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# Epigallocatechin gallate supported iron particles on dye decolorization and textile wastewater treatment

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**ABSTRACT**

This study investigated decolorization of mono azo methyl orange dye using epigallocatechin gallate supported iron. Camellia sinensis leaves extract and ferric chloride solution were utilized to synthesize the particles and those were characterized for size, crystal nature, structure pattern, elemental composition, attachment of epigallocatechin gallate to iron using particle size analyzer, x-ray diffractometer, transmission electron microscopy, energy-dispersive X-ray spectroscopy, fourier transform infra-red spectroscope, respectively. The results demonstrated that the preferred orientation of ferric iron with epigallocatechin gallate. The maximum dye decolorized was 77% and the solution was analyzed for metabolites using gas chromatography – mass spectrometry. The results demonstrated almost 60% of parent dye molecules were reduced to butadienyl benzene, benzene dicarbonitrile (m/z 128) and 17% were mineralized into 1-buten 4-yl (m/z 55). The observation of 2% parent molecule (m/z 327), 7% dimethyl amino phenyl azo methyl oxy benzene, butyl azo benzene sulfonate (m/z 253) in degraded solution confirmed that still 9% of high molecular structure with azo bond exist, and hence the 77% decolorization efficiency. The application of iron particles in treating real textile wastewater showed 85% color removal in 30 min. This study confirms that epigallocatechin gallate supported iron could be a promising material for textile wastewater treatment.

*Keywords:* Epigallocatechin gallate; Camellia sinensis; Iron; Textile wastewater treatment; Dye decolorization

## 1. Introduction

Textile dyeing is a well-known polluting industry and provide unsafe environment to more than 2.5 million people [1–4]. This is mainly due to the generation of huge amount of colored wastewater, failure of existing methods to treat colored wastewater [5,6]and discharging partially treated or untreated wastewater into the environment [7–9]. Textile dye is the major pollutant in the wastewater, possesses chromophore and auxochrome bonds, responsible for the color. Thus, the textile dye decolorization in environmental sustainable way needs research, which has been emphasized in recent years [10–12]. It is essential to identify an efficient and economic technique to decolorize dye organic compounds.

The investigation of nano sized iron particles in the removal of organic and inorganic pollutants is emerging in recent years, as the iron particles are highly reactive, non-toxic, and cost-effective [13–15]. The nano sized iron particles aggregate quickly due its magnetic properties, reducing the reactivity towards the pollutant. Addition of supports could enhance the stability of iron particles. The green supports are reported as non-toxic [16] and identified as an environmental friendly capping agent to stabilize iron particles. No studies have reported the decolorization pathway of dye molecules in the presence of epigallocatechin gallate supported iron particles and their application to textile dyeing wastewater treatment. Very limited studies utilized green supported nano iron particles, synthesized from Camellia sinensis leaves extract for dye decolorization [17–20]. However, Hoag et al. [17] and Shahwan et al. [19]

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reported dye decolorization using green supported nano iron particles was possible only in the presence of hydrogen peroxide catalyst.

This study investigated the decolorization pathway of mono azo methyl orange dye using epigallocatechin gallate supported iron. Camellia sinensis leaves extract were utilized to synthesize stabilized nano iron particles. The polyphenolic compounds present in the extract were involved in capping coordination. The synthesized particles were characterized for its size, crystal nature, structure pattern, elemental composition, polyphenolic compounds attachment to iron. The factors involved in the maximum decolorization of the dye were studied. The variation in pH throughout dye decolorization was monitored. The decolorization mechanism was analyzed, the degradation products were identified and the respective decolorization pathway is proposed. The particles were analyzed for reusing capacity and verified for the potential of textile wastewater treatment.

## 2. Materials and methods

### 2.1. Materials

Ferric chloride ($FeCl_3$), methyl orange ($C_{14}H_{14}N_3NaO_3S$, C.I. 13025), methanol, hydrochloric acid, and sodium hydroxide were all purchased from Merck India Private Ltd. All the chemicals used in this study are of analytical grade and utilized without further purification.

### 2.2. Synthesis of epigallocatechin gallate supported iron particles

The Camellia sinensis leaves were collected, thoroughly washed with deionized water, dried under shade, and finely powdered. Mixing 100 g of leaves powder into 1 L of deionized water, heating to 80°C for one hour in water bath facilitated in the preparation of 100 g/L concentration of leave extract. Then the extract was cooled down to room temperature and vacuum filtered. A solution of 0.5 M $FeCl_3$ was prepared. The solid ferric iron hydrolyzed water and contributed hydrogen ion to lower pH of the solution. The acidic pH was observed in $FeCl_3$ solution. The hydroxide ions combined with $Fe^{3+}$ and produced ferric oxides and hydroxides.

