



## Facile solvo-combustion synthesis of crystalline $\text{NaTaO}_3$ and its photocatalytic performance for hydrogen production



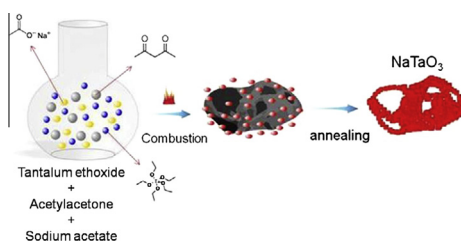
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### HIGHLIGHTS

- The development of an innovative solvo-combustion method for the synthesis of  $\text{NaTaO}_3$ .
- The acetylacetone was used as template and fuel material.
- The  $\text{NaTaO}_3$  was synthesized for first time at low temperature with large surface area.
- The presence of  $\text{Na}_2\text{Ta}_4\text{O}_{11}$  and nanosteps improve the photocatalytic hydrogen production.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Significant attention has been paid on the photocatalytic production of hydrogen from water splitting by using semiconductor materials. The  $\text{NaTaO}_3$  phase has been studied for this purpose due to its high potential to generate charge carriers by absorbing the photons energy. This work reports for the first time the synthesis of  $\text{NaTaO}_3$  powder through an innovative solvo-combustion reaction using the acetyl acetone as template and fuel. It can be mention some advantages of this method such as compounds with high crystallinity, large surface area and specific morphology at nanometer level under low temperature, 180 °C. In order to remove organic material and increase phase crystallinity, this powder was annealed at different temperatures up to 700 °C. All the reaction products were characterized and its photophysical and textural properties were determined. The XRD analysis results showed that crystalline phase of  $\text{NaTaO}_3$  is obtained from the as-prepared material at 180 °C. At higher temperature, the presence of a second phase,  $\text{Na}_2\text{Ta}_4\text{O}_{11}$ , was observed. The TEM micrographs revealed that  $\text{NaTaO}_3$  has hierarchical cubic morphology in the nanometer level with the formation of nanosteps between  $\text{NaTaO}_3$  particles. The materials presented high specific surface area values around 30–120 m<sup>2</sup> g<sup>-1</sup>, which are twice at 180 and 600 °C larger than other reported works. The UV–visible analysis shows a band gap value ( $E_g$ ) around 4.0 eV. It was found that there is a synergy effect between crystallinity of the phase and specific surface area that is the responsible to improve the hydrogen production around three times compared with the results obtained when the pure phase of  $\text{NaTaO}_3$  was employed. Also the presence of the second crystalline phase,  $\text{Na}_2\text{Ta}_4\text{O}_{11}$ , in small concentration and the formation of nanosteps between  $\text{NaTaO}_3$  nanoparticles contributed to enhance of photoactivity of  $\text{NaTaO}_3$ . Finally, when  $\text{RuO}_2$  (1 wt.%) was added as co-catalyst, the efficiency of hydrogen production was increased considerably, reaching around 50 mmol of  $\text{H}_2$  after 5 h of reaction, which is higher than other reports in literature for this material.

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## 1. Introduction

Several researches have been conducted in order to produce alternative clean energy sources. In this way, photocatalytic  $H_2$  production by semiconductor photocatalysts has been extensively studied as a potential means for supplying clean  $H_2$  using solar energy as primary source [1–5]. Various semiconductor materials have been researched as water splitting photocatalysts like tantalates, titanates, niobates and tungstanates due to their unique electron shell structure [3–9]. From these materials, it may be expected that tantalates show better photocatalytic water splitting efficiency since their conduction band presents Ta 5d orbital with lower energy level than Ti 3d orbital in titanate phases [5]. Additionally, the modifications of electronic structure, crystal structure or physicochemical properties could be enhancing the activity of catalytic materials for water splitting reaction [4–10]. Therefore, perovskite-type sodium tantalates have attracted much attention from researchers due to most efficient water splitting catalysts reported, with the band gap up to 4.0 eV [11–13].

In this way, several studies have focused on modification of  $NaTaO_3$ , doped with lanthanum or samarium, using different co-catalysts such as  $RuO_2$ , NiO, Au or Pt [12,13]. Moreover, the synthesis method of  $NaTaO_3$  also plays an important role to improve its photocatalytic performance. Synthesis methods such as solid state reaction, sol–gel, hydrothermal, solvo-thermal, have been employed to adjust the band gap energy, the crystalline phase structure, particle morphology and specific surface area of  $NaTaO_3$  [11,14–16]. Those reports indicate that large surface area can effectively improve the activity of the catalyst because the photocatalytic reaction mainly occurs on the particle surface [17,18].

