

A ^{222}Rn Source for Low-background Liquid Scintillation Detectors

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ABSTRACT

A technique for producing a radioactive source suitable for use in a low-background liquid scintillation detector is described. ^{222}Rn was concentrated from air to prepare liquid scintillator sources spiked with 10^6 Bq/m^3 of the radioisotope. Air was stripped of CO_2 and water vapor, and passed over cooled charcoal which trapped the radon. The accumulated radon was desorbed and transferred into a pseudocumene-based scintillator. These sources have been used for position calibration in the Counting Test Facility (a 5 m^3 spherical liquid scintillation detector) at the Laboratori Nazionali del Gran Sasso.

INTRODUCTION

Concentrated ^{222}Rn makes an ideal radioactive source for a variety of applications. ^{222}Rn , with a 3.8 day half life, is sufficiently long-lived to permit it to be used over a period of days, yet it decays away quickly enough that it doesn't pose a risk for long term contamination. We describe here the preparation of a radon source by concentrating radon from the air. This method created radioactive ^{222}Rn sources, ~ 1 Bq/ml, without requiring special precautions necessary with utilizing long-lived Ra isotopes. (^{222}Rn is a daughter of ^{226}Ra , and radon sources can be produced from radium decay.) Our particular implementation was for a small volume ($\sim 1\text{-}5\text{ cm}^3$) source of ~ 1 Bq for position calibration in the Counting Test Facility (CTF).

The CTF is a large-volume, low-background liquid scintillator detector located in the Laboratori Nazionali del Gran Sasso (LNGS) in Italy [1]. Its principal objective was to demonstrate that sufficient radiopurity could be achieved in large volume of liquid scintillator for use as a solar neutrino detector. The detector employs grading shielding, including an active shield within the scintillator region to only accept scintillation events within a central fiducial volume of the scintillator. The active shielding requires accurate reconstruction of the spatial location of the scintillation event. Photon arrival times at 100 phototubes contained within a water shield distributed around the scintillator volume are employed to reconstruct the event position. Small radioactive sources were moved to precise locations within the scintillator volume to calibrate the reconstruction algorithm and to determine the spatial resolution of the event reconstruction. Radon spiked scintillator sources were identified as the most desirable to calibrate the event position reconstruction within the CTF detector [2].

The source consisted of ^{222}Rn spiked scintillator contained in a quartz vial which could be inserted into the detector. The scintillator was a simple two component scintillator, 1.5 g 2,5 diphenyl oxazole (PPO) per liter of 1,2,4-trimethyl benzene (pseudocumene). Radon offered several advantages as a radioactive source material for

the CTF, especially the ease with which it could be detected. In the energy range of interest (0.2-1 MeV) the background rate in the CTF was approximately 2×10^{-2} Bq. Fortunately, in the ^{222}Rn decay sequence shown in Figure 1, there are time-correlated events. In particular, the decay of ^{214}Bi followed by the decay of ^{214}Po with a mean lifetime of 236 μs was used to distinguish radon events. The detection electronics for the CTF were specifically designed to detect both the energy (by number of photons detected) and time correlation of scintillation events [1]. The background coincidence rate of events correlated with mean delay times of 10 ns - 5 ms was less than 10 events per day. The time correlated ^{214}Bi - ^{214}Po decay sequence in a 0.1 Bq radon source was easily detected above the background in the CTF. Events tagged by time correlation as arising from the decay of ^{214}Bi - ^{214}Po could in turn be employed for energy calibration of the scintillation detector.

The second important advantage of the ^{222}Rn source was it was sufficiently short-lived that accidental contamination of the CTF detector by the source would not be fatal. The radon would decay with a decay time of 5.5 days, and the only residual activity after one month would result from ^{210}Pb - ^{210}Bi - ^{210}Po decay. A 1 Bq source that released its contents into the CTF would contribute a long-lived background of $< 4 \times 10^{-3}$ Bq in the energy window of 250-800 keV, which was below of initial background level.

