Wastewater treatment by dielectric barrier discharge plasma

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Abstract. This paper presents the influence of atmospheric non-thermal plasma on household wastewater by using dielectric barrier discharge (DBD) plasma generator. The model was designed in a vertical coaxial tube shape. The high voltage was supplied from high frequency AC power supply to the model for plasma generation at 0 (control group), 10, and 15 kV. Wastewater was released from the top of glass tube passing through DBD corona discharges that was generated between the needle tips and the glass tube, flowed down to the storage tank, and pumped to the top of the model again. The treatment process was operated for 30 minutes with the flow rate of water at 2 min/L. From the experimental results, it shows that all plasma treatment conditions could improve the wastewater quality. The best results were found at 15 kV plasma treated wastewater; the alkalinity, dissolved oxygen (DO), and conductivity were increased from the control group for 10.48%, 10.09%, and 17.79%, respectively. In addition, biochemical oxygen demands (BOD), and chemical oxygen demand (COD) were decreased from the control group for 7.5 times and 37.5%, respectively. An offensive odor of wastewater could be also improved.

1. Introduction
Water usage is probably the most daily important consumption in every household, for example; drinking, cooking, bathing, washing clothes and dishes, flushing toilets, and others. Inevitably, the water quality of post-usage water is consequently degraded causing wastewater. However, household wastewater is neglectly concerned, and usually released to the natural river without pre-treatment process resulting in environmental pollution [1-2].

Generally, wastewater remediation has many treatment processes such as physical, chemical, and biological wastewater treatment [2-4]. However, some organic pollutant in wastewater could not be completely treated by the aforementioned processes. Recently, many extensive applications of plasma technology with liquid solution have been continuously developed, such as plasma activated water for biological and medical applications, decontamination of volatile organic compounds, and purification of wastewater [5-10].

Dielectric barrier discharge (DBD) plasma is one of plasma technique that is suitable for a variety of applications including wastewater treatment [11]. This is due to the fact that the plasma generated by DBD generator is not in-contact with the high voltage electrode; therefore, it is safe to be applied on the sensitive and conductive material [12-14]. Moreover, many useful active, reactive species, and UV radiations could be effectively produced by DBD technique comparing to that of other plasma techniques [13-15].

In this paper, the DBD technique has been proposed for wastewater treatment. The plasma model was designed for circulated wastewater treatment process. It is aimed to be able to be inserted in
household sewers for pre-treatment process before releasing it to the natural water source. In this time, the parameters affected on the wastewater treatment efficiency, which are treatment time and plasma generation supplied voltage, were studied. The electrical characteristics of plasma and optical emission spectroscopy (OES) were also discussed.

2. Experimental setup

The schematic drawing of experimental setup and DBD plasma generator are shown in figure 1. This model is in a vertical coaxial tube shape. 12-cm-diameter outer glass tube and 10-cm-diameter outer aluminum tube are bonded with 80 aluminum needles on the inner surface of the tube, called an anode. The inner aluminum tube, acting as a cathode is covered with the 2-mm-thick glass tube. A gap between the tip of needle and the outer surface of glass tube is 1 cm.

The electrical characteristics of discharge current and voltage are observed by oscilloscope (Siglent SDS2304). The high-voltage probe (PINTEX HVP-18HF) is connected across the plasma generator for discharge voltage measurement. The discharge current is calculated from the voltage drop across 3.33-Ω monitored resistor, placed between cathode and ground.

The cycle of treatment process has started by supplying 14.6 kHz AC high voltage source to the anode, causing the air plasma generated between electrode gaps. Afterwards, the household wastewater was released on the top of glass tube, flowed down through air DBD corona discharges generated between the needle tips and the glass tube. The treated wastewater will flow down to the storage tank, and be pumped back to the top of the model again.

