



Sorption equilibrium, kinetic, thermodynamic, and desorption studies of Reactive Red 120 on *Chara contraria*

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ABSTRACT

Potential use of *Chara contraria* for sorption of Reactive Red (RR) 120 was investigated. The sorption process was highly dependent on various operating variables like particle size, adsorbent dose, pH, temperature, ionic strength, dye concentration, and contact time. Before and after the sorption of RR 120, adsorbent surfaces were characterized by FTIR-ATR spectroscopy. Equilibrium data was well predicted by Freundlich model. Kinetic sorption of RR 120 on *C. contraria* was well described by sigmoidal Logistic model. Activation parameters derived from Arrhenius and Eyring equations indicated that interaction was mainly physical sorption. Thermodynamic parameters showed that sorption process onto macro-alga was a spontaneous and endothermic nature. Low value of desorption suggested that both strong and weak binding forces could be present between dye and adsorbent. Overall, *C. contraria*, potentially low-cost adsorbent can be used for the removal of RR 120.

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1. Introduction

Reactive azo dyes are extensively used in dyeing processes in textile industry because of their simple dyeing procedures and good stability during washing process [1,2]. About 20–40% of these dyes remain in the effluents. Charges of such wastewater into receiving waters can be hazardous on exposed organisms due to their reactivity, toxicity, and stability [3]. Azo dyes constitute the largest group of mutagenic and carcinogenic xenobiotic pollutants. Disposal of these dyes and remediation of associated contaminated sites remains a worldwide concern [3,4]. Therefore, prior to discharge of wastewaters to aquatic ecosystems, removal of recalcitrant pollutants has become a necessity for living Earth.

Reactive dyes which contain one or more azo bond ($-N=N-$), exhibit a wide range of different chemical structures, primarily based on substituted aromatic and heterocyclic groups [4,5]. Use of reactive dyes is now the most important method for dyeing of cotton and cellulose fibers [6]. They have high resistance to chemical, photochemical, and biological degradation and highly persistent in natural environment. Due to the complex structure and stability of the dyes, conventional biological treatment methods are ineffective [2,4,7,8]. Moreover, their removal from wastewater by conventional coagulation, membrane filtration, and activated

sludge processes is difficult, since reactive dyes are highly soluble in water [1]. These processes have considerable energy requirements and thus impose high economic and environmental costs [2,9,10]. Therefore, alternative simple methods to work alongside these are sought. Sorption is known to be a promising technique, which has great importance due to ease of operation and comparable low cost of application in treating of textile dyeing effluents [8,10].

Sorption of reactive dyes has been studied in detail on various adsorbents such as bacteria, fungi, algae, aquatic plant, chitosan etc. [2,4,11]. In order to investigate sorption efficiency for removing of Reactive Red (RR) 120, different adsorbents such as *Lentinus sajor-caju* [12], *Spirogyra majuscula* [7], pistachio husk [6], *Hydrilla verticillata* [4], chitosan [11], palm-oil industry waste [13], clay [14] were used. For this purpose, potential use of *Chara contraria* for removing of RR 120 has not been found in literature. However, this adsorbent had been used to remove Lanaset Red (LR) G [15,16] and to decolorize malachite green [17].

C. contraria was chosen in the present study, since it is widespread underwater meadows and so relatively cheap adsorbent for sorption process. Besides, it was reported that this alga had considerable potential for the removal of dyes from aqueous solution [15,17]. Therefore, aim of this study was (i) to investigate the potential use of dried biomass of *C. contraria* for sorption of Reactive Red (RR) 120 from aqueous solution; (ii) to evaluate effects of particle size, adsorbent dose, pH value, temperature, ionic strength, initial dye concentration, and contact time on the sorption in the batch system; (iii) to investigate sorption kinetic models, equilibrium models, thermodynamic parameters, and

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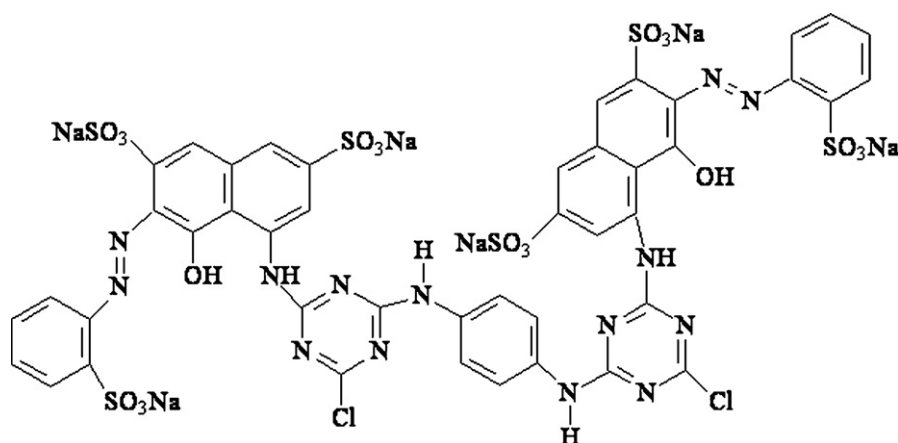


Fig. 1. Chemical structure of Reactive Red 120.

