

Effects of metastable atoms on breakdown voltage in Argon DBD

T Yoshinaga and H Akashi

National Defense Academy of Japan, 1-10-20 Hashirimizu, Yokosuka, Kanagawa
239-8686, Japan

yoshinag@nda.ac.jp

Abstract. A one-dimensional fluid calculation is performed to find out the effects of metastable atoms in alternating-current (AC) driven dielectric barrier discharge (DBD) of argon. Dependences of the breakdown voltage and the density of the metastable state atoms on the driving frequency and the pressure are investigated to clarify the importance of the secondary electron emission by metastable atoms and the metastable-metastable ionization reaction. The results suggest that such processes can be significant when the driving frequency becomes several hundred Hz under several Torr.

1. Introduction

DBD is applied in a variety of chemical plasma processes and light sources to generate non-equilibrium plasmas [1]. Dielectric layers play an important role by suppressing excessive current which leads to the damaging of electrodes. They are also important since the electrons necessary for self-sustaining discharge are yielded via secondary electron emission. While the dielectric materials (e.g. MgO) are regarded to have higher secondary electron emission coefficient (γ) than metals, those effects have not been evaluated well. Two major experimental methodologies have been attempted to estimate γ . One is a direct measurement in which the accelerated ion beam is bombarded and the emitted electron current is measured [2]. This method can measure γ defined as the ratio of the electron yield to the injected ion flux, while the injected ions tend to have higher energy levels (over tens of eV) than that of interest in DBD operation (at most ~ 10 eV). The other method measures the breakdown voltage (V_{bd}) between two parallel planar electrodes and deduce the secondary ionization coefficient (γ') using the Townsend's criterion for the self-sustaining discharge [3-6]. This method is rather close to the practical discharges in terms of the energy level of bombarding particles. While γ and γ' are strongly connected with each other, those values would not be exactly the same since γ' includes the backscattering effect via collisions with the background gas particles. The experimental approaches often take into account the contributions only from the ions. However, there exist other kinds of excited species in actual discharges. For example, it is well known that the effect of metastable atoms cannot be neglected especially in AC-driven DBDs because of their long lifetimes [7].

In this work we examine the effect of the secondary electron emission by metastable atoms and the metastable-metastable ionization reaction by investigating the discharge characteristics such as V_{bd} in relation to the driving frequencies (f) and pressures (p_0) using a one-dimensional numerical model.



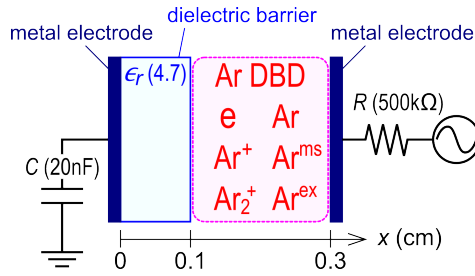


Figure 1. Schematic of one-dimensional fluid model for argon DBD.

Table 1. Reaction processes included in the present model.

No.	Reaction	Rate coeff. (cm ³ s ⁻¹)	Ref.
1	$e + \text{Ar} \rightarrow \text{Ar}^{\text{ms}} + e$	$k_{\text{ms}}(\varepsilon_e)$	[8,9]
2	$e + \text{Ar} \rightarrow \text{Ar}^{\text{ex}} + e$	$k_{\text{ex}}(\varepsilon_e)$	[8,9]
3	$e + \text{Ar} \rightarrow \text{Ar}^+ + 2e$	$k_i(\varepsilon_e)$	[8,9]
4	$e + \text{Ar}^{\text{ms}} \rightarrow \text{Ar}^+ + 2e$	$k_{\text{cui}}(\varepsilon_e)$	[8,10]
5	$\text{Ar}^{\text{ms}} + \text{Ar}^{\text{ms}} \rightarrow \text{Ar}^+ + \text{Ar} + e$	5×10^{-10}	[11]
6	$\text{Ar}^{\text{ex}} \rightarrow \text{Ar} + h\nu$	10^{-9} ^a	[8]
7	$\text{Ar}^+ + 2\text{Ar} \rightarrow \text{Ar}_2^+ + \text{Ar}$	2.5×10^{-31} ^a	[8,12]
8	$\text{Ar}_2^+ + e \rightarrow 2\text{Ar}$	8.8×10^{-8}	[13]

^a Dimensions of reaction 6 and 7 are (s⁻¹) and (cm⁶ s⁻¹).

