TIME AND FREQUENCY DOMAIN MEASUREMENTS FOR PLUTONIUM METAL RINGS

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ABSTRACT

The $^{252}$Cf-source-driven noise analysis method has been used to measure the neutron multiplication factor for subcritical systems and to identify fissionable materials and configurations. These measurements with the moderator-ring configuration have shown that the $^{252}$Cf-source-driven noise methods, both in the time and frequency domain, can be used to identify and distinguish the $^{242}$Pu from the $^{239}$Pu ring. Signatures for each ring can be used to track and confirm the presence of these two types of plutonium rings. Because of the sensitivity of these signatures to mass, these measurements can also confirm the plutonium mass. These preliminary measurements have not explored the full range of capability of this method for this application.

ACKNOWLEDGMENTS

During the week of February 22, 1993, cooperative measurements between Oak Ridge National Laboratory (ORNL) and Los Alamos National Laboratory (LANL) were performed at LANL, TA-55, PF-4, Rooms 329 and 330, as part of an ongoing LANL-ORNL program. The success of these measurements required the cooperative efforts of NMT-3, NMT-8, HS-1, and JCI. In particular, we acknowledge the efforts, capabilities, and professional attitudes of J. D. Olivas, K. M. Vigil, and T. H. Abeyta of NMT-5; D. R. Zerwekh and L. R. Jaramillo of HS-1; along with S. D. Owens, R. A. Vigil, and E. Griego of NMT-3.

INTRODUCTION

The $^{252}$Cf-source-driven noise analysis method has been used to measure the neutron multiplication factor for subcritical systems and to provide identification of fissionable materials and configurations. In the latter application, the method provides measurements that can be used to confirm the materials present. This method is an active neutron and gamma ray interrogation technique where $^{252}$Cf fission neutrons and gamma rays are incident upon the sample of interest and where the subsequently produced neutrons and gamma rays, from induced fissions, are measured both in the time and frequency domain. The Fourier frequency analyzer transforms blocks of time domain pulses from the detectors to produce the various frequency spectra on-line. The frequency content of the signals is produced by the fluctuations of the induced fission chain multiplication and scattering processes in the sample of interest. Both time and
frequency analysis measurements were performed with two plutonium metal rings to investigate the use of these methods to identify the isotopic composition of the metal rings: one mainly of $^{239}\text{Pu}$ with a mass of 3162 g and the other mainly of $^{242}\text{Pu}$ with a mass of 3491 g.

Previous measurements with uranium metal have demonstrated the usefulness of time and frequency analysis measurements for distinguishing fissile from nonfissile uranium metal. The time domain measurements with the plutonium metal rings of different isotopic compositions are very similar to previous measurements with uranium metal. The frequency domain measurements were also performed because they provide a variety of signatures that can also be used to distinguish fissile from nonfissile plutonium. The recent use of frequency analysis signatures for verification of depleted uranium at the Oak Ridge Y-12 Plant showed some advantages over the time domain for the uranium metal application. This paper presents the results of these measurements performed at LANL, TA-55, PF4, during the week of February 22, 1993.

