LA-UR- 10-03849

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Intended for: 51st INMM Annual Meeting
Baltimore, MD
July 11-15, 2010
(Full Paper)

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Initial Measurements of BN-350 Spent Fuel in Dry Storage Casks using the Dual Slab Verification Detector

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ABSTRACT

The Dual Slab Verification Detector (DSVD) has been developed, built, and characterized by Los Alamos National Laboratory in cooperation with the International Atomic Energy Agency (IAEA) as part of the dry storage safeguards system for the spent fuel from the BN-350 fast reactor. The detector consists of two rows of ^{3}He tubes embedded in a slab of polyethylene which has been designed to be placed on the outer surface of the dry storage cask. By performing DSVD measurements at several different locations around the outer surface of the DUC, a signature "fingerprint" can be established for each DUC based on the neutron flux emanating from inside the dry storage cask. The neutron fingerprint for each individual DUC will be dependent upon the spatial distribution of nuclear material within the cask, thus making it sensitive to the removal of a certain amount of material from the cask. An initial set of DSVD measurements have been performed on the first set of dry storage casks that have been loaded with canisters of spent fuel and moved onto the dry storage pad to both establish an initial fingerprint for these casks as well as to quantify systematic uncertainties associated with these measurements. The results from these measurements will be presented and compared with the expected results that were determined based on MCNPX simulations of the dry storage facility.

INTRODUCTION

The ability to safeguard spent nuclear fuel is strongly dependent on the technical capabilities of establishing and maintaining continuity of knowledge (COK) of the spent fuel as it is released from the reactor core and either reprocessed or packaged and stored at a storage facility. While the maintenance of COK is often done using continuous containment and surveillance (C/S) on the spent fuel, it is important that the measurement capabilities exist to re-establish the COK in the event of a significant gap in the continuous C/S by performing measurements that independently confirm the presence and content of Plutonium (Pu) in the spent fuel. The types of non-destructive assay (NDA) measurements that can be performed on the spent fuel are strongly dependent on the type of spent fuel that is being safeguarded as well as the location in which the spent fuel is being stored.

The BN-350 Spent Fuel Disposition Project was initiated to improve the safeguards and security of the spent nuclear fuel from the BN-350 fast-breeder reactor and was developed cooperatively to meet the requirements of the International Atomic Energy Agency (IAEA) as well as the terms of the 1993 CTR and MPC&A Implementing Agreements. The unique characteristics of fuel from the BN-350 fast-breeder reactor have allowed for the development of an integrated safeguards measurement program to inventory, monitor, and if necessary, re-verify Pu content of the spent fuel throughout the lifetime of the project. This approach includes the development of a safeguards measurement program to establish and maintain the COK on the spent fuel during the repackaging and eventual relocation of the spent-fuel assemblies to a long-term storage site.

As part of the safeguards measurement program, the Pu content of every spent-fuel assembly from the BN-350 reactor was directly measured and characterized while the spent-fuel assemblies were being stored in the spent-fuel pond at the BN-350 facility using the Spent Fuel Coincidence Counter (SFCC) [1]. Upon completion of the initial inventory of the Pu content of the individual spent-fuel assemblies, the assemblies were repackaged into welded steel canisters that were filled with inert argon gas and held either four or six individual spent-fuel assemblies depending on the type of assembly that was being packaged. This repackaging of the spent-fuel assemblies was performed in order to improve the stability of the spent-fuel assemblies for long-term storage and increase the proliferation resistance of the spent fuel. To maintain the capability of verifying the presence of the spent-fuel assemblies inside the welded steel canisters, measurements were performed on the canisters using the Spent Fuel Attribute Monitor (SPAM), which was a neutron coincidence counter very similar in design to the SFCC, but with a larger operational volume to accommodate the canisters. [2] The analysis of the neutron coincidence data from the measurements of the welded steel canisters using SPAM yielded a Canister Attribute (CA) that was shown to be proportional to the total amount of Pu mass that was present inside the canister and comparable to the CA values that were calculated based on the previous SFCC measurements. [3]

