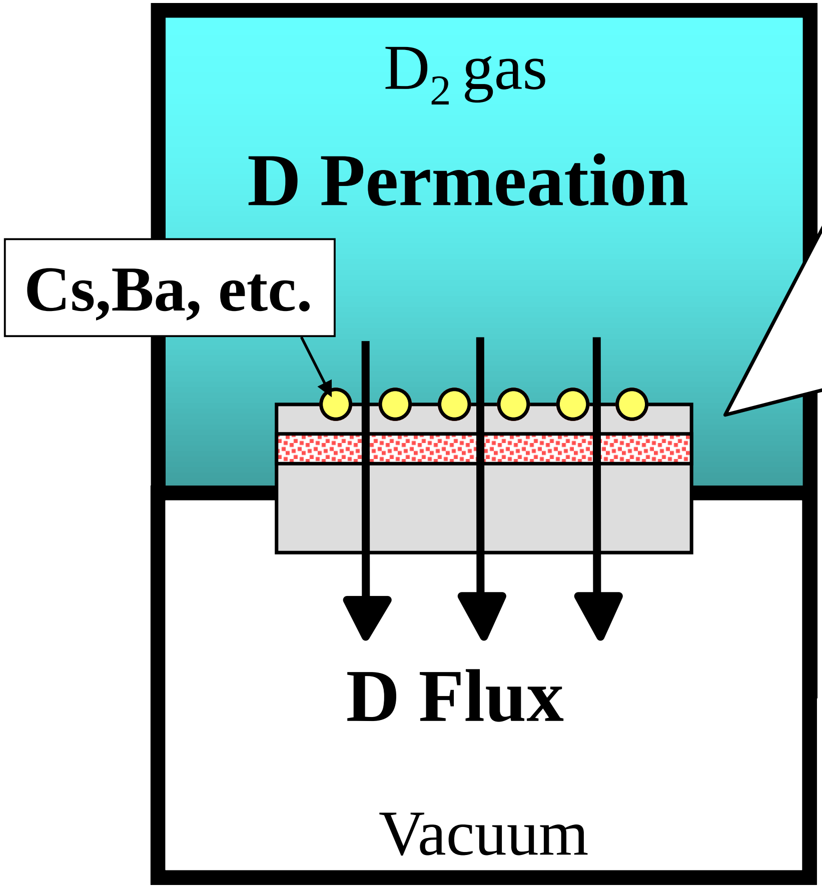
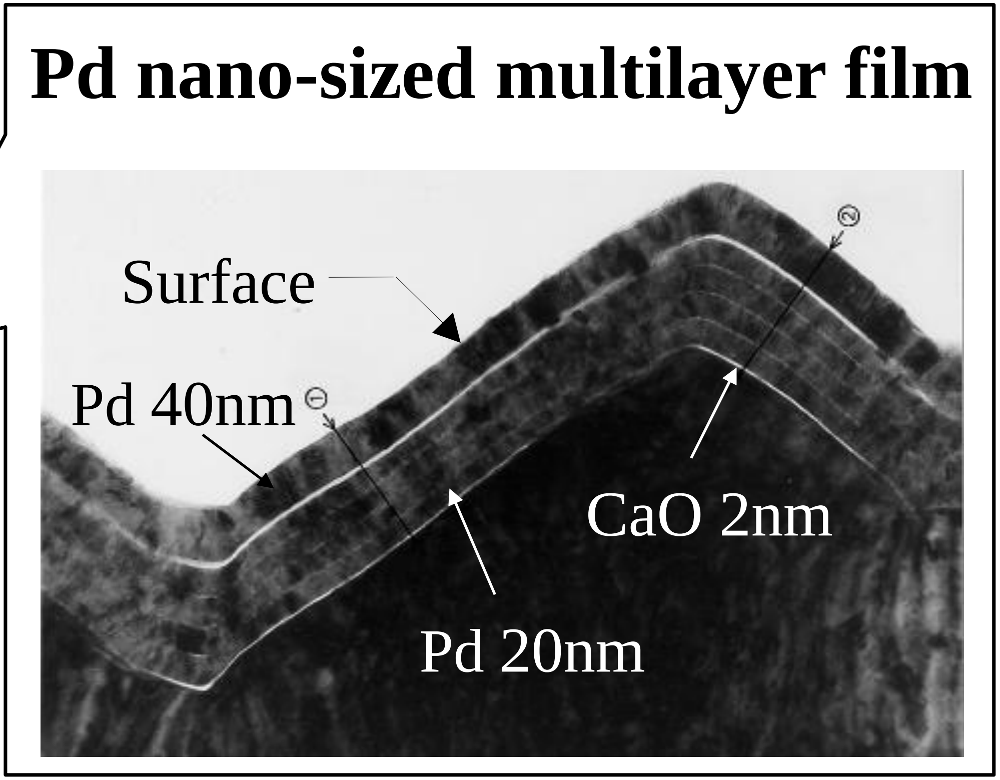
1. Introduction

Features of Our Method

D_2 gas permeation through nano-structured Pd complex





Reactions observed in the case of Gas Permeation

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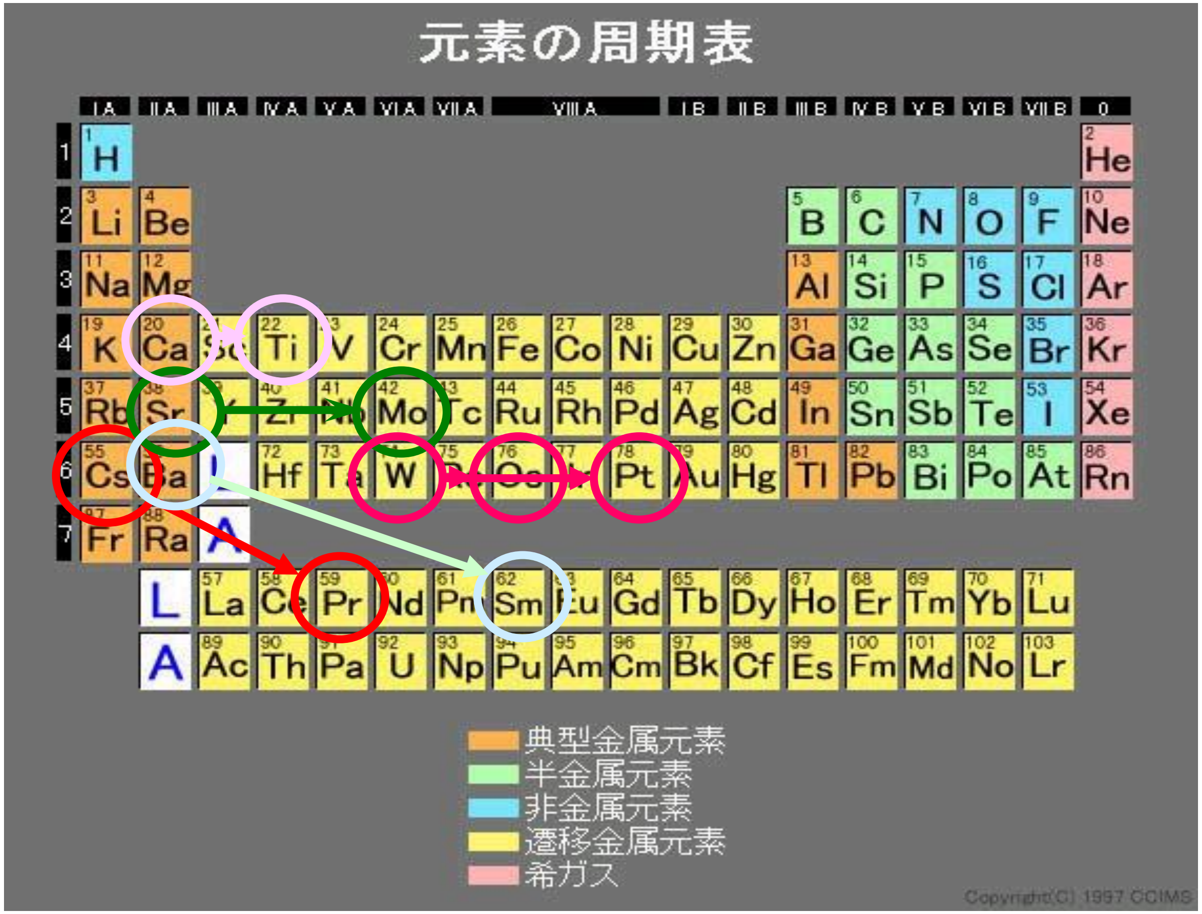


1)Alkali metals; Electron Emitter
2)2d, 4d, 6d; α capture reactions

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

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

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

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

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

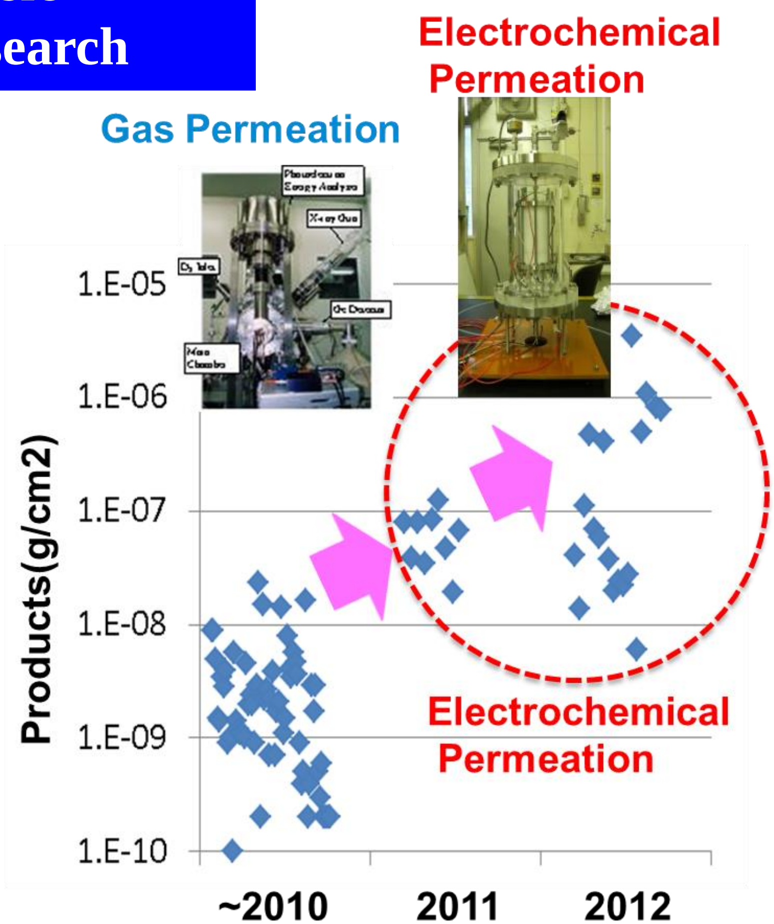
$$^{184}_{74}W \xrightarrow{2d(\alpha)} {}^{188}_{76}Os$$

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

Progress in Permeation Experiments

～2010
Gas Permeation ;Basic Research

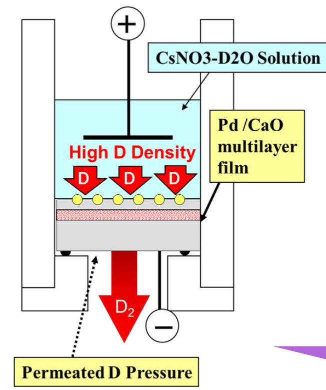
2011～2012
Increase of Products for Future Application



