Original Article

Comparing Fenton Oxidation with Conventional Coagulation Process for RR198 Dye Removal from Aqueous Solutions

Behnaz Esrafili[1] Reza Jalilzadeh Yengejeh[2]*

1. M.Sc., Department of Environmental Engineering, Science and Research Branch, Islamic Azad University, Tehran, Iran
2. Assistant Professor, Department of Environmental Engineering, Ahvaz Branch, Islamic Azad University, Ahvaz, Iran

***Correspondence to**: Reza Jalilzadeh Yengejeh
R.jalilzadeh@iauahvaz.ac.ir

(Received: 1 Mar. 2017; Revised: 5 Jun. 2017; Accepted: 15 Oct. 2017)

Abstract

Background and purpose: The wastewater discharged from textile industries is one of the main environmental pollutants that causes many problems for the environment if it is not treated or discharged. The present study compared Fenton oxidation process with coagulation and flocculation using the natural coagulant of Sodium Alginate in order to remove Reactive Red 198 Dye.

Materials and methods: This study was carried out in an experimental scale, in which the effects of pH, concentration of the dyeing substance, concentration of iron sulfate and hydrogen peroxide for the oxidation of Fenton and the effects of pH, concentration of coagulant, concentration of dyeing substance, and the Coagulant Aid of Sodium Alginate were all investigated.

***Results*:** The results of the current study indicated that in the Fenton process, the efficiency of RR 198 Dye removal under acidic conditions (pH=3) at optimal conditions was achieved to be 96.2% for the dye with the concentration of 20 mg/l. By using 70 mg/l of the coagulant of Poly-Aluminum Chloride in coagulation and flocculation process, 71.2% of the dye removal was obtained for the initial concentration of the dye as 20 mg/l. Also, by adding 50 mg/l of Sodium Alginate to the optimal concentration of Poly-Aluminum Chloride, the dye removal increased up to 92.1%.

***Conclusion*:** Although under optimal conditions, the efficiency of coagulation process with coagulant aid was only 4% less than the efficiency of Fenton process, considering the advantages of Fenton oxidation including lack of production of excessive sludge, a higher efficiency was gained at large doses of dye.

Key words: RR198 Dye; Fenton; Coagulation; Poly-Aluminum Chloride; Sodium Alginate

Citation: Esrafili B, Jalilzadeh Yengejeh R*. Comparing Fenton Oxidation with Conventional Coagulation Process for RR198 Dye Removal from Aqueous Solutions .2017; 5 (4):26-37

1. Introduction

The wastewater produced by textile industries is highly contaminant in terms of quality and has a huge volume in terms of quantity. It usually has a large amount of organic dyeing compounds (1, 2). Azo dyes contain one or several Azo bonds (-N=N-), and are one of the most important synthetic dyeing groups. Some of the Azo dyes and their impurities like amines are toxic and even carcinogenic for some living creatures (3). Coagulation and flocculation are important chemical processes of treatment for destabilizing colloid particles and Dye removal (4, 5). Another similar technique is advanced oxidation, currently used for treatment of undegradable organic compounds, such as dyeing materials, and organic chemicals, as well as toxic compounds. It is able to oxide organic compounds into carbon dioxide, water, and organic acid. Advanced Oxidation Processes include Fenton Process, Photocatalystic, Electrochemical, and Ultrasonic Methods (6-8). The methods of advanced oxidation are based on production of hydroxyl radical (HO`) in the medium. These radicals have the highest efficiency in the oxidation of organic compounds thanks to high potential of oxidation-reduction of the radicals and the non-selectivity of the compounds oxidized by them. During a catalysis process, H_2O_2 is converted to hydroxyl radical, possessing the highest oxidizing power after fluorine (9, 10). Fenton process is one of the most common and practical processes among the advanced oxidation processes producing hydroxyl radicals through the reaction of Ferro ion with hydrogen peroxide under acid conditions. Hydroxyl radical attacks organic compounds causing decomposition of organic compounds (11, 12). Mansoorian observed high efficiency of the Fenton oxidation process and over 95% removal of textile dye (13). Similarly, El Haddad et al. (2014) reported acidic pH as suitable for effective removal of Reactive Dye in the Fenton process (14). Coagulation tests were carried out in two steps. In the first step, poly aluminum chloride alone was used as coagulant, and in the second step, sodium alginate was used as a coagulant aid together with coagulant in order to enhance the efficiency of the process for the removal of RR198 dye. The results of these two processes were compared with the results of Fenton oxidation tests, and the efficiency of both methods was examined. This study aimed to investigate and figure out a treatment method to remove Reactive Red 198 Dye which is very common. Advanced oxidation process as well as Fenton coagulation and flocculation were applied in this study to remove Reactive Red 198 Dye.

