



Formation of phosphorylated $Zr_xSi_{1-x}O_2/Al_2O_3$ self-assembled membrane for cleaning oily seawater



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ABSTRACT

In order to deeply clean seawater containing oil, hydrolyzed polyacrylamide (HPAM) and suspended solids, phosphorylated $Zr_xSi_{1-x}O_2/Al_2O_3$ (PZSA) particles were firstly prepared through co-hydrolysis, silanization and phosphorylation, followed by coating of Al_2O_3 and then employed as a functional layer to form PZSA self-assembled membranes on porous supports. PZSA particles were characterized by scanning electron microscope (SEM), energy dispersive X-ray detector (EDX), Fourier transform infrared (FT-IR) and X-ray diffraction (XRD) while the self-assembled membranes were analyzed using SEM. Moreover, the optimum formation conditions of PZSA self-assembled membrane thickness were determined. The membranes formed under the optimal conditions were used to treat oily seawater and the quality of permeate water was investigated and analyzed. The results indicate that the particle size of PZSA is mainly distributed between 400 and 600 nm, Zr elements have been successfully doped into the lattice of SiO_2 and $\gamma-Al_2O_3$ has been coated on the surface of $Zr_xSi_{1-x}O_2$. Meanwhile, PZSA self-assembled membranes with desirable separation and selective adsorption properties perform attractive oil and COD retention ratio and water yield. Therefore, PZSA self-assembled membranes have the potential application in treating and cleaning oily seawater.

1. Introduction

Membrane processes have received significant attention as a promising technology for cleaning oily sewage [1] because of their high efficiency, deeply cleaning and no chemical additives compared to traditional separation methods [2–4]. But then, membrane fouling problem still remains one of major obstacles to the wider applications of membrane technology [5,6]. Recently, many strategies such as surface modification, doping hydrophilic polymers [7,8] or inorganic nanoparticles [9–11] into casting solutions have been investigated to enhance anti-fouling and hydrophilic properties of polymer membranes. Though these methods can improve the performance of polymer membranes to some degree, it may not solve the inherent deficiencies of polymer membranes such as high energy consumption, high membrane costs, poor selectivity, dramatic loss of permeation flux and complex preparation process of composite membrane [10,11]. These drawbacks also seriously hamper the widespread application of polymer membranes. Therefore, now looking for a type of membrane technology with low energy consumption and cost, high permeation flux and selectivity, simple preparation process becomes an important task to all researchers.

In recent years, dynamic membrane technology represents a promising substitution for conventional membrane technologies due to high retention efficiency of oil and suspended solids, high permeate flux, simple preparation process and low cost etc [12,13]. In addition, dynamic membrane can be simply formed on the surface of porous supports using a feed solution containing fine particles [14]. According to its formation process, dynamic membrane could be mainly divided into three basic categories: self-forming membrane, pre-coated membrane and composite dynamic membrane [15,16]. Many studies have indicated that the composite dynamic membrane made from two or more kinds of coating materials seemed to exhibit better separation performance in comparison to the dynamic membrane from single coating material [17–19]. For example, Yang et al. [17] successfully prepared the Kaolin/ MnO_2 composite dynamic membrane with high permeate flux and good retention ratio for the oily wastewater treatment. Wang et al. [18] found that the dextran-Zr composite dynamic membrane was able to reject hemoglobin completely, simply at the expense of a smaller permeate flux. In addition, the coating materials, such as $Mg(OH)_2$, MnO_2 , $Fe(OH)_3$, ZrO_2 , TiO_2 and so on, have already been reported to prepare the dynamic membranes for treatment of oily wastewater [20–24]. The membrane coating material

Abbreviations: PZSA, phosphorylated $Zr_xSi_{1-x}O_2/Al_2O_3$; HPAM, hydrolyzed polyacrylamide; SZP, phosphorylated Zr-doped hybrid silica; PSF, polysulfone

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plays a crucial role in the performance of dynamic membrane treatment systems. However, the above mentioned materials which can form dynamic membrane indeed lack the good selectivity, functionalized surfaces, good size and spherical shape of particle, limiting the enhancement of properties of dynamic membrane. Therefore, we also need to further study and develop new functional materials for promoting dynamic membrane technology progress.

Generally, after pretreatment of oily seawater, there are many new pollutants such as flocculants (hydrolyzed polyacrylamide HPAM) and surfactants, etc [25,26]. The existence of HPAM leads to the increase of wastewater viscosity and emulsification, and HPAM molecules with unique linear structure easily wrap with oil droplets of wastewater through the channels of conventional membrane [27]. For these reasons, conventional membrane technology could not effectively clean pretreated oily seawater with HPAM. In recent years, adsorption is considered as particularly attractive and effective process for removing contaminants and cleaning wastewater. Especially, γ - Al_2O_3 with large specific surface area and strong electric adsorption capacity has been widely used in the purification of water [28–30]. Some studies [31] have shown that excellent electric adsorption performance of Al_2O_3 is also highly effective to remove HPAM in wastewater. However, Al_2O_3 -coated diatomite does not show the performance of separating soluble oil and the grain size is larger. As a result, the purpose of the simultaneous separation of oil, HPAM and suspended solids in oily sewage can not be achieved through the formation of the above mentioned membranes. Therefore, we need to further study and develop new functional materials for promoting composite membrane and dynamic membrane technology progress, achieving the purpose of the simultaneous separation of oil, HPAM and suspended solids.

