Application of surface coatings for radon mitigation

Low Radioactivity Techniques - 2019

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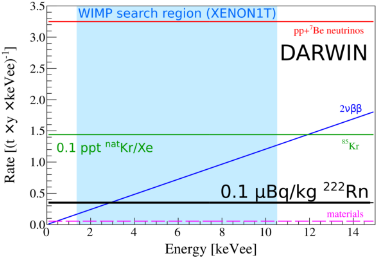
May 22, 2019

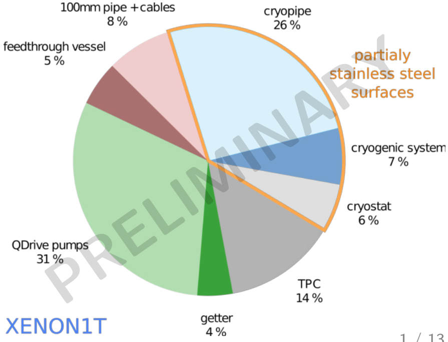
Motivation for radon mitigation



JCAP 10, 016 (2015)

- XENON1T achieved radon budget $10\,\mu\mathrm{Bq/kg}$
- $^{222}\mathrm{Rn}$ is a homogeneous internal background source
- No fiducialization possible, have to tackle it at the source

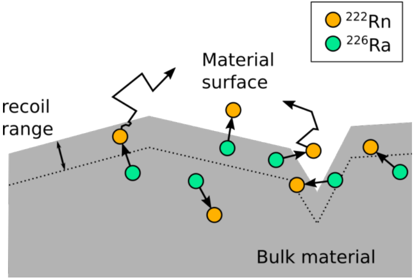
- Mitigation strategies: Screening, Distillation (see H. Simgen & E. Miller), **Surface coating**
- Significant fraction is emanated from **stainless steel surfaces**



Radon mitigation strategy

Emanation by recoil

- $^{226}\mathrm{Ra} \xrightarrow{4.8\ \mathrm{MeV}} {}^{222}\mathrm{Rn}$
- Recoil range of $^{222}\mathrm{Rn}$ in steel $\lesssim$ 20 nm



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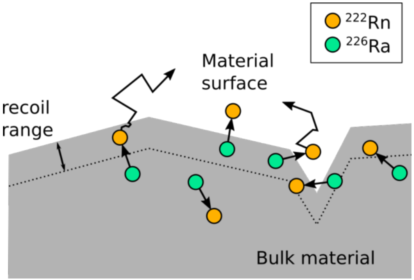
Emanation by diffusion

- Influenced by diffusion constant, temperature and half-life of the radon isotope (i.e. $T_{1/2}^{222Rn} \gg T_{1/2}^{220Rn}$)



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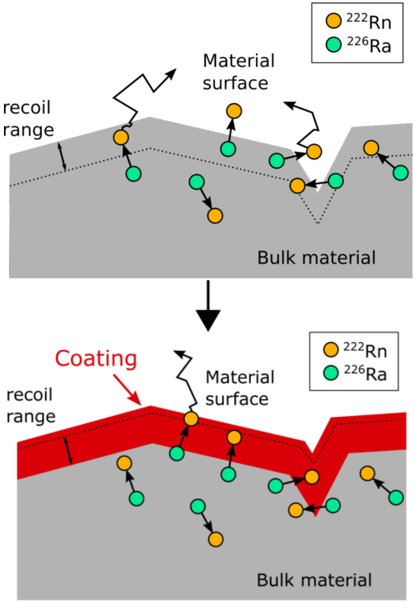
Coating approach

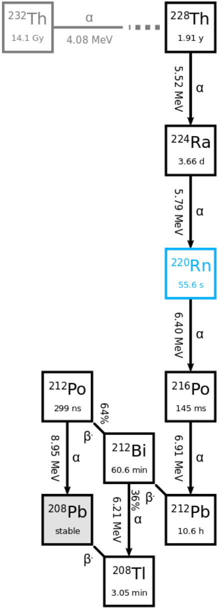
- Block recoil **and** reduce diffusion of radon
- Need for (radio-) pure coating material



Available Samples

Thoriated welding rods

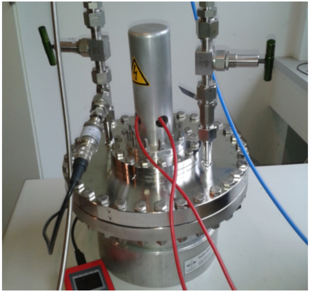
- Tungsten rods containing 4% of natural ThO_2
- \+ Sufficiently active ^{220}Rn source
- \+ Good availability
- \- Metallic surface but not stainless steel
- \- ^{222}Rn emanation only little

