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DEVELOPMENT OF AN NDA SYSTEM FOR HIGH-LEVEL WASTE FROM THE CHERNOBYL NEW SAFE CONFINEMENT CONSTRUCTION SITE

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ABSTRACT

In early 2009, preliminary excavation work has begun in preparation for the construction of the New Safe Confinement (NSC) at the Chernobyl Nuclear Power Plant (ChNPP) in Ukraine. The NSC is the structure that will replace the present containment structure and will confine the radioactive remains of the ChNPP Unit-4 reactor for the next 100 years. It is expected that special nuclear material (SNM) that was ejected from the Unit-4 reactor during the accident in 1986 could be uncovered and would therefore need to be safeguarded. ChNPP requested the assistance of the United States Department of Energy / National Nuclear Security Administration (NNSA) with developing a new non-destructive assay (NDA) system that is capable of assaying radioactive debris stored in 55-gallon drums. The design of the system has to be tailored to the unique circumstances and work processes at the NSC construction site and the ChNPP. This paper describes the Chernobyl Drum Assay System (CDAS), the solution devised by Los Alamos National Laboratory, Sonalysts Inc., and the ChNPP, under NNSA's International Safeguards and Engagement Program (INSEP). The neutron counter measures the spontaneous fission neutrons from the ^{238}U, ^{240}Pu, ^{244}Cm in a waste drum and estimates the mass contents of the SNMs in the drum by using of isotopic compositions determined by fuel burnup. The preliminary evaluation on overall measurement uncertainty shows that the system meets design performance requirements imposed by the facility.

INTRODUCTION

During the catastrophic nuclear accident at the Chernobyl Nuclear Power Plant (ChNPP) in 1986, unknown amount of special nuclear material (SNM) was ejected from the Unit-4 reactor. During the accident remediation, this SNM was buried under concrete and soil. It has lied there undisturbed for more than twenty years. Although technically under safeguards, neither Ukraine nor the International Atomic Energy Agency (IAEA) has been able to assay or verify this material because it is virtually inaccessible. This was acceptable to the international community so long as there was no chance that the SNM could be excavated and removed from the site. With the recent start of construction of the New Safe Confinement (NSC), this approach for safeguarding the SNM buried at the Chernobyl site is no longer viable. The NSC is the structure that will replace the present containment structure and will confine the radioactive remains of the ChNPP Unit-4 reactor for the next 100 years. The area around the Shelter is being excavated in order to lay the foundations of the NSC and its supporting infrastructure. It is expected that SNM could be uncovered during the excavation process and would therefore need to be safeguarded.

In early 2009, preliminary excavation work has begun in preparation for the construction of the NSC. ChNPP requested the assistance of the United States Department of Energy/National Nuclear Security Administration (NNSA) with developing a new NDA system that is capable of assaying radioactive debris stored in 55-gallon drums. The design of the system has to be tailored to the unique circumstances and work processes at the NSC construction site and the ChNPP.

Upon the request from DOE/NNSA, the Los Alamos National Laboratory (LANL) has developed the Chernobyl Drum Assay System (CDAS) to support safeguards measurements at ChNPP for the waste from the NSC excavation site. The CDAS measures the spontaneous fission neutrons from the ^{238}U, ^{240}Pu, ^{244}Cm in a waste drum and calculates the mass contents of the SNM in the drum. The latest capabilities of coincidence counting in the MCNPX transport code were used to calculate the singles, doubles, and triples count rates of the ^{3}He detectors. The system was shipped to ChNPP and installed at the Temporary Storage Facility (Building 12) in November 2009. This paper describes the design specifications and operating characteristics of the CDAS. The expected performance of the counter, with its measurement uncertainty and the applicability of the technology to ChNPP safeguards, is also discussed.

SOURCE TERM ANALYSIS

Burnup history of a spent nuclear fuel sample is critical information for the quantitative measurement of isotopic SNM contents in a sample. In addition, the matrix surrounding the sample needs to be characterized to account for the neutron multiplication and moderation in the measurement system. This crucial information is unknown in the ChNPP waste. The only available information on the source term is typical burnup value averaged over the reactor core based on the operation record at the time of the accident and gamma measurement data measured during drumming process at the excavation site.

Chernobyl Unit 4 was brought into operation in December 1983. When the unit was shut down for medium term maintenance that was scheduled for 25 April 1986, the core contained 1650 fuel assemblies with an average burnup of 10.3 GWD/MTU. Most of the fuel assemblies (75%) were from the first core charge and had a burnup of 10 – 15 GWD/MTU [1].

