

行政院國家科學委員會補助專題研究計畫簡易報告

常壓射頻電容耦合式電漿束之研究(三年)

Development and Characterization of an Atmospheric-pressure
Capacitively Coupled Plasma Jet (3 Year)

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計畫編號：NSC-96-2628-E-009-134-MY3

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計畫主持人：吳宗信

計畫參與人員：江明鴻 林逸民 國立交通大學機械工程學系

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行政院國家科學委員會專題研究計畫期中報告

常壓射頻電容耦合式電漿束之研究(第二年)

Development and Characterization of an Atmospheric-pressure Capacitively Coupled Plasma Jet (Second Year)

計畫編號：NSC-96-2628-E-009-134-MY3

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主持人：吳宗信 國立交通大學機械工程學系

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中文摘要

在過去十年中，大氣電漿逐漸替代低壓電漿作為材料表面處理的應用。其重要性是由於它相對低廉的設備費用，及更容易的操作條件和在更高的靈活性在工業上的應用。其中已經發展的大氣電漿源之中，又以非熱平衡『常壓射頻電容耦合式電漿束』（簡稱常壓電漿束）引起了最多的注意。主要乃因為其應用範圍相當廣闊，包括半導體製程光阻去除，清潔，消毒，表面改質和薄膜沈積等等，相當廣泛。

第二年，在實驗部分中，早期焦點放在改變各種測試條件下量測其電漿束出口處電漿相關的流體，化學及物理性質；後期再將焦點放在加入氧氣到原先氬與純氬及純氬比較對電漿性質的影響。並分別把氧氣加入常氣電漿束之機制，於出口區域延長自由基氧原子存活距離及增加其成份作為三個重要研究任務。除了重複第一年的量測外，在常氣電漿束出口位置針對不同氣體電漿使用光譜儀(OES)量測自由基原子種類及強度。

關鍵字：常壓電漿束，氬氣，氧氣，流體數值模組。

Abstract

In this proposed three-year project, we intend to develop and systematically characterize a non-thermal atmospheric-pressure capacitively coupled plasma jet (APPJ), which is able to generate reactive atoms or radicals of practical interest. Interested radicals may include oxygen- (electronegative) and nitrogen-based (electropositive) species, which have tremendous practical applications. Research approaches include both experiments and modelings.

In the second year, in the *experimental part* in the earlier stage we will focus on examining the postglow fluid dynamics, plasma physics and chemistry by varying all kinds of test conditions. In

later stage of this period, we will shift our focus to study the effects of the discharge by pure helium, pure argon and adding oxygen into the helium. Mechanism of introducing oxygen into the discharge, how to extend the downstream distance of the “alived” reactive oxygen atoms, and increase the oxygen atoms are the three most important tasks. In addition to all the measurements in the first year, distribution of the species concentrations, especially the oxygen atoms, shall be measured in the post-glow region using the OES system.

Keywords: atmospheric-pressure plasma jet, helium, argon, fluid modeling.

I. INTRODUCTION

It is well accepted that atmospheric-pressure plasmas do not have several inherited disadvantages as low-pressure plasmas do [Schutze *et al.*, 1998; Becker *et al.*, 2004; Moravej and Hicks, 2006]. Thus, there exists a great interest in atmospheric-pressure plasmas, as can be seen from several international conferences in the past five years [2000-2005]. Types of atmospheric-pressure plasmas [Schutze *et al.*, 1998 and references cited therein] include transferred arc and plasma torch, corona discharge, dielectric barrier discharge, and atmospheric-pressure plasma jet.

Plasma torch is one kind of thermal equilibrium plasmas, in which the temperature (10,000-20,000 K) of electrons, ions and neutrals are approximately the same, while the other three kinds are all non-thermal (“cold”) non-equilibrium plasmas, in which the electron temperature (10^4 - 10^5 K) is much higher than those of ions and neutrals (~300K). The former is limited in applications due to its high temperature, while the latter are more widely used due to its low temperature.

In corona discharges, plasmas are generated around the tip a needle within ~0.5 mm, where the electric field is very large and highly non-uniform. The corona discharge can be considered as a Townsend discharge or a negative glow discharge

depending upon the field and potential distribution. Micro-discharges and streamers are generated in the plasma. Operating voltage is in the range of 10-50 kV.