2. Materials and Methods

In this study, in order to compare the performance of Fenton and coagulation in removal of the dyeing material of Reactive 198, the effect of influential parameters as independent variables was investigated on dye removal followed by a presentation of a report on the efficiency of the contaminants removal as the process efficiency. The procedure of conductance of experiments has been based on a standard method (15).

An amount of one gram of Reactive 198 Dye purchased from Alvan Sabet Co. (Table 1) was brought to the volume of one liter with distilled water. For homogenization, it was placed on a stirrer with a slow rotation.

Table1. Chemical Compounds used in this research

Chemical Compound	Country / Company
Poly-Aluminum Chloride	Germany/ Merck
Alum	Germany/ Merck
Sodium Alginate	USA/ Sigma Aldrich
H_2O_2	Germany/ Merck
Deionized water	Germany/ Merck
Reactive Red 198 Dye	Iran/ Alvan Sabet

This solution was used as the stock solution for the preparation of samples with different concentrations. For adjustment of pH, Sulphuric Acid and Sodium Hydroxide 0.1 M were utilized. Figure 1 demonstrates the structure of the applied dye and Figure 2 represents the structure of the coagulant aid substance of sodium alginate (2, 16).

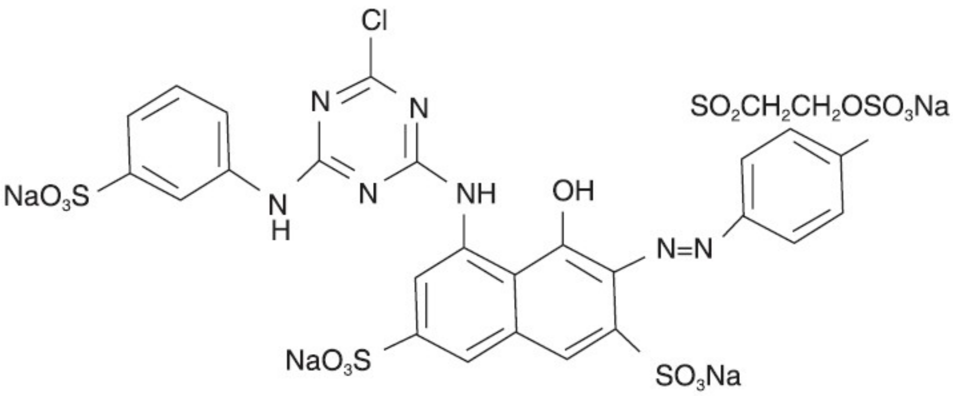

Figure 1. Chemical structure of the dyeing substance of Red Reactive 198(2)

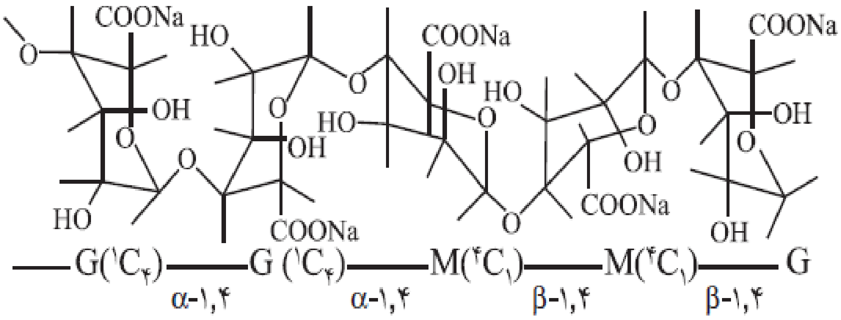

Figure 2. Molecular structure of the sodium alginate (2, 3, 29).

In the Fenton oxidation process and coagulation, the best performance of each step is considered to be constant in the next stage. In Fenton oxidation, first the effect of pH (3, 5, 7, and 9) and then the influence of concentrations of hydrogen peroxide at concentrations of 2, 4, 6, and 10 mmol at the optimal pH of the previous stage were experimented. The effect of the concentration of Iron Sulfate was also investigated between the values of (10, 15, 25, and 50 mg/L) followed by the evaluation of the effects of the concentration of the primary substance of the dyeing material with respect to the concentrations of the conducted studies at four concentrations (10, 20, 30, and 50 mg/L). It is worth mentioning that the desired concentrations were selected according to other studies in relation to the removal of the widely used dye and its relative amount in aqueous resources.

