Chemical separation of ^{139}Ce from a ^{nat}La target using LN resin

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Endohedral metallofullerenes are clathrate compounds in which metal atoms and metal carbon clusters are included in a fullerene, which is a cage-like molecule composed of sp^2 carbons.[1] In particular, radioactive metallofullerenes encapsulating radioisotopes (RIs) are expected to be applied to radiopharmaceuticals such as a drug delivery system for transporting an RI to affected organs by chemical modification of the fullerene surface. We have studied the production of endohedral metallofullerenes containing lanthanide and actinide elements and their chemical properties. In these studies, it is necessary to use well-characterized radioactive Ce-encapsulated metallofullerenes as the comparators for the metallofullerenes encapsulating RIs which are carrier-free or without any stable isotopes such as promethium and actinide elements. So far, we have used chemically separated ^{139}Ce produced by the ^{139}La$(d, 2n)^{139}$Ce reaction in the RIKEN AVF cyclotron with the target material of ^{nat}La for our metallofullerene research.[2] However, it is necessary to use the highly toxic $K_2Cr_2O_7$ as an oxidizing agent for Ce in the solvent extraction method by di-(2-ethylhexyl) phosphoric acid (HDEHP), which we employed so far, and is not preferable for the application of radioactive metallofullerenes for nuclear medicine in future work. Therefore, we use LN resin (Eichrom Technologies, Inc.) (see Fig. 1), which is an extraction chromatography resin specialized for the separation of rare-earth elements using HDEHP, for the separation of ^{139}Ce from a target material containing La in large quantities. The purpose of this work was to separate and purify a trace amount of ^{139}Ce from La as a target by extraction chromatography using LN resin.

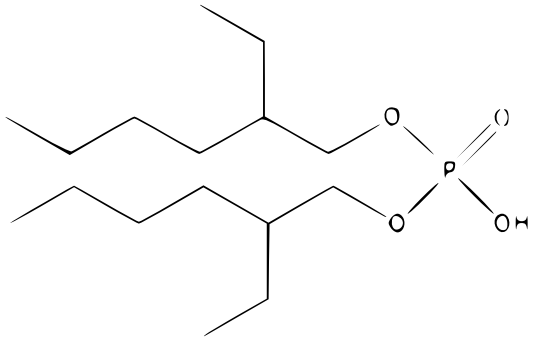

Fig. 1. Structure of the LN resin extractant.

About 1 g of a La plate as a target was irradiated with 24 MeV deuterons at a beam intensity of 5 μA in the RIKEN AVF cyclotron, and ^{139}Ce was produced by the ^{139}La$(d, 2n)^{139}$Ce reaction. The irradiated La target was dissolved in concentrated nitric acid to obtain a ^{139}Ce solution. A polyethylene column with a hight of 50 mm and a diameter of 7 mm was filled with LN resin and loaded with 100 μL of ^{139}Ce solution. A total of 25 mL of 0.15 M nitric acid was added to this column, and the eluate was collected every 5 mL. Then, a total of 10 mL of 6 M nitric acid was added to wash the column. Gamma rays with an energy of 165 keV of ^{139}Ce emitted from each fraction were measured with a Ge semiconductor detector. After gamma-ray measurement, 4-(2-pyridylazo) resorcinol (PAR) was added to each fraction to confirm the elution position of La by the ultraviolet/visible (UV/vis) absorption.

Figure 2 shows the elution behavior of ^{139}Ce during extraction chromatography, monitored by the relative radioactivity of 165 keV emitted from each fraction. The shaded fractions indicate the fractions containing La confirmed by UV/vis absorption. From these results, it is found that ^{139}Ce was sufficiently separated from La and that the proposed separation method is excellent for the separation of ^{139}Ce from a large amount of La used as a target material. Using this purified ^{139}Ce, we are currently researching metallofullerenes encapsulating ^{143}Pm and carrier-free endohedral metallofullerenes encapsulating ^{177}Lu.

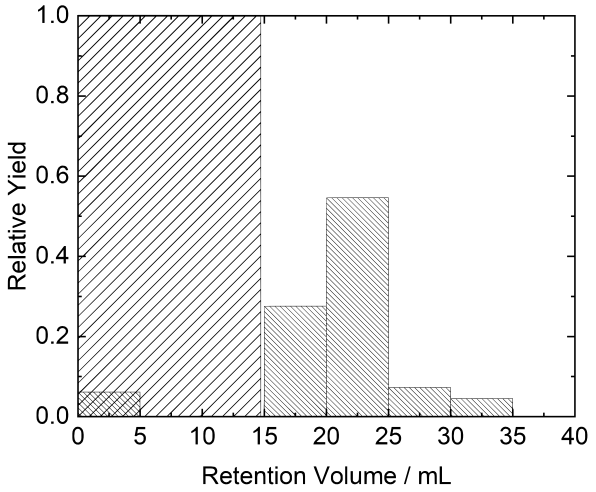

Fig. 2. Elution behavior of ^{139}Ce on LN resin. The shaded fractions indicate the fractions containing La, as confirmed by UV/vis absorption. The radioactivity in each fraction was normalized to that of loaded ^{139}Ce.

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References

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2) K. Akiyama *et al.*, RIKEN Accel. Prog. Rep. **53**, 194 (2019).