

Electrokinetic remediation for textile wastewater contaminated soil

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Graphical abstract



Abstract

Soil contamination due to industrial activities is very common and discharging of partially treated or untreated textile wastewater into nearby environment in developing countries contaminated soils. The remediation of textile wastewater contaminated soils containing residual organic dyes and inorganic salts is risky. It is required to identify an economic and eco-friendly in-situ remediation technique for the textile wastewater contaminated soil. This paper investigates the effective removal of residual textile dyes and inorganic salts from the soil using electrokinetic remediation. Laboratory scale acrylic reactors were utilized and the soil was collected from the contaminated agricultural site. The experiment was optimized for different electrode combination (Iron-iron, Iron-stainless steel, Iron-aluminium, Iron-graphite), electrode distance (3 cm, 5.5 cm, and 10.5 cm) and electrical potential gradient (3V/cm, 4V/cm, and 5V/cm). Tap water was used as electrolytes in order to prevent further contamination during remediation process. An electroosmotic flow of 256 mL was observed at cathode reservoir for Iron-graphite electrodes and these

electrodes removed 93% electrical conductivity and 82% of total organic carbon from soil. It is found that electroosmosis is the dominant mechanism by which residual textile dyes were removed. The proposed remediation technique is found efficient, economic and eco-friendly compared to the existing technologies.

Keywords: Textile dyes, inorganic salts, agricultural soil, electrolytes, electrodes

1. Introduction

The global impact on natural resources has increased over the past 150 years and the environmental concern on soil has begun to emerge especially in the past two decades (Rodríguez-Eugenio et al., 2018). Soil contamination due to industries is a widespread threat globally; textile industry is one of the major threats to the environment, as the wet processing utilizes lot of toxic chemicals, water and generates large amount of wastewater. Discharge of partially or untreated textile wastewater into nearby environment is practiced in developing countries for the last few decades and it affects aquatic organisms, plant growth, groundwater, soil quality, and human health. Similar situation was observed for the last three decades in Tiruppur, a textile hub in Tamil Nadu, Southern India, where deterioration of agricultural soil quality was observed and reported in a recent study (Raman & Mkandawire, 2021). The textile wastewater contaminated soil consists of residue of mixed reactive dyes and inorganic salts, affect soil fertility and it is challenging to remediate the soil. It is mandatory to identify and propose an economic and environmental sustainability in-situ remediation technique for the textile wastewater contaminated soil.

Textile dye spiked soil was remediated using bioremediation (Bhatt *et al.*, 2000; Khalid *et al.*, 2011) and electrokinetic separation followed by electrochemical oxidation (Lee *et al.*, 2009). However it required longer duration and specific laboratory conditions to achieve effective remediation. Plant biomass of subabul seeds

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remediated textile wastewater contaminated soil (Jayanthy et al., 2014). The results confirmed that adsorption of plant biomass could restore soils upto 50% and the remediated soil was again used for the cultivation of green gram. Biostimulation using microbes was coupled to remove dye residue and inorganic salts but acetic acid was used as catholyte, which would contaminate the soil system further (Annamalai et al., 2018). Salinization of agricultural land is another serious environmental challenge due to textile industries pollution and leads to an excessive increase of sodium, chloride and sulfate salts in the soil. Recently, researchers have reported that electrokinetic technology is an effective technique to remediate soil salinity and laboratory studies were conducted for the restoration of soil which contained mainly Na, Cl, and SO₄ ions (Cho et al., 2012; Kim et al., 2012; Lee et al., 2012). In all the studies, Na and Cl were removed easily from the soil (>97%) but the removal of SO₄ was only 62.3%, 25.6% and it was challenging.

