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Synergetic degradation of Acid Orange 7 by fly ash under ultrasonic irradiation

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ABSTRACT

The present work aimed to investigate the feasibility of fly ash, a by-product of coal combustion, for enhanced degradation of an azo dye Acid Orange 7 (AO7) under ultrasonic irradiation. X-ray diffraction and energy dispersive X-ray analysis indicated that ultrasonic irradiation did not change the crystal structure of fly ash. The decolorization efficiency of AO7 by the combined process could reach 76.7%, while ultrasonic process alone only removed 3.8% of AO7 within 60 min. A synergetic effect between fly ash and ultrasonic irradiation was firstly observed. The decolorization of AO7 fitted the first-order rate kinetics, and the K_1 was $0.0246\,\text{min}^{-1}$ for the combined process. Radical quenching experiment by iso-propanol (ISP) indicated that 24.8% of AO7 decolorization was contributed to hydroxyl radicals, indicating that the contribution of hydroxyl radicals was not as significant as expected. Theoretic analysis revealed that the sizes of fly ash cover the estimated resonance size of cavitation bubbles generated in this experiment. As such, fly ash particles could act as nucleus to generate more cavitation bubbles and enhance the ultrasonic degradation efficiency. Fly ash combined with ultrasonic irradiation has the great potential for practical treatment of wastewater containing organic pollutants.

Keywords: Water treatment; Synergetic effect; Ultrasonic irradiation; Acid Orange 7; Hydroxyl radicals

1. Introduction

Along with several decades of development, advanced oxidation processes (AOPs) are considered to be one of the most powerful technologies for the treatment of wastewaters containing toxic organic pollutants. AOPs are all characterized by the production of OH radical (2.72 V/NHE), which is the most reactive radical in aqueous solution [1]. Hydroxyl radicals are non-selective and they are capable of destroying organic pollutants into biodegradable chemicals or even completely mineralize them into water, carbon dioxide, and inorganics. Meanwhile, AOPs usually operate at or near ambient temperature and pressure, which is favorable for decontamination and purification.

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Ultrasonic irradiation, one of the typical AOPs, has received increasing attention in the past decades due to its capability of eliminating environmental contaminants efficiently [2–8]. The degradation of contaminants by ultrasound is mainly attributed to acoustic cavitation, which undergoes the formation, growth, and implosive collapse of bubbles, along with the intense localized temperature and pressures, electrical charges as well as plasma effects. High-frequency ultrasound was proved to be capable of breaking down non-polar pollutants such as hydrocarbons and aromatic contaminants [9–10]. Totally, the ultrasonic degradation process is based on pyrolytic degradation and reactive oxygen species (ROSs) oxidation resulted from the sonolysis of aqueous solutions [5]. These ROSs include OH radicals, H_2O_2, and HO_2^-, among which OH radicals are considered to be more responsible for ultrasonic degradation.

However, the efficiency of ultrasonic irradiation decreases as function of reaction time as oxidation intermediates become more polar. In addition, some undesirable and toxic intermediates may be accumulated in the process [5,11–13]. As a result, some combinative processes such as sono catalytic and sono photo catalytic process were developed recently to enhance the degradation efficiency [2,4,5,8,14]. In sono catalytic process, catalysts are used to lower reaction activation energy so that the efficiency can be improved significantly. Although various materials have been developed as catalysts for sono catalytic process [2,15,16], a low-cost catalyst is highly needed, which is always desirable in practical application.

Large quantity of fly ash is produced annually as a by-product of coal combustion in power stations. However, only a small portion of the industrial waste is utilized while most of them have to be discharged into landfills. Using fly ash for environmental decontamination is a good strategy to advance its utilization. The chemical composition of fly ash is typical of the most common glassy ternary system ($CaO–Al_2O_3–SiO_2$) with significant amounts of transition metal oxides. Wang and Zhu investigated sonochemical treatment of fly ash and then applied it for enhanced dye adsorption [17]. Fenton-like reactions combining ultrasound were studied by Song and Li, and they found that fly ash was a good Fenton-like catalyst which could achieve higher degradation efficiency compared with kaolinite, diatomaceous, and activated carbon [18]. Nakui and co-workers reported that coal ash (53–106 μm) increased the degradation of phenol under ultrasonic irradiation while the adsorption of phenol on the coal ash could be neglected [7]. Besides, coal ash was considered to have no catalytic property for the formation of OH radicals, and the enhanced degradation was regarded as a consequence of the surface roughness of the particles. In this study, fly ash from a local thermal power plant was used to assist the degradation of an azo dye Acid Orange 7 (AO7) under ultrasonic irradiation. A series of parameters such as fly ash dosage, solution pH, and glassware shape were investigated in terms of dye removal. Considering reaction mechanism, the contribution percentage of OH radicals and the resonance size of cavitation bubbles were discussed.

