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Treatment of methylene blue in aqueous solution by electrocoagulation/micro-crystalline cellulosic adsorption combined process

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ABSTRACT

The continuous application and consequent discharge of dyes from a variety of industries cause severe ecological outcomes. The research reports the treatment of a vastly applied textile dye methylene blue (MB) by electrocoagulation (EC)/micro-crystalline cellulosic (MCC) adsorption combined process. The influence of operating parameters of pH (5–9), current density (7.40–33 mA/cm^2), MCC dose (0–7 g/L), and dye concentration (25–75 mg/L) on the decolorization efficiency was studied. The MCC surface characterization with elemental composition and functional group detection was performed by scanning electron microscopy–energy-dispersive X-ray and Fourier-transform infrared spectroscopy. A profound effect of MCC dose was observed on the decolorization efficiency with 87.56% at zero doses to 99.9% at a dose of 7 g/L in EC/MCC adsorption combined process in a pyrex glass reactor. At optimal conditions, pH 9, current density 22.15 mA/cm^2, MCC dose 5 g/L, and C_0 = 50 mg/L, the 98.75% MB removal efficiency was achieved. The study explored the effective performance of MCC in the EC combined process and potential for the abatement of dye effluents.

Keywords: Adsorption; Cellulose; Electrocoagulation; Methylene blue

1. Introduction

Water is the fundamental necessity for every living organism and one of the most precious assets on earth. The usage of water is increasing owing to the hasty industrialization, growing population, and urbanization [1]. Water pollution has become the most significant environmental issue and it is escalating at an alarming rate due to the industrial usage of dyes and heavy metals. The textile industry plays a vital role in the social, economic, and cultural development of nations worldwide [2]. Due to the wide-ranging volume of water being utilized in dyeing and finishing processes, textile industries are one of the principal originators of wastewater [3,4]. The treatment of wastewater is a challenging topic from a global point of view.

Dyes are organic compounds that adhere to surface and impart color to the fibers frequently used in textile industries and discharged in effluents. Dyes are low biodegradable due to high molecular weight and complex structures and are harmful to aquatic life as well as for human beings

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[5]. Approximately 700,000 tons of dyes are manufactured annually, and 2%–3% dyes are lost into industrial wastewater [6]. The average concentration of dyes in real dye-house wastewater discharges varies 0.01–0.25 g/L [7]. Organic compounds that originate from dyes can be the root cause of allergies, human mutations, skin irritation, and even cancer and are fatal to aquatic life [7–9]. It is therefore imperative to eliminate dyes from the outlet streams before discharge into watercourses. Many conventional processes through physical and chemical methods are used for dye removal such as Fenton reagents, adsorption, chemical oxidation, ozonation, coagulation, ion exchange, membrane separation processes, biological degradation, and electrochemical methods [10–15]. Recently the advanced oxidation processes (AOPs) have gained much attention for the treatment of organics in the wastewater. AOPs operate through the generation of super acting hydroxyl radicals which rapidly attack the organics in the wastewater due to highly reactive and non-selective nature [16–18]. Numerous AOPs such as ozonation, Fenton processes, Photocatalysis, and electro-oxidative processes have been found efficient for the abatement of organics in the wastewater. However, these possess some disadvantages like the high cost of O_3, low water solubility, and the formation of some hazardous by-products in solutions are observed in ozonation based processes [16–18]. Sometimes the presence of high radical scavenging agents in the real wastewater suppresses the application of AOPs [17,18]. Fenton based processes may generate unwanted iron sludge and require low pH levels which are not practical [17,18]. The electrocoagulation and adsorption are the two eco-friendly and cost-effective techniques for wastewater treatment [19]. The combined process application reduces capital cost and treatment time requirements [19].

Electrocoagulation is an electrochemical technique in which current is passed through cathode and anode which are submerged in an electrolyte. The electric current results in the dissipation of the anode within the wastewater with the evolution of gas bubbles [20]. Agglomerates of hydroxide complexes are formed, and particulates are neutralized with the coagulant flocs removed by flotation or sedimentation. The electro-coagulation phenomenon provides a reliable, environmentally compatible, and cost-effective approach for wastewater treatment [21,22].

The adsorption technique offers the flexibility, low cost, ability to handle toxic compounds, ease of operation, and inhibits the formation of harmful substances [23]. Due to the high cost of available commercial adsorbents, biosorbents are focused and attractive due to the renewability, low cost, easy and abundant availability. Micro-crystalline cellulose (MCC) is an organic, renewable biodegradable polysaccharide and thermoplastic substance formed by the esterification of natural cellulosic materials such as wood, cotton, recycled paper, and sugar-cane. MCC has shown high adsorptive capabilities in previous research works [24–26]. MCC is a lignocellulosic material with salient features like non-toxicity, biodegradability, renewability, crystallinity, water insolubility, and good mechanical strength which make it an effective green adsorbent for the organic pollutants [24,27]. MCC has been applied as an adsorbent for dye treatment in previous research reports [24,25,28]. The present work involves the combined application of MCC adsorption and electrocoagulation processes for the treatment of methylene blue (MB) dye.

