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Microwave-assisted regeneration of spent activated carbon from paracetamol wastewater plant using response surface methodology

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ABSTRACT

The spent activated carbon used in the petrochemical plant was regenerated by microwave heating. The response surface methodology is utilized to optimize the process parameters. The results revealed that the optimum parameters were: regeneration temperature of 600°C, regeneration time of 10 min and microwave power of 500 W, with the methylene blue number and yield being 202.5 mg/g and 42.22%, respectively. The regeneration activated carbon under optimal parameters was characterized by nitrogen adsorption at 77 K, scanning electron microscope (SEM) and Fourier transform infrared spectroscopy (FTIR). The Brunauer–Emmett–Teller surface area, total pore volume and average pore diameter are estimated to be 987.5 m^2/g, 1.11 ml/g, and 4.49 nm, respectively. The effect of regeneration cycles on methylene blue number of activated carbon were studied, indicating that the activated carbon still had a certain of adsorption capacity. The activated carbon was characterized by FTIR and SEM. The results of SEM indicate that impurities which covered the surface of activated carbon are cleaned. It can be seen from FTIR analysis that functional groups of activated carbon are changed through microwave heating.

Keywords: Activated carbon; Microwave heating; Response surface; Methylene blue number

1. Introduction

Activated carbon is a most important material with abundantly developed pores and huge specific surface area. Moreover, it is widely used in water treatment [1,2], organic contaminants adsorbing [3] and catalytic supports [4]. Activated carbon is available in many forms including powders, granular, carbon fibers, and molecular sieves [5]. With the continuous development of industrial technology, activated carbon has a wide application prospect in some areas such as double electrochemical capacitors material [6–8], storage of gas fuel, and so on [9–11].

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Paracetamol (N-acetyl-p-aminophenol or acetaminophen) which is a pharmaceutical analgesic and antipyretic agent is widely used in the modern society [12–17]. The main reaction in the paracetamol production process is shown in Fig. 1. Activated carbon is used as adsorbent in paracetamol wastewater treatment, and adsorbs impurities and removes color from wastewater. The impurities are N-(2-hydroxyphenyl) acetamide, N-(4-hydroxyphenyl)-propanamide N-(3-chl -oro-4-hydroxyphenyl)acetamide, N-phenylacetamide, and so on. In the production process, powdered activated carbon was thrown into reactor from top to bottom, reacted with waste water sufficiently and filtered from the wastewater [18,19].

The regeneration technique of activated carbon is widespread in the world. It is applied to deal with spent activated carbon because it causes a large number of resources wastes, expendable item and great cost. In recent years, a wide variety of regeneration techniques in the literature treating spent activated carbon have been reported including electrochemical process [20,21], thermal treatment [22,23] chemical regeneration [24], and microwave regeneration [25–27].

Microwave belongs to the portion of the electromagnetic spectrum with wavelengths from 1 mm to 1 m and corresponding frequencies are in the range of 300 MHz–300 GHz [28]. In the recent years, microwave heating is one of the most effective methods and widely used in regeneration process of activated carbon [29]. The conventional heating method has some disadvantages such as longer heating time and higher energy loss and so on. However, microwave heating can solve these problems [30]. Microwave heating is being increasingly utilized for variety of applications, as heating is uniform, where the absorbed microwave readily transforms into heat inside the particles by dipole rotation and ionic conduction. The materials receive energy through dipole rotations and ionic conduction in microwave heating [31]. Activated carbon has an excellent microwave absorbing properties, which can obviously reduce reaction time and save energy [32].

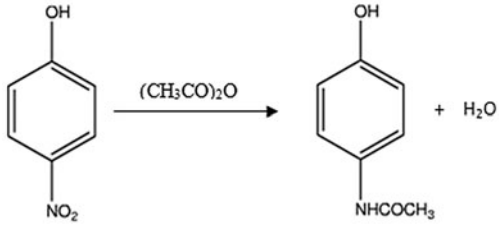

Fig. 1. The main reaction in the paracetamol production process.

Response surface methodology is an optimization technique based on multivariate statistics which includes experiment design, statistical model, and process optimization [33–36]. In the present study, spent activated carbon has been regenerated in order to improve adsorption capacity of activated carbon and realize recycling via microwave heating. The experiment is designed by Design Expert 7.1.5. Regeneration temperature, regeneration time and microwave power are three main parameters and corresponding methylene blue number and yield of activated carbon are investigated in this experiment.

