Radon background in astroparticle physics experiments - *Past achievements and future challenges*

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Radon – Some properties

- Noble gas.
- Heaviest existing gas → not **so** noble.
- No stable isotopes → All radon is radioactive.
- Part of natural (primordial) radioactivity.
- Short-lived: ^{222}Rn (3.82 days) by far the longest living, rest much shorter
- Followed by a chain of radioactive heavy metals (a-, β- and γ-emitters)

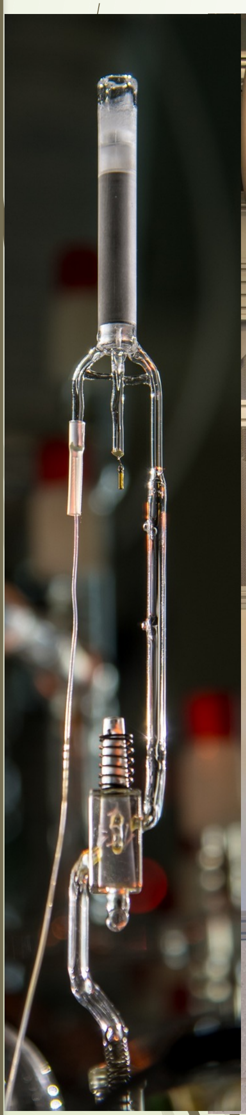
Radon detection (at low level)

- All radon detection by measurement of radioactivity
- Alpha emitter: „High“ energy signal

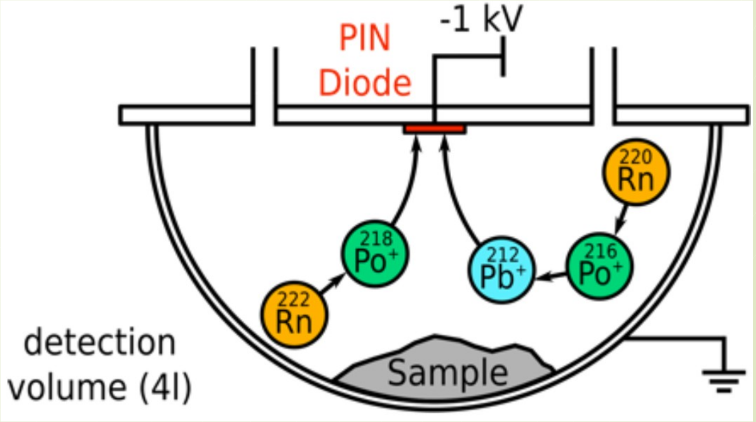
- Low-level → Requires radon collection
  - By cryo-trapping
  - By electrostatic collection of charged daughters
- Low-level → Requires clean materials and vacuum

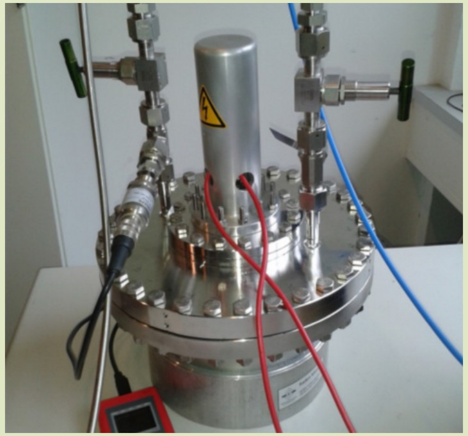
- Warning: ICP-MS / NAA has fantastic sensitivity for U/Th. But radioactive may be broken, so no information about Rn!

Material screening by ^{222}Rn emanation

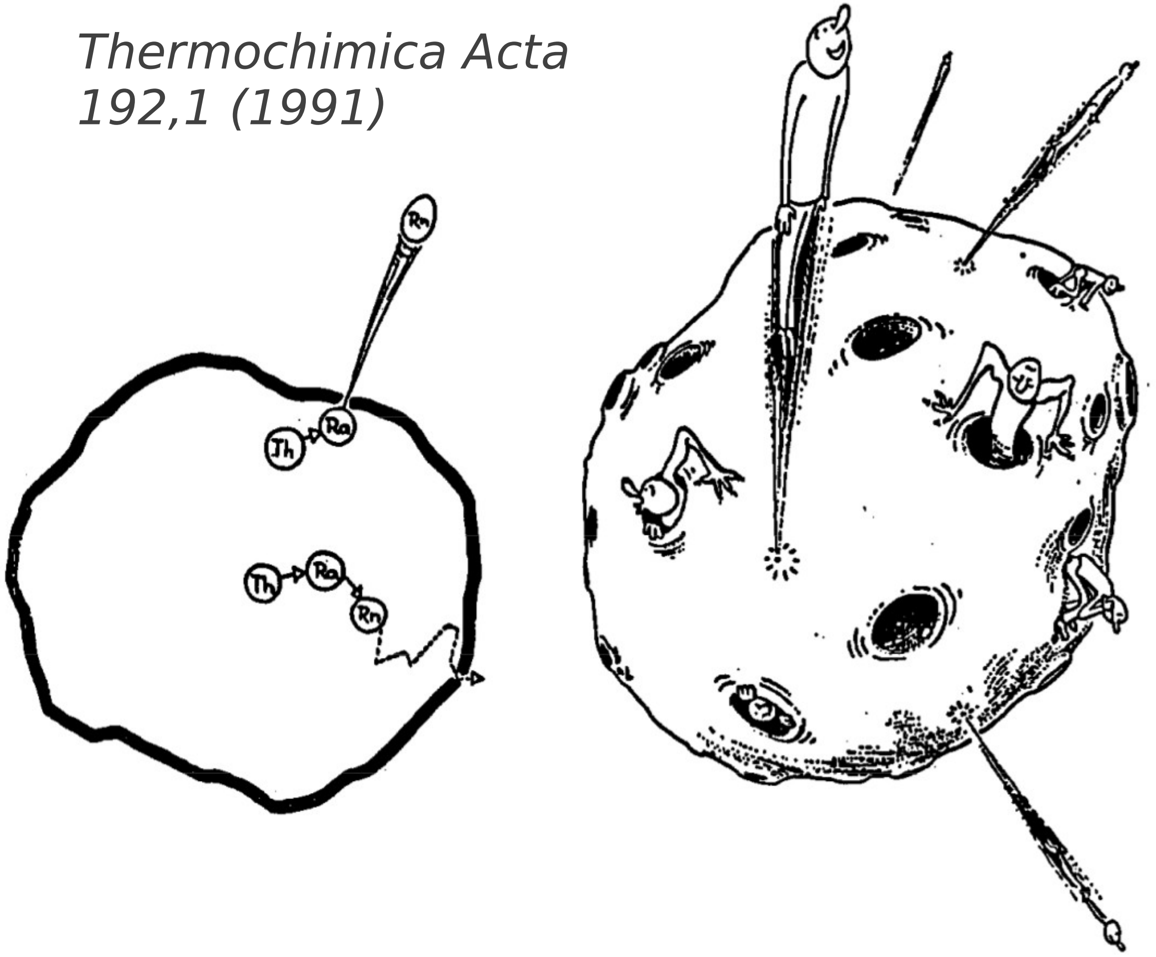


- ^{222}Rn-emanation rate provides complementary information
- MPIK ^{222}Rn infrastructure:
  - >20 ultralow background miniaturized proportional counters
    - Sensitivity: ~10 atoms.
    - 8 parallel counting lines
  - Fully automated ^{222}Rn concentration system (AutoEma).
  - ~15 sample vessels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





The radon emanation process

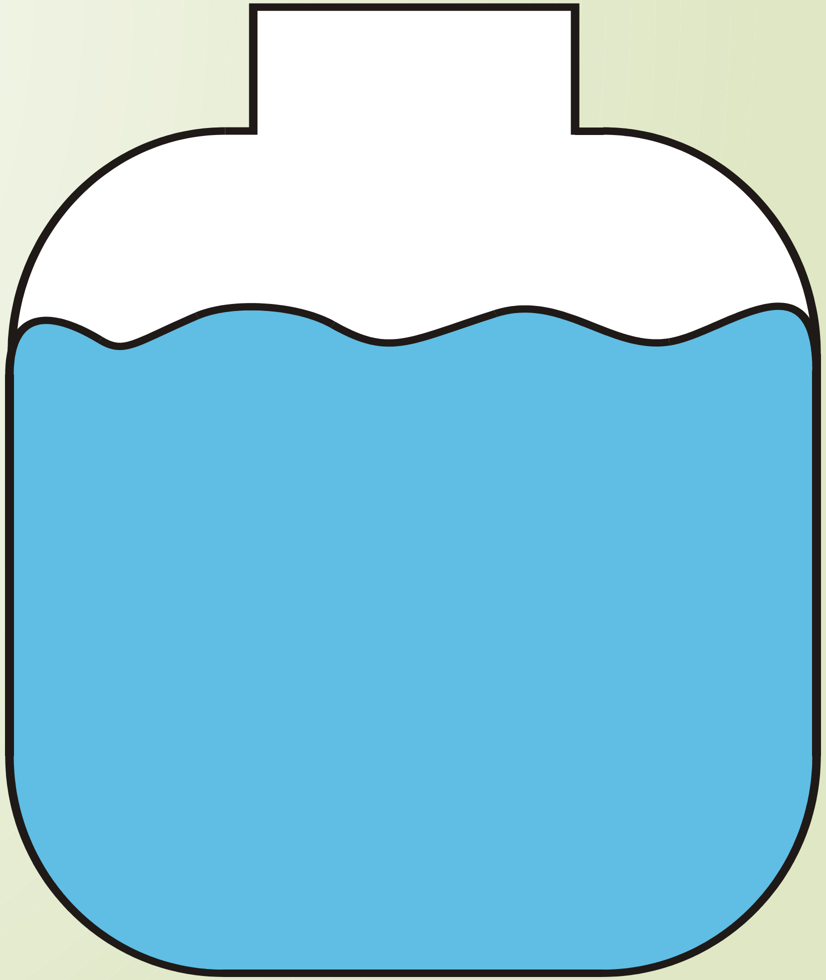


Thermochimica Acta 192,1 (1991)

Fig. 2. Scheme of the release of radon from the sample by recoil and diffusion mechanisms.

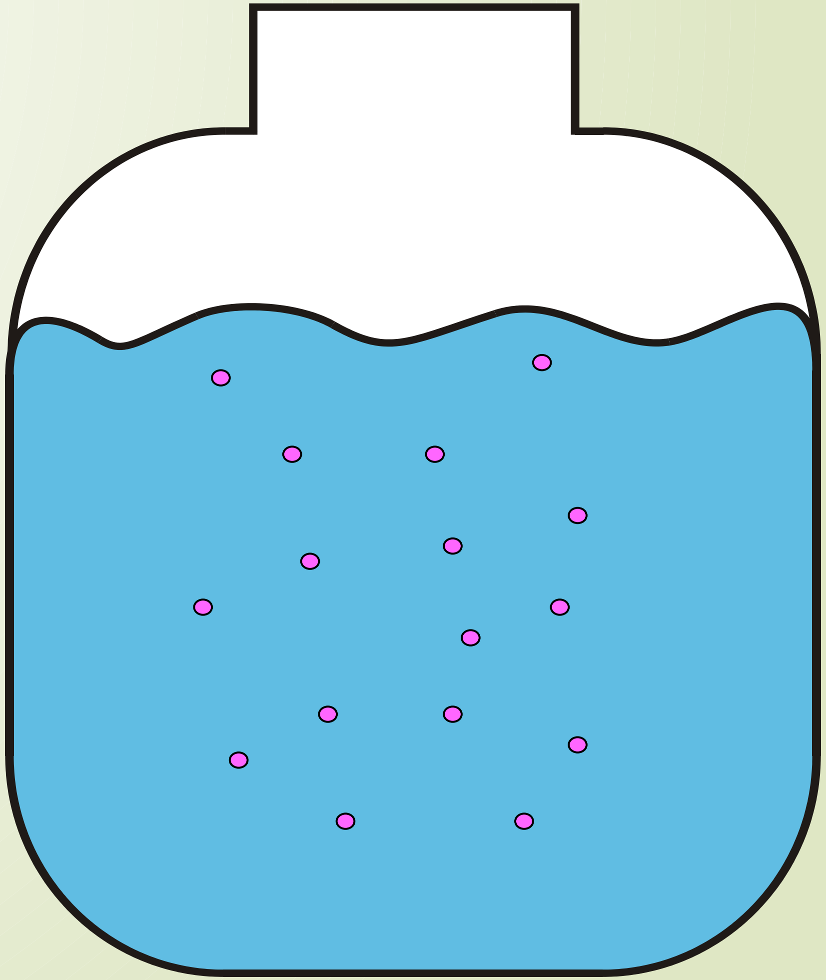
- Recoil-driven emanation:
  - O(5 MeV) α-decay creates O(100 keV) nuclear recoil
  - Recoild range 10 – 100nm, depending on material
  - Independent of temperature
- Diffusion-driven emanation
  - Strongly depends on temperature
  - Diffusion coefficient varies a lot between materials

Radon sources in a generic experiment



Radon sources in a generic experiment

- Intrinsic radium (uranium/thorium) contamination



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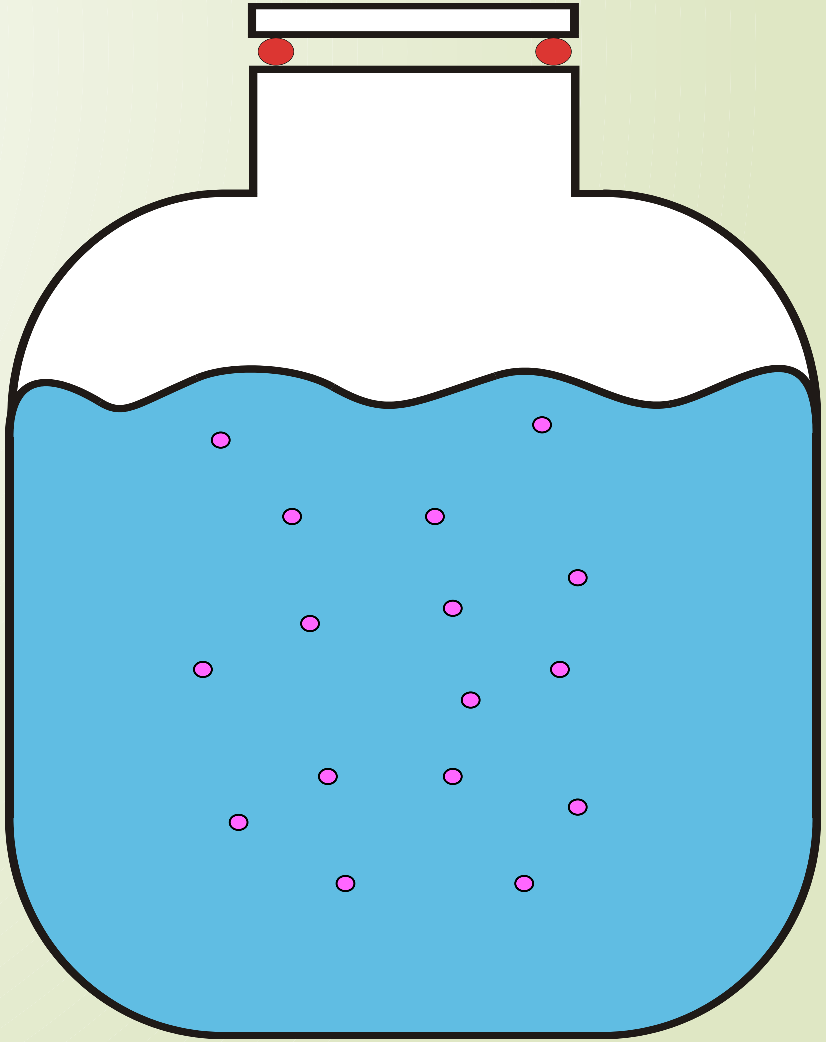
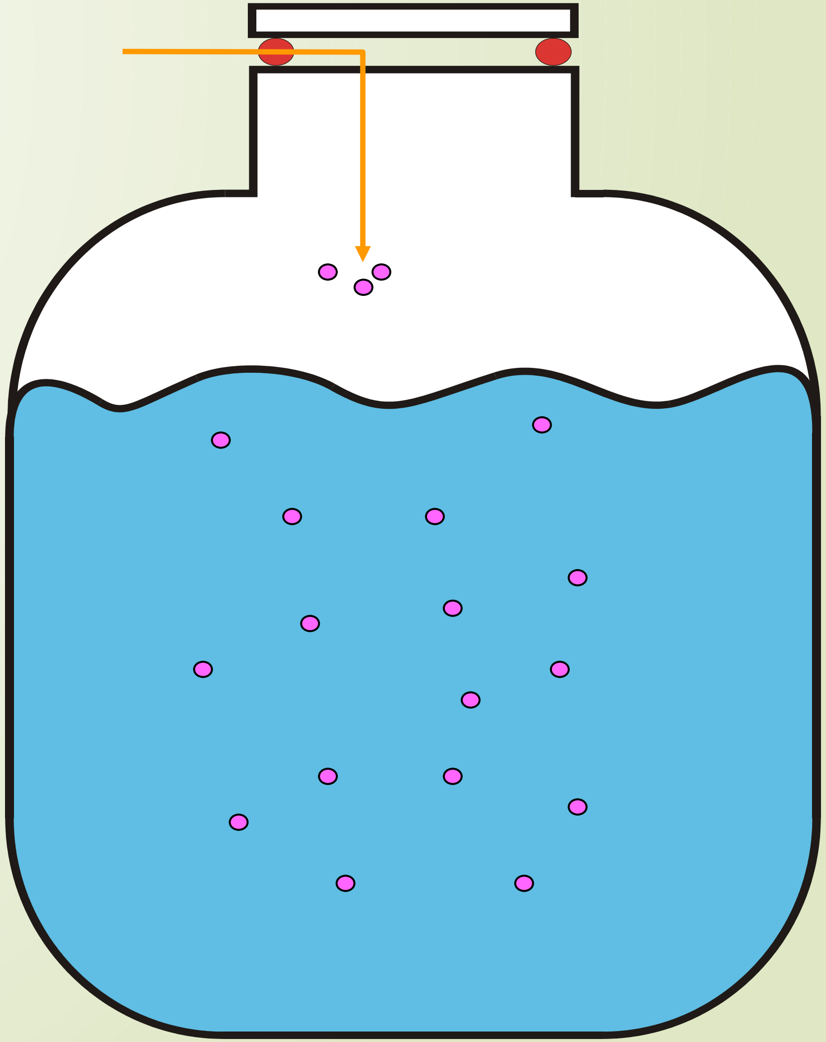


