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Citation: *AIP Conf. Proc.* **1549**, 185 (2013); doi: 10.1063/1.4818105

View online: <http://dx.doi.org/10.1063/1.4818105>

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# Radiopurity Measurement of Acrylic for DEAP-3600

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**Abstract.** The spherical acrylic vessel that contains the liquid argon target is the most critical detector component in the DEAP-3600 dark matter experiment. Alpha decays near the inner surface of the acrylic vessel are one of the main sources of background in the detector. A fraction of the alpha energy, or the recoiling nucleus from the alpha decay, could misreconstruct in the fiducial volume and result in a false candidate dark matter event. Acrylic has low levels of inherent contamination from  $^{238}\text{U}$  and  $^{232}\text{Th}$ . Another background of particular concern is diffusion of  $^{222}\text{Rn}$  during manufacturing, leading to  $^{210}\text{Pb}$  contamination. The maximum acceptable concentrations in the DEAP-3600 acrylic vessel are ppt levels of  $^{238}\text{U}$  and  $^{232}\text{Th}$  equivalent, and  $10^{-8}$  ppt  $^{210}\text{Pb}$ . The impurities in the bulk acrylic will be measured by vaporizing a large quantity of acrylic and counting the concentrated residue with ultra-low background HPGe detectors and a low background alpha spectrometer. An overview of the acrylic assay technique is presented.

**Keywords:** poly(methyl methacrylate), assay, gamma assay,  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ , ultra trace level, low background

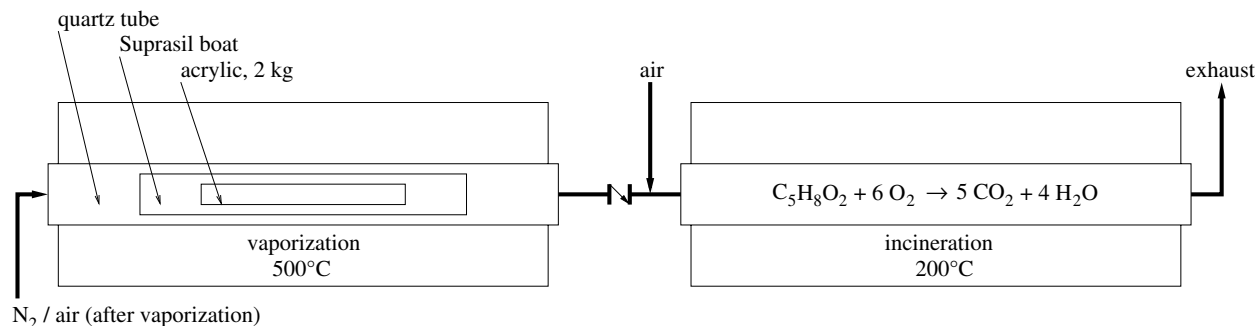
## BACKGROUND REQUIREMENTS FOR THE ACRYLIC VESSEL

DEAP-3600 is a single-phase liquid argon dark matter detector that will increase sensitivity to the spin-independent WIMP-nucleon cross section to  $10^{-46}$  cm<sup>2</sup>. Currently in the installation phase at SNOLAB, the detector will contain 3600 kg of liquid argon in a 2-in. thick acrylic vessel with an inner radius of 85 cm. Scintillation light will be detected by the surrounding 255 high quantum efficiency photomultiplier tubes (PMT's). Argon provides excellent discrimination between electromagnetic interactions and nuclear recoils based on scintillation time. All potential sources of backgrounds have been studied and mitigated, see Ref. [1] for a summary. This paper relates to surface backgrounds – alpha decays from the surface of the acrylic vessel. Acrylic has trace levels of  $^{238}\text{U}$  and  $^{232}\text{Th}$ , and perhaps more challenging is  $^{222}\text{Rn}$  diffusion from air into the materials used to make acrylic. Controlling radon exposure is of utmost importance. Close collaboration continued at every stage of manufacturing to produce an extremely clean acrylic vessel. From the bulk acrylic, the background budget is 0.01 background events in the inner 1000-kg fiducial volume at the 15–30 keV<sub>ee</sub> WIMP energy region of interest during 3 years of live time. Table 1 shows the corresponding maximum allowable concentrations for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{210}\text{Pb}$ , as calculated by a Monte Carlo model [2].

