

A method to calibrate a neutrino detector using the positron emitter ^{68}Ge

Andreas Piepke*, Brian Cook

Department of Physics 161-33, Caltech, Pasadena, CA 91125, USA

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Abstract

A ^{68}Ge source can be used to calibrate the positron detection efficiency of a liquid scintillation neutrino detector. A method to dissolve radioactive ^{68}Ge in pseudocumene-based liquid scintillator is described. The homogeneity of the source distribution has been studied and results for two Ge compounds are discussed. Light yield and light attenuation length of the liquid scintillator are not affected by the Ge doping.

1. Introduction

Liquid scintillation detectors have found widespread use in basic research and applied physics as they are cost effective and can be constructed in any desired shape and size. In particle physics, their high proton content makes them a good choice for the detection of electron anti-neutrinos through the inverse beta decay of the proton: $\bar{\nu}_e + p \rightarrow e^+ + n$.

The present study was motivated by the Palo Verde reactor neutrino experiment [1]. Here, a 12 ton segmented Gd-loaded liquid scintillation detector is being used to search for neutrino oscillations near a nuclear reactor. The detector is comprised of 66 acrylic cells, each 9 m long, arranged in a 6×11 array. The liquid scintillator, now designated as BC 521B, was developed in collaboration with NE Technology Ltd. The neutrino signal is discriminated from energetic background neutrons by making use of the detector segmentation: the positron kinetic energy, deposited in one cell, is detected in prompt coincidence with the annihilation radiation, which penetrates to surrounding cells. The reaction neutron is detected through capture on Gd in a delayed coincidence. A precise calibration of the positron efficiency is crucial as the goal of the experiment is an absolute determination of the neutrino-induced event rate. A measured rate significantly below that expected would signal the presence of neutrino oscillations and hence new physics.

A calibrated β^+ -activity, dissolved in the liquid scintillator of a dedicated detector element, is needed to allow an in situ measurement of the positron detection probability. A comparison of the calculated and measured shape of the positron spectrum tests how well we understand the energy

calibration and corrections for the absorption of annihilation radiation in the “positron-like” cell.

As positron source we have selected ^{68}Ge as it has a reasonably long half life, a high endpoint energy, to resemble the reactor induced positron spectrum (0–6 MeV) as close as possible, and shows only little gamma emission so as not to interfere with the annihilation radiation. The differences in the energy distribution of reactor induced and calibration positrons will result in different admixtures of annihilation in flight. The resulting systematic difference of about 2% will be corrected in the data analysis. Fig. 1 shows a simplified level scheme of the ^{68}Ge decay [2].

In the next sections we describe how to dissolve ^{68}Ge in pseudocumene (trimethylbenzene, $\text{C}_6\text{H}_3(\text{CH}_3)_3$) and mineral-oil-based liquid scintillator and how to perform a precise calibration of its activity.

2. Chemical preparation of the source

Carrier free ^{68}Ge is commercially available only in the form of germanic acid (H_2GeO_3) in dilute HCl (typically 0.5 molar or less). In this form it cannot be dissolved in an organic solvent like pseudocumene. It is known that ^{68}Ge

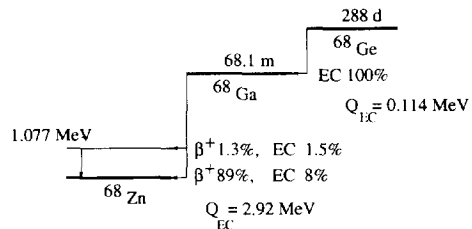


Fig. 1. Simplified level scheme of the ^{68}Ge decay.

* Corresponding author. E-mail andreas@citnp1.caltech.edu.

can be recovered as $^{68}\text{GeCl}_4$ by distillation from a 6M solution [3,4]. In this form it is soluble in pseudocumene, which makes up 35% of our liquid scintillator. A local radiochemical company made such a source for us by dissolving 37.4 kBq of ^{68}Ge in the form of GeCl_4 plus 19.1 mg of stable GeCl_4 carrier in 420 μl toluene, which was added to 183 ml of liquid scintillator.

