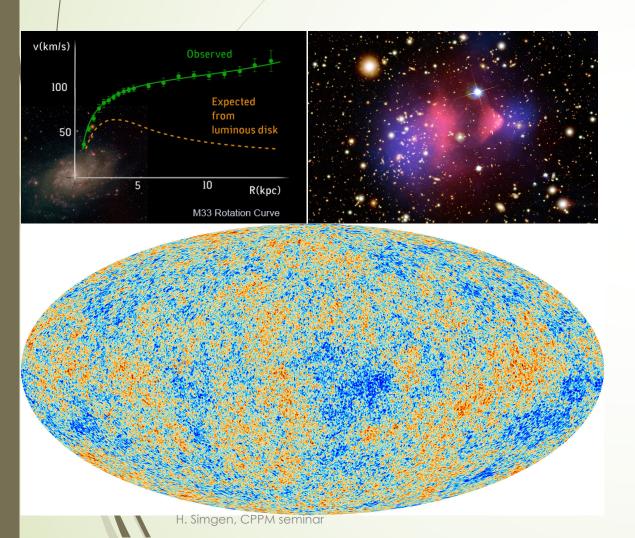
Noble, but Troublesome: Radon in Dark Matter Experiments

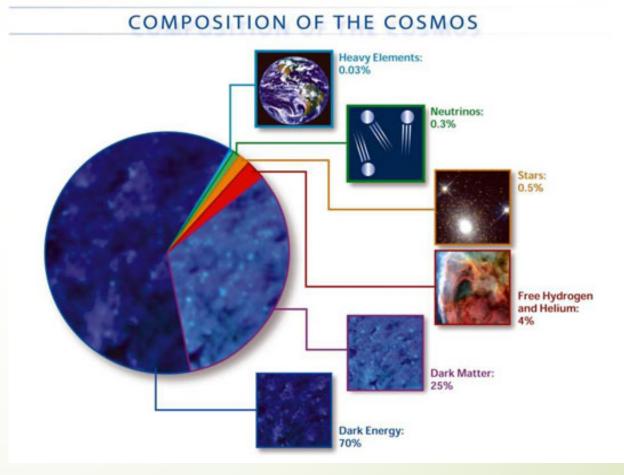
Hardy Simgen

Max-Planck-Institute for Nuclear Physics



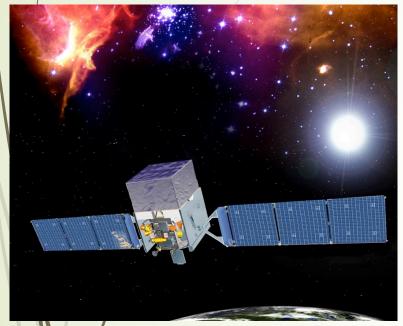
Dark Matter in the Cosmos





3

Indirect detection



$$\chi + \chi \to f + \bar{f}$$

Direct detection



$$\chi + N \rightarrow \chi + N$$

Production

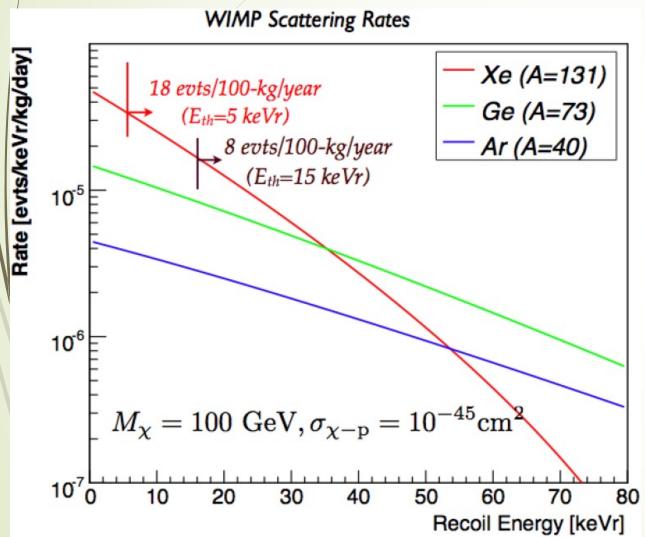


$$p+p \rightarrow \chi + ...$$

- 3 complementary channels
- very different techniques

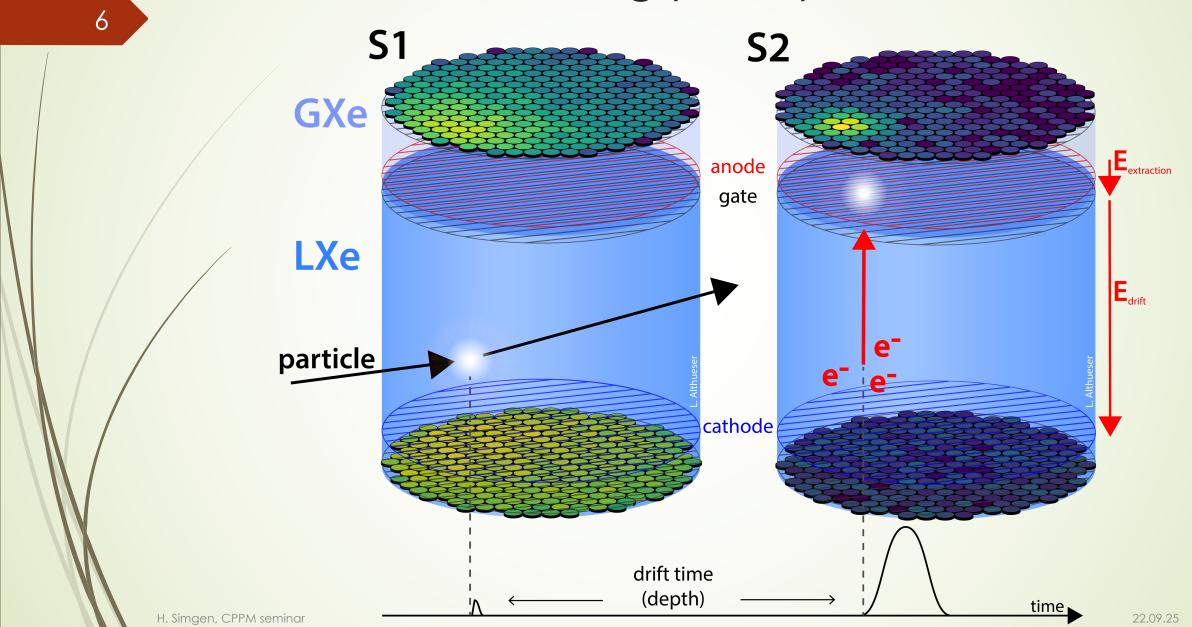


Why xenon?

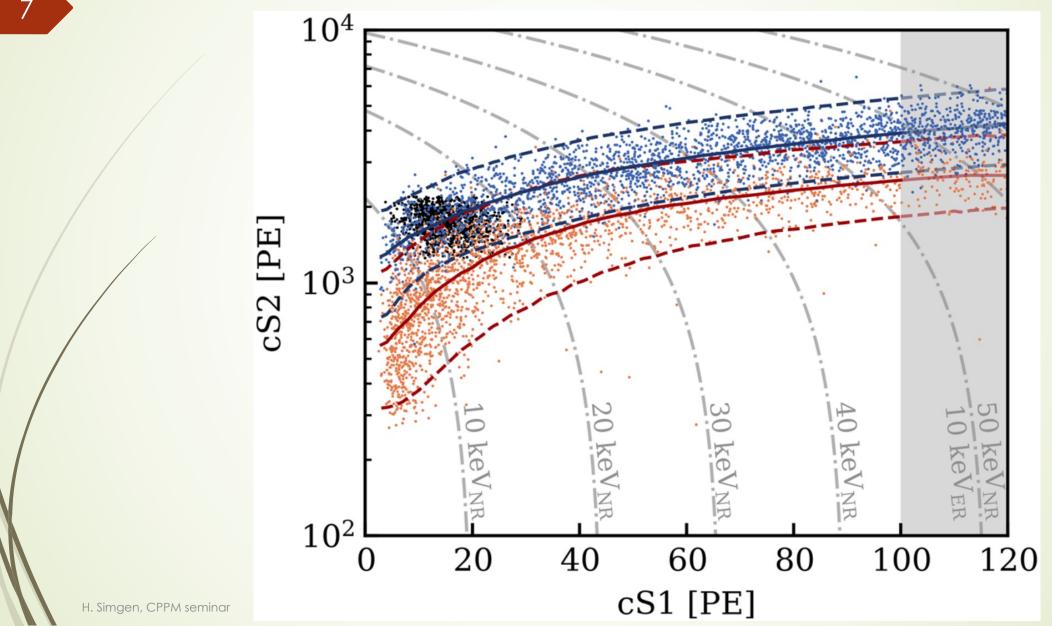


- High mass number (131) →
 relatively high rate
- LXe: Good self-shielding
- 178 nm scintillation photons:
 No wavelength shifter necessary
- No long-lived radioactive isotopes
- ¹²⁹Xe and ¹³¹Xe → spindependent interactions

