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Experimental ion mobility measurements for the LCTPC collaboration — Ar-CF₄ mixtures

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ABSTRACT: In this paper we present the results of the ion mobility measurements made in pure carbon tetrafluoride (CF₄) and gaseous mixtures of argon with carbon tetrafluoride (Ar-CF₄) for pressures ranging from 6 to 10 Torr (8–10.6 mbar) and for low reduced electric fields in the 10 Td to 25 Td range (2.4–6.1 kV·cm⁻¹·bar⁻¹), at room temperature. The time of arrival spectra revealed only one peak throughout the entire range studied which was attributed to CF₃⁺. However, for Ar concentrations above 70%, a bump starts to appear at the left side of the main peak for reduced electric fields higher than 15 Td, which was attributed to impurities. The reduced mobilities obtained from the peak centroid of the time-of-arrival spectra are presented for Ar concentrations in the 5%–95% range.

KEYWORDS: Charge transport and multiplication in gas; Ionization and excitation processes; Gaseous detectors; Ion sources (positive ions, negative ions, electron cyclotron resonance (ECR), electron beam (EBIS))

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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 [4]) for the International Linear Collider (ILC), where ionization of charge particles created during collisions of 1 ms-long bunch trains produces large number of electrons and ions. In the gas amplification stage at the endcaps of the TPC, electron signals are amplified and more ions are produced. To neutralize these ions a gating device will be installed at some distance from the gas amplification stage. It is, therefore, important to know the ion drift mobility for defining this distance and to ensure a high ion reduction while minimizing the thickness of the endplate.

Argon (Ar) has been chosen as the main filling gas for several detectors, while the choice of the quencher is not consensual and is determined by different parameters [3]. Recently, carbon tetrafluoride, CF₄, has been considered for the LCTPC mixture eventually with the addition of isobutane (iC₄H₁₀) [4]. CF₄ has intrinsically many desirable properties: the highest known electron drift velocity and a very low electron transverse diffusion, even in the absence of a magnetic field [3], making it a suitable additive for fast radiation detectors.

Thus, in the present work we take the first step to comprehend the Ar-CF₄-Isobutane mixture (95:3:2) behaviour for the LCTPC Collaboration, by studying the mobility of ions in argon-carbon tetrafluoride (Ar-CF₄) 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 [5–19].

The experimental setup used in the present work, described in detail in [5], 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 higher pressures [13].

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:

$$v_d = KE \quad (1.1)$$

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 [20, 21]. K is usually expressed in terms of reduced mobility K_0 , suppressing the dependence of the mobility values on the gas pressure. Thus

$$K_0 = KN/N_0 \quad (1.2)$$

where N is the gas number density and N_0 is the Loschmidt number ($N_0 = 2.68678 \times 10^{19} \text{ cm}^{-3}$ for 273.15 K and 101.325 kPa according to NIST [22]). The mobility measurements are usually presented as a function of the reduced electric field E/N in units of Td ($1 \text{ Td} = 10^{-21} \text{ V}\cdot\text{m}^2$) and the reduced mobility obtained from equation (1.2) is expressed in terms of $\text{cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$.

1.2 Langevin's theory

According to the Langevin's theory [23], the ions' mobility when the electrostatic hard-core repulsion becomes negligible compared to the neutral polarization effect, can be given by the following equation,

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

where α is the neutral polarisability in cubic angstroms ($\alpha = 1.64 \pm 0.01 \text{ \AA}^3$ for Ar [24] and $\alpha = 3.86 \pm 0.01 \text{ \AA}^3$ for CF_4 [25]) 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 [21]. Although generally accepted, Langevin theory has some known limitations in its application, namely with ions that may undergo a resonant charge transfer, in which case it fails to provide correct values for the ion's mobility [13].

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 when dealing with mixtures

of gases. Blanc found that the mobility of ions in gaseous mixtures, obeyed a simple relationship which can be expressed as follows:

$$\frac{1}{K_{\text{mix}}} = \frac{f_1}{K_{g1}} + \frac{f_2}{K_{g2}} \quad (1.4)$$

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 [26].

2 Method and experimental setup

The mobility measurements presented in this study were obtained using the experimental system described in [5, 15]. 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 an electric field created applying an adequate voltage across its electrodes. After gaining enough energy, the electrons will ionize the gas molecules found 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 that is 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 voltage being applied to the GEM (i.e. without drifting ions), from 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 flash of 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 equation (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.

