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Saturation Effects in Liquid Argon Doped with Methane*

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Abstract

Measurements of the charge collected for alpha and beta particles in liquid argon, doped with methane concentrations of up to 30%, have been measured. It is found that, for concentrations of ≥10% mole fraction of methane, the saturation constant in Birks' law does not change with increasing methane concentration, making the mixture similar to a warm liquid in behavior. The significance of the measurements for compensation in liquid argon hadron calorimetry is also discussed.

1. Introduction

In hadronic calorimetry, it is important to strive to design an instrument that has the same response for hadrons as for electromagnetic particles (i.e.. $e/h=1$). If $e/h \neq 1$, there is a constant term in the energy resolution that can dominate at high energies, and the pulse height will not scale as $1/\sqrt{E}$.

It has been very clearly shown by Wigmans in his landmark work[1], that in order to achieve compensation ($e/h=1$) in a hadron calorimeter there are certain requirements that must be met:

1) The calorimeter must be of the sampling type.
2) The sampling medium must contain a substantial amount of hydrogen.
3) The converter must be of a high-Z material such as lead or uranium.
4) The signal integration time must be long enough to be sensitive to the recoil protons.
5) The sampling fraction of the calorimeter must be tuned to account for the type of converter used and the response of the sampling material to recoil protons.

In a purely hadronic shower, on the order of 40% of the particle energy is invisible. Some of this energy escapes in the form of neutrinos and muons from the pion and kaon decays. Likewise, many of the neutrons produced escape, and the energy spent in breaking up the nuclei is also not seen.

The number of neutrons produced in a shower is proportional to the fraction of the shower that is purely hadronic. If there is hydrogen present in the calorimeter's readout, elastic scattering of the neutrons off the protons will be an important source of energy loss for the neutrons. If the only hydrogen present is in the readout, the energy going into recoil protons contributes directly to the signal, provided that the readout does not saturate too greatly from the high dE/dx of the recoil protons. Thus, a calorimeter that is more sensitive to the neutrons than to the direct ionization of the charged particles can be made compensating. Since only a small fraction of the missing energy is in the form of neutrons, a sampling calorimeter must be used to reduce the sensitivity to direct ionization. Also, since the number of neutrons produced in the shower is related to the fraction of nucleons in the converter that are neutrons, a high-Z material is necessary. There are about three times as many neutrons from a uranium converter than from lead. The increase in Z also lowers the response to electromagnetic interactions, through what is often erroneously called the "transition effect."

The above neutron energy loss mechanism requires several hundreds of nanoseconds. Thus, the signal integration time must be of sufficient length in order for the process to be effective. This time requirement puts the prospects of compensation at very-high-rate machines, such as the proposed Superconducting Super Collider (SSC), in question.

Hadron calorimeters that are constructed with the proper response to the direct ionization from the charged particles, and to recoil protons from neutron scattering, typically have energy resolutions of 30-35%/√E. Wigmans believes that for practical calorimeters, an energy resolution of 20%/√E is the ultimate limit. The key points are that a sampling calorimeter must be used with hydrogen in the readout. To date, the only calorimeters that have displayed compensation and good performance have used plastic scintillator as the readout material.

There are several major calorimeters using lead or uranium as the converter material and liquid argon (LAr) as the readout (e.g., SLD, D0, E-706, NA34). Since the response of LAr to neutrons is negligible, these are not compensating calorimeters. It is natural to search for a hydrocarbon that has a high solubility in LAr with the hopes of achieving compensation.

2. Doped Liquid Argon

In the past, LAr has been doped with three types of dopants for a variety of reasons. Small amounts ($\leq 0.5\%$) of methane and ethylene have been added to "cool" the excess electrons[2], thus increasing the electron mobility and decreasing diffusion. Improvements of greater than a factor of two in mobility have been achieved in this way.

Xenon has been used to dope LAr in order to increase the ion yield[3]. With its lower ionization potential, the xenon converts the excited states in the argon into free charge. This is similar to the Penning effect in gases[4]. With the addition of 1.6% xenon, the collected charge was increased by 13%.