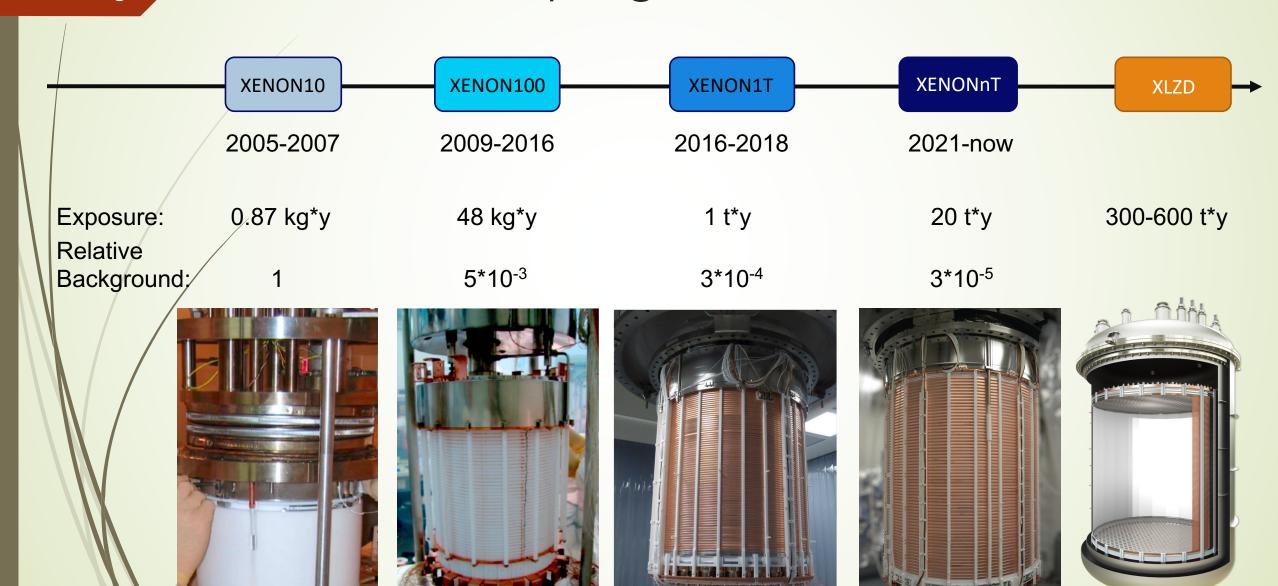
XENONnT working principle



Detector calibration

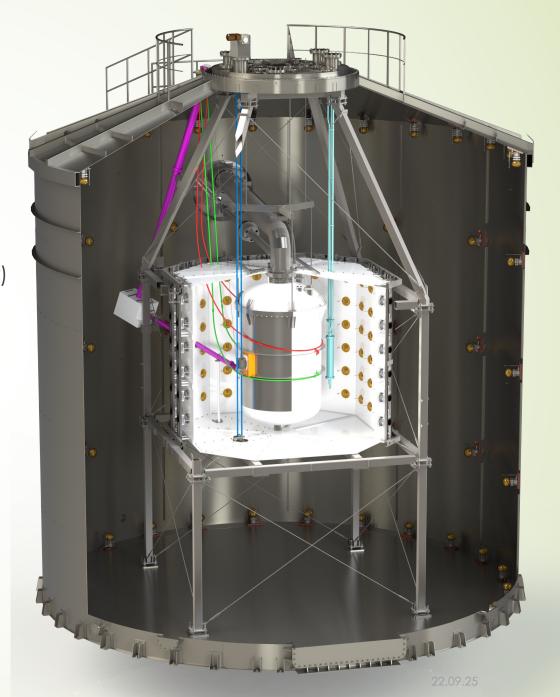


The XENON program

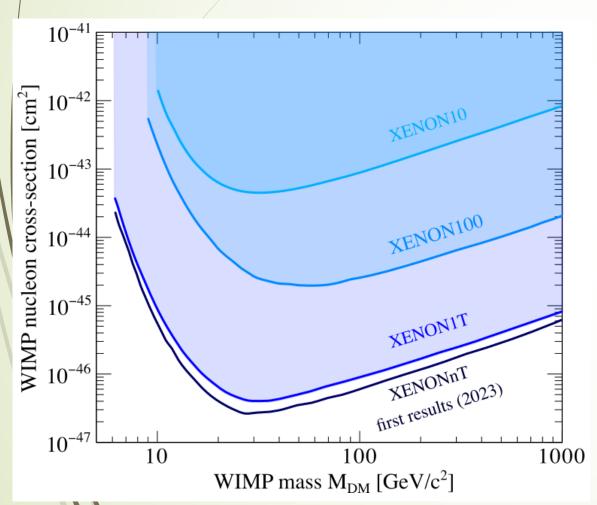


XENONnT

- 3 nested detectors
 - Water Cherenkov detector
 - Neutron veto system (Gd-loaded water)
 - Dual phase xenon TPC
- Many auxiliary systems
 - GXe purification system
 - LXe purification system
 - 2 xenon distillation columns
 - Cryogenic system
 - Calibration system
 - Xenon storage system
 - **...**



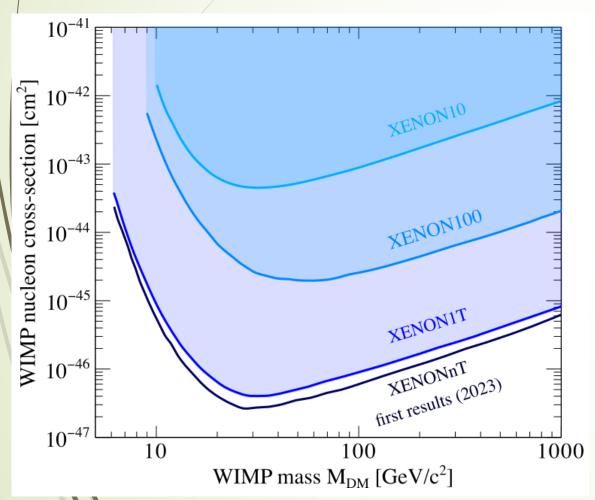
XENON experiments: WIMP results

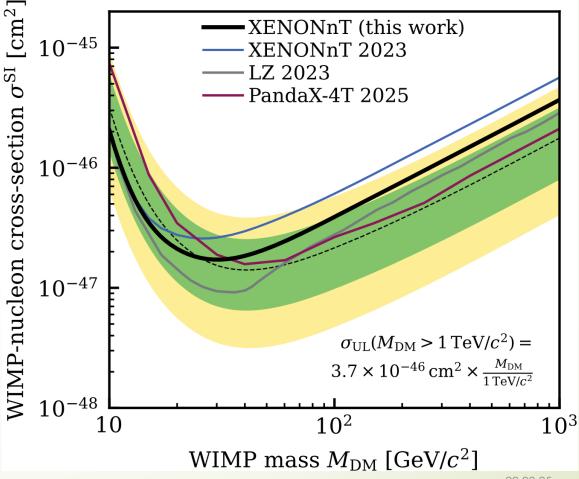


- Dual phase LXe TPCs have boosted direct dark matter search
- 4 orders of magnitude sensitivity improvement

XENON experiments: WIMP results

arXiv: 2502.18005



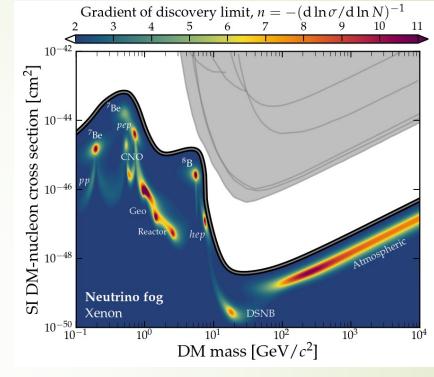


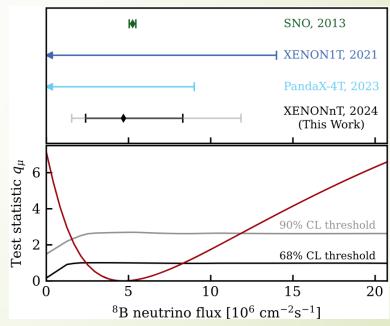
XENONnT: First signals from neutrinos

- DM sensitivity improved → neutrino signal within reach
- Special low-threshold analysis
- Very different background
- 2.7σ evidence for solar ⁸B neutrinos via CeνNS

Component	Expectation	Best fit
AC (SR0)	7.5 ± 0.7	7.4 ± 0.7
AC (SR1)	17.8 ± 1.0	17.9 ± 1.0
ER	0.7 ± 0.7	$0.5^{+0.7}_{-0.6}$
Neutron	$0.5^{+0.2}_{-0.3}$	0.5 ± 0.3
Total background	$26.4_{-1.3}^{+1.4}$	26.3 ± 1.4
8 B	$11.9_{-4.2}^{+4.5}$	$10.7^{+3.7}_{-4.2}$
Observed	3	7

PRL 133, 191002 (2024)





Electronic recoil background evolution

Background [events/t/y/keV]	XENON100 [PRD 83, 082001] (2011)	XENONnT [PRL 129, 161805] (2022)
Detector materials	1160	1.7
⁸⁵ Kr	860	0.6
²²² Rn (²¹⁴ Pb)	<140	5.9

► XENON100: ²²²Rn subdominant

Small detector → Material radioactivity dominant

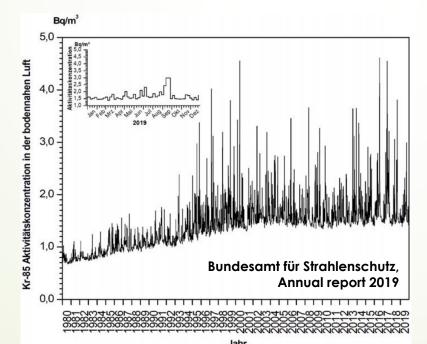
Kr problem not yet solved

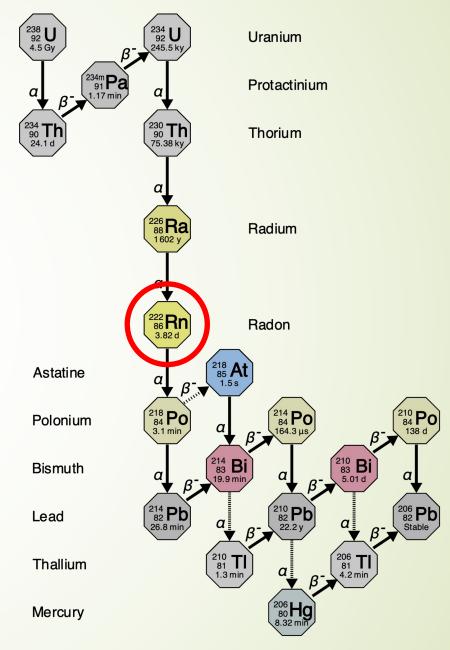
► XENONnT: ²²²Rn dominant

²²²Rn versus ⁵⁸Kr

14

	⁸⁵ Kr	²²² Rn
Half-life	10.4 years	3.8 days
Decay product	stable	Radioactive daughters
Origin	Man-made	Natural
Source	Nuclear fission	²³⁸ U chain



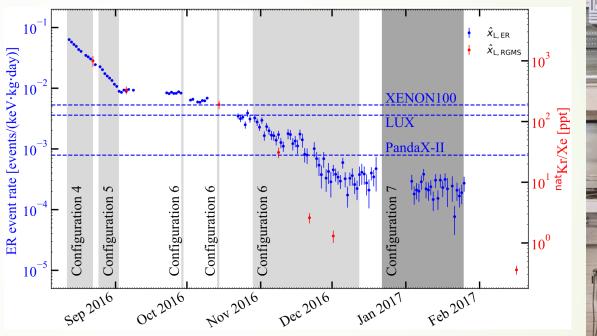


⁸⁵Kr in air (southern Germany)

