Desalination and Water Treatment
www.deswater.com
doi: 10.5004/dwt.2021.27755

238 (2021) 231–239
October

Fenton-microwave pretreatment of activated sludge for efficient biogas generation

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Received 24 April 2021; Accepted 26 August 2021

ABSTRACT

Excess activated sludge produced from textile treatment plants represents an environmental and an economical problem. The aim of this study is to optimize sludge disintegration by Fenton-microwave pretreatment (FMT) which helps improve its digestibility. The response surface methodology was applied. Three independent factors are selected: microwave power, microwave treatment time and the concentration of H_2O_2 ([H_2O_2]). Two responses were measured: total extracellular polymeric substances (tEPS) and soluble chemical oxygen demand (sCOD). The results of the optimization of FMT were a maximum sCOD of 2,045.2 mg/L, a tEPS of 7,155.6 mg/L with a microwave specific energy (M_{SE}) of 50,004 kJ/kg TS in optimal conditions of 800 W, 90 s and a 79 mg/g of [H_2O_2]. Experimental results were close to predicted results. Hence, models are globally validated but need to be better improved. FMT proved to be more efficient than the control in biogas production.

Keywords: Activated sludge disintegration; Fenton-microwave pretreatment; Response surface methodology; Anaerobic digestion

1. Introduction

Activated sludge is used as a biological purification method in wastewater textile treatment [1]. After cellular oxidation, the dissolved organic matter present in the wastewater transformed into a decanted colossal matter [2]. Decanted sludge is discarded after dewatering [3]. Transporting sludge to landfills presents an economic problem, mainly due to transport costs [4]. On the other hand, the accumulation of large quantities of sludge poses a major environmental problem due to their composition in undesirable elements [5]. It is, therefore, necessary to find solutions to valorize this waste.

Anaerobic digestion is considered as an efficient method to reduce sludge volumes and promote biogas production [2,6]. It consists of organic matter fermentation using anaerobic fauna [2,6]. Nevertheless, the rigid arrangement of the microbial cell walls of activated sludge, which acts as the protective layer of the internal cell products, reduces its solubility and consequently reduces its hydrolysis [7].

To overcome this limitation, many conventional sludge pretreatments have been proposed to increase their solubilization, which improves biogas production and reduces retention times.

Conventional chemical methods consist of the use of chemicals such as Sodium Hydroxide, calcium hydroxide and nitrite to dissociate EPS matrix [8–10]. These methods showed improvement in sludge digestion, but still have many disadvantages like high chemical cost and toxicity of some alkali like Na^+ [11].

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Fenton's reaction is one of the most effective advanced oxidation processes used in sludge chemical pretreatment [12]. The high efficiency of this method results from the formation and the release of hydroxyl radicals $HO^{\bullet}$ generated during hydrogen peroxide decomposition in the presence of Fe^{2+} ions (Eq. (1)) [12]. The Fe^{3+} can be reduced again to Fe^{2+} (Eq. (2)) [13]. The advantage of Fenton reaction is its simplicity and availability [13].

$$H_2O_2 + Fe^{2+} \rightarrow Fe^{3+} + HO^{\bullet} + OH^- \quad (1)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^{\bullet} + H^+ \quad (2)$$

Conventional thermal treatment consists of applying thermal energy via convection, allowing the destruction of the chemical bonds of the cell membrane [14]. However, this method can generate recalcitrant compounds that induce the inhibition of anaerobic biodegradability substrate [15]. Also, it has a prolonged disintegration time and a high temperature that can reach up to 270°C which requires a huge amount of electrical energy [14]. This makes the process economically infeasible [7,15].

Microwave irradiation is one of the most potent heat alternative treatment techniques for sludge disintegration [7]. It has proved to be an efficient tool for cell destruction, DCO liquefaction, EPS dissociation and biogas production while preserving rapid heating, high reaction rate, ease of control, compactness, space-saving and energy efficiency over the conventional thermal processes [16–18].

In microwave irradiation, electromagnetic energy penetrates the material and converts to heat inside the particle. Heat is generated within the material and is conveyed towards the outside [19,20].

Despite the advantages of the microwave and Fenton reaction on sludge disintegration, they still have many disadvantages like treatment cost and energy consumption, which can be minimized during their combination [19,21].

Previous studies showed that combined treatments can improve sludge solubility while reducing energy consumption and treatment costs [7,22].

Qi and Li [23] reported that Microwave-assisted Fenton-like processes not only can achieve treatment efficiency similar to that of stand-alone Fenton processes, but also reduce reagent dosage and shorten the reaction time. Eswari et al. [7] reported that H_2O_2 coupled to microwave irradiation of dairy waste activated sludge allowed not only reducing treatment costs but also enhancing the dissociation of extracellular polymeric substances (EPS) matrix. It subsequently improved the methane yield generation compared to conventional microwave disintegration.

However, understanding the interactions modality between the combinations of two techniques is still more complex.

Response surface methodology (RSM) is a statistical technique that can be applied to understand complex systems and analyze data in which a response of interest is influenced by different independent variables and to evaluate the simultaneous effects of several factors [24]. In addition, RSM is applied to search for the optimum conditions for desirable responses [8,25,26]. These reasons convinced several industrials and researchers to adopt RSM in processes evaluation.

The main objectives of the present study are to improve activated sludge disintegration potential by a Fenton reaction combined with a microwave treatment in an optimized system using response surface methodology with Box–Behnken design. Total extracellular polymeric substance (tEPS) and soluble chemical oxygen demand (sCOD) were used as an indicator for measuring sludge disintegration. Specific energy (SE) was used to control energy costs. After that, anaerobic digestion assay was carried out.

2. Material and methods

2.1. Samples collection

Dewatered aerobic activated sludge (DAS) was collected from a textile wastewater treatment plant in Ksar Hellal, Monastir, Tunisia after a filter press process. The collected aerobic sludge was stored at 4°C for subsequent study.

