Treatment of rice grain-based distillery effluent using hybrid electrocoagulation–microfiltration processes: performance and optimization

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ABSTRACT

The distillery industry has tremendous contribution to the Indian economy. Nowadays poor grade rice grain is used as a raw material for alcohol production instead of sugarcane molasses due to its availability throughout the year. However, wastewater generated from the distillery industry creates a serious effect on humans, plants, and animals. The biodigester effluent (BDE) generated from the rice grain-based distillery industry has a high organic load in terms of the chemical oxygen demand (COD = 7,020 mg/dm³) and color appearance (color = 485 PCU). Present work is an attempt to treat BDE by using electrocoagulation (EC) and electrocoagulation followed by microfiltration (EC-MF) processes. The maximum COD removal of 98.6% and color removal of 99.9% were achieved by the EC-MF process at their optimum condition. On the other hand, maximum COD removal of 87% and 80% were achieved with individual EC and MF processes respectively. The zero discharge is the need of the distillery industry therefore the EC-MF process is well applicable to treat BDE.

Keywords: Electrocoagulation; Economic analysis; Microfiltration; Biodigester effluent; Organic load

1. Introduction

Nowadays industrial development is increasing day by day due to the production of different products as per customer demand. During industrial production huge quantity of freshwater is required for different purposes such as fabricating, processing, washing, diluting, cooling, and transporting, and generates a large amount of industrial wastewater. If this wastewater is met to any freshwater stream, then the water source is fully contaminated. Hence most of industries treat wastewater properly and maybe reused or released it to a sanitary sewer or surface water in the environment. The alcohol industries are an important segment of the Indian economy because it is an important raw material for the manufacture of several products and also it is good blending material for gasoline. Nowadays alcohol-based hand sanitizers are playing a major role to avoid the spread of Covid-19. In India, around 60% of alcohol is still produced by sugarcane molasses that enhances the load of the former [1]. On the other hand, India is a major rice producing country therefore, a poor grade rice grain may be a good substitute in place of sugarcane molasses for the production of ethanol [2]. Furthermore, the availability of rice grain throughout the year is an additional advantage for the rice grain-based distillery industry.

For the production of a single liter of ethanol, 16-L water is required [3]. After the use of water, it is discharged as wastewater which is commonly known as a spent wash (SW).

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SW contains very high chemical oxygen demand (COD) (35–45 kg/m³) and biochemical oxygen demand (BOD) (7–10 kg/m³) value [4]. Hence SW is first treated in biodigester where around 60% of organic load reduces. In other words, biodigester effluent (BDE) still contains significant COD (14–18 kg/m³), and BOD (2.8–4 kg/m³) [5]. If BDE is directly discharged into any water receiving body it can damage the aquatic system. Therefore, without any prior treatment, it cannot be discharged into any water receiving body.

Several techniques such as wet oxidation [6], bio adsorption [7], thermolysis [8,9], coagulation [10], adsorption [11], and electrochemical degradation [12] are used for the treatment of BDE. To the best of our knowledge, these individual processes do not provide complete removal of organics from BDE. Because zero-liquid discharge is mandated, treatment schemes have evolved to adapt to this requirement. Recently a number of works published on the treatment of different industrial wastewater using electrocoagulation (EC) integrated with other techniques. Prajapati et al. [13] used electrocoagulation followed by coagulation for the treatment of BDE and achieved 98% COD removal. Sen et al. [14] examined the effect of EC combined with the adsorption process for the treatment of textile effluent and found 97% COD reduction. Feng et al. [15] observed 87.3% COD reduction from electroplating effluent using electrocoagulation combined with biological treatment. Apart from this EC combined with the microfiltration (MF) process provides almost organic removal from the BDE. In addition, EC integrated with the MF process is well effective in order to improve MF retention and mitigate membrane fouling. Basu et al. [16] used a combined EC-MF process to remove COD, turbidity, and color from the molasses distillery wastewater. Maximum COD removal of 90%, color removal of 88%, and turbidity removal of 99% were achieved at their optimum condition. A similar type of work was reported by Lebron et al. [17] for the treatment of vinasse using integrated coagulation, MF, and nanofiltration process, and the removal efficiencies for COD, color, and ions was achieved higher than 90.6%.

