Comparative Efficacy of Coagulation-Flocculation and Advanced Oxidation Process (AOP: Fenton) for Textile Wastewater Treatment

Atkeeya Tasneem[1], Protima Sarker[1*] and Md. Khabir Uddin[2]

[1]*Department of Environmental Science and Disaster Management, Noakhali Science and Technology University, Noakhali-3814, Bangladesh.*
[2]*Department of Environmental Sciences, Jahangirnagar University, Dhaka-1342, Bangladesh.*

Authors' contributions

This work was carried out in collaboration among all authors. Author AT designed the study, Investigated and wrote the protocol and the first draft of the manuscript. Author PS managed the literature search, reviewed and revised the manuscript. Author MKU supervised and managed the analyses of the study. All authors read and approved the final manuscript.

Original Research Article

ABSTRACT

Textile effluent contains dye and harmful chemicals which are difficult to remove. This study was conducted to find out the cost effective and efficient textile effluent treatment process. In this regard, coagulation-flocculation and Fenton treatment process were applied in which pH, Dissolve Oxygen, TOC (Total Organic Carbon), COD (Chemical Oxygen Demand), TDS (Total Dissolve Solid) and EC (Electro-conductivity) were analyzed before and after the treatment. Conventional Jar test was used for coagulation and flocculation process where chemical coagulants ($FeSO_4$ and $Ca(OH)_2$) were added to different doses. On the other hand, Hydroxyl (OH.) radical, a byproduct of reaction between hydrogen peroxide (H_2O_2) and ferrous sulphate ($FeSO_4$) was used in Fenton treatment processas a strong oxidant capable of oxidizing various organic compounds. In coagulation-flocculation process the maximum reduction of pH (6.3-6.8), TDS (400 mg/l), EC (1110 μs/cm), COD (86.56%) and TOC (97.81%) was found after using dose 1 (1 g of FeSO4 and 1 g of

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*Corresponding author: E-mail: protima_das37@yahoo.com;

Ca(OH)2) which was within Bangladesh standard value. And DO (Dissolves Oxygen) was increased maximum 4.4 mg/L than the untreated (1.2 mg/L) effluent. In Fenton treatment process, maximum TOC removal efficiency was found at pH-1 as 89.45%. The COD content was also remarkably reduced as 63.98% at pH-2. It was observed that in comparison of removal efficiency coagulation-flocculation process was more efficient than Fenton treatment process. Concurrently both processes were proved very cost effective, time saving and easily operative for industrial use.

1. INTRODUCTION

Textile industries are one of the largest users of water and produce a large amount of wastewater during textile processing such as, dyeing and finishing process [1]. These industries produce complex wastewater containing dyestuff, surface-active materials and textile additives also [2]. Waste water generated from this industries contains high color, high biochemical oxygen demand (BOD), chemical oxygen demand (COD), pH, temperature, turbidity and toxic chemicals [3].

In Bangladesh, it was estimated that about 3000 garment factories are operating in Dhaka. Textile industries in Bangladesh discharged about 217 million m^3 of wastewater in 2016 containing a wide range of pollutants and will reach 349 million m^3 by 2021 if these industries continue using conventional dyeing practices [4]. And the untreated effluent has been discharged into the rivers from nearby textile factories with several major sources of contaminants being outside the city area Gazipur, Tongi, Savar and Asulia [5,6].

Textiles wastewater causes serious water pollution which is very alarming for Bangladesh. The highly colored effluent severely affects photosynthesis of aquatic plants mostly and impacts on aquatic ecosystem due to low light penetration and oxygen consumption [7]. As a result, the normal life cycle of aquatic habitats of the water bodies are constrained (bioaccumulation, bio concentration, bio magnification) or sometimes may extinct which also affects the aquatic ecosystem as well as the total environment. So, it requires proper treatment of wastewater before discharge in environment [8,9].

In this perspective, several techniques have been used in the treatment of textile effluent. Such as, adsorption [10,11], membrane filtration [12,13], electro-coagulation [14], electrolysis [15], biosorption [16], reverse osmosis [17] and chemical oxidation [18]. Coagulation-flocculation process is an example of physico-chemical processes available for the treatment of textile effluent. The coagulation-flocculation (CF) process is used alone or combined with other methods in order to remove suspended solids and organic matter as well as providing high color removal in the textile industry wastewater [2]. Colloid particles are removed from water via coagulation-flocculation process. These test can be performed using a jar-test [19]. In coagulation process, the electrostatic attraction between oppositely charged soluble dye and polymer molecules coagulates the effluents while flocculation allows destabilized particles to flake and be sedimented by gravity [20].

