

Research Paper

Can electrocoagulation be an effective post-treatment option for SBR treated landfill leachate and municipal wastewater mixture?

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ABSTRACT

A combined process of sequencing batch reactor (SBR) and electrocoagulation for co-treatment of landfill leachate and municipal wastewater was assessed. SBR was used in the first instance for co-treatment of 20% (v/v) landfill leachate and municipal wastewater mixture. Effluent from SBR was subjected to electrocoagulation for post-treatment, with aluminum as sacrificial anode and stainless steel as cathode. Direct current at a density of 257 A/m² was applied during the electrocoagulation process. In electrocoagulation, spacing between the two electrodes was varied at 5 and 7 cm to assess its influence on treatment efficiency. SBR was effective to remove 65% chemical oxygen demand (COD), 77% total suspended solids (TSS), 89% ammonia, 80% nitrate, 64% phosphate and post-treatment by electrocoagulation resulted in an overall 98% COD, 98% TSS and 99% ammonia, nitrate and phosphate reduction efficiency with 5 cm of electrode spacing. Respectively, final COD, ammonia and TSS was 37, 1 and 98 mg/L after 150 min of electrocoagulation which met the Indian standards for the discharge of treated wastewater. The results highlight that SBR followed by electrocoagulation as post-treatment can be an effective option for the treatment of landfill leachate and municipal wastewater mixture.

Key words | co-treatment, electrocoagulation, leachate, municipal wastewater, SBR

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INTRODUCTION

Landfill leachate is aqueous effluent generated from municipal solid wastes dumps due to the presence of inherent moisture and rainwater percolation through wastes along with physical, chemical and microbiological decomposition of organic matter (Foo & Hameed 2009). Leachate contains dissolved organic and inorganic substances like ammonia, calcium, magnesium, iron, sulphate, chloride and metals such as cadmium, copper, chromium, lead and several xenobiotic compounds, leachate is known to produce adverse

effects on the environment and human health (Wiszniewski *et al.* 2006).

Landfill leachate generation and its poor management has been the major concern in open waste dumps. The option of constructing an on-site landfill leachate treatment facility may not be both economically and practically feasible at all the locations. Moreover, depending on the local climatic conditions, leachate generation may also cease during the dry months of the year which works against the

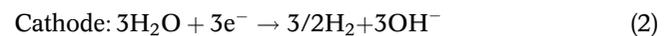
setting up of an on-site treatment facility. Co-treatment of landfill leachate with municipal wastewater in sewage treatment facilities appears to be a good option. Though there have been some studies exploring this option, there remains much scope for further research to develop an efficient process (Wiszniewski *et al.* 2006; Capodici *et al.* 2014; Ranjan *et al.* 2016). Biodegradation of organic pollutants is favored due to dilution of leachate with wastewater and easy adaptation ability of the activated sludge. Most of the co-treatment studies used some pre-treatment prior to biological treatment (Uygur & Kargi 2004; Xiao *et al.* 2013). Site-specific conditions and the complex nature of highly varying landfill leachate makes it necessary to develop combined treatment technology as stand-alone physico-chemical or biological treatment are often not able to achieve the higher levels of treatment required (Kulikowska & Klimiuk 2008).

Stabilized or old landfill leachate typically has a low biochemical oxygen demand/chemical oxygen demand (BOD/COD) ratio, thus co-treatment of landfill leachate and municipal wastewater helps to enhance this ratio and makes the wastes amenable for microbial treatment. Among microbial treatment systems, Sequencing Batch Reactor (SBR) has several advantages such as smaller area requirement, low surplus biomass production, highly flexible operation even with fluctuating quantities and organic loading rate and no need of any external support medium for growth (Neczaj *et al.* 2005). SBR is ideally suited to carry out both organic matter and nutrients (N and P) removal in a single reactor by applying sequential aerobic and anoxic phases.

