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Study on adsorption of octenylsuccinate by sepiolite

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ABSTRACT

Sepiolite was selected and studied for the adsorption of octenylsuccinate in the waste solution resulting from the production of octenylsuccinic starch. Scanning electron microscopy showed that sepiolite had a high surface area, and Fourier transform infrared spectroscopy indicated the formation of bonds between sepiolite and octenylsuccinate. The effects of reaction temperature, initial octenylsuccinate concentration, pH, adsorption time, and sepiolite dosage on adsorption efficiency (%AE) were examined by single-factor experiments. Significant parameters were further optimized by Box–Behnken response surface methodology. The theoretical optimum value of 100% was obtained when the initial octenylsuccinate concentration, pH, adsorption time, and sepiolite dosage of 20 mL solution were 0.048 mol/L, 3.12, 5.40 h, and 771.32 mg, respectively. Under these optimum conditions, the real %AE was 99.07%.

Keywords: Sepiolite; Adsorption; Octenylsuccinate; Response surface methodology (RSM)

1. Introduction

Activated carbon is currently the most widely used adsorbent for a range of compounds in solution, but its high cost in replacement and regeneration is prohibitive of larger scale use in industry [1]. Currently, several mineral adsorbents such as sepiolite, zeolite [2], bentonite [3], tremolite amianthus [4] are considered to be efficient alternatives to activated carbon as economical adsorbents for the removal of hazardous substances from aqueous solutions [5–7]. Sepiolite is a natural clay mineral with a unit-cell formula $(Si_{12})(Mg_8)(O_{30})(OH)_4(OH)_2{\cdot}8H_2O$ [8]. Advantageous qualities of sepiolite include its micro-fibrous structure, giving a theoretically high surface area, along with high chemical and mechanical stability, contributing to strong adsorption capacity [9,10]. Due to its low cost and abundance [1], sepiolite is widely used as an adsorbent in wastewater treatment, such as adsorption of copper (II) ions [11] and removal of a textile dye [12].

Octenylsuccinic starch (OS starch) is a modified starch that has been altered chemically in order to impart amphiphilic properties on hydrophilic native starch [13]. OS starch is used as an emulsifier and colloidal stabilizer in products such as beverages and salad dressings [14]. Normally, OS starch is prepared by the esterification of granular native starch with octenylsuccinic anhydride (OSA, Fig. 1(a)) catalyzed by basic conditions, followed by filtering and washing from the system [15]. As a consequence, octenylsuccinic

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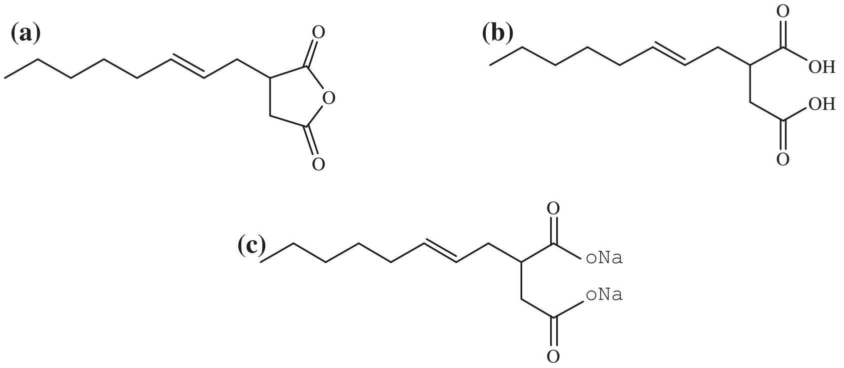

Fig. 1. Structures of OSA (a), octenylsuccinic acid (OS acid) (b), and octenylsuccinic sodium (OS-Na) (c).

acid (OS acid, Fig. 1(b)) and octenylsuccinic sodium (OS-Na, Fig. 1(c)) can remain in reaction solution after pH adjustment. In this study, OS acid and OS-Na are replaced by octenylsuccinate.

In order to achieve environmentally sustainable industrialized production, the octenylsuccinate in waste solution needs to be reclaimed. To our knowledge, however, octenylsuccinate is not readily biodegradable in the environment. Compared with conventional methods like biological fermentation [16,17], chemical adsorption would be a more economical choice and the chemical oxygen demand value of waste solution would also be effectively decreased. To achieve this, sepiolite after grinding and sieving was selected to adsorb octenylsuccinate [18].

Despite that sepiolite has been widely used in the removal of dyes and metal ions from wastewater [1], relatively few studies are concerning the adsorption of octenylsuccinate in waste solution. The objective of the present study was to investigate the adsorption efficiency (%AE) of sepiolite in removing octenylsuccinate from the waste solution of OS starch. Sepiolite was characterized by X-ray fluorescence (XRF), scanning electron microscope (SEM), and Fourier transform infrared (FT-IR). The effects of reaction temperature, initial octenylsuccinate concentration, pH, adsorption time, and sepiolite dosage were examined, and the % AE was evaluated and optimized by response surface methodology (RSM).

2. Materials and methods

2.1. Materials

The OSA reagent was supplied by Energy Chemical Co., Ltd. (Shanghai, China). Sepiolite was provided by Jiacheng nonmetal mining industry Co., Ltd. (Zhejiang, China). HPLC grade acetonitrile was purchased from SayFo Technology Co., Ltd. (Tianjing, China). Trifluoroacetic acid (TFA) was obtained from Sinopharm Chemical Reagent Co., Ltd. HPLC grade purified water was purchased from Wahaha Group Co., Ltd. (Hangzhou, China). Other chemicals were of analytical grade.

