Electrical and spectral characteristics of a tube-to-plate helium plasma generated using dielectric barrier discharge in water

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Abstract: A tube-to-plate dielectric barrier discharge He plasma was directly generated in water by 8 kHz sinusoidal excitation voltage. It was found that the average power, rotational temperature of OH, electronic excitation temperature, electron temperature, and electron density increased linearly with the increase of applied voltage and with the value in the range of 3.65-11.13 W, 427-529 K, 4355-4533 K, 1.15-1.96 eV, and 2.56×10^{14} - 5.76×10^{14} cm⁻³, respectively. Furthermore, the optical emission spectrum indicated that active He*, O*, N₂*, N₂⁺*, and OH* species existed in He plasma.

Keywords: Dielectric barrier discharge, Optical emission spectrum, Electron temperature, Electron density

1. Introduction

In the past decades, dielectric barrier discharges (DBDs) have attracted growing attention in organic pollutants removal from water [1-5]. It is demonstrated that electrical discharges of DBD applied in water could directly or indirectly cause organic compound degradation by

physical and chemical effects. Physical effects include the contribution of ultraviolet light and shock waves produced by plasma discharges which depend on their intensity [5-7]. Chemical effects include direct formation of reactive species such as OH^* , O^* , H^* , H_2O_2 and O_3 , which are able to react readily with a large group of organic chemicals to form more oxidized intermediate products [5-7]. Furthermore, the DBDs are stable, which can not only prevent spark formation, eliminate electrode etching and corrosion, but also lead to an uniform discharge distribution over the entire electrode area compared with other discharges including electrohydraulic discharge, corona discharge, arc discharge, and spark discharge [8].

Currently, the electrical discharge of DBD in water used to remove organic contaminants could be divided into two main groups, namely electrical discharge inside the reactor immersed in water and direct electrical discharge in water. The plasmas need to transfer a section of distance before reacting with organic pollutants for the electrical discharge inside the reactors which are immersed in water [9-12], leading to disappear of short lifetime active species before chemical reactions. It could seriously affect the degradation efficiency of organic pollutants in water, because the short lifetime active species such as OH^* and O^* play an important role for organic pollutants removal. On the other hand, the direct electrical discharge in water could avoid this problem. Those active species produced by directly electrical discharge in water could sufficiently react with organic pollutants before disappearing, which is very beneficial for organic pollutants degradation. However, the report related to direct electrical discharge of DBD in water is rare to see.

H. Wu et al. generated DBD plasma by a needle-to-plate electrodes in water to investigate the feasibility of aniline degradation in water [13]. Since the discharge was ignited without

pre-existing gas bubbles, a large fraction of energy was converted to heat for producing gas bubbles which were subsequently ionized by a large electric field. It is generally accepted that the electrical discharge in water is initiated in the gas phase-bubbles[14]. If the bubbles are preformed by injecting a gas into water, the energy balance could be improved with more energy used to generate chemically active species and the breakdown field strength could be obviously reduced [15]. B. G. Rodríguez-Méndez et al. generated pulse DBD plasma by a wire-to-cylinder electrodes under pre-existing gas bubbles in water [16], but the applied voltage was as high as about 25 kV. Moreover, the discharge ignited by alternating current power supply requires higher applied voltage for this kind of device, because it does not inject large energy into bubbles in a short duration time (nano to microseconds) to accelerate electrons which ionize gas molecules to produce plasma.

For these reason, a DBD He plasma was generated using a tube-to-plate electrodes in water and powered by a sinusoidal excitation voltage at 8 kHz. The discharge was ignited with a low applied voltage in water, because the seed charge was generated in the quartz glass tube in advance and transferred by electrical force into bubbles which were preformed through injecting He gas into water before discharge. Thus, the discharge path was easily formed in water. The aim of this work is to analyze the discharge characteristics of DBD He plasma in water. The conduction current, displacement current and average power were determined by electrical diagnosis, and the rotational temperature of OH, electron excitation temperature, electron temperature and electron density were obtained by optical emission spectroscopy.