In dielectric barrier discharge, which is often called “silent discharge”, it consists of two electrodes with at least one is coated with dielectrics. The discharge is maintained through a series of micro-discharges, lasting for 10-100 ns, and randomly distributed in space and time, which are often called “streamers”. Operating voltage in the range of 5-25 kV. Two major drawbacks are shared for the corona discharge and the dielectric barrier discharge. One is the high breakdown voltage and the other is the difficulty of maintaining a uniform glow like low-pressure plasma.

In atmospheric-pressure plasma jet, it often consists of two coaxial electrodes through which a mixture of gases flow. By applying 13.56 MHz RF power to the central electrode at a voltage in the range of several hundred volts, the gas discharge can be ignited and maintained over a fairly large range of conditions. Normally the atmospheric-pressure plasma jet is operated in the “abnormal glow” regime, in which the voltage increases with increasing current, while the low-pressure plasma is often operated in the “normal glow” regime, in which the voltage remains constant with increasing current [Schutze *et al.*, 1998].

In this report, we describe the progress of the project in the second year, which setting up related experimental facilities and instrumentation for preliminary measurements. We focus to study the effects of the discharge by pure helium, pure argon and adding oxygen into the helium. Mechanism of introducing oxygen into the discharge, how to extend the downstream distance of the “alived” reactive oxygen atoms, and increase the oxygen atoms are the three most important tasks.

II. EXPERIMENTAL METHOD

Fig. 1 illustrates the sketch of a co-axial APPJ, which is powered by a RF source (13.56 MHz) with a matching box between the RF power and APPJ to reduce the power reflection from the APPJ. The APPJ consists of two 60 mm long concentric electrodes made of metal that may be stainless steel or aluminum. The inner diameter of the outer grounded electrode is 5 mm without water cooling, while the inner electrode is directly connected to the RF power source. Gap spacing between two electrodes is 1 mm and is adjustable if necessary. Pure helium, argon, and adding oxygen into the helium is passed through the annular space between the electrodes.

Basic discharge parameters, such as the input RF power, RF voltage, RF current and phase angle, are measured by an RF impedance analyzer (MKS VI-probe-4100). The voltage and current wave form of the discharge were investigated by a current probe (Tektronix TCP312) and a high-voltage probe (Tektronix P6015A) through a digital oscilloscope

(Tektronix TDS1012B). The optical emission spectral intensity of the APPJ is measured using a monochromator (PI Acton SP 2500) with a Photomultiplier tube (Hamamatsu R928). Typical test conditions include RF power in the range of 2-70 W and calculated average velocity between the annular space in the range of 13-26.5 m/s, which corresponds to the Reynolds numbers (based on the gap distance) in the range of 190-2000. Surface hydrophilicity of the polypropylene (PP) film surface was measured using the contact angle for wetting of the glass surface was measured using a contact angle machine (KRÜSS GH100). In addition, the gas temperature (measured by K-type thermocouple) at the position of $z=4$ mm is generally low in the range of 50-70 °C for flow rate in the range of 10-20 slm at 16 W of various feed gas, which is safe for PP film surface cleaning.

III. RESULTS AND DISCUSSIONS

Preliminary results of the experiments is briefly summary as follows.

Experiments

The images of various feed gas in glow and post-glow region

Fig. 2 shows the typical image of glow and post-discharge region for He, He+0.3% O₂, and argon discharges at the condition of 10 slm and 16 W. For pure argon discharge, the light intensity looks lighter than pure helium and He+ 0.3% O₂ discharge. The helium becomes darker with 0.3% of traced oxygen addition. These color changes can be clearly explained from the OES measurement, as typically shown in Fig. 9-11, which will described in detail next.

The breakdown process of various feed gas

Fig. 3 (a)-(c) shows breakdown process of co-axial APPJ for helium, He+0.3% O₂, and argon feed gas, respectively. The breakdown voltage of helium is generally larger than that of argon at the same current as expected. Argon plasma operate region is shorter than helium plasma, and He+0.3% O₂ plasma operate region is longer than helium and argon plasma.

