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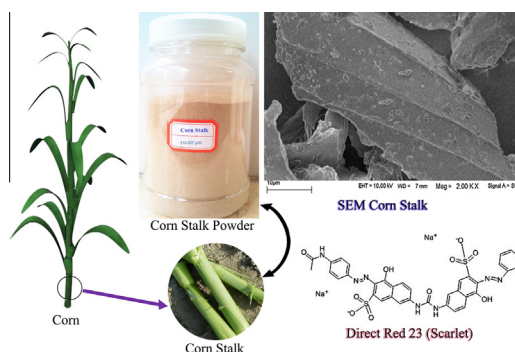
Removal of Direct Red 23 from aqueous solution using corn stalks: Isotherms, kinetics and thermodynamic studies

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HIGHLIGHTS

- Magnetic corn stalk was used as an adsorbent for the removal of Direct Red 23.
- The equilibrium data fitted well with the Freundlich model.
- Adsorption of Direct Red 23 onto corn stalk was favored at low pH.
- The pseudo-second-order kinetic model fits better with experimental data.

GRAPHICAL ABSTRACT



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ABSTRACT

The objective of this study was to assess the suitability and efficiency of corn stalk (CS) for the removal of diazo dye Direct Red 23 (DR23) from aqueous solutions. The effect of different variables in the batch method as a function of solution pH, contact time, initial dye concentration, CS amount, temperature, and so forth by the optimization method has been investigated. The color reduction was monitored by spectrophotometry at 503 nm before and after DR23 adsorption on the CS, and the removal percentage was calculated using the difference in absorbance. The sorption processes followed the pseudo second order in addition to intraparticle diffusion kinetics models with a good correlation coefficient with the overall entire adsorption of DR23 on adsorbent. The experimental equilibrium data were tested by four widely used isotherm models namely, Langmuir, Freundlich, Tempkin and Dubinin–Radushkevich (D–R). It was found that adsorption of DR23 on CS well with the Freundlich isotherm model, implying monolayer coverage of dye molecules onto the surface of the adsorbent. More than 99% removal efficiency was obtained within 10 min at adsorbent dose of 0.2 g for initial dye concentration of 10–90 mg L⁻¹ at pH 3. Various thermodynamic parameters, such as Gibbs free energy, entropy, and enthalpy, of the ongoing adsorption process have been calculated. Judgment based on the obtained results of thermodynamic values shows the spontaneous and endothermic nature adsorption processes on adsorbent.

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Introduction

Textile industries produce huge amounts of polluted effluents that are normally discharged to surface water bodies and ground-water aquifers. These wastewaters cause many damages to the ecological system of the receiving surface water [1,2] and create a

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lot of disturbance to the groundwater resources. consequently, an enormous amount of wastewater, along with solid residues, is produced. These wastewaters have an intense coloration, because approximately 20% of the total production is lost to the environment during the fiber coloring step [3]. The discharge of these colored pollutants to ecosystems is a dramatic source of esthetic pollution and has an adverse effect on aquatic life [4]. Several dyes are used in textile dyeing and they are comprised of two components: a chromophore group, responsible for the color and a functional group that binds to the textile fiber. Dyes are completely or partially soluble in various substrates such as textile materials, paper, leather, etc. Around 60% of the dyes used in textile plants are azo dyes, characterized by azo groups ($-\text{N}=\text{N}-$) bound to the sp^2 hybrid carbon atoms, and they are classified as mono, di, tri, and tetra azo dyes, with the azo groups mainly bound to the benzene and naphthalene rings [5]. Coloration is the first parameter taken into account, at least from the esthetic viewpoint, in effluent treatment. The need to develop processes to remove color which ensure low contaminant levels has led to several researches and publications focused on textile effluents [6]. Considerable efforts have been made by many researchers to find appropriate treatment systems in order to remove pollutants and impurities of wastewaters emanated from different industries, in particular, textile industry [1–7]. Many chemical and physical methods of dye-removal, including photocatalytic degradation [7], membranes [8] and adsorption techniques [9,10] have been used from time to time. Adsorption process is noted to be superior to other removal techniques because it is economically cost effective, simple [2] and it is capable to efficiently treat dyes in more concentrated form [9].

