Regular Article – Experimental Physics

Large-scale liquid scintillation detectors for solar neutrinos⋆

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Abstract. Large-scale liquid scintillation detectors are capable of providing spectral yields of the low energy solar neutrinos. These detectors require > 100 tons of liquid scintillator with high optical and radiopurity. In this paper requirements for low-energy neutrino detection by liquid scintillation are specified and the procedures to achieve low backgrounds in large-scale liquid scintillation detectors for solar neutrinos are reviewed. The designs, operations and achievements of Borexino, KamLAND and SNO+ in measuring the low-energy solar neutrino fluxes are reviewed.

1 Introduction

The detection of solar neutrinos has evolved over the past 60 years. Borexino, KamLAND and SNO+ are the third generation of Solar Neutrino Detectors. They are large-scale liquid scintillation detectors that have the sensitivity to obtain real time spectral resolution of low energy neutrino fluxes including the pp and CNO solar neutrinos. They are real-time, calorimetric, and spectral detectors that are unraveling the details of solar neutrino physics.

The first generation solar neutrino experiments were radiochemical detectors. Homestake measured the integrated flux of solar neutrinos with $E_\nu > 814$ keV based on the production of ^{37}Ar from ν_e capture by ^{37}Cl [1, 2]. Gallex and Sage (based on ν_e capture in ^{71}Ga to produce ^{71}Ge) extended observations of a solar neutrino deficiency over the energy range $E_\nu > 233$ keV. Homestake, SAGE and Gallex detected the integral neutrino rate with energies above specific thresholds by radio-chemical methods [3, 4]. The cross-section for neutrino capture rates is small which necessitated time integrated measurements. The detector needed to be shielded from high-energy cosmic radiation, but fortunately the radio-chemical detectors are largely insensitive to low-energy α, β and γ radiations from natural radioactivity.

Kamiokande and its successor Super Kamiokande, represented the second generation solar neutrino experiment. They obtained real time fluxes of the high energy ^{8}B neutrinos by detecting the Čerenkov radiation produced by electron-neutrino scattering [5–7]. The ^{8}B neutrinos

have energies > 5 Mev which produce detectable Čerenkov light. The ^{8}B neutrino flux measured by Kamiokande showed a clear deficit compared to the standard solar model [8–10].

The Sudbury Neutrino Observatory (SNO) was also a Čerenkov detector. It employed heavy water (D$_2$O) to measure the charged current reaction D(ν, e$^+$)nn induced by high-energy ^{8}B neutrinos [11, 12]. The positron was detected by Čerenkov radiation. SNO also measured the neutral current reaction D(ν,ν)pn by detecting the neutrons. The charged and neutral current measurements demonstrated that the deficit of neutrinos detected in the early solar neutrino experiments was due to neutrino oscillations [12].

Large-scale liquid scintillation detectors are also capable of detecting temporal variations in the solar neutrino spectrum extending the low energy cutoff to ~ 250 keV. But, the ubiquitous backgrounds from α, β and γ radiation due to natural radioactivity, most of which appear at energies < 2 MeV, are also detected by liquid scintillation detectors. To detect solar neutrinos by liquid scintillators the background from radioactive impurities should be reduced to be less than solar neutrino signal.

Borexino was proposed in 1990 as a ^{11}B loaded liquid scintillation detector to probe the ^{8}B neutrino flux [13]; but Borexino changed its objective to be a low-energy scintillation detector to measure the ^{7}Be solar neutrino flux [14]. Detection of solar neutrinos requires a large mass (> 100 tons) of scintillator with extremely low radioactivity (< 10 nBq/ton). Funding agencies requested a demonstration of technology that could produce a scintillator with purity of ^{238}U and ^{232}Th $< 5 \times 10^{-16}$ g/g-scintillator on a scale of > 1 ton. The Borexino collaboration built the Counting Test Facility (CTF) that achieved those goals in a non-segmented 4 ton liquid scintillation detector with

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on-line scintillator purification [15]. The full scale Borexino solar neutrino detector with a fiducial mass of 100 tons scintillator was constructed from 1999 to 2005 and data acquisition commenced in 2007.

KamLAND was built in 1997 as a large-scale liquid scintillation detector to study neutrino oscillations from reactor anti-neutrinos. In 2002 KamLAND upgraded their detector to measure the ^{7}Be solar neutrino flux [16]. KamLAND introduced a scintillator purification campaign to reduce radioactive impurities that interfered with the low energy neutrino window (200-800 keV). The KamLAND collaboration recently reported a measurement of the ^{7}Be solar neutrino flux [17].

SNO+ was conceived as a large-scale liquid scintillator that repurposed the SNO heavy water Čerenkov detector [18]. The SNO+ detector is deeper underground than Borexino or KamLAND and will have lower cosmic ray background. Its primary focus is the measurement of double beta decay. It will also be used for detection of reactor neutrinos, geoneutrinos and solar neutrinos. SNO+ is in the final stages of commission and should be taking data in 2015–2016 [19].

Two other scintillation detectors for solar neutrinos are in the proposal phase. LENS (Low Energy Neutrino Spectroscopy) detects pp solar neutrinos in an Indium loaded liquid scintillator via a triple coincidence of an energetic electron followed by two delayed γ-rays [20]. The triple coincidence greatly reduces almost all background interference. LENA (Low Energy Neutrino Astronomy) is proposing a 50000 ton liquid scintillation detector in Finland for low energy solar neutrinos among other objectives [21].

Large-scale liquid scintillator detectors require purifying the scintillator for both radio-purity and optical purity. The scintillator components, the scintillator purification, the detector construction and the scintillator handling must be integrated for the detector to be successful. Borexino, KamLAND and SNO+ have addressed system integration issues. In this chapter we will review the operation of large-scale liquid scintillation detectors for solar neutrinos.

