LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-SR-212001

Accelerator Mass Spectrometric (AMS) Measurements of Plutonium Activity Concentrations and $^{240}Pu/^{239}Pu$ Atom Ratios in Soil Extracts Supplied by the Carlsbad Environmental Monitoring & Research Center

T. F. Hamilton, T. A. Brown, A. A. Marchetti, R. E. Martinelli, and S. R. Kehl

Final Report

March 2005

This report was prepared under a Work for Others Agreement (Project L9537) between the United States Department of Energy and the State of Alaska, Department of Environmental Conservation.

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

TABLE OF CONTENTS

TABLES AND FIGURES

Accelerator Mass Spectrometric (AMS) measurements of plutonium activity concentrations and $^{240}Pu/^{239}Pu$ atom ratios in soil extracts supplied by the Carlsbad Environmental Monitoring & Research Center.

T. F. Hamilton[a], T.A. Brown[b], A.A. Marchetti[b], R.E. Martinelli[a], and S.R. Kehl[a]

[a]Environmental Science Division, Lawrence Livermore National Laboratory,
PO Box 808, Livermore, CA 94551-0808

[b]Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory,
PO Box 808, Livermore, CA 94551-0808

Final Report

1 March 2005

Summary

Plutonium-239 (^{239}Pu) and plutonium-239+240 ($^{239+240}Pu$) activities concentrations and $^{240}Pu/^{239}Pu$ atom ratios are reported for a series of chemically purified soil extracts received from the Carlsbad Environmental Monitoring & Research Center (CEMRC) in New Mexico. Samples were analyzed without further purification at the Lawrence Livermore National Laboratory (LLNL) using accelerator mass spectrometry (AMS). This report also includes a brief description of the AMS system and internal laboratory procedures used to ensure the quality and reliability of the measurement data.

.

1.0 Materials and Methods

Each sample (N=20) received for analysis consisted of a dried residue contained in a small quartz vial supplied by Lawrence Livermore National Laboratory (LLNL). The samples were supplied blind to LLNL researchers but were known to be prepared from large volume soil extracts after acid leaching and ion-exchange chromatography. At the request of LLNL researchers, 2 samples were prepared and identified as process blanks. All samples contained a known amount of plutonium-242 (^{242}Pu) (~ca. 3.5×10^{9} atoms) added as an isotope dilution spike. The spike information was used to calculate the amount of plutonium present in each sample split. These data should not be confused with any measurement data on the total amount of plutonium present in the original soil samples.

The dried sample residues were rehydrated In nitric acid and the material transferred to plastic centrifuge tubes. Plutonium was coprecipitated on a small quantity of Fe hydroxide, the precipitate transferred to a quartz vial and ignited at 800°C. The material was then packed into an aluminum AMS target with about 3 mg of added niobium (Nb) metal powder to enhance ion production. The analytical scheme included analysis of Fe-Nb target blanks (N=6) as well as Certified Reference Materials, (CRM) 128 (^{239}Pu/^{242}Pu = 0.9993 ± 0.0003) and CRM 138 (^{240}Pu/^{239}Pu = 0.0863 ± 0.0001), traceable to the U.S. National Institute of Standards and Technology (NIST).

The heavy ion AMS system used for low-level plutonium isotope measurements at the Center for Accelerator Mass Spectrometry (CAMS) at LLNL is shown in Figure1

