



Development of an externally controllable sealed isotope generator[☆]

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HIGHLIGHTS

- Developed an externally controllable sealed isotope generator.
- Solid-liquid and solid-solid separation generators were applied.
- Maintained high extraction efficiency and low breakthrough during repeated extractions.

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ABSTRACT

An externally controllable sealed isotope generator has been proposed for radiation education activities. Column (⁶⁸Ge-⁶⁸Ga and ¹³⁷Cs-^{137m}Ba) and solvent extraction (⁶⁸Ge-⁶⁸Ga)-based isotope generators were applied as radioactive sources. These generators showed high milking efficiencies and low breakthrough after repeated uses, and are expected to promote the use of isotope generators without radioactive contamination or the emission of radioactive waste. This isotope generator provides a new concept for sealed radioisotope sources.

1. Introduction

In 1996, the International Atomic Energy Agency (IAEA) recommended two regulatory exemptions (doses and concentrations) for radionuclides (IAEA, 1996). This recommendation encourages the use of radionuclides to examine and clarify the properties of radiation and radionuclides through experiments in schools and universities because practical training can be conducted at universities and schools using radionuclides below the regulatory dose limit. However, in the case of open sources, the small amounts of radionuclides used in these experiments may result in radioactive contamination and the generation of radioactive waste. We previously developed a ⁶⁸Ge-⁶⁸Ga tin(IV) oxide column-based isotope generator for radiation education activities that uses radionuclides at lower levels than the legal exemption (Nozaki, 2016; Washiyama et al., 2015). The SnO₂ is used as one of the most common column packing materials in ⁶⁸Ge-⁶⁸Ga generators, and ⁶⁸Ga is eluted as Ga³⁺ with HCl while ⁶⁸Ge (Ge⁴⁺ form) remains adsorbed (Sudbrock et al., 2014). In SnO₂ based ⁶⁸Ge-⁶⁸Ga generator,

distribution coefficient (Kd) value for ⁶⁸Ge on SiO₂ was determined as high (2,113) in 1 M HCl, whereas it was low (0.30) for ⁶⁸Ga³⁺ (Chakravarty et al., 2016). ¹³⁷Cs-^{137m}Ba generator is in common use for education experiments in the educational facilities (SSERC Bull, 2006). Nickel ferrocyanide on anion exchange resin has been reported as a sorbent for ¹³⁷Cs-^{137m}Ba generator column (Watari et al., 1967; Nagai and Watari, 1968). During the process of ¹³⁷Cs loading on the nickel ferrocyanide anion exchange resin (Amberlite IRA-904), ¹³⁷Cs (¹³⁷Cs⁺ form) is adsorbed on the resin, whereas ^{137m}Ba (^{137m}Ba²⁺ form) is not retained by the resin. However, anion exchange resin, itself does not adsorb ¹³⁷Cs cation, ferrocyanide portion of the resin participates in the binding with ¹³⁷Cs (Nagai and Watari, 1968). The Kd values for ¹³⁷Cs on nickel ferrocyanide-anion exchange resin is not determined. Copper ferrocyanide-anion exchange resin, a kind of metal salt-ion exchange resin, showed particularly high affinity (Kd = 3500) for ¹³⁷Cs in H₂O (Watari et al., 1967).

In order to expand the use of above-mentioned isotope generators for radiation education investigations without radioactive

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Table 1
Relevant nuclear data for ^{68}Ge - ^{68}Ga and ^{137}Cs - $^{137\text{m}}\text{Ba}$ generator systems.

Generator	Parent nuclide			Daughter nuclide		
	Half life	Decay mode	γ emission	Half life	Main emission	Main photon (keV)
^{68}Ge - ^{68}Ga	270 d	EC	no γ	67.7 m	β^+	511
^{137}Cs - $^{137\text{m}}\text{Ba}$	30.0 y	β^-	no γ	2.55 m	γ	662 (IAEA, 2013)

contamination or the generation of radioactive waste, we proposed a sealed isotope generator that is controllable from the outside. In the present study, we report the radioisotopic evaluation of column (^{68}Ge - ^{68}Ga and ^{137}Cs - $^{137\text{m}}\text{Ba}$) and solvent extraction (^{68}Ge - ^{68}Ga)-based generators in a basic study and demonstrated that the decay and growth of daughter radioactivity can be measured from outside of the sealed generator by milking daughter nuclides from parent ones using a prototype generator. The relevant nuclear data for ^{68}Ge - ^{68}Ga and ^{137}Cs - $^{137\text{m}}\text{Ba}$ generator systems are given in Table 1.

2. Materials and methods

2.1. Preparation of a tin(IV) oxide column for the chromatographic separation of ^{68}Ga from ^{68}Ge

The adsorbent SnO_2 for chromatographic separation was made by the following process. Tin (IV) chloride ($\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$) (Wako Pure Chemical Industries Ltd., Osaka, Japan) solved in distilled water was precipitated by the addition of sodium hydroxide aqueous solution. The $\text{SnO}_2 \cdot n\text{H}_2\text{O}$ gel was precipitated by neutralization of SnCl_4 (1.5 M) to pH 4.0 ± 0.1 , and collected and washed with centrifugation. The gelatinous mass spread over a large watch glass and warmed gently to be moist soft lumps. The lumps were pressed quite mildly onto 500 μm meshes of a sieve to be divided into particles. The particles were kept over potassium acetate a few days for mild drying. The particles were sieved through 250 μm meshes. Similar treatment of the particles was repeated by the use of 100 μm mesh sieve. The particles were sieved into two sizes: 212–106 μm and 106–38 μm , and kept over potassium acetate. After a week or more, the particles were dipped in 0.3 M HNO_3 and gently refluxed for 3 h. To remove the fine particles by decantation, suspension solution of HNO_3 was replaced by 1 M HCl and suspended. This process was repeated for several times. Prepared particles (sieved into 212–106 μm) in approximately 0.2 mL were packed into a disposable 1-mL plastic syringe in order to make a generator to test performance.

