Mode Transition in Magnetic-Pole-Enhanced Inductively Coupled Oxygen Plasma

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Abstract

Electronegative oxygen gas is used in many technological and industrial applications. In this paper, the vital parameters are investigated in two distinctive operational modes (E- & H-modes) of the “magnetic-pole-enhanced inductively coupled plasma” (MaPE-ICP) driven in oxygen with a small amount of actinometer (4% argon) for various applied powers (40-300 W) and gas pressures (15-60 mTorr). The key plasma parameters including electron density and electron temperature are determined by a Langmuir probe, whereas the dissociation fraction is estimated by actinometry which is a renowned optical emission spectroscopy technique. The determination of plasma parameters as a function of gas pressure and applied power is required to understand the discharge kinetics and upgrade the processing applications. The results obtained in the MaPE-ICP are compared with the traditional ICPs, and some interesting similarities and differences between the two schemes are reported.

Keywords

Inductively coupled plasma; Optical emission spectroscopy; Actinometry; Langmuir probe; Plasma parameters;

Introduction

Electronegative gases are used in many plasma technologies owing to their reactive nature. Oxygen is one of the simple, economical, and widely available electronegative gas which is frequently employed in various industrial and technological applications such as sterilization and decontamination of medical instruments [1], etching [2], nanofabrication [3], and modification of polymer film surfaces for tissue engineering [4]. Just like any other low temperature reactive plasma, the generation of active species in oxygen discharges (like atomic oxygen) is mainly governed by the free electrons. Therefore, the determination of plasma parameters including electron temperature, electron density, and dissociation fraction of oxygen molecules as a function of discharge control parameters (gas pressure and applied power) is very essential to understand discharge kinetics and optimize plasma processing [5].

Radio frequency (RF) driven “inductively coupled plasma” (ICP) discharges fulfil foremost requirements of reliable processing applications due to high plasma density, low contamination from reactor sputtering, easily controllable ion energy, electrodeless design, and low sheath voltage [6-9]. As compared to the traditional ICP setup, the “magnetic-pole-enhanced inductively coupled plasma” (MaPE-ICP) scheme is a new design which presents additional desirable features including relatively higher plasma density, lower electron temperature, enhanced power coupling efficiency, large-scale capacity, and excellent spatial uniformity. These attractive features warrant the efficient employment of the MaPE-ICP in state-of-the-art plasma processing applications [10, 11].

Similar to the traditional ICPs, the MaPE-ICP discharge also displays two distinctive operational modes based upon the power coupling mechanism. At lower RF powers, capacitive coupling is prevalent, whereas inductive coupling is dominant at higher RF

powers. Accordingly, the low power mode is termed as “electrostatic mode” (E-mode) and the higher power mode as “electromagnetic mode” (H-mode). The E-mode is traditionally excited by the electric field of the coil. In contrast, the H-mode is primarily produced by the induced electric field of the coil’s current. These two modes normally exhibit extremely different electrical properties and discharge characteristics [12-15].

The existence of two distinctive operational modes makes the ICPs very attractive field of study because of the fascinating features of the discharge and the underlying complicated physics. There are many theoretical and experimental studies wherein different plasma parameters are investigated in H-mode of the oxygen ICPs for various operational discharge parameters [16-19]. However, very little work has been done in E-mode or both modes simultaneously in the oxygen ICPs [20, 21]. To the best of our knowledge, there is no report in two distinctive modes of the planar coil MaPE-ICP driven in oxygen or its comparison with the traditional ICPs.

In this paper, the vital parameters are reported in two distinctive operational modes of the MaPE-ICP driven in oxygen with an actinometer (4% argon) for different RF powers and gas pressures. The fundamental plasma parameters like electron density and electron temperature are determined by a single-tip Langmuir probe. Moreover, the dissociation fraction is evaluated by actinometry method using optical emission spectroscopy. The results are compared with previous findings in the traditional ICPs and some interesting similarities and differences are observed between the two schemes. This report will be a good extension to the related studies.

The paper is organised as follows. **Section 2** illustrates materials and methods including plasma generation system along with the applied plasma diagnostic techniques. In **Section 3**, the experimental results are presented and briefly discussed. The major findings are concluded in **Section 4**.

2. Materials and Methods

2.1. Plasma Generation System

Figure 1 presents a schematic diagram of the experimental arrangement. The plasma was generated in the MaPE-ICP discharge system which has been discussed elsewhere in detail [10, 11]. The major difference between the traditional ICP and the MaPE-ICP is that a ferrite core is added in the later. The presence of ferrite core not only improves the power coupling efficiency of the RF coil by focussing the magnetic field lines inside the discharge chamber but it also increases mechanical strength of the MaPE-ICP source which allows us to reduce the thickness of dielectric window and thus the magnetic field strength inside the chamber can be further enhanced.

