



Short communication

# Hydrothermal synthesis of $\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$ nano-octahedrons with photocatalytic activity for overall splitting of water



Hun Xue<sup>a,b</sup>, Yanying Chen<sup>a,b</sup>, Nan Ding<sup>b,c</sup>, Qinghua Chen<sup>a,b,\*</sup>, Yongjin Luo<sup>a,b</sup>, Xiping Liu<sup>a,b</sup>, Liren Xiao<sup>b</sup>, Qingrong Qian<sup>a,b,\*</sup>

<sup>a</sup> College of Environmental Science and Engineering, Fujian Normal University, Fuzhou 350007, China

<sup>b</sup> Fujian Key Laboratory of Pollution Control & Resource Reuse, Fuzhou 350007, China

<sup>c</sup> College of Chemistry and Chemical Engineering, Fujian Normal University, Fuzhou 350007, China

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## ABSTRACT

$\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$  nano-octahedrons were successfully prepared via a facile hydrothermal method from  $\text{Sb}_2\text{O}_5$ . The as-prepared  $\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$  was characterized by X-ray diffraction (XRD), UV–vis diffuse reflectance spectra (DRS), scanning electron microscopy (SEM), transmission electron microscopy (TEM), high-resolution transmission electron microscopy (HRTEM) and energy dispersive spectrometer (EDS). Acidity of the precursor mixture had a positive effect on crystallization of  $\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$ .  $\text{RuO}_2$ -loaded  $\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$  showed photocatalytic activity for overall water splitting without adding hole scavengers to produce  $\text{H}_2$  and  $\text{O}_2$  under Xenon lamp irradiation.  $\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$  prepared at low pH exhibited superior photocatalytic activity possibly due to its high degree of crystallization.

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## 1. Introduction

Energy shortage and environmental pollution are the two major problems for the human society. Hydrogen generation by photocatalytic splitting of water is considered an ideal solution to these problems. Since the discovery of photocatalytic splitting of water on  $\text{TiO}_2$  electrodes by Fujishima and Honda in 1972 [1], this new method of using semiconductor and solar energy to produce clean energy has attracted increasing attention.

One type of such semiconductor photocatalysts is composite oxides, which consisted of p-block metal ions ( $\text{In}^{3+}$ ,  $\text{Sn}^{4+}$ ,  $\text{Sb}^{5+}$ ,  $\text{Ge}^{4+}$ , or  $\text{Ga}^{3+}$ ) with  $d^{10}$  configuration such as  $\text{MIn}_2\text{O}_4$  ( $\text{M} = \text{Ca}$  or  $\text{Sr}$ ) [2–4],  $\text{NaNbO}_2$  [5],  $\text{LaInO}_3$  [4],  $\text{Sr}_2\text{SnO}_4$  [6],  $\text{Bi}_2\text{Sn}_2\text{O}_7$  [7],  $\text{M}_2\text{Sb}_2\text{O}_7$  ( $\text{M} = \text{Ca}$  or  $\text{Sr}$ ) [8],  $\text{CaSb}_2\text{O}_6$  [8],  $\text{NaSbO}_3$  [8],  $\text{Gd}_2\text{MSbO}_7$  ( $\text{M} = \text{Fe}$ ,  $\text{In}$ ,  $\text{Y}$ ) [9],  $\text{Zn}_2\text{GeO}_4$  [10–12],  $\text{CuGaO}_2$  [13], and  $\text{MGe}_2\text{O}_4$  ( $\text{M} = \text{Zn}$  or  $\text{Ni}$ ) [14–17]. Their photocatalytic activity may be related to their unique electronic and crystal structures. The highly dispersive conduction band, originating from the hybridization of s and p orbitals of metal elements and the internal fields by the dipole moment inside the distorted  $\text{MO}_6$  ( $\text{M} = \text{In}$ ,  $\text{Sn}$ ,  $\text{Sb}$ , or  $\text{Ga}$ ) octahedra or  $\text{GeO}_4$  tetrahedron, may play a critical role in electron–hole separation upon photoexcitation [6,9,10,13,18]. Composite p-block metal oxides have recently become a hot research area owing to high efficiency as photocatalysts [19–21]. However, some

composite p-block metal oxides are not active for the overall water splitting without hole scavengers, even modified with cocatalysts such as Pt,  $\text{NiO}_x$  or  $\text{RuO}_2$  acting as active sites, which can promote the separation of photogenerated charge carriers and thus suppress their loss via recombination [17,21,22].

