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The treatment of azo dyes found in textile industry wastewater by anaerobic biological method and chemical oxidation

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ABSTRACT

The treatment of synthetic wastewater containing azo dyes found in textile industry wastewater was carried out by anaerobic biological method and chemical oxidation. The main target of this study was to compare different treatment methods and to evaluate the effect of different parameters on treatment effectiveness. In the microbial process, the results have shown that increasing the residence time, the amount of yeast extract and the addition of microorganisms originally growing on forest residues had positive effects on the dye removal. In the catalytic wet peroxide oxidation process, CWPO, the reaction conditions were optimized at 0.5 g/L activated carbon loading with 2 mL H_2O_2/300 mL solution (35 wt%), at 80 °C, in 2 h with pH = 3. At the optimum conditions, approximately 93% of the dye was removed. At these optimized conditions, the CWPO process was tested with real textile industry wastewater. The percentage of dye removal with this wastewater was 50%. The adsorption effect of the activated carbon was also investigated. At pH = 7, the removal by just adsorption was around 15%. But in acidic conditions (pH = 3) and at higher temperatures the adsorption effect of activated carbon increased. Adsorption and oxidation performances were compatible at 80 °C, however, at lower temperatures the adsorption effect was more considerable than the oxidation. It can be concluded that, generally the decolorization was accomplished by 60% adsorption and by 40% oxidation.

1. Introduction

The textile industry is known to be one of the major sources of water pollution. The quantity of wastewater from the textile industries has been increasing together with the growing demand for textile products. The wastewater contains organic dyes and their breakdown products which may have toxic and/or mutagenic effects on life. The release of textile industry wastewater into the environment can cause serious health and environmental problems [1].

Biological methods offer potential advantages over physical and chemical methods by being cheap and environmental friendly. In a biological treatment different microorganisms such as aerobic and anaerobic bacteria and fungi have been found to catalyze dye decolorization. Promising results were obtained by Dos Santos et al. in accelerating dye decolorization by adding mediating compounds and/or changing process conditions [2]. Generally the processes occurring in microbial ecosystems are governed by the action of many different microorganisms, all of which have a high degree of interaction with each other. It is believed that the interacting microbial population together can perform certain ecological functions better than each member of the population can do separately [3]. A consortium of mixed culture of microorganisms has several advantages for degrading compounds like those present in the textile effluents containing dyes. Normally, a consortium is more stable with changes in pH, temperature, and feeding composition when compared to pure cultures [4,5]. Also a consortium has more chances of complete mineralization of the dye present in the effluent as they work in co-metabolism style compared with a single strain [6]. Anaerobic microorganisms may be preferred for decolorization of azo dyes in textile wastewaters because of their properties [7]. For example, they generate electrons to cleave the azo bond [8].

The catalytic wet peroxide oxidation (CWPO) appears to be a potentially efficient process when compared with the other advanced oxidation processes, because the oxidizing properties of hydrogen peroxide are strong and a catalyst is used in this process. In the CWPO processes, the redox properties of dissolved transition metals (e.g., Fe, Cu, Ce) are used to generate hydroxyl radicals under mild reaction conditions in the presence of hydrogen peroxide. Although CWPO has been widely used for the oxidation of wastewater streams with high organic (TOC) content, the utility of this system is limited because of the restricted viable pH range and the need for the recovery of the homogeneous catalyst.

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Nomenclature

CWPO	catalytic wet peroxide oxidation
COD	chemical oxygen demand
C_0	initial absorbance value
C_f	final absorbance value
D	diameter
h	height
pH_{pzc}	point of zero charge
TOC	total organic carbon
λ_{max}	absorbance peak point

The use of a heterogeneous catalyst overcomes these drawbacks [9].

Activated carbons have proved their catalytic and adsorbent activity in the oxidation of organic pollutants [9–12]. Activated carbons can act as an adsorbent for the organic compounds and also as a catalyst for their oxidation. The generation of the active oxidizing species on the carbon surface can potentially increase the process efficiency [12]. The surface chemistry of carbon materials is basically determined by the acidic and basic character of their surface, and can be changed by treating them with oxidizing agents either in the gas phase or in a solution. These treatments fix a certain amount of functional groups occurring on the surface chemistry of activated carbon complexes such as: carboxyls, lactones, phenols, ketones, quinones, alcohols, and ethers that make the carbon materials more hydrophilic and acidic, decreasing the pH of their point of zero charge and increasing their surface charge density [13].

In the present work, typical reactive dyes found in the textile industry wastewater were selected and treatment of them by both anaerobic biological method and chemical oxidation method was evaluated. An innovative aspect of the study is to show the availability of two alternative treatment processes and to compare them. In addition to the main innovative aspect, different from the present studies in literature, in the biological method, as a nitrogen source yeast extract and as carbon source rice husks were used. In the chemical method, the CWPO process was applied in the presence of activated carbon to propose a treatment technique in which adsorption and catalysis is realized in a single step, taking advantage of the performance of activated carbon as adsorbent and catalyst.