The details of the experimental setup have been described earlier [22]. Briefly, the MaPE-ICP source is fitted on top of a cylindrical stainless steel chamber, where the gap between the lower electrode and dielectric window is 50 mm. The system is driven by a 13.56 MHz RF power source via a “matching network”. A base pressure of approximately 10^{-5} Torr was obtained by the turbo molecular pump and then the desired gases are filled inside the plasma chamber by mass flow meters.

Figure 1. A schematic diagram of the experimental setup along with the plasma diagnostic system

2.2 Plasma Diagnostics

2.2.1 Langmuir probe

A Scientific Systems RF-compensated sigle-tip Langmuir probe diagnostic was utilized to acquire the “current and voltage” (I - V) characteristics of the discharge. To circumvent any direct current (DC) shifting in the plasma potential, this probe setup comprises a reference electrode as well as a software controlled cleaning facility of the cylindrical tip made up of tungsten wire having length of 8.5 mm and diameter of 0.2 mm. Druyvesteyn method was employed to obtain the “electron energy probability function” (EEPF), $g_p(\varepsilon)$, from the probe’s I - V characteristics, by taking the second derivative as follows [23]:

$$g_p(\varepsilon) = \frac{1}{\sqrt{\varepsilon}} f_e(\varepsilon) = \frac{2\sqrt{2m_e}}{e^3 S_p} \frac{d^2 I}{dV^2} \quad (1)$$

where ε is energy of electrons, $f_e(\varepsilon)$ is “electron energy distribution function”, e and m_e are electronic charge and mass, while V , I and S_p are probe voltage, current, and tip area respectively.

Using the measured EEPF, the electron density (n_e) and electron temperature (T_e) were acquired as follows:

$$n_e = \int_0^\infty \varepsilon^{1/2} g_p(\varepsilon) d\varepsilon \quad (2)$$

$$T_e = \frac{2}{3} \langle \varepsilon \rangle = \frac{2}{3n_e} \int_0^\infty \varepsilon^{3/2} g_p(\varepsilon) d\varepsilon \quad (3)$$

here $\langle \varepsilon \rangle$ is the average energy of electrons.

The plasma skin depth (δ_p) was determined by the following relation:

$$\delta_p = \left(\frac{m_e}{n_e e^2 \mu_0} \right)^{1/2} \quad (4)$$

here μ_0 is the permeability of free space.

2.2.2 Optical Emission Spectroscopy

A non-invasive “optical emission spectroscopy” (OES) diagnostic of the plasma was carried out using 1-meter high resolution McPherson–2061 monochromator, connected to a “photo-multiplier tube” (PMT–R928) and an auto-ranging “Pico-ammeter” (Keithley–485). To calibrate the wavelength of the monochromator, a mercury lamp was used. The spectra of emission intensity as a function of wavelength were recorded for various gas pressures and RF powers. In order to obtain the final spectra, the profiles of emission intensity were very carefully normalized to the spectral responses of the diffraction grating, optical window, and photomultiplier tube.

Actinometry is a renowned OES technique in which the addition of small amount ($\leq 5\%$) of suitably chosen inert gas (e.g. argon) into the reactive plasma allows the determination of active species concentration and molecular dissociation fraction [24]. In the present work, the dissociation fraction of oxygen molecules is evaluated by actinometry method using the following relation [25].

$$D_f = \frac{I_{844}}{I_{750}} \frac{\nu_{750}}{\nu_{844}} \frac{A_{ij}^{2P_1}}{A_{ij}^{3P}} \frac{\sum A_{ij}^{3P}}{\sum A_{ij}^{2P_1}} \frac{K_{Ar}^{dir}}{K_O^{3P}} \frac{[Ar]}{[O_2]} - \frac{K_{de}^{3P}}{K_O^{3P}} \quad (5)$$

Here $[O_2]$ and $[Ar]$ are respectively the pre-discharge densities of oxygen and argon gases. I_{750} and I_{844} are the recorded emission intensities of Ar (750.4 nm) and O (844.6 nm) lines from the discharge, ν is the transition frequency, while A_{ij} is the transition probability and $\sum A_{ij}$ is the “cumulative transition probability” from level i to level j . Besides, K_{Ar}^{dir} is the rate coefficient for excitation via electron collision of the ground state Ar to the upper level $2p_1$, whereas K_O^{3P} and K_{de}^{3P} are the rate coefficients for “electron-impact direct excitation” from the ground-state O to $O(2p^3 \ ^3P)$ and “electron-impact dissociative excitation” from the ground-state O_2 to $O(2p^3 \ ^3P)$, respectively.