In the coagulation and flocculation experiments, first the pH of 4, 5, 6, 7, 8, and 9 were experimented. After determining the optimal pH, the effect of the concentration of the coagulant of Poly-Aluminum Chloride at values of 50, 70, 90, 110, 130, and 150 was investigated. The effect of Sodium Alginate was also examined at three concentrations (30, 50, and 80 mg/L) of the optimal pH along with different concentrations of the coagulant. Next, the effect of the concentration of the primary substance of the dyeing material was explored considering the concentrations of the conducted studies at concentrations of 5, 10, 15, 20, 30, and 50 mg/L. Coagulation and flocculation process was fulfilled using jar test apparatus with stirring of 100 rpm for one minute at first, and 30 rpm for 20 minutes afterward. Next, after 30 minutes of rest or sedimentation, and passing the samples through the filter, the rate of dye absorption and reduction was read via spectrophotometer. Moreover, in Fenton tests after making the sample and adding the desired amounts of ferrous sulphate and hydrogen peroxide, the samples were immediately tested in shaker (Ika KS260 basic) at different time periods (Table2).

Table2. Equipment used in this research

Instrument	Model
Jar Test	wisd laboratory instruments
Spectrophotometer	T80 UV/VIS spectrometer
Shaker	Ika KS 260 basic
pH meter	Crison

The Dye concentration was measured using a Spectrophotometer (T80, UV/VIS) of the wavelength of 518 nm at the wavelength at which the maximum absorption of this substance occurs.

3. *Results*

In the current study, the appropriate pH level for Fenton reaction was found to be about 3. Usually, in pHs about 3, iron deposits as Fe (OH)3, and causes the decomposition of hydrogen peroxide as oxygen and water. In addition, the formation of bivalent iron complexes at higher pHs reduces its concentration in the environment (Figure 3).

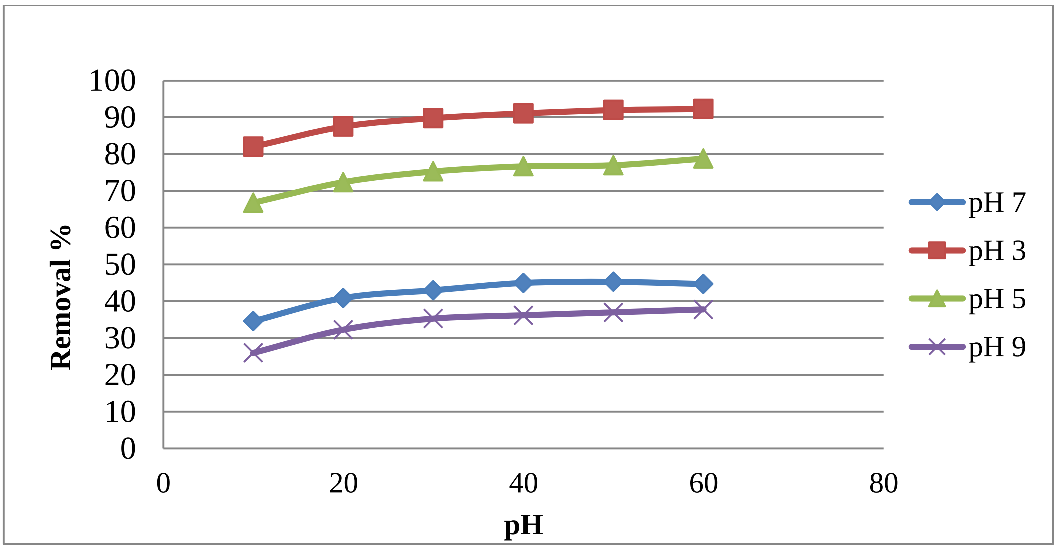

Figure 3. Effect of pH (Dye con: 20 mg/L, H_2O_2: 2 mmol , Fe: 15 mg/L)

According to Figure 4, the optimal concentration of hydrogen peroxide was obtained to be 4 mmol. When the concentration increased from 2 to 4 mmol, there was also an increase in dye removal, whereas at the concentration of 6 mmol, it not only did not increase, but also diminished to some extent. In a similar vein, with further elevation of concentration until 10 mmol, a further declination was documented in dye removal. When the time prolonged from 10 to 20 min, dye removal increased by 8%, but thereafter the elevation was very slow.

