

Cationic dye adsorption using porous composite membrane prepared from plastic and plant wastes



Chia-Hao Lin^a, Chin-Hau Gung^a, Jeng-Yue Wu^a, Shing-Yi Suen^{a,b,*}

^a Department of Chemical Engineering, National Chung Hsing University, Taichung 402, Taiwan

^b Center of Nanoscience and Nanotechnology, National Chung Hsing University, Taichung 402, Taiwan

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ABSTRACT

This study focuses on the utilization of composite membrane made from plastic and plant wastes in cationic dye adsorption. The chopped polyethylene terephthalate (PET) bottle was dissolved in the solvent and mixed with water bamboo husk particles to form the porous composite membrane via water-vapor-induced phase inversion. The pristine particles and composite membrane were characterized in morphology, pore structure, surface area, and ion-exchange capacity. In batch process at pH 4, the saturated capacities of methylene blue and methyl violet 2B adsorption onto the composite membrane with 30 wt.% particles were 307–326 mg/g membrane. The pure PET membrane showed negligible dye adsorption. Moreover, it is found that the cationic dyes were adsorbed onto the composite membrane through a combination of electrostatic interaction, hydrophobic interaction, and hydrogen bonding. A high desorption efficiency (>90%) was then attained by the use of 1 M KSCN in 80% methanol as desorbent. In the flow-through process with one 25 mm composite membrane disc at 1 mL/min, the dye breakthrough volumes were 105–135 bed volume ($V_{\text{effluent}}/V_{\text{adsorbent}}$). It is also verified that the prepared composite membrane could be successfully reused in three consecutive adsorption/desorption cycles without deterioration in dye removal and recovery performance.

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1. Introduction

Adsorption process has received worldwide prominence for treating aqueous effluents with recalcitrant pollutants owing to its convenience and effectiveness [1–6]. The applied adsorbents include natural and synthetic materials, whose adsorptivity varies from one to another. For process improvement, the pursuit of cheap and easily-available adsorbents is important and necessary. In recent decades, the biosorbents made from plant wastes attract great interest due to their advantages in low expense, accessibility, and eco-friendliness [4–6]. Almost all the parts of plant wastes (e.g. peel, leaf, seed, hull, shell, stalk, coir, cob, pomace, pulp, etc.) could be utilized as adsorbents [1,4–39], since they contain COOH, OH, or phenolic groups which could provide several types of bindings (e.g. charge interaction, hydrogen bonding, or hydrophobic interaction) [4,6,31]. Millimeter-to-micrometer particles are prepared from raw waste materials for achieving sufficient surface area. Based on these features, the low-priced biosorbents have been demonstrated effectual for removing cationic pollutants from water [7–39].

Nevertheless, the efficient application of particulate adsorbents is still challenged by a few problems: energy consumption for particle mixing and final solid–liquid separation in batch operation; low accessible flow velocity and high pressure drop in packed-bed process for small particles. A more suitable design to conquer these problems would be making the adsorbent in the form of membrane, which provides small bed height for higher flow rate and lower pressure drop as well as large pores for convective flow [2,3,40–46]. A popular method to produce the membrane with target functionality is to fabricate porous composite membrane via filling the particulate adsorbents in polymeric matrix (also named as mixed matrix membrane). In several researches [40–46], synthetic ion-exchange resins, natural clays, and plant waste particles (banana peel, tea waste, and pomelo peel) were taken as the fillers to prepare the composite membranes with commercial polymers. Their applications in protein or pollutant adsorption were verified efficacious and simple.

The present study aims to employ the PET (polyethylene terephthalate) bottle waste as polymeric matrix and water bamboo husk particles as the fillers for making the composite membrane. Both raw materials are from the wastes. PET bottles could be obtained everywhere as the containers of drinking water and pop sodas. They are difficult to degrade, causing a disaster of environmental pollution. Water bamboo (*Zizania caduciflora* Turcz.) is one popular agricultural product in Asia. Its outer husks are usually dumped to trash field

* Corresponding author. Tel.: +886 4 22852590; fax: +886 4 22854734.

E-mail address: sysuen@nchu.edu.tw, sysuen@dragon.nchu.edu.tw (S.-Y. Suen).

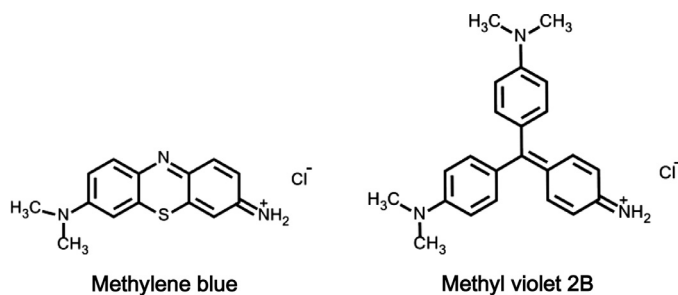


Fig. 1. Schematic diagram of cationic dye structures.

or burned down to ashes. If these wastes can be reused as membrane materials, the trash quantity in the earth would be decreased evidently. Water bamboo husk is mainly composed of cellulose, hemicellulose, and lignin [47], with small contents of protein, fat, and ash [48]. Our previous study [39] had proved that it contains COOH and OH groups and could be successfully applied in trapping heavy metal ions. In this study, the particles made from water bamboo husks would be blended with PET to form the composite membrane for cationic dye adsorption. The properties of the prepared composite membrane and its adsorption efficiency would be systematically evaluated.