Cow manure was freshly collected from a cow raising plant in Swassi, Monastir, Tunisia. The collected inoculum was also stored at 4°C for anaerobic digestion.

2.2. Analytical methods

sCOD was analyzed using HACH experimental kits. Kits mixed with the liquid part of sludge were incubated for 2 h at 150°C and color shift corresponding to COD was detected by HACH (3900) spectrophotometer. tEPS were extracted within a modified Morgan method [27]. A solution of 10% of sludge was prepared with 0.05% NaCl solution then dewatered by centrifugation in 50 mL tubes at 8000 rpm for 15 min. The supernatant contains soluble EPS. The sludge pellet in the tube was then re-suspended into 10 mL of 0.05% NaCl solution. Then, a heat NaCl solution at 70°C was added to the mixture to get a total volume of 50 mL with a warm temperature of 50°C. Sludge suspension was then sheared by a vortex mixer for 1min, followed by centrifugation at 8,000 rpm for 15 min. The supernatant contained loosely bound EPS. For the extraction of tightly bound EPS, the sludge pellet left in the centrifuge tube was re-suspended in a 0.05% NaCl solution to its original volume of 50 mL. The sludge suspension was heated to 60°C in a water bath for 30 min, and the sludge mixture was then centrifuged at 8,000 rpm for 20 min. The supernatant contained tightly bound EPS of the sludge. Initial haracteristics of dewatering activated sludge are shown in Table 1.

Table 1
Initial characteristics of dewatering activated sludge

Parameters	Value
Total solids, %	14.34 ± 1.3
Fixed solids, %	8 ± 0.14
Volatile solid, %	92 ± 0.14
sCOD, mg/L	13 ± 0.9
tEPS, mg/L	785 ± 15

2.3. Fenton-microwave pretreatment

A commercial microwave oven (800 W, 2450 MHz frequency, Samsung, Model ME732K) and a vessel made up of poly-tetra-fluoro-ethylene (PTFE) were used to pre-treat the DAS. The sludge disintegration was done with 10 g of DAS diluted in 20 mL of distilled water. Microwave treatment time and microwave power were tested for effective sludge solubilization.

The microwave specific energy input (M_{SE}) during the microwave disintegration experiment can be calculated as follows [7]:

$$M_{SE}\left(\text{kJ/Kg TS}\right)=\frac{P\times T}{V\times \text{TS}} \tag{3}$$

where M_{SE} = microwave specific energy input (kg/kg TS); P = microwave power consumed during the experiment (kW or kJ/s); T = treatment time (s); V = volume of the sample taken for the experiment (L); TS = total solids of the sample taken (kg/L).

Microwave power consumed was calculated as follows

$$P=\text{Power}\left(\%\right)\times \text{Total power of microwave} \tag{4}$$

Disintegrated sludge induced by microwave was followed by a Fenton reaction pretreatment.

2.4. Fenton reaction's pretreatment

All Fenton experiments were run with 10% of dewatered pretreated sludge in a total water volume of 100 mL. The pH of the solution was adjusted to 3 with H_2SO_4 and experiments were taking place at ambient temperature in 100 mL capacity glass beakers. Magnetic stirrers were used to provide continuous mixing during the Fenton experiments. In order to initiate the Fenton reaction, an appropriate amount of ferrous sulfate $Fe(SO_4)\cdot 7H_2O$ was added and steered for 2 min. To achieve reaction, an amount of $H_2O_2$was added to the sludge solution. The ratio of 0.07 g Fe^{2+} / 1 g of H_2O_2 was added [12]. After 1 h of reaction time, the pH of the solution was adjusted to 11 with NaOH 20% in order to achieve the maximum amount of $Fe(OH)^3$ precipitation. Then, an excess of sodium thiosulphate was added to block Fenton reaction. Subsequently, sCOD and tEPS of pretreated sludge were determined.

2.5. Biogas production assay

All experiments were performed in 1 L glass reactor bottles with 50 g of substrate and 10 g of fresh cow manure inoculums diluted in water. The substrates were control (untreated DAS) and Fenton-microwave pretreatment (FMT) samples. The pH of mixtures was adjusted initially at 8. The bottles containing samples were purged with nitrogen gas for about 2 min to maintain strict anaerobic conditions. Then the bottles were kept in a water bath at 40°C during a hydraulic retention time of 20 d. Reactors were shaken twice a day. The biogas was measured using the replacement liquid method with a 3 M NaOH solution to absorb available carbon dioxide in the biogas [28].

2.6. Design of experiments and optimization using RSM

RSM was applied to optimize factors influencing sludge disintegration using the MINITAB 17 software via the Box Behnken experimental design. RSM allows developing a mathematical correlation between responses (sCOD and tEPS) and three independent selected variables, including microwave applied power, microwave treatment time and $[H_2O_2]$. Experimental levels of variables, in coded and actual values are presented in Table 2.

The experimental results of RSM were fitted using the second-order polynomial regression equation.

$$Y=b_0+\sum_{i=1}^{k}b_iX_i+\sum_{i=1}^{k}b_{ii}X_i^2+\sum_{i}^{i<j}\cdot\sum_{j}b_{ij}X_iX_j \tag{5}$$

where Y: the predicted response, b_0: the constant coefficient, b_i: the linear coefficients, b_{ii}: the quadratic coefficients, b_{ij}: the interaction coefficients; X_i,X_j: are factors variables in codified form.

2.7. Statistical analysis

Analysis of variance (ANOVA) and multiple regressions were used to evaluate the adequacy of the model. The significance of each variable was determined by applying P-value which is related to F-value (confidence level of 95%).

ANOVA, diagnostics plots, model graphs which allow analyzing the effects of variables individually, the combination between factors, and optimization prediction of the levels of each variable for maximum response were obtained using the MINITAB 17.