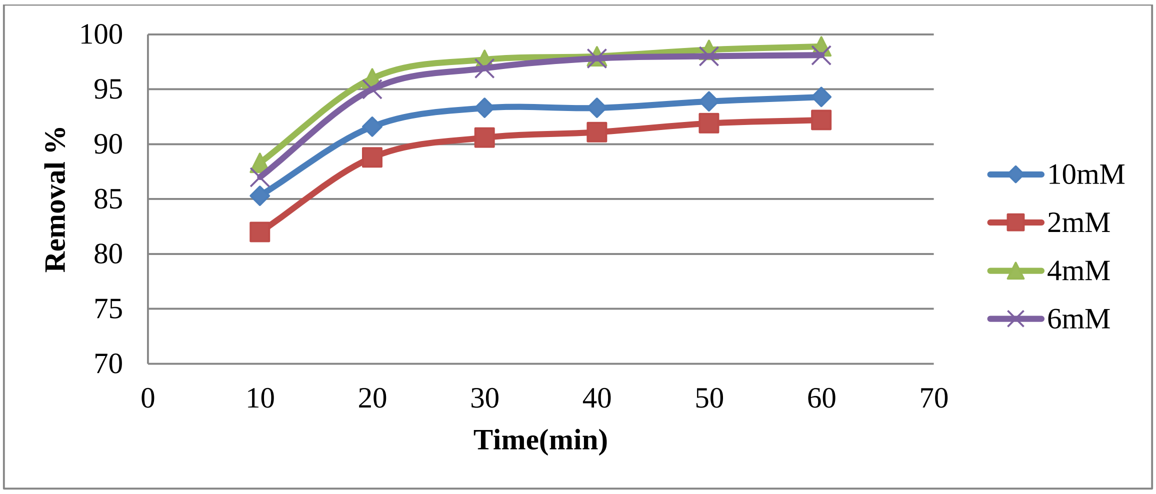

Figure 4. Effect of pH (Dye con: 20 mg/L, H_2O_2: 2 mmol , Fe: 15 mg/L)

According to Figure 5, the impact of iron sulfate at different concentrations (10, 15, 25, and 50 mg/L) on the process indicated that as the concentration increased from 10 to 15 mg/L, dye removal was seen to grow by 6.2% within 20 min. However, with further elevation of concentration up until 25 mg/L, the development was observed to be only 1% within the optimal time (20 min). Similarly, at the concentration of 50 mg/L of iron sulfate, the trend of dye removal decreased significantly, based on which the concentration of 15 mg/L was chosen as the optimal concentration.

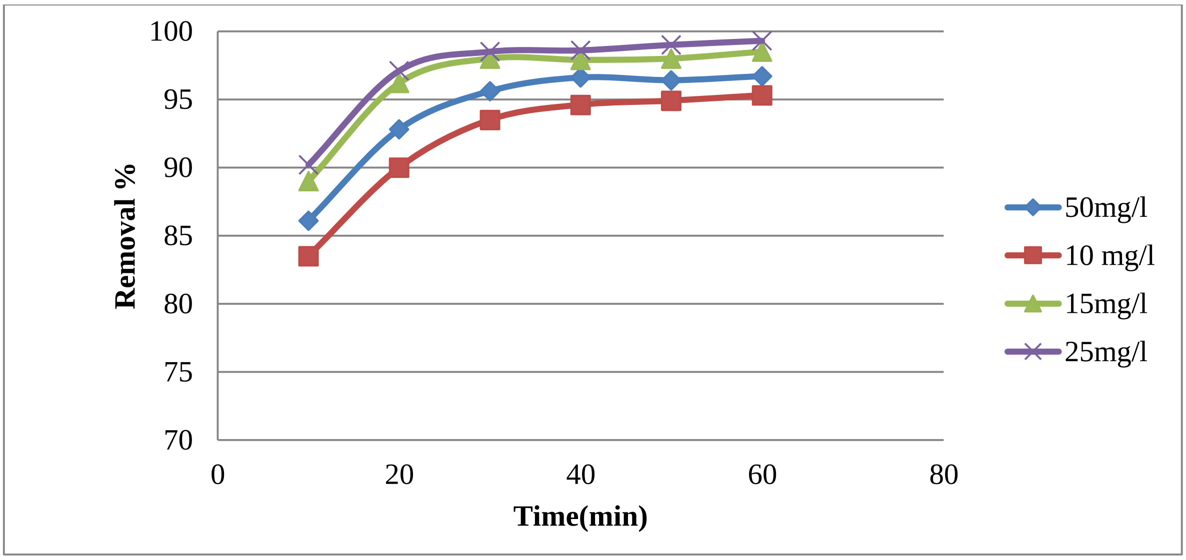

Figure 5. Effect of Fe (Dye con: 20 mg/L, H_2O_2: 4 mmol, pH=3)

The reduction is due to the absence of radicals to remove the dye in solution. In other words, higher amount of organic matter has less opportunity for degradation when faced with the produced hydroxyl radicals. Figure 6 demonstrates that as the dye concentration increased, the degree of dye removal diminished. This lowered removal was very trivial between the concentrations of 10 and 20 mg/L. Figure 6 demonstrates that as the dye concentration increased, the degree of dye removal diminished. This lowered removal was very trivial between the concentrations of 10 and 20 mg/L, but when the concentration increased up to 50 mg/L, the decline in removal became more dramatic. At all four concentrations, the optimal time for removal was found to be 20 min.

