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Green iron particles in textile and dyebath wastewater decolorization

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ABSTRACT

The world is fronting water quality crisis and industrialization has augmented the illegal discharge of wastewater to the environment. Textile is one of such industry, generates large amount of colored wastewater and discharges untreated wastewater into nearby environment, poses major threats to the environment worldwide. The textile wastewater consists of the spent water from pretreatment, dyeing and post treatment process of the textiles. However the dyebath wastewater is highly concentrated and consists of mixture of unfixed dyes. The effective and eco-friendly treatment of textile wastewater is challenging. It is essential to remediate the dyebath wastewater in order to maintain environmental sustainability. In this paper, the feasibility of green iron particles on real textile and dyebath wastewater decolorization was investigated. Green iron particles were synthesized using green tea leaves extracts and grape leaves extracts. The synthesized particles were characterized for size, morphology, crystal structure, elemental analysis, functional groups attached and specific surface area. 2–6 g/L of green iron was utilized to decolorize 63%–89% of highly concentrated dyebath wastewater and 80%–92% of textile wastewater. The pH 10.1–10.8 of wastewater was reduced to 8.1–8.9. Green iron removed nearly 50%–54% of chemical oxygen demand from the wastewater and concluded that partial mineralization was possible. The reusability of green iron particles was achieved for 3 cycles. Thus the present investigation confirmed the feasibility of green iron particles in textile and dyebath wastewater decolorization.

Keywords: Green iron particles; Green tea leaves; Grape leaves; Dyebath wastewater; Textile wastewater

1. Introduction

The tremendous growth in population, urbanization and industrialization in the recent years utilized most of the water resources in the world and degraded the environment [1]. It is clear that future demands for water could not be met unless the wastewater is properly managed. Textile is one such industry, utilizes enormous amount of water, manufactures 60 billion kg of fabric annually by releasing huge amount of colored wastewater and pollutes the environment [2]. It is reported that more than 50 sites in the world discharges colored wastewater from dyeing industry and it poses a complex threat to the environment of surrounding population [3]. In the wet processing of fabrics, three stages of treatment process are involved, that is, pretreatment, dyeing and post-treatment of the fabrics. The pretreatment improves the white color and texture of the fabrics using bleaching chemicals and the wastewater consists of mostly washed bleaching agents. Dyeing is the most complex step utilizes hundreds of dyes and auxiliary chemicals [4]. The global dyes and pigments market is projected to grow from $40.71 billion in 2022 to $56.91 billion by 2029. The dyeing wastewater consists of purely high concentration of unfixed dyes that contributes

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more in the textile wastewater [5]. The post treatment process includes several times of cold and hot washing of fabrics after dyeing, and the respective rinse wastewater consists of diluted colored wastewater [6]. The water requirement for dyeing purpose varies from 36–176 L/kg of fabric. The wastewater generation during dyeing process alone fluctuates 35–175 L/kg with an average of 105 L/kg of fabric [7]. Hence, the real textile wastewater consists of combined wastewater from all the three processes. The dyebath wastewater has more dye concentration when compared to the combined textile wastewater [8].

The conventional treatment methods are inefficient to treat textile wastewater containing different textile dyes, alkalis, organic and inorganic salts [9] and this led to the development of advanced treatment methods [10]. The application of nanotechnology [11] and the potential of iron in removing various organic contaminants have been successfully reported [12]. Studies of iron particles on textile dye decolorization are receiving more focus in recent years [13], as the particles are highly reactive, non-toxic, and cost-effective [14]. Iron particles aggregate quickly with respect to time, due its magnetic property and in order to stabilize iron particles, various supports are developed to provide strong inter-particle electrostatic repulsions [15]. In recent years, plant based green supports are introduced and the major benefits of this biosynthesis process is simple and quick, neither used nor generated any toxic chemicals, cost-effective and eco-friendly [16]. In general, plant extract is comprised of polyphenols, flavonoids, etc. in which polyphenols are known to be strong reducers [17] and effective metal chelators, forming stable complexes with iron [18]. Polyphenols are well known as antioxidants and several epidemiological studies have verified [19] that those have beneficial effects on human health [20]. In addition, iron possesses octahedral geometry, could coordinate up to three polyphenol groups [21].

Green supports are claimed recently to improve the stability, reactive as well as to enable eco-friendly remediation [22], still very few studies demonstrated the promising of green iron particles in textile wastewater decolorization [23]. However, the decolorization of dyebath wastewater is the real challenging [24]. In this study, the feasibility of green iron particles on real textile and dyebath wastewater decolorization was investigated and reported. The dye concentration was calculated from dyebath, rinse bath recipes and mass balance study. The wastewater was collected from the textile industries in Tiruppur, Tamil Nadu, Southern state in India. The characterization of wastewater before and after treatment with synthesized iron particles is performed. It is mandatory to evaluate the potential of green iron particles on real textile and highly concentrated dyebath wastewater treatment before it is recommended for any further industrial applications.

