Effects of gas sparging and mechanical mixing on sonochemical oxidation activity

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ABSTRACT

The effects of air sparging (0–16 L min\(^{-1}\)) and mechanical mixing (0–400 rpm) on enhancing the sonochemical degradation of rhodamine B (RhB) was investigated using a 28 kHz sonoreactor. The degradation of RhB followed pseudo first-order kinetics, where sparging or mixing induced a large sonochemical enhancement. The kinetic constant varied in three stages (gradually increased → increased exponentially → decreased slightly) as the rate of sparging or mixing increased, where the stages were similar for both processes. The highest sonochemical activity was obtained with sparging at 8 L min\(^{-1}\) or mixing at 200 rpm, where the standing wave field was significantly deformed by sparging and mixing, respectively. The cavitational oxidation activity was concentrated at the bottom of the sonicator when higher sparging or mixing rates were employed. Therefore, the large enhancement in the sonochemical oxidation was attributed mainly to the direct disturbance of the ultrasound transmission and the resulting change in the cavitation-active zone in this study. The effect of the position of air sparging and mixing was investigated. The indirect inhibition of the ultrasound transmission resulted in less enhancement of the sonochemical activity. Moreover, the effect of various sparging gases including air, N\(_2\), O\(_2\), Ar, CO\(_2\), and an Ar/O\(_2\) (8:2) mixture was compared, where all gases except CO\(_2\) induced an enhancement in the sonochemical activity, irrespective of the concentration of dissolved oxygen. The highest activity was obtained with the Ar/O\(_2\) (8:2) mixture. Therefore, it was revealed that the sonochemical oxidation activity could be further enhanced by applying gas sparging using the optimal gas.

1. Introduction

The removal of non-biodegradable and recalcitrant pollutants including phenols, dyes, natural organic matters, chlorinated compounds and personal care products (PPCPs) from water is a major task for the protection of aquatic ecosystems and the sustainable use of water resources [1–3]. Advanced oxidation processes (AOPs) have captured the attention of researchers for the effective treatment of conventional and emerging recalcitrant pollutants due to their ability to produce highly reactive oxidizing species such as hydroxyl radicals and sulfate radicals. It is common to use various chemicals including ozone, hydrogen peroxide, persulfate, and catalysts (TiO\(_2\), Fe(0), Fe(II)/Fe(III)) with wave energy (ultraviolet and ultrasound) and electrochemical technologies in AOPs [4–8].

Acoustic cavitation can be considered a “green chemistry” technique because various chemical and physical effects can be induced with no chemicals [9,10]. Although highly synergistic effects between ultrasound technology and chemicals or other AOPs have been widely reported [5,7,11–13], some researchers have reported remarkable enhancement of the sonochemical activity with no chemicals by altering the geometry of the liquid body, where the ultrasonic energy spreads out and cavitation occurs [14–20]. Asakura et al. investigated the effect of the liquid height/volume on the sonochemical oxidation activity under various frequency conditions and suggested an empirical relationship between the applied frequency and the liquid height at which the highest sonochemical activity was achieved [19,20]. Son et al. evaluated the input power, applied frequency, and liquid height/volume and suggested optimal liquid height ranges for maximizing the sonochemical activity depending on the applied frequency and input power [16–18]. They also reported an enhancement of the sonochemical activity by changing the probe position in the vessel in 20 kHz probe systems [14,15].