2. Materials and methods

2.1. Regeneration of spent activated carbon

The specific property of original powdered activated carbon is: methylene blue number of 225 mg/g, iodine number of 405 mg/g, and surface area being of 1,139.8 m^2/g, respectively. Spent activated carbon is received from a paracetamol factory in Liaoning province of China which has 45 mg/g of methylene blue number, 103 mg/g of iodine number and 252.6 m^2/g of surface area. The proximate analysis of spent activated carbon is presented as follow: moisture 32.71%, volatile16.29, fixed carbon 50.06% and bash 0.94%.

Fig. 2 shows a self-made microwave furnace which is employed in the regeneration experiments. It consists of a lot of systems such as microwave power control system (with the output power of 0–1.5 KW and the microwave frequency of 2.45 GHz) and the temperature control system the temperature control system (temperature controlled by the input microwave power during the activation process, measured by the thermoelement type of nichrome-nickel silicon armor, placed such that it touched raw, with a measurement precision of ±5℃). The regeneration process utilizing no protecting gas can be carried out by

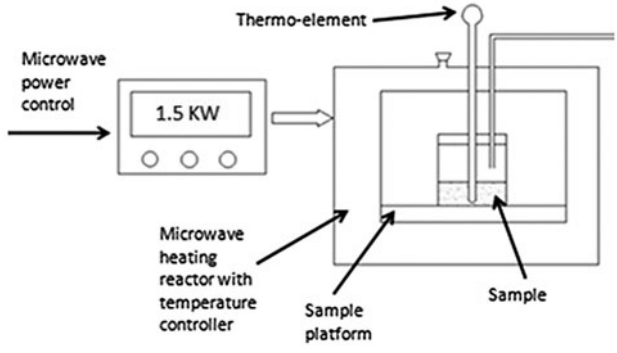

Fig. 2. Schematic of microwave heating equipment.

regeneration time (5–15 min), regeneration temperature (500–700℃) and microwave power (300–500 W). The ranges of experimental conditions are determined by preliminary single parameter experiments. The size of microwave furnace is 40 cm × 30 cm × 24 cm and the microwave is generated by continuous wave magnetron which converts alternating current power to microwave power. The microwave power converts to thermal energy because of the dielectric loss of internal materials in microwave heating. The material was placed into corundum crucible, and the height and width of the material in the corundum crucible is about 3 cm and 3.5 cm (about 20 g each time), respectively, then putted into microwave furnace. When the sample was cooled to room temperature, and got the sample from the microwave furnace followed by put into electric dry oven at the temperature of 105℃ for 3 h, which were stored for further characterization such as methylene blue number, yield, and so on.

2.2. Properties measurement techniques and characterization of activated carbon

The yield of activated carbon is based on the following equation: $W_A/W_B \times 100\%$, where W_A is the weight of regenerated activated carbon, W_B is the weight of the spent activated carbon. The standard testing methods of PR China (GB/T12496.10–1999) [37] is used for testing the methylene blue adsorption number of activated carbon after heated in the microwave. Iodine number is tested for the activated carbon according to the National Standard Testing Methods of PR China (GB/T12496.8–1999) [37]. The nitrogen adsorption of activated carbon is carried out at 77 K using an automatic adsorption apparatus (Autosorb-1-C, Quantachrome). The Brunauer–Emmett–Teller (BET) surface area of the sample is calculated by the BET equation. The total pore volume is estimated to be the equivalent liquid volume of the adsorbate (N_2) when the pressure is 0.99. The pore volume distribution of activated carbon is evaluated by the non-localization density functional theory (NLDFT). The microstructure is analyzed by the scanning electron microscope (SEM, Philips XL30ESEM-TMP). The Fourier transform infrared spectroscopy (FTIR) is applied to identify the chemical function groups present in the activated carbon. FTIR spectra are operating in the range of 4,000–400 cm^{-1} using AVATAR 360 (Thermo Nicolet , AVATAR, FTIR, 360).

3. Results and discussion

3.1. Response analysis and verification of the regression model

The central composite design (CCD) which is the standard of response surface method is selected for the optimization of parameters. The total number of the experiment is 20 including 8 factorial points, 6 axial points, and 6 replicates at the central points. The experiment conditions and values are listed in Table 1. The dependant variables selected are regeneration temperature (X_1), regeneration time (X_2), and microwave power (X_3). Microwave power is applied to control the heating rate of spent activated carbon. The upper and lower limits of the process variables are shown in Table 1, which is decided based on the literature pertaining to regeneration of activated carbon.

What's more, the results of methylene blue number (Y_1) and yield of activated carbon (Y_2) has been shown in Table 2. A total of 20 sets of experiments are necessary for a 2^3 full factorial design, which consists of 8 factor points, 6 pivot points, and 6 center points.

