Radon emanation suppression by surface coating

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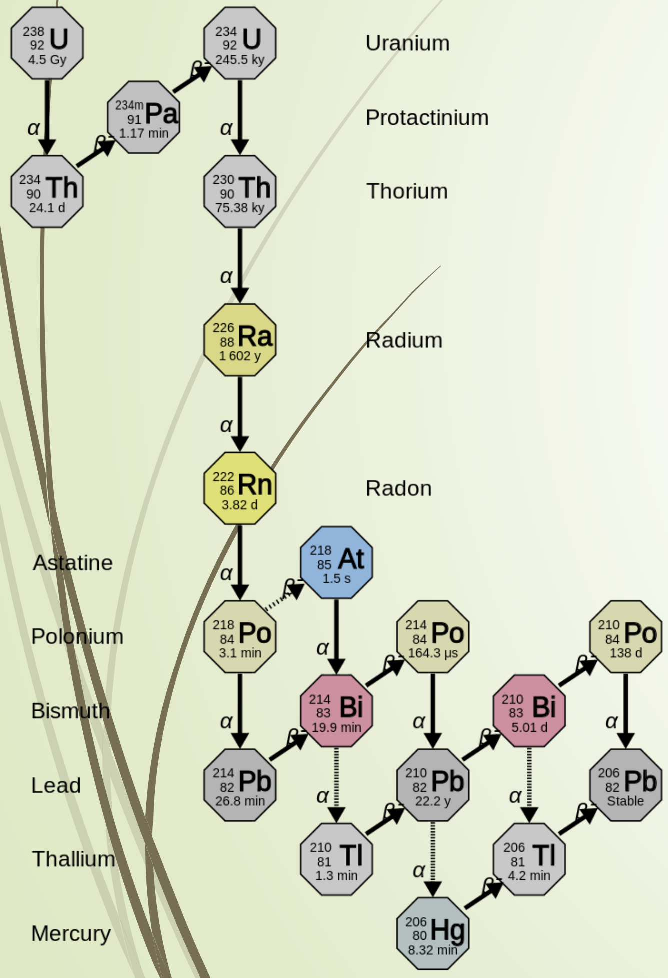
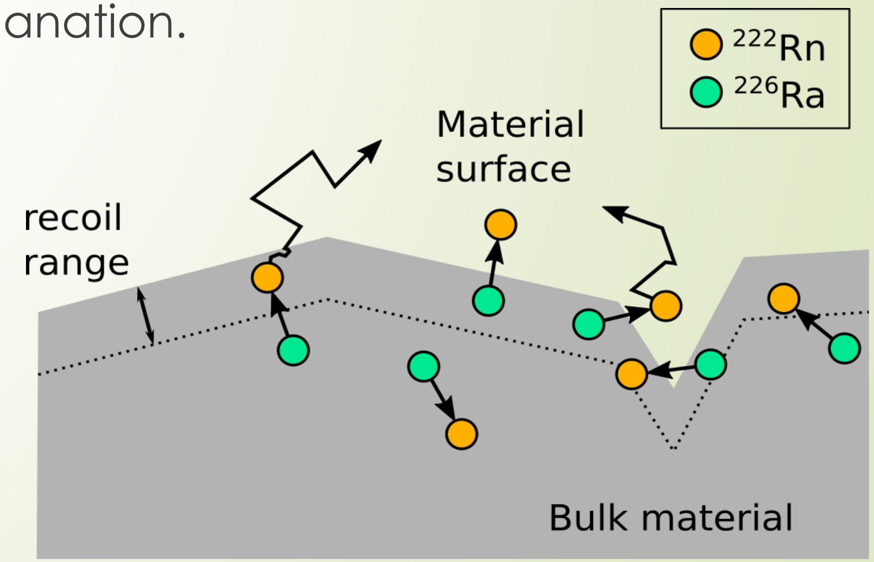
Radon background in low energy astroparticle physics experiments



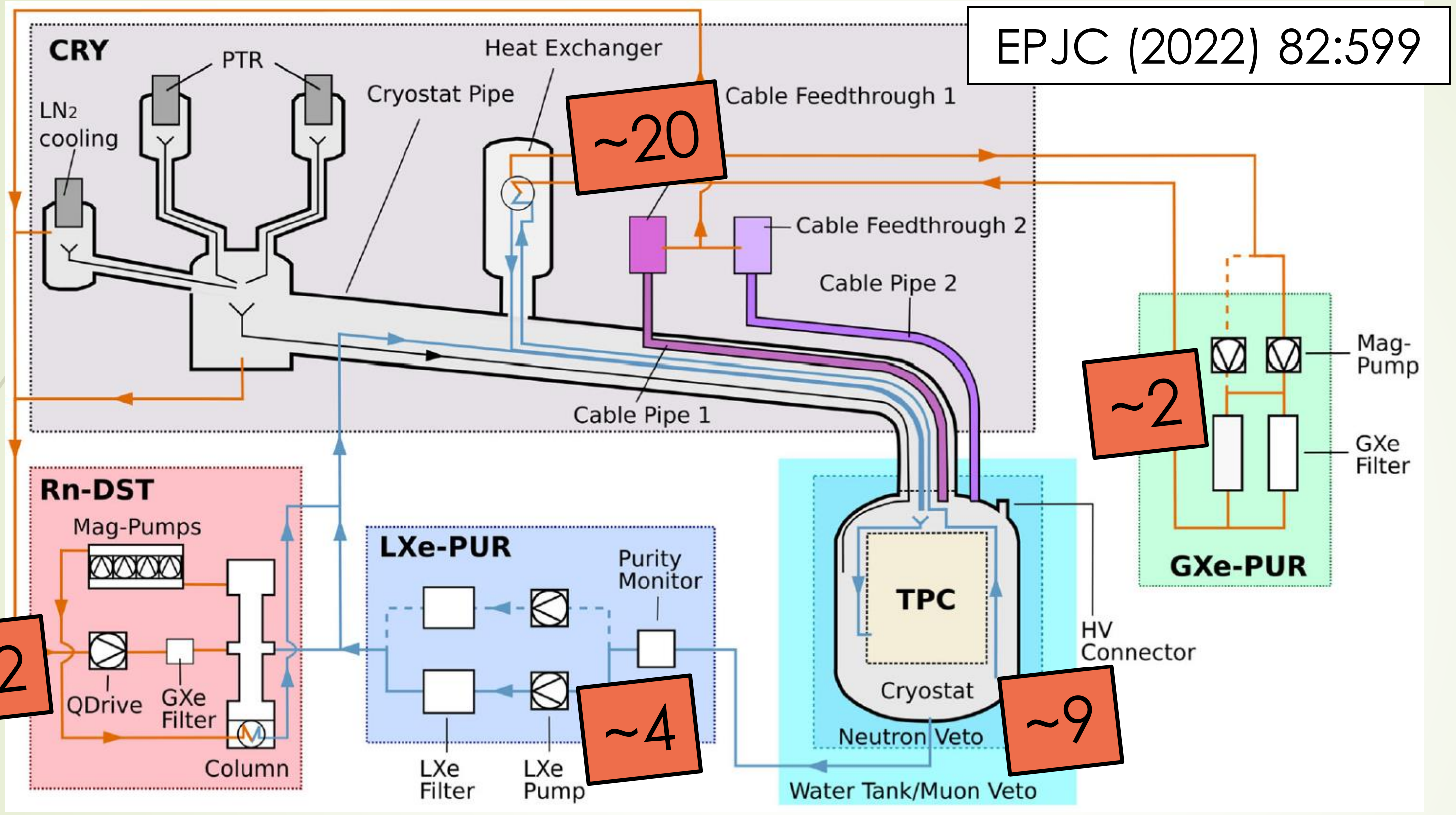
- Radon (in particular ^{222}Rn) is serious source of background in many astroparticle physics experiments.
- Noble gas: Diffusion!
- Followed by chain of radioactive isotopes.
- Not only radon in environment (atmosphere), but also radon sources in detector (part of natural uranium/thorium decay chains).
- Recoil- and diffusion-driven radon emanation.
- Radon mitigation requires
  - Radon removal

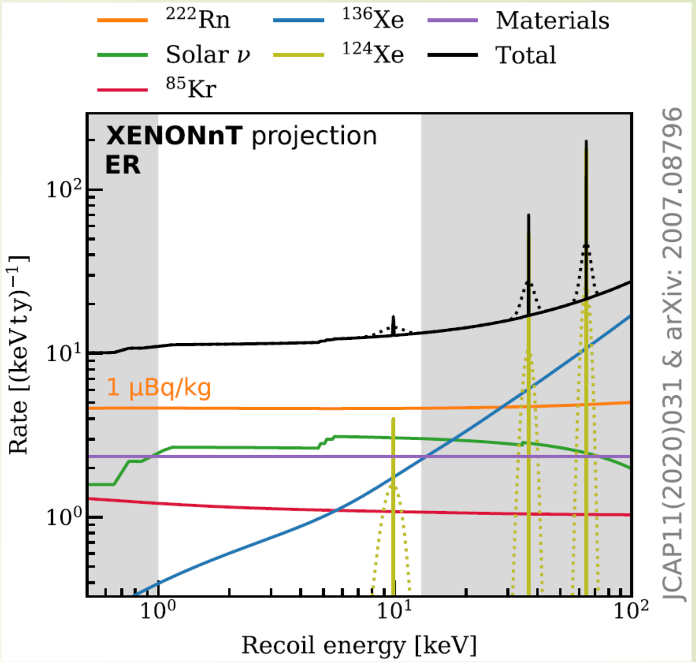
    and
  - Radium removal (mother element).