The current research examines the performance of the electrocoagulation (EC)/MCC adsorption combined process for the treatment of vastly applied textile dye MB in aqueous solution. The study explores the potential of MCC for dye abatement in the EC combined process. MCC was characterized by pH_{pzc}, scanning electron microscopy (SEM), energy-dispersive X-ray (EDX) and Fourier-transform infrared spectroscopy (FTIR) and Brunauer–Emmett–Teller (BET) techniques. The effect of operating parameters of pH (5–9), current density (16–33 mA/cm^2), MCC dose (0–7 g/L), and dye concentration (25–75 mg/L) on the decolorization efficiency was studied. A kinetic study was performed.

2. Materials and methods

2.1. Chemicals

MCC was purchased from Eisen-Golden Laboratory (USA) in powdered form. MB was purchased from AK Scientific, Inc., (USA). Aluminum electrodes were purchased from the local market. NaCl, NaOH, and HCl were procured from (Merck, USA).

2.2. Experimental and analytical methods

The experimental setup mainly consists of an aerated electrochemical reactor containing two aluminum electrodes (Fig. 1). The anode and cathode were arranged in a vertical position 1 cm apart and attached to a digital power supply. The electrodes were smoothed with fine-grained sandpaper, rinsed with 1N HCl and distilled water before each experimental run. MB solution (volume 1 L) was prepared before each run by dissolving a fixed amount of dye in distilled water. The batch experiments were performed at 25°C by taking 1L of MB solution, adding 2 g of NaCl electrolyte, and supplying air to the base of the reactor. During each experimental run, continuous aeration was provided at a rate of 3 L/min for agitation and flotation purposes. The current was set and then coagulation was initiated. Samples were taken after specific time intervals, permitted to settle, filtrated, and then analyzed using a UV/VIS Spectrophotometer (Perkin Elmer 35, US), the concentration of MB was estimated at 665 nm. The color removal efficiency Y_{COL} was determined as [29]:

$$Y_{COL}(\%) = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

where C represents the concentration of MB before and during experimentation (mg/L).

2.3. Characterization techniques

The characterization of MCC morphology and composition was inspected by SEM, and EDX using the Shimadzu XD-3 (Japan) analyzer before and after adsorption. The FTIR spectrum of the MCC before and after adsorption was obtained by Shimadzu 8400s spectrophotometer. The measurement of pH_{pzc} of cellulose was carried out by pH titration

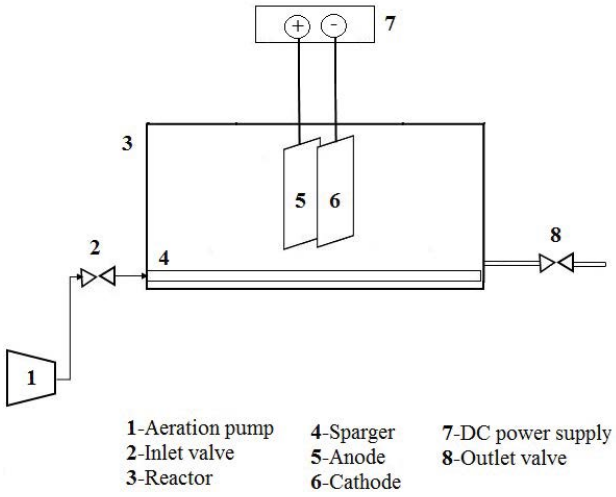

Fig. 1. Reactor setup.

procedures. The surface area of cellulose was determined by the BET analyzer (PMI, USA).

3. Results and discussion

3.1. Characterization results

The SEM images and EDX spectrum of MCC before and after adsorption are shown in Fig. 2. The MCC showed an irregular porous surface before adsorption (Fig. 2a) and a considerable change in the structure with surface coverage was observed after adsorption (Fig. 2b). The EDX spectra show the variation in peak intensities of the particular component before and after treatment (Fig. 2) and additional peaks were also observed in Fig. 2b which appear after adsorption analysis, which depicts the alteration of the aspects of the cellulose and confirms the adsorption process. The EDX composition of MCC is given in Table 1.

