ENERGY RESOLUTION IN LIQUID ARGON DOPED WITH ALLENE

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1. INTRODUCTION

Liquid argon has been studied for detection medium with large volume and good energy and position resolutions(1). When ionizing radiation passes through liquid argon, a number of electron-ion pairs and excitons are produced along the track. The electrons escaping recombination with ions are collected at an electrode by an external applied electric field. The recombining electrons and the excitons emit scintillation photons. The charge and scintillation signals are compensative and the relative yields of those depend on the type of ionizing radiation and on applied electric field(2,3).

If the projectile ionizes liquid argon heavily, as is the case for alpha particles, scintillation yield is large because of the high probability of recombination. The scintillation from liquid argon is in vacuum ultraviolet region(9-10.5 eV), and its detection efficiency is low. Consequently, the energy resolution by scintillation measurement for alpha particles is not good (~ 10 % FWHM for 5.3 MeV). The energy resolution by charge measurement is also not good due to small fraction of unrecombined electrons.

It is advantageous to dope liquid argon with molecules which have an ionization potential that is lower than the energies of the scintillation light(4-11). This process will result in an increase in the number of electrons collected and a corresponding improvement in the energy resolution. In particular, allene (C_3H_4) is such a molecule whose ionization potential in liquid argon is estimated to be about 8.5 eV. And its photoionization quantum efficiency has been reported to be about 0.6(11), which is the highest value among the dopants investigated.

We have examined the energy resolution for 5.305 MeV alpha particles, and describe the present status of the experiment on the effect by allene doped in liquid argon. The preliminary result for 976 KeV electrons are also presented.

2. EXPERIMENTAL

The detector is a parallel plate ionization chamber. The diameter of the discal electrodes is 25 mm, and the gap between them is 5.3 mm. A Po-210 source of 5.305 MeV alpha particles was deposited in the center of the cathode (K).

The argon gas was purified for a few days by Ba-Ti getters(12). Allene was purified by two kinds of method: "(a) a small quantity purification" and "(b) mass purification". In case of (a), the weight of allene was about 4 g, while it was about 100 g in case of (b). Allene was degassed at -110 °C for about 10 ((b) 30) seconds and purified by the use of molecular sieves (3A) at about -80 °C for several houres, which had been activated at 300 °C under vacuum. And it was degassed again at -110 °C for about 5 ((b) 10) seconds just befour use.

The detector and the gas supply system were evacuated to 10^{-7} torr after being baked at 130 °C.

The argon gas was liquefied by immersing the detector envelope in liquid argon bath. Three kinds of method were tried for mixing allene with argon. In case of (A), allene was added after two thirds of the required volume of argon was liquefied in the detector envelope, when Po source was immersed in liquid argon, and then the rest volume of argon was liquefied. In case of (B), allene was added to argon after one third of the required volume of argon was liquefied. In case of (C), the argon and allene were mixed in gaseous phase. The mixture gas was then liquefied in the detector envelope.

3. RESULTS

3.1 Allene purification

Two kinds of purification methods for allene were tried which were mentioned in Sec. 2. Figure 1 shows the collected charge as a function of the applied electric field. The value of 23.6 eV was used for the W-value in liquid argon[13]. In case of "mass purification", the pulse hight was higher than that in case of "small quantity purification". Figure 2 shows the energy resolution as a function of the applied electric field. The resolution was improved in case of "mass purification". These two figures show that electronegative impurities were excluded more completely in case of "mass purification". The difference between the two methods was the duration of degas before and after purification. Degas process must be effective for the purification of allene in this system.

3.2 Change of allene concentration

Figure 3 shows the charge response of liquid argon doped with allene in various concentrations for the mixing method (C). Low concentrations of allene resulted in low yield, presumably because the absorption length for photons becomes significantly larger than the thickness of the detector for small concentrations of allene.

Figure 4 shows the resolution as measured both in the pure argon and allene mixture by the mixing method (C) as a function of applied electric field. It is interesting that the applied electric field above about 2 kV/cm degrades the resolution as measured in allene mixture 69 ppm. The mixtures with lower concentrations exhibit only a slight degradation in energy resolution with increasing electric field. The intrinsic resolution is also plotted in figure 5 which is obtained by quadratically substracting the electric noise contribution from the measured resolution. The best intrinsic resolution was 1.4 % FWHM at an applied field of 5.7 kV/cm in allene mixture of 4 ppm(figure 6).

3.3 The method of mixing allene and argon

Figure 7 shows the relation between the collected charge and the applied electric field for 5.305 MeV alpha particles for three different mixing methods. Figure 8 shows the resolution as a function of the applied electric field.

