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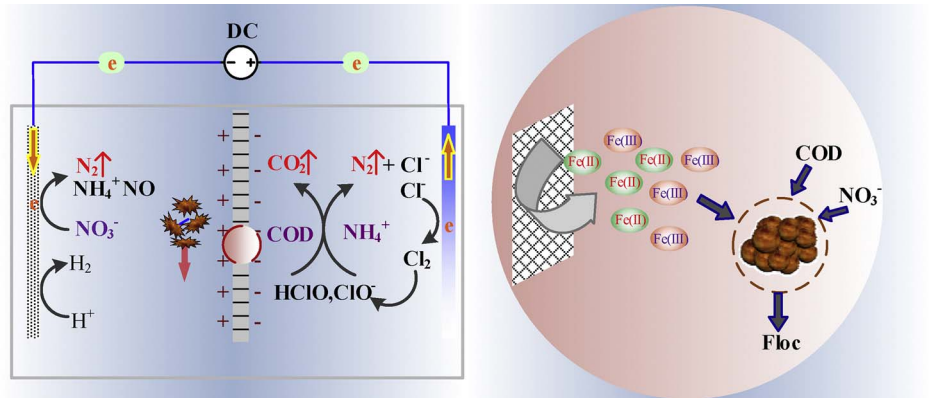
Tertiary treatment of landfill leachate by an integrated Electro-Oxidation/Electro-Coagulation/Electro-Reduction process: Performance and mechanism

Jing Ding[a], Liangliang Wei[a], Huibin Huang[a], Qingliang Zhao[a,*], Weizhu Hou[a], Felix Tetteh Kabutey[a], Yixing Yuan[a], Dionysios D. Dionysiou[b]

[a] State Key Laboratory of Urban Water Resources and Environment (SKLUWRE), School of Environment, Harbin Institute of Technology, Harbin 150090, China
[b] Environmental Engineering and Science Program, Department of Chemical and Environmental Engineering, University of Cincinnati, Cincinnati, OH 45221, United States

GRAPHICAL ABSTRACT



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ABSTRACT

This study presents an integrated Electro-Oxidation/Electro-Coagulation/Electro-Reduction (EO/EC/ER) process for tertiary landfill leachate treatment. The influence of variables including leachate characteristics and operation conditions on the performance of EO/EC/ER process was evaluated. The removal mechanisms were explored by comparing results of anode, cathode, and bipolar electrode substitution experiments. The performance of the process in a scaled-up reactor was investigated to assure the feasibility of the process. Results showed that simultaneous removal of carbonaceous and nitrogenous pollutants was achieved under optimal conditions. Ammonia removal was due to the free chlorine generation of EO while organic matter degradation was achieved by both EO and EC processes. Nitrate removal was attributed to both ER and EC processes, with the higher removal achieved by ER process. In a scaled-up reactor, the EO/EC/ER process was able to remove 50–60% organic matter and 100% ammonia at charge of 1.5 Ah/L with energy consumption of 15 kW h/m^3. Considering energy cost, the process is more efficient to meet the requirement of organic removal efficiency less than 70%. These results show the feasibility and potential of the EO/EC/ER process as an alternative tertiary treatment to achieve the simultaneous removal of organic matter, ammonia, nitrate, and color of leachate.

* Corresponding author.
E-mail addresses: zhql1962@163.com, qlzhao@hit.edu.cn (Q. Zhao).

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1. Introduction

During the process of sanitary landfilling or temporary storage for waste incineration, leachates are produced as a result of rainwater percolation, moisture presence and biological process of the solid wastes [1]. The treatment for landfill leachate is a great challenge because of the leachate complex, refractory, and varied composition [2]. Biological treatments, including anaerobic and aerobic processes, are commonly used as first step to remove most organic pollutants from raw landfill leachate [3]. Then physical-chemical processes are followed to effectively treat the complex biological effluent and deal with the residual pollutants. Such processes include multistage membrane technology [3,4], coagulation/precipitation methods [5,6], activated carbon filtration process [7], and advanced oxidation processes [7,8].

Electrochemical treatment has been successfully applied in complex wastewater treatment and has attracted much attention due to its environmental compatibility, high removal efficiency, easy operation, and possibility for automation [9,10]. Among the electrochemical technologies, the most studied for the treatment of landfill leachates are EO and EC. Previous studies investigated the behavior of EO for landfill leachate on the removal of organic compounds, color, and ammonia nitrogen [11–13]. Anodic oxidation in EO using different anode materials such as dimensional stable anode (DSA, Ti based metal coated with Ru, Ir or Rh oxides) [11,14,15], boron-doped diamond (BDD) [16,17], thin film oxides (PbO_2, SnO_2) [13,17], and carbon-based electrode [2,18], can achieve direct and indirect oxidation of organics and ammonia by active chlorine species (ClO^-, HClO, and Cl_2) or hydroxyl radicals (·OH) [12]. Apart from anode materials, research has also focused on the effect of operating parameters, for example chloride concentration, current density, electrolysis time, initial pH, and role of additional electrolytes [11–13]. Owing to its high efficiency and versatility, EO has been applied as pretreatment and post treatment technology for raw leachate and its biological effluent [2,19]. The EO as pretreatment process was evaluated to improve the biodegradability of sanitary landfill leachate and achieved an increase of the ratio between biochemical oxygen demand (BOD) and chemical oxygen demand (COD) (BOD/COD) from 0.3 to 0.88 [20]. Turro et al. used EO for the treatment of stabilized landfill leachate and obtained 90% COD, complete color, and total phenol removal under the optimal operating conditions [11]. Del Moro et al. investigated the comparison of EO as a standalone and as post treatment process, indicating the economic superiority of EO combined with biofilter [15].

