In situ Accelerator-Based Characterization of CaO/Sr/Pd Samples under Deuterium Permeation

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- **Deuterium permeation through Pd/(CaO+Pd)/Pd sample induced nuclear transmutations;**

$^{133}Cs \rightarrow ^{141}Pr$, $^{88}Sr \rightarrow ^{96}Mo$, $^{138}Ba \rightarrow ^{150}Sm$ and $^{137}Ba \rightarrow ^{149}Sm$.

1. Y. Iwamura, M. Sakano and T. Itoh; Jpn. J. Appl. Phys. 41 (2002) 4642-4650.
2. Y. Iwamura, T. Itoh, M. Sakano, S. Kuribayashi, Y. Terada, T. Ishikawa and J. Kasagi; Proc. ICCF11, 2004, Marseilles, France.

In the present work;

- **The objective is**
  - **to investigate the dependence of the transformation rate on material, D flux, flow direction, temperature, *etc*.**
  - **to find the optimum sample structure.**
  - **from a point of view of enhancing the transformation yield.**
- **Diagnostic methods;**

  ***in situ* and simultaneous measurements of compositional change during D permeation by accelerator analyses;**

  PIXE (Particle induced X-ray emission analysis)

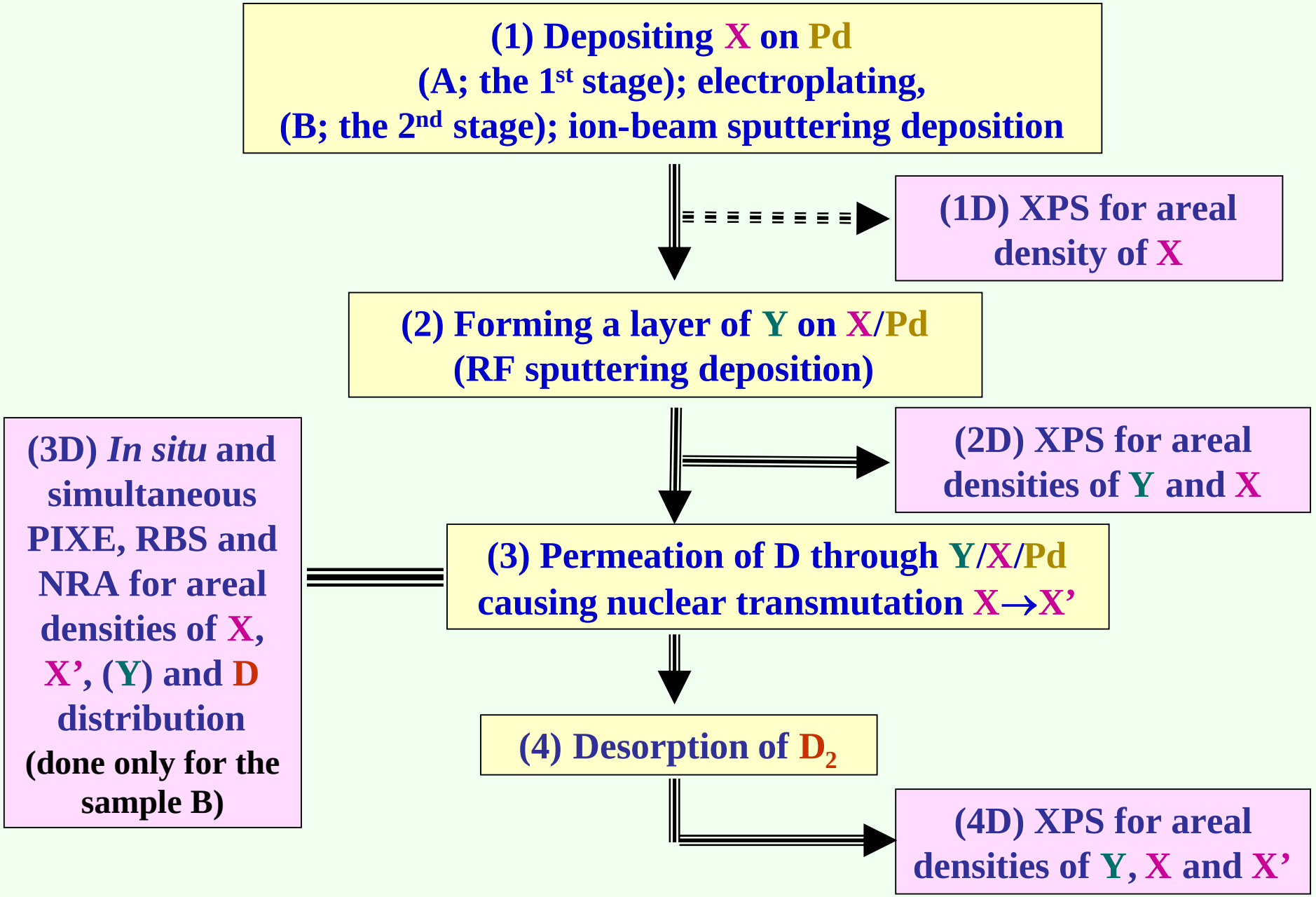
  RBS (Rutherford backscattering spectroscopy)

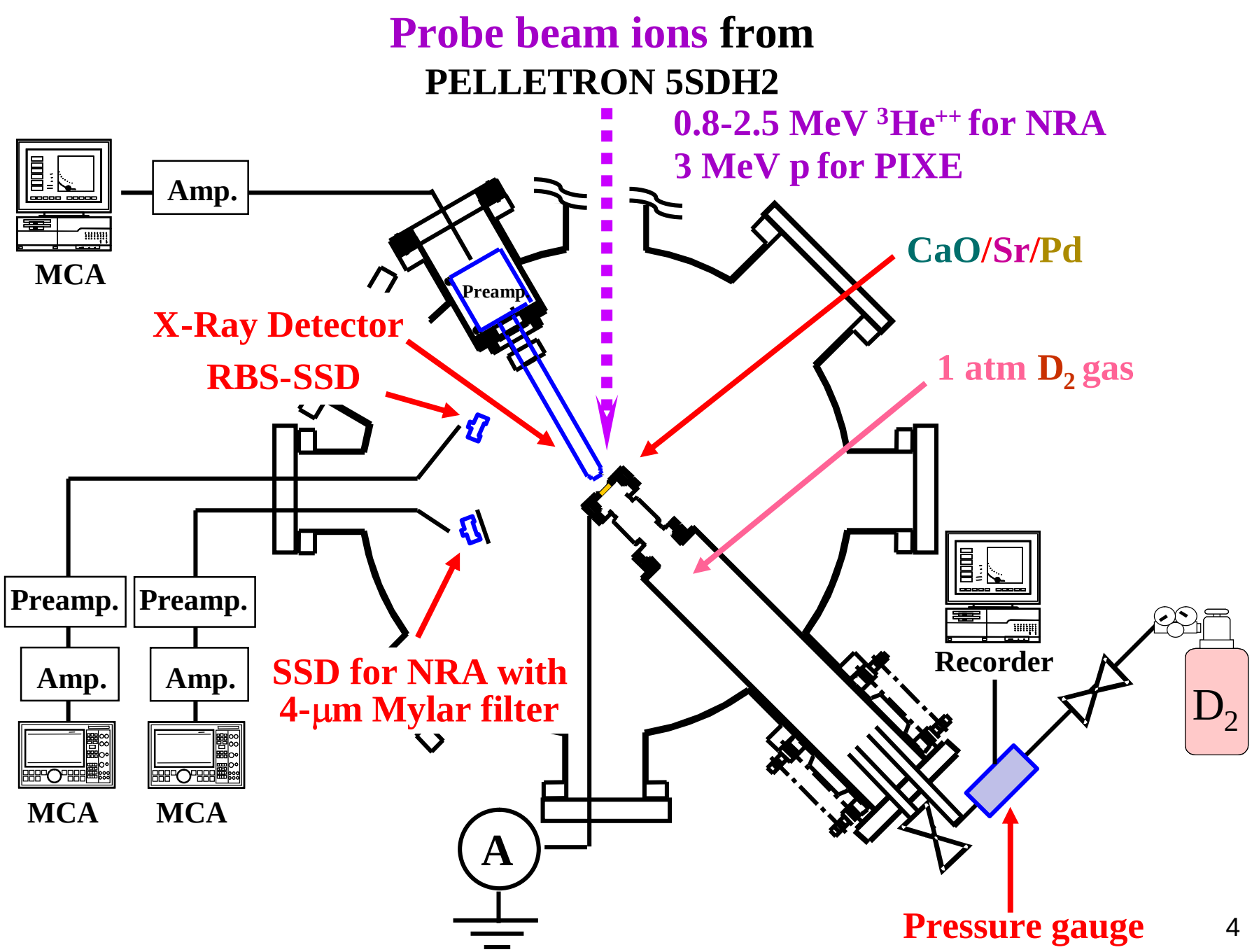
  NRA (Nuclear reaction analysis)

