Removal of Methylene Blue in Aqueous Solution by Microwave and Fly Ash Combined Process

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Keywords: Fly ash, Microwave, Methylene blue, Removal

Abstract. The combined process of microwave and fly ash is used to remove methylene blue pollutant from aqueous solution in this study. The microwave radiation time, fly ash dosage and pH value on removal process are varied separately to study the effect of these experimental factors on removal performance. The experimental results show that microwave and fly ash have obvious synergistic effect to remove the pollutant from solution. The combined process of microwave and fly ash shows its good removal efficiency for methylene blue from aqueous solution. Radiation time, fly ash dosage and pH value have significant influence on removal rate of methylene blue. With the increase of radiation time, fly ash dosage and pH value, the removal rate of methylene blue increases. Compared with fly ash alone process, the combined process of microwave and fly ash is more effective to treat pollutant.

Introduction

Fly ash (FA) discharged from coal-fired power plants has received more and more concern and attention. With the increasing demand of electrical energy, the yield of fly ash will become larger. If without effective reuse or disposition, fly ash may bring pollution and harm to its surrounding environment. Hence, there are more and more researches focusing on using fly ash to minimize the negative effect of fly ash on environment. Yao et al. mentioned that about 20% of the fly ash generated is being used in concrete production; other uses include road base construction, soil amendment, zeolite synthesis, and fillers in polymers [1]. Wee reviewed the availability and the potential of fly ash using in the carbon dioxide capture and storage (CCS) technology [2]. Fly ash is also used in water treatment field. Chen and Wang [3-6] proved the effectiveness of the combined technology of O_3 and fly ash to degrade herbicides pollutant from aqueous solution.

As a new and developing technique, microwave radiation process (MW) shows its wide applications in household, medical, industrial fields, etc. Because microwave has its special property in rapid and selective heating, microwave is also effectively used in the field of environmental technology and chemical technology including pyrolysis, phase separation and extraction processes, soil remediation, remediation of hazardous and radioactive wastes, coal desulphurization, sewage sludge treatment, chemical catalysis and organic/inorganic syntheses [7]. Remya and Lin pointed that microwave is also a useful technique for water and wastewater treatment [7]. Microwave alone, or coupled with others oxidants or other oxidation processes were used to treat ammonia nitrogen, chlorophenol derivative, phenol, pentachlorophenol, malachite green, dimethoate, and other organic pollutants in water or in wastewater. The combined processes include MW-UV (ultraviolet light), MW-H_2O_2, MW-UV-H_2O_2, MW-UV-TiO_2, MW-Fenton, MW-AC (activated carbon), MW-Persulfate ($S_2O_8^{2-}$), etc. These combined processes are helpful to widen microwave's application scope and to enhance its treatment efficiency for water and wastewater treatment. When it comes to the linking of fly ash and microwave, microwave is generally used to modify fly ash or to converse it to zeolite [8]. However, the combined removal process of microwave and fly ash directly using in water treatment is rarely reported. Thus, this paper focuses on the removal dyes pollutant from aqueous solution using combined process of MW-FA. Methylene blue (MB), a widely used cationic dyes, is selected as the model dyes pollutant. During the producing process and operating process, methylene blue can be discharged into the water environment or soil environment. Due to its high emission, high chromaticity and complex structure, wastewater of methylene blue is difficult to be biodegraded and maybe remain in the environment for relatively long periods of time. It is reported that methylene blue can cause increased pulse rate, vomiting, shock, heinz body formation, cyanosis, jaundice, quadriplegia, and tissue necrosis in humans [9]. Hence, the treatment of methylene blue pollutant has its practical significance and investigative value. The removal performance and influence factors of methylene blue from aqueous solution are discussed in this study using combined process of microwave and fly ash.

Experimental Part

The model pollutant used in laboratory was methylene blue (AR, purchased from Shanghai Maikun Chemical Co. Ltd.). The reaction solutions were prepared with deionized water. pH value of solution was adjusted using sulfuric acid solution or sodium hydroxide solution. Solution pH was measured using a pH meter (PHS-3C, Shanghai Precision and Scientific Instrument Corporation). The fly ash used in this study was taken from a power plant of Hebei province. A certain amount of fly ash was added into the reaction solution and mixed well. Samples were withdrawn from the reactor and filtered through 0.22 μm filter membrane, and then analyzed using spectrophotometer (722N, Shanghai INESA Analytical Instrument Co. Ltd) at 664 nm. The microwave oven (NN-GF599M) was purchased from Panasonic Corporation. The bottle of reaction solution was placed in a water bath to keep constant temperature 20°C before experiment. If no particular mention, the experimental conditions were bellows: the dosage of fly ash was 0.5 g fly ash added into 50 mL reaction solution; the pH value of solution was initial pH value without any pH adjustor; the power of microwave was middle fire; the initial concentration of methylene blue was 20 $mg \cdot L^{-1}$; the reaction time was 4 min.