Effects of electrode material on discharge characteristics

The delivered power to the load, i.e. the delivered power, is equal to $(P_{forward} - P_{reflected})$. Corresponding formula for calculating the electrical properties of the discharge are written as follows:

$$\text{The delivered power: } P_{deliv} = V \cdot I \cdot \cos\theta \quad (1)$$

$$\text{The impedance: } Z = V/I \quad (2)$$

$$\text{The resistance: } R (real) = \cos\theta \cdot V/I \quad (3)$$

$$\text{The reactance: } X (image) = \sin\theta \cdot V/I \quad (4)$$

$$\text{The phase angle: } \theta = \tan^{-1}(X/R) \quad (5)$$

Fig. 4, 5, 6 and 7 shows the θ - P curve, R - P , X - P and Z - P curve for various feed gas discharge, respectively.

Gas temperature distribution in the APPJ

Fig. 8 shows temperature distribution of the discharged gas with 10 and 20 slm of He, He+0.3% O₂ and Ar at delivered power of 16 W. Fig. 8 shows temperature distribution of the discharged gas with 10 and 20 slm of He, He+0.3% O₂ and Ar at delivered power of 16 W. The temperature of gas plasma is decrease fathering downstream and increasing flow rate increase.

Optical emission spectrum distribution

Fig. 9 shows a typical optical emission spectra in the range 200-900 nm for a helium discharge with 10 slm and the delivered power of 16 W. For the He discharge, the NO lines appear at 226, 236, 247, 259 and 271 nm, respectively, OH line at 309 nm, N₂ at 337 and 618 nm, N₂⁺ at 391 nm, N I at 472 and 517 nm, O II line at 672 nm, O III line at 741 nm and O line at 777 nm were detected. Some important reactions for helium along with the rates are shown below [shakir *et. al*, 2006]:

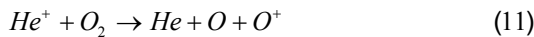
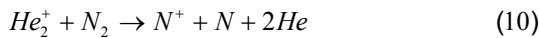
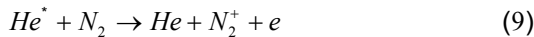
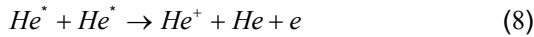
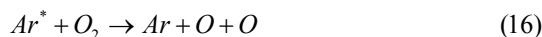


Fig. 10 shows a typical optical emission spectra in the range 200-900 nm for a He+0.3% O₂ discharge with 10 slm and the delivered power of 16 W. For the He+0.3% O₂ discharge, O III line at 760 nm and O line at 777 nm were detected. The oxygen plasma is a typical negative plasma that He+0.3% O₂ spectrum intensity is darker than pure helium gas discharges. Fig. 11 shows a typical optical emission spectra in the range 200-900 nm for an argon discharge with 10 slm and the delivered power of 16 W. For the He discharge, the NO lines appear at 226, 236, 247 and 258 nm, respectively, OH line at 309 nm, N₂ at 336 nm, N I at 472 and 517 nm, O II line at 672 and 776 nm, O III line at 741 nm and Ar line at 710 nm were detected. Some important reactions for argon along with the rates are shown below [shakir *et. al*, 2006]:



We attribute this to the enhanced UV emission, which in turn was absorbed by the ozone and

generates more oxygen radicals. More detailed mechanism will be provided during the conference meeting.

PP film surface cleaning

Fig. 12 shows the contact angles of PP film after treatment of the co-axial APPJ using various working gases with stationary (time: 5~20 sec) stage. For a stationary PP film, it becomes highly hydrophilic with contact angle of 15~20 degrees after 5 seconds of Ar co-axial APPJ treatment. We attribute this to the enhanced UV emission, which in turn was absorbed by the ozone and generates more oxygen radicals (O III 741 nm, O II 672 nm, O I 777 nm). More detailed mechanism will be study with measurement reactive neutral gas (O₃, NO, NO₂) concentration at post-glow region in future.