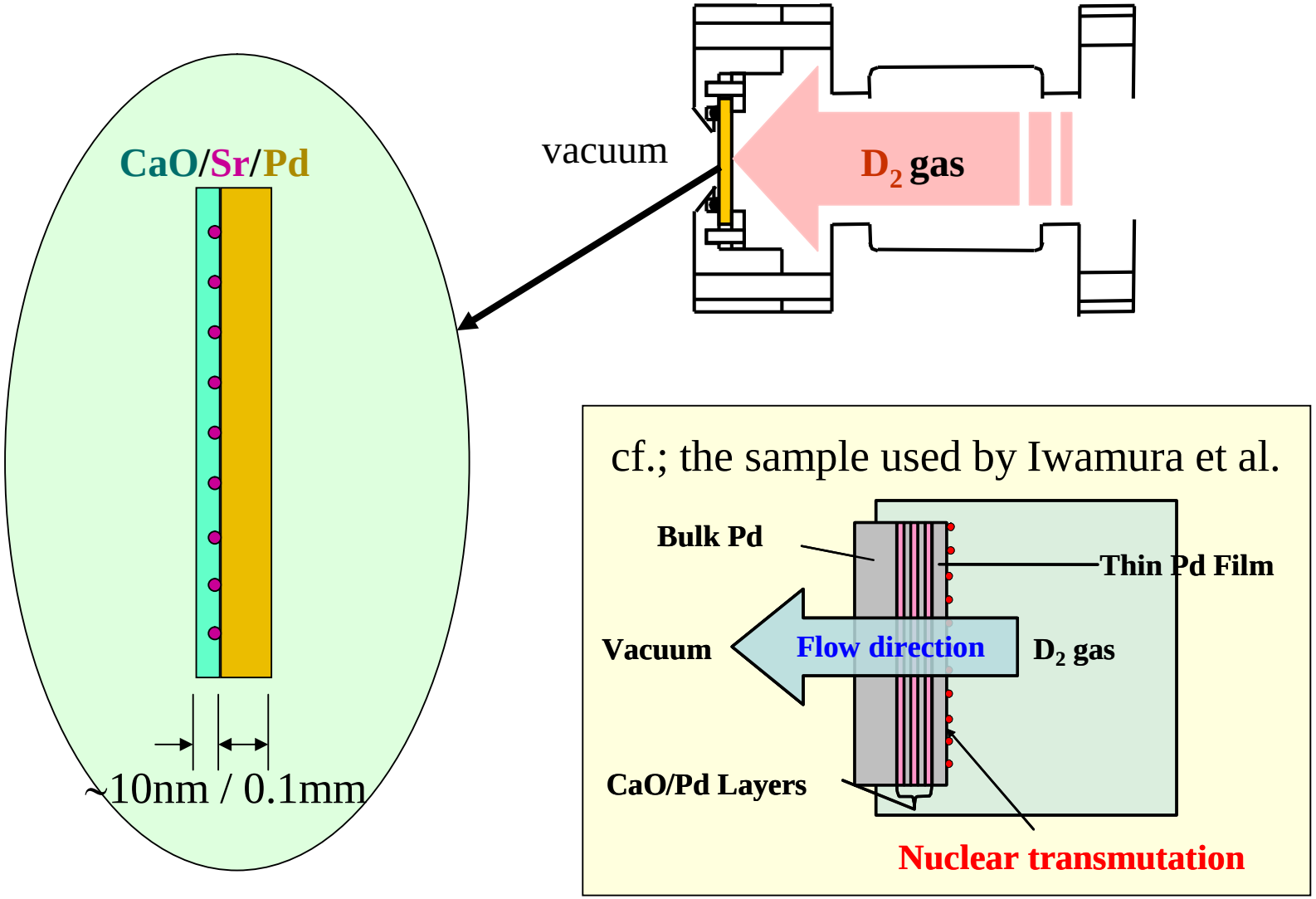
  ERDA (Elastic recoil detection analysis)

  as well as conventional XPS and Desorption measurements.

Experimental procedure; in the present work, X =Sr, Y=CaO

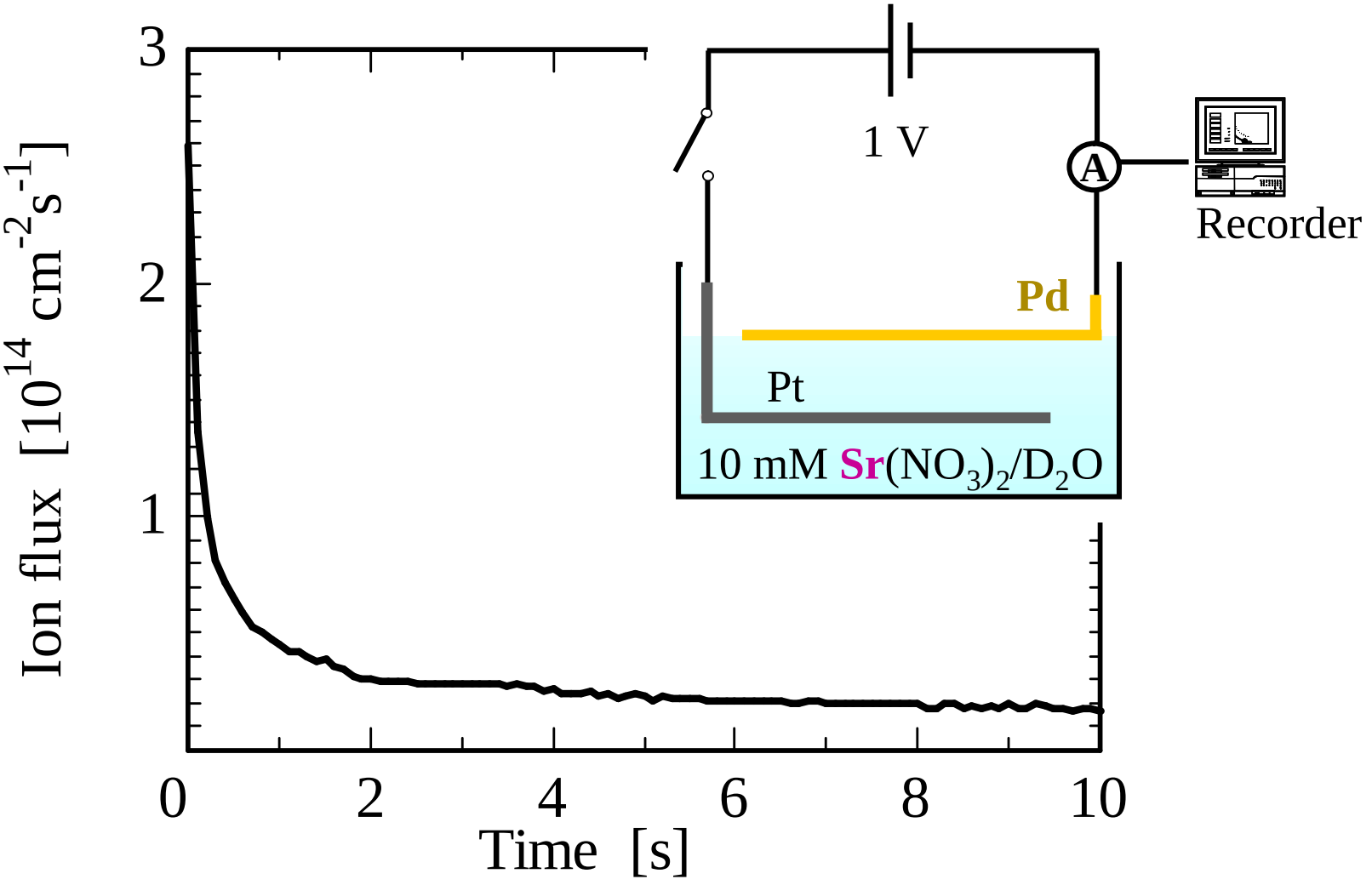



Probe beam ions from
PELLETRON 5SDH2
0.8-2.5 MeV $^3He^{++}$ for NRA
3 MeV p for PIXE
Amp.
MCA
Preamp.
CaO/Sr/Pd
X-Ray Detector
RBS-SSD
1 atm D_2 gas
Preamp.
Preamp.
Amp.
Amp.
MCA
MCA
SSD for NRA with
4-μm Mylar filter
Recorder
D_2
A
Pressure gauge



