Progress on Transmutation Experiments induced by D_2 gas permeation

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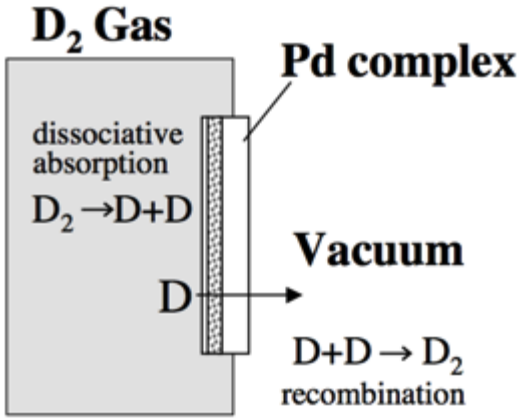
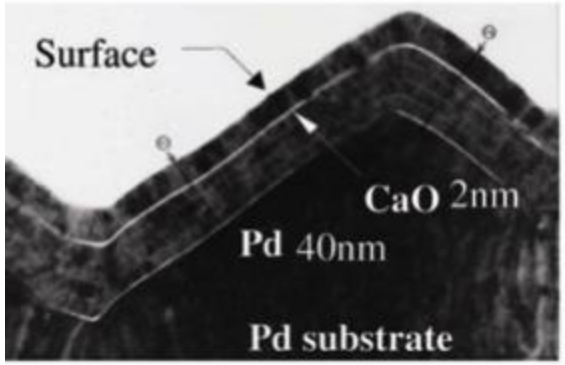
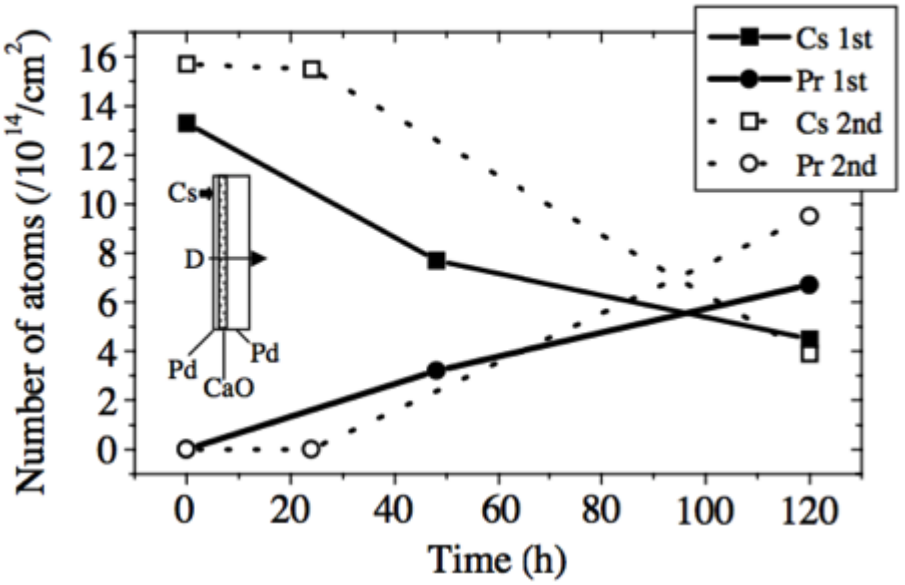
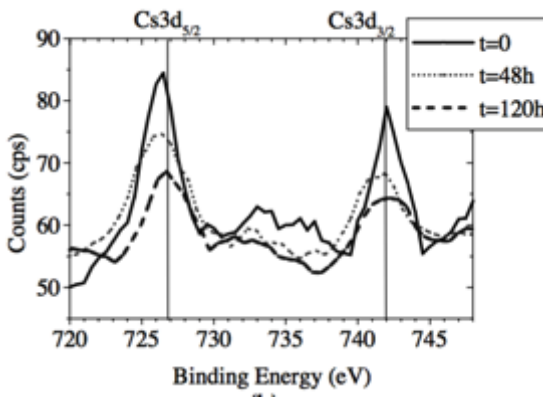
Cs → Pr Transmutation MHI Experiments

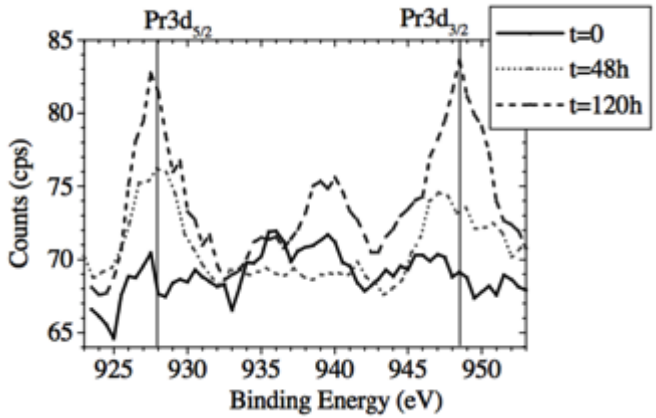
$$^{133}_{55}Cs \rightarrow ^{141}_{59}Pr$$











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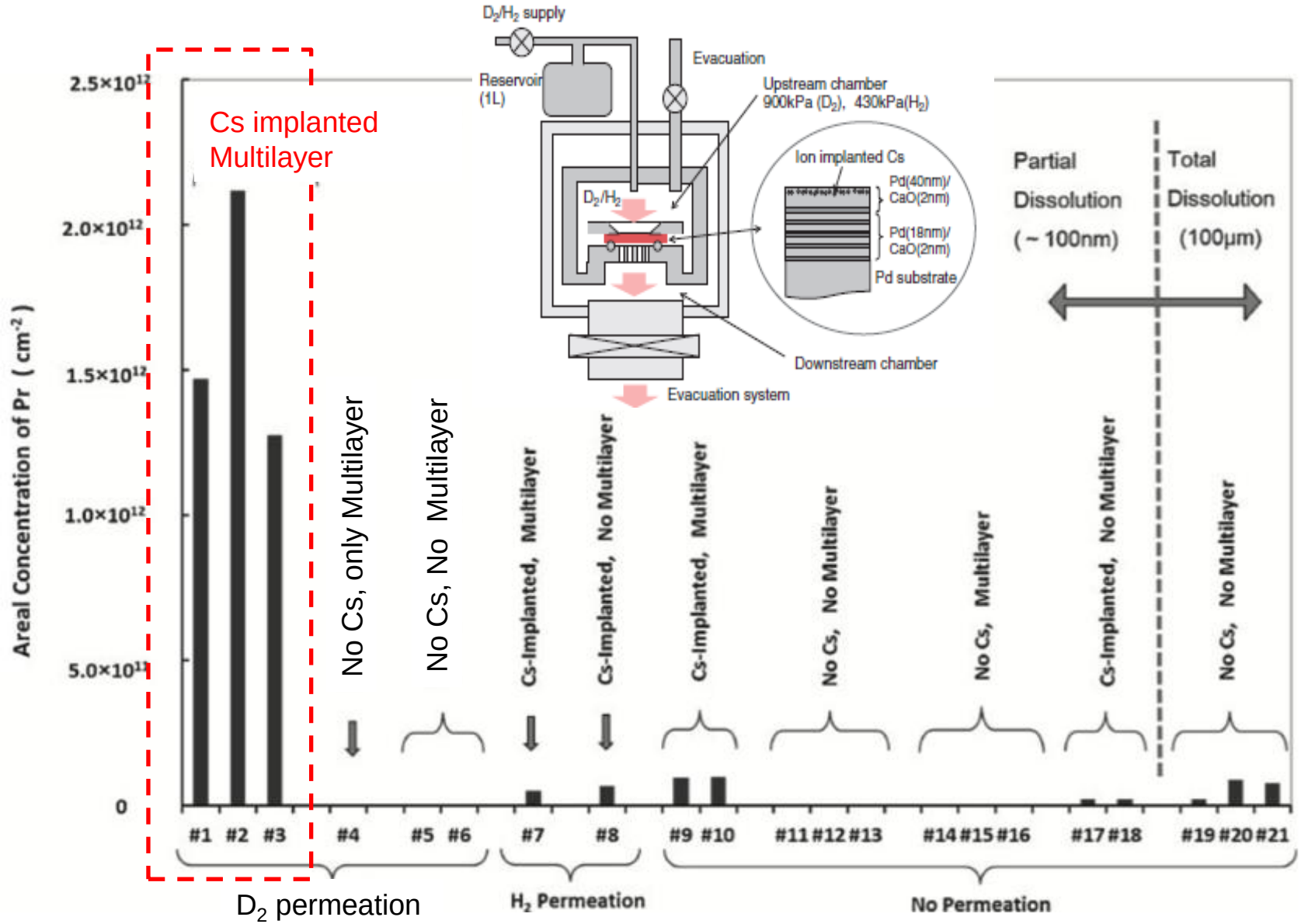
Replicated experiment of Toyota lab



Hioki et al. JJAP 52 (2013) 107301

Object

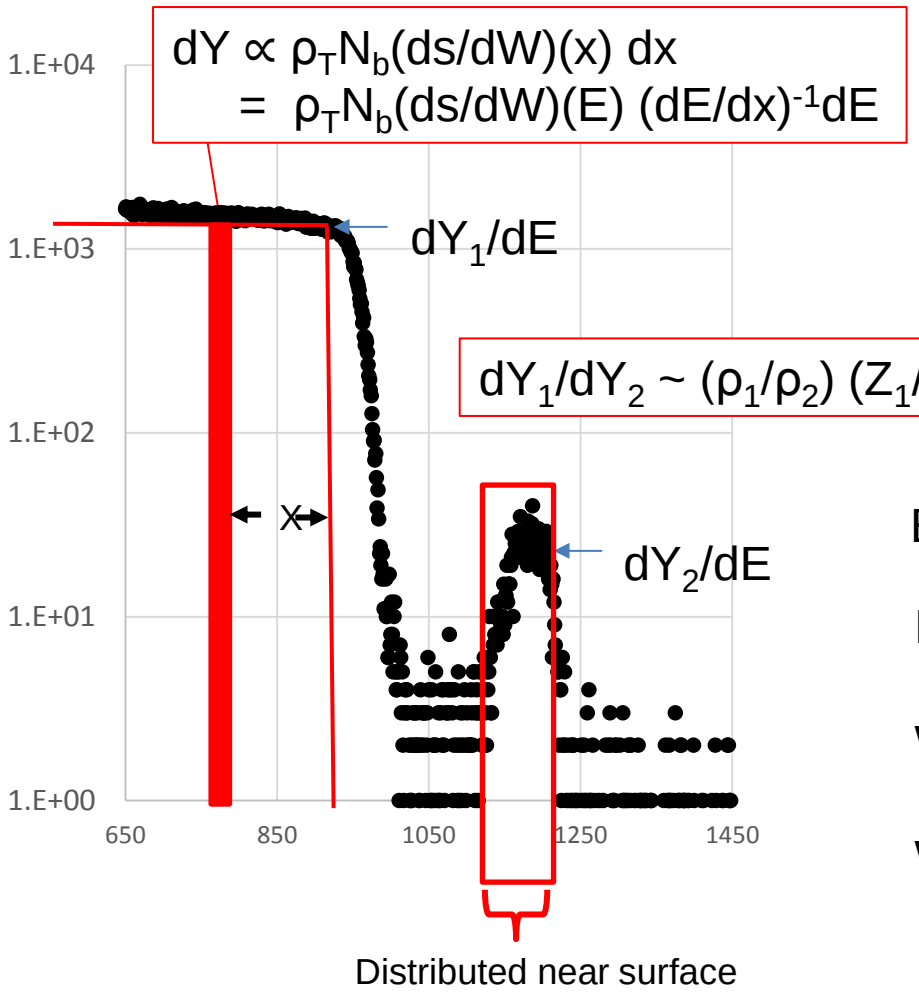
Successive various measurements showed existencs of ^{141}Pr
XPS, ICP-MS, Tof-SIMS, In-situ XRF at SPring8

