NMIS PLUS GAMMA RAY SPECTROMETRY FOR VERIFICATION OF WATER AND B_4C CONTENT OF THE ISOLATING MATERIAL FOR STORAGE AT HEUMF

J. S. Neal, S. A. Pozzi, J. D. Edwards, and J. T. Mihalczo
Oak Ridge National Laboratory
P. O. Box 2008, Oak Ridge, Tennessee 37831-6010, USA

ABSTRACT

The Nuclear Materials Identification System (NMIS) with gamma ray spectrometry can be used to verify the water and B_4C content of BoroBond™, which is the material in the rackable can storage boxes (RCSB) of the Highly Enriched Uranium Materials Facility (HEUMF) at the Y-12 National Security Complex. The water in the BoroBond™ slows down neutrons so that they can be captured by the boron. The fast neutron transmission measured by NMIS depends on the water content, and the capture events in boron depend on both the water content and the B_4C content. Having determined the water content from fast neutron transmission measurements, the B_4C content can be estimated from the measurement of the 478 keV boron-capture gamma rays.

A variety of measurements were performed with BoroBond™ blocks of varying thickness, B_4C content, and water content, as well as with a mockup of a RCSB of fixed water and B_4C content. These measurements were aimed at establishing a method to verify the water and B_4C content of the RCSBs at the production site, or subsequently at the Y-12 National Security Complex before or during the operating lifetime of the RCSB at the HEUMF.

INTRODUCTION

BoroBond™, a ceramic material containing natural boron carbide (neutron absorber) and water (neutron attenuator), is the filler material of the Rackable Can Storage Boxes (RCSBs) that will store highly enriched uranium in cans at the Highly Enriched Uranium Materials Facility (HEUMF) at the Y-12 National Security Complex [1,2]. Both attenuation and absorption influence nuclear criticality safety of the fissile material stored in RCSBs. To characterize the neutron attenuation and neutron absorption properties of this material, the Oak Ridge National Laboratory (ORNL) has performed an extensive series of measurements which included: fast neutron and gamma time-of-flight transmission using the Nuclear Materials Identification System (NMIS), and activation analysis with gamma ray spectrometry using a high purity germanium (HPGe) detector. These measurements were performed for a series of 12x12-in. square blocks of thickness varying from 2 to 12 in., with nominal natural B_4C contents of 0, 2.3, 4.6, and 9.1 wt%, and two water contents achieved by baking the blocks to remove approximately 5/6 of the water. These measurements were also performed with a special RCSB of BoroBond™ material with nominal 4.6% natural B_4C.

The purpose of this work was to develop methods for quantification of the water and B_4C content of the RCSBs at the production site, upon receipt at Y-12, or at any time of their useful life in the HEUMF. Another purpose was to provide data that can be used to verify Monte Carlo neutron transport theory methods that are used for criticality safety analysis. The latter is important since this material has never been used for an isolating material in a highly enriched uranium metal storage facility.

Herein, we describe the measurement methods, the configuration of the sources, detectors, and materials, the data obtained from some of the measurements, the analysis of the data, and recommendations for quantification of both the water and B_4C content of the RCSBs. The special RCSB used in the measurements was missing the steel top of its container, thus exposing the BoroBond™ to air.

FAST NEUTRON TRANSMISSION MEASUREMENTS

Four sets of test blocks, with 8 blocks in each set, of varying boron concentration and thickness were provided in order to investigate water concentration sensitivity as a function of hydrogen concentration and block thickness. Nominal block dimensions were 12x12x2 inches and 12x12x4 inches with nominal ${}^{nat}B_4C$ concentrations of 0, 2.3, 4.6, or 9.1 weight percent.