2 Liquid scintillators

Liquid scintillators detect solar neutrinos via neutrino electron (ν-e$^-$) scattering. The initial goal for Borexino and KamLAND was the mono-energetic ^{7}Be-ν line at 0.862 MeV; the profile of the recoil electrons is a unique "flat box" with a spectral (Compton) edge at 0.66 MeV. In principle, the profile is slightly different for ν_e and $\nu_{\mu,\tau}$, however, the difference is too small to be resolved. The ^{7}Be-ν signal window is (0.25–0.8) MeV, set mainly by background considerations (^{14}C). In this window, the nominal scattering cross-section is $\approx 5 \times 10^{-44}$ cm^2 (at ≈ 1 MeV). Thus, for a 100 ton detector and a neutrino flux $\phi_\nu(^7\text{Be}) = 4.8 \times 10^9$ cm^{-2} s^{-1} [22], an event rate of ≈ 50/da is expected (assuming no neutrino oscillations).

2.1 Mechanism of scintillation

Liquid scintillators are comprised of an aromatic solvent, a primary fluor, optional secondary fluors and photomulti-plier tubes. Ionizing-radiation, such as neutrinos, scatters electrons. These scattered electrons dissipate their energy by collisions with solvent molecules. Aromatic molecules have a high cross-section for excitation of π-π^* transitions. The electronically excited solvent molecules transfer their energy molecule to electronically excite fluor molecules. Energy transfer between solvent and fluor occurs by both radiative and non-radiative process. The excited fluor molecules dissipate a fraction of their energy into molecular vibrations (heat). When the excited fluor molecules then relax to their ground state they emit photons which are detected by photomultiplier tubes.

A detailed description of the photo physics associated with liquid scintillators can be found in the monograph by Knoll [23]. Ranucci provides more discussion of the physics of the Borexino scintillator [24].

2.2 Chemical properties of scintillator components

Choice of liquid scintillator components for a large-scale detector is based on several factors:

1) Optical properties of the scintillator components
2) Cost and availability of scintillator components.
3) Materials of construction and chemical compatibility with scintillator components.
4) Ability to purify the scintillator components.
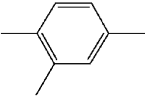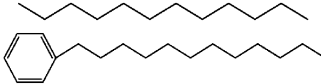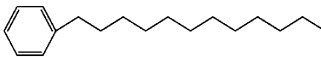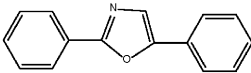5) Phototube characteristics.

The scintillator solvent must have: 1) high aromatic content; 2) good optical clarity at the wavelength of the propagated and detected light; 3) chemical stability and compatibility with confinement over long periods of time; 4) acceptable cost and availability. The ability to purify the scintillator solvent is also recommended for long term operation.

Most liquid scintillators are based on methylated benzene derivatives: toluene, xylene, pseudocumene (PC). Scintillator components used by Borexino, KamLAND and SNO+ are shown in table 1. Borexino chose PC as its scintillator solvent. PC is readily available from petroleum refining operations. It has good purity and can be repurified by distillation and/or water extraction.

Freshly distilled PC has good optical clarity, its optical attenuation length is > 7 m at 375 nm. PC is stable in the absence of oxygen; when exposed to oxygen and metal surfaces PC is slowly oxidized turning the solvent yellow and reducing the optical attenuation length. The optical attenuation length of freshly distilled PC is 7.4 m at 375 nm [25]. If the PC is kept free of oxygen and contaminants it retains good optical clarity indefinitely. However, laboratory tests showed exposure to oxygen from the air and stainless steel at room temperature for 3 weeks reduced the optical attenuation length to < 2 m at 375 nm [26].

Benzene derivatives are toxic and flammable; they must be handled with care. Borexino designed their fluid handling system to always have the solvent in vacuum tight vessels and piping under a nitrogen blanket to assure safe-handling [27]. An inert blanket, such as nitrogen, was important for maintaining the optical clarity of the Borexino scintillator over periods of several years.

Table 1. Scintillator solvents and fluor.

	Chemical structure	Normal Boiling point	Flash point
Pseudocumene (1,2,4 trimethyl benzene)		170 °C	44 °C
Dodecane (C12H16)		214 °C	71 °C
Linear Alkylbenzene		270–320 °C	130 °C
PPO (2,5 diphenyl oxazole)		∼ 350 °C	> 150 °C

KamLAND employed a mixture of a low volatility alkane, *e.g.* dodecane, and PC. The dodecane has no C=C double bonds. It has a longer attenuation length than PC between 280–350 nm, and it is less susceptible to chemical degradation. The mixed solvent in KamLAND has an optical attenuation length of ∼ 15 m of at 360 nm, compared to ∼ 8 m for neat PC. The mixed solvent in KamLAND has a higher flash point for potential combustion than neat PC.

SNO+ proposes to use single component solvent, linear alkyl benzenes (LABs). LABs are feedstocks for detergents making them almost as economical and available as PC. The LABs are less volatile and less toxic than the simple aromatics. LABs are a less aggressive solvent for PMMA (poly methyl methacrylate) than PC, making LABs more suitable to use with the acrylic sphere in SNO+. The LABs may be likened to fusing the dodecane and PC from KamLAND into a single molecule.

2.3 Scintillator fluors

Fluors red shift the wavelength of scintillation light to increase light transmission and improve photon detection. The three key elements for the primary fluor are: 1) matching light absorption of the primary fluor to the light emission by the aromatic solvent; 2) having a large Stokes shift between light absorption and light emission; 3) matching the wavelength of light emitted by the primary fluor to the wavelength sensitivity of the photomultiplier tube. A high overall quantum efficiency of light absorption and emission at longer wavelength by the fluor improves the scintillator sensitivity.

