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The study of TSS reduction in industrial wastewater by nano-coagulants

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Abstract

The main objective of this study is to investigate the effect of coagulants and nano-coagulants in different concentrations to reduce the TSS of industrial wastewater using the Taguchi statistical method. Two chemicals, iron chloride and aluminum sulfate, and two nanoparticles of iron oxide and alumina, were used as coagulants within the wastewater flocculation process at Beshel industrial complex. A L16 orthogonal array with the four factors of control at four levels each and two interactions was employed for optimization of coagulants and nano-coagulants concentration to reduce total suspended solids (TSS) in industrial wastewater. The concentration of iron chloride 1.5 g/l, concentration of iron oxide nanoparticles 1.5 g/l, concentration of aluminum sulfate 1.5 g/l and concentration of alumina nanoparticles 1.5 g/l were the optimal conditions to reduce the TSS of industrial wastewater.

Keywords: Taguchi statistical method; Industrial wastewater; TSS; Nano-coagulant

1. Introduction

Most water reuse applications prior to the last decade were producing secondary quality water for industrial or agricultural purposes [1,2,3]. Coagulation and flocculation are relatively simple and cost-effective, provided that chemicals are available and dosage is adapted to the water composition. The most common coagulants in wastewater treatment are aluminum sulphate and iron salts [4,5]. In addition, nanotechnology involves tailoring of materials at the atomic level to achieve unique properties, which can be suitably manipulated for some desired applications [6,7,8].

Malakootian et al. studied the parameters turbidity, heavy metals' concentration, color, phosphate, coagulants concentration, exposure time, TSS, pH and COD. Finally, the use of coagulation and flocculation process combined with chemical sequestration in the removal of organic and inorganic pollutants in wastewaters of automotive industry is very effective and can be used in water treatment of automotive industry [9]. In other study, the coagulation process was evaluated in treatment of municipal wastewater on the basis of organic material (COD) and suspended solids (TSS) removal efficiency. Their research evidenced once again that coagulation process can assure the limits of COD, TSS and RA for municipal wastewater treatment plants if the process is well optimized and operated [10]. In 2018, the enhanced treatment of ceramic-tile industry wastewater was investigated by modified coagulation–flocculation process using combination of poly-aluminum chloride (PAC) with anionic (A300), cationic polymer (C270) and nonionic polymers. The effects of pH, PAC coagulant dose alone and with polymers dose in various combinations was studied by jar tests. Zarei et al. resulted the coagulation–flocculation process as well as the PAC combination with anionic, cationic, and nonionic polymers can be used as an effective method for treatment of ceramic-tile wastewater. Also, the treatment cost for one cubic meter of ceramic-tile wastewater using the PAC + anionic as well as PAC + anionic and non-ionic polymers was 22.96 percent less than the PAC-alone method [11].

The objective of this work is the study of coagulants and nano-coagulants effect in different concentrations to reduce the TSS of industrial wastewater. Ferric chloride and aluminum sulphate,

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and two nanoparticles of iron oxide and alumina, were used as coagulants in the wastewater flocculation process at Beshel industrial complex.

Experimental

Materials: In this work, iron oxide nanoparticles (30-50 nm) and alumina nanoparticles (30-50 nm) were purchased from Nanotech Company (India). Iron chloride and aluminum sulfate was also purchased from Merk Company (Germany).

Design of Experiments (DOE): A L16 orthogonal array with the four factors of control at four levels each and two interactions was employed for coagulation process by adding iron oxide nanoparticles, alumina nanoparticles, iron chloride and aluminum sulfate in various concentrations. The analysis of laboratory results using Taguchi method and signal-to-noise analysis finally led to the determination of the optimal conditions in terms of the mentioned factors. An orthogonal array was designed for experiments using Qualitek-4 software considering the four variables of iron oxide nanoparticles concentration, alumina nanoparticles concentration, iron chloride concentration and aluminum sulfate concentration each at 4 different levels (Tables 1 and 2).

