1. Introduction
Arsenic is toxic for the human body. Therefore, waste materials and factory effluents containing arsenic must be treated and stored using the appropriate methods. In mining, arsenic is commonly found in the form of enargite (Cu₃AsS₄) and tennantite (Cu₁₀Fe₂AsS₁₃) sulfide minerals that are present in the copper ore deposits. Recently, disposal of the highly-concentrated arsenic accrued by the repeated processing of flue cinder in copper smelting has become difficult because of the increased arsenic concentration in the copper mineral. Because of the use of sulfuric acid to leach copper in the flue cinder, arsenic removal and storage materials must be stable under acidic conditions. Scorodite (FeAsO₄·2H₂O) has been studied as a promising storage material of arsenic. Scorodite releases little arsenic when the pH value of the solution is changed. Scorodite is synthesized by oxidation in an acidic solution containing divalent iron [Fe(II)] and pentavalent arsenic [As(V)]. A larger particle size is preferred because a low surface-to-volume ratio makes it difficult to dissolve scorodite in an acidic solution. Large scorodite particles (>10 μm) are synthesized at high temperature (>90°C) solution using stirring in general. Therefore, the reaction temperature is an important factor to synthesize scorodite with a large particle size. The purpose of the present study is to apply agglomeration effect of ultrasound irradiation for the synthesis of scorodite particles as large particle size without synthesis of fine particles.

2. Experimental
Arsenic acidic the solution [As(V)] was prepared using Na₂HAsO₄·7H₂O, H₂SO₄, and ion-exchange water. Then, Fe(II) solution was added to the As(V) 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 solution was placed directly above the transducer. The temperature of the irradiated solution was controlled at 50 or 70°C using hot water circulation around the flat-bottom flask. Before the sonication of the solution, oxygen gas (100 ml/min) was flowed into the solution for 20 min to replace the air with oxygen gas in the flask. 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) measurement, scanning electron microscope (SEM) observation, and ultraviolet-visible absorption spectra (UV-vis) measurement.

3. Results and Discussion
First, scorodite was successfully synthesized at low solution temperature of 50°C. The intensity of the XRD peaks differs between two conditions (stirring and ultrasound irradiation). The crystallinity of particles synthesized by ultrasound irradiation is higher than that obtained by stirring. In this study, we investigated the effect of reaction temperature during ultrasound irradiation on the size and morphology of the synthesized scorodite particles.
consider that the agglomeration effect of ultrasound irradiation during the particle growth process reduced the number of nuclei for crystal growth and enhanced the supply of solute from the solution to the agglomerated particles. However, such the low reaction temperature (50°C) does not synthesized scorodite particles as large size (>10 μm) for reaction time of 3 h under both conditions of stirring and ultrasound irradiation. Therefore, the temperature of the reaction solution was raised to 70°C. Fig. 2 shows the SEM images of precipitated samples at 70°C using ultrasound irradiation. The particles synthesized at 70°C by ultrasound irradiation or stirring were polyhedral shape. The particle size obtained by stirring was 3–5 μm, and that by ultrasound irradiation was a mixture of large (>10 μm) and small (<1 μm) particles. From this result, at 70°C, large scorodite particles with polyhedral shape were obtained by oxidation promotion and dissolution enhancement of the agglomerated particles using ultrasound irradiation. However, small particles may come from the nucleation by cavitation during ultrasound irradiation. It has been reported that the frequency of ultrasound decides the generation amount of oxidant by cavitation and around 200 kHz effective to generate oxidants. So, we relatively confirmed cavitation effect using KI method at different solution temperature. 0.1 M KI solution (50 ml) was irradiated at 50 or 70°C for 30 min. Fig. 3 shows the I3− molar concentration of irradiated solution at 50 or 70°C. From this result, at 70°C, we consider that the small particles generated by the generation of cavitation bubbles during ultrasound irradiation. Also, the generated I3− molar concentration was decreased with a rise in temperature from 50°C to 70°C. Therefore, by increasing the solution temperature (>70°C), we expect to be able to obtain large scorodite particles without synthesis of fine particles because the generated I3− molar concentration was decreased with a rise in temperature from 50°C to 70°C.

4. Conclusion

To investigate the effect of reaction temperature on the size and morphology of scorodite synthesized using ultrasound irradiation, we performed the synthesis of scorodite particles at different temperature (50 or 70°C) for 3 h with oxygen flow. At 50°C, the sizes of scorodite particles were 1–3 μm (stirring) and about 3 μm (ultrasound). It is difficult to synthesize scorodite particles as large size (>10 μm) under stirring or ultrasound irradiation at 50°C. At 70°C, the particles size obtained by stirring was 3–5 μm, and that by ultrasound irradiation was a mixture of large (>10 μm) and small (<1 μm) scorodite particles. In result of KI method, we consider that the small particles generated by the generation of cavitation bubbles. Therefore, by increasing the solution temperature (>70°C), we expect to be able to obtain large scorodite particles without synthesis of fine particles because the generated I3− molar concentration was decreased with a rise in temperature from 50°C to 70°C.

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