These two figures shows a clear difference between the methods. The data casts some doubt on our knowledge of the real concentration of allene. Figure 7 indicates larger concentrations for methods "A" and "B" than for "C". In this case, the resolution must be worse as predicted from figure 4 and this is inconsistent with the experimental results shown in figure 8. This fact might indicate the presence of electronegative impurities.

4. Discussion

4.1 Collected charge

An equation[7,8,10,11] was derived for the dependence of collected charge Q, including the effects of photoionization, on the applied field strength E. The result is

$$Q(E)=Q_a(E)+[\eta'(q_a N_i-Q_a(E))Y_b(E)+\eta' q_a N_{ex} Y_b(E)]\eta_p. \quad (1)$$

Q_a represents the portion of Q that is produced by direct ionization. N_i and N_{ex} represent the numbers of initially produced ion-electron pairs and excitons. The value of N_{ex}/N_i is 0.21[13] The probability that an isolated ion-electron pair produced through photoionization does not recombine is denoted by Y_b. The quenching factor for alpha particles is denoted by q_a which is 0.71-0.76 depending on the electric field[2,3]. η' is defined to be the product of ϕ and η_g, where ϕ is the quantum efficiency for photoionization and is the fraction of the number of photons absorbed in the detector volume to that emitted from an alpha particle track. The value of η_g was caluculated to be 0.57 for the mixture of 80 ppm allene and 0.40 for 8 ppm. η_p is the factor which expresses the positive ion effect on charge induced on the anode by ion-electron pairs produced through photoionization. The value of 0.6 is obtained for by fitting equation(1) to the experimental data for the mixture 82 ppm of allene as shown by a dashed line in figure 9[11].

4.2 Energy resolution

The factors which affect the energy resolution of alpha particles are estimated in this experiment[9]. (1)statistical fluctuation in the number of ion-electron pairs as expressed by the Fano factor(less than 0.2 % FWHM); (2)statistical fluctuations in the loss of electrons due to recombination(less than 0.5 % FWHM); (3)statistical fluctuations in the number of photoionization events(1 % FWHM); (4)fluctuation in the ionization density distribution along the track(negligible); (5)variation of recombination depending on the emission angle of alpha particles to the direction of the electric field(may be very significant); (6)variation in the value of due to the range of the alpha particles and variation of emission angle(may be very significant, especially high concentrations of allene); (7) variation of the local concentration of allene during the measurement(small); (8)electron attachment to electronegative impurities(unknown); (9)thickness of the radioactive source and irregularities of the surface of the electrodes (negligible); (10)electronic noise in the amplifier(about 450 e or 11 KeV).

In the best case, the measured energy resolution(1.7 % FWHM) is considered to be determined almost only by the statistics of the number of associated species and electronic noise. To improve the resolution further, the concentration of allene must be small so that the collisional energy transfer from excited argon atoms to allene molecules becomes negligibly small and the fluctuation in the value of η_{p} is reduced. And the size of the detector must be large so that the absorption length for photons is much smaller than the dimension of the detector for such dilute allene mixture.

5. RESULTS FOR ELECTRONS

The energy resolution has been examined for 976 KeV Bi conversion electron in liquid argon[14-18]. It is also expected that the energy resolution will be improved by adding allene to liquid argon if recombination process determines the energy resolution[19,20].

The gridded ionization chamber was used in this experiment. The diameter of the electrodes are 28 mm. A Bi-207 source was deposited in the center of the cathode. The grid is made of gold-plated tungsten wires of 10 μm in diameter strung on a tungsten alloy frame with a spacing of 150 μm. The distance between the cathode and the grid is 3.5 mm and that between the grid and the collector is 2 mm. Allene was purified by the "mass purification" method.

Figure 10 shows the charge collection as a function of electric field. Figure 11 shows the energy resolution as a function of electric field. These two figures indicate that the increase of collected charge dose not improve the energy resolution in case of electron radiation.

The best spectrum is shown in figure 12. 31 KeV FWHM was obtained for 976 KeV electrons in liquid argon doped with 65 ppm allene.

6. CONCLUSIONS

The addition of allene to liquid argon greatly improves the energy resolution of 5.305 MeV alpha particles. The best intrinsic resolution is 1.4 % FWHM obtained for 4 ppm allene doped liquid argon. In case of 976 KeV electron radiation, energy resolution was not improved by adding allene to liquid argon. The best resolution is 31 KeV FWHM obtained for 65 ppm allene doped liquid argon.

