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NEW PHOTOSENSITIVE DOPANTS FOR LIQUID ARGON*

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Abstract

Thirteen photosensitive dopants for liquid argon are presented, and the criteria for selecting prospective new dopants are discussed. A substantial improvement in energy resolution for 5.5-MeV alpha particles is measured in liquid argon when a photosensitive dopant is added.

1. Introduction

The addition of photosensitive dopants to liquid noble gases was first suggested by Policarpo[1] for the detection of UV photons originating outside of the liquid. When a material is dissolved in a liquid, its threshold for photoionization is lowered due to polarization of the solvent.[2] Thus it was hoped that liquid photoionization detectors could be constructed capable of detecting lower-energy photons than could be achieved with gaseous photoconverters.

The first successful doping of a liquid noble gas with a photosensitive material was reported by Anderson[3] where liquid argon (LAr) was doped with triethylamine (TEA) and trimethylamine (TMA). The objective of that work was to convert the scintillation photons from the LAr itself into detected charge. Working primarily with alpha particles, which yield a large quantity of recombination photons, an increase in collected charge of 30% and 60% was measured for dopings with TEA and TMA, respectively. An improvement in the energy resolution of the alpha particles was also measured, even under conditions where equal amounts of charge were collected in the doped and undoped LAr. In that work, it was also suggested that these dopants could be used with liquid xenon to convert proportional scintillation into charge amplification. This has been verified by S. Suzuki et.al.[4]

In this paper we will introduce eleven additional photosensitive dopants for LAr. The criteria for choosing these and prospective new dopants will also be discussed.

2. Choice of Dopants for Testing

The materials tested were chosen primarily by two criteria: their ionization potentials in the gas phase, I_g, and by their estimated solubility in LAr. The first criterion is straightforward. LAr produces UV photons due to excitons and recombination, with a mean wavelength of 3000Å (9.5eV).[5] A photosensitive material dissolved in LAr will have its ionization threshold lowered by about 0.7eV.[3] This sets an upper limit on I_g of about 10.2 eV. The only sample tested that did not meet this criterion was acetylene with I_g = 11.4eV. This was tested because acetylene and argon is a well-known Penning mixture in the gas phase, and it was hoped to see purely non-radioactive energy transfer.[6]

The estimation of the solubility in LAr of the materials to be tested was a little more difficult. Henry's law states that for nonpolar solvents the mole fraction of solute is proportional to its vapor pressure. Unfortunately, vapor pressures of organic materials are seldom available for LAr temperatures. Thus we were forced to extrapolate the existing data, usually for temperatures around room temperature, down to 90 K. When vapor pressure information was not available, we required the materials to have a low boiling point. We also assumed that polar materials would form colloids and freeze out, so materials with large known dipole moments were also avoided.

A third obvious criterion for choosing materials for testing is that they have a small electron affinity. Unfortunately, this information was not available for most materials, and thus did not play a large role in our decisions.

3. Experimental Setup

Most work was done with an ^{241}Am source providing 5.5-MeV alpha particles, detected with a 1.43mm ionization gap in the LAr. Since at 1.0kV mm^{-1}, about 90% of the electrons liberated by an alpha particle are lost due to recombination,[7] such a source is a very sensitive probe for the performance of photosensitive dopants. It is believed that the efficiency for photon production due to recombination in LAr is 100%. But, for the high charge densities from alpha particles, only 71% of this light escapes because of quenching[8] This quenching is not seen with the low charge densities from beta particles.

A ^{106}Ru source was also used, providing beta particles with a maximum energy of 3.5 MeV. The chamber used for the test with beta particles had a gap of 2.2mm and was followed by a second ionization chamber used in coincidence with the first. Triggering on beta particles that deposit a substantial amount of energy in the second gap allowed us to select minimum ionizing particles. Since, however, there is little recombination from beta particles, except at very low electric fields, this source was not very useful for evaluating dopants.

The argon used was condensed from the gas, taken from dewars of LAr. Only in two tests was the argon gas purified with a Hydrox purifier.

