



The performance of Electro-Fenton oxidation in the removal of coliform bacteria from landfill leachate

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ABSTRACT

Leachate pollution is one of the main problems in landfilling. Researchers have yet to find an effective solution to this problem. The technology that can be used may differ based on the type of leachate produced. Coliform bacteria were recently reported as one of the most problematic pollutants in semi-aerobic (stabilized) leachate. In the present study, the performance of the Electro-Fenton process in removing coliform from leachate was investigated. The study focused on two types of leachate: Palau Borung landfill leachate with low Coliform content (200 MPN/100 m/L) and Ampang Jajar landfill leachate with high coliform content ($>24 \times 10^4$ MPN/100 m/L). Optimal conditions for the Electro-Fenton treatment process were applied on both types of leachate. Then, the coliform was examined before and after treatment using the Most Probable Number (MPN) technique. Accordingly, 100% removal of coliform was obtained at low initial coliform content, whereas 99.9% removal was obtained at high initial coliform content. The study revealed that Electro-Fenton is an efficient process in removing high concentrations of pathogenic microorganisms from stabilized leachate.

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1. Introduction

Continuous population growth and industry development has led to increase in waste generation. To date, land filling is the preferred option for the disposal and management of solid urban wastes (Tengruai et al., 2007). In spite of the number of advantages, the resulting highly polluted industrial wastewater has been a cause for significant concern, especially because land filling is the most common technique for solid waste disposal (Ghafari et al., 2005). Landfill leachate is the liquid that has seeped through solid waste in a landfill, obtaining extracted, dissolved, or suspended materials in the process (Christensen et al., 2001). Landfill leachate is a potentially polluting liquid, which may cause harmful effects on the groundwater and surface water surrounding a landfill site, unless returned to the environment in a carefully controlled manner (Scottish Environment Protection Agency (SEPA), 2003). Regardless of the concentration changes, which depend on a complex set of interrelated factors, the complexity of the landfill leachate can be categorized on the basis of four major groups of pollutants: dissolved organic matter, inorganic macro-components, heavy metals, and xenobiotic organic compounds (Aziz et al., 2004). The environmental impact of leachate is affected by leachate strength, proper leachate collection, and efficiency of leachate treatment. Leachate requires treatment to minimize the amount of pollutants to an acceptable level prior to discharge into

water networks (Aziz et al., 2010). In this context, the presence of pathogenic microorganisms in leachate was reported in literature (Bodzek et al., 2006; Mwiganga and Kansime, 2005; Grisey et al., 2010). Fecal coliform, as an indicator of the presence of bacteria, is a serious concern in the long term (Mangimbulude et al., 2009). The presence indicator of bacteria does not necessarily pose health risk, but their presence implies the potential presence of pathogenic bacteria (Haller et al., 2009). A high concentration of Coliform bacteria (0.66×10^4) was found in landfill leachate (Umar et al., 2011). The pathogenic bacteria can contaminate water; thus their presence must be controlled to avoid potential health hazards (Grisey et al., 2010). Accordingly, significant contaminant levels of coliform in groundwater from leachate and untreated wastewater have been reported in literatures. A strong correlation ($r = 0.98$) was reported between coliform in drinking water and incidence of diarrheal diseases. Significant correlations with giardiasis, the hepatitis virus, and parasitic diseases were investigated (Yassin et al., 2006; Abu Amr and Yassin, 2008). Several applications and techniques have been applied on landfill leachate to reduce the environmental risk of high organic and toxic pollutants, such as coagulation/flocculation, electro-coagulation, Fenton, Photo-Fenton, Electro-Fenton, and Ozonation (Ilhan et al., 2008; Mohajeri et al., 2010a; Atmaca, 2009; Tatsi et al., 2003; Primo et al., 2008; Tizaoui et al., 2007).

A new technique that uses Ozone/Fenton in advanced oxidation process was also recently employed (Abu Amr and Aziz, 2012). However, none of them had been reported to be effective in removing bacterial content from the effluent. Recently, Electro-Fenton

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treatments have been increasingly used for landfill leachate treatment because of its powerful and effective removal of organic and toxic elements (Atmaca, 2009; Mohajeri et al., 2010b). Moreover, Electrochemical oxidation has been reported to be effective for both viral and bacterial inactivation (Drees et al., 2003; Fang et al., 2006; Morita et al., 2000). In a recent study, 94% COD and 95% color removals were obtained using Electro-Fenton on stabilized leachate (Mohajeri et al., 2010b). Although it has good removal efficiency, which meets the acceptable organic levels and thus allows the effluent to be discharged into the environment, unlike different treatment techniques (i.e. Fenton process) which may require further treatment process prior to final discharge, the effect of Electro-Fenton oxidation in removing coliform bacteria from leachate has not been investigated, making doubtful the microbial quality of the treated effluent and thus the risk of its discharge to environment. The current study focused on two types of leachate with different coliform bacterial contents. The removal efficiency against coliform bacteria after being subjected to Electro-Fenton process was investigated. The study aims to evaluate the microbial quality of the treated effluent with stabilized leachate through the Electro-Fenton process.