2. Experimental

2.1. Experimental set-up

2.1.1. Microbial process

First a batch process was performed to determine what substances were released from the rice husks, if any. This experiment was performed in 250 mL Erlenmayer flasks and two parallel continuous systems were used. Both systems consist mainly of cylindrical plexiglass reactors and pumps. In the first continuous system, two or four reactors (1st and 2nd reactors have 450 mL volume, 3rd and 4th reactors have 430 mL volume) connected in series were used and in the second system just two serial connected reactors (each reactor has 450 mL volume) were used.

2.1.2. Catalytic wet peroxide oxidation

The experimental set-up shown in Fig. 1 mainly consists of a reactor, a condenser and suitable measuring devices for temperature, pressure, and air flow rate. The main part of the system was a 500 mL (h = 500 mm; D = 35 mm) stainless steel reactor equipped with an electrical wires wrapped around, a temperature controller (PID-1/32 DIN model), and a pressure indicator. The condenser mounted at the top of the reactor was used to condense the vapors formed during the reaction and to send the condensate back to the reactor. The experimental setup has been explained elsewhere in detail [14].

2.2. Experimental procedure

2.2.1. Microbial process

The general experimental procedure followed can be summarized as follows: in the batch experiment 8 g of rice husks and 250 mL of 0.9% NaCl solution are mixed in 250 mL Erlenmayer flasks under anaerobic conditions. Samples are taken from the flasks every two days and then they are scanned with Perkin Elmer Lambda 35 UV/VIS spectrophotometer. In the continuous systems the reactors are filled with rice husks. The connection tubes between the reactors and feed bottles are washed with ethanol for

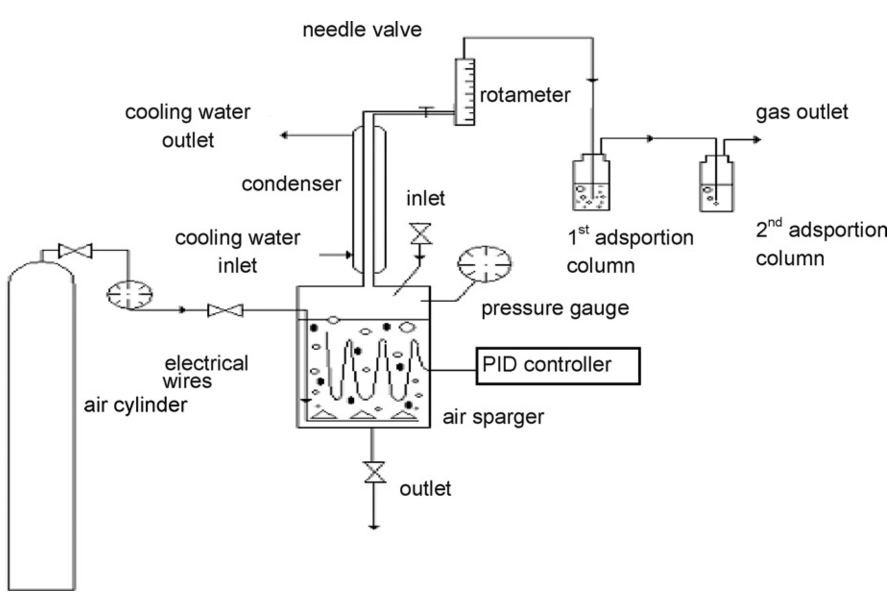
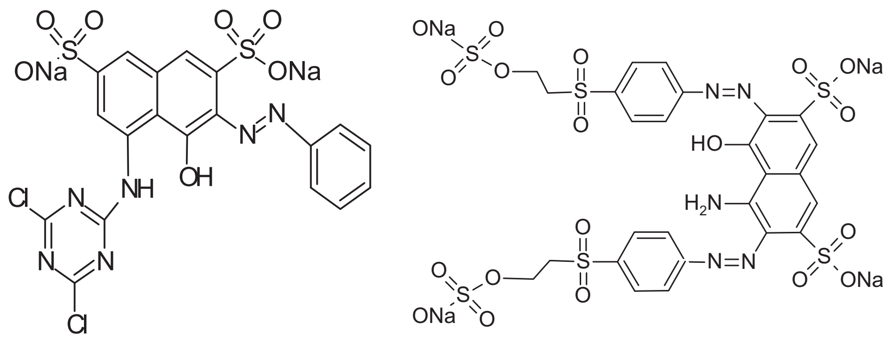


Fig. 1. Schematic view of the experimental set-up of wet peroxide oxidation process.



Procion Red MX-5B

Reactive Black 5

Fig. 2. The structures of the dyes.

sterilization. A desired synthetic dye mixture is prepared by using tap water and then put into an autoclave at 121 °C for 15 min for sterilization to ensure that there are not any microorganisms in the synthetic dye solution. The speed of the pump (C6-MIDI multi-channel pump, Watson Marlow SCI) is changed to provide a flow rate between 0.351 mL/min and 0.981 mL/min in order to maintain the required residence time. To determine the dye removal efficiency of the microorganisms, samples are taken every two days from each reactor and also from the orginal dye mixture and then analyzed.

