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Degradation of phenol wastewater by a new electromagnetic induction photo-catalytic reactor

X C Yuan^{1,2}, Q H Meng¹, J Y Sun¹, Y Yan¹, L Li¹, G C Li¹ and D Li¹

¹School of Chemistry and Chemical Engineering, Jiangsu Key Laboratory of Green Synthetic Chemistry for Functional Materials, Jiangsu Normal University, Xuzhou 221116, P.R. China

E-mail: yxingcheng@163.com

Abstract. A new inductive photo-catalytic reactor was obtained by the alternative magnetic field and optical coupling, which was driven by AC supply. In the cylinder reactor, UV-LED lights with the wavelength of 375-380nm were evenly distributed, and the phenol solution was used as simulated wastewater. The effects of initial phenol concentration, pH, TiO₂, H₂O₂, alternative magnetic frequency, current, and reaction time on the phenol degradation were investigated under an imposed alternative magnetic field. The optimized conditions and results were as follows: phenol concentrations of 15mg/L, pH of 7, H₂O₂ of 15μL, TiO₂ of 0.18g and alternative magnetic frequency of 12 KHz and current of 2A. With these conditions, the phenol degradation ratio reached 47.1% in 1 h reaction time. The new reactor is very promising for the effective treatment of refractory organic pollutants.

1. Introduction

Contamination of surface water due to refractory organic pollutants is posing a direct threat to human health and environment, which has become an important research topic about how to deal with these pollutants in the global. Many studies have shown that semiconductor photo-catalytic technology has many advantages to remove refractory organic pollutants, such as high removal efficiency, no secondary pollution, quick decomposition rate, and easy operation [1-3]. This technology is a kind of extremely promising purification technology and has received widespread attentions.

The core technology of semiconductor photo-catalytic oxidation is the light source, and developing light sources with long lifetime and uniform illumination has attracted much interest [4-6]. There are two types of light sources for photo-catalytic reactors: solar and artificial light; the latter source includes a high-pressure mercury lamp, fiber, and LED. Traditional photo-catalytic reactors usually use high-pressure mercury lamps as the light source, whose illumination intensity is high, but decays very fast, resulting in short lifetime. Moreover, the light from mercury lamps irradiates only from a side or surface, resulting in low light efficiency. Ollis and Marinangel proposed that fiber should be used as the transmission medium and photo-catalyst substrate [7]. The photo-catalytic process can be more environment-friendly, safer due to the strong and flexible fiber, lower energy radiation, and long distance lightning [8]. However, fiber reactors are expensive and cannot provide uniform illumination, i.e., the closer the light source, the stronger the illumination by the fiber. Recently, LED lights have become one of the most potential light sources. Compared to traditional mercury lamps, LED lights can convert electric power into optical energy. Moreover, when DC is used as the power source, LED



lights have simple modulation, long lifetime, high photon efficiency, no toxicity, high safety, and low volume; they have been widely used in the degradation of pollutants by photo-catalytic.

In this study, UV-LED lights with wavelength of 375-380nm were used to manufacture inductive photo-catalytic reactors. Based on electromagnetic induction, the micro-inductive LED light sources were evenly distributed in a cylinder reactor wrapped up with coils. Thus, an inductive photo-catalytic reactor was obtained by the alternative magnetic field and optical coupling, which was driven by AC supply. Phenol solution was used as the simulated wastewater. The effects of initial phenol concentration, pH, TiO_2 , H_2O_2 , alternative magnetic frequency, current, and reaction time on the phenol degradation were investigated under an imposed alternative magnetic field.

2. Materials and methods

2.1. Chemicals and reagents

AEROXIDE TiO_2 P₂₅ was purchased from Beijing Entrepreneur Science and Trading Co., Ltd. CH_3OOH were purchased from Sigma-Aldrich and HPLC grade. HCl, NaOH, H_2O_2 , $\text{C}_6\text{H}_5\text{OH}$ were analytical-grade and obtained from Shanghai Chemicals Factory, China. The laboratory water was obtained from Wahaha drinking pure water.

2.2. Experimental set-up

The new electromagnetic induction photo-catalytic reactor was shown in Figure 1.

