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Analysis of Magnetic Flocculation Process Conditions for Domestic Sewage Treatment by Response Surface Method

Jiawei Hu[1,a], Jun Li[2,a,*], Changwen Wang[a], Yu Chen[a], Yan Li[a], Guoyang Liu[a]
[a]Key Laboratory of Beijing for Water Quality Science and Water Environment Recovery Engineering,
Beijing University of Technology, Beijing 100124, China

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Abstract. Response surface methodology was applied to analyze the magnetic flocculation process conditions for domestic sewage treatment. Box-Behnken central composition experiments design was applied. The dosage of poly aluminum chloride (PAC), polyacrylamide (PAM), magnetic particle (MP) was chosen as causal factors. The effects of these factors on soluble orthophosphate (SOP), chemical oxygen demand (COD) removal efficiency were analyzed by using RSM. The pollutant removals in domestic sewage were developed to describe by regression models. The results show that the optimum conditions were PAC 309mgL^{-1}, PAM 1.74mgL^{-1}, MP 187mgL^{-1}. Based on these conditions, the SOP and COD removal efficiency were 96.15% and 45.38% respectively. It confirms that statistic models were reliably, the magnetic flocculation operating conditions for contaminant removals in domestic sewage were appropriated.

1. Introduction

The inorganic flocculants (such as aluminums salt, ferric salt) and organic polymer solvents used as electrolyte for the flocculation process. Flocculation technology is considering as the most extensive in water treatment at present. It significantly decreased the load of further water treatment process and the cost of whole flow about water processing [1-3]. Many valuable achievements by developing new type flocculants [4,5], or researching on the combination of flocculation process and other technologies [6,7] through the domestic and international experts.

Through magnetic technology binding water treatment process can enhance and improve removing ability and efficiency of the system [8,9]. There are two methods about the magnetic technology applied to flocculation process: improved flocculating conditions after inoculated magnetic seed [10-12], or adding magnetic combined with flocculants to strengthen the flocculation efficiency [13]. Flocculating constituent rapid separation is beneficial to water quality security guarantee.

Response surface methodology (RSM) usually used for bioengineering, chemical engineering and pharmaceutical engineering to prescription screening and technology optimization [14-16]. Using multiple regression equation to fit causal factors and response values, estimation of regression equation coefficient, solving the extreme value or target value to attain the expected process or parameter optimization purpose. It is more scientific than orthogonal test. The author used RSM to optimize the magnetic flocculation process parameters for domestic sewage treatment in experimental study. The feasibility of experimental design method for flocculation process was

1,a Jiawei Hu : hujiaweiyly@hotmail.com
2,a, Jun Li : jglijun@bjut.edu.cn
* Corresponding author. Tel.: +86-10-67391726

studied. The experimental research can provide technology exploration and investigative basis for further domestic sewage disposal.

2. Experimental

2.1 Materials

The water samples was fed from Beijing University of Technology domestic sewage, the soluble orthophosphate (SOP) and chemical oxygen demand (COD) as the main characterization parameters of water quality (Table 1). Analytically pure poly aluminum chloride (PAC) (the content of $Al_2O_3 \geq 28\%$) and polyacrylamide (PAM) as the flocculants and coagulant-aid were made into certain concentration for standard solutions to reserve. The magnetic particle's specific surface area is $8.921 m^2/g$, content of Fe_3O_4 is 59% (highest peak of XRD atlas) (Fig. 1).