Fast neutron transmission depends strongly on water content in the blocks. Source neutrons for these measurements were provided by ${}^{252}Cf$ electroplated on one plate of a parallel plate ionization chamber that produces a timing pulse each time ${}^{252}Cf$ spontaneously fissions. The fission neutrons have an energy distribution similar to that of uranium fission but slightly higher in energy. The emitted fission gamma rays and fast neutrons enter the BoroBond™ blocks, and their transmission is measured by a proton recoil scintillation detector placed on the opposite side from the source. The times of detection are measured after Cf fission with the Nuclear Materials Identification System (NMIS) [3]. Figure 1 presents time-of-flight measurements for blocks with nominal ${}^{nat}B_4C$ concentrations of 4.6% and varying thickness.

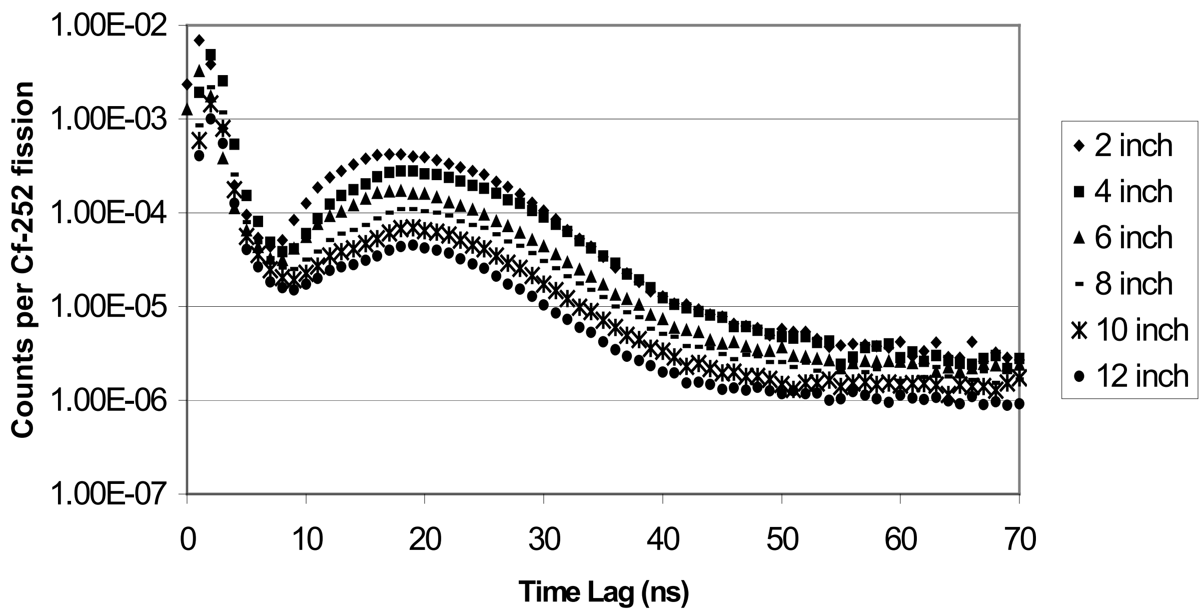

Fig. 1. Time-of-flight transmission measurement with the plastic scintillator for unbaked blocks with ${}^{nat}B_4C$ concentrations of 4.6% as a function of thickness.

A number of simulations were performed with the Monte Carlo code MCNP-PoliMi [4] for blocks of varying water content. As expected, attenuation of the neutrons from the source increases with water content. A feature was extracted from the transmission measurements to be related to the water content of the blocks. The feature is the total integral of the counts in the neutron peak. The

Monte Carlo calculational results were fit to an exponential function of water content. The results of the measurements are shown in Figure 2. As it can be seen, there is good agreement between the Monte Carlo simulation and the measurements. These Monte Carlo calculations used nominal atomic compositions. Although this was done for BoroBond™ blocks, the same procedure can be used for the actual RCSBs once an initial calibration measurement is performed.

