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An overview of electro-oxidation processes performance in stabilized landfill leachate treatment

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ABSTRACT

Among numerous techniques, electro-oxidation occupies a noticeable place in landfill leachate treatment. In recent years, electro-oxidation processes have been shown to be effective alternative for the removal of refractory organic compounds from stabilized landfill leachate. This paper aims to provide an overview about the effectiveness and mechanism of electro-oxidation processes employed for stabilized landfill leachate treatment. A review of the recent literature published on the electro-oxidation processes (i.e. electrochemical, electro-Fenton, photoelectro-Fenton, photoelectrochemical, and electrochemical peroxidation) performance and efficiency in stabilized leachate treatment are summarized and presented in this study. This study concluded that, although the high treatment cost caused by the energy consumption may limit electro-oxidation techniques, electro-oxidation processes are still a promising and effectual technology for the reduction of pollutants in stabilized landfill leachate.

Keywords: Electro-oxidation; Landfill; Stabilized leachate; Organic compounds removal

1. Introduction

Production of leachate that can seep into the ground and contaminate the ground water, surface water, and soil is generally a main drawback associated to municipal sanitary landfill disposal method [1]. Typically, landfill leachate contains large amounts of organic pollutants, ammonia-nitrogen, inorganic salts, and heavy metals [2–7]. The impact of landfills is also long term. This is due to the fact that landfills will continue to produce leachate and release biogas for a long period after closure. Primo et al. [5] reported that landfill leachate is a type of extremely polluted wastewater produced from municipal sanitary landfill. Organic substances (BOD_5 and COD) and NH_3-N are two main chemical features of environmental concern in landfill leachate.

The quantity and quality of landfill leachate vary with time, because deposited wastes consist of a wide range of inorganic, organic, and/or xenobiotic compounds that have an effect on the composition and the environmental potential of formed leachate [8]. Foo and Hameed [9] stated that the chemical composition of landfill leachate is influenced by a number of factors including seasonal precipitation,

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waste composition, and, mainly, the age of the landfill. Renou et al. [10] and Bashir et al. [11] indicated that the age of the landfill site is one of the main variables that affect the leachate characteristics. As a landfill becomes older, the biological decomposition of the deposited waste shifts from a relatively shorter initial period to longer decomposition periods commonly referred to as acidogenic and methanogenic phases [10–14]. Thus, leachate generated from young landfills (i.e. <5 years old) normally contains a huge quantity of biodegradable organic substance mainly volatile fatty acids (VFA) and are characterized by high COD (10,000 mg/L) and BOD_5/COD ratio. On the contrary, aged landfill (i.e. >10 years old) produces stabilized leachate that contains biorefractory compounds such as humic and fulvic acids, and are distinguished by a low COD of less than 4,000 mg/L and BOD_5/COD ratio of less than 0.1 [10]. Li et al. [15] reported that as the landfill leachate age increases and further stabilized, biodegradable fraction of organic substances in leachate reduces, and accordingly, conventional biological treatment methods are no longer adequate to achieve the level of decontamination required to decrease the negative effects of landfill leachate on environment. Therefore, alternative physico-chemical processes have been practiced [8,10]. Examples of the physico-chemical processes used for stabilized leachate treatment include: electro-oxidation processes [16,17], Fenton reaction [18,19], ozonation [20], ion-exchange [11,21,22], coagulation/flocculation [2,4], adsorption [23], air stripping [24], or combinations of two methods or more.

According to the literature, advanced oxidation processes (such as electrochemical oxidation, Fenton oxidation, electro-Fenton oxidation, photoelectro-Fenton, photoelectrochemical (PEC), electrochemical peroxidation (ECP), etc.) have been proved highly capable and efficient in reducing refractory organic substance and color as well as in oxidizing ammonia from raw and pretreated landfill leachate [16,19,25–30]. Furthermore, electro-oxidation can also achieve a considerably high efficiency in the removal of organic compounds from leachate compared to other physiochemical technologies, which only bring about phase transfer of the contaminants in question and do not involve chemical destruction. Nevertheless, the efficiency of the electro-oxidation techniques depends strongly on the experimental conditions and on the nature of the electrode materials [25]. Several electrodes made from different materials have been tested in electrochemical oxidation of different kinds of wastewater [25]. Recently, graphite carbon [31], aluminum [32,33], Ti/PbO_2 anode/stainless steel cathode [34], boron-doped diamond (BDD) [35], iron [36], stainless steel anode/carbon cathode [37], carbon anode/stainless steel cathode [38], and Ti/RuO_2–IrO_2 anode/stainless steel cathode [39] have been employed in the treatment of stabilized landfill leachate. Among AOPs, a combination of electrochemical oxidation and Fenton reagent has been considered to be a capable and attractive treatment technology for degradation of stabilized leachate. In recent years, there has been an increased focus on the use of electrochemical methods, which can produce $^{\bullet}OH$ as the main oxidizing agent by different ways [26,27].

The aim of this study is to provide an overview of electro-oxidation processes performance in treatment of old (stabilized) landfill leachate. Furthermore, to highlight and summarize the effectiveness of stabilized leachate treatment, various treatment combinations such as electro-Fenton, photoelectro-Fenton, PEC, ECP, and combined biochemical treatment and electrochemical oxidation are used. This review paper was prepared on the basis of the most recent scientific published literature.

2. Electrochemical oxidation mechanism

As mentioned previously, electrochemical oxidation process has been considered as one of the most effective techniques in degrading a range of refractory compounds in landfill leachate, textile wastewater, simulated wastewater, olive mill wastewater, paper mill effluents, and industrial paint wastewater [26–30,40,41]. According to Li et al. [26], this can be attributed to the role of a highly reactive radical intermediate such as hydroxyl radical ($^{\bullet}OH$) as an oxidant. The radicals with an oxidation potential (E^0) of 2.80 V can be produced in electrochemical oxidation systems and can rapidly degrade recalcitrant organics such as aromatic, chlorinated, and phenolic compounds. Radha et al. [27] concluded that the mechanism of electrochemical oxidation of wastewater is a complex phenomenon involving coupling of electron transfer reaction with a dissociate chemisorptions step.

