

# Change of the argon-based atmospheric pressure large area plasma characteristics by the helium and oxygen gas mixing

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Available online 16 November 2006

## Abstract

Oxygen and helium gases, often used in many plasma processes, were added to argon-based glow plasmas, produced at the atmospheric pressure, in order to study the controllability of the plasma characteristics by the supply gas mixing. Based on the electrical and optical diagnostics, the plasma parameters, such as the breakdown voltage, the rotational temperature, and the plasma uniformity, and their changes due to the gas mixing were investigated. The experimental results showed that the helium gas addition reduced the breakdown voltage (from 430 V to 300 V), the rotational temperature (from 465 K to 360 K), and the plasma uniformity. On the other hand, a small amount of oxygen gas increased the breakdown voltage (from 435 V to 463 V) and the rotational temperature (from 520 K to 600 K) due to various energy loss channels of the oxygen gas. The experimental results showed that it was possible to control the plasma characteristics by the gas mixing. Crown Copyright © 2006 Published by Elsevier B.V. All rights reserved.

**Keywords:** Gas mixing; Atmospheric plasma; Large area plasma

## 1. Introduction

Atmospheric pressure plasmas have recently been utilized for various industrial applications due to many advantages [1–3]. One of the main topics of the atmospheric pressure plasma studies is the possibility of replacing conventional low pressure plasmas for various processes. In order to accomplish the purpose, certain discharge conditions with a low breakdown voltage, a large volume, a high spatial uniformity, discharge stability, and so on are required. An argon gas, typically used for the atmospheric pressure plasma generation, is appropriate for producing relatively large and uniform plasmas, but the argon plasmas have relatively high gas temperature. On the other hand, helium plasmas usually exhibit low breakdown voltage and low gas temperature. Furthermore, oxygen is often used in industrial applications, such as in ashing and surface modification, because of its high chemical reactivity. Since each gas has its unique characteristics along with advantages and disadvantages for certain applications, the influence of the gas (He, O<sub>2</sub>) mixing on the argon-based atmospheric pressure plasma characteristics should be studied. Thus, based on the plasma characteristics for each gas as mentioned above, it was investigated

how the gas mixing could affect the plasma characteristics so as to take advantages of each gas.

## 2. Experimental setup

A schematic diagram of the plasma source is shown in Fig. 1. It has a double electrode structure with two side-discharge gaps, which provide not only a volumetric but also a stable plasma generation. The plasma was generated in the ambient air with argon, helium, and oxygen gases supplied through an array of gas holes. The bottom-discharge gap distance was fixed as 3 mm for this work. The central electrode was powered by a 13.56 MHz rf source with a matching network. The bottom electrode consisted of a copper mesh and a glass plate in order to observe the plasma. The images show a bottom view and a side view of the plasma when the plasma filled the whole discharge volume. The typical plasma area was 200 mm × 50 mm and the height was 3–5 mm.

## 3. Results and discussions

### 3.1. Argon and helium mixing

Fig. 2(a) shows the change in the breakdown voltage as the helium gas flow rate was varied from 0 to 10 lpm with the argon

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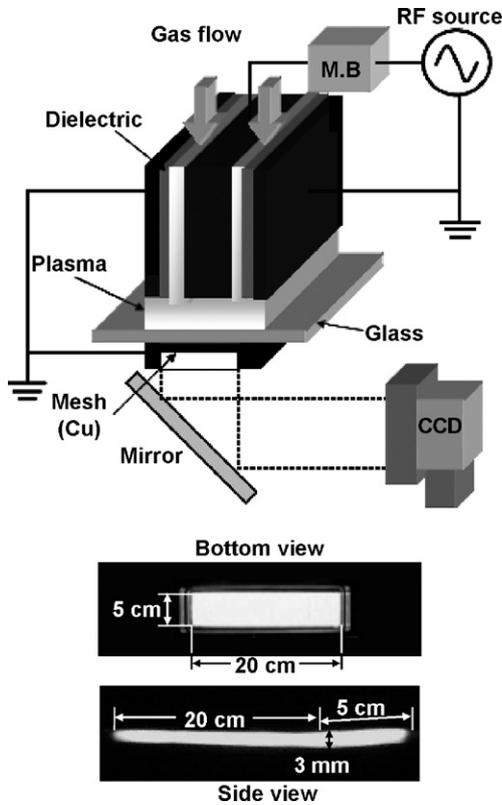