2. Materials and methods

2.1. Materials and apparatus

AO7 was purchased from Beijing Chemical Reagents Company. It was selected as a model compound and used without further purification. Fly ash was obtained from Xinli thermal power plant located in Zhengzhou City (Henan province, China). Sonication was performed in a 40 kHz ultrasonic cleaning bath (AS3120A, Tianjin Aotesai Instrument Co., China). Deionized (DI) water was used to prepare solutions.

2.2. Characterization

The morphologies of raw fly ash and used fly ash were recorded on a Philips Quanta-2000 scanning microscope coupled with an energy-dispersive X-ray (EDX) spectrometer. Their X-ray diffraction (XRD) patterns were recorded on a Rigaku D/MAX-3B X-diffractometer.

2.3. Degradation of AO7 by fly ash under ultrasonic irradiation

Degradation of AO7 was carried out in a conical flask. The volume of the conical flask was 500 mL, which contained 400 mL of AO7 solution ($10\,mg\,L^{-1}$) at neutral pH. A water-circulating unit was used to control the water temperature in ultrasonic bath. The dosage of fly ash was 3 g in 400 mL of AO7 solution unless otherwise stated. Samples were collected and filtered through a 0.45-μm membrane before analyzing. The concentration of AO7 was determined by measuring the absorbance at a fixed wavelength (484 nm), which is corresponded to the maximum absorption wavelength, by an UVmini-1240 spectrophotometer (Shimadzu). In the alcohol quenching test, the dosage of ISP was 0.05 mol L^{-1}. The decrease of apparent rate constant (pseudo first-order) K_1 resulted from ISP quenching can be used to indicate the contribution percentage achieved by hydroxyl radicals, which is calculated as the following equation [19,20]:

$$\text{Contribution percentage} = (1 - K_1(\text{quenching})/K_1) \times 100\% \quad (1)$$

3. Results and discussion

3.1. Characterization of fly ash

Fig. 1 shows the SEM images of raw fly ash and used fly ash for the degradation of AO7. It can be seen that fly ash particles have different shape and their size varies from 1 to 50 μm. Some of them aggregate together forming large particles. No significant change was observed in the morphologies of these fly ash particles after 2-h sonocatalytic reaction for degradation of AO7. The EDX analysis in Fig. 2 shows that the components of raw fly ash and used fly ash are almost same. The major constituents are amorphous carbon, quartz (SiO_2), mullite ($3Al_2O_3{\cdot}2SiO_2$), hematite (Fe_2O_3), magnetite (Fe_3O_4), lime (CaO), and gypsum ($CaSO_4{\cdot}2H_2O$). The atomic ratios of O, C, Si, Al, Ca, and Fe achieved 41.8, 28.0, 14.6, 11.0, 1.1, and 0.9%, respectively. A very weak N peak (3.1%, atomic ratio) was found for the used fly ash. It might be attributed to the residual AO7 molecule or its degradation products as no N element was detected on the raw fly ash. In addition, the intensity of Ca peak slightly decreased after the ultrasonic reaction, which indicates that lime or gypsum might be dissolved into water during the reaction.