The next step in securing the spent fuel from the BN-350 reactor is the packaging of the steel canisters into Dual-Use Casks (DUCs) and the transportation of the DUCs to a remote dry storage site. The Dual Slab Verification Detector (DSVD) has been developed, built, and deployed by Los Alamos National Laboratory in cooperation with the IAEA to perform measurements of the neutron flux emanating from inside the DUC at several locations around each cask to establish a neutron "fingerprint" that is sensitive to the contents of the cask. An initial series of DSVD measurements have been performed on dry storage casks that have been loaded with canisters of spent fuel and moved onto a storage pad at the BN-350 facility to both establish an initial fingerprint for these casks as well as to quantify systematic uncertainties associated with these measurements. The results from these measurements will be presented in this paper and compared with the expected results that were determined based on MCNPX simulations of the dry storage facility.

BN-350 DRY STORAGE SAFEGUARDS

A single DUC holds eight spent-fuel canisters, with one canister placed in the center of the cask, and the remaining seven canisters placed in a circular pattern surrounding the central canister. The walls of each DUC are 45 cm thick and contain multiple layers of steel and concrete as indicated in Figure 1a). The composition and thickness of the DUC's walls significantly reduces the amount of radiation flux that passes through the outer surface of the DUC from the spent fuel. Based on neutron transport calculations that were performed using MCNPX [4], the attenuation properties of the walls of the DUC, along with the spatial distribution of the canisters throughout the interior of the DUC, make it impractical to directly verify the presence of Pu within the DUC through any type of neutron coincidence measurements that are made on the outer surface of the DUC.

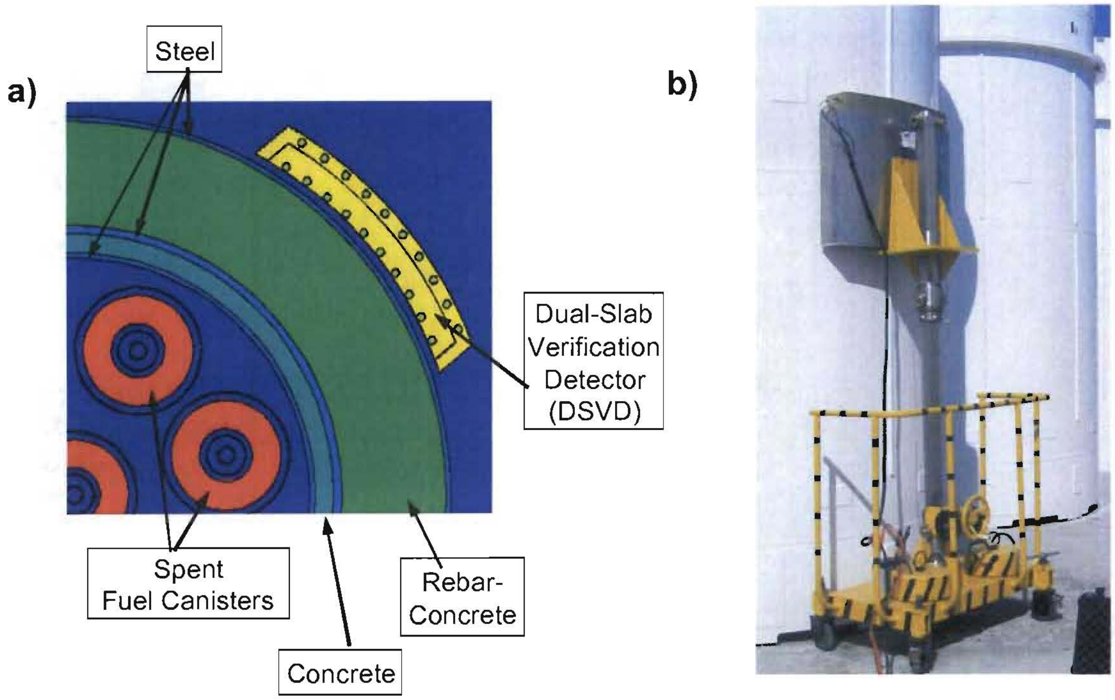

Figure 1a) Top view of the dual-use cask with the dual-slab verification detector. The numbers over the two spent-fuel canisters indicate the angular position within the DUC. **b)** Picture of DSVD detector in position alongside a BN-350 spent fuel dry storage cask.

