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A PEG-tannic acid decorated microfiltration membrane for the fast removal of Rhodamine B from water

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ABSTRACT

A polyether sulfone (PES) microfiltration membrane was decorated by the deposition of mixture of polyethylene glycol (PEG) and tannic acid (TA). It was used as an adsorbent for the removal of Rhodamine B (RhB) from water and had a high removal ratio (> 98.9%) at high flux up to 4777 L/(m^2·h) in dynamic test. The mixture of PEG and TA formed a stable suspension but it transformed to a viscous mixture during permeating the narrow channel of membrane, which resulted in high dispersion of the TA within the membrane and good dye adsorption. The turbulence and high driving force in the dynamic test ensured good contact of dye and the adsorbent, which led to good dye removal, while in stationary adsorption test it had only moderate dye adsorption. Molecular dynamics study revealed the molecular weight of PEG had an effect on the aggregation morphology of PEG and TA, as well as the exposure of TA on the surface, leading to different removal performance. Our study provided a simple and quick method to prepare adsorbent for the fast removal of dye from water.

1. Introduction

The removal of dye from water had attracted wide attention due to the billion tons of wastewater every year from textile industry. For the removal of dye, many technologies had been developed. Chemical degradation [1,2] and physical removal [3] were widely studied. Chemical degradation usually included photocatalysis [4,5] and electrochemical processing [6]. The physical removal was also widely studied using membrane [7,8] or adsorbent [9,10]. The membrane technique has been considered as one of the most practical one. Conventional ultrafiltration or nanofiltration usually suffered from a problem of poor water permeation only around tens of L/(m^2·h·bar) [7,11,12]. According to Hagen–Poiseuille Law $J = \varepsilon r^2 \Delta p/(8L\mu)$, the flux J of liquid was determined by the surface porosity ε, the effective pore radius r, the pressure Δp, the liquid viscosity μ, and the thickness of the membrane L. Therefore, ultrathin membranes, with thickness from tens to hundreds of nanometers and the high flux up to thousands of L/(m^2·h·bar) have been prepared by carbon nanotube, graphene and protein [13–19] as well as other materials [20].

Meanwhile, according to the equation above, another pathway should be available, that is, to increase the flux via enlarging the pore size and porosity. However, although some sponges with large pore size were reported to have good adsorption performance, they usually worked more slowly than the ultrathin membrane [9,21–24]. In addition, the preparation of adsorbent usually involved chemical vapor deposition (CVD) or other time-consuming method, which was not suitable for industrial application. As a result, we began to consider materials prepared by non-covalent bond like hydrogen bond [25–27] or coordinate bond [28,29] because the assembly was much easier and promising for field utilization. Tannic acid (TA) had strong interaction with dye [30–32] and it could be assembled with polymer [33,34]. However, at present the mixture was mainly used for gas separation or drug capsuling. Further study was required to adjust to dye removal.

In the present study, we demonstrated a polyether sulfone (PES) microfiltration membrane, which was decorated by polyethylene glycol (PEG) together with tannic acid (TA), could be used as a planar adsorbent for rapid dye removal. The mixture of PEG and TA had a special property, that it was a stable suspension in the water, but the suspension formed a viscous aggregation during the decoration and deposition. This property allowed the tannic acid to adhere to the membrane to achieve high dye removal efficiency.

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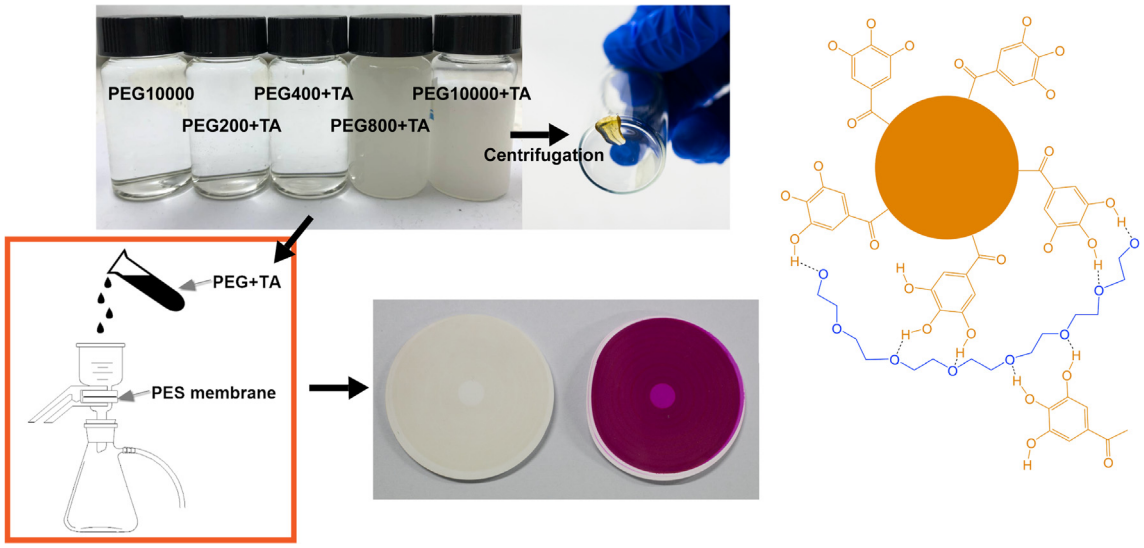