2.2.2. Catalytic wet peroxide oxidation

For a typical run, synthetic wastewater which contains Reactive Black 5 and Procion Red MX-5B is prepared with distilled water. The required amount of hydrogen peroxide and activated carbon is added and the pH of the wastewater is adjusted to the desired value. The prepared wastewater is charged to the reactor and cooling water is supplied to the process. The mixture is heated to the desired temperature. When the reactor reaches the desired temperature, the air flowrate is initiated and the reactor is pressurized. The desired air flow rate is adjusted with the help of a needle valve. During the experiment temperature, pressure and air flow rate are recorded at every 10 min. At the end of the experiment, a sample from the mixture is taken for analyzing the dye concentration.

2.3. Properties of wastewater

2.3.1. Microbial process

Two different kinds of synthetic wastewater compositions were used. The first synthetic dye solution (ww I) was prepared as a mixture of Reactive Black 5 and Procion Red MX-5B (Fig. 2) at equal concentrations of 200 mg/L each. In addition 1 g/L yeast extract was also added.

The second synthetic dye solution (ww II) which is more similar to real textile industry wastewater was used [2]. The composition of the solution was 12 g NaCl, 0.049 g $Na_3PO_4 \cdot 12H_2O$, 0.024 g $NaNO_3$, 0.255 g $KHSO_4$, 200 mg Reactive Black 5 dye, 200 mg Procion Red MX 5B, the soap and the washing solution from cotton, and 1 g of yeast for 1 L solution.

The both synthetic dye solutions were prepared at a pH around 7.

2.3.2. Catalytic wet peroxide oxidation

In order to have comparative experimental results, the CWPO experiments were performed using the same synthetic dye solution as used in the microbial process which was a mixture of Reactive Black 5 and Procion Red MX-5B at equal concentrations of 200 mg/L each at a pH around 7. In addition to this, a real textile industry wastewater was also tested using the CWPO process. Some properties of the original industrial wastewater were pH = 10, COD = 7250 mg/L.

2.4. Experiments

2.4.1. Microbial process

As a preliminary study, batch experiment was performed for background information to see if any substances were released from the rice husks that could interfere with the spectrofotometric analyses. The retention time was higher in the batch system when compared to the continuous systems and hence it was observed more clearly that the microorganisms were capable of degrading the compounds which might be released.

In the two parallel continuous systems the experiments were performed to investigate the effect of the following parameters explained below.

2.4.1.1. Continuous system 1. In this system effect of the wastewater composition, the residence time, the amount of yeast extract, and the wastewater flow rate was tested. In order to investigate the effect of wastewater composition two different synthetic wastewaters were prepared as explained in Section 2.3.1. The flowrate of the synthetic wastewater was adjusted as 0.351 mL/min. The experiments were lasted until the system reached the steady state conditions. In the experimental set in which the residence time effect on the performance was investigated, the number of reactors was increased from two to four to ensure a longer residence time and the feed flow rate was 0.351 mL/min. The system was also inoculated with a rinse solution from the forest residues (50.5 g wood chips of forest residues rinsed with 400 mL 0.9% NaCl) in order to add new microorganisms which had different capability for degradation of organic compounds. In order to investigate the effect of the yeast extract addition, the amount of yeast extract was increased from 1 g/L to 2 g/L in the synthetic dye mixture. The effect of the wastewater flow rate was also tested by increasing the rate from 0.351 mL/min to 0.803 mL/min.

2.4.1.2. Continuous system 2. Parallel to the experiments in continuous system 1, other experiments in continuous system 2 were performed to investigate addition of solution from forest residues and the effect of wastewater flowrate. The system had two reactors connected in series. The system was inoculated with a rinse solution from the forest residue so different kinds of microorganisms were added to the system in order to investigate the effect of the wood chips solution. The purpose was to add different kind of bacteria and fungi, which support themselves on complex organic

