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Pretreatment of Municipal Wastewater by Enhanced Chemical Coagulation

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ABSTRACT: The efficiency of existing municipal wastewater treatment plants has been affected by the increase of incoming organic load caused by the expansions in developing countries. In the present investigation, the effect of Chemically Enhanced Primary Treatment (CEPT) process was studied on the enhancement efficiency of a municipal wastewater treatment plant in Tehran, Iran. Jar test results showed an increase in COD, phosphorus, turbidity and TSS removal by the increase in dosage of aluminum sulfate and ferric chloride as coagulants. Results revealed COD, phosphorus, turbidity and TSS removals of 38, 66, 68 and 69 %, for alum at 80 mg/L and 60, 73, 49 and 48 % for ferric chloride at 70 mg/L as the optimum doses, respectively. Ferric chloride revealed more efficient results compared with alum. The result of tests to find the optimum pH of two coagulants revealed that pH of 8.2 for both chemicals is the optimum performance condition. Therefore, CEPT can be used as an efficient method in conventional municipal wastewater treatment plants to reduce the organic load of biological treatment and enhance the removal of nutrients.

Key words: Coagulant, COD, Jar test, Municipal wastewater, Phosphorus, Primary treatment

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INTRODUCTION

Nowadays, the treatment and disposal of pollutants, as one of the most important issues in environmental engineering, becomes even more complex given in place limit in terms of disposal options. From historical point of view in previous centuries (by the end of 1900) sufficient attention was not paid to the issue of wastewater treatment due to readiness in releasing untreated wastewater into recipient sources. But in the early of 20th. century, failure to provide wide and sufficient areas for disposal of untreated wastewater, particularly in mega cities, culminated into applying more effective methods in wastewater treatment (Metcalf and Eddy, 1991). Former experiences especially in 1980s revealed that wastewater treatment projects in developing countries not just requires technological facilities but these facilities could be established exploiting human and financial resources. Therefore, these projects are often constructed in big urban areas enjoying great human and financial sources and this fact limits their development into poor areas (Sonune and Ghate, 2004). To solve the above mentioned problems, Chemically Enhanced Primary Treatment (CEPT) is tenable as an appropriate, executive and effective method (Kurniawan, *et al.*, 2005). This technology not only brings proper and comparable results in terms of reducing the COD, turbidity and TSS in comparison with current systems, but also implies a very cost effective and productive method to upgrade the capacity of conventional plants (Olive, 2002). CEPT is a process in which the chemicals (generally metal salts or polymers) are added for pretreatment purpose. These chemicals conglomerate the suspending solid particles via coagulation and flocculation processes (Zhou, *et al.*, 2004). The accumulated particles or flocs have high sedimentation velocity and consequently the treatment performance of parameters such as COD, Phosphorus, turbidity and TSS will be improved. This process can be performed in sedimentation tanks of conventional treatment plants and is cheaper, simpler with higher performance comparing with conventional systems (Harleman and Morrisey, 1992; Harleman and Murcott, 1992, 2001a, 2001b). Among other advantages over conventional practices, it requires half of the necessary volume of sedimentation ponds in comparison to conventional methods, shows more discharge rate instead of smaller needed space required for installation of necessary facilities, has appropriate elimination function for wide range of wastewaters with various specifications, appropriately complies if being added to the various treatment facilities (Olive, 2002) and also bears numerous economic advantages in terms of production and exploitation (Harleman, *et al.*, 1997).

Chemical Enhanced Primary Treatment includes coagulation followed by sedimentation and removal of flocs in a sedimentation unit. Dispersed solids inside wastewaters include non sedimentary suspending materials or particles with very negligible sedimentation velocity, in which the colloids are as constituents of major part of non sedimentary particles. Since most of natural colloids have negative charges that expel the similar charges, these particles grant stability to a suspension. When a coagulant is added to wastewater, disintegrates and via hydrolyzing the metal ion, metal hydroxide ionic complexes with high positive charge will be formed. Since these complexes have high positive charges, they absorb to the surface of colloids and by reduction of negative charge, they are being made to be neutralized and condensed via Vandervalce forces. This absorption is strengthened by water turbulence (flocculation) and particles with proper sedimentation capability will be formed (Reynolds and Richards, 2000). Regarding the modality of phosphorus removal by coagulants it can be said that adding these chemicals to wastewater causes the cations of these salts to combine with anionic insoluble phosphates inside the suspension and creates insoluble metal phosphate. Given the light weight, this particle has not enjoyed sedimentary virtue yet and will be precipitated using flocculation (Daniels and Parker, 2003). In fact, upon adding coagulation salts, although wastewater receives Fe^{2+}, Fe^{3+} and AL^{3+}, phosphorus removal in pH lower than 6.5 will be accompanied with formation of insoluble substances of $AlPO_4$, $FePO_4$ and in pH higher than 6.5 with aluminum and Iron oxide and hydroxide (Irdemez, *et al.*, 2006; Jiang and Lioyd, 2000).

