Experimental ion mobility measurements in Ne-N$_2$

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Abstract: Data on ion mobility is important to improve the performance of large volume gaseous detectors, such as the ALICE TPC or in the NEXT experiment. In the present work the method, experimental setup and results for the ion mobility measurements in Ne-N$_2$ mixtures are presented. The results for this mixture show the presence of two peaks for different gas ratios of Ne-N$_2$, low reduced electric fields, $E/N$, 10–20 Td (2.4–4.8 kV·cm$^{-1}$·bar$^{-1}$), low pressures 6–8 Torr (8–10.6 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, 2], and also in IMS (Ion Mobility Spectrometry) a technique used for the detection of narcotics and explosives [3]. In order to fully understand and model these detectors it is important to have detailed information on the transport properties of ions.

Using the experimental method described in detail in [4], the mobility of ions in neon-nitrogen (Ne-N\textsubscript{2}) gas mixtures is 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\textsuperscript{-1}·bar\textsuperscript{-1}), extending previous studies developed in our group for other gases [4–13].

1.1 Ion mobility

Under a weak and uniform electric field a group of ions will eventually reach a steady state characterized by a drift velocity [3], \(v_d\), expressed by:

\[
v_d = KE
\]  

(1.1)

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

\[
K_0 = KN/N_0
\]  

(1.2)

where \(N\) is the gas number density and \(N_0\) is the Loschmidt number (\(N_0 = 2.6867 \times 10^{19} \text{ cm}^{-3}\)).

The mobility values can be presented as a function of the reduced electric field \(E/N\) in units of Townsend (1 Td=10\textsuperscript{-17} V·cm\textsuperscript{2}).
1.2 Langevin’s theory

According to Langevin’s theory [14], one limiting value of the mobility is reached when the repulsion becomes negligible compared to the polarization effect. This limit is given by the following equation,

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

where \( \alpha \) is the neutral polarisability in cubic angstroms (\( \alpha = 0.394 \, \text{Å}^3 \) for Ne [15] and \( \alpha = 1.74 \, \text{Å}^3 \) for N\(_2\) [15]) and \( \mu \) is the ion-neutral reduced mass in atomic mass units. The Langevin limit is the value of \( K \) in the double limit of low \( E/N \) and low temperature, conditions which ensure the dominance of the polarization attraction over other atomic interactions (e.g. hard sphere repulsion), describing well our experimental conditions: low pressure, low temperature and low reduced electric fields.

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, \( K_{\text{mix}} \), can be expressed as follows:

\[ \frac{1}{K_{\text{mix}}} = f_1 \frac{1}{K_{g1}} + f_2 \frac{1}{K_{g2}} \]  (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 [16].

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 GEM that is inside a gas vessel. The photoelectrons released from the CsI film trigger an electron avalanche inside the GEM holes, where they 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 acts as Frisch grid while the second, 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 is subtracted from the signal, gaussian curves are fitted to the time of arrival spectra from which the peak centroids are obtained. Since the peaks’ centroid corresponds to the average drift time of the ions along a known distance, the drift velocity and mobility can then be calculated. One important feature of the method is the capability of controlling the voltage across the GEM (\( V_{\text{GEM}} \)), and so the energy gained by the photoelectrons as they move across the GEM holes. This characteristic proves to be a great advantage since it enables the identification of the primary ions based on their ionization energies. 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.

3 Results and discussion

The mobility of the ions originated in Ne-N\textsubscript{2} mixtures have been measured for different reduced
electric fields $E/N$ (from 10 Td up to 20 Td) and different pressures (in the 6–8 Torr pressure range)
at room temperature (298 K).

The range of the reduced electric field values used to determine the ions’ mobility is limited
due to two distinct reasons: 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\textsuperscript{-1} bar\textsuperscript{-1}), which has been attributed to collisions between the ions and impurity
molecules.

A background work on the mobilities and ionization processes of Ne [5] and N\textsubscript{2} [6] in their
parent gases has already been performed in our group.

