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Observation and modeling of ^{222}Rn daughters in liquid nitrogen

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ABSTRACT

The results of alpha spectrometric measurements of the activity of ^{222}Rn daughters dissolved in liquefied nitrogen are presented. A direct detection method of ionized alpha-emitters from the ^{222}Rn decay chain (^{214}Po and ^{218}Po) in a cryogenic liquid in the presence of an external electric field is shown. Properties of the radioactive ions are derived from a proposed model of ion production and transport in the cryogenic liquid. Ionic life-time of the ions was found to be on the order of 10 s in liquid nitrogen (4.0 purity class). The presence of positive and negative ions was observed.

1. Introduction

Ultra-pure liquefied gases can serve as a passive shielding material and a cooling medium for the low background experiments searching for rare events. Additionally, scintillation light induced by internal radioactive decays or external radiation sources may be used as a 4-π anti-coincidence, further reducing the background. Although the liquid is highly purified it may still contain traces of intrinsic radioactive contamination. The surrounding material of the supporting structures and the cryostat container also present a source of internal background.

The natural presence of ^{226}Ra in all construction materials is a constant source of alpha emitters, belonging to the ^{222}Rn decay chain (^{214}Po and ^{218}Po) (Fig. 1). ^{222}Rn is a noble gas able to diffuse from the bulk to the surface of porous materials. It is also emitted from surfaces contaminated with ^{226}Ra. Consequently ^{222}Rn dissolves in cryogenic liquids, entering the surrounding active volume of a detector. Ions produced in the decays of ^{222}Rn and its daughters may reach vicinity of e.g. the germanium detectors by means of convection flows or electrostatic drifting of the radioactive ions. Such processes present a constant in time source of radioactive background. Thus, ^{222}Rn contamination results in a severe background (caused by energetic alpha- and beta-decays from the ^{222}Rn decay chain) which is difficult to control. Inve stigation of the behaviour of ^{222}Rn and its progeny in cryogenic liquids is thus of great importance for ultra-low background experiments using liquefied gas as shielding, a target or a detecting medium (e.g. two phase Time Projection Chamber in the DarkSide experiment [1], liquid argon as a passive shielding material and a cooling medium for the germanium detectors in the GERDA [2] experiment).

2. Detection of ^{214}Po and ^{218}Po in liquid nitrogen

2.1. Experimental setup

The experimental setup is presented in Fig. 2. A bare Si-PIN diode (Hamamatsu model S1223-01 with removed glass window) is immersed in liquid nitrogen under normal pressure, contained in a 32 L dewar. The diode is wired using a standard 50 Ω coaxial cable. The housing of the diode is grounded and connected to the signal cable shield. The diode is operated under a nominal voltage of +28 V (relative to the ground), specified for the best energy resolution for registering 5 MeV α-particles in air (5 mm distance). The chosen diode type operates stably in the cryoliquid for long periods of time, extending over two weeks. The diode is connected to a standard spectrometry electronics chain – charge preamplifier and the active filter amplifier. The readout system consists of a multi channel analyzer, recording alpha-energy spectrum data in fixed time windows (series of measurements). Changes in the dewar gross weight (32 L of total volume) are registered on-line to calculate losses of the cryoliquid due to evaporation (boil-off).

The dewar is electrically isolated from the ground. The metal housing of the dewar is wired to a bipolar high voltage (HV) power supply. Applying positive bias to the housing has the same effect as biasing the diode with negative high voltage of the same magnitude. In this biasing scheme the Si-PIN diode, having lower potential than the dewar, attracts cations. Reversely, negative bias

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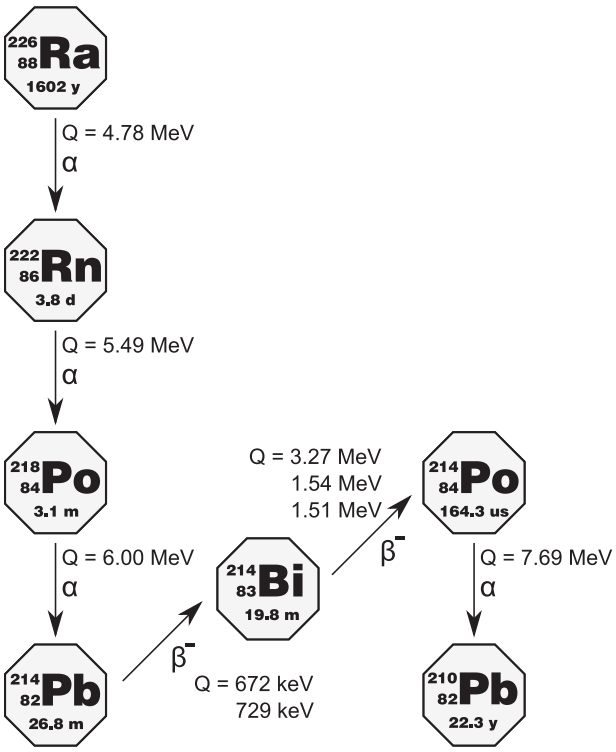

Fig. 1. ^{222}Rn decay chain.