Fig. 1. Schematic of the decoration process of membrane and the filtration test. On the right showed the hydrogen bond formed between PEG (blue) and TA (orange). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2. Materials and methods

2.1. Materials

PEG with different molecular weights (MWs) were obtained from Kelong Chemical (Chengdu, China). Polyethylene oxide (PEO) with MW of 200,000 was obtained from BASF. Tannic acid (TA) and Rhodamine B (RhB) were bought from Aladdin. All the materials were used as received. PES membrane with diameter of 50 mm and pore size about 0.1 μm was supplied by Nengda (Zhejiang, China).

2.2. Decoration of membrane and separation test

The process of decoration of membrane and separation test was shown in Fig. 1. Generally, PEG and TA were dissolved in water (1 wt% for the both). Then the two solutions were mixed together (at a volume ratio of V_{PEG}:V_{TA} = 1:1, unless specially mentioned) to obtain a white suspension (for PEG 200, 400 and 600, a transparent mixture was obtained). The suspension was formed between PEG and TA via hydrogen bond as shown in Fig. 1 (on the right). The hydrogen bond weakened the hydration capacity of PEG and TA to make the mixture less soluble and led to the formation of suspension. 20 ml of the suspension was filtered by a PES membrane under vacuum, then the suspension adhered to the membrane and the membrane turned to light yellow. In addition, on the middle top of Fig. 1 showed a resin-like yellow solid obtained from the PEG10000 + TA suspension after centrifugation. It was viscous at first, but solidified after several days. For PEG from 800 to 6000, similar substance was obtained but it was more fragile. It was worth noting that the suspension didn't transform without centrifugation. This would be discussed later.

For the dynamic separation test, the simulated wastewater was prepared by dissolving the RhB into deionized water (5 mg/l). Then the water was filtrated using the membrane in a common filter driven by a water vacuum pump. For each 100 ml, the filtrate was collected and tested by an ultraviolet photometer (UV 1800, Shimadzu) to calculate the residue concentration and rejection ratio as followed:

$$Rejection\ ratio = (1-C_F/C_0) \times 100\%$$

where C_F and C_0 were the concentrations of dye in the filtrate and the as-prepared wastewater.

Another stationary adsorption test was carried out by submerging the as-prepared sample into 100 ml of wastewater for 48 h (30 °C). The concentration of wastewater was analyzed at different time to calculate the adsorption capacity:

$$q_e = (C_0-C_t)V/W_0$$

where the q_e was the adsorption capacity (mg/g), C_0 and C_t the initial concentration and the concentration at a given time of RhB and V was the volume of wastewater. The W_0 was the weight of the decorated membrane.

2.3. Characterization

The PEG, TA and their mixture was tested by FTIR (Nicolet 6700) and differential scanning calorimetric (DSC) analysis (DSC1, Mettler Toledo). The decorated membrane was characterized by scanning electron microscopy (SEM, Hitachi), attenuated total reflection FTIR (ATR-FTIR, Nicolet 6700).

2.4. Molecular dynamics (MD) simulation

Since it was difficult to understand the structure of the mixture, the MD simulation of PEG and TA system was conducted to discover the effect of PEG MW on the aggregation morphology of the mixture using the Forcite package of Materials Studio. Three PEGs of different lengths (containing 14, 46 and 226 repeat units of ethylene oxide, whose MW was 618.76, 2028.45 and 9957.99 to model the PEG600, PEG2k and PEG10000 molecule respectively) as well as TA were constructed and packed into a periodic cell containing 1000 water, 2 TA and 16/5/1 PEG (600/2000/10,000) molecules. The rectangular cells had the same initial length of 5 nm along x and y direction. The MD simulation was conducted at 298 K and the inter-atom force was described with COMPASS27 force field. The geometry optimization was run for 5000 steps to get a proper initial model, and then model was simulated in NPT ensemble first for 100 ps (step size = 1 fs) and then followed by simulation in NVT ensemble for another 2 ns. The Nosé-Hoover thermostat was used for temperature control and Berendsen method was applied for pressure control.

In order to study the aggregation of TA, radial distribution functions (RDF) $g(r)$ was analyzed by the equation below

$$g(r) = \frac{N(r)}{4\pi r^2 \rho} dr \quad (1)$$

where $N(r)$ was the number of particles over a distance between r and $r + dr$ from a given reference atom, and ρ represented the number density, which is taken as the ratio between the total number of atoms