3. Results and discussion

3.1. Application of FMT for sludge disintegration

In this study, FMT pretreatment method was evaluated to enhance sludge disintegration.

Table 2
FMT pretreatment independent variables and their levels

		Levels of variables		
		Low	Mid	High
Combined pretreatment	Independent variables	–1	0	1
Fenton-microwave disintegration	Microwave pretreatment time (Mtt) (s)	40	100	160
combined pretreatment	Microwave power (Mp) (w)	100	450	800
	$[H_2O_2]$ (mg/g TS)	27	54	81

Several factors influence the effectiveness of the combined pretreatment [22,23]. Therefore, they influence sCOD and tEPS. To understand sCOD and tEPS released from DAS using FMT, response surface methodology has been applied via the Box–Behnken design.

3.1.1. Response regression equation

By applying multiple regression analysis on the experimental data, the full quadratic polynomial Eqs. (6) and (7) were derived to explain sCOD and tEPS models (Table 3).

3.1.2. Model adequacy

The full quadratic equation model was chosen as an appropriate model that can express the adequate correlation between responses and the independent variables. Indeed, the regression coefficients R^2 relative to the sCOD and tEPS are high ($R^2 > 90\%$) (Table 4).

ANOVA for the second-order polynomial models showed that all responses models were significant, as was clear from Fisher's *F*-test ($p < 0.05$) (Tables 5 and 6).

Concerning the significance of the factors, microwave treatment time (Mtt) and microwave power (Mp) had a significant effect on sCOD of FMT. All factors had a significant effect on tEPS of FMT.

Residual plots were used to examine the goodness-of-fit in regression and ANOVA (Figs. 1 and 2).

The normal probability plot of the residuals was used to verify the supposition that the residuals are normally distributed. Figs. 1a and 2a show that the normal probability plot of the residuals approximately followed a straight line.

The residuals vs. fits plot were applied to verify the supposition that the residuals are randomly distributed and have constant variance. Figs. 1b and 2b show that the points fall randomly on both sides of 0, with no recognizable patterns in the points.

The residuals vs. order plot were applied to verify the supposition that the residuals are independent from one another. Independent residuals show no trends or patterns when displayed in time order. Figs. 1c and 2c show that the residuals on the plot fall randomly around the center line.

There is no evidence to indicate that the regression terms are correlated with each other. We can see that the model is adapted to describe the responses by the response surface methodology.

3.1.3. Main effects plot analysis

The main effects plot was used to examine differences between level means for one or more factors.

Fig. 3 shows the main effect plots of tEPS. Different levels of the factors affect the response differently. Microwave treatment time and microwave power had a positive main effect on response for FMT. In fact, the increase of Mtt from 40 to 160 s led to the increase of tEPS from 5,050 to 7,250 mg/L and the increase of Mp from 100 to 800 w led to the increase of tEPS from 4,750 to 7,250 mg/L. However, $[H_2O_2]$ had both a positive and a negative main effect: the increase of the factor from 27 to 57 mg/g led to the increase of tEPS from 4,750 to 6,250 mg/L but the increase of the same factor from 54 to 81 mg/g induces a lower decrease of the response from 6,200 to 6,000 mg/L. The negative main effect of H_2O_2 observed in tEPS of FMT was explained by the fact of using excess reagents such as ferrous salt or hydrogen peroxide which has led to secondary reactions [13]:

$$Fe^{2+} + HO^{\bullet} \rightarrow Fe^{3+} + OH^{-} \tag{6}$$

$$H_2O_2 + {}^{\bullet}OH \rightarrow H_2O + HO_2^{\bullet} \tag{7}$$

So, the reagents have become typical radical scavengers, instead of generating new quantities of radicals [29].

Fig. 4 shows the main effect plots of sCOD. Mtt and Mp had a positive main effect only in the range level 40–100 s and 100–450 W respectively, where the first response was increased from 1,150 to 1,625 mg/L and the second was increased from 700 to 1,750 mg/L. For a higher value, there is no main effect of the two factors. $[H_2O_2]$ had a low positive main effect. In fact, the increase of the factor from 27 to 81 mg/g led to a lower increase in the response from 1,370 to 1,510 mg/L.

Treatment time and thermal power were proved as determinant factors on sludge pretreatments [23,30]. In most cases, their increase induced improvement disintegration. They acted simultaneously on solids biomass and bounded EPS matrix and they typically control how much energy was introduced to sludge [31,22]. In Fenton reaction, H_2O_2 concentration is a highly important factor affecting the

Table 4
Regression analysis of tEPS and sCOD models

Pretreatment	FMT	
Response	tEPS	sCOD
S	344.542	137.288
R^2 value	97.81	97.90
Adjusted R^2	93.86	94.12
Predicted R^2	64.94	66.41

Table 3
Regression equations of FMT

Pretreatment method	Regression equations
FMT	sCOD = –1,649 + 26.43 Mtt + 5.452 Mp + 17.1 $[H_2O_2]$ – 0.0790 Mtt × Mtt – 0.004465 Mp × Mp – 0.1430 $[H_2O_2]$ × $[H_2O_2]$ – 0.00843 Mtt × Mp – 0.0681 Mtt × $[H_2O_2]$ + 0.01823 Mp × $[H_2O_2]$ tEPS = 1,409 – 22.5 Mtt – 1.05 Mp + 123.7 $[H_2O_2]$ + 0.2053 Mtt × Mtt + 0.00359 Mp × Mp – 0.953 $[H_2O_2]$ × $[H_2O_2]$ + 0.00376 Mtt × Mp – 0.063 Mtt(s) × $[H_2O_2]$(mg/g) + 0.0142 Mp × $[H_2O_2]$

Table 5
ANOVA analyses of tEPS quadratic model

Source	Sum of squares	Df	Mean square	F-value	P-values Prob. > F
Model	26,494,742	9	2,943,860	24.80	***0.001***
Linear	21,527,254	3	7,175,751	60.45	***0.000***
Mtt	8,127,504	1	8,127,504	68.47	***0.000***
Mp	10,860,713	1	10,860,713	91.49	***0.000***
$[H_2O_2]$	2,539,037	1	2,539,037	21.39	***0.006***
Square	4,828,616	3	1,609,539	13.56	***0.008***
Interaction	138,872	3	46,291	0. 39	0.766
Error	593,544	5	118,709		
Total	27,088,287	14			

Significance values are in bold-italic.

