



Recycling of textile dyeing wash-off liquor using Fenton technology

Ayesha Siddique*, Irfan Ahmed Shaikh, Azhar Ali, Sana Islam, Anum Tariq, Khudajah Ikram

College of Earth and Environmental Sciences, University of the Punjab, Lahore, Pakistan, emails: ayeshaabubakar0@gmail.com (A. Siddique), Irfan.cees@pu.edu.pk (I.A. Shaikh), Azhar.cees@pu.edu.pk (A. Ali), Islamsana86@gmail.com (S. Islam), Anum.cess@pu.edu.pk (A. Tariq), Khudajah.ikram@gmail.com (K. Ikram)

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ABSTRACT

Significant problems of the textile industry are high water consumption and a large amount of complex wastewater generation. The current study focused on the recycling of textile wastewater in the process after treatment using Fenton process. The application was carried out lab-scale synthetically produced wastewater after dyeing with Reactive Orange 122, Yellow 84, Blue 221, and Red 223. The optimum value for Fenton treatment were selected pH: 3, FeSO₄: 500 mg/L, H₂O₂: 525 mg/L, and time: 60 min. The treated wastewater was recycled thrice in the process after treatment. The color change was ascertained in terms of color measurement, color fastness, and color strength. Results revealed that the fabric quality wasn't compromised in any way, as the color difference of the first two samples of all dyes was in the acceptable tolerance limit, except for the Red 223. Washing fastness showed no change in the results of fabric standard and samples as most of the samples showed excellent results. *K/S* results displayed that the samples of Orange 122 and Yellow 84 were lower; however, Blue 221 and Red 223 samples were greater than the standard. It was concluded that Fenton treatment was best suited for wash-off wastewater except Red 223, which was further recycled in the dyeing process. Thus, the application can save a huge amount of water consumption as well as effluent generation in the textile industry.

Keywords: Wastewater; Color difference; Textile treatment; Recycling

1. Introduction

Water is vital for life, and a valuable reserve for any country. Whenever there is an increase in agricultural activities and industrialization, there is an immense increase in water demand [1–3]. Pakistan ranks highest in water scarcity, which in turn is imposing strain on sustainable use and recycling of water [4]. Textile is a diverse industrial sector with a global economic impact. In Pakistan, the textile industry is contributing towards the country's growing economy and consumes not only humongous quantities of freshwater in terms of surface and groundwater sources, but also generates a huge quantity of wastewater with a high pollution load, leading to massive negative environmental

impacts [5–7]. The water demand can be met by opting for water-efficient processes and treatment methods [8]. Wash-off stages in the dyeing process of textile industries consume a lot of water, as the dyed fabric has to go through the washing steps several times to improve its color properties [9,10]. For the manufacturing of one kg of textile product, 200 L of water is used, mainly during the application of chemicals on fabrics, and the washing process of the final textile item [11]. Furthermore, wastewater generated from textiles contains contaminants such as acids, salts, alkali, dispersants, leveling agents, and different kinds of dyes [12].

Likewise, dyes used in printing and dyeing mainly constitute of aromatic and heterocyclic compounds having complex structures which makes them stable and

* Corresponding author.

recalcitrant, and consequently, hard to degrade [13]. The most commonly used dyes in the textile industry are reactive dyes, because of their efficient fastness properties [14]. Reactive dyes which are generally used for dyeing of cotton (cellulosic compound) are highly stained compounds having chromophore and a substituent group which gets attached to the fabrics with a covalent bond. These dyes adsorb on the fabric, the unfixed dyes are washed, which leads to excessive use of water [15], and about 8%–20% of the dyes are discharged off into the wastewater resulting in high pollution load [16,17].

There are many techniques for the treatment of wastewater such as physico-chemical methods (adsorption, coagulation, membrane technology, and electrochemical processes), advanced oxidation processes (AOP), and biological treatments (aerobic and anaerobic) [18]. Among several available feasible wastewater treatment options, advanced oxidation processes have prime importance [19]. Biological treatment methods have been used for many decades, but complex industrial effluents demand novel and promising methods with better efficiency and cost-effectiveness. AOPs including electrocoagulation and Fenton treatment are the best examples that are used to treat wastewater by oxidizing the organic pollutants [20]. These methods are very reactive, having the capability to destroy organic and recalcitrant compounds in the water far more efficiently, as compared to other methods [19]. AOPs make use of hydroxyl radicals, which are very fast oxidizing agents having an oxidizing potential of 2.33 V, by the virtue of which they react with the dyes at a high reaction rate [5].

Similarly, chemical methods like the Fenton process have provided a favorable and economically feasible method for destroying organic and hazardous contaminants in wastewater [21]. Fenton's reagent is a mixture of iron salt and hydrogen peroxide in which iron salt stimulates the breakdown of H_2O_2 to destroy organic pollutants that are indestructible by any biological means. Both soluble and insoluble dyes can be effectively removed by this process [22]. Fenton process is very effective in wastewater treatment, as it removes color and COD, and transforms organic compounds into water and carbon dioxide after destructive oxidation reaction. Colored effluent is the main problem which is mainly generated by dyeing and finishing processes in textile operations in the industry [23]. Reuse application of wastewater after its treatment in industrial processes has been recommended by many researches, as water can be conserved in terms of reuse after treating it [24–26].

