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# Treatment of Azo Dye (Acid Orange II) Wastewater by Pulsed High-Voltage Hybrid Gas-Liquid Discharge

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## Abstract

Pulsed high-voltage electrical discharge was used in treating azo dye (Acid Orange II, AO7) wastewater. Effects of initial pH,  $\text{Fe}^{2+}$  concentration, discharge mode, conductivity of initial aqueous solution and the type of bubbling gas (air,  $\text{O}_2$ ,  $\text{N}_2$ ) on AO7 degradation were studied. A new gas-liquid discharge (NGL) mode, by which a plate was exposed in air and needles were emerged in liquid, displayed a remarkably better AO7 degradation than gas-liquid discharge (GL) mode and liquid discharge (L) mode. The lower conductivity of aqueous solution and the higher oxygen concentration in bubbled gas were benefit to degradation process. In addition, owing to the multi-point structure and self-provided  $\text{Fe}^{2+}$ , the electrode with iron mesh-plate discharge showed more competitive degradation performance than that with multi-needle-plate discharge. The oxidant of  $\cdot\text{OH}$  and  $\text{H}_2\text{O}_2$  induced by the discharge process were also studied to explore the degradation processes. By NGL mode with iron mesh electrode and bubbled air in reactor, the degradation efficiency of AO7 was up to 85.8% when the initial pH, conductivity and  $\text{Fe}^{2+}$  concentration were 2.28, 200

$\mu\text{S}\cdot\text{cm}^{-1}$  and  $1.635\text{ mmol}\cdot\text{L}^{-1}$ , respectively.

*Keywords:* Azo dye (AO7), degradation, wastewater treatment, pulsed high-voltage hybrid gas-liquid discharge, iron mesh

## Introduction

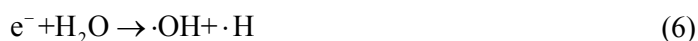
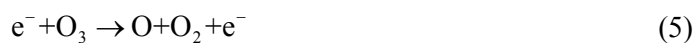
Water is indispensable to life as well as industry. However, wastewater from industry leads an alarming situation that requires immediate attention and effective treatment. It is estimated that 17 to 20% of industrial water pollution comes from textile dyeing and treatment plants<sup>1</sup>. Most of dyes are toxic, and some intermediates, which have potential carcinogenicity and mutagenicity, may be generated during the synthesis and degradation of these dyes. The intermediate products, especially some of the amines, are hard degradable.<sup>1,2</sup> Several methods have been applied to remove the dye components from wastewater, such as ozonization, activated carbon absorption, biodegradation and advanced oxidation processes (AOPs).<sup>3</sup>

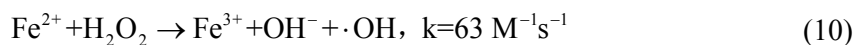
AOPs, which have unique potential in terms of degradation efficiency due to the generation of a specific kind of oxidants, such as  $\cdot\text{OH}$ ,  $\text{H}_2\text{O}_2$  and  $\text{O}_3$ , are also accompanied with UV photolysis, photocatalysis, ozone oxidation, sonochemistry, supercritical water oxidation, electrical discharge plasma technology, Fenton, high-energy electron irradiation, and etc.<sup>4-7</sup> Although most AOPs have succeeded in decolorizing effluents and lowering dye concentration to ppm level, the application of them is still limited due to the low degree of mineralization and the introduction of certain explosive and expensive additives ( $\text{H}_2\text{O}_2$ ,  $\text{O}_3$ ).<sup>8-10</sup>