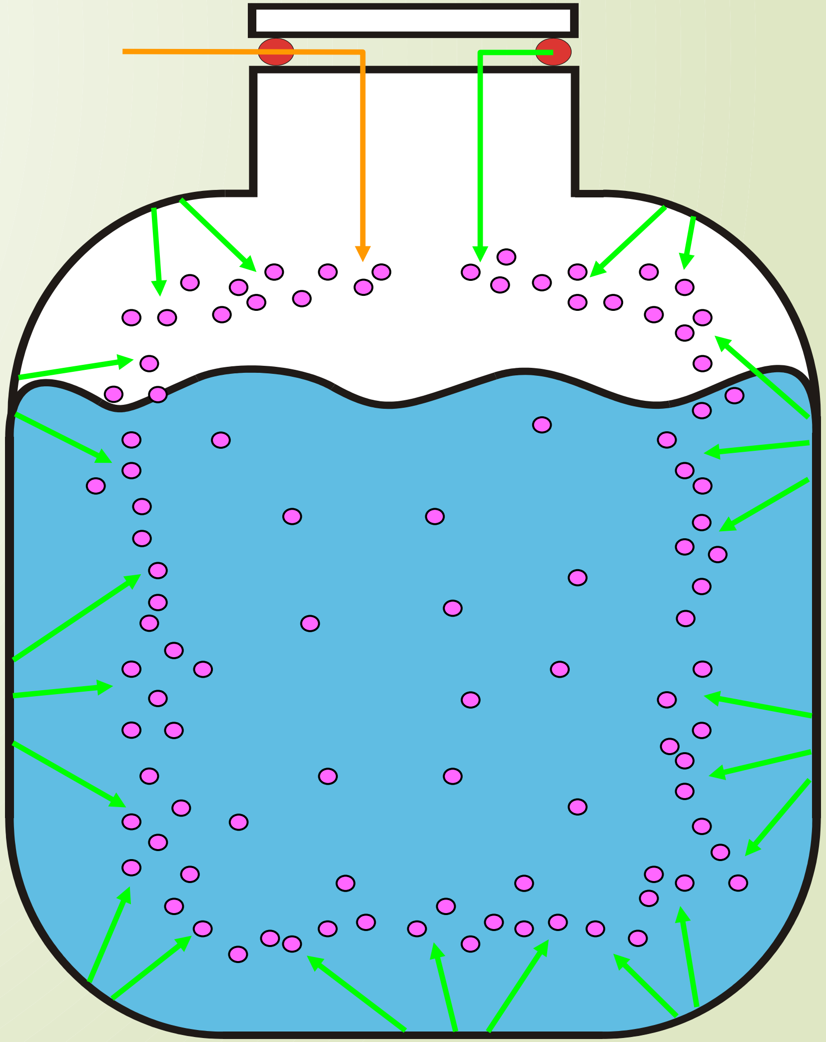
Radon sources in a generic experiment

- Intrinsic radium (uranium/thorium) contamination
- Diffusion through seals



Radon sources in a generic experiment

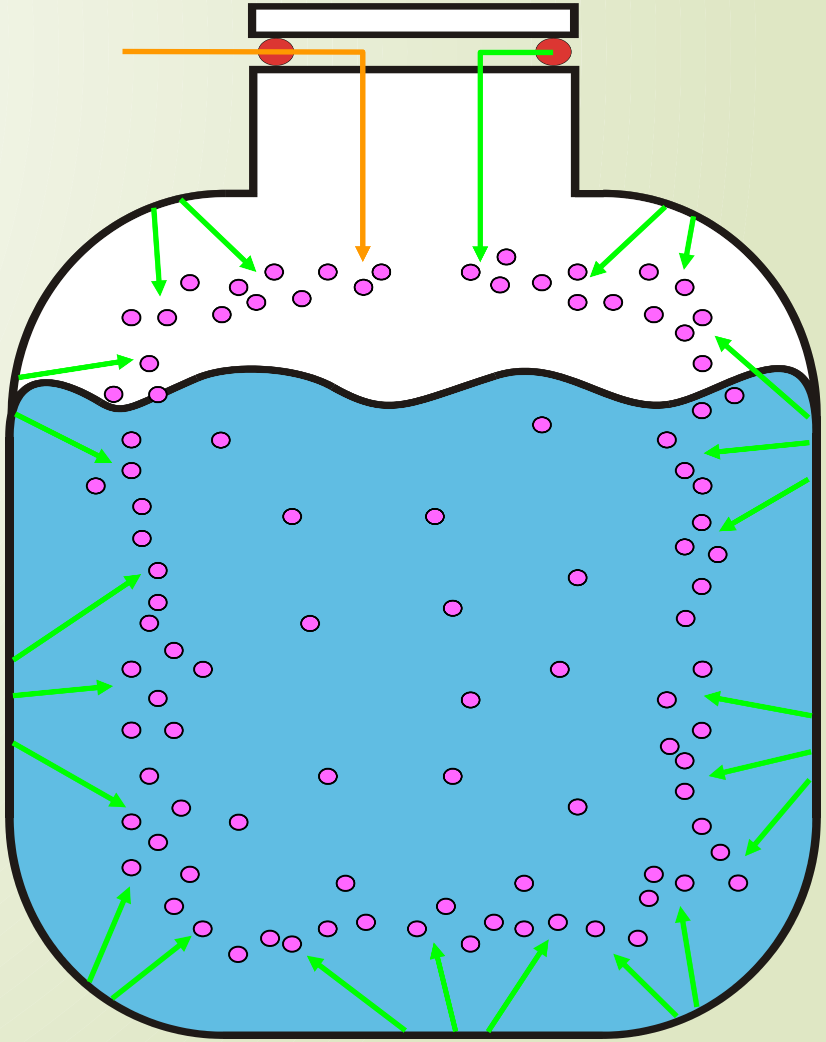
- Intrinsic radium (uranium /thorium) contamination
- Diffusion through seals
- Emanation from vessel and instrumentation



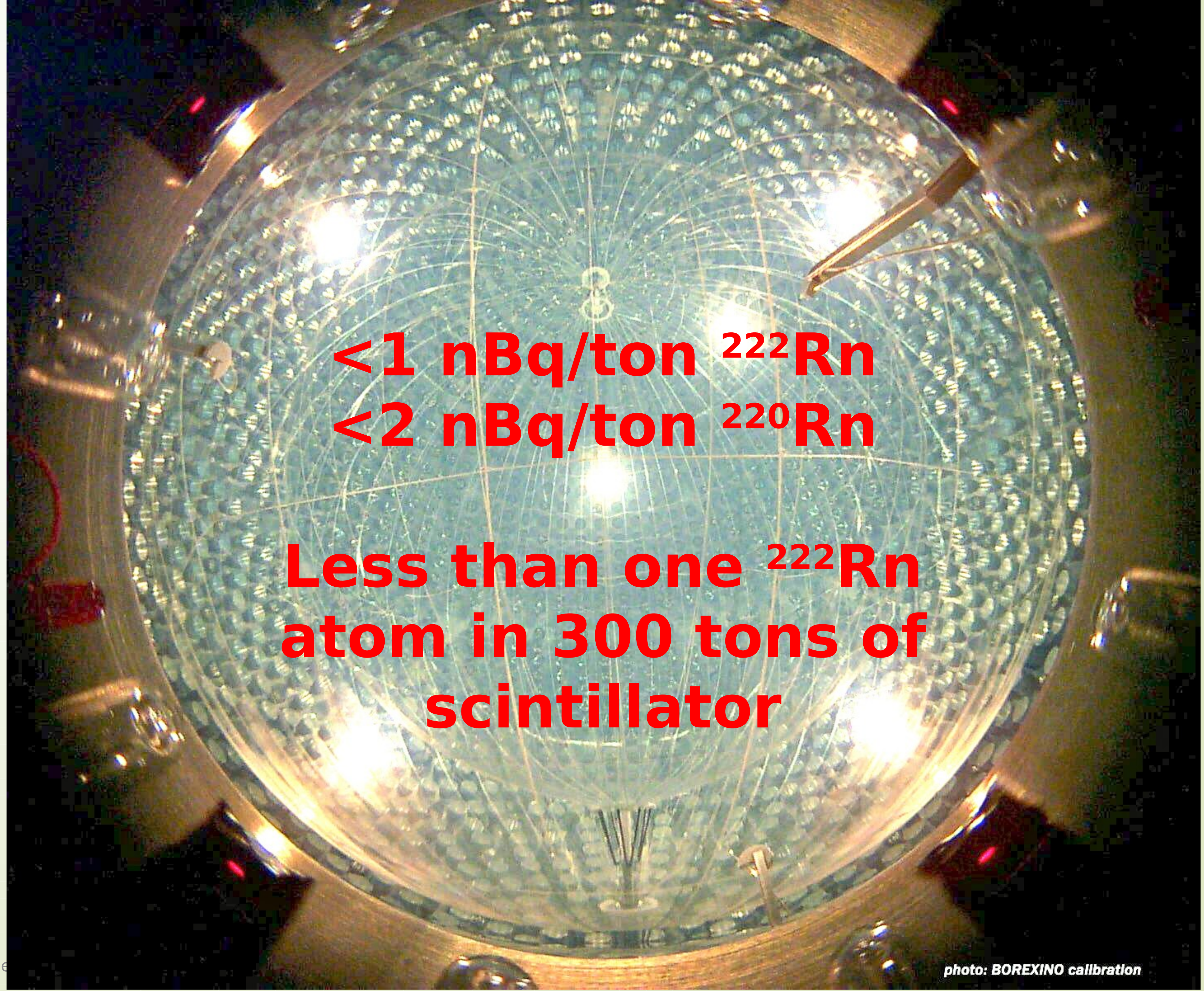
Radon sources in a generic experiment

- Intrinsic radium (uranium /thorium) contamination
- Diffusion through seals
- Emanation from vessel and instrumentation

- A successful radon mitigation strategies must address all radon sources!



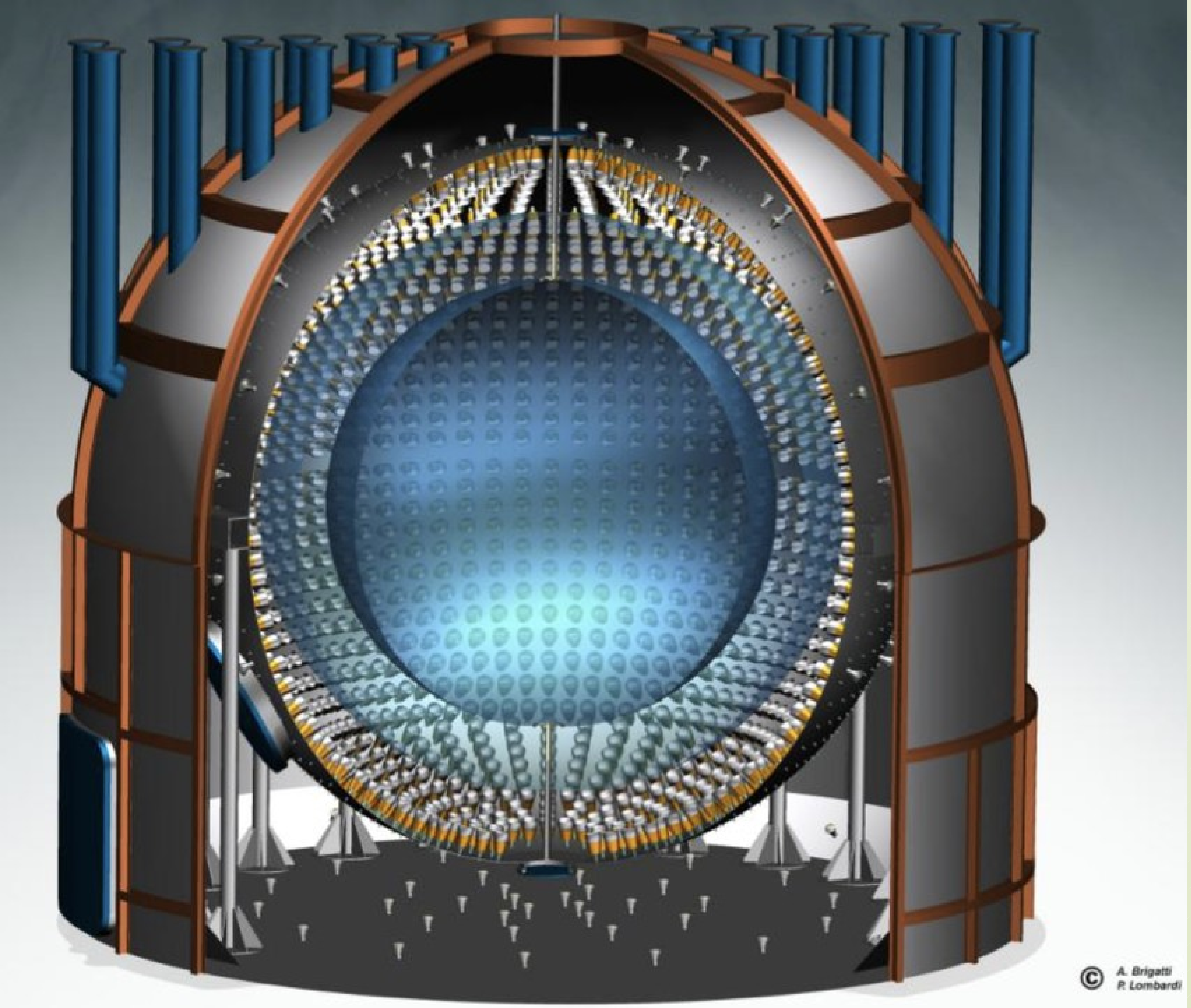
BOREXINO



<1 nBq/ton ^{222}Rn
<2 nBq/ton ^{220}Rn

Less than one ^{222}Rn atom in 300 tons of scintillator

photo: BOREXINO calibration

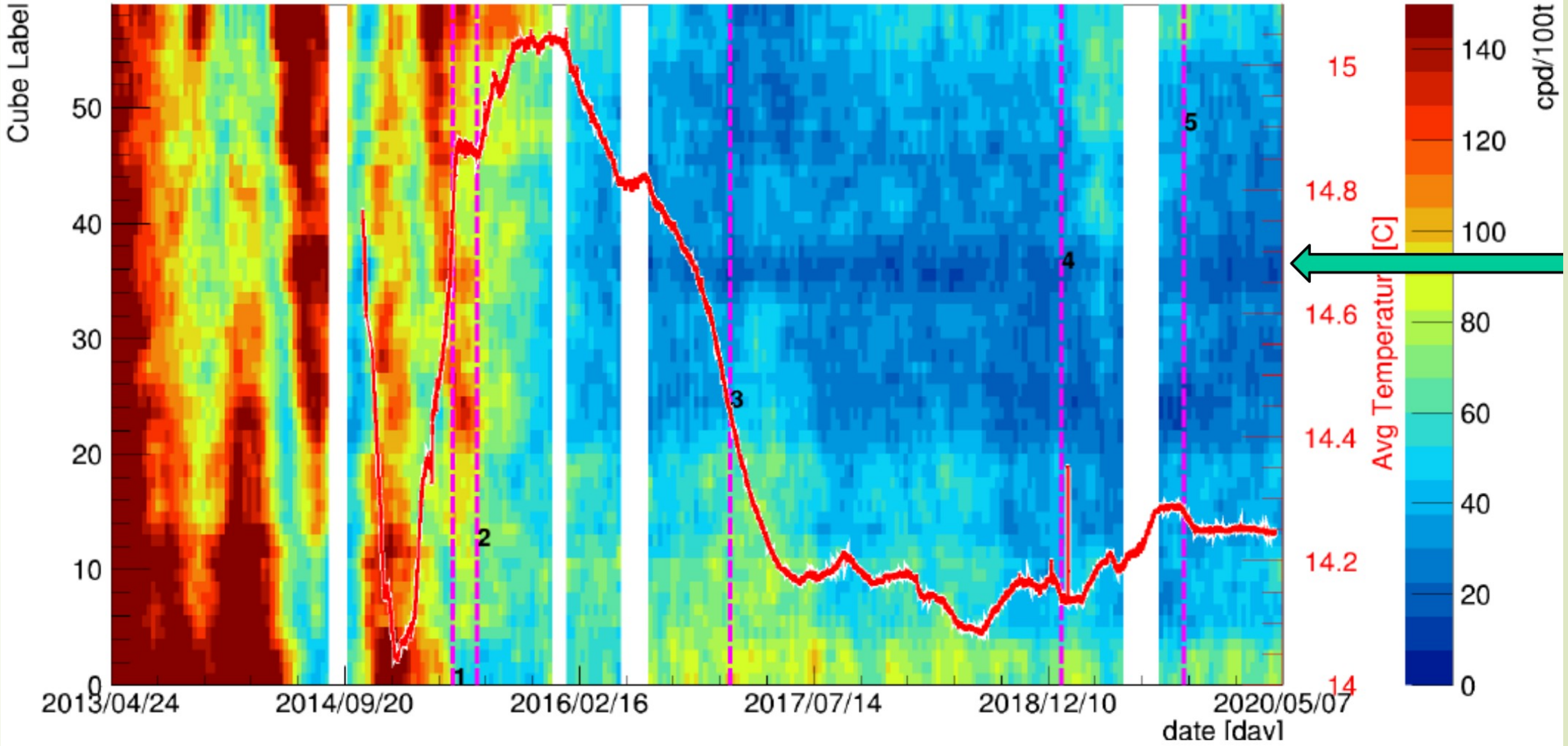


BOREXINO

- Liquid target
  - „easy“ to purify
  - Repeated purification possible
- Minimization of vessel material (nylon balloons)
- Minimization of seals
- Thorough ^{222}Rn screening campaign (complementing bulk impurity screening program)
- Thermal stability → No convection → Inner „fiducial“ volume remains clean!