The final empirical models in terms of faded parameters for methylene blue number (Y_1) and yield of activated carbon (Y_2) are shown in Eqs. (1) and (2):

$$Y_1 = 197.30 + 6.62\,X_1 + 12.68\,X_2 - 8.88\,X_1^2 - 15.50\,X_2^2 - 7.55\,X_3^2 \quad (1)$$

$$Y_2 = 42.02 - 6.58\,X_1 - 4.46\,X_2 - 1.95\,X_3 - 1.56\,X_1^2 + 1.57\,X_2^2 \quad (2)$$

The qualities of the model developed are always evaluated using the correlation coefficient value (R^2) which is 0.9552 for Eq. (1) and 0.9471 for Eq. (2), respectively. The results of the analysis of variance (ANOVA) have been carried out in Tables 3 and 4 to prove the validity of the model. Table 3 is ANOVA for response surface quadratic model on methylene blue adsorption number. The F-value is 23.71 and Prob. > F is less than 0.0001, which proves that the model is significant. The model parameters X_1, X_2, and the interaction terms (X_1^2, X_2^2, X_3^2) have been investigated to be significant according to the low "p" value (≤0.05), based on the ANOVA. Table 4 is ANOVA for response surface quadratic model on yield of activated carbon. The F-value is 19.98 and Prob. > F is less than 0.0001 which suggests that the model is significant.

Table 1
Independent variables and their levels used for central composite rotatable design

Factors	Code	Levels				
		−1.682	−1	0	1	1.682
Regeneration temperature (℃)	X_1	431.82	500	600	700	768.18
Regeneration time (min)	X_2	1.59	5	10	15	18.41
Microwave power (W)	X_3	163.64	300	500	700	831.36

Table 2
Experimental design matrix and results

Run	Regeneration temperature X_1 (℃)	Regeneration time X_2 (min)	Microwave power X_3 (W)	Methylene blue number Y_1 (mg/g)	Yield Y_2 (%)
1	500.00	5.00	300.00	142.5	52.11
2	700.00	5.00	300.00	165.0	40.15
3	500.00	15.00	300.00	172.5	48.29
4	700.00	15.00	300.00	187.5	34.24
5	500.00	5.00	700.00	150.0	50.13
6	700.00	5.00	700.00	165.0	38.74
7	500.00	15.00	700.00	180.0	46.25
8	700.00	15.00	700.00	180.0	28.55
9	431.82	10.00	500.00	157.5	48.55
10	768.18	10.00	500.00	180.0	27.91
11	600.00	1.59	500.00	127.5	58.11
12	600.00	18.41	500.00	172.5	36.06
13	600.00	10.00	163.64	165.0	49.25
14	600.00	10.00	836.36	180.0	40.03
15	600.00	10.00	500.00	202.5	42.22
16	600.00	10.00	500.00	195.0	41.91
17	600.00	10.00	500.00	187.5	41.83
18	600.00	10.00	500.00	202.5	42.01
19	600.00	10.00	500.00	195.0	41.79
20	600.00	10.00	500.00	202.5	42.13

Table 3
ANOVA for response surface quadratic model on methylene blue adsorption number

Source	Sum of squares	Degree of freedom	Mean square	*F*-value	Prob. > *F*
Model	7,672.89	9	852.54	23.71	<0.0001
X_1	597.60	1	597.60	16.62	0.0022
X_2	2,196.08	1	2,196.08	61.07	<0.0001
X_3	78.43	1	78.43	2.18	0.1705
X_1X_2	63.28	1	63.28	1.76	0.2142
X_1X_3	63.28	1	63.28	1.76	0.2142
X_2X_3	7.03	1	7.03	0.20	0.6678
X_1^2	1,135.20	1	1,135.20	31.57	0.0002
X_2^2	3,464.31	1	3,464.31	96.33	<0.0001
X_3^2	821.38	1	821.38	22.84	0.0007

Table 4
ANOVA for response surface quadratic model on yield of activated carbon

Source	Sum of squares	Degree of freedom	Mean square	*F*-value	Prob. > *F*
Model	1,012.10	9	112.46	19.88	<0.0001
X_1	590.64	1	280.20	104.93	<0.0001
X_2	271.42	1	67.09	47.97	<0.0001
X_3	51.91	1	8.41	9.18	0.0127
X_1X_2	8.82	1	0.92	1.56	0.2403
X_1X_3	1.19	1	0.63	0.21	0.6569
X_2X_3	2.35	1	0.001	0.42	0.5334
X_1^2	34.93	1	14.12	6.17	0.0323
X_2^2	35.70	1	4.08	6.31	0.0308
X_3^2	7.25	1	4.62	1.28	0.2840

The model parameters X_1, X_2, X_3 and the interaction terms (X_1^2, X_2^2, X_3^2) are significant, based on low "*p*" value (≤0.05).

