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A rapid Fenton treatment technique for sewage sludge dewatering

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HIGHLIGHTS

- The Fenton reaction time was shortened to 5 min.
- Through optimization experiments, 96% SRF reduction efficiency was achieved.
- The composite conditioner acted both as an oxidant and a coagulant.
- The results were confirmed by high-pressure filtration experiments.

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ABSTRACT

The study aimed to develop a rapid and practical Fenton treatment technique for sludge dewatering. The results indicated that the Fenton reaction time could be shortened to 5 min under suitable conditions. In addition, an orthogonal experimental design method was used to optimize the rapid Fenton treatment process. The optimal conditions were as followings, pH 3, Fe^{2+} of 50 mg/g DS (dry solids), H_2O_2 of 30 mg/g DS and lime of 50 mg/g DS, at which the specific resistance to filtration (SRF) reduction efficiency of 96% was achieved. The dosage of Fe^{2+} was demonstrated to be the most significant factor affecting sludge dewaterability. Furthermore, the lowest extracellular polymeric substances (EPS) concentration in the sludge supernatant was measured under the optimal conditions, suggesting that Fenton's reagent acted both as an oxidant and as an effective coagulant for the sludge fragments. The morphology of the sludge cake exhibited a relatively regular and spongy block-like structure, which could maintain high permeability during high-pressure filtration. With the novel filter press achieved the squeezing pressure up to 40 MPa, the water content of sludge cake could be reduced to 55.1 ± 0.6%. Additionally, the economic assessment shows that the rapid Fenton treatment combined with high-pressure filtration can be a suitable technique for sewage sludge dewatering.

1. Introduction

Currently, the main method of sewage sludge treatment is conditioning with organic polymers before mechanical dewatering, which limited by its drawbacks, and achieved the sludge water content of 75–80% [1]. Previous studies have indicated that the high affinity for water of extracellular polymeric substances (EPS) confines the dewaterability of sludge significantly [2]. The addition of organic polymers has no positive influence on the high EPS content and the water retention in the sludge flocs, and so fails to achieve deep dewatering of sewage sludge. New methods such as microwave conditioning [3], electrolysis [4] and chemical oxidation [5,6] have been developed to disrupt EPS or to release the bound water, which both enhance sludge dewaterability.

Fenton and Fenton-like peroxidation have been widely investigated as alternative methods for sludge conditioning [7,8]. Utilizing iron-catalyzed decomposition of H_2O_2 in acidic solutions, the Fenton reaction produces hydroxyl radicals ($^{\cdot}OH$), which effectively collapse and degrade EPS [9,10]. However, the optimal conditions of Fenton treatment for sludge conditioning were inconsistent in previous studies. For instance, Neyens et al. [11] reported that the optimal pH, reaction time and dosages of Fe^{2+} and H_2O_2 were 3, 60–90 min, 1.67 mg/g and 25 mg/g DS (dry solids), respectively. But Zhang et al. [12] stated the optimal dosages of Fe^{2+} and H_2O_2 to be 31.9 mg/g and 33.7 mg/g DS, while the initial pH of the sludge was adjusted to 5 and the reaction time to 30 min. Meanwhile, considerable research also suggested the dewatering performance of the sludge conditioned by Fenton's reagent varies greatly under different conditions, requiring further studies to illuminate this issue.

Furthermore, as well known, few municipal sewage treatment plants actually apply the Fenton reaction to sludge dewatering, due to the obvious shortcomings associated with the currently

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Nomenclature

SRF	specific resistance filterability	VS	volatile solids content
CST	capillary suction time	TOC	total organic carbon
DS	dry solids content	SEM	scanning electron microscope
EPS	extracellular polymeric substances	ANOVA	analysis of variance
TS	total solids content	D_F	fractal dimension value

available Fenton treatment process. Firstly, a lack of efficiency compared to polymer conditioning, with reaction times usually ranging from 30 to 90 min in previous studies [8,12]. For a large-scale sewage treatment plant, a rapid and efficient technology is needed to deal with the large quantities of sludge. Moreover, skeleton builders such as quick lime, cement and fly ash were used at relatively high dosages in combination with Fenton's reagent. During the pressure filtration process, skeleton builders can form a permeable, rigid lattice structure in sludge cakes, which increase the rate and extent of sewage sludge dewatering [13,14]. Skeleton builders could tremendously improve the dewatering performance achieved by Fenton treatment, and had been used at concentration up to 850 mg/g DS [15], which would lead to an expansion in sludge volume. Finally, the previous studies on the optimization of the treatment process seldom paid attention to the influence of pH value. Consequently, there has no convincing conclusion on the optimal pH for Fenton peroxidation. Lu et al. [16] claimed that there was no significant effect on the dewaterability of sludge conditioned with Fenton's reagent at pH values ranging from 2.5 to 7.0, while Neyens et al. [11] reported optimal pH values of 3 and 5, respectively. However, both the initial and final pH of the Fenton reaction should be regarded as important experimental parameters.

