IOP Conference Series: Materials Science and Engineering

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To cite this article: Dongji Liu *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **490** 022040

IOP Conf. Series: Materials Science and Engineering **490** (2019) 022040 doi:10.1088/1757-899X/490/2/022040

Preparation of Adsorbent from Fly Ash for Methylene Blue Wastewater Treatment

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Abstract: Fly ash contains SiO_2 and Al_2O_3, which account for more than 60% of the total. This is similar to the composition of zeolite which is frequently be used for adsorbent. Therefore, fly ash is used to prepare adsorbent for wastewater treatment by microwave-assisted hydrothermal method. The optimal reaction conditions were determined: alkali-ash ratio of 15:1, reaction temperature of 100℃ and reaction time of 0.5h. The removal rate of methylene blue is up to 97.6%. The adsorbent was characterized by SEM and FT-IR. Large number of silicon (aluminum) oxygen tetrahedrons and the porous lattice structure contribute high adsorption performance of the adsorbent.

1. Introduction

Fly ash is a kind of solid waste produced from coal burning in thermal power plants. The common uses of fly ash include preparation of cementing material, bricks and aerated block [1, 2]. However, high value use of fly ash has always been the target. It is a reasonable approach to prepare zeolite or adsorbent from fly ash which can be used for the treatment waste water.

At present, the methods for synthesizing zeolite mainly include hydrothermal synthesis method, alkali melting method and salt heating method, etc. Wang zhimei et al. [3] summarized various synthesis methods. Hydrothermal synthesis and alkali melting method are the two methods most widely used by scholars at present. Peng Shaohao [4] et al. found that the adsorbent synthesized by alkali melting-hydrothermal method has good selenium removal effect, and the maximum adsorption is up to 5.36mg/g. Wang guanghui [5] et al. synthesized NaA zeolite by alkali melting-hydrothermal method and the removal for Methylene blue can reach more than 99%. Microwave heating is the body heating caused by the loss of meson in the electromagnetic field of materials, which is different from common heating methods [6]. Miki Inada [7] et al. synthesized NaP1 zeolite by microwave-assisted hydrothermal method. The CEC value reached 200meq/100g. However, the process of synthesizing zeolite from fly ash is complicated and the requirements for synthesis conditions are relatively strict.

In this paper, fly ash was used to prepare adsorbent with the method of microwave-assisted hydrothermal for methylene blue treatment. The effect of some factors as alkali-ash ratio, reaction temperature and reaction time are investigated to obtain the optimal conditions.

2. Experimental

2.1. Materials
Fly ash is provided by Xibaipo power plant and it's chemical composition is shown in the Table 1. Sodium hydroxide, hydrochloric acid, and methylene blue were bought from market.

Table 1. Chemical composition of fly ash (%)

Material	CaO	MgO	SiO_2	Fe_2O_3	Al_2O_3	SO_3	Loss
Fly ash	4.662%	0.831%	49.432%	5.400%	36.506%	0.735%	6.280%

2.2. Pretreatment of fly ash
Firstly, put fly ash into a muffle furnace and calcined at 500℃ for 1h to remove impurities such as C and S. Then put the calcined fly ash into a three-necked flask. Add an appropriate amount of hydrochloric acid solution (the dosage of hydrochloric acid should immerse the fly ash completely) [8], heating and stirring at 100℃ for 1h.

2.3. Synthesis of adsorbent
A certain amount of pretreated fly ash sample is put into flask, adding NaOH solution (2mol/L) and aging for 1h. Then the mixture is transferred to a microwave reactor for constant temperature reaction, then filter.

The pH of filtrate is adjusted to about 6 with the HCI (2mol/L), transferred it to microwave reactor, heating and crystallizing. The reaction conditions should be controlled at 80℃ for 1.5h; then filering and drying to obtain the adsorbent.

2.4. Measurement of removal rate of methylene blue
The methylene blue standard solution is prepared according to GB/T12496.10-1999. The standard curve was measured at 664nm with ultraviolet spectrophotometer. The equation is as follows.

$$A=0.2386C_e \tag{1}$$

Where A is absorbance, C is concentration of methylene blue.

The adsorbent was mixed with 15mg/L methylene blue at a ratio of 500:1. Oscillating for 20min. Measureing the absorbance and calculating the removal rate according to the following formula.

$$\eta=(C_0-C_e)/C_0 \tag{2}$$

Where η is methylene blue removal rate, C_0 is initial concentration of methylene blue.