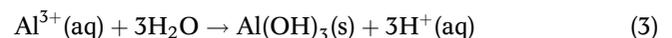
Çeçen & Çakiroğlu (2001) reported that when leachate COD is 50% of total initial COD, a significant decrease in reaction rates can occur due to the inhibitory effects of leachate. Another study on co-treatment with activated sludge in batch mode, a semi-continuous fed batch and a continuous flow fed batch, found that negative effects of leachate in municipal wastewater treatment plant could be avoided by PAC addition. Ferraz *et al.* (2014) observed that co-treatment in submerged aerobic biofilter performance was better at 2% volumetric ratio rather than 5%. These studies show that at higher leachate concentration hard biodegradable organic matter decreased mainly due to dilution rather than biodegradation. Neczaj *et al.* (2008) reported that the

best effluent quality was achieved for co-treatment of landfill leachate with dairy wastewater in SBR at organic loading of 0.8 kg BOD₅/m³.d and 10 d hydraulic retention time (HRT). These authors pointed out that the removal efficiency of SBR decreased with increased organic loading rate or decreased HRT.

Electrocoagulation has gained attention as a cost-effective physico-chemical treatment process which leads to higher treatment efficiency. Electrocoagulation is a combination of conventional coagulation, flocculation and electrochemistry in wastewater treatment (Sahu *et al.* 2014). Sacrificial anode (commonly aluminum or iron) undergoes electrolytic oxidation during the process forming metal-based coagulants which can remove a wide variety of contaminants and the cathode splits the water molecule to produce hydrogen (Equations (1) and (2)) (Elazzouzi *et al.* 2017). Al³⁺ and OH⁻ forms different Al³⁺ monomeric and polymeric species which are finally converted to Al(OH)₃, which contributes to sweep floc formation (Equations (3) and (4)) (Elazzouzi *et al.* 2017; Changmai *et al.* 2019).



Overall reactions occurring:



Electrocoagulation has been applied both in batch mode and continuous mode for removal of organic matter, suspended solids and color from a variety of wastewaters such as olive processing wastewater (Benekos *et al.* 2019), textile wastewater (Naje *et al.* 2016), printing ink wastewater (Papadopoulos *et al.* 2019), licorice processing wastewater (Abbasi *et al.* 2020), and municipal wastewater (Nawarkar & Salkar 2019). A combination of electrocoagulation with other treatments such as advanced oxidation processes and membrane filtration have been recently studied and found to be promising for water and wastewater treatment (Flores *et al.* 2018; Azerrad *et al.* 2019; Changmai *et al.* 2019). The literature shows that electrocoagulation has

been widely studied at laboratory scale for water and wastewater treatment, whereas studies at pilot scale are rather less. Recently, electrocoagulation research has also focused on electrode materials and different reactor configurations to optimize the process for efficient treatment of different wastewater (Naje *et al.* 2016; Flores *et al.* 2018; Nawarkar & Salkar 2019).

There are very limited studies on SBR followed by electrocoagulation for wastewater treatment. Biological processes followed by a physico-chemical treatment of wastewater offers several advantages such as improved efficiency, lesser sludge formation and low quantities of byproduct generation. The scientific literature shows that a combination of SBR + electrocoagulation has not been reported for co-treatment of landfill leachate and municipal wastewater. The major aim of the present study was to test the feasibility of combined SBR and electrocoagulation for co-treatment of landfill leachate and municipal wastewater for COD, total suspended solids (TSS), ammonia, nitrate and phosphate removal. Additionally, in electrocoagulation the influence of two different electrode spacings (5 and 7 cm) as one of the major process affecting parameters was also studied.

MATERIALS AND METHODS

Landfill leachate and wastewater sampling

Landfill leachate was collected in clean HDPE (high density polyethylene) containers from an open municipal solid waste dumpsite located at Jhiri village, 20 km away from Ranchi, Jharkhand, India. The dumpsite is not an engineered facility and wastes are disposed of without taking any scientific precautions involved in a designed engineered landfill. Landfill leachate is old or stabilized in nature as the dumping site is 15 years old (Foo & Hameed 2009; Ranjan *et al.* 2016). Jhiri dumping site is spread over 42 acres of land where approximately 700,000 kg of mixed wastes are dumped daily. Municipal wastewater was collected every day from the wastewater treatment plant at Birla Institute of Technology, Mesra, Ranchi. Both the wastewaters were stored in the laboratory under cold storage until further use.