The FTIR spectrum of MCC before and after treatment is given in Fig. 3. The strong peak encompassing 3,700–2,882 cm^{-1} depicts the hydroxyl groups – (OH-stretching), the

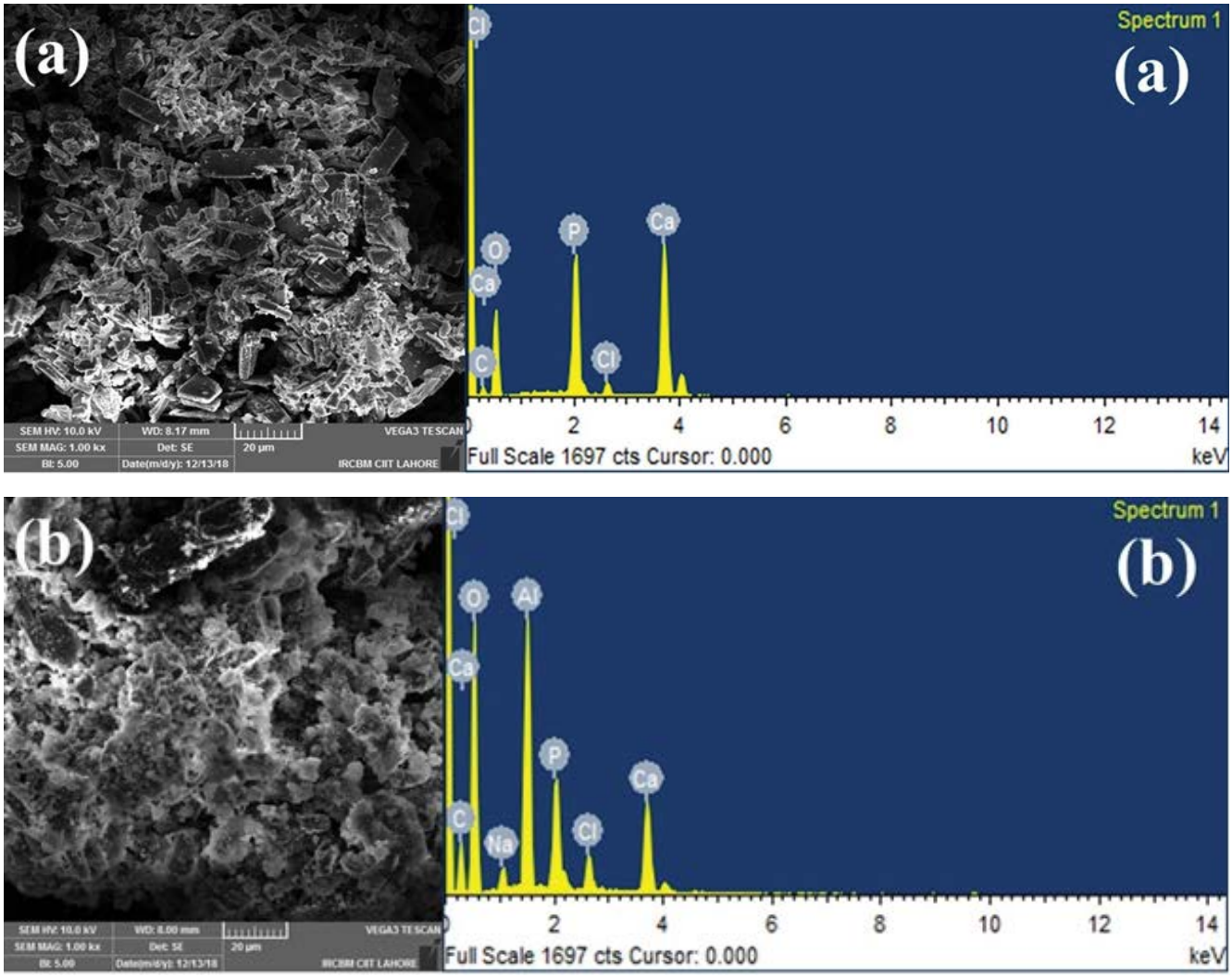

Fig. 2. SEM-EDX of MCC (a) before and (b) after adsorption.

Table 1
EDX composition of MCC

Element	Weight %	
	Before adsorption	After adsorption
C	11.27	20.99
O	53.30	54.13
P	13.28	5.19
Cl	1.43	1.87
Ca	20.72	5.75
Na	–	1.39
Al	–	10.68

bond at 2,000–1,500 cm^{-1} is associated with the occupancy of the C–H bond [19,30]. The peaks of 1,732 and 1,655 cm^{-1} are attributed to as the carbonyl group –C=O, 1,508 cm^{-1} denotes the aromatic rings, while 1,215 and 1,470–1,430 cm^{-1} incorporated with the C–O in phenols and $–CH_3$ respectively [19,30]. MCC characteristics are given in Table 2.

