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Size and Morphology of Scorodite Particles Synthesized Using Ultrasound Irradiation
超音波照射を用いて合成したスコロダイト粒子の粒径およびその形態

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1. Introduction

Wastewater contaiminated with arsenic, such as arsenic acid mine drainage, has been treated through the coprecipitation method using iron hydroxide generated by neutralization of the treatment solution. However, iron hydroxide is not suitable for the storage of arsenic because it dissolves in acidic solutions and releases the adsorbed arsenic.[1,2] Recently, scorodite has been studied as a promising storage material of arsenic.[3,4] Scorodite releases little arsenic when the pH value of the solution is changed. Therefore scorodite is an attractive material for storing arsenic. Scorodite is synthesized through a solution method using arsenic (As(V)) acidic solution and iron (Fe(II)) acidic solution.[5] Large scorodite particles(> 10 μm) are synthesized at high temperature (>90°C solution) in general. Particle size of scorodite can vary but larger ones are preferred because low surface-to-volume ratio makes it difficult to dissolve into the acidic solution. The purpose of our study is to apply ultrasound irradiation for synthesizing large scorodite particles(> 10 μm).

2. Experimental

As(V) acidic solution was prepared using a $Na_2HAsO_4 \cdot 7H_2O$, H_2SO_4, and ion-exchange water. Then, Fe(II) solution was added to the arsenic solution. Finally Fe(II)-As(V) solution (50 ml) was adjusted to a Fe/As molar ratio of 1.5. As(V) concentration of the solution was 20 g/L. sonication was performed with an ultrasonic generators (TA-4021; KAIJO) and submersible transducers (KAIJO). The Output and the frequency of the transducer were adjusted to 200 W and 200 kHz. A submersible transducer was placed at the bottom of a tank filled with water, and a flat-bottom flask containing the sample solution was placed directly above the transducer. Before the sonication of the solution, oxygen gas (100 ml/min) was flowed into the solution for 20 min to purge air in the flask. The temperature of the irradiated solution was controlled at about 50, 60, 70, 80 and 95°C using a heating system. **Fig. 1** shows the experimental apparatus of ultrasound irradiation. Sonication or stirring (1000 rpm) for the solution was conducted at 70°C for 1 h in oxygen gas atmosphere. The precipitates from the above process were filtered using a 0.45 μm pore diameter membrane filter. After drying, the precipitates were analyzed using X-ray diffraction (XRD) and scanning electron microscope (SEM).

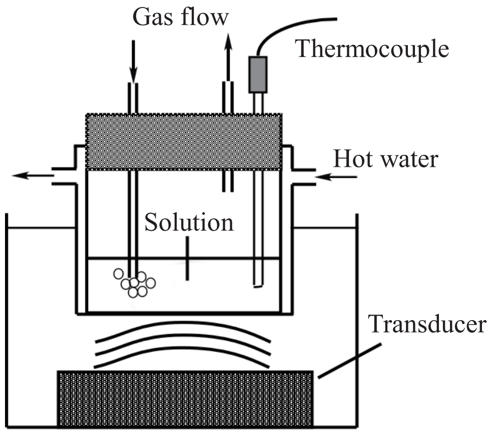

Fig. 1 Experimental apparatus of this study.

3. Results

We first confirmed effects of solution temperature and pH value on the synthesis of scorodite without using sonication. **Fig. 2** shows XRD patterns of precipitated samples synthesized by stirring at different temperatures with pH values fixed at 1.0. **Fig. 3** shows SEM images of those precipitated samples. XRD patterns of precipitated samples indicate that scorodite was successfully synthesized at each temperature. XRD peak intensity of samples was dependent on solution temperature. The XRD peak intensity became higher as the solution temperature was increased. Thus, the crystallinity of particles increases at high solution temperature. SEM observation results indicated that particles were generated by agglomeration of fine particles at 60°C and their particles size were < 1 μm. However, the morphology and size of particles were drastically changed at > 70°C synthesis conditions.

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The morphology was polyhedral shaped and the size was 1-2 µm. We also investigated the effect of pH value on particles using the solution of pH 2.0. As this pH, the particle size was 3-5 µm at 70°C, larger than that synthesized at pH 1.0. Therefore, among different synthesis conditions tested, pH value at 2.0 and oxidation temperature at > 70°C are the best condition to synthesize large size scorodite as a crystallinity structure.

