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Ag nanoparticles loading of polypyrrole-coated superwetting mesh for on-demand separation of oil-water mixtures and catalytic reduction of aromatic dyes

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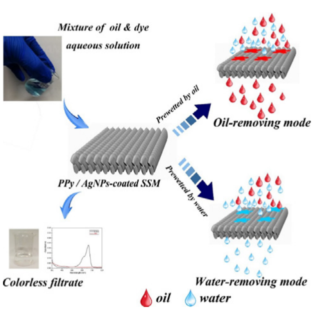
GRAPHICAL ABSTRACT

Underwater superoleophobic and underoil superhydrophobic PPy/AgNPs-coated mesh was fabricated to achieve on-demand oil-water separation and catalytic reduction of soluble aromatic dyes.



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ABSTRACT

Herein, a catalytic mesh with unique wettability, high oil-water separation efficiency and excellent catalytic performance towards aromatic dyes was fabricated. Polypyrrole (PPy) was firstly pre-coated on pristine stainless-steel mesh (SSM) surface *via* cyclic voltammetry approach. Subsequently, a simple electrodeposition process was performed to prepare and anchor Ag nanoparticles (AgNPs) onto the PPy-coated SSM surface. The PPy-coated mesh with anchored AgNPs was denoted as PPy/AgNPs-coated SSM. The obtained PPy/AgNPs-coated SSM exhibited dual superlyophobic properties and were able to achieve on-demand separation to deal with various of light oil ($\rho_{oil} < \rho_{water}$) and heavy oil ($\rho_{oil} > \rho_{water}$)-water mixtures. Importantly, benefitting from AgNPs on mesh surface, the obtained PPy/AgNPs-coated SSM exhibits exceptional catalytic activity. As proof-of-concept three typical aromatic dye molecules (methylene blue, rhodamine B and Congo red) can be effectivity degraded. Additionally, the degradation of aromatic dyes and oil-water separation were achieved simultaneously when the PPy/AgNPs-coated SSM was converted to water-removing mode. Therefore, the present work is of great significance to the development of novel oil-water filtration membranes and can open a new avenue towards the practicability of metal nanoparticle catalysts in wastewater treatment.

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1. Introduction

Serious water contamination with toxic organic pollutants has become one of the global environmental concerns caused by global industrialization and growing population. Insoluble oils and soluble dyes are the major contaminants that have a severe impact on ecosystem especially the fresh water for drinking. Removing such toxic and carcinogenic contaminants with high efficiency and low energy consumption is becoming a worldwide challenge [1–5]. Hitherto, many accessible techniques have been developed for wastewater treatment, such as oil-water separation, membrane distillation, photocatalytic degradation, absorption and so forth [6–9].

Oil-water separation technique based on superwetting membranes is broadly utilized for wastewater purification. In the last few years, superwetting materials (especially the filtration membranes) have gained considerable attention due to their excellent oil/water separation performance, energy saving and good oil/water selectivity [10–14]. Thereby, there were many significant efforts devoted to fabricating superhydrophobic - superoleophilic (named as oil-removing type) and superhydrophilic – underwater superoleophobic (water-removing type) membranes [15–24]. For example, superhydrophobic poly (vinylidene fluoride) (PVDF) membranes [25] and Cu_2S - Cu_2O composite ($Cu_2S@Cu_2O$) films [26] were fabricated to oil-water separation, respectively. Such oil-removing types are considered as an appropriate candidate for separating heavy oil ($\rho_{oil} > \rho_{water}$) from water. Nevertheless, the superhydrophilic - underwater superoleophobic (water-removing types) filters exhibited a poor separation efficiency to deal with heavy oil-water mixtures [16,23,24,27–31], which are more appropriate to separate light oil ($\rho_{oil} < \rho_{water}$) from water. To achieve on-demand oil-water separation, smart stimuli-responsive nanomaterials were widely exploited to fabricate filters with switchable separation mode between oil-removing and water-removing mode. But the conversion required continuous external stimuli, such as light illumination, temperature, electric field, pH and so on [32–40]. There is still an urgent need to fabricate such filtration membranes that can achieve switchable separation mode without any external stimulus, but these membranes are still scarcely reported. Recently, our group reported the polyaniline- and polypyrrole-coated meshes that can achieve conversion between oil-removing and water-removing mode [41–44]. However, such filtration membranes exactly cannot remove water-soluble organic dyes, especially the massive volume of aromatic dyes from the printing, textiles, leather and cosmetic industrials [45,46].

