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Fenton and Photo-Fenton Oxidation Processes for Degradation of 3-Aminopyridine from Water

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Abstract

3-Aminopyridine (3AP), used in manufacture of anti-inflammatory drugs and also as a plant growth regulator is one of the emergent contaminant, because of its toxic and carcinogenic potential and hazardous effect on natural environment. The objective of present study was therefore to investigate chemical treatment like the advanced oxidation technologies employing the classic Fenton and Photo-Fenton oxidation wherein effect of operating conditions like pH, Hydrogen peroxide (H_2O_2), Iron salts (both ferrous and iron extracted from laterite soil) and reaction time are optimized using synthetic 3-Aminopyridine solutions. In the present study, for 3AP conc. ranging from [10-80mg/L] under Fenton's oxidation at pH 3, optimum ratio of [H_2O_2]/[Fe^{2+}] :: [24-40]/[1] showed upto [90-77%] removal efficiency. Studies on use of laterite iron replacing the traditional ferrous iron, also showed comparable removal efficiencies upto [82-65%] for [H_2O_2]/[laterite iron] :: [32-53]/[1]. Moreover Photo-Fenton oxidation studies showed 100% removal for conc. range (10-30mg/L) under both iron salts. For Fenton's oxidation, optimum reaction time of 5.0 hrs for 10-30mg/L to 7.0 hrs for 40-60mg/L and finally to 8.5 hrs for 70-80mg/L of 3AP was required. Whereas photo-Fenton reaction studies required much less reaction time equal to 1.5 hrs for 10-30mg/L to 2.0 hrs for 40-60mg/L and 4.0 hrs for 70-80mg/L. Also Chemical oxygen demand (COD) removal was increased in case of Photo-Fenton oxidation indicating improved mineralization. Fenton and photo-Fenton methods can be considered as an effective advanced oxidation methods at ambient conditions. Also iron extracted from laterite soil can be used effectively in Fenton's reagent instead of traditional ferrous salts to treat polluted water bodies containing 3-Aminopyridine.

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1. Introduction

Pharmaceutical and Personnel Care Products (PPCPs) continues to grow worldwide on par with many agrochemicals. PPCPs are in part subjected to the metabolism of the user. The excreted metabolites plus some unaltered parent compounds get released into the sewage. Thus these chemicals enter the environment, where they are considered *pseudo-persistent* because the transformation/removal rate from the environment is compensated by the rate of replacement [1].The detection of (PPCPs) in the aquatic environment has added a new dimension to water quality programs in developing regions of the world. PPCPs may cause infertility, birth defects, ovarian failure and growth retardation. Declined sex ratios in Canada and the United States have also been reported. Research and published data are increasingly showing evidence of deleterious impacts of PPCPs on the ecosystem. Feminizations of fishes and gulls and sexual abnormalities in alligators due to exposure of PPCPs have been reported [2]. Likewise Pseudo-persistancy has been observed in nitrogen containing heteroaromatic pharmaceutical compounds like pyridine. These compounds have received immense attention recently, because of their presence in the environment and their toxic and carcinogenic potential and hazardous effect on natural environment [3]. Furthermore, some of the pyridine derivatives like aminopyridines can be toxic to certain life forms [4, 5]. Pyridines are rated as priority pollutants by United States Environmental Protection Agency (USEPA) [6]. Many pyridines of commercial interest find application in market areas where bioactivity is important. 3-Aminopyridine is also used as a plant growth regulator and also in azo dye in hair dyes and pharmaceutical drugs like Piroxicam, Tenoxicam, Ampiroxicam (anti-inflammatory drugs) [7].Treatment of pharmaceutical wastewater containing 3AP has always been troublesome to reach the desired effluent standards. Also there is addition of similar substances due to municipal sewage discharges. Moreover it is resistant to biological degradation [5]. In such a case, chemical pre-treatment like the advanced oxidation technologies particularly Fenton and Photo-Fenton oxidation processes which rely on the generation of very reactive oxidizing agents, i.e. free radicals such as the hydroxyl radical (•OH) are found effective owing to their high oxidation potential (+2.80 eV) in aqueous solution . These processes can adequately increase the biodegradability and remove toxicity of the wastewater prior to biological treatment [8].

2. Experimental Details

All the chemicals required for carrying out the aforesaid study with selected pyridine derivative viz; 3-Aminopyridine (3AP), extra pure (98% assay) were purchased from Merck. (India). The experiments were conducted at ambient temperature (27 ± 3^0C) in batch reactors. A 1000 mL solution of required 3AP concentration was prepared from the stock 3AP solution and was taken in a 2 litre reactor. Appropriate amount of Fe^{2+} concentration from the 1000 mg/L stock solution, freshly prepared from $FeSO_4.7H_2O$, was added to the reactor bath and stirred with magnetic stirrer. Also stock solution of iron extracted from laterite soil of NITK campus, India was prepared as per standard procedure. The iron from laterite soil was extracted using procedure given by Olanipekun [9]. Required amount of H_2O_2 was added to the reactor bath to initiate the reaction. For the experiments, pH (digital pH meter, Model EQ-610-Equiptronics) was adjusted after adding appropriate iron and H_2O_2 solution, stirred with magnetic stirrer (Model EQ-772-Equiptronics).pH of the solution was adjusted using 0.1NH_2SO_4 and 0.1NNaOH. The mixture of 3AP solution and Fenton's reagent (Fe^{2+} and H_2O_2) was stirred with magnetic stirrer during treatment. The experiment of Photo-Fenton oxidation is similar except stirring is carried out in presence of UV light (253.7nm) in specially designed UV