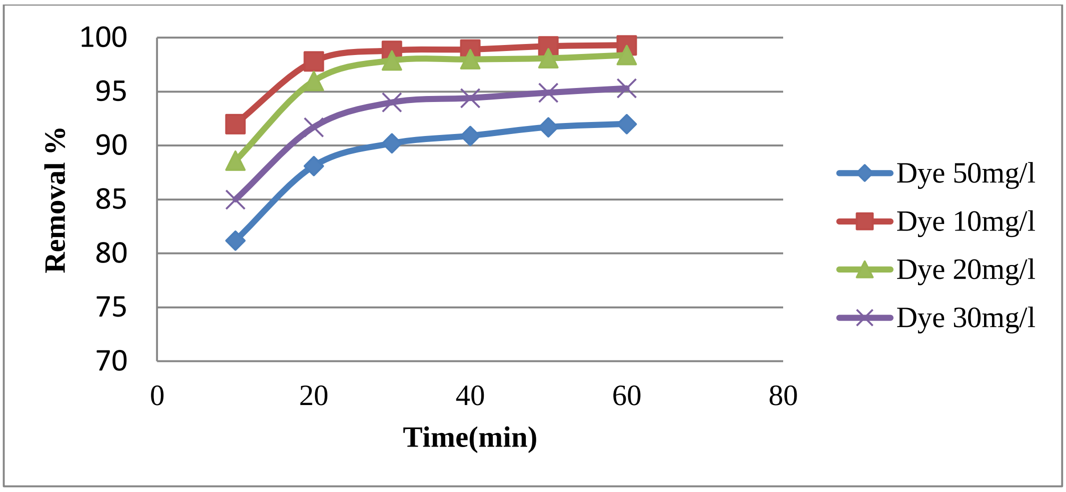

Figure 6. Effect of Dye con (H_2O_2: 4 mmol , pH=3, Fe: 15 mg/L)

In order to obtain the optimal pH, experiments were conducted at different pHs for both coagulators. Both diagrams had an ascending trend in the beginning. As is shown in Figure 7, the coagulator of the poly-aluminum chloride had the highest level of removal at pH=8, though further elevation of pH led to lowered removal.

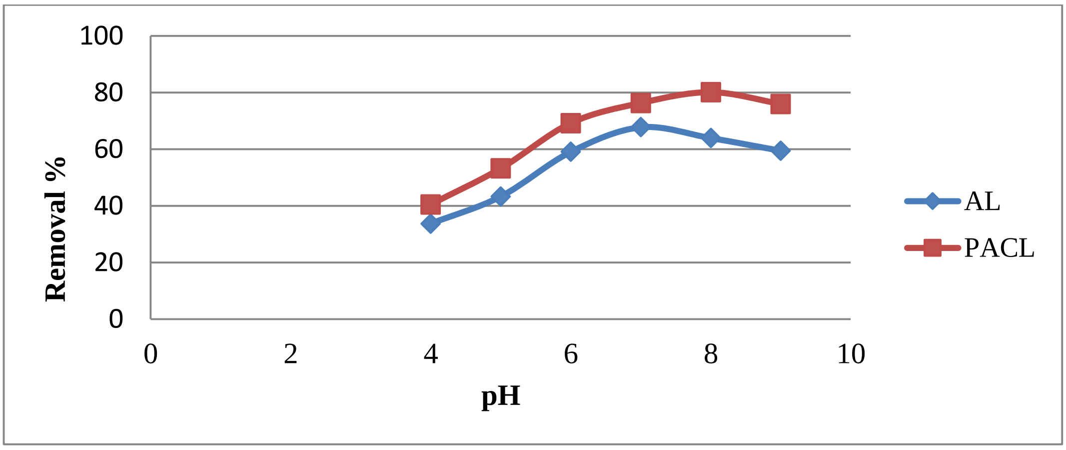

Figure 7. Effect of pH (Dye con: 20 mg/L, coagulant: 95 mg/L)

Figure 8 reveals the relationship between different concentrations of the coagulator of poly-aluminum chloride with dye removal levels. As the concentration of the coagulator increased from 50 to 70 mg/L for both concentrations of 20 and 50 mg/L, the efficiency grew by 27.7 and 25.4%, respectively. However, thereafter the trend of elevation became slower.