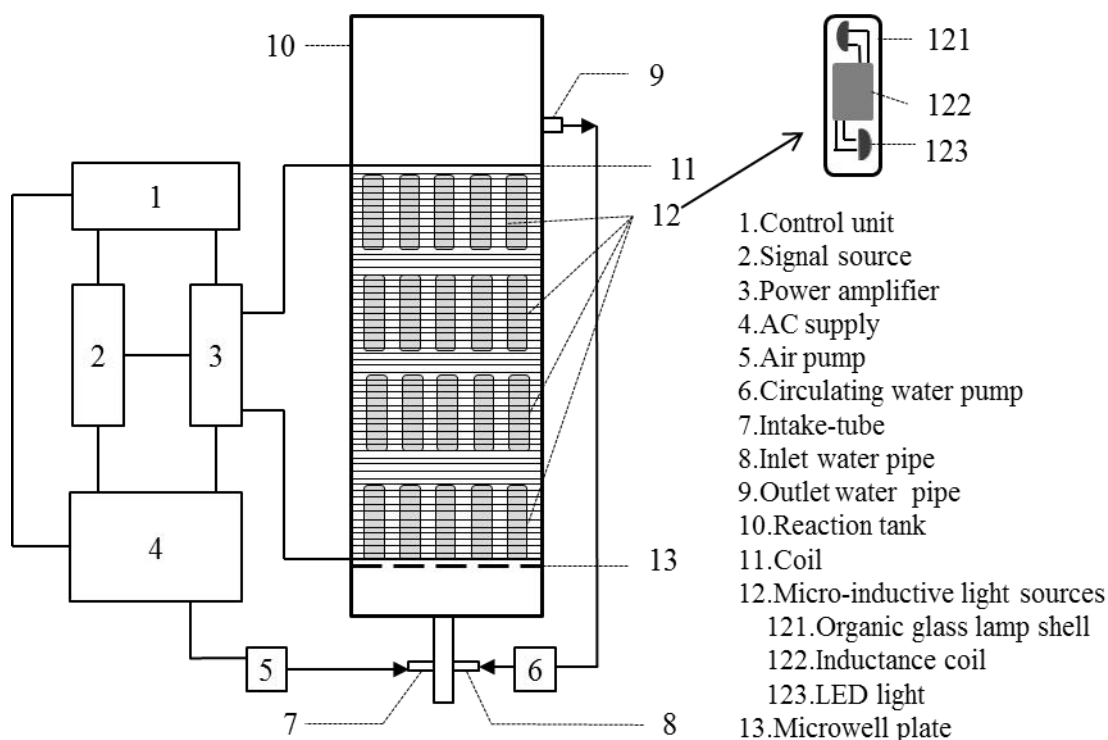


Figure 1. Schematic diagram of experimental set-up.

This photo-catalytic system was composed mainly of two parts: high-frequency AC (alternating current) supply and photo-catalytic reactor. The core was the inductive photo-catalytic reactor. The signal source of the high-frequency AC supply provided sinusoidal alternative signal; after amplification using the power amplifier, the signal worked on the coils that wrapped on the external wall of the reactor, thus generating an alternative magnetic field. Now, the inductive coils inducted energy from the alternative magnetic field on evenly filling micro-light sources in the reactor, which generated current to power the UV-LED lights and emitted UV light in a certain wavelength. The

organic wastewater could be catalytically degraded by the alternative magnetic and optical fields. The reaction liquid was removed through the top outlet using a circulation pump and added through the bottom inlet. The photo-catalytic reaction was repeated continuously. An air pump was used to introduce air from the bottom of the reactor to provide sufficient air and prevent the concentration of catalysts. Because of the even distribution of inductive light sources in the reactor, the efficiency of light sources was enhanced.

The structure of micro-inductive light sources was shown in Figure 1 (label 12). This was constituted by two UV-LED pins with parallel anti-phase connection, then connected to two output pins of inductive coils, and sealed by transparent synthetic glass. Cost-effective LED lights with the wavelength of 375-380 nm were selected for the device as the light sources. The micro-inductive light sources were densely placed in the reactors at multiple layers. When electromagnetic induction was applied in photo-catalytic reactors, it was not necessary to connect to power supply. The light sources could be evenly distributed in the reactors. Thus, the disadvantage of the breakdown of one LED light leading to the malfunction of all of them could be avoided.

