



Development of a Gd loaded liquid scintillator for electron anti-neutrino spectroscopy

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Abstract

We report on the development and deployment of 11.3 tons of 0.1% Gd-loaded liquid scintillator used in the Palo Verde reactor neutrino oscillation experiment. We discuss the chemical composition, properties, and stability of the scintillator elaborating on the details of the scintillator preparation crucial for obtaining a good scintillator quality and stability. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

The Palo Verde (Arizona) reactor neutrino experiment is a long baseline disappearance neutrino oscillation search [1]. Its goal is to test the $\nu_\mu \leftrightarrow \nu_e$ solution of the atmospheric neutrino anomaly [2] utilizing the inverse beta decay $\bar{\nu}_e + p \rightarrow e^+ + n$ as detection reaction. The Palo Verde detector is a finely segmented device minimizing correlated backgrounds by exploiting fast triple coincidences between detector elements. The positron energy deposit in coincidence with the annihilation quanta firing at least two neighboring elements identify an

e^+ . A delayed neutron capture signal completes the anti-neutrino signature.

The 11.3 tons of hydrocarbon-based liquid scintillator (H:C \approx 2) serve simultaneously as target and detector. The measured kinetic energy of the e^+ , being stopped in the liquid scintillator, allows the reconstruction of the $\bar{\nu}_e$ energy. As the e^+ energy is rather low (1–5 MeV), a high light yield scintillator is vital for obtaining good energy resolution.

The correlated positron–neutron signature of the detection reaction is a powerful tool for the reduction of random backgrounds. The Gd-loaded scintillator was chosen for the Palo Verde project because it offers two important advantages over pure hydrocarbon-based scintillator:

- A large thermal neutron capture cross section of the isotopes $^{155,157}\text{Gd}$ (61 400 and 255 000

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barns) shorten the neutron capture time. For 0.1% Gd loading (by weight) the neutron capture time is $\tau \approx 30 \mu\text{s}$, compared to $\tau \approx 180 \mu\text{s}$ for capture on the proton in unloaded scintillators.

- A release of a high energy (8 MeV) gamma cascade after thermal neutron capture on Gd results in a neutron capture signal well above the radioactivity backgrounds.

The Palo Verde detector consists of 66 acrylic tanks (900 cm \times 12.7 cm \times 25.4 cm) containing the liquid scintillator. Acrylic was chosen to obtain efficient light collection into the 5" photomultiplier attached to either side of the tank by exploiting total reflection. This choice requires long term compatibility of the cells with the liquid scintillator and excludes the use of pure solvents like xylene (dimethylbenzene) or pseudocumene (1,2,4-trimethylbenzene, PC). Goal of the project was to formulate a liquid scintillator containing not more than 40% PC diluted with mineral oil and dissolve the Gd in this mixture in a way compatible with the acrylic. Early developments of this cocktail were done in collaboration with NE Technology Ltd. The relatively large tank length places strict requirements on the transparency of the scintillator. We will discuss the scintillator formulation chosen to satisfy these requirements. We will present our measurements of the scintillator properties and finally, comment on the stability of the liquid.

The technical details given in this paper might be of interest for other projects also; for example large (100 tons) liquid scintillation solar neutrino detectors, loaded with Gd or Yb, are being discussed [3].

2. Scintillator formulation

To assure compatibility of the scintillator with the acrylic tanks a mixture of pseudocumene and mineral oil was chosen. As Gd dissolves in neither of the above liquids it was necessary to prepare a soluble and acrylic-compatible Gd compound. The compound prepared was gadolinium 2-ethylhexanoate [$\text{Gd}(\text{CH}_3(\text{CH}_2)_3\text{CH}(\text{C}_2\text{H}_5)\text{CO}_2)_3 \cdot x\text{H}_2\text{O}$]. It can be synthesized from gadolinium chloride, oxide, or nitrate. Early preparations in the project

were made from the nitrate, since a sufficiently radio-pure source was available. However, solutions from the nitrate were found to be unstable (solid–liquid phase separation) when handled, especially when exposed to air. The oxide, Gd_2O_3 , was converted to the 2-ethylhexanoate in an aqueous reaction, collected by filtration, and dried. The resultant compound was purity tested and then dissolved in a scintillation solvent, along with a primary fluorescent additive, a spectrum shifter, an antioxidant, and small amounts of two additional solvents to help keep the gadolinium compound in solution. The emission peaks of the primary fluor and spectrum shifter were 365 and 425 nm, respectively.

