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Homogenous VUV advanced oxidation process for enhanced degradation and mineralization of antibiotics in contaminated water



Mojtaba Pourakbar, Gholamreza Moussavi*, Sakine Shekoohiyan

Department of Environmental Health Engineering, Faculty of Medical Sciences, Tarbiat Modares University, Tehran, Iran

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ABSTRACT

This study was aimed to evaluate the degradation and mineralization of amoxicillin (AMX), using VUV advanced process. The effect of pH, AMX initial concentration, presence of water ingredients, the effect of HRT, and mineralization level by VUV process were taken into consideration. In order to make a direct comparison, the test was also performed by UVC radiation. The results show that the degradation of AMX was following the first-order kinetic. It was found that direct photolysis by UVC was able to degrade 50 mg/L of AMX in 50 min, while it was 3 min for VUV process. It was also found that the removal efficiency by VUV process was directly influenced by pH of the solution, and higher removal rates were achieved at high pH values. The results show that 10 mg/L of AMX was completely degraded and mineralized within 50 s and 100 s, respectively, indicating that the AMX was completely destructed into non-hazardous materials. Operating the photoreactor in continuous-flow mode revealed that 10 mg/L AMX was completely degraded and mineralized at HRT values of 120 s and 300 s. It was concluded that the VUV advanced process was an efficient and viable technique for degradation and mineralization of contaminated water by antibiotics.

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1. Introduction

The emerging contaminants are increasingly threatening drinking water quality throughout the world (Bagheri and Mohseni, 2015; Van Doorslaer et al., 2014). Various emerging contaminants such as pharmaceutical drugs, endocrine disruptors, polycyclic hydrocarbons and surfactants are found in aquatic environments and treated wastewater. Among the emerging contaminants, pharmaceutical compounds are of great importance due to their diversity and wide applications for both human and veterinary (Dorival-García et al., 2013). High detection frequency in the environment and the increasing bacterial resistance formation are the two main features associated with antibiotics, therefore there is a focus point of research on antibiotics removal from contaminated waters (Van Doorslaer et al., 2014). In the recent years, the residues of antibiotics in the environment have led to the growing international concern.

To protect human and environment health against the emerging contaminants, they must be removed from the contaminated water (Moussavi et al., 2015a). The advanced oxidation processes (AOPs) are among the promising methods with a good removal efficiency for the most classes of pharmaceuticals (Verlicchi et al.,

2015). The OH is the primary oxidant generated in the AOPs by chemical, photochemical or electrochemical methods (Verlicchi et al., 2015; Yahya et al., 2014) which reacts with many organic contaminants in water and converts them into more hydrophilic compounds. Different AOPs have been developed and studied so far for the removal of antibiotics in water. These include UV-oxidation (Sørensen et al., 2015), photocatalysis (Lofrano et al., 2016; Sarkar et al., 2015), $\text{UV}/\text{H}_2\text{O}_2$ (Jung et al., 2012; Zuurro et al., 2014), UV/O_3 (Liu et al., 2014), $\text{H}_2\text{O}_2/\text{O}_3$ (Hamdi El Najjar et al., 2013; Lin et al., 2009), $\text{UV}/\text{H}_2\text{O}_2/\text{O}_3$ (Qin et al., 2015), catalytic ozonation (Moussavi et al., 2015a), Fenton reagent and its derivatives (Santos et al., 2015; Yahya et al., 2014), sonolysis (De Bel et al., 2009) and wet air oxidation (Li et al., 2013).

With the increased use of UV radiation in drinking water treatment, much attention has been placed on developing UV-based AOPs for the removal of pollutants from water. $\text{UV}/\text{H}_2\text{O}_2$, as the most widely studied UV-based AOP, is considered as an effective process for decomposition of organic contaminants and has been applied in the full-scale drinking water treatment plants (Kruithof et al., 2007). The majority of UV-based AOPs are dependent on the external chemical addition, which could be considered as a limiting factor for their full-scale application.

The vacuum ultraviolet (VUV) irradiation is a new class of the chemical-less AOP in which the radical species are generated from the photolysis of the water molecules (Gonzalez and Braun, 1995; Imoberdorf and Mohseni, 2011b; Moussavi et al., 2014).

* Corresponding author.

E-mail address: moussavi@modares.ac.ir (G. Moussavi).



In addition, the photolysis of molecular oxygen dissolved in water by VUV photons produces ozone (Eqs. (3) and (4)) that further contribute in removal of contaminants (Moussavi et al., 2014).



Accordingly, the reactive oxidizing species can be produced in the VUV process without need to any external chemicals making the process simple to construct and operate. The efficacy of VUV process has been investigated for oxidation of a few natural and synthetic organic compounds (e.g., Bagheri and Mohseni, 2015; Imoberdorf and Mohseni, 2011a; Moussavi et al., 2014). Although, to the best of our knowledge, no reports could be found on investigating this technique for oxidation of antibiotics in contaminated water.

