



# Comparison of Photo Fenton and H<sub>2</sub>O<sub>2</sub>/UV Advance Oxidation Process in treating textile Wastewater for Reduction of COD and Color

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**Abstract:** Contaminants in textile wastewater are characterized as Recalcitrant, which usually contains high concentrations of organic compounds, expressed as high COD, a varied range of pH, low biodegradability, and the presence of colors. Such waste is difficult to treat by conventional wastewater treatment methods because the majority of the dyes present in textile wastewater are non-biodegradable and have negative effect to microorganisms. In this study two advanced oxidation process namely photo Fenton and H<sub>2</sub>O<sub>2</sub>/UV process investigated to compare their pollutant removal efficiency from live textile wastewater in terms of color removal and reduction of COD. The result shows that even though both process are capable of removing pollutant from the wastewater photo Fenton process has better reduction efficiency compared to H<sub>2</sub>O<sub>2</sub>/UV with a deviation of 19.4 % in decolorization and 25.6% in COD Reduction.

**Keywords:** Advanced oxidation process; COD Reduction; Decolorization, Photo-Fenton process, H<sub>2</sub>O<sub>2</sub>/UV process,

## I. INTRODUCTION

Industrialization advancements in dye production and dye effluents have a huge toll on the environment because of dye effluent. From various industries, Textile industries consume the maximum percentage of dyes, which is around 56 % of the world's total yearly production [1]. Various carcinogenic and toxic chemical compounds are introduced into marine life, in addition to humans and animals Due to the discharge of dye effluent into the water bodies. There are many classes of dyes, from those dyes azo and diazo dyes, which are widely used in printing processes and industrial dyeing [1]. Roughly, 70 % of dyes commonly used in industries are azo dyes [2]. Treating these dyes (Azo/diazo dyes) by conventional treatment processes is difficult to degrade. Non-biodegradable components pose a major hazard to the environment and every living thing living in it. Industries like textile production, dye manufacturing industries, etc. Face a real problem to removing the residual dyes from their Wastewater to a satisfactory and sometimes acceptable level before they discharge their effluents into the stream, land, etc. Treatments of effluents containing dyes and chemicals by conventional treatment processes (flocculation, trickling filters, electro-dialysis, etc.) are not effective toward all dyes [3], [4].

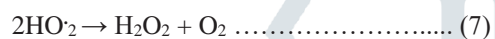
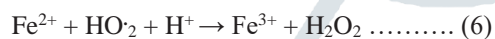
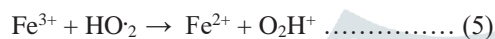
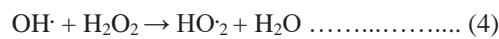
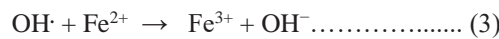
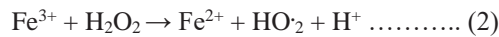
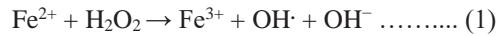
## II. ADVANCE OXIDATION PROCESS

Advanced Oxidation Processes (AOPs) are chemical treatment processes that generate hydroxyl radicals (OH<sup>•</sup>) designed to remove non-biodegradable compounds present in water and mostly in wastewater through oxidation by reacting with OH<sup>•</sup> radicals which are highly reactive. This radical is the strongest oxidizing species and has an oxidation power of 2.8 eV. It produces total mineralization by converting almost all recalcitrant compounds into CO<sub>2</sub> and H<sub>2</sub>O or partial mineralizing them, which means converting these compounds into more biodegradable substances so that they can be removed easily by a biological treatment

system. AOP are powerful systems capable of converting contaminants within a short reaction time into harmless substances and used for the treatment of effluents generated from different industries such as pulp and paper petrochemical, chemical, dye manufacturing, textile, and other industries. These advanced oxidation processes are considered suitable processes for handling the refractory pollutants present in industrial wastewater to improve the water quality. There are several technologies included in AOP such as Fenton reagent which is the combination of iron and hydrogen peroxide, photo-Fenton, ozonation,  $H_2O_2/UV$ , etc. the difference between these processes is that the source of radical generation.

### III. FENTON RELATED AOPs

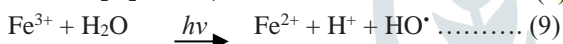
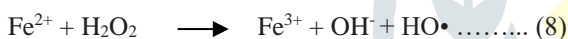
To understand the photo Fenton process, we have to understand the Fenton process. Fenton process is the combination of  $Fe^{2+}$  and  $H_2O_2$ . In Fenton-Related AOPs  $H_2O_2$  have to be activated to generate  $OH\cdot$  and iron ( $Fe^{2+}$ ) is the most commonly used to activate  $H_2O_2$  and generate hydroxyl radicals. This hydroxyl radical attacks organic molecules by abstracting hydrogen atoms. Organic molecules are then converted to  $CO_2$  and  $H_2O$ . The reactions involved in this process (Fenton related radical production) are



When it comes to removing recalcitrant compounds, most often this process is suitable as a treatment. Although, the major downside of the Fenton reaction process is the iron sludge waste production at the end of the process [5].