Table 1. Factors and their levels: the study of iron oxide nanoparticles concentration, alumina nanoparticles concentration, iron chloride concentration and aluminum sulfate concentration in coagulation process

Coagulation and flocculation process: Experiments were performed according to the orthogonal array that was designed to prepare the nanocomposite coating by considering 4 variables including iron oxide nanoparticles concentration, alumina nanoparticles concentration, iron chloride concentration and aluminum sulfate concentration at 4 different levels (Table 2). It was prepared 16 containers containing 100 ml of wastewater. During stirring, iron chloride, iron oxide nanoparticles, aluminum sulfate and alumina nanoparticles were added. The solution inside the 16 containers was first passed through filter paper separately and then the filtered solutions were used to measure the biological oxygen demand (BOD).

TSS measurement: In this work, first the weight of the filter paper was measured by digital scale. Then 50 ml of the primary wastewater sample (as control sample) and 16 samples (wastewater samples with coagulant compounds according to Table 3-4) were passed from through the filter paper. After filtration, the filter was placed in an oven at 103 °C for 1 hour. Then it was put in the desiccator for 20 minutes to remove the filter paper moisture. Finally, the filter paper was weighed again by digital scale. Total suspended solids (TSS) in mg / L were calculated as follows:

TSS= (dry weight of residue and filter paper - dry weight of filter paper alone in mg)/ volume sample in lit.

Table 1

Factors and their levels: the study of iron oxide nanoparticles concentration, alumina nanoparticles concentration, ferric chloride concentration and aluminum sulphate concentration in coagulation process

Factors	Level 1	Level 2	Level 3	Level 4
ferric chloride concentration (g/l)	0.4	0.8	1.5	3
Iron oxide nanoparticles concentration (g/l)	0.4	0.8	1.5	3
Aluminum sulphate concentration (g/l)	0.6	1	1.5	2
Alumina nanoparticles concentration (g/l)	0.6	1	1.5	2

Table 2

Levels of each factor in experiments to optimize industrial wastewater treatment process by coagulation method

Experiment number	Ferric chloride concentration	Iron oxide nanoparticles concentration	Aluminum sulphate concentration	Alumina nanoparticles concentration
1	1	1	1	1
2	1	2	2	2
3	1	3	3	3
4	1	4	4	4

5	2	1	2	3
6	2	2	1	4
7	2	3	4	1
8	2	4	3	2
9	3	1	3	4
10	3	2	4	3
11	3	3	1	2
12	3	4	2	1
13	4	1	4	2
14	4	2	3	1
15	4	3	2	4
16	4	4	1	3

Results and discussions

The total suspended solids (TSS) were measured in the wastewater from the Beshel industrial complex equal to 504 mg/l. In addition, the TSS testing was performed for 16 experimental samples in two replications and the percentage of TSS reduction was shown in Table 3.The output variable used in statistical analysis was the percentage of TSS reduction, which is a measurable physical quantity. The analysis considered in this research was signal-to-noise analysis using S/N ratio. The term "signal" represents the desirable control factors during the process and "noise" represents the undesirable effect. The S/N ratio can be calculated by the following equation [12]:

$$(S/N) = -10 \log(MSD) \qquad (1)$$

Where MSD is the mean squared deviation, which was obtained using following equation [12]:

$$MSD = \frac{\sum_{i=1}^{n}(yi)^2}{n} \qquad (2)$$

Where n is the total number of repetitions of the experiment and y_i is the result of each experiment. Note that there are three main categories of the quality characteristics, they are: 1. smaller is better, 2. nominal is better and 3. bigger is better. To obtain high percentage of TSS reduction, the 'bigger is better' quality characteristic was chosen. The average of S/N ratio for different levels of each factor to reduce the TSS including ferric chloride concentration, iron oxide nanoparticles concentration, aluminum sulphate concentration, and alumina nanoparticles concentration were presented in Figures (1) to (4).

Figure 1 showed the effect of ferric chloride concentration in flocculation process at four levels to reduce the TSS. As seen in Figure 1, the highest percentage of reduction of TSS was obtained at the level 3 from the concentration of ferric chloride (1.5 gr/l). As it is known, the S/N ratio increases with increasing the concentration of ferric chloride from 0.4 to 1.5 g/l, which indicates a further decrease in TSS at this concentration (1.5 g/l). But, the S/N ratio was 31.3 at the level of 4 which indicates a decrease in the TSS reduction. In fact, the coagulation–flocculation mechanism is proposed based on zeta potential measurement as the criteria to define the electrostatic interaction between pollutants and coagulant–flocculant agents [5].

