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Optimum turbidity removal by coagulation/flocculation methods from wastewaters of natural stone processing

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ABSTRACT

Recent expansions of stonecutting industries have led to increased water contamination. In this study, ferric chloride ($FeCl_3$, $6H_2O$), alum (Al_2 $(SO_4)_3$, $16H_2O$), and aluminum chloride ($AlCl_3$) are used as coagulants additives to treat wastewater of stone cutting industries. Response surface method (RSM) is used to optimize the operating variables such as coagulant dosage and pH to remove Total Suspended Solids (TSS). Additionally, one of the common RSM algorithms called central composite design is used to design laboratory analyses networks. Eleven tests are designed and conducted for each of three coagulants. Results show that the highest removal efficiency of TSS and turbidity using coagulant ($FeCl_3$, $6H_2O$) at pH 10.12 and dosage of .519 (g/l) is 88% and residual turbidity equals to 19 NTU. Alum (Al_2 $(SO_4)_3$, $18H_2O$) shows the optimum performance at pH 10.62 and dosage of 1.092 (g/l) with a rate of 87% removal of TSS and residual turbidity 18 NTU. Furthermore, coagulants ($AlCl_3$) at pH 9.23 and dosage of 0.780 (g/l) indicates the optimum removal efficiency of 80% TSS and residual turbidity of 19 NTU. Three coagulants used in this study have shown good results in the basic pH too.

Keywords: Treatment; Stonecutting wastewater; Ferric chloride; Alum; Aluminum chloride

1. Introduction

The increasing growth of stonecutting industries and extensive use of all types of stones in the construction projects has increased its wastewater by 25–40%, which requires an accurate environmental management [1]. An important issue in construction stone processing is the treatment and recycling the resulted mud water which is expensive. Suspending sediments in wastewaters of stonecutting industries could decrease the efficiency of machineries causing the darkness of stone and preventing the proper polishing and shining. Also, it could clog the pipelines. Two methods of traditional and modern are used for water recycling of stonecutting factories. Traditional method needs numerous ponds for stonecutting wastewater and also a vast area of land for mud collection. Modern method uses the filter press system and chemicals; hence, it does not need the construction of water ponds and mud treatment pools. Its benefit is returning unused land of factory that can be used for other beneficial purposes. To have a sustainable development strategy, environmental remedies is needed to control water pollution. Coagulation process

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is extensively used in wastewater treatment as it is efficient and simple to operate [2,3]. In this process, many factors may influence on the efficiency, such as type and dose of coagulant [4–8], pH [8–12], speed of mixture and timing [13,14], and temperature [15,16].

The main purpose of coagulation process is to use the polymer coagulants for the flocculation of outspread particles. Such coagulants have different types as their most important specifications are molecular weight and charge density [17,18]. Coagulants change the flocculated particles to greater particle size and lead their sedimentation. Connections of polymer and charge neutralization are two common mechanisms in flocculation. The flocculants are known to adsorb on particle surface through hydrogen bonding, ion binding, electrostatic interactions, chemical bonding, hydrophobic interactions, and van der Waals forces [18–22]. Ching et al. [23] analyzed the dynamics of coagulation of kaolin particles by ferric chloride. In another study, Bayraktar et al. [24] used marble solution for the flocculation of suspended particles and confirmed that the anion polymer with high molecular weight leads to better results for water purification and sedimentation rate. Poon and Chu [25] analyzed the use of ferric chloride and anion polymer in sedimentation and coagulation process by chemicals (CAPS). Seyrankaya et al. [26] studied the effect of pH and type of polymer on marble flocculation conditions and observed that they are ineffective, while the weak ionized anion flocculants had better function for water sedimentation and purification rate. Nishkov and Marinov [27] indicated that 31% anion polymer offers better sedimentation rate on Bulgarian marble.