Pseudocumene was the solvent of choice, for its high scintillation efficiency, lower solvency toward acrylic, and higher flash point than xylene. To increase the hydrogen content, and further decrease the solvent attack on the acrylic tanks, a high purity mineral oil was added as a final component of the finished scintillator solution. The solution is optimized at 60% oil by volume as this is the minimum necessary to dilute the pseudocumene enough that it does not harm the acrylic tank. A higher concentration of oil than this decreases the scintillation light yield, since this is a function of pseudocumene content (an aromatic liquid which inherently scintillates, whereas the oil is a saturated hydrocarbon and does not). Also, the higher the oil content, the lower the possible Gd content, as this also is a function of the pseudocumene solvent content.

We found that the fully blended scintillator, contained in 200 l steel drums did not remain stable (a single-phase liquid) during the transport over ground via truck from Ohio to Arizona. To solve this problem, pseudocumene-based “concentrate” was blended in Ohio and additional pseudocumene and the mineral oil was blended on site in Arizona. This procedure is described below.

3. Scintillator blending

The Gd “concentrate” (BC-521C) as prepared by Bicron at their Ohio factory was shipped to Palo Verde in 200 l stainless-steel drums. There

it was blended with the mineral oil (Witco Scintillator Fluid), and 1,2,4-trimethylbenzene (Koch Chemicals obtained through Bicon Inc.). The proportions used were 1:1:3 by volume for the Gd concentrate, PC and mineral oil to arrive at a Gd loading of 0.1% by mass for the blended scintillator. Special care was taken to select hardware being compatible with the used solvents. As the concentrate and mineral oil do not mix well (rapid addition of the mineral oil may lead to irreversible precipitation of the Gd compound) care was taken to mix those components in a controlled way.

The blending was done in two 200 l stainless-steel drums equipped with air driven stirrers having stainless-steel shafts and impellers. With each drum processing about 170 kg of scintillator; the load of one acrylic cell. The liquids were moved by air driven diaphragm pumps (Nylon body and Teflon diaphragm). Separate pumps were used for the mineral oil, the two other components and the blended scintillator. All piping to come in contact with pure Gd concentrate or PC was made from Teflon while PVDF valves were used. For the mineral oil polypropylene tubing was used. The blending drums were placed on electronic scales for a precise determination of the scintillator weight.

For the blending each component was first pumped into an intermediate stainless-steel vessel placed on an electronic scale to determine its weight and then drained by gravity into the mixing drum. First, the concentrate and PC were mixed. Then the mineral oil was added at a rate not exceeding ~ 3 l/min. During the blending process the liquid was regularly inspected for precipitation of the Gd compound. The stirrers were operated at a low speed to avoid excessive agitation of the liquid. The final mixture was stirred for at least 1 h. The liquid was then pumped through a 200 nm Gelman “HiFLO Sol-Vent DCF” filter directly into the acrylic tank. The filter was changed after every batch of scintillator. A 2 l sample was retained for every batch blended.

The liquid scintillator could not be bubbled with nitrogen for removal of dissolved oxygen as this bears the risk to destabilize the mixture leading to precipitation of the Gd compound. The acrylic tanks were flushed with Ar before the filling in order to remove any residual vapors left over from

the bonding and to displace the air. In addition, all tanks were pressure tested prior to filling to detect possible leaks.