3.2. Methylene blue number

Fig. 3 shows the effect of regeneration temperature and regeneration time on the methylene blue number. As can be seen from Fig. 3, methylene blue number can reach more than 200 mg/g when regeneration temperature and regeneration time are 600℃ and 10 min, respectively. The increase in regenerating temperature as well as the regeneration time contributes to the reduction in the residual organic content of the sample due to promotion of evaporation rate at low temperatures while through thermal cracking at high temperatures, both contributing to the opening of the pores. Similar observations have been reported by Duan et al. [38] about the relationship of the regeneration temperature and regeneration time on activated carbon regeneration.

Fig. 4 shows the three-dimensional response surfaces of the combined effect of microwave power and regeneration temperature on the methylene blue number at a regeneration time of 10 min. The trend of Fig. 4 is similar to Fig. 3. It can be seen that the methylene blue number of regenerated carbon increases with increase in regeneration temperature and microwave power.

Fig. 5 shows the comparison of predicted vs. the experimental data for methylene blue number of activated carbon. The response data of experiment data is

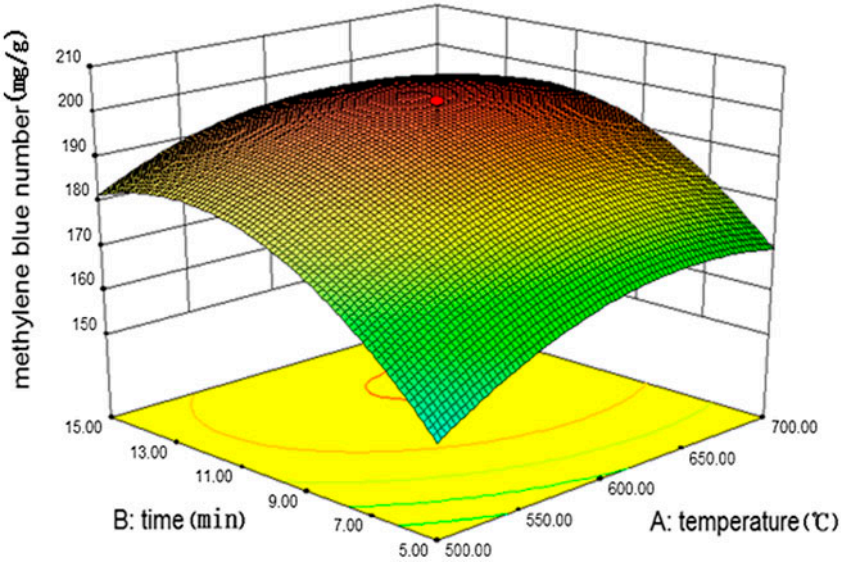

Fig. 3. Three-dimensional response surface plot of methylene blue number: effect of regeneration temperature and regeneration time on the methylene blue number (microwave power: 500 W).

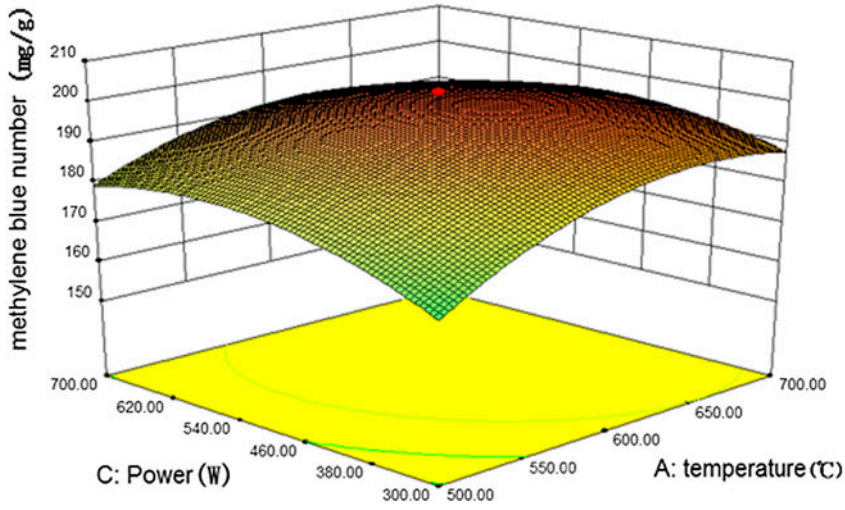

Fig. 4. Three-dimensional response surface plot of methylene blue number: effect of regeneration temperature and microwave power on the methylene blue number (regeneration time: 10 min).