A final consideration in our method of source preparation was that the Borexino collaboration would not permit "long lived" radioactive sources to be brought into the confines of the low-background CTF detector. In particular we were prohibited from bringing a radium source in the underground laboratory, leading us to exploit the naturally occurring radon in the underground water and air.

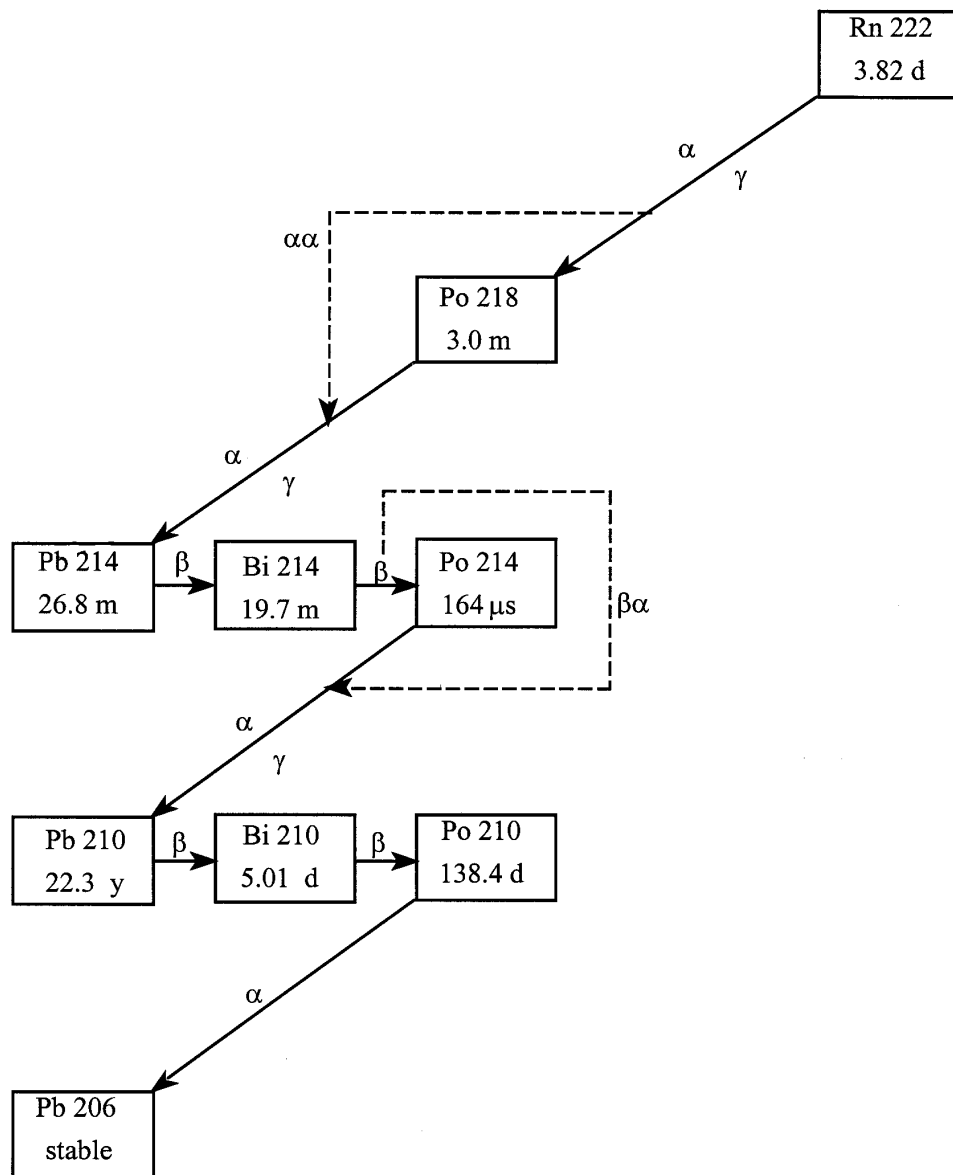


Figure 1. Decay chain of ^{222}Rn . ^{222}Rn decays can be tagged by observing the ^{214}Bi - ^{214}Po delayed coincidences with a mean decay time of 236 μs .