Table 1. Water quality of experiment.

quality parameters	contents	Analysis methods / instruments
SOP (mgL^{-1})	6.9-7.2	molybdate method (765 UV-Spectrophotometer)
COD (mgL^{-1})	263-280	potassium dichromate method (5B-3F COD Instrument)

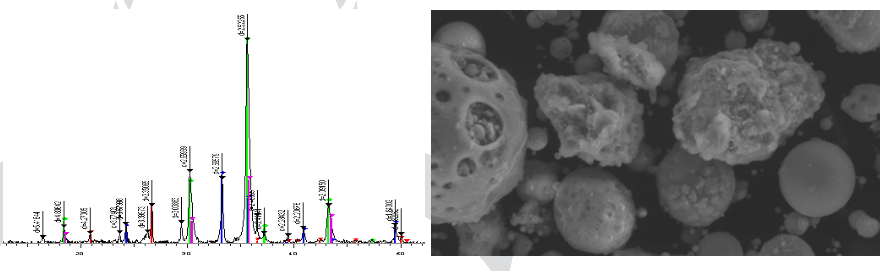


Fig. 1. XRD atlas and SEM photo of magnetic particle

2.2 Jar test

Thorough mixing of 1000mL sample and proper MP in the coagulation beaker. Setting the agitator (ZR4-6 coagulation experimental agitator) program: mixed section 288rpm, flocculation section 69rpm (the program was determined by multiple tests), auto-adding reagents. Starting up the agitator, auto-adding PAC in mixed section and PAM in flocculation section, settlement 10min after the end of flocculation process. Extraction of samples from supernatant to test according to the standard methods which were listed in Table 1.

2.3 Experimental design

The Design Expert 7.0 software was applied to design the experimental process. The first stage of the experiment was to study the effect of reagents dosage on the COD removal efficiency; The second stage was the Box-Behnken Design which was applied to design of the experiment about the effect of the dosage of PAC (X_1), PAM (X_2), MP (X_3) on SOP and COD removal efficiency (Y_1 and Y_2) (Table 2). Quadratic regression model and imitative equation obtained by RSM. And the optimal process condition of magnetic flocculation for domestic sewage treatment obtained by variance analysis.

3. Results and discussion

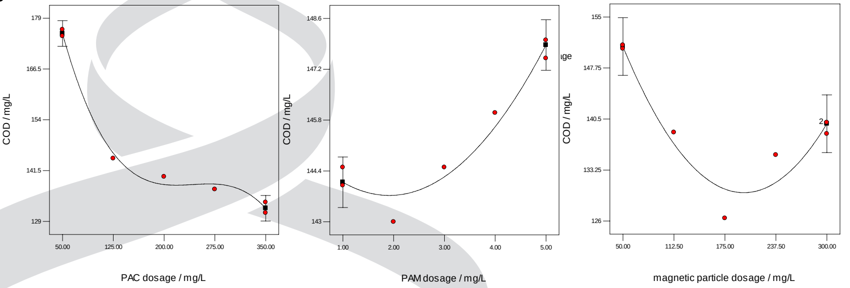
The reagents dosage will directly affect the flocculation efficiency. Comprehensive study on the effect of flocculants (PAC) on flocculation effect and coagulant aids (PAM and MP) on flocs particle. RSM as the research method to analyze the test results.

Table 2. Analytical factors and levels for RSM.

Factors	Coded symbols	Levels		
		-1	0	1
PAC dosage (mgL^{-1})	X1	50	200	350
PAM dosage (mgL^{-1})	X2	1	2.5	4
MP dosage (mgL^{-1})	X3	50	175	300

3.1 Effect of reagents dosage on the COD removal efficiency

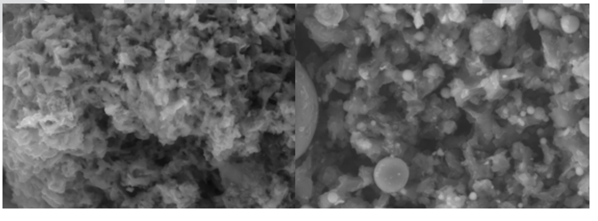
Fig. 2 (a) shows that the PAC dosage as the main factor would directly affect the flocculation efficiency. Improving the PAC dosage leads to a rapid decrease of content of residual COD. The phenomenon shows that the Al_{13} and its aggregates by PAC hydrolysis can adsorption with suspensibility COD particles which with negative electrification, then the higher charge and great molecular weight gives full play of the charge neutralization and adsorption bridging action. The suspensibility COD particles and hydroxide precipitation gel by continued PAC hydrolysis will form the flocs particle [17]. With the settling of floccus, the suspensibility COD particles will be effectively removed.



