

## Research Article

# Low-Temperature Synthesis of $\alpha$ -BiTaO<sub>4</sub> Photocatalyst by the Flux Method

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Low-temperature phase BiTaO<sub>4</sub> ( $\alpha$ -BiTaO<sub>4</sub>) was successfully synthesized by the flux method using Bi<sub>2</sub>O<sub>3</sub>-B<sub>2</sub>O<sub>3</sub> as the flux. According to previous reports,  $\alpha$ -BiTaO<sub>4</sub> has been mostly synthesized via a solid state reaction, which requires heating at 900°C for more than 48 h. In this study,  $\alpha$ -BiTaO<sub>4</sub> was successfully synthesized in 4 h at 750°C using the flux method. The impact of varying reaction conditions on the products was analyzed by X-ray diffraction and a scanning electron microscopy analysis. The grain size of  $\alpha$ -BiTaO<sub>4</sub> was dependent on reaction conditions such as the reaction temperature and solute concentration. The photocatalytic activity of the obtained  $\alpha$ -BiTaO<sub>4</sub> was evaluated for the degradation of phenol. It was found that flux-synthesized  $\alpha$ -BiTaO<sub>4</sub> exhibited a higher photocatalytic activity than  $\alpha$ -BiTaO<sub>4</sub> synthesized using the solid state method.

## 1. Introduction

Since the demonstration of water splitting using a TiO<sub>2</sub> electrode under ultraviolet light irradiation in 1972 [1], semiconductor photocatalysis has been actively studied. Among different photocatalysts, TiO<sub>2</sub> has been studied extensively and is currently the most common photocatalyst because it shows high photocatalytic activity under ultraviolet light, is chemically stable, is harmless to the human body, and has a low cost. However, the band gap of TiO<sub>2</sub> is too wide to respond to visible light. Thus, to exploit sunlight, the development of new photocatalysts that utilize visible light is needed.

It has been known that metal oxides containing bismuth, such as Bi<sub>2</sub>WO<sub>6</sub> [2], Bi<sub>2</sub>MoO<sub>6</sub> [3], BiVO<sub>4</sub> [4], and BiTaO<sub>4</sub> [5], are good visible light driven photocatalysts. The visible light response of these materials could be attributed to the presence of the Bi<sup>3+</sup> lone-pair electrons. It was clarified that the band gap narrowing was due to the contribution of Bi 6s orbitals to the valence band formation [5, 6]. In addition, it has been found that many metal oxides consisting of TaO<sub>6</sub> octahedral units can degrade organic contaminants or decompose water under ultraviolet light or visible light

irradiation [7, 8]. Therefore, we focused our attention on BiTaO<sub>4</sub>.

BiTaO<sub>4</sub> can be crystallized in two different forms, the triclinic (high-temperature phase,  $\beta$  type) and orthorhombic (low-temperature phase,  $\alpha$  type) systems. The low-temperature orthorhombic form transforms to the high-temperature triclinic form at 1150°C [9]. Generally,  $\beta$ -BiTaO<sub>4</sub> has been synthesized via a solid state reaction. Moreover, there are many reports of the synthesis of  $\beta$ -BiTaO<sub>4</sub> via liquid phase reactions such as the coprecipitation [10], polymeric precursor [11], and solution methods [12]. The photocatalytic activity of  $\beta$ -BiTaO<sub>4</sub> synthesized by the solid state reaction [5, 13, 14] and the solution method [12] have also been reported. On the other hand,  $\alpha$ -BiTaO<sub>4</sub> has been mostly synthesized via a solid state reaction. However, to avoid the formation of impurity phases, the reaction must be heated at 900°C for more than 48 h [15]. Unless this condition is fulfilled, the product is neither highly crystalline nor of a single phase [15]. Therefore, it is difficult to obtain high-quality  $\alpha$ -BiTaO<sub>4</sub> by this solid state reaction. Only one other method for the synthesis of  $\alpha$ -BiTaO<sub>4</sub>—a hydrothermal method—has been reported [16]. However, this method also requires a very long reaction time and

the products are not powders, but crystals. Moreover, the use of an Au tube is necessary because  $\text{KHF}_2$  serves as the solvent. For these reasons, the hydrothermal method is impractical. Furthermore, there have been no reports on the photocatalytic activity of  $\alpha\text{-BiTaO}_4$ . However, in the case of  $\text{BiTa}_{1-x}\text{Nb}_x\text{O}_4$  ( $0 \leq x \leq 1$ ), which is a solid solution of  $\text{BiTaO}_4$  and isostructural  $\text{BiNbO}_4$ , the low-temperature phase shows higher photocatalytic activity than the high-temperature phase [7, 13]. Therefore, the low-temperature phase of  $\text{BiTaO}_4$  is expected to show high photocatalytic activity.