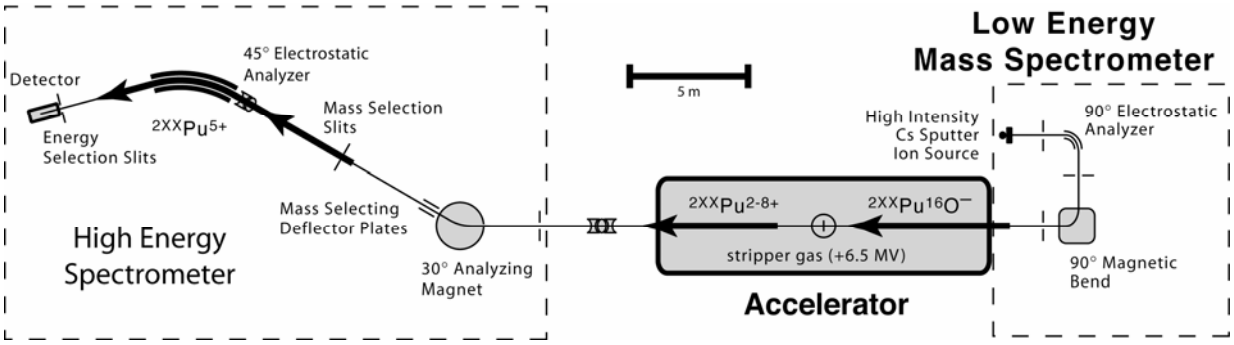

Figure 1. Heavy isotope accelerator mass spectrometry system at LLNL (Brown et al., 2004)

Negative plutonium ions (e.g., $^{239\text{-}244}$Pu^{16}O^{-}) are produced from sample targets using a cesium splutter source and injected into the accelerator through a 90° electrostatic analyzer. Negative ions entering the accelerator are stripped, converted to positive ions and accelerated into the high-energy spectrometer of the AMS system.

The high-energy spectrometer was designed to selectively transport 5+ ions to the detector using electrostatic deflection of desired ions through image slits coupled with fast mass switching of the low-energy

spectrometer and quasi-continuous normalization to the reference isotope (e.g., $^{242}Pu^{5+}$). Ions are counted on a two-anode, longitudinal field gas ionization detector with sufficient resolution to allow clean rejection of interfering ions at 4+ and lower charge states

2.0 RESULTS

^{239}Pu and $^{239+240}Pu$ activity concentrations and $^{240}Pu/^{239}Pu$ atom ratios in each of the 20 samples received from CEMRC are reported in Table 1. The results are expressed in units of μBq per sample (± 1 standard deviation) based on the ^{242}Pu spike information supplied to LLNL researchers. Some samples produced unusually high residual ion-currents but this phenomenon does not appear to have affected data quality.

The total measurement efficiency for the analysis was about $\sim 5\times10^{-5}$ with observed plutonium background concentrations based on Fe-Nb blank targets falling below $\sim 1\times10^{5}$ atoms. The Minimal Detectable Activity (MDA) for the measurement system at the time of analysis was estimated to be around 4×10^{-4} μBq and 2×10^{-3} μBq of ^{239}Pu and $^{239+240}Pu$, respectively.

Measurement data for AMS targets prepared from certified CRMs were all within stated uncertainties and showed no evidence of mass-dependent biases. For example, the mean weighted measured versus expected (theoretical) ratios of $^{239}Pu/^{242}Pu$ (N=12) in CRM 128 and $^{240}Pu/^{239}Pu$ (N=6) in CRM 138 were 1.006 ± 0.004 and 1.014 ± 0.014, respectively.

3.0 CONCLUSION

A series of purified soil extracts received from the Carlsbad Environmental Monitoring & Research Center were analyzed by accelerator mass spectrometry at the Lawrence Livermore National Laboratory. All test samples contained measurable quantities of plutonium in excess of that contained in quality control blank targets. Based on supplementary quality control analyses, it is expected that the accuracy and precision of all measurements performed under this agreement are within the stated uncertainties.

4.0 ACKNOWLEDGEMENT

The authors wish to thank Dr. Jeffrey Daniels for review of this report, and Jennifer Luna and Patricia Lopez for secretarial support.

REFERENCES

Brown, T.A., Marchetti, A.A., Martinelli, R.M., Cox, C.C., Knezovich, J.P., and Hamilton, T.F. (2004). Actinide measurements by accelerator mass spectrometry at Lawrence Livermore National Laboratory, Nucl. Instru. Meth. Phys. Res., **B223-224**, 788–796.