The effects of pH and density of polyacrylate anions (PAA) on behavior of suspended particles coagulation of two types of travertine and marble stones by settlement rate and turbidity index were analyzed by Ersoy [28]. The results showed that density of polymer charge and pH of solution plays critical role in flocculation of stonecutting wastewater. The type of natural stone has an important effect on the sediment hydraulics but is ineffective on the turbidity of fresh water. In marble solution, anion polymer-34% has the best function of flocculation in terms of precipitate velocity and also turbidity. In travertine solution, anion polymer-28% gives the best precipitate velocity, while anion polymer-34% gives the best treating in lower doses. High-pH doses accelerate the precipitate velocity of both types of suspended particles of natural stone and increase the turbidity of fresh water. For acceptable turbidity, we may select pH up to 11, anion polymer-28% for the flocculation of travertine solution, and pH up to 11 and polymer-34% for marble solution. Physicochemical treatment of wastewater resulted from the marble processing was analyzed by Jar's test machine in room temperature (20°C) [29]. Agrofloc 100, $FeCl_3$ and $Al_2(SO_4)_3$ were used as coagulant. The coagulant dose was selected at 100, 200, 300, 400, and 500 ppm, and the results showed that all doses of Agrofloc 100, $FeCl_3$ and $Al_2(SO_4)_3$ are good for coagulation and flocculation. Nasserdine et al. [30] studied environmental management of stonecutting industry in Herbon. Ten experimental plans were produced in the different places of Herbon for reducing and removing stonecutting wastes. The analysis of current methods and Jar test was used for the optimization of water recycling and treatment facilities. Based on the optimized doses and turbidity results, it was confirmed that the required turbidity may be reached by using the flocculants by optimized rate of 1.5 mg/l. Completion of trial projects removed the output wastewater of stonecutting and improved the quality of water recycled by 44–99%. This is itself a factor for reducing long-term costs of each factory on the study. The turbidity may be result of colloidal particles from different resources including organic colloidal [31–34], textile colors [35], and solid particles in sizes in the wastewater of different industries [25,36]. Flocculation was not sufficient for treatment of wastewater, while the methods of flocculation plus coagulation perform better treatment. Among the coagulants used, the best result was met at pH 6 and 9 and $AlCl_3$ in terms of turbidity removal from the wastewater of natural stone processing by flocculation [37]. Kushwaha et al. [38] studied the use of inorganic metallic coagulants including aluminum sulfate and iron sulfate in the treatment of diary wastewater. The results showed that in optimized pH 8, using iron sulfate by 800 mg/l and aluminum sulfate by 500 mg/l removed COD by 69.2 and 66.5%. Aguilar et al. [39] reported that when the aluminum sulfate and iron sulfate are used together, the quality of treatment increases. Basaran and Tasdemir [40] determined specifications of natural stone powder coagulation in the presence of different polymers. Jar test was used for determining its coagulation behavior. The test was performed in the presence of different polymers with different doses and pHs. Further studies showed that aluminum chloride ($AlCl_3$) has the best efficiency in the removal of turbidity to the aluminum sulfate and iron sulfate [37].

In this study, optimum coagulant and dose of remedies and optimum pH of wastewater was analyzed through sampling the sludge of different stonecutting factories. Samples were tested with different chemical materials as coagulant. Response surface method (RSM) also was used for optimization. RSM is an algorithm of mathematical and statistical techniques for experimental modeling, evaluating the effect of several factors and searching the optimized conditions [41,42]. RSM has been used for the calculation of optimized parameter in the different processes [43,44]. The experimental procedure executed in environment laboratory at KNTU. The results obtained from the tests will be first classified and then analyzed by Minitab and Matlab software. Experimental model will be developed and finally the accuracy of it would be verified.

2. Method and materials

2.1. Experimental site

The main center of natural stone processing is in Shamsabad Industrial city, Tehran province. It is located near to Hassanabad City in Fashapouyeh district of Ray, with latitude of 35˚21´6´´ and longitude of 51˚13´9´´, 40 km South of Tehran. Its altitude is close to 965 m. Its short distance to the Tehran metropolitan have imposed an important role of transferring none environmentally friendly industries out of Tehran. Shamsabad serves more than 300 natural stone cutting and processing factories [32].

2.2. Wastewater sample analysis

Wastewater used in this study was collected from the stonecutting industries of Shamsabad in summer 2014. The collected samples are from Sirjan porcelain processing by 20-liter plastic gallons and carried to the laboratory. They were filtered by 0.1-mm mesh. The gallons were shaken well before samples preparation for performing each test. The samples were kept isolated in the laboratory. The physical and chemical analysis of the wastewater is shown in Table 1.