#### a) Photo-Fenton AOPs

The Photo-Fenton process is a reaction of  $H_2O_2$  and  $Fe^{2+}$  in the presence of UV-vis light radiation. After the reaction of  $H_2O_2$  and  $Fe^{2+}$ , the ferric ions ( $Fe^{3+}$ ) will be produced with additional formation hydroxyl radical (8). The ferric ions ( $Fe^{3+}$ ) are then photo catalytically converted back to ferrous ions ( $Fe^{2+}$ ) in the presence of UV light as shown in (9). This shows that the Photo-Fenton process reduces the iron sludge waste, which encountered when applying Fenton reagent AOP [6].



The degradation of contaminants occurs when the hydroxyl radicals formed in these two reactions react with organic pollutants present in the wastewater. When UV radiation is applied to the reaction of the oxidant and reagent, the rate of decomposition of organic pollutants becomes faster. This is due to the photochemical reduction of  $Fe^{3+}$  back to  $Fe^{2+}$  as shown in (9). The degradation rate is significantly influenced by the pH of the solution [7].

### IV. MATERIALS AND METHOD

Raw textile wastewater (sample) was collected from textile industry to perform the experiment. The peak wavelength and Absorbance was found using shimadzu UV spectrophotometer. COD was measured with closed reflux titration method with 0.1M of Ammonium Ferrous Sulphate ( $Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$ ). The pH of the wastewater measured using Hana digital pH meter. The Characteristics of the wastewater shown in Table 1. Analytical grade  $H_2O_2$  30 % (w/v) and photo reactor fitted with 16W medium pressure UV light with wavelength of 254 nm used in the experiment for both processes. For photo Fenton process analytical grade Ferrous Sulphate heptahydrate ( $FeSO_4 \cdot 7H_2O$ ) used as a catalyst. The pH of the wastewater adjusted before every experiment using 1N NaOH and  $H_2SO_4$ . The experimental procedure in photo Fenton process start by adjusting the pH of the wastewater followed by addition of  $FeSO_4 \cdot 7H_2O$ . After 15 minute homogenization on the stirrer  $H_2O_2$  was added and the UV light turned on immediately and start the timer. In  $H_2O_2/UV$  process the experiment is the same without the addition of iron ( $FeSO_4 \cdot 7H_2O$ ) in the process.

Table – 1, Characteristics of the raw textile wastewater (sample)

pH	7	Color at 728 nm $\lambda$ (Abs)	0.930
COD (mg/l)	1500	TDS (mg/l)	2900

## V. RESULT AND DISCUSSION

### A. Photo Fenton process

#### 1. pH adjustment

To optimize the optimum pH value the effect of pH on decolorization and reduction of COD of the wastewater was studied by varying pH value from 1 to 5 and keeping the  $\text{H}_2\text{O}_2$  and  $\text{Fe}^{2+}$  dose constant. This process was carried out by Fenton reagent process since both Fenton reagent and photo Fenton process share same initial pH operating condition. The initial  $\text{H}_2\text{O}_2$  dose was decided as per the stoichiometric requirement with respect to COD, as illustrated bellow [25].

$$1\text{g COD} = 0.0625 \text{ mol of } \text{H}_2\text{O}_2 = 2.425 \text{ g of } \text{H}_2\text{O}_2 \dots (10)$$

Based on the initial COD of the wastewater (1500 mg/L), doses of  $\text{H}_2\text{O}_2$  required for the process were calculated. The initial  $\text{Fe}^{2+}$  dose for pH optimization is decided by a molar ratio of 10:1 ( $\text{H}_2\text{O}_2:\text{Fe}^{2+}$ ). The degradation of pollutants for each pH Value (1, to 5) with every 30-minute reaction time interval was studied the effect of pH on the decolorization and COD reduction efficiency of the wastewater was studied by varying pH values and keeping the  $\text{H}_2\text{O}_2$  and  $\text{Fe}^{2+}$  dose constant, where the  $\text{H}_2\text{O}_2$  dose was 93.75 mM and  $\text{Fe}^{2+}$  dose was 9.375 mM for 2 hrs of reaction time and volume of the wastewater sample was 1L. As the result shows when pH increases from 1-5 the decolorization and COD reduction efficiency increases up to a pH range of 3 from 7% to 82% in decolorization and from 5 to 78.1 % in COD reduction. But later as the pH range increases from 3 to 5 the decolorization and COD removal slowly decreases to 70 % and 40% respectively. The detailed result illustrated in Fig. 1. From this it can be concluded when that the pH increase above than pH=3 it leads to a decrease in both decolorization and COD reduction efficiency. The optimum pH value found to be pH=3 with 82% and 78% of decolorization and COD reduction efficiency respectively. This result agreed with the literature included in this paper.

