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Discharge characteristics of a radio-frequency capacitively coupled Ar/O_2 glow discharge at atmospheric pressure

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ABSTRACT

In this study, the discharge characteristics of the developed atmospheric pressure homogeneous and cold plasma source in Ar/O_2 glow discharge driven by radio-frequency (13.56 MHz) are investigated experimentally by means of electric measurements. The electron density is estimated to be in the order of 10^{11} cm^{-3} in the abnormal discharge regime and is reduced by half in amount when the oxygen is mixed into argon plasma at oxygen-to-argon ratio of 0.3 and 0.6 vol.% at the same input power. The estimated electron temperature assumes the value of 1.4 eV in the abnormal discharge regime and the addition of oxygen to the argon plasma at the oxygen-to-argon ratio smaller than 1.0 vol.% does not alter the electron temperature appreciably.

1. Introduction

Nonequilibrium oxygen plasmas have a variety of applications, such as etching, cleaning, sterilization, and thin film deposition. In this regard, most works are carried out in vacuum environment containing several tenths to several Torrs of oxygen gas [1]. One of alternative solutions to generate oxygen plasmas is an atmospheric pressure glow discharge (APGD) at open environment with the rare gas, such as He or Ar, diluted with O_2 [2–9]. For it operates in the absence of vacuum chamber and system, the costs of equipment investment and maintenance are greatly reduced and it is ready to be used in a pattern of continuous in-line processing. It has been studied that the radio-frequency (rf) capacitively coupled plasma discharge is an important approach to generate a stable, homogeneous, arcless plasma discharge at atmospheric pressure, e.g., one-dimensional plasma source (line source) employed for large-area surface treatment [2–9]. The addition of several tenths percent of oxygen in carrier gas is capable of yielding a high concentration of reactive species, which is comparable to a pure oxygen discharge at low pressure of several Torrs. Furthermore, at high gas pressure the damage on the substrate is prevented from bombarding by accelerated ions [3]. However, when the economic argon is used as a carrier gas and the reactive gas, such as O_2 and N_2, is added to produce reactive species, several issues arise with respect to its ignition, stability, and restricted chemical and physical operation window.

In our previous work, we have developed the argon rf-APGD in a bare metal electrode configuration by introducing a dielectric strip between bare metal electrodes in its gas inlet end [9]. The allowable oxygen-to-argon ratio reaches 1.0 vol.% and the generated Ar/O_2 plasma discharge is characterized by a low gas temperature and good spatial homogeneity. And its treatment efficacy is demonstrated by the practical use in ashing of carbon black. In this work, we evaluate the important plasma parameters in the rf-APGD, the electron density n_e and the electron temperature T_e, to inspect the discharge characteristics of the Ar/O_2 plasma discharge by means of measuring its electric characteristics and establishing an equivalent electric circuit model of the plasma discharge and with use of an energy balance equation.

2. Experimental procedure

The experimental setup is illustrated in Fig. 1 where the reactor used in this study is composed of two parallel plate electrodes made of stainless steel, the surface of which for discharge is polished smoothly with sandpaper. Both electrodes are rectangular shape of 60×90 mm^2 and cooled with chilled water; the upper electrode is connected to a rf power supply (13.56 MHz) via a manual L-type matching box; the lower one is grounded. The gap spacing between the pair of electrodes is $d = 1.6$ mm. A dielectric strip of 1 mm thick, 60 mm long, and 2.3 mm wide is applied on the lower electrode near the gas inlet port to construct a gas passage with a narrow cross section of 0.6×60 mm^2. The introduction of this gas inlet passage has been shown to be important and necessary in enhancing the oxygen-to-argon ratio up to 1.0 vol.% while exciting an stable Ar/O_2 plasma discharge at atmospheric pressure. The power supply is equipped with a pair of meters for simultaneously monitoring forward power and reflected

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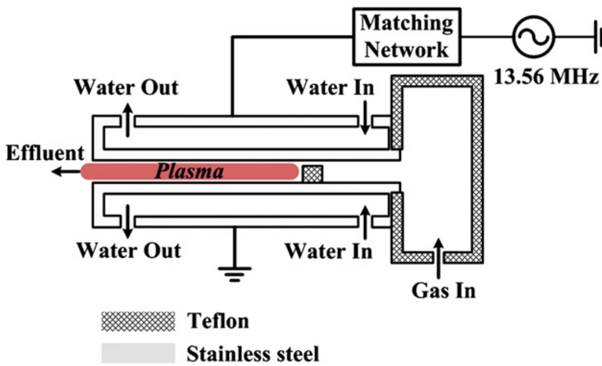

Fig. 1. Schematic diagram of the experiment setup.

power; therefore, the input power is able to be measured directly, when the complete matching is achieved by adjusting the tuning capacitor in matching box. A Pearson 4100 current probe and a high-voltage probe P6015A are connected directly to the powered electrode to capture current and voltage waveforms. In the experimental procedure, the argon gas (99.995% pure) is excited first and then the oxygen gas is added into argon discharge gradually till to a certain amount. The total gas flow rate is set at 28 l/min and the flow rate of argon and oxygen is best controlled by precise mass flow controllers, for a fluctuation of gas flow may deteriorate the discharge stability when the electronegative gas is fed. The detailed experiment setup and its discharge images could be found in [9] for a good illustration.