A third type of additive used in LAr is the photosensitive dopants[5]. These convert the scintillation light into collected charge. Thirteen such dopants have been discovered for LAr[6]. Working with such dopants as tetramethylgermanium and allene, the charge collected for alpha particles was increased by factors of over 2.5 and 8 for electric fields of 1.0 kV/mm and 0.1 kV/mm, respectively. A substantial improvement in the alpha-particle resolution was also measured.

The addition of a dopant to LAr to increase its neutron sensitivity would constitute a fourth class of dopants. There are two criteria that such a dopant must meet in order to be successful. First, it must be sufficiently soluble to allow a substantial fraction of the neutron's energy to be deposited in the readout. Secondly, the saturation effects in the mixture must not be so large that only a small fraction of the deposited energy is recorded.

2.1. Solubility

The first requirement of solubility in LAr is somewhat difficult to address. One tool that has been useful for the selection of materials to try as photosensitive dopants has been Henry's law which states that for nonpolar solvents, the mole fraction of solute is proportional to the vapor pressure. Table I gives the estimated vapor pressures, at LAr temperature, for hydrogen and the simplest of the hydrocarbons.

Hydrogen has the highest vapor pressure. It has been shown that at a temperature of 87 K, the hydrogen pressure must be about 20 atm in order to have a mole fraction of 0.2 in the LAr[7]. At a temperature of 140 K this pressure is reduced to about 10 atm. This concentration is too low to be of interest for hadron compensation, and of course, these pressures are far too high for a calorimeter of any practical size.

Of the hydrocarbons listed in Table I, methane has the highest vapor pressure by at least four orders of magnitude. We have made mixtures with up to 50% methane by volume (44% mole fraction). Since methane has the highest solubility of the hydrocarbons in Table I, and is the simplest of the hydrocarbons, it is the only material that we have seriously considered for this type of doping.

2.2. Saturation

The recoil protons of interest have energies in the few MeV range. These protons have a dE/dx over 100 times larger than minimum ionizing particles (mip) in the LAr. Therefore, it is important that the readout does not lose too much of the signal from these protons to saturation. Thus, one needs to know the saturation effects of the readout as a function of dE/dx.

Unfortunately, to our knowledge, this information does not exist for pure LAr, let alone doped LAr. One solution that has been used before is to assume that the saturation in liquids is the same as for scintillators[1]. Then one uses Birks' law to describe the effects in scintillators:

$$\frac{dL}{dx} = \frac{dE/dx}{1 + kB\ dE/dx} \tag{1}$$

where L is the amount of light produced by a particle of energy E, normalized in such a way that L/E=1 for a mip. This form has its advantages in that measurements need only be measured at two values of dE/dx in order to determine the constant kB and the normalization. This can be easily done in the laboratory with alpha-particle and beta-particle sources. For our measurements we have rewritten eqn.1 as:

$$\frac{dQ}{dx} = \frac{C\ dE/dx}{1 + kB\ dE/dx} \tag{2}$$

where Q is the amount of charge collected and C is a constant that allows the absolute amount of charge collected to be determined from our results.

There are two weaknesses in the calculations made below. The first is that it has not been shown that pure or doped LAr obeys Birks' law. Since we are interested in recoil protons with energies of about 2-5 MeV, which do not have ionization densities too different from alpha particles, we feel that this assumption is justified. There will be different from alpha particles, we feel that this assumption is justified. There will be further support for this assumption discussed later in the paper.

The second weakness is that the dE/dx calculated for the alpha particles is taken to be the average dE/dx over the length of the track. This simplification does not introduce a large error to the results.

3. Experimental Setup

A collimated ^{106}Ru source was used to provide beta particles with a maximum energy of 3.5 MeV. The chamber used for the test with the beta particles had a gap of 2.15 mm 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 mip's. The electric field was kept constant in the second gap so that the trigger level did not change with change in collection voltage. An ^{241}Am source providing 5.5-MeV alpha particles, detected with a 1.39 mm ionization gap, was used.

