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A comparative study for treatment of white liquor by different applications of Fenton process



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Abstract In this paper, the treatability of white liquor by conventional (CFP), modified (MFP) and electro-Fenton oxidation processes (EFP) was investigated depending on the COD parameter. Based on the experimental results, up to 62.4%, 58.4% and 54.9% COD removals by the CFP, MFP and EFP were achieved, respectively. It was observed that adjustment of initial pH to acidic values is not required in the CFP. The optimal operational conditions were found to be $[\text{Fe}^{2+}] = 500 \text{ mg/L}$, $[\text{H}_2\text{O}_2] = 1000 \text{ mg/L}$ at pH 7.3 (original pH) in the CFP, $[\text{Fe}^0] = 1250 \text{ mg/L}$, $[\text{H}_2\text{O}_2] = 1000 \text{ mg/L}$ at pH 3 in the MFP, and $I = 1.0 \text{ A}$, $[\text{H}_2\text{O}_2] = 1500 \text{ mg/L}$ at pH 3 in the EFP, respectively. As a result, the CFP has been determined as a more efficient alternative treatment method.

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1. Introduction

The pulp and paper industry is a water intensive industry and its high water consumption is one of the most important environmental concerns in this industry. The amount and characteristics of paper industry effluents depend upon the type of manufacturing process, type of the raw materials, process technology applied, management practices, internal recirculation of the effluent for recovery, and the amount of water to be used in the particular process (Pokhrel and Viraraghavan, 2004). While

the wastewater originating from the pulp making process is classified as black liquor, pulp bleaching effluent is called as white liquor. White liquors may contain dissolved lignin, carbohydrate, organic and inorganic chlorine compounds such as chlorate and chlorophenols, volatile organic compounds such as chloroform and carbon disulfide and color (Pokhrel and Viraraghavan, 2004). Discharge of this kind of completely untreated wastewaters may cause scum formation, slime growth, thermal impacts, color problems, loss of esthetic beauty and toxic effects on living organisms in the environment. Therefore, the paper industry wastewaters should be treated before their discharge.

In order to treat the paper industry wastewaters, though the conventional coagulation and activated sludge processes can be applied on the pulp and paper mill effluents containing various organic and inorganic non-biodegradable materials and color; quality of the treated effluent cannot meet the environmental regulations in many cases (Amat et al., 2005). In addition, the non-biodegradable fraction of the organic compounds can be accumulated in the waste biological sludge

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(Tambosi et al., 2006). Thus, for the treatment of the pulp and paper industry effluents, physico-chemical and chemical treatment processes, such as adsorption (Shawwa et al., 2001), air flotation (Gubelt et al., 2000), coagulation (Gokcay and Dilek, 1994; Tong et al., 1999), Fenton oxidation (Sevimli, 2005), foto-Fenton oxidation (Xu et al., 2007), electro-coagulation (Parama Kalyani et al., 2009; Zaied and Bellakhal, 2009), electro-Fenton oxidation (Selvabharathi and Kanmani, 2010), photo-catalytic oxidation (Amat et al., 2005), sedimentation (Thompson et al., 2001), ozonation (Sevimli, 2005) and sonication (Shaw and Lee, 2009) attracted more attention as alternative ways to be used in combination with a biological treatment process. Among all these processes, the mechanisms of adsorption, coagulation, flotation and sedimentation are based on the phase transfer from the liquid phase to solid phase and cause the problem to remain unresolved in the solid phase. Although ozonation and photo-catalytic oxidation methods are effective to remove the pollutants from the wastewater, they are not attractive owing to their high capital and operational costs. Sonication was reported as an inefficient method for COD removal from the wastewater of pulp and paper kraft mill (Shaw and Lee, 2009). But, when compared with these methods, the Fenton oxidation process is the commonly used advanced oxidation process which is utilized successfully for the treatment of various industrial wastewaters (Aydin et al., 2002; Gogate and Pandit, 2004; Mahiroglu et al., 2009; Güçlü et al., 2012), because it is cost effective, easy to handle and efficient treatment technique. The Fenton process has two distinct stages, namely Fenton's oxidation which is based on the formation of hydroxyl radicals (OH^\bullet) by the reaction of hydrogen peroxide (H_2O_2) and ferrous ion (Fe^{2+}) in the acidic medium and Fenton's coagulation which is mainly simple ferric coagulation following the oxidation stage. The Fenton oxidation process can be easily modified by using zero valent iron (ZVI) instead of Fe^{2+} as the catalyst iron source. This modified process is named as the modified Fenton process (MFP) in this study. Furthermore, the Fenton process can be combined with the electro-coagulation process, which is called as the electro-Fenton process (EFP).

