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## **TREATMENT OF TEXTILE WASTE WATER USING H<sub>2</sub>O<sub>2</sub>/UV SYSTEM**

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Treatment of textile wastewater with the Advanced Oxidation Process (AOP) is based on using the H<sub>2</sub>O<sub>2</sub>/UV system. The optimum condition for treatment of an effluent sample was determined by experimenting on a synthetic dye solution prepared using the blue sulfur dye. Different parameters that affect the reaction rate were tested (UV intensity of 18, 36 and 54 W, initial dye concentration of 70, 80, 90 and 100 ppm, pH 3, original pH of the solution equal to 7.29 and 10 as well as the catalyst dose). Absorbance was measured to determine the decolorization efficiency and then the total organic carbon was measured for the reactions at optimal conditions to assure that decolorization is accompanied by degradation.

*key words: wastes processing, waste water, water purification, decolorization*

### **INTRODUCTION**

Textile industry ranks top among most consumers. This is due to its raw materials, namely cotton, wool and man-made fibers, which are considered as sources of wealth (Hebiesh, 2000). In Egypt, this industry employs about one million manpower and the lives of many Egyptian families are connected with the textile industry (agriculture, trade and services). Furthermore, many research institutes and large companies are associated with the textile industry.

In Egypt, the textile production emerged thousand of years ago and it was founded on scientific basis in 1927. Egypt became an exporter of textile yarn in 1949. In the past the textile production had depended on the export especially to the European Union. Currently, the trade and export of textile is a complex business, in which many

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different aspects, such as marketing, access to the right distribution channels and compliance with the national legislation of the country to which goods will be exported, must be taken into account.

The problems start with the use of pesticides during the cultivation of the natural fibers or by emission during the production of synthetic fibers. Textile raw materials, essentially, cotton, wool, and synthetic fibers pass through several mechanical and chemical stages before reaching the consumer in the form of fabrics or ready-made garment. Certain chemicals which are used in the textile production chain can cause environmental and health problem. These problems may occur during the production process. With respect to emission or occupational health problem e.g., statistical significance RR-4.7 (RR, is a relative risk of individual substance, i.e., the frequency of adverse effects in relation to cumulated doses) was established for exposure to aromatic amines. For those contacting with aniline dyes the relative risk (RR) made up 2.4. The risk to develop bladder cancer in powder shops (RR-3.2) was attributed to the hazards of dyes and diphenylamine. In leather-shoe shops and textile industry the exposure to dyes is not safe (RR-6.1) (Nizamova, 1991; and Kampan et al., 2000). Other problems related to the chemicals used, which appear in the final product and cause, for instance, allergic skin reactions or even cases of cancer.

Textile wastewaters exhibit low BOD to COD ratios ( $<0.1$ ) indicating non-biodegradable nature of dyes (Pagga and Brown, 1986; Reife, 1993). Fifteen percent of the total world production of dyes is lost during dyeing process and is released in the textile effluents. The release of those colored wastewaters in the ecosystem is a dramatic source of esthetic pollution, eutrophication, and perturbations in aquatic life. As international environmental standards are becoming more stringent (EPA, 1998), technological systems for the removal of organic pollutants, such as dyes, have been recently developed. Among the heterogeneous photocatalysis is the Advanced Oxidation Process or AOP, which can be successfully used to oxidize many organic pollutants present in aqueous systems.

Decolorization of dye effluents has therefore acquired increasing attention. During the past two decades, the treatment of spent textile dyeing wastewater by traditional methods has proven to not be effective for many wastewater treatment facilities. Conventional activated sludge treatment is the typical treatment used today, though activated sludge was not originally used for treatment of industrial wastes, particularly textile wastes containing dyes and surfactants (Yang et al., 1998). Additional textile treatment methods such as combination of biological, physical, and chemical methods including coagulation/flocculation, electrochemical oxidation and activated carbon adsorption, reverse osmosis, ozone and oxidative/reductive chemical processes are all techniques that can be used for treating textile wastewater. Biological processes are not effective enough to decolorize dyes of high photostability while ordinary physical/chemical processes are not strong enough to decompose dye. Hence sludge generations are the principal weakness of these processes (Slokar and Majcen, 1998). The AOP decomposes the chromophore of the dye (Ferrero, 2000; Kurbus et al.,

2002) and consequently realizes complete decolorization, so advanced oxidation processes are being considered as emerging technology to handle large volumes of textile waste water.

