

Sonochemical green technology using active bubbles: Degradation of organic substances in water

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This review introduces recent studies on the sonochemical degradation of organic substances in aqueous solutions. Since the main advantage of this method is absent in added chemicals, this is applicable to many kinds of solutes with safely operation. Sonochemical degradation of organic substances can be applied for not only water treatment, but also food processing. In this review, organic substances are categorized as aromatic compounds, organic halogen compounds, polysaccharides, and microorganisms. Sonochemical degradation occurs in the three regions centered at cavitation bubbles. The optimum ultrasonic frequencies for the degradation are depended on the type of organic substances and are related to the chemical or physical effects of cavitation. The reaction kinetics and mechanism of sonochemical degradation are also discussed.

Keyword: Sonochemical degradation, Ultrasonic cavitation, Water treatment, Food processing, Sonochemistry

Introduction

Ultrasonic treatment degrades organic pollutants in wastewater including a wide range of chemical substances of carcinogens, medicines, dyes, and polymers. Such substances are difficult to degrade by activated sludge method which is conventional biological wastewater treatment. Ultrasound also has applied for disinfection to microorganisms. Further, even polysaccharides in foods can be degraded. Several advanced oxidation methods such as Fenton, ozone, and ultraviolet light have been explored to degrade these persistent chemicals. Among them, the ultrasonic method has several advantages. Ultrasonic irradiation generates active chemical species, in requiring for no additional chemicals, without production of harmful products. This method is applicable to treat many kinds of wastewater including coloring materials and polymers. Operation is simple with ensuring-safety. However, disadvantages are a lack of reaction selectivity and large-scale reactor. Since it employs water only at ordinary temperatures and pressures, ultrasonic method is a green process and technology.

In ultrasonic method, cavitation with high-power ultrasound waves of frequencies in the range of 20 kHz to several megahertz generates fine bubbles, originating from bubble nuclei, growing the resonance size under acoustic pressure fluctuations, and then rapidly collapsing it [1]. So, during the bubble collapse, the temperature reaches to thousands of degree Kelvin and to the pressure of several hundred atmospheres. Simultaneously, shock waves, shearing stress, and jet flow are also generated in ultrasonic water medium. Such non-equilibrium reaction media can localize high-temperature and high-pressure field [2] and become the source of the chemical and physical effects.

Figure 1 illustrates bubble in bulk aqueous medium. In an aqueous system, sonochemical degradation of organic compounds occurs in the three regions indicated in inside and interface of the bubble and in bulk solution. The region I locates inside the collapsing cavitation bubble, and the adiabatically compressing generates high temperatures and high pressures existing here. Since the inside bubble

contains water and volatile substances, pyrolysis leads to the formation of radicals of OH and atomic substances. At the interface region II for the immediate surroundings region between the cavitation bubble and the bulk solution, pyrolysis of solutes like aromatic and organic halogen compounds occurs with the abundant local hydroxyl radicals. Region III locates the bulk solution at ambient temperature, permitting the reactions between solutes and the hydroxyl radicals, when they escape from the bubble. Further, on the collapse, shock wave, shearing stress, and jet flow are also generated and cause physical effects of ultrasound in the region III. Aromatic compounds and organic halogen compounds in water are mainly degraded by direct pyrolysis in regions I and II and/or by reactions with radicals in region II. On the other hand, polysaccharides and microorganisms are degraded by physical effects in region III in addition to the chemical effects in regions II and III.

Generally, ultrasonic power was evaluated by calorimetry [3]. In the chemical effect, the evaluation on the chemical species is conducted using potassium iodide [4], coumarin [5], terephthalate [6], and others. On the other hand, since it is hard to evaluate the physical effects, aluminum foil erosion has been used for qualitative estimation. Recently, however, aqueous viscosity changes by polymer degradation [7] is proposed for the evaluation of physical effects.

This review introduces recent works on the sonochemical degradation of organic substances using active bubbles, which are produced in ultrasonic process. Organic substances are classified into aromatic compounds, organic halogen compounds, polysaccharides, and microorganisms. Important results and degradation mechanisms are explained to demonstrate the effectiveness of the ultrasonic method.

