EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Production of ^{226}Ra-implanted high-quality radon sources for detector characterization

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Abstract:

Reliable radon sources are a key ingredient for calibration and development of detectors for the radioactive noble gas ^{222}Rn. A novel approach to produce such sources is by the implantation of ^{226}Ra into a metallic carrier substrate. In a proof-of-principle study it was shown that a shallow implantation depth of about 8 nanometers allows to produce a recoil dominated ^{222}Rn source. In contrast to a diffusion driven radon release, this process is largely independent of environmental parameters such as humidity, temperature and pressure. Therefore, such sources feature an excellent stability of their radon emanation rate.

Due to a large interest in our community for such sources, we propose the production of twenty further samples. Given the long half-life of ^{226}Ra, these implantations can be carried out at times where no proton beam is available. Obtaining these samples will enable the development of future radon detectors and investigations of coating based radon barriers. Both will be necessary for future liquid xenon-based experiments used for direct dark matter detection. Furthermore, these sources can be applied for the calibration of large-scale liquid xenon detectors.

Requested shifts: 12 shifts (without protons) split into up to 2 runs over $\sim$ 6 months

1 Introduction and motivation

The naturally occurring, radioactive noble gas ^{222}Rn is part of the primordial uranium decay chain and formed in the alpha decay of ^{226}Ra. Its measurement has many applications ranging from radioprotection [1] over modeling of green house gas emissions [2] to prediction of seismic events [3]. But also in experiments searching for extremely rare processes, such as the direct detection of dark matter [4, 5] or the neutrinoless double beta decay [6], the radon-induced background plays an important role.

The calibration of radon detectors, requires radon sources featuring a well known radon release rate. Their radon emanation should not depend on any environmental factors, such as temperature or pressure, nor should they out-gas any unwanted impurities. Most commercial radon sources, however, are either liquid radium solutions or porous materials from which ^{222}Rn is released via diffusion. Both have significant shortcomings with regard to their purity and stability, opening the need for new kinds of radon sources. In a proof-of-principle study conducted in 2017, two radon sources have been successfully produced by the implantation of ^{226}Ra into stainless steel at the ISOLDE facility (see Fig. 1). Their thorough characterization [7] showed that this novel approach allows to produce radon standards with a very high quality. Another recent study carried out independently at the RISIKO facility in Mainz, Germany [8], supports this conclusion.

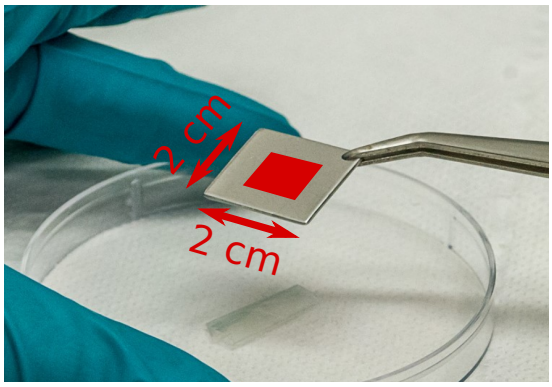

Figure 1: Photograph of the stainless steel sample. The approximate extend of the ^{226}Ra implantation is marked in red.

The two obtained sources have been successfully applied for several studies such as for the characterization of alpha spectrometers [9, 10], determination of the detection efficiency of novel radon detectors [11] as well as for studies on a novel radon mitigation technique using metallic surface coatings [12]. One source is considered to be used for the calibration of the radon-induced background in the XENONnT [13] dark matter detector.

To meet the existing demand and interest in these versatile radon sources, we propose to implant twenty further samples with ^{226}Ra. The substrates to be implanted will be mostly stainless steel, but also other metal plates (copper and titanium). Optionally, we are interested to implant a few insulator samples (PTFE and quartz glass) or semiconductors (silicon and germanium). The exact list of samples will be fixed as soon as the feasibility is clarified with the ISOLDE experts. Section 2 of this proposal summarizes the main results and experiences gained in the proof-of-principle study. Based on these findings, a detailed beam-time estimate for the proposed production is then given in section 3.