2.2. Characterization of sepiolite

The specific surface area of the sepiolite derived from N_2 adsorption isotherms was measured at liquid nitrogen temperature using a Micromeritics ASAP 2020 M instrument.

Particle-size analysis was done by laser diffractometry using a Particle Size Analyzer (Microtrac S3500, America) by suspending sepiolite sample in ethanol and decanting through the analyzer beam.

The chemical composition of the sepiolite sample was determined by XRF spectrometer (ZSX Primus II) after pelleting.

According to the previous studies [19,20], SEM (Hitachi S-4800) was performed at 5.0 kV to investigate the surface morphology of sepiolite. Samples were sputter-coated with a thin layer of carbon before microscopic observation.

The FT-IR spectra were collected with 2.0 mg of sample in KBr pellets using a Tensor 27 spectrometer (Bruker Optik GmbH, Ettlingen, Germany) applying a RT-DLaTGS internal detector in the region of 400–4,000 cm^{-1}, with a resolution of 4 cm^{-1}. The mass of each sample was 2.0 mg.

2.3. Adsorption experiments

An octenylsuccinate solution was applied to simulate the waste solution of OS starch. OSA (1~3 g) was weighed into a 500 mL brown bottle, which contained 200 mL of distilled water. The solution was stirred overnight with a magnetic stirring bar to ensure complete hydrolysis of OSA. Sodium hydroxide solution (1 N) and hydrochloric acid (1 N) were used to adjust the pH of the solution at 3.0~7.0 by a pH meter (Model ZD-2, Shanghai INESA and Scientific Instrument Co., Ltd). After the pH was stable for 30 min, the octenylsuccinate solution could be used for adsorption.

Sepiolite was dried at 150℃ in an oven for 2 h, and then cooled and collected in a bottle. For adsorption, sepiolite (300~900 mg) was weighed accurately in the sampling bottle to adsorb 20 mL of the octenylsuccinate solution at different concentrations and pH values. The sampling bottle was placed in a constant temperature oscillator (Model DKZ-2, Shanghai Jinghong Laboratory Equipment Co., IL) during the adsorption.

Reaction temperature, initial concentration, pH, adsorption time, and sepiolite dosage were considered to be the important parameters of adsorption behavior, according to the literature [8].

To compare different temperatures, OSA (2 g) was dissolved with 200 mL distilled water (the concentration was 0.048 mol/L). The pH was controlled at 4.0, and then the solution (20 mL) was absorbed by 600 mg sepiolite. During the adsorption (4 h), the solution was heated at 25, 30, 35, 40, and 45℃, respectively.

To compare different concentrations of octenylsuccinate, 0.024, 0.036, 0.048, 0.060, and 0.072 mol/L octenylsuccinate solutions were prepared by dissolving different weights of OSA (1.0, 1.5, 2, 2.5, and 3 g) in 200 mL distilled water. The different octenylsuccinate solutions (20 mL) were absorbed by 600 mg sepiolite for 4 h at pH 4.0 in 35℃ water bath.

In order to compare different pH, the pH of octenylsuccinate solution (0.048 mol/L) was controlled at 3.0, 4.0, 5.0, 6.0, and 7.0, respectively. Then the octenylsuccinate solution (20 mL) was absorbed by 600 mg sepiolite at 35℃ for 4 h.

To compare different adsorption times, the pH of octenylsuccinate solution (0.048 mol/L) was maintained at 4.0. Then the solution (20 mL) was absorbed by 600 mg sepiolite at 35℃ for different time (2.0, 3.0, 4.0, 5.0, and 6.0 h).

In order to compare different sepiolite dosages, octenylsuccinate solution (0.048 mol/L) was prepared and the pH was controlled at 4.0. The solutions were absorbed by 300, 450, 600, 750, and 900 mg sepiolite, respectively. During the adsorption (4 h), the solution was heated at 35℃.

For the RSM experiments, OSA (1, 2, and 3 g) was dissolved with 200 mL distilled water (the concentrations were 0.024, 0.048, and 0.072 mol/L). The pH was adjusted to 3.0, 4.0, and 5.0, respectively. The solution (20 mL) was absorbed by sepiolite (300, 600, and 900 mg) at 35℃ for various times (2, 4, and 6 h). Different concentrations of octenylsuccinate after adsorption were obtained and measured to study the adsorption of sepiolite.

2.4. High-performance liquid chromatography (HPLC) analysis

The HPLC system was a Dionex Ultimate 3000 Series (DU, Sunnyvale, California) consisting of a LPG-3400A pump, an automatic injector with a 20 μL loop, and a diode array detector (DAD-3000) equipped with a work station computer. An Extend C_{18} column (5 μm, 250 × 4.6 mm; Aglient) was employed at 30℃. The HPLC conditions were the same as our previous work [21]. The HPLC standard curve of octenylsuccinate in the range of 0.08–2 μg octenylsuccinate was employed in Eq. (1):

$$Y = 1.756X + 18.911 \tag{1}$$

where Y was total peak area (mAU*s), and X was octenylsuccinate content (mol). The correlation coefficient value of the equation was 0.9996, indicating the best-fitting curve.

2.5. Determination of octenylsuccinate by HPLC

The determination of octenylsuccinate referred to the previous method with some modifications [21].

The octenylsuccinate solution (20 mL) was prepared for adsorption. After adsorption, the solution was centrifuged and 5 mL of supernatant was collected and pipetted into a 50 mL volumetric flask, filling with water (pH 3, prepared by distilled water with 1.0 mol/L HCl). The solution was then analyzed by HPLC, yielding total peak area (Y) and the corresponding weight of octenylsuccinate (X, mol) by Eq. (1).