Table 6
ANOVA analyses of sCOD quadratic model

Source	Sum of squares	Df	Mean square	F-value	P-value Prob. > F
Model	4,395,019	9	488,335	25.91	***0.001***
Linear	2,771,834	3	923,945	49.02	***0.000***
Mtt	286,146	1	286,146	15.18	***0.011***
Mp	2,429,910	1	2,429,910	128.92	***0.000***
$[H_2O_2]$	55,778	1	55,778	2.96	0.146
Square	1,330,568	3	443,523	23.53	***0.002***
$[H_2O_2] \times [H_2O_2]$	40,128	1	40,128	2.13	0.204
Interaction	292,617	3	975,316	5.18	0.050
Lack of fit	94,240	3	31,413		
Total	4,489,259	14			

Significance values are in bold-italic.

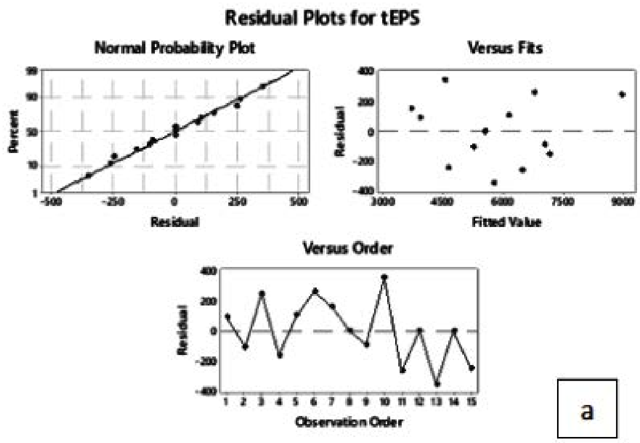

Fig. 1. Residual plots for tEPS.

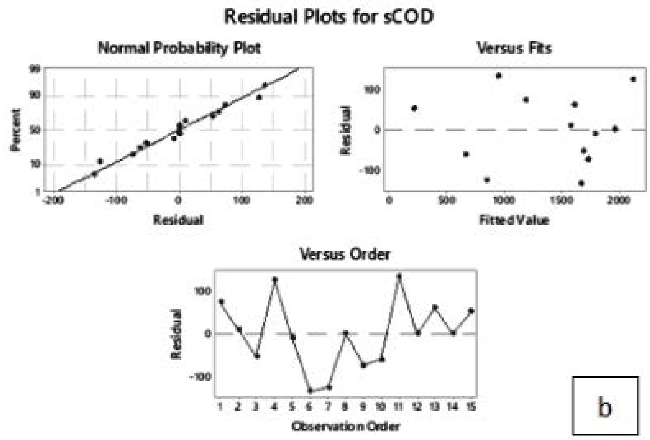

Fig. 2. Residual plots for sCOD.

degradation efficiency of organic matter [3,32]. However, H_2O_2 must be added with a limited amount. In excessive H_2O_2 disturbs Fenton process and increases operational costs [31].

3.1.4. Contour plots analysis

Contour plots provide a two-dimensional view during which all points having an equivalent response are connected to supply contour lines of constant responses. The

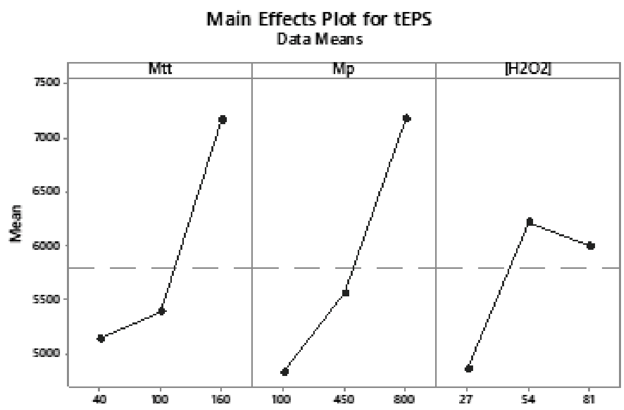

Fig. 3. tEPS main effects plot diagrams relative to FMT.

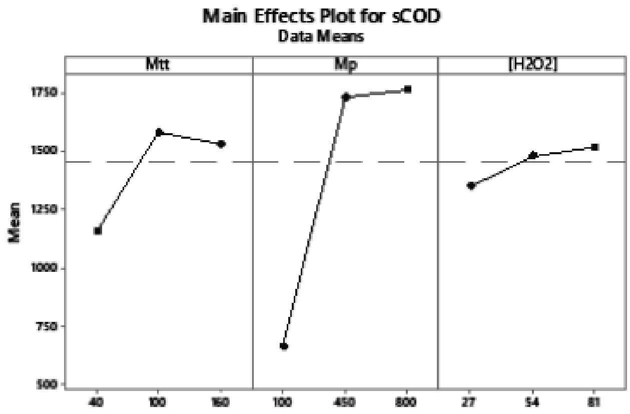

Fig. 4. sCOD main effects plot diagrams.

contour plot (Fig. 5a) shows the relationship between microwave power and microwave treatment time on tEPS. [H_2O_2] was in a mid-level. According to this figure, Mtt and Mp had a synergic effect on the response. In fact, the increase of the two factors simultaneously allows for the increase in EPS solubility. Darker regions indicate higher EPS solubility, which can exceed 8825 mg/L. The valleys in the lower left of the graph represent Mtt–Mp combinations that result in a low solubility. It was also observed that [H_2O_2] and Mtt also had a synergic effect on the response (Fig. 5b). In fact, for any fixed value of [H_2O_2], the increase of Mtt allows for the increase of tEPS. And for any fixed value of Mtt, the increase of [H_2O_2] allows for a low increase of the tEPS which can be up to 7,269 mg/L. Darker regions in the upper right of Fig. 5c indicate higher tEPS which can exceed 7,282 mg/L. This shows that the combination [H_2O_2]-Mp had synergic effects on the response.

