Beta spectrum of ¹¹⁵In

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Liquid scintillator solutions based on the solvent phenethyl alcohol are loaded with the salt indium trifluoroacetate to concentrations as high as 200 g In/l. Using a scintillator solution loaded with 51 g In/l in a low background chamber with muon anticoincidence and Pb walls 20 cm thick, a β^- decay spectrum from ¹¹⁵In is obtained and fitted with a sixth order polynomial in $(E_{endpoint} - E)/E_{endpoint})$. From these data we find $T_{1/2}(^{115}In) = 4.41(25) \times 10^{14}$ yr and $E_{endpoint}(^{115}In) = 482(15)$ keV. These low background experiments also show that indium loaded liquid scintillators are excellent neutron detectors. Their sensitivity for epithermal neutrons in particular is excellent. It is expected that these results will be useful in designing a solar neutrino capture detector of the type suggested by Raghavan.

$\begin{bmatrix} \text{RADIOACTIVITY} & ^{115}\text{In; measured } T_{1/2}, E_{\beta}. & \text{In-loaded liquid scintillator, } 90 \\ & \text{keV FWHM at 390 keV.} \end{bmatrix}$

I. INTRODUCTION

This work on the β^{-} decay of ¹¹⁵In is an outgrowth of a program at Bell Laboratories to develop a detector of solar neutrinos based on the neutrino capture reaction, ${}^{115}In(\nu_e, e^{-}){}^{115}Sn^*$. As pointed out by Raghavan (Ref. 1), ¹¹⁵In provides a unique ν detection signature which could be the basis for a direct counting low threshold detector. Unlike radiochemical experiments such as ${}^{71}\text{Ga}(\nu, e^{-}){}^{71}\text{Ge}$ which have been proposed² to measure the integrated flux of solar p-p neutrinos, an ¹¹⁵In detector could count neutrinos directly and would be capable of yielding the energy spectrum of the detected solar neutrinos, thus distinguishing in a single experiment the p-p neutrinos from the ⁷Be solar neutrinos. In this paper we report on the properties of indium loaded liquid scintillators and on the properties of the ¹¹⁵In β decay. The parts of this work relating more directly to neutrinocapture counting are reported elsewhere.^{3, 4, 5}

The natural β decay of ¹¹⁵In is an unavoidable background in any direct counting detector containing indium, and may be of major importance in an indium loaded ν -capture detector due to the low data rates expected. To evaluate specific detector designs it is necessary to know the ¹¹⁵In lifetime, and the endpoint and shape of its β spectrum. To measure these properties requires a detector that is heavily loaded with indium in such a way that all of the decay particles are recorded with good energy resolution. We have considered as possible detectors: scintillators, or semiconductor charge sensitive detectors with high indium content, and gas proportional counters with indium coatings on the walls or loaded into the gas. Of these the most convenient for our initial studies appeared to be a liquid scintillator loaded with a soluble salt of indium. The next section will describe the properties of our indium loaded scintillator, and Sec. III will report on the use of these loaded liquids to measure the β decay spectrum of ¹¹⁵In.

II. INDIUM LOADED SCINTILLATORS

The most light efficient liquids⁶ contain no metal loading and are based on the pure solvents xylene or toluene with the addition of one or more fluorescent compounds such as PPO (2,5 diphenyloxazole) that emit light from short lived states at a wavelength matched to the photomultiplier sensitivity. The short lifetime of these optical states reduces the opportunity for radiationless quenching by impurities in the solution. The introduction of indium into such solutions is a difficult technical problem because xylene and toluene are very poor solvents for the ionic indium compounds which are the most common and stable indium derivatives. The organoindium compounds (trimethylindium, etc.) do dissolve in xylenes, but these compounds tend to be unstable, sensitive to moisture, and spontaneously flammable in air, and thus are clearly not suitable for a large detector assembly. Beard and Kelly⁷ used a liquid scintillator containing 1% In by weight in the form of indium trihexanoate acid. It was our hope to load a liquid scintillator with considerably more indium.

Because it is most convenient to work with ionic indium compounds, we sought a solvent that would be both a good primary scintillator and able to dissolve a large amount of salt. Alcohols such as

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benzyl alchohol appear to be the best choice. The incorporation of a toluenelike π -electron system offers the potential for efficient scintillator action, and dielectric constant (13) is high enough to dissolve ionic compounds. The strong binding of hydroxyl groups to In^{+3} also suggests that alcohols would be good solvents.