H. Simgen, CPPM seminar

Kr removal in XENON1T / XENONnT

- Distillation column built for XENON1T
- Measured separation ~640.000 with 99% Xe recovery
- At least up to 6.5 kg/h throughput
- Lowest achieved concentration: 26 ppq
- Reliable Kr/Xe analytics with ppqsensitivity required



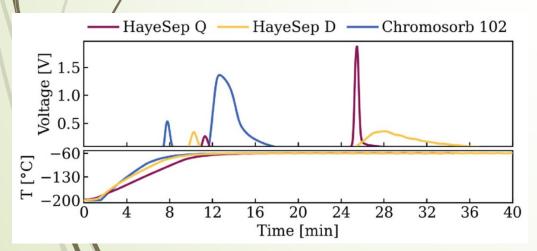
EPJC 77:275 (2017)



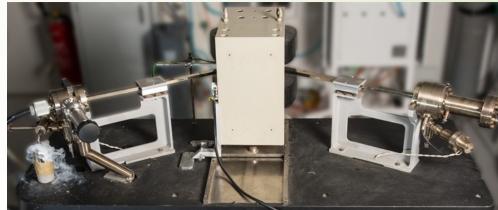
Rare Gas Mass Spectrometry 16 (RGMS) at MPIK

EPJC 74:2746 (2014)

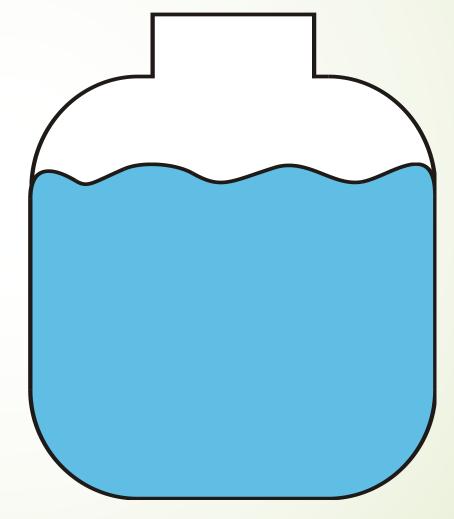
- Requires efficient removal of Xe \rightarrow gas chromatography.
 - Homemade UHV-qualified system
 - Low mass packed columns
- 8 ppg detection limit
- Future: Automated RGMS (Auto-RGMS)
 - Fully automated operation (Improved repeatability)
 - Improved separation columns



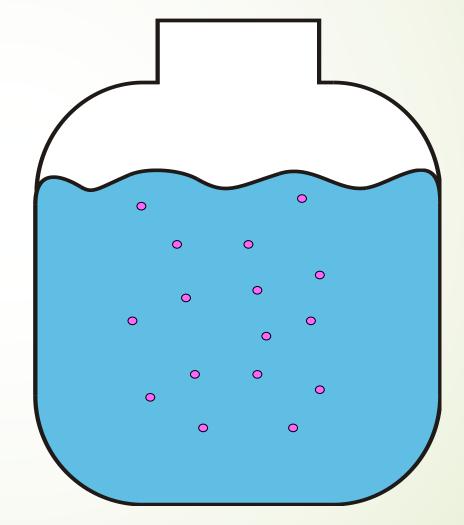
EPJC 85:576 (2025)



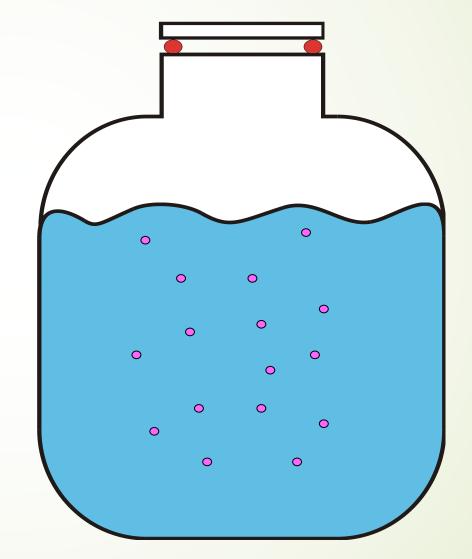




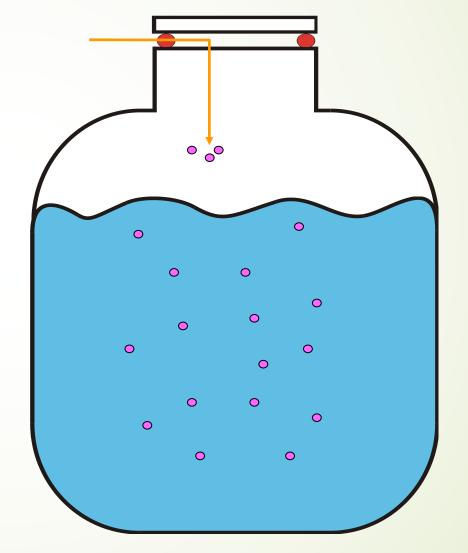
 Intrinsic radium (uranium/thorium) contamination



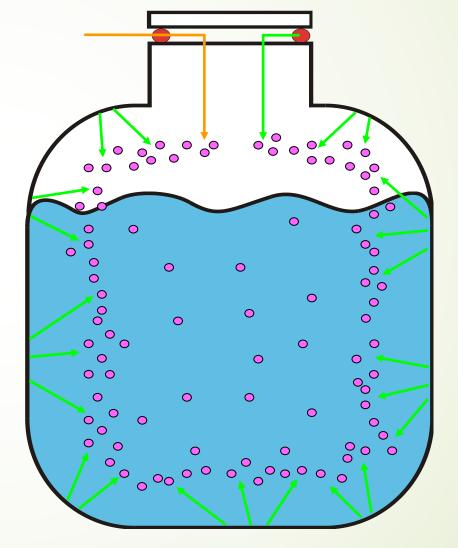
 Intrinsic radium (uranium/thorium) contamination



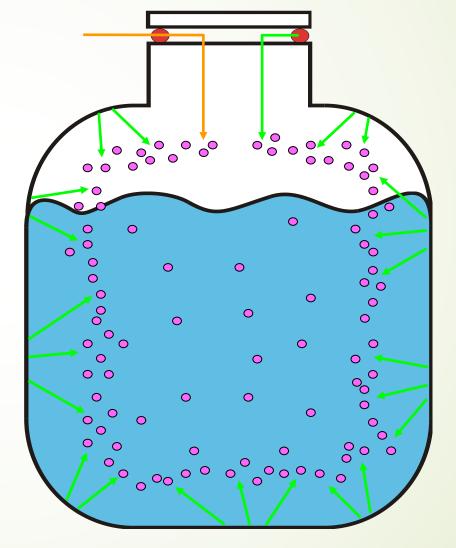
- Intrinsic radium (uranium/thorium) contamination
- Diffusion through seals



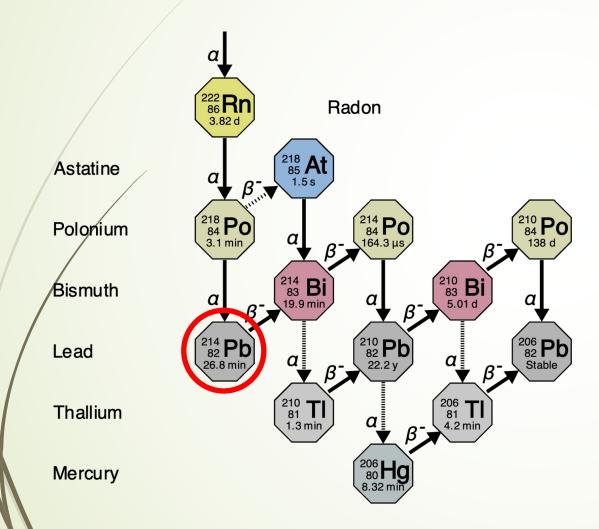
- Intrinsic radium (uranium /thorium) contamination
- Diffusion through seals
- Emanation from vessel and instrumentation



- Intrinsic radium (uranium /thorium) contamination
- Diffusion through seals
- Emanation from vessel and instrumentation
- A successful radon mitigation strategies must address all radon sources!



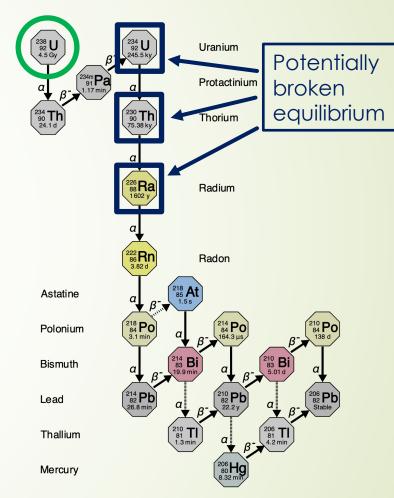
²²²Rn in the XENON experiments



- Dark Matter signal is low-energetic (<10 keV)
- Alpha emitter ²²²Rn, ²¹⁸Po, ²¹⁴Po: High energy events (MeV)
- Long-lived daughters out of equilibrium
- 214Bi-214Po coincidence
- Background only due to ²¹⁴Pb: Isolated beta emitter

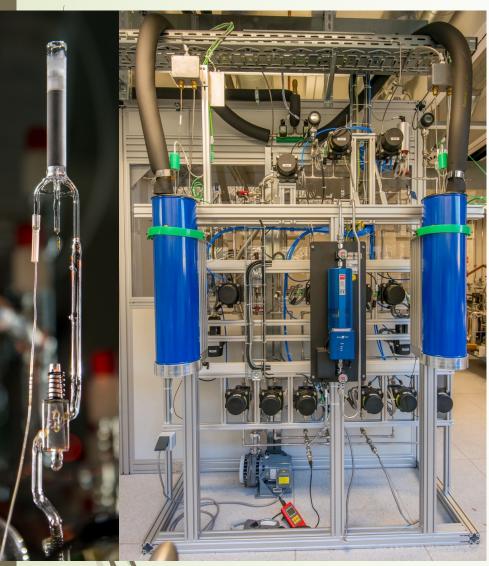
Radon detection (at low level)

- All radon detection by measurement of radioactivity (Alpha decays)
- Low-level → Requires radon collection
 - By cryo-trapping
 - By electrostatic collection of charged daughters
- Low-level → Clean materials and vacuum
- Warning: ICP-MS / NAA has fantastic sensitivity for U/Th. But radioactive may be broken, so no information about Rn!