In order to investigate the efficacy of VUV irradiation on antibiotics removal, amoxicillin (AMX) was selected in the experiment. AMX is a drug belonging to the class of β -lactam antibiotics that has a broad spectrum against both gram-negative and gram-positive bacteria. Almost 65% of the global antibiotic market is belonging to β -lactam antibiotics (Githinji et al., 2011). In β -lactam group, AMX is one of the most widely used drug (Marlière et al., 2000) which is widespread in the aquatic environment including wastewater. It is reported that AMX concentration in the effluent of pharmaceutical manufacturing plants may reach as high as 170 mg/L (Chen et al., 2011). Therefore, the effective removal of AMX from the industrial wastewater is of great importance from the environmental protection point of view. Several studies have been conducted for AMX removal including AOP (Jung et al., 2012), membrane filtration (Li et al., 2004) and adsorption process (Moussavi et al., 2015a).

In the present study, we investigated the efficacy of VUV process as a new class of AOP as compared with the UVC for the oxidation and mineralization of AMX under different experimental conditions. The VUV process was also tested in the continuous mode for oxidizing AMX in both distilled water and tap water.

2. Materials and methods

2.1. Experimental setup

Experimental tests were conducted in an annular photoreactor operated in both batch and continuous mode. The reactor was made of Pyrex with an internal diameter of 25 mm and a height of 400 mm into which a quartz sleeve with diameter of 15 mm was longitudinally inserted at the axial center. For the VUV experiments, a 5.7 W low-pressure mercury UV lamp (Heraeus Co.) emitting radiation at 254 nm (flux of 56 $\mu\text{W}/\text{cm}^2$ at 1 cm from lamp surface) and 185 nm (< 10%; flux of 5 $\mu\text{W}/\text{cm}^2$ at 1 cm from lamp surface) was installed inside the sleeve. The same structure was used for UVC tests with a 9 W low-pressure mercury lamp (OSRAM Co.) emitting with a dominant wavelength of 254 nm (flux of 60 $\mu\text{W}/\text{cm}^2$ at 1 cm from lamp surface). The photoreactor was wrapped with aluminum foil to prevent the emission of UV radiation into lab.

2.2. Experimental procedure

The AMX stock solution was prepared by dissolving 1 g of AMX powder obtained from a local pharmaceutical manufacturing plant in 1 L of distilled water. The synthetic working solution was prepared at initial concentrations of 10, 25, 50 and 100 mg/L by sufficient diluting the aliquots of the stock solution in distilled water. Then the pH of the solution was regulated at the desired value using 1 N NaOH or HCl. The photoreactor was operated in an up-flow mode in which the AMX solution entered at the bottom of the reactor by a peristaltic pump, flowed upward and exited from the top of the reactor. The reactor was operated both in batch and continuous mode. In the batch experiment, the effluent was recycled into the influent tank. The effects of solution pH, initial AMX concentration, water ingredients and the presence of *tert*-butanol as a radical scavenger on the degradation of AMX were evaluated under batch operation of the reactor. For the batch experiments, 170 mL of AMX solution with the desired composition was transferred to the inlet tank and it was injected to the reactor with flow rate of 1 L/min in a circulating mode. The influent tank was sampled at a given time intervals for analyzing the target parameters. In continuous-flow operation, the effect of hydraulic retention time (HRT) on AMX degradation was evaluated. In these tests, the AMX solution was pumped from the influent tank to the reactor and the effluent was collected in the effluent tank. The effluent was sampled at each HRT after passing 2 times of that HRT. All experiments were conducted at room temperature.

2.3. Analysis

AMX concentration was measured using an Agilent HPLC (Eclipse Plus C18 column; 3.5 μm , 4.6 \times 100 mm) with a UV detector at a wavelength of 190 nm. The mobile phase was a mixture of buffer phosphate with pH=4.8 and acetonitrile with a volumetric ratio of 60/40 with an injection flow rate of 1 mL/min. The pH level of samples was measured using a JENWAY, 3505 pH Meter. Duplicate tests were performed for each case and the mean values were used to represent each evaluation. The percentages of AMX degradation were calculated using the following Equation:

$$\text{AMX degradation(\%)} = \frac{(C_0 - C_t)}{C_0} \times 100 \quad (5)$$

where C_0 and C_t are the AMX concentration in the solution before and after the reaction, respectively. The mineralization of AMX in VUV process was also evaluated by measuring the TOC of the solution using a TOC analyzer (Shimadzu Co.). The degree of mineralization of AMX was calculated using the following equation:

$$\text{AMX mineralization(\%)} = \frac{(\text{TOC}_0 - \text{TOC}_t)}{\text{TOC}_0} \times 100 \quad (6)$$

where TOC_0 and TOC_t are the TOC concentration of solution before and after the reaction, respectively.

The kinetic of AMX degradation and mineralization was assessed by the pseudo-first-order kinetic model (Eq. (3)). The goodness of the model fitness with the experimental results was evaluated based on the determination coefficient (R^2) value.

$$\ln\left(\frac{C_t}{C_0}\right) = -k_1 t \quad (7)$$

where C_t and C_0 are the concentration of the AMX or TOC in the solution before and after the reaction, respectively, and k_1 is the pseudo-first-order reaction rate constant.

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