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USING ACTIVATED AND FUNCTIONALIZED OF *CONOCARPUS* L. LEAVES CARBON FOR REMOVAL OF DISPERSE YELLOW DYE FROM ITS AQUEOUS SOLUTIONS

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ABSTRACT : The textile industry is the key user of dyes and hence the major source of water pollution, which harm aquatic as well as human life. Dead leaves of *Conocarpus* L. were collected in order to preparethe activated carbon. The first part of the collected leaves was functionalized with nitric acid and the second one was treated with EDTA-Na$_2$. Experiments of adsorption were conducted at lab as a function of pH, contact time and initial dye concentration. The results showed that the maximum removal efficiency of the disperse yellow with a percentage of 94.11% and adsorption per unit mass with a value of 5.64 mg/l by activated carbon functionalized with the nitric acid was found in the acidic medium. The activated carbon that prepared from *Conocarpus* L. functionalized proved to be a new promised efficient adsorbent for the removal of the disperse yellow from its solutions.

Key words : Aqueous solutions, disperse yellow dye, carbon, *Conocarpus* L.

INTRODUCTION

The textile industry is the key user of dyes and hence the major source of water contamination, which affect aquatic as well as human life. Removing synthetic dyes from waste-water has become a challenge preventing their environmental hazard in particular for water bio-systems because it has been estimated that the total dyes consumption in thetextile industry worldwide is more than 10,000 tons per year and about 10–15% of these dyes are released as effluents during the dyeing processes (Gomez *et al*, 2007; Gupta *et al*, 2013; Salama, 2017). The dyes used in textile industry divided to anionic (acidic, direct, reactive), cationic (all basic dyes) and nonionic (disperse dyes) groups (Rasoulifard *et al*, 2010). Removing of dyes from the textile waste-water by conventional wastewater treatment methods is very difficult. Different methods for dyes and color removal, such aerobic and anaerobic microbial degradation, coagulation and chemical oxidation, membrane separation process, electro-chemical, filtration, softening and reverse osmosis have been suggested from time to time (Robinson *et al*, 2001). Nowadays; adsorption is the most operational method of dyes removal technique using low cost adsorbents. Adsorption is a well-known equilibrium separation process for water decontamination applications and has been found to be superior to other techniques for water reuse in terms of initial cost, flexibility and simplicity of design and ease of operation (Kannan *et al*, 2001; Ashoka and Inamdar, 2010).

MATERIALS AND METHODS

Preparation of dye solutions (adsorbate)

An accurately weighed 1g of the disperse yellow dye (C20H17CIN2O3) dissolved in deionized water was used to prepare a stock solution (1000 mg/l). The solution that used in the experiment for the required concentration was obtained by successive dilutions. Final concentration of the dye that was determined by using absorbance values was measured before and after adsorption experiments with UV-visible spectrophotometer at 446nm wavelength.

Preparation of thermal activated carbon (adsorbent)

To prepare activated carbon, dead leaves of *Conocarpus* L. tree were collected, washed with deionized water, areal dried, grinded, sieved (0.6 ml) and placed in the oven at 400°C for 120 minutes. The prepared activated carbon was divided into two parts, the first was treated with (0.5N)HNO$_3$ and washed with deionized water to reach pH 6.5. The second was treated with (0.02N) EDTA-Na2.

Suggested modified method for functionalized the thermal activated carbon

A new modified method for functionalized the thermally activated carbon was suggested by current study based on the previous studies (Youssef *et al*, 2013; Anna *et al*, 2015). The surfaces of thermally activated carbon were modified with (0.5N)HNO_3 at a ratio of 50g of activated carbon to 500ml of acid in conical flasks with a glass stopper. Then, the conical flasks were agitated with a water bath shaker for 2h at 90°C and 150 rpm. The resulted material was filtered and extensively washed with a hot deionized water until the cleansing water pH value was approximately 6.5. The carbon was dried in vacuum at 110°C for 4 h. The treated AC was marked as AC A.

