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Low Radioactivity Techniques

UNIWERSYTET JAGIELLOŃSKI W KRAKOWIE

Removal of ^{222}Rn daughters from metal surfaces

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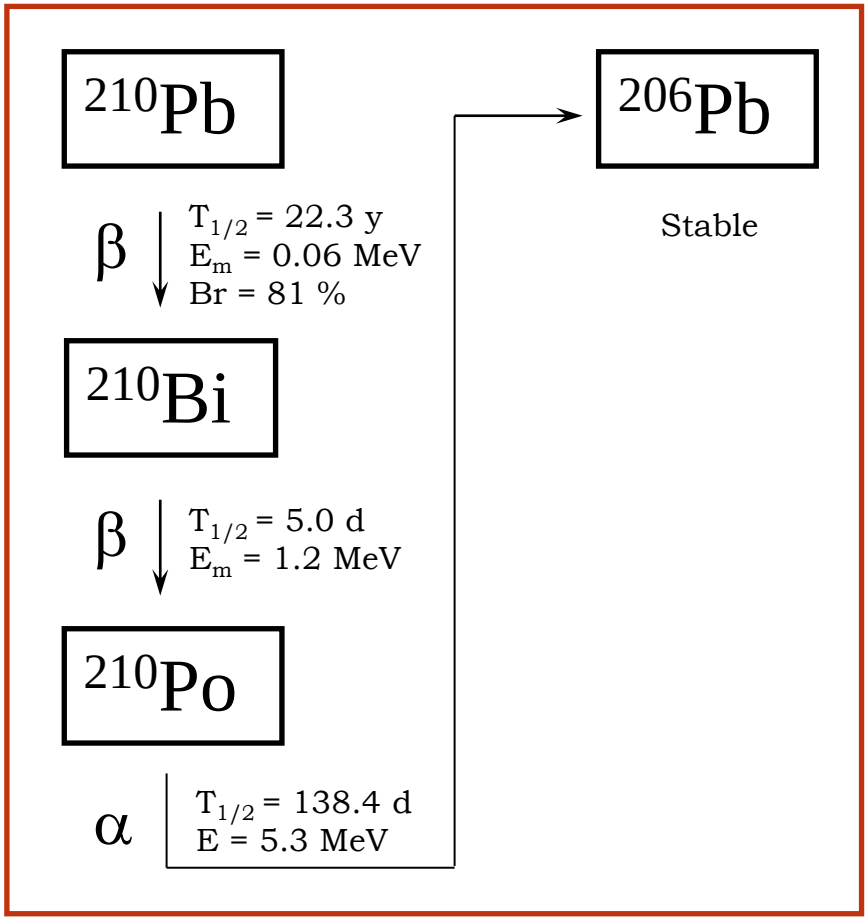
Outline

- Motivation
- Etching/electropolishing of artificially contaminated samples
- Tests of naturally contaminated samples
- Bulk ^{210}Po assay
- Conclusions

Motivation



- Disequilibrium in the chain
  $A_{Rn} \neq A_{pb}$
- In fresh material usually
  $A_{Pb} \neq A_{Po}$
- Each isotope should be investigated separately
- Pb may be plated out or implemented into the surface thus Rn-free atmosphere is needed
- ^{210}Bi:
  - pure β-emitter
- ^{210}Po:
  - degraded αs
  - source of ns (α,n) important for DM searches
  - difficult to remove from surfaces

Artificially contaminated samples

- Samples in form of discs with 50 mm diameter
- To increase the sensitivity samples were artificially loaded with ^{210}Pb, ^{210}Bi and ^{210}Po:
  - placed in a strong ^{222}Rn source for several months (^{210}Po specific activities of ≈100 Bq/m^2)
- Screening of ^{210}Po with an *alpha* spectrometer:
  - 50 mm Si-detector, bkg ≈2 α/d (1 – 10 MeV)
  - sensitivity ≈20 mBq/m^2 (100 mBq/kg, ^{210}Po)
- Screening of ^{210}Bi with a *beta* spectrometer:
  - 2 × 50 mm Si(Li)-detectors, bkg ≈0.18/0.40 cpm
  - sensitivity ≈10 Bq/kg (^{210}Bi)
- Screening of ^{210}Pb (46.6 keV line) with a *gamma* spectrometer:
  - 16 % HPGe detector with an active and a passive shield

Artificially contaminated samples

Activity reduction factors after etching / electropolishing

			Germanium	
Isotope	Copper	Stainless steel	NPGe	HPGe
^{210}Pb	50 / 300	100 / 400	100 / –	700 / –
^{210}Bi	50 / 300	100 / 800	400 / –	800 / –
^{210}Po	1 / 400	20 / 700	1000 / –	100 / –

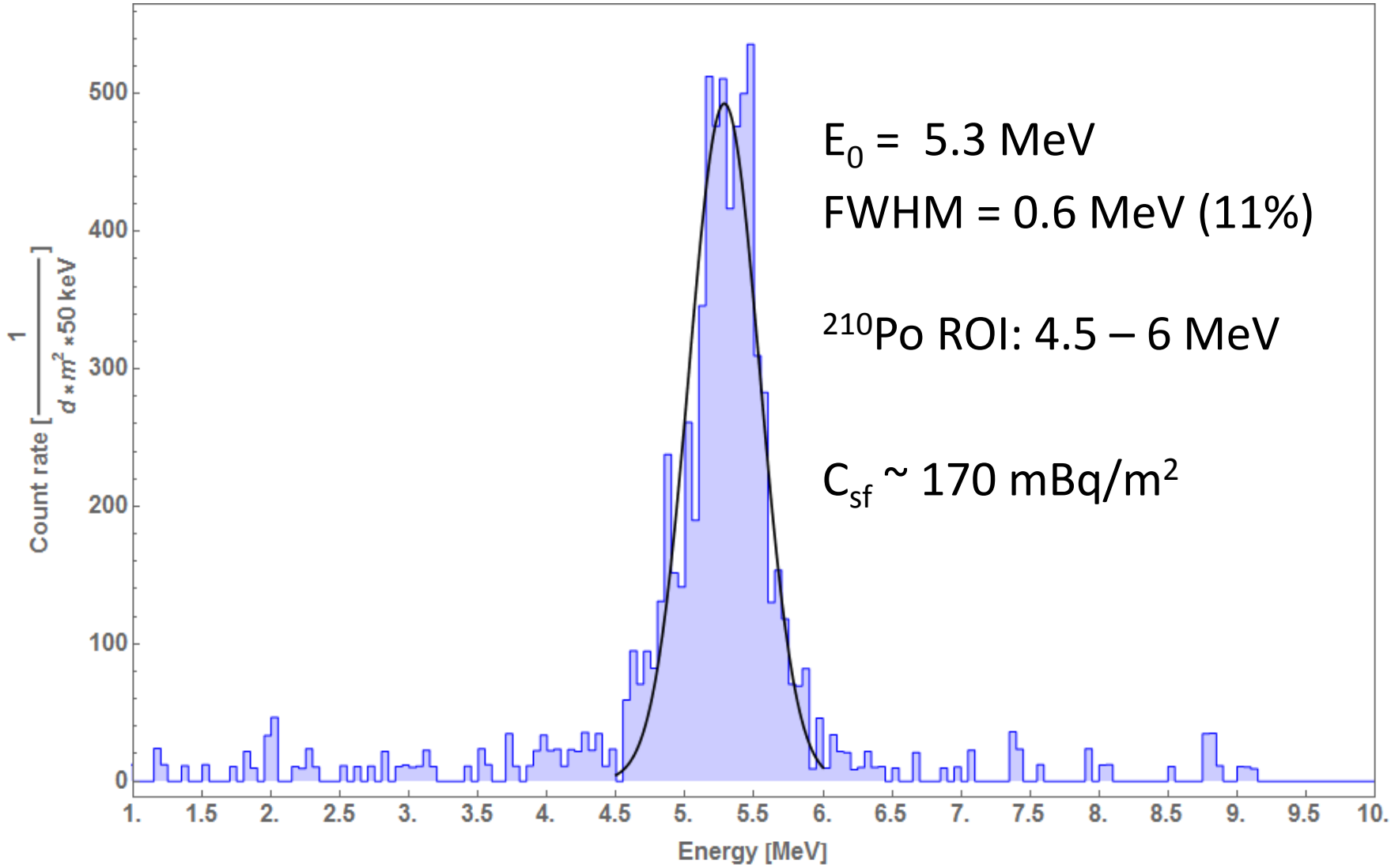
- Copper
  - etching: 5 min in (1% H_2SO_4 + 3% H_2O_2) and 5 min in 1% citric acid
  - electro-polishing (electrolyte): 85 % H_3PO_4 + 5 % 1-butanol ($C_4H_{10}O$)
- Stainless steel
  - etching: (20 % HNO_3 + 1.7 % HF) and 15 % HNO_3
  - electro-polishing (electrolyte): 40 % H_3PO_4 + 40 % H_2SO_4 + 3 % CrO_3
- Germanium
  - etching: CP4 solution (45.45 ml HNO_3 + 27.27 ml HF + 27.27 ml CH_3COOH + 0.5 ml Br for 100 ml solvent) done by Canberra-France in Lingolsheim in cooperation with MPP Munich