2. Materials and methods

2.1. Landfill leachate characteristics and sampling

Leachate samples were obtained from two landfill leachate ponds: the Palau Burung Landfill Site (PBLs) and the Ampang Jajar Landfill Site (AJLS). PBLs is located in the Byram Forest Reserve, in Penang, Malaysia. It has an area of 62.4 ha, of which 33 ha is currently operational and receives about 2200 t of daily municipal solid wastes. It is equipped with a natural marine clay liner and three leachate collection ponds (Bashir et al., 2011). On the other hand, AJLS is located in Seberang Perai, at the mainland of the Penang State of Malaysia, at latitude of 5°24'N and longitude 100°24'22"E. It has a total surface area of 17 acres. The landfill receives both municipal and industrial wastes. It was started as a dumpsite in the early eighties. It has neither a leachate collection system nor a defined space for waste dumping. During operation, the landfill receives approximately 650 t of solid waste per day (Bashir et al., 2011). About 20 L of leachate was collected manually in plastic containers on April 19 and 28, 2012. The samples were immediately transported to the laboratory, characterized, and cooled at 4 °C to minimize biological and chemical reactions. The average characteristics of the leachate used in the experiments are summarized in Table 1. The microorganism survival depends on the initial COD concentration, Although the concentration of COD in PBLs (1400–1600 mg/L) were reported higher than that in AJLS (120–250 mg/L), however, the lower coliform content in PBLs is attributed to the high salinity level (TDS = 11414 mg/L) of leachate as a major limiting factor for growth of bacteria (Umar et al., 2009). Sample collection and preservation were performed according to the Standard Methods for the Examination of Water and Wastewater (APHA, 2005).

Table 1
Characteristics of leachate collected from PBLs and AJLS Penang, Malaysia.

Parameters	PBLs range	AJLS range
pH	8–8.5	7–8.5
COD (mg/L)	1400–1600	250–450
Color (PtCo.)	3500–4000	130–300
TDS (mg/L)	11414	2420
Coliform (MPN/100 ml)	130–200	<240,000
Temperature (°C)	25–30	25–30

2.2. Experimental procedure

A total of 18 Electro-Fenton experiments were conducted in laboratory scale on both two types of leachate with nine experiments for each, using a 500 mL beaker as reactor. The initial pH of the leachate was adjusted to desired values using concentrated sulfuric acid or sodium hydroxide, 5 min before adding the Fenton reagents. A METTLER-TOLEDO 320 pH meter was used for the pH measurements. Before the measurements, the pH meter was calibrated using standard buffers (pH 4.0, 7.0, and 10.0) at room temperature. Direct current (DC) power supply (DAZHENG, PS 305D, 30 V, 3A) was used to provide the desired current. A pair of anodic and cathodic aluminum electrodes, each was 3 cm × 5 cm (active electrode area dipped in leachate) in dimension and 3 cm apart. The conditions for the experiments were determined based on optimum conditions obtained on the same type of leachate (Mohajeri et al., 2010b). For each run, a pre-defined amount of ferrous sulphate hepta hydrate (FeSO₄·7H₂O) and hydrogen peroxide (30%) were used to activate the E-Fenton reactions. H₂O₂/Fe²⁺ was used at a molar ratio of 1:1. About 500 mL of leachate was placed in an electrolytic cell. The desired amounts of iron (Fe²⁺) and hydrogen peroxide (H₂O₂) were added, before the electrical current was turned on. All tests were conducted at room temperature (28 ± 2 °C) and atmospheric pressure. The leachate was thoroughly stirred with a magnetic stirrer at appropriate time intervals, the DC power source was turned off, and the reaction was ended. Then, the samples were allowed to settle for 30 min (for solid sedimentation), and the supernatant was collected for water quality measurements. To evaluate the effectiveness of Electro-Fenton compared with Fenton reaction; experiments on the Fenton oxidation of the landfill leachate were conducted at laboratory scale for both leachate types, with 500 mL beakers used as batch reactor. Reagent-grade sodium hydroxide and sulfuric acid (Merck, GR grade) were used for pH adjustments (pH = 3) (Mohajeri et al., 2010a). The optimum amounts of ferrous sulfate heptahydrate and hydrogen peroxide (Merck, GR grade, 31%) The optimal dosages of Fenton reagent for both the Fenton and the electro-Fenton treatments (0.05 mol L⁻¹ (1700 mg/L) H₂O₂ and 0.05 mol L⁻¹ (2800 mg/L) Fe²⁺) with H₂O₂/Fe²⁺ molar ratio 1:3 were determined based on optimum conditions obtained in the same type of leachate (Mohajeri et al., 2010a).

2.3. Analytical method

Coliform bacteria were examined before and after the Electro-Fenton reaction. The Most Probable Number (MPN) technique was used to estimate the bacterial population size, as the coliform group (World Health Organization, WHO, 1984). The MPN test is a statistics-based test that estimates the number of fecal coliforms in a water sample based on the degree of lactose fermentation by the organisms in the sample. In this test, a series of tubes of phenol red lactose broth are inoculated with measured amounts of water to determine whether the water contains any lactose-fermenting bacteria that produce gas. 25 mL bottles were used in the serial dilution process. About 1 ml aliquots were accurately transferred into the bottles marked with the dilution (1 × 10, 1 × 10⁻¹, 1 × 10⁻², 1 × 10⁻³, and 1 × 10⁻⁴). Each was prepared from a set of 5 bottles with these serial dilutions starting from the highest dilution. The bottles were incubated for 2 d (48 h) at 37 °C. The number of tubes, which showed color change from red with blue to yellow acid and gas production, was counted. The number was compared with the MPN statistical tables that give the numbers of coliform per 100 mL (McQuillen, 1951). Furthermore, Chemical oxygen demand (COD) was examined before and after both Fenton and Electro-Fenton oxidation to compare and evaluate the removal efficiency of different organics concentrations. COD concentration was

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