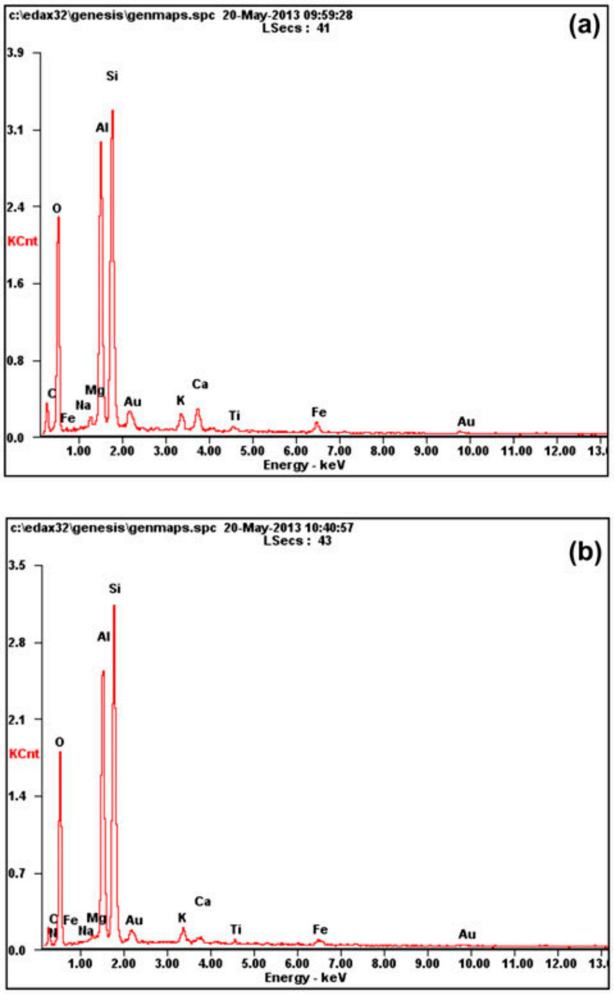

Fig. 2. EDX graphs of raw fly ash (a) and used fly ash (b).

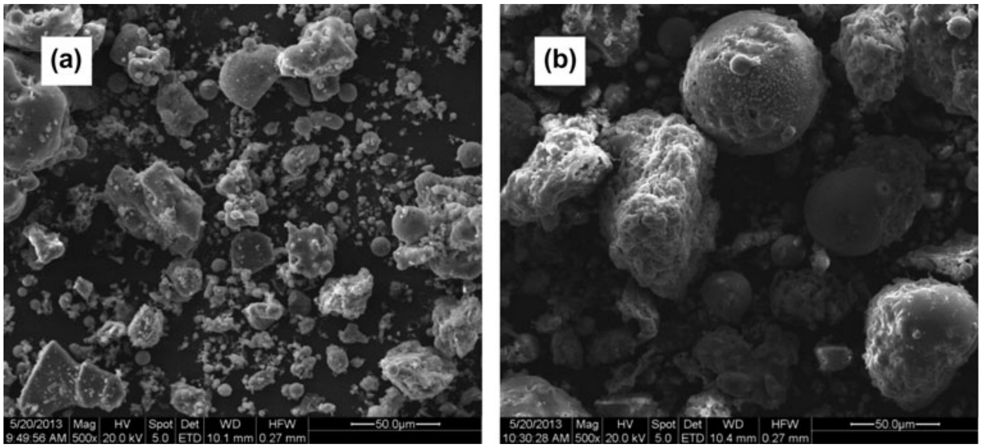

Fig. 1. SEM images of raw fly ash (a) and used fly ash for synergetic degradation of AO7 (b).

The XRD patterns of the raw fly ash and used fly ash samples are presented in Fig. 3. It can be seen that there is no significant profile change for the two samples, suggesting that no phase transform of fly ash happened during the ultrasonic treatment. Both samples consist of mullite and quartz, which are consistent with previous study [21]. Mullite does not naturally exist in coal. It is assumed to form during combustion by the thermal decomposition of natural alumino silicates such as kaolinite [22].