There are a number of strontium antimonate composite oxides such as  $\text{Sr}_2\text{Sb}_2\text{O}_7$ ,  $\text{SrSb}_2\text{O}_6$  and  $\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$ , all of which contain p-block metal ion  $\text{Sb}^{5+}$  [18].  $\text{Sr}_2\text{Sb}_2\text{O}_7$  has been reported to be a photocatalyst for water splitting without hole scavengers, and displays photocatalytic activity for the degradation of organic pollutants in gas phase and liquid phase [10,18,23]. However, photocatalytic performance of  $\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$  has never been reported to the best of our knowledge. Here, we synthesized nano-octahedral  $\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$  via a facile hydrothermal method from  $\text{Sb}_2\text{O}_5$  and for the first time, reported the ability to split water without adding hole scavengers under UV light.

## 2. Experimental

## 2.1. Synthesis

$\text{Sr}_{1.36}\text{Sb}_2\text{O}_6$  samples were synthesized by a hydrothermal method from  $\text{Sr}(\text{CH}_3\text{COO})_2 \cdot 0.5\text{H}_2\text{O}$  and  $\text{Sb}_2\text{O}_5$  as precursors. In a typical synthesis, the precursor  $\text{Sr}(\text{CH}_3\text{COO})_2 \cdot 0.5\text{H}_2\text{O}$  (0.88 g, 4.08 mmol) and  $\text{Sb}_2\text{O}_5$  powder (0.97 g, 3.00 mmol) mixed with 70 ml deionized water under vigorous stirring, were adjusted to different pH values (1, 5, 7, or 11) by nitric acid or sodium hydroxide solution. The suspension was placed in a Teflon-lined stainless steel autoclave for hydrothermal reaction at

\* Corresponding authors at: College of Environmental Science and Engineering, Fujian Normal University, Fuzhou 350007, China.

E-mail addresses: [cqhuar@fjnu.edu.cn](mailto:cqhuar@fjnu.edu.cn) (Q. Chen), [qrqian@fjnu.edu.cn](mailto:qrqian@fjnu.edu.cn) (Q. Qian).

180 °C for 48 h. The resulting precipitate was washed with distilled water and absolute ethanol several times, and then dried in the air at 80 °C.

1 wt.% RuO<sub>2</sub>-loaded Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> was prepared by an impregnation method [14,17]. In detail, the as-prepared Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> samples was impregnated with ruthenium carbonyl complex, Ru<sub>3</sub>(CO)<sub>12</sub> in THF, dried at 80 °C, and oxidized in the air at 500 °C for 5 h.

## 2.2. Characterization

X-ray diffraction (XRD) patterns were recorded on a Bruker D8 Advance X-ray diffractometer using CuK $\alpha$  radiation operating at the accelerating voltage and the applied current of 40 kV and 40 mA, respectively. Samples were scanned at a scanning rate of 0.004° s<sup>-1</sup> over the range of 2 $\theta$  = 10°–70°. UV–visible absorption spectra of the powders were collected on a UV–visible spectrophotometer (Cary 500 Scan Spectrophotometers, Varian, USA) using BaSO<sub>4</sub> as a reflectance standard. Field emission scanning electron microscopy (SEM) (JSM-6700F) was used to characterize the morphology of the samples. The transmission electron microscopy (TEM) and high resolution transmission electron microscopy (HRTEM) were measured by JEOL model JEM-2100 instrument at the accelerating voltage of 200 kV. The powder particles were ultrasonically dispersed in ethanol and a drop was dripped on a carbon film coated on a 3 mm diameter fine-mesh copper grid.

## 2.3. Photocatalytic activity measurements

The photocatalytic activity was evaluated in a Pyrex top-irradiation reaction vessel connected to a glass closed gas-circulation system. 100 mg of RuO<sub>2</sub>-loaded Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> powder was suspended in 100 ml distilled water under magnetic stirring. The reaction system was evacuated several times to remove air prior to irradiation. A 300 W xenon lamp which has emission lines in the wavelength range of 350–760 nm was used as the light source. The evolved gases were detected on line by a gas chromatograph (Shimadzu GC-8A) equipped with a thermal conductive detector (TCD) and a 5 Å molecular sieve column. Argon was used as the carrier gas.

## 3. Results and discussion

Fig. 1a shows the XRD patterns of samples obtained at different pH values. All peaks of the products obtained at low pH values (pH = 1 and 5) can be indexed according to JCPDS card 810735 of Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub>, confirming the identity of the products to be Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub>. According to the XRD traces, a mixture of Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> and SrSb<sub>2</sub>O<sub>6</sub> (JCPDS card 820519) is obtained when the pH value of the precursor mixtures increases to 7 and 11. It agrees with the observation that pH value plays a critical role in controlling the composition of the final products in a hydrothermal process. It can also be observed that lower pH value of the precursor mixture contributes to stronger diffraction peaks. This indicates positive effect of acidity on the crystallization of Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub>.