To understand the concept of the EC-MF process, the mechanism of both techniques EC and MF are explained in detail. EC is based on the supply of direct current on a metal electrode (generally iron or aluminium) that is submerged in the solution. BDE contains different types of organics and each organic has individual functional groups. These functional groups pass net negative charges. On the other hand, positive metallic ions are released from the anode that combines with the negative ions. Therefore, small particles are converted into heavy flocks in form of metal hydroxide and settle down at the bottom of the reactor through gravity [18] [Eq. (1)].

$$xM^{3+} + yH_2O \rightarrow M_x(OH)_y^{(3x-y)+} + yH^+$$
 (1)

Apart from this, during the MF contaminated solution is passed through a special pore-sized (PS) membrane (PS = $0.1-10 \mu$ m) to separate microorganisms and suspended particles from the process liquid with defined pressure [19]. If highly contaminated water is directly passed through MF based membrane module, it can reduce the fouling time of

the system consequently cost of the process rises [20]. As during the EC treatment most of the pollutants are already removed, it is a good feed for the MF process.

Around 100% removals of COD and color from the BDE have been the novelty of this work. To achieve this goal several experiments have been conducted using the EC-MF process for the removal of organics from BDE. In this context, the aim of this work has been to evaluate the feasibility of the EC-MF process for the BDE treatment. Both aluminium and iron electrode used with different operation condition (pH and current density (CD)) has been assessed aiming at MF rejection improvement, membrane fouling mitigation, and propose a sustainable BDE treatment that allows its reuse. Additionally, the technical feasibility of BDE treatment has been also analyzed.

2.1. Effluent

The BDE used in this work was collected from the Chhattisgarh Distillery Pvt. Ltd., Kumhari, Chhattisgarh. To avoid any changes in the sample it was preserved at 4°C. Chemicals (i.e., H_2SO_4 and NaOH having a purity of 99%) used were of analytical grade (AR) and purchased from the Merck Company, Mumbai (India). Iron and aluminium were purchased from the local market. Double distilled water was used throughout the experiments (made in a laboratory). The experimental setup of the Tangential flow filtration (TFF) system was imported from the Bionet Company, Murcia (Span) (Model: M1).

2.2. Experimental

The lab-scale batch experimental setup (reactor) used for the electrocoagulation studies as presented in Fig. 1, made by perspex glass had a capacity of 1.2 dm³ (105 mm × 98 mm × 120 mm).1 dm³sample was taken in the reactor. Four rectangular electrodes dimension of 70 mm × 70 mm (i.e., effective surface area of 4,900 mm²) were fitted in a glass reactor. The detailed design of the EC setup is presented in Table 1.

A fixed gap of 2 cm was taken between two electrodes. A sufficient gap of 2 cm was maintained between the bottom of the reactor and electrode for the proper stirring (i.e., using magnetic stirrer). DC (direct current) suppler current range of 0–5 Å and voltage range of 0–30 V were used for the supply of direct current. Treated samples were collected in fixed time intervals for COD and color measurement.

Tangential flow filtration (TFF) experiments were performed to evaluate the water recovery efficiency of the microfiltration (MF) process as presented in Fig. 1. To perform the TFF process, pre-electrocoagulated effluent was placed in a 1 L feed tank. Initially, the permeate outlet was closed and feed was recirculated through the membrane module (type of membrane hollow fiber) with the help of a pump (Flow feed pump inbuilt with experiment setup). The membrane had different average pore diameters of 0.1, 0.2, and 0.45 μ m, and a filtration area of 0.04 m². Setup was already equipped with measuring instruments to measure the different parameters like pressure, flow rate, etc. MF was performed at concentrated mode filtration, the feed flow rate was 1.1 L/min, the feed pressure was 20 kPa and up to a



Fig. 1. Schematic diagram of the EC-MF bench scale units

Table 1
Characteristics of electrode and reactor

Electrodes		
Material (anode and cathode)	Iron	Aluminium
Shape	Rectangular	Rectangular
	plate	plate
Size of each plate (mm)	70×70	70×70
Thickness (mm)	2	2
Effective area (mm ²)	4,900	4,900
Plate arrangement	Parallel	Parallel
Submergence	Full	Full
Reactor		
Make	Perspex glass	
Dimensions (L × W × H) (mm)	(105 × 98 × 120))
Volume (dm ³)	1.2	
Electrode gap (mm)	20	
Туре	Up flow	
Mode of operation	Batch	

recovery rate of 60%. The permeate side pressure was essentially atmospheric pressure. After 5 min of operation, once the steady state had been reached, the permeate outlet was opened, and permeate was collected in the permeate tank. Permeate flux was calculated based on the rate of permeate collection in permeate tank. Further, permeate was analyzed for COD, and color measurement.