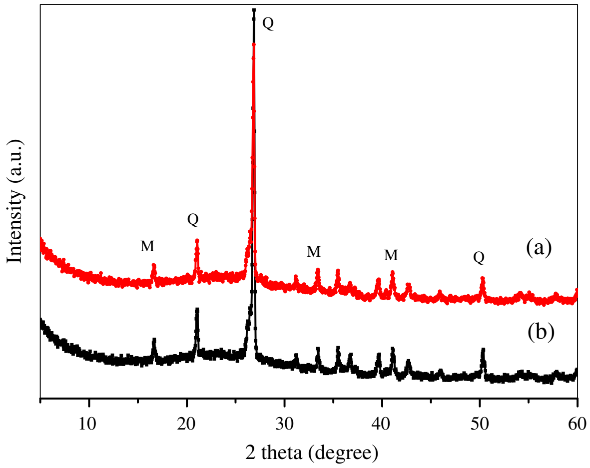

Fig. 3. XRD patterns of raw fly ash (a) and used fly ash (b). M = mullite, Q = quartz.

3.2. Destruction of AO7 and synergetic effect during the combinative degradation process

The removal of AO7 by fly ash under ultrasonic irradiation was investigated. Fig. 4 shows the UV–visible spectral changes of AO7 solution as a function of ultrasonic irradiation time. The major absorbance band at 484 nm and shoulder band 430 nm are ascribed to the hydrazone and azo form of AO7. In ultraviolet region, the bands located at 310 and 230 nm are due to the naphthalene and benzene rings of the dye, respectively [23]. As shown in Fig. 4(a) and (b), the four absorbance bands of the dye almost disappeared after 120-min degradation. Meanwhile, the bands at 484 and 430 nm disappeared more quickly and significantly compared with other two bands. The absorbance at 255 nm by 0, 60, and 120 min achieved 0.33, 0.215, and 0.253 respectively. This indicates that the chromophores of the dye are easily destructed while the naphthalene and benzene rings are difficult to degrade. It is worth to note that the absorbance at 255 nm firstly decreased and then increased due to the oxidation of the aromatic rings, which is consistent with our previous study [24]. It was also observed by other researchers that ultrasonic irradiation is capable of cleaving the aromatic rings [25].

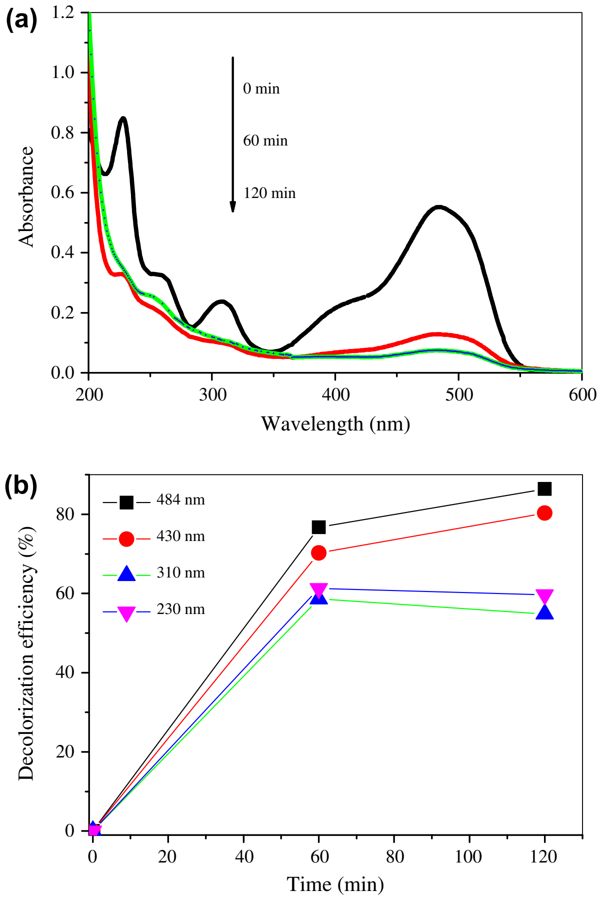

Fig. 4. UV–Visible spectral changes of AO7 as a function of ultrasonic irradiation time (a) and the decolorization efficiencies at 484, 430, 310 and 230 nm at 60 min and 120 min (b).