EXPERIMENTAL PROCEDURES AND RESULTS

The sources were prepared in Hall C of the underground laboratories at the LNGS. The abundance of radon in the air (100 Bq/m^3) and water (10^4 Bq/m^3) of the laboratories provided a good source of radon. The overall procedure was to trap radon

onto a cooled carbon adsorbent, and then transfer the radon into the scintillator liquid. Trapping radon onto charcoal is a well know procedure going all the way back to Rutherford [3], and is the basis for collecting samples for measuring radon in air [4,5]. Typically air is passed over a charcoal adsorbent for a period of time to collect radon, and the radon content on the adsorbent is measured. Radon adsorption onto carbon increases with decreasing temperature [6,7]. Water vapor and carbon dioxide in the air compete with radon for adsorption onto carbon, and hence limit the amount of radon that can be adsorbed [8,9]. To achieve higher adsorbed radon concentrations onto carbon we remove both carbon dioxide and water vapor from the air, and then adsorb radon onto a cooled carbon trap.

Water, carbon dioxide and volatile organics are at concentrations in air more than ten orders of magnitude greater than radon, and these species are more effectively adsorbed on carbon adsorbents than radon, necessitating their removal without depleting the radon. Even after concentrating the radon it still had an insignificant partial pressure, so it required careful handling to transfer the radon into the scintillator without excessive dilution. To put the concentrations into perspective, the air in Hall C of LNGS is at 15°C, with 70% relative humidity, and has radon at >100 Bq/m³. The CO₂ concentration is ~500 ppm (slightly greater than outdoor air). All these values are compared on a common basis in Table I.

Table I
Concentrations in Air

Species	Typical Concentration Range	Molecular Concentration (molecules/m ³)
Water Vapor	50% relative humidity 20°C	2.87×10^{23}
Carbon Dioxide	300 ppm	7.44×10^{21}
Radon	25 Bq/m ³	1.19×10^7

The apparatus for trapping radon in charcoal is shown in Figure 2. Air containing radon is blown through the system by a small diaphragm pump. The air passes through a concentrated solution of NaOH that removes CO₂, through a Drierite column to remove water, and through a cryogenically cooled charcoal trap. The typical air flow rate was varied from 100 cm³/s to 500 cm³/s.

The radon trap was a stainless steel tube 1-cm diameter 12-cm long closed at one end with a tee on the top. A 2-mm tube was fed through the tee to the bottom of the 1-cm tube. The annular region was filled with 5 g charcoal (Cameron-Yakima, Inc. CYVCC Coconut Shell Carbon 8 x 30). The charcoal in the radon trap was prepared by pulling a vacuum on it with a mechanical pump while heating to 100-150°C for approximately one-half hour. This pre-treatment of the charcoal was crucial to the final results. Heating in vacuum removed adsorbed water and carbon dioxide that collected on the charcoal from exposure to air. The presence of adsorbed water and CO₂ reduced the adsorption capacity of the charcoal for radon.

The CO₂ trap consisted of 75 g NaOH in 50-ml water. This was a saturated solution at 20°C with an excess of solid NaOH present in the bottom of the trap. Air was forced through a porous glass frit immersed in the aqueous NaOH solution producing bubbles ~100 μm in diameter. In separate experiments we measured the effectiveness of this technique for removing CO₂ from air. At a flow rate of 80 cm³/s the CO₂ level was reduced to ~60 ppm (from an ambient of 450 ppm) and the water vapor pressure was 6 mbar (from an ambient of 20 mbar or 20,000 ppm) after sparging through the solution. At a flow rate of 160 cm³/s the CO₂ level after sparging was ~175 ppm and the water vapor pressure was 8 mbar.

