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Adsorption of Anionic Dyes Using Monoionic and Binary Systems: a Comparative Study

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Abstract: Layered double hydroxide Mg_2Al-LDH with Mg^{2+}/Al^{3+} molar ratio of 2 was prepared via urea method and used to remove two dyes, namely methyl orange (MO) and Congo red (CR) in monoionic and binary systems by batch adsorption. The results showed by X-ray diffraction analysis of Mg_2Al-LDH confirm the crystal structure of the Mg_2Al-LDH material. The adsorption of MO and CR in binary systems decreased in the case of the mixture, compared to MO and CR alone with 98 and 70% of MO and CR, respectively. According to this study's results, it could be concluded that Mg_2Al-LDH adsorbent can be used effectively to remove anionic dyes from industrial wastewater.

Keywords: monoionic and binary systems; anionic dyes; adsorption; wastewater treatment.

1. Introduction

Because of their accumulation (in both effluents and the environment), the use of chemicals leads to serious environmental problems directly or indirectly (chronic toxicity). These issues arise from these compounds or their products of degradation. It is estimated that more than $7x10^5$ tons and about 10000 different kinds of dyes and pigments are produced yearly worldwide. It is estimated that 10 to 15% of the dye is lost in effluents [1]. A wide range of techniques has been developed to remove dyes from wastewater, such as coagulation/flocculation [2,3], oxidation/ozonation [4], membrane separation [5,6], photodegradation [7] along with biological processes [8]. Adsorption is one of the most popular and widely practiced due to its low-cost, ease of operation, high efficiency and efficiency, and a wide range of implementation in various conditions [9]. Adsorption processes are reported as an effective and efficient method to decolorize wastewater. It involves the transfer of dye particles on the adsorbent, resulting in clearer effluent [10].

Layered double hydroxides (LDHs) are intensively investigated because of their high anionic exchange capacity [11], reuse, larger surface area, porosity, and fundamental properties [12]. They have advantages over commercially available adsorbents in terms of low cost, high adsorption properties, and non-toxicity. The use of LDH could bring significant economic and environmental benefits to the wastewater treatment industries [13].

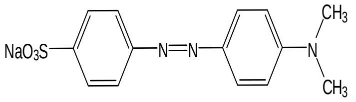
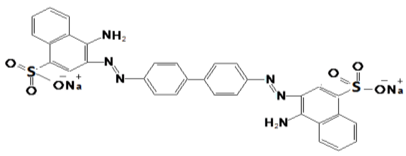
To our knowledge, little is known about MO and CR's use for the adsorption of synthetic clay, hence the originality of this work. The main aim of this work was to investigate the adsorption of MO and CR onto the synthetic clay to evaluate the behavior of two dyes in mono-ionic and binary systems

2. Materials and Methods

2.1. Preparation of methyl orange and Congo red dyes.

Methyl orange and Congo red used in this study were purchased from Panreac (Barcelona, Spain). A Stock solution of 1g.L^{-1} in methyl orange and Congo red dyes were prepared with double-distilled water. The working solutions were obtained by diluting the stock solution. Table 1 shows the molecular structures and some physicochemical characteristics of used dyes

Table 1. Physicochemical characteristics of used methyl orange and Congo red dyes.

Name (nm)	**Molecular structure**	**Nature**	**Molar mass (g. mol^{-1})**	**λ_{max}**
Methyl Orange		Anionic	327.3	464
Congo red		Anionic	696.66	497

2.2. Preparation and characterization of Mg_2Al-LDH.

The same conditions and experimental description from synthesized adsorbent Mg_2Al-LDH were given in detail in previous reports [1,14].

A detailed characterization of Mg_2Al-LDH adsorbent has been done using different techniques, as was described in our previous report. X-ray diffraction analysis of Mg_2Al-LDH confirms the crystal structure of the Mg_2Al-LDH material [1,15].

Figure 1 presents the SEM images of Mg_2Al-LDH. The two images present the morphology of the platelets, which reflected a good crystallinity.