The dopants were added to the evacuated test dewar to a pressure of 5 to 100 Torr, depending on the material. The argon was then condensed. After a

measurement, a fraction of the doped LAr was removed and more argon was added. This was repeated several times, giving decreasing concentrations of dopant. A single dopant would typically be studied over a range of concentrations that varied by a factor of 100. The solubility was estimated to be the concentration where no colloids could be seen in the mixture. The dopants were used as received from the supplier without purification.

4. Test Results

The materials that we have found to be successful as photosensitive dopants for LAr are listed in Table I, according to their value of I_g. The dipole moment and estimated pressure at 90 K are also given. The charge collected for 0.1 kV mm^{-1} and 1.0 $kVmm^{-1}$ are tabulated, normalized to the charge collected from alpha particles in pure LAr at the same electric fields. The concentrations listed are those for which the best performance was measured. Table II lists the dopants that did not show an increase in collected charge. Their values of I_g , dipole moments, and estimated pressure at 90 K are also given.

Figure 1 shows the charge collected as a function of electric field for 5.5-MeV alpha particles in pure LAr and with several of the dopants. There are a variety of responses. Materials such as TEA dimethylether(DME) and TMA seem to drop in performance at higher electric fields when compared to isobutylene (iso-B) and tetramethylgermanium (TMG). The behavior of methyl mercaptan (CH_3SH) is different than any dopant tested, but its repulsive odor discourages a repeat of the measurement.

Figure 2 shows a typical response of a dopant as a function of concentration. At the highest concentrations, the charge collected at the lowest electric fields is slightly depressed. This is probably due to a slight increase in recombination due to the presence of the organic molecules. At considerably lower concentrations, the response is reduced at all electric fields. This is due to the range of the photon becoming a substantial fraction of the gap size.

The effect of purification of the argon can be seen in Fig. 3, where the respones for TMG and allene are shown for purified and non-purified argon. The improvement with purification is most pronounced at low electric fields.

The improvement in the response to beta particles is also shown in Fig. 3. The addition of TMG improved the charge collection at low electric fields. At high fields, the collected charge was only increased by about 8%. This is probably due to conversion of excitons and not due to recombination photons.

In our earlier work, we estimated that the photon conversion efficiency for TMA at a field of 1.0 $keVmm^{-1}$ was 16% (see comments added in proof). Thus, one can estimate the efficiencies of the other dopants in Table I at that electric field from Table I. Both TMG and allene have efficiencies of about 45%.

As before, we noted that the energy resolution for the alpha particles was improved when a photosensitive dopant was added. The energy resolution as a function of charge collected for 5.5-MeV alpha particles in pure LAr and LAr

doped with allene is shown in Fig. 4. One can see that the improved resolution cannot be attributed to the increase in charge collected with the dopants but to an improvement in the statistics in the collection of the electrons. The resolution of 10.2% FWHM in pure LAr was measured at 2.0kVmm^{-1} while the resolution of 6.1% FWHM in the allene doped LAr was measured at only 0.1 kVmm^{-1}.

5. Discussion

When comparing Tables I and II, one can see that the criteria for selecting potential photosensitive dopants are fairly sound. No successful dopant had an I_g above 10eV, and they all have fairly small dipole moments. Their estimated pressures at 90 K are all above 10^{-9} Torr, with the exception of tetramethyltin (TMT) and TMG. But, as we said, these pressures are only estimates. It should also be noted that the solubility is proportional to, and not equal to, the vapor pressure.

The materials in Table II all tend to have low estimated pressures implying poor solubility. Nitric oxide failed because it proved to have a high electron affinity. Benzene and tetrakis(dimethylamino)ethylene (TMAE), which were expected to work, are now seen to have failed because of their low vapor pressures.

6. Conclusion

We have presented thirteen photosensitive dopants for LAr. The data presented should not be considered definitive but simply a starting point for future work. For one thing, we have not addressed the problem of purifying the dopants. This has been shown to be important in the cases of TEA and TMA.[13] We have only studied the importance of purifying the argon used with two of the dopants. The objective of this work is to present a list of materials that are known to work, and to try to come to some understanding of the criteria for choosing new materials. A great deal more work is left to be done.