Numerous researches have already been conducted in various countries particularly in developing countries. In coastal resort city of Rivera in Brazil which is facing with four times increase in basic population in summer time the wastewater treatment plant always experienced problems. In 2000, Yu and Bourke implemented CEPT system using 50 mg/L of ferric chloride with 0.5 mg/L of anionic polymer as coagulant aid, which was able in 60 and 85% reduction of BOD and TSS contents, respectively. In another instance, in 2001, conducted study by Harleman and Morcutt on economic assessment of implementing CEPT system in Rio de janeiro treatment facilities revealed that this system not only dose not require ,major capital investments but also can increase the capacity of existing treatment facilities without any requirement to change the current plan of working systems (Olive, 2002). The objective of the implemented study in UK by Song, *et al.* in 2003 was to develop a treatment system that can effectively reduce the concentration of pollutants in tannery wastewater to environmentally acceptable levels and that can greatly reduce the cost of discharging the effluent. During coagulation process, in optimum pH 7.5, 30-37 % of COD and 38-46 % of TSS were removed by and ferric chloride as coagulants, respectively. Also, ferric chloride produced better results comparing with (Song, *et al.*, 2003). In 2003, Delgado, *et al.*, conducted an experimental laboratory scale study using aluminum sulfate, Ferric chloride, and Poly aluminum chloride to obtain the required water quality (3-5 NTU) in turbidity in the discharges of the secondary treatment from a conventional wastewater treatment plant in Spain. The Poly aluminum chloride showed the best performance in wastewater natural pH and 50 mg/L of coagulant dosage (Delgado, *et al.*, 2003). In 2005 another research was conducted by Mahvi et al. in Iran to study the CEPT system performance on the waste of largest Iranian industrial complex into the municipal wastewater treatment plant. Applying of lime and FeO_3 as chemical coagulants, reduced the BOD, COD and TSS levels 27-53, 25-59, 46-94 percent, respectively and showed that the pretreated wastewater can be discharged into municipal wastewater treatment plant in terms of quality (Mahvi, *et al.,* 2005). In 2005, Amuda et al. studied the performance of CEPT pretreatment process on the wastewater of an beverage industry in Nigeria, using $Fe_2(SO_4)_3.3H_2O$ as coagulant and also in conjunction with neutral Poly Acrilamide Polyelectrolyte. The results revealed that using coagulant at 500 mg/L dosage removes 78, 75 and 74% of COD, Phosphorus and TSS contents, respectively, and applying coagulant aid at about 25 mg/L dosage enhances the removal of said parameters 93, 96 and 94% ,respectively. The results of this experiment were reported as completely appropriate for biological refining stage (Amuda, *et al.,* 2005).

On the basis of the above discussion, the main objective of this research was to investigate the feasibility of treating municipal wastewater by CEPT process. This was achieved by conducting laboratory settleability studies and jar tests using alum and ferric chloride. Furthermore, the optimum conditions under which the wastewater would be treated were investigated.

MATERIALS & METHODS

The samples were taken from discharge of primary sedimentation ponds in a municipal wastewater treatment plant (with the system of active sludge for treatment) in Tehran, Iran. Sampling was done at 8.00 AM and the samples were put under CEPT process using Jar test. The consumed raw samples for two coagulants were taken concurrently in all series of the tests and were completely similar.

The Jar tests were conducted using 6 jar system made by HACH Company, USA. whose blades rotates by an electrical engine. Before starting the work, the raw wastewater temperature and PH were measured using digital PH meter EUTECH 30. In every test, one liter of completely mixed sample was sprinkled inside followed by addition of specified concentration of given coagulant to each jar. The coagulation was done through one minute rotation in 120 rpm immediately followed by 15 minutes rotation at 40 rpm and then left them for 30 minutes for full sedimentation in the jars. In the next stage, the samples were taken from depth of 5 cm of supernatant to measure the COD, Phosphorus, turbidity and TSS parameters. Then, the samples were transferred into the environmental laboratory, University of science and Technology in order to measure specified parameters. COD measurement was made by Spectrophotometer HACH, DR-4000 using Reactor Digestion HACH method No. 8000. The samples digestion took place for 2 hours in accordance with manufacture method by COD reactor. Turbidity was measured by turbidimeter HACH model 2100N and reported in NTU. TSS was measured using paper filter S&S 589, Black Ribbon, according to the TSS measuring Method (Nazeri and Ekhtiarzadeh, 1995). In the present research ferric chloride and aluminum sulfate or alum made by Merck, were applied as coagulants. Stock solutions of these two chemicals at 10000 mg/L were produced by mixing 10 grams of every chemical in distilled water and then adjusting the volume in to one liter. Consequently, all of these solutions were kept in dark and air tight containers away from light. For adjusting the pH the lime solution and one normal Sulfuric acid prepared from Merck made acid 95% were used in the series of tests intended for determining optimum pH of every coagulant.

