Electron temperature and density measurement of a dielectric barrier discharge argon plasma generated with tube-to-plate electrodes in water

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Abstract: A dielectric barrier discharge argon plasma was generated with tube-to-plate electrodes in the water by a sinusoidal excitation voltage at atmospheric pressure. Time-averaged optical emission spectroscopy was used to measure the plasma parameters, of which the rotational temperature of OH was obtained by comparing the simulated spectrum with the measured spectrum at the $A^2\Sigma^+ \rightarrow X^2\Pi$ band transition and the electronic excitation temperature was determined by Boltzmann's plot method. Furthermore, the emission intensity ratio of atomic argon lines $\lambda = 811.5$ nm to $\lambda = 750.4$ nm was used to determine the electron temperature and the Stark broadening of the hydrogen Balmer H_α line was applied to measure the electron density. It has been found that the electron temperature and density in this argon plasma were in the range of 1.02-1.43 eV and on the order of 10^{14} cm⁻³, respectively, and the excitation temperature, rotational temperature, and electron density increase with the increase of applied voltage. Besides, the properties of the argon dielectric barrier discharge were also studied by electrical diagnosis.

Keywords: Dielectric barrier discharge, Conduction current, Electron temperature, Electron

density

1. Introduction

Recent year, the plasma-based oxidation methods used to remove the organic contaminants in the water have been increasingly attracted because the hydrogen peroxide, ozone, hydroxyl, oxygen radicals, and other chemically active species can be generated by the electrical discharges in or in contact with the water [1-5]. Many plasma generation methods, such as electrohydraulic discharge, corona discharge, dielectric barrier discharge (DBD), microwave discharge, radio-frequency discharge, arc discharge, spark discharge and others, have been applied [6]. Among these methods, the DBD had a unique advantages that were stable and can be easily produced under ambient conditions to realize uniform micro-discharge over the entire electrode area [7, 8]. Currently, the electrical discharge of DBD in the water used to remove organic contaminants can be divided into two main groups. One was that the DBD plasmas was generated in the inside of reactors immersed in the water and transferred a section of distance with flowing working gas, and then injected into the water by bubbling or aeration [6, 9-11]. under this condition, the most active species does not react with organic pollutants in the water because the transferred time of plasma in the inside of reactor much larger than lifetime of active species that was about dozens of nanoseconds. However, the short lifetime active species such as OH and O radicals play an important roles for organic pollutants removal [12, 13], so the degradation efficiency of organic pollutants will obviously decrease. Another was that the DBD plasmas was directly generated in the water. Under this condition, the active species can sufficiently react with organic pollutants in the water, hence the degradation of organic pollutants was relatively high. However, there was very little the

reports with related to that the DBD plasmas were directly generated in the water. P. Manoj Kumar Reddy et al. and B. G. Rodríguez-Méndez et al. respectively reported the DBD plasmas generated with a wire-to-cylinder electrodes under pre-existing gas bubbles in the water [14, 15], but the DBD plasma generated by a tube-to-plate electrodes in the water has not been reported up to now. Furthermore, the wire-to-cylinder electrodes were more difficult to develop the intense electric field necessary to initiate electrical discharges in the water than the needle-to-plate electrodes [16].

Currently, It was generally accepted that the electrical discharges in the water were initiated in the gas phase-bubbles, which were either formed due to local heating of water or injecting gas into the water [17]. The bubbles externally generated by injecting gas into the water between the powered and grounded electrodes may be preferable than in that without pre-existing bubbles for several reasons. The electrical breakdown in the water requires electric field strength of about 2 MV/cm (depending on the conductivity of the water) [18] and converts a large fraction of the input energy into heat to produce gas bubbles that were subsequently ionized by large electric field. On the contrary, the pr-existing bubbles between the powered and grounded electrodes improve the energy balance with more energy used to generate chemically active species and the breakdown field strength also reduces to be about 30 kV/cm [18]. Furthermore, the chemical and physical processes occur in the gas-liquid interfaces at which plasma were generated through ionizing gas bubbles and then interacts with the organic contaminants dissolved in the water [12, 19].

