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Reuse of the treated textile wastewater and membrane brine in the wet textile processes: distorting effects on the cotton fabric

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ABSTRACT

In this work, a pilot plant consisting of granular activated carbon (GAC), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) processes was tested for the reuse of biologically treated textile wastewater (BTWW). The treated wastewaters were reused in the dyeing and bleaching of cotton fabric. To achieve zero discharge, NF brine was treated by lime soda then reused in the dyeing process as salt liquor. The main polluting parameters monitored in the posttreatments were: particle size distribution, divalent hardness ions, conductivity, soluble chemical oxygen demand (SCOD), and color. To determine the effects of reuse on the wet textile processes, color difference (d*E*), Berger whiteness degree, scanned electron microscopy images, and elemental analysis were followed on the treated cotton fabric. Application of UF, following GAC treatment, reduced SCOD concentration from 195 to 86 mg L^{-1}. Over 99% Pt–Co and $color_{540}$ were removed by NF/RO membrane treatments. Both NF and RO permeates were found to be suitable for reuse. After treatment of NF brine by lime-soda softening process, 89.5 and 93.5% reductions in total hardness and SCOD were achieved, respectively. The reuse of softened NF brine was found to be acceptable, since d*E* values in dyeing process were less than 1.0.

Keywords: Advanced treatment; Brine; Membrane filtration; Reuse; Salt recovery; Textile wastewater

1. Introduction

Textile industry, which mainly consists of washing, scouring, bleaching, mercerizing, dyeing, and finishing processes, not only demands large amount of water (9 trillion gallons/year), but also uses large amount of various hazardous chemicals, such as many different dyes, sulfite, metals, various organic halogens, waxes, sizes, anti-static agents, electrolytes, solvents, enzymes, formaldehyde, surfactants, phenols, softening, biocides, and lots of other additives. Many of these toxic chemicals remain in effluent after biological or chemical treatment. As a result of this, textile wastewaters may be toxic even after the biological and chemical treatment [1]. Today, strict environmental regulations

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[2,3] for the control of different hazardous compounds in textile wastewater and also water shortages in arid area have forced the textile industry to use new technologies and develop new cost effective treatment approaches for the recycling of water from waste stream.

In order to meet discharge requirements, textile wastewaters are usually treated by chemical and activated sludge biological treatment processes [4,5]. However, effluent of textile wastewater secondary treatment plant needs further treatments for reuse [6–8]. Residual soluble chemical oxygen demand (SCOD), color, monovalent, and divalent ions in the secondary treated textile wastewater are some important parameters restricting the reuse of treated wastewater in the textile wet processes. In recent years, many different advanced treatment methods, such as ozonation [9,10], photocatalytic oxidation [11], Fenton's reagent [12], electrochemical treatment [13], adsorption [14,15], coagulation [16], membrane separation processes [17–20], and their different combinations [21–24], have been attempted for the removal of remaining pollutants in the biologically treated textile wastewater. Currently, membrane filtration processes are the only known advanced treatment methods to remove remaining hazardous pollutants from the effluent of textile wastewater treatment plants [17,25]. However, the brine stream, generated from the membrane process, contains a wide variety of concentrated toxic organic and inorganic dissolved solids with total dissolved solid concentration up to several thousand mg L^{-1}. Most of the large plant operations that are not permitted to dispose the saline and toxic membrane reject stream are seeking for efficient brine recovery technologies [26–28]. Lagoon evaporation has usually been used to decrease the volume of the brine, allowing the remaining concentrate to be handled as a solid [8,29]. This practice is limited by its large site requirements and is only practicable in arid regions. Consequently, alternate brine treatment technologies for zero liquid discharge are being investigated, which include freeze concentration, brackish water reverse osmosis (RO)-evaporation system, membrane distillation/crystallization [30,31], and hypochlorite generation [32] from the brine stream. However, considering economic and energetic terms, the cost of these systems is typically much higher than that of implementation of a membrane treatment facility [8]. Thus, in recent years, some works were attempted to reuse salt solution as dyeing process liquor to handle toxic brine stream [25,27,28,33].

The treatment requirement for reuse may change from process to process depending on the key pollutants in each process, such as iron species in bleaching process, hardness in dyeing process, and color in washing and scouring processes. Thus, general process water standards should be developed for each process to reduce treatment cost of water recovery systems and enhance environmental and economical performance of the textile factories. There are only few studies in terms of effects of water pollutions on the quality of textile wet process [33,34]. In this view, still many investigations should be done to determine the level of organic and inorganic pollutants in the reclaimed water for each wet process without deforming the final textile product quality. In this work, a pilot plant, consisting of adsorption, coagulation, oxidation, and membrane separation processes, was investigated to find out cost-effective advanced treatment method for each process. The treated wastewaters, taken from each treatment method, were reused in the dyeing, bleaching, washing, and rinsing processes, and changes in textile quality parameters were comparatively investigated for the determination of possible negative effects of reuse. The brine stream of membrane processes were treated by lime-soda softening method then reused in textile dyeing process for new clean production and zero discharge approaches in the cotton textile industry.

2. Material and methods

2.1. Analytical methodology

Malvern ZS90 nanosizer with particle size measurement range from 0.3 nm to 5 μm was used for the determination of particle size distribution (PSD). UV–vis spectrophotometer (Hach Lange DR 2500) was employed for recording UV–vis scan in the wavelength range of 200–600 nm, and also for measuring color as Pt–Co and absorbance value at 540 nm. General water quality parameters, such as pH, conductivity, sulfite, ammonia nitrogen, total chromium, oil and grease, total suspended solid (TSS), SCOD, and total hardness, were analyzed according to the respective methods outlined in the Standard Methods [35]. NaOCl (Merck, CAS N#: 7681-52-9) was used to obtain 0.3 mg L^{-1} residual chlorine in the feed wastewater for the control of bio-fouling on ultrafiltration (UF) membrane. Moreover, residual chlorine was measured by the standard iodometric titration method [35]. Scanned electron microscopy (SEM) images and elemental analysis of cotton fabric surface were performed using The Philips XL30 ESEM-FEG/EDAX high-resolution environmental scanning electron microscopy and microanalysis system.

