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Experimental ion mobility measurements in Ne-CF₄

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ABSTRACT: In this paper we present the results of the ion mobility measurements made in gaseous mixtures of neon with carbon tetrafluoride (Ne-CF₄) at a total pressure 8 Torr (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 only one peak, which was attributed to CF_3^+ . The reduced mobilities obtained from the peak centroid of the time-of-arrival spectra are presented for Ne concentrations in the 5%–95% range.

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

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1 Introduction

The mobility of charged particles in a gaseous media, K, defined as the ratio between the particle drift velocity, v_d , and the applied electric field, E:

$$K = \frac{v_{\rm d}}{E} \tag{1.1}$$

is an important parameter in gaseous detectors, since the detector response depends on its value, specially on the electron mobility. Nevertheless, in cases where the production of electron-ion pairs is large, i.e., when charge multiplication is the amplification mechanism, the detection rate can be limited space charge effects that result from the accumulation of charge and consequent field distortion. In these cases, the knowledge of the ion mobility can help to infer the rate detection limit [1]. Examples of this kind of detectors are micropattern gas detectors (MPGD), Transition Radiation Detectors (TRD) and Time Projection Chambers (TPC) [1–5]. Noble gases, like Xe, Ar and Ne are usually the preferred filling gases of detectors, but it is often necessary to add a molecular gas in low percentages to avoid some undesired effects, like the large electron diffusion, spurious pulses due to scintillation, among others [1, 6, 7]. The choice of the additive depends on the detector kind and its purpose, with CF_4 being considered in some cases such as in the NEXT [8] and sPHENIX [9] experiments. Still, the ion mobility in these mixtures is scarce or non-existing.

In this work we present the results for the ion mobility in mixtures of Ne-CF₄, which are being considered for the sPHENIX TPC [9]. The measurements were performed in an existing experimental system [10] for a total gas pressure of 8 Torr and for reduced electric fields, E/N, in the 10-25 Td range, with N the gas number density. For this range of E/N, the energy gained by the ions between collisions with the gas atoms/molecules is lower than the thermal energy and so v_d is proportional to the electric field, with K becoming constant, i.e., independent of E/N. It is usual to present the results of ion mobility in terms of the reduced mobility, K_0 , defined as:

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

where *N* is the gas number density and N_0 is the Loschmidt number ($N_0 = 2.68678 \times 10^{19} \text{ cm}^{-3}$ for 273.15 K and 101.325 kPa according to NIST [11]). 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·m²) and the reduced mobility obtained from equation (1.2) is expressed in units of cm²·V⁻¹·s⁻¹.

Since our experimental system does not allow a direct identification of the ions, to identify which ions are drifting and reach the collecting grid, the possible ion formation cross-sections and the reactions between the primary ions and the gas atoms/molecules are searched for in the literature. Also, a comparison between our results and the predicted ones, using both the Langevin theory [12] and Blanc's law [13] is made. The reduced mobility predicted by the Langevin theory, which assumes that the neutral polarization is the main responsible for the K_0 value at low electric field and low temperature, is given by:

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

where α is the neutral polarisability in cubic angstroms ($\alpha = 0.394 \text{ Å}^3$ for Ne [14] and $\alpha = 3.86 \text{ Å}^3$ for CF₄ [15]) and μ is the ion-neutral reduced mass in unified atomic mass units.

Although Langevin theory is usually still valid at room temperature [16], it has some limitations, namely in the drift of large molecules or when charge transfer mechanisms are involved during the ion's drift [17]. Concerning Blanc's law, it predicts the mobility of ions in binary gases, K_{mix} , given by:

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

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; f_1 and f_2 are the molar fraction of each gas in the binary mixture [13].