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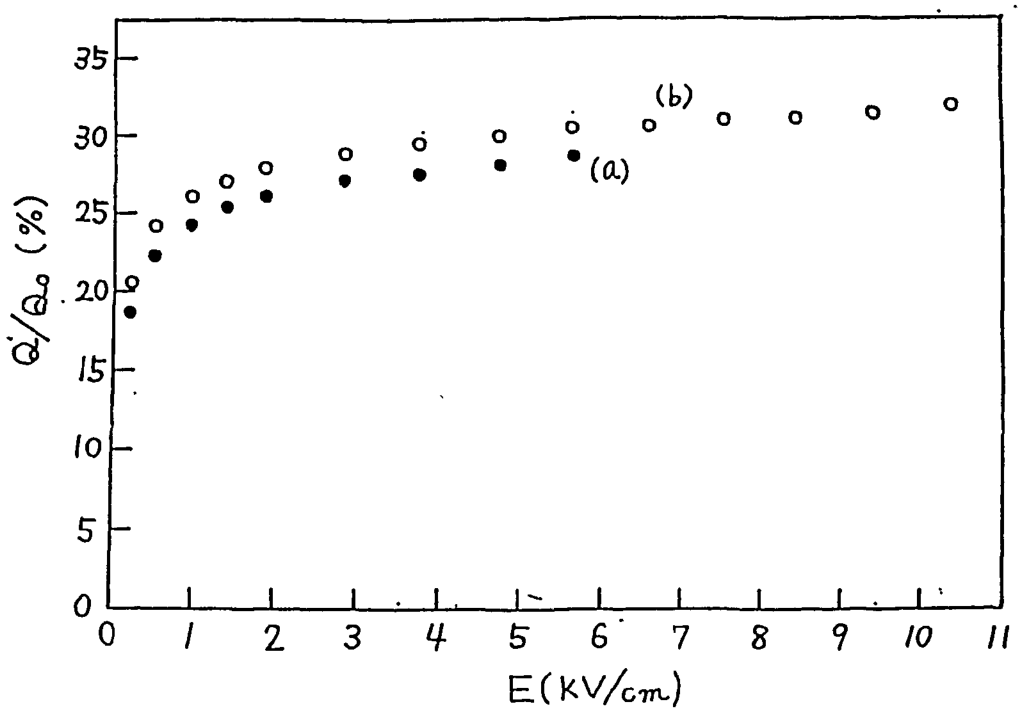

Fig. 1 Collected charge Q'/Q_0 vs applied electric field E for 5.305 MeV alpha particles in case of "(a)small quantity purification" and "(b)mass purification".

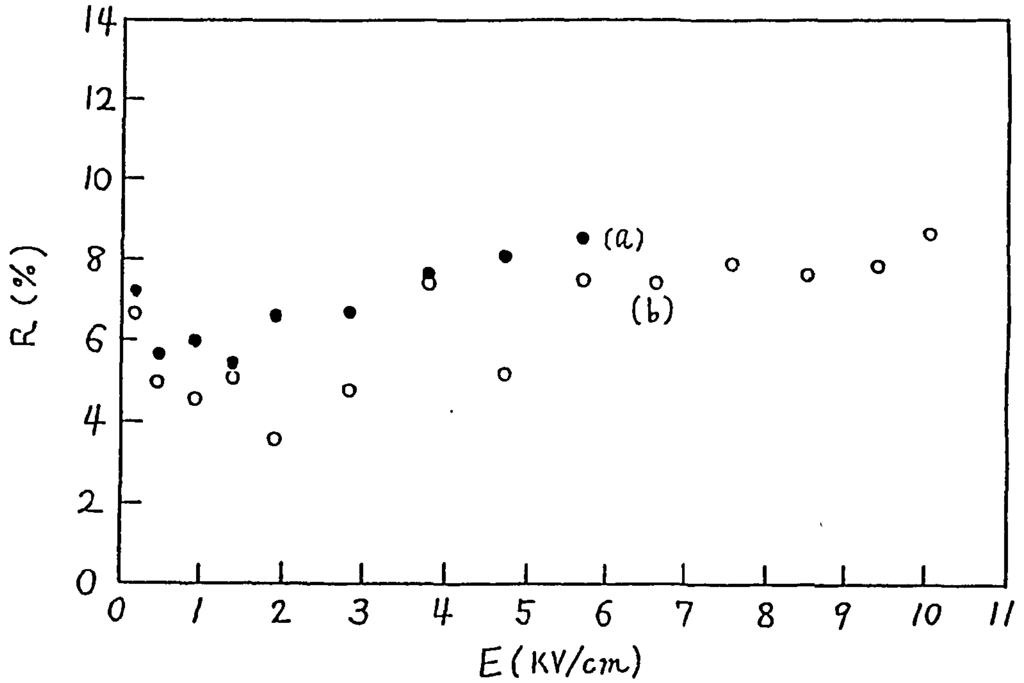

Fig. 2 Enegy resolution vs applied electric field E for 5.305 MeV alpha particles in case of "(a)small quantity purification" and "(b)mass purification"

