CORRELATION MEASUREMENTS WITH ^{252}Cf TO CHARACTERIZE FISSILE MATERIAL

J. K. Mattingly, J. T. Mihalczo, T. E. Valentine, J. A. Mullens, J. March-Leuba, T. Uckan
Instrumentation and Controls Division, Oak Ridge National Laboratory
P. O. Box 2008 MS6004, Oak Ridge, Tennessee 37831-6004

INTRODUCTION

Measurements using ^{252}Cf as a timed source of neutrons and gammas have in recent years undergone significant maturation. These methods use ^{252}Cf as an observable source of spontaneous fission neutrons and gammas in conjunction with one or more neutron- and/or gamma-sensitive detectors to measure the time-distribution of correlated detector counts following (a) an observed ^{252}Cf-fission event and/or (b) a counting event in another detector. Detection of ^{252}Cf spontaneous fission is frequently achieved via use of a small ionization chamber in which the ^{252}Cf is contained – in this case the timing of source emission events is *random*. However, one application subsequently described uses a neutron-absorbent "shutter" to modulate ^{252}Cf emissions to produce a neutron source with *deterministic* timing. Other applications, frequently termed *noise-analysis* measurements, transform the time-distributions to the frequency domain. Collectively, these correlation methods use ^{252}Cf to "excite" the fissile material and the response of the material is measured by an array of detectors and analyzed using standard time-correlation and/or frequency-analysis techniques.

In recent years numerous advances have been made in the application of these methods to in-situ, or field measurements directed at characterizing various configurations of fissile material in operational facilities.

SUMMARY OF APPLICATIONS

Californium-source-driven neutron noise analysis measurements were performed at the Missouri University Research Reactor under a variety of fuel configurations to obtain subcritical benchmarks that were subsequently used to infer *k*-effective and to estimate its bias from Monte Carlo calculations.[1] These measurements were used to reduce excess conservatism applied storage and transportation of fuel elements and consequently significantly reduce the costs of shipping fuel elements within the Savannah River Technology Center. Furthermore, similar ^{252}Cf-driven noise analysis measurements recently evaluated the subcriticality of an HEU storage array at the Oak Ridge Y-12 Plant for nuclear criticality safety purposes.[2]

A technique to measure the mass flow rate of ^{235}U in UF$_6$ gas was developed and implemented to monitor the down-blending of weapons-grade UF$_6$ to reactor-grade UF$_6$ in the Russian Federation in support of the HEU Transparency Agreement between the United States and Russia.[3] This technique uses a moderated ^{252}Cf-source modulated by a neutron-absorbent "shutter" to induce timed "bursts" of fission in the UF$_6$ gas. Delayed gammas from the fission fragments are detected downstream from the ^{252}Cf-source, and their timing and intensity (relative to the source modulation) are respectively used to determine the flow rate and concentration of ^{235}U in the gas. This method is also used to trace the feed-gas through the down-blending process.

Time-correlation measurements using a ^{252}Cf ionization chamber were applied at the former Oak Ridge Gaseous Diffusion Plant to estimate the spatial distribution, mass, and hydration of deposits of UO$_2$F$_2 \cdot n$H$_2$O in process piping.[4] The time-distribution of detector counts following ^{252}Cf-fission was used to discriminate between transmitted and scattered gammas and neutrons. Above 6 MeV, the total neutron cross-section of UO$_2$F$_2 \cdot n$H$_2$O is essentially independent of hydration (expressed in terms of the hydrogen to uranium atomic ratio, H/U) for H/U between 1 and 30, so the deposit thickness was estimated from high-energy (> 8 MeV) neutron transmission. Subsequently, the estimated thickness was used in conjunction with the measured gamma transmission to estimate the deposit density and hence its hydration. Scanning measurements were performed to estimate the azimuthal and axial distribution of thickness and density in order to reconstruct the spatial distribution of the deposit and to estimate its total mass. The results of these measurements were used by nuclear criticality safety specialists to estimate the subcriticality of the deposit and to formulate a safe plan for its removal.

At the Oak Ridge Y-12 Plant, active neutron interrogation measurements using the Nuclear Materials Identification System (NMIS) are routinely performed to identify receipts and inventory of nuclear weapons components, trainers, and other uranium items (e.g., metal castings) in support of nuclear materials control and accountability (NMC&A).[5] The NMIS uses small (~ 1 µg) ^{252}Cf ionization chambers and an array of fast plastic scintillators to measure the time-distribution of correlated counting events in each detector following (a) a ^{252}Cf

fission event and (b) a counting event in each other detector. These time-correlation "signatures," and their frequency-domain counterparts, are accumulated in real-time by the NMIS. The signatures have been used to verify the declared identity of receipts and items in storage using two different approaches. One approach matches the "fingerprint" formed by the collection of signatures measured from an "unknown" item to one member of a library of "reference" fingerprints acquired from items whose identity is unambiguously known. Another approach uses features of the signatures, e.g., gamma transmission and neutron production via induced fission, to estimate physical attributes, e.g., mass and enrichment, of the inspected item. The "fingerprint-matching" technique is applied to the initial verification of receipts and the periodic verification of inventory. The "attribute-estimation" technique has been applied to estimate the mass and enrichment of hundreds of items in storage. Both techniques are applied in-situ under conditions that often preclude the use of alternative methods, e.g., passive gamma spectrometry, due to the significant and variable gamma background that is frequently encountered in storage facilities.

CONCLUSIONS

Time-correlation and frequency-analysis methods using ^{252}Cf as an active neutron source have evolved to sufficient maturity to be of significant potential benefit to numerous real-world applications directed at characterizing fissile material. These applications include reactor physics measurements, nuclear criticality safety, nondestructive analysis and non-intrusive monitoring of enrichment (or de-enrichment) processes, and inventory-tracking for NMC&A.

REFERENCES

[1]T. E. Valentine, J. T. Mihalczo, R. C. Kryter, and V. C. Miller, "Subcritical Noise Analysis Measurements with Fresh and Spent Research Reactor Fuel Elements," ORNL/TM-13752, Oak Ridge National Laboratory, 1999.

[2]J. T. Mihalczo, T. E. Valentine, and L. D. Phillips, "Subcritical Measurements for an HEU Storage Vault at the Y-12 Plant Using the 252Cf-Source Driven Frequency-Analysis Method," *Nuclear Safety 38(1)*, January 1997, 10 – 24.

[3]J. March-Leuba, T. Uckan, J. Sumner, R. Vines, D. Powell, and E. Mastal, "Commissioning Measurements and Experience Obtained from the Installation of a Fissile Mass Flow Monitor in the Ural Electrochemical Integrated Plant (UEIP) in Novouralsk," 40th Annual Meeting of the Institute of Nuclear Materials Management, Phoenix, Arizona, July 1999.

[4]T. Uckan, M. S. Wyatt, J. T. Mihalczo, T. E. Valentine, J. A. Mullens, and T. F. Hannon, "Fissile Deposit Characterization at the Former Oak Ridge K-25 Gaseous Diffusion Plant by 252Cf-Source-Driven Measurements," ORNL/TM-13642, Oak Ridge National Laboratory, 1998.

[5]J. T. Mihalczo, J. K. Mattingly, T. E. Valentine, T. Uckan, L. G. Chiang, R. B. Perez, J. A. McEvers, J. A. Mullens, S. S. Hughes, C. R. Turner, M. Conger, "Field Use of NMIS at Oak Ridge," Y/LB-16,019, Oak Ridge Y-12 Plant, 1999.