Despite the fact that no direct identification of the ions is possible with the present system configuration, a procedure based on the control of the voltage across the GEM and the knowledge of the dissociation channels for the possible ions involved together with their product distribution and rate constants, has allowed to solve the ion identification problem.

3 Results and discussion

The mobility of the ions originated in Ar-CF₄ mixtures has been measured for different reduced electric fields E/N (from 10 to 25 Td), 8 Torr pressure 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: one is the occurrence of electric discharges at high E/N , and the other is the deterioration of the time of arrival spectra for very low values of E/N (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, unless mentioned.

3.1 Argon (Ar)

As reported in a previous work [5], in pure Ar two different types of ions are observed for electron impact ionization with energies of about 20 eV: the atomic and the dimer rare gas ions. While the atomic ion (Ar^+) is a direct result of electron impact ionization (equation (3.1)), the dimer ion (Ar_2^+) is, at our working pressures, the result of three-body collision between Ar^+ and two neutrals. The formation process of both can be represented as follows:



The atomic ion has a lower mobility than the dimer one, due to the resonant charge transfer process between Ar^+ ions and Ar atoms, that results in the slowing down of these ions.

Table 1. Ionization products, ionization cross sections for electron impact (25 eV) on Ar [27], appearance energy (A.E.) [28] and respective reaction rates.

Reaction	Cross Sec. (10^{-16}cm^2)	A. E. (eV)	Rate Const.	Ref.
$e^- + \text{Ar} \rightarrow \text{Ar}^+ + 2e^-$	1.24	15.70	—	[27, 28]
$\text{Ar}^+ + \text{Ar} \rightarrow \text{Ar} + \text{Ar}^+$	—	—	$4.6 \times 10^{-10} \text{ cm}^3 \cdot \text{s}^{-1}$	[13]
$\text{Ar}^+ + 2\text{Ar} \rightarrow \text{Ar}_2^+ + \text{Ar}$	—	—	$2.2 \times 10^{-31} \text{ cm}^6 \cdot \text{s}^{-1}$	[29]

3.2 Carbon tetrafluoride (CF_4)

In pure carbon tetrafluoride (CF_4), the mobilities of the ions formed in the parent gas were measured for different reduced electric fields E/N (10 - 45 Td) and for pressures in the 6–10 Torr range at room temperature (293 K) for a constant V_{GEM} of 25 V.

A typical example of a time of arrival spectrum is shown in figure 1, for a pressure of 8 Torr, a reduced electric field of 15 Td, and voltage across the GEM, V_{GEM} , of 25 V.

The primary ion identification is very important, specially when dealing with molecules with several atoms such as CF_4 . By fixing the GEM voltage, it is possible to identify the primary ions produced in the GEM holes and possible reaction channels, that will ultimately lead to the identification of the final ions.

In table 2 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 cross sections for CF_4 primary ionization products and the total cross section provided in [30], 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.

As seen in table 2, 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.