2.3. Experimental procedures

The phenol solution was used as the simulated wastewater. The effects of the initial phenol concentration, initial pH, TiO₂, H₂O₂, alternative magnetic frequency, current, and reaction time on the phenol degradation were investigated under imposed alternative magnetic field. The experiment was designed as follows:

TiO₂ and H₂O₂ were added into 250 mL phenol solution. The temperature was controlled by circulating water at 30 °C. The air flow rate was controlled using a compressor. The light source intensity was controlled using a regulated power supply. A cycling pump was used to circulate the reaction liquid. The current intensity and frequency of alternative magnetic field were adjusted. A stirrer was used to mix the reaction liquid. After 20 minutes, a sample was collected from the reactor and centrifuged using the high-speed centrifuge at 18,000 rpm for 5 min. The phenol was analyzed in the supernatant to calculate the degradation ratio.

2.4. Analysis

Analysis of phenol was performed using a HPLC system (Agilent, USA, 1100 Series high-performance liquid chromatography). The column temperature was 30 °C and a sample volume of 20 µL was injected from the sampler. Chromatographic separation was achieved on a Kromasil 100-5C₁₈ (250 × 4.6mm, E56403, Alltech, USA), the mobile phase consisted of 55% water and 45% methanol, the flow rate was 1.000 mL/min. The determination wavelength was set at 270 nm.

The phenol degradation efficiency was calculated using the following equation:

$$\eta = \frac{A_0 - A}{A_0} \times 100\%$$

η — The degradation efficiency of phenol (%);

A_0 — The peak area of phenol before degradation;

A — The peak area of phenol after degradation.

3. Results and discussion

3.1. Effect of initial concentration and pH on the phenol degradation

The phenol solutions with initial concentrations of 10 mg/L, 15 mg/L, 20 mg/L, 25 mg/L respectively and the initial pH of 7 were used about 250ml. And other operating parameters were TiO₂ of 0.20 g, H₂O₂ of 50µL, air flow rate of 60 L/h, current of 1 A, alternative magnetic field of 8 KHz, and the reaction time of 1 h respectively. The results were shown in Figure 2. The initial concentration was shown an effect on the phenol degradation with a certain extent. When the phenol concentration was 15 mg/L, the degradation percentage reached as high as 25.4%. Although the concentration of organic

pollutants in industrial wastewater is usually unknown, a proper pollutant concentration can enhance the degradation.

The phenol solutions with initial pH of 4,5,6,7,8,9,10,11 respectively and initial concentration of 15mg/L were used about 250ml. And other operating parameters were TiO₂ of 0.20 g, H₂O₂ of 50μL, air flow rate of 60 L/h, current of 1 A, alternative magnetic field of 8 KHz, and the reaction time of 1 h respectively. The results were shown in Figure 3. The initial pH of the solution was shown significantly effect on the phenol degradation. In an acidic solution, the photo-catalytic degradation became more efficient at higher pH. The highest photo-catalytic degradation was achieved around neutral pH. However, the higher pH would cause lower photo-catalytic degradation in alkaline solutions. As the pH>9, OH⁻ and ArO⁻ would be adsorption on the TiO₂ surface adsorption, which resulted in competing with each other and reduced the amount of •OH. Therefore, when the pH value is relative taller, phenol degradation rate can be reduced.

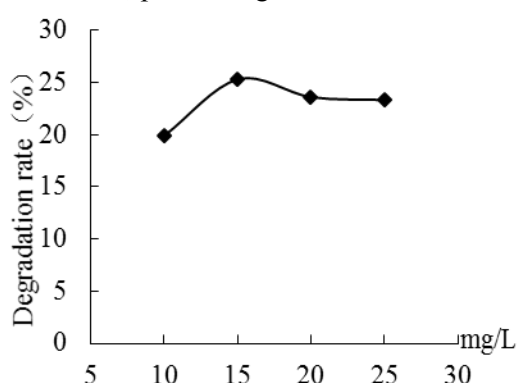


Figure 2. Effect of initial concentration on the phenol degradation.

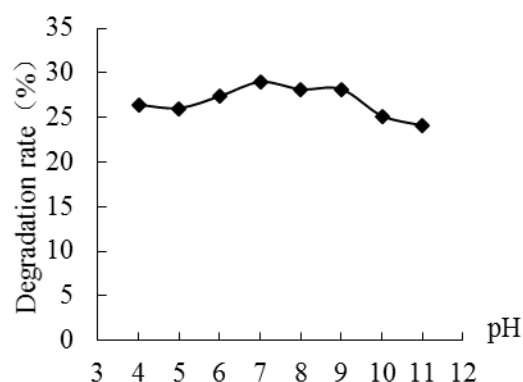


Figure 3. Effect of pH on the phenol degradation.

