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ORIGINAL PAPER

Effective treatment of domestic sewage to reuse in textile dyeing and catalytic treatment of generated dye wastewater

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Abstract

Many industries face a critical sustainability issue due to increased freshwater scarcity for their water-intensive activities, especially textile industries. It is essential to find alternatives to freshwater resources to sustain industries, which can be achieved by reusing the efficiently treated domestic wastewater. This study aims to address the efficient treatment of domestic sewage for its reusability in textile dyeing applications and the treatment of generated dyeing wastewater for domestic purposes. Domestic sewage was efficiently treated using sequential microbial indigenous reactors combined with disinfection and sand filtration. The removal efficiency of chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammoniacal nitrogen (NH_4^+-N), total nitrogen (TN) and total organic carbon (TOC) were 93.8% ± 1.04, 94.9% ± 0.9, 93.8% ± 5.7, 86.3% ± 3.8 and 92.3% ± 2.1, respectively, achieved. The final treated sewage had COD and BOD of 16 ± 5.3 mg L^{-1} and 3.84 ± 1.26 mg L^{-1}. It was reused for the dye fabrication on a textile cloth and compared with the quality obtained using freshwater. The bursting strength value of both the dyed clothes remained the same (8.53 Kgs/sq.cm). The colour strength value of clothes dyed using treated water was higher than freshwater dyed clothes. Generated dye effluent was treated using advanced oxidation reactors followed by a bio-reactor. The removal efficiency was COD, 95.1% ± 0.58, BOD, 89.4% ± 3.02, TOC, 93.0% ± 1.82, TN, 76.7% ± 6.1 and NH_4^+-N, 58.4% ± 10.1. Thus, the study demonstrated the efficient treatment of domestic sewage for reusability in the textile dyeing process, followed by textile effluent treatment.

Keywords Cotton dyeing · Hydroxyl radical · Immobilised cell reactors · Textile process · Treated sewage

Introduction

India is likely to face twin edged problems to deal with dwindling freshwater availability and increasing wastewater generation in the coming decades. The exponential population, urbanisation and industrialisation rise rapidly, leading to severe water stress (Ram and Irfan 2021). India generates about 38,354 MLD of municipal sewage in significant cities, but only 30% of sewage is treated (Chaudhari et al. 2020; Thongam and Chaturvedi 2021). As freshwater becomes an increasingly scarce resource, industries have been advised by pollution control authorities in India to develop economically feasible reuse technologies for zero-liquid discharge (ZLD) (Singh et al. 2020). The textile industry is widely known to employ the most water-intensive industrial activities that rely only on groundwater (Fito et al. 2020; Kazancoglu et al. 2020; Mirza et al. 2020). It is estimated that the textile process consumes about 100–200 L of water to process one kilogram of fabric (Maqbool et al. 2020; Tayyab et al. 2020). So, the textile industry faces the challenge of freshwater availability and desperately needs an alternative source to sustain the sector (Bommavaram et al. 2020; Sachidhanandham and Periyasamy 2020; Haque et al. 2021). The treated sewage can be considered an alternative resource to groundwater in the textile dyeing process to save freshwater resources (Tak 2021). Researchers have not yet explored the reuse of treated domestic sewage in textile dyeing applications to the best of our knowledge. So, the present investigation was focused on the effective treatment of domestic

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sewage and reusing the treated sewage for dyeing on textile cotton cloth. The activated sludge process majorly treats domestic sewage due to its good biodegradability (Chen et al. 2020; Xing et al. 2021). The major disadvantage of this process is sludge production and the active microorganisms washed off in the treated water (Hallaji et al. 2019). It also has sludge disposal difficulties and low life of the treatment processes (Otieno et al. 2019). To overcome these demerits, effective treatments technologies for domestic sewage which can provide reusable water for textile dyeing processes are required. The proposed sequential immobilised microbiological reactors can fulfil the current requirement. Thus, the study's objective was to treat and reutilise the treated sewage for the dye fabrication on cotton cloth by the wet-processing method. In order to meet and verify with ground water dyeing process, the treated sewage used dyed cloths are compared using instrumental analysis. As the process consumes 100–200 L water per kg of fabric, the same amount of dye wastewater generated which is another serious environmental problem needs to be addressed. The generated the dye wastewater can contain the unused reactive dyes of about 10 to 50% w/v, detergents and organic pollutants accounting for chemical oxygen demand (COD) (Bidu et al. 2021). If the dye wastewater is discharged without treatment into water bodies, it can percolate into the land and contaminate groundwater sources which create fatal adverse effects on both humans and animals (Bharagava and Chowdhary 2018; Younas et al. 2021).

The various advanced oxidation processes (AOPs) used to treat the dye wastewater have poor COD removal efficiency with disadvantages and limitations. The AOPs of Fenton treatment have a short range of working pH(2–3.5) and sludge disposal difficulties (Wang and Tang 2021; Brillas 2022; Wang et al. 2022). The ozonation process can remove COD from 50 to 60% within 90 min, but it has the shortcoming of the release of amine compounds, high operating cost, low lifespan, formation of toxic by-products and poor efficiency in alkaline conditions (Katheresan et al. 2018; Nidheesh et al. 2020).

Hence, another objective of the present investigation was to develop a suitable treatment option for the treatment of dye wastewater generated by the dyeing process. This study proposes a combination of chemical and immobilised biological oxidation methods for dye wastewater treatment. The benefits of the treatment used in this study include no secondary pollutant emission, groundwater contamination avoided, recycling of treated wastewater, low cost, convenient to use, producing non-toxic by-products and do not require any sophisticated or expensive mechanical equipment.

This research article comprises the integrated work of three objectives, such as the treatment of domestic sewage by immobilised microbial oxidation reactors, comparison of reuse of treated sewage with freshwater for dye fabrication on cotton cloth along with its characterisation and, finally, the catalytic oxidation of dye wastewater generated in the dyeing process.

This research work was carried out in Environmental Science Lab, CSIR—Central Leather Research Institute (CLRI), Chennai, India, from Jan 2021 to Aug 2021.

Materials and methods

All the chemicals used in this study were analytical grades bought from S D Fine-chem Limited. The domestic sewage used in this study was collected from the residential colony located in Adyar (City: Chennai, State: Tamil Nadu), India.

Treatment of domestic sewage using immobilised microbial oxidation reactors

The objective and principles of all reactors used in this study are discussed in supplementary information in Section S1 with Fig. S1-S5, including actual reactor images. The brief descriptions of the reactors are given below.