For these reasons, a DBD Ar plasma was generated with a tube-to-plate electrodes and powered by a 8 kHz sinusoidal excitation voltage in the water. The Ar gas was directly injected

into the water through a tube power electrode and the discharge was ignited with a low applied voltage in the water, because the seed charge was generated in the quartz glass tube in advance and transferred by electrical force and gas flow into the bubbles. The aim of this work was to analyze the discharge characteristics such as conduction and displacement current, discharge power, rotational temperature of OH, electron excitation temperature, electron temperature, and electron density.

2. Experimental setup

A schematic diagram of experimental apparatus and discharge photograph were shown in Fig. 1. The power electrode was a stainless steel tube powered by a sinusoidal excitation voltage at 8 kHz, which had an inner diameter of 1.8 mm, an outer diameter of 2 mm, and a length of 250 mm. The power electrode was tightly attached on the inside wall of the quartz glass tube that had an inner diameter of 2 mm, an outer diameter of 3 mm and a length of 200 mm, and its open end was in the position of 3 mm away from the exit of the quartz glass tube to prevent contacting water and generating seed charge at gas phase in advance. The quartz glass tube was vertically immersed in the water at a depth of 30 mm. The water was held in a transparent quartz glass vessel that consists of the cylindrical quartz glass tube and the quartz glass plate. The cylindrical quartz glass tube had an inner diameter of 30 mm, an outer diameter of 32 mm and a length of 70 mm, and the quartz glass plate had a 50 mm length, 50 mm width, and 1 mm thickness. The distance between the open end of power electrode and the bottom of quartz glass vessel was 10 mm. A copper foil was tightly attached on the outside surface of quartz plate as a ground electrode, which had a 40 mm length, 40 mm width, and 0.2 mm thickness.

The working gas of Ar (99.999%) was injected into the water through the power electrode controlled by mass flow controller at Ar flow rate of 2 L/min. Therefore, the gas bubbles were formed in between the power and ground electrodes. The applied voltages were measured by a high voltage probe (Tektronix P6015A), the currents were measured through a 20 Ω resistor in series with the ground electrode, and the electrical signals were recorded via a digital oscilloscope (Tektronix MDO3012). The Lissajous plot was obtained through a 1 μ F capacitor in series with the ground electrode. In order to collect the optical emission of the DBD plasma, an optical fiber was located in the position that is 2 and 3 mm away from the cylindrical quartz glass tube and the quartz glass plate respectively, and the spectral intensities were recorded by a spectrometer (Acton Research Spectrapro-2500i) with 300 grooves/mm gratings (spectral lines, 300-900 nm) and 2400 grooves/mm gratings (OH line, 306-312 nm and H $_{\alpha}$ line, 656.3 nm) and slit width of 20 μ m. The discharge images were obtained by a Canon digital camera EOS 5D Mark III.

3. Experiment results and discussions

3.1. Electrical discharge characteristics

Fig. 2 shows the equivalent circuit diagram of the DBD Ar plasma device. As shown in Fig. 2, C_p and R_p represent the equivalent capacitance and the resistance of the DBD Ar plasma respectively, and C_d represents the dielectric capacitor between the power and ground electrodes. Accordingly, the DBD Ar plasma device was essentially a parallel circuit that consists of the capacitor C_d and the series circuit of capacitor C_p and resistor R_p . Also, $U_{tot}(t)$, $I_{tot}(t)$, $I_{dc}(t)$, and $I_{dp}(t)$ represent the externally excited voltage, total current, conduction current, and displacement current, respectively.

