Letter to the Editor

Improving the performance of the liquid argon TPC by doping with tetra-methyl-germanium

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Abstract
In order to recover the charge lost by electron–ion recombination, we doped pure liquid argon with a photosensitive hydrocarbon, tetra-methyl-germanium (TMG), in the 3 ton ICARUS TPC. A charge increase of 25% to 220% was observed for different electric fields and for energy densities ranging from 1.6 to 32 MeV/cm.

The 3 ton liquid argon TPC has been in operation since May 1991 with pure liquid argon (LAr). In these three years we measured the basic parameters of the detector responses, such as the spatial resolution, the electron diffusion coefficient, the correlation of collected charge with electric field and energy density, by cosmic rays and external gamma ray sources [1]. In addition we have been continuously monitoring the stability of the liquid argon purity, the effectiveness of the recirculation system, and the reliability of the electronics read-out. So far no degradation of any part of this detector has been found.

As is well known, an ionizing particle in liquid argon will produce electron–ion pairs and excitons along the track. Depending on the ionization density and electric field, some of the pairs recombine and emit vacuum ultraviolet (VUV) photons with energy distribution peaked at 128 nm (9.7 eV). On the other hand, photon emission from excitons exhibits energy distribution peaking at the same energy (9.7 eV). With our data by minimum ionizing muons (mip), stopping muons and stopping protons, we have measured the collected charge as a function of energy density (1.6 to 32 MeV/cm) and electric field (100 to 500 V/cm) in pure liquid argon. We found that the electron escape probability depends heavily on these two parameters. The percentage of free electron yield can vary from 70% to 14% at different energy densities and electric fields. This nonlinear detector response may degrade the particle identification capability of the liquid argon TPC. A possible solution to improve the linearity of the detector response is to introduce photosensitive dopants able to convert part of the scintillation light, either from electron–ion recombination or by direct excitation, into additional free electron-ion pairs, thus enhancing the linearity as a function of the deposited energy density and electric field. We chose TMG as photosensitive dopant because of the following advantages:
1) TMG is not absorbed by the Oxisorb™ in the recirculation system. Allene, that could have been another candidate, is heavily absorbed by the Oxisorb.

2) TMG can be easily purified to an electron lifetime level better than 10 μs [2].

3) TMG has a large photoabsorption cross section of 62 Mbarn and has acceptable quantum efficiency [3].

The operation detail of the 3 ton liquid argon TPC is described in our previous paper [1]. In this detector the three dimensional track image is defined by the collection wire plane (x), the induction wire plane (y) and the electron drift time (z). The charge information is extracted from the signals on the collection wire plane which has a wire pitch of 2 mm and is horizontally oriented. The TMG used in this experiment has been supplied by Wiley Organics Co., USA, with a purity level of 99.9%. It was further purified by means of molecular sieves (4A + 13X) and vacuum distillation at −70°C and then finally transferred to a glass container at a temperature of about −50°C. The glass container was connected to the output of the Oxisorb in the recirculation system, where the TMG vapor is mixed with the purified argon gas and the mixture is liquefied to the bottom of the detector dewar. In Fig. 1 we show the vapour pressure of TMG measured as a function of temperature before doping. In the same picture we also show, as a comparison, the vapour pressures of TMS and TMSN [4]. As expected the TMG values lay in the range determined by these two hydrocarbons. The rate of TMG mixing with argon gas was controlled by varying the relative values of TMG vapour pressure to argon gas pressure. In order to increase the mixing rate, the TMG was heated up to about 80°C (surface temperature of the glass container) by an infrared lamp, corresponding to about 1.5 bar TMG vapour pressure. The purity of liquid argon was frequently measured both with the built-in laser lifetime monitor [5] and the wire chamber with crossing muons [6] during the whole doping period. The electron lifetime was always above 3 ms; this corresponds to an impurity concentration in liquid argon lower than 0.1 ppb.

Fig. 1. Measurement of the TMG vapour pressure as a function of temperature. The data of TMS and TMSN are reported from Ref. [3].

Fig. 2. Time evolution of (a) free electron lifetime and (b) charge yield measured by means of crossing muons at an electric field of 300 V/cm during the LAr doping with TMG.

Fig. 2a illustrates the lifetime evolution measured using crossing muon data.

The charge yield by mip muons in 2 mm was measured continuously and analyzed every 8 h at a fixed electric field of 300 V/cm. A 25% charge increase was observed already a few hours after doping TMG with 1.3 ppm (Fig. 2b). This encouraging result indicates that charge compensation by converting scintillation light to free electrons is really taking place. Fig. 3 illustrates the relation of the collected charge as a function of the energy deposition at an electric field of 200 V/cm and TMG concentration of 1.3 ppm and 3.5 ppm. Data come from the analysis of the stopping muon and proton events.

Fig. 3. Correlation between collected charge and deposited energy at an electric field of 200 V/cm and TMG concentration of 1.3 ppm and 3.5 ppm. Data come from the analysis of the stopping muon and proton events.
obtained from the analysis of the stopping muon and proton events at an electric field of 200 V/cm and TMG concentration of 1.3 ppm and 3.5 ppm. In this analysis (see Ref. [6] for details) the charge density is related to the energy density in the following way. First we obtained the $dQ/dx$ vs range relation and the $dE/dx$ vs range relation by reconstructing the experimental data and the Monte Carlo data describing the experimental set of events respectively. Hence the range is eliminated to obtain the $dQ/dx$ vs $dE/dx$ relation. It is evident that the linearity of the detector response is significantly improved. In fact at energy density of 1.6 MeV/cm, the number of collected electrons increase from 65% to 86% of the saturation value, while at energy density of 32 MeV/cm the increase is much more pronounced (from 14% to 46%).

Another important characteristic of TMG is its photoabsorption cross section. It determines the minimum TMG concentration level required in order to limit the spread of the electron–ion pairs produced by the scintillating light within the wire pitch of the liquid argon TPC. The high density tracks, identified as stopping protons, provide an opportunity to measure directly the photon mean free path (mfp). In this measurement, the high density proton track is a source which emits the VUV photons. We expect that the electron–ion pairs produced from VUV photons not only enhance the primary proton track but also produce new signals on a few wires adjacent to the stopping point of primary proton track. In the experiment, we did observe such signals at low TMG concentration. The pulse heights of such signals as a function of the wire positions from the stopping point of the proton track yield the mfp information of the VUV photons. By comparison of the experimental data with a Monte Carlo simulation, we determined values of the photon mfp of 5.5 and 2.5 mm at TMG concentrations of 1.3 and 3.5 ppm respectively. The average value of photoabsorption cross section, derived using the above results, is about 62 Mbar. This result is a factor of 2.4 smaller than the one reported by Anderson who achieved it, 147 Mbar, by a different method [7]. A deeper investigation about this discrepancy, including error analysis, will be reported in a following paper.

The stability of TMG concentration was continuously monitored at TMG concentrations of 1.3 ppm and 3.5 ppm by measuring the photon mfp. No significant change was observed after the recirculation system purified 3000 l and 5400 l of liquid argon in 25 and 45 days respectively. This result confirms that the Oxisorb operating in the recirculation system does not absorb the TMG molecules.

From the data presented in this letter, we can conclude that we observed for the first time the enhancement of the charge released by stopping cosmic muons and protons at various energy densities (ranging from 1.6 to 32 MeV/cm) in the ICARUS 3 ton LAr TPC doped with TMG. The performance of the detector is greatly improved and is remarkably stable in time.

References