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A new water-based liquid scintillator and potential applications

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ABSTRACT

In this paper we describe a new type of scintillating liquid based on water. We describe the concept, preparation, and properties of this liquid, and how it could be used for a very large, but economical detector. The applications of such a detector range from fundamental physics such as nucleon decay and neutrino physics to physics with broader application such as neutron detection. We briefly describe the scientific requirements of these applications, and how they can be satisfied by the new material.

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1. Introduction

Considerable interest has emerged in deep underground large detectors capable of detecting rare events from natural or man-made sources [1–2]. Much of the interest derives from the possibility of detecting neutrino interactions over long distances to fully determine the mixing phenomena that has been uncovered in the last two decades [3]. In particular, the detection of CP violation in the neutrino sector, using an accelerator produced beam, needs a capable detector with efficient mass (fiducial mass multiplied by detection efficiency) in excess of 100 kTon [4–5]. Such a detector, if located deep underground, could have significant potential for detection of nucleon decay and astrophysical sources of neutrinos. The most common type of such a detector is a water Cherenkov counter in the style of Irvine–Michigan–Brookhaven [6] or Super-Kamiokande [7]. In light of the above scientific interest, the Long Baseline Neutrino Experiment (LBNE) has been proposed. Along with a very intense neutrino beam from Fermilab, LBNE could include a 200 kTon water Cherenkov detector located 4850 ft underground in the former Homestake gold mine. In this report, we propose an additional enhancement to such an important detector facility via the dissolution of scintillation liquids in the ultrapure water. The addition of scintillator to the water Cherenkov detector will have two consequences: ionizing particles below the Cherenkov threshold in water (with $\beta < 0.75$) become detectable by scintillation light, and very low energy electromagnetic energy deposits could become detectable if backgrounds can be controlled.

Pure liquid scintillator (LS) has been the choice of detection medium for several neutrino experiments, with the common features of (1) high light yield at 30–50% anthracene, (2) adequate attenuation length ($\sim$15 m after purification), and (3) long stability (3+ years with careful chemical treatment). However, high cost, poor compatibility with materials and extensive liquid handling often raise special concerns, particularly regarding chemical safety in the confined spaces of underground laboratories. Water-based liquid scintillator is of great interest to large scale physics experiments, e.g. nucleon decay, long baseline accelerator neutrinos, geo-neutrino, etc., due to the relative simplicity of liquid handling and cost-efficiency. Although its photon yield is less than that of pure liquid scintillator, the superior attenuation length of water, $\sim$100 m, as achieved in Super-Kamiokande [7] and SNO [8] may well compensate to give an acceptable photoelectron (p.e.) yield.

Commercial scintillation cocktails designed for measuring radioactive α- or β-emitting nuclides in aqueous solution are of similar chemical composition and use similar energy-transfer mechanism as the scintillator described here; however their opaque micro-emulsion and instability in water (often breaking up into two phases in a few weeks) are unsuitable for use as a detection medium for neutrino experiments, which require long attenuation length and extended chemical stability over years. We have studied scintillating liquids with strong potential for mass production using water as a solvent for organic liquid scintillator. The scintillation process mainly takes place in the solute, which is a classic aromatic liquid with a high density of π-electrons for energy transfer. Conventional liquid scintillator is

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a non-polar molecule and therefore must be engineered by the use of a proper surfactant to become miscible with water. The resulting material can be adjusted to have sufficient photon yield, attenuation length, and chemical stability. The photon yield of aqueous scintillator compared with pure liquid scintillator is not expected to be linear with respect to the percentage of scintillation liquid in the water. In the description below, we briefly describe the physics motivation, the material chemistry and preparation methods, preliminary performance measurements, and possible improvements.

