Mobility of non thermal electrons and ions in very high density and purified nitrogen corona discharge

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Abstract. The mobility of nonthermal electrons and ions have been determined in very high and purified nitrogen corona discharge. Corona discharge was generated by plasma generator system with point-to-plane electrodes geometry configuration. Characteristic I-V has been done by using stabilized DC high voltage generator high up to 20 kV. Graph current of saturation corona unipolar for variation voltage and the distance between electrodes is curved with the semi-parabolic equation. We used I-V characteristic to determine charge particles mobilities. In this research high purified and slightly purified nitrogen gas. For negative corona, we found that the mobility of charged particles is greater than negative ions and smaller than thermal electron mobility. Particles engaging and moving in the corona with high purification the gas are non-thermal electrons, positive ions for positive corona. From the current-voltage characteristic, we can conclude that the regime of our discharge in very pure gases (N2) corresponds to the current regime limited by space charge. By analogy with what has been observed in a "discharge tube" this regime is similar to an abnormal glow discharge regime. We have seen previously that the variation with the density of the ionic mobility is in good agreement with the direct measurements by the time of flight method.

1. Introduction
Corona discharge is one of the techniques for generating cold plasma. Cold plasma at atmospheric pressure is very much utilized primarily in the printing industry, drying on painting, drying on food products. The basic research of corona discharges have been known since the pioneering work of Warburg, Townsend, Thompson [1], who laid the basis of the many subsequent experimental investigations and theories. Corona discharge is a low energy electrical discharge with non-thermal ionisation that takes place in the vicinity of an electrode of sufficiently low radius of curvature, in a gas at a pressure close to the atmospheric [2]. Corona discharges at atmospheric pressure are used to control environmental contamination by emission gas and fly as [3] can be used as an ion-driven wind generator [4]. In addition to corona at atmospheric pressure, there are also studies of corona at high pressures and liquids [6]. Electrical and spectroscopic diagnostics of corona discharge in nitrogen and argon very high pressure gasses, with pressure from 1 MPa to 100 MPa was carried out [7]. In this research the gas used to generate corona discharges has undergone a very high purification. The
reactor for the experiment can be filled with gas up to 100 bars. the novelty of this research is the level of nitrogen gas was perfectly purified and the high-density nitrogen molecules.

2. Methods
The electrical measurements (current, pause time between pulses) were made by a device developed in the laboratory (Figure 1). The high voltage was provided by a $0 \pm 20 \text{kV}$ stabilized DC voltage generator (Spellman RHSR/20PN(60) connected to the tip while the planar electrode was grounded. The intensity of the average current was measured either with a galvanometer (Sefram Vérispot) or a Keithley electrometer (model 610C). The current pulses were visualized using a preamplifier assembly followed by a Tektronix oscilloscope (Model 7633). In the pulsed current domain, pulses of light emitted by a photomultiplier (model 56AVP DARIO), whose spectral response was between 300 and 650 nm (maximum sensitivity at 400 nm) and the pulses, are simultaneously detected. current.

![Figure 1](image-url)  
**Figure 1.** Experimental set up

3. Results and Discussion
In negative polarity for the purified nitrogen, the current increases from a value of some tens of microamperes for voltage ($V$) equal threshold voltage ($V_s$) to reach the saturation zone and then the arc zone. In the perfectly purified nitrogen (the same gas passing 20 times in the purification circuit before the measurement), the current-voltage characteristic is slightly different. The current for $V = V_s$ goes directly from a non-detectable value to more than 100 $\mu$A. The higher the density of the gas, the higher the threshold voltage ($V_s$), and the higher the current at $V = V_s$. This characteristic is shown in Figure 2. We did not observe a transition to the arc and therefore saturation current was not detected although significant current values (up to 1 mA and sometimes 1.5 mA) were measured.
Figure 2. I-V curves for nitrogen perfectly purified and slightly purified in negative corona.

The curves in Figure 2 present a comparison of current-voltage characteristics in negative polarity, for a pressure of 0.5 MPa, between a perfectly purified gas and a slightly purified gas. In the little purified nitrogen, the current-voltage characteristic I (V) can be decomposed into different regions that correspond to different operating regimes. We try to explain this variation by analogy with what has been observed in a "discharge tube" [8,9].
- AB can be likened to the Townsend discharge and priming regimes;
- BC can be considered as the subnormal and normal regimes of glow discharges;
- CD can be considered as the abnormal regime of the glow discharge;
- D is the point of transition to the electric arc.

In highly purified nitrogen, we see in Figure 2 that the discharge regime is directly in the abnormal regime of the glow discharge (C'D'). In this regime the current is a direct current and the discharge is almost stationary. It should be noted that all analyzes in this study were made in this scheme.

Figure 3. I-V curves for nitrogen at different pressures in negative corona.
Figure 4 shows the evolution of the I/V versus V ratio in nitrogen gas at different pressures. We note that Vo is practically independent of pressure. For all the pressures studied, we have Vo \( \approx \) 1700 volts. However, for the low-pressure gas (for example 0.2 MPa), the threshold voltage V\text{threshold} \( \approx \) 2000 volts, which is close to Vo, but for the gas at high pressure (for example 5 MPa) V\text{threshold} \( \approx \) 7800 volt, the threshold voltage is then about 5 times higher than Vo. Although Vo is not zero, we consider that the discharge regime is close to a current regime limited by space charge.

