



## Original article

# Preparation of a novel composite electrode based on N-doped TiO<sub>2</sub>-coated NaY zeolite membrane and its photoelectrocatalytic performance



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## ABSTRACT

For the first time the preparation of the N-doped TiO<sub>2</sub>-coated NaY zeolite membrane (N-doped TiO<sub>2</sub>/NaY zeolite membrane) as an electrode material for photoelectrocatalysis has been achieved and reported. The XRD, SEM, UV–vis and XPS techniques were used to characterize the structure of the N-doped TiO<sub>2</sub>/NaY zeolite membrane. The results verified that the surface of the N-doped TiO<sub>2</sub>/NaY zeolite membrane was coated by TiO<sub>2</sub> nanoparticles of ca. 20 nm size and exhibited a distinct red-shift in the UV–vis spectra compared to N-doped TiO<sub>2</sub>. The photoelectrocatalysis performance of the N-doped TiO<sub>2</sub>/NaY zeolite membrane electrode was evaluated by phenol degradation. The results revealed it is a promising novel electrode material for application of photoelectrocatalysis in the removal of organic contaminants in waste water.

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## 1. Introduction

Contamination of natural waters is considered one of the most serious threats to human beings. The combined technology involving use of photocatalytic (PC) and electrochemical (EC) processes for the removal of harmful organic compounds in wastewater has developed rapidly in recent years [1,2]. Photoelectrocatalysis (PEC) represents an advantage over photocatalysis since it applies a potential across a photoanode on which the catalyst is supported. This configuration allows for a more effective separation of the charges generated ( $e^-/h^+$ ) in the process, thus increasing the lifetime of the electron–hole pairs [3]. The actual implementation of PEC technology is critically dependent on the characteristics of the semiconductor electrode materials. Among the various electrode materials studied, TiO<sub>2</sub> is the most common candidate for the destruction of organic pollutants due to its high photocatalytic activity, good stability, non-toxicity and low cost. However, its wide band gap ( $\sim 3.2$  eV of anatase) limits the effective application of TiO<sub>2</sub> under solar irradiation [4,5]. Therefore, many composite electrode materials involving TiO<sub>2</sub> have been investigated, such as, TiO<sub>2</sub>/SrTiO<sub>3</sub> [6], WO<sub>3</sub>/TiO<sub>2</sub> [7], Ti/TiO<sub>2</sub>

annotates [1], Bi<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> [2], Au-NPs/TiO<sub>2</sub> annotate [8] and Cr-doped TiO<sub>2</sub> annotates [9]. The doping with nonmetal elements in a TiO<sub>2</sub> nanostructure has shown encouraging results for the development of visible, light-active photoelectrocatalysts [10–12]. To achieve pre-concentration of pollutants and resolve the difficult separation of TiO<sub>2</sub> particles, immobilization of TiO<sub>2</sub> on an adsorbent, or an inert support, to form integrated photocatalytic adsorbents (IPAs) has been recommended [13]. Using IPAs, degradation of pollutants can be achieved by the simultaneous effects of physical adsorption by the adsorbent and photochemical degradation by the immobilized TiO<sub>2</sub>. To realize this, TiO<sub>2</sub> particles are typically dispersed on supports with high surface area such as activated carbon [14,15], and zeolite [16].

Zeolites consisting of three-dimensional structures of SiO<sub>4</sub> and AlO<sub>4</sub> tetrahedrally linked by oxygen atoms to form a cage-structure possessing good adsorption abilities, uniform channels and regular pores. Thereby, TiO<sub>2</sub> can be facily supported in the existing channels or surface of the zeolite particles. Recently, the studies involving TiO<sub>2</sub>/zeolite IPAs have exhibited good photocatalytic performance for the removal of organic compounds such as dyes [17–19], humicacids [20] and pharmaceutical compounds [21] attributed to the advantages of IPAs. With respect to electrocatalysis, the zeolite modified electrodes (ZME) exhibited some good features for the fast, easy and economical electrooxidation of formaldehyde [22]. Furthermore, ZME also have been employed in

