Capacitively Coupled RF Discharge Breakdown in Gas Mixtures

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Keywords : breakdown, RF discharge, gas mixtures, Penning ionization

Recently many studies on gas breakdown in the parallel plate capacitively coupled RF discharges have been made experimentally and numerically. Therefore breakdown curves (dependence of the breakdown voltage on the filling pressure) for various gases have been obtained so far, although a few papers exist for the breakdown in gas mixtures which are widely used in the present plasma technologies.

In these studies the breakdown curves for many gases are known to have the V-shaped characteristics made up of two (right and left) branches. In the right branches where electrons are well trapped between the electrodes the breakdown voltage shows the monotonous increase with the pressure, and the characteristics have been explained frequently by the electron production and loss processes such as the direct ionization and the diffusion loss. Now there also exist some gases, for instance, Ne and He which show another type of breakdown curve, that is L-shaped one. In these gases it has been suggested that the excited particles play an important role on breakdown. So it is interesting to study the role of ionization processes other than the direct ionization in RF breakdown.

The Penning ionization is a very useful ionization process in the breakdown of gas mixture because it can decrease the breakdown voltage remarkably as shown in DC and microwave discharges. However, the Penning process has been hardly studied in RF gas breakdown so far. In this paper we experimentally study RF gas breakdown in Ne-N 2, Ne-Ar and He-N 2 gas mixtures where the Penning ionization is to be expected. Therefore our interest is specifically focused on the breakdown characteristics at the small values of the mixing ratio x (N 2/Ar/Ne(He)+N 2/Ar). The breakdown voltages \( V_{Sp-p} \) are measured at the frequency of 13.56 MHz using a parallel plate capacitively coupled RF discharge device, and the measurements were made allowing the gas flow to avoid the accumulation of impurities.

In Fig. 1 (a), the breakdown curves of Ne-N 2 gas mixtures are shown taking \( x \) (%) as a parameter. As can be seen from this figure, the addition of a small amount of N 2 has no great influence on the characteristics of the left branch, while \( V_{Sp-p} \) in the right branch decreases and then increases with increasing \( x \) in some pressure range. The upper limit of this pressure range where the decrease of \( V_{Sp-p} \) appears is \( pL \approx 400 \) torr \( \cdot \) mm. About 22 \% decrease of \( V_{Sp-p} \) is obtained as the maximum value at 1.2 torr for \( L =26.4 \) mm. Since the decreases of \( V_{Sp-p} \) are not observed in Ar-N 2 mixtures where the Penning ionization does not occur definitely, the decreases in the right branches of Ne-N 2 mixtures are considered to be attributed to the Penning effect. Fig. 1 (b) shows the breakdown characteristics of Ne-Ar mixtures. Here the reason we choose this mixture system is as follows: the ionization potential of Ar is approximately equal to that of N 2, and therefore it is expected that there exists the difference in the degree of Penning effect between rare and molecular gas mixture and rare and rare gas mixture. The breakdown characteristics obtained are similar to that of Ne-N 2 mixtures. The addition of small amount of Ar brings about the decrease of the breakdown voltage and it reaches 32 \% at 2 torr for \( L =26.4 \) mm. The upper limit of the pressure range where the Penning effect appears is larger in Ne-Ar mixtures than in Ne-N 2 ones and its value is \( pL \approx 1050 \) torr \( \cdot \) mm. This result shows that there exists the difference between the characteristics of the right branches of these two mixture systems, and suggests that the number of electrons of higher energy which plays an important role in the production of metastable atoms is larger in Ne-Ar mixtures. The conventional Boltzmann analysis really shows that the average electron energy in Ne-Ar mixtures is higher than in Ne-N 2 mixtures. He-N 2 system which means another example of the Penning effect is also studied. The measurements results not shown here indicate that the maximum decrease ratio of voltage is about 30 \% at 0.3 torr.