NIM A 676 (2012) 140
NIM A 676 (2012) 149

XIA Large Surface Detector

- Only ^{210}Po studied
- Low background, large surface (LBS) alpha spectrometer
- Ar as counting gas (3.5 L/min)
- Sample size: 43 × 43 cm or 30 cm diameter disc, 1 mm thick
- PSD + veto guard (alphas, *mid-airs*, *ceiling*, *rounds*)

Copper sheet with surface ^{210}Po (air-born)



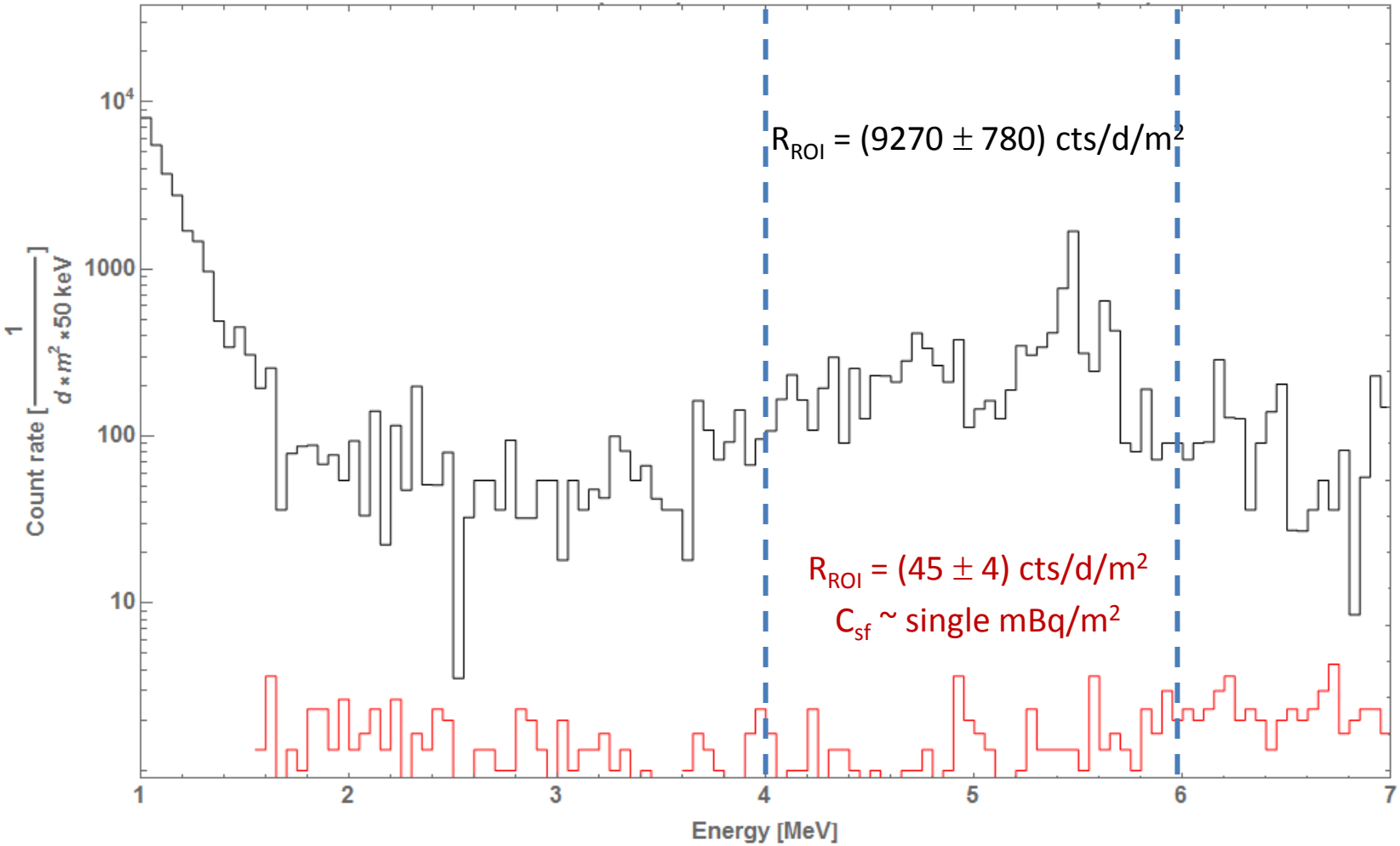
E_0 = 5.3 MeV

FWHM = 0.6 MeV (11%)