Fenton process is the simplest and cheapest AOP (Advanced Oxidation Process) which consists in the production of hydroxyl radicals by the iron (II) catalyst and Hydrogen peroxide [21, 22,23]. The molecules of textile dyes are structurally different with low or no biodegradability. To remove the color it needs to break off the conjugated unsaturated bonds in molecules [19]. Organic pollutants of textile effluent need to be oxidized for discharge purpose. Here, hydroxyl radical is achieved by the decomposition of hydrogen peroxide catalyzed by ferrous ions in acidic conditions and under mild conditions of temperature and pressure.

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO\bullet + OH^-$$

In this perspective, one of the effective processes is Fenton's reagent which is efficient, cost effective, time saving and easily applicable process [24]. Considering all the these issues the current study was conducted to evaluate and compare the efficiency of coagulation-flocculation and advanced oxidation process (Fenton) using different doses of coagulant and pH for removing BOD, COD, TOC, EC and TDS from the textile effluent.

2. MATERIALS AND METHODS

2.1 Effluent Sample Collection

The raw effluent sample was collected in plastic jars of 5L of capacity from 'Pakiza Dyeing and Printing Industries Ltd.' located at Savarupazilla in Dhaka, Bangladesh. These plastic sample jars were washed first, then rinsed thoroughly with de-ionized water, after that labeled and transported to the laboratory and finally stored in the refrigerator at 4ºC temperature for further treatment. The entire experiment was conducted in the laboratory of the Department of Environmental Science and Water Research Center located at Jahangirnagar University, Savar, Dhaka.

2.2 Chemicals and Reagents

Different chemicals and reagents were used in laboratory analyses as follows-Ferrous sulphate ($FeSO_4$), Calcium hydroxides $Ca(OH)_2$, Polymer, Conc. (30%) Hydrochloric acid (HCl), Conc. (35%) Hydrogen peroxide (H_2O_2) and COD Digestion Reagent. These reagents were purchased from the Hach Company.

2.3 Analytical Methods

The physical parameters such as, Color, Temperature (°C), EC (µS/cm) and TDS (mg/l) were measured using portable photometer (Model: HI96727, HANNA instrument), thermometer, electrical conductivity meter (HI8033) and Total Dissolved Solid Meter (HANNA, HI8734).

Chemical parameters such as, pH was measured using pH Meter (HM-30P, pH Meter), DO (mg/L) was measured using DO Meter (970 DO_2 Meter, Jenway, UK). Besides, TOC (mg/L) was measured with Total Organic Carbon Analyzer (TOC-L CPN E200), BOD (mg/L) was determined by 5-days incubation (20°C) method and COD (mg/L) was determined by Gravimetric method [25] and Titrimetric method [26].

2.4 Jar Test for Coagulation-flocculation

Chemical Coagulation was conducted with the help of jar test. For this test, 4 jars were taken, each of which contained 1 liter of raw sample. Then different doses of $FeSO_4$ and Ca(OH)**2** were added for the chemical coagulation-flocculation treatment as follows:

Dose-1: 1 g of $FeSO_4$ and 1 g of $Ca(OH)_2$
Dose-2: 2 g of $FeSO_4$ and 2 g of $Ca(OH)_2$
Dose-3: 3 g of $FeSO_4$ and 3 g of $Ca(OH)_2$
Dose-4: 4 g of $FeSO_4$ and 4 g of $Ca(OH)_2$

Later, those chemicals were mixed rapidly (in 200 rpm) with the effluent for five minutes (5 mins). Then polymer was added for flocculation and mixed slowly (in 30 rpm) with the effluent for fifteen minutes and kept the mixture for thirty minutes for settling down. After that filtration was done usingsand filter and then filtrated effluent was collected and its physico-chemical parameters (BOD, COD, TOC, EC, TDS, pH, DO) were measured again.

