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Removal of reactive yellow 145 by adsorption onto treated watermelon seeds: Kinetic and isotherm studies

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ABSTRACT

The aim of this study is to valorize the watermelon seeds treated with hexane for the removal of reactive yellow 145 from aqueous solutions. The effects of physico-chemical parameters such as pH, adsorbent dose, temperature and dye concentration on the removal of dyes were evaluated. The kinetic study of the adsorption of reactive yellow 145 on watermelon seeds adsorbent perfectly followed the pseudo-second-order kinetic model. The Langmuir, Freundlich, Temkin, Elovich and Dubinin-Radushkevich models were studied. It was proved that the experimental data fitted well to the Freundlich model, which signifies that the phenomenon of adsorption is done in multilayers on heterogeneous surfaces. The finding of this paper asserts that this material is a good and low-cost adsorbent that can be used for the removal of dyes from aqueous solutions.

1. Introduction

Nowadays, The wastewater has become an increasingly big concern to the scientific community all over the world (Borah et al., 2015). The synthetic dyes are usually used in several industries such as printing, food, textile, pulp mills, cosmetics, etc., poses a serious problem for the environment and living creatures. The removal of these effluents toxic is a environmental requirement to save this earth.

Several methods were used for the elimination of these toxic dyes, for instance, cloud point extraction, ultra-filtration, sorption, oxidation, coagulation-flocculation, etc… The adsorption is an efficient and economical method of treating aqueous effluent. Some of the benefits of the adsorption method are possible for the regeneration at the economical price, availability of known process equipment, sludge-free operation and recovery of the sorbate (Kapdan and Kargi, 2002). The adsorption process needs a sufficient quantity of adsorbents. Recently, the researchers think of new biomaterials from the agricultural waste which will be more economical, easy to regenerate and have a high capacity for adsorption (Tamez Uddin et al., 2017). In literature, a large category of adsorbents prepared from agricultural wastes have been used for removal pollutant from wastewater, including, Agricultural waste: coconut shell (Pino et al., 2006), Watermelon shell (Banerjee et al., 2012), rice straw (Ding et al., 2012), banana peel (Moubarak et al., 2014), apple peels (Enniya et al., 2018), potato peel (Öktem et al., 2012), Sugar wastes (Anastopoulos et al., 2017), olive wastes (Anastopoulos et al., 2015). Watermelon is one of the major underutilized fruits grown in the warmer part of the world, melon fruit contains large quantities of seeds. These seeds must be used like adsorbents in some studies (Salman et al., 2015) to eliminate the lead II (Samra et al., 2014) or phenol (Malunjkar and Ambekar, 2015) or methylene blue (Salman et al., 2015) in aqueous solutions, but the preparation of these biosorbent is differed from a study to other.

In the current work, the objective is to valorize the powder of watermelon seeds to remove reactive yellow 145 dye from an aqueous solution. The characterization of this adsorbent was made by FTIR, DRX, SEM, specific surface and elementary analysis. The Adsorption on batch mode was evaluated by the variation of temperature, adsorbent dosage, pH and initial concentration of dye. The equilibrium adsorption has been analyzed by the Langmuir, Freundlich, Temkin, Elovich and Dubinin-Radushkevich isotherms models and the kinetic data was tested on the pseudo-first and pseudo-second-order models.

2. Material and methods

2.1. Preparation of adsorbent

Watermelon seeds were collected, washed several times in running tap water followed by distilled water and dried at room temperature for 24 h. Dried samples were crushed to a powder by electric mortar. Then an extraction of fat for the watermelon seeds was produced by the soxhlet method with hexane. After that, the material was placed in an oven for 8 h at 80 °C, crushed and passed through sieves and particles

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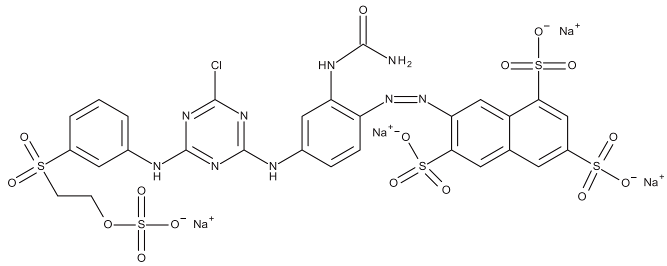

Fig. 1. Chemical structure of RY145 dye.

were collected under 200 µm. The sample obtained after all these steps was subsequently stored and named watermelon seeds treated (WST).

2.2. Preparation of adsorbate

The reactive yellow 145 (RY145) dye used in this study is an organic compound of chemical formula $C_{28}H_{20}ClN_9S_5.4Na$ (Fig. 1). Reactive yellow 145 is an anionic dye had a wavelength of 419 nm with the molecular weight equal to1026.20 g/mol (Aguedach et al., 2005; Kazi et al., 2016). The reactive yellow 145 dye used to prepare the stock solution by dissolving a specific amount of the dye in distilled water and then we made series of dilution to obtain the desired concentrations.