The concept behind the AOP is exposure of a strong oxidizing agent to ultraviolet (UV) light generates hydroxyl free radicals, which are stronger than any other oxidants (Bolton and Cater, 1994). The hydroxyl radicals generated after activation have a higher oxidation potential (2.8 V) than hydrogen peroxide (1.78 V) and so dye decolorization is feasible. Advanced oxidation is one of the potential alternatives to decolorize and reduce recalcitrant wastewater loads from textile dyeing and finishing effluents. This process implies generation and subsequent reaction of hydroxyl radicals, which are the most powerful oxidizing species after fluorine (Legrini et al., 1993).

When a water or wastewater containing  $H_2O_2$  is irradiated with UV, hydroxyl radicals are formed which are very powerful oxidizing species (Shu et al., 1994; Namboodri and Walsh, 1996). Furthermore, the  $H_2O_2/UV$  process has an additional advantage in that there is no sludge formation during any stage of the treatment (Galindo and Kalt, 1999).

## MATERIALS AND METHODS

To experiment on the efficiency of the  $H_2O_2/UV$  method, the Bleu-N-RM-2114 dye was purchased from the Engineering Company of Textile (Tenth of Ramadan City). The dye was classified as the sulfur dye. It was used directly as received from the textile industry without further purification. The synthetic dye solutions were prepared by dissolving a defined quantity of the dye into a 1-dm<sup>3</sup> Erlenmeyer flask and diluting it with tap water. They were protected from the light and kept in dark. The registration number of the dye is K3-01-0004, the trade name is Bleu-N-RM-2114, and the IUPAC nomenclature of the dye is sodium-1-amino 4-(2-methyl-5- (methyl phenyl sulfamyl amino) phenyl amino) arthraquinone-2-sulphamate.

Before the oxidation experiments were conducted, it was necessary to choose the appropriate concentration of the dye solution. A standard curve was drawn and concentrations of 70, 80, 90 and 100 ppm were selected for experimentation. The decolorization of the dye solution and wastewater was monitored spectrophotometrically with a UV-visible diffuse reflectance Cecil 7200 spectrometer, the maximum absorbance peak was determined by scanning the dye solution with 4000 nm/mm speed, band width 2 nm, and wavelength range from 190 to 900 nm. The maximum absorbance peak was at wavelength 576 nm for the synthetic dye solution and 462 nm for the effluent sample. In order to evaluate the extent of mineralization, total organic carbon measurements were performed using a "Phoenix 8000" Total Carbon Analyzer.

The wastewater sample used in the present study was effluent from the final clarifier at the Engineering Company of Textile (Tenth of Ramadan City). The effluent

was a murky, orange/maroon color and relatively free of particulate matter and it was directly analyzed for absorbance, total organic carbon, metals and the temperature was measured. Irradiation was performed in a cylindrical Pyrex batch reactor of volume  $350\text{ cm}^3$ , fitted with a chamber for the UV lamp to be placed in it. A low-pressure mercury-vapor fluorescent discharge lamp of 18 W consisting of a tubular glass envelope, emitting short-wave ultraviolet radiation (Philips PI-S, with emission 253.7 to 350 nm), was used. The lamp was placed horizontally in the specified chamber. The cylindrical Pyrex had a source of aeration, to assure continuous homogenization of the dye solution with the reactant and also continuous supply of oxygen.

#### TREATMENT BY $\text{H}_2\text{O}_2/\text{UV}$

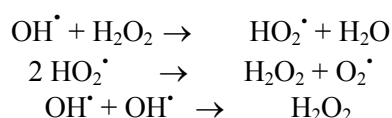
Synthetic dye solutions were injected in the reactor and dosed with different  $\text{H}_2\text{O}_2$  volumes (2.5, 5, 7 and  $10\text{ cm}^3$ ) to study the effect of  $\text{H}_2\text{O}_2$  concentration on the dye degradation. Also experiments were conducted by changing the pH conditions (3, 7.29, 9 and 11), the initial dye concentrations (70, 80, 90 and 100 ppm) and varying UV intensities (18 W, 36 W and 54 W) to study the effect of these factors on the degradation efficiency. Furthermore, samples were taken every 10 minutes and analyzed for UV/visible absorbance to study the decolorization of the solution and the kinetics of the reaction. Afterwards the most optimal conditions for degradation and decolorization of the synthetic dye solution were chosen and then applied on the wastewater sample. As previously mentioned,  $300\text{ cm}^3$  of the wastewater was placed in the reactor, and treatment took place using the optimal condition. The samples were withdrawn at defined time intervals and analyzed for UV/visible absorbance to determine the decolorization efficiency and the kinetic of reaction. At the end of the reaction time, a each sample was taken and analyzed for the total organic carbon content in order to assure mineralization of the wastewater.