In order to have a stable solution it is necessary to use the In^{+3} salt of a strong monovalent acid, HX, that will not undergo chemical reactions of the type $InX_3 + 3ROH - 3RX + In(OH)_3$. This reaction is likely if, for example, X = Cl. The product, benzyl chloride, is a good electron trap and it is also an effective scintillation quencher. Indium acetate is not appreciably soluble in benzyl alcohol but indium trifluoroacetate, $In(CF_3CO_2)_3$, is quite soluble.

Solutions of indium trifluoroacetate in benzyl alcohol proved to be good scintillator solutions when various secondary fluors were added. The efficiency decreases as the indium concentration



FIG. 1. Dashed curve: scintillation efficiency of phenethyl alchol with 6 g/l PPO (2, 5-diphenyloxazole) and 0.6 g/l Bis-MSB [p-bis(o-methylstryryl) benzene] as a function of indium concentration in the form of indium trifluoroacetate. Solid curve: same as above but with 5% of the solvent replaced with ethylene glycol. The solutions were tested in a 93 ml quartz cell coated with BaSO₄ reflector and coupled to a 5 cm diam. quartz EMI 9750QR photomultiplier. The 478 keV Compton edge from a ¹³⁷Cs γ -ray source was used to determine the light yields relative to a solution of p-xylene plus 5 g/l butyl PBD [2-(4-biphenyl)-5- (p-tert-butylphenyl)-1, 3, 4-oxadiazole]. This p-xylene solution is the most light efficient liquid scintillator known.

is increased. This is to be expected on the basis of several modes of interaction. First, In^{+3} is expected to be a moderately effective heavy-atom fluorescence quencher and further, as a moderately acidic ion, it may complex with the weakly basic secondary fluors. It may also be an electron scavenger; any compound of the latter type is a quencher.

The use of trifluoroacetate also gives us the advantages of a salt that is relatively easy to prepare in the anhydrous form, to prevent hydrolysis and formation of the insoluble oxide or hydroxide, and one that can be freed from excess acid by storage in a vacuum oven containing strong alkali. This is important because protons are very efficient scintillation quenchers.

Benzyl alcohol has one disadvantage that caused us to use phenethyl alcohol, the next higher alcohol and a poorer solvent. The oxidation of benzyl alcohol to benzaldehyde occurs fairly readily in the presence of air and the latter is a potent



FIG. 2. Measured spectra using a 1.25 l cell with 51.2 g In/l scintillator. This solution has a density of 1.142 g/cm³ and contains the proportions: 1319.1 g phenethyl alchol, 69.9 g ethylene glycol, 301.1 g indium trifluoroacetate, 8.0 g PPO (2,5-diphenyloxazole), and 0.8 g Bis MSB [p-bis(o-methylstyryl) benzene]. The ¹¹³Sn and ²⁰⁷Bi conversion electron sources were put in the scintillation liquid by means of thin-walled tubular needles of glass with 10 μm thick walls. The energy scale for this and the other spectra in this work were calibrated individually for each solution using as the three reference energies, the 1063 keV conversion sum peak of ²⁰⁷Bi, the 393 keV conversion sum peak of ¹¹³Sn, and either the 75 keV or the 24 keV x-ray photopeak from these sources. The energy calibrations included a quadratic term to account for scintillator nonlinearities.

quencher. This type of degradation is far less likely to be a problem with phenethyl alcohol and we chose it in the interest of long-term stability. In order to improve the solubility of indium trifluoroacetate in phenethyl alcohol, we added a small amount of ethylene glycol to complex some of the In^{+3} .

It is conceivable that other indium salts can serve in place of indium trifluoracetate but it will be difficult to obtain such high indium concentrations. The possible improvement in solution clarity is a compensating consideration.