2. Physics requirements

The two main physics signatures of relevance for this work are proton decay, specifically the super-symmetry (SUSY) favored decay, $p \to K^+ \bar{\nu}$, and low energy neutron detection and tagging. The most important features of the water-based scintillation medium are acceptable light yield, superior attenuation length, long chemical stability, and good time response (the ability to follow a time sequence of decays or interactions over a large dynamic range from nanoseconds to milliseconds). In addition to these physics goals, the scintillation light should allow lower energy thresholds and additional reconstruction capability for low energy neutrino detection. But here we will mainly consider the proton decay and neutron tagging physics signatures to understand the detector requirements.

2.1. Proton decay

One of the unique predictions of Grand Unified Theories (GUTs) is a finite nucleon lifetime through baryon number violation. A recent review of the subject is in Ref. [9]. The two recent important inputs into GUTs are the apparent unification of the three running coupling constants of the Standard Model [3] and very small masses of neutrinos recently discovered in neutrino oscillations.

Many recent analyses have suggested that GUT theories with a gauge coupling unification scale of $\sim 2 \times 10^{16}$ GeV could be the theory that unites all particles and interactions [9]. A key prediction of these models is proton decay. The two decay modes commonly considered are $p \to \pi^0 e^+$ and $p \to K^+ \bar{\nu}$. The first mode is considered to be the leading mode in many GUTs, in particular non-SUSY GUTs with typical predicted lifetime of $\sim 10^{35 \pm 1}$yrs. It is also the mode with the best limits because water Cherenkov counters have high efficiency to this mode. The current limit is $\tau(p \to \pi^0 e^+) > 8.2 \times 10^{33}$ yrs at 90% C.L. [10] obtained by the Super-Kamiokande collaboration.

On the other hand, the mode $p \to K^+ \bar{\nu}$ is favored by a number of SUSY GUTs with typical predictions in the range of $10^{34 \pm 1}$ yrs. The search for this mode in large water Cherenkov detectors is hindered by the kinematics of this decay. The momentum of the K^+ in this two body decay is 339 MeV/c (kinetic energy of 105 MeV), which is below the Cherenkov threshold for water. Therefore both final state particles are not visible in a water Cherenkov detector. The charged kaon has a range of about 13 cm in water. It decays with a lifetime of 12.4 ns into two main modes: $K^+ \to \mu^+ \nu_\mu$ and $K^+ \to \pi^+ \pi^0$, with branching ratios of approximately 63% and 21%, respectively. Other decay modes have much smaller branching ratios and are more difficult to reconstruct. The two main K^+ decay modes are visible in a water Cherenkov detector. The experimental signature for this decay is a time coincidence of the proton decay and the kaon decay. Nuclear de-excitation of the remaining nucleus can tag the time of the proton decay in a water Cherenkov detector.

The current best limit of $\tau(p \to K^+ \bar{\nu}) > 2.8 \times 10^{33}$yrs at 90% C.L. also comes from Super-Kamiokande [12]. When a proton bound in an oxygen atom decays, it produces a nitrogen ion in an excited state that decays about a third of the time via a detectable photon $\sim$6 MeV. This photon was used to tag the proton decay to obtain the above limit [11]. This method suffers from low efficiency because of the branching fraction of the nitrogen as well as the efficiency for detecting the 6 MeV γ-ray.

In a large water-based scintillation detector, the K^+ would deposit $\sim$90 MeV of visible energy, assuming a Birk's constant of about 0.01 for the saturation of the scintillator, for a factor of 3.75 increase in efficiency with respect to the Cherenkov detector. The large ($>$ 150 MeV) amount of visible energy from the subsequent K^+ decay would allow improved discrimination of shorter-lived kaons and approximately double the efficiency with respect to the Cherenkov detector. The overall increase in efficiency would thus be 7.5 and greatly extend the sensitivity to proton decay. For example, a 100 kTon detector could reach a lifetime limit of 10^{35} yrs in 10 yrs of run-time, essentially covering the entire range from current GUTs.

