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Coagulation and electrocoagulation for co-treatment of stabilized landfill leachate and municipal wastewater

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ABSTRACT

Landfill leachate and municipal wastewater at various ratios (1:20, 1:10, 1:7 and 1:5) were subjected to coagulation and electrocoagulation (EC). Alum was used in conventional coagulation at pH 6 and aluminum plate as electrode was used in EC at a current density of 386 A/m^2 with 5 cm inter electrode spacing. Treatment efficiency was assessed from removal of chemical oxygen demand (COD), total suspended solids (TSS), turbidity, ammonia, nitrate and phosphate. At 1:5 ratio of landfill leachate to municipal wastewater, highest COD removal was with 3.8 g/L alum whereas highest turbidity removal was with 3.3 g/L alum during coagulation. EC exhibited almost similar removal efficiency for all the parameters at different ratios tested except for COD which was considerably higher at 1:20 ratio. Aluminum consumption from electrode was 0.7 g/L following EC as compared to 3.8 g/L alum used in coagulation. The amount of sludge produced was found to be higher with EC as compared to coagulation which could be due to the fact that the electrochemical method was performed for a longer duration than conventional coagulation. For minimal sludge generation, EC reaction time should be ~30 min. Further studies with EC process on costing and sludge generation will help to advance the technology for wastewater treatment.

Key words | aluminum electrode, aluminum sulphate, coagulation, electrocoagulation, landfill leachate, municipal wastewater

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INTRODUCTION

Open dumping of municipal solid wastes, often in unlined sites, continues to be followed in many countries, which creates several environmental problems. One of the major challenges in solid wastes dumpsites is landfill leachate management, mainly due to high variations in its composition and quantity throughout the year. Landfill leachate are complex wastewater generated from waste dumps due to precipitation, biochemical processes in disposed wastes and inherent water content of waste itself which penetrates through the waste layers (Adeolu *et al.* 2011). Quantity and quality of landfill leachate generated in the landfills depends upon various factors such as moisture content, compaction, refuse composition, dumpsite age, liquid wastes co-disposal, pretreatment, particle size, density, precipitation, groundwater intrusion, irrigation, recirculation, settlement, vegetation, cover, gas and heat generation and transport (Renou *et al.* 2008). In addition, climatic factors such as precipitation, seasonal variations, intensity of sunlight, and humidity also play an important role in determining the leachate quality. Leachate represents potential threats to the environment as it may pollute aquatic systems and surrounding soils (Adeolu *et al.* 2011; Xie *et al.* 2016). Even after years of landfill closure, leachate continues to form due to slow natural waste biodegradation processes in landfill, necessitating its capture, storage, treatment and disposal (Labanowski *et al.* 2010).

Landfill is generally classified into three stages based on its age, young (<5 y), medium (5–10 y) and stabilized (>10 y)

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(Foo & Hameed 2009). Biochemical oxygen demand/chemical oxygen demand (BOD/COD) ratio of young landfill leachate is 0.5–1.0, medium landfill leachate is 0.1–0.5 and old landfill leachate is <0.1. Among the different landfill leachate characteristics, BOD/COD ratio is regularly used as the best representative of landfill leachate age as these are directly indicative of leachate's level of biodegradability. Young landfill leachate contains elevated concentrations of easily degradable organic matter such as volatile fatty acids and has a high BOD/COD ratio. The BOD/COD ratio in stabilized landfill leachate decreases with time as it is the non-biodegradable part of COD that largely remains.

An on-site landfill leachate treatment system may be difficult to establish and operate mainly due to higher costs and practicality issues such as leachate availability throughout the year, which may be the case in most landfills. Therefore, treatment of leachate with municipal wastewater in sewage treatment plants can be a good option. In addition to this, owing to the varying nature of leachate, mixing of landfill leachate with municipal wastewater helps in sustaining the stability required for leachate treatment to meet the stringent discharge standards. Stabilized landfill leachate is particularly difficult to treat due to a low BOD/COD ratio signifying the presence of high amounts of refractory compounds (Ranjan *et al.* 2016). In such cases, often a combination of treatment is required rather than a standalone treatment system for effective landfill leachate treatment. Physico-chemical processes appear to be better suited for both pre-treatment and post-treatment for stabilized landfill leachate.

