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Removal of tartrazine from aqueous solutions using adsorbents based on activated carbon and *Moringa oleifera* seeds

Isabela Maria Reck [a], Rebecca Manesco Paixão [b], Rosangela Bergamasco [b], Marcelo Fernandes Vieira [b], Angélica Marquetotti Salcedo Vieira [c, *]

[a] Postgraduate Program in Food Science, State University of Maringa – UEM, Av. Colombo, 5790, Maringa, Parana, CEP: 87020-900, Brazil
[b] Department of Chemical Engineering, State University of Maringa – UEM, Av. Colombo, 5790, Maringá, Paraná, CEP: 87020-900, Brazil
[c] Department of Food Engineering, State University of Maringa – UEM, Av. Colombo, 5790, Maringa, Parana, CEP: 87020-900, Brazil

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A B S T R A C T

In this study, a new and low-cost adsorbent, obtained from the *Moringa oleifera* seed, was compared to activated carbons of babassu coconut (ACBC) and bone (ABC), to remove dye from aqueous solution. Brunauer-Emmet-Teller analysis, scanning electron microscopy and X-ray diffraction were used to characterize the adsorbents. Adsorption with *Moringa oleifera* was able to remove more than 95% of the dye, reaching a maximum capacity of 91.27 mg g^{-1} at 15 °C. The results demonstrate that the kinetic data were well described by the models of pseudo-second order for the activated carbon of babassu coconut and pseudo-first order for both *Moringa oleifera* seeds and activated bone carbon. Isothermal data using the Langmuir and Freundlich equations were adjusted, with a better fit to the experimental data given by the Freundlich equation. The thermodynamic parameters show that adsorption is an endothermic process for both carbon materials and an exothermic process for the *Moringa oleifera* seeds. An actual wastewater assay was performed demonstrating the effectiveness of the Moringa adsorbent. This results show that *Moringa oleifera* seeds can be an effective alternative to the use of activated carbon for dye removal in contaminated waters, since it is a natural adsorbent and does not undergo the carbon activation processes.

1. Introduction

Many industrial processes generate colored effluents. Food industry is a large consumer of water and releases significant amounts of dyes with their effluents, as well as textiles. There are over 100000 different kinds of commercially available synthetic colors, and considerable percentages are being discharged daily in aqueous effluents (Almeida et al., 2009). Synthetic dyes are a significant pollutant class because they cause imbalance in aquatic ecosystems due to their complex molecular structures consisting of aromatic rings and azo functional groups (N = N), which are stable and difficult to remove (Bonetto et al., 2015).

Synthetic dyes absorb and reflect sunlight, interfering with the growth of aquatic species by inhibiting photosynthesis (Pereira and Alves, 2012). Moreover, these dyes may have harmful effects on organisms depending on the concentration and exposure time. Therefore, there is an urgent need to remove synthetic dyes from effluents before they are discharged into the water system.

Tartrazine, whose IUPAC (International Union of Pure and Applied Chemistry) name is trisodium (4E)-5-oxo-1-(4-sulfonatophenyl)-4-[(4-sulfonatophenyl) hydrazono]-3-pyrazolecarboxylate, is a synthetic water-soluble anionic dye. This substance can cause major allergic reactions, particularly among asthmatics and people with aspirin intolerance (Goscianska and Pietrzak, 2015). Thus, wastewater containing tartrazine, in various concentrations, must be treated before discharge.

Given the adverse effects of synthetic dyes in aquatic ecosystems, the search for appropriate alternatives to effluent treatment technologies is very important. Adsorption with activated carbon has proven to be very effective in the treatment of industrial waste (Gautam et al., 2015b; Peláez-Cid et al., 2016). In addition, conventional coagulation/flocculation processes are not the most suitable for dye removal from wastewater. The most commonly used coagulants such as aluminum and ferric salt, still have some

* Corresponding author.
E-mail addresses: isabelareck@hotmail.com (I.M. Reck), rebeccapaixao@gmail.com (R.M. Paixão), rosangela.bergamasco@gmail.com (R. Bergamasco), marcelofvieira@hotmail.com (M.F. Vieira), amsvieira@uem.br (A.M.S. Vieira).

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disadvantages. They usually produce large quantities of dangerous sludge with residual metal. Aluminium based coagulants present potential adverse effects in the public health (Wei et al., 2017). Moreover aluminum is not biodegradable and can cause problems of disposal and treatment of the large amount of sludge generated (Bongiovani et al., 2014). Therefore, the adsorption is one of the most promising techniques for the treatment and removal of pollutants and dangerous chemicals, especially with the activated carbon.

The final properties of the obtained product depend not only on the characteristics of the used material but also on the activating agent used and the conditions of the activation process (De Filippis et al., 2013). However, the use of commercially available activated carbon is limited because of the high manufacturing costs.

Therefore, recent research has been focused on searching for economical and effective adsorbents for adsorption (Habila et al., 2014). Methods for obtaining activated carbon from waste, such as pyrolysis (De Filippis et al., 2013) and co-pyrolysis (Habila et al., 2014) are used by produces useful gas and oil fraction (tar) with a high energetic value and a solid residue (char) that could be used as the cheap carbon-based material. Besides the study of alternative methods to reduce the cost of production of the coals, the search for organic wastes to be used as a precursor of activated carbon (Gautam et al., 2015a) and adsorbents (Pavan et al., 2014; Daneshvar et al., 2017), has also intensified in the sense of reducing environmental liabilities.