Instead of directly verifying the plutonium content in the spent fuel as had been done using the SFCC and SPAM detectors, an indirect verification technique has been developed that uses passive neutron measurements at four specific locations around the exterior of the DUC to establish a signature fingerprint for each DUC that is sensitive to the presence of each individual canister within the DUC. Similar verification techniques have been previously demonstrated by Abhold, et al. [5] and Turner and Swinhoe [6]. By maintaining an archived set of fingerprint data for each DUC and comparing a current set of fingerprint measurements with the archived data, a determination can be made as to whether or not the contents of an individual DUC have changed since the last verification measurement. In the case of the BN-350 spent fuel, the technique must be sensitive to the removal of approximately one single canister of spent fuel from a DUC. To achieve this level of sensitivity, the design of the DSVD was optimized based on MCNPX calculations of the transport of neutrons from the eight spent-fuel canisters to the surface of the DUC.

The DSVD consists of two rows of 10 ^{3}He proportional counters (4 atm pressure, 2.54 cm diameter, 61 cm long) embedded in polyethylene with the inner and outer rows of counters separated by a 1.5 mm thick sheet of cadmium. The detector is 65.9 cm tall and 82.6 cm wide with the inner surface of the front polyethylene slab curved with the same radius as the outer surface of the DUC so as to put all of the proportional counters in the front slab at the same distance away from the center of the DUC. A picture of the DSVD in position to make a measurement on a DUC is shown in Figure 1b). Because the neutrons travel through a large amount of moderating material before encountering the DSVD, a minimal amount of polyethylene was placed between the DUC and the inner slab counters in order to optimize the number of neutrons that pass through the counters. The total polyethylene thickness of the inner slab is 8.2 cm while the thickness of the back slab is 4.5 cm.

The inner row of ^{3}He counters is designed to be mostly sensitive to the neutron flux from the DUC of interest, while the outer row of counters are sensitive to the background neutrons from the neighboring DUCs. This arrangement allows for a method for suppressing background neutrons and determining the neutron flux emitted by the DUC of interest. The measured neutron flux from the DUC of interest in the DSVD (φ_i) is given by

$$\varphi_i = \frac{(C_i - C_{ib}) - R_f(C_o - C_{ob})}{\varepsilon_{ii} - R_f\varepsilon_{oi}} = \frac{C_{cor}}{\varepsilon_{ii} - R_f\varepsilon_{oi}} \quad (1)$$

where C_i and C_o are the measured count rates in the inner and outer slabs, respectively, C_{ib} and C_{ob} are the measured ambient background count rates in the inner and outer slabs, respectively, ε_{ii} and ε_{oi} are the detection efficiencies of the inner and outer slabs, respectively, for neutrons from the DUC of interest, and R_f is given by

$$R_f = \frac{\varepsilon_{io}}{\varepsilon_{oo}} \quad (2)$$

where ε_{io} and ε_{oo} are the detection efficiencies of the inner and outer slabs, respectively, for neutrons from outside the DUC of interest [5]. Because this technique is intended as a means to verify that no changes to the contents of a dry storage cask has occurred since the last inspection of the casks, the main parameter of interest is the relative difference between the measured flux at each location during the current set of measurements, φ_{iv}, and the measured flux during the previous set of measurements, φ_{ib}. The relative difference in these two sets of measurements, f, is given by:

$$f = \frac{\varphi_{iv} - \varphi_{ib}}{\varphi_{ib}} = \frac{\varphi_{iv}}{\varphi_{ib}} - 1 = \frac{C_{cor,v}}{C_{cor,b}} \cdot \frac{(\varepsilon_{ii} - R_f\varepsilon_{oi})_b}{(\varepsilon_{ii} - R_f\varepsilon_{oi})_v} - 1 \quad (3)$$