2. Experimental setup

Fig. 1 shows the schematic diagram of experimental apparatus and discharge photograph.

The power electrode is a stainless steel tube, 2 mm in outer diameter, 1.8 mm in inner diameter, and 250 mm in length. In order to generate seed charge in gas phase in advance and prevent contacting with water, the power electrode is tightly inserted into quartz glass tube and the open end is placed in the distance of 3 mm away from the exit of quartz glass tube. The quartz glass tube owns an inner diameter of 2 mm, an outer diameter of 3 mm, and a length of 200 mm and it is vertically immersed in water at a depth of 30 mm. The tap water is held in a transparent quartz glass vessel which consists of a quartz glass plate and a cylindrical quartz glass tube. The cylindrical quartz glass tube is 32 mm in outer diameter, 30 mm in inner diameter, and 70 mm in length. The quartz glass plate is 50 mm length, 50 mm width, and 1 mm thickness. Distance between the open end of power electrode and the bottom of quartz glass vessel is 10 mm. A copper foil is tightly attached on the outside surface of quartz plate as a ground electrode which is 40 mm length, 40 mm width, and 0.2 mm thickness.

The gas bubbles were formed between the power and ground electrodes by injecting working gas of He (99.999%) into water which was controlled by mass flow controller at flow rate of 2 L/min. The applied voltages and currents were measured by a high voltage probe (Tektronix P6015A) and a 20 Ω resistor in series with the ground electrode, and the electrical signals were recorded via a digital oscilloscope (Tektronix MDO3012). An optical fiber was used to collect the optical emission of DBD plasma and located in the position which was 2 and 3 mm away from the cylindrical quartz glass tube and the quartz glass plate respectively. 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). The resolution and slit width of spectrometer were 0.02nm

and 20 μm , respectively. 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 a typical waveforms of applied voltage $U_{\text{tot}}(t)$, total current $I_{\text{tot}}(t)$, conduction current $I_{\text{dc}}(t)$, and displacement current $I_{\text{dp}}(t)$ at the peak applied voltage of 8.2 kV. The measurement methods of conduction and displacement currents have been reported in detail in the previous studies [17, 18]. In Fig. 2, the phase difference between total current and applied voltage waveforms is almost 90° , which indicates that the impedance of DBD device is capacitive. The amplitude of displace current is much smaller than that of conduction current, in other words the conduction current takes up a large proportion in total current. It is indicated that the bulk of input energy is used to generate He plasma, thus the average power could be regarded as the effective power. A lot of filaments appear on the waveforms of total current and conduction current in the every half-period of applied voltage, which is in well agreement with the typical current waveforms of atmospheric DBD in gas phase [19, 20]. The peaks of conduction current $I_{\text{dc1}}(t)$ are 154.76 and -32.94 mA and the corresponding peaks of total current $I_{\text{dcp1}}(t)$ are 150.91 and -29.08 mA, respectively.

Fig. 3 shows the variations in average power versus peak applied voltage. The average power is determined by the well-defined expressions, $P_{\text{ave}} = \frac{1}{T} \int_0^T U_{\text{tot}}(t) I_{\text{tot}}(t) dt$, where T is one discharge cycle to be about 125 μs . As shown in Fig. 3, when the peak applied voltage increases from 6 to 10 kV, the average power linearly increases from 3.65 to 11.13 W.