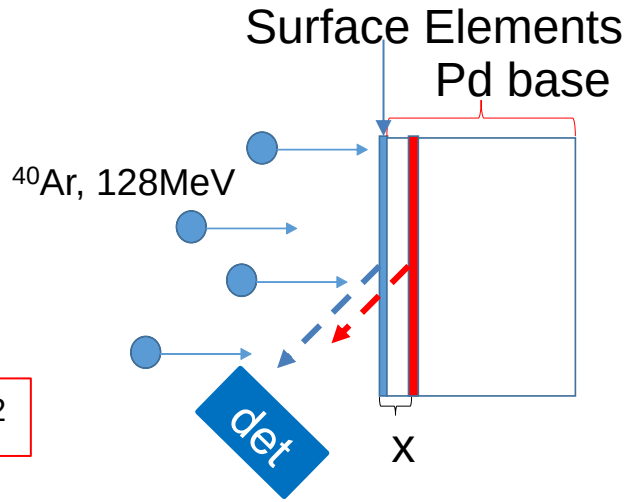
There is a criticism that the mass of the nucleus was not identified, since the employed mass spectrometry, such as ICP-MS, cannot exclude possibilities of chemical compound objects. Direct assignment of the nucleus (141Pr) by using nuclear physics approach is highly demanded.

Therefore, we employed the Rutherford Backscattering Spectroscopy (RBS) to identify ^{141}Pr for direct nuclear mass assignment.

Rutherford Backward Scattering (RBS)

Scattered by Nucleus; Mass determined by kinematics
Advantage for large A and Z





Example: Compare W on surface / Pd

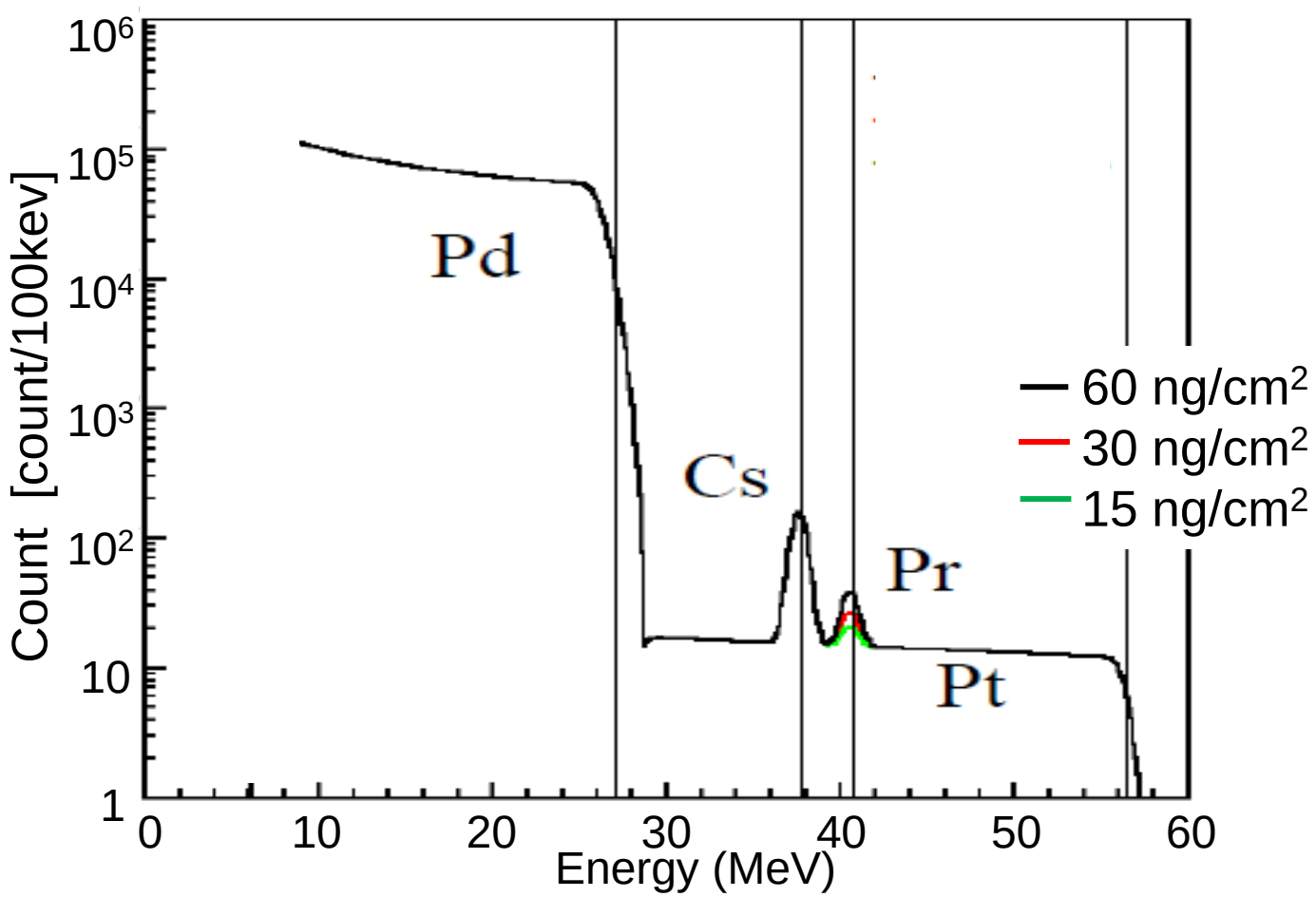
Pd density : 6.8×10^{22} atoms/cm^3
$(Z_{Pd}/Z_W)^2 = (46/74)^2 = 0.386$
W is more sensitive than Pd by 2.6 times

W density : ~ 5.4×10^{20} atoms/cm^3

Expected RBS Spectrum by irradiation 128-MeV ^{40}Ar

(1) Pt impurity in thick Pd :100 ppm
(2)^{133}Cs:uniformly distributed in 0 - 20 nm depth; 200ng/cm2
(3)^{141}Pr: uniformly distributed in 0 - 10 nm depth;

— 60 ng/cm^2 — 30 ng/cm^2 — 15 ng/cm^2



Cs transmutation sample by D_2 gas Permeation

Washing a Pd Sample with Acetone

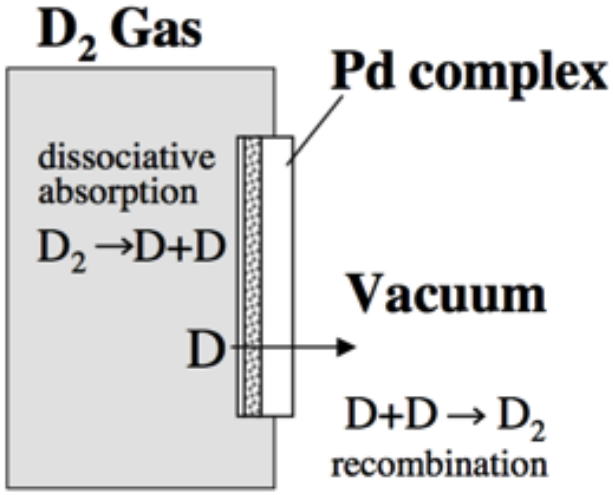
900℃ 10H Annealing under Vacuum Condition