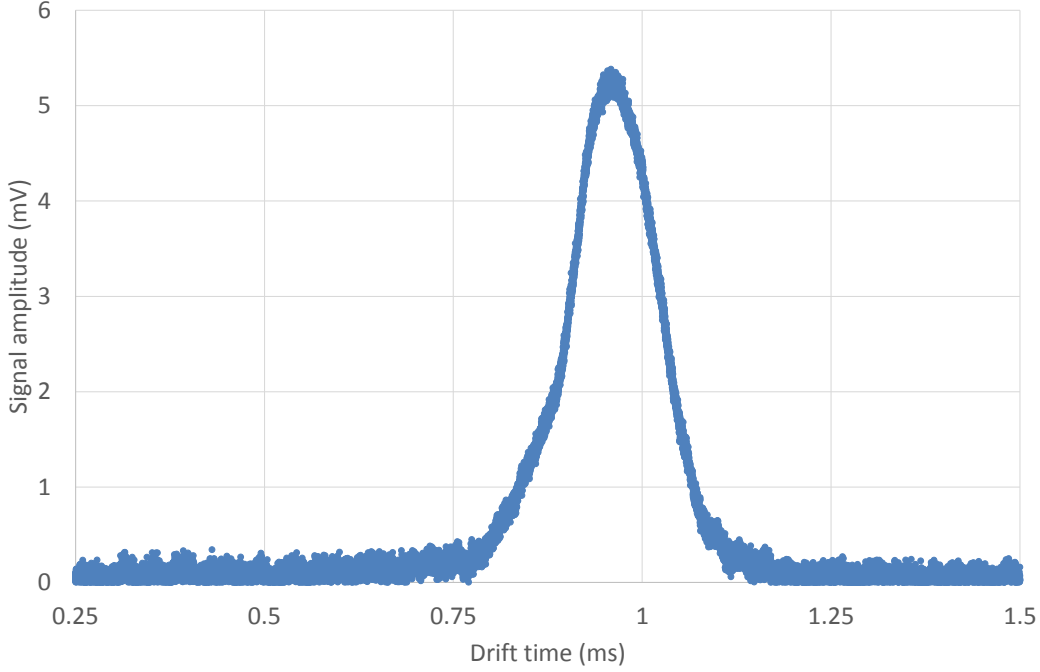


Figure 1. Time-of-arrival spectrum averaged over 128 pulses for pure CF_4 at a pressure of 8 torr, temperature of 293 K and a reduced electric field of 15 Td, with a voltage across GEM of 25 V.

Table 2. Ionization products, ionization cross sections for electron impact 25 eV on CF_4 [30], appearance energies (A.E.) [31], and respective product distribution.

Reaction	Cross Sec. (10^{-16}cm^2)	A. E. (eV)	Prod. Dist.
$e^- + \text{CF}_4 \rightarrow \text{CF}_3^+ + \text{F} + 2e^-$	1.011	15.69	96.1%
$e^- + \text{CF}_4 \rightarrow \text{CF}_2^+ + 2\text{F} + e^-$	0.038	21.47	3.6%
$e^- + \text{CF}_4 \rightarrow \text{CF}^+ + \text{F} + e^-$	0.0017	29.14	0.16%
$e^- + \text{CF}_4 \rightarrow \text{C}^+ + 2\text{F}_2 + e^-$	0.0007	34.77	0.007%
$e^- + \text{CF}_4 \rightarrow \text{F}^+ + \text{CF}_3 + e^-$	0.0007	35	0.007%

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.

To check the correctness of this assumption the trend of the experimental mobility values was compared with the polarization limit from Langevin's formula (equation (1.3)). Figure 2, shows the reduced ion mobility as a function of the reduced electric field in the 10 to 45 Td range, at 8 Torr, with a V_{GEM} of 25 V and at room temperature, together with the polarization Langevin limit ($E/N \rightarrow 0$ Td) for CF_3^+ ion.

The measured mobility tends to $1.12 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, a value in good agreement with the calculated value from equation (1.3) for CF_3^+ , $1.13 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, although it deviates from those obtained by other authors [32, 33]. The difference can be explained by the possibility of the

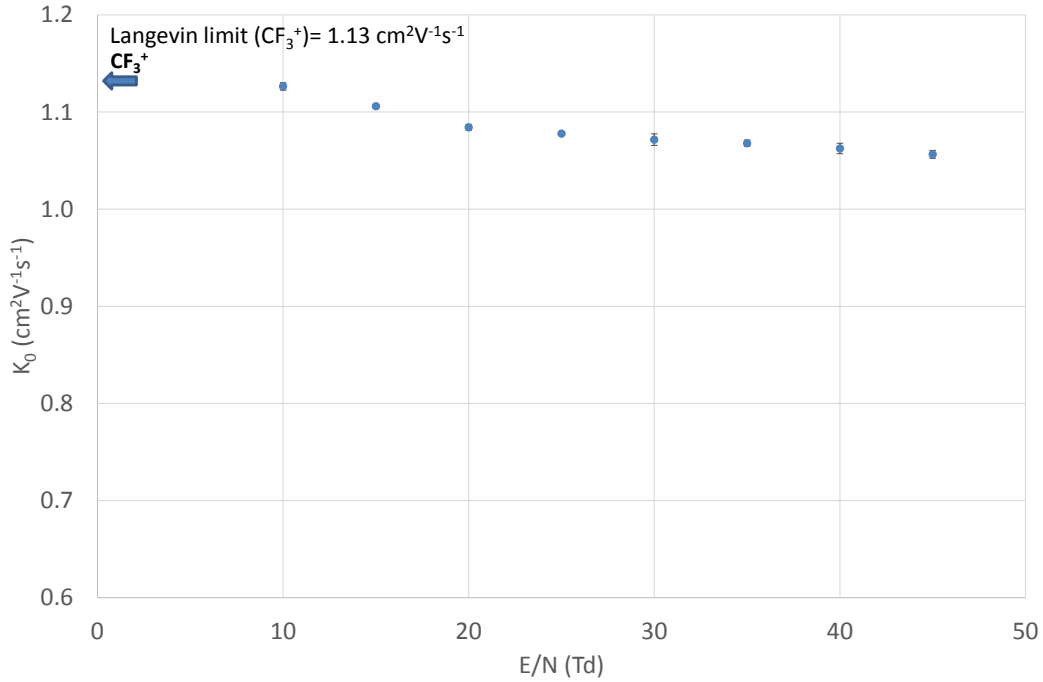


Figure 2. Reduced mobility of the ions produced in CF_4 for a pressure range of 6-10 Torr and for E/N values in the range 10-45 Td range.