Mtt and Mp had a synergic effect on the response (Fig. 6a). In fact, the increase of the two factors simultaneously allows increasing in sCOD. The ridge running in the upper middle of the graph indicates higher sCOD which can exceed 2,102 mg/L. It was also observed that the combination between [H_2O_2] and Mtt did not influence response variation significantly (Fig. 6b). For [H_2O_2]-Mp combination, the increase in Mp for any fixed value of [H_2O_2] led to an increase of sCOD. However, the increase of [H_2O_2] for any fixed value of Mp, led to a low increase of sCOD. An optimal value of the response was 2,174 mg/L (Fig. 6c).

Several studies showed the influence of Mtt and Mp which had a direct relationship with M_{SE} on organic matter disintegration in a combined chemical-microwave pretreatment. Qi and Li [23] showed that the increase of the two factors to 593 W and 8 min led to the increase in sludge disintegration. But a further increase limited SD. Kim et al. [30] showed that when they used a chemo-thermal pretreatment, there was a decrease in SD when SE input exceeded 20,000 kJ/kg TS. In fact, there exists a peak point past which additional energy input was attributable to a rigorous boiling that occurs and which might induce partial evaporation of released lysates from the medium [33].

3.1.5. Optimized conditions

After the evaluation of factors and combined factors on combined pretreatment efficiency using RSM, the optimization process leads to define optimal pretreatment conditions that improve sludge disintegration considering the relationship between all factors at the same time.

Optimization diagram for FMT (Fig. 7) illustrates the effect of each factor (columns) on tEPS, sCOD, SE and composite desirability (rows).

Optimization results are illustrated in Fig. 7 with overall desirability of 0.82 under optimal operating conditions of 800 W, 89.7 s of Mtt, and an [H_2O_2] of 65 mg/g TS. Under these conditions, the Minitab software provided a maximum sCOD value of 2,045.3 mg/L, a maximum tEPS value of 7,155.6 mg/L with specific target energy, which did not exceed 50,000 kJ/kg ST.

Experimental results (Table 7) were close to predicted results. Hence, our models are globally validated but need to be better improved.

Several studies have shown the performance of microwave pretreatment on sludge disintegration, which led them to use it in a combined process. Peng et al. [34] used combining microwave irradiation with sodium citrate addition for sewage sludge disintegration. Doğan and Sanin [35] used alkaline solubilization combined with microwave irradiation [35]. As for Kavitha et al. [2] they used a disperser-induced microwave disintegration of municipal waste activated sludge.

Compared with conventional thermal pretreatment, which requires a specific energy input of 72.000 KJ/kg TS [36], FMT found to consume significantly lower power. The M_{SE} spent in this study for FMT was relatively greater than that used by other investigators. Eswari et al. [7] showed using H_2O_2-microwave disintegration, that an optimal dissociation of EPS matrix (46.6%) was achieved with a 0.05 mg/g SS of H_2O_2, 259.2 W and minimal specific energy input of 10810 kJ/kg TS. Using RSM, Yang et al. showed that an optimal SE obtained using combined chemo-microwave disintegration pretreatment was 38,400 kJ/kg ST for a maximum disintegration degree of 65.8% [37]. Using less SE can be explained because of sludge properties, such as sample volume and concentration, which highly depend on SE [38].

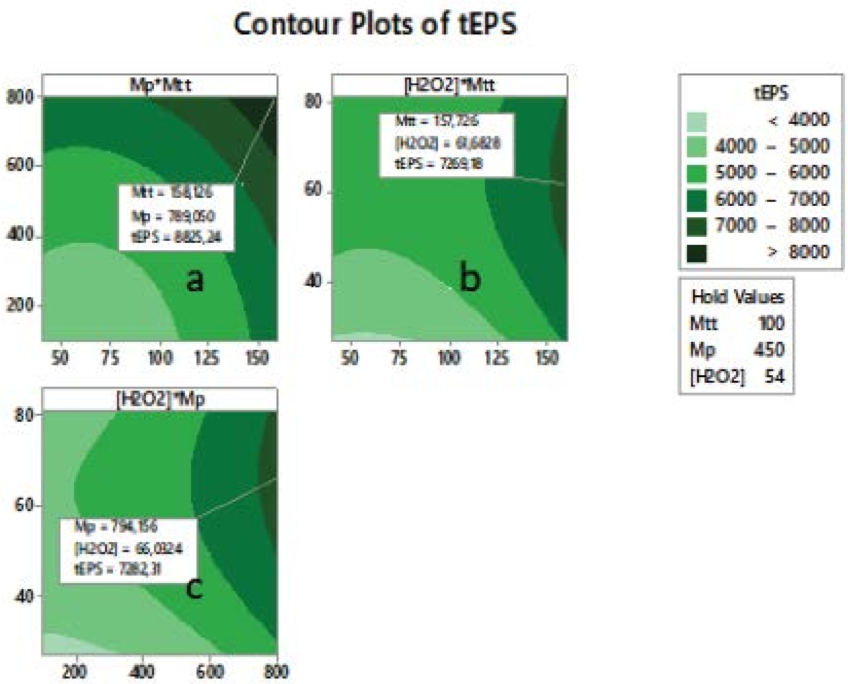

Fig. 5. tEPS contour plots.