Pilot tests are needed to confirm optimal conditions based on laboratory results. Liu et al. [15] performed such scaling up tests for the dewatering of sewage sludge conditioned by Fenton's reagent. However, these tests might lack of representation and accuracy due to the limitations of their dewatering equipment. The filter press, a major component of dewatering set-ups, used in previous studies usually could not generate filtration pressure much greater than 2.0 MPa, leading to the inadequate dewatering. To obtain reliable results, a novel filter press which could achieve high filtration pressure (40.0 MPa) was utilized in the pilot-scale experiments.

This study was carried out to explore a more rapid and practical Fenton treatment technique for sludge dewatering and to provide an example of proactive treatment engineering. Experimental conditions, including reaction time, pH and dosage of Fe^{2+}, H_2O_2 and lime were optimized through single-factor experiments and orthogonal experimental design. Based upon the results obtained in laboratory investigations, pilot-scale experiments using high-pressure were conducted to consummate the whole sludge treatment technique. Meanwhile, the mechanism of sludge dewatering in the conditioning system was investigated.

2. Materials and methods

2.1. Materials

Sludge samples were collected from the secondary sedimentation tank of a local wastewater treatment plant in Guangzhou, China. Samples were transported to the laboratory in polypropylene containers and stored at 4 °C for about 12 h. The supernatant was decanted to acquire the denser sludge samples that were used in this study (characteristics shown in Table 1). The sludge homogenization was performed by rapid stirring before taking smaller samples for characteristics measurement and experiment.

In Fenton experiments, H_2SO_4 (Analytical grade; Guangzhou Chemical Reagent Factory, China) was used to adjust the initial pH of sludge. $FeSO_4 \cdot 7H_2O$ and H_2O_2 solution (30 wt.%; industrial grade; Damao Chemical Reagent Factory, China) were used as the sources of Fe^{2+} and H_2O_2. Quick lime (Industrial grade; Tianjin Fuchen Chemical Reagents Factory, China) was used both as a neutralizer and a skeleton builder.

2.2. Experimental procedure

2.2.1. Laboratory-scale investigations

Samples of raw sludge (500 ml) were prepared in 1000 ml beakers and mixed well in a jar test apparatus (ZR4-6, Zhongrun, China) and adjusted to the desired pH prior to Fenton treatment. The characteristics of the sludge samples were mentioned in Table 1. Fe^{2+} ($FeSO_4 \cdot 7H_2O$) was added to the sample and the Fenton reaction was then initiated by addition of H_2O_2. The sample was stirred at 400 rpm for the desired reaction time. After the reaction, the sample was conditioned and neutralized by lime.

Initially, a series of single-factor experiments were conducted to obtain the optimal reaction time. After that, an orthogonal design $L_{16}(4^5)$ was chosen to evaluate the combined effects of the four variables (pH, Fe^{2+}, H_2O_2 and lime). The levels of the four factors were defined according to our previous tests, as shown in Table 2.

The specific resistance filterability (SRF) and capillary suction time (CST) were examined as evaluation parameters. Simultaneously, total organic carbon (TOC), DNA in the sludge supernatant and the final pH of the treated sludge were analyzed to aid our investigation into the mechanisms of sludge dewatering. Furthermore, loosely bound EPS (LB-EPS) and tightly bound EPS (TB-EPS) [17] concentration of several sludge samples was measured to optimize the mechanism investigation. In order to assess the disintegration degree of sludge and cells, the DNA concentration of the sludge supernatant was measured. DNA is a unique material inside the cell. So the content of DNA is usually used to characterize the degree of the cell rupture.

2.2.2. Pilot-scale experiments for constituting an integrated technique

Optimal conditions from the laboratory studies were evaluated by a novel sludge dewatering system, which was an important part of a whole sludge treatment technique. The highlight of this system is the spring filter press, which is predominantly steel and can achieve the squeezing pressure up to 40 MPa. The sludge conditioning and dewatering system is illustrated in Fig. 1. The hydraulic system can stably generate high pressure and transfer it to the filter plates. Since each filter plate contains six steel springs, the volume of the treated sludge can be further reduced during the pressing process, resulting in the improvement of dewatering effect. Raw sludge was transferred to a 100 L conditioning tank and then conditioned according to the desired dosage of the composite conditioner and Fenton reaction time.