

Removal of Various Pollutants from Leachate Using a Low-Cost Technique: Integration of Electrolysis with Activated Carbon Contactor

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Abstract Landfill leachate contains a high concentration of organic pollutants that are active agents in water pollution. This study was conducted to remove various pollutants from landfill leachate through electrolysis and activated carbon (AC) treatments. A simple electrolytic reactor was designed to investigate the removal efficiency of these treatments for biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSSs), and total dissolved solids (TDSs) from landfill leachate at different electric current densities (CDs) and retention times (RTs). The results showed that the highest removal efficiencies for BOD and COD were 75.6 and 57 %, respectively, under a 7-V

current for 4 h. It was also found that BOD, COD, TSS, and TDS removal efficiencies improved in proportion to an increase in CD and RT. However, pH gradually increased with an increase in CD and RT. A number of treated leachate samples were further polished by AC filtration to compare the effect of this additional process on the removal of color, BOD, COD, TSS, and TDS. This secondary treatment resulted in a higher removal of color and other pollutants than electrolysis alone. At 4 h RT, the BOD removal efficiency was 54.6 % at 3 V and 66.4 % at 5 V, and the efficiency increased to 61.5 and 70.5 %, respectively, after treatment by AC filtration. Under the same conditions, COD removal efficiency increased from 7.5 to 38.5 % at 3 V and from 31.1 to 49.5 % at 5 V. TSS and TDS removal efficiencies were also significantly improved by AC filtration. It is therefore concluded that 7 V of CD and 4 h of RT are the optimum parameters for removing pollutants from leachate and that the secondary treatment of AC filtration is an efficient method of further polishing.

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1 Introduction

The modern world produces a large volume of wastewater including municipal, industrial, commercial, and institutional wastewater. Nowadays, the electrocoagulation process (ECP) attracts a great attention in treating

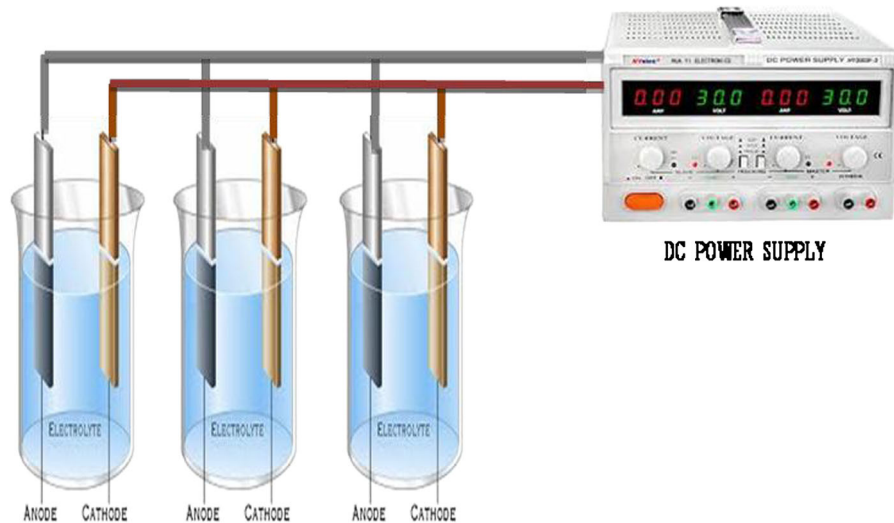


Fig. 1 Schematic diagram of electrolysis process

industrial wastewaters because of its versatility and environmental compatibility. This method is characterized by easy operation, absence of equipment for adding chemicals, shortened retention period, simple low-cost equipment, and minor amount of precipitate/sludge which sediments rapidly (Kobyta et al. 2006). The process is considered as an effective and reliable technology that provides an environmentally compatible method for reducing a large variety of pollutants (Chen 2004; Mollah et al. 2001; Rajeshwar et al. 1994). Moreover, the salt content of the liquid during ECP does not increase appreciably as in the case of chemical treatment (Mollah et al. 2001).