Abbreviations: AOPs, advanced oxidation processes; DO, dissolved oxygen; RhB, rhodamine B; SCL, sonochemiluminescence; SL, sonoluminescence

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Recently, an enhancement of the sonochemical activity was reported when the degree of instability of the liquid body in the sonorreactor increased. Hatanaka et al. reported an increase in the sonochemiluminescence (SL) intensity using flow circulation and mechanical stirring [21]. Kojima et al. induced mechanical flow in a sonorreactor using an overhead stirrer (0–350 rpm) and reported enhancement of the sonochemical activity by monitoring the sonochemiluminescence (SCL) intensity and triiodide ion production [22]. Bussemaker and Zhang investigated the effect of mechanical mixing on the sonochemical activity (SCL and $\text{H}_2\text{O}_2$) with variation of the applied frequency and power, demonstrating that the mixing rate influenced the sonochemical activity [23]. The effect of the sparging gas was also evaluated, and the mixing effect of sparging and consequent enhancement of the sonochemical activity were discussed [24–26]. The enhanced sonochemical activity was largely attributed to violent mixing in the liquid phase and an increase in the number of the cavitation-active bubbles due to anti-coalescence.

In our recent research, we investigated the effect of air sparging at various positions in a sonorreactor and reported a large enhancement of the sonochemical activity. This enhancement was attributed not only to violent mixing in the sonorreactor, but also the significant change in the sound energy field and cavitation-active zone [26]. In this study, the effect of air sparging and mechanical mixing on the sonochemical activity is investigated to understand the relationship between the degree of inhibition of ultrasound transmission and the enhancement of the sonochemical activity. The oxygen mass transfer under ultrasound irradiation and air sparging is analyzed to quantify the observed ultrasongic degassing. The sonochemical oxidation activity is analyzed by monitoring rhodamine B degradation with variation of the ultrasound power, air sparging rate, and mechanical mixing rate conditions. Rhodamine B is one of recalcitrant pollutants in aqueous environments and has been widely investigated in AOPs. In addition, the effects of the sparging position and dissolved gases on the sonochemical activity are studied.

2. Materials and methods

2.1. Chemicals

Rhodamine B and luminol (3-aminophthalhydrazide) were acquired from Sigma–Aldrich Co. (St. Louis, USA). Sodium hydroxide (NaOH) was acquired from Junsei Chemical Co., Ltd. (Tokyo, JPN). All chemicals were used as received.

2.2. Experimental setup

A 28 kHz bath-type sonorreactor ($L \times W \times H$: 330 mm $\times$ 220 mm $\times$ 450 mm) equipped with a transducer module (Miraue Ultrasonic Tech., Bucheon, KOR) at one side of the bottom was used in this study (Fig. 1). The ultrasonic module ($L \times W \times H$: 195 mm $\times$ 195 mm $\times$ 90 mm) was equipped with nine 28 kHz PZT (lead zirconate titanate) transducers. The liquid height/volume was determined based on the wavelength of the applied frequency and the wavelength was calculated as follows:

$$\text{Wavelength} (\lambda) = \frac{c}{f} \quad (1)$$

where $c$ is the speed of sound in water (1500 m s$^{-1}$) and $f$ is the applied frequency (28 kHz). The liquid height from the module to the liquid surface was 107 mm (2$\lambda$) and the corresponding total liquid volume was 11.2 L. The temperature was maintained at 20 $\pm$ 2°C using a cooling system (WiseCirc® WCR-P8, DAIHAN Scientific Co., KOR). The liquid body in the reactor was open to air.

The working input electrical power was 130 ± 10 and 205 ± 10 W, as measured using a power meter (HPM-300A, ADPower, KOR). The ultrasonic energy, designated as the calorimetric power in this study, was calculated as follows:

$$P_{\text{cal}} = \frac{dT}{dt}C_pM \quad (2)$$

where $P_{\text{cal}}$ is the ultrasonic/calorimetric energy, $dT/dt$ is the rate of increase of the liquid temperature, $C_p$ is the specific heat capacity of the liquid (4.184 J g$^{-1}$K$^{-1}$ for water), and $M$ is the mass of the liquid.