A ratio 1g activated carbon:10 ml (0.02 N)EDTA-Na2 was agitated in a 500ml conical flask for 3 h at 150 rpm. Then was filtered and washed with a hot deionized water repeatedly to get rid of excess non reacted EDTA-Na2. Next, the carbon was dried for 4h at 110°C and the material was marked as AC E.

Experiments of adsorption

Serial experiments of adsorption were performed at lab temperature as a function of pH (2, 4, 6, 8 and 10), contact time (5, 15, 30, 60 and 90 minutes) and initial dye concentration (10, 20, 30, 60, 90 and 120 mg/l) using a shaker with constant agitation speed of 225 rpm.

Calibration curve

For developing the calibration curve for the disperse yellow dye, standard dye solutions of 0–90 mg/l concentrations were prepared.

RESULTS AND DISCUSSION

Effect of pH

The pH factor is very important for dye removal in the adsorption process. The pH of the medium will control the electrostatic magnitude charges, which are imparted by ionized dye molecules and causing the variation of the adsorption rate with the pH of the usedmedium (Brahim *et al*, 2014; Etim *et al*, 2015). The effect of pH on removal percentage of disperse yellow dye from solutions and adsorption by using AC A and AC E investigated to be varied in the ranges of pH 2, 4, 6, 8 and 10 at fixed concentration of dye 60 mg/l, adsorbent (AC A and AC E) dosage of 1g and contact time of 60 minutes.

The results of Figs. 2 and 3 showed that the highest removal percentage and adsorption per unit mass of the disperse yellow dye by AC A and AC E were (94.11% and 35.22%) and (5.64 and 2.11) mg/g at pH 2 respectively. It was found that the maximum percentage of removal and adsorption of the dyeon AC A and AC E were at pH2 and no significantly change occurred when the initial pH increased from 2 to 8. Nitric acid showed that it was very effective oxidizing agents due to introduction of a significant number of oxygenated acidic functionalities onto the carbon surface that mainly include carboxylic, lactone and phenolic hydroxyl groups (Gómez-Serrano *et al*, 1993; Yoshinobu, 1993; Figueiredo *et al*, 1999; Lei *et al*, 2002). The carboxyl,hydroxyl and other groups of the carbon were deprotonated to produce negative surface charge, which in turn leads to an electrostatic attraction with H^+ ions intheacidic medium (pH2). Then a strong competition occurred on AC surfaces between dye cations and H^+ ions for occupying the exchangeable sites on AC. In contrast, as the pH increased to 8 there was a reduction in the adsorption efficiency due to the low solubility of the dye in a medium with pH between 2-8. As the solubility of disperse dye in aqueous solution is low, it has a tendency to accumulate at the surface of adsorbents (Ramakrishna and Viraraghavan, 1997). Thus, pH2 was chosen as the best pH value for the further experiments. A similar observation was reported by Narayanan *et al* (2015) for the adsorption of the disperse yellow dye onto *Saccharum spontaneum* surfaces.

The effect of contact time

The contact time effect on removal of disperse yellow dye from its solutions and its adsorption on AC A and AC E were studied for the dye concentration of 60mg/l, of adsorbent (1g) and contact time (5, 15, 30, 60 and 90 minutes) at pH 2.

When AC N and AC W were used, the removal of the dye (75.33% and 13.77%respectively) was fast in the first 5 minutes for AC A and at 60 minutes the percentage of the removed dye reached to (94.11% and 34.00%, respectively) and the amounts of the dye adsorbed per unit mass of AC A and AC E increased from 4.58 to 5.64 and 0.82 to 2.04 mg/g, respectively (Figs. 4 and 5). It is evident from results that the amount of removed dye increased with increasing of contact time because of the adsorption effect to take place, the adsorbent and the adsorbed material must contact for a certain period of time (Kýlýç and Janabi, 2017).

