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A novel approach for removing an industrial dye 4GL by an Algerian Bentonite

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ABSTRACT

Adsorption processes of 4GL on two different bentonite-based sorbents were compared: i) Sodium-bentonite in the presence of a cationic surfactant CTAMB; ii) organobentonite alone. This latter was prepared by exchanging the inorganic cation of bentonite with a quaternary ammonium cation at 100% of the clay's CEC. Batch adsorption studies were conducted to evaluate the effect of various parameters such as the quaternary ammonium cation (CTAMB) loading, contact time and initial 4GL concentration.

From the kinetic study, it is interesting to note that Organobentonite exhibits faster kinetics compared to Na-bentonite/CTAMB system. The comparison between isotherm plots makes clear that the presence of CTAMB in solution changes the 4GL isotherm. Compared to organobentonite, the adsorption capacity is higher when CTAMB was present in solution. In this case, the results of 4GL adsorption by bentonite obey to Langmuir model. While for organobentonite, all models seem to be applicable. The difference between the two methods is confirmed by XRD analysis.

The performance shown by Organobentonite and Na-bentonite/CTAMB system was suitable compared to other adsorbents, reflecting a promising future utilization in wastewater treatment.

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1. Introduction

The textile industry effluents contain an important amount of colour, dissolved surfactant and a large amount of suspended solids and hence are an important pollution problem in developing countries. Among these dyes and pigments, many are toxic and have carcinogenic and mutagenic effects that affect aquatic biota and humans [28,44]. Actually, coloured effluents have to be adequately treated before they are discharged into the environment.

Acid dyes are water-soluble and anionic. They are widely used to dye wool, nylon, and silk fibers. Research efforts have been deployed to improve bentonite adsorption property of acid dye. Recently, bentonite modification has been focused on: acid activation [16,20], Cr(III)-intercalating [8], aluminium pillaring [9], surfactant modification [3,18,36,

37], Na⁺ exchanging [3], and layered double hydroxide [38]. All these modification techniques are effective for acid dye removal, especially the surfactant-modified bentonite. In the last 20 years, many reports [44,47] showed that the surfactant-modified clays displayed higher adsorption capacity than the original clay. Although the modification of clays with surfactants increases their cost significantly, the resultant increase in adsorption capacity is higher. However, clay derivatives may be promising sorbents for environmental and purification purposes.

The adsorption capacities of acid dyes on acid and certain metal-modified bentonite is approximately 0.1–0.3 mmol/g [20]. The cationic surfactant-modified bentonite can adsorb 0.78–6.03 mmol of dye per gram [36], which is >20 times the adsorption capacity of other adsorbents. Results show that surfactant-modified bentonite is efficient for acid dye removal.

In wastewaters coming up from pigments, dyestuffs or textile manufactures, cationic surfactants coexist always with dyes [31,40]. Cationic surfactant such as quaternary ammonium compounds can significantly reduce the activity of cells, which may inhibit the biodegradation of other pollutants [41].

In order to reduce the cost and simplify the process of wastewater treatment, a new process including both the synthesis of Organobentonite and its adsorption removal of organic pollutants from wastewater was proposed in this research work. In this process, the adsorbate and cationic surfactant were dispersed in wastewater,

Abbreviations: Ci, the initial concentration of dye, mg/L; Ce, the concentration of dye in solution after equilibrium, mg/L; V, the total volume of the solution, L; M, the mass of the adsorbent, g; q_e, the amounts of dye adsorbed at equilibrium time, mg/g; q_t, the amounts of dye adsorbed at time t, mg/g; K₁, the pseudo-first order rate constant, min⁻¹; K₂, the pseudo-second order rate constant, g/mg min; q_m, the amount of solute adsorbed per weight of adsorbent in forming a complete monolayer on the surface, mg/g; b, Langmuir constant related to the energy, L/mg; k and n, Freundlich constants.

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and then the sodium-exchanged bentonite was added. Thus, the organic pollutants were removed from water accompanied with the synthesis of Organobentonite [19,23]. Especially, the presence of cationic surfactant in the wastewaters of pigments, dyestuffs, rubber products or agriculture chemical manufactures, may achieve the process by the addition of bentonite and simultaneously remove both the surfactants and organic pollutants. It is also hoped that the biodegradability of the wastewater would be improved by the removal of the non-biodegradable organic pollutants and antimicrobial cationic surfactants after that the wastewater being treated by the one-step process.

In this paper the adsorption properties of Organobentonite in aqueous phase and bentonite in the presence of both organic and surfactant contaminations were studied. From these aspects, our work is concentrated on the adsorption of anionic dye Supranol Yellow 4GL on surfactant–clay complexes using two methods: One method consists to add the surfactant to the solution containing the pollutant and the complex CTAMB-4GL is then adsorbed onto bentonite (Na-bentonite/CTAMB system). Another method consists to adsorb the pollutant (4GL) onto bentonite exchanged with CTAMB (organobentonite).

2. Materials and methods

2.1. Preparation of Organobentonite (adsorbent)

The starting clay used in this study is supplied by ENOF (an Algerian manufacture specialized in the production of non-ferric products and useful substances) at the rough state. This clay has undergone a purification protocol composed of several stages: disintegration to disperse the layers of clay and recovery of the fraction lower than 2 μm . The recovered suspension was treated with an acid solution (0.05 N) to eliminate carbonates, and then saturated with sodium chloride (0.1 N) three times. Thus, the recovered clay known as sodium bentonite was washed several times and was dried at ambient temperature.