2.2. Process review

In this study, the biologically treated textile wastewater (BTWW) originated from five cotton fabric plants were used as a feed wastewater of the pilot plant. The central biological treatment plant that consists of a balance, neutralization, activated sludge treats 9,000 m^3 d^{-1} wastewater. Bleaching, rinsing, washing, dyeing, and finishing processes are the main wastewater resources in all factories. Ion-exchanged underground water was used as process water in all wet processes in the factories. Characterization of effluent of biological treatment plant was given in Table 1.

2.3. Pilot plant

The pilot membrane treatment systems consisted of coagulation, sand filtration, granular activated carbon (GAC), UF, nanofiltration (NF), and RO units (Fig. 1). Wastewater was taken from the discharge point of the central biological treatment plant of five different cotton fabric dyeing and finishing plants. The pilot plant was operated under different conditions and treated wastewaters were taken after each process to reuse into washing, rinsing, dyeing, and bleaching of 100% cotton fabric. The brine stream of NF membrane was treated by lime-soda softening method to reuse as saline dyeing liquor. The chlorination of the feed was practiced for the control of bio-fouling on UF membrane. Antiscalant (Perma Treat PC-191, Nalco Company) was used at a concentration of 3 mg L^{-1} before NF and RO membrane units for the prevention of membrane clogging. Membrane specifications and operation conditions were given in Table 2.

2.4. Lime-soda softening of the NF brine

Lime and then soda (Na_2CO_3, CAS N#: 497-19-8) were added to the NF brine. As a result of lime addition, calcium and magnesium were removed from wastewater at pH > 10 by precipitating as $CaCO_3$ and $Mg(OH)_2$. Thus, lime-soda softening was studied at pH 11 by adding appropriate amounts of $Ca(OH)_2$ (Merck CAS N#: 1305-62-0) and Na_2CO_3 (Merck). After lime-soda addition, solution was mixed for 2 min at 200 rpm and after 1 h settlement time, supernatant was taken and used for the dyeing of cotton fabric at laboratory scale.

Table 1
Chemical and physical properties of BTWW

Parameter	Value
pH	7.0–8.9
Conductivity (mS cm^{-1})	5.9–7.0
SCOD (mg L^{-1})	140–260
TSS (mg L^{-1})	50–90
Total hardness (mg L^{-1} as $CaCO_3$)	240–375
Sulfite (mg L^{-1})	0.78–1
Ammonia nitrogen (mg L^{-1})	1–2
Total chromium (mg L^{-1})	<0.03
Oil and grease (mg L^{-1})	3–5
Color (Pt–Co)	400–700

2.5. Quality parameters of treated cotton fabric

Treated textile wastewater types were taken from the pilot treatment and used for the textile processes of bleaching, dyeing, and rinsing of 100% cotton plain-woven fabric. As applied in the cotton textile industry, a mixture of 25 mg L^{-1} H_2O_2 (35%, Merck, CAS N#: 8007-30-5) and 25 mg L^{-1} NaOH (Merck, CAS N#: 1310-73-2) was used at 95°C for the bleaching of 100% cotton fabric. After 45 min of bleaching time, the treated fabric was rinsed for 10 min with 1 mg L^{-1} acetic acid solution and then washed, rinsed, and dried at room temperature. Dystar Remazol Yellow RR, Remazol Red RR, Remazol Blue RR dyes were used for the light green and dark green reactive dyeing owing to their extensively usage in textile industry. These dyes are composed of 50–60% dyestuff, 30–40% inorganic salt, and 5% other additives. Remazol Yellow RR and Remazol Red RR are anionic dyes. No halogens or heavy metals are included in Remazol Blue RR. Their detailed structures and maximum absorbance values are given in Table 3. Bleached 100% cotton fabric was dyed for 1 h at 60°C to obtain light and dark green colors according to the recipes given in Table 4.

Prior to the Berger whiteness and color difference measurements, fabric samples were conditioned for 24 h under conditions of 20°C and 65% relative humidity. To evaluate the effect of treated wastewater reuse on the bleaching and dyeing processes, Berger whiteness and color difference values were measured using a DATACOLOR 600 spectrophotometer, under D65 light, according to ISO 105 Standard Methods [36]. Berger whiteness degree was calculated by the formula as given below.

$$W_{\text{Berger}} = Y + a \cdot Z - b \cdot X \quad (1)$$

where X, Y, and Z are CIE tristimulus values and numerical parameters for 2° and 10° observers. Numerical parameters of "a" is 3.440 and 3.895; "b" is 3.448 and 3.904 for 2° and 10° observers, respectively [37].

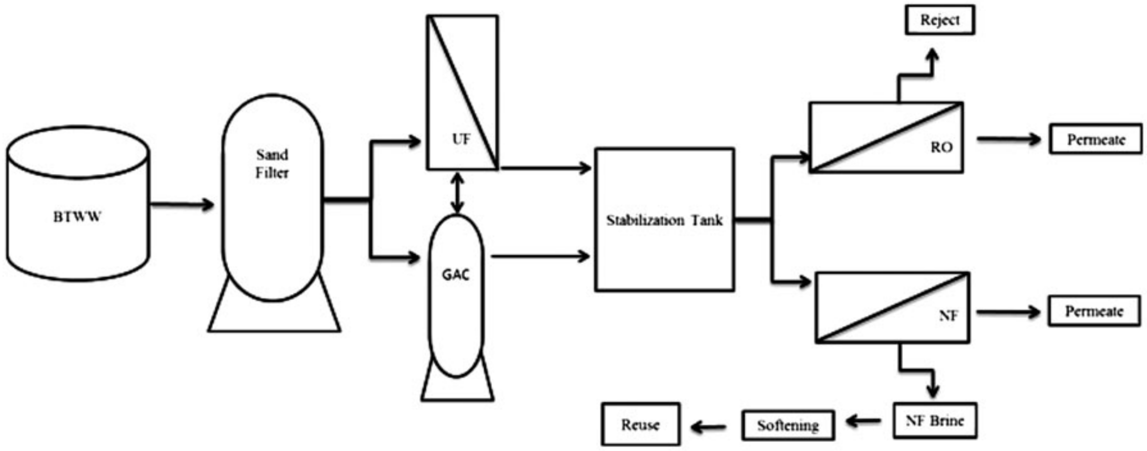

Fig. 1. Scheme of pilot membrane treatment system.