If the performance of the detector were perfectly stable as a function of time, then ε_{ii}, ε_{oi}, and R_f would be constant as a function of time, which implies that

$$(\varepsilon_{ii} - R_f\varepsilon_{oi})_b = (\varepsilon_{ii} - R_f\varepsilon_{oi})_v \Rightarrow$$

$$f = \frac{C_{cor,v}}{C_{cor.b}} - 1$$

Because it is not realistic for the performance of the DSVD to be perfectly stable as a function of time, it is necessary to perform calibration checks to determine how the performance of the detector has changed since the last measurements were performed. By making measurements on the same known neutron source before and after each verification measurement campaign, one can eliminate the need to determine the absolute detector efficiencies and simply determine the performance of the detector relative to the previous baseline measurements. Hence the fingerprint equation can be written as

$$f = \frac{C_{cor,v}}{C_{cor,b}} \cdot CF - 1 \quad (4)$$

where CF is the correction factor which accounts for the change in the response of the DSVD to the known neutron source between the previous baseline measurement and the current verification measurement. It should be noted the corrected count rate for the verification measurement ($C_{cor,v}$) in Equation 4 is decay corrected based on an average set of Pu isotopics that have been calculated for the BN-350 spent fuel assemblies using a combination of the the REBUS-3 fuel-cycle analysis code [7] and the ORIGEN point depletion code [8].

DSVD MEASUREMENTS OF BN-350 SPENT FUEL

DSVD verification measurements of the BN-350 spent fuel in dry storage casks occurred while the casks were being staged for transport at the storage pad that is located near the BN-350 facility in Aqtau, Kazakhstan. The measurements were performed during two separate measurement campaigns that were separated by several months in time. During the initial measurement campaign, calibration and ambient background measurements were performed inside the BN-350 reactor over the spent fuel pond. These measurements consisted of a background measurement with no source present, and a normalization measurement using an 11.1 Ci AmLi source that is stored on site. The ambient background count rates that were measured during the first campaign were 2.47 ± 0.1 cts/s and 4.07 ± 0.1 cts/s for the inner and outer slab detectors, respectively. These measured rates were assumed to be the background rates that would be encountered at the dry storage pad, and were subsequently used to perform background subtraction for both the normalization measurements with the AmLi source and the verification measurements at the dry storage pad. Due to time constraints that were encountered during the first measurement campaign, no ambient background measurements were possible at the dry storage pad. For the second measurement campaign, ambient background measurements were performed outside near the dry storage pad before and after the verification measurements were performed. The average measured ambient background count rates for both the inner and outer slabs for the second measurement campaign was 6.4 ± 0.1 cts/s.

Two normalization measurements were performed during the first measurement campaign with an AmLi source located along the central axis of the DSVD at 30 cm from the inner surface of the DSVD and then 30 cm from the outer surface of the DSVD. The location of the AmLi source relative the detector was the same as the initial characterization measurements that were performed at LANL [9]. The source strength of the AmLi source used in the measurements at the BN-350 was calculated to be 6 x 10^5 n/s ± 10% based on the manufacturer's certificate. While normalization measurements were only performed prior to the verification measurements during the first measurement campaign, normalization measurements were performed at the beginning and end of each day during the second measurement campaign. To improve the systematic uncertainty associated with the normalization measurements, a fixture was used during the second measurement campaign to fix the location of the AmLi source at the appropriate position relative to the inner and outer surface of the DSVD in order to improve the repeatability of the measurements. The amount of variation that was observed in the normalization measurements throughout the first and second measurement campaigns can be observed in Figure 2 which shows a) ε_{ii}, b) ε_{oi}, and c) R_f as a function of the measurement number. Based on a series of measurements that were performed with the AmLi source removed from the fixture, rotated by 90 degrees, and replaced into the fixture, the estimated systematic error associated with the normalization measurements for ε_{ii} and ε_{oi} was 0.3%