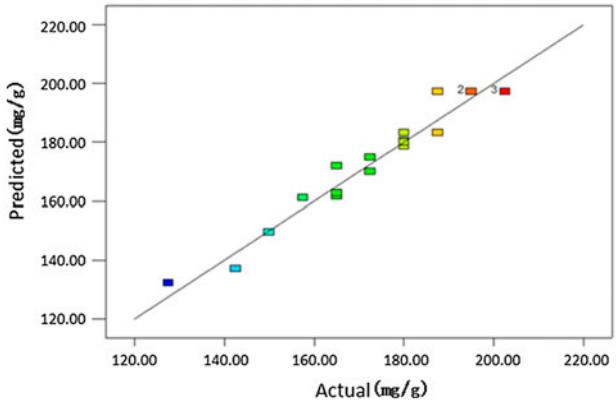

Fig. 5. Predicted vs. experimental adsorption of methylene blue number.

measured in a particular run while the predicted values are used the model Eq. (1) to evaluate. Hence, it can be concluded that the two-factor interaction model is suitable to describe the correlation between experimental parameters and methylene blue adsorption capacities. As can be observed in the Fig. 5, the experimental data are evenly distributed on the both sides of the model prediction, which indicates that the suitability of the model is developed in capturing the correlation between the process and response variables.

3.3. Activated carbon yield

Three-dimensional response surfaces plot of yield with respect to the regeneration temperature and regeneration time is shown in Fig. 6, while Fig. 7 shows the effect of regeneration temperature and microwave power on the yield of activated carbon. Similar conclusions have been reported by Duan et al. [39] in literature pertaining to regeneration activated carbon. It can be found from Figs. 6 and 7 that the yield of activated carbon decreases with the increase in regeneration temperature, regeneration time, and microwave power. Among the three variables, the influence of regeneration temperature is observed to be more significant than any other factors. Because the experiment doesn't have any other protecting gas, and the part of activated carbon surface has been burned. The yield of activated carbon is not high and it is average around 40% decreases with increasing of three parameter values.

Fig. 8 is the yield of activated carbon between predicted number and the experimental number. According to Fig. 8, it can be seen that experiment value is evenly distributed on both sides of the predict line basically, which indicate that the selected model can reflects independent and dependent variables in the microwave regeneration.

3.4. Verification of optimization results

According to the prediction of response values and parameters by Design Expert 7.1.5, the results between predicted and experimental in the optimal conditions are shown in Table 5. The predicted methylene blue number is 2.64% different from experimental, and yield is 0.48% different from experimental. It can be found that the response surface model is effective from these values. By testing, the iodine number of the regenerated activated carbon is 326 mg/g.

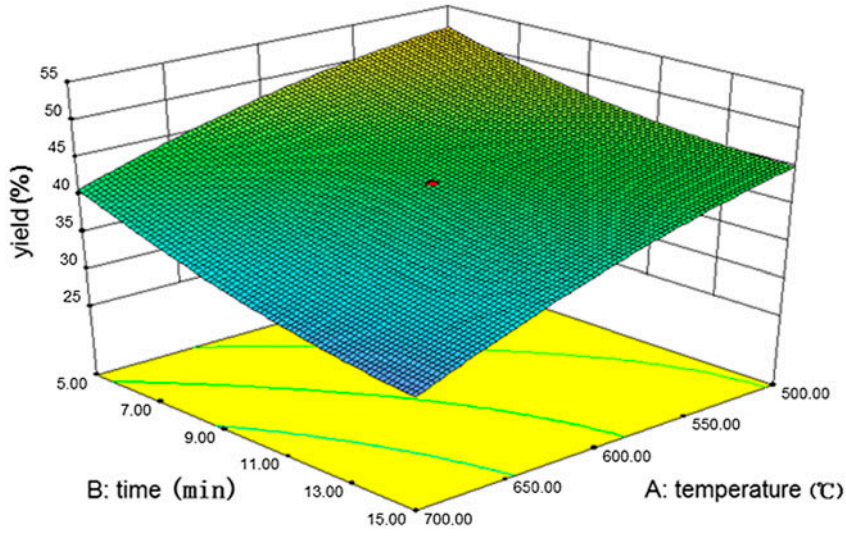

Fig. 6. Three-dimensional response surface plot of activated carbon yield: effect of regeneration temperature and regeneration time on yield (microwave power: 500 W).