**DESCRIPTION OF THE METHOD**

The $^{252}\text{Cf}$-source-driven noise analysis method uses neutrons from a $^{252}\text{Cf}$ spontaneous fission source to induce fissions in the sample. These fissions produce secondary and scattered particles that are detected in one (for time domain measurements) or two (for frequency domain measurements) detectors. In the time domain measurement, the detector pulses are sorted in time according to their time of occurrence after $^{252}\text{Cf}$ fission. The time of occurrence of $^{252}\text{Cf}$ spontaneous fission is provided by an ionization chamber that has a $^{252}\text{Cf}$ deposit on one electrode. Thus, each spontaneous fission of the source produces an electronic pulse defining the time of $^{252}\text{Cf}$ fission. In the frequency domain measurements, signals from the $^{252}\text{Cf}$ source ionization chamber and two detectors are processed to obtain (1) auto power spectral densities, $G_{11}$, $G_{22}$, and $G_{33}$, where the subscripts designate detector 1 (the source ionization chamber), detector 2, and detector 3, and (2) cross-power spectral densities (CPSDs), $G_{12}$, $G_{13}$, and $G_{23}$. From these quantities, the coherences $\gamma_{12}^2$, $\gamma_{13}^2$, and $\gamma_{23}^2$ and a ratio of spectral densities $R = G_{12}^* G_{13} / G_{11} G_{23}$ are obtained. The fluctuations of the fission chain multiplication and scattering processes in the sample produce a fluctuating signal that has a variety of frequency components that can be resolved with Fourier analysis. The frequency spectrum of the signals is used for verification in much the same way as Fourier analysis is used in voice identification. These time and frequency analysis signatures have a variety of characteristics that can be used to distinguish fissile from nonfissile material. They can be used to identify material with inherent sources since some of these parameters depend on both $^{252}\text{Cf}$ and the inherent source strength and some only depend on the $^{252}\text{Cf}$ source. Some of the measured parameters are also dependent on background, while others are not. The independence of background of some of the signatures makes this method very useful for warehouse or storage applications in close proximity to other material. In fact, measurements have been performed with hundreds of units in containers within 6 m of the verification station without affecting the verification procedure. This independence of the background permits the identification of secondaries through the first stage of dismantlement because the presence of other neutron sources from plutonium do not change some of the signatures.

**SOURCES AND DETECTORS**

The $^{252}\text{Cf}$ sources used for this measurement were 0.365 in. in diameter, and a sketch is shown in Fig. 1. The californium sources used in these measurements had intensities of $-4.5 \times 10^4$ and $1.145 \times 10^6$ fissions/s. The detector for the time domain measurement was a liquid scintillator (NE 213) with pulse shape discrimination. This 1.7-in.-OD, 1.7-in.-thick detector was surrounded by 0.125-in.-thick lead on the front face and sides. For the frequency analysis measurements, 1.7-in.-OD, 2.0-in.-thick plastic scintillators with 0.125-in.-thick lead on the front face and sides were used.

**PREMEASUREMENT CALCULATIONS USING MONTE CARLO TRANSPORT THEORY METHODS**

Previous measurements with uranium metal showed that the difference between depleted and enriched uranium metal could be enhanced by the use of polyethylene reflector around the uranium metal parts. This results from the fact that, for unmoderated fissile material, the polyethylene adds a longer time component to the decay of the neutron population that results from thermalized neutrons in the polyethylene returning to the uranium metal,
producing fission on a longer time scale. This effect was evaluated for the $^{239}$Pu ring by calculating the time domain results using the KENO-NR Monte Carlo neutron transport code for a ring with a central polyethylene plug and both 1- and 2-in.-thick polyethylene reflectors on the top and bottom of the ring. No radial reflector was used in these measurements because the detector was placed adjacent to the radial surface of the plutonium ring. The premeasurement calculations showed that the polyethylene configuration of Fig. 1 was sufficient for this enhancement, and thus these measurements were performed with this thickness of polyethylene. This effect was also confirmed by MCNP-DSP calculations.

**MATERIAL DESCRIPTION**

The plutonium isotopic compositions of the two metal rings are given in Table 1. The polyethylene for these measurements had an average density of 0.916 g/cm$^3$. The stainless steel (SS 304) disc (4.62 in. OD, 0.87 in. thick) had a density of 8.87 g/cm$^3$. The top discs and those internal to the ring had 0.40-in.-diam axial holes to accommodate the $^{252}$Cf source. The outside diameter of the large polyethylene pieces of Fig. 1 was 4.62 in. and that for the inner discs was 2.5 in. The thickness of the top polyethylene disc was 0.97 in., while the lower large polyethylene disc had a thickness of 1.0 in. at the outside and 0.5 in. on the inside. The aluminum ring under the plutonium metal dimensions were: 0.25 in. thick, 3.25 in. ID, and 4.75 in. OD.