To overcome the disadvantages, the electrical discharge plasma technology was

proposed in recent years, which can generate a large number of oxidizing species, including  $\cdot\text{OH}$ ,  $\cdot\text{O}$ ,  $\cdot\text{H}$ ,  $\text{H}_2\text{O}_2$ ,  $\text{O}_3$ , etc. As one of the electrical discharge plasma technologies, pulsed high-voltage discharge technology has received much more attention in recent few decades. By this method, various physical and chemical effects can be obtained, such as pyrolysis, UV photolysis, electrohydraulic cavitation, as well as formation of oxidizing species: radicals ( $\cdot\text{H}$ ,  $\cdot\text{O}$ ,  $\cdot\text{OH}$ ) and molecules ( $\text{H}_2\text{O}_2$ ,  $\text{O}_3$ , etc).<sup>11</sup> Meanwhile,  $\text{H}_2\text{O}_2$  and  $\text{O}_3$  could be transformed to  $\cdot\text{OH}$  or other radicals by a series of reactions (Eqs. 1-9).<sup>12</sup> Therefore, in this case, the amount of  $\text{H}_2\text{O}_2$  and  $\text{O}_3$  is vital to dyes degradation.

Due to the limited capabilities of the transformation from  $\text{H}_2\text{O}_2$  and  $\text{O}_3$  to  $\cdot\text{OH}$  and other radicals, many researches added ferrous salt or other metal salts to induce Fenton or Fenton “like” processes.<sup>13,14</sup> The typical Fenton reaction is showed in Eq. 10.





Pulsed high-voltage discharge was widely used to the degradation of textile dyes. Locke et al.<sup>15</sup> used a gas-liquid hybrid reactor to degrade phenol. The results showed that the gas-liquid hybrid discharge was superior to gas discharge and liquid discharge. Grymonpré et al.<sup>16</sup> introduced  $\text{Fe}^{2+}$  into the discharge process to promote phenol decomposition via Fenton reaction (Eq. 10). Wang et al.<sup>17</sup> degraded Acid Orange II (AO7) by using a multi-needle-to-plate high-voltage pulsed corona discharge system, and the results showed that increasing the number of needles could enhance the degradation performance. Zheng et al.<sup>12</sup> used a circulatory airtight reactor system, in which the degradation efficiency could reach up to 91%, to degrade methyl orange (MO) solution. Li et al.<sup>18</sup> combined the pulsed discharge technology with the semiconductor photocatalysis technology by inducing  $\text{TiO}_2$  nanoparticles in degrading AO7.

In this work, Acid Orange II (AO7) was selected as a simulation wastewater containing azo dyes. Effects of initial pH and conductivity, ferrous iron concentration, discharge mode and various bubbled gas types on the degradation of AO7 were investigated and the optimal parameters were obtained. In addition, considering that the iron mesh owns a large number of points like needles and can generate sufficient  $\text{Fe}^{2+}$  at low pH, an iron mesh covered needle-plate discharge, instead of needle-plate discharge, was first applied to degrade the simulation wastewater. A comparison of AO7 degradation by using discharge with additional  $\text{Fe}^{2+}$  and that with iron mesh was then performed. The existence of oxidant of  $\cdot\text{OH}$  and  $\text{H}_2\text{O}_2$  were also studied to

explain the degradation processes.

## Experimental

### Materials and reaction system

The experimental apparatus consists of a pulse power supply and a non-thermal plasma-based water treatment reactor system (Fig. 1). The voltage and output capacity are 30 kV and 3  $\mu$ F respectively. The pulse rising time is less than 100 ns. The reactor is composed of pin-electrodes, cooper plate-electrode ( $\Phi=50$  mm,  $\delta=3$  mm), rubber tube, air pump (ACO-003) and a Plexiglas cylinder ( $\Phi=120$  mm,  $h=180$  mm). There are two flow meters (LZB-10) for the control of gas velocity. Six needles are 16 steel hypodermic needles and the needle tips were encased by silicone insulation, with only 1 mm of the needles were exposed beyond the silicone insulator.