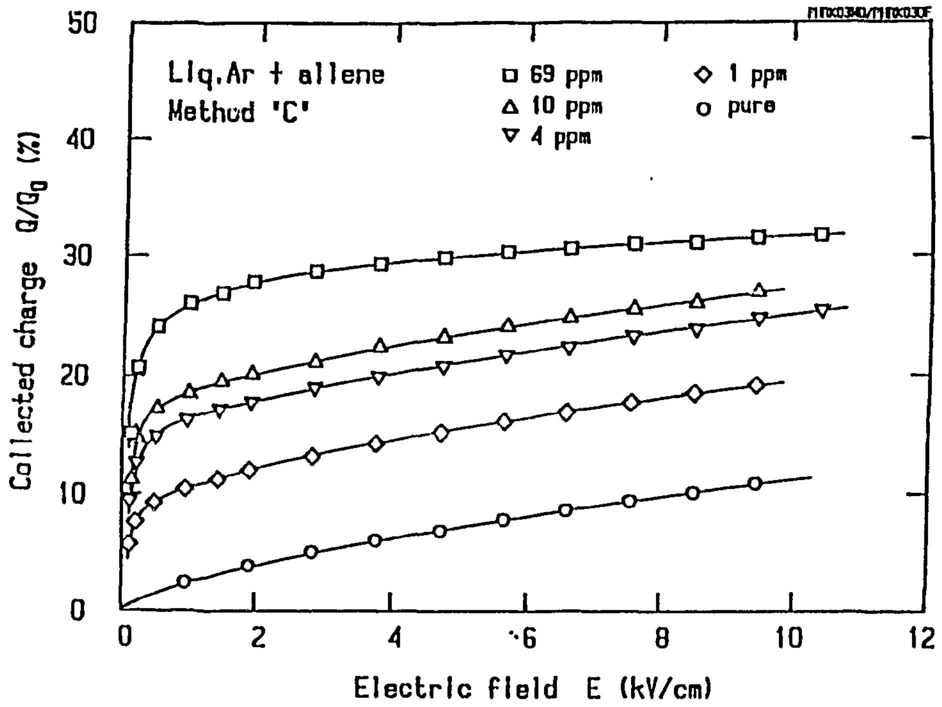

Fig. 3 Collected charge Q/Q_0 vs applied electric field E for 5.305 MeV alpha particles in allene doped argon and pure liquid argon.

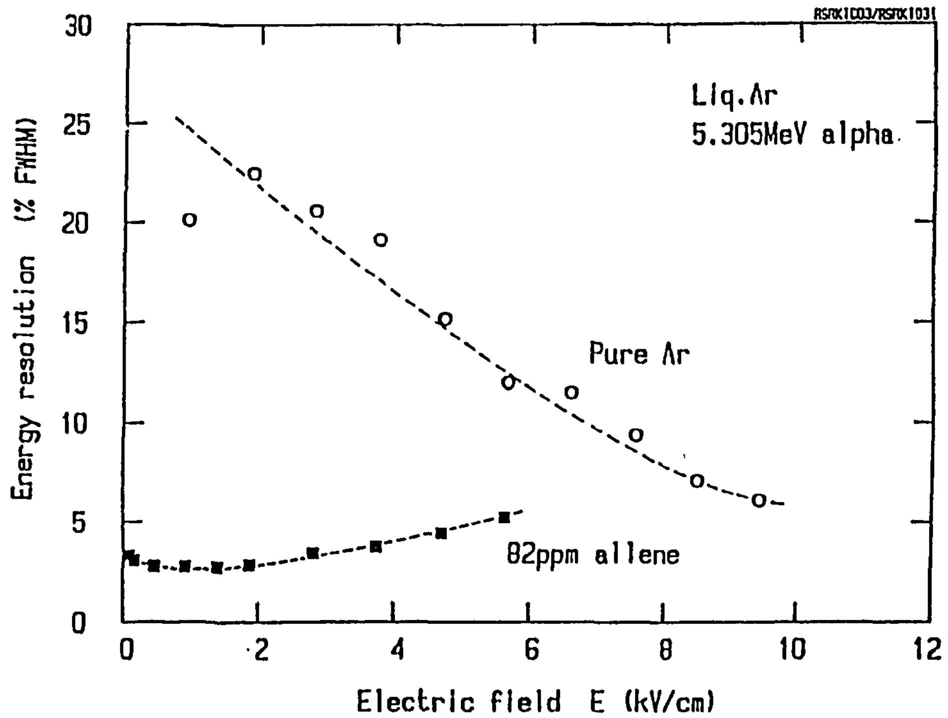

Fig. 4 Energy resolution vs applied electric field E for 5.305 MeV alpha particles in allene doped argon and pure liquid argon.