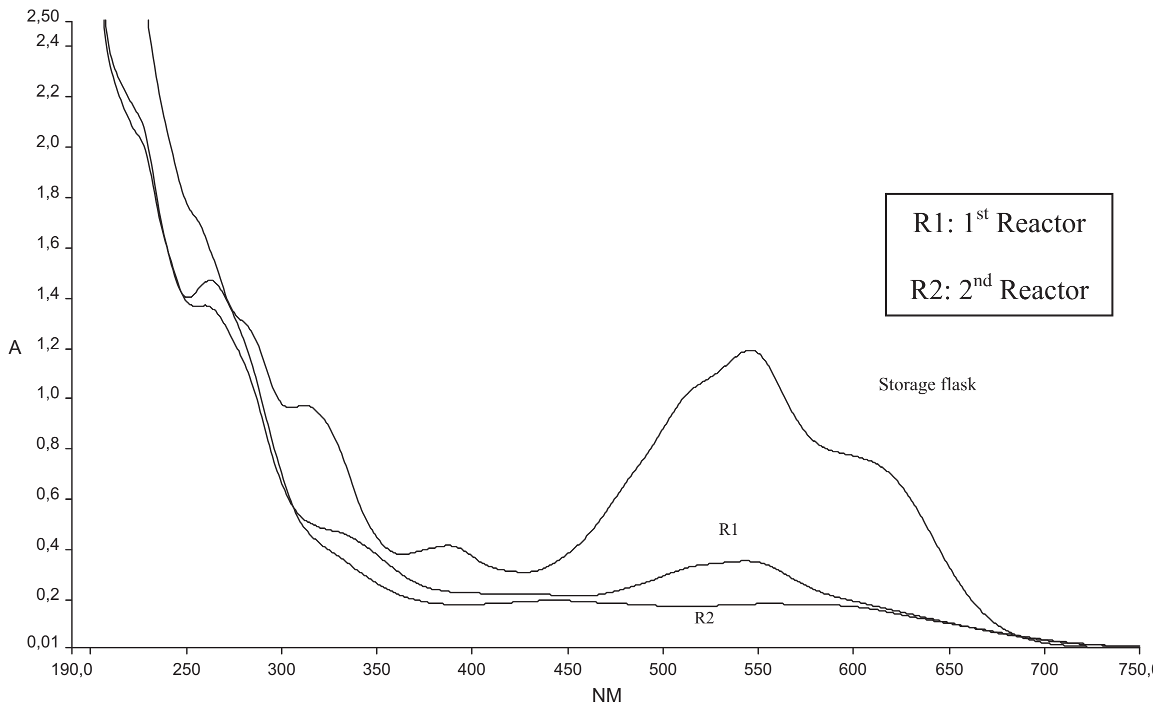


Fig. 3. Absorbance data of the dye wastewater and the outlets of the reactors in the 21st day of the process after start-up of continuous system 1.

polymers and aromatic molecules. The flow rate of the wastewater (ww II) was 0.311 mL/min.

Finally, the flow rate of wastewater was increased from 0.311 mL/min to 0.637 mL/min to investigate the effect of increasing the flow rate and hence residence time.

2.4.2. Catalytic wet peroxide oxidation

The second section of the study was organized in five sets to determine the appropriate conditions for the removal of azo dyes by CWPO. In each set, all the experiments were performed using 300 mL solution of a mixture of 200 mg/L of Procion Red MX-5B and 200 mg/L of Reactive Black 5 at atmospheric pressure, with a continuous air flow rate of 0.23 L/min.

In the first set, the effect of the activated carbon loading was investigated with addition of different concentrations (0.5 g/L; 1 g/L; 1.5 g/L and 2 g/L) with a pH = 7, at a temperature of 50 °C and addition of 2 mL of H_2O_2 for 300 mL solution. In the second set, the effect of hydrogen peroxide loading was studied by varying the amount of H_2O_2 (2, 4, 8 mL) for 300 mL solution with a pH = 7, at 50 °C and 0.5 g/L activated carbon loading. In order to investigate the effect of temperature, in the third set the experiments were performed at four different temperatures (50, 60, 80 and 90 °C), operating conditions were pH = 7, 0.5 g/L activated carbon loading and 2 mL H_2O_2 for 300 mL solution. In the third set, in order to determine the optimum pH value, different pH values (7–2) were tested with 0.5 g/L activated carbon loading and 2 mL H_2O_2 at 50 °C for 300 mL solution. Finally, in the last set, to find the optimum reaction time several experiments were performed for different experiment times: 5, 10, 15, 30, 45, 60, 90, 120, 150, 180 min with 0.5 g/L activated carbon loading and 2 mL of H_2O_2 at 50 °C.

In order to investigate the adsorption effects of the activated carbon four different trials were performed in a three necked balloon with the dye solution (200 mg/L Reactive Black 5 and 200 mg/L Procion Red MX-5B) and 0.5 g/L activated carbon. The adsorption effect of the activated carbon was tested under different pH values (pH = 3 and pH = 7) at 50 °C. And in addition to this, at pH = 3 the reaction temperature was raised to the value of the optimized conditions (80 °C). And all the results of these trials were compared with the CWPO at the optimized conditions.

2.5. Product analysis

2.5.1. Microbial process

The samples were scanned with a Perking Elmer Lambda 35 UV/VIS spectrophotometer, Perkin Elmer Lambda 35 UV/VIS, and analyzed by Perkin Elmer UV winlab ver: 2.85.04 software between 90 nm and 750 nm wavelengths, the λ_{max} of Reactive Black 5 is 597 nm and, the λ_{max} is 538 nm for Procion Red MX-5B [15,16]. The λ_{max} is 550 nm for the mixture of these dyes. Because of this reason, the absorbance data was investigated at around 550 nm. The dye removal efficiencies were calculated as shown below:

$$X\ (\%) = \frac{C_0 - C_f}{C_0} \times 100 \quad (1)$$

where C_0 and C_f are the initial and final absorbance values of the dye.

