

Regular Article

Synthesis and application of a new carboxylated cellulose derivative. Part III: Removal of auramine-O and safranin-T from mono- and bi-component spiked aqueous solutions



Filipe Simões Teodoro^a, Megg Madonyk Cota Elias^a, Gabriel Max Dias Ferreira^a, Oscar Fernando Herrera Adarme^b, Ranylson Marcello Leal Savedra^c, Melissa Fabíola Siqueira^c, Luis Henrique Mendes da Silva^d, Laurent Frédéric Gil^{a,*}, Leandro Vinícius Alves Gurgel^{a,*}

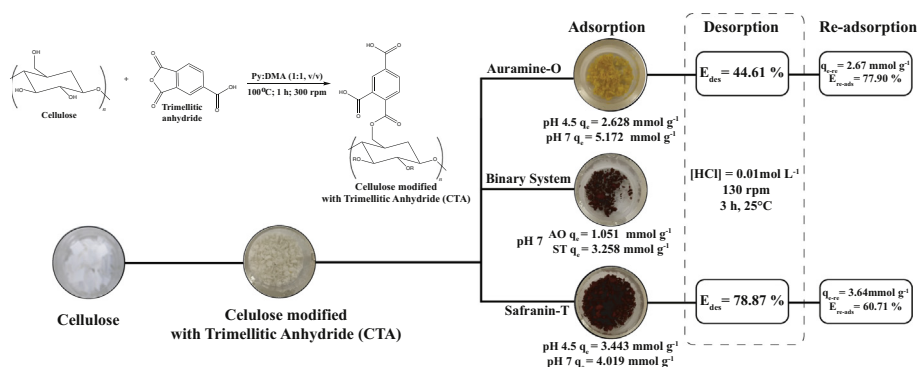
^a Grupo de Físico-Química Orgânica, Departamento de Química, Instituto de Ciências Exatas e Biológicas, Universidade Federal de Ouro Preto, Campus Universitário Morro do Cruzeiro, s/n°, Bauxita, 35400-000 Ouro Preto, Minas Gerais, Brazil

^b Laboratório de Química Tecnológica e Ambiental, Departamento de Química, Instituto de Ciências Exatas e Biológicas, Universidade Federal de Ouro Preto, Campus Universitário Morro do Cruzeiro, s/n°, Bauxita, 35450-000 Ouro Preto, Minas Gerais, Brazil

^c Laboratório de Polímeros e Propriedades Eletrônicas de Materiais, Departamento de Física, Universidade Federal de Ouro Preto, Campus Universitário Morro do Cruzeiro, s/n°, Bauxita, 35400-000 Ouro Preto, Minas Gerais, Brazil

^d Grupo de Química Verde Coloidal e Macromolecular, Departamento de Química, Centro de Ciências Exatas e Tecnológicas, Universidade Federal de Viçosa, Av. P. H. Rolfs, s/n°, 36570-000 Viçosa, Minas Gerais, Brazil

GRAPHICAL ABSTRACT



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ABSTRACT

In the third part of this series of studies, the adsorption of the basic textile dyes auramine-O (AO) and safranin-T (ST) on a carboxylated cellulose derivative (CTA) were evaluated in mono- and bi-component spiked aqueous solutions. Adsorption studies were developed as a function of solution pH, contact time, and initial dye concentration. Adsorption kinetic data were modeled by monocomponent kinetic models of pseudo-first- (PFO), pseudo-second-order (PSO), intraparticle diffusion, and Boyd, while the competitive kinetic model of Corsel was used to model bicomponent kinetic data. Monocomponent adsorption equilibrium data were modeled by the Langmuir, Sips, Fowler-Guggenheim, Hill de-Boer, and Konda models, while the IAST and RAST models were used to model bicomponent equilibrium data. Monocomponent maximum adsorption capacities for AO and ST at pH 4.5 were 2.841 and 3.691 mmol g^{-1} , and at pH 7.0 were 5.443 and 4.074 mmol g^{-1} , respectively. Bicomponent maximum adsorption capacities for AO and ST at pH 7.0 were 1.230 and 3.728 mmol g^{-1} . Adsorption enthalpy changes

* Corresponding author.

E-mail addresses: legurgel@iceb.ufop.br, legurgel@yahoo.com.br (L.V.A. Gurgel).

1. Introduction

Contamination of ground and surface water by synthetic dyes due to the discharge of untreated or partially treated industrial wastewater is a serious environmental problem and is considered to be a threat to humans and aquatic organisms [1,2]. Synthetic dyes have various industrial uses including in cosmetics, food processing, leather tanning, rubber, paper, plastics, printing and dye manufacturing, and textiles [1,3].

The discharge of synthetic dyes into the hydrosphere is problematic primarily due to dyes' potential to color water and reduce sunlight penetration into water, as well as dyes' resistance to photochemical and biological degradation [1,4]. In addition, synthetic dyes and their degradation products include mutagenic compounds, carcinogenic compounds, and compounds that may increase the chemical oxygen demand (COD) and biochemical oxygen demand (BOD) of water resources [1,3]. The complex aromatic chemical structures of synthetic dyes make them very stable to heat, oxidizing agents, photochemical degradation, and biodegradation, which makes their removal from industrial wastewaters difficult [1,3].