Various physico-chemical processes such as adsorption, advanced oxidation processes, ammonia stripping and coagulation-flocculation have been used for leachate treatment (Renou *et al.* 2008). Coagulation-flocculation has been found to be useful in COD removal and total suspended solids (TSS) removal up to 90% depending on the contaminants and coagulant types (Boumechhour *et al.* 2012). Coagulation-flocculation involves the destabilization of colloidal particles charge by the addition of coagulants which leads to the formation of flocs through collisions of unstable particles and their aggregation as a soft mix which gets separated from liquid by settling or by application of dissolved air floatation (Canizares *et al.* 2009). pH, coagulant dose and settling time are some of the important processing parameters that influence the coagulation-flocculation process for wastewater treatment (Ayoub *et al.* 2001).

An alternative technique to coagulation-flocculation that has been successfully applied to treat various industrial wastewaters is electrocoagulation (EC) (Khemis *et al.* 2006). EC is a process that forms coagulants through electrodissolution of sacrificial anode(s), usually aluminum or iron, which leads to the generation of hydrolysis products that destabilize various pollutants. Destabilized pollutants aggregate to form flocs which are skimmed from the surface when bubbles of hydrogen produced at the cathode either allow floatation or sedimentation to occur (Ricordel & Djelal 2014). Increasing the application of EC as the preferred treatment can be attributed to the easy automation, easy operation, no need to add chemicals and low operating costs compared to the conventional coagulation process (Lacasa *et al.* 2011). The main process influencing parameters in EC are pH, current density, electrode material, electrodes spacing and reaction time (Fernandes *et al.* 2015). EC can be a suitable option for landfill leachate treatment due to its high electrical conductivity and chloride concentration (Labanowski *et al.* 2010). High electrical conductivity of effluent has the capability to limit temperature variations of solution known as Joule effects (Donini *et al.* 1994). In addition to this, chloride ions in the effluent controls electrode dissolution by increasing the conductivity of solution which in turn can decrease the energy consumption (Labanowski *et al.* 2010).

The major aim of this study was to compare conventional coagulation and EC as a pre-treatment option with aluminum as coagulant for co-treatment of stabilized landfill leachate and municipal wastewater at different ratios. COD, TSS, turbidity, ammonia, nitrate and phosphate removal efficiency were used to evaluate both the processes.

MATERIALS AND METHODS

Landfill leachate and municipal wastewater collection

Municipal wastewater and landfill leachate mixture was used as influent for coagulation and EC experiments. Landfill leachate samples were obtained from an unlined open MSW dumpsite in Ranchi, Jharkhand, India. Open dumping

of MSW in Jhiri dumping yard in Ranchi has been ongoing for the last 15 years in an area of 22 acres. Ranchi's elevation from mean sea level is 651 m. Climate is subtropical with heavy rainfall during the monsoon season (June–September). Summer months are from March to June and winter from November to February. Mean annual temperature is 29.6 °C (maximum) and 18 °C (minimum). Mean annual precipitation is 1,400 mm whereas mean number of rainy days are 75 and most of the rainfall occurs during June to September (Source: Indian Meteorological Department). The dumpsite receives ~700,000 kg of mixed wastes daily. Landfill receives waste from residential areas, commercial establishments, vegetable and fruit markets, meat and fish markets and from hotels and restaurants. No segregation and compaction of waste is carried out at the landfill and covering of soil over deposited waste is also not carried out, exposing the waste to all the environmental processes. Leachate samples were collected from a pond formed in a large depression in the landfill area. For each sampling event five different sampling points were selected, one in the center and four in the periphery of the leachate pond to obtain an aggregate sample of 5 L. Leachate samples were collected using a clean HDPE bottle tied to a telescopic rod. Landfill leachate samples were collected twice in a month and kept under cold storage in the lab. Prior use leachate samples were always brought to room temperature (20–25 °C) for about 2 h. Landfill leachate samples were mixed manually for re-suspension of settled solids before carrying out the experiments. Raw municipal wastewater was collected from the equalization tank of sewage treatment plant located at Birla Institute of Technology, Mesra, Ranchi. Landfill leachate and municipal wastewater were thoroughly mixed to formulate different ratios (1:20, 1:10, 1:7 and 1:5) for each batch run of coagulation and EC.

Coagulation-flocculation experiments

Coagulation-flocculation experiments were carried out in standard jar test apparatus using 1 L beakers with 500 mL wastewater mixture. Experiments were conducted in batch mode to study the influence of coagulant dosage on coagulation-flocculation. Aluminum doses used were 2.8, 3.3, 3.8, 4.3, 4.8 and 5.3 g/L. Experiments were conducted at the respective ratios of 1:20, 1:10, 1:7 and 1:5 landfill leachate and municipal wastewater with one ratio at a time. For experiments, beakers were placed on the jar test apparatus followed by the addition of wastewater mixture and agitation was started. Once the required rpm for rapid mixing was attained, different doses of alum were added for the treatment process. The contact time consisted of 1 min of rapid mixing at 200 rpm and 20 min of slow mixing at 60 rpm followed by a settling time of 30 min for sedimentation. Initial pH was adjusted to 6 using 1 N H_2SO_4 for all the coagulation experimental runs. Treatment efficiency was determined by sampling at the start and completion of reaction to measure COD, TSS, turbidity, ammonia, nitrate and phosphate.