As seen in these cases the decrease of breakdown voltage is certainly observed in RF breakdown, but the decrease ratio for each of gases is relatively small compared with other discharge systems such as DC and microwave discharges. So this thing suggests that in RF breakdown there exist some competitive ionization processes accompanied by excited particles including the Penning ionization, and numerous metastable particles are consumed in ionization processes except the Penning one.
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The RF (13.56 MHz) gas breakdown in Ne-N₂, Ne-Ar and He-N₂ mixture systems are investigated experimentally in a parallel plate capacitively coupled device with varying the pressure, the mixing ratio and the electrode separation, specifically focussing on the breakdown at the small values of the mixing ratio. The breakdown voltage curves obtained have the V-shaped or L-shaped characteristics. The breakdown voltage decreases with the addition of a small amount of N₂ or Ar in each mixture system, and the ratios of decrease in breakdown voltage to rare gas breakdown voltage in these mixture systems are about 22, 32 and 30 % at the maximum, respectively. The decrease of breakdown voltage is observed in a certain restricted pressure range, which is narrower in Ne-N₂ system than in Ne-Ar one. This feature in Ne-N₂ system is probably due to the decrease of electron energy attributed to the vibrational excitation of N₂ by electron impact. Compared with the DC breakdown and the microwave one, the decrease ratio in the RF breakdown is relatively small, suggesting that there exist some competitive ionization processes accompanied by excited particles including the Penning ionization.

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1. Introduction

Recently many studies on gas breakdown in the parallel plate capacitively coupled RF discharges have been made experimentally and numerically\(^{(1)-(3)}\). Therefore breakdown curves (dependence of the breakdown voltage on the pressure) for various gases have been obtained so far, although a few papers\(^{(4)-(8)}\) exist for the breakdown in gas mixtures which are widely used in the present plasma technologies.

In these studies the breakdown curves for many gases are known to have the V-shaped characteristics made up of two (right and left) branches. In the right branches the breakdown voltage shows the monotonous increase with the pressure, and electrons are well trapped between the electrodes because of the smaller amplitude of the oscillatory motion of electrons compared to the electrode separation. So the characteristics of the right branches have been explained frequently by the electron production and loss processes such as the direct ionization and the diffusion loss\(^{(7)}\). Now, another type of breakdown curve, L-shaped one, has been found out for Ne\(^{(6)-(8)}\). The feature in L-shaped curves is that the behaviour of the right branch shows the flat characteristics in the broad range of pressure, and it has been suggested that the metastable particle plays an important role in ionization processes in the right branch\(^{(8)}\). There also exists an interesting idea that in relatively higher pressure range of the right branch the photo-electrons released from the walls due to the radiation from the resonance states of Ne are important in the breakdown\(^{(9)}\).

The suggestion that the excited particles take part in the RF breakdown leads us to another ionization process, namely the Penning ionization. The Penning ionization is a very useful ionization process in the breakdown of gas mixture because it can decrease the breakdown voltage remarkably as shown in DC\(^{(10)}\) and microwave discharges\(^{(11)-(12)}\). However, the Penning process has been hardly studied in RF gas breakdown so far\(^{(8)-(13)}\). Therefore it is interesting to investigate not only the direct ionization but also the Penning ionization in the breakdown of RF discharges.

In this work we obtain the breakdown voltage curves for Ne-N₂, Ne-Ar and He-N₂ systems in RF discharges with varying the mixing ratio of N₂ or Ar. Specifically our interest is focused on the breakdown characteristics at the small values of the mixing ratio. Then we analyze the measurement results using the conventional Boltzmann analysis based on the two-term approximation and compare our results with those obtained in other discharge systems.

2. Experimental Setup

The experiments were performed at the frequency of 13.56 MHz using the parallel plate capacitively coupled RF device (640 × 640 × 200 mm\(^3\)) in which aluminum electrodes (500 × 400 mm\(^2\))
are equipped as shown in Fig. 1. The electrode separation $L$ can be varied from 26.4 to 77.4 mm and in the present experiments we adopted the two cases of 26.4 and 77.4 mm as $L$. For Ne-N$_2$ and Ne-Ar systems premixed gases were used and for He-N$_2$ system these two gases were mixed with mass flow controllers adjusting each flow rate. Ne and He respectively contain N$_2$ of 4 and 0.1 ppm at the maximum as an impurity. To avoid the accumulation of impurities, the measurements were made allowing the gas flow with varying the flow rate from 14 to 200 ccm. The effect of gas flow on the breakdown voltage was hardly observed except for Ne over 50 ccm. So the flow rates were fixed at 24 ccm for Ne based mixtures and at 200 ccm for He based ones.

