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Ra-226/Ac-225 chemical separation R&D at SCK•CEN

The radiochemistry group of SCK•CEN possesses a purified Th-229 source since the beginning of 2017, routinely providing Ra-225, Ac-225, and Bi-213 tracers. This Th-229 had to be separated from long-lived Ac-227 and Ra-226 contaminants. The source originated from an Ac-227 production program in the 1970s, where highly radioactive Th-228 was co-produced by neutron capture of Ac-227 in the BR2 reactor when irradiating large Ra-226 targets. After 45 years in interim storage, the near-complete decay of Th-228 revealed the presence of useful quantities of Th-229, and Ra-226/Ac-227 impurities from the thorium separation process. SCK•CEN remained in possession of this large purified Ra-226 stock.

Easy access to significant quantities of Ac-225 and Ra-225 tracers had sparked interest and research in the field of Ra/Ac separation chemistry, mainly because cost-efficient large scale production of non-carrier added Ac-225 is conceivable using the strategic Ra-226 stock as the starting material. Protons, deuterons, neutrons, and photons can be used to produce Ac-225 using Ra-226 as a target material. Especially fast neutron and photon production pathways benefit from a large Ra-226 target mass.
Extraction chromatography experiments have been performed to identify a number of feasible separation and purification pathways to produce pure Ac-225 free from Ra-226 and its progeny (Pb, Bi, Po). Diglycolamide (DGA) and dialkylphosphoric acid (HDEHP) type solid phase extraction resins were tested as an approach to obtain high separation factors between Ra and Ac. Combining these two resin types was the key to efficient separation systems, as they work in the opposite acidity range. The choice of mineral acid (HNO3, HCl) influences the decontamination of Ac-225 from the progeny of Ra-226. The combination of a DGA or HDEHP resin with a U-TEVA and/or crown ether extraction resin was determined to be beneficial for removal of the progeny of Ra-226.

As a pre-purification step, precipitation in strong mineral acid (HNO3, HCl) can remove the vast majority of Ra-226 from the solution. Especially when large target masses are used, this process becomes advantageous. The choice of acid can be influenced by the irradiation target compound.

Ra-226 recycling throughout the process is necessary to avoid significant losses to the aqueous and solid waste. Decontamination of aqueous streams can be achieved by precipitation as RaSO4, or co-precipitation as Ba(Ra-226)SO4. RaSO4 can be converted to RaCO3, and re-dissolved in a mineral acid.
These separation experiments have offered a valuable insight into the Ra/Ac separation chemistry.

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