(a) (b) (c)

Fig. 2. Effect of reagents dosage on the COD removal efficiency ((a)-PAC, (b)-PAM, (c)-MP)

The coagulant aid PAM can affect the flocs size [18]. PAM will further shorten the spacing in particles by the charge neutralization action. Because the active groups with long-train structure of PAM can produce the adsorption bridging action, the flocs diameter increases gradually will be beneficial to subsidence rapidly. Fig. 2 (b) shows that the excess PAM dosage can increase residual COD. Because excess adding PAM will damage the adsorption bridging action by encapsulated the surface of suspensibility COD particles.



(a) (b)

Fig. 3. (a)-SEM photos of flocculating constituents; (b)-SEM photos of magnetic flocculating constituents.

MP which has positive charge plays strengthening flocculation effect. Adding MP can increase the solid particles quantity in the water which will be increasing the amount of flocs by multiple collisions. As the condensation core, MP adsorption and negative electrification suspensibility COD particles will form magnetic flocs by electrostatic force action [19]. Fig. 2 (c) shows that the excess MP dosage can increase the content of suspensibility COD. The structure of magnetic flocs is differ from general flocs (Fig. 3).

3.2 RSM analysis of magnetic flocculation process

BBD of 3 factors 3 levels type that is a standard plan of Design Expert software, was used in experimental design about the dosage of PAC (X_1), PAM (X_2) and MP (X_3) to the effect of SOP and COD removal efficiency (Y_1 and Y_2) (Table 3). The fitting formula (expressed by coding values) of quadratic regression model as follows:

$$Y_1=+91.17+31.97x_1-2.21x_2-2.63x_3+4.65x_1x_2+0.85x_1x_3+0.055x_2x_3-27.24x_1^2-4.18x_2^2+5.11x_3^2. \quad (1)$$

$$Y_2=+44.88+4.47x_1+0.18x_2+0.50x_3+0.88x_1x_2+0.000x_1x_3+0.25x_2x_3-2.04x_1^2-1.65x_2^2+1.08x_3^2. \quad (2)$$

There is no significant difference between experimental data of Table 3 and predictive values of statistical model. It shows that the test data reliable. Through the variance analysis, PAC dosage (X_1) is the significant ($p<0.05$). The PAC as the main factor would directly affect the flocculation efficiency. The data of regression analysis (Table 4) shows that, model fitting coefficient R^2 of fitting formula (1) is 95.24% and modified fitting coefficient R^2(adj) is 89.12%. Model (1) is high fitting degree and reliability. There is similar conclusion about model (2). Fig. 4 is the response surface graphs.

Table 3. Box-Behnken design and experimental data

Run	X_1 (mgL^{-1})		X_2 (mgL^{-1})		X_3 (mgL^{-1})		Y_1 (%)		Y_2 (%)	
	code	actual	code	actual	code	actual	Exp.	Pre.	Exp.	Pre.
1	1	350	0	2.5	-1	50	99.37	99.53	48.74	47.89
2	-1	50	0	2.5	-1	50	50.42	40.55	39.09	38.94
3	-1	50	0	2.5	1	300	37.02	33.59	39.09	39.94
4	0	200	0	2.5	0	175	83.93	91.17	47.72	44.88
5	-1	50	-1	1	0	175	30.21	34.64	38.08	37.41
6	1	350	0	2.5	1	300	89.37	99.23	48.74	47.89
7	-1	50	1	4	0	175	12.07	20.93	36.05	36.01
8	0	200	0	2.5	0	175	98.58	91.17	45.19	44.88
9	1	350	-1	1	0	175	98.14	89.28	44.57	44.38
10	0	200	-1	1	-1	50	91.56	97.00	43.06	43.88
11	0	200	0	2.5	0	175	91.22	91.17	43.91	44.88
12	0	200	1	4	-1	50	91.47	92.47	43.55	44.88
13	0	200	0	2.5	0	175	90.76	91.17	44.05	44.88
14	0	200	0	2.5	0	175	91.35	91.17	43.55	44.88
15	0	200	-1	1	1	300	92.63	91.64	44.57	44.38
16	1	350	1	4	0	175	98.6	94.17	46.05	46.72
17	0	200	1	4	1	300	92.76	87.33	46.05	45.23