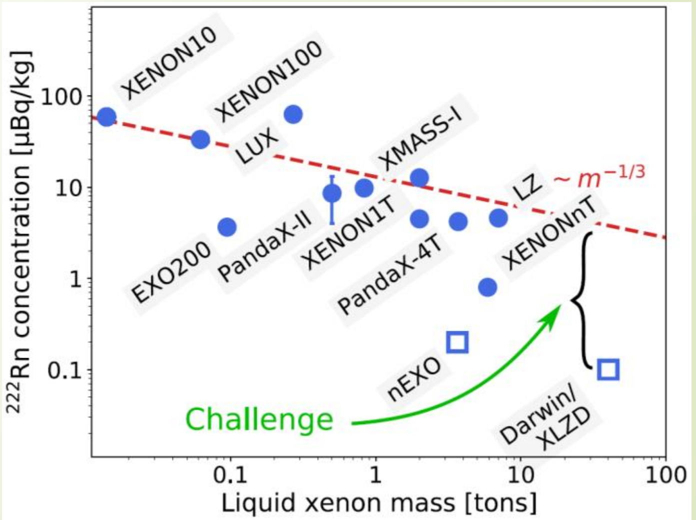


Radon background in XENONnT

All units [mBq]



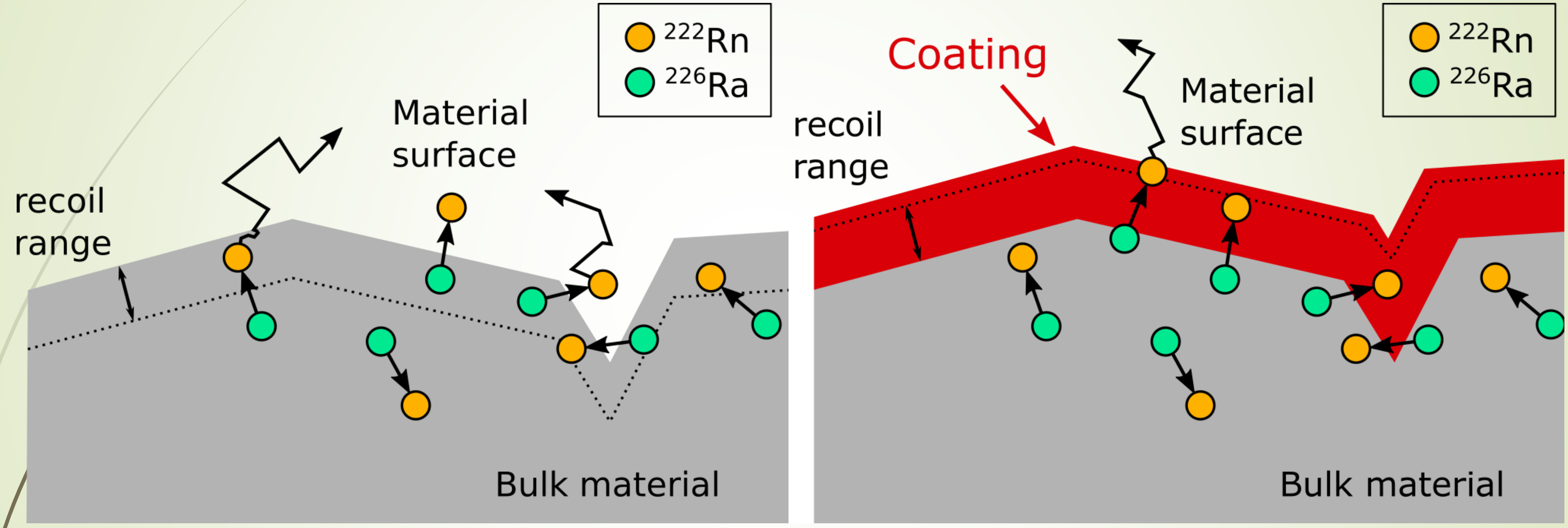




- ^{222}Rn is dominant background source in XENONnT.
- Various sources distributed over entire experimental infrastructure.
- <1 µBq/kg achieved (material selection and cleaning, online Xe distillation).
- Future experiments require even stronger radon supression.

→ Novel radon mitigation techniques!

Radon mitigation by surface coating



- Idea: A Rn-tight, clean (Ra-free) surface coating blocks Rn-emanation.
- Should work for recoil-driven (86 keV) AND diffusion-driven emanation.

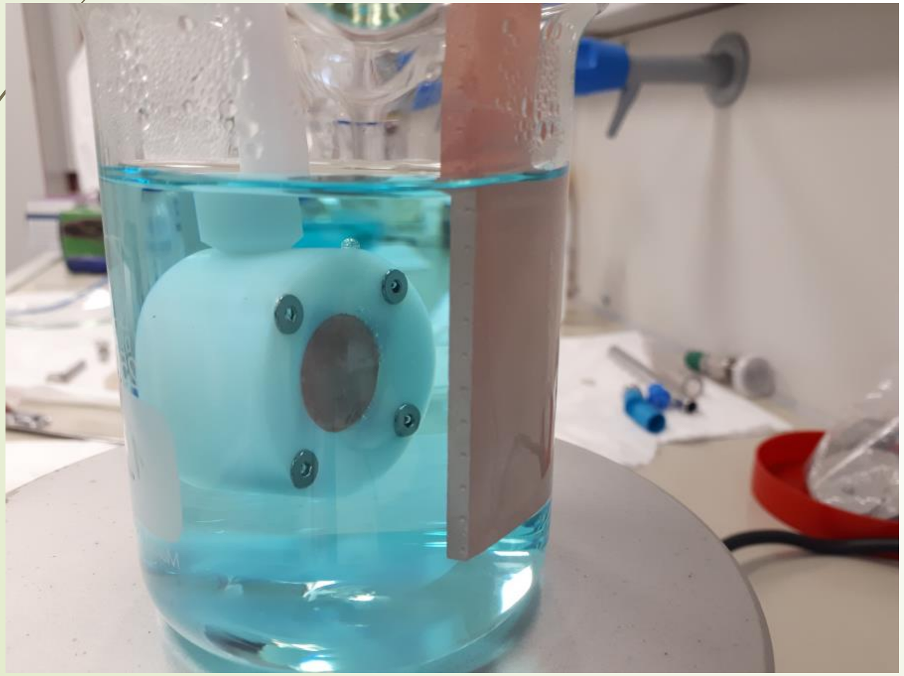
Rn reduction factors for thoriated weldig rods

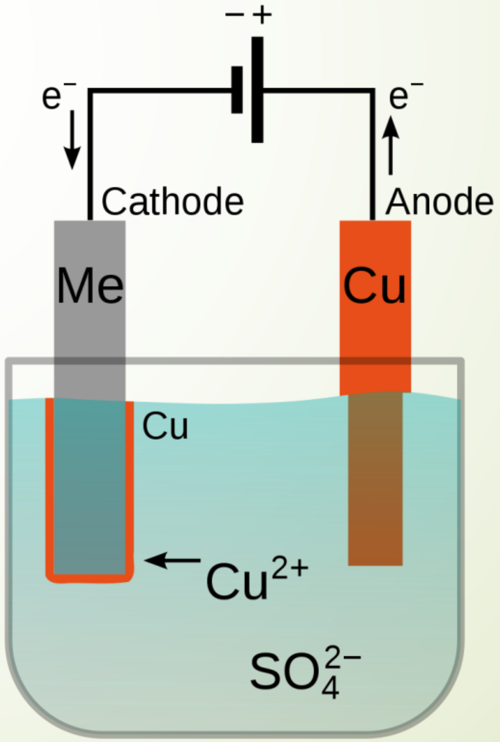
Method	Coated material	Company	^{220}Rn reduction	^{222}Rn reduction
Physical vapor deposition (sputtering)	Ti	Europcoating	1 – 5	~1
Chemical vapor deposition	C-H	Innovative Coating Solutions	~3	~1,5
Plasma deposition	Cu	Dr. Laure	2 – 20	---

- Measurements with electrostatic detectors (^{220}Rn) and proportional counters (^{222}Rn).
- 4% thoriated welding rods: First simple metallic Rn-emanating sample.
- Little ^{220}Rn reduction and essentially no ^{222}Rn reduction observed.
- Best result for „hot“ plasma depsoition, but re-evaporation due to hot substrate.

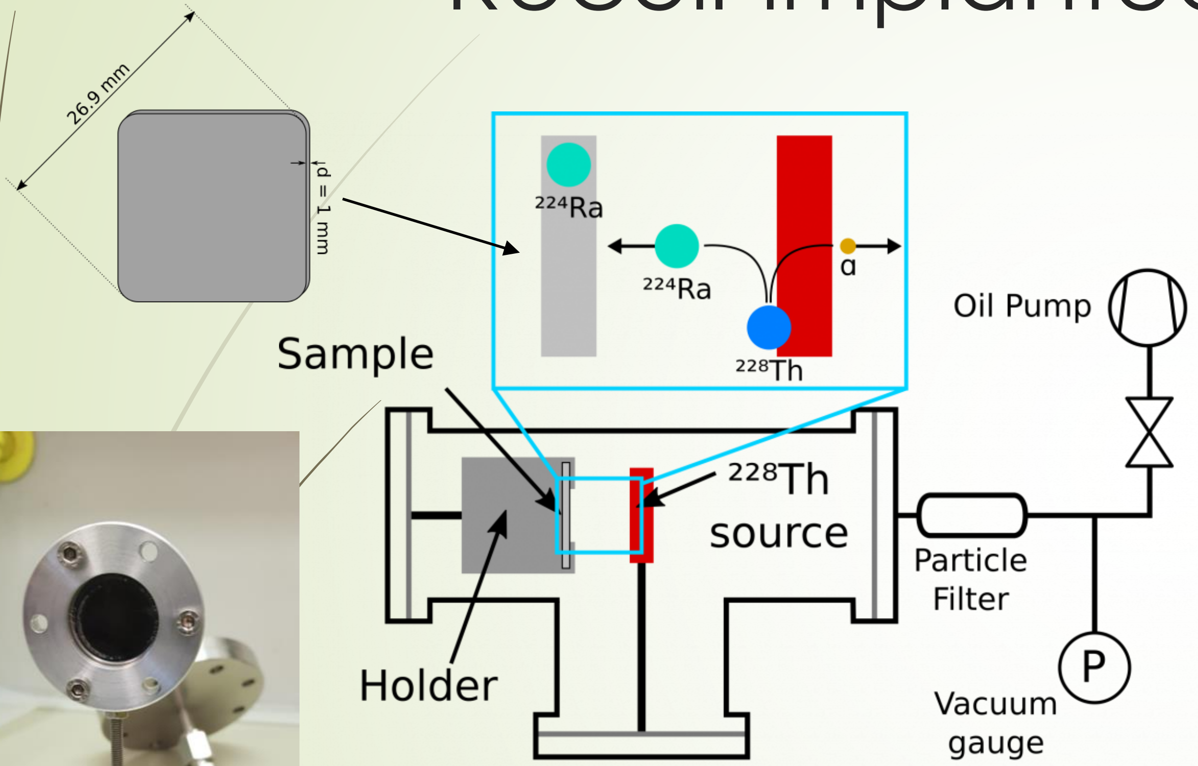
Copper electro-deposition

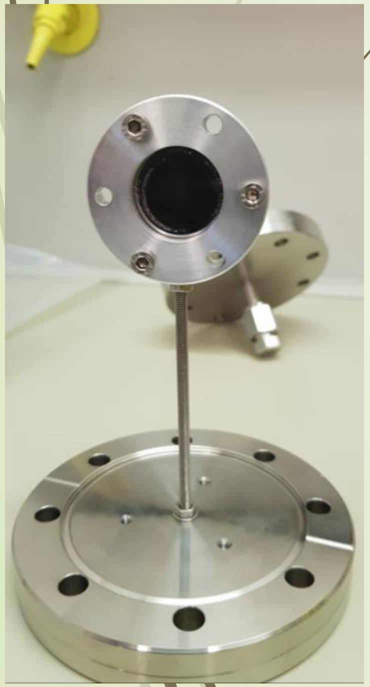
- Motivated by experience with electro-formed copper (clean!).
- In-house development at MPIK: ~5 µm thick Cu layer.
- Good mechanical stability on tungsten.
- Efficient blocking of ^{224}Ra alpha-decay.
- ^{220}Rn reduction factor of ~100 for thoriated tungsten rods observed!