Given an ever-increasing lack of water due to tremendous utilization, this study focuses on finding a feasible solution for saving water, and reducing water contamination, by treating the wash-off wastewater with the Fenton oxidation process, and reusing it in subsequent wash-off steps.

2. Materials and methods

2.1. Materials

Five grams of knitted and bleached, 100% pure cotton fabric was used. NaCl and Na_2CO_3 were used as fixation and exhaustion agents, respectively, along with acetic acid

which was used for neutralization. $FeSO_4 \cdot 7H_2O$ (molecular weight: 151.91 g/mol, odorless, turquoise in color, and soluble in water) and 30% w/w H_2O_2 (molecular weight: 34.015 g/mol, molarity = 0.89 M, normality = 1.78 N, almost colorless (pale blue) liquid, soluble in water, has a boiling point of 423 K and melts at 272.4 K, oxidizing and bleaching agent) were used as Fenton's reagent for the Fenton oxidation treatment. Sulfuric acid (H_2SO_4 , 98%) and sodium hydroxide (NaOH, 0.1 M) were used for maintaining pH. Reactive dyes were used because of their extensive application in the textile industry, and these dyes were used without any further purification. Properties of dyes are given in Table 1.

2.2. Synthetic dyeing process

Isothermal dyeing was done with the dye of 5% shade depth, and a 1:10 liquor ratio was maintained. For standard dyeing, 0.25 g of dye and four grams of salt, along with 50 mL of water was taken in a beaker to make the solution. Afterward, 5 g of fabric was dipped into the water and placed on a thermostatic water bath. 1 g of Na_2CO_3 was added when a temperature of 60°C was attained. Continuous mixing was done for 45 min to avoid any uneven patches and discoloration on fabric. After 45 min, the beaker was removed from the water bath and subjected to the wash-off process. The same process was done for all of the four dyes.

2.3. Production of wash-off liquor

After isothermal dyeing, the fabric was squeezed and sent through six stages of the wash-off process to remove extra unfixed dyes. A liquor ratio of 1:10 was maintained. The wastewater collected from wash-off process was analyzed, and then subjected to treatment. The process sequence of dyeing and wash off is given in Fig. 1.

2.4. Analytical work

Fenton treatment process parameter optimization was done for the samples and standards by evaluating and controlling various factors viz. pH, time, and concentrations of $FeSO_4$ and H_2O_2 . Color removal efficiency (%) was evaluated with the help of the given formula. Color removal efficiency (%) = $(A_i - A_f)/A_i \times 100$, where A_i is the dye solution absorbance before treatment, A_f is the dye solution absorbance after treatment. Color difference (ΔL^* , Δa^* , Δb^* , ΔC^* , ΔH^* , and ΔE^*) and color strength (K/S) values were evaluated by SF600 spectrophotometer with the visible range of 400–700 nm. Samples washed off with Fenton treated wastewater were subjected to CIELAB test to check the change in the shade [27]. The K/S values of dyed fabrics were calculated by the Kubelka–Munk equation which is $K/S = (1 - R)^2/2R$, where K is the absorption coefficient, S is the scattering coefficient, R is the reflectance of dyed samples. This reflectance-based value is the decreased ratio of light obtained by absorption and scattering [28]. Standards of American Association of Textile Chemists and Colorists (AATCC) were followed in which fastness to washing and rubbing (dry and wet) was determined according to the 61-2013 and 8-2013 standards [29] with the help of Gyrowash and Crock meter. Washing fastness was performed according to the ISO

Table 1
Properties and chemical arrangements of synthetic dyes used

Dyes names (C.I Reactive)	Commercial name	Molecular weight (g/mol)	λ_{\max} (nm)	Chemical structures
Orange122	Jackofix Orange ME2RL	1,034.27	490	
Yellow 84	Jackofix Yellow HE4R	1,628.22	226	
Blue 221	Synozol Blue KBR	1,082.83	520	
Red 223	Synozol Red GD	1,041.26	490	

105:C06/C2S [30]. Results were evaluated with the help of AATCC chromatic transference scale which ranges from 1 (poor) to 5 (excellent).

2.5. Experimental setup

2.5.1. Treatment with Fenton oxidation process

The wastewater was treated chemically with the Fenton process in which 500 mg of FeSO_4 and 525 mg H_2O_2 was added, which resulted in the precipitate formation, and appearance of pale yellowish wastewater. One hour later, the precipitates settled down, and discolored water separated as supernatant.

2.5.2. Recycling of Fenton treated wash-off

Wastewater treated by the Fenton process was subsequently used in the wash-off procedure of other dyed fabrics.