cluster formation [34], $\text{CF}_3^+(\text{CF}_4)$, that would be favoured at higher pressures (close to atmospheric) and whose expected value is close to those observed experimentally by these authors. Another explanation could be the fact that the measurements in those references were taken in conditions where the Langevin limit is not strictly applicable as discussed in [21].

3.3 Argon-carbon tetrafluoride (Ar- CF_4) mixture

In carbon-tetrafluoride (Ar- CF_4) mixture, from pure CF_4 and to pure Ar, only one peak was observed throughout the entire range of mixtures studied as can be seen in figure 3. Figure 3 presents several time of arrival spectra for different Ar- CF_4 mixtures (20%, 50%, 70% and 90% of Ar), for a total pressure of 8 Torr, using a V_{GEM} of 25 V and a reduced electric drift field of 15 Td, at room temperature.

The ion responsible for the only peak observed is thought to be the same identified in pure CF_4 , i.e. CF_3^+ . Even though the electron impact ionization cross section for Ar [27] is higher than that for CF_4 [31], anticipating a predominance of Ar^+ down to 45% Ar, these ions rapidly undergo a charge transfer reaction with CF_4 :



with a reaction rate of $7.0 \times 10^{-10} \text{ cm}^3 \cdot \text{s}^{-1}$ [35] much faster than the competitive channel for the production of Ar_2^+ (reaction (3.2)), justifying the appearance of a single peak in the entire mixture range studied. This charge transfer reaction also leads to the suppression of the remaining alternative channels which would produce CF_2^+ , CF^+ , C^+ and F^+ .

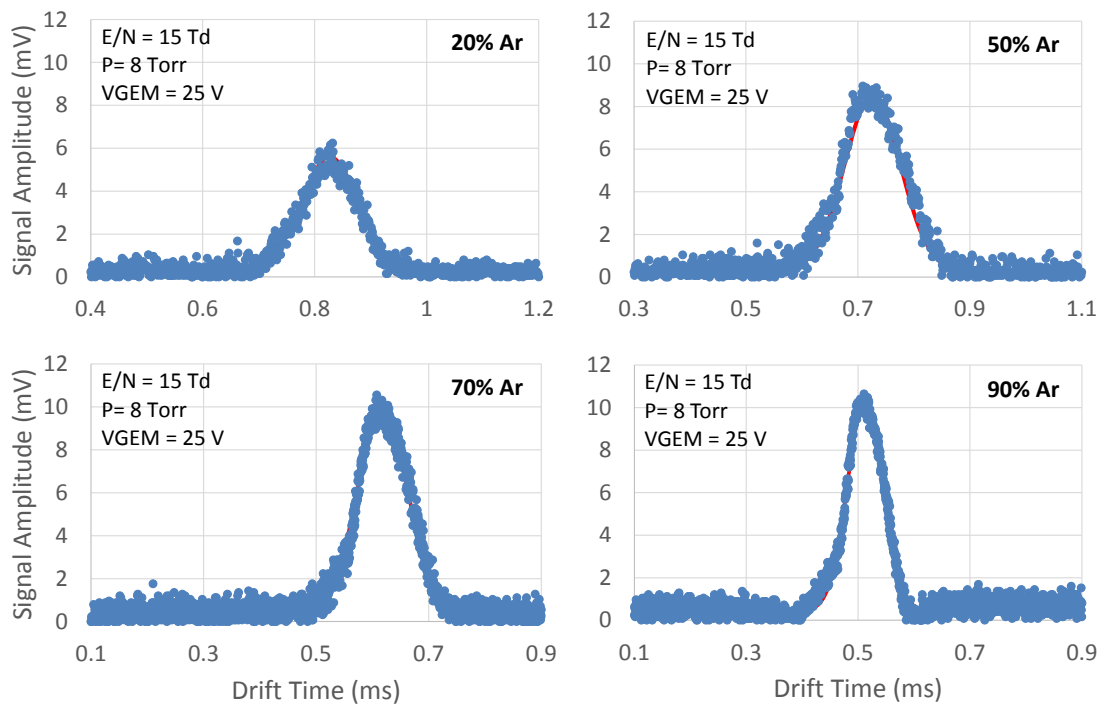


Figure 3. Time-of-arrival spectra (average of 128 pulses) recorded for several Ar-CF₄ mixtures (20%, 50%, 70% and 90% of Ar) at total pressure of 8 torr, reduced electric field of 20 Td, V_{GEM} of 25 V and room temperature (293 K).