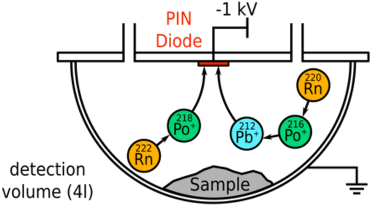


Measurement (Radon Monitor)

- Electrostatic collection of charged radon daughters
- Detection efficiency for $^{222}\mathrm{Rn}$ regularly monitored ($\approx 33\,\%$)
- Measurement of α energy using a (windowless) silicon PIN diode
- Identification of various α lines possible
- Relative measurement before/after coating:

$$R = \frac{A_{before}(Rn)}{A_{after}(Rn)}$$

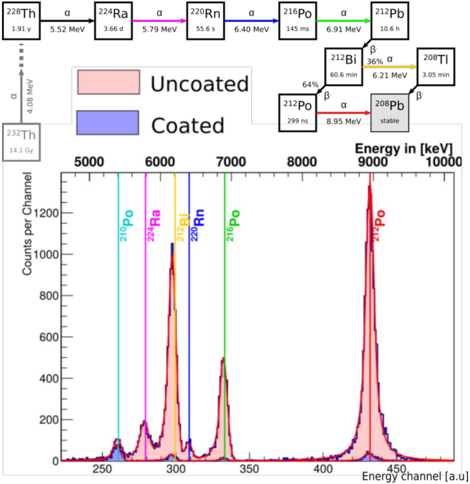




Measurement (Radon Monitor)

Energy spectrum of thoriated welding rods

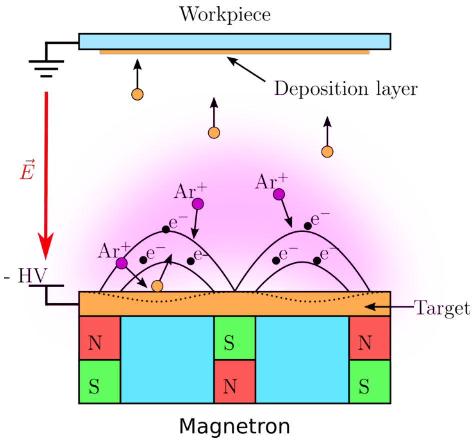
- Coating largely reduces the amount of emanation
- $^{224}\mathrm{Ra}$ is visible only in uncoated spectrum
- Constant $^{210}\mathrm{Po}$ signal from contamination on the PIN diode



→ 10 $\mu\mathrm{m}$ electrodeposited copper layer

Coating techniques I (Sputtering)

- Performed by *EuropCoating* (German company)
- Process in low pressure argon atmosphere
- Deposition of $400 - 800\,\mathrm{nm}$ titanium
- Work piece at room temperature
- Thronton model predicts surface micro-structure

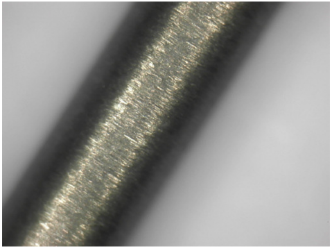


Coating techniques I (Sputtering)

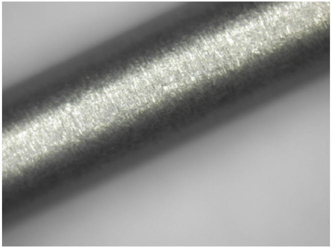
Results

Reduction factors	$^{222}\mathrm{Rn}$	$^{220}\mathrm{Rn}$	$^{224}\mathrm{Ra}$
sample #1	2.2 ± 1.7	4.51 ± 0.07	> 25
sample #2	1.3 ± 0.2	1.49 ± 0.01	3.9 ± 0.6
sample #3	-	1.18 ± 0.01	>50

- Thronton model $\rightarrow$ vertically aligned, porous grains
- Higher workpiece temperature required



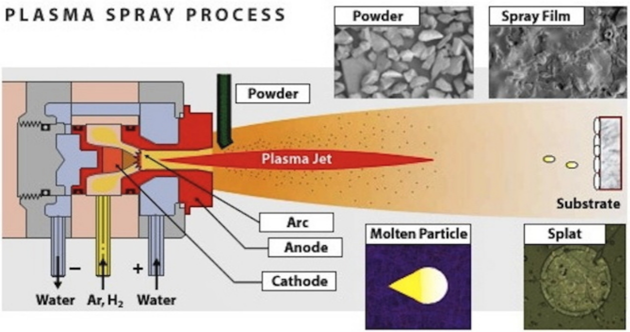
$\rightarrow$ uncoated



$\rightarrow$ coated

Coating techniques II (Plasma deposition)