3.2. Gas temperature and electronic excitation temperature

Gas temperature T_g (K) is an important parameter in any plasma processing application, and for atmospheric plasmas it is close to the rotational temperature T_{rot} (K) of OH [21-23]. In order to determine the rotational temperature of OH, the simulated spectrum is compared with the experimentally recorded spectrum in the $A^2\Sigma^+ \rightarrow X^2\Pi$ band transition from 306 to 312 nm using LIFBASE software which is a well known spectral analysis software to determine the rotational and vibrational temperature of molecules and electronic bands, such as OH (A-X), NO (A-X, B-X, C-X, D-X), CH (A-X, B-X, C-X), CF (A-X, B-X), etc [24, 25]. The gas temperature could be determined when the simulated spectrum fits well with the measure one. Considering the small fluctuation of OH spectrum intensity during discharge, OH spectrum in the range of 306-312 nm was measured for 10 times at the same peak applied voltage and then the error bars of OH rotational temperature were determined by a well known formula of standard error of the mean [26, 27]. The error bars of electronic excitation temperature, electron temperature, and electron density were also determined with above-mentioned method. Fig. 4 shows the variation in rotational temperature versus peak applied voltage and inset graph is 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. As shown in Fig. 4, the rotational temperature increases linearly with the increase of peak applied voltage. When the peak applied voltage increases from 6 to 10 kV, the rotational temperature increases from 427 to 529 K. Fig. 5 shows the optical emission spectrum in the range from 250 to 850 nm. It clearly indicates that the DBD He plasma is dominated by excited He, OH ($A^2\Sigma^+ \rightarrow X^2\Pi$), N_2 ($C^3\Pi_u \rightarrow B^3\Pi_g$), N_2^+ ($B^2\Sigma_u \rightarrow X^2\Sigma_g$), H_α (656.3 nm), H_β (486.1 nm), and O (777.4 nm). Moreover, the intensity of OH line (309 nm) is the largest in optical emission spectrum from 250 to 850 nm, which shows

that a lot of OH radicals are generated in water. The OH radicals are vital oxidants that dominate the plasma oxidation in water treatment, and their reactions with organic compounds may be differentiated into three different mechanisms: abstraction of hydrogen atom, electrophilic addition to unsaturated bond and electron transfer [8]. Thus, the DBD He plasma generated by tube-to-plate electrodes could be used for water purification.

Fig. 6 shows the variation in electronic excitation temperature versus peak applied voltage and the inset graph is the Boltzmann's plot of excitation temperature at a peak applied voltage of 9 kV. The electronic excitation temperature is determined by choosing the neutral He atomic lines 388.9, 447.2, 492.2, 501.6, 587.6, 706.5, and 728.1 nm, respectively, under the Boltzmann approximation [28, 29]. As shown in Fig. 6, when the peak applied voltage increases from 6 to 10 kV, the electronic excitation temperature linearly increases from 4355 to 4533 K, which presents a similar tendency with the curve of average power versus peak applied voltage. It may be due to fact that the electrons could obtain more energy from external electric field with the increase of peak applied voltage, so their excitation temperature quickly increases to generate a large amount of active species by inelastic collisions with molecular species.

3.3. Electron temperature

Under the condition of local thermodynamic equilibrium (LTE), the electron temperature T_e (eV) is approximated by electron excitation temperature T_{exc} (K) in reported work [30, 31]. However, the atmospheric pressure DBD plasmas usually work in non-LTE where electron temperature and electron excitation temperature are different. It is therefore necessary to introduce another way for diagnosing the electron temperature. It is demonstrated that the

emission intensity ratio of two helium lines, which involve the transition $3^3S \rightarrow 2^3P$ (706.5 nm, threshold at 20.96 eV) and transition $3^1S \rightarrow 2^1P$ (728.1 nm threshold at 21.21 eV), could be used to determine the electron temperature which assumes that the electrons obey a Maxwellian electron energy distribution [32, 33]. Under our experimental condition, He excited states (3^1S) and (3^3S) are produced from the ground state by a direct electron impact with atomic helium, and the collisional induced quenching reactions of He excited states (3^1S) and (3^3S) with nitrogen, oxygen, and water molecules are neglected because the density of these molecules is much lower than that of helium in the bubbles. Considering the collisional induced quenching of He excited states (3^1S) and (3^3S) in helium, the intensity of He line 706.5 nm could be expressed as

$$I_{706.5} = C_{706.5} h\nu_{706.5} A_{ij}^{706.5} n_e k_e^{706.5} n_{He} (A_i^{706.5} + k_q^{706.5} n_{He})^{-1} \quad (1)$$

