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Comparison of Fenton and electro-Fenton processes for oxidation of phenol

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ABSTRACT

This study compares the performance of Fenton and electro-Fenton processes for phenol degradation. The effects of operational parameters such as initial pH, current density, concentration of phenol and hydrogen peroxide on phenol removal was investigated in electro-Fenton process. The degradation and mineralization efficiency increased with an increase in hydrogen peroxide concentration and current density and decreased with an increase in initial phenol concentration and initial pH. It was found that optimum pH, current density and H₂O₂ concentration were 3.0, 1 mA/cm² and 500 mg/L, respectively. Under the optimized conditions, the phenol and chemical oxygen demand (COD) removal efficiency reached 93.32% and 87.5%, respectively. In conventional-Fenton process, the effect of hydrogen peroxide and Fe²⁺ concentrations on the degradation and mineralization of phenol removal were investigated. The results indicated that the conventional-Fenton process only yields 59% mineralization. An estimation of the operating costs of the processes investigated showed that electro-Fenton was the more economical system to treat the phenol containing wastewater.

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1. Introduction

Phenol and its compounds are present in the wastewaters from many industrial sectors such as oil refineries, petrochemical and ceramic plants, coal conversion activities, pharmaceuticals, pesticides, dyes, plastics, explosives and herbicides and phenolic resin production" (Pigatto et al., 2013). These compounds are very hazardous due to their poor biodegradability, high toxicity and ecological aspects. The presence of phenol in drinking water and irrigation water represents a serious health hazard to humans, animals, plants and microorganisms. Phenol is a potential carcinogen of human, which raises considerable health concerns, even at low concentrations. Phenol is classified by the USEPA as a priority pollutant. (Subramanyan, 2014). "It is therefore essential to develop effective treatment techniques for the removal

of phenol in wastewater" (Nidheesh and Gandhimathi, 2012). "Treatment of phenolic wastewater to harmless level is difficult for many biological and chemical processes due to its high solubility and stability in water" (Kavitha and Palanivelu, 2004). Advanced oxidation processes (AOPs) such as Fenton (Neyens and Baeyens, 2003; Bautista et al., 2008) and electro-Fenton (Nidheesh and Gandhimathi, 2012; Yuan et al., 2006) are successfully used for the removal of phenol. Especially, electro-Fenton which is a new advanced oxidation process induced by electrochemistry has attracted much interest (Chang et al., 2004).

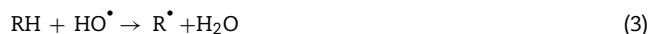
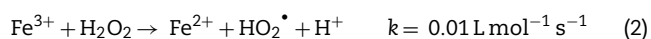
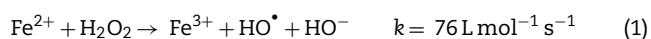
"The generally accepted mechanism of the Fenton process proposes that hydroxyl radicals are produced in accordance with Eq. (1), while the catalyst is regenerated through Eq. (2), or from the reaction of Fe³⁺ with intermediate organic radicals" (Eqs. (3)–(5)) (Bautista et al., 2008).

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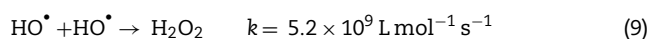
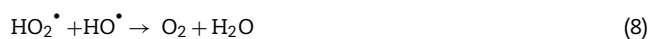
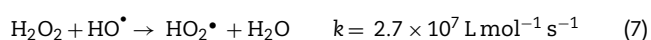
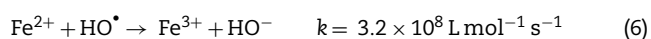
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Nevertheless, a number of competitive reactions can also occur (Eqs. (6)–(9)), which negatively affect the oxidation process:



In electro-Fenton (EF) process, pollutants are destroyed by the action of Fenton's reagent in the bulk together with anodic oxidation at the anode surface. Electro-Fenton process is classified into four categories depending on Fenton's reagent addition or formation. In type 1, hydrogen peroxide and ferrous ion are electro-generated using a sacrificial anode and an oxygen sparging cathode respectively. In type 2, hydrogen peroxide is externally added while ferrous ion is produced from sacrificial anode as shown in Eq. (10)



In type 3, ferrous ion is externally added and hydrogen peroxide is generated using an oxygen sparging cathode. In type 4, hydroxyl radical is produced using Fenton reagent in an electrolytic cell and ferrous ion is regenerated through the reduction of ferric ions on the cathode (Babuponnusami and Muthukumar, 2013).