Table 3

Levels of each factor in experiments to optimize industrial wastewater treatment process by coagulation method

Experiment number	TSS reduction%	TSS (iteration2) mg/l	TSS (iteration1) mg/l
1	37.99	312.53	312.52
2	28.54	360.2	360.1
3	46.05	271.92	271.93
4	36.43	320.36	320.37
5	37.66	314.18	314.18
6	37.84	313.29	313.28
7	41.88	292.92	292.92
8	38.57	309.6	309.61
9	42.21	291.25	291.24

10	41.43	290.16	290.16
11	41.97	292.46	292.47
12	45.06	276.90	276.89
13	29.92	353.18	353.19
14	34.94	327.88	327.89
15	39.58	304.52	304.52
16	41.63	294.18	294.17

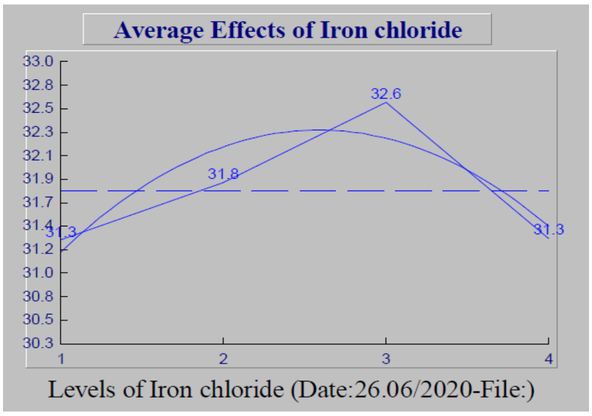

Fig. 1. Average of S/N ratio for different levels of ferric chloride concentration to reduce the TSS

As seen in Figure 2, the highest percentage of reduction of TSS was obtained at the level 3 from the concentration of iron oxide nanoparticles (1.5 gr/l). The S/N ratio for iron oxide nanoparticle concentration was 31.3 at the level 1, 31.1 at the level 2, 32.5 at the level 3, and 32.1 at the level 4.

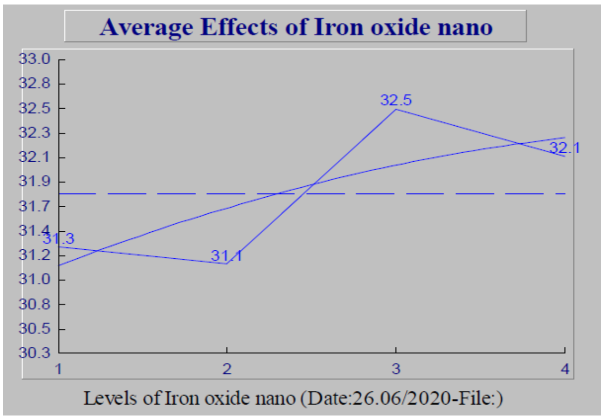

Fig. 2. Average of S/N ratio for different levels of iron oxide nanoparticles concentration to reduce the TSS

Figure 3 showed the percentage of TSS reduction in the wastewater was obtained at level 3 from the concentration of aluminum sulphate (1.5 g/l). In the flocculation process, aluminum sulphate acts similarly to ferric chloride. After adding aluminum sulphate (or ferric chloride) to wastewater, the aluminum (or ferric) reacted with the bicarbonates or hydroxides in the wastewater to form aluminum (or ferric) hydroxide or aluminum (or ferric) bicarbonate as a precipitate. As shown in Figure 4, the S/N ratio for alumina nanoparticle concentration was 32.1 at the level 1, 30.7 at the level 2, 32.4 at the level 3, and 31.8 at the level 4. Therefore, the best performance occurred at level 3 of this factor (concentration of 1.5 g/l) to reduce the TSS in wastewater.