A micro-porous glass sparger equipped with a glass pipe was used to continuously supply gas, including air, $\text{N}_2$, $\text{O}_2$, Ar, $\text{CO}_2$, and an Ar/$\text{O}_2$ (8:2) mixture, to the liquid. The gas sparging also induced violent liquid mixing effect in the reactor without a mechanical stirrer. The sparger was located 1 cm above the center of the transducer module (“②” in Fig. 1), where the highest sonochemical oxidation was observed among the various vertical positions evaluated in our previous study [26]. The concentration of dissolved oxygen (DO) was measured at several points before and after ultrasound irradiation using a DO meter (ProODO; YSI Inc., USA). Before each experiment, the solution was saturated with an applied gas and the degree of gas saturation was checked by measuring the DO concentration. The gas flow rate ranged from 1 to 16 L min$^{-1}$. A stainless-steel overhead stirrer was placed at the same position of the sparger, as shown in Fig. 1. The mixing speed ranged from 50 to 400 rpm. In order to investigate the effect of sparging or mixing without direct contact with the ultrasound transmission, the sparger or stirrer was placed at “③” (vertical position: 1 cm above the transducer module; horizontal position: 6 cm away from the right-hand-side wall).

2.3. Quantification and visualization of sonochemical oxidation

To quantitatively analyze the sonochemical oxidation process, the degradation of rhodamine B was evaluated using a spectrophotometric method measured at 554 nm (Vibra S60, Biochrom Ltd., UK). The initial concentration of the rhodamine B solution was 5 mg L$^{-1}$, where the solution was prepared with tap water. No significant difference in water quality was observed during the experiments. The ultrasound irradiation time was 180 min and the concentration of rhodamine B was monitored at 20 min intervals. All measurements were repeated three times; the average values are reported herein.

The sonochemically active zone was visualized using luminol solution (0.1 g L$^{-1}$ luminol and 1 g L$^{-1}$ NaOH) in a completely dark room [14,26]. SCL images were acquired using an exposure-controlled digital camera (g58; Sony Corp., JPN).

3. Results and discussion

3.1. Ultrasonic degassing and gassing

The effects of the ultrasound treatment and air sparging on the mass transfer of oxygen from air to water (the absorption of oxygen in water) were investigated using the 28 kHz sonorreactor and the DO meter. The water in the sonorreactor was degassed by sparging with nitrogen gas and the initial DO concentration was then determined, giving a value of 2.1 ± 0.2 mg L$^{-1}$. Fig. 2 shows the change in the DO concentration under various conditions. When the DO concentration is below the saturation/equilibrium concentration, oxygen is absorbed into the water spontaneously until the concentration reaches the saturation concentration. The mass transfer coefficient can be obtained according to the following equations:

$$\frac{dC}{dt} = K_i a (C_s - C) \quad (3)$$

$$\ln(C_s - C) = -K_i a t + \ln(C_s - C_0) \quad (4)$$

where $C$ is the DO concentration (mg L$^{-1}$), $C_s$ is the DO saturation concentration (mg L$^{-1}$), $C_0$ is the initial DO concentration (mg L$^{-1}$), and $K_i a$ is the observed volumetric mass transfer coefficient (min$^{-1}$) [27]. In this study, the DO saturation concentration was 8.9 ± 0.4 mg L$^{-1}$ at
the temperature of the room (20 °C). For the "No US and No Sparging" system, no significant increase in the DO concentration (ΔDO: +0.16 mg L⁻¹) was observed over the course of 10 min, even though the difference between the saturation DO concentration and the actual DO concentration, which is considered as the driving force for oxygen mass transfer, was very large. The DO concentration did not increase noticeably during the given time period probably because of the relatively large water volume (11.2 L) compared with the water surface area (330 mm × 220 mm). Mass transfer might occur violently only in the region adjacent to the water surface. However, increasing the DO concentration in the region of the water surface region might contribute marginally to the bulk DO concentration without agitation in the reactor.