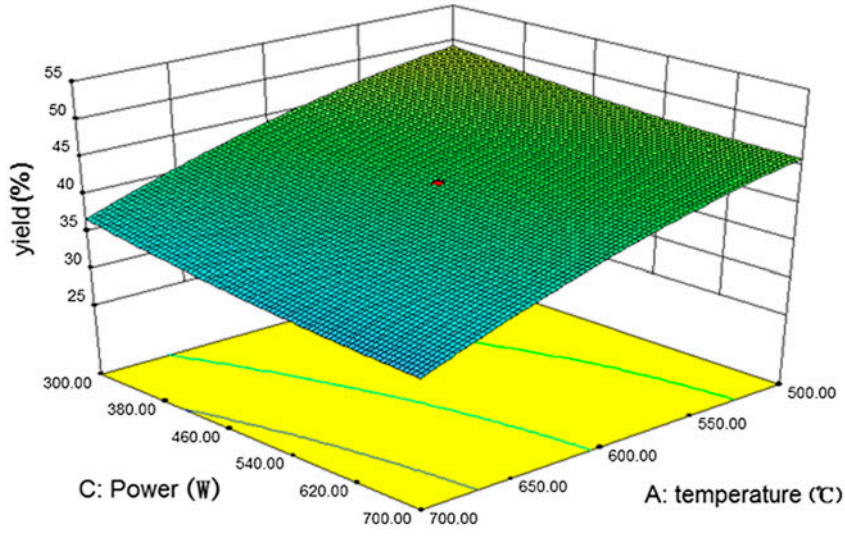

Fig. 7. Three-dimensional response surface plot of activated carbon yield: effect of regeneration temperature and microwave power on yield (regeneration time: 10 min).

3.5. Pore structure and surface area analysis

The nitrogen adsorption isotherms of spent and regenerated activated carbon which have been estimated under optimal conditions using the Autosorb instrument at 77 K are shown in the Fig. 9. Nitrogen adsorption isotherm is measured over a relative pressure (P/P_0) range from approximately 10^{-7}–1. The BET surface area is calculated from the isotherms using the BET equation (Gregg and Sing, 1982). It can be found from Fig. 9 that the isotherms of spent and regenerated activated carbon pertain to intermediate type II [40] based on IUPAC classification, which meaning the significant pores of regenerated activated carbon is mesopores. It can be seen that the isotherm of regenerated activated carbon is above the isotherm of spent activated carbon, which indicates that regenerated activated carbon has a relatively good adsorption properties.

Fig. 10 shows the cumulative pore volume distribution chart for spent and regenerated activated carbon.

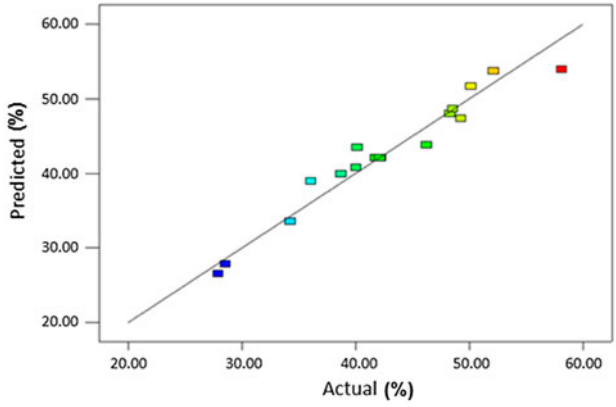

Fig. 8. Predicted vs. experimental activated carbon yield.

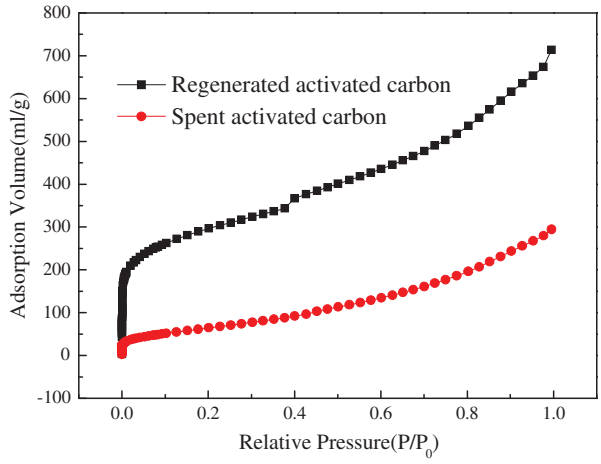

Fig. 9. Nitrogen adsorption isotherm of the spent and regenerated activated carbon.

