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Effects of the Liquid Conductivity on Pulsed High-voltage Discharge Modes in Water

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Abstract: Spark, stream and corona pulsed high-voltage discharges in water induced by the various initial conductivities have been examined in this paper. The discharge modes changed from spark to corona discharge with the liquid conductivity increasing. The apparent production of OH radical and quantum yield generated by spark discharge in distilled water were 11.57 μmol/L and 0.0978 photon/s, respectively. A preliminary study on acid fuchsine (AF) treatment indicated that higher AF removal efficiency has been achieved by spark discharge. The process of degradation showed that the oxidative effects through OH radical oxidation did not play an important role and did increase with the discharge mode changing to spark discharge.

Keywords: Pulsed high-voltage discharge, spark discharge, corona discharge, quantum yields.

Electrical discharge produced by pulsed high-voltage is a newly developed method for degradation of organic pollutants in water[1, 2]. High degradation efficiencies of phenol[3, 4], acetophenone[5] and organic dyes[6, 7] in aqueous solutions have been achieved using corona discharges. And other discharge modes formed by adjusting the electrical gap distance[8]. Clements *et al.*[9] found that the streamer length decreased as the water conductivity rose from 10 to 125 μS/cm and maximum streamer length obtained at water conductivity of 8 μS/cm. However, few studies paid attention on the discharge modes induced by various initial conductivities in water and effects of them on organic pollutant degradation. As industrial wastewater is generally conductive, it is essential to explore the form of pulsed high-voltage discharges with various initial conductivities.

A basic mechanism for the treatment of organic compound is considered to proceed through oxidative pathways in which the active species, such as OH radical, play an important role, photolysis and pyrolysis pathways in which the physicochemical characteristics of solution had little effects on the organic compounds degradation[1]. The objective of the paper is to probe the discharge modes induced by various initial conductivities and to determine the production of the active species such as OH radical and the quantum yields on the intensive ultraviolet from the pulsed discharge plasma. Acid fuchsine (AF), one of the biorefractory dyes, is selected as the model pollutant in order to explore the degradation ability and the possible degradation mechanism.

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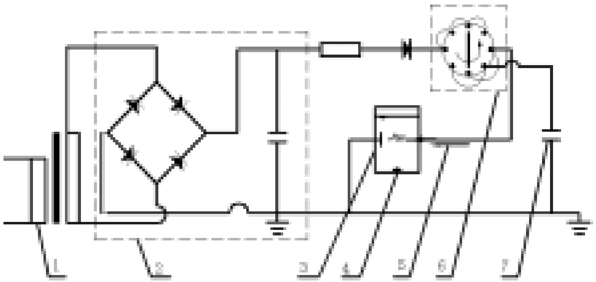
Figure 1 Schematic diagram of the experimental apparatus



1. Transforming double spark gap, 2. Voltage rectifying circuit, 3. Reactor, 4. Magnetic stirring bar, 5. Air injected, 6. Rotating double spark gap, 7. Storage capacitor.

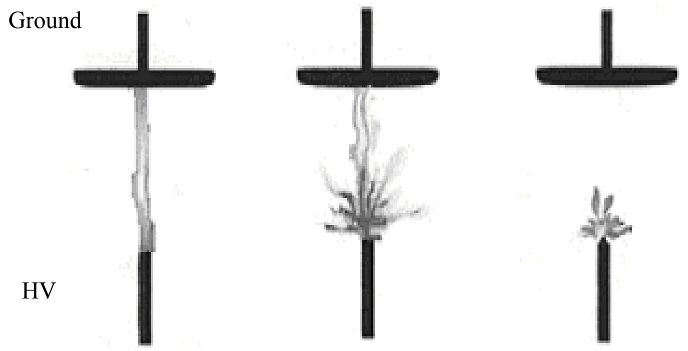
Figure 1 showed the schematic diagram of the experimental apparatus. The pulsed power supply consisted of a transformer (10 KVA), voltage rectifying circuit, rotating double spark gap, and storage capacitance (2000 pF). The reactor was a glass cylinder with 50 mm inner diameter containing electrodes in a point-to-plane geometry. The point electrode was a stainless hypodermic needle (common injection needle 7 #) inserted through silicon sealant. The needle tip was 30 mm from the copper disk as a ground electrode. Air generated by the brake compressor was continuously injected into the reactor through the tip of the needle electrode. There was a magnetic stirring bar at the bottom of the reactor to keep the solution well mixed.