^{210}Po ROI: 4.5 – 6 MeV

C_{sf} ~ 170 mBq/m^2

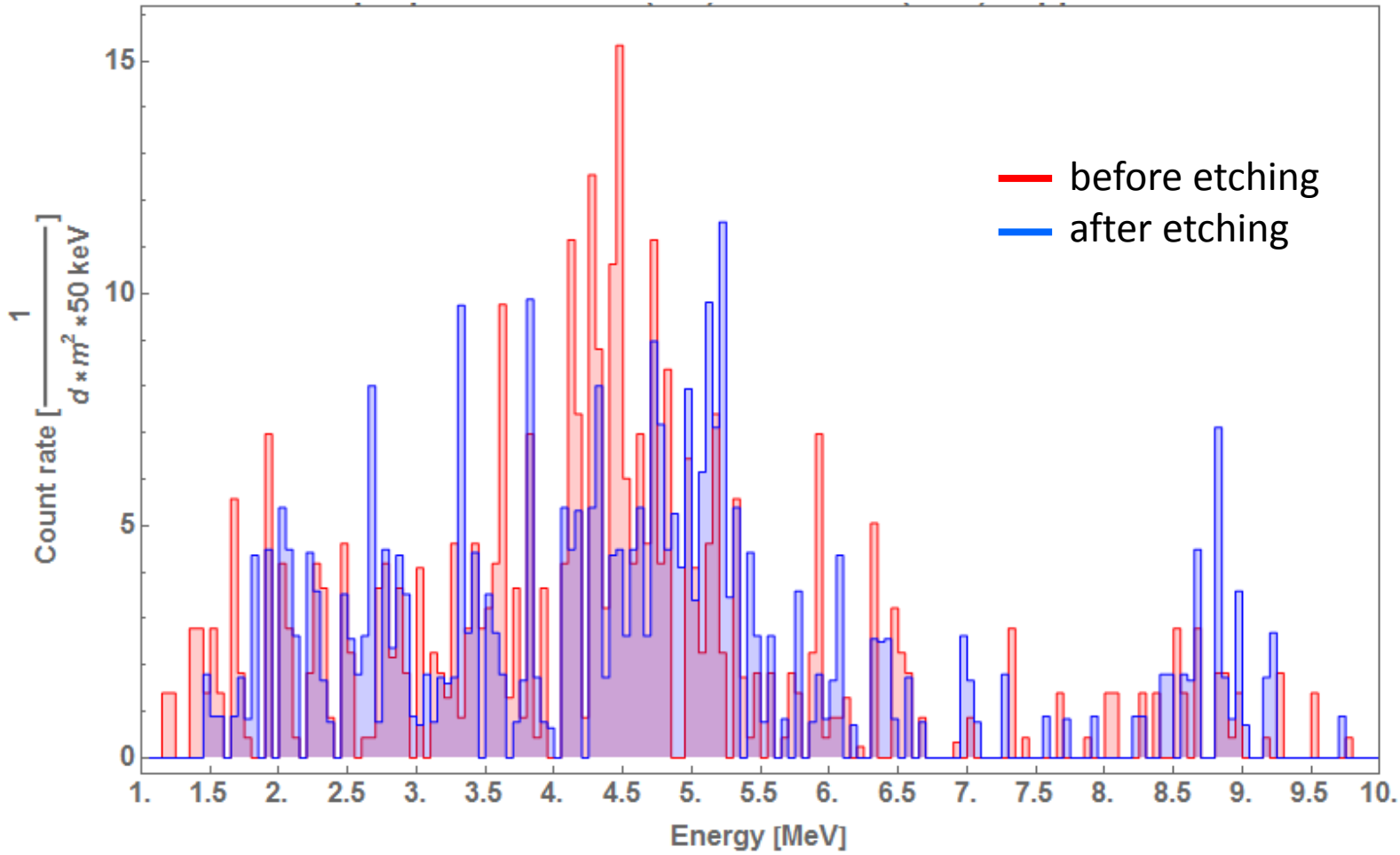
XIA Large Surface Detector – background



- Low background ORTEC α detector (40 mm diameter) at LNGS (*Appl. Rad. Isot. 81 (2013) 146*) vs. XIA Ultra-Low 1800 on the surface: **factor 200 improvement**

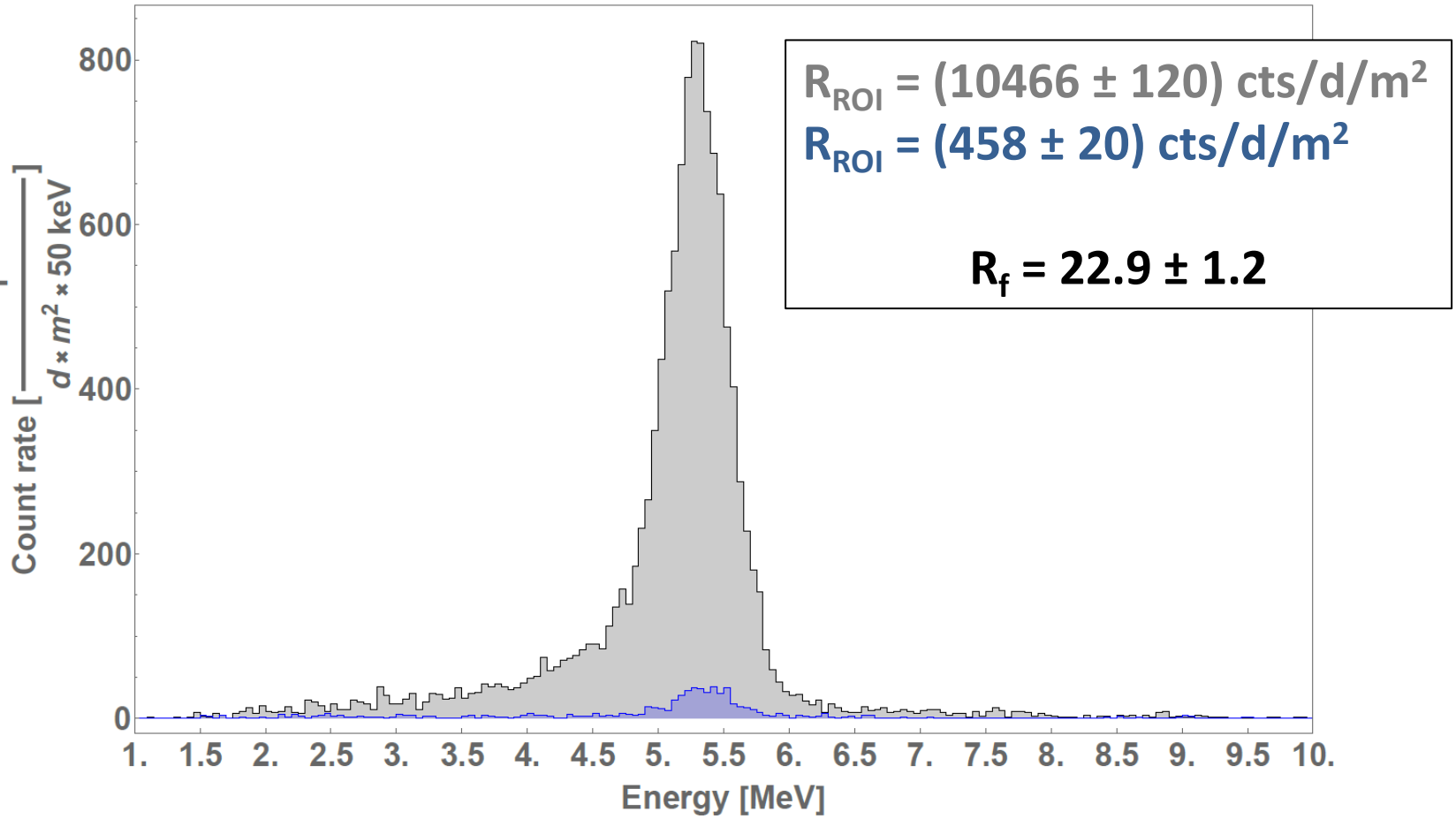
Appl. Rad. Isot. 81 (2013) 146

Etching of clean Cu (ETP)