In a previous study, Deng [42] investigated the implementations of electrochemical oxidation process in landfill leachate treatment. Deng [42] presented the most common conceptual circuit diagram that represents the design of electrochemical reactor, including a direct current (DC) power supply, a cathode, an anode, and the electrolyte (a medium that provides the ion transport mechanism between the anode and the cathode necessary to maintain the electrochemical process). The schematic diagram of electrochemical reactor is demonstrated in Fig. 1. The electrochemical oxidation of contaminants in

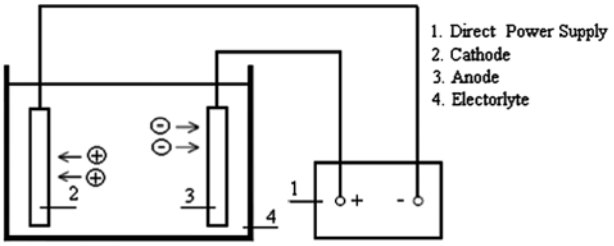

Fig. 1. Schematic diagram of electrochemical reactor [42].

waste water is accomplished through two different approaches, as illustrated in Fig. 2 [43–48]:

(i) Direct anodic oxidation, where the organic compound degradation can occur directly over the anode through the adsorbed •OH, or chemisorbed active oxygen in the anode surface (often called "anodic oxidation, direct oxidation or electrochemical incineration").

(ii) Indirect oxidation (organic compounds are treated in the bulk solution by means of species generated in the electrode).

Chemical reaction with electro generated species from water discharge at the anode such as physisorbed hydroxyl radical (•OH) or chemisorbed such as oxygen in the lattice of a metal oxide (MO) anode. The action of these oxidizing species leads to total or partial decontamination, respectively.

Li et al. [26] and Scialdone et al. [49] stated that during indirect oxidation, the agents produced on the anode, that are responsible for oxidation of inorganic and organic matters may be chlorine and hypochlorite, hydrogen peroxide, and ozone. Furthermore, during electrolysis, two species of active oxygen can be electrochemically produced on oxide anodes (MO_x). One is the chemisorbed "active oxygen" (oxygen in the oxide lattice, MO_{x+1}), responsible for the electrochemical conversion through (Eq. (1)). While the other is the physisorbed "active oxygen" (adsorbed hydroxyl radicals, •OH), responsible for electrochemical combustion through Eq. (2) [16];

$$R + MO_{x+1} \rightarrow RO + MO_x \tag{1}$$

$$R + MO_x({}^{\bullet}OH)_z \rightarrow CO_2 + zH^+ + ze + MO_x \tag{2}$$

where R symbolizes organic matters and z represents the number of absorbed •OH on anode. Throughout the electrochemical oxidation of leachate, the contaminants removal may be mainly attributed to indirect oxidation, utilizing chlorine/hypochlorite formed by anodic oxidation of chlorine originally existing or added in the leachate. On the other hand, direct anodic oxidation may slightly destroy pollutants adsorbed on the anode surface [26,42]. A series of reactions involving indirect oxidation during electro-oxidation are shown in Eqs. (3)–(9).

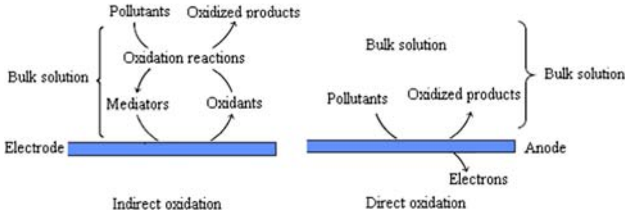

Fig. 2. Schemes of pollutant removal pathways in electrochemical oxidation process (direct and indirect oxidation) [26,33].

Anodic reactions:

$$2Cl^- \rightarrow Cl_2 + 2e^- \tag{3}$$

$$6HOCl + 3H_2O \rightarrow 2ClO_3^- + 4Cl^- + 12H^+ + 1.5O_2 + 6e^- \tag{4}$$

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \tag{5}$$

Bulk reactions:

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \tag{6}$$

$$HOCl \rightarrow H^+ + OCl^- \tag{7}$$

Cathodic reactions:

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \tag{8}$$

$$OCl^- + H_2O + 2e^- \rightarrow Cl^- + 2OH^- \tag{9}$$

Scialdone et al. [49] reported that hypochlorite (OCl^-) generated in bulk solution (Eqs. (6) and (7)) is a strong oxidant that can oxidize aqueous organic compounds. Due to the low cost of chloride, its relatively high solubility and strong oxidizing properties of the produced active chlorine, indirect electrochemical oxidation of pollutants leachated by electrochemically generated chlorine was the most commonly practiced method.

In addition to its effectiveness in refractory organic compounds removal, electrochemical oxidation methods were investigated in ammonia removal. Liu et al.

[50] and Li and Liu [51] studied the electrolytic degradation and kinetic modeling of electrolytic degradation of ammonia using Ti/IrO_2 and RuO_2/Ti anode, respectively.

The selection of electrode materials is extremely significant as it affects the selectivity and the efficiency of the process. The electrode materials must have the following properties [52]:

- High physical and chemical stability; resistance to erosion, corrosion, and formation of passivation layers.
- High electrical conductivity.
- Catalytic activity and selectivity.
- Low-cost/life ratio. Whereas the use of inexpensive and durable electrode materials must be favored.

3. Electrochemical oxidation performance in stabilized landfill leachate treatment

Nowadays, there is a continuously increasing concern for the development of advanced oxidation processes (AOPs), which is considered a highly competitive treatment technology for the removal of organic pollutants not treatable by conventional techniques due to their high chemical stability and/or low biodegradability [53–60]. The effectiveness of electrochemical oxidation of landfill leachate in various studies is illustrated in Table 1. Furthermore, the initial leachate characteristics and optimum operation conditions are presented in Table 2.