2 Method and experimental setup

The experimental setup has already been used and described in previous papers [10, 17–21]. The ion's velocity is obtained by measuring, with a digital oscilloscope (Tektronix TDS 2022B), the time that the ions take to drift under a uniform electric field, from a GEM [22] where they are originated to a grid located 4.273 cm above it. A flash from the UV lamp hits a CsI photocathode, that is deposited in the top GEM surface, originating a number of photoelectrons that are guided to the GEM holes due to the potential voltage difference applied between the GEM surfaces. In their drift through the GEM holes, these electrons originate ions through collision with the gas atoms/molecules, and these ions start their drift, under the influence of a uniform electric filed, until they reach a Frisch grid, 0.5 mm below the collecting grid. Between the Frisch grid and the collecting grid a high electric field is applied and this charge signal is recorded in the oscilloscope, originating a time spectrum, with the flash from the UV lamp being the time trigger for the ion formation. An average of 128 spectra is made and the background noise is subtracted for every time spectrum obtained. The peaks' centroid in the spectra correspond to the time the ions took to drift through a known distance (4.273 cm) and from these, the drift velocity and the ion mobility can be obtained. As already mentioned, since no direct ion identification is possible, the knowledge of the primary ions originated and the possible reactions between these ions and the gas molecules and their reaction rates, together with the Langevin limit for the ion mobility and Blanc's Law, help in the identification of the ions responsible for the different peaks in the obtained time spectra.

Since impurities can alter the ions' mobility, before each experiment the vessel was vacuum pumped down to pressures of 10^{-6} to 10^{-7} Torr. No measurement was considered until the signal in the oscilloscope stabilised, and, after that, all measurements were done in a 2-3 minutes time interval to ensure minimal contamination of the gas mixture, mainly due to outgassing processes.

Although the measurements are performed at low pressure (up to a few tens of Torr) the results have been consistent with data obtained at higher pressures [17, 23, 24]. The exceptions are if the reaction times (pressure dependent) involved in the formation of the ions observed are of the order of magnitude of the drift time; in these cases the ion mobility measured will be affected by the pressure.

3 Results and discussion

The mobility of the ions originated in Ne-CF₄ 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 E/N values considered in this work is within the required conditions of low reduced field, i.e. E/N < 30 Td for the working pressures used. A background work on the mobilities and ionization processes of Ne [18, 20] and CF₄ [21] in their parent gases has already been performed in our group.

3.1 Neon-Carbon Tetrafluoride (Ne-CF₄) mixture

In neon-carbon tetrafluoride (Ne-CF₄), only one peak can be observed for the mixture range studied as can be seen in figure 1, where the drift spectra for several Ne-CF₄ percentages (20%, 50%, 80% and 90% of Ne) are displayed, at 8 Torr, 293 K and 15 Td with a V_{GEM} of 25 V.

In figure 1 there are two striking features both of which occur with increasing Ne concentration in the mixture: one is the increase in the mobility of the ions that results in a shift in the peaks towards lower drift times and the other is a decrease in their abundance which can be seen from the decrease in the peak area, more pronounced for Ne concentrations above 70%. The higher mobility with increasing Ne concentration, can be explained by the lower Ne mass when compared to that of the CF₄ molecule, which implies a higher probability of lower reduced mass ion-neutral collisions (μ) which leads to a higher mobility (see eq. (1.3)).

The most probable candidate ion for the peak observed is CF_3^+ . Since the electron impact ionization cross section at 25 eV in CF_4 [25] is about forty times greater than the cross section in Ne [26], it is expected that CF_4 ions (mainly CF_3^+ as seen in [21]) will be produced more abundantly than Ne ions down to CF_4 concentrations of about 2.5% CF_4 . Additionally, the appearance energies for CF_4 ions, in particular CF_3^+ [27], are much lower than those for Ne ions [28]. As a result a charge transfer from Ne⁺ to CF_4 is also expected, and the Ne⁺ ions produced will rapidly react with CF_4 molecules [29], which prevents them from appearing in the time-of-arrival spectra. These possible reactions are shown in table 1. In this table it can also be seen that the competing three-body reaction that would lead to the formation of Ne₂⁺ ($k = 5.6 \times 10^{-32} \text{ cm}^6 \cdot \text{s}^{-1}$) is less probable (and much slower) than the charge transfer reactions. As a result, Ne ions will also lead to the formation of the same ion as in pure CF_4 [21], namely CF_3^+ .

Figure 2 shows the inverse of the reduced mobility obtained experimentally in the different Ne-CF₄ mixtures at 8 Torr and for E/N of 15 Td and 20 Td at room temperature, together with Blanc's law prediction for the main candidate ions — CF₃⁺ (green), Ne⁺ (orange) and Ne₂⁺ (blue).



Figure 1. Time-of-arrival spectra of an average of 128 pulses recorded for several Ne-CF₄ mixtures (20%, 50%, 80% and 90% of Ne) 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).

