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Photolysis kinetics and influencing factors of bisphenol S in aqueous solutions

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Abstract

The photodegradation of bisphenol S (BPS) in aqueous solutions was studied under different conditions. Photolysis and kinetics were investigated, as were the photolysis mechanism and the influences of initial pH value, light source, and environmental substances in water. The results showed that the photolysis of BPS occurred under UV light, and the rate increased with light source intensity. The photolysis of 5.0–50.0 mg/L BPS in water followed first-order kinetics: the rate was $\gamma = 0.0161C_{\text{BPS}}$ under a 40-W UV-lamp, and the degradation half-life was 43.1 min. Due to its absorption of light, direct photolysis of BPS was a predominant pathway for BPS but was not obviously affected by reactive oxygen species. The results confirmed that the photolysis rates of BPS in alkaline water solution were faster than those in acidic and neutral water solution because of the ionization of BPS. The photodegradation rate of BPS increased in the presence of chloride and ferric ions, while the rate was inhibited by nitrate and phosphate in aqueous solution.

Key words: bisphenol S; photolysis; kinetics; influencing factors

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Introduction

The environmental effects of endocrine disruptors have been of increasing scientific and public concern over the past decade (Fenner-Crisp et al., 2000; Peré-Trepaut et al., 2004; Sichilongo and Torto, 2009). Endocrine disruptors are suspected of accumulating in living organisms and aquatic environments (Gong et al., 2009; Markman et al., 2007), causing irreversible damage to reproductive systems and, thus, threatening the sustainability of many animal species (Andrea, 2008).

Bisphenol S (BPS), 4,4'-sulphonyl diphenol, has been widely used as a monomer in the production of epoxy resins (Rwei et al., 2003), cyclic carbonates (Kim et al., 2001), and sulphonated poly(ether ketone ether sulphone) (S-PEKES) (Changkhamchom et al., 2010). It is also an important chemical additive in pesticides, dyestuffs, color-fast agents, leather tanning agents, dye dispersants, and fiber improvers. Thus, production of and demand for BPS in China are increasing continuously (Liu, 2005) and due to its widespread application, there is a high risk of human exposure to BPS.

Phenolic compounds are highly toxic environmental pollutants that seriously threaten human health (Fan et al., 2007). Different phenolic compounds possess different environmental behaviors and have differing ecological effects and toxicities. Animal experiments have demonstrated that BPS, which can promote lymphocyte proliferation, is also

an environmental endocrine disruptor (Li et al., 2003). To date, however, very little is known about the behavior of BPS in aquatic environments.

The goal of the present study was to examine the photodegradation process and reaction kinetics of BPS in aqueous solutions. Factors affecting direct photolysis of BPS were studied in detail, including initial pH value and irradiation light source. The photolysis of BPS in the presence of chloride, ferric, nitrate, and phosphate ions were also investigated. Because chloride ions, ferric ions, nitrate, and phosphate are common components of natural water, this study may help in understanding the photochemical behavior of BPS in aquatic environments.

1 Experimental

1.1 Materials and chemicals

Bisphenol S was obtained from the Beijing Chemical Reagent Co., Ltd. (China). $\text{Fe}_2(\text{SO}_4)_3$, $\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$, sodium chloride and sodium nitrate were purchased from the Shanghai Chemical Reagent Co., Ltd. (China) and were used to provide ferric ions, phosphate ions, chloride ions and nitrate ions, respectively. Isopropanol and sodium azide were purchased from the Tianyu Chemical Reagent Co., Ltd. (China). High-performance liquid chromatography (HPLC)-grade acetone and ethyl acetate were obtained from Tedia Ltd. (Fairfield, USA). All experimental solutions were prepared by dissolving the reagents directly in

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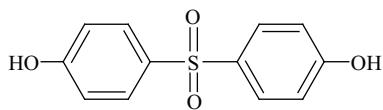
twice-distilled water. All stock solutions were refrigerated after preparation and used within one month. The different pH values of the aqueous solutions, pH 3.0, 5.0, 7.0, 9.0, and 11.0, were determined using a pH meter and were obtained by adjusting the solutions as needed with 0.1 mol/L H₂SO₄ or 0.1 mol/L NaOH.