- In partnership with Dr. Laure Plasmatechnologie (Stuttgart, Germany)
- Coating with $\approx 1\,\mu\mathrm{m}$ of Copper
- Powder condensates on work piece surface



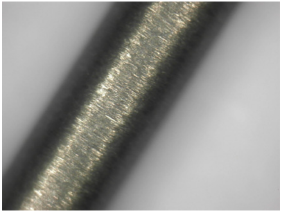
https://www.sciencelearn.org.nz

Coating techniques II (Plasma deposition)

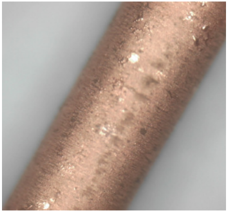
Results

Reduction factors	$^{220}\mathrm{Rn}$
$\varnothing$ 1 mm rod	2.27 $\pm$ 0.02
$\varnothing$ 4.8 mm rod	22.0 $\pm$ 1.8

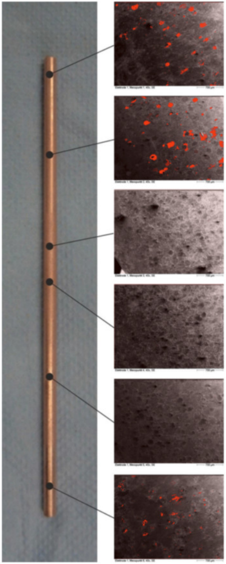
- Complete suppression of $^{224}\mathrm{Ra}$ observed
- Test coatings $\rightarrow$ reproducibility difficult



$\rightarrow$ uncoated



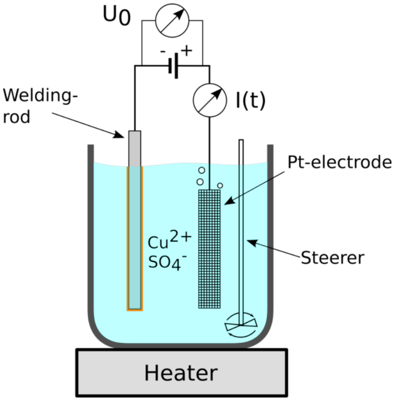
$\rightarrow$ coated



- SEM/BSE analytics

Coating techniques III (Electro-deposition)

Reduction factors	coating thickness	$^{220}\mathrm{Rn}$	$^{222}\mathrm{Rn}$
sample #1	$10\,\mu\mathrm{m}$	107 ± 14	-
sample #2	$5\,\mu\mathrm{m}$	47 ± 4	-
sample #3	$5\,\mu\mathrm{m}$	129 ± 3	7.8 ± 2.6
sample #4	$2\,\mu\mathrm{m}$	12 ± 4	-

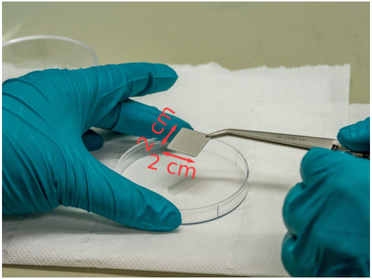


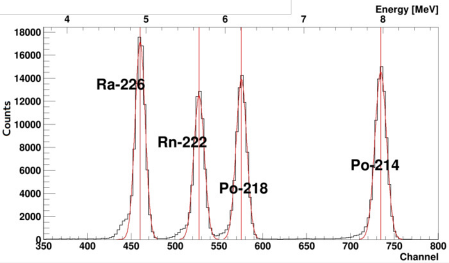
- Tests with electro-deposition of copper have been made at MPIK
- Used electrolyte: $\mathrm{CuSO_4}$ (0.05 mol/l) in $\mathrm{H_2SO_4}$ (1 mol/l)
- Good adhesion achievable by heating bath to 40 °C and steering

What’s next?

Implanted stainless steel

- Stainless steel samples implanted with $^{226}\mathrm{Ra}$ by ISOLDE facility @ CERN
- \+ Sufficiently active source ($\approx 2\,\mathrm{Bq}$ $^{222}\mathrm{Rn}$ emanation)
- \+ Stainless steel surface
- \+ Two samples produced so far
  - Implantation energy: $30\,\mathrm{keV}$ $\rightarrow$ shallow implantation depth $7\,\mathrm{nm}$
- \- LHC & ISOLDE are now in long shutdown (until 2021)





$\rightarrow$ α-spectrum of the sample

What's next?