According to the equivalent circuit diagram given in Fig. 2, following equations were obtained:

$$\frac{dU_{tot}(t)}{dt} = \frac{I_{dp}(t)}{C_d}, \quad (1)$$

$$I_{tot}(t) = I_{dp}(t) + I_{dc}(t). \quad (2)$$

If the dielectric capacitor C_d was determined, the displacement current $I_{dp}(t)$ can be calculated using the applied voltage $U_{tot}(t)$ from the Eq. (1). The dielectric capacitor C_d was determined by the applied voltage $U_{tot}(t)$ and the displacement current $I_{dp}(t)$ measured at a peak applied voltage of 2.5 kV. When the air replaces the argon as the working gas the discharge does not occurs between the power and ground electrodes at a peak applied voltage of 2.5 kV, so the measured current by a digital oscilloscope was displacement current $I_{dp}(t)$, as shown in Fig. 3(a). Therefore, the dielectric capacitor C_d was easily determined by Eq. (1) with the data in Fig. 3(a). Fig. 3(b) shows the capacitance waveform of C_d for three periods. In Fig. 3(b), the two peaks appeared symmetrically in the every half-period, which arise due to numerical singularities occurring during the zero crossing of the denominator [20, 21]. Accordingly, the average value of C_d was determined without considering the effect of peaks equal to 3.29 pF. The discharge current was determined through subtracting the displacement current from the total current using Eq. (2). This measuring method has been detailedly reported in the previous studies [22, 23]. Fig. 4 shows the waveforms of applied voltage, total current, conduction current, and displacement current at a peak applied voltage of 9 kV. As presented in Fig. 4, the waveforms of total current and applied voltage have a phase difference of nearly 90° , which indicates that the impedance of the DBD device was capacitive. The amplitude of conduction current was much close to the total current due to that the displacement current takes up a small

proportion of the total current, which was indicated that the average power can be regarded as the effective power. In the every half-period of the applied voltage, a lot of filaments appear on the waveform of conduction current, which was similar to the typical characteristics of atmospheric pressure DBD in the gas phase [24, 25]. The peaks of conduction current $I_{dc1}(t)$ were 149.34 and -145.32 mA and the corresponding peaks of total current $I_{dcp1}(t)$ were 154.23 and -150.38 mA, respectively.

Fig. 5 shows the variations in discharge power versus peak applied voltage. The discharge power was determined by a Lissajous plot obtained through a 1 μ F capacitor in series with the ground electrode [26]. Regarding the results shown in Fig. 5, the average power increases linearly with the increasing peak applied voltage. As the peak applied voltage increases from 6 to 10 kV, the average power increases from 3.41 to 10.47 W.

3.2. Gas temperature and electronic excitation temperature

The rotational temperature T_{rot} (K) of OH was normally accepted as an accurate estimate of the gas temperature of the atmospheric-pressure plasmas [27, 28]. Therefore, the emission spectrum of the $A^2\Sigma^+ \rightarrow X^2\Pi$ band transition of OH in the range from 306 to 312 nm was used to determine the gas temperature that was obtained when the best fits were achieved between the measured and simulated spectrum of the $A^2\Sigma^+ \rightarrow X^2\Pi$ band transition of OH. Fig. 6 shows the variations in rotational temperature versus peak applied voltage and inset graph was the measured and simulated spectrum of the $A^2\Sigma^+ \rightarrow X^2\Pi$ band transition of OH at a peak applied voltage of 9 kV. Regarding the results shown in Fig. 6, the rotational temperature increases linearly with the increase of peak applied voltage. As the peak applied voltage increases from 6 to 10 kV, the rotational temperature increases from 608 to 738 K.

Fig. 7 shows the optical emission spectrum in the range from 300 to 900 nm. Besides the Ar emission lines, the excited H (H_α at 656.3 nm and H_β at 486.1 nm), O ($3p^5P \rightarrow 3s^5S^0$ at 777.4 nm and $3p^3P \rightarrow 3s^3S^0$ at 844.6 nm), OH ($A^2\Sigma^+ \rightarrow X^2\Pi$), N_2 ($C^3\Pi_u \rightarrow B^3\Pi_g$), and N_2^+ ($B^2\Sigma_u \rightarrow X^2\Sigma_g$ at 391 nm) were detected in the emission spectrum [29, 30], which indicates that the DBD Ar plasma device can be used for water purification. The neutral Ar 696.5, 706.7, 727.3, 738.4, 751.5, 763.5, 794.8, 800.6, 826.5, 840.8, and 852.1 nm for the transition ($3s^23p^5$) $4p \rightarrow (3s^23p^5)4s$ were chosen to determine the electronic excitation temperature T_{exc} (K) under a Boltzmann approximation [27, 28, 31]. Fig. 8 shows the variation in electronic excitation temperature versus peak applied voltage and the inset graph was the Boltzmann's plot of excitation temperature at a peak applied voltage of 9 kV. As presented in Fig. 8, the electronic excitation temperature increases linearly with the increasing peak applied voltage, which was in well agreement with the conclusion reported by Qian et al. [32]. As the peak applied voltage increases from 6 to 10 kV, the electronic excitation temperature increases from 3382 to 3565 K, which also indicates the presence of energetic electrons that were capable of generating a large of active species by inelastic collisions with molecular species.

