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Experimental ion mobility measurements in Xe-CF$_4$ mixtures

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ABSTRACT: In this paper we present the results of the ion mobility measurements made in gaseous mixtures of xenon with carbon tetrafluoride (Xe-CF$_4$) for pressures ranging from 6 to 10 Torr (8-10.6 mbar) and for low reduced electric fields in the 10 to 25 Td range (2.4-6.1 kV·cm^{-1}·bar^{-1}), at room temperature. The time-of-arrival spectra revealed one or two peaks depending on the gas relative abundances, which were attributed to CF$_3^+$ and to Xe$_2^+$ ions. However, for Xe concentrations above 60%, only one peak remains (Xe$_2^+$). The reduced mobilities obtained from the peak centroid of the time-of-arrival spectra are presented for Xe concentrations in the 5%-95% range.

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Contents

1 Introduction

Measuring the mobility of ions in gases is relevant in several areas, from physics to chemistry, e.g. in gaseous radiation detectors modelling and in the understanding of the pulse shape formation [1–3]. One of these examples is the upcoming LCTPC (Linear Collider TPC) for the International Linear Collider (ILC), where the estimate of the ion disk composition and evolution during the drift is essential to see where to stop the ions during the 1st millisecond of their drift (i.e. during a bunch train crossing) for the design of the gating device and dimensioning of the gap between the gating device and the readout the information on the ion mobility is essential.

Xenon (Xe) is commonly adopted as the main filling gas for several detectors, while the choice of the quencher is determined by different parameters [3]. Nowadays carbon tetrafluoride, CF_4, has been thought for the LCTPC mixture.

So, in the present work we studied the mobility of ions in xenon-carbon tetrafluoride (Xe-CF_4) gas mixtures in the 6 to 8 Torr (8 to 10.6 mbar) range and for reduced electric fields commonly used in gaseous detectors, extending previous studies developed in our group for other gases [4–18].

The experimental setup used in the present work (described in detail in [4]) allows the measurement of ion mobility in gas mixtures. Initially thought for high pressure, it was converted into a low pressure gas system, since lowering the operation pressure provided a wider scope of application and more detailed information on the fundamental processes involved in the ion transport and also allowed to reduce the inherent operation cost. Still, the results have been consistently in accordance with data obtained at high pressure [12].

1.1 Ion mobility

As a result of an interaction process in a gas mixture, a group of ions is originated and during their movement under the influence of low uniform electric field, these ions will collide with neutral gas molecules, losing energy in collisions while gaining energy from the electric field, eventually reaching a steady state. The resulting average velocity of the group of ions, v_d, also known as drift velocity can be given by:

$$v_d = KE \tag{1.1}$$

where K is the mobility of the ions and E is the intensity of the drift electric field. For low E/N, v_d is proportional to the electric field, i.e., when the energy gained from the field between collisions is below the thermal energy [19, 20]. K is usually expressed in terms of reduced mobility K_0, suppressing the dependence of the mobility values on the gas pressure. Thus

$$K_0 = KN/N_0 \tag{1.2}$$

where N is the gas number density and N_0 is the Loschmidt number ($N_0 = 2.68678 \times 10^{19}$ cm^{-3} for 273.15 K and 101.325 kPa according to NIST [21]). The mobility measurements are usually presented as a function of the reduced electric field E/N in units of Td (1 Td = 10^{-21} V·m^2) and the reduced mobility obtained from equation (1.2) is expressed in terms of cm^2·V^{-1}·s^{-1}.

1.2 Langevin's theory

According to the Langevin's theory [22], one limiting value of the ions' mobility is reached when the electrostatic hard-core repulsion becomes negligible compared to the neutral polarization effect. This limit is given by the following equation,

$$K_{pol} = 13.88 \left(\frac{1}{\alpha \mu} \right)^{\frac{1}{2}} \tag{1.3}$$

where α is the neutral polarisability in cubic angstroms (α = 4.044±0.01 Å^3 for Xe [23] and α = 3.86±0.01 Å^3 for CF$_4$ [24]) and μ is the ion-neutral reduced mass in unified atomic mass units.