2. Materials and methods

2.1. Materials

The outer husks of water bamboo were collected from the local markets in Taichung, Taiwan. The PET material was the plastic bottle of pure water bought from Vedan Enterprise Corp., Taiwan. Two cationic dyes adopted in this study were methylene blue trihydrate (80% purity, MW = 374.2, λ_{\max} = 668 nm, ICN Biomedicals, USA) and methyl violet 2B (75% purity, MW = 379.5, λ_{\max} = 575 nm, Sigma-Aldrich, USA). Their molecular structures are plotted in Fig. 1. These dyes were used as received.

2.2. Preparation and characterization of water bamboo husk particles

The water bamboo husks were cut into small pieces (about 1 cm size), washed with deionized water, and then dried in an oven at 60 °C for 24 h. The chopped husks were then ground into particles using an industrial grinder (RT-02A, Rong Tsong Precision Technology, Taiwan) at 30,000 rpm (940 W) for 3 min. The obtained particles were further sieved using a 450-mesh sieve (Retsch, Germany). For decoloration, the fine particles were immersed in deionized water at 50 °C and shaken for 24 h. Finally, the filtered particles were dried in an oven at 60 °C for 24 h. The photos of water bamboo husk waste and the prepared powders are depicted in Fig. 2.

The 90Plus Particle Size Analyzer of Brookhaven Instruments (USA) was employed to analyze the particle size limitation of fine particles. The average pore diameter and surface area of water bamboo husk particles were measured by a Micromeritics BET (ASAP 2010, USA). Prior to BET measurement, the particles were degassed under vacuum at 120 °C for 24 h.

As for the measurement of ion-exchange capacity, 0.05 g of water bamboo husk particles were submerged in 20 mL of 0.1 N HCl, and shaken for 24 h at room temperature. The particles were then constantly rinsed with deionized water for 24 h to remove the acid trace. Subsequently, the particles were equilibrated with 20 mL of 0.01 N NaOH solution for 24 h. The ion-exchange capacity was determined from the alkalinity reduction in NaOH solution by back titration using 0.01 N HCl and calculated as $(M_{O,NaOH} - M_{E,NaOH})/\text{weight of particles}$, where $M_{O,NaOH}$ and $M_{E,NaOH}$ are the moles of NaOH before and after the equilibration, respectively.

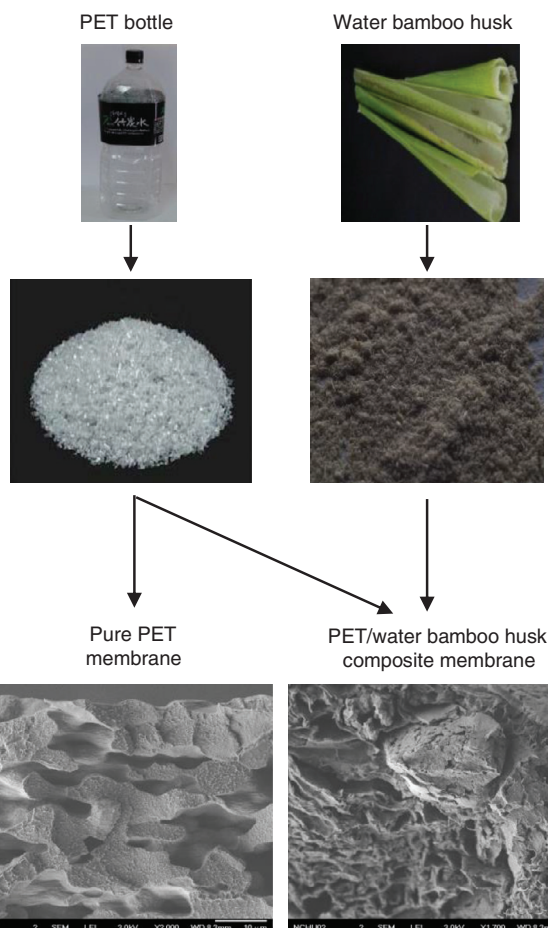


Fig. 2. Photos of original waste materials and the cross-sectional SEM for pure PET membrane and PET/water bamboo husk composite membrane.

2.3. Preparation and characterization of PET/water bamboo husk composite membrane

As shown in Fig. 2, the PET bottle was cut into 1 cm × 1 cm pieces in the beginning. 3.605 g of PET pieces (70 wt.%) were dissolved in 12 g of phenol/tetrachloroethane (7/3, w/w) at 95 °C for 24 h. 1.545 g of water bamboo husk particles (30 wt.%) were then added in PET solution and stirred uniformly for 24 h. The above casting solution was spread over a smooth glass plate using a doctor blade (200 μm) to form a film. The film was exposed to an air of >90% relative humidity for 10 min (water-vapor-induced phase inversion), followed by solvent evaporation in normal atmosphere at room temperature for 24 h (dry phase inversion). Finally, the film with glass plate was immersed in deionized water bath at room temperature until the membrane was detached from the glass plate. The prepared membrane was dried at room temperature for 24 h prior to use. It is noteworthy that the composite membranes with 40 wt.% particle content were also prepared in this study under two different conditions. In the first case, the same polymer solution (3.605 g PET + 12 g solvent) was used. However, the addition of 40 wt.% water bamboo husk particles (2.403 g) led to a high viscosity in the casting solution and failed for a proper casting. A similar phenomenon was also observed in the literature [42,46]. In the second case, the whole amount of polymer and particles was kept constant. That is, the casting solution (17.15 g) contained a lower quantity of polymer (3.09 g) but more particles (2.06 g). The resulted composite membrane became brittle and was partially cracked as water passed through in the flow process. Subsequently, 30 wt.% was considered as the optimal particle content for the composite membrane.

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