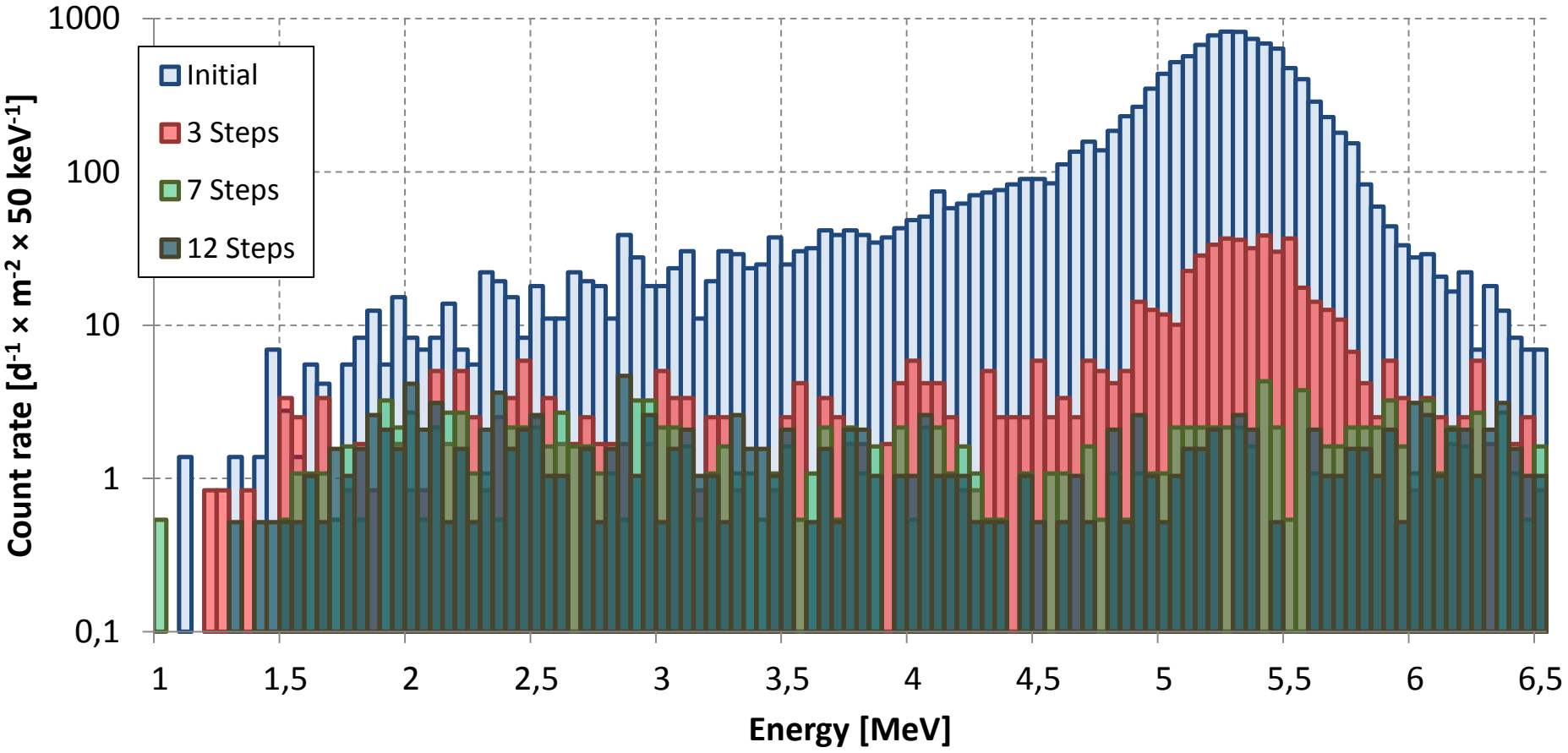
- Etching procedure: 5 min in a mixture of: 1% H_2SO_4 + 3% H_2O_2
- Washing in high-purity deionized water (18 MΩ × cm)
- No effect observed for surface ^{210}Po (at the level of ~1 mBq/m^2)

Dynamic etching

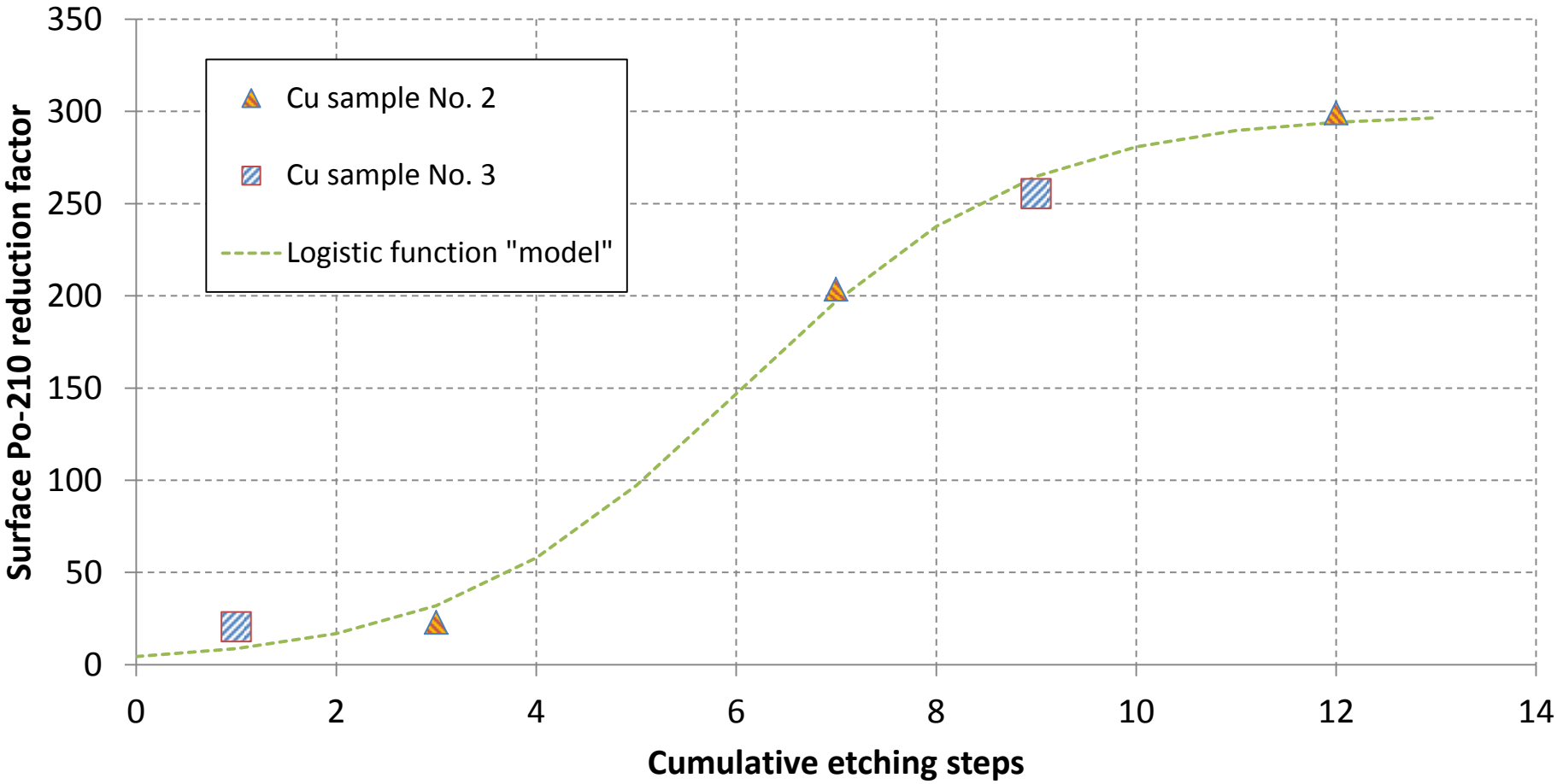


- Etching procedure: 4 × 1 min in a mixture of 1% H_2SO_4 + 3% H_2O_2
- Passivation with 1% citric acid at the end
- Washing in high-purity deionized water (18 MΩ × cm)