2. Numerical Model

A simple one-dimensional fluid model is adopted in this study. Figure 1 shows the discharge gap configuration with the external circuit. The grounded side of the parallel plane electrodes with cross section of 4 cm² is covered with the dielectric material with relative permittivity (ϵ_r) at 4.7 and thickness (d_b) of 0.1 cm. The discharge gap (d) is 0.2 cm. The plasma is assumed to be uniform on the plane parallel to the electrodes, so that the transport parameters vary only with the distance from the electrodes. It is also assumed that the plasma is consisted with electrons (e), ions (Ar^+ , Ar_2^+), metastable atoms (Ar^{ms}) and other excited atoms (Ar^{ex}) besides the ground-state argon atoms (Ar). There are an external resistor ($R = 500$ kΩ) connected on the powered side and a capacitor ($C = 20$ nF) on the grounded side as experiments [6]. The continuous-wave (CW) sinusoidal voltage with amplitude at 500 V is applied. The one-dimensional equation of continuity in discharge space is denoted by,

$$\frac{\partial n_j}{\partial t} = -\frac{\partial \Gamma_j}{\partial x} + S_j, \quad (1)$$

$$\Gamma_j = W_j n_j - D_j \frac{\partial n_j}{\partial x}, \quad (2)$$

where the subscript j represents the particle species (e , Ar^+ , Ar_2^+ and Ar^{ms}). n_j , Γ_j , S_j , W_j , and D_j represent the particle density, flux, production (or loss) term, drift velocity and diffusion coefficient of the particle j , respectively. Only W_j of charged particles can have finite values. The advection of Ar^{ex} is not calculated in this study since the de-excitation dominates because of its short lifetime (estimated as 10^{-9} s). The reactions for S_j included in the present calculation are listed in table 1. The above swarm parameters are deduced using the Monte-Carlo simulation with the same cross-sections described in [8]. The conservation of electron energy is considered as follows,

$$\frac{\partial (n_e \varepsilon_e)}{\partial t} = -\frac{5}{3} \left(\Gamma_e \varepsilon_e - D_e n_e \frac{\partial \varepsilon_e}{\partial x} \right) - e E \Gamma_e + S_\varepsilon. \quad (3)$$

Here, ε_e is the mean energy of electrons, E the electric field and S_ε the gain and loss term of electron energy via inelastic reactions. It is assumed that the electron velocity distribution is Maxwellian. Time evolutions of the particle densities and the electron energy are computed using equations (1) – (3) coupled with the Poisson's equation,

$$\frac{\partial E}{\partial x} = e(n_{\text{Ar}^+} + n_{\text{Ar}_2^+} - n_e)/\epsilon_0. \quad (4)$$

At the plasma boundaries, the secondary electrons flow into the discharge space both from the metal electrode and the dielectric surface. The fluxes $\Phi_{2\text{nd}}$ are given by,

$$\Phi_{2nd} = \gamma_i(\Gamma_{Ar^+} + \Gamma_{Ar_2^+}) + \gamma_m \Gamma_{Ar^{ms}} + \gamma_p \Gamma_p, \quad (5)$$

where γ_i , γ_m and γ_p represent the secondary electron emission coefficient of ions, metastable atoms and photons, respectively. Γ_p is the photon flux onto the electrodes. The values of γ will be different depending on the influx species and electrode materials.