SBR startup and operation

A laboratory scale SBR was made using a glass jar with a working volume of 3 L, with dimensions of 16 cm (dia) and 25 cm (total height). A magnetic stirrer was used for mixing the reactor contents and an aquarium pump was used for aeration. For SBR startup, activated sludge collected from an extended aeration-based sewage treatment plant was added to the reactor as inoculum. During the startup period, the reactor was fed only with municipal wastewater. Nutrients were added to the system to maintain COD/N/P ratio at 100:6:2, to facilitate rapid build-up of activated biomass.

Once the biomass concentration remained stable for several cycles, the co-treatment process was started by feeding 2% (v/v) of landfill leachate with municipal wastewater. Landfill leachate concentration in the influent was slowly increased to 20% (v/v) of the daily volumetric exchange ratio. For the anoxic phase reaction, sodium acetate was added to SBR to improve the denitrification efficiency. BOD/COD ratio was always maintained at 0.5. For co-treatment experiments, SBR was operated at room temperature at 6 d HRT and 30 d Solids Retention Time (SRT). One SBR cycle of 24 h was divided into: 5 min of fill phase, 16 h of oxic phase (air supply + mixing), 7 h of anoxic phase (mixing only), 50 min of sedimentation phase and 5 min of decant phase. Samples for various parameters analysis were collected from SBR at the start of every day's cycle, end of the oxic phase and finally, at the end of the anoxic phase. SBR was run for several cycles to attain steady state conditions and only when effluent quality was stable for at least three cycles was data collected from the next three cycles. Steady state was confirmed to have been reached once there were no large differences in effluent COD, ammonia, nitrate and phosphate (Ranjan *et al.* 2016).

Electrocoagulation treatment

A bench scale electrocoagulation set-up was made with borosilicate glass of 1 L capacity with 0.7 L as the working volume. A schematic of SBR + electrocoagulation for co-treatment of landfill leachate and municipal wastewater mixture is shown in Figure 1. SBR effluent for post-treatment purposes was added into the electrocoagulation reactor.

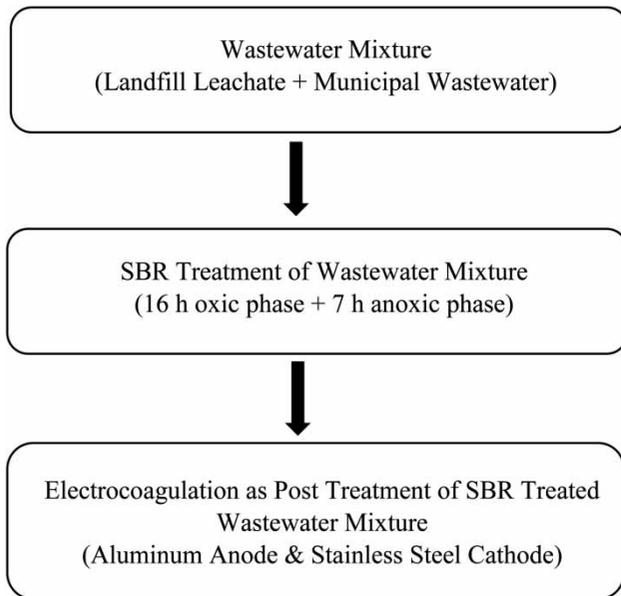


Figure 1 | Flow chart of landfill leachate and municipal wastewater mixture treatment in SBR + electrocoagulation.

Two different electrodes were used as anode and cathode. The cathode was made up of stainless steel and aluminum and was used as the sacrificial anode which undergoes electrolytic oxidation to form metal oxides which act as coagulant. The dimensions of both the electrodes were $7 \times 6.5 \times 0.4$ cm. Electrodes were connected to a direct current (DC) power supply where the voltage was maintained at 12 V generating current density of 257 A/m^2 . Two different electrodes spacings were tested to determine their effect on electrocoagulation. The inter electrode gap was varied at 7 cm (mode 1) and 5 cm (mode 2) during the treatment. Electrocoagulation reactor contents were mixed with a magnetic stirrer during the experiment. Experiments were performed for 150 min and samples were taken from the reactor at the start and then at 30 min intervals for different analysis. All the experimental runs were carried out in triplicate and the data presented are the average of these runs.