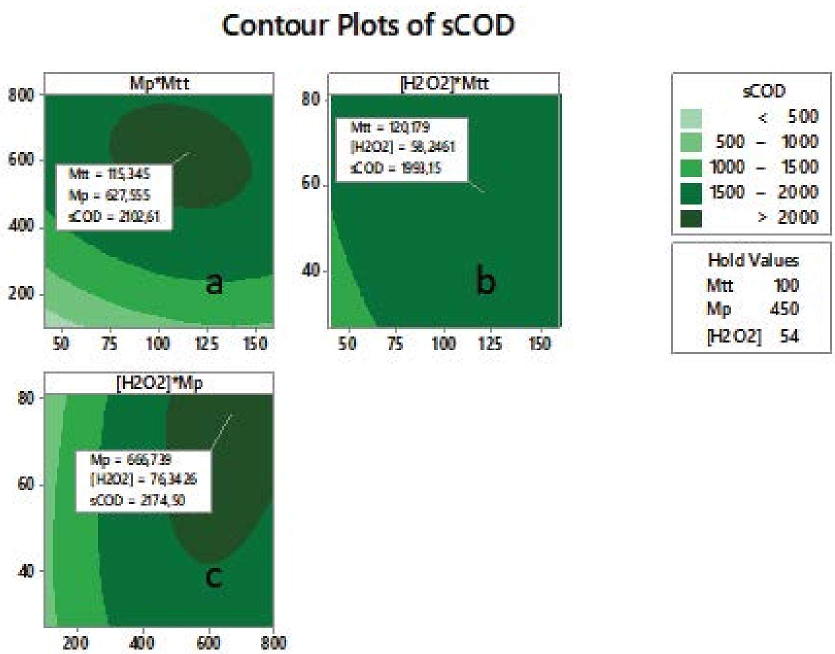

Fig. 6. sCOD contour plots.

3.1.6. Biogas production assay

Biogas production assay was executed to evaluate the efficiency of FMT on biogas production compared to the untreated sludge (control). Fig. 8 shows the total biogas produced during the digestion of substrates samples. Untreated sludge produced a lower total volume of 583 mL equivalent to 88.3 mL/g VS due to sticky flocs and harder cell walls that made digestion difficult. Biogas production was found to be greater for FMT samples compared to the control and the total biogas produced was calculated to be 868 mL equivalent to 131.5 mL/g VS at the end of the

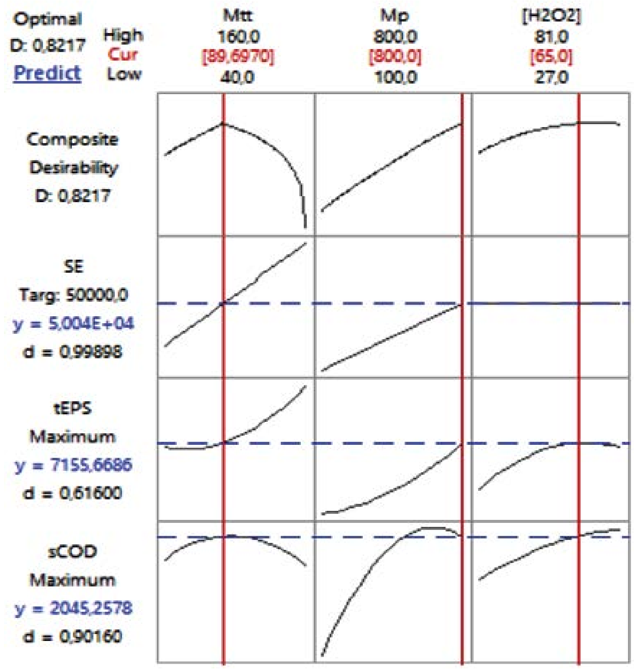

Fig. 7. Optimization diagram of FMT.

Table 7
Experimental results of FMT, under optimized conditions

Pretreatment method	FMT
Experimental results	tEPS = 6,937 mg/L ± 41.0 sCOD = 1,800 mg/L ± 32.2

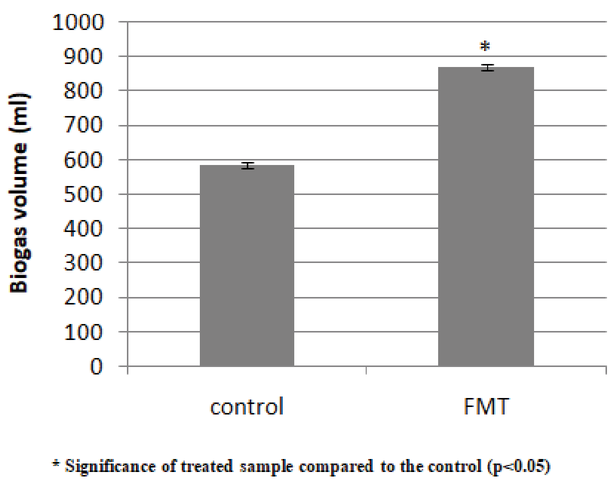

Fig. 8. Total biogas production from FMT and control sludge.

experiment (p < 0.05). The efficiency of FMT was due to the enhancement of sludge disintegration with temperature and Fenton reaction prior to the anaerobic digestion [39]. Many researchers proved the efficiency of hybrid pretreatment methods for enhancing biogas production. Eskicioglu et al. showed that maximum biogas production from sludge using microwave disintegration coupled with advanced oxidation was found to be 289.6 mL/g VS at the end of a retention time of 34 d [33]. Eswari et al. [7] showed that maximum biogas production from sludge using H_2O_2-Microwave pretreatment was found to be 250 mL/g VS at the end of a retention time of 30 d compared to control (36 mL/g VS).