The inherent problem with this source is that GeCl_4 is highly reactive. It tends to adhere to the surfaces of the container and react with residual water in the scintillator.

We developed a chemically stable source containing Ge in the form of tetra-*n*-butylgermane ($\text{C}_{16}\text{H}_{36}\text{Ge}$) which also dissolves in pseudocumene.

18.5 MBq of ^{68}Ge in the form of carrier-free germanic acid in 0.5 ml of 0.001M HCl was purchased from Los Alamos National Laboratory. The compound was converted to GeCl_4 and the excess HCl removed using the following procedure.

As GeCl_4 is quite volatile and reacts violently with water, all glassware that was to come in contact with it was dried prior to use and then sealed under a dry nitrogen atmosphere. The germanium tetrachloride was handled only after being cooled in an ice bath to prevent evaporation.

2.1. Isotope exchange

100 μl , or 3.7 MBq of ^{68}Ge , of the radioactive germanium solution was added to a dry, cooled (ice bath) 10 ml round-bottom flask, sealed under nitrogen using a gas-tight calibrated syringe¹. The sample was injected through a rubber septum to prevent any contact with air. To this solution was added 200 μl of concentrated hydrochloric acid to bring the solution to about 8M in HCl and convert the germanic acid to germanium tetrachloride. Approximately 0.5 g (270 μl) of stable germanium tetrachloride was added and the mixture stirred for 30 min in an ice bath to extract the radioactive ^{68}Ge into the GeCl_4 carrier. After stirring, the mixture was allowed to stand for several minutes after which 200 μl of GeCl_4 , now doped with ^{68}Ge , was extracted by syringe from the bottom of the flask with the intention of leaving most of the lighter HCl behind.

2.2. Synthesis of the tetra-*n*-butylgermane

200 μl (1.75 mmol) of GeCl_4 doped with ^{68}Ge was transferred into a dried 100 ml flask, sealed under nitrogen and cooled in an ice bath². Through a rubber septum, 5.0 ml of hexane was injected by syringe to act as a reaction solvent. While stirring at 0°C, 7.3 ml (11.7 mmol) of a 1.6M *n*-butyl-lithium in hexane solution was slowly added (the

reaction is exothermic) by syringe. The reaction was stirred for 30 min and then allowed to warm to room temperature and stirred for another hour. The flask was again cooled in an ice bath, and 10 ml of distilled water was added and the mixture stirred vigorously for 10 min. The solution was transferred to a 100 ml separatory funnel and allowed to stand for several minutes while the immiscible liquids separated. The lower layer, consisting of water containing lithium chloride, was drained from the funnel leaving behind the hexane containing the tetra-*n*-butylgermane. This washing was repeated with another 10 ml of distilled water and then once more, using a saturated sodium chloride solution (brine). Finally, the washed hexane solution was poured into a flask containing approximately 2 g of anhydrous magnesium sulfate (MgSO_4) and was stirred vigorously for about 20 min in order to absorb the remaining water. The solution was poured through a fine fritted filter to remove the magnesium sulfate, and the hexane solution was collected in a flask. After collection, the filter still contained a significant amount of ^{68}Ge , so 10 ml of hexane was added to the filter for a second collection. The hexane solution was finally dissolved in about 50 ml of liquid scintillator to make the calibration solution. This solution contained a total of about 0.7 MBq of ^{68}Ge , or 20% of the amount started with.

The described procedure resulted in a radioactive solution containing 18 kBq/ml of ^{68}Ge and 112 ppm of stable Ge carrier³. Li was not detectable in the final solution; its concentration has been determined to be below 30 ppb. The Gd content of the liquid scintillator was measured to be 0.1% by weight before and after the addition of the Ge. A second source with 370 Bq/ml of ^{68}Ge and 213 ppm of stable Ge carrier was prepared to allow experiments with varying Ge concentration.