EC is an effective and simple method for treating wastewater, such as landfill leachate [1,5,21], tannery [22,23], and textile industrial effluent [9,24], which are often tainted with color substances, suspended solids, organic, inorganic, and heavy metal pollutants that could be flocculated. In the EC process, the coagulation agents are generated in situ by the EO of "sacrificial electrode", undergo destabilization, and aggregate to form flocs with contaminants [25]. The most widely used anode materials for EC are iron and aluminum, which always hydrolyze to polymeric iron and aluminum hydroxides. Iron electrode material is considered to be more appropriate for EC in leachate tertiary treatment because it is less toxic and more acceptable in the environment [26]. Lacasa et al. also indicated that the cost of EC by iron is lower than that by aluminum [27]. Electro-Flotation (EF) always takes place in EC since hydrogen bubbles are produced at the cathode and some flocculated pollutants are floated by natural buoyancy to the surface. Important operating factors influencing the EC performance include inter-electrode distance, mechanical stirring, current density, pH, and conductivity [1,9,28]. The EC processes have been shown high effectiveness not only in the removal of color [26], refractory organic matter [5], and heavy metals [23], but also in the elimination of inorganic contaminants, such as nitrate [27,29] and phosphates [27]. The feasibility of EC on treating leachate was examined, achieving high removal efficiency for COD, turbidity, color, UV_{254}, and 40% nitrate removal, but the process was found not appropriate for the removal of ammonia from leachate [21]. Besides EC, the ER process is also popular for nitrate removal [30,31]. In the ER process, electrons from the cathode surface react with nitrate to form intermediate nitrite [32] and nitrogen gas as the final product [33]. In our previous study, nitrate removal with graphite felt (GF) as cathode in dual-chamber cell showed high performance and broad application prospects [34].

In recent years, the combination of electrochemical methods has the potential to eliminate various contaminants and enhance the treatment efficiency [35]. The significance of this work is to explore an integrated electrochemical process that employs iron bipolar electrode and evaluate its performance as an alternative treatment process for the removal of both carbonaceous and nitrogenous contaminants in landfill leachate. There are only few articles in the literature that deal with topics related to bipolar electrochemical system. Mameri et al. reported the use of bipolar electrodes resulted in increase of the anode area and reduction in electrolysis time [36]. Various bipolar electrodes with different forms of anodes were applied to expand the capabilities of electrochemical processes by various researchers. The continuous aluminum bipolar ECEO-EF reactor was designed and evaluated for simultaneous removal of ammonia and phosphate for wastewater effluent [37]. Llanos and Cotillas carried out an integrated electro-disinfection/EC process with iron or aluminum bipolar electrode to remove *Escherichia coli* and turbidity for urban wastewater treatment and reuse [38,39]. Peroxielectrocoagulation/EO/EF process was constructed with aluminum bipolar electrodes for effective removal of organic pollutants from the olive mill wastewater and achieved increase in biodegradability index (BOD/COD) from 0.29 to 0.46 [40].

In our previous studies, we investigated the performance of ER on nitrate reduction [34] and an integrated EO/ER process on simultaneous removal of ammonia, nitrate, and coliforms [41]. Motivated by our previous results, an EO/EC/ER process, consisting of DSA as anode, GF as cathode, and perforate iron plate as bipolar electrode, was developed and applied for the treatment of actual (field) leachate biological effluent in this study. The design of bipolar electrode between anode and cathode could achieve simultaneous removal of residual COD, ammonia, and nitrate in the leachate. To the authors' best knowledge, this integrated process of EO/EC/ER has not yet been reported. The aim of this work was to evaluate the performance of this EO/EC/ER process for the removal of organic and nitrogen contaminants. Performance evaluation included the effects of several parameters, such as chloride concentration, pH, type of anode materials, inter-electrode gap, and flow velocity. The removal mechanism of this process was explored by comparing results of four parallel experiments. In addition, the feasibility of this integrated process was evaluated in a scaled-up reactor on the basis of pollutants removal and energy consumption.

2. Materials and methods

2.1. Experimental set-up and operation

Experiments were conducted in batch mode, with recirculation (Fig.1). BDD or DSA (Ir-Ru oxide cover, DSA_1 or Ir-Ta-Sn oxide cover, DSA_2) were used as anode, GF or titanium net (Ti) was served as cathode, while iron net (Fe) was introduced between anode and cathode as bipolar electrode. The BDD used was silicon based electrode and was purchased from Neocoat SA, Switzerland. The perforated iron electrodes were polished and cleansed in distilled water before use. Details of DSA, GF and titanium electrodes are available in the literature [34,41]. A potentiostat (Zhaoxin Corp., JPS-3005, 0–30 V, China) supplied electric power for this process. The design of the experiments is shown in Table S1. The electrochemical process was carried out in a 0.12 L reactor with immersed electrode area of 10 cm^2 for model and actual leachate. A scaled-up reactor of 2.8 L volume and the electrode area of 100 cm^2 was applied for actual leachate to evaluate the feasibility of this EO/EC/ER process.