The adsorption efficiency (%AE) of octenylsuccinate on sepiolite was calculated by Eqs. (2) and (3):

$$C = \frac{10^4 X}{20 \times 10^{-3}} \tag{2}$$

$$\%\mathrm{AE} = \frac{C_o - C}{C_o} \times 100\% \tag{3}$$

where C was the concentration of octenylsuccinate after adsorption (mol/L), C_o was the initial concentration of octenylsuccinate (mol/L), 10^4 was the dilution factor,

20×10^{-3} was the volume of the octenylsuccinate solution (L).

2.6. Experimental design for response methodology

Key parameters were identified based on the results of single-factor experiments. Levels of the key parameters, including initial octenylsuccinate concentration, pH, adsorption time, sepiolite dosage, and interactions between variables, were analyzed and optimized by Box-Behnken RSM with four factors and three levels, including three central points. The high, middle, and low levels of each variable were coded as +1, 0, and −1, respectively [22–24]. Experimental design was shown in Table 1. *A*, *B*, *C*, and *D* were the coded values for concentration, pH, time, and sepiolite dosage, respectively.

The effectiveness of this model was evaluated by the coefficient of determination (R^2) and analysis of variance (ANOVA).

2.7. Statistical analysis

Experiments were performed in triplicate. Data were analyzed by an ANOVA procedure using software Design-Expert (Version 8.0.5, State-Ease Inc., Minneapolis, Minnesota, USA). Tukey's studentized range (HSD) was done by SAS version 9.2 (SAS Institute, Inc., Cary, NC).

3. Results and discussion

3.1. Characterization results of sepiolite

The BET surface area was measured to be 44.74 m^2/g from N_2 adsorption isotherms. Besides, the sepiolite sample yielded an average particle size (D_{v50}) of 7.86 μm.

XRF was performed to determine the chemical composition of the sepiolite, which was listed in Table 2. It showed a high proportion of SiO_2 and MgO while other minerals were present in trace amounts.

As seen in Fig. 2, sepiolite had a fibrous shape and polyporous surface which indicated high surface areas. Hence, sepiolite offered a potential for adsorption of octenylsuccinate from the washing solution of OS starch [25].

Fig. 3 shows the FT-IR spectra of natural sepiolite (a), sepiolite after adsorption (b), and OSA (c). Sepiolite (771.32 mg) adsorbed 20 mL of the octenylsuccinate solution under optimum conditions of initial octenylsuccinate concentration 0.050 mol/L, pH 3.12, adsorption time 5.40 h, followed by vacuum freeze-drying to get the sepiolite after adsorption. As shown in Fig. 3(a), bands at 3,693 cm^{-1} were corresponding to the vibrations of the Mg-OH group [20]. Bands at 1,433, 1,016, and 460 cm^{-1} originated from Si–O–Si vibration [7]. In comparison with natural sepiolite (a), sepiolite after adsorption (b) had five additional bands. The bands occurring at 2,938, 2,925, and 2,856 cm^{-1} were due to the C–H stretching vibration of chain methylene ($-CH_2-$) [26], supporting the intercalation of the side chain of OSA (c). Carboxylate bands occurring at 1,560 and 1,458 cm^{-1} were indicative of the new chemical bond formation of octenylsuccinate and sepiolite, showing the typical chemical adsorption of octenylsuccinate [27].

3.2. Factors affecting adsorption

3.2.1. Effect of temperature on adsorption process

Fig. 4(a) shows that there are few changes on %AE at the temperatures from 25 to 40℃, and %AE was lower in 45 than in 40℃. Tsai et al. found that the higher temperature could weaken the attractive force between adsorbate and adsorbent [8,28,29]. However, there was no significant effect on %AE in the temperature range of 25–45℃ in this study (Values are not significantly different ($p < 0.05$)). Considering the convenience of experimental operation, 35℃ was selected as the following experimental condition.

3.2.2. Effect of initial octenylsuccinate concentration on adsorption process

As shown in Fig. 4(b), when the initial octenylsuccinate concentration increased from 0.024 to

Table 1
Factors and levels of Box-Behnken experiment design

	Factor			
Level	Concentration *A* (mol/L)	pH *B*	Time *C* (h)	Sepiolite dosage *D* (mg)
−1	0.024	3.0	2.0	300
0	0.048	4.0	4.0	600
1	0.072	5.0	6.0	900

Table 2
Chemical analysis of the sepiolite sample

Compound	SiO_2	MgO	Al_2O_3	Fe_2O_3	CaO	K_2O	Na_2O	P_2O_5	SO_3	SrO	Cl	LOI*
wt%	54.800	18.600	0.223	0.056	0.740	0.049	0.024	0.021	0.011	0.010	0.008	25.457

*Lost on ignition.

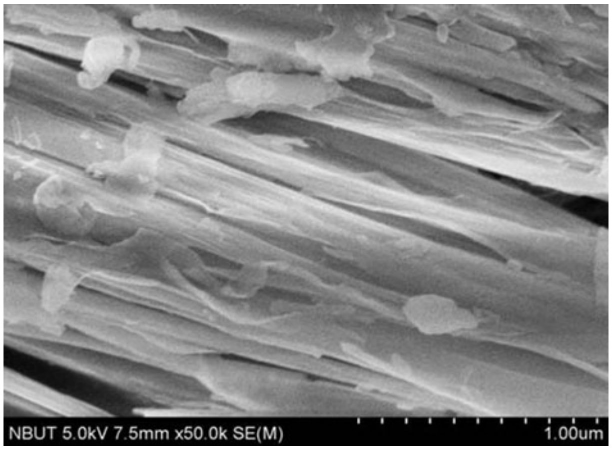

Fig. 2. SEM image of sepiolite.