The LAr was condensed from the gas from a dewar of welding-grade argon. Research-grade methane was used, purified by a Hydrox purifier. Tests have shown that the use of the purifier did not substantially change the data.

4. Test Results

As expected, the addition of methane decreased the amount of charge collected, with the greatest difference for the alpha particles. A typical set of data, for both alpha and beta particles, and for methane concentrations of 0, 5, 15, 25, and 35% mole fraction, is shown in Fig. 1. One must remember that since the beta particles deposit charge across the gap, when working in the pulse mode, the induced charge is only 1/2 of the charge actually liberated[8]. From the data in Fig.1 and other measurements, we were able to determine the constants C and kB in eqn. 2.

4.1. kB

Figure 2 shows the value of the constant kB as a function of methane concentration for electric fields of 1 kV/mm and 2 kV/mm. Because of the changing density of the mixture, dQ/dx and dE/dx have been divided by the density. Thus kB has the units of g $\mathrm{Mev^{-1}cm^{-2}}$.

The most striking thing about Fig. 2 is that the value of the constant kB saturates for methane concentrations of ≥10%. Thus, above this concentration, the amount of charge collected from alpha particles tracks the charge collected from beta particles. The values for the fits in this figure, for ≥10% methane, are 0.037 and 0.024 g $\mathrm{Mev^{-1}cm^{-2}}$ for 1 and 2 kV/mm, respectively.

It appears that there are two components to the charge liberated by heavily ionizing particles in liquids: a heavily ionizing fraction and a lightly ionizing fraction. With the addition of methane, the heavily ionizing component completely disappears, leaving only the lightly ionizing component. Thus, methane-doped LAr looks like a hydrocarbon, where the signal from alpha particles is believed to be mostly due to the delta rays that are produced[9]. It has also been shown[10] that the dE/dx dependence of warm liquids is fairly well fit by the theory of Jaffé. This fit is very similar in form to Birks' law and adds support to its use, at least for methane dopings of ≥10%.

Figure 3 shows kB as a function of electric field for LAr doped with 10% of methane and for pure LAr. Since the value of kB depends only on the electric field for methane concentrations ≥10%, one only needs to determine the value for one concentration. A plot of other concentrations show no significant differences from this figure.

A plot of kB as a function of electric field for LAr doped with 15 ppm of the photosensitive dopant allene is also shown in Fig.3. It can be seen that the addition of the dopant makes a dramatic reduction in the saturation properties of LAr, particularly at low electric fields. One would hope that the addition of such a dopant would lessen the saturation in methane-doped LAr. Unfortunately, methane has a high cross section for the absorption of the UV light produced in argon, and the addition of a few per cent to the allene doped LAr removed the effect of the allene completely.

4.2. C

The value of C as a function of methane concentration for electric fields of 1 kV/mm and 2 kV/mm is shown in Fig. 4. This figure reflects the decrease in collected charge with increase in methane concentration. As the methane concentration increases, the thermalization distance of the electrons decreases. Thus, a greater fraction of the electrons reach thermal energy in the high electric field of the positive ion and are lost to recombination. Like kB, C has its greatest change at concentrations of up to 10%. Above that amount, the slope is very gradual. The above data indicates that with a collection field of 2 kV/mm, the signal for a mip would be 54% and 47% of the value for pure LAr for methane mixtures of 10% and 20%, respectively.

5. Discussion

It has been shown for minimum ionizing particles that methane can be added to LAr to concentrations of up to 20% mole fraction while maintaining 50% of the original signal pulse height. This is important if one hopes to achieve compensation in LAr calorimeters. There are two major problems with this mixture. The first is the safety problem. Since methane has a lower vapor pressure than LAr, and in a mixture it has a vapor pressure equal to the vapor pressure of pure methane times its mole fraction (Raoult's rule), the mole fraction of methane in the mix will increase if the liquid is allowed to boil off. This could happen in the case of a spill. Thus the methane would concentrate and could cause a serious fire hazard.