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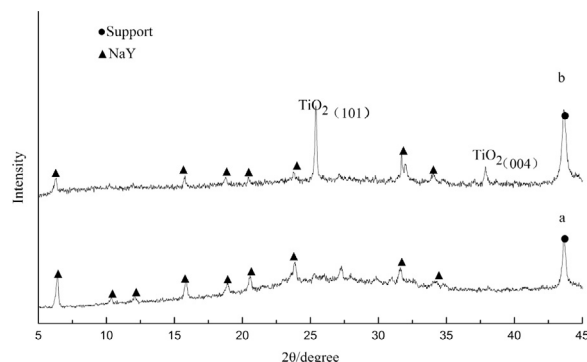
the oxidation of ascorbic and uric acid, cysteine, methanol and ethanol and so on [23–25]. Zeolite membranes, as the intergrown material of zeolite crystals, have potential advantages in many applications, such as electrocatalysts and an electrolyte membrane for fuel cells [13] and appears promising to be developed as a novel integrated photocatalytic adsorbent for the removal of organic compounds. However, to date, the study of electrode material based on zeolite membrane-supported N-doped TiO<sub>2</sub> used in photoelectrocatalysis has never been reported. This work prepared a novel photoelectrocatalytic electrode material based on N-doped TiO<sub>2</sub>-coated NaY zeolite membrane, and studied its photoelectrocatalytic performance for phenol degradation.

## 2. Experimental

**Preparation of zeolite membrane-based electrode material:** The porous stainless steel (stn stl) disc measuring 20 mm in diameter and 2 mm in thickness with a porosity of 75% and an average pore size of 0.2 μm was polished with sandpaper (500 mesh), then treated in an aqueous solution of sodium hydroxide (10 mol L<sup>-1</sup>) for 12 h, dried at 120 °C for 24 h and transferred into a desiccator until further use (refer to our previous work [26] for the preparation of the Y-type zeolite membrane). The precursor of N-doped TiO<sub>2</sub> was synthesized using sol-gel method [27] and *n*-butylamine as the doping N source. The N-doped TiO<sub>2</sub>-coated NaY zeolite membrane was prepared by using the dip-coating method. One side of the supported NaY zeolite membrane was dipped in the precursor of N-doping solution for 30 s, and the procedure was repeated at least three times. After drying in air at 60 °C, the substrate was calcined at 500 °C for 6 h. After cooling, the resulting N-doped TiO<sub>2</sub>-coated NaY zeolite membrane was washed several times with deionized water until pH 7 and dried at r.t.

**Material characterization:** The formation of zeolite membranes was confirmed by X-ray diffraction (XRD) using a Bruker-AXS D8 powder diffractometer with Cu Kα (λ = 0.154 nm) radiation (40 kV and 30 mA). The surface morphology was examined by a scanning electron microscopy (SEM) (S4800II, Hitachi) at an acceleration voltage of 15 kV. The UV-vis light absorption spectrum was obtained on a Varian Cary 5000 spectrophotometer equipped with an integrating sphere assembly. The XPS analysis adopted ESCALAB 250Xi spectrometer (Thermo Scientific).

**Measurement of catalytic performance:** The photoelectrocatalytic activity of the above prepared electrode material was studied by using probe pollutants-phenol under simulated solar-light radiation flux (Source: xenon lamp, PLS-SXE300, 300 W, 200–2500 nm in the wavelength) and an electric field of 8 V. Herein, the supported N-doped TiO<sub>2</sub>-coated NaY zeolite membrane, as electrode material, was used as the anode and the porous stn stl disc was used as cathode. The degradation rate of phenol was calculated by measuring the varied levels of the concentration of phenol before and after reaction via UV-vis spectrophotometer (Lambda 850, Perkin Elmer Co.). The evaluation was performed in