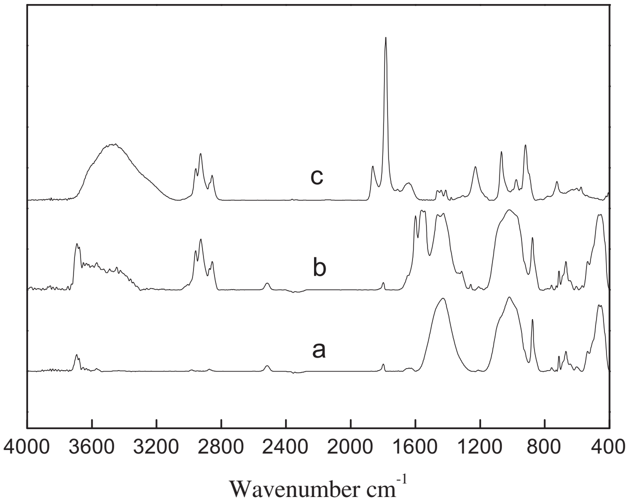

Fig. 3. FT-IR images of natural sepiolite (a), sepiolite after adsorption (b), and OSA (c).

0.060 mol/L, the %AE remained stable in the range of 94~96%, suggesting that the unsaturated adsorption sites kept the high %AE. However, the %AE decreased to 85% when the concentration was 0.072 mol/L. The sharp drop of %AE could be explained that an excessive of initial octenylsuccinate concentration increases the saturation of adsorption sites during the adsorption process [30]. The corresponding adsorption capacity q (mol/g), the amount of octenylsuccinate adsorbed on sepiolite, increases from 7.59×10^{-4}~2.00×10^{-3} mol/g with the change of the initial octenylsuccinate concentration (0.024~0.072 mol/L).

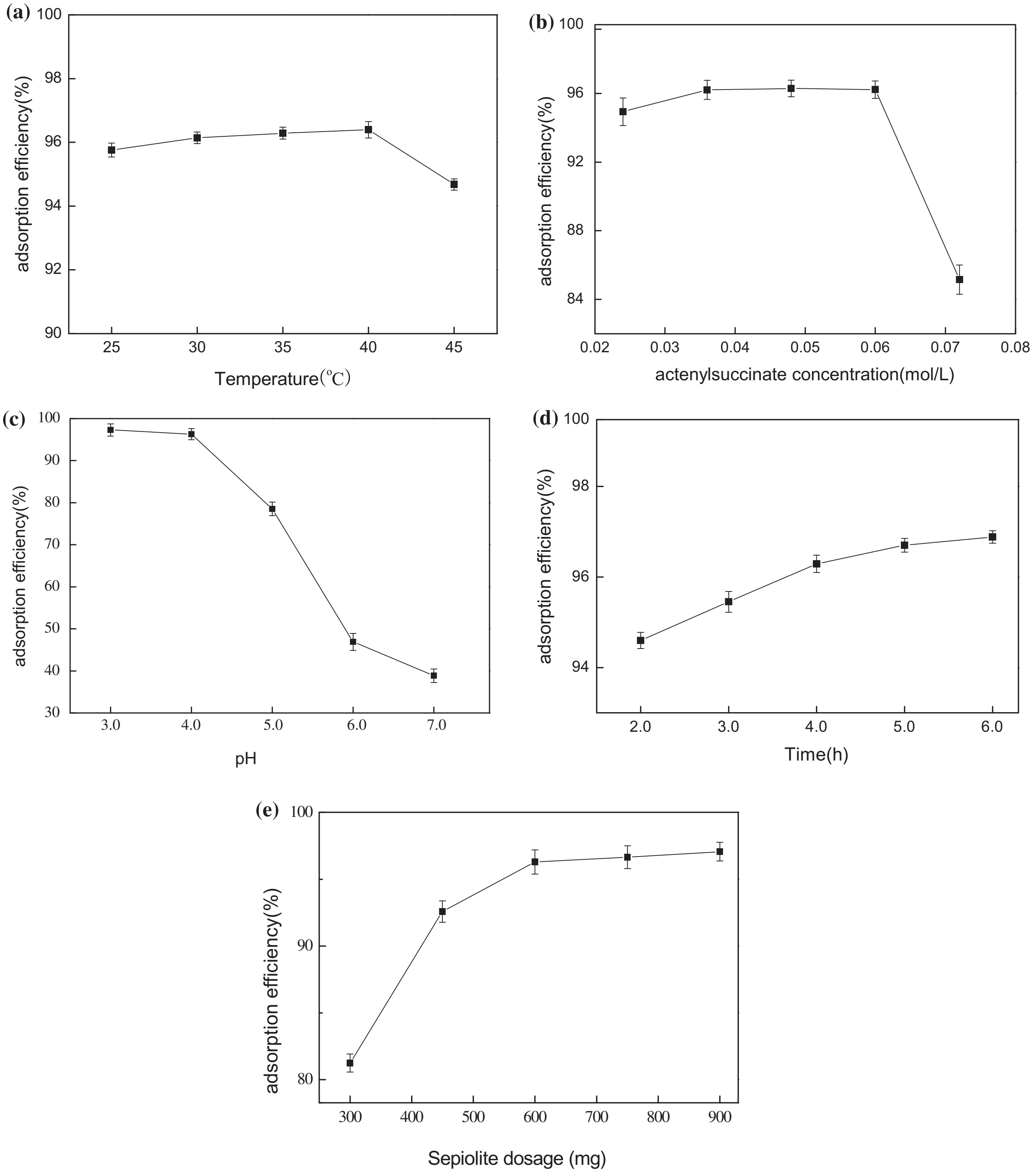

Fig. 4. Effects of reaction temperature (a), initial octenylsuccinate concentration (b), pH (c), adsorption time (d), and sepiolite dosage (e) on adsorption efficiency (%AE).

