Sonocatalytic determination of acoustic power for degradation of Diethyl phthalate

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1. Introduction

Diethyl phthalate (DEP) was one of the most frequently identified phthalates with high water solubility and short-chain in diverse environmental samples including surface water, drinking water, and sea water and so on. DEP was difficult to degrade biologically and photo-chemically [1].

However, few reports on DEP removal by ultrasound (US) could be found for degradation effectively. These processes were successfully applied for degradation of aliphatic, aromatic, polycyclic aromatic and halogenated hydrocarbons, azo dyes and for some pesticides compound as well [6].

Recent observations have indicated that TiO$_2$ particles can enhance the oxidizing power of ultrasound even in the absence of ultraviolet irradiation. Although the application of heterogeneous catalysts in an ultrasonic system has been reported previously, TiO$_2$ was found to have a higher oxidizing power and a specific mode of action in this system. The presence of a heterogeneous catalyst seems to increase the rate of formation of cavitation bubbles by providing additional nuclei site [7], which increase the pyrolysis of H$_2$O molecules and formation of OH radical. Furthermore, it is known that the flushes of single bubble sonoluminescence(SBSL), and sonoluminescence caused by bubble implosion may induce the excitation of TiO$_2$ in the ultrasonic system[9].

The aim of this study is to determine the kinetics of sonocatalytic degradation of Diethyl phthalate using frequency and power.

2. Experiment

DEP obtained from Sigma Aldrich (USA, purity $>$ 99%), characteristics of DEP are shown in the table. This table indicates that DEP is non-volatile compound.

The experiments were performed in a cup horn Type reactor which was surrounded by cylindrical water jacket, a constant temperature (16$\pm$2 °C) was maintained, Fig.1 shows a schematic diagram of the reactor. Ultrasonic waves were emitted from a transducer with ultrasonic controller (Miraes Ultrasonic Tech., Korea) .The frequency of 500 and 35 kHz were used and the electric power of ultrasound was fixed at 8.9, 26.1, 41.9 and 58.9 W.

The initial concentration of DEP was 0.2 Mm and total volume of aqueous phase was 500 ml. Samples were taken from the reactor for analysis of DEP at predetermined intervals (i.e. 0, 15, 30, 60 and 90 min) The concentrations of DEP was determined by using GC-MSD.

Table1. Characteristics of DEP

<table>
<thead>
<tr>
<th>Molecular formula</th>
<th>C$<em>{12}$H$</em>{14}$O$_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molecular weight (g/mol)</td>
<td>222.24</td>
</tr>
<tr>
<td>Water solubility at 25 °C (mg/L)</td>
<td>1080</td>
</tr>
<tr>
<td>Log K$_{ow}$</td>
<td>2.42</td>
</tr>
<tr>
<td>Henry’s law constant (atm-m$^3$/mole)</td>
<td>6.10$\times$10$^{-7}$</td>
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Fig. 1 Schematic diagram of the experimental apparatus for sonocatalytic process.

3. Results and Discussion

Fig. 2 shows the degradations of DEP by sonolysis and sonocatalysis at 35 kHz. The pseudo first order kinetic constants for degradation of DEP by sonocatalysis were 1.9$\times$10$^{-3}$, 3$\times$10$^{-3}$, 4.3$\times$10$^{-3}$, 6$\times$10$^{-3}$ min$^{-1}$ at 8.9, 26.1, 41.9, 58.9 W respectively. And the combined effect was highest at high power in sonocatalysis.

In this study, the faster degradation of DEP with sonocatalysis was much efficient as compared...
to sonolyis was believed to be due to the higher efficiency for the production OH radical produced by bubble collapse[8].

The sonocatalytic degradation was higher at high power and frequency of 500 kHz as shown in Fig. 3. The degradation rates were 9.3×10⁻³, 1.14×10⁻², 1.52×10⁻², 2.23×10⁻² min⁻¹ respectively. That means the rate of degradation at 500 kHz was 3-5 times more efficient than lower frequency. The mineralization of DEP was highest at US+TiO₂ at 500 kHz.

The beneficial effect of degradation rate was believed to be due to increase cavitation activity occurring at higher levels of power. As power increased, the number of collapsing cavities also increased, thus leading to enhanced degradation rates [7]. The rate of degradation increased with increasing power and addition of TiO₂ catalyst at high frequency.

This was presumably so since the amount of energy transmitted per unit volume increased with increasing power in presence of catalyst.

4. Conclusion

The kinetics of sonocatalytic process for degradation of DEP according to power and frequency were studied. According to previous studies, the faster degradation of DEP was believed to be due to the higher efficiency for the production OH radical as well as to the ultrasonic physical (bubble collapse and mixing) and TiO₂ (catalyst) processes[6]. For the range of experimental conditions, the rate constant of DEP degradation did indeed appear to vary linearly with high power and high frequency.

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References