The study of a new type of micro-electrolysis technology on nitrobenzene wastewater treatment

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Keywords: Fly ash; Iron-carbon micro-electrolysis; Nitrobenzene wastewater; Single-factor experiment; Orthogonal experimental.

Abstract. In this work, a new type of micro-electrolysis technology using combined materials containing fly ash, iron scraps and rare earth Gd on the treatment of toxic and harmful nitrobenzene wastewater was presented. Fly ash was applied instead of activated carbon in the combined materials and single-factor experiment was conducted to determine and optimize the relevant influence factors on the removal rate of nitrobenzene. Both the main and minor factors and the optimum levels influencing the nitrobenzene removing property were determined via the orthogonal experimental design. The results showed that the rank of each factor for removing ratio of nitrobenzene from better to less was respectively: pH, rare earth Gd dose, mass ratio of fly ash and iron and reaction time. The optimal combined conditions were 2, 12mL, 2:1and2.0 h respectively. The experiment data of the removal rate of nitrobenzene was 83.3%. In addition, the removal efficiency was improved by 13.6% with the addition of rare earth Gd.

Introduction

Nitrobenzene wastewater has the characteristics of high toxicity, high oxygen demand and difficult biodegradability, etc. Nitrobenzene is now recognized as one of the high toxic and carcinogenic substances in the world. It is also one kind of harmful wastewater needed to be focused on in China [1,2].

Iron-carbon micro-electrolysis technology is one of the methods to treat organic wastewater [3~5], which is based on electrochemical principle of metal corrosion and dissolution. Nowadays, the researches on micro-electrolysis are inclined to the consideration of iron carbon micro-electrolysis. However, few researches are focused on the problem of replacement of filler, especially on replacing original filler with solid waste. The rare earth has the special 4f electronic structure, unusually rich electronic energy levels and very lively chemical properties. Studies show that the unique catalytic effect and strong adorability of rare earth metals have been confirmed in the field of water treatment [2,6]. This paper proposes a new kind of micro-electrolysis method to treat the toxic and harmful nitrobenzene wastewater by a micro-electrolysis method, which replaces active carbon with coal ash, use waste iron scraps and a certain amount of catalytic rare earth Gd. This paper provides a new way for the recycling of solid waste coal ash and seek a new treatment method for nitrobenzene wastewater which containing toxic and harmful organic pollutants to realize treating wastes with wastes and reducing the emission of pollutants.

Experimental

Waste iron scraps after wash and soak iron scraps of particle size 0.5 ~3mm with washing powder and dilute sulfuric acid, rinse with distilled water, and set aside. Coal ash (Tangshan Datang International DouHe Power Plant): firstly, wash with water and dry it, then, immerse 48 h by analog nitrobenzene solution to make it saturated. The nitrobenzene solution with a concentration of 100mg/l was used as the analog nitrobenzene wastewater.

The treated waste iron scraps and coal ash were mixed in a certain proportion in the 250ml beaker, and then a certain amount of analog nitrobenzene wastewater was added. The mixture was stirred in

the speed 150 r/min. The main influence factors on the treatment effect of analog nitrobenzene wastewater was investigated, including ratio of coal ash/iron scraps, pH, reaction time, and the dosage of rare earth Gd.

Nitrobenzene remove rate is calculated using the formula following:

$$\eta = \text{ $(C_0$-$Ct) /$C_0\times100\%}$$
 where, η —the nitrobenzene removal rate of wastewater in the reaction C_0 —the nitrobenzene concentration of initial wastewater C_t —the nitrobenzene concentration of treated wastewater

Results and Discussion

Single-factor experiment

The influence of the mass ratio of coal ash/iron scraps on nitrobenzene removal rate. 100ml nitrobenzene wastewater with concentration of 100mg/L and 1g iron scraps were mixed. The mass ratio of coal ash/iron scraps were changed from 1:1 to 3.5:1 in order to investigate the influence of the mass ratio of coal ash/iron scraps on nitrobenzene removal rate. The experimental results were shown in Figure 1. With the increase of the mass ratio of coal ash/iron scraps, the nitrobenzene removal rate was on a downward trend. When the mass ratio of coal ash/iron scraps was 1:1~1.5:1, the nitrobenzene removal rate was larger, up to 65.2~63.1%.

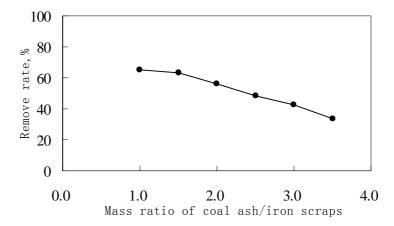


Fig.1 The influence of the mass ratio of coal ash/iron scraps on nitrobenzene removal rate

The influence of pH on nitrobenzene removal rate. The mass ratio of coal ash/iron scraps was1:1. To investigate the influence of pH on nitrobenzene removal rate. The pH values were adjusted to 2, 3, 4, 5 and 6, respectively. The experimental results were shown in Figure 2.

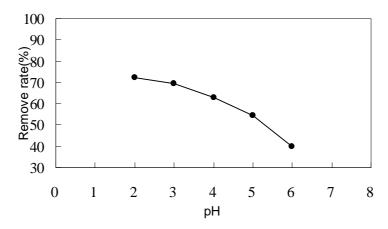


Fig.2 The influence of pH on nitrobenzene removal rate

The lower pH was conducive to the removal of aniline nitrobenzene. The removal rate of nitrobenzene was higher when pH was 2~ 3, up to about 70%. After that, with the increase of pH, the removal rate of nitrobenzene decreased rapidly. As can be seen, the pH was determined as 2~ 3.