To adjust for the calculational bias, the calculated values were adjusted to the experimental value at 100% water content. The resulting functional dependence on water content was fit with an exponential, which can be inverted to give the water content (w) as a function of the neutron counts per Cf fission (n)

$$w = -90.1 \cdot \ln n - 475.8$$

This equation can be used to find the water content independently of the B_4C content of the 8 in. blocks. The normalized neutron counts (n) were found by integrating the transmission measurements for time lags 6 to 60 ns. In the case of the blocks where the water content was reduced by bake-out, the errors are greater than for the non-baked blocks. This difference might be explained by considering the error introduced by the bake-out process.

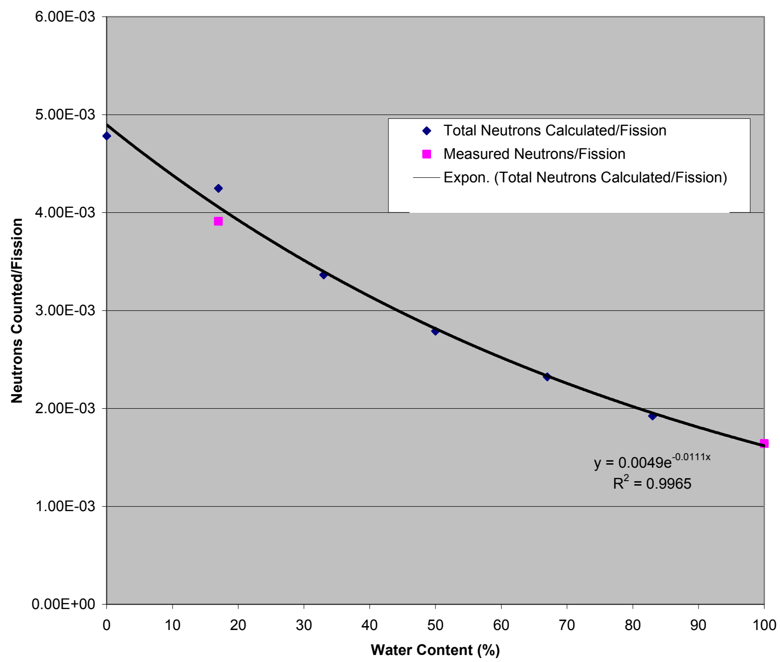

Fig. 2. Area of neutron peak transmission simulations as a function of water content for 8-in.-thick blocks of 4 wt% B_4C and measurements.

Fast neutron time-of-flight transmission measurements were also performed for the mockup RCSB with two different source locations. The first is shown in Figure 3, which is a photograph of the mockup RCSB with a source in one fissile storage location and a plastic scintillation detector in another. The mockup RCSB was oriented vertically to minimize floor reflection effects. In actual verification measurements, the RCSB will be located horizontal about 3 feet off the floor, and floor effects will be corrected. The fissile storage locations were numbered 1 to 6 starting at the upper right of this photo 1, 2, 3 across the top right to left and 4, 5, 6 across the bottom left to right. For these measurements, the source was located in the center of fissile location 1 and the detector sequentially in locations 2, 3, 4, 5, and 6. Then, the source was placed in location 2, and the detectors in locations 1, 3, 4, 5, and 6.

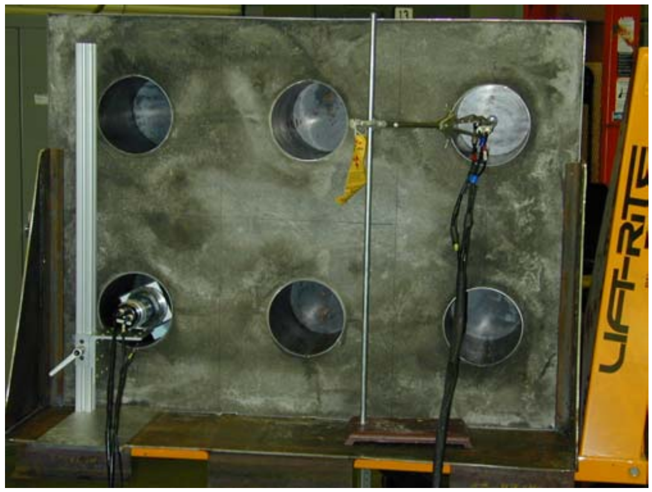

Fig. 3 Photograph of the mockup RCSB with the Cf source in fissile location 1 and the detector in fissile location 4.