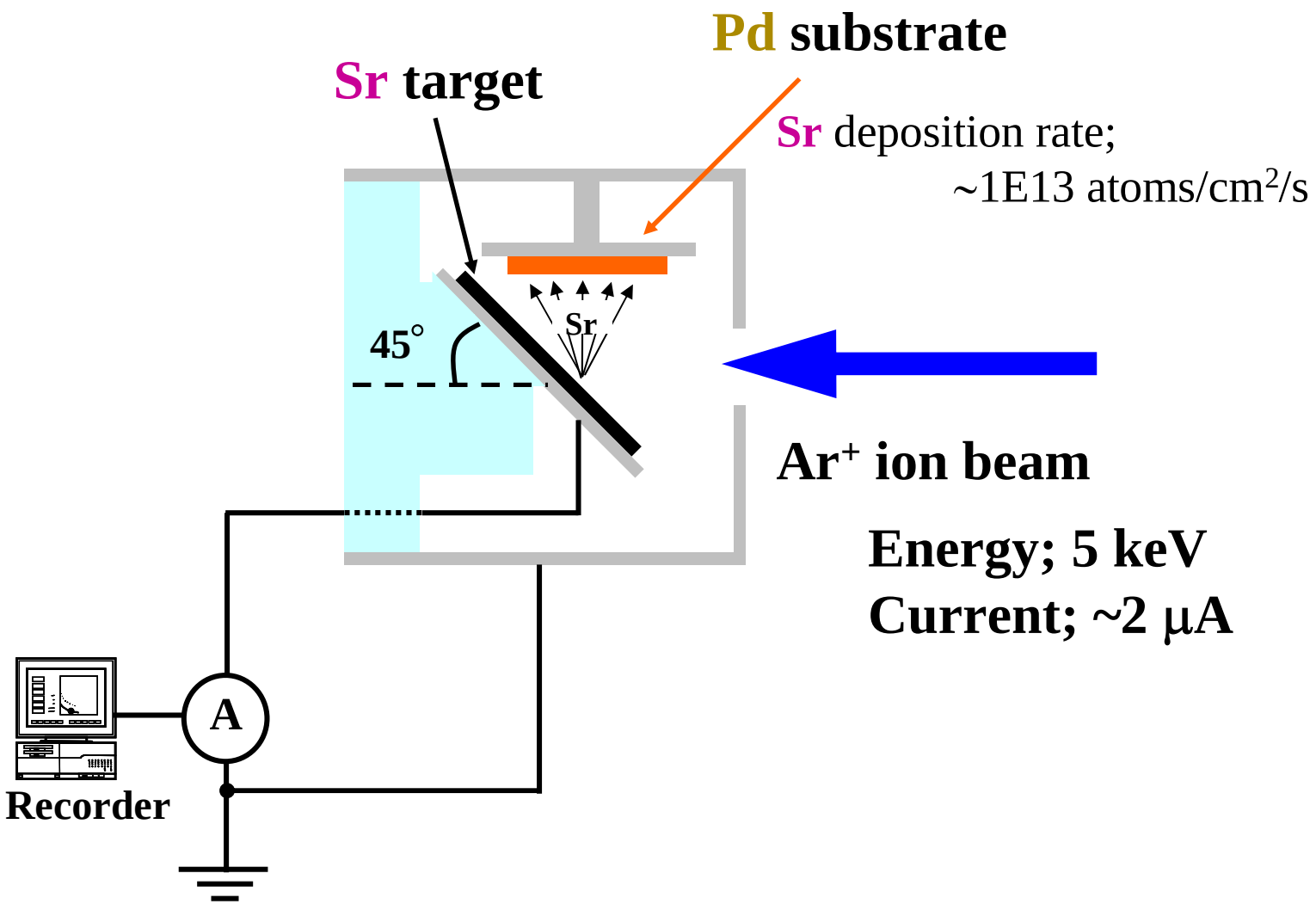
Deuterium permeates through the CaO(~10nm)/Sr/Pd sample from the rear surface out to vacuum.

(1) Depositing Sr on Pd ; (A) electrochemical method



Sr atoms are deposited on one side of a 0.1-mm-thick Pd foil as contamination with an areal density of the order of 10^{14} cm^{-2}.

(1) Depositing Sr on Pd ; (B) ion beam sputtering

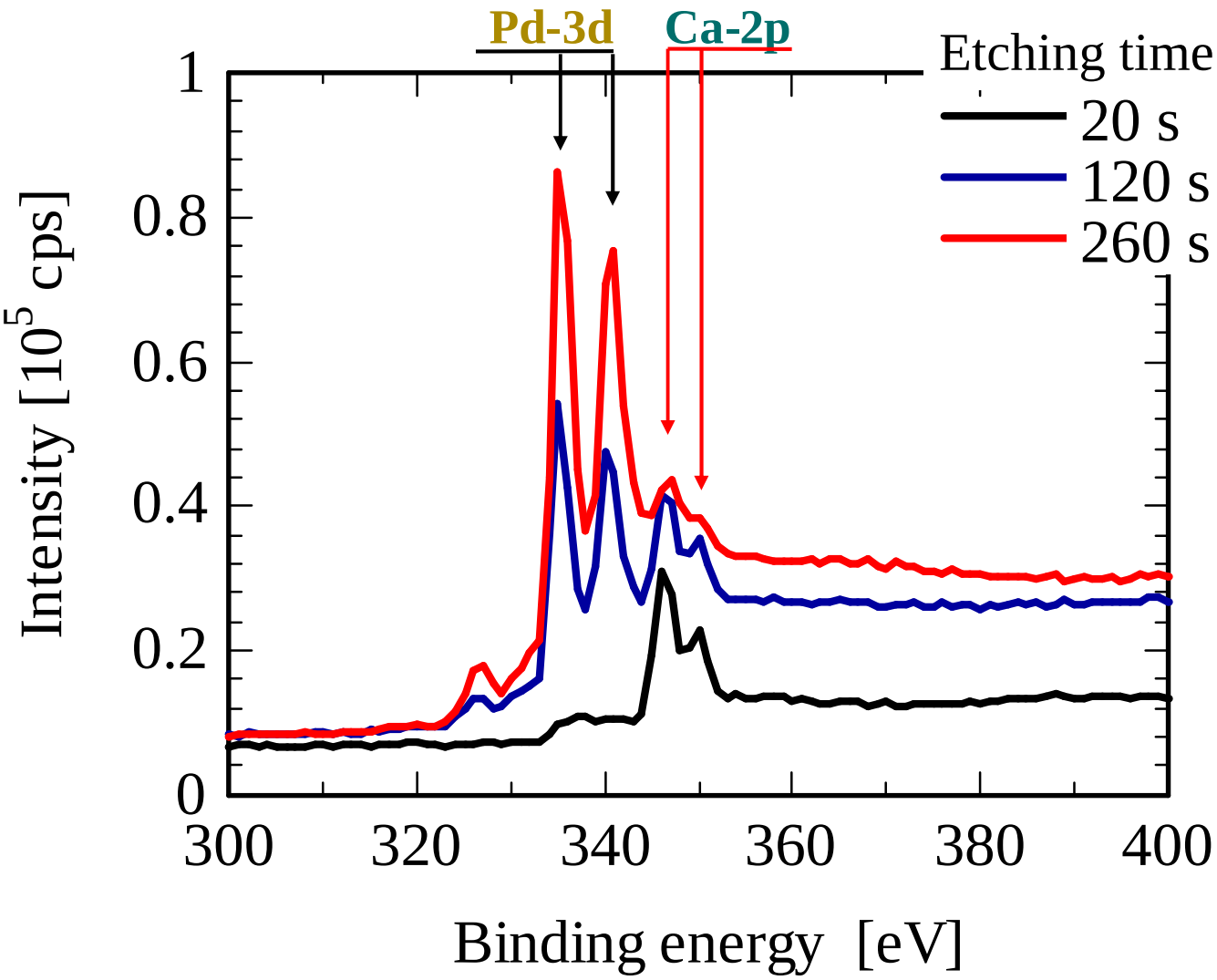


(2) Forming a layer of CaO on Sr/Pd ; RF sputtering deposition

RF sputtering condition

- Cathode (sputter target); CaO
- Anode (sample); Sr/Pd
- Ar pressure; 0.1 Torr
- RF power; 100 W
- Plasma exposure time; 5 - 30 min.
- Sample temperature; 450 K
- CaO deposition rate; 0.1 - 0.5 nm/min

(2D) XPS characterization after forming the layer of CaO on Sr/Pd



Change in the sample structure is reflected in XPS spectra after repetitive etching of the CaO/Sr/Pd surface.

