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ORIGINAL ARTICLE

Removal efficiency of (AO7) Acid Orange 7 color using the advanced oxidation of modified UV/H2O2 with zero-valent iron

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ABSTRACT
Dyes are mainly organic materials that have one or more benzene rings. They are highly toxic to aquatic environments and do not decompose easily in the environment. Thus, it is essential for them to be refined with effective and inexpensive methods before evacuation to the environment. Nowadays, many researchers have used zero-valent metals such as iron to refine organic pollutants so that using iron powder to remove the environment pollutants are up to date researches today. In this study, Zero valence iron powder is used in the presence of hydrogen peroxide and UV radiation to decompose the Acid Orange7 color. The research parameters in this study are pH, the initial concentration of the color solution, the initial concentration of iron powder, the initial concentration of hydrogen peroxide, and the contact time. The results of the tests showed that by increasing the contact time, the initial concentration of iron powder and the initial concentration of hydrogen peroxide, the removal efficiency increases while color removal efficiency increases by reducing the pH and initial concentration of color. It was determined that the highest removal efficiency occurs on pH=3, the contact time of 120 minutes, 3 ml in 150 ml hydrogen peroxide and 0.3g in 150ml iron powder in color solution. Also, the results showed that the process efficiency of zero-valent iron powder is acceptable in the presence of UV light and hydrogen peroxide. According to the results of this research, this process can be used more effectively in refining colored wastewater.
Keywords: *Textile wastewater - chemical oxidation - zero valent iron - AO7*

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INTRODUCTION

Textile industry is one of the most important industries of each country and it is one of the greatest consumers of water resources and in the following wastewater producer. Textile industry wastewater is always considered by environmental experts to reduce the pollution of water resources often due to solids and fibers, dyes, detergents, starch and a variety of different types of mineral salts and heavy metals, nitrogen and phosphorus in bands dyes, thermal pollution, drastic changes in pH, fat, and oil. Dyes in textile wastewater are mainly parts of aromatic compounds. These compounds have two or more benzene rings and they are considered as resistant compounds in the environment due to having multiple connections. Dyes are mainly organic compounds which are produced naturally or artificially and they are toxicity for some aquatic and they have adverse effects on the environment, in addition to creating ugly scenes in the environment. Thus, removing these substances prior to evacuation to the environment is essential. One of the dyes that sometimes made problems for the environment is a type of azo color called Acid Orange 7. In a study, anaerobic refining is used to remove Acid Orange 7 color. The study has shown that color removal is possible only when the active biomass is used with a nutritional assistance [1]. One of the ways that researchers recently have focused on is using advanced oxidation processes, which have provided acceptable results in recent years. In this process, free active radicals such as hydroxyl active ($^{\bullet}OH$) is produced which has a strong oxidation power to destroy organic pollutants[1].

[1] $E_{OH\bullet}$= 2.80 eV

The advantage of the advanced oxidation processes is that they ultimately destroy the organic materials to CO2 and H2O, i.e. mineral decomposition. Another advantage of this process is that it can destroy the low concentrations of pollutants [2]. In order to increase the efficiency of advanced oxidation processes, these processes are typically used as a combination of several advanced oxidation processes. Fe^0/UV, Fe^0/H_2O_2, and H_2O_2/UV processes are considered an advanced oxidation process, in which hydroxyl radicals are formed (Fig. 1) [3, 4].

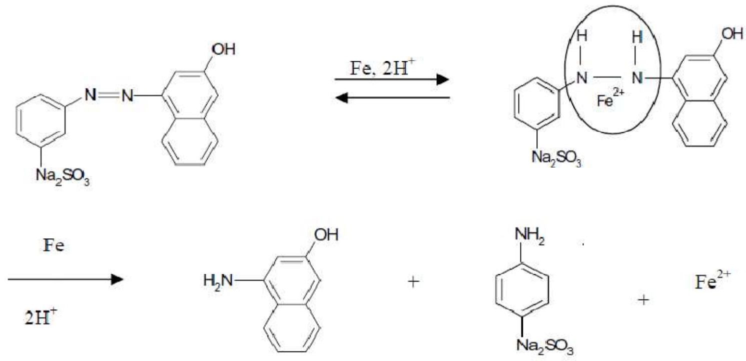

Figure 1. Analysis of Acid Orange 7 in Fe^0/H_2O system [3,4]

In H_2O_2/UV process, hydroxyl radical is produced according to (Equation 1):

$$H_2O_2 + UV \rightarrow 2OH^0 \qquad (1)$$

This process can convert colored wastewaters into the water, carbon dioxide, and nontoxic without creating secondary pollutants. The major advantage of this process is the very strong oxidation power, lack of sludge production, method simplicity, and relatively low investment costs [5].