## RESULTS AND DISCUSSION

#### EFFECT OF $\text{H}_2\text{O}_2$ DOSE

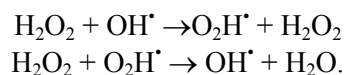
The effect of  $\text{H}_2\text{O}_2$  dosage (ranging from  $3\text{ cm}^3$  to  $10\text{ cm}^3$ ) on the decolorization efficiency was investigated while stabilizing all the conditions of the reaction and altering the  $\text{H}_2\text{O}_2$  doses. The conditions of the treatment experiment are listed as follows: UV power 18 Wt, dye concentration 70 ppm, reaction time 3hrs, pH 7.29, volume of the dye solution  $300\text{ cm}^3$ , room temperature. The effect of the  $\text{H}_2\text{O}_2$  dose on the decolorization efficiency is shown in Figure 1. It can be observed that the decolorization efficiency increases with increasing hydrogen peroxide dose up to a point where it reaches a maximum and then starts to decrease, where the decolorization efficiency

reached 90.69% at H<sub>2</sub>O<sub>2</sub> dose equal to 10 cm<sup>3</sup> (3.9 wt %), whereas above this dose the decolorization efficiency decreased to 82.3% when the applied dose was 12 cm<sup>3</sup> (4.8 wt %) which is consistent with the available literature (Stefan et al., 1996, Chu, 2001 and Ghaly et al., 2001). Daneshvar et al., (2004) reported that in the decolorization of AO7 the same behavior is observed. This behavior is due to the fact that hydrogen peroxide is a scavenger for hydroxyl radicals according to the reaction given in the following equation (Buxton, 1988; Dionysiou et al., 2004):

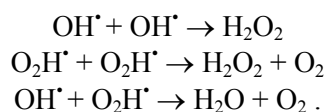


When enough hydrogen peroxide is present in the solution, it starts to compete with the dye for reaction with hydroxyl radicals since HO<sub>2</sub><sup>•</sup> is less reactive than the OH<sup>•</sup> radical. An increased level of hydrogen peroxide has a diminishing effect on the reaction rate (Aleboye et al., 2005). In addition, the OH<sup>•</sup> radicals generated at a high local concentration will readily dimerize to H<sub>2</sub>O<sub>2</sub>. Therefore, it is important to optimize the applied dose of hydrogen peroxide to maximize the performance of the UV/H<sub>2</sub>O<sub>2</sub> process and minimize the treatment cost. The optimum dose for this experiment is 10 cm<sup>3</sup> (3.9 wt %) where the decolorization efficiency reached 90.69%, whereas above this dose the decolorization efficiency decreased to 82.3%, when the applied dose was 12 cm<sup>3</sup> (4.8 wt %). The hydrogen peroxide concentration is an important parameter to adjust and control the decolorization of dyes in the UV/H<sub>2</sub>O<sub>2</sub> reactor. Degradation of the color is due to the hydroxyl radicals generated upon photolysis of hydrogen peroxide (Namboodri and Walsh, 1996; Arslan and Balcioglu, 1999). Several studies have proposed different reaction mechanisms for this photolysis. It is widely accepted that the main interaction between H<sub>2</sub>O<sub>2</sub> with UV radiation and free radicals are well represented by the following reactions (Alfano et al., 2001).

#### *Propagation*

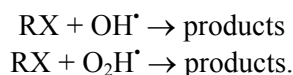


#### *Termination*



The powerful oxidizing hydroxyl radicals react with the dye molecules resulting in the destruction of their intrinsic color.