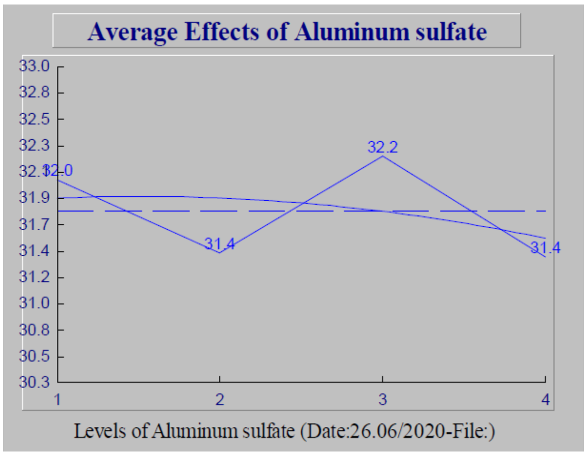

Fig. 3. Average of S/N ratio for different levels of aluminum sulphate concentration to reduce the TSS

The optimal conditions for achieving the highest efficiency of TSS reduction are presented in Table 4. The optimum concentrations of ferric chloride, iron oxide nanoparticles, aluminum sulphate, and alumina nanoparticles obtained 1.5 g/l for four coagulants. As illustrated, the optimal conditions did not occur in any of the 16 experiments. The TSS was measured equal to 47.3% under optimal conditions. This table also shows that the most effective factor in reducing the TSS was the alumina nanoparticles concentration (31.25%). After alumina nanoparticles concentration, the contribution of iron oxide nanoparticles concentration (28.52%), Ferric chloride concentration (22.97%) and aluminum sulphate concentration (17.26%) have the greatest effect on reducing the TSS in the wastewater, respectively. Because the surface to volume ratio in nanoparticles is high, so more surface is in contact with the colloids and the coagulation process is better. For this reason, the two nanoparticles of iron oxide and aluminum have better conditions than ferric chloride and aluminum sulfate for decreasing TSS in the wastewater [7 ,8].

This work was a comparative study between two coagulants and two nano-coagulants to obtain the optimal conditions for the treatment of industrial wastewater. The experiments for each coagulant were carried out by varying the dosage in concentration (0.4%-6% and 0.6%-2%) to find the optimum dosage. The supernatants were decanted and their values for TSS were measured. The analysis of variance and signal-to-noise ratio showed the optimum concentrations of ferric chloride, iron oxide nanoparticles, aluminum sulphate, and alumina nanoparticles was 1.5 g/l for four coagulants to reduce the TSS in wastewater, respectively. This affirms results of other studies that increasing the dosage of coagulant increases the treatability performance until agglomeration saturation is attained, whereby the performance starts to decline or stabilize [11, 12]. Coagulant concentration is one of the most important parameters on the performance of coagulation and flocculation process. Accordingly, low and high concentrations can have a negative impact on coagulation and flocculation performance and this performance is important in terms of process and economy. According to results of a study carried out by Duad (2015) by increasing the concentration of coagulants, removal rate of contaminants increased so that optimum concentration of aluminum sulfate and ferric sulfate for the removal of turbidity, COD, Color, and TSS was obtained from 120 to 200 mg/l [13]. whereas in this work, optimum concentration of ferric chloride, Iron oxide nanoparticles, aluminum sulfate, and Alumina nanoparticles to decrease TSS was obtained about 1.5 g/l. Since colloids have negative charges, they are absorbed faster by materials having the opposite charge and their settlement arrangements is provided by becoming weightier. The obtained results were in accordance with a study conducted by Duad et al. in 2015. When the concentration of coagulant is optimal beyond the limit, positive charges are formed around colloids for re-stabilization of colloidal particles and cannot be removed by prekinetic flocculation; thus, the removal efficiency comes down.

Conclusions

In this work, it was studied the effect of coagulants and nano-coagulants in different concentrations to reduce the TSS of industrial wastewater using the Taguchi statistical method. The analysis of variance and signal-to-noise ratio showed the optimum concentrations of ferric chloride, iron oxide nanoparticles, aluminum sulphate, and alumina nanoparticles was 1.5 g/l for four coagulants to reduce the TSS in wastewater, respectively. In addition, the most effective factor to reduce the TSS in industrial wastewater was the alumina nanoparticles concentration equal to 31.25%.