The Drierite column was filled with CaSO₄ pellets, which adsorbed water from the air. The calcium sulfate had a color additive that changed color when the pellets were saturated with water vapor. Flow through the system was halted when the front corresponding to water absorption by the calcium sulfate reached 3/4 the length of the

column. The Drierite column reduced the water vapor pressure to < 0.5 mbar, but did nothing to reduce the CO_2 level in the air.

The dry, "CO₂-free" air then passed into the radon trap. This trap consisted of charcoal that was cooled in a cryogenic bath. A temperature bath of frozen ethanol (-117°C) was used to cool the trap. At this

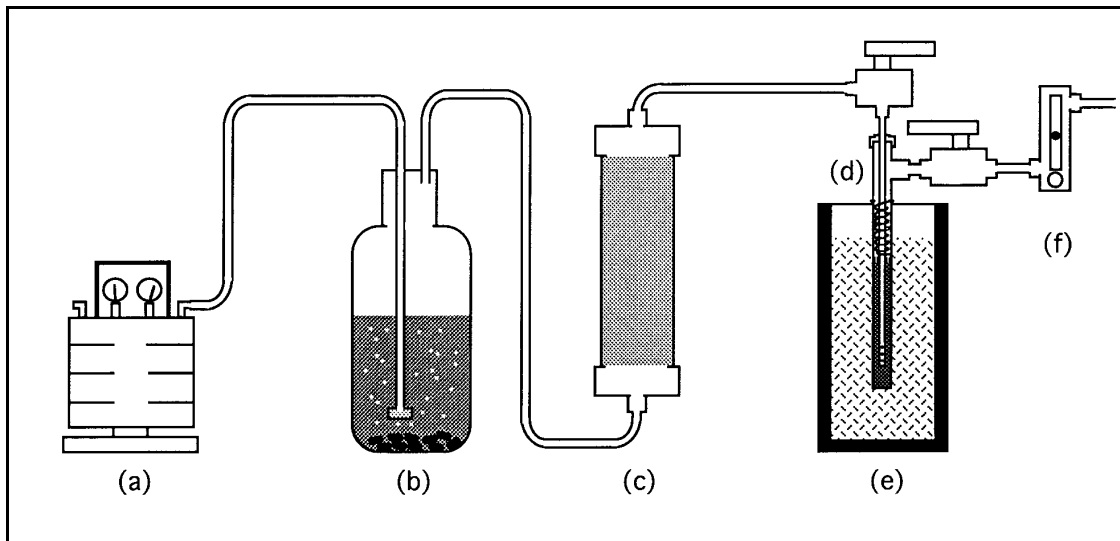


Figure 2. Radon trapping apparatus which consists of a) a diaphragm pump, b) CO₂ trap, c) Drierite column, d) charcoal trap surrounded by e) cryogenic bath, and f) a flowmeter.

temperature radon (bp -62°C) is trapped on the charcoal while nitrogen (bp -196°C) and oxygen (bp -183°C) are not. The CO₂ (sublimation pt. -79°C) and water removal steps were necessary because these gases would be trapped on the charcoal at frozen ethanol temperatures and saturate the charcoal before an adequate amount of radon could be trapped. The air passed through a flowmeter after it left the charcoal trap and was returned to the atmosphere.

Normally air was passed through the system at a flow rate of 200 ml/s for nearly two hours. The Drierite was usually near exhaustion at this point. If 100% of the radon in the air was adsorbed on the charcoal, the charcoal would contain more than 100 Bq of radon activity. By replenishing the CaSO₄ in the Drierite column and the NaOH in the

CO₂ trap, more air could be passed through the system. In one case, the airflow was continued for 12 hours, and a stronger source was produced.