Fe^0/UV process is considered as one of the advanced oxidation processes. In this process, hydroxyl radicals are produced in the absence of hydrogen peroxide which is one of the main advantages of this method. In fact, this is a Fenton process, in which hydrogen peroxide is not used. In this process, iron powder in aqueous environments causes catalyzing the production of hydroxyl radical in the presence of UV light according to (Equation 2):

$$Fe^0 + UV \rightarrow Fe^{+2} + 2e^- \qquad (2)$$

$$Fe^{+2} + H_2O_2 \rightarrow Fe^{+3} + OH^0 OH^-$$

Hydrogen peroxide in this reaction can be achieved through OH2 radicals in wavelengths less than 698nm (Equation 3):

$$2H_2O + UV(< 698nm) \rightarrow OH_2{}^0 + 3H^+ \qquad (3)$$

$$OH_2{}^0 (O_2{}^{0-}) + H^+ \rightarrow H_2O_2 + H_2$$

Based on these reactions, Fe^0/UV process causes producing Ferro ions and hydrogen peroxide will be generated through producing $OH_2{}^0$ radicals by UV light without the need to add hydrogen peroxide. Therefore, according to the above reactions, a combination of the above methods can greatly increase the efficiency of the process [5,6].

In a study, zero-valence iron powder was used to decompose Acid Orange 7 in the presence of ultrasonic waves that he results showed that color removal increased in the presence of ultrasonic waves and color removal reaction follows the equation [7].

The electro-Fenton process was used to remove Acid Orange 7 in a research. In this study, hydrogen peroxide was produced by adding some electrolyte solution and depletion of dissolved oxygen in the color solution. Electrolytes perchlorate, chloride, and sulfate was used in pH=3 that color removal was achieved in 90 minutes [8]. In another study, microwave signals were used to decompose Acid Orange 7 in the presence of hydrogen peroxide that decomposed the color by produced radicals from decomposing hydrogen peroxide. In addition, the color analysis which was performed by microwaves also significant [9]. Now, in this study, we want to assess the efficiency of UV/H2O2 advanced oxidation with zero-valent iron in the efficiency of AO7 pollutants from aqueous solutions.

MATERIALS AND METHODS

This study was began in order to investigate the removal of AO7 color from an aqueous solution based on UV/H2O2 advanced oxidation with zero-valent iron. For this purpose, the required materials for the test (Table 1) were prepared and used.

Table 1. Types of the required chemicals for the study

Name of chemical	Application	Production	Description
Sulfuric acid	PH adjustment	Merck	One normal
Soda	PH adjustment	Merck	One normal
Iron powder	Oxidation solution	Merck	The effective size 150 microns and purity of 98%
Liquid hydrogen peroxide	Oxidation solution	Merck	
Acid Orange 7	Test	Alvan Sabet Hamadan	97%
Filter paper		Merck	Pore diameter 45 microns
UV lamp	Oxidation solution	Nozhan Teb	40 V

The chemical structure of Acid Orange 7 color can be seen in Figure 2.

$$Fe^{+2} + OH_2^{\,0}(O_2^{\,0-}) \rightarrow Fe^{+3} + H_2$$

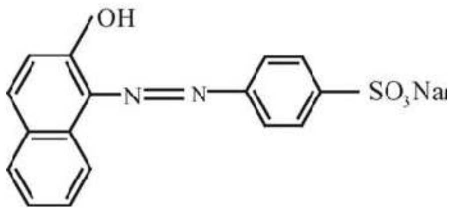

Figure 2. The chemical structure of the Acid Orange 7 color

Preparation of standard curve

In order to prepare the standard curve, initially the maximum absorption wavelength was determined for color Acid Orange 7 487nm by UV/VIS spectrophotometer model 1700 manufactured by Shimadzu Co. in Japan. Then, a specific concentration of color was prepared and their absorption was read by spectrophotometer and the standard curve was prepared in the following way.