Explore further techniques

- Multi-layer coatings for enhanced diffusion mitigation (e.g. copper + nickel)
- Polymer coatings (e.g. diamond like carbon)

More systematic studies

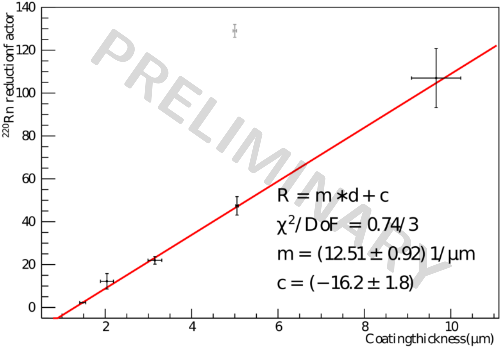
- Determine $^{222}\mathrm{Rn}$ reduction factor (ISOLDE samples)
- Dependence of radon diffusion in coatings at different temperatures
- Temperature stability of coatings
- Mechanical stability of coatings (adhesion, hardness, ...)
- Compatibility of coating with LXe purity (see G. Eurin's talk)

Summary

- Radon mitigation will be an important aspect of several future rare-event searches.
- Application of surface coatings has proven to be an effective method against radon emanation
- Radon reduction factors of the order of 100 have been achieved

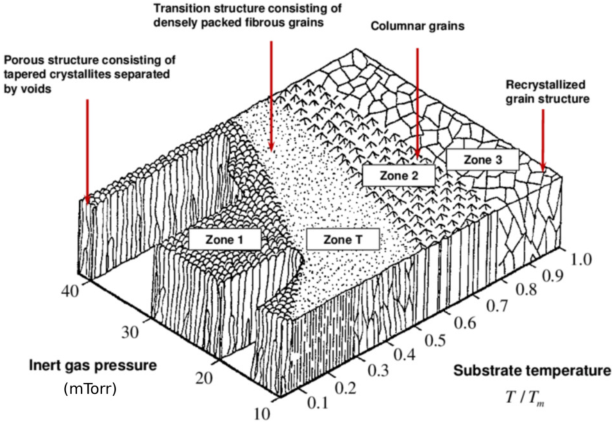
More results are on the way
Stay tuned!

Dependence of $^{220}\mathrm{Rn}$ reduction factor (layer thickness)



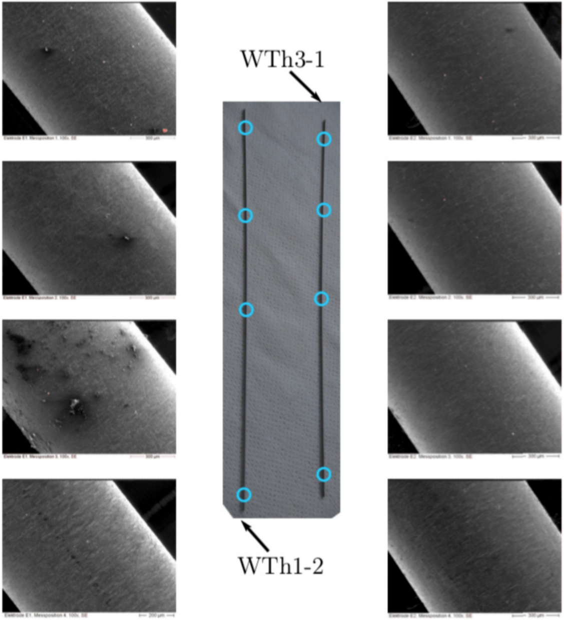
- Combining all copper coatings (electro- and plasma)
- Linear dependence of reduction factor with coating thickness
- Behavior still needs to be explained by a model (diffusion)
- Outlier @ 5 $\mu\mathrm{m}$ might be due to different detection efficiency

Thronton model (sputtering)



- Low radon reduction achieved, despite high $^{224}\mathrm{Ra}$ suppression
- Recoil emanation well controlled but insufficient diffusion tightness
- Thronton model zone 1 $\rightarrow$ probably not ideal

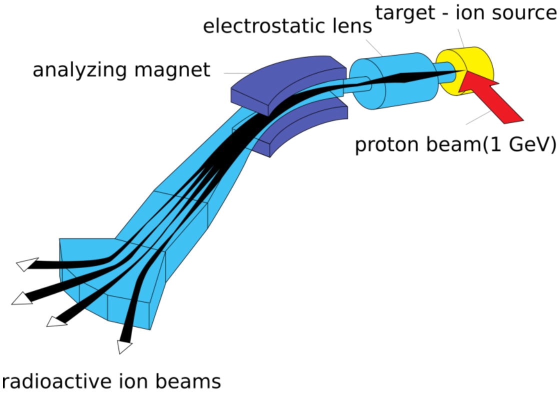
Sputtering investigation



- Sample #2 vs. Sample #1
- Investigation with SEM/BSE $\rightarrow$ no significant defects
- Large difference in $^{224}\mathrm{Ra}$ suppression still unclear