3.1 Ne-N\textsubscript{2} mixture

In neon-nitrogen (Ne-N\textsubscript{2}) mixtures with N\textsubscript{2} concentrations higher than 10% only one peak is
observed, as can be seen in figure 1. The ion responsible for this peak is the same ion as in pure
N\textsubscript{2} according to the cross sections and rate constants displayed in table 1, i.e. N\textsuperscript{+}\textsubscript{4}. Since the total
ionization cross section for electron impact (at an energy of 23 eV) in Ne is 0.0166±0.001×10\textsuperscript{-16}
cm\textsuperscript{2} [17] — about 18 times lower than that for N\textsubscript{2} (0.492±0.025×10\textsuperscript{-16} cm\textsuperscript{2} [18])), it is expected
that even at low N\textsubscript{2} concentrations (down to about 5% of N\textsubscript{2}), N\textsubscript{2} ions are still the ones preferentially
produced. For N\textsubscript{2} concentrations below 10% in the mixture another peak becomes visible as can be
observed in figure 1. In table 1 the possible reactions are summarized together with the respective
ionization cross sections or rate constants.

Figure 2 shows the evolution of the fraction of ion species present as a function of time for N\textsubscript{2}
concentration of 10% (figure 2a), and 50% (figure 2b), for a total pressure of 8 Torr. The relative
abundance of the different ion species was calculated using both electron impact ionization cross
sections and the reaction rates summarized in table 1.

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. A careful analysis of figure 2 can help to explain figure 1, where
the time-of-arrival spectra for several Ne-N\textsubscript{2} mixtures (5%, 10%, 50% and 90% of N\textsubscript{2}) at a pressure
of 8 Torr, temperature of 298 K and for a reduced electric field of 15 Td with a voltage across GEM
of 23 V, are displayed. As can be inferred from table 1, at very low N\textsubscript{2} concentrations (up to about
5% of N\textsubscript{2}), the production of Ne\textsuperscript{+} ions will be more abundant, leading to the same ions as in pure
Ne, while above this and up to 10% of N\textsubscript{2}, both Ne and N\textsuperscript{+}\textsubscript{4} ions will be produced.

Observing figure 2a, at the instant corresponding to the detected ions’ drift time taken from
the peaks’s centroid of figure 1 (10% N\textsubscript{2}), about 0.15 ms, it is possible to conclude that the ions
Figure 1. Time-of-arrival spectra averaged over 128 pulses for several Ne-N$_2$ mixtures (5%, 10%, 50% and 90% of N$_2$) at a pressure of 8 Torr, temperature of 298K and for a reduced electric field of 15Td with a voltage across GEM of 23 V (background noise subtracted).

Table 1. Summary of possible reactions and respective rate constants or cross section for electron impact ionization of the different primary ions for 23 eV.

| Reaction                        | Rate Constant | Cross section (23 V) | Ref. |
|---------------------------------|---------------|----------------------|------|
| $e^{-} + Ne \rightarrow Ne^{+} + 2e^{-}$ | –             | $0.0166\pm0.001\times10^{-16}$ cm$^2$ | [17] |
| $Ne^{+} + Ne \rightarrow Ne + Ne^{+}$ | $3.4\pm0.85\times10^{-10}$ cm$^3$.s$^{-1}$ | – | [19] |
| $Ne^{+} + 2Ne \rightarrow Ne^{+}_{2} + Ne$ | $4.4\pm0.4\times10^{-32}$ cm$^6$.s$^{-1}$ | – | [20] |
| $e^{-} + N_{2} \rightarrow N_{2}^{+} + 2e^{-}$ | –             | $0.492\pm0.025\times10^{-16}$ cm$^2$ | [18] |
| $N_{2}^{+} + N_{2} + M \rightarrow N_{4}^{+} + M$, $(M = N_{2})$ | $5.5\pm0.3\times10^{-29}$ cm$^6$.s$^{-1}$ | – | [21] |
| $N_{2}^{+} + N_{2} + M \rightarrow N_{4}^{+} + M$, $(M = Ne)$ | $1.4\times10^{-29}$ cm$^6$.s$^{-1}$ | – | [21] |
| $Ne^{+} + N_{2} \rightarrow N_{2}^{+} + Ne$ | $1.1\pm0.44\times10^{-13}$ cm$^3$.s$^{-1}$ | – | [19] |

responsible for the peaks observed will be Ne ions ($Ne^{+}/Ne^{+}_{2}$)$^1$ for the peak with higher mobility and the N$_4^+$ ions for the peak with lower mobility. In this case the expected fraction of each species collected is about 83% of N$_4^+$, 8.9% of Ne$^+$ and 6.8% of Ne$^+_2$ and 1.3% of N$_2^+$, whereas the ions formed at the GEM are 78% N$_2^+$ and 22% Ne$^+$. Further increasing the concentration of N$_2$ in the mixture will lead mainly to the formation of N$_4^+$. Looking at figure 2b (50% N$_2$), at about 0.28 ms, the drift time of the ion responsible for the spectrum in figure 1 (50% N$_2$), the expected fraction of each ion species is 99.8% of N$_4^+$, with the remaining representing only 0.2%, while the ions formed at the GEM are 97% N$_2^+$ and 3% Ne$^+$.