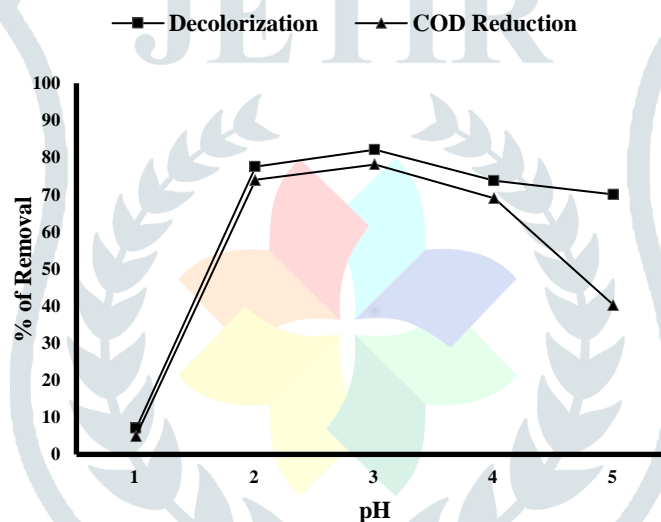


Fig 1. Degradation efficiency and effect of pH on Decolorization and COD Reduction

#### 2. $\text{H}_2\text{O}_2$ Adjustment for photo Fenton process

The dose of  $\text{H}_2\text{O}_2$  was optimized by varying the  $\text{H}_2\text{O}_2$  dose from 50 - 150 mM, using the optimum pH and keeping the  $\text{Fe}^{2+}$  concentration and reaction time constant. The degradation efficiency that the change of  $\text{H}_2\text{O}_2$  dose from 50 to 150mM has on decolorization and reduction of COD of the wastewater was studied. The effect of  $\text{H}_2\text{O}_2$  on the decolorization and COD reduction efficiency of the wastewater was studied by varying the  $\text{H}_2\text{O}_2$  dose and keeping the rest of the variable parameter constant ( $\text{Fe}^{2+}$  =9.375Mm, pH=3, 2hrs of reaction time) and summarized in Fig.2. As the result shows with increasing  $\text{H}_2\text{O}_2$  dose from 50 up to 93.75mM the degradation efficiency increases from 86.2 to 96.7% in decolorization and from 85% to 92.53% in COD reduction. This shows that more hydroxyl radicals are available and utilized in the process to degrade the pollutant present in the wastewater however as the  $\text{H}_2\text{O}_2$  dose increases from 93.75mM to 150Mm the degradation efficiency slowly starts to decrease from 92.53% to 74% in COD reduction and from 96.7% to 76.3% in decolorization. This indicates that the increase in  $\text{H}_2\text{O}_2$  dose has a positive effect on the degradation but that is only up to a certain point as hydrogen peroxide itself inhibits the process by reacting with hydroxyl radicals and producing  $\text{HO}_2^{\cdot}$  which is less reactive than  $\text{OH}^{\cdot}$ . When the concentration of  $\text{H}_2\text{O}_2$  becomes excess in the system  $\text{H}_2\text{O}_2$  consumes  $\text{OH}^{\cdot}$  radical which is called the scavenging effect of  $\text{H}_2\text{O}_2$  [8]. The effect of each utilized dose of  $\text{H}_2\text{O}_2$  on the rate of degradation in both decolorization and COD reduction at 30 min intervals is illustrated in Fig. 3. (a) and (b) respectively. As the result shows in all cases of the results when a concentration of 93.75mM of  $\text{H}_2\text{O}_2$  has utilized the degradation efficiency is better than every other  $\text{H}_2\text{O}_2$  concentration used in the process with 78.4%, 82.8%, 88.7%, 96.7% of decolorization and 74.4%, 75.4%, 80.8% and 92.53% of COD reduction at each 30 minutes time interval.

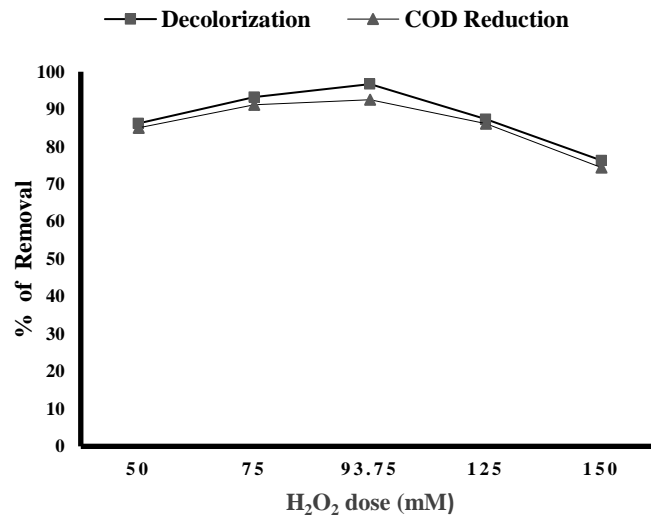


Fig. 2. Degradation efficiency and effect of H<sub>2</sub>O<sub>2</sub> Dose on Decolorization and COD Reduction in photo Fenton process