The second problem with adding methane to LAr is saturation for particles with a high dE/dx. If one assumes that pure and methane-doped LAr obey Birks' law, and naively applies the value of kB for the pure LAr to doped LAr, the contribution to the signal of the recoil protons will be over-estimated by a factor of three. Thus, a substantially greater amount of methane must be added than was previously thought. The ideal additive would be a photosensitive dopant with a high solubility, but many likely materials have been tested[6] and none found.

The hadron response of a methane-doped LAr calorimeter remains to be calculated for lead and uranium converter. There are at present plans to dope the D0 test calorimeter at Fermilab. Such doping could substantially improve the performance of LAr calorimetry.

Table I: Some Vapor Pressures at 87 K

Material	**Formula**	**Estimated Pressure*** (torr)
Hydrogen	H_2	$1.2x10^5$
Methane	CH_4	$5.6x10^1$
Acetylene	C_2H_2	$2.0x10^{-3}$
Ethylene	C_2H_4	$2.4x10^{-2}$
Ethane	C_2H_6	$1.6x10^{-3}$
Propylene	C_3H_6	$9.5x10^{-6}$
Propane	C_3H_8	$5.4x10^{-6}$

* Estimated using the Clausius-Clapeyron equation and pressures taken from ref.11.

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Figure Captions

Figure 1	Charge liberated by 5.5 MeV alpha particles and the induced charge per mm for beta particles in methane doped LAr. Curves are shown for methane concentrations of 0, 5, 15, 25, and 30% mole fraction.
Figure 2	kB as a function of methane concentration for electric fields of 1 and 2 kV/mm.
Figure 3	kB as a function of electric field for pure LAr and, for LAr doped with 10% methane and 15 ppm allene.
Figure 4	C as a function of methane concentration for electric fields of 1 and 2 kV/mm.

