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Effect of valence state on chromatographic fractionation of molybdenum isotope in aqueous hydrochloric acid solutions

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Abstract

The Mo(V) and Mo(VI) isotope fractionation using typical anion-exchange resins was investigated in 0.10 M (M = mol / dm^3) HCl solutions at 308 and 338 K. The examined resins were benzimidazole-type anion-exchange resin embedded in high-porous silica beads, which has two types of functional groups consisting of 1-methylbenzimidazole and 1,3-dimethylbenzimidazole (AR-01(Cl form)), weakly basic porous-type WA20 resin (WA20(Cl form)), and PA316 resin, which is one of porous-type strongly basic anion-exchange resin (PA316(Cl form)). As a result, the isotope separation coefficients of Mo(VI) species were obtained by using the isotope fractionation curve of Mo(VI) species with AR-01(Cl form) and WA20(Cl form) resins while we couldn't observe the Mo(VI) isotope fractionation in case of PA316 resin and the Mo(V) isotope fractionation using these resins. It may be suggested that these tendencies for the Mo isotope fractionation are caused by the weak adsorption of Mo(VI) species onto PA316 resin and the strong adsorption of Sn species used for reduction of Mo(VI) species for these resins.

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1. Introduction

^{99m}Tc, a metastable isomer of ^{99}Tc, is of great interest from the viewpoint of the medical use of nuclear diagnostics due to the half life of $T_{1/2}$ = 6.015 h and 143 keV [1]. In case of Japan, ^{99}Mo, a raw material of ^{99m}Tc, has been imported from foreign countries such as Canada, Netherlands, and Belgium, etc. [2]. Nowadays, most ^{99}Mo is produced by using nuclear research reactors with highly enriched ^{235}U (HEU), which has intrinsically some serious worries for nuclear proliferation. These reactors have been getting decrepit and the realistic costs for specialized facilities for chemical treatments, storages, and the disposal of large amounts of highly radioactive wastes are not reasonable [3]. Recently, some researchers have suggested that ^{99}Mo can be produced using the respective reactions of ^{98}Mo(n, γ)^{99}Mo, ^{100}Mo(n, 2n)^{99}Mo, and ^{100}Mo(p, x)^{99}Mo reactions [2,4]. Before their nuclear reactions, it also has been required to enrich ^{98}Mo or ^{100}Mo isotope for preparation of the enriched ^{99}Mo isotope because of comparatively low natural abundance of ^{98}Mo and ^{100}Mo isotopes. The Mo isotope fractionation in chemical reactions is also particularly interesting for geochemists [5-7]. Our works which have examined the mechanisms of Mo isotope fractionation in solutions, may contribute to the understanding the nature of the isotope fraction of Mo in the natural world. Some researchers have studied the chemical enrichment of various nuclides by using chromatography [8-23]. In analogy of them, we have also performed some chromatographic isotope separation experiments of hexavalent Mo species using the synthesized benzimidazole-type anion-exchange resin embedded in high-porous silica beads in hydrochloric acid solutions [22]. It has been known that Mo species has various chemical forms in aqueous solutions and seven stable isotopes in nature [22]. In other words, the systematic understanding of adsorption and desorption behavior of Mo species is inevitable, compared with other elements. However, the chemical data on isotope fractionation of medium-heavy elements such as Mo are not well-known. Little information on the effect of different valence states on chromatographic fractionation of Mo isotopes in hydrochloric acid solutions is available.

From these backgrounds, we have examined the effect of valence states on Mo isotope fractionation behavior in the hydrochloric acid solutions using typical anion-exchange resins (see Figure 1) such as benzimidazole-type anion-exchange resin embedded in high-porous silica beads, which has two types of functional groups consisted of 1-methylbenzimidazole and 1,3-dimethylbenzimidazole (AR-01(Cl form)), weakly basic porous-type WA20 resin (WA20(Cl form)), and PA316 resin, which is one of porous-type strongly basic anion-exchange resin (PA316(Cl form)).