

Carbon Electrode and Buleleng Seawater in Degrading the Leachate in Bengkala Village of Buleleng Regency by Using Electro-Oxidation Techniques

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ABSTRACT. This research aims to study the effectiveness of using carbon electrodes and Buleleng seawater in degrading leachate through electro-oxidation techniques. Electro-oxidation is a waste processing technology that cannot be biologically degraded, has high efficiency and degradation speed, is easy to operate, has low cost, and is environmentally friendly. Leachate is taken from the dumping ground in Bengkala village of Buleleng Regency. The volume of leachate used was 50 mL. The influence of the surface area of the electrode, the distance between electrodes, voltage, contact time, and the amount of seawater were also studied. All treated leachates were then measured for COD, DO, absorbance, BOD, and pH. The results showed that Buleleng sea water contains chlorine ions 16.862 g/L. COD, DO, absorbance, BOD, and pH of leachate before electro-oxidation 38.763x10³, 0, 17.1, 0, and 9 mg/L respectively. The surface area of the electrodes, the distance between electrodes, voltage, contact time, and the amount of seawater used were most effective in degrading leachate in 10.551 cm², 3 cm, 13.5 volts, 1.5 hours, and 560 mL respectively.

Keywords: Buleleng seawater, carbon electrodes, degrading leachate, electro-oxidation technique

INTRODUCTION

Leachate is the liquid produced from a dumping ground. Leachate contains toxic organic compounds, ammonia, heavy metals, xenobiotics, and inorganic salts (Keyikoglu et al., 2021); (Siddiqua et al., 2022). The organic compounds such as dichloromethane; 1,2 dichloroethane, trichloroethane, tetrachloroethane, toluene chlorophenol, aldrin, lindane, and PCBs found in leachate are hazardous to human health. Other heavy metals carried in the leachate are Cu, Cd, Cr, Fe, and Pb (Mojiri et al., 2021).

Various attempts have been done to degrade leachate, including flocculation/coagulation (Paula Floriano Santos et al., 2022), activated adsorption (Peres et al., 2018), microbial treatment (Quraishi et al., 2019), fenton oxidation (Göde et al., 2019a), advanced oxidation (Samarghandi et al., 2018) (Moreira et al., 2015), microalgae usage (Hernández-García et al., 2019), biogeochemical process (Zhou et al., 2016), continuous-flow catalytic ozonation (De Brito et al., 2019). These efforts are not very acceptable since they cannot degrade old leachate and are ineffective in degrading organic acids and ammonium. Therefore, pretreatment is required for

adsorbent and large analyte derivatives (Göde et al., 2019b); (Anqi et al., 2020).

Electro-oxidation is a simple technique to degrade leachate. Efficiency electro-oxidation techniques depend on factors such as the electrode material type, electro-oxidation time, pH, and concentration of the electrolyte solution. This technique has some advantages such as being easy to implement, simple procedure, and being able to degrade more comprehensive leachate. Electrode and electrolyte solutions are essential parts of an electro-oxidation technique. Electrode and electrolyte media often used in engineering electro-oxidation are Pt electrodes in a NaCl electrolyte solution, PbO₂/Ti, SnO₂/Ti, Fe, and Al electrodes in a solution of Na₂SO₄ (Xiaocai Yu et al., 2019); (Xia et al., 2019); (Aggadi et al., 2021). The use of such electrodes and media demands high cost. This obstacle can be overcome by using carbon electrodes and seawater media.

Seawater consists of 96.5% of all known water on the Earth and covers 71% of the Earth's surface. The concentration of major cations in surface seawater is sodium (Na⁺). The major anion in surface seawater is chloride (Cl⁻). Chloride anions became essential for environmental monitoring and directly led to metal corrosion. It was a major ongoing challenge in

shipbuilding and maritime transport. Anions chloride in seawater has three main industrial uses: disinfection of water and installations, control of biofouling, and preventing the transport of aquatic invasive species. Once in contact with seawater, chlorine reacts rapidly with water constituents (e.g. bromide ions, ammonia, and nitrogen-containing compounds) to form a range of oxidative species (e.g. bromine and N-haloamines), termed "chlorine-produced oxidants" (CPOs) or "total residual oxidants" (TRO). The chemical nature of CPOs and their concentration are a function of two categories of parameters related to treatment modality (e.g. chlorine dose) and water quality (e.g. temperature, pH, ammonia concentration, and organic constituents) (Chen & Compton, 2023). On the other hand, chlorine anion is very useful for the treatment of water pollution (Yao et al., 2021); (Samarghandi et al., 2020).