3. Results and discussion

In Fig. 2, the discharge current and voltage in RMS values versus the input power measured in the glow discharge at total gas flow rate of 28 l/min are shown for three different oxygen-to-argon ratios of 0.0, 0.3, and 0.6 vol.%, respectively. With increasing input power, both discharge current and voltage increase simultaneously. In Fig. 2(a), the turning points (O, P, and Q for three cases) on the curves connecting two linearly varying segments of the discharge voltage versus input power indicate the transition from normal to abnormal glow discharge. With the data in Fig. 2, a current and voltage characteristic (CVC) curve is obtained for these three different oxygen-to-argon ratios and correspondingly plotted in Fig. 3. The turning points are labeled by O, P, and Q respectively for three cases in Fig. 3. Since the discharge covers partial area of electrodes in normal glow discharge and it is difficult to define the discharge area in the current experiment setup, we pay our attention to the abnormal discharge regime to inspect the dependence of the electron temperature and the electron density on the input power and the different oxygen-to-argon ratios. In fact, it is in this abnormal discharge region that the practical applications are carried out to implement a large-area surface treatment. It is notable that the discharge current increases linearly with the applied voltage even for admixture of oxygen into argon in this abnormal discharge regime and the maximum value of the discharge current in termination of the abnormal glow discharge for these three oxygen-to-argon ratios are almost identical at the RMS value of 2.8 A, while the corresponding values of input power are quite different, as shown at 220, 340, and 380 W for the oxygen-to-argon ratios of 0.0, 0.3, and 0.6 vol.%, respectively.

The spatiotemporal structure of O_2 plasma discharge between parallel plates driven by rf of 13.56 MHz at pressure of 0.5 Torr has been investigated by using the relaxation continuum model by considering the elementary particles, O_2^+, O^+ O_2^-, O^-, electrons, and O in O_2 [10,11]. The numerical simulation shows that even in pure oxygen discharge, the dominant carriers of the total current are still electrons in the bulk plasma region due to large electron mobility, although the number density of heavy charged particles, O_2^+ and O^-, are two orders of magnitude more than the electron density. For our case of mixture of no more than 1.0 vol.% of oxygen into argon, the oxygen concentration is almost equal to that in pure oxygen discharge at pressure of several tenths Torr; however, the mean-free-path length of particles is reduced greatly due to high density of argon atoms at atmospheric pressure. And taking into account the fact that the conduction current predominates over the displacement current in the bulk of plasma and the total current continuity is conserved in the circuit, it is reasonable to assume that the electron density n_e is proportional to the amplitude value of applied current I_{rf} as follows,

$$n_e = \xi I_{rf} \tag{1}$$

in which the coefficient ξ is a complex function of input power and oxygen-to-argon ratio and trivially dependent of n_e. The structure of

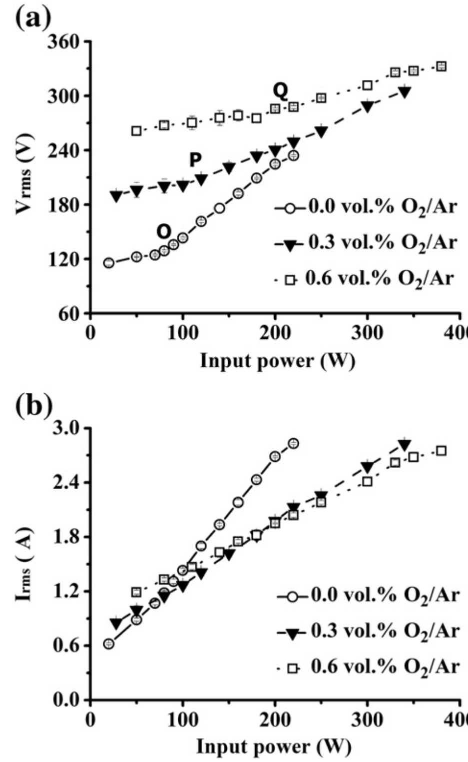

Fig. 2. Plots of (a) discharge voltage (RMS) versus input power and (b) discharge current (RMS) versus input power for oxygen-to-argon ratios of 0.0 vol.% (dot), 0.3 vol.% (triangular), and 0.6 vol.% (rectangular) at total gas flow rate of 28 l/min; and O, P, and Q are the transition points from normal to abnormal discharges.

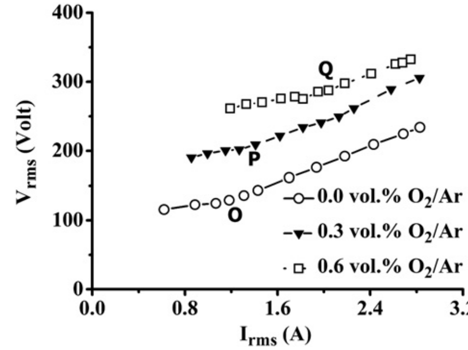

Fig. 3. Plot of discharge voltage versus discharge current in RMS for oxygen-to-argon ratios of 0.0 vol.% (dot), 0.3 vol.% (triangular), and 0.6 vol.% (rectangular) at total gas flow rate of 28 l/min and O, P, and Q are the transition points from normal to abnormal discharges.