Bashir et al. [31] applied electrochemical treatment for stabilized landfill leachate using graphite carbon electrodes and sodium sulfate (Na_2SO_4) as electrolyte. The optimum conditions were found as 1,414 mg/L initial COD concentration, 79.9 mA/cm^2 current density, and 4 h reaction time. This achieved 70% BOD_5 removal, 68% COD removal, and 84% color removal. Bashir et al. [31] mentioned that electrochemical treatment of stabilized landfill leachate using graphite carbon electrode was effective in BOD_5, COD, and color removals. However, the color concentration of the treated samples increased at low influent COD and high current density owing to corrosion of electrode material. Mohajeri et al. [32] studied the electrochemical oxidation process to eliminate COD and color from semi-aerobic stabilized landfill leachate collected from Pulau Burung Landfill Site, Penang, Malaysia. Experiments were carried out in a batch laboratory-scale system in the presence of NaCl as electrolyte and aluminum electrodes. In optimized condition, maximum removals for COD (49.3%) and color (59.2%) were achieved at current density of 75 mA/cm^2, electrolyte concentration 2,000 mg/L, and reaction time 218 min. Isa et al. [33] studied the effectiveness of electrochemical treatment in treating semi-aerobic landfill leachate. The electrolytic reactor assembly included a pair of aluminum electrodes, a power supply, and NaCl as electrolyte. At optimum conditions, (2000 mg/L initial COD, about 17 mA/cm^2 current density, 4 h reaction time, and 1 g/L NaCl), COD removal, BOD_5 removal, and color removal were estimated to be 82.2, 81.5, and 95%, respectively. Panizza et al. [34] studied the anodic oxidation of a raw leachate from an old municipal solid waste landfill located near the city of La Spezia (Italy) using an electrolytic flow cell equipped with a Ti grid coated with a lead dioxide (Ti/PbO_2) as the anode and stainless steel as the cathode. The method succeeded by removing 80% of COD. They found that the contribution of the indirect process mediated by the active chlorine electro generated from the chloride ions oxidation to the reduction of the organic load of the effluent was larger than that of the $^{\bullet}OH$ mediated process. Also, they concluded that an increase in the applied current intensity, pH level, and temperature accelerated the removal of COD and improved the current efficiency. Papastavrou et al. [35] focused on the treatment of landfill leachate that biologically pretreated via electrochemical oxidation. Electrochemical oxidation over a boron-doped diamond electrode achieved about 90% COD removal in 4 h with the resulting stream having a COD content as low as 50 mg/L. According to Papastavrou et al. [35], an increase in current intensity from 15 A to 21 A had no realistic effect on the overall COD removal, which followed first-order kinetics. Orkun and Kuleyin [36] found that the optimum COD removal (65.85%) by using iron electrode was obtained with typical operating conditions: current density, 30 mA/cm^2; operation time, 180 min; conductivity, 16.4 ms/cm; and initial pH, 6.54. Misra et al. [37] studied the treatment of landfill leachate by using a batch electrolytic parallel plate (Stainless Steel/Carbon) and fixed bed three-dimensional electrode reactor using granular-activated carbon as a particle electrode. A maximum of 78.2% COD removal was achieved. They concluded that electro-oxidation of leachate in three-dimensional electrode reactor required less electrical energy compared to parallel plate configuration. Rao et al. [38] considered the electro-oxidation of high-strength leachate from an industrial landfill using three-dimensional carbon bed electrode reactor (TDR). They reported that a 60–64% COD and 66–73% TOC were removed during 1 h. The removal of COD and TOC slowly improved with treatment time, and 76–80% COD and 83–85% TOC removals were

Table 1
Electrochemical oxidation efficiency of landfill leachate in various studies

Leachate type	Electrode materials		Removal efficiency (%)					Ref.
	Anode	Cathode	COD	Color	Ammonia	TKN	BOD	
Raw leachate	Ti-TiO_2-RuO_2-IrO_2	Copper	78		99.6			[17]
Raw leachate	Graphite carbon	Graphite carbon	68	84	–	–	70	[31]
Raw leachate	Aluminum	Aluminum	49.3	59.2	–	–	–	[32]
Raw leachate	Aluminum	Aluminum	81.5	95	–	–	82	[33]
Raw leachate	Ti/PbO_2	Stainless steel	80	–	–	–	–	[34]
Biologically pretreated leachate	Boron-doped diamond (BDD)	Boron-doped diamond (BDD)	90	–	–	–	–	[35]
Raw leachate	Iron	Iron	65.8	–	–	–	–	[36]
Raw leachate	Stainless steel	Carbon	78.2	–	–	–	–	[37]
Raw leachate	Carbon	Stainless steel	80	–	–	97	–	[38]
Raw leachate	Ti/RuO_2–IrO_2	Stainless steel	26.5	–	81	–	–	[39]
Raw leachate	Aluminum	Aluminum	59	–	14	–	–	[60]
	Iron	Iron	35	–	11	–	–	
Raw leachate	Aluminum	Aluminum	–	–	–	66	91.7	[61]
Raw leachate	Aluminum plates	Aluminum plates	45	–	–	–	–	[62]
Biologically pretreated leachate	BDD	Stainless steel	–	–	100	–	–	[63]
Raw leachate	30% RuO_2 and 70% TiO_2 coated titanium	30% RuO_2 and 70% TiO_2 coated titanium	73	86	49	–	71	[64]
Raw leachate	(PbO_2/Ti), binary Ru–Ti oxide-coated titanium (DSA type), and ternary Sn–Pd–Ru oxide-coated titanium (SPR)	(PbO_2/Ti), binary Ru–Ti oxide-coated titanium (DSA type), and ternary Sn–Pd–Ru oxide-coated titanium (SPR)	92		100	–	–	[65]
Raw leachate	Aluminum	Aluminum	70	56	24	–	–	[66]
Raw leachate	Ti/IrO_2–RuO_2	Zirconium	90	100	–	–	–	[67]
Pretreated leachate	Ti plate coated with platinum and iridium dioxide	Steel plates	56.7		>95	–	–	[69]
Raw leachate	BDD	Stainless steel	89		>95	–	–	[68]
Biologically pretreated leachate	BDD	Stainless steel cathode	43.3	–	–	–	–	[69]
Raw leachate	BDD	Stainless steel	100		100			[70]

Table 2
Initial leachate characteristics and optimum operation conditions in various studies