**TABLE 1.** Limits in bulk acrylic

	( $\mu\text{Bq/kg}$ )	(g/g)
$^{238}\text{U}$	3.7	$0.3 \times 10^{-12}$
$^{232}\text{Th}$	5.3	$1.3 \times 10^{-12}$
$^{210}\text{Pb}$	31	$1.1 \times 10^{-20}$

The acrylic assay is based on the vaporization technique used by SNO, which achieved 0.5-ppt sensitivity for  $^{238}\text{U}$  and  $^{232}\text{Th}$  [3]. To measure the radiopurity of acrylic at such low levels, 10 kg of acrylic will be vaporized and the residue will be collected by rinsing with heated acids. Count rates can be increased from the expected 1 count/day (cpd) level by vaporizing even more acrylic. For  $^{238}\text{U}$  and  $^{232}\text{Th}$ , gammas from further down the chains, specifically from  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ , will be measured with an ultra low background HPGe detector. The  $^{210}\text{Pb}$  will be measured with a well detector, and also by its  $^{210}\text{Po}$  daughter with an alpha counter.



**FIGURE 1.** The acrylic vaporization system first vaporizes PMMA in an  $N_2$  atmosphere, and then destroys MMA by incineration. A block of acrylic is contained in a cylindrical Suprasil boat. Air is added to the incineration furnace. After vaporization is complete, carbon is removed from the boat by adding air to the vaporization furnace.

## THE ACRYLIC VAPORIZATION SYSTEM

Located at the surface site of SNOLAB, the acrylic vaporization system is the core of the acrylic assay. Acrylic is poly(methyl methacrylate). When heated, the PMMA polymer tends to breakup into its MMA monomer [4]. The boiling point of MMA is  $(100.6 \pm 0.2)^\circ\text{C}$  [5], and therefore the primary product of vaporizing acrylic is MMA vapor. Although the exposure to MMA is not considered a hazard, MMA has a strong and unpleasant odor. For this reason, the acrylic vaporization system is designed for pyrolysis of acrylic followed by removal of MMA by combustion. The system consists of two large furnaces, each 1.2 m in length, for vaporization and incineration. See Fig. 1. A block of acrylic,  $6 \times 6 \times 50$  cm with a mass of 2 kg, is cut on a dedicated saw. The acrylic is placed in a 75-cm cylindrical boat made of Suprasil, an ultra pure synthetic quartz. To contain the acrylic when it melts, the ends of the boat are flared inward. The boat sits inside a 15-cm diameter and 1.5-m long quartz tube in the vaporization furnace. A flow of  $N_2$  gas is initiated and the vaporization furnace is heated to  $500^\circ\text{C}$ . The  $N_2$  gas is boiloff from a large liquid nitrogen storage tank. The lid of the furnace may be opened to observe the vaporization, or incineration. During vaporization, bubbles form on the surface, the block foams and expands, and there is a change from colorless to yellow. The vapor is white in color. The MMA vapor flows from the vaporization furnace to the incineration furnace, and the connecting section must be maintained above  $100^\circ\text{C}$ . After a 0.5-psi check valve, air is injected into the incineration furnace to provide oxygen for combustion,  $C_5H_8O_2 + 6 O_2 \rightarrow 5 CO_2 + 4 H_2O$ . The incineration furnace is filled with steel wool to provide a large surface area and is operated at  $200^\circ\text{C}$ . The products move to the exhaust line. An excessive amount of char accumulates in the quartz boat; it has been estimated to be as much as 15% of the initial mass of PMMA [4]. This solid material would make rinsing and counting difficult. To remove carbon, air is added to the vaporization furnace after vaporization is complete. Carbon combines with oxygen to form carbon dioxide, which goes to the exhaust,  $C + O_2 \rightarrow CO_2$ . The acrylic vaporization system can accommodate 2 kg each day, however, on the order of 10 kg is required to increase count rates. After having vaporized five 2-kg blocks in the same boat, the boat is moved into a cleanroom. The boat is placed on heated rollers at 1 rpm and rinsed with aqua regia, 1:3 vol/vol concentrated nitric acid and concentrated hydrochloric acid. There are two 1-h rinses with 50 ml of aqua regia, during which time the acids reach approximately  $45^\circ\text{C}$ , followed by a 5-min rinse with 20 ml of ultra pure water (UPW). The effluent is collected and the volume can be reduced by evaporation using a PTFE beaker directly on a hot plate. An FEP bottle can be used to store the effluent.