3.2. Effect of pH

pH has a prime role in both the EC and adsorption processes. In the EC process, the formation of various metal hydroxide species is a function of the prevailing pH. The surface charges on the surface of the adsorbent are strictly controlled by the dominant pH. The decolorization efficiency was studied at different pH values in the range (pH 5–9) and results given in Fig. 4 depict that MB removal was highest at alkaline pH 9. The MB percentage removal was enhanced from 86.11% to 98.75% by pH rise from 5 to 9 after 20 min. The lower MB removal efficiency at acidic zone may be due to the presence of predominant cationic species (Al^{3+} and $Al(OH)^{2+}$) formed at pH 5 in EC process which repel cationic MB dye species while $Al(OH)_3$ formed is insoluble in water at neutral pH which captures the dye molecules and as a consequence, the removal efficiency enhances [21,31]. At pH 9, $Al(OH)_3$ formed are transformed into dominant anionic species $Al(OH)_4^-$, which accelerates the removal efficiency by fast capturing the cationic MB molecules by electrostatic interactions [19]. Moreover, the surface of MCC at pH = 9 > PZC = 6.6 ± 0.2 becomes strongly negatively charged and attracts the cationic MB molecules on the surface by physical and chemical bonding.

3.3. Effect of current density

Current density (CD) is an essential parameter for controlling the rate of electrochemical reactions in the EC process as it determines the formation rate of flocculant. The CD effect on MB removal was studied by applying CD in the range from 7.40 to 33.07 mA/cm^2. The results (Fig. 5) show that by increasing CD, removal efficiency is increased. High CD leads to accession in the amount of metal hydroxide due to the increased anodic dissolution of Al, more mass diffusion, bubble formation, and turbulence which results in the enhancement of MB removal efficiency [31–33]. In comparison with the conventional EC process, cellulose significantly enhances the percentage of removal on the same CD. The MB removal efficiencies of 90.62%, 98.75% and 99.6% were obtained at CD of 16.23, 22.15 and 33.07 mA/cm^2 respectively. At the studied lower CD values of 10.98 and 7.40 mA/cm^2, the MB removal efficiency declined to 80.52% and 73.77% respectively. This may be ascribed to the fact that the rate of electrochemical reactions becomes sluggish at the lower CD values [19]. Moreover, the decreased Al anodic dissolution at lower CD causes a decline in the coagulants production in the solution thereby decreasing the MB removal [19].

The anode consumption and the amount of sludge generated at the studied CD values were examined and results are shown in Fig. 6. It was observed that when the CD values were increased from 7.4 to 33.07 mA/cm^2, the anode consumption increased from 0.575 to 2.08 g/g dye, while the sludge amount increased from 0.29 to 1.68 g/g dye respectively. The increase in the CD causes a faster anodic dissolution by accelerating the electrochemical reactions which in turn increases the anode consumption [19,34]. The rate of Al^{3+} ion generation greatly increases with the CD, which increases the production of metal hydroxide coagulants in the solution and leads to giving more flocs as MB trappers thereby increasing the sludge amount [19,34]. A similar effect was reported in some previous studies [34,35].

All the experimental runs were commenced at 25°C, but a 5°C–6°C rise in the temperature of the solution was observed at the end of the combined process treatment at higher CD values, which may be due to a slight dissipation of electrical energy into heat energy [19]. The same effect has been reported in some previous research works [36,37]. The rise in temperature may have a positive effect on the MB removal efficiency as by the reduction in the solution viscosity and enhancing the ion mobility which accelerates the collisions frequency and MB entrapment by electro-flocs [19,38].

3.4. Effect of MCC dose

The effect of MCC dose on MB removal efficiency was studied on a scale of 0–7 g/L. The results (Fig. 7) show that the addition of MCC as adsorbent has a strongly positive effect on the percentage removal of MB. At 0, 1, 3, 5 and 7 g/L doses of MCC, the achieved removal efficiencies were 87.56%, 92.4%, 96.18%, 98.75%, and 99.52% respectively after 20 min. The larger the MCC dose, the more is the availability of sorption sites, increased diffusion and high availability of surface area are the factors which enhance the dye removal efficiency [31,39,40]. The results presented in Fig. 7 show that at zero dose of MCC, the MB removal efficiency of 87.56% was achieved which enhanced to 98.75% respectively when 5 g/L dose of MCC was employed. Thereby the results show an increase in the MB removal efficiency by 11.19% when 5 g/L MCC dose was added to the reactor at optimum conditions.