For the purpose, the wastewater was neutralized with the basic solution of NaOH, till precipitates formed. After filtration, completely colorless wastewater was obtained which can be reused. The wastewater was thrice recycled; hence, three samples of each of the dye were used along with the reference (standard). The workflow of this research is shown in Fig. 2.

3. Results and discussion

3.1. Optimization of parameters for Fenton process

Process optimization was ascertained by observing the impact of various parameters on color removal efficiency. Color removal was evaluated with the assistance of visual discoloration. Spectrophotometric analyses were also used at the λ_{\max} of dying wastewater to evaluate the color removal efficiency of the treated samples. Both techniques almost

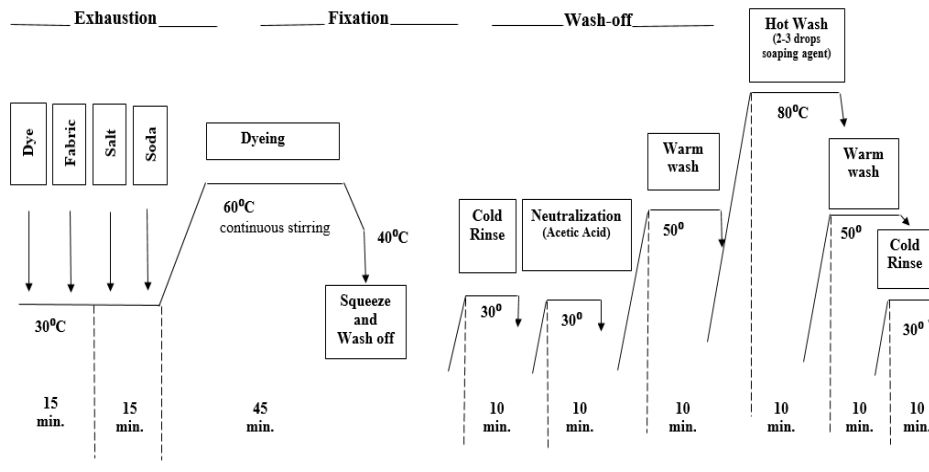


Fig. 1. Process route of isothermal dyeing and wash-off.

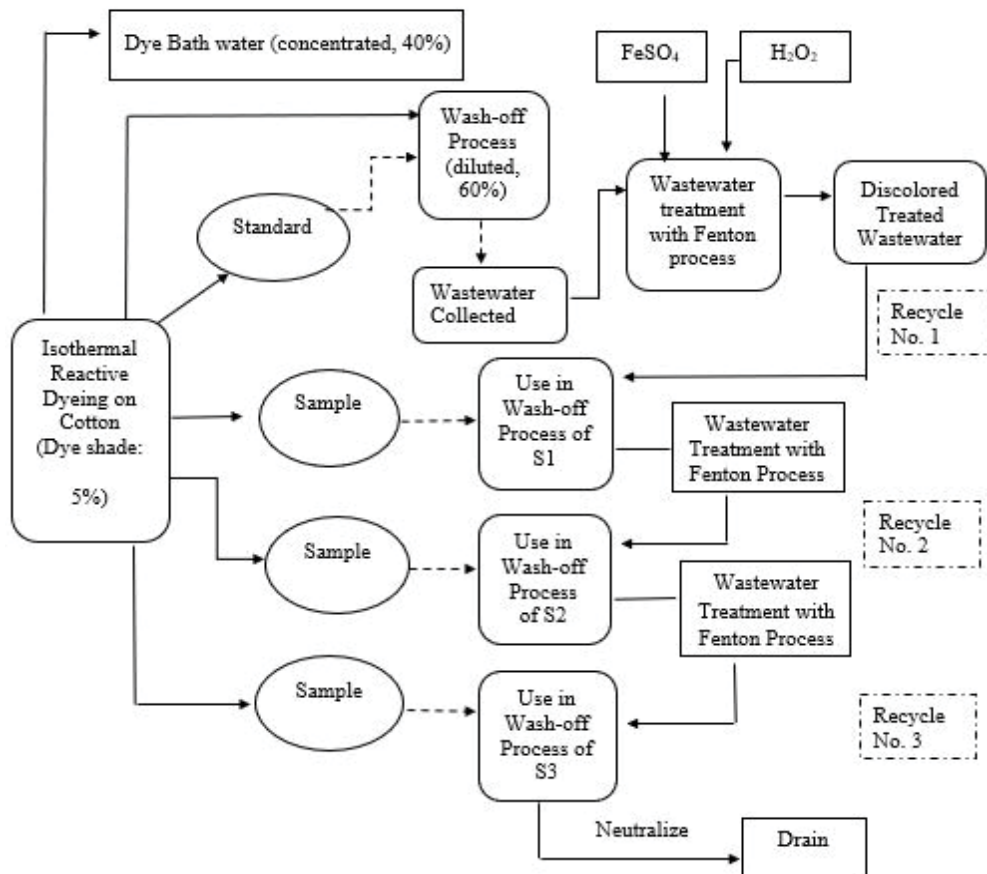


Fig. 2. Workflow of recycling of Fenton treated wash-off wastewater.

showed the same results, that is, discoloration at the same point.