TABLE I

Properties of Photosensitive Dopants Tested

		I_g (eV) [a]	Dipole Moment debyes [b]	Estimated Pressure 90°K (Torr) [b]	Charge Collected* [c] (LAr≡1) 0.1kVmm^{-1}	1.0kVmm^{-1}	Concentration (ppm)
TEA	$(C_2H_5)_3N$	7.50	0.66	--	2.2	1.3	47
TMA	$(CH_3)_3N$	7.82	0.612	3×10^{-8}	3.4	1.6	110
TMT	$(CH_3)_4Sn$	8.25/8.76	--	4×10^{-12}	3.0	1.6	1.5
Cyclohexene	C_6H_{10}	8.95	--	--	2.1	1.3	3.6
1,3-Butadiene	C_4H_6	9.06	0	4×10^{-7}	4.6	1.9	17
Cis & Trans 2 Butene	C_4H_8	9.13	0(trans)	5×10^{-8}	3.6	1.6	72
TMG	$(CH_3)_4Ge$	9.2/9.29	--	3×10^{-10}	7.4(9.8)	2.6(2.7)	15
Isobutylene	C_4H_8	9.23	0.5	5×10^{-7}	4.9	1.8	16
Methyl Mercaptan	CH_3SH	9.44	1.52	2×10^{-8}	2.0	2.0	15
Pentene (Technical)	C_5H_{10}	9.5	--	1×10^{-9}	3.1	1.5	7
Allene	C_3H_4	9.53	0	1×10^{-5}	6.5(8.7)	2.5(2.7)	14
TMS	$(CH_3)_4Si$	9.86	0.525	8×10^{-9}	4.6	1.8	5.8
DME	$(CH_3)_2O$	10.0	1.30	5×10^{-8}	3.6	1.4	14

a) Ref. 9

b) Ref. 10

c) ( ) Purified LAr used

triethylamine (TEA), trimethylamine (TMA), tetramethyltin(TMT), tetramethylgermanium (TMG), tetramethylsilane (TMS), dimethylether (DME)

TABLE II

Properties of Unsuccessful Dopants Tested

Material		I_g (eV) [a]	Dipole Moment (debyes) [b]	Estimated Pressure 90 K (Torr) [b]
TMAE	$(C_{10}H_{24}N_4)$	5.36	--	10^{-22}
Propylamine	$(CH_3)_3N$	7.82	1.17	8×10^{-11}
DEA	$(C_2H_5)_2NH$	8.01	0.92	9×10^{-11}
1,3-Cyclohexadiene	C_6H_8	8.25	0.44	--
DMS	$CH_3)_2S$	8.69	1.50	2×10^{-9}
Benzene	C_6H_6	9.24	0	5×10^{-16}
Nitric Oxide	NO	9.25	0.16	3.3
Acetone	C_3H_6O	9.7	2.88	2×10^{-11}
Acetylene	C_2H_2	11.4	0	6×10^{-4}

a) Ref. 9; Ref 11-TMAE

b) Ref. 10; Ref. 12-TMAE

tetrakis(dimethylanino)ethylene (TMAE), diethylamine (DEA), dimethylsulfer (DMS)

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Figure Captions

Figure 1 Charge collected as a function of electric field for 5.5-MeV alpha particles in pure LAr and in LAr with a variety of dopants.

Figure 2 Charge collected as a function of electric field for 5.5-MeV alpha particles in pure LAr and LAr doped with TMG at different concentrations.

Figure 3 Charge collected as a function of electric field for 5.5-MeV alpha particles in purified LAr and unpurified LAr when doped with TMG and allene. The response to beta particles in pure LAr and LAr doped with TMG is also shown.

Figure 4 Energy resolution as a function of charge collected for 5.5-MeV alpha particles in pure LAr and LAr doped with allene.