The laboratory analyses of oxide elements were performed by X-ray fluorescence (XRF). Table 2 shows the XRF analyses. The results were reported without considering probable loss of ignition (LOI) of samples. LOI is a test used in XRF major element analysis which consists of strongly heating a sample of the material at a specified temperature, allowing volatile substances to escape or oxygen is added, until its mass ceases to change.

Table 1
Physical and chemical analyses of the wastewater

TS (g/l)	TSS (g/l)	TDS (g/l)	Turbidity (NTU)	pH
13.48	3.51	9.97	1,275	7.27

Table 2
Oxide elements of wastewater

Oxide elements	wt%
CaO	95.6
MgO	0.89
Na_2O	0.51
SiO_2	0.33
Fe_2O_3	0.28
Al_2O_3	0.58
K_2O	0.015

2.3. Supplement chemicals

The chemical materials used in all tests are presented as follows. It shall be noted that all chemical materials used are made by Merck Co. (Germany). H_2SO_4 (1 M), NaOH (1 M), $18H_2O$, $Al_2(SO_4)_3$, $FeCl_3$, $6H_2O$, $AlCl_3$. The compounds have iron and aluminum as traditional coagulants in water and wastewater treatment industry. We used alum, ferric chloride, and aluminum chloride as coagulant in this study. The coagulants in previous researches had acceptable results for removing the suspended particles and turbidity. Distilled water was used in the study and was produced by using Pars Azma machine. Jar test machine with six steel blades, made by Zag Chemistry Co., with capability of adjusting speed and duration time. Jar test machine was used in this research for quick and gradual mixtures of flocculation process. All chemicals were weighted by using balance model MP-300G, made by Japanese AND Co., with accuracy of 1 mg. All pH measurements were measured by portable Martini Instrument Co., model MI805 with accuracy of 0.01. The remained turbidity after tests was measured by Lovibond machine, model PCcheckit with accuracy of 0.1 NTU.

2.4. pH and coagulant dose optimization

In this research, Central Composite Design, (CCD) and RSM were used for optimizing two important variables including dose of coagulant and pH. Before designing the tests, few introductory tests were performed to find the more suitable efficiency of coagulant and pH dose. Then, the different efficiencies were chosen for coagulant and pH dose for the different coagulants. For ferric chloride, coagulant dose was selected from 0.3 to 1 g/l and pH from 6.5 to 9.5. For alum, the dose of coagulant was chosen from 0.3 to 1 g/l and pH from 6.5 to 9.5. Totally, 33 tests were performed for all coagulants. It means that 11 tests were performed per coagulant. Standard jar with six steel blades, made by Zag Chemical Co., with capability of

Table 3
Mixing speed and mixing durations in different tests

Factor	Limits	Value
Mixing speed (rpm) Fast	80–250	200
Mixing time (min)	1–5	2
Mixing speed (rpm) Slow	30–60	60
Mixing time (min)	10–55	15

setting speed of rotation and time for mixture was used for the tests. Matlab and Minitab software's were used for the analysis of the obtained results (Table 3). By developing a model for the laboratory results, the derivation and verification of Quadratic models was analyzed for four methods performances. Finally, the process was optimized in RSM to determine optimized conditions of TSS and turbidity removal.

3. Results and discussions

3.1. Ferric chloride

The tests were designed in CCD method in 0.3–1 g/l and 6.5–9.5 for ferric chloride as coagulant and pH dose. The TSS (suspended particles removal %) was determined as follows:

$$\mathrm{TSS}(\%) = \left(\frac{\mathrm{TSS_{in}} - \mathrm{TSS_{out}}}{\mathrm{TSS_{in}}}\right) \times 100 \quad (1)$$

where $\mathrm{TSS_{in}}$ is TSS before stonecutting wastewater treatment processes; and $\mathrm{TSS_{out}}$ is TSS after stonecutting wastewater treatment processes. Table 4 shows the results of 11 ferric chloride coagulant laboratory tests.

