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Study of radon reduction in gases for rare event search experiments

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ABSTRACT

The noble elements, argon and xenon, are frequently employed as the target and event detector for weakly interacting particles such as neutrinos and Dark Matter. For such rare processes, background radiation must be carefully minimized. Radon provides one of the most significant contaminants since it is an inevitable product of trace amounts of natural uranium. To design a purification system for reducing such contamination, the adsorption characteristics of radon in nitrogen, argon, and xenon carrier gases on various types of charcoals with different adsorbing properties and intrinsic radioactive purities have been studied in the temperature range of 190–295 K at flow rates of 0.5 and 2 standard liters per minute. Essential performance parameters for the various charcoals include the average breakthrough times (τ), dynamic adsorption coefficients (k_a) and the number of theoretical stages (n). It is shown that the k_a-values for radon in nitrogen, argon, and xenon increase as the temperature of the charcoal traps decreases, and that they are significantly larger in nitrogen and argon than in xenon gas due to adsorption saturation effects. It is found that, unlike in xenon, the dynamic adsorption coefficients for radon in nitrogen and argon strictly obey the Arrhenius law. The experimental results strongly indicate that nitric acid etched Saratech is the best candidate among all used charcoal brands. It allows reducing total radon concentration in the LZ liquid Xe detector to meet the ultimate goal in the search for Dark Matter.

1. Introduction

Modern rare-event search experiments require low-radioactivity Time Projection Chambers (TPCs) to achieve high detection sensitivities. Noble gases such as argon (Ar) and xenon (Xe) are well-suited as target media for Dark Matter (DM) and Neutrinoless Double Beta Decay (NDBD) [1–3]. They are both excellent scintillators. Both are easily ionized by particles and can be easily purified of electronegative impurities [4] to achieve efficient charge transport. The challenge of every DM and NDBD experiment is to suppress radioactive backgrounds. Natural Ar and Xe do not have intrinsic long-lived isotopes but, during the production cycle, Xe can be contaminated with ^{85}Kr from the atmosphere. ^{39}Ar is produced in the atmosphere by cosmic rays scattering from ^{40}Ar. Both radioactive isotopes decay primarily by β-emission and their presence in detectors may limit ultimate sensitivities. Fortunately online distillation systems can be employed to remove these radioactive isotopes [5]. Radon (^{222}Rn), with a half life of 3.8 days, is another isotope that must be eliminated from TPC detectors. ^{222}Rn is a daughter of ^{238}U and is continuously supplied from warm detector components (e.g. cables, feedthroughs, etc.).

In the past charcoals have been used in low background experiments (Borexino [6], XMASS [7], SNO+ [8] and CUORE [9]) to reduce ^{222}Rn in the detectors by trapping the ^{222}Rn in charcoal long enough for it to decay but, to the best of our knowledge, available charcoal adsorbents have not been systematically studied. A gas system was fabricated to study ^{222}Rn reduction methods using commercial charcoal brands: Calgon Carbon (OVC 4 × 8) [10], Shirasagi (G2 × 4/6-1) [11], Saratech (Blücher GmbH.), nitric acid (HNO_3) etched Saratech (Blücher GmbH.) [12] and Carboact (Carboact International) [13]. The studies were performed within the scope of the LUX-ZEPLIN (LZ) DM experiment [14]. The physical conditions of charcoals ranged between 190 K and 295 K for ^{222}Rn in nitrogen (N_2), Ar and Xe as carrier gases with flow rates of 0.5 and 2 standard liters per minute (slpm).

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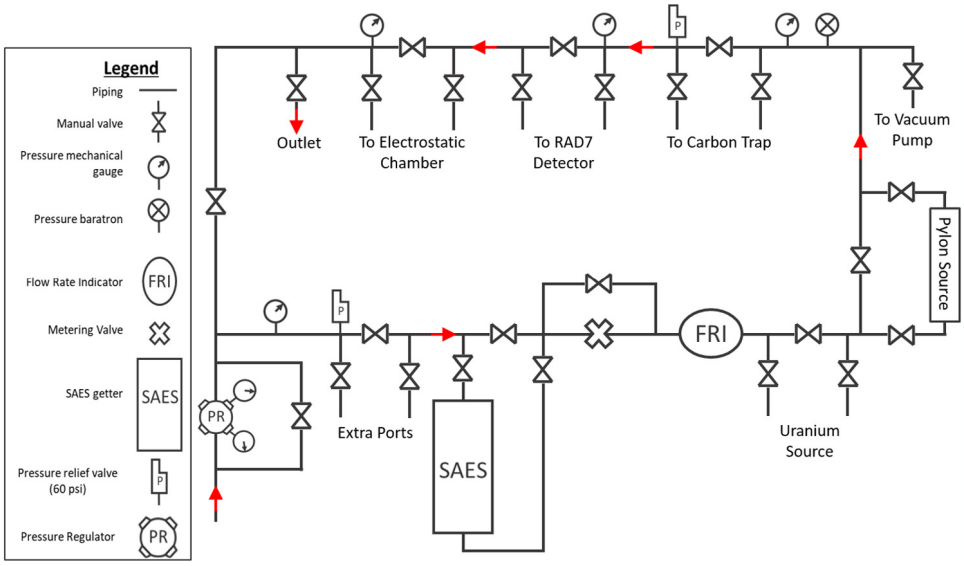

Fig. 1. Schematic view of the ^{222}Rn reduction and evaluation gas system.

