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Experimental ion mobility measurements in Xe-CH₄

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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-CH₄ mixtures are presented. The results for this mixture show the presence of two distinct groups of ions. The nature of the ions depend on the mixture ratio since they are originated by both Xe and CH₄. The results here presented were obtained for low reduced electric fields, E/N , 10–25 Td (2.4–6.1 kV · cm⁻¹ · bar⁻¹), at low pressure (8 Torr) (10.6 mbar), and at room temperature.

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], 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 TRDs' of the Multi-Wire Proportional Chambers type (MWPCs) [7]. Xenon (Xe) is considered to be the best choice as the main gas, while the choice of the best quencher is not unanimous [3]. One effective quenching gas is methane (CH₄) but, due to its flammability its usage is limited [3]. Still, xenon-methane (Xe-CH₄) mixtures are used in high energy physics experiments such as DØ [8, 9], HERMES [10] and PHENIX TRDs [11, 12], being 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 [13]) 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 [14].

In this work, the mobility of ions in Xe-CH₄ gas mixtures was measured at 8 Torr (10.6 mbar) and for reduced electric fields commonly used in gaseous detectors, 20 Td (4.5 kV · cm⁻¹ · bar⁻¹), extending previous studies developed in our group for other gases [13–25].

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]:

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

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

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

with N the gas number density and N_0 the Loschmidt number ($N_0 = 2.68678 \times 10^{19} \text{ cm}^{-3}$ for 273.15 K and 101.325 kPa according to NIST [26]). The mobility values are commonly presented as a function of the reduced electric field E/N in units of Townsend ($1 \text{ Td} = 10^{-17} \text{ V} \cdot \text{cm}^2$).

1.2 Langevin's theory

According to Langevin's theory [27], one limiting value of the mobility is reached when the electrostatic hard-core repulsion becomes negligible compared to the neutral polarization effect [28]. This limit is 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 ($\alpha = 4.044 \text{ \AA}^3$ for Xe [29] and $\alpha = 2.62 \pm 0.01 \text{ \AA}^3$ for CH_4 [30]¹) and μ is the ion-neutral reduced mass in unified atomic mass units. Although the Langevin limit only applies rigorously for real ion-neutral systems only in the double limit of low E/N and low temperature, it still predicts the low-field mobility at room temperature with relatively good accuracy [28], which is the case in our experimental conditions. Although generally accepted, Langevin theory, has some known limitations in its application, namely with ions that undergo resonant charge transfer, where it fails to provide correct values for the ion's mobility [14].

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:

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

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 [31].

¹Fundamental information on molecular polarizabilities.

2 Method and experimental setup

The mobility measurements presented in this study were obtained using the experimental system described in [13]. 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. 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.

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, which has been providing correct and consistent results for several gas mixtures.

3 Results and discussion

The mobility of the ions originated in Xe-CH₄ mixtures has been measured for different reduced electric fields E/N (from 10 Td up to 25 Td), at 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 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 $1.2 \text{ kV} \cdot \text{cm}^{-1} \cdot \text{bar}^{-1}$), which has been attributed to collisions between the ions and impurity molecules.

Previous work on the mobilities and ionization processes of Xe [13] and CH₄ [22] 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 [32, 33].

3.1 Xenon (Xe)

Regarding the pure xenon (Xe) case, only one peak is observed for electron impact energy of about 20 eV using a reduced electric field of 15 Td and a pressure of 8 Torr at room temperature. The ion

responsible for the peak observed is the Xe dimer ion (Xe_2^+). While the atomic ion (Xe^+) is a direct result of electron impact ionization [34], Xe_2^+ is the result of the following reaction:



3.2 Methane (CH_4)

In pure CH_4 , two peaks were observed and reported in a previous work [17]. These two peaks were identified as corresponding to CH_5^+ (peak with higher mobility) and to a 2-carbon ion group (C_2H_n^+) plus C_3H_7^+ (peak with lower mobility), which result from reactions involving the primary ions and CH_4 molecules. Other authors have observed three peaks in the same conditions as already discussed in [17]. These primary ions can be found in table 1, where we summarize the possible reactions due to electron impact in CH_4 for electron energies of 20 eV, together with the respective cross sections, appearance energies and the product distribution. The probabilities presented were obtained using the cross sections for CH_4 primary ionization products and the CH_4 total cross section provided in [38], which allowed us to infer the product distribution of the primary ionization.

Table 1. Ionization products, ionization cross sections for electron impact (20 eV) on CH_4 [38], appearance energies (A.E.) [39] and respective product distribution.