The optimum percentage of suspended particles removal by ferric chloride was obtained at 87.037% at pH 10.12 and ferric chloride of 0.65 g/l. Then, the results obtained from test were analyzed by MATLAB in RSM method and CCD design. The measured data correlated with quadratic polynomial model. In order to optimize the process, the results are described as follows:

$$Y = 79.115 + 3.808\,X_1 + 0.862\,X_2 + 1.513\,X_1^2 - 2.832\,X_2^2 - 1.997\,X_1\,X_2 \quad (2)$$

where Y is removal percentage of TSS, pH is given as X_1, and ferric chloride is given as X_2.

Fig. 1 shows the optimized point by model appears in base pH and middle coagulant dose in which pH 10.12 and dose 0.519 g/l. In this point, the maximum removal percentage of suspended particles was 87.864%. After test in the introduced optimized point by model, turbidity was obtained at 19 NTU and the removal percentage of turbidity was 98.15% according to the initial turbidity of wastewater at 1,275 NTU. Also, at the optimized point introduced by the model, the height of sludge after sedimentation was 9 mm, while the height of graded tube was 210 mm. Volume of sludge allocates about 4.28% of the sample volume in graded tube.

3.2. Alum

The suspended particles removal maximized at 83.127 when pH 10.62 and alum dose of 0.65. In the next step, the results obtained from the tests were analyzed by Minitab and Matlab software with RSM method and CCD design that is shown in Table 5. The measured data correlated with quadratic polynomial model in order to optimize the process.

Table 4
Results of ferric chloride coagulant tests

#	pH	Dosage (g/l)	Sediment depth (mm)	pH Final	Turbidity (NTU)	TSS (%)
1	9.5 ± 1	1 ± 1	8	7.25	41	81.319
2	9.5 ± 1	0.3 ± 1	5	7.15	20	84.783
3	6.5 ± 1	1 ± 1	7	6.50	24	77.500
4	6.5 ± 1	0.3 ± 1	5	6.61	19.9	70.973
5	10.12 ± 1.414	0.65	9	7.47	20	87.037
6	5.89 ± 1.414	0.65	5	6.90	20	76.549
7	8	1.141 ± 1.414	6	7.10	34	73.750
8	8	0.156 ± 1.414	3	7.24	21	72.455
9	8	0.65	6	6.90	20	77.889
10	8	0.65	5	6.90	23	79.435
11	8	0.65	6	6.83	21	80.019

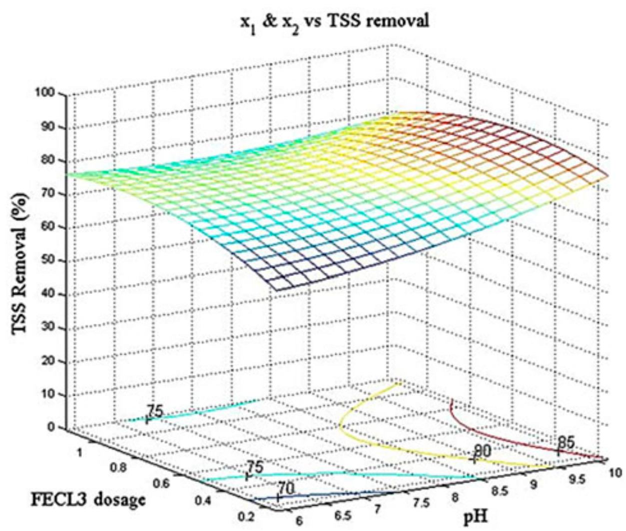

Fig. 1. Contour of ferric chloride coagulant for TSS removal percentage.

3.2.1. Alum in the first stage of analyses

In the first stage of analysis obtained for alum coagulant, all factors governing the proposed models, i.e. the 1st degree (X_1 and X_2) and 2nd degree parameters such as X_1^2 and X_2^2 plus interactions of two primary factors ($X_1 \times X_2$), were considered in order to reach a correlation on importance of such factors. The results are described as follows:

$$Y = 66.902 + 8.757\,X_1 + 4.475\,X_2 + 0.085\,X_1^2 - 5.195\,X_2^2 + 5.646\,X_1\,X_2 \quad (3)$$

Fig. 2 shows the optimized point is at base pH and medium alum dose in which pH 10.62 and alum 1.09 g/l. The optimum model shows maximum percentage of suspended particles removal is equal to 86.876%.

