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Spectral Diagnosis of Acetone Species Under Atmospheric Pressure Argon Plasma Condition

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A new approach to dielectric barrier discharge plasma jet at atmospheric pressure is presented in this paper. There are few studies have investigated the optical emission spectroscopy diagnosis of acetone species in intermediate frequency argon/acetone plasma at atmospheric pressure. Argon/acetone plasma jet was generated from a nozzle using an AC discharge in intermediate frequency at atmospheric pressure. The spectral lines of argon and acetone plasma emission were obtained and analyzed by using HR2000 spectral diagnosis equipment. It is significant for the application of optical emission spectroscopy diagnosis of volatile organic reagent in argon plasma and is important to the concept of plasma polymerization of volatile organic reagent. There exist large differences in the activity species of acetone plasma at atmospheric pressure and under vacuum condition and oxygen is the most important influencing factor. In addition, the spectral line 706.35 and 727.38 nm are chosen to estimate the electron excitation temperature. The experimental results indicate that spectral diagnosis has been proved a workable method.

Key Words: Spectral diagnosis, Optical emission spectroscopy, Atmospheric pressure plasma jet, Acetone species.

INTRODUCTION

Scientific investigation of atmospheric gas discharges dates back to the 19th century. In the former, discharges at the interface between a dielectric solid and the gases are undesirable since they usually lead to chemical degradation and physical erosion and may eventually result in failure of the solid. In the latter case, materials processing, such chemical changes are attributes. Dielectric barrier discharge derives from the fact that at least a dielectric material covers one of two planar or cylindrical electrodes connected to an ac power supply. The main advantage of this type of electrical discharge is that a 'cold' (non-equilibrium) plasma conditions in atmospheric or near-atmospheric pressure gases can be maintained in an economic and reliable way, a fact that has opened many application areas like ozone generation, excitation of gas lasers and excimer lamps, plasma displays, surface treatment of polymers and many others[1-6].

For the reason of atmospheric dielectric barrier discharge plasma jet being non-equilibrium plasma, one important parameter to describe such plasma is the electron excitation temperature, which is a magnitude greater than the ion and the gas temperatures. In order to derive the electron excitation temperature of the atmospheric dielectric barrier discharge plasma by optical emission spectroscopy, a Boltzmann plot was applied[7-9].

There are few studies have investigated plasma diagnosis of acetone species by optical emission spectroscopy. Using argon as a background gas, the spectral lines of argon/acetone plasma jet with intermediate frequency at atmospheric pressure can be recorded and analyzed.

EXPERIMENTAL

The experimental system is produced that mainly consists of two plane-parallel stainless steel electrodes and a quartz tube (internal aperture 5 mm and external aperture 8 mm) is placed between two electrodes (Fig. 1). Argon flows through the tube and out into the environmental air. A sinusoidal type of voltage with several tens kV is applied to the electrodes and the frequency of power supply is varied in the range from 1-20 kHz. Plasma spectroscopic emissions are recorded by using HR2000 High-resolution Spectrometer (Ocean Optics) and all signals will be directly sent to computer for data processing immediately during the experiment.

RESULTS AND DISCUSSION

Fig. 2 demonstrates the stream of atmospheric pressure argon plasma jet had a length of *ca.* 3.5 cm outside of the tube

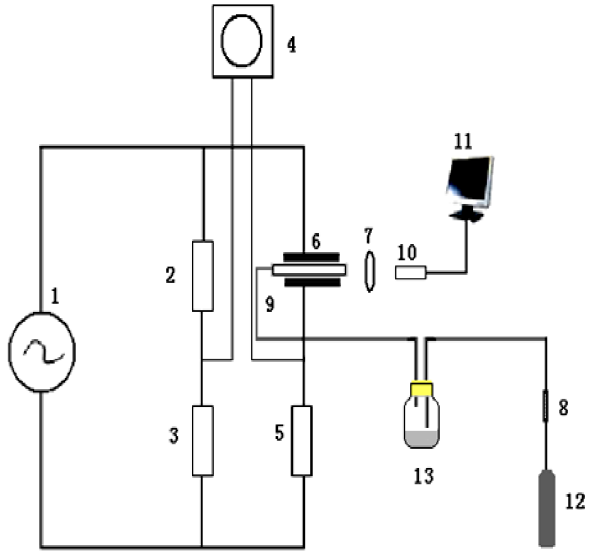

Fig. 1. Experimental arrangement of spectroscopic diagnostics of atmospheric pressure plasma jet. 1. Power; 2,3. Resistance (R_2/R_3 = 1000); 4. Digtial oscilloscope; 5. Current-sampling resistance (R = 100 Ω); 6. Electrodes; 7. Lens system; 8. Flowmeter; 9. Quartz tube; 10. Optical fiber; 11. PC; 12. Gas bottle; 13. Acetone