3. Results and discussion

Figure 2 shows waveforms of a discharge driven at $f = 20$ Hz under $p_0 = 5$ Torr. Positive polarity of the discharge current corresponds to the direction in which the positive voltage is applied on the powered electrode and the positive ions flow towards the grounded dielectric electrode ($-x$ direction in figure 1). Secondary electron emission coefficients (γ_i , γ_m , γ_p) are all set at 0.01 for both of the metal and dielectric electrodes. The breakdown occurs with the inrush discharge current (I) with a peak at ~ 10 μ A at $t \sim 0.1$ s as shown in figure 2 (c). The gap voltage (V_g) is ~ 250 V at this moment. After this breakdown I immediately decreases and V_g stops increasing and is kept constant at ~ 235 V until the source voltage (V_s) starts to decrease as shown in figures 2 (c) and (d). There remains a small current flowing after the breakdown and the positive charge of ions accumulates on the dielectric barrier (q) as shown in figure 2 (c). This charge compensates the excessive voltage impressed between the discharge gap despite the increasing V_s . The spatially averaged electron density ($\langle n_e \rangle$) increases and decreases along with I , while the spatially averaged density of metastable atoms ($\langle n_{ms} \rangle$) shows a quite slow decrease after the breakdown due to the long lifetime and the small diffusion coefficient as shown in figure 2 (a). The spatially averaged electron energy ($\langle \epsilon_e \rangle$) has risen upto ~ 10 eV before breakdown and remains roughly constant until V_g starts to decrease at $t \sim 0.115$ s as shown in figure 2 (b). Then, $\langle \epsilon_e \rangle$ decreases to almost zero when the polarity of V_g is reversed ($t \sim 0.12$ s). The magnified waveforms when breakdown occurs [figures 2 (e) and (h)] show that I around 1 μ A increases exponentially with t as well as $\langle n_e \rangle$ with the time constant at $\tau_{1\mu A} [= I/(dI/dt)] \sim 5$ μ s. As shown in

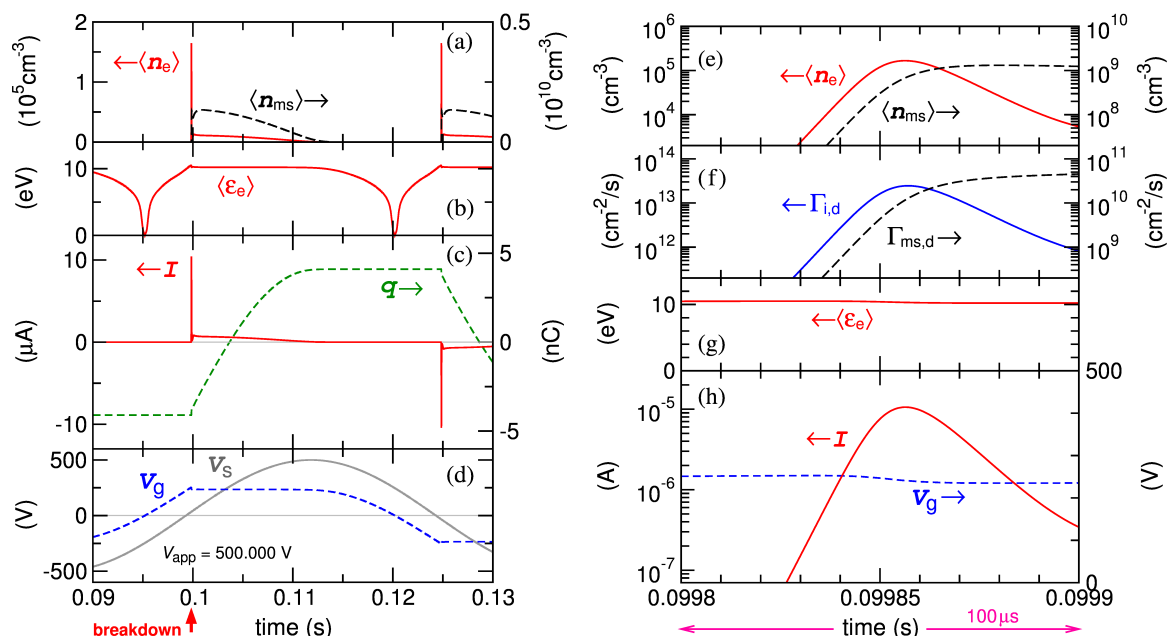


Figure 2. Typical waveforms of a discharge driven at 20 Hz. (a): Spatially-averaged density of electrons $\langle n_e \rangle$ and metastable atoms $\langle n_{ms} \rangle$. (b): Spatially-averaged electron energy $\langle \epsilon_e \rangle$. (c): Discharge current I and accumulated charge on the dielectric layer q . (d): Applied voltage V_s and gap voltage V_g . (e)-(h): Magnified waveforms around breakdown event. (e): Density ($\langle n_e \rangle$ and $\langle n_{ms} \rangle$). (f): Particle flux of ions $\Gamma_{i,d}$ and metastable atoms $\Gamma_{ms,d}$ onto the dielectric layer. (g): Electron energy ($\langle \epsilon_e \rangle$). (h): Discharge current I and gap voltage V_g .