3.2. Impact of pH on color removal of dyes

The pH was determined by keeping reaction conditions constant (FeSO_4 concentration = 500 mg/L, H_2O_2

concentration = 525 mg/L, and time = 60 min) during the whole experimentation series, as the literature extensively claims pH to be a crucial parameter for the Fenton process [31]. Results revealed that maximum color removal (98%) was attained at a pH of 3 (Fig. 3). An increase in the pH to 3.5 and greater, color removal efficiency showed a significant subsequent decrease. This decrease in efficiency is due

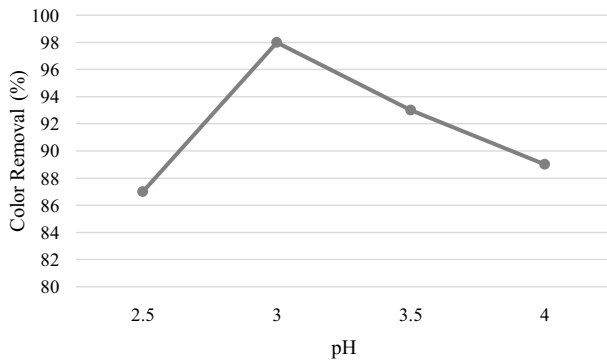


Fig. 3. Color removal efficiency of dyes at different pH at reaction conditions: FeSO_4 concentration = 500 mg/L, H_2O_2 concentration = 525 mg/L, and time = 60 min.

to the fact that the stability of Fe^{2+} is reduced because of less solubility of Fe ions at high pH levels; hence, reducing the hydroxide ions (OH^-) production. In addition, Fe reacts with OH^- and produces $\text{Fe}(\text{OH})_2$ resulting in the precipitation of $\text{Fe}(\text{OH})_3$, which does not have the capability to react with H_2O_2 . Therefore, the degradation rate is greatly reduced [32]. The optimum value obtained for pH is 3 where the color removal was at its best. Similar results have been reported by other studies as well [25,33–37].

3.3. Impact of reaction time on color removal of dyes

Reaction time plays an important role in the discoloration of wastewater. The impact of time on the removal rate of dyes is demonstrated in Fig. 4. Setting reaction conditions constant (FeSO_4 concentration = 500 mg/L, H_2O_2 concentration = 525 mg/L, pH = 3), a time range of 30, 45, 50, and 60 min was set. Results revealed a direct association between time and color removal efficiency. It was observed that the maximum color removal (98%) happened at 60 min. Reaction time showed an increasing trend; that is, as time increased, the color removal efficiency also increased which proved that the removal efficiency might be attributable to the oxidation of organic pollutants with OH^\bullet radicals [20]. Previous studies have reported similar results where 60 min was the optimum value of time for the Fenton process [38,39].

3.4. Impact of FeSO_4 concentration on color removal of dyes

Fig. 5 illustrates the impact of FeSO_4 dose on the color removal of dyes. The range of FeSO_4 was set at different concentrations of 50, 250, 500, and 700 mg/L, by keeping pH = 3, H_2O_2 concentration = 525 mg/L, and time = 60 min. A maximum discoloration rate of 98% was observed at 500 mg/L. Results indicated that with an increase in the FeSO_4 concentration, the removal rate also increased. This is due to the fact that Fe^{2+} promotes the generation of hydroxyl radicals. However, by increasing FeSO_4 concentration from 500 to 700 mg/L, removal efficiency significantly decreased (90%). This happens because an increase in Fe concentration beyond this amount reduces the OH^- concentration gave the scavenging effect of OH^- [40,41]. The same trend

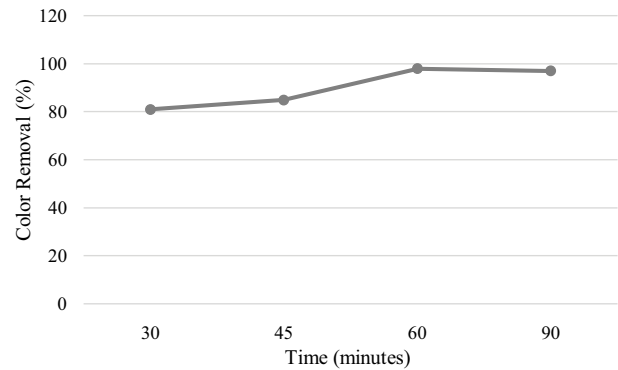


Fig. 4. Color removal efficiency of dyes at different reaction time at reaction conditions: FeSO_4 concentration = 500 mg/L, H_2O_2 concentration = 525 mg/L, and pH = 3.

