Nonequilibrium Atmospheric Pressure Ar/O\textsubscript{2} Plasma Jet: Properties and Application to Surface Cleaning

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Abstract In this study an atmospheric pressure Ar/O\textsubscript{2} plasma jet is generated to study the effects of applied voltage and gas flux rate to the behavior of discharge and the metal surface cleaning. The increase in applied voltage leads to increases of the root mean square (rms) current, the input power and the gas temperature. Furthermore, the optical emission spectra show that the emission intensities of metastable argon and atomic oxygen increase with increasing applied voltage. However, the increase in gas flux rate leads to a reduction of the rms current, the input power and the gas temperature. Furthermore, the emission intensities of metastable argon and atomic oxygen decrease when gas flux rate increases. Contact angles are measured to estimate the cleaning performance, and the results show that the increase of applied voltage can improve the cleaning performance. Nevertheless, the increase of gas flux rate cannot improve the cleaning performance. Contact angles are compared for different input powers and gas flux rates to search for a better understanding of the major mechanism for surface cleaning by plasma jets.

Keywords: nonequilibrium plasma jet, gas flux rate, plasma cleaning, the temperature of gas

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(Some figures may appear in colour only in the online journal)

1 Introduction

Metal surfaces are generally coated with protective lubricants or oils. The removal of these organic coatings is a preliminary stage of any further surface treatment [1]. Usually, metal surfaces are primarily cleaned by wet chemistry. However, aqueous cleaning, in many cases, will bring environmental, safety, and health problems. Moreover, there does not seem to be an efficient way to dispose of the waste [2]. As an alternative cleaning method, low-pressure plasma is cheaper, more effective, and environmentally friendly [3–7]. Nevertheless, the use of these low-pressure plasmas need vacuum apparatus and installations, which brings expensive operating costing and poor efficiency. Therefore, the use of plasmas generated at atmospheric pressure has attracted considerable attention in process of surface cleaning due to hypothermal gaseous temperature, dismiss of vacuum installation, generation of sufficient reactive species [8–12]. A plasma jet is an example of an atmospheric pressure plasma, which can be produced in open space rather than in conned discharge gaps, which is characterized by flexibility, compact, and efficiency [13–16]. Owing to these attractive features, the atmospheric pressure plasma jet is applied to the metal surface cleaning [17,18]. In previous research, various plasma jets have been produced by microwave, radio frequency, pulse [19–22], and so on. However, these plasma jet sources result in an expensive budget due to the plasma generator.

This article reports a modified device which comprises double cylindrical quartz glass tubes and two electrodes. The modified device can work under AC voltage for realizing a high efficiency on plasma cleaning with inexpensive costing. We will focus on showing the change in electrical properties and water contact angle with discharge voltage and gas flux rate, and searching for a better understanding of the major mechanism for surface cleaning by plasma jets.

2 Experimental installation

Fig. 1 shows a schematic diagram of the modified plasma apparatus for the atmosphere plasma jet. As shown in Fig. 1, the apparatus is composed of double quartz glass tubes (an inside and an outer quartz tubes), two powered electrodes (a stainless injection needle and one aluminum foil ring) and the ground electrode (the other aluminum foil which is tightly wrapped
around the quartz tube). The metal surface sprayed with lubricants (a compound of various types of alkanes, i.e., \( C_m H_n \)) is placed at a distance of 1.5 cm from outer quartz glass tubes nozzle in the process of cleaning. The apparatus is designed to help enable it to process the surface of any shape and size. Pure argon gas is introduced into the inside quartz glass tube via the stainless needle by mass flux controller at flux rate of 2 slm, 3 slm, 4 slm and 5 slm, respectively. The 2% additive \( O_2 \) is forced into the outer quartz tube through a side channel. Both of the powered electrodes are supplied by alternating current power at voltage of 6.5 kV, 7 kV, 7.5 kV, 8 kV, 8.5 kV and 9 kV, respectively.

Fig. 1 Sketch of the experimental apparatus

In this study, the waveforms of the voltage and the current are measured by digital oscilloscope (Tektronix DPO 4104) with high-voltage probe (P6015 A) and current probe (Pearson 4100), respectively. The power consumed during the discharge is computed according to Q-V Lissajous figures by a digital oscilloscope. Gaseous temperature is measured by a fiber thermometer (FISO FOT-L-SD) with an accuracy of ±5 k. The electronically excited species generated are obtained using optical emission spectroscopy (Acton 2500i). The cleaning performance in the lubricants is investigated by the contact angle measurement system (Kino SL200B) after plasma cleaning for 20 s.