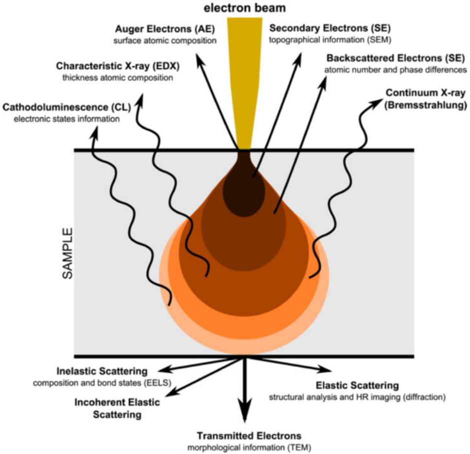
Ion Separation Online Device (ISOLDE) @ CERN

- Protons from pre accelerator of LHC @ $1.4\,\mathrm{GeV}$
- Irradiation of uranium-carbide target
  $\rightarrow$ production of various isotopes
- Separation of desired isotope by el/mag filter



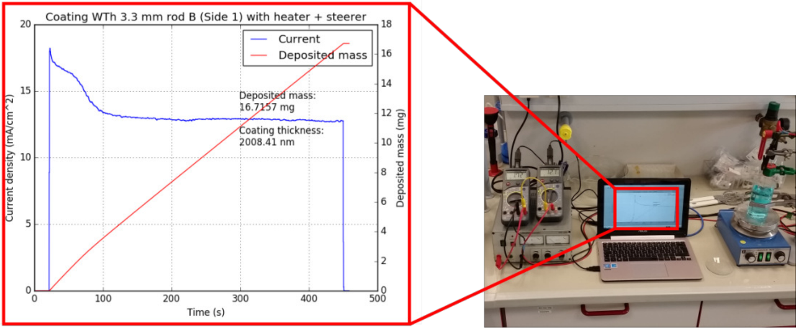
Back Scatter Electron (BSE) analysis

- Integrated detector in some **S**canning **E**lectron **M**icroscopes
- Signal strength depends on Z^2 of the material ($Z_{Cu} = 29, Z_W = 74$)
- High Z material will "shine" through the coating up to depths of about $1\,\mu\mathrm{m}$

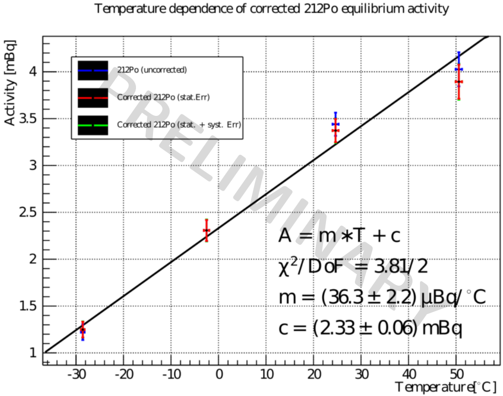


Electro-deposition setup

- Current is constantly read out and digitized
- Time integration of current → total deposited mass
- Allows for an on-line estimation of the layer (average) thickness

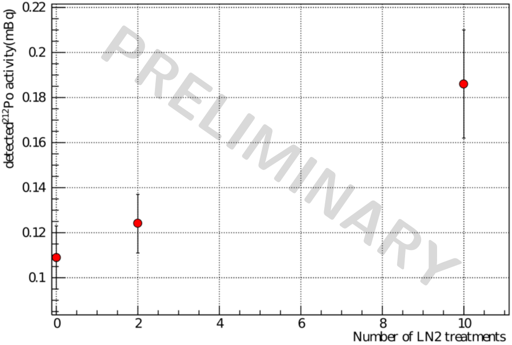


Dependence of $^{220}\mathrm{Rn}$ reduction factor (temperature)



- Measurment of a sputtered sample (titanium)
- Thoriated tungsten welding rod as base material

Dependence of $^{220}\mathrm{Rn}$ reduction factor (LN2 treatment)



- Electrodeposited copper (10 $\mu\mathrm{m}$)
- Thoriated tungsten welding rod ($\varnothing$ 4.8 mm) as base material
- Repeated temperature shock by direct submersion of the sample in liquid nitrogen (LN2)