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Short-lived Rn-222 daughters in cryogenic liquids

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Abstract. In this paper a detection method of $\alpha$ emitters from $^{222}\text{Rn}$ decay chain, present in cryogenic liquids, using bare Si-PIN diodes immersed in the liquids is presented. Properties of ionized $^{222}\text{Rn}$ daughters deduced from conducted measurements are outlined. Life-time of positive ions was found to be of the order of 10 s, and nonzero content of electronegative ions was observed.

Keywords: Alpha spectroscopy, Cryogenic liquids, Screening, Low-background measurements

PACS: 23.60.+e, 29.30.Ep, 29.30.Kv

INTRODUCTION

Cryogenic liquids are extensively used in ultra-low background experiments. The liquids serve as a passive shielding and a coolant (e.g. the GERDA experiment [1]) or as a target and detecting medium (two phase TPC in the DarkSide experiment [2]). The ultra-low background experiments are designed to achieve both the lowest possible background levels and to maximize their sensitivity to registering rare events. Therefore reduction of the intrinsic background is essential and the content of radioactive impurities in the cryogenic liquids being in use ought to be investigated. Techniques such as material selection, purification and cleaning, signal analysis and cryogenics can be employed.

$^{222}\text{Rn}$ belongs to the $^{238}\text{U}$ decay chain present in all materials used for construction of the experiments. As an inert noble gas, radon is able to diffuse to the surface of some materials and enter surrounding active volumes of the detectors. This process presents a constant source of radioactive contamination, caused by energetic alpha- and beta-decays of $^{222}\text{Rn}$ daughters with accompanying gamma radiation.

Radon daughters may form positive ions as a result of $\alpha$ and $\beta$ decays in the cryogenic liquids. Electrostatic forces present in some setups (biased bare germanium diodes in the Gerda experiment or TPC in the DarkSide experiment) are able to drift the ions. As a result concentration of the radioactive ions in active volumes of the detectors builds up.

Similar mechanism of ion drifting may be a source of increased background for other isotopes present in liquid gases, like presence of $^{42}\text{K}$, a daughter of $^{42}\text{Ar}$, in natural argon.

IONS IN CRYOLIQUIDS

To better understand the behaviour of radioactive ions in cryogenic liquids we discuss here processes responsible for formation of the ions and the electron-cation pairs after $\alpha$- and $\beta$-decays.

Ionization

Radioactive decays in liquefied gases produce electron-ion pairs along the track of an alpha or beta particle. Loss of the released kinetic energy $Q$ is dominated by electronic interactions and collisions in traversed matter. The deposited energy $Q$ is therefore expended in the production of electron-ion pairs $N_i$, excitation of atoms and sub-excitation electrons, and directly in atomic motion. The average energy required to produce one electron-ion pair $W$ is defined as $W \approx Q/N_i$. E.g. the value of $W$ for liquid argon is $23.6\pm0.3$ eV, small compared to the typical $Q$ value (several MeV for $\alpha$-decays).

Atoms recoiled in alpha decays also become charged after the decay. Recoil energy of the ion (typically 100 keV per recoiled atom) is dissipated in subsequent slowing down collisions with the surrounding matter, creating electron-ion pairs (with pair production energy $W$). Initially negatively charged recoil atom (due to emission of an alpha particle), easily strips off its valence electrons in interactions with the surrounding matter. In consequence the atom becomes...
positively charged. If such created cation is multiply charged, then the process of recombination described later leads to the singularly charged cation. Likewise in the case of $\beta^-$-decay, in which the recoil energy is much lower, the atom is already initially positively charged and no electron stripping occurs.

**Recombination**

The cations (recoil ions and ions of the medium) eventually recombine with the freed electrons. The *initial* recombination (also called germinate recombination) is important for understanding the differences in amounts of ions produced in $\alpha$ and $\beta$-decays. The *bulk* recombination is related to the nonzero impurity content of the cryoliquid.

An electron freed from an atom in ionization process may neutralize the cation of its origin. High ionization density of $\alpha$-decays leads to more efficient initial recombination. Electron-cation pairs produced in $\beta$-decays are less prone to initial recombination due to lower ionization density.

Presence of an external electric field reduces initial neutralization. Mobility of free electrons $\mu_\text{e}$ as a response of a charged particle to the pulling electric field in dense medium, is high (e.g. $\mu_\text{e} = 1.5 \times 10^2 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) compared to the mobility of heavier ions (e.g. $\mu_\text{ion} = 2.4 \times 10^{-4} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for $^{226}\text{Th}$ in liquid xenon [4]). Consequently, electric field rapidly separates the electrons from the almost immobile ions. Effective charge dissociation depends upon the field strength, and facilitates preservation of the cations during the initial recombination.