1.2 Photochemical experiments

Photochemical experiments were conducted in a photochemical reaction chamber using two kinds of lamps as the irradiation source: a 40-W metal halide lamp (MHL, $\lambda \geq 313$ nm) from Wuhan Yaming Lamp Co., Ltd. (China) to simulate UV-Vis light (Zhou et al., 2010; Zhang et al., 2006), and a 20-W and a 40-W UV lamp (UV, 254 nm) from Yuelu Irradiation Lamp Co., Ltd. (Changsha, China) to simulate UV light. The lamp was hung over quartz tubes (outer diameter 1.8 cm, length 13.5 cm) at a fixed distance of 8.0 cm. The temperature of the irradiated solution was maintained at 25±1°C. Each reaction solution was collected at selected time intervals and then analyzed by HPLC.

1.3 Analytical methods

The molecular formula of BPS is C₁₂H₁₀O₄S, and its structure is shown as:



The UV absorbance spectrum of the BPS solution was recorded with a UV-1102 spectrophotometer (Techcomp, China), and the characteristic absorbance peak was 258 nm with a molar absorption coefficient $\epsilon_{\text{max}} = 2.07 \times 10^4$ L/(mol·cm).

The concentration of BPS in the reaction process was measured by HPLC (Agilent 100, USA) using a device fitted with a Discovery C18 reversed-phase column (150×4.5 mm, 5 μm) at 30°C. The mobile phase composition was ethyl acetate-acetone (60:40, V/V) at a flow rate of 1.0 mL/min. Samples were analyzed with a UV photodiode array detector at a wavelength of 258 nm. Measurements

were made in triplicate for each experiment with less than 5% error.

2 Results and discussion

2.1 BPS photolysis process

The UV spectra of the BPS photolysis process in water are presented in Fig. 1. The initial spectrum (before irradiation) had two maxima at 234 and 258 nm, and two minima at 219 and 240 nm. The absorbance curves showed a flattening tendency with irradiation time. It is evident from Fig. 1 that as photolysis time increased, both absorption maxima disappeared, indicating that the concentration of BPS was lost as photodegradation proceeded.

2.2 Photolysis kinetics

According to the theory of chemical kinetics (Fu et al., 2006), the photolysis rate (γ , mg/(L·min)) can be defined as:

$$\gamma = -\frac{dC_{\text{BPS}}}{dt} \quad (1)$$

where, t (min) is the photolysis time and C_{BPS} (mg/L) is the concentration of BPS in the photolysis process. The direct photolysis for different initial BPS concentrations was conducted in pure water. Figure 2 shows that the photolysis rate increased with increasing initial BPS concentration; the fitted curve of initial rate versus initial BPS concentration followed first-order law in the solutions. The photolysis kinetic equation is:

$$\gamma = k_{\text{BPS}} C_{\text{BPS}} = 0.0161 C_{\text{BPS}} \quad R^2 = 0.9820 \quad (2)$$

where, k_{BPS} (min⁻¹) is the photolysis rate constant. The degradation half-life of BPS was 43.1 min.

2.3 Effect of pH and light source

Figure 3 shows that the BPS degradation rate did not vary significantly with pH for pH values from 3.0 to 7.0. However, the degradation rate increased significantly for pH values of 9.0 and greater. After 40 min of radiation, the