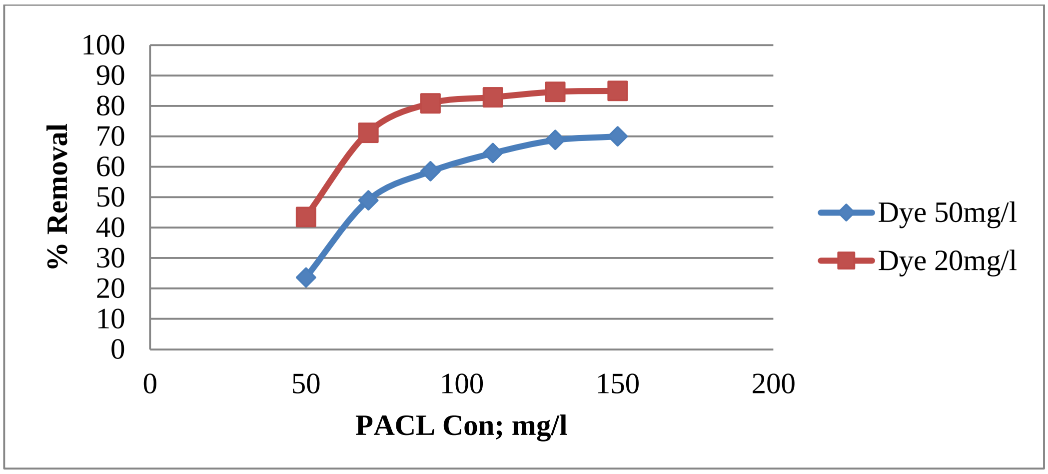

Figure 8. The effect of PACL ((Dye con: 20-50 mg/L, pH=8).

According to Figure 9, it is observed that at low concentrations of the dyeing material (concentrations of 5-20 mg/L), the changes are minor. However, when the concentration of the dyeing substance increased from 20 to 50 mg/L, then a dramatic decline was observed in the degree of removal, where at the optimal concentration of 70 mg/L for the concentration of the dyeing material with a concentration of 20 and 50 mg/L, the level of removal was achieved to be 71.3% and 47.8%, respectively.

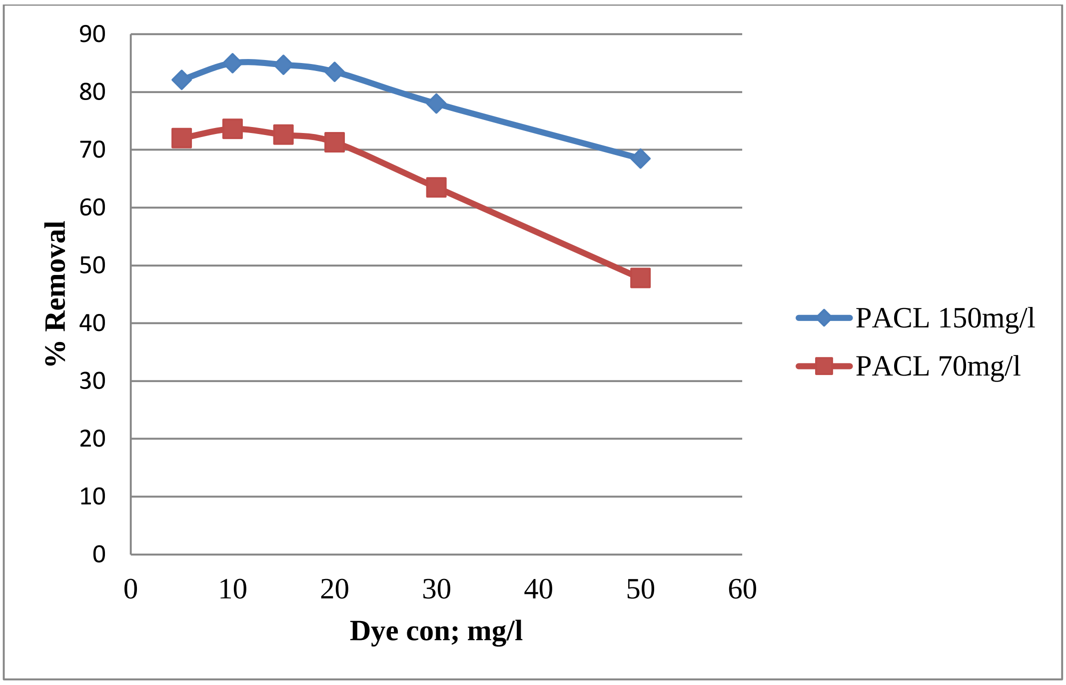

Figure 9. The effect of dye concentration with PACL (con: 70 and 150 mg/L, pH=8).

Figure 10 indicates the effect of the Coagulant Aid of sodium alginate on dye removal. As can be observed, this coagulator has been added at three concentrations of 30, 50, and 80 mg/L to different doses of the coagulator of poly-aluminum chloride. In this experiment, the concentration of 50 mg/L has been obtained as the optimal concentration in this diagram.

Figure 11 demonstrates the comparison of the processes in this experiment. As can be seen, the Fenton process has the highest efficiency equal to 96.2% at optimal conditions (iron sulfate 15 mg/L, and hydrogen peroxide 4 mmol within 20 min).

The process of coagulation and flocculation in which a Coagulant Aid had also been used, had a higher removal percentage in comparison with the process without sub-coagulator, where in coagulation with poly-aluminum chloride with a concentration of 70 mg/L, the level of removal was 71.2%. Through the addition of 50 mg/L Coagulant Aid of sodium alginate, the removal percentage has reached by 92.1% showing a 21% increase.

