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Ultrasound coupled with Fenton oxidation pre-treatment of sludge to release organic carbon, nitrogen and phosphorus☆

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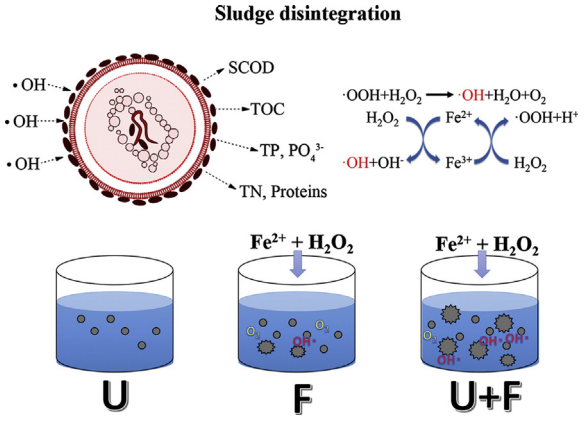
HIGHLIGHTS

- Combined ultrasound–Fenton pre-treatment was proposed for sludge disintegration.
- Ultrasound–Fenton significantly increased carbon, nitrogen and phosphorus release.
- Higher level of OH• was detected after combined disintegration than Fenton.

GRAPHICAL ABSTRACT



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ABSTRACT

We focused on the effects of ultrasound and Fenton reagent in ultrasonic coupling Fenton oxidation (U + F) pre-treatment processes on the disintegration of wastewater treatment plant sludge. The results demonstrated that U + F treatment could significantly increase soluble COD, TOC, total N, proteins, total P and PO_4^{3-} concentrations in sludge supernatant. This method was more effective than ultrasonic (U) or Fenton oxidation (F) treatment alone. U + F treatment increased the soluble COD by 2.1- and 1.4-fold compared with U and F alone, respectively. U + F treatment increased the total N and P by 1.7- and 2.2-fold, respectively, compared with F alone. After U + F treatment, sludge showed a considerably finer particle size and looser microstructure based on scanning electron microscopy, and the highest OH• signal intensity increased from 568.7 by F treatment to 1106.3 using electron spin resonance. This demonstrated that U + F treatment induces disintegration of sludge and release of organic carbon, nitrogen and phosphorus better.

☆ The English in this document has been checked by at least two professional editors, both native speakers of English. For a certificate, please see: http://www.textcheck.com/certificate/8amrcQ.

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Notations

COD	chemical oxygen demand
TOC	total organic carbon
TC	total carbon
TS	total solid
N	nitrogen
P	phosphorus
SE	standard error
SEM	scanning electron microscope
U	ultrasound treatment
F	Fenton oxidation treatment
U + F	ultrasonic coupling Fenton oxidation treatment
A2/O	anaerobic–anoxic–oxic
ICP-AES	inductively coupled plasma-atomic emission spectrometry
ESR	electron spin resonance
DMPO	5,5-dimethyl-1-pyrroline-N-oxide
OH•	hydroxyl radicals

