



Investigation of acid red 88 oxidation in water by means of electro-Fenton method for water purification



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HIGHLIGHTS

- Oxidation of AR88 with hydroxyl radicals was investigated for water purification.
- Rate constant of oxidation of AR88 was calculated as $(1.57 \pm 0.06) \times 10^{10} \text{ M}^{-1} \text{ s}^{-1}$.
- Intermediates of AR88 were determined by spectroscopic and chromatographic methods.
- Effects of current on the evolutions of formed carboxylic acids were examined.
- A general reaction route for AR88 oxidation was proposed for the first time.

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ABSTRACT

In this study, electro-Fenton method was applied to acid red 88 (AR88) containing aqueous solutions for the removal of it from water. The mineralization of AR88 has been achieved by oxidation with hydroxyl radicals. These radicals were produced simultaneously by the electro-Fenton method using an electro-chemical cell including a carbon felt cathode and a platinum anode. Applied current and concentrations of catalyst and supporting electrolyte were optimized to obtain the best effective parameters of 500 mA, 0.1 mM and 75 mM, respectively. The absolute rate constant for the oxidation reaction of AR88 with hydroxyl radical was determined as $(1.57 \pm 0.06) \times 10^{10} \text{ M}^{-1} \text{ s}^{-1}$. Total organic carbon (TOC) analysis was performed to determine whether the organics were converted to carbon dioxide or not. A two-hour electrolysis at 500 mA is enough to remove 87% of initial TOC values of 0.25 mM AR88 solution. Electro-Fenton treatment of AR88 led to the formation of five aromatic intermediates, five short-chain carboxylic acids and three inorganic ions. Identified intermediates and complete mineralization of AR88 allowed us to propose a mineralization pathway for first time in the literature.

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1. Introduction

Azo dyes, a group of synthetic dyes, contain an azo group ($-\text{N}=\text{N}-$) and aromatic groups to form different colors (Wu and Wang, 2001; Liu et al., 2013). They are used widely in many industries such as textile, paper, printing, dye, pharmaceutical, food, cosmetics, plastic and electronic industries and, they constitute almost 30% of total dye markets (Erkurt, 2010; Naveenraj et al., 2013). Most of azo dyes have been released to environment after use in an industrial process. Discharging of them to nature brings a lot of harmful effects on the released ecosystems since some of them are toxic, mutagenic and carcinogenic agents (Naveenraj et al., 2013).

Moreover, they show resistance to biodegradation in the environment because of their stable chemical structures.

Acid Red 88 (AR88) is the first red azo dye, discovered in latest 19th century by BASF (Chudgar and Oakes, 2003). It has been widely used in a lot of industries like other azo dyes. Therefore, it continuously subjected to water and environment. The removal of AR88 from water systems has a great importance because of its different effects such as carcinogenic and toxic effects on the released environment and ecological systems. In this frame, photocatalytic degradation of AR88 has been investigated (Anandan et al., 2008). While photocatalytic degradation is a useful method, the degradation rate of AR88 is too slow. Furthermore, the use of high amount of catalyst and the high cost of irradiation sources (UV lamps) limit the application of photocatalytic methods. The combination of photocatalysis and sonolysis was also searched for the

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degradation of AR88 (Madhavan et al., 2010). Although the degradation rate of AR88 increased in this process, the total degraded amount of AR88 was still too low. Another classical method is the adsorption of AR88 on different materials. In this manner, magnetic $ZnFe_2O_4$ spinel ferrite nanoparticles was used as an adsorbent for the removal of AR88 from water (Konicki et al., 2013). However, the capacity of the adsorbent was relatively low and as a result of this some of AR88 still remained in the water. Azolla rongpong, a fresh water macro alga, was used a biosorbent for the purification of water (Padmesh et al., 2006). Biosorption has the same limitations with the adsorption processes. Biodegradation was also used for AR88 removal from water (Khehra et al., 2006). In this process, degradation rate was too slow and it takes days to remove a small amount of AR88. Moreover, this process brings new pollutants to treated water systems. Finally, the hydrodynamic cavitation process was applied to AR88 solutions containing Fe–TiO₂ as a catalyst and hydrogen peroxide as an oxidant (Saharan et al., 2012). The use of high amount of catalyst (1.8 g L⁻¹) and hazardous chemicals like hydrogen peroxide and not reaching complete degradation are the disadvantages of this method.

The development of advanced oxidation processes (AOPs) has lately allowed the effective removal of persistent organic pollutants from water or soil systems. These methods can be characterized by the formation of highly reactive species, mainly hydroxyl radical ($\cdot OH$). This radical can be obtained by different methods such as chemical, electrochemical, and photochemical methods (Swaminathan et al., 2003; Özcan et al., 2008a; Brillas et al., 2009; Oturan and Aaron, 2014). This radical is the main oxidant in these systems and it reacts with resistant organic molecules and leads to their degradation by dehydrogenation, redox or hydroxylation reactions (Brillas et al., 2009). Electrochemical methods are good alternatives for the production of hydroxyl radical (Brillas et al., 2009;

Oturan et al., 2011; Sirés and Brillas, 2012; Oturan, 2014; Sirés et al., 2014; Ammar et al., 2015).

