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ASSESSMENT OF A Cu-PEROVSKITE MATERIAL IN A HETEROGENEOUS ELECTRO-FENTON PROCESS FOR THE DEGRADATION OF ORGANIC DYES CONTAMINANTS IN A WIDE RANGE OF pH

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These days, the textile industry produces large volumes of tinted wastewater containing a high amount of organic contaminants (Pazdzior et al., 2018). Among them, dyes are considered particularly relevant in industrial effluents due to it is difficult to use conventional treatment technologies for their elimination (Cintra Fernandes et al., 2018; Cruz-Rizo et al., 2017). Some processes such as coagulation-floculation, flotation, adsorption, membranes or reverse osmosis have been reported for the dyes removal. However, they are not effective for the complete decoloration treatment (Cintra Fernandes et al., 2018; Cruz-Rizo et al., 2017; Saleh and Taufik, 2019a). Alternatively, advanced oxidation processes (AOPs), mainly photochemical and electrochemical systems, have showed promising potential for removal of dyes from industrial wastewater streams (Saleh and Taufik, 2019b; Poza-Nogueriras et al., 2018). The electro-Fenton process has been received much attention in order to improve the classical Fenton system by in-situ hydrogen peroxide electrogenereration in the cathode via reduction of dissolved oxygen. Until now, most of the works reported in literature have studied homogeneous electro-Fenton systems under acid pH conditions (Poza-Nogueriras et al., 2018). Current efforts are focused on the development of active heterogeneous catalyst in a wider range of pH. Perovskite-type oxides (ABO_3) provides remarkable red-ox properties and highly mobility of oxygen and electrons through their crystaline structures (Carrasco et al., 2016). These features makes perovskite-type oxides promising heterogeneous Fenton-like catalysts. At present, the use of perovskite materials in electro-Fenton-like reactions is scarce and limited to acid pH conditions (Ben Hammouda et al., 2019). Thus, the aim of this work is the evaluation of Cu-perovskite material ($LaCu_{0.5}Mn_{0.5}O_3$) as heterogeneous electro-Fenton-like catalyst in the degradation of methylene blue (MB) as model pollutant.

$LaCu_{0.5}Mn_{0.5}O_3$ perovkiste was synthetized following a modification of Pecini method proposed by Carrasco et al. (2016). Electro-Fenton experiments were performed in a undivided batch electrochemical chamber working at room temperature (22 ± 5 ºC) under continuous stirring (200 rpm). The cathode was a graphite plate (9 cm^2) and the anode was titanium mesh coated Iridium mix metal oxides (Magneto special anodes B.V., The Netherlands) with a separation gap of 2 cm between them. A direct current of 2 V was supplied by a power supply (HQ PS3003, Helmholt Elektronik A/S, Denmark). Both electrodes were connected through a titanium wire of 1 mm diameter with an external resistor of 10 Ω for monitoring the current intensity in the chamber along the reaction time. The chamber was continously bubbled with air (20 mL/min) by a peristaltic pump. In a typical run, 130 mL of a methylene blue solution (5-10 mg/L) in mixture with 50 mM of Na_2SO_4 was placed in the chamber after appropriate

adjustment of the pH (3, 5 or 7). Powder Cu-perovskite was added in the desired catalytic loading (0.6-2.5 g/L). Afterthat, the power supply was turned on, starting the reaction. Several aliquots were withdrawn along the reaction for the determination of MB removal as indicator of the catalyst activity.

Different experiments were carried out to assess the effect of two operating conditions: pH and perovskite dosage. **Figure 1** shows the evolution of methylene blue concentration for electro-Fenton experiments performed at different pH **(a)** and perovskite concentration **(b)**.

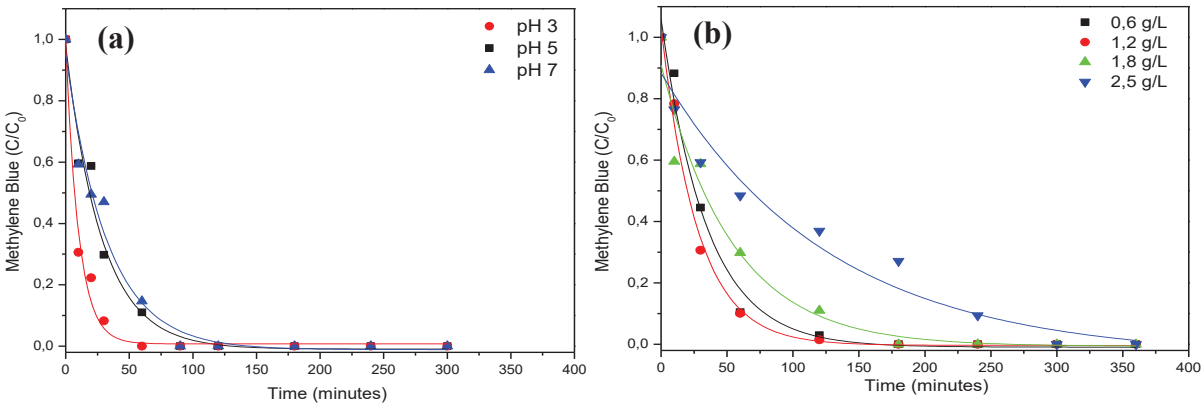

Figure 1. Removal of MB (**a**) at pH 3, 5 and 7 with initial concentartion of $LaCu_{0.5}Mn_{0.5}O_3$ of 1.85 g/L and $[MB]_0$= 5 mg/L; (**b**) at diferent perovskite concentration (0.6, 1.2, 1.85 and 2.5 g/L) with pH 5, $[MB]_0$= 10 mg/L.

A total decoloration of MB was obtained after 30 minutes at pH 3, but it was also noteworthy the activity of $LaCu_{0.5}Mn_{0.5}O_3$ at higher pH conditions of 5 and 7. This fact is particularly remarkable as efficiency of homogeneous electro-Fenton systems is normally limited to acid pH about 2-3 (Ben Hammouda et al., 2019). The current intensity in these reactions were in the range of 5 and 10 mA (0.55 and 1.1 mA/cm^2). The decrease of the catalyst concentration until 0.6 g/L allowed a better electrocatalytic performance of Cu-perovskite. These results seem to be also in agreement with the data reported in literature (Ben Hammouda et al., 2019), as an excess of electro-active metal species can promote the scavenging of hydroxil radicals. Additionallly, the presence of powder heterogeneous particles of Cu-perovskite could increase the internal resistance for the electrons transfer from anode to cathode.

As conclusion, $LaCu_{0.5}Mn_{0.5}O_3$ showed a remarkable activity for the removal of MB at circumneutral pH. These results suggest that the perovskite would be a promising option for organic compounds removal by electro-Fenton process working with neutral water pH.

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