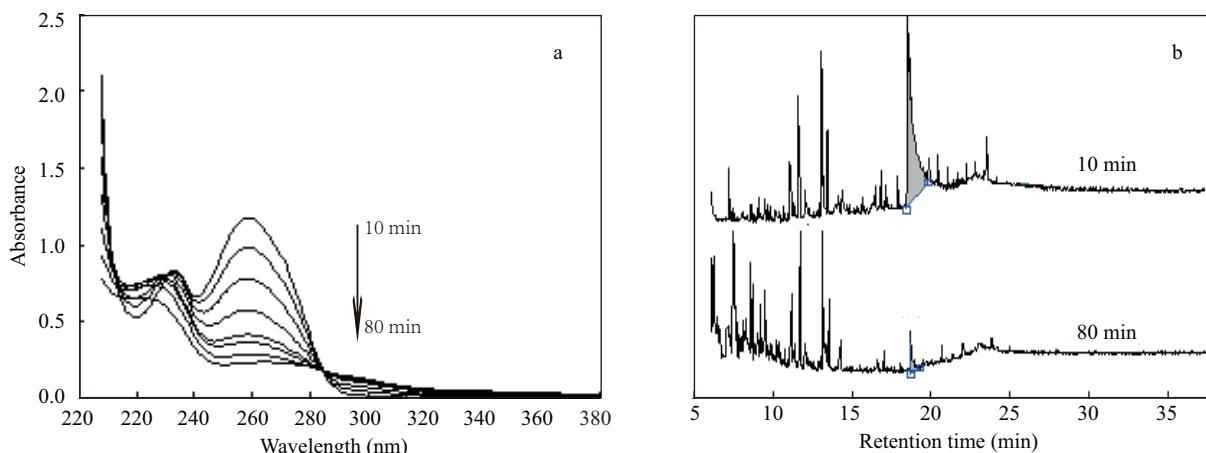


Fig. 1 (a) Absorbance of BPS in twice-distilled water versus wavelength change during photolysis at 10-min intervals under a 40-W UV lamp, (b) HPLC spectra of reaction solution in photolysis 10 min and photolysis 80 min.

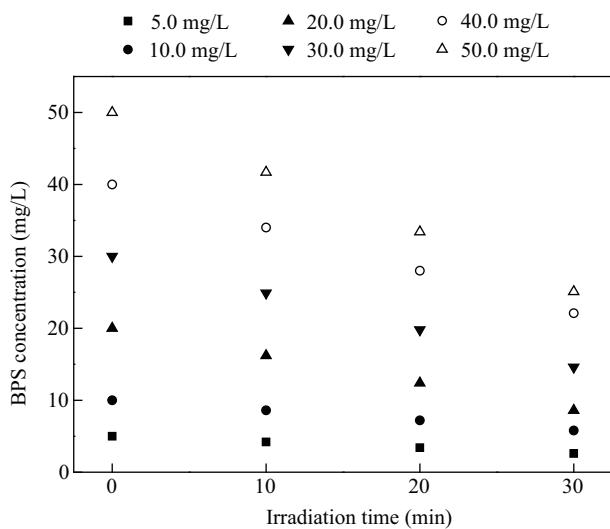


Fig. 2 Concentrations of BPS during the direct photolysis reaction under a 40-W UV lamp, for different initial concentrations in the range of 5.0–50.0 mg/L.

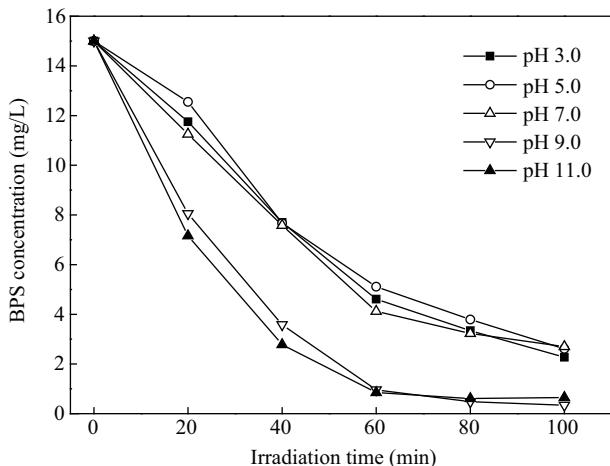


Fig. 3 BPS concentration versus time in water of varying pH with initial BPS concentration of 15.0 mg/L.

degradation efficiency of BPS was 48.7% at pH 3.0, but 81.5% at pH 11.0.

