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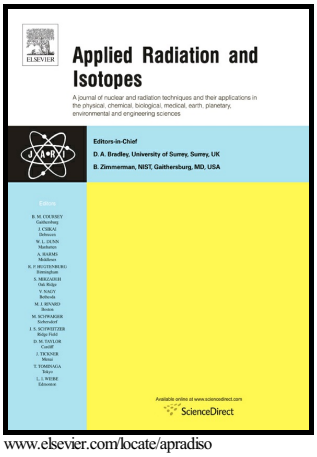

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Thorium-229 quantified in historical Thorium-228 capsules

Sven Boden[1], Koen Vints[1], Simone Cagno[1]*, Dominic Maertens[1], Karen Van Hecke[1], Thomas Cardinaels[1,2]

1. Belgian Nuclear Research Centre (SCK•CEN), Boeretang 200, B-2400 Mol, Belgium

2. KU Leuven, Department of Chemistry, Celestijnenlaan 200F, P.O. Box 2404, B-3001 Heverlee, Belgium

* Corresponding author: simone.cagno@sckcen.be

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Abstract

Thorium-229 is a valuable, but scarce, radionuclide for nuclear clock applications or targeted alpha therapy. While it is mostly produced by the decay of ^{233}U, ^{229}Th can also be produced by neutron irradiation of ^{226}Ra. At SCK•CEN, capsules containing mainly ^{228}Th (by-product of ^{226}Ra irradiation) were characterized to quantify the present amounts of ^{229}Th, ^{228}Th, ^{227}Ac, ^{226}Ra with high resolution gamma spectroscopy, after a decay period of 40 years in which ^{228}Th has decayed. High purity ^{229}Th was quantified, and after recovery using radiochemical separation processes, it can be used to support ongoing research.

1. Introduction

Thorium-229 is used as a generator of ^{225}Ac and ^{213}Bi for Targeted Alpha-immunotherapy (TAT) (see e.g. Elgqvist *et al.*, 2014 and Jurcic *et al.*, 2015), and for nuclear clocks (see e.g. Karpeshin and Trzhaskovskaya, 2015 and Jeet *et al.*, 2015). Thorium-229 and its daughters do not occur in nature and belong to the decay series of ^{233}U, bred from ^{232}Th. The aged ^{233}U stockpile is the only large source of ^{229}Th in the world (about 9.3 GBq – 250 mCi), but is not likely to increase due to safety and security issues (Alvarez, 2013). Moreover, it is not radioisotopically pure due to ^{232}Th and ^{228}Th (decay of ^{232}U) impurities. While for medical applications this is not a particular concern (besides additional shielding for ^{228}Th), isotopic impurities in ^{229}Th could be a significant issue for nuclear clocks (Stellmer *et al.*, 2015 and Von der Wense *et al.*, 2015).

The current driving need for ^{229}Th is medical research and an increase in ^{225}Ac/^{213}Bi production is needed (Nuclear Science Advisory Committee, 2015). Many new possible production routes for these three isotopes have been explored via particle accelerator or nuclear reactor (see e.g. Harvey, 2011; Hogle *et al.*, 2016; IAEA, 2009). While a few routes are promising, no relevant amounts have been produced yet, and the scarcity of ^{229}Th and its daughter nuclides limits ongoing research. Therefore, attempts are made to retrieve ^{229}Th from historical sources.

In 1969-1974, SCK•CEN and Union Minière conducted a project to produce a gram amount of ^{227}Ac (the so-called *Actinium Programme*), by irradiating ^{226}Ra in the high flux reactor BR2. Substantial amounts of ^{228}Th were co-produced during irradiation, and SCK•CEN developed radiochemical methods to separate the complex mixture of target material and irradiation products (Monsecour *et al.*, 1974; Monsecour *et al.*, 1977; Baetsle *et al.*, 1972). During the *Actinium Programme*, ^{227}Ac was separated after each irradiation cycle, while the ^{226}Ra target material was purified and recycled for a