The prepared $FeCl_3$ solution was mixed to leave extract in a 1:1 volume ratio and the black precipitates were observed. When $Fe^{3+}$ ions were added to the polyphenolic compounds in extract, the weak hydroxyl bonds attached to the polyphenolic compounds were modified. The trivalent iron replaced hydrogen ions; bonded to the oxygen and hydroxide molecules. The observed precipitates confirmed the attachment of polyphenolic compounds to iron particles. Jovanovic et al. [21] reported that the presence of epigallocatechin gallate polyphenolic compound is large amount in Camellia sinensis leaves extract. The formation of epigallocatechin gallate supported iron particles is illustrated in Fig. 1. The synthesized particles were vacuum filtered, dried, stored in vials, and kept in a desiccator until further use.

### 2.3. Characterization of epigallocatechin gallate supported iron particles

The particle size was observed based on dynamic light scattering technique using nano particle analyzer SZ-100 (Horiba, Japan). The crystal structure was examined using Ultima-IV X-ray diffractometer (Rigaku, Japan) with Cu Kα radiation. The operating voltage and current was maintained at 40 kV and 30 mA. The size, morphology, and the elemental analysis were studied using transmission electron microscopy (TEM) with Energy-dispersive

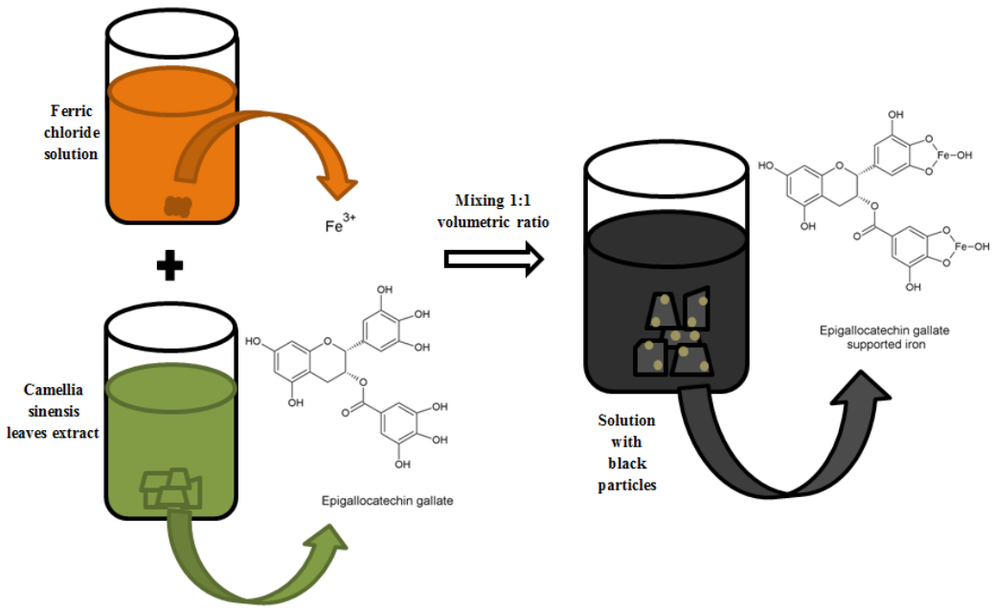



Fig. 1. Schematic presentation of epigallocatechin gallate supported iron synthesis.

X-ray spectroscopy (EDS), which was acquired using Tecnai G2 Spirit (FEI, Netherlands). The functional groups attached to freshly synthesized and used particles were monitored using the spectrum of Fourier transform infrared (FTIR)spectroscope (Perkin–Elmer Instrument Co. Ltd., USA).

### 2.4. Decolorization experiment

Methyl orange was utilized as a model dye to understand the behaviour of epigallocatechin gallate supported iron particles on decolorization. The dye contains mono azo class (–N=N-) chromophore as well as sulfonate (–$SO_3H$) auxochrome bonds, responsible for its color. It is an anionic dye. 100 mg/L of stock solution was prepared and the serial dilutions were made from the stock solution as required. The standards of 1.0–30.0 mg/L of dye was prepared from the stock solution and the respective absorbance were monitored using UV-Vis spectrophotometer (Jasco Inc., Japan) at $\lambda_{max}$ = 464 nm and at $\lambda_{max}$ = 505 nm. The standard calibration curves were utilized to determine dye concentrations before and after the treatment with epigallocatechin gallate supported iron. The reaction between dye molecules and epigallocatechin gallate supported iron particles was studied by adding the particles to 30 mL dye solution. The initial pH of dye was observed as 6.0. Digital pH meter was utilized to monitor the pH. The decolorized solution was collected at every 5, 10, 20, 30, 60, 120, and 180 min. The supernatant was utilized to monitor change in dye concentration, after centrifugation and decantation process. All experiments were done in triplicate to obtain the results with an error <5%. The dye decolorization efficiency was calculated as follows in Eq. (1):

