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Optimization of integrated ultrasonic-Fenton system for metal removal and dewatering of anaerobically digested sludge by Box-Behnken design

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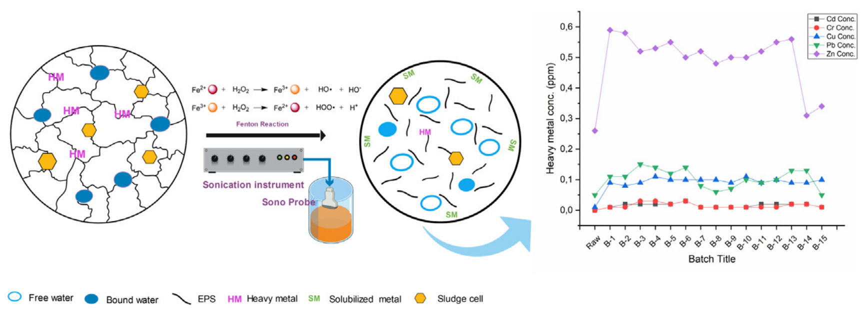
HIGHLIGHTS

- Fenton (Fe^{2+}/H2O2) and alternative Fenton (Fe^{2+}/NaOCl) systems were investigated.
- Dewaterability and metal leaching responses were optimized by Box-Behnken design.
- Ultra-sonicated Fenton significantly increased the metal removal from sludge.
- Optimal operating conditions were 36 mM of Fe^{2+}, 320 mM H_2O_2 and 30 min sonication.

GRAPHICAL ABSTRACT



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ABSTRACT

This study reveals the optimization of ultrasonic-Fenton process for the treatment of sludge taken from a local municipal wastewater treatment plant after anaerobic digestion. Box-Behnken design (BBD), a common approach of response surface methodology (RSM), was applied to evaluate and optimize the individual and interactive effects of three process variables, namely Fe^{2+} dose, H_2O_2 amount and sonication time for Fenton-ultrasonication method. Five dependent parameters including total organic carbon (TOC), extracellular polymeric substances (EPS), as LB-EPS and TB-EPS, and metals such as Zn and Cu were considered as the responses to investigate. According to the results of analysis of variances (ANOVA), five modelling equations are proposed that can be used to operate the design space with high regression coefficient R^2. Modelling results suggest that Fenton parameters, such as: H_2O_2 and Fe^{2+} dosage had the significant effects on the overall removal of TOC; whereas, sonication improved the metal removal from the sludge sample. Based on response surface methodology, best performance is achievable under the following conditions: 36 mM of Fe^{2+}, 320 mM H_2O_2 with 30 min of sonication respectively for all of the responses.

1. Introduction

Aerobic and anaerobic digestion are the most common functioning process for sludge generation in the wastewater treatment system and generates sludge at a high amount of 0.2–0.3 kg/m^3 treated water (Turovskiy and Mathai, 2005). For maintaining a continuous operation of wastewater treatment plants (WWTPs), periodic disposal of sludge from the system is needed; which is now considered as a major environmental concern. Moreover, sludge particles aggregate as a colloidal suspension which contains >90% water. Therefore, dewatering of sludge is

Abbrevatations: TOC, Total organic carbon; RSM, Response surface methodology; Fenton + US, Fenton + ultrasonication; BBD, Box-Behnken design; ICP-OES, Inductively coupled plasma-optical emission spectrometry; CCD, Central composite design; EPS, Extracellular polymeric substances; PP, Polypropylene; AOPs, Advanced oxidation processes; TSS, Total suspended solid; UV, Ultraviolet; WW, Wastewater; WWTPs, Wastewater treatment plants.

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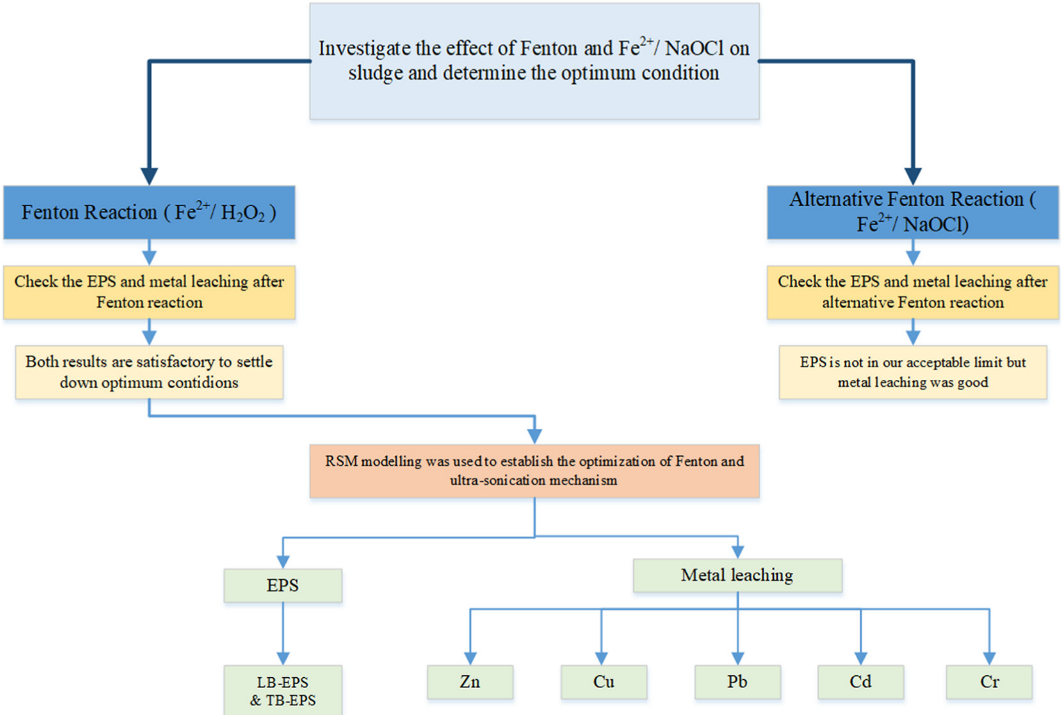

