

## Photocatalytic Performance of Barium-doped Strontium Tantalate

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### Abstract

*Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub>, a layered perovskite compound, has been reported to possess most excellent photocatalytic properties among the layered perovskite materials. Recently, we have demonstrated that Ba<sub>5</sub>Ta<sub>4</sub>O<sub>15</sub> that was prepared under a mol ratio of Ba: Ta=1:1 has high photocatalytic performance as well as Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub>. In this study, the photocatalyst samples with a mol ratio of Sr: Ba: Ta = (1-x): x: 1 were prepared. The maximum photocatalytic performance was obtained for x= 0.2, which is three times as high as that of undoped Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub>.*

**Keywords :** photocatalyst, water-splitting reaction, tantalate, perovskite

### 1. Introduction

Hydrogen energy is expected as alternative future energy to fossil fuel. The cyclic and clean photocatalytic reaction to split water into H<sub>2</sub> and O<sub>2</sub> attracts attention as one of the promising methods to produce hydrogen. A series of mixed oxides containing the Ta<sup>5+</sup> ion is known to show a high photocatalytic performance in this decomposition reaction [1-4]. In particular, La<sup>3+</sup> and alkali earth ions-doped NaTaO<sub>3</sub> exhibited excellent activity [5-7]. On the other hand, an alkali-earth tantalate with layered perovskite structure, particularly Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub>, has excellent photocatalytic property [8]. In our previous work [9], Ba<sub>5</sub>Ta<sub>4</sub>O<sub>15</sub> with a small amount of tetragonal BaTa<sub>2</sub>O<sub>6</sub>, which was prepared by a mol ratio of Ba: Ta = 1: 1, has been proved to have high photocatalytic performance as well as Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub>. In this study, several amount of Ba was doped into Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub>, and the effect of Ba substitution for the Sr site on the photocatalytic activity was investigated.

### 2. Experimental and Results

The photocatalyst samples were prepared by the polymerized complex method. The compositional ratio of Sr: Ba: Ta was set to (1-x): x: 1, where x = 0, 0.1, 0.15, 0.2, 0.4, 0.6, 0.8 and 1. The prepared polymerized complex resins were thermally calcinated at 1273K for 24 hours after calcinations at 1073K for 2 hours.

Constituent phases of the obtained photocatalyst samples were identified by a conventional powder x-ray diffraction

(XRD). The mass fraction for each phases and the specific surface area of the samples were analyzed by Rietveld method (RIETAN-2000) [10] and BET method, respectively.

NiO (0.2 wt. %) was used as a co-catalyst and these supported photocatalysts were prepared by a conventional impregnation method. The photocatalytic reaction was carried out in an inner irradiation type quartz reactor connected to closed gas circulation system with a dead volume of 450 ml. The photocatalyst samples of 1.0 g were suspended in well degassed water in the reactor. A high-pressure mercury lamp was used as ultraviolet light source. The produced gases were analyzed by gas chromatography.

Fig. 1 shows the XRD patterns of photocatalyst samples with different Ba content. The samples of  $x \leq 0.15$  consist of the single phase of Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub>. A little Ba<sub>5</sub>Ta<sub>4</sub>O<sub>15</sub> phase appears as the second phase at  $x = 0.2$ . Moreover, tetragonal BaTa<sub>2</sub>O<sub>6</sub> appears at  $x = 0.4$ , then three phases coexist. Both samples of  $x = 0.8$  and 1.0 were identified as a mixture of Ba<sub>5</sub>Ta<sub>4</sub>O<sub>15</sub> as the main phase and tetragonal BaTa<sub>2</sub>O<sub>6</sub> as the sub phase. Each peak of the diffraction patterns shifts to the lower angle side as an amount of Ba increases.