Initial concentration	Optimum operation conditions	Ref.
COD, 1,140 mg/L; ammonia, 800 mg/L; and pH, 7.9	Reaction time (RT), 7 h; and current density (CD), 5.4 A/dm^2	[17]
BOD$_5$, 80 mg/L; BOD$_5$/COD, 0.043; COD, 1,870 mg/L; color, 2,970Pt–Co; and pH, 8.9	Na$_2$SO$_4$, 1 g/L; initial COD conc., 1,414 mg/L; RT, 4 h; and CD, 79.9 mA/cm^2	[31]
BOD$_5$, 237 mg/L; COD, 2,600 mg/L; BOD$_5$/ COD, 0.09; color, 3,250 Pt.Co; and pH, 8.0	NaCl, 2 g/L; RT 218 min; and CD,75 mA/cm^2	[32]
BOD$_5$, 366 mg/L; COD, 3,032 mg/L; color, 4,970 Pt-Co; and BOD$_5$/COD, 0.12	NaCl, 1 g/L; initial COD conc., 2,000 mg/L; RT, 4 h; and CD 17 mA/cm^2	[33]
COD, 780 mg/ L; BOD$_5$/COD < 0.1; and pH = 8.20	pH, 8.2; *T*, 50°C; flow rate = 420 L/h; RT; 3 h; I = 2A	[34]
COD, 500 mg/ L	RT, 4 h; I, 15 A	[35]
BOD$_5$, 6,400 mg/L; COD, 11,000 mg/L; Ammonia, 1,247 mg/L; and pH, 7.95	pH, 6.54; *T*, 25°C; RT, 3 h; and CD, 30 mA/cm^2	[36]
COD, 17,100–18,400 mg/L; TKN, 2,000– 2,240 mg/L; BOD$_5$/ COD, 0.05; and pH,8.9	RT, 6 h; I, 3 A	[38]
BOD$_5$, 560 mg/L; COD, 3,782 mg/L; Ammonia, 3,143 mg/L; and pH, 8.4	RT, 3 h; and CD 60 mA/cm^2	[39]
BOD$_5$, 5,270 mg/L; COD, 12,860 mg/L; Ammonia, 2,240 mg/L; BOD$_5$/COD, 0.4; and pH, 8.2	RT, 30 min; and CD, 63.1 mA/cm^2	[60]
BOD$_5$, 132 mg/L; TKN, 372 mg/L; and pH, 7.6	NaCl, 2.9 g/L; pH, 7.6 RT, 38.8 min; and CD, 10.99 mA/cm^2	[61]
COD, 380 mg/L; and BOD$_5$/COD, 0.18	N/A	[62]
BOD$_5$, 70 mg/L; COD, 380 mg/L; and BOD$_5$/COD, 0.18	RT, 6 h; and CD, 30–90 mA/cm^2	[63]
BOD$_5$, 640 mg/L; COD, 4,434 mg/L; BOD$_5$/COD, 0.14; and pH, 8.35	Flow rate, 2,000 L/h; RT, 3 h; and CD, 116 mA/cm^2	[64]
COD, 4,100 mg/L; Ammonia, 2,600 mg/L; and BOD/COD, 0.2	NaCl, 7.5 g/L; RT, 4 h; CD, 0.15 A/cm^2	[65]
COD, 28,200–34,200 mg/L; Ammonia, 1,599–1,631 mg/L; BOD$_5$/ COD ratio, 0.09–0.12; and pH, 7.6–8.9	Stirring speed, 150 rpm; and CD, 50 mA/cm^2	[66]
BOD$_5$, 120 mg/L; COD, 2,960 mg/L; BOD$_5$/COD, 0.4; and pH, 8	*T*, 80°C; RT, 4 h; and CD, 32 mA/cm^2	[67]
BOD$_5$, 4–20 mg/L; COD, 400–500 mg/LAmmonia, 5–15 mg/L; and pH, 8–9	RT, 1 h; and CD, 20 mA/cm^2	[69]
COD, 860 mg/L; Ammonia, 780 mg/L; and pH, 8.16	RT, 1 h; and CD, 30 mA/cm^2	[68]
COD, 5,800 mg/L; Ammonia, 1,210 mg/L; and pH, 8.4	RT, 6 h; and CD, 50 mA/cm^2	[69]
COD, 4,430 mg/L; BOD$_5$/COD 0.14; and N-NH$_4$ 1,930 mg/L and pH, 8.35	*T*, 20°C; RT, 6–8 h; and CD, 30 mA/cm^2	[70]

obtained at 6 h. About 57–64% TKN was removed during 1 h. In this study, about 97% TKN was removed in 6 h at applied current of 3 A. Zhang et al. [39] compared the performance of a two-dimensional electrode reactor with a same size three-dimensional electrochemical reactor (leachate characteristics were pH 8.4, COD 2091 mg/L, and ammonia 2,531 mg/L) at 60 mA/cm^2 of current density and 3 h of reaction time. In the two-dimensional electrode reactor, 20.2% COD removal efficiency was obtained, while COD removal efficiency increased to 26.5% in the three-dimensional electrochemical reactor. However, ammonia removal efficiency was as high as 81.1% in the three-dimensional electrochemical reaction compared with only 57.7% in the two-dimensional electrode reactor. The employment of electrochemical oxidation for treatment of Odayeri (Turkey) landfill leachate showed that over 59% of COD and 14% of ammonia removal were accomplished by using aluminum electrodes, 30 min contact time, and a current density of 631 A/m^2 [61]. Contreras et al. [62] investigated landfill leachate treatment by using aluminum electrode. The optimal conditions obtained were time 38.8 min, pH 7.6, current density 109.9 A/m^2, and NaCl 2.9 g/L. The results showed that 96.9% of turbidity, 97.0% of Al, 99.5% of Fe, and 66% of total Kjeldahl nitrogen were removed. Furthermore, the sludge formed exhibited a good floc size, which separated in less than 10 min by classical sedimentation [62].