2. Materials and methods

2.1. Wastewater collection and characterization

The wastewater was collected from textile industries in Tiruppur, Tamil Nadu, India. Two samples of dyebath (DBWW1, DBWW2) and five samples of textile wastewater (TWW1, TWW2, TWW3, TWW4, TWW5) were collected. The wastewater consists of the spent water from pretreatment, dyeing and post-treatment process of the textiles. However the dyebath wastewater consists of wastewater from dyeing process alone and it consists of mixture of unfixed reactive dyes, chemicals such as Glauber salt (sodium sulfate) and soda ash (sodium carbonate). These chemicals were added in order to enhance the fixation property of the dyes to the fabrics [25]. The dye concentration was calculated from dyebath, rinse bath recipes and mass balance. The collected wastewater and dyebath wastewater was characterized for pH, total dissolved solids (TDS), chlorides (Cl), sulfates (SO_4), sodium (Na), electrical conductivity (EC), chemical oxygen demand (COD) as per the American water works association standard methods for the examination of water and wastewater [26] and the total organic carbon (TOC) was determined using standard method [27].

2.2. Mass balance

Two samples of different dyebath wastewater and rinse wastewater were collected from the industries (Fig. 1) and the respective fabric dyeing recipe was also received. With the help of the recipes and collected samples, the total solids (TS) were found out. Both recipe and collected sample data were compared. The dye fixation was calculated as 83%–86% and the unfixed reactive dyes concentration was computed. The variation in total solids was observed as 1.21 g for first sample, 19.11 g for second sample (Fig. 2a and b). The highest variation in total solids of second sample might be due to the error in collecting samples before proper mixing of the dye, and also the error variation was less than 0.05%.

The respective dye concentration in dyebath and rinse wastewater was determined based on mass balance (Fig. 3a and b). For 86% dye fixation, it found out that the concentration of dye in dyebath wastewater as 160 mg/L and rinse wastewater as 28 mg/L, respectively. Similarly for 83% dye fixation, the dye concentration in dyebath wastewater and rinse wastewater was calculated as 183 and 30 mg/L, respectively. Thus, the textile wastewater is the mixture of dyebath as well the rinse wastewater in dyeing process in addition to the wastewater received from pretreatment and post-treatment process in the fabrics dyeing. The mass balance study demonstrates the existing maximum dye concentration in dyebath wastewater, and the significance of decolorization of highly concentrated dyebath wastewater, as it plays major role in the color of textile wastewater.

2.3. Synthesize of green iron particles

The green iron particles GT-nZVI was synthesized from *Camellia sinensis* (green tea) leaves extract [28] and GP-nZVI was synthesized from *Vitis vinifera* (black grapes) leaves extract [23], as reported in the earlier study. A solution of 0.5 M $FeCl_3$ was prepared and mixed with green extract separately in 1:1 volume ratio. The observed precipitates confirmed the formation of green iron particles GT-nZVI and GP-nZVI (Fig. 4) respectively. The synthesized green particles were vacuum filtered, dried, stored in vials, and kept in a desiccator until further use.

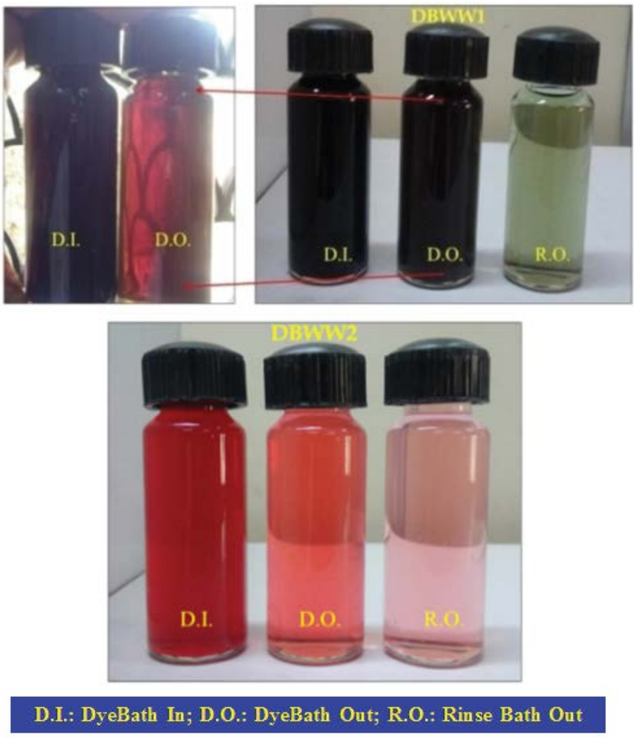


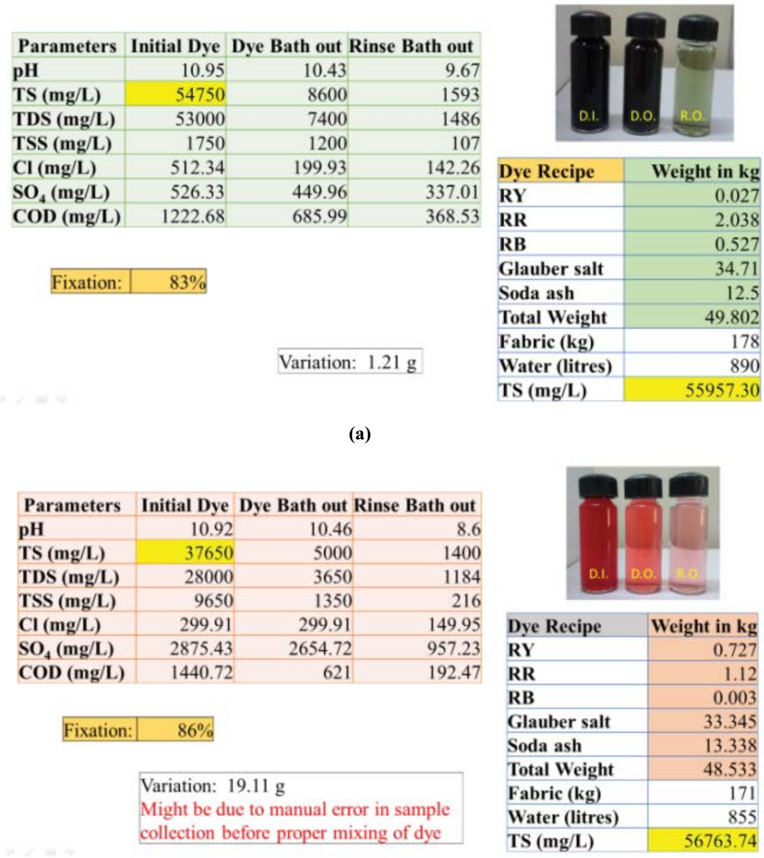
Fig. 1. Dyebath wastewater (DBWW1, DBWW2).

