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Photodegradation of phenol and cresol in aqueous medium by using Zn/Al + Fe mixed oxides obtained from layered double hydroxides materials

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ABSTRACT

ZnAlFe layered double hydroxides (LDHs) were synthesized by the co-precipitation method at different M_{II}/M_{III} ratio. The solids were calcined at 723 K obtaining the respective mixed oxides with high specific surface areas (138–70 m²/g) and semiconductor properties (band gap values of 2.54–2.04 eV). The photocatalytic activity of these materials was tested for the photodegradation of aqueous solutions containing phenol and *p*-cresol (40 ppm), in presence of a UV light source. A disappearance of 98% of phenol and a total photodegradation of *p*-cresol were obtained after 6 and 4 h, respectively.

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1. Introduction

Due to increasing water demands and/or long periods of drought, removing pollutants from industrial wastewaters is becoming an important area of research, since the quality of drinking water available in the world is decreasing. Additionally, stricter wastewater discharge standards continue to be introduced worldwide, in an effort to reduce the environmental impact of industrial processes [1].

Phenol and phenol derivatives used as raw materials in petrochemical and chemical industries are considered one of the most common organic water pollutants because of its high toxicity, even at low concentrations. Several technologies are available to remove industrial organic wastes, such as biological, thermal and chemical treatments and the named advanced oxidation processes (AOPs) [2–5]. Among the AOPs, the photodegradation, a technique which employs semiconductors as photocatalysts, provides a promising method for the elimination of these pollutants in water [2,6,7]. TiO₂, ZnO and SnO₂ are the semiconductor materials most widely reported as photocatalysts [8–14]. However, layered double hydroxides (LDHs) have been recently reported as a good alternative for the photodegradation of pollutant organic compounds [4]. In particular, ZnAl LDHs are successful photocatalysts for the degradation of organic compounds like methyl-orange [4], methylene blue [15] and phenol [16,17] in aqueous media.

Layered double hydroxides (LDHs) belong to a class of anionic mineral clays, with structure derived from mineral brucite, Mg(OH)₂. When a fraction of Mg²⁺ ions is isomorphously substituted by a trivalent ion such as Al³⁺ or Fe³⁺, the positive charge generated on the metal hydroxide slab is compensated by the inclusion of anions generating the general formula $[M_{1-x}^{2+}M_x^{3+}(\text{OH})_2]^{x+}(A^{n-})_{x/n} \cdot y \cdot \text{H}_2\text{O}$, where M²⁺ and M³⁺ are divalent and trivalent metal ions, respectively, and Aⁿ⁻ is an intercalate anion, being CO₃²⁻ the most common [18–20]. By means of a controlled thermal decomposition, the LDHs were transformed into the respective mixed oxides.

Since ZnO [12,13] and Fe₂O₃ oxides [21,22] are well known as photocatalytically active semiconductors, in the present work the synthesis by the co-precipitation method of ZnAlFe LDHs is reported. The obtained materials were characterized by X-ray diffraction (XRD), nitrogen adsorption and UV–vis spectroscopy. The photocatalytic activity of the solids was tested towards the decomposition of the organic pollutants phenol and cresol in aqueous medium.

2. Experimental

Double layered materials containing ZnAlFe at different M_{II}/M_{III} ratio were prepared at constant pH by the co-precipitation method, using aqueous solutions of Zn(NO₃)₂·6H₂O, Al(NO₃)₃·9H₂O and Fe(NO₃)₃·9H₂O (J.T. Baker Analyzed Reagent) as a source of metals. The solutions were added dropwise in a glass reactor vessel containing 800 mL of bidistilled water under vigorous magnetic stirring; the pH of the solution was adjusted to 9 by adding NH₂CONH₂ as precipitant agent. The resulting suspension was

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