$$\text{Dye decolorization efficiency } (\%) = \left(1 - \frac{C_t}{C_o}\right) \times 100 \quad (1)$$

where $C_0$ = dye initial concentration, and $C_t$ = dye concentration after reaction time in minutes. The decolorization was investigated by varying epigallocatechin gallate supported iron dose (0.2–2.0 g/L), initial dye concentration (20–100 mg/L), and pH (2–10). The pH of the dye was varied using sodium hydroxide solution (1.0 M) or hydrochloric acid (1.0 M). At an optimized condition, the maximum dye decolorization was achieved and the concentrate of the degraded dye solution was collected using a rotary evaporator. The residue was preserved using ethtanol and analyzed using a gas chromatography – mass spectrometry (GC-MS) QP 2010 plus (Shimadzu, Japan). The degraded dye intermediates and products were observed and the respective decolorization pathway is proposed.

## 3. Results and discussion

### 3.1. Characterization of epigallocatechin gallate supported iron particles

Most of the particles size was in the range of 140–160 nm. The size distribution is presented in Fig. 1a. The X-ray diffraction pattern had no indication of any peaks along 2θ, shown in Fig. 1b. This revealed that the particles were amorphous in nature due to the preferred orientation of iron particles with epigallocatechin gallate. Hence the morphology showed irregular clusters of epigallocatechin gallate capped iron particles presented in Fig. 1c. The structure pattern indicated that the aggregation of iron particles was prevented using green supports. The EDS analysis confirmed the presence of high carbon and oxygen content in addition to iron element. The FTIR spectra recorded several peaks in the range of 500 $cm^{-1}$ to 4000 $cm^{-1}$ presented in Fig. 1d. The freshly synthesized particles showed broad peak at 3161 $cm^{-1}$ and sharp peak at 1606 $cm^{-1}$, those confirmed the presence of –OH bonds and C=C stretching vibration. The medium intense peak at 1415 $cm^{-1}$ was recognized as in-plane bending vibration of –OH bond in synthesized particles. The peaks at 842 $cm^{-1}$ and 798 $cm^{-1}$ were related to C-O-C stretching vibration and aromatic rings. The FTIR spectrum of utilized epigallocatechin supported nano iron particles showed that the peaks were disappeared (Fig. 1d) after several cycles of dye decolorization experiments.

### 3.2. Decolorization of methyl orange dye

The initial dye concentration was fixed as 20 mg/L. The maximum dye color removal was 77% at epigallocatechin gallate supported iron dose of 1.0 g/L. This is due to the release of ferric iron from epigallocatechin gallate compound into the dye solution. The ferric iron in dye solution hydrolyzed water and released hydrogen ions. The hydrogen ions were bonded to nitrogen in the azo bond, formed azonium ions in the dye structure. This aided cleavage of azo and auxochrome bond in the dye structure. If the dose was increased to 1.2 g/L, 1.5 g/L, 2.0 g/L, the dye removal was reduced to 71%, 64%, and 59% respectively. This is due to the maximum release of ferric iron generated large amount of hydrogen ions, increased pH of dye solution. Thus the intensity of chrome was increased in dye solution. Hence, the dose was optimized to attain maximum decolorization, presented in Fig. 3a. The addition of epigallocatechin gallate supported iron to the dye solution reduced its initial pH 6.0. This was confirmed by recording the pH throughout the decolorization experiment and it is shown in Fig. 3b. All reactions attained equilibrium within 60 min of reaction. The pH was monitored at 5, 10, 20, 30, 60 min of reaction time. The initial pH was reduced to 2.8 in 5 min and it was again reduced to 2.5 until 30 min. The generated hydrogen ions set up the reduction process. The hydroxide ions were attached to nano iron particles and formed oxide, hydroxide layers on the surface. After 30 min of reaction time, the pH was increased to 3.5. This demonstrated that the amount of hydroxide ions not utilized was increased. After reducing dye molecules, all the nano iron particles were converted into iron oxides and iron hydroxides, and then induced adsorption process. The dye decolorization efficiency for different epigallocatechin gallate supported iron dose was presented with respect to reaction time in Fig. 3c. The initial concentration of dye was varied from 20 to 100 mg/L with the optimized 1.0 g/L dose of epigallocatechin gallate supported iron and showed 53% of dye decolorization when the dye concentration was increased to 100 mg/L. More dye molecules required higher dose of epigallocatechin gallate supported iron particles to get the maximum decolorization. At the minimum initial concentration,