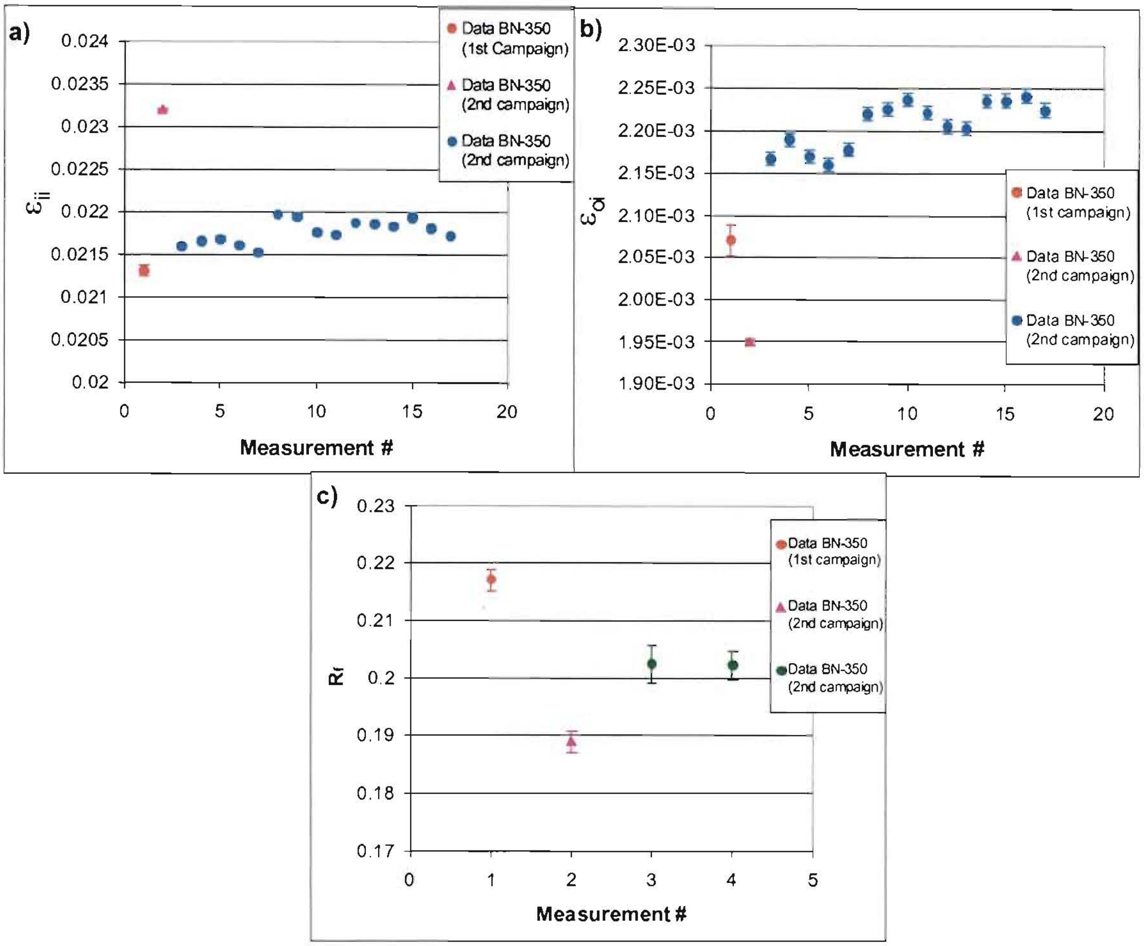

Figure 2. Results from the AmLi normalization measurements for **a)** ε_{ii} **b)** ε_{oi} and **c)** R_f from the first two verification measurement campaigns at the BN-350. The pink triangles indicate the normalization measurements that were performed during the second campaign without the AmLi source fixture. The error bars shown in **a) - c)** are due to statistics only and does not include the uncertainty in the source strength.

and 0.5%, respectively.

