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Validation Testing of Accelerator Mass Spectrometry Plutonium Bioassay Measurements Conducted at the Lawrence Livermore National Laboratory

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The Lawrence Livermore National Laboratory (LLNL) has recently developed an accelerator mass spectrometry (AMS) measurement system designed specifically for low sensitivity, high sample throughput, and robust measurements of actinide elements. The aim of this AMS research and development effort was to improve upon the existing bioassay measurement technologies being used to assess occupational, military, and public exposures to plutonium isotopes. Specifically, the newly developed AMS measurement system offers a number of potential operational and technological advantages for radiation protection programs compared to the more classical bioassay detection technologies that are based on alpha-spectrometry or competing newer measurement technologies such as fission track analysis. The U.S. National Institute of Technology and Standards (NIST) have independently verified the accuracy and reliability of the AMS system at LLNL for low-level plutonium bioassay measurements. In fact, results of plutonium-239 (^{239}Pu) and plutonium-240 (^{240}Pu) measurements in spiked, synthetic-urine samples met ANSI 13.30 criteria for both precision and accuracy at all test levels. Currently, this new measurement technology is being used to improve on the quality and reliability of plutonium exposure assessments in the Marshall Islands, as well as to verify potential workplace intakes of plutonium at LLNL. The AMS measurement technique is continuously tested and verified by analysis of quality-control (QC) samples prepared by researchers from the Oak Ridge National Laboratory (ORNL). These external quality-control samples were prepared as spike additions to aliquots of human urine collected from a low-background donor. The QC samples are normally offered with a declared ^{239}Pu concentration range of between 30 to 200 μBq per sample, and a $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio that approximates that observed in integrated worldwide-fallout deposition, i.e., ~ 0.2 . Based on ANSI 13.30 criteria, the combined measurement bias and precision of the analysis over the duration of the program has been -0.8% and 5.3% , respectively, for ^{239}Pu ; and $+5.2\%$ and 10.7% , respectively, for ^{240}Pu . Under routine operating conditions the AMS system background is equivalent to about ~ 1 μBq of ^{239}Pu and ~ 3 μBq of ^{240}Pu , far exceeding the requirements of the latest U.S. Department of Energy (DOE) regulation stated in *10 CFR 835* for *in-vitro* bioassay monitoring of plutonium isotopes. Based on these externally validated results and the operational experience of researchers at the LLNL Center for Accelerator Mass Spectrometry (CAMS), the AMS measurement system at LLNL is considered to be the *cutting edge* technology for the stated purpose, and vastly improves on the capabilities essential for monitoring compliance with the standards for occupational safety and risk management at LLNL, and elsewhere in the DOE complex.