Researches in sonochemical green process in Asia

Asian researchers actively study degradation of organic substances in aqueous solutions by ultrasound. In Japan, sonochemical green process have been widely investigated from 1990s [8]. Since entering 2000s, Korean researchers started to report many papers [9]. Recently, we can frequently read new papers written by Chinese researchers and recent works in Malaysia, India, Saudi Arabia, and Iran are also introduced in this review.

Aromatic compounds and organic halogen compounds

Many of aromatic compounds and organic halogen compounds in water are effectively degraded by ultrasonic irradiation. In recent trends of sonochemical degradation of aromatic compounds, high toxic chemicals and coloring substances are used. Table 1 lists recent examples of the sonochemical degradation of aromatic compounds and organic halogen compounds.

Benzophenone-3 [10], bisphenol A [11,12], di(2-ethylhexyl) phthalate [13], and 2-phenoxyethanol [14] as classified in endocrine-disrupting compounds were degraded sonochemically at various frequencies. Vega et al. [10] used ultrasound from 215 to 1134 kHz at the same ultrasonic power measured by calorimetry and reported maximum degradation rates at 574 kHz, increasing with the increased ultrasonic power. The degradation kinetics obeyed well with a pseudo-first-order model and benzophenone-3 was degraded at the bubble/liquid interphase. Bisphenol A was more rapidly decomposed at 400 kHz than at 200 kHz [11], obeying pseudo-first-order kinetics in the degradation rate. They indicated that the degradation reaction depended on the concentration and pH. At higher concentrations, the experimental results were applied to Langmuir–Hinshelwood mechanism. At low pH and a bulk temperature of 40 °C, interestingly, bisphenol A degradation was enhanced. Both the hydroxylation of the benzene ring and the attack on the connecting carbon were found to be the dominating mechanisms of bisphenol A degradation, and the latter mechanism proceeded more

significantly.

In carcinogens like 3,3'-diaminobenzidine [15], 5-methylbenzotriazole [16], and ethyl paraben [17], the degradation of 3,3'-diaminobenzidine was ultrasonicated at 43, 141, and 500 kHz reacting in 500 kHz for the optimum [15] and 5-methylbenzotriazole was effectively degraded at 640 kHz ultrasound in oxygen-saturated aqueous solution [16]. The decomposition followed pseudo-first-order kinetics, and the decomposition rate dropped significantly in the presence of tertiary butyl alcohol using as a hydroxyl radical scavenger. This indicated that the decomposition mechanism was proposed based on the reactions of hydroxyl radical substitution to the benzene ring, hydrogen atom abstraction by hydroxyl radical on benzylic carbon, and consequences of the autoxidation reactions. Triazole ring opening was explained by the high-temperature conditions in the vicinity of cavitation bubbles. Uddin and Okitsu [18] examined the effects of sodium sulfate and sodium chloride on the sonochemical degradation of 1,4 benzoquinone and hydroquinone at 200 kHz. The highest degradation rate was obtained in the presence of 0.443 M sodium sulfate for 1,4 benzoquinone, and the degradation rate of 1,4-benzoquinone was 10.6 times that of hydroquinone. On the other hand, the addition of sodium chloride was little in the effect on the degradation of hydroquinone, whereas negative effects were observed for the degradation of 1,4-benzoquinone.

Other harmful chemicals such as fungicides [19], pesticides [20], medicines [21-23], and polycyclic aromatic hydrocarbons [24-26] were also degraded by ultrasonic cavitation. Diphenhydramine, the first-generation antihistamine, was rapidly degraded by ultrasound at 640 kHz [21]. Further, similar to other chemicals, the degradation rate of diphenhydramine was also fitted to pseudo-first order kinetics [10,11]. Dibenzothiophene, a polycyclic aromatic hydrocarbon, in deionized water was effectively degraded at 352 kHz [24]. When natural water or seawater was used instead of deionized water, there was no noticeable decreases in the degradation rate.