3.2.3. Effect of pH on adsorption process

Adsorption of octenylsuccinate onto sepiolite was investigated in the range of pH 3–7. Due to the availability restraint of Extend C_{18} column, pH that was lower than 3 was not involved. The %AE of sepiolite decreased with increasing pH and the optimum pH was 3 (Fig. 4(c)). In previous study, Uğurlu et al. reported that hydrogen ions (H^+) in the solution changed the specific surface charge of activated sepiolite [12]. Under these experimental conditions, more H^+ will be available at lower pH. The surface of sepiolite becomes more positively charged, thereby increasing the electrostatic attractions between negatively charged octenylsuccinic ions and positively charged adsorption sites, which lead to an increase in the octenylsuccinate adsorption [12,15,31,32]. With the gradual increase of pH, less positive charge develops on the surface of sepiolite, resulting in electrostatic repulsion between negatively charged sepiolite surface and negatively charged octenylsuccinic ions. Thus, the increase in concentration of H^+ (low pH) contributes to the increase in %AE of sepiolite.

3.2.4. Effect of adsorption time on adsorption process

Fig. 4(d) shows that an increase in time (2~6 h) resulted in a slight increase in the %AE (94.5~97.0%). The maximum %AE of sepiolite was observed after

Table 3
Box-Behnken experimental design matrix

No.	A	B	C	D	%AE
1	−1	−1	0	0	98.08
2	−1	1	0	0	84.43
3	1	−1	0	0	95.05
4	1	1	0	0	51.68
5	0	0	−1	−1	69.90
6	0	0	−1	1	95.08
7	0	0	1	−1	85.28
8	0	0	1	1	93.08
9	−1	0	0	−1	97.06
10	−1	0	0	1	97.81
11	1	0	0	−1	46.86
12	1	0	0	1	95.69
13	0	−1	−1	0	98.11
14	0	−1	1	0	97.15
15	0	1	−1	0	56.54
16	0	1	1	0	59.72
17	−1	0	−1	0	97.98
18	−1	0	1	0	97.57
19	1	0	−1	0	71.90
20	1	0	1	0	89.13
21	0	−1	0	−1	75.48
22	0	−1	0	1	98.20
23	0	1	0	−1	57.86
24	0	1	0	1	59.08
25	0	0	0	0	90.88
26	0	0	0	0	89.41
27	0	0	0	0	88.32

Table 4
ANOVA for quadratic model

Source	Sum of squares	df	Mean square	*F* value	Prob > *F*	Significance
Model	7,256.82	14	518.34	20.86	<0.0001	Significant
A	1,252.97	1	1,252.97	50.43	<0.0001	Significant
B	3,096.37	1	3,096.37	124.63	<0.0001	Significant
C	87.59	1	87.59	3.53	0.0849	
D	945.19	1	945.19	38.04	<0.0001	Significant
AB	220.82	1	220.82	8.89	0.0115	Significant
AC	77.79	1	77.79	3.13	0.1022	
AD	577.92	1	577.92	23.26	0.0004	Significant
BC	4.28	1	4.28	0.17	0.6853	
BD	115.56	1	115.56	4.65	0.0520	
CD	75.52	1	75.52	3.04	0.1068	
A^2	6.53	1	6.53	0.26	0.6174	
B^2	574.18	1	574.18	23.11	0.0004	Significant
C^2	0.72	1	0.72	0.029	0.8673	
D^2	154.13	1	154.13	6.20	0.0284	Significant
Residual	298.13	12	24.84			
Lack-of-fit	294.83	10	29.48	17.86	0.0541	Not significant
Pure error	3.30	2	1.65			
Cor total	7,554.95	26				

Notes: Values of "Prob > F" less than 0.050 indicated that model terms were significant. C.V. %: 6.02; *R*-squared: 0.9605; adjusted *R*-squared: 0.9145; Pred *R*-squared: 0.7742.