2.5.2. Catalytic wet peroxide oxidation

Removal of dye was analyzed spectrophotometricaly in a UV/VIS spectrophotometer at 550 nm (NOVA 400 Merck UV-spectroquant). For evaluating the performance of the reactions, the dye removal efficiencies were calculated by Eq. (1).

3. Results and discussion

3.1. Microbial process

The experiments in continuous systems are not performed in replicates, but each experiment was run for at least three weeks.

3.1.1. Continuous system 1

The percentage of dye removal in the case of using the synthetic dye solution which contains just the two different azo dyes (each dye had a concentration of 200 mg/L) and yeast extract (1 g/L) was around 78.8% at 0.351 mL/min feed flow rate. An example for the absorbance data of the dye wastewater and the outlets of the reactors at the 21st day of the process after start-up of continuous system 1 can be seen in Fig. 3.

When changed to the more realistic wastewater composition (ww II) the dye removal percentage decreased to 29.5%. This might be explained by the negative effect of the washing up detergent, fats, and salts on the activity of the microorganisms.

In order to improve the performance of the system with wastewater composition which is more similar to real wastewater, the numbers of reactors were increased from two to four which increased the residence time approximately twice. Feed flowrate was 0.351 mL/min. The system was also inoculated with rinse solution from forest residues (50.5 g wood chips of forest residues rinsed with 400 mL 0.9% NaCl) in order to add new microorganisms which had different capability for degradation of organic compounds.

It can be concluded that the percent of dye removal increased to 43% in the outlet of the second reactor after microbes from forest residues were inoculated, when compared to before it was only 29.5% before. The increase in the number of reactors also had a positive effect on the dye removal; after passing all four reactors the dye removal obtained was 78.6%.

Increasing the amount of yeast extract in wastewater composition affected the activity of the microorganisms considerably. The dye removal increased from 78.6% to 89.2%. Yeast extract is a nitrogen source for microorganisms so increasing the amount of yeast extract might facilitate the growing of microorganisms in the system and hence the dye removal increased.

The percent of dye removal obtained was 82.7%. The dye removal decreased from 89.2% to 82.7% after increasing the flowrate more than double. It might be concluded that the microorganisms adapted with increased capacity to the increased flowrate.

3.1.2. Continuous system 2

The percent for the same wastewater composition 85.4% decolorization was obtained in system 2 after adding the forest residue solution, compared to 78.8% in system 1 earlier. Inoculating the system with a forest residue solution increased the dye removal capacity for the system.

There was an 82.8% removal after increasing the flowrate whereas 85.4% of dye removal had been obtained before the flowrate increase. The flowrate was increased approximately two fold but the difference between 85.4% and 82.8% was not big in proportion to flowrate change. Although increasing the flowrate two fold, dye removal decreased only 3%. It can be concluded that the biofilters can manage fluctuations as increased flow leading to reduced residence time with only a slight decrease in decolorization efficiency.

The results of the microbial process are given in Table 1.

Table 1
Results of the Experiments in the Microbial Process.

Experiments	Changing parameters	Results (% dye removal)
Continuous system I	Synthetic dye solution	78.8
	New wastewater composition	29.5
	Increasing the residence time	78.6
	Increasing the amount of yeast extract in wastewater	89.2
	Increasing the wastewater flowrate	82.7
Continuous system II	Washing the system with forest residues solution	85.4
	Increasing the wastewater flowrate	82.8

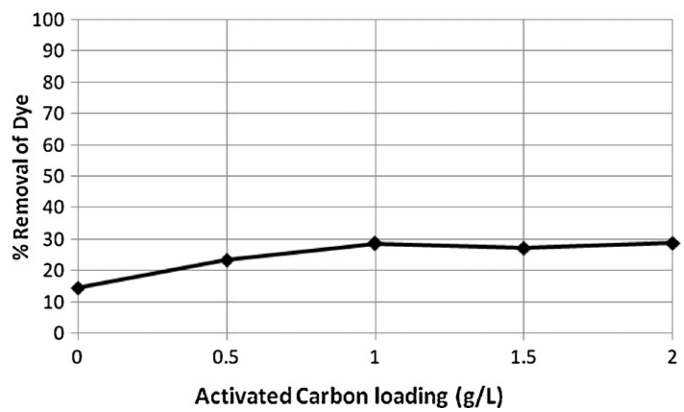


Fig. 4. The effect of the activated carbon loading on dye removal.

3.2. Catalytic wet peroxide oxidation

In most cases the results gained obey the experimental theory. However in the very few cases when the replicates were inevitable, the experiments were repeated.

3.2.1. Determination of optimum operating parameters

Generally, it can be concluded that when the amount of activated carbon was increased the dye removal also increased. For example, when there was no activated carbon, the % removal of dye was 14.5% but after adding 0.5 g/L activated carbon, the % removal of dye increased to 23.5%. At the higher loading values, the effect of activated carbon was negligible. Fig. 4 shows the results gained. When the activated carbon loading increased from 0.5 g/L to 2 g/L, the removal of dye increased from 23% to 28%. Therefore when the results were evaluated, 0.5 g/L of activated carbon loading seemed to be an optimum value.