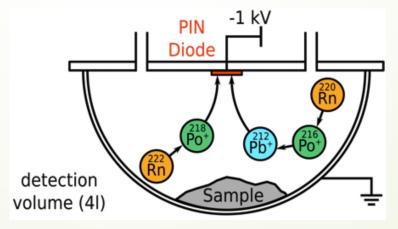


By http://commons.wikimedia.org/wiki/User:BatesIsBack

Material screening by ²²²Rn emanation



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





H. Simgen, CPPM seminar

The radon emanation process

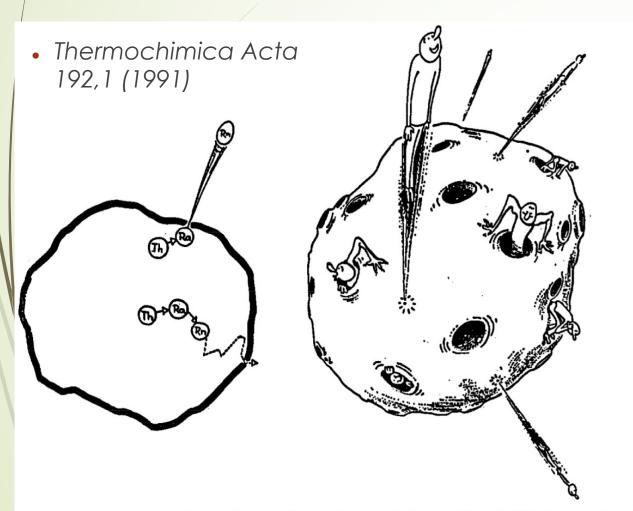


Fig. 2. Scheme of the release of radon from the sample by recoil and diffusion mechanisms.

- 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 emanation from charcoal

- Many charcoals screened at MPIK:
 - 30 years ago: Discovery of "CaroAct"
 - 15 years ago: Discovery of "Saratech"

MIM A 903, 267 (2018)

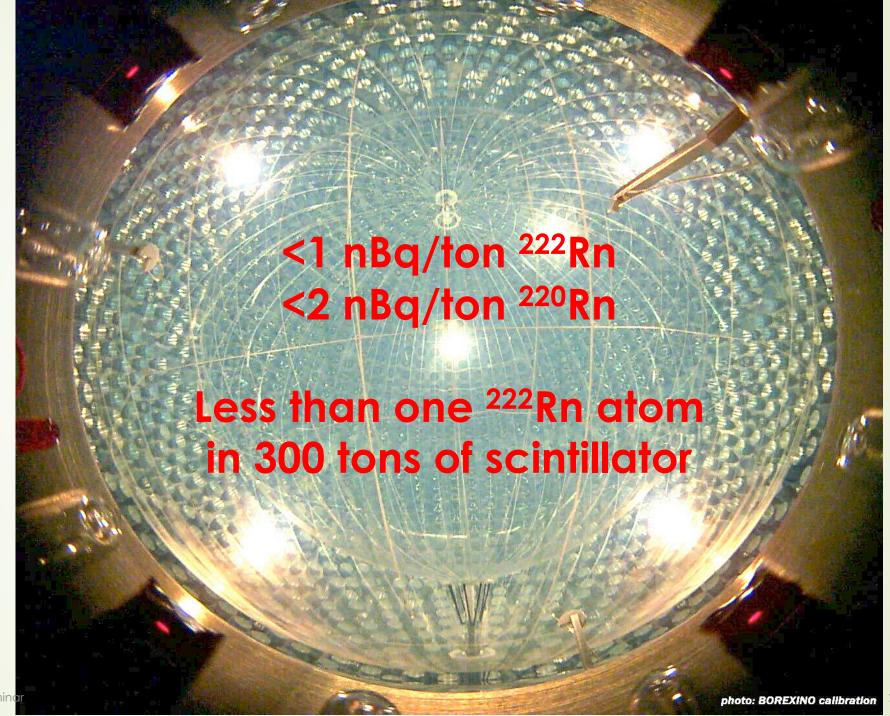
Charcoal	Specific activity (mBq/kg)	Price (USD/kg)	References
Calgon OVC 4 × 8	53.6 ± 1.3	6	This work
Shirasagi G2 \times 4/6–1	101.0 ± 8.0	27	This work
Saratech	1.71 ± 0.20	35	This work
HNO ₃ etched Saratech	0.51 ± 0.09	135	This work
Carboact	0.23 ± 0.19	15,000	This work
Carboact	0.33 ± 0.05	15,000	[28]

²²²Rn emanation from Saratech charcoal

- 222Rn release rate strongly depends on extraction parameters
- 226Ra activity (HPGe spectroscopy): (7 ± 1) mBq/kg
- Gamma-ray measurement of HNO₃-cleaned charcoal ongoing.
- 222Rn emanation into xenon?

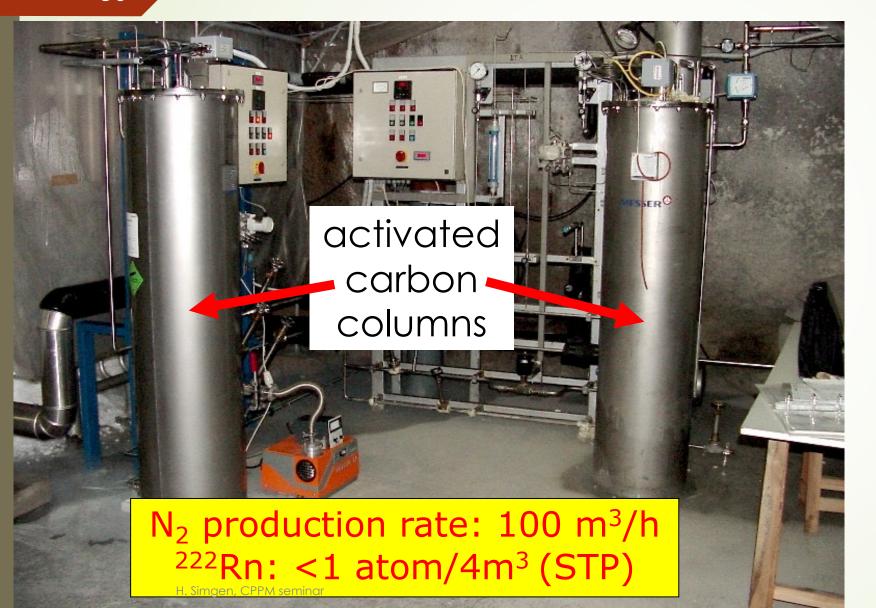
Charcoal	Extraction conditions	²²² Rn emanation rate [mBq/kg]
Saratech	Room temperature	<0.3
Saratech	200 °C and prolonged extraction	3.5 ± 0.2
HNO ₃ -cleaned Saratech	200 °C and prolonged extraction	1.0 ± 0.1

BOREXINO



High Purity nitrogen for BOREXINO

30

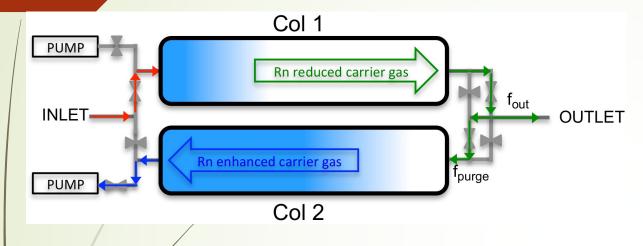


- Radon-free nitrogen for scintillator sparging
- Large flow-rate (100 m³/h)
- Simple concept withlittle maintenance needed
- Lqiuid nitrogen cooled columns
- Purifications of liquid nitrogen (less efficient, but high throughput)
- Key: Low ²²²Rn emanating activated carbon

Appl. Rad. Isot. 52 (2000) 691

Improving adsorption gas purification

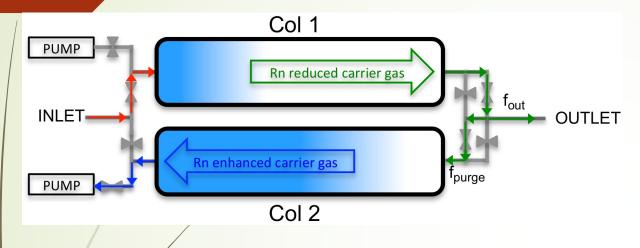
31



- 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

32

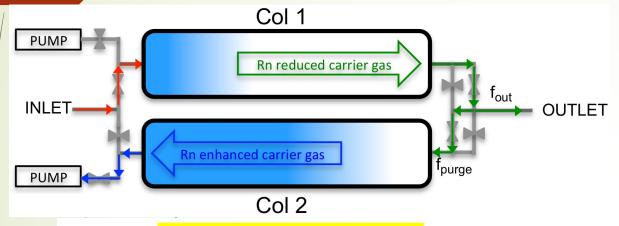


- Improving performance by doubling columns and avoiding equilibrium
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- Gain in radiopurity → purer gas
 - JINST 16 P07047 (2021)
- New adsorber materials → José Busto
 - Carbon molecular sieves
 - metal organic frameworks
 - molecular cages
 - carbon aerogel

•

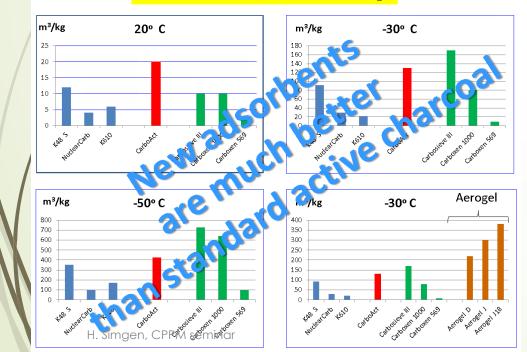
Improving adsorption gas purification

33



- Improving performance by doubling columns and avoiding equilibrium
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K_factor for several materials in N₂



