Borexino: A Scintillator Purification System for a Large Scale Solar Neutrino Experiment

An on-line purification system for a large-scale low-background liquid scintillation detector has been tested with the 5-ton liquid scintillation detector, the Counting Test Facility at Gran Sasso. Counter-current water extraction removes polar and charged impurities such as ions and metal salts. Vacuum distillation of the entire scintillator has been applied to remove both radioactive impurities and chemical impurities that affect the optical properties of the scintillator. Filtration with a 0.05 μm particulate filter removed suspended dust particles that may contain U, Th and K. Nitrogen stripping was employed to remove dissolved water and noble gases (e.g. ^{85}Kr) from the scintillator. The system has effectively reduced the internal back in the 5 ton liquid scintillation detector from 470±90 events/d to the detection limit of the detector (±45 events/d) in the energy window $250<E<800$ keV.

1. INTRODUCTION

Very low backgroud detectors are required to identify rare events, including solar neutrinos, searches for dark matter, double beta decay events, and searches for proton decay. Liquid scintillation detectors employed for these searches require a large mass (~100 tons) of scintillator with extremely low radioactivity (<10 nBq/ton). Liquid scintillation will detect all radioactive decays, which necessitates removal of radioactive sources in the scintillator. To achieve this high radiopurity standard chemical separation processes may be applied on-line to the scintillator.

The concept of on-line purification has been tested with a 5 ton liquid scintillation counting test facility (CTF) at the Laboratori Nazionali del Gran Sasso (LNGS). The principle objective of the CTF is to demonstrate the technical feasibility of purifying liquid scintillator to a level adequate for the Borexino solar neutrino experiment.

The CTF contains the liquid scintillator within a 2 m diameter sphere constructed of 0.5 mm thick nylon. The scintillator containment vessel is suspended within 1000 tons of ultrapure water shielding. Scintillation light is detected with 100 phototubes with light collectors mounted in the water viewing the scintillator vessel. The liquid scintillator tested in the CTF was a solution of 1.5 g/L of PPO (2,5 diphenyloxazole) in pseudocumene (1,2,4 trimethylbenzene). A description of the CTF, and the data acquisition and analysis in the CTF is published elsewhere [1,2].

2. RADIOPURITY REQUIREMENTS FOR A SOLAR NEUTRINO DETECTOR

Borexino is a solar neutrino experiment designed to detect the ^{7}Be neutrino flux by neutrino-electron scattering in a large scale liquid scintillation detector. The monoenergetic ^{7}Be neutrino signature will appear as a step decrease in the intensity of the energy spectrum at 665 keV. Based on the standard solar model (SSM) the count rate for the ^{7}Be neutrinos is 0.5 events/day/ton-scintillator within the energy window 250 - 800 keV [3,4]. The principal impurities anticipated in Borexino are summarized in Table 1. The concentration of ^{14}C, which decays by β-emission with an end point of 156 keV, must be below 10^4 atoms/ton scintillator to avoid a pileup on the phototubes giving accidental coincidences. ^{14}C and ^{3}H are part of the molecular structure of the scintillator and cannot be removed; the scintillator components are chosen to be derived from petroleum where millions of years in subterranean rock formations has allowed these to decay. Phase separation processes, such as extraction or distillation can remove impurities that form homogeneous solutions with the scintillator, such as soluble organometallic complexes. Lastly, dust particles in the colloidal range of 2 - 2000 nm may contain a variety of radioactive impurities such as ^{238}U, ^{232}Th and ^{40}K. Larger particles can be removed by filtration, whereas phase separation processes may more effectively remove the smaller particles.

3. PURIFICATION SYSTEM FOR THE COUNTING TEST FACILITY

The purification of the scintillator focused on removal of dust, potassium from the fluor, and radon and its progeny. To minimize problems associated with Rn emanation, materials of construction were chosen to have a low U content and low permeability to noble gases. ^{210}Pb from the decay of ^{222}Rn may be deposited on the surface of the scintillator containment vessel from exposure to radon. This surface contamination is a continual source of radioactivity, which can be identified by positional reconstruction of the scintillation events. However, minimizing the surface contamination can improve the sensitivity of the detector, so during construction of the scintillator containment vessel exposure to air was kept to a minimum to keep the ^{210}Pb deposits to a minimum.

The scintillation efficiency and attenuation of the emitted light during transit through the large volume of the scintillator are also critical for a large liquid scintillation detector, and any purification methods for improving radiopurity of the scintillator must not degrade the optical properties of the scintillator [5,6]. The optical attenuation length at 420 nm, as measured with a visible spectrophotometer, was used as an indication of the optical clarity of the scintillator to indicate any degradation of the scintillator.

