



## Research article

## Degradation of Penicillin G by heat activated persulfate in aqueous solution

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## ABSTRACT

We used Heat Activated of Persulfate (HAP) to decompose Penicillin G (PEN G) in aqueous solution. The effect of pH (3–11), temperature (313–353 K), and initial concentration of Sodium Persulfate (SPS) (0.05–0.5 mM) on the decomposition level of PEN G were investigated. The residue of PEN G was determined by spectrophotometry at the wavelength of 290 nm. Also, the Chemical Oxygen Demand (COD) was measured in each experiment. The Total Organic Carbon (TOC) analysis was utilized for surveying the mineralization of PEN G. In addition, based on Arrhenius equation, the activation energy of PEN G decomposition was calculated. The results indicated that the maximum PEN G removal rate was obtained at pH 5 and by increasing the doses of SPS from 0.05 to 0.5 mM, the PEN G decomposition was enhanced. It was found that an increase in temperature is accompanied by an increase in removal efficiency of PEN G. The activation energy of the studied process was determined to be 94.8 kJ mol<sup>-1</sup>, suggesting that a moderate activation energy is required for PEN G decomposition. The TOC measurements indicate that the HAP can efficiently mineralize PEN G. Besides, the presence of the scavengers significantly suppressed the HAP process to remove the PEN G. Overall, the results of this study demonstrate that using HAP process can be a suitable method for decomposing of PEN G in aqueous solutions.

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## 1. Introduction

Antibiotics play an important role in contamination of ground-water and surface water resources due to their high consumption in treating of bacterial infections (Robinson et al., 2007; Walter and Vennes, 1985). These contaminant compounds are identified in high concentrations (ppm) in hospital wastewater, lower concentrations (ppb) in municipal wastewater, and much lower (ppt) in surface water resources (Brown et al., 2006; Ferdig et al., 2005). Due to long residence times and persistence because of reducing chemistry and relatively low microbial rates in groundwater, a considerably high level of antibiotics has been reported in groundwater (Ji et al., 2014). As a result, they can cause microbial resistance to the antibiotics and destruct the microbes used in wastewater treatment (Kümmerer, 2009).

Previous studies have shown that antibiotics are not totally removed during biological treatment; rather, they enter water resources through effluent of wastewater treatment plants (Balcioglu and Ötger, 2004). Since these compounds are non-biodegradable (Alaton et al., 2004), they are not effectively removed through conventional municipal wastewater treatment processes. In the recent studies, various methods have shown the ability to remove pharmaceutical compounds (Esplugas et al., 2007; Radjenović et al., 2008; Rivera-Utrilla et al., 2009). Some of these advanced treatments such as biofiltration, adsorption on activated carbon, and reverse osmosis (Daghrir et al., 2012; Snyder et al., 2006) are utilized for removing of antibiotics but these methods only transfer contaminants from one phase to another one (Ikehata et al., 2008; Nasuhoglu et al., 2012). Among different methods, advanced oxidation process has been successfully used for destructing resistant pollutants (Taghi Ghaneian et al., 2016). The basis of this process is to produce hydroxyl radicals OH<sup>•</sup> and Sulfate radicals (SO<sub>4</sub><sup>•-</sup>) (Saïen et al., 2011). The SO<sub>4</sub><sup>•-</sup> radical is a very powerful oxidant with oxidation-reduction potentials of 2.5–3.1 V (Neta et al., 1988). SO<sub>4</sub><sup>•-</sup> can effectively decompose organic compounds (Yang et al., 2011). Generally, SO<sub>4</sub><sup>•-</sup> radicals directly change the organic

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compounds into organic radical cations by separating electrons from organic compounds (Tan et al., 2013a). By activation through heat, UV, and transition metals,  $SO_4^{\cdot-}$  radical is usually generated from Sodium Persulfate ( $Na_2S_2O_8$ , SPS) or Potassium Peroxymonosulfate ( $KHSO_5$ , PMS) (Liu et al., 2013; Yang et al., 2010). Liu et al. (2018b) investigated degradation of trichloroethene by persulfate activated with  $Fe^{2+}$  and zero-valent iron. Degradation of UV-filter benzophenone-3 in aqueous solution using persulfate catalyzed by cobalt ferrite was studied by Pan et al. (2017), where they reported a removal efficiency of 91 percent achieved in 6 h at initial pH of 7, catalyst dosage of 500 mg/L, and initial concentration of 1.3  $\mu$ M for benzophenone-3. Degradation of sulfloxazole using peroxymonosulfate activated by copper-cobalt oxides (Yao et al., 2017), degradation of tetracycline hydrochloride by electro-activated persulfate (Liu et al., 2018a), degradation of sulfolane using activated persulfate with UV and UV-Ozone (Izadifard et al., 2017), and degradation of methyl paraben using UV-activated persulfate (Dhaka et al., 2017) also were reported by other researchers.