Reaction	Cross Sec. (10^{-16} cm^2)	A.E. (eV)	Prod. Dist.
$\text{CH}_4 + e^- \rightarrow \text{CH}_4^+ + 2e^-$	0.892	12.65±0.40	56.7%
$\text{CH}_4 + e^- \rightarrow \text{CH}_3^+ + \text{H}^- + e^-$	0.512	13.58±0.10	32.6%
$\text{CH}_4 + e^- \rightarrow \text{CH}_3^+ + \text{H} + 2e^-$		14.34±0.10	
$\text{CH}_4 + e^- \rightarrow \text{CH}_2^+ + \text{H}_2 + 2e^-$	0.169	15.10±0.10	10.7%

Table 2 presents a summary of the chemical reactions, their product distribution, and respective reaction rates for the reactions between the primary ions displayed on table 1 and CH_4 molecules, at room temperature [40].

Table 2. Ionization reactions, product distribution and rate constants for the collisions of the primary ions with CH_4 . Adapted from [40, 41].

Reaction	Prod. Dist.	Rate Const. ($10^{-9} \text{ cm}^3 \text{ s}^{-1}$)
$\text{CH}_4 + \text{CH}_4^+ \rightarrow \text{CH}_5^+ + \text{CH}_3$	1.00	1.140±0.171
$\text{CH}_4 + \text{CH}_3^+ \rightarrow \text{C}_2\text{H}_5^+ + \text{H}_2$	1.00	1.100±0.165
$\text{CH}_4 + \text{CH}_2^+ \rightarrow \text{C}_2\text{H}_4^+ + \text{H}_2$	0.70	1.300±0.195
$\text{CH}_4 + \text{CH}_2^+ \rightarrow \text{C}_2\text{H}_5^+ + \text{H}$	0.30	
$\text{CH}_4 + \text{C}_2\text{H}_5^+ \rightarrow \text{C}_3\text{H}_7^+ + \text{H}_2$	1.00	0.00009±0.0000135

The reactions presented on table 1 and 2 corroborate the explanation of the results obtained for pure CH_4 , justifying the attribution of the most intense peak to CH_5^+ (lighter, thus with higher mobility) and the smaller one to a group of ions which include C_2H_4^+ , C_2H_5^+ and C_3H_7^+ (heavier, thus with lower mobility).

3.3 Xe-CH₄ mixture

In xenon-methane (Xe-CH₄) mixtures, two distinct groups of ions are observed for all mixture compositions studied, from pure Xe to pure CH₄, as can be seen in figure 1, where the drift spectra for several Xe-CH₄ mixtures (10%, 50%, 70% and 95% of Xe) are displayed, at 8 Torr, 293 K and 15 Td with a V_{GEM} of 20 V. The ions responsible for the several peaks were found to depend on the mixture ratio, suggesting that they are originated by both CH₄ and Xe.

