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Review

Preparation and characterization of a novel polyethyleneimine cation-modified persimmon tannin bioadsorbent for anionic dye adsorption

Xiaojuan Li [a], Zhongmin Wang [a, **], Jingliang Ning [a], Mingmin Gao [a], Wenbin Jiang [a], Zhide Zhou [b, ***], Guiyin Li [b, *]

[a] School of Materials Science and Engineering, Guilin University of Electronic Technology, Guilin, 541004, China
[b] School of Life and Environmental Sciences, Guilin University of Electronic Technology, Guilin, Guangxi, 541004, China

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A B S T R A C T

A novel and recyclable bioadsorbent (PTP) has been prepared by the cationization of persimmon tannin (PT) using polyethyleneimine (PEI) for application in the removal of the anionic dye methyl orange (MO) from aqueous solution. The physicochemical properties of the prepared PTP were characterized by Fourier transform infrared spectroscopy, scanning electron microscopy, Zeta potential measurements, Brunauer-Emmett-Teller and thermogravimetric analysis. Systematic batch adsorption experiments were carried out with pH, bioadsorbent dosage, initial MO concentration and contact time. Kinetic regression analysis indicated that the adsorption processes followed the pseudo-second order model. The equilibrium isotherm was in good fit with the Freundlich model with a maximum adsorption capacity of 225.74 mg/g. Thermodynamics data revealed that the adsorption of MO onto PTP was feasible, spontaneous and endothermic. A possible biosorption mechanism was presented where electrostatic interactions, hydrogen bonding, and π-π interactions dominated the adsorption of MO onto PTP. Moreover, the regeneration of the PTP was easily achieved and MO removal efficiency remained high (81.47%) after six cycles. The actual sewage treatment simulation was evaluated and the PTP had a good preference to adsorption MO. All these results indicated that PTP could be considered a high performance and promising candidate for the effective removal of anionic dyes from aqueous solutions.

Contents

* Corresponding author.
** Corresponding author.
*** Corresponding author.
E-mail addresses: zmwang@guet.edu.cn (Z. Wang), zzdlgy@163.com (Z. Zhou), liguiyin01@163.com (G. Li).

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1. Introduction

As a result of rapid industrialization and population expansion, the contamination of water with metal ions and organic dyes has increased significantly to the extent that it has attracted worldwide attention (Visa and Duta, 2013). Organic dyes, especially azo dyes, are commonly employed to color products such as textiles, pulp and paper, dyestuffs, and plastics, which leads to the discharge of large volumes of colored wastewater into the environment (Fan et al., 2015; Salima et al., 2013). Azo dyes are the most widely used type of synthetic dyes in textile and garment applications. About 10%–15% of the dyes are discharged into the environment without treatment and may seriously affect the health of the contact. Azo dyes can decompose and produce more than 20 kinds of carcinogenic aromatic amines under certain conditions. After being activated, it can change the DNA structure of human body to cause lesions and induce cancer (Jing et al., 2016; Sanchez-Martin et al., 2010). In addition, organic dyes tend to exhibit excellent stability to light and heat, and are resistant to biodegradation due to their complex aromatic structures (Tanhaei et al., 2015; Dias and Petit, 2015). Therefore, azo dyes wastewater must be detoxified before discharge.

At present, among the numerous techniques reported for the treatment of dye-containing wastewater, adsorption is considered one of the most promising methods due to its simplicity, effectiveness, and low cost (Dotto et al., 2017; Fan et al., 2015; Gao et al., 2010). In general, the process costs associated with such dye removal techniques depend mainly on the cost of the adsorbent and its regeneration. To date, biomaterials, modified clays, activated carbon materials, polymeric resins, and zeolites have been applied as adsorbents to remove dyes (Sahraei et al., 2017; Arshadi et al., 2016; Ghaedi et al., 2012; Munagapati and Kim, 2016). Among these adsorbents, biomass-based adsorbents (biomaterials) have received increasing attention because of their widespread availability, low-cost, and biodegradability (Tripathi et al., 2014; Chen et al., 2011; Zhao and Zhou, 2016; Song et al., 2017). Chen et al. (2011) utilized surfactant modified silkworm exuviae to explore the adsorption mechanism of methyl orange in aqueous solutions. Zhao and Zhou, 2016 reported the use of a bioadsorbent extracted from Salvia miltiorrhiza Bge (red sage) to remove methylene blue from wastewater. In addition, Song et al. (2017) demonstrated a high removal efficiency of dyes from wastewater using a cellulose nanocrystal-reinforced keratin bioadsorbent.

As an alternative potential bioadsorbent, persimmon extract (or persimmon tannin, PT), which contains multiple adjacent phenolic moieties, has received growing attention in recent years. Indeed, this material, which is obtained from young astringent persimmons and is a valuable waste material commonly discarded from the juice industry (Zhou et al., 2016), has been applied in the removal of dyes and metal ions from aqueous solutions in recent years (Karaman et al., 2014). In our previous work, we reported the preparation of various PT-based bioadsorbents and subsequent investigations into their adsorption characteristics towards metal cations present in aqueous solutions (Liu et al., 2013; Cui et al., 2013; Zhou et al., 2015).

In the context of our current study, we wished to prepare a low-cost, high-efficiency, and recyclable bioadsorbent through modifying PT to enhance its adsorption properties. For this purpose, we selected polyethylenimine (PEI), a cationic polyelectrolyte containing branched chains in addition to primary, secondary, and tertiary amino groups (1:2:1 ratio), due to its high charge density (23.3 mequiv/g in aqueous solution when fully protonated) and good biocompatibility (Jing et al., 2016; Yu et al., 2013). To the best of our knowledge, this is the first time that the PEI cationic modification of PT has been carried out with the aim of treating dye-containing wastewater. The as-prepared PTP was characterized by various of methods, such as FTIR、SEM、TGA、Zeta potential、BET. The adsorption performs of methyl orange (MO) from aqueous solutions has been investigated in detail. In addition, a possible adsorption mechanism will be proposed based on the adsorption kinetics, isotherms, and thermodynamics. Finally, the adsorption-desorption and regeneration of PTP and the actual sewage treatment simulation were evaluated in the context of practical applications.

2. Material and methods

2.1. Materials

PT powder extracted from astringent persimmon was donated by the Guangxi Huikun Company of Agricultural Products (Guangxi, China). PEI with a molecular weight of 600 was purchased from Aladdin Industrial Corporation (Shanghai, China). Glutaraldehyde (25% w/w aqueous solution) was obtained from Xilong Science Co., Ltd. (Guangdong, China). Methyl orange (MO), methylene blue (MB) and rhodamine B (RB) were purchased from Reagent Three factory (Shanghai, China). $AgNO_3$ and $Pb(NO_3)_2$ were purchased from Tianjin North Union Fine Chemicals Development Co., Ltd (Tianjin, China). All other chemicals were of reagent grade and were employed without any further purification.

2.2. Synthesis of the PTP bioadsorbent via the PEI cationic modification of PT

PT powder (10.0 g) and 25% glutaraldehyde (40.0 mL) were mixed with ultrapure water (700 mL) in a 1000 mL three-necked round-bottom flask for 30 min at 200 rpm and 323 K. PEI (3.0 g) was added and the mixture allowed to react for 6 h. After that, the suspended solution was filtered, and washed three times with ultrapure water. The washed sample was then suspended in ultrapure water (500 mL) prior to spray drying to obtain the desired PTP bioadsorbent. The conditions employed for the spray drying procedure were as follows: inlet temperature, 473 K, outlet temperature, 383 K, and feed rate, 30 mL/min. The cationization process