In order to prepare the standard curve of Acid Orange 7 color, color solution with 2, 4, 8, 10, and 20ml concentration was prepared and their absorption was read based on percentage using UV/VIS spectrophotometer. Table 2 shows the results of the adsorption of the desired colors. In Figure 3, the standard curve of Acid Orange 7 color is shown.

Table 2. The read absorption of Acid Orange 7 color solutions

Color solution (mg/L)	Absorption (%)
2	0.226
4	0.448
6	0.682
8	0.917
10	1.149
20	2.46

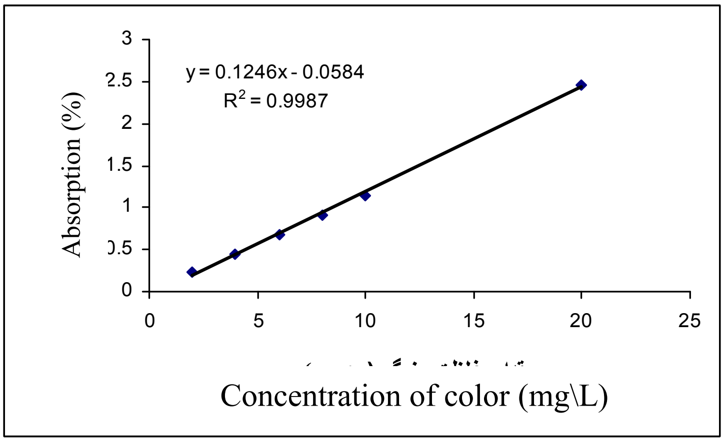

Figure 3. Standard curve diagram for Acid Orange 7

Process efficiency of iron powder was determined in the presence of hydrogen peroxide and UV radiation in the removal of AO7. The investigated parameters in this process were pH, contact time, initial concentration of color solution, the initial concentration of hydrogen peroxide and initial concentration of iron powder that in this stage, the removal efficiency of both colors were determined by fixing the 3 parameter and changing other two parameters. For example, at constant pH 7, the contact time of 120 minutes and 3 mL constant concentration of hydrogen peroxide, different concentrations of iron powder and color were added and the color removal efficiency was determined. In this study, changes of all parameters and their mutual impact on each other were studied.

Surveying the effect of pH on the efficiency of the process

In advanced oxidation system, since the investigated parameters were 5, the color removal efficiency was surveyed by fixing the 3 parameter and changing other two parameters in this system. For example, in the fixed time of 2 hours, 50mg\l the color constant concentration and 3ml per 150ml the constant concentration of hydrogen peroxide of color solution, different concentrations of iron powder at different pH were added and the effect of pH was determined.

Surveying the effect of iron powder concentration

In this system, the color removal efficiency was surveyed by fixing the 3 parameter and changing other two parameters. For example, in a fixed time of 2 hours, 3ml per 150ml the constant concentration of hydrogen peroxide of color solution and constant pH of 7 in different concentrations of color and different amounts of iron powder was added to the system and the system performance was determined.

Surveying the effect of contact time

In advanced oxidation system, the efficiency of contact time was measured by fixing the 3 parameter and changing other two parameters. For example, in 50mg\L the constant concentration of color, 3mL in 150mL the constant concentration of hydrogen peroxide of color solution and constant pH of 7, different amount of powder was added to the system and sampling was done at determined contact times and therefore, the remaining color concentration was determined.

Surveying the effect of the initial concentration of color

In this system, testing was repeated as well as the previous method. For this purpose, in the constant pH of 7, 0.3g in 150ml the constant concentration of iron powder and the fixed contact time of 120 minutes, different values of the initial concentration of color were added to the system in the different concentration of hydrogen peroxide and thus, the efficiency of the process were determined.

