Disintegration of sewage sludge with bifrequency ultrasonic treatment
Jianguo Jiang, Shihui Yang, Maozhe Chen and Qunfang Zhang

ABSTRACT
A bifrequency ultrasonic generating trough was used to improve disintegration of sewage sludge from thickener tanks. Ultrasonic operating parameters such as frequency, power, and time were optimized. Ultrasonic treatment successfully disintegrated the floc structure of sewage sludge and promoted the release of organic matter and metal ions such as Ca$^{2+}$ and Mg$^{2+}$. After ultrasonic treatment, soluble chemical oxygen demand (SCOD) in the sludge solution increased 11.28–49.06%, while the concentrations of Ca$^{2+}$ and Mg$^{2+}$ rose 5.5–25.0% and 2.7–19.0%, respectively. The ultrasonic frequency of 25 kHz was most effective in disintegrating sludge, and high energy densities and longer treatment time increased SCOD and metal ion release. The optimal parameters that promoted the increase SCOD were 25 kHz, 75 W/L and 60 s. Under these conditions, double-frequency ultrasonication was more effective than single-frequency ultrasonication.

Key words | bifrequency, disintegration, floc structure, metal ion, SCOD, ultrasound

INTRODUCTION
Sewage sludge is the main solid waste produced in wastewater treatment processes (Aparicio et al. 2009). The cost of sludge treatment represents 25–40% of the total disposal cost for the sewage treatment plant (Fan & Li 2006). Over the last three decades, treatment and disposal of sewage sludge have become increasingly important (Mikkelsen & Keiding 2002; Carballa et al. 2006; Parravicini et al. 2006; Samaras et al. 2008). Sludge is comprised of suspended flocs that are composed of dispersed microbial bacteria bridged by extracellular polymeric substances (EPS), cations (e.g. Ca$^{2+}$), and other cell granules (Houghton & Stephenson 2002). Anaerobic digestion (AD) is a widely used method for sewage sludge stabilization technology (Lu et al. 2008). However, the sludge mesophilic digestion technology widely used in China has several disadvantages such as low reaction speeds, long retention times, and use of large-volume reactors, complex operation, and low methane content. Therefore, sludge digestion tanks in sewage treatment plants in China have been built but are not efficient for sludge digestion. Because the cell walls of microbes are relatively resistant to biodegradation, the hydrolysis reaction of the microbial cell walls appears to be the speed-limiting step of AD (Pons et al. 2008).

Some novel pretreatment processes, such as ultrasound (Grönroos et al. 2005; Krupp et al. 2005; Yin et al. 2006; Na et al. 2007), acid (Neyens et al. 2005), ball mill (Nah et al. 2000), hydrothermal (Catallo & Comeaux 2008), and electrochemical (Glendinning et al. 2007) treatment, had been proved efficient on the improvement of anaerobic digestion. Ultrasonic sound waves can produce a series of extreme conditions in liquids, including the discharge of electricity, a momentary increase in temperature by thousands of degrees, pressure increases of hundreds of atmospheres (Tiehm et al. 2001), and hyper-velocity jet flow. Ultrasonic waves transmit in solutions and produce unidirectional forces simultaneously to accelerate the transfer and diffusion of substances (Yu et al. 2007).
This can be used to replace mechanical stirring or to remove substances from surfaces, and can be described as the mechanical effect of ultrasonic waves. Conversely, ultrasonic waves transmitted longitudinally in solution make the liquid alternate between compression and expansion phases. Small bubbles existing in the liquid expand rapidly during the expansion phase and then encounter sudden adiabatic compression during the compression phase of the wave, promoting temperature and pressure increases and the release of energy. Furthermore, during this cycle a new micronucleus is produced by cavitation (Feng & Huang 1993). When sludge is treated with ultrasonic waves, the micro-bubbles in the sludge crack, and the instantaneous high pressure and temperature break the bonding forces within the sludge floc, thereby disintegrating the sludge.