#### *Decomposition*



The  $\text{OH}^\bullet$  radicals are capable of oxidizing organic compounds mostly by hydrogen abstraction

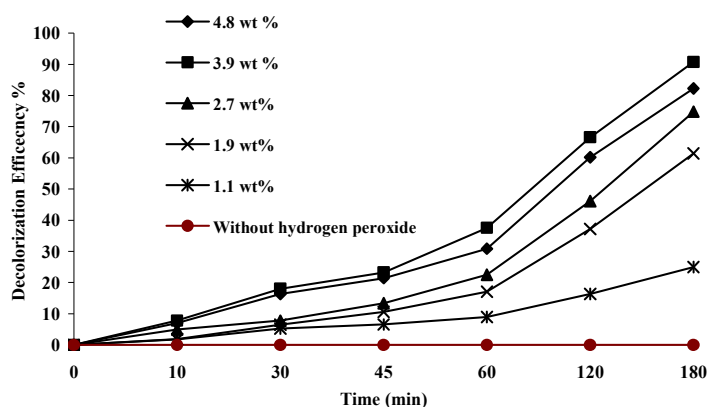
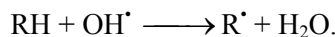


Fig. 1. Effect of  $\text{H}_2\text{O}_2$  concentration on the decolorization efficiency of dye solution using  $\text{H}_2\text{O}_2/\text{UV}$  system

This reaction generates organic radicals which by addition of molecular oxygen yield peroxy radical. These intermediates initiate thermal chain reactions of oxidative degradation leading finally to carbon dioxide, water and inorganic salts. Electrophilic addition of  $\text{OH}^\bullet$  radicals to organic  $\pi$  (double bond) systems constitutes another mechanism of oxidative degradation.

#### EFFECT OF UV POWER

The effect of the UV power on the decolorization efficiency was also studied, by testing UV intensities of 18, 36 and 54 W. The experiments were performed as mentioned earlier, while stabilizing all the other conditions. These conditions are:  $\text{H}_2\text{O}_2$  dose 3.9 wt %, dye concentration 70 ppm, reaction time 3hrs, pH 7.29, volume of dye solution 300  $\text{cm}^3$ . As shown in Figure 2, when the UV power was 18 W, the decolorization efficiency was 7.84% after 10 min, and then, at the end of the 3 hrs of decolorization, the efficiency reached 90.69%. By increasing the UV power to 36 W, the decolorization efficiency became 21.84% after 10 min, and finally the decolorization efficiency reached 94.7% at the end of the 3 hrs. The UV power was further increased to 54 W to study its effect on the decolorization efficiency. The data showed that after 10 min the decolorization efficiency was 24.93% after 10 min, then at the end of the 3 hrs the decolorization efficiency was 100%. This reveals that increasing the UV power from 18 to 54 W the removal efficiency increased from 90.69% to

100%. This increase in decolorization is due to increased production of hydroxyl radical. At low UV power, the rate of photolysis of  $H_2O_2$  into hydroxyl radical ( $H_2O_2+h\nu \rightarrow 2\cdot OH$ ) is reduced (Yang et al., 1998).

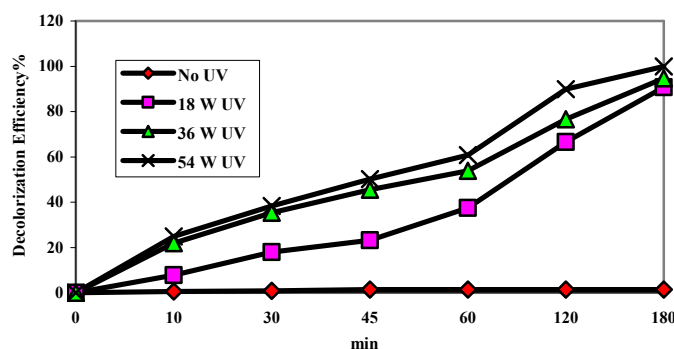


Fig. 2. Effect of UV power on decolorization efficiency

Therefore, the best optimal condition for the experiment is utilizing the UV power of 54 W, because it yields the maximum decolorization percentage.