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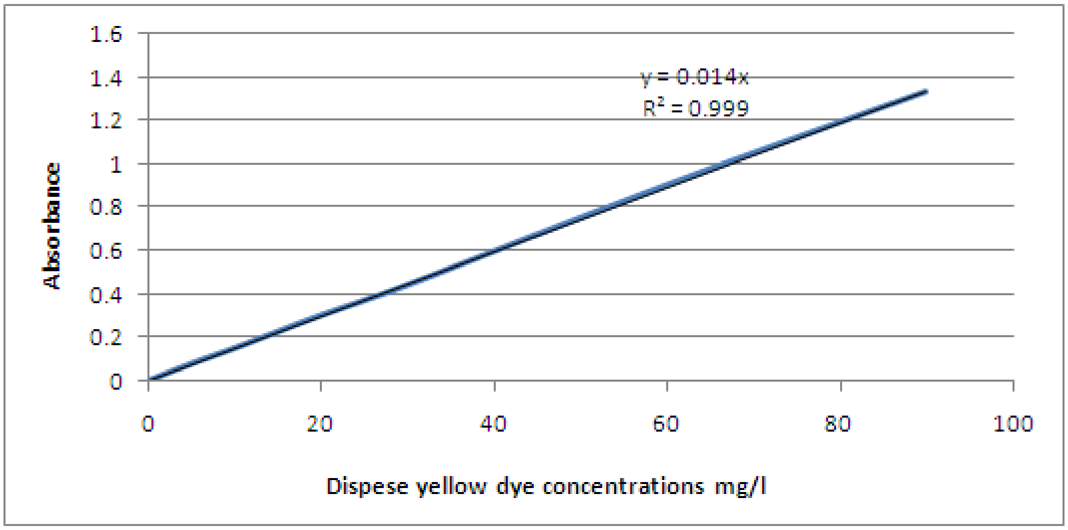

Fig. 1: Calibration curve for disperse yellow dye.

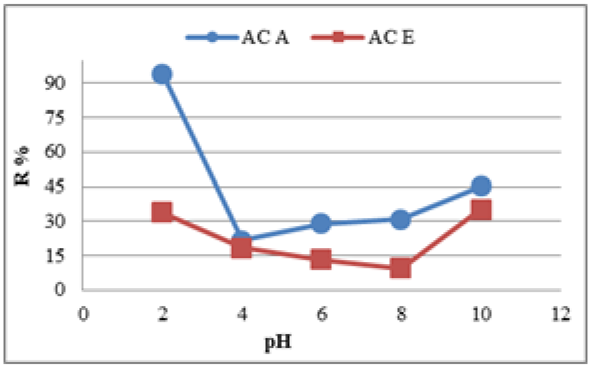

Fig. 2 : The pH effect on dye removal (%).

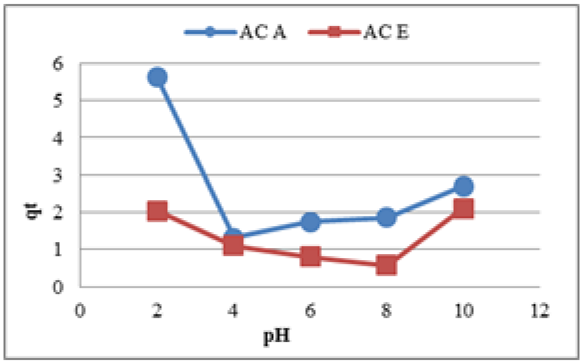

Fig. 3 : The pH effect on dye adsorption (qt mg/g).

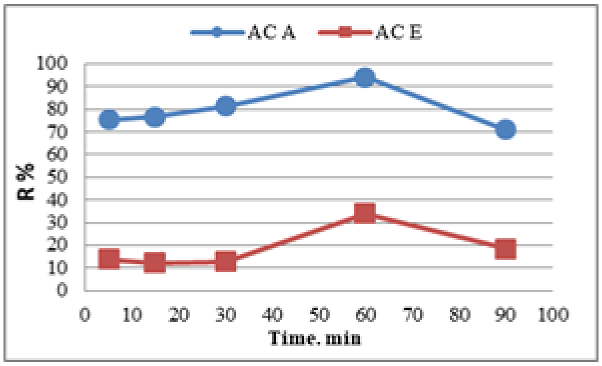

Fig. 4 : The contact time effect on dye removal (%).

