



Electro-Fenton process for implementation of acid black liquor waste treatment

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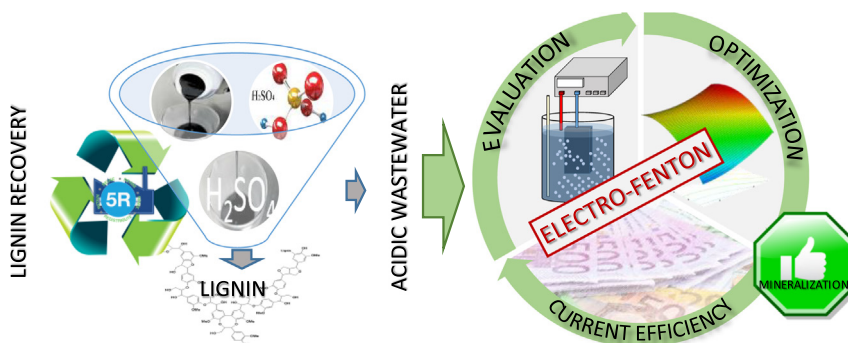
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HIGHLIGHTS

- Electro-Fenton demonstrated to be a feasible treatment for acid black liquor waste.
- Optimization of operational conditions following a response surface methodology.
- Significant reduction of color, total phenolic content and COD in wastewater.
- Environment-friendly treatment performed with high efficiency and low energetic cost.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, an eco-friendly solution for the remediation of wastewater generated in the lignin recovery process from eco-industrial paper mill has been proposed in their way towards a more circular economy strategy. Thus, the application of the electro-Fenton process for the degradation of the non-recovered lignin and other organic compounds form a scarcely studied acid black liquor waste (ABLW) was successfully performed. This treatment was able to operate in a range of COD loads (0.5–19.5 mg O₂·L⁻¹) showing high degradation values of the ABLW determined by the abatement of color, total phenolic content and COD. Then, the optimization of the working conditions for the design of a sustainable treatment system with optimum efficiency was carried out using a response surface methodology. The experiment carried out in the calculated optimal conditions for the electro-Fenton degradation process (current intensity 132.5 mA, catalyst dosage of 0.10 mM, and temperature 40 °C) showed a COD removal of 74.82% and current efficiency 77.79%, close to the theoretical value predicted by the model 73.12% and 77.06%, respectively. In addition, the identification of the final products permitted to confirm the mineralization efficiency.

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1. Introduction

Paper and pulp industry and its adverse effects and impacts into the environment have been seen as a source of water pollutants for a long

time (Lindholm-Lehto et al., 2015). Inside the production process, and during Kraft pulp production, wood is heated in an alkaline medium to dissolve the lignin and separate the cellulose, generating a solution, known as black liquor (Dieste et al., 2016). Based on black liquor properties and high lignin content, the extraction and use of this lignin increases the value created from the wood material and reduce the releases from the productive process (Doherty et al., 2011). For this

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reason, the lignin recovery from black liquor has attracted the attention of the scientific community.

Nowadays, several processes have been developed to recover lignin from black liquor. Among them, the more usual solution involves a combination of precipitation and solid separation processes. The acidic precipitation of black liquor is one feasible process for achieving this objective (Gouveia et al., 2012). Although after this treatment it is possible to recover a high proportion of Kraft lignin, the process also generates an ABLW with high Chemical Oxygen Demand (COD) and that is thrown in a stream water causing pollution. As result of this, an efficient treatment method is required to overcome the contamination by phenolic compounds, fatty acids, tannins, lignin and its derivatives, which some of them are listed and classified as priority pollutants by the United States Environmental Protection Agency (Ugurlu et al., 2006). In this context, several methods have been investigated such as adsorption on activated carbon, photo-catalysis using TiO₂, activated carbon post-treatment, anaerobic treatment; autohydrolysis and organosolv process and several types of advanced oxidation processes (Araujo et al., 2002; Oliveira et al., 2013; Sevimli et al., 2014; Ugurlu and Karaoglu, 2009; Wang et al., 2016). Among them, electro-Fenton could be a good alternative based on the high COD and acid medium of the waste, conditions that are desirable to perform this advanced oxidation process, which is considered as one of the most effective oxidative process (Brillas and Martínez-Huitle, 2015; Moreira et al., 2017; Sirés et al., 2014; Solano et al., 2016).