Table 1.	Reactions involving	Ne atomic ions and its ioniza	tion products with CF ₄ , adapted from	om [<mark>29</mark>].	
	Reaction	Prod. Dist.	Rate Const	t. $(10^{-10} \mathrm{cm}^3 \cdot \mathrm{s}^{-1})$	

reaction	1100. 0100.	
$Ne^+ + CF_4 \rightarrow CF_3^+ + F + Ne$	0.955	
$Ne^+ + CF_4 \rightarrow CF_2^+ + 2F + Ne$	0.045	1.34 ± 0.40

 K_{g1} and K_{g2} in Blanc's law (equation (1.4)), were obtained either using experimental values from literature [30, 31] in the case of Ne⁺ and Ne⁺₂ in Ne and [21] in the case of CF⁺₃ in CF₄ or, when not existing, by using the Langevin limit formula (equation (1.3)). Ions moving in CF₄ have already been seen to follow fairly well the values obtained from the Langevin polarization limit [21]. For this reason the mobility value of Ne⁺ and Ne⁺₂ ion in CF₄ used in this work are the ones obtained from the Langevin limit, 1.74 and $1.34 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, respectively. As mentioned before, Langevin theory has some limitations, namely in Ne, yielding mobilities lower than the experimental values. A correction was introduced in terms of the Ne polarizability in order to provide a more adequate fitting to the experimental data, as seen in [17]. The values were obtained using the ion mass-mobility data for Ne in [17] and the mass of CF₃⁺, yielding a mobility value of 6.64 cm² · V⁻¹ · s⁻¹.



Figure 2. Reduced mobility of the ions produced in the Ne-CF₄ mixture for a pressure of 8 Torr and for a E/N of 15 Td (black) and 20 Td (red) at room temperature. The dashed lines represent the mobility values expected from Blanc's law for CF₃⁺ (green), Ne⁺ (orange) and Ne₂⁺ (blue).

Figure 2 shows that the experimental values closely follow Blanc's law curve for CF_3^+ , confirming the initial assumption that this is the ion responsible for the signal observed.

In table 2 the results obtained for the ion mobilities of CF_3^+ in Ne-CF₄ mixtures are summarized. No significant variation of the mobility was observed in the range of E/N values studied, as can be seen in figure 2.

Ne-CF ₄ Mixture	Mobility
(% of Ne)	$(\mathrm{cm}^2 \cdot \mathrm{V}^{-1} \cdot \mathrm{s}^{-1})$
5%	1.15 ± 0.01
10%	1.19 ± 0.02
15%	1.27 ± 0.01
20%	1.30 ± 0.01
30%	1.45 ± 0.01
40%	1.66 ± 0.01
50%	1.77 ± 0.01
60%	2.06 ± 0.02
70%	2.53 ± 0.02
80%	3.15 ± 0.03
85%	3.63 ± 0.02
90%	4.26 ± 0.02
95%	5.12 ± 0.04

Table 2. Mobility of the ions observed for the Ne-CF₄ mixture ratios studied, obtained for E/N of 15 Td, at 8 Torr and 293 K.

4 Conclusion

In the present work we measured the reduced mobility of ions originated by electron impact in Ne-CF₄ mixtures at a total pressure of 8 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 throughout the range of mixtures studied. This peak is due to CF_3^+ ions, with our experimental results showing consistency with those expected from Blanc's law throughout the entire range of mixtures (from 5 to 95% Ne).

Increasing Ne concentration in the mixture resulted in a higher mobility which is explained by the ion/neutral lower reduced mass, while not depending significantly on E/N.

At higher pressures (close to the atmospheric one) the formation of clusters is favoured and so it is expected that the ions formed will be $CF_3^+(CF_4)$ as discussed in [21], with the theoretical values obtained from Langevin's polarization limit being close to the experimental data obtained at atmospheric pressure in pure CF_4 [32, 33]. Theoretical estimates (using Blanc's law and Langevin's polarization limit formula) can be used for the mobility of the cluster ion at atmospheric pressure, which should range from 0.94 to $6.2 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, depending on the mixing ratio from 0 to 100% Ne, respectively.

Future work is expected with other gaseous mixtures. It is our intention to proceed this line of investigation using different binary mixtures of Ne, CF_4 , CH_4 , C_2H_6 and iC_4H_{10} which are of interest for low energy X-rays detection.

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