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ABSTRACT: Data on ion mobility is important to improve the performance of large volume gaseous detectors. In the present work the method, experimental setup and results for the ion mobility measurements in Xe-CO$_2$ mixtures are presented. The results for this mixture show the presence of only one peak for all gas ratios of Xe-CO$_2$, low reduced electric fields, E/N, 10-25 Td (2.4-6.1 kV·cm^{-1}·bar^{-1}), low pressures 6-8 Torr (8-10.6 mbar), at room temperature.

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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], and also in IMS (Ion Mobility Spectrometry) a technique used for the detection of narcotics and explosives [4].

One of these examples are the so-called Transition Radiation Detectors (TRDs), used for particle identification at high momenta [5, 6]. The choice of the gas mixture for such detectors is determined by several parameters such as high electron/ion velocity and low electron diffusion, which are of key importance [3] as they influence the rate capability and signal formation of detectors such as the Multi-Wire Proportional Chambers (MWPCs) [7]. Xenon (Xe) is considered to be the best choice as main gas, while the choice of the best quencher is not unanimous [3]. One effective quenching gas is methane, (CH$_4$) but, due to its flammability its usage is limited [3], and is usually substituted by carbon dioxide (CO$_2$). Presently, xenon-carbon dioxide (Xe-CO$_2$) mixtures are widely used in high energy physics experiments such as ALICE [6, 8] and CBM TRD [6, 9] and in order to fully understand and predict their performance it is important to have detailed information on the transport properties of ions in these gas mixtures.

The experimental setup used in the present work (described in detail in [10]) allows the measurement of ion mobility in gas mixtures. Initially thought for high pressure, it was converted into a low pressure gas system. 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 pressure [11].

In this work, the mobility of ions in Xe-CO$_2$ gas mixtures was measured at pressures in the 6 to 8 Torr (8 to 10.6 mbar) range and for reduced electric fields commonly used in gaseous detectors, 15 Td (3.6 kV·cm^{-1}·bar^{-1}), extending previous studies developed in our group for other gases [10–20].

1.1 Ion mobility

Under a weak and uniform electric field a group of ions will eventually reach a steady state. In this conditions, the average velocity of this group of ions, also known as drift velocity v_d, is proportional to the electric field intensity [4]:

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

where K is the mobility of the ions, expressed in units of $cm^2 \cdot V^{-1} \cdot s^{-1}$ and E the intensity of the drift electric field. The ion mobility, K, is normally expressed in terms of the reduced mobility K_0,

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

with N the gas number density and N_0 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 values are commonly presented as a function of the reduced electric field E/N in units of Townsend (1 Td=10^{-17} V.cm^2).

1.2 Langevin's theory

According to Langevin's theory [22], one limiting value of the mobility is reached when the electrostatic hard-core repulsion becomes negligible compared to the neutral polarization effect [24]. 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 ($\alpha = 4.044$ Å^3 for Xe [23] and $\alpha = 2.93$ Å^3 for CO_2 [23]) and μ is the ion-neutral reduced mass in unified atomic mass units. Although the Langevin limit applies rigorously for real ion-neutral systems only in the double limit of low E/N and low temperature, it predicts the low-field mobility at room remperature with relatively good accuracy [24], which is the case in our experimental conditions. Although generally accepted, Langevin theory, has some known limitations in its application, namely with CO_2, where it fails to provide correct values for the ion's mobility [11].

1.3 Blanc's law

In binary gaseous mixtures Blanc's law has proven to be most useful when determining the ions' mobility. According to this law the reduced mobility of the ion in the binary mixture, K_{mix}, can be expressed as follows:

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

where K_{g1} and K_{g2} are the reduced mobility of that same ion in an atmosphere of 100% of gas #1 and #2 respectively and 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 [10]. 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 GEM that is inside a gas vessel. The photoelectrons released from the CsI film are guided through the GEM holes, ionizing 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. A pre-amplifier is used to convert the charge collected into a voltage signal, and the time spectra are recorded in a digital oscilloscope. After the background subtraction from the signal, gaussian curves are fitted to the time of arrival spectra from which the peak centroids are obtained. Since the peaks' centroid correspond to the average drift time of the ions along a known fixed distance (4.273 cm), the drift velocity and mobility can then be calculated. One important feature of the system is the capability of controlling the voltage across the GEM (V_{GEM}), which limits the maximum energy gained by the electrons as they move across the GEM holes, narrowing the variety of possible primary ions produced. Identifying the primary ions will allow to pinpoint secondary reaction paths that lead to the identification of the detected ions which has been providing correct and consistent results with this experimental setup.