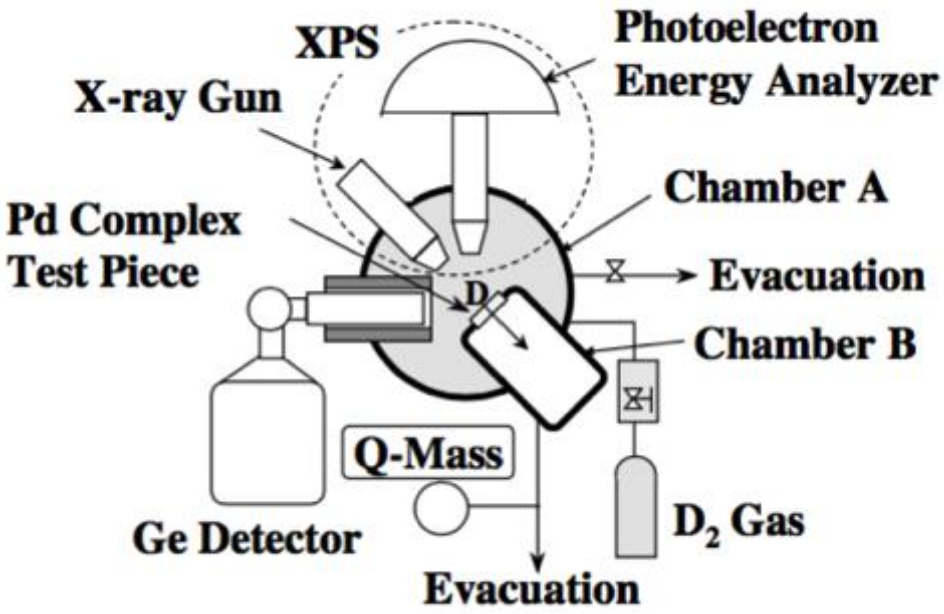
Washing the Sample with Aqua Regia

Fabrication Pd-CaO Multilayer using Ion Beam Spattering

Cs Deposition

D2 Gas permeation



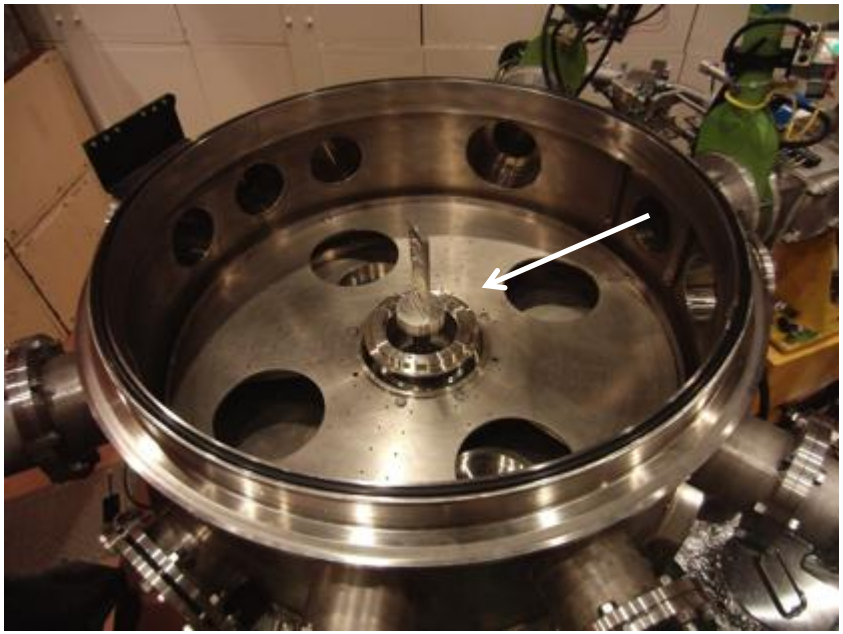


Sample for RBS was provided by MHI

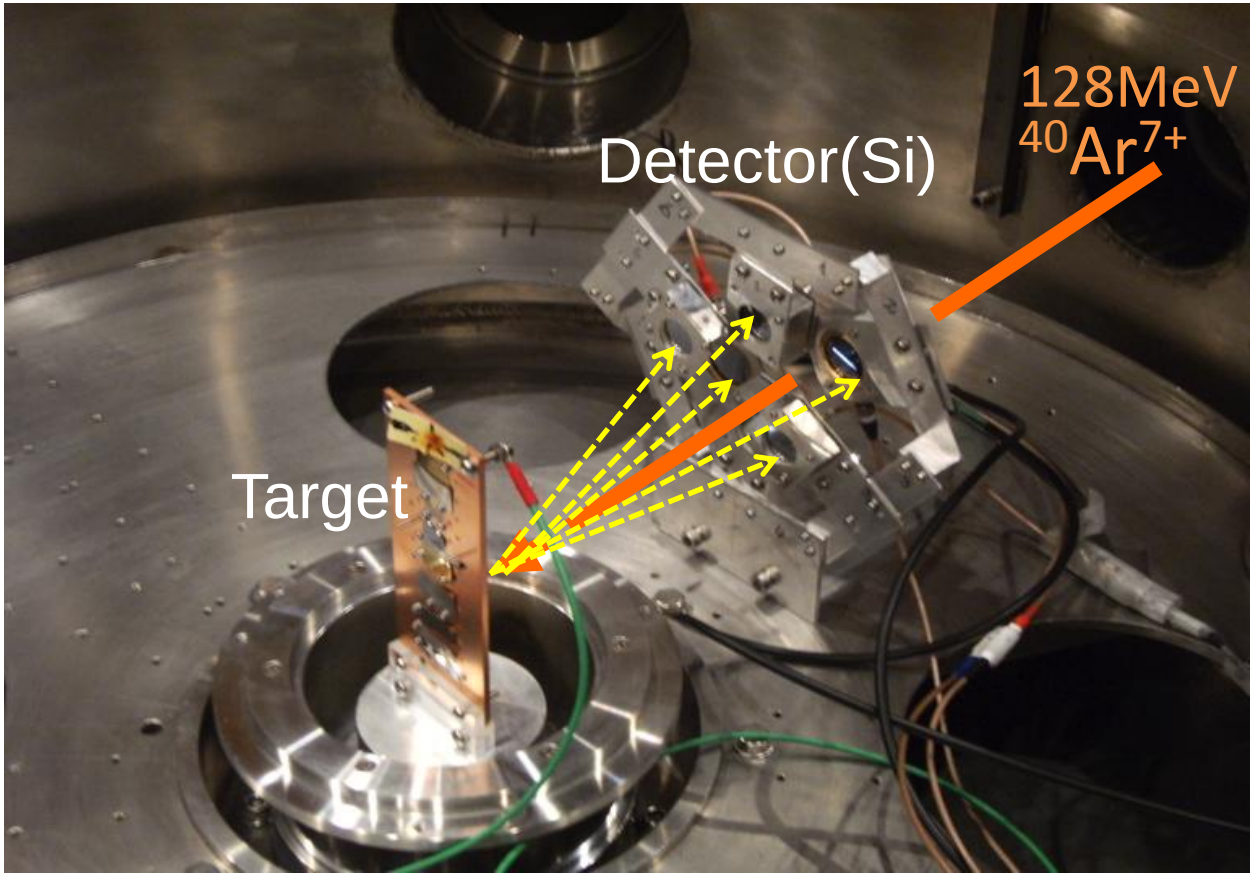
Cyclotron Radioisotpe Center, Tohoku University

ECR ion source + 930 AVF Cyclotron

Large Scattering chamber



Geometric Arrangement of Beam, Target and detector



Beam:

$^{40}Ar^{7+}$ 128MeV

~ 1 particle nA

Si detector:

300μm in thickness

300 mm^2x3, 450 mm^2x2

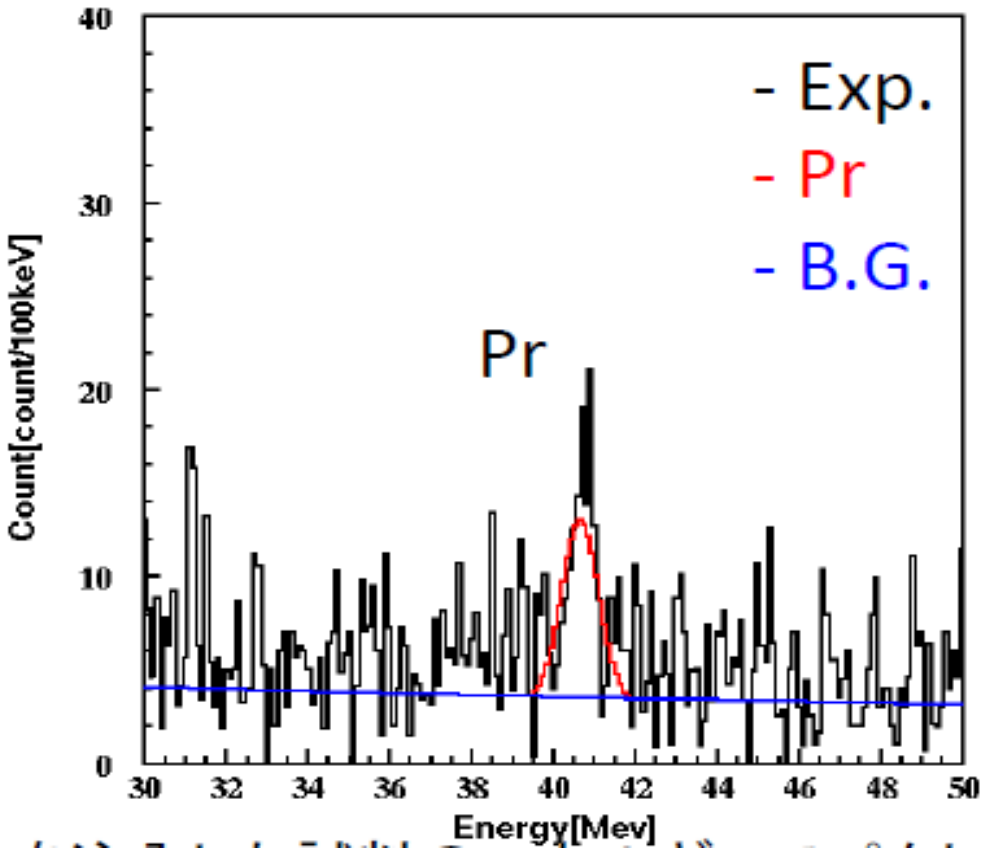
$\Theta = 165°$

total $\Delta\Omega = 0.083$ sr

Several-hour measurement for a sample to identify Pr of 10 ng/cm^2

Sensitivity of Pr identification

RBS spectrum Pr-implanted Pd/CaO foil



Pr

Implanted：23.4 ng/cm^2

Measured：18.9±4.8 ng/cm^2

Reproduced well !!

Pt BG: ~ 95 ppm of Pd

By measuring several hours,10 ng/cm^2-Pr can be detected with the confidence level of 99%　(or statitical significance of 2.6σ).

＊test peace was provided by Toyota Central Research and Development Laboratories

Sample Target

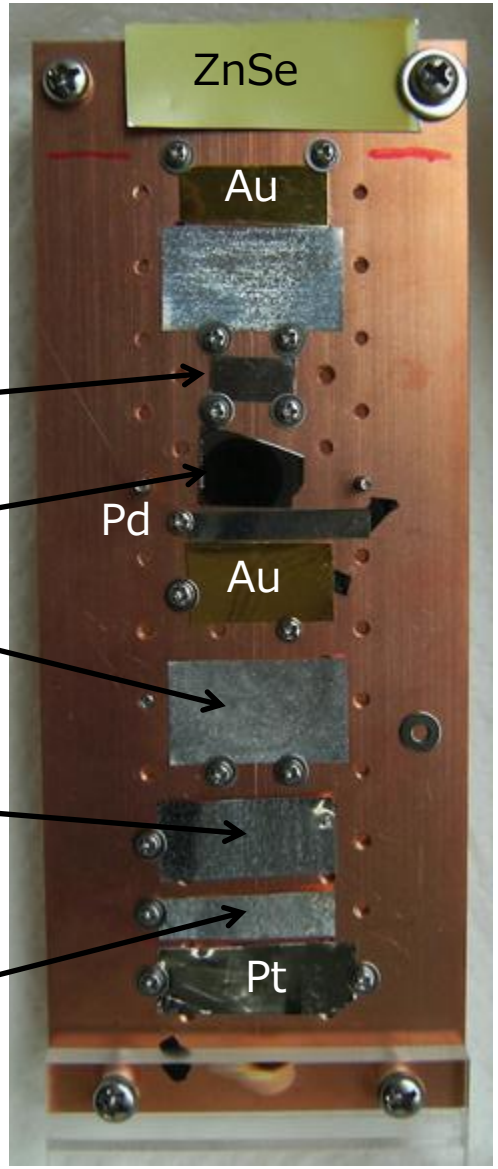
test peace
Pr Implantation 10^{14}
Pr:23.4ng/cm^2