2.3. Analytical method

The standard dilution method is used for the measurement of the BOD (5-d) of given the sample at 20°C [21]. For this, an airtight BOD bottle was filled with 300 mL of sample and put in an incubator at 20°C for 5 d. Further, the value of dissolved oxygen was measured in the original sample and after 5 d of incubation. The BOD value of the sample is a difference between the initial and final value of dissolved oxygen. A gravimetric test is used for the evaluation of the total suspended solids. In this technique a water sample is passed through Whatman filter paper (number 42) and received residue on filter paper was dried to a constant weight at $105^{\circ}C \pm 1^{\circ}$. The final weight of residues is the value of the suspended solid [21]. Total solid is the summation of suspended solids and dissolved solids (i.e., total solid is measured by evaporating a sample to dryness and weighing the sample) [21].

A dichromatic closed reflux method was used for the determination of the COD of a given sample using solution A (contains mercury (II) sulphate) and solution B (contains sulphuric acid, potassium dichromate) as per Standard Methods [21]. In this method, sample is diluted as per requirement and 3 mL was put in a cylindrical tube. Further reagent A (0.3 mL) and reagent B (2.3 mL) are taken in the tube. These solutions are shifted in an incubator (Loveland, Hach Company) at 150°C for 2 h. Finally, samples are permitted to cool to room temperature and COD values are determined by measuring the absorbance of the digested assay solution at λ = 620 nm in a UV-Visible spectrophotometer (Thermo, England make). The value of the color of the given sample was measured by the absorbance capacity at the wavelength (λ_{max} = 475 nm) of the solution using a UV spectrophotometer [22]. The percentage of COD and color removal from permeate was calculated by the following equation [23];

$$\% \operatorname{Removal} = \frac{\left(C_o - C_t\right) \times 100}{C_o}$$
(2)

where C_o is the initial concentration of effluent (mg/dm³) and C_t is the concentration of treated effluent (mg/dm³). The characteristics of the original effluent are given in Table. 2.

Table 2 Characteristics of rice grain-based biodigester effluent

Parameter	Range
рН	7.4
COD (mg/dm ³)	7,020
BOD (mg/dm ³)	1,130
BOD/COD	0.161
TDS (mg/dm ³)	12,272
TSS (mg/dm ³)	6,534
Color (PCU)	485

3. Results and discussion

3.1. Electrocoagulation treatment of BDE

3.1.1. Effect of pH

The initial pH of the solution is a dominant parameter of an electrocoagulation treatment as reported by several authors [24–26]. The oxidation of Fe²⁺ ions to Fe³⁺ ions depend on the pH of the solution as well as the dissolved oxygen concentration [27]. It is reported that [28] in a solution of acidic media (pH > 7) the oxidation of Fe²⁺ cations goes slowly in contact with dissolved oxygen [Eq. (3)]. Apart from this when the solution has neutral (pH = 7) or alkaline media (pH > 7), Fe²⁺ is immediately converted into ferrous hydroxide [Eq. (4)] that is readily oxidized by dissolved oxygen to iron (III) hydroxide [Eq. (5)]. Overall adsorption mechanism is more dominant in the present study [29].

$$Fe^{2+} + O_2 + 2H_2O \rightarrow Fe^{3+} + 4OH^-$$
 (3)

$$Fe^{2+} + 2OH^{-} \rightarrow 2Fe(OH)_{2}$$

$$\tag{4}$$

$$4Fe(OH)_{2} + O_{2} + 2H_{2}O \rightarrow 4Fe(OH)_{3}$$
(5)

Therefore, several experiments were performed to investigate the effect of pH on COD and color removal at varying pH range (pH 2-10) with iron electrode while other operating parameter were remained constant (i.e., $CD = 102 \text{ A/m}^2$ and EG = 2 cm). It could be observed from Fig. 2a that the percentage COD reduction were increased 53% to 83% with increase in pH from pH 2 to pH 8. However further increased in pH, the percentage COD reductions were decreased. Also, percentage color reductions were increased 35% to 50% from pH 2 to pH 8 and above pH 8 color reductions were decreased (Fig. 2b). Overall maximum COD and color reduction were achieved at pH 8. This may be due to the fact that maximum organic removal achieves when a balance occurs among availability of H⁺ ions, amount of Fe³⁺ released by an iron anode, and negative ions possessed by the different functional groups present in the organics [30,31].