Dynamic etching – 3, 7, 12 cumulative steps

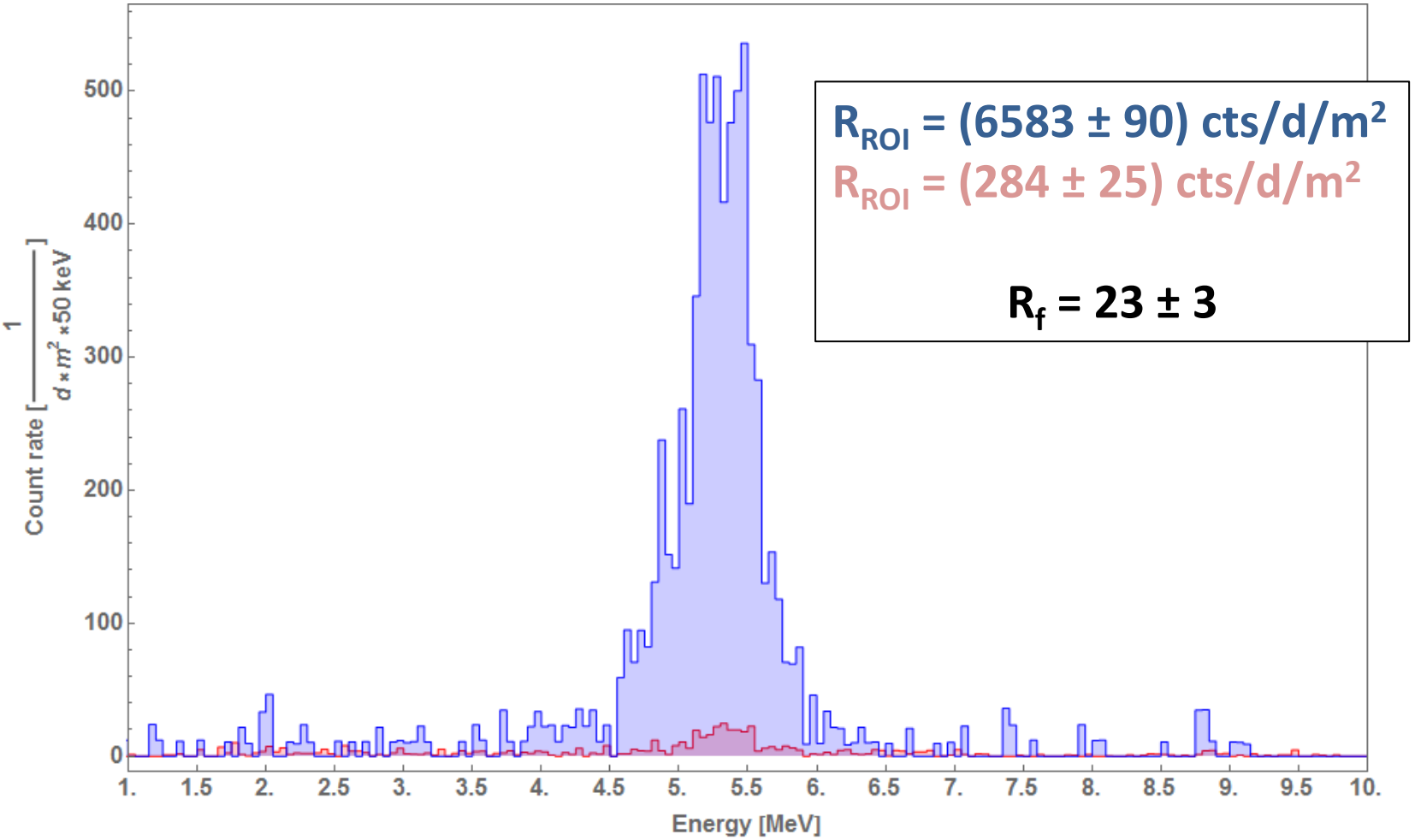


Dynamic etching



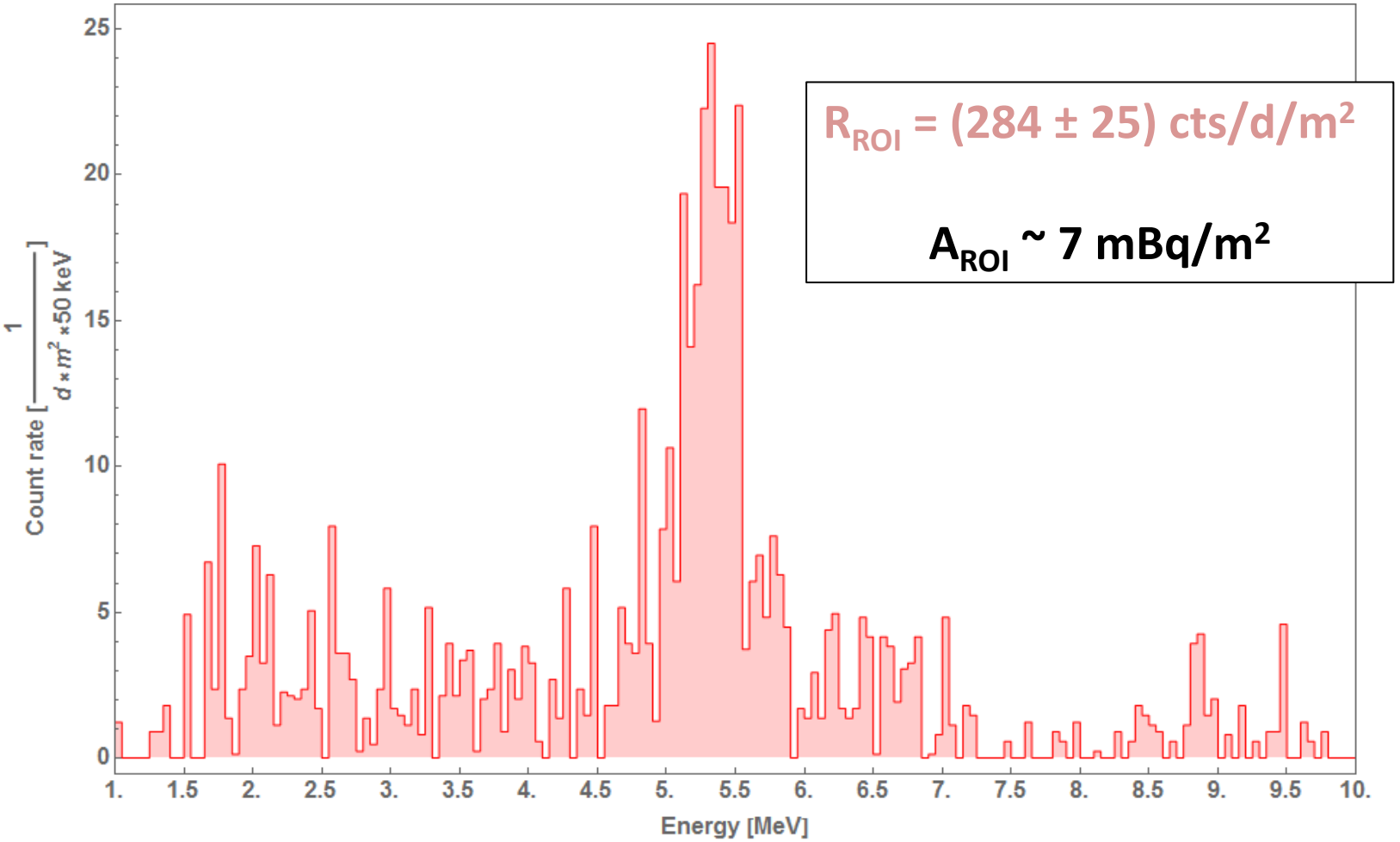
- Energetic washing n-times always with fresh reagents

