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Disinfection of wastewater effluents with the Fenton-like process induced by electromagnetic fields

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ABSTRACT

This research work is focused on the application and assessment of effectiveness of the Fenton-like processes induced by radiofrequency for the inactivation of faecal bacteria (*Escherichia coli* and *Enterococcus* sp.) present in treated urban wastewater effluents. Fenton processes were carried out at near neutral pH (pH 5) with different iron sources, such as iron salts (ferric chloride, 5, 50 and 100 mg/L Fe^{3+}), magnetite (1 g/L) and clay (80 g/L), hydrogen peroxide (25 mg/L) and in absence and presence of radiofrequency. Two different electromagnetic field intensities (1.57 and 3.68 kA/m) were used in Fenton processes induced by radiofrequency. Different agents used in the Fenton processes induced by electromagnetic fields (iron source, hydrogen peroxide and RF) were analyzed individually and in combination under the same experimental conditions. First assays of ferromagnetic material/H_2O_2/radiofrequency processes achieved promising results in terms of bacterial inactivation. For instance, Fe^{3+}/H_2O_2/Radiofrequency achieved a maximum level of *E. coli* inactivation of 3.55 log after 10 min of treatment. These results are higher than those obtained in absence of radiofrequency. The thermal activation of iron atoms allows the Fenton reaction to intensify, increasing the final yield of the treatment. On the other hand, different behavior was observed in the inactivation of *E. coli* and *Enterococcus* sp. due to the structural differences between Gram-negative and Gram-positive bacteria.

1. Introduction

Despite that biological pollution is an important part of the wastewater composition, generally the municipal wastewater treatment plants are not designed with the aim to remove it (Mosteo et al., 2013). The origin of this type of pollution derives from excrements of animals and human beings, and their consequences mainly depend of the kind of microorganisms,

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their concentration and the use of this polluted water. The urban wastewaters present a highly variable microbiological composition, depending on factors such as size of population or main economic activity. As a consequence, in recent years the reuse of treated urban wastewater is gaining in importance.

The presence of biological pollutants and hazardous persistent organic and inorganic compounds, can limit not only the discharge to public water, but also their reuse. The Spanish Royal Decree 1620/2007 regulates the reuse of treated urban wastewater in Spain. This establishes the maximum concentrations of contaminants permitted taking into account the type of water reuse: urban, agricultural, industrial, recreational or environmental. The pathogens controlled by this law are *Escherichia coli* and intestinal nematodes in all cases, and Legionella sp. and Salmonella sp. in several cases such as industrial use.

E. coli is the most commonly used indicator of faecal contamination in the quality control of water and wastewater. These Gram-negative bacteria inhabit the intestinal tract of humans and warm-blooded animals. Their presence in water samples is almost always indicative of faecal pollution and the possible existence of other enteric pathogens such as *Salmonella* spp., *Yersinia* spp., *Shigella* sp., etc. These enteric bacteria are responsible for minor gastrointestinal diseases. However, depending on the strain virulence, severe illness or death can occur. Another faecal pollution indicator used in the quality control of water is *Enterococcus faecalis* (Gram-positive). The use of *Enterococcus* sp. as an indicator, bacteria is frequently suggested as an alternative to *E. coli*. Their main advantage lies in their greater resistance and their inability to grow in some environments such as soil, water and others. It is therefore important to develop efficient methods to inactivate *Enterococcus* sp. and *E. coli* prior to water reuse. We have modified the caption of Fig. 2.

In order to obey the current legislation (RD 1620/2007), it is often required to treat water with filtration techniques (conventional and membrane processes), chemical precipitation, disinfection (chlorination, ozonization, UV-C irradiation), activated carbon adsorption, ionic exchange, ponds, wetlands and other biological systems. The application of Advanced Oxidation Processes could be used as an alternative to conventional technology in the removal of hazardous pollutants and microorganisms' pathogens present in wastewater, enhancing the reuse and decreasing the risk to human health and the environment.