The surfactant used to modify the bentonite is a cationic surfactant, Cetyltrimethyl-ammonium bromide (CTAMB 99%), of chemical formula $\text{C}_{19}\text{H}_{42}\text{NBr}$ provided by ACROS ORGANICS.

The organic clay (organobentonite) was prepared by exchanging the inorganic cation of bentonite with a quaternary ammonium cation at 100% of the clay's CEC according to the procedure described earlier [2]: the quantity of organic cation added to the bentonite was determined by:

$$f = \frac{M_{\text{cation}}}{\text{CEC} \times M_{\text{clay}} \times \text{GMW}_{\text{cation}} \times X} \quad (1)$$

where f : fraction of cation exchange capacity satisfied by organic cation (dimensionless), M_{cation} : mass of organic cation required to achieve 100% of CEC (mass), CEC: cation exchange capacity of clay (equivalents/mass), M_{clay} : mass of clay (mass), $\text{GMW}_{\text{cation}}$: gram molecular weight of organic cation (mass/mol), and X is the moles of charge per equivalent (mole/equivalent).

Theoretically, the cation exchange capacity represents the maximum amount of organic cation that can be exchanged onto the clay's surface. The synthesis of organoclay was performed by the following procedure: 1 g of Na-bentonite was first dispersed in about 100 mL of deionised water, to which a desired amount of cetyltrimethylammonium bromide was slowly added. The concentration of CTAMB was 100% CEC of bentonite. The mixtures were stirred for 24 h. All products were washed until free bromide anions (tested by AgNO_3), dried at 60 °C and finally ground in an agate mortar.

2.2. Adsorbate

The selected dye is an industrial colouring substance used in the synthesis for dyeing the chemical textiles. It is synthesized and marketed

under the commercial code: Yellow Supranol 4GL [22]; it is used without further purification. Its maximum wavelength of adsorption at 410 nm is determined by a UV–Visible spectrophotometer SAFAS. For every investigated system, the complete UV–visible spectra were measured in the presence of the CTAMB, to examine the effect on the spectrum shapes and thus to avoid misinterpretation of the spectrophotometric determinations.

2.3. Methods

Powder X-ray diffraction (PXRD) patterns were obtained with a Siemens D501 X-ray diffractometer using $\text{Cu K}\alpha$ radiation ($\lambda = 1.5415 \text{ \AA}$) and fitted with a graphite back-end monochromator. The samples were scanned from 2° to 70° (2θ) using steps of 0.08° and a counting time of 4 s per step. The attenuated total reflectance infrared spectra (ATR-FTIR) were collected on an FTIR Nicolet 5700 (Thermo Electron Corporation) spectrometer equipped with a Smart Orbit accessory.

2.4. Kinetic and equilibrium experiments

The adsorption experiments were determined by using the batch equilibration technique, and were conducted in laboratory by contacting a specific volume of Supranol yellow 4GL (adsorbate) with a quantity of bentonite or Organobentonite as an adsorbent.

The effect of cetyltrimethylammonium bromide ions on 4GL adsorption was assessed through the determination of CTAMB isotherm in the presence of 4GL at 100 mg/L concentration.

In order to evaluate kinetics data, 2 g of adsorbent was added to 1 L of known initial concentration: 100 mg/L of 4GL (Organobentonite alone or Na-bentonite/CTAMB system) and the mixture was allowed to agitate at the stirring speed of 300 rpm. In the preliminary adsorption test, negligible effect of the stirring speed was observed. At given time intervals, about 2.5 mL of the solution was sampled using a 10 mL syringe, then centrifuged and the equilibrium concentrations of the adsorbate were calculated, after measuring their absorbance values at wavelength of 410 nm by UV–vis spectroscopy (SAFAS bundle of type MC²).

Isotherm experiments were carried out at 25 ± 1 °C using a water bath shaker. 100 mL of solutions of varying concentration of 4GL from 200 to 1400 mg/L were placed in vials. An equal amount 0.2 g of Organobentonite was introduced into each vial, and stirred for 60 min. The collected samples were then centrifuged and analyzed as mentioned above.

The adsorption isotherms in the presence of CTAMB (Na-bentonite/CTAMB system) were carried out by shaking 0.2 g of sodium-exchanged bentonite with 100 mL of solution containing a variable initial concentration (200–1400 mg/L) of 4GL mixed with 5 mmol/L of CTAMB. After 60 min stirring, each sample was centrifuged. The supernatant was analyzed at 410 nm.

Controls containing 4GL solution and no adsorbent were measured for each adsorption experiment. The controls were used to obtain a calibration curve for each measurement.

For the adsorption experiment, the mass of the 4GL lost from solution during equilibration, was assumed to be adsorbed by the adsorbent:

$$q_e = \frac{(C_i - C_e) \times V}{M} \quad (2)$$

where q_e is the mass of solute adsorbed by the adsorbent (mg/g), C_i is the initial concentration of solute (mg/L), C_e is the solution phase solute concentration at the end of the adsorption experiment (mg/L), V is the volume of solution (L), and M is the mass of the adsorbent (g).

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