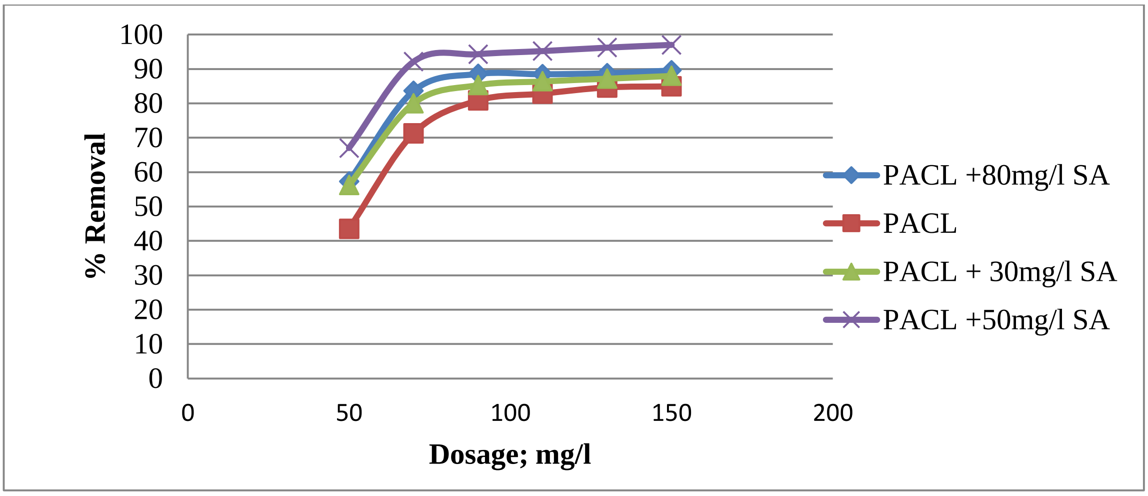

Figure 10. The effect of sodium alginate (dye con: 20 g/L, pH=8 and PACL)