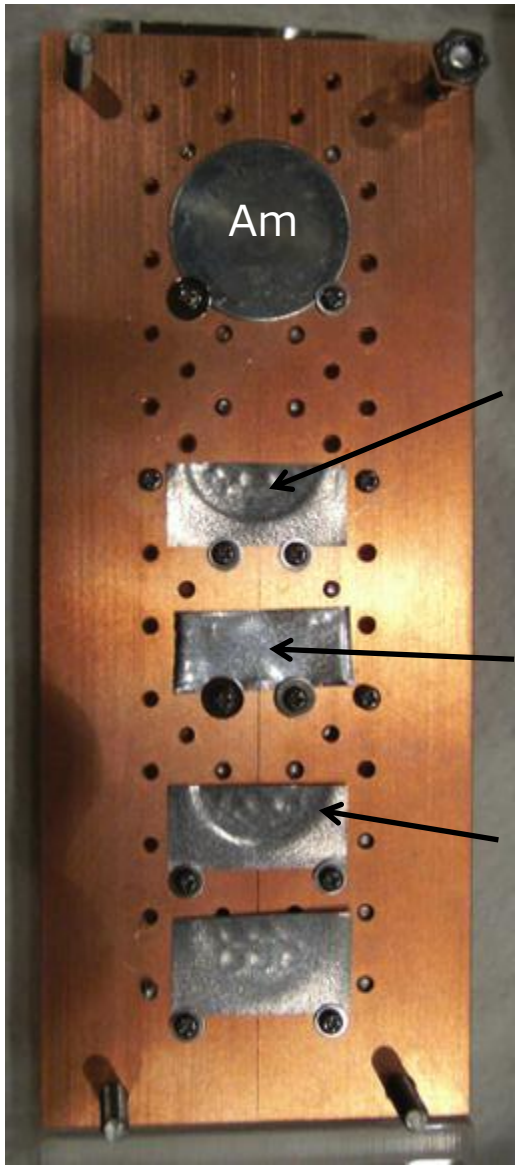
Au(10nm)/Si

FG sample（SK137）
Cs:134ng/cm^2
Pr:74ng/cm^2

Ref Sample
(Only Multilayer)

test peace
Cs Implantation





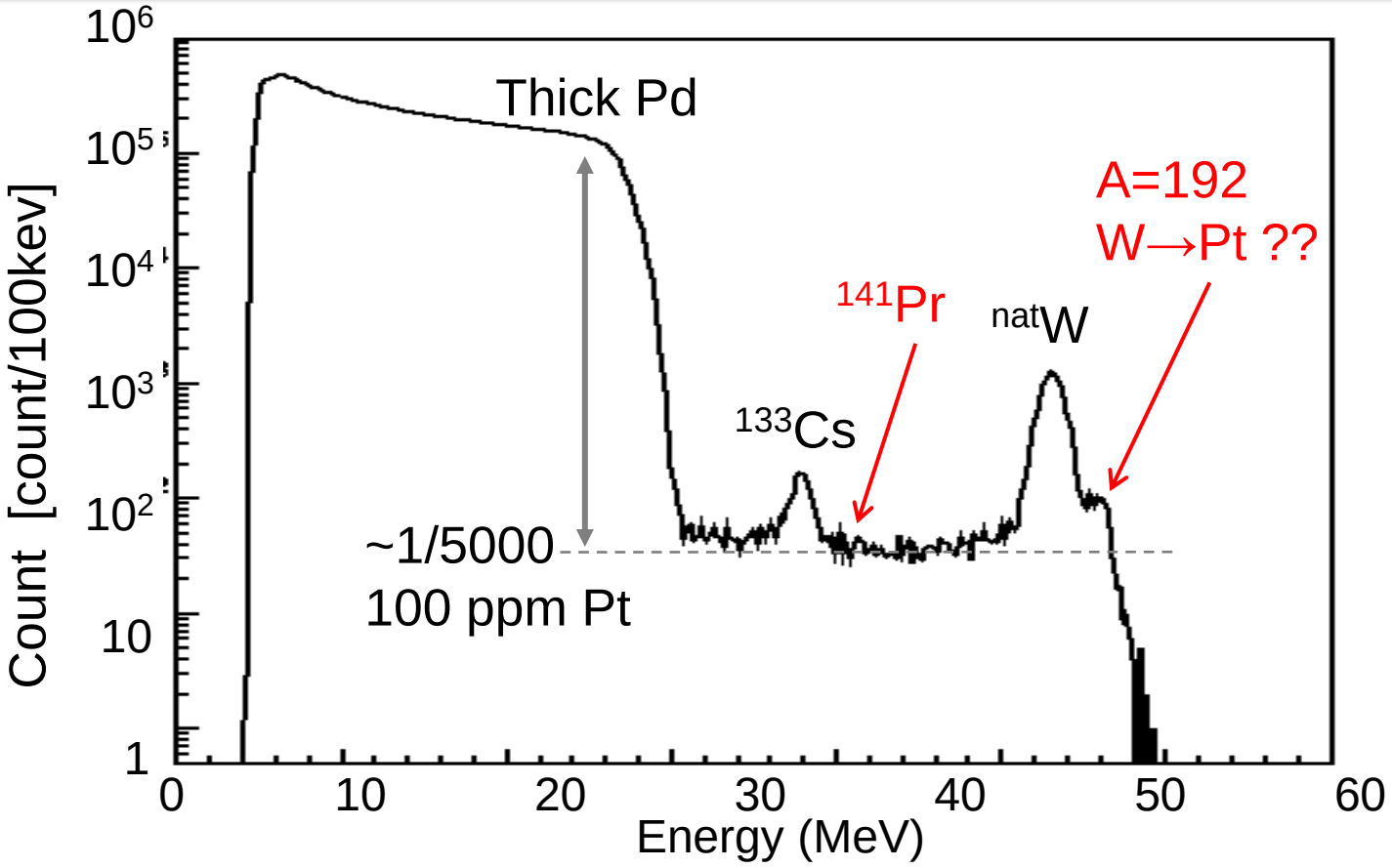
FG sample（SK132）
Cs: 94.6 ng/cm^2
Pr: 51 ng/cm^2

Ref Sample
(Cs Dope,
No D_2 Permeation)

FG sample（SK130）
Cs: 57.7 ng/cm^2
Pr: 50.8 ng/cm^2

RBS spectrum : with D_2 gas permeation

Summed spectrum of foreground samples



BG: mainly Pt impurity in thick Pd ~ 100 ppm→BG determine a detection limit.

^{133}Cs : distributed 0 - 80 nm in depth

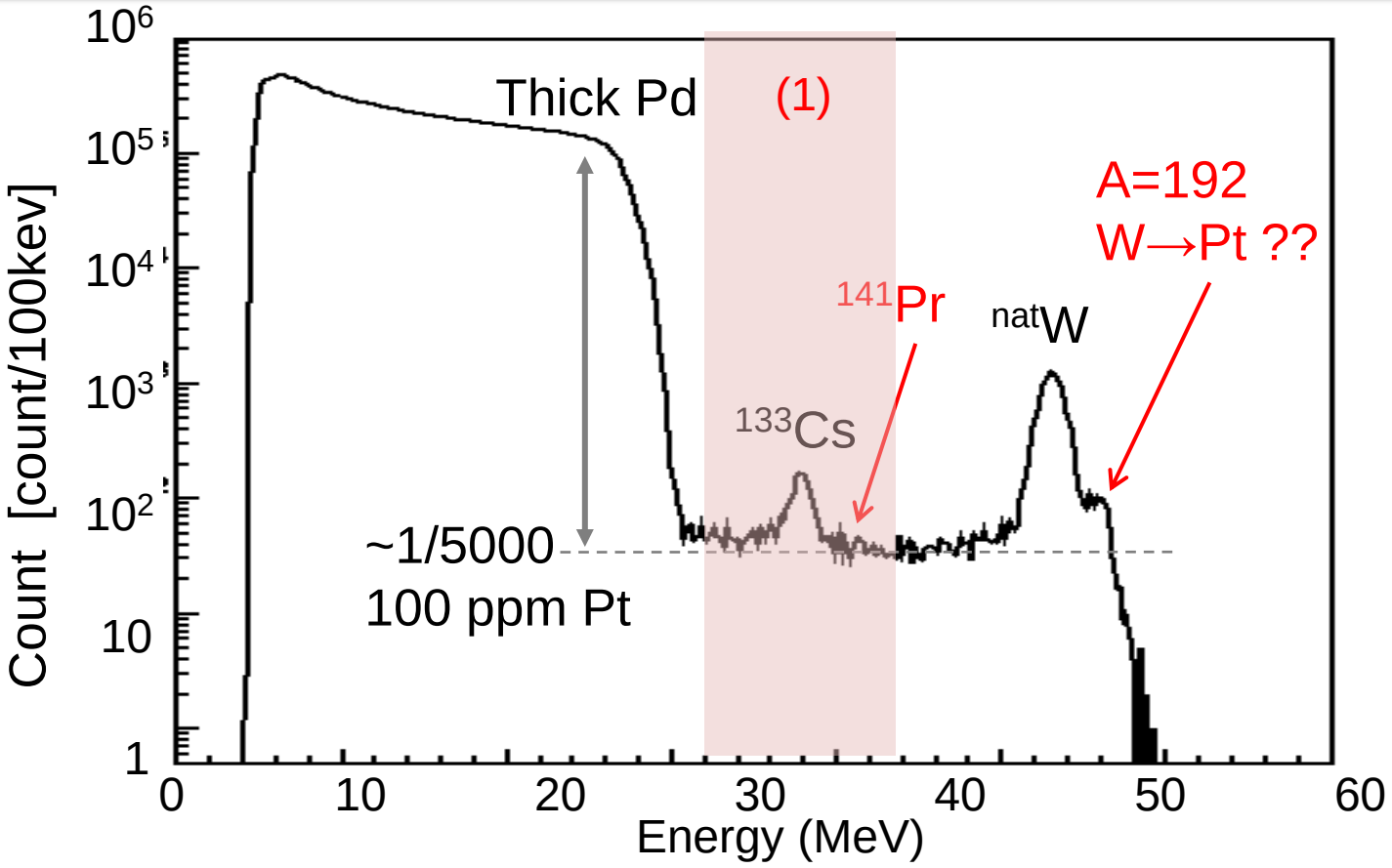
^{nat}W: distributed 0 - 80 nm in depth contamination of W (182,183,184,186) of multilayer

(1) A tiny peak at A=141 (^{141}Pr)

^{141}Pr exists on the surface. not observed for without D_2 ➡ consistent with MHI report ➡