3.3. Electron temperature

The electron temperature T_e (eV) was determined from the emission intensity ratio of two argon lines that involve the transition $2p_1 \rightarrow 1s_2$ (750.4 nm, threshold at 13.48 eV) and the transition $2p_9 \rightarrow 1s_5$ (811.5 nm threshold at 13.07 eV). It was demonstrated that the emission intensity ratio of these two lines can be used accurately to estimate the spatial evolution of T_e [33]. Under our DBD Ar plasma condition, the electronic excited states of Ar ($2p_1$) and ($2p_9$) were produced from the ground electronic state by a direct electron impact and the nitrogen,

oxygen, and water molecular densities in the bubbles comparing to the argon density were very small, so the collisional induced quenching of Ar excited states (2p₁) and (2p₉) in those molecules were neglected. Considering the collisional induced quenching of Ar excited states (2p₁) and (2p₉) in the argon, the intensity of Ar line $\lambda = 750.4$ nm can be expressed as

$$I_{750.4} = C_{750.4} h\gamma_{750.4} A_{ij}^{750.4} n_e k_e^{750.4} n_{Ar} (A_i^{750.4} + k_q^{750.4} n_{Ar})^{-1} \quad (3)$$

The intensity of Ar line $\lambda = 811.5$ nm was given by

$$I_{811.5} = C_{811.5} h\gamma_{811.5} A_{ij}^{811.5} n_e k_e^{811.5} n_{Ar} (A_i^{811.5} + k_q^{811.5} n_{Ar})^{-1}, \quad (4)$$

where the quantity C contains all optical and geometrical parameters; $h\gamma$ was the photon energy of respective transition; A_i was the sum over all optical transition probabilities A_{ij} for the excited state and is equal to the reciprocal of the natural lifetime τ_0 ; and A_{ij} was equal to $A_i = 1/\tau_0$, since the only optically allowed transition from each of the excited states was observed, respectively [34]. $k_q^{750.4}$ and $k_q^{811.5}$ denote the quenching rate coefficients of Ar excited states (2p₁) and (2p₉). The data of radiative lifetimes and quenching rate coefficients for Ar (2p₁) and (2p₉) atoms take from the literature [35]. n_e and n_{Ar} denote the electron and argon density. The argon density n_{Ar} was determined by the ideal gas equation $n = p/kT_g$, where the gas pressure p (Pa) takes the value of 1.01×10^5 Pa and the gas temperature T_g (K) was obtained from the Fig. 6.

The electron excitation rate coefficient k_e was determined by integrating the cross sections over an assumed Maxwellian distribution

$$k_e = \int_0^\infty \sigma(\varepsilon) f(\varepsilon) \sqrt{\frac{2e}{m_e}} d\varepsilon, \quad (5)$$

where m_e was the electronic mass, $\sigma(\varepsilon)$ was the electron collision cross section for excitation [36], and $f(\varepsilon)$ was the electron energy distribution function. The ratio of $k_e^{811.5}$ to $k_e^{750.4}$ as a

function of electron temperature was shown in Fig. 9. Taking the intensity ratio of Eq. (4) to Eq. (3), we obtain

$$\frac{k_e^{811.5}}{k_e^{750.4}} = \frac{C_{750.4} h \gamma_{750.4} I_{811.5} a_{ij}^{750.4}}{C_{811.5} h \gamma_{811.5} I_{750.4} a_{ij}^{811.5}} \quad (6)$$

with

$$a_{ij}^{750.4} = \frac{A_{ij}^{750.4}}{A_i^{750.4} + k_q^{750.4} n_{Ar}} \quad \text{and} \quad a_{ij}^{811.5} = \frac{A_{ij}^{811.5}}{A_i^{811.5} + k_q^{811.5} n_{Ar}} \quad (7)$$

