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# Treatment of olive oil mill wastewater by combined process electro-Fenton reaction and anaerobic digestion

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### Abbreviations:

OMW: olive mill wastewaters

COD: chemical oxygen demand

BOD<sub>5</sub>: biological oxygen demand

TSS: total suspended solids

LMM: low molecular mass

AF: anaerobic filter

VFA: volatile fatty acids

## ABSTRACT

In this work, we investigated an integrated technology for the treatment of the recalcitrant contaminants of olive mill wastewaters (OMW), allowing water recovery and reuse for agricultural purposes. The method involves an electrochemical pre-treatment step of the wastewater using the electro-Fenton reaction followed by an anaerobic bio-treatment. The electro-Fenton process removed 65.8% of the total polyphenolic compounds and subsequently decreased the OMW toxicity from 100% to 66.9%, which resulted in improving the performance of the anaerobic digestion. A continuous lab-scale methanogenic reactor was operated at a loading rate of 10 g chemical oxygen demand (COD) l<sup>-1</sup> d<sup>-1</sup> without any apparent toxicity. Furthermore, in the combined process, a high overall reduction in COD, suspended solids, polyphenols and lipid content was achieved by the two successive stages. This result opens promising perspectives since its conception as a fast and cheap pre-treatment prior to conventional anaerobic post-treatment. The use of electro-coagulation as post-treatment technology completely detoxified the anaerobic effluent and removed its toxic compounds.

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## 1. Introduction

Treatment and disposal of olive mill wastewater (OMW) represents one of the main problems for olive oil producing countries of the Mediterranean area. Tunisia is one of the largest olive oil producers in the world with an average annual production of 450,000 tons. This results in a by-product of 600,000 m<sup>3</sup> OMW. These liquid residues are 100–150 times more heavily loaded with pollutants than ordinary domestic wastewater (Sabbah et al., 2004). The high polluting activity of OMW is linked with their high content of organic molecules, especially polyphenolic mixtures (4–10 g l<sup>-1</sup>) with different molecular weights (Hamdi, 1992),

as well as their acidity and high concentration of potassium, magnesium and phosphate salts (Arienzo and Capasso, 2000). Besides aromatic compounds, OMW contain other organic molecules including nitrogen compounds, sugars, organic acids, and pectins (Della Greca et al., 2000), that increase their organic load (chemical oxygen demand (COD) = 80–200 g l<sup>-1</sup>; biological oxygen demand (BOD<sub>5</sub>) = 50–100 g l<sup>-1</sup>). Furthermore, the physico-chemical characteristics of OMW are rather variable, depending on climatic conditions, olive cultivars, degree of fruit maturation, storage time, and extraction procedure.

Many pollution disposal methods, such as concentration, evaporation, incineration, ultrafiltration/reverse osmosis,

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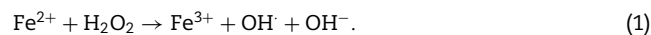
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lime precipitation, aerobic treatment, lagooning, codigestion, etc were tested on OMW, none of them led to industrial applications. However, anaerobic digestion seems to have some clear advantages that would make it the process of choice (Borja et al., 1992). Indeed, this treatment process produces energy (methane) and a digested effluent with a significant reduction of the organic load (Marques, 2001). However, many problems concerning the high toxicity and inhibition of biodegradation of these effluents were encountered during anaerobic treatments, because some bacteria, such as methanogens, were particularly sensitive to the organic contaminants present (Andreoni et al., 1984). The phenolic compounds severely limit the possibility of using anaerobic digestion (Sayadi et al., 2000). Therefore, the elimination of phenolic compounds from OMW was considered as an important objective in order to reduce its toxicity and to permit the occurrence of microbial fermentation. For this, research turned to a more promising alternative, namely the physico-chemical pre-treatment to remove the toxic compounds of OMW (Beccari et al., 1999).

In recent years, there has been increasing interest in the use of electrochemical technologies for the treatment of wastewaters. This technique was found to be successful in removing pollutants in various industrial wastewaters (Lin and Chang, 2000; Ciardelli and Ranieri, 2001; Lai and Lin, 2004). In two recent investigations, Inan et al., (2004) and Adhoum and Moncer (2004), an electrochemical method was used for decreasing the organic matter in OMW. Both investigators found efficient removals of COD, colouration and polyphenols content by electrolysis process using aluminium and iron electrodes. However, a relatively new chemical oxidation method that has not received much attention for OMW or other industrial wastewater treatment is the electro-Fenton method (Lin and Chen, 1997). This method represents a combination of the electrochemical process and the Fenton oxidation. It is based on the fact that hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) can be used as an oxidant in advanced oxidation processes to decompose refractory or toxic wastewaters (Kusvuran et al., 2004). As indicated in reaction (1), when the ferrous ion reacts with  $\text{H}_2\text{O}_2$  it will generate strong oxidant hydroxyl radicals ( $\text{OH}^\cdot$ ).