The effect of pH on COD, and color removal efficiencies were studied with aluminium as a sacrificial electrode. The experiments were performed at varying pH (pH



Fig. 2. Effect of initial pH on (a) percentage COD reduction and (b) color removal (COD concentration = $7,020 \text{ mg/dm}^3$, color = 485 PCU, CD = 102 A/m^2 , EG = 2 cm, anode iron).

2–10) at constant CD (102 A/m²) and EG (2 cm). The results obtained during the EC treatment are presented in Fig. 3a and b. It was found that percentage COD and color reduction increased from 48% to 78% (Fig. 3a) and from 33% to 52% (Fig. 3b), respectively at pH ranges from 2 to 8. In other words, the different results of COD and color reductions were obtained at varying pH. This may be due to the quality and quantity of hydroxide ions generated at particular pH. BDE contains number of components such as proteins, carbohydrates, and melonidin. These have various functional groups which pass a net negative charge that reacts with aluminium and its positively charged hydroxides, for example, $AlOH^{2+}$, etc. The quantity of electrode loss, formation of Al^{3+} , $Al(OH)_{2'}$, $Al(OH)_{3'}$, and monomeric and polymeric hydroxide species varied with pH [12,32].

The economic consideration was also an important parameter to optimize the treatment process. The electrical energy consumption was estimated by following equation [33,34]:

Power consumption =
$$\frac{Uit}{v}$$
 (6)

where *U* is the cell voltage (volt), *i* is the current (Å), *t* is the reaction time (h) and *v* is the volume of BDE in the reactor (m³). Power consumption with respect to different pH is presented in Fig. 4 and it is found that power consumption



Fig. 3. Effect of pH on (a) percentage COD reduction and (b) color reduction (COD concentration = $7,020 \text{ mg/dm}^3$, color = 485 PCU, CD = 102 A/m^2 , EG = 2 cm, anode, aluminium).

increases with increase in pH. Power consumption of 15.1, 16.9, 17.5, 20.9, and 21.6 kWh/m³ found at pH of 3.5, 5, 6.5, 8, and 9.5 respectively at CD of 102 A/m², EG of 2 cm, and *t* of 100 min.

3.1.2. Effect of CD

CD is a key parameter during the electrocoagulation process [35,36]. Hence the impact of varying CD on the COD, and color removal for a two-, four-, and eight number of plate configuration in parallel arrangement were observed. The maximum COD removal of 65%, 87% and 54% were obtained with two-, four-, and eight plate configurations respectively as shown in Fig. 5. This may be due to the overdosing of metal ions can destabilize the colloids. Hence the effect of CD on COD and color removal during EC treatment of BDE was investigated with varying CD of 51–204 A/m² (four plate configuration), pH of 8, EG of 2 cm, and anode material of iron.

The variation of the current density range has oriented the characteristics and the amount of pollutants to be removed from given wastewater, for instance from 25–300 A/m² [37–39]. The selection of the optimum current density is evaluated by considering other operating parameters such as pH of solution etc. For long time utilization of electrocoagulation reactor current density should be near about 102 A/m² [40].



Fig. 4. Effect of initial pH on energy consumption (COD concentration = $7,020 \text{ mg/dm}^3$, color = 485 PCU, CD = 102 A/m^2 , EG = 2 cm, anode iron, reaction time = 100 min).



Fig. 5. Effect of number of electrode on (a) percentage COD reduction and (b) color reduction (COD concentration = $7,020 \text{ mg/dm}^3$, color = 485 PCU, pH = 8, EG = 2 cm, anode iron).

The results obtained during EC treatments are presented in Fig. 6a and b. As indicated by Fig. 6a, the COD reductions were increased from 68% to 87% with an increase in CD from 51 to 153 A/m². Further increase in CD, the percentage COD reductions would decrease. Similarly, percentage color reductions were increased from 41% to 62% with an increased in CD from 51 to 153 A/m² (Fig. 6b). Further, increased CD percentage color reductions were also



Fig. 6. Effect of CD on (a) percentage COD reduction and (b) color reduction (COD concentration = $7,020 \text{ mg/dm}^3$, color = 485 PCU, pH 8, EG = 2 cm, anode iron).

decreased. It is also demonstrated from Fig. 6a and b that most of COD and color reductions of 84% and 50%, respectively were achieved at CD of 102 A/m². When CD rose up to 153 A/m² marginal COD, and color reduction of 87% and 62% were observed. In other words, higher values of CD are not the assurance of larger COD and color reduction efficiencies. Because organics removal depends on additions of metal ions (Fe³⁺) releases by anodic material and negative ions generates by the functional groups. These functional groups are naturally in built with organics and the individual functional groups has involved in a particular organic component [12,37]. An excess amount of metal ions can destabilize the particles of BDE [31,38].