The removal rate was calculated using the proportion of the removed amount in initial amount of methylene blue. And the removed amount is obtained from the initial concentration minus the concentration after treatment.

In order to simplify the expression later, microwave, fly ash and methylene blue are abbreviated to MW, FA and MB separately.

Result and Discussion

Promotion of MW on FA Performance. The removal rate of methylene blue using FA alone and combined process of MW-FA were experimented and analyzed to show the promotion of microwave on fly ash (Fig. 1).

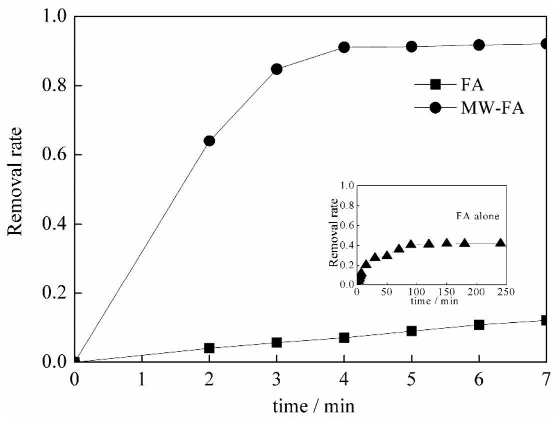

Fig. 1 Promotion effect of microwave on fly ash to remove methylene blue

From Fig. 1, it can be seen that the removal of methylene blue by fly ash is enhanced greatly by microwave. At the reaction time 7 min, the removal rate of methylene blue by fly ash alone is only 12.09%, which is small and shows that the adsorption of fly ash doesn't work significantly during the reaction period (in 7 min). The subfigure of Fig. 1 shows the adsorption performance of fly ash alone in enough long period. The relative stable value appeared after 100 min indicates that the maximum removal rate of fly ash is around 40%, which shows the maximum of adsorption capacity of fly ash is a relatively small value. In MW-FA combine process, the removal rate of methylene blue reaches above

90% at 7 min. Hence, the promoting effect of microwave on fly ash is obvious. Remya and Lin reviewed that the reason of higher degradation efficiency of MW-absorbing materials like activated carbon is the hot-spot formation [7]. The promoting effect of MW on FA is also explained using this point. The temperature of reaction solution with fly ash was observed to be higher than that without fly ash at same microwave power and same radiation time. It is proved that the heating performance of microwave is better and faster than that of other heating methods. The chemical compositions of fly ash such as Fe_2O_3, MgO, TiO_2, K_2O, CaO, Na_2O, Al_2O_3 and other trace metal compositions may act as catalytic effect [10]. Fly ash can play its role as direct absorbent of methylene blue or absorbent of microwave and then to remove methylene blue. And the addition of fly ash changes the pH value of reaction solution, which can influence the dissociation extent of initial organic. The assistant of microwave energy leads to the polarization of molecules. These different roles can be integrated into the overall improvement of microwave on fly ash. Fig. 1 also shows that the radiation time of microwave has a great influence on the removal efficiency of methylene blue. In MW-FA experimental system, with the increase of radiation time of microwave, the removal rate of MB goes up firstly in 4 min, and then becomes relatively stable.

Effect of fly ash dosage. The removal rate of methylene blue was determined in FA alone system and MW-FA system with different dosages of fly ash (Fig. 2).