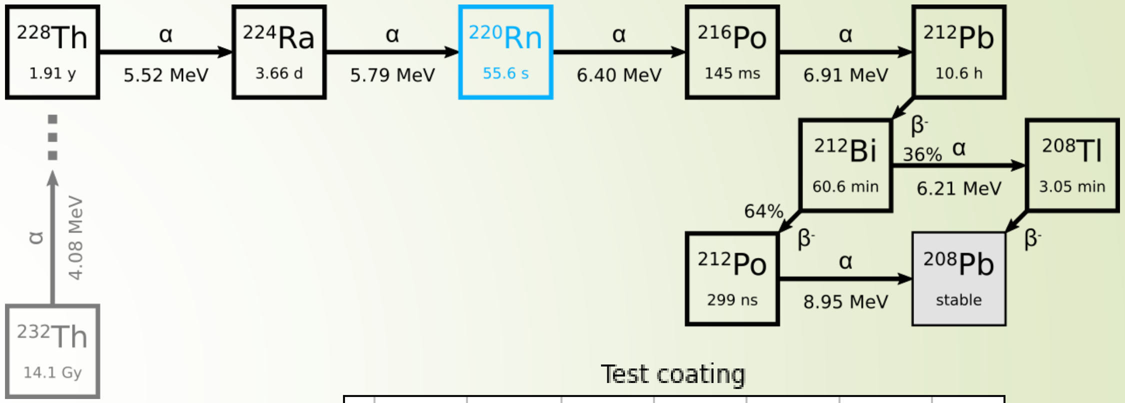




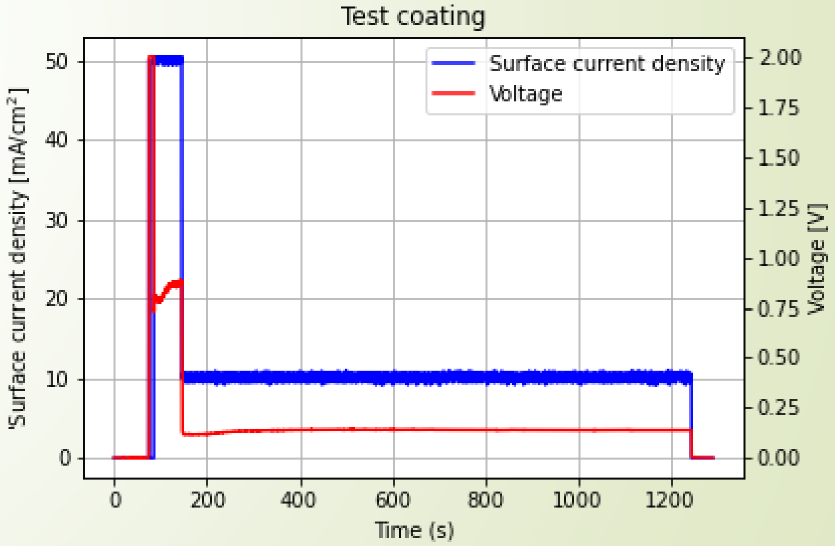
From tungsten to stainless steel (SS): Recoil-implanted ^{224}Ra





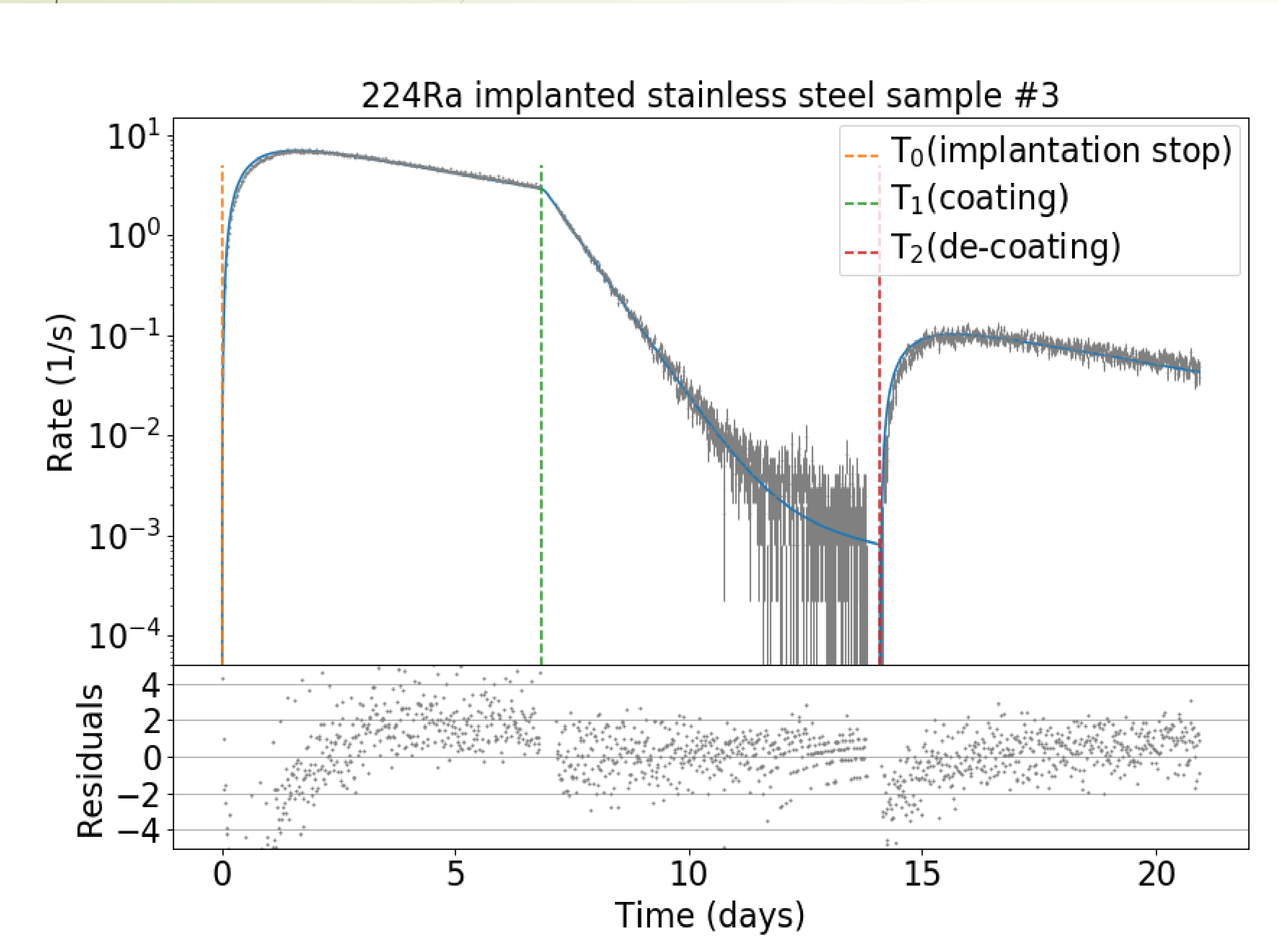


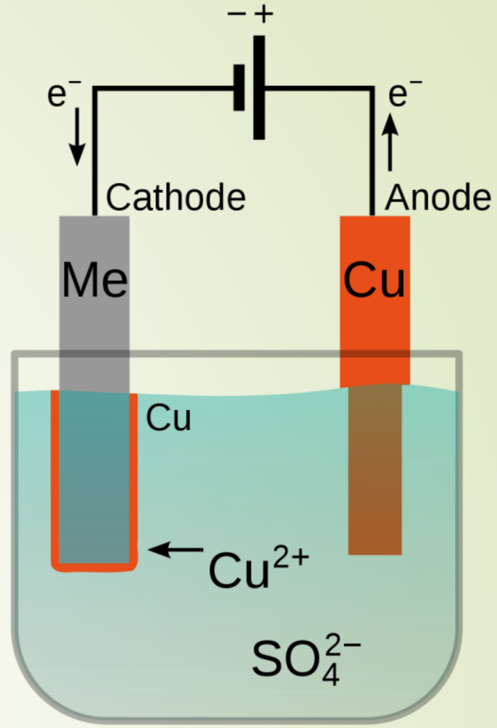
- ^{228}Th source recoils ^{224}Ra into SS disc.
- → Short-lived (t_H=3.7 days) ^{220}Rn-emanating SS sample.
- Easy to produce, relatively high activity.
- Re-consider plasma deposition with improved samples.



Procedure modification : Adhesion layer (~1 μm) for mechanical stability.

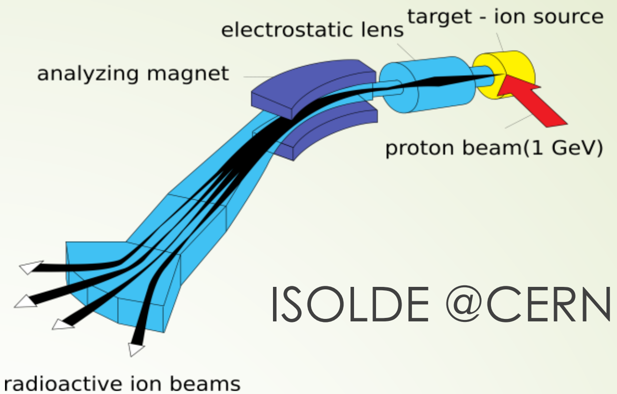
Coating (and de-coating) of ^{224}Ra-implanted SS disc



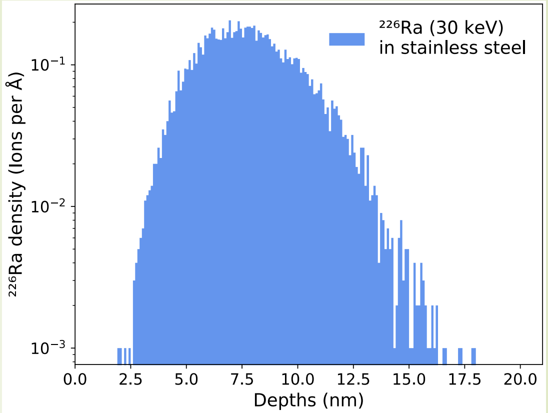


- ^{224}Ra decays on SS disc.
- ^{220}Rn is emanated and measured through ^{212}Po alpha-decays.
- Possible issue: Activity wash-off in electrolyte.
- Implantation → coating → de-coating
  - Upper limit from coating.
  - Lower limit from de-coating.
- Results for reduction factor R: **20 < R < 1000.**

