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Original article

Fast and efficient removal of alizarin yellow dye (azo dye) from water and wastewater samples using modified nanoclay

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ABSTRACT

A fast and efficient method has been developed for removal of Alizarin Yellow dye using modified nanoclay. Montmorillonite (MMT) was modified by a facile and one-step procedure with diethylenetriamine (DETA) and was used as an adsorbent. The effects of pH value of the dye solution, adsorbent dose, adsorption time and the initial dye concentration on the Alizarin Yellow adsorption onto the composite were investigated. The DETA-MMT had a high uptake capacity in room temperature and could remove Alizarin Yellow dye of about 85 % with 6 g/L of adsorbent, in only 2 min. Langmuir and Freundlich isotherms were employed for the study of the adsorption of Alizarin Yellow dye onto DETA-MMT. The method was applied to the removal of Alizarin Yellow in different tap water, river water and industrial wastewater samples.

1. Introduction

Colored wastewater is one of the most important sources of environmental pollution and because of their visibility and bearing toxic chemicals, it is very important to be treated before released into the environment. In recent years, several methods have been used for treatment of dye effluents (Pourreza and Elhami, 2010; Saad, et al. 2010). Synthetic dyes have been used in many industries such as textile, paper printing, food, pharmaceutical, leather and cosmetics. Azo dyes, one of the greatest groups of synthetic dyes, have one or more azo bounds (-N=N-). Removal of azo dyes from colored effluents due to their complex composition, toxicity, poor degradability and high solubility, have attracted great interest (Perju and Dragan, 2010).Conventional methods such as chemical oxidizing method and biological treatment cannot effectively remove of dyes from effluents, so finding an effective technique is necessary (Shiau and pan, 2004). Adsorption is an effective method for removal of dyes. Clays are applied as adsorbents for heavy metal ions (Shahmohammadi-Kalalagh, et al., 2011; Abollino, et al., 2008; Gu, et al., 2010) and dyes (Wang and Wang, 2008; Syed, et al., 2010). Montmorillonite is nanoclay that widely used in various science fields due to their large cation exchange capacity, high specific surface area, good swelling capacity and high platelet aspect ratio and ease with which their surface can be modified (Ahmad, et al., 2009). Presence of inorganic cations such as Na+ and Ca2+ on the basal planar surface of montmorillonite layers makes it hydrophilic in nature and hence, shows the clay ineffective for absorption of aliphatic and relatively anionic compounds. It is possible to modify the surface properties of MMT greatly by replacing the natural inorganic cations with other cation such as metal ions (Abollino, 2008) and quaternary amines (Xi, et al., 2004; manocha, et al., 2007).

In this study, modified montmorillonite with diethylenetriamine (DETA-MMT) was used for removal of alizarin yellow (AY) dye from water samples. Alizarin yellow is a mordant dye, suitable for the dyeing of wool and nylon (Salman, et al., 2011). It usually exists as a sodium salt. The effects of pH value of the dye solution, adsorbent dose, adsorption time and the initial dye concentration on the adsorption of AY on DETA-MMT have been investigated. The adsorption isotherms for AY dye onto DETA-MMT were also studied.

2. Materials and methods

2.1. Materials

All the used chemicals had an analytical grade and doubled distilled water was used throughout the study. Montmorillonite (sodium form) was supplied by F.C.C. (China).

A stock solution of 1000 mg/L of AY dye was prepared by dissolving 0.50 g of the reagent in water and diluting to as much as 500 mL in a volumetric flask. The desired concentrations were obtained by successive dilutions.

2.2. Preparation of adsorbent

The montmorillonite (5 g) was suspended in 500 mL distilled water and stirred magnetically for about 24 h at room temperature. After this period, 5 mL DETA added and the solution pH using hydrochloric acid was adjusted (pH=3.5). The suspension was stirred for 2 h in 50 oC. The white precipitation was filtered and washed several times with distilled water. Then, the precipitation was dried in an oven at 50°C for 48 h.