With an increase of the H_2O_2 loading in the process, the removal in dye of the system increased. When the amount of H_2O_2 increased from 2 mL to 4 mL, the removal of dye increased (from 23% to 27%) very slowly. When the hydrogen peroxide dosage exceeds 4 mL, the removal percentage decreased as seen in Fig. 5. One possible reason for this small decrease is explained in the study performed by Quintanilla et al. Above all sites of the activated carbon charged in the solution are already occupied and so the formation of hydroxyl radicals on the surface of the activated carbon is stopped. Moreover,

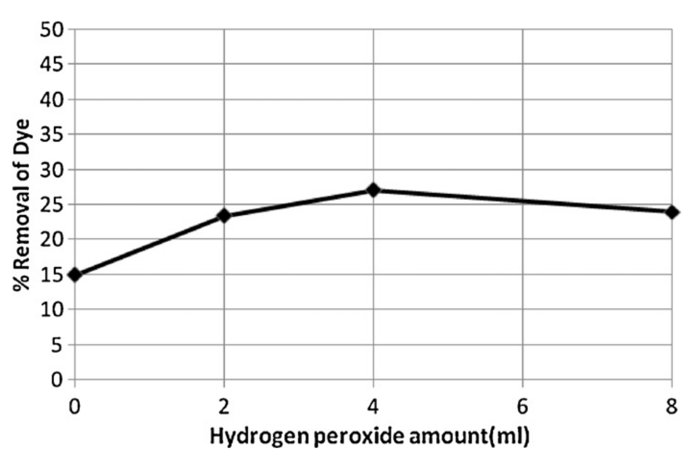


Fig. 5. The effect of H_2O_2 loading on dye removal.

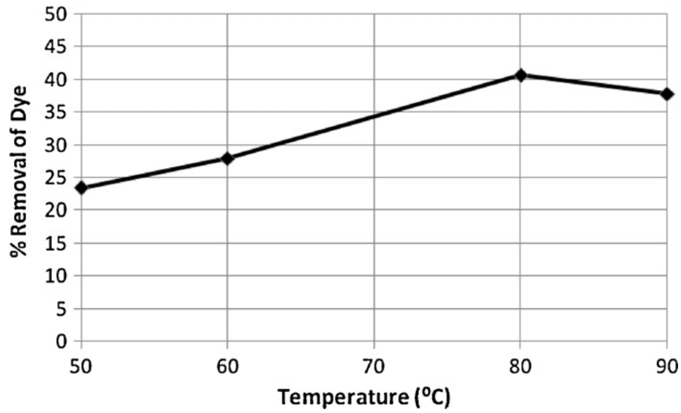


Fig. 6. The effect of temperature on dye removal.

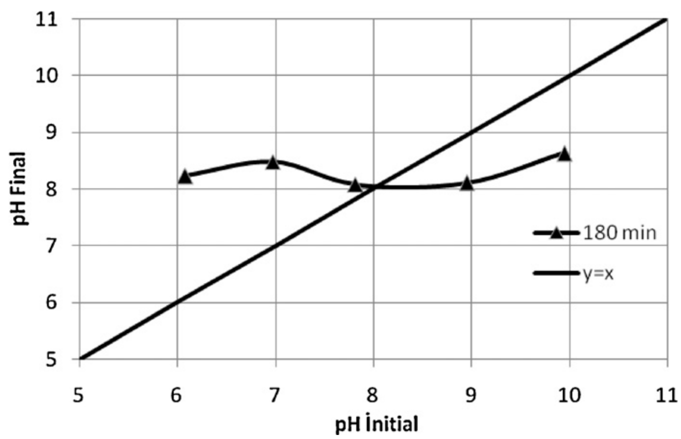


Fig. 7. The point of zero charge of pH (pH_{pzc}) of activated carbon.

a competitive reaction between hydrogen peroxide in excess and already yielded hydroxyl radicals could be possible. In that case, using less hydroxyl radicals in the oxidation reactions of azo dyes is probable [11]. When the results were evaluated and when the economics of the process are considered, 2 mL H_2O_2 in 300 mL solution seemed as an optimum value. Because at values higher 4 mL of H_2O_2, excess hydrogen peroxide goes in the reaction with the produced hydroxyl radicals and hence less hydroxyl radicals are present in the system.

Fig. 6 shows that the removal of dye incresed from 23% to 40% as a consequence of increasing the temperature from 50 °C to 80 °C. This is because higher temperature increased the reaction rate between H_2O_2 and the catalyst, thus increasing the rate of formation hydroxyl radicals. But increasing the temperature from 80 °C to 90 °C decreased the removal. The reason to obtain lower removals is that at higher temperatures the thermal decomposition of H_2O_2 resulted in the reduction of its effective concentration towards making hydroxyl radicals [17].