Figure 1 shows the absorption and emission spectra for scintillator solvent, PC, and scintillator fluor, PPO, used in Borexino and KamLAND. The electronic excited states in PC relax by loss of vibrational energy causing emission to be red shifted from absorption (Stokes shift). The Stokes shift for PC is relatively small. The overlap of the emission and absorption results in the self-attenuation of the primary scintillation light. Primary fluors, such as
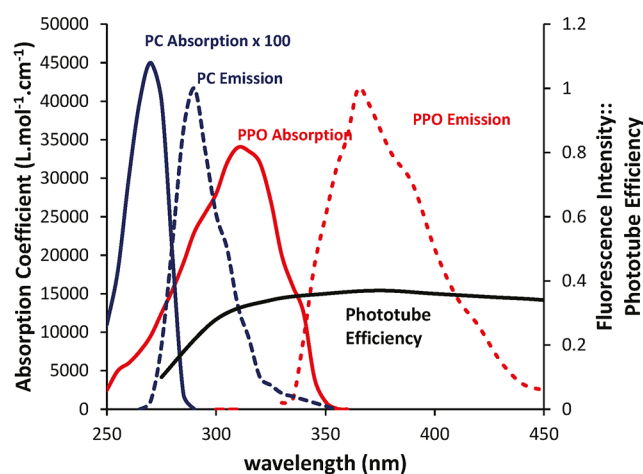

Fig. 1. Absorption and emission spectra for PC and PPO. The phototube efficiency is for Thorn EMI phototubes used in the Counting Test Facility.

PPO (2,5 diphenyl oxazole) have a larger Stokes shift with less overlap between the absorption and emission spectra. Secondary fluors, such as bis-MSB (1,4-Bis(2-methylstyryl) benzene) can be used to shift the emitted photons to even longer wavelength.

PPO absorbs radiation at the wavelength of PC emission and then emits photons at longer wavelengths that reduce self-attenuation from PC and PPO. The photons emitted from the PPO are transmitted through the scintillator and the glass of the photomultiplier tube. As shown in fig. 1 the absorption coefficient for PPO is a factor of 100 greater than the absorption coefficient of PC. A low concentration of PPO is sufficient to absorb the photons emitted from PC and re-emit them at longer wavelength. As shown in fig. 1 the wavelength for maximum PC emission is 365 nm, which closely matches the Borexino phototube response [28].

Scintillation light emanates in all directions. After emission light can be absorbed and re-emitted. Absorption

and re-emission alter the timing distribution of the photon arrival times to the phototubes. Florescence decay, light yield and scattering have to be quantified for the choice of scintillator mixture employed in each detector. Light scattering and reflections in the CTF were quantified using a window source, in which a shield limited the solid angle of the detector directly illuminated [29]. Light arriving at the shielded phototubes revealed that in the CTF 28% of the light is scattered over a distance of 1 m in the scintillator. *Ex situ* and *in situ* source experiments were employed to measure absorption and scattering coefficients and fluorescence decay times for Borexino [30]. Similar measurements were undertaken by KamLAND as well. KamLAND reported light yields were 20% greater than initially expected as a result of absorption-reemission [16].

2.4 Scintillator event discrimination

Liquid scintillators are sensitive to any type of ionizing radiation, including beta particles, alpha particles, gamma rays, muons, geoneutrinos, etc. Discrimination of scintillator excitation by non-neutrino sources is important to improving the sensitivity of the liquid scintillator to detect the low-energy solar neutrino spectrum.

The decay times from α-particle excitation are longer than for β-particle excitation. Pulse shape discrimination can tag α-events with 90–99% efficiency, depending on the composition of the scintillator and on the α-energy [31, 32]. Time correlated $\alpha - \beta$ scintillation events from ^{214}Bi-^{214}Po and ^{212}Bi-^{212}Po were used by both Borexino and KamLAND to identify the contributions of ^{238}U and ^{232}Th to the detector backgrounds. The time correlated $\alpha - \beta$ Bi-Po tags are particularly useful in tracing the origins of persistent leaks of Rn, either from air laden with Rn or Rn emanation from materials of construction [33].

The scintillation light yield and decay times from excitation by γ-ray and α-particle excitation were obtained in laboratory samples for the CTF [34] and KamLAND [35, 36]. The light yield of the PC-PPO scintillator used in the CTF was approximately 10^4 photons/MeV of energy deposited by fully relativistic particles such as β-rays and secondary electrons from γ-rays. For slower particles, the photon yield decreases [23]. The yield for 7.7 MeV α-particles from ^{214}Po decay is approximately 1000 photons/MeV. Scintillation from α-excitation was distinguished from β-excitation from the long time fluorescence tail [23, 37, 38].

3 Purity requirements

The standard solar model without neutrino oscillations predicts the ^{7}Be in Borexino should be 50 events/da in a 100 ton fiducial volume. Ideally the neutrino count rate should be 10–100 times the background rate in the detector. The required backgrounds are so low that only the CTF, Borexino or KamLAND detectors can measure the necessary backgrounds. In order to guide development of a large scintillation detector the potential background rates

in the scintillator were estimated assuming likely chemical forms of the impurities. Table 2 summarizes various radioactive impurities and the scintillation count rate that might be encountered without purification or special handling. Strategies tested for reduction of the radioactive impurities are also indicated in table 2.

The ^{14}C concentration in the scintillator limits detection of the pp neutrino. The ^{14}C β-decay has an endpoint of 156 keV, but finite energy resolution and pile-up creates a tail in the β energy spectrum that extends to higher energy and can interfere with the pp edge at 250 keV. The ^{14}C/^{12}C should be $\leq 10^{-18}$ for detection of the pp solar neutrino flux. Modern carbon of biological origin has an isotopic ratio ^{14}C/^{12}C of about 10^{-12}. At the earths surface ^{14}C is continuously produced from ^{14}N, via the reaction ^{14}N(n, p)^{14}C. The carbon is incorporated into organic matter via photosynthesis. Petroleum deposits have been shielded from cosmic radiation deep underground and are millions of years old. Consequently, most of the ^{14}C present in the petroleum precursors has decayed away; a small residual concentration persists formed by underground nuclear reactions that produce ^{14}C from a sequence of α-decay and (n,p) reactions. The Borexino collaboration determined the abundance of ^{14}C in a petroleum-derived scintillator to be ^{14}C/^{12}C $\sim 10^{18}$ [39]. The ^{14}C levels is dependent on the geology of the crude oil source that was employed to make the scintillator solvent and may vary by an order of magnitude.