Sequential oxic-anoxic reactor (SOAR)

The SOAR reactor was designed as detailed by Mahesh et al. 2017 with a working volume of 7.5 L. It consisted of three compartments, connected internally, and each compartment was filled with circle polypropylene to facilitate microbial growth on the inner and outer surfaces of the material. The total suspended solids (TSS) present in the sewage was separated and digested by microorganisms present in SOAR. The two reactors of SOAR-I and SOAR-II were used in the treatment scheme of domestic sewage.

Fluidised bio-bed oxidation reactor (FBBOR)

The FBBOR reactor was fabricated using the acrylic sheet with a total volume of 17 L and a working volume of 15 L in breadth 21 cm and height 42 cm. The FBBOR reactor consisted of carbon silica matrix (CSM) immobilised with bacterial cells. The CSM and immobilised CSM were prepared as reported in previous studies by Prabhakaran et al. (2021). The immobilised bacteria can metabolise the fragmented organics in dye wastewater into CO_2 and H_2O. The advantages of the FBBOR reactor are high bacterial population density, easy treated water separation and repeated continuous operation without new culture addition. It also has the advantages of plug flow operation by maintaining the immobilised cells as a stationary phase, small footprint requirement, high volume-based purification efficiency, amenability to scale up, high operation flexibility and a good prospect for large-scale application.

Packed bio-bed oxidation reactor (PBBOR)

The PBBOR reactor was fabricated in a poly vinyl chloride (PVC) pipe with a total capacity of 3000 ml and a working volume of 2500 ml. The bottom of the reactor (diameter 9 cm) was filled with coarse gravel (5 cm height) and medium gravel (5 cm height). Then it was filled with fine gravel to the height of 5 cm and packed with CSM of 600 μm size to the height of 40 cm. The air diffuser was placed at the bottom of the CSM bed to inject air at the rate of 0.6 kg cm^{-2} into the packed bed reactor. The flow of wastewater to be treated was in the counter-current mode.

Treatment of domestic sewage

The sewage was treated through the biological treatment method using sequential immobilised cell reactors as shown in Fig. 1a. It comprised the hydraulic scheme of SOAR-I, FBBOR, SOAR-II, PBBOR, disinfection using sodium hypochlorite (hypo) and sand filtration. The raw sewage was pumped to the SOAR-I to remove TSS, and then, the outlet from the SOAR-I reactor was treated by an FBBOR reactor for the biological oxidation of the organic pollutant. The FBBOR outlet was carried to SOAR-II to remove biological suspended solids; then, the SOAR-II outlet was further treated in a PBBOR reactor to oxidise the remaining organic contaminants. The PBBOR outlet was disinfected by adding hypo 10 mg L^{-1} and then sand filtered. This treatment was studied under continuous process with a feed of 10 times raw domestic sewage. The entire treatment was characterised by physicochemical parameters of pH, COD, biochemical oxygen demand (BOD), TSS, total dissolved solids (TDS), ammoniacal nitrogen (NH_4^+-N), total Kjeldahl nitrogen (TKN), sulphates, sulphide and iron were characterised with duplicates by following American Physical Health Association (APHA) standard methods (APHA 2017).

Reutilisation of treated sewage for dye processing with characterisation

Dyeing process on textile cloth

The reactive brilliant red K-2G dye was used for the dye fabrication study. The textile cotton cloth of dimensions 10 cm × 10 cm was taken for dye fabrication by the wet-processing method. It was done by a dyeing machine (Model HTAI/HT-220V/380V) for dye fabrication on cotton cloth using treated sewage. The dyeing process was started with bleaching of cotton cloth by adding hydrogen peroxide 2% (v/v). It whitened the cotton cloth material by removing the undesired natural colour compounds like chlorophyll, xanthophyll and carotene. Then Basopal FB PK (0.8% v/v) was added and heated up to 90 ℃ to remove residual peroxide from the bleached cloth. The cloth was mixed well with acetic acid (1% v/v) at pH4, and then the stabilising agent of Sirix SB (0.5% v/v) was added and washed for 20 min. Further, the reactive dye was added to water at pH6.5 with salt and caustic soda and then heated to 60 ℃. The fabricated dye cloth continued for shade running up to 30–180 min, followed by hot water wash at 90 ℃. Then the dyed fabric was washed at pH4 by adding acetic acid 1% (v/v) for 40 min and soaping with hot water at pH6. Finally, the dyed cloth was dried well to remove moisture. The dye fabrication on cotton cloth was also done using freshwater (groundwater) as a control for a comparison study with treated sewage used dyed fabric two times. The dyed cloth by treated sewage was

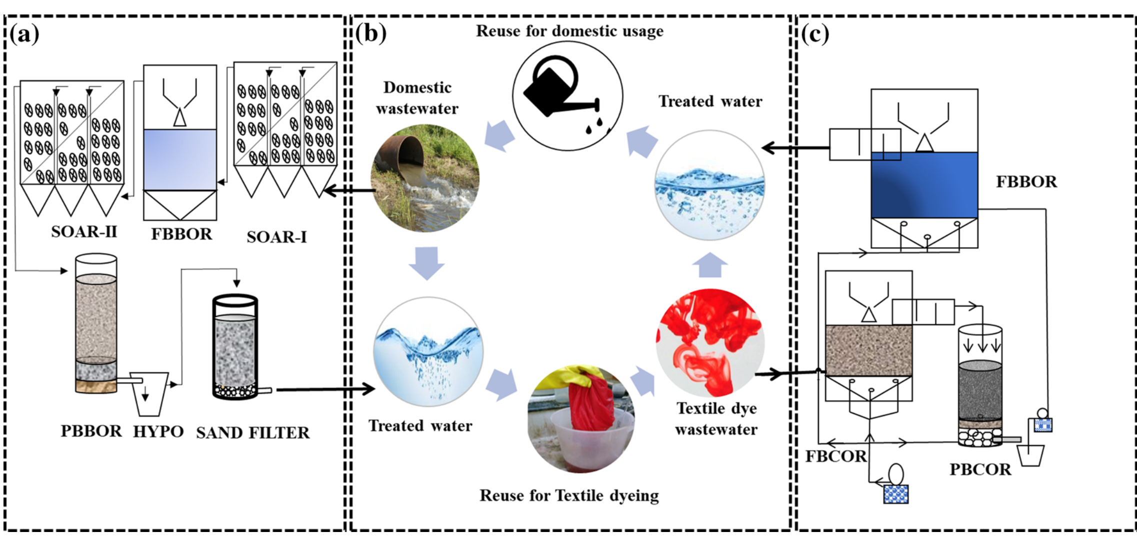

Fig. 1 Schematic diagram of domestic sewage treatment (**a**), reutilisation of the treated wastewaters (**b**) and treatment of generated dye wastewater

labelled as DCTS, and freshwater was labelled as DCFW (Fig. 2).