Surveying the effect of the initial concentration of hydrogen peroxide

In this section, the impact of the initial concentration of hydrogen peroxide in the presence of UV radiation and iron powder was investigated. For this purpose, tests were done fixing the 3 parameter and changing other 2 parameters. For example, in the constant pH of 7, the constant contact time of 2 hours, 50mg\l the initial concentration of color, and different amounts of iron powder concentration, different concentration of hydrogen peroxide were added to the system and thus, the amount of color reduction was calculated.

RESULTS AND DISCUSSION

The removal tests results of AO7 color are given in Figure 4 to 13 using ZVI/UV/H_2O_2.

Table 3. The removal tests results of AO7 color using ZVI/UV/H_2O_2 (hydrogen peroxide concentration: 3ml per 150ml, iron powder concentration: 0.3g in 150ml and pH=7)

Color concentration / Contact time	75 mg/L	50 mg/L	25 mg/L
30 (minutes)	41.54	29.64	13.57
60 (minutes)	19.81	17.35	1243
120 (minutes)	11.88	8.39	6.64

According to the results of Table 3, by increasing the initial concentration of color, the removal efficiency of color decreases but by increasing the contact time, the efficiency of the process increases.

Table 4. The removal tests results of AO7 color using ZVI/UV/H_2O_2 (hydrogen peroxide concentration: 3ml per 150ml, iron powder concentration: 0.3g in 150ml and contact time: 2 hours)

pH / Color concentration	11	7	3
25 mg/L	20.17	7.31	0.41
50 mg/L	38.61	9.47	0.821
75 mg/L	43.47	11.53	1.023

Results of Table 4 show that increasing pH and the initial concentration of color both reduce the efficiency of the process.

Table 5. The removal tests results of AO7 color using ZVI/UV/H_2O_2 (hydrogen peroxide concentration: 3ml per 150ml, iron powder concentration: 0.3g in 150ml and color concentration: 50mg\L)

pH / contact time	11	7	3
30 (minutes)	43.65	22.16	2.17
60 (minutes)	40.51	16.43	1.58
120 (minutes)	37.23	9.04	0.79

Results of Table 5 showed that increasing the contact time increases the color removal and increasing pH decreases the efficiency of the process.

Table 6. The removal tests results of AO7 color using ZVI/UV/H_2O_2 (hydrogen peroxide concentration: 3 ml per 150 ml, iron powder concentration: 0.3g in 150 ml and pH=7)

Color concentration / Iron powder concentration	75 mg/L	50 mg/L	25 mg/L
0.1(g)	37.25	19.37	11.21
0.2(g)	24.71	12.32	7.34
0.3(g)	11.41	7.87	5.14

Results of Table 6 showed that increasing the concentration of iron powder increases the color removal while increasing the initial concentration of color reduces the efficiency of the process.

Table 7. The removal tests results of AO7 color using ZVI/UV/H_2O_2 (hydrogen peroxide concentration: 3ml per 150ml, color concentration: 50mg\L and pH=7)

Iron powder concentration / Contact time	0.3(g)	0.2(g)	0.1(g)
30 (minutes)	29.66	34.27	40.62
60 (minutes)	20.14	23.64	33.64
120 (minutes)	9.21	12.46	19.21

Results of Table 7 showed that by increasing the concentration of iron powder and contact time, the color removal efficiency increases.

Table 8. The removal tests results of AO7 color using ZVI/UV/H_2O_2 (hydrogen peroxide concentration: 3ml per 150ml, color concentration: 50mg\L and contact time: 2 hours)

pH / Iron powder concentration	11	7	3
0.1(g)	41.52	24.71	1.87
0.2(g)	38.68	11.27	1.24
0.3(g)	36.43	9.25	0.71

In Table 8, it was shown that increasing the concentration of iron powder increases the color removal efficiency while by increasing pH, the system performance decreases.

Table 9. The removal tests results of AO7 color using ZVI/UV/H_2O_2 (contact time: 2 hours, iron powder concentration: 0.3g in 150ml, and pH=7)

H_2O_2/UV / Color concentration	3 mL/150 mL	2/25 mL/150 mL	1/5 mL/150 mL
25 mg/L	7.11	8.74	10.25
50 mg/L	9.02	11.21	12.24
75 mg/L	11.36	13.41	15.25

The results of Table 9 show the increasing the initial concentration of hydrogen peroxide increases the color removal efficiency. Also, this table shows that increasing the initial concentration of the color solution reduces the efficiency of the process.

