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Adsorption of Bezathren dyes onto sodic bentonite from aqueous solutions

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ABSTRACT

The aim of the present work was to investigate the adsorption of synthetic textile dyes, such as Bezathren-Blue, Bezathren-Green and Bezathren-Red onto sodium bentonite (Bt-Na⁺). Adsorption experiments were performed under batch process, to assess the performance of Bt-Na⁺ for the removal of Bezathren-dyes, using initial dyes concentrations, pH of solution, contact time and temperature as variables. According to results, the uptake of Bezathren-dyes by Bt-Na⁺ was rapid and the maximum sorption was observed at lowest pH. The maximum uptake capacities (q_m) for Bezathren-Blue, Bezathren-Green and Bezathren-Red were 35.08 mg/g, 32.88 and 48.52 mg/g respectively. Different types of adsorption isotherms and kinetic models were used to describe the Bezathren-dyes adsorption behavior. The experimental results fitted Freundlich model and the pseudo-second order kinetic models well. The results suggested that Bt-Na⁺ is suitable as a sorbent material for recovery and adsorption of Bezathren dyes from aqueous solutions.

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1. Introduction

Synthetic dyes are extensively used in different processing industries such as textile leather, printing, cosmetic, drug and food [1]. Based on their structure, synthetic dyes can be classified into 20–30 groups. But, the largest class of synthetic dyes in the color index is acid dyes, and this class of dyes is anionic compounds and mostly is azo, anthraquinone or triarylmethane group. Anthraquinone based dyes account for about 15% of colorants and have structures based on quinines. These dyes are very resistant to degradation and its color will fade away for a long time [2].

Dyes removal from industrial wastewater is an environmental issue, due to their high organic loads, low biodegradability, toxicity, mutagenicity and carcinogenicity; even in low concentrations, color imparts an anesthetic appearance to water [3]. The selection of adequate methods for effective removal of dyes from wastewaters depends on many factors: the dye chemical structure, specific behavior at different pH values, concentration in wastewater etc. Various physical, chemical and biological processes are used for dyes removal: coagulation-flocculation, precipitation, biodegradation, adsorption, ion exchange, membrane processes, chemical oxidation and solvent extraction [4,5]. However, all these conventional methods are not comparable to adsorption technique in term of efficiency, operating cost, process flexibility and ease of

operation. Further all these techniques were found to be inefficient and incompetent because of the fairly high solubility and stability of the dyes toward light, oxidizing agents and aerobic digestion. A comprehensive survey indicates that adsorption technique was the most appropriate and efficient one [6].

A large number of studies have been dedicated to finding suitable and cheap adsorbents for the treatment or removal of dyes from water and wastewater. The conventional adsorbent is the activated carbon but many other low cost adsorbents such as sepiolite, kaolinite, montmorillonite, smectite, bentonite, zeolite and alunite have been investigated for this purpose [7–13].

Some of clay minerals possess a high adsorption capacity toward several classes of dyes and their adsorption capabilities are comparable to those activated carbons. From the recent studies of the adsorption of dyes using clay minerals it can be seen that some of natural clay minerals (mostly is montmorillonite/bentonite) show significant dye removal capacities; while others still need modification in order to enhance its adsorption capacities [14].

The adsorption performance of clay minerals and its modified forms also depend strongly on class of dye. Many of natural clay minerals have a high adsorption capacity for binding basic (cationic) dyes but often hardly to remove dyes from other groups or classes of dyes [15].

In this regard, montmorillonite-rich materials like bentonites exhibit highly interesting properties, e.g. high specific surface area, cation-exchange capacity (CEC), porosity, and tendency to retain water or other polar and non-polar compounds [16]. The

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Bentonite is the clay mineral used in this study. It is clay mainly composed of montmorillonite which is a 2:1 type aluminosilicate. Bentonite has the capacity to exchange these cations with the ones present in aqueous solutions of organic or inorganic salts. This property is mainly responsible for the great adsorbent power of bentonite, especially toward ions in solution. The bentonite was chosen for this work because of its natural abundance and low cost, compared with other clay types, bentonite has excellent adsorption properties and possesses adsorption sites available within its interlayer space as well as on the outer surface and edges [17]. Therefore, bentonite has recently been employed in many separation applications with or without modification [18].

The aim of the present work is to investigate the possibility of Bt-Na⁺ as an adsorbent for removal of Bezathren dyes (Anthraquinone based dyes), which is, namely Red, Blue and Green, from aqueous solution by adsorption. Effects of pH and temperature on the adsorption process are also investigated. The adsorption capacity of Bezathren dyes with Bt-Na⁺ was carried out using two kinetic models, which are the pseudo-second order and pseudo-first order. Finally, the experimental data were compared using two isotherm equations, which are Langmuir and Freundlich.

2. Experimental

2.1. Bentonite sample

The natural bentonite used in this study was obtained from deposits in the area of Maghnia (west of Algeria); it was supplied by ENOF Ltd. Company (Algeria).

The chemical composition was found to be as follows: 62.48% SiO₂, 17.53% Al₂O₃, 1.23% Fe₂O₃, 3.59% MgO, 0.82% K₂O, 0.87% CaO, 0.22% TiO₂, 0.39% Na₂O, 0.04% As, 13.0% loss on ignition at 950 °C. The mineralogical analysis showed that the native crude clay mineral contains preponderantly Montmorillonite (86 wt.%); the clay composition also includes Quartz (10%), Cristoballite (3.0%) and Beidellite (less than 1%) [16,19].