- The CaO layer thickness x is calculated from the intensity ratio of these peaks, $Y_{\text{Ca-2p}}/Y_{\text{Pd-3d}}$;

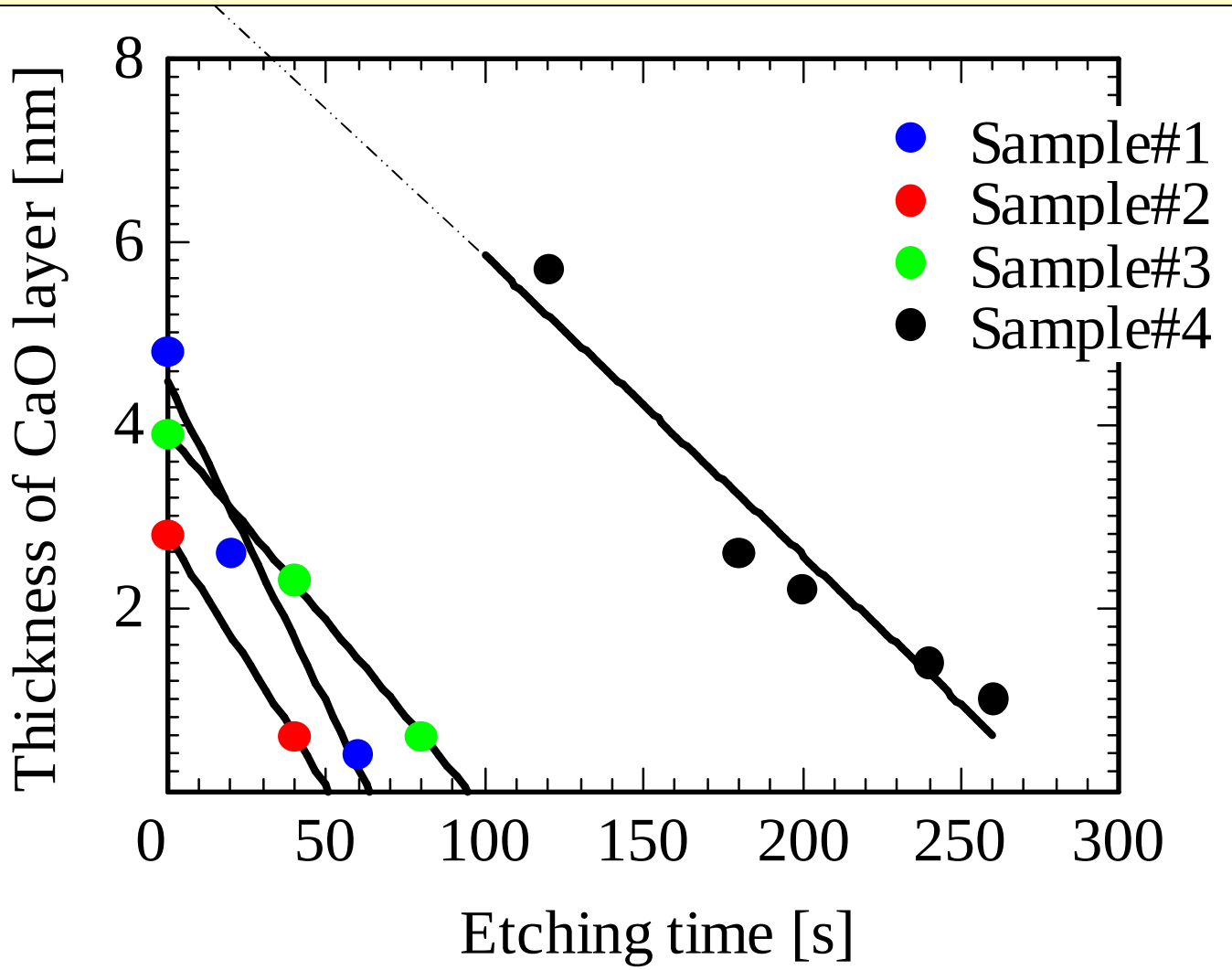
$$\exp\left(\frac{x}{\lambda_{\text{CaO}-\text{Pd}}}\right)\cdot\left(1-\exp\left(-\frac{x}{\lambda_{\text{CaO-Ca}}}\right)\right)=\frac{n_{\text{Pd}}\cdot\sigma_{\text{Pd}}\cdot\lambda_{\text{Pd-Pd}}\cdot Y_{\text{Ca-2p}3/2}}{n_{\text{Ca}}\cdot\sigma_{\text{Ca}}\cdot\lambda_{\text{CaO-Ca}}\cdot Y_{\text{Pd-3d}5/2}}.$$

where

$$Y_{\text{Ca\_2p}3/2}=\phi_{\text{p}}\cdot S\cdot n_{\text{Ca}}\frac{d\sigma_{\text{Ca}}}{d\Omega}\Delta\Omega\cdot\int_0^x\exp\left(-\frac{t}{\lambda_{\text{CaO-Ca}}}\right)\text{d}t,$$

$$Y_{\text{Pd-3d}5/2}=\phi_{\text{p}}\cdot S\cdot n_{\text{Pd}}\frac{d\sigma_{\text{Pd}}}{d\Omega}\Delta\Omega\cdot\exp\left(-\frac{x}{\lambda_{\text{CaO-Pd}}}\right)\int_0^\infty\exp\left(-\frac{t}{\lambda_{\text{Pd-Pd}}}\right)\text{d}t.$$

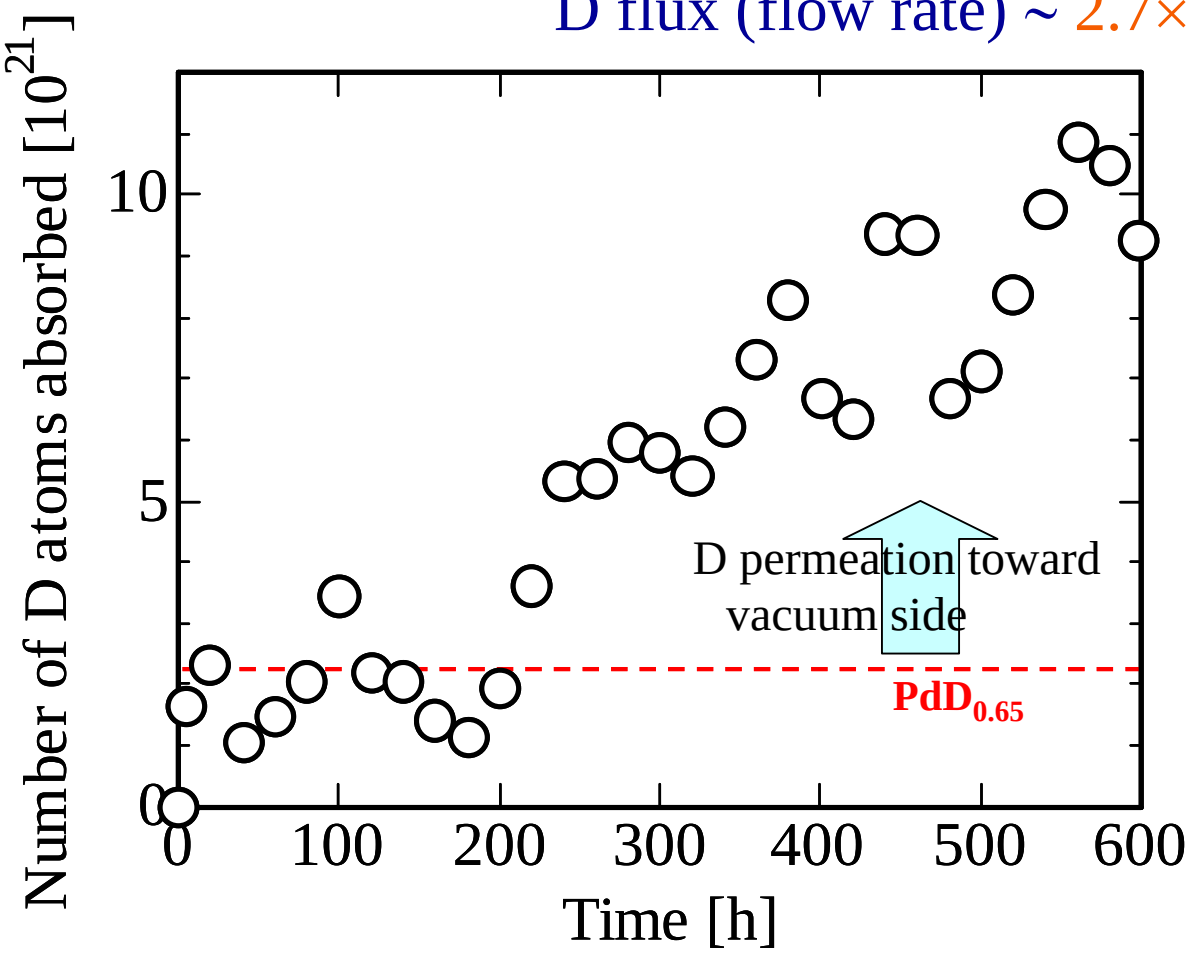
(2D) XPS characterization after forming the layer of CaO on Sr/Pd