BOREXINO: Thermal stabilization

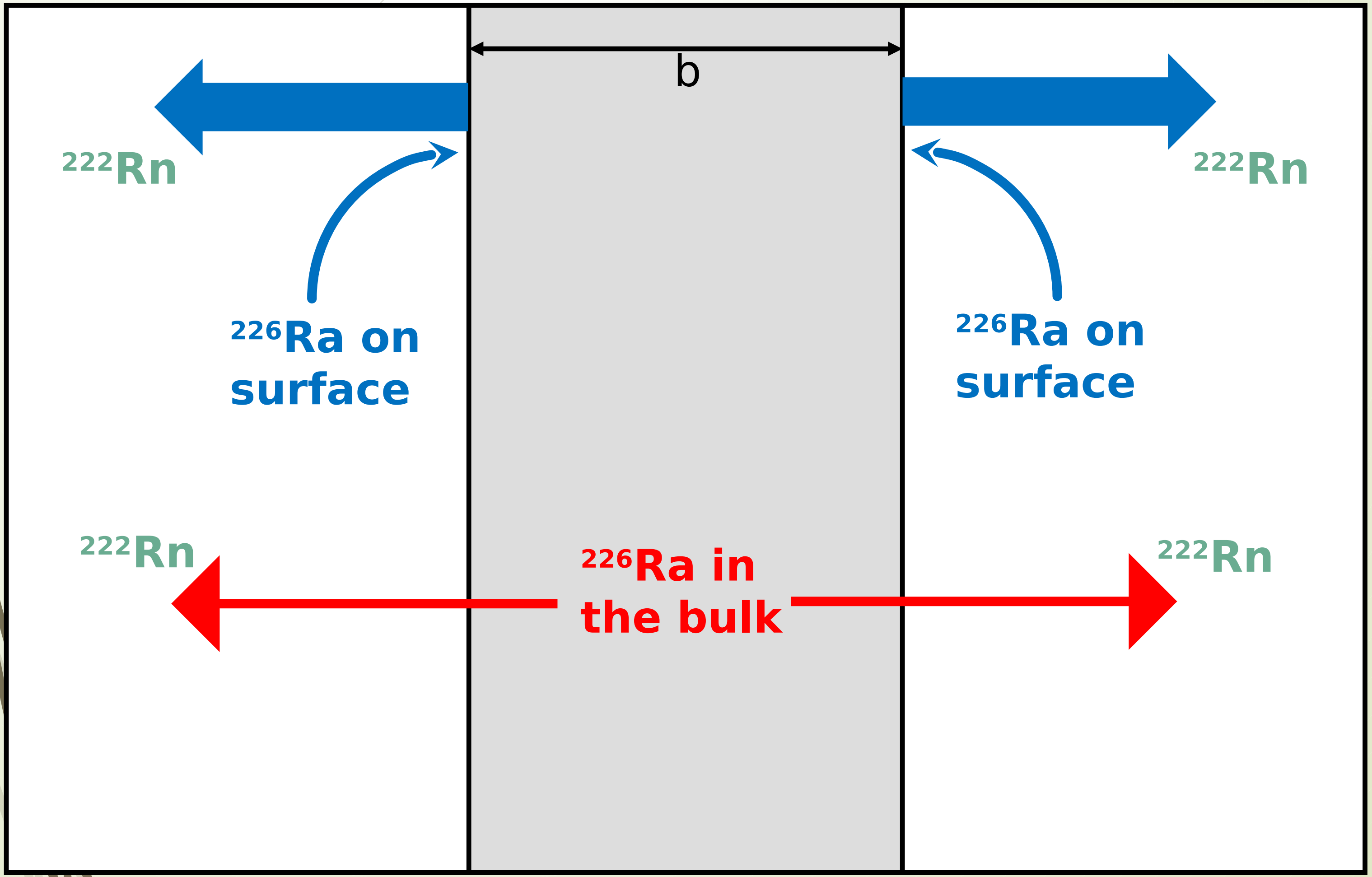
G. Ranucci, Neutrino 2020



- Switch off convection to achieve ultimate purity!

Nylon film during measurement @ MPIK

Radon in the BOREXINO nylon vessel

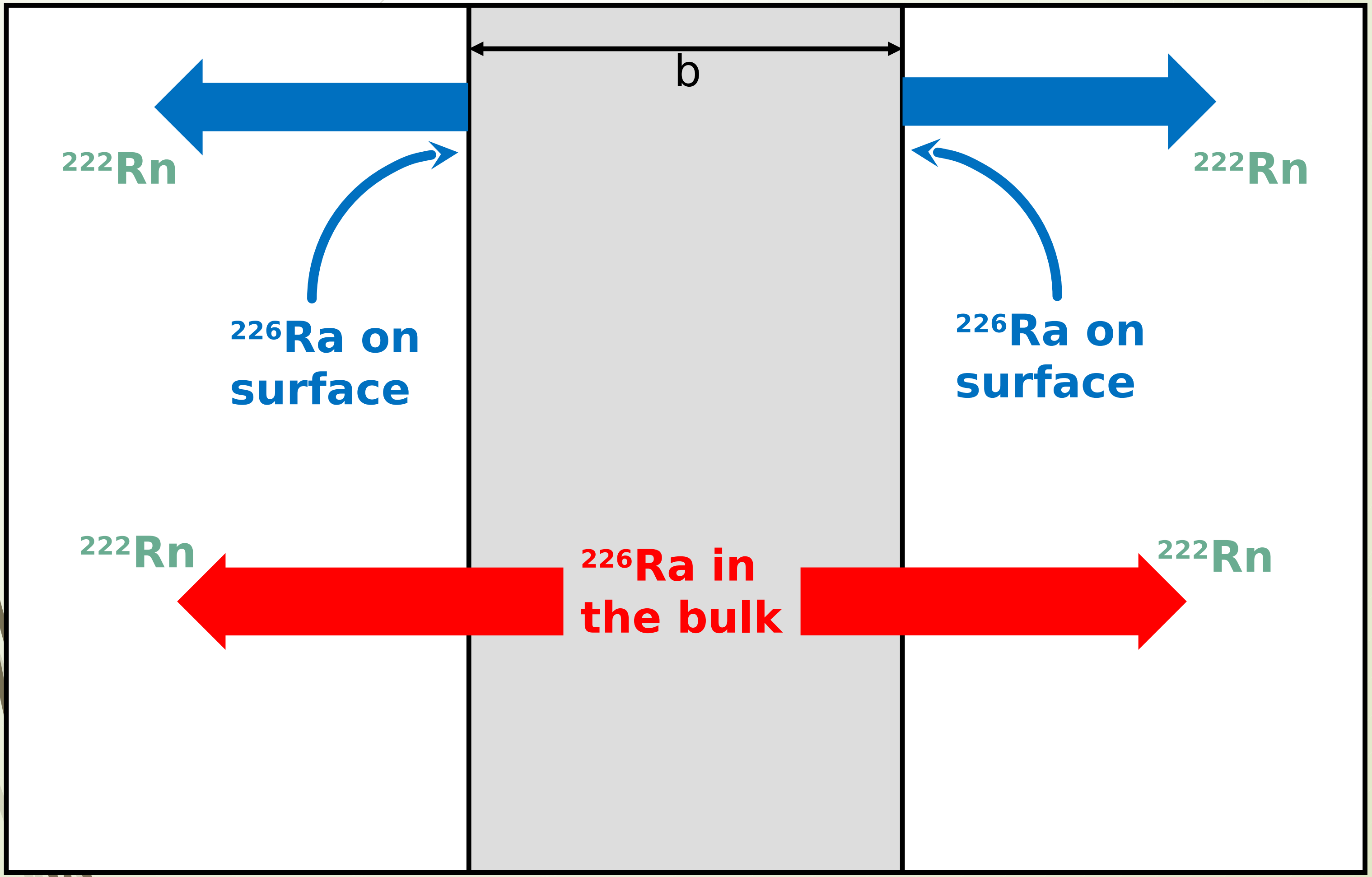


- Radon measurement from thin nylon foild yields
  - Contribution from surface

    plus

  - Unknown fraction of bulk contamination
- For ^{222}Rn mitigation it is crucial to know its origin.
- Nylon can absorb a lot of water (10% of its weight)
  - „widening“ of polymers
  - much faster radon diffusion

Radon in the BOREXINO nylon vessel



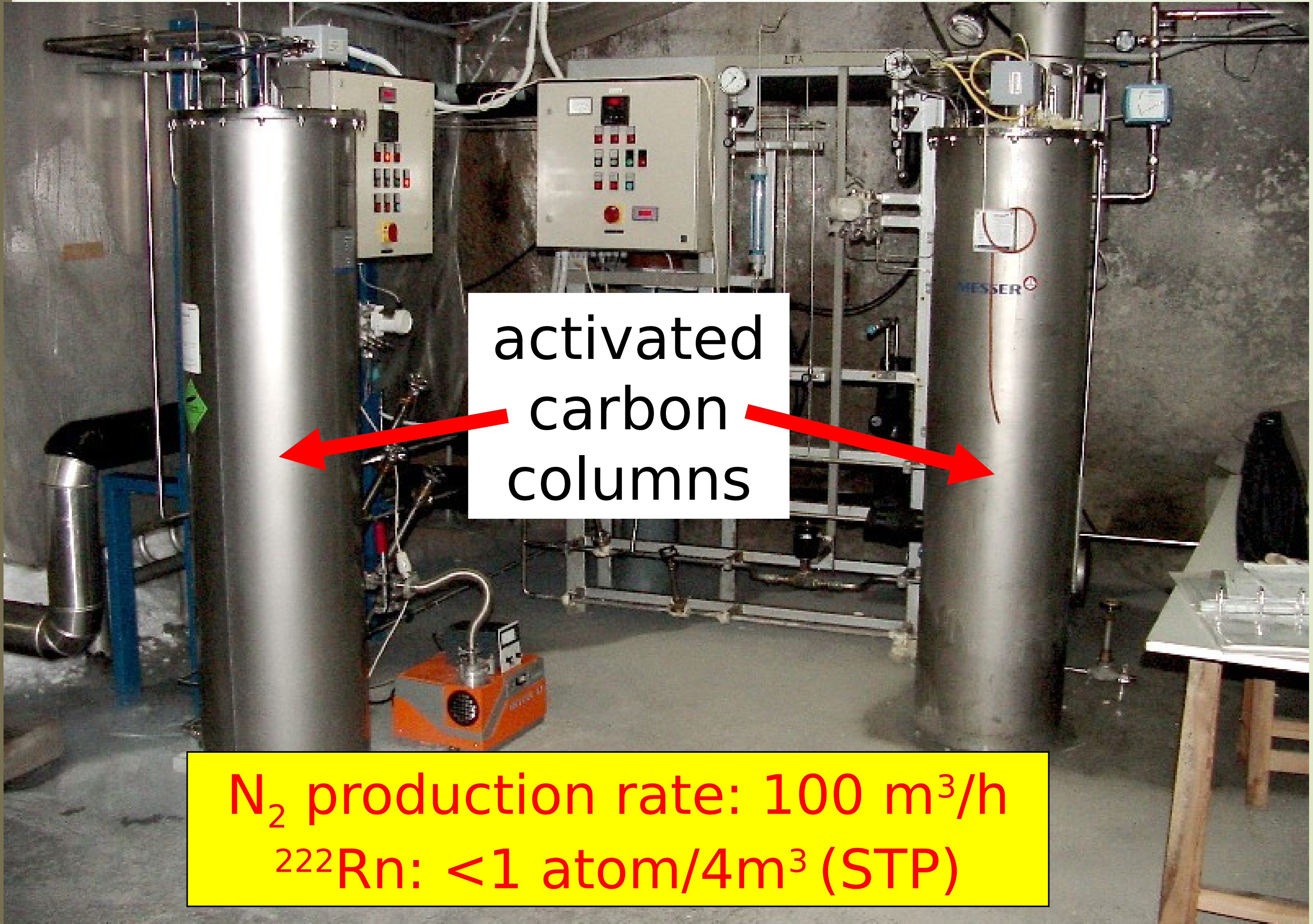
- d_{dry} = (270 ± 30) µm
- d_{humid}= (7 ± 1) µm = 40 x d_{dry}
  (NIM A 449 (2000) 158-171)

- 1-dimensional, stationary model of diffusion:
  - $A_{dry} = A_{surf} + 0.144 \times A_{bulk}$
  - $A_{humid} = A_{surf} + 0.988 \times A_{bulk}$

- Disentanglement of bulk and surface contribution

- Finally upper limits for both:
  - A_{bulk} <15 µBq/kg
  - A_{surf} <0.9 µBq/m²

High Purity nitrogen for BOREXINO



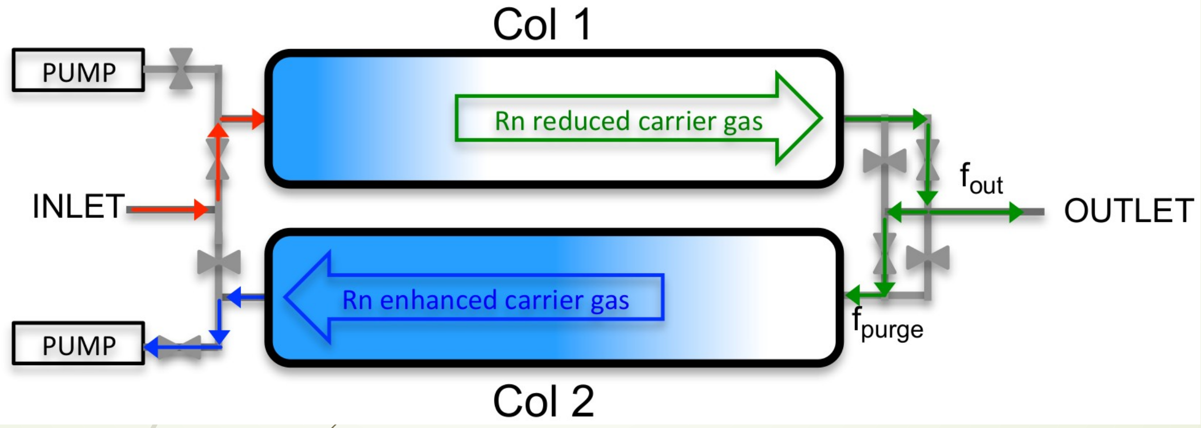
N_2 production rate: 100 m^3/h

^{222}Rn: <1 atom/4m^3 (STP)

- Radon-free nitrogen for scintillator sparging
- Large flow-rate (100 m^3/h)
- Simple concept withlittle maintenance needed
- Lqiuid nitrogen cooled columns
- Purifications of liquid nitrogen (less efficient, but high throughput)
- Key: Low ^{222}Rn emanating activated carbon

Appl. Rad. Isot. 52 (2000) 691

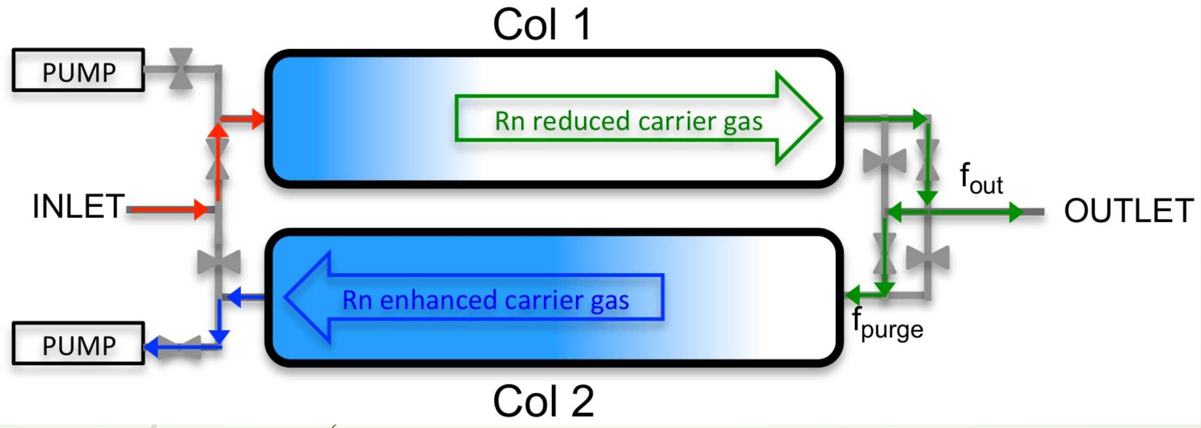
Improving adsorption gas purification



- Improving performance by doubling columns and avoiding equilibrium
- Vacuum swing adsorption technique can reduce amount of adsorbens
- Gain in radiopurity → purer gas

JINST 16 P07047 (2021)