The general design of the scintillator purification system employed distillation and water extraction followed by filtration and nitrogen stripping. Distillation and water extraction were proven effective for removing ^{7}Be impurities from liquid scintillator components in the most sensitive laboratory experiment [7].

Table 1
Radioactive Impurities in Liquid Scintillators

U, Th and K - most likely source is suspended dust (siliceous particles).
U and Th exist at ca. 10 ppm in the earth's crust
K is 2.4% of earth's crust

$$\frac{1\ \mu g\ dust}{ton\ \text{scintillator}}\ \text{produces}\ \begin{cases} 10 \quad events/da \quad for \quad U \quad and \quad Th \\ 100 \quad events/da \quad for \quad K \\ in \quad 100 \quad ton \quad fiducial \quad volume \end{cases}$$

K -Neutron activation measurements have shown substantial potassium contamination in the fluor (PPO), K ~ 10^{-6} g/ g-PPO in as purchased material. At the design concentration level of 1.5 g PPO/Liter-PC the acceptable level of K in PPO is 10^{-11} g-K/g-PPO.

^{85}Kr - 10.7 yr half life contained in air at ~1 Bq/m^3
1 liter air exposure produces a background of 75 events/day

^{7}Be - Produced cosmogenically from ^{12}C, with a 55 day half life.
Equilibrium exposure at surface will produce a background of 50,000 events/day
that will require 1 ½ year to decay to <10 events/day

^{222}Rn - Contained in underground air of LNGS at ~100 Bq/m^3 and underground water at ~10,000 Bq/m^3. Rn emanation from materials of construction will also result from U(Ra) decay chain. Rn will decay away with 5.5 day decay time, but will leave ^{210}Pb and its daughters ^{210}Po and ^{210} Bi)with a 22 year half life.

Water extraction was preferred for on-line purification. It could be applied to the scintillator mixture without altering its composition, it could be done at room temperature avoiding any thermal degradation of the scintillator and avoids potential hazards associated with heating pseudocumene above its flash point (43°C). Water extraction is not effective at romoving chemical impurities that may effect the optical properties of the scintillator, and it may result in potential problems associated with temperature fluctuations.

When contacted with water during extraction the scintillator becomes saturated with water. The solubility of water in pseudocumene at 20°C is approximately 50 ppm, and decreases with decreasing temperature. Decreasing the temperature of a water-saturated scintillator results in water condensation, making the scintillator cloudy. To avoid water condensation the water is stripped from the scintillator after water extraction. Water has a higher vapor pressure than pseudocumene, and may be stripped from the scintillator by flowing dry nitrogen through the scintillator. The water content in the scintillator is reduced by 90% in a single equilibrium stage at 15°C with a mass flow ratio of nitrogen to scintillator of 0.04 (0.03 m^3 gaseous N_2 per liter of scintillator). The nitrogen stripping operation has two additional benefits. It strips oxygen from the scintillator, whose presence quenches the scintillation. Second, the nitrogen strips soluble radioactive gas impurities from the scintillator including ^{222}Rn and ^{85}Kr.

Distillation of the scintillator mixture is more complicated. In a batch distillation the PPO/pseudocumene mixture will be depleted of the less volatile PPO. With a continuous feed it is possible to keep a high concentration of PPO in the liquid of the boiler vessel, such that the equilibrium vapor composition above the liquid corresponds to 1.5 g PPO/L-pseudocumene. By maintaining a concentrated PPO/pseudocumene solution in the boiler vessel the scintillator was continuously distilled with negligible loss of PPO. The concentration of PPO required was dependent on the temperature and pressure in the system. Typically the distillation was carried out at pressures of ~ 100 torr at 125°C, where the liquid in the bottom of the boiler vessel had a composition of circa 500 g PPO/L pseudocumene (PPO and pseudocumene are totally miscibile at 125°C).

The general strategy adopted for on-line scintillator purification was to employ water extraction followed by nitrogen stripping. Vacuum distillation would be used intermittently to remove chemical impurities that may degrade the optical properties of the scintillator, as well as removing the radioactive impurities.