2.4. Characterization of green iron particles

Green iron (GT-nZVI, GP-nZVI) particles were characterized for particle size, crystal structure, morphology, elemental analysis, functional groups attached and specific surface area. The particle size was determined using Nanoparticle Analyzer SZ-100 (Horiba, Japan) and the crystal structure was observed using Ultima IV X-ray Diffractometer (Rigaku, Japan) at 40 kV voltage and 30 mA current. The morphology and elemental analysis was performed using transmission electron microscopy (TEM) coupled with energy-dispersive X-ray spectroscopy (EDS) using Tecnai G2 Spirit (FEI, The Netherlands). The functional groups attached were recognized with Fourier-transform infrared (FTIR) spectrum recorded using PerkinElmer (Instrument Co., Ltd., USA). The Brunauer–Emmett–Teller (BET) specific surface area was measured using Micromeritics ASAP 2020 (Micromeritics, USA).

2.5. Decolorization of wastewater

The green iron particles of 1–6 g/L dose was added to the 30 mL of textile wastewater, dyebath wastewater and mixed properly. The decolorization was monitored at every 5, 10, 15, 30, and 60 min. The wastewater treatment using green iron particles was further characterized for pH, TDS, Cl, SO$_4$, Na, EC, COD, and TOC. The color removal was monitored using UV-Vis Spectrophotometer and measured at multiple



Parameters	Initial Dye	Dye Bath out	Rinse Bath out
pH	10.95	10.43	9.67
TS (mg/L)	54750	8600	1593
TDS (mg/L)	53000	7400	1486
TSS (mg/L)	1750	1200	107
Cl (mg/L)	512.34	199.93	142.26
SO$_4$ (mg/L)	526.33	449.96	337.01
COD (mg/L)	1222.68	685.99	368.53

Fixation: 83%

Variation: 1.21 g

Dye Recipe	Weight in kg
RY	0.027
RR	2.038
RB	0.527
Glauber salt	34.71
Soda ash	12.5
Total Weight	49.802
Fabric (kg)	178
Water (litres)	890
TS (mg/L)	55957.30

(a)

Parameters	Initial Dye	Dye Bath out	Rinse Bath out
pH	10.92	10.46	8.6
TS (mg/L)	37650	5000	1400
TDS (mg/L)	28000	3650	1184
TSS (mg/L)	9650	1350	216
Cl (mg/L)	299.91	299.91	149.95
SO$_4$ (mg/L)	2875.43	2654.72	957.23
COD (mg/L)	1440.72	621	192.47

Fixation: 86%

Variation: 19.11 g
Might be due to manual error in sample collection before proper mixing of dye

Dye Recipe	Weight in kg
RY	0.727
RR	1.12
RB	0.003
Glauber salt	33.345
Soda ash	13.338
Total Weight	48.533
Fabric (kg)	171
Water (litres)	855
TS (mg/L)	56763.74

(b)

Fig. 2. Determination of dye fixation of dyebath wastewater (a) DBWW1 and (b) DBWW2.