Improving adsorption gas purification



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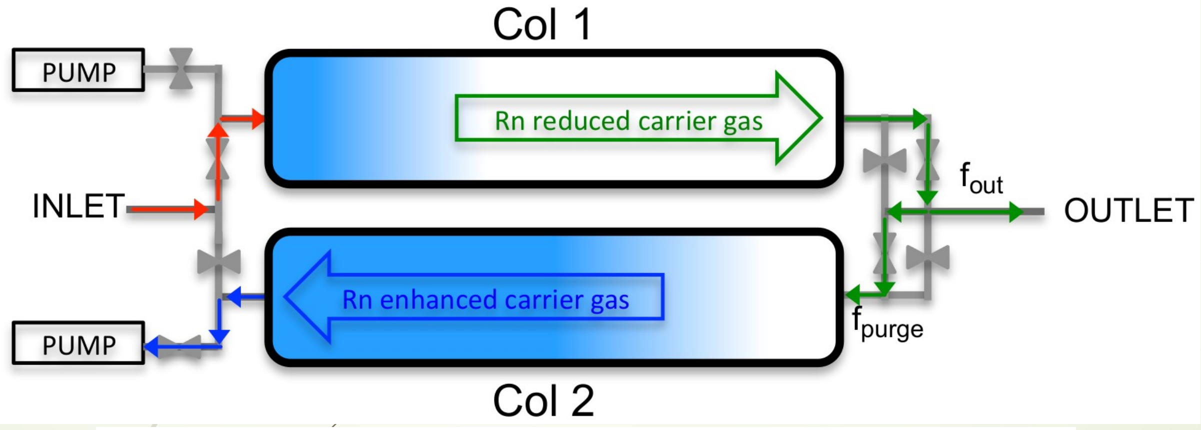
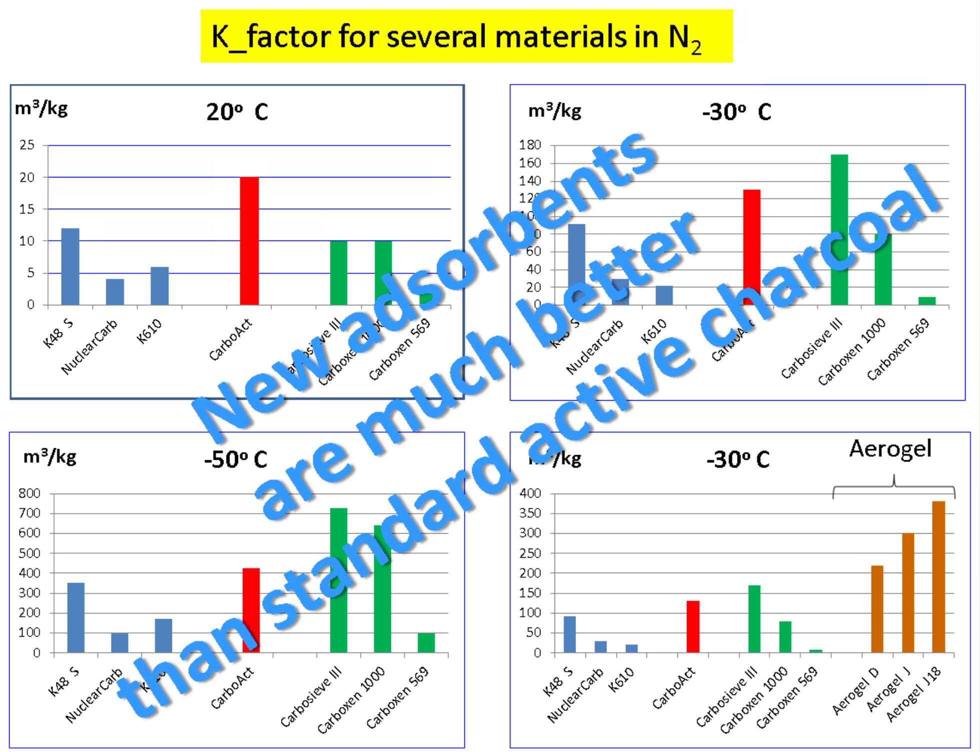
- New adsorber materials → José Busto
  - Carbon molecular sieves
  - metal organic frameworks
  - molecular cages
  - carbon aerogel
  - …

Improving adsorption gas purification




K_factor for several materials in N_2

New adsorbents are much better than standard active charcoal

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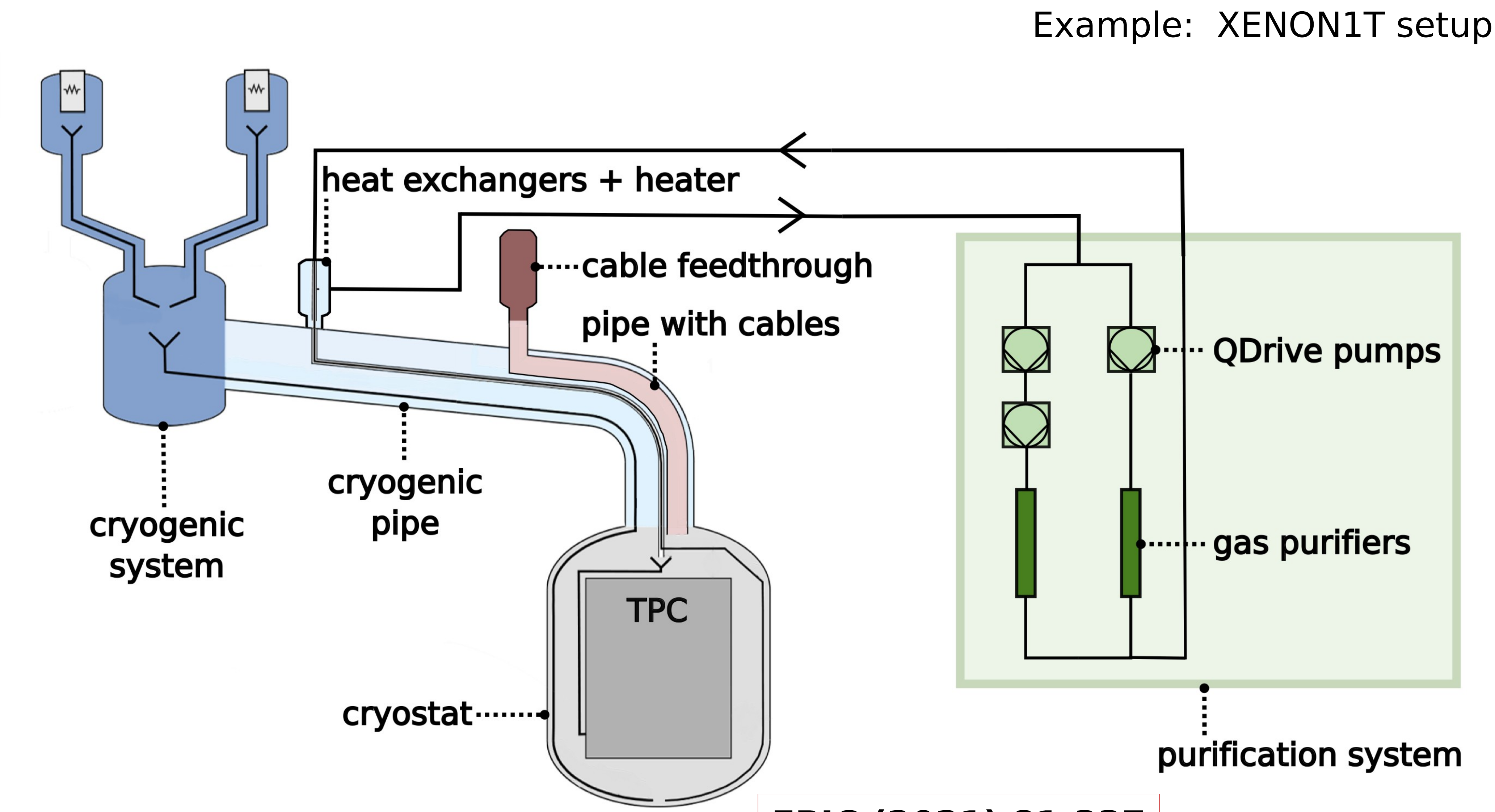
J. Busto,
GDR Nov. 22

^{222}Rn emanation rate of some stainless steel vessels

Vessel	Size	^{222}Rn emanation rate [mBq]
BX storage tank HT2	2.1 m^3	1.2 ± 0.4
BX storage tank D330	1.6 m^3	7.1 ± 1.2
GERDA cryostat	64 m^3	13.7 ± 1.9
XENON1T cryostat	~2 m^3	1.8 ± 0.3
XENONnT cryostat	~4.5 m^3	1.9 ± 0.2
BX water extraction column	0.6 m^3, 608 m^2 (SS packings)	4.8 ± 0.7 --> **8 µBq/m^2**

Understanding ^{222}Rn source

Example: XENON1T setup



EPJC (2021) 81:337

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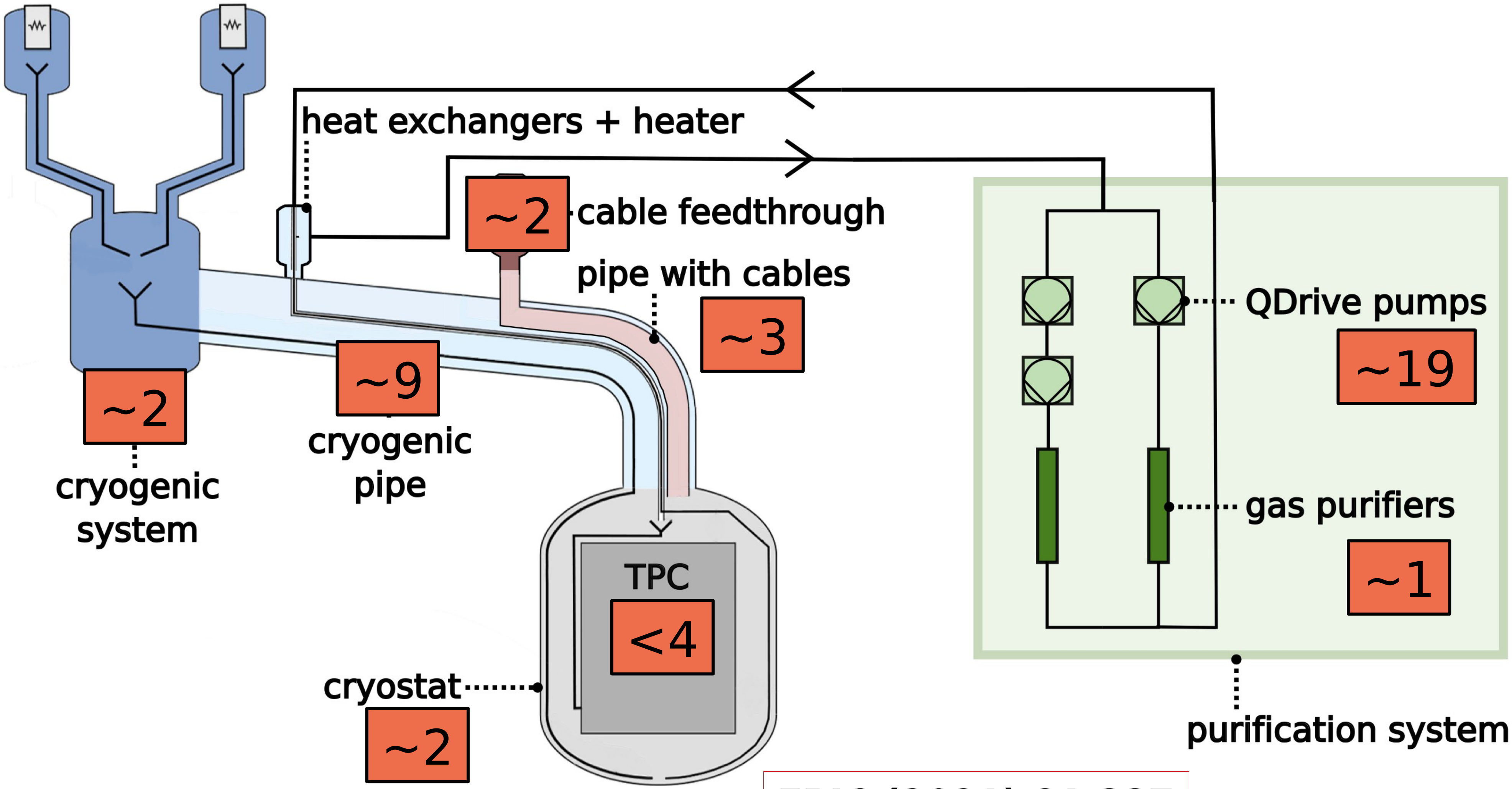
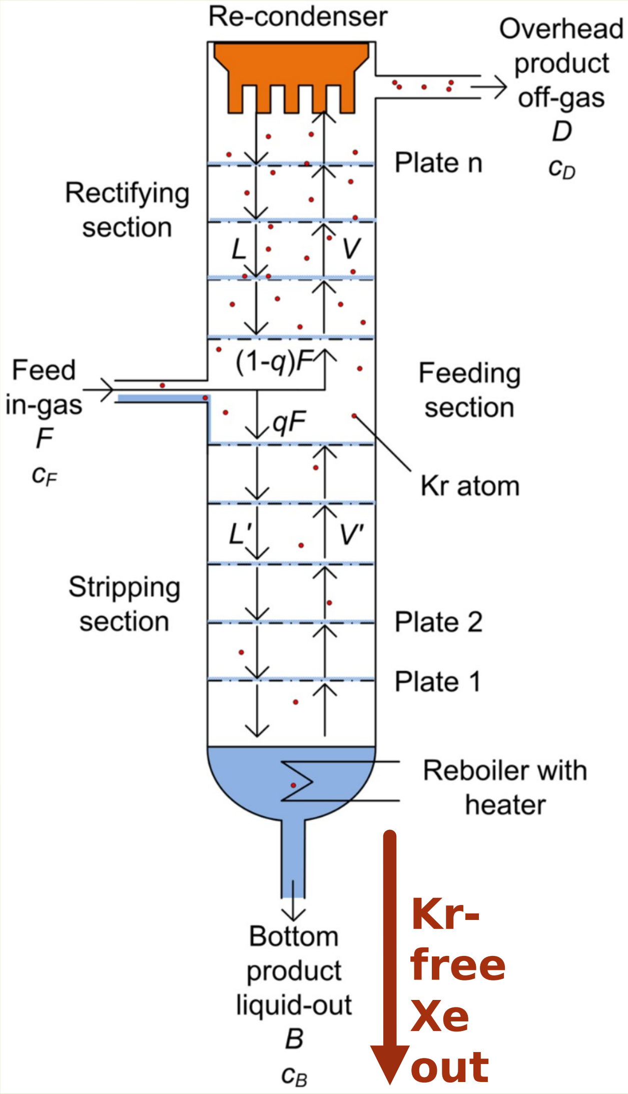


EPJC (2021) 81:337

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
- **0.8 μBq/kg achieved**
- **Future: Further upscaling?**
  - Need to process entire budget in O(^{222}Rn half-life)
  - 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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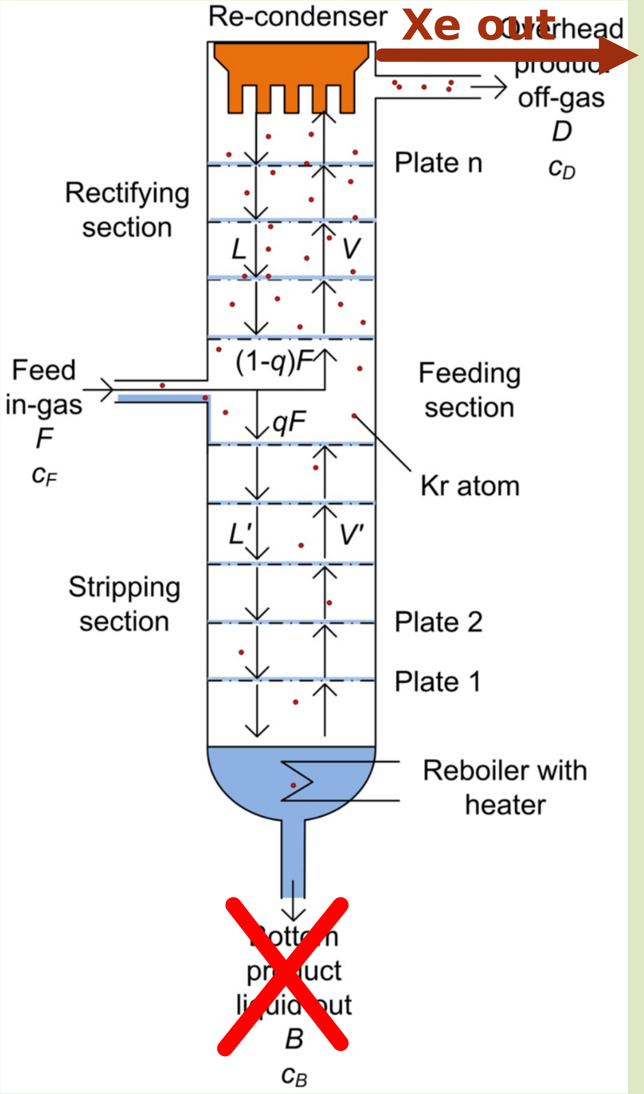
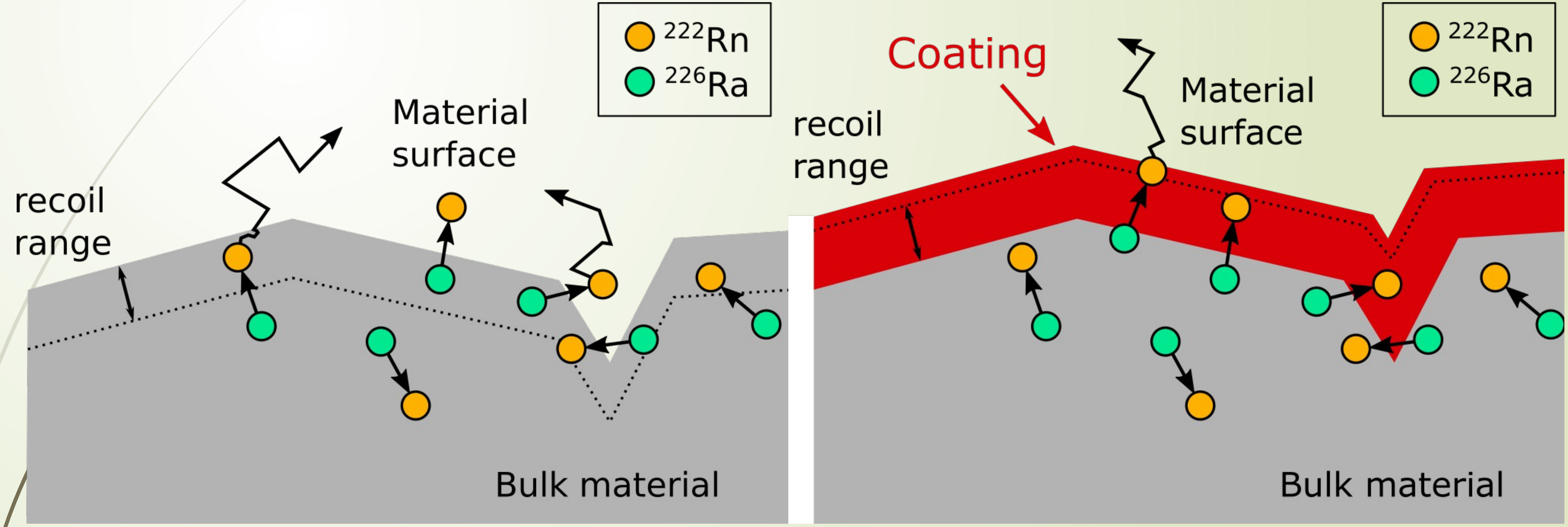