2 Production of ^{226}Ra-implanted samples at ISOLDE

The long half-life of ^{226}Ra of approximately 1600 years allows to perform the implantation without the need for simultaneous proton irradiation of the target (i.e. *off-line*). In the 2017 proof-of-principle study, ^{226}Ra ions were released from an uranium carbide target (UC611), which had been irradiated by 1.19×10^{18} protons during previous usage throughout almost 7 days [14]. Subsequently, ionization was achieved by surface ionization, yielding a measured beam current of ^{226}Ra ions of approximately 3 pA. The beam energy was set to 30 keV and mass number 226 has been selected using the general purpose separator (GPS). A total amount of 1×10^{12} radium ions have been collected into two stainless steel samples with a size of 2 cm×2 cm (see Fig. 1) during a net time of about 8 hours. During the implantation, the ion beam was swept such that a square area of 1 cm×1 cm across each sample's surface was covered.

The number of implanted radium ions $N_{226\mathrm{Ra}}$ can be directly converted into the sample's radium activity $A_{226\mathrm{Ra}}$ via

$$A_{226\mathrm{Ra}} = \lambda_{226\mathrm{Ra}} \cdot N_{226\mathrm{Ra}} \,, \tag{1}$$

where $\lambda_{226\mathrm{Ra}}$ is the decay constant of ^{226}Ra. From this, an implanted activity of about 7 Bq per sample is expected. After the sample arrived back to Heidelberg, the activity in each sample was confirmed using HPGe and alpha spectrometry. Furthermore, their radon emanation rates were determined using miniaturized proportional counters [15, 16]. The results along with their uncertainties are summarized in Table 1, while more details on the applied measurement techniques and analyses can be found in [7, 12].

Table 1: Comparison of the results from HPGe spectrometry, alpha spectrometry and radon emanation of the two implanted stainless steel samples.

Measurement	Result (Bq)	
	Sample a	*Sample b*
Implantation	about 7	about 7
γ-spectrometry	$7.4 \pm 0.1_{\,\mathrm{stat}} \pm 0.9_{\,\mathrm{syst}}$	$8.4 \pm 0.3_{\,\mathrm{stat}} \pm 1.0_{\,\mathrm{syst}}$
α-spectrometry	$8.70 \pm 0.06_{\,\mathrm{stat}}\,{}^{+2.0}_{-1.8\,\mathrm{syst}}$	$9.13 \pm 0.10_{\,\mathrm{stat}}\,{}^{+0.7}_{-0.4\,\mathrm{syst}}$
^{222}Rn emanation	$2.07 \pm 0.03_{\,\mathrm{stat}} \pm 0.04_{\,\mathrm{syst}}$	$2.00 \pm 0.03_{\,\mathrm{stat}} \pm 0.04_{\,\mathrm{syst}}$

The implantation distribution of ^{226}Ra ions as a function of depth has been simulated using the SRIM code [17] and is shown in the left of Figure 2. From this, we expect that an ion beam energy of 30 keV yields an average implantation depth of 7.9 nm with a spread of 2.3 nm [18]. Using this distribution as an input, the radon emanation fraction f_{em} can be modeled. The right panel of Figure 2 shows how this fraction depends on the incident ion beam energy (for more details, see [12]). An excellent agreement is found between this prediction and the observed emanation fractions of both samples, which suggests that ^{222}Rn is predominantly released via recoil. This is further supported by the absence of a