IV. CONCLUSIONS AND FUTURE WORKS

In the current report, we have described the progress of our project in experiments and simulation of APPJ using helium and argon gases. Important conclusions are summarized as follows:

1. A test rig has been set up and verified for operation of APPJ using RF power.
2. Preliminary measurements of electrical properties, velocity fields, and temperature distribution have been made. Several interesting electrical properties have been found for oxygen addition into helium, which requires further investigation to understand the underlying physics.
3. A parallelized 2D fluid modeling code using finite-difference method has been developed and tested.
4. A preliminary simulation using concentric APPJ with RF power is conducted. Results are reasonably explained.

Future work includes detailed measurements of plasma properties within the APPJ at various operating conditions. In addition, the development of the N-S equation solver using cell-centered finite-volume method is currently in progress and will be combined with the present fluid modeling code to fully simulate the APPJ, which the project aims.

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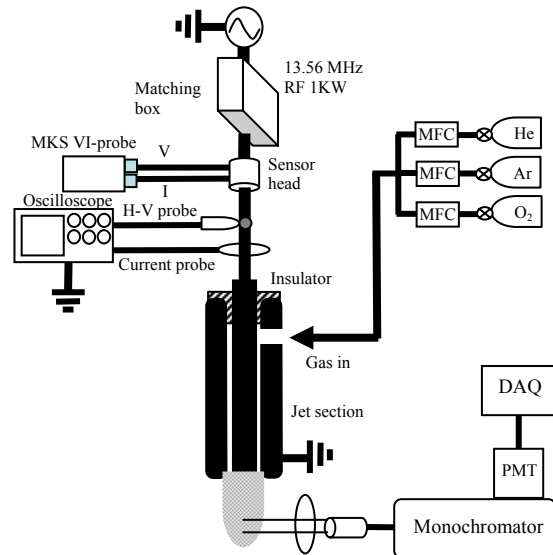


Fig. 1 Sketch of an OES for measuring plasma spectrum intensity distributions in the post-glow region of a co-axial APPJ.

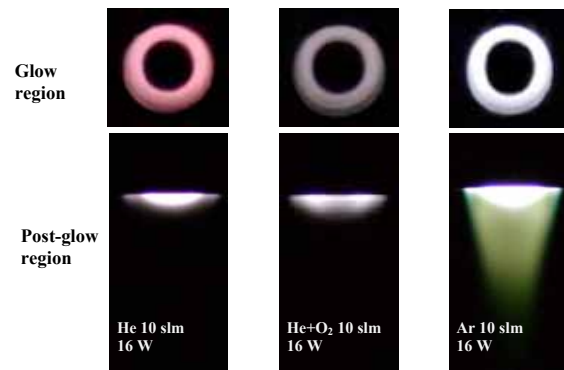
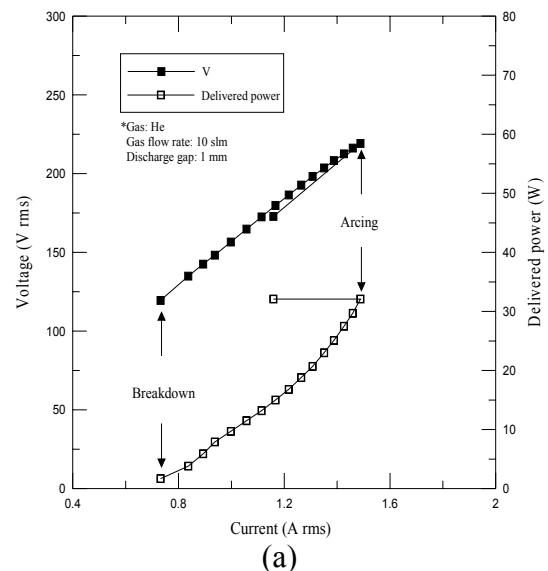


Fig. 2 Images of plasma glow and post-glow region for He, He+0.3% O₂, and argon discharges. Other discharge parameters are: gas flow rate=10 slm, delivered power=16 W, and gap=1 mm.