3. Measurement of the source strength

In the neutrino oscillation experiment we plan to measure the event rate to a statistical accuracy of 5%. To reach such a precision the absolute strength of ^{68}Ge must be known and calibrated to 1-2% accuracy. As the chemical efficiency of the procedure described in the previous section is not known, the source must be calibrated experimentally. Therefore, a calibration method, based on an HP Ge detector, was developed. An extended source presents problems because the self-absorption and solid-angle corrections are difficult to estimate with the required precision. However, by exploiting the coincidence of the positron and annihilation radiation, one can make use of counting rate ratios whereby the various efficiencies cancel.

The apparatus used for the calibration consists of a small cylindrical acrylic tank measuring 10 cm in diameter and about 6 cm in length filled with radioactive scintillator. A

¹ The technique was brought to our attention by David Jamriska of Los Alamos National Laboratory who supplied us with a complete description of the procedure.

² Marcus Boehm of Ligand Pharmaceuticals advised us on how to synthesize the tetra-*n*-butylgermane.

³ The Ge carrier, Li and Gd concentration measurements were performed by Elemental Research Inc. of Vancouver (Canada) by ICPMS.

3-inch photo-multiplier tube is coupled to one side of the tank and the apparatus placed up against an HP Ge detector which is enclosed in a lead house to attenuate natural radioactivities from the walls. A large plastic scintillator above the entire apparatus rejects cosmic muons. In the scintillator, the positron loses energy and then annihilates, producing $E = 511$ keV gammas, which are detected in the HP Ge detector.

The source activity A in Bq is then evaluated as follows: let $N_{\gamma 511}$ be the counting rate observed in the annihilation peak in the germanium detector spectrum. Let N_s be the counting rate of the scintillator in some energy interval, and let N_c be the rate at which both the germanium detector and the scintillator fire together in the correct energy intervals. Let the efficiency of the germanium detector for annihilation gammas be given by ϵ_{Ge} . Let b_0 and b_1 be the positron branching ratios to the ground state and $E^* = 1.077$ MeV excited state of ^{68}Zn , respectively (see Fig. 1). Then define ϵ_{s0} and ϵ_{s1} as the positron detection efficiencies in the scintillator of positrons from the ground state branch and the excited state branch, respectively. These efficiencies are not equal because they include threshold (spectral fraction) cuts. The annihilation radiation produced by the two decay branches is only weakly affected by the different admixture of annihilation in flight. We can neglect its influence on the germanium detection efficiency. We can write the following expressions:

$$N_{\gamma 511} = 2A(b_0 + b_1)\epsilon_{\text{Ge}}, \quad (1)$$

$$N_s = Ab_0\epsilon_{s0} + Ab_1\epsilon_{s1}, \quad (2)$$

$$N_c = Ab_0\epsilon_{s0}2\epsilon_{\text{Ge}} + Ab_1\epsilon_{s1}2\epsilon_{\text{Ge}}. \quad (3)$$

Thus

$$\begin{aligned} \frac{N_s N_{\gamma 511}}{N_c} &= \frac{A(b_0\epsilon_{s0} + b_1\epsilon_{s1})2A\epsilon_{\text{Ge}}(b_0 + b_1)}{2A\epsilon_{\text{Ge}}(b_0\epsilon_{s0} + b_1\epsilon_{s1})} \\ &= A(b_0 + b_1). \end{aligned} \quad (4)$$

The activity

$$A = \frac{N_s N_{\gamma 511}}{(b_0 + b_1)N_c}$$

is thus determined independently of the individual detector efficiencies. They cancel because the experimental conditions are the same for the singles and coincidence measurements (apart from a small correction due to annihilation radiation scattering inside the scintillator). Indeed to ensure precisely the same experimental conditions, the singles and coincidence data are taken together, and coincidences are identified off line so that any gain drift or other environmental change affects both data sets identically.