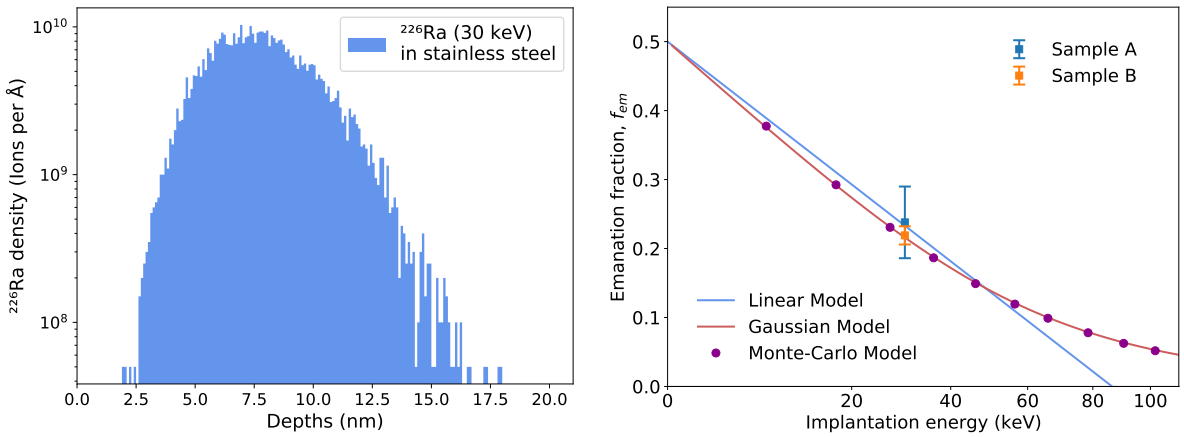

Figure 2: **Left:** Expected distributions of the implanted ^{226}Ra ions as a function of the depth below the samples surface. Simulation performed using the SRIM code [17]. **Right:** Dependence of the expected radon emanation fraction due to recoil as a function of the ratio between the mean implantation depths and the average ^{222}Rn recoil range. The obtained fraction of both samples closely matches this expectation.

significant temperature variation of the radon emanation rate from ambient temperature down to -30°C [7].

2.1 Short-lived contamination

Shortly after the implantation, additional activity from several short-lived isotopes was observed on the sample. Both samples showed a significant activity originating from the side bands of mass number 226 in the isotopes ^{225}Ra as well as ^{227}Th. This was determined using an alpha spectrometric measurement (see [7, 12] for details). Since both isotopes have half-lifes of 15 days and 19 days respectively, they decayed away rapidly and did not affect the usability of the sources. Additionally, a trace contamination of ^{139}Ce ($T_{1/2}$ = 138 days) was found on one of the two samples by HPGe spectrometry. While the presence of ^{225}Ra and ^{227}Th may be expected due to imperfect mass separation, the ^{139}Ce contamination was completely unexpected and hints towards a contamination during handling, which should be avoided. Fortunately, its activity decayed away sufficiently fast and left no persisting impact on the usability of the sample. Still, the total activity from short-lived contaminants might require an additional *cool-down* time of about 1 week before the samples can be removed from the implantation chamber as well as an additional 4 weeks prior to their shipment.

3 Beam time estimate for a further implantation

To estimate the implantation duration t for each sample, the required number of ^{226}Ra ions N_{ions} needs to be calculated. This follows directly from the desired radon emanation rate $R_{222\text{Rn}}$, scaled by the achieved radon emanation fraction f_{em} and the radium decay

constant $\lambda_{226\mathrm{Ra}}$ (see also equation 1)

$$t = \frac{e}{I_{\mathrm{beam}}} \cdot \frac{R_{222\mathrm{Rn}}}{f_{\mathrm{em}} \cdot \lambda_{226\mathrm{Ra}}}, \tag{2}$$

where I_{beam} corresponds to the average ion beam current and e is the elementary charge.

From the experience gained in the proof-of-principle study, we can expect an ion beam current of $I_{\mathrm{beam}} = 3\,\mathrm{pA}$ and an emanation fraction of $f_{\mathrm{em}} = 25\%$ at an ion beam energy of 30 keV (see Figure 2). The implantation of 20 samples with a radon emanation rate of $R_{222\mathrm{Rn}} = 1\,\mathrm{Bq}$, will then require a **total beam time of $\mathbf{T_{beam}} = 86$ hours**. Where each sample needs to receive roughly $N_{\mathrm{ions}} = 3 \times 10^{11}$ ^{226}Ra ions.

4 Summary

Table 2: Summary of beam and source parameters for the proposed implantation

Required Isotope	Target	Ion source	Beam current	Number of samples	Ions per sample	Requested shifts	Protons required
^{226}Ra	UC_x	Surface	3 pA	20	3×10^{11}	12	No

Based on the successful proof-of-principle study carried out in 2017, and the favorable properties of the obtained ^{222}Rn sources, we propose the production of 20 further ^{226}Ra implanted samples. Assuming that a similar beam current of 3 pA can be obtained, this should be possible within a total of 86 hours of beam time. It should be noted, that this implantation can be carried out without the need for simultaneous proton irradiation of the target (i.e. *off-line*). Table 2 summarizes the key parameters for the proposed project.

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DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
SSP-GLM chamber	☒ To be used without any modification ☐ To be modified
Different substrates for the ^{226}Ra implantation: Stainless steel and other metallic (titanium, copper) and possibly non-metallic samples (PTFE, quartz glass, silicon and germanium)	☒ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT
Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	

	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			