Based on these facts, the focus of this work was on  $\alpha\text{-BiTaO}_4$ . The flux method was selected for the synthesis of  $\alpha\text{-BiTaO}_4$  because this method involves a low-temperature reaction and a short reaction time. In addition,  $\text{Bi}_2\text{O}_3\text{-B}_2\text{O}_3$  was selected as the flux because it is considered as difficult to contaminate the product as an impurity and its eutectic temperature is comparatively low. Furthermore, the photocatalytic activity of  $\alpha\text{-BiTaO}_4$  synthesized by the flux method and the known solid state reaction was measured.

## 2. Experimental

**2.1. Synthesis of  $\alpha\text{-BiTaO}_4$ .**  $\alpha\text{-BiTaO}_4$  was synthesized by the flux method with  $\text{Bi}_2\text{O}_3\text{-B}_2\text{O}_3$  as the flux. Reagent-grade  $\text{B}_2\text{O}_3$  (Wako Pure Chemical Industries, Ltd.),  $\text{Bi}_2\text{O}_3$ , and  $\text{Ta}_2\text{O}_5$  (Kojundo Chemical Lab Co., Ltd.) were used for the synthesis of  $\alpha\text{-BiTaO}_4$ . The molar ratios of the raw materials are given in Table 1. The total amount of the reagents was 3 g for each condition. The mixture was placed in an alumina crucible, reacted for 4 h, and then cooled down to room temperature. Reaction temperatures are also provided in Table 1. After the reaction, the products were separated from the remaining flux using nitric acid ( $2.5 \text{ mol L}^{-1}$ ) in an ultrasonic bath for 5 h.

As a reference sample,  $\alpha\text{-BiTaO}_4$  was synthesized via a solid state reaction.  $\text{Bi}_2\text{O}_3$  and  $\text{Ta}_2\text{O}_5$  were weighed out in an appropriate stoichiometric ratio and ground for several minutes using an agate mortar and pestle. During the grinding, a small quantity of acetone was added to uniformly mix the materials. The mixture was then placed in an alumina crucible and preheated in air at  $700^\circ\text{C}$  for 48 h, and finally heated at  $900^\circ\text{C}$  for 48 h.

**2.2. Characterization.** The products were characterized by powder X-ray diffraction (XRD) analysis using a Rigaku RINT-2200 with  $\text{Cu K}\alpha$  radiation ( $\lambda = 1.54178 \text{ \AA}$ ) and operated at 40 kV and 40 mA with a scan speed of  $2^\circ \text{ min}^{-1}$  and a step width of  $0.01^\circ$ . The morphology and grain size of the products were investigated by scanning electron microscopy (SEM) using a JEOL JSM-6390LA operating at 10 kV.

**2.3. Evaluation of Photocatalytic Activity.** The photocatalytic activity of  $\alpha\text{-BiTaO}_4$  was evaluated on the basis of the photodegradation of phenol. As-prepared  $\alpha\text{-BiTaO}_4$  (0.1 g) and an  $\text{H}_2\text{O}_2$  aqueous solution (0.5 mL, 30 wt%) were added to a phenol solution (100 mL,  $50 \text{ mg L}^{-1}$ ). A 300 W Xe lamp

TABLE 1: Synthesis conditions of  $\alpha\text{-BiTaO}_4$  by the flux method.

Run no.	$\text{Bi}_2\text{O}_3 : \text{B}_2\text{O}_3$ (molar ratio)	Flux <sup>a</sup> : $\text{Ta}_2\text{O}_5$ (molar ratio)	Reaction temperature/ $^\circ\text{C}$
1	38 : 62	30 : 7	750
2	38 : 62	30 : 7	800
3	38 : 62	30 : 7	900
4	38 : 62	30 : 0.5	750
5	38 : 62	30 : 2	750
6	38 : 62	30 : 4	750
7	38 : 62	30 : 9	750

<sup>a</sup>Flux =  $\text{Bi}_2\text{O}_3 + \text{B}_2\text{O}_3$ .