The intensity of He line 728.1 nm is given by

$$I_{728.1} = C_{728.1} h\nu_{728.1} A_{ij}^{728.1} n_e k_e^{728.1} n_{He} (A_i^{728.1} + k_q^{728.1} n_{Ar})^{-1} \quad (2)$$

where the quantity C contains all the optical and geometrical parameters; $h\nu$ is the photon energy of respective transition; A_i denotes the sum over all optical transition probabilities A_{ij} for the excited state and is equal to the reciprocal of radiative lifetime τ_0 ; and A_{ij} is equal to $A_i = 1/\tau_0$, since the only optically allowed transition from each of the excited states is observed, respectively [34]. n_e and n_{Ar} denote the electron and argon density and the argon density n_{Ar} is determined by the ideal gas equation. $k_q^{706.5}$ and $k_q^{728.1}$ denote the quenching rate coefficients of He excited states (3^1S) and (3^3S). The data of radiative lifetimes and quenching rate coefficients for He excited states (3^1S) and (3^3S) are taken from the literatures [35-37].

The electron excitation rate coefficient k_e is determined by integrating the cross sections

over an assumed Maxwellian distribution

$$k_e = \int_0^\infty \sigma(\varepsilon) f(\varepsilon) \sqrt{\frac{2e}{m_e}} d\varepsilon, \quad (3)$$

where $\sigma(\varepsilon)$ is the electron collision cross section for excitation [38], $f(\varepsilon)$ is the electron energy distribution function, and m_e is the electronic mass. Fig. 7 shows the variation in ratio of k_e^{7065} to k_e^{7281} versus electron temperature. Taking the intensity ratio of Eq. (1) to Eq. (2),

we obtain

$$\frac{k_e^{7065}}{k_e^{7281}} = \frac{C_{7281} h \gamma_{7281} I_{7065} a_{ij}^{7281}}{C_{7065} h \gamma_{7065} I_{7281} a_{ij}^{7065}} \quad (4)$$

with

$$a_{ij}^{7065} = \frac{A_{ij}^{7065}}{A_i^{7065} + k_q^{7065} n_{He}} \quad \text{and} \quad a_{ij}^{7281} = \frac{A_{ij}^{7281}}{A_i^{7281} + k_q^{7281} n_{He}}. \quad (5)$$

Thus, the electron temperature could be determined by a fitting function shown in Fig. 7 and the ratio of k_e^{7065} to k_e^{7281} is calculated using Eqs. (4) and (5). The electron temperature as a function of peak applied voltage is shown in Fig. 8. When the peak applied voltage increases from 6 to 10 kV, the electron temperature increases from 1.15 to 1.96 eV which is much higher than the electronic excitation temperature shown in Fig. 6. However, it is in a good agreement with the electron temperature of 1.87 eV in DBD He plasma generated by ring-to-ring electrodes at atmospheric pressure and 1.4 eV in plane-to-plane He DBD plasma at atmospheric pressure [39, 40]. It is a fact that the electronic excitation temperature is equal to the electron temperature only when LTE is valid. Furthermore, the electron temperature and electronic excitation temperature of DBD plasmas present a large difference and normally satisfy the relation of $T_e > T_{exc}$ in non-LTE [39]. It may be due to that the helium inelastic collisions cause a decrease of helium excitation temperature, so the relation $T_e > T_{exc}$ is

satisfied [41].