Fig. 10 also suggests that the amount pores of regenerated activated carbon are in the mesoporous range.

Moreover, the details of pore structure of spent activated carbon and regenerated activated carbon are shown in Table 6. As can be seen from Table 6, the pore volume, average pore diameter, and BET surface area of regenerated activated carbon has a large increased, which means that the microwave activating process has an excellent effect on the regeneration of spent activated carbon. Compared with the spent activated carbon, the regenerated activated carbon shows a remarkable increase in the adsorption volume, with corresponding BET surface area of 987.5 m^2/g.

The regeneration activated carbon treated the iodine solution as the target adsorbate, and did adsorption–desorption experiments under the optimal experimental conditions. Fig. 11 shows the effect regeneration cycle on methylene blue number of regeneration activated carbon. We can find that the methylene blue number of regeneration activated carbon decrease with the increase in regeneration cycle. It is because that the increase in regeneration cycle can lead to the collapse of the pore structure formed formally, and the accumulated a certain amount of carbide in pore will also cause of regeneration activated carbon adsorption performance degradation. The five cycles of the methylene blue number of regeneration activated carbon is 137.5 mg/g, which is only 67.9% of the first cycle methylene blue number of activated carbon.

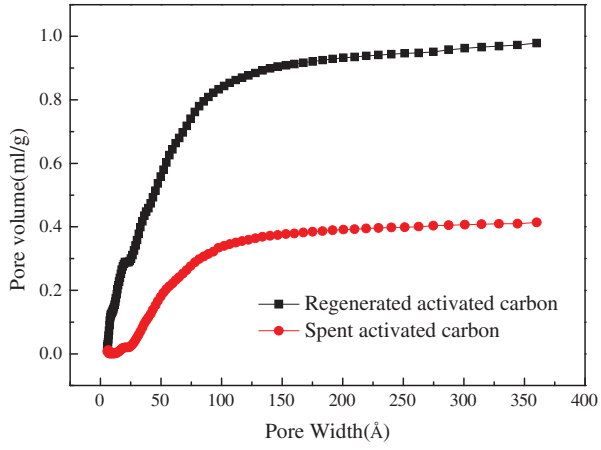

Fig. 10. Cumulative pore volume distribution chart for spent and regenerated activated carbon.

3.6. SEM analysis

The microscopic structure of the spent activated carbon and regenerated activated carbon is shown in Fig. 12. It can be found that the surface of the spent activated carbon is smooth and is covered a lot of impurities in Fig. 12(A). However, as shown in

Table 5
Validation of process optimization

Regenerated temperature (℃)	Regenerated time (min)	Microwave power (W)	Methylene blue number (mg/g)		Yield (%)	
			Predicted	Experimental	Predicted	Experimental
600	10	500	197.3	202.5	42.02	42.22

Table 6
Details of pore structure of spent activated carbon and regenerated activated carbon

Subject	Spent activated carbon	Regenerated activated carbon
BET Surface area (m^2/g)	252.6	987.5
Total pore volume (cm^3/g)	0.46	1.11
Average pore diameter (nm)	7.24	4.49
Micropore volume (%)	4.76	29.50
Mesopore volume (%)	95.24	70.50

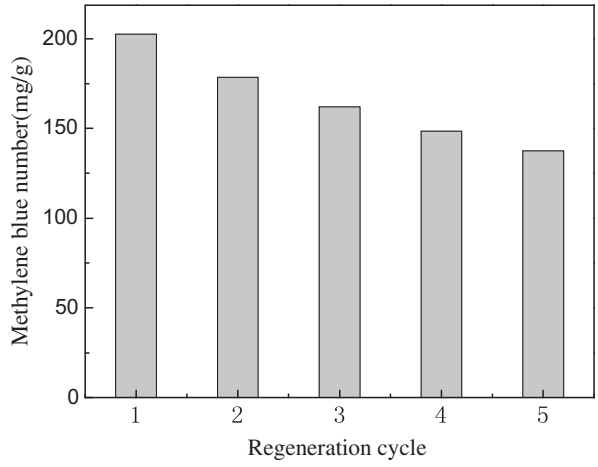

Fig. 11. The effect regeneration cycle on methylene blue number of regeneration activated carbon.

