



Efficient mineralization of the antibiotic trimethoprim by solar assisted photoelectro-Fenton process driven by a photovoltaic cell



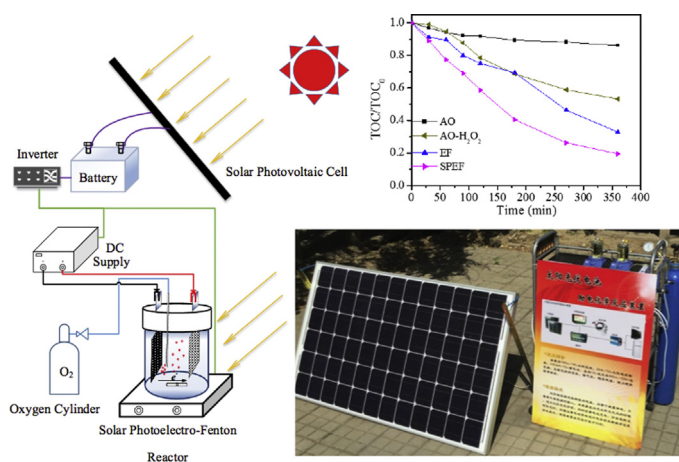
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HIGHLIGHTS

- A novel photovoltaic solar photoelectro-Fenton (SPEF) system was developed.
- Efficient mineralization of trimethoprim (TMP) was achieved.
- Aromatic intermediates, carboxylic acids and inorganic products were detected.
- A comprehensive TMP degradation pathway in SPEF process was proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, a novel self-sustainable solar assisted photoelectro-Fenton (SPEF) system driven by a solar photovoltaic cell was developed for the efficient mineralization of antibiotic trimethoprim (TMP) in water. A comparative degradation of 200 mg L⁻¹ TMP by RuO₂/Ti anodic oxidation (AO), anodic oxidation with H₂O₂ electrogeneration (AO-H₂O₂), electro-Fenton (EF) and SPEF was investigated. SPEF was proved to exhibit the highest oxidation power, i.e., more than 80% TOC was removed after 360 min SPEF treatment of 200 mg L⁻¹ of TMP under optimal conditions at pH 3.0, 1.0 mM Fe²⁺ and 18 mA cm⁻². Influences of current density, pH, initial Fe²⁺ and initial TMP concentration on SPEF process were also studied. Ten aromatic intermediates generated from hydroxylation, carbonylation and demethylation reactions were identified using UPLC-QTOF-MS/MS system during the SPEF treatment, together with three carboxylic acids (oxamic, oxalic and formic acids) and two inorganic ions (NH₄⁺ and NO₃⁻) measured. Therefore, a reasonable pathway of TMP degradation in SPEF process was proposed.

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1. Introduction

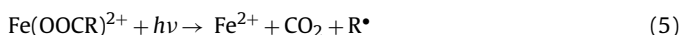
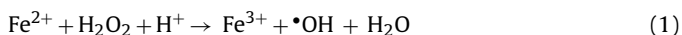
Usually taken as a potentiator to increase other antibiotics' antibacterial activity (e.g., sulfamethoxazole), trimethoprim (TMP) is one of the most important antibiotics commonly used for the

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treatment of urinary, respiratory or gastro tract infections for both humans and animals [1–3]. TMP is incompletely metabolized during human therapy and about 80% of TMP is excreted in its pharmacologically active form, leading to its subsequent discharge into municipal sewerage [4]. Since TMP cannot be efficiently removed through conventional biological processes in wastewater treatment plants (WWTPs), it has been detected in surface water and secondary effluents released from WWTPs at concentrations ranging from 0.003 to 4.30 $\mu\text{g L}^{-1}$. Even at trace concentration, it may cause negative effects for increasing the risk of bacterial resistance [5–7]. Therefore, there is an urgent need to develop more efficient treatment approaches for the degradation of TMP to avoid the potential adverse effects on the aqueous environment.

Recent studies have proved that advanced oxidation processes (AOPs) are promising alternative techniques for TMP removal [8]. For example, Kim et al. found that 5 mg L^{-1} TMP was completely removed within 20 min under 8.9 W electric barrier discharge plasma treatment [9]. Abellán et al. observed that 50% of initial total organic carbon (TOC) was removed with TiO_2 dose higher than 0.5 g L^{-1} when 100 mg L^{-1} TMP was treated using photocatalytic process for 360 min [10]. Li et al. studied the elimination of pharmaceuticals and personal care products (PPCPs) including 2 mg L^{-1} TMP using Fenton treatment, and up to 30% TOC decay was obtained under a constant $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ molar ratio of 2.5 [11]. Electro-Fenton (EF) was also successfully applied to treat the TMP wastewater, which was based on the continuous generation of $\cdot\text{OH}$ via Fenton's Reaction (1) in acidic aqueous medium [12], where H_2O_2 was supplied from two-electron reduction of dissolved oxygen (Eq. (2)) and Fe^{2+} was regenerated from one-electron reduction (Eq. (3)) [13]. Mansour et al. investigated the combined EF process with activated sludge treatment for the degradation of 0.2 mM TMP, where complete TMP removal was achieved at 0.69 mM Fe^{2+} , 466 mA for 30 min electrolysis. Although only 12% and 21% TOC were removed after 30 min and 60 min treatment, the biodegradability was significantly improved [14]. More recently, Moreira et al. examined the comparative degradation of 20 mg L^{-1} TMP by EF, photoelectro-Fenton (PEF) and solar photoelectro-Fenton (SPEF) processes, which were performed in a lab-scale flow plant equipped with compound parabolic collectors (CPCs) and an electrochemical filter-press cell with a boron-doped diamond (BDD) anode and a carbon-PTFE air-diffusion cathode. The relative oxidation ability increased in the order: $\text{EF} < \text{PEF} < \text{SPEF}$ [15]. The higher oxidation ability of SPEF owing to the more potent UV intensity supplied by sunlight was explained by the following reactions [16–21]: (i) the photolysis of photoactive Fe(III)-hydroxyl complexes like $\text{Fe}(\text{OH})^{2+}$, generating Fe^{2+} and $\cdot\text{OH}$, and (ii) the quick photodegradation of Fe(III)-carboxylate intermediates via Eqs. (4) and (5).