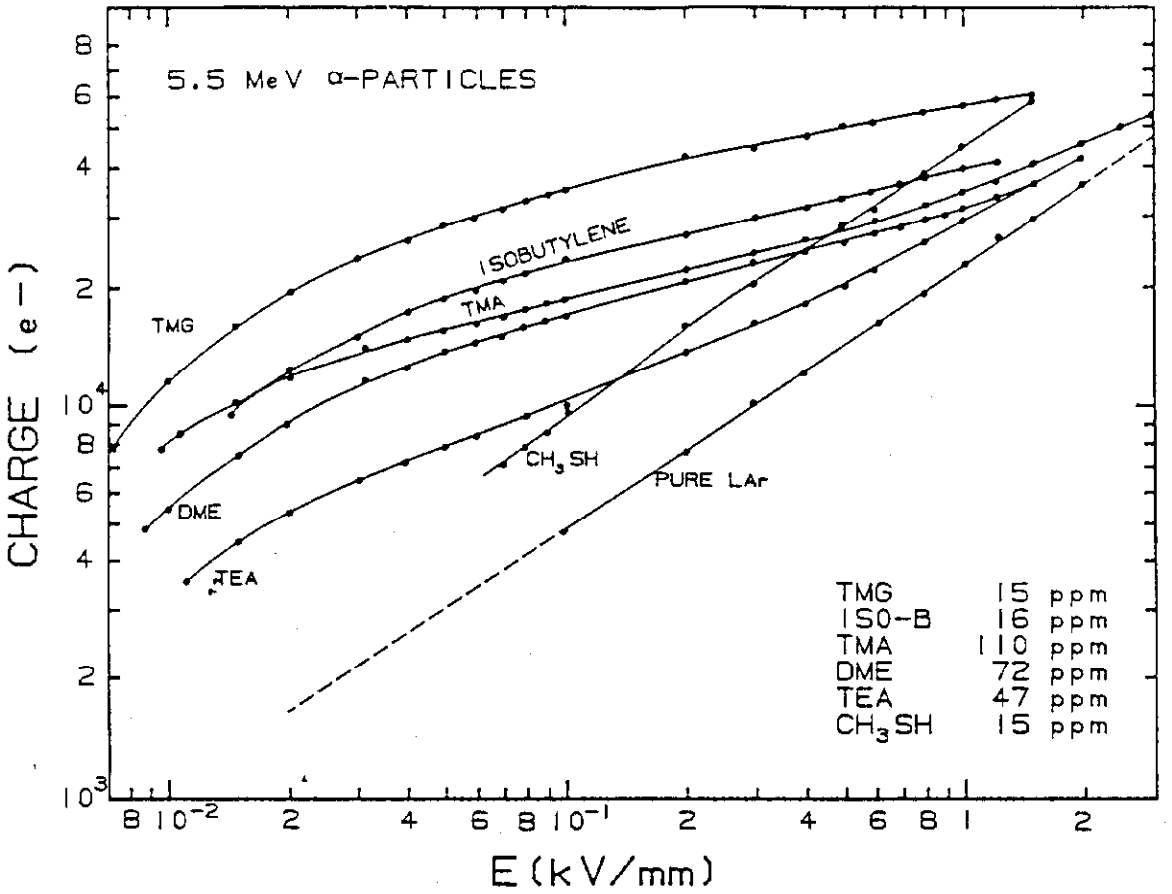

Figure 1