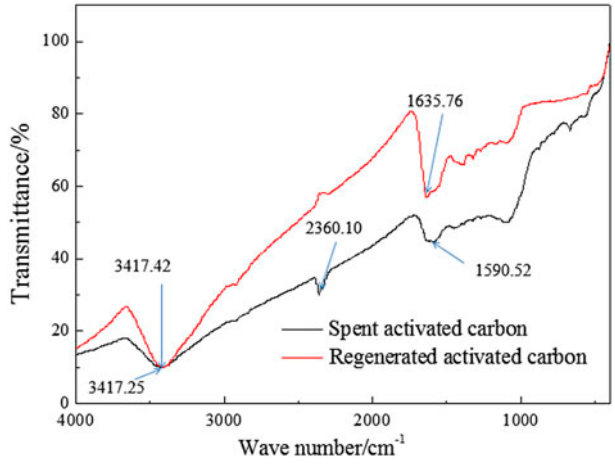

Fig. 13. FT-IR spectra of the spent and regenerated activated carbon.

Fig. 12(B), the impurities on the surface of the activated carbon are almost removed. At the same time, the surface of regenerated carbon becomes more and more drape and forms lots of new pores.

3.7. FTIR analysis

Fig. 13 shows the results of FTIR characterization. It can be found from the Fig. 13 that the overall shapes of the spectra between the spent activated

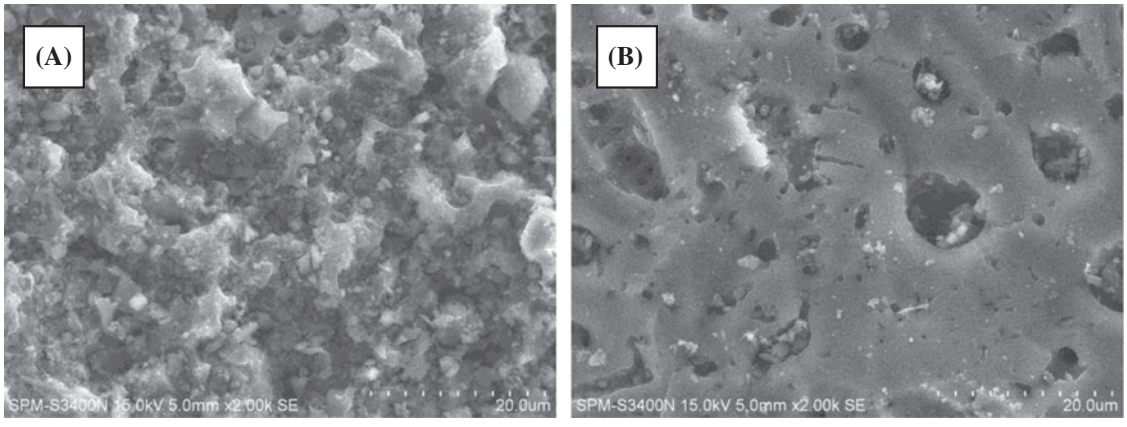

Fig. 12. SEM images of the spent (A) and regenerated (B) activated carbon.

carbon and regenerated carbon have a little different. The spectrum of regeneration activated carbon has two peaks at 3,417.42 and 1,635.76 cm^{-1} and the spectrum of spent activated carbon has three peaks at 3,417.25, 2,360.10, and 1,590.52 cm^{-1}. As can be seen from Fig. 13, the broad peak at 3,417.42 cm^{-1} is assigned to O–H stretching vibration, the band at 1,635.76 and 1,590.52 cm^{-1} indicate that there may be exist C=O functional group. There is a little different between FTIR results of spent activated carbon and regenerated activated carbon. The adsorption peak of spent activated carbon in 2,360.10 cm^{-1} can be assigned to C≡C antisymmetric stretching [41].

4. Conclusion

Spent activated carbon which is used in paracetamol adsorption has been regenerated via microwave heating. The effects of three vital process parameters, including regenerated temperature, regenerated time, and microwave power on the methylene blue number and yield are investigated systematically. The experiments are designed by Design Expert 7.1.5 in the range of 500–700˚C, 5–15 min, 300–700 W, respectively. As a result, the methylene blue number and yield are 202.5 mg/g and 42.22%, respectively, under optimal conditions such as regeneration time of 10 min, regeneration temperature of 600˚C and microwave power of 500 W. The regenerated activated carbon under optimal conditions is characterized by nitrogen adsorption at 77 K, SEM, and FTIR. The BET surface area, total pore volume, and average pore diameter that are at the optimized process conditions are estimated to be 987.5 m^2/g, 1.11 ml/g, and 4.49 nm, respectively. The methylene blue number of regeneration activated carbon is only 137.5 mg/g after five cycles and still have a certain of adsorption capacity. The results of SEM indicate that impurities covered on the surface of activated carbon are basically cleaned. It can be observed from FTIR analysis that functional groups of regeneration activated carbon are changed through microwave heating.

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