Table 10. The removal tests results of AO7 color using $ZVI/UV/H_2O_2$ (color concentration: 50mg\L, iron powder concentration: 0.3g in 150ml, and pH=7)

H_2O_2/UV / Contact time	3 mL/150 mL	2/25 mL/150 mL	1/5 mL/150 mL
30 (minutes)	16.11	17.62	19.52
60 (minutes)	12.54	14.57	16.23
120 (minutes)	9.13	11.43	12.37

According to the data shown in Table 10, it can be concluded that increasing the initial concentration of hydrogen peroxide and contact time increases the efficiency of the process.

Table 11. The removal tests results of AO7 color using $ZVI/UV/H_2O_2$ (contact time: 2 hours, color concentration: 50mg\L, and pH=7)

H_2O_2/UV / Iron powder concentration	3 mL/150 mL	2/25 mL/150 mL	1/5 mL/150 mL
0.1(g)	24.64	31.2	38.76
0.2(g)	11.75	17.4	26.43
0.3(g)	9.22	11.63	12.21

As shown in Table 11 by increasing the initial concentration of hydrogen peroxide and also by increasing the initial concentration of iron powder, the removal efficiency of the system is increased substantially.

Table 12. The removal tests results of AO7 color using $ZVI/UV/H_2O_2$ (contact time: 2 hours, iron powder concentration: 0.3g in 150ml, and color concentration: 50mg\L)

pH / H_2O_2/UV	11	7	3
1/5 mL/150 mL	43.11	12.71	4.17
2/25 mL/150 mL	39.19	11.53	2.14
3 mL/150 mL	37.21	9.37	0.53

According to Table 12, it can be concluded that increasing pH has a negative impact on the efficiency of the process and reduces the efficiency of the process while increasing the initial concentration of hydrogen peroxide at different pH increases the efficiency of the process.

Figure 4 shows the effect of pH change on the removal efficiency of AO7 color using $ZVI/UV/H_2O_2$. The results showed that in $ZVI/UV/H_2O_2$ system, by increasing pH from 3 to 11, the removal efficiency for Acid Orange 7 decreases from 98% to 24%.

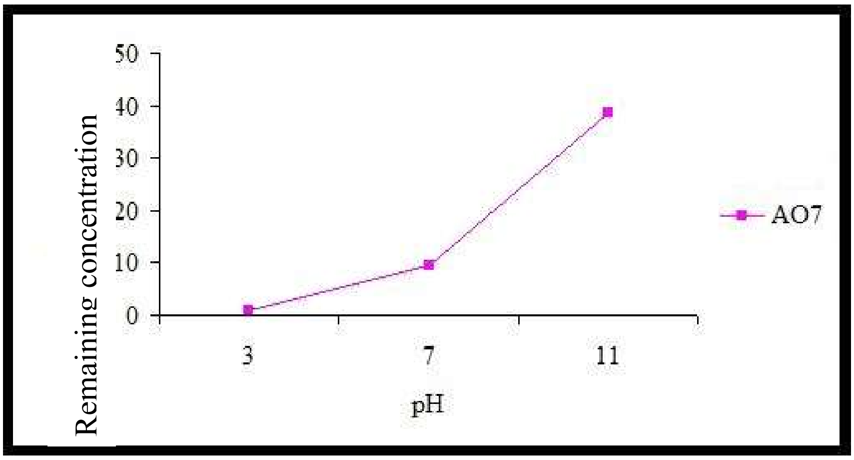

Figure 4. Evaluating the impact of pH in removal of Acid Orange 7 color in $ZVI/UV/H_2O_2$ system (hydrogen peroxide concentration: 3 ml per 150 ml, iron powder concentration: 0.3g in 150 ml, and initial concentration of color: 50mg\L)

In the present study, the reason for increasing the efficiency of the advanced oxidation process of zero-valence iron powder in the presence of UV rays and hydrogen peroxide and also, acidification of the environment are the following cases:

- In case of adding hydrogen peroxide to the zero valent iron powder system, practically, the production of free and regenerative electrons becomes pale and the process will go ahead towards semi-Fenton process and more production of hydroxyl radicals.