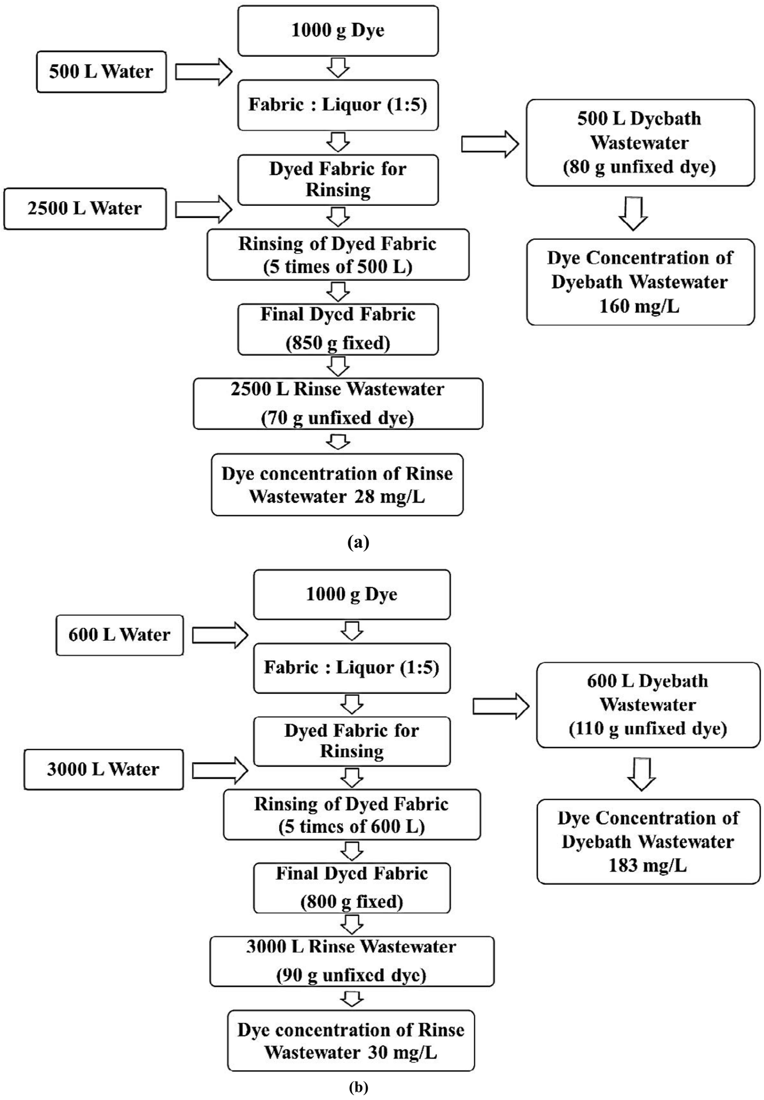


Fig. 3. Mass balance for unfixed dye concentration in dyebath wastewater.

wavelengths of 436, 525, 620 nm after removing the turbidity by centrifugation. The green iron dose was varied and optimized for maximum dye decolorization. The repeatability of the experiment confirms the precision of results; hence all the experiments were triplicated and obtained the results with a standard deviation error of less than 5%.

3. Results and discussion

3.1. Wastewater characterization

The textile wastewater samples (TWW1, TWW2, TWW3, TWW4, TWW5) and dyebath wastewater samples (DBWW1, DBWW2) were characterized for pH, TDS, Cl, SO_4, Na, EC, COD and TOC and presented in Table 1. As discussed earlier in mass balance study (section 2.2), the alkaline nature of dyebath out is higher than textile wastewater, which is a mixture of pretreatment, dyeing and post-treatment wastewater. The initial pH of dyebath out wastewater was 10.6–10.8, and the textile wastewater was 10.1–10.4.

The EC and TDS values are lesser in dyebath wastewater, compared to textile wastewater, as the dyebath wastewater mainly consists of unfixed reactive dyes, Glauber salts and soda ash. However, the textile wastewater consists of all the inorganic salts utilized for pretreatment, post-treatment process in addition to the dyeing process. The initial COD and TOC value ranges from 782–1,785 mg/L and 264–638 mg/L, respectively. The major cations and anions exist in the textile wastewater were Cl, SO_4, and Na, as these ions mostly present as auxochrome in the dye structure [6].

3.2. Characterization of green iron particles

The average particle size of green iron particles (GT-nZVI and GP-nZVI) were found as 120–140 nm (Fig. 5) and in a recent study, it was reported as 172 nm [22], more than 100 nm as it is capped by polyphenolic compounds [29]. The X-ray diffraction spectrum of both green iron particles showed no peaks (Fig. 6), validates the amorphous nature of iron particles with polyphenolic compounds and