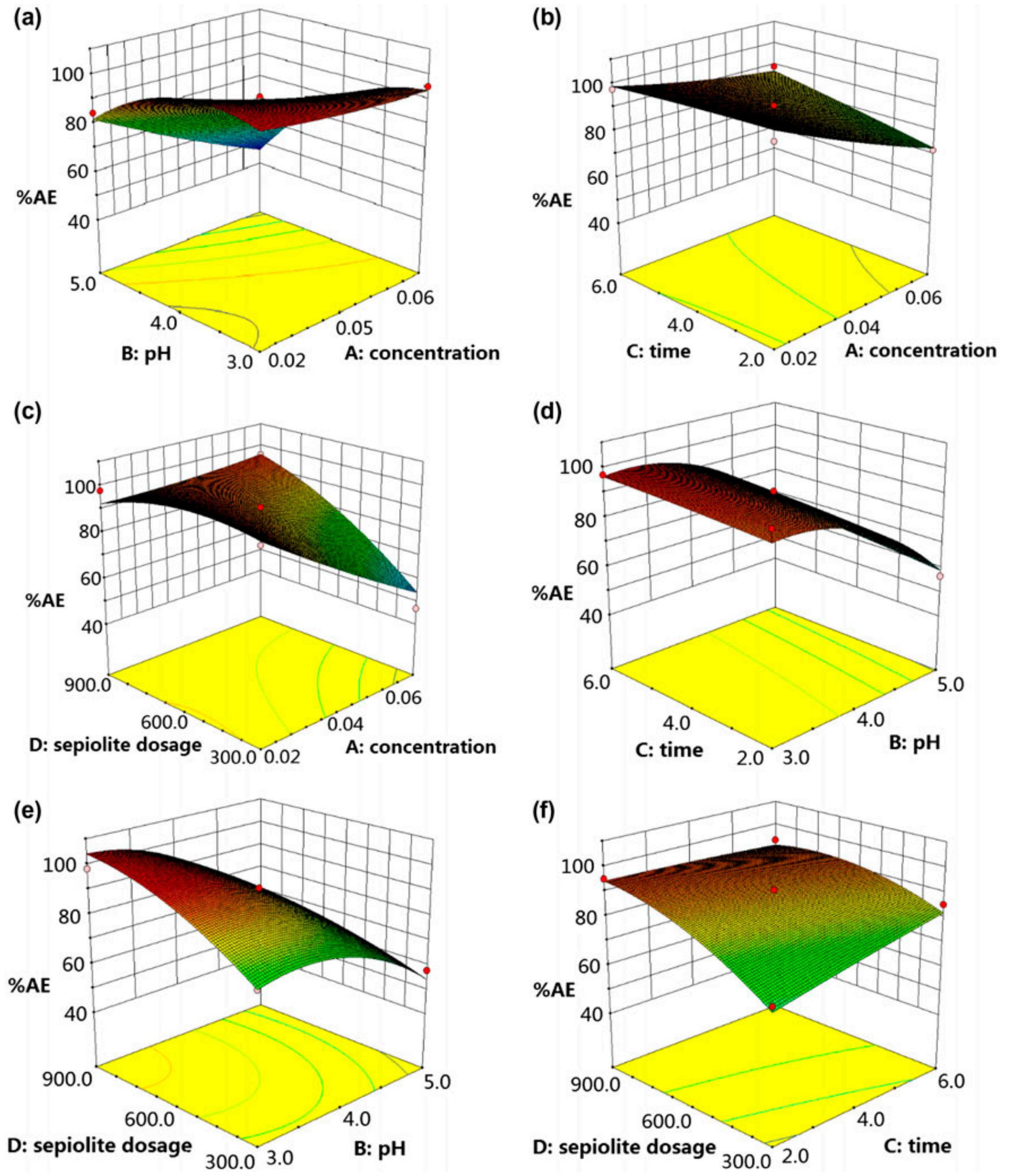

Fig. 5. Response surface plots of various parameters on adsorption efficiency (%AE): effect of concentration and pH on %AE (a); effect of concentration and time on %AE (b); effect of concentration and sepiolite dosage on %AE (c); effect of pH and time on %AE (d); effect of pH and sepiolite dosage on %AE (e); effect of time and sepiolite dosage on %AE (f).

5 h, beyond which there was no further increase in adsorption. The point corresponding to the maximum adsorption capacity was defined as the adsorption equilibrium time [33,34]. Doğan [9] reported the strong chemical binding of methylene blue dyes to sepiolite required a long contact time for the attainment of equilibrium, which supports the current finding that adsorption of octenylsuccinate by sepiolite also requires extended contact time in order to achieve a high %AE.

3.2.5. Effect of the sepiolite dosage on adsorption process

Fig. 4(e) presents the effect of the amount of sepiolite on the octenylsuccinate removal. As seen in Fig. 4(e), the %AE increased from 82 to 96% with increasing sepiolite dosage from 300 to 600 mg. It is because the number of available adsorption sites remains unsaturated due to the increase in adsorbent dosage, which boosts the %AE [30]. However, the %AE had no significant increase and tended to be stable in 96% within the range of sepiolite dosages (600~900 mg). One important reason is that an increase in sepiolite dosage decreases the adsorption density during the adsorption process, thereby enhancing the electrostatic interactions [11]. Another reason may be ascribed to the aggregation of sepiolite particles when the adsorbent concentration is high, resulting in a decrease in total surface area of adsorbent (sepiolite).

3.3. Design and results of tests analysis

Experimental results of RSM were shown in Table 3. The software Design-Expert was used to get the regression equation of %AE in Eq. (4):

$$\begin{aligned}\%\,\mathrm{AE} = {} & 89.54 - 10.22A - 16.06B + 2.70C + 8.88D \\ & - 7.43AB + 4.41AC + 12.02AD + 1.03BC \\ & - 5.37BD - 4.35CD + 1.11A^2 - 10.38B^2 \\ & - 0.37C^2 - 5.38D^2 \end{aligned} \tag{4}$$

where %AE was the response value, and A, B, C, D were the coded values of initial octenylsuccinate concentration, pH, adsorption time, sepiolite dosage, respectively.

The significance of quadratic model was analyzed by ANOVA. The results were given in Table 4.

The fitness of the model was checked by the coefficient of determination (R^2). The higher R^2 value indicated the aptness of the model [35]. The statistical significances of independent variables and their interactions were evaluated based on p-values. The model of the p-value was smaller than 0.0001, pointing to the accuracy of the model. Meanwhile, the R^2 for the response was 0.9605 which showed only 3.95% of the total variations could not be explained by the model. In addition, the value of lack-of-fit was 17.86 and the corresponding p-value was 0.0541 (>0.05), indicating that lack-of-fit was not significant. Therefore, all the results showed that the model had a good-fit of the experiment.