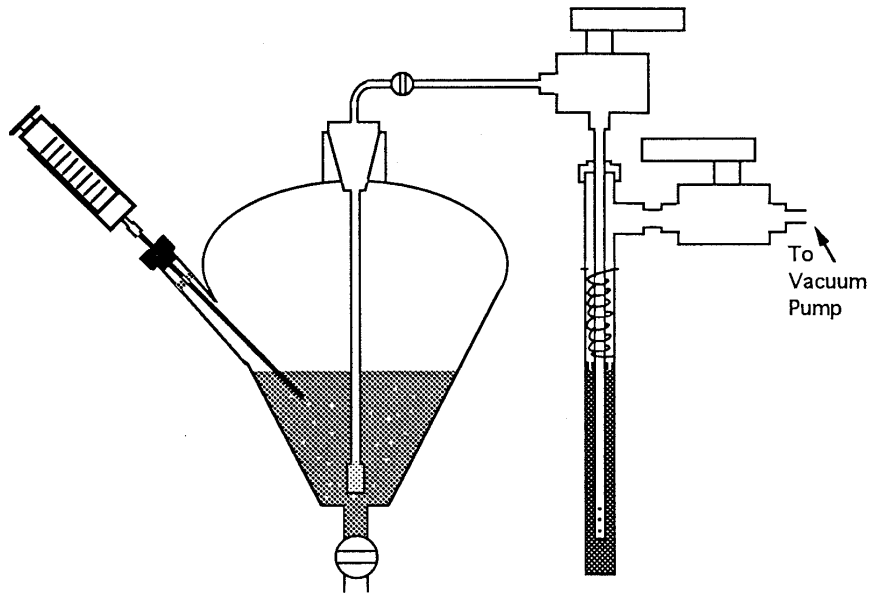


Figure 3. Separatory funnel used to transfer radon from the charcoal trap to the 50-ml scintillator sample in the flask. The syringe is used to remove radon-spiked scintillator from flask and to fill the sample vial. The syringe is not present during the transfer from the trap. The valve at the top of the funnel is connected to the radon trap as shown.

The radon trapped in the charcoal was transferred into a sample of scintillator using the apparatus shown in Figure 3. The radon trap was detached from the air flow system, and connected to a flask containing approximately 50 ml of scintillator. The scintillator in the flask was identical to that which was present in the CTF, nominally 1.5-g of PPO per liter of pseudocumene. The scintillator was thoroughly sparged with nitrogen or helium to removed any dissolved oxygen (dissolved oxygen quenched the fluorescence yield of the scintillator). The flask was attached to the air inlet side of the radon trap. A vacuum pump was attached to the air outlet from the trap, and the trap was evacuated while being maintained in the frozen ethanol bath at -117°C . After evacuation,

the charcoal trap was isolated, and the trap was removed from the ethanol bath and warmed using electric heat tape. A pressure gauge connected to the trap showed a rise in pressure as the heating caused radon (and CO₂ and water) to desorb from the charcoal. After heating, a small amount of helium was introduced into the trap through the valve where the vacuum pump had been connected. This helium increased the pressure in the trap; the valve between the trap and the scintillator flask was opened permitting gas to flow into the scintillator. The gas bubbled through a frit, exposing the scintillator to concentrated radon. The mass transfer rate of the desorbed radon from the gas phase into the liquid was low, and the helium carrier gas forced more gas through the scintillator facilitating the radon transfer in a short period of time.

Even with the use of helium, this procedure had a rather low efficiency. It was estimated that at least 100 Bq of radon was trapped in the charcoal. This radon was extracted into 50 ml of scintillator. The partition coefficient of radon in the vapor to radon in the scintillator was estimated from the Ostwald coefficients of radon in pseudocumene and xylene of 11.5 [10,11]. The vapor volume in the trap and above the scintillator was approximately 20 cm³. If all the radon in the inlet air had been trapped on the charcoal and equilibrium between gaseous radon and dissolved radon was established, a source solution with strength of 2 Bq/ml would have been obtained. The source solution typically had strength of 0.5 Bq/ml, indicating an overall efficiency for radon collection and transfer of 25%.

The radon-spiked scintillator was transferred to source vials for insertion into the detector. Selection and preparation of the source vials were important to reduce the risk of contamination of the CTF detector. The source vials were made of quartz, with a refractive index similar to the CTF scintillator. The vials were cleaned by soaking in 10% nitric acid at 60°C for several hours, rinsed with DI water and dried with nitrogen prior to use.

