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Degradation of biologically treated landfill leachate by using electrochemical process combined with UV irradiation

Shuhu Xiao [a,b], Jianfeng Peng [a,b], Yonghui Song [a,*], Dongsheng Zhang [b], Ruixia Liu [a,b], Ping Zeng [a,b]

[a] *State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China*
[b] *Department of Urban Water Environmental Research, Chinese Research Academy of Environmental Sciences, Beijing 100012, China*

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ABSTRACT

Leachate originated from landfills, where municipal solid wastes (MSWs) were disposed, was of complex compositions that could have a high environmental impact. The primary aim of this work was to provide a feasible and effective way to remove biorefractory organics and ammonium–nitrogen from landfill leachate by electrochemical process combined with UV irradiation. A nonphotoactive RuO_2/Ti electrode was selected as anode and comparative treatments were investigated. At the natural pH and without addition of electrolyte, about 100% of color and NH_3–N, and 80% of TOC in the leachate were removed with a current density of 50 mA cm^{-2} followed by a final COD reduction from 560 mg L^{-1} to less than 50 mg L^{-1}, which was by far better than single electrochemical process. Moreover, the kinetic analysis also proved that a synergetic effect was observed by combining UV irradiation with electrochemical process in the treatment of landfill leachate. The variations of active chlorine and ESR results together indicated that the *in situ* electrogenerated active chlorine and *in situ* photogenerated radical species in the bulk of solution should be responsible for the synergetic effect. In addition, excitation emission matrix (EEM) and gel permeation chromatography (GPC) spectra indicated that the humic and fulvic substances in the leachate were preferentially degraded.

1. Introduction

Sanitary landfill is still the most popular way for municipal solid wastes (MSWs) treatments in China and some other countries [1,2]. However, the leachate derived from the rainfall, run off of surface drainage and decomposition of organic waste, is a dark grey and foul smelling solution. It possesses high values of biological oxygen demand (BOD) and chemical oxygen demand (COD). Due to its toxic potential, the leachate might represent an environmental problem [3,4]. In an effort to control the pollution caused by landfill leachate, many treatment processes have been studied. Biological treatment processes [5–12], including anaerobic and aerobic processes, were quite effective for young landfill leachate with a high BOD_5/COD. But they failed to treat the old or biologically treated landfill leachate which contained a large percentage of recalcitrant organic molecules. As a result, physico-chemical processes (including coagulation [13], adsorption [14], membrane filtration [15], O_3 [16], Fenton [17], UV/H_2O_2 [18], UV/Fenton [19], photocatalysis [20], electrochemical process [21–23], etc.) may be used for organics abatement as a main or supplementary treatment.

Up to now, electrochemical process has been proved to be promising for wastewater treatment mainly due to its high effectiveness and easy operation. Especially in the case of landfill leachate, which has good conductivity and rich chloride ions [23], the pollutants are destroyed by either direct or indirect oxidation process in the electrochemical process. In a direct anodic oxidation process, the pollutants are first adsorbed on the anode surface and then destroyed by the anodic electro-transfer reaction. While in an indirect oxidation process strong oxidants, such as active chlorine including hypochlorite and hypochlorous acid are *in situ* electrochemically generated. Thus, the organic contaminants and NH_3–N are then degraded in the bulk solution by oxidation reactions of the generated oxidants.

Recently, the combination of electrochemical and photochemical process in the presence of Cl^- has been proven to be more efficient in removing pollutants than individual electrochemical and photochemical process [24,25]. It was reported in our previous study [26] that active oxidants could be efficiently generated and NH_3–N was completely degraded by electrochemical process combined with UV irradiation in the presence of chlorides. Herein, a 10 W low-pressure mercury lamp was introduced to the electrochemical treatment of landfill leachate. In comparison with the electrochemical process, an obvious increase in the removal of color, NH_3–N, TOC and COD was observed in the electrochemical process combined with UV irradiation. The results were expected to

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* Corresponding author. Tel./fax: +86 10 84928380.
E-mail address: songyh@craes.org.cn (Y. Song).

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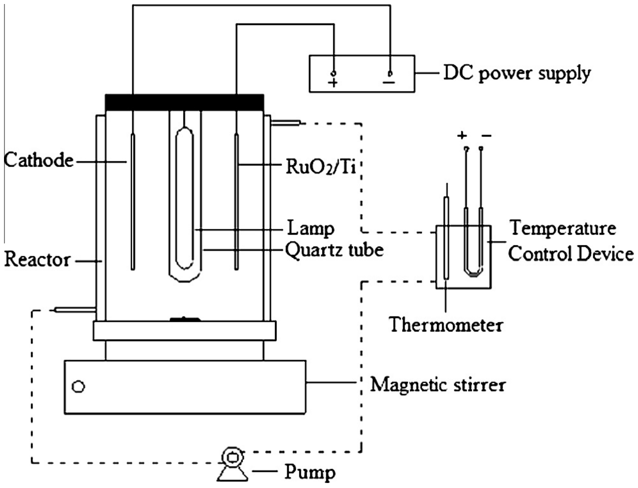

Fig. 1. Schematic diagram of lab scale batch mode photassisted electrochemical oxidation reactor.

provide basic information for the application to post-treat the biorefractory landfill leachate.