A total of 11 343 kg of liquid scintillator was prepared in this way.

4. Scintillator quality

The following scintillator properties were routinely tested for at least one sample of blended scintillator for every batch of Gd concentrate:

- Light attenuation length at 440 nm
- Light yield
- Gd loading

Achieving an acceptable light attenuation length was the most challenging task during the scintillator development. Before working with production size samples, discussed below, we went through four generations of prototypes. In this prototype-phase emphasis was given to the optimization of the solvent balance and quality of the raw ingredients. While the light yield and Gd loading were satisfactory from the beginning, early samples did exhibit problems in their transparency.

In the following we will discuss our data on the scintillator properties.

4.1. Attenuation length

The light attenuation length was measured using a vertically oriented 1.5 m long, 3 cm diameter stainless-steel tube filled with liquid. A blue LED equipped with a 440 nm wavelength filter and focussed to parallel by means of a 7-cm-focal-length lens, placed one focal length away from a pinhole aperture, illuminates a phototube at the other end of the tube. The LED was operated in pulsed mode to minimize background through scintillation light. The average pulse height, seen by the phototube, was then measured for varying liquid height and fitted to an exponential to obtain the attenuation length (scattering plus absorption).

Fig. 1 shows the results obtained through those measurements. The first entry labeled CF denotes a measurement done with a batch of fluid composed of 36% PC and 64% mineral oil mixed in the

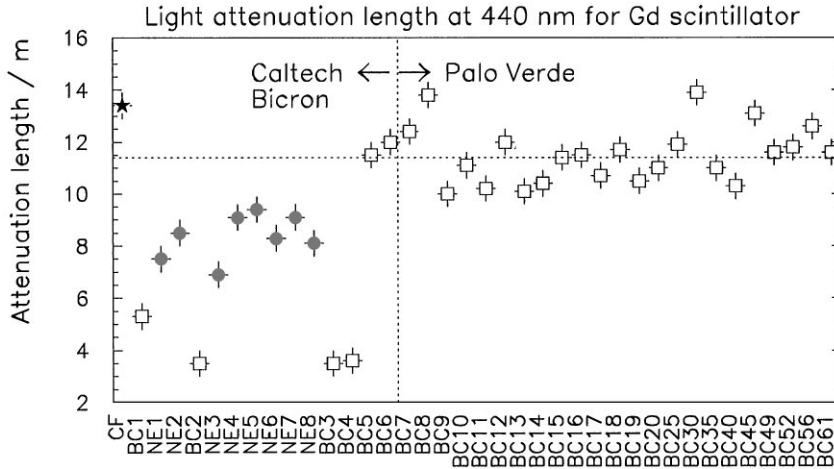


Fig. 1. Light attenuation length at 440 nm as measured for cleaning fluid (CF, star), NE scintillator (NE, circle) and Bicron scintillator (BC, square). The scintillator batches are identified by successive numbering, as given in the figure. All batches to the right of the vertical line were blended in Palo Verde, while the rest were mixed at Caltech or Bicron.

hardware described before. The good attenuation length obtained demonstrated that all used vessels and plumbing had the necessary purity before starting to work with the scintillator. This liquid was also used to test one of the acrylic tanks for surface impurities.

The batches labeled NE in Fig. 1 were early prototypes made by NE Technology Ltd. The first batches blended by Bicron (labeled BC1–4 in Fig. 1) before transportation showed a rather high light absorption. The fact that samples BC5 and BC6, blended at Caltech from Gd concentrate like the NE samples NE1–8, were of much better transparency shows that the blended scintillator is too fragile for a long transport. After switching to the on-site blending attenuation lengths consistently larger than 10 m were achieved for the bulk of the scintillator. For the first three drums of Gd concentrate, the light attenuation length of every scintillator batch was tested (BC7–20), for the later samples only one per concentrate batch. The average attenuation length for samples blended from BC5 on was 11.4 m.