Among different technologies the available, electrokinetics is an efficient in-situ treatment technology that remediates soil from both organic and inorganic contaminants. With the application of low intensity direct current across electrode pairs, the contaminants are transported towards respective electrodes depending on their charge and the removal is based on electromigration, electroosmosis, and electrophoresis mechanisms. Even though the electrokinetic technique is utilized mostly to remove heavy metals, it has also been applied to treat organic and mixed contaminants in the recent years (Annamalai et al., 2018; Paiva et al., 2018). However, the electrolyte was consumed among ammonium acetate, sodium dodecyl sulphate, potassium sulfate, sodium sulfate, and hydrochloric acid to remove Na, Cl, SO₄ from soil. The utilized electrodes IrO₂-RuO₂-TiO₂/Ti and Ti electrodes are costly and electrolytes are toxic (Annamalai et al., 2018, 2016, 2014). Hence it is mandatory to find cost-effective electrodes and ecofriendly electrolytes for the remediation of textile wastewater contaminated soils. The potential of electrokinetics in the remediation of textile wastewater contaminated soil with cost-effective electrodes, ecofriendly electrolytes needs detailed investigation.

This paper investigated the electrokinetics remediation using three laboratory scale reactors (R1 - 10 cm x 5 cm x 5.5 cm; R2 - 15 cm x 5 cm x 5.5 cm; R3 - 20 cm x 5 cm x 5.5 cm), made of acrylic for textile wastewater contaminated soil. The soil was collected from the contaminated agricultural field and the soil was packed in the reactor as in-situ condition such as moisture content and density. The electrokinetic remediation was operated at an electrical potential gradient of 3-5V/cm. The removal of textile dyes and inorganic salts were thoroughly investigated using iron (Fe), stainless steel (SS), aluminium (Al), graphite (Gr) electrodes. Tap water was used as electrolytes in order to prevent further contamination. The influencing parameters such as distance between electrodes and the variation in electrical potential gradient were investigated in the remediation process.

2. Materials and Methods

2.1. Collection and characterization of textile wastewater contaminated soil

The contaminated soil from agricultural land from Tiruppur was collected till depth of 15 cm. The samples were dried in shade for two days, powdered if its size was more than 2 mm and analyzed for pH, EC (IS 2720 – Part 21, 26), TOC (Walkley and Black, 1934), Cl, SO4, Na, K, Ca, Mg as per the standard methods (Rice *et al.*, 2017) by extracting the ions from soil using water (1:5) (Margesin and Schinner, 2005).

2.2. Fabrication of reactors and experimental set up

Three laboratory scale reactors (R1 - 10 cm x 5 cm x 5.5 cm; R2 - 15 cm x 5 cm x 5.5 cm; R3 - 20 cm x 5 cm x 5.5 cm), made of acrylic were fabricated for textile wastewater contaminated soil remediation, consists of compartments for soil, anolyte, and catholyte. The soil length designed for reactors R1, R2, and R3 was 2.5 cm, 5 cm, and 10 cm, respectively. All compartments were divided by perforated sheets and filter papers were placed to restrict the movement of soil particles towards electrolyte compartment, as presented in Figure 1. The study was conducted on field contaminated soil, saturated with 20% moisture content and well packed to attain the field density of 1.6 g/cc. The soils utilized were 100 g for R1, 200 g for R2 and 400 g for R3. The electrolytes were filled, the electrodes were placed in the respective compartments and connected to a regulated DC power supply (Model: L3202) purchased from AP Lab Power Supply. An electrical potential gradient of 3-5V/cm was applied to perform electrokinetics. The removal of textile dyes and inorganic salts were studied using iron (Fe), stainless steel (SS), aluminium (Al), graphite (Gr) electrodes and their details are presented in Figure 2. Increase in the contact area of electrodes, increases the removal of contaminants from the soil. Hence, preliminary study was conducted using the different shapes of the electrodes and finally rectangular size electrodes were optimized and utilized for further investigation. Tap water was used as electrolytes in order to prevent further contamination during experiments and the distance between electrodes on remediation process were studied, as presented in Figure 3.