Several studies have addressed the use of ultrasonic treatment for disintegration of sewage sludge domestically and abroad, but these studies utilized single-frequency ultrasonic treatment for sludge disintegration. Few studies have considered a change of ultrasonic frequency and ultrasonic generator arrangements. In the present investigation, a bifrequency ultrasonic generating trough was designed to disintegrate sewage sludge, and the influence of ultrasonic treatment on sludge disintegration was investigated using ultrasonic parameters such as frequency, electrical power density, treatment time, and generator arrangement mode (single side or double sides). These studies provide basic data for the application of ultrasonic technology in the field of sludge treatment.

**MATERIALS AND METHODS**

**Materials**

The sludge used in these experiments was sampled from a sludge thickener pool at a wastewater treatment plant in Beijing. The excess sludge treatment included thickening, anaerobic digestion, and dewatering. The basic characteristics of the sludge sample from the thickener pool are shown in Table 1.

<table>
<thead>
<tr>
<th>pH</th>
<th>Moisture content (%)</th>
<th>SCOD (mg L(^{-1}))</th>
<th>SS (mg L(^{-1}))</th>
<th>VSS (mg L(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.84</td>
<td>97.73</td>
<td>728.92</td>
<td>5,786</td>
<td>4,293</td>
</tr>
</tbody>
</table>

**Equipment**

At present, no sophisticated ultrasonic equipment for sludge treatment exists in the Chinese market. In laboratories, sludge ultrasonic research depends on the cell disruption instrument, which may have problems such as maldistribution of energy caused by the monopolar probe and difficulties in the adjustment of operation parameters. Based on test requirements and sludge characteristics, we designed and produced a special bifrequency ultrasonic generating trough (Figure 1).

This ultrasonic generator has the following characteristics:

1. Easily adjustable operation parameters: the frequency covers 19, 25, 40, and 80 kHz; the electrical power of every side of the generator covers 200 W to 500 W; the treatment time covers 0 to 60 min.

2. Bifrequency: the generators above and underneath the trough can be controlled so that single-frequency or double-frequency ultrasonic waves can be generated to treat sludge.

3. Uniformly distributed ultrasonic treatment: the size of the surface of the ultrasonic transducer is 510 mm \(\times\) 260 mm, under which 10 evenly distributed ultrasonic detectors are located. Figure 2 shows the acoustic power distribution, determined by the dyeing method (Qiping et al. 2000; Minglei et al. 2004), of a single ultrasonic detector and the equipment used in this experiment. A cavitation bubble is characterized by tiny points, showing more uniformity in Figure 2(b).
Wide application for media: as shown in Figure 1, the protective cover is filled with water to avoid the burning of the transducer when little water exists in the media. Hence, the system can be used for treatment of sewage sludge before and after dewatering.

Method

Sludge (4 L) was placed in the trough and exposed to different ultrasonic fields by adjusting the frequency, electrical power density, and treatment time. All of the parameters are conveniently adjusted by electric control system through digital display control system (Figure 1). Firstly, electrical power density and treatment time were fixed; the optimal frequency of the equipment was selected by comparing the effects of treatment. Then, the optimal frequency was fixed; the optimal electrical power density and optimal treatment time were selected respectively. In addition, the frequency mode was studied.

The characteristics of the sludge before and after treatment were examined. The sludge microstructure was observed by electron microscopy, soluble chemical oxygen demand (SCOD) was determined by the potassium dichromate method, and the concentrations of Ca\(^{2+}\) and Mg\(^{2+}\) were determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES) after pretreatments with acid and filtration.

The sludge sampling and ultrasonic treatments were conducted for 12 hours, and the characteristics of non-treated sludge and treated sludge were synchronously analyzed in 12 hours after sampling. If the analysis of characteristics could not be completed in 12 hours, the sample and the treated sample were pretreated and stored in a refrigerator at 4°C to prevent sludge metamorphism and filamentous bulking, even so, the analysis of characteristics were completed in no more than 24 hours after sampling.

RESULTS AND ANALYSIS

Changes in the sludge floc structure

Ultrasonic energy spreads throughout the sludge with alternate strengths and breaks the tiny bubbles in the solution. The consequent shear stress and transient high temperature and pressure damage the bonding forces among the sludge flocs, changing the floc structure (Guangmin et al. 2003). Figure 3 shows the microstructure of sludge flocs before and after ultrasonic treatment (25 kHz, 100 W/L, 60 s).