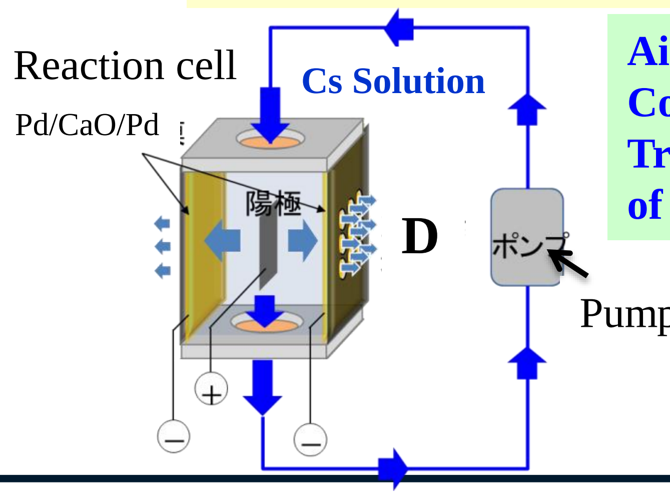
2013～
Consecutive Processing

Batch Processing

Addition of fixed quantity of Cs



Consecutive Processing



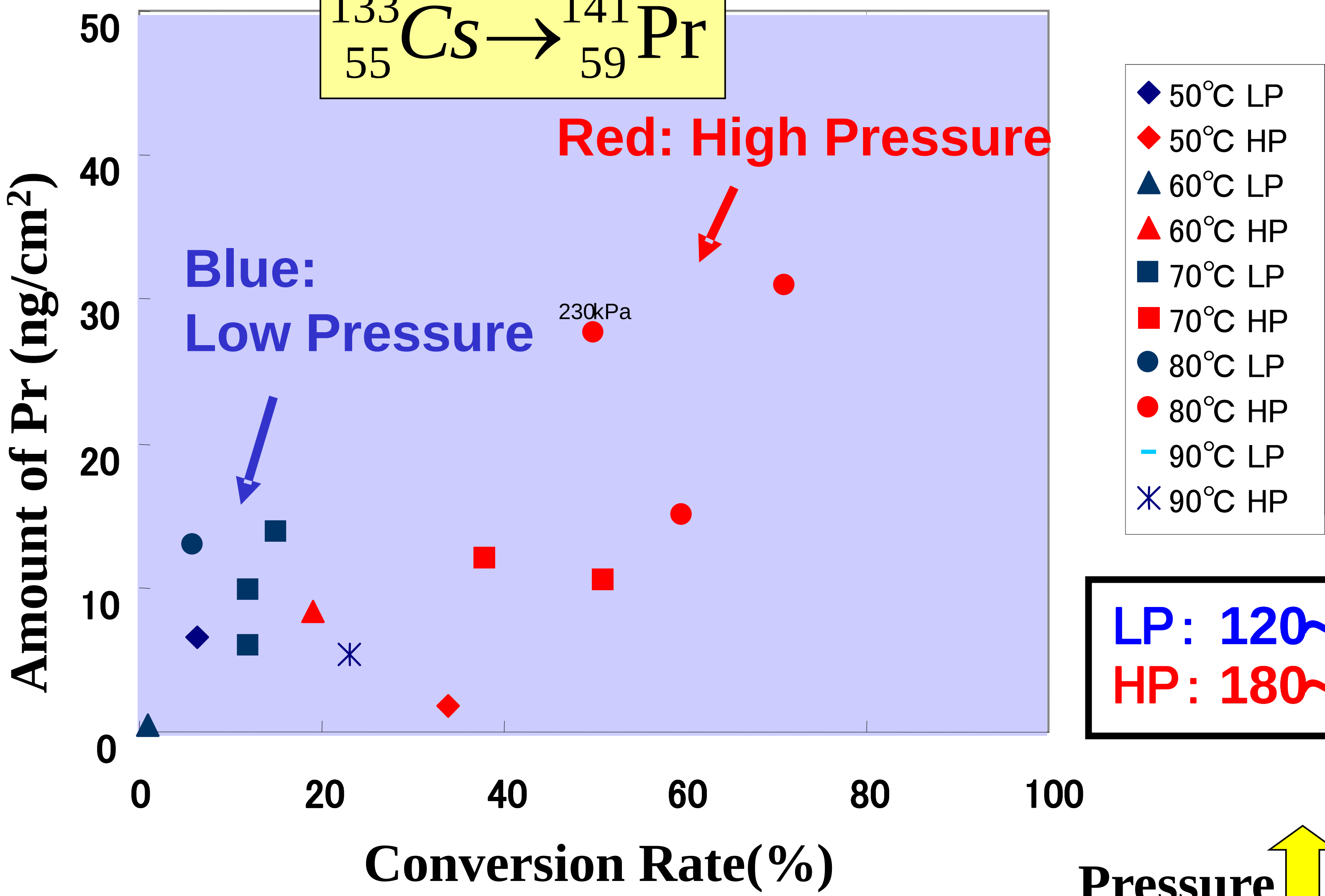
Aiming Consecutive Transmutation of Cs

2. Results of Batch Process Experiments

2-1. Transmuted Products Analyzed by ICP-MS, SIMS and XPS

Pr Dependence on D_2 gas pressure

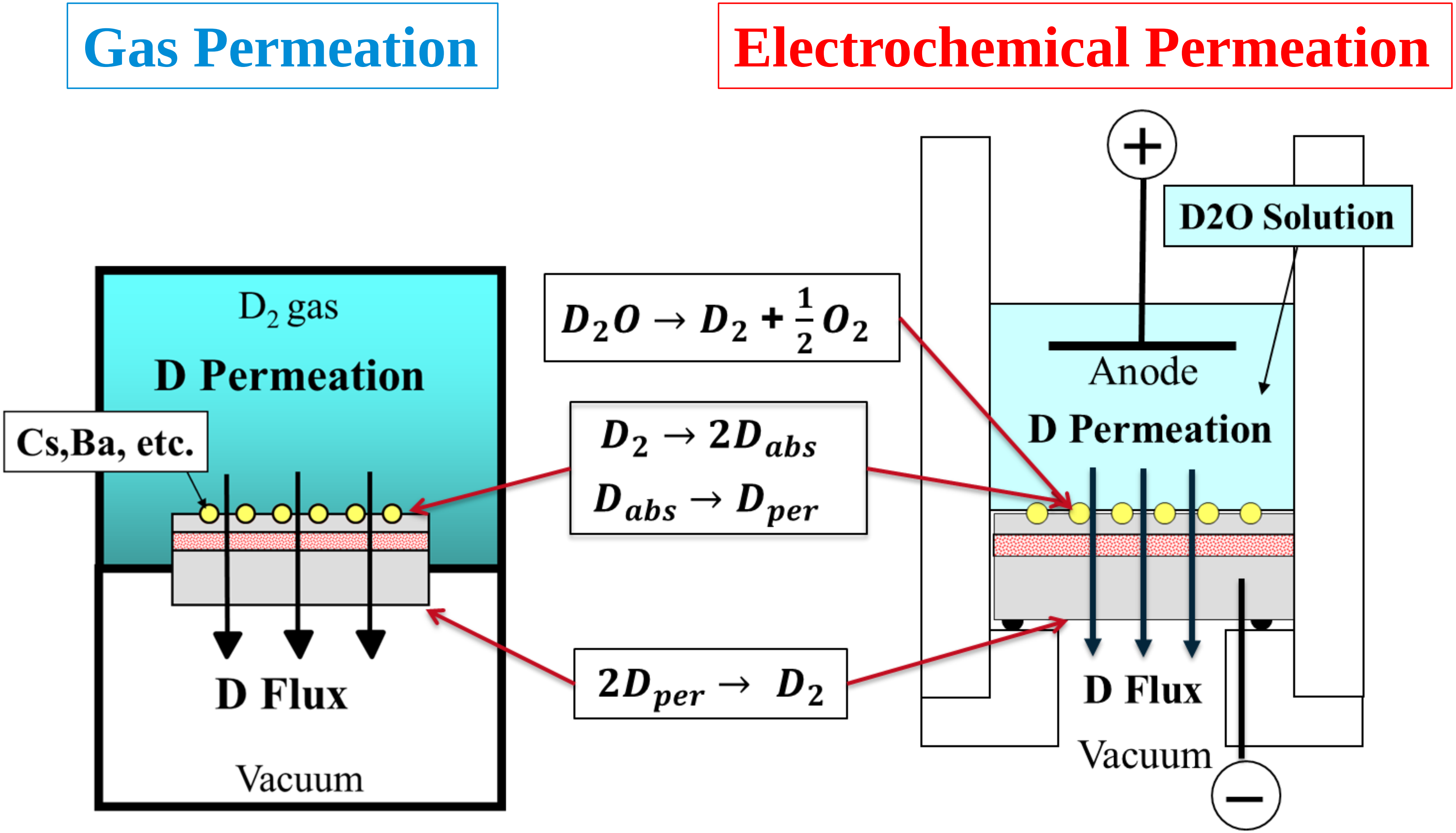
$$ {}^{133}_{55}Cs \rightarrow {}^{141}_{59}\mathrm{Pr} $$