Figure 1. (a) The schematic drawing of experimental setup and (b) DBD plasma generator.
The treatment process was operated for 10, 20, and 30 minutes with 2-min/l water flow rate while the AC high voltage was supplied to the plasma generator at 0 (control group), 10, and 15 kV. All experimental groups were studied on the alkalinity, temperature, conductivity, dissolved oxygen (DO), biochemical oxygen demands (BOD), and chemical oxygen demand (COD). The results of plasma treated wastewater in comparison with the control group were also analyzed.

3. Experimental results

3.1. Electrical characteristics

Figure 2 shows the electrical characteristics of discharge voltage and current during plasma irradiation at 15-kV supplied source voltage by comparing the plasma irradiation between released wastewater (with water) and non-released wastewater (without water) to the system. Micro discharge currents had been observed in each interval of negative and positive rising edge of discharge voltages for both cases. However, the micro discharges could be effectively generated in the positive rising edge compared to that of negative rising edge. The electrical characteristics of plasma without released wastewater case have higher maximum discharge voltage and current than that of the released wastewater case. Releasing of wastewater covering the glass tube could decrease electrical resistance between electrode gap, resulting in discharge voltage reduction. That is because the wastewater has more conductivity than the air.

![Figure 2. The electrical characteristic of discharge voltage and current during plasma irradiation at 15 kV consist of (blue) Non-released wastewater and (red) Released wastewater.](image)

| Supplied AC source voltage (kV) | Discharge voltage (kV<sub>rms</sub>) | Discharge current (A<sub>ms</sub>) | Peak of discharge current (A) |
|---------------------------------|-------------------------------------|----------------------------------|------------------------------|
| 10                              | 3.05                                | 1.49                             | 2.40 - 7.20 and (-3.00) - 0  |
| 15                              | 5.04                                | 1.63                             | 4.20 - 11.41 and (-8.40) - (-0.60) |

The discharge voltage and current during the wastewater treatment with the plasma at 10 and 15-kV supplied source voltage in table 1 demonstrates that the increase of supplied source voltage from 10-kV to 15-kV can increase discharge voltage (V<sub>ms</sub>), discharge current (I<sub>ms</sub>), and peak of discharge current.
current ($I_p$). The $V_{rms}$, $I_{rms}$, and $I_p$ were increased for 65.24%, 9.40%, and 58%-75%, respectively. From the data shown in this table, an increase of discharge current would produce more amounts of radical species, which is benefit for better wastewater treatment.

3.2. Plasma characteristic

Figure 3 displays the optical emission spectroscopy (OES), measured by using a CCD spectrometer (Newport 71S100087) during the wastewater treatment at 10 and 15-kV supplied source voltage for observing radical species in plasma. The air plasma generation can produce several reactive nitrogen species (RNSs) and reactive oxygen species (ROSs) such as hydroxyl ($OH^-$) radical (306-315 nm), oxygen (O) radical (780-790 nm), nitric oxide (NO), and nitrogen (N2) radical (300-400 nm) [16-19]. As shown in the figure, the radical species at 15-kV supplied source voltage have more relative intensity than that of radical species at 10-kV supplied source voltage due to the higher discharge current. Since the RNSs and ROSs play an important role in oxidative destruction of organic pollutant in the wastewater; therefore, the usage of 15-kV supplied source voltage for plasma generation is expected to be the suitable wastewater treatment condition.

![Figure 3. The OES of air plasma during plasma wastewater treatment at 10 and 15-kV supplied source voltage.](image)

3.3. Wastewater treatment analysis

The first investigated parameter was alkalinity, which was measured by pH meter (Horiba U–10). The 50-ml wastewater sample was measured by pH meter for 2 min. As shows in figure 4, the experimental results indicate that all plasma treated wastewater conditions have more alkalinity than that of the control group. The increase of treatment times increases alkalinity of the treated wastewater. The alkalinity of treated wastewater at 10 and 15-kV supplied source voltage is increased from the control group for 3.54% and 10.48%, respectively. The best condition for alkalinity improvement of wastewater is 15-kV supplied source voltage with 30-min treatment time, which is 7.80. This is due to the fact that the hydroxides ($OH^-$) will react with alkali metals and alkaline earth metals in household wastewater, which affects on alkalinity of wastewater after treatment. These beneficial radicals can be produced from $NO_3^-$, $NO_2^-$, and $O_3$ that are generated from air plasma [20-21], and can be also produced from water molecule ($H_2O$) activated by air plasma [22].
Figure 4. The alkalinity of plasma treated group and control group.