2. Apparatus

2.1. Gas system

^{222}Rn adsorption on different types of charcoals was measured while entrained in N_2, Ar, and Xe carrier gases. A schematic view of the ^{222}Rn reduction system is shown in Fig. 1. The radon reduction system was designed and constructed at the University of Michigan.

The plumbing assemblies were welded (HE Lennon Inc., Swagelok, Michigan) [15] from 1/4" ultra-high vacuum (UHV) pipes and valves. After completion, the system was helium leak checked using a residual gas analyzer (RGA100, SRS), and no leaks were detected within the detection limits (5×10^{-14} Torr). Water vapor and other gas impurities may significantly affect the performance of a charcoal trap. Therefore, only boiled-off N_2 and Ar gases as well as research purity grade Xe gas were used for the measurements. Moreover, Xe and Ar gases were continuously purified during the measurements with a SAES high temperature getter (Model: PS3-MT3-R-1) composed of zirconium to remove oxygen, water vapor, nitrogen and other impurities [16]. The getter bed operating temperature was in the range of 350–400 °C. When pure N_2 was used, the getter was turned off and bypassed to prevent exothermic reaction leading to irreversible damage, and possibly explosion, to the zirconium cartridge. Before commencement of each set of measurements with carrier gases and charcoals, the moisture content of the trap was purged with boiled-off N_2 gas for 12 h. The charcoal trap was baked at 100 °C and evacuated with scroll and turbo-molecular pumps (Agilent Technologies). The charcoal trap and the UHV gas system were evacuated to at least 10^{-5} Torr for 12 h before the measurements began.

The experimental procedure was as follows. At the beginning of each measurement the gas was flowed through the charcoal trap and two independent radon detectors connected in series to evaluate radon background levels in the gas system and in the investigated charcoal trap. Uranium ores were initially used as ^{222}Rn source, but their activity was very weak. Therefore, they were replaced by a Pylon-1025 source (Electronics Development Company, Ltd.) containing dry radium (^{226}Ra) with an activity of 103 kBq and encapsulated in an aluminum cylinder to prevent its leakage [17]. The carrier gas was diverted through the source for up to 3 min resulting in an injection of a sharp, short pulse of ^{222}Rn into the charcoal trap. The carrier gas continued flowing through the trap for the duration of each individual measurement. The gas flow rate was controlled with a UHV gas regulator and a metering valve. It was measured with a UHV mass flow meter (Model:179A01314CR3AM, MKS) [18]. The precision and accuracy of the flow meter were 0.2% and 1% of the full scale, respectively. The pressure in the gas system was regulated using a check-valve installed at the outlet port. The mass flow meter was operated with a single channel power supply readout (Model: 246C, MKS) [18]. The gas system included mechanical gas pressure gauges and a baratron absolute pressure transducer with an analog read-out before and after the trap to measure the gas pressure. The measurements were made at different gas pressures to study their impact on ^{222}Rn adsorption on charcoals. The gas system was equipped with pressure relief valves (PRV) rated to 60 pounds per square inch (psi). When average ^{222}Rn breakthrough times were measured in Ar and N_2 carrier gases, the gases were not recovered in view of their low cost. The system handled 16 kg of Xe gas contained in two aluminum cylinders with volumes of 30 l each. The xenon gas was cryogenically transferred between the two cylinders using liquid N_2. The mass of the Xe gas was measured with tension load cells (FL25-50 kg, Forsentek, China) [19].

2.2. Vacuum-jacketed cryostat and charcoal traps

In order to select the optimal radon adsorbent, different types of charcoals were investigated. Charcoals were contained in modified conflat UHV vessels (Kurt J. Lesker, USA) [20]. The dimensions of the vessels were 3.4 cm and 6.4 cm in diameter and 12.6 cm and 35 cm in length with corresponding volumes of 0.1 l and 1.1 l, respectively. Charcoals Calgon OVC 4 × 8 (Calgon, 50 g, 0.1 l), Shirasagi (G2 × 4/6-1, 45 g, 0.1 l), Saratech (70 g, 0.1 l; 650 g, 1.1 l), HNO_3 etched Saratech (650 g, 1.1 l) and Carboact (241 g, 1.1 l) were selected for their different properties such as porosity, density, surface area, radioactive background as well as relative cost. Before assembly, the traps and their UHV components were thoroughly cleaned in methanol. All charcoals, except specially treated etched Saratech, were then rinsed with deionized (DI) water. The traps were equipped with fine stainless steel (SS) meshes as well as 60 μm UHV filter gaskets on the inlet and outlet to prevent escape of fine charcoal grains and dust into the main volume of the gas system. The pressure difference between the inlet and outlet of the charcoal trap was measured to be less than 1 psi. The charcoal bed layers were compressed with a SS spring to maintain stable and uniform packing. The meshes and springs were cleaned in an ultrasonic bath with methanol prior to assembly. The charcoal traps were equipped with calibrated platinum RTD PT100 (Omega) temperature sensors [21] embedded in the charcoal layers. The temperatures of the inner volume in the vacuum-jacketed cryostat and the inner volume of the charcoal trap agreed within 0.5%. The vacuum-jacketed cryostat was well suited (Cryofab, Cryogenic equipment, USA) [22] to store charcoal traps and