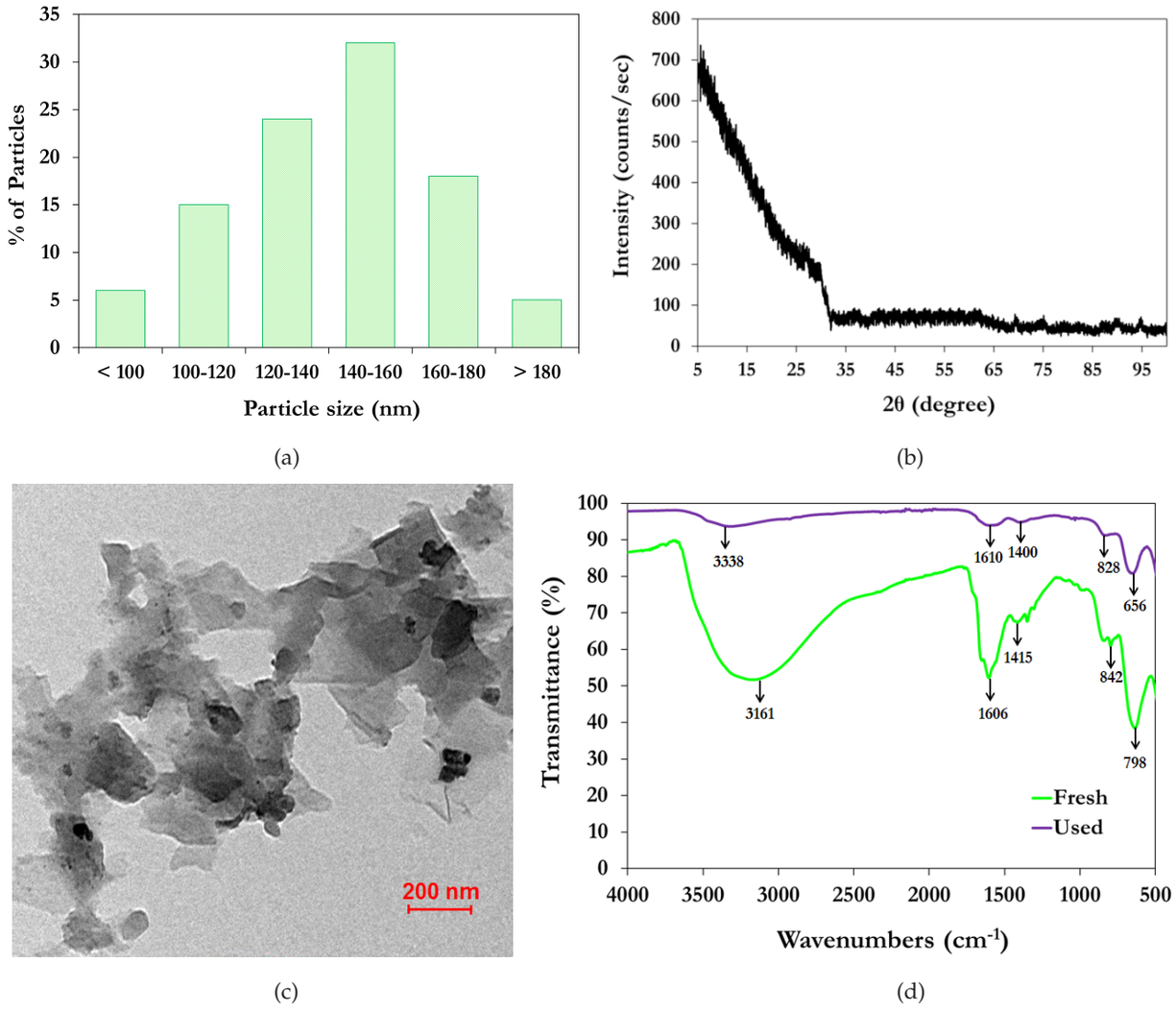



Fig. 2. Epigallocatechin gallate iron characterization: (a) Particles size distribution (b) XRD pattern (c) TEM image (d) FTIR spectra.

the maximum dye decolorization was observed, shown in Fig. 3d. The effect of pH on dye decolorization was studied by varying initial pH to 2, 4, 6, 8, and 10. In acidic pH, the decolorization was largely accomplished by the reduction of dye molecules. In basic pH, it was attributed to the adsorption, as discussed earlier. The respective changes in decolorization efficiency due to different initial pH are presented in Fig. 3e. The study showed 81% to 73% decrease in decolorization efficiency, when the pH was increased from 2 to 10. The influence of pH is very low in decolorization when compared to initial dye concentration and epigallocatechin gallate supported iron dose. All reactions reached equilibrium in 30 min of reaction time and no changes in dye decolorization were observed after 60 min.