(iv) to clarify desorption of this dye. Moreover, Logistic model was fitted to experimental data to get more information about the sorption of RR 120 such as sorption rate (μ) and maximum sorption value (A).

2. Experimental

2.1. Adsorbent

C. contraria used in the study, was obtained from a freshwater pool in Durukaynak village of Adiyaman, Turkey. Collected sample was washed twice with tap water, dried at 80 °C for 24 h, and sieved into three particle sizes (125–250, 250–500, and >500 μm mesh sizes). It was stored in air tight polyethylene bottle.

Infrared spectra of the adsorbent before and after RR 120 sorption were taken using a Fourier transform infrared equipped with an attenuated total reflection spectrometer (Perkin-Elmer Spectrum 100 FTIR-ATR Spectrometer).

2.2. Adsorbate

Azo reactive dye, Reactive Red (RR) 120 (Procion Red HE-3B; MF: $\text{C}_{44}\text{H}_{24}\text{Cl}_2\text{N}_{14}\text{O}_{20}\text{S}_6\text{Na}_6$; MW: 1469.98; λ_{max} : 515 nm) was obtained from Sigma (Sigma-Aldrich Chemical Co., St. Louis, USA). The chemical structure is shown in Fig. 1. Stock dye solution (1 g L^{-1}) was prepared by dissolving accurately weighed quantity of RR 120 in distilled water. Dye solution with desired concentrations was prepared by diluting the stock dye solution with suitable volume of distilled water.

2.3. Sorption studies

Sorption of RR 120 on *C. contraria* was investigated as functions of particle size (>500 to >125 μm), adsorbent dose (0.5–4.0 g L^{-1}), initial pH value (pH 1–9), temperature (298–313 K), ionic strength (0.001–1.0 M NaCl), initial dye concentration (25–200 mg L^{-1}), and contact time (0–180 min) under the aspects of sorption isotherms, sorption kinetics, activation energy, thermodynamic, and desorption studies.

The pH of solutions was adjusted to desired value with 0.1 M HCl and/or 1.0 M NaOH solutions. Experiments were performed with 100 mL of sorption solution (with desired concentration and pH) and desired adsorbent in 250 mL conical flask. These flasks were stirred on the orbital shaker at 150 rpm for 180 min, which was more than ample time for equilibrium.

At predetermined time (0, 5, 10, 15, 30, 45, 60, 90, 120, 150, and 180 min), withdrawn samples from the flasks were centrifuged to

precipitate suspended biomass at 6000 rpm for 10 min. The residual RR 120 concentration in the supernatant liquid was analyzed using UV/VIS spectrophotometer (Jenway 6305) at λ_{max} 515 nm. Sorption experiments were performed as duplicate and mean values were used in sorption analyses. The amount of dye adsorbed per adsorbent mass was calculated by the use of the following equations.

$$q_t = \frac{(C_o - C_t) \times V}{M} \quad (1)$$

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

where q_t and q_e show the amount of RR 120 adsorbed on the macro alga at time t and at equilibrium (mg g^{-1}), respectively. C_o , C_t , and C_e represent the dye concentrations (mg L^{-1}) at initial, at t time, and at equilibrium in the solution, respectively. V is the volume of solution (L), and M is the mass of adsorbent (g).

For sorption studies, various theoretical equations are applied to experimental data in order to find a model which adequately predict kinetic or equilibrium data. The fitting procedure was performed by use of commercial computer software SigmaPlot version 11 (Systat Software, Inc., CA, USA) via the Marquardt-Levenberg algorithm. The validity of models was evaluated using the coefficient of determination (R^2) and the sum of squares errors (SSE).

2.4. Thermodynamic study

Thermodynamic experiments were performed by agitating dye solutions of different concentrations (25–200 mg L^{-1}) with 1 g L^{-1} *C. contraria* at different temperatures (298, 303, and 308 K).

2.5. Desorption study

For desorption studies, RR 120 dye solution (1 g L^{-1}) were mixed with *C. contraria* at pH 1 and 25 °C for 180 min. Remaining dye concentration in the solution was measured in order to calculate adsorbed dye on the adsorbent. After then, adsorbent was filtered and dried in the vacuum oven at 80 °C for 24 h. RR 120 loaded adsorbent was allowed to contact with 50 mL of distilled water at different pH value (pH 8, 9, 10, and 11) in 100 mL conical flasks at 25 °C, agitated on the orbital shaker at 150 rpm for 180 min. The amount of dye desorbed was determined by using spectrophotometer (Jenway 6305) at 515 nm. The percentage of dye desorbed from the adsorbent was calculated:

$$\text{Desorption (\%)} = \frac{\text{mass of desorbed}}{\text{mass of adsorbed}} \times 100 \quad (3)$$

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