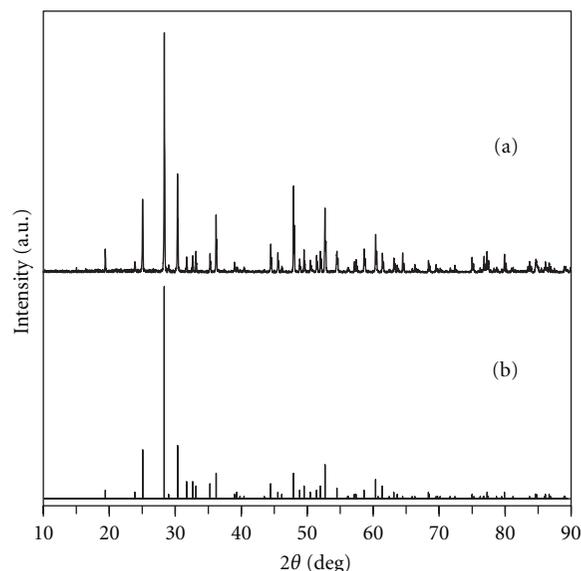


FIGURE 1: XRD pattern of (a) the product synthesized by the flux method at  $900^\circ\text{C}$  for 4 h (run no. 3) and (b)  $\alpha\text{-BiTaO}_4$  JCPDS.

( $\lambda > 300 \text{ nm}$ ) was used as the light source. Before illumination, the solution was magnetically stirred for 60 min in the dark to reach adsorption-desorption equilibrium between the photocatalyst and phenol. At every 30 min intervals, 5 mL of the suspension was sampled and centrifuged to remove the photocatalyst for analysis of phenol concentration. The adsorption spectrum of the centrifuged solution was recorded on an ultraviolet-visible spectrophotometer using a Shimadzu UV mini 1240 at a wavelength of 270 nm.

## 3. Results and Discussion

**3.1. Comparison of Products Obtained from Flux Method and Solid State Reaction.** Figure 1 shows the XRD pattern of the product synthesized by the flux method at  $900^\circ\text{C}$  for 4 h. All diffraction peaks of the product were indexed to  $\alpha\text{-BiTaO}_4$  (JCPDS 01-072-7514), indicating that the product was a single phase of  $\alpha\text{-BiTaO}_4$ . Therefore,  $\alpha\text{-BiTaO}_4$  can be synthesized by the flux method with a significantly shorter reaction time than the solid state reaction.

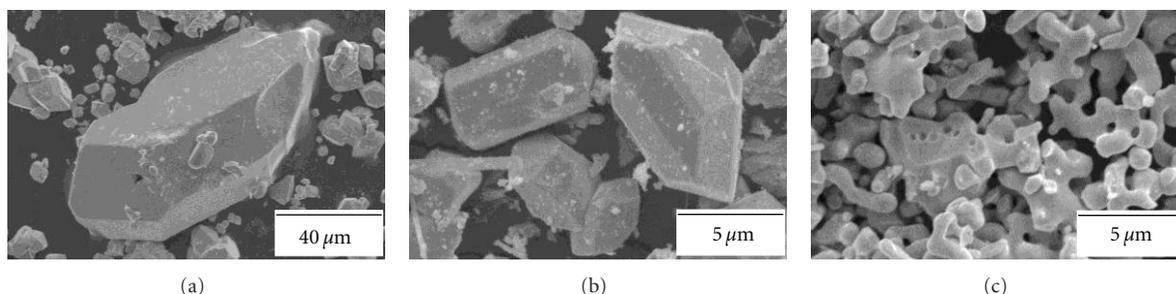


FIGURE 2: SEM images of  $\alpha$ -BiTaO<sub>4</sub> synthesized ((a), (b)) by the flux method at 900°C for 4 h (run no. 3) and (c) by the solid state reaction.

