



# Graphene oxide/triethanolamine modified titanate nanowires as photocatalytic membrane for water treatment

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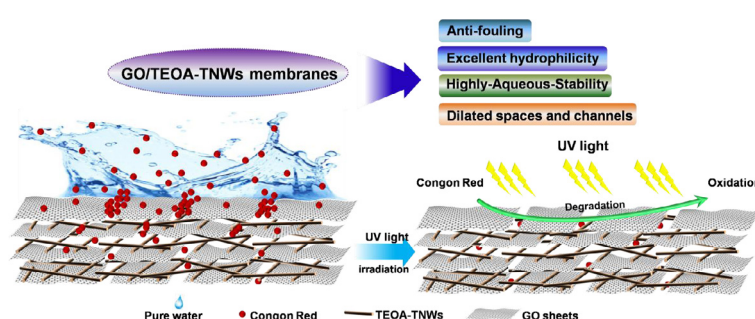
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## HIGHLIGHTS

- GO/TEOA-TNWs composite has been prepared via a one-step synthesis approach.
- Intercalation of TNWs into GO layers can produce more 2D nanochannels.
- The GO/TEOA-TNWs membranes exhibit anti-fouling ability for water treatment.
- The GO/TEOA-TNWs membranes show high water flux and rejection to organic dyes.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Graphene oxide (GO) membranes are a promising candidate for water treatment, such as sewage purification and desalination, due to its high mechanical strength, superior flexibility and hydrophilic property. However, one of their major application challenges is the membrane fouling, resulting in membrane blocking and poor water flux. Here, we incorporate triethanolamine (TEOA) modified titanate nanowires (TNWs) with GO membranes to resolve the fouling problem by taking advantage of enhanced membrane photocatalytic properties. The results show that the composite membranes exhibited remarkable photocatalytic degradation of Congon Red dyes under UV-light during filtration separation, and membrane fouling could be significantly alleviated after UV-light irradiation, leading to high water treatment performance for long-term cycling use. Furthermore, intercalation of TNWs between GO layers can produce a great quantity of nanochannels inside the membranes such that significantly increases the water flux. Besides, the presence of TEOA can evidently improve the aqueous stability for GO membranes, which is also an essential requirement for practical application. Our work demonstrates that endowing GO membranes with photocatalytic properties is an effective approach to resolve the fouling problem. And the as-prepared GO/TEOA-TNWs composite membranes are a promising material with anti-fouling function and highly-aqueous-stability for long-term practical water treatment applications.

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## 1. Introduction

Water pollution from natural organic matters, industrial dyes, microorganisms and heavy metals is a rising worldwide environ-

mental issue [1–4]. Highly efficient and clean water treatment technologies are urgently needed for both of research and industry. The membrane technologies have been widely applied in the water treatment industry due to its high efficiency, low energy-consuming and easy operation [5,6]. Beyond the traditional porous polymer membranes, graphene oxide (GO) based membranes, re-stacking from two-dimensional GO nanosheets, have been

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regarded as a promising candidate for water purification and desalination [7–10]. By taking advantages of its high mechanical strength/flexibility, excellent hydrophilic surface properties and 2D interconnected nanofluidic channels for ion and molecular transport, GO based membranes have exhibited extraordinary separation performance in respect of water flux and pollutant molecular/ions rejection [11–15]. However, one of their major application challenges is the membrane fouling, resulting in GO membranes blocking and poor water flux, which would be much more serious when separating organic pollutants in water [16–18]. In addition, owing to the electrostatic repulsion between GO nanosheets in aqueous solution, the aqueous stability of neat GO membranes is poor and they are normally tended to disintegrate [19]. Therefore, long-term practical applications of GO based membranes for water treatment have been significantly hindered by the above two main problems.

Recently, a strategy of endowing GO membranes with photocatalytic properties to degrade the organic pollutant has been proposed to resolve the membrane fouling problem [20,21]. Sun and coworkers reported a bifunctional GO-TiO<sub>2</sub> microspheres deposited polymer filtration membrane, which could synchronously degrade the organic pollutants during water filtration, resulting in a high water flux and anti-fouling performance [22]. Xu and coworkers synthesized GO-TiO<sub>2</sub> nanoparticles filtration membrane and found out that incorporation of TiO<sub>2</sub> photocatalyst into GO membranes can not only reduce the membrane fouling problem by enhanced photocatalytic properties but also produce much more dilated space and nanochannels during GO nanosheets in the membrane [23]. However, many of the current reports pay main attention on the initial pollutants degradation performance. The recycling ability and physical stability of GO composite membranes with photocatalytic properties for long-term use are needed to be focused on as well.