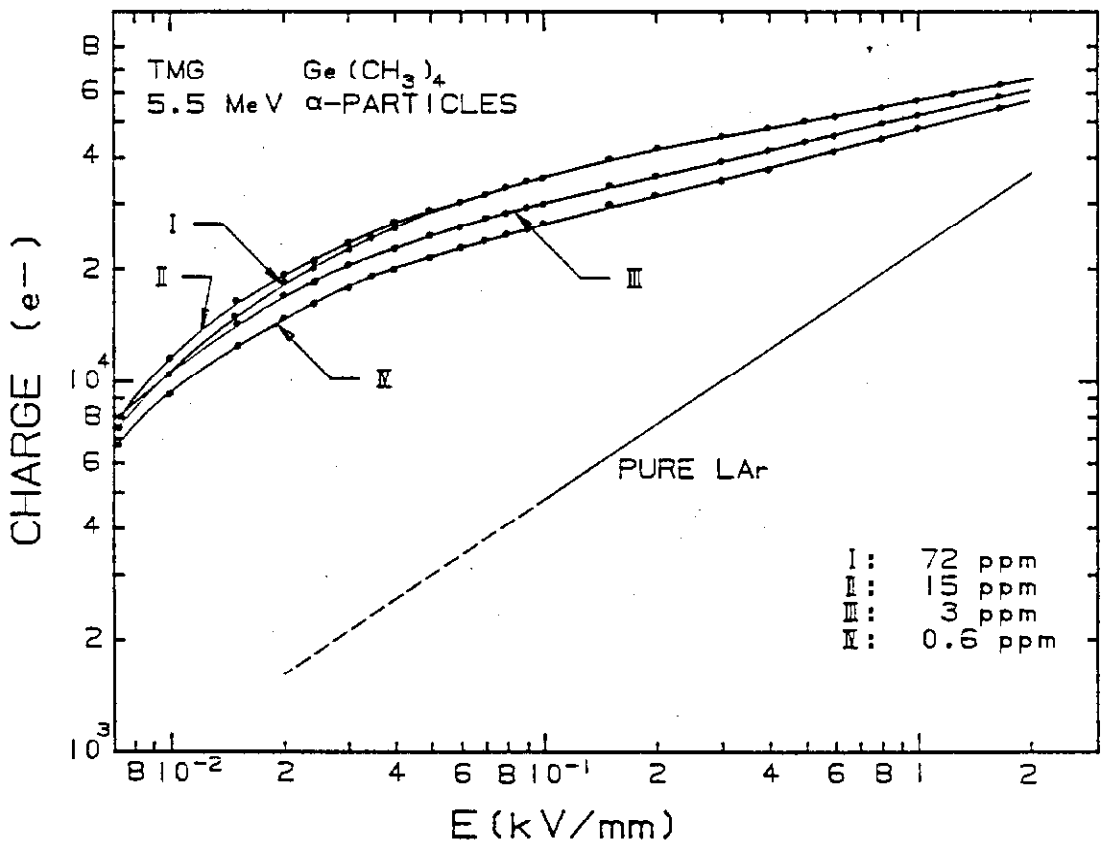

Figure 2

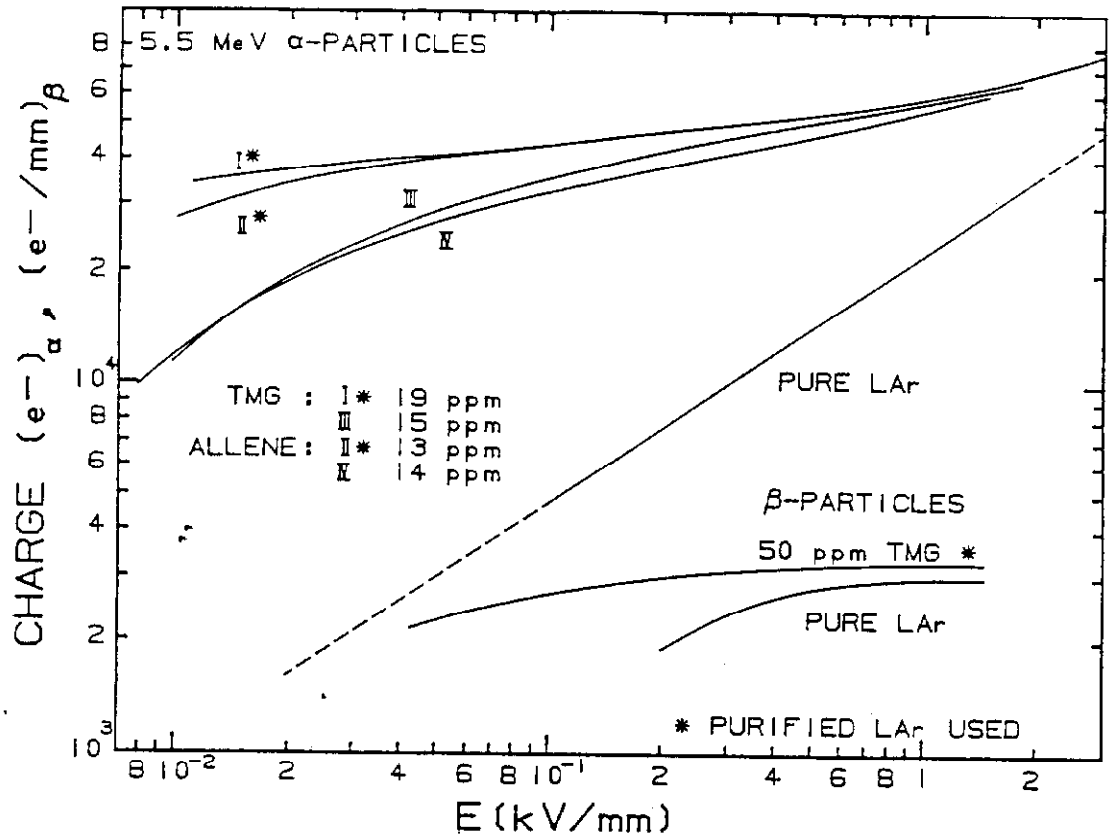