2.2. Preparation of a nickel ferrocyanide-anion exchange resin column for the chromatographic separation of $^{137\text{m}}\text{Ba}$ from ^{137}Cs

Nickel ferrocyanide-anion exchange resin was prepared using a previously described method (Watari et al., 1967; Nagai and Watari, 1968) with the following modifications. Amberlite IRA-904 (18–30 mesh; ORUGANO Co., Ltd., Tokyo, Japan) was converted to ferrocyanide in 0.5 M potassium ferrocyanide solution. The resin was then washed with distilled water and suspended in 1 M nickel chloride solution. The nickel ferrocyanide-anion exchange resin that formed was washed with distilled water. This process was repeated four times. Approximately 0.2 mL of the prepared nickel ferrocyanide-anion exchange resin was packed into a disposable 1-mL plastic syringe in order to make a generator to test performance. A polypropylene sheet (P-50R, pore size of 50 μm ; FILTEREN Co., Ltd., Saitama, Japan) was packed at the bottom of the syringe in order to retain the resin.

2.3. Radioisotopic evaluation of the column-based isotope generator

^{68}Ge was produced by the proton irradiation of a gallium metal target, was recovered by in-situ extraction using a cocktail of 1 M HCl and 30% H_2O_2 (1:1) in the National Institutes of Quantum and Radiological Science and Technology (Chiba, Japan). The recovered crude ^{68}Ge solution was further purified by passing through a AG50W-X8 column to eliminate tiny amount of Ga as well as H_2O_2 , and was prepared in 1 M HCl (Nagatsu et al., 2015). A total of 1.06 kBq/10 μL of ^{68}Ge solution was applied to the tin(IV) oxide-packed column (0.2 mL). The isotope generator was eluted by infusing 1 mL of 1 M HCl using a disposable 1-mL plastic syringe. Column and elute activities were measured with an auto well-type NaI (Tl) scintillation counter (Accu-FLEX γ 8001, Hitachi Ltd., Tokyo, Japan). ^{68}Ga elution efficiency was obtained by the equation as follows.

$$\text{Elution efficiency} = \frac{^{68}\text{Ga activity in the elute}}{\text{Total } ^{68}\text{Ga activity in the column}} \times 100(\%)$$

^{68}Ge leakage (breakthrough) was evaluated from ^{68}Ga present in the elute a week after the end of elution as follows.

$$\text{Breakthrough} = \frac{^{68}\text{Ga activity in the elute 1 week after the elution}}{^{68}\text{Ga activity eluted from the column}} \times 100(\%)$$

All calculations were performed using decay and geometry corrected counts.

In order to examine the elution properties of the generators, isotope generators ($n = 5$) were eluted with reused eluents that did not contain a significant amount of the daughter nuclide. The radioactivity of daughter nuclides in the eluate completely decayed by the next elution (milking). The assessment of elution efficiency and breakthrough was repeated ten times in six months.

^{137}Cs , with specific activity of 1.61×10^8 Bq/g in 0.1 M HCl, was purchased from the Japan Isotope Association (Tokyo, Japan). A total of 3.4 kBq/20 μL of $^{137}\text{CsCl}$ solution was applied to the nickel ferrocyanide-anion exchange resin column (0.2 mL). The isotope generator was eluted by infusing 1 mL of distilled water with a disposable 1-mL plastic syringe. Column and elute activities were measured with a well-type NaI(Tl) scintillation counter, and $^{137\text{m}}\text{Ba}$ elution efficiency was assessed as follows.

$$\text{Elution efficiency} = \frac{^{137\text{m}}\text{Ba activity in the elute}}{\text{Total } ^{137\text{m}}\text{Ba activity in the column}} \times 100(\%)$$

^{137}Cs leakage (breakthrough) was evaluated from $^{137\text{m}}\text{Ba}$ present in the eluent an hour after the end of elution as follows.

$$\text{Breakthrough} = \frac{^{137\text{m}}\text{Ba activity in the elute 1 hr after the elution}}{^{137\text{m}}\text{Ba activity eluted from the column}} \times 100(\%)$$

In order to investigate the elution properties of the generators, isotope generators ($n = 5$) were eluted with reused eluents that did not contain a significant amount of the daughter nuclide. The assessment of elution efficiency and breakthrough was repeated ten times in six months. A one-way analysis of variance ANOVA with Dunnett's test was used to test the significant difference of data in breakthrough.

2.4. Radioisotopic evaluation of a solvent extraction-based isotope generator

The extraction of ^{68}Ga with organic solvents was employed for the preparation of the hydrophobic ^{68}Ga compound. 8-Hydroxyquinoline (oxine) was used as an extractant according to a previously described method (Ehrhardt and Welch, 1978). Extraction was performed in a glass test tube (16.5 \times 108 mm, round bottom) with a polytetrafluoroethylene screw cap. Oxine dissolved in ethanol (50 μg /50 μL) was added to ^{68}Ge (1.06 kBq) containing 5 mL of 1 M acetate buffer solution (pH 5). After the addition of 5 mL organic solvent, chloroform, or 1-octanol (octanol), the ^{68}Ga -containing sample was shaken in a sealed glass test tube for 2 min, and ^{68}Ga was extracted with an organic

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