

## The effect of ultrasound irradiation on arsenic adsorption from aqueous solution into compounds prepared from quarry waste 廃石を用いたヒ素の除去における超音波の効果

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### 1. Introduction

The coprecipitation of pentavalent arsenic with trivalent iron compounds such as iron hydroxide and ferric oxide has been used as one of the methods to adsorb arsenic in aqueous solution [1]. The valence of arsenic in aqueous solution is usually trivalent or pentavalent, and arsenic mainly forms As(III) as arsenious oxide in the effluent. Compared to the adsorption processes of As(III) using trivalent iron compounds, the adsorption processes of As(V) is easier to handle [2]. The adsorption process of As(III) requires the oxidation from As(III) to As(V) using oxidants. Therefore it is crucial to oxidize As(III) to As(V) with high efficiency. Ultrasound irradiation has been used as an oxidation method of As(III) [3]. In this study, we performed the removal of As(III) in the aqueous solution using ultrasound oxidation and trivalent iron compounds.

### 2. Experimental

The schematic design of the experimental apparatus is shown in Fig.1. The sonication was performed with an ultrasonic generator (TA-4021; KAIJO) and a submersible transducer (KAIJO). The output and frequency of this device was adjusted to 200W and 200kHz. A submersible transducer was put on bottom of a tank filled with water, and a round bottom flask containing a sample solution was placed right above the transducer. The temperature of the irradiated solution was controlled to 25°C. As<sub>2</sub>O<sub>3</sub>, Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O and FeCl<sub>3</sub>·6H<sub>2</sub>O were used as As(III), As(V) and Fe(III), respectively. NaOH was used for pH adjustment.

In order to confirm the oxidation performance of this equipment, 60ml of As(III) solution (10ppm) was sonicated for 15min or 30min. The oxidation ratio of As(III) to As(V) was evaluated using a molybdenum blue method.

As(III) removal experiments were conducted as follows. First, 60ml of As(III) solution (10ppm) was sonicated for 30min. After irradiation, 100ppm of Fe(III) was added into the

sonicated solution and the pH was adjusted to 6 with NaOH to generate precipitates. The treated solution was stood for 60min to precipitate Fe-As compounds. The precipitates were filtered through a 0.45μm membrane filter. The residual As concentration was measured using an ICP-OES instrument (SPS5510; SEIKO INSTRUMENTS INC.). To argue for the advantage of sonication, we conducted same experiments without ultrasound irradiation. In such case 100ppm of Fe(III) was added into 10ppm of As(III) or As(V) solution and then the solution was stood for 90min.

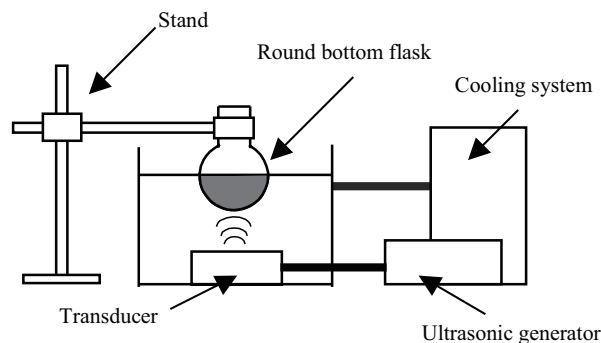
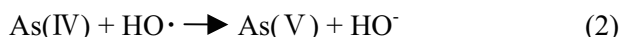
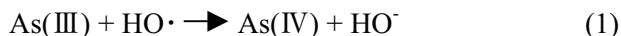


Fig.1 Schematic design of the experimental apparatus.