Table 1. Raw wastewater characteristics in first stage

pH	Temp (°C)	COD (mg/L)	Phosphorus (mg/L)	Turbidity (NTU)	TSS (mg/L)
8.26	24.8	173.4	15.80	15.70	77.50

Table 2. Results of final pH by Alum and Ferric chloride in first stage

Dosage (mg/L)	pH	$FeCl_3$ Dosage (mg/L)	pH	$FeCl_3$ Dosage (mg/L)	pH
0	8.26	0	8.26	50	7.80
8	8.25	4	8.20	70	7069
25	8.14	6	8.16	100	7.54
70	7.90	9	8.13	140	7.39
120	7.71	14	8.06	180	7.24
200	7.50	30	7.92	220	7.09

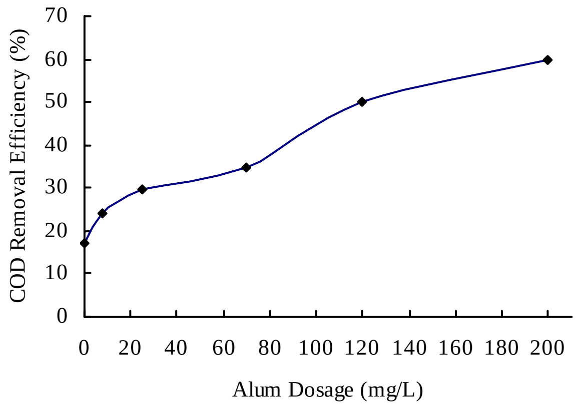

Fig.1. COD removal efficiency by Alum

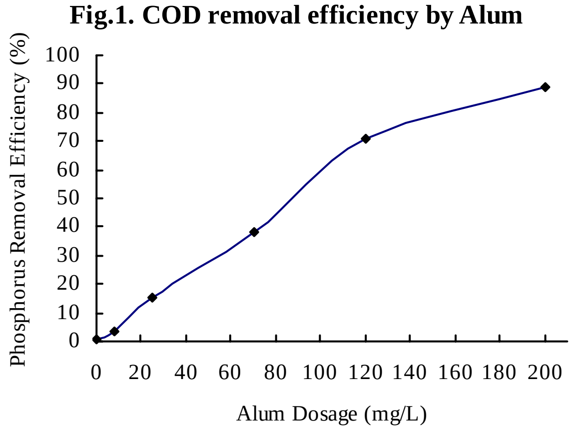

Fig.2. Phosphorus removal efficiency by Alum

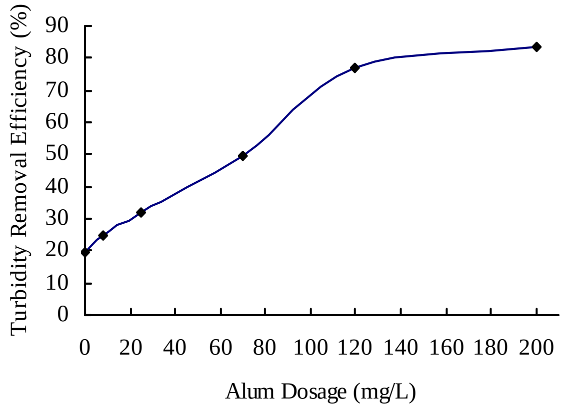

Fig. 3. Turbidity removal efficiency by Alum

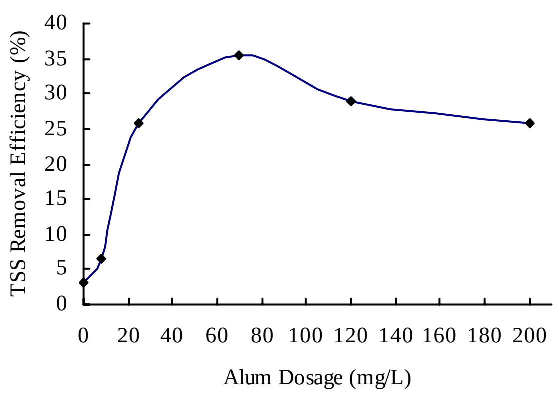

Fig. 4. TSS removal efficiency by Alum

RESULTS & DISCUSSIONS

In the present study, 2 stages of tests were considered in order to determine the optimum conditions for every coagulant. First stage included determining optimum dosage of every coagulant in natural pH of wastewater and in second stage the optimum pH of performance for every coagulant in resultant dosage from last stage was specified. Therefore, a series of Jar test in which alum dosage ranging from 0 to 200 mg/L and two series of Jar tests by ferric chloride dosage ranging 0 to 220 mg/L were tested for implementing the first stage of the tests. In Table 1 the specifications of raw wastewater, used in Jar tests at the first stage are shown and the resultant final pH by and ferric chloride are observable in Table 2.The results due to the removal efficiency of COD, phosphorus, turbidity and TSS by and ferric chloride are shown in Figs. 1-8, respectively.