<table>
<thead>
<tr>
<th>Table 1. Characteristics of plutonium Metal Rings</th>
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<tbody>
<tr>
<td><strong>Quantity</strong></td>
</tr>
<tr>
<td>Total Mass, g</td>
</tr>
<tr>
<td>plutonium Isotopics, %</td>
</tr>
<tr>
<td>238</td>
</tr>
<tr>
<td>239</td>
</tr>
<tr>
<td>240</td>
</tr>
<tr>
<td>241</td>
</tr>
<tr>
<td>242</td>
</tr>
<tr>
<td>Outside Diameter, cm</td>
</tr>
<tr>
<td>Inside Diameter, cm</td>
</tr>
<tr>
<td>Height, cm</td>
</tr>
<tr>
<td>Method of Fabrication</td>
</tr>
</tbody>
</table>

**MEASUREMENT CONFIGURATION**

For contamination control, both rings were wrapped in a double-thick (~2-in.-wide) plastic material with overlap such that there were four layers of 0.005-in.-thick plastic around the ring. Each ring was then bagged out of the glove box in a large, 0.020 in.-thick plastic bag-out-bag, such that the ring could be configured with polyethylene reflector as shown in Fig. 1. A polyethylene plug with an axial hole was inserted inside the ring to position the source. The stainless steel disc located on top of the polyethylene reflector, as shown in Fig. 1, compressed the plastic wrap and bag-out-bag in a reproducible way. The configuration was placed on a 1/16-in.-thick steel table ~24 in. from the wall and 33 in. from the concrete floor. The $^{252}$Cf source was positioned in the center of the central polyethylene plug, as shown in Fig. 1. The height from the top of the steel plate was 5.0 in. above the steel table for the $^{242}$Pu ring and 5.06 in. for $^{239}$Pu ring. The $^{239}$Pu ring has a lower density than the other ring since it has approximately the same dimensions but less plutonium. The detectors were placed adjacent to the radial surface of the bagged plutonium ring at the vertical center of the ring.

**TIME DOMAIN MEASUREMENTS**

The neutron and gamma ray detector system utilized a liquid scintillator (NE 213, 1.7-in. diam) with pulse shape discrimination for neutron-gamma ray separation. The threshold for neutron detection was 1.5 MeV, and the probability that a neutron incident on the detector is counted was 0.20. The neutron-gamma ray discrimination was not perfect, so a small number of gamma ray counts appear in the neutron channel, and some neutron counts appear in the gamma ray channel. This is observable in the rising portion of the neutron time distribution. The time distributions of neutrons at the detector after $^{252}$Cf fission (correlated to $^{252}$Cf fission) for both rings are compared in Fig. 2a. The time scale of the abscissa has been adjusted so that the time of the $^{252}$Cf fission, plus the gamma ray flight time to the detector, is plotted at 25 ns so that the rising portion of the signal is displayed. The comparison for longer times is given in Fig. 2b. To confirm that the time domain measurements were reproducible, three different time bin intervals were used. The distributions consist of: directly transmitted particles that comprise the rising portion of the signal, scattered particles, and particles from the induced fission chain multiplication process in the ring. Because the $^{242}$Pu metal ring is composed primarily of a threshold fission isotope, it has significantly less induced fission, and the time distributions terminate ~20 ns after the $^{252}$Cf fission (Fig. 2b). For the $^{239}$Pu ring, the distribution persists...
for a long time (>300 ns). This effect is the result of the polyethylene moderator, which slows down neutrons, causing fission in the $^{239}$Pu ring. The $^{252}$Cf source for the time domain measurements contained 0.073 $\mu$g of $^{252}$Cf and had a spontaneous fission rate of $\sim$45,000 fissions/s, whereas the spontaneous fission rate of the material of the $^{242}$Pu ring ($2.6 \times 10^6$ fissions/s total for the $^{242}$Pu ring) was a factor of 58 higher than the $^{252}$Cf source fission rate. Despite the high inherent spontaneous fission rate of the $^{242}$Pu ring, which results in neutrons at the detectors not correlated with $^{252}$Cf fission, these measurements were performed in $\sim$15 min. One time domain measurement was performed in 5 min and indicated that this was sufficient for distinguishing the $^{242}$Pu ring from the $^{239}$Pu ring. This measurement time could be reduced at least a factor of 10 with larger sources and currently available processors. These time domain measurements showed excellent discrimination against neutrons from events other than $^{252}$Cf-induced fission, because those neutrons and gamma rays from fission chains initiated by other neutron sources are not time correlated with $^{252}$Cf fission and only produce random background. The peak to background ratio for the measurements with the $^{242}$Pu ring was 66, while that for the $^{239}$Pu ring was 867. The peak to background ratio is related to the ratio of induced fissions from $^{252}$Cf neutrons to the inherent spontaneous fission of the ring material. The minimum time-correlated count rate in these measurements was $\sim$5000 counts per minute for the $^{242}$Pu ring. The characteristics of these time domain measured data are sufficiently different that they can be used to distinguish the $^{242}$Pu ring from the $^{239}$Pu ring.