Based on the research studies, leachate in the Bengkala Village of Buleleng Regency is classified as mature and is dominated by organic compounds. Its maturity is based on a comparison of leachate BOD/COD, which is 0. The characteristics of leachate in Bengkala village are: black, muddy, smelly, and itchy. Such leachate problems trouble the inhabitants living close to the dumping ground. This concern is rapidly increasing for the ground is higher than its surroundings. The inhabitants are worried that leachate might contaminate their water sources. To anticipate such matters, a comprehensive, cheap, and easy treatment needs implemented. The possible treatment to anticipate the arising problem is by applying an electro-oxidation technique, i.e. using carbon electrodes and seawater.

Seawater such as Buleleng seawater and carbon electrodes have not been used to treat water pollution like leachate. This paper studies the effect of the electrode surface area, the distance between electrodes, voltage, contact time, and the amount of seawater to degrade Bengkala leachate. Thus, the effectiveness of carbon electrodes and the volume of seawater were studied to degrade leachate.

EXPERIMENTAL SECTION

Materials

The materials used are $K_2C_2O_7$ (Merck, 99.9%), H_2SO_4 (Merck, 98%), $HgSO_4$ (Merck, 98%), water distillation, Ag_2SO_4 (Merck, 98%), $C_8H_5KO_4$ (Merck, 98%), carbon electrode (graphite 90 %). All materials are pro analysis grades. Whatman 42 filter papers were used during the experiment. At the same time, other materials used were leachate and Buleleng seawater. Leachate was collected from the Bengkala dumping ground and the seawater was taken from the Buleleng sea.

Instrumentations

The instruments used were glass apparatus, magnetic stirrer, analytical balance (Mettler Toledo AB54S), pH meter (Hanna H198103), magnetic stirrer

plate (Thermolyne Cimarec 1), COD reactor (Hanna instruments HI-839800 25), double-beam UV-VIS (spectrophotometer UV-1800) and, DO Meter (Hanna HI9146).

Sampling techniques of leachate and Buleleng seawater

Leachate samples were collected from the leachate retention pond at the Bengkala Landfill, Kubutambahan District, Buleleng Regency, Bali Province. The sampling and storage techniques were based on SNI 6989.59:2008 (Badan Standarisasi Nasional, 2008). The steps for leachate sampling were as follows: leachate was collected at several points and at different depths, each containing 2 litres. The leachate samples were mixed to ensure homogeneity and filtered.

The Buleleng sea is mostly fed by a river, so the salinity of it varies. Estuary waters were a sample in this research. The method of sampling Buleleng seawater refers to SNI 6964.8:2015 (Badan Standarisasi Nasional, 2015). Seawater samples were taken during high tide and low tide with oligohaline salinity zones (0.5-5 psu), mesohaline salinity zones (5-18 psu), and polyhaline salinity zones (18-30 psu). Each zone was taken 2.5 litres, then mixed. Buleleng seawater was filtered, and its chlorine content was determined.

Identifying COD, DO, absorbance, pH, and BOD in leachate

COD was determined by the closed reflux method and measured with a Shimadzu UV-vis double beam at $\lambda = 600$ nm. The absorbance was measured with a Shimadzu UV-vis double beam at a wavelength (λ) 511 nm. DO was measured with a metric DO and pH was measured by pH metric. BOD determination was done by using the following equation:

$$BOD_5 = [(DO_{to} - DO_{t5}) - (DO_{blanko-to} - DO_{blanko-t5})]P \quad (1)$$

In this case, the DO measurement $DO_{to} = 0$ day incubation sample bottles, $DO_{t5} = DO$ at 5 days of incubation measurement sample bottles, $DO_{blanko-to} = DO$ on measuring 0 day incubation bottles blank, $DO_{blanko-t5} = DO$ at 5 days of incubation bottles measurement blank, and $P =$ the dilution factor.

Identifying the effectiveness of carbon electrodes

The effectiveness of electrodes in wastewater treatment is greatly influenced by several factors. These factors include: anode surface area (Pierpaoli et al., 2021), the distance between electrodes, voltage variation, and electro-oxidation time (Huang et al., 2024).