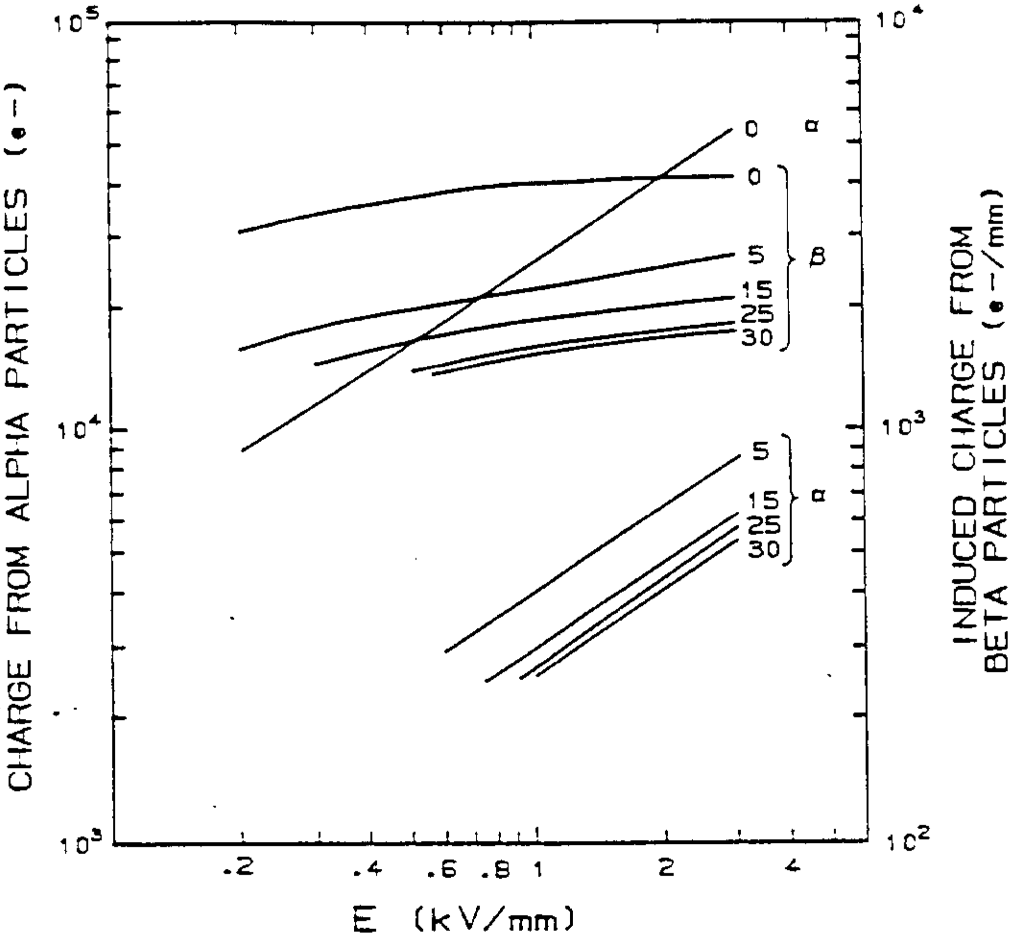

Figure 1

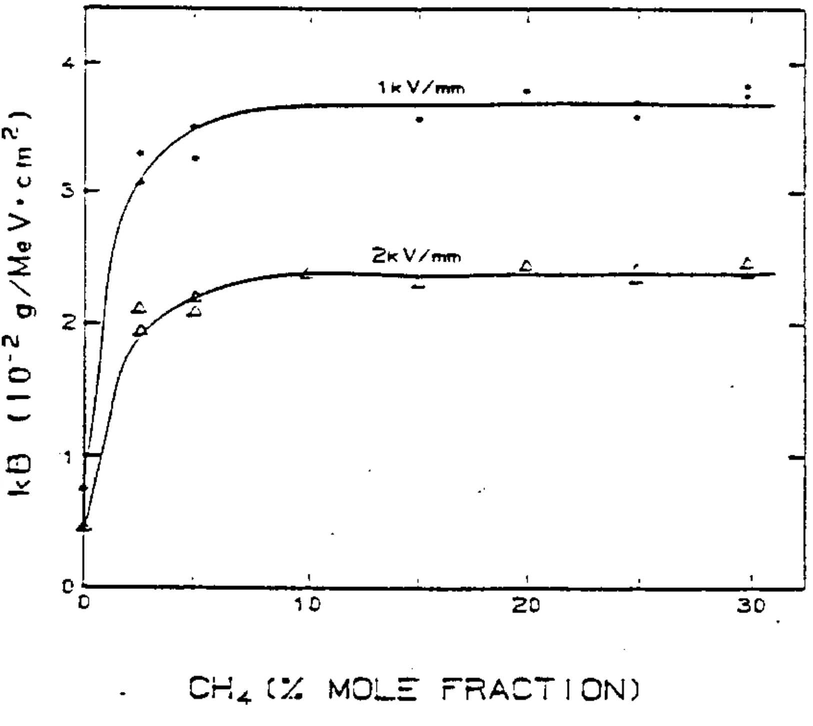

Figure 2