A dye decolorization efficiency of 41.5% was achieved within 60 min by fly ash alone as a consequence of dye adsorption, while the decolorization efficiency only reached 3.8% by ultrasonic irradiation alone. However, combining ultrasonic irradiation and fly ash achieves a much higher decolorization efficiency of 76.7%. AO7 degradation kinetics by fly ash alone and ultrasonic irradiation in the presence and absence of fly ash was investigated and the results are also illustrated in Fig. 5. The pseudo first-order kinetics of the three processes was simulated as:

$$\ln(C_0/C) = K_1 t \qquad (2)$$

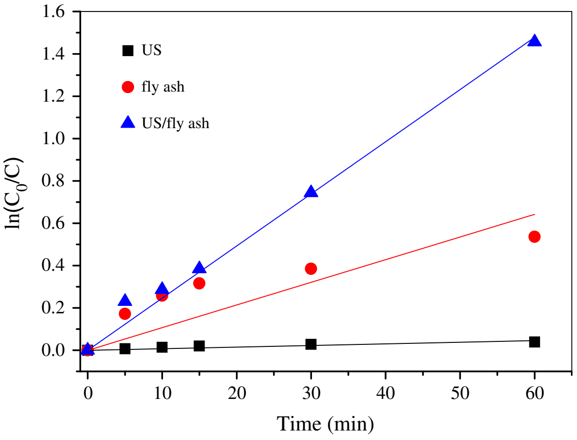

Fig. 5. Pseudo-first-order degradation of AO7 by ultrasonic irradiation (US) alone, fly ash alone and US combinative with fly ash (US/fly ash).

where C is the dye concentration at time t, C_0 is the initial dye concentration, and K_1 is the apparent first-order rate constant. The apparent first-order kinetics well fit the three processes and the K_1 for fly ash adsorption, ultrasonic irradiation, and the combined process are $0.0107\,\text{min}^{-1}$, $7.55 \times 10^{-4}\,\text{min}^{-1}$, and $0.0246\,\text{min}^{-1}$ ($R^2 = 0.92$, 0.86, 0.99). The K_1 for the combined process is 2.15 times that of the sum of K_1 values of the other two processes, indicating a significant synergetic effect.

3.3. Optimization of operational parameters

The effect of fly ash dosage was investigated from 0.5 to 3.0 g. The results are illustrated in Fig. 6. The dye decolorization efficiency increased with the increasing of fly ash dosage, which indicates that it is a favorable measure to enhance the degradation efficiency by increasing the catalyst dosage. The increase in decolorization efficiency was attributed to the increasing surface area and active sites of fly ash. Meanwhile, compared with the dosage of 2.5 g, the decolorization efficiency at the dosage of 3.0 g did not improve significantly. The similar degradation performance was also observed during sonocatalytic degradation by other researchers [2,7,15]. This might be attributed to the acoustic energy attenuation of ultrasonic wave and the decrease of nucleation site with increasing amount of fly ash.

The effect of solution pH on the decolorization process in the presence of fly ash and ultrasonic irradiation was illustrated in Fig. 7. It shows that the solution pH has a significant impact on the dye decolorization. The highest decolorization efficiency was observed at pH 7.0, which is determined by the degradation capability of the reactive system as well as the adsorption of the dye onto fly ash. As the aforementioned EDX result, the main constituents of fly ash are silica and alumina. The zero point charges (ZPC) of silica and alumina are 2.3 and 8.2, respectively [26]. As such, the ZPC of fly ash is deduced to be near neutral pH, indicating that fly ash is positively charged under acidic conditions and negatively charged under alkaline conditions. Considering the pK_{a1} (11.4) and pK_{a2} (~1) of AO7 [27], AO7 molecules are more positively charged under acidic conditions while more negatively charged under alkaline conditions. As a result, more AO7 molecules are adsorbed onto fly ash at neutral pH conditions, which facilitates the sonocatalytic degradation on the active sites.

The effect of glassware shape was investigated by adding the same volume of AO7 solution into colorimeter tube, beaker, and conical flask. The result is

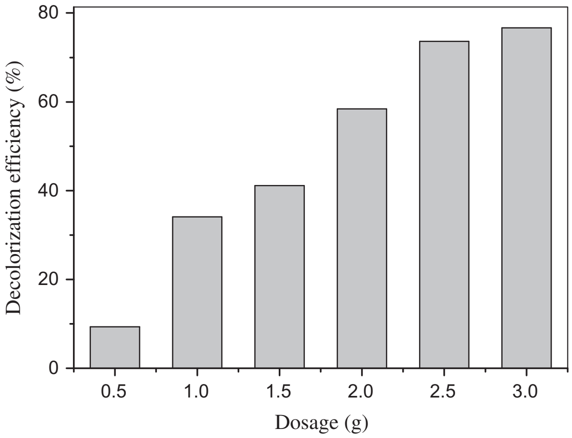

Fig. 6. Effect of fly ash dosage on AO7 decolorization. Reaction time, 60 min.