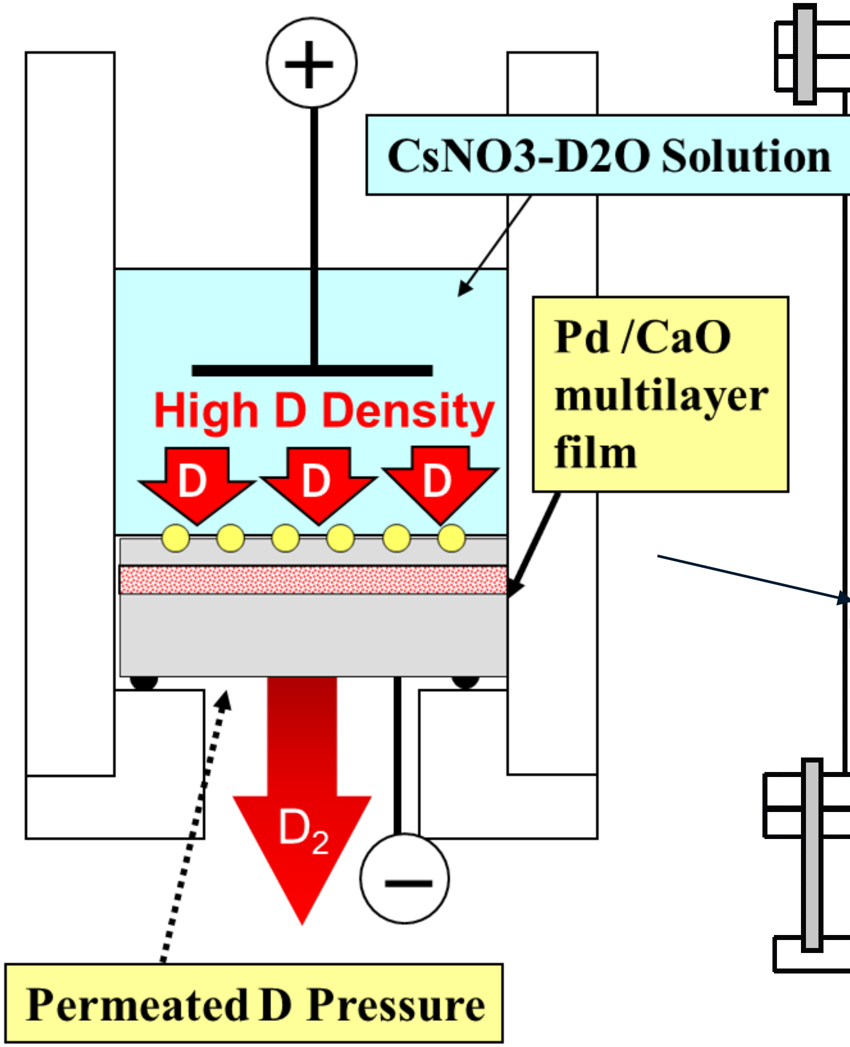
LP : 120～130kPa
HP : 180～230 kPa

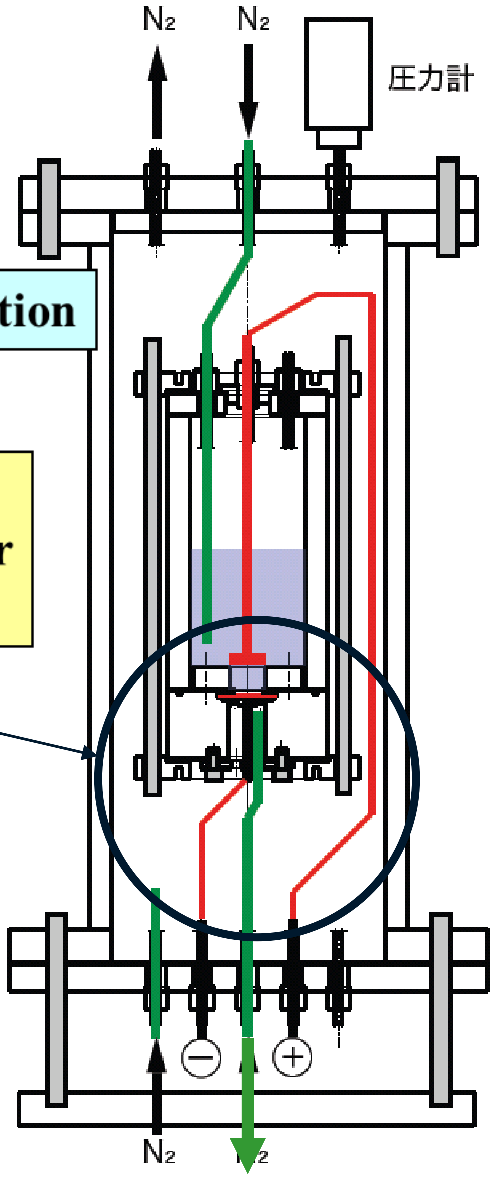
Pressure ↑ → Pr ↑

Gas vs. Electrochemical Permeation

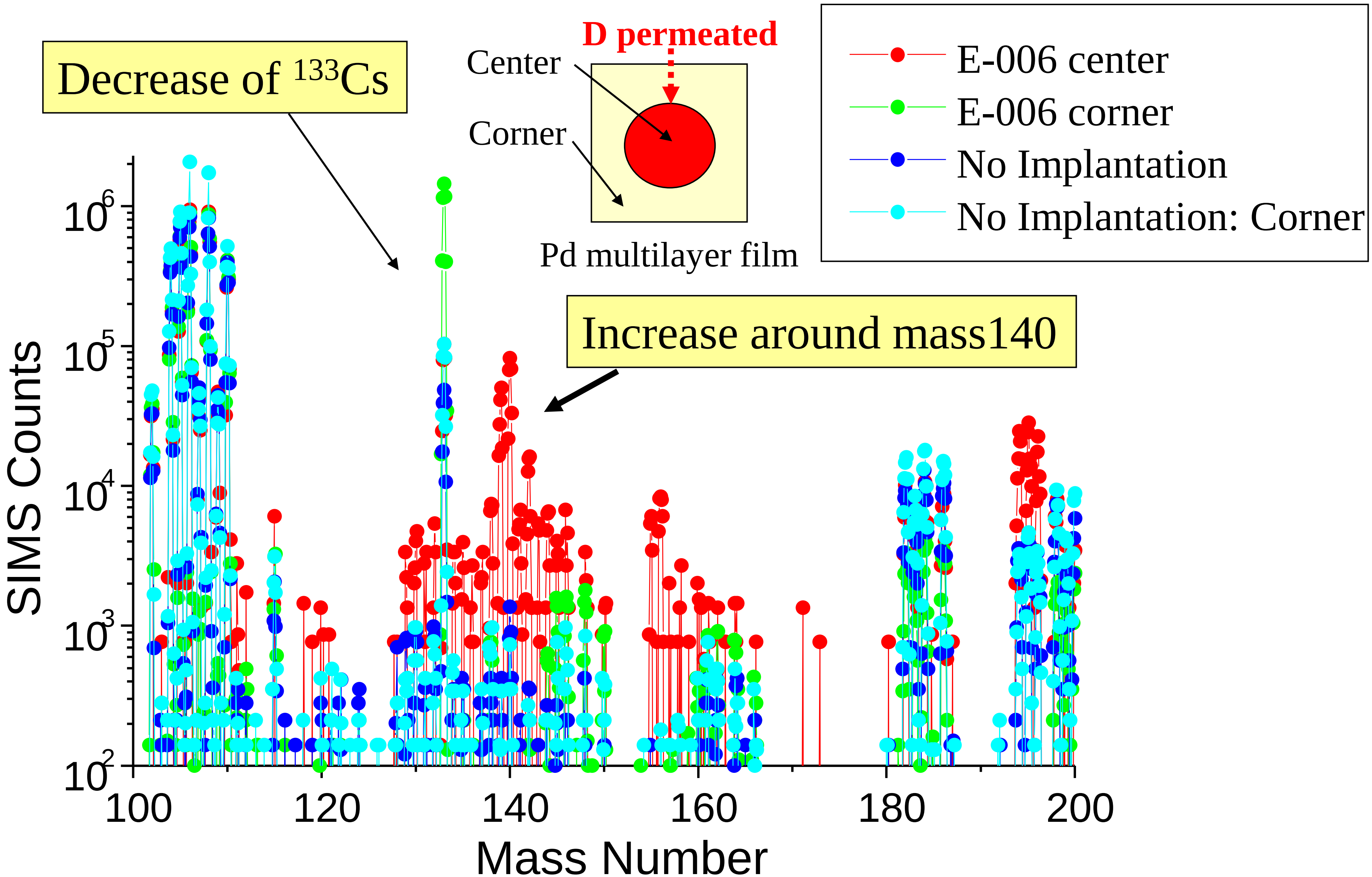


Experimental Apparatus aiming Increase of D Density

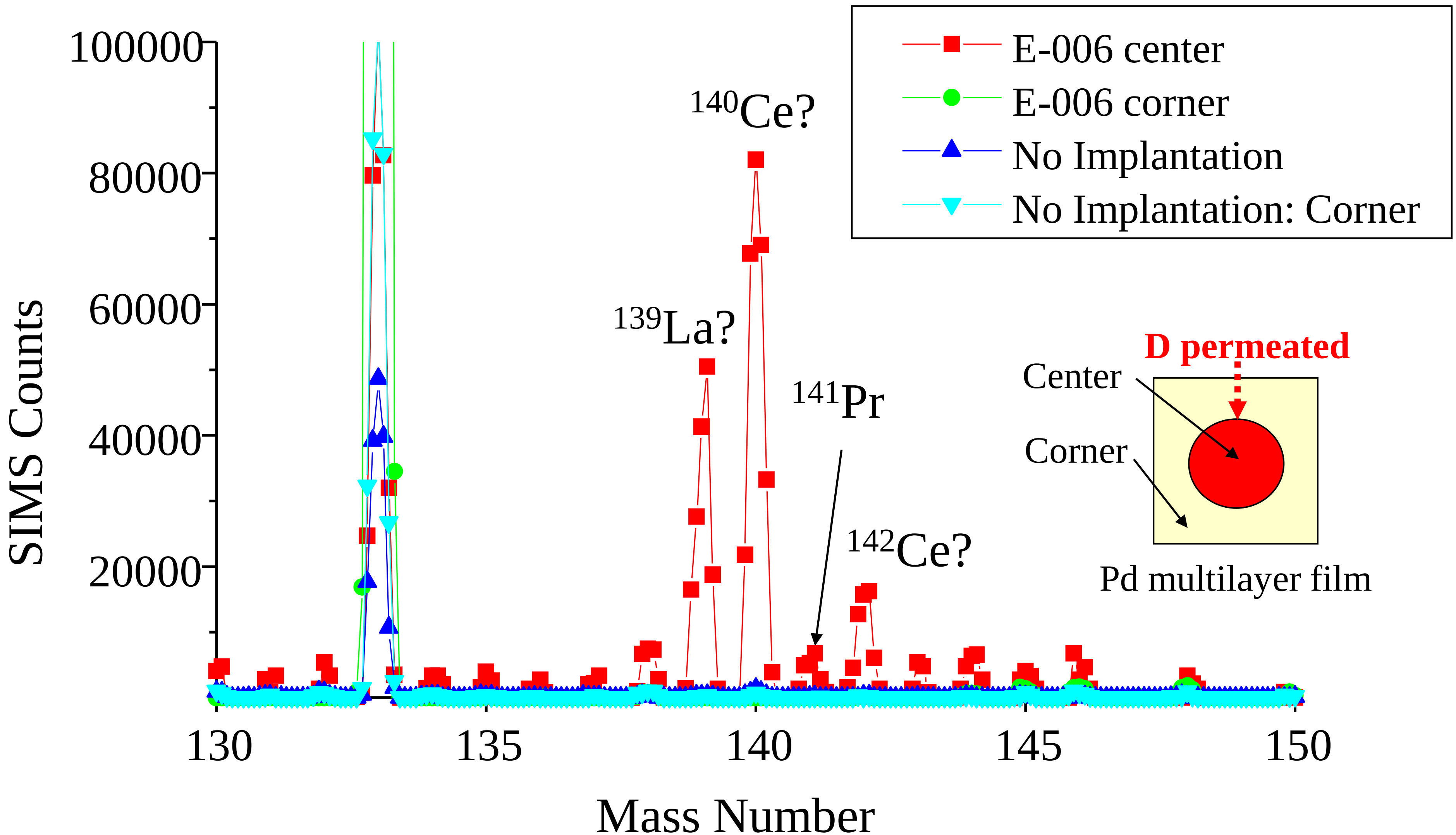




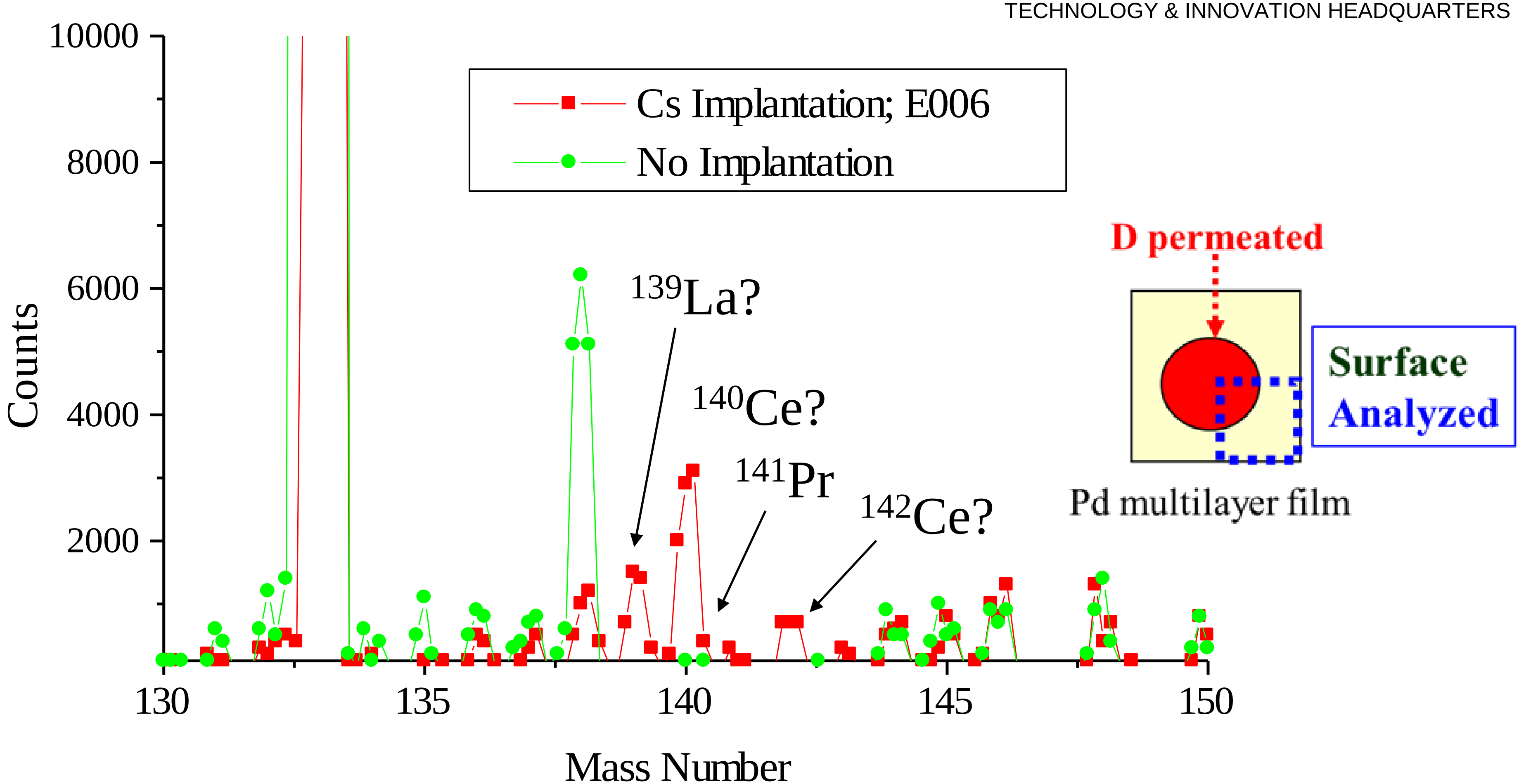
SIMS Analysis; E006 Wide Spectra



SIMS Analysis; E006



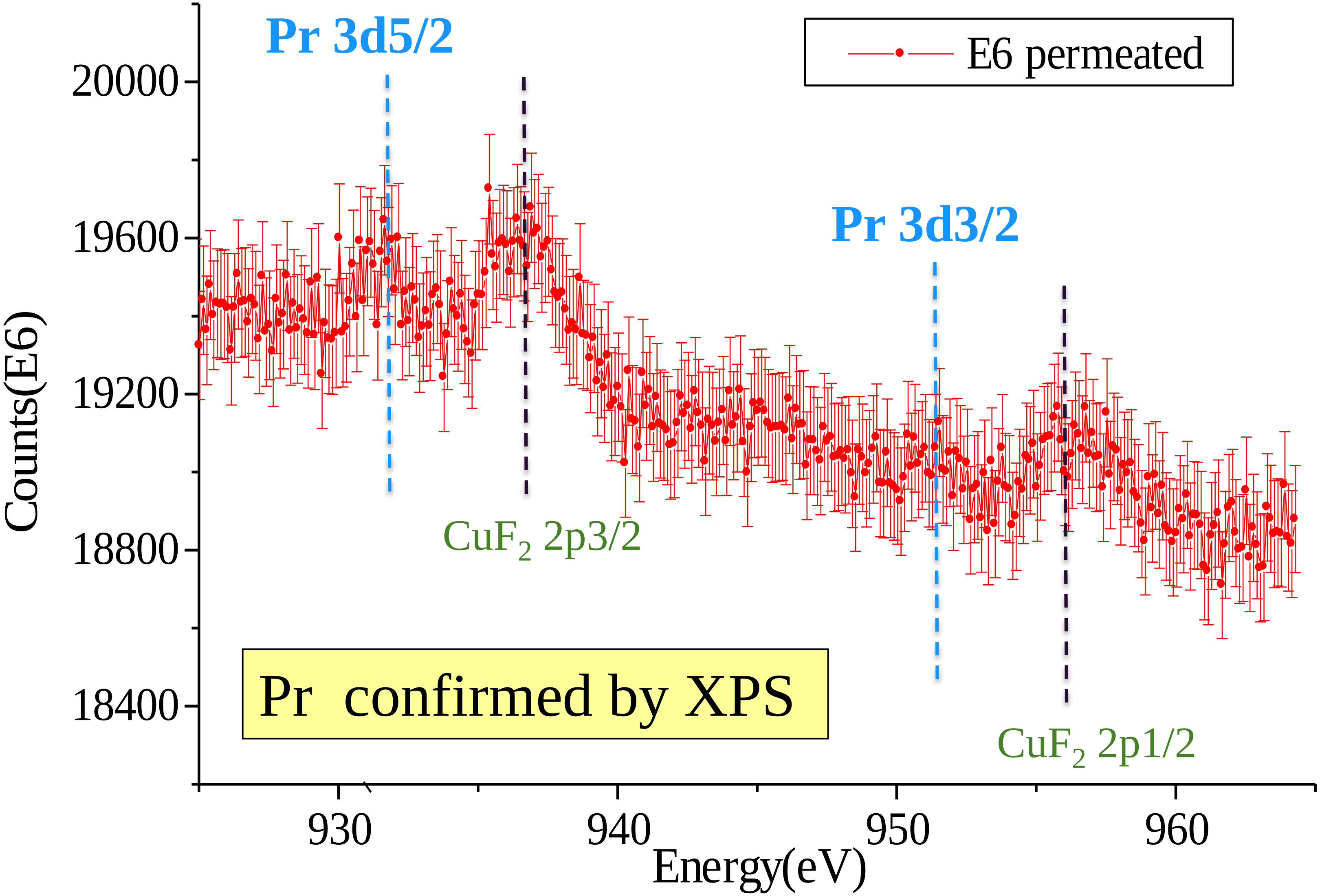