The influence of reaction time on nitrobenzene removal rate. The effects of reaction time on nitrobenzene removal rate experimental results were shown in Figure 3. We can see that with the extension of reaction time, the nitrobenzene removal rate was continuously increasing. When the reaction time was 2.0h, the nitrobenzene removal rate reached 70.5%, and the increasing trend became gradually stable after 2.0h. So the reaction time was determined as 2.0h.

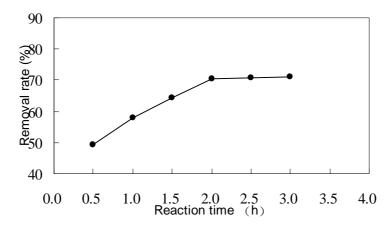


Fig. 3 The influence of reaction time on nitrobenzene removal rate.

The influence of rear earth Gd dosage on nitrobenzene removal rate. The concentration of rare earth Gd is 6.16g/L. The rear earth Gd dosage were changed from 0ml to 15ml in order to investigate the influence of rear earth Gd dosage on nitrobenzene removal rate. The experimental results were shown in Figure 4. The nitrobenzene removal rate increased with the increase of rare earth Gd dosage. The removal rate of nitrobenzene reached 82.7% when the dosage of rare earth Gd was 9ml. The removal rate of nitrobenzene was about 13.6% higher than that of without rare earth Gd. So the dosage of rare earth Gd was determined as 9 ml.

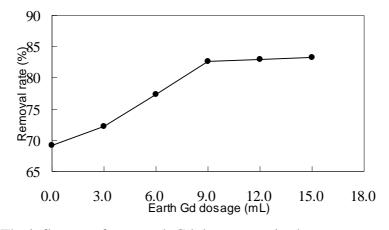


Fig. 4 The influence of rear earth Gd dosage on nitrobenzene removal rate

The orthogonal experiment

According to the single factor experiment results, the main factors and the level range of each factor in orthogonal experiments of nitrobenzene wastewater was confirmed, which treated by micro-electrolysis method that replace active carbon with coal ash. These factors are expressed by A, B, C and D, respectively. The main influence factors include pH, reaction time, mass ratio of coal

ash/iron scraps and the dosage of rare earth Gd. Choose three levels of each factor. Design the Orthogonal table as $L_9(3^4)$. As shown in Table 1 and the results were shown in Table 2.

Table 1 Orthogonal table

Factors levels	A (pH)	B (Reaction time/h)	C (Mass ratio of coal ash/iron scraps)	D (Dosage of rare earth Gd/ml)
1	2.0	1	1:1	6
2	3. 0	1.5	1.5:1	9
3	4. 0	2	2: 1	12

Table 2 Orthogonal experiment results table

NO. Factors	A (pH)	B(Reaction time/h)	C (Mass ratio of coal ash/iron scraps)	D (Dosage of rare earth Gd/ml)	Removal rate of nitrobenzene /%
1	2.0	1	1:1	6	76. 5
2	2.0	1.5	1.5:1	9	80. 7
3	2.0	2	2: 1	12	83. 3
4	3. 0	1	1.5:1	12	75. 4
5	3. 0	1.5	2: 1	6	67. 2
6	3. 0	2	1:1	9	71. 7
7	4.0	1	2: 1	9	65. 4
8	4.0	1.5	1:1	12	64. 8
9	4.0	2	1.5:1	6	63. 9
K1	240. 5	217. 3	213. 0	207. 6	
K2	214.3	212. 7	220. 0	217.8	
K3	194. 1	218. 9	215. 9	223. 5	
R	46. 4	6. 2	7. 0	15. 9	

The experimental results showed that: the order of influence factors of nitrobenzene wastewater treatment by micro-electrolysis was pH > dosage of rare earth Gd > mass ratio of coal ash/iron scraps > reaction time. The optimal combination was $A_1B_3C_3D_3$. Namely, pH2, the dosage of rare earth Gd solution 12ml, the mass ratio of coal ash/iron scraps 2:1, reaction time 2.0h. Under the optimal experimental conditions, the removal rate of nitrobenzene was83. 3%.

Conclusions

In this work, the micro-electrolysis method could replace active carbon with coal ash and use waste iron scraps and catalytic rare earth Gd to treat the toxic and harmful nitrobenzene wastewater. The experimental results showed that the order of influence factors of nitrobenzene wastewater treatment by micro-electrolysis was pH > dosage of rare earth Gd > reaction time > mass ratio of coal ash/iron scraps. The optimal combination was $A_1B_3C_3D_3$. Namely, pH2, the dosage of rare earth Gd solution 12ml, the mass ratio of coal ash/iron scraps 2:1, reaction time 2.0h. The rare earth Gd on nitrobenzene wastewater treatment had a better catalytic effect, which was an important factor affecting the removal rate of nitrobenzene. The nitrobenzene removal rate increased about 13.6%.

Acknowledgements

This work was supported by the construction of Key Laboratory fund project of North China Institute of Science and Technology (Basic scientific research business of the Central University, Grant No. 3142014017)

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