To investigate the possibility of using the gamma measurement data for sample burnup estimation, the gamma measurement data from the site were analyzed with Origen-ARP code [2]. The validation of the Origen-ARP code is also established by comparing the results with isotopic composition data provided by the site for several spent fuel assemblies; the reliability of the isotopic data were evaluated with Russian originated FUEL code and confirmed by IAEA. Table 1 summarizes the specifications of the ChNPP-4 spent fuel assemblies used for the burnup code validation.

Table 1. Specifications of the ChNPP-4 Spent Fuel Assemblies for Burnup Code Validation.

Fuel Assembly No.	Burnup (GWD/MTU)	Cooling (yrs)	Initial Enrichment (%)	Power (MWD/Assembly)
1.20.4208.82	10.898	23	2.0	1250
1.20.0933.83	2.825	25	2.0	324
1.20.6805.84	13.932	20	2.0	1598
2.20.7619.79	14.368	23	2.0	1648
4.20.34089.91	13.836	12	2.0	1587

The result of comparison between Origen-ARP and FUEL code for major actinides in a typical ChNPP-4 spent fuel assembly of 10.8 GWD/MTU burnup is shown in Fig. 1. As observed from the figure, the Origen-ARP code results show good agreements with FUEL results for major actinides. The important major actinide ^{235}U mass is agreed within 5% of the difference between two codes, while ^{239}Pu is agreed within 10% of the difference. The major spontaneous fissionable isotopes ^{238}U, ^{240}Pu, and ^{244}Cm are agreed within 8% of the difference. Total U and Pu mass are agreed within 1% and 8% respectively.

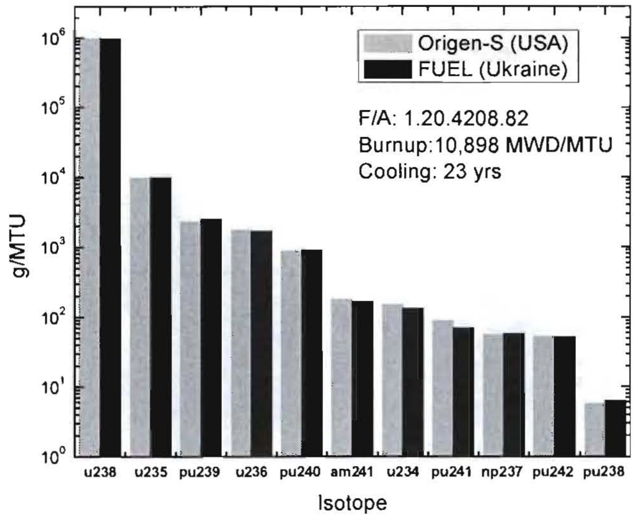


Fig. 1. Comparison of SNM mass inventory in a typical RBMK-1000 spent fuel.

Origen-ARP calculated burnup response curve as a function of activity ratio between ^{134}Cs and ^{137}Cs was generated using RBMK-1000 library as shown in Fig. 2. The burnup of the ChNPP spent fuel assembly is best fitted with the exponential decay function with following coefficients and an adjusted R^2 value of 0.999. In order to validate the burnup curve derived using Origen-ARP, burnup estimations were performed for several waste drums using the gamma measurement data provided by the ChNPP facility.

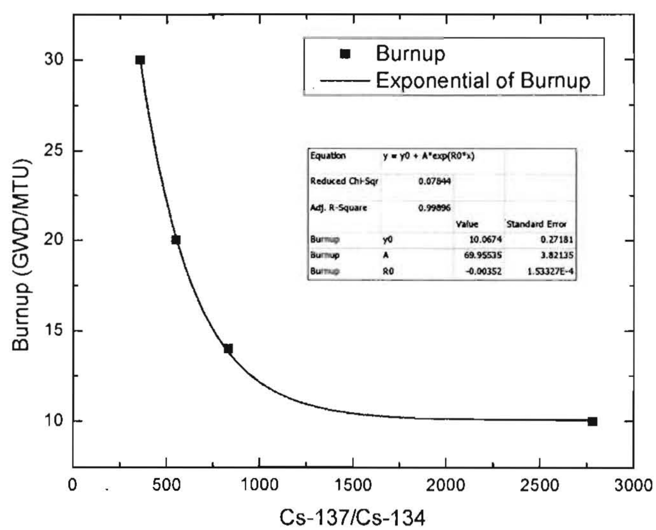


Fig. 2. Burnup response curve as a function of activity ratio Cs-137/Cs-134.