### 3. Results and Discussion

Fig.2 shows the ratio of oxidation As(III) to As(V) in the solution treated by sonication. The oxidation ratio became high with increasing irradiation time. The oxidation ratio of As(III) to As(V) after sonication for 15min and 30min showed around 20% and 30%, respectively. The oxidation mechanism of As(III) to As(V) using sonication has been reported by Neppolian et al. [3]. In aqueous solution, OH radical is generated by sonication. As(III) is oxidized to As(IV) by donating an electron to the OH radical (reaction1). The resulting As(IV) is highly unstable and it further undergoes oxidation to As(V) with OH radical (reaction2).



The results of our experiment indicate that the OH radical generated by ultrasound irradiation at 200kHz contributed to the oxidation of As(III) to As(V).

**Fig.3** shows residual As concentrations in the As(III) and the As(V) solutions treated by a trivalent iron compound either following ultrasound irradiation or without ultrasound irradiation. After the precipitation treatment of As(V) solution with the trivalent iron, residual As concentration showed a low value close to the value of effluent standard (under 0.1ppm). However, the precipitation treatment of As(III) solution with the trivalent iron showed 0.7ppm of residual As concentration. Thus, we could confirm that As(III) was more difficult to remove as compared to As(V) using the trivalent iron. On the other hand, residual As concentration decreased to about 0.2ppm after 30min of ultrasound irradiation followed by 60min of the trivalent iron treatment. This will be because As(III) was oxidized to As(V) by ultrasound irradiation, and the As(V) was easily adsorbed from the aqueous solution into the trivalent iron compound.

Now we are planning As adsorption from aqueous solution into compounds prepared from quarry waste of green tuff (Akita, Japan). **Fig.4** shows the photograph of green tuff. Green tuff has a potential as a coprecipitation material for As because green tuff contains about 2.3% of iron [4].

#### 4. Conclusions

We confirmed that As(III) was oxidized to As(V) by ultrasound irradiation. In addition, it became evident that the treatment of As(III) solution with ultrasound irradiation in combination with the application of trivalent iron caused residual As concentration in the solution lower than that treated only by the trivalent iron. In this preliminary study, we carried out the removal of As by forming precipitation with Fe(III). In the future, we will examine the removal of As with the use of quarry waste.

#### References

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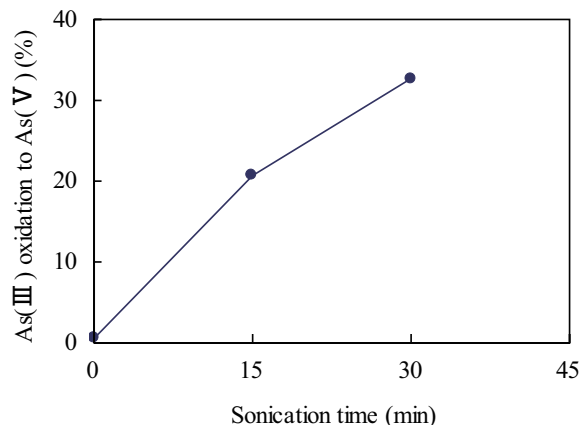


Fig.2 Variation in the oxidation ratio of As(III) to As(V) treated by 200kHz sonication.

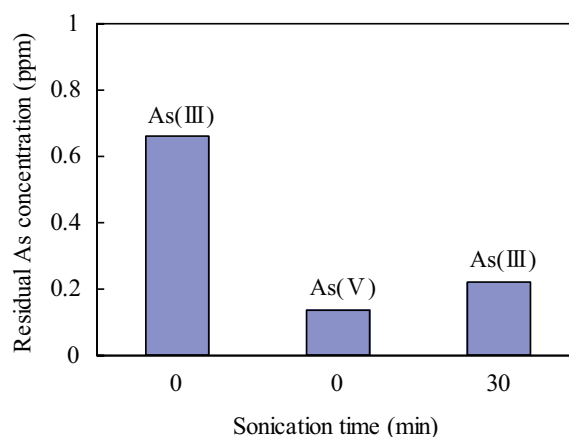


Fig.3 Effect of sonication on As removal by iron precipitate.



Fig.4 Green tuff waste.