Electropolishing of copper



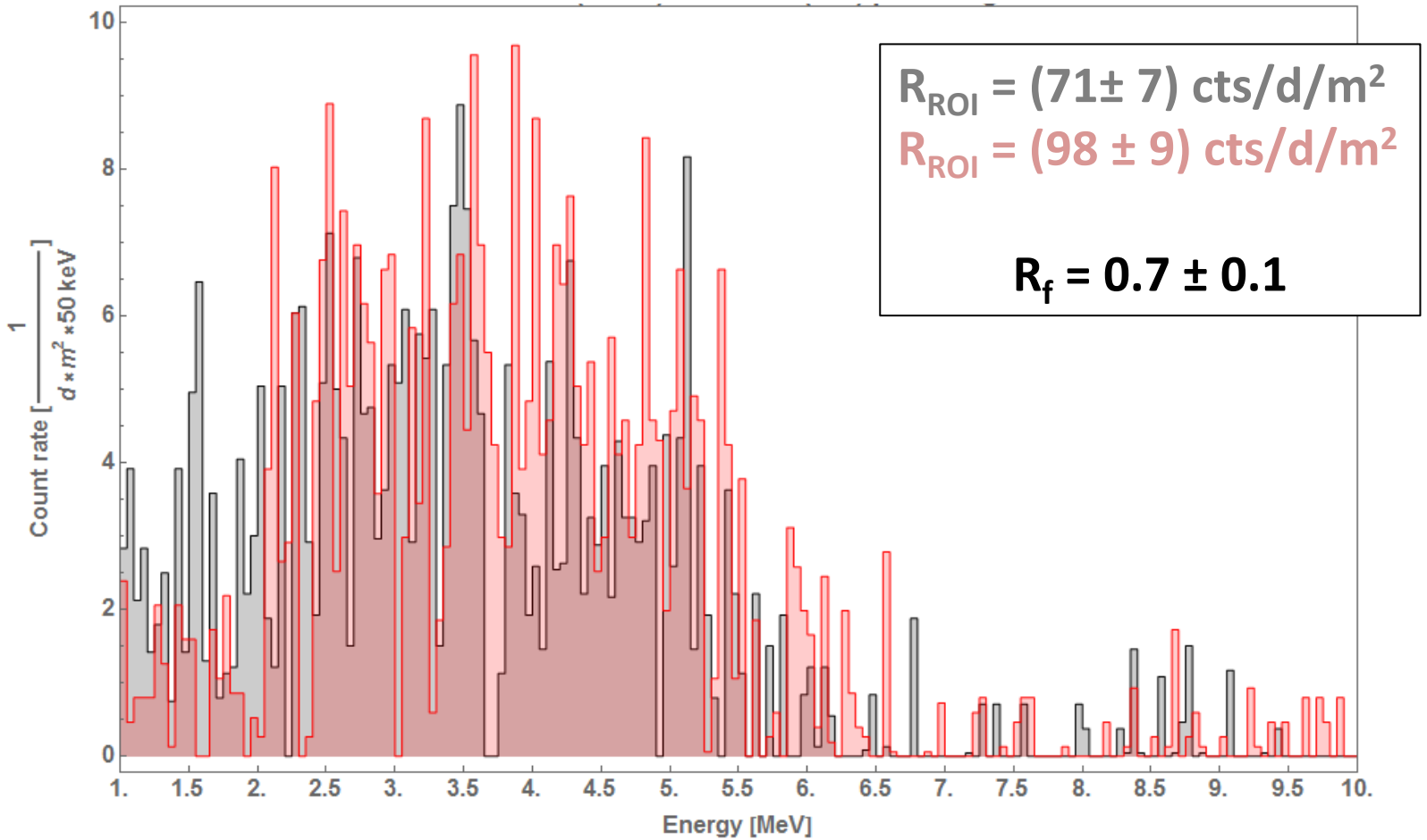
- Polishing solution: 95% H_3PO_4 + 1% 1-butanol
- Polishing conditions: 2.5 A/dm², 3 V, 20 min, 2 cm plate distance, room temperature

Electropolishing of copper



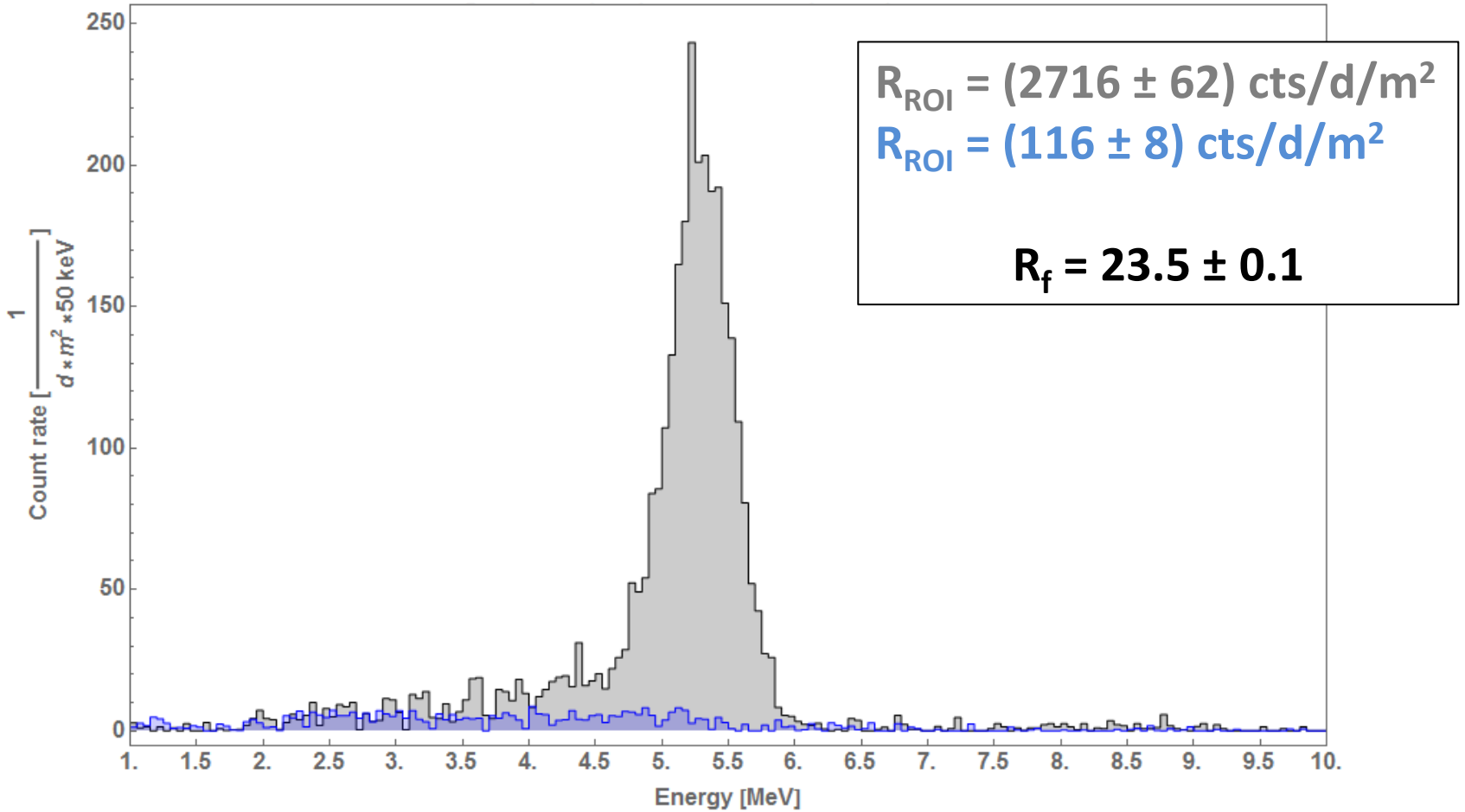
- Polishing solution: 95% H_3PO_4 + 1% 1-butanol
- Polishing conditions: 2.5 A/dm², 3 V, 20 min, 2 cm plate distance, room temperature

