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Characteristics of OH production by O_2/H_2O pulsed dielectric barrier discharge

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ABSTRACT

We studied characteristics of OH production by pulsed dielectric barrier discharge (DBD) with O_2/H_2O gas mixtures. We found that the light emission from the O_2/H_2O DBD was due to OH (A–X) transition. Therefore, it is allowed to measure the spatial distribution and the intensity of OH (A–X) by simply direct photography of discharge image. The OH (A–X) intensity became weaker approaching the dielectric barriers and away from the electrodes along the direction of the axis of and the surface of the electrodes, respectively. Regardless of the specific operating parameters, the intensity of OH (A–X) linearly increased with discharge power. At a similar level of the discharge power, increasing the applied voltage was most effective for the OH production. In addition, we found that the production of OH radical could be enhanced by increasing O(1D) radical. Our study not only introduced a simple method for the semi-quantification of OH (A–X) in the O_2/H_2O DBD, but also suggested a way for the optimization of OH production in a plasma process.

1. Introduction

Today, non-thermal plasma (NTP) is widely applied in the field of material synthesis, surface treatment, plasma medicine, environmental remediation, and fuel reforming [1–5]. In NTP, the produced chemically active species, such as energized electrons, ions, atoms, radicals and reactive molecules, are responsible for plasma-induced reactions [6]. Hydroxyl (OH) radical is a one of the most important reactive species because it has very strong oxidation ability. For the plasma medicine, the OH radical is especially important because its excited state can emit a light peaked at 308 nm, which is the most effective radiation wavelength for the treatment of psoriasis [7].

Hydroxyl radical generation has been studied with various gas discharges and different diagnostic technologies. Using laser-induced fluorescence (LIF) of OH, highly time and spatially resolved measurements have been reported with Ar/H_2O [8], He/H_2O [9], $N_2/O_2/H_2O$ [10] and $N_2/O_2/H_2O$/HCs [11] plasma. Nevertheless, the LIF-based technology requires a rather complicated experimental arrangement. Considering this fact, some alternative methods have been adopted to measure the OH radical. The spatially distribution of OH was investigated in humid air [12] and $N_2/O_2/H_2O$ [13] corona discharge using optical emission spectroscopy. The quantification of OH radical in Ar/H_2O corona discharge was studied by monitoring CO oxidation [14]. Also in Ar/H_2O discharge, spatially averaged OH concentration was measured by a resonant absorption spectroscopy [15]. Despite great efforts have been devoted, the OH production and consumption pathways in a plasma process are still not well understood. For example, in humid oxygen-enriched gas plasma, OH generation is usually attributed to $H_2O + e \rightarrow H + OH + e$ and $O(1D) + H_2O \rightarrow 2OH$. However, the contribution of each mentioned reaction to the OH generation is still unclear.

Dielectric barrier discharge (DBD), a type of NTP having at least one layer of a dielectric material in between two electrodes to limit electrical current within each microdischarge channel, is characterized as a mature, stable and reliable NTP [16,17]. For traditional AC energized DBD, once an electrical geometry is fixed, increased applied voltage does not result in the increase in the reduced field

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intensity, because a breakdown voltage in the AC DBD is insensitive to the applied voltage [18]. This may limit the application of the AC DBD to where requires a certain range of mean electron energy. Recently, pulsed DBD has been proposed because a breakdown field intensity and consequent mean electron energy in a discharge gap could be varied with the amplitude of the applied voltage [19].

In this paper, we report the characteristics of OH production by pulsed DBD with O_2/H_2O gas mixtures. Time-resolved discharge emission spectra were measured by a monochromator equipped with a streak camera. By using a digital single-lens reflex camera and Image-Pro+ software, the spatial distribution and intensity of OH (A–X) were investigated. The experimental results were evaluated in terms of the applied peak voltage (U_{peak}), the frequency (f), the discharge gap distance (d), and the relative humidity (RH). OH production and consumption pathways were discussed based on the results of a numerical analysis of O_2/H_2O discharge. Detailed physical characteristics of DBD were also discussed.

2. Experimental setup

Fig. 1 shows a schematic diagram of experimental setup. Our experiment was carried out in a plate-to-plate DBD reactor, which was composed of two stainless electrodes (Φ10 mm) and two alumina plates (50 × 50 × 1 mm^3). The two alumina plates were spaced by alumina spacers (50 × 17.5 mm^2) with thickness from 1 to 7 mm to form a discharge space with different gap distances. The DBD reactor was installed in an acrylic resin box having two quartz windows for light-detection, and two holes for gas flow in and out.

The DBD reactor was energized by a pulsed power source (DP-30K10, Peec) with 3 μs in full width at half maximum (FWHM) and 1 μs in rise time. A digital oscilloscope (TDS7104, Tektronix) was used for voltage–current characterization. The applied voltage was measured by a 1000:1 high voltage probe (P6015A, Tektronix), while the discharge current was monitored by a current sensor (A6312, Tektronix). Discharge power was obtained through the time integration of the product of voltage and current, multiplied by an applied frequency.