To achieve the above physics sensitivity, such a 100 kTon water-based liquid scintillator detector would have to satisfy four requirements: (1) the detector has to be housed deep underground, at least 3000 m-water-equivalent, to limit the backgrounds from cosmic ray interactions, in particular muon interactions that produce strange particles, such as neutral K^0 mesons, that can mimic the proton decay signature [13] by entering the detector and charge exchanging into K^+. (2) there must be sufficient detected scintillation light from the K^+. The number of detected photons needs to be well above the dark noise expected from the photo sensors during the time interval from the decay of the proton to the decay of the kaon. We determine this threshold to be approximately $d_t = 50$ detected photons for the K^+ energy deposit of $T_v = 90 MeV$, which includes correction for saturation. From this requirement we can compute the scintillation light yield and the attenuation length needed for the scintillator. (3) the light from the event must be able to travel to the photo-sensors without significant absorption. The attenuation length of light includes both the effects of absorption and of scattering. Here for a conservative estimate we will assume that absorption dominates. Since the detector dimensions will be approximately 50 m, the attenuation must be $\lambda > 20$ m in the appropriate wavelength of $\sim 400 \pm 50$ nm. (4) Lastly, the pulse shape (or time signature) of the K^+ track must be cleanly separated from the pulse shape from the K^+ decay products. The last requirement implies that the pulse broadening due to intrinsic scintillation processes as well as scattering and other detector related effects must be less than the 12.4 ns of K^+ lifetime.

The most likely photo-sensor for this application will be a photo-multiplier tube, which has a typical photocathode efficiency of 20% in the 400 nm wavelength range. We will assume that 25% of the surface area of the detector is covered by photocathode, so that $\varepsilon_p \sim 5\%$ of the light that reaches the wall of the detector is converted into p.e. signals. If L is the average length for the flight path of photons to the photomultiplier tubes then we can compute an approximate relationship between the needed light yield (I_G) per unit ionization energy deposited (in MeV) versus the attenuation length of the light in the appropriate wavelength band (typically $\sim$400 nm)

$$I_G = \frac{d_t}{T_v \varepsilon_p} \times e^{L/\lambda}$$

We set $L = 25$ m, the resulting relationship between the required light yield and the attenuation length is plotted in Fig. 1.

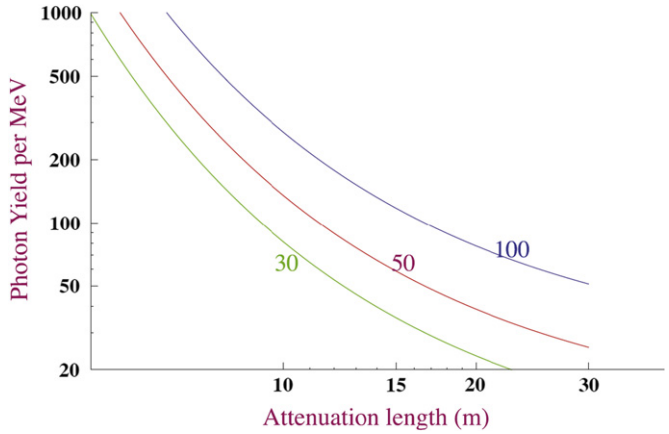

Fig. 1. The required light yield per MeV as a function of attenuation length for 3 different values of detected numbers of photo-electrons (30, 50, or 100 p.e. as indicated) for the kaon track in the $p \rightarrow k^+ \bar{\nu}$.

2.2. Neutron tagging

A neutron with initial energy of a few MeV or less, rapidly loses energy in hydrogenous materials such as water or liquid scintillator. Once thermalized, it can be captured on hydrogen with an emission of a 2.2 MeV photon. The neutrons can also be captured on isotopes loaded into the scintillator that have large cross-sections for neutron capture and a larger release of energy. For example, two stable isotopes of gadolinium (Gd^{155} and Gd^{157}) with large abundances (about 15% each) are known to have very large cross-sections with 8 MeV of average photon energy release. The γ-ray energy is observed mainly through Compton scattering electrons, many of which can be below the Cherenkov threshold.