The pH value should be lower than the point of zero charge, pH_{pzc}, of the activated carbon in order to maximize adsorption [18,19]. Because of this reason, pH_{pzc} of activated carbon used in the experiments was investigated and it was founded at a pH = 8. The graph for determination of pH_{pzc} of activated carbon is given in Fig. 7. So the experiments were performed at a lower pH than pH_{pzc}. Generally, the dye removal increased from neutral to strong acidic conditions (the removal 23% at pH = 7 and 56% at pH = 2). Fig. 8 shows the results of this set. The best removal rate was obtained at a pH = 2, but pH = 3 was chosen as the optimum value when it was evaluated regarding corrosion and environmental conditions.

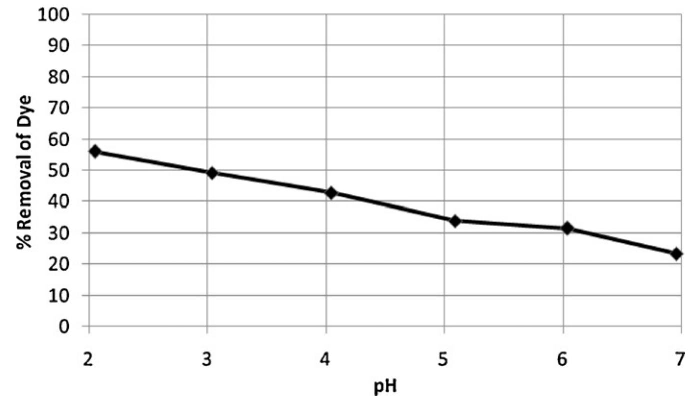


Fig. 8. The effect of pH on dye removal.

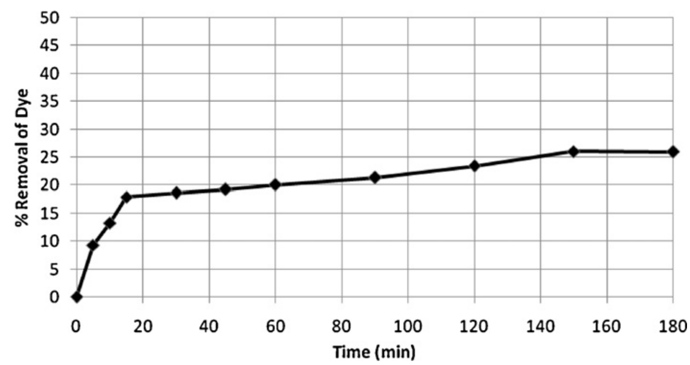


Fig. 9. The effect of reaction time on dye removal.

Al-Degs et al. reported that, the adsorption capacity of the activated carbons increased in acidic conditions, but decreased in the basic solutions. These results can be explained also by the oxidation properties of hydrogen peroxide and hydroxyl radicals that depend on the pH of the solution [20].

Generally, the dye removal has increased with increasing the reaction time (Fig. 9). The removal of dye incresed rapidly for the first 15 min and increased slightly between 15 min and 150 min. After 150 min, the effect of the reaction time was not negligible.

3.2.2. Experiments performed at optimum conditions at different reaction times

In the study, as the next step, the experiments were performed under the optimized conditions for the CWPO of the mixture of Reactive Black 5 and Procion Red MX-5B. These conditions were

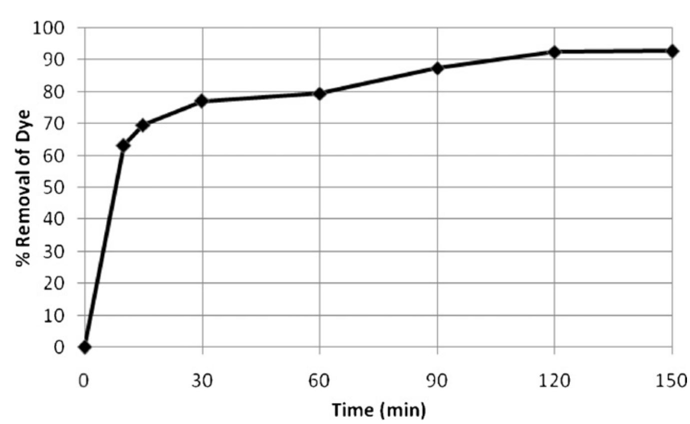


Fig. 10. The effect of CWPO process at the optimum conditions on dye removal.