EC experiments

EC was carried out in a bench scale setup of 0.75 L at respective ratios of 1:20, 1:10, 1:7 and 1:5 landfill leachate and municipal wastewater. A stainless steel plate was used as cathode and an aluminum plate was used as sacrificial anode. Both the electrodes were of $0.07 \times 0.065 \times 0.004$ m dimensions. The distance between anode and cathode was kept at 5 cm in the reactor. The electrodes were connected to a DC power supply providing a current density of 386 A/m^2 at 12 V. A magnetic stirrer was used to agitate the contents of the EC mixture. All the runs were performed at room temperature without any pH adjustment. The addition of salt as supporting electrolyte was not needed for increasing the electrical conductivity of wastewater mixture, hence all the experiments were performed with the initial conductivity of wastewater mixture. EC time was 90 min while samples were retrieved at 30 min intervals for physico-chemical analysis. Before each run of EC, aluminum electrodes were cleaned and weighed; after the process the electrode was scraped to remove the layers formed during electrolysis and weighed to estimate the amount of aluminum consumed during treatment.

Chemical analysis

Physico-chemical characteristics of landfill leachate and municipal wastewater were carried out to assess the treatment efficiency of both the processes. Various chemical analyses were performed as per the standard methods

which are briefly described here (APHA 1998). pH and electrical conductivity of samples were determined using a multi parameter meter (HORIBA, Japan). TSS in samples was measured using pre-weighed 0.45 μm filter connected to vacuum filtration apparatus. The residue retained on the filter was dried to a constant weight at 105 °C, cooled in a desiccator and weighed. The process of drying, cooling in the desiccator and weighing was carried out until a constant weight was recorded. COD in samples was analyzed without any delay using the open reflux method, in the presence of excess potassium dichromate under highly acidic conditions. After the digestion, unreduced potassium dichromate was titrated with ferrous ammonium sulfate to determine the oxidizable substances present as oxygen equivalent. For this analysis, turbidity was determined in samples immediately using a nephelometer before the measurements samples were gently agitated. NH_3-N analysis was carried out soon after sampling following the phenate method. NO_3-N was estimated immediately after sampling using a nitrate ion electrode (YSI, USA). PO_4^{3-} was measured using the stannous chloride method as per the direct procedure detailed in *Standard Methods* (APHA 1998). Sludge production was determined from the final treated wastewater mixture that was subjected to coagulation and EC after 30 min of settling time in a graduated measuring cylinder. The data reported consist of the average of duplicate analysis from all the experimental runs.

Table 1 | Physico-chemical characteristics of municipal wastewater and stabilized landfill leachate

Parameter	Municipal wastewater	Stabilized landfill leachate
pH	7.0 ± 0.3	7.8 ± 0.4
EC (mS/cm)	0.74 ± 0.2	8.9 ± 1.9
TDS	450 ± 40	6,700 ± 3,500
TSS	460 ± 20	4,400 ± 2,500
BOD	240 ± 100	60 ± 45
COD	500 ± 140	3,850 ± 230
Ammonia	60 ± 2.2	638 ± 4
Nitrate	0	63 ± 4
Phosphate	40 ± 4	55 ± 17

All values are in mg/L except pH and EC. $n = 10$ for municipal wastewater and landfill leachate.

RESULTS AND DISCUSSION

Landfill leachate and municipal wastewater characteristics

Landfill leachate is in a stabilized condition as evident from high COD and low BOD_5 (Table 1). The BOD_5/COD ratio signifies the proportion of biodegradable organic matter in the leachate. Young leachate contains high concentrations of easily degradable organic matter resulting in a high BOD_5/COD (>0.5) ratio compared to stabilized landfill leachate (<0.1) (Foo & Hameed 2009). Leachate from the dumpsite in Ranchi had a very low BOD_5/COD ratio (0.015), representing low biodegradability of organic matter. Leachate pH was slightly higher than neutral, higher ammonia and comparatively lower nitrate concentration also substantiated that the landfill is in a methanogenic phase. Municipal wastewater contained moderate BOD and COD concentration.

