

The effect of gas sparging on sonochemical oxidation reactions

Hyeon-jae Lee, Wontae Lee, Younggyu Son[†]
(Department of Environmental Engineering, Kumoh National Institute of Technology)

1. Introduction

Acoustic cavitation can occur in aqueous phase when ultrasound with enough intensity is irradiated. The sonochemical effects including pyrolysis and radical reactions and the sonophysical effects including microjet and shockwave are created by the cavitation events. Some novel processes have been developed using these unique cavitation effects in environmental, chemical, material, and energy engineering fields.

Advanced oxidation processes (AOPs) are state-of-the-art water/wastewater treatment processes for the removal of recalcitrant pollutants such as chlorinated compounds, phenols, dyes, pharmaceuticals, and endocrine disrupting compounds. Recently ultrasound is considered as one of powerful AOPs and green processes because acoustic cavitation can induce the degradation and mineralization of aqueous pollutants without chemicals.

To enhance the degradation and mineralization reactions of aqueous pollutants in sonochemical processes, various methods including the combination with other AOPs, the addition of chemicals and solid materials, dissolved gas saturation, and mechanical agitation have been applied. Among these enhancement methods, the last two seemed to be cost effective and easily applicable. Moreover post treatment processes for the removal of additives and byproducts are not required.

In this study the effect of continuous gas sparging on sonochemical oxidation reactions was investigated in a 36 kHz pilot scale sonoreactor. The gas sparging enabled to provide gas molecules consistently during the cavitation events and induce violent mixing in the whole sonoreactor. The sonochemical oxidation was quantified using the KI dosimetry and luminol images were taken for the visualization of cavitation active zone.

2. Experimental

The reactor in this study was an upward-irradiation sonoreactor, also called a standing-wave type reactor, as shown in Fig. 1.

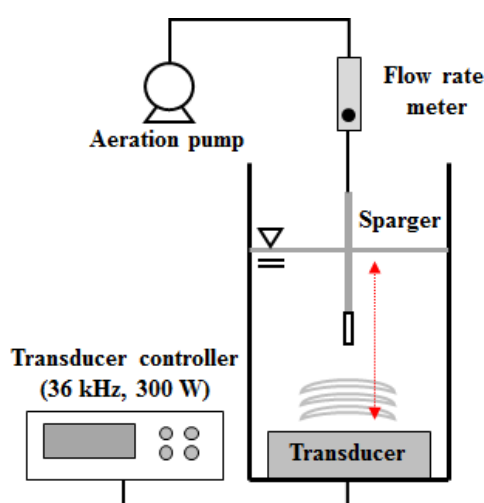


Fig. 1 Schematic of a 36 kHz pilot-scale sonoreactor used in this study.

The transducer module (200 X 200 X 100 mm³) including nine transducers were equipped at the bottom of the sonoreactor (210 X 210 X 500 mm³). The frequency and input power were 36 kHz and 300 W, respectively. The overall liquid height was determined in our preliminary test and the liquid height of 250 mm (6 λ) was applied. The equivalent liquid volume was 11.8 L.

The pyrex-glass gas sparger was placed 1 cm above the transducer module in the center of the sonoreactor. Various gases including air, N₂, N₂/O₂ (50:50), and O₂ were supplied using a gas cylinder or a air pump at the flow rates of 3, 6, and 9 L/min.

The KI dosimetry was used to quantify the sonochemical oxidation reactions. The irradiation time was 30 min. The triiodide ion concentration was measured using a UV-vis spectrophotometer (Biochrom Libra S60) at 350 nm. The dissolved oxygen was measured using a DO meter (YSI Pro20).

[†]e-mail address: yson@kumoh.ac.kr

3. Results and discussion

The Fig. 2 shows the variation of the concentration of sonochemically generated triiodide ion for the gas sparging of air, N₂, N₂/O₂, and O₂. Compared with the value at 0 L/min (no sparging), relatively high concentrations were observed for most sparging cases. The higher flow rate resulted in more severe sonochemical oxidation reactions (higher concentration of triiodide ion). The enhancement by the gas sparging might be mainly due to the violent mixing induced by rising bubbles from the sparger placed at the bottom of the sonoreactor. Kojima et al. reported that the sonochemical efficiency value significantly increased as the mixing intensity increased in a 490 kHz sonoreactor.

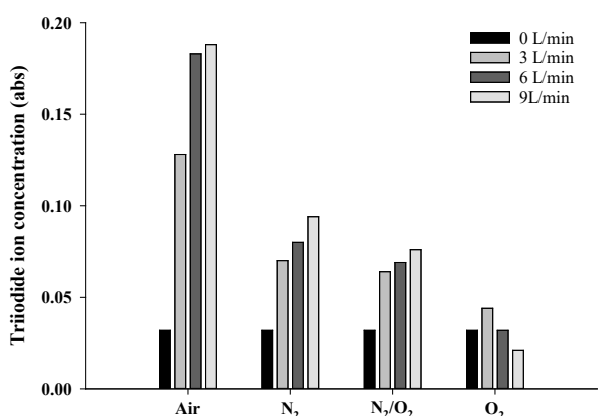
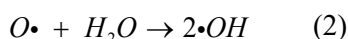


Fig. 2 The variation of the concentration of triiodide ion for air, N₂, N₂/O₂, and O₂ at the gas flow rate of 0, 3, 6, and 9 L/min.

It is generally known that oxygen molecules can be the source of oxidizing radicals such as hydroxyl radical during acoustic cavitation events. Pétrier et al. reported that faster degradation of 4-chlorophenol in oxygen-saturated condition than in argon-saturated condition due to more generation of OH radicals expressed as follows:



In this study, however, very low concentrations of the triiodide ion were observed for the O₂ sparging and higher flow rate did not result in more sonochemical oxidation reactions.

To investigate the effect of oxygen supply on

sonochemical oxidation, the concentration of dissolved oxygen (DO) was measured for the rate of 6 L/min at the beginning and the end (30 min later) of the experiment. The initial and final DO saturation ratios were 105%/106%, 24%/26%, 200%/192%, and 460%/463% for air, N₂, N₂/O₂, and O₂, respectively. It seemed that sonochemical oxidation reactions were significantly inhibited by excessive oxygen. For the O₂ sparging, the inhibition by excessive O₂ was much larger than the enhancement by high rate mixing and it resulted in lower concentration of the triiodide ion at higher gas flow rate.

Acknowledgment

This research was supported by the Korea Ministry of Environment (MOE) as “Geo-Advanced Innovative Action” Program (Project No. 2012000550003 and 2015000560002)

References

1. D.H. Bremner, R. Molina, F. Martínez, J.A. Melero, and Y. Segura: *Appl. Catal. B: Environ.* **90**, (2009) 380.
2. M. Lim, Y. Son, B. Park, and J. Khim, *Jpn. J. Appl. Phys.* **49**, (2010) 07HE06.
3. Y. Son, M. Lim, J. Khim, L.-H. Kim, and M. Ashokkumar, *Chem. Eng. J.* **183**, (2012) 39.
4. R.A. Torres, J.I. Nieto, E. Combet, C. Pétrier, and C. Pulgarin, *Appl. Catal. B: Environ.* **80**, (2008) 168.
5. B. Neppolian, A. Doronila, F. Grieser, and M. Ashokkumar, *Environ. Sci. Tech.* **43**, (2009) 6793.
6. Y. Kojima, Y. Asakura, G. Sugiyama, and S. Koda: *Ultrason. Sonochem.* **17** (2010) 978.
7. C. Pétrier, E. Combet, and T. Mason, *Ultrason. Sonochem.* **14**, (2007) 117.