Table 2
Specifications of membranes used in UF, NF, and RO processes

	Ultrafiltration	Nanofiltration	Reverse osmosis
Model	dizzer® S 0.9 MB 6.0	Filmtech NF90-4040	Filmtech BW30-4040
Material	Polyvinylidene fluoride (PVDF)	Polyamide thin film composite	Polyamide
Active area	2 × 6 m^2	3 × 7.6 m^2	2 × 7.6 m^2
Flux ($L(m^2 h)^{-1}$)	30	33	49
Operating pressure (bar)	2.5r	8	17
Feed flow rate ($m^3 h^{-1}$)	0–6	3.6	2.5
Salt rejection (%)	–	>97	>99
Antifouling agent	Chlorine	Antiscalant	Antiscalant
Maximum feed SDI	–	5	5

Table 3
Designation of dyes (Dystar) used for dyeing process of cotton fabric

Color index name	Chromophore group	λ_{max} (nm)	Reactive anchor system	CAS N#
Remazol Yellow RR	Monoazo	420	Vinylsulphonyl (VS)	183185-50-4
Remazol Red RR	Monoazo	540	Vinylsulphonyl (VS) Monohalogentriazine (MHT)	183185-49-1
Remazol Blue RR	Anthraquinone	590	Vinylsulphonyl (VS)	183185-48-0

Table 4
Composition of light and dark green reactive dyeing solutions

	Light green	Dark green
Yellow RR (%)	0.25	1
Red RR (%)	0.01	0.04
Blue RR (%)	0.15	0.6
NaOH (48 Be°) (mlL^{-1})	50	90
NaCl ($g L^{-1}$)	25	60
Na_2CO_3 ($g L^{-1}$)	10	10
Sequestering agent ($g L^{-1}$)	0.3	0.3

The d*E* values of specimen fabrics were taken directly from the printout of color measurement device. d*E* value was originally calculated based on CIELAB color measurement system. Components of the color measurement system were positioned on a three-dimensional space. The lightness of the color (L^* = 0 yields black and L^* = 100 indicates diffuse white), position of the color in color space between red/magenta and green (a^*, negative values indicate green while positive values indicate magenta), and position of the color in color space between yellow and blue (b^*, negative values indicate blue and positive values indicate yellow). Color value differences, (Δ) of L^*, a^*, and b^* between sample color and reference color were calculated as:

$$\Delta L^* = L^*\text{sample} - L^*\text{reference} \quad (2)$$

$$\Delta a^* = a^*\text{sample} - a^*\text{reference} \quad (3)$$

$$\Delta b^* = b^*\text{sample} - b^*\text{reference} \quad (4)$$

Calculated ΔL^*, Δa^*, and Δb^* values were used to calculate total color difference of $\Delta \text{E}, (\text{d}E)$ as follows;

$$d\text{E}(\Delta E) = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$$

3. Results and discussion

3.1. Treatment performance of pilot plant

In this study, the preliminary experiments (conductivity, color, total hardness, pH, COD, etc.) were performed to investigate the performance of GAC, UF, and UF- and GAC-combined system as pretreatment of NF/RO units.

It is known that GAC adsorption is efficient at low pH values [38]. Therefore, pH of BTWW was adjusted to 6.8 during operation of the pilot plant. The treated wastewater was used for the dyeing and bleaching of 100% cotton fabric as applied in the cotton textile factory. Process water and biologically treated wastewater were used as benchmark for comparison.

Fig. 2 represents PSDs of different treatment processes. Two different PSDs (250 and 1,000 nm) were observed in the sand-filtered BTWW. After GAC adsorption, average PSD of BTWW decreased from 980 to 480 nm. Average PSD was found to be 230 nm in the UF- and GAC-combined system treated wastewater; however, average pore size of UF membrane was around 20 nm. Deposited particles on the GAC will form new granular particles by mutual combination with organic compounds and smaller particles [39–41]. In a similar manner, generally, mean particle size in the UF and NF permeate is larger than the pore size of related membranes [42,43]. Thus, enlargement of PSD after UF- and GAC-combined system treatment was attributed to the aggregation of some small organic and inorganic particles in the UF permeate and GAC surface.

SCOD concentration of biologically treated wastewater decreased from 195 to 86 mg L^{-1} by GAC treatment. After UF process, color and SCOD removal efficiencies were found to be insignificant (<3%) (Table 5). The pilot plant was operated to investigate the performance of the UF and GAC-combined treatment option. Chlorination of the feed until remaining 0.3 mg L^{-1} residual-free chlorine obtained in wastewater was recommended for the control of bio-fouling on the surface of UF membranes [44]. This condition of 0.3 mg L^{-1} residual chlorine was also followed before UF unit. Upon application of prechlorination followed by UF process, substantial increases as from 56 to 88% for Pt–Co and 61 to 98% for color_{540} removals were attained (Table 5 and Fig. 3). Konsowa and colleagues [45] reported that preozonation enhanced the dye removal performance of GAC process and the reason of which was attributed to the mineralization of the dye molecules by ozone. In a similar manner, also in this study, the enhanced color removal after prechlorination could be explained by the oxidation of the coloring organics by free chlorine.