Coagulation process for co-treatment of landfill leachate with municipal wastewater

Different ratios (1:20, 1:10, 1:7 and 1:5) of landfill leachate with municipal wastewater were treated using coagulation at different alum dosages. The ratios used in this study were selected to simulate the conditions which might be in the reasonable range for field-scale applications. Moreover, it has been reported that higher leachate concentration affects the co-treatment process and leachate should not exceed 20% of the total wastewater mixture (Çeçen & Çakiroglu 2001; Mojiri *et al.* 2016; Ranjan *et al.* 2016).

The removal efficiency of COD, TSS, turbidity, ammonia, nitrate and phosphate at different ratios of landfill leachate and municipal wastewater are shown in Figure 1(a)–1(f). Among the different ratios tested, COD removal at the lowest dilution (1:5) was found to be better and the maximum COD removal was reached with the 3.8 g/L dose for all the ratios tested except for the 1:7 ratio which was at 3.3 g/L. TSS removal with 1:20 and 1:10 ratio increased up to 3.3 g/L alum dose, thereafter any increase in dose did not have any significant effect. The highest TSS removal at the 1:20 and 1:10 ratios could be due to the presence of lower solids concentrations as the leachate volume was low compared to the

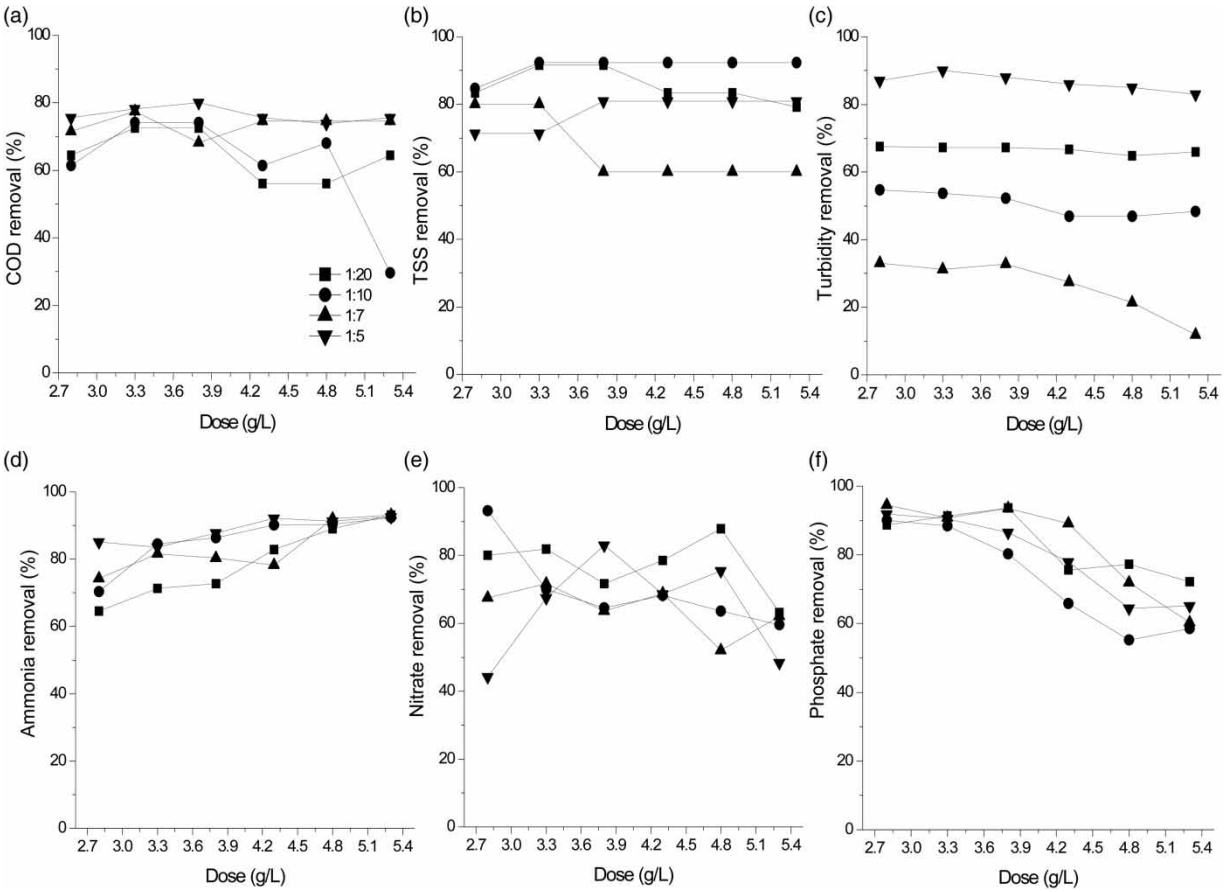

Figure 1 | Effect of coagulant dosage during coagulation on the removal efficiency of (a) COD, (b) TSS, (c) turbidity, (d) ammonia, (e) nitrate, and (f) phosphate from landfill leachate and municipal wastewater at 1:20, 1:10, 1:7 and 1:5 ratio.