Such a spark configuration was used to separate the charging and discharging phases of the capacitor. When the first gap was closed, the storage capacitor was charged and subsequently, when the first gap was opened and the second one was closed, the capacitor was switched to the electrode system. The experiments for determination of active species were taken in distilled water. A little KCl was added in order to change the liquid conductivity. AF, a usual dye, was dissolved in distilled water at initial concentration of 50 mg/L for the degradation experiments. The concentration was analyzed by UV-visible absorption spectroscopy at λ=544 nm on a spectrophotometer (Techcomp UV8500). The apparent production OH radical was calculated indirectly by with the absorbance value at 508 nm[10], since OH radical could oxidate $Fe(phen)_3^{2+}$ to $Fe(phen)_3^{3+}$. The quantum yields were determined by colorimetry using ferric oxalate[11].

Figure 2 presented the effects of initial conductivities on electrical discharge modes, spark, stream and corona discharge, where initial conductivities in distilled water were 12 μS/cm, 70 μS/cm and 300 μS/cm respectively. The results indicated that streamer production and propagation were affected by initial conductivity and streamer length decreases evidently along with the increase of the conductivity, which coincided Clements's conclusion. Streamer could continuously touch the counter electrode thus bridging the gap and forming spark discharge when the conductivity was 12 μS/cm.

Effects of various discharge modes on the active species generated, such as OH radical, were shown in the **Figure 3**. The production of active species increased with discharge time, and after 20 min it decreased slowly, because OH radical reacted each other and formed H_2O_2 at the termination reaction stage. Joshi[12] obtained the similar

Figure 2 Schematic diagram of electrical discharge modes on various initial conductivities



(a) Spark discharge with conductivity of 14 μS/cm; (b) Stream discharge with conductivity of 70 μS/cm; (c) Corona discharge with conductivity of 300 μS/cm

Table 1 Quantum yields on various reaction systems

Reaction systems	Quantum yields (photon/s)
Spark discharge	0.0978
Stream discharge	0.0478
Corona discharge	0.0385

results. The production in case of spark discharge was higher than that in corona discharge. After 30 min discharge treatment, the apparent production of OH radical was 11.57 μmol/L, 9.87 μmol/L and 6.01 μmol/L, respectively, in sequence by spark, stream and corona discharge. In order to assess the intensity of ultraviolet from the pulsed discharge plasma, quantum yields were determined. It can be seen from **Table 1** that quantum yield of 0.0978 photon/s in case of spark discharge was two times more than 0.0385 photon/s in corona discharge, which indicated that the ultraviolet intensity was higher in case of spark discharge. In conclusion, the power efficiency of spark discharge was significantly higher than that of other discharges. The mode of spark discharge was adopted at later experiments.

Similarly AF removals were dependent on discharge modes. From **Figure 4**, AF removal was 97.8% in case of spark discharge, and it was expected to be significantly higher than 48.6% in case of corona discharge. In order to explore the effects of OH radical oxidation on AF degradation, 0.0854 mmol/L Na_2CO_3, one of the most representative natural OH radical scavengers[12], was added. From **Figure 4**, it can be observed that the presence of scavenger really inhibited the degradation rate. AF removals were 45.6% and 79.4% in the presence of scavenger in case of corona and stream discharges, which fell 3% and 8% respectively as compared with those in the absence of scavenger. These results indicated that in the process of AF degradation the oxidative pathways through OH radical oxidation did not play an important role in case of mixed and corona discharge, and the effects of OH radical oxidation did increase with the discharge mode changing to spark discharge.

Figure 3 Effects of various discharge modes on apparent production of OH radical

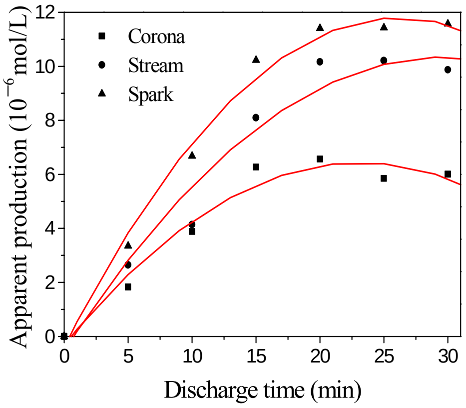
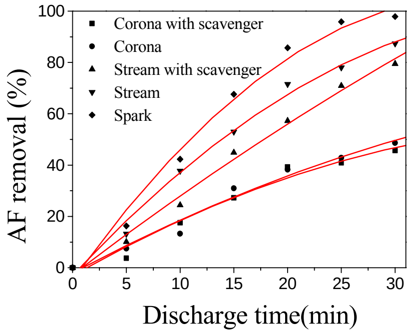


Figure 4 Effects of various discharge modes on AF removal



In conclusion, the discharge modes were subjected to the liquid conductivity. Higher power efficiency can be achieved in case of spark discharge, and OH radical oxidation did not play an important role in case of stream and corona discharge. This technology of pulsed high-voltage discharge is certainly a positive method to achieve quick mineralization. It will be expected to be valuable for environmental research and application. Further work on effects of photolysis and pyrolysis and identification of byproducts is required.

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