Figure 1. Fabricated reactors (R1, R2, and R3) and DC power supply



Figure 2. Electrodes utilized for electrokinetics



Figure 3. Distance between electrodes on electrokinetic remediation

Table 1. Contaminated soil properties before and after remediation

Physicochemical Properties	Contaminated Soil APS9	Remediated Soil after Electrokinetics				
		APS9-1	APS9-2	APS9-3	APS9-4	
рН	8.9	7.7	7.8	7.9	7.6	
EC (dS/m)	4.8	0.9	1.5	2.3	0.3	
TOC (%)	6.1	2.3	3.4	4.2	1.1	
Cl (g/kg)	7.6	0.8	2.2	4.9	0.2	
SO ₄ (g/kg)	4.2	1.3	2.8	3.5	0.5	
Na (g/kg)	8.2	1.0	2.3	4.9	0.2	
K (g/kg)	0.5	0.3	0.4	0.4	0.3	
Ca (g/kg)	1.8	1.4	1.6	1.7	1.3	
Mg (g/kg)	0.2	0.2	0.2	0.2	0.2	

Table 2. Characteristics of anolyte before and after remediation

Physicochemical Properties	Ten Weter TDW	TPW at Anode Reservoir after Remediation				
	Tap water TPW	TPWA-1	TPWA-2	TPWA-3	TPWA-4	
рН	7.0	3.2	3.5	3.7	3.0	
EC (dS/m)	0.21	0.48	0.42	0.38	0.52	
TDS (mg/L)	389	964	756	532	1156	
COD (mg/L)	0	0	0	0	0	
TOC (mg/L)	0	0	0	0	0	
Cl (mg/L)	112	546	432	381	658	
SO ₄ (mg/L)	41	224	196	147	289	
Na (mg/L)	68	34	46	48	22	
K (mg/L)	23	19	20	21	18	
Ca (mg/L)	82	61	68	73	55	
Mg (mg/L)	49	26	29	32	23	

Table 3. Characteristics of catholyte before and after remediation

Physicochemical Properties	Tap Water TPW —	TPW at Cathode Reservoir after Remediation			
		TPWC-1	TPWC-2	TPWC-3	TPWC-4
рН	7.0	10.8	10.6	10.4	11
EC (dS/m)	0.21	0.59	0.45	0.38	0.67
TDS (mg/L)	389	1326	1146	1085	1548
COD (mg/L)	0	759	632	517	886
TOC (mg/L)	0	286	214	182	325
Cl (mg/L)	112	163	147	126	185
SO ₄ (mg/L)	41	98	85	72	104
Na (mg/L)	68	821	695	482	867
K (mg/L)	23	96	84	72	109
Ca (mg/L)	82	165	147	123	186
Mg (mg/L)	49	107	98	82	114

2.3. Detailed investigation of effective removal of residual textile dyes and inorganic salts from soil

The cost-effective electrodes and eco-friendly electrolytes were opted for maximum removal of textile dyes and inorganic salts from soil. Depending upon electrode distance (3 cm, 5.5 cm, and 10.5 cm), electrode combination (Fe-Fe, Fe-SS, Fe-Al, and Fe-Gr), electrical potential gradient (3 V/cm, 4 V/cm, and 5 V/cm), and the electrokinetic experiments were conducted for 8 h. The pH was monitored for every 30 min at electrode reservoirs using digital pH meter. The electroosmotic flow during the experiment was monitored and the flow of electrolytes with contaminants into cathode reservoir was collected in a beaker. The anolyte was filled into the anode reservoir using burette arrangement.