2.5 Fenton Process

Fenton process is considered as one of the advanced oxidation processes. Hydroxyl ($OH^.$) radical is produced in this system as a byproduct of reaction between hydrogen peroxide (H_2O_2) and ferrous sulphate ($FeSO_4$). The hydroxyl ($OH^.$) radical acts as a strong oxidant capable of oxidizing various organic compounds [27,28]. Different pH ranges were considered for Fenton process as follows:

Sample 1: Adjust pH in 4
Sample 2: Adjust pH in 3
Sample 3: Adjust pH in 2
Sample 4: Adjust pH in 1

Samples were taken in four jarsof 500 ml each for four different pH and iron catalyst ($FeSO_4$) of ten milliliters (10 ml of $FeSO_4$ inppm) was added to each sample. Then 3 ml (30% concentrated) of hydrogen peroxide (H_2O_2) was added slowly to each sample again. After that the effluent and chemicals were mixed up for five minutes (5 mins) and kept the mixture for thirty minutes (30 mins) for settling down. Thence filtration was done with sand filter and filtrated effluent was collected and its physico-chemical parameters (BOD, COD, TOC, EC, TDS, pH, DO) were measured again.

3. RESULTS AND DISCUSSION

3.1 Physico-chemical Characterization of Textile Effluent

pH, DO (Dissolve Oxygen), EC (Electro-conductivity), COD (Chemical Oxygen Demand), TDS (Total Dissolve Solid) and TOC (Total Organic Carbon) were measured before and

after treatment. DO value was 1.2 mg/L which was lower than the Department of Environment, Bangladesh standard (2008) [29]. BOD and COD values were measured in the range of 138 to 181 mg/L and 448 to 520 mg/L respectively which were higher than the standards of DoE [29]. Here, physico-chemical parameters of raw effluents are shown in Table 1.

3.2 Changes of Parameters after Coagulation-flocculation Treatment

a) Changes in COD: COD is one of the important parameters to determine wastewater quality. The COD value of raw water of 'Pakiza Dyeing & Printing Textile Industry' was 522 mg/L. After applying various doses (dose-1, 2, 3 & 4) of ferrous sulphate and calcium hydroxide in coagulation process COD value decreased to 70 mg/L (86.59% decrease) at dose-1, 235 mg/L (54.98% decrease) at dose-2, 286 mg/L(45.21% decrease) at dose-3 and 251 mg/L(51.916% decrease) at dose-4 as shown in Fig. 1.

b) Changes in TOC: Total Organic Carbon is an important parameter of industrial effluent the presence of which indicates unhealthy condition for water. Prior to the treatment, the TOC value of textile industry was 12.8 mg/L, which was very high. After the treatment with coagulation-flocculation method, the value of TOC decreased to 0.28 mg/L, 1.43 mg/L, 0.98 mg/L and 1.72 mg/L at dose-1, 2, 3 and 4 respectively as shown in Fig. 2.

c) Changes in pH: The raw effluent collected from textile was alkaline in nature and the pH value was >10.5. After the treatment, the pH value of the treated sample decreased to 6.85 at dose-1, 6.75 at dose-2, 6.55 at dose-3 and 6.30 at dose-4 as shown in Fig. 3.

d) Changes in DO: The DO level of the raw effluent was very low, which was 1.2 mg/L. After coagulation-flocculation treatment, the DO level increased than the initial value (1.2mg/L). When dose-1 was applied, the DO level of the effluent

Table 1. Physico-chemical parameters of raw effluents

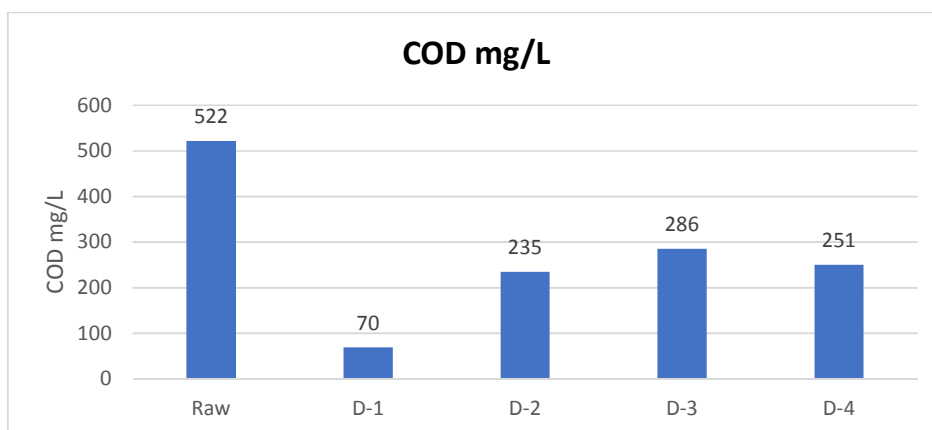| Physico-chemical parameters	Wastewater sample	Effluents quality standard (DoE 2008) [29]
Color	Dark deep green	Transparent
Temperature (ºC)	45	40
EC (µs/cm)	2300	1200
TDS (mg/L)	1260	2100
Turbidity (FTU)	48.28	5
pH	10.5	6-9
DO (mg/L)	1.2	>5
BOD (mg/L)	265	50
COD (mg/L)	522	200
TOC (mg/L)	12.8	…….

