## Production of arsenic RI tracer from gallium oxide target by alpha beam irradiation

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Arsenic (As) is toxic to living beings. Therefore, it is considered one of the most important environmental pollutants. Contamination of soil by As significantly influences the living environment and agriculture. Phyto extraction is an effective method to remove As from the contaminated soil.<sup>1)</sup> Thus it is necessary to determine the plant species that can efficiently extract As to achieve effective As removal. We need to investigate which plant species more efficiently extract arsenic to achieve effective arsenic removal. We believe that plant imaging using radioisotope (RI) is effective for that purpose. Arsenic is known to be stable in the trivalent and pentavalent oxidation states. However, it is suggested that the pentavalent arsenic species are predominant in soil due to the biological activity of transforming microorganisms.<sup>2)</sup> Therefore, the arsenic species in the same oxidation state as in soil are preferred for our present purpose. It was recently reported<sup>3,4)</sup> that the  $^{72}$ Se $(T_{1/2} = 8.5 \text{ d})/^{72}$ As $(T_{1/2} = 26.0 \text{ h})$  generator was prepared from a germanium target irradiated with protons. The tracer from the method, however, includes beta emitters useless for the positron imaging. In this study, we aim to develop a method for the tracer preparation of  $^{74}$ As ( $T_{1/2} = 17.77$  d), a positron emitter using the reaction system of a gallium (Ga) target with alpha particles for less contamination of beta emitters in the tracer. The <sup>74</sup>As radioactivity was measured by gamma-ray spectrometry with a high-purity germanium detector.

0.1 g of  $Ga_2O_3$  powder was formed into pellets  $(10 \text{ mm}\phi)$ . Each pellet was covered with an aluminum foil (10  $\mu$ m thickness) and used as a target. The target was irradiated with an alpha beam (29 MeV, 1.5  $p\mu A$ ) for 30 min. Following processes for tracer preparation were started after the byproducts such as  $^{71}As$  $(T_{1/2} = 65 \text{ h})$  and <sup>72</sup>As decayed out at least 4 weeks after the irradiation. The irradiated  $Ga_2O_3$  was dissolved in 1 mL of 12 M NaOH by heating up to 80°C. 917 mg of non-irradiated  $Ga_2O_3$  was dissolved in 9 mL of 12 M NaOH and added to the target solution, which was neutralized into a pH of 4 to 5 with ca. 2 mL of 6 M HCl and changed into the suspending solution. The suspension was subjected to solvent extraction with 5 mL of 1 M Di-(2-ethylhexyl)phosphoric acid (HDEHP) organic solution with either of benzene, toluene, and 1-octanol by shaking for ca. 1 h. The aqueous phase from solvent extraction with toluene or benzene was processed into the following chemical treatment after precipitation of

Table 1. Arsenic yields extracted into aqueous phase.

solvent	benzene	toluene	1-octanol
yield (%)	$66.5\pm0.8$	$65.8\pm 0.9$	$40.2\pm0.5$



Fig. 1. A part of the scheme for As tracer preparation.

NaCl by adding ethanol.

The supernatant collected from the aqueous solution was treated after measuring its radioactivity, as depicted in Fig. 1, where a chromatography column of 2 mL anion exchange resin of Muromac<sup>®</sup>  $1 \times 8 \ 100-200$ mesh (Cl<sup>-</sup> form) filled into the 30 Muromac<sup>®</sup> Minicolumn M (7.5 mm $\phi \times 45$  mm) was used. This column was flushed with 20 mL of H<sub>2</sub>O, 20 mL of 1 M NaOH, and 20 mL of H<sub>2</sub>O in a sequential order. The ion form of the resin was changed from Cl<sup>-</sup> to OH<sup>-</sup> in the preparative conditioning. The supernatant was charged into the column, which was flushed with 20 mL of H<sub>2</sub>O to remove ethanol and undesired species. After that, <sup>74</sup>As was eluted with 20 mL of 0.1 M HCl.

Table 1 presents the results of solvent extraction. The recoveries of <sup>74</sup>As isotope are nearly the same for benzene and toluene except for 1-octanol. The loss of <sup>74</sup>As in coprecipitation with NaCl was ca. 1.7%. The column chromatographic process in Fig. 1 lost ca. 10% of the charged <sup>74</sup>As onto the column. We successfully prepared an <sup>74</sup>As tracer solution including 60% of the produced radioactivity in a Ga<sub>2</sub>O<sub>3</sub> target combining solvent extraction and anion exchange column chromatography.

## References

- 1) M. Lei et al., Env. Sci. and Poll. Res. 25, 124 (2018).
- 2) R. E. Macur et al., Env. Sci. and Tech. 38, 104 (2004).
- 3) M. Jahn et al., Radiochim. Acta 98, 807 (2010).
- 4) D. E. Wycoff *et al.*, J. Chromatogr. A **1340**, 109 (2014).

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