Rn-free Xe out

Radon mitigation by surface coating



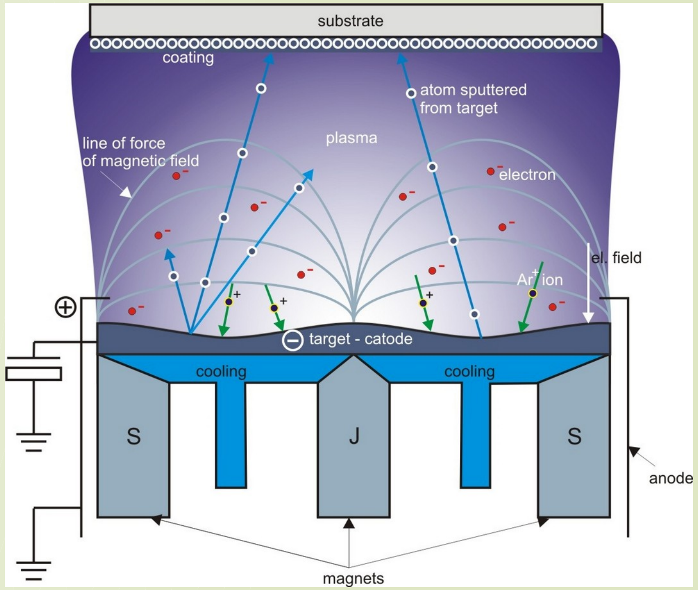
- Idea: A Rn-tight, clean (Ra-free) surface coating blocks Rn-emanation
- Should work for recoil-driven (86 keV) AND diffusion-driven emanation

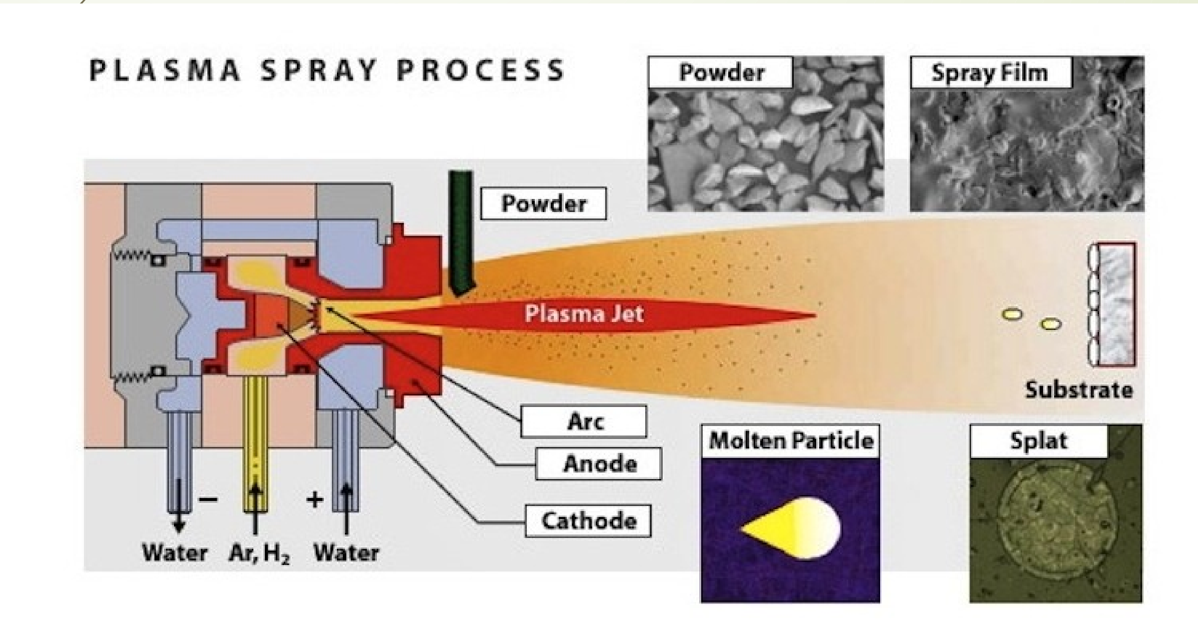
Radon mitigation by surface coating

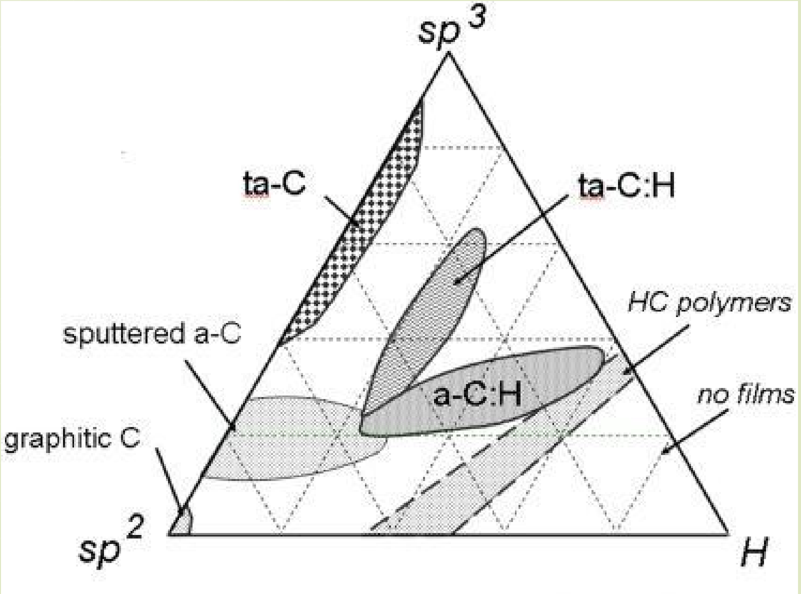
- Countless different coating techniques exist: Where to start?
- Required properties: Tight, homogeneous, stable (also at low temperature)
- Coating cleanliness may be ultimate limitations --> start with the cleanest!
  - Vacuum processes
    - Physical Vapor Deposition (PVD)
    - Chemical Vapor Deposition (CVD)
  - In-house electro-deposition
    - Experience from electro-formed Cu
- Samples: Metallic (ideally stainless steel) with high radon emanation rate
- Initially 4% thoriated tungsten welding rods
  - Wrong substrate tungsten
  - Wrong isotope ^{220}Rn: Much shorter half-life than ^{222}Rn

Investigated vacuum coating techniques

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







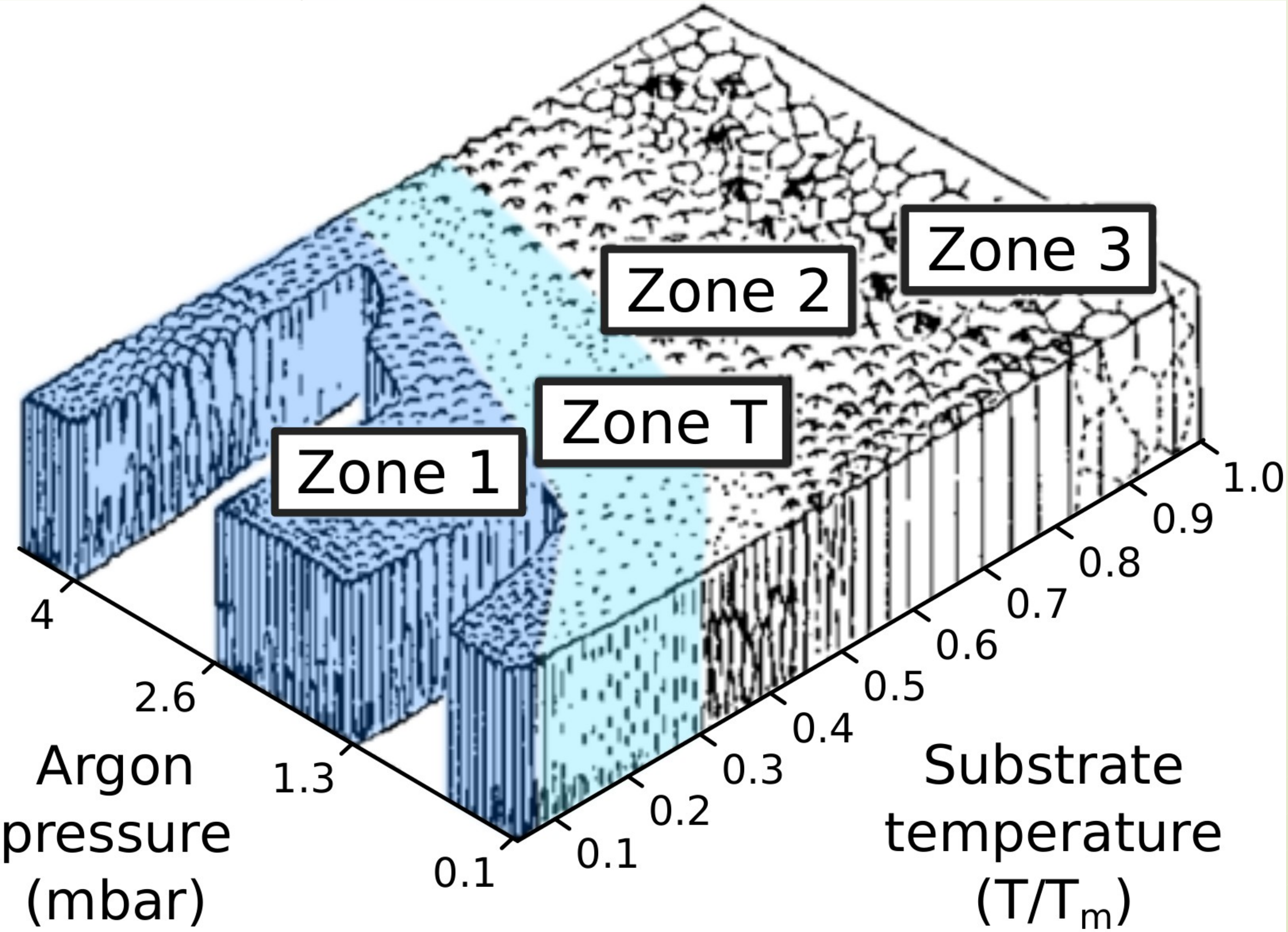
J. Robertson, Materials Science and Engineering, 2002 (129-281)

Results: ^{222}Rn emanation reduction factors

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

- Little ^{220}Rn reduction and essentially no ^{222}Rn reduction
- Best result for „hot“ plasma depsoition, but re-evaporation due to hot substrate

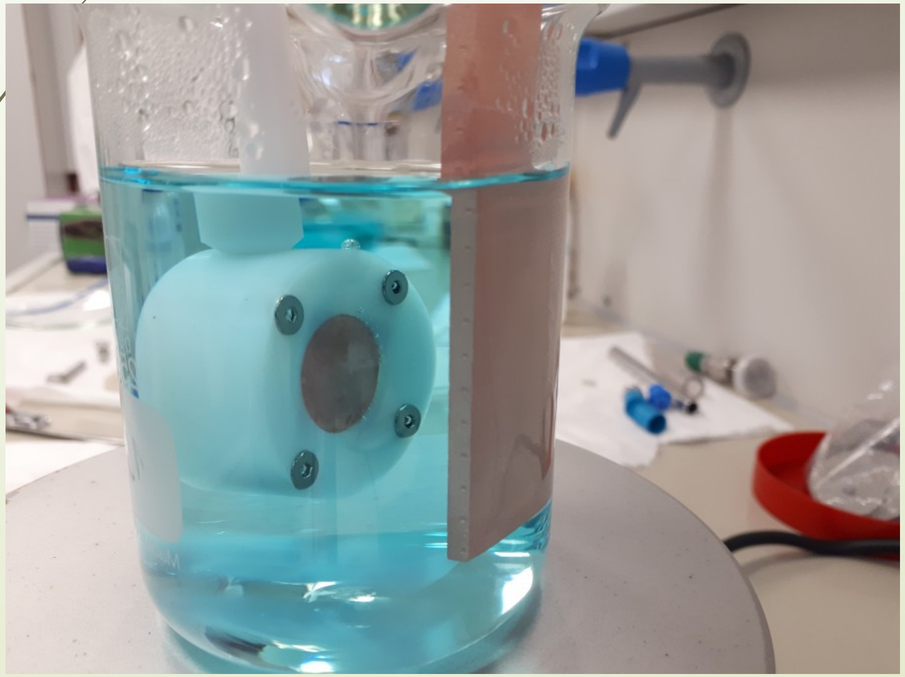
Thornton structure zone model:

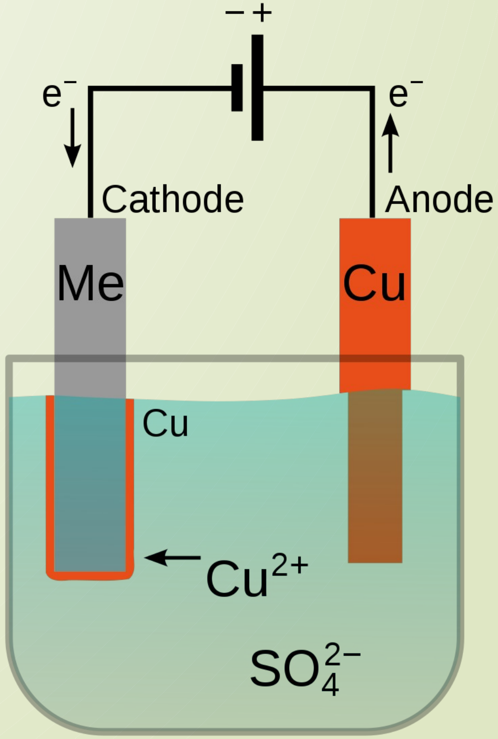


- Sputtering is low temperature process
- Growth of vertically aligned grain boundaries
- May block reactive gases (corrosion protection), but „diffusion highway“ for noble gases.
- Focus on high temperature applications (plasma coating) or non vacuum-growth technique.
- But what about CVD?

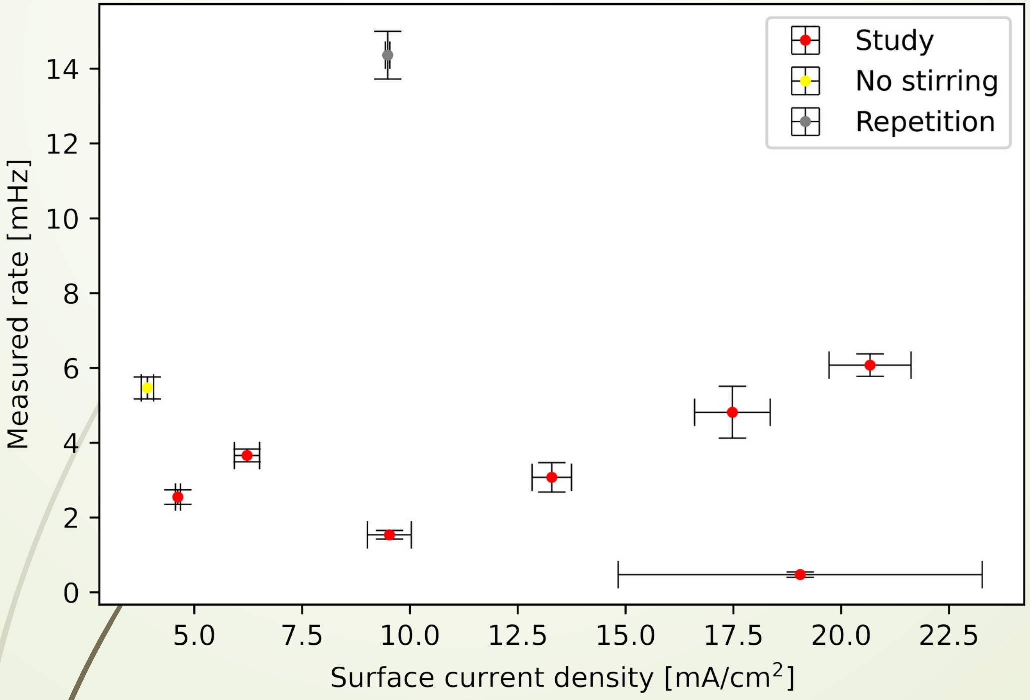
Copper electro-deposition

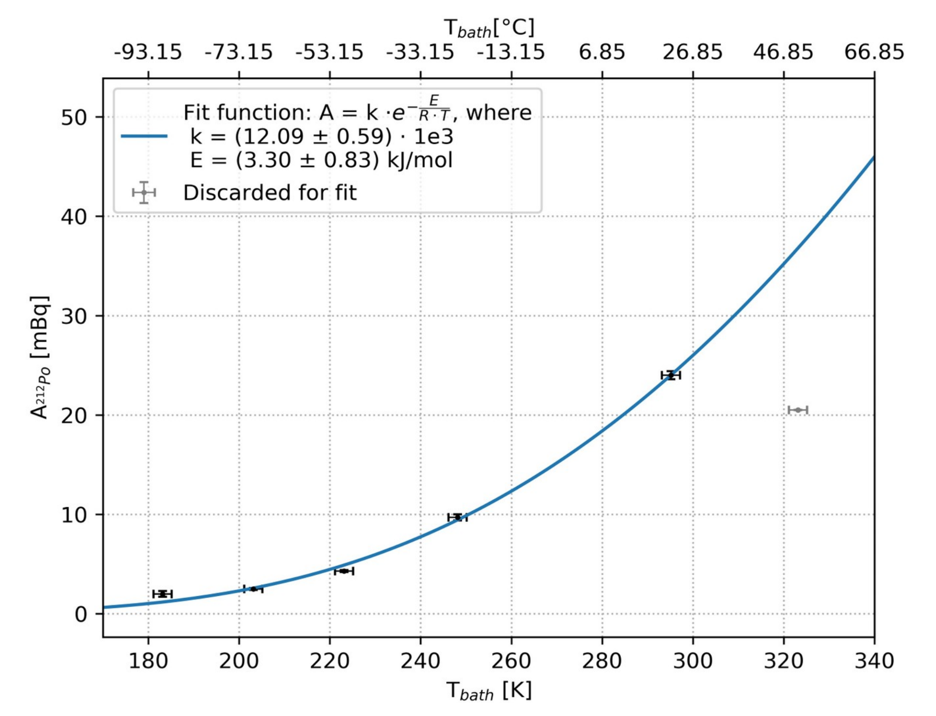
- Motivated by experience with electro-formed copper (clean!)
- In-house development at MPIK: 5 µm thick Cu layer
- Anode made from copper
- Good mechanical stability on tungsten
- Efficient blocking of ^{224}Ra alpha-decay
- ^{220}Rn reduction factor up to 100 observed!





