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Anahtar Kelimeler: DEET (N,N-diethyl-m-toluamide), yüzey suyu, LC-MS/MS, katı faz ekstraksiyonu (SPE), Orta ve Güney Türkiye

S-022

DEGRADATION OF CHLORAMPHENICOL (CAP) BY ELECTRO-FENTON PROCESS USING GRAPHENE OXIDE-AMORPHOUS FEPO4 AND GRAPHENE OXIDE-MAGNETITE (FE3O4) AS STABLE HETEROGENEOUS CATALYSTS

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Aim: Global freshwater reserves are rapidly depleting in the World. The purification of domestic wastewater and the release of it to the environment are very important. However, degradation of antibiotics is not effective with traditional systems. To eliminate these kind pollutants from water, more powerful processes, namely advanced oxidation processes (AOPs) was developed[1]. The AOPs are based on the in situ generation of strong oxidizing agent hydroxyl radicals (•OH) and their high oxidation power. One of AOPs is electro-Fenton (EF). In EF, hydrogen peroxide was continuously produced due to cathodic reduction of oxygen gas. Hydrogen peroxide reacts with ferrous ions which added catalytic amount to form hydroxyl radical.

$H_2O_2 + Fe^{2+} \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^-$ (1)

Chloramphenicol (CAP) is a broad-spectrum antibiotic exhibiting activity against both Gram-positive and Gram-negative bacteria, as well as other groups of microorganisms. It extensively used since the 1950s in the treatment of domestic animals all over the world due to low cost and ready availability[2]. However, Chloramphenicol is, in certain susceptible individuals, associated with serious toxic effects in humans including bone marrow depression, particularly severe in the form of fatal aplastic anemia[3].

For this reason, the use of this drug is forbidden by China, Japan, Canada, United States, Australia and European Union. But, it is still used legally in some countries. In the recent studies showed that it has been detected in several matrices, such as plants[4], milk[5], surface water[6] and sewage treatment plants[7].

Recently, graphene-based hybrid materials have been used for environmental applications in water purification for the adsorption of organic[7] and inorganic[8] contaminants, membrane technology [9] and the catalytic process[10].

In this study, GO-magnetite (Fe_3O_4) and GO-$FePO_4$ was synthesized and used as heterogeneous Fe^{2+} source for degradation of CAP in electro-Fenton process. These heterogeneous catalysts are magnetic materials and separate from solutions and will be reused. The effects of catalyst type, pH, current and time on decomposition and mineralization were studied.

Material and methods: Synthesis of GO-$FePO_4$: The graphene oxide (GO) was prepared Hummers method[11]. GO was dispersen in deionize water (DI) by ultrasonication for 30 min to obtain a homogeneous suspension.$NH_4H_2PO_4$ and $Fe(NO_3)_3$ solutions were added into GO solution.pH was adjust at 1.5. The precipitate was dried in air at 70°C.

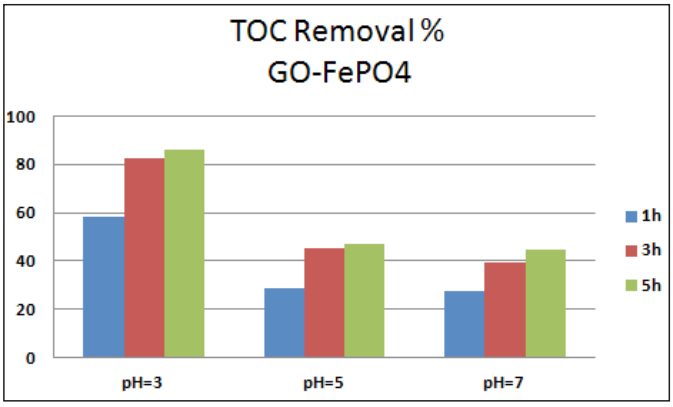
Synthesis of GO-Fe_3O_4: Firstly, GO was dispersed in DI water under ultrasonic shaking for 3h. After that $FeCl_2.4H_2O$ and $FeCl_3.6H_2O$ solutions were added into the GO solution.Finally, 16 ml of %30 NH_3 solution was quickly added into the GO solution and pH was adjust at 10. The preciapiate was respectively washed with ethanol and water, then was dried in air at 70°C.

Electro-Fenton process: Electrochemical experiments were performed at constant current with a DC power supply (TT-T-ECHNI-C) in a 250 mL undivided cylindrical glass cell. 0.05M Na_2SO_4 was used to enhance the conductivity and 0.5M H_2SO_4 solution was added to bring the desired pH value. Electrolyses of 200 mL CAP (80 mg/L) solutions were performed on a carbon feltworking electrode (50 cm^2 = 12.5×4 cm) by applying a constant current of 200, 300, and 400mA. The counter electrode was Pt gauze(6 cm^2) placed on the center of the cell. The solution was stirred with a magnetic bar. A catalytic quantity of Fe^{2+}, GO-$FePO_4$ or GO-Fe_3O_4 was added to solutions before starting the electrolysis. Prior to the electrolysis, oxygen gaswas bubbled for 15 min to saturate the aqueous solution, which was fed with pure O_2 at 20 mL/min during the experiments.

Degradation of CAP: CAP concentration was analyzed spectrophotometrically on a UV–vis spectrometer (Shimadzu UV-160A) at 280 nm by measuring the absorbance of the untreated and treated samples at the maximum wavelength.

Mineralization of CAP: The total organic carbon (TOC) of the initial CAP solution and samples was determined with TOC cell test (Merck).

Results: When the effect of the current was examined, the highest mineralization was obtained at 300 mA in EF with ferrous ion at pH 3.0. Further experiments were done in this current. The effects of the prepared GO-composites on mineralization were investigated at different pH and dosage. The most effective mineralization for both GO-composites was found at pH 3 (Fig 1).



While 85% mineralization was provided with GO-$FePO_4$ at pH 3.0, this value remained at 45% at pH 5. The most effective amount of catalyst was observed as 0.5 g / L.

The SEM image of GO-FePO4 was given Fig 2. The prepared catalyst was found to have an amorphous structure.