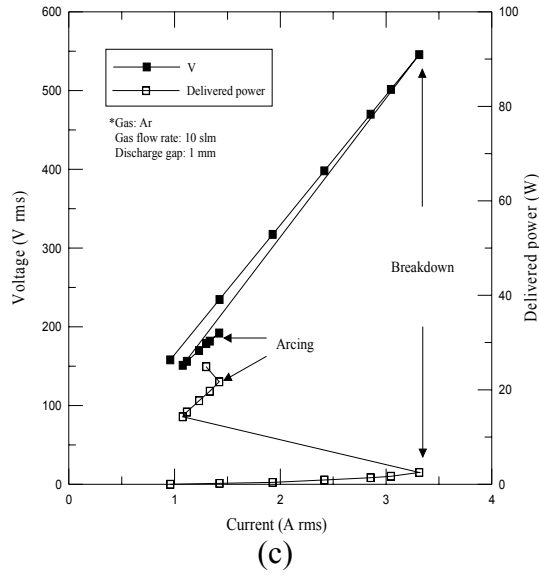
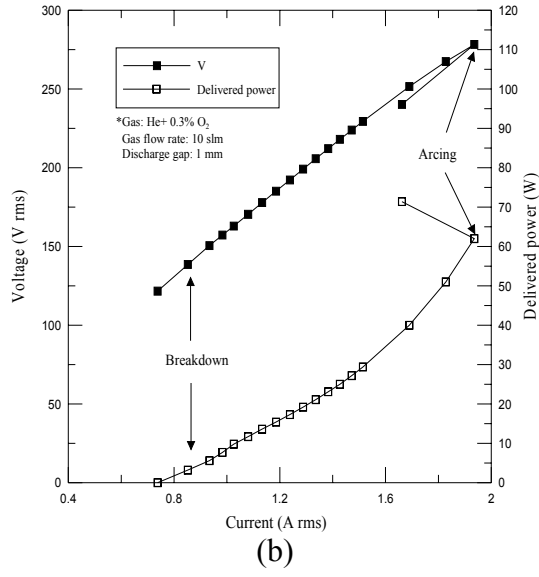


Fig. 3 The breakdown process of co-axial APPJ for various feed gas. (a) helium, (b) He+0.3% O₂ and (c) argon.

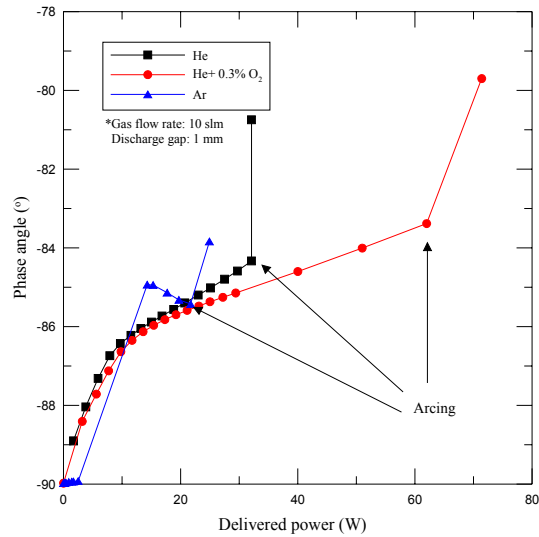


Fig. 4 Phase angle as a function of delivered power for He, He+ 0.3% O₂, and argon discharges. Other discharge parameters are: gas flow rate=10 slm, and gap=1 mm.

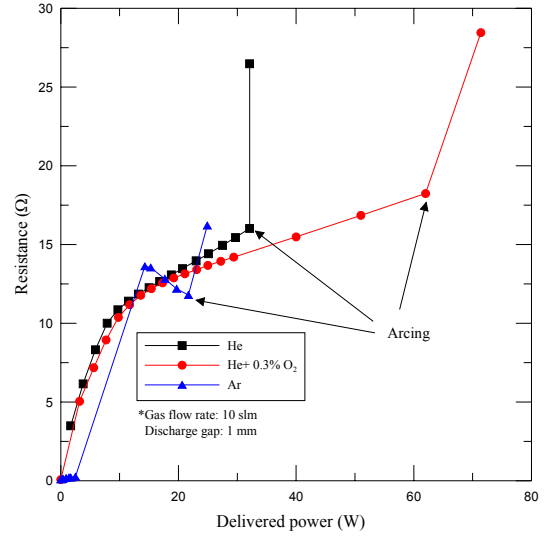


Fig. 5 Resistance as a function of delivered power He, He+ 0.3% O₂ and argon discharges. Other discharge parameters are: gas flow rate=10 slm, and gap=1 mm.