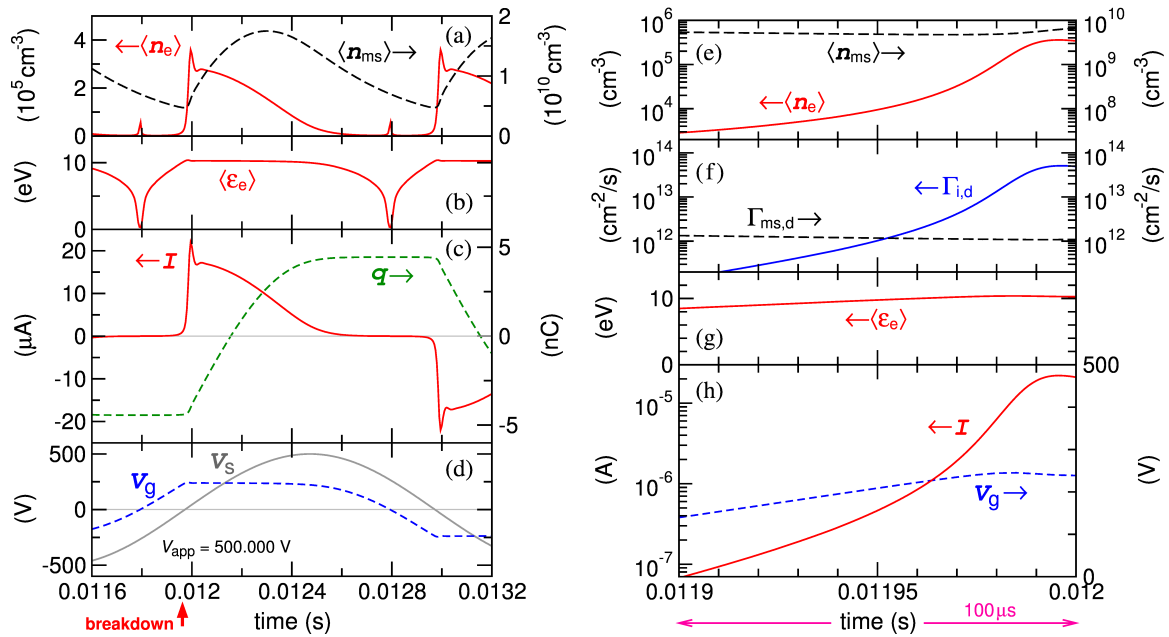


Figure 3. Discharge waveforms driven at 500 Hz. Plotted parameters are the same as shown in figure 2.

figure 2 (f) the influx of ions flowing onto the dielectric layer ($\Gamma_{i,d}$), which shows an exponential increase as I and $\langle n_e \rangle$, is found to be $10^3 - 10^4$ times higher than the metastable atoms' flux ($\Gamma_{ms,d}$). Here, $\Gamma_{i,d}$ represents the summed flux of monomer and dimer ions ($\Gamma_{Ar^+} + \Gamma_{Ar_2^+}$), while the monomer ions flux is several hundred times higher than the dimer ions flux. Since the three-body reaction will hardly occur under such low pressure discharge, dimer ions' density is low compared with the monomer ions' density. The secondary electron emission by photons can be negligible since the photon flux ($\Gamma_{p,d}$) is estimated at $\sim 1 - 100 \text{ cm}^{-2}/\text{s}$, being much less than even $\Gamma_{ms,d}$ in this discharge. Therefore, ions are the most significant species for the secondary electron emission at the breakdown as long as γ_i does not become several orders of magnitudes smaller than γ_m .