Electro-Fenton method, one of the most important electrochemical advanced oxidation processes (EAOPs), is a very useful method for the electrochemical production of hydroxyl radicals. In this method, hydroxyl radical is formed electro-catalytically by the Fenton's reaction (Eq. (1)) (Sirés et al., 2007; Özcan et al., 2008b, 2009a). This reaction requires the mixing of hydrogen peroxide and Fe^{2+} to form hydroxyl radical (Eq. (1)). In the electro-Fenton process, this reaction is conducted catalytically via electrochemistry by forming hydrogen peroxide (Eq. (2)) and Fe^{2+} (Eq. (3)) from the reduction of oxygen and Fe^{3+} on a cathode surface, respectively. Some of the used oxygen is formed on the anode surface at the same time (Eq. (4)). The obtained hydroxyl radical can be used for the effective degradation of organic pollutants (Zhou et al., 2012; Loaiza-Ambuludi et al., 2013a). This method is a good alternative to whole decomposition of hazardous substances to protect the environment and eco-system since it is a fast and economical method and doesn't need any harmful chemical. In this frame, electro-Fenton and solar electro-Fenton methods were used for the degradation of AR88 in a flow system with a boron doped diamond anode and carbon-PTFE air-diffusion cathode (Ruiz et al., 2011). Some short-chain carboxylic acids and inorganic ions were determined as degradation products, but determination of aromatic intermediates and degradation rate constant of AR88 have not been reported yet.

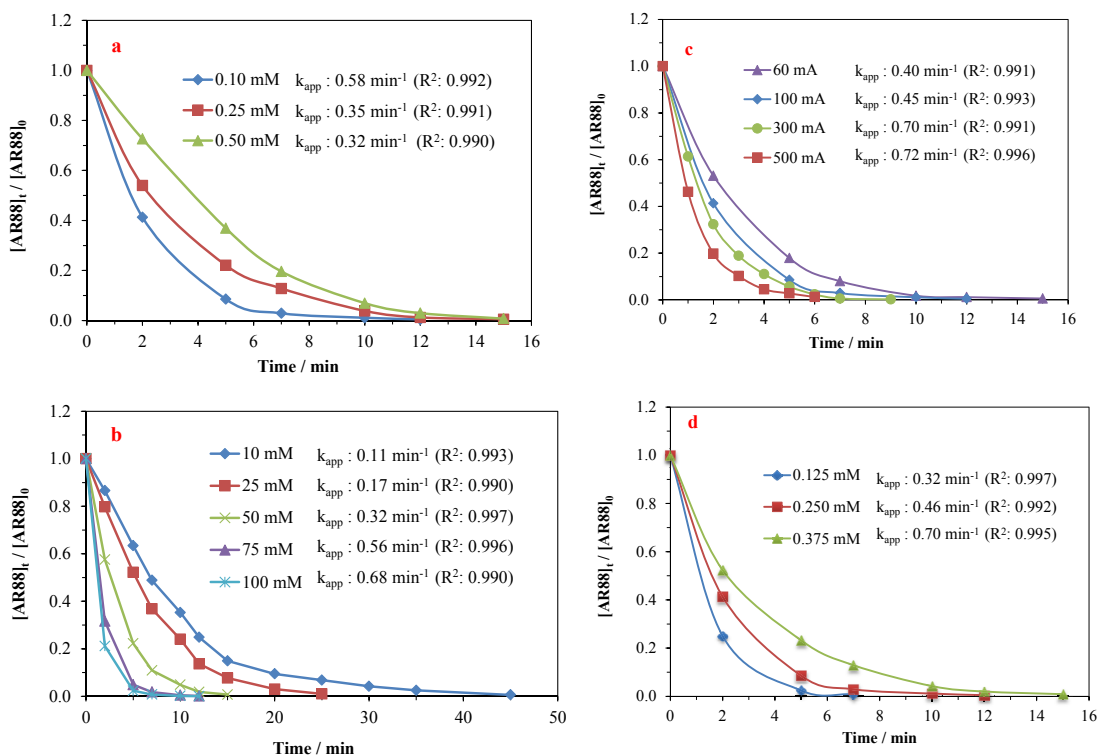
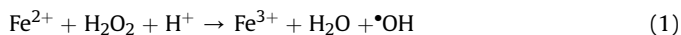


Fig. 1. Normalized concentration-time profiles of AR88 solutions (0.175 L) during the electro-Fenton treatment in aqueous media (pH: 3) at room temperature (25 °C). Experimental conditions; a) $[AR88]_0 = 0.25$ mM, $[Na_2SO_4] = 0.05$ M, I: 100 mA, $[Fe^{3+}] = 0.1$ mM (\blacklozenge), 0.25 mM (\blacksquare) and 0.50 mM (\blacktriangle). b) $[AR88]_0 = 0.25$ mM, $[Fe^{3+}] = 0.2$ mM, I: 100 mA, $[Na_2SO_4] = 10$ mM (\blacklozenge), 25 mM (\blacksquare), 50 mM (\times), 75 mM (\blacktriangle) and 100 mM (Ж). c) $[AR88]_0 = 0.25$ mM, $[Na_2SO_4] = 0.075$ M, $[Fe^{3+}] = 0.1$ mM, I: 60 mA (\blacktriangle), 100 mA (\blacklozenge), 300 mA (\bullet) and 500 mA (\blacksquare). d) I: 100 mA, $[Na_2SO_4] = 0.075$ M, $[Fe^{3+}] = 0.1$ mM, $[AR88]_0 = 0.125$ mM (\blacklozenge), 0.25 mM (\blacksquare) and 0.375 mM (\blacktriangle).

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