ICP-MS Analysis; E006



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

Different Tendency from D_2 gas permeation

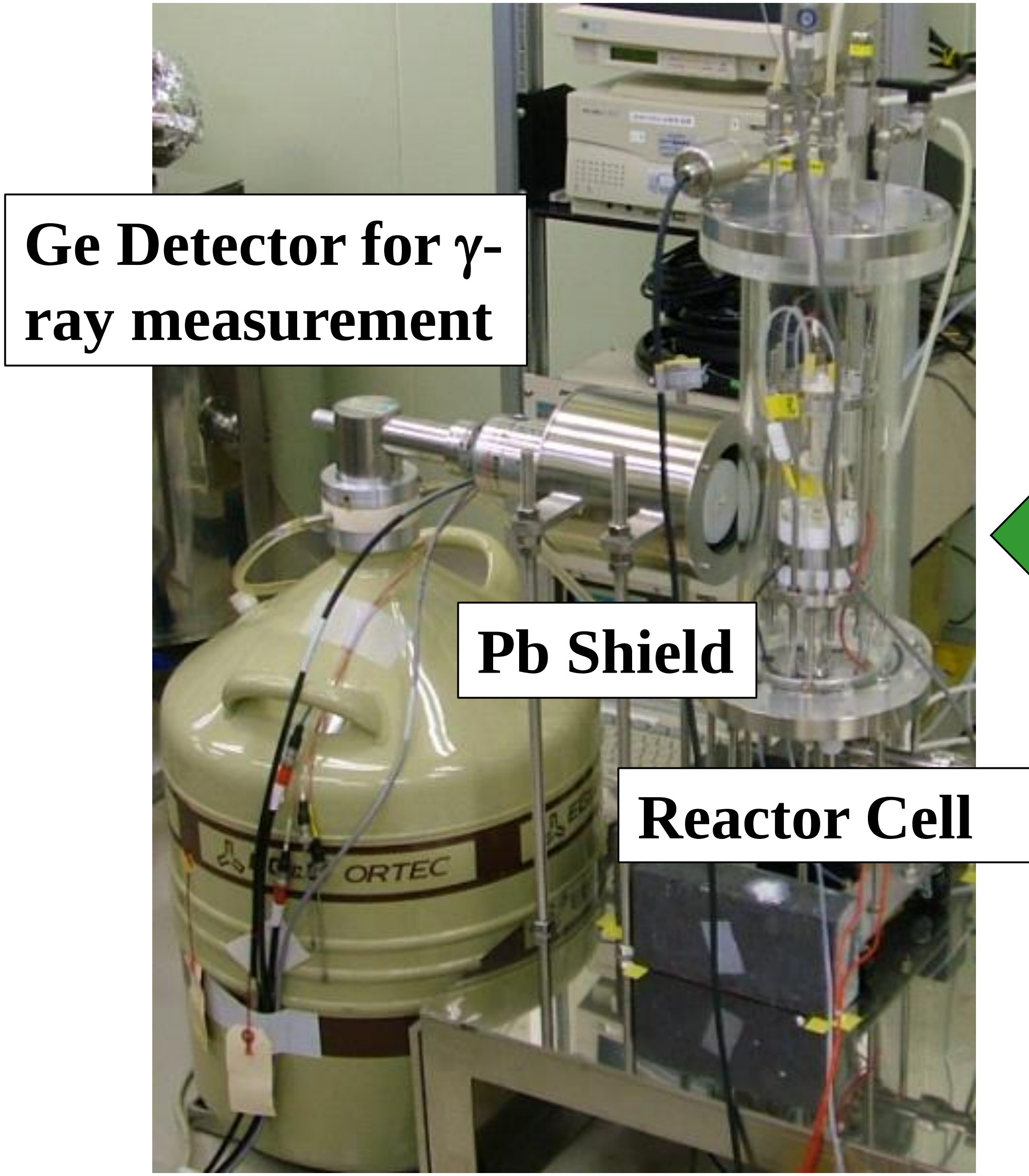
Confirmation of the products by XPS

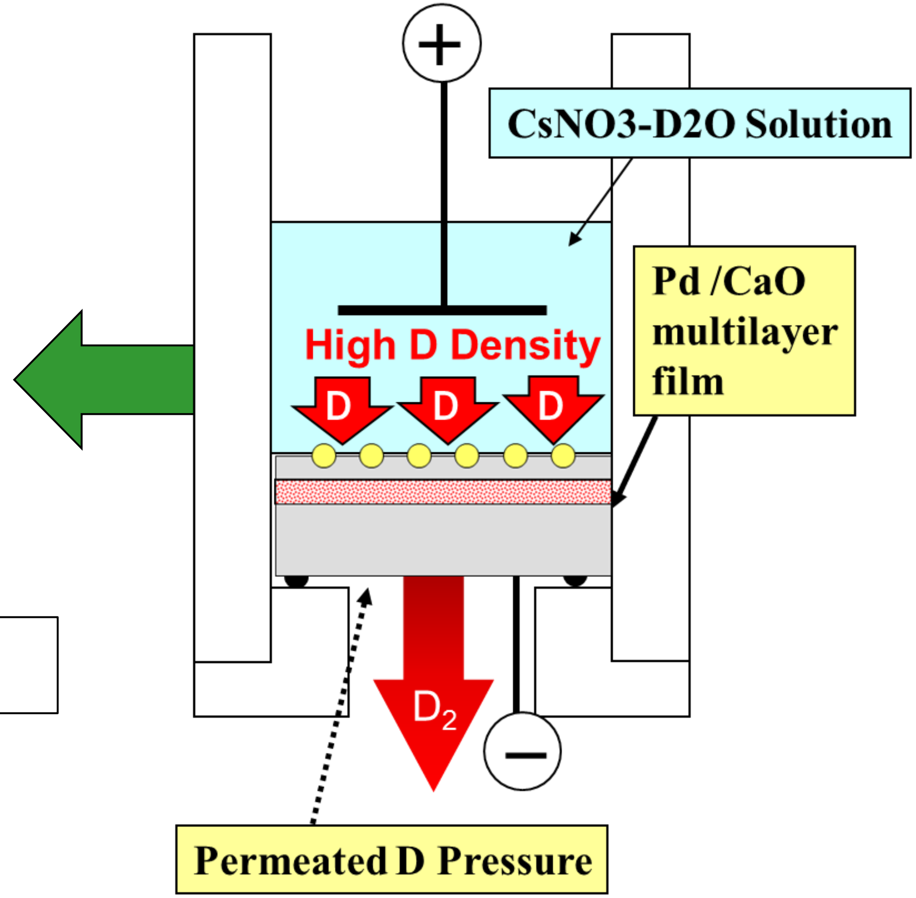


Pr confirmed by XPS

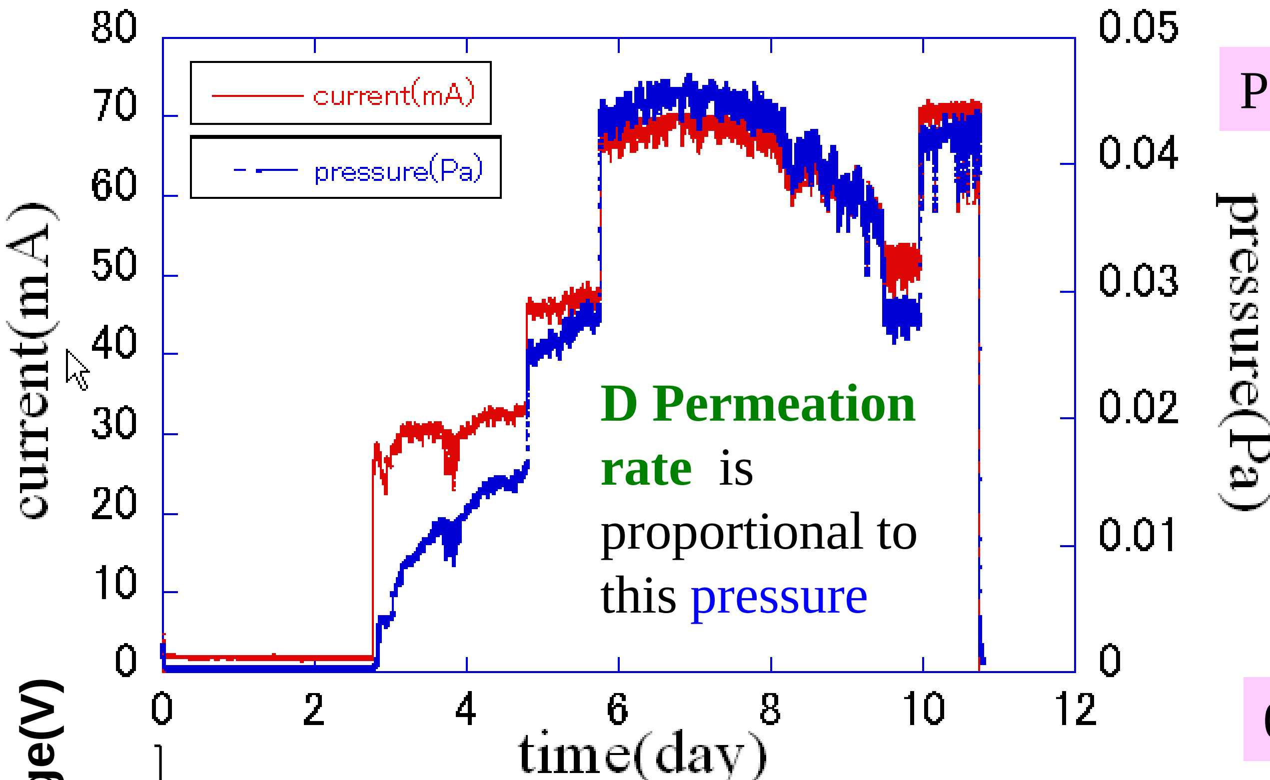
2-2. Observation of γ-ray peaks

Introduce a Gamma-ray Detector



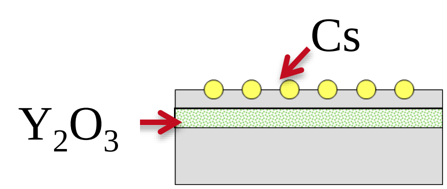