3.4. Electron density

Stark broadening of hydrogen Balmer line, H_α (656.3 nm), could be used to determine the electron density with a reasonable accuracy when its value is higher than $\sim 10^{14} \text{ cm}^{-3}$ [32-44]. Under our experimental condition, the broadening of recorded H_α line results from the contributions of Stark and Van der Waals, instrumental, and Doppler broadenings. The Doppler and instrumental broadenings cause the appearance of Gaussian profiles and the Stark and Van der Waals broadenings could be approximated by Lorentzian profiles. The observed profile is the convolution of Gaussian and Lorentzian profiles and assumed to be a Voigt function. The FWHMs (full-width at half-maximum) of Gaussian and Lorentzian profiles are related to the fore-mentioned broadenings and could be obtained 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 $\Delta\lambda_V$ represent the FWHMs of Doppler, instrumental, Stark, and Van der Waals broadenings, respectively [45].

The instrumental broadening is calibrated by a He–Ne laser at 632.8 nm and $\Delta\lambda_{inst}$ is determined by a Gaussian fitting to be about 0.06 nm. The FWHM of Doppler broadening is determined from the gas temperature T_g [46]

$$\Delta\lambda_D \text{ (nm)} = 7.16 \times 10^{-7} \lambda_0 \left(\frac{T_g}{M} \right)^{0.5}, \quad (6)$$

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

The FWHM of Van der Waals broadening could be expressed by formula [43]

$$\Delta\lambda_V \text{ (nm)} = \frac{5.736}{T_g^{0.7}}. \quad (7)$$

The FWHM of Stark broadening is related to the electron density by [44]

$$\Delta\lambda_s \text{ (nm)} = 0.549 \times \left(\frac{n_e}{10^{17}} \right)^{0.67965}, \quad (8)$$

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

Accordingly, the electron density is obtained by the FWHM of Stark broadening which could be obtained from Voigt profile by separating (deconvolution) other broadening components. Fig. 9 shows the Voigt fitting of H_α line profile at a peak applied voltage of 9 kV and the electron density versus peak applied voltage. When the peak applied voltage increases from 6 to 10 kV, the electron density linearly increases from 2.56×10^{14} to $5.76 \times 10^{14} \text{ cm}^{-3}$. This result seems reasonable if compared with the electron density of other work, for example, the electron density is $2 \times 10^{14} \text{ cm}^{-3}$ in DBD He plasma generated by needle-to-plate electrodes at atmospheric pressure [47] and the electron density was in the order of magnitude of 10^{14} cm^{-3} in atmospheric DBD He plasma produced by needle-to-ring electrodes [37].

4. Conclusions

An atmospheric DBD helium plasma generated by tube-plate electrodes in water has been studied. By the electrical and spectral diagnosis it was found that the conduction current took up a large proportion of total current and the electron temperature was much larger than electronic excitation temperature. When peak applied voltage increases from 6 kV to 10 kV, the average power, rotational temperature of OH, electronic excitation temperature, electron temperature, and electron density were in the range of 3.65-11.13 W, 427-529 K, 4355-4533 K, 1.15-1.96 eV, and 2.56×10^{14} - $5.76 \times 10^{14} \text{ cm}^{-3}$, respectively. Moreover, the optical emission spectrum showed that the active species such as O^* and OH^* were produced in DBD He plasma, thus the tube-to-plate DBD helium plasma device could be used for water purification.

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Caption of figures

Fig. 1. Schematic diagram of experimental setup and discharge photograph at a peak applied voltage of 8.2 kV and Ar flow rate of 2 L/min.

Fig. 2. Applied voltage and total current waveforms (a) and displacement and conduction

currents waveforms (b) at a peak applied voltage of 8.2 kV.

Fig. 3. Average power versus peak applied voltage.

Fig. 4. 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. 5. Optical emission spectrum of 250-550 nm (a) and 550-850 nm (b) at a peak applied voltage of 9 kV.

Fig. 6. 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. 7. Ratio of electron excitation rate coefficient k_e^{7065} to k_e^{7281} as a function of electron temperature.

Fig. 8. Electron temperature versus peak applied voltage.

Fig. 9. Voigt fitting of H_α line profile at a peak applied voltage of 9 kV (a) and electron density versus peak applied voltage (b).