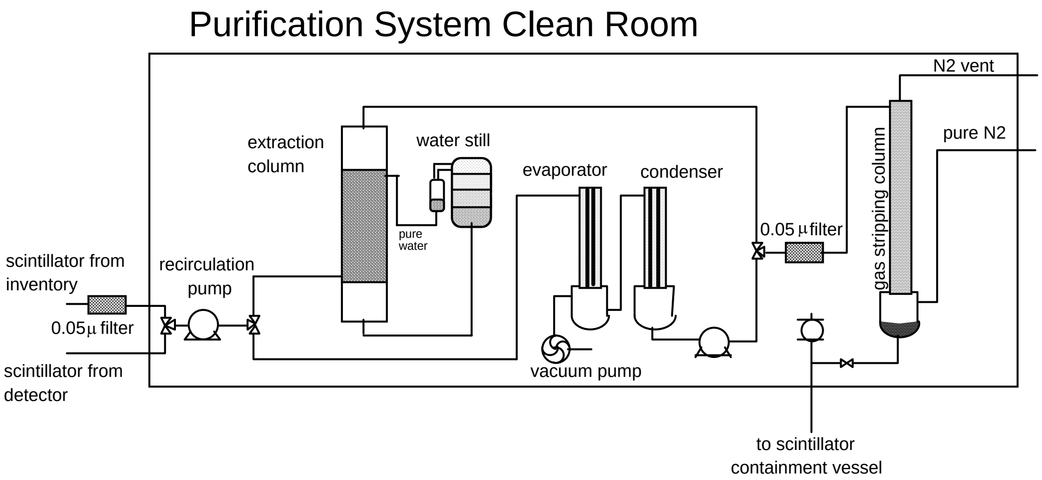

Figure 1. Schematic of the purification system in the counting test facility. The scintillator is circulated from the scintillator containment vessel in the detector through either counter current water extraction, or vacuum distillation. After water extraction or distillation the scintillator is filtered through a 0.05 μm filter and stripped of water and dissolved gases in a nitrogen stripping column. The scintillator is then returned to the detector containment vessel.

The inside of the CTF purification plant. The pipe in the center leads down to the scintillator containment vessel. The water extraction column is to the right rear of the photo. The nitrogen stripping column is in the foreground on the right of photo. At the bottom of the stripping column is the liquid holding vessel where the scintillator level is controlled to fix the liquid pressure head on the scintillator containment vessel. The distillation subsystem is to the left. The steel columns going from the ceiling down are the falling film evaporator and condenser. Near the floor at the rear center and to the front left are pumps used for moving the scintillator within the purification plant. The vessels and piping has all been insulated to minimize heat losses.

Since dust particles may introduce U, Th, and K, particulate matter is a serious concern as a source of radioactive impurities. To minimize particulate matter stainless steel vessels and tubing were first mechanically polished to an Ra 15 finish (so-called non-particulating surfaces) followed with electropolishing and final cleaning with ultrapure water and nitrogen in clean-room conditions. In addition, the scintillator was passed through a teflon 0.05 micron particulate filter (the finest available), prior to the nitrogen stripping.

The scintillator is contained within a nylon vessel suspended in the water shield of the CTF detector. The scintillator is purified on-line by removing scintillator from the containment vessel, passing it through the purification system and returning it to the containment vessel. The scintillator volume is 5000 L and the total volume of scintillator within the purification system is less than 50 liters, so the detector volume does not change significantly during purification. Since the purified scintillator is mixed back in with the scintillator in the active detector the removal of impurities decreases approximately exponentially with a time constant equal to the residence time of the system (residence time = [volume of detector]/[volumetric flow rate of purification system]). For a flow rate of 50 liter/hr the residence time is 100 hours, so the time constant for reduction of impurities in the CTF is approximately 4 days.

A schematic diagram of the purification system for the CTF is shown in Figure 1. Detailed process and instrumentation diagrams (PID drawings) are available elsewhere [8,9].

4. PERFORMANCE OF THE PURIFICATION SYSTEM

The computer control of the processes providing a trouble free automated system operating 24 hours a day with a minimum of operator attention. The selection of equipment, materials, and cleaning procedures was judged satisfactory. We were unable to detect radon emanation from any of the vessels and piping in the purification system, and could only set an upper limit of radon emanation of 0.02 Ra/m^2/d. The only significant problem encountered was radon contamination of the scintillator by the nitrogen stripping operation due to a radon leak in the boil off nitrogen system. In our efforts to identify the radon the entire purification system was made vacuum tight, and not merely fluid tight, as is normally the case for such systems. By thorough leak checking with a helium leak detector, and modification of some seals, the entire system was sealed to the equivalent of a high vacuum system.

After the final vessel filling, the scintillator was undisturbed for several weeks to allow radon to decay, and evaluate the ^{238}U and ^{232}Th backgrounds, and the ^{14}C background. The ^{222}Rn eventually reached a constant level of 1.5 ± 0.6 counts/day, as determined by measurement of the ^{214}Bi-^{214}Po beta-alpha delayed coincidences (236 ms mean time), corresponding to a ^{238}U concentration of (3.5±1.3) x 10^{-16} g/g-scintillator. The ^{232}Th concentration based on the ^{212}Bi-^{212}Po delayed coincidence (432 ns mean time) was 4.4 +1.5/-1.2 x10^{-16} g/g. The ^{14}C spectrum in the range 50-150 keV was fit to a simulation. From the total area of the ^{14} C spectrum the ratio of ^{14}C to ^{12}C was $\frac{^{14}C}{^{12}C} = (1.85 \pm 0.13) \times 10^{-18}$ [10].

