

Spectral Diagnosis of Atmospheric Pressure Plasma Jet

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A self-made atmospheric pressure plasma jet reactor on intermediate frequency is presented in this paper. The working gas argon went through the quartz tube and the plasma was generated from a nozzle at atmospheric pressure. The active species generated in the plasma were investigated by optical emission spectroscopy. Through the analysis of the emission spectrum in a series of experiments, it is confirmed the existence of some free radicals, by analysis of the relative strength of active species and the possible dissociation process. There exist large differences in the activity species of plasma between in air environment and in the quartz tube and nitrogen is the most important factor. The experimental results indicate that spectral diagnosis has been proved a workable method and optical emission spectroscopy diagnosis is significant for the research of chemical principle in plasma. Thus, it is important to the application in plasma polymerization and material modification.

Keywords: Spectral diagnosis, Optical emission spectroscopy, Atmospheric pressure plasma jet, Plasma polymerization.

INTRODUCTION

Atmospheric pressure plasma jet is an effective method to produce non-equilibrium plasma at atmospheric pressure and it has become a hot topic in the field of low temperature plasma¹⁻⁴. Atmospheric pressure plasma jet device is applied not only in the field of physics, but also in the material surface modification⁵, sterilization⁶, medical science⁷, thin film deposition⁸, spectroscopy diagnosis⁹, active substance detection^{10,11}. According to the researches in recent years, it's very difficult to monitor and analyze the internal plasma chemical reaction process in atmospheric pressure plasma jet and it's also difficult to know the change of active components from the jet into the air. The main methods of diagnosis of plasma parameters contain microwave, probe and emission spectrometry. Compared with other methods, emission spectrometry has the advantages of simple device, without direct contact and can be performed real-time. So the main work of this paper is to analyze active substances and groups which are generated during the discharge and analyze the relative intensity of spectrum lines *via* the method of emission spectrum.

EXPERIMENTAL

Experimental device: Fig. 1 shows a self-made atmospheric argon plasma jet experimental device. The quartz tube

(inner diameter 1.0 mm, wall thickness 0.5 mm) is used as the barrier dielectric and aluminium electrode was applied around the quartz tube. High voltage AC of intermediate frequency 18-20 kHz was used. Optical emission spectroscopy was recorded by using HR4000 high-resolution spectrometer (Ocean Optics) and all signals would be directly sent to the computer for data processing by operating software SpectraSuite.

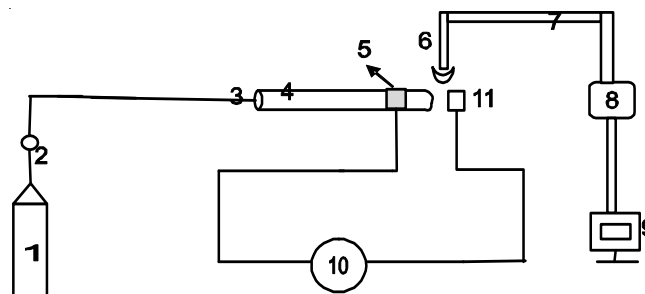


Fig. 1. Atmospheric pressure plasma jet device of spectra diagnosis

Spectral measurement: In this paper, optical fiber probe was placed vertically on the side of quartz tube to record spectrum as shown in Fig. 2. The purpose is to analyze particles (groups) before and after the atmospheric pressure plasma jet injected into the air.

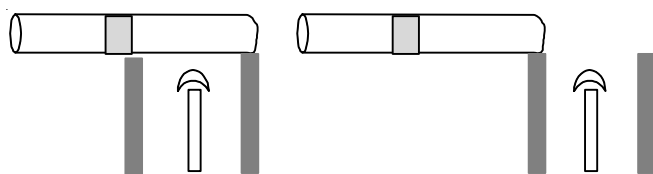


Fig. 2. Optical fiber probe position of measurement plasma in or out quartz tube

RESULTS AND DISCUSSION

When the discharge began, the plasma is dark purple; with the discharge power rose and the argon continuous accessed, plasma became bright, stable and dispersed. Figs. 3-5 are respectively the ultraviolet-visible band, visible-infrared band, visible band spectrum of argon plasma.