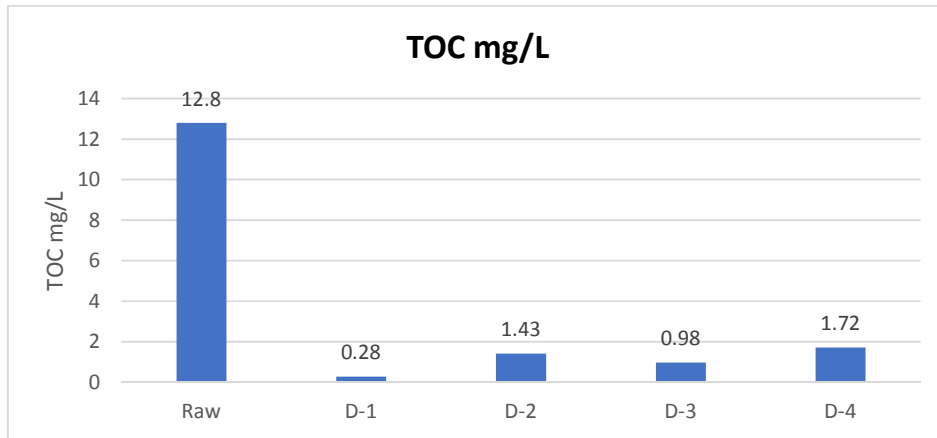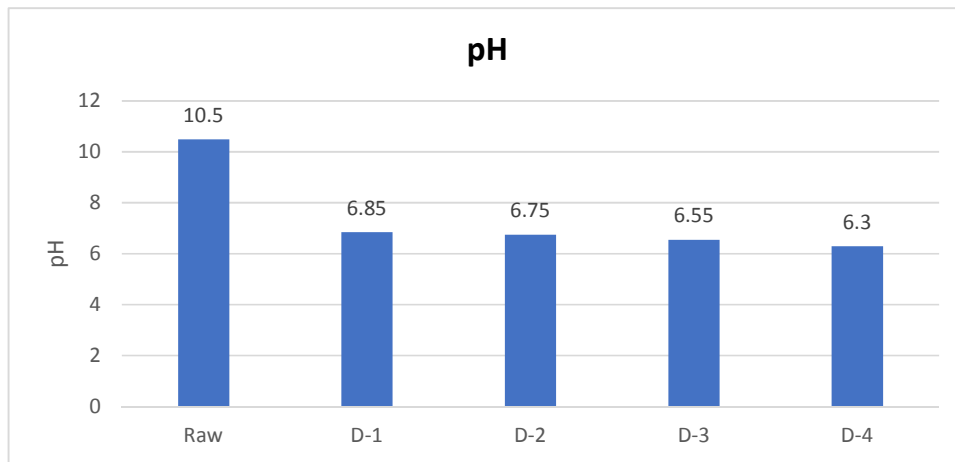**Fig. 1. Changes in COD at different doses**