The thickness of the CaO layer has been calculated from the XPS yield of the Pd-3d peak relative to that of Ca-2p to be about 10 nm for the sample #4.

(3) Permeation of D through CaO/Sr/Pd (the sample B)

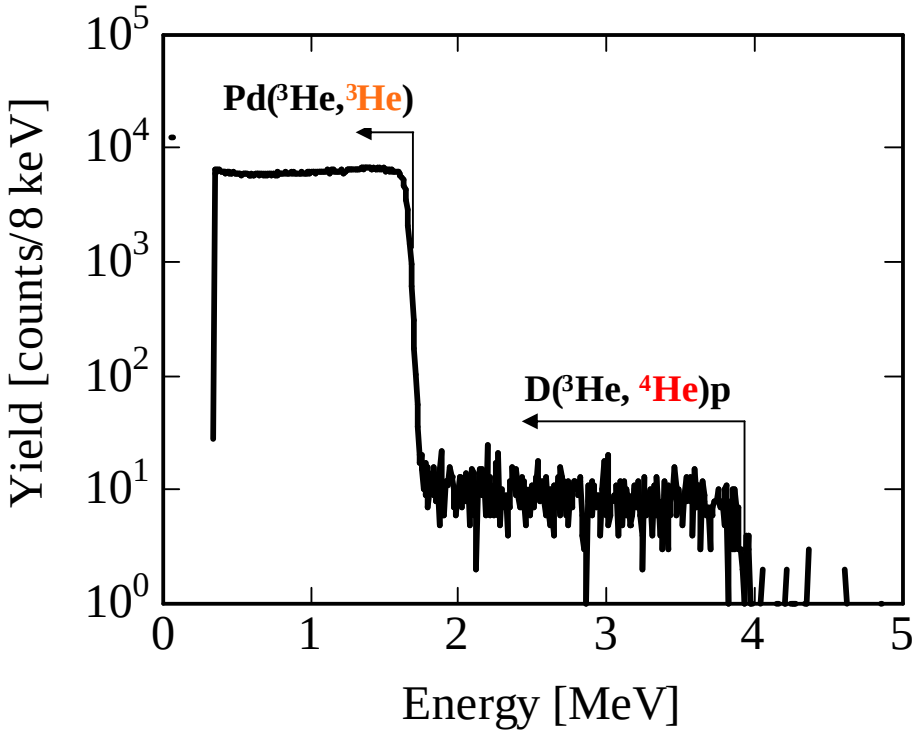
D flux (flow rate) ~ 2.7×10^{15} $cm^{-2}s^{-1}$
(=0.02 sccm)

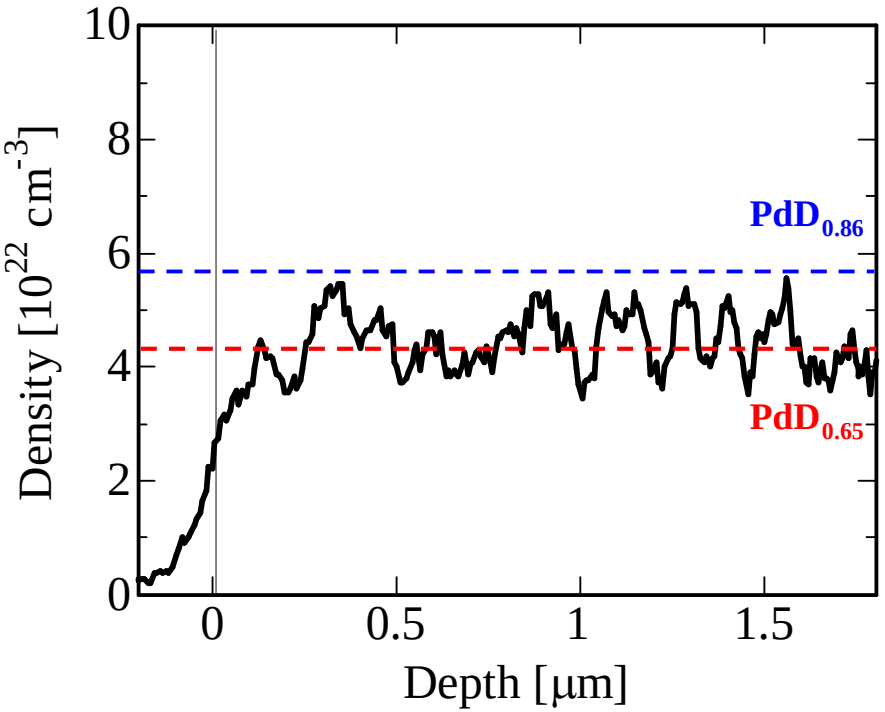


During D permeation, pressure in the reservoir has been monitored to calculate the number of absorbed/transmitted D atoms.

(3D) NRA during permeation of D through CaO/Sr/Pd (the sample B)