Fig. 4 shows XRD patterns of samples synthesized using 200 kHz irradiation or stirring. XRD patterns of precipitated samples indicate that scorodite was successfully synthesized using 200 kHz irradiation. The intensity of XRD peaks was similar to that in stirring condition. Thus, the crystallinity of particles synthesized by ultrasound irradiation is almost the same as that synthesized by stirring. **Fig. 5** shows SEM images of precipitated samples synthesized using 200 kHz irradiation or stirring. Two samples had nearly identical polyhedron shape. However, the particle size was obviously different. The scorodite particles synthesized by ultrasound irradiation were larger. Most particles were > 10 µm. In contrast, the particle size of scorodite synthesized using stirring was approximately 3-5 µm. Despite that high solutions temperature weakens the oxidation effect of ultrasound irradiation, particle sizes are still larger than stirring. Thus, it appears that oxidation effect and agglomeration effect of ultrasound irradiation assist the growth of scorodite particles.

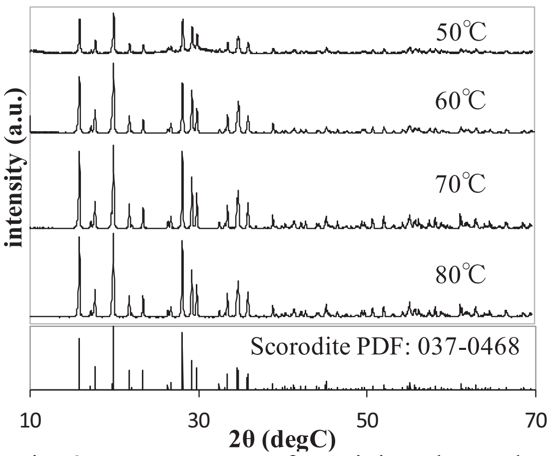

Fig. 2 XRD patterns of precipitated samples synthesized at each temperature for 1 h by stirring.

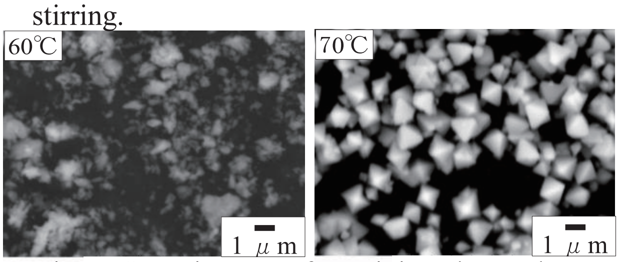

Fig. 3 SEM images of precipitated samples synthesized at 60°C or 70°C.

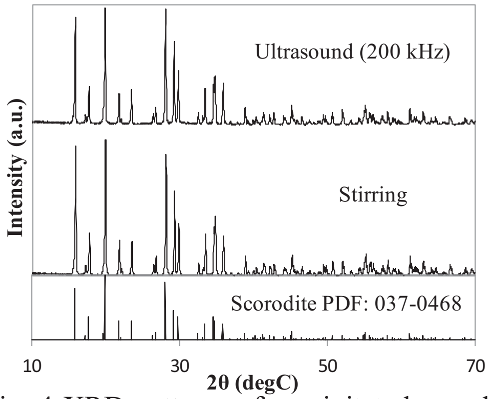

Fig. 4 XRD patterns of precipitated samples synthesized at 70°C for 1 h by ultrasound irradiation or stirring.

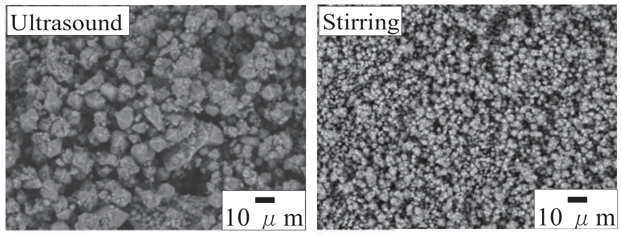

Fig. 5 SEM image of precipitated samples.

4. Conclusions

Scorodite was successfully synthesized in large size using 200 kHz ultrasound irradiation (pH 2.0, 70°C). Particle size of scorodite obtained by ultrasound synthesis (about 10 µm) was larger than that obtained by stirring synthesis (about 3-5 µm). Although the oxidation effect of ultrasound irradiation is reduced by the high solution temperature, particle sizes are larger than stirring. Thus, Oxidation effect and agglomeration effect of ultrasound irradiation appear to assist the growth of scorodite particles.

Acknowledgment

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