3.2.2. Alum in the second stage of analyses

In the 2nd stage of analysis of the results obtained for alum coagulant, the effect of 2nd degree parameter X_1^2 was not considered in the model. The factors governing the model, i.e. the 1st degree parameters, X_1 and X_2, and 2nd degree parameter such as X_2^2, and interactions of two effective factors ($X_1 \times X_2$), were considered in order to reach an optimum model, described as follows:

$$Y = 66.982 + 8.757\,X_1 + 4.475\,X_2 - 5.22\,X_2^2 + 5.646\,X_1\,X_2 \quad (4)$$

Fig. 3 shows the optimized point predicted by model occurs in pH base and high range dose in which pH 10.62 and alum of 1.092 g/l. In this point, the maximum percentage of suspended particles removal happens at 86.745%. After test in the introduced optimized point by model, turbidity was obtained at 18 NTU and the removal percentage turbidity was 98.588% according to the initial turbidity of wastewater at 1,275 NTU. In the optimized point introduced by the model, the height of sludge after sedimentation was 13 mm while the height of graded tube was 210 mm. Volume of sludge allocates about 6.19% of the sample volume into graded tube.

3.3. Aluminum chloride

According to the results, the optimum TSS% removal was 79.825 when pH 9.5 and aluminum

Table 5
Results of alum coagulant tests

#	pH	Dosage (g/l)	Sediment depth (mm)	pH Final	Turbidity (NTU)	TSS (%)
1	10(+1)	1(+1)	12	7.47	38	77.419
2	10(+1)	0.3(-1)	5	8.14	35	57.738
3	7(–1)	1(+1)	6	6.16	46	52.000
4	7(–1)	0.3(–1)	3	6.33	47	54.902
5	10.62(+1.414)	0.65(0)	18	8.50	18.3	83.128
6	6.38(–1.414)	0.65(0)	4	6.61	36	53.571
7	8.5(0)	1.141(+1.414)	8	6.20	36	64.516
8	8.5(0)	0.156(–1.414)	3	7.36	59	51.064
9	8.5(0)	0.65(0)	6	7.27	42	65.322
10	8.5(0)	0.65(0)	8	7.27	43	68.182
11	8.5(0)	0.65(0)	6	7.01	53	67.213

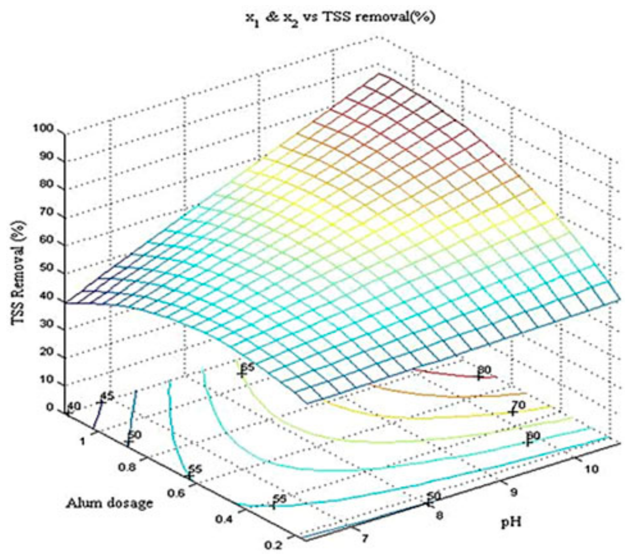

Fig. 2. Contour of alum coagulant TSS removal percentage (1st stage).

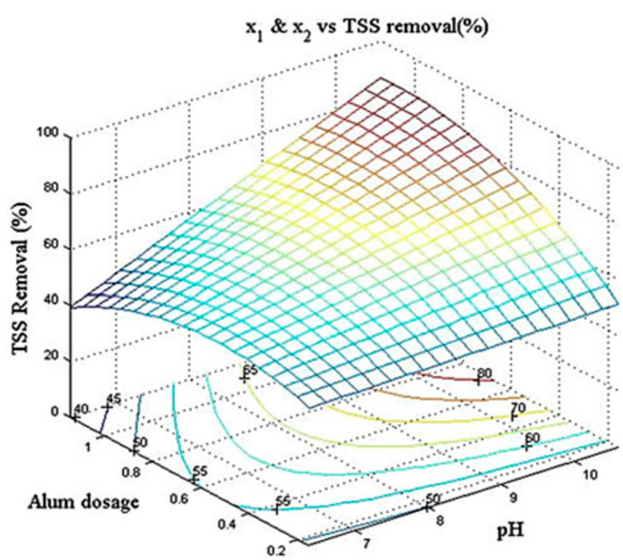

Fig. 3. Contour of alum coagulant TSS removal percentage (2nd stage).