3 Results and discussion

3.1 Electrical properties

The typical waveforms of applied voltage with the blue line and the current in the red line are measured by digital oscilloscope (Tektronix DPO 4104) with high-voltage probe (P6015 A) and current probe (Pearson 4100), respectively. The power consumed during the discharge is computed according to Q-V Lissajous figures by a digital oscilloscope. Gaseous temperature is measured by a fiber thermometer (FISO FOT-L-SD) with an accuracy of ±5 k. The electronically excited species generated are obtained using optical emission spectroscopy (Acton 2500i). The cleaning performance in the lubricants is investigated by the contact angle measurement system (Kino SL200B) after plasma cleaning for 20 s.

The energy consumed in the discharges can be obtained by using Q-V Lissajous figures. As shown in the inset of Fig. 4, the typical Q-V Lissajous figure under applied voltage of 7 kV at the gas flux rate of 5 slm with the discharge frequency of 30 kHz is demonstrated, the area of which is the energy consumed in a period of discharge. After getting the energy dissipated one period, the power dissipated in a period of discharge can be calculated according to

\[
P = \frac{W}{T} = W f. \tag{1}\]

The energy consumed during the discharge with variation of the applied voltage at different flux rates of 2 slm, 3 slm, 4 slm, and 5 slm is simultaneously shown in Fig. 4. The results show that the consumed energy increases when applied voltage increases at the same flux rate. This is easy to understand because a higher voltage can induce a stronger discharge, as shown in Fig. 3, which can lead to more consumed energy. Meanwhile, at the same applied voltage the consumed energy decreases as a function of the gas flux rate, as shown in Fig. 4. The decrease in consumed energy can be related to the convective heat transport by gas flow [23]. More Joule heat losses because of the increase in the gas flux rate can make the gas temperature decrease, and cause the local gas intensity increase according to equation of gas state, which in turns lead to the reduction of increasing applied voltage from 6.5 kV to 9 kV. However, the rms current decreases with increasing gas flux rate from 2 lpm to 5 lpm at the same applied voltage.

Fig. 2 The typical waveforms of applied voltage with the blue line and the current with the red line for Ar (5 slm) with an addition of 2% \( O_2 \) at peak of applied voltage 7 kV.

Fig. 3 The plot of the rms current versus applied voltage at serial of Ar flux rate 2 slm, 3 slm, 4 slm and 5 slm.
electric field. So very naturally the electrical discharge grows weak when the gas flux rate increases, as shown in Fig. 3, and the consumed energy will also decrease when the gas flux rate increases, as shown in Fig. 4.

**Fig. 4** Input power versus discharge voltage at Ar flux rate of 2 slm, 3 slm, 4 slm and 5 slm

Through the above analysis, it is crucial to study the variation of gas temperature when the gas flux rate increases. In this study, the gas temperature is measured directly by use of optical fiber temperature sensor, as shown in Fig. 5. It is found that the gaseous temperature increases when the applied voltage increases at the same gas flux rate, while it decreases when the gas flux rate increases at the same applied voltage, which indicates that the gas flux rate has a big influence on the gas temperature, and has a further influence on the discharge current and the input power.

**Fig. 5** Gas temperature versus applied voltage at serial of Ar flux rate 2 slm, 3 slm, 4 slm and 5 slm

### 3.2 Estimation of metastable argon density

Fig. 6 presents the typical optical emission spectrum of the Ar/O₂ plasma jets in a range of 700-860 nm under applied voltage of 7 kV at the gas flux rate of 5 slm. The intensities of Ar⁺ (763.5 nm) in optical emission spectrum are recorded to qualitatively study the change of the metastable atoms’ density as a function of the applied voltage at different gas flux rates (see Fig. 7). It is found that at the same gas flux rate, the intensities of Ar⁺ (763.5 nm) increase when the applied voltage increases. As mentioned above, both the input power and discharge current increase when the applied voltage increases, which would help the atom of Ar to get more chances of collision, and then more Ar⁺ can be generated, as shown in Fig. 7. It is also shown in Fig. 7 that under the same applied voltage, the intensities of Ar⁺ decrease rather than increase with increase of the gas flux rate. The reason for the reduction in the intensities of Ar⁺ is mainly due to the reduction in the discharge current and input power when the gas flux rate increases. The decrease of the discharge current and input power reduces the collision chance of the atom of Ar; as a result, the intensities of Ar⁺ decrease.