3.2. Effect of TiO₂ and H₂O₂ additives on the phenol degradation

The catalyst TiO₂ was added to the phenol solutions with 0.14g, 0.16g, 0.18g, 0.20g, 0.22g respectively. And other operating parameters were initial concentrations of 15mg/L, pH of 7, H₂O₂ of 50μL, air flow rate of 60 L/h, current of 1 A, alternative magnetic field of 8 KHz, and the reaction time of 1 h respectively. As shown in Figure 4, the photo-catalytic degradation ratio first increased with the amount of catalyst and then decreased after reaching its peak value. Under the UV irradiation, the higher quality of TiO₂ concentration in the solution produced more electron/hole and more •OH, which would speed up the reaction [9-10]. However, excessive TiO₂ concentration reduced the light energy utilization and made the degradation effect weaker due to the incident ultraviolet shielding and scattering effect. The optimum amount was determined to be 0.18 g in this study.

Similarly, the catalyst H₂O₂ was added to the phenol solutions with 5μL, 10μL, 15μL, 25μL, 50μL, 100μL, 150μL respectively. And other operating parameters were initial concentration of 15mg/L, pH of 7, TiO₂ of 0.18g, air flow rate of 60 L/h, current of 1 A, alternative magnetic field of 8 KHz, and the reaction time of 1 h respectively. The amount of H₂O₂ was shown to affect the photo-catalytic degradation of phenol by TiO₂ to a certain extent. The photo-catalytic degradation ratio first increased with the amount of H₂O₂ and then decreased after reaching the peak value at a dosage of 15μL, as shown in Figure 5. With the increase of H₂O₂ dosage, the amount of •OH increased in the solution, which promoted the organic matter degradation under the condition of light. Excessive H₂O₂, however, not only consumed •OH, but the decomposition of H₂O₂ was also invalid [11].

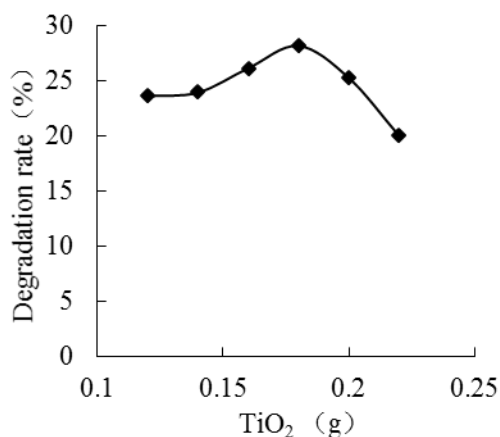


Figure 4. Effect of TiO₂ on the phenol degradation.

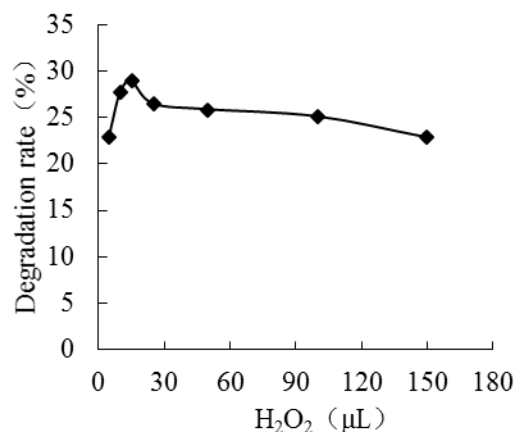


Figure 5. Effect of H₂O₂ on the phenol degradation.

3.3. Effect of magnetic field and coil current on the phenol degradation

The frequency of alternative magnetic field was changed from 1 to 20 KHz. And other operating parameters were initial concentration of 15mg/L, pH of 7, H₂O₂ of 15μL, TiO₂ of 0.18g, air flow rate of 60 L/h, current of 1A and the reaction time of 1 h respectively. As shown in Figure 6, the applied alternative magnetic field was shown to affect the photo-catalytic degradation of phenol. At low frequency, the effect of applied alternative magnetic field was not obvious. At the higher frequency, the magnetic field intensity became higher, resulting in the stronger UV light, a stronger coupling between the magnetic field and UV light, and the higher photo-catalytic degradation ratio of phenol.