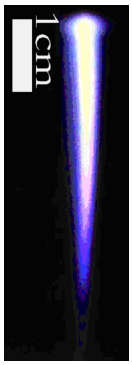

Fig. 2. Demonstration of atmospheric pressure argon plasma jet

and a homogeneous cylindrical shape. Fig. 3(a-c) show dielectric barrier discharge with different gas at atmospheric pressure in quartz tube. It is found that three photographs have different colours in visible band. Optical emission spectroscopy comprises several techniques that form the important means we have for plasma diagnosis and analysis. We measure spectra emitted by atoms and ions with optical transitions in the wide wavelength range from *ca.* 294-740 nm. This range includes the ultraviolet and visible light (from violet at 380-740 nm). With optical emission spectroscopy, we can determine the chemical composition of plasma qualitatively.

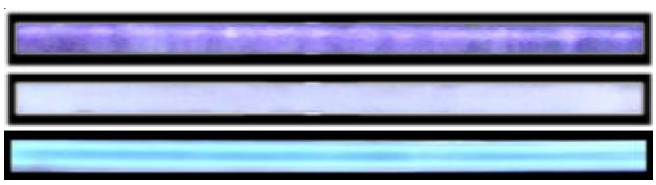

Fig. 3. Photograph of dielectric barrier discharge at atmospheric pressure in quartz tube. (a) Air; (b) argon; (c) argon/acetone

In general, the mean of optical emission spectroscopy analysis of atmospheric plasma is more difficult than that of low-pressure plasma at pure gas filled case. At low pressure, a simple spectral structure should be observed in plasma emission spectroscopy. In contrast, at the case of the atmospheric pressure, the recorded spectroscopic emission is much complicated due to many elements plasma emissions as shown in Fig. 4. Based on literature[10,11], part of argon plasma lines has been identified for approximately quantitative calculation of plasma parameters, such as electron excitation temperature, in order to control the process of materials surface modification.

Assuming we have a homogeneous and optically thin plasma, the relative transition probabilities of two different lines (indicated by suffixes 1 and 2) may be determined from the following expression[12]:

$$\frac{I_1}{I_2} = \frac{A_1 g_1 \lambda_2}{A_2 g_2 \lambda_1} \exp\left[-\frac{(E_2 - E_1)}{kT_e}\right] \quad (1)$$

Here A is the transition probability, I the total intensity, λ the wavelength, E the excitation energy, k the Boltzmann constant and g the degeneracy of the upper level. Eqn. 1 implies that the upper levels involved are in partial local thermodynamic equilibrium (LTE); for our plasma this condition is easily satisfied[13]. The natural logarithm of eqn. 1 can be written as the following expression:

$$T_e\left(\ln\frac{I_1\lambda_1}{A_1 g_1} - \ln\frac{I_2\lambda_2}{A_2 g_2}\right) = \frac{1}{k}(E_1 - E_2) \quad (2)$$

According to eqn. 2, the slope of the straight line, which fits about E/k and ln (Iλ/gA), is just electron excitation temperature. Considering some influence factors, such as self-absorption, signal-to-noise ratio and mutual interference of emission spectroscopy. And in eqn. 2, g, A, E were studied by literature[10-12]. A straight line of least squares fit about E/k and ln (Iλ/gA) can be achieved. According to eqn. 2, the slope of the straight line is just electron temperature. The measurement method of electron excitation temperature is practical and helpful to obtain other related parameters so that the process of material modification will be controlled.

Fig. 4 shows emission spectra of argon/acetone plasma jet at atmospheric pressure. The spectrum lines of neutral argon atom spectrum lines in the range 680-740 nm are recognized. And part of argon/acetone plasma lines has been identified, as shown in Table-1.

TABLE-1
OPTICAL EMISSION SPECTROSCOPY DIAGNOSIS OF ARGON/ACETONE PLASMA SPECIES

Species	Wavelength (nm)
OH	308.23, 309.15
CH	312-318, 389.08, 431.45
H_γ	434.04
CO	451.18
C^+	336.39, 392.24, 426.21, 503.62, 514.97
C^{2+}	385.45, 418.59, 466.03, 501.43, 516.49
Ar(I)	427.55-429.79, 512.79, 516.27
Ar(II)	473.54, 487.85, 516.71

Compare with optical emission spectroscopy of acetone at low pressure recorded by Okoshi *et al.*[14], Fig. 4 shows the relatively strong emission of the OH band around 309 nm at

TABLE-2
SPECTRAL LINES DATA OF ARGON[16]

