



Removal of Reactive Dye Types (Methylene Blue, Direct Blue 15 and Reactive Black 5) from Water with Different Methods

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HIGHLIGHTS

- > The UV effect on the different methods used for the removal of reactive dyes has been demonstrated with the designed experimental system.
- > It was found that UV combined Fenton and biosorption methods gave better dye removal results for all dyes tested.

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ABSTRACT

Synthetic paints attract a lot of attention due to the width of their usage areas and the amount of excessive use. Particularly, the damage caused to the water by the part remaining from the paint used and disposed of in the environment is visible from time to time. Removal of synthetic dyes used from these waters is an important research topic. In our research, we planned to investigate the removals comparatively by using the biosorbent we developed in different combinations. Therefore, chemo-biosorbent was obtained by modifying it with biosorbent and nano-iron produced from sunflower and chitosan, and a reaction medium was created using Methylene Blue (MB), Direct Blue 15 (DB) and Reactive Black 5 (RB) dyes. In addition, dye removals of chemosorption, biosorption, Reactions with UV combined methods under the same optimum conditions were compared to find out which removal was most appropriate. As a result of the reaction, it was observed that UV application contributed significantly to the experimental conditions. It was observed that UV increased the dye removal at varying rates in the range of 0.27-4.42% biosorption and Fenton reactions. Accordingly, it was understood that UV did not have a positive contribution in the chemo-biosorption reaction, which we performed using nano-iron in its structure. Considering the cost evaluation, such studies will contribute to the selection of the preferred method for dye removal.

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1. Introduction

Many industries around the world use synthetic dyes. It is stated that these are more than 8×10^5 tons per year. In addition, it has been reported that more than half of these dyes enter the waters as waste. These mixed dyes cause both temporary and permanent damage to the ecosystem. Although some of these damages can be seen as economic losses, more serious damages that cannot be resolved in a long time are factors affecting living things such as water and soil pollution [1]. For these reasons, the removal of waste dyes from water has become a very important research topic. In addition, many organic pollutants need to be removed from soil, water and air, and basically, the same principles can be used to remove such pollutants [2].

Although there are different classifications of the methods in the removal of synthetic or natural pollutants in water, it has been observed that they can be grouped into 4 main groups as follows. Electrochemical, Fenton reaction, oxidation techniques containing peroxide, physicochemical methods including adsorption, ion exchange, coagulation, membrane method using different filtration techniques and biological techniques using enzymes and similar [3].

By using one or more of these techniques, it is desired to obtain the least damage to the environment from industrial wastewater. For this reason, the contribution of the methods used in dye removal is very important. During the removal of synthetic dyes from wastewater with UV, which is one of the electrochemical methods, radicals are formed in the reaction vessel with the effect of UV and these radicals occur when the synthetic dye is destroyed. Only UV is not an adequate and suitable method for removing paint from water. However, it is suitable for use as a combined method of removing small amounts of dye from wastewater.

Fenton reaction is an important method that is widely preferred in synthetic dye removal processes. Fenton, ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$) and Fenton-like ($\text{H}_2\text{O}_2/\text{Fe}^{3+}$) methods can be used for this purpose. If UV / visible irradiation ($\lambda < 600$ nm) is added to this method, the method is called the photo Fenton method and the method is quite advanced. The reason for this is the desire to prevent the decrease in the rate of the Fenton reaction by the complete depletion of Fe^{2+} during the Fenton reaction and its conversion to Fe^{3+} . Thanks to UV, Fe^{3+} ions are regenerated to Fe^{2+} and thus the continuity of the Fenton reaction is ensured [4, 5].

In this research, which we aimed to do, firstly, it was planned to prepare the reaction medium in which chitosan and sunflower waste as biosorption agent, chitosan sunflower modified with nano-iron synthesized by green synthesis method for chemisorption, and H_2O_2 , an oxidation agent, were added for Fenton process. And, to compare these reactions, it was planned to compare the results obtained from the methods by determining the effect on dye removal by using a UV light source.

2. Materials and Method

2.1. Chemicals and Dye Removal

The materials used for biosorption, chemisorption and Fenton reaction were synthesized as previously mentioned in

Turgut [6]. For this purpose, after the sunflower was obtained in September, it was used by sorting, drying and shredding. After the nano-iron used in the chemisorption environment was synthesized using the green synthesis method and the method of Nadaroğlu et al [7–9], chitosan was combined with sunflower material and modified. The obtained material was used for chemisorption, and then the Fenton reaction was carried out under the same conditions with the addition of H_2O_2 . Then, to examine the effect of UV on these reaction environments, the reaction was carried out under optimum conditions by installing a UV lamp and temperature-controlled reaction mixtures in a closed environment designed under the same conditions.