Second treatment approach was the investigation of the role of RO and NF applications followed by prechlorination and UF- and GAC-combined treatment scheme. Fouling capacity of wastewater in NF/RO systems is characterized by the silt density index (SDI). NF/RO system requires SDI lower than 5 given by membrane supplier. Successful application of UF membranes in decreasing SDI to very low levels as SDI < 1.5 has been reported by Ciardelli and colleagues [46]. Therefore, an increase in SDI higher than 5 was not expected before NF and RO membranes treatment [28]. Both NF and RO permeates were found to be efficient to meet the reuse standards [18,47]. Total hardness level into both NF and RO permeates were found to be under 5 mg L^{-1} as $CaCO_3$ (APHA STM, 1998). Over 99% Pt–Co and color_{540} were removed after NF and RO membrane treatments. SCOD concentrations in RO and NF permeates were found to be 10 and 25 mg L^{-1}, respectively.

3.2. Effects of reuse on the textile dyeing and bleaching processes

Residual SCOD, color, and hardness are the main problems avoiding wastewater reuse. Most of the remaining SCOD present in the biologically treated wastewater can be considered nonbiodegredable, thus known as refractory SCOD. In the textile wastewater, refractory organics include nonbiodegredable dyes, surfactants, phenols, biocides, natural polymers, coating additives, and cotton derivatives. Color in the effluent of biological treatment plant is generally resulted from remaining refractory organic dyes. Eighty-five percent reactive, 10% disperse, and 5% some other colored dyes are used in all cotton factories. Thus, in this work, reactive dyes were selected

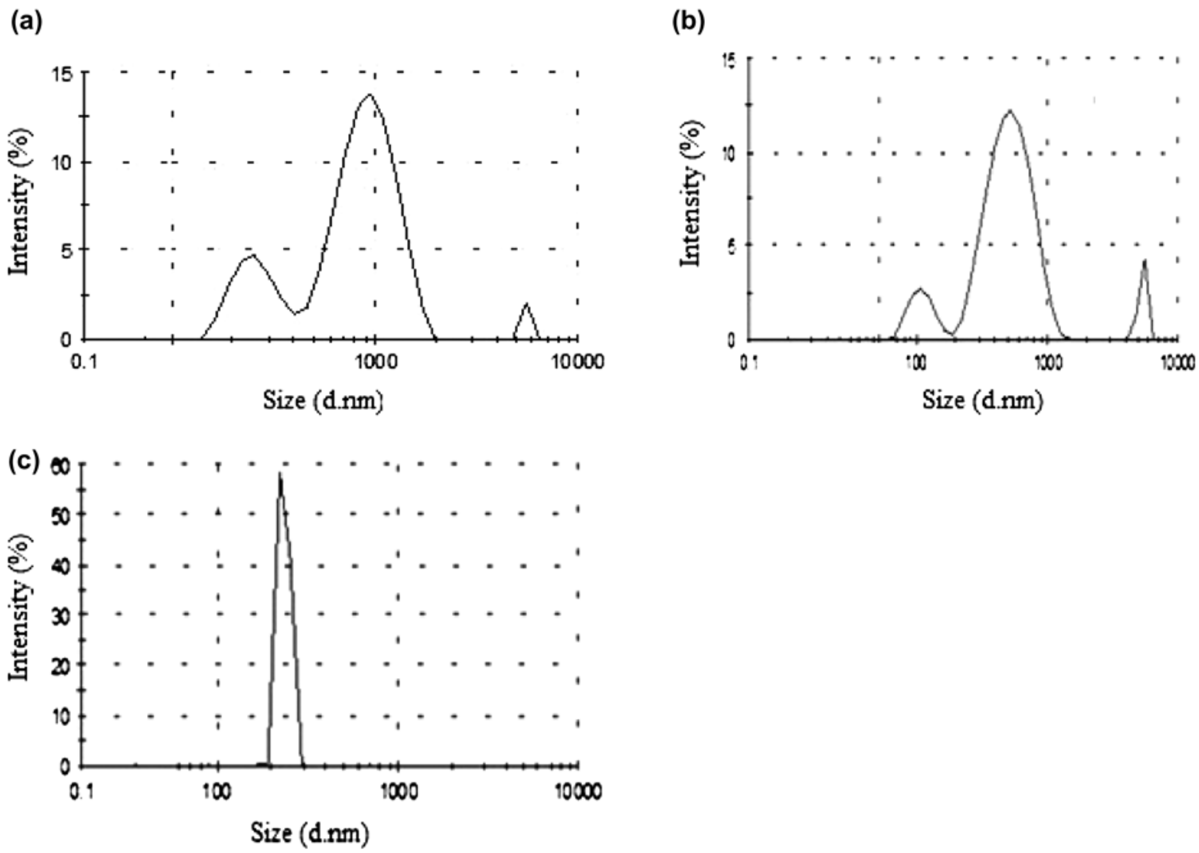

Fig. 2. PSD by intensity of: (a) biologically treated wastewater after sand filtration, (b) GAC adsorption, and (c) UF and GAC combined treatment systems (pH = 6.8 and conductivity = 6.9 mS cm^{-1}).

Table 5
The performance of different advanced treatment configurations of BTWW after adjusting pH to 6.8

	pH	Conductivity (mS cm^{-1})	SCOD (mg L^{-1})	Color (Pt–Co)	$Color_{540}$ (cm^{-1})	Total hardness (mg L^{-1})	TSS (mg L^{-1})
Raw water	8.2	6.9	195	462	0.14	255	84
GAC	6.8	6.9	86.4	203	0.08	215	20
UF	6.8	6.9	190	455	0.13	–	–
UF and GAC	6.8	6.8	65	56	0.02	220	NAD[b]
NF permeate	5.5	0.51	25	3.2	0.001	<DL[a]	–
RO permeate	5.6	0.06	10	0	0.008	<DL[a]	–
Softened NF brine	6.9	17.06	112	78	0.05	712	68

[a]DL: Detection limit.
[b]NAD: Not available data.

for the evaluation of the effects of reuse in the dyeing process. The treated samples, taken after each process in the pilot plant, were used in the dyeing and bleaching of cotton fabric samples, and process water was used as a benchmark. Figs. 4 and 5 show the effects of reuse of treated wastewaters on the dyeing quality. Following the reuse of NF and RO treated wastewaters in dyeing process, color was observed to be uniform and color difference (d*E*) found to be less than 1.0 indicating that the treated wastewaters could express no adverse effects on the dyeing process. Reuse of UF- and GAC-combined system treated and untreated