Typical transmission measurement distributions with the source placed in location 1, and the detectors in locations 2 through 6 are shown in Figure 4.

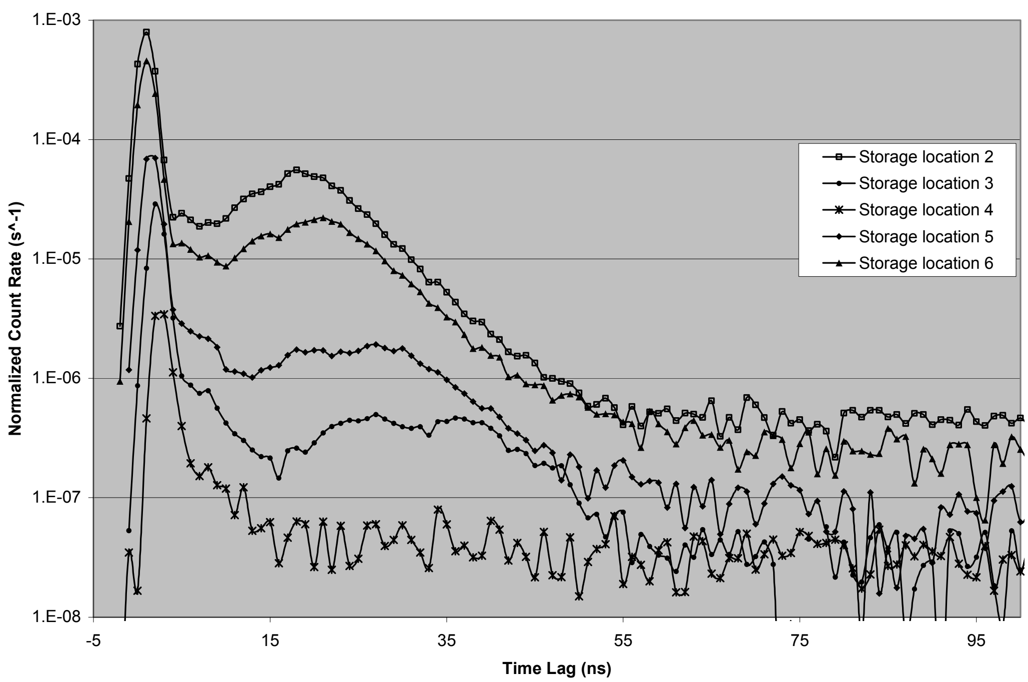

Fig. 4. Time-of-flight transmission measurement results for the source in location 1 and the detector in location 2 to 6.

Integrals of the neutron peak of the transmission measurements are given in Table 1. The table also shows the integrals for the measurements performed with the source in location 2, and the detector in locations 1, 3, 4, 5, and 6. These integrals are related to the product of the detection efficiency and the number of neutrons per fission transmitted between fissile storage locations in the RCSB. Fission neutron attenuation is larger with greater spacing between holes. For non-adjacent holes the attenuation is an order of magnitude higher than for adjacent holes. These low transmissions reflect the good fast neutron isolation properties of BoroBond™ of these thicknesses.

Table 1. Integral of neutron peak per Cf fission for mockup RCSB transmission measurements with source in fissile storage locations 1 and 2[a]

Source position	Integral of neutron peak for detector in fissile storage locations					
	1	2	3	4	5	6
1	--	8.17E-04	1.65E-05	4.83E-06	5.14E-05	3.82E-04
2	8.50E-04	--	8.27E-04	5.46E-05	4.20E-04	5.21E-05

[a]Numbers in this table refer to fissile storage locations.