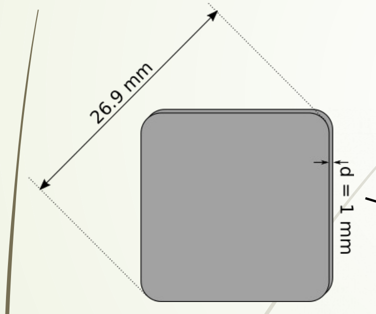
Optimization of Cu coating procedure and ^{220}Rn reduction results

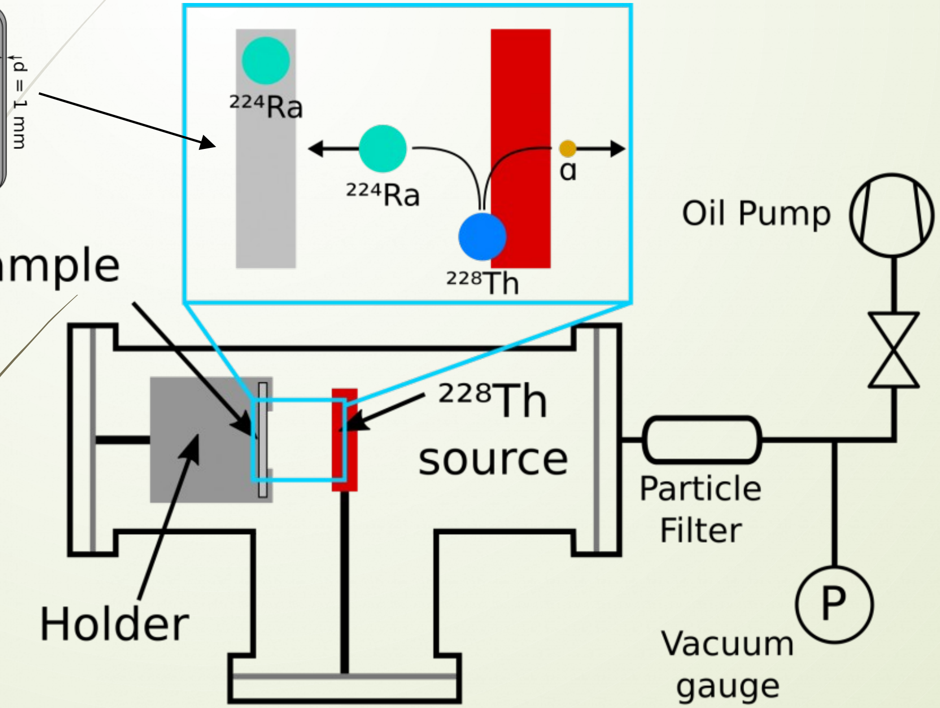


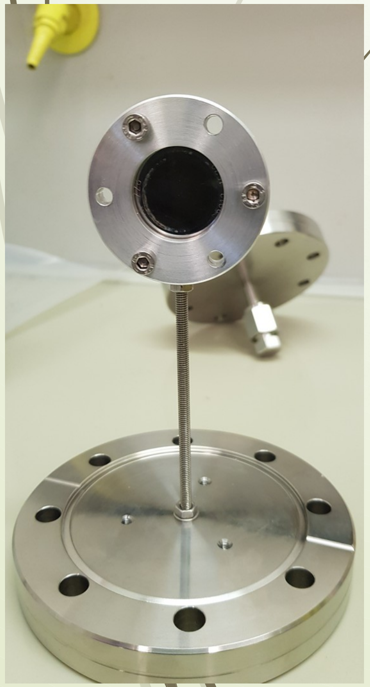


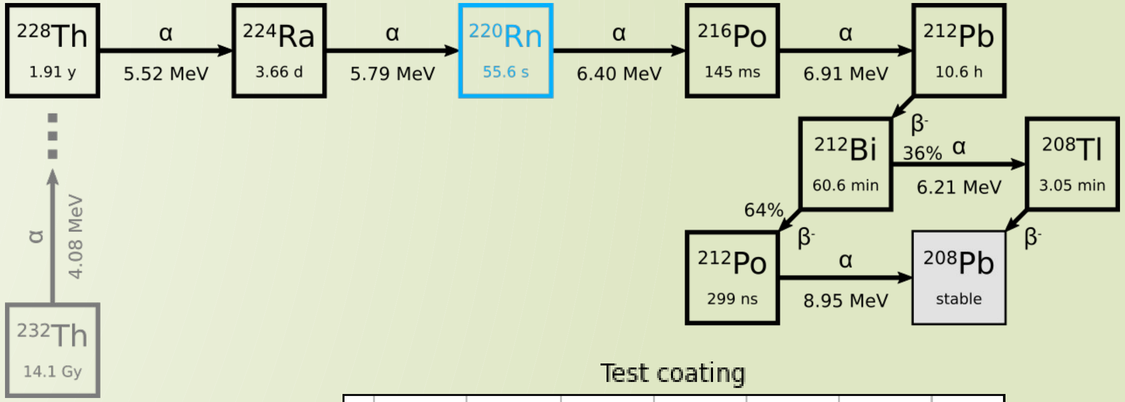
- Optimum surface current density identified.
- Avoid whisker growing by careful parameter control.
- Diffusion-driven emanation confirmed by tests at different temperatures.
- Even hints for slight ^{222}Rn reduction.

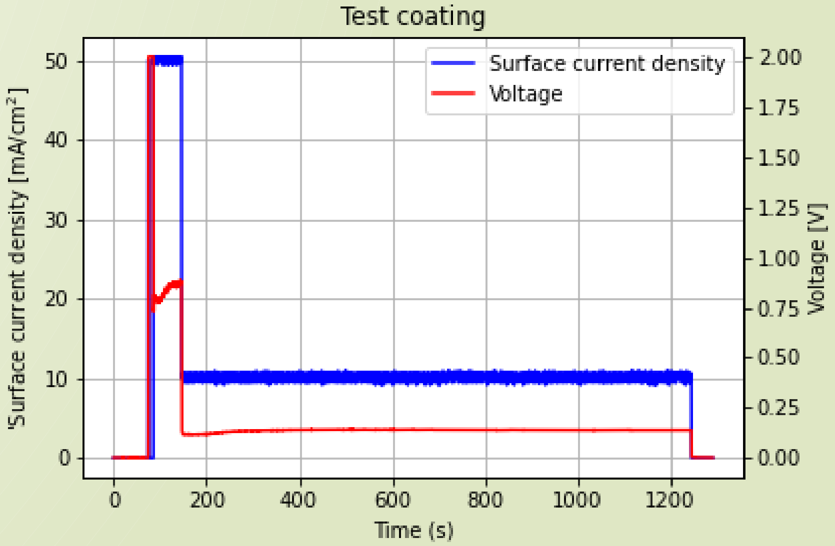
From tungsten to stainless steel: Recoil-implanted ^{224}Ra







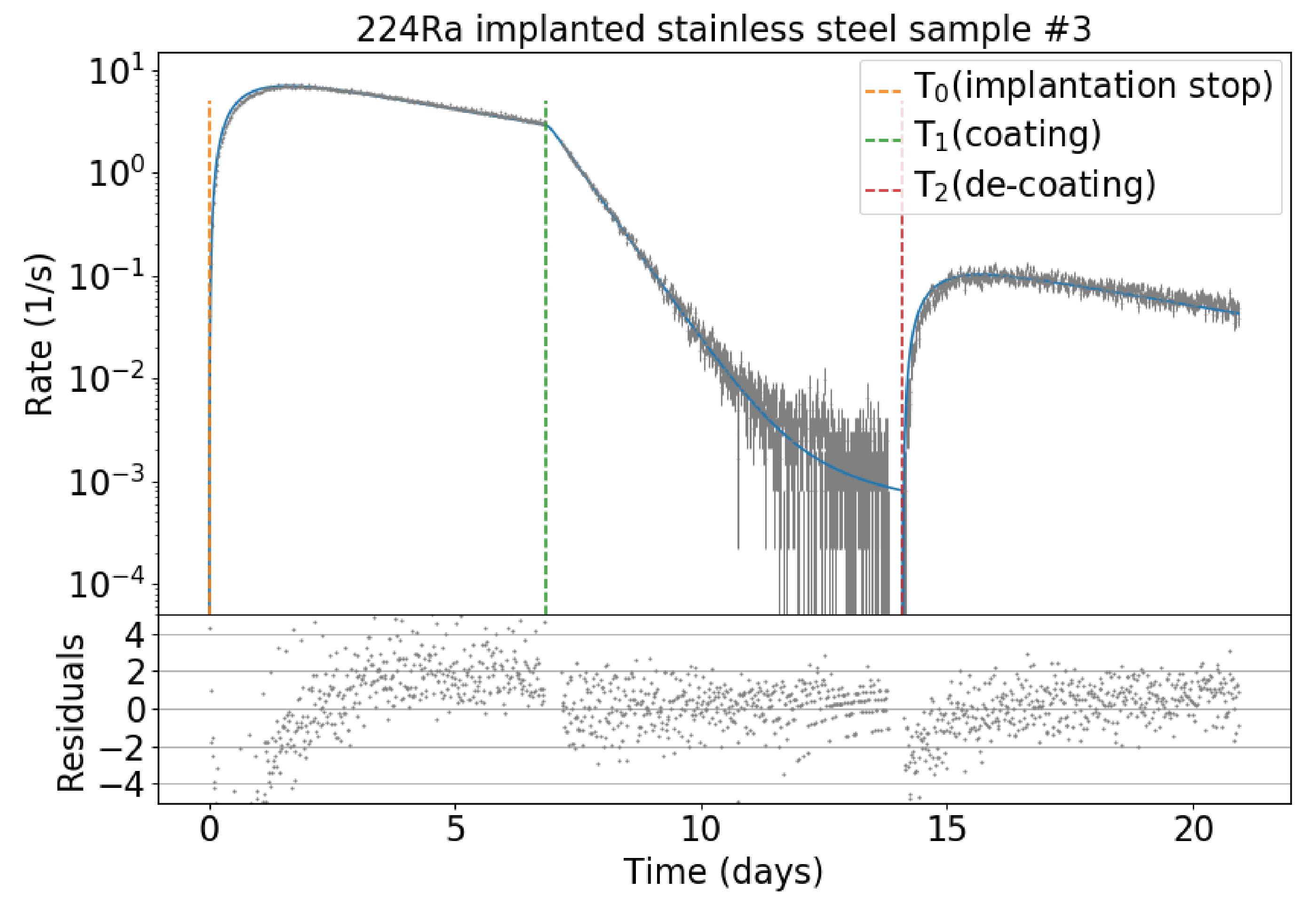


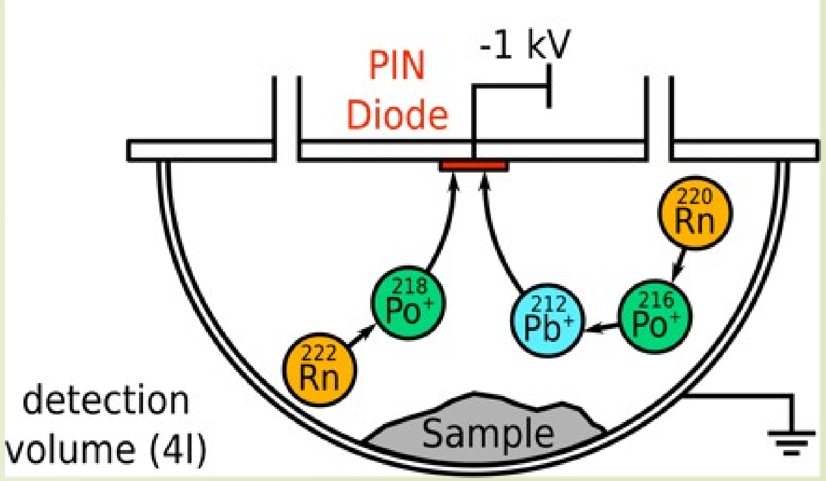


- 228Th source recoild 224Ra into SS disc
- Short-lived (t_H=3.7 days) ^{220}Rn-emanating SS disc
- Easy to produce, relatively high activity

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

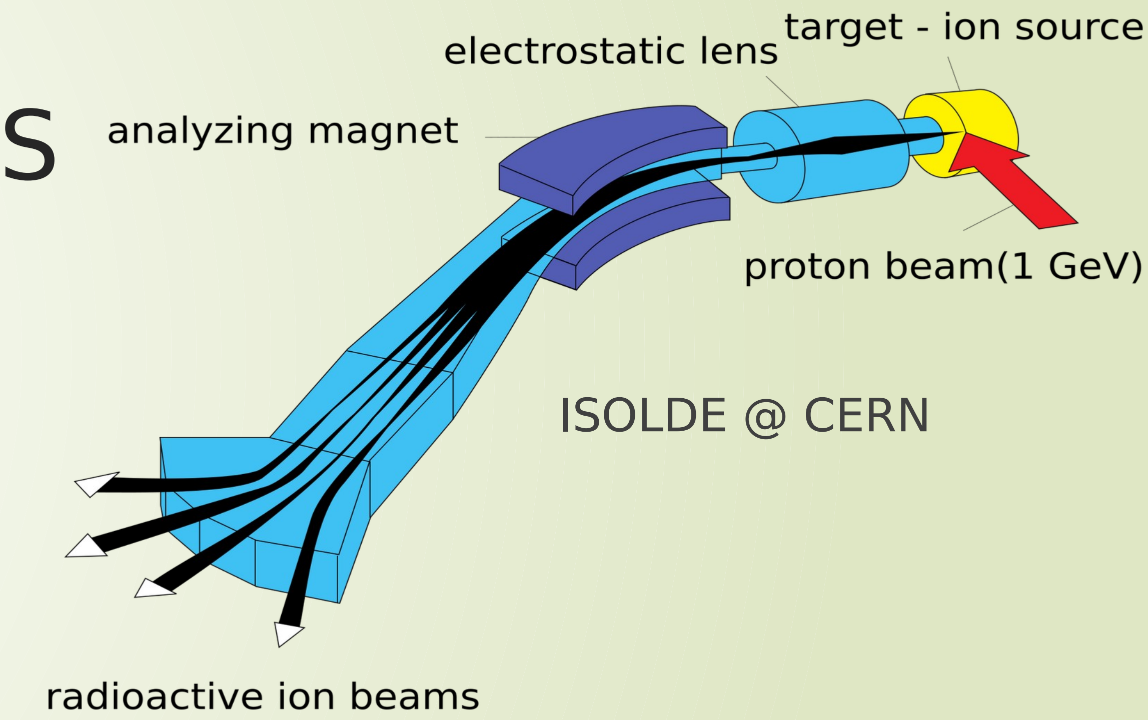
Coating (and de-coating) of ^{224}Ra-implanted SS disc



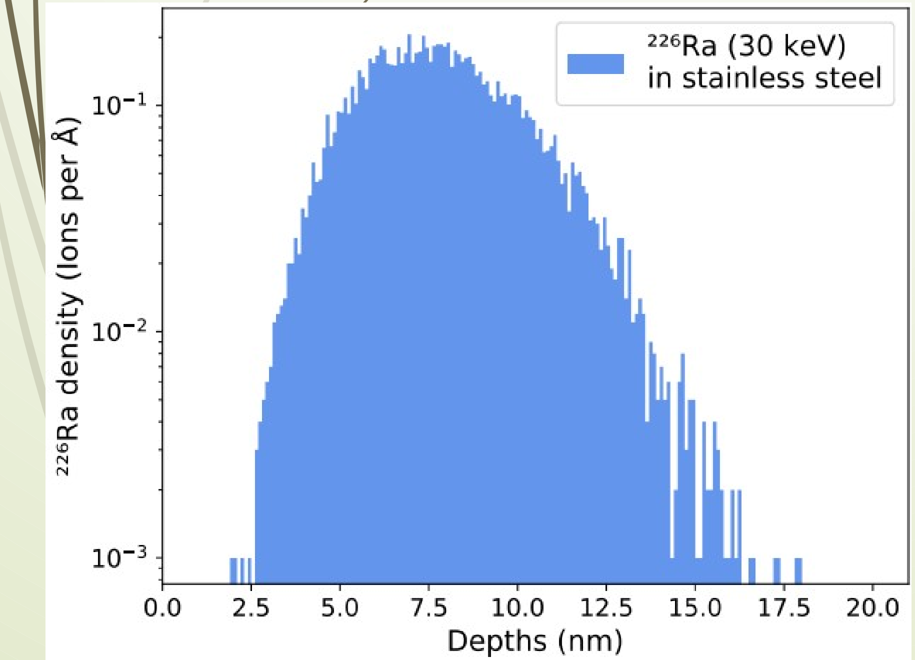


- ^{224}Ra decays on SS disc
- ^{220}Rn is emanated and decays in electrostatic radon monitor
- Charged ^{212}Pb (^{220}Rn-daughter) is collected on PIN diode
- Counting of ^{212}Po α-decays
- Possible issue: Activity wash-off in electrolyte
- Procedure: Implantation → coating → de-coating
- Upper limit from coating, lower limit from de-coating
- Results for reduction factor R: **$20 < R < 1000$**