**FREQUENCY DOMAIN MEASUREMENTS**

The frequency domain measurements were performed at a digitizing rate of 36 MHz, much less than that required for measurement of the full frequency content of the signal for these systems. However, prior measurements over limited frequency range have been shown to be useful for determining isotopic composition of uranium metal. The two detectors for the frequency domain measurements were located adjacent to the outer surface of the bagged plutonium ring, 180° apart. They were plastic scintillators (1.7 in. diam) detecting both gamma rays and neutrons without distinction. The threshold for neutron detection was $-0.5$ MeV, and the probability that a neutron hitting the front face of the detector is counted was 0.40 for both detectors.

Frequency domain measurements were performed for both rings with and without a $^{252}$Cf fission source. In this paper, only data for a measurement with each ring are presented for comparison. The CPSDs, $G_{12}$ and $G_{13}$, and coherence, $\gamma_{12}$ and $\gamma_{13}$, are related to the fraction of common information in two signals between a detector signal and the $^{252}$Cf source ionization chamber. In the time domain measurements, the time distribution of counts in a detector was measured with respect to $^{252}$Cf fission. In the frequency domain measurements, the detector channels are designated as 2 and 3 is and the signal from the $^{252}$Cf ionization chamber processed as channel 1. The CPSDs, $G_{12}$ and $G_{13}$, and the coherence between detectors and the $^{252}$Cf source are the result of the common information in the two signals, which arises from their correlation through the fission chain multiplication process. A comparison of the CPSD and coherence between one detector and the $^{252}$Cf source is given in Figs. 3 and 4. These functions correlating the other detector with the source are similar and not shown. These quantities are clearly different and well beyond the statistical uncertainties. The difference in these quantities can be used to distinguish the rings. Other frequency domain functions also show differences clearly related to the ring materials.

**SUMMARY**

These measurements with the moderator-ring configuration of Fig. 1 have shown that the $^{252}$Cf-source-driven noise methods, both in the time and frequency domain, can be used to identify these rings and thus distinguish the $^{242}$Pu from the $^{239}$Pu ring. The signatures for each ring can be used to track and confirm the presence of these two types of plutonium material. These signatures can be used to confirm the plutonium mass because of their sensitivity to mass. These preliminary measurements have not explored the full range of capability of this method for this application.

**REFERENCES**


Figure 1. Sketch of the Measurement Material Configuration Without Detectors

Figure 2a. Comparison of Time Domain Signatures for Both Rings up to 35 ns After Californium Fission (t = 0 for Californium fission plotted at 25 ns)

Figure 2b. Comparison of Time Domain Signatures for Both Rings up to 180 ns

Figure 3. The Cross Power Spectral Density Between One Detector and the Californium Source as a function of frequency for the plutonium Metal Rings

Figure 4. The Coherence Between One Detector and the Californium Source as a function of Frequency for the plutonium Metal Rings