As shown in Fig. 4, there was almost no difference between the UV absorption spectra at acidic and neutral pH. The maximum peak absorbance was at 258 nm. However, in alkaline solutions, the absorption spectra of BPS increased sharply in wavelength from 285 to 305 nm. This change was attributed to the ionization of BPS in aqueous solutions of pH > 9.0 (Juan, 1999). The BPS phenolate anion, originating from the deprotonation of the phenolic hydroxyl group, made the absorption spectra shift to longer wavelengths, causing a significant increase in absorbance at $\lambda = 295$ nm ($\epsilon_{\text{max}} = 3.41 \times 10^4$ L/(mol·cm)). The red shift produced in alkaline solution enhanced the absorbency of BPS. Thus, alkaline pH appears to enhance the degradation of BPS.

To study the influence of different light sources on the photodegradation efficiency of BPS, aqueous solutions containing 15.0 mg/L BPS were irradiated separately under a 40-W MHL lamp (10.5 $\mu\text{W}/\text{cm}^2$), a 20-W UV lamp (3.6 mW/cm²), and a 40-W UV lamp (7.1 mW/cm²). Figure 5 shows that after irradiation for 120 min, the

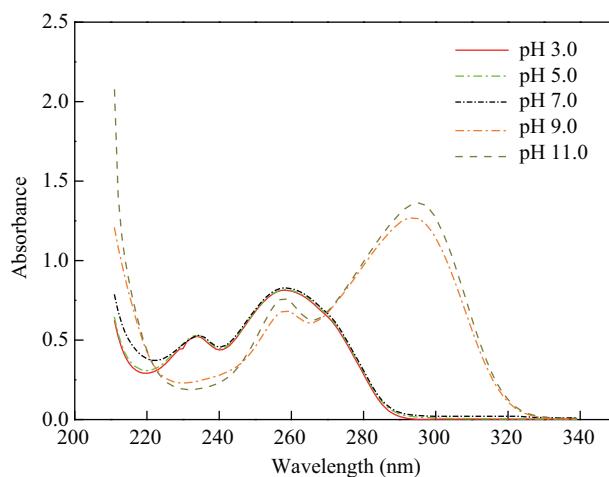


Fig. 4 UV absorption spectra of 10.0 mg/L BPS in aqueous solution at different pH values.

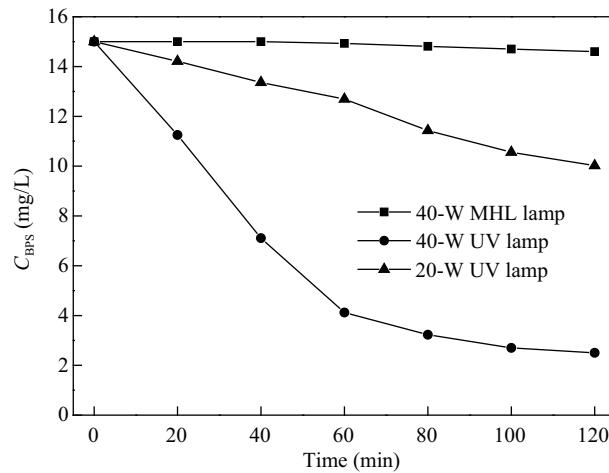


Fig. 5 BPS concentration versus time under irradiation from different light sources.

degradation efficiencies of BPS from lowest to highest were the MHL lamp (2.7%), the 20-W UV lamp (33.2%), and the 40-W UV lamp (83.3%). This is because BPS in water has almost no absorption of light at wavelengths above 320 nm (Fig. 4), and the MHL lamp used in this study only produced light at wavelengths > 313 nm. These results also indicate that the photodegradation of BPS is more efficient under higher powered lamps.