During the first measurement campaign, the casks were located on the storage pad in a single row with a 1 meter separation between neighboring casks. To aid in positioning the DSVD on the side of the cask, the outer surface of each cask has 4 unique markings at 90 degree intervals that are fixed in relationship to the interior structure of the DUC. By aligning the placement of the DSVD on the outer surface of the DUC with these markings, the amount of uncertainty associated with the positioning and repositioning of the DSVD on the DUC was reduced. To further aid in reproducing future measurements on each DUC, a fiducial mark was made on the cask where the lower-left corner of the DSVD sits during the measurements. To estimate the systematic uncertainty associated with repositioning the DSVD at the four locations around the outside of the cask, a series of measurements were performed where the DSVD was moved from its measurement location by 20 mm in all four directions (up, down, left and right). The effect that was observed in the count rates of the inner and outer slabs with the movement of the DSVD was approximately 1%. It is estimated that the error in repositioning the DSVD at the same spot of the DUC should be less than 20 mm because of the markings that are present on each cask. Because it was not possible to perform daily normalization measurements using the AmLi source during the first measurement campaign, a set of two daily confirmation measurements were performed during the course of the measurements which involved repeating the measurement at one position on a cask on two consecutive days. Based on taking the standard deviation of the repeated measurements, the estimated systematic uncertainty in the inner slab and outer slab measurements due to repositioning the detector at a given location on the cask are ± 1% and ± 2 %, respectively.

Based on the known loading of the casks, the construction of the casks, the orientation of the casks on the dry storage pad, and MCNPX simulations of the dry storage measurement scenario, a series of calculations were performed to determine the expected count rates for the inner and outer slab for the casks that were measured during the first campaign. Figure 3 shows the measured count rates (pink squares) and the calculated count rates (blue diamonds) for the inner slab (top row) and outer slab (bottom row) detectors for two spent fuel casks. Both the measured and calculated count rates were normalized relative to the average measured count rate in either the inner or outer slab detector

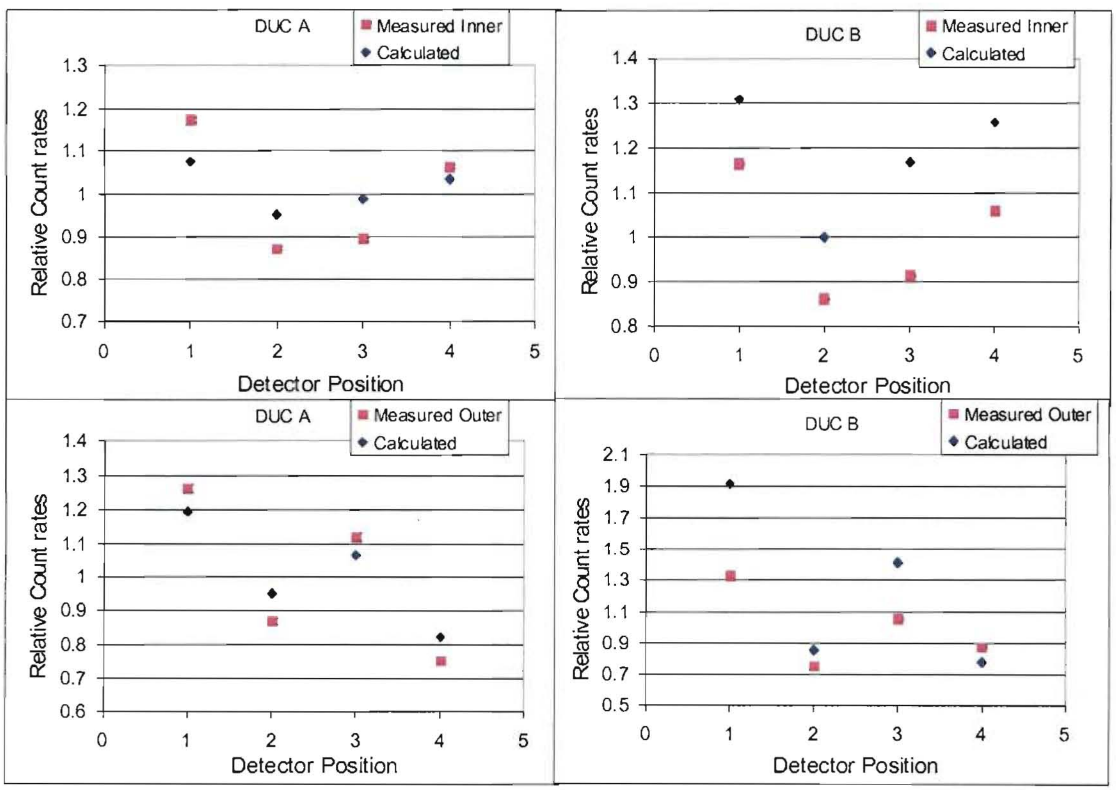

Figure 3. Comparison between the measured relative count rates in the inner (top row) and outer (bottom row) slab detectors for two spent fuel casks labeled DUC A (left column) and DUC B (right column) and the calculated relative count rates that are based on Monte Carlo simulations of the measurement scenario. Both the measured and calculated count rates were normalized relative to the average measured count rate for that particular slab detector and spent fuel cask that was measured.

