Separation and purification of ^{139}Ce tracer for metallofullerene synthesis

K. Akiyama,[*1,*2] K. Amekura,[*1] and H. Haba[*2]

Recently, clinical tests have been conducted on radionuclides of group 3 elements including lanthanides, and some radionuclides were newly approved as radiopharmaceuticals. Metallofullerenes encapsulating lanthanide elements are attractive for application in electronic devices and pharmaceuticals. In particular, it is expected that rich π electrons on a metallofullerene surface enable the production of various metallofullererene derivatives by the addition of various functional groups and that the encapsulation of radionuclides by these derivatives could be applied as radiopharmaceuticals. For such applications, excellent radionuclides, such as ^{139}Ce, emitting monochromatic γ rays and have a half-life of several months are required. In this paper, we report the separation and purification of a ^{139}Ce tracer from its target material for metallofullerene synthesis.

Cerium-139 was produced from the ^{139}La$(d, 2n)^{139}$Ce reaction using a ^{nat}La target irradiated with 5 μA of 24 MeV deuterons for 5 h at the RIKEN AVF cyclotron. After the irradiation, the La target containig ^{139}Ce was dissolved in nitric acid. For the separation and purification of ^{139}Ce from La, the solvent extraction method was employed and was executed according to the procedure shown in the Fig. 1. The solution samples obtained in a series of this extraction procedure were measured using a Ge semiconductor detector to check the remaining ^{139}Ce radioactivity in each sample.

The remaining amounts of the target material La and oxidant Cr were determined by optical absorption measurement and were found to be 2.1% and 3.9%, respectively. The amount of remaining La in a single separation process is still much larger than the amount of the ^{139}Ce tracer. However, it has been found that by repeating this separation procedure, it would be possible to obtain a sufficiently purified ^{139}Ce tracer.

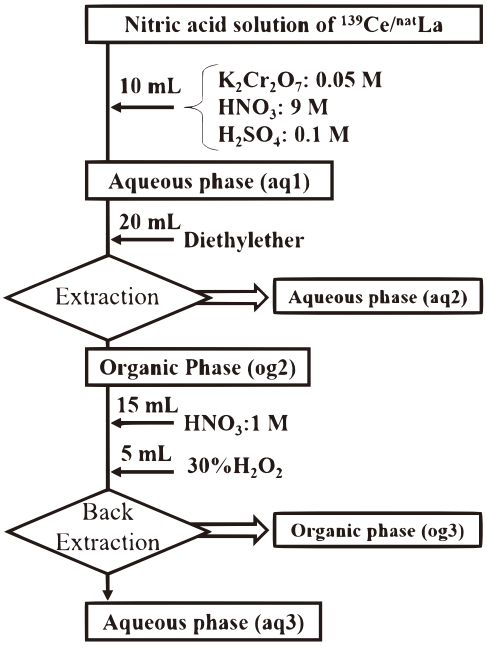

Fig. 1. Schematic diagram of ^{139}Ce separation from a of La target.

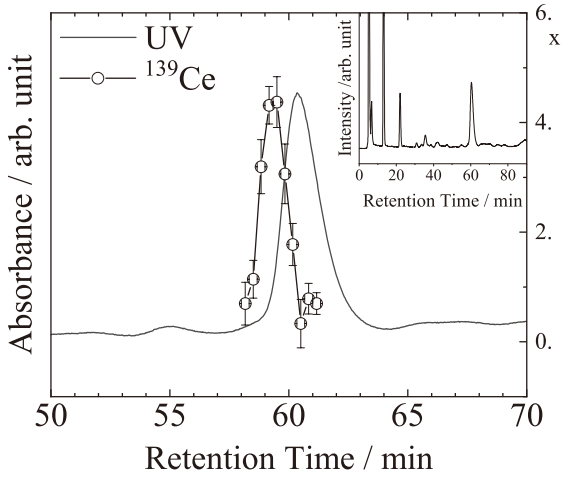

Fig. 2. HPLC elution behavior of ^{139}Ce metallofullerene together with UV-monitored chromatogram of metallofullerenes produced in this study.

As mentioned above, the separated ^{139}Ce tracer still includes large amounts of La and Cr. However, if the amounts of these impurities are sufficiently small relative to that of the added carrier element and do not affect the fullerene production, the purity of ^{139}Ce can be said to be sufficient in this experiment. To confirm this, Tb_4O_7 was added to a ^{139}Ce tracer solution, and this solution was employed for the metallofullerene production under the same conditions as those previously reported.[1] In Fig. 2, the high-performance liquid chromatography (HPLC) elution behavior of ^{139}Ce metallofullerene (open circles) on a Buckyprep column (Nacalai Tesque, Inc., efluent: tolunene) is shown together with a chromatogram aquired using UV absorption (gray solid line). The most dominant elution peak found in both chromatograms are due to M@C_{82}.[2] It is reported that the HPLC retention time of M@C_{82} on a buckyprep column increases as the atomic number of the encapsulated lanthanide atom increased.[3] The observed UV-monitored HPLC elution peak around 60 min occurs after that of Ce metallofullerene and is considered to be derived from Tb@C_{82} because no HPLC elution peak with sufficient intensity and faster retenton time than that of Ce@C_{82} is observed. In conclusion, the observed elution peak is derived from Tb@C_{82} and the amount of the remaning La with this separation method is found to be sufficiently small.

*1 Department of Chemistry, Tokyo Metropolitan University
*2 RIKEN Nishina Center

References

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