The effect on COD and color removal by varying number of electrodes for two-, four-, and eight plate configurations in parallel arrangement were also observed for aluminium electrode. The maximum COD removal of 49%, 80% and 44% with two-, four-, and eight plate configurations respectively as presented in Fig. 7. This may be due to the overdosing on metal ions can destabilize the colloids. Hence the effect of CD on COD and color removal during electrocoagulation process is investigated at different CD ranges of 51-204 A/m² (four plate configuration) pH of 8, EG of 2 cm, and metal anode of aluminium. It could be seen from Fig. 8a and b that percentage COD and color reduction increased from 62% to 80% and from 38% to 57%, respectively with a treatment time of 90 min. at CD of 51-204 A/m². Aluminium based electrode provided lesser COD and color removal efficiencies than iron electrode at same CD range of 51–204 A/m².



Fig. 7. Effect of CD on (a) percentage COD reduction and (b) color reduction (COD concentration = $7,020 \text{ mg/dm}^3$, color = 485 PCU, pH = 8, EG = 2 cm, anode aluminium).

This may be due to COD, and color removal from the BDE depends on the type of pollutants present in the solution. BDE contains several types of organics such as reduced carbohydrate, proteins, etc. and these organics have own functional groups for example protein have amino groups that release net negative charges [31,39]. Fe³⁺ combined more effectively with these negative ions as compared to Al³⁺ and increases the effective size of particles consequently iron based electrode offered better results than aluminium anode [1,41].

3.2. Electrocoagulation followed by microfiltration treatment of BDE

3.2.1. Effect of BDE concentration on permeate flux

The concentration of the solution plays an important role during the MF process [42,43]. If the solution has a higher concentration of pollutants, then it affects the efficiency of the membrane module. Because the module applies more effort to treat the solution and also small particles may adhere to the pores consequently causing the fouling of the membrane. Therefore, solutions that have lower concentration enhance the life of membrane module. The effect of BDE concentrations on permeate collection efficiency was observed during MF process using membrane of different pore size (0.1, 0.2, and 0.45 μ m). Applied pressure of 20 kPa, effluent flow rate of 1.1 L and filtration area of



Fig. 8. Effect of CD on (a) percentage COD reduction and (b) color reduction (COD concentration = $7,020 \text{ mg/dm}^3$, color = 485 PCU, CD = 102 A/m^2 , EG = 2 cm, anode aluminium).

0.04 m² were remain constant for all MF experiments. The BDE of different concentration included untreated effluent (COD of 7,020 mg/dm3), iron anode-based EC treated effluent (COD of 1,123.2 mg/dm3), and aluminium based EC treated effluent (COD of 1,544.4 mg/dm³) was applied through MF experimental setup. Results obtained during MF treatment are presented in Fig. 9. It could be seen from Fig. 9 that permeates flux were increased with decreased in effluent concentration. The maximum permeate flux was achieved 29 × 10⁵, 71 × 10⁵ and 56 × 10^5 m³/m²s for untreated effluent, iron electrode-based EC treated effluent and aluminium electrode-based EC treated effluent respectively with same PS (0.45 µm). It confirmed that the MF treatment provided better result with lower concentration feed. This may be due to presence of high quantity of particle in BDE can block the path of water supply in MF module consequently for better results feed concentration should be poor. In addition, permeates flux were found poor decreased in PS of MF module. The permeate flux were decreased 22 × 10⁵–29 × 10⁵ m³/m²s from PS of 0.45–0.1 μ m for untreated effluent, 66×10^5 – 71×10^5 m³/m²s from PS of 0.45-0.1 µm for iron electrode-based EC treated effluent, $48 \times 10^{5} - 56 \times 10^{5} \text{ m}^{3}/\text{m}^{2}\text{s}$ from PS of 0.45–0.1 µm for aluminium electrode-based EC treated effluent. This may be due to easy passes of feed in larger PS membrane module [44,45].