Figure 4. Laboratory scale electrokinetic remediation

All the experiments were triplicated in order to obtain the results with a standard deviation error of less than 5%. The initial characterization of electrolytes for pH, TDS, Cl, SO₄, COD, Na, K, Ca, and Mg was done. The pH at electrode reservoirs and electroosmotic flow were monitored during the experiment. Once the experiment was over, the electrolytes were collected and characterized for pH, TDS, Cl, SO₄, COD, Na, K, Ca, and Mg. The soil was sliced into equal parts of 2.5 cm width along the longitudinal direction, and then characterized for pH, EC, Cl, SO₄, TOC, Na, K, Ca, and Mg. The detailed study on laboratory scale electrokinetic remediation is presented in Figure 4.

3. Results and Discussion

For different electrodes (Fe-Fe, Fe-SS, Fe-Al, and Fe-Gr), the reactor R1 packed with contaminated soil of electrode distance of 3 cm and tap water as electrolyte were used for electrokinetic remediation at 5V/cm electrical potential gradient. The characteristics of contaminated soil, anolyte and catholyte before and after remediation using Fe-Fe (APS9-1, TPWA-1, and TPWC-1), Fe-SS (APS9-2, TPWA-2, and TPWC-2), Fe-Al (APS9-3, TPWA-3, and TPWC-3), and Fe-Gr (APS9-4, TPWA-4, and TPWC-4) electrodes are presented in Tables 1, 2 and 3.



Figure 5. Electroosmotic flow using different electrodes



Figure 6. Effect of electrodes in the pH of anolyte and catholyte

The high electroosmotic flow of 256 mL was observed at cathode reservoir for Fe-Gr electrodes and these electrodes only removed 93% EC from the soil in 3 h, whereas Fe-Fe, Fe-SS, and Fe-Al removed 81% in 4 h, 69% in 5 h and 52% in 6 h respectively (Figure 5). However the remediation of soil was monitored for 8 h and the pH at anolyte, catholyte were reported at every 30 min as presented in Figure 6. In detail, Fe-Gr electrodes removed 97% of Cl, Na and 88% of SO₄ as presented in Figure 7. 88-89% SO₄, and more than 97% Na and Cl removal using Fe-Gr electrodes from saline soils were reported earlier by Lee *et al.* (2012).

The electroosmotic movement of reactive textile dyes towards cathode was observed and Fe-Gr electrodes removed 82% of TOC from soil whereas Fe-Fe, Fe-SS, and Fe-Al removed 63%, 44%, and 31% respectively, as presented in Figure 8. Similar observation of anionic dye movement towards cathode was reported by Lee *et al.* (2009) though boron doped diamond anode and NaCl electrolyte was utilized. This confirms that the Fe-Gr electrodes demonstrated excellent removal of EC and TOC from soil. For the electrode distance of 3 cm of R1 packed with contaminated soil, tap water as electrolytes and Fe-Gr electrodes were utilized to optimize the electrical potential gradient. The experiment was conducted at 3V/cm, 4V/cm and 5V/cm for 3 h and the respective removal of Cl, SO4, Na, EC and TOC from the soil is presented in Figure 9. In order to achieve the maximum removal from the soil, the experiment was conducted for 8 h and the respective removal was observed for 3V/cm in 8 h and 4V/cm in 5 h. Higher the electrical potential gradient, quicker was the removal of contaminants from the soil. To optimize the electrode distance the reactors R1, R2 and R3, tap water as electrolyte and Fe-Gr electrodes, were utilized, and the experiment was conducted at 3V/cm for 10 h. The pH of anolyte and catholyte was monitored for every 1 h during the experiment, (Figure 10).