1. Introduction

Sludge is the primary solid waste created during the treatment of municipal sewage. The total amount of domestic wastewater discharged in 2012 was 6.8×10^{10} tons, of which household wastewater contributed 4.6×10^{10} tons (Wen and Liu, 2013). Given that the average solid-containing rate of municipal sewage is 0.02%, sludge yield reached 6.8×10^{7} tons (moisture: 80%). Large amounts of activated sludge without proper disposal poses a threat to ecological systems (Rai et al., 2004). On the other hand, sludge is a potential resource because it contains large quantities of organic matter that can be converted into biogas through anaerobic digestion (Yen and Brune, 2007; Sosnowski et al., 2003). Sewage sludge contains considerable amounts of nutrients (Sinha et al., 2014; Tyagi and Lo, 2013; Zhou et al., 2008), especially phosphorus (0.5–0.7% total solid [TS]) and nitrogen (2.4–5.0% TS) (Metcalf et al., 1991). These nutrients exist mainly in proteinaceous and zoogloea forms (Tyagi and Lo, 2013). To recover N and P from sewage sludge, it is necessary to perform an N and P-solubilisation process to release phosphate into the supernatant. To recover nutrients, including N and P, the structure of sludge zoogloea must be broken down. Biopolymers are released following destruction of the cell structure. Various sludge disintegration methods have been investigated for pre-treatment; these methods disrupt cell walls, resulting in lysis of sludge cells (Appels et al., 2008). Possible pre-treatments include mechanical, thermal, chemical and biological methods. Recently, it has been reported that these pre-treatments including ultrasonic methods could overcome the rate-limiting step and so reduce the digestion time (Khanal et al., 2007). Organic matter was transferred from sludge flocs to the aqueous phase after ultrasonic pre-treatment, which improved the digestibility and dewaterability (Khanal et al., 2007). Fenton oxidation is commonly used to degrade organics and kill microorganisms during wastewater treatment. During Fenton oxidation, the sludge structure is destroyed and water and organics are released (Chen and Pignatello, 1997). This reagent also oxidises odorous substances, kills pathogenic bacteria and stabilises the sludge (Chen and Pignatello, 1997; Pham et al., 2010; He and Wei, 2010). Problems such as a relatively low efficiency and high energy consumption arise using only a single disintegration method, while some studies have demonstrated that the combination of various methods with different disintegration modes improves the disintegration effect in a synergistic manner. Kim (Kim et al., 2010) and Jin (Jin et al., 2009) evaluated alkali coupled with ultrasound as a pre-treatment for sludge disintegration. Kim (Kim et al., 2010) reported that alkali could reduce the energy consumption of the ultrasound method. Jin (Jin et al., 2009) found that the applying order of alkali and ultrasound treatment had a significant effect on the release of sludge COD. Xu (Xu et al., 2010) examined sludge pre-treatment using ultrasound coupled with ozone and showed that ultrasound improved the oxidative ability of ozone and significantly improved its utilisation ratio. Many disintegration methods are available, each of which has advantages and disadvantages. Single Fenton oxidation and ultrasound are most commonly used for wastewater treatment (Pham et al., 2010; Mohapatra et al., 2011). Herein, we report ultrasound coupled with Fenton oxidation pre-treatment of sludge to promote release of organic carbon, nitrogen and phosphorus. The technique combines the advantages of the mass transfer and cavitation effect of ultrasound and the strong oxidation function of Fenton reagent, resulting in improved disintegration of sludge. By comparing carbon, phosphorus and nitrogen release with the degree of sludge disintegration after U + F treatment, the mechanism of sludge disruption was investigated and discussed.

2. Material and methods

2.1. Excess sludge

Excess sludge was obtained from the Xiaojiahe Wastewater Treatment Plant in Beijing, China. Its sewage treatment capacity is 40,000 m^3 per day. The sewage treatment plant uses the A2/O processing system for municipal wastewater treatment, which is the most common method of treating sewage in most Chinese cities. The excess sludge was obtained from the secondary sedimentation tank, in which gravity thickening occurs following aerobic digestion. In order to prevent its deterioration, the sludge was stored in a refrigerator at 4 °C and the experiments had to be completed within 10 days. All the data presented in Table 1 and Figs. 1–6 of the manuscript are representative of at least three parallel experiments in which all samples were assayed in triplicate. The variance analysis has been done by using the standard deviation method and the error bars have been added in Table 1 and Figs. 1–4 of the manuscript.

2.2. Experimental set-up

A probe-type ultrasound generator was used in this paper. Ultrasound (20 kHz) was used according to previous reports (Jiang et al., 2009, 2014a,b; Tiehm et al., 2001).

2.2.1. Supernatant separation

The sludge samples was centrifugated at 5000 rpm for 5 min, then filtrated by a quantitative filter paper (0.53 mm, 120 μm).

2.2.2. Fenton reagent preparation

As reported in our previous study (Jiang et al., 2014a), the optimum dose of Fenton reagent for sludge disruption was prepared as follows: 2.0 g of $FeSO_4 \cdot 7H_2O$ were added to 1 L of sludge and stirred uniformly, after which 1.7 mL of 30% H_2O_2 was added to the system, in which the concentrations of Fe^{2+} and H_2O_2 were 0.4 and 0.50 g/L, respectively. A hydrochloric acid solution (18 wt.%) was used to adjust the pH to 3.

To investigate the effect of processing time on U, F and U + F, the ultrasonic energy density was fixed at 720 W/L. To investigate the effect of ultrasonic energy density, the ultrasonication duration was 20 min. For U + F, Fenton oxidation time was fixed at 20 min, followed by

Table 1
Characteristics of the excess sludge analysed in this study.

No.	pH	Moisture	COD	TOC	TC	Proteins	N	P
Units		Content%	mg O_2/L	mg/L	mg/L	mg/L	mg/L	mg/L
Mean	6.82	98.8	17868.4	89.3	147.7	767.9	98.8	268.8
SE	0.06	0.2	697.5	8.6	8.5	37.8	12.7	36.9