Carbon electrodes are tested for effectiveness by varying the surface area of the carbon electrodes, the distance between electrodes, the voltage supplied, and contact time. The most effective surface area was determined by variations of the surface area of the carbon electrodes dipped were respectively 2.620, 5.130, 7.640, and 10.551 cm^2 . The diameter and length the of carbon electrodes used were 0.8 and 5 cm, respectively. Then, the carbon electrodes were

dipped into a mixture of leachate (50 mL) with Buleleng seawater (660 mL). The electrodes are then connected to 13.5 volts for 0.5 hours.

The most effective distance between electrodes was determined by varying the distance between the two electrodes: 3, 6, 9, and 12 cm. The carbon electrodes were dipped into a mixture of leachate (50 mL) with Buleleng seawater (660 mL) and connected to a 13.5 volts voltage source for 0.5 hour with the most effective surface area of electrode.

The most effective voltage was determined based on distance between electrodes and the surface area the most effective. The voltage was varied at 3, 6, 9, and 13.5 volts. The voltage source was passed through the carbon electrodes for 0.5 hour in nto a mixture of leachate (50 mL) with Buleleng seawater (660 mL).

The most effective contact time was achieved under the most effective conditions of electrode surface area, distance between electrodes, and voltage. A total of 50 mL of leachate and 660 mL of seawater were used. The leachate electro-oxidation contact time varied between 0.5, 1, 1.5, and 2 hours.

All leachates treated under various conditions, including varying surface area, distance between electrodes, voltage, and contact time, were then measured for COD, DO, absorbance, BOD, and pH. The most effective conditions for various variations were determined based on COD, DO, absorbance, BOD, and pH values by comparing one data set with another and with existing research findings in journals. The data were then narrated based on supporting theory.

Identifying the effectiveness of the seawater volume

The effectiveness of electrolyte solutions used in wastewater treatment is highly influenced by the number of particles in the solution. The number of NaCl particles has a significant impact on the indirect reaction of chlorine in wastewater treatment (Huang et al., 2024). The effectiveness of the number of NaCl

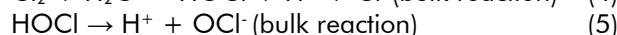
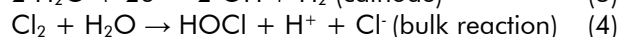
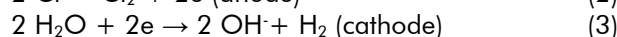
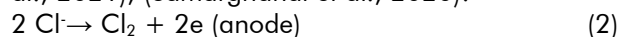
particles can be varied by the amount of NaCl volume at the same concentration, and this method is used to test the effectiveness of the Buleleng seawater.

Testing the effectiveness of the seawater volume was done by varying the amount of the seawater during electro-oxidation. Variations in seawater amounts were 360, 460, 560, and 660 mL. The seawater was then mixed with 50 mL of leachate and electrooxidized at the most effective surface area, electrode distance, voltage, and contact time. The treated leachates were finally tested for COD, DO, absorbance, BOD, and pH.

RESULTS AND DISCUSSION

Chlorine levels in Buleleng seawater

Buleleng sea water contains chlorine ions 16.862 g/L. Chlorine ion is oxidized to chlorine (Cl₂) during the electro-oxidation process at the anode. Chlorine formed at the anode can react with water to form hypochlorite acid (HOCl). Chlorine and hypochlorite acid are a strong oxidizing agent that oxidizes leachate (Agustina, Bagastyo, & Nurhayati, 2019). The reaction in the electro-oxidation process can be written (Yao et al., 2021); (Samarghandi et al., 2020):



The level of COD, DO, absorbance, BOD, and pH leachate before treatment

The levels of COD, DO, absorbance, and pH leachate before treatment can be seen in **Figure 1**. **Figure 1** shows that the levels of COD and absorbance leachate were very high, while the levels of DO and BOD leachate were zero. High COD indicated that much substance was dissolved (pH = 9) and there was much oxygen used in the leachate, so leachate absorbance was very high. Hence, there was no free oxygen in the leachate (DO = 0) and consequently, there were no microorganisms living in the leachate