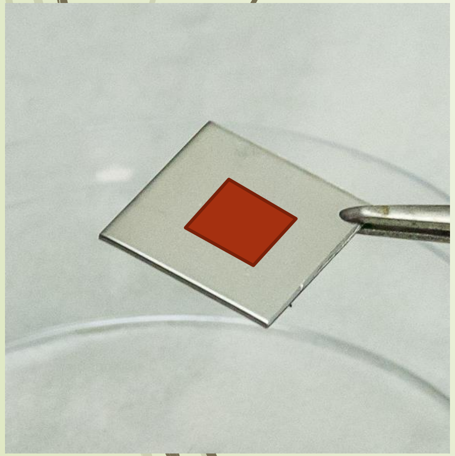
^{222}Rn on stainless steel



ISOLDE @CERN



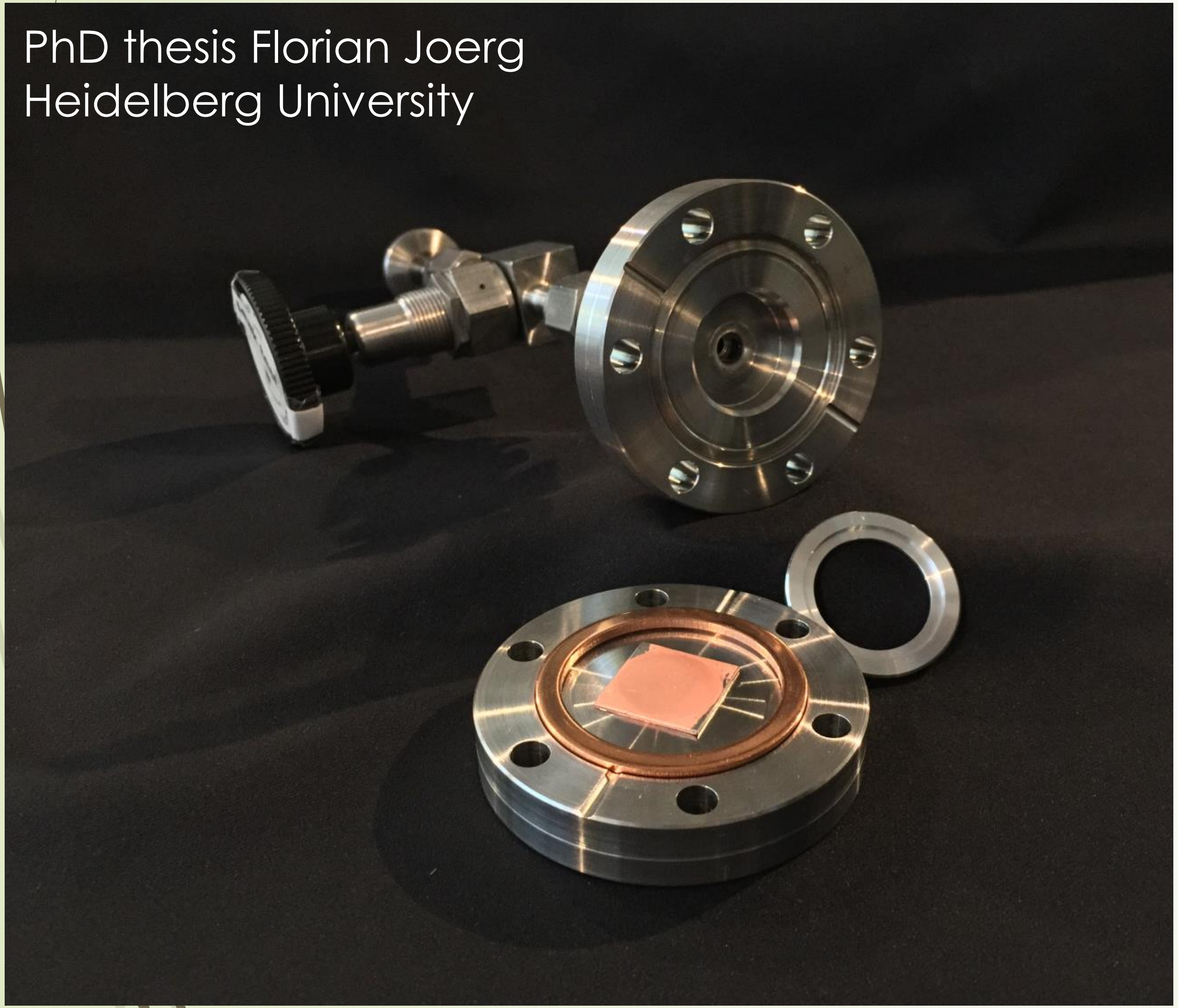
- ^{226}Ra-implanted stainless steel discs (2cm x 2cm).
- Produced at ISOLDE facility (CERN).
- 30 keV implantation energy.
- 2 test samples produced in 2017.
- 20 new samples approved (not only stainless steel), production in 2023.



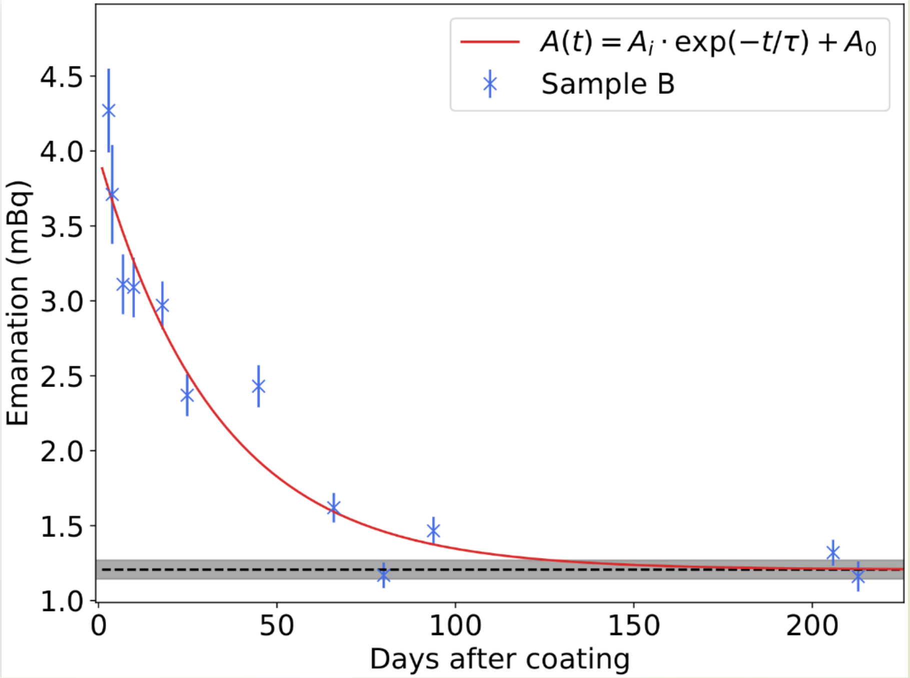
Measurement		Result (Bq)
^{222}Rn emanation	a	2.07 ± 0.03 (stat) ± 0.04 (syst)
	b	2.00 ± 0.03 (stat) ± 0.04 (syst)
γ-spectrometry	a	7.4 ± 0.1 (stat) ± 0.9 (syst)
	b	8.4 ± 0.3 (stat) ± 1.0 (syst)
α-spectrometry	a	8.7 ± 0.1 (stat) $^{+2.0}_{-1.8}$ (syst)
	b	9.1 ± 0.1 (stat) $^{+0.7}_{-0.4}$ (syst)

- Sample characterization published in *Appl. Rad. Isot. 194 (2023) 110666.*
- Alpha measurement:
  - ~8.5 Bq ^{226}Ra activity.
  - Central deposition confirmed.
- Gamma measurement confirms alpha measurement.
- Direct ^{222}Rn emanation test with proportional counters:
  - Sample a: (2.00 ± 0.05) Bq
  - Sample b: (2.07 ± 0.05) Bq
- Wipe test: Less than 1% of activity removed.

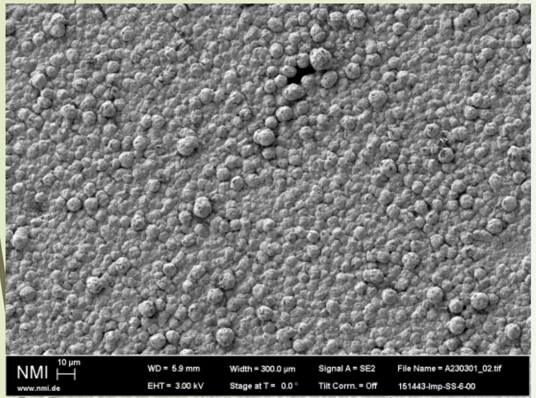
Coating of the 1st ISOLDE sample

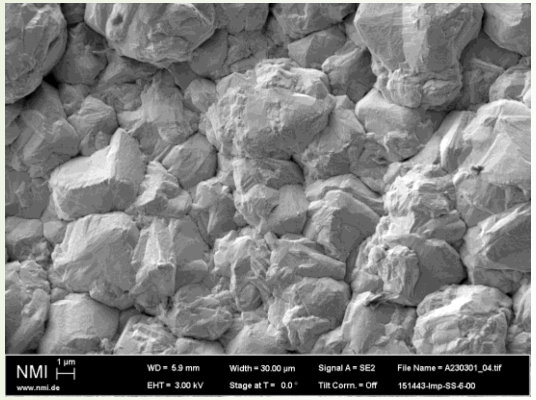


- Standard MPIK electro-chemical copper-on-steel coating recipe applied.
- Unexpected large ^{222}Rn reduction factor!
- Increased over time from 450 to 1700: Copper layer re-arrangement?