Another series of measurements was performed with different source-detector locations. In this configuration, the ^{252}Cf source was placed in a different location, as shown in the photograph of Figure 5. Two 1-in.-diam and 6-in.-deep source holes were drilled in the RCSB. The one on the left is designated source hole A, and the one on the right as source hole B. These source holes were each equidistant between four fissile storage locations. This geometry is that recommended for the at-the-factory verifications. These source holes would be provided in each RCSB. Sources in these holes will provide unambiguous repeatable location of the sources for this type of measurement, provide a storage location for the sources when not in use at the factory for verification, and, because of symmetry of the source-detector locations, indicate the uniformity of the BoroBond™ of the RCSB. Because of symmetry, responses B1, B6, A3, A4 and B2, B5, A2, A5 should be alike. The results of these measurements are given in Table 2.

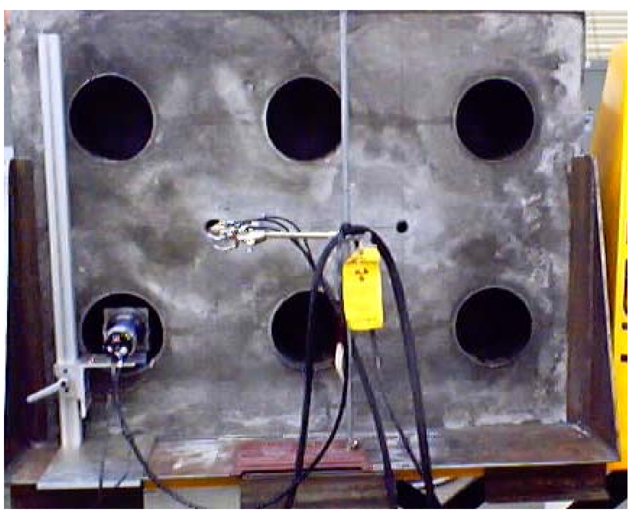

Fig. 5. Photograph of measurement setup for the mockup RCSB transmission measurements with Cf source located in hole A and detector in fissile storage location 4.

Table 2. Integral of neutron peak per Cf fission for the mockup RCSB transmission measurements with source in holes A and B[a]

Source position	Integral of neutron peak for detector in fissile storage locations					
	1	2	3	4	5	6
A	--	3.10E-03	3.11E-03	3.50E-03	3.46E-03	--
B	2.83E-03	2.94E-03	--	--	3.81E-03	3.61E-03

[a] Numbers in this table refer to fissile storage locations.

The values of Table 2 are higher than those of the previous configuration (Table 1) because there is less material between the source and the detector. The increased response of the detectors in the lower fissile storage locations is the result of floor reflection and can be corrected for. The low transmissions indicate the good neutron isolation properties of BoroBond™ at these thicknesses.

GAMMA SPECTROSCOPY MEASUREMENTS

The boron capture process depends on both the boron and water content, and different boron and water contents can produce the same neutron captures in boron. After an initial set of measurements, half of the blocks were baked at 140°C for approximately 24 hours in order to reduce the water content, and thus vary the hydrogen concentration. Measurements would then be considered for use as boron concentration calibration standards for the RCSB material. Prompt Gamma Neutron Activation Anlaysis (PGNAA) techniques were employed in this study.

Figure 6 presents the arrangement of (6) ^{252}Cf pellet sources, a 4x4x4 inch tungsten alloy block, and a high-purity germanium (HPGe) detector against a 12-inch block. The tungsten block provided a high-density shield for lowering ^{252}Cf source gamma flux at the HPGe detector. This arrangement of source and detector can also be used adjacent to the RCSBs.

