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Title:

Measurement of Actinides in Environmental Samples at Micro-Becquerel Levels by Accelerator Mass Spectrometry.

Abstract:

The need for ultra-sensitive actinide measurements continues to expand in the fields of environmental stewardship, nuclear isotope forensics, radiobioassay and environmental research. We have developed a heavy isotope accelerator mass spectrometry (AMS) system at Lawrence Livermore National Laboratory's Center for Accelerator Mass Spectrometry. The system was designed particularly for the measurement of actinide concentrations and isotopic ratios. A fast isotope switching capability has been incorporated in the system, allowing flexibility in isotope selection and for the quasi-continuous normalization to a reference isotope spike.

Initially, our utilization of the system has concentrated on the measurement of Pu isotopes. Under current operating conditions, background levels equivalent to $<10^6$ atoms are observed during routine ^{239}Pu and ^{240}Pu measurements. Measurements of samples containing 10^{13} ^{238}U atoms demonstrate that the system provides a ^{238}U rejection

factor of $\sim 10^7$. Recently, we have utilized the high dynamic range of the AMS system in measuring samples whose Pu contents ranged from $<10^6$ (background) to $>10^{11}$ Pu atoms. Measurements of known materials, combined with results from an externally organized intercomparison program, indicate that our ^{239}Pu measurements are accurate and precise down to the μBq level ($\sim 10^6$ atoms).

The development of the heavy isotope system was undertaken with particular interest in the measurement of environmental samples, including soils, sediments, waters, air filters, tissue samples, and human urine. The high rejection of interferences, including molecular interferences, and low susceptibility to matrix components, provided by the AMS technique are of particular relevance for such complex samples. These two factors significantly reduce demands on sample preparation chemistry for Pu analyses, allowing relatively simple, cost-effective procedures to be employed. The sensitivity of the AMS technique indicated by our Pu measurements suggests that the further optimization of sample preparation procedures will allow sample size reductions. When such sample preparation improvements are combined with the high sample throughput capabilities of our AMS system, the result will be a rapid and cost-effective measurement technique for actinides in environmental samples.

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