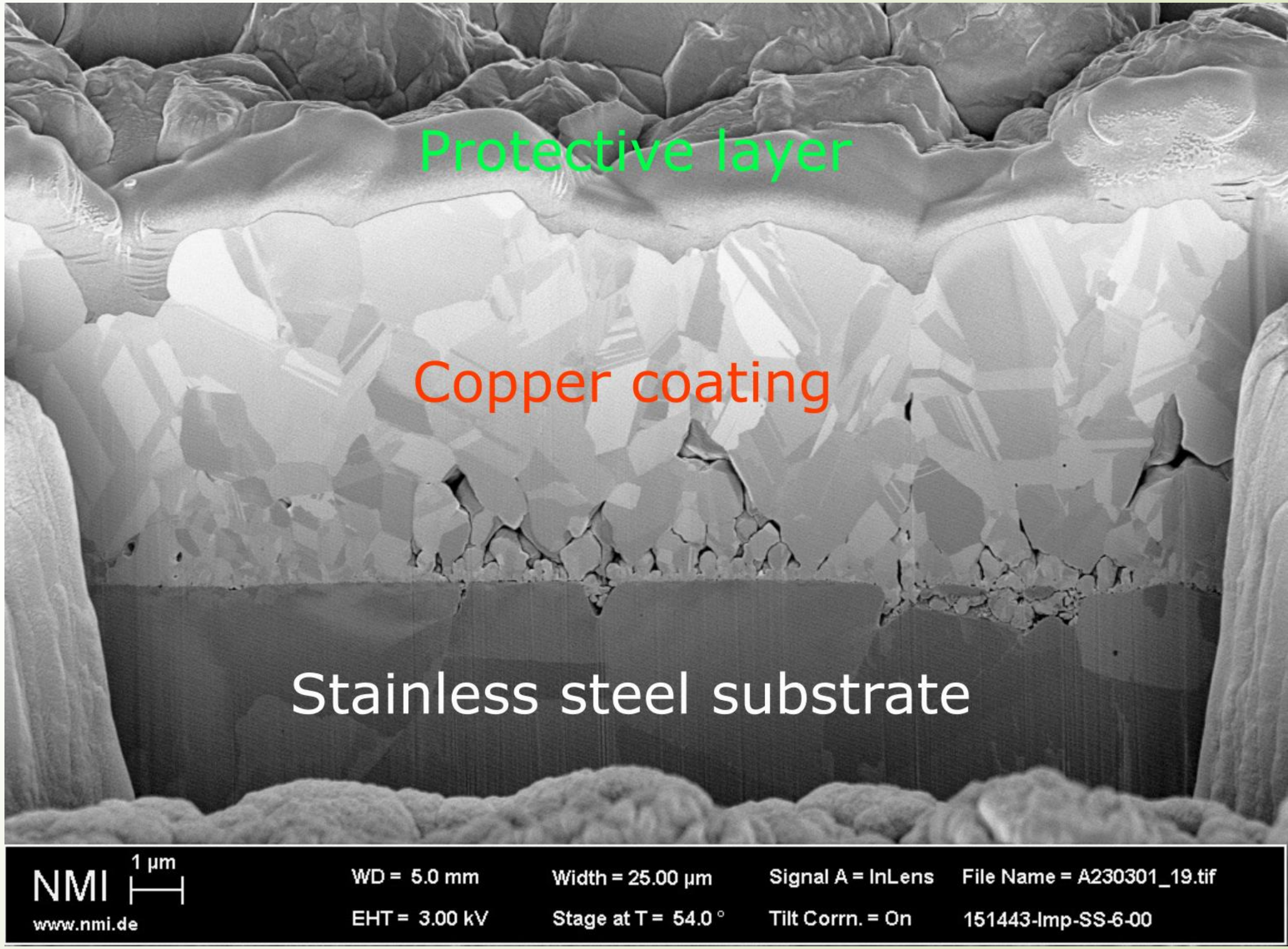


Secondary Electron Microscopy (SEM) investigation of our coating





- Rough surface texture with spherical structures.
- Unhomogeneous adhesive layer with small grain size and holes.
- Tight cover layer with larger grains.



Summary and outlook

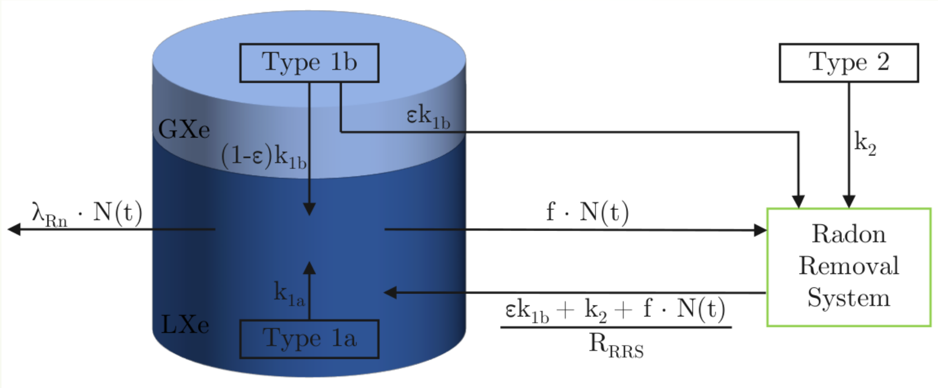
New electrode design for coating of cylindrical vessel

- Surface coating is a promising technology for radon mitigation.
- Electro-deposition of copper showed best reduction results.
- Future work focus on:
  - Coating characterization (SEM, X-ray, …)
  - Purity of coating:
    - Cleanliness of electrolye (Deposition of impurities).
  - Up-scaling:
    - Development of new setup to coat ~15 liter vessel.
  - New ISOLDE samples will allow for more systematic studies.

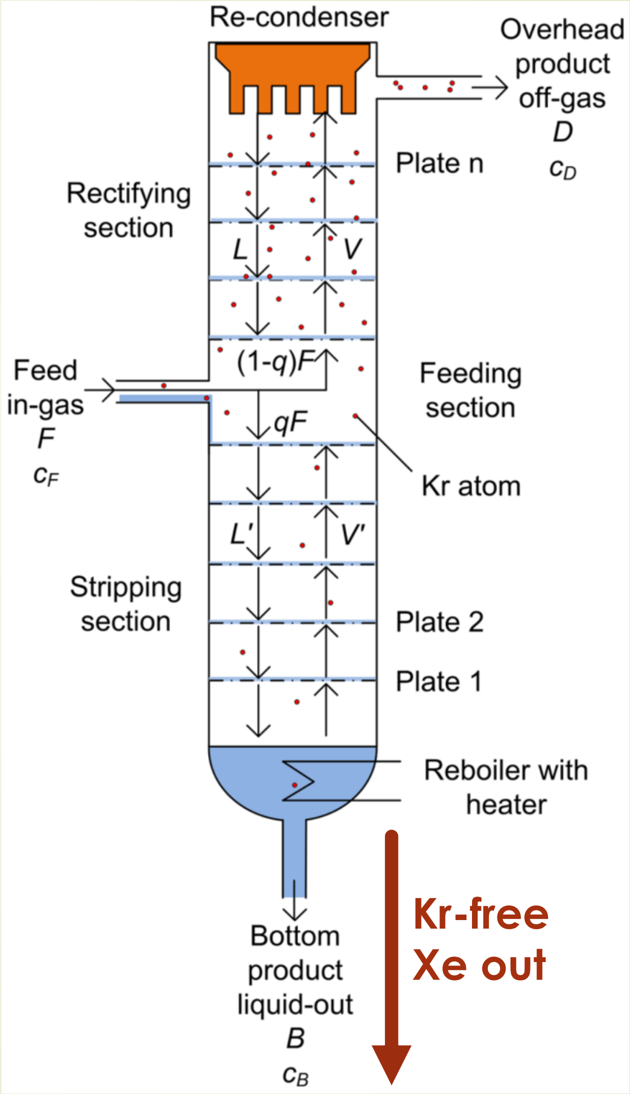
Backup

Radon removal by xenon distillation

- Radon is less volatile than xenon → reverse operation mode w.r.t. Kr removal.
- Feasibility demonstrated in Xenon100: EPJC (2017) 77:358
- Applied in XENON1T: 4.5 µBq/kg achieved: EPJC (2021)81:337
- High flow radon removal system developed for XENONnT: arXiv: 2205.11492
- **1.7 µBq/kg achieved**



- Upscaling only possible for high throughput system.
- Processing speed for DARWIN must be >=10 tons/day.
- Efficiency in power consumption and xenon holdup versus radon reduction is crucial.



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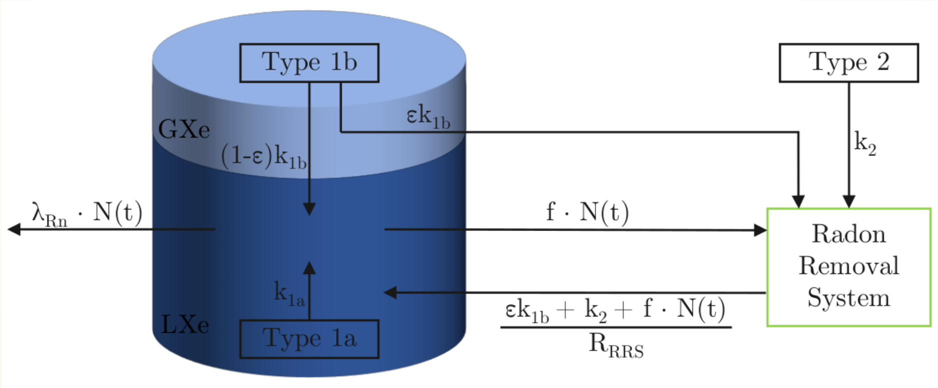
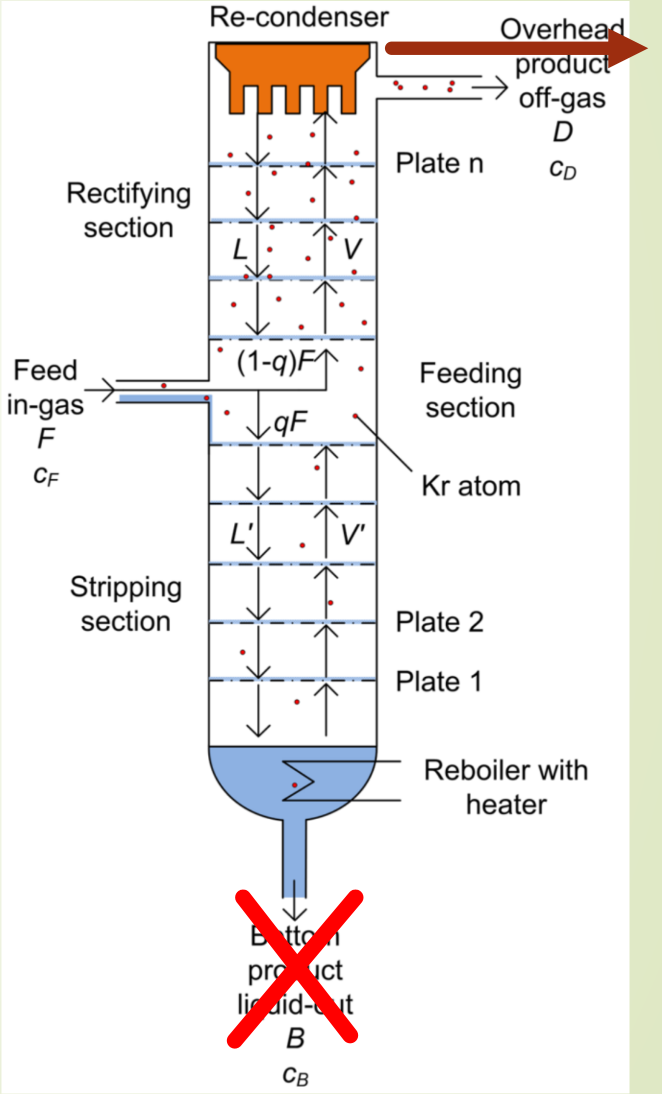


Rn-free Xe out



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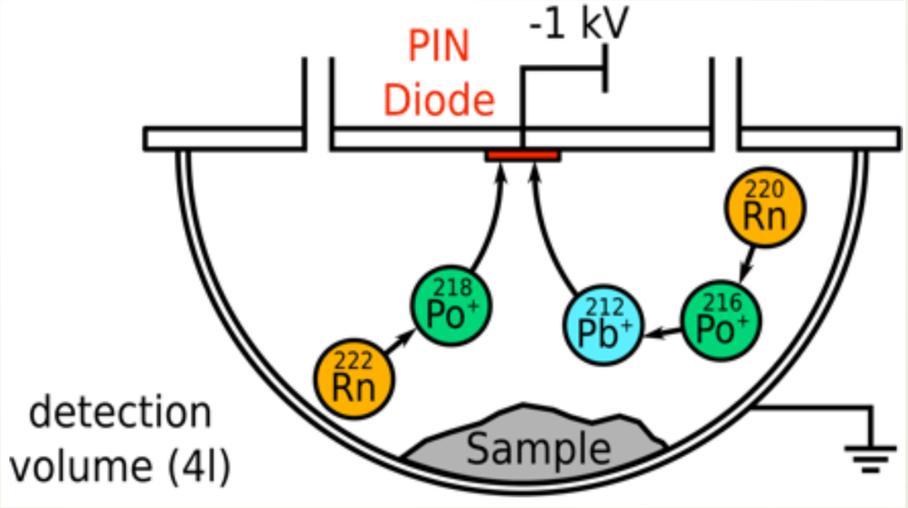
- Processing speed for DARWIN must be >=10 tons/day.
- Efficiency in power consumption and xenon holdup versus radon reduction is crucial.