Fig. 9. Effect of pore size (PS) on pure water flux using untreated effluent and EC treated effluent for both anode material iron and aluminium as a feed (Feed flow rate = 1.1 L/min, feed pressure = 20 kPa).

3.2.2. Effect of PS on untreated effluent

The selection of the best PS of membrane for any particular wastewater depends on the concentration and size of particles. Because if the PS has a higher value, then harmful particles may also pass from pores, and the efficiency of the process influences while PS have smaller then the chances of fouling may be enhanced. Therefore, the value of PS should be optimum. To investigate the effect of different PS on COD and color removal efficiencies of untreated BDE, several MF experiments were performed at varying PS of 0.1–0.45 µm, filtration area of 0.04 m², applied pressure of 20 kPa, and effluent flow rate of 1.1 L. The concentration of untreated BDE in term of COD was 7,020 mg/dm³. The results obtained are presented in Fig. 10a and b. The percentage COD reduction decreased from 80% to 64.9% with increased in PS from 0.1-0.45 µm (Fig. 10a). In other words, maximum COD reduction of 80% was achieved for PS of 0.1 µm with process time of 30 min and further increased in PS percentage COD reduction decreased. This may be due to membrane module with lower PS does not pass smaller particles even the applied pressure is high and pure permeate obtains as a product. However, lower PS of membrane module receives fouling in a short period of treatment and their cleaning can enhance the cost of the process. Furthermore, the percentage color reduction also was decreased from 85% to 71% with increase in PS from 0.1-0.45 µm (Fig. 10b). The color reduction followed the pattern of COD reduction. This may be due to insoluble organics of BDE provided major contribution in color appearance and these organics are separated through the MF process [46].

3.2.3. EC treated effluent followed by MF treatment

It is well known fact that membrane separation process provided better results with solution of lower concentration. Therefore, BDE is first treated by electrocoagulation process where sufficient amount of organic load has been reduced. To achieve the almost removal of organics from

(a) 100

90

80

70

60

50

40

20

10

0

0

5

10

15

Time (min)

20

25

COD reduction

\$ 30

(b)

100

90

80

70



% color reduction 60 50 40 30 20 10 0 0 5 10 15 20 25 Time (min)

Fig. 10. Effect of PS on (a) percentage COD reduction and (b) color reduction (feed = untreated effluent, COD concentration = 7,020 mg/dm³, color = 485 PCU, feed flow rate = 1.1 L/min, feed pressure = 20 kPa).

electro-coagulated BDE, it is further treated by membrane separation process. The effect of PS during MF treatment on COD and color removal of iron based pre-electrocoagulated effluent (COD of 1,123.2 mg/dm3) was carried out at range of 0.1-0.45 µm and results are presented in Fig. 11a and b. Maximum COD reduction of 99.9%, 99.6%, and 98.6% were achieved with PS of 0.1, 0.2, and 0.45 μ m respectively. On the other hand, color reductions of 99.9% for PS of 0.1 µm, 99.6% for PS of 0.2 µm, and 99.1% for PS of 0.45 µm were achieved. Therefore, optimization point of view PS of 0.45 µm was found most effective to treat BDE. Because permeate collection and fouling rate of larger PS membrane is much better than smaller PS membrane [47]. Furthermore, this (larger PS) type of membrane is most cost effective.

Aluminium electrode based pre-electrocoagulated effluent (COD of 1,544.4 mg/dm³) was also further treated by MF treatment. Experiments were performed with PS range of 0.1-0.45 µm, filtration area of 0.04 m², applied pressure of 20 kPa, and effluent flow rate of 1.1 L. The results obtained after MF experiment are presented in Fig. 12a and b. Maximum COD and color reduction were increased 98.1%-99.6% and 98.5%-99.9% with PS of 0.45-0.1 µm, respectively. The COD and color reductions of aluminium electrode based pre-electrocoagulated effluent were less than iron electrode based pre-electrocoagulated effluent. This may be due to higher organic concentration of

Fig. 11. Effect of PS on (a) percentage COD reduction and (b) color reduction (feed = iron-based EC treated effluent, COD concentration = 1,123.2 mg/dm³, color = 242.5 PCU, feed flow rate = 1.1 L/min, feed pressure = 20 kPa).