Finally: ^{222}Rn on SS

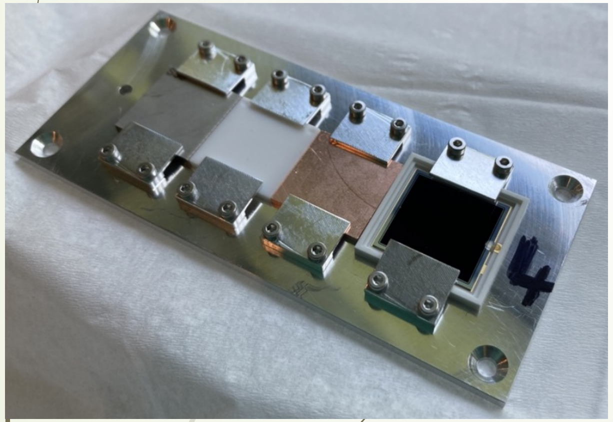


ISOLDE @ CERN

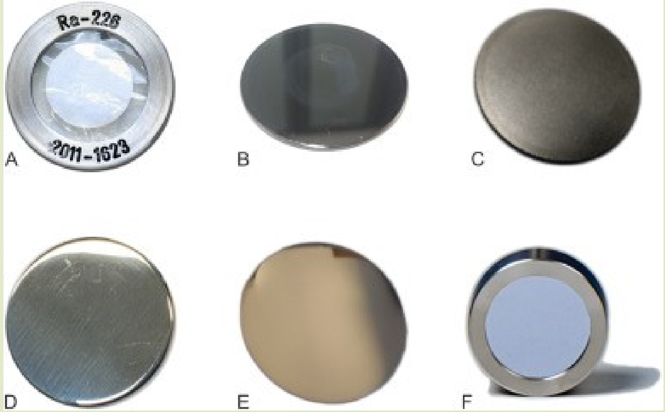


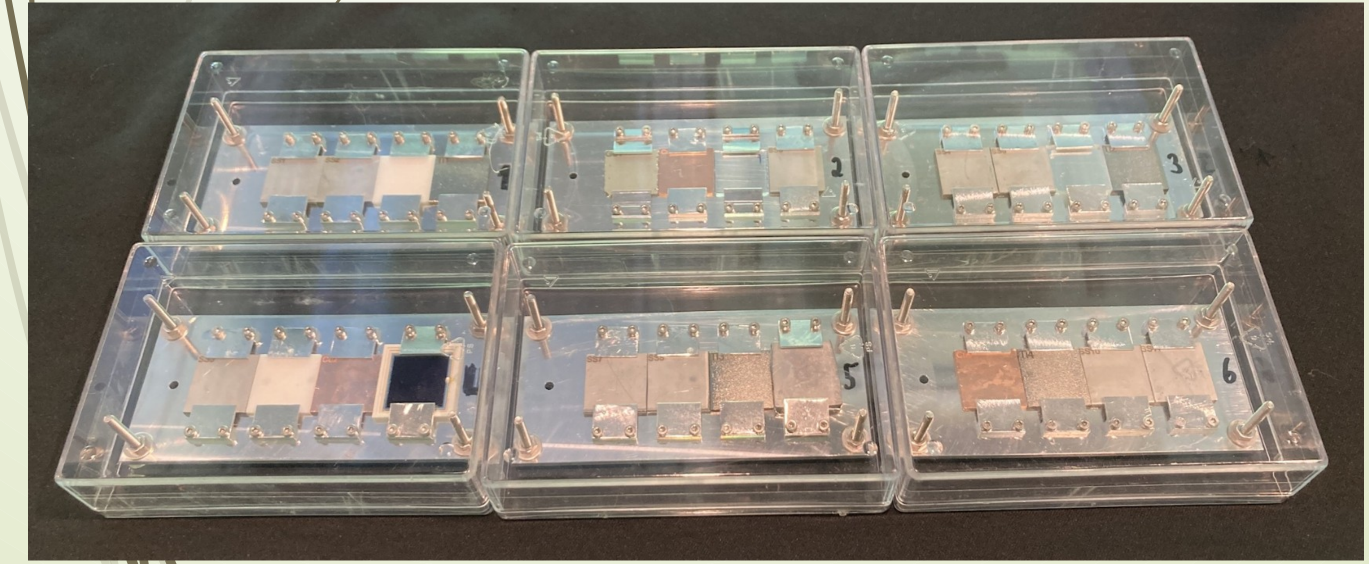
- ^{226}Ra-implanted stainless steel discs (2cm x 2cm)
- Produced at ISOLDE facility (CERN)
- 30 keV implantation energy
- Ion range distribution simulation in SS (SRIM):
  - $\mu = 7.9$nm, $\sigma = 2.3$nm
- 2 test samples produced in 2017
- Summer beam time for 20 new samples approved

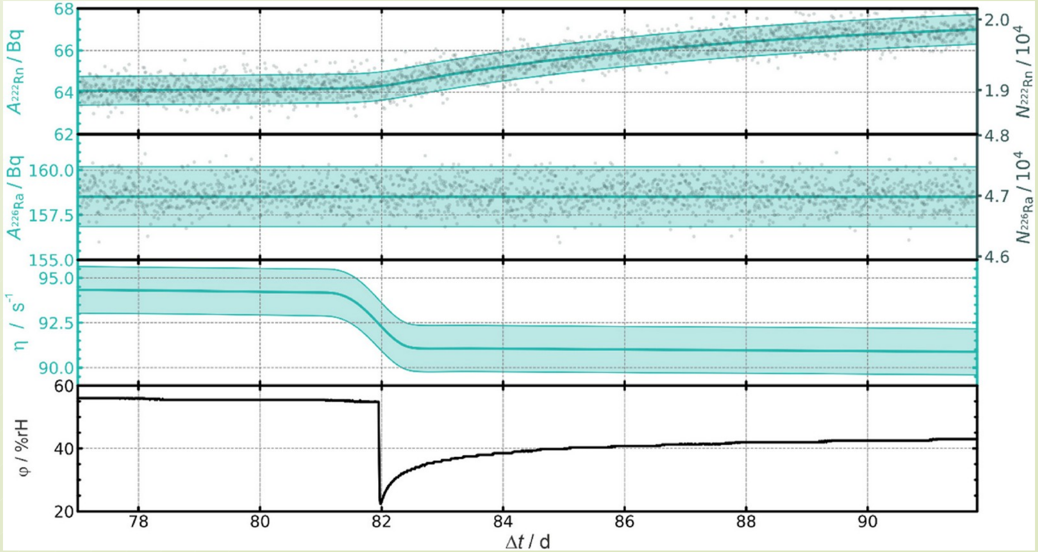
Novel ^{222}Rn sources



- ^{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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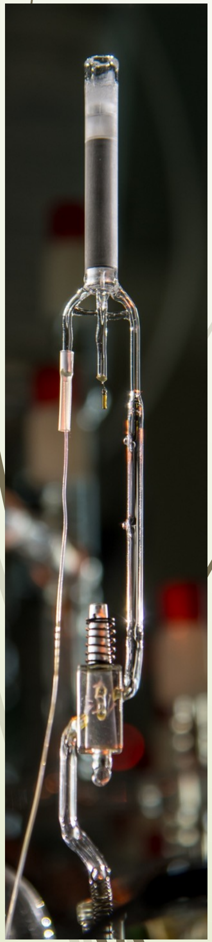
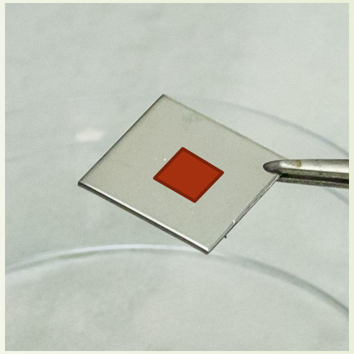
ISOLDE sample characterization





- Alpha measurement:
  - Short-lived contaminants (from ^{235}U chain)
  - ~8.5 Bq ^{226}Ra activity
  - Central deposition confirmed
- Gamma measurement confirms alpha measurement
  - Unexpected ^{139}Ce discovered (t_H = 137.6 d)
- 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

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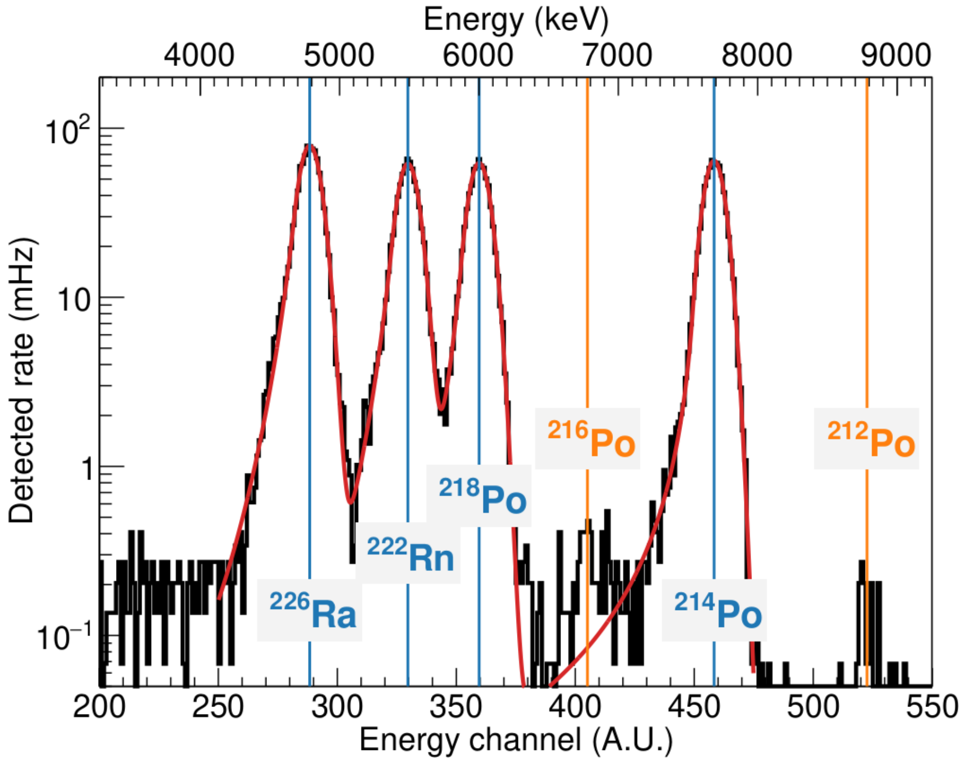
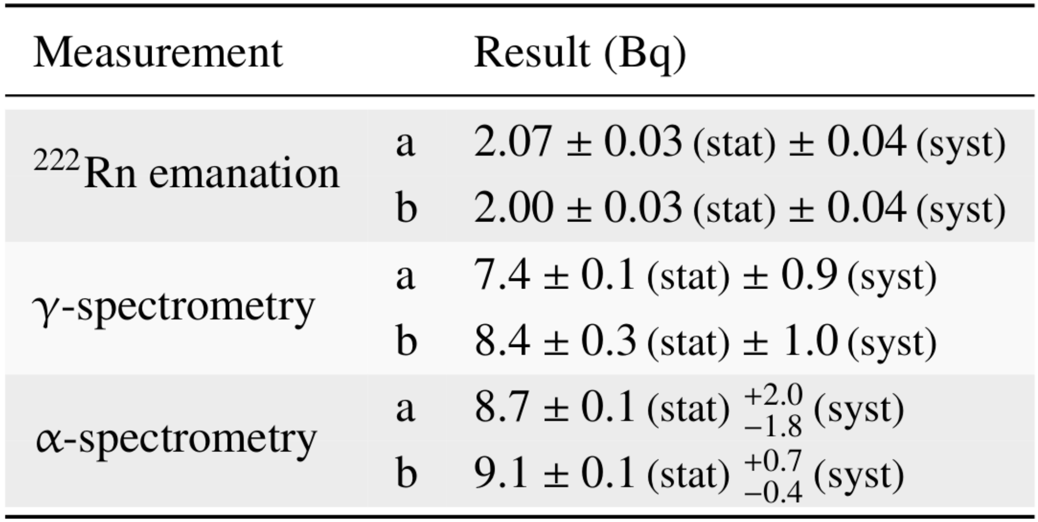
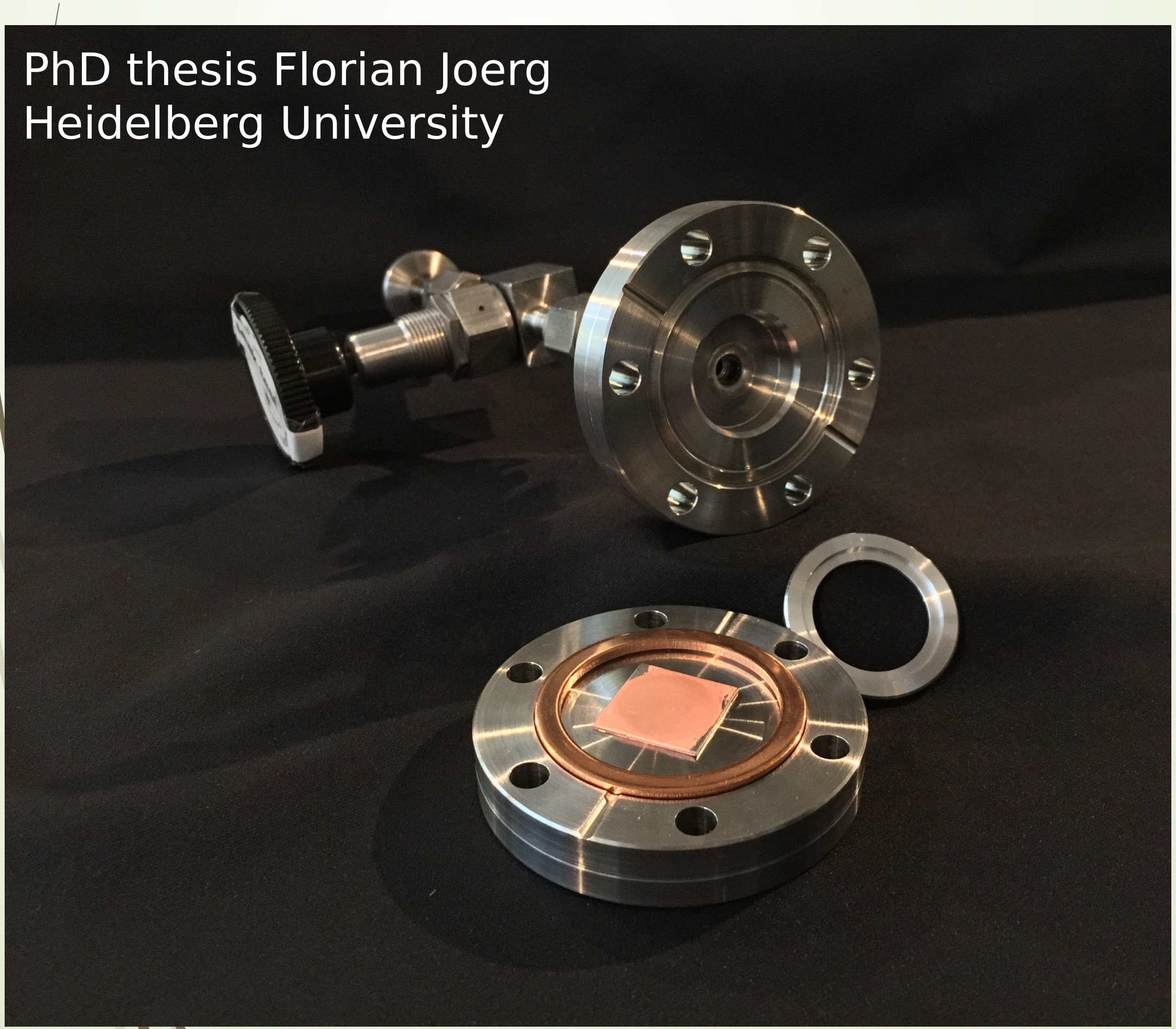




Measurement		Result (Bq)
^{222}Rn emanation	a	2.07 ± 0.03 (stat) ± 0.04 (syst)
	b	2.00 ± 0.03 (stat) ± 0.04 (syst)
γ-spectrometry	a	7.4 ± 0.1 (stat) ± 0.9 (syst)
	b	8.4 ± 0.3 (stat) ± 1.0 (syst)
α-spectrometry	a	8.7 ± 0.1 (stat) $^{+2.0}_{-1.8}$ (syst)
	b	9.1 ± 0.1 (stat) $^{+0.7}_{-0.4}$ (syst)