Although the Langevin limit is meant to be applied for real ion-neutral systems in the double limit of low E/N and low temperature, it predicts the low-field mobility at room temperature with relatively good accuracy [20]. Although generally accepted, Langevin theory has some known limitations in its application, namely with ions that may interact by resonant charge transfer, in which case it fails to provide correct values for the ion's mobility [12].

1.3 Blanc's law

Blanc's empirical law, which resulted from Blanc's work in the mobility of ions in binary gaseous mixtures, has proven to be most useful when determining ions' mobility using mixtures of gases. Blanc found that the mobility of ions in gaseous mixtures, obeyed a simple relationship which can be expressed as follows:

$$\frac{1}{K_{mix}} = \frac{f_1}{K_{g1}} + \frac{f_2}{K_{g2}} \tag{1.4}$$

where K_{mix} is the reduced mobility of the ion in the binary mixture; K_{g1} and K_{g2} the reduced mobility of that same ion in an atmosphere of 100% of gas #1 and #2 respectively; f_1 and f_2 are the molar fraction of each gas in the binary mixture [25].

2 Method and experimental setup

The mobility measurements presented in this study were obtained using the experimental system described in [4]. A UV flash lamp with a frequency of 10 Hz emits photons that impinge on a 250 nm thick CsI film deposited on the top of a Gas Electron Multiplier (GEM) placed inside a gas vessel. The photoelectrons released from the CsI film are guided through the GEM holes by the electric field created by applying an adequate voltage across its electrodes. After gaining enough energy, the electrons will ionize the gas molecules encountered along their paths. While the electrons are collected at the bottom of the GEM electrode, the cations formed will drift across a uniform electric field region towards a double grid; the first one acts as Frisch grid while the second one, at ground voltage, collects the ions' charge. This charge collected at the collecting grid is converted from current to voltage by a pre-amplifier originating a time-of-arrival spectrum that is recorded in a digital oscilloscope (Tektronix TDS 2022B), set to continuously average 128 pulses, and fed to a PC for further processing. After subtracting the background spectra, obtained without the voltage applied to the GEM (i.e. without drifting ions), to these time-of-arrival spectra, Gaussian curves are fitted to the peaks in the spectra using Matlab. The trigger in the system is set by the UV flash lamp, providing the initial time information.

Since the peaks' centroid corresponds to the average drift time of the ions along a known distance (4.273 cm), the drift velocity is determined, and the mobility can then be calculated using expression (1.1). The system relies on the voltage across the GEM (V_{GEM}) to control the maximum energy of the electrons, which helps in the primary ion identification. Identifying the primary ions will allow to pinpoint secondary reaction paths that lead to the identification of the detected ions.

Since impurities play an important role in the ions' mobility, before each experiment the vessel was vacuum pumped down to pressures of 10^{-6} to 10^{-7} Torr and a strict gas filling procedure was carried out. No measurement was considered until the signal stabilised, and all measurements were done in a 2-3 minutes time interval to ensure minimal contamination of the gas mixture, mainly due to outgassing processes.

Despite the fact that no direct identification of the ions is possible with the present system, using an elaborate procedure based on the method described together with the knowledge of the dissociation channels, product distribution and rate constants it is possible to solve the ion identification problem.

3 Results and discussion

The mobility of the ions originated in Xe-CF_4 mixtures has been measured for different reduced electric fields E/N (from 10 to 25 Td), for a total pressure of 8 Torr and at room temperature (293 K).

The range of the reduced electric field values used to determine the ions' mobility is limited by two factors: the occurrence of electric discharges at high E/N and the deterioration of the time-of-arrival spectra for very low E/N values (below 5 Td or 1.2 kV·cm^{-1}·bar^{-1}), which has been attributed to collisions between the ions and impurity molecules.