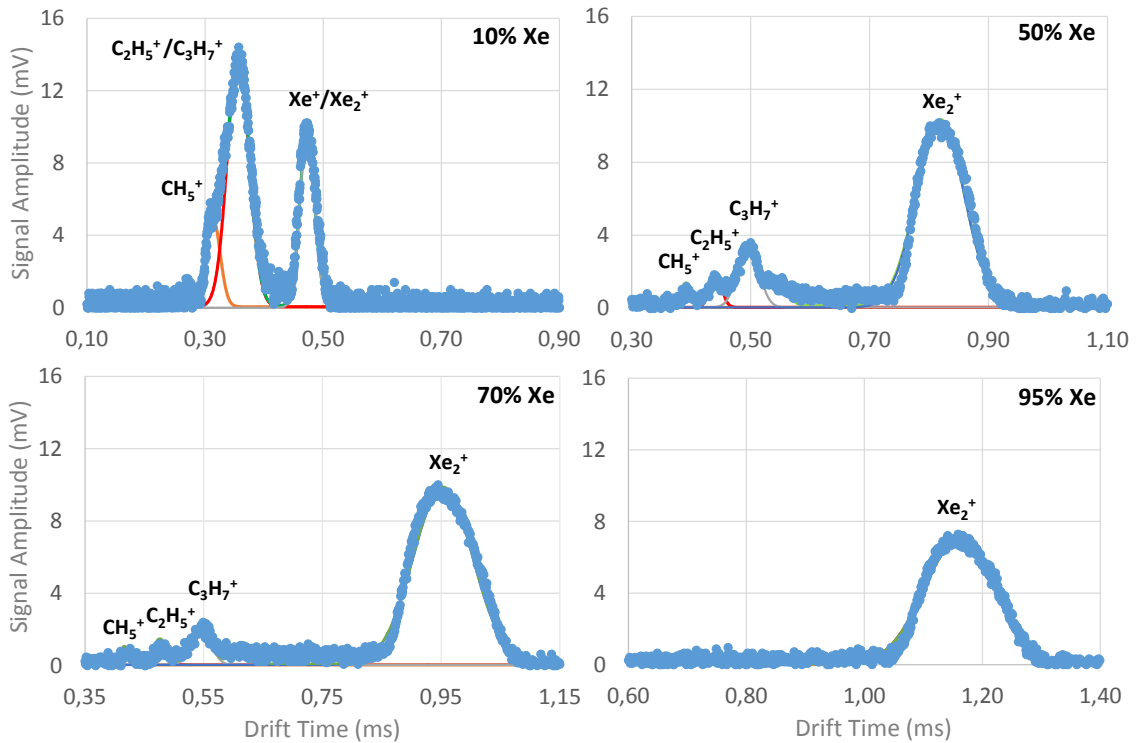


Figure 1. Time-of-arrival spectra averaged over 128 pulses for several Xe-CH₄ mixtures (10%, 50%, 70% and 95% of Xe) at a pressure of 8 Torr, temperature of 293 K and for a reduced electric field of 20 Td with a voltage across GEM of 20 V (background noise subtracted).

Looking at figure 1, there are two relevant aspects both as a result of increasing Xe concentration in the mixture: one is the decrease in mobility of the different ions observed, and the other is the change of the dominant ion species present, which can be identified by a decrease in the area of the group of ions with higher mobility, and an increase in the peak area of the second group of ions, indicating that the faster group of ions are generated from CH₄ molecules while the second, and slower group, is originated by Xe atoms.

As for the shift of the peaks towards higher drift times in the drift spectrum (decreasing ion mobility) with increasing Xe concentration, it can be explained by the lower CH₄ mass when compared to that of the Xe atom, which implies a lower reduced mass (μ in the Langevin limit eq. (1.3)) in ion-neutral collision, thus a lower mobility.

From the experimental results it is possible to conclude that the ions observed depend on the relative abundance of the gases. Starting from pure CH₄ and up to 5% Xe, only two peaks are

observed. The ions responsible for these peaks are the two groups identified in pure CH₄: CH₅⁺ (the peak with higher mobility) and C₃H₇⁺ (the most intense peak). As mentioned previously, the CH₅⁺ is originated by CH₄⁺ through the reaction,



whose probability may be further enhanced by the charge transfer of Xe⁺ to CH₄. C₃H₇⁺ comes from a two-step reaction. The first is the production of C₂H₅⁺, an intermediary product of CH₄, which will further react with CH₄ leading to the formation of C₃H₇⁺:



Increasing Xe concentration from 5% up to 20% causes the appearance of a new peak, as can be observed in figure 1 for 10% Xe, with lower mobility (at the right side) of the C₃H₇⁺ peak. This peak, is clearly related to the availability of Xe, since its area increases with Xe concentration. Between 5% and 20% Xe, reaction (3.1) is very slow, and is not completed during the drift time of the group of ions. So, Xe⁺ is expected to be the ion responsible for the appearance of this peak. Once formed, and since the ionization energy of Xe⁺ is lower than that of the ions originated by CH₄, it is expected that Xe⁺ won't transfer its charge to CH₄ (see table 3 and 1). Between 20% and 80% Xe, in addition to the already mentioned peaks, another one starts to appear at the left side of the peak attributed to C₃H₇⁺. The ion considered to be responsible for this peak is C₂H₅⁺, which has lower mass, thus is expected to have higher mobility. This ion results from the longer reaction time for the formation C₃H₇⁺ (eq. (3.4)) at lower CH₄ abundance, allowing C₂H₅⁺ ions to reach the collecting grid. Considering the Xe ions' group, the increasing concentration of Xe in the mixture will lead to the formation of Xe₂⁺, which will replace Xe⁺. Finally, for Xe concentrations above 80% and up to pure Xe, only one peak is observed which, from the evolution observed, was attributed to Xe₂⁺, leading to higher diffusion.

Table 3. Ionization products, ionization cross sections for electron impact (20 eV) on Xe [34], appearance energy (A.E.) [35] and respective reaction rates.