3. Results and Discussion

In this experiment, the pressure range of 15 – 60 mTorr was explored for various RF powers (40 – 300 W). For all the pressures explored here, the discharge was found to operate in E-mode for 40 – 80 W and H-mode was detected for 100 – 300 W. The existence of two distinctive operational modes in the MaPE-ICP was affirmed by the abrupt variation in luminosity of the discharge and plasma parameters.

The variation of electron density (n_e) with applied RF power at a constant gas pressure of 45 mTorr is shown in Figure 2. It is noticed that n_e readily increases with RF power in both modes of the discharge as expected because the energy of electrons increases with power and thus enables them to cause further excitation/ionization in the discharge. Consequently, n_e is increased with RF power. In the beginning, the discharge generates in E-mode at lower RF powers (40 – 80 W), with much lower $n_e \sim 10^8 - 10^9 \text{ cm}^{-3}$. However, once the RF power is increased to 100 W or above, a transition to H-mode occurs where significant increase in the measured value of n_e ($\sim 10^{10} - 10^{11} \text{ cm}^{-3}$) and plasma luminosity is observed.

Figure 2. The variation of electron density (n_e) with applied RF power at **45** mTorr gas pressure

Figure 3 shows the pressure dependence of n_e in two distinctive operational modes; the E-mode (40 W) and the H-mode (300 W). In both modes, n_e is slightly increasing with gas pressure but the pressure dependence of n_e is rather weak comparing to its power dependence in view of the fact that the value of n_e is tremendously increased ($\sim 10^8 - 10^{11} \text{ cm}^{-3}$) by varying the RF power from 40 to 300 W.

Figure 3. The pressure dependence of electron density (n_e) in E- and H-modes

The observed power dependence of n_e in both modes is similar to the traditional oxygen ICP [20], but the mode transition in the MaPE-ICP is observed at relatively lower RF power. It simply reflects that the MaPE-ICP system is more economical due to its enhanced power coupling efficiency. Conversely, the pressure dependence of n_e in the H-mode is not

similar between the two schemes. It is observed that n_e does not vary with pressure in the traditional ICP but increases with pressure in the MaPE-ICP. In addition, the pressure and power dependences of n_e are almost similar to our earlier study in the argon MaPE-ICP [22]. However, the value of n_e in the oxygen MaPE-ICP is found to be lower and transition to H-mode takes place at much higher RF power than the argon MaPE-ICP. This difference can be attributed to multiple energy loss processes in the oxygen discharge such as higher collision frequencies, dissociative recombination, and negative ion formation [12].

The measured value of n_e demonstrates that certain critical electron density is essential for steady H-mode of the discharge which is in agreement with theoretical criterion of Lee *et al.* [26], where the steady operation of H-mode in the ICP is linked to plasma skin depth (δ_p) by the condition $\delta_p \leq \sqrt{2/3} R$. Here R is radius and $\sqrt{2/3} R$ is “critical dimension” of the plasma device. The critical dimension of our MaPE-ICP system is 13.8 cm. **Figure 4** presents the power dependence of δ_p at a constant pressure of 45 mTorr. The value of δ_p decreases with RF power in both modes but the remarkable decrease is observed during the E-H mode transition (80 – 100 W), where the electron density shows an abrupt jump. The value of δ_p is much higher in E-mode (40 – 80 W) than H-mode (100 – 300 W) due to the tremendous difference of n_e in two distinctive operational modes of the discharge. Besides, the value of δ_p in H-mode is smaller than critical dimension of the MaPE-ICP system as predicted by Lee *et al.* [26].

Figure 4. The variation of skin depth (δ_p) with applied RF power at a fixed pressure of 45 mTorr

The dependence of electron temperature (T_e) on applied RF power at a constant pressure of 45 mTorr is depicted in **Figure 5**. The value of T_e decreases with applied power in the E-mode and a jump is observed during E-H mode transition (80 – 100 W). The decrease in T_e is most probably induced by the increase in electron-electron collisions and stepwise ionizations which usually amplify with plasma density [27, 28]. It is also noticed that T_e increases slightly with RF power in H-mode of the discharge which is an indication of the “neutral gas heating” because an elevated gas temperature at a constant pressure results in the reduction of neutral density, which consequently increases T_e [29]. It is also interesting to mention that T_e is showing an abrupt jump during the mode transition, just like the electron density, plasma skin depth and emission intensity, indicating that T_e is a good alternative parameter for confirmation of “E-H mode transition” in the oxygen MaPE-ICP. This result is also consonant with our earlier prediction in the argon MaPE-ICP discharge [22].