The range of E/N values considered in this work is within the conditions of low reduced field, i.e. $E/N < 30$ Td for the working pressures used.

3.1 Xenon (Xe)

For pure Xe, with the experimental setup and conditions used of reduced electric fields E/N (10–25 Td) and for a pressure of 8 Torr, at room temperature (293 K) and for a constant V_{GEM} of 25 V, two types of ions are produced: Xe^+ and Xe_2^+. The atomic ion Xe^+ is produced directly by electron impact. The homonuclear diatomic ion Xe_2^+ is produced either by Hornbeck-Molnar process (a two-body association involving an excited atom and another in the ground-state), the dominant formation process below about 1 Torr,

$$Xe + Xe^{**} \rightarrow Xe_2^+ + e^- \tag{3.1}$$

or by conversion of the primary ions Xe^+ through a three body process, which is favoured at high pressure,

$$Xe^+ + 2Xe \rightarrow Xe_2^+ + Xe. \tag{3.2}$$

The corresponding K_0 values, compatible with those obtained in [4].

3.2 Carbon tetrafluoride (CF$_4$)

In pure carbon tetrafluoride (CF$_4$), only one peak was observed for different reduced electric fields E/N (10–25 Td) and for a pressure of 8 Torr, at room temperature (293 K) and for a constant V_{GEM} of 25 V.

In table 1 the possible reactions resulting from the electron impact in CF$_4$ for electron energies up to 25 eV, together with their (respective) cross-sections, appearance energies and the product distribution are summarized. The probabilities indicated for the product distribution were calculated using the partial and total electron impact cross sections for CF$_4$ provided in [26], which allowed to infer the product distribution of the primary ionization. To the best of our knowledge these primary ions (CF_3^+, CF_2^+, CF^+, F^+ and C^+) are long-lived at our working pressures, which means that once formed they will remain unaltered during their drift time.

Table 1. Ionization products, ionization cross sections for electron impact 25 eV on CF$_4$ [26], appearance energies (A.E.) [27], and respective product distribution.

Reaction	Cross Sec. (10^{-16}cm^2)	A. E. (eV)	Prod. Dist.
$e^- + CF_4 \rightarrow CF_3^+ + F + 2e^-$	1.011	15.69	96.1 %
$e^- + CF_4 \rightarrow CF_2^+ + 2F + e^-$	0.038	21.47	3.6 %
$e^- + CF_4 \rightarrow CF^+ + F + e^-$	0.0017	29.14	0.16 %
$e^- + CF_4 \rightarrow C^+ + 2F_2 + e^-$	0.0007	34.77	0.007 %
$e^- + CF_4 \rightarrow F^+ + CF_3 + e^-$	0.0007	35	0.007 %

As seen in table 1, the most probable ion is CF_3^+. In addition CF_2^+, CF^+, C^+ and F^+ are also produced but with a much lower probability, up to 3.9 % of the total number of primary ions. Considering the relative abundance expected and our experimental system's limitations (e.g. signal-to-noise ratio), it is highly probable that it will only be possible to observe the peak corresponding

to CF_3^+ ion, eventually with the residual (up to 3.6%) contribution of CF_2^+, which would be expected to have a slightly higher mobility due to its smaller mass.

Comparing the experimental mobility values with the polarization limit from Langevin's formula (equation (1.3)) it was possible to verify that the measured mobility tends to 1.12 cm^2 V^{-1} s^{-1}, a value in good agreement with the calculated value from equation (1.3) for CF_3^+, 1.13 cm^2 V^{-1} s^{-1}, although it deviates from those obtained by other authors [28, 29]. The difference can be explained by the possibility of the cluster formation [30], $CF_3^+(CF_4)$, that would be favoured at higher pressures (close to atmospheric) and whose expected value is close to those observed experimentally by these authors.