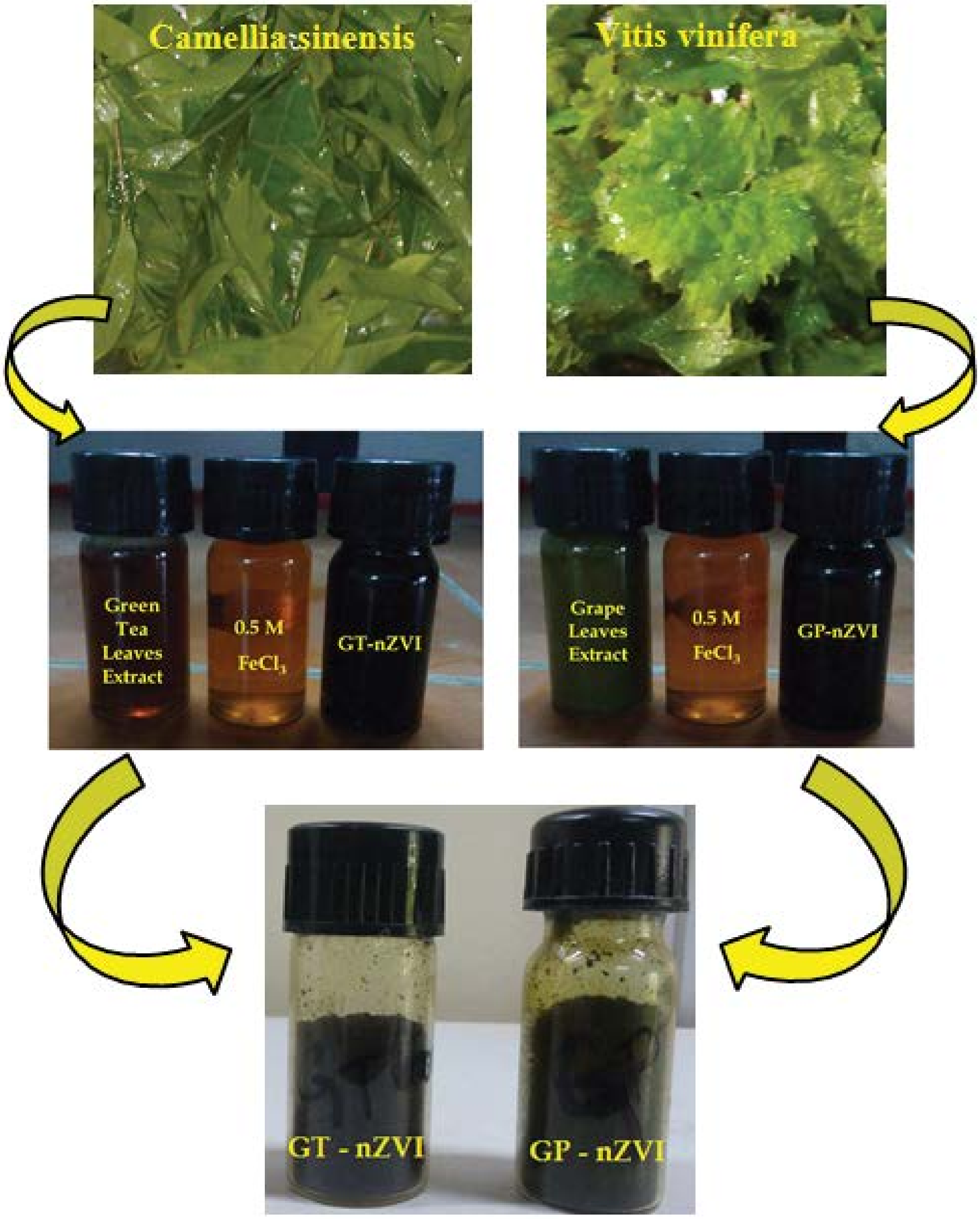


Fig. 4. Synthesis of green iron particles (GT-nZVI and GP-nZVI).

Table 1
Characterization of textile and dyebath wastewater

Parameters	TWW1	TWW2	TWW3	TWW4	TWW5	DBWW1	DBWW2
pH	10.3	10.1	10.3	10.2	10.4	10.8	10.6
EC (dS/m)	16.35	17.33	22.58	11.87	21.14	11.87	4.91
TDS (mg/L)	7,264	8,965	10,486	9,634	9,885	7,856	3,856
COD (mg/L)	1,652	1,288	945	853	1,304	1,785	782
TOC (mg/L)	584	436	337	308	459	638	264
Cl (mg/L)	4,458	5,179	7,482	6,732	7,281	214	328
SO_4 (mg/L)	354	512	488	464	459	5,780	2,587
Na (mg/L)	1,897	2,044	2,693	2,361	2,265	2,456	432

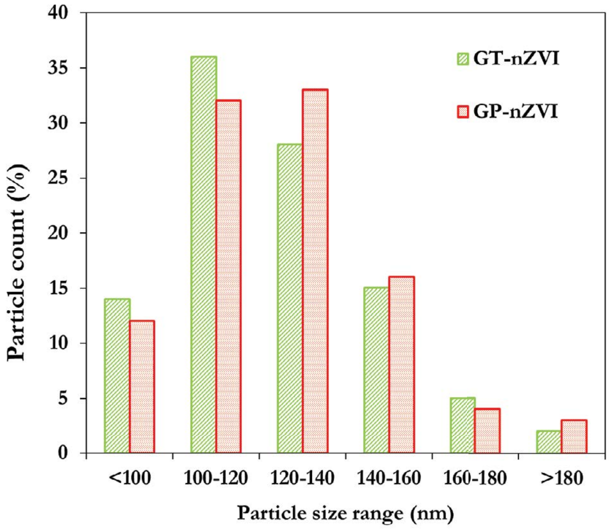


Fig. 5. Particle size of GT-nZVI and GP-nZVI.

similar observations were reported in the recent study [20]. The amorphous nature of green iron particles were further recognized with TEM images as shown in Fig. 7.

The FTIR spectra of green iron (GT-nZVI and GP-nZVI) showed a broad envelope due to OH stretching vibration of 3,400–3,000 cm^{-1} as reported in an earlier study [30]. The widening is due to the hydrogen bonding of OH groups and the sharp peak at 1,567 and 1,588 cm^{-1} confirms the stretching vibrations of aromatic ring of polyphenolic compounds in green iron, that is, GT-nZVI and GP-nZVI [31]. The peaks observed in the fingerprint region confirmed the presence of Fe–O bond in GT-nZVI and GP-nZVI (Figs. 8 and 9). Hence, from the observed FTIR spectra, it is confirmed that polyphenolic compounds are mainly involved in complexation with Fe^{3+} in green iron [32] and

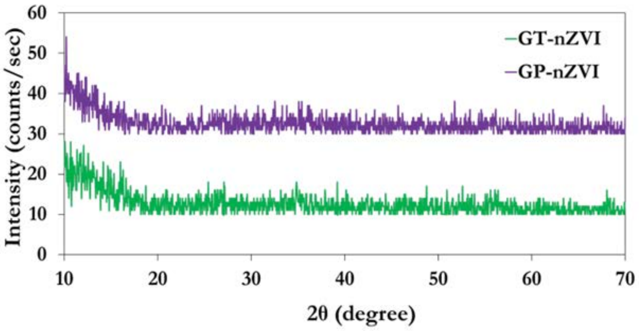


Fig. 6. Crystal structure of GT-nZVI and GP-nZVI.

thus amorphous morphology [33]. The BET surface area was measured as 14.6 m^2/g for GT-nZVI, and 15.08 m^2/g for GP-nZVI respectively. Recent studies reported 6.67 and 5.8 m^2/g of surface area for green iron [34] and Gao et al. [22] reported 54.27 m^2/g of surface area for 172 nm of green iron. However, the increase in particle size reduces the specific surface area and there by the reactivity [35].