Although SPEF process was considered as a cost-effective process, energy still had to be supplied for maintaining the electrolysis [22–24]. The application of photovoltaic cell might be a suitable way for further reduction of energy consumption for renewable sunlight was used as energy source [25–28]. For example, Figueroa et al. [29] found that the EF process coupled with a solar photovoltaic cell could efficiently mineralize 0.061 mM Reactive Black 5 (115 mg L^{-1} COD) with 90% of initial COD removed and 0.1 mM Acid Green 25 (62 mg L^{-1} COD) with 80% COD removed after 2.33 h treatment, as well as industrial sample (230 mg L^{-1} COD) with 50% COD

removed after 7.5 h treatment. Garcia-Segura et al. [30] studied the degradation of Direct Yellow 4 (DY4) diazo dye using a combined process of SPEF and photovoltaic cell, in which the EF and the solar photo-Fenton reactions were conducted separately, where photo-Fenton reaction took place in solar CPCs and the solutions were recirculated by a pump. 96–97% TOC reduction was achieved when 0.32 mM DY4 was treated by SPEF process at pH 3.0, 5.0 A and 0.50 mM Fe^{2+} .

In this paper, a combined SPEF system coupled with photovoltaic cell was developed for treating 200 mg L^{-1} TMP in a quartz reactor equipped with a RuO_2/Ti anode and an activated carbon fiber (ACF) cathode. Besides, a battery system charged by solar cells was used for powering the DC power supply and the magnetic stirrer. The degradation kinetics and mineralization ability of SPEF process were explored by conducting comparative degradation by anodic oxidation (AO), anodic oxidation with H_2O_2 electrogeneration (AO- H_2O_2), EF and SPEF; the effects of current density, pH, initial Fe^{2+} content and initial TMP concentration on the mineralization rate of SPEF were also assessed. The intermediates generated in SPEF process were identified by ultra-performance liquid chromatograph (UPLC) coupled to a quadrupole-time-of-flight mass spectrometer (QTOF-MS/MS) and a general TMP degradation pathway was proposed.

2. Experimental

2.1. Chemicals

Trimethoprim ($\text{C}_{14}\text{H}_{18}\text{N}_4\text{O}_3$, MW 290.32, >98%) provided by Tokyo Chemical Industry was employed without further purification. Analytical grade anhydrous sodium sulfate, heptahydrated ferrous sulfate, sulfuric acid and sodium hydroxide were obtained from Sinopharm Chemical Reagent Co. Ltd., China. Solutions were prepared with water generated from a Millipore Synergy UV system, with a conductivity of 18.2 $\text{M}\Omega\text{ cm}$ at 25 °C. HPLC grade acetonitrile and formic acid were obtained from Fisher Scientific. The ACF felt (BET 1237 $\text{m}^2\text{ g}^{-1}$) was purchased from Shandong Xuesheng Technology Co. Ltd., China, and its surface properties like BET area, total pore volume and pore size had been provided in our previous paper [31].

2.2. Experimental system

The SPEF treatment system consisting of the SPEF unit and the photovoltaic power unit is displayed in Fig. 1. In SPEF unit, a 125 mL cylindrical quartz cell with a coupled quartz lip was used, which could collect the direct solar radiation well. In the AO process, the anode and cathode were both 4 cm \times 5 cm RuO_2/Ti mesh electrodes, while in the anodic oxidation with electrogenerated H_2O_2 (AO- H_2O_2), EF and SPEF conditions, a 4 cm \times 5 cm ACF felt was attached to one of RuO_2/Ti mesh electrodes to act as cathode. Prior to each electrolytic trial, the ACF felt was immersed in 200 mg L^{-1} TMP solutions for several times to preclude the adsorption effect of TMP on ACF felt. The SPEF experiments were conducted from 9:00 to 15:00 h in sunny and clear days during the summer (from July to September) of 2014 in our laboratory of Beijing, China (latitude 39°57'09"N, length 116°21'14"E). The photovoltaic power unit incorporated a UD 100-12 battery (12V100Ah, Santak Kexing Electronic Co. Ltd., China) and a solar cell panel (size 1250 \times 750 \times 40 mm, Guangzhou Z.T. Solar Technology Co. Ltd., China) with solar to electrical energy efficiencies of 18% under 40,000 lx. Working voltage and currents were 18 V and 8.33 A, respectively. The panel was made up of 72 mono-crystalline silicon solar cells; connected in 12 rows and 6 columns, with an active surface area of 12 \times 10 cm^2 per cell. The inclination of the solar

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