3.3 Xenon-carbon tetrafluoride (Xe-CF$_4$) mixture

In xenon-carbon tetrafluoride (Xe-CF$_4$) mixtures, one or two peaks can be observed depending on the mixture composition, from pure Xe to pure CF$_4$, as can be seen in figure 1, where the drift spectra for several Xe-CF$_4$ mixtures (20%, 50%, 80% and 90% of Xe) are displayed, at 8 Torr, 293 K and 15 Td with a V_{GEM} of 25 V.
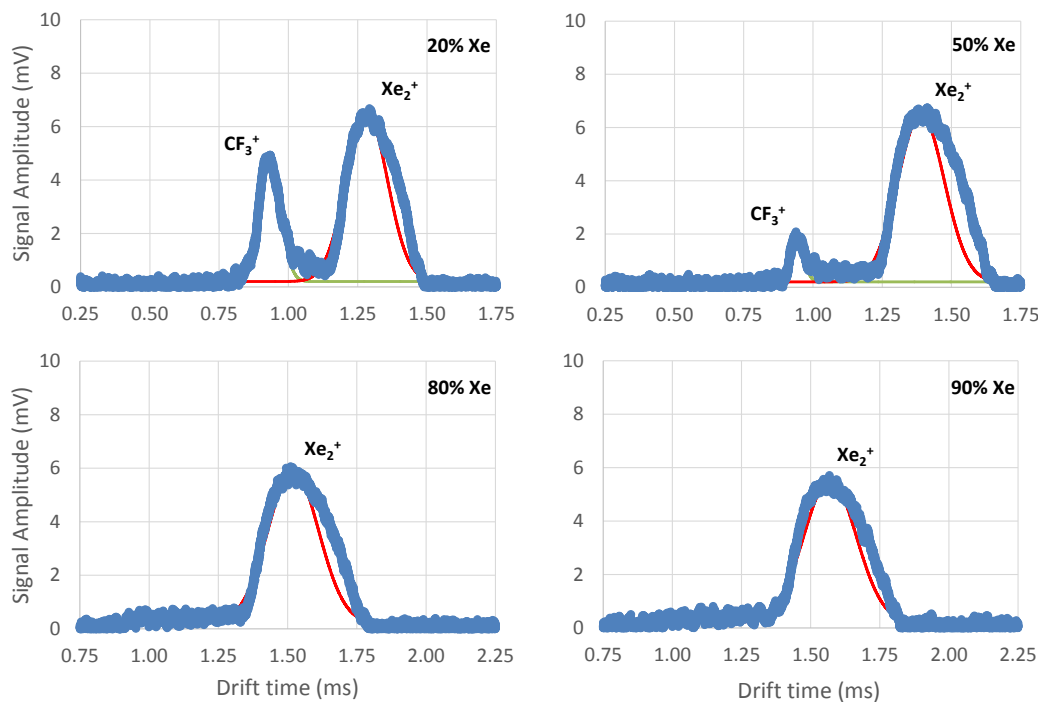

Figure 1. Time-of-arrival spectra of an average of 128 pulses recorded for several Xe-CF$_4$ mixtures (20%, 50%, 80% and 90% of Xe) at a total pressure of 8 Torr, a reduced electric field of 15 Td, a V_{GEM} of 25 V and at room temperature (293 K).

In figure 1 there are two stricking features both of which are a result of the increasing Xe concentration in the mixture: the decrease in mobility of the different ions observed — more pronounced in the lower mobility peak — and the change in the relative abundace of ion species present (even changing the dominant ion species), which can be perceived by a decrease in the area of the peak corresponding to the higher mobility, and an increase in the area of the other

peak. Considering the shift of the peaks towards higher drift times (decreasing ion mobility) with increasing Xe concentration, it can be explained by the higher Xe mass when compared to that of the CF_4 molecule, which implies a higher reduced mass in ion-neutral collisions (μ in equation (1.3)), thus a lower mobility. Regarding the changes observed in the different peaks' area, it suggests that the faster group of ions comes from CF_4, while the second, and slower group, is originated by Xe atoms.