3. Results and discussion
The effects of alkali-ash ratio (Ratio of the volume of Sodium hydroxide solution to mass of fly ash), reaction temperature and reaction time on the adsorption capacity of the prepared adsorbent were investigated. The removal rate of methylene blue wastewater was used as target value.

3.1. Alkali-ash ratio
The experiments were performed with the alkali-ash ratio of 5, 7, 10, 15, 20. The experimental results are shown in Figure 1.

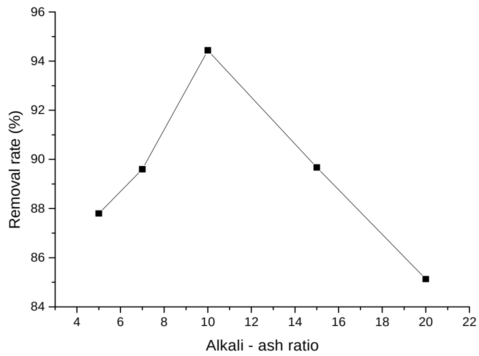

Figure 1. Effect of alkali-ash ratio on methylene blue removal rate

As can be seen from Figure 1, the maximum removal rate of methylene blue is 94.44% when the alkali-ash ratio is 10. With the increase further in alkali-ash ratio, the removal rate decreases. The proper range of alkali-ash ratio falls within 7~10.

3.2. Reaction temperature

The alkali-ash ratio was set to 10:1. Under this condition, the removal effect of methylene blue by the adsorbent was investigated when the reaction temperature was at 70℃, 80℃, 90℃, 100℃ and 105℃. The results are shown in Figure 2.

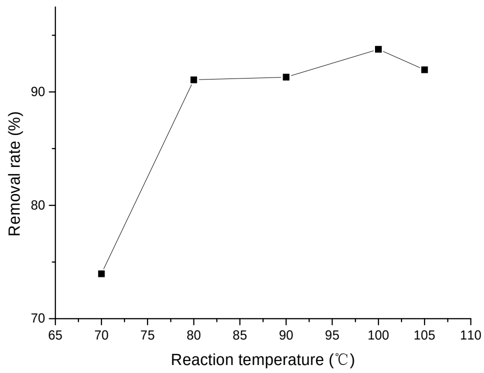

Figure 2. Effect of reaction temperature on methylene blue removal rate

It can be seen that the removal rate of methylene blue increases with the increase in reaction temperature. When the temperature increases from 70℃ to 80℃, the removal rate of methylene blue increased rapidly from 73.96% to 91.07% and then the curve tends to smooth.

3.3. Reaction time

The alkali-ash ratio and reaction temperature were set at 10:1 and 80℃ respectively. Under these conditions, the removal effect of methylene blue was investigated with the reaction time. The results are shown in Figure 3.

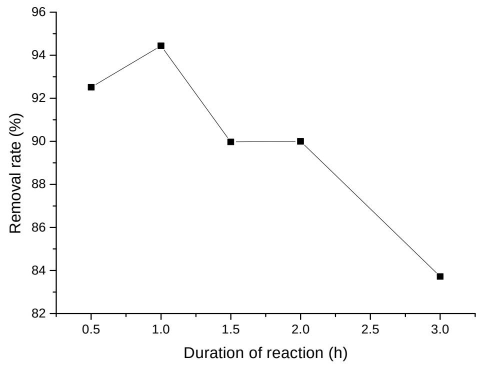

Figure 3. Effect of reaction time on methylene blue removal rate

It can be seen that the removal rate of methylene blue increases first and then decreases with the increase of temperature. When the reaction time was 1h, the removal rate reached the maximum. At this time, the removal rate was 94.44%.

3.4. Orthogonal test
Three factors with three levels were selected for orthogonal experiment design. See Table 2. The results are shown in Table 3.

Table 2. Orthogonal matrix factors and horizontal table

A Alkali-ash ratio	B Reaction temperature(℃)	C Reaction time(h)
7:1	80	0.5
10:1	90	1
15:1	100	1.5

It can be seen from the Table 3 that the change of alkali-ash ratio has the most significant effect on the adsorption of methylene, followed by the reaction temperature and the reaction time. The optimal factor/level combination is A3B3C1.

It has been verified that the removal rate of methylene blue under the combination of factors/levels of A3B3C1 can reach 97.6%.