As shown in Fig 1, increase in alum concentration increases removal efficiency of COD. The slope of curve in low alum dosages was higher in which a reduction in case of raise in concentration could be observed. In this way, by considering COD as removal objective, 80 mg/L is selected to apply to the process. In term of phosphorus removal in Fig 2, it can be observed that the slope of curve in low dosages of coagulant is higher than in case of increase in alum concentration. By selecting phosphorus removal as treatment goal, 100 mg/L could be applied to achieve the best results. In term of turbidity in fig 3, the process of increase in turbidity removal can be observed by increment of coagulant concentration, although the removal efficiency is almost steady due to alum dosage increment in high concentration of coagulant. In this case, dosage of 100 mg/L for alum reveals the best results for turbidity removal. Regarding TSS, as it could be seen in Fig .4, removal efficiency increases by alum concentration until 80 mg/L, although decreases gradually in higher concentrations. This feature could affect determining the optimum dosage of this coagulant which could be observed 80 mg/L from Fig .4. pH alteration process in Table 2 reveals reduction accompanied with increase of coagulant concentration. As the natural limit in treated wastewater pH is advised and the need to neutralize pH in case of defying from natural range, the final pH of pretreated wastewater will be effective in determining the optimum dosage of alum as coagulant. The results observed in removal efficiency of blanks (samples in which the coagulant's concentration is zero), particularly for two parameters of COD and turbidity are considerable. As it can be observed in Fig. 1 for blank, 17 % of COD reduction is being observed just by mixing and sedimentation, although in the same sample the parameter of turbidity showed removal of 20%. The removal in phosphorus and TSS are 1 and 3 % in blank, respectively. It looks that the performance of primary sedimentation system in the studied treatment plant is not effective properly and more organic matter removal could be obtained by enhancement of primary sedimentation system performance.

Another important point is the high removal of phosphorus obtained upon applying alum, which reached to 72 and 89% at 120 and 200 mg/L, respectively. To explain this, it can be said that although the chemistry basis of phosphorus removal by alum is not completely known, but it reduces possibly via occurring the complex reactions and absorption by flocs. In this way, the phosphorus will be removed by formation of insoluble sediment of $AlPO_4$ .In fact; it is assumed that $AlPO_4$ will be trapped inside the floc and then will be precipitated. But another important point is that the phosphorus removal is dependent to pH and the optimum pH for this removal by , as Reynolds and Richards explained is 5.5 to 6.5 (Reynolds and Richards, 2000). Given the final pH results in Table 2, it could be said that increasing of alum dosage, coincided with pH reduction and this in turn increased the phosphorus removal by alum dosage increment. This result is justified by Song et al. in England by using alum and ferric chloride as coagulant for an industrial wastewater treatment in 2003. Results of their Experiments as shown in fig. 5 revealed pH reduction by increase of coagulant dosage (Song, *et al.*, 2003).

In results of tests by ferric chloride as coagulant, according to Fig. 6, the rate of COD removal increases in lower dosages of coagulant and fells down by increasing in concentration. The important point in this curve is high removal efficiency of ferric chloride in low concentrations. Hence, by adding just 9 mg/L of chemical coagulant, 40% of COD removal is tenable. In this case, by considering COD as removal objective, 50 mg/L is selected to apply to the process. From Fig.7 in phosphorus removal, it could be observed that the curve is steeper in high concentrations of ferric chloride than low dosages, although at higher concentrations is going to reach to a relatively steady state. The mechanism goes for turbidity removal, the TSS removal reduces with increase in concentration of coagulant, as in 220 mg/L, 64% increase in TSS comparing with raw sample is measured. Range of 50-70 mg/L for coagulant dose seems to be optimum for TSS removal. Observing the pH alterations process in Table 3, indicates that like alum, it reduces with increase in coagulant concentration as it was observed by the experiment results of Song, *et al.* in 2003 and its impact should be considered as a limit factor in determining the ferric chloride optimum dosage.At the end of this stage of tests, as the results showed, by considering COD removal, as one of the most effective methods for performance enhancement of aeration tanks and phosphorus removal for reducing possibility of eutrification, 80 and 70 mg/L were selected as optimum dosage of alum and ferric chloride in wastewater natural pH, respectively.

