Preliminary Results of Uranium and Plutonium Efficiency Measurements Using a Thermal Ionization Cavity Source Interfaced with a Finnigan MAT 262 Mass Spectrometer

Riciputi, L.R., Ingeneri, K.B., (Oak Ridge National Laboratory, Oak Ridge, TN, 37831);
Hedberg, P.M.L. (Safeguards Analytical Laboratory, IAEA, Siebersdorf, Austria)

Evolving goals of safeguards environmental sampling of uranium and plutonium demand decreased sample sizes. Thermal ionization mass spectrometry (TIMS), with samples loaded on metallic filaments for vaporization and ionization before mass analysis, has been the mainstay analytical technique for precise isotope ratio measurement and, using isotope dilution, abundance determination. However, the ionization efficiency (ratio of element ionized to element available) of TIMS is poor for uranium and plutonium. Thus, improved ionization efficiency would increase analytical sensitivity and reduce sample size requirements, enhancing environmental sampling safeguards methods. We have developed a thermal ionization cavity (TIC) source that is interfaced with a commercial TIMS instrument (MAT 262) at the IAEA Safeguards Analytical Laboratory. The sample is loaded in a cavity bored into a metal (tungesten, W or rhenium, Re) rod, which is heated by electron impact. The source's confined geometry and ability to operate at much higher temperatures provide the potential for enhanced ionization efficiency compared to traditional TIMS sources.

Although source geometry and operating parameter optimization is just beginning, efficiency tests were undertaken to benchmark progress. Tests used individual resin beads containing ~10 ng of uranium. W cavities loaded with a single bead and enough graphite to fix the bead yielded efficiencies between 0.02 and 0.04%. Re cavities provided higher efficiencies (0.1 to 0.6%). Comparison experiments using single Re filaments yielded lower efficiencies (0.001 to 0.01%). Increased graphite in W cavities improved efficiency (0.03 to 0.08%) but changed temporal ionization behavior; similar behavior was not observed with Re. Addition of Re-C slurry in W cavities further improved efficiency (0.1-0.2%). Limited plutonium efficiency tests used resin beads loaded with 30pg of plutonium. Two W cavities yielded efficiencies of 0.08%; Re cavities ranged from 0.4 to 1.2%. Although preliminary, these results match or exceed many of the best reported TIMS efficiencies. Various observations, including maximized source lense voltages and deposit patterns on slits, suggest that transmission is far from optimized, and that ionization efficiency is significantly higher than effective efficiency measured in these tests.

Research sponsored by N.S.P.O., U.S.D.O.E. under contract DE-AC05-00OR22725 with Oak Ridge National Laboratory, managed and operated by UT-Battelle LLC.