RBS spectrum : with D_2 gas permeation

Summed spectrum of foreground samples



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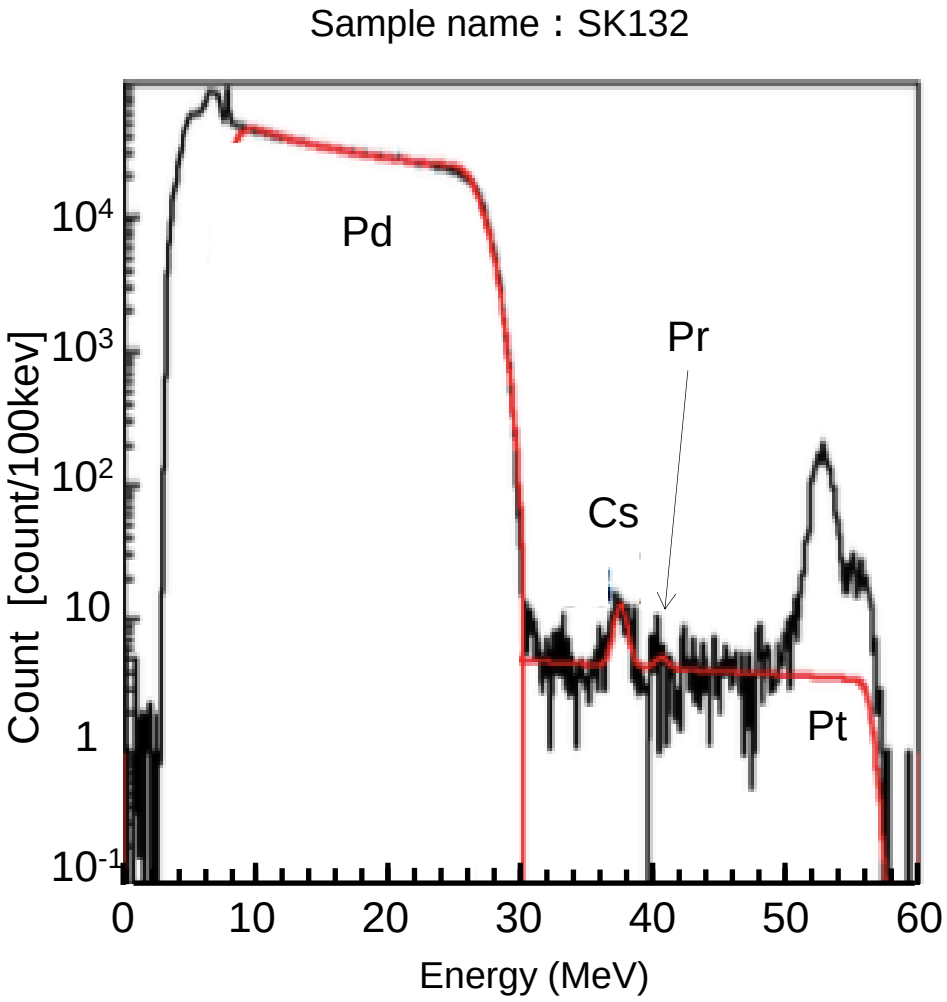
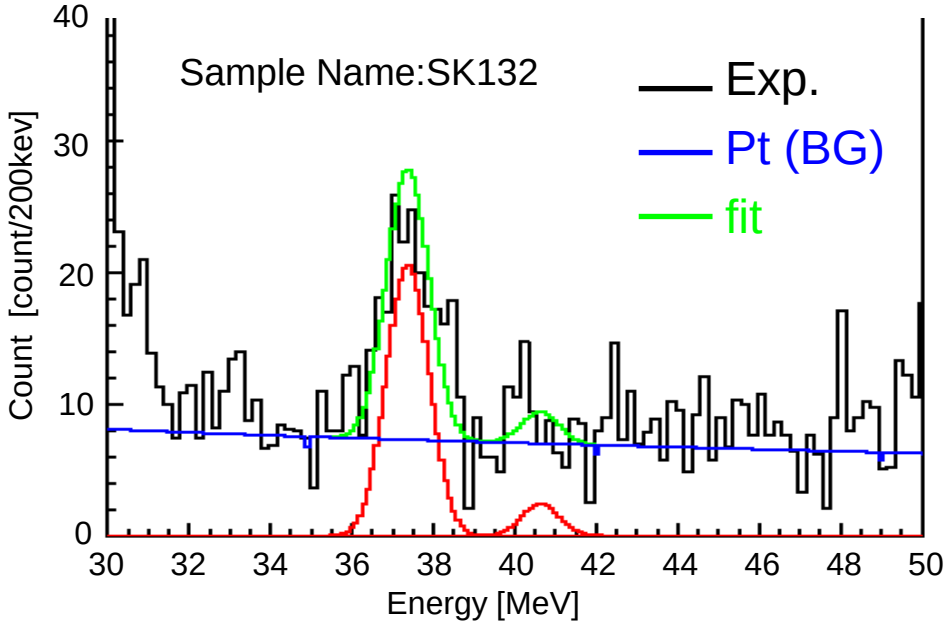
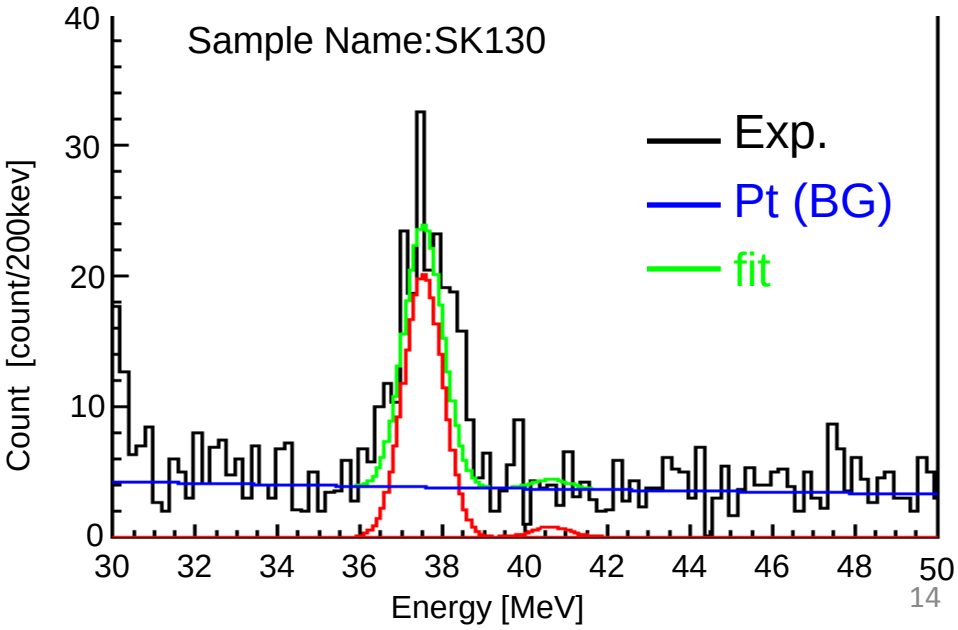
RBS spectrum with D_2 gas permeation: Cs, Pr detection







Pr amount comparison of ICP-MS and RBS

Sample Name	Analytical method	Cs(ng/cm$^{2)}$	Pr(ng/cm^2)
SK130	ICP-MS	57.7	50.8
	RBS	94.1±10.9	< 6
SK132	ICP-MS	94.6	51
	RBS	37.8±5.9	< 5.1±2.6
SK137	ICP-MS	134	74
	RBS	19.7±7.8	< 11
Pr test Sample	Pr Implanted		23.4
	RBS		19±5

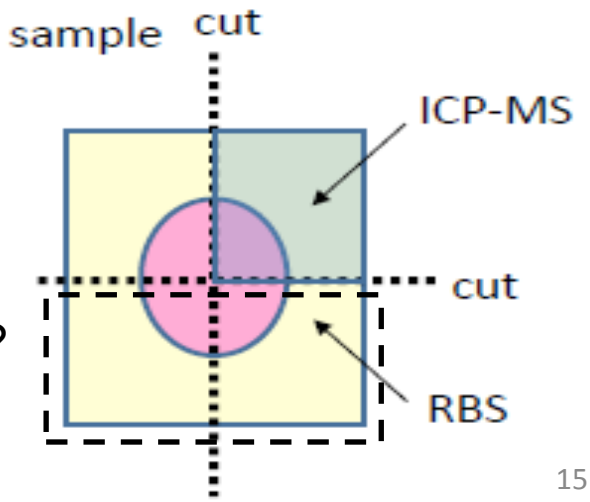
Comparing between the results of ICP-Ms and RBS, quantitative agreement is not good.

Y(RBS)/Y(ICP) = 0.15 ~ 1.63 for ^{133}Cs

< 0.15 for ^{141}Pr

→ Measurement points of ICP-MS and RBS are different ?

→ may be due to the local distribution of ^{141}Pr

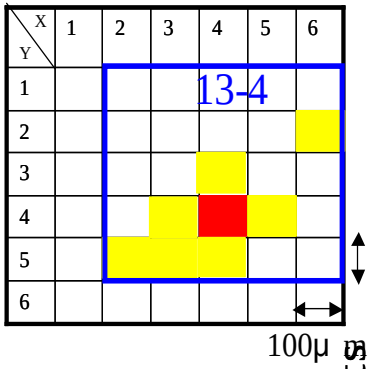


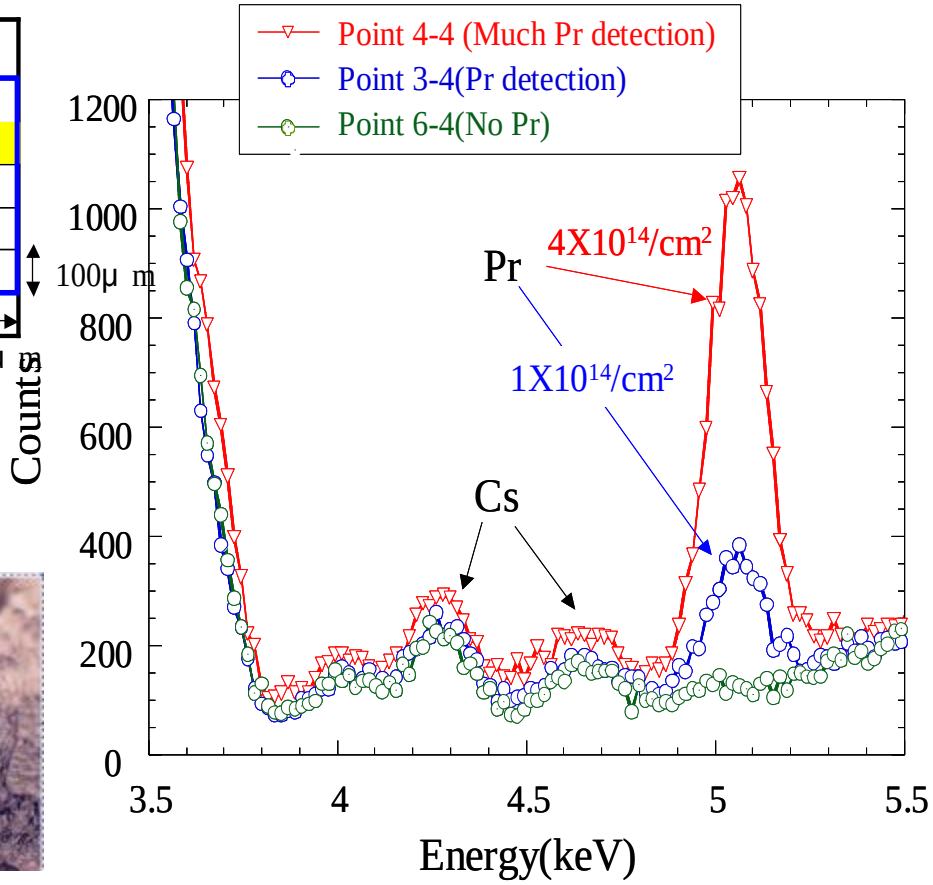
Non-uniform distribution of Pr (XRF @ SPring-8)