Based on our previous work [32,33], PZSA particles are prepared and then are used for the formation of self-assembled membranes on porous supports through combining the advantages as below: first, PZSA particles have various point defects inside the structure and lots of hydroxide radicals on the surface, which could enhance the hydrophilicity and permeate flux of membranes; second, the lipophilic chains of PZSA particles formed by silanization modification could effectively adsorb oil in seawater; finally, γ - Al_2O_3 coated on the surface of SZP has an excellent adsorption effect on HPAM in seawater, which makes it difficult for linear HPAM to pass through the small channels of membranes. Therefore, PZSA self-assembled membranes with hydrophilicity, anti-fouling, selective adsorption performance and good membrane channels are used in the treatment of oily seawater.

In this paper, to effectively remove oil, HPAM and suspended solids in oily seawater, PZSA particles were firstly prepared and then employed as a functional layer to form PZSA self-assembled membranes on porous supports. PZSA particles and PZSA self-assembled membranes were characterized and studied. The optimum formation conditions of PZSA self-assembled membrane thickness were determined. In addition, PZSA self-assembled membranes were used to treat oily seawater and the quality of permeate water was investigated and analyzed.

2. Experimental

2.1. Materials and reagents

Zirconium oxychloride ($\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$, AR grade, 99.0%), tetraethyl orthosilicate (TEOS, AR grade, 28.0%), phosphorous acid (H_3PO_3 , AR grade, 99.0%), cetyltrimethylammonium bromide (CTAB, AR grade, 99.0%) and aluminum nitrate ($(\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O})$, AR grade, 99.0%) were purchased from Tianjin Guangfu Fine Chemicals Co., Ltd. Formaldehyde (HCHO, AR grade, 37.0%) was provided by Tianjin Bodi Chemical Co., Ltd. (3-aminopropyl)triethoxysilane (APTES, AR grade, 97.0%) was obtained from Chenguang coupling agent Co., Ltd. Anhydrous ethanol ($\text{C}_2\text{H}_5\text{OH}$, AR grade, 99.8%), glacial acetic acid (CH_3COOH , AR grade, 99.5%) and aqueous ammonia ($\text{NH}_3 \cdot \text{H}_2\text{O}$, AR

Table 1
The detailed composition of diatomite.

Component	Content
SiO_2 [wt%]	≥ 85.0
Al_2O_3 [wt%]	≤ 5.0
Fe_2O_3 [wt%]	≤ 2.0
CaO [wt%]	≤ 1.0
MgO [wt%]	≤ 0.9
Pb [mg L^{-1}]	8

grade, 25.0%) were bought from Tianjin Jiangtian chemical technology Co., Ltd. Diatomite was obtained through Qingdao Sanxing Diatomite Co., Ltd. The detailed composition of diatomite is shown in Table 1. All reagents and chemicals were used as received.

2.2. Preparation of PZSA particles

PZSA particles with the diameter of around 400–600 nm were prepared in our laboratory. PZSA particles were prepared through co-hydrolysis, silanization and phosphorylation, followed by coating of Al_2O_3 . The preparation process of PZSA particles is schematically shown in Fig. S1. The synthesis approach of SZP particles is similar to the literature [33], and the detailed procedures are as follows:

$\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$ and TEOS were firstly mixed fully in a conical flask, and APTES was then added to the conical flask for mixing. Then the whole mixture under magnetic stirring reacted for 3 h at 30 °C and stirred to form the gelatin form, and followed by being settled in room temperature for 12 h, then dried for 24 h in 100 °C. Then spread in the mixed solution of H_3PO_3 and HCHO, refluxed under the temperature of 80 °C for 6 h, then cooled down to room temperature, repeatedly centrifuged and washed. And then dried in 100 °C to constant weight, and finally samples of SZP particles were obtained.

The prepared SZP particles were dissolved in distilled water, adding a certain amount of $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, magnetic stirring solution for 10 min. Then $\text{NH}_3 \cdot \text{H}_2\text{O}$ solution was added dropwise into the mixture whose pH had been adjusted to 9.0, under magnetic stirring reacted for 10 h at 40 °C. Afterwards, the reaction solution was cooled down to room temperature, dried to a constant weight at 60 °C after being washed to neutral centrifugal. Finally, samples of PZSA particles with different diameters were obtained.

2.3. Formation of PZSA self-assembled membranes

The experimental device of self-assembled membrane is schematically shown in Fig. 1. The self-assembled membrane system consists of coating solution tank, oily seawater tank, pump, self-assembled membrane module, flowmeter, pressure gauge, valves and piping. The magnetic stirrer was used in the coating solution tank to ensure that diatomite and PZSA particles suspension were well dispersed. The formation process of PZSA self-assembled membranes is divided into two steps. Firstly, a certain concentration of diatomite suspension in the

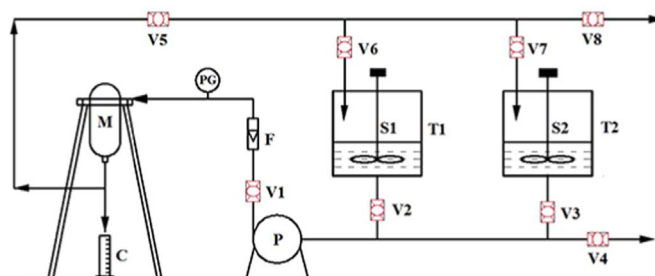


Fig. 1. Schematic diagram of experiment setup: M self-assembled membrane module; F flowmeter; P pump; C measuring cylinder; PG pressure gauge; V1–8 valves; T1 coating solution tank; T2 oily seawater tank; S1, S2 stirrer.

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