Analysis

Characterization of landfill leachate and municipal wastewater was carried out following the analytical procedures detailed in the standard methods (APHA 1992). pH and electrical conductivity (EC) were monitored using a HORIBA

multi-parameter meter on a regular basis to check the stability of the SBR. Co-treatment efficiency of the SBR and electrocoagulation was assessed by analyzing the samples for removal of COD, ammonia, nitrate, phosphate and TSS as per the standard methods (APHA 1992). For this purpose, samples were allowed to settle and later filtered through $0.45 \mu\text{m}$ filter paper for further analysis. MS Excel and SigmaPlot (Ver. 13) were used for processing the data to prepare figures and tables. Effluent characteristics were subjected to univariate Analysis of Variance (ANOVA) to determine the statistical significance among inter-electrode spacing and process efficiency.

RESULTS AND DISCUSSION

Characteristics of municipal wastewater and landfill leachate

Municipal wastewater and landfill leachate were characterized to determine the strength of wastewaters (Table 1). Low BOD:COD ratio and slightly high pH indicated that the leachate was in a stabilized state (Ranjan et al. 2016). It is known that the BOD/COD ratio of leachate from a young landfill is 0.5–1.0, medium-age landfill is 0.1–0.5 and old landfill is <0.1 (Foo & Hameed 2009). Landfill

Table 1 | Mean ($n = 20$, \pm S.D.) physico-chemical characteristics of municipal wastewater and landfill leachate

Parameters	Municipal wastewater	Landfill leachate	Indian Standards for Treated Wastewater Discharge
pH	7 ± 0.3	7.9 ± 0.4	6.5–9.0
EC (mS/cm)	0.7 ± 0.2	9 ± 2	–
TDS (mg/L)	470 ± 40	$6,900 \pm 2,000$	–
TSS (mg/L)	450 ± 35	$6,800 \pm 2,500$	100
BOD (mg/L)	260 ± 80	80 ± 40	30
COD (mg/L)	460 ± 90	$4,600 \pm 1,800$	250
Ammonia (mg/L)	45 ± 2	310 ± 100	50
Nitrate (mg/L)	0	35 ± 7	10
Phosphate (mg/L)	10 ± 2	50 ± 35	5
BOD/COD ratio	0.6	0.02	–

leachate is generally classified into three stages based on its age, young (<5 years), medium (5–10 years) and old (>10 years) (Foo & Hameed 2009). Among the different landfill leachate characteristics, the BOD/COD ratio is regularly used as the most representative of landfill age as these are directly indicative of leachate's biodegradability. High ammonia concentration and low nitrate concentration in landfill leachate revealed that the landfill is in a methanogenic phase. Municipal wastewater contained moderate BOD and COD concentration as the source was from student hostels, offices, and faculty and staff residences at the university and there was no mixing of any type of industrial wastewater. Landfill age plays an important part in its composition, especially on organic compounds and ammonia concentration, as it has been reported that landfill reaches the methanogenic stage in very short periods, such as within four years (Kulikowska & Klimiuk 2008). It has been reported that old landfills are often characterized by low BOD/COD ratio and high ammonia concentration (Kulikowska & Klimiuk 2008; Ranjan *et al.* 2016).

Co-treatment of landfill leachate and municipal wastewater in SBR

Removal efficiencies of COD, ammonia, nitrate, phosphate, and TSS following co-treatment of 20% (v/v) landfill leachate and municipal wastewater in SBR are presented in Table 2. COD removal efficiency was 65% after the co-treatment. Uygur & Kargi (2004) reported 62% COD

removal following landfill leachate treatment in SBR. Capodici *et al.* (2014) found 88% COD reduction during co-treatment of landfill leachate and wastewater using SBR. The comparatively moderate level of COD reduction achieved in this study could be due to the presence of higher concentrations of refractory substances such as the diverse mixture of humic acids, hydrocarbons, proteins, lipids, carbohydrates, carboxylic acids, amino acids, surfactants, detergents, wetting agents, emulsifiers and hydrophilic acids, commonly found in stabilized leachate (Wiszniewski *et al.* 2006).