Figure 3 shows the waveforms of a discharge driven at $f = 500 \text{ Hz}$, while other discharge conditions are the same with the discharge shown in figure 2. Similarly to the discharge at $f = 20 \text{ Hz}$, I increases rapidly when V_g reaches some particular level (breakdown occurs) and V_g becomes almost constant (between 230 - 240 V) after the breakdown as shown in figures 3 (c) and (d). After the inrush of the initial current, however, I does not decrease immediately and forms a long tail in the order of $10 \mu\text{A}$ for $\sim 300 \mu\text{s}$. A long decay time can be found also in $\langle n_e \rangle$ as shown in figure 3 (a). The time evolution of $\langle n_{ms} \rangle$ is of the largest difference from the case at $f = 20 \text{ Hz}$. It appears to increase and decrease independently of the other parameters and remains finite throughout the discharge cycle [see figure 3 (a)]. The long decay time of $\langle n_{ms} \rangle$ would be originated by that the metastable atoms' motion is not governed by the electric field but only by the slow diffusion adding to their long lifetimes. Both the metastable-metastable ionization (reaction 5 in table 1) and the secondary electron emission can occur throughout the discharge due to the remaining metastable atoms, yielding electrons and ions. This suggests that $\langle n_{ms} \rangle$ would be a significant parameter in high frequency operations. Since $\Gamma_{ms,d}$ is found to be similar to $\Gamma_{i,d}$ ($\sim 10^{12} \text{ cm}^{-2}/\text{s}$) around the breakdown ($t \sim 0.01195 \text{ s}$) the metastable atoms would have a comparable contribution to the ions for the secondary electron emission as shown in figure 3 (f). I and $\langle n_e \rangle$ do not increase exponentially but in a complex and slower way in contrast to the discharge at 20 Hz as shown in figures 3 (e) and (h). The time constant at $0.1 \mu\text{A}$ ($\tau_{0.1\mu\text{A}}$) is $\sim 30 \mu\text{s}$, while it decreases to $\tau_{1\mu\text{A}} \sim 13 \mu\text{s}$ at $1 \mu\text{A}$ and $\tau_{10\mu\text{A}} \sim 8 \mu\text{s}$ at $10 \mu\text{A}$.

Since it is difficult to define the breakdown voltage exactly, we simply focus on the voltage when I reaches $1 \mu\text{A}$ ($V_{1\mu\text{A}}$). Figure 4 (a) shows the dependence of $V_{1\mu\text{A}}$ on f with different γ_m . Without the

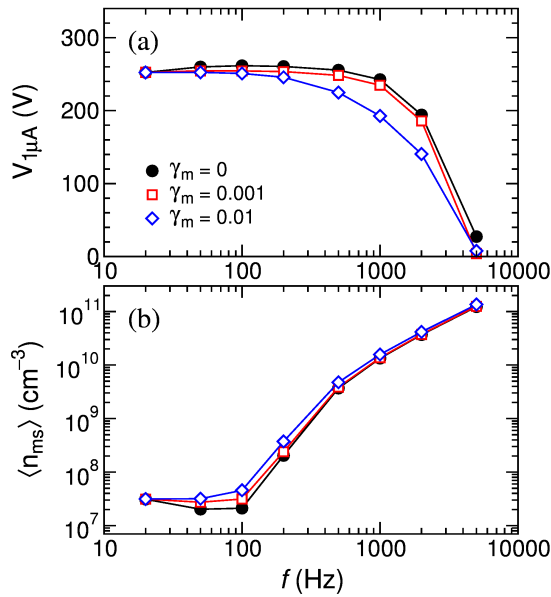


Figure 4. Dependence of (a) breakdown voltage $V_{1\mu A}$ and (b) metastable atoms density $\langle n_{ms} \rangle$ on driving frequency with different γ_m .