Figure 3(a) shows the morphology of the raw sludge. Sludge flocs gather to form a floc group. A large amount of lacuna (the void surrounded with floc) provides a stable place for water in the sludge. Figure 3(b) shows the morphology of the treated sludge. In comparison with Figure 3(a), ultrasonic treatment disintegrated the floc joints in the sludge and significantly decreased the degree of floc aggregation. The destructive effect of ultrasonic treatment on sludge is more visible under 40× magnification (Figure 3c and d). With ultrasonic treatment, the particle size of sludge flocs decreased and the structure of flocs became looser. These results confirm that ultrasonic treatment of sludge can disrupt flocs, change their structure, decrease particle size, and affect the compactness of the sludge floc.

Change of sludge SCOD

Ultrasonic cavitation in sludge causes sludge flocs to break and disrupts cells within the sludge. As a result, the organic matter parcelled among the sludge flocs and the extracellular polymer on the surface of cells enters the water phase and increases the SCOD of the sludge solution (Chu et al. 2001).

(1) Ultrasonic frequency

The ultrasonic frequency determines the degree of mechanical and chemical disruption. The sludge in this study
was treated with different ultrasonic frequencies to determine the optimal frequency for sludge treatment. Modulation of ultrasonic parameters such as frequency, power, time, and ultrasonic mode demonstrated that frequency was the most important parameter for the disintegration of sewage sludge. The treatment of sludge at any power and at several time points had nearly identical effects on the sludge (as estimated by SCOD) within several frequency ranges. Thus, the ultrasonic treatment parameters of 75 W/L and 60 s were selected for analyzing the influence of frequency on the sludge SCOD (Figure 4).

Figure 4 indicates that the degree of SCOD in sludge differs with ultrasonic frequency. The treatment of sludge at ultrasonic frequencies of 19, 25, 40, and 80 kHz promoted 36, 49, 21, and 13% increases in SCOD, respectively, compared with SCOD in raw sludge. Hence, the treatment of raw sludge at 25 kHz frequency appeared to be optimal for promoting the digestion of organic material.

The relationship between cavitation and ultrasonic frequency underlies the effects of ultrasonic frequency on disintegration. According to the principles of sonochemistry, a close relationship exists between the ultrasonic cavitation threshold in sludge liquid and the ultrasonic frequency. When the resonant frequency of ultrasonic
waves is consistent with the most probable distribution of nucleus size in sludge liquid, the maximum sonochemical yield is obtained, and any frequency that deviates from the resonant frequency causes a decrease in sonochemical yield (Zhao et al. 1994; Huang et al. 1995; Feng et al. 1997). We conjecture that in our experiment, the ultrasonic frequency of 25 kHz was consistent with the most probable distribution of nucleus size in the sludge and therefore resulted in the highest level of sludge disintegration.

(2) Ultrasonic energy

Ultrasonic energy determines the amount of cavitation bubbles and the value of shear stress produced in the sludge solution. In this experiment, the influence of electrical power density and treatment time on sludge SCOD was investigated. The sludge was treated at 25 kHz, and the overall ultrasonic energy was adjusted by changing the input electrical power density and treatment time. The resultant changes in sludge SCOD are indicated in Figure 5.

When the treatment time was 10 s, the sludge SCOD increased 10–11% irrespective of electrical power density (Figure 5). With a treatment time of 15 s, SCOD gradually increased to 816, 857, 878, and 941 mg/L when treated with increasing power densities of 50, 75, 100, and 125 W/L, respectively. Treatment with a electrical power density of 125 W/L achieved a 29% increase in SCOD, which was 2.4-fold the SCOD increase at 50 W/L. Treatment for 20 s resulted in a similar trend for the electrical power density effect on sludge SCOD, but the overall SCOD increase was larger because of the extended treatment time. With a 20 s treatment time, the sludge SCOD reached 858, 965, 932, and 964 mg/L when treated with power densities of 50, 75, 100, and 125 W/L. With a 30 s treatment time, there were no obvious differences in SCOD at higher power densities. However the treatment of sludge for 60 s caused SCOD to increase to 1,086, 989, and 987 mg/L with power densities of 75, 100, and 125 W/L, respectively. Thus, the electrical power density of 75 W/L promoted the highest SCOD in this experiment.