Nitrification efficiency in SBR was 89% at the end of the oxic phase. The pH in SBR was always maintained in the range of 7–8 which supports and induces the growth of ammonia oxidizing bacteria and nitrite oxidizing bacteria, resulting in significant ammonia removal through nitrification. The major process of ammonia removal was nitrification in SBR. pH was always in the range favorable for nitrification to occur. Nitrification can be inhibited as pH drops to less than 6. Further, ammonia removal by volatilization requires pH more than 8.2 which was not the condition in SBR. Uygur & Kargi (2004) recorded 44% of ammonia nitrogen removal during direct leachate treatment in SBR. Neczaj *et al.* (2007) found 75% ammonia removal efficiency for ultrasound pre-treated landfill leachate using SBR. Denitrification efficiency was 80% at the end of the anoxic phase. The slightly lower nitrate reduction recorded in the present study may be due to the toxic effects of landfill leachate that may have affected the

Table 2 | Variations in COD, ammonia, nitrate, phosphate, TSS and turbidity during co-treatment of landfill leachate and municipal wastewater in SBR

Co-treatment efficiency

Parameters	SBR influent	SBR effluent	Reduction (%)
COD (mg/L)	1732 ± 68	609 ± 44	65 ± 1.2
Ammonia (mg/L)	144 ± 5.4	16 ± 2.1	89 ± 1.1
Nitrate (mg/L)	38.1 ± 2.1 (nitrification initial)	116 ± 0.9 (nitrification final)	–
Nitrate (mg/L)	116 ± 0.9 (denitrification initial)	23 ± 1.4 (denitrification final)	80 ± 1.4
Phosphate (mg/L)	2.2 ± 0.2	0.8 ± 0.3	64 ± 7.8
TSS (mg/L)	5314 ± 192	1205 ± 78	77 ± 2.3
Turbidity (NTU)	87 ± 3.5	18 ± 0.7	79 ± 0.01

growth of denitrifying bacteria (Ksiazek *et al.* 2014). The present study also achieved efficient phosphate and TSS removal at 64 and 77%, respectively. SBR operated for nitrification-denitrification was effective to remove ammonia, nitrate, phosphate and COD. Since the quality of SBR effluent was not within the discharge criteria, it was subjected to electrocoagulation for post-treatment to enhance COD, nutrients and TSS removal efficiency.

Electrocoagulation treatment

Changes in pH, EC, TSS, COD, ammonia, nitrate, and phosphate of SBR effluent following electrocoagulation as post-treatment are presented in Figure 2(a)–2(f) (7 cm – mode 1) and Figure 3(a)–3(f) (5 cm – mode 2). The major reason for using two different electrodes spacing is the fact that the distance between the electrodes has a significant effect