XRF study shows: Existing probability of Pr strongly depends on location

Average value of Pr in 13-4 4.4×10^{13} atoms/cm^2

100 micron beam;SP-24, 13-4

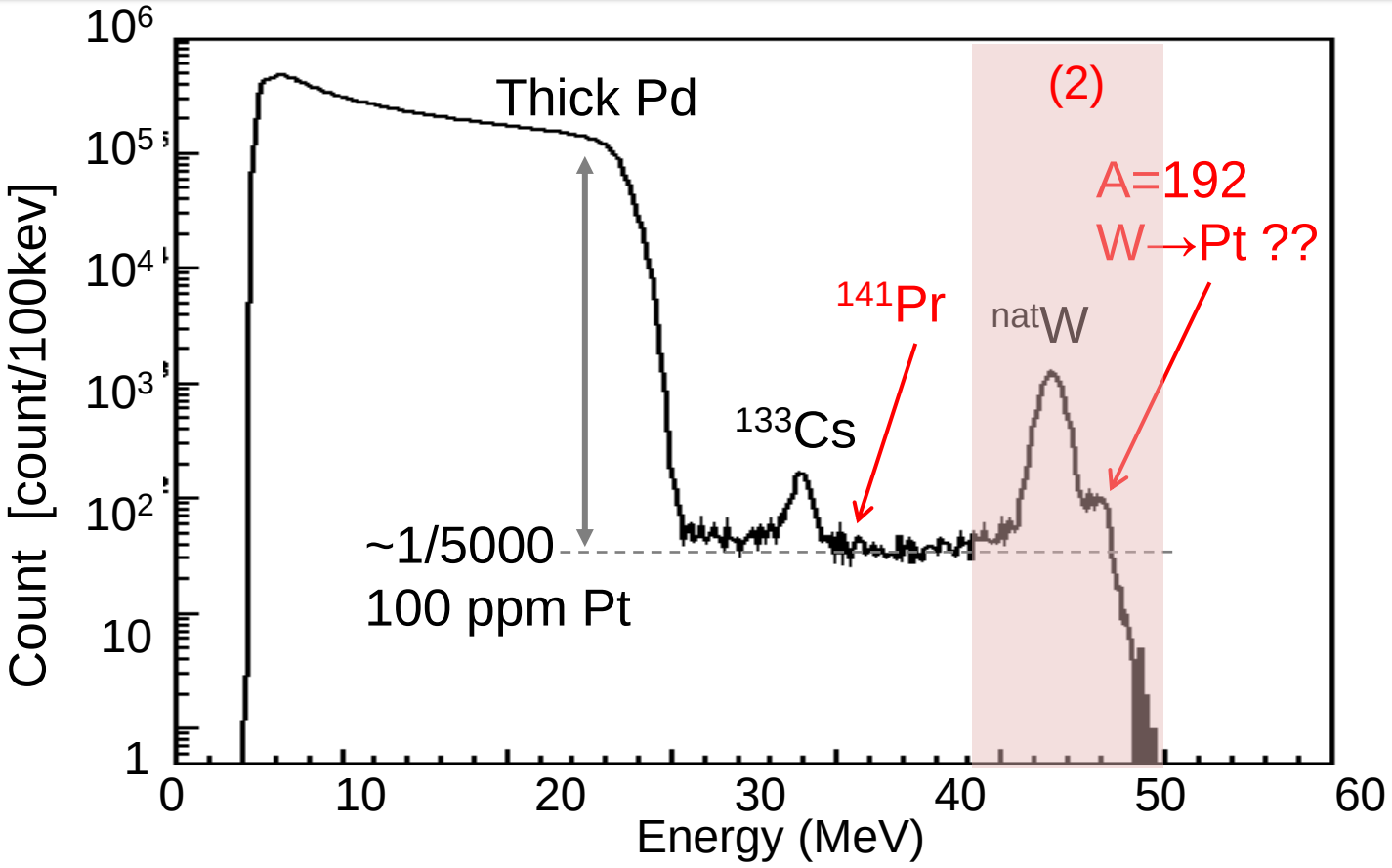




Y. Iwamura et al., *Condensed Matter Nuclear Science,* World Scientific, New Jersey, p.178, 2006.

RBS spectrum : with D_2 gas permeation

Summed spectrum of foreground samples



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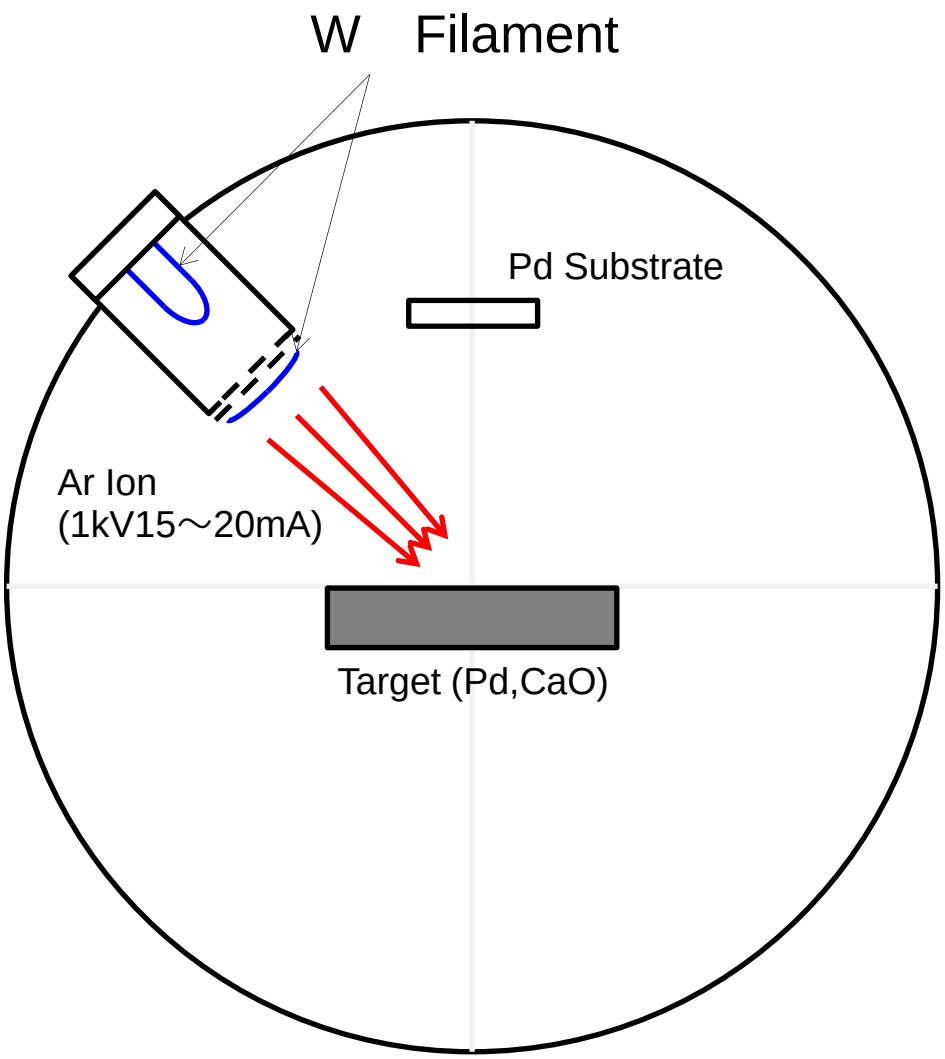
(1) A tiny peak at A=141 (^{141}Pr)

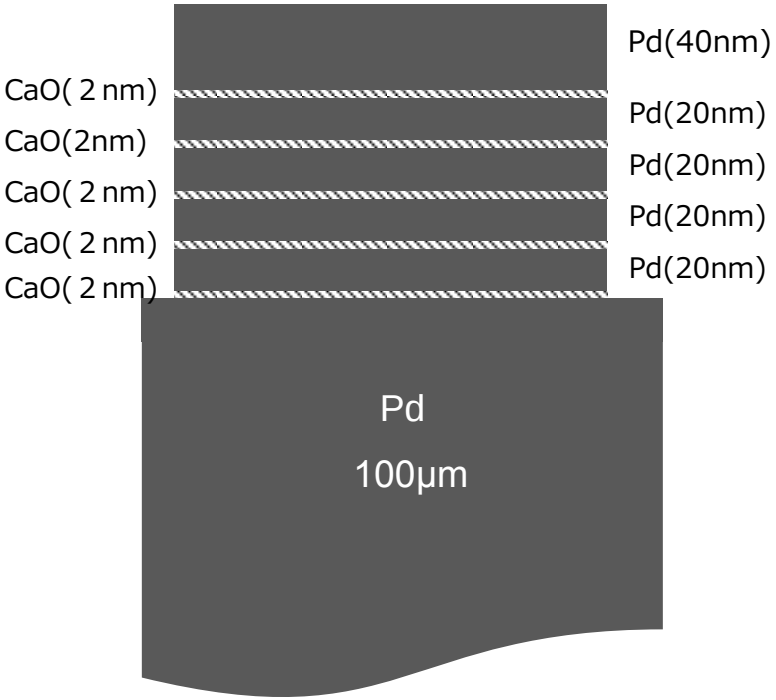
^{141}Pr exists on the surface. not observed for without D_2 ➡ consistent with MHI report ➡