Fig. 2. Changes in TOC at different doses

Fig. 3. Changes in pH at different doses

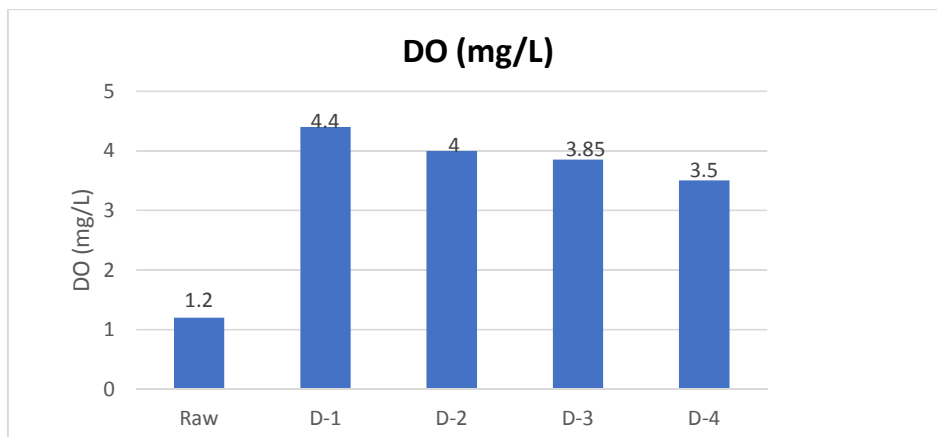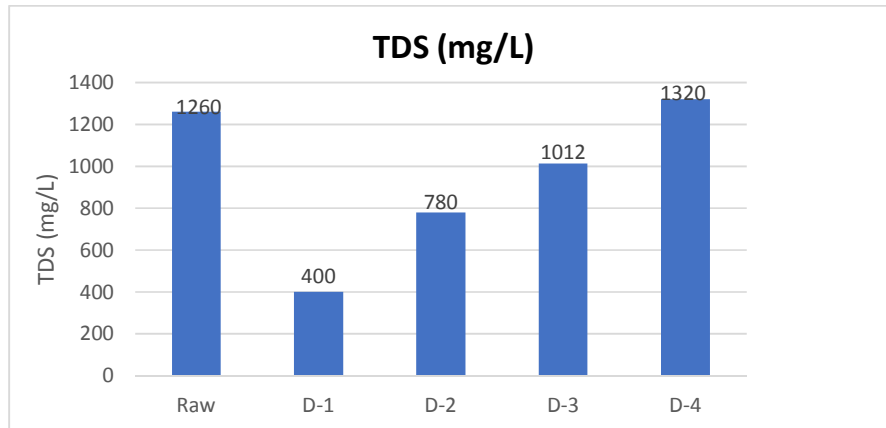**Fig. 4. Changes in DO at different doses**

TDS (mg/L)

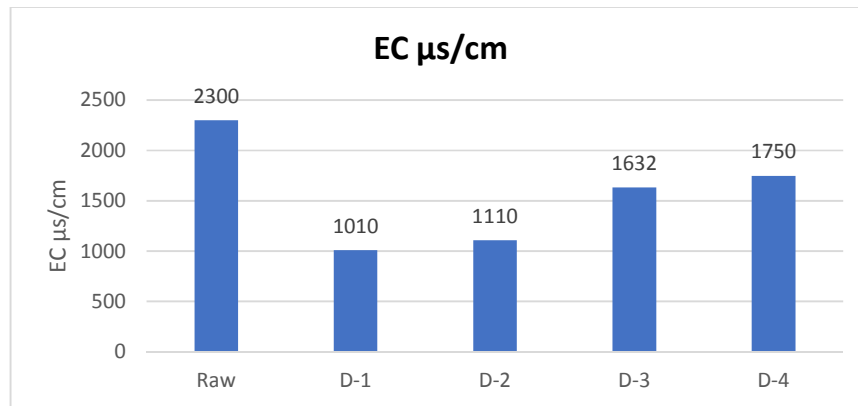**Fig. 5. Changes in TDS at different doses**

EC µs/cm

Fig. 6. Changes in EC at different doses

increased to 4.4 mg/L. After that dose-2, dose-3 and dose-4 were applied gradually which resulted in the changes of DO level to 4 mg/L, 3.85 mg/L and 3.5 mg/L respectively as shown in Fig. 4.

e) Changes in TDS: The Total Dissolved Solids (TDS) of the raw effluent was 1260 mg/L. After coagulation-flocculation treatment, the TDS value changed gradually to 400 mg/L at dose-1, 780 mg/L at dose-2, 1012 mg/L at dose-3 and 1320 mg/L at dose-4 as shown in Fig. 5.

f) Changes in EC: Initial EC was 2300 mg µs/cm. After the treatment, it changed gradually to 1010 µs/cm at dose-1, 1110 µs/cm at dose-2, 1632 µs/cm at dose-3 and 1750 µs/cm at dose-4 as shown in Fig. 6.