4.2. Light yield

The light yield of the scintillator was measured using a ^{207}Bi conversion electron source, irradiat-

ing a 2" diameter Petri dish containing 14 ml of the sampled scintillator. The resulting liquid height of 7 mm was chosen to stop the ~ 1 MeV conversion electrons (range ~ 5 mm) within the liquid while optimizing the solid angle towards the photomultiplier. The sample was placed on a 5" photomultiplier. To monitor PMT stability a 1" diameter NaI detector, separated by a thin Pb shield from the source, registering its γ -radiation was placed next to the sample onto the same PMT and both spectra were collected in parallel. The position of the ^{207}Bi conversion electron peak in the measured spectrum is a measure of the average pulse height and hence the light yield. A mixture of PC with 4 g/l PPO and 100 mg/l bisMSB, deoxygenated by bubbling with Ar before the measurement, served as a light yield standard. We assume a light yield of 80% anthracene for this mixture. Fig. 2 shows the measured light yield of scintillator samples. The light yield was verified for at least on scintillator batch per concentrate lot. The light yield was consistently 56% anthracene for the 22 tested samples.

4.3. Gd loading

The Gd concentration of the liquid was measured using X-ray fluorescence after thermal neutron capture on Gd, as described in Ref. [4]. The

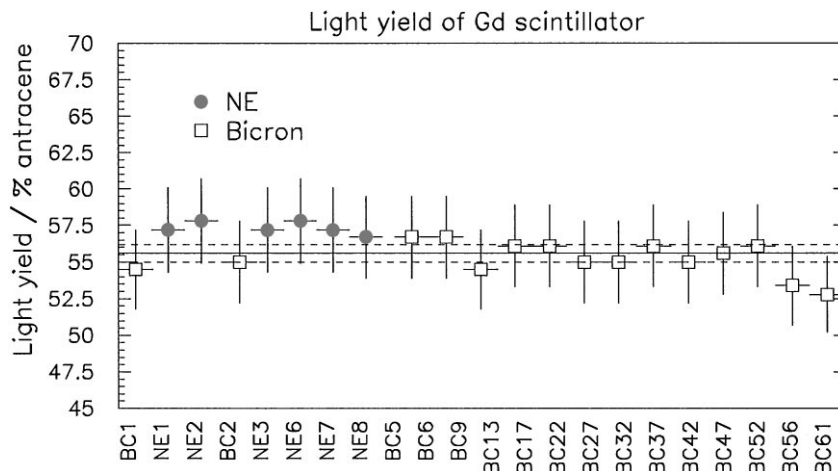


Fig. 2. Light yield of different batches of Gd scintillator.

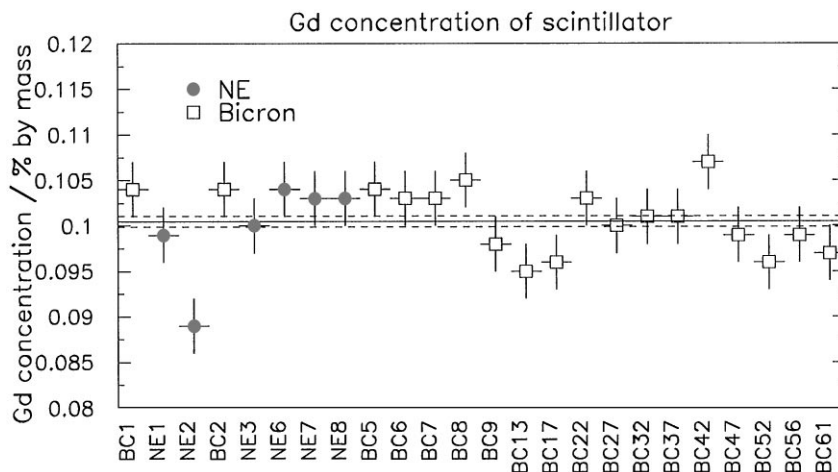


Fig. 3. % Gd loading (by mass) of scintillator batches.