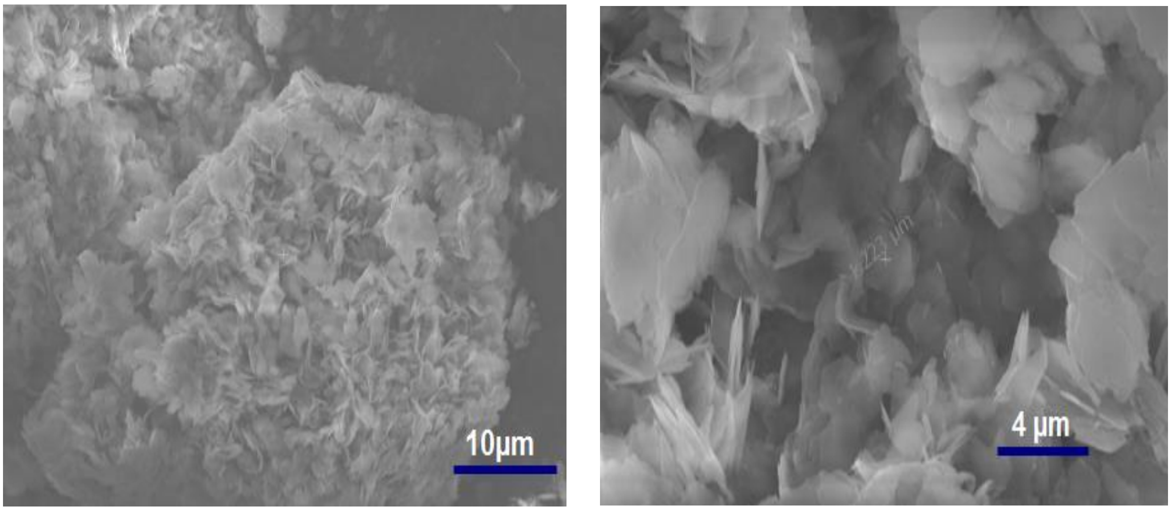


Figure 1. Scanning electron microscopy image of Mg_2Al-LDH material.

Figure 2 displays the infrared spectra of Mg_2Al-LDH, the broad absorption at 3395 cm^{-1} is assigned to O-H stretching of the water molecule in the brucite-like layers [16,17], the weak band at 1632 cm^{-1} is due to the bending vibration (deformation mode of H–O–H (H–O–H)) of interlayer water molecules in LDH [18,19]. The stretching and bending modes of the CO_3^{2-} group are observed at 1354 cm^{-1} [20]. The bands at 652– 455 cm^{-1} regions are ascribed to M-oxygen-M stretching (M= Mg and Al) [21,13].

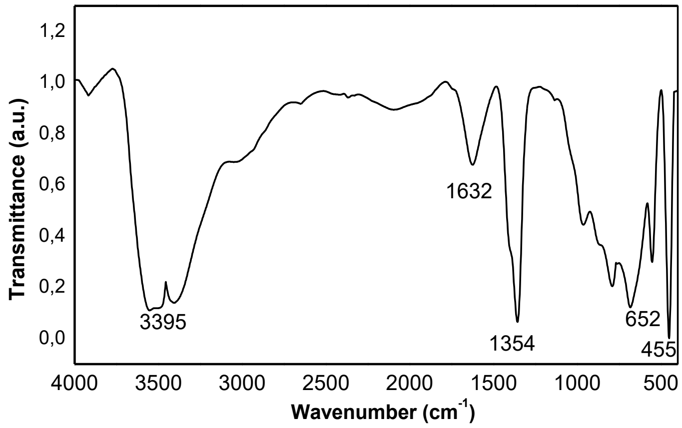


Figure 2. FTIR spectra of Mg_2Al-LDH.

2.3. Batch studies.

The adsorption of methyl orange (MO) and Congo Red (CR) onto synthetic clay in monoionic and binary systems was carried out in a batch system. The adsorption of two dyes was carried out in a thermostatic bath by adding 0.02 g of our adsorbent into a flask containing 40 mL of the mixture of two dyes (MO+CR) by varying the contact time (1–180 min) and composition dye (90% MO+10% CR;70% MO%+30% CR; 50% MO + 50% CR; 70% CR % +30 % MO; 90 % CR+10 % MO). After a suitable time, the dye solution was separated from the adsorbent by centrifugation for 15 min at 3000 rpm.