The traditional wastewater treatment methods such as chemical precipitation, filtration, air stripping, and microwave irradiation are expensive. This can be attributed to their high operational cost due to the use of expensive equipments (Bonmati and Flotats 2003; Jeong et al. 2006; Kim et al. 2006; Sanz et al. 2003). Wastewater treatment by ECP is a modern treatment that removes the pollutants through two processes; electrocoagulation and electroflotation (Holt et al., 2005). It is a fast process and needs simple equipments. Recently, electrolysis has been effectively applied to treat various types of wastewater such as poultry wastewater (Kobyta et al. 2006; Yetilmezsoy et al. 2009), cattle slaughterhouse wastewater (Un et al. 2009), latex wastewater (Vijayaraghavan et al. 2008), tannery wastewater (Costa and Olivi 2009), textile wastewater (Bayramoglu et al. 2004; Can et al. 2003; Daneshvar et al. 2006; Kobyta et al. 2003; Lin and Chen 1997; Lin

and Peng 1994), urban wastewater (Pouet and Grasmick 1995; Rahman et al. 2014; Rizvi et al. 2013), tar sand and oil shale wastewater (Renk 1988), chemical fiber plant wastewater (Lin and Lin 1998), laundry wastewater (Wang et al. 2009), and industrial wastewater (Mahmoud and Hoadley 2012). It was also successfully tested to treat various food industry wastewaters such as yeast wastewater (Khristoskova 1984), olive oil wastewater (Adhoum and Monser 2004; Inan et al. 2004), restaurant wastewater (Chen et al. 2000a, b), egg process wastewater (Xu et al. 2002), and oily wastewater (Calvo et al. 2003; Chen et al. 2002; Ibanez et al. 1995). Zaleschi et al. (2014) studied on xanthene dye removal from aqueous solutions by electrocoagulation. The electrochemical treatment was successfully tested using

Table 1 Characteristics of raw leachate and standard effluent quality

Parameters	Mean values	Malaysian standard ^a
pH	7.35	6–9
COD (mg/l)	1168.96	400
DO _{Initial} (mg/l)	8.41	7–9
BOD (mg/l)	657.55	20
BOD/COD ratio	0.56	–
TSS (mg/l)	44.38	0
TDS (mg/l)	938.17	50

^a Sewage and Industrial Effluents, 1979

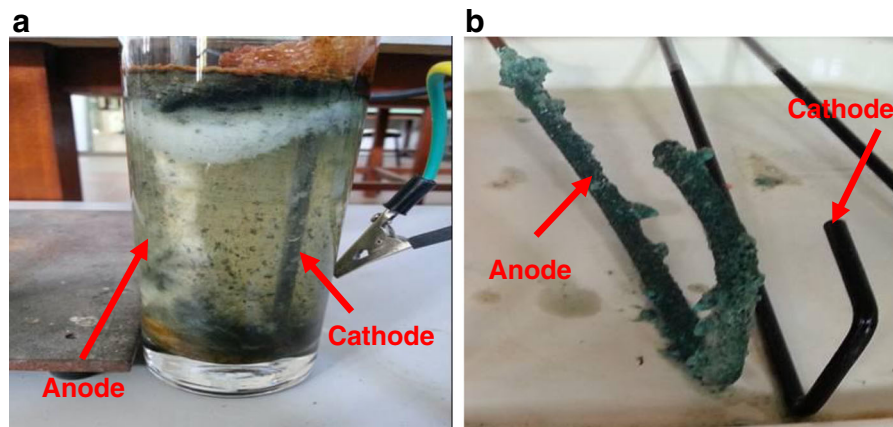


Fig. 2 Reactions at anode and cathode

solutions containing a mixture of phenol and formaldehyde simulating an industrial effluent (Fornazari et al. 2012) and using effluent from the pharmaceutical industry (Domínguez et al. 2012).