In figure 2 the photocatalytic activity is illustrated as a function of Ba content  $x$ . Each photocatalytic activity of H<sub>2</sub> and O<sub>2</sub> increases with a progress of Ba substitution for Sr sites of Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub> and reaches to a maximum at  $x = 0.2$ . The maximum values are 6400 mol/h and 3200 mol/h for H<sub>2</sub> and O<sub>2</sub> (stoichiometric composition), respectively, which are three times as high as those of undoped Sr<sub>2</sub>Ta<sub>2</sub>O<sub>7</sub>. The maximum activity obtained at solid solubility limit of Ba, or under existence of a little second phase of Ba<sub>5</sub>Ta<sub>4</sub>O<sub>15</sub>, which suggests that the lattice strain introduced by Ba substitution

improves photocatalytic activity. Each activity drops down with a further increase of Ba doping and becomes minimum at  $x = 0.4$  where the third phase of tetragonal  $\text{BaTa}_2\text{O}_6$  appears. The each minimum value is rather lower than that of  $\text{Sr}_2\text{Ta}_2\text{O}_7$  at  $x = 0$ . As  $x$  increases, each activity improves again with a decrease of  $\text{Sr}_2\text{Ta}_2\text{O}_7$  phase and reaches to the secondary peak (3400  $\mu\text{mol/h}$  for  $\text{H}_2$  and 1700  $\mu\text{mol/h}$  for  $\text{O}_2$ ) at  $x = 0.8$  where two phases of  $\text{Ba}_5\text{Ta}_4\text{O}_{15}$  and tetragonal  $\text{BaTa}_2\text{O}_6$  coexist.

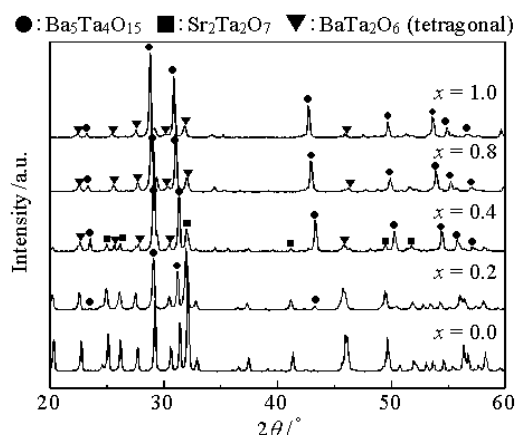


Fig. 1. XRD patterns of samples of Sr: Ba: Ta = (1-x): x: 1 calcinated for 24 hours at 1273K.

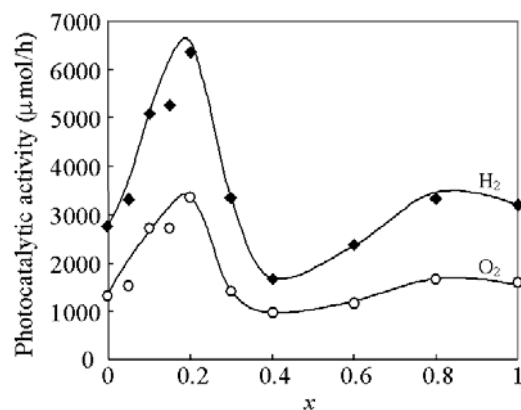


Fig. 2. Variation of photocatalytic activity with Ba content,  $x$ ; Sr: Ba: Ta = (1-x): x: 1.

Specific surface area of undoped  $\text{Sr}_2\text{Ta}_2\text{O}_7$  ( $x = 0$ ) was 9  $\text{m}^2/\text{g}$ , which greatly exceeded others of 3  $\text{m}^2/\text{g}$ . This result indicates that the specific surface areas of the photocatalysts

prepared in this work did not almost affect their  $\text{H}_2\text{O}$ -decompositional activity as compared to other factors such as a lattice strain.

### 3. Summary

The results of this study are summarized as follows.

- (1) The maximum activity is obtained at solid solubility limit of Ba, or under existence of a little second phase of  $\text{Ba}_5\text{Ta}_4\text{O}_{15}$  at  $x = 0.2$ ; Sr: Ba: Ta = (1-x): x: 1.
- (2) A photocatalytic activity drops down with a further increase of Ba doping and becomes minimum at  $x = 0.4$  where the third phase of tetragonal  $\text{BaTa}_2\text{O}_6$  appears.
- (3) The increase of photocatalytic activity may be caused by crystal distortion due to doping of Ba ion. The specific surface areas of the photocatalysts prepared in present work did not almost affect their  $\text{H}_2\text{O}$ -decompositional activity.

The maximum photocatalytic activity achieved in this study is in the highest level in a series of tantalum photocatalysts reported so far.

### 4. References

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