Time-dependent experiments have been carried out for a better understanding of the formation of the Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> nanocrystallites. The unreacted Sb<sub>2</sub>O<sub>5</sub> is presented besides Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> production when the reaction time is less than 2 h. However, pure Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> can be obtained when the hydrothermal time is longer than 6 h. The XRD diffraction peaks of Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> become stronger with increasing hydrothermal time, indicating the enhanced crystallinity (Fig. 1b). Based on the above controlled results, possible chemical reactions involved in the synthesis of Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> particles can be formulated as the following:

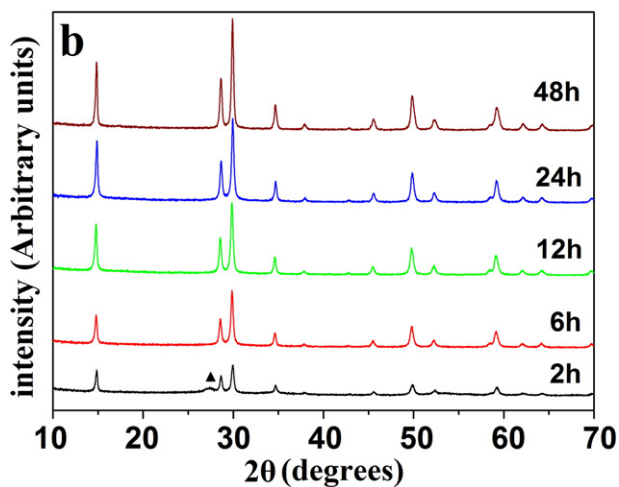
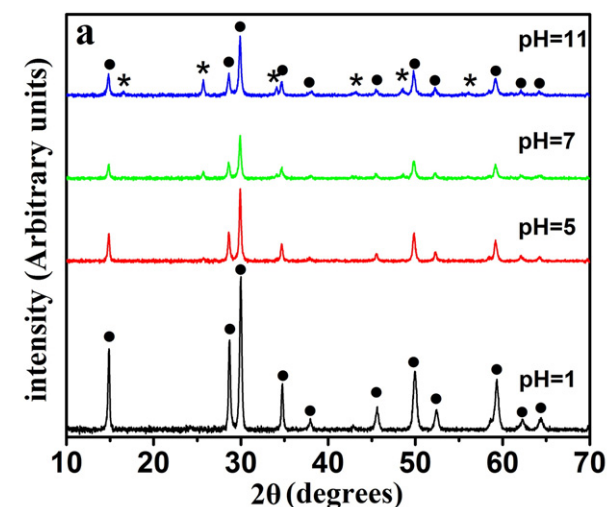
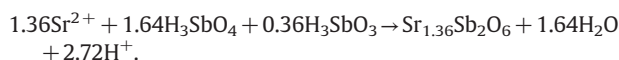
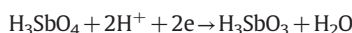
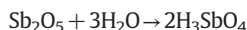


Fig. 1. XRD patterns of the samples prepared (a) at 180 °C for 48 h at different pH values (1, 5, 7, and 11) of the precursor mixtures. (●) Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub>; (\*) SrSb<sub>2</sub>O<sub>6</sub>. (b) at pH 1 for different times (2 h, 6 h, 12 h, 24 h, 48 h). (▲) Sb<sub>2</sub>O<sub>5</sub>.

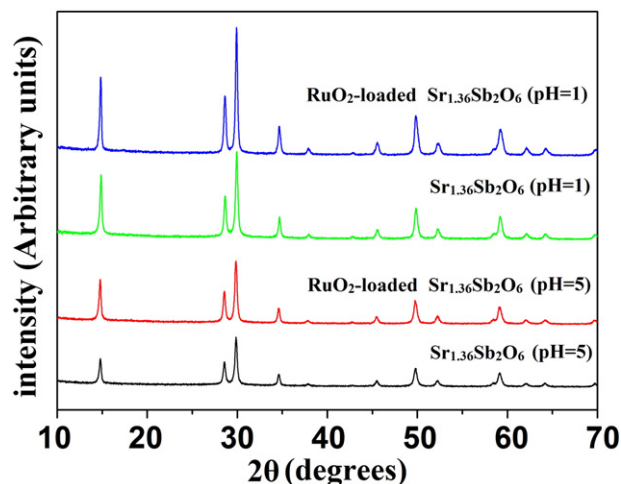


Fig. 2. XRD patterns of Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> and 1 wt.% RuO<sub>2</sub>-loaded Sr<sub>1.36</sub>Sb<sub>2</sub>O<sub>6</sub> prepared at 180 °C for 48 h at pH 1 and 5.

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