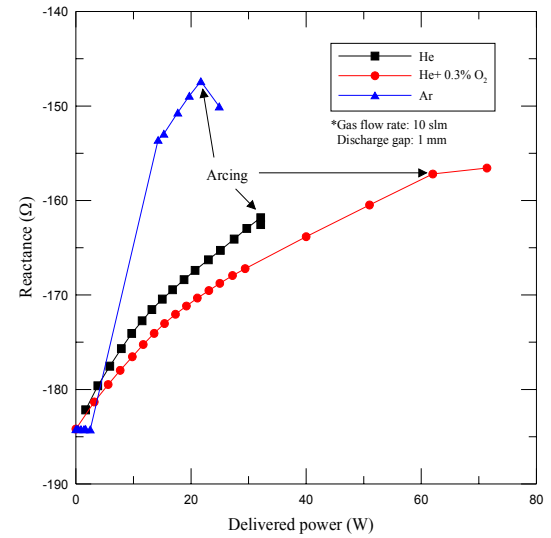


Fig. 6 Reactance as a function of delivered power He, He+0.3% O₂ and argon discharges. Other discharge parameters are: gas flow rate=10 slm, and gap=1 mm.

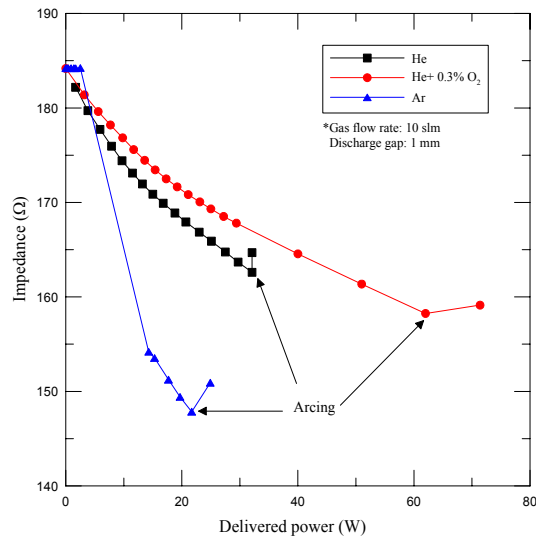


Fig. 7 Impedance as a function of delivered power He, He+ 0.3% O₂ and argon discharges. Other discharge parameters are: gas flow rate=10 slm, and gap=1 mm.

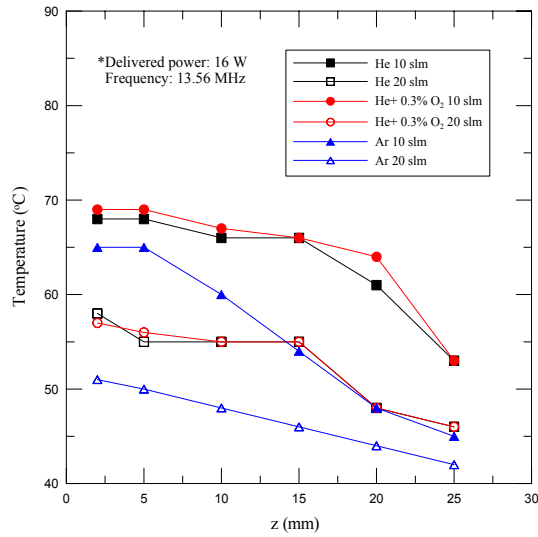


Fig. 8 Temperature distributions in the post-glow jet region of He, He+ 0.3% O₂ and argon discharges. Other discharge parameters are: $z=1-25$ mm, gas flow rate=10-20 slm, input power=16 W, gap=1 mm.