3.5. Effect of initial dye concentration

The effect of dye initial concentration was studied by varying the concentration from 25 to 75 mg/L. The results (Fig. 8) show that with an increment in dye concentration,

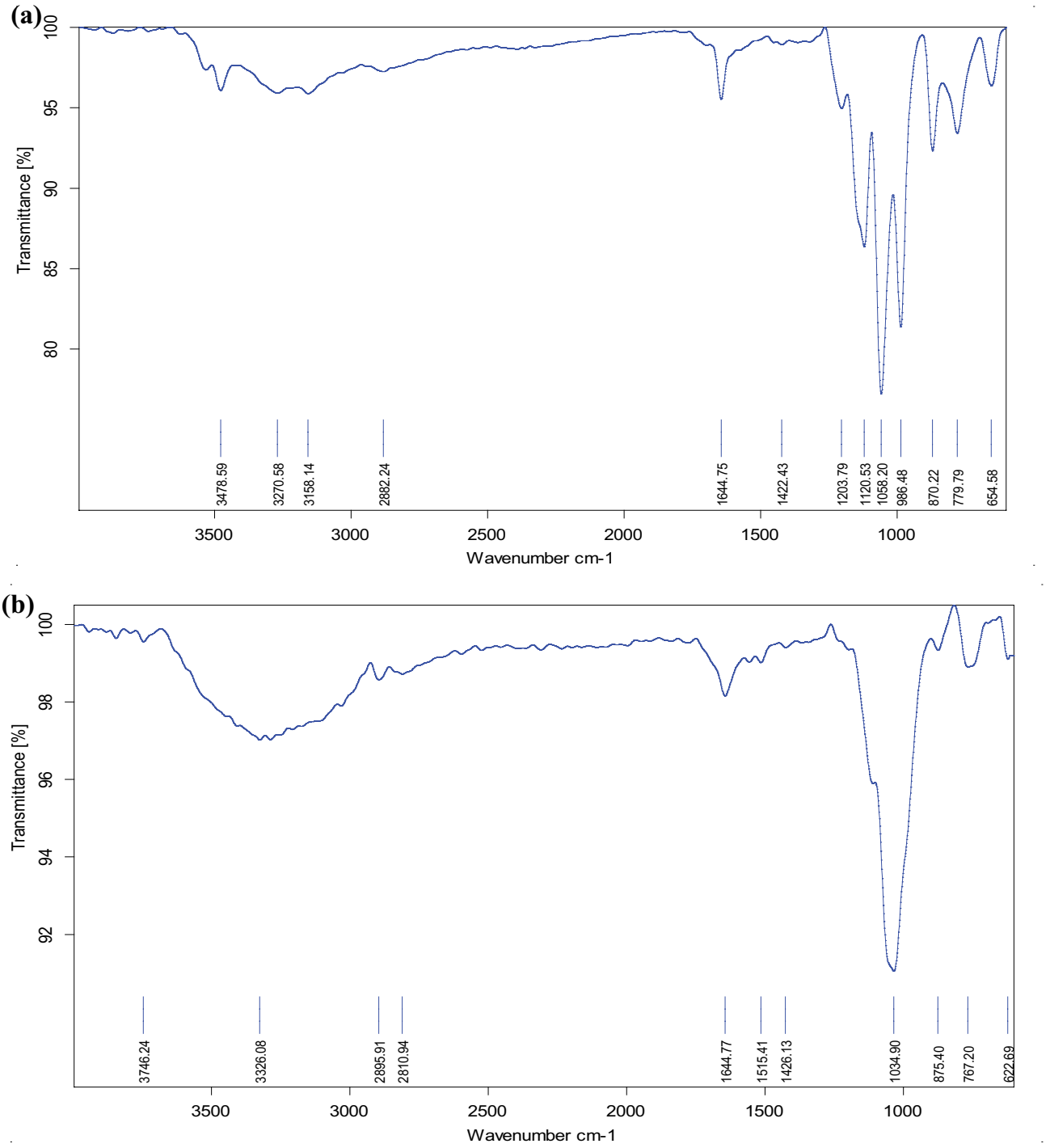

Fig. 3. FTIR of MCC (a) before and (b) after adsorption.

the removal efficiency of dye is reduced. The highest removal percentage (99.82%) at lowest dye concentration (25 mg/L) may be due to the vacant active sites available in the aluminum flocs during the initial stage of EC process but at higher concentration (75 mg/L) removal efficiency is decreased to 91.22%, as the adsorption capacity of flocs becomes insufficient, repulsion of adsorbate occurs due to lack of binding sites [21,41].