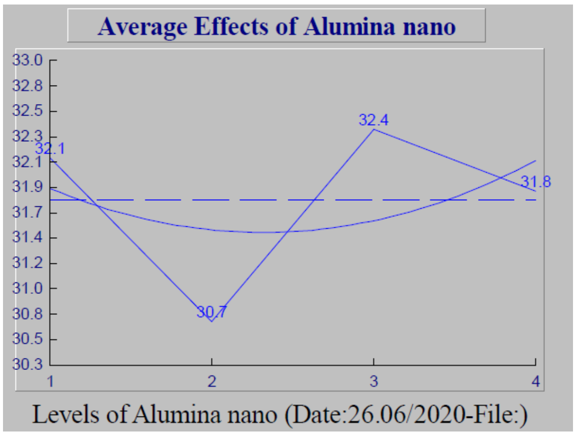

Fig. 4. Average of S/N ratio for different levels of alumina nanoparticles concentration to reduce the TSS

Table 4. Optimal levels and the contribution of factors in reducing the TSS

	Factors	Level Desc,	Level	Contribution(%)
1	Ferric chloride concentration (g/l)	1.5	3	22.97
2	Iron oxide nanoparticles concentration (g/l)	1.5	3	28.52
3	Aluminum sulphate concentration (g/l)	1.5	3	17.26
4	Alumina nanoparticles concentration (g/l)	1.5	3	31.25

References

[1] Patel, H. and Vashi, R.T. (2013) Comparison of naturally prepared coagulants for removal of COD and color from textile wastewater, Global NEST Journal 15, 522–528.

[2] Renaut, F., Sancey, B., Badot, P.M. and Crini, G. (2009) Chitosan for coagulation/flocculation processes, an eco-friendly approach, European Polymer Journal 45, 1332–1348.

[3] Ebeling, J.M., Sibrell, P.L., Ogden, S.R. and Summerfelt S.T. (2003) Evaluation of chemical coagulation/flocculation aids for the removal of suspended solids and phosphorus from intensive recirculating aquaculture effluent discharge, Aquacultural Engineering 29, 23–42.

[4] Amuda, O.S., Amoo, I.A. and Ajayi, O.O. (2006) Performance optimization of coagulant/ flocculant in the treatment of wastewater from a beverage industry, Journal of Hazardous Materials 129, 69–72.

[5] Babu, R. and Chaudhuri, M. (2005) Home water treatment by direct filtration with natural coagulant, Journal of Water and Health 3, 27–30.
[6] Ghorbani, H.R. and Molaei, M. (2017) Antibacterial nanocomposite preparation of polypropylene-Silver using Corona discharge, Progress in Organic Coatings 112, 187–190.
[7] Ghorbani, H.R., Alizadeh, V., Parsa Mehr, F., Jafarpour golroudbary, H., Erfan, K. and Sadeghi Yeganeh, S. (2018) Preparation of polyurethane/CuO coating film and the study of antifungal activity, Progress in Organic Coatings 123, 322–325.
[8] Ghorbani, H.R. and Molaei, M. (2017) Optimization of coating solution for preparation of antibacterial copper-polyethylene nanocomposite, Materials Research Express 4, 065017.
[9] Irfan, M., Butt, T., Imtiaz, N., Abbas, N., Ahmad Khan, R. and Shafique, A. (2017) The removal of COD, TSS and colour of black liquor by coagulation–flocculation process at optimized pH, settling and dosing rate, Arabian Journal of Chemistry 10, S2307–S2318.
[10] Abu Hassan, M.A., Li, T.P. and Noor, Z.Z. (2009) Coagulation and flocculation treatment of wastewater in textile industry using chitosan, Journal of Chemical and Natural Resources Engineering 4, 43-53.
[11] Nasr, F.A., Abdelfattah, I. and Shana, A. M. (2019) Cost-effective Physicochemical Treatment of Carpet Industrial Wastewater for Reuse, Egyptian Journal of Chemistry 62, 609- 620.
[12] Naik, S.S. and Setty, P.Y. (2011) Optimization of parameters for denitrification of wastewater using fluidized bed bioreactor by Taguchi method, International Journal of Biotechnology Applications 3, 97-101.
[13] Daud Z, Awang H, Aziz AbdulLatif A, Nasir N, Ridzuan MB, Ahmad Z. Suspended solid, color, cod and oil and grease removal from biodiesel wastewater by coagulation and flocculation processes. Procedia Soc Behav Sci. 2015; 195: 2407–2411