A (initial octenylsuccinate concentration), B (pH), and D (sepiolite dosage) were significant factors to the adsorption process of sepiolite (p-value < 0.0001). However, the p-value of time was 0.0849 (>0.05) which implied the time was not a significant factor. Besides, there were significant interaction between A and B (initial octenylsuccinate concentration and pH), A and D (initial octenylsuccinate concentration and sepiolite dosage) whose p-value was 0.0115 (<0.05) and 0.0004 (<0.05). B^2 (the square of pH) and D^2 (the square of sepiolite dosage) were also significant model terms ($p < 0.05$).

3.4. Analysis of responsive surface plots

The optimum value of various parameters was represented in the three-dimensional surface plots. The %AE of sepiolite altered significantly by changing in pH from 3 to 7. In Fig. 5(a, d, e), it decreased with the increase in pH. The maximum efficiency was obtained at the lowest pH (pH 3) and the minimum efficiency occurred at the highest pH (pH 5). For sepiolite dosage, %AE increased with increasing sepiolite, which was shown in Fig. 5(c, e, f). It demonstrated that the higher amount of sepiolite facilitated the adsorption behavior ranging from 300 to 900 mg. In Fig. 5((a)–(c)), increasing the concentration was followed by the decrease in %AE, indicating that the concentrated solution was not beneficial for the %AE. In Fig. 5(b, d, f), time had no significant impact on the %AE. In summary, the concentration, pH, and sepiolite dosage were the significant parameters of %AE of sepiolite.

According to the software Design-Expert, maximum %AE (100%) was obtained with initial octenylsuccinate concentration of 0.050 mol/L, pH of 3.12, time of 5.40 h and sepiolite dosage of 771.32 mg. Under these optimum conditions, the real %AE was 99.07%, indicating effective optimization.

4. Conclusions

Sepiolite had a considerable potential to remove octenylsuccinate in adsorption system as an adsorbent because of its high surface area, which was confirmed by high specific surface area (44.74 m^2/g) and SEM analysis. FT-IR spectroscopy showed that sepiolite had five additional bands after adsorption, indicating that new chemical bond formation of octenylsuccinate and sepiolite.

The %AE of the adsorption of octenylsuccinate onto sepiolite was investigated at different temperatures, initial octenylsuccinate concentrations, pHs, adsorption times, and sepiolite dosages. Based on the single-factor experiments, selected conditions were optimized by RSM, and a well-fitted mathematical model was established. At 35˚C, the optimum conditions were determined as follows: initial octenylsuccinate concentration of 0.050 mol/L, pH of 3.12, adsorption time of 5.40 h, and sepiolite dosage of 771.32 mg, respectively. The real %AE was 99.07%, which was close to the theoretical value (100%).

In summary, sepiolite was successfully used as a low-cost material for the removal of octenylsuccinate from the de facto waste solution of OS starch. However, the adsorption model and reclaiming of octenylsuccinate in waste solution requires further researches.

Supplementary material

The supplementary material for this paper is available online at http://dx.doi.10.1080/19443994.2015.1026277.

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References

[1] S. Çoruh, F. Geyikçi, S. Elevli, Adsorption of neutral red dye from an aqueous solution onto natural sepiolite using full factorial design, Clay. Clay. Miner. 59(6) (2011) 617–625.

[2] B. Armağan, M. Turan, M.S. Çelik, Equilibrium studies on the adsorption of reactive azo dyes into zeolite, Desalination 170(1) (2004) 33–39.

[3] A.S. Özcan, A. Özcan, Adsorption of acid dyes from aqueous solutions onto acid-activated bentonite, J. Colloid Interface Sci. 276(1) (2004) 39–46.

[4] G. Peng, L.J. Wan, L.X. Wang, Removal of Pb(II) from aqueous solution using tremolite amianthus, Asian. J. Chem. 24(8) (2012) 3457–3460.

[5] Q.Y. Sun, L.Z. Yang, The adsorption of basic dyes from aqueous solution on modified peat-resin particle, Water Res. 37(7) (2003) 1535–1544.

[6] S. Çoruh, F. Geyikçi, U. Çoruh, Removal of Cu^{2+} from copper flotation waste leachant using sepiolite: Full factorial design approach, Acta Geodyn. Geomater. 10 (4) (2013) 453–458.

[7] S. Lazarević, I. Janković-Častvan, D. Jovanović, S. Milonjić, D. Janaćković, R. Petrović, Adsorption of Pb^{2+}, Cd^{2+} and Sr^{2+} ions onto natural and acid-activated sepiolites, Appl. Clay. Sci. 37(1–2) (2007) 47–57.

[8] A. Özcan, E.M. Öncü, A.S. Özcan, Adsorption of acid blue 193 from aqueous solutions onto DEDMA-sepiolite, J. Hazard. Mater. 129(1) (2006) 244–252.

[9] M. Doğan, Y. Özdemir, M. Alkan, Adsorption kinetics and mechanism of cationic methyl violet and methylene blue dyes onto sepiolite, Dyes. Pigments 75(3) (2007) 701–713.

[10] O. Özdemir, M. Çınar, E. Sabah, F. Arslan, M.S. Çelik, Adsorption of anionic surfactants onto sepiolite, J. Hazard. Mater. 147(1–2) (2007) 625–632.