**Fig. 6** The example of optical emission spectrum (700-860 nm) of the plasma jet for Ar (5 slm) with addition of 2% O₂ at peak of applied voltage 7 kV

**Fig. 7** Emission intensity of metastable atoms versus applied voltage at Ar flux rate of 2 slm, 3 slm, 4 slm and 5 slm

In order to improve cleaning effect, 2% additive O₂ is introduced into the discharge gas. The addition of O₂ to Ar can generate some reactive oxygen species, which are essential to metal surface cleaning. As shown in Fig. 8, the intensities of atomic oxygen O (777.4 nm and 844.6 nm) in optical emission spectrum are recorded to qualitatively study the change of atomic oxygen as a function of the applied voltage at different flux rate. Intensities of O (777.4 nm)
and O (844.6 nm) increase as applied voltage increased from 6.5 kV to 9 kV for the same gas flow rate. Based on the above results, the discharge current and input power increase with the increasing applied voltage, consequently, the inelastic electron-impact collisions with oxygen molecule can happen more frequently, as follows:

\[ e + O_2 \rightarrow O + O^- \]  
\[ e + O_2 \rightarrow O + O + e \]

consequently, more atomic oxygen (777.4 nm and 844.6 nm) can be generated with increasing applied voltage at the same gas flux rate. Simultaneously, the intensities of Ar\(^*\) increase with the increase of the applied voltage, as a result, more atomic oxygen can be generated, as follows:

\[ Ar^* + O_2 \rightarrow O + O^* + Ar \] 
\[ Ar^* + O \rightarrow O^* + Ar \]

It is also shown in Fig. 8 that for a certain applied voltage, intensities of O (777.4 nm) and O (844.6 nm) decrease when the gas flux rate increases, which is in accordance with the reduction in discharge current and input power with increase of the gas flux rate. The decrease of discharge current and input power causes the occurrence of reactions 1 and 2 to be greatly reduced, and then the generation of O (777.4 nm) and O (844.6 nm) suffers a corresponding decrease. As mentioned above, Ar\(^*\) also contributes to the generation of O (777.4 nm) and O (844.6 nm) (reactions 3 and 4); however, the intensities of Ar\(^*\) decrease when the gas flux rate increases. Consequently, the intensities of O (777.4 nm) and O (844.6 nm) further decrease when the gas flux rate increases.

3.3 Contact angle measurement

The lubricant on a metal surface is a compound of various hydrocarbons and non-hydrocarbons, primarily including cycloalkanes, alkanes, aromatic hydrocarbon, and nitrogenous, oxygenated organic compounds. In metal surface cleaning by the Ar/O\(_2\) plasma jet, it is believed that electrons produced in the Ar/O\(_2\) plasma jet have high energy, and they can drive metastable atoms to be produced; therefore, the high-energy metastable atoms can also physically break directly or partly the bonds of hydrocarbon and non-hydrocarbons. Furthermore, the monomer component produced during the process described above chemically react with oxygen atoms to form volatile gas, such as CO\(_2\) and H\(_2\)O as follows:

\[ C_mH_n + O \rightarrow CO_2 + H_2O \]

Consequently, oil contamination can be cleaned away from the surface by the plasma jet. The contact angles, which provide information on the hydrophilicity of the surface, are an essential techniques to evaluate the surface cleanliness and performance of plasma cleaning [24]. In this paper, the contact angles on the surface are measured after Ar/O\(_2\) plasma jet treatment to assess the effect of plasma cleaning, as shown in Fig. 9. It is found in Fig. 9 that at a certain gas flux rate, the contact angles decrease sharply with the increase of applied voltage, which indicates that increased of applied voltage can effectively improve the effect of cleaning. By contrasting the data among Fig. 4, Fig. 7, Figs. 8 and 9, it can be found that when applied voltage increases, the input power increases simultaneously, and then higher power can produce more Ar\(^*\) and more atomic oxygen. As a result, more Ar\(^*\) with high energy can physically break more molecule bonds of the lubricants. In the meantime, more atomic oxygen react chemically with the monomer units produced during the collision, so more lubricants can be removed with an increase of the applied voltage. Through the above analysis, it can be found that Ar\(^*\) and O play a vital role in plasma cleaning.
then Ar* and O also decrease, which means that the physical collision and chemical reaction with the lubricant on the surface cannot occur more frequently. As a result, the contact angles increase when the gas flux rate increases. As the gas flux rate increases, the Ar/O$_2$ plasma jet cannot get a better cleaning performance. So it is believed that the Ar/O$_2$ plasma jet with higher applied voltage and lower gas flux rate can improve cleaning performance.

4 Conclusion

This study indicates that the plasma jets with different applied voltages and gas flows exhibit different cleaning performances. This paper briefly analyzed the factors which affect cleaning effect according to the experimental results. The results of this study show that increasing applied voltage can lead to the increase in the discharge current and input power, and finally cause intensities of O and Ar* to increase. This then improves the chances of chemical reactions and physically collision with the lubricants on the surface and, consequently, has a better cleaning effect. However, with increasing gas flux, the decrease of the discharge current and input power finally leads to a decrease of the intensities of O and Ar*, which reduces the chances of chemical reactions and physical collision with the lubricants on surface. Consequently, a plasma jet with a higher gas flux cannot get a good cleaning effect. The results also indicate that O and Ar* are key factors that produce a cleaning effect in plasma cleaning.

References


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