## ULTRA LOW BACKGROUND GERMANIUM DETECTORS

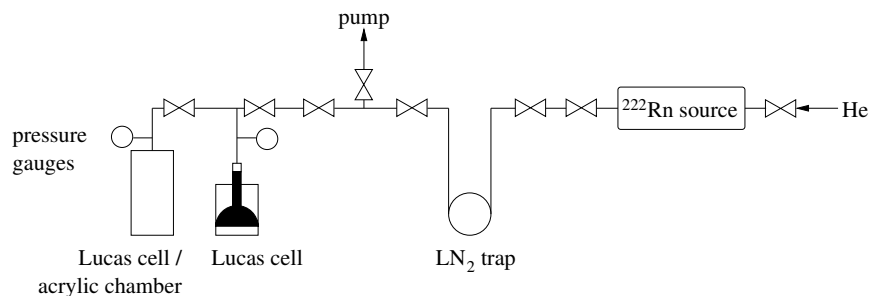
The effluent will be measured with custom ultra low background high purity germanium (HPGe) detectors underground. SNOLAB has a well-established gamma assay program with a  $200\text{-cm}^3$  coaxial detector. Another  $400\text{-cm}^3$  p-type coaxial detector and a  $300\text{-cm}^3$  well detector were purchased by Queen's University for assay of components in the DEAP and SNO+ detectors. The well detector has high efficiency and low background for the 46.539-keV gamma from  $^{210}\text{Pb}$ , which has a 4.25% branching fraction. The well has a diameter of 21 mm and a depth of 66.5 mm. A 3-ml PTFE bottle, selected to fit in the well detector, contains the effluent and is placed directly in the detector.

## LOW BACKGROUND ALPHA SPECTROMETRY

In parallel to the well detector measurement,  $^{210}\text{Pb}$  will be measured by the 5.304-MeV alpha from its  $^{210}\text{Po}$  daughter. Polonium begins to volatilize at approximately  $100^\circ\text{C}$  [6], and should be removed during vaporization. Storing the effluent for a month allows  $^{210}\text{Po}$  to grow-in. Polonium spontaneously deposits on various metals, including nickel [7]. The nickel disks are  $\frac{1}{2}$  in. in diameter and were cut from approximately 0.03-in. thick Nickel 200. A small hole is drilled close to the edge of each disk. The disks are then polished; some have been polished by hand and some electropolished. The sample is transferred from an FEP bottle to a PTFE beaker, and the FEP bottle is rinsed with three 5-ml portions of hydrochloric acid. The PTFE beaker is placed on a  $100^\circ\text{C}$  hot plate monitored with an RTD, a resistance temperature detector. Once the sample has evaporated to near dryness, 50 ml of UPW is added followed by 10 ml of hydrochloric acid and 100 mg of ascorbic acid. A length of PTFE tape is threaded through the hole in the disk for handling. To degrease the nickel disk, it is dipped in nitric acid, then hydrochloric acid, and finally UPW. The disk is submerged for 2.5 h while the sample is stirred and heated to  $55^\circ\text{C}$ . The magnetic property of nickel on the stirrer provides the agitation. At the end of deposition, the disk is dipped in UPW and dried with boiloff  $\text{N}_2$  gas. Two low background 450-mm<sup>2</sup> ion-implanted-silicon detectors are used with battery-biased sample holders. The negative bias keeps the positively charged recoiling nuclei from contaminating the detector. Both counters are at the specification of 6 cpd at 3–8 MeV,  $(7.6 \pm 1.2)$  cpd and  $(5 \pm 1)$  cpd. An encapsulated, collimated  $^{241}\text{Am}$  source was used to measure the resolution for the counters, 19 keV and 24 keV, and to determine that the effect of the dead layer is negligible. Due to geometry, the counting efficiency is 15%; only  $^{210}\text{Po}$  from one side of the disk will reach the detector, and there is a solid angle based on the 8-mm separation between the nickel disk and the detector. The first acrylic samples that were measured for practice were consistent with the background of the counters at the  $^{210}\text{Po}$  energy window, set at 3.5–5.35 MeV, and correspond to a sensitivity of  $10^{-19}$  g/g  $^{210}\text{Pb}$ .