Table 2
MCC characteristics

BET area (m^2/g)	0.048
Pore size (cc/g)	18.62
PZC	6.6 ± 0.2

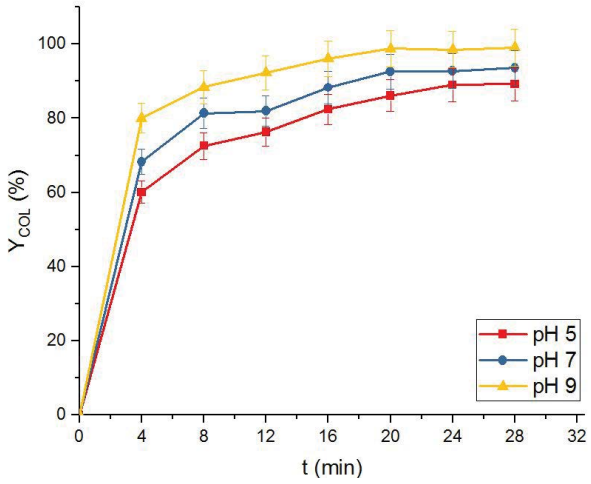

Fig. 4. Effect of pH on MB removal (C_0 = 50 mg/L; CD = 22.15 mA/cm^2, MCC dose = 5 g/L).

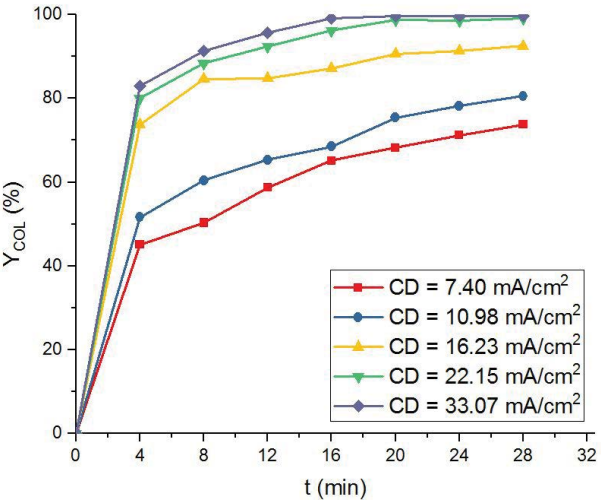

Fig. 5. Effect of CD on MB removal (MCC dose = 5 g/L; C_0 = 50 mg/L; pH = 9).

3.6. Sludge characterization

The sludge generated in the reactor was analyzed by techniques SEM, EDX, and FTIR. The SEM results shown in Fig. 9a reveals a highly porous and gelatinous texture of the surface. Fig. 9b shows the EDX analysis of the sludge and the elemental composition is given in Table 3. The main components found in the sludge were Al 49.11%, O 29.6%, and C 17.91%. Fig. 9c shows the FTIR results of sludge. The broad sharp peak at 3,339.18 cm^{-1} corresponds to the O–H stretches of the formed prime coagulants $Al(OH)_4^-$ in the reactor [42,43]. The peak at 1,625.64 cm^{-1} indicates the H–O–H stretching vibrations [42,43]. The peak at 1,145.17 cm^{-1} indicates the Al–O–H stretch of the generated Al-flocs [42,43]. The peaks in the range 700–800 cm^{-1} indicate the N–H stretching of coagulated dye molecules [42,43].

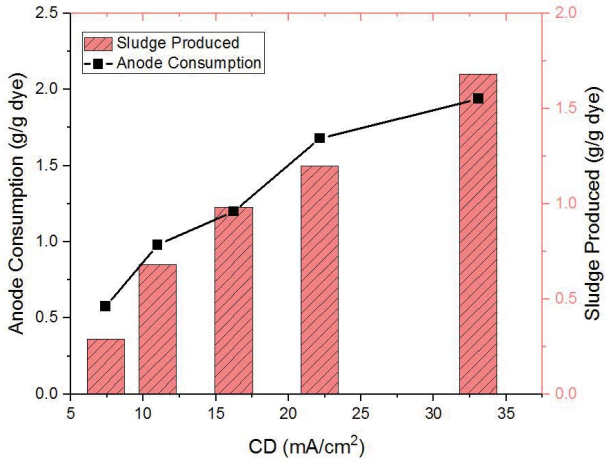

Fig. 6. Anode consumption and sludge produced (MCC dose = 5 g/L; C_0 = 50 mg/L; pH = 9).

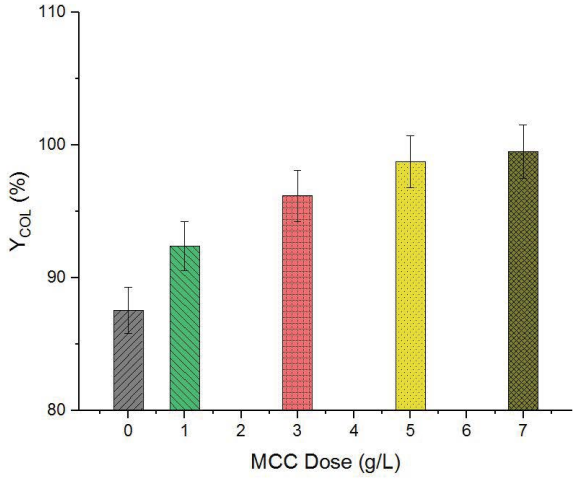

Fig. 7. Effect of MCC dose on MB removal (CD = 22.15 mA/cm^2; pH = 9; C_0 = 50 mg/L; t = 20 mins).