Electrochemical oxidation tests were carried out also by Labanowski et al. [63] in a laboratory pilot using aluminum plates. The leachate used in this study came from the landfill of Crezin (Limoges, France), and it presented low biodegradability and high concentration of macromolecules. Labanowski et al. [63] achieved 45% reduction of COD. Moraes and Bertazzoli [65] investigated treatment of leachate collected from sanitary landfill in Rio Claro (Brazil) using flow electrochemical reactor. The collected leachate was treated via electrochemical oxidation in a pilot-scale flow reactor, using oxide-coated titanium anode. The experiments were conducted under a constant flow rate of 2,000 L/h, and the effect of current density on chemical oxygen demand, total organic carbon, color, and ammonium removal was investigated. At a current density of 116.0 mA/cm^2 and 180 min of processing, the removal rates obtained were 57% for TOC, 86% for color, 73% for COD, and 49% for ammonium. Chiang et al. [66] evaluated the efficiency of various anode types including graphite, PbO_2/Ti, binary Ru–Ti oxide-coated titanium (DSA type), and ternary Sn–Pd–Ru oxide-coated titanium (SPR) for landfill leachate via electrochemical oxidation process in the presence of different concentrations of chloride ions. The removal effectiveness of COD was decreased in the order SPR > DSA > PbO_2 > graphite due to the higher chlorine/hypochlorite production efficiency of the SPR anode. A 92% COD removal and a total ammonium removal were obtained after 240 min by the SPR at 0.15A/cm^2 when the chloride content was 7,500 mg/L. Anglada et al. [70–74] clarified the different phenomena that take place during the electro-oxidation process of landfill leachate at a pilot plant scale with boron-doped diamond (BDD) anodes. The evolution of the level of chloride ions, chlorate, inorganic carbon, pH, and redox potential was found to be interrelated. Effectively, they applied two mathematical models for describing the oxidation kinetics of COD and ammonium. In their study, Yong et al. [75] indicated that the influence of pH value on the treatment efficiency of landfill leachate was correlated with the concentrations of chloride ion and heavy metal ion. Lei and Baoshan [76] reviewed the technical applicability and the treatment performance of various electrochemical oxidations for landfill leachate. Lei and Baoshan [76] summarized the theorem of the process and its applications in landfill leachate treatment.

From the previous studies, it can be concluded that electrochemical oxidation is still a promising and influential technology, particularly for low BOD_5/COD ratio or high-toxic landfill leachate where biological methods are not effective [64]. However, the forthcoming research is required to elucidate the influence of pH, and the hazard of chlorinated organics formation throughout electro-oxidation of landfill leachate [64]. In this regard, the formations of undesirable oxidation by-products such as nitrate ions [64] as well as chlorinated organic substances [77] were recently detected. According to the literature, in the presence of chloride, there are some reactions that can be occurred leading to the formation of undesirable by-products such as chloramines. Chloramines (very toxic substances) are produced due to the presence of ammonia and free chlorine in the electrolysis system, and their prevention is very significant for environmental protection [78–83].

In addition to organic compound removal from landfill leachate with a low BOD_5/COD ratio, Anglada et al. [82] addressed the concern of formation of chlorinated organic compounds and nitrate ions as a result of organic matter and ammonia and/or organic nitrogen electro-oxidation in the presence of chloride ions. In their study, the process was evaluated on COD, total nitrogen, and color removal, as well as on the formation of nitrate, nitrite, and chlorinated organics. At applied current of 18 A and reaction time of 8 h, high levels of decolorization (84%), COD removal (51%), and ammonia removal (32%) were achieved. However, the concentration of chlorinated organics increased continuously with treatment time reaching values of 1.9 μg/L, 753 mg/L, and 431 μg/L of trihalomethanes (THMs), haloacetonitriles (HANs), and haloketons (HKs), respectively [82]. In another study, Perez et al. [83] investigated the formation of oxidation by-products during the electrochemical removal of ammonium using BDD electrodes from landfill leachate containing chlorides. Two different levels of chloride concentration were studied i.e. low chloride concentrations (0–2,000 mg/L), and high chloride concentrations (5,000–20,000 mg/L). At high chloride concentration, the formation of nitrogen gas and the rate of ammonium removal were favored [83]. Moreover, chloride was also oxidized during the electrochemical treatment leading to the formation of free chlorine responsible of the ammonium oxidation, together with undesirable products such as chloramines, chlorate, and perchlorate [83]. Perez et al. [83] reported that the concentration of chloride had an important influence on the formation kinetics of the oxidation by-products. In view of that, at low chloride concentrations, chlorate appeared like an intermediate compound leading to the formation of perchlorate. However, at high chloride concentrations, chlorate formation was delayed significantly and perchlorate was not detected during the experimental time [83].

On the other hand, the main problems to be tackled before the full-scale employment ofelectrochemical oxidation is the reduction of operating cost and the development of competent and stable electrode materials [52]. Therefore, in order to improve the application of electro-oxidation of landfill leachate, improvement of electrodes material for more economical and effective electrodes is exclusively required [64]. Recently, various researchers focused on the energy consumption [17,34,36,67,70,73]. The energy consumption for the electrochemical treatment was evaluated by Nisha Priya et al. [17] taking into consideration the electrolysis time, current intensity, and voltage. According to their study, the leachate treatment required 193.35 KWh/kg COD of energy which can be decreased to 121.00 KWh/kg COD if 1,000 mg/L of chloride was added [17]. According to Panizza et al. [34], the specific energy consumption necessary to reduce the organic load to below the disposal limit was 90 kWh/m^3. Bouhezila et al. [67] used electrochemical reactor in leachate treatment. The findings in this study show that the removal efficiencies of COD, total nitrogen, color, and turbidity were respectively 70, 24, 56, and 60% with Al electrodes and 68, 15, 28, and 16% with Fe electrodes. Electrical energy consumption and operating cost were 0.022 kWh/L, 0.54 US$/m^3 with Al electrodes, and 0.019 kWh/L, 0.47 US$/m^3 with Fe electrodes.