Heat activation may effectively shorten the reaction time and lead to a higher drop in consumption of SPS compared to the other mentioned methods (Tan et al., 2013a). Thermo-activated persulfate oxidation of sulfamethoxazole was studied by Ji et al. (2015) and introduced as an effective approach for the treatment of SMX and other sulfonamides in waters.

Penicillin G (PEN G) is the first discovered antibiotic by a human that used for treating bacterial diseases (Peterson et al., 2012) and is one of the most widely used antibiotics (Brown et al., 2006). The present research was conducted to study the feasibility of using Heat-activated SPS (HAP) to remove PEN G from aqueous solution. It is identified that the reaction between SPS and heat (Eq. (1)) leads to generation of  $SO_4^{\cdot-}$  ( $E = 2.6$  eV) (Tan et al., 2013a).



## 2. Materials and methods

### 2.1. Chemicals and apparatus

PEN G (Penicillin G sodium salt) and Sodium Persulfate were prepared from Sigma Aldrich (USA) Company. The specifications of PEN G and SPS are shown in Table 1. To measure chemical oxygen demand (COD), Mercury (II) Sulfate, Potassium Dichromate, Silver Sulfate, and Sulfuric acid were purchased from Merck Company (Germany). All chemicals were of analytical grade. Double distilled water was utilized in all experiments. The pH was adjusted using HCl and NaOH. The temperature of solutions was set by a shaker incubator (MaxQ Mini 4450, Thermo Scientific). A spectrophotometer (LUV-100A model) was applied to measure the

concentration of residual PEN G and a Dr5000 spectrophotometer (HACH) was used to determine the COD concentration. Also, to investigate the mineralization of PEN G, the total organic carbon (TOC) was determined through TOC analyzer (ANATOC Series II).

### 2.2. Experimental procedure

A batch system was used to conduct the experiments. Effects of pH, initial concentrations of SPS, temperature, and the presence of organic and inorganic scavengers were studied. Before each experiment, PEN G and SPS stock solutions were freshly prepared through the double distilled water. The used reactor was filled with 250 ml of the prepared solution. In each experiment, to control interferences, blank sample (SPS-free) was utilized and little changes were observed in PEN G decomposition (<5%). The residual of PEN G also was determined by spectrophotometry at the wavelength of 290 nm. Besides, to determine the residual concentration of PEN G and its intermediate products, COD and TOC were measured. Also, before adding SPS to the solution, the solution temperature was set. To ensure a complete mixing during the experiment, the shaker speed was set at 100 rpm. The results were reported as the average of three replications of each experiment. The apparent activation energy to decompose PEN G at the studied temperatures was calculated through Arrhenius equation (Eq. (2)) (Ghauch and Tuqan, 2012; Wang and Liang, 2014).

$$k = A \exp\left(\frac{-E_a}{RT}\right) \quad (2)$$

Where  $A$  is the frequency factor,  $E_a$  ( $J \text{ mol}^{-1}$ ) is the apparent activation energy,  $R$  ( $8.314 \text{ J mol}^{-1} \text{ K}^{-1}$ ) is the ideal gas constant, and  $T$  (K) is the reaction absolute temperature (Ghauch et al., 2012b).

Kinetic study of PEN G was also investigated. The degradation of PEN G by HAP was fitted by the Langmuir-Hinshelwood as the following equation (Taghavi et al., 2018a):

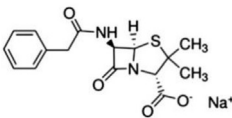
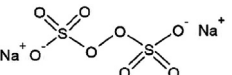
$$\ln \frac{C_0}{C_t} = kt \quad (3)$$

where  $C_0$  and  $C_t$  are concentrations of PEN G at initial and contact time of  $t$ , respectively, and  $k$  is the rate constant that is obtained from the slope of the straight-line of the plot of  $\ln(C_0/C_t)$  versus  $t$  (Taghavi et al., 2018b).

Also, the effects of organic and inorganic hydroxyl radical scavengers on the PEN G degradation were determined using chloride and carbonate as inorganic radical scavengers, and chloroform,  $Na_2$ -EDTA, t-butanol, and benzoquinone as organic radical scavengers. In this stage, the initial PEN G concentration, SPS dosage, pH, and temperature were assigned to be constant at 0.02 mM, 0.5 mM, 5, and 353 K, respectively. Subsequently, both

**Table 1**

Physical and chemical properties of PEN G and SPS.

| Chemical Name       | Chemical structures   | Molecular formula       | Molecular weight ( $g \text{ mol}^{-1}$ ) |
|---------------------|---|-------------------------|---|
| Penicillin G sodium |  | $C_{16}H_{17}N_2NaO_4S$ | 356.37                                    |
| Sodium Persulfate   |  | $Na_2O_8S_2$            | 238.09                                    |

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