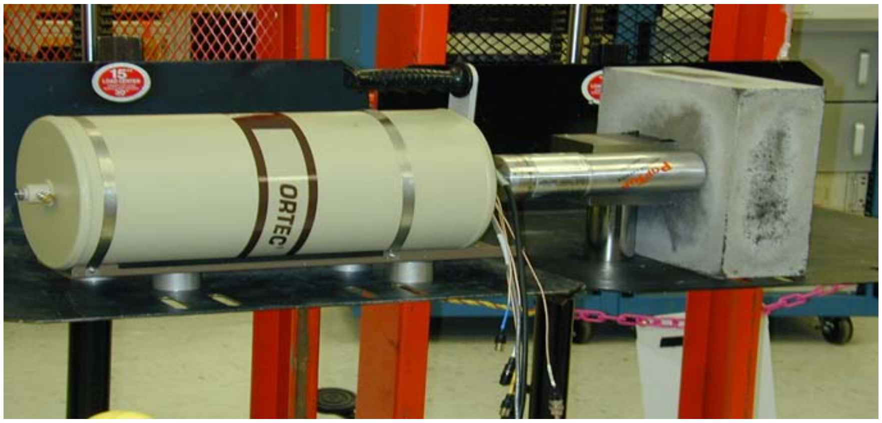

Fig. 6. Experimental arrangement (side view).

In order to determine the sensitivity of the measurement method to B_4C content for a given material thickness, data are plotted in Figure 7 as counts in the boron peak versus nominal B_4C content.

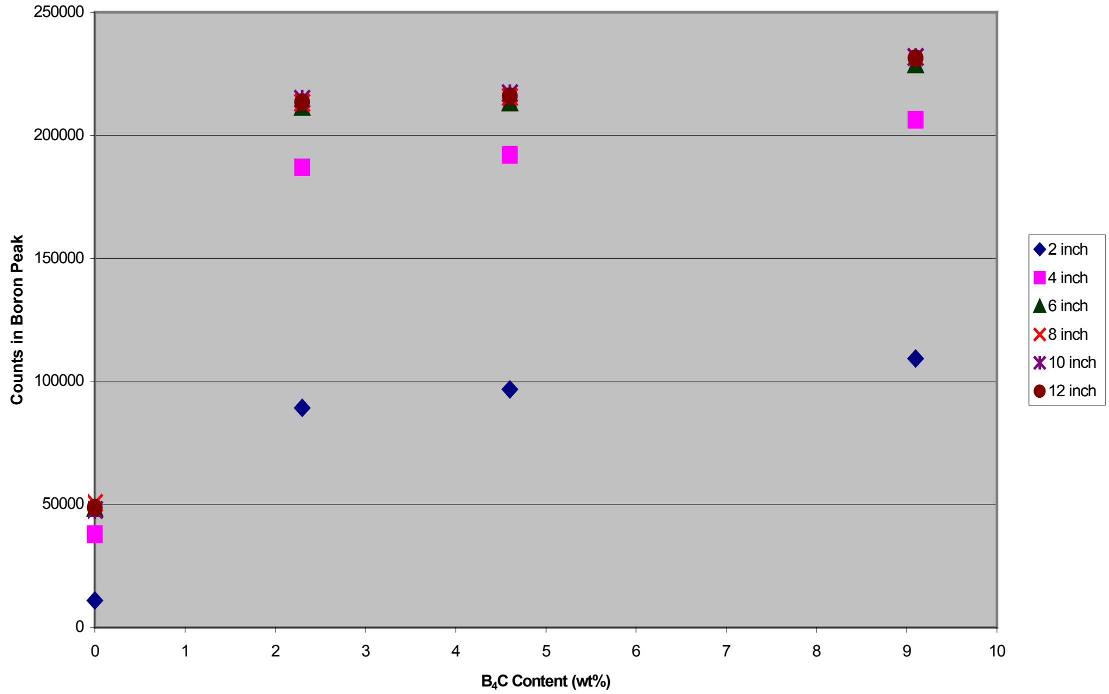

Fig. 7. Variance-weighted average values for counts in the boron peak as a function of the nominal boron concentration (unbaked).