Fig. 1. An experimental set up for the atmospheric pressure plasma source and the generated plasma images in the bottom view and the side view.

gas flow rate fixed at 10 lpm. As increasing the helium gas flow rate, the breakdown voltage was decreased from 425  $V_{pk}$  to 300  $V_{pk}$  as expected, because of the dilution of the argon gas and the Penning ionization of the argon atoms by the metastable states of the helium [2].

Fig. 2(b) describes the discharge current and voltage characteristics when the plasma was generated at a fixed input power of 300 W. In this case, the helium gas addition reduced the discharge current and increased the discharge voltage. It was resulted from the lower power coupling efficiency of the helium gas compared to the argon gas [1]. Current ( $I$ ) versus voltage ( $V$ ) curves are shown in Fig. 2(c) at various helium gas flow rates. When the current was less than 1 A, the discharge current increment was relatively smaller for all the helium gas flow rates. This is due to the fact that the plasma was generated only in the side gaps and the gas breakdown with a sizable current was not initiated in the bottom gap yet [3]. But even so, it is believed that a small amount of charged particles traveled down from the side gaps to the bottom gap, acting as seed electrons for the gas breakdown in the bottom gap. It was confirmed that the capacitance calculated from the slope of the  $I$ – $V$  curve in the low current range was almost same with the capacitance of the simple vacuum parallel plate capacitor. When the plasma was generated in the bottom-discharge gap, however, a small voltage change brought about a large current increase. In addition, as more helium gas was added, the measured voltage was higher, because of the higher discharge resistance due to the lower power coupling efficiency.