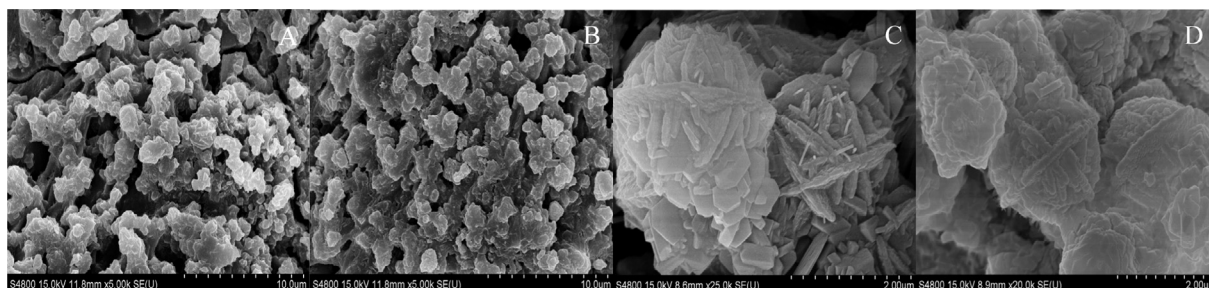
**Fig. 1.** XRD patterns of the porous stn stl supported NaY zeolite membrane (a) and N-doped TiO<sub>2</sub>/NaY zeolite membrane (b).

100 mL phenol solution at a concentration of 90 mg/L under magnetic stirring. After the completion of degradation, the N-doped TiO<sub>2</sub>/NaY zeolite membrane was regenerated by calcination at 500 °C. The recycling performance of the composite electrode material was also studied.

## 3. Results and discussion

**Fig. 1** shows the XRD patterns of the as-synthesized NaY zeolite membrane (a) and the N-doped TiO<sub>2</sub>/NaY zeolite membrane (b). The resulting as-synthesized NaY membrane displays the standard peak of the FAU-type structure (JCPDF:39-1380) as shown in **Fig. 1a**, confirming that the as-synthesized NaY zeolite membrane has been successfully grown on the porous stn stl disc. After coated by TiO<sub>2</sub> precursor and then calcined, the substrate-supported NaY zeolite membrane appears as the fresh peaks at 2θ = 25.3° and 37.8°, which are attributed to the anatase crystal structure of TiO<sub>2</sub>. This suggests that the TiO<sub>2</sub> particles have been successfully coated on the NaY zeolite membrane. **Fig. 2** shows SEM images of zeolite membrane before and after coated by TiO<sub>2</sub>. It can be observed that the NaY zeolite membrane is of relatively better integrity. In contrast to the magnification images, the NaY zeolite membrane consists of the flower-like intergrown crystals (**Fig. 2C**), and the surface of the N-doped TiO<sub>2</sub>/NaY zeolite membrane is fully coated by TiO<sub>2</sub> nanoparticles with the size of ca. 20 nm (**Fig. 2D**).

**Fig. 3** shows the UV-vis spectra of the N-TiO<sub>2</sub> and the N-doped TiO<sub>2</sub>/NaY zeolite membranes. Besides the typical UV absorption band in the spectra of both samples, a new absorption band appears in the visible range of between 400 nm and 600 nm. This verifies that there is an existing N-Ti banding in N-TiO<sub>2</sub>. Compared to the N-doped TiO<sub>2</sub> without the supporting NaY zeolite membrane, the visible absorption band of the N-doped TiO<sub>2</sub>/NaY zeolite membrane take place as a distinct red-shift. It is likely to be ascribed to the effect of the nano-pore cavity of porous zeolite. Since a part of N-TiO<sub>2</sub> nanoparticles are encapsulated in the nano-pore cavity by the



**Fig. 2.** SEM images of supported NaY zeolite membrane (A and C (magnification)) and N-doped TiO<sub>2</sub>/NaY zeolite membrane (B and D (magnification)).

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