2.4 Effect of chloride, ferric ions, nitrate and phosphate

Sodium chloride (NaCl) is present in almost every soil and aquatic environment (Kochany et al., 1990); thus, natural water contaminated with BPS likely also contains chloride. To examine the effect of the ionic strength of chloride on the photolysis of BPS, different concentrations of NaCl were added to aqueous solutions of 15.0 mg/L of BPS before irradiation. Figure 6 shows that a higher degradation rate of BPS was achieved with higher concentrations of NaCl. The initial degradation rates were 0.041, 0.128 and 0.280 mg/(L·min) of BPS in the presence of 0, 0.01, and 0.05 mol/L of NaCl, respectively. The increasing ion strength evidently increased the degradation of BPS during photolysis, as observed in previous research on organic compounds such as chloromycetin (Zhou et al., 2010). This

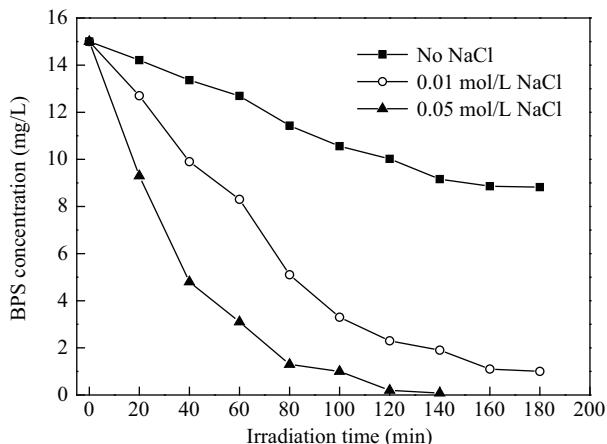


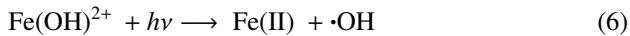
Fig. 6 BPS concentration change with time in the presence of different concentrations of NaCl under a 20-W UV lamp.

might be the result of the photosensitization of halide ions. Combined with data documented in the literature (Li et al., 1991), photosensitization caused by halide ions might be as follows:



Reaction (3) is the photochemical reaction, and Cl· radicals are produced; Reaction (4) of Cl· radicals and halide ions results in the production of Cl₂· radicals, which are the precursors of phenol oxidation processes. The P stands for the products of the oxidized BPS.

Iron is one of the most abundant transition metals in the environment. Thus, the effect of ferric ion concentrations on the photolysis of BPS was examined. Figure 7 shows that only 4.3% of the BPS was degraded by MHL light irradiation in the absence of ferric ions. The addition of ferric ions to the BPS solution enhanced BPS degradation under MHL light, especially after 80 min of irradiation. These findings are similar to those of studies on Fe(III)-pyruvate complexes (Wang et al., 2006) and Fe(III)-citrate complexes (Zhang et al., 2006). According to previous studies (Zhou, 2010), photolysis of ferric ions under UV-Vis light produces ·OH radicals as follows:



The ·OH radical has a high oxidizing potential for the degradation of organic pollutants in water. The ·OH produced from the photolysis of ferric ions might play an important role in oxidation and promote the photodegradation of BPS in aqueous solutions.

However, when pH was adjusted to 5.0 and 7.0, the BPS degradation efficiency reduced from 56.5% to 6.3%, respectively and to 5.5% after 180 min irradiation. The addition of ferric ions no longer increased BPS photodegradation efficiency because of the lower photoactivity of ferric ions at pH values greater than 3.0. Moreover, ferric ions are partially or completely precipitated when pH is 5.0 or 7.0. Results indicate that pH value has a great effect on the promotion of ferric ions.