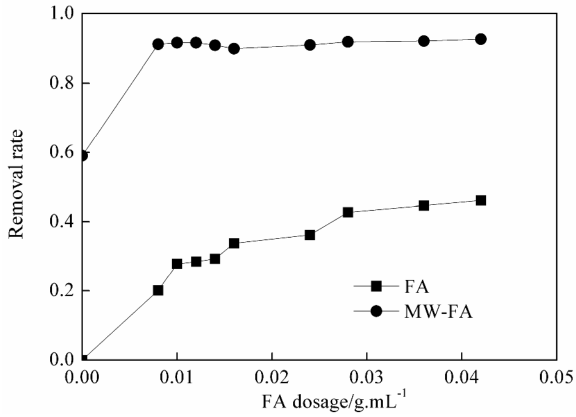

Fig. 2 Removal rate of MB at different FA dosages

From the figure, it can be seen that the dosage of fly ash has positive effect on MB removal. In MW-FA system, with the increase of FA dosage, the removal rate (4 min) of methylene blue is enhanced sharply, and then basically maintains at around 90%. In FA alone system, the removal rate (1 h) of methylene blue increases gradually with increasing of FA dosage. Fly ash provides active surface to absorb methylene blue directly or absorb the energy from microwave and therefore to promote reaction of methylene blue. From the subfigure in Fig. 1, the maximum capacity of fly ash absorption is relatively low. Apart from the adsorption ability of fly ash, the hot-spot is another contribution. With the increase of fly ash, hot-spot formation is enhanced in the microwave field. The MW-FA system is more effective and stable than FA alone system.

Effect of pH value. During chemical and physical treatment of wastewater, pH value is a non-ignorable factor in most cases. Fig. 3 shows the removal rate of methylene blue at different pH values of reaction solution.

From Fig. 3, it is proved that pH value of reaction solution has strong influence on removal of methylene blue. In the pH range from 2.5 to 9.5, with the increase of pH value, the removal rate increases slowly from 22.39% to 39.33%. In the pH range from 9.5 to 12.5, the removal rate is rising rapidly from 39.33% at pH=9.5 to 80.98% at pH=12.5, and then reaches above 90%. The removal rate of pH value 12.5 climbs to 96.70%. Hence, the elevating of pH value helps to remove methlyene blue from aqueous solution. The change of pH value can affect the surface charge of fly ash and methylene blue, and also affect the degree of ionization and speciation of methylene blue during the process, therefore change the adsorption behavior and capacity. Hence, the impact of pH value is positive.

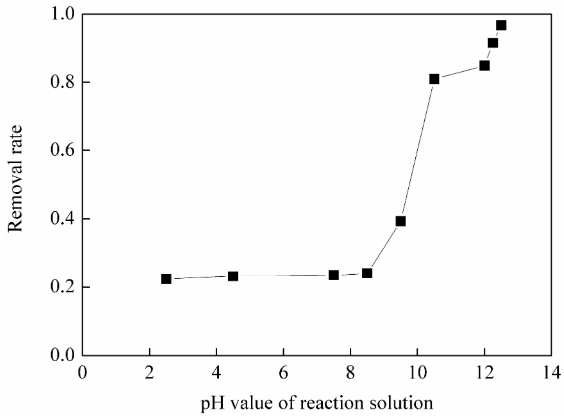

Fig. 3 Removal rate of MB at different pH values

Conclusions

In order to expand the application range of microwave and fly ash, the removal of methylene blue using FA alone system and MW-FA combined system was examined. The removal of methylene blue by fly ash is enhanced greatly by microwave, which shows the improvement effect of fly ash and microwave. The combined process of fly ash and microwave has good removal efficiency in a short treatment time. The removal efficiency of methylene blue increases with the increase of microwave radiation time, fly ash dosage and pH value of solution. These experimental factors have strong positive effect on treatment of methylene blue.

Acknowledgements

This work was financially supported by the Fundamental Research Funds for the Central Universities (No. 2014MS123).

References

[1] Z. T. Yao, X. S. Ji, P. K. Sarker, J. H. Tang, L. Q. Ge, M. S. Xia, Y. Q. Xi: Earth-Science Reviews. Vol. 141(2015), p. 105-121.

[2] J. H. Wee: Applied Energy. Vol. 106(2013), p. 143-151.

[3] L. Chen: Advances in Engineering Research. Vol. 84(2016), p. 624-627.

[4] L. Chen: Advances in Engineering Research. Vol. 84(2016), p. 628-631.

[5] L. Chen, Y.Y. Wang: Degradation of Dicamba in Aqueous Solution Using Ozonation Assisted by Fly Ash. ICEEMA 2015, 2015, 6.

[6] L. Chen: Influence Factor of Dicamba Degradation Using O_3-FA Process in Aqueous Solution. ICEEMA 2015, 2015, 6.

[7] N. Remya, J.-G. Lin: Chemical Engineering Journal. Vol. 166(2011), p. 797-813.

[8] S.S. Bukhari, J. Behin, H. Kazemian, S. Rohani: Fuel. Vol. 140(2015), p. 250-266.

[9] S. Banerjee, G. C. Sharma, M.C. Chattopadhyaya, Y. C. Sharma: Journal of Environmental Chemical Engineering. Vol. 2(2014), p. 1870-1880.

[10] Y.-L. Song, J.-T. Li: Ultrasonics Sonochemistry. Vol. 16(2009): 440-444.