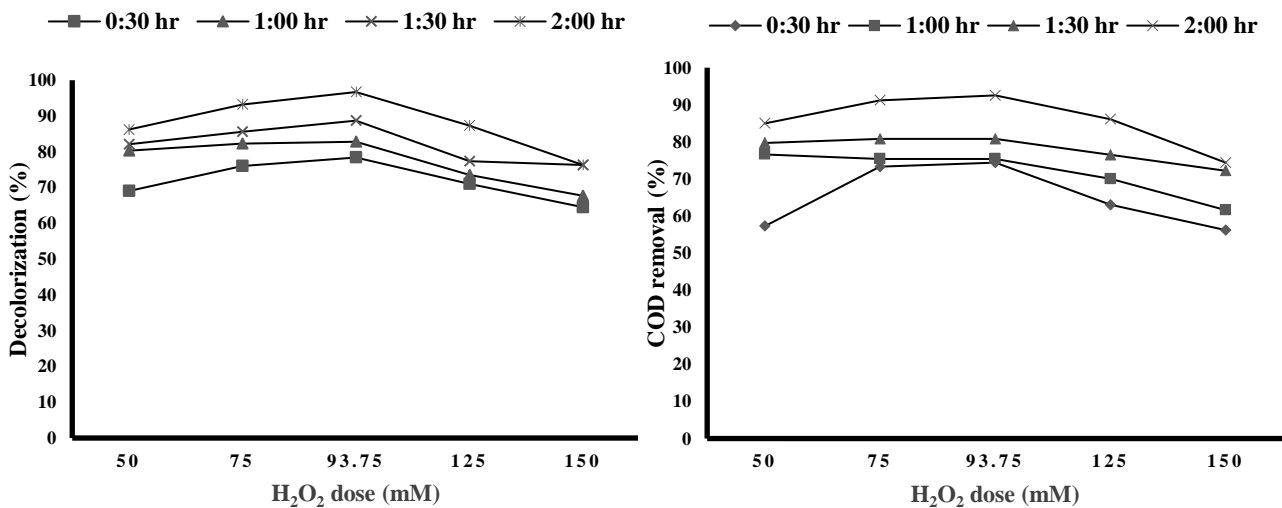


Fig. 3. Rate of (a) decolorization (b) COD reduction at every 30 minutes Reaction Time Interval at different H<sub>2</sub>O<sub>2</sub> Dose in photo Fenton process

### 3. Fe<sup>2+</sup> dose adjustment

The dose of Fe<sup>2+</sup> was optimized by varying the Fe<sup>2+</sup> dose from 2mM – 12mM, using the optimum pH and concentration of H<sub>2</sub>O<sub>2</sub>. The degradation efficiency that the change of Fe<sup>2+</sup> dose from 2mM to 9.375mM has on decolorization and reduction of COD of the wastewater was studied. The effect of Fe<sup>2+</sup> on the decolorization and COD reduction efficiency of the wastewater was studied by varying the Fe<sup>2+</sup> dose (2mM – 12mM) and using the optimized independent operation parameters (H<sub>2</sub>O<sub>2</sub>=93.75mM, pH=3) for 2hrs of reaction time. The result is illustrated in Fig. 4. As the result shows with increasing Fe<sup>2+</sup> dose from 2mM up to 8mM the degradation efficiency increases from 77.1% to 98% in decolorization and from 74.4% to 93.6% in COD reduction. Further increasing the Fe<sup>2+</sup> dose leads to a reduction of degradation performance from 98% to 62% in decolorization and from 93.6% to 54.1% in COD reduction after 2hrs of reaction time. This shows that excess Fe<sup>2+</sup> concentration in the system also affects the process by reacting with hydroxyl radical which is illustrated in (4). This will make hydroxyl radical less available in the system which ultimately affects the degradation efficiency. The effect of each utilized dose of Fe<sup>2+</sup> on decolorization and COD reduction at 30 min intervals for each dose is illustrated in Fig. 5 (a) and (b) respectively. Better degradation efficiency achieved when Fe<sup>2+</sup> dose is set to 8mM with 84.3%, 84.3%, and 98% of decolorization efficiency, and 81.8%, 84%, 89.3% and 93.6% of COD reduction efficiency at each 30 minutes time interval of reaction time.