Figure 5. The variation of electron temperature (T_e) with applied RF power at 45 mTorr

Figure 6. The variation of electron temperature (T_e) with gas pressure in E- and H-modes

Figure 6 shows the variation of T_e with increase in pressure for two distinctive operational modes of the MaPE-ICP. It is clear that T_e is decreasing with pressure in both modes of the discharge because the collision frequency of the electrons increases due to reduction in mean free path. Thus the mean electron energy is reduced with increase in pressure. However, T_e is higher in E-mode than H-mode, which is completely inverse to the reported behaviour in traditional oxygen ICP [20]. The higher T_e observed in E-mode is due to the fact that the measured skin depth is much higher in this mode comparing to the H-mode of the discharge. Moreover, it is also observed that T_e in the MaPE-ICP is quantitatively lower than the traditional ICPs, under similar operational discharge parameters [20, 21]. This recommends the MaPE-ICP for material processing where lower T_e is needed.

Figure 7. The power dependence of dissociation fraction (D_f) at 45 mTorr gas pressure

The power dependence of dissociation fraction (D_f) at a constant pressure of 45 mTorr is displayed in **Figure 7**. The value of D_f increases with RF power in both modes of the MaPE-ICP which is obvious because the number of energetic electrons increases with power and thus the rate of molecular dissociation in the discharge is increased. Similarly, the value of D_f also increases with gas pressure in both modes as shown in **Figure 8**. This is due to the fact that the number of parent oxygen molecules available in the gas increases with pressure. The measured value of D_f in E-mode is found to be less than 1% but increases up to 29% in H-mode of the discharge. The observed pressure and power dependences of D_f are dramatically identical to the measured n_e indicating that the generation of active species in oxygen discharge is primarily controlled by the free electrons

Figure 8. The pressure dependence of dissociation fraction (D_f) in E- and H-modes

The measured values of dissociation fraction (D_f) in the MaPE-ICP are significantly different than the reported values of D_f in the traditional ICPs. In particular, Fuller *et al.* [30] estimated $D_f \sim 1\%$ for RF power of 400 W and gas pressure of 10 mTorr by actinometry method. Corr *et al.* [20] reported $D_f \sim 10\%$ for 220 W RF power and 50 mTorr gas pressure using laser-induced fluorescence. Clearly, the measured values of D_f in the MaPE-ICP are considerably greater comparing to the traditional ICPs, under similar discharge control parameters. This indicates that the MaPE-ICP driven in electronegative oxygen gas can be extremely useful for processing applications due to its much better efficiency of active species generation.

4. Conclusions

In this study, the key plasma parameters are explored in two distinctive operational modes (E- & H-modes) of the MaPE-ICP driven in oxygen for different discharge control parameters. It is noticed that the value of electron density upraises with RF power and gas pressure in both operational modes of the discharge, but the remarkable increase is observed during “E-H mode transition”. The value of electron temperature decreases in E-mode and slightly increases in H-mode with increasing RF power, however, it reduces with pressure in both modes. The dissociation fraction is increased with gas pressure and RF power in both operational modes of the MaPE-ICP and the trend is dramatically identical to the measured electron density indicating that the generation of active species is primarily controlled by the free electrons. The trends and values of measured plasma parameters in the MaPE-ICP show some interesting similarities and differences than the traditional oxygen ICPs. The mode transition in the MaPE-ICP occurs at relatively lower RF power suggesting that it is a more economical plasma system. In contrast to the traditional ICP, the electron temperature in H-mode is found to be lower than E-mode of the MaPE-ICP. Besides, the measured electron temperature is also qualitatively lower than the traditional ICP indicating that the MaPE-ICP is very promising for plasma applications where lower electron temperature is needed. However, the remarkable difference is observed in the values of dissociation fraction of oxygen molecules, which are much higher in H-mode of the MaPE-ICP. These features highly recommend the use of MaPE-ICP source for modern high-tech plasma applications.

Acknowledgements

This research is supported by the Plasma Physics Project for “Gomal University D.I.Khan” under “Higher Education Commission” (HEC) of Pakistan. Author A. W. Khan would like to appreciate the financial assistance of HEC during his Ph.D. studies.

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