Time-resolved optical emission spectroscopy (OES) was carried out with a monochromator (C5094, Hamamatsu) equipped with a streak camera (C7700, Hamamatsu) and a grating (100 grooves/mm). The calibration of the monochromator was performed using a mercury lamp. The monochromator system was synchronized to the power source using a signal delay generator (DG535, Stanford Research Systems). A digital single-lens reflex camera (EOS7D, Canon) installed with a quartz lenses (EF-S 18-135 mm f/3.5–5.6 IS, Canon) was used for direct photography.

Mass flow controllers (D07 series, Sevenstar) were used to control the flow rate of oxygen (99.999%, Wugang) gas. To control the humidity, a portion of the oxygen gas was bypassed to a bubbler filled with deionized water. The temperature of the bubbler was kept constant at 298 K. A desired relative humidity was obtained by controlling the ratio of the humidified oxygen gas to the dry oxygen gas. The total gas flow rate was fixed at 100 ml/min.

3. Experimental results

A typical voltage and current waveform for a single-pulse applied voltage is shown in Fig. 2. A group of current spikes could be identified in the primary voltage pulse. These spiky current correspond to filamentary microdischarges in the discharge gap. They were randomly distributed in time and each spike had about a 10 ns duration. It appears that the intensity of the discharge increased with as the applied voltage increased. The discharge pattern from the pulsed DBD with a mixture of O_2/H_2O appeared to be in the typical filamentary microdischarge mode [20] of DBD. After the primary voltage peak, several parasitic voltage peaks caused by voltage reflections from the primary pulse were observed. When their amplitudes return to a size large enough to breakdown a pre-ionized medium, the resultant reflected voltage could induce a secondary discharge [5].

Fig. 3 shows typical discharge images for the single-pulse applied voltage. The images were taken by a streak camera when the grating was centered at 0 nm (grating acts like an ordinary mirror). No light emission could be detected with the O_2 DBD, while an obvious discharge light emission was observed at around 30 μs with the O_2/H_2O DBD (RH = 100%).

To indentify the observed light, the grating was then centered at 500 nm to cover possibly interested spectral range for the O_2/H_2O DBD. Time averaged (over 100 μs) discharge emission spectra are shown in Fig. 4 which also includes the spectra of O_2 DBD for a comparison purpose. As expected, no molecule band or atomic line could be observed with the pure oxygen DBD. The same phenomenon was reported for O_2 DC corona discharge [21]. When water vapor was added, we observed a band centered at 308 nm which

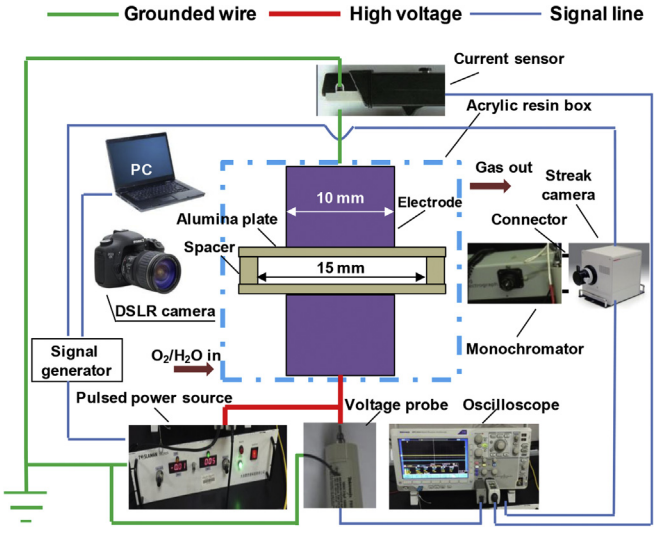

Fig. 1. A schematic of experimental setup.

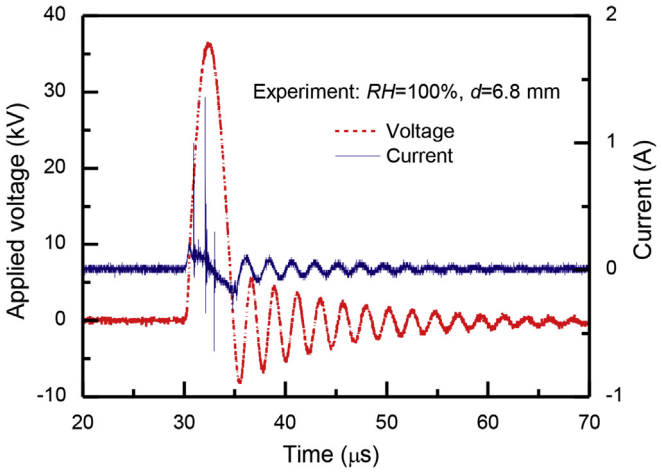

Fig. 2. Typical voltage and current waveform at d = 6.8 mm and RH = 100%.