The UV-Vis spectra of dye solution before and after reaction with epigallocatechin gallate supported iron are presented in Fig. 3(f). The strong absorbance bands at 464 nm and 252 nm confirmed the presence of azo bond and aromatic ring in methyl orange dye structure. The spectral band at 464 nm disappeared suddenly due to the addition of epigallocatechin gallate supported iron and a new band at 505 nm was formed. Such a red shift (from 464 to 505 nm) termed as bathochromic shift, was observed clearly in Fig. 3f. This occurred due to protonation of nitrogen in azo bond, as discussed earlier in this section and also reported in previous studies [19,22]. The appearance of a new band at 247 nm confirmed the formation of benzoquinone compounds, as a result of dye decolorization. The intensity of chrome in dye solution was reduced with respect to reaction time but complete decolorization was not achieved due to the permanent shift of chrome. This might be specific to this mono azo methyl orange dye, could be verified with other diazo and anthraquinone dyes by further studies.

### 3.3. Decolorization mechanism analysis

The decolorization mechanism was studied by identifying intermediates, and the products using GC-MS results. Several peaks were observed from retention time

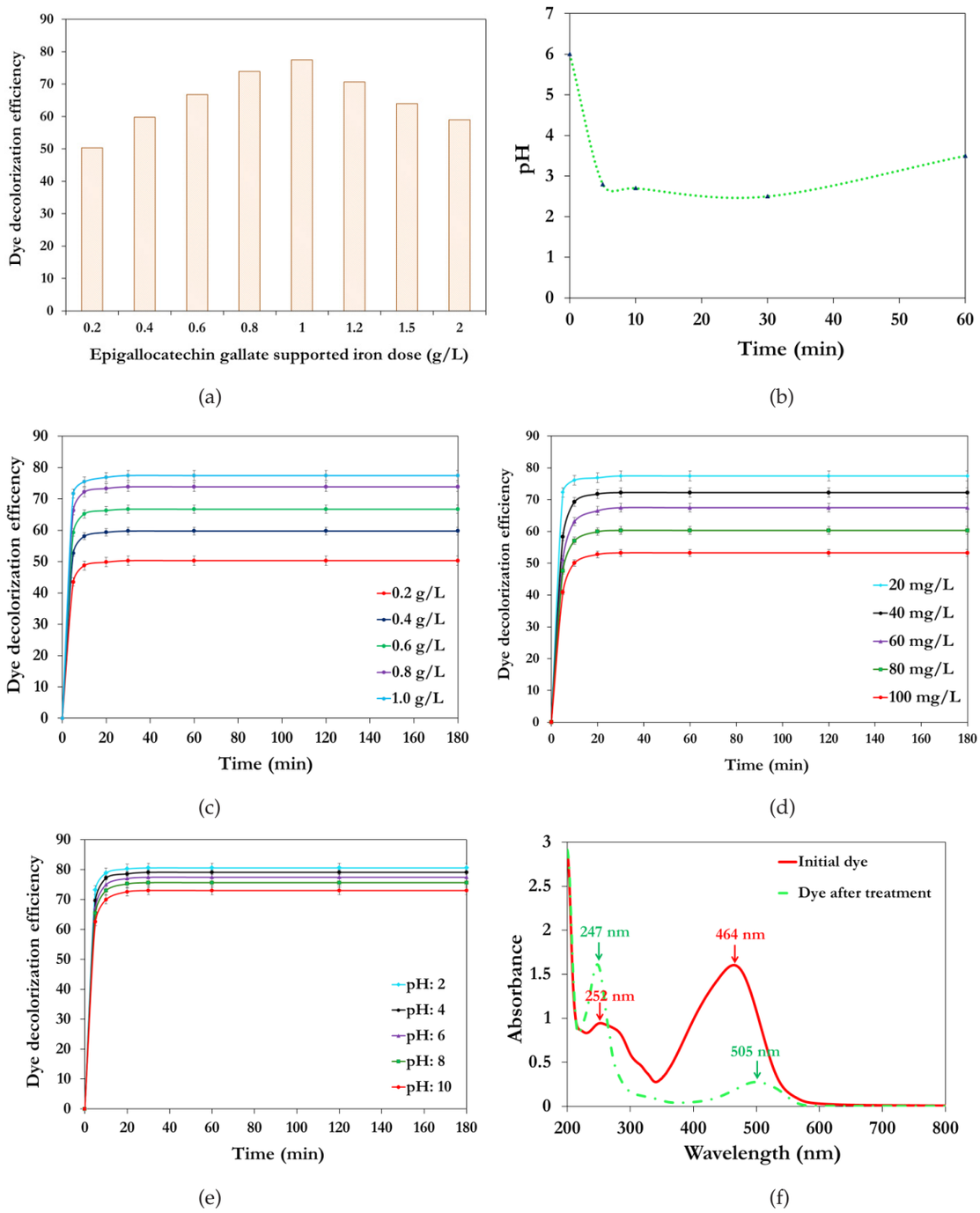
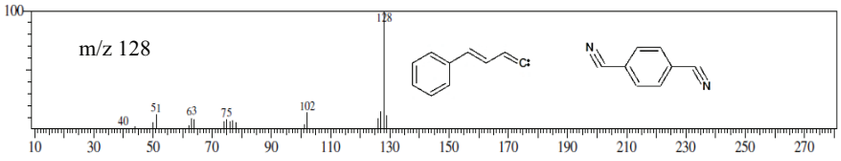