3.3 Changes of Parameters after Fenton Treatment

a) Changes in COD: The COD value was 522 mg/L. After the adjustment to various pH values

(pH-1, pH-2, pH-3 & pH-4), the COD value changed to 430 mg/L(17.62% decrease), 188 mg/L(63.98% decrease), 244 mg/L(53.25% decrease) & 240 mg/L(54.02% decrease) at different pH values (pH-1, pH-2, pH-3 & pH-4 respectively) as shown in Fig. 7.

b) Changes in TOC: Initially the TOC level was 12.8 mg/L. After the treatment with Fenton process, TOC level changed to 1.35 mg/L, 2.01 mg/L, 15.6 mg/L & 17.7 mg/L at different pH values (pH-1, pH-2, pH-3 & pH-4 respectively) as shown in Fig. 8.

3.4 Comparison of Coagulation & Fenton Process for COD and TOC Removal

COD and TOC both are important parameters to determine the water quality. In case of COD, the initial value was 522 mg/L which was higher than the standards of DoE [29]. High COD level indicates toxic condition and the presence of biologically resistant organic substances [30]. When dose-1 was applied in coagulation-flocculation treatment and pH-1 was adjusted in

Fenton treatment, a huge difference between the experimental value and the standard value had been noticed.

After that different doses (dose-2, 3 &4) and different pH values (pH- 2, 3 & 4) were experimented in the same manner which resulted in the change of COD value to a significant state near about the standard level of DoE (2008)[29]. In coagulation-flocculation process, for dose-1 COD level was found withmaximum reduction rateas 86.56%where in Fenton process the maximum reduction was noticed as 63.98% at pH-2. Here, Fig. 9 shows the comparison.
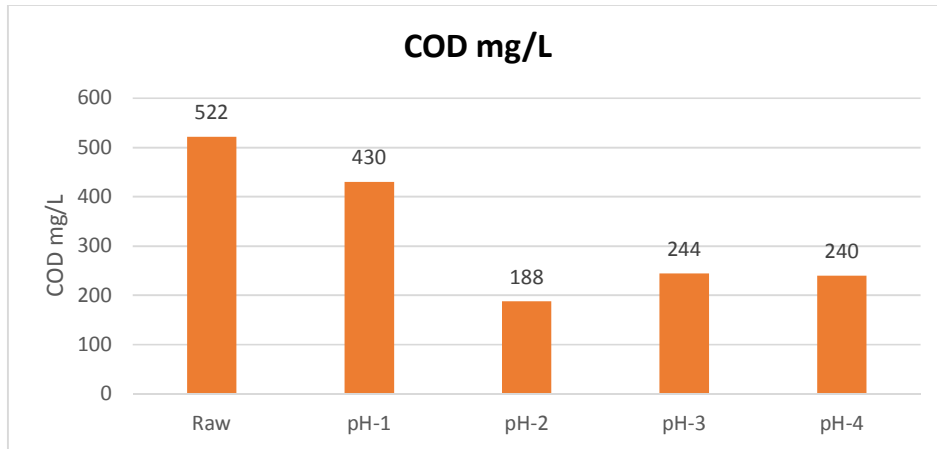

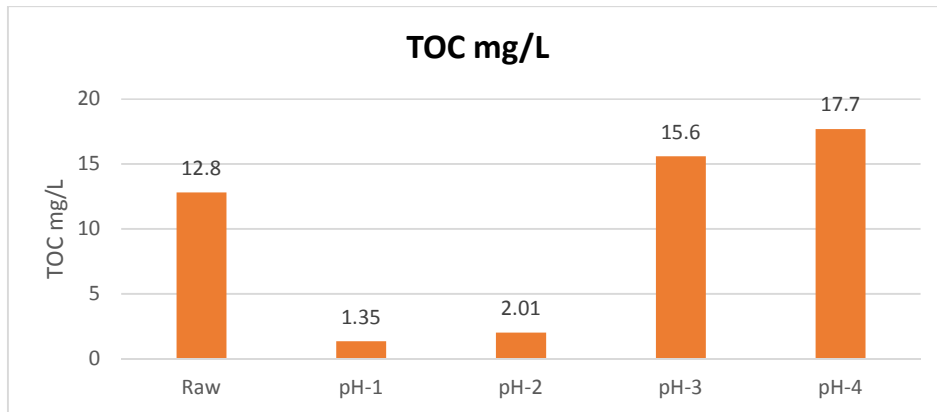**Fig. 7. Changes in COD at different pH**