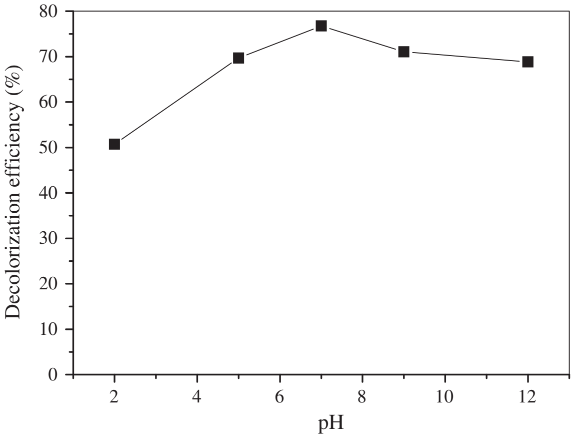

Fig. 7. Effect of solution pH on AO7 decolorization.

presented in Fig. 8. The bottom of the three vessels was smooth and flat. The bottom area of beaker and conical flask was similar, while the bottom area of colorimeter tube was one-twentieth that of beaker used. The decolorization efficiencies of the dye in the three different reactors are very close between 59.1 and 66.3%. However, some researchers found that both reactor diameter and liquid height significantly influenced the degradation of methyl orange under 200-kHz indirect sonication [28]. The frequency set in this study was 40 kHz. Though the glassware shape had a minor influence on dye degradation in this research, it can be deduced that the reactor shape could dramatically affect the degradation efficiency under some reaction conditions.

3.4. Reaction mechanism

AOPs are characterized by the production and utilization of hydroxyl radicals. Along with the generation of cavitation bubble by ultrasonication, thermolytic reactions occur in the hot cavitation bubble itself or at the interfacial region between the gaseous bubble and the surrounding liquid. Hydroxyl radicals and H_2O_2 can be generated by sonolysis alone according to the following reactions [29]:

$$H_2O))))) \rightarrow H^{\cdot} + HO^{\cdot} \tag{3}$$

$$H^{\cdot} + O_2 \rightarrow {}^{\cdot}HO_2 \rightarrow HO^{\cdot} + 1/2O_2 \tag{4}$$

$$O_2 \rightarrow 2 \cdot O \tag{5}$$

$$^{\cdot}O + H_2O \rightarrow 2HO^{\cdot} \tag{6}$$

$$HO^{\cdot} + HO^{\cdot} \rightarrow 2H_2O_2 \tag{7}$$

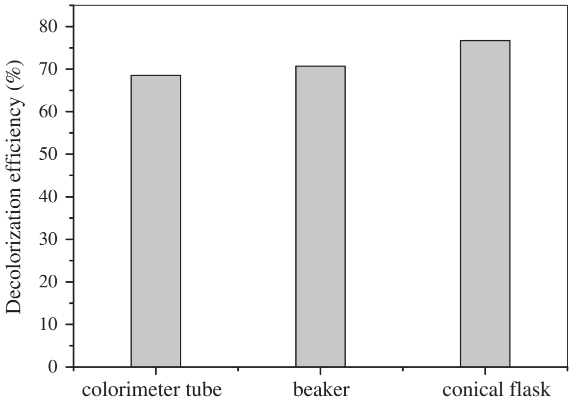

Fig. 8. Effect of glassware shape on AO7 decolorization. Reaction time, 60 min.

Meanwhile, the formation of cavitation bubbles can be increased by the heterogeneous nucleation of bubbles, generating hot spots in the solution. These hot spots can cause H_2O molecules to pyrolyze to form more OH radicals as well. From this point of view, sonocatalytic process is characterized by the generation of hydroxyl radicals as other AOPs.