The aim of this study is to compare electro-Fenton and conventional-Fenton process for phenol removal. The effects of experimental parameters such as initial pH, current density, concentration of phenol and hydrogen peroxide on the degradation and mineralization of phenol were investigated. Operating costs were also calculated for electro-Fenton and conventional-Fenton process to compare their economic feasibility.

2. Materials and methods

2.1. Materials

Phenol was obtained from Merck (99.5%). The synthetic phenolic wastewaters were prepared by dissolving an appropriate amount of phenol in deionised water. All chemicals used for the analysis were also obtained from Merck. Iron electrodes (99%) were used in the experiments as the cathode and anode materials with dimensions of 45 mm × 60 mm × 2 mm. The total effective surface area of iron electrodes was 58.5 cm².

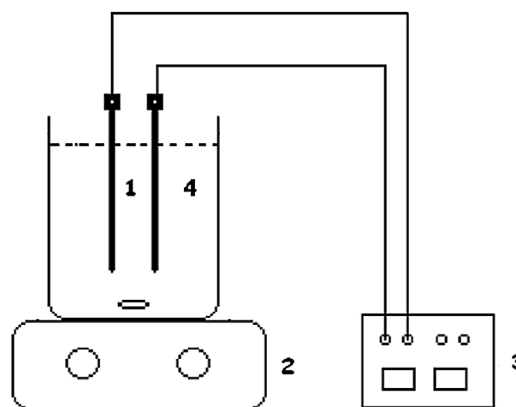


Fig. 1 – Experimental apparatus (1) iron electrodes, (2) magnetic stirrer, (3) power supply, (4) wastewater.

2.2. Electro-Fenton experiments

Electrochemical reactor was made of plexiglas with the dimensions of 102 mm × 102 mm × 90 mm and equipped with iron electrodes. The experimental apparatus is illustrated in Fig. 1. Anode and cathode were mounted parallelly and situated approximately 4 cm apart from each other. The electrodes were supplied from a digital DC power supply. Experiments were carried out by varying the pH of the solution (pH 3.0–7.0), current density (1–5 mA/cm²), concentration of phenol (50–500 mg/L) and concentration of hydrogen peroxide (0–1000 mg/L). In each run, 500 ml of synthetic phenolic wastewater at specified initial concentrations was prepared. The pH and conductivity of synthetic wastewater were adjusted to desired value using 1 M H₂SO₄ and 0.05 M Na₂SO₄ solutions, respectively. Then hydrogen peroxide was added at desired concentrations. After synthetic wastewater placed in to the electrochemical reactor the power supply was initiated. At the end of the run, the pH was adjusted to 10 using 1 M Ca(OH)₂ and the solution was filtered and samples were analyzed for phenol and COD.

2.3. Conventional-Fenton experiments

In the conventional-Fenton study, the effects of the H₂O₂ and Fe²⁺ concentrations on the phenol and the COD removal were investigated. Fenton experiments were performed at initial phenol concentration of 250 mg/L. H₂O₂ concentrations ranged between 250 and 1000 mg/L and Fe²⁺ concentrations ranged between 15.0 and 60.0 mg/L. During the experimental studies, the volume of the synthetic wastewater was 500 mL. The synthetic wastewater was placed in a beaker and the pH value was adjusted to 3.0. Fenton experiments were carried out by the addition of ferrous ion and hydrogen peroxide to synthetic wastewater, then the mixture was agitated for 5 min. At the end of the run, the pH was adjusted to 10 using 1 M Ca(OH)₂ and the solution was filtered and phenol and COD content were analyzed.

2.4. Analytical methods

Phenol and Chemical Oxygen Demand (COD) were analyzed according to standard Methods (APHA, 1995). Phenol was determined by the 4-aminoantipyrine method by UV-Vis spectrophotometer (Termospectronic Helios). COD was determined by closed reflux method. The COD of the samples were analyzed using a thermoreactor (NOVA 60) and a photometer

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