



Exploring the advanced oxidation/reduction processes in the VUV photoreactor for dechlorination and mineralization of trichloroacetic acid: Parametric experiments, degradation pathway and bioassessment



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HIGHLIGHTS

- Simultaneous advanced oxidation/reduction of TCAA in VUV process.
- Complete degradation and efficient mineralization of TCAA in VUV process.
- Complete dechlorination of TCAA in VUV process.
- Efficient detoxification of TCAA in the VUV process as compared with that in UVC.

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ABSTRACT

The present study reports degradation, dehalogenation and mineralization of trichloroacetic acid (TCAA) exploring advanced oxidation/reduction processes (AORPs) in a chemical-less VUV reactor. The performance of VUV process was almost independent of solution pH. The complete degradation and dechlorination of TCAA up to initial concentration of 50 mg/L achieved in the VUV process at a neutral solution pH within a relatively short reaction time of 30 min. The observed first-order degradation rate of TCAA increased from 2.35 to 12.20 mg/L.min with the increase of initial concentration from 1 to 100 mg/L. The major water anions did not considerably influenced the decomposition of TCAA. The hydroxyl radical and hydrated aqueous electrons simultaneously contributed in degradation and dechlorination of TCAA in the VUV process. Around 87% of TCAA (10 mg/L) was mineralized in the VUV process with 30 min. The degradation intermediates determined using LC/MS were found to be simple non-chlorinated substances mainly formic and acetic acids. A low energy was consumed in VUV to attain the efficient decomposition of TCAA. The VUV process was efficiently operated in continuous-flow mode for degradation, dechlorination and mineralization of TCAA. Therefore, the VUV photoreactor is an emerging and feasible chemical-less AORP which can be efficiently operated in continuous-flow regime for high-rate decomposition and mineralization of halogenated organic compounds.

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1. Introduction

Due to its low cost, high efficacy, and simple operation, chlorination is one the most common method of choice for disinfection of drinking water in the world [1]. However, the chlorinated disinfection by-products (DBPs) formed in the chlorination process from the reaction between free chlorine (HOCl and OCl⁻) and organic matters in water is a growing concern because of their strong health effects. It is well known that the chlorination of waters containing organic matters produces chloroacetic acids (CAAs). CAAs

are very toxic and known as carcinogenic compounds for human; the carcinogenic risk of CAAs is 100 times higher than that of trihalomethanes [2]. The human uptake CAAs mainly through consumption of the contaminated water [3,4]. The presence of CAAs in the water supplied for drinking is of great public health concern. The WHO has set 0.2 mg/L as the guideline value for trichloroacetic acid (TCAA) in drinking water [5]. Therefore, to protect the human health against the carcinogenic CAAs, the contaminated waters must be efficiently treated for these compounds, either in point-of-use or in point-of-entry, before drinking. TCAA is also used in various industrial applications as an etching, a pickling agent for surface treatment of metals, a swelling agent and solvent in the plastic industry, an auxiliary in textile finishing, and as an additive

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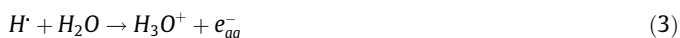
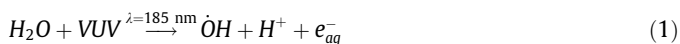
to improve high-pressure properties in mineral lubricating oils [6]. Therefore, these industries may have high concentrations of TCAA in their effluent required to be treated before discharging to the environment.

Advanced oxidation processes (AOPs) have recently emerged as a significant class of technologies for accelerating the oxidation and destruction of a wide range of organic contaminants in the contaminated water. In the classical AOPs, highly reactive hydroxyl radicals ($\cdot\text{OH}$) generated from the chemical, photochemical, ozone- and photo-catalytic, and electrochemical homogeneous or heterogeneous processes are the main oxidative specie contributed in the oxidation/degradation mechanisms of organic contaminants [7]. To date, the degradation of CAAs has been studied in some of the classical AOPs including photolysis [8], UVC/ $\text{K}_2\text{S}_2\text{O}_8$ [9], O_3/UV , $\text{H}_2\text{O}_2/\text{UV}$, $\text{O}_3/\text{H}_2\text{O}_2$, $\text{O}_3/\text{H}_2\text{O}_2/\text{UV}$ [10], Fenton/ UV/TiO_2 [11], format/ UV/TiO_2 [12] and $\text{UV}_{400}/\text{TiO}_2$ [13] AOPs. Although these processes reported to some extent efficient degradation of MCAA and DCAA, the dechlorination of TCAA was slow and incomplete [10]. In addition, in chemical-based AOPs including those examined for CAAs degradation, the $\cdot\text{OH}$ is typically generated at the presence of externally added chemicals/catalysts. Need the external additives make the AOPs cost-intensive, complex to operate due to needing the continuously regulating amount of additives (chemicals/catalysts), and need to efficient separation of the chemical residuals or spent catalysts from the treated water or off gasses (in ozone based AOPs). Therefore, these critical defects impede the application of the chemical-based AOPs for advanced treatment of drinking water.