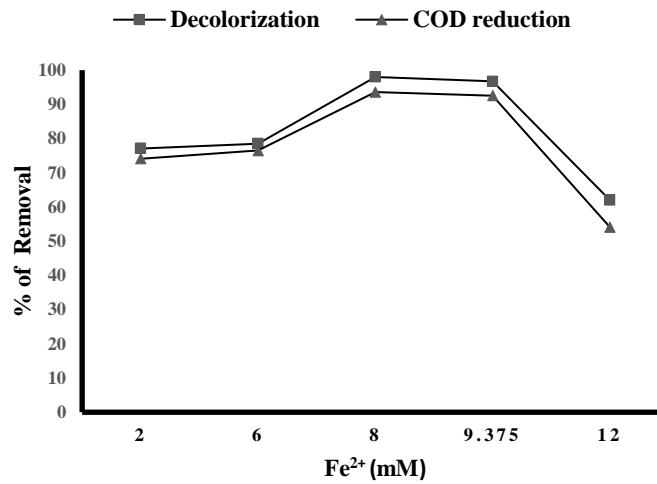


Fig. 4. Degradation efficiency and effect of Fe<sup>2+</sup> on Decolorization and COD Reduction

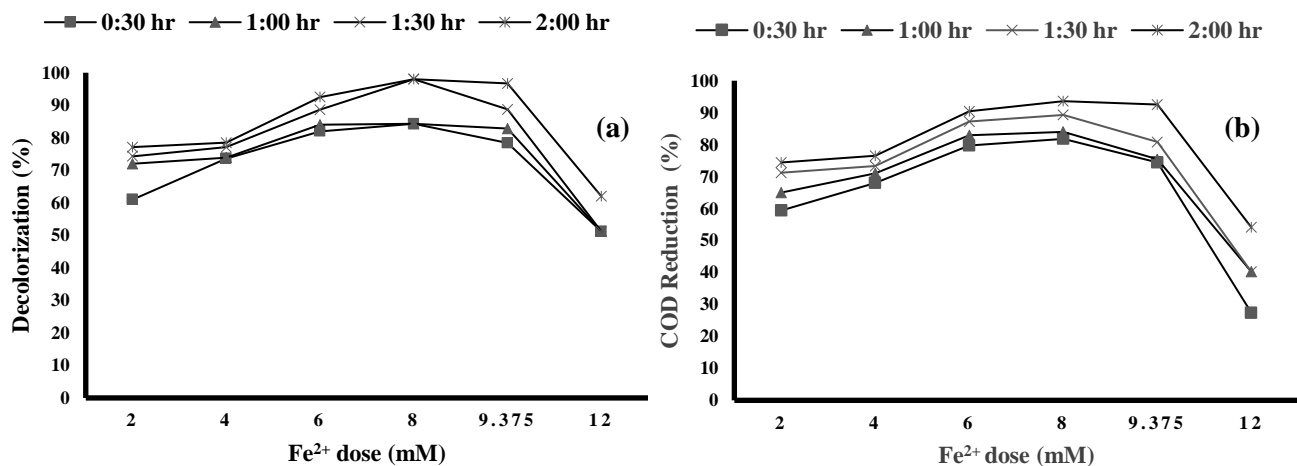


Fig. 5. (a) Rate of decolorization and (b) Rate of COD reduction decolorization at every 30 minutes Reaction Time Interval at different Fe<sup>2+</sup> Dose, H<sub>2</sub>O<sub>2</sub>= 93.75Mm, pH=3, reaction time = 2hrs

#### 4. Reaction Time Optimization for Photo Fenton process

Reaction time has been optimized by varying time of reaction and using all the optimized variable parameters and the result are illustrated in Fig. 6. As it shown even though time has a Positive effect on the final performance in terms of total mineralization but increasing the reaction time from 120 minutes (2hrs) up to 180 minutes (3hrs) doesn't have a significant change with 94.6% of COD reduction efficiency compared to that of 120 min (2hrs) of reaction time performance efficiency with 93.6% of COD reduction.

### B. H<sub>2</sub>O<sub>2</sub>/UV process

#### 1. pH adjustment

Adjustment of the optimum pH value has been carried out by varying pH value from 2 to 7 and keeping the dose of H<sub>2</sub>O<sub>2</sub> and time of the reaction constant (H<sub>2</sub>O<sub>2</sub> = 93.75Mm, time of reaction 2hrs). The initial H<sub>2</sub>O<sub>2</sub> dose was decided as per Eqn. (10). To find out what is the optimum pH range for better performance for this process study was carried out. The result illustrated in Fig. 7 shows that better efficiency achieved in the pH range of 2 up to 3 where 3 being the optimum pH value with 70% of decolorization and 56.2 % of COD reduction efficiency. When pH range increases from 3 up to 7 the decolorization and COD reduction efficiency decreases to 47.4% and 27.4% respectively.