Instrumental characterisation of dyed cloths

The dyed cloths (DCFW and DCTS) were subjected to thermal stability in thermogravimetric analysis (TGA) using Q50 (V20.6 Build 30) instrument. It was done from 30 to 800 ℃ under the reduced nitrogen atmosphere at a temperature gradient of 20 ℃/min. The surface morphology of dyed cloths was analysed using field emission scanning electron microscopy (FESEM) (FEI Quanta-250). It was done by passing the fine beam of energised electrons to scan in a series of parallel tracks “spot by spot”. The energy-dispersive X-ray analysis (EDX) was carried out on dyed cloths using the instrument FEI Quanta FEG 2000 to determine the elemental composition. ATR-FTIR (attenuated total reflection–Fourier transform infrared spectroscopy) of model JASCO FTIR-4200 used for dyed cloths at the incident 2θ angle of 45° with a resolution of 4 cm^{-1} and scanning speed of 2 mm/sec.

Treatment of the dye wastewater using catalytic and bio-oxidation reactors

Fluidised bed catalytic oxidation reactor (FBCOR)

FBCOR reactor was fabricated similar to the geometry of FBBOR reactor, but they differed in catalyst function. The FBCOR is a chemical oxidation reactor with hydrogen peroxide (H_2O_2) and potassium permanganate ($KMnO_4$) used as oxidants to generate hydroxyl radicals in dye wastewater treatment. The CSM was used to sustain the hydroxyl radical production for the oxidation of organics in wastewater.

Packed bed catalytic oxidation reactor (PBCOR)

The PBCOR reactor was fabricated as illustrated by Natarajan et al. 2020. The PBCOR reactor was packed with CSM to generate hydroxyl radicals with molecular oxygen from atmospheric air at ambient conditions. The PBCOR is also a chemical oxidation reactor in which the hydroxyl radicals are employed for the catalytic oxidation of organic compounds present in dye wastewater. The catalytic oxidation reaction was initiated by small potassium permanganate and hydrogen peroxide doses. The biological oxidation reactor of FBBOR used in the dye wastewater treatment is similar to the structure and function used in the sewage treatment.

Treatment of dye wastewater and instrumental characterisation

The dye wastewater generated in dye processing was treated in combined chemical and sequential biological reactors of FBCOR, PBCOR and FBBOR. It was done in a continuous treatment process with four times feed of raw dye wastewaters. The ultraviolet–visible (UV–Vis) spectrum of untreated and treated dye wastewater was recorded using a CARY 100 spectrophotometer in the 200–800 nm wavelength range. The spectrophotometer of Varian Cary Eclipse was used with an excitation source from a Xe lamp in the wavelength range of 200–800 nm to the dye wastewater. The Fourier transform infrared spectrophotometer (FTIR) of Perkin-Elmer was used to study the dye wastewater. The rapid nitrogen gas drier was used to dry the dye wastewater at low temperatures (70 ℃) to avoid escaping any organic compound. The dried samples were mixed well with spectroscopy grade KBr (Merk, Darmstadt, Germany) in a mortar. Then it was pelletised with a thickness of 1 mm and diameter of 13 mm. The pellets were analysed in the wavenumber range of 4000 to 400 cm^{-1} with five scans to investigate the functional groups present in the dye wastewater. The entire dye wastewater treatment was characterised for physicochemical parameters of pH, ORP, COD, BOD, TDS, TOC, TN, NH_4^+-N, sulphate and chloride in duplicates by following the APHA standard methods.

Results and discussion

Characterisation and treatment of domestic sewage

The physicochemical characteristics of pH, oxidation reduction potential (ORP), COD, BOD, TDS, TSS, TKN, NH_4^+-N, hardness, iron, sulphate and sulphide of sewage are presented in Table 1. It was the average of parameters analysed ten times. The average total COD of the

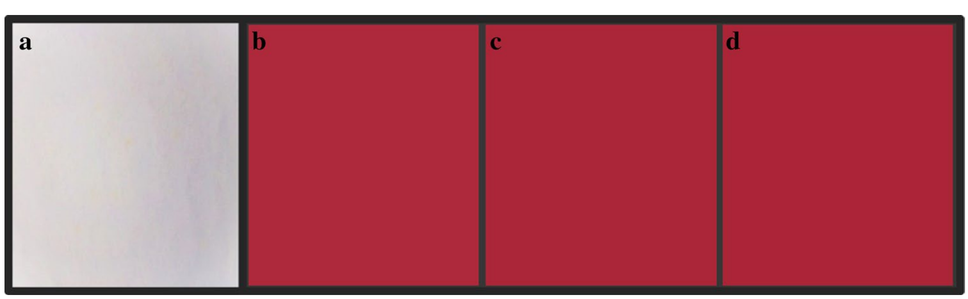

Fig. 2 Colour comparison of cotton cloth (**a**), DCFW (**b**) and DCTS (**c**, **d**)

wastewater was 336 ± 63.07 mg L^{-1} with soluble COD of 256 ± 56.03 mg L^{-1}. The average BOD of the water was 76.67 ± 15.74 mg L^{-1} with the BOD:COD ratio in the range of 0.29615-0.30469. The biodegradability of the sewage was very high so subjected to biological oxidation methods.

The sewage treatment scheme consisted of SOAR-I-FBBOR-SOAR-II-PBBOR. The optimisation studies of sewage treatment schemes are discussed in supplementary information section S2 with Fig. S6-S10. The hydraulic retention time (HRT) was fixed as 3 h for SOAR-I, 6 h for FBBOR, 3 h for SOAR-II and 1 h for PBBOR. The CSM dosage in the FBBOR reactor was fixed optimum as 30 g L^{-1} based on the pollution parameters removal efficiency. The PBBOR treated water was disinfected by adding sodium hypochlorite 10 mg L^{-1} and then sand filtered. The final treated water was characterised by physicochemical parameters and is shown in Table 1. The removal efficiency of sewage treatment under optimised conditions showed the COD, BOD, NH_4^+-N, total nitrogen (TN) and total organic carbon (TOC) by 93.9 ± 1.04%, 94.9% ± 0.9, 93.8% ± 5.7, 86.3% ± 3.8 and 92.3% ± 2.19 in w/v, respectively (Fig. 3).