Figure 1 shows the light efficiency of our phenethyl alcohol scintillator solution as a function of indium concentration. The light yield falls roughly exponentially with increasing indium concentration, reaching e^{-1} at 80 g In/l, an improvement over Beard and Kelly of an order of magnitude in indium loading efficiency. Figure 2 shows measured spectra from three radiation sources (²⁰⁷Bi, 113 Sn, and 51 Cr) taken with a 1.25 liter fused silica cell containing 51.2 g In/l in the phenethyl alcohol scintillating solution. The quartz cell has a diameter of 13.5 cm is 9.8 cm long, and is coated with BaSO₄ white reflector paint. One EMI 9791 KA photomultiplier tube is optically coupled to each of the two ends of the cylindrical cell. The energy pulses from the two tubes were summed and stored in a 1024 channel analyzer gated by coincidences $(2\tau = 100 \text{ ns})$ between the two anode pulses.

The ⁵¹Cr spectrum illustrates the photopeak efficiency of the detector for a 320 keV γ -ray. The Bi²⁰⁷ spectrum shows the Pb x-ray photopeak at 75 keV and the 8% conversion line at 1.063 MeV. The ¹¹³Sn spectrum shows the In x-ray photopeak at 24 keV and a strong 49% conversion line at 393 keV. The conversion electron sources were put



FIG. 3. The measured full width at half maximum (FWHM) energy resolution as a function of the excitation energy for two indium loaded scintillator solutions.



FIG. 4. Raw β spectrum obtained with 51.2 g In/l scintillator and background spectrum obtained with 51.2 g Sn/l loaded scintillator. Inset shows the ¹¹⁵In-¹¹⁵Sn nuclear energy levels.

in the liquid at the center of the quartz cell by means of thin walled tubular needles of glass 200 μ m diam. by 10 μ m thick. Such spectra allow a direct measure of the resolution of the indium loaded liquid. Figure 3 shows the measured FWHM resolution in keV vs the excitation energy in keV for this solution and also for a 100.2 g In/l solution.

III. BETA SPECTRUM OF ¹¹⁵In

A potentially serious problem in using ¹¹⁵In as a detector of solar neutrinos arises because ¹¹⁵In is naturally β ⁻ unstable, decaying to the ¹¹⁵Sn ground state (see Fig. 4 inset). To evaluate the background to be expected from this natural radioactivity we have measured the 115 In β spectrum. The cell filled with a 51.2 g In/l solution was placed in a Pb brick enclosure with bottom and walls 20 cm thick and a 10 cm thick top. The two photomultipliers were counted in coincidence with one another and in anticoincidence with a 100 cm $\times 40 \text{ cm} \times 1.25 \text{ cm}$ thick muon sensitive detector resting on the top of the Pb cover, 25 cm above the center of the quartz cell. Figure 4 shows the energy spectrum obtained with the indium loaded solution along with a background spectrum which was obtained by replacing the In loaded solution with a solution containing 51.2 g Sn/l in the form of tetrabutylin in xylene and napthalene. A characteristic feature of both spectra is the prominent peak at about 50 keV. This low energy



FIG. 5. In-Sn difference spectrum obtained by subtracting from the 51.2 g In/l spectrum (in Fig. 4), the spectrum of an unloaded phenethyl alcohol scintillation solution; then subtracting from the 51.2 g Sn/l spectrum (in Fig. 4), the spectrum of an unloaded paraxylene scintillation solution; and finally by subtracting this Sn difference spectrum from the In difference spectrum. To do these subtractions the spectrum from each scintillation solution was individually calibrated at three energies using the ²⁰⁷Bi and the ¹¹³Sn conversion sources as described in the caption of Fig. 2. This involved computing for each data point of each spectrum the keV/channel and the corresponding (counts/s)/keV. Because indium and tin have similar atomic numbers, the two metal loaded solutions should have similar detection efficiencies for background γ rays and charged particles. In this sense, this difference spectrum is expected to be a measure of the radioactivity associated with the indium alone.

peak occurred in all of our liquid scintillation spectra, in both the loaded and the unloaded solutions, so it is not connected with the heavy metal loading, and it can be removed from the data by subtraction as was done in Fig. 5, below. We did not establish the origin of this 50 keV peak, but we suspect that it may be due to the ¹⁴C in all our organic solvents. The ¹⁴C β^- decay is characterized⁸ by a 5700 year half-life, a 156 keV endpoint, and a 45 keV average β energy.

