



One-pot hydrothermal synthesis of zeolite/sodium tantalate composite and its photodegradation of methyl orange



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ABSTRACT

Sodalite/NaTaO₃ composite was prepared by a one-pot hydrothermal synthesis method. Sodalite and NaTaO₃ grow interpenetrated, and the resulting composites have similar morphology as the pure sodalite. The sodalite/NaTaO₃ composite has a lower band gap of 3.35 eV due to the heterogeneous doping effect, and exhibits an enhanced photodegradation of methyl orange under UV irradiation as compared to the pure NaTaO₃. A slight structure distortion is found for NaTaO₃ after removing sodalite by acid washing the sodalite/NaTaO₃ composite, and such result further confirms the co-growth of the two crystals. This one-pot hydrothermal method opens up new avenues for the preparation of photocatalytic composites.

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1. Introduction

Perovskite-type sodium tantalate (NaTaO₃) has been reported as an efficient photocatalyst for overall water splitting under ultraviolet (UV) irradiation [1,2]. However, it only can be activated in the low-UV region because of its large band gap ($E_g \approx 4$ eV). Reducing the band gap is the only way to achieve the visible-light activity, and the doping technique is widely used to narrow the band gap [3–7]. Doping generally includes the introduction of heterogeneous elements [3–5] and self-doping [6,7]. It has been intensively investigated since Asahi et al. reported nitrogen-doped TiO₂ in 2001 [8]. To date, nearly all non-metals have been explored as dopants to modify TiO₂ structure for improving its photocatalytic activity [9]. Nitrogen-doped NaTaO₃ has also been reported and it shows enhanced visible-light photocatalytic activity [10–12]. The effect of codoping to improve the photocatalytic performance of NaTaO₃ was investigated by using hybrid density functional calculations. The band gap is reduced significantly for the Mo–N, C–N and N–P codoped NaTaO₃ to shift the absorption curve to the visible region [12,13].

Zeolites are a class of microporous crystalline solids with uniform pores and channels (<2 nm), and they are widely used in the areas

such as catalysis, ion exchange, separation, and adsorption [14]. Zeolites have been used in zeolite/titania composites to promote their photocatalytic activities, such as photocatalytic degradation of dyes [15,16], aquatic pollutants [17], and photocatalytic oxidation of nitrogen oxide [18]. Zeolites could also increase the grain size of the zeolite/titania composites, rendering it easier to separate and reuse [15,16]. As hydrothermal synthesis is an effective method for preparing both crystalline ceramic powders [7,19,20] and zeolites [21,22], it is possible to combine ceramic powders and zeolites together in a one-pot hydrothermal synthesis.

In this work, Ta₂O₅ was used as the starting material, and it transfers into NaTaO₃ in alkaline solution under hydrothermal condition (150 °C) with the co-growth of zeolite sodalite. Microsized composites were prepared with similar morphology as pure zeolite sodalite. The resulting interpenetrated sodalite/NaTaO₃ composites are expected to show synergism of both zeolite sodalite and NaTaO₃. The band gap of NaTaO₃ is also found to be red shifted due to the “doping effect” of heterogeneous zeolite.

2. Experimental

2.1. One-pot hydrothermal synthesis of zeolite/NaTaO₃ composite

Zeolite/NaTaO₃ composite was prepared by a one-pot hydrothermal synthesis method. Typically, 6 g of NaOH was dissolved in

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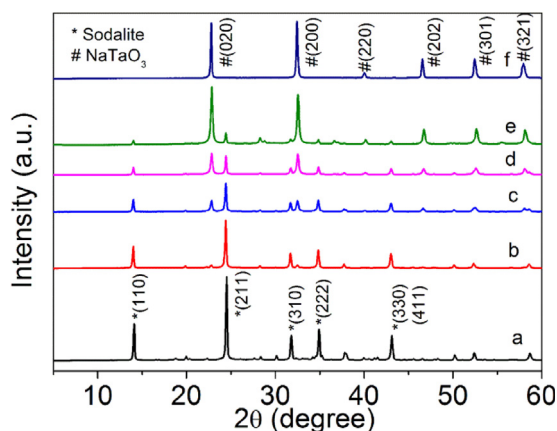


Fig. 1. XRD patterns of sodalite/NaTaO₃ composite synthesized at 150 °C with different amounts of Ta₂O₅: ZT-0 (a), ZT-0.1 (b), ZT-1 (c), ZT-2 (d), ZT-4 (e), and pure NaTaO₃ (f).

47.5 g of deionized water under stirring. The solution was divided into two equal parts. 5.89 g of NaAlO₂ and 0–4 g of Ta₂O₅ were added to one solution, and 7.75 g of silica sol (30% SiO₂) was added to another solution under stirring. The silica sol solution was then poured into the NaAlO₂/Ta₂O₅ solution under stirring for 4 h. The hydrothermal synthesis was carried out at 150 °C for 24 h. After the reaction, the samples were obtained by centrifugating and washing with deionized water for three times, and then dried at 80 °C overnight. The samples were denoted as ZT-*n*, where *n* represents the amount of Ta₂O₅ (g) used. For example, ZT-4 stands for the composite prepared with 4 g of Ta₂O₅. Pure NaTaO₃ was prepared by hydrothermal synthesis at 150 °C for 24 h using the solution prepared by mixing 4 g of Ta₂O₅, 6 g of NaOH and 47.5 g of deionized water.