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.

The method described together with the knowledge of the dissociation channels, product distribution and rate constants represent a valid, although elaborate, solution to the ion identification problem.

3 Results and discussion

The mobility of the ions originated in Xe-CO_2 mixtures has been measured for different reduced electric fields E/N (from 10 Td up to 25 Td) and at 8 Torr pressure at room temperature (293 K).

The range of the reduced electric field values used to determine the ions' mobility is limited by two distinct factors: one is the electric discharges that occur at high E/N values; the other is the observed 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.

Previous work on the mobilities and ionization processes of Xe [10] and CO_2 [18] in their parent gases has already been performed in our group.

The range of E/N values considered in this work is within the validity conditions of Blanc's law, this is, in the region of low E/N [26, 27].

3.1 Xe-CO_2 mixture

In xenon-carbon dioxide (Xe-CO_2) mixtures, one peak is observed for all mixtures studied, from pure Xe to pure CO_2, as can be seen in figure 1, where the drift spectra for several Xe-CO_2 mixtures (5%, 25%, 50% and 95% of Xe) are displayed, at 8 Torr, 293 K and 15 Td with a V_{GEM} of 20 V.

The ion responsible for the peak was found to depend on the mixture ratio according to the cross sections and rate constants in table 1: for Xe concentrations down to 15% the ions are from Xe (Xe_2^+/Xe^+), while below this concentration the main ion is believed to be a CO_2 ion ($CO_2^+ \cdot CO_2$).
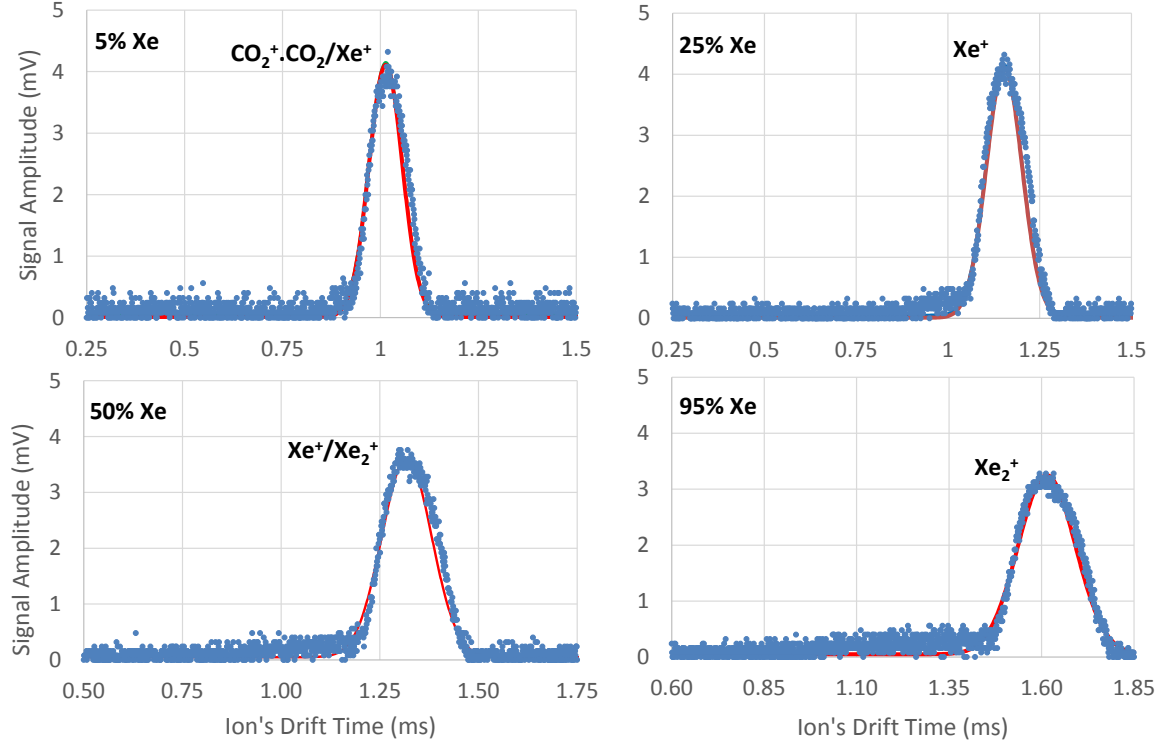
Figure 1. Time-of-arrival spectra averaged over 128 pulses for several Xe-CO_2 mixtures (5%, 25%, 50% and 95% of Xe) at a pressure of 8 Torr, temperature of 293 K and for a reduced electric field of 15 Td with a voltage across GEM of 20 V (background noise subtracted).