![Figure 4. I/V versus V curves for nitrogen at different pressures, negative polarity.](image)

### 3.1 Non-thermal electronic mobility

The "apparent" mobility of charge carriers in negative polarity was calculated from the slope of the straight lines I (V) (in all tests the curves I (V) are straight lines), following the Sigmond model. In Figure 5 we can observe the behavior of the apparent mobility as a function of the density of the gas. This mobility decreases and follows, as a first approximation, a decrease in \( N^{-1} \).

The work of Wada et al [Wad81] gives, for the mobility of thermal electrons (with \( E/N \approx 10^{-19} \text{ Vcm}^2 \)), values 25 times higher than ours in a density range between \( 10^{20} \text{ cm}^{-3} \) and \( 10^{21} \text{ cm}^{-3} \). For higher densities, the variation of the mobility of the thermal electrons no longer follows an \( N^{-1} \) law. This is a "negative effect" of density on mobility. On the other hand, our results always follow a variation in \( N^{-1} \) and that at least up to \( N = 2.4 \times 10^{21} \text{ cm}^{-3} \).

However, in our tests, the reduced E/N field in the drift region varies between \( 0.5 \times 10^{-17} \text{ V cm}^2 \) and \( 2.5 \times 10^{-17} \text{ Vcm}^2 \). This field is too high for the electrons to be in thermal equilibrium with the gas. In the same figure we show the results of Allen and Prew [10] concerning the mobilities of non-thermal electrons for a domain of E/N similar to ours. Our apparent negative polarity mobility is in good agreement with Allen and Prew results [10]. We can say that the charge carriers in our experiments were indeed non-thermal electrons.
Figure 5. Mobility according to the density of nitrogen. Negative charge carriers. □ K-app. our results ♦ non-thermal electronics [10] ▼ thermal electronic head [11]

3.2 Ionic mobility

The tests are conducted here with a positive tip, the charge carriers created are then positive ions. The dependence of K+ with the density of nitrogen is shown in Figure 6. In this same figure, we gather the results of Gee et al [12] concerning the mobilities of N2+ ions in nitrogen gas, which are obtained by the method of flight time. We notice a good agreement between our results and those of these authors. Using the advanced Langevin theory, we plot the theoretical mobility curve for N2+ ions in Figure 6. We obtain a good agreement between this curve and our experimental values as well as those of Gee et al [12]. When the carriers are ions (positive polarity point for example), we have shown that the mobility can be deduced from the slope p of the line I (V). We have seen previously that the variation with the density of the ionic mobility is in good agreement with the direct measurements by the time of flight method. On the other hand, if our measurements are made in a gas without purification or in a gas that has remained in the cell for several days, the mobilities obtained will be very low [14]. We present the results for a "low degree of purification" gas of our predecessor in argon by indirect measurement and compare them to our own values for non-thermal electrons (Figure 6). We can see that the mobilities of the carriers in a gas without purification are between those of the non-thermal electrons corresponding to the electric field existing in the transport zone and those of O2 ions. Depending on the tests and the degree of purification of the gas, the apparent mobility of the negative carriers is given by:

\[ K_{\text{app}} = \frac{S^n K_i X_i}{S^n_i X_i} \]

where \( K_i \) is the mobility of ions (O2 eg.) or electrons and \( X_i \) their respective molar fraction.
Figure 6. Mobilities of ions in nitrogen as a function of density. □ K-app. our results • negative ions [13] ▼ positive ions [12], ▲ K +, positive ions [13], ----- curve according to the theory of Langevin evolved

By extrapolating the experimental points to densities corresponding to the unpurified liquid argon of Bonifaci [15] in which the mobility of the charge carriers corresponds to that of the O$^{2-}$ [16] ions, we can conclude that the higher the density increases the higher the molar fraction of O$^{2-}$, so the apparent mobility becomes the mobility of O$^{2-}$ ions. We retrace in the same figure the theoretical Langevin curves approach quantum and corrected by the resonance charge transfer (here called the evolved Langevin theory) [13]. According to this theory, we report the cause of measurement accuracy (such as current, pressure, density, and voltage measurements). It should be noted that the advanced Langevin theory is still valid for N$_2^+$ ions in the gas up to the critical pressure. The differences between the mobility of ions come from the polarizability differences $\alpha$ and the molar masses [1, 17]. Therefore, if one calculates the mobility from the Langevin theory, mobility is decrease as function of gas pressure or density.

4. Conclusion
We have demonstrated, following the phenomenon observed visually and with the characteristic current voltage, the existence of the corona discharge in very pure nitrogen gas. The shapes of geometry (the distance between two electrodes, the polarity, the radius of curvature of the tip etc.) are the decisive parameters for initiating a corona discharge. These parameters intervene on the electric field in the vicinity of the active electrode. Therefore, Weisler's conclusion must be examined and verified that a corona discharge cannot be generated in very pure nitrogen gas. From the current-voltage characteristic, we can conclude that the regime of our discharge in very pure gases (N$_2$) corresponds to the current regime limited by space charge. By analogy with what has been observed in a "discharge tube" this regime is similar to an abnormal glow discharge regime. The good agreement between the values deduced from our I-V characteristics and the mobility flight time measurements has shown us that our indirect measurement method is reliable and is equivalent to a direct measurement by flight time. In addition, the advantage of our assemblies comes from the facility (implementation) of installation. From the comparison of the results on the mobilities of well purified gases with those poorly purified, we have shown that the system of purification and circulation of the gas can remove the content of electronegative impurities (e.g., oxygen) from the gas. Our study has
made it possible to specify the nature and the characteristics of the charge carriers but a modeling of non-thermal electrons in a dense medium remains to be done.

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