Table 1 Characteristics of domestic sewage and treated domestic sewage

Parameters*	Domestic sewage	SD ±	Treated sewage	SD ±
pH	7.83	0.35	6.05	0.15
ORP mV	-119.4	42.03	53.2	20.6
TS	807.5	124.89	550	60.5
TDS	820	77.65	550	60.5
TSS	200.83	83.10	BDL	BDL
T.COD	336	63.07	16	5.3
S.COD	256	56.03	16	5.3
BOD	76.67	15.74	3.84	1.26
BOD:COD	0.2994	0.2809	0.24	0.2377
NH_4^+-N	44.1	15	2.72	2.5
TKN	21.84	5.58	5.04	1.5
TOC	164	21.20	12.51	4.12
TN	81.37	0.68	11.07	2.31
Sulphates	115.57	15.10	94.82	12.54
Sulfide	2.14	0.42	BDL	BDL
Iron μg L^{-1}	2.1	1.32	0.263	0.015
Chlorides	193.56	20.60	138.96	25.13
Hardness	250	44.35	160.5	26.51
TVCB(CFU/ml)	1×10^8	1×10^2	4×10^1	$1\times10^{0.5}$
TVCC(CFU/ml)	8×10^7	1×10^3	1×10^1	$1\times10^{0.5}$

*All are expressed in mg L^{-1} except pH if not specified

*All parameters are an average of 10 times

BDL below detection limit

Many researchers reported the treatment of sewage and reuse for irrigation purposes. Nasr et al. (2019) reported the removal efficiency of COD by 78%, BOD by 84% and TSS by 92% while using the compact biological unit comprising anaerobic, aerobic biofilm and settling. De Anda et al. (2018) used a septic tank, anaerobic up-flow filter and wetland with plants for sewage treatment and reported the removal of organic pollution by 95%, nutrients by 50% and total coliform by 99.6%. Guadie et al. (2021) reported the reuse of municipal wastewater treated by an aerobic–anoxic system, in which they reported that the removal efficiency of COD, BOD, TSS, NH_4-N and TN was 91–94%, 92–95%, > 96%, 92%, and 88%, respectively. But, the advantage of the present study is retaining microorganisms without washing them into treated effluent, as all are immobilised aerobic reactors.

Characterisation of dye fabricated cotton cloths

The final treated sewage was reused (Fig. 1b) for the dye fabrication on cotton cloth as discussed in materials and methods section. The dye fabricated cloths of DCFW and DCTS were subjected to instrumental analysis, and their characteristics are discussed below.

Colour comparison study and analysis of delta E and bursting strength of dyed cloths

The visual image of cloths is shown in Fig. 2, where cotton cloth (a), DCFW (b), and DCTS (c, d). It was found that plain cotton cloth was uniformly dyed in both DCFW and DCTS with no visual changes. The DCTS analysed in two trials looked similar in c and d of Fig. 2. Ćurić et al. (2021) reported the utilisation of treated textile wastewater for washing dyed fabric and found no significant difference with the standard sample.

The delta *E* value represents the dye affinity or neutral affinity onto fabric cloth, and the bursting strength indicates the strength of the fabric. The higher the neutral affinity

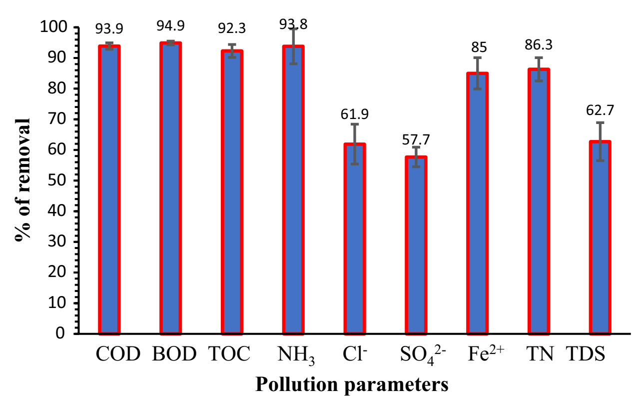

Fig. 3 Removal efficiency of sewage treatment process

value means stronger the fixation of dye onto fabric and vice versa. The neutral affinity and bursting strength of DCTS were compared with DCFW as shown in Supplementary Table S1. The neutral affinity of DCFW was taken as 100% and compared with two trial studies of DCTS (Table S1a, b) which showed 110.3 and 113.2% due to high dye fixation. The bursting strength value of DCTS and DCFW remained the same at 8.53 Kgs/sq.cm.

Analysis of colour strength (K/S) value of dye fabricated cloth

The absorbance coefficient (K) and transmittance coefficient (S) and their ratio measure the fabric's colour strength. The colour strength (K/S) values are used in textile industries to determine the depth of the colour of dyed cloths. It was measured at $\lambda_{360-750}$ nm for the DCTS and DCFW as shown in Fig. 4a. The K/S value of two trials of DCTS (a, b) was higher than that of DCFW. This may be because the treated sewage increases the fibre's external surface pores, attracting the dye ions for more colour development and firm bonding (Khan et al. 2021).

Thermogravimetric analysis (TGA) of dyed cloths

The TGA was carried out on dyed cloths to determine the thermal stability (Fig. 4b). The dyed fabrics were almost burnt entirely and DCTS showed similar thermal stability