 $PS = 0.1 \, \mu m$

-PS = 0.2 μm

PS = 0.45 µm

30

 $PS = 0.1 \, \mu m$

 $PS = 0.2 \mu m$

 $PS = 0.3 \, \mu m$

30

35

35

aluminium-based EC treated effluent consequently it takes more time to pass from the membrane module and also enhances the fouling factor of membrane [48].

The effect of pressure on also investigated for pressure range of 15 to 25 kPa for both aluminium and iron based pre-electrocoagulated effluent. Experiments were performed with PS of 0.45 µm, filtration area of 0.04 m², and effluent flow rate of 1.1 L. Maximum COD reduction of 99.2%, 98.6% and 97.8% and color reduction of 99.7%, 99.1% and 98.3% were achieved at 15, 20 and 25 kPa respectively for iron based pre-electrocoagulated effluent (Fig. 13a and b). While, maximum COD reduction of 99.1%, 98.9% and 97.9% and color reduction of 99.7%, 99.5% and 97.7% were obtained at 15, 20 and 25 kPa respectively for iron based pre-electrocoagulated effluent (Fig. 14a and b). In other words, not any significant changes obtained with pressure variation and pollutant almost removed however pressure of 20 kPa is most acceptable due to better result and it is cost efficient also.

3.3. Optimization

In the above discussion, it is clear that individual EC treatment did not provide the required results. Also, individual MF treatment was not given the expected output. Therefore, EC followed by MF treatment is selected for



Fig. 12. Effect of PS on (a) percentage COD reduction and (b) color reduction (feed = aluminium based EC treated effluent, COD concentration = $1,544.4 \text{ mg/dm}^3$, color = 232.8 PCU, feed flow rate = 1.1 L/min, feed pressure = 20 kPa).

better results. In the case of EC treatment iron electrode provided better results (COD was reduced from 7,080 to 1,123.2 mg/dm³) than aluminum electrode (COD was reduced from 7,080 to 1,544.4 mg/dm³). Additionally, the cost of iron electrode is less than aluminium electrode and also iron is comparatively less harmful than aluminium [2,49]. Therefore, iron-based EC treatment should be selected for further treatment through MF. On the other hand, in case of MF treatment, the pore size of membrane enhances the cost of process. In other words, higher the PS of membrane reduces its cost. It is clear from Fig. 8a and b that the percentage COD and color removals did not vary significantly with PS. Therefore, membrane module with PS of 0.45 µm should be selected for effluent obtained from EC treatment. In other words, iron-based EC treated effluent should be further treated by membrane module with PS of 0.45 µm. The final results by EC-MF process in term of COD reduction of 98.1% (COD reduced from 1,123.2 to 21.34 mg/dm³) and color reduction of 99.1% (color reduced from 242.5 to 2.18 PCU) were achieved. Therefore, EC-MF process has achieved desired concentration as suggested by different regulatory agencies.

If untreated effluent is directly treated through membrane separation technique it can enhance the cost of process because the availability of the least amount of permeate and also fouling of membrane due to the high organic load. However, recycling of permeate through membrane module may provide a better result than other



Fig. 13. Effect of pressure on (a) percentage COD reduction and (b) color reduction (feed = iron-based EC treated effluent, COD concentration = $1,123.2 \text{ mg/dm}^3$, color = 242.5 PCU, feed flow rate = 1.1 L/min, PS = 0.45μ m).

techniques but treating this way can enhance the treatment time. Apart from this EC-MF process is more acceptable than other techniques.

4. Conclusion

This work is based on the investigation of performance and optimization of EC and EC-MF processes towards COD and color reduction from BDE. The results show that EC-MF (electrode combination of Fe–Fe) gives better COD, and color removal efficiencies than MF and EC alone. The effect of various operating parameters like pH of the solution and CD on EC treatment was studied. The maximum COD and color reduction efficiency of 84% and 50% respectively was achieved with EC treatment (electrode combination of Fe–Fe) at their optimum condition. This effluent was further treated by the MF process. EC-MF process provided excellent results in term of COD reduction of 98.1% and color reduction of 99.1%. Hence, it is recommended that EC-MF process could be successfully applied for the treatment of BDE.

Declaration of competing interest

The author has not any conflict of interest.



Fig. 14. Effect of pressure on (a) percentage COD reduction and (b) color reduction (feed = aluminium-based EC treated effluent, COD concentration = $1,544.4 \text{ mg/dm}^3$, color = 232.8 PCU, feed flow rate = 1.1 L/min, PS = $0.45 \mu \text{m}$).

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