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Title: Use of Accelerated Mass Spectrometry for Plutonium Urinalysis

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Abstract

The needs for low-level detection of plutonium isotopes are not currently being met by existing technologies, especially in the areas of human bioassay and radiation protection of workers. Conventional alpha-particle spectrometric methods commonly employed around the DOE complex have detection limits for plutonium-239 (^{239}Pu) on the order of 250 to 350 μBq, well above the requirements to satisfy regulatory standards for internal dosimetry programs. Consequently, a long-standing 'technological shortfall' has existed in providing adequate radiation protection for workers potentially exposed to Pu. Alternative techniques have included fission track analysis (FTA), inductively coupled plasma mass spectrometry (ICP-MS) and thermal ionization mass spectrometry (TIMS). Many of these techniques suffer from one or more of the following problems: (1) uncertainty in levels of recovery and/or removal of fissionable uranium; (2) isobaric or other interferences from molecular species, high background count rates, high sample preparation and/or measurement costs, and limited throughput for routine bioassay measurements. Previous studies conducted at the Australian National University (Canberra, Australia) had demonstrated that accelerator mass spectrometry (AMS) has the potential to avoid many of these disadvantages and provide a low-level detection capability for ^{239}Pu in the attogram range (<1 μBq).

We were able to demonstrate the viability of the LLNL AMS for detection of low levels of Pu isotopes for a number of different applications. The final spectrometer included a 45-degree cylindrical electrostatic analyzer (ESA) designed to improve on uranium (U) rejection. Total rejection of U is presently in the order ~10^7 or better, yielding reproducible backgrounds on the order of <1 μBq for ^{239}Pu. Instrumental precision was limited by time-dependence in the negative ion output. We developed a fast-switching data-acquisition mode to average out the signal over several passes, and improve on both precision and accuracy. Over one hundred analyses have now been been performed on artificial and human urine. AMS offers high efficiency, high rejection of interferences, and low susceptibility to matrix components; it also provides for rapid measurement throughput (>50 unknowns per 24 h) and a large dynamic range. This work clearly demonstrates that the CAMS AMS spectrometer at LLNL is well suited for detection of μBq concentrations of ^{239}Pu in urine, and provides for a rapid, cost-effective and robust measurement technique.

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