Table 4. Results of regression analysis for the Box-Behnken design.

Model term	Std. Dev	R^2	R^2(adj)
Y_1	9.13	95.24%	89.12%
Y_2	1.48	92.86%	83.67%

The response surface graphs show that, PAC dosage would directly affect the flocculation efficiency. There is obvious peak and tendency on the surface. Compared with the significant effect the settlement rate, PAM and MP would not affect the flocculation efficiency. It is showed on the surface.

3.3 Optimization of magnetic flocculation process

The target values of SOP and COD removal efficiency were 95% and 45% respectively; the optimum conditions by RSM predictive values were PAC 309mgL^{-1}, PAM 1.74mgL^{-1}, MP 187mgL^{-1}. Through three times verification test, the average SOP and COD removal efficiency are 96.15% and 45.38% respectively, relative error less than 5%. The results show that the response surface models reliable, optimization process parameters can reach the experimental target. Magnetic flocculation process can effectively remove the pollutants of the domestic sewage. There is no obvious removal effect about ammonia nitrogen under the optimum conditions.

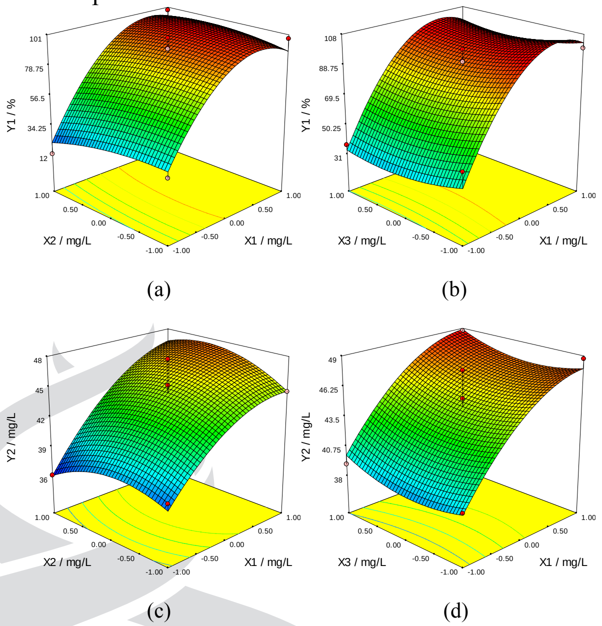


Fig. 4. Response surface showing the effects of different factors on pollutants removal efficiency

4. Conclusion

Response surface methodology was applied to analyze the magnetic flocculation process conditions for domestic sewage treatment. The experimental results show that the response surface models with high reliability, which can used in prediction and optimization about pollutants removal efficiency. The dosage of PAC is directly affecting the flocculation efficiency; MP can enhance flocculation process and improve sedimentation effect of flocs. The massive water disposal throughput has been playing an important role in emergency water treatment. The optimum conditions of magnetic flocculation process: PAC 309mgL^{-1}, PAM 1.74mgL^{-1} and MP 187mgL^{-1}; the SOP and COD removal efficiency were 96.15% and 45.38% respectively under the optimum conditions. There is no obvious removal effect about ammonia nitrogen. Therefore, Response surface methodology can reach the optimum process conditions and effectively reduce limitation of orthogonal test method. It can be provided reference for further experimental research on a pilot-scale.

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