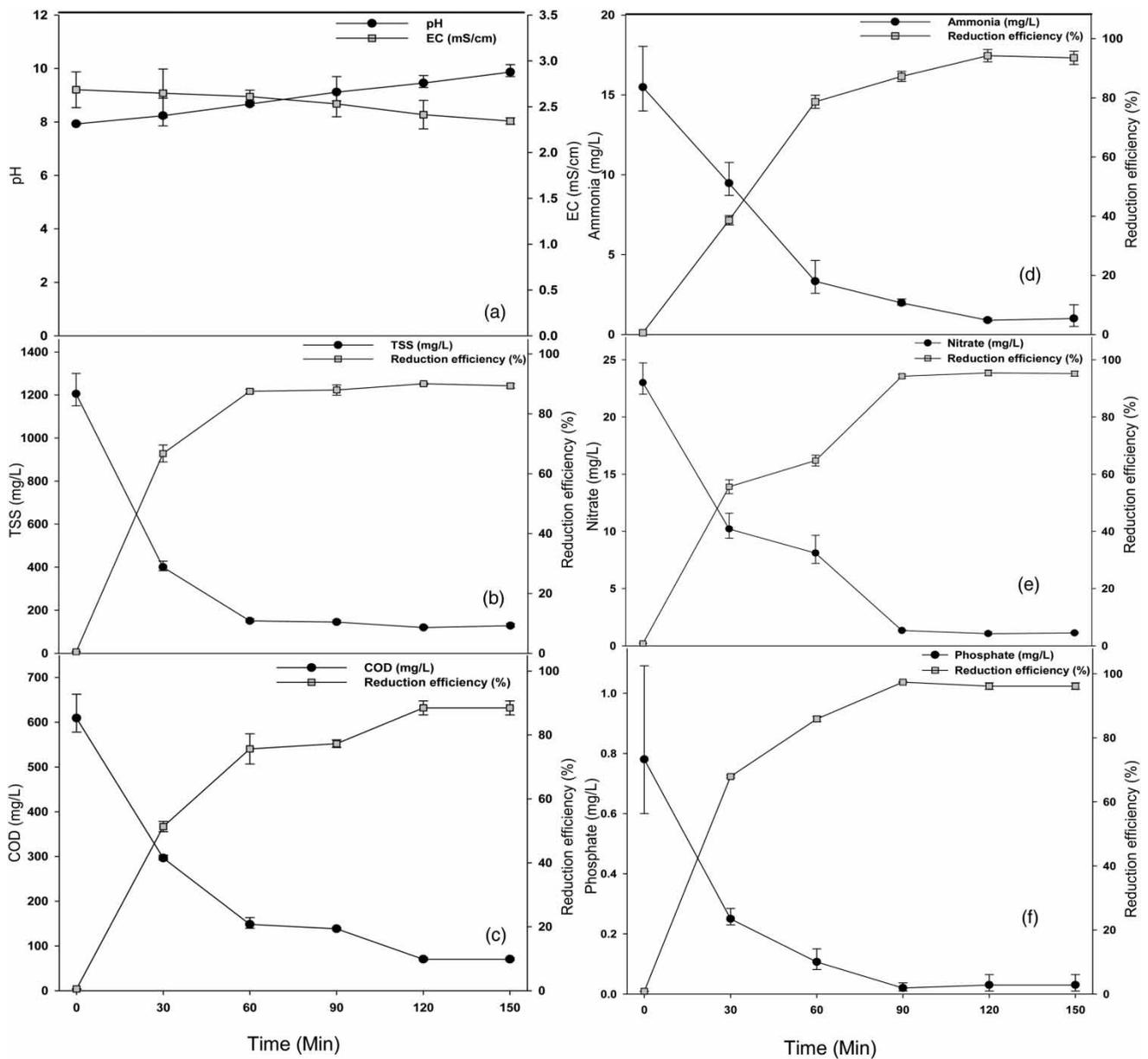


Figure 2 | Variations in (a) pH and EC, (b) TSS, (c) COD, (d) ammonia, (e) nitrate, and (f) phosphate during electro-coagulation with 7 cm of electrode spacing (mode 1).