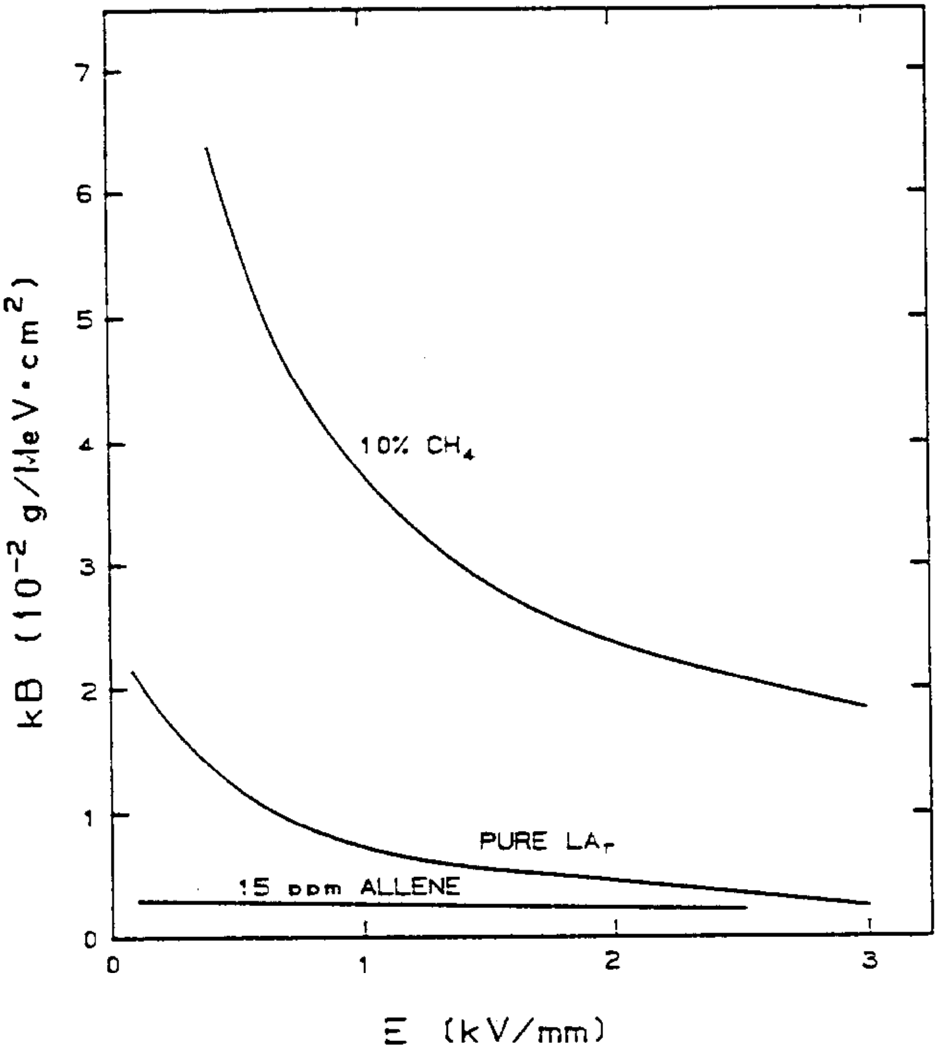

Figure 3

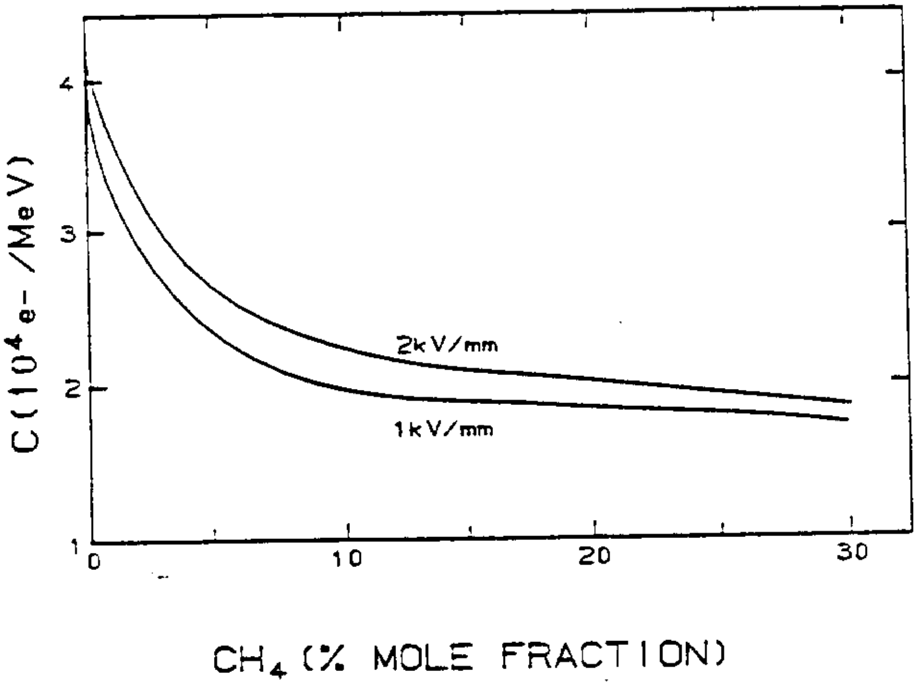

Figure 4