The morphology of the product was observed by SEM and compared with that of  $\alpha$ -BiTaO<sub>4</sub> synthesized by the solid state reaction. As can be seen from Figures 2(a) and 2(b), flux-synthesized  $\alpha$ -BiTaO<sub>4</sub> was a euhedral crystal. On the other hand, as shown in Figure 2(c), solid state-synthesized  $\alpha$ -BiTaO<sub>4</sub> showed necking between particles. Therefore, it was concluded that flux-synthesized  $\alpha$ -BiTaO<sub>4</sub> had a higher crystallinity, which is advantageous for photocatalysis. However, the grain size of the product obtained using the flux method was 5–90  $\mu\text{m}$ , whereas that of the material prepared via the solid state reaction was 2–5  $\mu\text{m}$ . Therefore, in terms of the specific surface area, solid state-synthesized  $\alpha$ -BiTaO<sub>4</sub> had properties that are more advantageous for photocatalysis. For this reason, it was necessary to examine reaction conditions in order to obtain small crystals. It was determined that the large crystals were formed because the solubility of the solute in the flux is high, and the transport rate of the solute is fast. Thus, reaction temperature, which directly influences both of these features of the reaction, was examined.

### 3.2. Influence of Reaction Temperature on Product Formation.

Figure 3 shows the XRD patterns of the products synthesized at 750°C and 800°C for 4 h. As can be seen from Figures 3(a) and 3(b), the products were a single phase of  $\alpha$ -BiTaO<sub>4</sub> when prepared at both temperatures. This result indicates that  $\alpha$ -BiTaO<sub>4</sub> can be synthesized using the flux method at a reaction temperature lower than that of the conventional solid state reaction, which requires heating at 900°C.

Figure 4 shows the SEM images of the products shown in Figure 3. As can be seen in Figures 4(a) and 4(b), when reaction temperature was 750°C, grain size was 1–30  $\mu\text{m}$ . However, Figures 4(c) and 4(d) show that when reaction temperature was 800°C, grain size increased to 2–60  $\mu\text{m}$ . As mentioned in Section 3.1, the grain size at reaction temperature 900°C was 5–90  $\mu\text{m}$ . Therefore, these results indicate that grain size decreases with reaction temperature. The cause is thought to be decreased in the solubility of the solute in the flux due to the lowering of reaction temperature. And it is also the cause that the transport rate of the solute reduces because the viscosity of the solution increases as the reaction temperature decreases.

However, even when reaction temperature was as low as 750°C, grain size was still too large (1–30  $\mu\text{m}$ ) compared with that of solid state-synthesized  $\alpha$ -BiTaO<sub>4</sub> (2–5  $\mu\text{m}$ ). Yet,

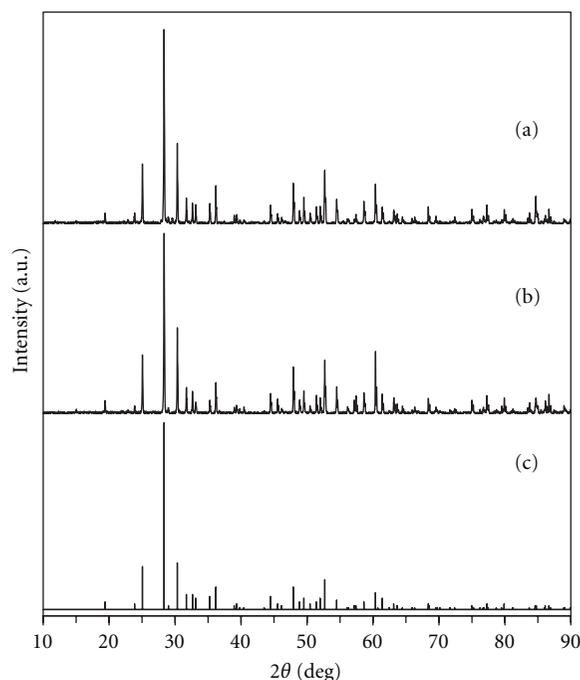


FIGURE 3: XRD patterns of the products synthesized by the flux method at (a) 750°C (run no. 1), (b) 800°C (run no. 2), and (c)  $\alpha$ -BiTaO<sub>4</sub> JCPDS.

when the mix ratio of the flux is Bi<sub>2</sub>O<sub>3</sub> : B<sub>2</sub>O<sub>3</sub> = 38 : 62, the eutectic temperature is 722°C, and thus, the synthesis cannot be performed at a lower temperature. Therefore, 750°C was selected as the optimum reaction temperature. As a result, it was necessary to examine the other reaction conditions in order to find a way to produce smaller crystals. Here, the trend of the broad grain size distribution for flux-synthesized  $\alpha$ -BiTaO<sub>4</sub> was the focus of attention. The broad range of grain sizes is likely owing to the high viscosity of B<sub>2</sub>O<sub>3</sub>, which is a component of the flux. Because the viscosity of B<sub>2</sub>O<sub>3</sub> is high, the diffusion of Ta<sub>2</sub>O<sub>5</sub>, which is the solute, does not occur uniformly in the reaction system and a concentration gradient of dissolved Ta<sub>2</sub>O<sub>5</sub> in the flux occurs. Therefore, solute concentration, which is thought to affect the grain size of the product, was investigated.