Model proposed by Thomas and Imel [5] describes the probability of survival of the charge $C_0$ (electron-cation pairs), depending on the type of the energy deposition processes leading to electron-ion pair formation ($\alpha$- or $\beta$-decay) and on the external electric field:

$$C = C_0 \frac{\ln(1 + \xi)}{\xi}, \quad \xi = \frac{N_t \alpha}{4a^2 \mu_\text{e} E}$$

where $N_t$ is, as previously, the number of electron-cation pairs produced within a box of dimensions $a$, $\alpha$ the recombination coefficient, $\mu_\text{e}$, electron mobility, and $E$ is the electric field strength. The value of $\xi$ is usually treated as a free parameter and fitted to the data, e.g. the result of the fit provides $\xi_\alpha E = 840 \text{ V/cm}$ for 364 keV electrons, and $\xi_\alpha E = 470 \text{ kV/cm}$ for 5.64 MeV $\alpha$-decays [5]. The significant difference in the $\xi$ values for $\alpha$ and $\beta$ particles originates from the different ionization densities caused by $\alpha$ and $\beta$ radiation (through $N_t$ and $a$).

E.g. the probability of cation survival $\xi_C = C/C_0$ during the germinate neutralization in the electric field $E = 30 \text{ V/mm}$, produced by $\beta$-particle is $\xi_{C\beta} = 48\%$, whereas in the case of $\alpha$ radiation the probability is $\xi_{C\alpha} = 0.47\%$, over 100 times lower.

The cations may also be directly neutralized by the free electrons or electronegative impurities. Typical impurity content of technical grade liquid nitrogen or argon is at the level of ppm (parts per million) or more.

The first ionizing potential (IP) is the energy required to remove a bound shell electron from a neutral atom. The second ionizing potential is a potential associated with the removal of a second electron from a singly ionized atom. The first ionizing potentials of $^{222}\text{Rn}$ daughters range from 11 eV for $\text{Rn}$ to 7.3 eV for $\text{Bi}$. The second IPs are order of magnitude higher ($\sim 149\text{ eV}$ for $\text{Bi}$). The cations may also be directly neutralized by the free electrons or electronegative impurities. Typical impurity content of technical grade liquid nitrogen or argon is at the level of ppm (parts per million) or more.

The differences in the first IPs of recoil ions and gap energies of the medium atoms exceeding 2 eV allow electron exchange process between the neutral recoil atom (possibly neutralized right after the decay) and an ion of the medium. Therefore the recoil atom may be recharged in certain circumstances, and may exhibit extended ionic life-time. Higher values of the second IPs of the elements disallow multiple ionization ($Y^{++}$, $Y^{+++}$ etc.) in the electron exchange process, favoring singular ionization.

**Ion transport**

The cation of interest can survive long enough to be transported by the electric forces with characteristic mobility $\mu_{\text{ion}}$. Velocity of the ion is proportional to the applied electric field [cite]:

$$\vec{v}(r) = \mu_{\text{ion}} \vec{E}(r)$$

Range of the ion depends on the ionic life-time $\tau$ or nuclear life-time, whichever is significantly shorter, and on external electric field $E$, i.e. its strength and shape. The cations are drifted along the electric field lines, from higher to lower potential. Daughter ions forming chemical compounds or complexes may also carry negative electric charge.
FIGURE 1. Sketch of the measurement setup for $^{222}$Rn doped liquid nitrogen (not to scale). Si-PIN diode is operated under nominal voltage of $+28$ V relative to the ground. The dewar metal housing is usually biased to $\pm 2$ 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.

FIGURE 2. Energy spectrum recorded by the Si-PIN diode in liquid nitrogen doped with $^{222}$Rn, obtained for the positive polarization of the dewar ($+2$ kV) and the time dependence of the $^{214}$Po activity after changing the bias polarization ($2$ kV $\rightarrow -2$ kV). After the change marked with vertical dashed line negative ions become drifted towards the diode.

EXPERIMENTAL SETUP AND RESULTS

The experimental setup presented in Fig. 1 was used to study ion transport mechanism in liquefied nitrogen. Bare Si-PIN diode was immersed in a dewar filled with the cryoliquid. The diode was connected using a standard 50 $\Omega$ coaxial cable. Housing of the diode was grounded and connected to the signal cable shield. The diode was operated under nominal voltage of $+28$ V (relative to the ground). The diode was connected to a standard spectrometry electronics chain - charge preamplifier and the active filter amplifier. The readout system consisted of a multi channel analyzer, recording alpha-energy spectrum data in 900 s time windows. Changes of the dewar gross weight (32 l of total volume) were registered to calculate losses of the cryoliquid due to evaporation (boil-off).

The dewar was electrically isolated from the ground, the metal housing was wired to a bipolar high voltage (HV) power supply. By applying a positive bias to the housing cations were drifted towards the diode. Reversely, negative bias on the dewar put the diode on higher potential, attracting anions.

The first measurement followed at least 3 h after doping the liquid with $^{222}$Rn, to achieve decay (secular) equilibrium in the volume of the cryoliquid between $^{222}$Rn and its daughters. The recorded signal of $^{222}$Rn daughters $\alpha$-decays was corrected for $^{222}$Rn decay and liquid losses in off-line analysis. A typical spectrum recorded by the diode is shown in Fig. 2 for the dewar biased to $+2$ kV.