This  $\text{Fe}^{2+}/\text{H}_2\text{O}_2$  system, often referred to as Fenton's reagent (Fenton's), has dual functions of OH radical peroxidation as well as ferrous/ferric coagulation.

During this process, the non-biodegradable organics and toxic pollutants present in the wastewaters such as polyphenols are usually destroyed by direct or indirect anodic oxidation via the production of oxidants such as hydroxyl radicals and complex coagulants that promote the flocculation of the matter (Israilides et al., 1997; Panizza et al., 2000; Chen et al., 2002).

This paper will attempt to apply the electro-Fenton process to reduce the organic load and the toxicity of OMW in order to improve the anaerobic digestion in terms of biomethane yield. Electro-coagulation was assayed as a post-treatment for complete detoxification and colour removal allowing water recovery and reuse for agricultural purposes.

## 2. Materials and methods

### 2.1. OMW characterisation

Fresh OMW was obtained from an olive oil continuous processing plant located in Sfax (southern Tunisia). The OMW was characterised by high total suspended solids (TSS) content, COD concentration up to  $100 \text{ g l}^{-1}$  and polyphenols up to  $12 \text{ g l}^{-1}$ . Raw OMW was pre-decanted in a 120 l decanter before being treated by electro-Fenton in order to remove suspended solids (Fig. 1).

To confirm the role of electro-Fenton in polymerising and removing the highly polymerised phenolic fraction, experiments were carried out with a low-molecular-mass (LMM) polyphenolic fraction ( $<2 \text{ kDa}$ ) obtained by the ultrafiltration of crude OMW using a polysulphone  $2 \text{ kDa}$  cut-off membrane. The purpose of this ultrafiltration was to study the effect of electro-Fenton reaction on the toxic fraction of OMW which is composed of LMM phenolics such as simple phenolics (hydroxytyrosol, tyrosol, p-OH benzoic acid, p-OH phenyl acetic acid, vanillic acid, caffeic acid, coumaric acid, vanillin, ferulic acid, catechol, methylcatechol), tannins, antocyanins, catechin (Sayadi et al., 2000). The C18-HPLC chromatogram of this OMW phenolic fraction is presented in Fig. 2.

### 2.2. Electro-Fenton and electro-coagulation treatment

Preliminary experiments were carried out in a  $0.25 \text{ l}^{-1}$  glass reactor for the electro-Fenton of OMW fraction. The aqueous solution of reactants was homogenised by magnetic agitation to avoid concentration gradients. The electro-Fenton reactor was formed by one pair of anodic and cathodic electrodes (cast iron plates) which were positioned approximately  $1.5 \text{ cm}$  apart from each other and were dipped in the effluent. The total effective surface area of electrodes was  $0.2 \text{ dm}^2$ . The current input was supplied by a convergy power supply. In each run, approximately  $0.2 \text{ l}$  of OMW fraction was placed in the electrolytic cell. The pH of the solution was adjusted to 4.  $\text{H}_2\text{O}_2$  was added to the electrolytic cell before the electrical current was turned on. A batch study was conducted to optimise parameters like  $\text{H}_2\text{O}_2$  concentration and current density governing the electro-Fenton process. These parameters were examined in the range of  $0\text{--}1.5 \text{ g l}^{-1}$  and  $1.25\text{--}10 \text{ A dm}^{-2}$ , respectively. The optimum  $\text{H}_2\text{O}_2$  concentration and current density were found to be  $1 \text{ g l}^{-1}$  and  $7.5 \text{ A dm}^{-2}$ , respectively. At these conditions, maximum removal of monomer concentration, COD and colour were attained. For this reason, these conditions were chosen as the optimised parameters and were subsequently used for preparing the pre-treated OMW for the biomethanisation. Experiments of electro-Fenton of crude OMW were conducted in a  $5 \text{ l}$  glass reactor using iron electrodes having an effective surface area of  $150 \text{ dm}^2$  (Fig. 1). In each run,  $3 \text{ l}$  of crude OMW were treated and operated in batch mode.

Electro-coagulation of anaerobic effluent was carried out in the same reactor as for the electro-Fenton of crude OMW without stirring. This electrolysis process lasted  $2 \text{ h}$  at  $1.8 \text{ A dm}^{-2}$  and without adjustment of pH.

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