chloride dose equals to 1. While the results at pH 10.12 and dose 0.65 are equal to 79.489% and at pH 8 and dose 0.65 equal to 79.32% that are very close. In the next step, the results obtained from the test were analyzed by Minitab and Matlab software in RSM method and CCD design. The measured data (Table 6) correlated with quadratic polynomial model in order to optimize the process (Eqs. (5) and (6)).

3.3.1. Aluminum chloride in the first stage of analyses

In the first step of analysis of results obtained for aluminum chloride coagulant, all governing factors which means 1st degree, X_1 and X_2, and 2nd degree parameters as X_1^2 and X_2^2, and also interaction of two effective factors ($X_1 \times X_2$) were considered to reach an optimum model, described as follows:

$$Y = 77.082 + 5.52\,X_1 + 1.711\,X_2 - 3.161\,X_1^2 - 2.504\,X_2^2 + 1.354\,X_1\,X_2 \quad (5)$$

Fig. 4 shows the optimized point simulated by the model at pH base and medium range dose in which pH 9.45 and dose of 0.884 g/l. In this point, Maximum removal percentage was 80.365%.

3.3.2. Aluminum chloride in the second stage of analyses

In the 2nd step of analysis of the results obtained for aluminum chloride coagulant, all governing factors such as 1st degree, X_1 and X_2, and 2nd degree parameters, X_1^2 and X_2^2 were considered in the analyses. But, the interaction of two effective factors ($X_1 \times X_2$) were not considered as influential factors. The optimum model is described as follows:

$$Y = 77.082 + 5.52\,X_1 + 1.711\,X_2 - 3.161\,X_1^2 - 2.504\,X_2^2 \quad (6)$$

Fig. 5 shows the optimized point simulated by the model occurs in pH base and top range dose in which pH 9.23 and dose of 0.780 g/l. In this point, the optimum percentage of suspended removal equals to 80.36%. After test in the introduced optimized point by model, turbidity was 25 NTU and removal percentage was 98.039% according to the initial turbidity of wastewater at 1,275 NTU. The parameter of pH*-dosage was obtained at $p = 0.16$ in the first step of the modeling, (p is the probability of results at $\leq \alpha = 0.05$). In second step, aluminum chloride was chosen as optimized coagulant in model for the removal of suspended solids.

According to the results from the selected models for ferric chloride coagulant, in both governing models, 1st and 2nd degree parameters plus interaction between two factors were impressive. Interaction pH*pH was not considered on the selected model for alum

Table 6
Results of aluminum chloride coagulant tests

#	pH	Dosage (g/l)	Sediment depth (mm)	pH Final	Turbidity (NTU)	TSS (%)
1	9.5(+1)	1(+1)	13	7.16	29	79.825
2	9.5(+1)	0.3(−1)	6	7.48	40	72.449
3	6.5(−1)	1(+1)	9	6.33	54	66.667
4	7(−1)	0.3(−1)	4	6.61	54	64.706
5	10.12(+1.414)	0.65(0)	14	6.91	25	79.487
6	5.89(−1.414)	0.65(0)	6	6.25	55	63.043
7	8(0)	1.141(+1.414)	11	6.23	34	74.118
8	8(0)	0.156(−1.414)	5	6.94	53	71.042
9	8(0)	0.65(0)	6	6.52	32	75.581
10	8(0)	0.65(0)	8	6.52	29	76.344
11	8(0)	0.65(0)	7	6.32	26	79.320