Auramine-O and safranin-T are classified as basic dyes [5], which can be used to dye wool, silk, acrylic fibers, and leather in textile industries [6]. Auramine-O can cause acute oral toxicity, carcinogenicity, cytotoxicity, DNA damage, genotoxicity, and mutagenicity [7]. Safranin-T can cause carcinogenicity, mitochondrial toxicity, mutagenicity, and nucleic acid damage [7]. According to Terinte et al. [8], most conventional dyeing takes place in developing countries, where environmental legislation is often insufficient and/or not implemented properly. A survey describing the percentages of unfixed dyes that may be discharged in the effluent for different dyes, fabrics, and applications was recently reported by Terinte et al. [8].

Three categories of treatment technologies have been applied to effluent containing synthetic dyes: physical, chemical, and biological [3,9]. Physical treatments include adsorption by activated carbon, membrane filtration and separation, ion-exchange, irradiation, and electrokinetic coagulation. Chemical treatments include oxidative processes, photochemical destruction, and electrochemical destruction. Biological treatments include aerobic and anaerobic treatment, among others [1,3,9]. Despite the development and application of technologies for treatment of wastewaters containing synthetic dyes, cost effective, efficient, and rapid water treatment at the commercial level is still a challenging problem for the scientific community [1]. Although adsorption can treat large flow rates, producing high quality effluents [1], if the adsorbent material cannot be cleaned by desorption and reused, the applicability and usefulness of this treatment method is limited. Thus, research to develop suitable adsorbents with high adsorption capacity, good economic viability, versatility for removal of various organic and inorganic pollutants, and better regeneration capacity is still needed.

In the first and second parts of this series of studies [10,11], the synthesis and application of a new adsorbent material (CTA) using cellulose as a solid support was described in detail, as well as the adsorption of Co^{2+} , Cu^{2+} , and Ni^{2+} from mono- and multi-component aqueous solutions. The high adsorption capacity and selectivity exhibited by the CTA adsorbent for removal of metal

ions from mono- and multi-component aqueous solutions, in addition to CTA's high capacity for basic dye adsorption compared to previously studied adsorbents (Supplementary Table 1), motivated our research group to begin adsorption studies for the removal of dyes from aqueous solutions. As pointed out by Gusmão et al. [12], the use of the same adsorbent for various environmental applications is desirable because it makes its production economically feasible.

In the third part of this series of studies, the removal of the basic dyes auramine-O (AO) and safranin-T (ST) using the CTA adsorbent was assessed as a function of solution pH, contact time (kinetics), and initial dye concentration in spiked mono- and bi-component aqueous solutions. Dye removal was also studied at two pH values (i.e. 4.5 and 7.0). A suitable desorption process was developed to evaluate the regeneration and reusability of the CTA adsorbent. The enthalpy of the adsorption process was also evaluated and assessed by isothermal titration calorimetry, and the removal mechanism of both dyes was evaluated based on thermodynamic data and molecular quantum mechanics calculations.

2. Material and methods

2.1. Material

Auramine-O (MW = 303.83 g mol $^{-1}$; $\text{C}_{17}\text{H}_{21}\text{N}_3\cdot\text{HCl}$; C.I. 41000; $\lambda_{\text{max}} = 434$ nm; purity $\leq 100\%$) and safranin-T (MW = 350.84 g mol $^{-1}$; $\text{C}_{20}\text{H}_{19}\text{N}_4\text{Cl}$; C.I. 50240; $\lambda_{\text{max}} = 530$ nm; purity $\leq 100\%$) were purchased from Vetec (Brazil). Anhydrous citric acid ($\text{C}_6\text{H}_8\text{O}_7$; 99%) and trisodium citrate ($\text{Na}_3\text{C}_6\text{H}_5\text{O}_7\cdot 2\text{H}_2\text{O}$; 99%) were purchased from Synth (Brazil). Dye solutions used in the adsorption studies were prepared in buffer solutions consisting of 0.05 mol L $^{-1}$ citric acid/sodium monocation (pH from 2.13 to 3.13), 0.05 mol L $^{-1}$ sodium monocation/sodium dication (pH from 4.13 to 5.15), and 0.05 mol L $^{-1}$ sodium dication/sodium trication (pH from 6.13 to 7.13). Other materials and chemicals used in this investigation are described in the first part of this series of studies [10].

2.2. Adsorption experiments

Monocomponent adsorption studies of AO and ST on the CTA adsorbent were performed at two pH values (i.e. 4.5 and 7.0), while bicomponent adsorption studies of AO and ST on the CTA adsorbent were only carried out at pH 7.0, which was the pH of highest adsorption capacity for both dyes. All adsorption studies were performed in duplicate.

2.2.1. Adsorption of dyes onto CTA as a function of solution pH

For a typical monocomponent experiment, CTA samples (20.0 mg) weighed in cylindrical glasses (1.8 mm height \times 2.2 mm diameter) were added to 250 mL Erlenmeyer flasks containing 100.0 mL of buffered dye solution of a known concentration (0.932 mmol L $^{-1}$) at pH values varying from 2.13 to 7.14. Flasks were stirred at 130 rpm and 25 °C in a shaker incubator (Marconi, model MA-830) for 24 h. Then, the suspensions were centrifuged (Excelsa® II centrifuge, model 206 BL) at 3600 rpm for 10 min to separate the solid and liquid phases. The absorbance of the supernatant phase was measured on a UV-Vis spectrophotometer (Biospectro, model SP-220) at 434 nm for AO and 530 nm for ST.

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