Therefor, the electron temperature can be determined by a fitting function shown in Fig. 9 and a ratio of $k_e^{811.5}$ to $k_e^{750.4}$ calculated using Eqs. (6) and (7). Fig. 10 shows the variation in electron temperature versus peak applied voltage. As regards the results in Fig. 10, the electron temperature increases with the increase of applied voltage. When the peak applied voltage increases from 6 to 10 kV, the electron temperature increases from 1.02 to 1.43 eV. Moreover, the electron temperature was much higher than the electronic excitation temperature shown in Fig. 8, which were in accordance with the typical characteristics of DBD Ar plasmas generated at atmospheric pressure. Generally, the electron temperature is in the range of 2000-5000 K while the electron temperature was in the range of 1-3 eV for DBD Ar plasmas generated at atmospheric pressure [32, 37- 39]. It may be due to that the argon inelastic collisions cause a decrease of the argon excitation temperature so that the relation $T_e > T_{exc}$ was satisfied [27].

3.4. Electron density

It was well known that the Stark broadening of the hydrogen atomic line H_α (656.3 nm) can be used to determine the electron density when the discharges are operated at atmospheric pressure [19, 40, 41]. The total broadening of the H_α line results from the contribution of different mechanisms and each mechanism was independent and plays a different role on

broadening the line depending on the plasma conditions. In our experimental condition (gas pressure of 1 atm and T_g in the range of 608-738 K), the relevant sources of broadening necessary to consider include the Stark, Doppler, Van der Waals, and instrumental broadenings, among which the Doppler and instrumental broadenings cause the appearance of Gaussian profiles and the Stark and Van der Waals broadenings can be approximated by Lorentzian profiles. The observed line profiles were the convolution of Gaussian and Lorentzian profiles and can be expressed a Voigt profiles. The full-width at half-maximum (FWHM) was the parameter widely used to characterize the broadened line profiles. The FWHM of the Gaussian and Lorentzian profiles ($\Delta\lambda_G$ and $\Delta\lambda_L$) were obtained respectively by $\Delta\lambda_G = \sqrt{\Delta\lambda_D^2 + \Delta\lambda_{inst}^2}$ and $\Delta\lambda_L = \Delta\lambda_S + \Delta\lambda_V$, where $\Delta\lambda_D$, $\Delta\lambda_{inst}$, $\Delta\lambda_S$, and $\Delta\lambda_V$ represents the FWHM of the Doppler, instrumental, Stark, and Van der Waals broadenings, respectively [42]. The instrumental broadening was calibrated by using a He–Ne laser at 632.8 nm and $\Delta\lambda_{inst}$ was determined by a Gaussian fitting, which was found to be 0.06 nm.

The FWHM of Doppler broadening was determined from the gas temperature T_g [43]

$$\Delta\lambda_D \text{ (nm)} = 7.16 \times 10^{-7} \lambda_0 \left(\frac{T_g}{M} \right)^{0.5}, \quad (8)$$

where the gas temperature T_g was given in Kelvin, M was the H atom expressed in atomic mass units and λ_0 was the wavelength of H_α line in nm.

The FWHM of Van der Waals broadening can be expressed by formula [40]

$$\Delta\lambda_V \text{ (nm)} = \frac{5.736}{T_g^{0.7}}. \quad (9)$$

The FWHM of Stark broadening was related to the electron density by [41]

$$\Delta\lambda_S \text{ (nm)} = 0.549 \times \left(\frac{n_e}{10^{17}} \right)^{0.67965}, \quad (10)$$

where electron density was in the unit of cm^{-3} .