Cosmogenic derived ^{7}Be is formed by the reaction of cosmic neutrons and protons with ^{12}C. It has a half-life of 53 days. Secular equilibrium at the earths surface would produce a background of 10^5 events/da. The Borexino group showed that ^{7}Be could be removed by distillation or multistage water extraction [40]. The Borexino collaboration also made special effort to move the scintillator solvent from the refinery in Sardinia to the underground laboratory as fast as possible to minimize the formation of cosmogenic ^{7}Be in the scintillator.

Other radioactive impurities enter the scintillator from the air or dust. These impurities are reduced by a three prong attack. 1) The scintillator handling and containment systems must be cleaned to minimize dust and remove surface contamination due to radon decay daughters. 2) The scintillator containment and handling system must be leak tight to keep out radioactive gas impurities, especially ^{85}Kr. 3) The scintillator must be purified to remove pre-existing radioactive impurities. Large-scale liquid scintillators require extraordinary cleanliness and leak-tightness of the entire purification system. Three specific requirements were applied to Borexino.

1) Vacuum leak tightness for total system leak rate $< 10^{-5}$ mbar-L/s

2) Suspended particulate counts in liquids flowing through the system should be below IEST-STD-CC1246D level 30 [41, 42].

3) Washing with detergent and chelating agents followed by de-ionized (DI) water rinsing of all surfaces to reduce surface deposits.

Table 2. Radiopurity requirements for low-energy scintillation Detectors (estimates based on Borexino Detector).

Radio-isotope	Source	Typical level in scintillator without purification	Removal strategy	Level achieved in Borexino (< 1 cpd/100 ton)
^{14}C	Scintillator	^{14}C/^{12}C $\sim 10^{-12}$	Petroleum derivative (old carbon)	^{14}C/^{12}C $\approx 2 \times 10^{-18}$
^{7}Be	Cosmic ray activation of ^{14}C	2.7×10^3 cpd/ton corresponding to equilibrium for cosmic ray activation of ^{12}C to ^{7}Be at earths surface	Distillation and underground storage	< 0.01 cpd/ton
^{222}Rn and its progeny	Underground air Underground water	40 Bq/m^3 (2×10^7 cpd/ton) 50 Bq/m^3	Nitrogen stripping, Rn decay	$< 1\,\mu$Bq/m^3 (scintillator) $\sim 30\,\mu$Bq/m^3 (water)
^{238}U and its progeny	Suspended dust, stainless steel, nylon	10^4 cpd/ton (for the entire decay chain) $< 10^{-12}$g-U/g-scintillator 1 g-dust suspended in 1 ton of scintillator with 10^{-6}g-U/g-dust	Distillation, filtration	$< 10^{-18}$g-U/g-scintillator
^{232}Th and its progeny	Suspended dust, stainless steel, nylon	10^4 cpd/ton $< 10^{-12}$g-Th/g-scintillator 1 g-dust suspended in 1 ton of scintillator with 10^{-5}g-Th/g-dust	Distillation, filtration	$< 10^{-18}$g-Th/g-scintillator
^{40}K	Contaminant in fluor Underground dust	2700 cpd/ton (10^{-6}g-K/g-PPO) 1 ppm (10^{-6} g-K/g-dust)	Water extraction, filtration and distillation of fluor solution	$< 10^{-14}$g-K/g-scintillator
^{39}Ar	Air	200 cpd/ton Ar absorption into PC from air with ^{39}Ar $= 13$ mBq/m^3-air	Nitrogen stripping, Leak tight system	< 500 nBq/m^3-N$_2$
^{85}Kr	Air	4.3×10^4 cpd/ton Kr absorption into PC from air with ^{85}Kr $= 1.1$ Bq/m^3-air	Nitrogen stripping, Leak tight system	< 100 nBq/m^3-N$_2$

Vacuum leak tightness eliminated the influx of ^{39}Ar, ^{85}Kr and ^{222}Rn from air. Borexino designed and built the scintillator handling system (containment vessel, purification system, and filling system) to have a total leak rate of $< 10^{-8}$ m^3-air/hr. The leak rate specification was based limiting the total ^{85}Kr background to < 1 cpd in the 100 ton fiducial volume. It was assumed that all the Kr from any air leak was absorbed by scintillator during during detector filling. Air was assumed to have a ^{85}Kr content of ~ 1 Bq/m^3-air:

$$[\text{Air Leak Rate}] < \frac{\left(1\,\frac{\text{ton PC}}{\text{hr}}\right)\left(\frac{0.1\,\text{cpd-}^{85}\text{Kr}}{100\,\text{ton-PC}}\right)}{\left(10^5\,\frac{\text{cpd-}^{85}\text{Kr}}{\text{m}^3\text{-air}}\right)} \tag{1}$$

$$< 10^{-8}\,\frac{\text{m}^3\text{-air}}{\text{hr}}. \tag{2}$$

Micron-sized particulates are suspended in the air and in the flowing fluids due to Brownian motion. Assuming the dust particulates have concentrations of ~ 1 ppm of U and 10 ppm Th [43], the tolerable dust concentration is $\sim 10^5$ g-dust/ton-scintillator to keep the U and Th background < 1 cpd/100 ton-scintillator. Precision cleaning involving detergents, chelating agents and rinsing with filtered water can reduce the suspended particulate count to the tolerable level [44, 45]. During fabrication and assembly of the scintillator purification and fluid handling systems the internal surfaces of the system may be exposed to air containing ^{222}Rn. Radon decay deposits ^{210}Pb onto the exposed surfaces. ^{210}Pb accumulates and subsequently ^{210}Bi and ^{210}Po emanate from the surfaces as the ^{210}Pb decays ($t_{1/2}$ (^{210}Pb) $= 22$ yr) [46]. The ^{210}Bi creates a broad background from β-emission with an endpoint at 1.16 MeV ($t_{1/2}$ (^{210}Bi) $= 5$ d). ^{210}Po decays with a 5.4 MeV α-emission ($t_{1/2}$ (^{210}Po) $= 138$ d) that pro-

duces a scintillation peak at in the neutrino window at 425 keV (the light yield in the PPO/PC scintillator is reduced by 11 for α-particles compared to β-particles with the same energy [31, 34]). Precision cleaning reduces the amount of ^{210}Pb adsorbed on the surfaces of the purification system [47]. In Borexino all stainless steel surfaces were acid etched with nitric acid or a formic acid/citric acid mixture prior to precision cleaning to minimize the ^{210}Pb contamination. When any modifications to the purification system were done the exposed parts were all acid etched (metallurgists refer to this as pickling) and then precision cleaned.

