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Multivariate analysis of sludge disintegration by microwave–hydrogen peroxide pretreatment process

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HIGHLIGHTS

- Investigation of TSS, H_2O_2 dosage, pH and interactions on MW sludge pretreatment.
- Quadratic models were drawn for 16 response variables with good predictive ability.
- Models could optimize the treatment process for multiple disintegration objectives.

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ABSTRACT

Microwave irradiation (with H_2O_2) has been shown to offer considerable advantages owing to its flexible control, low overall cost, and resulting higher soluble chemical oxygen demand (SCOD); accordingly, the method has been proposed recently as a means of improving sludge disintegration. However, the key factor controlling this sludge pretreatment process, pH, has received insufficient attention to date. To address this, the response surface approach (central composite design) was applied to evaluate the effects of total suspended solids (TSS, 2–20 g/L), pH (4–10), and H_2O_2 dosage (0–2 w/w) and their interactions on 16 response variables (e.g., $SCOD_{released}$, pH, $H_2O_{2\ remaining}$). The results demonstrated that all three factors affect sludge disintegration significantly, and no pronounced interactions between response variables were observed during disintegration, except for three variables (TCOD, $TSS_{remaining}$, and $H_2O_{2\ remaining}$). Quadratic predictive models were constructed for all 16 response variables (R^2: 0.871–0.991). Taking soluble chemical oxygen demand (SCOD) as an example, the model and coefficients derived above were able to predict the performance of microwave pretreatment (enhanced by H_2O_2 and pH adjustment) from previously published studies. The predictive models developed were able to optimize the treatment process for multiple disintegration objectives.

1. Introduction

Conventional biological wastewater treatment processes, when applied widely, produce vast amounts of waste activated sludge (WAS), the treatment and disposal of which are difficult and expensive for municipal wastewater treatment plants (WWTPs). However, WAS is gaining prominence as a potential bio-resource, although sludge reutilization is impeded by factors such as the protective cell walls and matrix of extracellular polymeric substance (EPS). To overcome these obstacles, sludge pretreatment technologies have been developed incrementally in several previous studies [1,2]. Pretreated sludge has been demonstrated to be suitable for many purposes, including the following: (1) enhancing anaerobic digestion of WAS to improve biogas production [3–5] or energy recovery by microbial fuel cells [6]; (2) recovering nutrients (nitrogen and phosphorus) or material resources (e.g., proteins, VFAs for the production of polyhydroxyalkanoates) from sludge [7]; (3) improving sludge dewatering to promote reduction in sludge volume or sludge disinfection [8]; and (4) reducing sludge production by cryptic growth [9,10].

Methods for sludge disintegration by mechanical [11], thermal [12], and chemical [13] treatment have been proposed previously. Microwave (MW) pretreatment, which offers advantages such as the rapid application of direct heat and reduction of energy losses, is an alternative method to conventional thermal pretreatment and has garnered increasing attention recently [14,15]. In particular, microwave pretreatment was found to be superior to thermal treatment in terms of sludge solubilization and biogas production [16]. Moreover, changes in the dipole orientation of polar molecules occur during microwave irradiation, producing athermal

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(or non-thermal) effects [17]. For the solubilization of waste activated sludge, the effectiveness of microwave irradiation is affected by several factors, which have been investigated extensively [3]; such factors include MW output power [10,18], target temperature [19], and sludge concentrations [20,21]. However, investigation of these factors through conventional "change one factor at a time" methods have some shortcomings, including that these methods are laborious, time-consuming, and incapable of obtaining true optimal values owing to their inability to consider interactions among variables. Therefore, three-factor fixed-effect analysis of variance (ANOVA) determination has been adopted to evaluate the effects of high temperature (110–175 °C), MW intensity (1.25 and 3.75 °C/min), and sludge concentration (6 and 11.85%) on the solubilization of sludge [22]; the interaction of these factors was found to be significant at the 94% confidence interval. Moreover, the effects of heating pretreatment on the degree of solubilization of waste activated sludge are investigated using response surface analysis [21], and the conditions required to produce a maximum solubilization degree of 17.9% were predicted to be 400 W (output power 400–1600 W), 102 °C (target temperature 60–120 °C), and 2.3% TS (total solid concentration, 1–3%).

The addition of H_2O_2 in a closed-vessel MW digestion system has been reported to enhance the pretreatment of sludge markedly [23], and such advanced oxidation processes (AOPs) appear to offer promise as suitable technologies for the minimization [24] and pasteurization and stabilization [25] of excess sludge. A H_2O_2 dosing strategy to inhibit the adverse effects of catalase at low temperatures in an open system was developed in our previous work [26]. To optimize the microwave-enhanced advanced oxidation process (MW/H_2O_2–AOP), screening experiments (four factors: temperature, hydrogen peroxide dosage, mixing, and solids concentration) were conducted [20]; appropriate solids disintegration and nutrient release were determined to occur at 120 °C and 0.80 g H_2O_2/g dry sludge. Yin et al. [27] found initial sludge TS content and hydrogen peroxide dosage were the most significant factors controlling nutrient solubilization (i.e., more significant than heating temperature and heating time), maximum solubilization was obtained for 2.5% TS, 2 wt% hydrogen peroxide, and 5 min of microwave heating at 120 °C.