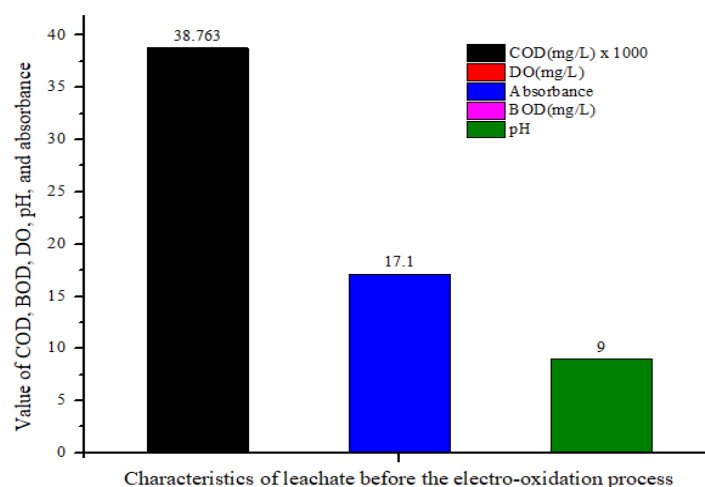


Figure 1. The value of COD, DO, absorbance, BOD, and pH of leachate before the electro-oxidation process

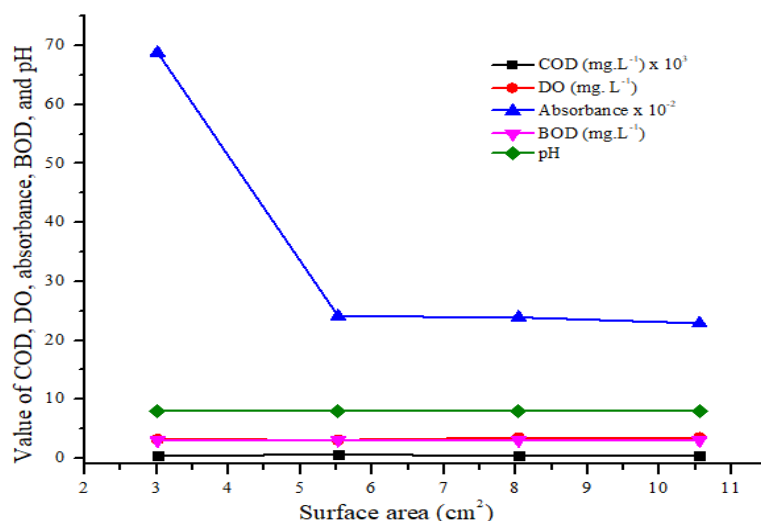


Figure 2. The value of COD, DO, absorbance, BOD, and pH at surface area variation of carbon electrodes

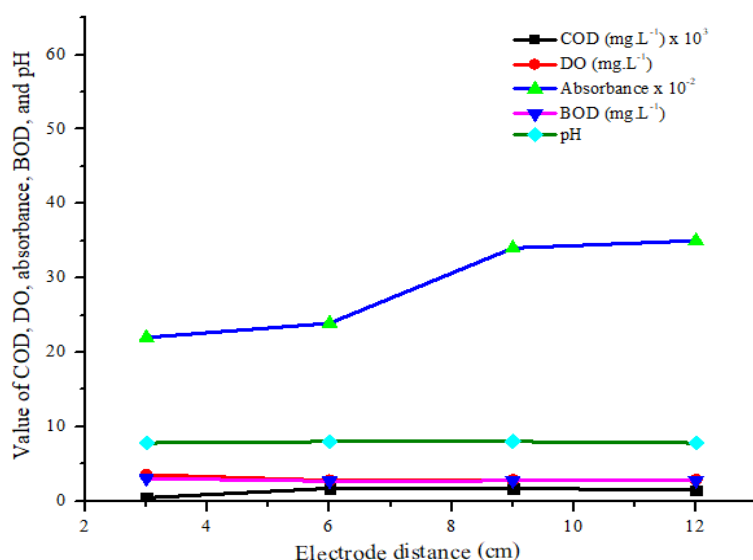


Figure 3. The value of COD, DO, absorbance, BOD, and pH at electrode distance variation of carbon electrodes

(BOD = 0). The BOD/COD value is very low (0), indicating that most of the organics in the leachate are compounds that are difficult to decompose biologically. Aerobic microbes are not able to consume most of these organic compounds, so the BOD is very low, the COD is still high because many organic compounds remain in recalcitrant form such as humic and fulvic compounds as well as aromatic acids and other complex compounds (Lindamulla, Nanayakkara, Othman, Jinadasa, & Herath, 2022). Therefore, when the BOD/COD ratio approaches zero, simple biological treatment is ineffective, so a chemical pretreatment process such as electro-oxidation is needed so that difficult-to-degrade compounds are broken down into more easily biodegradable forms.