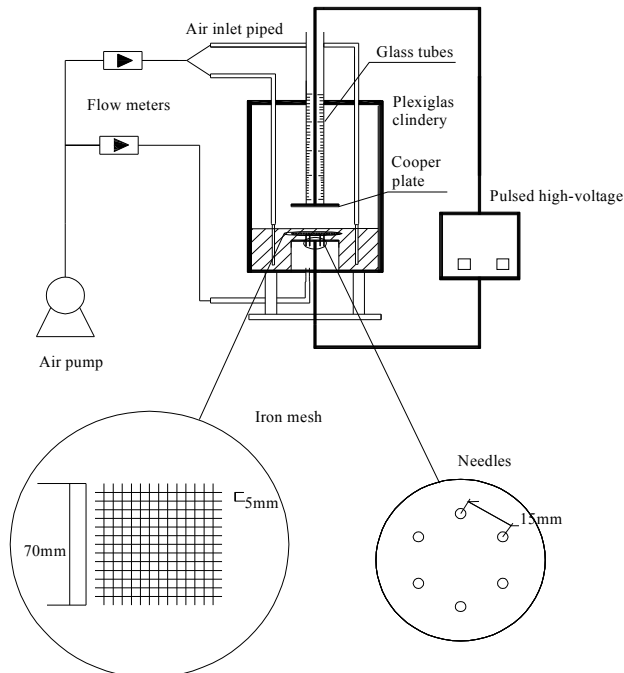


Fig. 1 Schematic of reactor and the details

## Experimental methods and analysis

As it was mentioned above, AO7 was selected as a simulation wastewater of azo dyes. 220 mL 40 mg•L<sup>-1</sup> AO7 solution was poured into the reactor with bubbling gas from needles, which was distributed by the air pump at 400 L•h<sup>-1</sup> and 500 L•h<sup>-1</sup>, respectively. A 70 mm × 70 mm iron mesh was used in this work, which contains grids with the side length of 5 mm. The plate-electrode was vertically placed above the surface of liquid, and needles were submerged in liquid phase. The distance between the plate and needles was 15 mm. H<sub>2</sub>SO<sub>4</sub> solution and Na<sub>2</sub>SO<sub>4</sub> solid were used to adjust the pH of dye solution and the initial electrical conductivity respectively. The green-vitriol was responsible for offering Fe<sup>2+</sup>. The conductivity and pH of the solution were measured by conductivity meter (DDS-11A) and pH meter (Denver UB-7), respectively. The concentration of AO7 solution was determined by calibrated UV-spectrophotometer (Model UV-1800, Shimadzu) with the wavelength of 484 nm. H<sub>2</sub>O<sub>2</sub> was analyzed following titanium potassium oxalate chromogenic method<sup>31</sup>. And the •OH was analyzed by Fe(phen)<sub>3</sub>(2+) spectrophotometry<sup>32</sup>.

The degradation efficiency of AO7 ( $\eta$ ) is calculated as:

$$\eta(\%) = \frac{C_0 - C}{C_0} \times 100\% \quad (11)$$

where  $C_0$  and  $C$  are the initial and residual concentration of AO7 solution, respectively.

## Results and Discussion

### Effect of discharge mode

The pulsed high-voltage discharge in liquid is corona or corona-like discharge, which generates hydroxyl and other hydrogen and oxygen radicals.<sup>19</sup> When it is related to gas discharge and the voltage is high enough, stream discharge and glow discharge might happen, during which ozone and hydroxyl and other radicals could be generated.<sup>15</sup> These radicals can improve the degradation of AO7. The gas-liquid discharge (GL) is conducted with the needles and plate electrodes submerged in liquid and the gas bubbled from both needles electrodes and distributors. The plate electrode is exposed above the liquid surface in NGL, which makes it difference from GL or L.

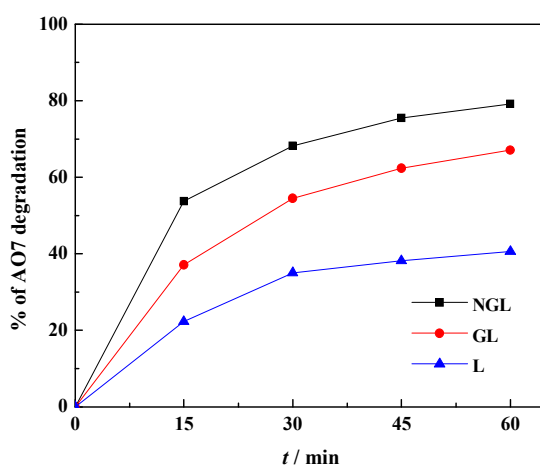


Fig. 2 Effect of discharge mode on AO7 degradation

As shown in Fig. 2, effects of various discharge modes on the degradation of dyes were investigated. The descending order of AO7 degradation efficiencies is NGL, GL and L, which is consistent with the results of literatures<sup>18,20</sup>. Since the bubbled gas led to the generation of ozone which could not be generated in liquid,<sup>21</sup> the degradation efficiency by GL was higher than that by L. In addition, bubbling gas