4 Large-scale detectors

The liquid scintillation detectors for solar neutrinos are based on graded shielding with the scintillator at the center of a set of concentric shells of increasing radiopurity. Cosmic radiation is reduced by locating the detectors more than 1000 m below ground. The meters of water equivalent (mew) shielding for the three detectors are: KamLAND (2700 mew), Borexino (3800 mew) and SNO+ (6000 mew). SNO+ will have significantly better cosmic ray shielding, which reduces the background from the generation of ^{11}C [48]. The organic scintillator was confined to the central volume of all three detectors. The outer volume the detectors was filled with water providing shielding from γ-radiation emanating from the surrounding rock. The water shield also serves as a muon detector. Scintillation events are vetoed when they coincide with muon induced Čerenkov emission in the water shield.

The scintillator volume was sub-divided into buffer and active scintillator regions for both KamLAND and Borexino. The phototubes are located on stainless steel spheres that confine the buffer region. The buffer provided shielding from the high local scintillation rate created by the phototubes. The organic buffers in KamLAND and Borexino were density matched to the scintillator [16, 49]. Density matching allowed use of thin film containment vessels for the scintillator, which reduced the background arising from the scintillator vessel.

SNO+ will have a single volume of liquid scintillator contained within the 12 m diameter acrylic sphere originally used for the heavy water used in SNO. The phototubes are mounted on a geodesic sphere and view the acrylic sphere. The phototube structure and acrylic sphere are in a cavern filled with water. SNO+ has a thick wall acrylic container for the scintillator which can withstand the buoyant forces of the scintillator. The scintillator in SNO+ will be a linear alkyl benzene solvent with ~ 2 g-PPO/L-LABs as the primary scintillator [18]. The LABs have been chosen to minimize softening of the acrylic by PC.

KamLAND has 1200 m^3 of liquid scintillator consisting of a mixed solvent of PC and dodecane with 1.5 g-PPO/L-solvent. The scintillator is contained within a 13 m diameter vessel of a transparent polymer laminate. KamLAND has a buffer of a mixture of dodecane and an isoparaffin contained between the scintillator vessel and

an 18 m diameter steel sphere supporting phototubes to detect the scintillation light.

Borexino has three liquid volumes inside the water shielding tank [27]. A 13.7 m diameter stainless steel sphere located inside a water shielding tank confines an outer buffer and serves as a support structure for the photomultiplier tubes. The central scintillator volume is contained within an 8.5 m diameter thin nylon vessel containing 1.5 g-PPO/L-PC [49]. A second nylon vessel 11.0 m diameter contains a passive shield composed of pseudocumene and 5.0 g/L DMP (dimethylphthlate). DMP quenches the scintillation of PC to suppress the high scintillation rates caused by γ-emission from the photomultiplier tubes. Quenching the scintillation light in the buffer region reduces the background from accidental coincidences. The second nylon vessel acts as a barrier against radon and other backgrounds originating from the phototubes and light collectors mounted on the stainless steel sphere. The volume between the stainless steel sphere and the second nylon vessel is also filled with the DMP/PC buffer mixture.

Borexino, KamLAND and SNO+ decided upon slightly different strategies to achieve the purity necessary for low energy solar neutrino detection. The key engineering aspects of liquid scintillation detectors are discussed below. Borexino and SNO+ chose 2-component scintillators which simplified quality control over procurement, fluid handling and purification methods of the scintillator. KamLAND used a 2-component solvent and a single fluor. KamLAND diluted the PC with dodecane to increase the optical attenuation length and reduced the flammability limit of the scintillator.

Both KamLAND and Borexino separated the phototubes from the fiducial volume by using thin film balloon type confinement, and surrounding the phototubes with only solvent and no fluor. SNO+ achieves the same effect by having the phototubes in water outside the acrylic sphere.

The CTF revealed a significant and unexpected background from ^{210}Po. It was suspected that the ^{210}Po was released from surfaces contaminated with ^{210}Pb. Exposure of the surfaces that will contact the scintillator to air with ^{222}Rn can leave ^{210}Pb deposits on the internal surfaces. Decay of the ^{210}Pb can release ^{210}Po into the scintillator where it can decay to ^{206}Pb. Borexino developed Radon filters to provide low Radon air during construction of the nylon scintillator confinement vessels [49].

5 Purification methods for liquid scintillators

The challenge for purification of low background liquid scintillators is that, except for the noble gases, the molecular identity of the impurities is unknown. In the absence of specific chemical identity purification methods were chosen to be applicable to the most likely molecular form of the potential radioactive impurities. The impurities identified as potentially most troublesome for a low background scintillation detector were:
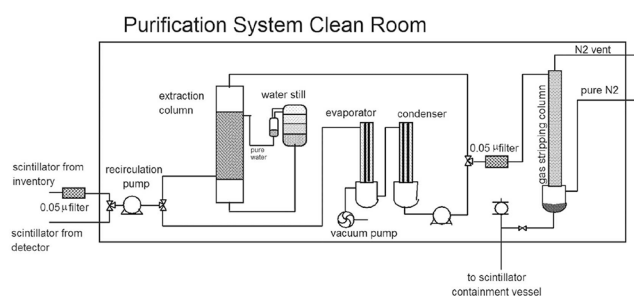
Fig. 2. Process flow schematic of the CTF on-line purification system. Reprinted from ref. [15] with permission from Elsevier.