1:5 ratio. TSS removal at the 1:7 ratio remained stable at 2.8 and 3.3 g/L whereas at 3.8 g/L it decreased and remained somewhat similar after this dose. TSS removal at the 1:5 ratio remained stable at 2.8 and 3.3 g/L whereas at 3.8 g/L it increased and remained somewhat similar after this dose. Maximum turbidity removal was at the 1:5 ratio at 3.3 g/L alum dose, whereas for other ratios tested the highest removal was at 2.8 g/L. Turbidity reduction decreased at the 1:5 ratio with >3.3 g/L alum dose and in other ratios turbidity removal declined after the 2.8 g/L alum dose. Ammonia removal increased with dose at all the ratios tested except at the 1:7 ratio where the ammonia removal decreased up to 4.3 g/L alum followed by an increase at dose >4.8 g/L. Ammonia gets removed at acidic pH as the ammonium ions get adsorbed on aluminum hydroxide precipitates formed following alum addition. Ammonia removal results are in accordance with the work of Trabelsi *et al.* (2013) where they found similar ammonia removal efficiency of SBR treated landfill leachate and municipal wastewater at ratios of 9:1, 8:2, 7:3 and 5:5. Nitrate removal was poor as compared to other parameters and did not exhibit any specific trend at all the ratios tested. Such a trend on nitrate removal could be attributed to the significant increase in wastewater electrical conductivity (see Figure 2(a)) which could have increased the competition between nitrate ions and coagulant counter ions for adsorption onto the metal hydroxide precipitates and which may also have reduced the width of double layers formed around precipitate complexes (Lacasa *et al.* 2011). Maximum phosphate removal with all the ratios tested was found to be at 2.8 g/L alum dose, thereafter phosphate removal mainly declined with the increase in alum dose. Phosphate was removed mainly by forming insoluble phosphoric compounds that precipitates and settles out from wastewater (Xie *et al.* 2005).

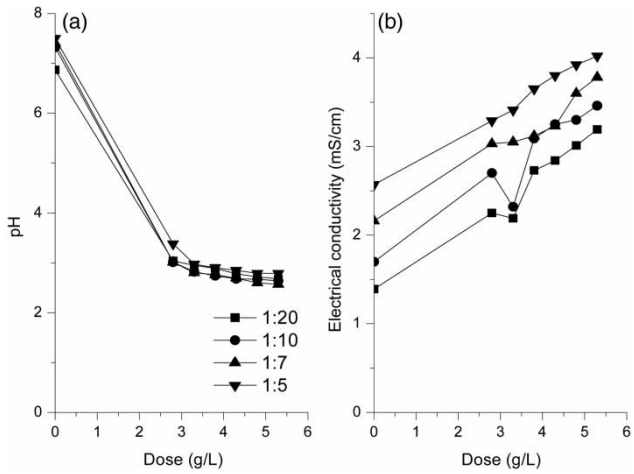

Figure 2 | Variations in (a) pH and (b) electrical conductivity during coagulation at different doses of aluminum sulphate at 1:20, 1:10, 1:7 and 1:5 ratios of landfill leachate and municipal wastewater.

Alum at 3.8 g/L dose for the 1:5 ratio of landfill leachate with municipal wastewater led to better treatment; respectively, COD, turbidity and TSS removal were 80, 88 and 81%. As the alum dose increased to 4.3 g/L, COD and turbidity decreased to 75 and 86% while TSS remained constant at 81%. Reasons for the higher treatment efficiency could be attributed to a significant change in pH from 7.5 to 3.0 (Figure 2), which removes colloids due to both charge neutralization and enmeshment of pollutants on aluminum hydroxide precipitates (Canizares *et al.* 2009; Gandhimathi *et al.* 2013). The initial pH for all the ratios of landfill leachate and municipal wastewater was adjusted to 6 for an efficient coagulation process, which decreased with an increase in dosage during the treatment due to the acidic nature of alum which consumes alkalinity (Figure 2(a)). As shown in Figure 2(b), the electrical conductivity of the treated wastewater increased with an increase in dose at all ratios of landfill leachate and municipal wastewater which was due to the dissociation of alum and corresponding drop in pH.