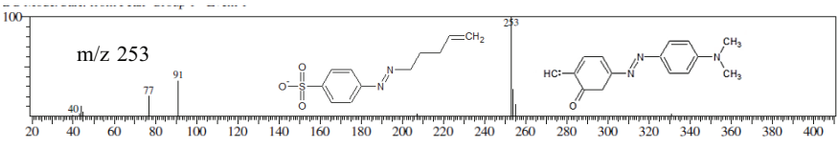
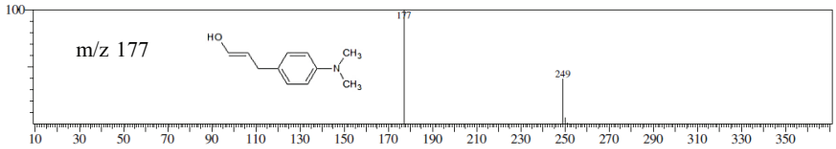
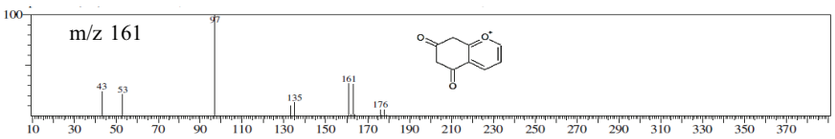
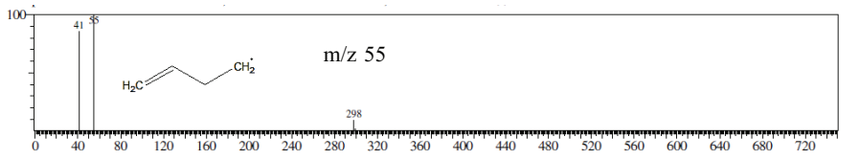
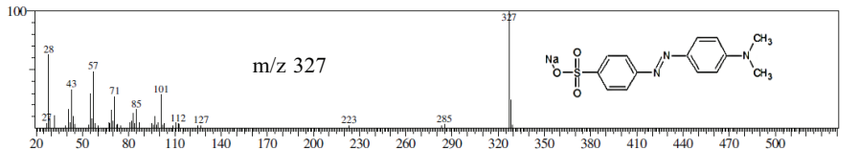
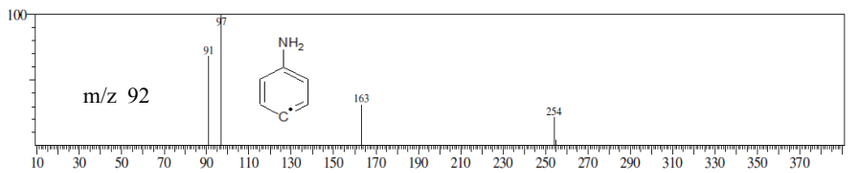
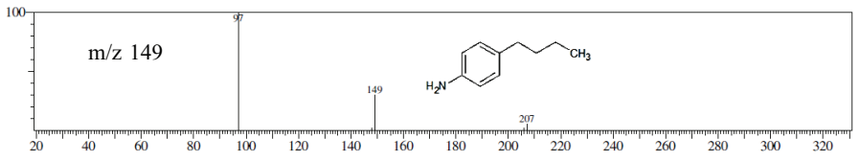



Fig. 3. (a) Epigallocatechin gallate iron dose optimized for maximum dye decolorization (b) Variation of pH during dye decolorization (c) Effect of epigallocatechin gallate iron dose (d) Effect of dye initial concentration (e) Effect of pH (f) UV-Visible spectra of dye before and after treatment with 1.0 g/L of epigallocatechin gallate supported iron – 20 mg/L initial dye concentration and pH: 6.

of 9.89–29.26 min. Through m/z ratio, two large peaks of butadienyl benzene or benzene dicarbonitrile (m/z 128), 1-buten 4-yl (m/z 55) as well six small peaks of dimethyl amino phenyl azo methyl oxy benzene or butyl azo benzene sulfonate (m/z 253), di methyl amino propyl hydroxyl benzene (m/z 177), 4-oxochromen-2-olate (m/z 161), parent molecule methyl orange (m/z 327), 4-amino phenyl (m/z 92), 2-methyl-phenylpropan-5-amine were identified is presented in Table 1. The relative abundance of the intermediates and products in the dye solution was also reported. With the presence of eight parent, transformation and degraded products, it is possible to under-