3.3. Decolorization of wastewater

The green iron dose was optimized as 2–6 g/L and utilized for the decolorization of textile and dyebath wastewater. Green iron (GT-nZVI and GP-nZVI) decolorized nearly 90%–92% of textile wastewater. However, in case of dyebath wastewater, GT-nZVI and GP-nZVI removed 63%–86% and 78%–89% of color, respectively only, as the dyebath has highly concentrated colored wastewater. The dyebath wastewater decolorization using green iron particles is shown in Fig. 10. The characterization of textile and dyebath wastewater after treatment with green iron particles (GT-nZVI and GP-nZVI) are presented in Tables 2 and 3. The pH of wastewater was reduced from 10.6–10.8 to 8.3–8.9

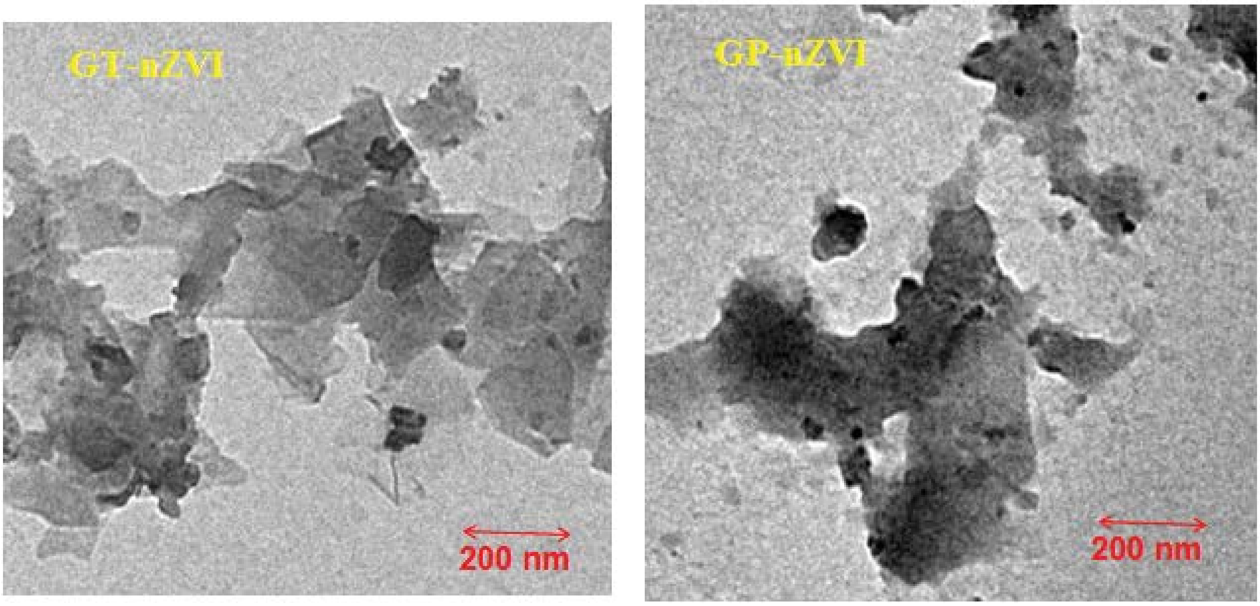


Fig. 7. TEM images of GT-nZVI and GP-nZVI.

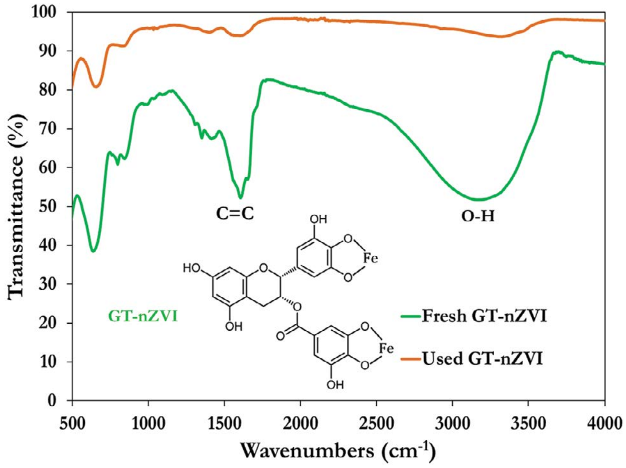


Fig. 8. FTIR spectra of fresh and used GT-nZVI.