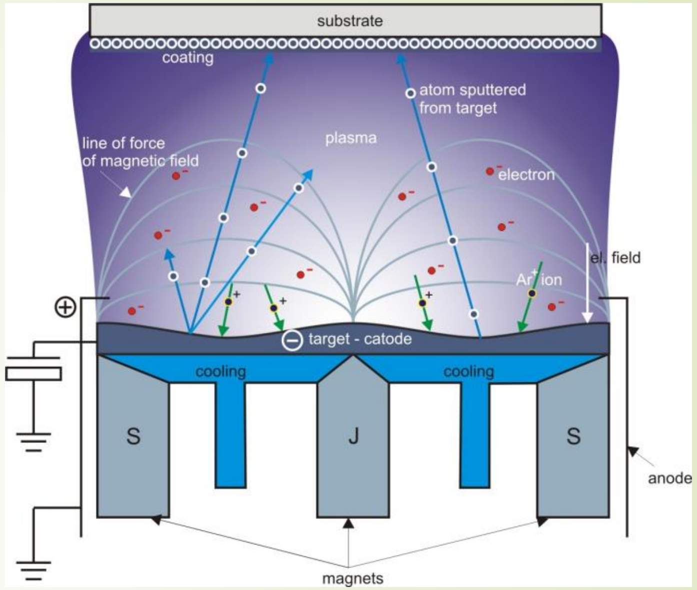
^{222}Rn emanation measurements at MPIK

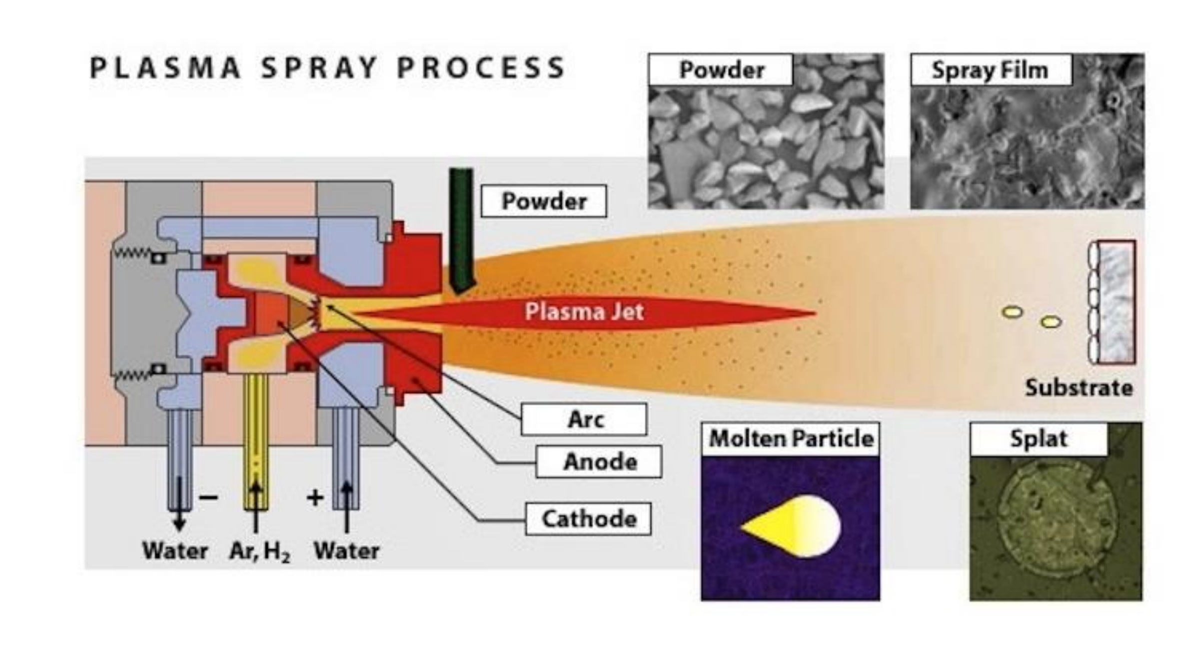
- MPIK ^{222}Rn infrastructure:
  - >20 ultralow background miniaturized proportional counters
    - Sensitivity: ~10 atoms.
    - 8 parallel counting lines.
  - Fully automated ^{222}Rn concentration system (AutoEma).
  - ~15 sample vessels (0.1 – 80 lit.).
  - 3 electro-static ^{222}Rn monitors.

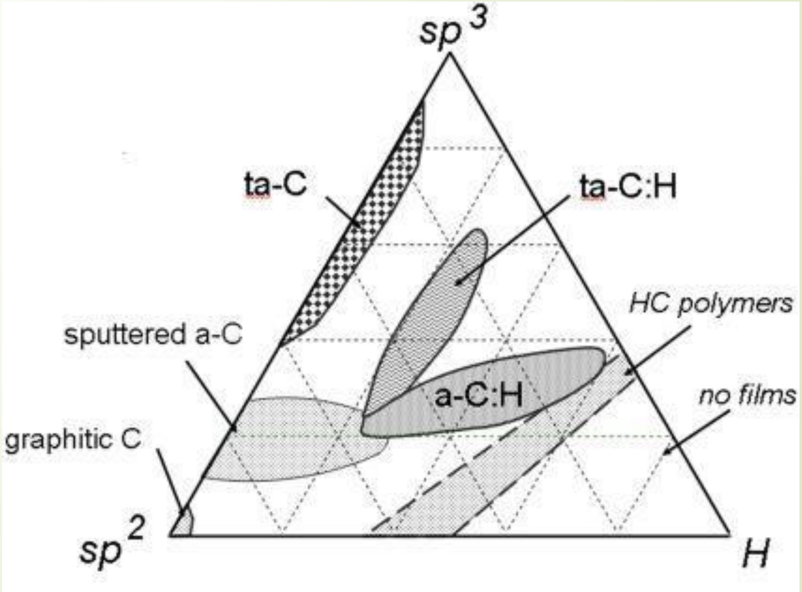


Investigated vacuum coating techniques

- Physical vapor deposition (PVD).
  - Titanium sputtering.
- Chemical vapor deposition (CVD).
  - Amorphous hydrogenated carbon coating (a-C:H).
- Copper plasma deposition.

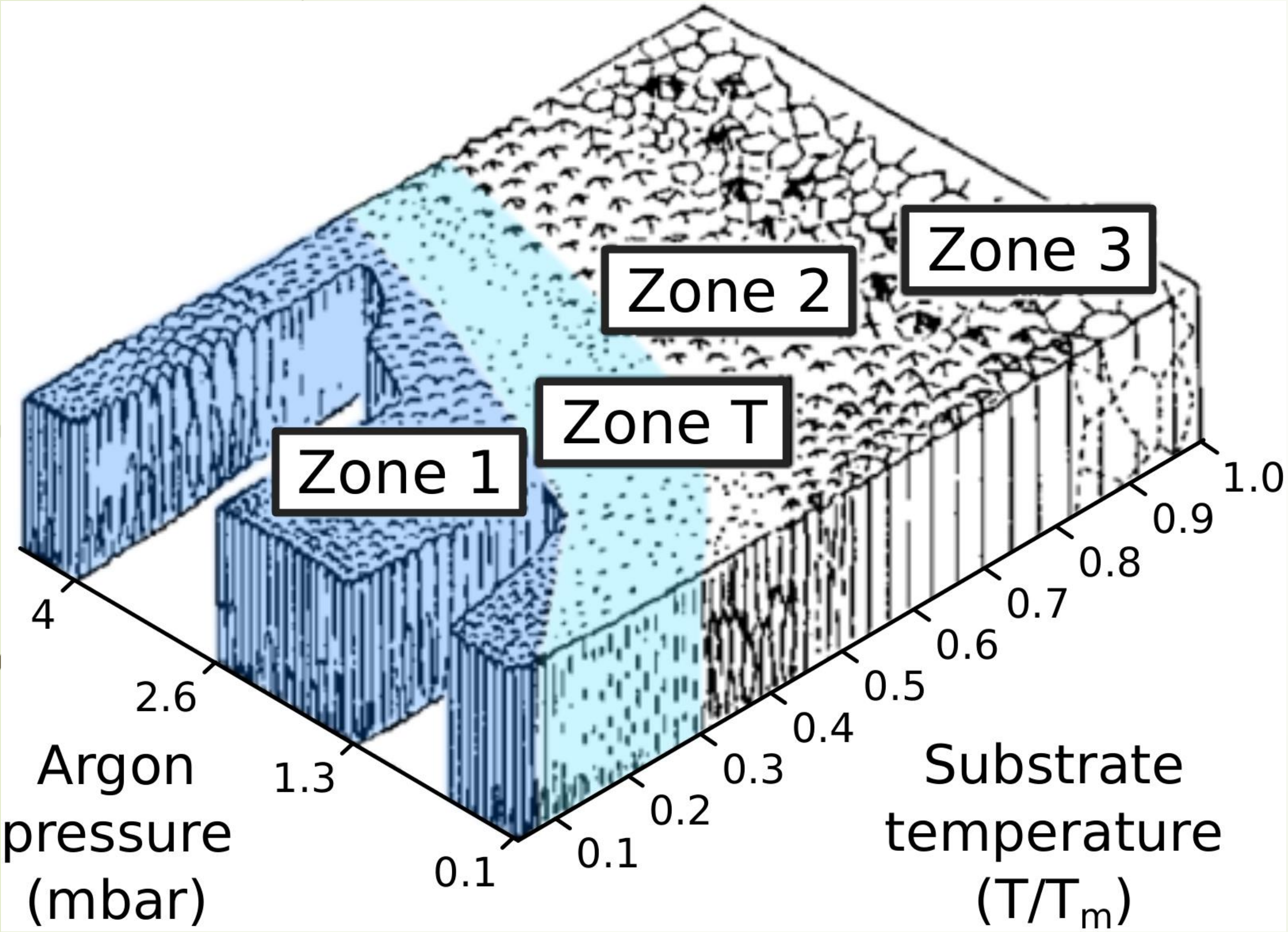






J. Robertson, Materials Science and Engineering, 2002 (129-281)

Thornton structure zone model:



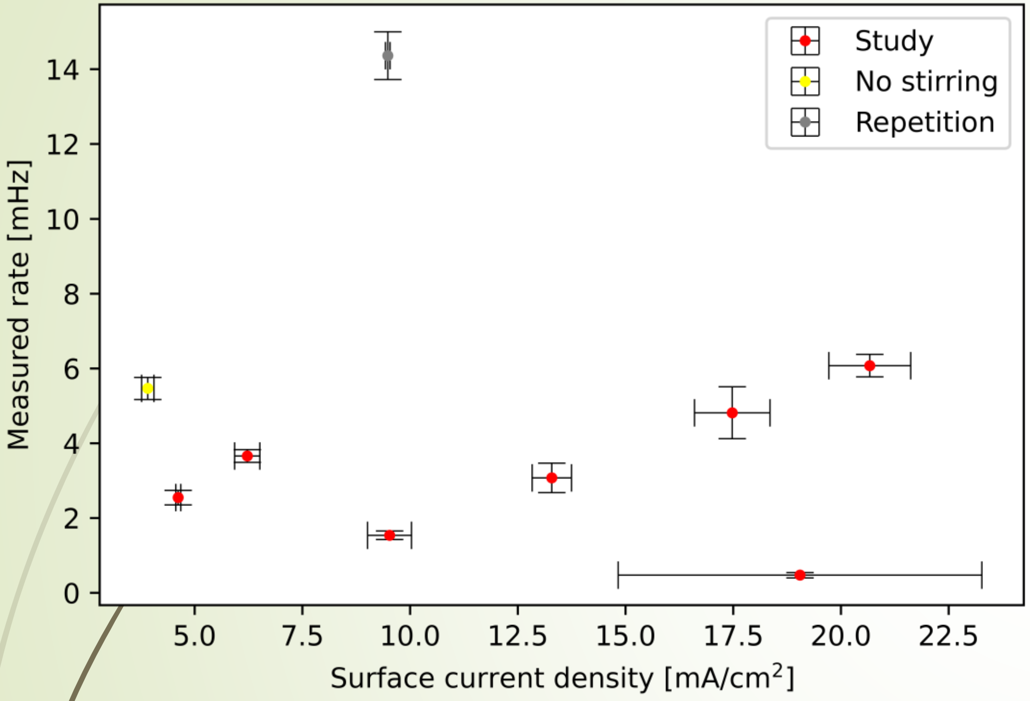
- Sputtering is low temperature process.
- Growth of vertically aligned grain boundaries.
- May block reactive gases (corrosion protection), but „diffusion highway“ for noble gases.
- Focus on high temperature applications (plasma coating) or non vacuum-growth technique.
- But what about CVD?

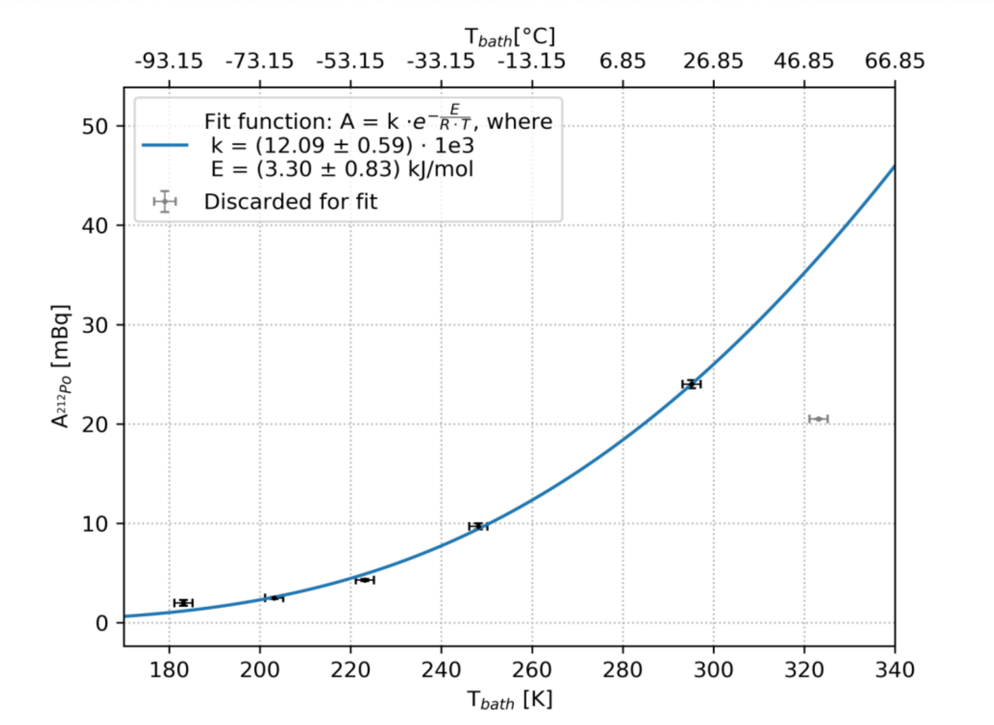
Summary of results from 1st phase

Method	Coated material	Company	^{220}Rn reduction	^{222}Rn reduction
Sputtering	Ti	Europcoating	1 – 5	~1
Plasma deposition	Cu	Dr. Laure	2 – 20	---
Chemical vapor deposition	C	Innovative Coating Solutions	~3	~1,5
Electro chemical	Cu on W	MPIK	O(100)	1 – 8
Electro chemical	Cu on SS	MPIK	20 – 1000	---

- Focus on electro-chemical plating (but don't forget plasma deposition).

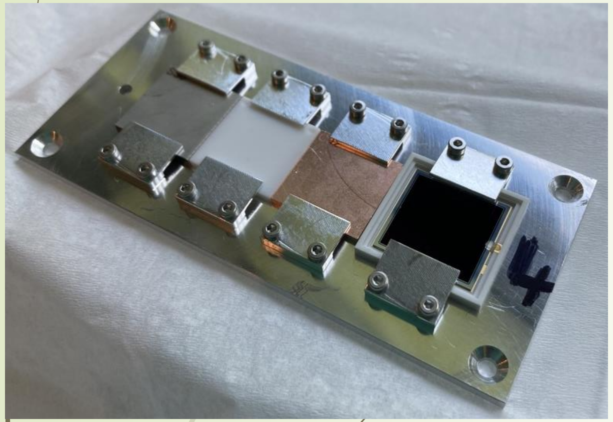
Optimization of Cu coating procedure and ^{220}Rn reduction results



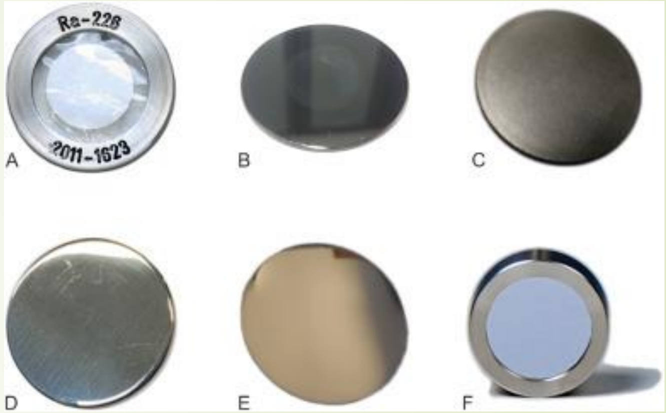


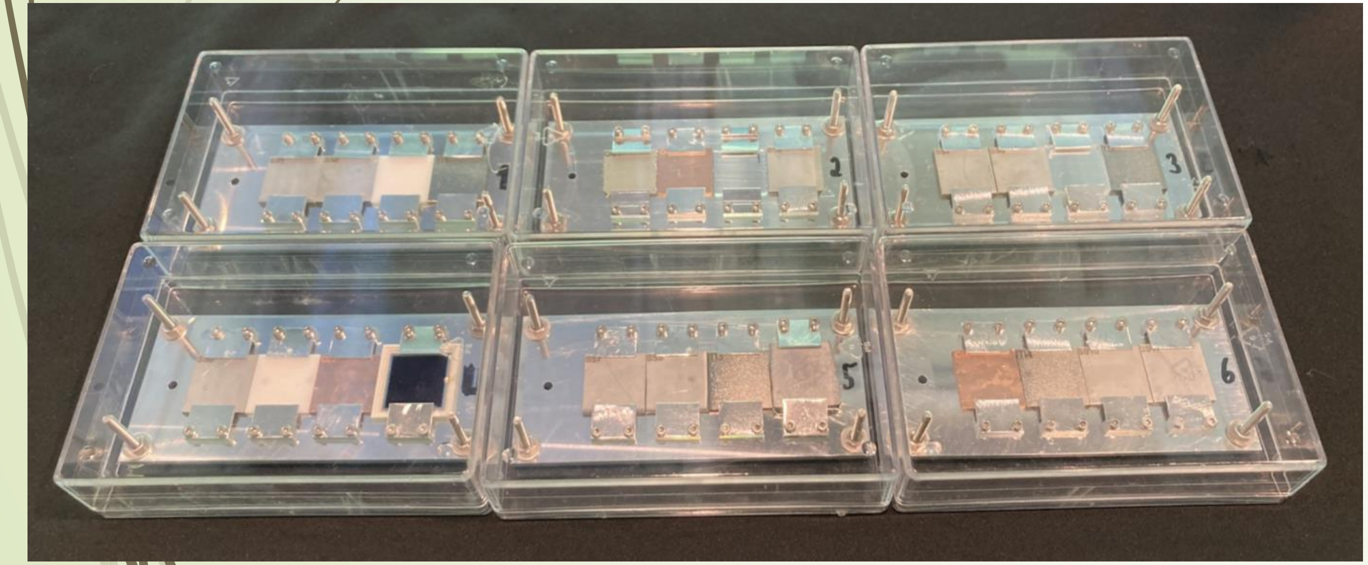
- Optimum surface current density identified.
- Avoid whisker growing by careful parameter control.
- Diffusion-driven emanation confirmed by tests at different temperatures.
- Even hints for slight ^{222}Rn reduction.