In the present work, we rationally incorporating triethanolamine (TEOA) modified titanate nanowires (TNWs) with GO membranes to resolve the fouling and long-term aqueous stability problems. TNWs were firstly synthesized by hydrothermal treatment of commercial TiO<sub>2</sub> nanoparticles. Following modification of TNWs with TEOA via a simple sonication mixing method, the composite membranes (GO/TEOA-TNWs) were fabricated by vacuum filtration of GO and TEOA-TNWs mixing suspension, which can be directly used for water filtration treatment. The results show that the composite membranes exhibited remarkable photocatalytic degradation of Congon Red dyes under UV-light during filtration separation, and membrane fouling could be significantly alleviated after UV-light irradiation, leading to high water treatment performance for long-term cycling use. Furthermore, intercalation of TNWs between GO layers can produce a great quantity of nanochannels inside the membranes such that significantly increases the water flux. Besides, the presence of TEOA can evidently improve the aqueous stability for GO membranes, which is also an essential requirement for long-term practical application. It was clearly demonstrated that the as-prepared GO/TEOA-TNWs composite membranes are a promising material with anti-fouling function and highly-aqueous-stability for long-term practical water treatment applications.

## 2. Experimental section

### 2.1. Chemicals and materials

TiO<sub>2</sub> nanoparticles (average particles size of 40 nm) were purchased from Klamar Chemical Reagent Co. Ltd. (Shanghai, China). The TNWs were synthesized by hydrothermal treatment of TiO<sub>2</sub> nanoparticles in 60 mL NaOH (10 M) solution, as reported

elsewhere [24]. Triethanolamine (TEOA) modified TNWs was obtained via a simple sonication mixing process. Typically, 0.5 g of TNWs with 100 mL H<sub>2</sub>O was under ultrasonic dispersion for 30 min. Then 10 mL of TEOA was added into the suspension and the mixture continued ultrasonic treatment for 3 h. After filtrating, washing and drying, TEOA modified TNWs was obtained. GO was synthesized following the modified Hummers method, as shown in our previous work [25].

### 2.2. Fabrication of GO and GO composite membranes

Pure GO membranes were fabricated by vacuum filtration of the as-prepared GO suspension through a porous cellulose membrane (47 mm in diameter, 0.45  $\mu$ m pore size). Typically, 2 mL of GO dispersion (0.05–0.35 mg/mL) was filtrated to fabricate GO membranes, and it was stabilized for 30 min under a pressure of 0.3 MPa by nitrogen filling. For the preparation of GO/TEOA-TNWs membranes, typically, 10 mg of as synthesized GO was well dispersed in 40 mL of deionized water. Then, 20 mg of TEOA modified TNWs was added to the GO suspension. And the mixture was sonicated for half hour. Finally, 2 mL of the mixture was filtrated to fabricate GO/TEOA-TNWs membranes, and it was stabilized for 30 min under a pressure of 0.3 MPa by nitrogen filling. The preparation of other GO composite membranes adopted similar method.

### 2.3. Test of membrane flux, rejection and photocatalytic properties

Water flux and rejection performance of GO and its composite membranes were investigated using a dead end membrane filtration system, as shown in Fig. S1. It consists of a feed tank pressurized with a nitrogen cylinder. The pressure can be adjusted by nitrogen filling. Congon Red solution (10 mg/L) was used to simulate dye-containing wastewater, and the dye concentration of the feed was determined by a UV–Vis spectrometer (UV-752 N). The flux was calculated on the basis of permeate mass divided by effective surface area and filtration time, with a unit of L/(m<sup>2</sup>h). A high-pressure 250 W mercury lamp was used as the UV-light source. When investigating the anti-fouling performance of GO/TEOA-TNWs composite membranes, the prepared membranes were both treated through a 5 h UV-light irradiation before the Congon Red removal process. After every 3 h filtration for Congon Red, intermittent UV-light irradiation is adopted. The continuous filtration of the membranes without UV-light irradiation was also conducted as a control experiment.

The separate photodegradation activity of GO/TEOA-TNWs composite membranes was also studied. The membranes (21.8 mg) were put into an aqueous solution (100 mL) containing of 1 mg Congon Red. Before UV-light irradiation, the suspensions were kept in the dark for 0.5 h to establish an adsorption-desorption equilibrium. The 752 N UV–Vis spectrophotometer with a detection wavelength at 664 nm was used to analyze the sampled suspension. The changes in maximum absorption versus irradiation time ( $C/C_0$  versus  $t$ ) were obtained, which were used to reflect the decrease in the Congon Red concentration.

### 2.4. Characterization

Fourier-transform infrared (FT-IR) spectra were recorded on a Nicolet 6700 spectrophotometer using KBr pellets. The charge properties of TNWs and TEOA-TNWs were analyzed using Zeta potentials measurement (Zetaplus, Brookhaven, USA). X-ray diffraction (XRD) was carried out using a Japan Rigaku D/MAX-2500 instrument with a Cu K $\alpha$  radiation and a scanning rate of 5 °/min. Morphology of the membranes was observed by a scanning electronic microscope (Nova, Nano SEM230, USA). X-ray photoelectron spectroscopy (XPS) analysis was conducted on Thermo

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