The chemicals used in the study were Chitosan (Sigma/Aldrich 75% purity), Acetic acid (CH_3COOH -Sigma/Aldrich 100%), Sodium hydroxide (NaOH-Merck 99-100%), Hydrochloric acid (Merck-HCl 37%), Hydrogen peroxide (H_2O_2 ; 35%).

The dyes used for dye solutions in the experimental stage are Reactive Black 5 (Sigma/Aldrich $\geq 50\%$), Direct Blue 15, (Sigma/Aldrich $\geq 40\%$) and Methylene Blue (MB, $\geq 40\%$), Iron (III) chloride (FeCl_3 ; Sigma/Aldrich 97%) and Sodium phosphate (Na_3PO_4 ; Merck $\geq 99\%$).

In order to understand how much of the dyes were removed from the reaction medium, the equation (1) given below was used. The % dye removal was calculated separately for all dyes at wavelengths that gave optimum absorbance, using the Equation 1.

C_o : Dye absorption

C_t : Result dye adsorption

$$\% \text{Dye removing} = \frac{C_o - C_t}{C_o} \quad (1)$$

2.2. Characterization

Characterization and identification of biosorption, chemisorption and Fenton reagents prepared for dye removal using different techniques (UV-VIS-NIR (Shimadzu UV-3600 Plus), SEM (Scanning Electron Microscope) (Zeiss brand), XRD (X-ray powder diffraction) (Panalytical Empyrean brand) and FT-IR (Fourier Transform Infrared) [6].

3. Results and Discussion

3.1. Investigation of the kinetics of MB, DB and RB dyes removal

Synthetic dye removal from wastewater, which is still an important research area today, is a very interesting research topic. Numerous publications are made each year on this research topic. As can be seen from these studies, it is aimed to achieve optimization by using different methods for this purpose. In our research, 3 different methods were tried on its removal from water by using 3 different synthetic dyes.

In addition, the reactions were repeated under the same conditions in a 30 Watt UV environment in order to have information about how the results would change if the UV effect was combined with these methods. The studies were

carried out in the UV reactor system designed by us in Figure 1.

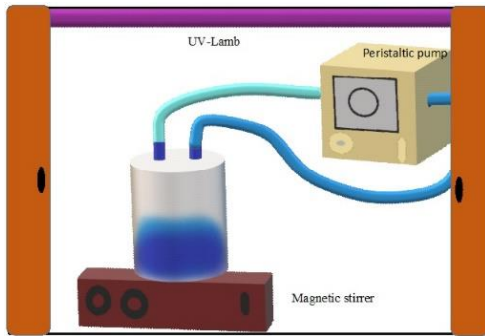


Figure 1 Reactor assembly used for the UV effect

Experimental environments were created to compare the 3 different dye removals we combined with this setup. Priya, K, and Palanivelu 2005 stated in their research that they achieved 95% paint removal in a very short time by using UV and Cl^- ion sources [3].

Nguyen et al., on the other hand, reported that they could remove 44.2-99.6% of different synthetic dyes in their research using UV and photo-catalyst [10, 11].

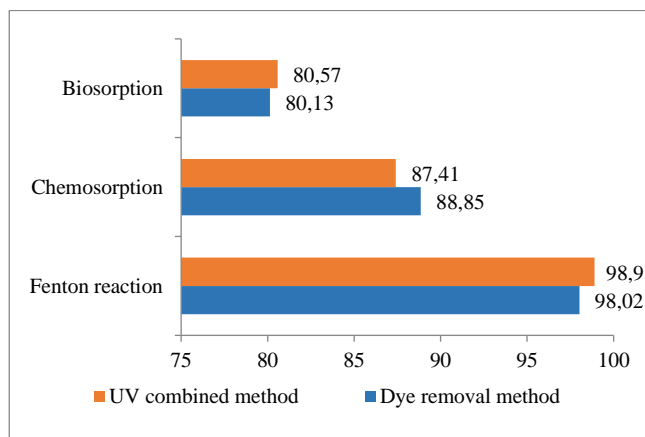


Figure 2 Optimum conditions in the Methylene Blue removal reaction (Time: 60 min., pH: 5, Temperature: 40 °C, Dye Concentration: 25 mg/L, Amount of Substance: 25 mg, Amount of H_2O_2 for Fenton Process: 600 μL)