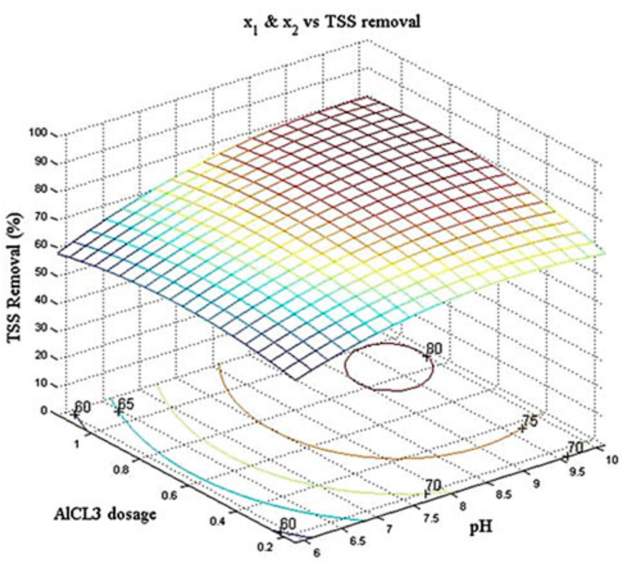

Fig. 4. Contour of aluminum chloride coagulant TSS removal percentage (1st stage).

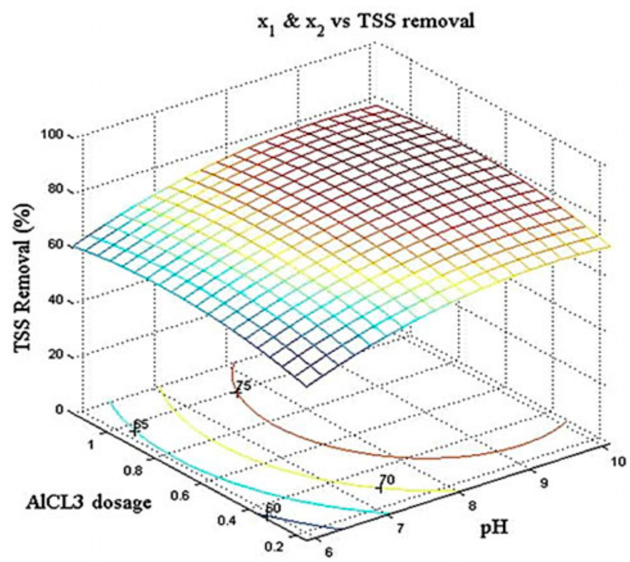

Fig. 5. Contour of aluminum chloride coagulant TSS removal percentage (2nd stage).

coagulant due to high value of p (results may be random). Moreover, pH*Dosage factor was not considered in the aluminum chloride coagulant due to high value of p in the model. Efficiency of ferric chloride coagulants with efficiency of 87.864% and alum with efficiency of 86.745% in removing of suspended particles is better than the aluminum chloride coagulant with 80.36%. For ferric chloride coagulant, the optimization point is at pH 10.12 and dose 0.519 g/l in which the removal of suspended particles was 87.864%. For alum coagulant, it is at pH 10.62 and dose 1.092 g/l in which the removal is 86.745%. And for aluminum chloride coagulant, it is at pH 9.23 and dose 0.780 g/l in which removal is 80.36%.

4. Conclusions

Efficiency of three types of coagulants was acceptable with respect to suspended particle removal and the height of generated sludge. As the height of sludge is low, less water is wasted. Therefore, ferric chloride coagulant with sludge height of 9 mm has the best result in comparison with alum with sludge height of 18 mm and also chloride aluminum with sludge height of 13 mm. Sedimentation velocity of suspended particles is high in high pH's according to the examiner's observations during the tests. Ferric chloride coagulant gave the best result with the lowest dose used compared to other two coagulants.

The optimized point of aluminum chloride coagulant is at pH with lower base value which needs less chemicals for setting pH. All three coagulants used in this study showed acceptable results in pH base, and the optimized points reached at pH base. The final pH (after coagulation and flocculation process) reaches to the neutral pH when initial pH is base in wastewater, which is good for reusing in stone cutting processes. Since the alkaline wastewater cause less problems than the acid wastewaters, the optimized points reaches at pH base for three coagulants. Compared to traditional method, RSM considers the effects of all factors with least laboratory experimental tests using the proper interaction of variables, reaching to optimum result.

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