4.2. Nitrogen Stripping:

Nitrogen stripping of the scintillator reduced the water content in pseudocumene to < 20 ppm. Nitrogen stripping in the CTF also removed ^{85}Kr, and reduced the total background. After the initial filling the presence of ^{85}Kr was detected by a time correlated events along a minor decay branch (0.43% branching ratio).

$$^{85}Kr \xrightarrow{15.6y} {}^{85}Rb^{*} + \beta(173keV)$$

$$^{85}Rb^{*} \xrightarrow{0.146\mu s} {}^{85}Rb + \gamma(514keV)$$

Along this branch 1.3±0.2 events/day were detected indicating a total ^{85}Kr background of 300 events/day (185 events/day have energies that fall in the 250-800 keV neutrino window). This level of ^{85}Kr was consistent with the Kr from 4 liters of air contacting the scintillator. Nitrogen stripping of the scintillator was carried out for 4 cycles (1 cycle refers to processing 5 m^3 of scintillator), with a scintillator flow of 30 liter/hr (25.8 kg/hr) and a nitrogen flow of 2.5 m^3/hr (3.1 kg/hr). After the nitrogen stripping operation the delayed coincidence of the ^{85}Kr decay decreased to 0.24±0.2 events/day, and the background count rate internal to the scintillator containment vessel was reduced from 470±90 events/day to 285±70 events/day. The reduction of the internal background was slightly greater than the estimated level of ^{85}Kr, suggesting other background impurities, such as 39Ar may have been reduced as well.

An increase in the radon level during nitrogen stripping indicated a radon leak. Since the radon was not accompanied by ^{85}Kr, but was accompanied by thoron the radon must have result from emanation - and not an air leak. Tests with the boil-off nitrogen system suggested that the radon emanation came from a source midway down in the tank, which could have been a welded joint or perhaps residual dust or dirt. By keeping the liquid level in the nitrogen boil off nearly full the rise in background due to radon was minimal.

4.2 Water Extraction

Water extraction of the scintillator was carried out for a total of five cycles with a scintillator flow of 30 liter/hr (25.8 kg/hr). The nitrogen stripping operation continued at the same conditions as outlined above.

Energy spectra of the backgrounds internal to the scintillator vessel before and after the water extraction are shown in Figure 2. The spectrum after the water extraction had a featureless decaying background. The increase in the background at low energy results principally from the tail of the ^{14}C peak. The spectrum prior to water extraction shows a distinct peak at 395 keV. The 395 keV peak has been identified as the alpha particles from the decay of ^{210}Po to ^{206}Pb.

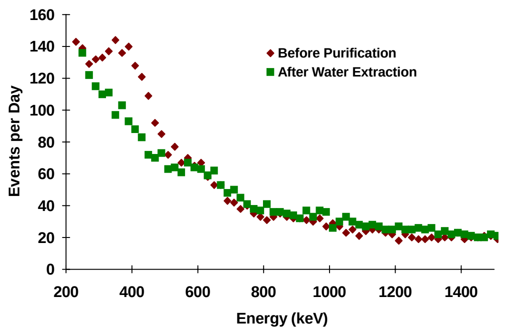

Figure 2. Spectral background of the CTF (■) before and (◆) after water extraction. Spectra correspond to events reconstructed internal to the scintillator containment vessel.

4.3. Distillation

The distillation system ran three complete cycles operating at 12 liter/hr (10.3 kg/hr). Samples drawn at routine intervals during the distillation process showed the PPO level in the scintillator was kept constant to within 1%, and the optical attenuation length at 420 nm was maintained at 7.5 m. Analyses of the background in the scintillator containment vessel after the distillation showed a small decrease in the ^{210}Po signal and a small decrease in the internal background rate, as shown in Table 2.

Table 2
Backgrounds* in the CTF During Purification Operations

Time Period	Total Background (events/day)	Total Internal Background (events/day)	^{85}Kr (events/d)	^{210}Po (events/d)
Initial filling	2595±215	470±90	300±45	250±40

After nitrogen stripping	2325±220	285±70	25±25	265±62
After water extraction	2075±195	21±47	25±25	<40
During distillation			50±25	0
After distillation	2005±195	35±67	25±25	0

*events in the energy range of the neutrino window, 250-800 keV

5. CONCLUSIONS

Purification of a large-scale liquid scintillator suitable for the Borexino solar neutrino detector has been demonstrated. A pseudocumene/PPO scintillator was purified in place achieving backgrounds corresponding to <10 events/day in the energy window 250-800 keV. Distillation, water extraction, filtration and nitrogen stripping were all demonstrated to be effective at removing the radioactive impurities they were designed to handle. All the construction materials internal to the purification plant (electropolished stainless steel and teflon) were found to be acceptable for handling high purity scintillator components.

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