



An effective route to transform scrap tire carbons into highly-pure activated carbons with a high adsorption capacity of ethylene blue through thermal and chemical treatments



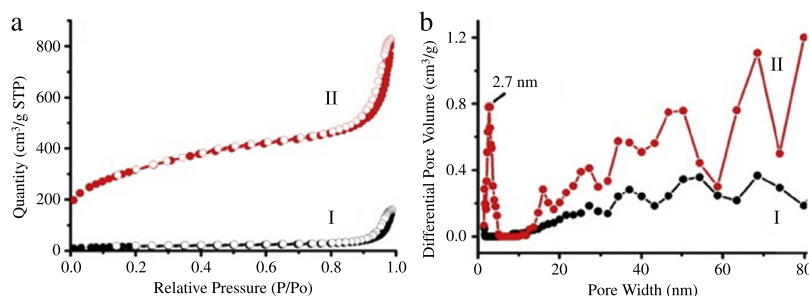
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HIGHLIGHTS

- Thermal and chemical treatments were performed to prepare HP-AC.
- HP-AC has a high adsorption capacity of 323 mg/g for MB.
- Meso-pore structures and oxygenated functional groups control the MB adsorption.
- Pseudo-second-order and Langmuir adsorption models is the adsorption route for MB.
- Electrostatic interaction is the main driving force for the adsorption of MB.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 24 September 2016
Received in revised form 4 March 2017
Accepted 20 March 2017
Available online 27 March 2017

ABSTRACT

A three-step treatment (thermal, chemical and activation) has been developed to effectively transform scrap tire carbons (STCs) into highly-pure activated carbons (HP-AC). The as-prepared HP-AC displays a specific surface area of 1048 m²/g (total pore volume of 1.16 cm³/g) with a yield of 21.4 wt%. Adsorption of methylene blue (MB) using HP-AC from aqueous solution under equilibrium and kinetic conditions was evaluated by varying initial MB concentration (0.15–20 ppm), contact time (0–120 min), solution pH (2–12) at 30°. The Langmuir isotherm model shown better fit to the equilibrium results than the Freundlich model. The adsorption capacity (q_m) of HP-AC is 323 mg/g. The kinetic adsorption results were well described by the pseudo-second-order model. XPS results reveal that electrostatic interaction between the oxygenated functional groups on HP-AC and MB is suggested as the main driving force for the adsorption process. This study demonstrates that HP-AC is an effective and low-cost adsorbent for the removal of MB from aqueous solution.

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1. Introduction

Water-soluble organic dyes are widely used as the coloration in various fields, e.g. paper, leather, textiles, and other industries. However, the release of the dyes into water systems is potentially harmful to living organisms including humans and wildlife. Methylene Blue (MB) is one of the most commonly used water-soluble organic dyes. Though MB is not strongly hazardous, however, on inhalation, it will cause rapid breathing, vomiting, shock, mental confusion, and nausea in humans. Therefore, it is essential to remove MB or other dyes from water systems as to reduce the impact to the environments. Although diverse techniques have been developed to remove dyes from water systems. Sorption is a commonly used method to remove the contaminants in water systems owing to its simplicity, high efficiency, and low cost. Activated carbon (AC) has been widely applied to remove dyes from the water systems (Gupta et al., 2012; Saleh and Gupta, 2014; Jawad et al., 2016; Wang et al., 2016), however, AC derived from conventional sources, e.g. coal, coconut shell, bamboo etc., is not beneficial for the applications in water treatments. A promising way is to obtain AC derived from biomass or industrial wastes. Herein, we aim to develop a kind of AC derived from the industrial waste with high adsorption capacity of MB.

Recycling of the industrial wastes, e.g. scrap tires, is arousing great interest amongst industrialized countries owing to the environmental impact and the demand for versatile energy resources. For the past three decades, many advances have been achieved in the recycling of scrap automobile tires, especially in the production of scrap tire carbons (STCs) (Darmstadt et al., 1995; Sahouli et al., 1996; Piskorz et al., 1999; Fernández et al., 2012; Mikulova et al., 2013) and oils (Quek and Balasubramanian, 2013; Kebritchi et al., 2013) through pyrolysis. However, the applications for STCs are significantly limited by their physicochemical properties, especially the low specific surface area, high sulfur content, and ash content. The ash content is the unfavorable for polymer reinforcement when the STCs are used as the reinforced fillers. In addition, for the transformation of STCs into activated carbons (AC), the ash components would also reduce the production yield owing to catalytic combustion of carbon during the activated process (Fierro et al., 2007). Normally, post-treatments to demineralize are required for further applications, especially in rubber reinforcements or as the precursor of AC. Accordingly, several post-treatment techniques using acids and bases have been conducted to purify STCs for reducing the ash content as low as possible. A previous study reported that acid treatments were more effective in reducing the ash content than base treatments (Mikulova et al., 2013). However, to date, it is difficult to obtain the ash content below 3.0 wt% by this process (Normally, the requirement of the ash content in polymer reinforcements should be lower than 1 wt%). This will significantly retard the use of STCs in polymer reinforcements. Therefore, more recently, much effort has been made to modify the STCs into porous carbons using steam or CO₂ activation (Betancur et al., 2009; Shan et al., 2011; González et al., 2006; Chaala et al., 1996; Mui et al., 2004; Suuberg and Aarna, 2007). These approaches are successful to open new routes for exploiting STCs as AC for removing pollutants from wastewater. Moreover, AC generated from scrap tires is a low-cost process comparing to other carbon sources, e.g. coal, woods, bamboo or others (Zhi et al., 2014). However, methods to generate highly-pure AC from STCs with high specific surface area, acceptable yield and high adsorption capacity of water pollutants is still a challenge and needed more studies. The bottleneck technique is to effectively purify STCs with a low ash content, which was reported to be a primary factor governing the specific surface area as well as the pore structures and the production yield of the as-prepared AC during the activated process. Here, we develop a new route—a three-step treatment for preparing highly-pure AC using STCs through thermal and chemical treatments. In this work, we can obtain highly pure STCs (HP-STCs) with an ash-content approximately 1.46 wt% (normally non-metal oxides are 0.48 wt%, e.g. SiO₂) and the sulfur content is around 0.22 wt%. The obtained HP-STCs is as clean as the commercial carbon blacks. Further activation of the HP-STCs by CO₂ at 900 °C can generate AC (noted as HP-AC) with specific surface area of 1048 m²/g and total pore volume of 1.16 cm³/g. Adsorption isotherm of the methylene blue on HP-AC was determined to have a maximum monolayer adsorption capacity of 323 mg/g. A Langmuir adsorption equilibrium behavior and a pseudo-second-order adsorption model are also discussed.

2. Materials and methods

2.1. Materials

Samples of STCs were supplied by a scrap tire pyrolysis plant in Taiwan, Kao Hsing Chang Co., using a low-pressure (200–600 Torr) vacuum pyrolysis at 450 °C. A commercial carbon black-N339 was supplied by China Synthetic Rubber Co. in Taiwan.

2.2. Preparation of highly-pure STCs with thermal and chemical treatments

For a typical run, the pristine sample was thermally treated at 800 °C in He flow of 45 sccm for 2 h, and followed by a treatment at 500 °C with a mixture of He/H₂ (1/3 = 60 sccm) for 2 h. After the thermal treatments, the sample was refluxed in an acid solution (HCl 2.0 M) for 4 h at 100 °C.

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