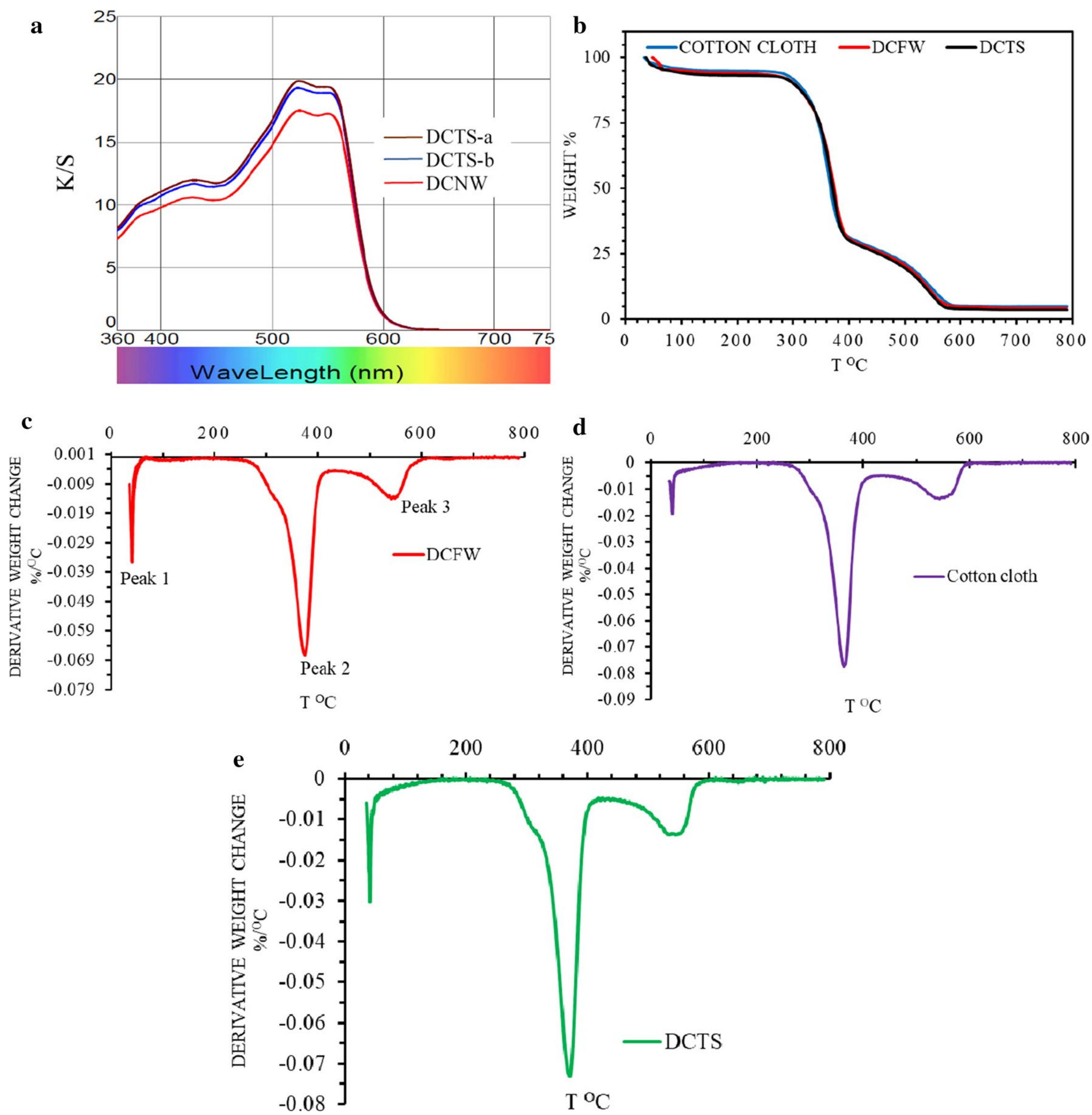

Fig. 4 Colour strength of DCFW and two trials of DCTS (**a**), TGA of cotton cloth, DCFW and DCTS (**b**) and the first derivative of TGA of cotton cloth (**c**), DCTS (**d**) and DCFW (**e**)

with DCFW. This was due to the no induced surface scaling formation using treated sewage. The TGA of cotton cloth, DCFW and DCTS showed the residue of 3.59 and 4.205% in w/w, respectively. The DCTS showed an increase in ash content by 0.615% w/w, and this may be attributed to the deposition or binding of inorganic salts present in the treated sewage. And also, DCTS showed that the percentage of weight loss was slightly higher than DCFW in the initial heating phase. This indicated that textile cloth had more affinity to dye during dye processing. The ionic atmosphere in treated sewage might induce polarisation of the functional groups in the cotton fibres to draw the dye molecules. This built a higher thickness of uniform dye throughout the surface of the cloth. The first derivative of TGA for cotton cloth, DCTS and DCFW is shown in Fig. 4c, d, e and calculated the enthalpy (ΔH value). It was found that the ΔH value for DCTS was comparatively less in peak two and peak three than DCFW and near to ΔH of cotton cloth due to releasing of exothermic energy (Table S2).

Field emission scanning electron microscope (FESEM) analysis of dyed cloths

The surface morphology of dyed cloths was analysed using FESEM in high and low resolution to determine the convoluted structure of bacterial swellings caused by the presence of bacteria and salts and also to determine the surface deposition of bacteria on the fibre and texture of the yarn after the dyeing process. The FESEM images in low and high resolution of DCFW (Fig. 5a, b) were compared with two trials studies of DCTS (Fig. 5c, d; e, f). It was observed that they do not show any significant changes in the yarn's morphology and texture due to the application of treated sewage for dye processing. And also, there was no evidence of damage in and on the fabric in terms of bacterial deposition, convolutions and swelling. This finding has also been observed by Yang et al. (2020) on the surface of the dyed cotton fabric.