The Fenton process has its origin in the discovery, reported in 1894, that ferrous ions strongly promote the oxidation of tartaric acid by hydrogen peroxide. The main reactions involved in the Fenton process are as follows (Barb et al., 1951):

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH^\bullet \ (k = 70\ M^{-1}\ s^{-1}) \quad (1)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^\bullet + H^+ \left(k = 1 - 2 \times 10^{-2}\ M^{-1}s^{-1}\right) \quad (2)$$

$$OH^\bullet + H_2O_2 \rightarrow HO_2^\bullet + H_2O \ \left(k = 1.7 - 4.5 \times 10^{-7}\ M^{-1}s^{-1}\right) \quad (3)$$

$$OH^\bullet + Fe^{2+} \rightarrow Fe^{3+} + OH^- \ (k = 3.2 \times 10^8\ M^{-1}\ s^{-1}) \quad (4)$$

$$Fe^{3+} + HO_2^\bullet \rightarrow Fe^{2+} + O_2 + H^+ \left(k = 1.2 \times 10^6\ M^{-1}s^{-1}\right) \text{ at pH 3} \quad (5)$$

$$Fe^{2+} + HO_2^\bullet + H^+ \rightarrow Fe^{3+} + H_2O_2 \left(k = 1.3 \times 10^6\ M^{-1}s^{-1}\right) \text{ at pH 3} \quad (6)$$

$$HO_2^\bullet + HO_2^\bullet \rightarrow H_2O_2 + O_2 \quad (7)$$

where k is the second order rate constant.

In the presence of dissolved iron ions (Fe^{2+}/Fe^{3+}), hydrogen peroxide is rapidly and efficiently decomposed, producing $OH^\bullet$ radicals (Eqs. (1) and (2)).

It is well known that at a pH of 2.8 the amount of dissolved iron in water is higher than at pH 5, and that when almost all iron added to water is dissolved, the formation of hydroxyl radicals increases (Pignatello et al., 2006). For this reason, the scientific community has established a pH of around 2.8 as the optimal pH in Fenton and photo-Fenton reactions. However, the process can also be carried out at near neutral pH (pH 5) avoiding the costs and drawbacks of acidifying and subsequent neutralization (Aleksic et al., 2010).

The use of radiofrequency (RF) in the treatment of urban wastewater is to date a research field which is considered to be largely unexplored, with the actuation mechanism yet to be fully understood. In recent years, some authors have applied the use of electromagnetic fields in the treatment of wastewater, i.e. combined with biological processes with the aim to increase the activity of the microbial fauna (Ji et al., 2010), electrocoagulation in the removal of humic acids (Ghernaout et al., 2009), in combination with hydrogen peroxide in the removal of organic pollutants (Sobiecka et al., 2008), and in the combination with Fenton processes in the removal of 4-chlorophenol and other hazardous pollutants (Krzemieniewski et al., 2003, 2004; Dębowski et al., 2007; Sobiecki et al., 2008, Kim et al., 2011).

The generation of hydroxyl radicals in the Fenton process increases with the increase of water temperature. However, this fact only occurs up to a certain point (Zazo et al., 2011), as well as, the increase of temperature produces the undesired thermal decomposition of hydrogen peroxide in water and oxygen. However, this fact has a high energetic consumption, which could be decreased with a specific thermal activation of iron, an active substance in the oxidant radical's generation. The thermal activation is produced by means of electromagnetic energy, induced by an electronic device, which activates ferromagnetic substances used in the Fenton process. Induction heating is a non-contact heating process that uses high frequency to heat materials which are electrically conductive. Induction heating is a high efficiency process traditionally used in industrial, medical and domestic applications (induction stove), etc. Several authors have reported the heating of conductive materials mainly due to the hysteresis effect and Foucault (Mühlbauer, 2008; Davies, 1990; Jiles and Atherton, 1986; Naidu, 1991).

The aim of this work was to evaluate the efficiency of Fenton processes induced by electromagnetic fields at near neutral pH (pH 5) to remove *E. coli* and *Enterococcus* sp. added in simulated wastewater effluents. Several iron sources (5, 50 and 100 mg/L Fe^{3+}; 1 g/L magnetite; 80 g/L clay) were evaluated in presence and absence of hydrogen peroxide (25 mg/L) and electromagnetic fields (1.57 and 3.68 k Am).