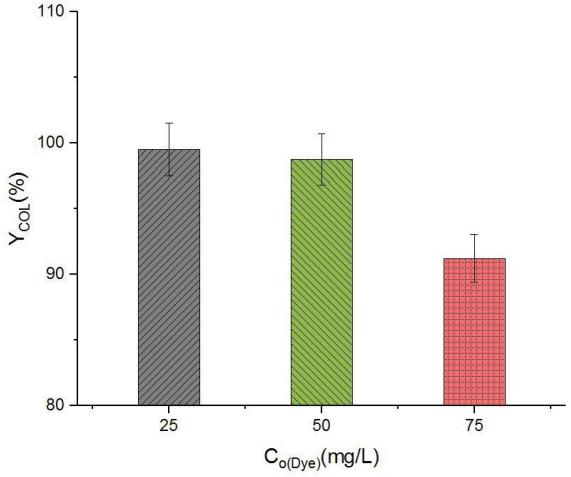

Fig. 8. Effect of initial dye concentration on MB removal (CD = 22.15 mA/cm^2; pH = 9; MCC dose = 5 g/L; t = 20 mins).

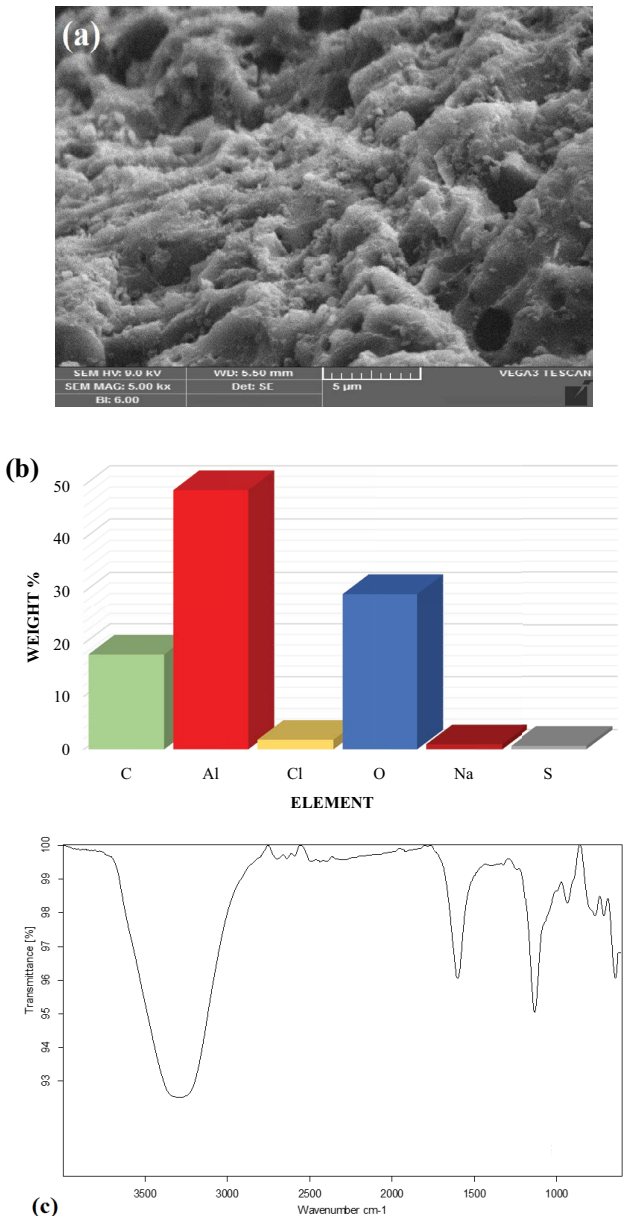

Fig. 9. Sludge analysis (a) SEM, (b) EDX and (c) FTIR.