In their study, Panizza et al. [34] mentioned that specific energy consumption necessary to reduce the organic load to below the disposal limit was 90 kWh/m^3. On the contrary, Anglada et al. [70] reported that the specific energy consumption required for reaching the disposal limits for COD and ammonia in Spain were 54 kWh/m^3 and 93kWh/m^3, respectively. Thus, according to Anglada et al. [70], the energy cost was €9.3/m^3. Nevertheless, the prices of energy cost are highly dependent on the particular country. Moreover, it should be kept in mind that energy price is significantly dependent on the required level of treatment. Additionally, it should be noted that the operating cost involves costs of chemicals, electrodes, and energy consumptions plus labor, maintenance, sludge dewatering, and disposal, and fixed costs [36].

4. Electro-oxidation related processes

4.1. Electro-Fenton (EF)

Fenton process has attracted great interest in view of its high capacity to generate hydroxyl radicals through decomposition of H_2O_2 by Fe^{2+} in acidic conditions [84,85]. According to Brillas et al. [86], the efficiency of Fenton oxidation process can be considerably improved by using electricity (electro-Fenton) for the generation of additional hydroxyl radicals. The electro-Fenton processes include electrochemical reactions for the *in situ* generation of the reagents used for the Fenton reaction. The generated reagents depend on solution conditions, cell potential, and nature of electrodes [87].

In the electro-Fenton process, Fenton process and electrochemicals are combined to increase the degradability of organic substances present in high-polluted wastewaters [87,88]. Under electrically assisted Fenton reaction, more $^\bullet OH$ radicals are generated and the oxidation of the organics to CO_2 can be enhanced in the same period [87–89]. The enhanced generation of hydroxyl radicals in the presence of electricity guarantees considerable enhancement in the reduction of pollutants. The electro-Fenton process has two different configurations. In the first one, Fenton reagents are added to the reactor from outside and inert electrodes with high catalytic activity are utilized as anode material. However, in the second configuration, only hydrogen peroxide is added from outside and Fe^{2+} is added from sacrificial cast iron anodes. The electro-Fenton process has been recently employed in stabilized landfill leachate treatment by different researchers [90–96].

The electro-Fenton process can proceed by the following chain reactions [84,87,94]. The Fe^{2+} initiates and catalyses the decomposition of H_2O_2, resulting in the production of hydroxyl radicals ($^\bullet OH$) in the solution. Hydroxyl radicals are also generated at the surface of a high oxygen overvoltage anode from water oxidation. This reaction increases oxidative potential of the process due to the increase of the $^\bullet OH$. The produced $^\bullet OH$ is the main oxidizing agent of organic matter, i.e. its conversion to CO_2, water, and inorganic ions. The electro-Fenton oxidation process can be illustrated by the following chain reactions:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + {}^\bullet OH \tag{10}$$

Hydroxyl radicals are also generated at the surface of a high oxygen overvoltage anode from water oxidation:

$$H_2O \rightarrow {}^\bullet OH + H^+ + e^- \tag{11}$$

Eq. (10) is propagated from ferrous ion regeneration mainly by the reduction of the produced ferric species with hydrogen peroxide:

$$H_2O_2 + Fe^{3+} \rightarrow Fe^{2+} + HO_2 + H^+ \quad (12)$$

Also the generated ferric ion from Eq. (10) can be reduced to ferrous ion by electrochemical regeneration of Fe^{2+} ions on the cathode surface:

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \quad (13)$$

Furthermore, ferrous ions can be quickly destroyed by hydroxyl radicals:

$$Fe^{2+} + {}^{\bullet}OH \rightarrow Fe^{3+} + OH^- \quad (14)$$

The generated hydroxyl radicals would react rapidly with most of the organic compounds, either by addition to a double bond or by abstraction of a hydrogen atom from aliphatic organic molecules:

$$RH + {}^{\bullet}OH \rightarrow H_2O + R^{\bullet} \quad (15)$$

The resulting organic radicals then react with oxygen to initiate a series of oxidation reactions leading to mineralization of the organics to produce CO_2 and H_2O.

The electro-Fenton process is a promising alternative for mineralization and reduction of recalcitrant organic compounds in landfill leachate [89–96]. Urtiaga et al. [90] concluded that, as a result of the advanced oxidation process that combines Fenton and electro-oxidation processes, the highly polluted raw landfill leachate characterized by an average value of COD = 4,430 mg/L and N-NH_4^+ of 1,225 mg/L has been transformed into an aqueous effluent with COD and ammonium contents below the discharge limits, COD < 160 mg/L, and N-NH_4^+ < 15 mg/L. The testing of the integrated process has been performed on-site at pilot scale. In another study, Mohajeri et al. [91] optimized the operating parameters in electro-Fenton process, for the removal of recalcitrant organics from semi-aerobic landfill leachate using response surface methodology (RSM). In their study, the efficiency of the essential process variables such as H_2O_2/Fe^{2+} molar ratio, current density, pH, and reaction time were determined. The optimum removals were determined as 94.07% of COD and 95.83% of color at pH 3 and H_2O_2/Fe^{2+} molar ratio was 1, contact time was 43 min, while current density was 49 mA/cm^2. However, applying cast iron plates for both anodes and cathodes with surface area of 22.6 cm^2 and yielded 72 and 90% removal of COD and color, respectively, for a treatment period of 20 min, constant DC current of 3A and H_2O_2 concentration of 2,000 mg/L at initial pH 3 [92]. Shi et al. [93] studied landfill leachate treatment by a three-dimensional electrode electro-Fenton process. The optimal operating conditions were found as current density of 57.1 mA/cm^2, electrode distance of 10 cm, initial pH 4.0, dosage of Fe^{2+} of 1 mmol/L, and air flow of 0.2 m^3/hr. Under these conditions, the COD and TOC removal efficiencies reached 80.8% and 73.26%, respectively. The biodegradability (BOD_5/COD ratio) was increased from 0.125 to 0.486. Zhang et al. [94] used Ti/ RuO_2- and IrO_2-type electrodes as anode materials and added the Fenton reagent from outside for the treatment of high-strength landfill leachate. Boye et al. [95] investigated the treatment of Istrana landfill leachate (Treviso, Italy) via electro-Fenton process. Whereas H_2O_2-assisted process was used together with iron anode coupled with Ti (Pt) cathode. The COD removal was in the range of 60–80% in 6 h for the leachate. They indicated that electro-Fenton technology is suitable for landfill leachate treatment. The electro-Fenton method together with chemical coagulation as pretreatment process before biological treatment was investigated by Lin and Chang [96]. The authors used two pairs of electrodes (cast iron plates) and they added H_2O_2 to the electrolytic cell before the electrical current was started.