1) micron and submicron particulates (dust) containing K, U, and Th
2) ^{222}Rn emanating from materials of construction
3) ^{85}Kr, ^{39}Ar and ^{222}Rn from air leaks
4) ^{210}Pb deposited on the surface of the scintillator containment vessel after exposure to ^{222}Rn (this produced background of ^{210}Po and ^{210}Bi)
5) ^{40}K in the fluor
6) partially oxidized aromatic chemical impurities that can reduce the optical quality of the scintillator

The Counting Test Facility of Borexino initiated the concept of on-line purification for liquid scintillators. Purification methods were chosen based on standard separations technology from the petrochemical industry. A process flow diagram for the on-line purification system used in the CTF is shown in fig. 2. The purification systems for Borexino, KamLAND and SNO+ have many of the same elements.

The CTF purification system was assembled and operated inside a class 1000 clean room. The clean room environment minimized particulates from entering the system during installation and maintenance. The control of particulates is important in the dusty underground environment of the large-scale scintillation detectors. SNO had also developed clean room techniques to prepare the heavy water using in their Čerenkov detector [50].

5.1 Filtration

Dust particles, primarily silicates and aluminates in the scintillator are a potential source of U, Th, K and other elements. The dust particles range in size from nm to microns. The CTF and Borexino employed commercial Teflon cartridge filters from Millipore or Pall with nominal cutoff size of $0.05\,\mu$m at the inlet and outlet of the purification system.

5.2 Water extraction

Water extraction is based on differential solubility of species in water and the organic scintillator. Charged impurities, such as metal salts and silicate dust particles
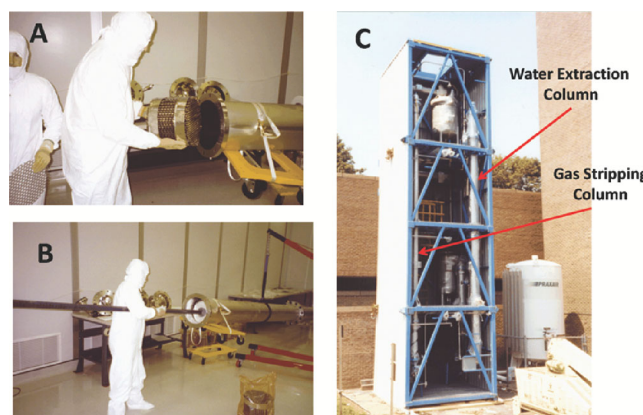

Fig. 3. Water extraction equipment for the Borexino scintillator. (A) Technician holding a section of structured packing used in the water extraction column. (B) Loading of the structured packing into the water extraction column. (C) One of the Borexino purification skids prepped for shipping to Gran Sasso. The water extraction column and the gas stripping columns are highlighted.

have a strong thermodynamic preference for polar solvents, such as water, compared to non-polar organic solvents. Water extraction works well for U, Th and K impurities in the scintillator.

Water extraction involves contacting the organic scintillator with purified water. Aromatic and aliphatic hydrocarbon liquids, such as PC, LABs and mineral oil are immiscible in water; the solubility of water in these solvents is $< 10\,$ppm by mass. When water and the scintillator are contacted, the impurities partition between the two phases. The partition coefficients for ionic species, such as K^+ or $UOCl^+$ may be as large as 10^6, which makes water extraction very efficient.

There are two essential process considerations for water extraction. (1) The contacting equipment must disperse the two liquid phases to create lots of interface for interphase mass transport. (2) There must be a continuous source of purified water to remove the impurities.

Good interfacial contact and mixing between the water and scintillator is achieved by passing the liquids countercurrent through a structured packing material. The structured packing, shown in fig. 3(A), breaks up and mixes the liquids flowing through the column. Water enters just below the top of the column and flows downward due to gravity. Scintillator has a density of $\sim 0.9\,$g$\cdot$cm^{-3} and enters just above the bottom of the column and flows upward. The sections of the column above the water feed and below the scintillator feed have no packing materials and serve as disengaging zones for water (at the bottom of the column) and scintillator (at the top of the column).

Countercurrent flow has the highest purity water contacting the purest scintillator product. In the CTF and Borexino the water for extraction is regenerated by distilling the water leaving the extraction column. A small amount of water, approximately 5–10%, containing U, Th and K impurities is disposed of from the water distillation.

Makeup water from a DI water plant is added to the water extraction feed to compensate for the water disposed of that contains the impurities.

Reuse and repurification of the water used in water extraction was important to achieve low background from ^{222}Rn and its daughters [51]. The Borexino collaboration observed deionization did not removed ^{222}Rn and the scintillator extracted the ^{222}Rn from the water. During the CTF operation the deionized water was stored for 30 days to permit the ^{222}Rn to decay, and then distilled and reused the water to minimize the introduction of ^{222}Rn.

A counter-current packed column was used for water extraction. The number of effective stages is a function of the diameter and height of the column, the type of packing and the flow rates of the water and scintillator. Engineering correlations were employed to design the extraction column. The Borexino water extraction column was designed to have approximately 4 theoretical stages at nominal scintillator flow rates of $1\,\mathrm{m^3\,hr^{-1}}$.

Water extraction can be applied to scintillator mixtures (solvent + fluors). It is carried out at ambient temperature. The main limitations of water extraction are: 1) it does not effectively remove chemical impurities that degrade optical clarity; 2) it does not remove Noble gas impurities ^{39}Ar and ^{85}Kr.

The scintillator leaving the water extraction column in saturated with water. If the temperature of the detector is lower than the temperature of the extraction column, water droplets will form in the scintillator. In the CTF and Borexino the water extracted scintillator was stripped by nitrogen to reduce the concentration of water dissolved in the scintillator before returning the scintillator to the detector.

5.3 Distillation

Distillation separates radioactive impurities having a different vapor pressure from the scintillator. Distillation is also effective at removing the chemical impurities that degrade optical clarity. PC/PPO scintillator was distilled on-line in the CTF. Borexino used distillation to purify the PC used in the scintillator and buffer regions during filling of the detector [51,52]. KamLAND performed on-line distillation of their scintillator [53]. SNO+ plans to use distillation to prepare their LAB solvent.

Distillation vaporizes a portion of a liquid mixture and then condenses that vapor back to liquid. The more volatile components are concentrated in the overhead vapor product and the less volatile species are concentrated in the bottoms liquid product. Metal salts and dust particles are low volatility, so the U, Th and K radioactive impurities are concentrated in the bottoms products of distillation. Partial oxidation of PC produces products that are less volatile than PC so distillation is also effective at removing chemical impurities that affect the optical clarity of the scintillator.