Besides, the ECP can be applied for the treatment of landfill leachate (Peng 2013; Tsai et al. 1997). Ilhan et al. (2008) investigated on landfill leachate treatment by ECP using aluminium and iron electrodes and observed that aluminium had more chemical oxygen demand (COD) removal (56 %) than iron electrode (35 %). It had higher performance than classical chemical coagulation process and it can be applied as a step of a joint treatment. Öztürk et al. (2013) studied the effect of seawater conductivity on the treatment of domestic and hazardous solid waste leachate by ECP. They concluded that adding seawater accelerated the process due to the increase of conductivity and

the presence of other ions in its composition. For suspended solids and sulphate ions, the removal efficiencies varied in the range of 50–70 %.

Kabuk et al. (2013) investigated on leachate treatment with ECP and optimization by response surface methodology. At optimum working conditions, 60.5 % COD removal, 92.4 % total suspended solids (TSSs) removal, 60.8 % total organic carbon (TOC) removal, 28.3 % total Kjeldahl nitrogen (TKN) removal, 99 % PO₄-P removal, and 28.9 % NH₃-N removal results were obtained. Fernandes et al. (2014) applied a combined ECP and anodic oxidation (AO) in order to improve the biodegradability of landfill leachate. During ECP, chromium was almost completely removed and zinc was partially removed; the remainder of the zinc was removed during AO. The concentration of iron increased during ECP and decreased during AO.

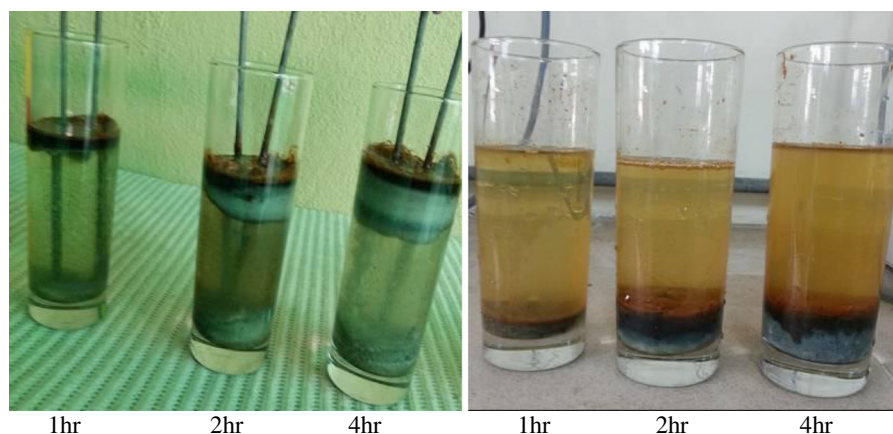


Fig. 3 Depositions of pollutants for electrolysis

Table 2 Deposition of pollutants on electrodes at 5 V of electric current

Retention time (h)	Deposition thickness (mm)
1	8
2	15
4	20

Rada et al. (2013) observed that electrochemical oxidation can remove 64–70 % of COD and 15–61 % of ammonium from landfill leachate. In order to achieve the discharging limits for COD and particularly for nitrogen, electrooxidation only is not sufficient, and other treatments are needed. They suggested applying electrooxidation in combination with hydrogen peroxide (electro-Fenton) when the main objective is to reduce the COD concentration under the discharging limit.

Therefore, in this study, a novel low-cost process integrating electrocoagulation with an activated carbon (AC) contactor is developed for the first time to improve the treatment of the increasing volume of leachate. The optimum pollutant removal efficiencies (for BOD₅, COD, TDS, TSS, and pH) are identified by extensive laboratory analysis. The proposed process is an eco-friendly, sustainable technique for leachate treatment, which reduces treatment cost and saves energy, and which also helps in protecting the environment.