Example of Gamma-Ray Detection; E16

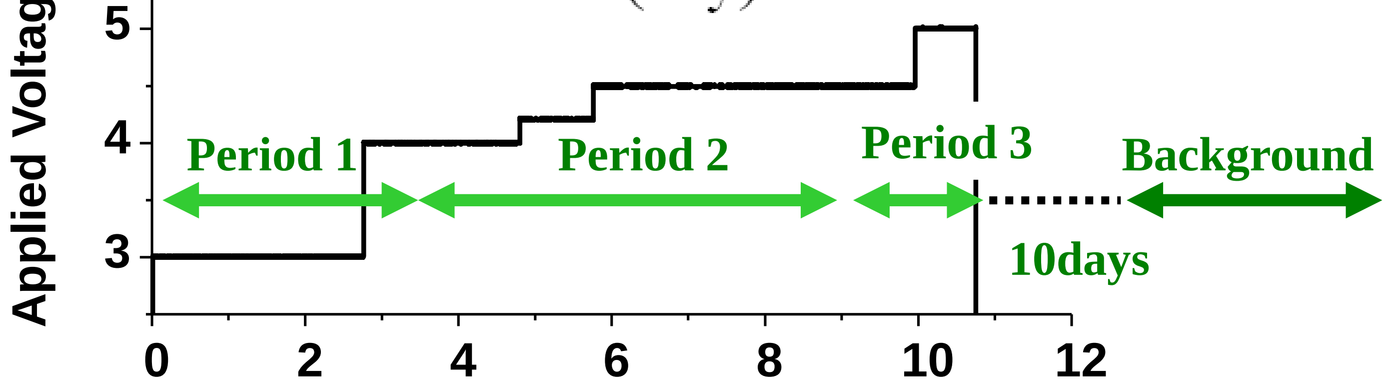


Pd/Y_2O_3/Pd multilayer film

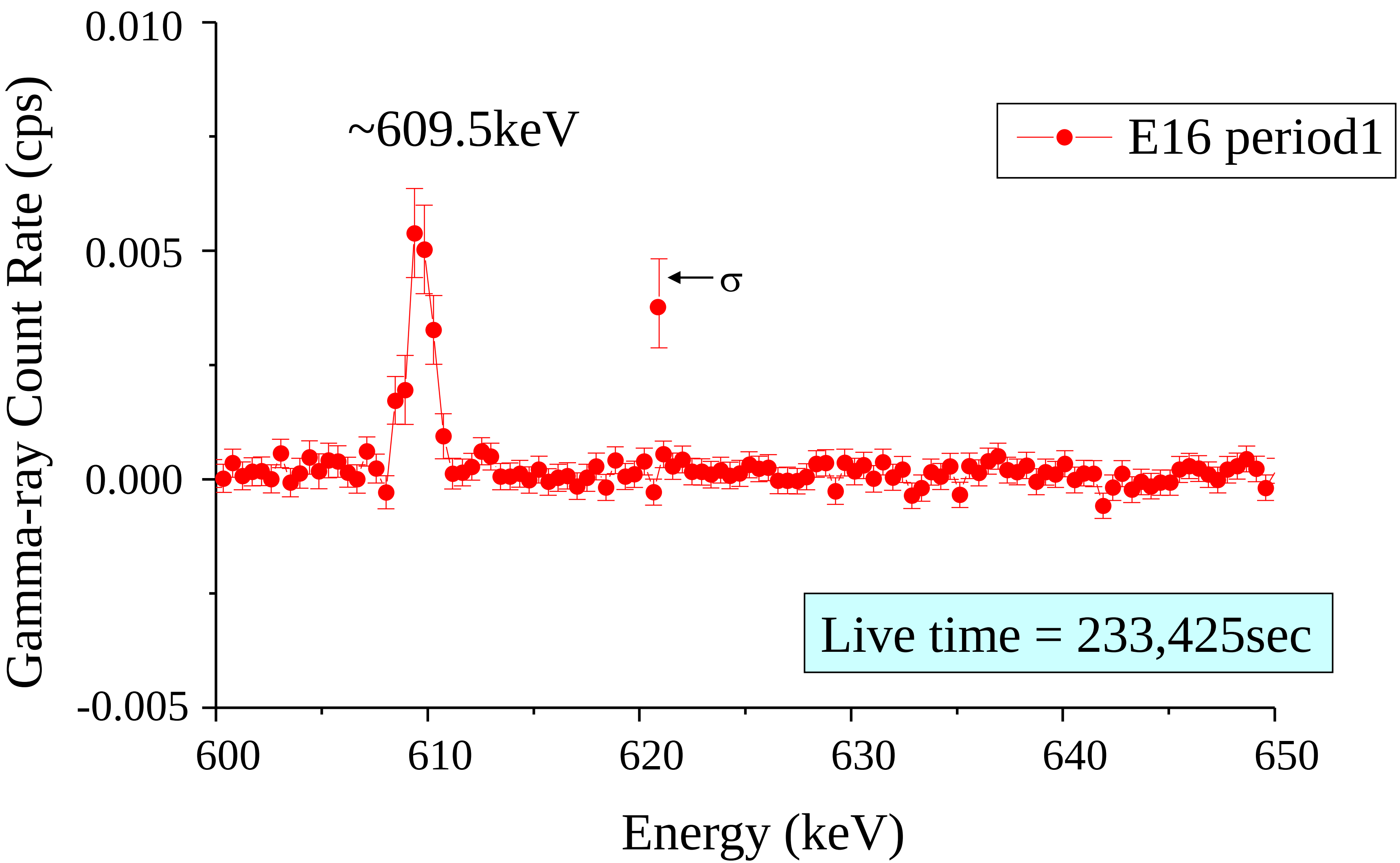
Cs ion implanted
1×10^{16}/cm^2 20kV



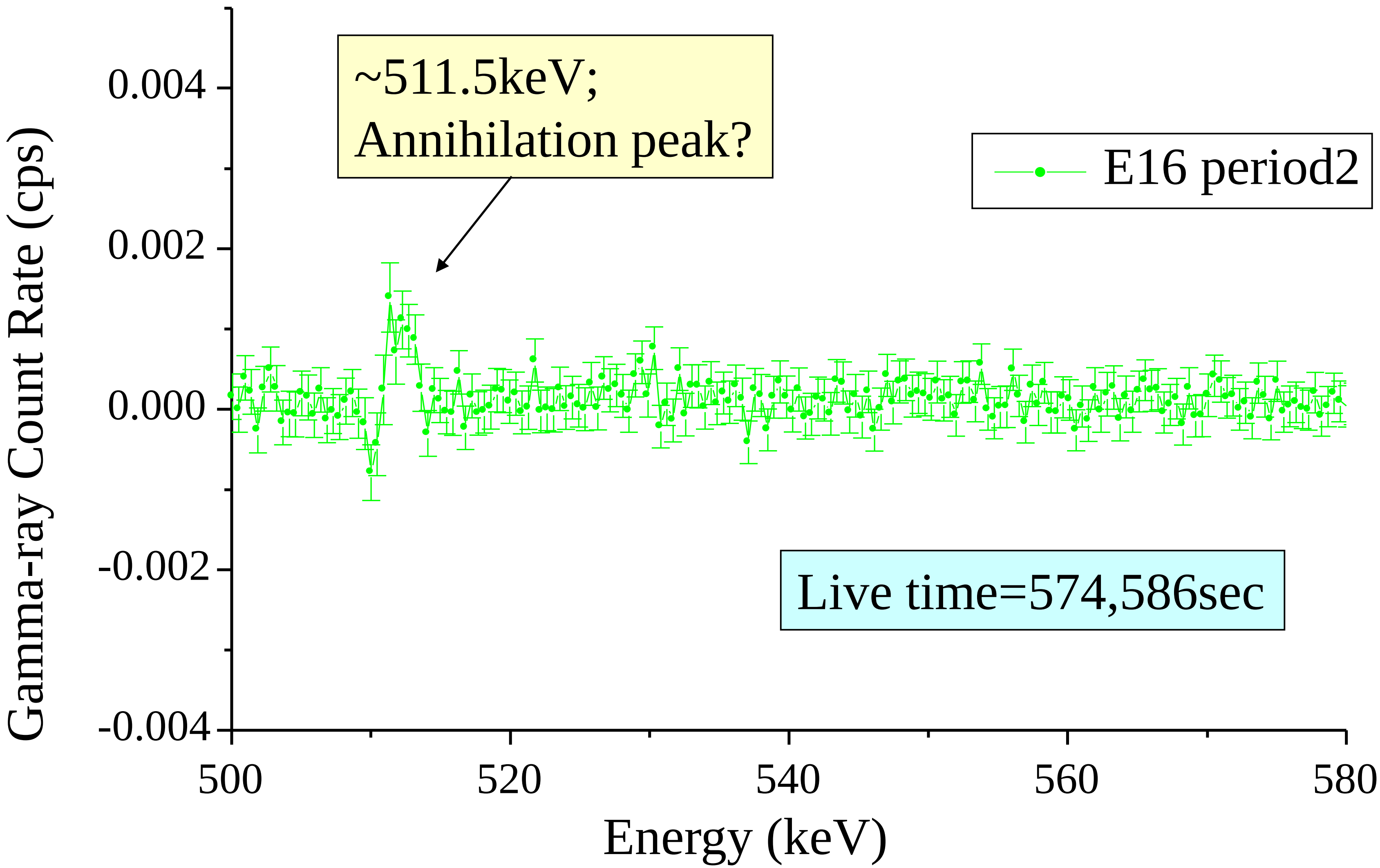
0.5M$CsNO_3$-D_2O Solution



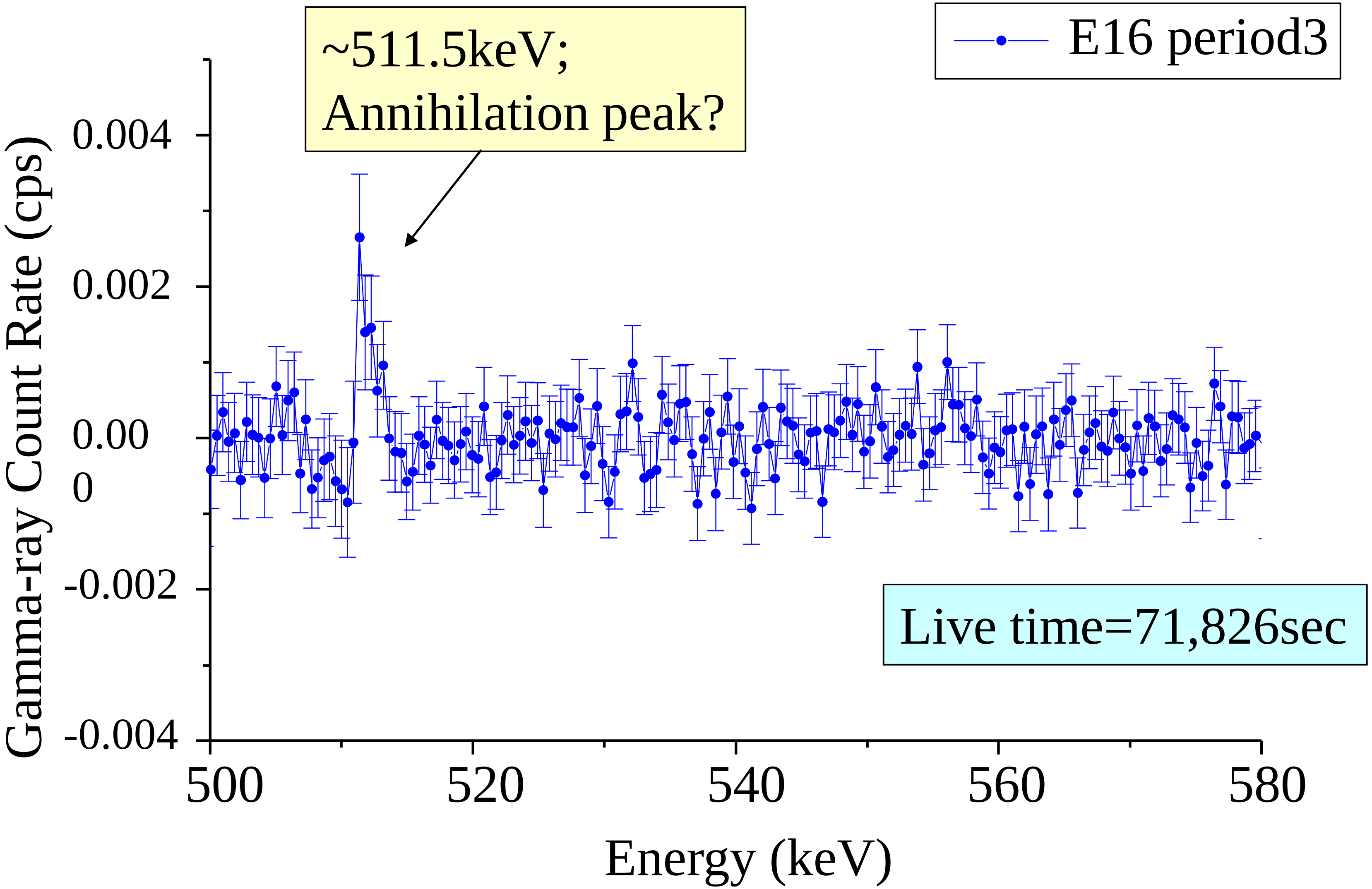
Gamma-ray Measurement (period 1)



Gamma-ray Measurement (period 2)



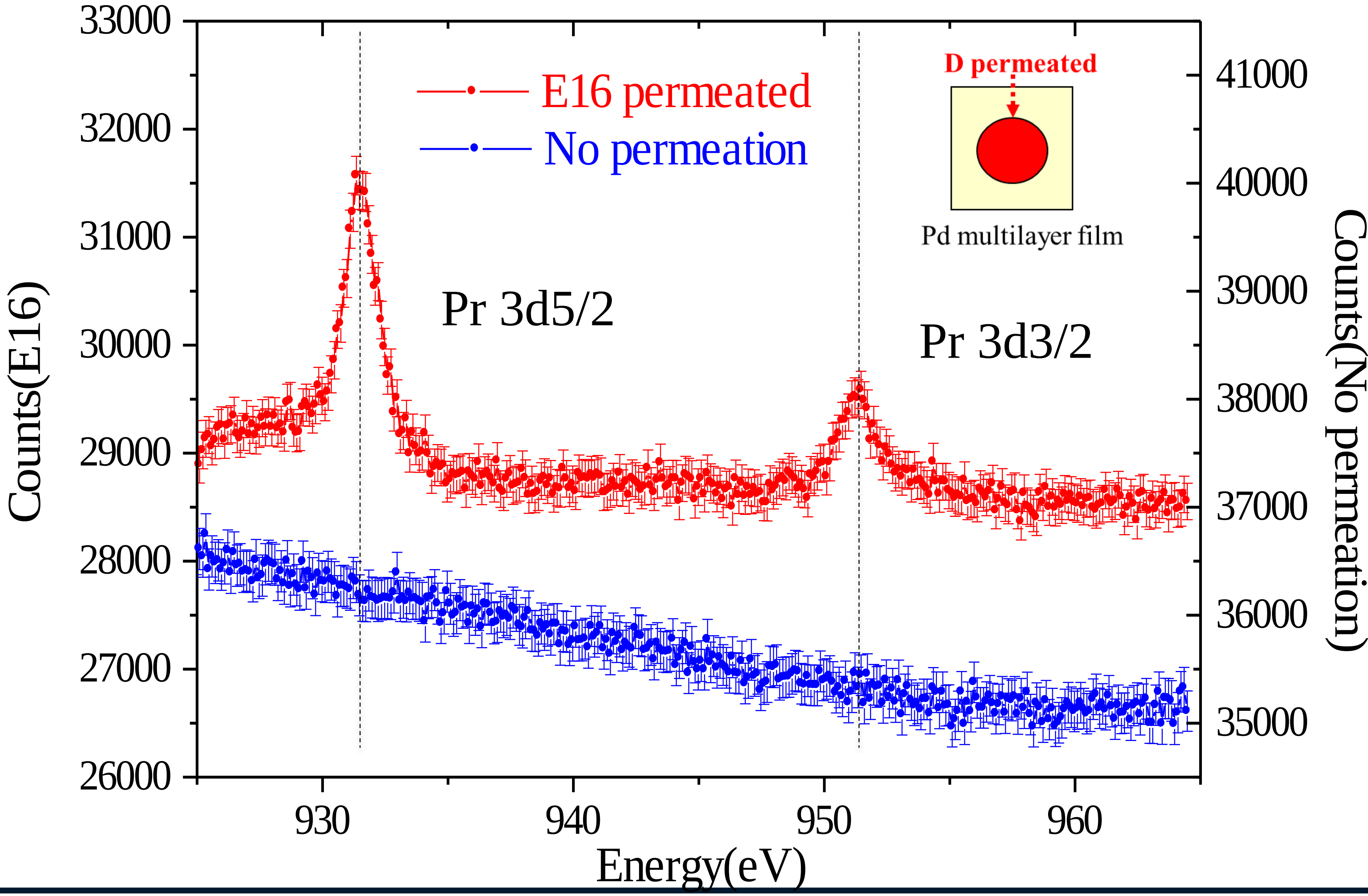
Gamma-ray Measurement (period 3)