W & Pt in Pd-CaO Multilayer

Ion beam Sputtering Apparatus

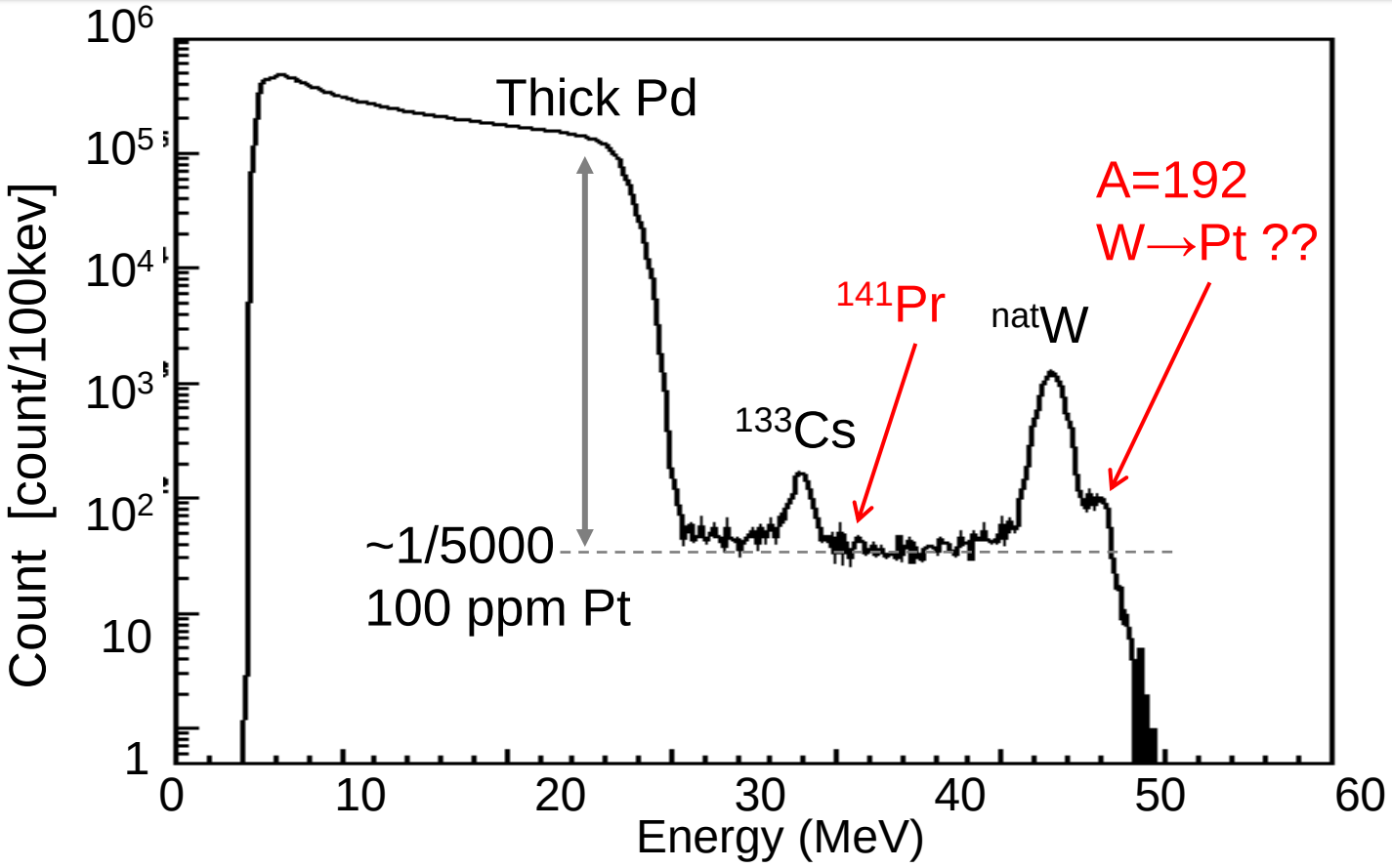


- Pd（multilayer＆Substrate) contain Pt about 100ppm
- Pd-CaO multiLayer contain W from ion source



RBS spectrum : with D_2 gas permeation

Summed spectrum of foreground samples



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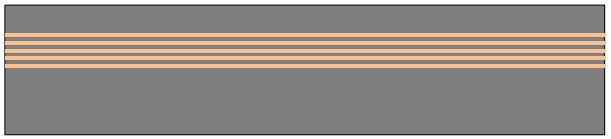
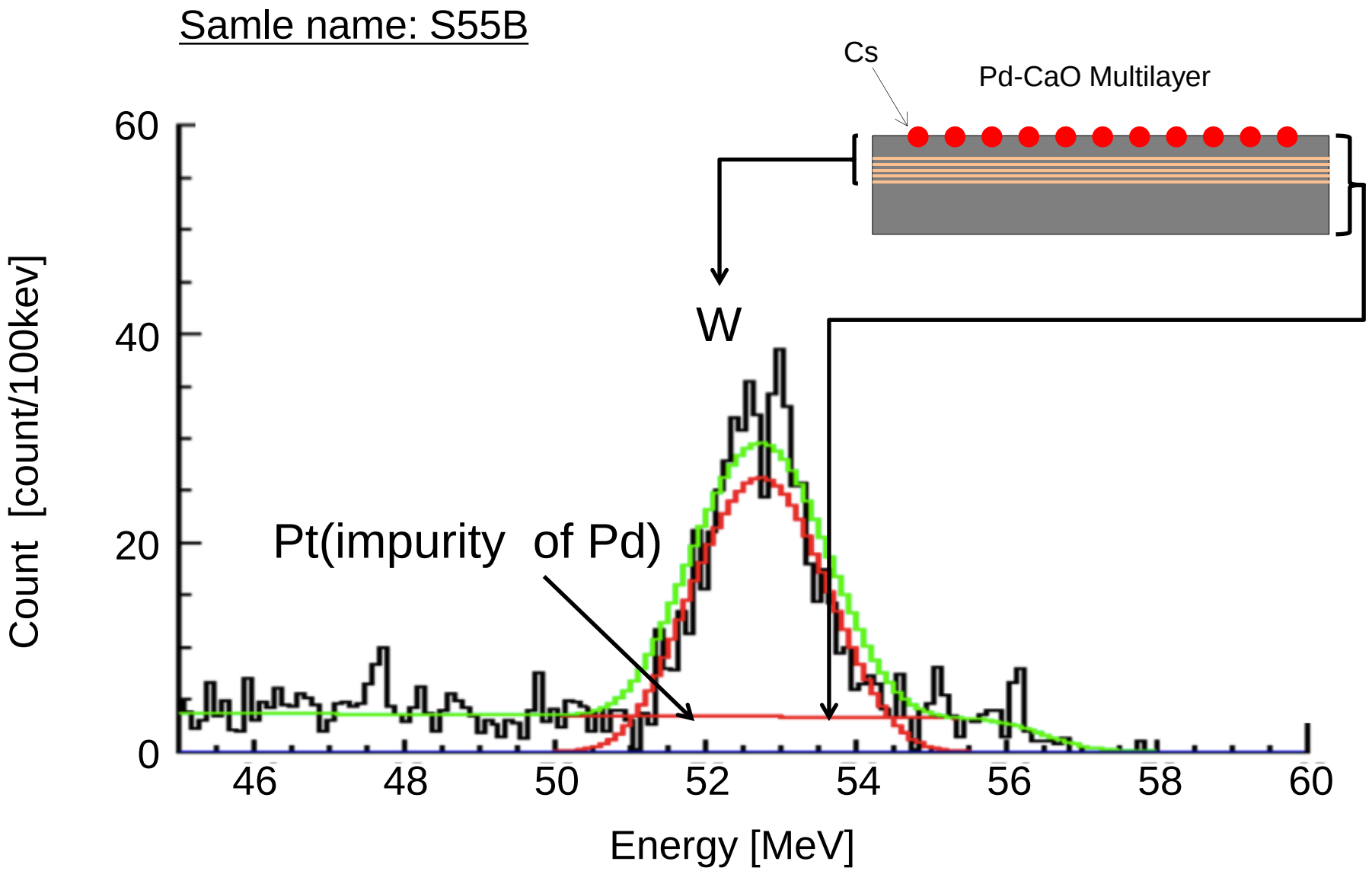
RBS spectrum
Pd multi-Layer without Cs dope & D_2 permeation