The stability and composites of sodalite/NaTaO₃ composites were tested by dispersing 0.5 g of powder in 25 ml of 1 M HCl under stirring for 4 h. The resulting products were obtained by centrifugating and washing with deionized water for three times, and dried at 80 °C overnight.

2.2. Characterization

Powder X-ray diffraction (XRD) patterns were recorded on a Rigaku Mini Flex 600 diffractometer with Cu Kα radiation (15 mA and 40 kV) at a scan rate of 2°/min with a step size of 0.02°. Scanning electron microscopy (SEM) images of the samples were taken with a JSM-7001 microscope (JEOL). Elemental mapping images were obtained by using Energy-Dispersive X-ray Spectroscopy (EDX) on a JEOL JSM 7001 microscope, and the element percentages were estimated by using AztecEnergy software (Oxford Instruments). The UV–vis absorption spectra were recorded on UV–vis spectrophotometer (UV-2600, SHIMADZU).

2.3. Photocatalytic degradation

Photocatalytic degradation was performed by degrading methyl orange (MO) in water under UV irradiation. A UV lamp (Pen-ray[®] 254 nm, UVP) was plunged into a quartz immersion well cooled by tap water. NaTaO₃ (50 mg) or sodalite/NaTaO₃ composite (ZT-4, 102.2 mg) was added into 50 ml of 20 mg/l MO aqueous solution. After desired time intervals of irradiation (0–6 h), the suspension was collected and filtered for UV–vis analysis (UVmini-1240, SHIMADZU) by recording the characteristic absorption of methyl orange (464 nm).

3. Results and discussion

0.1–4 g of Ta₂O₅ was added into the zeolite synthesis solution for the co-growth of zeolite/NaTaO₃ composite. Fig. 1a shows the pure sodalite structure prepared without addition of Ta₂O₅, displaying characteristic peaks of sodalite at (110), (211), (310), (222), (330) and (411). With the amount of Ta₂O₅ increases from 0.1 to 4 g (Fig. 1b–e), the sodalite characteristic peaks decrease fast, and the NaTaO₃ characteristic peaks (JCPDS No. 025-0863) [23,24] become stronger, indicating more and more NaTaO₃ presents in the sodalite/NaTaO₃ composites. Fig. 2 shows SEM images of pure sodalite, sodalite/NaTaO₃ composites and pure NaTaO₃. The pure sodalite in Fig. 2a is composed of microsized particles (1–3 μm). The sodalite/NaTaO₃ composites prepared with 0.1–4 g of Ta₂O₅ also show the similar morphology as the pure sodalite (Fig. 2b–e). For the pure NaTaO₃, it shows a typical cubic

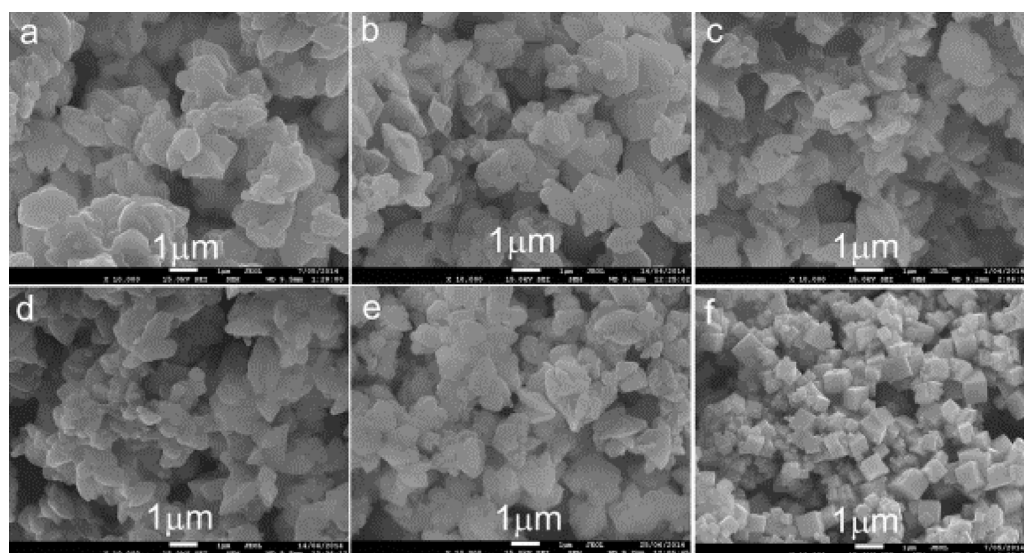


Fig. 2. SEM images of sodalite/NaTaO₃ composite synthesized at 150 °C with different amounts of Ta₂O₅: ZT-0 (a), ZT-0.1 (b), ZT-1 (c), ZT-2 (d), ZT-4 (e), and pure NaTaO₃ (f).

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