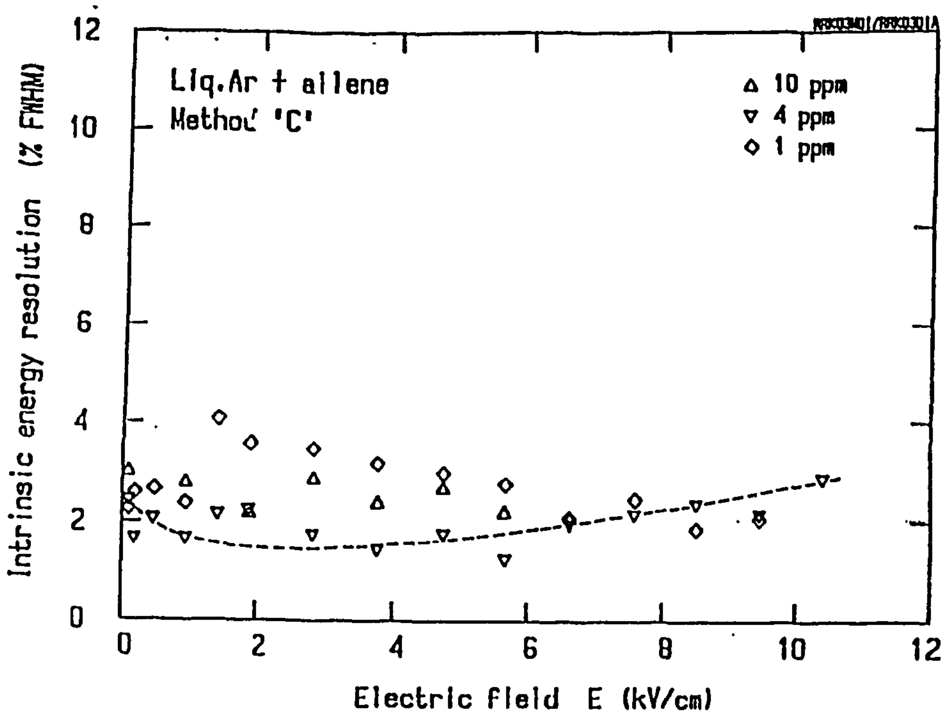

Fig. 5 Intrinsic energy resolution vs applied electric field E for 5.305 MeV alpha particles in allene doped argon and pure liquid argon.

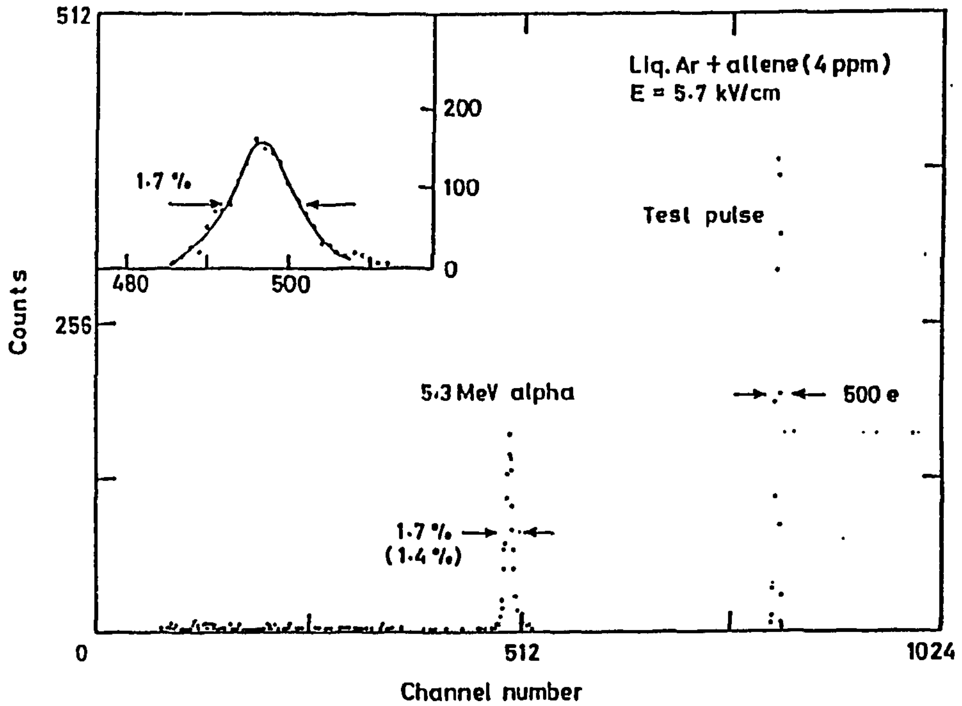

Fig. 6 The best spectrum for 5.305 MeV alpha particles taken with liquid argon+allene 4 ppm at 5.7 KV/cm.