3.1. The electronics readout

The data are recorded event by event on magnetic disk using a 486 PC and CAMAC electronics. The main event

trigger comes from an OR of the germanium detector and the photo-tube signal. This OR gates a LeCroy 2249 charge ADC which acts as a register, latching the logic state of four input lines which indicate whether or not the scintillator, the HP Ge detector, the muon veto, and a mercury pulser (described below) were active during the event, i.e., within 100 ns of each other. This module generates a CAMAC LAM which then causes a readout of the register ADC, a peak-sensing LeCroy 3511 ADC used to measure the HP Ge energy, a LeCroy 2249W charge ADC used to measure the scintillator energy, and a LeCroy 2228A TDC which measures the time difference between the HP Ge detector and the photo-tube. A mercury pulser (Ortec 448) firing at about 1 Hz is fed into the test input of the germanium detector preamp and used to monitor the stability of the preamp and shaping amplifier (Ortec 671) gain. The Ge detector calibration was found to be stable to better than 1% during all data runs.

3.2. Data analysis

The germanium singles rate, or $N_{\gamma 511}$, is determined by fitting a Gaussian plus a linear background to the 511 keV spectral peak and integrating the Gaussian. The no-source background, obtained by filling the cell with fresh scintillator, is subtracted, but this contribution is typically about 1% for the activities used. In this analysis, muons and pulser events are rejected by placing the appropriate cuts on the hit register, but any light deposit in the scintillator is disregarded. The photo-tube singles count rate, or N_s , is determined by integrating the measured beta spectrum over some energy range regardless of the germanium detector hit condition. Again, muon events are removed in this analysis and the no-source background is subtracted. Finally, the coincidence counting rate, or N_c , is determined by integrating the beta spectrum over an adjusted energy interval using the data subset generated by requiring that 511 keV be deposited in the germanium detector within 30 ns of the scintillator's firing as measured by the TDC. The source-correlated background is subtracted. It is due to Compton scattering of 1.077 MeV gammas. To estimate this contribution, a window is set in the germanium spectrum of equal width to the width of the 511 keV peak, but displaced by one peak width. A coincidence data set is then generated and subtracted. This source-related background is generally on the level of 2–3%. These three rates are then used to calculate the source activity as described above. The activity calculation was found to be independent of the scintillator energy interval used in the analysis, as expected.

In the analysis we also have to take into account that the average energy deposited in the liquid scintillator by the annihilation radiation is different for the single and coincidence data set. In fact, it turns out that a shift of the energy scale is all that is needed to correct for the removal of one annihilation quanta by requiring its full absorption in the Ge detector. As the energy and angular distribution of the anni-

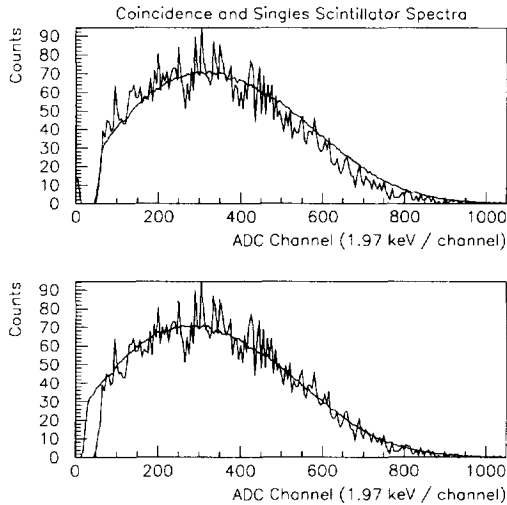


Fig. 2. The top figure illustrates the relative shift between the singles (smooth curve) and the coincidence spectra of ^{68}Ge . The singles spectrum has been normalized to the same area as the coincidence spectrum by multiplying by a factor of $\frac{1}{1.37}$. Both spectra are rebinned by a factor of five for clarity. The lower figure illustrates the match of the two spectra after shifting the singles spectrum by 33 channels (65 keV).

hilation radiation is completely independent of the positron energy, on average all positron energies are affected identically by the absorption of gamma energy. We use a maximum likelihood fit to determine the shift between single and coincidence scintillator spectra. The analysis interval of the coincidence data set is adjusted according to the fitted shift. The correction of the different absorption probabilities is typically in the -5% range. The error of this estimate is folded with the statistical errors assuming that they are independent. Fig. 2 illustrates this spectral adjustment.