### 3.3. Influence of Solute Concentration on Product Formation.

By varying the amount of Ta<sub>2</sub>O<sub>5</sub>, the influence of solute

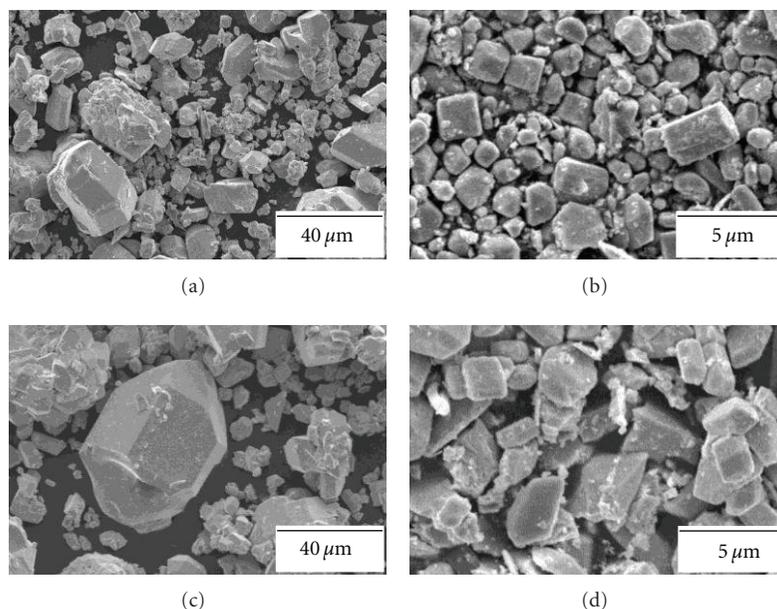


FIGURE 4: SEM images of  $\alpha$ -BiTaO<sub>4</sub> synthesized by the flux method at ((a), (b)) 750°C (run no. 1) and ((c), (d)) 800°C (run no. 2).

concentration on the products was investigated. Figure 5 shows the XRD patterns of the products synthesized at 750°C for 4 h in various solute concentrations. As can be seen from Figure 5(a), when the Flux:Ta<sub>2</sub>O<sub>5</sub> ratio was 30:0.5, the products were  $\alpha$ -BiTaO<sub>4</sub> and Bi<sub>4</sub>Ta<sub>2</sub>O<sub>11</sub>. This result indicates that when the concentration of Ta<sub>2</sub>O<sub>5</sub> is too low, a compound with two equivalents of Bi is formed owing to the presence of excess Bi relative to Ta. On the other hand, when the Flux:Ta<sub>2</sub>O<sub>5</sub> ratio was 30:9 (Figure 5(d)), the products were  $\alpha$ -BiTaO<sub>4</sub> and Ta<sub>2</sub>O<sub>5</sub>, which indicates that when the concentration of Ta<sub>2</sub>O<sub>5</sub> is too high, Ta<sub>2</sub>O<sub>5</sub> remains in the system. However, when the Flux:Ta<sub>2</sub>O<sub>5</sub> ratio was appropriate, such as 30:2 (Figure 5(b)) or 30:4 (Figure 5(c)), the product was a single phase of  $\alpha$ -BiTaO<sub>4</sub>. Figure 6 shows the SEM images of the products. These results indicate that when the Flux:Ta<sub>2</sub>O<sub>5</sub> ratio was 30:2 or 30:4, the grain size of  $\alpha$ -BiTaO<sub>4</sub> was 1–10  $\mu$ m, which is smaller than that obtained with the original ratio of Flux:Ta<sub>2</sub>O<sub>5</sub> = 30:7 (1–30  $\mu$ m, Section 3.2).