Electropolishing of clean stainless steel (SS 1.4301 (304), sheet No. 1)



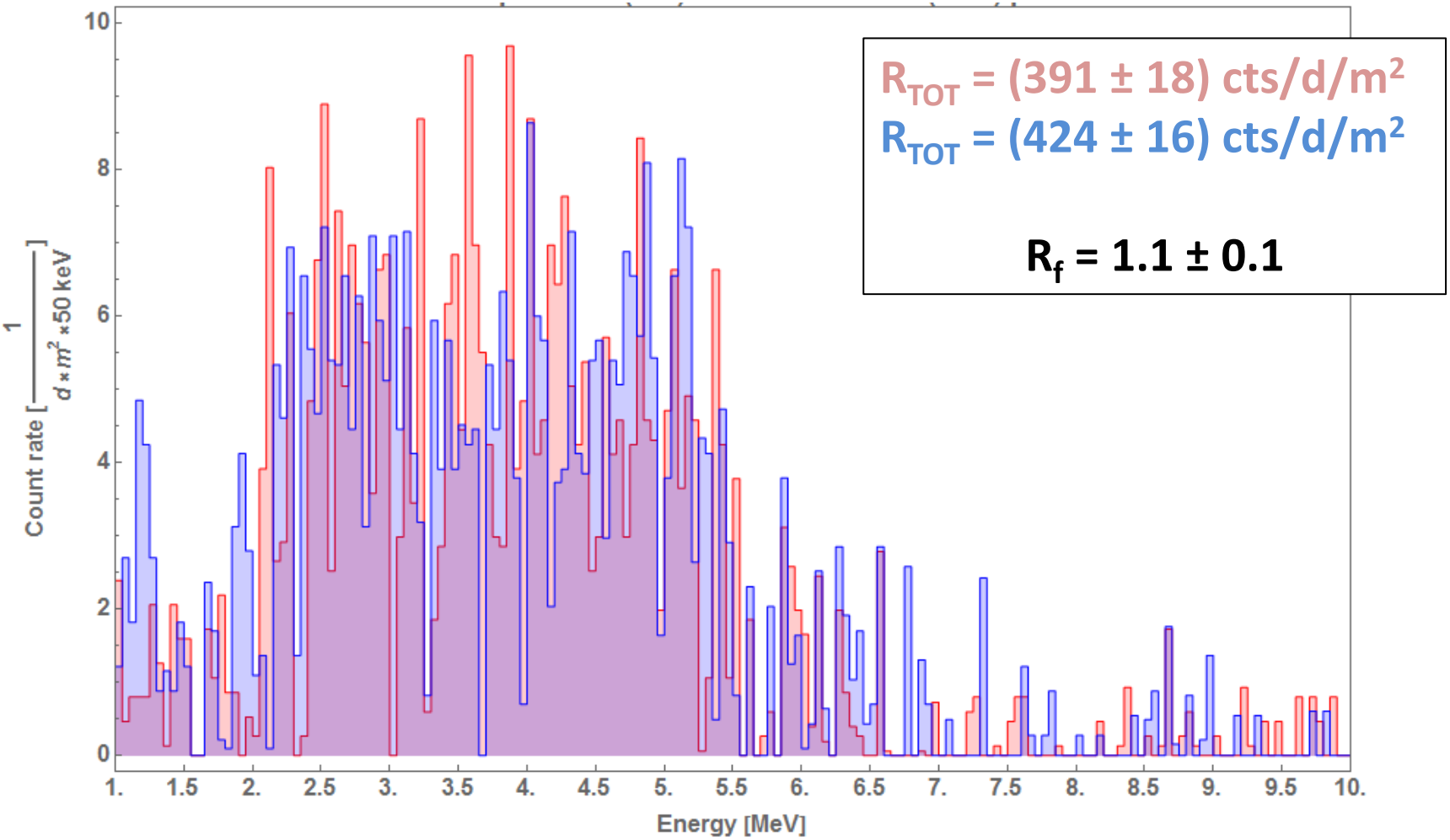
- Polishing mixture: 1:1 of 95% H_2SO_4 and 85% H_3PO_4
- Washing: 5 min in 15% HNO_3 and later in HP water
- Polishing conditions: 2.5 A/dm², 2 V, 25 min, 2 cm plate distance, T ≈ 50 °C

Electropolishing of stainless steel (SS 1.4301 (304), sheet No. 2)



- Polishing mixture: 1:1 of 95% H_2SO_4 and 85% H_3PO_4
- Washing: 5 min in 15% HNO_3 and later in HP water
- Polishing conditions: 2.5 A/dm², 2 V, 25 min, 2 cm plate distance, T ≈ 50 °C

Electropolishing of stainless steel (sheet No. 1 and 2 after electropolishing)



- No effect observed for surface ^{210}Po (at the level of ~1 mBq/m²)

Bulk ^{210}Po assay

First results for bulk ^{210}Po in various metals reported during the ICRM–LLRMT conference in Seattle (Sept. 2016)

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Studies of surface and bulk ^{210}Po in metals using an ultra-low background large surface alpha spectrometer

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ABSTRACT

First measurements of natural surface and bulk ^{210}Po specific activities for metals are reported. If covered with protective foils, the surfaces did not show indications of ^{210}Po and the obtained upper limits are in the range of single mBq m^{-2}. Weak bulk activities, in the range of 50 – 280 mBq kg^{-1}, were registered for Stainless Steel and Copper, while significant amounts of ^{210}Po, ~1.5 Bq kg^{-1}, were detected in Titanium. One special Teflon sample was investigated with respect to its bulk ^{210}Po.