The evolution of the proportion of the peaks observed (from CF_4 and Xe ions) with the mixture composition was compared with the relative abundance of primary ions after traversing the GEM holes, using a Monte Carlo simulation code described in detail in [31]. The simulation code uses the cross-sections for the scattering of electrons by Xe atoms and CF_4 molecules described in [32]. As in [31], a uniform field between anode and cathode inside the GEM holes was considered. Although this is not the real field geometry, it can provide sufficient information for the present purpose. The simulation reproduces the drift of 10^8 photoelectrons released from a CsI photocathode into Xe-CF_4 mixtures (at a typical pressure of 8 Torr and temperature of 293 K) until they reach the anode at a distance of 50 mm from the photocathode and calculates the proportion of primary ions (CF_3^+ or Xe^+) after this drift for the different gas mixtures. The results are displayed in figure 2, where it can be seen that, even at low Xe concentrations (down to about 15% of Xe), Xe ions are still the ones preferentially produced.
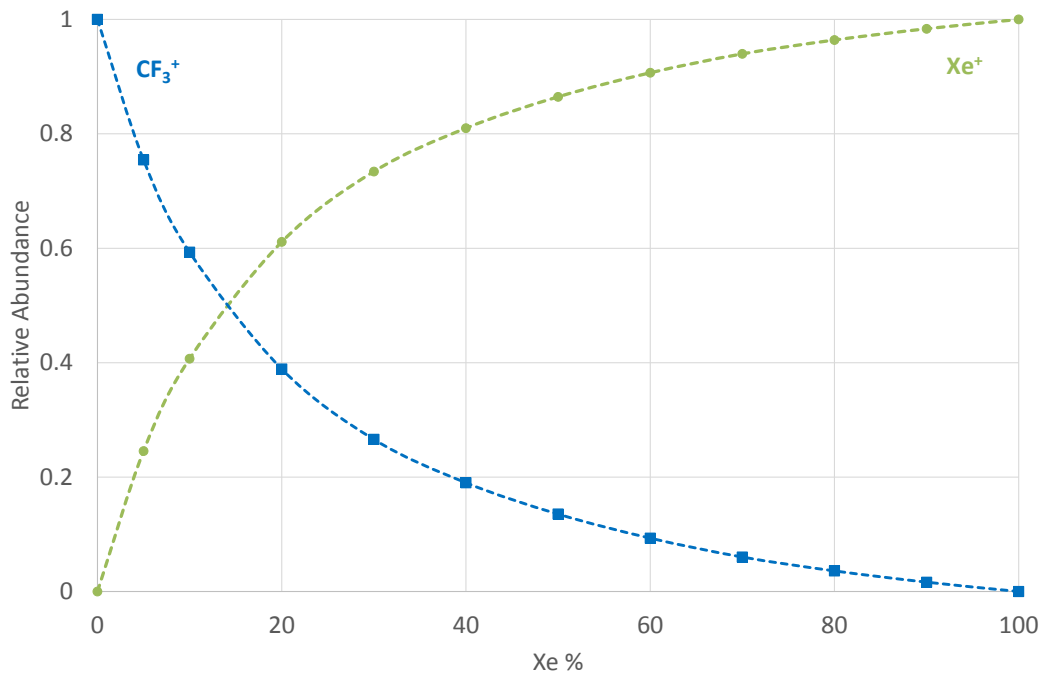

Figure 2. Monte Carlo calculated relative abundance of the fraction of ions produced at the GEM holes as a function of the Xe percentage in the mixture.