4.2. Photoelectro-Fenton (PEF)

The effectiveness of electro-Fenton method can be considerably enhanced in the presence of UV irradiation by a process called photoelectro-Fenton [88,97–101]. The catalytic effect of Fe^{2+} can be enhanced by assisting electro-Fenton process with UV irradiation. The photoelectro-Fenton process can increase the regeneration rate of Fe^{2+} in the presence of UV [46,90,97–99]. An increased concentration of hydrogen peroxide increases the oxidative capability of the process. Moreover, H_2O_2 produces two ${}^{\bullet}OH$ by photocatalytic effect of UV irradiation [46,100].

$$H_2O_2 + hv \rightarrow 2{}^{\bullet}OH \quad (16)$$

The degradation action of UVA irradiation is complicated and can be illustrated by Altin [94] and Tauchert [100]. According to Ting et al. [97] and Brillas et al. [86], PEF is a type of Fenton technology which uses the same conditions as that of the electro-Fenton process with simultaneous irradiation of UVA light. This accelerates the degradation rate of organic pollutants in the reaction and increases the regeneration rate of Fe^{2+} as well. Additional ${}^{\bullet}OH$ can also be observed because of the photolysis of $[Fe(OH)]^{2+}$ and

Fe(III) complexes as illustrated in reactions (17) and (18) [86,101].

$$(\text{Fe-OH})^{2+} + hv \rightarrow \text{Fe}^{2+} + {}^{\bullet}\text{OH} \tag{17}$$

$$\text{Fe(OOCR)}^{(2+)} + hv \rightarrow \text{Fe}^{2+} + \text{CO}_2 + \text{R}^{\bullet} \tag{18}$$

In their research, Ting et al. [97] used a set of sixteen 3 W UVA lamps fixed inside a cylindrical Pyrex tube (allowing wavelengths $\lambda > 320$ nm to penetrate) as an irradiation source. In addition to all the experimental conditions mentioned above, UV light with maximum wavelength of 360 nm was irradiated inside the reactor, supplying a photoionization energy input to the solution of 48 W. Environmental application of the PEF method is quite a new subject and previous studies are fairly limited [100,101]. In old landfill leachate, treatment using photo-Fenton process alone was studied by Primo et al. [5]. They reported photo-Fenton process as an efficient alternative for the treatment of biologically pretreated landfill leachate and the process achieved about 86% removal of COD. Altin [101] investigated the performance of utilizing photoelectro-Fenton in landfill leachate treatment. Fig. 3 shows the schematic diagram of photoelectron-Fenton reactor.

Altin [101] compared the effectiveness of photoelectro-Fenton technique with electro-Fenton technique and reported an additional 10% COD removal by photoelectro-Fenton process in comparison with electro-Fenton. Altin [101] indicated that the obtained results from the comparative experiments revealed that the PEF process was more effective than electro-Fenton and UV/H_2O_2 processes for the landfill leachate treatment efficiency.

Chunxia et al. [98] also applied PEF oxidation method to treat landfill leachate, which was pretreated by biochemical process and sampled from a landfill in Beijing City, China. They reported that the photoelectro-Fenton reaction could degrade landfill leachate effectively. Whereas, 78.9% of TOC, 62.8% of COD, and 100% of color were removed at 360 min electrolysis and under the operation conditions of 0.5 A current, 1 mmol/L Fe^{2+}, 250 mL/min O_2 at pH 3. Nevertheless, total decolorization and 60% of COD removal were obtained (stabilized leachate with a pH of 8, COD of 5,500 mg/L and $BOD_5 < 20$ mg/L) at reaction times of 300 min [98].

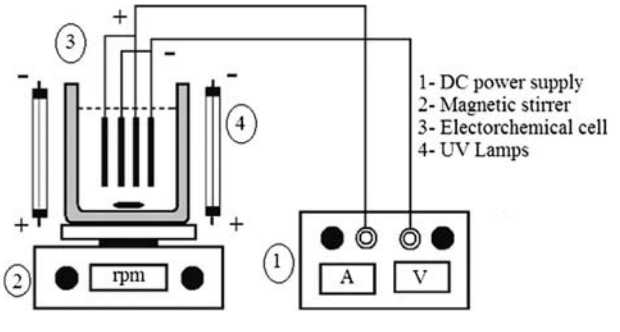

Fig. 3. Schematic diagram of photoelectron-Fenton reactor [101].

4.3. Photoelectrochemical

Recently, PEC oxidation has been employed as an effective method in removing organic contaminants. Zhao et al. [102] reported that the landfill leachate can be efficiently removed by photo-assisted electrolysis method. In their study, leachate was treated by PEC oxidation in a pilot-scale flow reactor, using UV light irradiation. At a current density of 67.1 mA/cm^2 and 2.5 h reaction time, the reduction rates obtained were 74.1% COD, 41.6% TOC, and 94.5% ammonium in the electrolysis process with UV light irradiation. In comparison, the removal rates of COD, TOC, and ammonium were decreased in the individual electrolysis process, respectively. Zhao et al. [102] indicated that the increase induced by the UV light irradiation was analyzed. Landfill leachate treatment via PEC procedure was also reported by Tauchert et al. [100]. The PEC process showed a low degradation capacity towards landfill leachate, basically due to the negative effect of its dark coloration in the photochemical reactions. By using a prior precipitation process, the efficiency of the PEC treatment was significantly improved, permitting 90% and 60% of decolorization and COD removal, respectively [100]. Taking into account the extremely complex character of the leachate and its usual resistance to conventional degradation processes, the result reported by Tauchert et al. [100] confirm the high potentiality of PEC processes to the remediation of recalcitrant residues. Xiao et al. [103] studied PEC treatment of landfill leachate with activated carbon fiber cathode and RuO_2/Ti anode. The formations of hydroxyl and chlorine radicals, which were disassociated from H_2O_2 and active chlorine by UV and Fe^{2+}, were proposed to be the main mechanism. Under the conditions of initial pH value of 2.0~5.1, current intensity of 15.0 mA/cm^2, and Fe^{2+} addition of 1 mmol/L, the removals of TOC reached 80% within 300 min [103].