Under the condition of this work, the FWHM of Stark broadening was determined by separating (deconvolution) the other broadening components from the Voigt profiles and the electron density was calculated using Eq. (10), as shown in Fig. 11. The electron density increases almost linearly with the increase of applied voltage. When the peak applied voltage increases from 6 to 10 kV, the electron density increases from 2.32×10^{14} to $5.56 \times 10^{14} \text{ cm}^{-3}$. In addition, the electron density of the DBD Ar plasma can be estimated from the expressions $n_e = J / (e \mu_e E)$ [26], where J was the current density, e was the electron charge, μ_e was the electron mobility, and E was the electric field sustained in the discharge region, respectively. The current density was determined by the conduction current divided by the discharge cross section. The conduction current was obtained from the Fig. 4(b) and the discharge cross section was determined by expressions $S = \pi(R_1^2 - R_2^2)$, where R_1 and R_2 were the outer and inner radius of the power electrode. the discharge cross section was equal to 0.005966 cm^2 . The electric field E takes an approximate value of argon breakdown threshold (2.7 kV/cm) [44] at 1 atm. The value of $\mu_e p$ for argon gas was $0.33 \times 10^6 \text{ cm}^2 \text{ torr/Vs}$ [44]. Under the condition shown in Fig. 4, the electron density was estimated to be $1.34 \times 10^{14} \text{ cm}^{-3}$. It was found that the electron density calculated by the two methods was in the same orders of magnitude, which also indicates the electron density calculated by Stark broadening of H_α line was reasonable.

4. Conclusions

A DBD argon plasma generated with tube-plate electrodes in the water has been studied. It was found that the average power, rotational temperature of OH, electronic excitation temperature, electron temperature, and electron density increase linearly with the increase of

applied voltage, and which were in the range of 3.41-10.47 W, 608-738 K, 3382-3565 K, 1.02-1.43 eV, and 2.32×10^{14} - 5.56×10^{14} cm⁻³, respectively, when peak applied voltage increases from 6 to 10 kV. Furthermore, by the optical emission spectrum it was found that the excited Ar, O, N₂, N₂⁺, and OH existed in the DBD Ar plasma.

Acknowledgments

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Caption of figures

Fig. 1. Schematic diagram of experimental setup and discharge photograph at a peak applied voltage of 9 kV and Ar flow rate of 2 L/min.

Fig. 2. Equivalent circuit diagram of the DBD plasma device.

Fig. 3. Applied voltage and displacement current waveforms (a) and capacitor C_d waveform for three periods (b) at a peak applied voltage of 2.5 kV.

Fig. 4. Applied voltage and total current waveforms (a) and displacement current and conduction current waveforms (b) at a peak applied voltage of 9 kV.

Fig. 5. Discharge power versus peak applied voltage.

Fig. 6. Rotational temperature of OH versus peak applied voltage and the inset graph was measured and simulated spectrum of the $A^2\Sigma^+ \rightarrow X^2\Pi$ band transition of OH in the range from 306 to 312 nm.

Fig. 7. Optical emission spectrum of 300-650 nm (a) and 650-900 nm (b) at a peak applied voltage 9 kV.

Fig. 8. Excitation temperature as a function of peak applied voltage and the inset graph was Boltzmann's plot of excitation temperature at a peak applied voltage of 9 kV.

Fig. 9. Ratio of electron excitation rate coefficient $k_e^{811.5}$ to $k_e^{750.4}$ as a function of electron temperature.

Fig. 10. Electron temperature versus peak applied voltage.

373

374 **Fig. 11.** Voigt fitting of the H_α line profile (a) and electron density versus peak applied voltage

375 (b).

Fig.1

Fig. 2

Fig.3(a)

Fig.3(b)

Fig.4(a)

Fig.4(b)

Fig. 5

Fig. 6

Fig. 7(a)

Fig. 7(b)

Fig. 8

Fig. 9

Fig. 10

Fig. 11(a)

Fig. 11(b)