In monoionic system, the percentage removal of dyes adsorbed at equilibrium was given by the following equation:

$$\text{Removal efficiency (\%)} = \frac{(C_0 - C_e)}{C_0} \times 100 \qquad (1)$$

where C_0 and C_e are the initial and equilibrium concentration (mg.L^{-1}) of dyes in solution.

In a binary mixture of A and B components, the optical densities d_1 and d_2 were measured at λ_1 and λ_2 [22].

$$C_A = \frac{k_{22}d_1 - k_{21}d_2}{k_{11}k_{22} - k_{12}k_{21}} \qquad (2)$$

$$C_B = \frac{k_{11}d_2 - k_{12}d_1}{k_{11}k_{22} - k_{12}k_{21}} \qquad (3)$$

3. Results and Discussion

3.1. Adsorption of MO and CR in monoionic system.

Figure 3 shows the adsorption kinetics in MO and RC by Mg_2Al-LDH at 100 mg. L^{-1}. The equilibrium was reached after 15 min for MO and 120 min for CR, with percentage removal 98 and 70% for MO and CR, respectively.

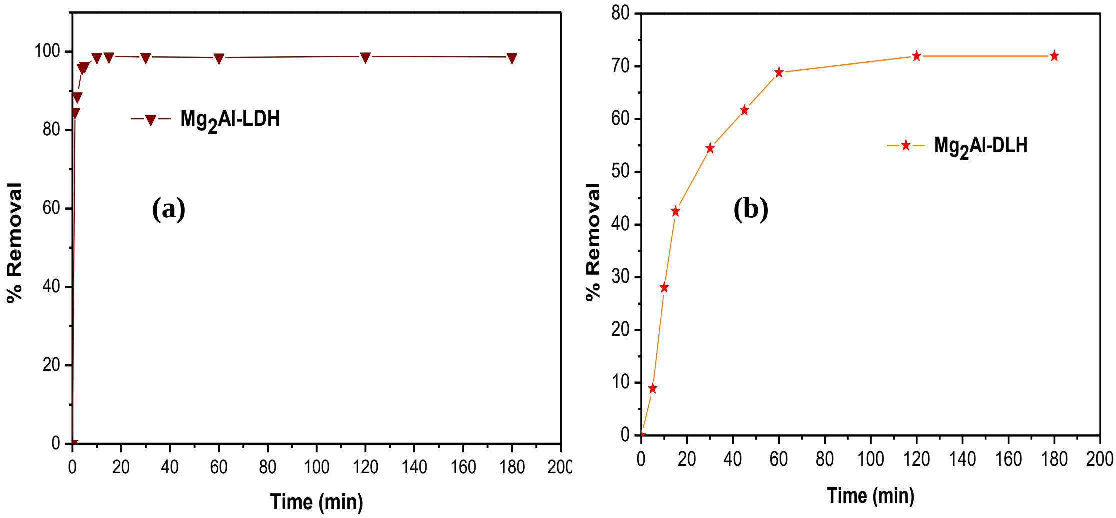


Figure 3. Adsorption kinetics in (**a**) MO; (**b**) RC by monoionic system.

3.2. Adsorption dyes in a binary mixture.

3.2.1. Calibration curve.

The calibration curves of MO and CR at maximum wavelengths: 464 and 497 nm. These curves allow us to determine the calibration constants (Figure 4).