In order to characterize the background more completely, spectra were also taken with a 97 g Sn/l solution and with unloaded solutions of phenethyl alcohol and *p*-xylene. These spectra show that the background depends only weakly on the heavy metal concentration and the solution density. Figure 5 is a difference spectrum which is believed to be a measure of the indium radioactivity. It was obtained by subtracting from the 51.2 g In/l spectrum, the spectrum of its solvent, and then subtracting from the 51.2 g Sn/l spectrum the spectrum of its solvent. The Sn difference spectrum was then subtracted from the In difference spectrum to give the spectrum of Fig. 5. Certain features of this spectrum are not characteristic of what one would expect from a pure β^- decay. The sharp rise below 50 keV is attributable to inexact cancellation of phototube noise and the low energy peak is (see above) due to slight inaccuracies in the relative energy calibrations at low energies. This is not unexpected. However, there is a sharp peak at ~160 keV which is unlike a β spectrum; and finally, the counting rate does not go exactly to zero at high energies (see \times 10 scale expansion of Fig. 5). It thus appears that there is some extra source of radioactivity present causing a background which our subtraction technique missed.

After a fruitless search for radioactive contaminants we saw from studying the density dependence of the background that the extra background was proportional to the amount of In at low concentrations, and saturated at high In concentration. This suggested a neutron activated background due to a parent with such a high neutron cross section



FIG. 6. γ rays characteristic of the decay of 54 min ¹¹⁶In. Both spectra were obtained in the low background Pb enclosure with a 5×5 cm NaI(Tl) detector adjacent to and counting in coincidence with the 51.2 g In/l scintillation liquid in the 1.25 l cell. The upper data were obtained with a neutron source near but external to the Pb chamber. The γ -ray photopeaks at 417 keV, 1090 keV, and 1293 keV are clear evidence that the In loaded scintillator is sensitive to neutrons producing 54 min ¹¹⁶In. The lower data obtained without an external neutron source show that some 54 min ¹¹⁶In is produced by ambient neutrons that are created in and pass through the walls of the Pb chamber.

that the In loaded scintillator was black to neutrons for concentrations above 50 g l. The thermal neutron absorption cross section for 95.7% abundant ¹¹⁵In is ~200 b, and its epithermal resonance integral is ~3300 b.⁹ The levels of 116 In which would be produced by neutron absorption by ¹¹⁵In are highly excited states (several MeV) that decay by prompt γ -ray emission leading eventually to the ¹¹⁶In ground state. The ¹¹⁶In ground state then β^{-} decays with $T_{1/2} = 54$ min to ¹¹⁶Sn, releasing ~3.3 MeV in one of several β - γ cascades. This β^- and γ spectrum could account for the failure of the background to vanish in Fig. 5. Moreover, there is a highly converted 164 keV ¹¹⁶In transition⁸ with $T_{1/2} = 2.2$ s which precedes the β decay and could account for the sharp peak

To test whether our detector is indeed sensitive to neutrons, a 5×5 cm NaI(Tl) detector was put in the Pb chamber next to the In loaded scintillator cell to search for the characteristic γ rays from In¹¹⁵ (n, γ) reactions. In Fig. 6 the lower data are the NaI(Tl) spectrum obtained in coincidence with pulses from the In scintillator cell; the upper data are a similar coincidence spectrum obtained with a Be (α, n) neutron source placed near the Pb casket. The appearance in the upper data of the ¹¹⁶In decay photopeaks expected⁸ at 417 keV (36%),

at low energies in Fig. 5.



FIG. 7. Neutron induced activity in the 1.25 l cell filled with 51.2 g \ln/l scintillator solution. To obtain this spectrum the conditions were those of the 51.2 g \ln/l spectrum of Fig. 4, except that a Be (α, n) source was placed near but external to the Pb chamber. The spectrum shown is a difference spectrum of this scintillator obtained with and without the external neutron source.

1090 keV (53%), and 1293 keV (80%) proves that our detector is sensitive to neutrons. The similarity in the overall shape of the upper and lower curves and the clear presence of the 1090 keV photopeak in the lower data confirms that ambient neutrons are present which are causing an extra background by activating the In.