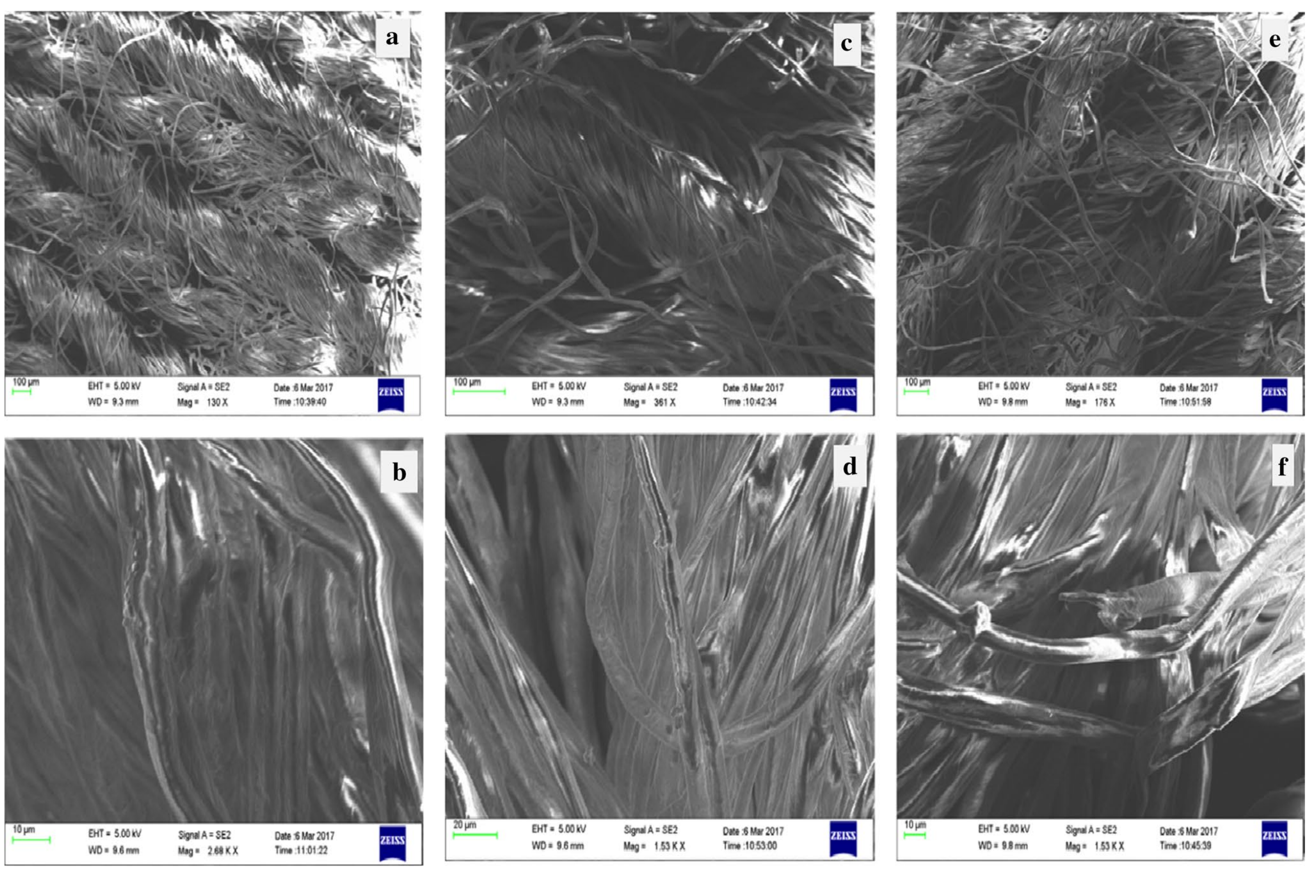

Fig. 5 The low- and high-resolution FESEM image of DCFW (**a**, **b**) and two-trial study of DCTS (**c**, **d**; **e**, **f**)

Energy-dispersive X-ray (EDX) analysis of dyed cloths

The EDX analysis was used to study the elemental composition of the dyed cloths. The two sharp peaks in all EDX spectrums were observed due to oxygen and carbon atoms (Fig. 6). This finding corroborates with the findings of Emam et al. (2021) and Hadid et al. (2021) for cellulose materials. DCFW (Fig. 6a), and two trials of DCTS (Fig. 6b, c) observed no change in the elemental composition. This was due to the no deposition of organic carbon, bacterial inclusion and bio-refractory organic compounds from treated sewage onto the cloth fibres during the dye processing. This gives evidence that the treated sewage does not contain any significant amount of bio-refractory organic compounds that cause deposition on cloth fibres. This also confirmed that the disinfection of the PBBOR outlet with sodium hypochlorite at the dosage of 10 mg L^{-1} was sufficient to lyse the pathogenic and non-pathogenic bacteria. The sand filtration of disinfected water eliminated the dead bacteria, so no deposition and impregnation of bacterial biomass took place during the dyeing of cloths.

Attenuated total reflection–Fourier transform infrared spectroscopy (ATR-FTIR) analysis of dyed cloths

The ATR-FTIR spectra of cotton cloth, DCFW and DCTS were studied and are shown in Fig. 7a. All ART-FTIR spectrum peaks resembled the same with cotton cloth except the peak at 1542 cm^{-1} in DCFW and DCTS due to the N–H stretching of -N=N- with force constant of 9.801 N/cm. It confirms that the binding of dyes onto the cotton fabric took place. The peak around 3336 cm^{-1} is attributed to O–H stretching of cellulose, and the peak at 2902 cm^{-1} is due to C–H Stretching. The peak at 2359 cm^{-1} may be due to C–O stretching, and the peak at 1636 cm^{-1} may be due to the C–O stretching of COOH. The peak at 1428 cm^{-1} and 1320 cm^{-1} corresponds to O–H in-plane coupling with C–H bending. The peak at 1035 cm^{-1} corresponds to the C–H in-plane bend, and the peak at 704 cm^{-1} is due to the C–H bending of alkynes. Hence, the reuse of treated sewage without any ultra-purification is suitable for dye fabrication in the textile process to get reliability as of DCFW.

The viability of dyed cloths against microorganisms

The DCFW and DCTS subjected to the microbial study are shown in the supplementary information (Fig. S11, S12). It showed the significant growth of bacteria and coliforms in DCFW. However, bacterial growth was not seen in the DCTS due to the usage of hypo in the treatment process. Thus, the DCTS is protected from the infection of microorganisms.

Treatment of dye wastewater generated in dye processing with characterisation

The treated sewage was used for textile processing in mechanical dyeing equipment to follow the industrial dyeing procedure. The spent dye wastewater (DWW) was further treated and discharged as shown in Fig. 1c. The scheme consists of the FBCOR-PBCOR-FBBOR process.

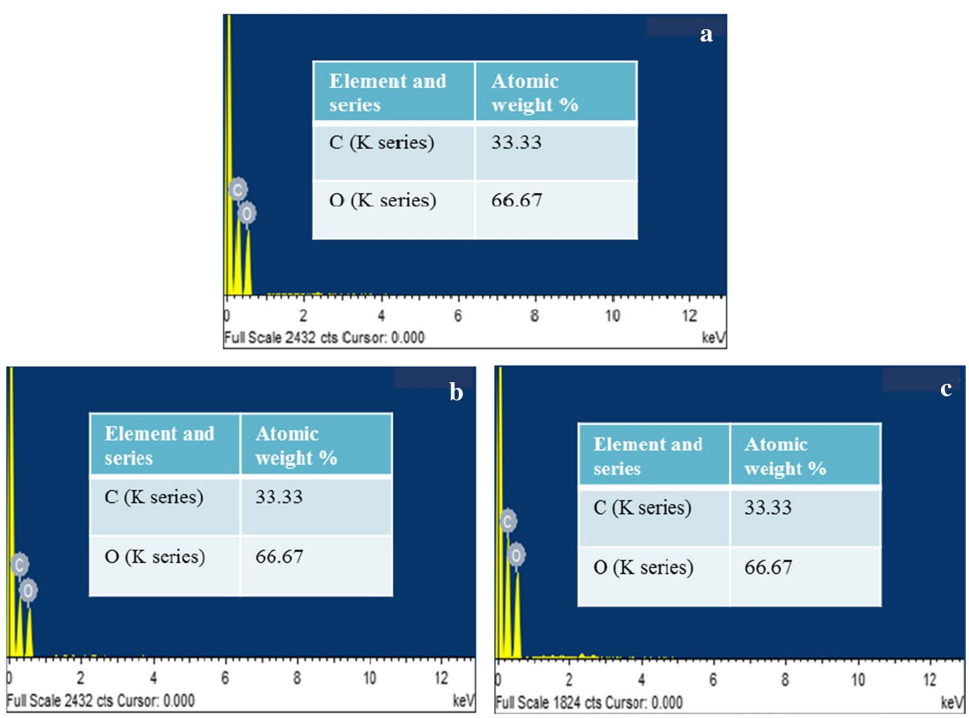

Fig. 6 EDX analysis of DCFW (**a**) and two trials of DCTS (**b**, **c**)