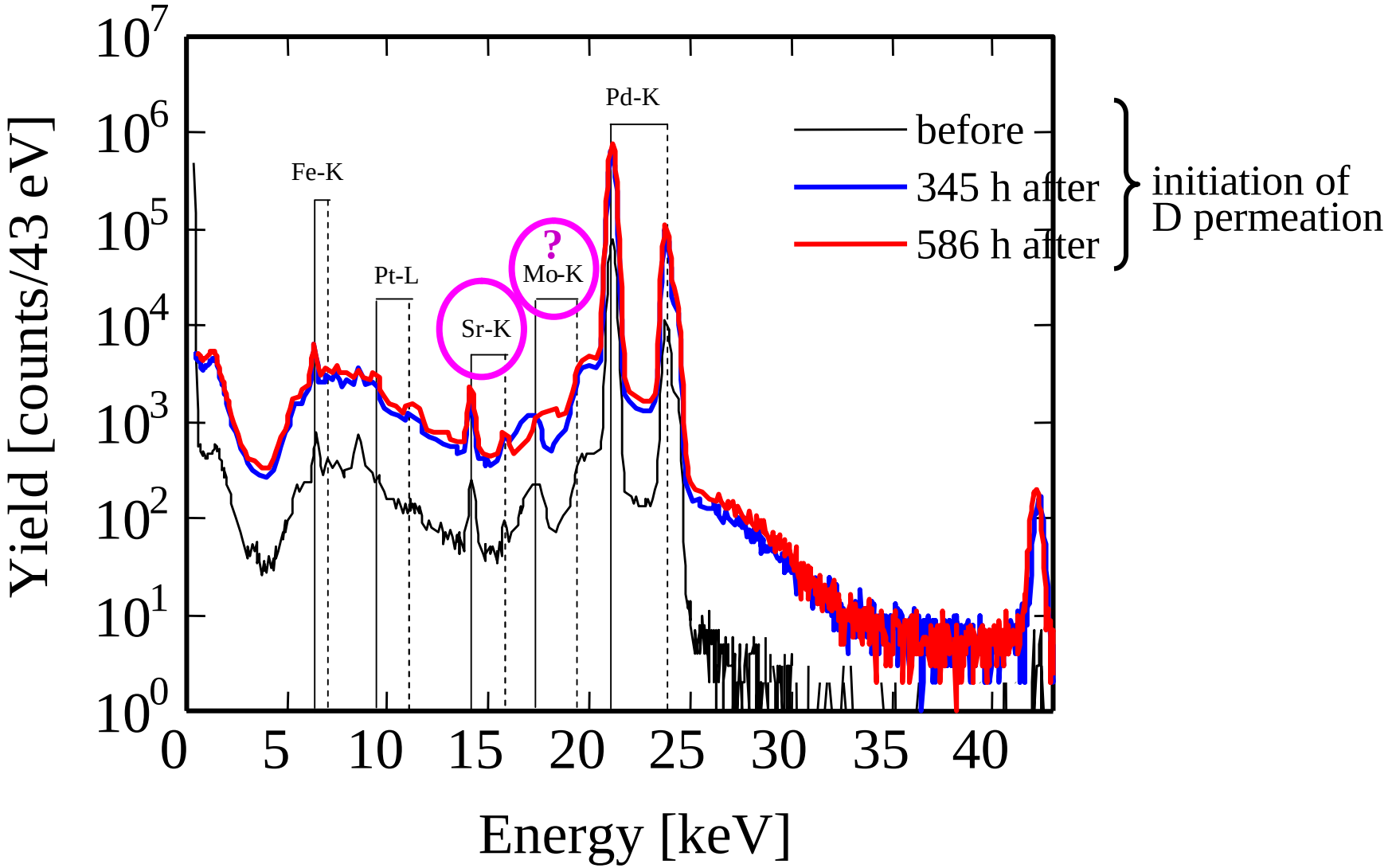
NRA using the D(^{3}He, ^{4}He)p reaction





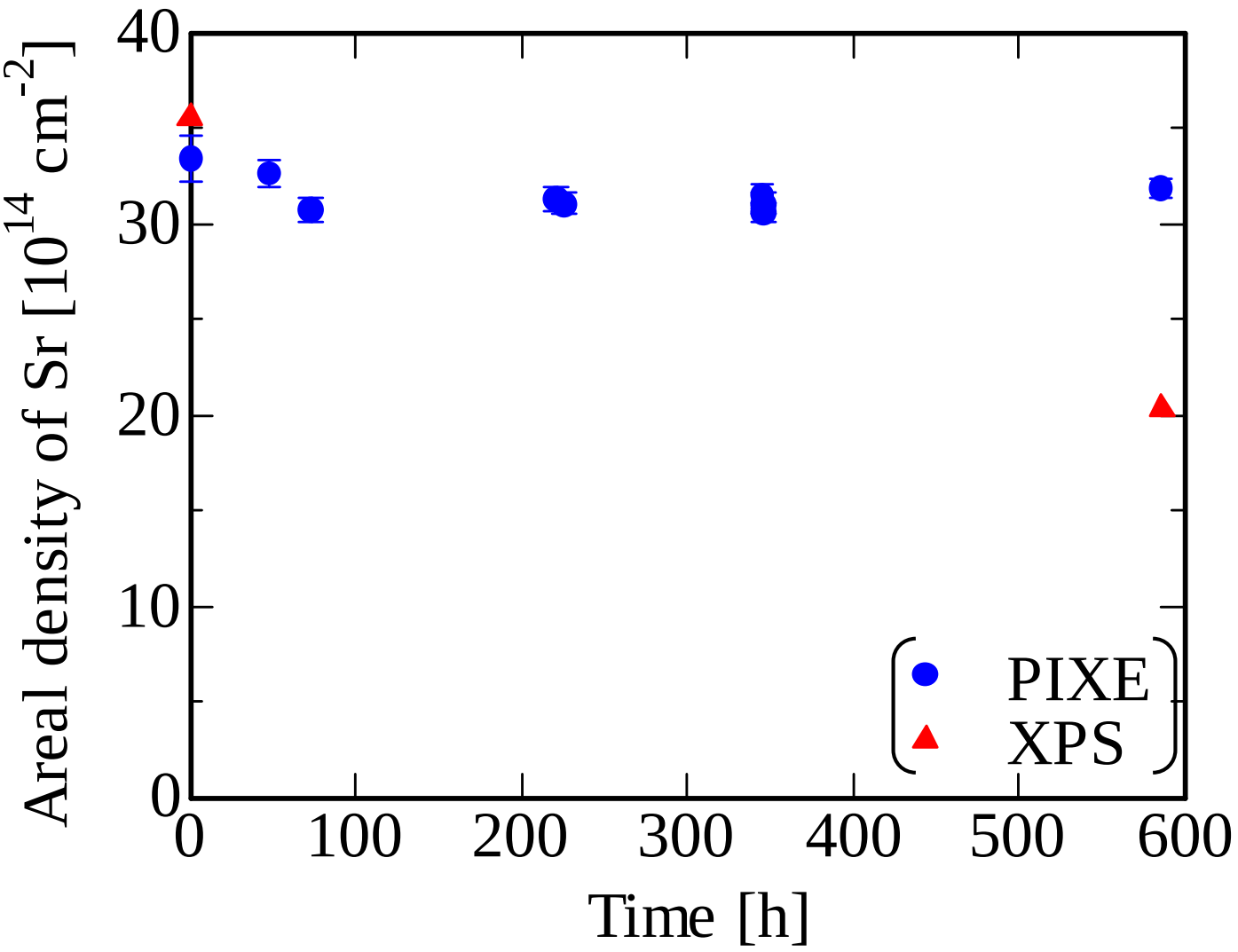
The NRA revealed the almost uniform composition of $PdD_{0.65}$ over the depth region of 0-1.8 μm.

(3D) PIXE analysis during permeation of D through CaO/Sr/Pd (the sample B)



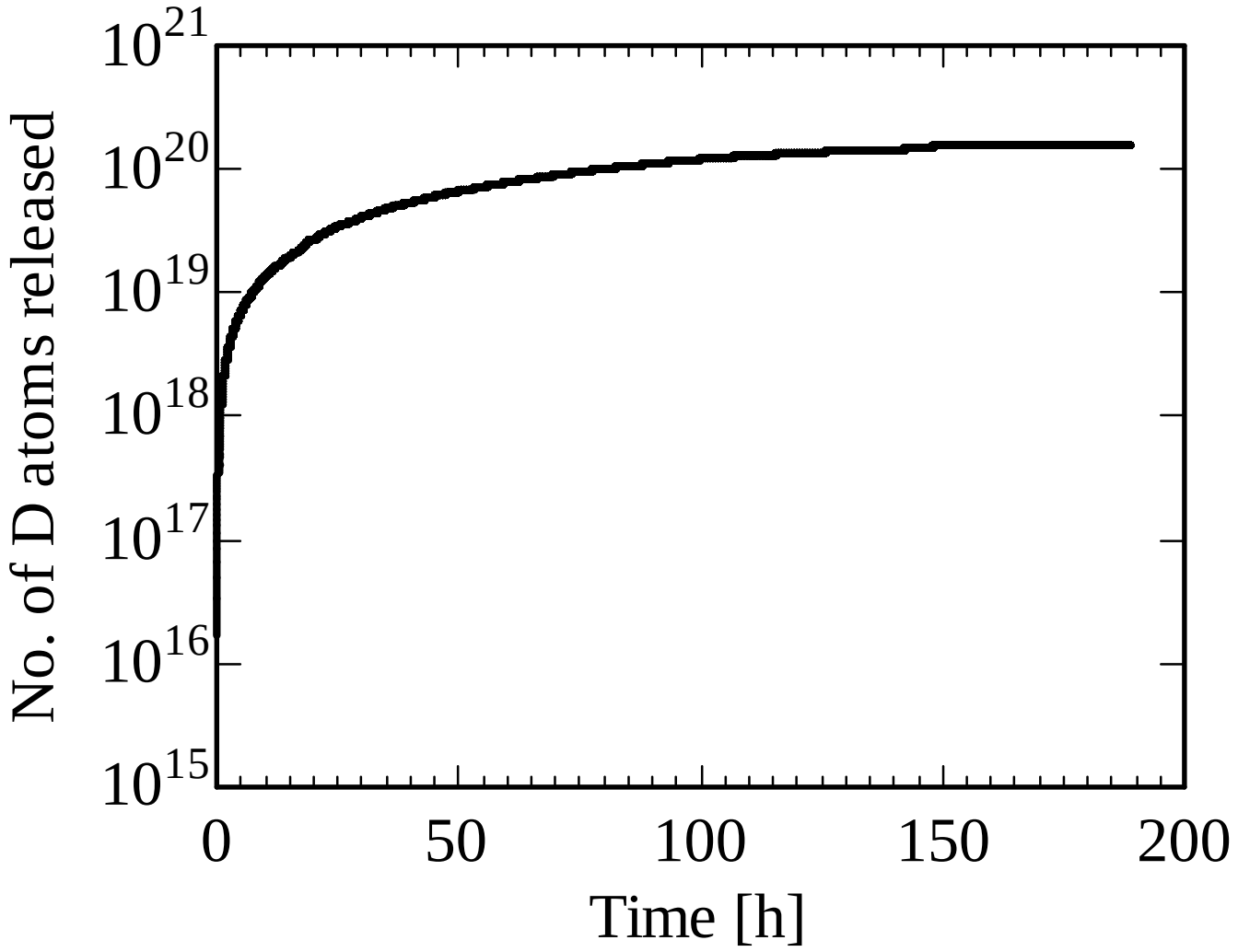
The PIXE spectra revealed existence of a variety of impurities.
The Mo-$K_{\alpha/\beta}$ seems to be interfered by unidentified broad peaks.