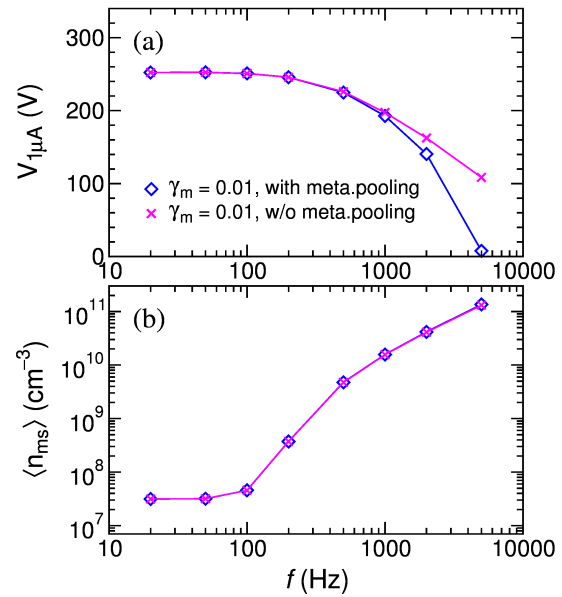


Figure 5. Comparison of frequency characteristics between the cases with and without the metastable-metastable ionization reaction process.

secondary electron emission by metastable atoms ($\gamma_m = 0$), $V_{1\mu A}$ is independent of f when f is lower than ~ 200 Hz, while it decreases with f over 500 Hz. $\langle n_{ms} \rangle$ at the same moment ($I = 1 \mu\text{A}$) increases with f when f becomes higher than 100 Hz as shown in figure 4 (b). Since the decrease of $V_{1\mu A}$ becomes remarkable as γ_m becomes higher, it is suggested that the secondary electron emission by the metastable atoms strongly affects the breakdown voltage under such higher frequency range. The condition under which $V_{1\mu A}$ appears to decrease at $f = 500$ Hz with $\gamma_m = 0.01$ corresponds to the condition when the secondary electron flux by the metastable atoms ($\gamma_m \Gamma_{ms,d}$) becomes comparable to that by ions ($\gamma_i \Gamma_{i,d}$) in figure 3. On the other hand, $V_{1\mu A}$ drops when $\langle n_{ms} \rangle$ becomes $\sim 10^{10} \text{ cm}^{-3}$ even with $\gamma_m = 0$, suggesting that the other effects of metastable atoms (e.g. metastable-metastable ionization or cumulative ionization from metastable atoms) are also taking place. Note that $V_{1\mu A}$ becomes almost zero at $f = 5$ kHz with any γ_m as shown in figure 4 (a). This indicates that the discharge becomes continuous and the current-carrying species remain in the discharge space throughout one discharge cycle.

Figure 5 compares the case with and without the metastable-metastable ionization process with $\gamma_m = 0.01$. The values of $V_{1\mu A}$ are quite similar under $f \lesssim 1$ kHz as shown in figure 5 (a). The difference between the two cases appears only under $f \gtrsim 2$ kHz, which is higher than the frequency level that the effect of γ_m appears. This suggests that higher $\langle n_{ms} \rangle$ is needed for the metastable-metastable ionization process than γ_m effect to affect $V_{1\mu A}$. This process would not be the primary loss mechanism of metastable atoms since $\langle n_{ms} \rangle$ takes the same value in both cases as shown in figure 5 (b). Considering the discharge driven at $f = 500$ Hz, $\langle n_e \rangle \sim 2 \times 10^4 \text{ cm}^{-3}$ and $\langle n_{ms} \rangle \sim 4 \times 10^9 \text{ cm}^{-3}$. The rate coefficient of cumulative ionization (k_{cui}) is roughly estimated at $\sim 10^{-7} \text{ cm}^3 \text{ s}^{-1}$ from $\langle \epsilon_e \rangle \sim 10$ eV around breakdown [see figure 3 (b)], besides the rate coefficient of metastable-metastable ionization (k_{mm}) is assumed at $5 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ (see table 1). The reaction rates of these processes become $\langle k_{cui} n_e n_{ms} \rangle \sim 8 \times 10^6 \text{ cm}^{-3} \text{ s}^{-1}$ and $\langle k_{mm} n_{ms}^2 \rangle \sim 8 \times 10^9 \text{ cm}^{-3} \text{ s}^{-1}$, respectively, showing that the metastable-metastable ionization process dominates the loss processes of metastable atoms. Therefore, the metastable atoms would be lost mainly via the diffusion process (the diffusion coefficient is assumed as $D_{ms} = 50 \text{ cm}^2 \text{ s}^{-1} \text{ Torr}^{-1}$ [14]) from the discharge space since the metastable-metastable ionization is not the dominant loss mechanism of metastable atoms.