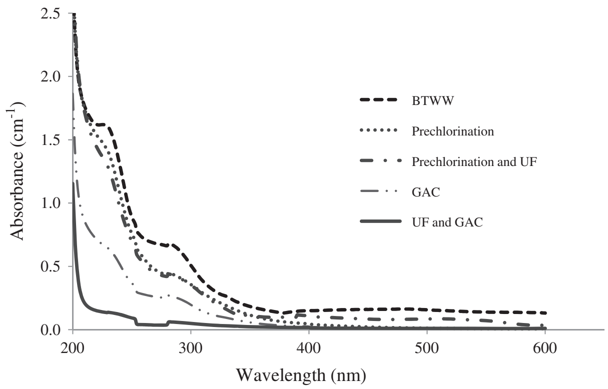

Fig. 3. UV–vis absorption spectra of treated wastewaters: (1) BTWW, (2) prechlorination, (3) prechlorination and UF, (4) GAC, and (5) UF and GAC (pH = 6.8, chlorination dose = 4 mg L^{-1}, and residual chlorine = 0.3 mg L^{-1}).

BTWW caused much lighter color on the cotton fabric, and d*E* values were found unacceptable as greater than 1.0. Lu et al. [48] also reported similar unexpected d*E* results for light color after using reclaimed wastewater. The result achieved using high colored BTWW improve that negative effect of reuse on the light color may be resulted from remaining color concentration. Moreover, SEM images (Fig. 6(a)–(f)) and SEM elemental analysis (Table 6) showed that precipitation of calcium and magnesium on the cotton surface could be the main reason of the adverse effect of the reuse. It could be concluded that NF or RO treatment with UF and/or GAC pretreatment schemes could be sufficient to reduce both SCOD and total hardness for the reuse of BTWW. This result reemphasized the observation made by Lu et al. [48] that membrane filtration is a feasible method for reusing of BTWW in a dye bath.

Due to the cost of treatment, instead of zero discharge, mixing treated waters with process water could also be a wastewater reuse strategy in the textile industries. Thus, in this work, the effect of different wastewater concentrations was also investigated using mixed-treated process waters at different ratios (100, 75, 50, and 25%). d*E* values showed that 25% diluted UF- and GAC-combined system treated wastewaters could be reused in the dyeing process without distorting dyeing quality (Figs. 4 and 5).

Some textile processes, such as prewashing, bleaching, and mercerizing, may not need to use softened water, and there is no water quality standard for each process. The main purpose of the bleaching process is to transform natural color of raw cotton fabric to a standard white color. Berger whiteness is the only requested quality parameter in the process. Treated wastewaters were used in the bleaching process to evaluate the effect of reuse and hardness ions on the bleaching process. Berger whiteness after reuse was shown in Fig. 7. In contrast, dyeing process, probably due to the white color of $CaCO_3$ precipitation on the surface, reuse of BTWW and GAC-treated wastewaters increased the whiteness of bleached cotton fabrics. Results reflect the fact that organic and hardness levels in BTWW do not have adverse effect on the whiteness level in the bleaching

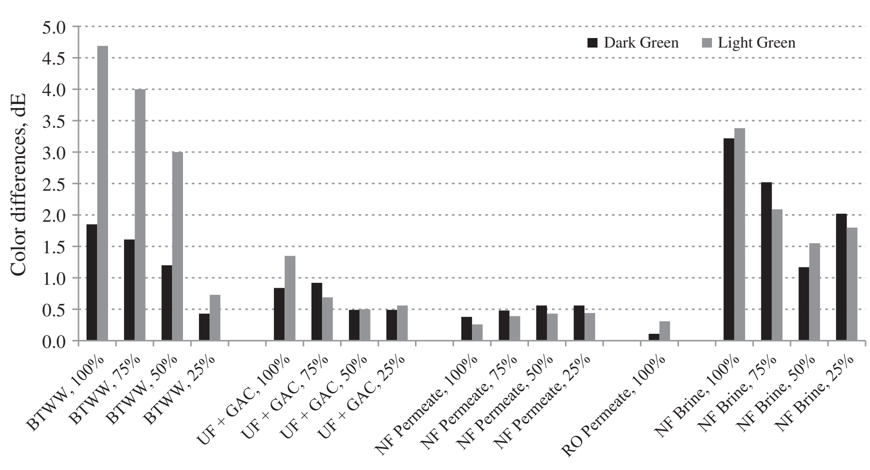

Fig. 4. Color difference, d*E*, values of cotton fabric dyed with mix of treated wastewater and process water (dyeing temperature, T_d = 60°C, NaCl concentration dyeing process = 25 g L^{-1} for light green and 60 g L^{-1} for dark green, Na_2CO_3 = 10 g L^{-1}) (Percentage values that are shown in Fig. 4 belong to the ratio of treated wastewater, deficient percentage is fulfilled with process water).