Fig. 1. Schematic diagram of the research work.

needed prior to disposal for the reduction of sludge handling costs as well as the transportation costs (Yu et al., 2016; Zhou et al., 2014a).

Dewatering of sewage sludge is a challenging endeavour because of their hydrated nature along with highly colloidal structure. In this regard, extracellular polymeric substances (EPS), mainly consisting of proteins and polysaccharides, have a substantial impact. Indeed, EPS can be divided into LB-EPS (loosely bound EPS) and TB-EPS (tightly bound EPS), and the LB-EPS has a significant effect on sludge dewaterability. In bacterial cell, EPS is attached to the surface and in activated sludge, this insoluble organic matter accounts for 50–90% of the total organic matter. Moreover, EPS has a strong water binding capability; which makes it difficult to remove the sludge floc's water by mechanical force (Hong et al., 2017; Qi et al., 2011).

Apart from dewatering, toxicity of the residual sludge should also be considered before disposal into the environment. In conventional water treatment systems, toxic metals tend to settle down in the residual sludge (Wong et al., 2006); which is often digested, incinerated, or deposited in landfills, and in some cases used as fertilizer in agricultural lands. All these processes can make a pathway for toxic metals to find their destination in the environment. In addition, the possibility to find pathogenic organisms in sludge, makes it potentially harmful to the environment and public health. Numerous conditioning methods including physical, biological, mechanical and chemical systems are used to treat sludge in WWTP (Wang et al., 2017). In aqueous medium, advanced oxidation processes (AOPs) are very promising and could be applied to degrade contaminated and biologically refractory organic pollutants. The key advantage of AOPs is the in situ generation of hydroxyl radicals (HO•), which can act as highly oxidizing agents in water (Cheng et al., 2016a; Laine and Cheng, 2007; Oturan and Aaron, 2014). Under optimum conditions, it is capable of converting a variety of organic pollutants into smaller molecular weighted compounds or even into water and carbon dioxide as well. Chemicals like ozone (Esplugas et al., 2007), hydrogen peroxide and Fe^{2+}/UV (del Moro et al., 2013) are often used in chemical oxidation techniques.

Among AOPs, Fenton technique is reported as an effective process in the detoxification of toxic and persistent organic pollutants in water, soil and sewage sludge due to its high degradation ability, short reaction time and simplicity in operation (Cheng et al., 2016b; He et al., 2015; Hu et al., 2018). The combination of hydrogen peroxide and ferrous ion is the main reagent for Fenton, leading to the formation of highly reactive hydroxyl radical ($HO^{•}$) along with the oxidation of Fe^{2+} to Fe^{3+} (Eq. 1) (Huang et al., 2017; Xiao et al., 2018; Zingaretti et al., 2018). Under the action of HO• radical, organic compounds (R-H or R) transform into the organic radicals (R• or HOR•) (Eqs. (2) & (3)), which further transforms

Table 1
Experimental range and levels of design of experiment.

Variables	Factors	Coded factor level		
		−1	0	+1
Fe^{2+} dose, mM	x_1	10	23	36
H_2O_2 dose, mM	x_2	100	230	360
Sonication time, min	x_3	10	20	30

Table 2
Regression co-efficients of model equations and their corresponding level of significance.

Y	x_1	x_2	x_3	x_{12}	x_{22}	x_{32}	$x_1 x_2$	$x_1 x_3$	$x_2 x_3$	Const.	R^2 (%)	R^2 (adj) (%)
TOC	7.19	26.42	−6.50			−9.13				9.75	74.59	68.06
LB-EPS	−2.867	2.701	−1.772		4.253	7.7	3.858	4.632		38.32	83.49	79.24
TB-EPS		14.33	−11.22	50.17	32.35	40.33	−18.91		17.48	−50.62	82.81	78.39
Zn	8.95	8.52		6.23		4.24		−8.78		100.08	81.67	76.96
Cu	175.7	−49.6	100.2	99.99	−99.7			−149.6		1500.7	81.07	76,20