measurements were done in comparison to standard solutions of $\text{Gd}(\text{NO}_3)_3$ dissolved in alcohol. The absolute Gd concentration of one scintillator batch was verified by mass spectroscopy in a commercial laboratory. In addition to this cross check a few early samples were also tested by neutron activation of ^{158}Gd , using a ^{252}Cf spontaneous fission source placed in a water moderator. The 363 keV γ -radiation following the β -decay of ^{159}Gd into ^{159}Tb ($T_{1/2} = 18.6$ h) was detected with a Ge detector.

As for the light yield at least one scintillator batch per concentrate lot was tested for its Gd loading to make sure that the detector is of reasonable uniformity. As can be seen in Fig. 3 the Gd loading was consistently 0.1%. The small scattering shows that the production process of the concentrate is well controlled. The low value of NE2 was confirmed by the observation of Gd precipitation in the transport drum. The solvent balance of all following batches was readjusted after this observation. For prototype samples kept at Caltech

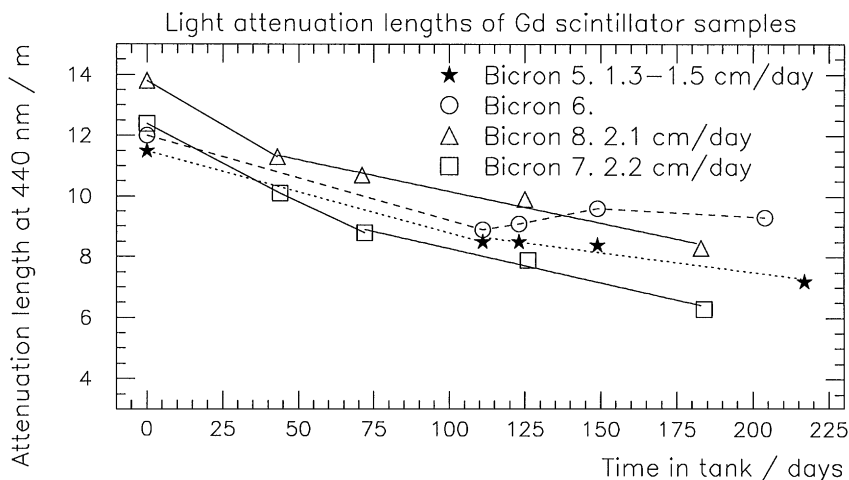


Fig. 4. Time development of the light attenuation length for Gd scintillator batches. Samples have been taken from the acrylic tank.

for several years no instability of the Gd loading was observed. The described loading scheme is quite stable if the liquid is not disturbed or agitated.

5. Scintillator stability

As neither light yield nor Gd loading showed any observable degradation over time we shall concentrate in the following on the time development of the scintillator transparency which is subject to aging. To test the stability of the transparency, samples from four scintillator batches have been taken from the acrylic tanks over time to study the transparency loss of the scintillator in its experimental environment. Fig. 4 gives the results of these repeated measurements. In all cases an initial decline stabilized, resulting in a loss rate of about 2 cm/day. This rate of aging is acceptable for our experiment. The early samples BC1-4 (see Fig. 1) showed a much more rapid loss of transparency. It is interesting to note that the aging process starts only after the blending. Test batches blended from concentrate retains exhibit the same attenuation length as the original liquid sample. The aging process is sped up by subjecting the liquid to elevated temperatures. In situ

measurements made for all scintillator batches by scanning the detector elements with a γ source confirm that the light attenuation has acceptable stability in time.

6. Conclusion

The formulation of a pseudocumene-based 0.1% Gd-loaded liquid scintillator having good long-term stability and compatibility with acrylic has been reported. The measurements done during development and deployment of the scintillator have been discussed. The details of the scintillator preparation, which in our view were important for obtaining scintillator of high transparency and stability, were reported.

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