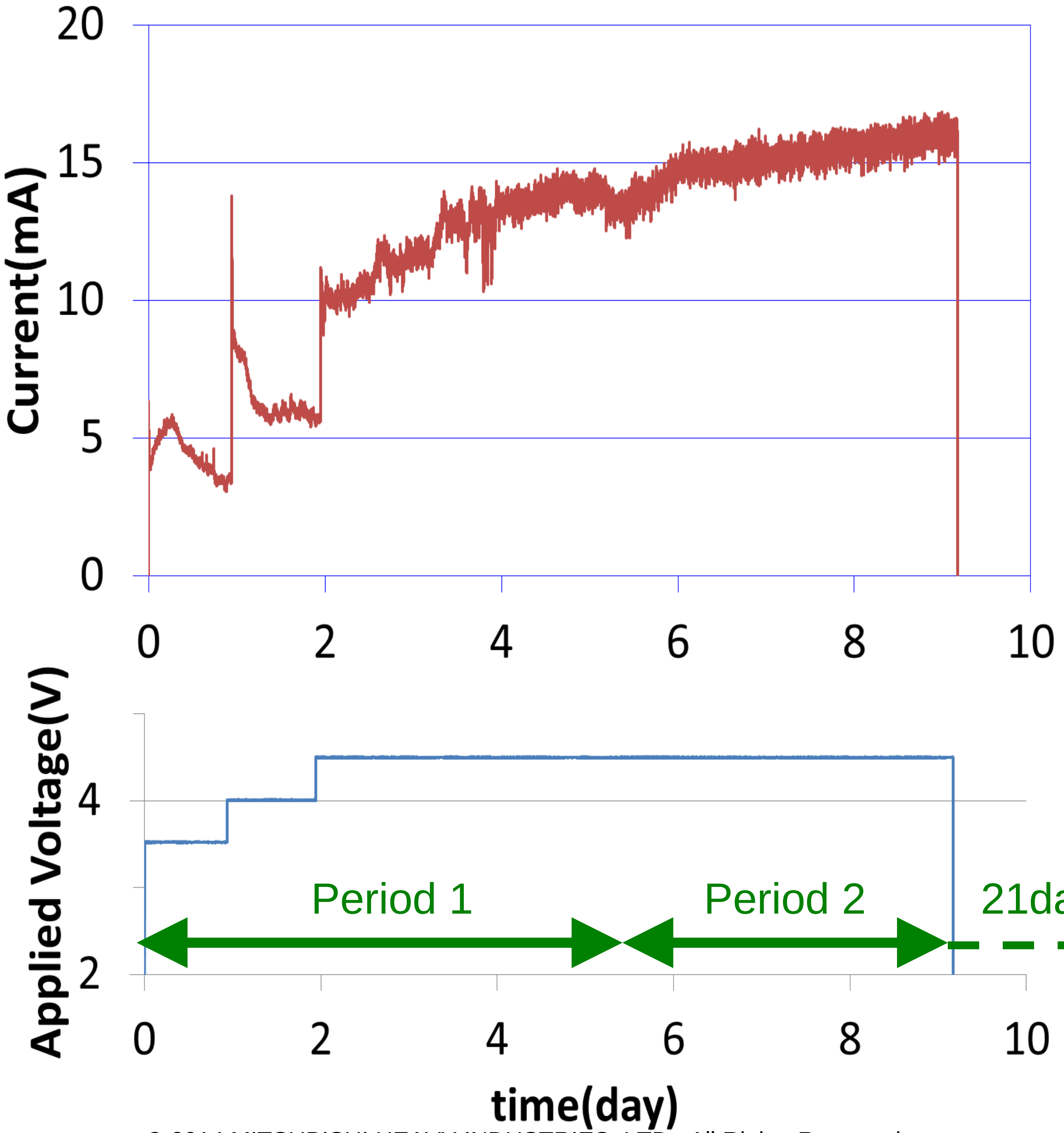


E16 Gamma-ray Measurement Summary

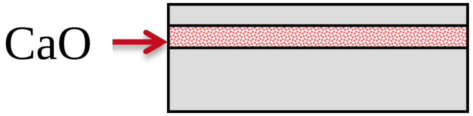
Time	Gamma-ray
Period 1	609.5keV gamma-ray detected No 511keV detected
Period 2	511.5keV gamma-ray detected No 609.5keV detected
Period 3	511.5keV gamma-ray detected No 609.5keV detected

Pr detected by XPS from the center of E16 sample



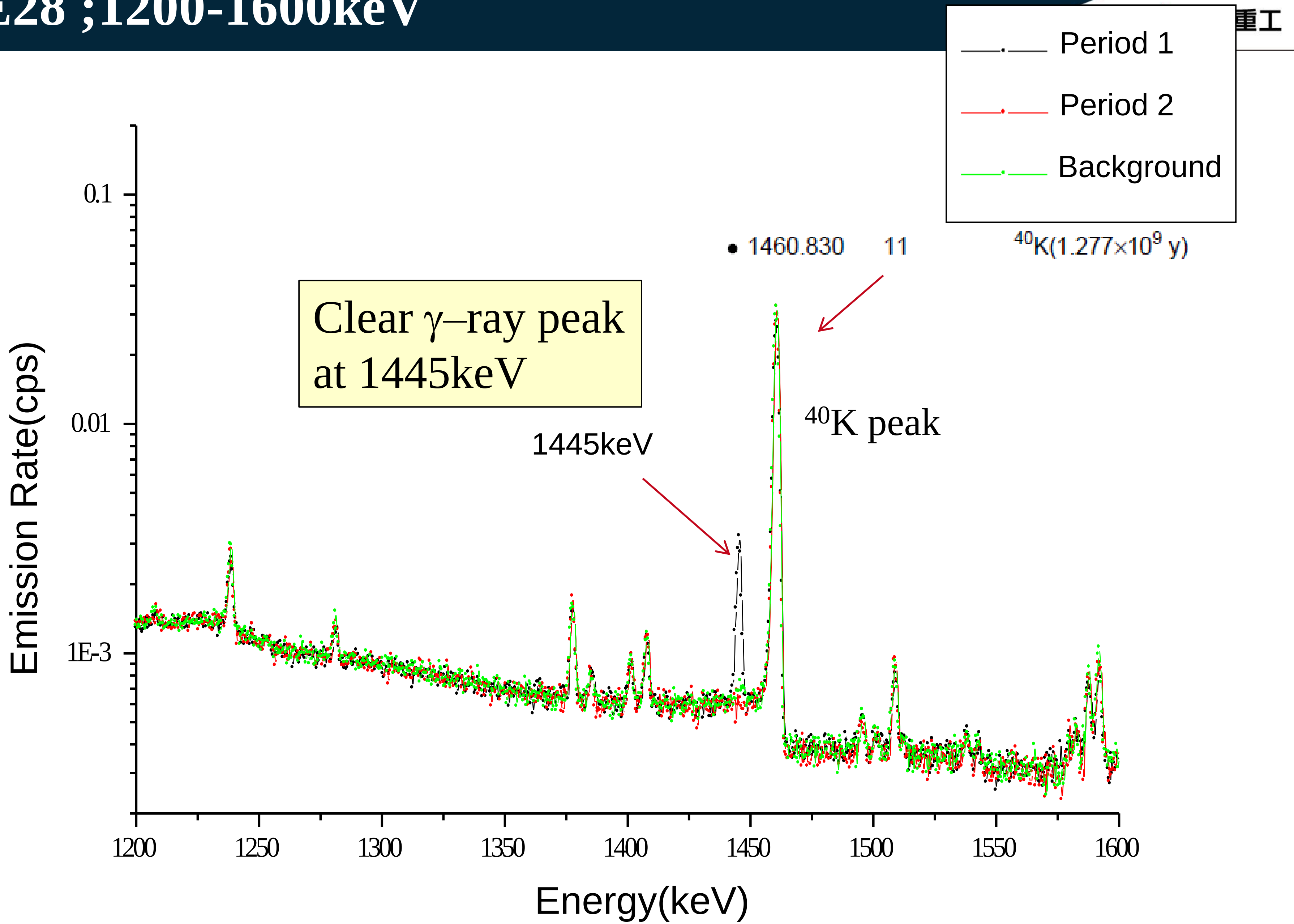


Pd/CaO/Pd multilayer film

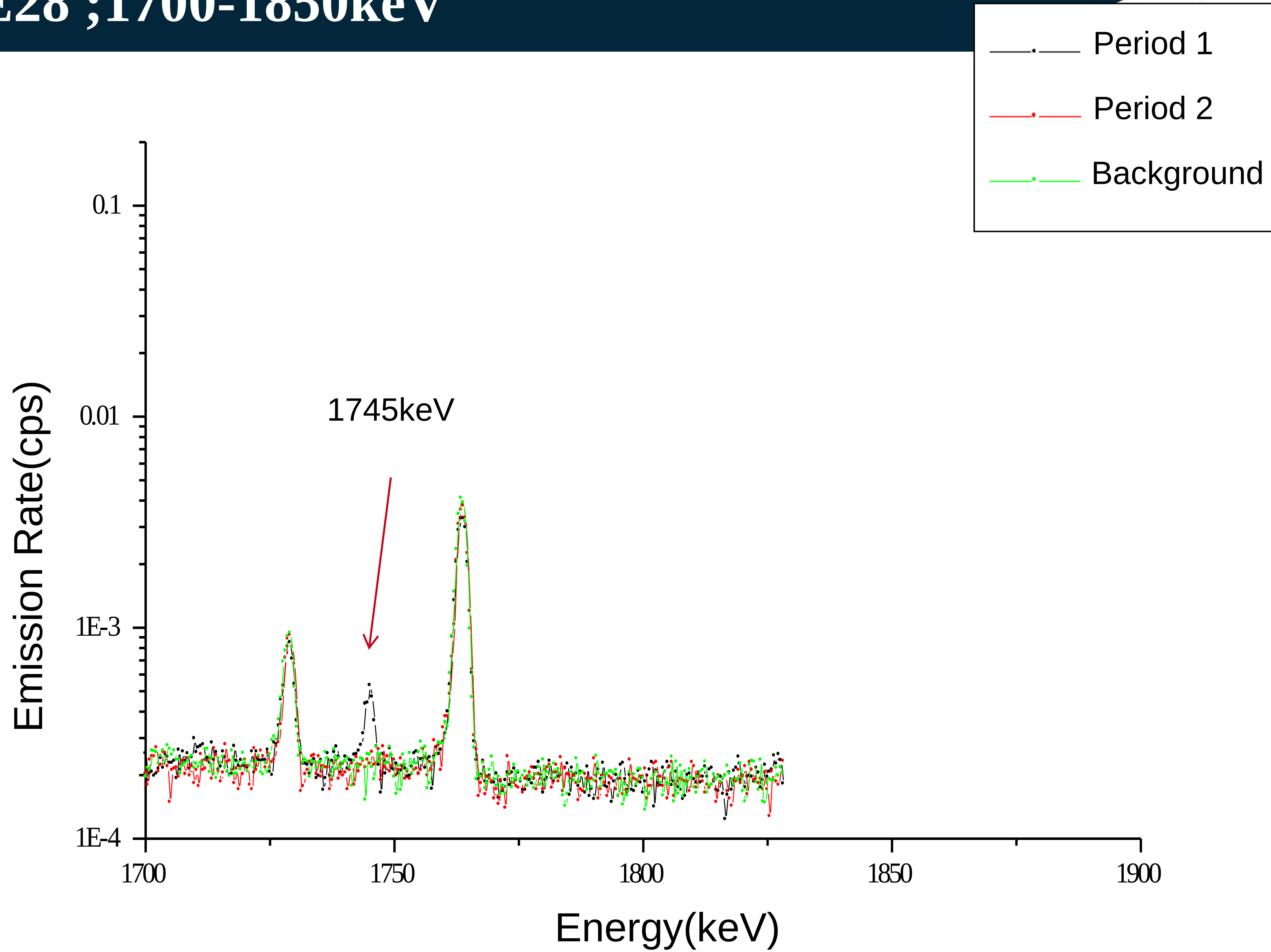


0.1MCsNO$_3$-D$_2$O Solution

E28 ;1200-1600keV



E28 ;1700-1850keV



Discussion on emitted γ-ray during E28 period1

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.

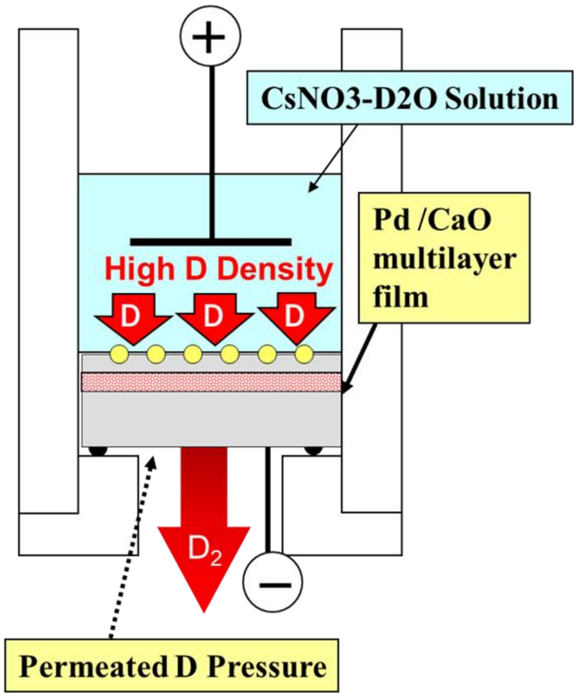
- **γ-rays from unstable nuclei**
- **γ-rays from excited nuclei**
- **Thermal neutron capture γ-rays**

Observed γ-rays seems to be attributed to minor short lived nuclei.