The radon dissolving procedure is directly followed by $^{214}$Po and $^{218}$Po $\alpha$-activity measurements in the cryoliquid. The $\alpha$-energy spectrum is recorded in 900 s real time windows, and stored as a multi-channel histograms. $^{214}$Po and $^{218}$Po energy peaks are selected from the spectrum (channels 5200 - 6700 for $^{214}$Po energy window, $Q = 7.69$ MeV), and the gross count rates in respective windows are corrected for $^{222}$Rn decay and liquid boil-off. The count rates are then analyzed as a function of time elapsed since the $^{222}$Rn doping and different bias voltages applied to the dewar. Change of the bias allows to select charge sign of the analyzed ions.

The Si-PIN diode registers only decays occurring up to 50 $\mu$m away from the semiconductor surface due to stopping of alphas in the cryoliquid. The time dependence of the $^{214}$Po count rate during the bias polarization change (preserving the HV magnitude or switching on of the bias) may be seen as a combination of two processes: a) decay of the diode surface residual activity, and b) time varying flux $\Phi(t)$ of the new ions attracted by the reversed bias:

$$A^+/- (t) = B(t, N_{Po}, N_{Pb}, N_{Bi}) + b + \Phi(t)$$  \hspace{1cm} (3)
TABLE 1. Parameters of investigated $^{214}$Po signal found for bias voltage changes (cf. Eq. 4), deduced from the proposed model. The $T_m$ value in boldface (transition $+2$ kV $\rightarrow$ $-2$ kV) was fixed as the fitting gave results beyond life-time of $^{222}$Rn daughters (the value corresponds to the mean life-time of $^{214}$Bi.)

<table>
<thead>
<tr>
<th>Bias voltage change</th>
<th>$\varepsilon$ (%)</th>
<th>$T_0$ (s)</th>
<th>$T_m$ (s)</th>
<th>$r$ (d$^{-1}$)</th>
<th>Ion type</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 $\rightarrow$ 2 kV</td>
<td>47.1(0.5)</td>
<td>5.9(0.1)</td>
<td>21.6(0.3)</td>
<td>4.4(0.2)</td>
<td>positive</td>
</tr>
<tr>
<td>2 $\rightarrow$ -2 kV</td>
<td>0.65(0.02)</td>
<td>0.4(0.1)</td>
<td>1188</td>
<td>11.80(1.0)</td>
<td>negative-like</td>
</tr>
<tr>
<td>-2 $\rightarrow$ 2 kV</td>
<td>55.6(0.3)</td>
<td>0.098(0.008)</td>
<td>9.56(0.06)</td>
<td>10.4(0.2)</td>
<td>positive</td>
</tr>
</tbody>
</table>

where $B$ is the Bateman expression for $^{214}$Bi activity (observed via $^{214}$Po $\alpha$-decays) as a function of time and $^{218}$Po, $^{214}$Pb and $^{214}$Bi deposited on the Si-PIN diode before the bias change (residual activities are not necessarily in secular equilibrium as different isotopes may exhibit different life-times and ionization probabilities), $b$ - background count rate, $\Phi$ - flux of the incoming ions.

The ions flux depends on ions life-time $T_{ion}(t)$. The life-time is a function of the local concentration of electronegative impurities (close to the diode surface). The impurity concentration change caused by presence of the E-field, diffusion or other transport processes, was modeled using a limited growth function:

$$T_{ion}(t) = T_0 T_m e^r t / (T_m + T_0 (e^r t - 1))$$

where $T_0$ is the minimum ionic life-time (related to the maximum local concentration of the impurities), $T_m$ is the maximum ionic life-time (minimum local concentration of the impurities), and $r$ is the impurity concentration change rate. (This function is a solution to a differential equation $\frac{dT}{dt} = r x(t)$, where $x(t) = \frac{T_{ion}(t)}{T_m}$, requiring $T_{ion}(0) = T_0$ and $T_{ion}(+\infty) = T_m$.) Fig. 2 shows the time dependence of the measured signal for change of the bias from 2 kV to -2 kV. Tab. 1 summarizes the fit parameters found for different studied scenarios.

CONCLUSIONS

We have shown the possibility to observe alpha-activity of positive and negative $^{222}$Rn-born ions, enhanced by electrostatic collection in cryogenic liquids. The maximum life-time of ions, varying in long time-scales, was found to be order of 10 s (depending on the quality of the cryoliquid). We also conclude, that the changes in local properties of the liquid influence the measurement results. This effect should be further investigated. We have also shown, that the Box model of germinate neutralization well describes our results. The amount of produced negative-like ions is below 1 %, and their life-time is mainly limited by the nuclear life-time.

We would like to pursue our activities further investigating $^{218}$Po and limiting sources of uncertainties. We also plan to employ electric field calculations to improve understanding of the germinate neutralization processes influence on production of the ions.

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REFERENCES