Reaction: The UV spectra of argon plasma was shown in Fig. 3, the main light emitting groups are OH and N₂. The OH peak value of plasma in the quartz tube is higher than outside the quartz tube. It is presumed argon carried a small amount of impurities of water molecules, which was dissociated into OH groups by the high voltage¹². When plasma went into the air, the air particles and the groups in the plasma collided with each other and cause OH spectral intensity reduced. The O atoms produced by the air and the OH groups react at the same time, reduced the concentration of OH groups¹³:

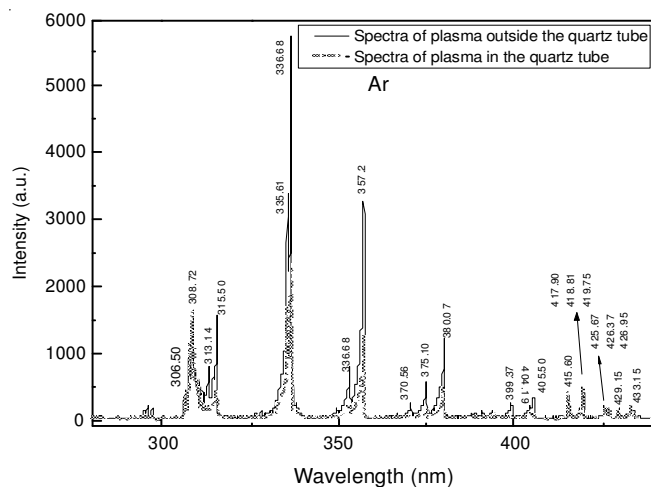


Fig. 3. Ultraviolet-visible band spectra of argon plasma

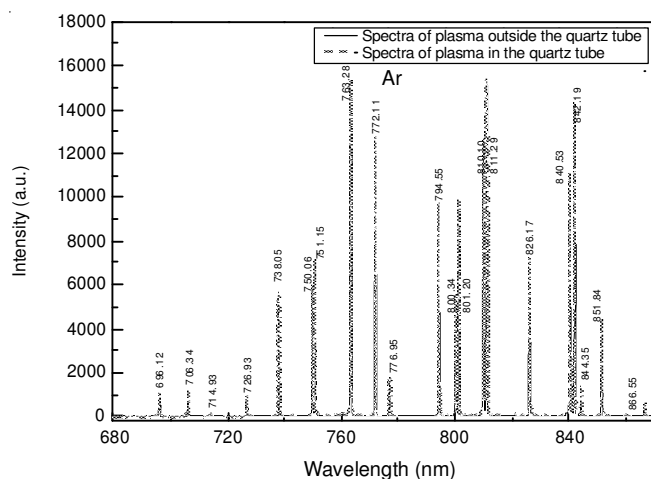


Fig. 4. Visible-infrared band spectra of argon plasma

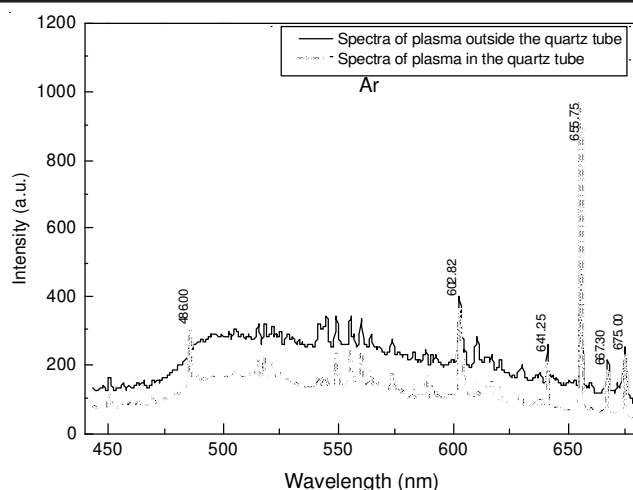
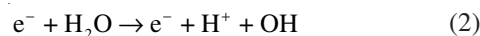
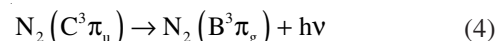
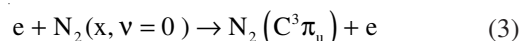


Fig. 5. Visible band spectra of argon plasma

The OH group is given by:



It is obvious to find the spectral lines of N₂ (337.17, 357.17, 380.70, 399.80 and 405.90 nm). This is due to the experiment was carried out at atmospheric pressure. It's difficult to completely remove the influence of air environment. But there is no spectral line of N. It may be because that life of dissociated N atoms is short and N atoms will compound into complex N₂ molecules¹⁴. Quartz tube is a relatively closed environment, so that there is no obvious spectral line. N₂ characteristic peaks' positions are 337.17, 357.17, 380.70, 399.80 and 405.90 nm (Table-1), which is mainly generated by the collision of electronic excitation¹⁴ ($\text{C}^3\pi_u \rightarrow \text{B}^3\pi_g$):



As shown in Fig. 4, the near infrared spectrum of argon plasma, characteristic peaks are mainly Ar and O (776.95, 844.35 nm). The relative intensity of spectra of plasma outside of the quartz tube is weaker than that in the quartz tube, which indicates that the active particles crashed with air and annihilated when contact with air. There are O characteristic peaks in near infrared spectrum and the relative ratio of plasma spectral line in the quartz tube is similar to that outside the quartz tube. It is suggested that the main source of O is not from the air, but from the plasma reaction. Oxygen atoms mainly through the following reaction:

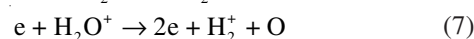
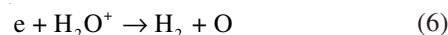
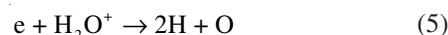


Fig. 5 shows that in the visible band spectrum, spectral intensity of plasma outside quartz tube is overall higher than the outside in quartz tube, presumably the active components is mostly from the air. In the peaks' positions 486 and 655.75 nm, the spectrum intensity in the quartz tube is higher than that outside the quartz tube. It is indicated that the peaks are due to plasma effects. Consulting literatures, the spectral line is of H spectrum line, by the formula 1, plasma reaction process may produce H, the recorded spectral lines was also confirmed

TABLE-1
OPTICAL EMISSION SPECTROSCOPY
DIAGNOSIS OF PLASMA SPECIES [Ref. 12-14]

Species	Wavelength (nm)	Corresponding spectral term
OH	305-315	$A^2\Sigma \rightarrow X^2\Pi$
N ₂	337.11	$C^3\Pi_u \rightarrow B^3\Pi_g$
N ₂	357.17	$C^3\Pi_u \rightarrow B^3\Pi_g$
N ₂	380.70	$C^3\Pi_u \rightarrow B^3\Pi_g$
N ₂	399.80	$C^3\Pi_u \rightarrow B^3\Pi_g$
N ₂	405.90	$C^3\Pi_u \rightarrow B^3\Pi_g$
H _β	486.12	Balmer (n=4-2)
H _α	656.20	Balmer (n=3-2)
Ar I	696.12	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	706.34	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	714.93	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	726.93	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	738.05	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	750.06	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	751.15	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	763.28	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	772.11	$3s^23p^5 4s \rightarrow 3s^23p^54p$
O	776.95	$3p^5P \rightarrow 3s^5S^0$
Ar I	794.55	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	800.34	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	801.2	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	810.1	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	811.29	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	826.17	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	840.53	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	842.19	$3s^23p^5 4s \rightarrow 3s^23p^54p$
O	844.35	$3p^5P \rightarrow 3s^5S^0$
Ar I	851.84	$3s^23p^5 4s \rightarrow 3s^23p^54p$
Ar I	866.55	$3s^23p^5 4s \rightarrow 3s^23p^54p$

the high concentration of H radicals produced in the plasma reaction. In addition, many peaks' positions are unable to verify the corresponding plasma groups and those will be stored for further research.

Conclusion

According to the record and analysis of the plasma emission spectra in and out of the quartz tube, active groups in the plasma does exist certain differences. Compared to

plasma emission spectrum in the quartz tube, N line is quite obvious outside the tube, while the OH spectrum is relatively weak and Ar and O spectra is weakened to some extent. This is due to plasma into the air, Ar, O and a part of OH groups collided with the air have been annihilated. Another part of OH groups reacted with O and O₃, the spectral line intensity reduced.

Through the emission spectral analysis, atmospheric argon plasma emission spectroscopy diagnosis is one of the effective methods to analyze complex plasma reaction process. Using plasma jet device can get a good outcome. For future application in material surface modification, plasma polymerization and other fields, this is a beneficial exploration and attempt.

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