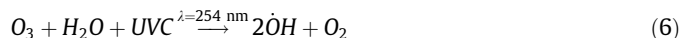
Another considerable point in elimination of CAAs from water using chemical-based AOPs is that for achieving complete degradation of these compounds some bonds need to be oxidized (e.g., C—C) while some others should be reduced (C—Cl). It means that both reductive aqueous electrons and oxidative have to be simultaneously presented in the reaction medium.

In the other word, chemicals-less process, high reaction rate (efficient in a relatively short reaction time), low energy consumption, production of non-hazardous residues for organism and the environment, and applicability are the main important criteria in selection and use of a specific control system [14,15]. To cope with above-mentioned criteria, advanced oxidation/reduction processes (AORPs) have been recently emerged as novel technologies. In AORPs the reactive oxidative ($\cdot\text{OH}$) and reductive (e_{aq}^-) species are concurrently generated and involved in the oxidation and reduction reactions [12,16,17] for accelerating the destruction of a wide range of contaminants including very toxic DPBs as CAAs.

Among them, the vacuum UV (VUV; 185 nm + 254 nm) is recently developed as a chemical less AORP in which both $\cdot\text{OH}$ and e_{aq}^- are simultaneously generated from photoionization (Eq. (1)) and homolysis (Eqs. (2) and (3)) of water molecules with high-energy VUV photons absorbed in water [12,16,17].



In addition, photolysis of oxygen dissolved in water by VUV photons leads to the formation of ozone (Eqs. (4) and (5)) [17,18], which in turn can contribute in the generation of extra $\cdot\text{OH}$ (Eq. (6)).



It is found from the above-mentioned reactions that high concentrations of oxidative radicals ($\cdot\text{OH}$) and reductive electrons (e_{aq}^-) are simultaneously generated in the VUV reactor without addition of external chemical/catalyst [16,18]. It allows exploring the AORPs, which are necessary to attain the complete degradation of halogenated organic contaminants [19,20]. It allows operating the VUV process more efficient, cost-effective and environmentally benign as compared with the conventional AOPs for degradation of halogenated contaminants [21].

The purpose of this study was to evaluate the feasibility of VUV photoreactor for degradation, dechlorination and mineralization of CAAs as the main groups of toxic DBPs exploring the AOP in simultaneous with advanced reduction process (ARP). TCAA was selected as the model of CAAs because of its profoundly detected in waters, its insufficient degradation in classical AOPs as well as its high solubility in water that is one of the main reasons for its high dispersion in the aquatic environment [2,8,10,22].

Accordingly, influence of the main operational parameters including solution pH, initial TCCA concentration, reaction time, presence of electron and radical scavengers, water anions, and continuous-flow operation was investigated on the degradation and dechlorination of TCAA. The kinetic of TCCA mineralization under optimum operational conditions was also examined. The degradation pathway and final products were also investigated.

2. Materials and methods

2.1. VUV photoreactor experimental set-up and procedure

All the experiments were conducted in a lab-scale photoreactor. The experimental set up is shown in Fig. 1. A tubular glass photoreactor was used in this study with an internal diameter of 25 mm and a height of 400 mm having a working volume of 100 mL. The effective length of the flow path of 350 mm. A synthetic quartz sleeve with the outer diameter of 15 mm was inserted in the photoreactor cell. A 5.7 W dichromatic low-pressure mercury UV lamp emitting UV at two distinct wavelengths of 254 nm (UVC; 90% with the flux of $54\ \mu\text{W}/\text{cm}^2$ at 1 cm from the lamp surface) and 185 nm (VUV; 10% with the flux of $5\ \mu\text{W}/\text{cm}^2$ at 1 cm from the lamp surface) purchased from Heraeus Co. was installed at the center of the sleeve. For the UVC experiments, the VUV lamp was replaced with UVC one (at the same power) emitting UV at only wavelength of 254 nm. The photoreactor was wrapped with an aluminum sheet to prevent the emission of UV radiation into the lab. All chemical used in the experiments were of analytical grade.

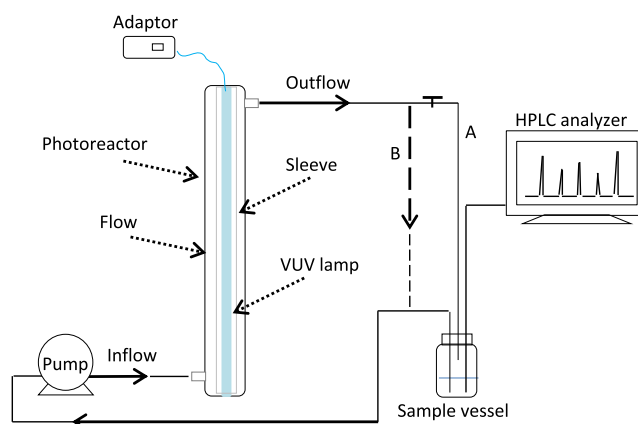


Fig. 1. The schematic of VUV experimental setup.

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