Biosorption, using carbons from plant and animal sources, is advantageous for water treatment because it is a process that uses low-cost materials to sequester pollutants from aqueous solutions, that is, it uses by-products of the agricultural, food and pharmaceutical industries, making it an economically viable process (Bhatnagar and Minocha, 2010).

The agricultural sector is constantly growing, particularly as trade relations strengthen between countries and demographics continue to put pressure on food production systems. Twelve percent of the total land area of the world is used for crop production in the agricultural sector, producing many different types of wastes and by-products (Grace et al., 2016). According to Maneerung et al. (2016) several studies have been effective in using activated carbon obtained from renewable sources, such as walnut shells, orange, rice and bamboo fibers, with the aim of using waste materials.

This means that the biosorption can be considered an alternative and low-cost treatment method. Activated bone carbon was studied along with others adsorbents by Ip et al. (2010) in Reactive Black 5 dye adsorption. The tartrazine removal from water was studied with carbons produced from other sources. Gautam et al., 2015a evaluated the ability of activated carbon obtained from Alligator weed (*Alternanthera philoxeroides*), while Habila et al. (2014) focused their studies on the evaluation of the simultaneous conversion of different types of wastes (palm, paper, and plastic wastes) into activated carbon via copyrolysis.

According to literature, many biomaterials can be applied according to their adsorption capacities, however, the activated carbon from bone and babassu coconut, are yet poorly exploited for dye removal from wastewater.

Carbon that goes through activation processes has an increased surface area, which facilitates the bioremediation of many contaminants. However, seeds capable of acting as biosorbents, as in the case of *Moringa oleifera* (moringa) seeds can also have a high potential for pollutant removal.

In this context, the seeds of *Moringa oleifera* have been studied for the removal of turbidity and other impurities in water. The use of moringa has two additional advantages in water treatment; it can be used as the primary source of activated carbon and has coagulants/flocculants in seeds (Obuseng et al., 2012). The use of moringa as a water treatment agent is perhaps one of the most interesting uses. According to Baptista et al. (2015), the seeds have an active compound that acts in colloidal systems to neutralize charges and form bridges between particles. This process is responsible for flake formation and sedimentation.

The aim of this study was to evaluate the removal of the yellow dye tartrazine from aqueous solutions using low-cost adsorbents, moringa seeds, compared to activate bone carbon (ABC) and the activated carbon of babassu coconut (ACBC), which according to literature (De Filippis et al., 2013; Ip et al., 2010; Jibril et al., 2013), present high capacity in treatment of water.

2. Material and methods

The dye used for the adsorption study was tartrazine yellow, the powder in its pure form, donated by the Duas Rodas Industrial Company of Jaraguá do Sul, SC. The seeds of *Moringa oleifera* were obtained in the city of Aracaju, SE, Brazil. The Tobasa - Tocantins Babaçu S/A company, Cinelandia/RJ region, provided the activated bone carbon (ABC) and the activated carbon of babassu coconut (ACBC).

2.1. Adsorbent preparation

Moringa seeds were peeled manually and then dried in an oven with air circulation (Digital Timer SX CR/42) at 105 °C (Akhtar et al., 2007) for 2 h. After drying, they were ground in a common processor for about 2 min to be standardized into a particle size of 28 mesh. This parameter was chosen after preliminary experiments with different particle sizes to verify this influence on tartrazine removal (data did not reported). However, the analysis reported no significant difference of this parameter in the dye adsorption process, so the mesh particle size 28 was chosen for removal of this pollutant due to its higher yield. The obtained powder was used as an adsorbent for adsorption assays. The ACBC was donated by Tobasa Bioindustrial Babaçu S.A company and ABC was donated Bonechar Carbon Brazil Ltda.

2.2. Adsorbent characterization

The methodology for determining the point of zero charge (pH_{PZC}) was adapted from Regalbuto and Robles (2004) and it is known as the “experiment of eleven points”. A plot of the difference between the final pH and the initial pH (pHf-pHi) against initial pH (pHi) was plotted, where pHf-pHi = 0 was taken as the pH_{PZC}. For microstructural characterization of carbons and moringa seeds, X-ray diffraction (XRD) analysis was performed on a D8-Advance diffractometer (Bruker). Samples were exposed to X-rays with a 2θ angle ranging between 5° and 80°, in 0.2° steps, with a CuKa 1.5406 Å tube lamp. The applied voltage and current were 40 kV and 30 mA, respectively. The evaluation of morphological characteristics of the moringa seeds and carbons were carried with a scanning electron microscopy (SEM) using a SS 550 Superscan electron microscope (Shimadzu). The characterization of the adsorbents was completed by obtaining their specific surface area and pore volume using the BET method (Brunauer et al., 1938), with ultrapure nitrogen adsorption at 77 K, using a Quantachrome NOVA 1000 series analyzer.

2.3. Adsorption kinetics

The adsorption kinetics were studied using a series of parameters previously analyzed, as such 50 mL of 15 mg L^{-1} of tartrazine