The detection of neutrons is important for one of the key low energy physics signatures. The detection of anti-electron neutrinos $\bar{\nu}_e$ can be made through the inverse beta decay process ($\bar{\nu}_e + p \rightarrow n + e^+$). This was the process that allowed direct detection of neutrinos [14]. This process has allowed detection of neutrinos from reactors and potential supernova explorations. For the future, a very large detector placed at a great distance from a reactor complex could refine the solar neutrino oscillation parameters. The same detector could be used for detection of diffuse neutrino flux from distant past supernovae [15].

Sensitivity to low energy neutrons in a large water-based liquid scintillator detector can be achieved by two means: capture on free protons or capture on Gd. The neutron can be captured on free protons with an average lifetime of $\sim$200 μsec in the liquid with a release of 2.2 MeV of γ-ray energy. Such detection will require good purity to suppress accidental backgrounds from radioactive impurities (uranium, thorium, and radon) in the detector and shielding from external sources. The detection efficiency can be enhanced if it is in coincidence with the primary event such as inverse beta decay even though the detection energy is small. An advantage of loading with Gd is that the time for neutron capture can be lowered to 30 μsec and also the energy release is much higher at $\sim$8 MeV allowing for easier detection. With a water-based scintillator most of the energy released by capture on Gd can become visible in contrast to a water Cherenkov detector in which only $\sim$1/2 of the energy is visible due to the Cherenkov threshold [16].

2.3. Other applications

Precision long-baseline neutrino oscillation measurements will rely on accurate knowledge of neutrino–nucleus interactions. To obtain this information, the detector near the neutrino beam source should have the same elemental composition as the far detector. A near detector containing water-based LS would be fully active and probe the same neutrino–oxygen interactions observed in a far, large water Cherenkov detector.

Lastly, inexpensive large neutron detectors could be useful for security monitoring of containers to detect transport of radioactive materials. They could also be useful for inexpensive active veto shields against cosmic ray muons.

3. LAB-based scintillating aqueous solution

3.1. Chemistry of water-based material

Organic liquid scintillator dissolved at an appropriate concentration in a water medium is essential to the success of the water-based liquid scintillator. Organic solvents are immiscible in water mainly due to the differences in polarities. A surfactant that contains lypophilic and hydrophilic groups is required to emulsify the organic liquid scintillator into the water solvent. Engineering of a complexing medium to stabilize the lypophilic and hydrophilic molecules in a water medium with appropriate optical transmission and long-term stability is our priority of this study. A series of conventional organic liquid scintillators, such as PC, PXE, PCH, and LAB, commercially capable in large quantity, have been systematically studied at Brookhaven National Laboratory (BNL). Their loading capabilities, optical properties, purification methods, proton densities, commercial availabilities, etc., are well understood [17]. A suitable ampliphilic surfactant that can reduce the liquid–liquid interfacial tension between organic solvent and water, and thus blend the liquid scintillator into the bulk water molecules is essential to stabilize the water–liquid scintillator solution. The degree of tension reduction is a function of surfactant concentration, which could also affect the optical and stability properties of the medium.

3.2. Preparation and performance

Linear Alkyl Benzene (LAB) is currently used in several neutrino experiments (Daya Bay [18], SNO+ [19], Reno [20], and LENS [21]). LAB has a long alkyl chain, 10–13 carbon atoms, attached to the benzene ring (where the unsaturated π-electrons are essential for emission of ultra-violet light due to incident ionizing radiation). LAB has several advantages as LS: (1) good optical transparency ($>$20 m at 430 nm) [22], (2) relatively high photon yield (30% of anthracene), and (3) high flash point (140 °C, compared to 38 °C for the commonly used pseudocumene as in Borexino [23] and KamLAND [24]).