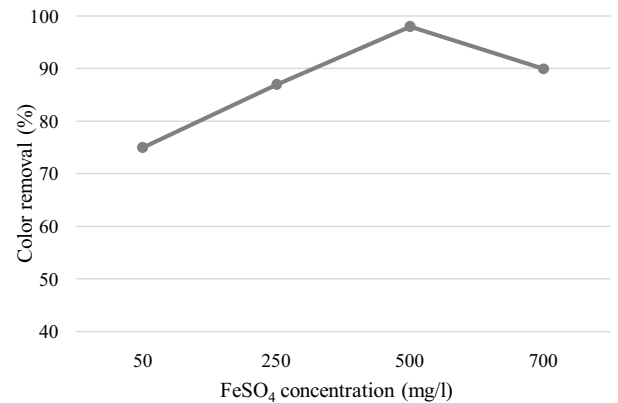


Fig. 5. Color removal efficiency of dyes at different concentrations of FeSO_4 at reaction conditions: pH = 3, H_2O_2 concentration = 525 mg/L, and time = 60 min.

was observed by a few other studies which showed that by increasing FeSO_4 concentration, rate of removal decreased [42]. Therefore, the optimum value of Fe concentration was found to be 500 mg/L [43,44].

3.5. Impact of H_2O_2 concentration on color removal of dyes

The effect of H_2O_2 on dyes' removal efficiency has been shown in Fig. 6. A concentration range of 175, 350, 525, and 680 mg/l was set. Reaction conditions were pH = 3, FeSO_4 concentration = 500 mg/L, and time = 60 min. It was seen that most color removal (95%) was obtained at 525 mg/L of H_2O_2 . Results demonstrated that by increasing H_2O_2 concentration, decolorization efficiency also increases because of the sufficient amount of OH^\bullet radical generation. Further increase in the concentration of H_2O_2 from 525 to 680 mg/L shows that the removal rate of dyes decreased to 90% [45]. This is due to the fact that H_2O_2 shows scavenging behavior where it generates the hydroperoxyl radical (HO_2^\bullet) which is comparatively less active than OH^\bullet radicals, which further reduces the concentration of OH^- in solution [37,46]. Higher values of H_2O_2 beyond a certain point can affect negatively

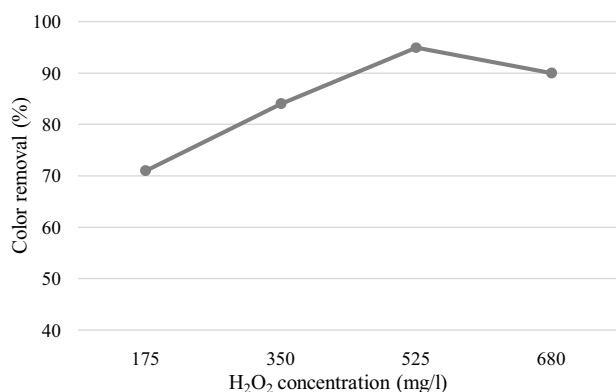


Fig. 6. Color removal efficiency of dyes at different concentrations of H₂O₂ at reaction conditions: pH = 3, FeSO₄ concentration = 500 mg/L, and time = 60 min.

as a result of the breakdown of H₂O₂ itself, which produces water and oxygen [47].

3.6. Color measurement

The CIELAB color scale was used to evaluate the color difference of dyed fabrics which were washed-off with the treated wastewater. The color difference was used as a measure for approving or rejecting the quality of the dyed fabric, as well as for matching the shade of dyed fabric against the standard. The L^* axis represents the lightness and darkness of the dye. The a^* axis represents the redness and greenness, while b^* axis represents the yellowness or blueness. The C^* is the difference in chroma and h^* is the difference in hue. The total color difference E^* is a single value that has been calculated by taking differences between the L^* , a^* , and b^* of the sample and standard. If the E^* value is either below one or equal to one the color difference is acceptable [48].

In case of C.I Reactive Orange 122, ΔL^* for sample 1 is lighter (1.14) while ΔC^* is duller (−0.73). Δa^* represents greener (−0.92), Δb^* represents bluer (−0.07). Whereas, ΔE^* is 0.72, which falls within an acceptable range of CMC (Color Measurement Committee of the Society of Dyes and Colorists of Great Britain) tolerance system. For the sample 2, ΔL^* is lighter (1.66), while ΔC^* is duller (−0.45); likewise, Δa^* is −0.65 while Δb^* is 0.03, representing greener or bluer shade. ΔE^* is 0.88 which is also acceptable. For sample 3, ΔL^* is lighter (2.13) and ΔC^* is duller (−0.63) while Δa^* represents greener (−0.84) and Δb^* represents bluer (−0.02) shade. ΔE^* is 1.11 which is not in an acceptable range. Summary of the color difference values are shown in Fig. 7.

In the case of C.I Reactive Blue 22, as shown in Fig. 8, ΔL^* for all of the three samples show lighter shade (0.90, 0.28, and 0.30). ΔC^* of all the three samples show that they are brighter than the standard (0.66, 1.27, and 0.95). Δa^* and Δb^* of all samples show redder (0.07, 0.67, and 1.03) and bluer (−0.66, −1.24, and −1.35) shades, respectively. ΔE^* of the first two samples is in the acceptable limit (0.72, 0.86) while the third sample is not in admissible limits (1.12).