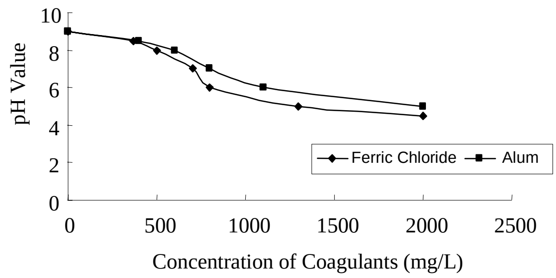

Fig. 5. Results of Final pH amounts by Song et al. 2003

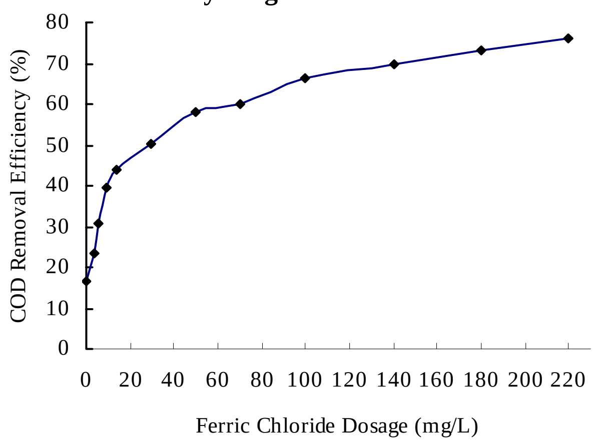

Fig. 6. COD removal efficiency by $FeCl_3$

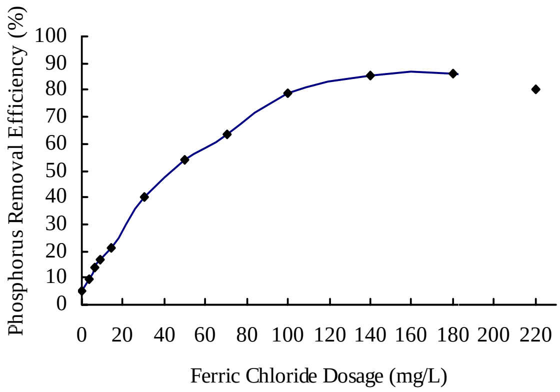

Fig. 7. Phosphorus removal efficiency by $FeCl_3$

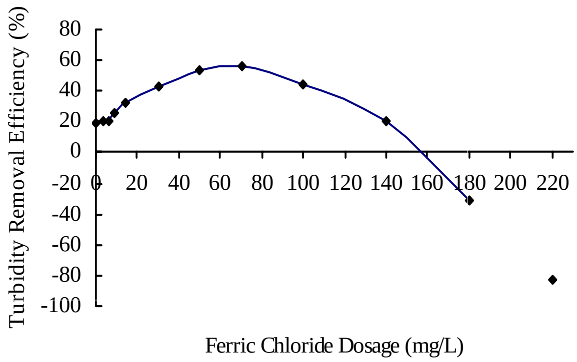

Fig. 8. Turbidity removal efficiency by $FeCl_3$

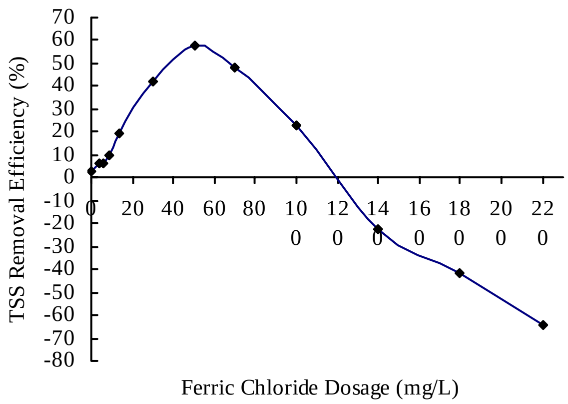

Fig. 9. TSS removal efficiency by $FeCl_3$

Table 3. Row wastewater characteristics in second stage

pH	Temp. (°C)	COD (mg/L)	Phosphorus (mg/L)	Turbidity (NTU)	TSS (mg/L)
8.21	27.3	161.8	20.78	22.20	155.00

It should be noted that selection of optimum dosage for every coagulant depends on parameters for treatment. But in the second stage of the tests for determining the optimum performance pH for each coagulant in optimum concentrations from previous stage, a series of Jar tests were implemented for alum and ferric chloride. Given that the best pH for alum is in the range of 4.5-8 and for ferric chloride is 4-12 (Reynolds and Richards, 2000), the amounts of 4, 5, 6, 7, 8, and 9 for alum and 5, 6, 7, 8, 9 and 10 for ferric chloride were selected as pH ranges for this stage of tests. It is notable that in each series of Jar tests, one Jar contained wastewater with natural pH namely near to 8 was tested without addition of any chemicals to adjust pH. In Table 3, the specifications of raw wastewater used in jar tests in second stage and in Table 4, the final obtained pH by alum and ferric chloride are shown. Also, the COD, Phosphorus, turbidity and TSS reduction results are shown in figures 9 to 12 for alum and 13 to 16 for ferric chloride, respectively.