2 Materials and Methods

2.1 Collection of Raw Leachate

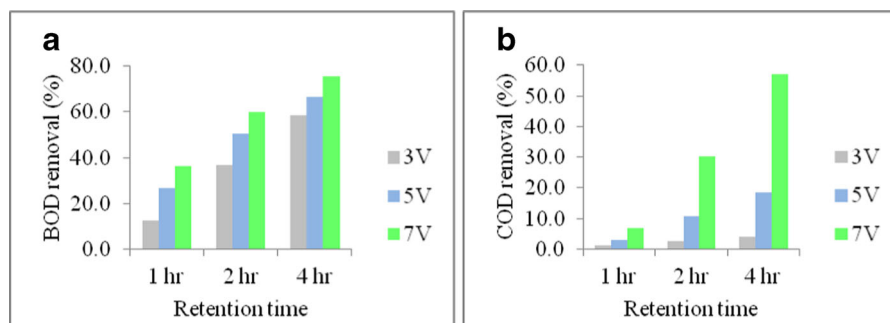
A number of raw leachate samples were collected from the Jeram Sanitary Landfill, Kuala Selangor, Malaysia.

The samples were taken from the ultimate discharge point where the leachate is collected by three main pipelines from the active phase (i.e., currently in operation). Each of the samples was collected in a 2-L bottle, transferred to the laboratory, and stored at 4 °C. Sampling continued for 5 months between October, 2012 and March, 2013.

2.2 Experimental Setup for Electrolysis and AC Filtration

A cylindrical glass reactor was designed to optimize the efficiency of the removal of pollutants from the leachate. The parameters studied were (i) different current densities (CDs) of 3, 5, and 7 V and (ii) different retention times (RTs) of 1, 2, and 4 h, without and with AC filtration to investigate their effects on the removal of various pollutants. The reactor measured 15 cm in height and 4.5 cm in diameter and had a capacity of 240 mL. A pair of electrodes made of iron was installed as the anode and cathode. Each electrode had a diameter of 3 mm and an effective surface area of 188 cm². The two electrodes were arranged in parallel with an acrylic band with 1-cm distance between them to obtain an efficient electric field. The electrodes were connected to a digital direct current (DC) power supply (Model: Zhaoxin RXN 3010 DC 12 V 30 A) that regulated the electricity. For one cycle of the electrolysis process, three cylindrical glasses were set up together in a parallel arrangement to ensure that each glass could receive the same voltage (Fig. 1). A glass of sample was removed after the stipulated RT of 1, 2, and 4 h, respectively.

After the raw leachate had been treated by the electrolysis process, the effluent was passed through a secondary treatment of AC filtration by using a laboratory

**Fig. 4** BOD and COD removal efficiency by electrolysis

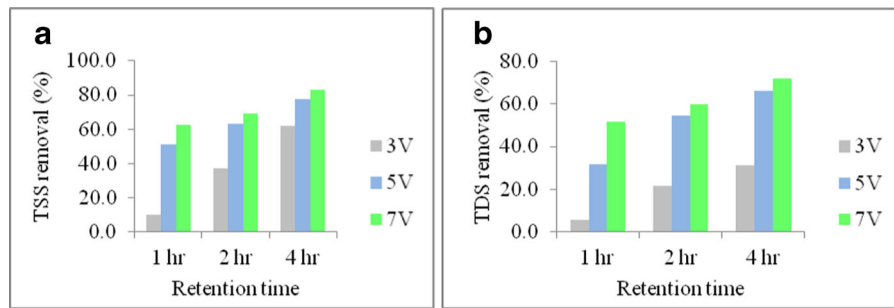


Fig. 5 TSS and TDS removal efficiency by electrolysis

plastic column filled with AC. The removal efficiency of the various parameters was investigated without and with this secondary treatment. The AC-filtrated leachate was prepared by sieving the leachate from the plastic column through fabric and pouring it into a beaker after the adsorption treatment.

2.3 Laboratory Analysis

The parameters studied were pH, biochemical oxygen demand (BOD), COD, TSS, and total dissolved solids (TDSs) according to the Standard Methods for the Examination of Water and Wastewater (APHA, 1998). Each test was repeated at least three times to confirm the accuracy of the experimental data. The parameters were measured and analyzed for raw leachate, after electrolysis and after AC filtration, so that the removal efficiencies could be compared for each stage.