Table 1. Summary of possible reactions and respective rate constants or cross section for electron impact ionization at 20 eV (references on the last collumn).

Reaction	Rate Constant	Cross section (20 eV)	Ref.
$e^- + Xe \rightarrow Xe^+ + 2e^-$	-	$2.43\pm0.12\times10^{-16}$ cm^2	[28]
$Xe^+ + Xe \rightarrow Xe + Xe^+$	2.5×10^{-10} cm^3.s^{-1}	-	[29]
$Xe^+ + 2Xe \rightarrow Xe_2^+ + Xe$	$2.0\pm0.2\times10^{-31}$ cm^6.s^{-1}	-	[30]
$e^- + CO_2 \rightarrow CO_2^+ + 2e^-$	-	$0.452\pm0.032\times10^{-16}$ cm^2	[31]
$CO_2^+ + CO_2 \rightarrow CO_2 + CO_2^+$	$3.7\pm0.37\times10^{-10}$ cm^3.s^{-1}	-	[32]
$CO_2^+ + CO_2 + M \rightarrow CO_2^+.CO_2 + M$	$2.1\pm0.3\times10^{-28}$ cm^6.s^{-1}	-	[33]
$CO_2^+ + Xe \rightarrow Xe^+ + CO_2$	$6.0\pm1.8\times10^{-10}$ cm^3.s^{-1}	-	[34]

From the total ionization cross section for electron impact (table 1) at an energy of 20 eV for Xe we can observe that the ionization probability for Xe atoms is about 5 times higher than that for CO_2 molecule, thus it is expected that even at low Xe concentrations (down to 15% of Xe), Xe ions are still the ones preferentially produced.

Using the reaction rates together with the electron impact ionization cross sections, the time evolution of the ion species for a total pressure of 8 Torr, was calculated and the results obtained for
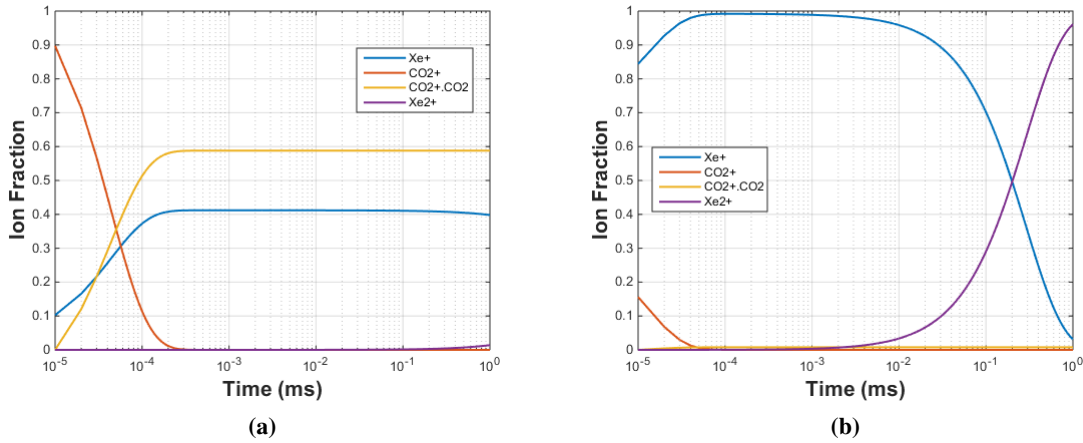
Figure 2. Fraction of ions that can be formed as a function of time for Xe-CO_2 mixtures of 5% (a) and 50% (b) of Xe, for a total pressure of 8 torr.