Reaction	Cross Sec. (10 ⁻¹⁶ cm ²)	A.E. (eV)	Rate Const.	Ref.
e ⁻ + Xe → Xe ⁺ + 2e ⁻	2.43	12.13	—	[34, 35]
Xe ⁺ + Xe → Xe + Xe ⁺	—	—	2.5×10 ⁻¹⁰ cm ³ · s ⁻¹	[36]
Xe ⁺ + 2Xe → Xe ₂ ⁺ + Xe	—	—	2.0×10 ⁻³¹ cm ⁶ s ⁻¹	[37]

Another relevant feature that can also be observed in figure 1 is the variation of the FWHM which is seen to increase with Xe concentration, and that can be related to the higher Xe mass.

The evolution of the proportion of the peaks observed (from CH₄ and Xe ions) with the mixture composition can be explained analysing the total cross sections for electron impact ionization of Xe and CH₄ ions from table 3 and 1, it can be seen that, at electron energy of about 20 eV, the ionization probability for Xe is about 1.5 times higher for than for CH₄. It is thus expected that, even at lower Xe concentrations (down to 40% of Xe), Xe ions will still be preferentially produced.

Although energetically favoured, references to the charge transfer between CH_4^+ and Xe (1) were not found in literature. The only related reference found was to a charge transfer from a doublet state of Xe^+ to CH_4 (2) [42]. Nevertheless, the prevalence of the charge transfer reaction represented by (1) would reinforce the experimental observations made, corroborating the presence and abundance of Xe ions at low Xe concentrations.



In figure 2 we plot the mobility obtained for the ions produced in Xe- CH_4 mixtures as a function of Xe percentage, for 8 Torr and 20 Td at room temperature (293 K), together with Blanc's law prediction for Xe_2^+ (red dashed line), Xe^+ (grey dashed line) and CH_5^+ (green dashed line), C_2H_5^+ (orange dashed line) and C_3H_7^+ (blue 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 making use of the Langevin limit.

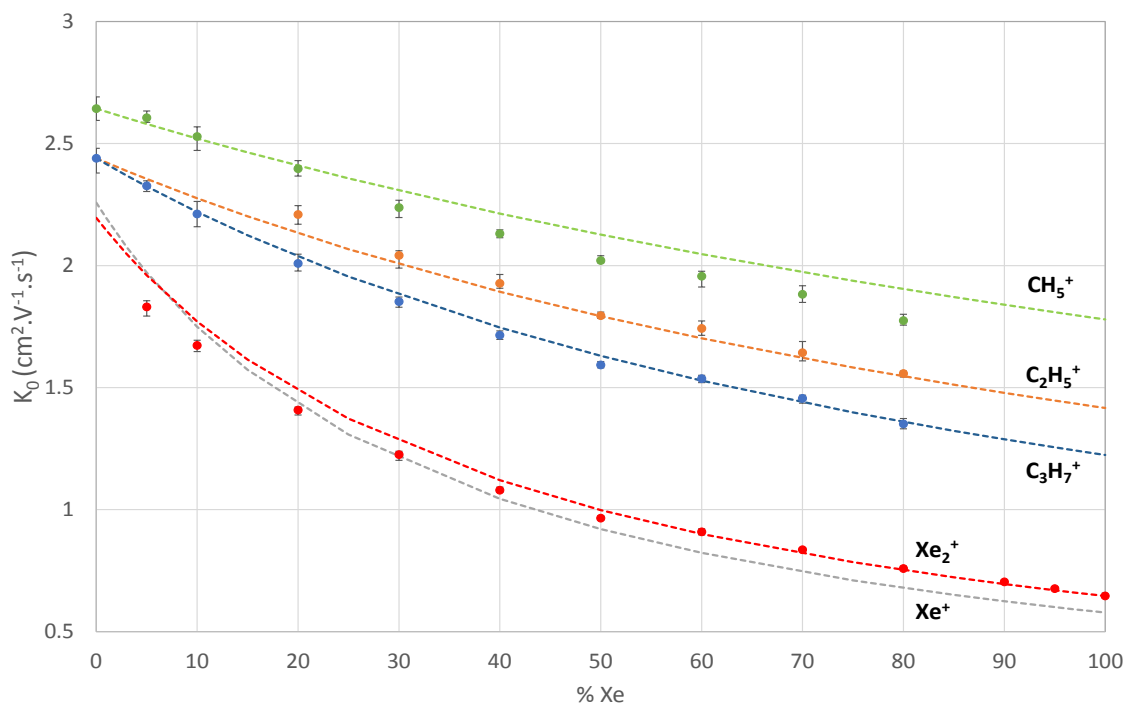


Figure 2. Reduced mobility of the ions produced in the Xe- CH_4 mixture for a pressure of 8 Torr and for a E/N of 20 Td at room temperature. The dotted lines represent the reduced mobility values expected from Blanc's law for Xe_2^+ (red), Xe^+ (grey) and for the ions originated by CH_4 - CH_5^+ (green), C_2H_5^+ (orange) and C_3H_7^+ (blue).