for that particular spent fuel cask. For the two casks shown in Fig. 3, a reasonable qualitative agreement exists between the calculated and measured count rates for both the inner and outer slab detectors. While the agreement in the overall count rates shown in Fig. 3 between the calculated and measured count rates is typical for the other casks measured in the first campaign, larger discrepancies have been observed between the calculated and measured fingerprint patterns for a number of casks that were measured in the first campaign. Work is currently ongoing to improve the agreement between the measured and simulated fingerprints for cask measured during the first campaign.

During the second measurement campaign, a series of sensitivity measurements were performed with the DSVD fixed to a specific measurement position on the side of a cask and set up to continuously acquire data while individual canisters were loaded into an empty cask. This provided an opportunity to not only determine how each canister contributes to the count rate observed within the DSVD, but also to benchmark the MCNPX calculations that were performed to determine the performance of the detector. Figure 4 shows the measured and calculated total count rates in both the inner and outer slabs as each canister was introduced into the cask. While a relatively good agreement exists between the measured and calculated count rates for the inner slab for the first 5 – 6 canisters that were introduced into the cask, the measured inner count rates for the last two canisters are much larger than the calculated rates. It should be noted that the first 5 canisters that were placed in the cask were placed in positions that were furthest away from the DSVD location. The next two canisters were placed in the positions that were closest to the DSVD, while the last canister was placed in the center of the cask. Work is currently ongoing to understand the large discrepancies between the calculated and measured count rates when the last three canisters were introduced into the cask, the reduction in the measured outer slab count rates when the last canister was introduced into the cask, and the overall discrepancies between the calculated and measured outer slab count rates.

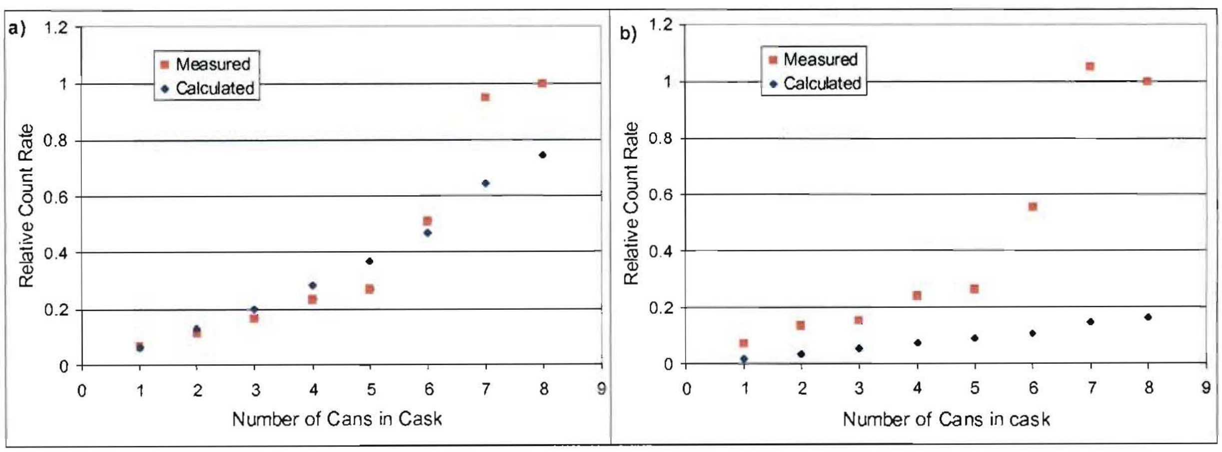

Figure 4. Measured (red squares) and calculated (blue diamonds) count rates in the a) inner slab and b) outer slab as a function of the number of canisters that had been loaded into an empty cask. The count rates shown in the figure are relative to the measured count rates in the inner and outer slab, respectively, with all 8 canisters in the cask.