[11] S. Çoruh, F. Geyikçi, Adsorption of copper (II) ions on montmorillonite and sepiolite clays: Equilibrium and kinetic studies, Desal. Water Treat. 45 (2012) 351–360.

[12] M. Uğurlu, Adsorption of a textile dye onto activated sepiolite, Micropor. Mesopor. Mater. 119(1–3) (2009) 276–283.

[13] M.C. Sweedman, M.J. Tizzotti, C. Schäfer, R.G. Gilbert, Structure and physicochemical properties of octenyl succinic anhydride modified starches: A review, Carbohyd. Polym. 92(1) (2013) 905–920.

[14] S. Park, M.-G. Chung, B. Yoo, Effect of octenylsuccinylation on rheological properties of corn starch pastes, Starch-Stärke. 56(9) (2004) 399–406.

[15] S. Tesch, Ch Gerhards, H. Schubert, Stabilization of emulsions by OSA starches, J. Food Eng. 54(2) (2002) 167–174.

[16] G. Lofrano, S. Meriç, G.E. Zengin, D. Orhon, Chemical and biological treatment technologies for leather tannery chemicals and wastewaters: A review, Sci. Total Environ. 461–462 (2013) 265–281.

[17] H.-J. Chen, Y.-Z. Lin, J.-M. Fanjiang, C. Fan, Microbial community and treatment ability investigation in AOAO process for the optoelectronic wastewater treatment using PCR-DGGE biotechnology, Biodegradation 24(2) (2013) 227–243.

[18] D. Qiu, C.K. Tang, G. Peng and X.J. Fang, Adsorption treatment method for waste water containing octenyl succinate, CN 103130298 A, 2013.

[19] E.H. Duan, J. Han, Y. Song, Y.N. Guan, W.X. Zhao, B.B. Yang, B. Guo, Adsorption of styrene on the hydrothermal-modified sepiolite, Mater. Lett. 111 (2013) 150–153.

[20] Y. Qiu, S.M. Yu, Y.F. Song, Q. Wang, S.S. Zhong, W.X. Tian, Investigation of solution chemistry effects on sorption behavior of Sr(II) on sepiolite fibers, J. Mol. Liq. 180 (2013) 244–251.

[21] D. Qiu, Y.J. Bai, Y.C. Shi, Identification of isomers and determination of octenylsuccinate in modified starch by HPLC and mass spectrometry, J. Food Chem. 135 (2) (2012) 665–671.

[22] S. Çoruh, S. Elevli, Optimization of malachite green dye removal by sepiolite clay using a central composite design, J. Global Nest. 16(2) (2014) 339–347.

[23] J. Lu, X.M. Feng, Y.Q. Han, C.H. Xue, Optimization of subcritical fluid extraction of carotenoids and chlorophyll a from Laminaria japonica Aresch by response surface methodology, J. Sci. Food Agric. 94(1) (2014) 139–145.

[24] D.P. Veetil, G. Mercier, J.F. Blais, M. Chartier, Optimization of operating parameters for the selective flotation of heavy metals from contaminated fine sediment using response surface model, Soil. Sediment. Contam. 23(2) (2014) 107–125.

[25] H.X. Cheng, K.F. Zeng, J.T. Yu, Adsorption of uranium from aqueous solution by graphene oxide nanosheets supported on sepiolite, J. Radioanal. Nucl. Chem. 298(1) (2013) 599–603.

[26] X.F. Liang, Y.M. Xu, G.H. Sun, L. Wang, Y.B. Sun, Y. Sun, X. Qin, Preparation and characterization of mercapto functionalized sepiolite and their application for sorption of lead and cadmium, Chem. Eng. J. 174(1) (2011) 436–444.

[27] Y. Bai, Y.C. Shi, D.L. Wetzel, Fourier transform infrared (FT-IR) microspectroscopic census of single starch granules for octenyl succinate ester modification, J. Agric. Food Chem. 57(14) (2009) 6443–6448.

[28] W.T. Tsai, C.W. Lai, K.J. Hsien, Adsorption kinetics of herbicide paraquat from aqueous solution onto activated bleaching earth, Chemosphere 55(6) (2004) 829–837.

[29] D. Singh, Effect of different factors on the adsorption of phosphamidon on two different types of Indian soil, Adsorpt. Sci. Technol. 16(8) (1998) 583–594.

[30] E. Demirbas, M.Z. Nas, Batch kinetic and equilibrium studies of adsorption of Reactive Blue 21 by fly ash and sepiolite, Desalination 243(1–3) (2009) 8–21.

[31] S. Karcher, A. Kornmüller, M. Jekel, Anion exchange resins for removal of reactive dyes from textile wastewaters, Water Res. 36(19) (2002) 4717–4724.

[32] A. Bousher, X.D. Shen, R.G.J. Edyvean, Removal of coloured organic matter by adsorption onto low-cost waste materials, Water Res. 31(8) (1997) 2084–2092.

[33] M.S. Chiou, H.Y. Li, Equilibrium and kinetic modeling of adsorption of reactive dye on cross-linked chitosan beads, J. Hazard. Mater. 93(2) (2002) 233–248.

[34] G. Han, Y. Huang, G. Li, Y. Zhang, T. Jiang, Detailed adsorption studies of active humic acid fraction of a new binder on iron ore particles, Miner. Process Extr. M. Rev. 35(1) (2014) 1–14.

[35] F. Amin, H.N. Bhatti, S. Rehman, Optimization of growth parameters for lipase production by Ganoderma lucidum using response surface methodology, Afr. J. Biotechnol. 10(28) (2011) 5514–5523.