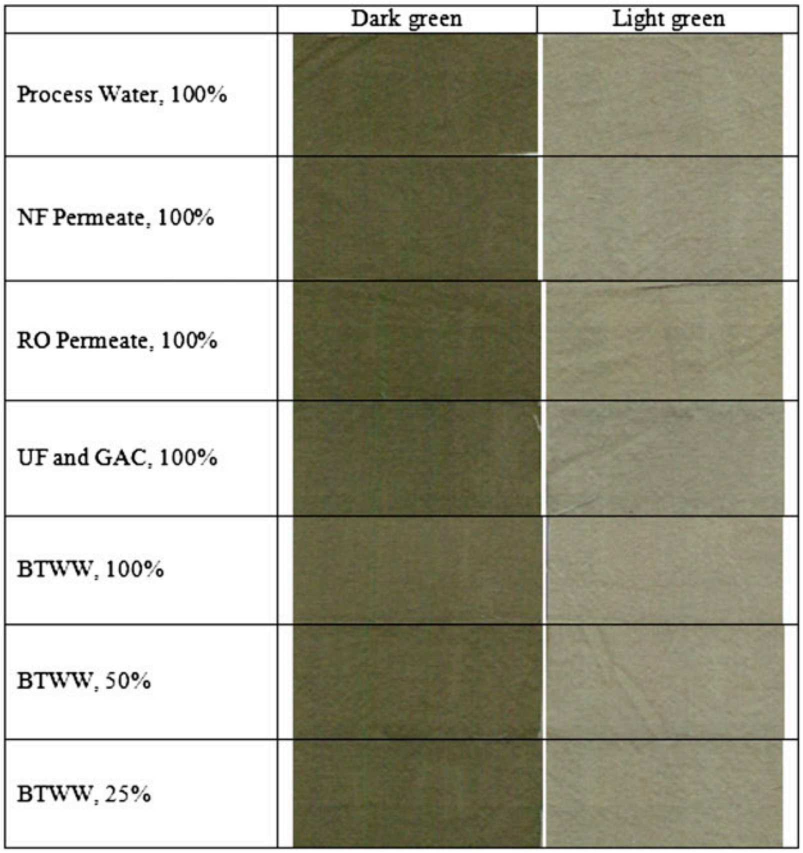

Fig. 5. Dark green and light green color images (Please check the online version for colored images.) of the cotton fabrics after dyeing (dyeing temperature, T_d = 60˚C, NaCl concentration dyeing process = 25 g L^{-1} for light green and 60 g L^{-1} for dark green, Na_2CO_3 = 10 g L^{-1}) with mix of treated wastewater and process water (Percentage values that are shown in Fig. 5 belong to the ratio of treated waste water, deficient percentage is fulfilled with process water).

process; however, scaling on the bleaching equipment should also be considered.

3.3. Softening of NF brine and reuse in reactive dyeing

Total dyes consumed in the cotton textile dyeing processes in five textile plants are composed of 85% reactive dyes. Reactive dyes are not biodegradable and also have poor fixation rate on the cotton fabric. Thus, the effluents of the textile industry could be mainly composed of reactive dyes. On the other hand, due to the poor fixation rate of reactive dyes, reactive dyeing of cotton requires excessive amounts of salt (30–100 g L^{-1} NaCl). Salts are not removed by secondary treatment systems and remain in the effluent of biologically treated wastewaters. In arid or semi-arid regions, discharging of saline wastewaters is not permitted due to its adverse toxic effects [49]. According to the BTWW, membrane brine streams contain around three times higher salinity, and thus textile industries are focusing on solving this problem for the reuse of BTWW. In this study, lime-soda softening method (1,500 mg L^{-1} lime and 500 mg L^{-1} Na_2CO_3) was used for the treatment of NF brine. Following lime-soda softening process, 89.5 and 93.5% reductions in total hardness and SCOD were attained, respectively. As given in material and methods part, 25 and 60 g L^{-1} salt (NaCl), used for light and dark green dyeing, gives 39 and 94 mS cm^{-1} conductivity levels, respectively. Thus, prior to the reuse of wastewater in dyeing process appropriate amount of salt was added into the BTWW and softened NF brine (17 mS cm^{-1}) to get same electrolyte condition as applied in dyeing process (Table 4).

The influences of the softened brine and its dilutions on the color difference values (dE) of light and dark green dyed samples were investigated (Fig. 8). The use of treated brine for both light and dark green fabric samples were found to be acceptable, since their determined dE values were less than 1.0. Use of NF