4.4. Electrochemical peroxidation

Paton et al. [104] investigated the feasibility of using ECP for the treatment of land fill leachate generated from Stewartby landfill (Bedfordshire, UK). The optimization of the process was conducted by varying electrode voltage, polarity switching rate, initial pH, and hydrogen peroxide concentration. This resulted in an observed 36% BOD_5 and 52% COD reduction by adjusting the pH to 4.0, applying an electrode voltage of 1.9V and switching speed of 5 cycles per minute [104]. Paton et al. [104] concluded that ECP process is effective in the treatment of landfill leachate, and suggested that with further optimization, ECP may serve as an attractive option to current biological leachate treatment methods.

In general, ECP was first explained by Scrudato and Chiarenzelli [105]. According to their study, ECP involves the oxidation of aqueous or suspended organic substance by the action of hydroxyl free radicals, generated from hydrogen peroxide and ferrous ions, which are electrochemically produces from cast iron or mild steel electrodes [105]. The resultant product is recognized as Fenton's reagent. Recently, ECP achieved a remarkable degradation rate of dye in wastewater [106].

4.5. Combined biochemical treatment and electrochemical oxidation

A synergistic combination of the biochemical treatment and electrochemical oxidation (SBEO) with sectional treatment on a boron-doped diamond (BDD) electrode is proposed for stabilized landfill leachate treatment by Zhao et al. [107]. The first stage involves the synergistic system of biochemical treatment and electrochemical oxidation. After that, the second stage was followed by individual biochemical treatment. The value of BOD_5/COD ratio was improved from 0.016 to 0.51. Moreover, the toxicity index was reduced to 30.2% from 82.5% at 2 h. That is, not only was the efficiency of biodegradability greatly improved, but the toxicity was also dramatically reduced. The TOC removal reached 83.1%. The authors studied the practicability and the efficiency of SBEO process based on the most important aspects such as BOD_5/COD ratio, toxicity, and TOC removals.

Chiang et al. [108] reported that an expensive energy loading is required for the individual electrochemical treatment of landfill leachate. Therefore, they used coagulation and adsorption pretreatments to aid the electrochemical oxidation treatment of landfill leachate. Electrochemical oxidation combined with pretreatment processes was found to achieve better results than the individual electrochemical oxidation process. COD and ammonium removal efficiencies of 90.3% and 80.1% were achieved by electrochemical oxidation combined with adsorption [108].

Treatment of old-aged landfill leachate by combined chemical and biological methods was conducted by Lin and Chang [106]. In this study, the electro-Fenton method assisted by chemical coagulation was employed and found to be highly efficient in treating a large amount of refractory organic and inorganic compounds. The sequencing batch reactor (SBR) was capable of further elevating the water quality of leachate effluent to the discharge or nonpotable reuse standard. The electro-Fenton oxidation employed was able to greatly enhance the COD removal of the leachate effluent from chemical coagulation. At an optimum pH 4, the oxidation process achieved about 100% of color removal. An optimum H_2O_2 dosage of 750 mg/L yields a very good COD, color, NH_3-N, and phosphorus removals in less than 30 min [109]. Treatability of landfill leachate by combining both up flow anaerobic sludge blanket reactor (UASB) and electrochemical oxidation was investigated [110–112]. The leachate with a 4,750 mg/L of COD and 1,310 mg/L of NH_3-N was first treated in a UASB reactor. The process removed about 66% of COD by Li et al. [112], which indicated that the treatment of landfill leachate by the combined system was very effective.

4.6. Combined membrane biological reactor and electrochemical oxidation

In general, the integrated membrane process is considered an efficient and cost-effective process in the removal of nonbiodegradable organic and nitrogenous matters from landfill leachate [113]. For instance, Laitinen et al. [114] assayed sequencing batch reactor and membrane bioreactor for the treatment of landfill leachate and found that the MBR effluent was significantly better in quality. Recently, Feki et al. [115] studied the integration of membrane bioreactor (MBR) with electrochemical oxidation process for the treatment of landfill-stabilized leachate. Electrochemical oxidation process using Ti/Pt, graphite, and PbO_2 electrodes was employed as a post-treatment method. The combination of MBR with electrochemical oxidation can be a technically suitable solution for stabilized leachate treatment with an efficient reduction of different parameters, particularly COD (85%), TKN (94%), and color (99%). Another application of electrochemical oxidation using Pi/Ti was applied as a post-treatment and after the biological process in order to reduce the residual ammonia and COD from Djebel Chekir (Tunisia) landfill leachate carried out by Aloui and co-authors [116]. They found that at a pH value of 9, current density of 4 A/dm^2, and electrolysis duration of 60 min, COD was reduced to 27 mg/L.

5. Conclusions

The electro-oxidation of municipal landfill-stabilized leachate can result in removing of a considerable amount of nonbiodegradable refractory matters (COD), color, and ammonia. Thus, electrochemical oxidation is a promising and powerful technology, mainly for low BOD_5/COD or high toxic landfill leachate which is hard to be treated biologically. During the electro-oxidation of leachate, the reduction of contaminants is mainly attributed to the indirect oxidation. On the other hand, direct anodic oxidation slightly destroys pollutants adsorbed on the anode surface. Using combination processes—such as EF, PEF, PEC, ECP, and combined biochemical treatment with electrochemical oxidation—can considerably improve the treatment performance efficiency. However, the main drawbacks to overcome before the full-scale implementation of electro-oxidation are the high electrical energy demand, which causes high operational costs, as well as the improvement of efficient and stable electrode materials. Minimization of energy consumption can be further achieved by the optimization of other reaction conditions, for example, operational pH, $H_2O_2:Fe^{2+}$ molar ratio, current density, and reaction time. On the other hand, future research is expected to develop more economical and effective electrode materials.

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