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

J. Busto, GDR Nov. 22

²²²Rn emanation rate of some stainless steel vessels

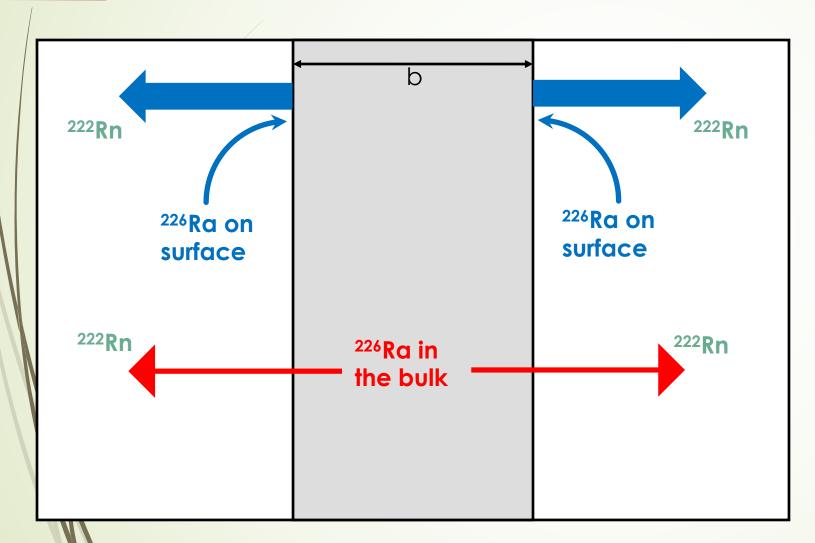
Vessel	Size	²²² Rn emanation rate [mBq]
BX storage tank HT2	2.1 m ³	1.2 ± 0.4
BX storage tank D330	1.6 m ³	7.1 ± 1.2
GERDA cryostat	64 m ³	13.7 ± 1.9
XENON1T cryostat	~2 m³	1.8 ± 0.3
XENONnT cryostat	~4.5 m ³	1.9 ± 0.2
Borexino water extraction column	0.6 m³, 608 m² (SS packings)	$4.8 \pm 0.7 \rightarrow 8 \mu Bq/m^2$

Nylon film during measurement @ MPIK

35

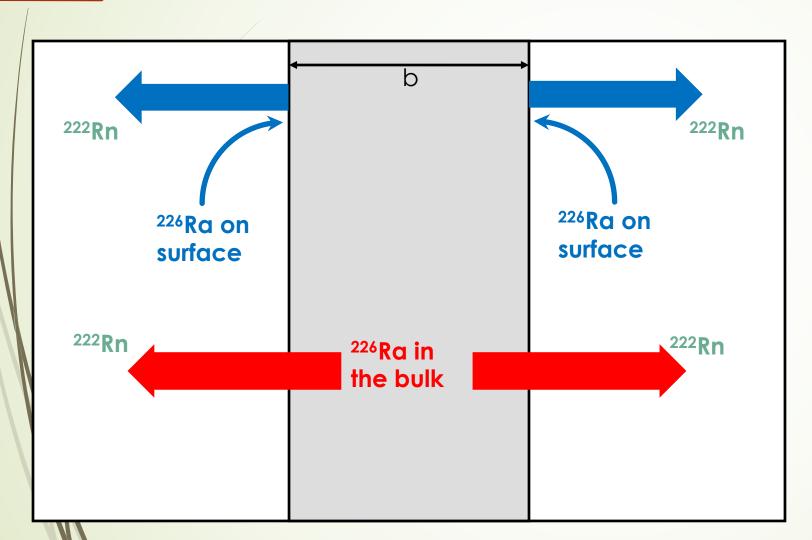


Radon in the BOREXINO nylon vessel



- Radon measurement from thin nylon foild yields
 - Contribution from surface plus
 - Unknown fraction of bulk contamination
- For ²²²Rn 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



- $d_{dry} = (270 \pm 30) \mu m$
- 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:
 - $A_{bulk} < 15 \mu Bq/kg$
 - $A_{surf} < 0.9 \mu Bq/m^2$

Understanding ²²²Rn source distributions

38

Example: XENON1T setup heat exchangers + heater cable feedthrough pipe with cables **QDrive pumps** cryogenic pipe cryogenic ····|·· gas purifiers system **TPC** cryostat. purification system EPJC (2021) 81:337

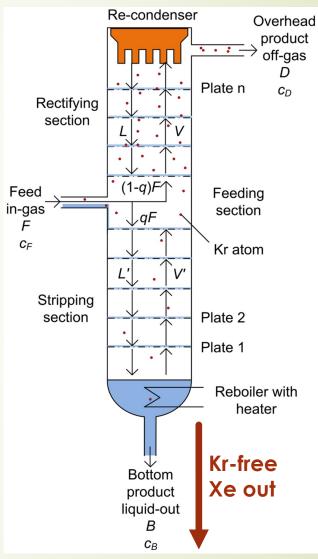
Understanding ²²²Rn source distributions

39

Example: XENON1T setup heat exchangers + heater cable feedthrough pipe with cables **QDrive pumps** ~19 cryogenic cryogenic pipe ·····|·· gas purifiers system ~1 **TPC** <4 cryostat. purification system EPJC (2021) 81:337

Radon removal by xenon distillation

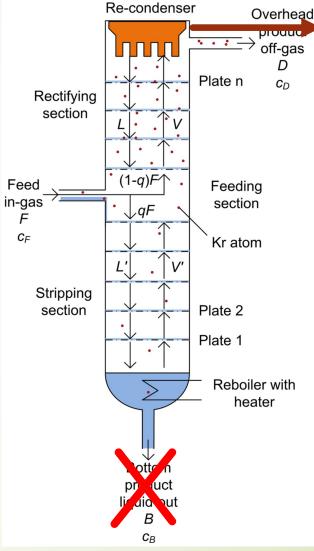
- Radon is less volatile than xenon → reverse operation mode w.r.t. Kr removal
- Feasibility demonstrated in Xenon100: EPJC (2017) 77:358
- Applied in XENON1T: 4.5 μBq/kg achieved: EPJC (2021)81:337
- High flow radon removal system developed for XENONnT: arXiv: 2205.11492
- 0.8 μBq/kg achieved
- /Future: Further upscaling?
 - Need to process entire budget in O(²²²Rn half-life)
 - Processing speed for XLZD 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



PandaX talk at LRT 2024 workshop

Surface Treatments to Boost the Sensitivity



Different types of polishing methods were tested on the same chamber to lower the radon background.







	7.4 SS detector	7.4 SS detector	7.4 SS detector
Polishing	Electrochemical	Mirror	Mirror + electrochemical
Roughness [um]	3.00 ± 0.44	0.12 ± 0.04	0.13 ± 0.03
Blank [mBq/m²]	9.08 ± 0.71	0.48 ± 0.14	0.33 ± 0.10
Bulk* [mBq/kg]		10.15±0.68	

*ICP-MS ²³⁸U-early chain measurement

Radon mitigation by surface polishing

- 222Rn emanation often due to surface impurities
- 2 effects of polishing
 - Removing material
 - Smoothening
- Tests with 4% thoriated tungsten → smoothening is the dominant effect
- Metallic samples → Rn diffusion negligible → ²²²Rn emanation only due to surface impurities

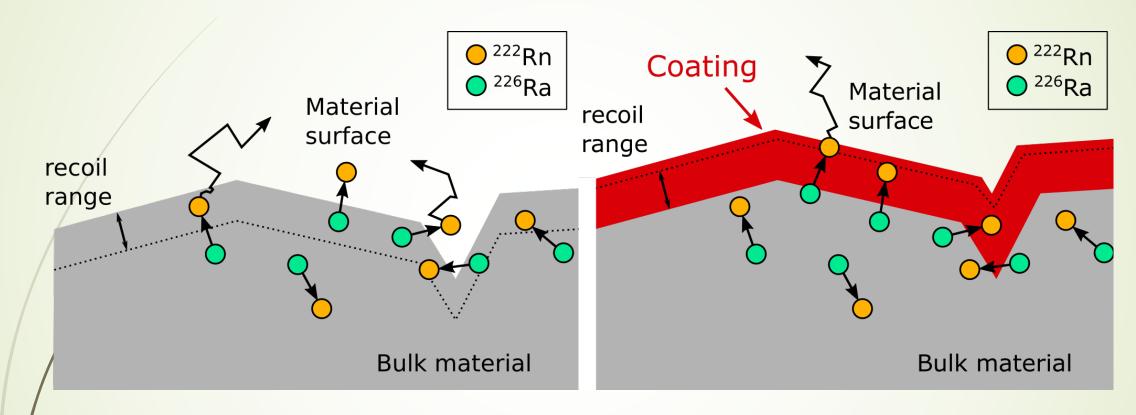


Roughness 2.5 micron



Roughness 1.5 micron

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

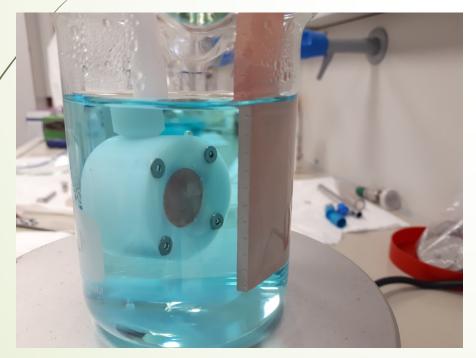
Radon mitigation by surface coating

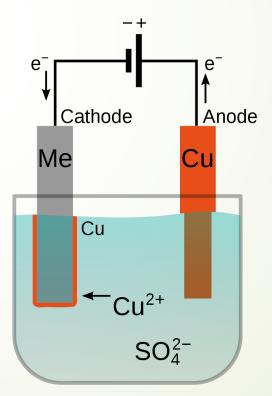


- Countless different coating techniques exist: Where to start?
- Required properties: Tight, homogeneous, stable (also at low temperature)
- Coating cleanliness may be ultimate limitations → start with the cleanest!
 - Vacuum processes
 - Physical Vapor Deposition (PVD)
 - Chemical Vapor Deposition (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 ²²⁰Rn: Much shorter half-life than ²²²Rn

Copper electro-deposition

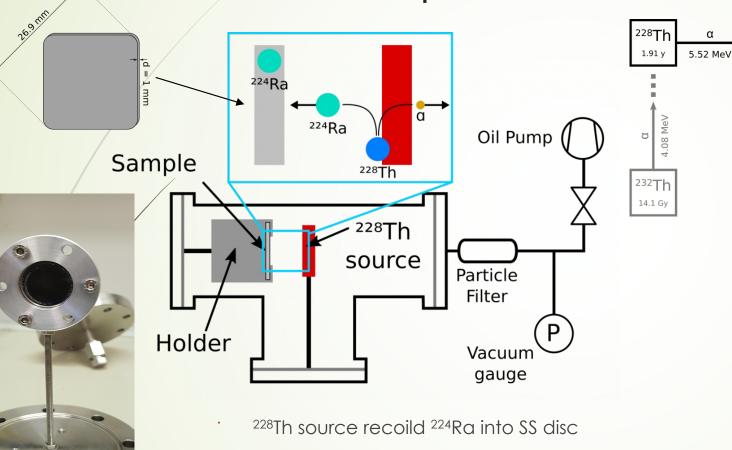
- Motivated by experience with electro-formed copper (clean!)
- In-house development at MPIK: 5 µm thick Cu layer
- Anode made from copper
- Good mechanical stability on tungsten
- Efficient blocking of ²²⁴Ra alpha-decay
- ²²⁰Rn reduction factor up to 100 observed!