(3D) PIXE analysis during permeation of D through CaO/Sr/Pd (the sample B)



Variation of the areal density of Sr during permeation of D for 590 hours. Results of ex-situ XPS are also plotted for comparison.

(4) Desorption of D_2



Outgassing of the sample after finishing D permeation was necessary for introduction into the XPS chamber. Total amount of desorbed D atoms measured with QMS indicates D/Pd~0.3.

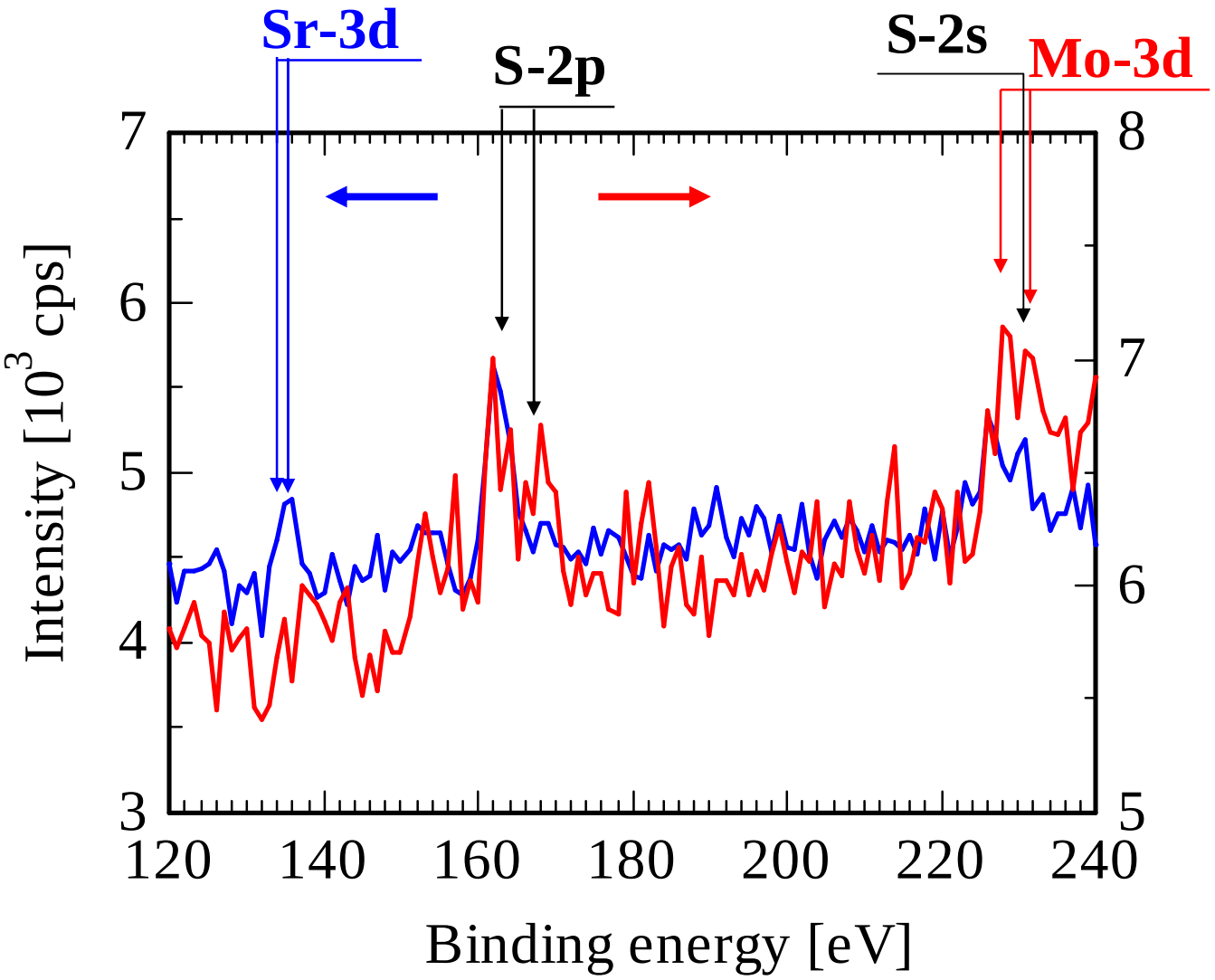
- The process of deuterium charging is described by a 1-dimensional solution of the diffusion equation for the sample with a thickness and area of a and S, respectively.
- One side of the sample is assumed to be opaque for deuterium, while the other facing to hydrogen gas to give a boundary condition for the deuterium density; $n(a) = n_0$.
- The number of deuterium atoms absorbed in the sample, $N_a(t)$, is given as a function of time by

$$N_a(t) = aSn_0\left[1-\sum_{s=0}^{\infty}\left\{\frac{8}{(2s+1)^2\pi^2}\cdot\exp\left(-\left(\frac{(2s+1)\pi}{2a}\right)^2 Dt\right)\right\}\right]$$

- A characteristic time $t_{1/2}$ for the number of deuterium atoms absorbed to reach a half of the saturation value, *i.e.*, $N_a(t_{1/2})/aS = N_a(\infty)/2aS = 2.9\times10^{22}$ cm^{-3}, is therefore given by