The PC/PPO scintillator mixture was distilled on-line in the CTF. Scintillator was evaporated at reduced pressure of 65 mbar and a temperature of $130\,^{\circ}$C in a falling film evaporator [25]. The vapor was condensed at $15\,^{\circ}$C.

Table 3. Purity requirements for nitrogen.

Impurity	Natural activity in air (Bq/m^3-air)	Purity requirement (Bq/m^3-nitrogen)
39Ar	$\sim 13 \times 10^{-3}$	40×10^{-9}
85Kr	1	10×10^{-9}

The reduced pressure distillation proceeded with smooth vaporization and avoided liquid drops being carried over from the evaporator to the condenser. KamLAND also employed reduced pressure distillation of their scintillator [53,54]. KamLAND employed three evaporators and condensers in their system.

Borexino distilled its scintillator solvent and fluor immediately before filling the detector. The Borexino system was designed to process $1\,\mathrm{m^3\,hr^{-1}}$ of PC in 6 sieve tray column that was $0.8\,\mathrm{m}$ diameter and $6\,\mathrm{m}$ tall [51]. The distillation column operated at $100\,^{\circ}$C and $100\,\mathrm{mbar}$. A bottoms product of 1–2% of the PC feed contained the radioactive impurities was discarded. SNO+ is also planning multistage columns for the LABs [57,58].

5.4 Gas stripping

Distillation and water extraction do not remove noble gas impurities (Ar, Kr and Rn) from PC. Borexino and KamLAND remove Ar, Kr and Rn impurities by gas stripping [16,51]. Borexino carried out gas stripping with counter current flow of the liquid PC and gaseous nitrogen in a column containing structure packing, similar to the packing used in the water extraction column shown in fig. 3. PC flows down through the structured packing as a thin film while N$_2$ flows upward counter-current to the PC. The liquid flows as a thin film which is important for effective stripping; diffusional mass transport of impurities to the liquid/gas interface is minimized with the thin liquid films. The effectiveness for gas stripping is a function of the ratio of the gas to liquid molar flow rates and the solubility of the Ar, Kr and Rn in PC [51].

The gas stripping packed column for Borexino was $0.15\,\mathrm{m}$ diameter and $10\,\mathrm{m}$ tall. It was designed to operate at a liquid flow of $900\,\mathrm{kg/hr}$ ($1\,\mathrm{m^3/hr}$), a vapor flow of $20\,\mathrm{kg\text{-}N_2/hr}$ temperature of $20\,^{\circ}$C and 0.1–1 bar pressure. Stripping efficiency scales inversely with gas solubility in PC. At the nominal operating conditions Ar and Kr concentrations are effectively reduced by a factor of > 100 from PC. Rn concentration is only reduced by a factor of ~ 3.

Gas stripping is preferred to vacuum stripping because the gas flow improves the interfacial mass transfer between liquid and gas permitting higher efficiency. Reducing the pressure improves the theoretical purity for the same number of theoretical stages, but the equipment does not function as efficiently so it is harder to get to the theoretical purity limit. Steam stripping is an alternative to vacuum stripping that was tested in Borexino [51] and is planned for in SNO+ [55].

For nitrogen stripping to achieve the required Ar and Kr levels in the scintillator the Ar and Kr levels in the N_2 must be very low; the required concentrations are given in table 3. The Borexino collaboration tested nitrogen supplies from different producers and identified a number of sources with sufficiently low Ar and Kr for Borexino [56]. SNO+ is proposing to employ steam stripping instead of N2 stripping. Because Noble gas solubility in water is very low the concentrations of Ar and Kr are easier to achieve.

Both Borexino and KamLAND applied gas stripping after water extraction or distillation of the scintillator [51]. The gas stripping reduced the dissolved water content in the scintillator, which kept water from phase separating into micron sized water drops and scattering the scintillation light. Scintillator leaving the water extraction column is saturated with dissolved water. If the temperature of the scintillator decreases the scintillator is supersaturated with water and fine drops phase separated causing light scattering.

5.5 Adsorption

Radioactive or optical impurities can be removed by adsorption onto high surface materials such as silica gels. The scintillator is passed through the column and polar impurities are sorbed onto the gel. The column has a finite capacity and must be changed periodically after the surface adsorption sites are saturated.

The Borexino collaboration tested adsorption to remove U and Th from a scintillator of PXE (phenylxylylethane) and p-terphenyl using silica gel and alumina adsorbents. Laboratory experiments with neutron activation analysis showed very promising results for removal of U and Th [57]. The low levels of U and Th were not achieved with the large-scale CTF. One cause of the poor performance with the CTF was PPO adsorbed onto the adsorbent and block U and Th adsorption.

KamLAND performed laboratory tests for adsorption of ^{212}Pb from scintillator [54]. They found promising results for adsorption of the Pb, but they also found that the fluor, PPO, was adsorbed as well. KamLAND also reported that silica gels had high K concentrations that could potentially be a problem for the scintillator. SNO+ has plans to use adsorbents with the LABs scintillator for metal ion sorption [58].

5.6 Prepurification of fluor

PPO (2,5, diphenyl oxazole) has been the choice of fluor for Borexino, KamLAND and SNO+. Borexino and KamLAND tested commercially available PPO and found U and Th levels to be acceptable but the K contamination was too high. Potassium was at the ppm level [25], and it must be reduced to $< 10^{-11}$ g-K/g-PPO.

PPO is solid below 70 °C. To simplify processing Borexino collaboration chose to purify a concentrated master solution of 100 g-PPO per 1 L-PC. Three methods of purifying the master solution were to reduce the K level in the master solution: 1) the master solution was filtered through a 0.05 micron Teflon filter; 2) the master solution was extracted with an equal volume of distilled water; 3) the master solution was batch distilled under vacuum at 80 °C; 95% of the PPO was collected in the distillate and 5% was rejected as bottoms containing the K impurity.