5% and 50% Xe concentrations are shown in figure 2a and figure 2b, respectively. As can be seen, the fraction of the different ion species present at the end of the drift distance will depend on the reaction time and on the total drift time. Figure 2 can help to interpret figure 1: down to about 8% Xe the production of Xe^+ ions will be more abundant, leading to the same ions as in pure Xe (Xe_2^+ and Xe^+), while below this Xe concentration, $CO_2^+.CO_2$ ions will be produced more abundantly.

According to figure 2a (5% Xe), the estimated fraction of each ion species collected can be determined. In this case, the species expected to be present are: 39.8% of Xe^+, 1.4% of Xe_2^+ and 58.8% of $CO_2^+.CO_2$, at the instant corresponding to the detected ions' average drift time (estimated from the peaks's centroid of figure 1 at 5% Xe, to be about 1 ms). In spite of the fact that two different ions ($CO_2^+.CO_2$ and Xe^+) are expected from the analysis of figure 2a, only one peak was observed in figure 1. This can be explained by the fact that both ions have similar mobilities, according to Blanc's law, therefore contributing to the same peak in the time spectra. Being energetically favourable, the charge transfer from $CO_2^+.CO_2$ to Xe could also explain the fact that only one peak was observed in figure 1. However, since data for this reaction is not available in the literature it was not considered in the analysis performed in figure 2.

Further increasing the concentration of Xe in the mixture will lead first to the formation of Xe^+ and ultimately to Xe_2^+. In fact, looking at figure 2b, at the instant corresponding to the detected ions' average drift time inferred from figure 1 for 50% Xe (about 1.3 ms), the estimated fraction of each ion species is 96.1% of Xe_2^+ and 3.1% of Xe^+ (adding up to 99.2%), while the ions formed at the GEM are expected to be 84.4% Xe^+ and 15.6% CO_2^+.

Thus, though CO_2^+ ions are produced at the GEM in both cases, with a comparatively higher abundance at 5% Xe, the end product is essentially a Xe ion, namely Xe_2^+ coming from Xe^+ through the three-body reaction:

$$Xe^+ + Xe + M \rightarrow Xe_2^+ + M \tag{3.1}$$

where M is an atom or molecule from the gas mixture, in our case Xe or CO_2, that removes the excess energy.

As for the Xe$^+$ ion, it can be originated either by direct electron impact ionization of Xe or by the charge transfer reaction,

$$CO_2^+ + Xe \rightarrow Xe^+ + CO_2 \tag{3.2}$$

which in the pressure conditions of this experiment is a much faster reaction than the three-body competing one:

$$CO_2^+ + CO_2 + M \rightarrow CO_2^+.CO_2 + M \tag{3.3}$$

as can be seen in figure 2b for 50% of Xe.

Since according to [35] there is no charge transfer between Xe$_2^+$ and CO$_2$ we expect that, once formed, that ion will remain unaltered through the rest of the drift distance.

Other relevant features include the variation of the FWHM and peak position (drift time) in the drift spectra that can also be observed in figure 1. The larger FWHM for the peak as Xe concentration increases can be explained by the higher longitudinal diffusion of the Xe$_2^+$ ion in the medium due to higher energy loss probability in collisions with higher mass particles.

As for the shift of the peak in the drift spectrum towards lower drift times (increasing ion mobility) with the decrease in Xe concentration, it can be explained by the lower CO$_2$ mass compared to the Xe atom, which implies a lower reduced mass (μ in the Langevin limit eq. (1.3)) in ion-neutral collision, implying a higher mobility.

In figure 3 we plot the inverse reduced mobility obtained for the ions produced in Xe-CO$_2$ mixtures as a function of Xe percentage, for 8 Torr and 15 Td, at room temperature (293 K), together with Blanc's law prediction for Xe$_2^+$ (blue dashed line), Xe$^+$ (green dashed line) and CO$_2^+$.CO$_2$ (orange dashed line). K_{g1} and K_{g2} in Blanc's law (eq. (1.4)), were obtained either using experimental values from literature or, when not possible, by extrapolating from existing experimental data.

As mentioned before, Langevin theory has some limitations, namely in CO$_2$, yielding mobilities higher than the experimental values. To address the lack of experimental values for Xe$^+$ and Xe$_2^+$ in pure CO$_2$ an alternative method was devised using the data in paper [11] for pure CO$_2$.