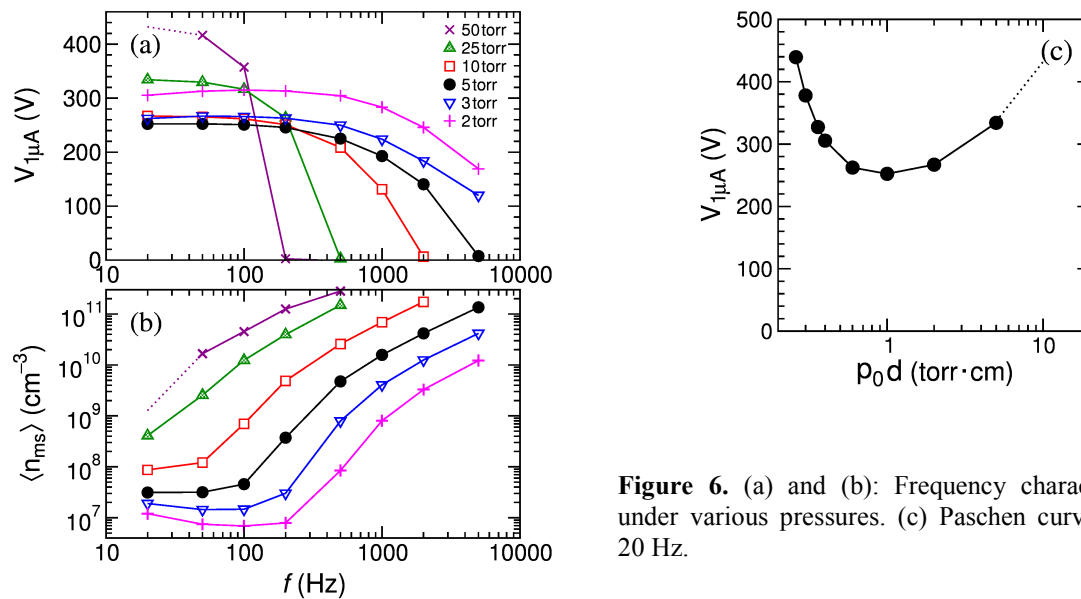


Figure 6. (a) and (b): Frequency characteristics under various pressures. (c) Paschen curve at $f = 20$ Hz.

Figures 6 (a) and (b) show the frequency dependences under different p_0 . The frequency at which $V_{1\mu A}$ decreases becomes lower under higher p_0 . The increase of $\langle n_{ms} \rangle$ with p_0 would be caused from the smaller diffusion coefficient of metastable atoms under higher p_0 . Although a breakdown occurs at $V_g \sim 430$ V under $f = 20$ Hz and $p_0 = 50$ Torr, I does not reach 1 μA . By plotting $V_{1\mu A}$ at $f = 20$ Hz against $p_0 d$ the Paschen curve can be obtained as shown in figure 6 (c). This characteristics resembles the experimentally obtained one (figure 10 in [4]) although the values in detail are different. The pressure of 5 Torr corresponds to the pressure at Paschen minimum ($p_0 d = 1$ Torr·cm).

4. Summary

The effects of metastable atoms in argon DBD between two parallel planar electrodes are investigated using one-dimensional fluid model. The breakdown voltage decreased with f when f exceeds several hundred Hz. Under $p_0 = 5$ Torr, the effect of γ_m first appears at $f \sim 500$ Hz. Next the metastable-metastable ionization process takes place at ~ 2 kHz. Under $f \lesssim 200$ Hz the effects of metastable state atoms do not appear. The metastable atoms' density builds-up higher and the breakdown voltage decreases more largely under higher p_0 . These results suggest that γ_m of the dielectric materials in DBD might be evaluated as well as γ_i by examining the breakdown voltage under various p_0 and f .

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