EC process for co-treatment of landfill leachate with municipal wastewater

The EC process using 386 A/m^2 current density at 5 cm inter electrode spacing was assessed for the removal of COD, TSS, turbidity, ammonia, nitrate and phosphate from landfill leachate to municipal wastewater mixture at ratios of 1:20, 1:10, 1:7 and 1:5 (Figure 3). Respective COD, TSS and turbidity removal recorded were 73, 53 and 88%, respectively, at the 1:5 ratio of landfill leachate in municipal wastewater after 30 min of reaction which resulted in 0.7 g/L of aluminum consumption (Figure 3(a)–3(c)). Results are in accordance with the work of Ilhan *et al.* (2008) which showed that an application of 348 A/m^2 current density led to 45% COD removal in 30 min reaction time. Further, as the current density was increased to 631 A/m^2, COD removal also increased up to 59% at the same electrolysis time. Hence, an increase in current density in the EC process results in better COD removal. Except for COD removal which was moderate at the low dilutions (1:5, 1:7 and 1:10), EC was found to work with similarly high efficiency for all other parameters regardless of leachate strength.

Ammonia, nitrate and phosphate were removed effectively by 87, 95 and 85%, respectively, after 30 min of electrolysis time with 1:5 ratio of landfill leachate to municipal wastewater (Figure 3(d)–3(f)). Ammonia was also removed around 30 min of electrolysis due to an increase in pH and temperature that converts ammonium into ammonia nitrogen which is stripped with gases formed around the cathode (Ilhan *et al.* 2008).

At alkaline pH with 1:5 ratio landfill leachate and municipal wastewater, COD and TSS were removed by electrolytic dissolution of aluminum anode which produces Al^{3+} and $Al(OH)_2^+$ species and OH^- on the cathodic surface, resulting in an increase in pH up to 90 min of reaction time (Figure 4(a)) (Canizares *et al.* 2009). Further, it has been reported that an increase in electrolysis time causes an increase in pH due to oversaturation of CO_2 in acidic medium which gets released from the effluent by purging of H_2 and O_2 bubbles. Electrical conductivity (Figure 4(b)) also plays an important a role as pH, high initial conductivity of wastewater causes high current efficiency that inhibits electrode passivation (formation of oxide layer on the electrode surface which prevents metal dissolution and electron transfer) (Liu *et al.* 2010).

Three major mechanisms involved in coagulation-based processes are electrical charge neutralization and particles separation, entrapment of particles by coagulant metal hydroxides and destabilization by adsorption to particle surface (vanLoon & Duffy 2011). When aluminum is added to