There were reported several dyes [27-29] and pigments [30-32] in water treated by ultrasonic

irradiation. Toluidine blue in water was degraded at 1700 kHz [27]. For a higher initial concentration, interestingly, the degradation rate increased significantly with increasing liquid temperature up to 50 °C and decreased thereafter. The effect of ultrasonication on the removal of phycocyanin from water at 120-800 kHz was investigated [30], and 200 kHz was found to be optimum.

On the other hand, organic halogen compounds were used as surfactants [33,34], antiseptic agents [35], and pesticides [36,37]. Shende et al. [33] investigated the sonochemical degradation kinetics of nonvolatile surfactant perfluorooctanoic acid and perfluorooctane sulfonic acid. By developing a Michaelis-Menten-type kinetic model, they empirically estimated the concentration of active cavity sites during sonochemical reactions. Ultrasonic degradation of triclosan was studied at different frequencies in the same ultrasonic power, resulting in the optimum frequency at 574 kHz [35]. Triclosan degradation occurred over the bubble surface. The observation of the degradation rates indicated effect of the triclosan bulk concentration, the rate of generation and recombination of radicals, and the reaction with hydroxyl radicals and the reactant.

Polysaccharides and microorganisms

Recently, polysaccharides have been often used in food processing. Polysaccharides and microorganisms in water are effectively degraded by the physical and chemical effects occurring in regions II and III, as shown in Fig. 1. In the case of polysaccharides, the scission of polymer chains mainly occurs because of shear force and radicals under ultrasonic medium. Polysaccharides are classified in cellulose [38,39], foods [40-43], and others [44-48] for ultrasonic degradation.

Kojima et al. [38] degraded cellulose, when ultrasounds at 20 and 500 kHz were used. The viscosity decreased with increasing sonication time. The degrees of the polymerization decreased from 230 to

150 after 30 min of sonication at 20 kHz or after 120 min of sonication at 500 kHz. Importantly, cellulose nearly retained its crystallinity after sonication. Yan et al. [39] sonicated cellulose in water using an ultrasonic bath at 53 kHz and indicated that ultrasound decomposed the β -glycosidic linkage and hydrogen bonds in cellulose.

With regard to foods, the degradation of polysaccharides in konjac [40,41], mushrooms [42], and sweet potato [43] was investigated. The ultrasonic treatments for konjac glucomannan in water caused dramatic reduction in apparent aqueous viscosity, average molecular weight, and aggregated particle size [40]. In the same way as cellulose, konjac glucomannan had less damage to the primary molecular structure under ultrasonic treatment at 20 kHz with 700 W. The effect of ultrasonic treatment on the physical properties and antioxidant activity of the polysaccharide in the mushroom *Auricularia auricula* was evaluated at 20 kHz with 500 W [42]. It was noted that ultrasonic waves destroyed the helical structure and changed the monosaccharide proportion in the mushroom. Interestingly, the antioxidant activity of the *Auricularia auricula* polysaccharide [42] and potato pectin [43] increased significantly after the degradation.

Other polysaccharides such as dextran [44, 45], curdlan [46], hyaluronic acid [47], and gum [48] were degraded by ultrasonication. Koda et al. [44] effectively degraded methyl cellulose, pullulan, dextran, and poly(ethylene oxide) in aqueous solution at 500 kHz. Pu et al. [45] performed the sonochemical degradation of six dextrans in water at 20 kHz with 600 W. In all samples, ultrasonic treatment reduced the molecular weight and polydispersity of dextran. Significant such macromolecule degradation was observed initially, and particularly for their samples in the initial molecular weight. It was indicated that a midpoint scission model was suitable to describing their understanding to the results.