From tungsten to stainless steel: Recoil-implanted ²²⁴Ra



Test coating 2.00 Surface current density Voltage current density [mA/cm²] - 1.75 1.50 ^{-1.25}∑ - 0.75 O 20 - 0.50 0.25 -0.00 800 1000 200

²¹⁶Po

145 ms

²¹²Po

299 ns

6.91 MeV

²¹²Bi

8.95 MeV

6.40 MeV

5.79 MeV

²¹²Pb

6.21 MeV

²⁰⁸Ph

208**T**|

3.05 min

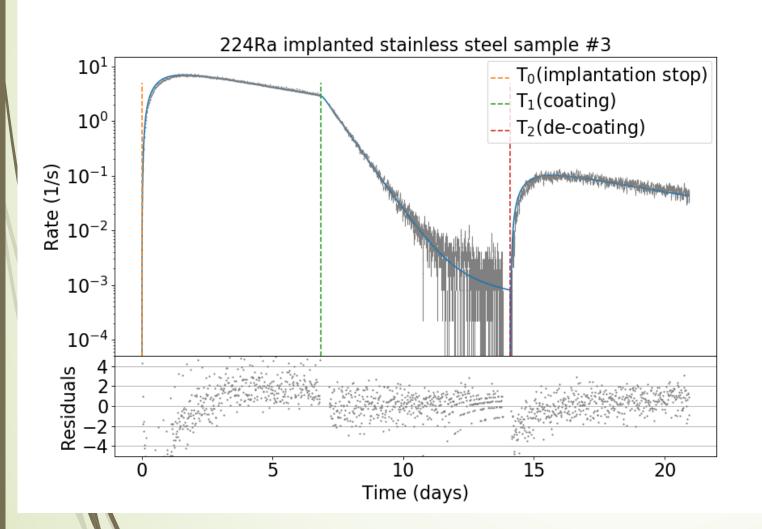
Procedure modification: Adhesion layer (~1 µm) for mechanical stability

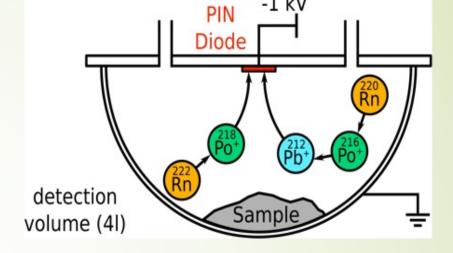
Time (s)

Short-lived (t_H=3.7 days) ²²⁰Rn-emanating SS disc

47

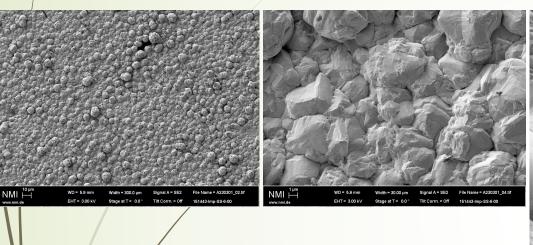
Coating (and de-coating) of ²²⁴Ra-implanted SS disc



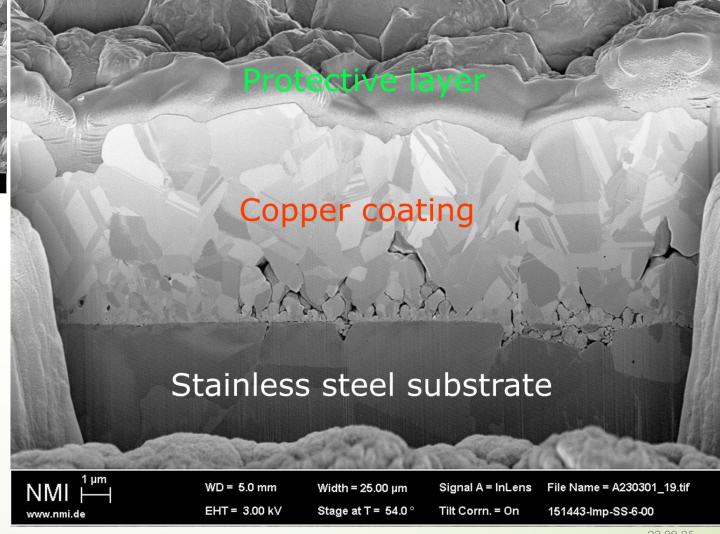


- ²²⁴Ra decays on SS disc
- ²²⁰Rn is emanated and decays in electrostatic radon monitor
- Charged ²¹²Pb (²²⁰Rn-daughter) is collected on PIN diode
- Counting of ²¹²Po a-decays
- Possible issue: Activity wash-off in electrolyte
- Procedure: Implantation → coating → de-coating
- Upper limit from coating,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

Secondary Electron Microscopy (SEM) investigation of our coating

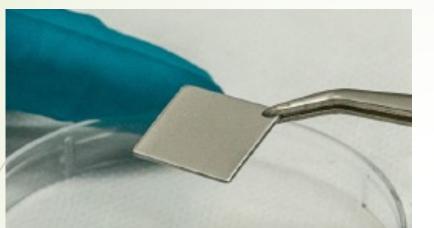


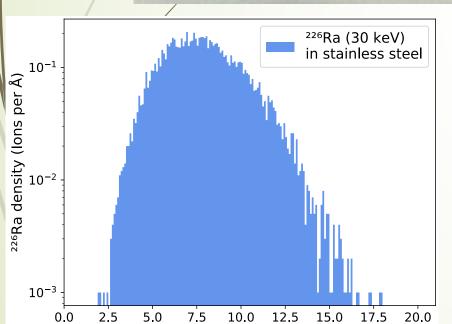
- Rough surface texture with spherical structures.
- Unhomogeneous adhesive layer with small grain size and holes.
- Tight cover layer with larger grains.



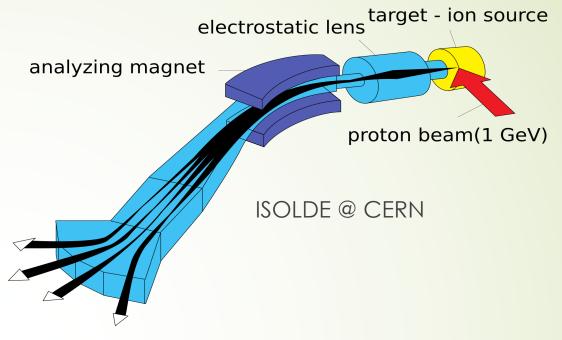
Finally: ²²²Rn on SS







50



- radioactive ion beams
- ²²⁶Ra-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

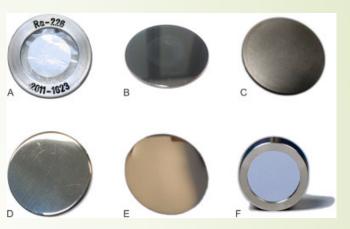
Depths (nm) H. Simgen, CPPM seminar

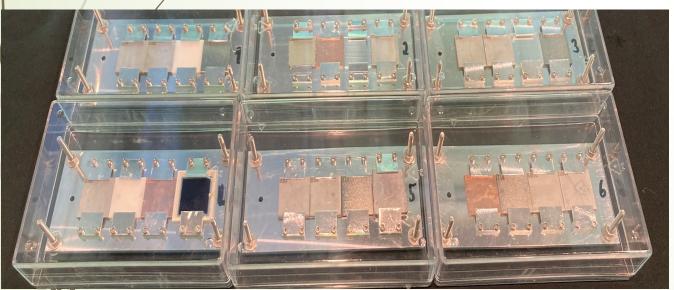
Novel ²²²Rn sources

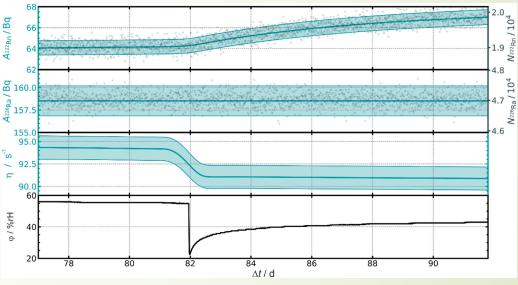
51



- ²²⁶Ra implantation in SS, Cu, Ti, Pb, Ge, Si, PTFE, SiO₂, acrylic.
- >30 samples.
- Similar development at PTB (Braunschweig): Traceable ²²²Rn sources.
- Deposition on an active Si detector allows online emanation rate monitoring.





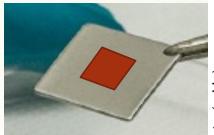


Appl. Rad. Isot. 196 (2023) 110726

H. Simgen, CPPM seminar

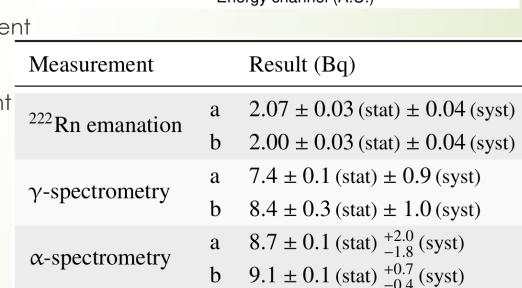
52

ISOLDE sample characterization



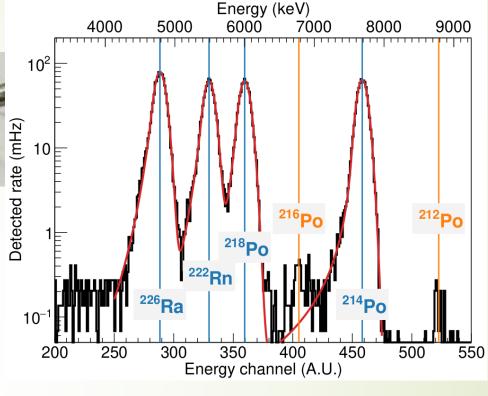


- Short-lived contaminants (from ²³⁵U chain)
- ~8.5 Bq ²²⁶Ra activity
- Central deposition confirmed
- Gamma measurement confirms alpha measurement
 - Unexpected ¹³⁹Ce discovered (†_H = 137.6 d)
- Direct ²²²Rn emanation test with proportional count
 - Sample a: (2.00 ± 0.05) Bq
 - Sample b: (2.07 ± 0.05) Bq
- Wipe test: Less than 1% of activity removed