It is well known that ultrasonic degassing occurs by the following mechanism: 1) dissolved gas molecules are transferred into the cavitation bubbles in the acoustic cavitation field; 2) the bubbles grow by rectified diffusion and coalescence; 3) bubbles larger than the cavitation-active bubbles float to the top by buoyant force and the dissolved gases are thus removed from the liquid [28–30]. Gondrexon et al. reported a newly defined equilibrium concentration of DO for ultrasound irradiation, which was obtained by considering both the ultrasonic gas desorption and concentration-difference-driving gas absorption...
In their study, the range for the natural equilibrium DO concentration was 8.5–9.2 mg L⁻¹, while the new equilibrium concentration was 5.6–5.9 mg L⁻¹. They found that gas absorption occurred via the formation of an acoustic fountain upon contact of the water in the reactor with the ambient air, and the degassing and gassing were balanced during ultrasound irradiation [31]. Yanagida also reported that the DO saturation decreased from 100% and then remained constant at approximately 50% as the irradiation time elapsed (1 MHz, Vₛ = 16 mL) [32]. Laugier et al. compared ultrasonic gassing and degassing under mechanical stirring and found that the ultrasound was more effective in the degassing processes (20 kHz, Vₛ = 1 L) [33].

In this study, it was observed that ultrasound irradiation had a positive impact on the recovery of the DO concentration. The volumetric mass transfer coefficient, K_La, for the “US” system was approximately 22 times larger than that obtained for the “No US and No Sparging” system (ΔDO: +2.47 mg L⁻¹). The enhanced oxygen mass transfer in the “US” system may be due to the ultrasound irradiation force, which can induce a macro-scale flow in the bulk water phase and move the transferred oxygen molecules down the water surface. The ripples on the water surface induced by the ultrasound irradiation might also contribute to the increased DO concentration. Note that ultrasonic degassing seemed to be operative, given that bubbles escaping from the water were visually observed during the ultrasound irradiation. Therefore, the ultrasonic degassing was considered to be negligible owing to the relatively large liquid volume (Vₗ = 11.2 L) compared with the liquid volumes in the abovementioned studies, whereas ultrasonic gassing occurred actively in this study. Moreover, further evaluation showed that the DO concentration remained constant in the initially air-saturated water during ultrasound irradiation. Air sparging (4 and 16 L min⁻¹) increased the DO concentration rapidly, and the application of ultrasound further enhanced the mass transfer (by ~10–20%). A higher gas flow rate induced a higher bubble density (or higher gas holdup) in the reactor and faster mass transfer despite the larger size of the bubbles [34–37]. In this study, larger bubbles and higher gas holdup were observed at higher flow rate. The concentration-difference driving mass transfer might also be significantly enhanced because of the large fluctuation of the water surface, as shown in Fig. S1. It is suggested that in the mechanism of ultrasonic enhancement of the mass transfer, the bubbles or bubble clusters may be dispersed and split in the cavitationally active zone, thereby increasing the interfacial area between the bubbles and liquid [33,36]. The bubble oscillation induced by the acoustic wave may also accelerate mass transfer between the air bubbles and water [38]. In an additional test, no significant difference in the DO concentration was observed for the initially air-saturated water (air-saturated DO conc.:
8.9 ± 0.4 mg L⁻¹ at room temperature) when sparging or sparging/ultrasound was applied. This is because the DO concentration in the atmosphere did not increase above the saturated concentration under normal conditions and the DO depleted by ultrasonic degassing or by consumption for the cavitation events was recovered immediately via ultrasonic gassing and air sparging.

