Adsorption of cationic dye from aqueous solution onto activated carbon prepared from olive stones

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HIGHLIGHTS

- Thermal carbonization is carried out at 800 °C and 60 min.
- The maximum adsorption of MB dye by OSAC occurred at a pH 5.
- Equilibrium data were best fitted to Temkin isotherm.
- Adsorption kinetics follows the pseudo-second-order kinetic model.

ARTICLE INFO

ABSTRACT

The use of agriculture solid waste as low-cost adsorbents is considered as an ecofriendly adsorbent due to their contribution in the reduction of costs for waste disposal. The present study investigates the adsorption efficiency of raw olive stone (OS) and activated carbon prepared from Egyptian olive stones (OSAC). Batch adsorption experiments were conducted under varying conditions of contact time, initial concentration of methylene blue dye (MB), adsorbent dosage, pH and temperature. The experimental equilibrium data were examined using Langmuir, Freundlich, Temkin, Dubinin–Radushkevich, and Harkins–Jura isotherms. The adsorption kinetic dye was analyzed using pseudo-first order, pseudo-second order and the intraparticle diffusion model. The results showed that the percentage of dye removal increased as the temperature increased but it decreased with the increase in initial dye concentration. The optimum pH required for maximum adsorption was found to be 5. Kinetic studies showed that the adsorption MB onto OS and OSAC followed pseudo-second order kinetic model. The results indicated that olive stone activated carbon could be used as a low-cost adsorbent for the removal of methylene blue from aqueous solution.

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1. Introduction

Discharging of dyes into water resources even in small amounts can affect the aquatic life, limit light diffusion and reduce the process of photosynthesis. Industries such as plastics, rubber, paper, textile, cosmetics and leather are sources for dye effluents. Many of these dyes are carcinogenic, mutagenic, and teratogenic and also toxic to human beings, fish species, and aquatic organisms. Dyes can also cause allergic dermatitis and skin irritation (Uğurlu et al., 2008).

Methylene blue (MB), a cationic dye, is mainly used for dyeing cotton and silk (Tan et al., 2008b). MB can cause eye burns, and if swallowed, it causes irritation to the gastrointestinal tract with symptoms of nausea, vomiting and diarrhea (Tan et al., 2008b), profuse sweating, mental confusion and methemoglobinemia. On inhalation, it can give rise to short periods of rapid or difficult breathing (Tan et al., 2008a). Acute exposure to methylene blue may increase heart rate, Heinz body formation, cyanosis, jaundice, quadriplegia, and tissue necrosis in humans (Vadivelan and Vasanth Kumar, 2005). Hence, the removal of dyes from aquatic wastewater is important for both environmental and public health concerns (Konicki et al., 2013).

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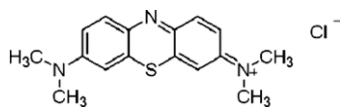 E-mail addresses: riham17@yahoo.com, riham.hazzaa@pua.edu.eg (R. Hazzaa).

Fig. 1. Chemical structure of methylene blue.

Many different techniques have applied for the removal of dyes from wastewater including photocatalytic degradation (Sohrabi and Ghavami, 2008; Sleiman et al., 2007), sonochemical degradation (Abbasi and Asl, 2008), micellar enhanced ultrafiltration (Zaghbani et al., 2008), cation exchange membranes (Wu et al., 2008), electrochemical degradation (Fan et al., 2008), adsorption/precipitation processes (Zhu et al., 2007), integrated chemical–biological degradation (Sudarjanto et al., 2006), solar photo-Fenton and biological processes (García-Montano et al., 2008), Fenton–biological treatment scheme (Lodha and Chaudhari, 2007) and adsorption on activated carbon (Hameed and Daud, 2008). Adsorption has been found to be superior to other techniques for water re-use in terms of initial cost, flexibility and simplicity of design, ease of operation. Activated carbon has been popular choice as an adsorbent for MB removal from wastewater due to its large specific surface area, low density, chemical stability, suitability for large scale production, variety of structural forms, and the ability to modify the pore structures but its high cost poses an economical problem (Rafatullah et al., 2010).