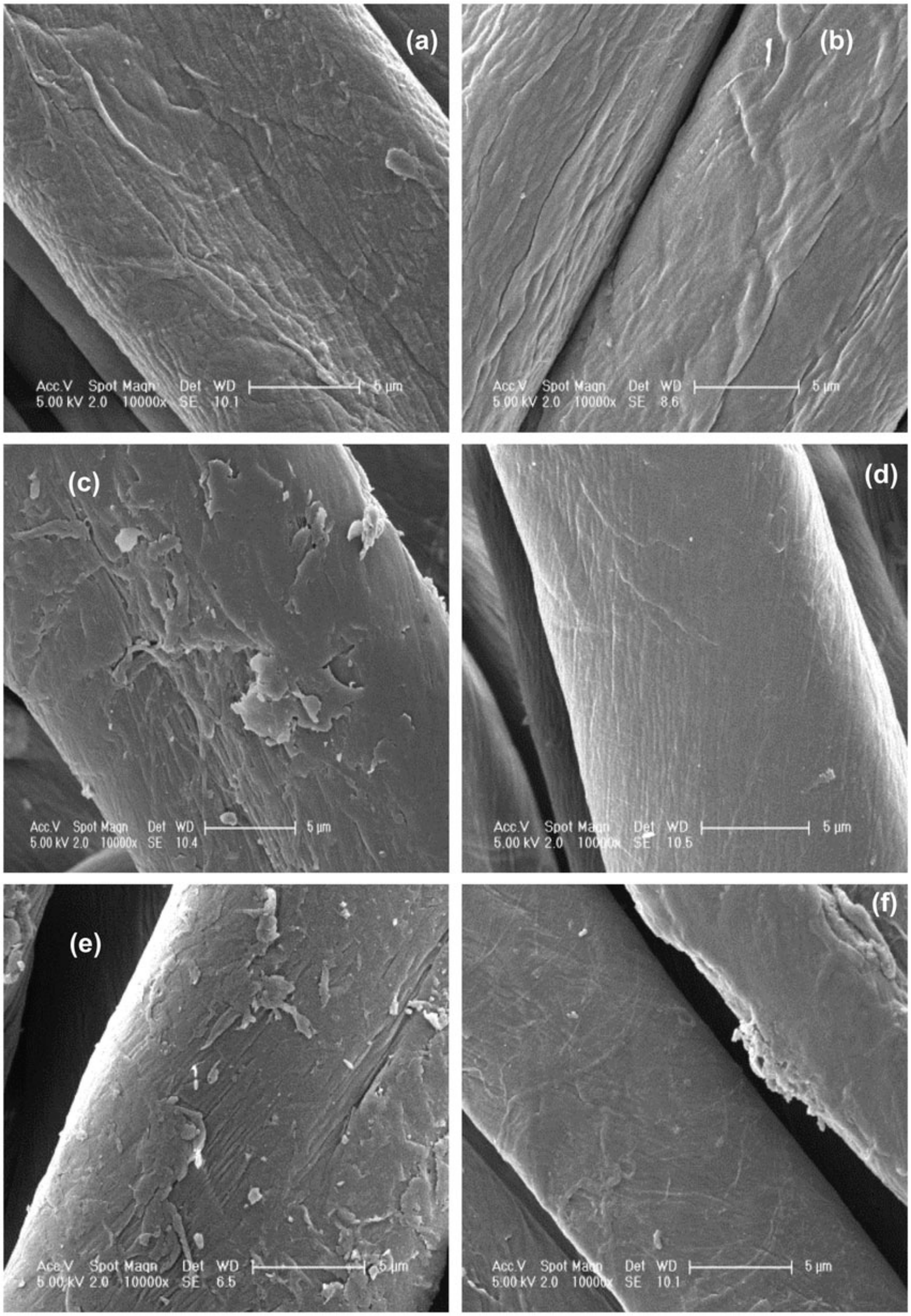

Fig. 6. SEM images of cotton fabric dyed with treated wastewaters: (a) process water, (b) NF permeate, (c) BTWW, (d) UF and GAC, (e) NF brine, and (f) softened NF brine 100%.

Table 6
SEM elemental analysis of the surface of the cotton fabrics dyed using effluent of different treatment configurations (dyeing temperature, T_d = 60°C, dyeing time = 1 h)

	NF effluent 100%	Softened NF brine 100%	Process water	Raw water	GAC effluent 100%
C	76.08	74.00	72.88	74.43	74.53
O	21.13	24.05	25.36	22.92	22.52
Mg	0.11	0.15	0.12	0.17	0.19
Ca	0.10	0.14	0.09	0.20	0.13
Fe	0.11	0.27	0.22	0.16	0.40

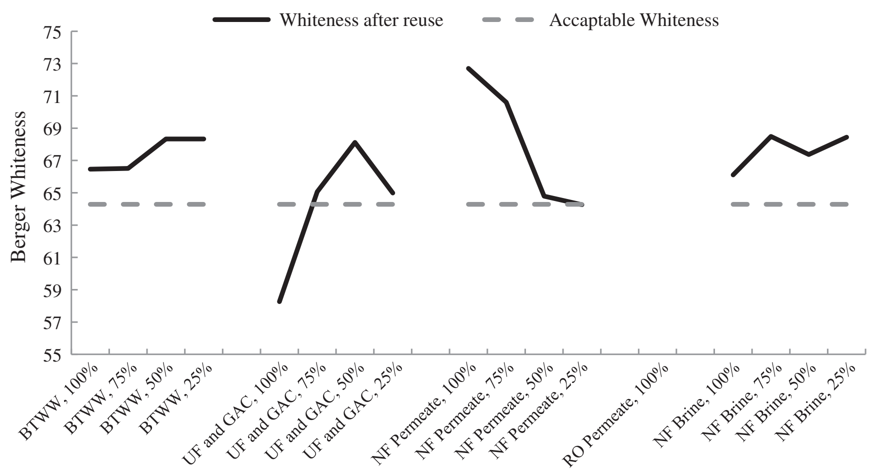

Fig. 7. Effect of treated wastewaters on the bleaching quality (bleaching time = 45 min, bleaching temperature T_b = 95°C, H_2O_2 = 25 mg L^{-1}, NaOH = 25 mg L^{-1}).

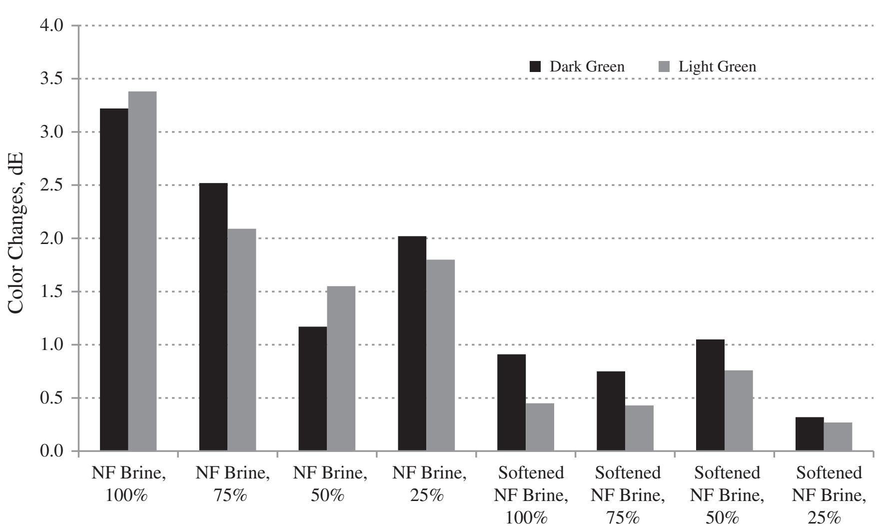

Fig. 8. Effect of softened NF brine on the reactive dyeing of 100% cotton fabric (pH of treated wastewater = 6.9, dyeing temperature, T_d = 60, dyeing time = 1 h, $Ca(OH)_2$ = 1,500 mg L^{-1}, and Na_2CO_3 = 500 mg L^{-1}).