Taking into consideration the above mentioned, the synthesis of  $NaTaO_3$  via a simple innovative solvo-combustion method is reported for the first time in this work. This material was tested as photocatalyst for hydrogen production under UV-light irradiation.

## 2. Experimental

### 2.1. Synthesis of $NaTaO_3$ via solvo-combustion method

Tantalum (V) ethoxide (99.98% Aldrich) and sodium acetate (98% DEQ) were used as chemical reagents to obtain  $NaTaO_3$  powder. For synthesis 3.96 mmol of ethoxide tantalum and 3.96 mmol of sodium acetate was placed in a flask and dissolved in 0.15 mol of acetyl acetone with 0.26 mol of ethanol. The mixture was stirred and refluxed until the temperature reached 70 °C. Then, 1 mL of nitric acid was added and the flask immediately placed onto a hot plate at 180 °C to rapidly evaporate the solvents. During evaporation, the sample spontaneously ignited, and in a few minutes, a spongy material was obtained. The as-prepared powder was ground and thermally treated in air for 2 h at different temperatures (400, 600 and 700 °C).

### 2.2. Characterization

The crystal structure and phase transformation of the obtained powder were characterized by X-ray powder diffraction (XRD) using a Bruker D8 Advance diffractometer with  $Cu K\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ). The morphology and particle size of the synthesized  $NaTaO_3$  were determined by Scanning Electron Microscope (SEM) in a JEOL 6490 LV and TEM using a FEI Titan G2 80–300 microscope. Phase crystallization and complete removal of organic material were determined by differential thermal analysis and thermogravimetric (DTA/TGA) analysis using a SDTQ600, TA Instruments apparatus at a heating rate of  $10 \text{ }^\circ\text{C min}^{-1}$  under air

flow. The energy band gap ( $E_g$ ) was determined by the Kubelka–Munk function using a UV–vis spectrophotometer (Lambda 35 Perkin Elmer Corporation) coupled with an integrating sphere. Specific surface area ( $S_{BET}$ ) was measured by  $N_2$  physisorption through the BET method using Quantachrome NOVA 2000e equipment.

### 2.3. Water splitting test under UV light irradiation

The water splitting was carried out at low pressure in a batch-type reactor using an inner irradiation cell and argon as carrier. Firstly, 300 mL of distilled water, without the presence of any sacrificial agent were bubbled with argon for 15 min. Then, 300 mg of material was dispersed into water under vigorous stirring. The system was kept at 100 Torr and the water temperature was maintained at 25 °C during the test using a cooling system. Afterwards, the batch-reactor was irradiated with a UV source, Hg lamp with intensity of 400 W, and heterochromatic irradiation ( $\lambda = 200\text{--}800 \text{ nm}$ ). The evolved hydrogen was analyzed using a gas chromatograph with a TCD detector and a packed column Haysep D 100/120. The analysis was carried out in 30 min intervals for 5 h and repeated three times to evaluate the stability of the catalyst. After each photocatalytic reaction, the sample was separated by centrifugation, and then reused for the next experiment.

## 3. Results and discussion

### 3.1. X-ray diffraction

It has been prepared the crystalline phase of  $NaTaO_3$  for first time at low temperature (180 °C) and in a few minutes. Fig. 1 shows the XRD patterns of  $NaTaO_3$ , as-prepared and thermal treated samples annealed at 400, 600 and 700 °C. It is evident that the as-prepared presents high crystallinity and gradually improves it

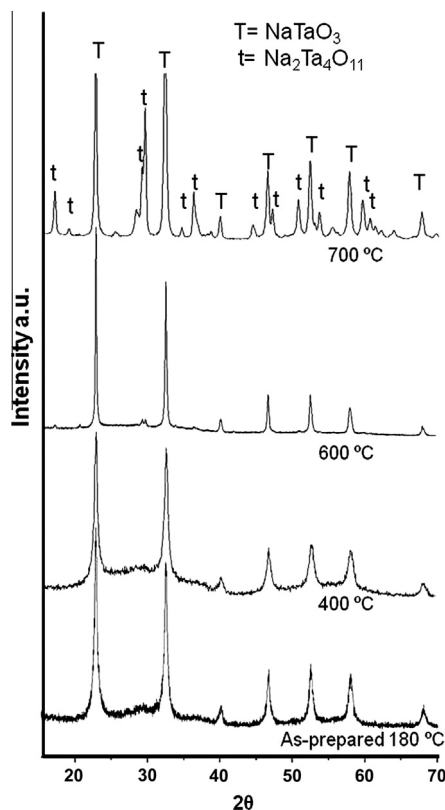


Fig. 1. XRD patterns of  $NaTaO_3$  powders prepared via solvo-combustion method.

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