Microorganisms were ultrasonically disinfected in fish aquaculture [49] and vegetable storage [50], and treated in sewage sludge [51,52]. In zooplankton and algae samples with ultrasound waves at 850

kHz, effectiveness was seen in disinfection of eukaryotic organisms even at low power in aquaculture system [49]. Sewage sludge was a byproduct produced at municipal wastewater treatment plant as irradiated by ultrasound waves at 20 kHz [51]. The waves destroyed the sludge floc structure and cell walls, released intracellular organic matter, and accelerated the hydrolysis process. Importantly, ultrasound irradiation improved the sedimentation and dewatering performance of sludge. Moreover, ultrasonication also promoted coagulation and improved the efficiency of the anaerobic digestion process and the final biogas production.

Conclusion

The recent experiments and results on the sonochemical degradation of aromatic compounds and organic halogen compounds in water have led to three main findings. The first finding is that the optimum ultrasonic frequencies for such degradation range between 200 and 600 kHz. These frequencies are known to have high chemical effects [4]. The second finding is that ultrasonic power measurement by calorimetry to discuss sonochemical effects at various frequencies is becoming popular. Ultrasonic power in the sample can be directly determined by calorimetry [3]. In addition, diagnostic methods [4-6] for reactive chemical species formed by the chemical effect were reviewed in this paper. The third finding is that most reactions follow pseudo-first-order kinetics and that some reactions at high concentrations change to follow zero order kinetics.

In the case of sonochemical degradation of polysaccharides, two observations have been made recently. The first is that the optimum ultrasonic frequencies are often below 100 kHz by strong physical effects [7]. The second is that ultrasonic irradiation decreases the molecular weight via chain scission but does not affect the primary structure of the polysaccharide. In microorganism, however, the optimum ultrasonic frequencies depend on the features of the microorganism. Those are degraded

by the chemical or physical effects of ultrasonic cavitation, or both. Since ultrasonic treatment has several advantages, this method is expected to develop in future as a new green process and technology.

Conflict of interest statement

Nothing declared.

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In this paper, sonochemical treatment for municipal sludge was investigated. Ultrasonicated sludge were improved the sedimentation and dewatering performance. Moreover, ultrasound can also improve the activity of excess sludge, and the efficiency of anaerobic digestion process and the final biogas production. These results indicate that sonochemistry is effective for municipal sludge treatment.

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Figure captions

Figure 1 Sonochemical degradation: Regions, mechanisms, and organic substances

Table captions

Table 1 Recent examples of sonochemical degradation of aromatic compounds and organic halogen compounds