Up to now, the treatment of black and dark brown liquors from the pulp and paper industries has been widely investigated. Sevimli (2005) carried out a comparative study for the post-treatment of corrugated board factory effluent via CFP and ozonation. In his study, the CFP was determined as a more feasible process with 83% chemical oxygen demand (COD) removal in comparison with ozonation. In the study of Kazmi and Thul (2007), only 62% COD removal was achieved by the CFP. Tambosi et al. (2006) reported a nearly 50% COD removal from the pulp and paper industry effluent by an Fenton-like process using Fe^{3+} as the catalyst iron. In another study, Selvabharathi and Kanmani (2010) reported 90% COD removal from the dark brown liquor by the EFP. In spite of those studies conducted with the black and brown liquors, the literature contains a small number of studies on the treatment of the white liquor from the paper industry with different applications of Fenton processes. Moreover, we could not meet any report related with the treatment of the white liquor via the Fenton process modified with ZVI and the electro-Fenton process using cast-iron electrodes. For this reason, the main aim of the present work is to provide more insights into the treatment of the real paper industry effluent by different modifications of the Fenton process and to

determine the influences of different operating parameters on the COD removal.

2. Experimental section

2.1. The wastewater and chemicals used

The composite wastewater (white liquor) sample used in this study was obtained from the discharge point of an existing wastewater treatment plant of a paper factory in Konya, Turkey. The treatment plant has physical treatment units (equalization, pumping station, clarifier and neutralization). The wastewater sample was preserved in the dark at 4 °C in a refrigerator and used without any dilution. No significant dissolution in the wastewater sample was observed during the oxidation experiments. The wastewater had a pH of 7.3, turbidity of 434 NTU, COD of 865 mg/L, Cl^- (chloride) of 390 mg/L and SO_4^{2-} (sulfate) of 630 mg/L.

Hydrogen peroxide (H_2O_2) (35% w/w), ferrous iron sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), iron powder (ZVI, Fe^0), sodium hydroxide (NaOH), sulfuric acid (H_2SO_4), manganese dioxide (MnO_2), sodium chloride (NaCl) and other chemicals were all of analytical grade and purchased from Merck (Germany). All chemicals were utilized as received without further purification. Distilled water was used in the preparation of all solutions.

2.2. Experimental procedures

Classical (CFP) and modified Fenton (MFP) oxidation experiments were carried out with 500 mL working volume in Pyrex-glass beakers using a standard jar test apparatus (Velp, FC6S). The experiments were done at room temperature (24 ± 2 °C) using varying $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ – H_2O_2 (for CFP experiments) and Fe^0 – H_2O_2 (for MFP experiments) dosages at different pH values in order to determine optimum dosages. The CFP and MFP experiments were conducted in three steps. The wastewater pH was first adjusted to the desired value. The second step was the addition of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ and Fe^0 in CFP and MFP, respectively. In the final step, H_2O_2 was added into the reaction mixture. After the additions of Fenton reagents, the wastewater was mixed at 90 rpm during the oxidation stage of the Fenton process. At the end of the oxidation stage, the solution pH was adjusted to about 7.5 using 6 N and 0.1 N NaOH solutions and the wastewater was mixed for 3 min at 30 rpm to form iron (oxy)hydroxide ($\text{Fe}(\text{OH})_3$) flocs. After 30-min precipitation, 25 mL sample was pipetted from the supernatant for the COD analysis.

The EFP experiments were conducted with 500 mL of wastewater in a Pyrex-glass reactor. The wastewater was mixed by magnetic stirrer to homogenize the reaction solution. Cast-iron anode and cathode plates were utilized in pairs in the electro-chemical reactor. The electrodes, dimensions of which are 8.8 cm \times 2.0 cm (with a thickness of 3.0 mm), were positioned approximately 7 cm apart from each other and were immersed about 5 cm into the white liquor. The electrical direct current (DC) input was supplied by a DC power supply (Good Will, Taiwan). In each experimental run, 500 mL of wastewater was pour into the reactor. The wastewater pH was adjusted to the desired value using 0.1 and 6 N H_2SO_4 solutions. H_2O_2 was added into the wastewater and the electrical current was immediately turned on. During the adjustment of the

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