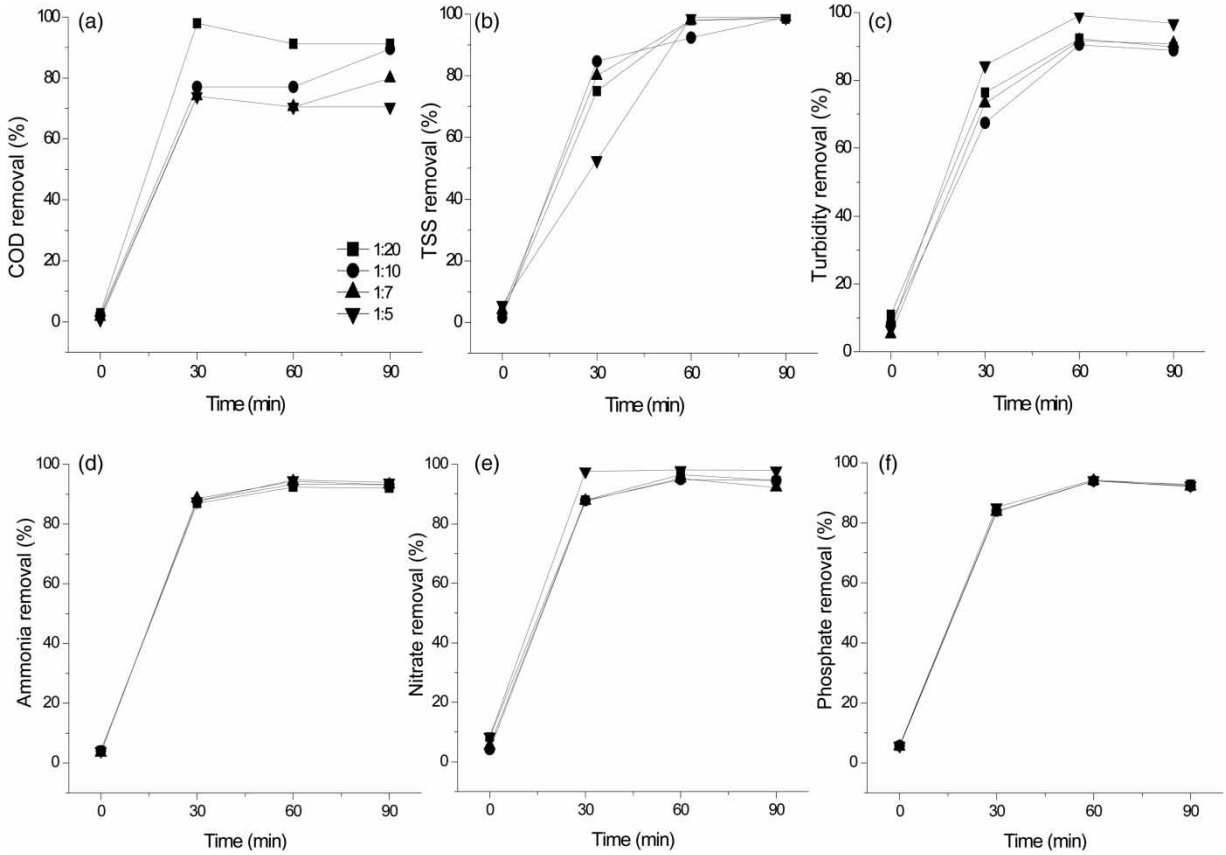

Figure 3 | Variations in (a) COD, (b) TSS, (c) turbidity, (d) ammonia, (e) nitrate, and (f) phosphate removal efficiency by EC at 1:20, 1:10, 1:7 and 1:5 ratio of landfill leachate and municipal wastewater.

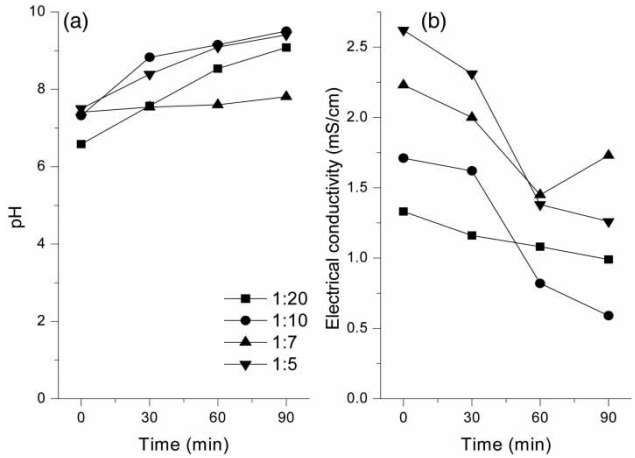

Figure 4 | Variations in (a) pH and (b) EC with time during EC at 1:20, 1:10, 1:7 and 1:5 ratios of landfill leachate and municipal wastewater.

wastewater it forms insoluble aluminum hydroxide, gelatinous floc which settles slowly through the wastewater, removing the suspended materials including nitrate and phosphate mainly by adsorption and sweep precipitation. Most of the treatment reported in this study was due to metal hydroxide formation, precipitation and adsorptive coagulation. For instance, it is known that nitrate can be chemically reduced to ammonia with aluminum powder only at pH in the range of 9–10.5. Ammonia can later be removed by air stripping or other thermal/chemical-based ammonia recovery methods; however, this process works only when the pH is in the mentioned range (Murphy 1991; Emamjomeh & Sivakumar 2005). Since pH was less than that required for chemical denitrification, the major mechanisms for nitrate removal in this study were sweeping coagulation and precipitation (Aghapour *et al.* 2016). Aluminum can react with phosphate to precipitate as highly insoluble aluminum phosphate, but other complex processes also occur such as aluminum hydrolysis which is readily converted to an insoluble hydrous oxide form which also aids in phosphate removal (vanLoon & Duffy 2011).

Both the coagulation and EC processes were found to be effective for removal of colloidal particles, suspended solids and nitrogenous compounds from different mixtures of landfill leachate and municipal wastewater. However, the coagulation process involves the modification of initial pH of effluent for an efficient removal process as compared to the EC process in which the mechanism is effective at the initial pH of the mixture of landfill leachate and municipal wastewater. pH adjustment will involve extra operational costs for the coagulation process in leachate treatment plants. In addition to this, a greater amount of aluminum dose was consumed in the coagulation process (3.8 g/L) as compared to the EC process (0.7 g/L) as shown in Figure 5(a) and 5(b). Further, nitrogen compounds were effectively removed by the EC process (87% NH_3-N and 95% NO_3^--N) whereas the coagulation process led to 80% NH_3-N and 63.6% NO_3^--N removal at 1:5 ratio of landfill leachate to municipal wastewater.