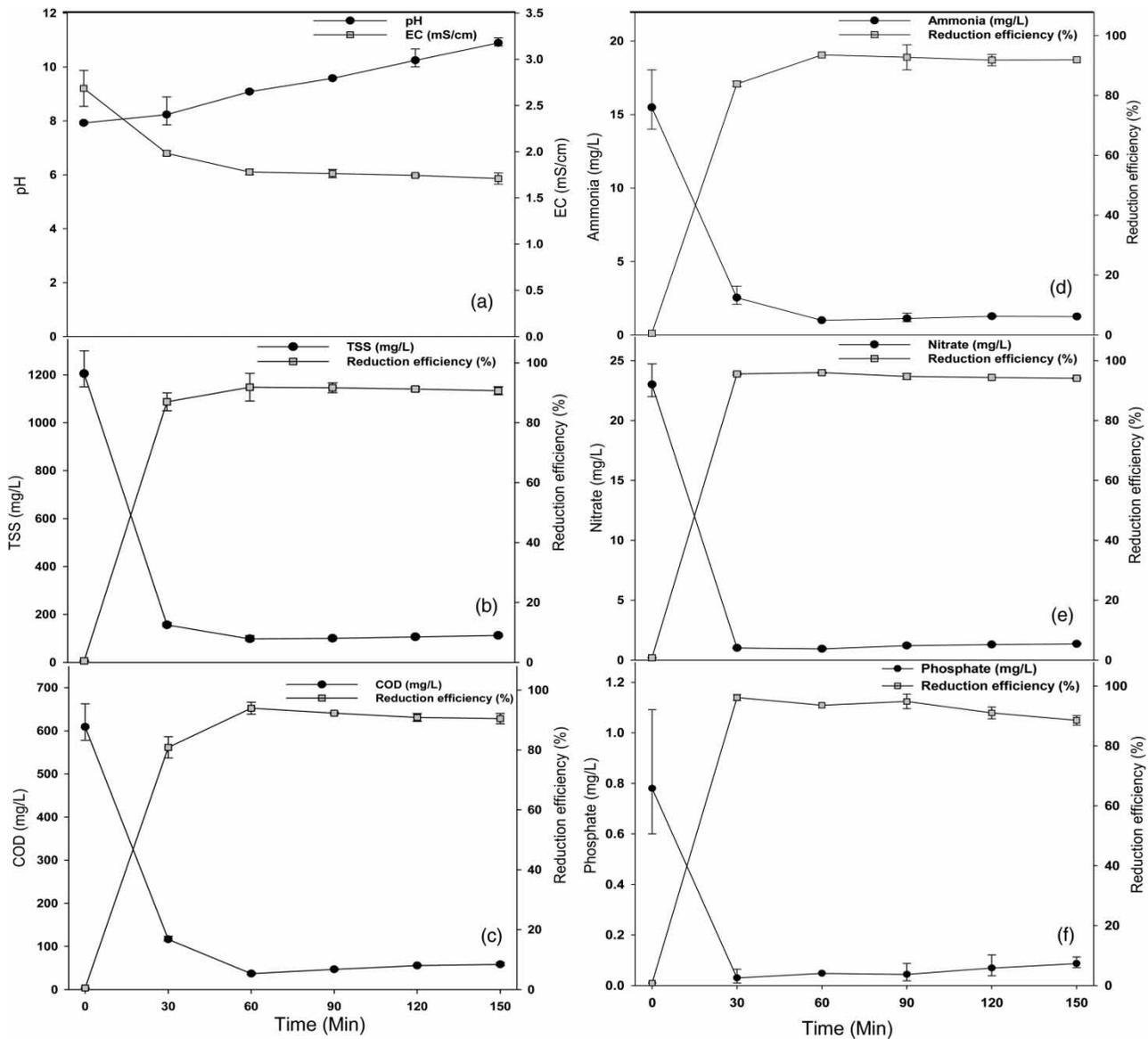


Figure 3 | Variations in (a) pH and EC, (b) TSS, (c) COD, (d) ammonia, (e) nitrate, and (f) phosphate during electro-coagulation with 5 cm electrode spacing (mode 2).

on the electrocoagulation efficiency for contaminant reduction (Sahu *et al.* 2014). Closeness of the electrodes enhances mass transfer characteristics, decreases ohmic loss and energy consumption during the process (Sahu *et al.* 2014).

The pH in the reactor slowly increased from 7.9 to 9.9 during mode 1 operation in electrocoagulation treatment while it increased to a final pH of 10.9 in mode 2. The pH increase is due to electrolytic oxidation of the sacrificial

aluminum anode which combines with hydroxide ions producing aluminum hydroxide (Elazzouzi *et al.* 2017). On the other hand, Li *et al.* (2011) concluded that increasing pH from acidic to basic increases the COD and ammonia removal efficiency as they found maximum COD reduction in the pH range of 7.5–10. The results revealed that maximum 88% COD and 90% TSS reduction could be achieved at 120 min reaction time with mode 1, while mode 2 was slightly more efficient whereby 94% COD and

92% TSS removal occurred at a shorter reaction time of 60 min. The total reduction of COD and TSS increased to 98% through combined SBR and electrocoagulation for co-treatment of landfill leachate and wastewater. The final COD concentration was 37 mg/L and the final TSS concentration was 98 mg/L which met the Indian standards (Central Pollution Control Board, India) for discharge of treated wastewater in inland surface water.