$^1$As can be see from figure 2a the conversion of Ne$^+$ to Ne$^+_2$ is incomplete for this mixture, so the mobility measured is in fact the result of the drift path travelled both as Ne$^+$ and Ne$^+_2$.
Figure 2. Fraction of ions that can be formed as a function of time for Ne-N$_2$ mixtures of 10% (a) and 50% (b) of N$_2$, for a total pressure of 8 torr.

In fact, above 10% N$_2$ the only ion expected is N$_4^+$ which results from N$_2^+$ through a three-body reaction:

$$N_2^+ + N_2 + M \rightarrow N_4^+ + M \quad (3.1)$$

where M is an atom or molecule from the gas mixture, in our case Ne or N$_2$. In this collision the excess energy is removed by a third body (M), preventing its dissociation back to N$_2^+$. As can be seen from table 1 the reaction time will depend on M. As a consequence the reaction time for the formation of N$_4^+$ will be affected by the chosen reaction partner (M), which can affect the signal shape if the drift time is of the same order of magnitude of the reaction time. A longer reaction time means that part of the drift path is spent as N$_2^+$, which has lower mobility than N$_4^+$, originating an asymmetry towards the right side of the N$_4^+$ peak, as can be observed in the drift spectra displayed in figure 1.

As for the N$_2^+$ ion, it can be originated either from direct electron impact ionization of N$_2$ or from the charge transfer reaction,

$$Ne^+ + N_2 \rightarrow N_2^+ + Ne \quad (3.2)$$

that has a lower reaction time than the competing one,

$$Ne^+ + 2Ne \rightarrow Ne_2^+ + Ne \quad (3.3)$$

in the pressure conditions of this experiment (as can be seen in figure 2b for 50% of N$_2$). Since to our knowledge there is no charge transfer between Ne$_2^+$ and N$_2$ we expect that, once formed, it will remain unaltered through the drift distance. Also, since the dissociation energy of N$_4^+$ (0.87 eV) is much larger than the kinetic energy of the ion under low reduced electric field, once N$_4^+$ is formed it will have a low probability of dissociating back into N$_2^+$ and N$_2$ [6].

Concerning the peak area, we can observe in figure 1 that it varies with N$_2$ concentration in the mixture, a feature somehow related to the availability of Ne. There is also a peak shift to lower drift times with decreasing N$_2$ concentration, which translates into an increase in the ion mobility.
This increase is due to the fact that the Ne atom has a much smaller mass than \( N_4^+ \) ion implying a much lower energy loss in elastic collisions with the gas atoms/molecules.

As mentioned, Blanc’s law can be used to predict the mobility of the ions in gaseous mixtures. In figure 3 we plot the inverse of the reduced mobility obtained for the ions produced in the Ne-N\(_2\) mixture as a function of the different mixture ratios studied for a pressure of 8 Torr and for \( E/N \) of 15 Td, at room temperature (298 K).

![Figure 3. Inverse of the reduced mobility of the ions produced in the Ne-N\(_2\) 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 \( N_4^+ \) (orange) and for \( N_2^+ \) (blue).](image)

Dashed lines representing Blanc’s law for the most abundant ion, \( N_4^+ \) (orange), as well as for \( N_2^+ \) (blue) are also displayed. In this case, in Blanc’s law (eq. (1.4)), \( K_{g1} \) and \( K_{g2} \) were obtained either by using Langevin’s formula (eq. (1.3)) for \( K_{(N_4^+/Ne)} \) or by selecting experimental values from literature.

Observing figure 3 it is possible to conclude that the ion mobility experimentally obtained roughly follows, within error bars, Blanc’s law for the most abundant ion down to 40% \( N_2 \), while below this \( N_2 \) concentration it deviates towards the \( N_2^+ \) theoretical value given by Blanc’s law. The same figure indicates that the ion species observed depends on the amount of \( N_2 \) in the mixture (a second peak appears as seen in figure 1 for 5% and 10% \( N_2 \) and also in figure 3).

In fact, starting from pure Ne, as a result of the reaction rates and pressures, we have initially the formation of \( Ne^+ \) and \( Ne_2^+ \), then \( Ne^+ \) and \( N_4^+ \) and finally only \( N_4^+ \) according to the different reaction channels discussed previously. In addition it is also possible to see that increasing \( N_2 \) leads to a significant decrease in the mobility of the \( N_2 \) ions present, i.e. \( N_4^+ \).