On the basis of these results, the optimum synthesis conditions for preparing the smallest grain size of  $\alpha$ -BiTaO<sub>4</sub> are given as follows: the mix ratio of the flux is Bi<sub>2</sub>O<sub>3</sub>:B<sub>2</sub>O<sub>3</sub> = 38:62, the mix ratio of the flux and solute is Flux:Ta<sub>2</sub>O<sub>5</sub> = 30:2 or 30:4, reaction temperature is 750°C, and reaction time is 4 h. Flux-synthesized  $\alpha$ -BiTaO<sub>4</sub> prepared under these conditions was considered to be the best sample for evaluation as a photocatalyst.

**3.4. Photocatalytic Activity.** The photocatalytic activity of flux-synthesized  $\alpha$ -BiTaO<sub>4</sub> with a small grain size (run no. 6) and solid state-synthesized  $\alpha$ -BiTaO<sub>4</sub> was evaluated for the degradation of phenol. Figure 7 depicts the photodegradation efficiencies of phenol as a function of irradiation time over the different  $\alpha$ -BiTaO<sub>4</sub> samples. As

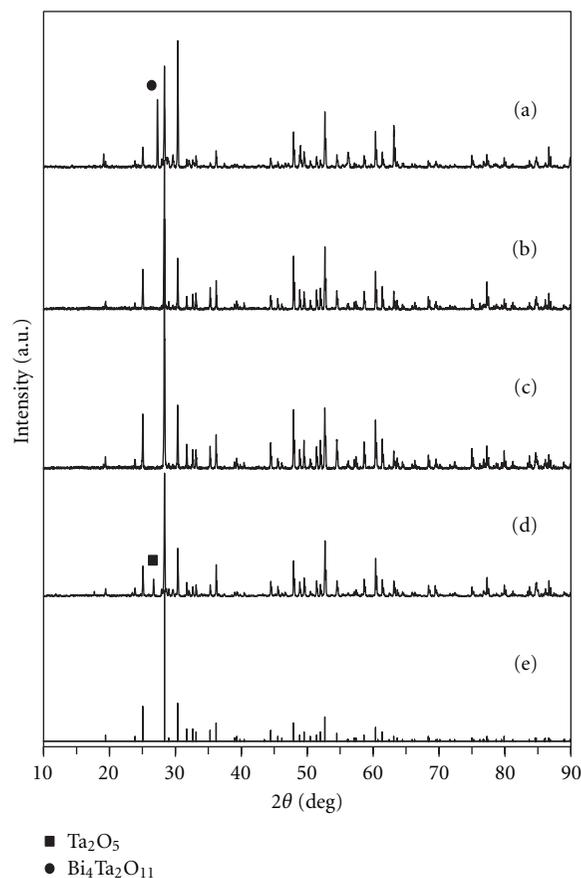


FIGURE 5: XRD patterns of the products synthesized by the flux method with a mix ratio of Flux:Ta<sub>2</sub>O<sub>5</sub> of (a) 30:0.5 (run no. 4), (b) 30:2 (run no. 5), (c) 30:4 (run no. 6), (d) 30:9 (run no. 7), and (e)  $\alpha$ -BiTaO<sub>4</sub> JCPDS.

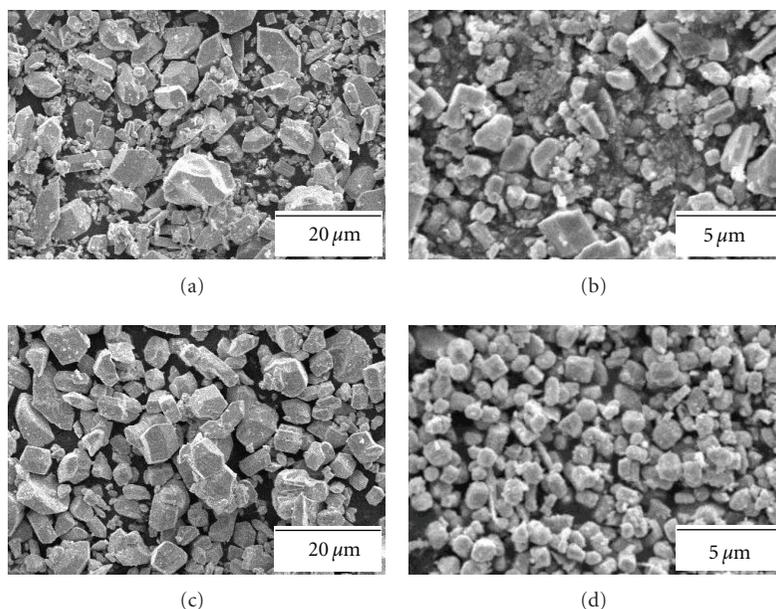


FIGURE 6: SEM images of  $\alpha$ -BiTaO<sub>4</sub> synthesized by the flux method with a mix ratio of Flux : Ta<sub>2</sub>O<sub>5</sub> of ((a), (b)) 30 : 2 (run no. 5) and ((c), (d)) 30 : 4 (run no. 6).