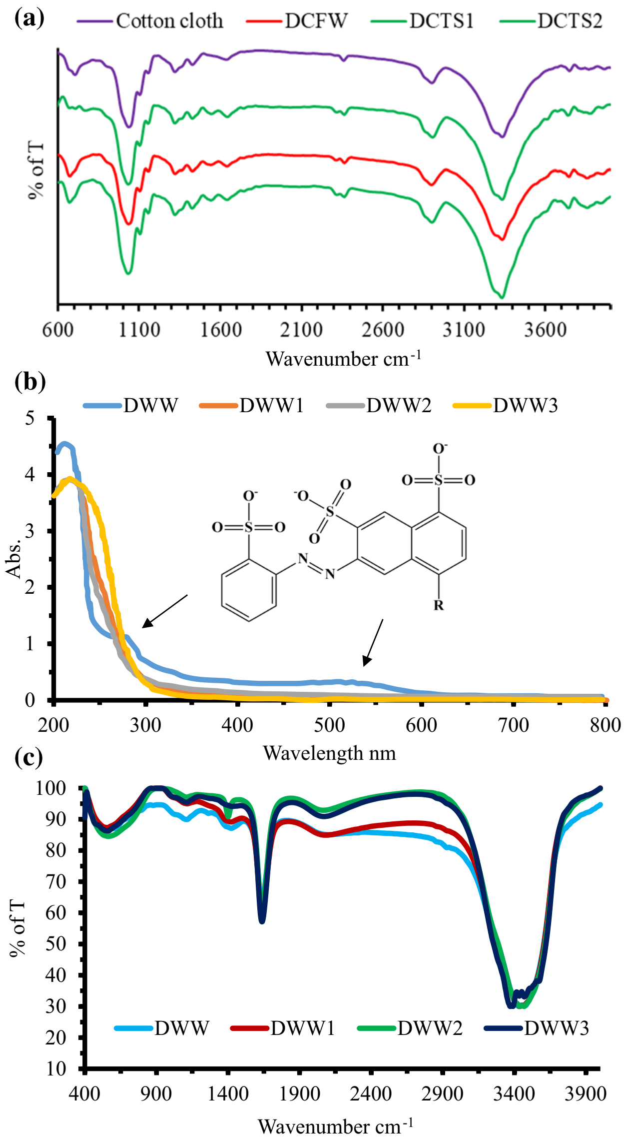

Fig.7 ATR-FTIR results of cotton cloth, DCFW and DCTS (**a**), UV–visible spectrum (**b**), and FTIR spectrum (**c**) of DWW treatment operations

The FBCOR and PBCOR reactors were used for the catalytic destruction and mineralisation of organic compounds present in DWW. Then the mineralised compounds were treated further in the FBBOR reactor using microorganisms. Table 2 shows the characteristics of raw DWW as COD, 1760 ± 51.2 mg L^{-1}; BOD, 98 ± 12.5 mg L^{-1} with the BOD: COD of 0.0559 ± 0.0548. The BOD:COD ratio suggests that DWW is less amenable to biological degradation, so the chemical treatment applied before microbial treatment to increase biodegradability. The optimisation studies of DWW are described in supplementary information section S3. The DWW treated at the optimised condition and the removal of pollution parameters for the FBCOR outlet (DWW_1),

Table 2 Characteristics of the treatment process of DWW

	pH	ORP mV	TDS g L^{-1}	COD	BOD	TOC	TN	NH_4^+-N	Sulphate	Chloride g L^{-1}
DWW	9.68 ± 0.51	−95.6 ± 25.3	38.76 ± 0.12	1760 ± 51.2	98.4 ± 12.5	477.8 ± 35.2	69.18 ± 6.5	15.4 ± 5.1	0.47 ± 0.12	28.29 ± 0.12
DWW_1 6 h	8.64 ± 0.35	−60.4 ± 15.6	36.54 ± 0.13	1040 ± 35.1	97.76 ± 6.4	416.5 ± 32.7	57.95 ± 5.2	21.6 ± 4.3	0.44 ± 0.15	27.3 ± 0.15
DWW_1 24 h	8.63 ± 0.37	−35.1 ± 14.3	34.63 ± 0.15	640 ± 25.6	52.48 ± 8.1	400.0 ± 25.1	47.51 ± 4.2	21 ± 3.5	0.58 ± 0.19	26.8 ± 0.13
DWW_2 1 h	8.19 ± 0.31	−1.5 ± 12.1	32.64 ± 0.12	320 ± 15.4	51.2 ± 5.4	132 ± 15.5	36.41 ± 4.9	18.4 ± 4.6	0.55 ± 0.14	25.84 ± 0.15
DWW_3 12 h	7.59 ± 0.25	32.5 ± 15.3	29.68 ± 0.10	240 ± 12.3	48 ± 6.7	50.32 ± 13.2	26.21 ± 7.5	12.4 ± 6.1	0.3 ± 0.12	25.8 ± 0.13
DWW_3 24 h	7.48 ± 0.26	57.9 ± 16.3	26.83 ± 0.15	85 ± 8.6	10.48 ± 3.4	33.4 ± 8.6	16.1 ± 4.5	6.4 ± 3.1	0.3 ± 0.13	25.8 ± 0.05

PBCOR outlet (DWW_2) and FBBOR outlet (DWW_3) are shown in Table 2. The overall performance of the DWW treatment system under optimised conditions was COD by 95.1% ± 0.58, BOD by 89.4% ± 3.02, TOC by 93.0% ± 1.82, TN by 76.7% ± 6.16 and NH_4^+-N by 58.4% ± 10.11 in w/v, respectively. The chemical oxidation method removed the pollutant by 81.8% ± 0.85, whereas the biological oxidation method removed 13.35% ± 0.54 in w/v. Thus, the chemical oxidation method was dominant due to the high oxidation potential of hydroxyl radicals generation in dye wastewater treatment.