CONTACT INFORMATION

Dr. Terry Hamilton
Deputy Division Leader
Lawrence Livermore National Laboratory
PO Box 808, L-642
Livermore, CA 94551-0808
Telephone: (925) 422-6621
Fax: (925) 423-6785
Email: hamilton18@llnl.gov

Alternate
Dr. Tom Brown
Center for Accelerator Mass Spectrometry
Lawrence Livermore National Laboratory
PO Box 808, L-397
Livermore, CA 94551-0808
Telephone: (925) 423-8507
Fax: (925) 423-7884
Email: brown92@llnl.gov

Accelerator Mass Spectrometric (AMS) Measurements of Plutonium Activity Concentrations and $^{240}Pu/^{239}Pu$ Atom Ratios in Soil Extracts Supplied by the Carlsbad Environmental Monitoring & Research Center.

Table 1. ^{239}Pu and $^{239+240}Pu$ activity concentrations and $^{240}Pu/^{239}Pu$ atom ratios in purified soil extracts received from the Carlsbad Environmental Monitoring & Research Center (CEMRC).

LLNL Sample ID	Sample Description	^{239}Pu	$^{239+240}Pu$	$^{240}Pu/^{239}Pu$
		(mBq, sample received)		atom ratio
RFC631	CEMRC NMSU; 105851	1.364 ± 0.017	1.464 ± 0.018	0.074 ± 0.003
RFC632	CEMRC NMSU;105852	0.424 ± 0.010	0.456 ± 0.010	0.075 ± 0.006
RFC633	CEMRC NMSU; 105853	3.373 ± 0.038	3.609 ± 0.039	0.070 ± 0.002
RFC634	CEMRC NMSU; 105854	0.283 ± 0.008	0.315 ± 0.008	0.113 ± 0.009
RFC635	CEMRC NMSU; 105855	0.246 ± 0.006	0.270 ± 0.007	0.100 ± 0.008
RFC636	CEMRC NMSU; 1345_TB	0.034 ± 0.003	0.036 ± 0.003	0.084 ± 0.021
RFC637	CEMRC NMSU; 105856	0.364 ± 0.009	0.421 ± 0.010	0.156 ± 0.010
RFC638	CEMRC NMSU; 105857	0.317 ± 0.008	0.366 ± 0.009	0.154 ± 0.011
RFC639	CEMRC NMSU; 105858	0.510 ± 0.010	0.595 ± 0.011	0.168 ± 0.008
RFC640	CEMRC NMSU; 105859	1.447 ± 0.023	1.617 ± 0.025	0.118 ± 0.006
RFC641	CEMRC NMSU; 1346_LCS	1.593 ± 0.019	1.607 ± 0.019	0.009 ± 0.001
RFC642	CEMRC NMSU; 105860	0.447 ± 0.009	0.526 ± 0.010	0.177 ± 0.009
RFC643	CEMRC NMSU; 105861	0.508 ± 0.011	0.607 ± 0.012	0.193 ± 0.010
RFC644	CEMRC NMSU; 105862	0.727 ± 0.013	0.863 ± 0.014	0.186 ± 0.008
RFC645	CEMRC NMSU; 105863	0.258 ± 0.007	0.302 ± 0.007	0.172 ± 0.011
RFC646	CEMRC NMSU; 105864	0.589 ± 0.011	0.690 ± 0.012	0.173 ± 0.008
RFC647	CEMRC NMSU; 105865	0.318 ± 0.009	0.365 ± 0.010	0.146 ± 0.012
RFC648	CEMRC NMSU; 105866	0.371 ± 0.008	0.431 ± 0.008	0.164 ± 0.009
RFC649	CEMRC NMSU; 105867	0.418 ± 0.011	0.496 ± 0.012	0.188 ± 0.013
RFC650	CEMRC NMSU; 1348_TB	0.002 ± 0.001	0.002 ± 0.001	_ _