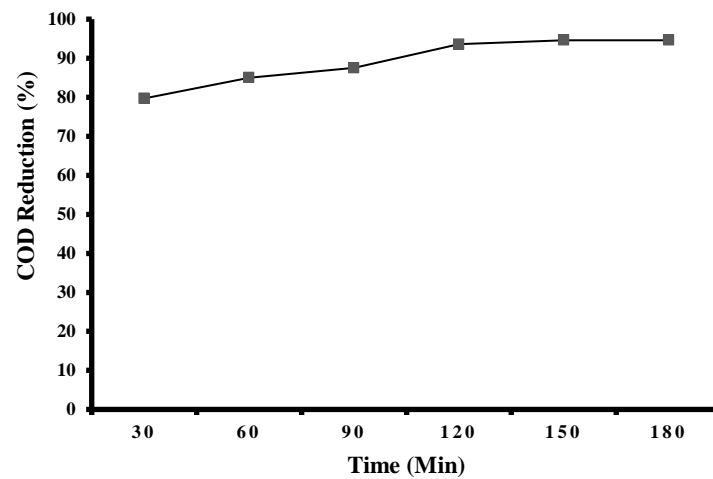


Fig. 6. Degradation efficiency and effect of reaction time on COD Reduction in photo Fenton process

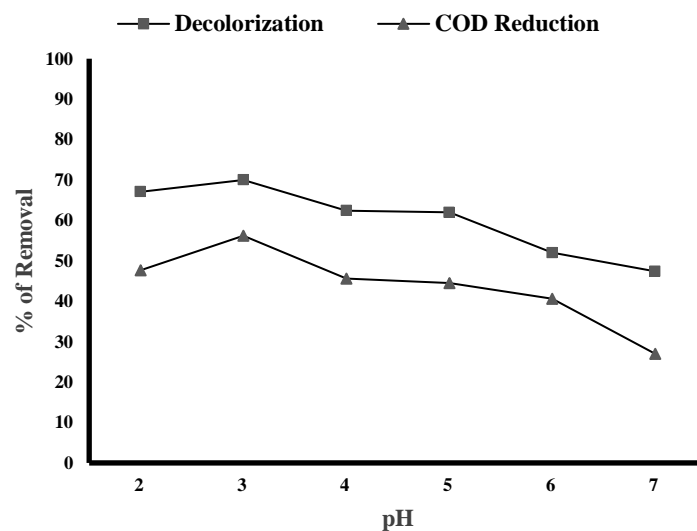


Fig. 7. Degradation efficiency and effect of pH in terms of Decolorization and COD Reduction

## 2. $H_2O_2$ Dose optimization for $H_2O_2/UV$ Process

The dose of  $H_2O_2$  was optimized by varying the  $H_2O_2$  dose from 25 - 125 mM, using the optimized pH and keeping the time of reaction constant. The degradation efficiency that the change of  $H_2O_2$  dose from 25 to 125mM has on decolorization and reduction of COD of the wastewater at every 30 minutes intervals was studied. The effect of excess concentration of  $H_2O_2$  has on the decolorization and COD reduction efficiency of the wastewater was studied by varying the  $H_2O_2$  dose and keeping the rest of the Independent variable parameter constant (pH=3, 2hrs of reaction time) and illustrated in Fig. 8. With increasing  $H_2O_2$  dose from 25 up to 75mM the degradation efficiency increases from 40% to 78.4% in decolorization and from 31.7 to 68% in COD reduction. This indicates that more hydroxyl radicals are available and utilized in the process to degrade the pollutant present in the wastewater when 75mM  $H_2O_2$  dose applied in the system. However as the  $H_2O_2$  dose increases from 75mM to 125mM the degradation efficiency slowly starts to decrease from 78% to 62.58% in decolorization and from 68% to 40.8% in COD reduction efficiency. This again can be explained by the scavenging effect of excess concentration of hydrogen peroxide in addition to auto decomposition of  $H_2O_2$  to water and oxygen [9]. The rate of degradation of pollutants for both decolorization and COD reduction at every 30 min intervals for each  $H_2O_2$  dose utilized in the process is illustrated in Fig. 9. (a) and (b) respectively. Better performance efficiency achieved when a concentration of 75mM of  $H_2O_2$  has used in the process with 70%, 70%, 74.1%, 78.4% of decolorization and 48.8%, 61.6%, 64.8% and 68% of COD reduction at each 30 minutes time interval.