The effectiveness of the carbon electrode distance degrading leachate is presented in **Figure 3**. Looking at the figure, the 3 cm carbon electrode distance showed a decrease in the levels of COD and absorbance, but an increase in the levels of DO

and BOD. The shortest distance between electrodes could degrade leachate the most since the number of ions gathered at each electrode was getting closer so that they could easily react with one another. Chlorine (Cl₂) formed in the anode quickly reacted with water collected at the cathode to form hypochlorite acid. As the formation of hypochlorite acids was getting faster, the amount of hypochlorite acid particles were rapidly increasing; henceforth, the leachate oxidized by hypochlorite acid was also increasing. The increasing amount of hypochlorite acids was also capable of decreasing the pH of the leachate from 9 to 8. This impact caused a decrease in the levels of COD and absorbance but in contrast an increase in the levels of DO and BOD. So, based on it, **Figure 3** shows that a carbon electrode distance of 3 cm was the most effective in degrading leachate.

Voltage variation effects on leachate are presented in **Figure 4**. The effects of voltage variation are seen from the DO, COD, absorbance, BOD, and pH in the

leachate after a given treatment. The greater the applied voltage, the less the value of COD and absorbance, but the greater the value of DO and BOD was. The addition of the voltage meant the addition of strong currents in the cell so that the amount of chlorine ions oxidized to chlorine increased (Veli et al., 2021). The increasing amount of chlorine caused a greater number of leachate to degrade. The amount of leachate degraded affects the increase of BOD and DO, but the decrease of absorbance and COD (Antony et al., 2020). Voltage variations do not impact pH values. However, voltage variations of 3-13.5 volts can decrease the pH from 9 to 8. This indicated that the formed hypochlorite acids also reacted with OH⁻ ions in the leachate (Ramprasad et al., 2019). So, based on it, **Figure 4** shows that voltage 13.5 volts was the most effective in degrading leachate.

The effectiveness of contact time is presented in **Figure 5**. The figure showed the value of COD and absorbance decreased, while the DO and BOD increased during the contact time of 0.5-1.5 hours. During those hours, the number of chloride ions oxidized into chloride gas and hypochlorite acids formed was also increased. The more the amount of chlorine and hypochlorite acids were formed, the more the amount of leachate degraded. This caused the levels of COD and absorbance to decrease, while on the contrary the DO and BOD were increasing during those hours.

When the contact time of 1.5-2 hours; COD, absorbance, DO, and BOD were relatively constant. This means that the number of chlorine and hypochlorite acid (HOCl) formed are relatively the same so the leachate degraded also stays the same. These impacts relatively give constant values of COD, absorbance, DO, and BOD.

The effect of contact time on the change of the pH of leachate is that the longer the contact time, the lower the pH of the leachate would be. Leachate acidity changes during the contact time of 0.5-2 hours were 5–3. When it compared with the initial leachate acidity (pH = 9), leachate acidity decreased significantly. The decrease in the pH of the leachate resulted from the increase in the amount of hypochlorite acids (Pieczyńska et al., 2019); (Moreno-Palacios et al., 2019); (Yao et al., 2021). So, based on it, **Figure 5** shows that contact times of 1.5 hours were the most effective in degrading leachate.

The effectiveness of the Buleleng seawater volume

The effectiveness of the Buleleng seawater volume can be examined in **Figure 6**. NaCl concentration increases, it can increase Cl⁻ ion concentration. The presence of chloride ions can increase the concentration of Cl₂, HOCl, and OCl species (Massoudinejad et al., 2015). Increasing the volume of seawater used in the leachate electro-oxidation process can increase the number of Cl⁻ ions, thereby increasing the indirect electro-oxidation reaction by active chlorine (Equation 2) (Moreira et al., 2017). In addition, increasing active chlorine concentration will increase chlorine production and decrease the selectivity for oxygen evolution (Equation 3), thereby increasing the leachate electro-oxidation reaction (Equation 7) (Jager et al., 2017).