The worldwide industrial uses of LAB (CEPSA corporation records the use of millions of tons per year) are for lubricants and organic solvents in application of manufacturing detergents, in which a sulfonic-acid (HO-S-O-OH) derivative of alkyl benzene (LAS) is produced. Briefly, the chemical basis of LAS is that the alkyl chain and benzene group act as a solvent to extract oils and grease (lypophilic), while the attached sulfonate group is soluble in water (hydrophilic). The unique lypophilic and hydrophilic properties, and associated benzene π-electron ring, acting as surfactant and liquid scintillator simultaneously, of LAS, make it a promising candidate for a water-based liquid scintillator (or organo-scintillator loaded water detector, see Fig. 2). The production quantity and cost of the LAS are very attractive for large-scale neutrino detectors.

Water based scintillator (WbLS) R&D based on LAS was initiated by the BNL neutrino group, and so far a range (as high as 40% LS) of organic liquid scintillators have been successfully

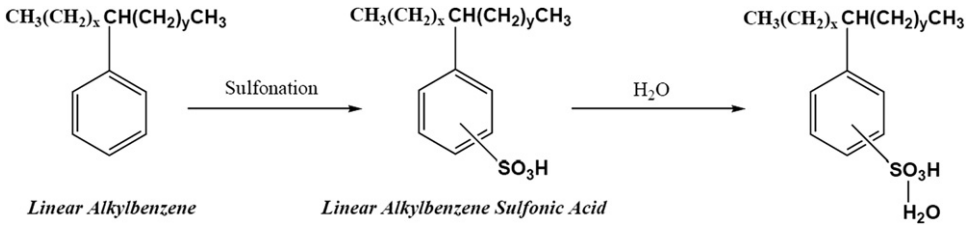

Fig. 2. Schematic of the chemical attachment of a LAB molecule to water using a surfactant.

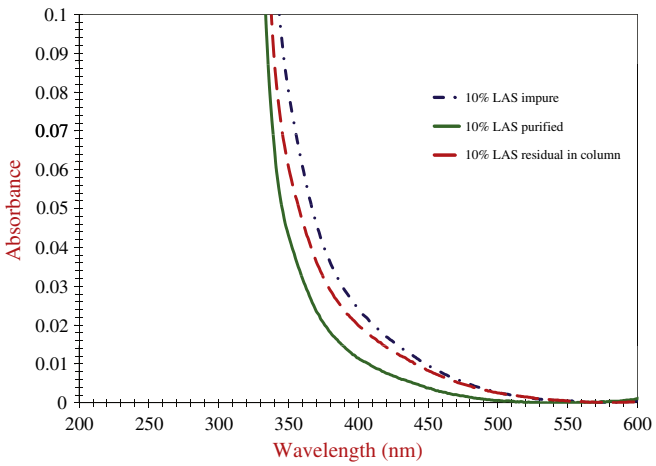

Fig. 3. Relative absorption through a 10 cm cell for water-based liquid scintillator with 10% loading of LAS before and after purification.

loaded in water using a newly developed synthesis recipe. These WbLS samples have been stable for more than 1.5yrs since preparation.

Selected UV transmissions of 10% organic scintillator in water before and after column purification are shown in Fig. 3. The 10% LAS residual-in-column represented the colored impurities removed via the purification steps. The optical transmission of the initial LAS material is not favorable (dark brown) for use as a particle detector, but can be improved greatly using dry-column purification. An optically improved LAS feedstock is under development, in collaboration with the vendor, and will be available in a few months. Fig. 4 shows some of these samples excited by an UV lamp at 250 nm (in the peak of LAS absorption). It should be noted that loading wavelength shifter (such as Carbostyril-124) [25] into water is different from the water-based liquid scintillator. The former only improves the Cherenkov counting efficiency, but has no capability of capturing incident UV photons induced by ionization radiation. As presented in Fig. 4, pure water loading with Carbostyril-124 did not emit blue light while WbLS samples with either PPO or Carbostyril-124 did.