Agricultural wastes are low-cost materials used to produce activated carbon (Hussein et al., 2008). Activated carbons are manufactured from a variety of materials such as olives stone (Rodriguez et al., 2008), almond hulls (Christopher et al., 2000), peach stone (Arriagada et al., 1997), apricot stone (Petrova et al., 2010) date stone (Hameed et al., 2009; Alhamed, 2009), pistachio shell (Yang and Lua, 2006), sugarcane bagasse (Amin, 2008), pomegranate peel (Amin, 2009). These activated carbons are used for water purification (Kobya et al., 2005; El-Sadaawy and Abdelwahab, 2014).

In Mediterranean countries, olive stones and residues are a cheap and quite abundant agricultural waste. In present study, olive stone collected from Siwa Oasis in Egypt has been used for preparing activated carbon. The objectives of the study were to investigate the following:

  (i) Utilization of OS as a feedstock for activated carbon production via thermal activation
 (ii) Effect of activation temperature and time on activated carbon production
(iii) Optimum conditions for adsorption of MB onto OS and OSAC
(iv) Effect of pH, temperature, contact time and dye concentration on MB adsorption efficiency
 (v) Most appropriate adsorption isotherms, kinetics and mechanism of sorption.

2. Materials and methods

2.1. Materials

Methylene blue dye, used as an adsorbate, was purchased from Merck and used without any further purification. The molecular weight (g/mol), molecular volume (cm^3/mol) and molecular diameter (nm) of MB are 319.85, 241.9 and 0.8, respectively. MB has molecular formula $C_{16}H_{18}N_3ClS$. Chemical structure of methylene blue is shown in Fig. 1.

A stock solution of 1000 mg/L was prepared by dissolving the required amount of dye in double distilled water. Serial dilutions were prepared from the stock solution. A calibration standard curve was constructed using a UV spectrophotometer (Shimadzu Model: UV 1601) at maximum wavelength of 640 nm. The pH of solution was adjusted with 0.1M HCl or 0.1M NaOH (Merck). All the adsorption experiments were carried out at room temperature (25 ± 2 °C). All chemicals were of analytical reagent grade.

2.2. Preparation of activated carbon

Olive stone (2 kg) were collected from Siwa oasis in Egypt, washed several times with distilled water to remove dirt, dried at 105 °C for 2 h until a constant weight was reached, crushed, milled and sieved to select particles size (0.4–0.8 mm) for use. The resulting material raw olive stones termed (OS) was used as an adsorbent. A part of the raw materials were subjected to thermal activation in absence of air. The olive stones were carbonized in stainless steel tube vertical reactor placed in the muffle furnace. The activation temperature and time tested were in the ranges of 400, 500, 600, 700, 800 and 900 °C for 30, 60 120 min. 30 g sample of olive stone were used for each experiment. The activated carbon prepared is termed (OSAC). The percentage yield from each prepared activated carbon was calculated from the following equation:

$$\text{Yield\%} = \frac{W_f}{W_i} \tag{1}$$

where, W_f, W_i the mass of the resulting (OSAC) and the initial mass of the raw olive stone respectively.

The microstructures of raw olive stone (OS) and activated carbons olive stone (OSAC) were examined using scanning electron microscopy (SEM). Scanning electron microscopy was carried out on samples that were gold-coated for electrical conduction (model SEM JEOL JSM 6360 LA Japan). Fourier transform infrared (FTIR) analysis was applied on both (OS) and (OSAC) to determine the surface functional groups, by using FTIR spectroscope from 4000–500 cm^{-1} (FTIR-2000, Perkin Elmer)

2.3. Adsorption experiments

Batch adsorption experiments were carried out using a thermostated horizontal shaker with agitation speed 150 rpm to reach equilibrium in a set of Erlenmeyer flasks (250 mL) where 100 mL of MB solutions with initial concentrations of 50–120 mg/L. One gram of the adsorbent was added into each flask covered with glass stopper.