At first it was assumed that the slopes for the data series in Figure 8 were linear over the range of 2.3 to 9.1 wt% B_4C. Using the variance-weighted average values and the variances for those average values, we estimated the sensitivity and associated error for each block thickness using the Bayesian Inference Using Gibbs Sampling (BUGS) program. The BUGS program was also used to estimate the uncertainty in wt% B_4C given counts in the boron peak by accounting for uncertainties in all of the linear model fitting parameters as well as errors in the measurements. Uncertainties for each block thickness are presented in Table 3.

Table 3. Estimated wt% B_4C uncertainty for each block thickness

Nominal Thickness (in.)	B_4C Uncertainty (wt% B_4C)
2	0.10
4	0.32
6	1.08
8	1.01
10	0.93
12	0.95

We concluded that B_4C content may be estimated to ±1 wt% B_4C for blocks of similar composition and geometry.

Measurements were made at points located on the perimeter of the mockup RCSB. The mockup RCSB was filled with 4.6 wt% B_4C BoroBond™. Measurement points were grouped by approximate BoroBond™ thickness. Approximate thicknesses were 4.3, 5.5, and 6 inches. Using the BUGS program, the predicted wt% B_4C for the ~6-in. thick RCSB measurements was calculated using variance-weighted average value of boron counts and the linear calibration model from above. The BUGS program calculation accounts for uncertainty in all linear model parameters as well as errors in the measurements. For the 6-in. RCSB measurements, BUGS predicted 4±1 wt% B_4C.

VERIFICATION METHOD

Unfortunately, no single method investigated could quantify both the hydrogen and B_4C contents independently. The fast neutron time-of-flight transmission method is sensitive to the water content and not to the B_4C content. Thus, fast neutron transmission can be used to determine the water content. Gamma spectrometry directly measures the gamma rays produced when a neutron is absorbed by boron. Without water present to slow the neutrons, the boron capture rate is greatly reduced. There are different combinations of water and B_4C content that yield the same measured result. However, if the water content is known from the fast neutron time-of-flight transmission measurements, the B_4C content can be obtained by the PGNAA method.

The fast neutron time-of-flight transmission and gamma ray spectrometry methods could be implemented as follows. The data could also be used to assess the uniformity of the BoroBond in the RCSB and archived for future comparisons. The fast neutron time-of-flight transmission measurements would use two ^{252}Cf sources in ionization chambers inserted in two specially provided 6-inch deep holes, each equidistant from four fissile storage can locations in the RCSB. Each fissile storage can location would contain one of six fast plastic scintillation detectors connected by cables to the associated electronics. The symmetry of the source-detector arrangement allows easy assessment of the uniformity of the BoroBond™ material in the RCSB. The source-shield-detector arrangement for gamma spectrometry was adjacent to the sides of the RCSB. So as to not have to correct for the decay of the source, it is recommended to use an Am/Be source (433 year half-life). With the water content determined from the fast neutron transmission, gamma ray spectrometry could then be used to quantify the B_4C content.

REFERENCES

[1] A. W. Krass, K. D. Lewis, J. J. Lichtenwalter, D. A. Tollefson, “Comparison of Absorber Materials for a Palletized Rack Storage System,” Transactions of the American Nuclear Society, 85, 159,2001.

[2] J. J. Lichtenwalter, D. A. Tollefson, L. E. Johnsen, A. W. Krass, K. D. Lewis, “Palletized Rack Storage System Absorber Material Testing,” Transactions of the American Nuclear Society, 85, 161 (2001).

[3] J.T. Mihalczo, J.A. Mullens, J.K. Mattingly, T.E. Valentine, “Physical Description of Nuclear Materials Identification System (NMIS) Signatures,” Nuclear Instruments and Methods in Physics Research A, 450, 531 (2000).

[4] E. Padovani and S.A. Pozzi, "MCNP-PoliMi ver. 1.0. User's Manual", CESNEF-021125 Library of Nucl. Eng. Dept., Polytechnic of Milan, Italy, November 2002