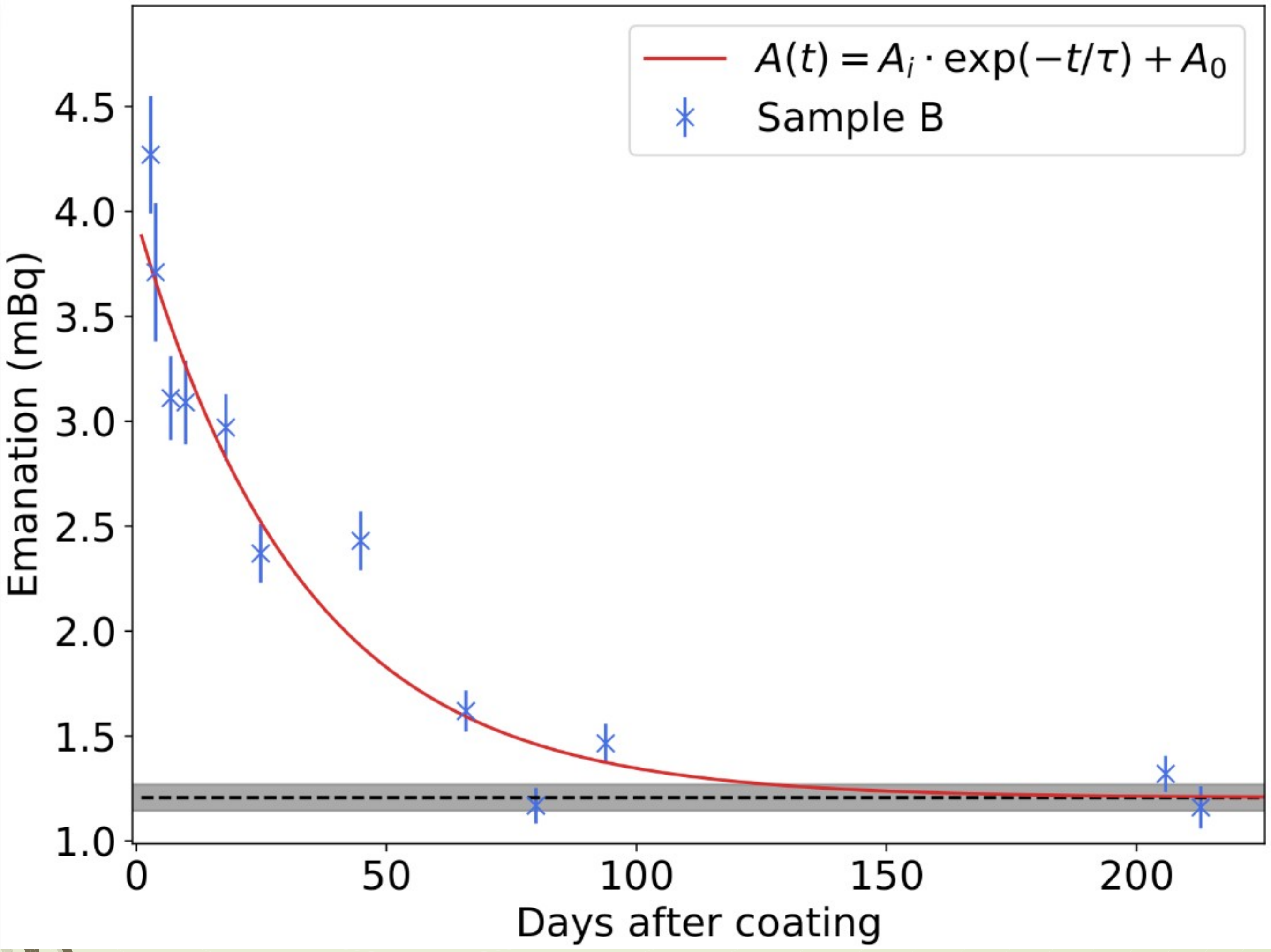
Coating of the 1st ISOLDE sample


PhD thesis Florian Joerg
Heidelberg University

- 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:

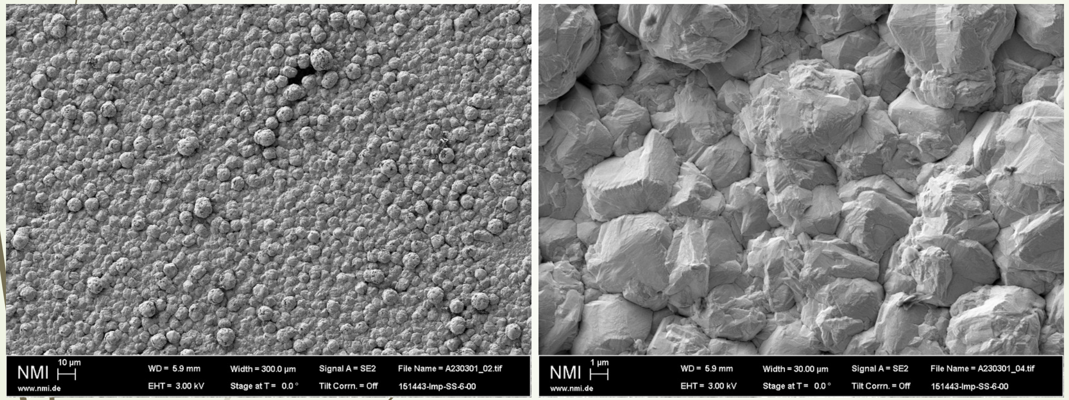
Activity [Bq]	^{226}Ra (186 keV)	^{222}Rn daughters
ISOLDE sample before coating	8.4 ± 1.0	6.0 ± 0.3
ISOLDE sample after coating	7.7 ± 1.0	7.2 ± 0.4
Electrolyte after coating	---	0.34 ± 0.02

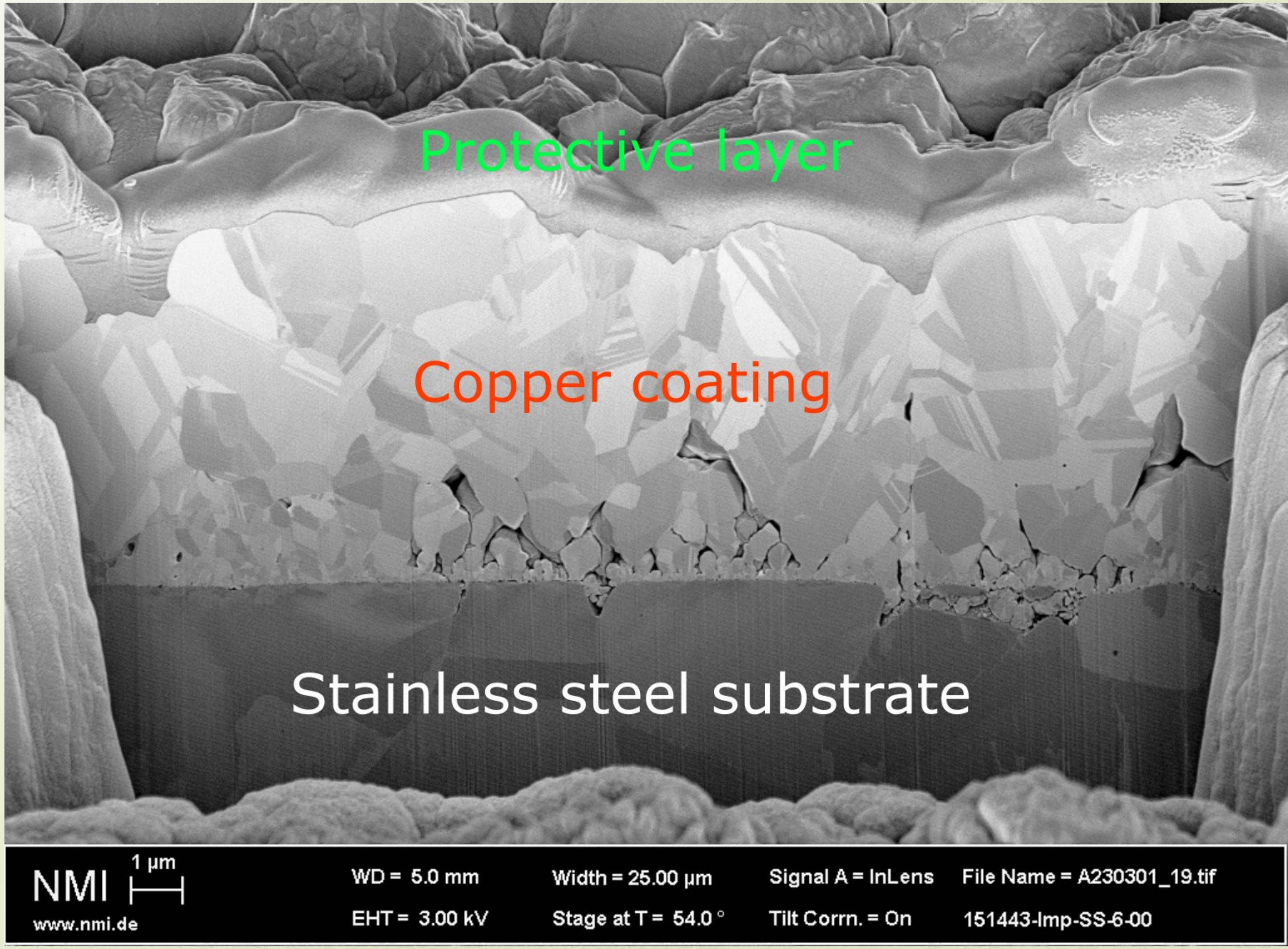
1st coated ISOLDE sample: Temporal development of ^{222}Rn emanation rate



- ^{222}Rn emanation rate was found to decrease
- Coating is getting “tighter”
- Oxidation?
  - But storage under protective atmosphere
- Re-crystallisation?
- **Final ^{222}Rn reduction factor: ~1700**
- Currently: Temperature dependency studies ongoing

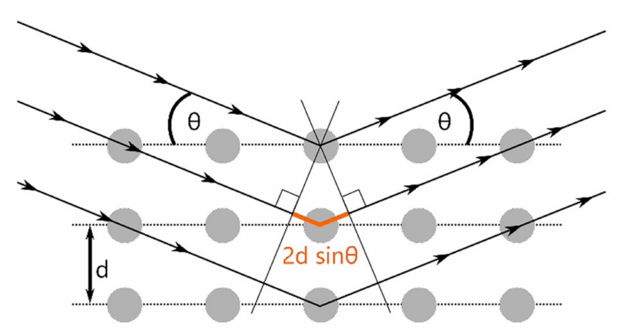
Secondary Electron Microscopy (SEM) investigation of our coating

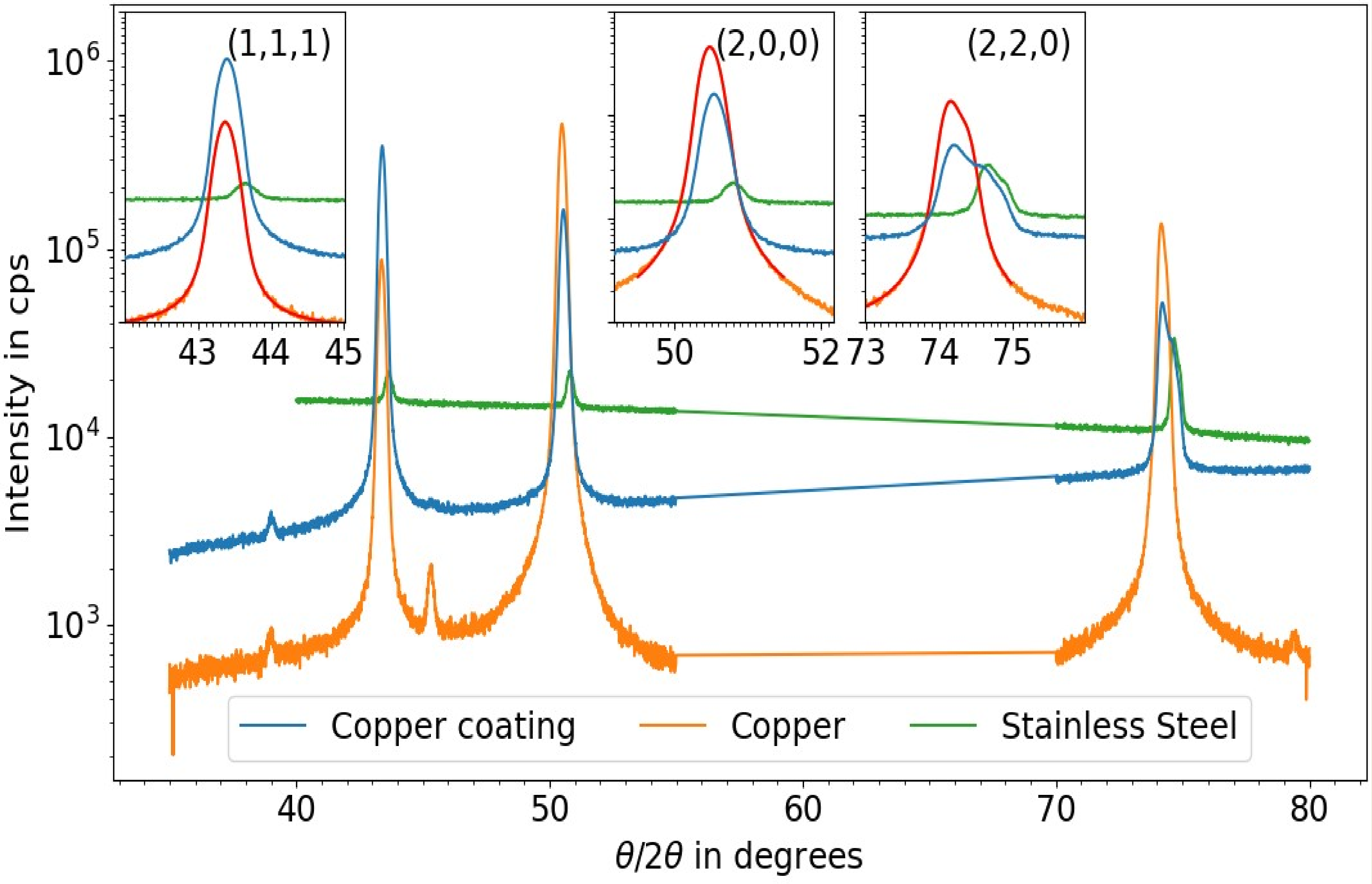




- Rough surface texture with spherical structures.
- Unhomogeneous adhesive layer with small grain size and holes.
- Tight cover layer with larger grains.

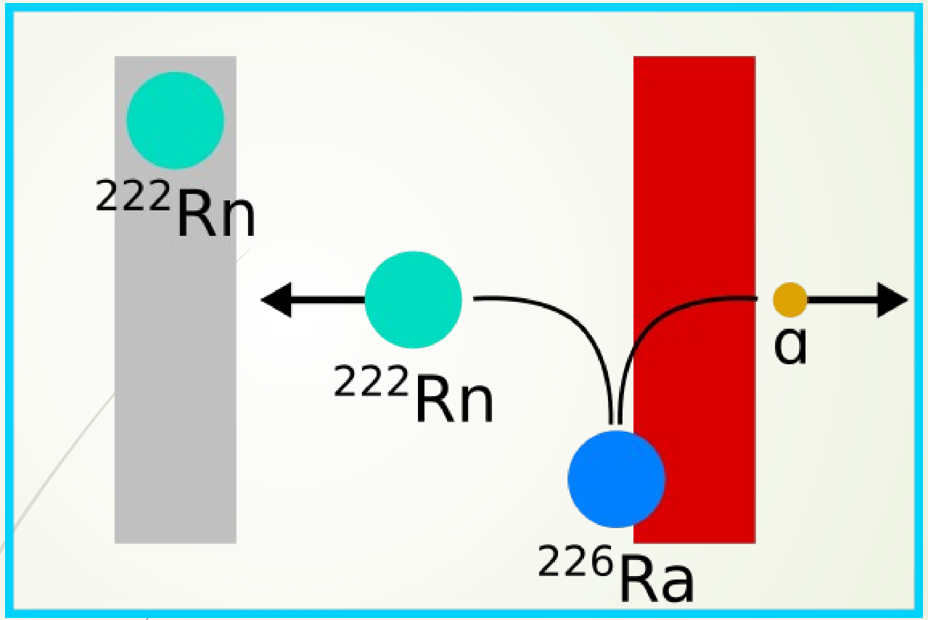
X-ray diffraction

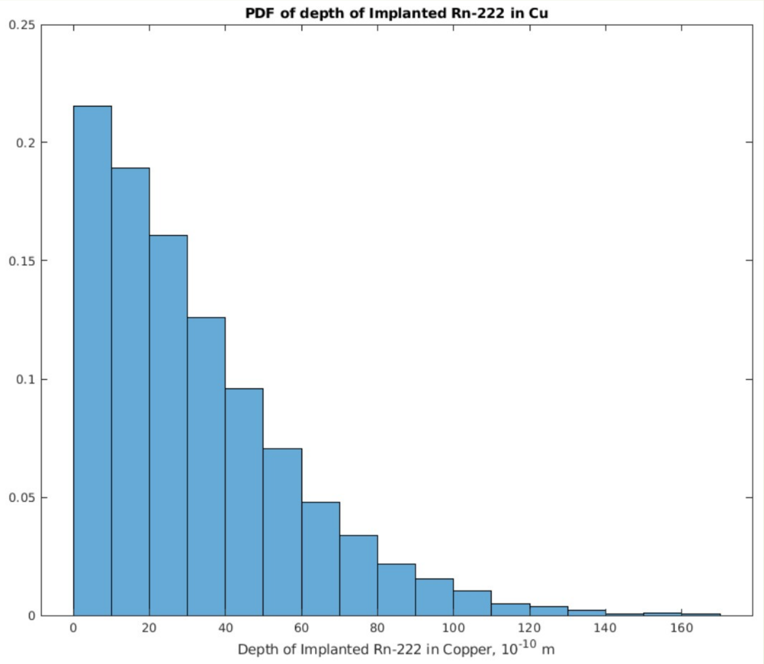




- Very preliminary study
- Done at Heidelberg University (IMSEAM: Institute for molecular systems engineering and advanced materials)
- Basic features as expected, but some unexplained effects:
  - Amplitude ratios doesn't always match expectation (directionality in lattice?)
  - Peak positions slightly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

Radon diffusion studies





- ^{226}Ra source: How to measure only ^{222}Rn emanation by diffusion (no recoil)?
- Need ^{222}Rn source without ^{226}Ra
- Recoil-implant ^{222}Rn in substrate and measure ^{222}Rn emanation
- Diffusion in metals is low → only ^{222}Rn close to surface relevant.
- Alpha counter → Total activity
- Proportional counter → Emanation fraction
- Simple diffusion model predicts emanated radon fraction:

$$R = \frac{Emanation(t)}{Activity(t)} \sim \sqrt{D \cdot t}$$

PRELIMINARY results

Material	Radon diffusion constant
Copper	$(5 \pm 2) \times 10^{-23}$ cm^2/s
HP Germanium	$\sim 2 \times 10^{-23}$ cm^2/s
HP Germanium (Li-doped)	$\sim 2 \times 10^{-22}$ cm^2/s
Stainless steel	$\sim 1 \times 10^{-22}$ cm^2/s
Quarz glass (amorphous)	$(4 \pm 2) \times 10^{-24}$ cm^2/s
Quarz crystal	$(3 \pm 1) \times 10^{-22}$ cm^2/s

$$R = \frac{Emanation(t)}{Activity(t)} \sim \sqrt{D \cdot t}$$