2.3. General procedure

In each adsorption experiment, 50 mL of dye solution of known concentration and pH was added to modified montmorillonite in a 250 mL erlenmeyer flask in room temperature (25±1 °C) and it was stirred by a shaker at 110 rpm. After 35 min the mixture was centrifuged at 3000 rpm and the concentration of AY in the solution was measured by spectrophotometeric method to be at 374.0 nm with Perkin-Elmer UV Visible Spectrophotometer (model: Lambda35). The percentages of AY removal is calculated based on the following equation:

$$\mathrm{Re}\,moval(\%) = \left(\frac{C_{0-}C_e}{C_0}\right) \times 100 \qquad (1)$$

In this equation, C0 is the initial dye concentration (before being mixed with the adsorbent), and Ce is the equilibrium concentration (after being mixed with the adsorbent). The equilibrium concentration was obtained through a calibration curve for the AY dye.

3. Results and discussion

Diethylenetriamine- montmorillonite was chosen as an adsorbent for removal of alizarin yellow dye. The preliminary experimental observations showed that AY dye is adsorbed on diethylenetriamine- montmorillonite, while montmorillonite did not have this ability.

3.1. Effect of pH

The adsorption of AY dye onto the composite as a function of pH was investigated at the initial dye concentration of 100 mg L−1 and the contact time of 30 min. Since AY dye is precipitated under acidic conditions at pH values lower than 4, the investigations were carried out at pH upper than 4.5. The effect of pH on the adsorption of AY onto composite is shown in Fig. 1. The removal of AY dye was maximum in the pH of 4.5. In the acidic solution, the adsorption process of the anionic dye AY by DETA-MMT is an electrostatic interaction, where the protonated amine groups of DETA-MMT interact with the anionic groups of the dye. While at high pH, more OH− ions present and compete with the anionic groups of AY for the adsorption sites of adsorbent, thus the available adsorption sites for AY decrease.

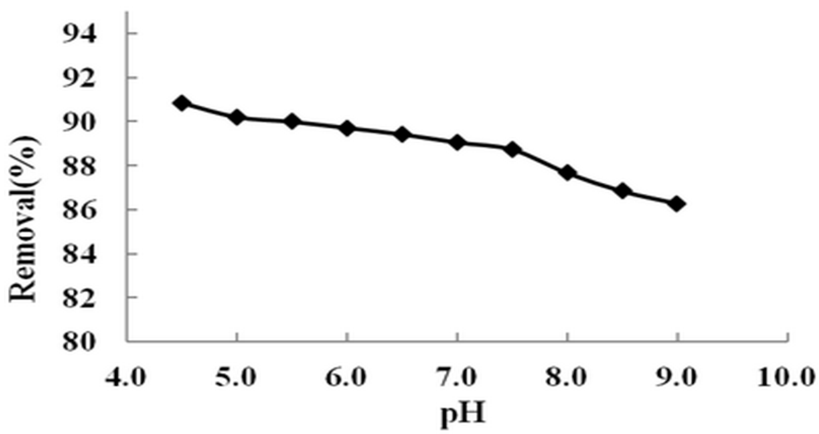

Fig. 1. Effect of pH on AY dye removal.

(Initial dye concentration 100 mg/L, adsorbent dosage=0.3 g/50 mL, temperature 25 °C, contact time=30 min, agitation rate= 120 rpm).

3.2. Effect of adsorbent dose

The influence of adsorbent dose on AY removal was studied by varying the adsorbent dose from 1.0 to 7.0 g L−1 at an initial AY concentration of 100 mg L−1 in 50 mL solutions. Increased adsorbent dosage implied a greater surface area and a greater number of binding sites available for the constant amount of AY. An adsorbent dose of 6 g/L was chosen as an optimum value. The results showed (Fig. 2) that 2 g/L of DETA-MMT is required for the 81% removal of AY from initial concentrations of 100 mg/L. However, for a removal of about 91%, 6 g/L of DETA-MMT was needed.