- UV radiation on the one hand will directly attack the molecules of dye and on the other hand, by the production of hydroxyl radicals in the presence of zero-valent iron increases the efficiency of the process.
- The presence of UV radiation accelerates the decomposition of hydrogen peroxide in the environment and increases the production of hydroxyl radicals in the environment.
- In the case of UV radiation in the environment, OH_2^0 radicals will not be formed [10,11].

The impact of changing the iron powder in ZVI/UV/H_2O_2 system was investigated by changing the contact time, pH, the initial concentration of color and initial concentration of hydrogen peroxide. The impact of changing the iron powder concentration on the color removal efficiency of the system is shown in Figure 5. The results of the tests showed that by increasing the iron powder concentration from 0.1g to 0.3g in 150mL in the system, the efficiency of the process increases. In ZVI/UV/H_2O_2 system, in contact time of 120 minutes, 7 = pH, 50mg\L the initial concentration of color and 3ml per 150ml the initial concentration of hydrogen peroxide, by increasing the initial concentration of iron powder from 0.1g to 0.3g in 150mL, the removal efficiency for Acid Orange 7 color reached 84% from 62%. In the advanced oxidation system, in the contact time of 120 minutes, 7 = pH, 3ml in 150ml the initial concentration of hydrogen peroxide and 50mg/L the initial concentration of color, by increasing the initial concentration of iron powder from 0.1g to 0.3g in 150ml, the removal efficiency for Reactive Black 5 color reached 76% from 48% and for Acid Orange 7 reached 84% from 62%. In the advanced oxidation system of ZVI/UV/H_2O_2, increasing the iron powder increases the production of hydroxyl radicals because increasing the initial concentration of iron powder increases the active surface of metal to contact with hydrogen peroxide and UV radiation. In this system, as well as zero-valent iron powder, increasing the concentration of zero valence iron powder can cause a rapid increase in the efficiency of the process [14-12].

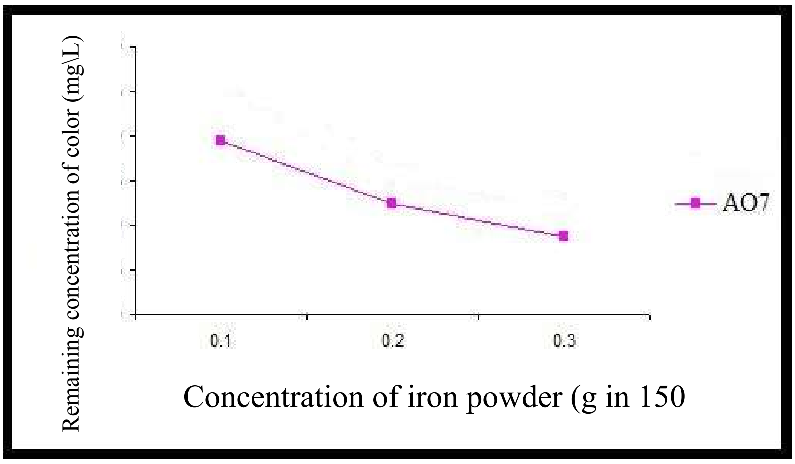

Figure 5. Evaluating the impact of iron powder concentration in removing Acid Orange 7 color in ZVI/UV/H_2O_2 system (7 = pH, 3 ml in 150 ml hydrogen peroxide, 120 minutes contact time and 50 mg/L initial concentration of color)