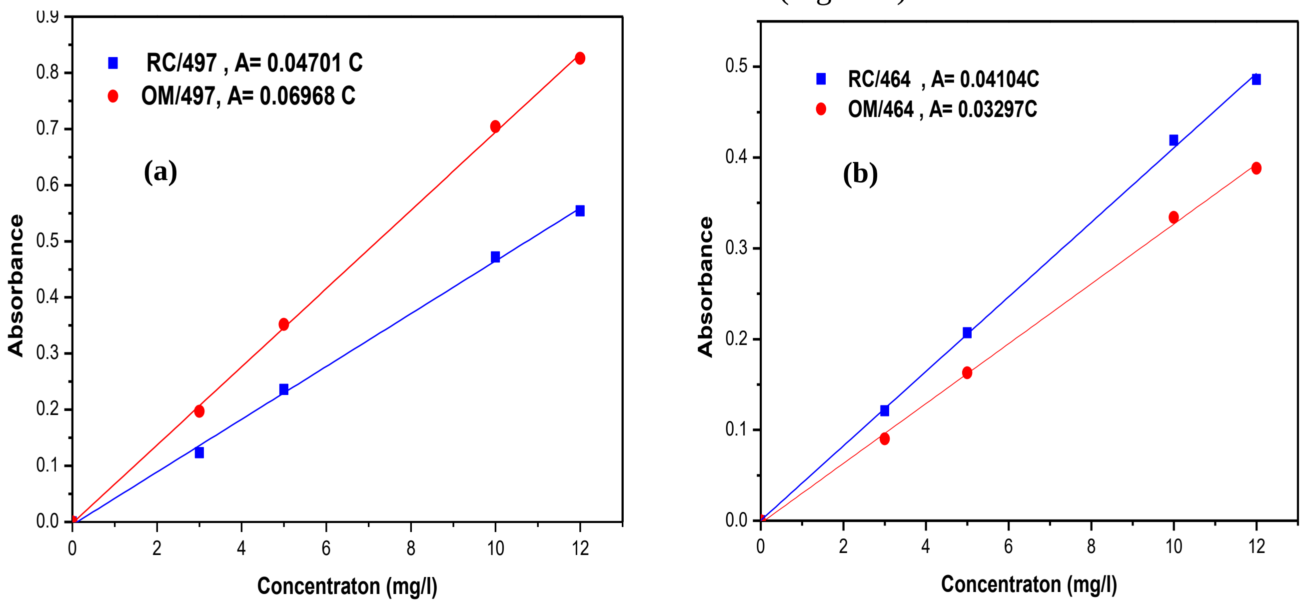


Figure 4. Calibration curve of MO and CR; (**a**) λ_{max}= 497nm ; (**b**) λ_{max} =464nm.

3.2.2. Adsorption of MO and CR in a binary system.

The adsorption of MO and CR dyes onto synthetic clay in a binary system (MO + CR) comparing with the same dyes taken individually has been investigated by varying the percentage of MO and CR in the binary mixture with the adsorbent dose, the initial dye concentration and the contact time have been fixed at 0.5 g.L^{-1}, 100 mg.L^{-1} and 180 min respectively, At the room temperature and the initial dye solution pH. The obtained results are illustrated in Figure 5. While analyzing these results, we noticed that MO and CR's adsorption decreased in the case of mixing, compared to MO and CR alone. This could be explained by the competitive effect between the two anionic dyes.

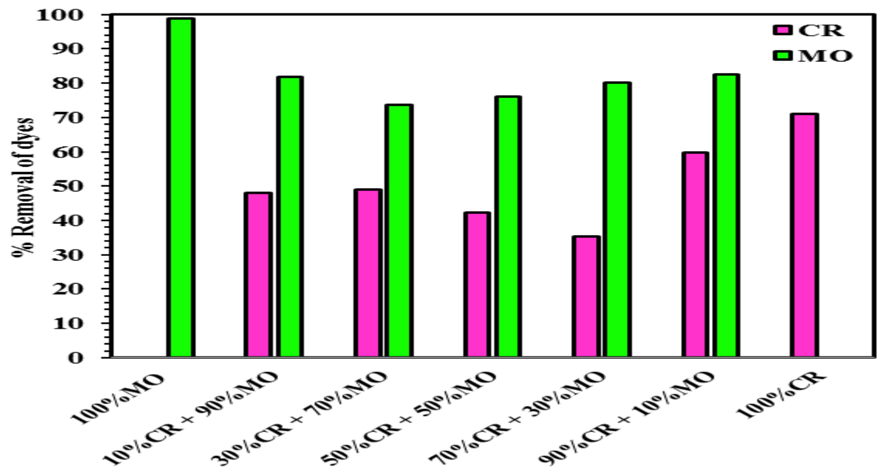


Figure 5. Adsorption of MO and CR in binary systems.

4. Conclusions

In this study, adsorption experiments for removing methyl orange and Congo red from aqueous solutions were performed using monoionic and binary systems. The results also show that these materials were found to be excellent adsorbent for the MO and CR. The adsorption of MO and CR in binary systems decreased in the case of mixing, compared to MO and CR alone. These exciting results offer several perspectives, such as recycling the material used; the synthesized carrier is also be tested for trapping other organic solvents.

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Conflicts of Interest

The present paper is an original work, and all the authors declare that they have no conflicts of interest

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