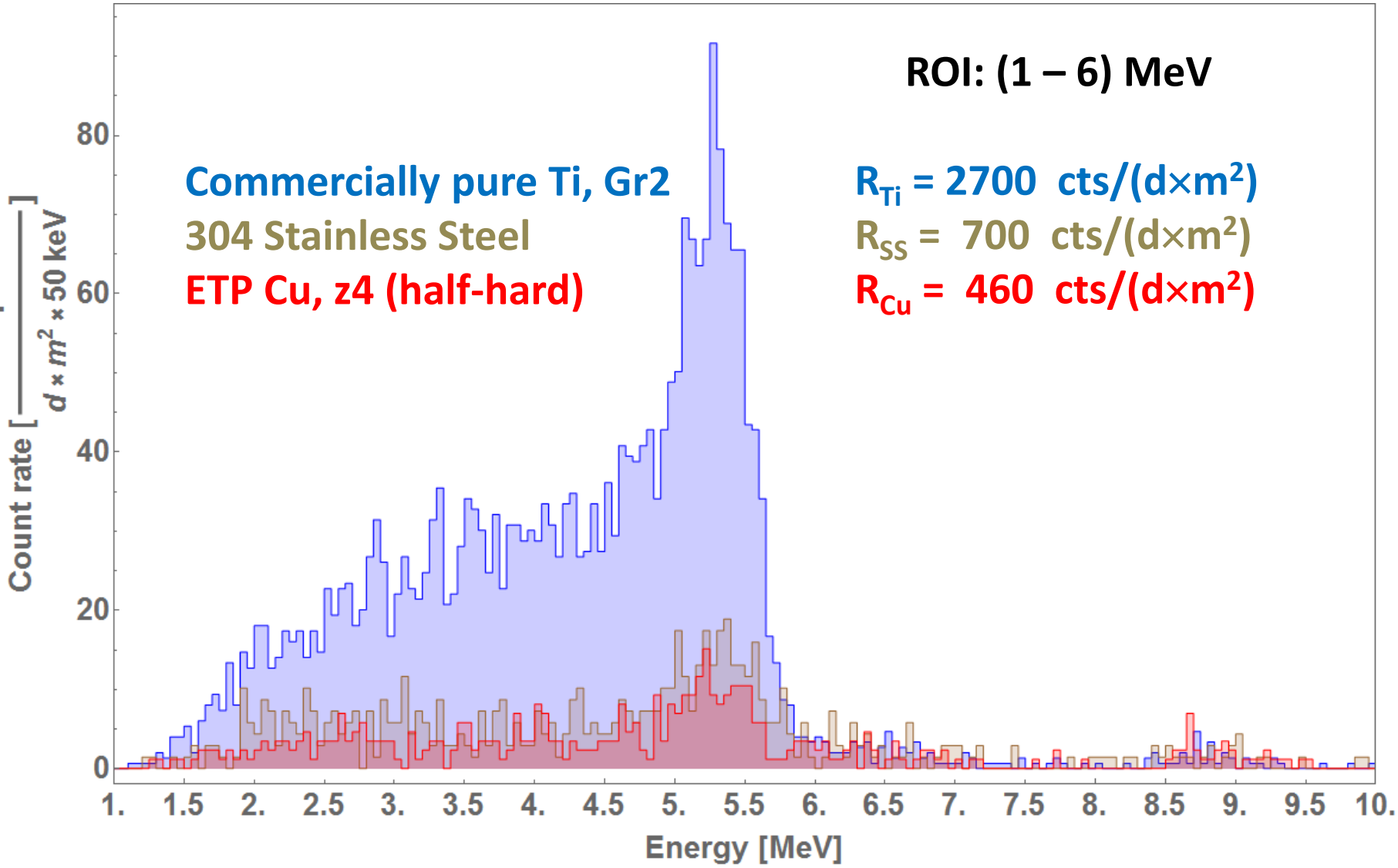
https://doi.org/10.1016/j.apradiso.2017.01.030

Bulk ^{210}Po assay

Material	Bulk ^{210}Po [mBq/kg]	Surface ^{210}Po [mBq/m^2]	Remarks
OF Copper	54	≤ 3	z4 (half hard)
ETP Copper	75	≤ 3	z4 (half hard)
„Old” ETP Copper	280	170	z4 (half hard)
Stainless Steel	80	≤ 3	Type 304
Titanium	1500	68	GR2
Teflon	$\leq$ 46	–	High purity, ATP

https://doi.org/10.1016/j.apradiso.2017.01.030

Bulk ^{210}Po assay



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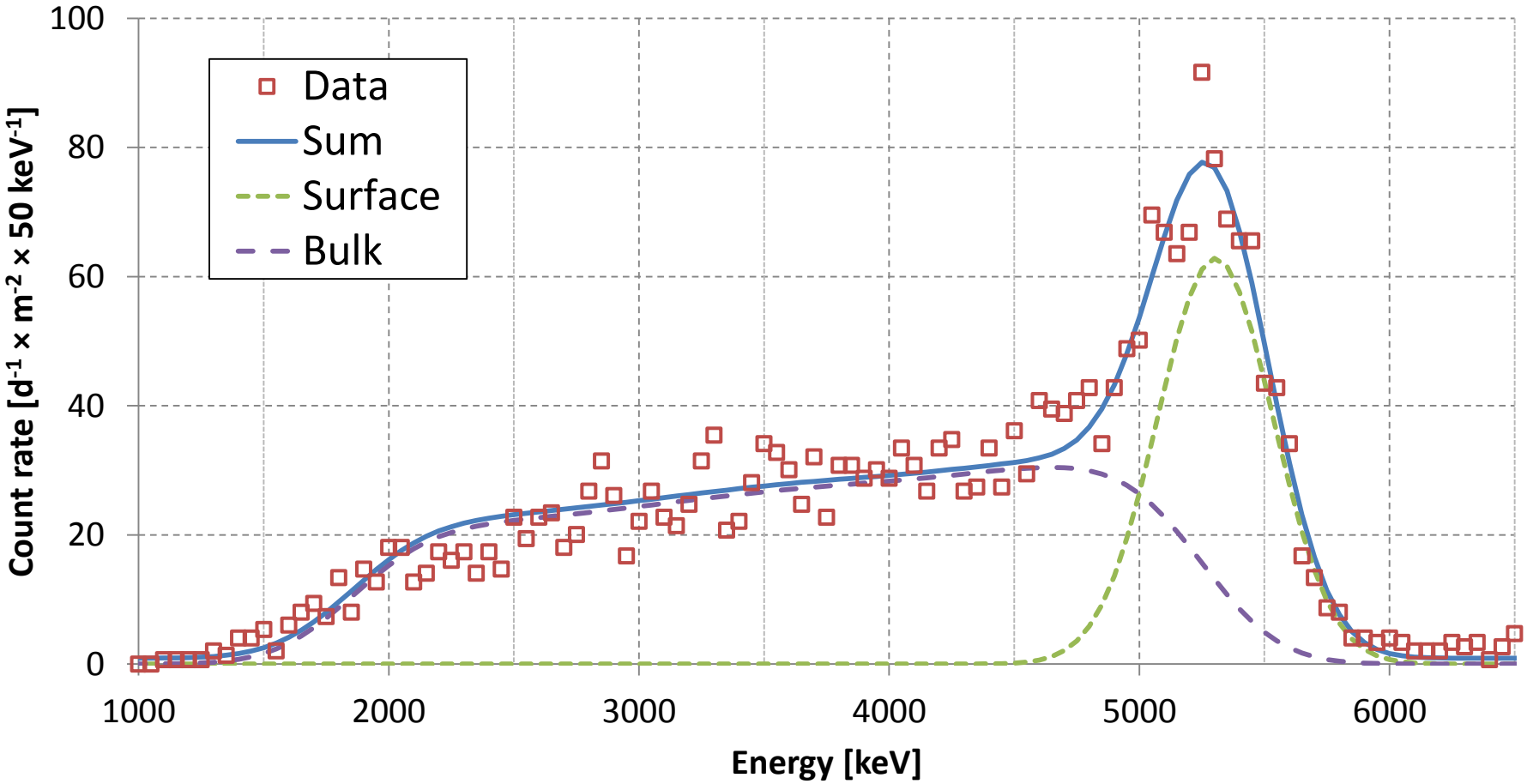
^{210}Po in/on Titanium after surface treatment (etching)



- The detection efficiencies precisely fixed (spectrum deconvolution)
- $C_{BULK} = (1.0 \pm 0.1)$ Bq/kg
- $C_{sf} = (18.7 \pm 1.0)$ mBq/m^2

Conclusions

- Copper/stainless steel surfaces protected against air (^{222}Rn) did not show indications of ^{210}Po.
- Etching/electropolishing removes ^{210}Pb, ^{210}Bi and ^{210}Po from metal surfaces, the effect seems to be material- and surface finish dependent. „*Static*" etching did not affect ^{210}Po on copper.
- At the level of mBq/m^2 proper etching/electropolishing does not contaminate surfaces with ^{210}Po.
- For the first time an effect of ^{210}Po activity reduction on copper surface after („*dynamic*") etching has been observed.
  High activity reduction factor obtained (~300).
- Applied electropolishing procedures reduce the natural ^{210}Po surface activity on copper/steel by a factor up to 25.
  There is still room for improvements.
- High-sensitivity alpha spectrometers allow for ^{210}Po surface and bulk activity measurements.