Direct Red 23 has diazo group, it has toxicity and carcinogenic nature. Despite this, very few studies have been conducted on the adsorption of Direct Red 23 from water. Abdelwahab et al. have studied the adsorption capacity of the untreated and activated rice husk on adsorption of Direct Red 23 from aqueous solution [11]. Ardejani et al. evaluated the removal of two dyes, Direct Red 23 and Direct Red 80, from textile wastewaters using orange peel as a low-cost adsorbent [12]. Holliman et al. have investigated the adsorption of Direct Red 23 on different samples of titania [13]. Tan et al. used *Rhizophora apiculata* bark as adsorbent for removal of Direct Red 23 [14]. Hebeish et al. have prepared cationized sawdust for removal of Direct Red 23 from aqueous solutions [15]. Achmad et al. have synthesized a novel adsorbent from *Uncaria Gambir* for removal of Direct Red 23 from aqueous solution [16]. Additionally, only a few studies have been done on the aqueous photocatalytic degradation of Direct Red 23. Zhu et al. have investigated photocatalytic degradation of Direct Red 23 and Acid Red 3B by supported $\text{TiO}_2 + \text{UV}$ in aqueous solution [17]. Song et al. have prepared $\text{SrTiO}_3/\text{CeO}_2$ composite for photocatalytic degradation of Direct Red 23 in aqueous solutions under UV irradiation [18]. Conducted also research on removal of Direct Red 23 by a variety of microorganisms [19] and ozonation combined with sonolysis [20].

Therefore, in this work, magnetic corn stalk was used for removal of anionic dye, Direct Red 23 (DR23), from aqueous solution. The effects of pH, temperature and initial dye concentration on dye adsorption by the corn stalk were investigated. The experimental data were analyzed using the Lagergren pseudo-first-order kinetic model, the pseudo-second-order model and the intraparticle diffusion model. Thermodynamics parameters, ΔG° , ΔH° and ΔS° , were also calculated.

Experimental

Materials and instrument

Corn Stalk (CS) were firstly separated, before their use, all the sorbents were washed thoroughly with deionized water and oven

dried at 25 °C for 24 h. Crushed in a ball mill and sieved the samples with stainless steel mesh screens to obtain different particle sizes of adsorbent.

All reagents used are of analytical grade chemicals and were obtained from Merck. Stock solutions of the test reagent were made by dissolving the dyes in doubly distilled water. The structure of the dyes, Direct Red 23, anionic direct dye; formula: $\text{C}_{35}\text{H}_{25}\text{N}_7\text{Na}_2\text{O}_{10}\text{S}_2$; Molecular weight = 813.72 gr mol^{-1} ; maximum wavelength = 503 nm) is shown in Table 1. The concentration of the dye was determined at 503 nm using an UV-Vis spectrophotometer (model 160A, Switzerland). A pH meter (model 827 lab, Hyderabad, India) was used to determine the pH of the solution. The morphology and size distribution of the CS was observed by scanning electron microscopy (SEM; Hitachi S-4160) under an acceleration voltage of 200 kV. The degree of crystallinity of the corn stalk was shown from X-ray diffractograms (X Philips X'Pert X-ray diffractometer) with $\text{Cu K}\alpha$ radiation for 2θ values over 10–80°. also analyzed with FTIR-8 400 S Shimadzu spectrophotometer in order to identify the functional groups in the fiber structure.

Batch mode adsorption studies

Batch mode adsorption studies were carried out by shaking 50 ml conical flasks containing 0.2 g of CS and 25 mL of DR23 solutions of desired concentration with adjusted pH on an orbital shaker machine at 200 rpm at 318.15 K. The solution pH was adjusted with 0.1 mol L^{-1} HCl and 0.1 mol L^{-1} NaOH solutions. At the end of the adsorption period, the supernatant solution was separated by centrifugation at 200 rpm for 10 min. Then the concentration of the residual dye was determined spectrophotometrically by monitoring the absorbance at 503 nm for DR23 using UV-Vis spectrophotometer. Percentage of dye removal was calculated using the following equation:

$$\% \text{DR23 Removal} = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

where C_0 (mg L^{-1}) and C_t (mg L^{-1}) are dye concentration initially and at time t , respectively. For adsorption isotherms, dye solutions different concentrations (10–100 mg L^{-1}) were agitated with known amounts of adsorbents until the equilibrium was achieved. Equilibrium adsorption capacity was calculated from the following equation:

$$q_e = \frac{(C_0 - C_t)V}{W} \quad (2)$$

where q_e (mg g^{-1}) is the equilibrium adsorption capacity, C_t (mg L^{-1}) is the dye concentration at equilibrium, V (L) is the volume of solution and W (g) is the weight of adsorbent.

The procedures of kinetic experiments were identical with those of equilibrium tests. At predetermined moments, aqueous samples (5 mL) were taken from the solution, the liquid was separated from the adsorbent magnetically and concentration of DR23 in solution was determined spectrophotometrically at a wavelength of 503 nm. The amount of DR23 adsorbed at time t (q_t (mg g^{-1})) was calculated by following equation:

$$q_t = \frac{(C_0 - C_t)V}{m} \quad (3)$$

where C_0 (mg L^{-1}) is the initial dye concentration, C_t (mg L^{-1}) the dye concentration at any time t , V (L) the volume of the solution and m (g) is the mass of the adsorbent.

Results and discussion

Characterization of adsorbent

The SEM images of the CS surface show the homogeneous and relatively smooth surface of the CS (see Fig. 1). Fig. 1 shows the

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