In the same way, the high-frequency current of coil was changed from 0.4A to 2A by increasing 0.2A. And other operating parameters were initial concentration of 15mg/L, pH of 7, H₂O₂ of 15μL, TiO₂ of 0.18g, air flow rate of 60 L/h, alternative magnetic field of 12 KHz and the reaction time of 1 h respectively. As shown in Figure 7, the coil current was shown to affect the photo-catalytic degradation of phenol. With the stronger coil current, the intensity of the alternative magnetic field became stronger, and the phenol degradation became more efficient. When the frequency of magnetic field was 12 KHz and the coil current was 2.0 A, the phenol degradation ratio reached as high as 47.1%.

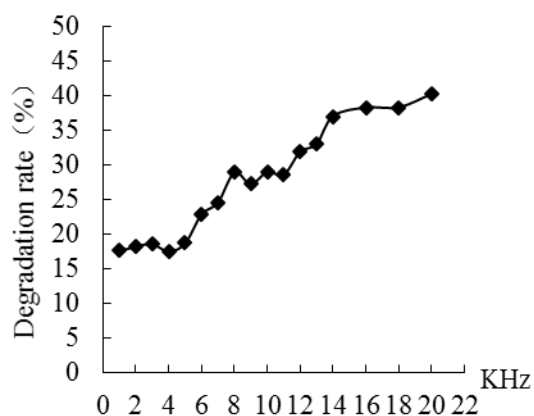


Figure 6. Effect of magnetic frequency on the phenol degradation.

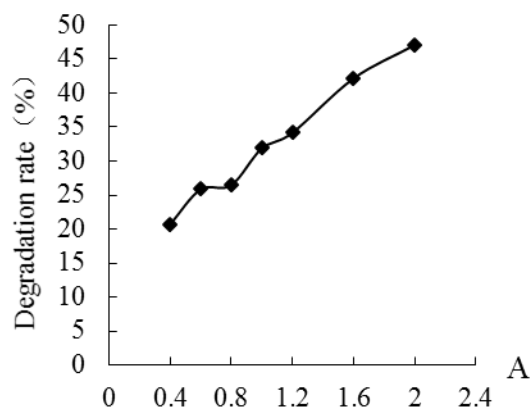


Figure 7. Effect of coil current on the phenol degradation.

3.4. Effect of reaction time on the phenol degradation

According to the above optimization of conditions, the operating parameters were initial concentration of 15mg/L, pH of 7, H_2O_2 of 15 μL , TiO_2 of 0.18g, air flow rate of 60 L/h, alternative magnetic field of 12 KHz respectively. And then the reaction time was changed with 0.5h, 1h, 2h, 3h, 5h, 7h, 13h and 23h. As shown in Figure 8, the reaction time was shown to affect the photo-catalytic degradation of phenol. With the longer reaction time, the degradation of phenol became much faster. Then, the photo-catalytic degradation decreased after a certain point. The phenol degradation reached as high as 97.9% after 23 h.

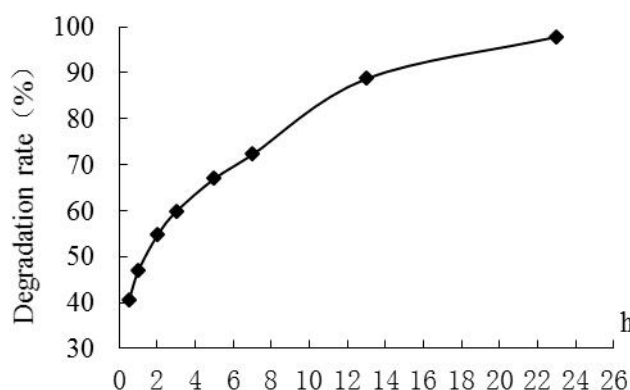


Figure 8. Effect of reaction time on the phenol degradation.

4. Conclusion

The new electromagnetic induction photo-catalytic reactor worked very well for the phenol degradation using the simulated wastewater. The optimized conditions and results were as follows: phenol concentration of 15mg/L, pH of 7, H_2O_2 of 15 μL , TiO_2 of 0.18g, air flow rate of 60 L/h and alternative magnetic field of 12 KHz and current of 2A. With these conditions, the phenol degradation ratio reached 47.1% in 1 h reaction time.

The new reactor generated electricity through electromagnetic induction without any external power. The photo-catalytic efficiency could be enhanced by coupling an alternative magnetic field and UV light and evenly distributing micro-light sources in the reactor. Thus, the lifetime of micro UV-LED light sources was extended, and the wavelength could be adjusted. So, the new reactor is very promising for the effective treatment of organic pollutants.

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