Actinide Characterization using UHEXRF

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Actinide characterization is typically achieved using radiation based detectors. While these have been the mainstay for radioactive elements, the main drawback in using these instruments is the lack of specificity and selectivity and usually large backgrounds. The primary advantage of such instrumentation is the ability to detect the emitted radiation through container walls. Often times this advantage of through container wall measurement outweighs the disadvantages. X-ray fluorescence can be used to detect elements through container walls, albeit, typically those container walls are either polymer based or thin glass. The high energy of the actinide K lines (above 90 keV) offers the ability to both excite and detect ultrahigh energy X-rays through container walls. Ultrahigh energy XRF refers to XRF above 80 keV. In this work, ultrahigh energy X-ray fluorescence (UHEXRF) utilizes high energy excitation, 122 keV, to directly detect and quantify uranium and plutonium through container walls. The excitation beam of 122 keV is produced on the 6-ID-D beam line of the Advanced Photon Source at Argonne National Laboratory. This energy is sufficient to excite both the uranium Ka1 and plutonium Ka1 lines at 98.428 keV and 103.653 keV respectively. Several sample types have been measured with this approach which include: dried spot residues of actual nuclear spent fuel, plutonium contaminated soils and plutonium spiked synthetic spent fuel dried spot residues. In addition, a mock fuel rod containing model fuel pellets consisting of thorium oxide and uranium oxide with a stearic acid binder was also characterized. The thorium-uranium mixture provides a convenient model for the uranium-plutonium heavy metal ratio of actual spent fuel. The mock fuel rod used a typical Zircaloy tubing of around 600 micrometers thick. The plutonium dried spot samples were compared with measurements from a commercial conventional micro X-ray fluorescence instrument with the Pu La1 line at 14.279 keV. The Pu spiked samples were used to create a calibration of Pu intensity with known mass in the deposit to quantify the Pu in the spent fuel samples and contaminated soils. Use of the Advanced Photon Source, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science by Argonne National Laboratory, was supported by the U.S. DOE under Contract No. DE-AC02-06CH11357. The authors would like to acknowledge the support of the DOE Defense Nuclear Nonproliferation R&D for financial support of this research effort.