Santhana Raj et al. (2021) reported the textile effluents remediation for water reuse by microbial treatment using Escherichia fergusonii followed by adsorption in activated charcoal. They reported combined removal efficiency of colour removal by 98.4%, COD by 92.5% and BOD by 85%. Ćurić et al. (2021) reported the textile wastewater treatment using ultrafiltration membrane technology to reuse the treated water for washing the dyed fabric. They stated that the COD, BOD and TOC removal efficiency was 79.8, 56.0, 82.4 and 81.0%, respectively. The major disadvantage of the treatment method is the high cost of the equipment, membrane fouling, decreasing permeate flux over the time and management of reject, adsorbed material and fouled membrane.

UV–visible spectroscopy and FTIR analysis of DWW treatment

UV–Vis spectra were recorded in the spectral range of $\lambda_{200–800}$ nm for the DWW treatment process (Fig. 7b). A peak at λ_{510} nm is responsible for the chromospheres of the azo functional compound present in the DWW (Shi et al. 2021; Amer and Hadi 2022). The DWW_1 showed hypo and hypochromic shifts due to the cleavage of bulk and complex molecules into simpler units. The complete removal of the chromosphere group was found in DWW_2 after the treatment of DWW_1. The DWW_2 was further treated in the FBBOR reactor, and its outlet shows a reduction in intensity by successive oxidation of organic compounds during the treatment process.

The FTIR spectrum (Fig. 7c) of DWW showed a broad peak at 3429.98 cm^{-1}, attributed to N–H and O–H stretching vibrations. The peaks at 1422.63 cm^{-1}, 1635.93 cm^{-1}, 1111.83 cm^{-1} may be corresponding to C–N, C=O (amide group) and C–O stretching of the organic compounds. A peak at 565.44 cm^{-1} corresponds to the vibrations of the benzene ring (Saifi et al. 2021; Yu and Hiramatsu 2022). The spectra of other wastewaters of DWW_1, DWW_2 and DWW_3 resembled DWW with a change in wavenumber. DWW_1 showed peaks at 3433.11, 1636.99, 1420.63, and 554.68 cm^{-1}. DWW_2 showed peaks at 3467.45, 1636.65, 1401.13, and 565.32 cm^{-1}. DWW_3 showed a peak at 3468.93, 1637.29, and 563.67 cm^{-1}. It was evident that the disappearance of the C–N stretching peak in the FBBOR reactor confirms the complete degradation of azo dye compounds.

Fluorescence spectroscopy analysis of DWW treatment

The fluorescence spectra were recorded for the DWW treatment process (Fig. S8). It was observed that the DWW is not fluorescence active because azobenzene is not an active fluorescent compound (Dudek et al. 2020; Cuétara-Guadarrama et al. 2021). The change in the excitation peak was observed in DWW_1 & DWW_2 due to their cis/trans isomerisation (Zink-Lorre et al. 2021). The DWW showed an excitation peak at λ_{620} nm due to the presence of dye compounds on further treatment in the FBCOR process. The excitation region was altered due to changes in the molecular structure by the oxidation process in DWW_1. The dye compounds disappeared entirely in DWW_2 and DWW_3. Thus, the fluorescence spectrum supported the findings of FTIR and UV–visible for the complete removal of azo dye compounds by the treatment process.

Mechanism of reactive dye degradation

In the degradation mechanism, the potential of hydroxyl radicals induced the polarisation of electrons in the conjugated π bonded electronic structure of reactive dye (1). The substituted aryl compound (reactive dye) was attacked by hydroxyl radical (2) and cleft the azo bond to form two hydroxylated substituted aryl compounds (3). The Ar–NH–NHOH (4) was deprotonated to form Ar–NH–N=O (5), and then the molecule could be cleaved by hydroxyl radical to form water and carbon dioxide with others (6). Similarly, the Ar–SO_2OH (7) attached by hydroxyl radicals resulted in water and carbon dioxide formation. The UV–visible spectrum showed the λ_{max} at 510 nm due to reactive dye present in DWW. The degradation of reactive dye was confirmed with UV–Vis, FTIR and fluorescence analysis and also the decrease in TOC along with COD confirmed the degradation of dye compounds in DWW. The treated dye wastewater can be again reused for domestic or other industrial purposes, and thus the sustainability achieved (Fig. 1b).

Economic analysis of the entire treatment process

Domestic wastewater treatment and its reuse application in the textile process would considerably benefit the industry. The treated sewage water had characteristics similar to groundwater sources; thus, it can be safely handled in the textile industry. The expenditure towards the reuse of treated sewage water is shown in detail in supplementary information section S4 by assuming the textile industry requires

1000 kilolitre per day (KLD). The proposed scheme's net saving on dyeing with the reuse of treated sewage water is 3783.8 USD/day.

Thus, by implementing the reuse of treated domestic sewage for the dyeing process and again treating the dye wastewater to reuse for domestic purposes able to reduce the water stress condition. And also, the sustainability of the industry and the environment in terms of freshwater can be achieved.

Conclusion

The study concludes that (i) the domestic sewage could be efficiently treated with sequential microbial reactors of SOAR-I, FBBOR, SOAR-II and PBBOR. It had COD, BOD, $NH4^+$-N, TN and TOC removal efficiency by 93.9% ± 1.04, 94.9% ± 0.9, 93.8% ± 5.7, 86.3% ± 3.85 and 92.3% 2.19 in w/v, respectively.

(ii) The treated sewage was reused for the dyeing process on the cotton fabric and compared with the freshwater. The treated sewage water did not affect the dyed fabric compared with freshwater. The dye fixation was confirmed with the instrumental analysis of ATR-FTIR, EDX, FESEM, delta *E* (%), bursting strength (Kgs/sq.cm) and K/S values. After dyeing, the EDX results showed similar results with the freshwater used cloth fibre. The SEM images did not contain any significant changes due to the usage of treated sewage water for textile processing, and also, there was no evidence of damage in and on the fabric. The ATR-FTIR results of the dyed fabric showed the presence of -N=N-, which indicated the presence of azo dyes. The bacterial growth was observed in the freshwater dyed fabric, but the treated water dyed fabric did not show any bacterial growth. Hence, the second objective of reutilising treated domestic sewage was also achieved and compared with freshwater.

(iii) The generated dye effluent from the dyeing process was treated using sequential reactors of FBCOR, PBCOR and FBBOR. It efficiently removed COD by 95.1% ± 0.58, BOD by 89.4% ± 3.02, TOC by 93.0% ± 1.82, TN by 76.7% ± 6.16 and NH_4^+-N by 58.4% ± 10.11 in w/v, respectively. The treated dye water can be reused for domestic usage, and thus the recycling process continues. Hence, the objective of treatment of dye wastewater was also achieved successfully. So, the proposed methods can sustain the environment and reduce water stress in terms of freshwater.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

Ethical approval This article does not contain any studies with human participants or animals performed by any of the authors.

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