



Enhanced removal performance of estriol by a three-dimensional electrode reactor

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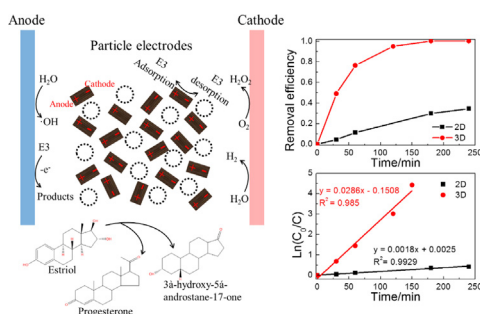
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HIGHLIGHTS

- TDER performed much better in removing E3 than two-dimensional electrode reactor.
- The possible degradation mechanisms of E3 were demonstrated.
- Two new E3 degradation intermediates were observed for the first time.
- Continuous-flow TDER achieved an 80% E3 removal efficiency with proper conditions.

GRAPHICAL ABSTRACT



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ABSTRACT

This study dealt with the treatment of estriol (E3), a typical endocrine disrupting chemical (EDC), using a three-dimensional (3D) electrode reactor (TDER). The reaction rate per unit area of TDER in batch mode was 3.23–5.75 times that of a conventional two-dimensional (2D) electrode reactor, while its energy consumption was only 1/7–1/5 of that of the latter. The kinetics analysis results indicated that the degradation of E3 in both systems followed pseudo first-order kinetics. The effects of initial E3 concentration, current density, electrode distance, and recirculation flow rate on the performance of TDER were investigated in batch mode. A current density of 1.0 mA/cm², electrode distance of 2 cm, and recirculation flow rate of 200 mL/min were identified as optimal operating parameters that could guarantee excellent E3 removal efficiency and energy consumption. Progesterone and 3 α -hydroxy-5 α -androstane-17-one were identified by UPLC/MS/MS and GC/MS for the first time as E3 degradation intermediates. A possible E3 degradation pathway was proposed based on the intermediates and the results of previous studies. The mechanism analysis results identified indirect oxidation as the main contributor to E3 degradation in batch TDER, with the repolarization of particle electrodes contributing to the degradation. Subsequently, TDER was operated in continuous-flow mode to treat a secondary effluent spiked with E3. With a hydraulic retention time of 50 min and a current density of 20 mA/cm², TDER achieved an E3 removal efficiency of 80%, thereby demonstrating the ability of continuous-flow TDER to produce a stable, high quality effluent. The aforementioned results highlighted 3D electrolysis as a promising alternative method for removing trace EDCs from secondary effluent in pretreatment or advanced treatment applications.

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1. Introduction

Over the past decades, the removal of endocrine disrupting chemicals (EDCs) received much research attention because of

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their wide presence in wastewater effluents, surface water, and other aquatic systems as well as their toxicity to aquatic life forms at extremely low levels (0.1 ng/L–10 ng/L) [1]. Among many types of EDCs, estrogens have high potential for disrupting the endocrine of organisms in aquatic ecosystems [2]. Natural estrogens or synthetic hormones, including estrone (E1), 17 β -estradiol (E2), estriol (E3, see Fig. 1) or 17 α -ethinyl estradiol (EE2), and diethylstilbestrol, have been widely detected in the effluents of sewage treatment plants (STPs) worldwide with concentrations ranging from not detected (ND) to 240 ng/L [3,4], most of which were much higher than the toxicity threshold. The continuous release of EDCs in wastewater effluents, which is a main source of reclaimed water, poses a huge risk for both public health and the environment [5]. Therefore, the EDCs in the effluents of STPs must be eliminated before such waste is discharged into the environment to ensure the ultimate safety of reclaimed water and preserve the ecosystem.

Trace organic contaminants are usually removed via conventional biological treatment [6–8], adsorption [9,10], membrane technologies [11], chemical oxidation [12–17], electrochemical methods [18–23], and combined processes [24,25]. Despite yielding promising results, these technologies have reported several drawbacks. For instance, biological methods cannot achieve a satisfactory removal efficiency [6], while ozonation and photocatalytic systems are very expensive and always encounter operational problems [26]. In this sense, electrochemical methods have emerged as promising alternatives for contaminant removal.

Electrochemistry has received increasing research attention because of its simple equipment, easy operations, environmental compatibility, and effectiveness in degrading non-biodegradable pollutants [27]. Electrochemical methods theoretically mineralize pollutants or generate biodegradable intermediates [28]. Among these methods, the emerging three-dimensional (3D) electrolysis technology addresses the problems encountered by conventional two-dimensional (2D) electrolysis, including its low current efficiency and mass transfer limitations [29]. 3D electrolysis can also achieve high electrolytic efficiency and low energy consumption by providing numerous micro-electrolysis cells as expanded reaction areas and promoting the electro-generation of oxidants (e.g., hydroxyl radicals [30], active chlorine [31], or S₂O₈²⁻ [32]). Therefore, 3D electrolysis has a great potential for removing trace organic pollutants in solutions with low conductivities.

