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Title page

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LOW LEVEL DETECTION OF PLUTONIUM IN HUMAN URINE USING ACCELERATOR MASS SPECTROMETRY

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Abstract

Internal dosimetry is an important part of nuclear safety for over 100,000 radiation workers in the United States. The U.S. Department of Energy (DOE) has established basic standards for occupational radiation protection in the Code of Federal Regulations, 'Occupational Radiation Protection', Title 10, Part 835 (10 CFR 835). Radiobioassay measurements on biological materials excreted or removed from the body are routinely used in internal dose evaluation programs. These programs require demonstrated compliance with annual dose limits, and should be able to confirm the presence of a radionuclide delivering a 50-year committed effective dose equivalent ($H_{E,50}$) in excess of 100 mrem (1 mSv). However, it has not always been feasible to confirm intakes that result in a 100-mrem $H_{E,50}$ for selected alpha emitting radionuclides such as plutonium because of the lack of adequate methods of detection. For example, conventional decay counting techniques such as alpha-spectrometry have a minimum detectable amount (MDA) for plutonium-239 (^{239}Pu) of 200-300 μBq or about an order of magnitude above that required to meet regulated dose controls. At the same time, guidance for implementation of internal dosimetry programs calls for the use of state-of-the-art technologies. The Lawrence Livermore National Laboratory (LLNL) has recently developed and demonstrated the viability of using Accelerator Mass Spectrometry (AMS) for ultra-trace plutonium isotope detection and measurement. AMS has a detection sensitivity of about 1 μBq, and avoids many of the disadvantages of using either conventional or other competing new technologies. It is no longer true that a technological shortfall exists in radiobioassay for plutonium urinalysis. For practical reasons, the availability of atom counting techniques such as AMS for low-level plutonium measurements cannot preclude the use of conventional methods within existing internal dosimetry programs. However, applications of this new technology will offer opportunity for managers of labs and nuclear facilities to selectively evaluate specific worker exposure scenarios and/or workplace practices, and help improve on overall worker protection and hazards management.

The new heavy-element AMS detection system for actinide analysis at LLNL was developed at the Center for Accelerator Mass Spectrometry (CAMS). Plutonium targets are prepared in an iron-niobium oxide matrix, and mounted on a sample wheel containing a negative ion spluttering source. Plutonium ions (PuO^-) are extracted from the source, and injected into the accelerator. Accelerated Pu^{5+} ions are then selectively filtered by a combination of magnetic and energy analysis, counted and identified on a gas-ionization detector. The final spectrometer includes 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 ~0.4 μBq of ^{239}Pu. We have also developed a fast-switching data-acquisition mode to average out the signal over several passes, and improve both precision and accuracy.

Several hundred analyses of plutonium have now been performed on artificial and human urine samples. AMS offers high efficiency, high rejection of interferences, and low susceptibility to matrix components for low-level detection of plutonium isotopes; it also provides for rapid measurement throughput (>50 unknowns per 24 h) and a large

linear dynamic range. Method validation has included participation in an interlaboratory exercise organized by the National Institute of Standards and Technology (NIST) (Figure 1). The results of this exercise clearly show that AMS technologies at LLNL are well suited for detection of μBq concentrations of ^{239}Pu in urine, and provides for an accurate and robust measurement technique.

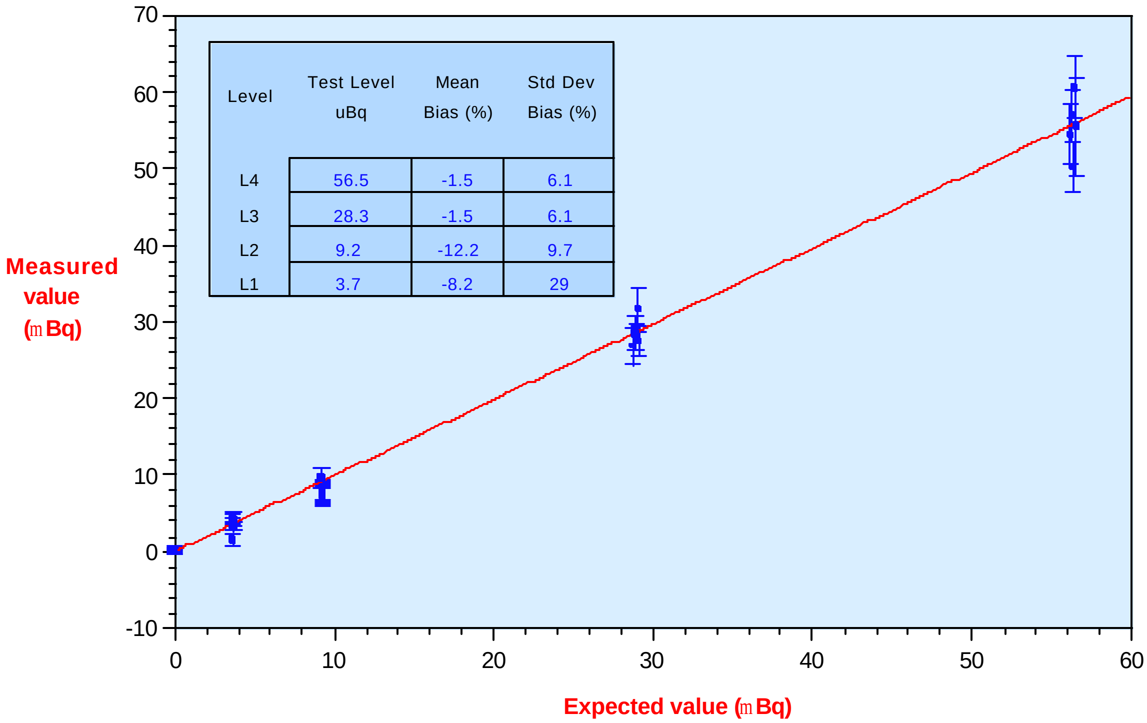

Figure 1. Results of a NIST interlaboratory exercise on low-level ^{239}Pu in synthetic urine.

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