Also in figure 3, it is possible to see that the ion drift time decreases, and that a slight bump is present at the left side of the main peak, throughout the whole range of mixtures studied. While the decrease in the drift time of the ions is related to the fact that Ar has a lower polarizability and lower mass than CF₄, a behaviour predicted by the Langevin limit (equation (1.3)), the origin of the small bump however remains unclear.

To identify the origin of both peak and bump, the ion fraction as a function of time was calculated for 95% of Ar at a total pressure of 8 Torr from the cross section data and reaction rates for the different primary ions expected to be present. The results are presented in figure 4.

From this figure, only one peak is expected to be present in the range of concentrations studied, which is confirmed to be CF₃⁺, since the reaction leading to Ar₂⁺ formation (equation (3.2)) is much slower than the one leading to CF₃⁺ ion formation (equation (3.3)). As for the bump, we can observe that at 0.5 ms the concentration of both CF₂⁺ and Ar₂⁺ are negligible, so these possibilities were ruled out. The fact that this bump is present throughout the entire range of mixtures studied, leads us to believe that it may be due to the presence of impurities, namely water vapor from the detector outgassing, which can also explain why the effect observed is independent of the gas composition.

In order to confirm the ion identification, Blanc's law was used as a cross checking method.

Figure 5 shows the experimental values of the reduced mobility for the ions produced in the Ar-CF₄ mixture, for different ratios, in the pressure range of 6-10 Torr and for E/N of 15 Td at room temperature, together with Blanc's law prediction for the main candidate ions - CF₃⁺ (orange), Ar⁺ (red) and Ar₂⁺ (green). K_{g1} and K_{g2} in Blanc's law (equation (1.4)), were obtained

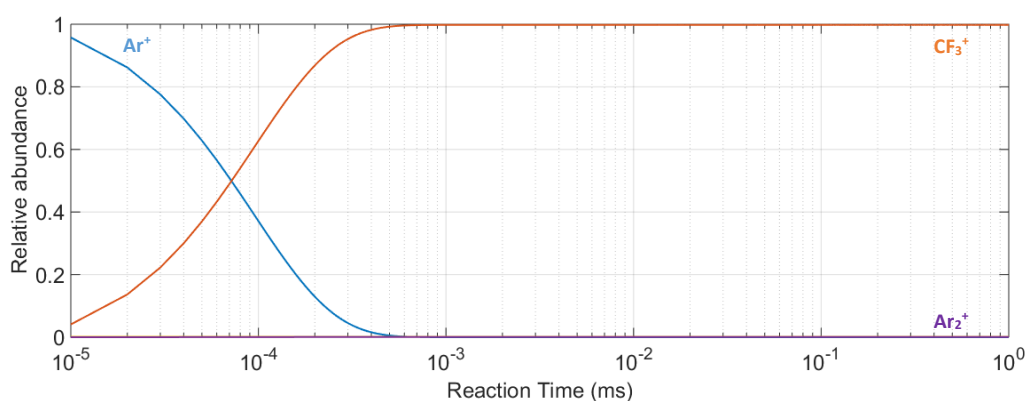


Figure 4. Fraction of the ions that can be formed as a function of the time in Ar-CF₄ mixtures with 95% of Ar at a total pressure of 8 Torr.

either using experimental values from literature or, when not existing, by using the Langevin limit formula (equation (1.3)).