The removal efficiency of Acid Orange 7 color in three time range of 30, 60, and 120 minutes in ZVI/UV/H_2O_2 system and in contrary, pH changes, the initial concentration of color, the initial concentration of iron powder and the initial concentration of hydrogen peroxide were studied. The results of the study showed that the removal efficiency increases with increasing the contact time. In the advanced oxidation system of ZVI/UV/H2O2, the removal efficiency of Acid Orange 7 reached 82% from 56% by increasing the contact time from 30 minutes to 120 minutes. Contact time is an important parameter in chemical reactions. Basically, a contact time or a balance time can be defined in chemical reactions for the desired reaction. Balance time is a time, in which the amount of the pollutants removal reaches a constant value. In other words, if the contact time be more than the balance time, the process would not be economically viable because the amount of removal is very low. In the current study, in the advanced oxidation system of ZVI/UV/H2O2, increasing the contact time increases the removal efficiency of color. This increase has a similar gradient for AO7 color. As previously mentioned, the efficiency of the advanced oxidation processes depends on producing hydroxyl radicals. Increasing the contact time in this type of system will increase the hydroxyl radicals' production. In this system, whatever the contact time increases, the color removal efficiency will increase and as seen on Figure 6, the reaction has an ascending trend and it is still not balanced. In this system, the metal iron surface will be constantly kept clean in UV radiation. On the other hand, OH_2^0 radicals will be decomposed by UV radiation. Also, direct exposure of UV rays will also decompose colors. Therefore, the system performance will continue [10 and 11].

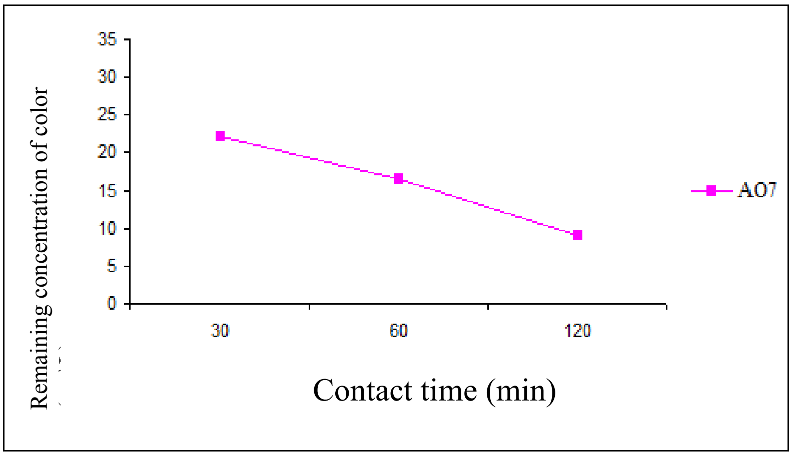

Figure 6. Evaluating the impact of contact time in removing Acid Orange 7 color in ZVI/UV/H_2O_2 system (3 ml in 150 ml hydrogen peroxide, 0.3mg in 150ml the initial concentration of iron powder, and 50 mg/L initial concentration of color)

In order to evaluate the effect of initial concentration of color on removal efficiency of AO7 color with concentrations of 25, 50, and 75mg\L at different pH, different concentrations of zero valence iron powder, various concentrations of hydrogen peroxide and contact times were considered and the color removal efficiency was calculated. The results of the tests are shown in Figure 7. In ZVI/UV/H2O2 system, in pH=7, the contact time of 120 minutes, 0.3g in 150ml concentration of iron powder and 3ml in 150ml the initial concentration of hydrogen peroxide, by increasing the initial concentration of color from 25 to 75mg\L, the removal efficiency of color reduced to 63% from 72%. In the advanced oxidation system of ZVI/UV/H2O2, since, hydroxyl radical production also remains constant under constant conditions, therefore, by increasing the amount of pollutants in the environment, the radicals will be consumed. Thus, by increasing the concentration of pollutants the efficiency of the process will be decreased [15].