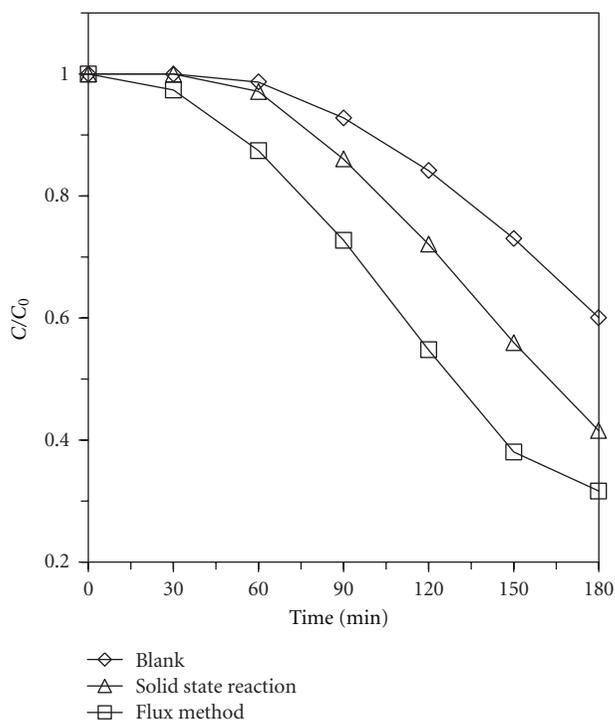


FIGURE 7: Photodegradation efficiencies of phenol as a function of irradiation time.

shown in Figure 7, both  $\alpha$ -BiTaO<sub>4</sub> samples showed a higher degradation efficiency of phenol than a blank solution, that is, the direct photolysis of phenol in the presence of H<sub>2</sub>O<sub>2</sub> without a catalyst. This result indicates that  $\alpha$ -BiTaO<sub>4</sub> can decompose phenol under light irradiation.

Furthermore, flux-synthesized  $\alpha$ -BiTaO<sub>4</sub> showed a higher degradation efficiency than solid state-synthesized  $\alpha$ -BiTaO<sub>4</sub>. This difference is probably due to the higher crystallinity of flux-synthesized  $\alpha$ -BiTaO<sub>4</sub>. Thus, we succeeded in improving the photocatalytic activity of  $\alpha$ -BiTaO<sub>4</sub> using the flux method as a synthesis technique.

#### 4. Conclusions

$\alpha$ -BiTaO<sub>4</sub> was successfully synthesized by the flux method at a lower reaction temperature and in a shorter reaction time than the conventional solid state reaction. The solid state reaction requires heating at 900°C for more than 48 h, whereas the flux method can be used to synthesize  $\alpha$ -BiTaO<sub>4</sub> at 750°C in 4 h. In addition, the grain size of  $\alpha$ -BiTaO<sub>4</sub> can be controlled by adjusting reaction temperature and solute concentration. The synthesis conditions that provide  $\alpha$ -BiTaO<sub>4</sub> with the smallest grain size were found to be as follows: a mix ratio of flux: Bi<sub>2</sub>O<sub>3</sub>:B<sub>2</sub>O<sub>3</sub> of 38:62, a mix ratio of flux: solute (Ta<sub>2</sub>O<sub>5</sub>) of 30:2 or 30:4, reaction temperature 750°C, and reaction time 4 h. Furthermore,  $\alpha$ -BiTaO<sub>4</sub> prepared under these conditions exhibited higher photocatalytic activity than that synthesized using the solid state method.

#### References

- [1] A. Fujishima and K. Honda, "Electrochemical photolysis of water at a semiconductor electrode," *Nature*, vol. 238, no. 5358, pp. 37–38, 1972.
- [2] L. S. Zhang, W. Z. Wang, L. Zhou, and H. L. Xu, "Bi<sub>2</sub>WO<sub>6</sub> Nano- and microstructures: shape control and associated visible-light-driven photocatalytic activities," *Small*, vol. 3, no. 9, pp. 1618–1625, 2007.