During the time between the first and second measurements campaigns, the casks that had been present at the storage pad at the BN-350 during the first campaign had been transported to the permanent storage location and were no longer available for verification measurements when the second measurement campaign occurred. While it was no longer possible to repeat verification measurements on casks that had been initially measured during the first campaign, repeat verification measurements were performed on a number of the casks that were present at the BN-350 storage pad during the second campaign. These measurements were performed to test the performance of the DSVD. For these measurements, the baseline and re-verification measurements were performed on different days during the measurement campaign. Figure 5 shows the measured deviation in the count rates between the baseline and verification measurements as a function off the measurement position for two of the casks. For the two casks shown in Fig. 5, the measured deviation in the fingerprint is consistent with no change having taken place in the contents of the two casks as expected. The error bars shown in Fig. 5 are the preliminary total error bars for the measurements which include both the statistical and systematic error components of the measurement. For the two casks shown in Fig. 5, the total error bars associated with the deviation in the fingerprints range from ± 2.5% to ± 3.5%.

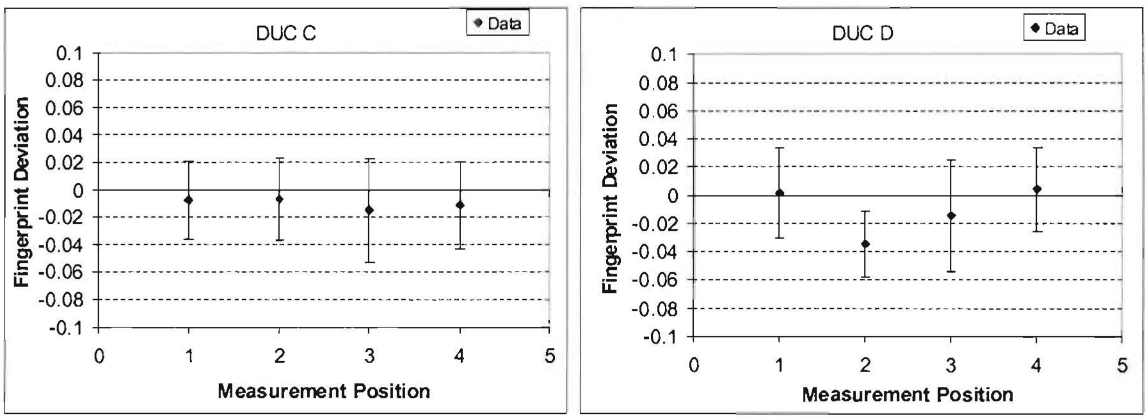

Figure 5. Deviation in measured count rates as a function of the measurement position for two DUCs that had been measured twice on separate days during the second measurement campaign. The error bars shown in the figure include both statistical and systematic errors.

SUMMARY

Measurements have been performed on BN-350 spent fuel in dry storage casks using the DSVD at to establish a neutron "fingerprint" for each cask. As part of the initial set of measurements that were performed, a sensitivity measurement was performed with the DSVD fixed at a given location on the outside of a cask as it was loaded with canisters of spent fuel. An initial analysis of this sensitivity data shows significant differences between the measured count rates in the inner and outer slabs of the DSVD counter and the calculated count rates that are based on MCNPX simulations of the measurements. Work is currently ongoing to rectify the differences between the calculated rates and the measured rates for this set of sensitivity measurements.

Repeat measurements were performed on several casks to test the stability of DSVD measurements. A preliminary analysis of these measurements have shown the total uncertainty associated with verifying the neutron "fingerprint" of a dry storage cask is on the order of ± 3%. This uncertainty estimate includes error associated with the normalization measurements using a single AmLi source as well as error associated with repositioning the DSVD on the side of the cask.

ACKNOWLEDGEMENTS

This work was sponsored by the Department of Energy, Office of Global Threat Reduction (NA-21).

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