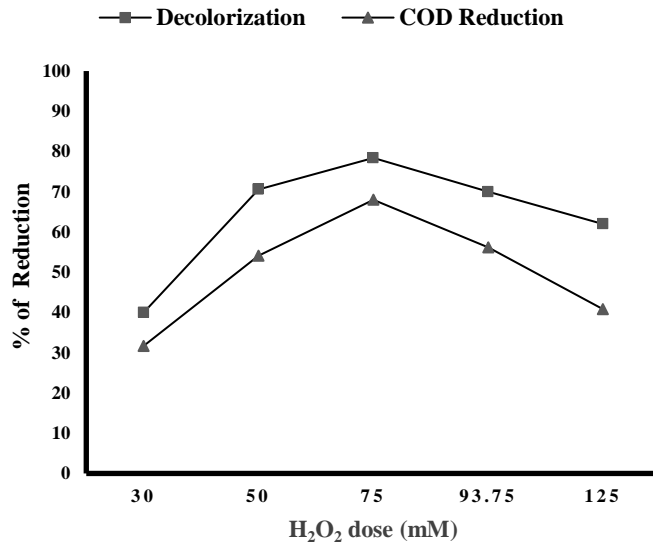


Fig. 8. Degradation efficiency and effect of H<sub>2</sub>O<sub>2</sub> Dose on Decolorization and COD Reduction in H<sub>2</sub>O<sub>2</sub>/UV process

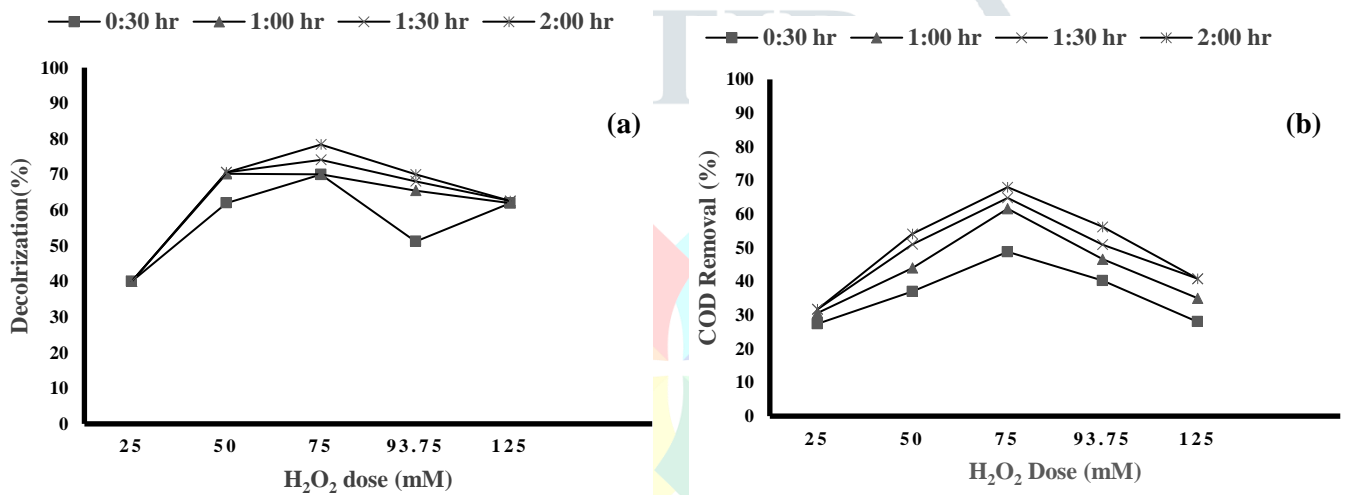


Fig. 9. (a) Rate of Decolorization and (b) Rate of COD reduction at every 30 minutes Reaction Time Interval at different Fe<sup>2+</sup> Dose, H<sub>2</sub>O<sub>2</sub>= 93.75Mm, pH=3, for 2hrs

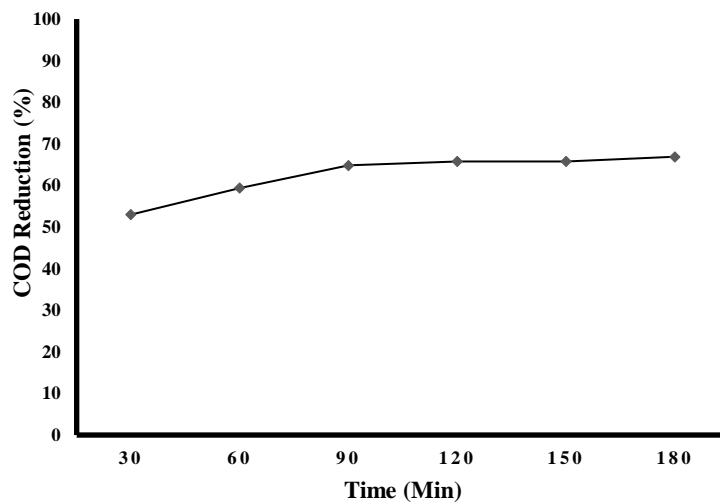


Fig. 10. Degradation efficiency and effect of reaction time in terms of COD Reduction in H<sub>2</sub>O<sub>2</sub>/UV process

### 3. Reaction Time optimization for H<sub>2</sub>O<sub>2</sub>/UV process

Reaction time has been optimized by increasing time of reaction to 3hrs (180min) and using all the optimized variable parameter. As it shown in Fig. 10 even though time has positive effect on the final performance in terms of total mineralization but increasing the reaction time from 120 minutes (2hrs) up to 180 minutes (3hrs) doesn't have change or significant deviation in the final COD reduction

efficiency with 67% of reduction compared to that of 120 min (2hrs) of reaction time performance efficiency with 68% of COD reduction.