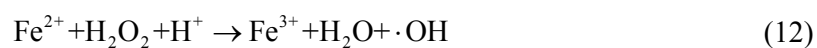
could enhance the reaction of oxide radicals with dye molecular by intensifying the turbulence. The AO7 degradation efficiency by NGL was much higher than that by GL. One reason is that the discharge in gas phase happens more easily than that in liquid phase as the dielectric constant of gas is less than that of liquid.<sup>22</sup> Due to the phenomenon that corona light became intensive, which could be seen easily due to the extra gas zone, the corona discharge became intensive too. The other reason is that much more O<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> are generated in NGL than that in GL and L.<sup>19,23</sup>

### Effect of initial pH value

It is well known that dyes are sensitive to pH.<sup>24</sup> Fig. 3a shows the effect of initial pH on AO7 degradation and the highest degradation efficiency was 79.2% when the pH was 2.28.

The pH of initial solution has an influence on the Fenton reaction, as it can promote the production of •OH. High initial pH leads to low H<sup>+</sup>. Due to the reaction shown in Eq. 12, the generation amount of •OH will be small, which results in low degradation efficiency when the initial pH is high. However, when the initial pH value is lower, AO7 molecular structure could be more stable and hard to be degraded.<sup>25</sup> Thus, with decreasing the pH value from 2.28 to 1.01, a significant reduction of AO7 degradation efficiency was observed. As shown in Fig.3b, the effect of pH on the AO7 degradation follows second-order-kinetics during the present study.

Rates constant is calculated by Eq. (13):



$$\frac{1}{C_0} - \frac{1}{C_t} = k_1 t \quad (13)$$

where  $C_0$ ,  $C_t$  and  $k_t$  are the initial and residual concentration of AO7 solution, and rate constants of kinetics equations, respectively. Table 1 shows the rate constants and  $R^2$  values under different conditions.

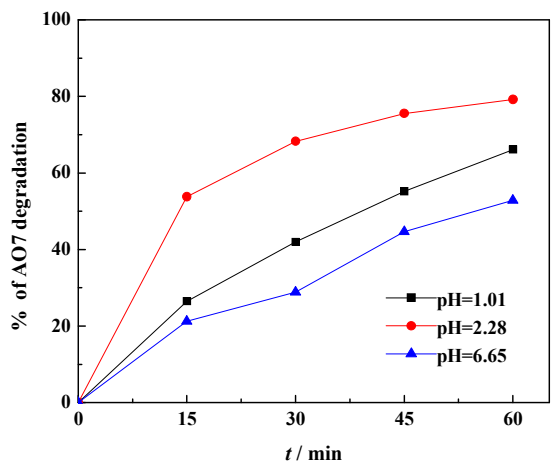


Fig. 3a Effect of initial pH value on AO7 degradation

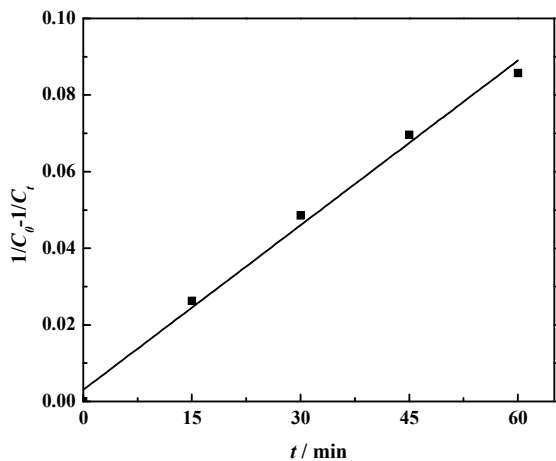


Fig. 3b Second-order-kinetics of  $40 \text{ mg} \cdot \text{L}^{-1}$  AO7 degradation (pH is 2.28)