Figure 5 explains the results of chemical oxygen demand (COD) that was measured from the plasma treated wastewater and control group by COD investigation methods. All treated wastewater and control group were brought into the chemical measurement process. The experimental results indicated that all plasma treated groups have better COD than the control group. The COD decreases when the treatment time increases at 15-kV supplied source voltage, but it slightly changes at all treatment times at 10-kV supplied source voltage. At 30-min treatment time, the COD of the treated wastewater at 10 and 15-kV supplied source voltage are improved from the control group for 25% and 37.5%, respectively. The best condition for COD enhancement of wastewater is 15-kV supplied source voltage with 30-min treatment time, which is 4000 mg/l while the control group is 6400 mg/l.

Another important parameter used for wastewater quality investigation is dissolved oxygen (DO) which is measured by DO meter (Si-Analytics HandyLab-2). As shows in the figure 6, the plasma treated wastewater groups contain better DO than that of the control group. It could be noticed that the DO increased with increase of treatment time. At 30-min treatment time, the DO of the treated wastewater at 10 and 15-kV supplied source voltage are improved from the control group for 7.05% and 10.09%, respectively. The best condition for DO improvement is 15-kV supplied source voltage with 30-min treatment time, which is 7.96 mg/l (control group is 7.23-mg/l).

Figure 5. The COD of plasma treated wastewater group and control group.
Figure 6. The DO of plasma treated wastewater group and control group.

Figure 7 shows the results of biochemical oxygen demand (BOD), measured from the treated wastewater group and control group by BOD measurement methods. The treated wastewater was brought into the measurement process to measure DO value in the 5th day (DO5). Then, the DO5 is calculated together with DO value at the 1st day (DO0) by finding products between the differences of DO5 and DO0, and 100 for BOD results. From the experimental results, plasma treated wastewater groups produced better BOD than that of the control group. The BOD continuously decreased when the treatment time increased. At 30-min treatment time, the BOD of plasma treated wastewater at 10 and 15-kV supplied source voltage are improved from the control group for 3.53 and 7.5 times, respectively. The best condition for BOD enhancement is 15-kV supplied source voltage with 30-min treatment time, which is 16 mg/l (control group is 120-mg/l).

Figure 7. The BOD of plasma treated wastewater group and control group.

The conductivity of treated wastewater and control group were measured by conductivity meter (Horiba U–10). As shows in figure 8, all treated wastewater groups have more conductivity than that of the control group. The increase of treatment time and supplied source voltage can increase the conductivity of the wastewater. The conductivity of treated wastewater at 10 and 15-kV supplied source voltage for 30-min treatment time are increased from the control group for 5.53% and 17.79%, respectively. This is due to the fact that the hydroxides (OH⁻) radicals, RNSs, and ROSs play a significant part in the oxidative destruction of organic pollutant, which affect on organic molecule size.
reduction. The small organic molecules could be dissolved well in water, resulting in an increase of water conductivity.

Figure 8. The conductivity of plasma treated wastewater group and control group.

4. Conclusion
This paper aims to apply the proposed model for household wastewater treatment. From the experimental results, it could confirm that the air DBD plasma can improve household wastewater. It could be noticed that the RNSs and ROSs, which take on an important role in the wastewater improvement, could be effectively produced at the higher supplied source voltage. The best condition for the wastewater improvement was at 15-kV supplied source voltage with 30-min treatment time; the alkalinity, DO, and conductivity were improved from the control group for 10.48%, 10.09%, and 17.79%, respectively. The BOD and COD were enhanced from the control group for 7.5 times and 37.5%, respectively.

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