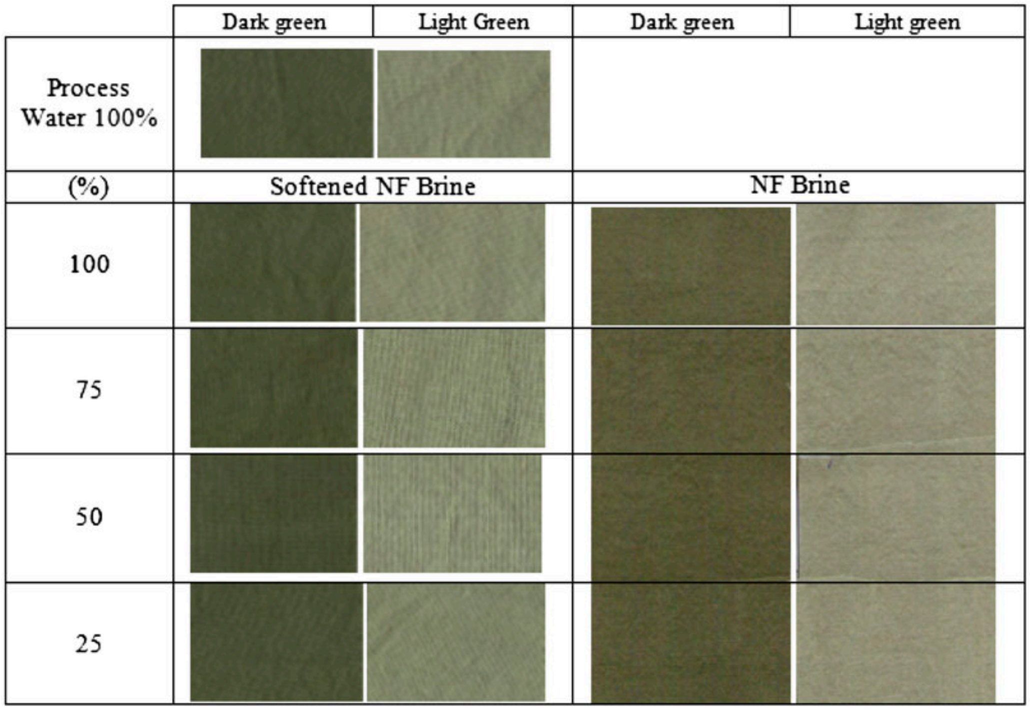

Fig. 9. Dark green and light green color images of the cotton fabrics after dyeing with mix of softened NF Brine and NF Brine treated wastewater and process water (dyeing temperature, T_d = 60°C, NaCl concentration dyeing process = 25 g L^{-1} for light green and 60 g L^{-1} for dark green, and Na_2CO_3 concentration = 10 g L^{-1}) (Percentage values that are shown in Fig. 5 belong to the ratio of treated waste water, deficient percentage is fulfilled with process water).

brine and their dilutions caused undesirable level of color differences (dE > 1.0), for both light and dark colors. Shu et al. reported that aggregation number would increase as the dye concentrated in the membrane brine [50]. Also, it is well known that with high concentration, either dyes or salts promote dye aggregations [51]. Otherwise, dye aggregation may be promoted by organic matter/ionic group content [52]. Results show that characterization of organic matter and different ion groups may have affected by aggregation, and thus dyeing efficiency. However, further investigation should be done to explain the effects of remaining organics and ion groups on the dyeing process. The enhancement of water quality of NF brine by lime-soda softening, visible color, and dE values (Figs. 8 and 9) are acceptable in the dyeing process. However, Mg^{2+} and Ca^{2+} concentrations on the dyed fabric samples increased after reuse of the softened NF brine (Fig. 6(f) and Table 6).

4. Conclusion

In this work, the major purpose was to test the applicability of GAC, UF, NF, and RO processes for the reuse of biologically treated textile wastewater. The treated wastewaters were further used for the dyeing and bleaching of cotton fabric. As expected, both NF and RO permeates (75% of textile wastewater) could be used directly in the textile processes. Otherwise, the study demonstrates that zero discharge may be possible in the textile industry since the softened NF brine (25% of textile wastewater) reused in the dyeing process as salt liquor. Based on the experimental results, the following points could be presented as conclusions:

- Application of GAC decreased PSD of BTWW from 980 to 480 nm and to 230 nm with UF pretreatment. Using of prechlorination following UF enhanced the treatment efficiency of GAC process and 56% Pt–Co and 61% $Color_{540}$ removals in GAC process increased to 88 and 98%, respectively. SCOD level in BTWW decreased from 195 mg L^{-1} to 86 mg L^{-1} and 56 mg L^{-1} after GAC and "pre-chlorination, UF and GAC" combined treatment schemes, respectively. Thus, it could be concluded that UF is important pretreatment option before NF membrane process for successful control of the organic and inorganic fouling problems.

- The treated samples, taken after each process in the pilot plant, were used in the dyeing and bleaching of 100% cotton fabric. Concerning the d*E* color change values, SEM images, and elemental analysis of surface, NF and RO treated samples were found to be suitable for the dyeing process. In spite of low SCOD level (56 mg L^{-1}) in the UF and GAC treated water, high color changes (d*E* > 1.0) were observed on the cotton fabric after reuse, the reason of which was attributed to hardness precipitation on the cotton fabric. Otherwise, dilution of UF and GAC treated water by 50% was found to be tolerable for the reactive dyeing.
- Reuse of wastewaters did not affect the bleaching quality of cotton fabric and contrary to the dyeing process, Berger whiteness level increased after reuse. The reason of the enhanced bleaching quality could be attributed to the bleaching effect of H_2O_2 in the bleaching liquor and also precipitation of Ca^{2+} ions as $CaCO_3$ on the surface of the cotton fabric.
- NF brine was used in the dyeing process at different dilutions. Adverse effects of NF brine decreased with increasing dilution rate. NF brine was softened by lime-soda treatment method and treated NF brine was used in the reactive dyeing as salt liquor to achieve a cost-effective reuse system. SEM images and d*E* values showed that reuse of the softened NF brine displayed no adverse effects on the reactive dyeing. It could be concluded that the recovery of salt without using expensive brine treatment methods could make NF membrane reuse systems cost effective for zero discharge.

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