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Degradation and Detoxification of Tetracycline by Sono-Fenton and Ozonation

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Abstract : Among a wide variety of pharmaceutical compounds, tetracycline antibiotics are one of the largest groups of pharmaceutical compounds extensively used in human and veterinary medicine to treat and prevent bacterial infections. Because it is water soluble, biologically active, stable and bio-refractory, release to the environment threatens aquatic life and increases the risk posed by antibiotic-resistant pathogens. In practice, due to its antibacterial nature, tetracycline cannot be effectively destructed by traditional biological methods. Hence, in this study, two advanced oxidation processes such as ozonation and sono-Fenton processes were conducted individually to degrade the tetracycline for investigating their feasibility on tetracycline degradation. Effect of operational variables on tetracycline degradation, release of nitrogen and change of toxicity were also proposed. Initial tetracycline concentration was 50 mg/L. To evaluate the efficiency of tetracycline degradation by ozonation, the ozone gas was produced by an ozone generator (Model LAB2B, Ozonia) and introduced into the reactor with different flows (25 - 500 mL/min) at varying pH levels (pH 3 - pH 11) and reaction temperatures (15 - 55°C). In sono-Fenton system, an ultrasonic transducer (Microson VCX 750, USA) operated at 20 kHz combined with H_2O_2 (2 mM) and Fe^{2+} (0.2 mM) were carried out at different pH levels (pH 3 - pH 11), aeration gas and flows (air and oxygen; 0.2 - 1.0 L/min), tetracycline concentrations (10 - 200 mg/L), reaction temperatures (15 - 55°C) and ultrasonic powers (25 - 200 Watts), respectively. Sole ultrasound was ineffective on tetracycline degradation, where the degradation efficiencies were lower than 10% with 60 min reaction. Contribution of Fe^{2+} and H_2O_2 on the degradation of tetracycline was significant, where the maximum tetracycline degradation efficiency in sono-Fenton process was as high as 91.3% followed by 45.8% mineralization. Effect of initial pH level on tetracycline degradation was insignificant from pH 3 to pH 6 but significantly decreased as the pH was greater than pH 7. Increase of the ultrasonic power was slightly increased the degradation efficiency of tetracycline, which indicated that the hydroxyl radicals dominated the oxidation of tetracycline. Effects of aeration of air or oxygen with different flows and reaction temperatures were insignificant. Ozonation showed better efficiencies in tetracycline degradation, where the optimum reaction condition was found at pH 3, 100 mL O_3/min and 25°C with 94% degradation and 60% mineralization. The toxicity of tetracycline was significantly decreased due to the mineralization of tetracycline. In addition, less than 10% of nitrogen content was released to solution phase as NH_3-N, and the most degraded tetracycline cannot be full mineralized to CO_2. The results shown in this study indicated that both the sono-Fenton process and ozonation can effectively degrade the tetracycline and reduce its toxicity at profitable condition. The costs of two systems needed to be further investigated to understand the feasibility in tetracycline degradation.

Keywords : degradation, detoxification, mineralization, ozonation, sono-Fenton process, tetracycline
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