2.3. Adsorption mechanism

The adsorption process was determined by batch mode of a known amount of the WST adsorbent by adding 100 mg of adsorbent into 250 mL conical flasks which contains specific volume 100 mL of dye solution of RY145. Adsorption experiments were made to take into consideration the effects of experimental parameters of initial dye concentrations (10, 20, 30, 40, 50 and 60 mg/L), the effect of adsorbent (50, 100, 150, 200, 250 and 300 mg), the effect of temperature (20, 30, 40 and 50 °C) and the pH (3, 5, 7, 9 and 11). The solutions pH was adjusted by adding HCl (0.1 N) or NaOH (0.1 N) solution and controlled by the pH-Metre (OHAUS STARTER 3100). Each experiment was replicated three times and the average reading was taken.

The conical flasks were carried out in an orbital shaker at 500 rpm for 3 h. The samples were centrifuged (Centrifuge 5415 R) at 12×10^3 rpm for 15 min and the remaining dye concentrations in the supernatant were recorded at the maximum wavelength (419 nm) by UV–VIS Spectrophotometer (JENWAY 6715).

The elemental analysis of WST adsorbent was obtained with a Panalytical's WD-XRF spectrometer, FT-IR spectra were achieved by using an IRTF Vertex 70, XRD of WST adsorbent was acquired out by a Panalytical'sX'Pert PRO MRD by using a copper anode tube λ(Cu) = 1.54 Å, the micrographs of samples were taken by using scanning electron microscopy (SEM) (FEI Quanta 200) and the surface areas were measured by the BET method (Brunauer - Emmett - Teller) using a Pore Size Micrometric (9320 model, USA).

The amount of dye adsorbed onto the WST at time t, q_t (mg/g), was determined according to equation Eq. (1):

$$q_t = \frac{(C_0 - C_t)}{W} \times V \qquad (1)$$

C_0 and C_t (mg/L) are the concentrations of RY145 at initial and time t (min), respectively. V (L) is the solution's volume, and W (g) is the weight of WST adsorbent.

The percentage of adsorption was obtained by the following equation Eq. (2):

$$\%\text{of Adsorption} = \frac{(C_0 - C_e)}{C_0} \times 100 \qquad (2)$$

C_e (mg/L) is the equilibrium concentration of RY145 (Slimani et al., 2017).

2.4. Adsorption kinetics

It is necessary to study the adsorption kinetics to obtain the information about the adsorption mechanism, which is important for efficiency of the process. The adsorption kinetics was evaluated by using both the pseudo-first and pseudo-second-order kinetics to determine the mechanism of RY145 onto WST.

The pseudo-first-order model is given as (Lagergren, 1898):

$$q_t = q_e(1 - e^{K_1 t}) \qquad (3)$$

The pseudo-second-order model is given as (Ho and Mc Kay, 2000):

$$q_t = \frac{K_2 . q_e^2 . t}{1 + K_2 . q_e . t} \qquad (4)$$

q_e and q_t (mg/g) are the amounts of the RY145 adsorbed at equilibrium and at time t(min), respectively, k_1(min^{-1}) is the adsorption rate constant of the pseudo-first-order and k_2 (g/mg.min)is the rate constant of the second-order-model.

2.5. Adsorption isotherm

Adsorption isotherm represents the mathematical models of the relationship between the adsorbed quantity and the adsorbate quantity remaining in a test medium at a defined temperature under equilibrium condition.

Several models of adsorption isotherm available for analyzing experimental parameters and for describing the equilibrium of adsorption. In the present work, Langmuir (Langmuir, 1918), Freundlich (Freundlich, 1906), Temkin (Temkin and Pyzhev, 1940), Elovich (Elovich and Larinov, 1962) and Dubinin–Radushkevich (Dubinin and Radushkevich, 1947) isotherm were used to investigate the adsorption behavior. Table 1 presents the non linear forms of these isotherms models.

2.6. Error analysis

Due to the inherent bias resulting from linearization, five different error functions of non-linear regression basin were employed in this study to evaluate the isotherm constants.

1. The sum of the squares of the errors (SSE):
   This most commonly used error function has one major drawback. The function will result in the calculated isotherm parameters providing a better fit at the higher end of the liquid phase concentration range. This is because the magnitude of the errors and hence the square of the errors will increase as concentration increases.

$$SSE = \sum_{i=1}^{n} (q_{e,calc} - q_{e,\exp})_i^2 \qquad (5)$$

2. The sum of the absolute errors (SAE):

Table 1
Isotherm models tested in this study.

Isotherm	Non linear form
Langmuir	$q_e = \frac{q_m KLCe}{1 + KLCe}$
Freundlich	$q_e = K_F (C_e)^{1/n}$
Temkin	$\frac{qe}{qm} = \frac{RT}{\Delta Q}\ln(KTCe)$
Elovich	$\frac{qe}{qm} = KECeexp(-\frac{qe}{qm})$
Dubinin-Radushkevich	$q_e = q_m exp(-\beta\, \varepsilon^2)$ (with $\varepsilon = R\,Tln(1+\frac{1}{qe})$