The PPO master solution was water extracted for the CTF [15, 25], but was both water extracted and distilled for Borexino [51, 59]. Distillation was semi-batch where master solution is continuously fed to an evaporator operating at ~ 200 °C and 30 mbar. Approximately 99% of the master solution is vaporized and the remaining 1% is allowed to concentrate in the bottom of the reboiler along with the low volatility impurities. Approximately 2–3 L of concentrated PPO ($\sim 99\%$ PPO/1% PC) was discharged and discarded from the evaporator bottoms every hour. The liquid product from the condenser is stripped by nitrogen in a column packed with stainless steel gauze.

PPO master solution distillation was done with no air exposure. Vacuum tightness was necessary to avoid contamination by Ar, Kr and Rn from the air, and to avoid any oxygen exposure. After distillation the PPO master solution was stored in a vacuum tight container under a nitrogen atmosphere. The PPO master solution is metered to mix with freshly distilled PC during the filling of the Borexino detector. Details about the Borexino fluid handling operations are detailed elsewhere [27, 60].

KamLAND purified PPO by water extraction at 75–90 °C, where both PPO and water are liquid [53, 54]. By heating all the equipment and piping above the melting point of PPO and below the boiling point of water KamLAND was able to contact the PPO liquid with liquid water. KamLAND also tested distillation of PPO in lab experiments, but they did not pursue that approach with the KamLAND detector.

6 Achievements

6.1 CTF

The Borexino collaboration built a 4 ton liquid scintillation counting test facility (CTF) to demonstrate the technology to produce high purity liquid scintillator detector; a description of the CTF and the purification system it employed is published elsewhere [25]. The primary achievements of the CTF were: 1) demonstration of purification of multi-ton quantities of liquid scintillator to $< 5 \times 10^{-16}$ g-U/g-scintillator; 2) ^{14}C/^{12}C $\sim 10^{-18}$; 3) demonstration of on-line purification of multi-ton quantities of quantities of scintillator; 4) identification of the need for high-purity gas stripping to remove ^{85}Kr and ^{39}Ar from the scintillator; 5) the need for a veto of muon related events. The CTF was valuable in evaluating alternative scintillator components and purification technologies. It was large enough to prove technologies at a scale relevant to a solar neutrino detector, yet small enough to permit economical testing.
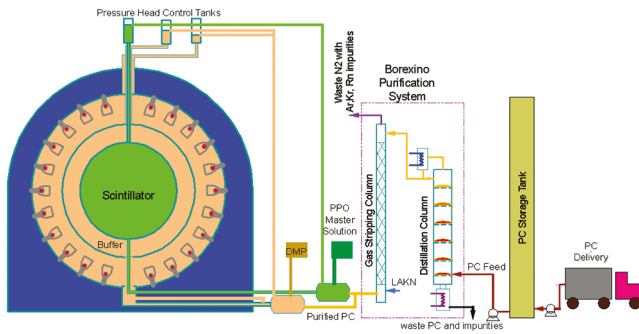
Fig. 4. Schematic of the Borexino solar neutrino detector at Gran Sasso. The entire detector is located 1400 m underground. The detector is at the left. The outer tank is filled with water. Inside the water tank is a stainless steel sphere that supports the phototubes and contains two pseudocumene buffer regions; at the center is a pseudocumene/PPO scintillator mixture. The delivery of pseudocumene and purification of the scintillator and buffer is illustrated to the right. Reprinted from reference [27] with permission from Elsevier.

6.2 Borexino

Borexino began operation as a detector for ^{7}Be solar neutrinos in 2007. The details of preparation of the scintillator and filling of the three zones of the detector are provided in papers by the collaboration [27,49,51]. The process schematic shown in fig. 4 illustrates the integration of the detector, the scintillator fluid handling and the scintillator purification. Detector filling required coordination of delivery of the solvent, on-line purification of the solvent, on-line mixing of the solvent and master fluor solution, controlling the fluid flow rates into the scintillator and buffer volumes to maintain equal hydrostatic pressures, and control of nitrogen pressures for blanketing the scintillator.

The key elements of successful handling the scintillator in Borexino are summarized below.

1) A concentrated master solution of PPO in PC was prepared by filtration, water extraction, distillation and gas stripping.

2) PC was delivered directly from the refinery on Sardinia to the underground laboratory by tanker truck and off loaded into $100 \, \mathrm{m}^3$ storage tanks. Shipping time was kept to < 3 days to minimize cosmogenic formation of ^{7}Be.

3) The PC was purified by distillation and nitrogen stripping. Purified PC was mixed in-line with the concentration PPO master solution to achieve the desired scintillator composition. The mixed scintillator was metered into the three volumes of the detector to maintain neutral buoyancy.

4) Purification and filling were all done with continuous blanketing of high purity nitrogen.

5) There has been multiple repurification of the scintillator by water extraction. Repurification involves filtration, water extraction and nitrogen stripping.

The initial purification of the PC during detector filling produced scintillator of sufficient quality to measure the ^{7}Be and ^{8}B solar neutrino fluxes [61,62]. The repurification by on-line water extraction in 2010–2011 reduced backgrounds to allow the first measurement of pp neutrinos, and to obtain limits on the pep neutrino flux [63–65]. Continued on-line purification has reduced the ^{210}Pb impurity levels that should provide improved limits on the pp and CNO neutrino fluxes.

6.3 KamLAND

KamLAND was originally built for detection of antineutrino from nuclear reactors. It looked for delayed coincidence events with energies $> 2.2 \, \mathrm{MeV}$ so impurities were not critical. It was converted to a solar neutrino detector by initiating scintillator purification in 2007 and a second campaign in 2008–2009. The initial filling of KamLAND used the scintillator components as received. The backgrounds after the initial filling were too high to detect the low energy solar neutrinos. Several purification campaigns of scintillator distillation reduced the backgrounds sufficiently to measure the ^{7}Be solar neutrino flux [17]. The ^{7}Be flux KamLAND measured is consistent with the value reported by Borexino [17,64].

6.4 SNO+

At this time the SNO+ experiment has not run for sufficient time to have data on the low energy neutrinos.

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