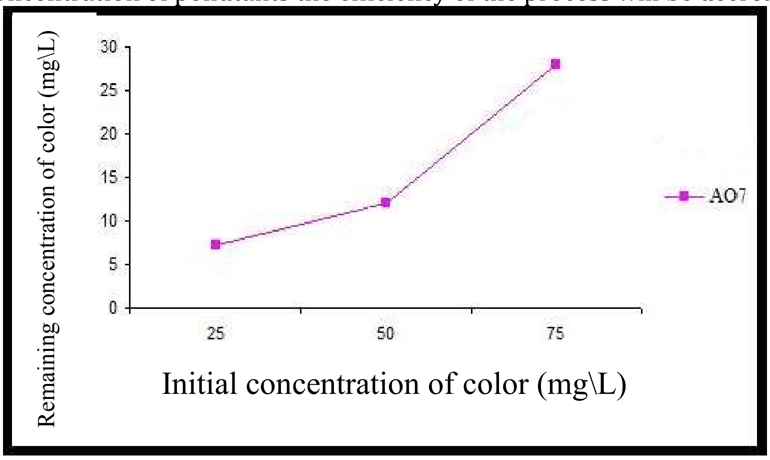

Figure 7. Evaluating the impact of the initial concentration of Acid Orange 7 color in ZVI/UV/H_2O_2 system (3 ml in 150 ml hydrogen peroxide, 0.3mg in 150ml the initial concentration of iron powder, 120 minutes contact time, and pH=7)

Hydrogen peroxide with the initial concentrations of 1.5, 2.25, and 3ml in 150ml in the advanced oxidation system of ZVI/UV/H2O2 has evaluated the removal efficiency of Acid Orange 7 color by changing the contact time, pH, initial concentration of iron powder and initial concentration of color. The results of the tests showed that by increasing the initial concentration of hydrogen peroxide in the system, the color removal efficiency increases. The results of these tests are shown in Figure 8. In the advanced oxidation system of ZVI/UV/H2O2, in pH=7, contact time of 120 minutes, 0.3g in 150ml concentration of iron powder and 50mg\L the initial concentration of color, by increasing the initial concentration of hydrogen peroxide from 1.5 to 3ml in 150ml, the removal efficiency of Acid Orange 7 color reached 82% from 72%.

In the advanced oxidation system of ZVI/UV/H2O2, increasing the initial concentration of hydrogen peroxide increases the efficiency of the process. In this system, as shown in Figure 8, increasing the initial concentration of hydrogen peroxide quickly increases the efficiency of the process. In the advanced oxidation system of ZVI/UV/H2O2, increasing the initial concentration of hydrogen peroxide increases the production of hydroxyl radicals and no OH_2^0 radicals will be formed in UV radiation. Thus, the efficiency of the process will continue with a sharp slope [2].

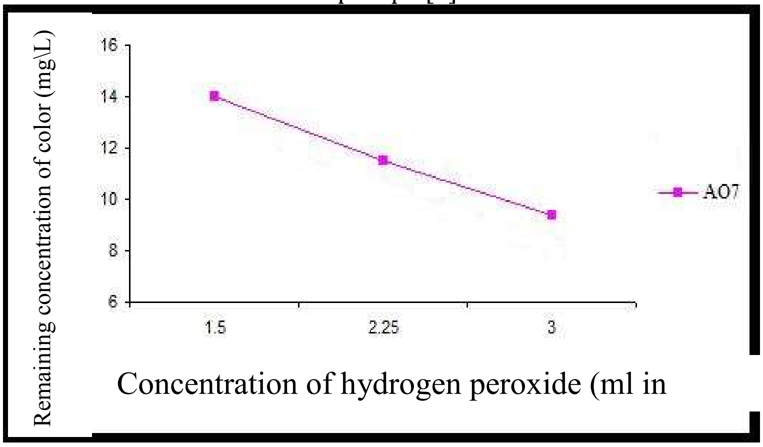

Figure 8. Evaluating the impact of the initial concentration of hydrogen peroxide in removing Acid Orange 7 color in ZVI/UV/H_2O_2 system (pH=7, 120 minutes contact time, 50 mg/L initial concentration of color, and 0.3mg in 150ml the initial concentration of iron powder)

CONCLUSION

In this study, the removal efficiency of Acid Orange 7 color was investigated using the advanced oxidation ZVI/UV/H2O2 system. The results of the tests showed that by increasing the contact time, the initial concentration of hydrogen peroxide and the initial concentration of iron powder, the color removal efficiency increases while by decreasing pH and the initial concentration of color, the color removal efficiency decreased. The maximum amount of color removal in the system was obtained in pH=3, contact time of 120 minutes, 3ml in 150ml hydrogen peroxide, 0.3mg in 150ml iron powder and 25 mg\L the initial concentration of color. The results indicated the acceptable removal of Acid Orange 7 color by advanced oxidation system with zero-valent iron powder.

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