3.2. Effects of air sparging and mixing

The sonochemical degradation of an aqueous pollutant under ultrasound irradiation (calorimetric power: 60 and 120 W) with air sparging (flow rate: 0, 1, 2, 4, 8, and 16 L min⁻¹) or mechanical mixing (0, 50, 100, 200, 300, and 400 rpm) was investigated using rhodamine-B (RhB) as a model pollutant at an initial concentration of 5 mg L⁻¹ in tap water. Fig. 3 shows the pseudo first-order reaction kinetic constants for the ultrasonic process, ultrasonic process with sparging, and ultrasonic process with mixing, obtained by applying Eq. (5) [39–41]:

\[
\frac{dC}{dt} = k[RhB] \cdot [-OH] = k'[RhB]
\]  

where \( k \) is the second-order reaction constant, \([RhB]\) is the RhB concentration, \([-OH]\) is the concentration of hydroxyl radicals generated by cavitation, and \( k' = k \cdot [-OH] \) is the pseudo first-order reaction constant. The concentration of the OH radical can be considered as constant under continuous and stable ultrasound irradiation [39] and the second-order rate equation reduces to the first-order rate equation. Previous researchers investigated the removal of RhB using advanced oxidation technologies and detected various degradation intermediates [41,42]. It was found that the \( K_{OH} \) value of all detected intermediates was much lower than that of RhB. Thus, the OH radicals were much more likely to react with the RhB molecules than the degradation byproducts. Pétrier et al. reported that a less hydrophobic compound (4-chlorophenol) started to degrade only after a more hydrophobic compound (chlorobenzene) was almost degraded when both compounds were present simultaneously under 300 kHz ultrasound [43]. Therefore, the assumption of a pseudo first-order reaction for RhB degradation in this study would be quite reasonable, and was confirmed from the \( R^2 \) values which were very close to unity, as shown in Fig. 3. No significant degradation was detected in the single and combined processes of air sparging and mechanical mixing without ultrasound irradiation. In addition, no significant difference was obtained for the experiments with and without the sparger or the stirrer in the reactor even though slight changes of the cavitation-active zone in SCL image were observed as shown in Fig. 4 [26].

The co-application of air sparging or mechanical mixing resulted in a significant enhancement (up to approximately 600% compared to the US single process) of the first-order degradation constant under the ultrasonic power of 60 and 120 W. The kinetic constant varied quite similarly in three stages as the sparging or mixing rate increased. In the first stage, the constant increased gradually, and the increase was more pronounced in the US/sparging processes. In the second stage, the constant increased exponentially with sparging at 8 L min⁻¹ and mixing at 200 rpm. Subsequently, the constant decreased slightly in the third stages. The kinetic constants in the third stage were still much higher than those in the first stage in this study. Note that the mechanism for enhancement of the cavitational degradation via sparging and mixing might be different due to differences in shape, size, materials, and other characteristics of the sparger or mixing blade.

Generally, it can be understood that the presence of a large amount of bubbles, solid materials, or strong fluid flow in the pathway of ultrasound transmission inhibits the distribution of the ultrasound and the cavitational activity in the system [37]. However, improved cavitational activity due to air/gas sparging [24,26,38,44], fluid flow [21,45], and mechanical stirring/mixing [21–23,25,46] was also reported previously. The enhancement mechanisms were suggested to proceed as follows: gas sparging, fluid flow, and mixing induced intensive liquid circulation in the reactor and enhanced movement of the target reactants to the more cavitationally active zone [22,24,26]. The fluid flow and mixing prevented coalescence of the bubbles and reduced the generation of large bubbles that were cavitationally inactive [21–23]. Nucleation for the cavitation events could be enhanced, and gas molecules essential for increasing the cavitational activity could be supplied by gas sparging [23,25,39].

Consistent with our recent study, it was also revealed in this study
Fig. 5. Relative sonochemiluminescence (SCL) intensities obtained from the images in Fig. 4. The number in parentheses represents the relative total intensity of SCL.