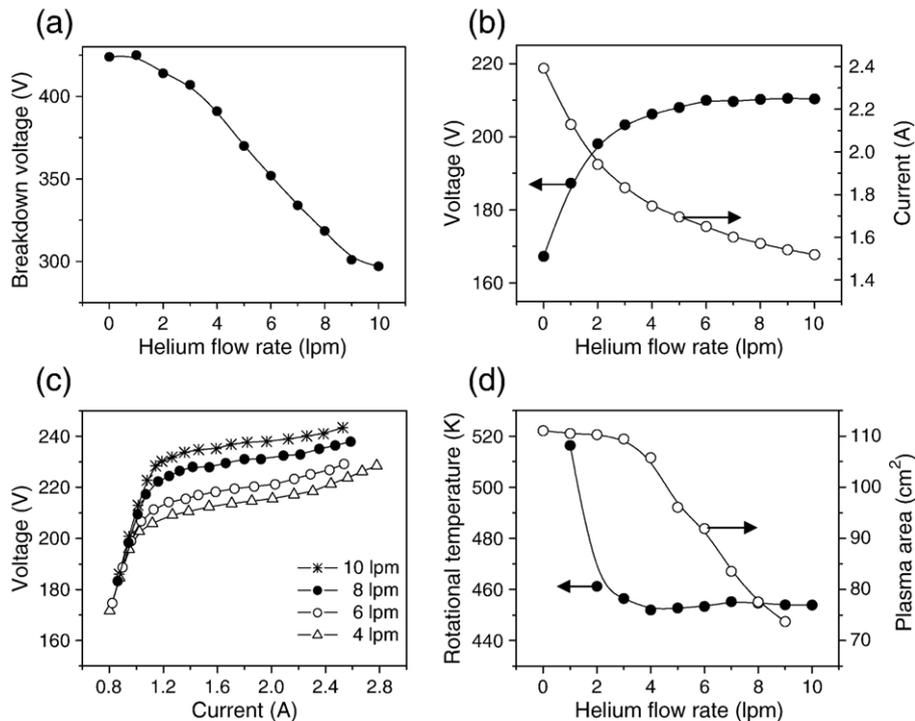


Fig. 2. (a) Breakdown voltages at different helium gas flow rates from 0 to 10 lpm with the argon gas flow fixed at 10 lpm. (b) Change of the discharge voltage and current. (c)  $I$ – $V$  curves for various helium gas flow rates. The helium gas addition brought about the discharge voltage decrease and the discharge current increase. (d) Increasing the helium gas amount resulted in the reduction of the rotational temperature and the plasma area.

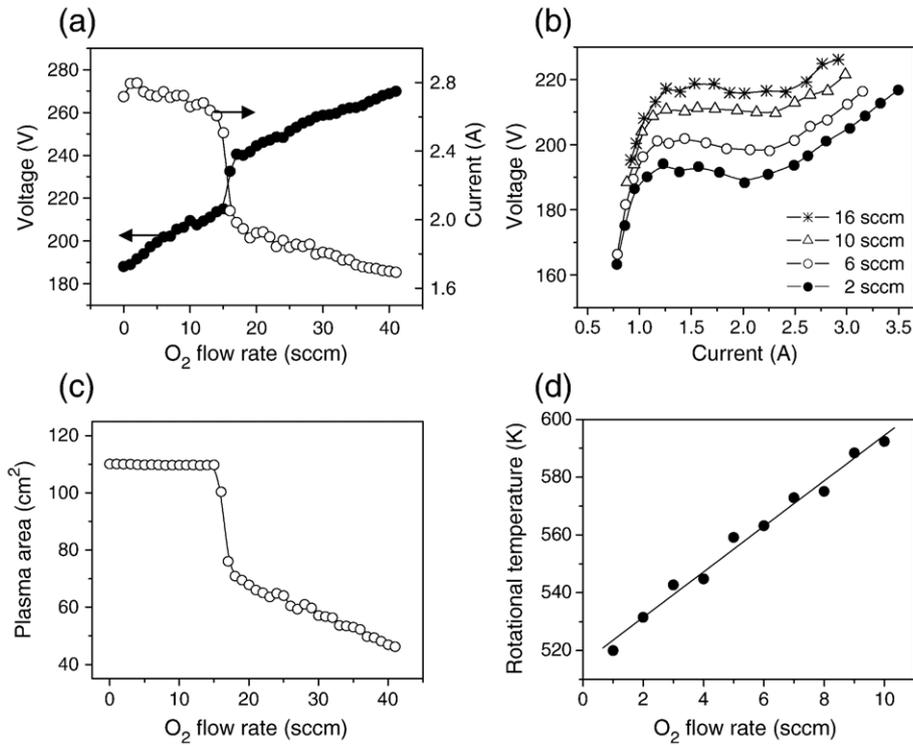


Fig. 3. (a) Change of the discharge voltage and current. (b)  $I$ - $V$  curves for various oxygen gas flow rates. (c) Dependence of the plasma area and (d) the rotational temperature on the oxygen gas flow rate.

From an application point of view, especially for the heavy particle processes, the gas temperature ( $T_{\text{gas}}$ ) is an important parameter for the atmospheric pressure plasmas. It is closely related to the rotational temperature ( $T_{\text{rot}}$ ) of the diatomic particles, present inside the atmospheric pressure plasma, since the rotational-translational relaxation is fast enough to equilibrate  $T_{\text{rot}}$  and  $T_{\text{gas}}$  [4]. Thus,  $T_{\text{gas}}$  can be indirectly known by measuring  $T_{\text{rot}}$ .  $T_{\text{rot}}$  was obtained in this experiment by analyzing the hydroxyl (OH) emission spectra, originated from the ambient air [5], and the results are shown in Fig. 2(d). As seen in the figure,  $T_{\text{rot}}$  was decreased until the helium flow rate of 3 lpm, which corresponds to the Ar:He gas ratio of 0.3. Afterwards,  $T_{\text{rot}}$  was nearly constant at 455 K. The change of the plasma area against the helium flow rate was investigated as well by analyzing the plasma images obtained at the bottom electrode made of the copper mesh and the glass plate. On the contrary, the plasma area was almost constant until the ratio of the argon and helium flow rates was less than 0.3, then, it was rapidly decreased thereafter. Fig. 2 suggests that there exists an optimal mixing ratio of argon and helium gas, guaranteeing a low gas temperature, a low breakdown voltage, and a uniform large plasma area.