To corroborate the preliminary UV-light test, a further study of light-yields relative to water was done with Cs^{137} source. The γ-Compton scattering (Fig. 5) data show that the photon production of 10% loaded WbLS is well above the background or water loaded with Carbostyril-124 alone even though the incident radiation rate is attenuated due to optical and chemical quenching. A detailed light-propagation study using a larger detector and high-intensity proton beams at different energies is essential to understand the attenuation factor. Yet the scintillation photons measured from the WbLS, compared to that of the Carbostyril-124-loaded pure water with and without $Cs^{137}\gamma$-rays, clearly indicate that the light obtained from the WbLS sample is from ionization induced by the incident γ-radiation.

As described in the physics requirements, the pulse duration from the scintillation media is another key factor for measuring fast decays. The timing properties of the emitted pulse were measured using time-resolved fluorescence spectroscopy. The water based scintillator sample was excited by a short pulse (1 ns FWHM) at 250 nm. The emission pulse measured at 329 nm was found to have two components with decay times of 3.8 ns and 10 ns. The spectrum of emitted light was also measured by steady-state fluorescence spectroscopy and is shown in Fig. 6 in comparison with pure LAB-PPO scintillator. This demonstrated the light-propagation of the WbLS is similar to that of pure LAB. However the contents and types of fluor and shifter in the WbLS needs to be optimized, as shown in the Fig. 6 of incomplete transition between LAS emission (280 nm) and PPO absorption (300 nm).

4. Deployment of water-based scintillator in a very large detector

This work has demonstrated the principle that a mass-producible, cost-effective water-based liquid scintillator can be achieved using the derivatives of linear-alkyl-benzene-based organic scintillator (linear alkyl benzene sulfonate, LAS). For the successful application of WbLS to large-scale physics experiments, several key characteristics, such as (1) adequate photon production, (2) long attenuation length, (3) fast decay time, and (4) chemical stability, require systematic study with the final synthesized recipe. Preliminary data show that 10% WbLS emits scintillation light through the ionization process, similar to but with less yield than LAB ($\sim$120 p.e./MeV at 15% photo-coverage). Nevertheless quenching phenomenon of WbLS is not yet understood. The optical property of the prepared WbLS material does not yet meet the requirements for a large detector. Conventional liquid scintillators are often slightly colored, and thus not suitable for large-scale neutrino experiment without purification. With dry-column exchange and short path vacuum distillation, the optical transmission was significantly improved. On-line purification schemes for the organic-water mixing system need to be developed. Lastly, pure LAB liquid scintillator is known to decay in the range of few to tens of nanoseconds and the WbLS decays with a similar lifetime, which makes the WbLS suitable for fast particle observation, like K^+ decay.

Additional studies with cosmic ray muons and high intensity proton beams (0.1–1 GeV) are planned to examine the light propagation phenomenon within the liquids. These will need a detector with several kilograms of material. In addition, other WbLS characteristics, such as organo-complexing species in water, light scattering and quenching, large-scale purification, etc., require further study. Stability of WbLS under various environments needs to be studied even though the current WbLS samples have been stable for $>$ 1.5yrs since preparation.

Loading organometallic ions into the WbLS to enhance the particle-capture signal is under development. Preliminarily loading