3 Results and Discussion

Leachate that contains a high level of BOD and COD indicates the presence of high organic load. The

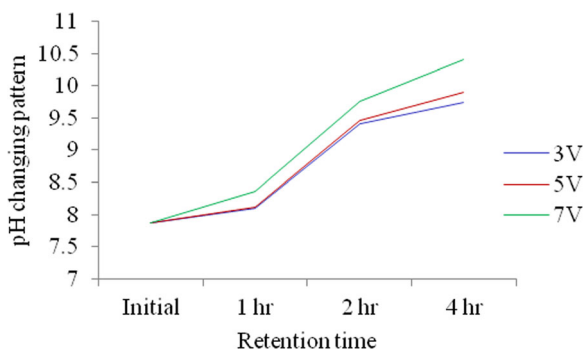


Fig. 6 pH increases due to electrolysis

collected raw leachate had a black color and an offensive odor. Table 1 shows the raw leachate characteristics alongside the standard effluent quality requirements for discharge into a watercourse as specified by the Effluent Quality (sewage and industrial effluents) Regulations, 1979 in the Environmental Quality Act, 1974 and enforced by the Department of Environment, Malaysia.

3.1 Effect of Electrolysis on Removal of BOD and COD

A huge number of bubbles were observed in the reactor during the electrolytic processing of the leachate (Fig. 2). A lot of organic materials in the leachate were oxidized and coagulated electrochemically and electroflotation occurred through the bubbles produced during electrolysis. The coagulated pollutants settled at the bottom of the reactor after stopping the process. The highest amount of sediment (coagulated pollutants) was observed at 4 h RT (Fig. 3). The highest deposition thickness of pollutants (20 mm) on the anode was also observed at 4 h RT (Table 2).

Results showed that BOD removal efficiency increased with increasing CD and RT (Fig. 4a). The