In the case of C.I Reactive Yellow 84, ΔL^* for sample 1 is lighter (0.27), ΔC^* is brighter (0.35), Δa^* is redder (0.86), and Δb^* is of bluer shade (−0.04). ΔE^* is within acceptable

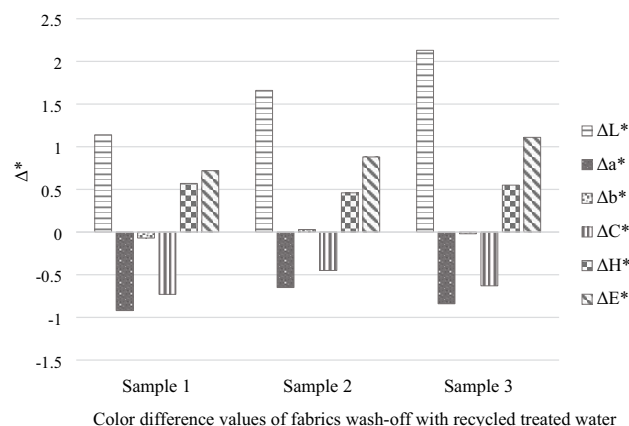


Fig. 7. Summary of color difference values of fabrics dyed with C.I Reactive Orange 122.

limits (0.59). For sample 2, ΔL^* shows lighter (0.67) while ΔC^* shows duller shade (−1.41). The values of Δa^* and Δb^* are −1.50 and −0.86, showing greener and bluer shade. The value of ΔE^* is acceptable (0.88). ΔL^* for sample 3 is −0.01, showing darker shade. ΔC^* shows duller shade (−2.28). Δa^* and Δb^* values are −1.95 and −1.58, respectively, representing that sample is exhibiting greener and bluer shade. ΔE^* is not acceptable (1.05). Results are given in Fig. 9.

In the case of C.I Reactive Red 223, as shown in Fig. 10, for sample 1, ΔL^* value is −1.08, showing darker shade. ΔC^* is −1.97 showing a shade duller than the reference. The values of Δa^* and Δb^* show that the sample is towards greener (−1.73) and bluer (1.11) shade. ΔE^* is 0.94, demonstrating that it is in tolerable limits. For sample 2, ΔL^* value is −0.77, while ΔC^* value is −2.03, showing darker and duller shades, respectively. Δa^* shows greener (−1.58) while Δb^* shows bluer (−1.77) shade. ΔE^* is 1.08, which is unacceptable.

The overall results after analyzing ΔE^* showed that the first two samples of dyes are in the acceptable limit. In the case of Red 223, water can be recycled only once, because red is the darker shade and the results of only the first sample are in an acceptable range. Potential of the Fenton process for the recycling of wastewater from the wash-off process has also been successfully determined by Islam et al. [15]. Similarly, Uygur [49] also demonstrated the reuse applicability of wastewater.

3.7. Wash fastness

Wash fastness test of samples washed off with Fenton treated wastewater was carried out with the help of multi-fiber strip that ranges from 5 (excellent) to 1 (poor). The test was carried out at 50°C. According to the result given in Table 2, there is no difference between the standard and samples. The results of cellulose acetate, acrylic nylon 6.6, wool worsted, and polyester terylene for C.I Reactive Blue 221, Yellow 84, and Red 223 are 4–5 (excellent), and in the case of Orange 122, results are 4 (good). This means there was no staining on fabric washed-off with Fenton treated wastewater. The results of un-mercerized cotton for C.I Reactive Blue 221, Yellow 84 is 4, while in case of Orange 122, results are in the range of 3 to 4. In case of

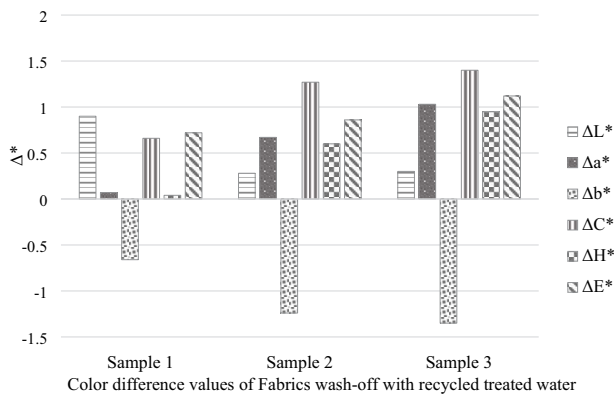


Fig. 8. Summary of color difference values of fabrics dyed with C.I Reactive Blue 221.

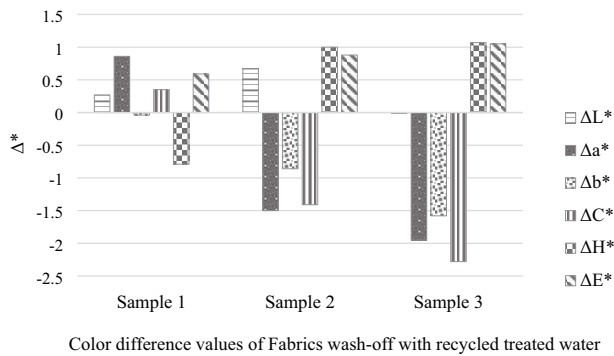


Fig. 9. Summary of color difference values of fabrics dyed with C.I Reactive Yellow 84.