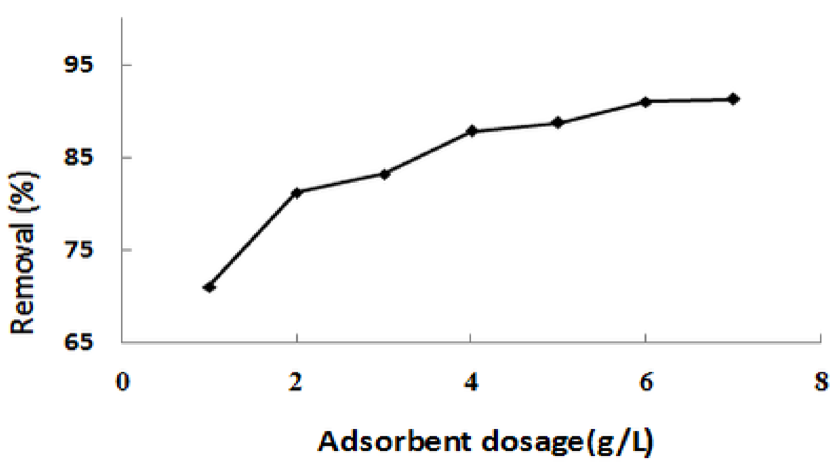

Fig. 2. Effect of adsorbent dose on AY dye removal.

(Initial dye concentration 100 mg/L, pH=4.5, temperature 25 °C, contact time=30 min, agitation rate= 120 rpm).

3.3. Effect of contact time and agitation rate

The adsorption of AY dye onto composite adsorbent has been investigated as a function of time in the range of 2-50 min (Fig. 3). In each adsorption experiment, 50 mL of dye solution of known concentration and pH was added to 0.3 g of adsorbent in 50 mL round bottom flask at room temperature and the mixture was stirred on a shaker at 120 rpm. As the results indicate, removal of AY using the adsorbent occurs quickly and it is not a highly time-dependent process. It was found that about 85% removal of AY occurs within 2 min. However, for a removal of about 90%, a time interval of 40 minutes was needed. This confirms a high and rapid adsorption of AY dye by DETA-MMT.

The agitation speed ranging between 80 and 120 rpm was maintained (Figure not shown). In the all speed, the removal was not varied significantly. An agitation speed of 110 rpm was chosen as an optimum value. The small effect of agitation showed that external mass transfer was not the rate limiting step.

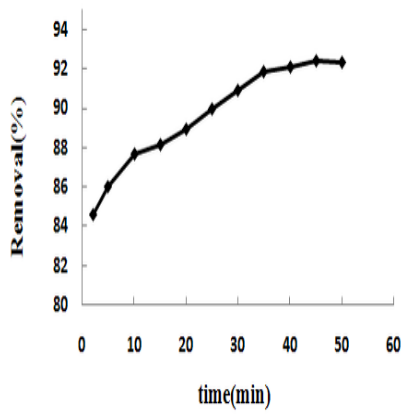

Fig. 3. Effect of contact time on adsorption of AY dye. (adsorbent dosage=0.3 g/50 mL, pH=4.5, temperature=25 °C, agitation rate= 120 rpm).

3.4. Study of initial concentration

In all the cases of optimization, the dye used had a concentration of 100 mg L-1. In order to study the possibility of dye removal in other concentrations with the same optimization condition, other concentrations were studied, as well. The results showed that the optimization condition was applicable for concentrations 10-200 mg L-1 and dye removal will be 92-96%.

3.5. Application

In order to test the reliability of the proposed removal methodology, it was applied to the removal of concentrations of AY from tap water, river water and industrial wastewater samples. For this purpose, 30 mL of each of the samples was treated under the general procedure. Spiking AY to the samples performed the validity of the procedure. The results presented in Table 1 show that good extraction efficiencies are obtained for the removal of AY spiked to river water, tap water and industrial wastewater samples.