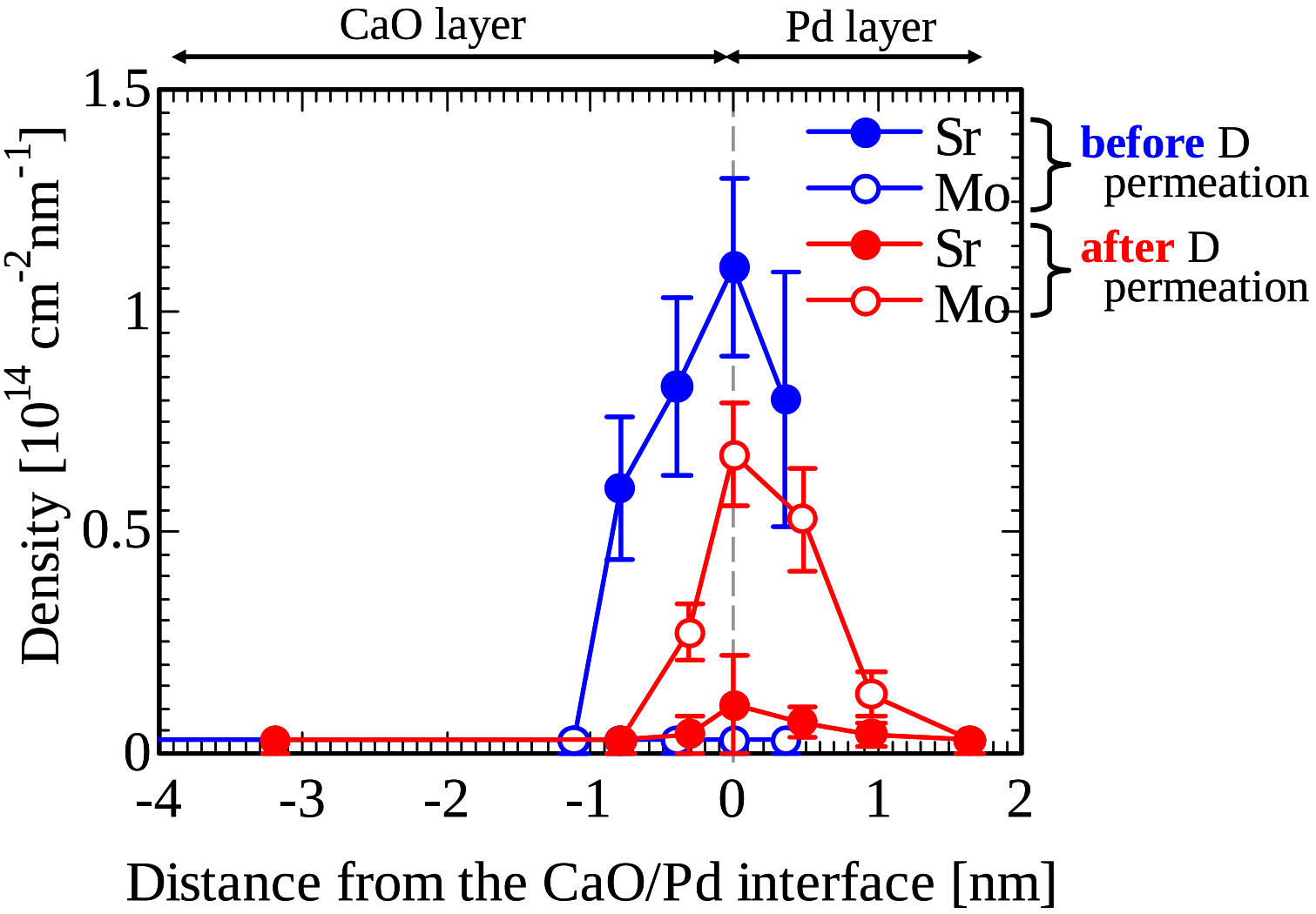
$$t_{1/2} = \frac{0.1967a^2}{D} = 6\times10^1 \text{ s}$$

(4D) XPS characterization of the sample A after D_2 Desorption



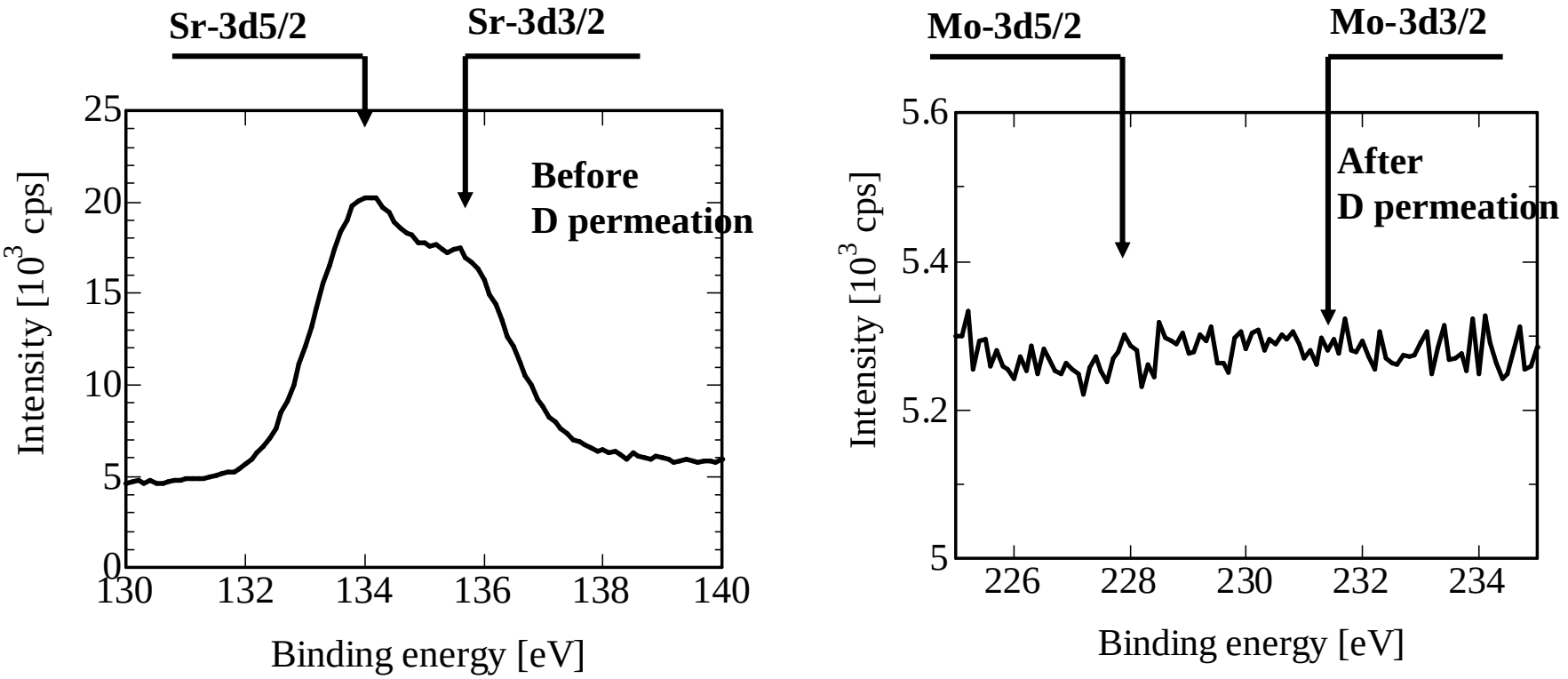
XPS spectra before and after D permeation. Increase in Mo-3d peaks in exchange for decrease in Sr-3d peaks is observed.

(4D) XPS characterization of the sample A after D_2 Desorption



XPS analysis implies nuclear transmutation near the CaO/Pd boundary: Sr atoms with areal density of 1.3×10^{14} cm^{-2} appear to be transformed to Mo atoms of 6.6×10^{13} cm^{-2} by D permeation.

(4D) XPS characterization of the sample B after D_2 Desorption



An order of magnitude increase in areal density of Sr (~2 monolayers) was realized by RF sputtering method. However, nuclear transmutation was not recognized for this sample.

Summary of changes in areal densities

	PIXE			XPS	
	Sr	**Mo**	**Others**	**Sr**	**Mo**
A (electroplating)	**-----**	**-----**	**-----**	**–1.3**	**+0.66**
B (I. B. sputtering)	**–2.5**	**<<+1.9**	**<<+1**	**–15**	**<<+0.11**
	areal densities in 1E14 cm^{-2}				

Sample A: Transmutation efficiency was ~50%.

Sample B: Decrease in areal density of Sr was not followed by consistent increase in other elements observable.

Summary

(1) Implication of nuclear transmutation Sr →Mo has been obtained in a system [vacuum/CaO/Sr/PdD$_x$/D$_2$], which is a little simpler than that used by Iwamura *et al*.

(2) The diagnostic method used to identify the elements was conventional XPS, giving the areal densities of 1.3×10^{14} cm^{-2} (Sr) and 6.6×10^{13} cm^{-2} (Mo).

(3) Extended analytical methods have been prepared; *in situ* and simultaneous PIXE, RBS and NRA/ERDA for areal densities of transmutation elements and deuterium distribution.

(4) Increase in the areal density of Sr to 3.3×10^{15} cm^{-2} by using ion beam sputtering method resulted in negligible transmutation yield.

Summary

(5) Further experimental work is necessary to confirm the phenomena;

-Dependence on the density of the atoms to be transmuted,
the areal density of the atoms,
the flux (flow rate) of D,
the sample structure,
…….

-Effect of surface contamination.