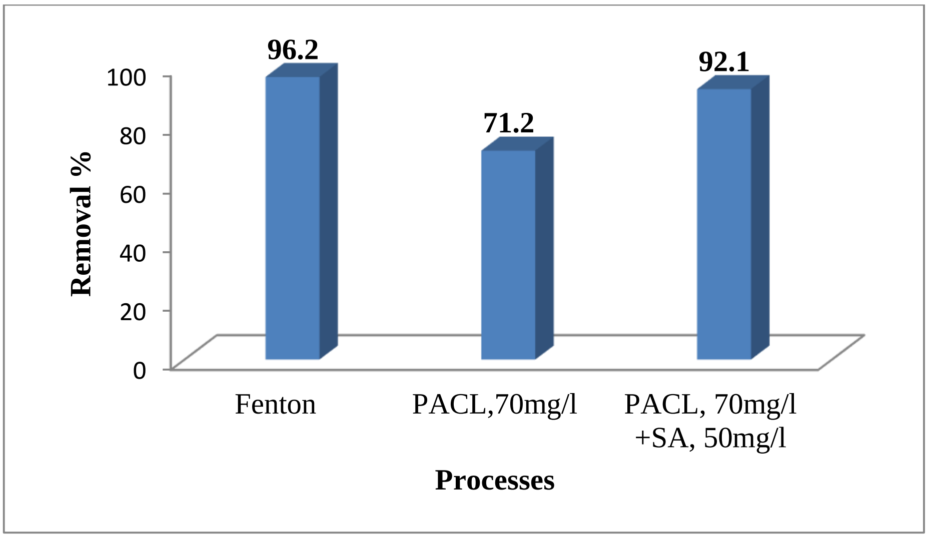

Figure 11. Comparison of two processes at optimal condition

4. Discussion

In this study, the appropriate pH for Fenton reactions was about 3. Usually, in pHs above 3, iron deposits as $Fe(OH)_3$, and caused the degradation of hydrogen peroxide into oxygen and water. Moreover, the formation of bivalent iron complexes at higher pHs resulted in the reduction of its concentration in the environment. In contrast, the reproduction of iron (II) through a reaction between Fe^{3+} and H_2O_2 in more acidic pHs was supposed to be prevented (EG1-2)(17). As the concentration of hydrogen peroxide increased from 2 to 4 mM, a significant increase was observed in decolorization. The increase of efficiency was due to greater amount of hydrogen peroxide in the system and subsequently the increased production of hydroxyl radicals, which finally caused the degradation of the dye (17). As the concentration increased to 10 mm, the efficiency reduced dramatically. As stated in other studies, the decrease of efficiency can be attributed to the excessive concentration of hydrogen peroxide and spontaneous decomposition of this oxidizing agent (18, 19). With the increase of iron sulfate from 10 to 15 mM, favorite decolonization was observed, because any increase in the level of iron concentration resulted in the production of more hydroxyl radical, and consequently the speed and rate of bleaching increased. Hodaifar et al. achieved the same results in their research (20). As the iron (II) ions exceeded over a certain limit, the rate of produced hydroxyl radicals increased greatly, too. The reaction between the produced hydroxyl radicals and H_2O_2 in solution, then, caused the fixation or even the reduction of decolorization rate. Many studies, such as those conducted by Hameed and Karthikeyan indicated that the excessive increase of Fe (II) concentration had an inhibitory effect on the rate of decolorization (21, 22). In this study, the rate of decolorization reduced about 10% as the concentration ranged from 10 to 50 mg/l. The reduction is due to the absence of radicals to remove the dye in solution. In other words, the higher amount of organic matter has resulted in the provision of less opportunity for degradation when faced with the produced hydroxyl radicals. As mentioned in a research conducted by Karthikeyan et al., in most advanced oxidation processes, a reference is made to the decrease of removal because of the increase of dye due to the fixation of hydrogen peroxide and consequently the unchanged amount of OH radicals (22). According to the researchers, pH is a significant factor in the coagulation and flocculation process. Each coagulant has also an optimum pH in which the coagulation and flocculation process occur for a certain concentration of coagulant in the shortest possible time and with the highest efficiency which, of course, varies according to the type of dye (23, 24). As can be seen in this study, the highest efficiency belonged to Poly Aluminum Chloride at pH=8. In a similar study, it was found that Poly Aluminum Chloride was effective in the removal of dye in a wide range of pH (pH= 7-10) (25). According to diagram (6), it seems that the concentration of 70 mg/l with the removal of 71.2% was documented to be a particular point or, in other words, was a critical point in the graph for the dye with concentration of 20 mg/l, because the slope of dye removal in this point has reached its maximum value. Therefore, the optimal concentration of Poly Aluminum Chloride was considered to be 70 mg/l to remove the dye. In their study, Bali et al. demonstrated that the effectiveness of decolorization at the concentration of 20 to 100 mg/l increased as the amount of coagulant increased, and then no change was observed (26). As it was observed, through an increase in the organic matter as the primary concentration of the dye, the rate of decolorization decreased. Zahrim (27) carried out a research via increasing the initial concentration of dye in coagulation and flocculation process with poly aluminum chloride as coagulant, and found that the removal of the dye decreased in the process (27). The rate of reduction varied according to the type of consumed dye and the kind of coagulant. In this study, through the increase of initial concentration of dye, a higher decrease was observed in the efficiency of coagulation process compared with Fenton process. Hence, in this study, it was found that an increase of coagulant concentration from 30 to 50 mg/l led to an increase in the dye removal, and as the dose increased to 80 mg/l, the dye removal decreased. In the research conducted by Caihone, the combination of alum and sodium alginate rather than alum alone had a more positive effect on the dye removal (2). The use of a polymer in combination with a conventional coagulant could enhance the rate of dye removal. Organic polymer compounds could also be useful at the coagulation because of their ability to produce large and compact flukes with settling characteristics (25, 27).

5. Conclusion

The present study aimed at removing the dyeing substance of reactive red 198, and to this end, it applied two methods of Fenton advanced oxidation plus coagulation and flocculation. The Fenton advanced oxidation process was found to be an effective method for removing dye, and based on the results, as compared with the application of poly-aluminum chloride coagulant, it had a higher efficiency in dye removal in itself. The addition of optimal dose of the Coagulant Aid of sodium alginate to the optimal concentration of poly-aluminum chloride resulted in 21% of dye removal, suggesting the effectiveness and efficiency of this natural coagulant on removal of reactive red 198. According to the obtained results, at the dye concentration of 20 mg/L, the application of poly-aluminum coagulant together with sodium alginate at optimal conditions became almost equal to the removal percentage of Fenton oxidation being only 4% lower. However, in this study, Fenton oxidation was more desirable thanks to its advantages including no production of huge amounts of sludge and no problem regarding the cost of discharging sludge. According to the results of this study, further investigation should be performed regarding the variables, such as temperature, the stirrer rate, and the application of these two methods on other textile dyes. Additionally, in order to increase the efficiency, further research could be conducted through comparing the Fenton oxidation with other advanced oxidation methods and using combinational processes for the elevation of the efficiency of the process, such as photo-electro-Fenton and so on.

Acknowledgement

This Article derived from M.Sc Degree Thesis of Ms. Behnaz Esrafili. It was performed under the supervision of Dr. Reza Jalilzadeh (Ph.D) at the Department of Environmental Engineering, College of Environment and Energy, Science and

Research Branch, Islamic Azad University, Tehran, Iran.

Conflict of interest

The authors declare no conflict of interest in this research.

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