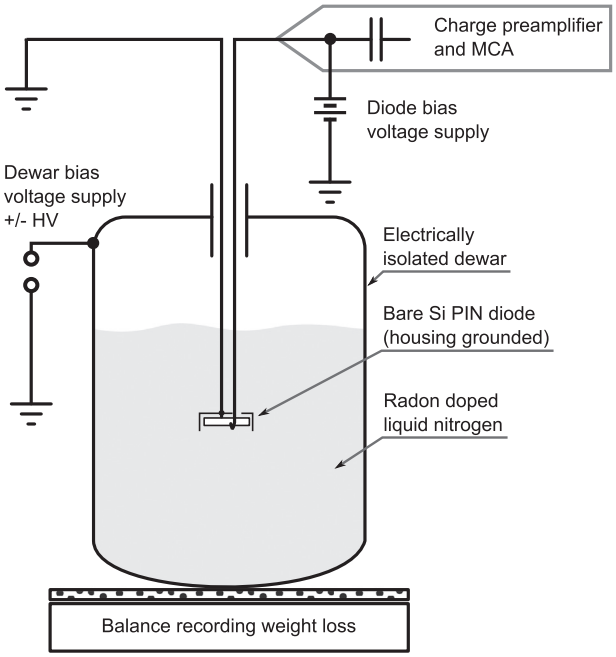

Fig. 2. Sketch of the measurement setup (not to scale). The Si-PIN diode is operated under a nominal voltage of +28 V relative to the ground. The dewar's metal housing is usually biased to ±2 kV. If the housing is biased to +2 kV, then the configuration is equivalent to approximately −2 kV applied to the diode housing and dewar grounded.

on the dewar puts the diode at a higher potential. Positive ions are then repelled from the diode.

2.2. ^{222}Rn doping procedure

An amount of radon activity on the order of 15 kBq from a radon generator is dissolved in liquid nitrogen (purity class 4.0) at the beginning of each measurement cycle (Fig. 3). The dissolved activity

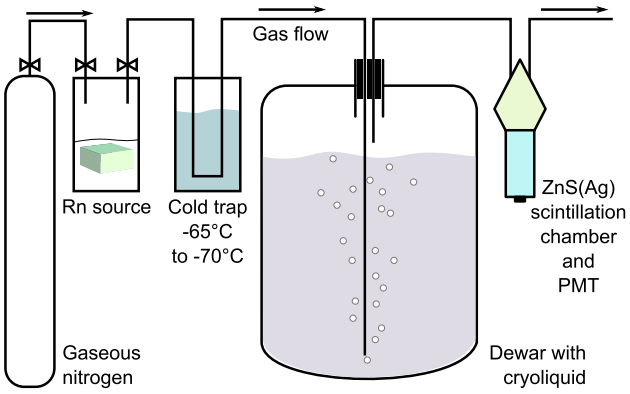

Fig. 3. Setup for dissolving gaseous ^{222}Rn in the liquid nitrogen (not to scale). Radon from the source, together with the gas carrier, is transported through a cold trap to reduce humidity content. Some of the ^{222}Rn atoms may freeze out on the cold parts of the piping operated in the cryogenic liquid. Consequently, ^{222}Rn dissolves with less than 100% efficiency.

ranges from roughly 10 to 15 kBq. The radon source consists of 0.1 L of ^{226}Ra solution (18.5 kBq of activity) enclosed in a tight silicon pipe and placed in a 4.6 L metal vessel. More than 90 % of the ^{222}Rn generated in the solution diffuses through the pipe walls into the vessel volume. The volume is then flushed with a carrier gas (nitrogen). The carrier gas enriched in radon is transported through a cold trap (temperature ranges from −65 to −70 °C) to remove traces of humidity. The thin gas outlet is submerged in liquid nitrogen. Some fraction of radon atoms may freeze out on the cold surfaces of the piping before entering the volume of the liquid. Bubbles of the carrier gas ascend to the surface partially dissolving in the cryogenic liquid. Radon activity enclosed in the bubbles is then entirely transferred to the cryoliquid. The exhaust gas is monitored for the residual radon activity using a ZnS(Ag) scintillation chamber. The whole procedure lasts for about 30 min, and uses a carrier flow of about 0.5 L/min to fully flush the radon source volume.

2.3. ^{214}Po and ^{218}Po alpha activity measurements

The first measurement is taken at least 3 h after the dissolving procedure, to achieve decay (secular) equilibrium in the volume of the cryoliquid between ^{222}Rn and its daughters. The recorded signal of ^{214}Po and ^{218}Po α-decays is corrected for ^{222}Rn life-time and liquid losses in off-line analysis. Background alpha-energy spectrum recorded by the Si-PIN diode immersed in the liquid nitrogen (no ^{222}Rn dissolved) is shown in Fig. 4. For comparison, a typical spectrum recorded in a ^{222}Rn doped nitrogen is shown in Fig. 5. The dewar was biased to +2 kV during these two measurements. Fig. 6 presents the energy spectrum recorded with −2 kV bias applied to the dewar. The shape of the α-peaks indicates that the drifted ions decay very close or stick to the active surface of the diode (the distance is much shorter than the range of alphas from ^{214}Po and ^{218}Po decays in liquid nitrogen).

Each time, the α-energy spectrum was recorded in 900 s real time windows, and stored as a multi-channel histograms. The ^{214}Po energy peak around Q=7.69 MeV is selected from the spectrum (channels 5200–7000) and the gross count rate in the respective window is corrected for ^{222}Rn decay and liquid boil-off. The count rate is then analyzed as a function of the time elapsed since the ^{222}Rn doping and changes of the bias voltages applied to the dewar. The sign of the analyzed ions was selected by the bias voltage polarity. Distance between the Si-PIN diode and the bottom of the tank may be changed to limit the collection volume of the ions.

Analysis of the ^{218}Po signal is rather difficult due to the background generated by ^{214}Po (cf. Fig. 5) in the energy range of the ^{218}Po alpha peak. The analysis is based on simultaneous fitting