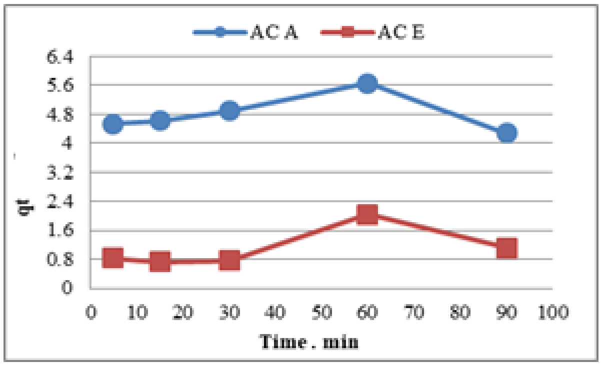

Fig. 5 : The contact time effect on dye adsorption (qt mg/g).

removed dye increased with increasing of contact time, the adsorbent and the adsorbed material must contact for a certain period of time (Kýlýç and Janabi, 2017).

The effect of initial concentration

The initial concentration is important factor to determine the process of dye removal. The concentrations of the disperse yellow dye were varied from 10 to 120 mg/l at a fixed adsorbent (AC A and AC E) dosage of 1g and contact time of 60 minutes with a pH value of 2. For AC A and AC E, the highest removal and concentration of the dye adsorbed per unit mass byAC A and AC were (94.1% and 34.0%) and (5.64 and 2.04) mg/g at 60 mg/l of dye concentrations, respectively (Figs. 6 and 7).

The percent removal of the dye by AC has increased quicklyat initial concentration compared with AC E because it offers important driving force to overcome all mass transfer resistances of the dye between the aqueous and solid phases, which increased the dye removal (Ahmad *et al*, 2008; Mulugeta *et al*, 2014). Then the removal rate gradually decreased from 94.1% to 47.22% as the dye concentration increases from 60 to 120 mg/l. The decline in the dye removal is due to the progressive decrease in

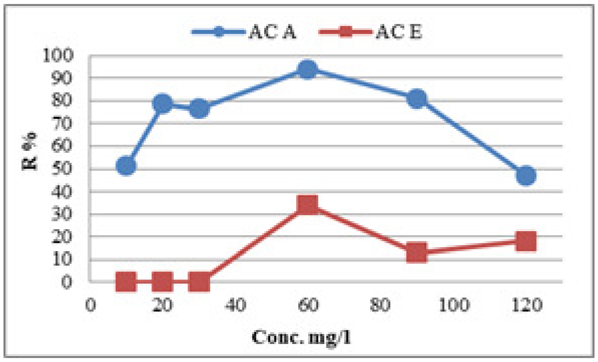

Fig. 6 : The concentration effect on dye removal (%).

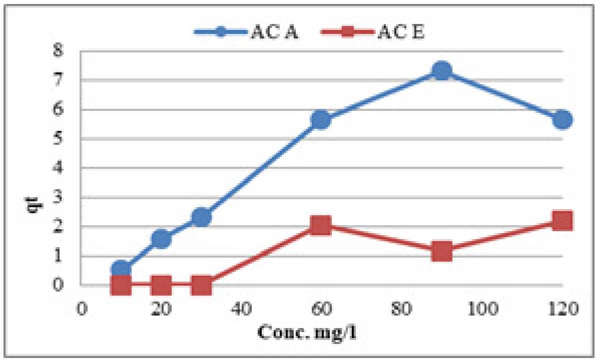

Fig. 7 : The concentration effect on dye adsorption (qt mg/g).

the electrostatic interaction between the dye and the adsorbent active sites (Narayanan *et al*, 2015).

CONCLUSION

The experimental results showed that the efficient removal of disperse dye from its solutions and adsorbed on AC A took place at an acidic medium (pH2). In the view of these results, it can be concluded that the new *Conocarpus* L. functionalized activated carbonthat was treated and functionalized with (0.5N)HNO_3 in acidic medium at pH2 was the better selection as a promised adsorbent for the removal of the disperse yellow from its solutions.

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