The values were obtained using the ion mass-mobility data for CO$_2$ in [11] and the masses of Xe$^+$ and Xe$_2^+$ yielding mobility values of 1.15$\pm$0.05 cm$^2 \cdot$V$^{-1} \cdot$s^{-1} and 1.05$\pm$0.10 cm$^2 \cdot$V$^{-1} \cdot$s^{-1}, respectively. A similar analysis of the ion mobility-mass data in Xe, obtained from [36] showed that in pure Xe the ion mobility follows fairly well the values obtained from the Langevin polarization limit. For this reason the mobility value of CO$_2^+$.CO$_2$ ion in Xe used in this work is the one obtained from the Langevin limit, 1.05 cm$^2 \cdot$V$^{-1} \cdot$s^{-1}, which helps to support this approach.

As seen in figure 3 the ion mobility experimentally obtained follows, within error bars, Blanc's law prediction for the most abundant ion in the different Xe percentages studied. In fact, from 100% down to 25% Xe the behaviour of Xe$_2^+$ is followed, while between this concentration and 15% it deviates towards Xe$^+$ predicted behaviour, and below 15% Xe it follows Blanc's law prediction for CO$_2^+$.CO$_2$. This behaviour is consistent with reaction rates and pressure data: in pure Xe the ion present is Xe$_2^+$. However, as Xe concentration decreases, this ion spends an increasing part of its drift time as Xe$^+$, with its mobility reflecting this growing contribution. As a consequence, the mobility of Xe$_2^+$ will decrease, eventually reaching the predicted value (Blanc's law) of Xe$^+$, for Xe concentrations from 25% down to around 15% of Xe. At about 15% Xe, as seen before, both primary ions (CO$_2^+$ and Xe$^+$) will be equally produced at the GEM, although only below 8% Xe the most abundantly produced ion changes to CO$_2^+$.CO$_2$, as until that concentration the charge exchange between
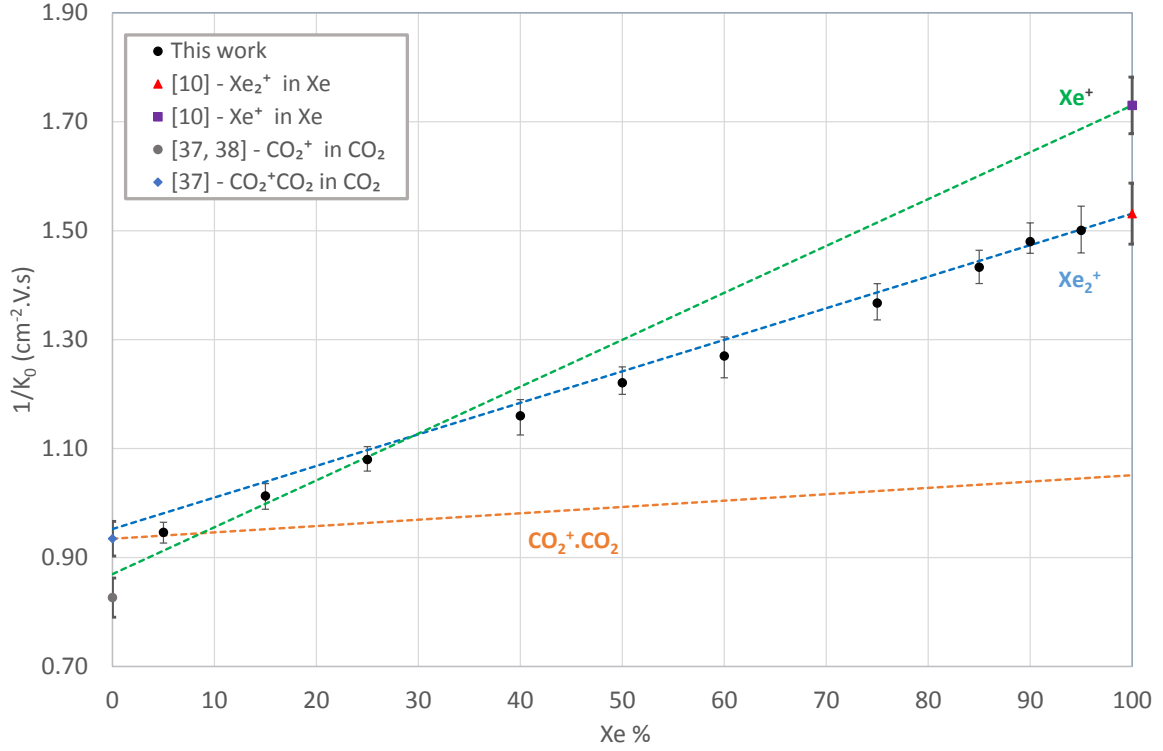
Figure 3. Inverse of the reduced mobility of the ions produced in the Xe-CO$_2$ mixture for a pressure of 8 Torr and for a E/N of 15 Td at room temperature. The dotted lines represent the mobility values expected from Blanc's law for Xe$_2^+$ (blue), Xe$^+$ (green) and for CO$_2^+$.CO$_2$ (orange).