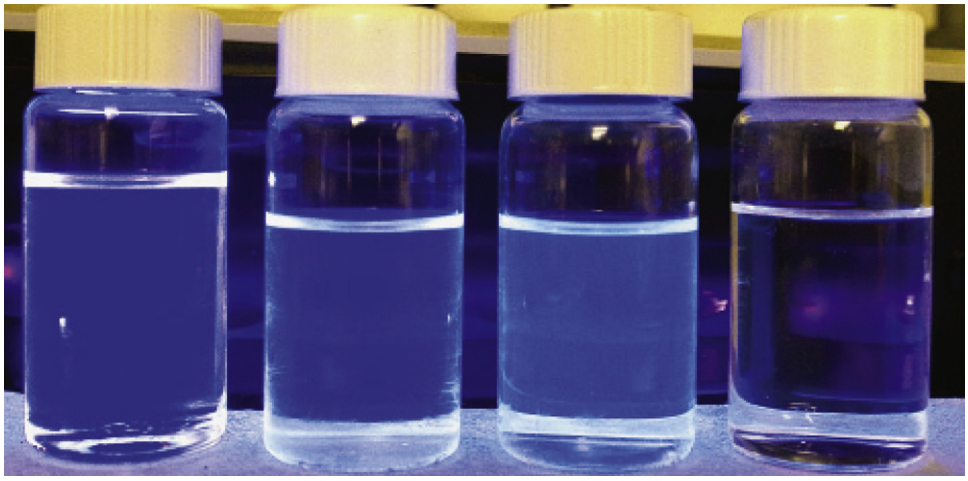

Fig. 4. Samples of water-based liquid scintillator samples illuminated by ultraviolet light. From left to right, samples are pure LAB based liquid scintillator, 10%-LAS loading in water with 3 g/L PPO, 10%-LAS loading in water with 3 g/L Carbostyril-124, and pure water with 3 g/L Carbostyril-124. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

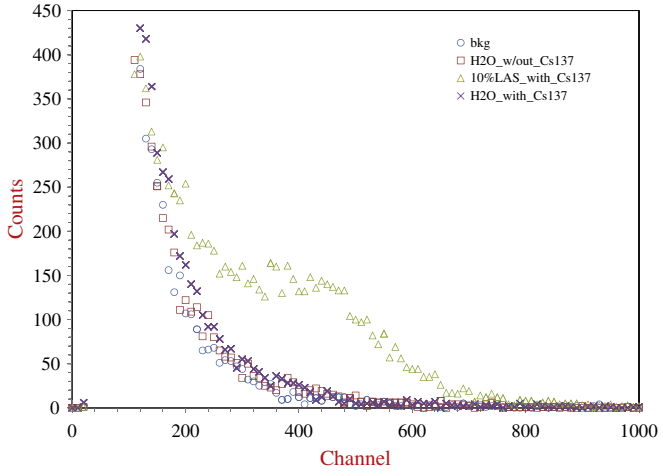

Fig. 5. Scintillation response from WbLS. The liquids were excited by external Cs^{137}.

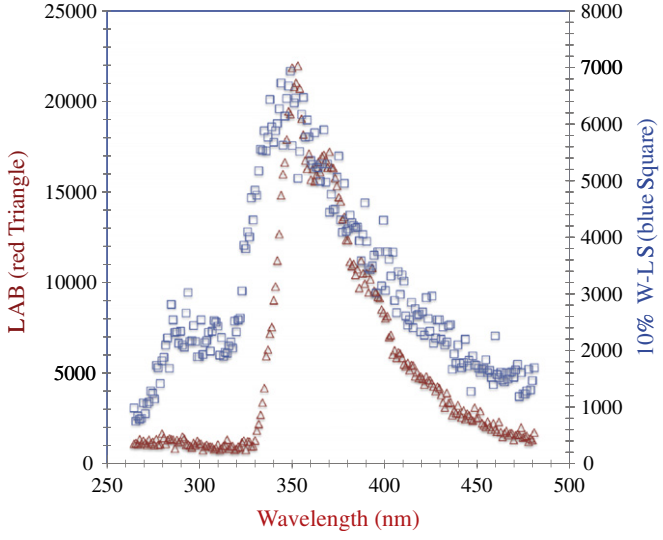

Fig. 6. Fluorescence response from scintillator liquids. The liquids were excited by 250 nm laser with a fixed intensity and observed to produce the plotted radiation. The red curve is for LAB with PPO and the blue is the WbLS with PPO (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of gadolinium into the WbLS has been tested and the results are very promising. Current samples of ~0.5% Gd in WbLS are available for in-lab tests; however loading parameters for large-scale production will require modification depending on the experimental needs. Eventually a larger WbLS (with and without metallic ions) demonstrator, at the scale of few tons, will be built for prototype-studies of anti-neutrinos from a nuclear reactor.

Acknowledgments

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