Samle name: M115

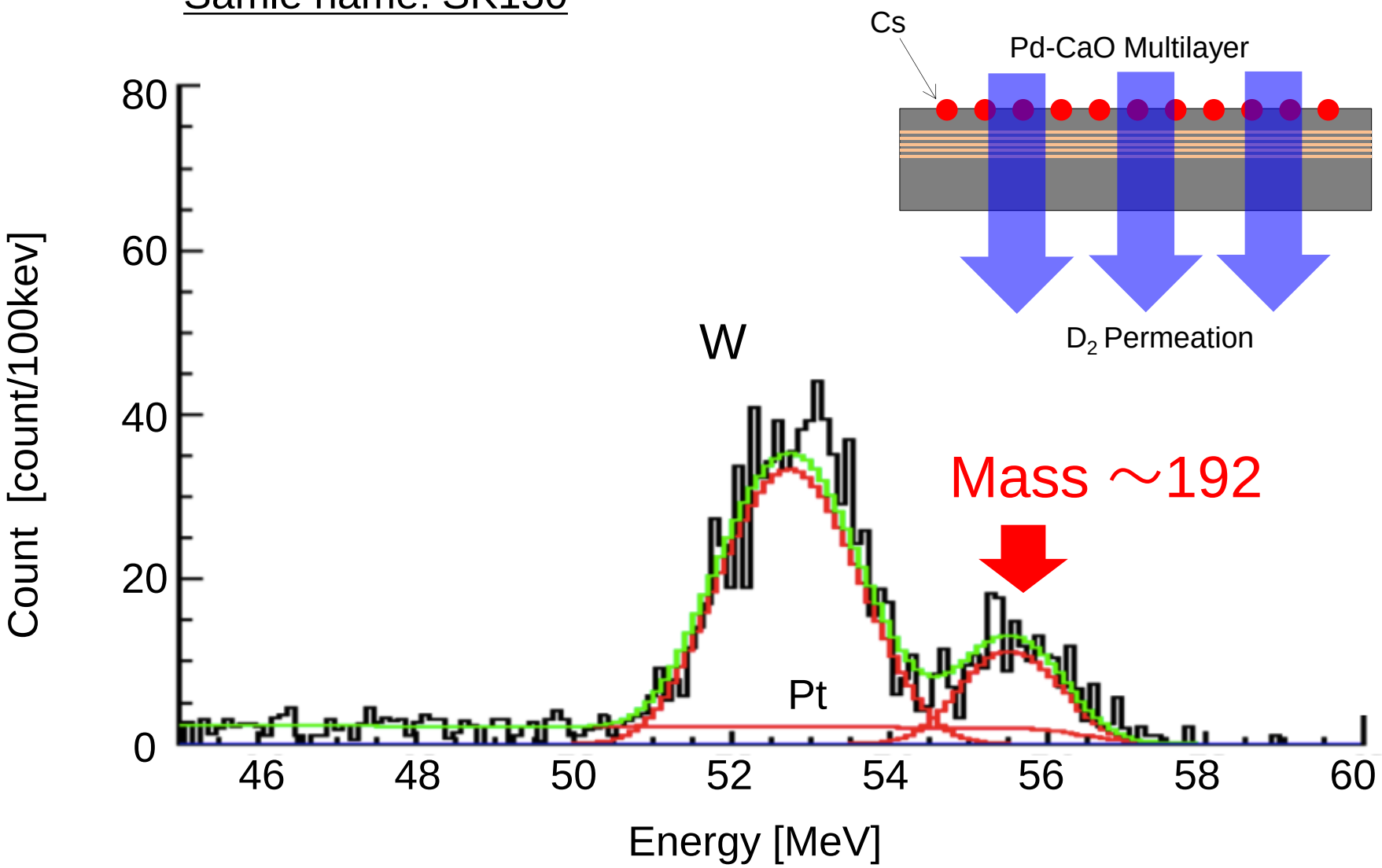


RBS spectrum
Cs Doped multilayer without D_2 permeation



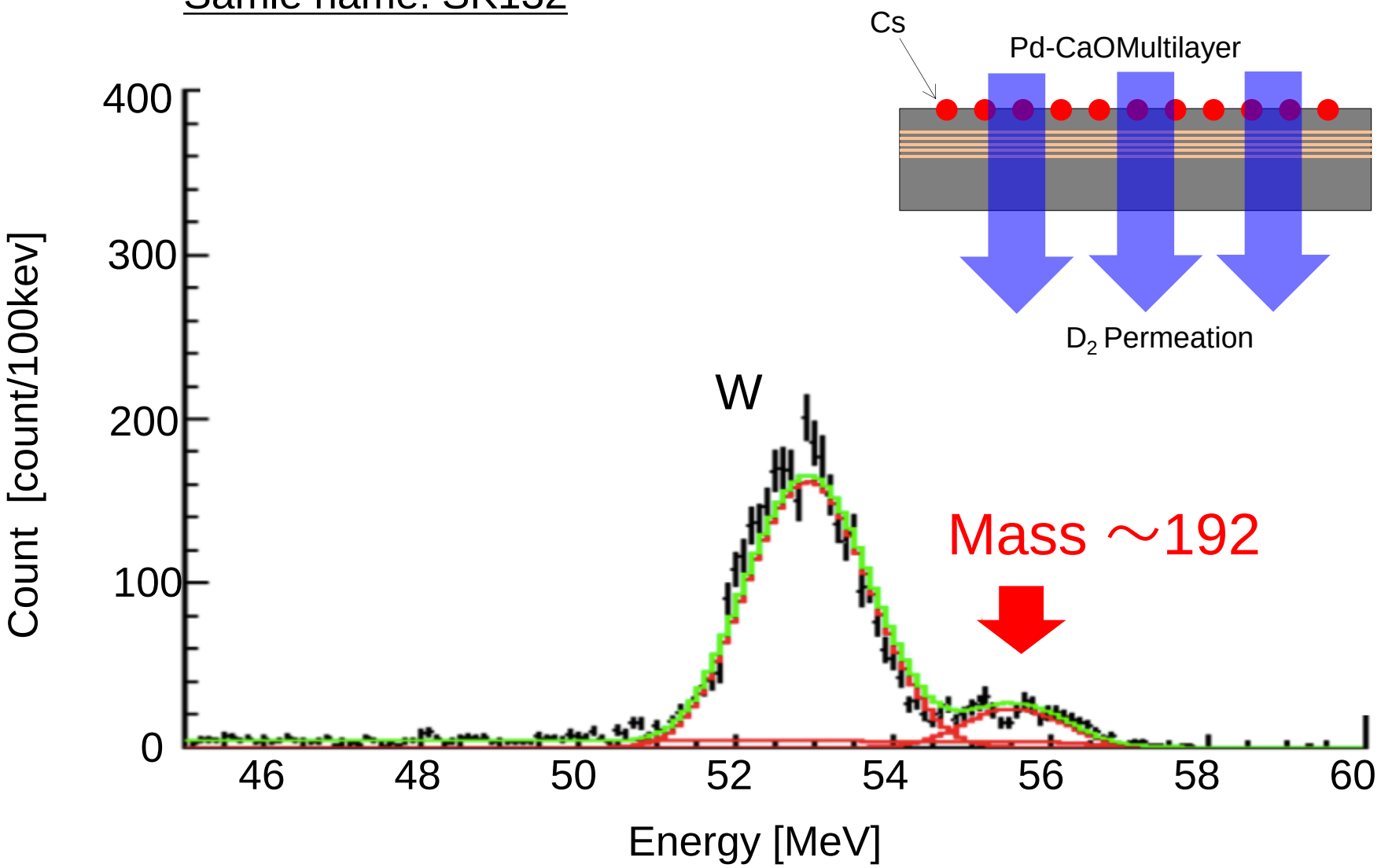
RBS spectrum
Cs Doped multilayer after D2 permeation

Samle name: SK130



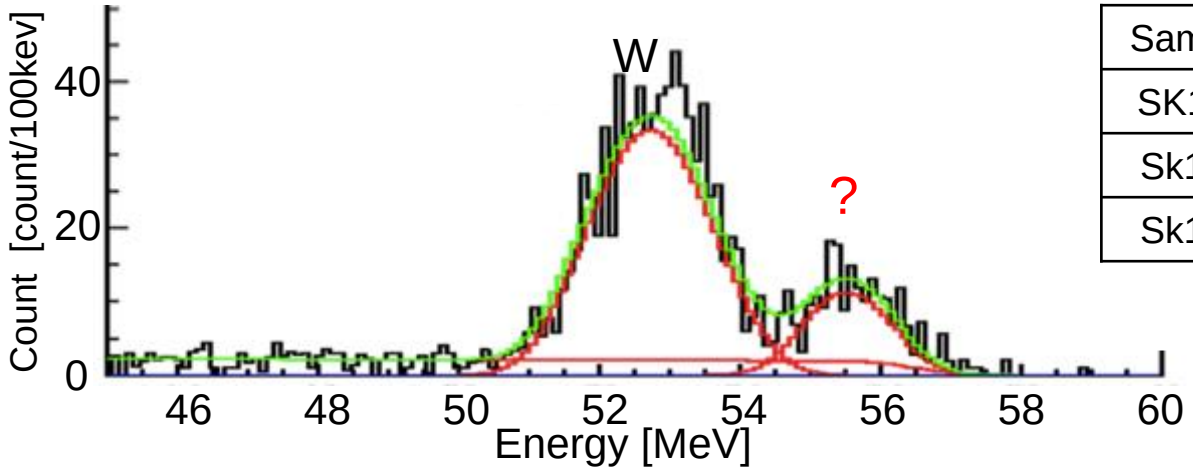
RBS spectrum
Cs Doped multilayer after D_2 permeation

Samle name: SK132

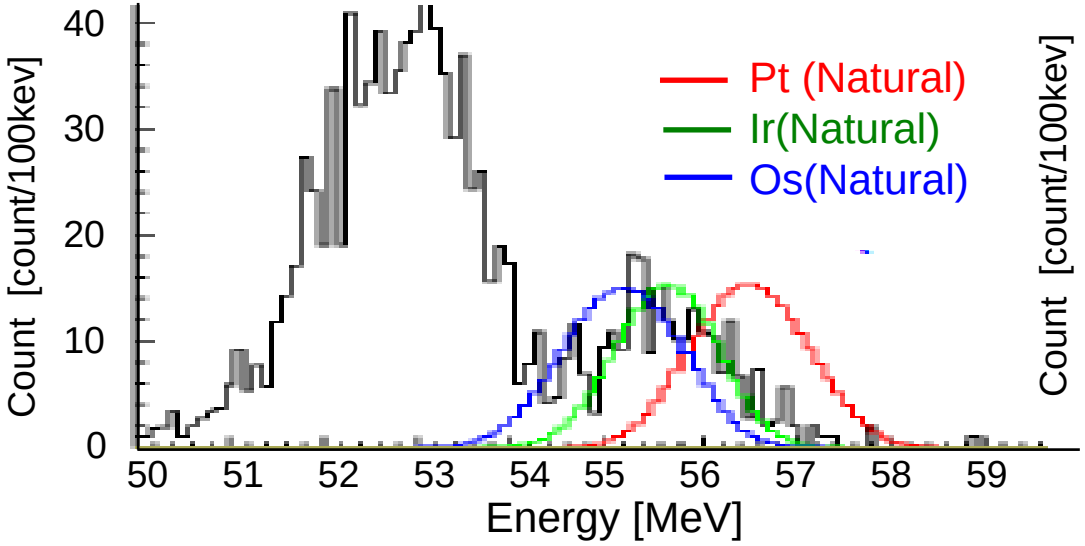


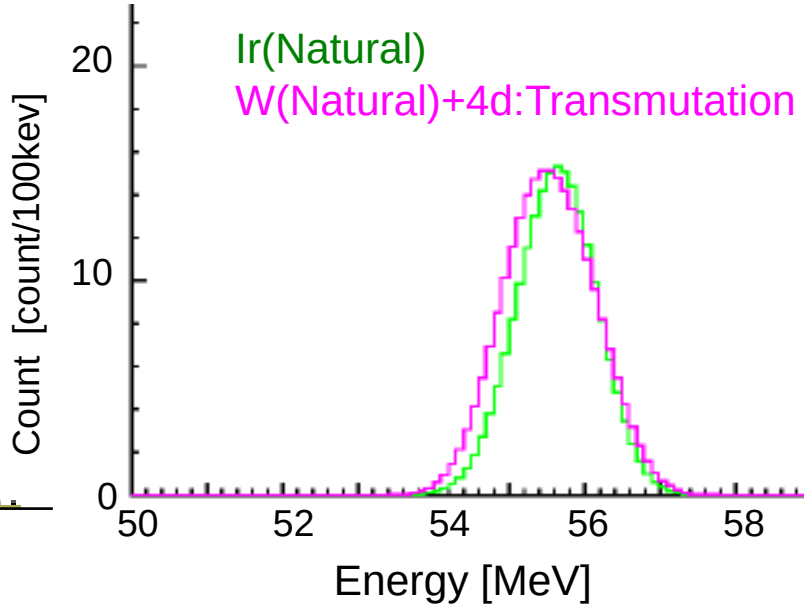
What is a peak A=192 Os? Ir?, Pt?

Samle name: SK130



Sample	W (ng/cm^2)	A~192 (ng/cm^2)
SK130	346±14	91.7±7.4
Sk132	697±13	90.8±5.0
Sk137	911±20	104±7





Ir natural :^{191}Ir (37%) ^{193}Ir(63%)

Conclusion

1.We performed Rutherford Backscattering Spectroscopy (RBS) to identify ^{141}Pr for direct nuclear mass assignment.

2. For FG samples, we could identify the ^{141}Pr events as well as ^{133}Cs, although the statistics is not enough. A comparison of the results between ICP-Ms and RBS shows that a quantitative agreement is not good.

$$Y(RBS)/Y(ICP) = 0.15 \sim 1.63 \text{ for } ^{133}Cs$$
$$< 0.15 \text{ for } ^{141}Pr$$

→ Measurement points of ICP-MS and RBS are different ?

→ may be due to the local distribution of ^{141}Pr

3. A broad peak indicating the existence of a nuclide with mass number around 192 was observed. There is a possibility that the impurity W in Pd/CaO multi-layer complex was transmuted into the elements of mass around 192.

Acknowledge

RBS measurement was partially supported by IMPACT Program of Council for Science, Technology and Innovation in 2015. Program name is “Reduction and Resource Recycle of High Level Radioactive Wastes with Nuclear Transmutation”.

The authors would like to thank
Mr. Ryo Tajima, Dr.Yuki Honda, and Prof. Hidetoshi Kikunaga for their corporations on RBS analysis, Research Center for Electron Photon Science, Tohoku University and
Mr. Hideki Yoshino and Mr. Masanao Hattori for their supports.

W+4d

$^{183}W(19\%) + 4d \rightarrow {}^{191}Pt \quad (\tau_{1/2} = 2.8\ d)$