Table 1
Intermediates and products identified by GC-MS

| Retention time (min) | Mass spectrum | Relative abundance (%) |
|---|---|---|
| 9.89 |   | 60 |
| 17.73 |   | 7 |
| 23.64 |   | 5 |
| 24.13 |   | 3 |
| 25.17 |   | 17 |
| 26.39 |   | 2 |
| 28.22 |   | 3 |
| 29.26 |   | 3 |

stand the dye decolorization pathway proposed in Fig. 4. The parent molecule sodium dimethyl amino phenyl azo benzene sulfonate (m/z 327) was modified into dimethyl amino phenyl azo methyl oxy benzene (m/z 253) and butyl azo benzene sulfonate (m/z 253) and the epigallocatechin gallate supported iron compound was disintegrated into 4-oxochromen-2-olate (m/z 161). The compound m/z 253 was reduced to di methyl amino propyl hydroxyl benzene (m/z 177) and sulfanilic acid (m/z 172). The organic compounds m/z 177 and m/z 172 were again reduced to 2-methyl-phenylpropan-5-amine (m/z 149), benzene dicarbonitrile (m/z 128) and 4-amino phenyl (m/z 92).

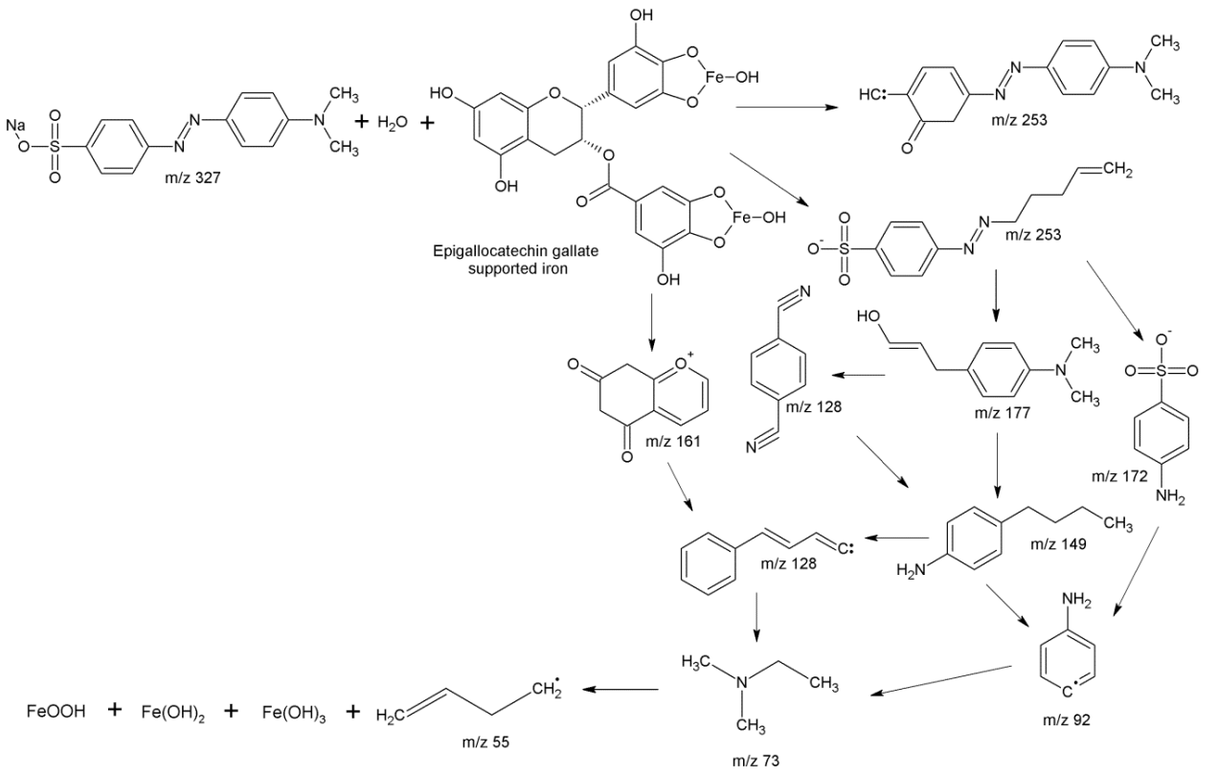



Fig. 4. Proposed dye decolorization pathway using epigallocatechin gallate supported iron.

