

## Adsorption of anionic dyes from aqueous solution on zeolite from fly ash-iron oxide magnetic nanocomposite

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**Abstract.** Magnetic zeolite/iron oxide nanocomposite was prepared by mixing zeolite synthesized from coal fly ashes with magnetite nanoparticles in suspension and was used for the removal of Reactive Orange 16 (RO16) and Indigo Carmine (IC) from aqueous solutions. The effect of various experimental parameters such as contact time, pH, adsorbent dose and temperature were investigated. The experimental data were analyzed using the pseudo-first-order and pseudo-second-order adsorption kinetic models. The experimental data fit the second-order kinetic model. The Langmuir and Freundlich isotherm models were tested for their applicability. Results indicated that according to the Langmuir isotherm, the maximum sorption capacities are 1.1 and 0.58 mg·g<sup>-1</sup> for RO16 and IC, respectively. Thermodynamic parameters showed that adsorption of dyes were endothermic and spontaneous in nature.

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**Key words:** magnetically modified zeolite, coal fly ash, nanocomposite, adsorption, dye removal

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## 1 Introduction

Magnetic separation technology as an efficient, fast and economical method for separating magnetic materials has been widely used in textile, biology, and environmental protection [1–4]. Adsorption is considered a simple and economical technique for wastewater treatment [5, 6].

The adsorbents combining magnetic separation technology with adsorption process have been widely used in environmental purification [7–9]. The main advantage of this technology is that it can dispose a mass of wastewater in a very short period of time and produce no contaminants such as flocculants [10]. Some examples are the use of activated carbon/iron oxide

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magnetic composites for the adsorption of volatile organic compounds [11], montmorillonite-iron oxide magnetic composites for the adsorption of metal cations [12] and NaY zeolite-iron oxide magnetic composite for removal of metallic contaminants from aqueous solution [13].

Most commercially available magnetic particles are rather expensive and cannot be used for large-scale processes. Magnetic modification of low cost adsorbents could lead to materials suitable for biotechnology and environmental applications.

Zeolite synthesized from Brazilian coal fly ashes offer an attractive and inexpensive material option for removal of contaminants. In our group zeolites synthesized from Brazilian coal fly ashes have been used as low-cost adsorbents to remove contaminants from aqueous effluents [14, 15] and zeolite from fly ash/iron oxide composites were used as adsorbents for the removal of metal ions of  $Zn^{2+}$ ,  $Cd^{2+}$  and  $Pb^{2+}$  from water [16, 17].

In the present study, magnetic nanocomposite of zeolite from fly ash was prepared by simple method and their adsorptive characteristic for the removal of dyes from aqueous solution was studied. The adsorption studies such as effect of contact time, pH, adsorbent dose and temperature were explored in batch experiments and thermodynamic, kinetic and adsorption isotherm analysis were used to elucidate the adsorption mechanism.

## 2 Materials and methods

### 2.1 Materials

All chemicals used in this study were of analytical grade. Reactive Orange 16 (RO16; C.I. 17757;  $C_{20}H_{17}N_3O_{10}S_3Na_2$ ; molar mass =  $601.54 \text{ g}\cdot\text{mol}^{-1}$ ) was purchased from Sigma-Aldrich (50% purity) and Indigo Carmine (IC; C.I. 73015;  $C_{16}H_8N_2Na_2O_8S_2$ ; molar mass =  $466.35 \text{ g}\cdot\text{mol}^{-1}$ ) was supplied by Vetec Quimica Fina Ltda (100% purity). Stock solutions were prepared in deionized water (Millipore Milli-Q) and the solutions for adsorption tests were prepared by diluting. The sample of coal fly ashes from baghouse filter was obtained from a coal-fired power plant located at Figueira City, in Parana State, Brazil.

### 2.2 Preparation of the zeolite from fly ash-iron oxide magnetic composite

First, coal fly ash was used as starting material for zeolite synthesis by means of hydrothermal treatment. In synthesis experiment, 20 g of fly ash was heated to  $100 \text{ }^\circ\text{C}$  in an oven for 24 h with 160 mL of  $3.5 \text{ mol}\cdot\text{L}^{-1}$  NaOH solution. The zeolitic material (ZC) was repeatedly washed with deionized water until pH 11 and dried at  $100 \text{ }^\circ\text{C}$  for 24 h [18]. Magnetite particles were prepared by adding of NaOH solution drop by drop into ferrous sulfate solution with agitation until the pH reached 11. The slurry was heated on a water bath. After that, the magnetite was washed with distilled water and dried at room temperature. The magnetite particles were redispersed in aqueous solution and ZC was added slowly with agitation. The zeolite ZC/magnetite ratio was 3:1(w/w). The obtained zeolite-iron oxide magnetic composite (ZM) was washed with distilled water, dried at room temperature and milled. The black product was attracted toward the magnet (Fig. 1).