The **Figure 6** shows that the addition of 360-560 mL volume of seawater caused a decrease in the values of COD and absorbance, but the DO and BOD values increased. However, when 560-660 mL volume of seawater was added; the levels of COD, absorbance, DO and BOD tended to be stable. This means the amount of 560 mL seawater is the best volume of seawater to degrade leachate since the

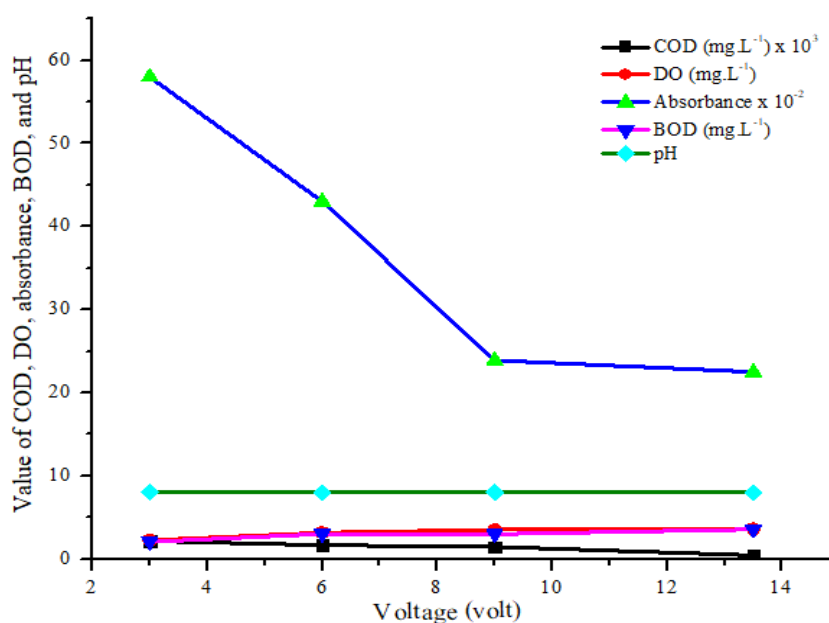


Figure 4. The value of DO, absorbance, BOD, and pH at voltage variation of carbon electrodes

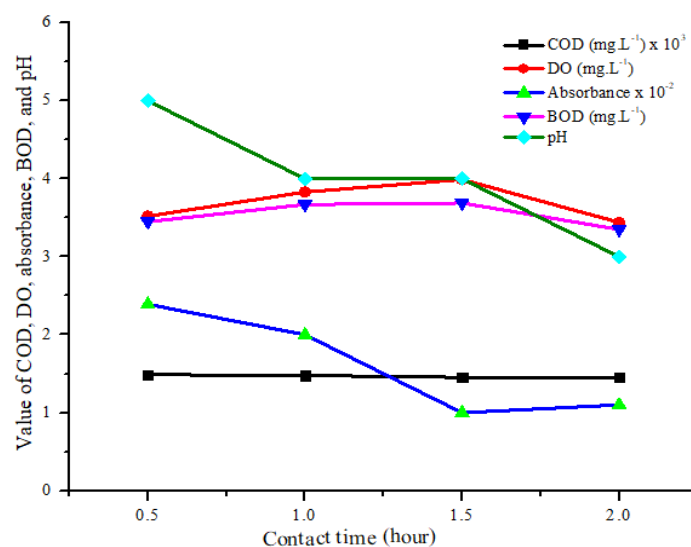


Figure 5. The value of DO, absorbance, BOD, and pH at contact time variation of carbon electrodes

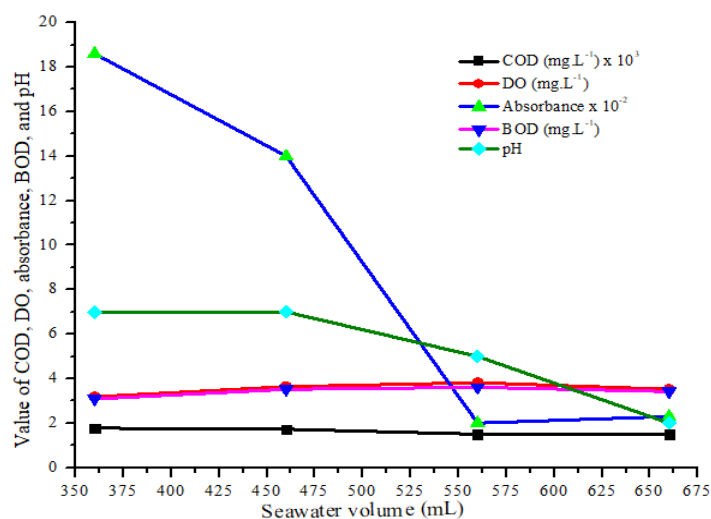


Figure 6. The value of DO, absorbance, BOD, and pH at seawater volume variation of carbon electrodes

amount of chlorine ions oxidized to form chlorine which then reacted with water to form hypochlorite acid is most effective in degrading leachate (Mandal, Dubey, & Gupta, 2017).