3. Preliminary Results on Consecutive Transmutation Experiments

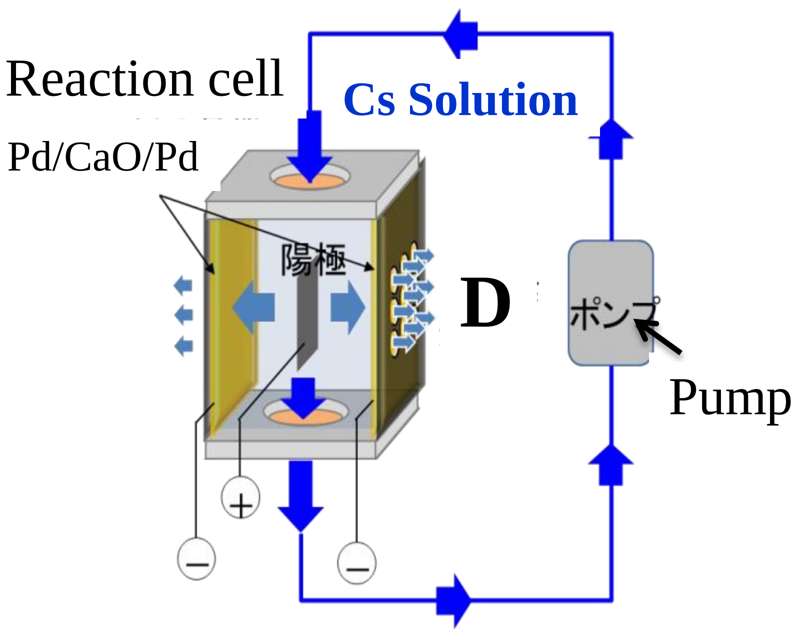
Batch vs. Consecutive Processing

Batch



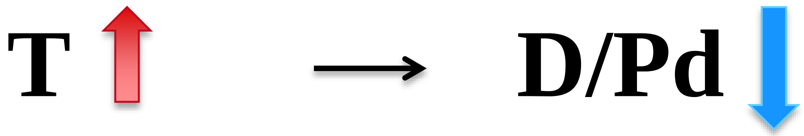
Addition of fixed quantity of Cs

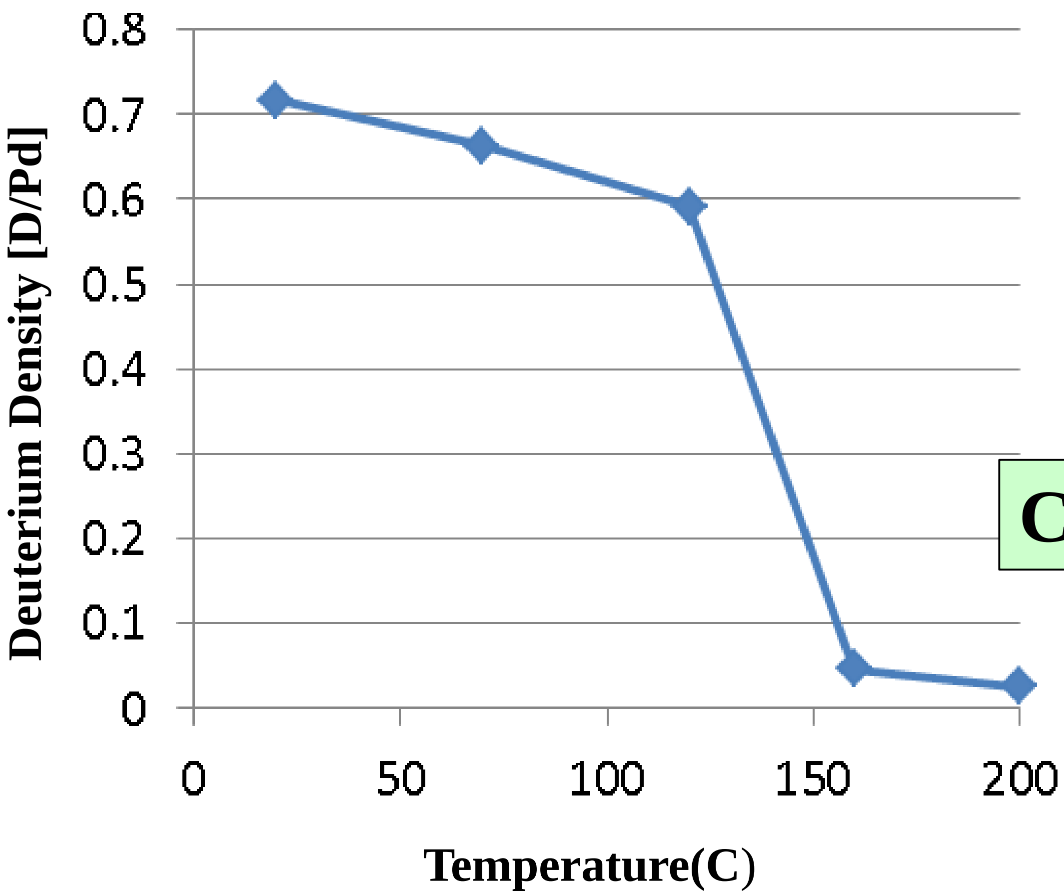
Consecutive



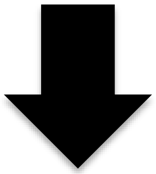
Circulation of Electrolyte

Cooling Pd Surface by Circulation of Electrolyte





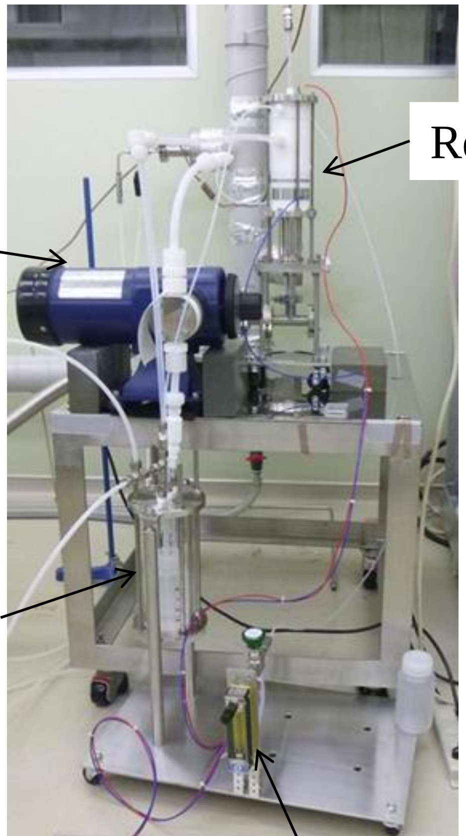
Cooling is important!

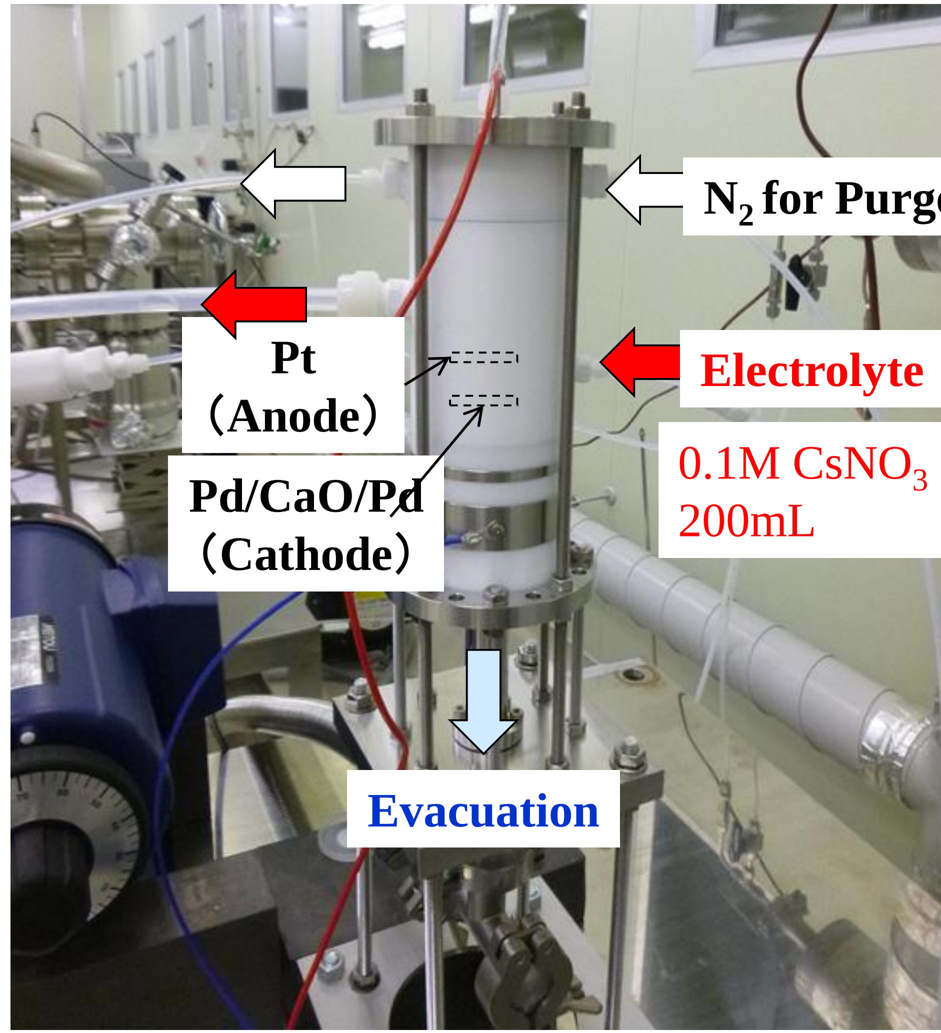


Circulation of Electrolyte

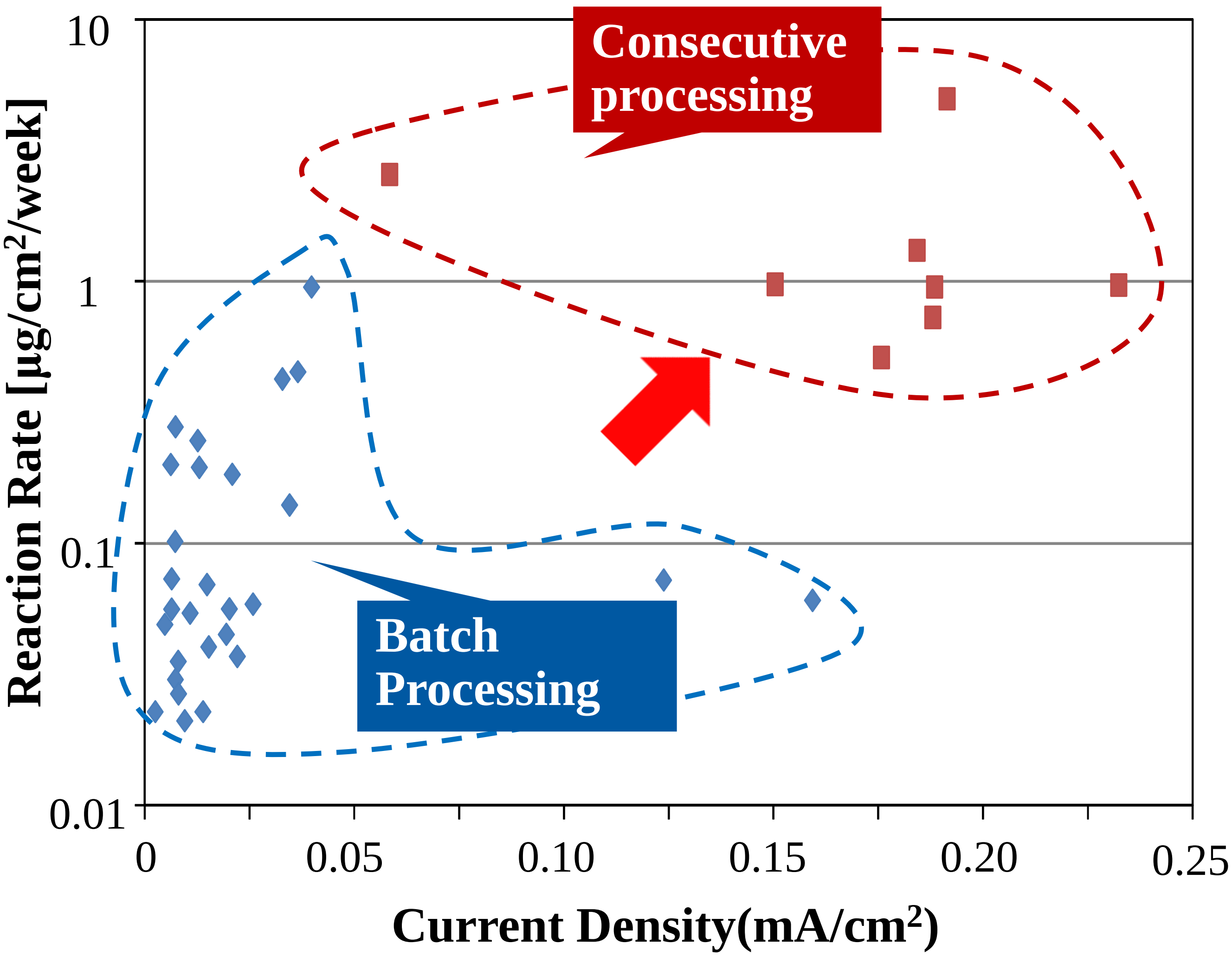
Experimental Setups for Consecutive Transmutati…

All Contact Surfaces with Electrolyte are made of Teflon to Avoid Contamination.