**Table 1**

Kinetics parameters under different pH

pH	concentration of $\text{Fe}^{2+}$ / $\text{mmol} \cdot \text{L}^{-1}$	degradation at 60 min / %	Rate constant / $(\text{mmol/L})^{-1} \cdot \text{min}^{-1}$	$R^2$
1.01	1.635	66.9	0.0008	0.9757
2.28	1.635	79.2	0.0014	0.9928

6.68	1.635	52.9	0.0004	0.9779
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### Effect of $\text{Fe}^{2+}$ concentration

Fig. 4a shows that various concentration of  $\text{Fe}^{2+}$  resulted in different degradation efficiencies, and the highest degradation efficiency was 79.2% when the concentration of  $\text{Fe}^{2+}$  was  $1.635 \text{ mmol}\cdot\text{L}^{-1}$ . The presence of  $\text{Fe}^{2+}$  significantly enhanced the degradation of AO7 by reacting with  $\text{H}_2\text{O}_2$  to produce more  $\cdot\text{OH}$ . When the concentration of  $\text{Fe}^{2+}$  was  $0.818 \text{ mmol}\cdot\text{L}^{-1}$ , less  $\cdot\text{OH}$  could be obtained than that was  $1.635 \text{ mmol}\cdot\text{L}^{-1}$  based on Eq. 11. The experimental data also shows that AO7 degradation efficiency decreased when iron concentration further increased. According to Eq. 11, there was not enough  $\text{H}_2\text{O}_2$  to react with  $\text{Fe}^{2+}$  when  $2.453 \text{ mmol}\cdot\text{L}^{-1}$   $\text{Fe}^{2+}$  was added. And then the reaction in Eq. 14 would happened.<sup>22</sup> Thus, the amount of  $\cdot\text{OH}$  reduced by reacting with excess  $\text{Fe}^{2+}$ . As a result, the AO7 degradation efficiency reduced. So there should be an optimal value for the initial  $\text{Fe}^{2+}$  concentration in solution. As shown in Fig.4b, the effect of concentration of  $\text{Fe}^{2+}$  on the AO7 degradation follows second-order-kinetics during the present study. Rate constant is calculated by Eq. 13. Table 2 shows the rate constants and  $R^2$  values under different conditions.



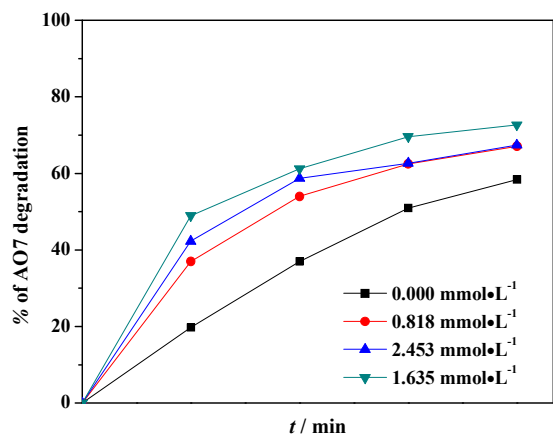


Fig. 4a Effect of ferrous iron concentration on AO7 degradation

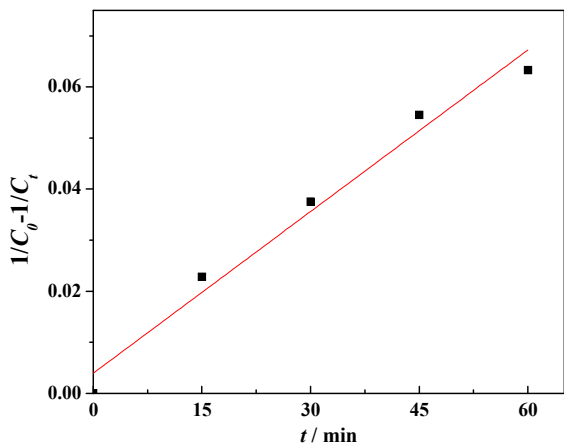


Fig. 4b Second-order-kinetics of 40 mg·L<sup>-1</sup> AO7 degradation at 1.635 mmol·L<sup>-1</sup> Fe<sup>2+</sup>