3.6. Isotherms of adsorption

In this study, Langmuir and Freundlich isotherms were employed for the study of the adsorption of AY dye onto modified montmorillonite. Such isotherms were achieved for an initial concentration of 10-200 mg L-1 in the previous optimization condition and a temperature of 25±2 0C; and qe is achieved through equation (2):

$$q_e = (C_0 - C_e)\frac{V}{W} \qquad (2)$$

In this equation, qe is the adsorption capacity in an equilibrium of (mg L-1); C0 is the initial concentration of dye (mg L-1); Ce is the equilibrium concentration (mg L-1); V is the volume of solution; and W is the weight of the adsorbent.

In order to study the adsorption of dye according to Langmuir Isotherm the following equation (3) was used, in which Q0 and b are the constants of Langmuir. Ce/qe was plotted vs. Ce. No linear relationship is found between the sorbet concentrations at equilibrium in the solid and liquid phases respectively (Figure not shown).

$$\frac{C_e}{q_e} = \frac{1}{Q_0 b} + \frac{1}{Q_0} C_e \quad (3)$$

The Freundlich model assumes a heterogeneous adsorption surface with sites that have different adsorption energies which are not equally available. This isotherm is achieved through equation (4). In this equation, Kf and n are the empirical constants and their values were obtained from the intercept (log Kf) and slopes (1/n) of linear plots of logqe versus logCe. For this isotherm, logqe was plotted vs. logCe.. The results have been presented in Fig. 4. The Kf and n values in Freundlich equation were found to be 2.49 and 1.1 respectively.

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \quad (4)$$

Table 1
Removal of Alizarin Yellow dye in spiked water samples.

sample	Added dye (mg/L)	Remained dye (mg/L)	Removal (%)
Tap water	50	5.97	88.50
	100	14.83	85.17
	200	20.66	89.67
River water	50	7.24	85.68
	100	11.58	88.42
	200	23.24	88.38
Industrial wastewater	50	5.35	89.29
	100	12.50	87.50
	200	23.78	88.11

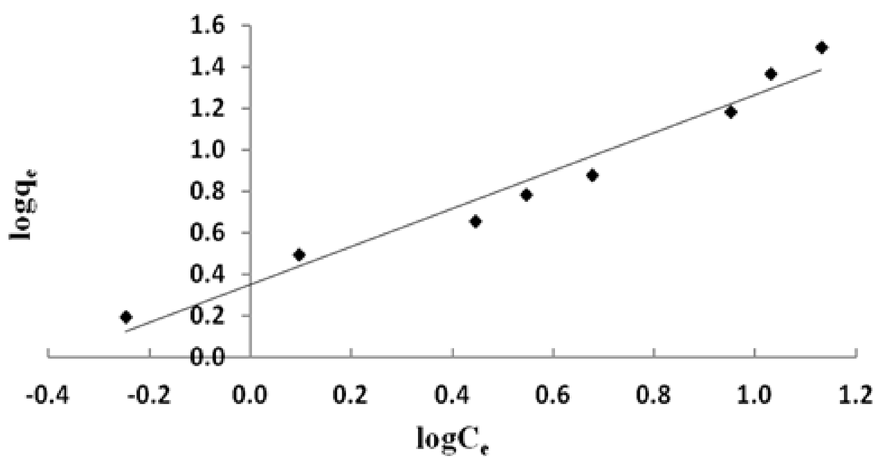

Fig. 4. The plot of AY adsorption on Freundlich isotherm model.

4. Conclusions

The present investigation evaluated the fact that the chemically modified montmorillonite with diethylenetriamine can be used as an effective and fast adsorbent for the removal of AY dye, azo dye, from different water samples. Contact time, adsorbent dosage and pH are the most effective parameters on adsorption of dyes. The effects of these parameters on the adsorption of AY dye was examined by optimal experimental conditions. From the experimental results, it was observed that for dye concentrations as much as 10–200 mg L-1, quantitative removal (more than 92%) is obtained in a single adsorption. The adsorption equilibrium data obeyed the Freundlich model in the concentration ranges studied. Simplicity of adsorbent preparation, effective and fast adsorbent were the main advantages in this method.

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