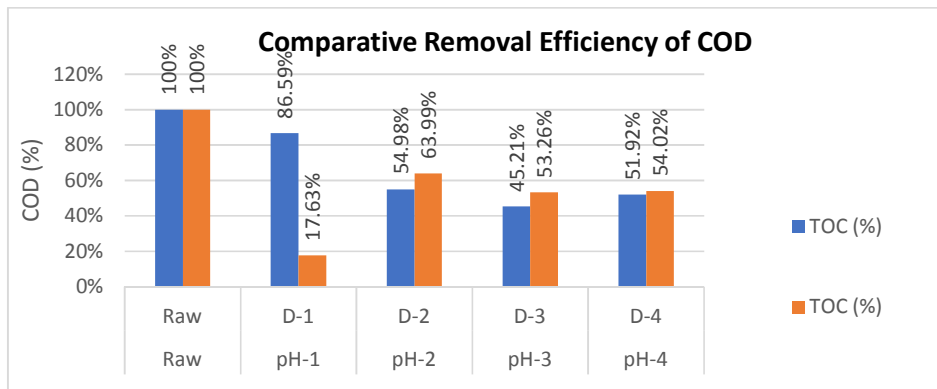**Fig. 8. Changes in TOC at different pH**

Fig. 9. Comparative Removal Efficiency of COD after Coagulation-Flocculationand Fenton process

Total Organic Carbon (TOC) is another parameter, the existence of which in wastewater indicates contaminated state. From Fig. 10 it was observed that 97.8125%, 88.828%, 92.34375% and 86.5625% removal efficiency were found at dose-1, dose-2, dose-3 and dose-4 respectively. While treated with Fenton process, the TOC removal efficiency was found 89.45% and 84.29% at pH-1 and pH-2 respectively. But the value increased more than the initial value (12.8 mg/L) at pH-3 and pH-4.

3.5 Discussion

The measured pH value of wastewater was 10.5 which was alkaline in nature. That value indicated the presence of negatively charged suspended particles (colored colloids and organic matter) which are usually observed in effluent with pH ≥ 4 [31]. This alkalinity has effects on the buffering capacity of the water systems and needs to be monitored in all cases.The pH of the effluent alters the physico-chemical properties of water which in turn adversely affects aquatic life, plants, and humans. After the coagulation-flocculation process, a significant reduction was observed in pH value (6.30-6.85) at different doses. That pH range followed the standard pH value set by the DoE [29] and the EPA [32].

The most significant measure of water quality is the dissolved oxygen (DO) [33]. The lower DO content could be due to intrusion of high organic load in the water which leads to oxygen depletion [34]. High value of Total Dissolved Solids reduces the light penetration into water and ultimately decreases the photosynthesis. The decrease in photosynthetic rate reduces the DO

level of wastewater which results in decreased purification of wastewater by microorganisms [35]. Initial DO value was very low (1.2 mg/L). While treated by coagulation-flocculation process, the level upgraded to (3.5-4.4) mg/L at different doses. It was observed that the level was near about the standard value >5 set by the DoE [29].