The bentonite was purified according to the method published in a previous study [14] and it was put in sodic form as follow: an amount of bentonite was dispersed in NaCl solution (1 M) with a 1/5 mass ratio and after agitation for 2 h, the solid was separated by centrifugation (rotational speed equal to 6000 rpm for 15 min), this operation was repeated three times. The solid was washed three times with distilled water and it was dried at 40 °C for three days.

The chemical composition of purified bentonite (Bt-Na⁺) was found to be as follows: 64.7% SiO₂, 18.1% Al₂O₃, 0.95% Fe₂O₃, 2.66% MgO, 0.8% K₂O, 0.61% CaO, 0.2% TiO₂, 1.43% Na₂O, 0.05% As, 10.0% loss on ignition [20,21].

The cation-exchange capacity (CEC) of bentonites was determined according to the ammonium acetate saturation method and was found to be 70 meq per 100 g of dry natural-Bt and 98 meq per 100 g of dry Bt-Na⁺. The BET specific surface area increase from 50 m²/g in natural-Bt to 95 m²/g in Bt-Na⁺ [16,22].

2.2. Dyes solutions

Bezathren dyes used in this study are classified as an anthraquinone dyes (Schema 1). They were provided from SOITEX Company (Tlemcen-Algeria).

- Bezathren-Blue: Dinaphthol [2,3-*a*:2',3'-*h*] phenazine-5,9,14,18-(6*H*,15*H*)-tetraone (C₂₈H₁₄N₂O₄); MW: 442.10 g/mol, Log P: 2.68, C Log P: 7.60.
- Bezathren-Red: 1, 4-diamino-2-methoxyanthracene-9,10-dione (C₁₅H₁₂N₂O₃); MW: 268.27 g/mol, Log P: 0.69, C Log P: 2.92.
- Bezathren-Red: 1, 4-bis (*p*-tolylamino) anthracene-9,10-dione (C₂₈H₂₂N₂O₂); MW: 418.49 g/mol, Log P: 6.32, C Log P: 9.32.

Dyes solutions at 1000 ppm were prepared by dissolving of dyes powder (0.1 g) in 100 mL of distilled water. On a Analytik Jena Specord 210 Plus UV-vis spectrophotometer, these compounds absorbed at wavelength (λ_{max}) 600.0 nm for Bezathren-Blue, 630.0 nm for Bezathren-Green and 510.4 nm for Bezathren-Red.

2.3. Adsorption and procedure

The method of adsorption used for this study, was carried out by a mixture of 10 mL of Blue and Green dyes solutions of known concentration (C₀ = 50 ppm), and 0.05 g of our Bt-Na⁺ and 15 mL of Red dye solution with 0.03 of Bt-Na⁺ in Erlenmeyer with stopper, under vigorous stirring (700 rpm) at room temperature (20 ± 2 °C). Both liquid and solid phases were separated by centrifugation; the liquid phase was measured by the UV-visible spectrometer.

The amount of adsorbed dyes at various equilibrium times (*q_t*, mg/g) was calculated using following relationship:

$$q_t (\text{mg/g}) = \frac{C_i - C_t}{W} \times V \quad (1)$$

The percentage removal of dyes was calculated using following equation:

$$\text{Adsorption (\%)} = \frac{C_i - C_e}{C_i} \times 100 \quad (2)$$

where C_i, C_t and C_e (mg/L) are the initial, time *t* and equilibrium dyes concentrations, respectively; V and W are the liquid volume (L) and the weight of dried used adsorbent (g).

2.6. Characterization techniques

The chemical composition of the bentonites was determined by X-ray fluorescence spectroscopy (Philips PW 3710) and XRD patterns of bentonites were collected on a Philips X-Pert diffractometer. Thermogravimetric analyses of bentonite (TGA) was performed using a Perkin Elmer TGA-7 thermogravimetric analyzer at a heating rate of 15 °C/min from 40 to 800 °C under nitrogen atmosphere (at 20 ml/min) in order to evaluate the thermal stability of our bentonite. IR spectrum was obtained with a Perkin Elmer 16PC spectrometer Model Fourier transform infrared spectrometer.

3. Results and discussion

3.1. Characteristic results of Bt-Na⁺

3.1.1. FTIR analysis of the Bt-Na⁺

The IR spectrum of Bt-Na⁺ (Fig. 1) reveals the presence of characteristic absorption bands of clay such as bands corresponds to Si-O, Si-O-M, and M-O-H (M=Al, Fe or Mg) existing between anions and cations located in octahedral and tetrahedral sheets, and OH groups. For example, bands between (3620–3640 cm⁻¹) can be associated to stretching vibrations of O-H groups coordinated to Al and Mg atoms (3640 cm⁻¹) or two Al atoms (3620 cm⁻¹) in octahedral sheets of bentonite; For the band centered at 1027 cm⁻¹, it characterizes the Si-O stretching vibrations; the stretching vibrations bands of Si-O-M^{VI} (M= Al, Mg, and Fe) located in octahedral sheets appears at 400–550 cm⁻¹ range [19,20]. For M^{VI}-OH bands (M^{VI}=Al, Mg, and Fe): Al^{VI}-OH vibrations occur at 920 cm⁻¹, sharing of the OH group between Fe and Al in octahedral sheets can move this peak until about 815–915 cm⁻¹, in the case of our sample of Bt-Na⁺, the peak appears at 913 cm⁻¹.

3.1.2. TG analysis of the Bt-Na⁺

For the dried Bt-Na⁺ curve (Fig. 2), 9.5% mass-loss was recorded at the temperature range of 30–200 °C and 4.75% mass-loss appeared at 350–800 °C. The first is due to desorption of water molecules, which were adsorbed onto the cations in the bentonite

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