## LEAD SPIKE TO MEASURE RECOVERY EFFICIENCY

To determine the loss of lead during vaporization, rinsing, and during  $^{210}\text{Po}$  collection, a measured amount of  $^{222}\text{Rn}$  was loaded into acrylic. The setup consists of a  $^{222}\text{Rn}$  source, a liquid nitrogen cooled stainless steel coil as a trap, two Lucas cells, and an empty acrylic chamber. See Fig. 2. The  $^{222}\text{Rn}$  source, RN-1025 from Pylon Electronics Inc., has an activity of 526.545 kBq equilibrium or 66239.419 mBq/min continuously available from  $^{226}\text{Ra}$  powder. The chamber is made from an acrylic tube, and acrylic cement is used to attach an acrylic disk to one end. An aluminum clamp with an O-ring sealed the chamber. The tubing is  $\frac{1}{4}$  in. stainless steel. A Lucas cell is an evacuated acrylic container coated with scintillating  $\text{ZnS}(\text{Ag})$  paint used for measuring radon by counting with a PMT. The procedure begins by evacuating the Lucas cells for 2 h and measuring the backgrounds. Upon reattaching the Lucas cells to the setup after counting, they are evacuated for another 30 min. The  $^{222}\text{Rn}$  source is purged with helium gas and then closed. The time is recorded while the source regenerates. The trap is evacuated and cooled with liquid nitrogen for 30 min. A valve at the source is opened and the  $^{222}\text{Rn}$  gas moves from the source to the trap by free expansion for 10 min. The source is returned to atmospheric pressure with helium gas. The trap is opened to the Lucas cell, and while keeping liquid nitrogen on the trap, the system is evacuated to low pressure. The trap is warmed with a heat gun for several minutes and for another 10 min the  $^{222}\text{Rn}$  redistributes. The Lucas cell is removed and counted after 3 h, at which point the alpha-emitting daughters,  $^{218}\text{Po}$  and  $^{214}\text{Po}$ , are approximately in transient equilibrium with the parent,  $^{222}\text{Rn}$ . To understand the transfer from the Lucas cell to the acrylic chamber and the connecting tubing, a transfer is first completed between two Lucas cells. The hot Lucas cell is opened to another Lucas cell that is attached where the acrylic chamber would be attached. Having the same volume, the Lucas cells obtain equal amounts of  $^{222}\text{Rn}$ . The amount of  $^{222}\text{Rn}$  in the tubing is determined from the difference between the initial amount and the amount in both Lucas cells. One Lucas cell is then opened to the evacuated acrylic chamber. After 2 weeks, while  $^{222}\text{Rn}$  decays to  $^{210}\text{Pb}$ , the acrylic chamber is vaporized and rinsed. The effluent is counted directly in the well detector, and can be used later for  $^{210}\text{Po}$  collection efficiency and alpha counting.



**FIGURE 2.** A measured amount of  $^{222}\text{Rn}$  is transferred to an evacuated acrylic chamber. Lucas cells measure the amount of  $^{222}\text{Rn}$  to understand the available volumes during free expansion. The acrylic chamber is sealed for 2 weeks while  $^{222}\text{Rn}$  decays to  $^{210}\text{Pb}$ . After vaporization, rinsing, and  $^{210}\text{Po}$  collection, the recovery efficiency of lead is determined.

## SUMMARY

Radiopurity of materials must be understood at unprecedented levels during such a sensitive dark matter experiment. Fabrication of the DEAP-3600 acrylic vessel involved extensive effort to minimize any contamination. An acrylic assay based on vaporization has been developed. After collection of the residue with heated aqua regia, samples are counted in low background HPGe detectors. In addition, ultra low background alpha counters are used to measure  $^{210}\text{Po}$  deposited on disks. The procedure will next be optimized for the lead recovery efficiency. These techniques will be the most sensitive yet for  $^{238}\text{U}$  and  $^{232}\text{Th}$  equivalent, and especially for  $^{210}\text{Pb}$ .

## ACKNOWLEDGMENTS

The design, installation, commissioning, operation, and analysis for the various aspects of the acrylic assay involved many people from the DEAP collaboration. From SNOLAB, we would like to thank L. Anselmo for assistance with sample preparation, vaporization, and rinsing, B. Morissette and the installation team for assembling the acrylic vaporization system, I. Lawson for measurements with the well detector, and C. Jillings, F. Duncan, O. Li, and R. Ford for useful discussions. From Queen's University, we acknowledge in particular the following people: B. Cai, M. Kuźniak, A. McDonald, A. Noble, P. Skensved, P. Rau, W. Rau, and M. Ward. This research is supported by the Natural Sciences and Engineering Research Council of Canada, the Canada Foundation for Innovation, and the Ontario Ministry of Research and Innovation.

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