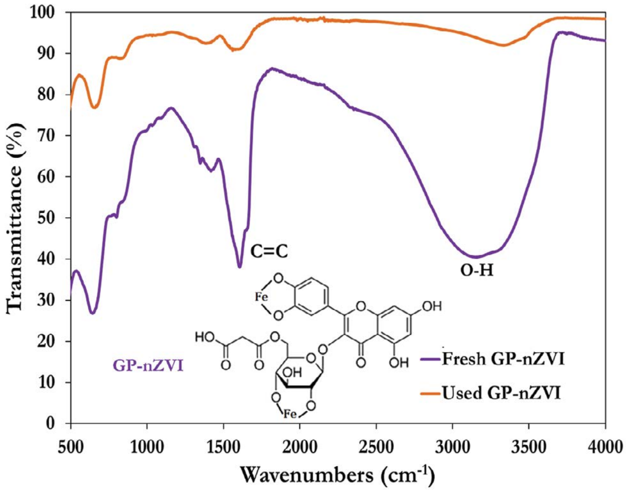


Fig. 9. FTIR spectra of fresh and used GP-nZVI.

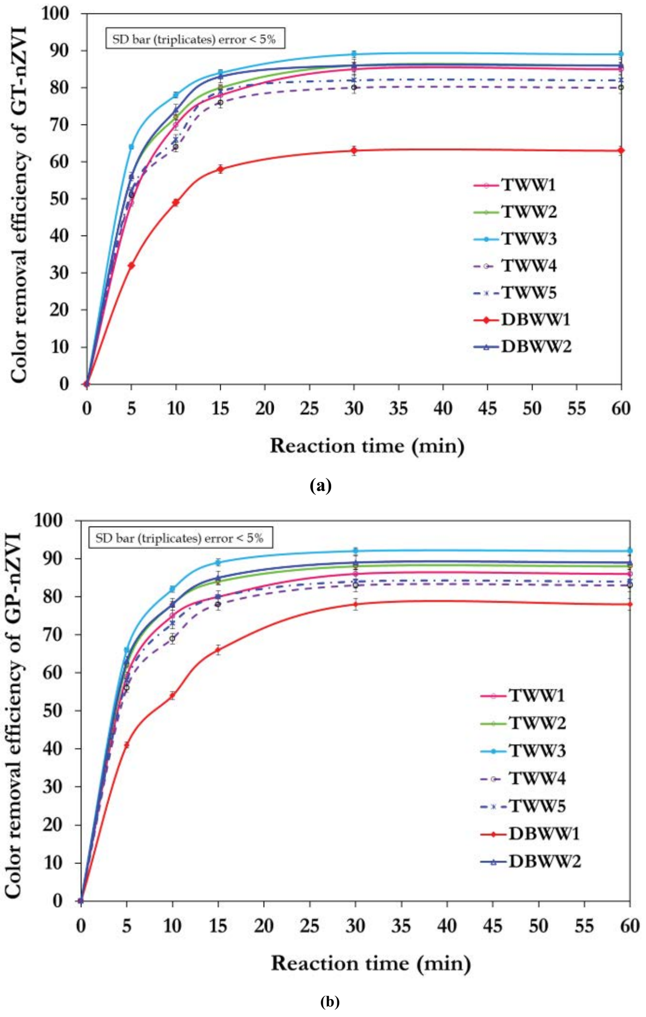
Fig. 10. Dyebath (DBWW2) wastewater decolorization using green iron particles.

Table 2
Textile and dyebath wastewater after treatment with GT-nZVI

Parameters	TWW1	TWW2	TWW3	TWW4	TWW5	DBWW1	DBWW2
pH	8.6	8.5	8.6	8.5	8.6	8.9	8.3
EC (dS/m)	14.32	15.28	20.56	9.93	18.54	9.51	4.63
TDS (mg/L)	7,011	8,867	10,268	9,475	9,622	7,112	3,362
COD (mg/L)	1,128	856	461	405	879	1,165	449
TOC (mg/L)	467	318	152	126	324	469	157
Cl (mg/L)	4,414	5,172	7,478	6,726	7,256	203	314
SO_4 (mg/L)	348	507	482	458	452	5,776	2,582
Na (mg/L)	1,886	2,034	2,678	2,346	2,247	2,252	424

Table 3
Textile and dyebath wastewater after treatment with GP-nZVI

Parameters	TWW1	TWW2	TWW3	TWW4	TWW5	DBWW1	DBWW2
pH	8.4	8.3	8.4	8.4	8.4	8.5	8.1
EC (dS/m)	14.28	15.11	20.23	9.14	18.36	9.14	4.56
TDS (mg/L)	6,987	8,856	10,156	9,463	9,589	7,105	3,346
COD (mg/L)	1,108	827	454	398	826	1,034	431
TOC (mg/L)	424	296	147	116	317	442	138
Cl (mg/L)	4,395	5,154	7,462	6,704	7,244	198	302
SO_4 (mg/L)	336	496	476	445	445	5,762	2,573
Na (mg/L)	1,882	2,028	2,667	2,339	2,235	2,236	418



(a)

(b)

Fig. 11. Textile and dyebath wastewater decolorization using (a) GT-nZVI (b) GP-nZVI.

in the presence of GT-nZVI and 8.1–8.5 in the presence of GP-nZVI. The organic loading due to green iron particles were also monitored distinctly and presented in Fig. 11. Reasonable reduction in COD was observed and the green iron, GT-nZVI and GP-nZVI removed nearly 48%–52% of COD, as shown in Figs. 12 and 13.