Alcohols are commonly used to estimate the contribution of hydroxyl radicals through scavenger quenching method [19,30,31]. ISP was used in this study to evaluate the production of OH radicals. As showed in Fig. 9, the K_1 for the degradation by ultrasonic irradiation and fly ash decreased from 0.0246 min^{-1} without ISP to 0.0185 min^{-1} in the presence of ISP, which means that the contribution percentage of hydroxyl radicals amounts to only 24.8% for the total reaction. Nevertheless, Nakui and co-workers considered that coal ash accelerated phenol degradation due to the increase in hydroxyl radicals [7]. Though OH radicals are generally considered to be responsible for the sono chemical degradation of organic pollutants, its contribution to the degradation of AO7 in this study was not as significant as expected in this research.

On the other hand, the degradation performance of ultrasonication is determined by the cavitation effect while the cavitation effect is dependent on the cavitation bubbles generated in the process. The generation of cavitation bubbles can be accelerated by the presence of heterogeneous catalyst which acts as nucleus [32]. Once the size of catalyst particle is in the same order of magnitude as the resonance size of cavitation bubbles, these catalysts could act as cavitation nucleus [33]. The resonance size of cavitation bubbles at given ultrasound frequency can be estimated as [6,32]:

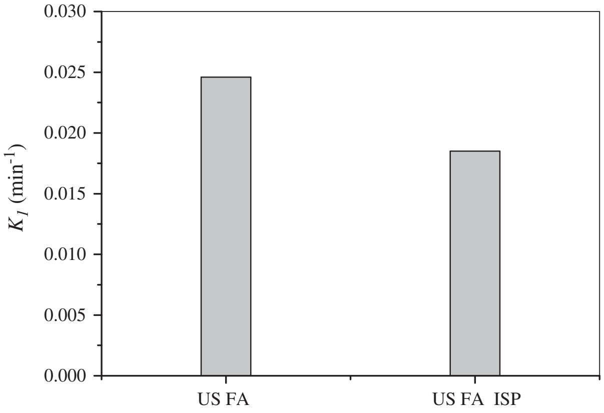

Fig. 9. Effect ISP quenching on K_1 for AO7 decolorization by fly ash under ultrasonic irradiation. Reaction time, 60 min.

$$f = \frac{\sqrt{\frac{3\gamma}{\rho}\left(P_0 + \frac{2\sigma}{\alpha}\right) - \frac{2\sigma}{\alpha\rho}}}{2\pi\alpha} \quad (8)$$

where f is the ultrasound frequency, α is the radius of bubbles, γ is the ratio of heat capacities of saturating gas at constant pressure and volume, ρ is the density of surrounding medium, σ is the surface tension, and P_0 is the ambient pressure.

In this experiment, f was 40 kHz, γ was 1.39 for air, σ was negligible, and water density (ρ) was 1.0 g cm^{-3}. Accordingly, the resonance size of cavitation bubbles in water is calculated to be 43.1 μm. As the resonance size of cavitation bubbles is in the size range of fly ash particles, they could act as nucleus to enhance the generation of more cavitation bubbles and concurrently improve the ultrasonic degradation performance. AO7 was actually subjected to sonocatalytic degradation by fly ash under ultrasonic irradiation. In addition, Nakui and co-workers considered that coal ash (53–106 μm in particle size) has no catalytic property for the formation of hydroxyl radicals [7]. The enhanced sonochemical degradation performance was regarded as a consequence of the surface roughness of the particles [7,34]. In short, both the sizes and surface roughness of fly ash particles might have contributed to the sonocatalytic degradation process.

4. Conclusion

AO7 was decolorized by fly ash under ultrasonic irradiation. Combining ultrasonic irradiation and fly ash achieved particularly higher AO7 degradation efficiency compared with individual process. A significant synergetic effect was firstly observed for the combined sonocatalytic process. Ultrasonic irradiation did not induce the phase conversion of fly ash or morphology change. It revealed that the contribution percentage of hydroxyl radicals amounted to only 24.8% for the total reaction. Theoretic analysis implied that fly ash particles could act as nucleus to generate cavitation bubbles and enhance the ultrasonic degradation efficiency.

Acknowledgments

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