### C. Comparison of Photo Fenton and H<sub>2</sub>O<sub>2</sub>/UV Process Based on Degradation Performance

According to the result of these two advance oxidation process at their optimum operating conditions in treating the textile wastewater comparisons has been carried out based on performance (decolorization and COD reduction) at every 30 minute reaction time and illustrated in Fig. 11 (a) and (b). As the result shows photo Fenton process has a beater rate of degradation and beater efficiency with 84.3%, 84.3%, 98%, 98% and 81.8%, 84%, 89.3, 93.6% degradation efficiency in decolorization and COD reduction respectively for every 30 minute reaction time interval. After 2hrs of reaction time photo Fenton process has better reduction efficiency compared to H<sub>2</sub>O<sub>2</sub>/UV with a deviation of 19.4 % in Decolorization and 25.6% in COD Reduction.

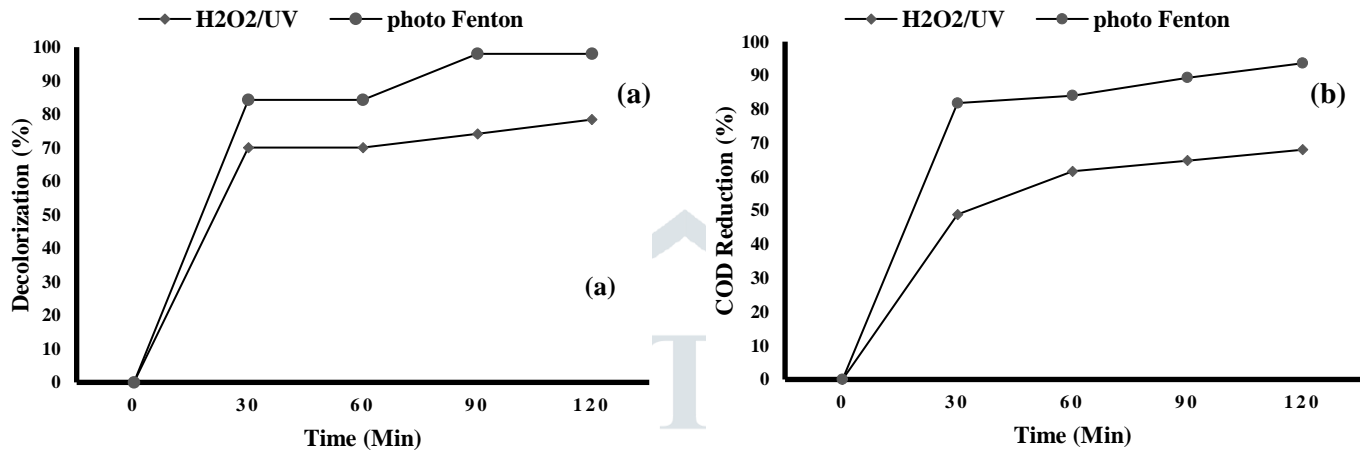


Fig. 11. Comparison of performance between H<sub>2</sub>O<sub>2</sub>/UV and photo Fenton process based on (a) decolorization (b) COD reduction

## VI. CONCLUSION

From this study it can be conclude that both photo Fenton and H<sub>2</sub>O<sub>2</sub>/UV process are effective in treating the textile wastewater. Independent variable parameters (pH, dose of H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup>, reaction time) influence both process's pollutant removal efficiency. Photo Fenton process has better pollutant removal efficiency with 98% in decolorization and 93.6% in COD reduction compared to that of H<sub>2</sub>O<sub>2</sub>/UV process with removal efficiency of 78.4% and 68% in decolorization and COD reduction respectively. Except for dose of H<sub>2</sub>O<sub>2</sub>, they share the same independent variable (reaction time = 2hrs, and pH=3) to achieve better performance. The optimum concentration of H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup> dose for photo Fenton process was 93.75mM/l and 8mM/l respectively. The optimum dose of H<sub>2</sub>O<sub>2</sub> in H<sub>2</sub>O<sub>2</sub> coupled with UV process was 75Mm/l. excess concentration of H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup> has negative effect on the process because they scavenge hydroxyl radical which ultimately affect the integrity of the processes performance.

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