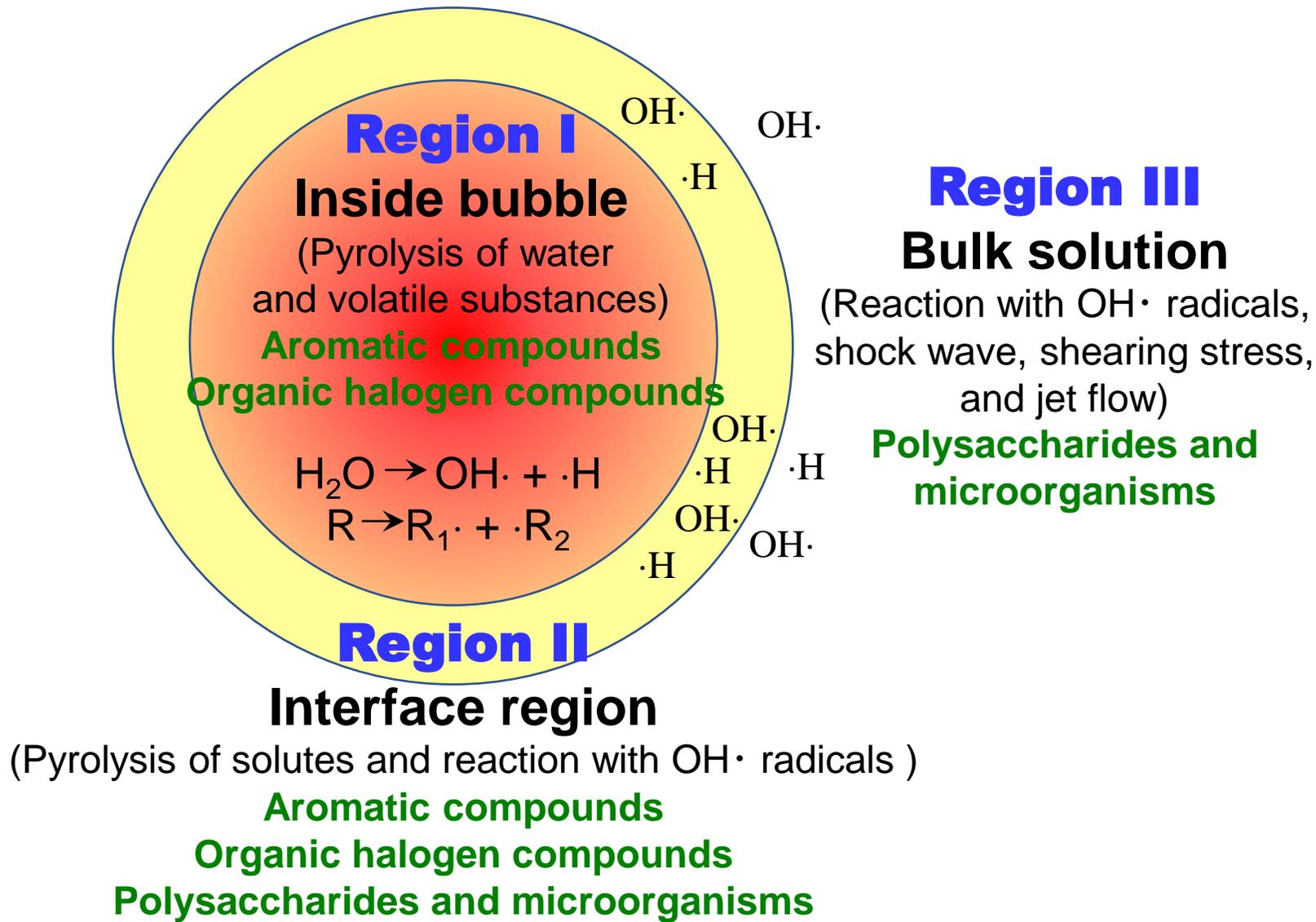


Fig.1 Yasuda

Table 1 Recent examples of sonochemical degradation of aromatic compounds and organic halogen compounds

Category	Chemical substances	Frequency (kHz)	Ultrasonic power	Temperature (°C)	Volume (ml)	Concentration	Ref.
Endocrine disrupting compounds	Benzophenone-3	215, 373, 574, 856, 1134	40-200 W/L (Calorimetry)	25	300	2-22 μ M	[10]
Endocrine disrupting compounds	Bisphenol A	200, 400	100 W (Electric power)	10-60	250-1250	5-200 μ M	[11]
Carcinogens	3,3'-Diaminobenzidine	43, 141, 500	10-100 W (Electric power)	-	200	0.1 mM	[15]
Carcinogens	5-Methylbenzotriazole	640	500 W (Electric power)	-	500	7.5-210 μ M	[16]
Harmful materials	1,4-Benzoquinone, hydroquinone	200	15 W (Calorimetry)	27	60	0.45 mM	[18]
Medicines	Diphenhydramine	640	396 W (Electric power)	10	500	2.8-160 μ M	[21]
Polycyclic aromatic hydrocarbons	Dibenzothiophene, bisphenol A	352	80 W (Electric power)	21	300	0.045-4.5 μ M	[24]
Dyes	Toluidine blue	1700	15 W (Calorimetry)	25-70	100	0.5-20 mg/L	[27]
Pigments	Phycocyanin	120, 200, 400, 800	60-100 W (Electric power)	-	100	4.75 mg/L	[30]
Surfactants	Perfluorooctanoic acid, perfluorooctane, sulfonic acid	575	77 W/L (Calorimetry)	21	200	0.2-45.45 μ M	[33]
Antiseptic agents	Triclosan	20, 215, 373, 574, 856, 1134	40-200 W/L (Calorimetry)	25	250, 300	1.7-11 μ M	[35]