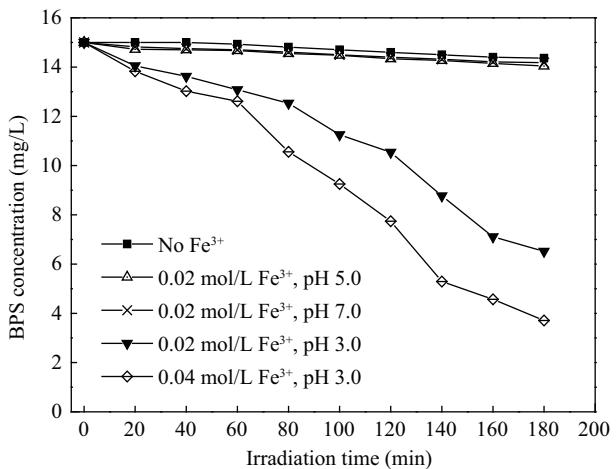


Fig. 7 BPS concentration change with time in the presence of different concentrations of ferric ions under a 40-W MHL lamp.

Excess nitrogen and phosphorus, usually in the form of nitrate and phosphate, are major causes of eutrophication of natural waters (Wu et al., 2008; Yang et al., 2008). The effects of nitrate and phosphate on the direct photolysis of BPS were tested at pH 7.0 by the addition of 0.01 mol/L and 0.05 mol/L nitrate and phosphate to the BPS solution. Figure 8 shows that adding 0.01 or 0.05 mol/L nitrate or phosphate to the solution inhibited BPS photolysis. After 180 min irradiation, the degradation efficiency for 15.0 mg/L BPS was 86.5% in water, 74.5% in the presence of 0.05 mol/L nitrate, and 66.5% in the presence of 0.05 mol/L phosphate. In addition, pale yellow photoproducts were produced in the nitrate medium, while pink photoproducts were produced in the phosphate medium. These colored products might affect the degradation efficiency of BPS by competing with BPS for light absorption, retarding the direct photolysis of BPS.

Moreover, the photolysis of BPS was different in the presence of nitrate or phosphate. Because of the photosensitization of nitrate (Sharpless et al., 2003; Nélieu et al., 2004), nitro radicals might be produced in the photochemical reaction, which could attack the benzene ring to form nitro compounds (Zhan et al., 2005). In the presence of excess nitrate, the generated nitro radicals might react with BPS at the beginning of the irradiation. This would result in the photodegradation rate of BPS in solutions containing nitrate being lower than that in water, as shown in Fig. 8a. Phosphate, however, is not an environmental sensitizer and would not influence BPS photolysis directly. Figure 8b shows that at the beginning of irradiation, the photodegradation rate of BPS in the presence of phosphate was the same as that in water. However, after irradiation for 40 min, the degradation efficiency of BPS in the phosphate medium slowed down. The reason behind this phenomenon might be that the intermediates generated in the photolysis of BPS increased with irradiation time and reacted with phosphate ions to form new compounds, which retarded further direct photodegradation of BPS. These results indicate that in eutrophic waters more attention should be paid to photoproducts, which were found to be more persistent than the BPS.