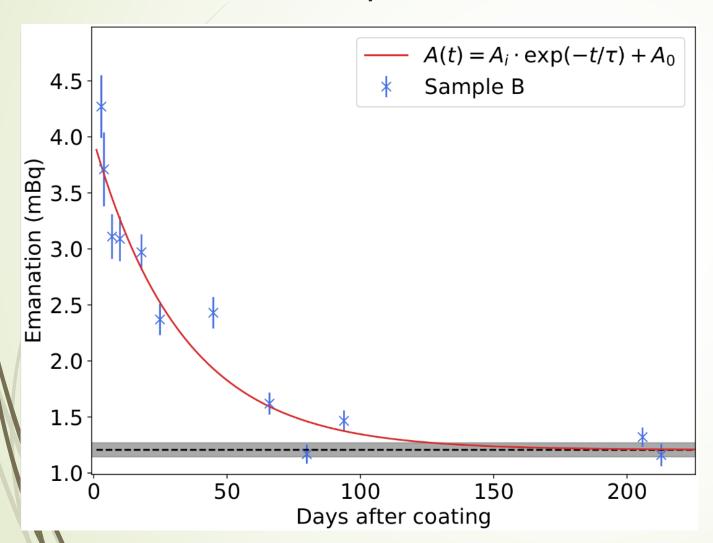
Coating of the 1st ISOLDE sample



- Standard MPIK electrochemical Cu-on-SS coating recipe applied
- ²²²Rn emanation rate
 - Before coating: (2.00 ± 0.05) Bq
 - After coating: (4.3 ± 0.3) mBq
- Unexpected large ²²²Rn 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 ²²²Rn emanation rate



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

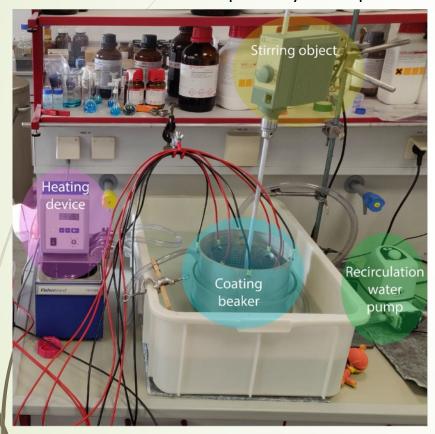
Upscaling

- All samples so far were few cm² size
- Applicability to XLZD requires (much) larger coated surfaces.
 - Larger coating vessels
 - Higher-current power supply
 - New copper electrode
 - Better temperature control
 - Improved mixing
- New samples:
 - Stainless steel cylinders
 - ~20cm height
 - ~20cm diameter
 - 1250 cm² surface



Upscaling

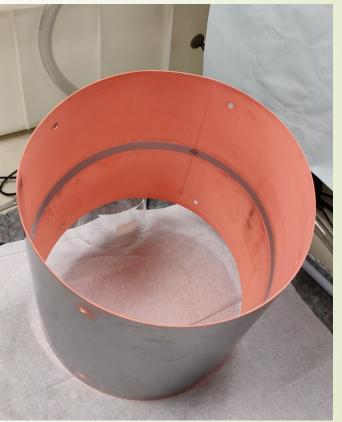
First tests with temporary setup



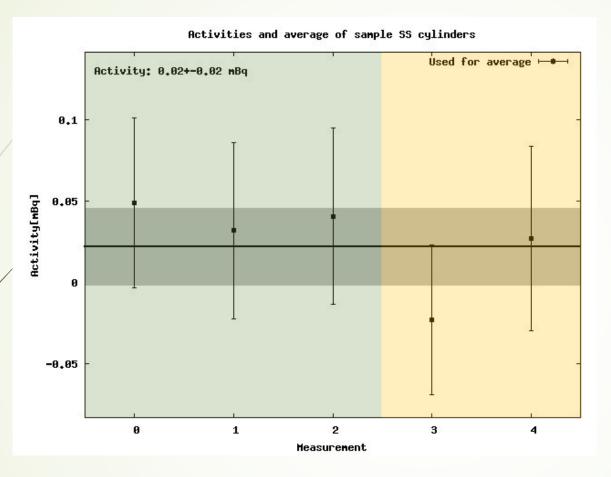
New copper electrode



First "large surface" coating



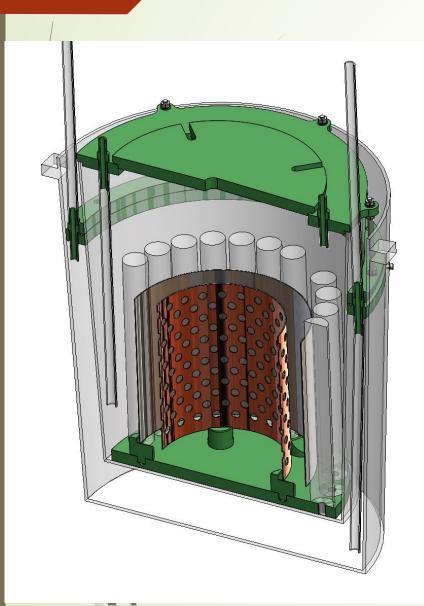
First radiopurity tests



- 222Rn emanation rate from five stainless steel cylinders measured before and after coating.
- No increase observed: Extra ²²⁶Ra introduced <80 μBq/m².

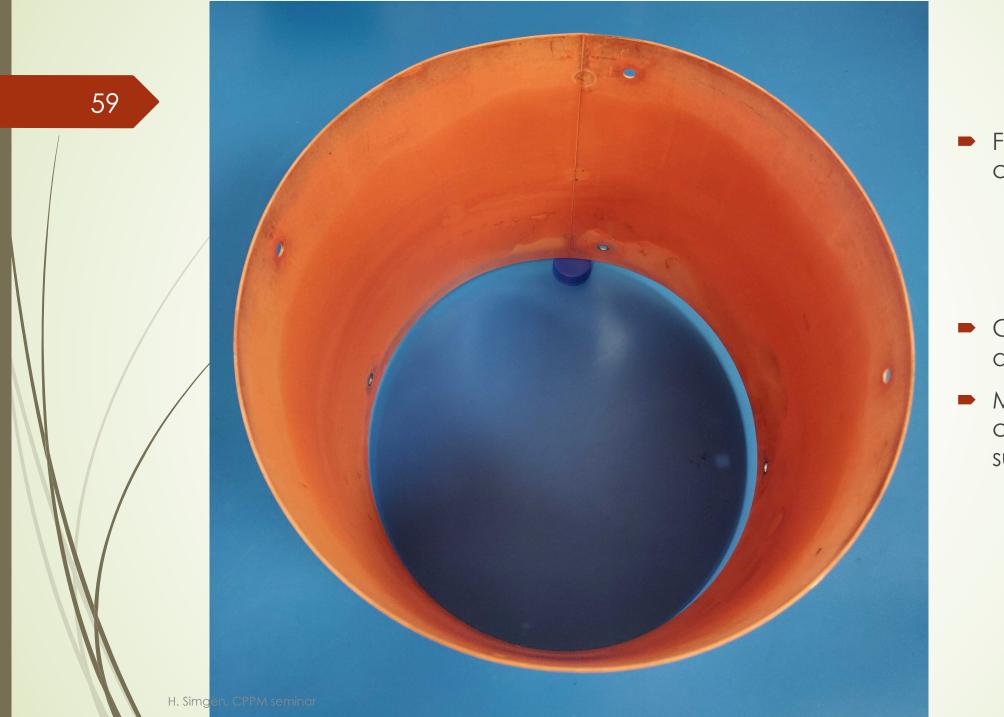


New dedicated coating vessel

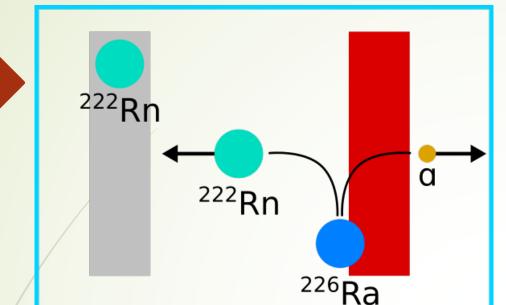


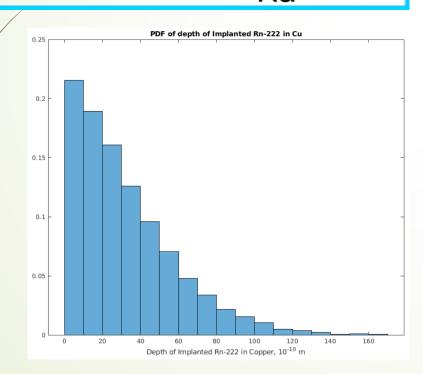


- Newly designed HDPE vessel.
- Sufficiently high.
- Stronger power supply unit.
- Electrolyte immersed in thermally controlled water bath.



- Fully coated inner cylindrical mantle.
 - ~20 cm height
 - ~20 cm diameter
 - → ~1250 cm² surface
- Coating is stable and homogenous.
- Meanwhile > 10
 cylinder mantles
 successfully coated.