Figure 7 shows the spectra of leachate before and after the electrooxidation process. Furthermore, it displays a prominent peak in the visible region (400–800 nm), indicating the presence of the azo chromophore group (–N=N–). Another peak found in the UV region (200–400 nm) represents unsaturated groups, such as benzene, naphthalenes, and heterocyclic rings (Droguett et al., 2020). The decrease of absorbance in the UV region was less pronounced than in the visible region because the aromatic structures of compounds in leachate were more stable than in the azo chromophore group. During the electrooxidation process by using a carbon electrode, there was a decrease in the peak at 400–800 nm, because the chromophore group became an intermediate substance before aliphatic carboxylic acids and

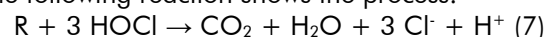
carbon dioxide were formed. This event accelerates the decrease in absorbance and disappearance of the peak at 400–800 nm. The decreasing absorbance in the UV region is lower than in the visible region because the aromatic structure is more stable than the azo chromophore group (Tang et al., 2020).

The effectiveness of the addition of seawater can be viewed from the reactions that occur during electro-oxidation. During electro-oxidation, the organic compounds in the leachate passed two processes, notably: partially and fully oxidized. In partial oxidation, the leachate was not fully oxidized (Veli et al., 2021). The reaction process can be seen as follows (Deng et al., 2020):



Where R: organic compounds; MO_x : anode oxide; MO_{x+1} : oxygen in the oxide lattice.

In the full oxidation, leachate was fully oxidized. The following reaction shows the process.



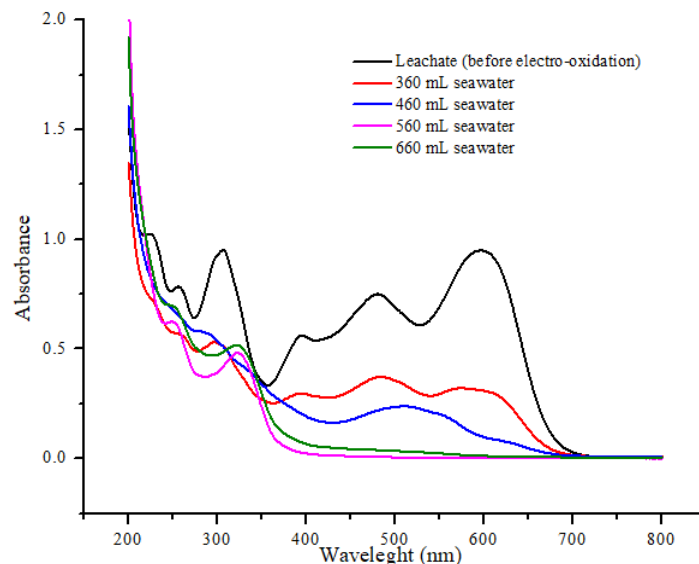


Figure 7. The absorbance spectra of leachate before and after electro-oxidation at seawater volume variation

The impacts of adding seawater volume upon the changes of the seawater leachate acidity showed that the more addition of seawater, the lower the leachate acidity would be. In the addition of 360 – 450 mL seawater, the pH of the leachate showed 8. When the addition of 450-660 mL of seawater, leachate pH decreased, i.e. 8 to 2. This happened because the number of hypochlorite acids formed was increasing and not all those acids reacted with leachate. The excess of hypochlorite acids had effects on the decrease of leachate acidity.

CONCLUSIONS

Based on the research results, it can be concluded that the Buleleng seawater and carbon electrodes can degrade leachate. Buleleng sea water contains chlorine ions 16.862 g/L. The best surface area of the carbon electrode to degrading leachate was 10.551 cm². The effective distance was found 3 cm for degrading leachate. The 13.5 volts was an effective voltage for degrading leachate. The effectiveness of contact time was found in 1.5 hours, and the amount of Buleleng seawater was 560 mL for degrading leachate.

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