The ChNPP has been using gamma instruments combined with INSPECTOR-2000 and ISOCS (In Situ Object Counting System) calibration software for the gamma activity measurements of the waste drums at the excavation site. The burnup estimations based on the INSPECTOR-2000 measured gamma activity ratios and the Origen-ARP derived burnup curve are summarized in Table 2. As shown in the table the estimated burnups are in the range of 10 – 16 GWD/MTU, which is reasonable considering the reported burnup range of 10 - 15 GWD/MTU for the ChNPP-4 spent fuel assemblies [1].

Table 2. Burnup Estimation for ChNPP Waster Drums using $^{137}Cs/^{134}Cs$ Activity Ratios.

Drum No.	INSPECTOR-2000 (kBq)				$^{137}Cs/^{134}Cs$	Estimated Burnup (GWD/MTU)
	Cs-134	Cs-137	Eu-152	Eu-154		
1	7.99	1.46E+04	1.98	6.28E+01	1,827.28	10.18
4	4.54E-06	3.45E-03	4.00E-06	2.10E-05	759.91	14.89
7	4.40E-06	7.12E-03	5.52E-06	4.52E-05	1,618.18	10.30
8	7.10E-05	5.03E-02	2.36E-04	3.08E-04	708.45	15.85
32	4.21E-05	3.10E-02		5.61E-04	736.34	15.31

ASSAY APPROACH

In order to accomplish the design requirement of the NDA system; which is quantitative measurement of U, Pu, and ^{235}U contents in a sample with an uncertainty less than 50%, a series of assay procedure has been developed. The approach is based on an assumption that the burnup information is provided by gamma measurements on a waste drum, and the information is reliable with reasonable uncertainty. With the burnup information, isotopic composition of the sample can be evaluated using a burnup calculation code, Origen-ARP. The isotopic composition of the sample is to provide spontaneous fission neutron yield and neutron multiplicity information of the sample.

In conventional neutron coincidence assay, singles and doubles rates are counted using a counter with several He-3 neutron coincidence detectors [3]. The physics of the coincidence assay is based on the point model equations as followings;

$$S = mF_0 \varepsilon \nu_{s1} M(1+\alpha),$$

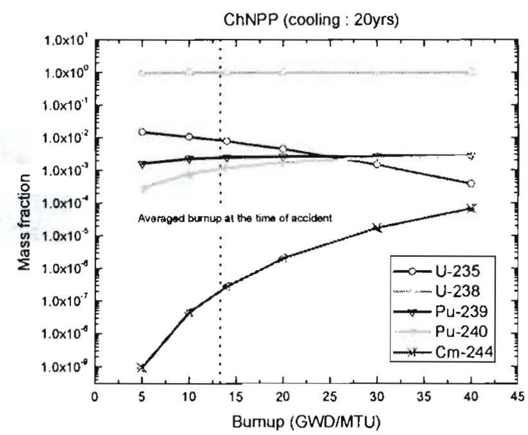
$$D = \frac{1}{2} mF_0 \varepsilon^2 f_d \nu_{s2} M^2 \left[1 + \left(\frac{M-1}{\nu_{i1}-1}\right) \frac{\nu_{s1}\nu_{i2}}{\nu_{s2}} (1+\alpha)\right],$$

where,

S, D = Singles (Totals) and Doubles (Reals) rates,
m = total mass of effective spontaneous fission isotopes,
M = neutron multiplication,
α = (α,n) to spontaneous fission neutron ratio,
ε = detection efficiency,
f_d = doubles gate fraction,
ν_{si} = factorial moments i of the spontaneous fission neutron multiplicity distribution,
ν_{ii} = factorial moments i of the induced fission neutron multiplicity distribution,
F_0 = spontaneous fission rate per unit mass.

Among the variables in the two equations, total mass of spontaneous fission isotopes *(m)*, neutron multiplication *(M)* , and (α,n) to spontaneous fission neutron ratio *(α)* are unknown, while neutron multiplicity distributions (v_{si} , v_{ii})can be estimated if isotopic composition of the sample is known. The doubles gate fraction (f_d) and detection efficiency *(ε)* are variables that are affected by matrices between neutron source term and detectors. With an assumption that the intrinsic neutron multiplication of SNM itself is negligible and net neutron multiplication is mostly affected by surrounding matrix, the variable *M* also can be a function of surrounding matrix. Therefore, most of the variables in the point model can be determined if the isotopic composition of the SNM is known and the surrounding matrix is characterized.

