Radiochemical separation of ²²⁷Ac in environmental samples using solidphase extraction with ion-imprinted polymer resin

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Introduction

Radiochemical separation of naturally occurring actinides in water often requires several steps. Accurate quantification of actinium (Ac) necessitates the use of an internal standard. However, the scarcity of long-lived Ac tracers poses a challenge for a wide application of ²²⁷Ac as an environmental tracer. Radioactive ²²⁷Ac (actinium-227, $t_{1/2}$ = 22 years) arises from the radioactive decay chain of ²³⁵U (uranium-235, $t_{1/2}$ = 7.04·10⁸ years, 0.7 % of the total U), is one of the rarest naturally occurring radioisotopes and is expected to be found at secular equilibrium with ²³⁵U. The direct predecessor of ²²⁷Ac in the ²³⁵U chain is ²³¹Pa (protactinium-231, $t_{1/2}$ = 3.276·10⁴ years) which has a high affinity to particles and sediments, unlike the ²²⁷Ac which is predominantly partitioned within the water column of lake and marine systems. This phenomenon enables to exploit ²²⁷Ac as a promising tracer for upwelling velocity and for providing information on water column stratification changes useful on 100-year scale.¹ Herein we propose a method for the radiochemical separation of ²²⁷Ac in water, employing a polymer resin imprinted with yttrium (III).²

Materials and Methods

To test the extraction of ²²⁷Ac in the scope of use as a geological tracer, water sampled from the Lake Geneva was spiked with ²²⁷Ac and ²⁴³Am tracers. The actinides were pre-concentrated in two steps by co-precipitation with iron hydroxide and calcium oxalates which were then wet-ashed by microwave-assisted acid digestion under pressure. The extraction of ²²⁷Ac and ²⁴³Am was achieved from samples at pH 3 on 5 mL columns containing 2 g of Y³⁺-imprinted polymer resin.³ ²²⁷Ac and ²⁴³Am were released from column with 2 M HCl, electrodeposited on stainless steel discs and stored for 120 days to enable an ingrowth of alpha-emitting progenies of ²²⁷Ac prior to alpha spectrometry counting.

Results

The experiments with water from Lake Geneva spiked with ²²⁷Ac and ²⁴³Am yielded a radiochemical recovery of ²⁴³Am in the range of 70-80 % using Y-imprinted resin. Initial counting shortly after the electrodeposition identified traces of ²²⁷Ac progenies (Figure 1, green trace). The samples were recounted after 120 days at equilibrium between ²²⁷Ac and its progenies and showed an ingrowth as expected (Figure 1, red trace). To quantify the ²²⁷Ac, ²¹⁵Po peak free from isotopic interferences was used. Naturally occurring ²³⁸U and ²³⁴U were also partly (30 % of total U in water) retained by Y-imprinted resin, requiring a further development of a radiochemical separation method.



Figure 1. Alpha-spectrum of ²²⁷Ac progenies in Lake Geneva water extracted by Y-imprinted resin in the presence of ²⁴³Am tracer shortly after the radiochemical separation (green trace) and after 120 days of ingrowth (red trace).

Conclusion

Tracer experiments with lake water showed that Y-imprinted resin can be applied for the determination of naturally occurring ²²⁷Ac. Combined with a very sensitive detection technique, *e.g.* accelerator mass spectrometry, it can pave the way to a broader adoption of ²²⁷Ac as an oceanographic tracer in environmental sciences.

References

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[2] Chauvin et al., Chem. Eur. J. 12 (2006) 6852-6864

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