High EC indicates a large amount of ionic substances like sodium, potassium, iron etc. in textile effluent [36]. Initially EC was measured as 2300 µs/cm. After coagulation-flocculation treatment, a substantial decrease had been observed at dose-1 and dose-2 as 1010 µs/cm and 1110 µs/cm respectively. Before the treatment, the TDS value of the effluent was 1260 mg/L which had exceeded the guideline limit [29]. High TSS and TDS detected can be recognized to high color and they may be major sources of the heavy metals. Increased heavy metals concentration in river sedimentscan grow suspended solid concentration [37]. During the dry season, the occasional dust re-suspension leads these metals into the atmosphere along with the particulates which canform health problems in the form of air pollution. Moreover, high concentration of TDS reduces water clarity and cloudy water absorbs more heat andblocks light penetrations. Therefore, intensified turbidity rises water temperature and inhibits photosynthesis [7]. This study found the value of TDS (after coagulation-flocculation treatment) satisfactory at dose-1 and dose-2 which was 400 and 780 mg/L respectively. But the level of TDS and EC increases after using high dose of coagulant. So, Dose 1 was effective for reducing EC and TDS of wastewater.
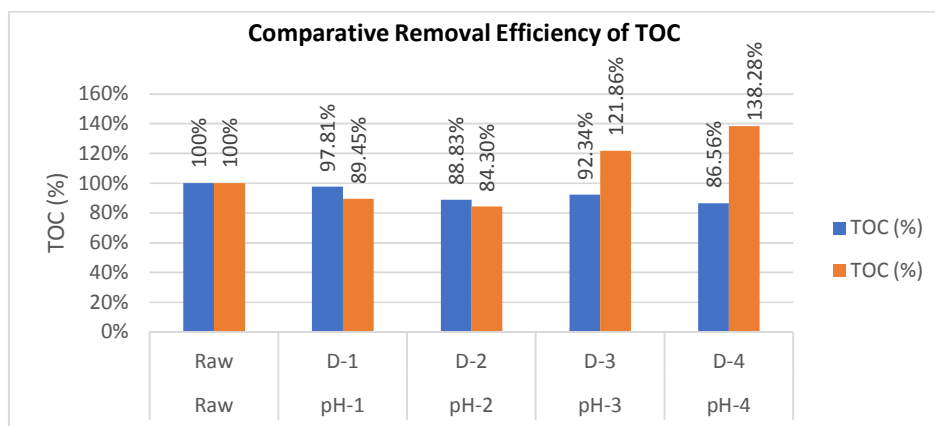

Fig. 10. Comparative removal efficiency of TOC after coagulation-flocculation & Fenton process

High COD level implies toxic state and the existence of biologically resilient organic substances [38]. COD value in sample effluent was 522 mg/L. Coagulation and flocculation test showed that COD removal was effective at all doses. Where, dose-1 reduced the maximum COD as 86.56%while dose-2, 3 and 4 reduced 54.98%, 45.21% and 51.916% respectively. So, it is better to avoid using high dose of coagulant. Abdel-Fatah et al. [39] also found 93.3% COD removal efficiency after using chemical coagulant. In Fenton test, COD removal was agreeable at pH-2, pH-3 & pH-4 where pH-2 reduced the maximum COD as 63.98%. But at pH-1 the removal rate of COD was found very poor as 17.62%. Amruta et al. [28] found 85% removal efficiency of COD using Fenton process.

Total Organic Carbon (TOC) is an important parameter of industrial effluent, the presence of which indicates unhealthy condition for water [40]. The TOC removal efficiency in coagulation-flocculation treatment was noticed noteworthy as97.81%, 88.82%, 92.34% and 86.56% at dose-1, dose-2, dose-3 and dose-4 respectively. Among these changes, dose-1 reduced the maximum TOC while in Fenton process, pH-1 and pH-2 showed the maximum reduction of TOC as 89.45% and 84.29% respectively. But at pH-3 and pH-4 the reduction rate was very unusual. However, in both treatment processes TOC removal efficiency was remarkable at all.

4. CONCLUSION

Textile industries are one of the most significant economic sectors in Bangladesh. But the impacts regarding this sector are causing alarming issues for environment day by day. The pollutants generated from this sectorhave already appeared as a big concern not only for health of humans/animals but also for aquatic and terrestrial ecosystems. This work analyzed the removal efficiency of EC, TDS, pH, DO, COD and TOC of textile effluent by applying coagulation-flocculation & Fenton treatment processes and make a comparison between TOC & COD removal efficiency. After coagulation-flocculation and Fenton processes, the values of the parameters (DO, TOC, COD, TDS, EC, and pH) were positively changed. Maximum removal efficiency of COD (86.56%) and TOC (97.81%) were found for using low dose of coagulant. And other parameters were also reduced after using low dose of chemical coagulant. So, it could reduce the cost of treatment process. In Fenton treatment process,

maximum TOC removal efficiency (97.81%) was examined at pH-1. From the study, it is observed that coagulation-flocculation and Fenton treatment processes are more cost effective and easier than other treatment methods which will lead to achieve the concept of "zero discharge" from the industrial production practices.

DISCLAIMER

ACKNOWLEDGEMENT

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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