The measurement procedures in two branches were different$^{15}$. In the right branches the breakdown voltage $V_{SP-p}$, peak to peak value, was measured with the high voltage probe for a certain gas pressure $p$ fixed increasing the RF voltage little by little until the breakdown occurs. In the left branches, to the contrary, the breakdown pressure was measured with the capacitance manometer for a certain RF voltage fixed increasing the total pressure so slowly from a sufficiently low pressure until the discharge starts. In these measurements the matching of the RF power was adjusted carefully to maintain the reflection power as low as possible. The breakdown voltages obtained were scattered in 6% around the average value in the right branches and pressures in 5% in the left branches.

### 3. Experimental Results

Figure 2 presents the typical oscillograms of RF voltage applied to the powered electrode and the optical emission signal from the discharge space at the breakdown. From this figure it is seen that the RF voltage begins to decrease with a certain delay time to optical emission signal and approaches to the steady state discharge voltage with the small self-bias. The delay time $\tau_{pV}$ is considered to be the time required for the space charge effect to come to distort the uniform electric field in the discharge space. The voltage value just before decreasing can be regarded as the breakdown voltage.

In Figs. 3 (a) and (b), the breakdown curves in Ne-N$_2$ mixtures are shown for $L$ =26.4 and 77.4 mm with the mixing ratio as a parameter. The typical L-shaped curve is obtained for Ne in Fig. 3 (a). It is rather important to clarify the whole pressure dependence in the right branch of the L-shaped curve which is considerably different from the V-shaped one. It has been shown experimentally that the similarity law for $V_{SP-p}$ holds on the product $pL$ in the right branches$^{15}$. Consequently, to study the whole pressure dependence in the right branch, it is sufficient to study the characteristics of the right branch for the large electrode separation ($L$ =77.4 mm). Figure 3 (b) shows that Ne breakdown voltage increases through three stages in the right branch and that the flat characteristics in Fig. 3 (a) correspond to the second stage.

In both these figures, it is found that the breakdown voltage decreases with the addition of a small amount of N$_2$. This decrease of $V_{SP-p}$ is considered to be attributed to the Penning ionization. In Ar-N$_2$ mixtures where the Penning process does not take place, the decrease of $V_{SP-p}$ was not observed certainly in spite of trying out several mixing ratios. Fig. 3 (a) shows that the dependence of $V_{SP-p}$ on the amount of N$_2$ is greatly influenced by the total pressure. At about 20 torr, relatively higher pressure, $V_{SP-p}$ increases monotonously with increasing the mixing ratio of N$_2$, while $V_{SP-p}$ decreases and then increases at 0.2-15 torr. This
means that the decrease of $V_{Sp-p}$ occurs in the restricted range of pressure. The decrease of $V_{Sp-p}$ in the restricted pressure range is similarly observed for $L = 77.4$ mm at the pressures below 5 torr as shown in Fig. 3 (b). The upper limit of the pressure range where the decrease of $V_{Sp-p}$ appears is $pL \approx 400$ torr $\cdot$ mm for both electrode separations as expected from the similarity law. About 22 % decrease of $V_{Sp-p}$ is obtained as the maximum value at 1.2 torr for $L = 26.4$ mm. Figure 3 (a) also shows that the addition of a small amount of N$_2$ has no great influence on the position of the left branch. This result is reasonable, because in the left branch of breakdown the interactions between electrons and electrodes must be more important than the atomic and molecular processes.