Ammonia and nitrate were almost completely removed from SBR effluent following electrocoagulation as post-treatment with respective final concentrations of 1 and 0.9 mg/L, respectively. Thus, 99% ammonia and nitrate removal were attained with both operating modes in electrocoagulation. Li et al. (2011) reported a maximum 39% ammonia removal during direct landfill leachate treatment using the electrocoagulation process. The reason for higher COD, TSS and total nitrogen reduction attained in the present study could be due to the use of SBR followed by electrocoagulation. Electrocoagulation also resulted in higher levels of phosphate removal, particularly with mode 2 operations which led to 99% phosphate removal in 30 min of reaction while with mode 1 it took 120 min to achieve 99% removal of phosphate. The electrocoagulation cycle time in this batch mode study was kept at 150 min to ensure that there was no change in removal efficiency once maximum removal was attained.

Univariate ANOVA test was used to establish the statistical relationship between inter-electrode spacing and

effluent characteristics where a significant difference was found among removal efficiencies at different electrodes distances studied for various parameters ($F_{3,86} = 31.38$, $P < 0.001$). ANOVA indicated that there is a positive impact of lesser inter-electrode spacing on electrocoagulation process efficiency where the best treatment was obtained at 5 cm compared to 7 cm. A summary of combined SBR + electrocoagulation treatment is presented in Table 3. The results of the present study indicated that a combination of SBR and electrocoagulation as a post-treatment option could be used as a suitable alternative to treat complex landfill leachate with municipal wastewater in an effective manner. Furthermore, compared to the 7 cm distance between the electrodes (mode 1), 5 cm spacing between the electrodes (mode 2) was found to be more efficient for electrocoagulation of landfill leachate and municipal wastewater mixture.

CONCLUSIONS

SBR was effective for both nitrification and denitrification as evident from ammonia and nitrate removal of 89 and 80%, respectively. SBR was also effective for phosphate removal, though COD reduction was moderate only. The results showed that SBR was efficient for direct co-treatment of landfill leachate and municipal wastewater, but the treated waste could not meet the discharge criteria as per the

Table 3 | Summary of combined treatment efficiency

Parameters	7 cm electrode spacing (maximum efficiency at 120 min of total reaction time)					5 cm electrode spacing (maximum efficiency at 60 and 30 min for phosphate)				Effluent Standards for Discharge (mg/L) (CPCB, India) ^a
	SBR initial (mg/L)	SBR final (mg/L)	Electro-coagulation initial (mg/L)	Electro-coagulation final (mg/L)	Electro-coagulation efficiency (%)	Combined process efficiency (%)	Electro-coagulation final (mg/L)	Electro-coagulation efficiency (%)	Combined process efficiency (%)	
COD	1732	609	609	70	88.5	96	37	94	98	250
TSS	5314	1205	1,205	120	90	98	98	92	98	100
Ammonia	144	16	16	0.9	94	99	1	94	99	50
Nitrate	116	23	23	1.1	95	99	0.9	96	99	10
Phosphate	2.2	0.8	0.8	0.02	97	99	0.03	96	99	5

^aEffluent standards for discharge in inland surface water as per the Central Pollution Control Board (CPCB), India.

Indian standards. Electrocoagulation was tested and found to be an efficient post-treatment option for SBR effluent as it enhanced the COD and TSS reduction. Electrocoagulation also removed ammonia, nitrate, and phosphate to high levels at a short reaction time of 30 min at 5 cm spacing. During electrocoagulation, as the pH increased with time, there was a consequent EC decline which led to the removal of COD, particulate matter and nutrients. The combined SBR + electrocoagulation proved to be an efficient process to co-treat landfill leachate and municipal wastewater. Further studies on linking SBR with electrocoagulation in continuous mode with important variables such as HRT, current density and different electrode materials combination, will help to optimize the co-treatment process.

ACKNOWLEDGEMENTS

The infrastructure and support of Birla Institute of Technology, Mesra is gratefully acknowledged for this research work. We also thank the anonymous reviewers for the comments which helped us to improve the manuscript.

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First received 24 May 2019; accepted in revised form 20 January 2020. Available online 17 February 2020