- [3] Y. Shimodaira, H. Kato, H. Kobayashi, and A. Kudo, "Photophysical properties and photocatalytic activities of bismuth molybdates under visible light irradiation," *Journal of Physical Chemistry B*, vol. 110, no. 36, pp. 17790–17797, 2006.
- [4] L. Zhou, W. Z. Wang, S. W. Liu, L. S. Zhang, H. L. Xu, and W. Zhu, "A sonochemical route to visible-light-driven high-activity BiVO<sub>4</sub> photocatalyst," *Journal of Molecular Catalysis A*, vol. 252, no. 1-2, pp. 120–124, 2006.
- [5] R. Shi, J. Lin, Y. J. Wang, J. Xu, and Y. F. Zhu, "Visible-light photocatalytic degradation of BiTaO<sub>4</sub> photocatalyst and mechanism of photocorrosion suppression," *Journal of Physical Chemistry C*, vol. 114, no. 14, pp. 6472–6477, 2010.
- [6] L. Wang, W. Z. Wang, M. Shang et al., "Visible light responsive bismuth niobate photocatalyst: enhanced contaminant degradation and hydrogen generation," *Journal of Materials Chemistry*, vol. 20, no. 38, pp. 8405–8410, 2010.
- [7] Z. G. Zou, J. H. Ye, K. Sayama, and H. Arakawa, "Photocatalytic and photophysical properties of a novel series of solid photocatalysts, BiTa<sub>1-x</sub>Nb<sub>x</sub>O<sub>4</sub> (0 ≤ x ≤ 1)," *Chemical Physics Letters*, vol. 343, no. 3-4, pp. 303–308, 2001.
- [8] Z. G. Zou, J. H. Ye, and H. Arakawa, "Optical and structural properties of the BiTa<sub>1-x</sub>Nb<sub>x</sub>O<sub>4</sub> (0 ≤ x ≤ 1) compounds," *Solid State Communications*, vol. 119, no. 7, pp. 471–475, 2001.
- [9] R. S. Roth and J. L. Waring, "Synthesis and stability of bismutotantalite, stibiotantalite and chemically similar ABO<sub>4</sub> compounds," *the American Mineralogist Journal*, vol. 48, no. 11-12, pp. 1348–1356, 1963.
- [10] H. Muthurajan, U. N. Gupta, B. Rituraj et al., "A coprecipitation technique to prepare BiTaO<sub>4</sub> powders," *Materials Letters*, vol. 62, no. 3, pp. 501–503, 2008.
- [11] C. G. Almeida, H. M. C. Andrade, A. J. S. Mascarenhas, and L. A. Silva, "Synthesis of nanosized β-BiTaO<sub>4</sub> by the polymeric precursor method," *Materials Letters*, vol. 64, no. 9, pp. 1088–1090, 2010.
- [12] R. Ullah, H. Q. Sun, H. M. Ang, M. O. Tade, and S. B. Wang, "Visible light photocatalytic degradation of organics on nanoparticles of bi-metallic oxides," *Separation and Purification Technology*, vol. 89, no. 6, pp. 98–106, 2012.
- [13] Z. G. Zou, J. H. Ye, and H. Arakawa, "Photocatalytic water splitting into H<sub>2</sub> and/or O<sub>2</sub> under UV and visible light irradiation with a semiconductor photocatalyst," *International Journal of Hydrogen Energy*, vol. 28, no. 6, pp. 663–669, 2003.
- [14] H. J. Zhang, G. Chen, X. Li, and Q. Wang, "Electronic structure and water splitting under visible light irradiation of BiTa<sub>1-x</sub>Cu<sub>x</sub>O<sub>4</sub> (x = 0.00 – 0.04) photocatalysts," *International Journal of Hydrogen Energy*, vol. 34, no. 9, pp. 3631–3638, 2009.
- [15] C. Y. Lee, R. Macquart, Q. Zhou, and B. J. Kennedy, "Structural and spectroscopic studies of BiTa<sub>1-x</sub>Nb<sub>x</sub>O<sub>4</sub>," *Journal of Solid State Chemistry*, vol. 174, no. 2, pp. 310–318, 2003.
- [16] V. I. Popolitov, "Crystallization of bismuth orthoniobate and orthotantalate under hydrothermal conditions," *Kristallografiya*, vol. 33, no. 1, pp. 222–224, 1988.



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