In summary, prolonged treatment times increased the sludge SCOD. According to the principles of sonochemistry, when the treatment time is invariable and the power increases, the cavitation yield shows a non-linear increase that is fast at the beginning and ultimately saturates (Lorimer 1990). We observed that with a long treatment time, SCOD increased as the electrical power density increased, but the threshold electrical power density was 100 W/L. The optimal parameters in this investigation were 25 kHz, 75 W/L and 60 s.

Changes in metal ion concentrations

Sludge floc is an aggregate of many single cells and is held together by extracellular polymeric substances (EPS). Inorganic particles such as Ca\(^{2+}\) and Mg\(^{2+}\) are needed to act as bridging ions (Novak et al. 1977; Zhou et al. 2004). At beginning, the solution concentration of Ca\(^{2+}\) and Mg\(^{2+}\) increased quickly as a result of the release of the Ca\(^{2+}\) and Mg\(^{2+}\) into the aqueous phase, then, the particle size of sludge became smaller during disintegration which enlarge the Specific Surface Area and accelerate the absorption, and the surface charge was negative, which could adsorb Ca\(^{2+}\) and Mg\(^{2+}\) (Wang et al. 2006). Therefore, the concentration of Ca\(^{2+}\) and Mg\(^{2+}\) went down.

Accordingly, the changes in metal ion concentrations in the sludge solution under different treatment conditions were investigated, and the results are shown in Figure 6.

(1) Ultrasonic frequency

When the electrical power density was 50 W/L, the Ca\(^{2+}\) and Mg\(^{2+}\) soluble concentrations in sludge treated at the ultrasonic frequency of 40 kHz were higher than those in sludge treated at 25 kHz. Furthermore, 40 kHz ultrasonic
treatment increased the Mg\(^{2+}\) concentration 14–19\%, in contrast to the small change with the 25 kHz treatment. Increasing the electrical power density to 75 W/L resulted in a small difference in the Ca\(^{2+}\) and Mg\(^{2+}\) concentrations at 25 and 40 kHz. The 25 kHz treatment gave a maximum Ca\(^{2+}\) concentration of 1,097 mg/L when the treatment time was 15 s, and the 40 kHz treatment resulted in a maximum of 919 mg/L when the treatment time was 20 s. With a short treatment time (<15 s), the 25 kHz ultrasonic treatment was better than the 40 kHz treatment for the release of metal ions, whereas with greater durations of treatment (>20 s), the 40 kHz treatment showed a greater ability to release metal ions. Increasing the electrical power density to 100 W/L at 40 kHz resulted in a maximum Ca\(^{2+}\) soluble concentration of 1,098 mg/L with a 15 s treatment; this treatment appeared to be more effective than treatment at 25 kHz for 20 s, which gave a maximum Ca\(^{2+}\) concentration of 1,097 mg/L. When the electrical power density was 125 W/L, the results were opposite those with 100 W/L. Almost no difference in Mg\(^{2+}\) release was observed between 100 and 125 W/L.

(2) Ultrasonic electrical power density

Increasing the electrical power density, which increases the input energy, had a significant effect on the release of Ca\(^{2+}\) and Mg\(^{2+}\) from the sludge solution. When the treatment time was 15 s, frequency was 25 kHz and the electrical power density was 125 W/L, the Ca\(^{2+}\) soluble concentration of treated sludge increased by 507 mg/L over that found in raw sludge. At 50 W/L, the increase was only 48 mg/L. For any given treatment time, an optimal electrical power density for the release of metal ions was observed.

(3) Ultrasonic treatment time

The concentration of metal ions in the solution increased with treatment time, but the rate of increase depended on the input power. With a small energy input, the metal...
ion concentration, especially the Mg$^{2+}$ concentration, increased slowly. When the electrical power density was greater than 75 W/L, treatment time affected the concentrations of both Ca$^{2+}$ and Mg$^{2+}$. At a electrical power density of 75 W/L, the Ca$^{2+}$ concentration increased to 1,079 mg/L and then decreased to 889 mg/L. At 100 W/L, the Ca$^{2+}$ concentration increased to 1,037 mg/L and then decreased to 928 mg/L. Under these conditions, the Mg$^{2+}$ concentration increased by 25% and was stable during long treatment times. These results suggest that 15–20 s is the optimal treatment time for the release of metal ions from sewage sludge.