In Fig. 10. COD removal by alum at 80 mg/L is shown, which describes that by increase in pH COD removal shows deep steep at first followed by steady steep indicating that natural pH (8.21) or 9 is the optimum pH. Selection of such range for optimum pH is confirmed by observing figures 11 for phosphorus, 12 for turbidity and 13 for TSS. From the Figs., it can be observed that at optimum pH (8.21), 38% of COD, 66% of phosphorus, 68% of turbidity and 69% of TSS could be removed. This logic governs the selection of 8.21 (natural pH) as optimum is completely economical given that there is no need to apply pH escalating facilities at the start of process and then pH neutralizing facilities at the end of process. Although it should be noted that selecting this case as the optimum situation not only dose not need pH alteration facilities but also shows no great difference with a situation deriving out of pH 9. Optimum performance of alum in the range of natural pH is confirmed by experimental results of Song, *et al.* in 2003 in England as shown in Fig.14 (Song, *et al.*, 2003). In Fig. 15, the removal of COD by 70 mg/L of ferric chloride indicates the deep steep of removal curve in low pHs and steady state in higher pHs revealing that the optimum pH ranging from 8.21 (natural pH) to 9.98. This issue is confirmed by observing figures 16 for phosphorus, 17 for turbidity and 18 for TSS. The removal efficiency obtained from the figures at optimum pH (8.21) for COD, phosphorus, turbidity and TSS is 60, 73, 49 and 48%, respectively.

Table 4. Results of final pH by alum and Ferric chloride in second stage

Adjusted pH (Coagulant: Alum)	Final pH	Adjusted pH (Coagulant: $FeCl_3$)	Final pH
3.57	3.57	4.83	3.19
4.97	4.20	6.19	6.02
5.95	5.36	6.90	6.95
7.18	7.25	8.21	7.72
8.21	7.87	8.99	8.10
9.00	8.39	9.98	9.40

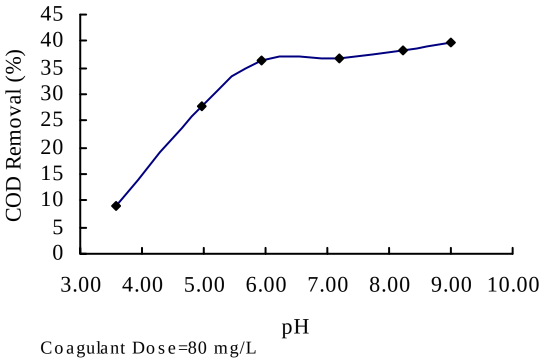

Fig. 10. COD removal efficiency by Alum in different pHs

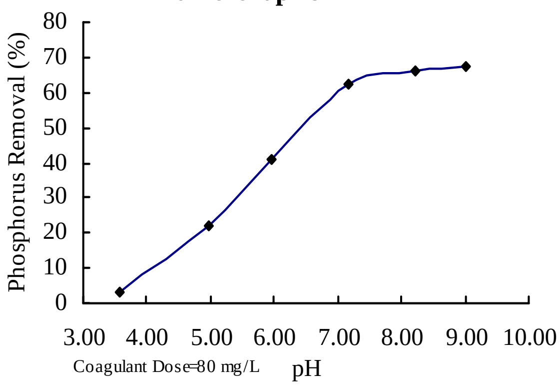

Fig. 11. Phosphorus removal efficiency by Alum in different pHs

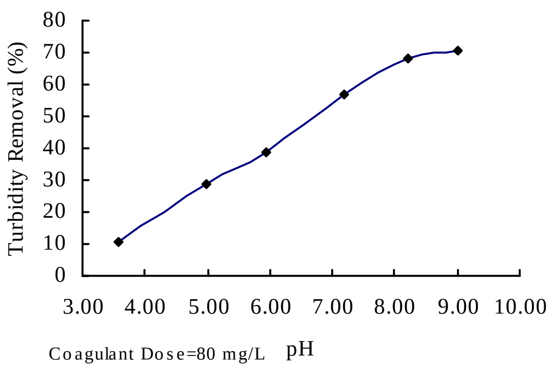

Fig. 12. Turbidity removal efficiency by Alum in different pHs

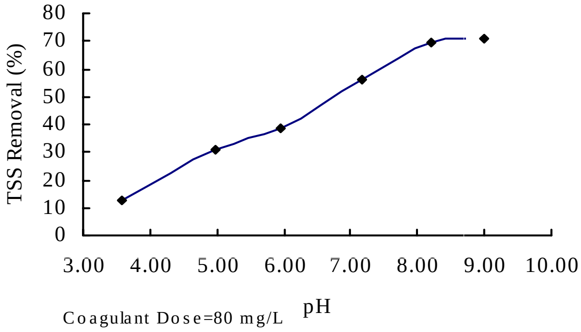

Fig. 13. TSS removal efficiency by Alum in different pHs

At the end of this experimental research, by considering some limits in removal of turbidity and TSS, results indicate more efficiency for COD and phosphorus removal by ferric chloride rather than alum. As shown in Fig. 20, this is confirmed by the experimental results of Song et al. by alum and ferric chloride as coagulants in 2003 in England (Song, *et al.*, 2003).

