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K.G. McIntosh, G.J. Havrilla, R.F. Gilmore, M.K. Holland

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Evaluation of high resolution X-ray monochromatic wavelength dispersive X-ray fluorescence instrument for determining Pu and U in nuclear reprocessing streams

K.G. McIntosh[1a], G.J. Havrilla[1], R.F. Gilmore, Jr.[2], M.K. Holland[2]

[1]Los Alamos National Laboratory, Los Alamos, NM 87545 USA

[2]Savannah River National Laboratory, Aiken, SC 29808 USA

[a]Corresponding author, email address: kmcintosh@lanl.gov

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Abstract

An alpha-prototype benchtop instrument was developed and tested for measuring Pu and U in spent fuel dissolver tanks in a nuclear fuel reprocessing plant. The high resolution X-ray (hiRX) instrument was designed with the aim of providing a new capability to measure samples with high radiation background with selectivity, accuracy, and precision using ultra-low volumes (7 microliters) of liquid. The instrument was tested with specimens from the operational reprocessing facility at Savannah River National Laboratory. The results showed that bias was <6% for Pu nitrate process solution and <18% for Pu in U/HNO_3 matrix. Instrumental precision was <1 % RSD while between microcell repeatability was <6 % RSD. The limit of detection for Pu in HNO_3 matrix was $3x10^{-5}$ g/L (100 s, 35 W). This study demonstrates that hiRX technology has the potential to provide rapid and simultaneous quantification of Pu and U without sample preparation, making it suitable for process monitoring and materials control and accountability applications.

1.0 Introduction

The verification of special nuclear material inventories as part of nuclear safeguards is of great importance with the worldwide increase in the number and complexity of nuclear facilities used to meet growing energy needs. International safeguards enable the peaceful use of nuclear materials by minimizing the potential for undetected diversion of nuclear materials at fuel reprocessing and disposition facilities. This is accomplished by containment, surveillance, and nuclear material accountancy, in which analytical measurements help to establish or confirm the amount of fissile material present in the nuclear fuel reprocessing facility. A suite of tools is needed to accurately determine Pu and U mass in a variety of materials, from solids including fuel rods and vitrified waste, to reprocessing solutions. These solutions encompass a range of matrices and chemical forms as materials move through separations and purifications processes, and can therefore present an analytical challenge for prompt and accurate determinations of Pu and U concentration. The fact these materials are highly radioactive adds an additional layer of complexity to the analysis, as safety and worker dose have to be considered along with typical criteria such as sensitivity and specificity when selecting an appropriate method. Common