(a) Air sparging

(b) Mechanical mixing
that deformation or elimination of the standing wave field in the bath-type sonoreactor via sparging and mixing directly adjacent to the transducer module may contribute to enhancing the cavitational activity [26]. Previous researchers briefly mentioned the deformation or elimination of the standing wave field [23,25]. Without sparging or mixing, however, it was reported that high cavitational activity could be obtained through spatially stable distribution of the cavitation bubbles, which could be attained when a stable standing wave field was formed [29,47]. As shown in Figs. 4 and 5, a gradual elimination of the standing wave field was observed as the sparging and mixing rate increased. At lower rates of sparging or mixing (2 L min\(^{-1}\) or 100 rpm), bright and repetitive stripes, providing evidence of formation of the standing wave field, were still observed and a relatively low enhancement of the sonochemical activity was achieved. However, the stripes diminished gradually, and no stripes were observed for sparging and mixing at higher rates. A large sonochemical enhancement was obtained from the disappearance of the standing wave field (from 8 L min\(^{-1}\) and 200 rpm for US 120 W).

Interestingly, the SCL intensity in the zone close to the transducer module increased markedly as the sparging and mixing rate increased. A large portion of the total SCL intensity was concentrated in the zone corresponding to 10% of the total liquid height under the conditions at which the standing wave field disappeared (from 8 L min\(^{-1}\) and 200 rpm for US 120 W). The images and profiles of the SCL indicated that large portion of the ultrasonic energy was trapped in the bottom zone and the remaining portion of the energy was largely attenuated as it was transmitted upward through the sparged bubbles and the movement of the blades. The concentration of the sonochemical activity in the bottom zone might be due to the presence of large amount of bubbles for air sparging and the formation of vortex-generated hollow filled with air and entrapped air bubbles drawn by the vortex movement for mechanical mixing. As shown in Fig. S1, the portion of air-filled space increased in the ultrasound transmission pathway as the sparging rate or mixing rate increased. The transmitted ultrasound could not induce formation of the stable and strong standing wave field because the liquid surface, acting as a reflector, was very unstable and the intensity of the ultrasound was very low. It would be reasonable to consider that the trapped ultrasonic energy in the bottom zone is the main contributor to the large enhancement of the kinetic constant because a large sonochemical enhancement and a high SCL intensity in the bottom zone were obtained when high sparging and mixing rates were employed. Recently, it was also reported that very high sonochemical activity could be obtained when a 20 kHz probe was placed very close to the bottom of the reactor, and the ultrasound irradiation was inhibited significantly by the bottom of the reactor [14].

Note that the significant change in the total SCL intensity, representing the large enhancement in the sonochemical activity due to sparging and mixing, was not observed in this study. The capture of light emitted from the reactions of luminol might be negatively affected by the movement of the sparged bubbles, fast rotation of the mixing blade, and sparging/mixing-derived fluid flow. Therefore, the present SCL analysis focused on the formation of the active zone in each image.

### 3.3. Effects of sparging/mixing position and sparging gases

The effect of the position of the sparger or the stirrer was...
investigated for two positions, i.e., 1 cm above the center of the transducer module, and 6 cm away from the right-hand-side wall. The former position was used for Sections 3.1 and 3.2 above and is represented as “0” in Fig. 1. The latter was used to apply sparging or mixing without direct contact with the ultrasound transmission and is represented as “②”. As shown in Fig. 6, air sparging or mechanical mixing at position “②” also induced violent fluid flow in the reactor, and higher activity was observed compared to the cases with no sparging or mixing (US 60 W and US 120 W). However, the enhancement of the cavitation activity at “②” was less than that at “0”; it appears that more direct disturbance of the ultrasound transmission by sparging or mixing induced much higher cavitation activity than that caused by the generation of violent flow in the reactor [26]. Indirect disturbance of the ultrasound transmission could also be induced by the violent flow generated at “②”. In addition, the enhancement of the cavitation activity via ultrasound transmission disturbance or the generation of violent flow seemed to be maximized under certain optimal conditions.

The simultaneous application of sparging and mixing at “0” and “②” resulted in reduced enhancement (approximately 10%) compared to the application of sparging or mixing individually under US 120 W. This might be due to increased attenuation of the ultrasound caused by the very high instability and heterogeneity of the liquid body in the reactor.