**Table 2**  
Kinetics parameters under different concentrations of Fe<sup>2+</sup>

concentration of Fe <sup>2+</sup> / mmol·L <sup>-1</sup>	pH	degradation at 60 min / %	rate constant / (mmol/L) <sup>-1</sup> · min <sup>-1</sup>	R <sup>2</sup>
0	2.28	58.4	0.0006	0.9904
0.818	2.28	67.1	0.0008	0.9923
1.635	2.28	72.7	0.0011	0.9790
2.453	2.28	67.4	0.0008	0.9598

Effect of species of bubbling gas

Many studies have proved that higher concentration of oxygen is benefit for degrading dyes.<sup>26,27</sup> As shown in Fig. 5, the degradation efficiency of AO7 with three bubbling gases followed the sequence: Oxygen > Air > Nitrogen. Since little O<sub>3</sub> was generated when nitrogen was bubbled, the degradation efficiency of AO7 with nitrogen bubbling was the lowest. It had been found that more H<sub>2</sub>O<sub>2</sub> and O<sub>3</sub> were generated with O<sub>2</sub> feeding<sup>26,27</sup> than that with other two gas feedings in the beginning of discharge process, according to Eqs. 1-4, and 9. However, excess O<sub>2</sub> might generate excess hydrogen, hydroxyl radical and oxygen atom which could lead to Eqs. 15-17.<sup>26</sup> As a result, the ozone can be consumed and the final degradation efficiency of AO7 by feeding O<sub>2</sub> was close to that by feeding air.

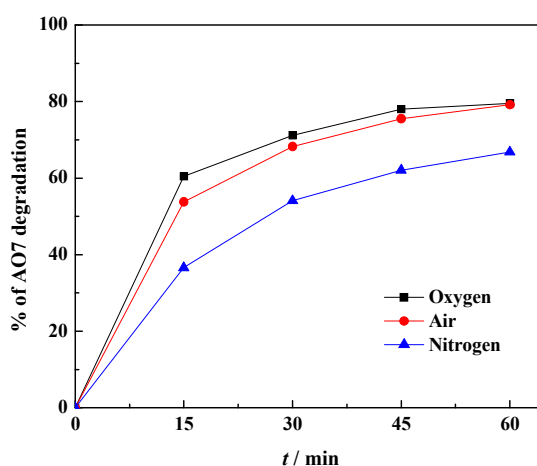


Fig. 5 Effect of species of bubbling gas on AO7 degradation

It can be concluded that higher concentration of oxygen in the bubbled gases can lead to higher degradation efficiency. With the consideration of cost, air is the best choice as the bubbling gas.

### **Effect of initial conductivity**

Liquid conductivity is one of the important parameters which affect the discharge mode (spark discharge, stream discharge, corona discharge) and radical emission intensity.<sup>28</sup> As the conductivity is increasing, the discharge mode changes from spark discharge to stream discharge, and then to corona discharge.<sup>29</sup> During this process, the discharge becomes weaker and weaker, leading to the degradation efficiency of AO7 lower and lower. As the initial conductivity increased from 200  $\mu\text{S}\cdot\text{cm}^{-1}$  to 800  $\mu\text{S}\cdot\text{cm}^{-1}$ , the discharge mode was not converted and the reduction of degeneration efficiency of AO7 was negligible, as shown in Fig. 6. Therefore, the degeneration efficiency of AO7 kept constant. However, the degeneration efficiency of AO7 decreased much more with the initial conductivity increasing up to 1500  $\mu\text{S}\cdot\text{cm}^{-1}$  or even 3000  $\mu\text{S}\cdot\text{cm}^{-1}$ .