To conclude, the parameters of frequency, electrical power density, and treatment time affected the metal ion concentration during ultrasonic treatment. The relationship between frequency and metal ion release differed with energy input. With less energy, ultrasonic treatment at 40 kHz accelerated the release of metal ions to a greater extent than treatment at 25 kHz. However, with more energy, the difference between 40 and 25 kHz decreased, and then an opposing trend was observed. Overall, increasing the electrical power density and treatment time accelerated the release of metal ions, but the release slowed with treatment times longer than 30 s.

**Effect of frequency mode on sludge disintegration**

According to the principles of sonochemistry, ultrasonic frequency affects the sonochemical yield. Thus, bifrequency and trifrequency treatments should give higher yields than single-frequency treatments. The effect of frequency on sludge disintegration was investigated using our bifrequency ultrasonic equipment, which could be adjusted to separately control the frequency both above and below the sludge. In this experiment, single frequency means that the ultrasonic energy was delivered from the bottom of the trough, and double frequency means that the ultrasonic energy originated from both the top and the bottom of the trough. The electrical power density of every side was 50 W/L, and the total electrical power density was 100 W/L.

**Effect on SCOD**

The SCOD values in the sludge treated with single and double frequencies are compared in Figure 7.

Without altering the frequency, electrical power density, or treatment time, these results indicate that bifrequency ultrasonic treatment is superior to single-frequency treatment for the disintegration of sludge. However, with a short treatment time, the disintegration did not differ between the two treatments. Conversely, when the treatment time was increased to 20, 30, or 60 s, the rate of SCOD increase with bifrequency treatment was 4.2, 15.4, or 23.5% greater, respectively, than the rate with single-frequency treatment.

**Effect on metal ions**

Metal ion release was similar between single-frequency and bifrequency treatments (Figure 8). The concentration of Ca$^{2+}$ initially increased rapidly and then decreased slowly; the concentration of Mg$^{2+}$ increased to a level that did not change further with time. The Ca$^{2+}$ concentration reached...
1,037 mg/L using the single-frequency protocol and 979 mg/L with the bifrequency protocol, followed by a decrease to 928 mg/L and 915 mg/L, respectively, with a treatment time of 20 s. The Mg²⁺ concentration reached a similar maximum with single-frequency (57 mg/L) and bifrequency treatments (60 mg/L) and thereafter remained constant for a 20 s treatment time.

These data indicate that bifrequency ultrasonic treatment, as compared with single-frequency treatment, affects sludge SCOD but does not augment metal ion release. Bifrequency treatment produced a higher chemical yield, probably owing to more cavitation bubbles and stronger shearing action; however, the electrical potential of the floc and the competition among ions of different valences appeared to restrict the adsorption reactions between metal ions and sludge floc.

CONCLUSIONS

(1) With ultrasonic treatment, the particle size of sludge floc decreased and the structure of the floc aggregate was destroyed.

(2) Ultrasonic treatment promoted the dissolution of organic substances in the sludge floc and increased SCOD of the sludge solution by 11–49%. Ultrasonic treatment at a frequency of 25 kHz was more useful than 19, 40, or 80 kHz for increasing SCOD in the sludge. Increasing the electrical power density and extending the treatment time also increased SCOD, with threshold values of 75 W/L and 30 s in this experiment.

(3) Ultrasonic treatment promoted the release of metal ions from the sludge; the concentration Ca²⁺ and Mg²⁺ increased by 6–25% and 3–19%, respectively, with ultrasonic treatment. The release of metal ions appeared to depend on the electrical power density. At low power, an ultrasonic frequency of 40 kHz accelerated the release of metal ions to a greater extent than treatment at 25 kHz; at higher powers, this difference between frequencies was lost, and a decreased efficacy of ion release was observed. Longer treatment times (> 30 s) were not more effective at releasing metal ions.

(4) Bifrequency ultrasonic treatment increased sludge SCOD to a greater extent than single-frequency treatment when all other parameters remained constant. However, there were no obvious differences in metal ion release between single and bifrequency treatments.

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