Radon diffusion studies

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

$$R = \frac{Emanation(t)}{Activity(t)}$$

$$\sim \sqrt{D \cdot t}$$

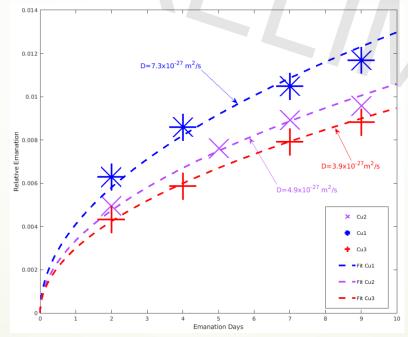
60

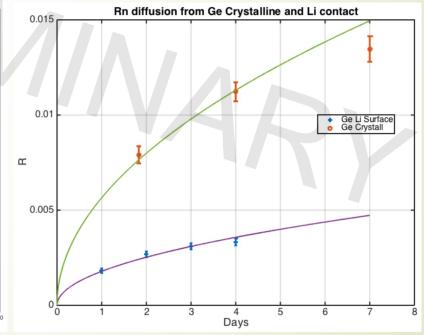
PRELIMINARY results

61

$$R = \frac{Emanation(t)}{Activity(t)}$$

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



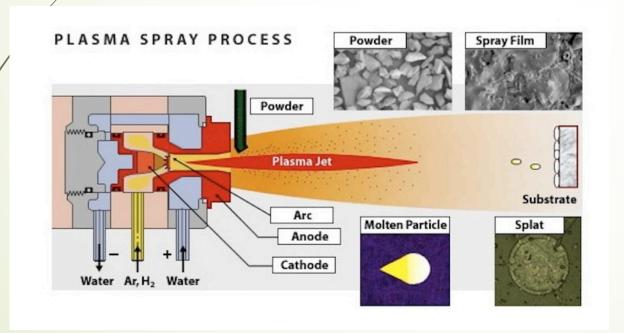


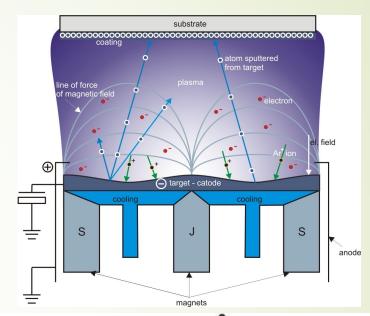


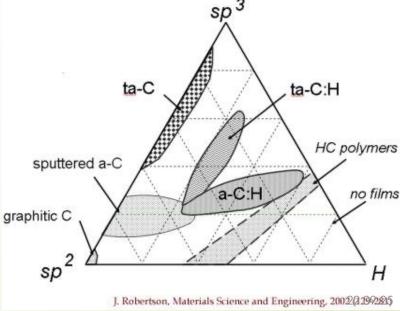
22.09.25

Investigated vacuum coating techniques

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





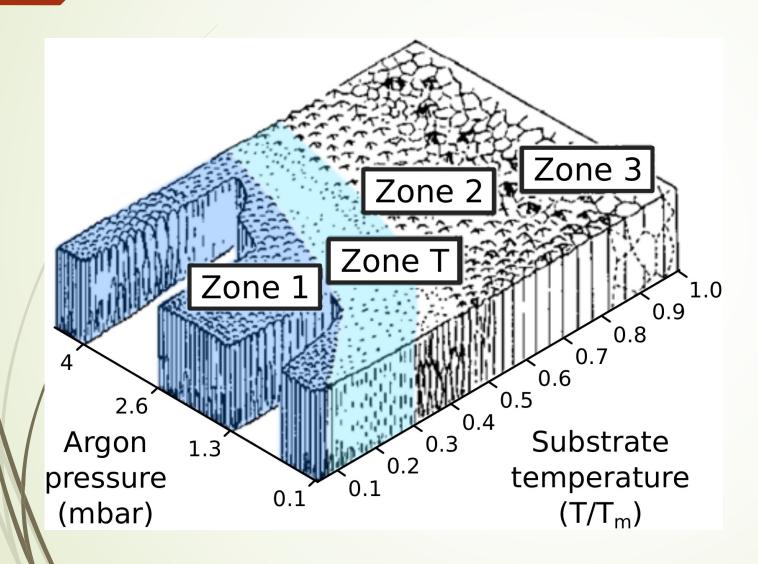


Results: 222Rn emanation reduction factors

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	С-Н	Innovative Coating Solutions	~3	~1,5

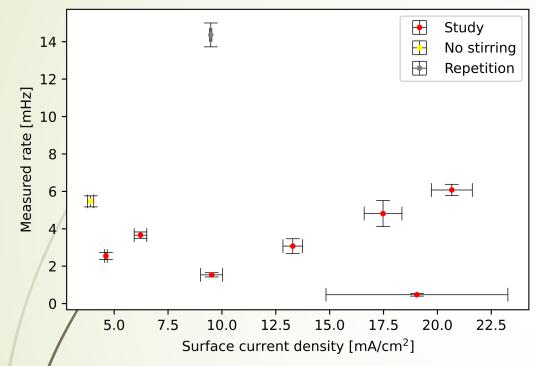
- Little ²²⁰Rn reduction and essentially no ²²²Rn reduction
- Best result for "hot" plasma depsoition, but re-evaporation due to hot substrate

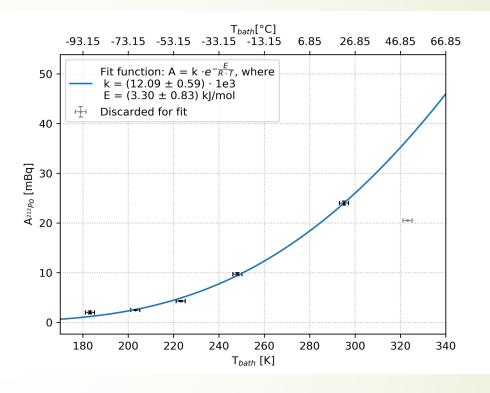
Thornton structure zone model:



- 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?

Optimization of Cu coating procedure and ²²⁰Rn reduction results





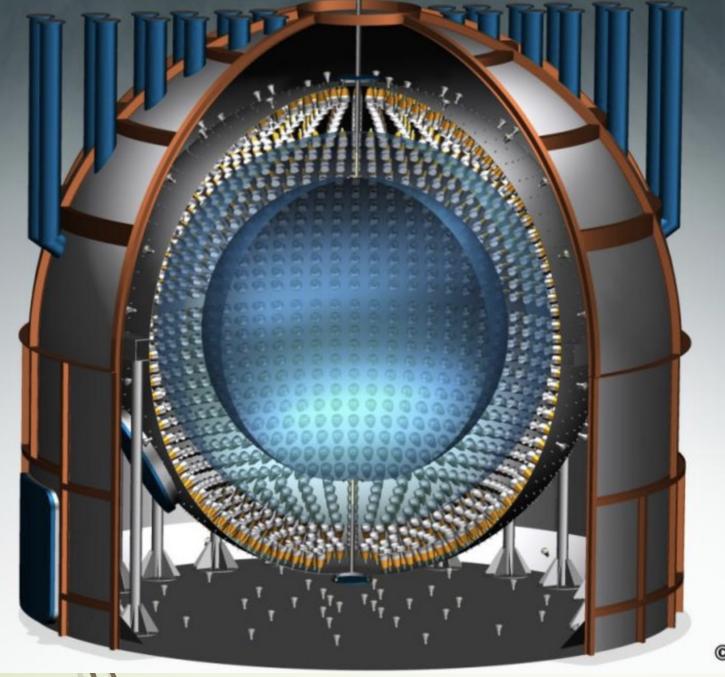


- Optimum surface current density identified.
- Avoid whisker growing by careful parameter control.
- Diffusion-driven emanation confirmed by tests at different temperatures.
- Even hints for slight ²²²Rn reduction.

H. Simgen, CPPM seminar

Radon – Some properties

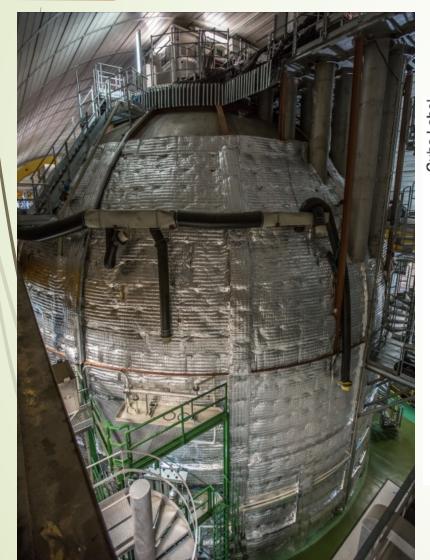
- Noble gas.
- Heaviest existing gas → not so noble.
- No stable isotopes → All radon is radioactive.
- Part of natural (primordial) radioactivity.
- Short-lived: ²²²Rn (3.82 days) by far the longest living, rest much shorter
- Followed by a chain of radioactive heavy metals (a-, β- and γ-emitters)

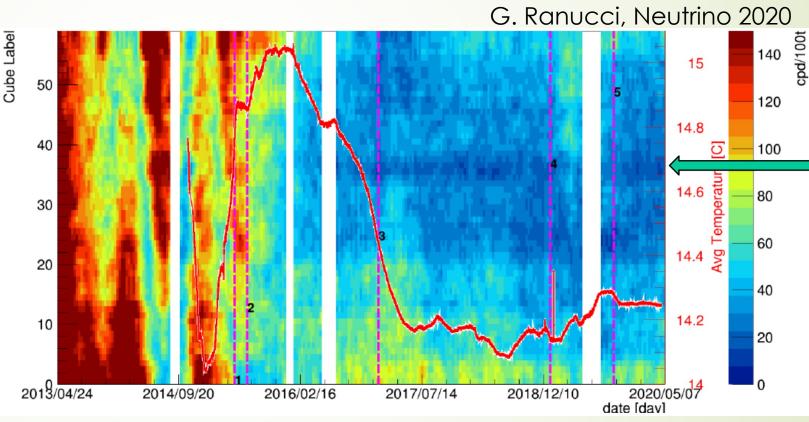


BOREXINO

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

BOREXINO: Thermal stabilization

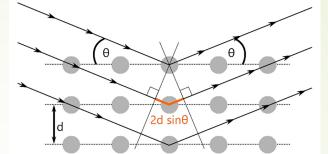




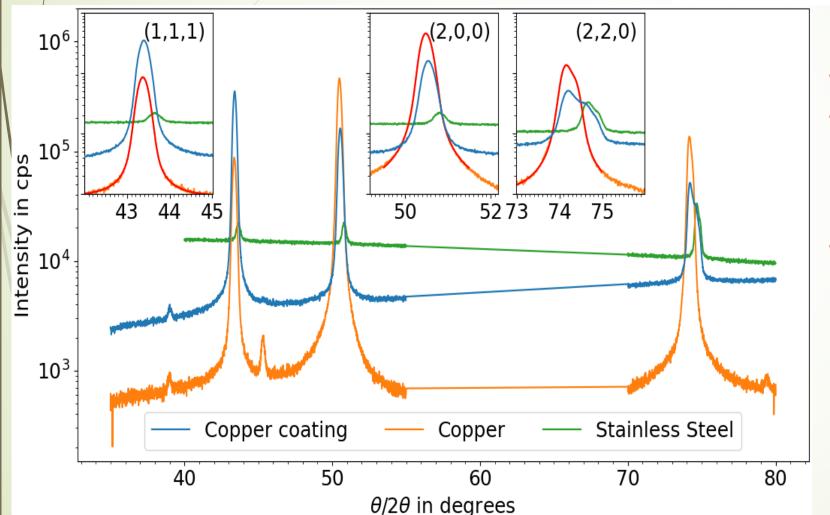
Switch off convection to achieve ultimate purity!

70

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