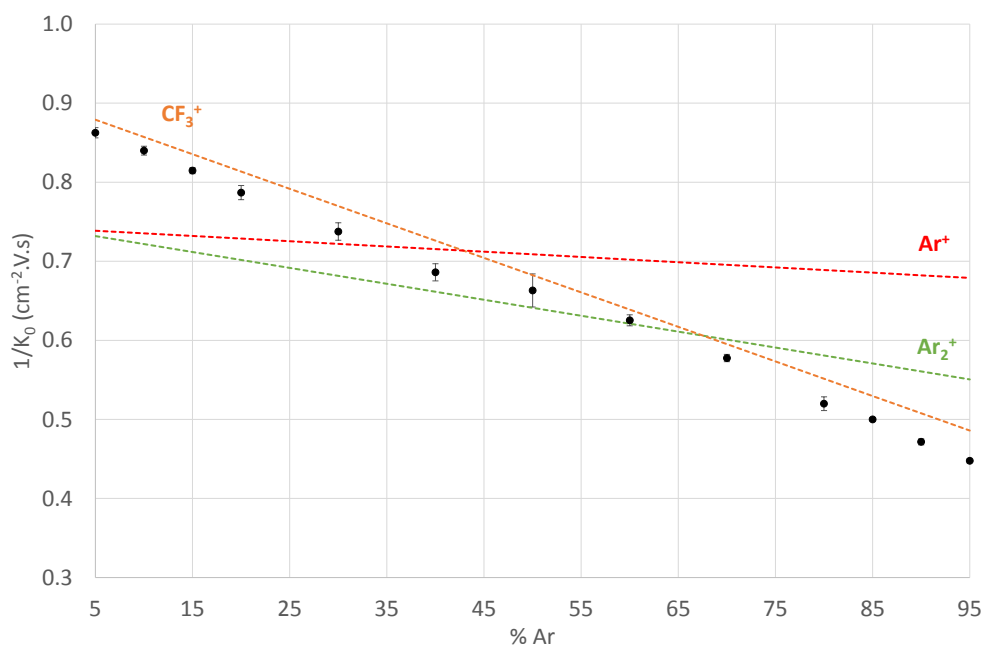


Figure 5. Reduced mobility of the ions produced in the Ar-CF₄ mixture for a pressure of 8 Torr and E/N of 15 Td at room temperature. The dashed lines represent the mobility values expected from Blanc's law for CF₃⁺ (orange), Ar⁺ (red) and Ar₂⁺ (green).

The single peak observed throughout the entire range of mixtures studied displayed a slight increase in mobility when even the slightest concentration of CF₄ was added to Ar. The ion mobility values obtained experimentally follow Blanc's law for the CF₃⁺ ion, for all the studied mixtures, with a deviation of about 5% from the theoretical curve. Table 3 summarizes the results obtained.

No significant variation of the mobility was observed in the range of pressures (6–8 Torr) and for the E/N values (10–25 Td) studied.

Table 3. Mobility of the peaks observed for the Ar-CF₄ mixture ratios studied, obtained for E/N of 15 Td, a pressure of 8 Torr at room temperature (293 K).

Mixture	Mobility (cm ² ·V ⁻¹ ·s ⁻¹)	Ion
5% Ar	1.159 ± 0.008	CF ₃ ⁺
10% Ar	1.191 ± 0.007	CF ₃ ⁺
15% Ar	1.228 ± 0.005	CF ₃ ⁺
20% Ar	1.271 ± 0.013	CF ₃ ⁺
30% Ar	1.356 ± 0.018	CF ₃ ⁺
40% Ar	1.458 ± 0.021	CF ₃ ⁺
50% Ar	1.509 ± 0.044	CF ₃ ⁺
60% Ar	1.599 ± 0.016	CF ₃ ⁺
70% Ar	1.731 ± 0.012	CF ₃ ⁺
80% Ar	1.924 ± 0.029	CF ₃ ⁺
85% Ar	2.000 ± 0.007	CF ₃ ⁺
90% Ar	2.120 ± 0.015	CF ₃ ⁺
95% Ar	2.234 ± 0.013	CF ₃ ⁺

4 Conclusion

In the present work we measured the reduced mobility of ions originated by electron impact with an energy up to 25 eV in pure CF₄ and Ar-CF₄ mixtures for pressures from 6 to 10 Torr, low reduced electric fields from 10 to 25 Td and different mixture ratios (from 0 to 100% Ar).

Only one peak was consistently observed for the different concentrations of Ar in the mixture which, is thought to be originated by CF₃⁺ as corroborated by Blanc’s law. Our experimental results show the same consistency within 5% with the ones predicted by the Blanc law throughout the mixture range studied. For high Ar concentrations, above 70%, a bump starts to appear at the left side of the main peak for reduced electric fields higher than 15 Td, which was attributed to impurities.

The experimental mobility values did not display a significant dependence over the studied range of pressure and E/N studied (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 Xe-CF₄, Ne-CF₄, Ar-IsoButane, Ar-CF₄-IsoButane (T2K mixture) the last one necessary for the LCTPC collaboration, of the International Linear Collider (ILC) experiment.

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