Better performance of ferric chloride comparing with alum can be verified by economical comparison. It can be observed that with the price of 7500 and 5000 Rials per kilogram for alum and ferric chloride respectively and selected dosage of those coagulants, total cost of pretreatment with ferric chloride would be less than that for alum. By this way, 70 and 80 mg/L are selected for alum and ferric chloride as optimum coagulant dosages, respectively and 8.21 (natural wastewater pH) is known as optimum pH for performance of both coagulants.

CONCLUSION

The experimental studies of CEPT process by alum and ferric chloride in municipal wastewater indicate that ferric chloride has more efficiency for COD and phosphorus removal in the same concentrations rather than alum. Although it shows some limits in removal of turbidity and TSS and in higher dosages the final turbidity and TSS exceeds even from the presented amounts in raw wastewater. Therefore, selection of appropriate coagulant and also optimum conditions of its performance largely depends on elected parameters that should be removed. Given all four parameters COD, phosphorus, turbidity and TSS as removal purpose, the dosage of 80 mg/L for alum and 70 mg/L for ferric chloride could be suggested as the optimum concentrations in natural pH of wastewater. It should be noted that test results indicate the natural pH as the optimum performance pH for both coagulants given the necessity of neutrality of discharged treated wastewaters. One of the other important results of this research via using ferric chloride as coagulant is that in lower rates (less than 10 mg/l) it can show appropriate results. With the current price of alum and ferric chloride and selected dosage of those coagulants, total cost of pretreatment with ferric chloride would be less than that for alum. Therefore, if the obtained removal results for given parameters in treatment plants are sufficient; this chemical could be applied in CEPT process with many economical justifications. Therefore, Chemical Enhanced Primary Treatment could be implemented as an effective method in municipal wastewater treatment improving the imposed load to aeration ponds, reducing the nutrient concentration and improving the efficiency of treatment.

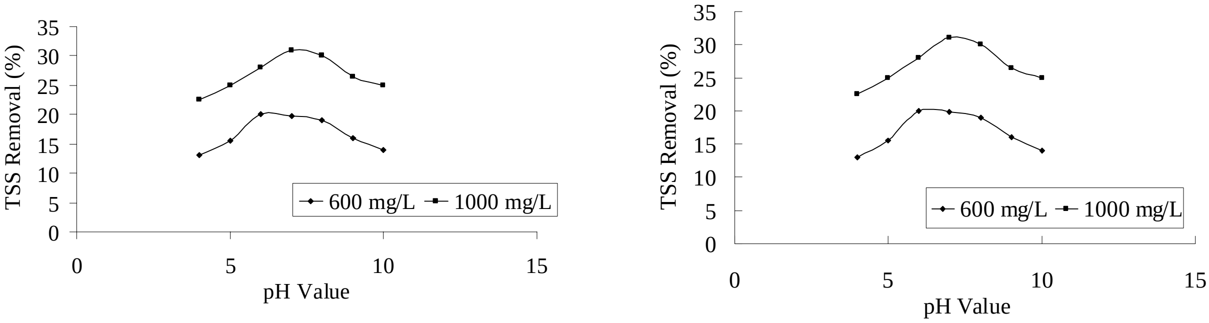

Fig. 14. COD and TSS removal by alum in different pHs (Song *et al.*, 2003)