Comparing the results from figure 2 for the relative abundance of the ions in Xe-CF_4 calculated through the Monte Carlo simulation with the peak area, obtained by adjusting a Gaussian fit to each peak in the experimental time spectra (figure 1), it is possible to see a good agreement between the two, indicating that once the primary ions are formed the relative abundance of CF_4 and Xe ions will remain approximately constant.

Blanc's law was also used as a cross checking method to verify the ion identification.

Figure 3 shows the reduced mobility of the ions produced in different Xe-CF$_4$ mixtures at 8 Torr and for E/N of 15 Td at room temperature, together with Blanc's law prediction for the main candidate ions — CF$_3^+$ (blue), Xe$^+$ (green) and Xe$_2^+$ (red). K_{g1} and K_{g2} in Blanc's law (equation (1.4)), were obtained either using experimental values from literature or, when not existing, by using the Langevin limit formula (equation (1.3)).
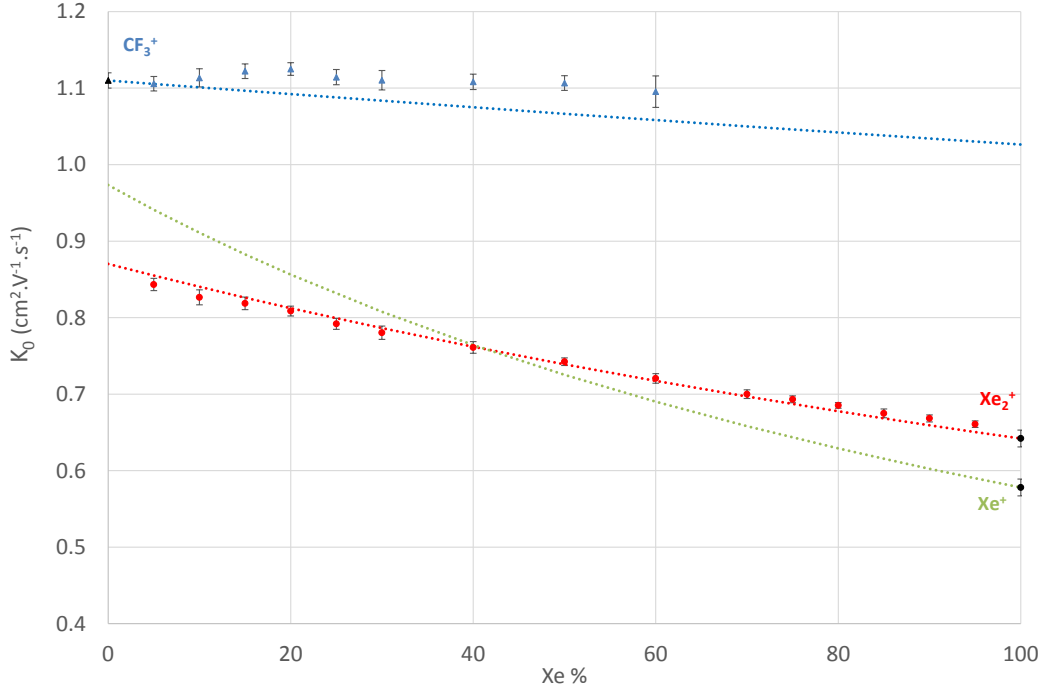

Figure 3. Reduced mobility of the ions produced in the Xe-CF$_4$ mixture for a pressure of 8 Torr and for a E/N of 15 Td at room temperature. The dotted lines represent the mobility values expected from Blanc's law for CF$_3^+$ (blue), Xe$^+$ (green) and Xe$_2^+$ (red).

Figure 3 shows that while the peaks corresponding to higher drift times follow closely Blanc's law curve for Xe$_2^+$, those corresponding to lower drift times are also well decribed (3% maximum deviation) by the same law for CF$_3^+$. The slight deviation observed may be caused by the collision-induced dissociation reaction between CF$_3^+$ and Xe [33],

$$CF_3^+ + Xe \rightarrow CF_2^+ + Xe + F \tag{3.3}$$

which is energetically favourable and would lead to an increasing formation of CF$_2^+$ with increasing concentration of Xe in the mixture and for which there is no reaction rate available.