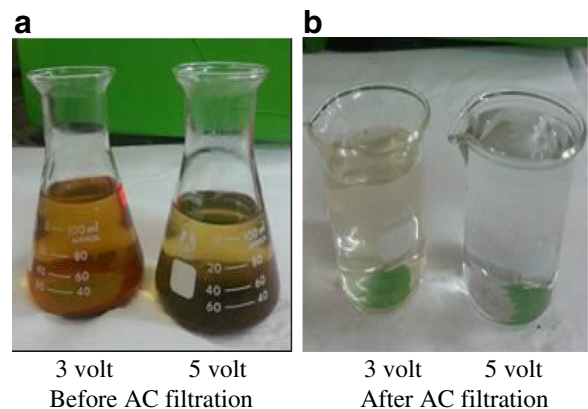


Fig. 7 Filtration of treated leachate through activated carbon

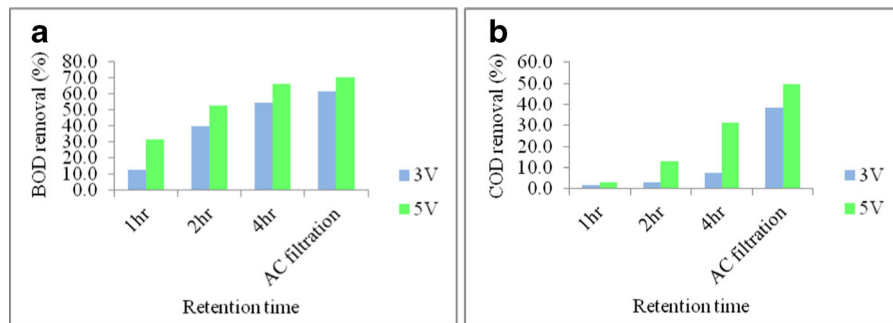


Fig. 8 Effect of AC filtration on BOD and COD removals

highest BOD removal efficiency (76 %) occurred at 7 V and 4 h RT while the lowest happened at 3 V and 1 h RT. The BOD removal efficiency was found to be proportional to CD and RT. Similarly, the highest COD removal (57 %) was observed at 7 V and 4 h RT and COD removal was also found to be proportional to CD and RT. Figure 4b shows the COD removal efficiency at different CD and RT. Kobya et al. (2006) state that an increase in CD results in an increase in the generation of ions and flocs, which increases the efficiency of COD removal. Yetilmeszooy et al. (2009) state that COD and color removal efficiencies are increased with increasing CD. Chen (2004) states that CD determines the production rate of coagulant and bubbles, which affects the growth of flocs. Cho et al. (2010) found that the highest percentage of removal of NH_4 (97 %), PO_4 (82 %), and color (83 %) from swine wastewater occurred at 7 V. Wang et al. (2009) studied the removal of COD from laundry wastewater at 3, 5, and 7 V and found that the highest removal efficiency occurred at 7 V. They also found that removal efficiency dramatically increases with increasing voltage, which is very similar to the findings of this study. It can therefore be concluded that

7 V could be the optimum CD for pollutant removal from landfill leachate.

The effect of CD or electrical potential (voltage) on the percentage COD and BOD removed from leachate is the most important parameter to consider when attempting to improve the efficiency of the electrodes in this process. According to Faraday's law, as CD increases, the efficiency of the ions released from the respective electrodes also increases. In addition, CD not only determines the coagulant dosage rate, but also the bubble production rate as well as the size and growth of the deposition, hence CD affects the overall efficiency and operating costs of the process.

3.2 Effect of Electrolysis on Removal of TSS and TDS

The TSS and TDS removal efficiencies were also investigated. The results showed that lower CD and RT resulted in less TSS and TDS removal (Fig. 5a). The highest TSS removal percentage (83 %) occurred at 7 V and 4 h RT and likewise the highest TDS removal (72 %) also occurred at 7 V and 4 h RT (Fig. 5b). The

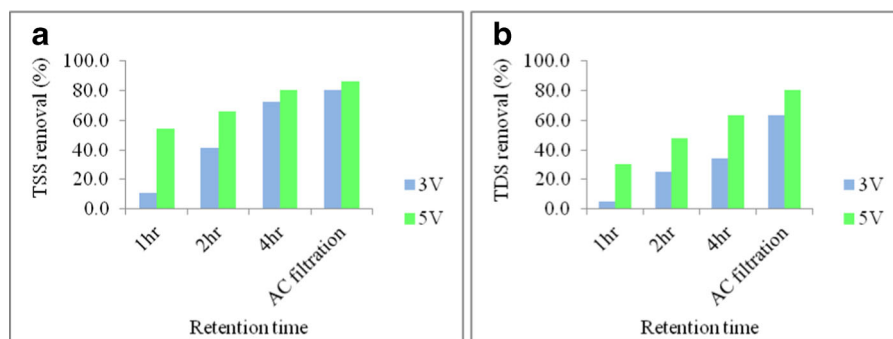


Fig. 9 Effect of AC filtration on TSS and TDS removals

TSS and TDS removal efficiencies were found to be proportional to CD and RT.

3.3 Effect on pH

The average initial pH of the raw leachate was 7.88 (i.e., slightly alkaline). The results showed that alkalinity increased with increasing CD and RT (Fig. 6). In fact, acidity or alkalinity depends on the presence of hydrogen or the hydroxyl ion concentration of a solution. A huge amount of hydrogen ions was removed during electrolysis through H_2 gas and produced a lot of OH^- ions through the electrodes, which could have been the cause of the alkalinity in all the samples. At 7 V and 4 h RT, the pH increased from 7.88 to 10.41. Note that the initial pH value was high in this experiment. Kobya et al. (2003) found that the highest removal efficiencies were achieved when the initial pH was below 8. It can therefore be suggested that the electrolysis process could be performed after modification of the initial pH (to below 8).