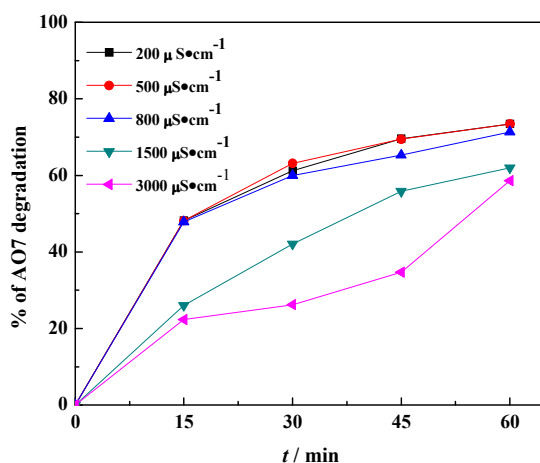


Fig. 6 Effect of initial conductivity on AO7 degradation

### The new type of electrode configuration

The type of electrode configuration is also important to discharge process, such as needle-plate, rod-rod, line-tube, ring-tube, plate-plate, and etc. Nowadays, the needle-plate reactor has attracted more and more attentions. The effect of different numbers of needles on discharge process has been studied by many researchers and the conclusion, that more needles are beneficial to degradation, was finally obtained.<sup>17,29</sup>

When a pulsed voltage is applied to the needle electrode, the accelerating electrons may ionize the bubbled gas and water molecules at the tip of needles in the reactor, forming active radicals, which can react with pollutants in water.<sup>30</sup> Therefore, increasing the number of needles can provide more discharge points, i.e, more radicals can be formed which is beneficial to the degradation of AO7. Thus, the multi-needle-plate reactor, rather than the original single-needle-plate reactor, is selected as the experimental equipment by more and more researchers.

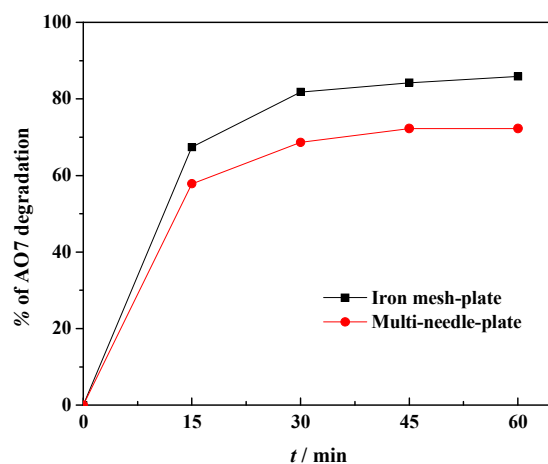


Fig. 7 Effect of different electrode configuration on AO7 degradation

The iron mesh-plate discharge is a type of discharge, in which needles are covered by an iron mesh. It can offer much more discharge points due to the special structure of iron mesh, which contains a large number of joints. Since the number of the iron mesh-plate discharge points is much larger than that of the multi-needle discharge, the degradation efficiency of AO7 by the iron mesh-plate discharge was higher than that by the multi-needle-plate discharge. In addition, the iron mesh could be treated as a source of  $\text{Fe}^{2+}$  at pH 2.28. Thus, the introduction of additional  $\text{Fe}^{2+}$  from other chemicals is unnecessary. Above all, iron mesh-plate discharge was superior to multi-needle-plate discharge. The comparison results of AO7 degradation efficiency by these two types of discharge are shown in Fig. 7.

#### Mechanism of high voltage discharge for the degradation of AO7

Degradation is due to the formation of chemical species, neutral molecules and



molecules in excited states such as highly reactive  $\bullet\text{OH}$ ,  $\text{NO}\bullet$ ,  $\bullet\text{O}_2$ ,  $\text{O}\bullet$ ,  $\text{HO}_2\bullet$ ,  $\text{H}\bullet$ ,  $\text{H}_2\text{O}_2$ ,  $\text{O}_3$ ... The most reactive species is  $\bullet\text{OH}$ , which has a highest oxidation potential. And  $\text{H}_2\text{O}_2$  can also transform into  $\bullet\text{OH}$  by Fenton reaction. So the contents of  $\bullet\text{OH}$  and  $\text{H}_2\text{O}_2$  were analyzed to prove the existence of oxidants. In order to avoid the interference of AO7 molecule, 220 mL deionized water was poured into the reactor as the discharge system. The pH was 2.28 and the concentration of  $\text{Fe}^{2+}$  was  $1.635 \text{ mmol}\cdot\text{L}^{-1}$  in the experiment for detecting  $\bullet\text{OH}$ . The same pH was chosen in the experiment for detecting  $\text{H}_2\text{O}_2$  but without  $\text{Fe}^{2+}$ .