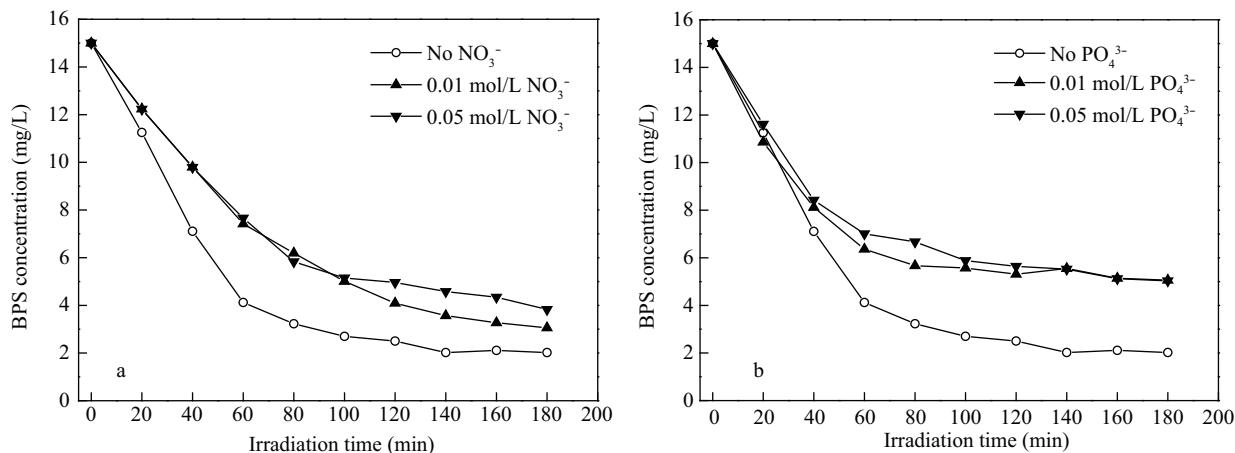


Fig. 8 BPS concentration change with time in the presence of different concentrations of nitrate (a) and phosphate (b) under a 40-W UV lamp.

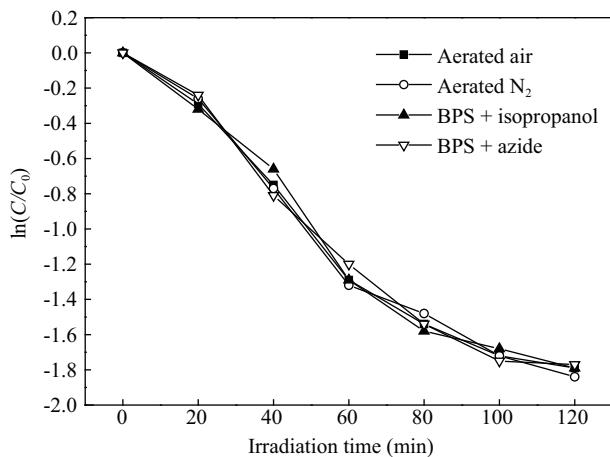


Fig. 9 Comparison of BPS photolysis under different conditions.

2.5 Photolysis mechanism of BPS

To study the degradation mechanism of BPS under UV light, the photolysis of BPS under different aerated conditions was examined, as shown in Fig. 9. Dissolved oxygen in water might participate in BPS degradation, except the direct photolysis of BPS as a consequence of UV-light absorption, by producing reactive oxygen species (ROS) such as $^1\text{O}_2$ (Chen et al., 2008) and $\text{O}_2^{\cdot-}$ (Draper et al., 1983), which may indirectly affect the degradation of BPS. However, as shown in Fig. 9, BPS degradation did not differ markedly between the aerated N_2 condition and the aerated air condition; thus, dissolved oxygen appeared to contribute very little to the photolysis process.

To determine whether $\cdot\text{OH}$ and $^1\text{O}_2$ were involved in the ROS-induced photodegradation of BPS, excess amounts of isopropanol and sodium azide were added in the aerated air condition as effective scavengers for $\cdot\text{OH}$ and $^1\text{O}_2$ (Yang et al., 2005), respectively. Figure 9 shows that BPS degradation rates were not suppressed by the presence of isopropanol or sodium azide and were similar to the BPS direct photolysis rate in pure water, suggesting that BPS degradation was primarily due to its absorption of light and ROS did not affect photolysis obviously.

3 Conclusions

The photodegradation of BPS in aqueous solutions can occur under UV light, and depends primarily on the direct photolysis of BPS as a consequence of its absorption of light. When initial concentrations of BPS ranged from 5.0 to 50.0 mg/L, the photolysis kinetics followed first-order law. Environmental factors, including pH, light source, and the presence of chloride, ferric, nitrate, and phosphate ions, influenced the photodegradation of BPS. High pH values increased the photolysis of BPS and the degradation efficiency of BPS was enhanced in the presence of chloride and ferric ions, but its photolysis was inhibited by the presence of nitrate and phosphate ions.

Acknowledgments

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