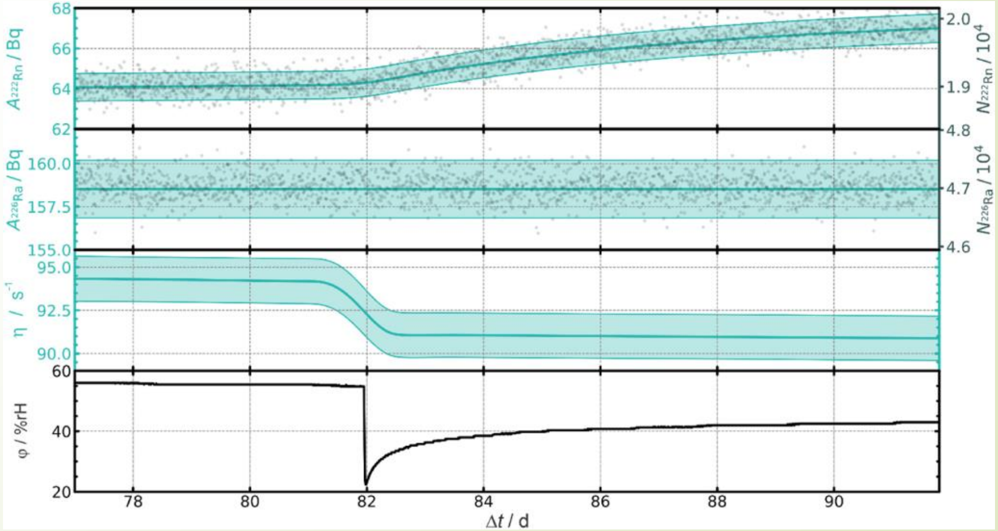
Novel ^{222}Rn sources



- ^{226}Ra implantation in SS, Cu, Ti, Pb, Ge, Si, PTFE, SiO_2, acrylic.
- 20 – 24 new samples.
- Similar development at PTB (Braunschweig) → Traceable ^{222}Rn sources.
- Deposition on an active Si detector allows online emanation rate monitoring.







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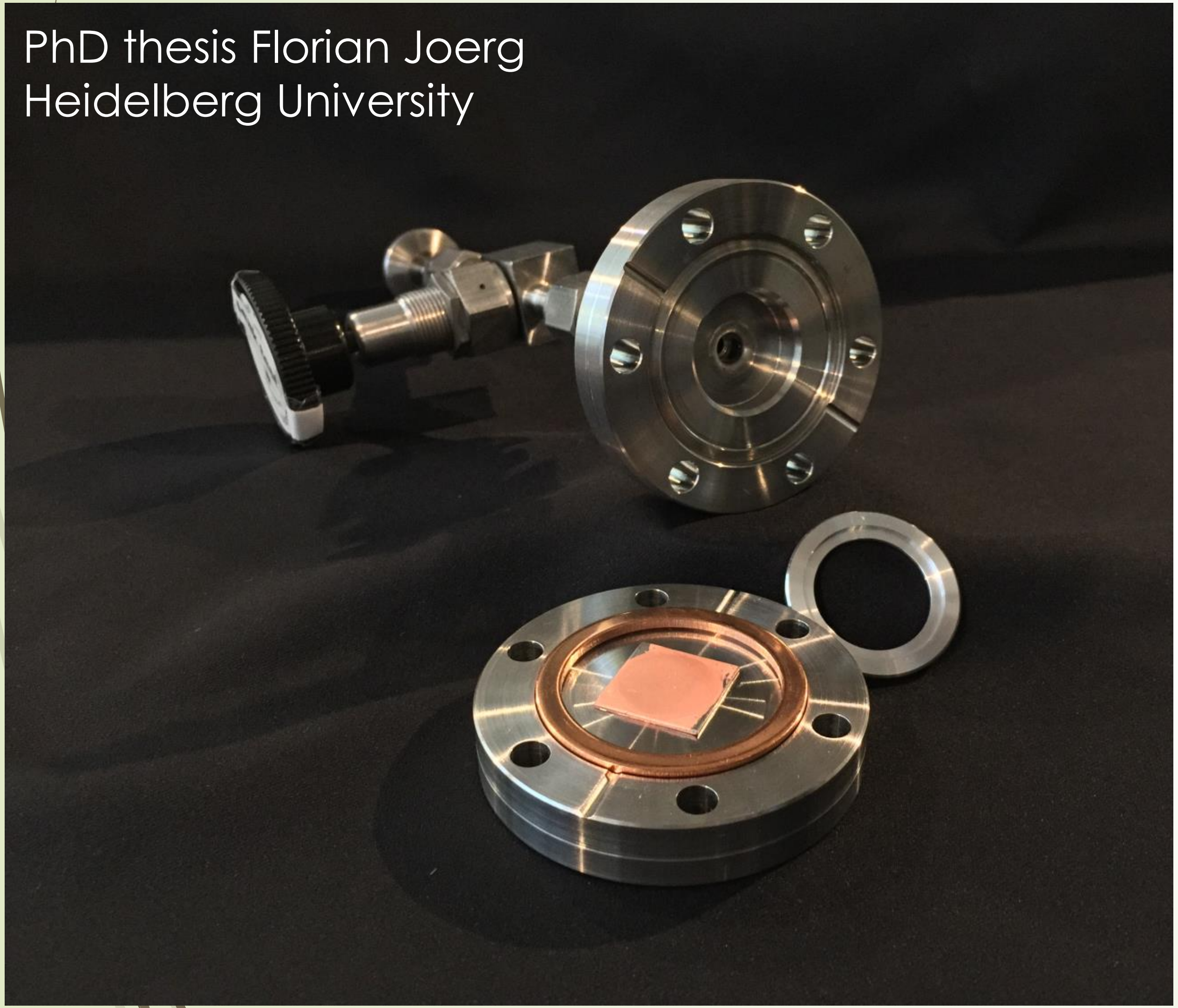
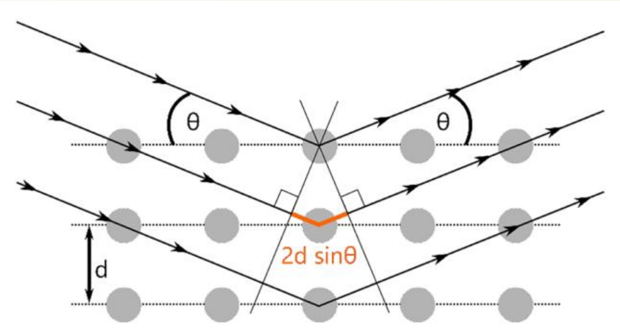
Coating of the 1st ISOLDE sample

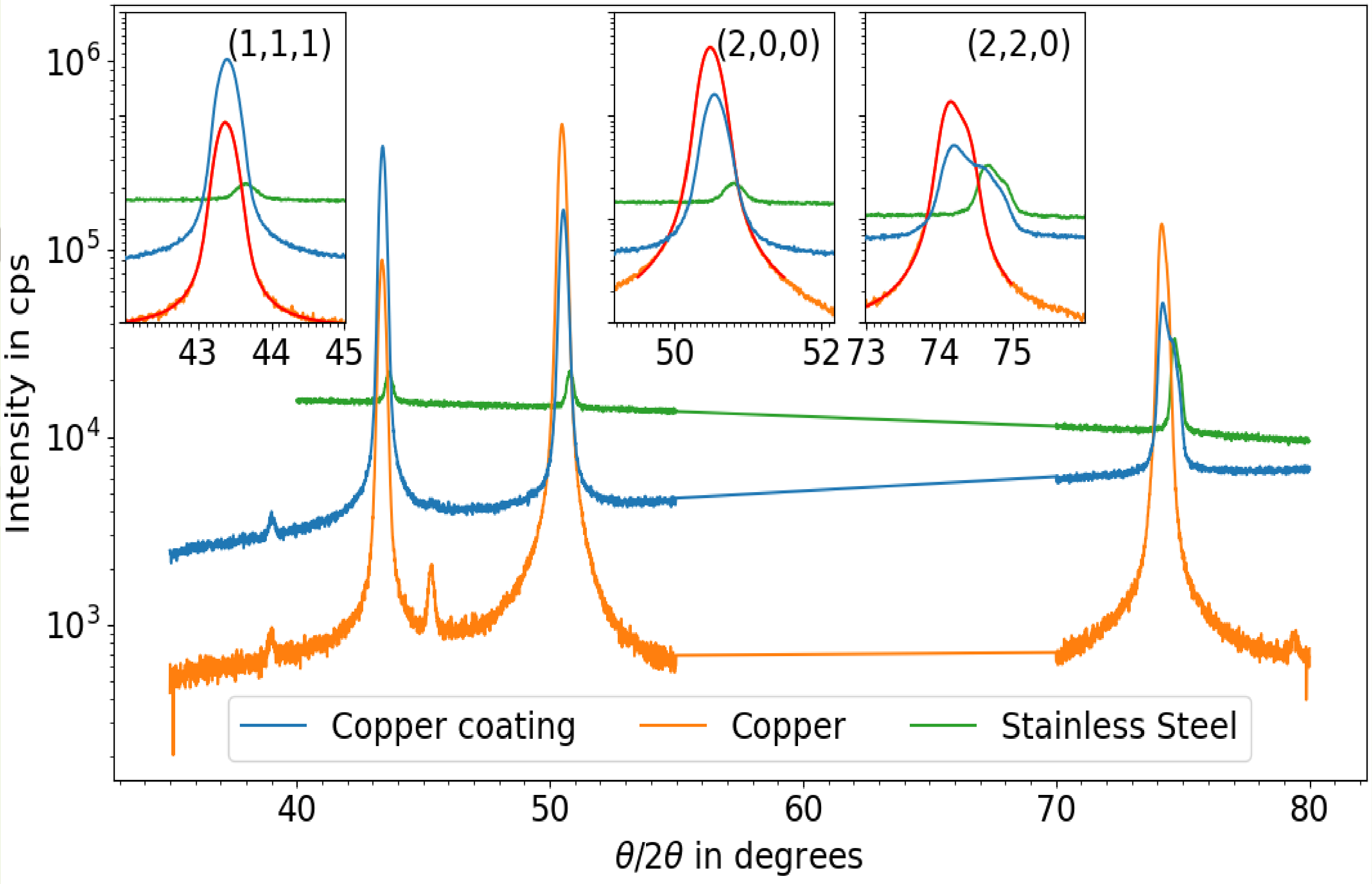


- Standard MPIK electrochemical Cu-on-SS coating recipe applied.
- ^{222}Rn emanation rate
  - Before coating: (2.00 ± 0.05) Bq.
  - After coating: (4.3 ± 0.3) mBq.
- Unexpected large ^{222}Rn reduction factor: ~465.
- Gamma spectroscopy results:

Activity [Bq]	^{226}Ra (186 keV)	^{222}Rn daughters
ISOLDE sample before coating	8.4 ± 1.0	6.0 ± 0.3
ISOLDE sample after coating	7.7 ± 1.0	7.2 ± 0.4
Electrolyte after coating	---	0.34 ± 0.02

X-ray diffraction





- Very preliminary study.
- Done at Heidelberg University (IMSEAM: Institute for molecular systems engineering and advanced materials).
- Basic features as expected, but some unexplained effects:
  - Amplitude ratios doesn't always match expectation (directionality in lattice?).
  - Peak positions slightly shifted (material stress?).
  - Not understood low intensity peaks.