FMT was more efficient to enhance anaerobic digestion compared to other pretreatments. In fact, using ultrasonic irradiation, cumulative biogas produced after 20 d of retention time was 100 mL [40]. Also, advanced oxidation of sludge using potassium ferrate allowed the production of a biogas volume of 130 mL after a retention time of 60 d [41].

However, electrochemical treatment of activated sludge was found to be more effective. Some authors reported that the biogas production rate exceeded 500 mL/g VS [42].

4. Conclusion

The present study explains the efficiency of sludge disintegration by a Fenton-microwave pretreatment method via a response surface methodology.

The effect of various operational parameters on tEPS and sCOD was investigated. The best optimization of sludge disintegration obtained with FMT was sCOD value of 2,045.3 mg/L, a maximum tEPS value of 7,155.6 mg/L with a target specific energy of 50,000 kJ/kg ST, under optimal operating conditions of 800 W, 89.7 s and 65 mg/g TS of H_2O_2.

Experimental results were close to predicted results. These prove that the models are globally validated but need to be more improved. RSM and experimental results proved that FMT was an efficient tool for enhancing sludge disintegration. Biogas production assay shows that the combined pretreatment has improved sludge digestibility compared to untreated sludge.

Acknowledgments

This research and innovative work has been carried out within the framework of the MOBIDOC device financed by the European Union within the framework of the EMORI program and administered by the ANPR.

References

[1] E. Ardern, W.T. Lockett, Experiments on the oxidation of sewage without the aid of filters, J. Chem. Technol. Biotechnol., 33 (1914) 523–539.
[2] S. Kavitha, J.R. Banu, J.V. Kumar, M. Rajkumar, Improving the biogas production performance of municipal waste activated sludge via disperser induced microwave disintegration, Bioresour. Technol., 217 (2016) 21–27.
[3] S. Pullket, Sewage Sludge as Source of Activated Carbon for the Removal of Endocrine Disrupting Chemicals in Wastewater, Thesis, Imperial College London, South Kensington, London, 2015.
[4] J. Choudhary, B. Kumar, A. Gupta, Feasible utilization of waste limestone sludge as filler in bituminous concrete, Constr. Build. Mater., 239 (2020) 117781, doi: 10.1016/j.conbuildmat.2019.117781.
[5] H. Hudcová, J. Vymazal, M. Rozkošný, Present restrictions of sewage sludge application in agriculture within the European Union, Soil Water Res., 14 (2019) 104–120.