CO$_2^+$ and Xe still makes Xe$^+$ the more abundant ion. As a consequence, the mobility of the observed ion approaches the predicted theoretical value for the CO$_2$ cluster ion. In spite of the fact that Xe$^+$ and CO$_2^+$.CO$_2$ have different masses, according to Blanc's law they have similar mobilities between 5%-15% Xe, thus becoming indistiguishable in the drift spectra. So, despite the presence of two different ions, only one peak is visible in the spectrum, corroborating what was discussed earlier in figure 2.

Finally, for high Xe concentrations the introduction of CO$_2$ leads to an increase in the mobility of the Xe ions present, *i.e* Xe$_2^+$. The ion mobility values measured in this experiment range from 1.06±0.02 (5% Xe) to 0.67±0.02 cm^{2}V^{-1}s^{-1} (95% Xe), for E/N = 15 Td and 8 Torr. No significant variation of the mobility was observed in the range of pressures (6-8 Torr) and of E/N (10-25 Td) studied. The results here presented are in good agreement with the experimental values obtained by other authors [7, 39] at standard pressure and similar E/N. Table 2 summarizes the results obtained.

4 Conclusion

In the present work we measured the reduced mobility of ions originated by electron impact in Xe-CO$_2$ mixtures at pressures ranging from 6 to 8 Torr, low reduced electric fields (10-25 Td) and different mixture ratios. The experimental results show that, for the range of concentrations studied, only one peak was observed in the time-of-arrival spectrum, although in some cases the peak can correspond to more than one ion. The ions responsible for these peaks are believed to be the ones originated in Xe (Xe$_2^+$ and Xe$^+$) for concentrations of Xe down to 15% while for concentrations

Table 2. Mobility of the ions observed for the Xe-CO$_2$ mixture ratios studied, obtained for E/N of 15 Td, at 8 Torr and 293 K.

Xe-CO$_2$ Mixture % of Xe	Mobility (cm^2·V^{-1}·s^{-1})
5%	1.06 $\pm$ 0.02
10%	1.03 $\pm$ 0.02
15%	0.99 $\pm$ 0.02
25%	0.93 $\pm$ 0.02
40%	0.86 $\pm$ 0.02
50%	0.82 $\pm$ 0.01
60%	0.79 $\pm$ 0.02
75%	0.73 $\pm$ 0.02
85%	0.70 $\pm$ 0.01
90%	0.68 $\pm$ 0.01
95%	0.67 $\pm$ 0.02

below 15% of Xe, the ions observed are mainly originated by CO$_2$, namely the cluster CO$_2^+$.CO$_2$. Between 25% and 15% Xe, the mobility of the only ion observed, Xe$_2^+$, starts to be affected by the increasing drift path travelled as Xe$^+$, therefore closely approaching the theoretical values given by Blanc's law for Xe$^+$. For lower Xe concentrations (around 8% of Xe), the ion mobility values approach the mobility of CO$_2^+$.CO$_2$ theoretical value. The ions' mobility was seen to increase with the increasing CO$_2$ concentration in the mixture with the behaviour following roughly Blanc's law through the entire range for the different ions present.

The mobilities calculated did not display a significant dependence either with pressure in the range studied (6-8 Torr), or with the E/N for the values used in this work (10-25 Td).

Future work is expected with other gaseous mixtures. It is our intention to extend the work on ion mobility using different mixtures of known interest such as Xe-C$_2$H$_6$, Xe-CH$_4$, Ar-CF$_4$, Ar-CF$_4$-Isobutane, Ne-CF$_4$ and Xe-CF$_4$.

Acknowledgments

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