Fig. 7 shows the effect of the sparging gas on RhB degradation using various gases including air, N\textsubscript{2}, O\textsubscript{2}, Ar, CO\textsubscript{2}, and an Ar/O\textsubscript{2} (8:2) mixture. The gas sparging was performed for 20 min before and during the experiment. The DO concentration was monitored to measure saturation with the sparged gas directly and indirectly, and the average DO concentration during the experiment for each case is also shown in Fig. 7. No significant variation in the DO concentration was observed. In our preliminary test, the effect of saturated gases on the sonochemical activity in the same system and it was found that the dissolved gas concentration varied significantly when the system was exposed to air. Therefore, we focused on the gas sparging effect on the sonochemical oxidation activity under the constant dissolved gas condition in this study.

It is well known in sonochemistry that the dissolved oxygen molecules play an important role in generating various oxidizing radicals such as hydroxyl radicals (\cdot\text{OH}), hydroperoxyl radicals (\cdot\text{OOH}), and oxygen atoms (\cdot\text{O}), while the dissolved argon molecules enhance the severity of the cavitation phenomena significantly [43,48–50]. In this study, however, the sonochemical oxidation activity was more significantly affected by the gas sparging rate than by the type and concentration of dissolved gases. When the DO concentration (0.3 (3% saturation) was increased to 41.0 mg L\textsuperscript{-1} (460% saturation; see Fig. 7), the difference between the reaction constants at the same sparging rate was not larger than expected from previous studies, except in the case of CO\textsubscript{2}. The dissolved CO\textsubscript{2} molecules appeared to act as a very powerful radical scavenger [11,51,52] and no activity was observed in spite of the violent sparging action. The order of magnitude of the reaction constants differed depending on the sparging rate. The reaction
constant for sparging at 4 L min\(^{-1}\) decreased in the order: Ar > O\(_2\) > air ≈ N\(_2\) > no sparging ≥ CO\(_2\). It was reported that the presence of excess Ar or O\(_2\) reduced the cavitational activity [25,48,50], and the order in which the sonochemical reactions was enhanced varied depending on the sonication frequency or the target reaction [49,50,53]. The highest reaction constant in this study was obtained for the mixture of Ar/O\(_2\) (8:2), termed the optimal gas condition for sonochemical oxidation [25,48]. Therefore, the data reveal that the sonochemical oxidation activity could be further enhanced by combination with gas sparging using the optimal gas mixture of Ar and O\(_2\).

4. Conclusions

The effect of air sparging and mechanical mixing on the sonochemical degradation of RhB was investigated in a 28 kHz sonoreactor. Sparging and mixing induced violent liquid flow and instability of the liquid body, and a large enhancement of the sonochemical oxidation activity was observed. Oxygen mass transfer tests indicated that no ultrasonic degassing effect was operative, rather ultrasound irradiation noticeably enhanced oxygen mass transfer from air to the liquid. The sonochemical oxidation activity was significantly enhanced by the application of air sparging or mechanical mixing with ultrasound irradiation. The pseudo first-order kinetic constant varied in three stages, i.e., it gradually increased, increased exponentially, the decreased slightly. Increasing the sparging or mixing rate led to increasing severity of the deformation of the standing wave field, and the cavitation activity was concentrated at the bottom region adjacent to the transducer module. These phenomena are considered the main origins of the enhanced sonochemical oxidation induction by sparging and mixing in this study. Therefore, direct inhibition of the ultrasound transmission may be critical for obtaining high cavitation activity, as further confirmed when sparging or mixing was applied on the other side of the transducer module in the reactor. All gases used for sparging, except CO\(_2\), enhanced the sonochemical activity, and the order of magnitude of the kinetic constants varied depending on the sparging rate.

CRediT authorship contribution statement

Jongbok Choi: Validation, Writing - original draft. Hyeonjae Lee: Methodology, Investigation. Younggyu Son: Conceptualization, Methodology, Writing - review & editing, Visualization, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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