[6] S. Maamri, M. Amrani, Biogas production from waste activated sludge using cattle dung inoculums: effect of total solid contents and kinetics study, Energy Procedia, 50 (2014) 352–359.
[7] A.P. Eswari, S. Kavitha, J.R. Banu, O.P. Karthikeyan, I.T. Yeom, H_2O_2 induced cost effective microwave disintegration of dairy waste activated sludge in acidic environment for efficient biomethane generation, Bioresour. Technol., 244 (2017) 688–697.
[8] C. Sun, R. Liu, W. Cao, K. Li, L. Wu, Optimization of sodium hydroxide pretreatment conditions to improve biogas production from asparagus stover, Waste Biomass Valorization, 10 (2019) 121–129.
[9] H. Junoh, C.H. Yip, P. Kumaran, Effect on $Ca(OH)_2$ Pretreatment to Enhance Biogas Production of Organic Food Waste, IOP Conference Series: Earth and Environmental Science, Volume 32, International Conference on Advances in Renewable Energy and Technologies (ICARET 2016) 23–25 February 2016, Putrajaya, Malaysia, 32 (2016) 012013.
[10] X. Liu, X. Huang, Y. Wu, Q. Xu, M. Du, D. Wang, Q. Yang, Y. Liu, B.-J. Ni, G. Yang, F. Yang, Q. Wang, Activation of nitrite by freezing process for anaerobic digestion enhancement of waste activated sludge: performance and mechanisms, Chem. Eng. J., 387 (2020) 124147, doi: 10.1016/j.cej.2020.124147.
[11] J. Oladejo, K. Shi, X. Luo, G. Yang, T. Wu, A review of sludge-to-energy recovery methods, Energies, 12 (2019) 60, doi: 10.3390/en12010060.
[12] S. Pilli, S. Yan, R.D. Tyagi, R.Y. Surampalli, Overview of Fenton pre-treatment of sludge aiming to enhance anaerobic digestion, Rev. Environ. Sci. Biotechnol., 14 (2015) 453–472.
[13] K. Michalska, K. Miazek, L. Krzystek, S.Ledakowicz, Influence of pretreatment with Fenton's reagent on biogas production and methane yield from lignocellulosic biomass, Bioresour. Technol., 119 (2012) 72–78.
[14] Z. Hanjie, Sludge treatment to increase biogas production,Skolan för arkitektur och samhällsbyggnad, Kungliga Tekniska högskolan, 2010.
[15] D.P. Rufus, R. Banu, D.D. Nguyen, G. Kumar, S.W. Chang, Effect of dispersion treatment on dairy waste activated sludge to hasten the production of biogas, Front. Energy Res., 7 (2019) 136, doi: 10.3389/fenrg.2019.00136.
[16] D.A. Jones, T.P. Lelyveld, S.D. Mavrofidis, S.W. Kingman, N.J. Miles, Microwave heating applications in environmental engineering—a review, Resour. Conserv. Recycl., 34 (2002) 75–90.
[17] V.K. Tyagi, S.L. Lo, Microwave irradiation: a sustainable way for sludge treatment and resource recovery, Renewable Sustainable Energy Rev., 18 (2013) 288–305.
[18] J.H. Jang, J.H. Ahn, Effect of microwave pretreatment in presence of NaOH on mesophilic anaerobic digestion of thickened waste activated sludge, Bioresour. Technol., 131 (2013) 437–442.
[19] J.R. Banu, R.Y. Kannah, S. Kavitha, M. Gunasekaran, G. Kumar, Novel insights into scalability of biosurfactant combined microwave disintegration of sludge at alkali pH for achieving profitable bioenergy recovery and net profit, Bioresour. Technol., 267 (2018) 281–290.
[20] A. Zaker, Z. Chen, X. Wang, Q. Zhang, Microwave-assisted pyrolysis of sewage sludge: a review, Fuel Process. Technol., 187 (2019) 84–104.
[21] M.H. Zhang, H. Dong, L. Zhao, D.X. Wang, D. Meng, A review on Fenton process for organic wastewater treatment based on optimization perspective, Sci. Total Environ., 670 (2019) 110–121.
[22] J.R. Banu, R.Y. Kannah, S. Kavitha, M.Gunasekaran, I.T. Yeom, G. Kumar, Disperser-induced bacterial disintegration of partially digested anaerobic sludge for efficient biomethane recovery, Chem. Eng. J., 347 (2018) 165–172.
[23] X. Qi, Z. Li, Efficiency optimization of a microwave-assisted Fenton-like process for the pretreatment of chemical synthetic pharmaceutical wastewater, Desal. Water Treat., 57 (2016) 11756–11764.
[24] N. Barrak, R. Mannai, M. Zaidi, M. Kechida, A.N. Helal, Experimental design approach with response surface methodology for removal of indigo dye by electrocoagulation, J. Geosci. Environ. Prot., 4 (2016) 50–61.
[25] T. Yu, Y. Deng, H. Liu, C. Yang, B. Wu, G. Zeng, F. Nishimura, Effect of alkaline microwaving pretreatment on anaerobic digestion, biogas production of swine manure, Sci. Rep., 7 (2017) 1–8.
[26] G.E. Box, J.S. Hunter, W.G. Hunter, Statistics for Experimenters: deSign, Innovation, and Discovery, Vol. 2, Wiley-Interscience, New York, 2005.
[27] J.M. Morgan, C.F. Forster, L. Evison, A comparative study of the nature of biopolymers extracted from anaerobic and activated sludges, Water Res., 24 (1990) 743–750.
[28] C.H. Pham, J.M. Triolo, T.T.T. Cu, L. Pedersen, S.G. Sommer, Validation and recommendation of methods to measure biogas production potential of animal manure, Asian-Australas. J. Anim. Sci., 26 (2013) 864, doi: doi: 10.5713/ajas.2012.12623.
[29] M.S. Lucas, J.A. Peres, Removal of COD from olive mill wastewater by Fenton's reagent: kinetic study, J. Hazard. Mater., 168 (2009) 1253–1259.
[30] D.H. Kim, E. Jeong, S.E. Oh, H.S. Shin, Combined (alkaline + ultrasonic) pretreatment effect on sewage sludge disintegration, Water Res., 44 (2010) 3093–3100.
[31] K. Izumi, Y.K. Okishio, N. Nagao, C. Niwa, S. Yamamoto, T. Toda, Effects of particle size on anaerobic digestion of food waste, Int. Biodeterior. Biodegrad., 64 (2010) 601–608.
[32] S. Şahinkaya, E. Kalıpcı, S. Aras, Disintegration of waste activated sludge by different applications of Fenton process, Process Saf. Environ. Prot., 93 (2015) 274–281.
[33] C. Eskicioglu, K.L. Kennedy, R.L. Droste, Enhancement of batch waste activated sludge digestion by microwave pretreatment, Water Environ Res., 79 (2007) 2304–2317.
[34] L. Peng, L. Appels, H. Su, Combining microwave irradiation with sodium citrate addition improves the pre-treatment on anaerobic digestion of excess sewage sludge, J. Environ. Manage., 213 (2018) 271–278.
[35] I. Doğan, F.D. Sanin, Alkaline solubilization and microwave irradiation as a combined sludge disintegration and minimization method, Water Res., 43 (2009) 2139–2148.
[36] I. Lee, J.I. Han, The effects of waste-activated sludge pretreatment using hydrodynamic cavitation for methane production, Ultrason. Sonochem., 20 (2013) 1450–1455.
[37] Q. Yang, J. Yi, K. Luo, X. Jing, X. Li, Y. Liu, G. Zeng, Improving disintegration and acidification of waste activated sludge by combined alkaline and microwave pretreatment, Process Saf. Environ. Prot., 91 (2013) 521–526.
[38] Y.C Bozkurt, O.G. Apul, Critical review for microwave pretreatment of waste-activated sludge prior to anaerobic digestion, Curr. Opin. Environ. Sustainability, 14 (2020) 1–9.
[39] C. Eskicioglu, A. Prorot, J. Marin, R.L. Droste, K. J Kennedy, Synergetic pretreatment of sewage sludge by microwave irradiation in presence of H_2O_2 for enhanced anaerobic digestion, Water Res., 42 (2008) 4674–4682.
[40] S. Şahinkaya, M.F. Sevimli, Sono-thermal pre-treatment of waste activated sludge before anaerobic digestion, Ultrason. Sonochem., 20 (2013) 587–594.
[41] A. Gonzalez, H. Guo, O. Ortega-Ibáñez, C. Petri, J.B. van Lier, M. de Kreuk, A. Hendriks, Mild thermal pre-treatment of waste activated sludge to increase loading capacity, biogas production, and solids' degradation: a pilot-scale study, Energies, 13 (2020) 6059, doi: 10.3390/en13226059.
[42] B. Yu, J. Xu, H. Yuan, Z. Lou, J. Lin, N. Zhu, Enhancement of anaerobic digestion of waste activated sludge by electrochemical pretreatment, Fuel, 130 (2014) 279–285.