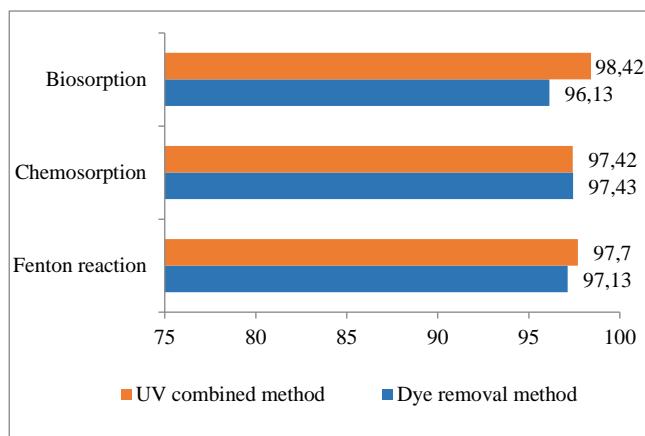


Figure 3 Optimum conditions in the Direct Blue 15 removal reaction (Time: 30 min., pH: 2, Temperature: 25 °C, Dye Concentration: 25 mg/L, Amount of Substance: 50 mg, Amount of H_2O_2 for Fenton Process: 900 μL)

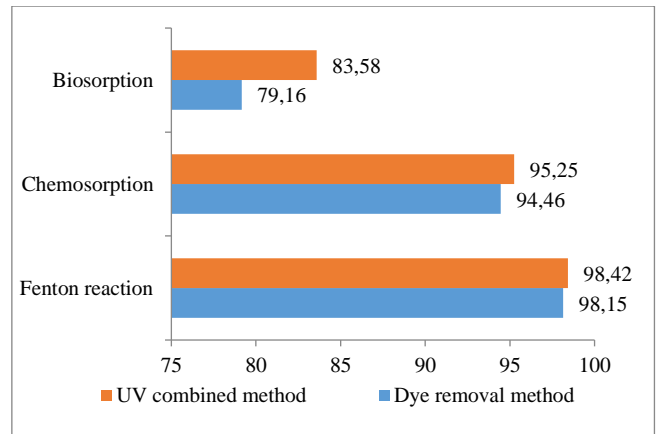


Figure 4 Optimum conditions in the Reactive Black 5 removal reaction (Time: 90 min., pH: 3, Temperature: 50 °C, Dye Concentration: 25 mg/L, Amount of Substance: 50 mg, Amount of H_2O_2 for Fenton Process: 600 μL)

In this study, as a result of the experiments, it was determined that optimum dye removal was achieved by the photo-Fenton reaction under optimum conditions. The removal percentages for the dyes were determined as MB:98.9%, DB15:97.7% and RB5:95.5%, respectively (Figure 2, Figure 3 and Figure 4) [12, 13].

Lucas and his group used Fenton and photo-Fenton reactions for the removal of RB5 dye in their dye removal studies at pH = 3.0. As a result of the reaction, they reported that RB5 could be removed from the water at a rate of 97.5% by the Fenton reaction and 98.1% by the photo-Fenton reaction under optimum conditions [14].

In another study, O'Dowd et al. stated that the optimum pH for the Fenton reaction is between pH 2-4 if Fe II, III is in solution, while when they are used as a heterogeneous catalyst, optimum results are obtained in the range where the pH is higher. In our research, it was observed that although we used heterogeneous catalysts, maximum dye removal was achieved between pH: 2-5 with the effect of nano-iron [15].

As can be seen from the figures drawn as a result of our experiments, the use of the UV combined method has generally positively affected the methods at different rates.

4. Conclusion

As a result of this research, thanks to the reactor we designed, a new dye removal mechanism has been created for dye removal from wastewater. When the obtained data are evaluated, it is clearly seen that UV has a positive effect on the Fenton reaction. It is understood that the reason for this is the photo-Fenton reaction. On the other hand, it was observed that UV conditions did not affect chemosorption positively. Although biosorption itself is not seen as very advantageous compared to other methods, it has been understood by the experimental results that the amount of biosorption with UV increases.

Conflict of Interest

Authors declare that they have no conflict of interest with any person, institution, or company.

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