3.4 Effect of AC Filtration on Electrolyzed Landfill Leachate

Filtration through AC was the secondary treatment applied after the primary process (electrolysis). The results revealed that AC filtration had an effect on the removal of pollutants after electrolysis. The leachate after AC filtration was almost clean and much clearer than that of after primary treatment only (Fig. 7). Figures 8 and 9 show that AC filtration greatly affected the removal efficiencies of BOD, COD, TSS, and TDS. At 4 h RT, BOD removal was 54.6 % at 3 V and 66.4 % at 5 V, and these efficiencies increased to 61.5 and 70.5 %, respectively, after AC filtration. Under the same conditions, COD removal increased from 7.5 to 38.5 % at 3 V and from 31.1 to 49.5 % at 5 V. Similarly, the increase in TSS removal efficiency was also notable, increasing from 72.7 to 80.1 % at 3 V and from 80.1 to 86 % at 5 V at 4 h RT. A significant improvement was seen in TDS removal efficiency, which was 34.3 % at 3 V and 63.4 % at 5 V at 4 h RT and increased to 63.4 and 80.7 %, respectively, after AC filtration. Figure 10 shows the effect of voltage on BOD and COD removals. The highest BOD and COD removal efficiencies (83 and 97 %, respectively) were achieved by applying 15 V of current density.

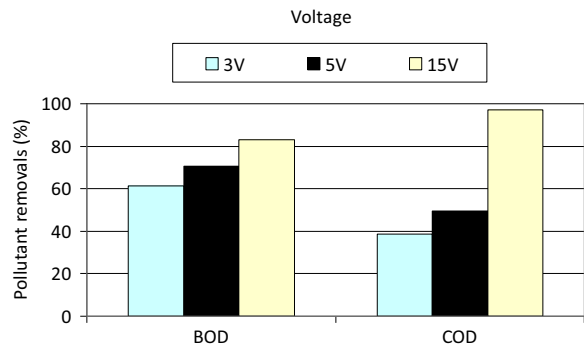


Fig. 10 Effect of voltage on BOD and COD removals

AC filtration helps to reduce the total amount of contaminants present in effluent. Activated carbon is a porous material that removes organic compounds from liquids through adsorption. In adsorption, the organic molecules contained in leachate are attracted and bound to the surface of the pores of the AC. Due to its high degree of microporosity, AC can easily adsorb pollutants from leachate and consequently can achieve higher removal efficiencies. However, it should be noted that the overall performance of an adsorption treatment process depends on the chemical composition and concentration of the contaminants.

4 Conclusions

Electrolysis is an effective way to remove pollutants from landfill leachate. A removal efficiency of 76, 57, 84, and 72 % was achieved for BOD, COD, TSS, and TDS, respectively, after primary treatment by electrochemical oxidation at a 7 V current density (CD) and 4 h of retention time (RT). The results show that the quality of the treated leachate after activated carbon (AC) filtration significantly improved in terms of suspended solids, color, and odor. At 4 h RT, the BOD removal efficiency was 54.6 % at 3 V and 66.4 % at 5 V, and these percentages increased to 61.5 and 70.5 %, respectively, after secondary treatment by AC filtration. Under the same conditions, COD removal efficiency also increased from 7.5 % at 3 V and 31.1 % at 5 V to 38.5 and 49.5 %, respectively. Similarly, the improvement in the proportion of TSS removal was also notable, increasing from 72.7 % at 3 V and 80.1 % at 5 V at 4 h RT to 80.1 and 86 %, respectively, after AC filtration. Likewise, TDS removal efficiency was improved significantly from 34.3 % at 3 V and 63.4 % at 5 V at 4 h RT to 63.4 and 80.7 %, respectively. This process is a low-cost

treatment which is environmentally friendly. It has no adverse effects and does not form any new toxic byproducts. It can be applied when pretreating domestic and industrial wastewater containing concentrated organic contaminants.

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