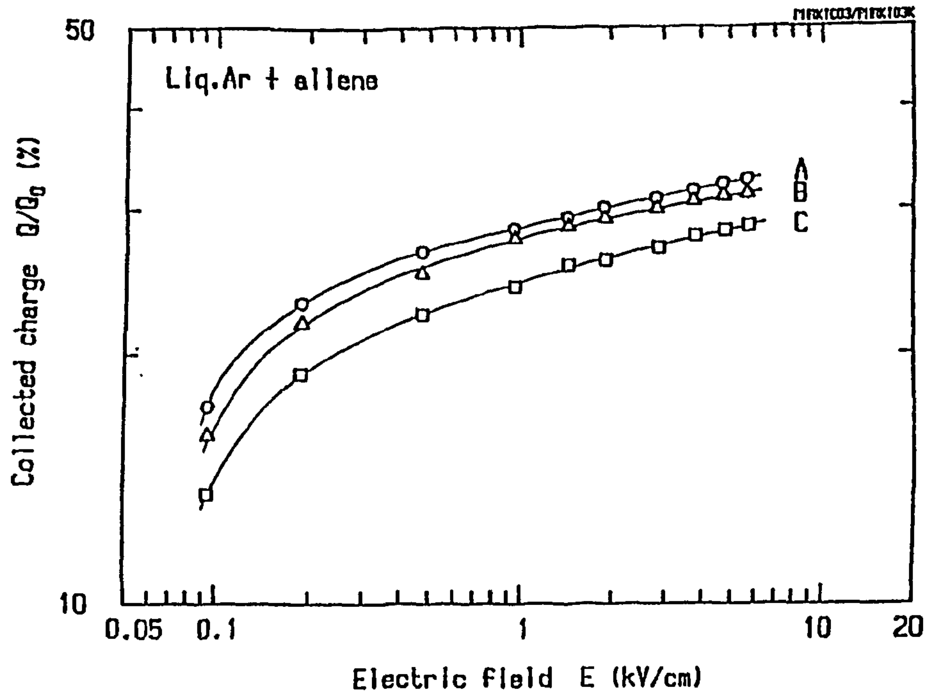

Fig. 7 Collected charge Q/Q_0 vs applied electric field E for 5.305 MeV alpha particles in allene doped argon for various mixing methods.

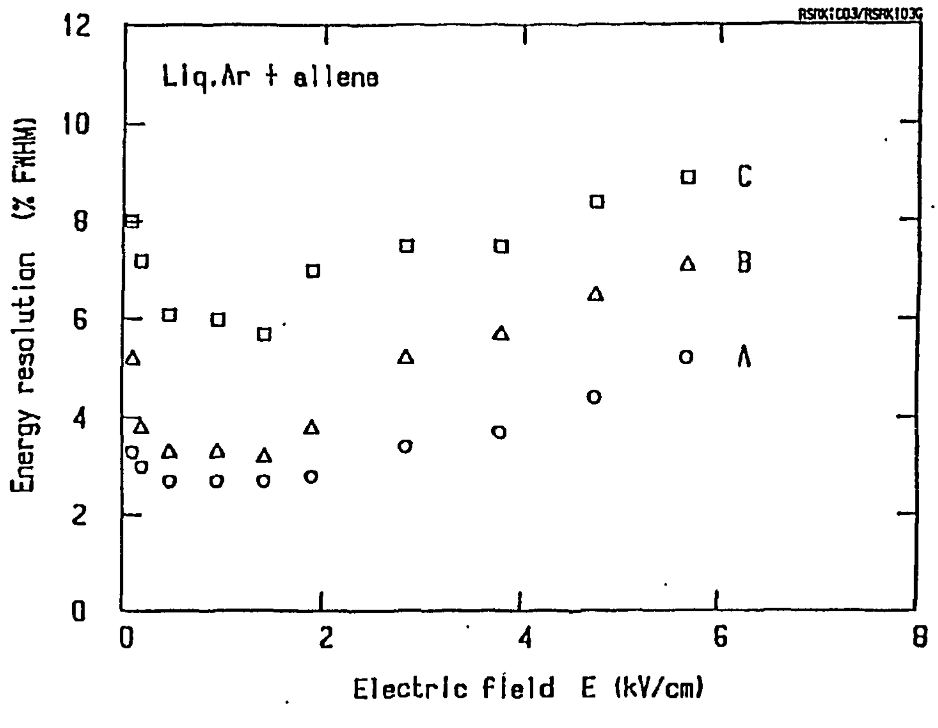

Fig. 8 Energy resolution vs applied electric field E for 5.305 MeV alpha particles in allene doped argon for various mixing methods.