2. Experimental

2.1. Apparatus

Electrochemical process (EC), photochemical process (PC), and electrochemical process combined with UV irradiation (PEC) were carried out in a 600 mL, single compartment, glass cell with a 3.5 cm-diameter quartz tube placed in the center and used as the UV bulb housing. The outside of the glass cell was covered with a circulating water system in order to keep the temperature at about 25 °C. The experimental setup was sketched in Fig. 1. The system was controlled by a DC power supply source AMRFL LPS302A (Dahua Instrument Corporation of Beijing). The 60 cm^2 area of non-photoactive RuO_2/Ti mesh (Hengli Ti Corporation of Beijing) was selected as anode, and Ti mesh with the same solid surface area was selected as cathode. The UV irradiation was provided by a 10 W low-pressure mercury lamp (253.7 nm) with the light intensity of 1.36 mW cm^{-2} as measured at distance of about 6 cm from UV lamp by a UV radiometer (Light and Electric Instruments Factory of Beijing Normal University).

2.2. Characteristics of landfill leachate

Leachate samples, gathered into reservoir and pretreated by three processes (anaerobic, aerobic and coagulation), were obtained from a sanitary landfill of Beijing and stored in the refrigerator at 4 °C until experiments. The characteristics of landfill leachate were summarized in Table 1. It was observed that the concentrations of COD and NH_3–N were still a little high and the amounts of removable substrates by more biological treatments might be very small because the biodegradability value (BOD_5/COD) was ca. 0.09. However, the conductivity and the concentration of Cl^- were both high enough. Thus, electrochemical process was just a choice for the leachate treatment.

Table 1
Characteristics of the landfill leachate.

Parameter	Content
pH	7.5–8.0
Conductivity (ms cm^{-1})	14.6–5.0
Alkalinity ($CaCO_3$)	6000–6500
COD	480–640
BOD_5	40–55
TOC	170–200
Color[a]	0.35–0.45
NH_3–N	600–800
Cl^-	4200–5000

Units in mg L^{-1}.
[a] Represented by the absorbance at 400 nm.

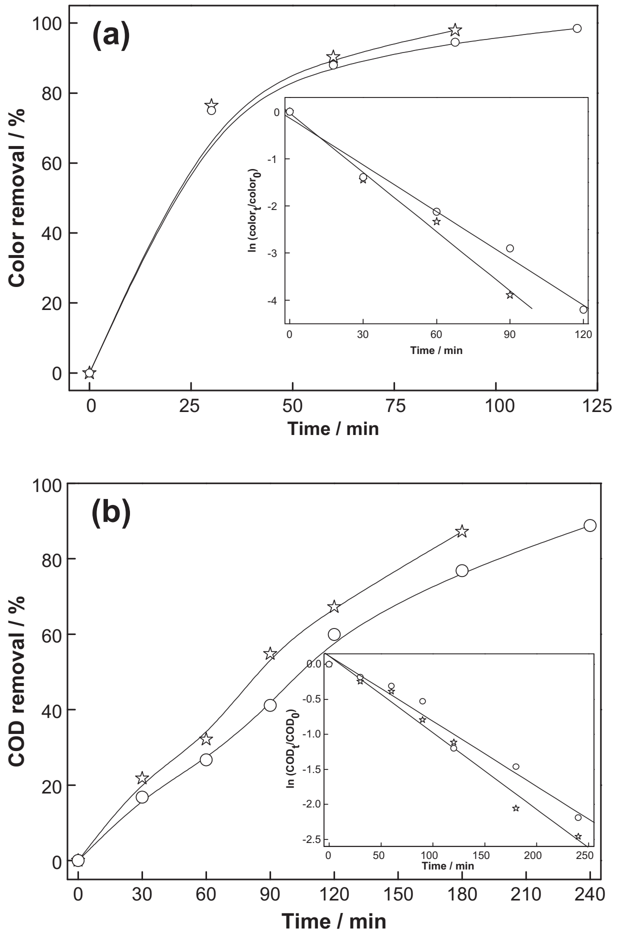

Fig. 2. Kinetic analysis for pseudo first-order removal of (a) color or (b) COD in the EC (○) and PEC (☆) processes. Natural pH, current density 50 mA cm^{-2}.

2.3. Analytical methods

Samples were withdrawn from the reactor at regular time intervals. UV–vis spectra of the samples were recorded between 200 and 600 nm on a U-3010 UV–vis spectrophotometer (Hitachi Co., Japan) equipped with 1 cm quartz cuvette. The pH values of samples were adjusted with NaOH or H_2SO_4 using an Orion 720APLUS Benchto meter (Thermo Orion Co., USA). The discoloration of the solution was achieved by measuring the absorbance at 400 nm [21]. The total organic carbon (TOC) was measured by a multi-N/C 3000 TOC analyzer (Analytik Jena AG Co., Germany) after the solution was filtered through a 0.45 μm filter. The chemical oxygen demand (COD), ammonia–nitrogen and active chlorine were determined according to the standard methods [27]. Fluorescence spectra were recorded on an F-4500 fluorescence spectrophotometer (Hitachi Co., Japan) by measuring the emission spectra in the range from 280 to 550 nm repeatedly at the excitation wavelengths from 220 to 420 nm. Spectra were then converted into an excitation emission