The compounds m/z 161 and m/z 149 were reduced to butadienyl benzene (m/z 128). The compounds butadienyl benzene (m/z 128) and 4-amino phenyl (m/z 92) were degraded into di-methyl-ethyl-amine (m/z 73), which was again mineralized into 1-buten 4-yl (m/z 55). Further mineralization into $CO_2$ and $H_2O$ may be possible; however they were not detected in mass spectrometry. Sixty percent of dye molecules were degraded into benzene dicarbonitrile (m/z 128), butadienyl benzene (m/z 128) and 17% of dye molecules were mineralized into lower molecular compounds (m/z 55). Still 2% of parent dye molecules (m/z 327) were observed in degraded solution and hence the decolorization efficiency was 77%.

### 3.4. Reusability of epigallocatechin gallate supported iron

The reusability of 1.0 g/L epigallocatechin gallate supported iron in treating 30 mL dye of 20 mg/L concentration at its initial pH 6 was studied. The green support was dissolved after several uses of dye decolorization and without capping agent the reactivity of iron particles was reduced, thereby the decolorization efficiency. The FTIR spectrum of the particles after several cycles of use on dye decolorization (Fig. 2d) was discussed already in section 3.1. The reusing capacity of epigallocatechin gallate supported iron for several dye decolorization experiment is illustrated in Fig. 5. The first 3-4 cycles achieved more than 50% dye decolorization and further usage reduced decolorization ability to 34%, 29%, 13%, 3% respectively.

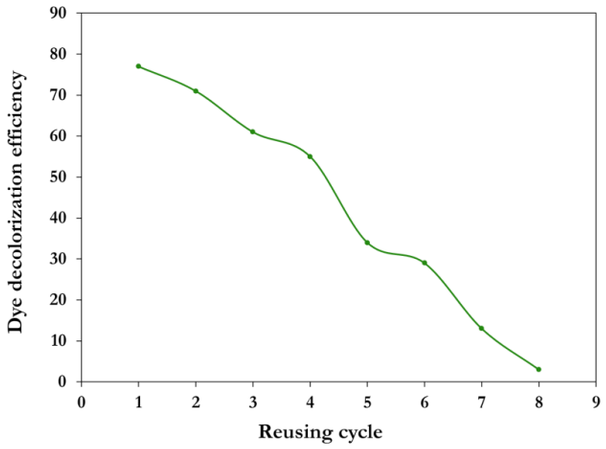



Fig. 5. Epigallocatechin gallate supported iron reusing capacity.

### 3.5. Decolorization of real textile dyeing wastewater

The efficiency of epigallocatechin gallate supported iron in treating real textile dyeing wastewater was evaluated. The textile dyeing wastewater was collected from an industry at Tiruppur, Tamil Nadu, India. A dose of 1.5 g/L of epigallocatechin gallate supported iron was added to 30 mL of wastewater. The initial pH 9.0 of wastewater was reduced to 7.4 and 85% color removal was observed in 30 min of reaction time. Hence this study demonstrated that

epigallocatechin gallate supported iron could treat real textile dyeing wastewater efficiently.

## 4. Conclusion and future perspectives

Epigallocatechin gallate supported iron were synthesized using Camellia sinensis leaves extract, non-toxic, green method. The synthesized particles were characterized for size, crystal nature, structure pattern, elemental composition and attachment of epigallocatechin gallate to iron. The dye decolorization efficiency of epigallocatechin gallate supported iron was investigated by varying dose (0.2–1.0 g/L), initial dye concentration (20–100 mg/L) and pH (2–10). The dye was decolorized for 77% and bathochromic shift of 464 to 505 nm was observed. In addition, the reduction in pH of dye as well protonation of azo bond in dye structure was noticed. The decrease in dye concentration was reported but no complete decolorization was observed, which confirmed that chromophore and auxochrome bonds were removed and modified. This chrome shift is particular to methyl orange dye molecules. It has to be verified with further study on widely using reactive dyes in textile dyeing industries. The reusability of epigallocatechin gallate supported iron is possible for 3-4 cycles, since the attached epigallocatechin gallate was diminished after several uses. Almost 60% of dye molecules were reduced to butadienyl benzene, benzene dicarbonitrile (m/z 128) and 17% of dyes molecules are mineralized into lower molecular compound 1-buten 4-yl (m/z 55). The observation of 2% of parent molecule (m/z 327) and 7% of dimethyl amino phenyl azo methyl oxy benzene, butyl azo benzene sulfonate (m/z 253) in degraded solution confirmed that still 9% of high molecular structure and azo bond exist, hence the 77% decolorization efficiency. The application of epigallocatechin gallate supported iron in treating real textile dyeing wastewater showed 85% color removal in 30 min. This study confirms that epigallocatechin gallate supported iron could mineralize dye molecules into lower molecular compounds and have the potential to treat textile dyeing wastewater efficiently.

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