Table 3. Response of orthogonal experiment

Experiment No.	Alkali-ash ratio	Reaction temperature	Reaction time	Blank	Experimental results
1	1	1	1	1	91.42
2	1	2	2	2	90
3	1	3	3	3	89.69
4	2	1	2	3	91.79
5	2	2	3	1	92.54
6	2	3	1	2	93.77
7	3	1	3	2	94.33
8	3	2	1	3	93.38
9	3	3	2	1	96.53
K1	90.370	92.513	92.857	93.297	
K2	92.370	91.973	92.773	92.700	
K3	94.747	93.330	92.187	91.620	
S	4.377	1.357	0.670	1.877	

3.5. Micro-structure of the absorbent
The microstructure was investigated by using SEM. As can be clearly seen from Figure 4, the surface morphology of the adsorbent is loose and porous, and the surface pores are highly developed, which is one important cause of strong adsorption capability for methylene blue.

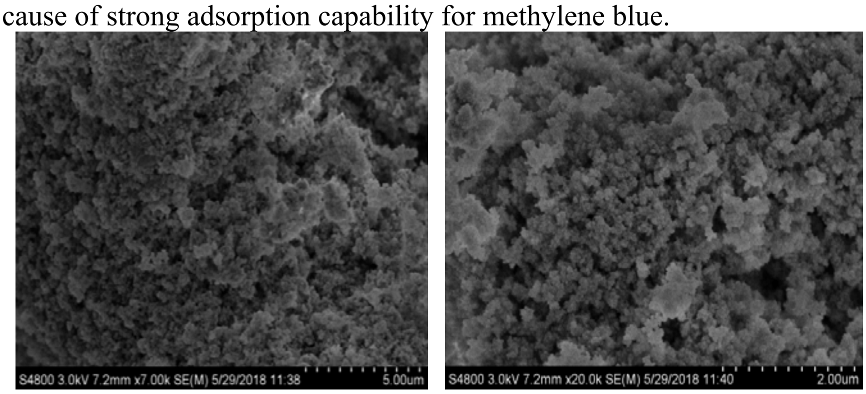

Figure 4. SEM photograph of the adsorbent

FT-IR was used for characterization of the absorbent. In Figure 5, the absorption peak at 3420.77cm^{-1} is the -OH stretching vibration peak of water. It is the skeleton vibration peak of silicon (aluminum) oxygen tetrahedron (SiO_4, AlO_4) at 1623.16cm^{-1}. The asymmetric stretching vibration peak of silicon (aluminum) oxygen tetrahedron is located 1094.82cm^{-1} and 799cm^{-1} is the bending vibration peak of silicon (aluminum) oxygen tetrahedron. Large number of silicon (aluminum) oxygen tetrahedrons and the lattice structure contribute high adsorption performance of the adsorbent [9].

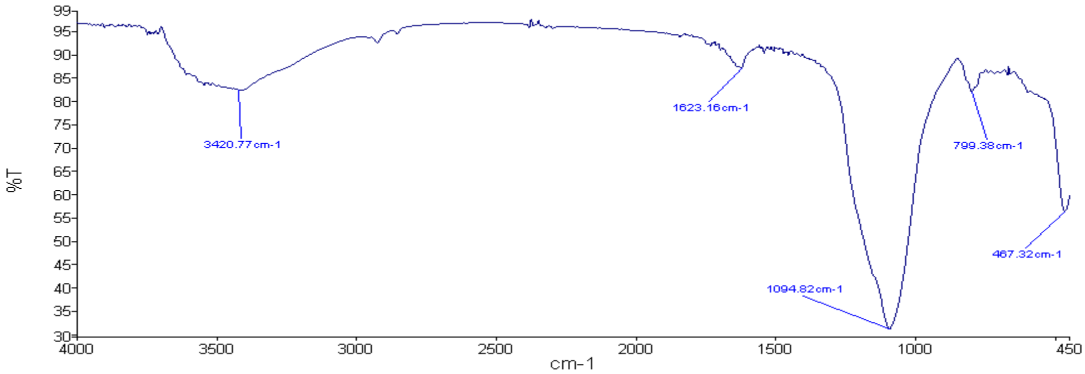

Figure 5. FT-IR spectrum of the adsorbent

4. Conclusion

The preparation of adsorbent from fly ash and removal effect of methylene blue from methylene blue wastewater were investigated through single factor experiments which including alkali-ash ratio, reaction temperature, reaction time. Then orthogonal experiment was used to obtain the optimal synthesis conditions: alkali-ash ratio of 15:1, reaction temperature 100℃, reaction time 0.5h. The removal rate of methylene blue is up to 97.6%.

The microstructure and characterization of the absorbent were studied by using SEM and FT-IR respectively. Large number of silicon (aluminum) oxygen tetrahedrons and highly developed porous and lattice structure contribute high adsorption performance of the adsorbent prepared.

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