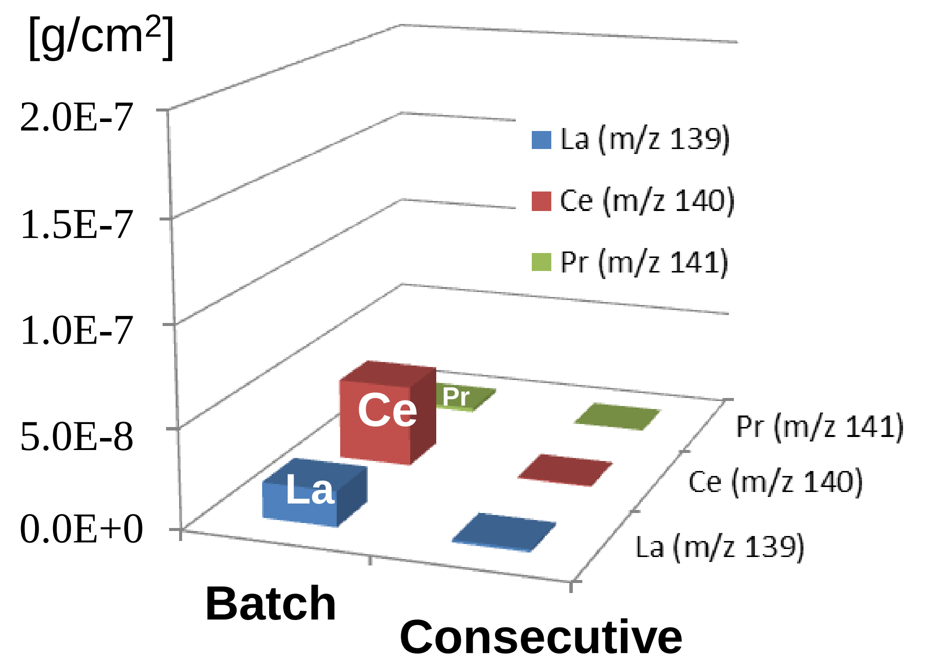
Reaction rates; Batch vs. Consecutive Processing

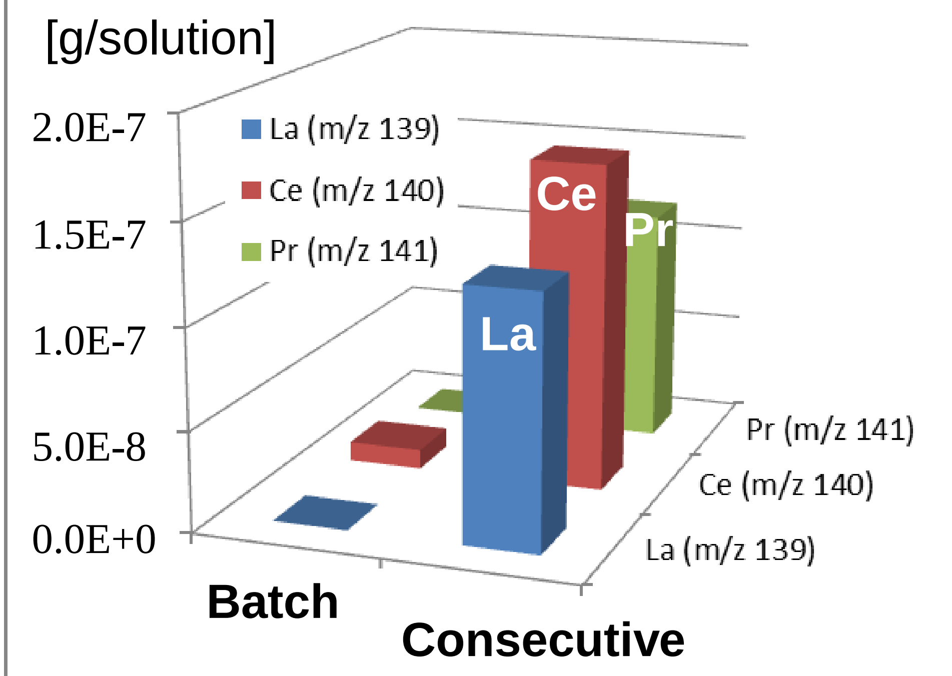


Products; Batch vs. Consecutive Processing

Pd/CaO/Pd Thin Film

0.1M $CsNO_3$ Solution





Batch : Products	→	**On the Pd Thin Film**
Consecutive : Products	→	**In Solution**

4. Replication Experiments by Toyota Central Research and Development Laboratories, Inc.

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Inductively Coupled Plasma Mass Spectrometry Study on the Increase in the Amount of Pr Atoms for Cs-Ion-Implanted Pd/CaO Multilayer Complex with Deuterium Permeation

Tatsumi Hioki[1*], Naoko Takahashi[1*], Satoru Kosaka[1], Teppei Nishi[1], Hirozumi Azuma[1], Shogo Hibi[1], Yuki Higuchi[1], Atsushi Murase[1], and Tomoyoshi Motohiro[2]

[1] *Toyota Central Research and Development Laboratories, Inc., Nagakute, Aichi 480-1192, Japan*
[2] *Green Mobility Collaborative Research Center and Graduate School of Engineering, Nagoya University, Nagoya 464-8603, Japan*
E-mail: l6908@mosk.tytlabs.co.jp; nao-t@mosk.tytlabs.co.jp

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To investigate the nuclear transmutation of Cs into Pr reported in this journal by Iwamura and coworkers, we have measured the amount of Pr atoms in the range as low as $\sim 1 \times 10^{10}$ cm^{-2} using inductively coupled plasma mass spectrometry for Cs-ion-implanted Pd/CaO multilayer complexes before and after deuterium permeation. The amount of Pr was initially at most 2.0×10^{11} cm^{-2} and it increased up to 1.6×10^{12} cm^{-2} after deuterium permeation. The increase in the amount of Pr could be explained neither by deuterium permeation-stimulated segregation of Pr impurities nor by external contamination from the experimental environment during the permeation. No increase in Pr was observed for permeation with hydrogen. These findings suggest that the observed increase in Pr with deuterium permeation can be attributed to a nuclear origin, as reported by Iwamura and coworkers, although the amount of the increase in Pr is two orders of magnitude less than that reported by them.

Experimental Setup

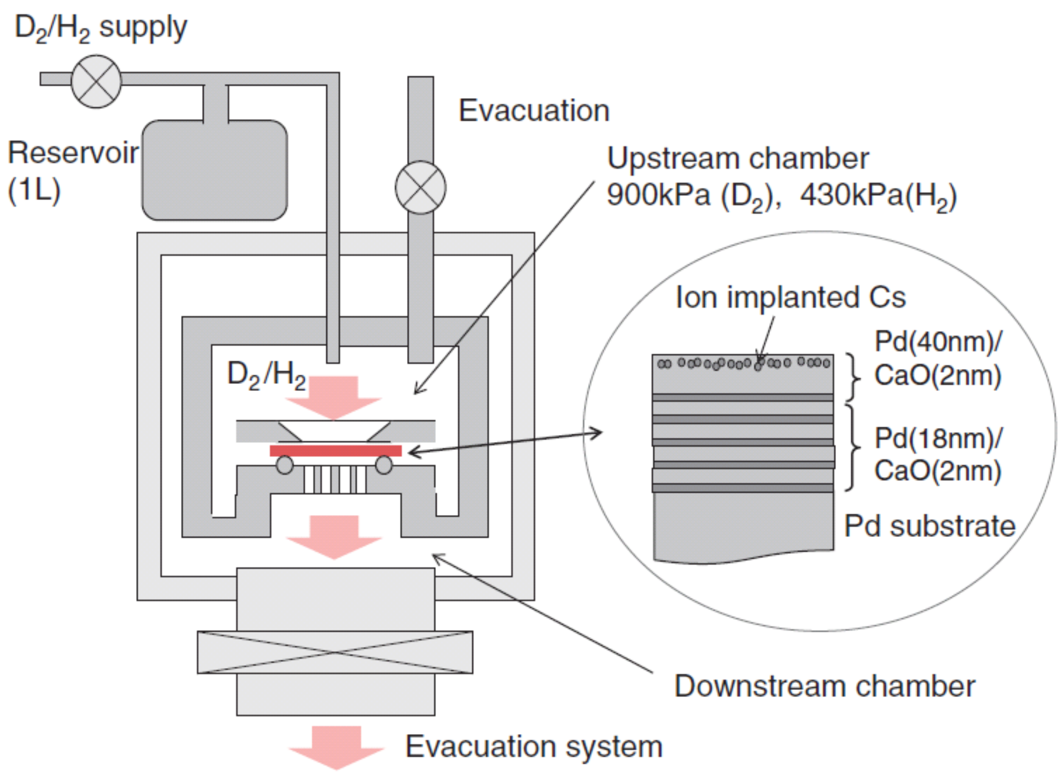

Fig. 1. (Color) Schematics of deuterium (/hydrogen) permeation system and Cs-ion-implanted Pd/CaO multilayer complex.

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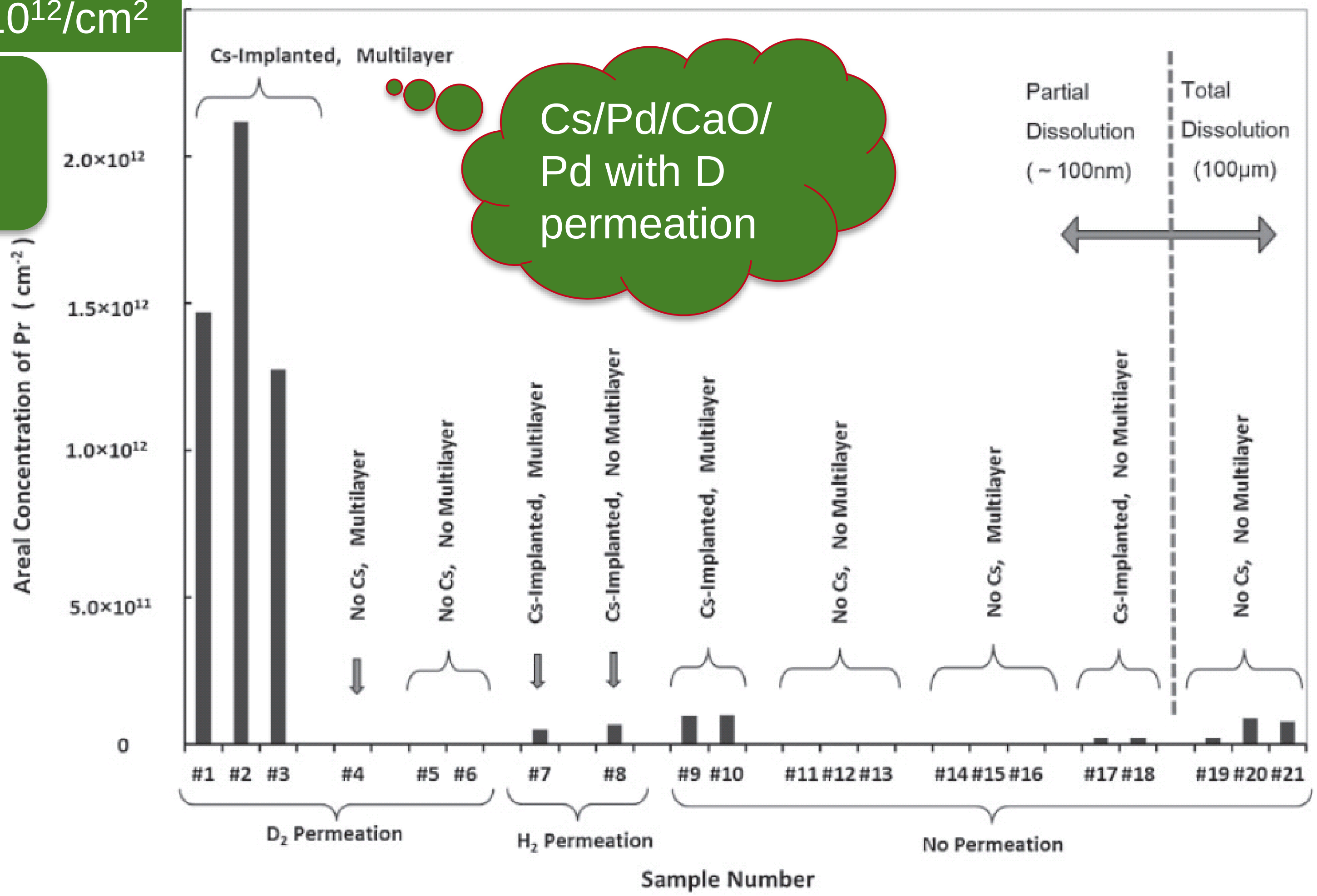
Results

2.5X10^{12}/cm^2

Pr



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Concluding Remarks

1. **Deuterium Permeation Induced Transmutation Reaction have been observed in the Pd complexes, which are composed of Pd and CaO thin film and Pd substrate.**
2. **Electrochemical permeation aiming the increase the local deuteron density near the surface of Pd made increase transmuted products.**
3. **Statistically significant γ-rays which have clear energy spectra were detected. At present, we have limited examples. Further study is necessary.**
4. **Preliminary consecutive transmutation experiments gave us higher reaction rates than batch processing up to now. Much products were recovered in the solution.**
5. **Toyota R&D Lab successfully reproduced permeation induced transmutation of Cs into Pr.**

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