Fig. 8 and Fig. 9 show that the concentration of  $\bullet\text{OH}$  increased with reaction time as well as  $\text{H}_2\text{O}_2$ . The increases of the two oxidants were approximately linear. And the concentrations of  $\bullet\text{OH}$  and  $\text{H}_2\text{O}_2$  were both about  $0.37 \text{ mmol}\cdot\text{L}^{-1}$  when the discharge time was 60min. With the increase of the reaction time, the electrical power injected into the system increased, which could increase the amount of active species (especially  $\bullet\text{OH}$  and  $\text{H}_2\text{O}_2$ ) produced. Then these species could react with AO7 molecule and degrade it into smaller molecules.

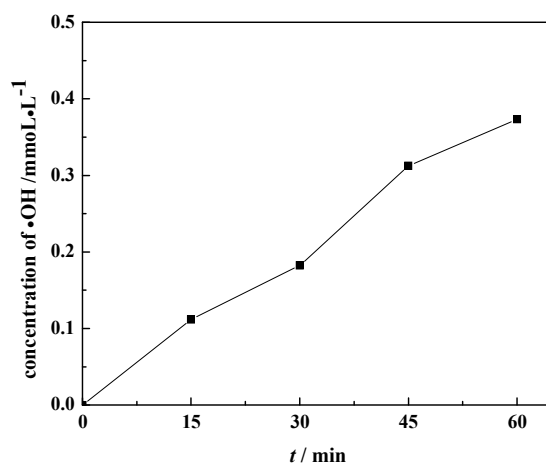


Fig. 8 The rate of production of •OH.

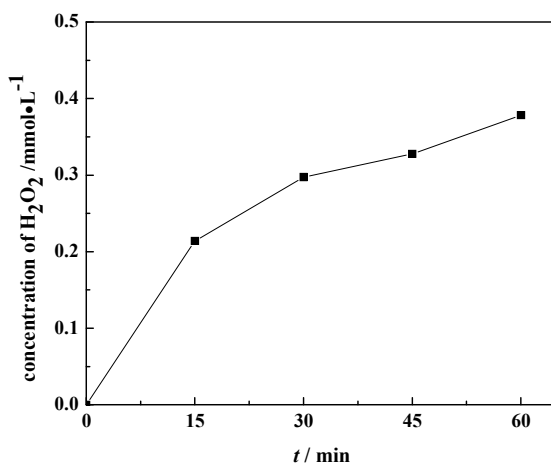


Fig. 9 The rate of production of H<sub>2</sub>O<sub>2</sub>

## Conclusions

Pulsed high-voltage electrical discharge with Fenton reaction was applied to the treatment of azo dye (Acid Orange II, AO7) simulation wastewater. NGL discharge was more effective than GL and L discharge. The iron mesh-plate discharge with

more discharge points was proved superior to the multi-needle-plate discharge on degrading the AO7. The high degradation rate, up to 85.8%, was achieved, when the initial pH was 2.28 and the concentration of  $\text{Fe}^{2+}$  was  $1.635 \text{ mmol}\cdot\text{L}^{-1}$ . It could be concluded that higher removal efficiency was obtained in pure oxygen than in air and in nitrogen. Changing the initial conductivity from  $200 \text{ }\mu\text{S}\cdot\text{cm}^{-1}$  to  $800 \text{ }\mu\text{S}\cdot\text{cm}^{-1}$  has not shown any significant change in degradation efficiency. However, the degradation efficiency dropped significantly when the initial conductivity became large enough. The oxidants induced in the discharge were also analyzed, both the concentrations of  $\cdot\text{OH}$  and  $\text{H}_2\text{O}_2$  increased as the process went on.

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