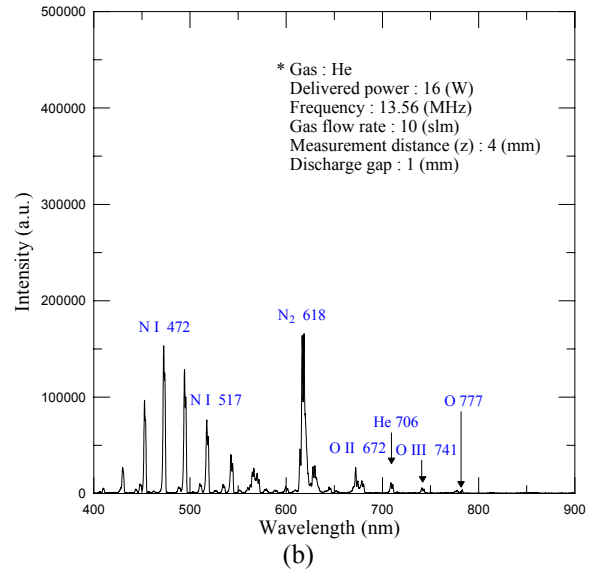
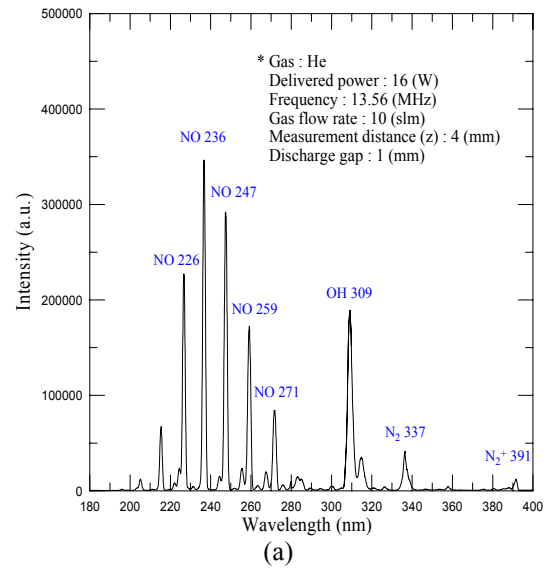


Fig. 9 Optical emission spectrum in the (a) UV and (b) VIS region for the post-glow helium plasma.

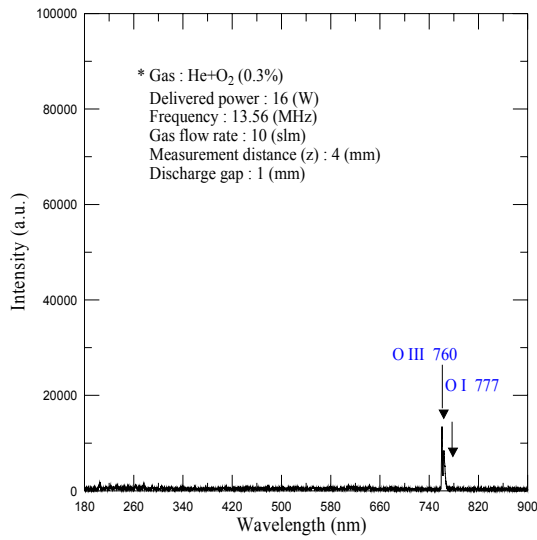


Fig. 10 Optical emission spectrum in the UV-VIS region for the post-glow He+0.3% O₂ plasma.