### 3.2. Argon and oxygen mixing

The oxygen gas is often introduced for particular processes utilizing atmospheric pressure plasmas. For instance, the oxygen gas is used in the ashing process for removing organic materials due to the high chemical activity of the oxygen

radicals [6]. In addition, the oxygen gas is one of the best sterilization agents [7]. Since only a small amount of oxygen gas can significantly change the plasma characteristics, obtaining the fundamental knowledge and the experimental results about the oxygen plasmas is important in determining the optimum operating conditions with certain plasma characteristics.

As shown in Fig. 3(a), the addition of oxygen gas up to 40 sccm to the argon (10 lpm) plasma increased the breakdown voltage and the discharge sustaining voltage while it decreased the discharge current due to many energy loss channels, such as molecular rotations and vibrations along with the high electron affinity of the oxygen. Similar to the results of the helium gas

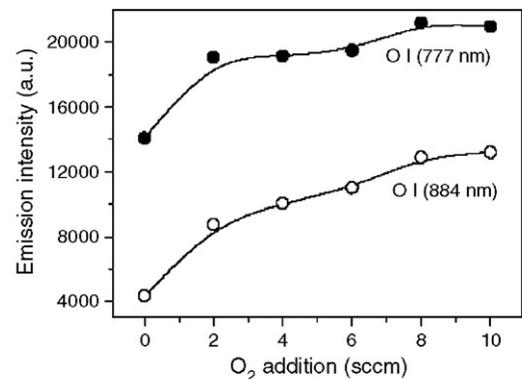


Fig. 4. Variation in the spectral intensity of the atomic oxygen lines (777 nm, 844 nm).

addition, the slopes of the  $I$ – $V$  curves for different oxygen gas flow rates are nearly overlapped in the low current range by the same reason. The discharge current drop and the discharge voltage increase with more oxygen can be explained by the plasma resistance increase, caused by the reduction in the electron density due to the high electron affinity of the oxygen.

The plasma area did not change until the oxygen gas flow rate of 15 sccm, but after that, an abrupt reduction in the plasma area was observed. Fig. 3(d) describes the oxygen effects on  $T_{\text{rot}}$  obtained from OH emission spectra, where  $T_{\text{rot}}$  was linearly increased from 520 K to 590 K as the oxygen gas was added up to 10 sccm. Fig. 4 shows a relative emission intensity of oxygen radicals with respect to the oxygen gas flow rate. As expected, the oxygen atom concentration was increased, which is good for activating chemical reactions for certain applications.

#### 4. Summary

The helium and oxygen gas mixing effects on the atmospheric pressure argon-based plasma were investigated. The helium gas addition by changing the mixing ratio with respect to the argon gas flow rate significantly reduced the discharge voltage and the rotational temperature. This may be very useful for certain atmospheric pressure plasma processes. However, too much addition of the helium gas reduced the power coupling efficiency and the plasma area. It was also observed that there existed an optimum mixing ratio, depending on the input power

level, at which a relatively low discharge voltage, a low gas temperature, a high uniformity, and a high power coupling efficiency were obtained. On the other hand, the addition of the oxygen gas in the argon-based plasma raised the breakdown voltage and the gas temperature while reducing the plasma area. These experimental results allow us to expect that the atmospheric pressure plasma characteristics can be effectively controlled by the gas mixing for applications with specific requirements.

#### Acknowledgement

This work was supported by KAIST.

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