In table 2 the results obtained for the ion mobilities of CF$_3^+$ and Xe$_2^+$ in Xe-CF$_4$ mixtures are summarized.

No significant variation of the mobility was observed in the range of E/N values (10-25 Td) studied.

Table 2. Mobility of the peaks observed for the Xe-CF$_4$ mixture ratios studied, obtained for E/N of 15 Td, a pressure of 8 Torr at room temperature (293 K).

Mixture	Mobility (cm$^2\cdot$V$^{-1}\cdot$s^{-1})	Ion
5% Xe	1.106 $\pm$ 0.009	CF$_3^+$
	0.843 $\pm$ 0.008	Xe$_2^+$
10% Xe	1.113 $\pm$ 0.012	CF$_3^+$
	0.827 $\pm$ 0.011	Xe$_2^+$
15% Xe	1.122 $\pm$ 0.009	CF$_3^+$
	0.819 $\pm$ 0.008	Xe$_2^+$
20% Xe	1.125 $\pm$ 0.008	CF$_3^+$
	0.809 $\pm$ 0.006	Xe$_2^+$
25% Xe	1.114 $\pm$ 0.009	CF$_3^+$
	0.792 $\pm$ 0.007	Xe$_2^+$
30% Xe	1.100 $\pm$ 0.013	CF$_3^+$
	0.780 $\pm$ 0.009	Xe$_2^+$
40% Xe	1.108 $\pm$ 0.001	CF$_3^+$
	0.761 $\pm$ 0.008	Xe$_2^+$
50% Xe	1.107 $\pm$ 0.009	CF$_3^+$
	0.742 $\pm$ 0.005	Xe$_2^+$
60% Xe	1.095 $\pm$ 0.021	CF$_3^+$
	0.721 $\pm$ 0.006	Xe$_2^+$
70% Xe	0.700 $\pm$ 0.006	Xe$_2^+$
75% Xe	0.693 $\pm$ 0.005	Xe$_2^+$
80% Xe	0.685 $\pm$ 0.004	Xe$_2^+$
85% Xe	0.675 $\pm$ 0.006	Xe$_2^+$
90% Xe	0.668 $\pm$ 0.005	Xe$_2^+$
95% Xe	0.661 $\pm$ 0.004	Xe$_2^+$

4 Conclusion

In the present work we measured the reduced mobility of ions originated by electron impact in Xe-CF$_4$ mixtures under different pressures (from 6 to 10 Torr), low reduced electric fields (from 10 to 25 Td) and different mixture ratios, for electron impact ionization energies up to 25 eV.

Only one peak was consistently observed for Xe concentrations above approximately 60%, thought to be originated by Xe$_2^+$, while a second peak starts to appear for Xe concentrations below this value and which was attributed to CF$_3^+$, with the expected abundance of both ions being in accordance with a Monte Carlo simulation carried out for the whole range studied.

Also, our experimental results seem to be relatively consistent with the ones predicted by the Blanc law throughout the mixture range studied (from 0 to 100% Xe) for the peak with lower mobility, Xe_2^+, while displaying a slight deviation for CF_3^+.

Increasing CF_4 concentration in the mixture resulted in a higher mobility of both ions observed. The experimental mobility values did not display a significant dependence over the studied range of pressure and E/N (6-10 Torr and 10-25 Td, respectively).

Future work is expected with other gaseous mixtures. It is our intention to proceed this line of investigation using mixtures such as Ne-CF_4, Ar-IsoButane, Ar-CF_4-IsoButane (T2K mixture) — the last one being considered for the LCTPC collaboration, of the International Linear Collider (ILC) experiment.

Acknowledgments

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