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Microwave-assisted Fenton's oxidation of amoxicillin

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HIGHLIGHTS

- ▶ MW-assisted Fenton's oxidation significantly improved amoxicillin degradation.
- ▶ Classic and microwave assisted Fenton's reaction were compared.
- ▶ MW-assisted Fenton's oxidation main parameters were optimized step-by-step.
- ▶ Total degradation was reached in 5 min, with a reduced amount of iron catalyst.
- ▶ A semi-empirical kinetic model was proposed to predict the two-stage decay curves.

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ABSTRACT

Advanced oxidation processes (AOPs) have been increasingly applied to emergent pollutants degradation. Although, homogeneous reaction by classical Fenton enables amoxicillin degradation, high iron catalyst concentrations are needed, raising environmental concerns.

This work proposed an innovative and cheap solution to degrade amoxicillin by combining microwave with Fenton's reaction. The main operational parameters were optimized step-by-step (hydrogen peroxide and ferrous ion concentration and microwave power). Amoxicillin oxidation was significantly improved over classical Fenton's reaction. In fact, in less than 5 min ($P = 162$ W, $[H_2O_2]_0 = 2.35$ mg L^{-1}, $[Fe^{2+}]_0 = 95$ μg L^{-1}) amoxicillin was no longer detected in the reaction system. A semi-empirical kinetic model was proposed to predict the two-stage decay curves at any conditions within the studied parameters and the adequacy of the model was statistically evaluated.

1. Introduction

The β-lactam compounds represent by far the most consumed class of antibiotics. In fact, amoxicillin (AMOX), a broad-spectrum antibiotic, is one of the most prescribed worldwide [1–3]. When ingested, it is partially excreted unmetabolised (about 80–90%) via urine and faeces into the domestic sewage and subsequently discharged to the wastewater treatment plants (WWTPs) [4]. However, most traditional WWTPs are not designed to remove this kind of compounds and therefore, they are released into the ecosystem and can cause serious pollution problems.

Recent studies have reported the environmental presence of amoxicillin in concentrations in the range of μg–ng L^{-1} [5–10]. Due to the persistence and bioaccumulation in the environment, this antibiotic may induce toxic effects, changing the natural balance of ecosystems. Moreover, the presence of broad spectrum antibiotics, such as amoxicillin, in aquatic environments, has promoted the formation of antibiotic-resistant bacteria, which cause human health problems. Therefore, it is necessary to develop new alternative methods to prevent environmental pollution, namely water contamination.

Traditional chemical and physical methodologies have been tested to treat waters polluted with antibiotics, but they were not sufficiently effective for most of these compounds. The conventional biological systems applied in WWTPs were unsuccessful in the removal of these organic compounds due to their recalcitrant and toxic nature to the microorganisms [11–13].

In recent years, advanced oxidation processes (AOPs) emerge as new alternatives for the treatment of these effluents containing antibiotics. Several studies on antibiotics degradation by AOPs have been published in recent years. Ozonation, Fenton, photo-Fenton, photolysis and semiconductor photocatalysis are some examples of this kind of processes. An overview of recently published papers on amoxicillin degradation in water matrices by AOPs is present in Table 1.

Although most studies reported in the literature lead to a complete removal of amoxicillin, the time required for treatment is usually long ($t > 30$ min). Furthermore, some methodologies as ozonation and semiconductor photocatalysis requires large

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