No.	λ (nm)	Spectral term	g	A ($10^{-6}s^{-1}$)	E (eV)	I (au)	ln (Iλ/gA)	E/k (10^5 K)
1	706.35	$^2P_3 \rightarrow {}^1S_5$	5	3.08	13.302	231.659	9.27	1.542
2	727.38	$^2P_2 \rightarrow {}^1S_4$	3	1.83	13.328	290.595	10.56	1.545

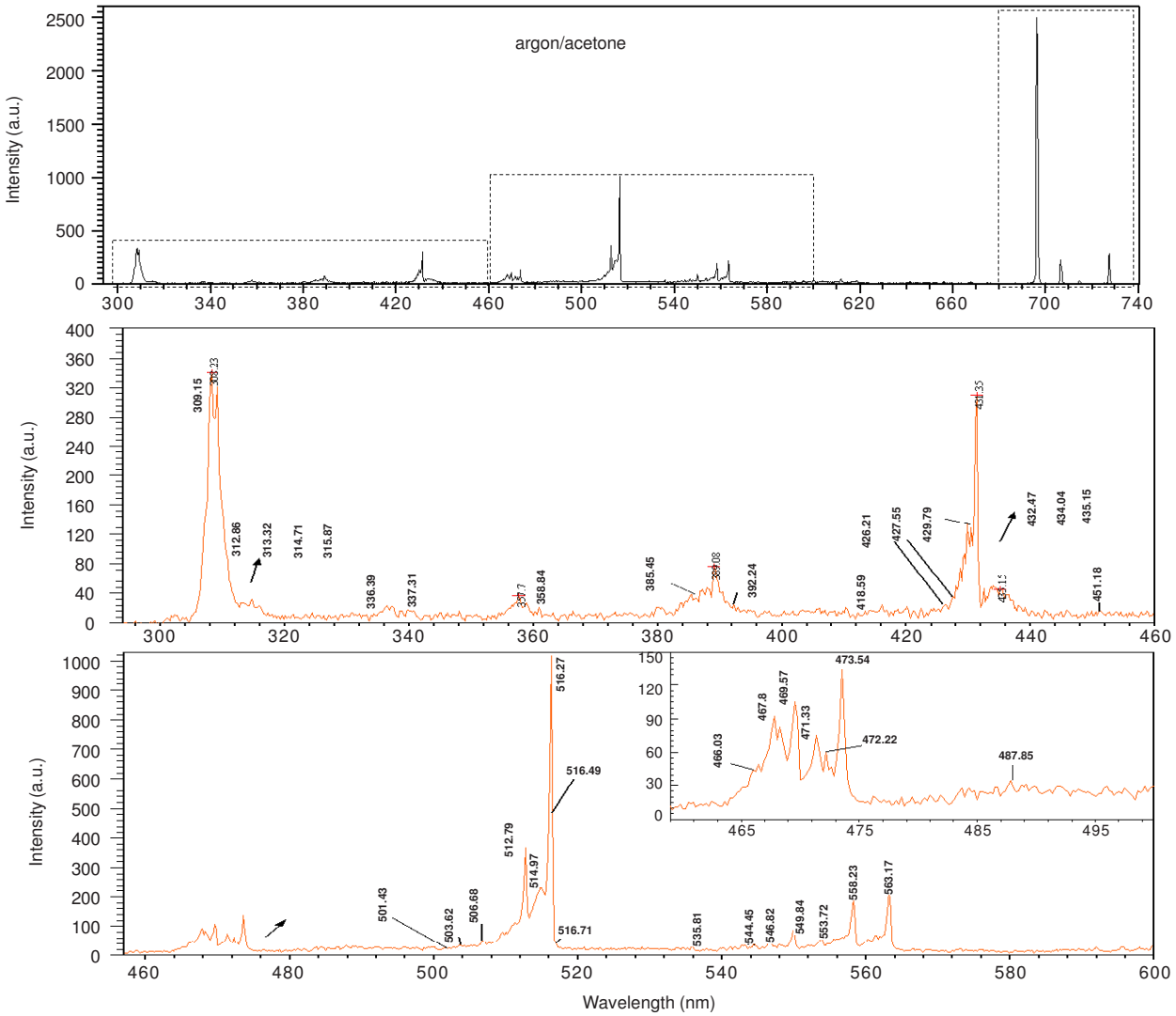

Fig. 4. Emission spectra of argon/acetone plasma jet at atmospheric pressure

atmospheric pressure, but emission lines of O^+ at 376 nm, H at 397 nm and C^+ at 406.8 nm have not been found and spectral line intensity of CO around 451.4 nm is lower than that at low pressure obviously. By analyzing acetone emission spectra at atmospheric pressure and under vacuum condition, there exist large differences in the activity species of acetone plasma and oxygen is the most important influencing factor[15].