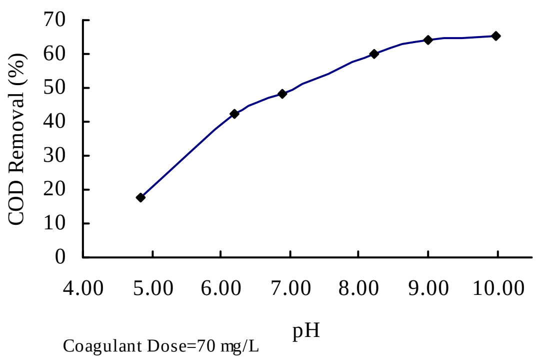

Fig. 15. COD removal efficiency by $FeCl_3$ in different pHs

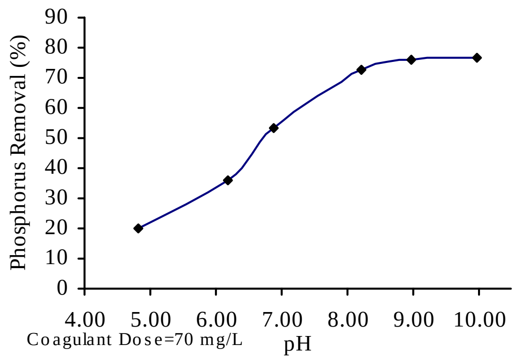

Fig. 16. Phosphorus removal efficiency by $FeCl_3$ in different pHs

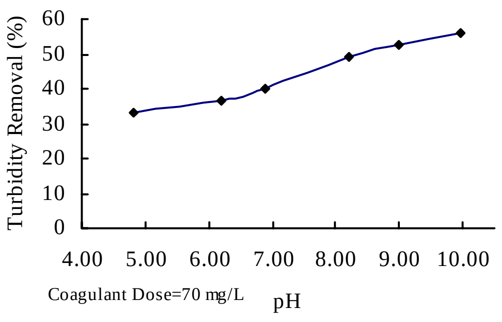

Fig. 17. Turbidity removal efficiency by $FeCl_3$ in different pHs

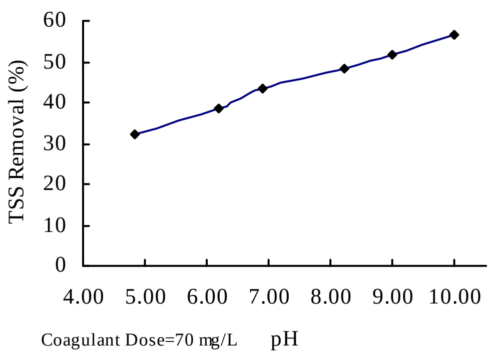

Fig. 18. TSS removal efficiency by $FeCl_3$ in different pHs

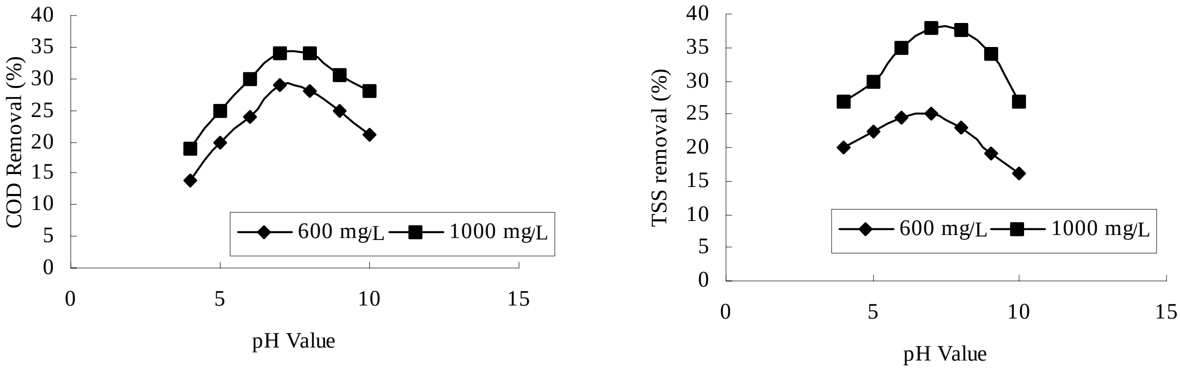

Fig. 19. COD and TSS removal by ferric chloride in different pHs (Song, *et al.*, 2003)

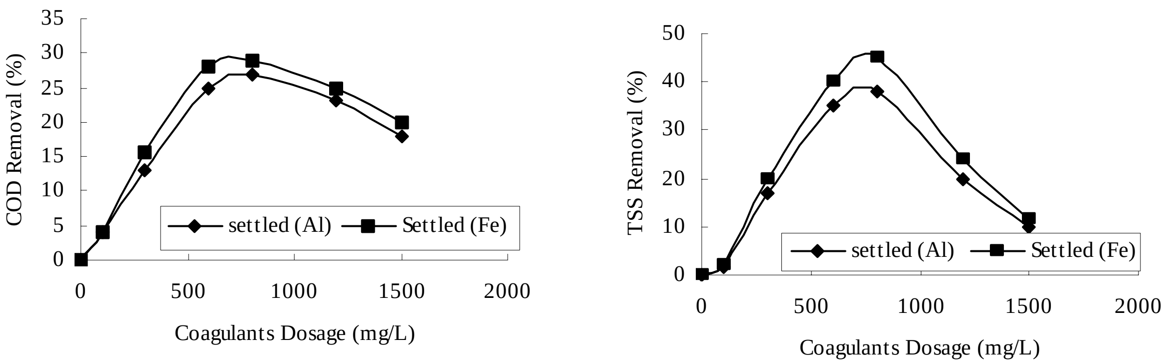

Fig. 20. COD and TSS removal in different dosages of coagulants (Song, *et al.*, 2003)

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