Red 223, the value is 3 for un-mercerized cotton because the red shade is deeper. Other studies have also shown no differences in reuse water and initial water [50,51].

In dry and wet crocking tests, the results for dry crocking for all of the four dyes are 4-5 which means excellent. But in case of wet crocking the results are 3-4. C.I Reactive Red 223 gives the results 3 (poor) which means this shade is darker than others. The results of the standard and samples are not different significantly. Results of other studies also showed no influence in wash fastness of dyeing [52,53].

3.8. Color strength (K/S)

K/S values were evaluated, and the comparison was done between samples and standards. The plain line shows the standard and other lines show the samples. The more the dye present in the fabric, the deeper is the shade, causing

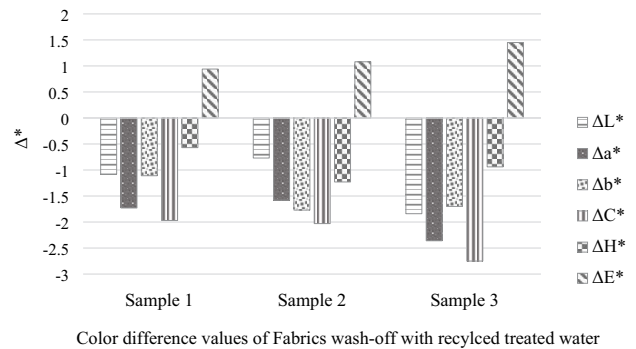


Fig. 10. Summary of color difference values of fabrics dyed with C.I Reactive Red 223.

Table 2

Wash Fastness results of dyed fabrics wash-off with untreated (standard) and Fenton treated recycled wastewater (samples)

Dyes	Staining							Crocking	
	Recycled treated water	Cellulose acetate	Un-mercerized cotton	Nylon 6.6	Polyester terylene	Acrylic	Wool worsted	Dry	Wet
Orange 122	Standard	4	3-4	4	4	4	4	4-5	4
	S1	4	3-4	4	4	4	4	4-5	4
	S2	4	3-4	4	4	4	4	4-5	4
	S3	4	3-4	4	4	4	4	4-5	4
Blue 221	Standard	4-5	4	4-5	4-5	4-5	4	4-5	3-4
	S1	4-5	4	4-5	4-5	4-5	4-5	4-5	4
	S2	4-5	4	4-5	4-5	4-5	4-5	4-5	4
	S3	4-5	4	4-5	4-5	4-5	4-5	4-5	4
Yellow 84	Standard	4-5	4	4-5	4-5	4	4-5	4-5	3-4
	S1	4-5	4	4-5	4-5	4-5	4-5	4-5	4
	S2	4-5	4	4-5	4-5	4-5	4-5	4-5	4
	S3	4-5	4	4-5	4-5	4-5	4-5	4-5	3
Red 223	Standard	4-5	3	4-5	4-5	4-5	4-5	4-5	3
	S1	4-5	3	4-5	4-5	4-5	4-5	4-5	3
	S2	4-5	3	4-5	4-5	4-5	4-5	4-5	3

S: Sample (fabrics wash-off with recycled wastewater).

higher K/S curves and vice versa [28]. Color strength (K/S) of samples of C.I Reactive Orange 122 and Yellow 84 are less than the standard at the maximum wavelength (λ_{max}) of 500 and 440 nm (Figs. 11 and 12) which shows first two samples are almost near the standard having low values of K/S . Fig. 13 shows K/S trend of C.I Reactive Blue 221 that all three samples are greater than the standard having elevated curves at a λ_{max} of 620 nm showing higher K/S values which means shade is deeper causing more dye to present on fabric. Fig. 14 shows K/S values of C.I Reactive Red 223 that the curve of first samples is almost the same as the standard, while the subsequent curve is slightly higher than the standard at λ_{max} of 530 nm showing high values of K/S because of deeper shade [54].