The mobility values in this experiment range from \( 6.41 \pm 0.06 \) to \( 2.37 \pm 0.03 \) cm\(^2\)V\(^{-1}\)s\(^{-1}\) for the slowest ion, and from \( 7.20 \pm 0.06 \) to \( 6.55 \pm 0.05 \) cm\(^2\)V\(^{-1}\)s\(^{-1}\) for the fastest ion, 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 \( E/N \) (10–20 Td) studied.
The mobility values of the ions observed for the Ne-N\textsubscript{2} mixture ratios studied, at $E/N$ of 15 Td, pressure of 8 Torr and at room temperature (298 K) are summarized in table 2.

**Table 2.** Mobility of the ions observed for the Ne-N\textsubscript{2} mixture ratios studied, obtained for $E/N$ of 15 Td, at 8 Torr and 298 K.

| Ne-N\textsubscript{2} Mixture | % of N\textsubscript{2} | Mobility (cm\textsuperscript{2}V\textsuperscript{−1}s\textsuperscript{−1}) | Mobility (cm\textsuperscript{2}V\textsuperscript{−1}s\textsuperscript{−1}) |
|-------------------------------|-----------------|------------------|------------------|
|                               |                 | $N^+_4$          | Ne\textsuperscript{+}/Ne\textsuperscript{2}_2 |
| 5%                            |                 | 6.41 ± 0.06      | 7.20 ± 0.06      |
| 10%                           |                 | 5.86 ± 0.05      | 6.55 ± 0.05      |
| 15%                           |                 | 5.30 ± 0.05      | –                |
| 25%                           |                 | 4.61 ± 0.06      | –                |
| 40%                           |                 | 3.80 ± 0.05      | –                |
| 50%                           |                 | 3.46 ± 0.08      | –                |
| 60%                           |                 | 3.19 ± 0.06      | –                |
| 75%                           |                 | 2.78 ± 0.04      | –                |
| 90%                           |                 | 2.47 ± 0.03      | –                |
| 95%                           |                 | 2.37 ± 0.03      | –                |

From 0 to 100% N\textsubscript{2}, the peaks observed were seen to vary in position and area, demonstrating that the ion or ions formed and their mobilities depend on the ratio of the two gases used.

**4 Conclusion**

In the present work we measured the reduced mobility of ions originated by electron impact in the Ne-N\textsubscript{2} mixtures using pressures from 6 to 8 Torr, low reduced electric fields (10–20Td) and different mixture ratios. The experimental results show that for the mentioned mixtures two peaks were observed in the range of concentrations studied. The ions responsible for these peaks are believed to be the ones originated in Ne (Ne\textsuperscript{+} and Ne\textsuperscript{2}+) for concentrations of N\textsubscript{2} up to 5% while for concentrations up to 10% of N\textsubscript{2}, the ions observed are Ne\textsuperscript{+} and N\textsuperscript{+}4. Above 10% N\textsubscript{2} the only ion observed is N\textsuperscript{+}4, which can be formed through different reaction channels. The ions’ mobility was seen to decrease with the increase of N\textsubscript{2} concentration in the mixture with the behaviour following roughly, Blanc’s law for N\textsuperscript{+}4 down to 40% N\textsubscript{2} and then gradually changing towards the N\textsuperscript{+}2 predicted behaviour. An asymmetry was observed in some spectra, this asymmetry is expected to be due to the different reaction paths discussed and that originate N\textsuperscript{+}4.

Additionally we verified that the mobilities calculated did not display a significant dependence either with pressure in the pressure range studied (6–8 Torr), or with $E/N$ in the range used in this work (10–20 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-CO\textsubscript{2}, Xe-CH\textsubscript{4}, Ar-CF\textsubscript{4}, Ne-CF\textsubscript{4} and Xe-CF\textsubscript{4}.
Acknowledgments

This work was supported by the RD51 Collaboration/CERN, through the common project “Measurement and calculation of ion mobility of some gas mixtures of interest”. André F.V. Cortez received a PhD scholarship from FCT-Fundação para a Ciência e Tecnologia (SFRH/BD/52333/2013). Pedro M.C.C. Encarnação was supported by FEDER, through the Programa Operacional Factores de Competitividade — COMPETE and by National funds through FCT-Fundação para a Ciência e Tecnologia in the frame of the Project QREN n.4825, Rad for Life.

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