#### EFFECT OF THE INITIAL DYE CONCENTRATION ON THE DECOLORIZATION EFFICIENCY

After studying the effect of the two previous factors, another factor was studied which is the effect of the initial dye concentration on the decolorization efficiency. This factor was studied by stabilizing all the other conditions as follows:  $H_2O_2$  dose 3.9 wt %, reaction time 3 hrs, pH 7.29, UV power 54 W, volume of the dye solution  $300\text{ cm}^3$ , room temperature. The results show that by increasing the initial dye concentration from 70 ppm to 100 ppm the removal efficiency decreased from 100% to 70.5%.

Hydroxyl radical is mainly responsible for dye decolorization and its concentration remains constant for all dye concentration. The increase in dye concentration increases the number of dye molecules and not the hydroxyl radical concentration and therefore the removal rate decreases. The increase in dye concentration may also induce a rise of the internal optical density and the solution becomes more and more impermeable to UV radiation. Then, hydrogen peroxide can only be irradiated by a smaller portion of UV light to form less free radicals and the color degradation rate decreases (Aleboyeh et al., 2005). Aleboyeh et al., (2003) obtained similar results for the acid orange 8, acid orange 7, and methyl orange dyes.

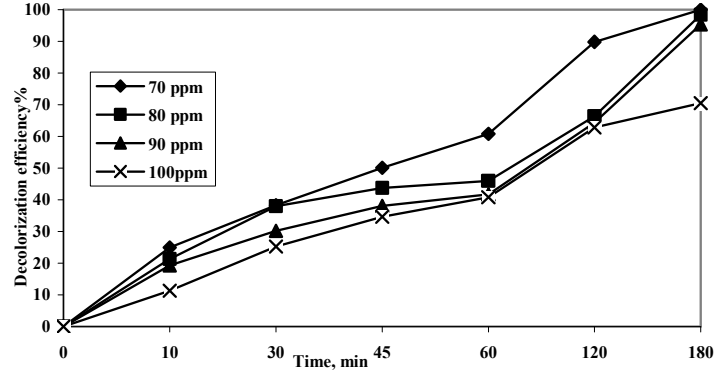
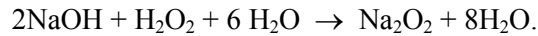


Fig.3. Effect of initial dye concentration on the decolorization efficiency of the blue dye solution using  $H_2O_2/UV$

#### EFFECT OF pH ON DECOLORIZATION EFFICIENCY

The effect of pH on the decolorization efficiency of the blue dye solution was studied by stabilizing all the other conditions and only changing the pH of the dye solution where  $H_2O_2$  dose was  $10 \text{ cm}^3$ , reaction time 3 hrs, UV power 54 W, dye concentration 70 ppm, volume of dye solution  $300 \text{ cm}^3$ . As shown in Figure 4, the decrease of the pH from the original conditions of the dye solution (pH=7.29) to 3.03 has slightly decreased the removal efficiency from 100% to 98.75% while increasing the pH from the original conditions of the dye solution (7.29) to 11.11 led to a decrease in the removal efficiency from 100% to 86.96%. It can be concluded that the pH increase leads to a decrease in the decolorization efficiency. This can be explained by the fact that part of  $H_2O_2$  is used for the oxidation of alkalis (NaOH) during the decolorization forming sodium peroxide ( $Na_2O_2$ ). As a result, less hydrogen peroxide is available for the formation of hydroxyl radicals and consequently the degradation of the dye decreases. This behavior is especially noticeable at higher concentration of NaOH as follows:



Oxidation with  $H_2O_2$  should therefore be performed in acidic medium, but acidification and neutralization increase the cost of such processes. Fortunately in our case there was no great effect of acidification on the decolorization efficiency of the synthetic dye solution.



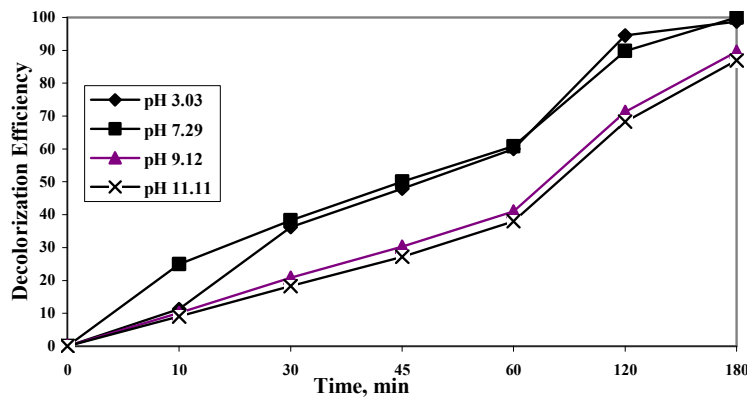


Fig.4. Effect of pH variation on the decolorization efficiency of the blue dye solution using  $H_2O_2/UV$