To achieve dual purification of wastewater containing insoluble oils and soluble dyes, semiconductor photocatalysts (e.g., TiO_2, ZnO, Ag_3PO_4) were employed to construct filtration membranes for integrated oil-water separation and dye degradation under UV light or even sunlight illumination. For example, a TiO_2 nanocluster-based mesh and a Ag_3PO_4-based double-layer polyester textile that can both purify oil- and dye-polluted water were fabricated [47–49]. However, the dye decomposition processes required additional UV light or even sunlight irradiation, which are required to use complex apparatus, energy-consuming and expensive. Recently, organic dye degradation over heterogeneous metal-based catalyst has received much attention owing to its easy operation and excellent catalytic performance. The catalytic activity of metal nanoparticles functions without introducing any additional energy. In this regard, metal nanoparticle as an ideal catalyst are promising for constructing oil-water separation membranes with dye degradation ability. However, the catalytic activity, reusability and stability of metal nanoparticle catalyst are largely restricted by two dilemmas [50,51]. Metal nanoparticle catalysts are prone to aggregate owing to small size. Additionally, nanoparticle catalysts are hard to collect from completed liquid reaction system and reuse. Liu et al. fabricated a polydopamine-coated stainless-steel mesh (SSM) anchored with AgNPs. The membranes exhibited superhydrophilic - underwater superoelophobic property and catalytic reduction capacity for aromatic dyes [52]. The polydopamine coating was served as a robust support can suppress AgNPs agglomeration to a large extent. The dye-polluted water can be purified, but the composite membranes can only operate water-removing mode to deal with light oil-water mixtures. The carbon nanotubes/core–shell polystyrene-Au nanoparticles composite membrane also appeared quite similar drawbacks [53].

To this end, a polypyrrole (PPy) -based catalytic mesh for on-demand oil-water separation and degradation of aromatic dyes was fabricated by a facile two-step electrochemical approach. PPy was firstly pre-coated on pristine stainless-steel mesh (SSM) surface *via* cyclic voltammetry technique. Subsequently, a simple electrodeposition process was performed to prepare and anchor Ag nanoparticles (AgNPs) onto the PPy-coated SSM surface. The PPy-coated mesh with anchored AgNPs was denoted as PPy/AgNPs -coated SSM. The as-prepared mesh exhibited underwater superoleophobic (UWSOB) and underoil superhydrophobic (UOSHB) properties. Therefore, the resulting meshes were able to separate both light oil ($\rho_{oil} < \rho_{water}$) and heavy oil ($\rho_{oil} > \rho_{water}$) from water. The obtained PPy/AgNPs -coated SSM displayed excellent catalytic performance towards organic aromatic dyes such as methylene blue (MB), rhodamine B (RhB) and Congo red (CR). Importantly, the degradation of aromatic dyes and oil-water separation were both achieved when PPy/AgNPs -coated SSM was converted to water-removing mode.

2. Experimental section

2.1. Materials

Pyrrole (98%, CP) and sodium borohydride ($NaBH_4$, 97%, AR) were purchased from KEFENG Chemical Reagent Co. Inc. Shanghai, China and Guangming Chemical Reagent Co. Inc. Shanghai, China, respectively. Silver nitrate ($AgNO_3$) from Chengdu KELONG Chemical Reagent Co. Inc. was of analytical grade. Methylene blue, Congo red and Rhodamine B were supplied by Aladdin Industrial Inc., Shanghai, China. All dye regents are all of analytical grade. SSMs (2300 mesh size) were obtained from Anping Material Tech Co. (Hebei, China). Ultrapure water (18.2 $M\Omega\ cm^{-1}$) was used throughout all the work.

2.2. Preparation of PPy-coated SSM and PPy/AgNPs -coated SSM

SSMs (3 cm $\times$ 3 cm) were ultrasonically rinsed with anhydrous ethanol, acetone and deionized water in sequence three times. The preparation methods were described in our previous work with little modification [41]. In brief, the formation and coating of PPy on pristine SSM were achieved through three-electrode cyclic voltammetry approach firstly. The saturated calomel electrode, platinum slice and a slice of cleaned SSM were performed as reference electrode, counter electrode and working electrode, respectively. 60 mL of 0.1 mol/L pyrrole in 1 mol/L hydrochloric acid solution was chosen as electrolyte. The voltage range was set from −0.2 to 0.9 V and scan velocity was adjusted to 50 mv/s. After 5 times CV numbers, PPy-coated SSM were took out and copiously rinsed with deionized water, following by drying in a 60 °C oven. Subsequently, PPy/AgNPs -coated SSMs were fabricated by a two-electrode electrodeposition process. PPy-coated SSM and a piece of cleaned pristine SSM were chosen as cathode and anode,