Figure 7 shows the scintillation spectrum induced by neutron capture from the external Be (α, n) source placed near the 1.25 l cell loaded with the 51.2 g/l indium solution. This spectrum is a measure of only the neutron induced activity in the scintillator liquid; the activity not associated with the external neutron source has been subtracted. The Pu-Be neutron source was located ~70 cm from the Pb enclosed In-loaded scintillator and was contained in a paraffin shield 17 cm thick. Its strength was estimated to be ~1.2×10⁶ n/s into 4π . The total activity (in Fig. 7) induced by the neutron source was 399 s^{-1} , which corresponds to ~300 neutron captures/second by the In scintillator after correcting the multiplicity effect of the 165 keV γ -ray decay. This agrees well with a simple solid angle estimate of 350 n/s (at the In scintillator) made by ignoring the effects of the Pb shielding and assuming a 6 cm attenuation length for the neutrons in the paraffin.

Assuming that the energy spectrum of the ambient neutrons and those from the Pu-Be source are similar, Fig. 7 can be used to correct Fig. 5 for the activity associated with the capture by ¹¹⁵In of ambient neutrons. Taking the ratio of the counts in the three signature photopeaks of the two spectra of Fig. 6 suggests that 1% of the spectrum of Fig. 7 should be subtracted from the spectrum of Fig. 5. In practice, enough of the Fig. 7 spectrum was subtracted to normalize the spectrum of Fig. 5 to zero counts between 1400 and 1500 keV. This required 0.91% of the spectrum of Fig. 7. This neutron corrected difference spectrum is shown in Fig. 8 over the range 0-800keV. The high energy portion of the spectrum was statistically consistent with zero counts over the entire range 550 to 1500 keV. We take this as additional evidence that the neutron sensitivity of the In accounts for the extra background seen in Fig. 5. Note also that the low energy peak seen in Fig. 5 has disappeared in this spectrum; it was clearly due to the 164 keV neutron-induced 2.2 s γ ray of ¹¹⁶In*. Under the assumption that there is no remaining background at low energy (from, e.g., ¹⁴C), the data points in Fig. 8 are the β decay spectrum of ¹¹⁵In as seen by a β ⁻ detector with the energy resolution of Fig. 3.

The total area of the envelope of count rate in the β spectrum of Fig. 8 (and in two other in-



FIG. 8. The β decay spectrum of ¹¹⁵In. The experimental points are obtained by correcting the In-Sn difference spectrum of Fig. 5 for the effect of ambient neutrons on In shown in Fig. 7. After this correction was made adjacent data channels were combined for improved clarity. The solid line is a polynomial fit to the data; the dashed line is the fit with the energy resolution of the In-loaded scintillator unfolded as described in the text. The fit shown was generated using the double Gaussian resolution function and the coefficients $a_2 = +0.0244$, $a_3 = -0.0237$, $a_4 = +0.0044$, $a_5 = -0.0038$, $a_6 = +0.0008$, and $E_{endpoint} = 467.8 \text{ keV}$.

dependent spectra not shown) is 16.0(9) counts/s from a total of 64.0 g indium (61.3 g ¹¹⁵In). Thus our measurement of the ¹¹⁵In decay half-life is $T_{1/2} = 4.41(25) \times 10^{14}$ yr. This value is lower than but consistent with the result of Watt and Glover,¹⁰ and also lower than the less accurate work of Beard and Kelly.⁷

The β endpoint energy cannot be obtained directly from Fig. 8 without taking into account the energy resolution of the scintillator. This is done in the dashed curve of Fig. 8 which shows the shape of the ¹¹⁵In β ⁻ decay spectrum deconvoluted from the energy resolution of the scintillator. To obtain this deconvolution the energy shape of the β decay spectrum, N(E), was approximated as a sum of orthogonal polynominals $(E_{endpoint} - E)/E_{endpoint}$

$$N(E) = \sum_{n=2}^{6} a_{n} P_{n} \left[\frac{E_{\text{end point}} - E}{E_{\text{end point}}} \right] E < E_{\text{end point}}$$

$$N(E) = 0, \quad E \ge E_{\text{endpoint}}$$

where

$$\begin{split} P_2(x) &= \sqrt{5}x^4, \\ P_3(x) &= 6\sqrt{7} \left[-\frac{5}{6}x^2 + x^3 \right], \\ P_4(x) &= 84 \left[\frac{15}{28}x^2 - \frac{3}{2}x^3 + x^4 \right], \\ P_5(x) &= 398 \left[\frac{7}{24}x^2 + \frac{7}{5}x^3 - \frac{21}{10}x^4 + x^5 \right], \\ P_6(x) &= 1787 \left[\frac{14}{99}x^2 - \frac{56}{55}x^3 + \frac{26}{11}x^4 - \frac{8}{3}x^5 + x^6 \right]. \end{split}$$

This spectral shape function N(E) was then folded with an energy resolution function R(E, E') characteristic of our scintillation apparatus,

$$f(E) = \int R(E, E') N(E') dE'$$

and the resulting function f(E) was fitted to the background subtracted experimental data of Fig. 8 and two other similar data runs.