3.3. Precision and reproducibility

The validity of the method was demonstrated using a calibrated ^{22}Na point source, placed between the acrylic tank, filled with pure scintillator, and the Ge detector. As the source was encapsulated, the positron energy could not be measured, instead we used a coincidence between the 1.275 MeV gamma line (measured with the scintillator) and the annihilation radiation (registered by the Ge detector). Our data analysis of the source activity was in good agreement with the nominal source strength.

Tests of the precision were made using ^{68}Ge . A cell was filled with scintillator to which a small quantity of ^{68}Ge had been added, and measurements of its activity were made. The source remained in the tank for several days during which repeated measurements were made to investigate the reproducibility of the activity measurement. Fig. 3 summarizes the results. The calculated activity showed good reproducibility. The fluctuations found in the results of the repeated measurements confirm that the statistical error is on the $\pm 1\%$ level as estimated in the error propagation.

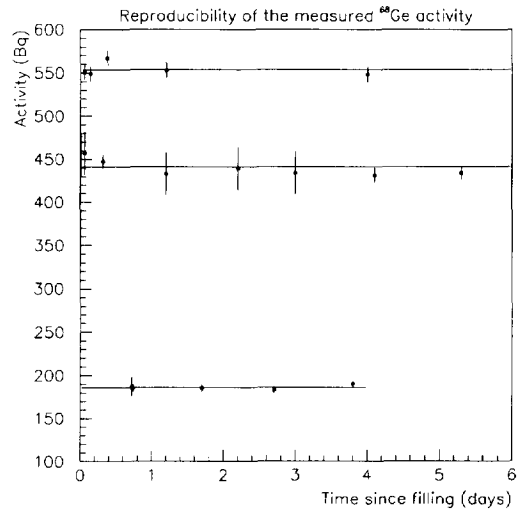


Fig. 3. Reproducibility of the ^{68}Ge activity measurements. Measurements made over several days during three separate runs are displayed. The average value is plotted as a horizontal line for different activities.

4. Homogeneity of the source distribution

Determinations of how much of the source activity was attaching to the walls of the acrylic container were made using the described calibration system. The source, dissolved in liquid scintillator, was placed into a virgin acrylic test tank where it remained for a length of time ranging from an hour to a month in various runs. The activity of the source was calibrated by the above method, and the scintillator was then dumped, the acrylic tank washed with clean scintillator, and then refilled with fresh, unloaded scintillator. The activity remaining in the tank was then again determined to estimate the fraction of the source bound to the cell walls. Measurements of this type were performed both for the $^{68}\text{GeCl}_4$ source and for the tetra-*n*-butylgermane sources. The source sticking was investigated as a function of the time spent in the cell and of the germanium carrier concentration in the scintillator sample. The results of these measurements are summarized in Table 1. It is clear from the table that the

Table 1
Investigation of ^{68}Ge source chemical stability. Source type **A** is the commercial source based on GeCl_4 . Type **B** is the tetra-*n*-butylgermane source synthesized at Caltech.

Source type	Ge concentration [ppb]	Exposure time [days]	Fraction sticking [%]
A	635	0.2	2.4 ± 0.6
A	1540	1.9	3.7 ± 0.4
A	276	4.0	21.6 ± 0.6
A	89	28	40 ± 1
B	938	4.0	0.11 ± 0.03
B	137	4.0	0.37 ± 0.5
B	3.3	4.2	1.9 ± 0.2
B	4480	5.3	0.15 ± 0.04

tetra-n-butylgermane source shows markedly better stability in the tank and a very low rate of sticking to the walls.