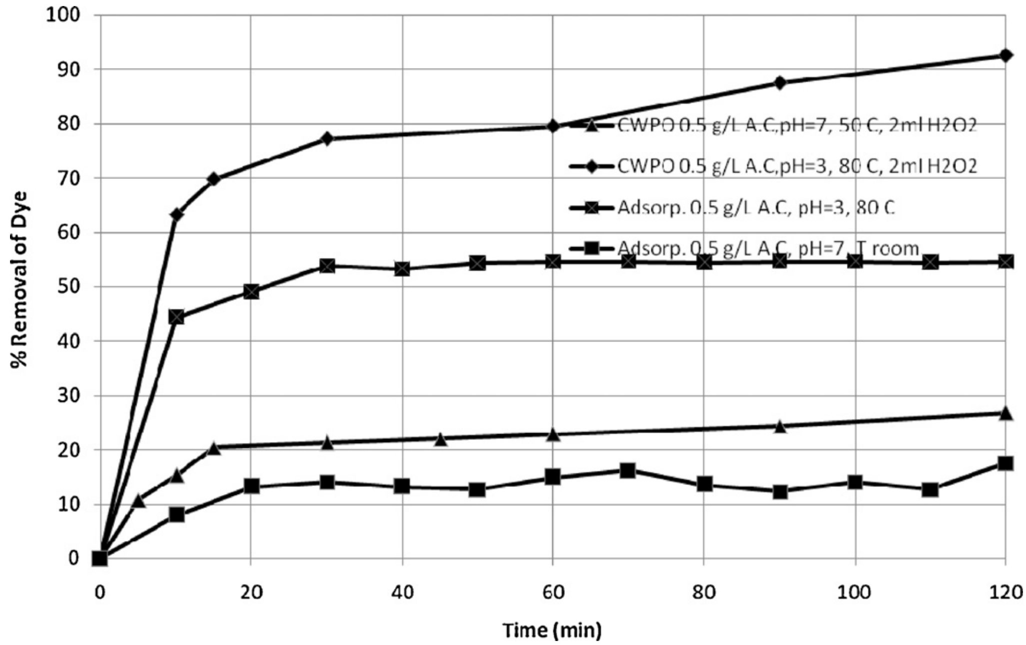


Fig. 11. Comparison between CWPO process and adsorption experiments.

0.5 g/L activated carbon loading, 2 mL H_2O_2/300 mL dye solution, a pH = 3 and T = 80 °C. The reaction time was 120 min. Under these conditions, 93% dye removal was achieved showing that CWPO can be recognised as an attractive wastewater techonology for removal of azo dyes. In Fig. 10, the results of the experiments performed are plotted.

3.2.3. Application of the CWPO process to the textile industry wastewater at the optimized conditions

As one of the main goals of this study, the optimized conditions determined were tested on wastewater which was supplied from a textile dyeing factory in Bursa in Turkey. The CWPO of this wastewater was performed at the optimized conditions. At these conditions, 46.0% of dye removal and 22.4% of COD removal was achieved, respectively. In the same conditions, the H_2O_2 amount was doubled to investigate if the H_2O_2 amount had a considerable effect. The dye removal increased just to 50.06% at the end of 2 h. Then, all the other conditions were kept constant except for the activated carbon loading, and it was doubled. Again the removal (49.30%) did not change considerably. The result obtained is not as much as expected. Because at these optimum conditions with using the prepared synthetic wastewater, the dye removal obtained was approximately 90% whereas with the textile industry wastewater, the removal obtained was around 50%.

3.2.4. Adsorption tests on activated carbon

The results gained on adsorption test are plotted in Fig. 11. When the results are investigated, it can be concluded that adsorption on activated carbon presents a low performance for dye removal at the higher pH (pH = 7). The dye removal based on just adsorption increased when the pH is decreased from pH = 7 to pH = 3. The adsorption effect of activated carbon increased in the acidic conditions and also at a higher temperature. Orfão et al., reported that, adsorption performance is highly affected by the pH of the solution, since it influences the surface charge of the activated carbons and consequently the intensity of the electrostatic interactions between dye molecules and the adsorbents surface [18]. It can be concluded that at higher temperatures the adsorption effect of activated carbon and the effect of oxidation on the removal were compatible. But at lower temperatures the adsorption effect of activated carbon was more effective than oxidation.

4. Conclusions

In this article, an investigation of a comparative study of anaerobic biological method and CWPO in dye removal of textile industry wastewaters was reported.

The main objective of the study was to show the availability of two alternative treatment processes and to compare them by evaluating the effect of different parameters on treatment effectiveness.

In the microbial process, parameters such as the wastewater composition, the residence time, the amount of yeast extract, wastewater flowrate (residence time), and the addition of microorganisms from wood chips were investigated. The highest removal obtained by the microbial process was approximately 89%.

In the CWPO process, the effects of activated carbon loading, H_2O_2 loading, pH, temperature and reaction time were investigated. At the optimized reaction conditions approximately 93% of the dye was removed. As a final step, the optimized conditions were tested with a real textile industry wastewater supplied from a factory located in Bursa. The percentage dye removal of this wastewater was 50%.

If both the processes are compared, it can be concluded that both have some advantages and disadvantages. The microbial process is an environmental friendly process but the treatment must last for a longer time to ensure the microorganisms adaptation. In the chemical oxidation process, the reaction time is less; however it should be studied under an acidic medium and by adding some oxidizing agents. This is limited by investment and operating costs. Consequently, the conditions should first be evaluated before deciding on which process that will be used.

It can be concluded that the microbial process and CWPO process are promising technologies which might be used to treat aqueous solutions containing azo dyes with good performance. The results would expedite the application of these methods to the treatment of wastewaters of textile industry.

Acknowledgements

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