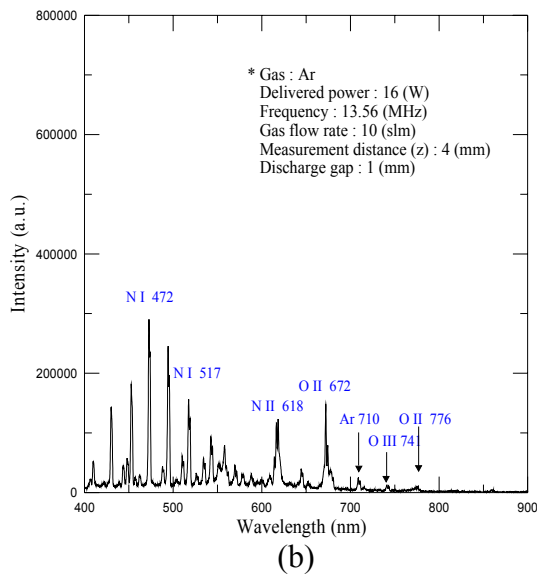
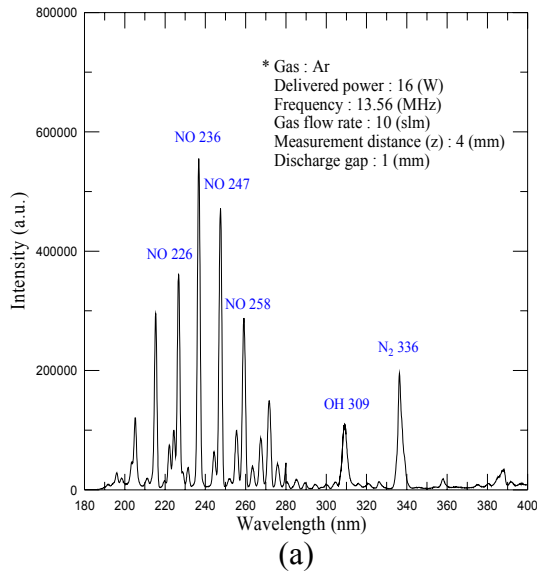


Fig. 11 Optical emission spectrum in the (a) UV and (b) VIS region for the post-glow argon plasma.

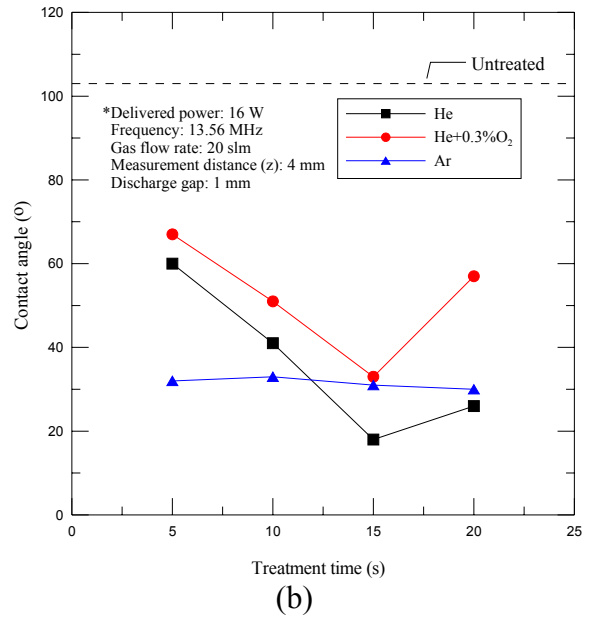
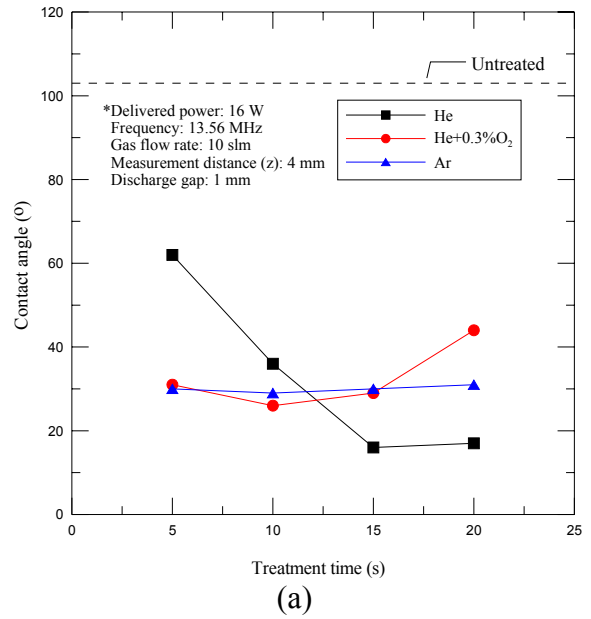


Fig. 12 Characteristic of the contact angle for wetting of a PP film after plasma treatment as a function of the treatment time in the post-glow region. Other discharge parameters are: $z=4$ mm, gas flow rate (a) 10 and (b) 20 slm, input power=16 W, gap=1 mm.