Figure 7. Major cation and anion removal using different electrodes



Figure 8. EC and TOC removal using different electrodes

The acidic pH at anode reservoir prevented the corrosion of Fe electrode. The soil was sliced into 2.5 cm width

along the length and analyzed for physicochemical properties after 10 h of experiment. The reduction of EC, TOC, Cl, SO₄ and Na in the soil along normalized distance from anode for reactors R1, R2, R3 is presented in Figures 11, 12 and 13. From the plots, it is obvious that minimum the electrode distance, maximum the removal of inorganic salts and textile dyes from the soil. From the results, it is found that the reactors R1, R2 and R3 removed EC and TOC from the soil in 6 h, 7h and 8 h respectively at 3V/cm. However, the performance of R1 and R2 showed better performance than R3 and the removal efficiency is shown in Figure 14. This is due to that the minimum the electrode distance increased the contaminant removal from the soil. The characteristics of anolyte and catholyte in the reactor R1, R2, R3 after remediation experiment are also presented in Figure 15. This confirms that the catholyte contains higher TDS due to the presence of residual reactive textile dyes, those consists of inorganic ions in chemical structure and as well, the large amount of Na, and other minor cations K, Ca, and Mg.



Figure 9. Effect of electrical potential gradient on soil remediation



Figure 10. Effect of electrode distance in anolyte and catholyte pH



Normalized Distance from Anode

Figure 11. Soil characteristics in the reactor R1 after remediation



Normalized Distance from Anode





Figure 13. Soil characteristics in the reactor R3 after remediation This demonstrated that the Cl and SO_4 moved towards anode and Na moved towards cathode. The reactors R1, R2, and R3 removed 96%, 91% and 74% of Cl, 86%, 76% and 48% of SO_4 , and 96%, 91% and 80% of Na respectively. Similar trend of observation on Cl, SO_4 and Na removal was reported on the remediation of saline soil by Lee *et al.* (2012) and Kim *et al.* (2012). Reactors R1, R2, and R3 removed 82%, 77% and 55% of TOC from the textile wastewater contaminated soil. This confirms that the movement of residual textile dyes towards cathode and it was observed by the color change in the cathode reservoir as presented in Figure 16. Similar inference was reported by Pazos *et al.* (2008) and Lee *et al.* (2009) earlier for the removal of Lissamine Green B and Acid Blue 25 dye respectively. This is due to the dominant mechanism of electroosmosis flow over electromigration; however the movement of dye towards anode was reported by Paiva *et al.* (2018) and Annamalai *et al.* (2018) but it lacks enough justification. The detailed investigations on electrokinetics demonstrated the effective removal of TOC and EC using Fe-Gr electrodes, at minimum electrode distance of 3 cm and in the presence of electrical potential gradient of 5V/cm.



Figure 14. Effect of electrode distance in electrokinetic remediation



Figure 15. Characteristics of anolyte and catholyte after electrokinetic remediation



Figure 16. Electroosmosis of residual textile dyes

4. Conclusion

An electroosmotic flow of 256 mL was observed at cathode reservoir for Fe-Gr electrodes and these electrodes removed 93% EC from the soil in 3 h, whereas Fe-Fe, Fe-SS, and Fe-Al removed 81% in 4 h, 69% in 5 h and 52% in 6 h respectively. In detail, Fe-Gr electrodes removed 97% of Cl, Na and 88% of SO₄. As well Fe-Gr removed 82% of TOC from soil whereas Fe-Fe, Fe-SS, and Fe-Al removed 63%, 44%, and 31% respectively. The reactors R1, R2, and R3 removed 96%, 91% and 74% of Cl, 86%, 76% and 48% of SO₄, 96%, 91% and 80% of Na and 82%, 77% and 55% of TOC from the textile wastewater contaminated soil. The detailed investigations on electrokinetic remediation of the textile wastewater contaminated soil demonstrated the effective removal of TOC and EC using Fe-Gr electrodes, at minimum electrode distance of 3 cm and in the presence of electrical potential gradient of 5V/cm. However the reactor R1 remediated only 100 g of soil but R2 of 5.5 cm electrode distance demonstrated similar performance by remediating 200 g of soil. It is found that electroosmosis is the dominant mechanism by which the residual textile dyes were removed and electromigration plays an important role in movement of EC from the soil towards electrodes. Thus the textile contaminated soil is remediated using environmental economic and sustainable in-situ technique.

Conflict of Interest

The authors have declared no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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