Microwave–H_2O_2 has been reported to perform better in substrate degradation under acidic and neutral conditions [28]. Moreover, the effects of acids (e.g., HCl and H_2SO_4) on sludge disintegration have been shown to enhance the release of ammonia [29]. Microwave-enhanced advanced oxidation processes have been adopted previously for the treatment of dairy manure at low pH [30]; such low-pH methods are known to offer advantages such as enhanced phosphorous release and promotion of the dewatering of sludge [31]. Conversely, combined MW irradiation (160 °C) with an alkaline pretreatment method (using NaOH, pH ~ 12.5) demonstrated that this technique could increase the solubilization ratio (in terms of SCOD/TCOD) to 0.37 [3].

Recently, Hong et al. [28] found that the degradation of rhodamine B (RhB) and methylene blue (MB) in the MW–H_2O_2 system was very competitive at extreme alkaline pH, suggesting that this may be an appropriate means of promoting degradation under highly alkaline conditions; this result is encouraging for the development of MW–H_2O_2 technology in alkaline wastewater treatment. Similarly, it has been shown that high pH may enhance the MW thermal effect, making MW techniques more suitable for application in sludge disintegration [15]. However, few studies to date have investigated the microwave–H_2O_2 system under alkali conditions.

A priori, and on the basis of a literature survey on MW and AOPs [23–28], TSS, H_2O_2, and pH were known to be the critical factors controlling sludge disintegration at mild temperatures (100 °C) [32]; however, the effect of pH (from acidic to basic) has not yet been fully recognized. In addition, the interactions among these three factors remain unclear. Moreover, the majority of previous researches have focused on the treatment of target sludge, neglecting the behavior of the oxidant H_2O_2 during this process. In practical engineering applications of this technology, it is always designed to achieve multi-objectives for the sludge disintegration unit; for example, the sludge can be treated to release more organic matter and less heavy metal. If recovery of N/P in the form of struvite is to be considered, phosphate and ammonia (and Mg^{2+}) release should be maximized, whereas the release of Ca^{2+} (which acts as an inhibitor) should be minimized. Therefore, there is great demand for a unifying predictive model that incorporates the above aspects of sludge pretreatment and will allow simultaneous quantitative evaluation of sludge disintegration and optimization of MW–H_2O_2 sludge pretreatment by pH adjustment for multi-objectives.

Response surface methodology (RSM) is a powerful statistical tool used to construct models and evaluate the influences of several individual factors and their interactions simultaneously. Accordingly, RSM has emerged as an important tool in the study of multifactor interaction. Typically, predictive models have been used to analyze and optimize operation parameters, thus allowing desirable responses to be attained while reducing the number of experiments.

The present study adopted multivariate analysis to investigate sludge disintegration and aimed to achieve the following: (1) investigate the effects of pH in isolation and in conjunction with uniform initial sludge concentration and H_2O_2 dosage, and assess the interactions among these factors during sludge disintegration based on RSM and 16 response variables; and (2) construct a unifying model to predict sludge solubilization and H_2O_2 usage, thus providing a simple way to estimate optimal values for multiple sludge pretreatment objectives.

2. Materials and methods

2.1. Raw materials

The WAS was obtained from returned sludge from the secondary settling tank of the Fangzhuang municipal wastewater treatment plant (WWTP), with a design capacity of 40,000 m^3/day, in Beijing, China. This WWTP was an A^2/O process plant and operated with sludge retention time (SRT) of approximately 15 days. The high VSS to TSS ratio in the sludge (79%) indicates that it consisted mainly of organic substances. Waste activated sludge was centrifuged before use and washed three times to avoid the interference of soluble matter. The sludge was stored at concentrations of 3% at 4 °C and diluted with distilled water prior to use to provide various set concentrations.

2.2. Experimental design

The effects of the studied variables (TSS, pH, H_2O_2 dosage) and their interactions on the response variables were investigated using a central composite design (CCD) method. The CCD method adopted allows the development of mathematical equations. TSS (X_1), pH (X_2), and the H_2O_2 to sludge ratio (X_3) were varied over the ranges 3–20 g/L, 2–12, and 0–2 (w/w), respectively, with corresponding central values of 11.5 g/L (X_1), 7 (X_2), and 1.0 (X_3). X_i denotes the real value of the three variables according to the experimental design (Table 1, actual values). The parameters were standardized according to the following equation.

$$x_i = \frac{X_i - X_i^o}{\Delta X_i} \tag{1}$$

where x_i was the coded value of the variable X_i, X_i^o was the value of X_i at the center point of the investigated area, and ΔX_i was the