Fig. 5 shows a straight line of least squares fit about E/k and ln (Iλ/gA). According to eqn. 2, the slope of the straight line is just the electron excitation temperature. The spectral line 706.35 and 727.38 nm are chosen to estimate the electron excitation temperature, as shown in Table-2. Although the experimental results are relatively rough, the measuring method for the electron excitation temperature is practical and helpful to obtain other related parameters.

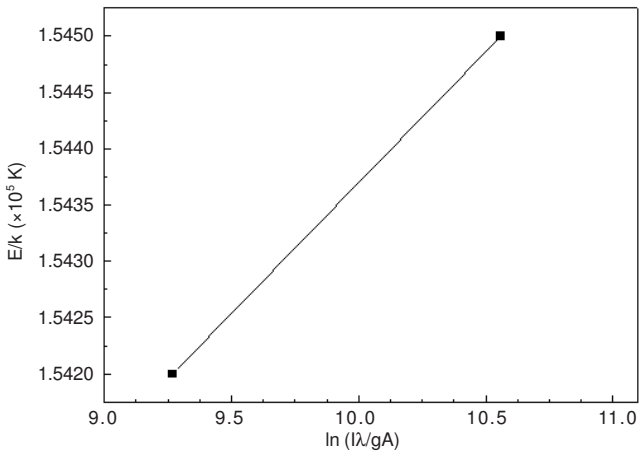

Fig. 5. Relationship between E/k and ln (Iλ/gA)

Conclusion

Atmospheric pressure plasma jet was produced to diagnose acetone species in argon by optical emission spectroscopy. The spectrum lines of neutral argon atom spectrum lines in the range 680-740 nm and argon/acetone plasma spectrum lines in the range 300-570 nm are recognized. An exciting result was observed that there exist large differences in the activity species of acetone plasma at atmospheric pressure and under vacuum condition and oxygen is the most important influencing factor. In addition, the electron excitation temperature of plasma jet in atmospheric pressure is quantitatively analyzed using relative intensity of argon spectrum lines. Therefore, optical emission spectroscopy diagnosis has been proved a workable method and it is great importance to atmospheric plasma diagnosis of chemical activity species. It is significant for the application of optical emission spectroscopy diagnosis of volatile organic reagent in argon plasma and is important to the concept of plasma polymerization of volatile organic reagent.

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REFERENCES

1. U. Kogelschatz, *IEEE Trans. Plasma Sci*., **30**, 1400 (2002).
2. U. Kogelschatz, *Pure Appl. Chem*., **62**, 1667 (1990).
3. J.H. Kim, J. Sohn, J.H. Cho, M.Y. Choi, I.G. Koo and W.M. Lee, *Plasma Process. Polym*., **5**, 377 (2008).
4. B. Eliasson and U. Kogelschatz, *IEEE Trans. Plasma Sci*., **19**, 309 (1991).
5. U. Kogelschatz, B. Eliasson and W. Egli, *J. Phys. IV (France)*, **7**, C4 (1997).
6. U. Kogelschatz, B. Eliasson and W. Egli, *Pure Appl. Chem*., **71**, 1819 (1999).
7. X.L. Tang, X.P. Feng, Z.G. Li, Y.-H. Yan and G. Qiu, *Spectrosc. Spect. Anal*., **24**, 1437 (2004).
8. G. Shanmugavelayutham, V. Selvarajan, P.V.A. Padmanabhan, K.P. Sreekumar and N.K. Joshi, *Curr. Appl. Phys*., **7**, 186 (2007).
9. S. Forster, C. Mohr and W. Viol, *Surf. Coat. Technol*., **200**, 827 (2005).
10. H. Meiners, *J. Quant. Spectrosc. Radiat. Transfer*, **9**, 1493 (1969).
11. P. Murphy, *J. Opt. Soc. Am*., **58**, 1200 (1968).
12. H.D. Campbell and A.J. Barnard, *J. Quant. Spectrosc. Radiat. Transfer*, **9**, 461 (1969).
13. H.R. Griem, Plasma Spectroscopy, McGraw Hill Book Co., New York, p. 148 (1964).
14. M. Okoshi, S. Higuchi and M. Hanabusa, *Appl. Surf. Sci*., **154**, 376 (2000).
15. A. Bradley, *J. Electrochem. Soc*., **119**, 1153 (1972).
16. L.F. Dong, J.X. Ran, Z.Q. Yin and Z.-G. Mao, *Spectrosc. Spect. Anal*., **25**, 1184 (2005).