For reference, the calibration of a detector element of the neutrino detector involves inserting about 1 kBq of ^{68}Ge activity into 200 liters of scintillator, where it would remain for about one day. Using the source described above (370 Bq/ml and 213 ppm Ge) results in a germanium concentration of about 3 ppb. Of course, the concentration to activity ratio can be adjusted arbitrarily. We have shown, through our measurements, that the tetra-n-butylgermane source remains in solution for a sufficient length of time. From Table 1 it is also clear that a neutrino measurement could not be based on the $^{68}\text{GeCl}_4$ source as a significant portion would adhere to the walls of the detector. The geometrical distribution would hence not resemble the neutrino signal.

5. Physical properties of the liquid scintillator

It is important to verify that the Ge loading into the scintillator does not significantly affect its physical properties, most critically, the light yield and attenuation length. To study the properties of the Ge-loaded scintillator we prepared a third tetra-n-butylgermane sample containing 120 ppm of stable Ge carrier. The measurements were done with a diluted scintillator sample containing 2 ppm Ge. This concentration is about a factor 700 higher than will be typically used for the calibration. The light attenuation length and light yield of this loaded scintillator was measured and compared to that of the unloaded scintillator. No significant differences were measured. The measurements are described in the following sections.

5.1. Attenuation length

The attenuation length was measured using the light from a blue LED, driven by a pulser, passing through a 440 nm filter and then emerging through a small pinhole. The rays are focused to parallel by a 7-cm focal-length lens placed one focal length away from the pinhole. The parallel beam shines down a 1.5 m hollow aluminum pipe with an inside diameter of about 3 cm. At the bottom of the pipe is a glass window behind which a photo-multiplier tube detects the emerging light. Liquid scintillator is filled into the aluminum pipe, and the response of the photo-tube as a function of the height of the liquid gives a measure of the attenuation length at 440 nm. Data are acquired using a peak-sensing ADC gated by the trigger output of the pulser driving the LED. This method of gating is used to minimize the background from actual scintillation light produced by natural radioactivities. This background was measured with the tube full of scintillator by triggering on the pulser, but unplugging the LED, and was found to contribute less than a part in 10 000. Measurements of the LED peak position were made at liquid heights of 20, 40, 60, 80, and 100 cm. The liquid was

then drained out and measurements were made at 90, 70, 50, 30, and 20 cm. This staggering of points improved the immunity of the measurement to tube gain drifts or changes in LED light output. We found light attenuation lengths of 8.1 ± 0.9 m and 8.8 ± 1.1 m for the loaded and unloaded scintillator, respectively.

5.2. Light yield

The light yield of the Ge-loaded and unloaded scintillators were compared using a Pyrex petri dish, about 2 inches in diameter and 0.5 inch deep which was optically coupled to a three-inch photo-tube, oriented vertically. 20 ml of scintillator was then pipetted into the dish and a ^{207}Bi source, which emits 1 MeV conversion electrons, was placed above the scintillator. The photo-tube output was then read out into a charge ADC. By comparing the position of the electron peak, which is proportional to the number of photoelectrons produced, using the Ge-loaded and unloaded scintillators, one can compare their relative light output. However, any change in the photo-tube gain between the two measurements would cause a shift of the peak position unrelated to the light yield. To monitor the tube gain, an LED, driven by a pulser, was positioned to shine onto the photo-tube. In this way we measured light yields of 151 ± 3 pe./MeV (photoelectrons/MeV) and 155 ± 3 pe./MeV for the Ge-loaded and unloaded scintillator, respectively. We conclude that the light yield of the scintillator is unaffected by a 2 ppm Ge loading.

6. Conclusion

We have developed a ^{68}Ge source which can be dissolved in pseudocumene and mineral-oil-based liquid scintillator. We have worked out a method which allows us to calibrate the activity of extended radioactive scintillator samples to 1% accuracy. We present experimental evidence that neither the light attenuation length nor the light yield of the scintillator is affected by the Ge loading. We have shown that the ^{68}Ge remains in solution if used in form of tetra-n-butylgermane. Thus we have demonstrated that such a source is well suited for measuring the positron detection efficiency of a segmented liquid scintillation neutrino detector.

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