Table 3
EDX composition of sludge

Element	Weight %
C	17.91
Al	49.11
Cl	1.8
O	29.6
Na	0.94
S	0.64

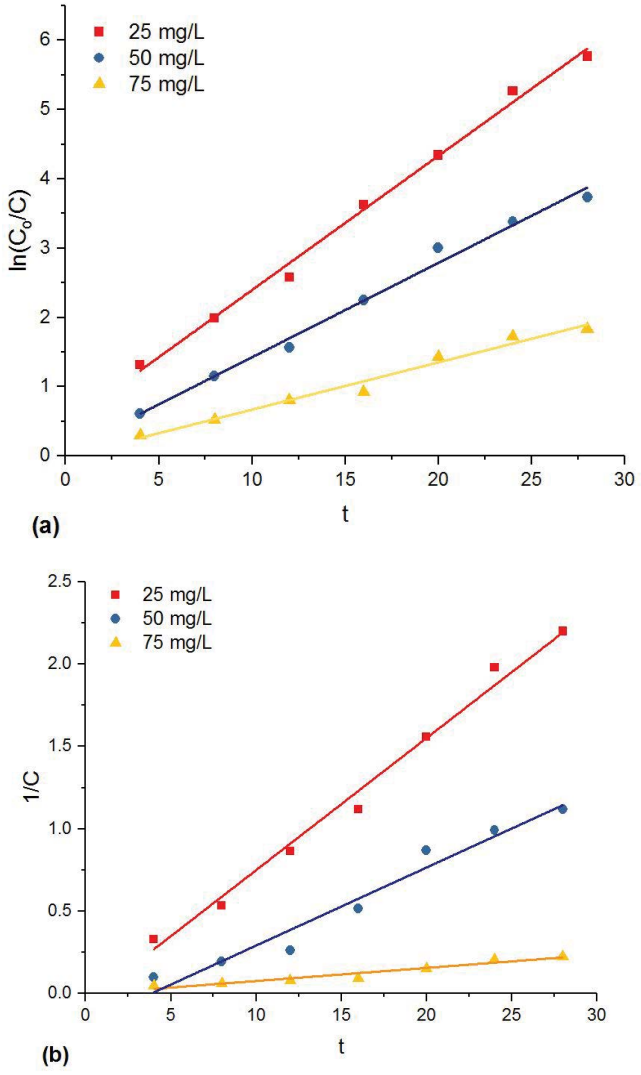

Fig. 10. Kinetic plots of MB decolorization (a) first-order and (b) second-order.

3.7. Dye removal mechanism

The electrocoagulation process is mainly driven by the metal electrode type utilized to generate the electro-flocs, surface texture of produced flocs, and the nature of the dye molecules [19]. The dye adsorption phenomenon is dependent on the surface charges of the adsorbent and the dye, surface area of adsorbent, and nature of adsorbent [7]. In the present study of combined electrocoagulation/MCC adsorption process, the results obtained at the optimum conditions, that is, pH 9, CD 22.15 mA/cm^2, and MCC 5 g/L and the characterization reveals that the electrostatic interactions were the dominant phenomenon in the MB removal. The EC with Al electrodes generates Al-based metal hydroxide in the reactor with $Al(OH)_4^-$ being the most dominant coagulant species at pH 9 [19] which have strong electrostatic interactions for cationic charged MB dye. The SEM results of sludge (Fig. 9a) reveal the uniform porous nature of Al sludge which easily adsorbs the MB dye molecules. The sludge FTIR (Fig. 9b) confirms the dominant availability of Al electro-coagulants in the solution as also

Table 4
Kinetic study parameters

Dye concentration (mg/L)	First-order		Second-order	
	K_1 (1/min)	R^2	K_2 (L/mg min)	R^2
25	0.194	0.993	0.081	0.974
50	0.136	0.987	0.047	0.952
75	0.0681	0.974	0.0075	0.94

confirmed by EDX results (Fig. 9c). Moreover, the MCC surface accompanied by good surface area behaves negatively charged at pH 9 and thereby strongly adsorbs the opposite charged MB molecules. The EDX of MCC (Fig. 2b) also confirms the abundant availability of the Al flocs in the reactor.

3.8. Kinetic study

The MB decolorization kinetics was studied by first-order and second-order models. Fig. 10 shows the linear plots of kinetic models and kinetic parameter values are given in Table 4. The MB decolorization followed the first-order kinetics with the highest value of K_1 = 0.194 1/min and R^2 = 0.993.

4. Conclusions

- The addition of MCC to the EC process caused a prompt enhancement in the MB dye removal at lower CD and contact time.
- The alkaline pH 9 was found highly suitable for MB removal in EC/MCC adsorption combined process and a positive effect of current density while a negative effect of dye concentration was observed on MB removal.
- The optimum MB removal of 98.75% was achieved at pH = 9, CD = 22.15 mA/cm^2, C_0 = 50 mg/L, time = 20 mins, with MCC dose = 5 g/L.
- The study revealed the effective performance of MCC in the EC combined process for the abatement of dye effluents.

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