Figure 3

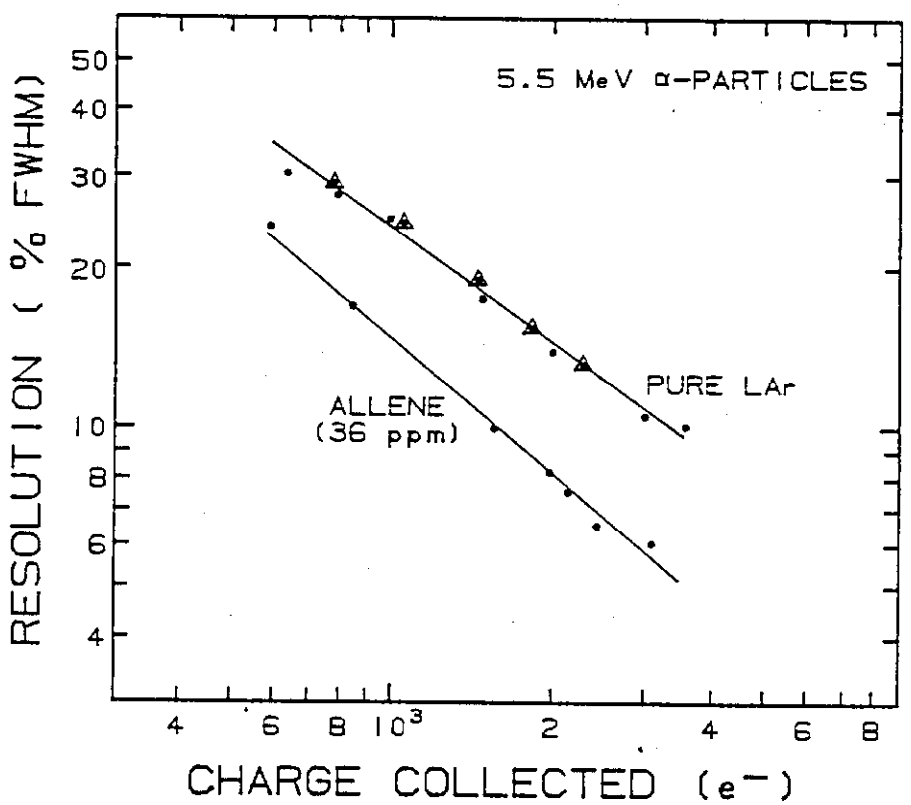

Figure 4