Electro-Fenton treatment is based on the Fenton's reaction with *in situ* generation of hydrogen peroxide and its reaction with an iron catalyst in order to generate a powerful oxidant, hydroxyl radical ($\cdot\text{OH}$), able to degrade a wide range of organic compounds. This process is considered as an environment-friendly technology involving the use of safe reagents: H₂O₂, Fe²⁺ ions, and the subsequent production of $\cdot\text{OH}$ (Eq. (1)). H₂O₂ is continuously electrogenerated (Eq. (2)) on the cathode and Fe²⁺ is constantly regenerated from Fe³⁺ (Eq. (3)–(6)) by direct reduction on the cathode or reduction processes involving H₂O₂ or intermediate organic radicals. Thus, this technology presents significant advantages such as low operational cost and high mineralization degree of pollutants (Brillas and Martínez-Huitle, 2015; Feng et al., 2013; Moreira et al., 2017; Oturan et al., 2008; Rodrigo et al., 2014; Rosales et al., 2012; Sirés et al., 2014).



Up to now, it is scarcely reported the application of electro-Fenton to the degradation of the pollutants present in ABLW. In our previous studies, the electro-Fenton process has been successfully applied to the degradation of different organic pollutants (Bocos et al., 2017; Iglesias et al., 2014; Iglesias et al., 2015).

The aim of this work was the design of a treatment system, with optimal efficiency, for the degradation of the non-recovered lignin and other organic compounds present in the acid black liquor supported on the potential application of the electro-Fenton process. For this purpose, the optimization of the working conditions was carried out using response surface methodology as tool. The degradation of the ABLW was followed by color decrease and COD abatement. In addition, the analysis of several phenols and carboxylic acids by high performance

liquid chromatography (HPLC) permitted to confirm the mineralization efficiency.

2. Experimental

2.1. Black liquor and reagents

Black liquor of a *Eucalyptus globulus* Kraft cooking was provided by ENCE (Pontevedra, Spain). Iron (II) sulphate heptahydrate (catalyst, 99%), sodium carbonate and Folin & Ciocalteu's phenol reagent were supplied by Sigma-Aldrich. Organic solvents were analytical grade from Fisher Chemicals and Rathburn Chemicals. Resorcinol, pyrocatechol, pyrogallol, *p*-hydroxybenzoic acid, hydroquinone, oxalic, succinic, glycolic, formic, acetic and oxamic acids were provided from Sigma-Aldrich and Merck. The solutions were prepared using purified water obtained by reverse osmosis technology (AquaMax-Basic 360).

2.2. Kraft lignin recovery and generation of ABLW

The black liquor was diluted with distilled water and then, pH was lowered to pH 2 with sulphuric acid (4 M) under stirring at 50 °C (Gouveia et al., 2012). The precipitated Kraft lignin was separated by means of a centrifugation process and the liquid named as ABLW was recovered for its treatment. The characteristic parameters have been summarized in Table 1.

2.3. Experimental procedure

Electro-Fenton experiments were carried out with a two-electrode system in a thermostatic undivided cylindrical glass reactor (250 mL) with cooling jacket to avoid concentration gradients and to perform the assays at different temperatures (from 20 to 40 °C). The carbon-felt cathode (19.0 × 6.0 × 0.6 cm, Carbon-Lorraine, RVG 2000) was placed on the inner wall of the glass cell whereas a double-side boron-doped diamond/Nb anode (2500 ppm B, 2.5 × 5.0 × 0.1 cm, Neocoat SA) with an active area of 17.5 cm² was located in the center of the reactor. Both electrodes were connected to a direct current (DC) power supply (HP model 3662) operating in a range of current intensities from 100 to 500 mA. The electrodes were immersed in 150 mL of a solution of ABLW and at natural pH of the solution (pH around 2–3). Electric parameters were recorded with a multimeter (Fluke 175, Fluke Corporation). Iron was added to the solution as catalyst and its dosage was evaluated by the experimental design in a range from 0.1 to 0.5 mM. To *in situ* electrogeneration of H₂O₂, air was continuously bubbled on the cathode at 1 L·min⁻¹. Samples were taken at the predetermined times and Fenton reaction was stopped by iron precipitation (NaOH 2 M).

Table 1
Characterization of ABLW.

Parameter	Value	Detected phenols	Concentration (ppm)
COD (g·L ⁻¹)	19.3	Pyrogallol	191.0
pH	1.7	Resorcinol	48.8
Total phenols (g·GA·L ⁻¹)	1.955	Pyrocatechol	53.9
Conductivity (mS·cm ⁻¹)	50.8	<i>p</i> -Hydroxybenzoic acid	14.0
Salts content (g·L ⁻¹)	76.5		
Water hardness (mg·L ⁻¹ CaCO ₃)	146.5		
TDS (ppm)	38,269		
Coloration	Light brown		

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