From 100% down to 20% Xe, in figure 2, the experimental values obtained for the several ions identified follow Blanc's law, with the exception of the one for CH_5^+ and Xe_2^+ which deviate from it. In the CH_5^+ case, this can indicate the increasing presence of CH_4^+ instead of CH_5^+ due to the longer reaction time for the formation of CH_5^+ for Xe percentages above 20%. Since CH_4^+ is likely to be affected by resonant charge transfer, this can explain the measured mobility values below the expected by Blanc's law.

As for Xe_2^+ , whose mobility is seen to be lower than the expected by Blanc's law, one possible explanation for this behaviour is the influence of its predecessor ion, Xe^+ , whose mobility is affected by resonant charge transfer, a phenomenon not accounted for in Blanc's law.

The ion mobility values measured were seen to vary with the relative abundance of the gases, but no significant variation of the mobility was observed in the range of E/N (10–25 Td) studied.

Table 4 summarizes the results obtained.

Table 4. Mobility of the ions observed for the Xe-CH₄ mixture ratios studied, obtained for E/N of 20 Td, at 8 Torr and 293 K.

Mixture	Mobility ($\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$)	Ion
5% Xe	2.61 ± 0.05	CH_5^+
	2.33 ± 0.02	C_3H_7^+
	1.83 ± 0.03	$\text{Xe}^+/\text{Xe}_2^+$
10% Xe	2.53 ± 0.05	CH_5^+
	2.21 ± 0.05	$\text{C}_2\text{H}_5^+/\text{C}_3\text{H}_7^+$
	1.67 ± 0.02	$\text{Xe}^+/\text{Xe}_2^+$
20% Xe	2.40 ± 0.03	CH_5^+
	2.21 ± 0.04	C_2H_5^+
	2.01 ± 0.03	C_3H_7^+
	1.41 ± 0.02	$\text{Xe}^+/\text{Xe}_2^+$
30% Xe	2.24 ± 0.04	CH_5^+
	2.04 ± 0.04	C_2H_5^+
	1.85 ± 0.02	C_3H_7^+
	1.23 ± 0.02	Xe_2^+
40% Xe	2.13 ± 0.02	CH_5^+
	1.93 ± 0.03	C_2H_5^+
	1.71 ± 0.02	C_3H_7^+
	1.08 ± 0.01	Xe_2^+
50% Xe	2.02 ± 0.02	CH_5^+
	1.79 ± 0.01	C_2H_5^+
	1.59 ± 0.01	C_3H_7^+
	0.97 ± 0.01	Xe_2^+
60% Xe	1.96 ± 0.03	CH_5^+
	1.74 ± 0.03	C_2H_5^+
	1.53 ± 0.01	C_3H_7^+
	0.91 ± 0.01	Xe_2^+
70% Xe	1.88 ± 0.03	CH_5^+
	1.64 ± 0.04	C_2H_5^+
	1.46 ± 0.02	C_3H_7^+
	0.84 ± 0.01	Xe_2^+
80% Xe	1.77 ± 0.02	CH_5^+
	1.56 ± 0.01	C_2H_5^+
	1.35 ± 0.02	C_3H_7^+
	0.76 ± 0.01	Xe_2^+
90% Xe	0.70 ± 0.01	Xe_2^+
95% Xe	0.68 ± 0.01	Xe_2^+

4 Conclusion

In the present work we measured the reduced mobility of ions originated by electron impact in Xe-CH₄ mixtures at pressures of 8 Torr, low reduced electric fields (10–25 Td) and different mixture ratios. The experimental results show that, for the range of concentrations studied, two different groups of ions were identified, one belonging to ions from CH₄ (CH₅⁺, C₂H₅⁺ and C₃H₇⁺) and the other to Xe ions (Xe⁺ and Xe₂⁺). The presence, abundance and mobility of the different ions present was seen to vary for the range of mixtures studied. Increasing CH₄ concentration in the mixture resulted in a higher mobility of the ions observed, with the behaviour roughly following Blanc's law through the entire range for the different ions proposed.

The mobilities measured did not display a significant dependence with the E/N for the range covered in this work (10–25 Td).

It is our intention to extend the work on ion mobility using different mixtures of known interest (for the applications already referred) such as Ar-CF₄, Ar-CF₄-Isobutane, Ne-CF₄ and Xe-CF₄.

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