Unfortunately, the resolution function R(E, E') of the apparatus is not known and is not readily measurable. To examine the sensitivity to R(E, E'), the data were analyzed two ways. For the first approach R(E, E') was assumed to be a simple Gaussian

$$R(E, E') = \frac{1}{2\pi\sigma^2(E')} \exp \left[-(E-E')^2/2\sigma^2(E')\right].$$

Here $\sigma(E) = 5.16\sqrt{E/2}(2 \ln 2)^{1/2}$, and is obtained by fitting the 51.2 g In/l data of Fig. 3 to a straight line. With this method Fig. 8 and the 2 other sets of similar data yielded a mean endpoint of 492.7 (13.6) keV, where the error quoted reflects the distribution of the three runs about the mean.

A slightly better fit was obtained by assuming R'(E, E') is the sum of two Gaussians



FIG. 9. Kurie plot of the β decay spectrum of ¹¹⁵In. Every point (x, y') on this plot was obtained from the corresponding point (x, y) of Fig. 8 by the transformation

$$y' = [y/(x+m_0c^2)(x^2+2xm_0c^2)^{1/2}F]^{1/2}$$

where F is the Fermi function for Z = 49. The meaning of the solid and dashed lines is as in Fig. 8. Background corrections are significant for the points below 120 keV (compare Figs. 4 and 5 with Fig. 8). For this reason the low energy data in this figure and in Fig. 8 may be subject to systematic uncertainties.

$R'(E, E') = \frac{4}{5}R(E, E') + \frac{1/5}{8\pi\sigma^2(E')} \\ \times \exp[-(E-E')^2/8\sigma^2(E')],$

with $\sigma(E)$ the same as before. With this double Gaussian assumption the mean endpoint becomes 470.6(5.2) keV, and again the quoted error reflects the statistical distribution of the three runs about the mean.

The difference between these values is an indication of the systematic error due to our ignorance of the details associated with the resolution function. Other systematic effects include small uncertainities in the energy calibrations. Our measured ¹¹⁵In β ⁻ endpoint then is taken to be the mean of these two methods, 482(15) keV, where the quoted error is believed to reflect the systematic as well as the statistical uncertainties. The ¹¹⁵In spectrum is shown as a Kurie plot, ¹¹ [N(E)/ $p(E+m_0c^2)F(Z)]^{1/2}$ vs E, in Fig. 9. The isotope ¹¹⁵In has a fourth forbidden nonunique β spectrum, and this figure represents the first such spectrum to be obtained with sufficient accuracy to show a departure from a straight line Kurie plot. The detailed shapes for transitions of this type have not yet been calculated.

IV. DISCUSSION

From the point of view of designing an indium loaded solar neutrino detector, this work demonstrates the following points: Useful scintillator solutions with indium concentrations in excess of

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100 g/l are achievable, but as the indium content increases both the light output and the energy resolution decrease. Other scintillator liquids and indium salt combinations may, of course, give better results. The decrease in scintillation light with increasing indium concentration is due to both increased quenching of the primary scintillations and to the reduced optical transparency of the loaded solutions. In a large volume ν -capture detector the transparency problem takes on increasing importance. We measure for the ¹¹⁵In half-life $4.41(25) \times 10^{14}$ yr and for the β endpoint 482(15) keV. Both of these numbers are somewhat lower than, but not in actual disagreement with, literature values. Finally, it is clear that these indium loaded liquids are marvelously sensitive neutron detectors. Indeed, no element in the periodic table has as large a resonance integral as does indium⁹ for epithermal neutrons between 0.5 eV and 15 MeV. The relatively high and stable indium loading achieved in this work suggests that neutron detectors based on these indium loaded scintillator liquids are practical. From the point of view of a ν -capture detector, the high neutron sensitivity means that reasonable care must be taken to shield any indium based solar neutrino detector array from neutrons.

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