4. Conclusion

This study was aimed to assess the efficiency of the Fenton process for the treatment of dyeing wastewater, and to evaluate the possibility of recycling Fenton treated

wastewater. The efficacy of this method was examined on cotton fabrics dyed with Reactive Orange 122, Reactive Yellow 84, Reactive Blue 221, and Reactive Red 223. Wash-off wastewater was treated with the Fenton process, and then reused in the washing process of other dyed fabrics. The effectiveness of the treatment process for the removal of dye color was evaluated by the optimization of factors such as pH, $FeSO_4$ concentration, H_2O_2 concentration, and reaction time at room temperature. It was analyzed that the process has a fast reaction rate and it also has financial advantages, because of its cost-effectiveness. Quality of dyeing was determined on the basis of color change properties, washing fastness, and color strength. Results indicated that the color difference values were in acceptable limits for the first two samples of Reactive Orange 122, Reactive Yellow 84, and Reactive Blue 221, while in case of Reactive Red 223, the only first sample was within the acceptable limit. Washing fastness results showed little variation between the standard and the samples. Color strength results indicated that most of the samples have a lower curve than the standard in case of Reactive Orange 122 and Yellow 84, while greater in case

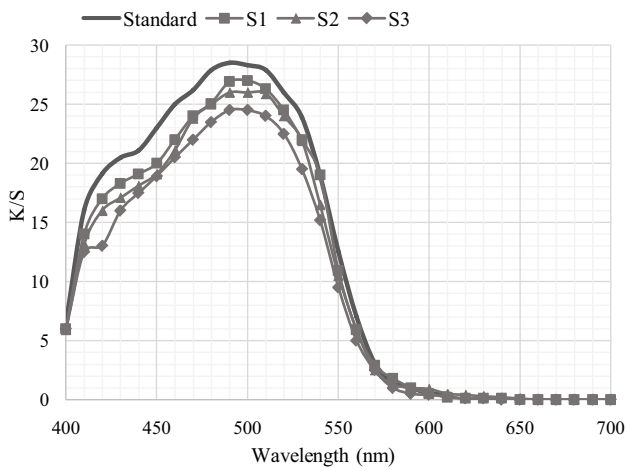


Fig. 11. Graph of wavelength plotted against K/S of fabrics dyed with C.I Reactive Orange 122.

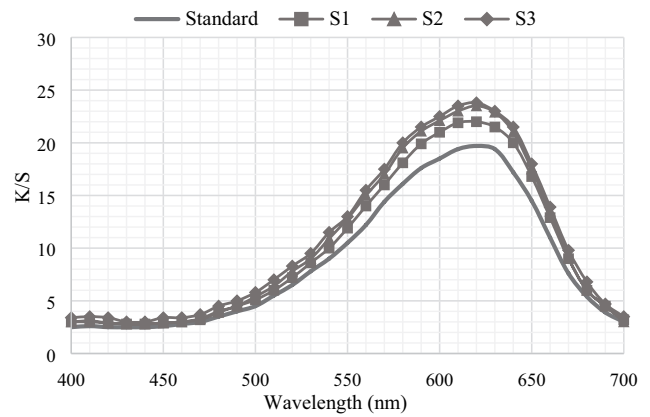


Fig. 13. Graph of wavelength plotted against K/S of fabrics dyed with C.I Reactive Blue 221.

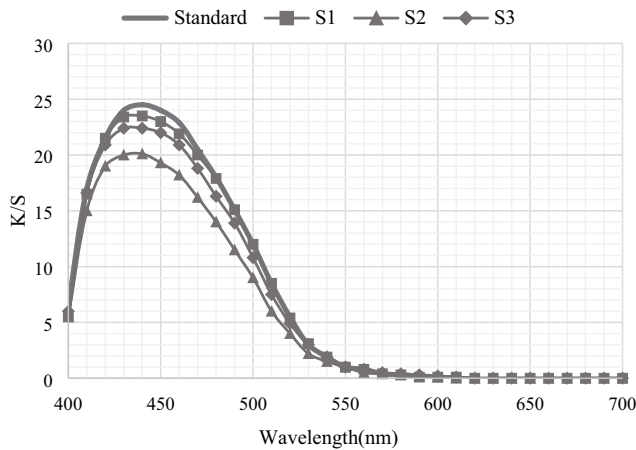


Fig. 12. Graph of wavelength plotted against K/S of fabrics dyed with C.I Reactive Yellow 84.

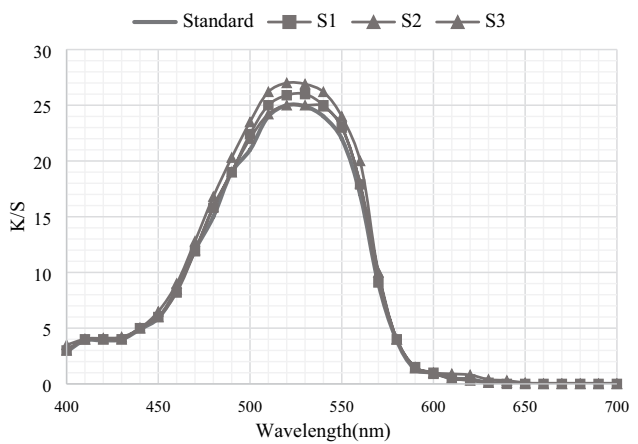


Fig. 14. Graph of wavelength plotted against K/S of fabrics dyed with C.I Reactive Red 223.

of Reactive Blue 221 and Red 223. Wash-off can be done successfully by recycling wastewater which is indicated by color difference results. Therefore, Fenton treatment proves to be an effective method for the recycling of wastewater in cotton fabrics' wash-off process without affecting its quality.

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