Figures 4 (a) and (b) show the breakdown curves with the mixing ratio as a parameter for Ne-Ar mixtures. Here the reason we choose this mixture system is as follows: the ionization potential of Ar is approximately equal to that of N$_2$, and therefore it is expected that there exists the difference in the degree of Penning effect between rare and molecular gas mixture and rare and rare gas mixture. The breakdown characteristics obtained are similar to that of Ne-N$_2$ mixtures. The addition of small amount of Ar brings about the decrease of the breakdown voltage and it reaches 32 % at 2 torr for $L = 26.4$ mm. Another case of the Penning combination of gases, He-N$_2$ system, is presented in Ne-Ar mixtures there is a minimum breakdown voltage as obtained at lower pressures, but in Ne-N$_2$ mixtures $V_{Sp-p}$ monotonously increases with the mixing ratio. This means that the upper limit of the pressure range where the decrease of voltage appears is larger in Ne-Ar mixtures than in Ne-N$_2$ ones, and its value is $pL \approx 1050$ torr $\cdot$ mm. The Penning effect is brought about by metastable particles and the production rates of them are approximately governed by the average electron energy. Therefore we evaluate the average electron energy in both mixture systems by using a two-term approximation of the Boltzmann analysis. The cross-sections used are those recommended by Hayashi for rare gases(14) and by Phelps for nitrogen(15). Figures 7 (a) and (b) show the dependence of the average electron energy $\langle \varepsilon \rangle$ on the mixing ratio for $L = 26.4$ mm. The dependences of $\langle \varepsilon \rangle$ on the mixing ratio at lower pressures (0.5 and 1 torr) are roughly the same in both systems, and consequently the production rates of metastable particles are expected to be approximately equal in both systems. It is true that the values in both systems of the total excitation frequency for Ne, which consists of all excitation processes taken into account in the present calculation, are different in only 15 % at the mixing ratio of 5 % and in 40 % even at 10 %. At relatively higher pressure (10 torr), with increasing the mixing ratio, $\langle \varepsilon \rangle$ decreases more rapidly in Ne-N$_2$ mixtures than in Ne-Ar ones, because of the vibrational excitation of N$_2$ by electron impact. This vibrational excitation is known as the fairly effective process which restrains the electron energy from rising. The result of analysis suggests that the lower decrease ratio in breakdown voltage at the higher pressure in Ne-N$_2$
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It is useful to compare our results with the data of other type of discharge systems. It is known that the breakdown voltage in the DC discharge system decreases drastically due to the Penning effect. For example, in Ne-Ar mixtures the breakdown voltage decreases about 75% with the addition of 0.01% Ar at $pL = 1000$ torr \cdot mm\textsuperscript{(10)}. The microwave discharge system is specifically interesting, because the electrons are trapped in the discharge space as same as the right branch of RF breakdown. It has been reported that in Ne-Ar mixtures the breakdown voltage decreases about 70% with the addition of 0.1% Ar at the frequency of 9.5 GHz\textsuperscript{(11)} and about 80% with 0.12% Ar at the frequency of 2.8 GHz\textsuperscript{(12)}. In our results the maximum decrease ratio in breakdown voltage is about 32% which is rather a small value in comparison with those in other discharge systems shown here.

5. Conclusions

We experimentally investigated the mixing effect of small amount of N\textsubscript{2} and Ar on the parallel plate RF gas breakdown in Ne-N\textsubscript{2}, Ne-Ar and He-N\textsubscript{2} mixture systems. The results obtained in this work are summarized as follows:

1. The breakdown voltage decreases with the addition of a small amount of N\textsubscript{2} or Ar in each mixture system. The maximum decrease ratios are about 22, 32 and 30% respectively.

2. The decrease of breakdown voltage is observed in a certain restricted range of pressure. This result is more noticeable in Ne-N\textsubscript{2} system than in Ne-Ar one and probably due to the decrease of electron energy attributed to the vibrational excitation of N\textsubscript{2} by electron impact.

3. The decrease ratio in breakdown voltage in RF breakdown is small in comparison with other discharge systems such as the DC discharge and the microwave one. This result suggests that numerous metastable particles are consumed in other ionization processes competitive with the Penning one.

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