Neutron Activation Methods to Determine Actinides in Biological Materials

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Instrumental neutron activation analysis (INAA) and radiochemical neutron activation analysis (RNAA), fission track analysis (FTA), and delayed neutron INAA can be used to complement alpha spectrometry and other techniques for the determination of actinides in biological tissues, including nutritional materials. Radiochemical separation of actinide using isotopic tracers followed by alpha spectrometry is normally the method of choice because of the low radiometric detection limits (L_d<3x10^{-4} Bq/nuclide) achievable and the isotopic information provided (e.g., ^{228,230,232}Pu, and $^{241,\ 243}$Am). However, for nuclides with very long half-lives (e.g., ^{232}Th, ^{235,238}U, and ^{239}Pu), detection limits can be greatly improved using neutron activation methods, such as INAA, RNAA, preconcentration NAA, FTA, and delayed neutron NAAA. These activation methods may be combining with alpha spectrometric methods to provide adequate detection limits and improved precision for both short-and long-lived actinide isotopes. We review applications of activation methods to the determination of actinides, including Th, U, Np, and Pu isotopes, in biological tissues and we discuss potential applications to higher actinides.

This laboratory has recently developed activation methods combine with alpha spectrometry for the determination of Th, U, and Pu isotopes in human tissues and other biological samples. Determination of ^{240}Pu/^{239}Pu ratios in human tissues by combining alpha spectrometry and FTA was used to evaluate Pu sources for U.S. Transuranium and Uranium Registries cases of occupational exposure. Plutonium was separated from other actinides by anion exchange using ^{238}Pu as a tracer and was electrodeposited onto V disks. Alpha spectrometry determined $^{239+240}$Pu because the major alpha peaks of ^{239}Pu and ^{240}Pu are not resolved using high-efficiency surface-barrier or ion-implanted Si detectors. Plutonium-239 was then determined on the disk by FTA in a thermal neutron fluence of 1.5x10^{15} cm^{-2} using Lexan as the track detector. Measurement of the ^{240}Pu/^{239}Pu ratio in tissues from one individual showed that it is possible to identify multiple exposures to Pu of different isotopic composition. In another study, the distribution of Th is isotopes in all major tissues and organs of an individual exposed only to ambient levels of Th was carried out by separation of Th by ion exchange using ^{229}Th as a tracer, electrodeposition onto V disks, and determination of ^{228,230}Th by alpha spectrometry followed by determination of ^{232}Th by INAA using the ^{232}Th (n, y) ^{233}Pa reaction. Detection limits for ^{232}Th by INAA were 5x10^{-6} Bq compared to 2x10^{-4} Bq by alpha spectrometry. A similar procedure has been used to determine ^{235}U in tissues from individuals exposed occupationally to depleted or natural U. The detection limit for ^{235}U by alpha spectrometry is poor because of tailing of ^{234U} alpha peals into the ^{235}U region of interest. In this procedure, ^{234,238}U are determined by alpha spectrometry using ^{232}U as a tracer, and ^{235}U is determined by INAA using the ^{235}U(*n, f*) ^{140}Ba→ ^{140}La reaction.

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