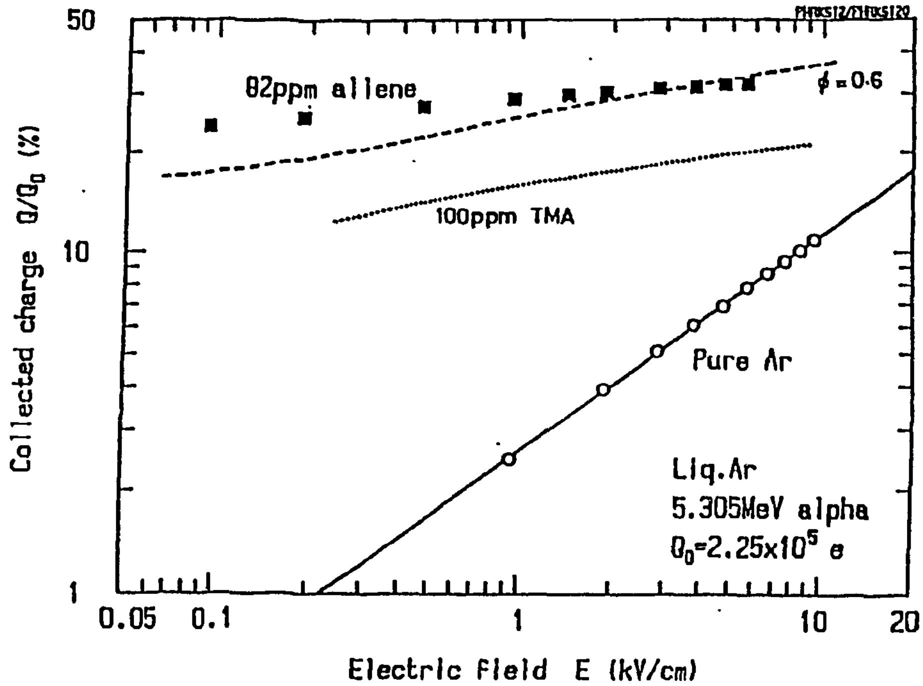

Fig. 9 Collected charge Q/Q_0 vs applied electric field E for 5.305 MeV alpha particles in 82 ppm allene doped argon and in pure argon.

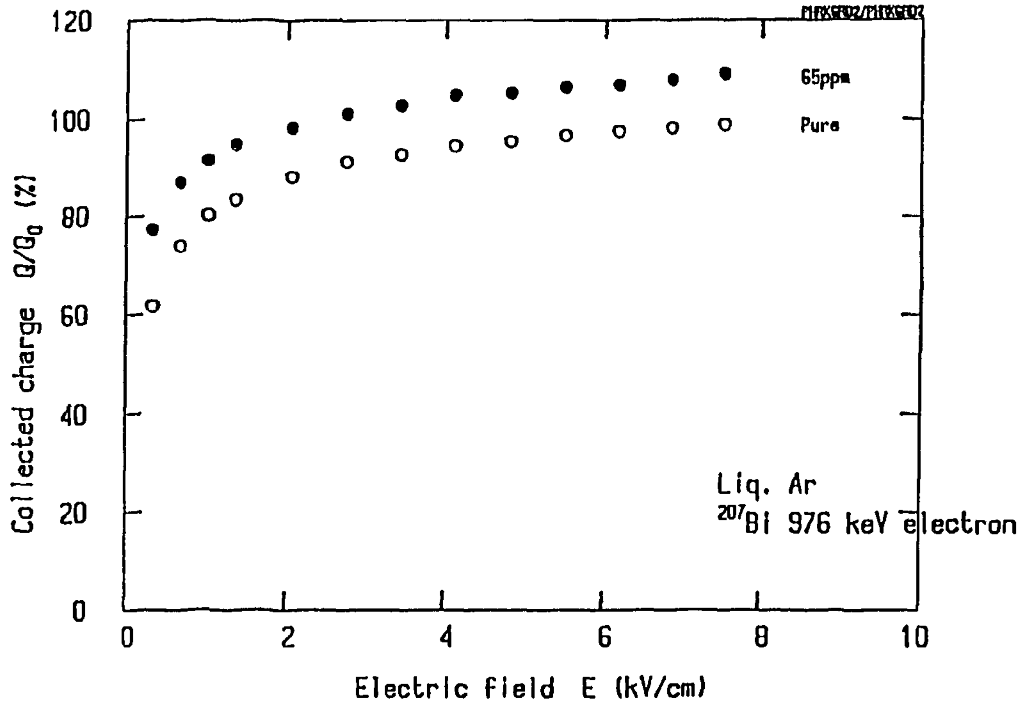

Fig. 10 Collected charge Q/Q_0 vs applied electric field E for 976 KeV electrons in 65 ppm allene doped argon and in pure argon.