The most favourable conditions which give a full decolorization rate are as follows:  $H_2O_2$  dose  $10 \text{ cm}^3$  (3.9 wt %), pH 7.29, initial dye concentration 70 ppm, UV intensity 54 W. The total organic carbon or TOC, a measure of the quantity of organically bound carbon that can be oxidized to  $CO_2$ , was analyzed before and after the decolorization process to prove that the decolorization of the solution is accompanied by degradation and mineralization of organic matter. It was found that the TOC of the original dye solution was  $62 \text{ mg/dm}^3$  and after treatment it reached  $4.34 \text{ mg/dm}^3$  (removal efficiency 93%). It can be concluded that although the decolorization efficiency reached 100% but the TOC removal did not reach that percentage. This may be attributed to the destruction of the dye into intermediates that are no longer visible and need smaller amounts of oxygen to completely oxidize, while at the same time these intermediates cause higher TOC values (Kurbus et al., 2002).

#### TREATING THE EFFLUENT SAMPLE USING $H_2O_2/UV$ SYSTEM

First, TOC was determined ( $700 \text{ mg/dm}^3$ ) before applying the  $H_2O_2/UV$  treatment method, then the treatment was carried under the optimum conditions of the  $H_2O_2/UV$  system:  $H_2O_2$  dose  $10 \text{ cm}^3$  (3.9 wt %), pH 7.29, initial dye concentration 70 ppm, UV intensity 54 W. After 5 hours the TOC was determined as  $105 \text{ mg/dm}^3$  (removal efficiency 85%).

#### CONCLUSIONS

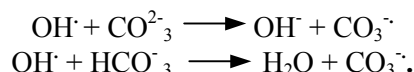
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of oxygen to completely oxidize while at the same time these intermediates cause higher TOC values (Kurbus et al., 2002). It can be noticed that the TOC removal percentage in the effluent sample is less than removal efficiency of the synthetic dye solution (except for the ozone system). This can be explained by two reasons:

1. presence of other than dyes refractory materials
2. increase in alkalinity which may be due addition of sodium carbonate.

Sodium carbonate is the common auxiliary chemical employed in textile processing operations where it is used in the dyeing path in order to adjust the pH of the bath as it plays an important role in fixing the dye on the fabrics and in the fastness of color.

The decrease in the degradation of the sample in the presence of carbonate ions is due to the hydroxyl scavenging property of carbonate ions which can be accounted from the following reactions:



It can also be noticed that the TOC removal is incomplete while the color has completely disappeared. This phenomenon may be due to the destruction of the chromophore of the dye which is responsible for the color appearance and transforming it into smaller fragments of molecules (intermediates). Extending the reaction time should completely eliminate the TOC.

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**Amin H., Amer A., El Fecky A., Ibrahim I.,** *Przeróbka wody odpadowej za pomoca sytemu  $H_2O_2/UV$ , Physicochemical Problems of Mineral Processing, 42 (2008), 17-28 (w jęz. ang)*

Przeróbka wód odpadowych przemysłu farbiarskiego metoda Advanced Oxidation Process (AOP) jest oparta na układzie  $H_2O_2/UV$ . Określono eksperymentalnie optymalne warunki przerobu odcieków dla syntetycznego barwnika *blue sulfur*. Badano parametry które wpływają na szybkość reakcji (intensywność UV na poziomie 18, 36 oraz 54 W, początkowe stężenie barwnika 70, 80, 90 oraz 100 ppm, pH 3 oraz naturalne wynoszące od 7.29 do 10, jak również ilość katalizatora). Mierzono absorbcję dla określenia wydajności odbarwiania oraz całkowitą ilość węgla organicznego dla optymalnych warunków reakcji zapewniających rozpadu barwnika w trakcie jego dekoloryzacji.

*słowa kluczowe: przeróbka odpadów, wody odpadowe, oczyszczanie wód, dekoloryzacja*