The sludge volume after 30 min of settling time was found to be higher with 90 min EC than 21 min coagulation for mixtures of stabilized landfill leachate and municipal wastewater. In general, EC has been reported to produce less sludge than the chemical coagulation process and the deviation found in the present study could be mainly due to the following two reasons which need to be studied further. First, it is known that generally alum-based flocs are lighter than iron-based flocs and in the EC process the amount of aluminum liberated was very low compared to the chemical coagulant dose used. Second, in the EC process sludge generation was tested after 90 min reaction time whereas the maximum treatment efficiency was consistently found at 30 min reaction time. Hence, applying a treatment time of 30 min or less may reduce the volume of sludge generated. Ricordel & Djelal (2014) have also reported that sludge volume increased with the EC time, mainly due to higher coagulant generation than that required. Thus, short EC time could be tested in future studies to improve the formation of large and dense flocs which could exhibit better settling velocities.

CONCLUSIONS

Conventional coagulation and EC processes were studied for co-treatment of landfill leachate and municipal wastewater at different ratios, i.e. 1:20, 1:10, 1:7 and 1:5. At the lowest dilution, 1:5 ratio, COD removal efficiency was slightly better with coagulation than EC. EC worked better at the highest dilution of 1:20 for COD removal than conventional coagulation. Other than this result on COD, at all other ratios tested EC exhibited better treatment efficiency when compared to the conventional coagulation. The EC process could remove pollutant at a significantly lesser dose of 0.7 g/L than the coagulation process which required 3.8 g/L alum. Further, the EC process can be carried out effectively without any pH adjustment of the

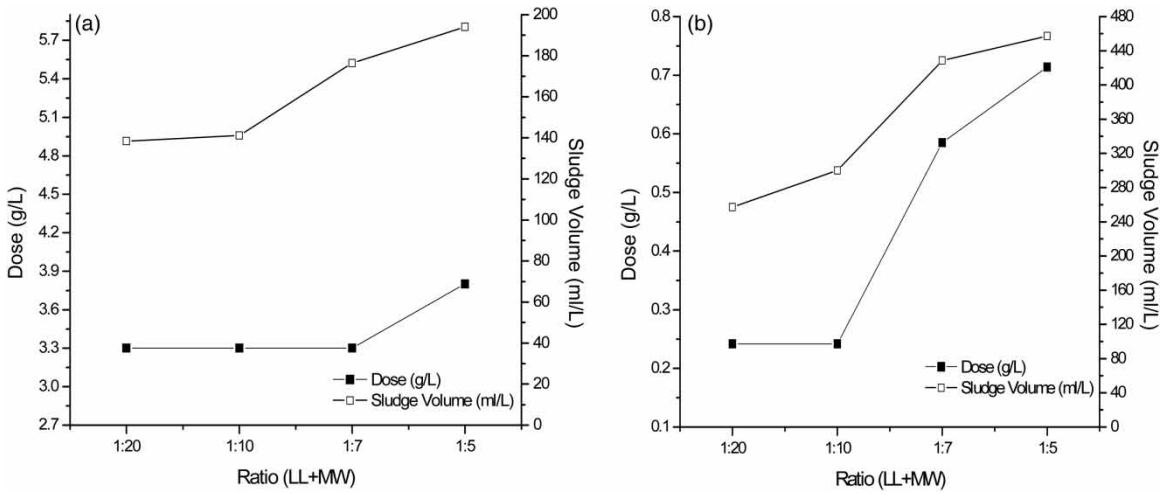

Figure 5 | Coagulant dose consumption and sludge volume generated at 1:20, 1:10, 1:7 and 1:5 ratio of landfill leachate and municipal wastewater with (a) coagulation and (b) EC.

initial reaction mixture whereas alum required pH adjustment to 6. Sludge production was moderately higher in EC in comparison to coagulation, mainly due to increased EC time, thus it can be suggested that EC time should be limited to <30 min for lesser sludge production. Overall, the results indicated that the EC process was moderately better than the coagulation process for pre-treatment of mixture of stabilized landfill leachate and municipal wastewater. Future studies on EC based on reaction time, settling time and electrodes longevity, along with sludge generated and cost estimates, will further improve the comparison between EC and conventional coagulation.

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