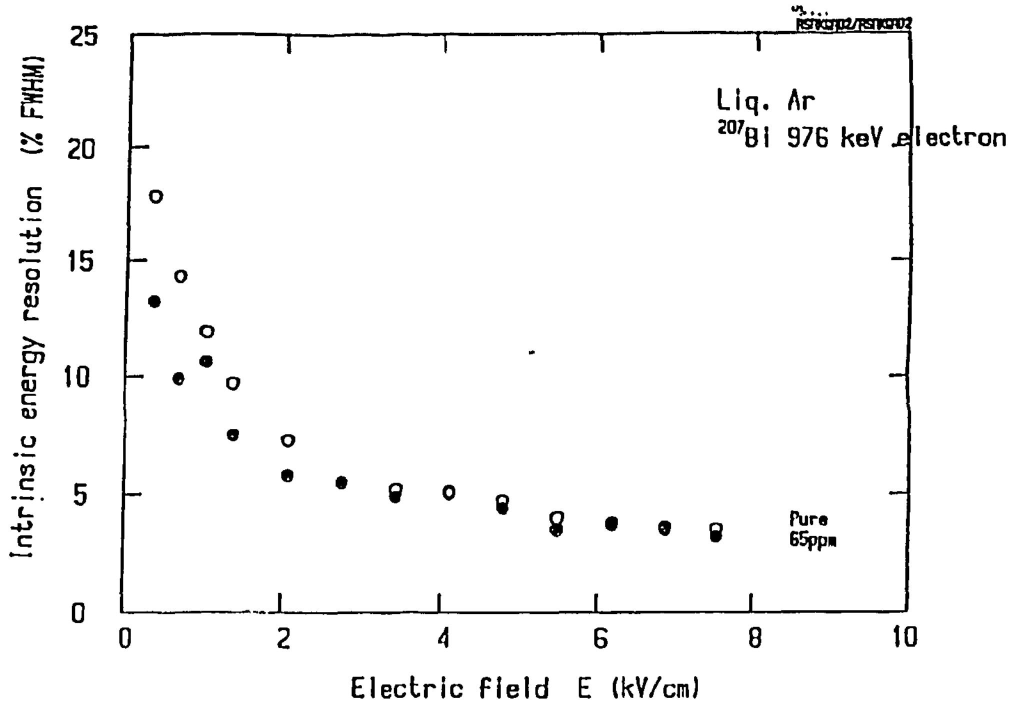

Fig. 11 Intrinsic energy resolution vs applied electric field E for 976 KeV electrons in 65 ppm allene doped argon and pure liquid argon.

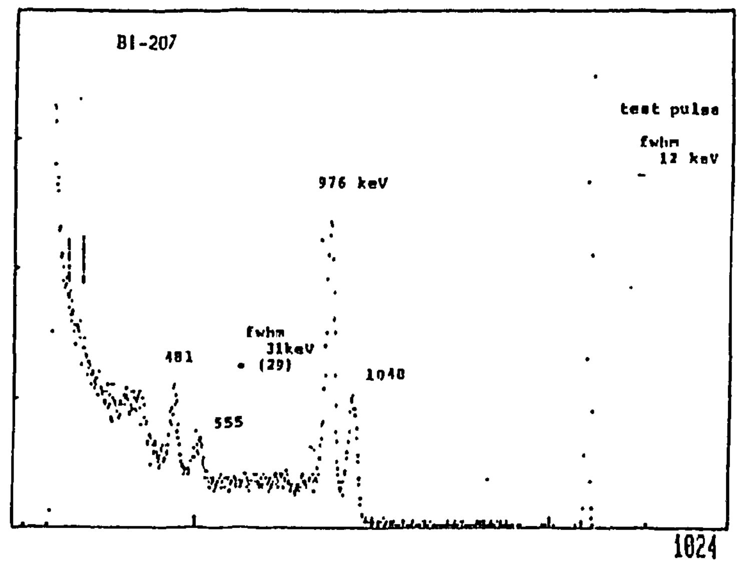

Fig. 12 The best spectrum for Bi-207 source, which was taken with liquid argon doped with 65 ppm allene at 7.5 kV/cm.