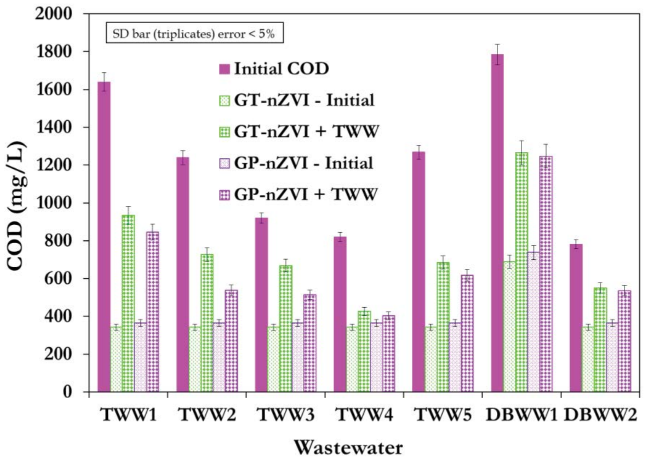


Fig. 12. COD monitored for green iron particles.

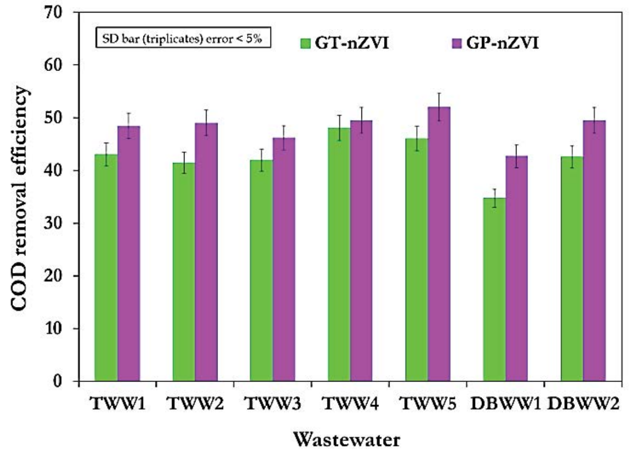


Fig. 13. COD removal efficiency of green iron particles.

3.4. Reusability of green iron particles

The reusability of green iron particles were examined by separating the utilized iron particles after decolorization, using decantation process and recycling further. The experiment was repeated for three times so that the results were verified with a standard deviation error of less than 5%. The reusability of green iron was possible

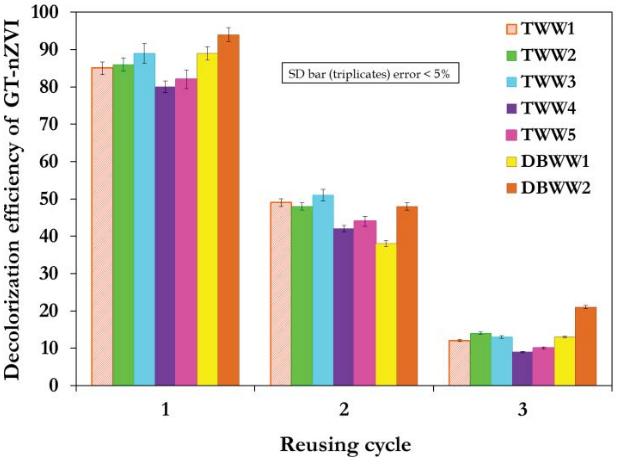


Fig. 14. Reusability of GT-nZVI in wastewater decolorization.

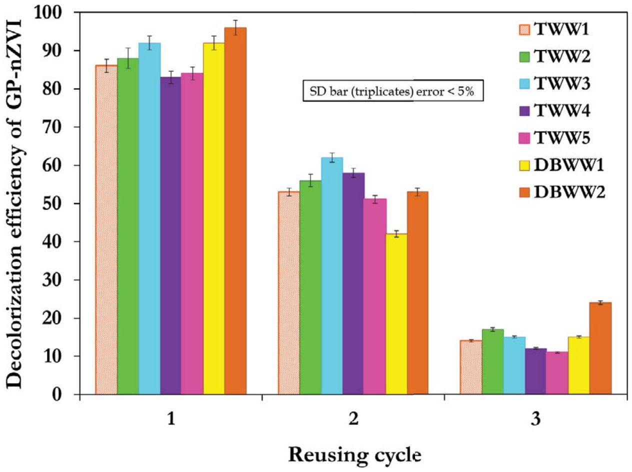


Fig. 15. Reusability of GP-nZVI in wastewater decolorization.

for 3 cycles; however, it showed 38%–51% decolorization in the second cycle of use, as presented in Figs. 14 and 15. GP-nZVI decolorized more than 50% of wastewater for first 2 cycles (Fig. 15). The used GT-nZVI and GP-nZVI particles were scanned using FTIR spectroscopy and the observed the spectrum shows no presence of C=C and O–H function group with the iron particles after few cycle of reuse (Figs. 8 and 9). This confirms that the potential of green iron particles on textile and dyebath wastewater decolorization. Out of the two green iron particles, GP-nZVI demonstrated better performance.

4. Conclusion

2 g/L of green iron was utilized for 80%–92% of textile wastewater decolorization, but 4–6 g/L of green iron of was required to decolorize 63%–89% of highly concentrated dyebath wastewater. The pH of textile (10.1–10.4) and dyebath (10.6–10.8) wastewater was reduced to 8.1–8.9 using GT-nZVI and GP-nZVI. The green iron removed 50%–54% of COD and TOC from the wastewater and concluded that only partial mineralization was possible. The reusability of green iron particles was applicable for first 2–3 cycles; however, it showed very minimal decolorization of 38%–51% in the second cycle of use. The present investigation confirms the feasibility of green iron in textile and dyebath wastewater decolorization.

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