Several different shapes of quartz vial were used, with volumes between 1 and 5 ml. Some of these are shown with their dimensions in Figure 4. These vials were sealed with threaded Teflon or nylon caps and Teflon tape.

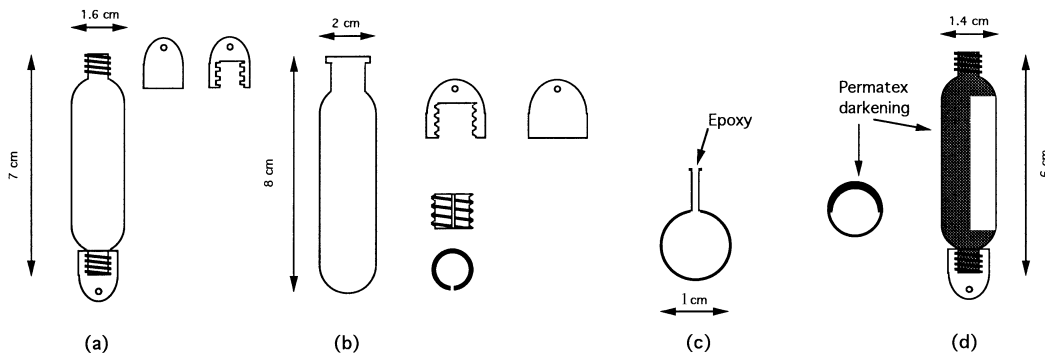


Figure 4 (a-d). Quartz source vials that were used for calibration in the CTF detector.

A specially prepared vial was made to study light scattering in the CTF; 180° of the inside of the vial was painted black with Permatex (Fig. 4d), so light could only escape in the non-blackened directions. The intensity of light for directly illuminated phototubes to the intensity for those tubes only receiving scattered light was measured [2].

In the transfer of the radon-spiked scintillator to the vials, it was necessary to minimize oxygen contamination because oxygen quenches the fluorescence of the scintillator. The scintillator in the flask mentioned above was sparged with nitrogen prior to radon transfer, and was kept isolated from air after the sparging. Spiked scintillator was removed from the flask with a syringe through a septum and then injected into the source vials. The source vials were flushed with nitrogen prior to filling and the nitrogen flow was continued until moments before the vial was capped. The spiked scintillator could not be sparged for oxygen removal because this would have removed the radon. Although the procedures for avoiding oxygen contamination were improved as more sources were prepared, most showed some evidence of oxygen quenching. This was

evidenced by a reduction of the energy of the ^{214}Bi β 's and the ^{214}Po α 's as compared with their positions from events occurring in the free volume of the CTF detector. As a result of the scintillation quenching by oxygen, the sources were not used for an absolute energy calibration.

There are other more efficient methods of preparing radon-spiked sources. Workers at NIST have developed a polyethylene encapsulated radium source that emanates radon at a constant rate over extended periods of time (they have tested it over the period of years) [12-14]. Their use has been principally as a source of radon emanation into water. We tested a variation on this approach placing radium source in a flask connected to a second flask containing the scintillator sample and allowing the radon to equilibrate between the scintillator liquid and the vapor. Direct counting with a germanium detector of the scintillator material after 1-day exposure to the radon suggested this was a good source. However, we were not permitted to use this source due to concern over potential contamination of the CTF by radium.

CONCLUSION

In this work, a technique was developed for producing radioactive "point" sources suitable for use in a large-volume low-background detector liquid scintillation detector. These sources were prepared by trapping of radon from the underground air and transferring it into scintillator liquid. Sources with an activity ~ 1 Bq/ml could be readily produced by the methods outlined here. This source preparation method was safe and utilized no controlled radioactive sources that might require special handling. The ^{222}Rn source produced events easily discriminated from the background through the use of time correlated decays.

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