3D electrolysis has been successfully applied in various types of wastewater treatment [33–36]. Nevertheless, most related works have focused on treating high concentrated organic wastewater with pollutant or COD concentrations ranging from mg/L to g/L before treatment, only few studies have examined the removal of trace-level pollutants, especially in secondary effluents, for water reuse purposes. Sakakibara et al. [37,38] published several studies on the removal of trace estrogens using a 3D electrode reactor (TDER) packed with Ti/Pt particles and on the regeneration of particle electrodes via ozonation and Fenton reaction. However, without indirect oxidation and particle repolarization, the treatment time may be extended for several days. Therefore, highly efficient 3D electrolysis technologies for treating trace EDCs must be developed, and the effects of the operating parameters, degradation

kinetics, and mechanisms in 3D electrolysis must be examined further.

This study focuses specifically on the treatment of E3. Given the high water solubility of estrogens as well as the metabolite and breakdown product of E1 and E2, E3 was often detected at higher levels and more frequently than E1, E2, and EE2 in both the influents and effluents coming from municipal wastewater treatment plants (WWTPs) [6,39]. However, most studies on the removal of estrogenic EDCs have focused on E1, E2, and EE2 [19–21], while only few have examined the degradation of E3 [2] and the related degradation mechanisms. The destruction of E3 in aquatic environment has also attracted limited research attention. Therefore, the removal characteristics of E3 warrants further study.

Based on the above studies and considerations, this work investigated the use of TDER for the electrochemical degradation of E3 and the treatment of secondary effluents. The kinetics of E3 degradation and the effects of several operating parameters, including initial E3 concentration (C₀), electrode distance, current density (j), and recirculation flow rate (Q), on E3 degradation were evaluated in batch mode. The potential mechanisms and possible intermediates of E3 degradation were proposed. TDER was then applied to treat real secondary wastewater spiked with E3 in continuous-flow mode to test its feasibility for practical applications.

2. Materials and methods

2.1. Chemicals and reagents

An E3 (Sigma–Aldrich, $\geq 97\%$) stock solution with a 1 g/L concentration was prepared by dissolving 0.1 g E3 in a 100 mL 80% methanol aqueous solution (methanol:water = 4:1 by volume). Methanol (99.9%), acetonitrile (99.9%), acetone (99.3%), and other organic solvents were of chromatographic grade and obtained from J.T. Baker, while all the other chemicals (e.g., Na₂SO₄, NaOH, and H₂SO₄) were of analytical grade and purchased from Sinopharm Chemical Reagent Co., Ltd. (Beijing, China). These chemicals were used directly without further purification. All solutions were prepared with Milli-Q water (≥ 18.2 M Ω ·cm) or deionized (DI) water.

2.2. Reactor configuration

Batch 3D electrolysis was performed in an undivided rectangular electrolysis reactor made of Plexiglas with an effective volume of 300 mL. Fig. S1(a) shows the schematic diagram of the batch TDER configuration. Recirculation batch type was used to facilitate the mass transfer. A Ti/IrO₂–RuO₂ plate and a pure Ti plate (5 × 10 cm, Longsheng Nonferrous Metal Co., China) were used as anode and cathode, respectively. The distance between the anode and cathode could be adjusted according to requirements. The space between the main electrodes was filled with approximately 50 g granular graphite particles (2 mm–3 mm in diameter and 5 mm–6 mm in length) and 70 g glass beads (2 mm–3 mm in diameter) with a volume ratio of 1:1. These materials were purchased from Shenzhen Xieli Graphite Material Co., China and used as the third electrode. To reduce the short circuit current and improve the repolarization of the particle electrodes, the particles were mixed evenly via manual vibration in a plastic basket before being packed to make the glass beads well distributed between the graphite granules. After each test, the particle electrodes were ultrasonic processed and washed several times before reuse. All electrochemical processes were performed in galvanostatic mode using a digital direct current (DC) power supply (DC 32V 2A, UNI-T UTP3703, China).

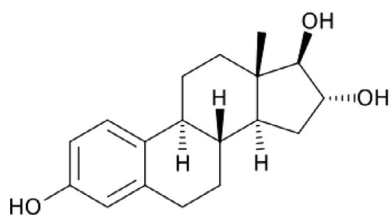


Fig. 1. Chemical structure of estriol.

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