As described in previous section, the isotopic composition of the SNM can be determined by the burnup estimation. In case of spent fuel assemblies from light water reactor, ^{244}Cm is the only major spontaneous fission neutron source because of its incomparably high spontaneous fission yield (1.080×10^7 fission/s-g). It is noted, however, that the mass fraction of ^{244}Cm in the ChNPP spent fuel is relatively lower than that of general light water reactor because of the considerably low burnup. The mass fractions of major actinides as a function of fuel burnup were calculated with Origen-ARP as shown in Fig. 3 (left). For the CDAS assay approach, therefore, it is reasonable to include ^{238}U and ^{240}Pu in addition to the ^{244}Cm as major spontaneous fission isotopes. The effective neutron multiplicity distributions are calculated for a given SNM with a specific burnup using MCNPX (Monte-Carlo N-Particle eXtened) transport code as shown in Fig. 3 (right) [4].



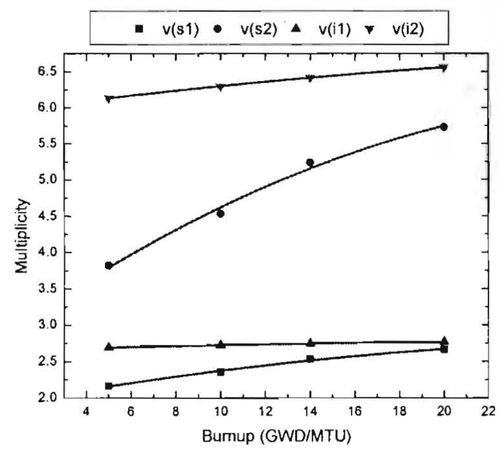


Fig. 3. Isotopic mass fractions (left) and effective neutron multiplicities (right).

The sample matrix in a waste drum has two primary effects on the neutrons: (1) energy reduction by scattering reactions, and (2) neutron absorption of the low-energy neutrons. The counter is designed with an optimum moderator (CH_2) thickness to be relatively insensitive to the energy reduction; however, as the hydrogen density in the drum increases, the absorption process significantly reduces the measured neutron signal.

In order to characterize the matrix perturbation in a waster drum, the Add-a-Source (AS) method is employed in the CDAS approach [5]. The basis of the Add-a-Source (AS) method is to measure the matrix perturbation to the counting rate from a small ^{252}Cf source ($\sim1\times10^6$ n/s) on the outside of the sample and use the information to correct for the matrix perturbation on the inside of the sample. To correct for the matrix perturbation on the neutron signal, the AS method measures each drum both with and without the ^{252}Cf source at the bottom of the drum. The AS method can provide necessary information on the doubles gate fraction (fd), detection efficiency (ε), and neutron multiplication (M) based on the detector response by active interrogation on the matrix using an external ^{252}Cf neutron source.

With the matrix dependent parameters determined by the AS measurement and the burnup dependent parameters according to burnup estimation, total spontaneous fission rate of the sample in a waste drum can be calculated as follows;

$$F = \frac{S-D}{(C_1-C_3)(1+\alpha)-C_2} \text{ (sf/sec)},$$

$$\text{where,} \quad C_1 = \varepsilon M \nu_{s1},$$
$$C_2 = \frac{\varepsilon^2 f_d M^2}{2} \nu_{s2},$$
$$C_3 = \frac{\varepsilon^2 f_d M^2}{2} \nu_{s2} \left[\frac{M-1}{\nu_{i1}-1}\right] \nu_{s1} \nu_{i2},$$
$$\alpha = \frac{{}^{S}/_{D}(C_2+C_3)-C_1}{C_1-{}^{S}/_{D} C_3}.$$

The isotopic composition data previously evaluated using burnup code is used to calculate the necessary SNM contents in the sample. Even though the net neutron multiplication by the matrix can be evaluated based on the Add-a-source method; the intrinsic net neutron multiplication in the source material itself and α-value (ratio of (α,n) neutron and spontaneous fission neutron) are unknown. A predictor-corrector approach is used to evaluate proper neutron multiplication and α-value in the system by comparing the calculated coincidence count rates with measurement results.

DESIGN CHARACTERISTICS

The CDAS is designed to measure a 200-L drum by passive and active neutron assay. It consists of six ^{3}He-based neutron detector slabs and a neutron source tray at the bottom as shown in Fig. 4. According to the design requirement, the system is designed to locate at the top of the KTZV-02 container and conduct measurement while an overhead crane is holding the drum. In order for the drum manipulator to be able to get through the sample chamber, three detector slabs are designed to have 15 cm distance from the drum surface, while the other three detector slabs are located adjacent to the drum surface with minimum clearance. The detector slabs adjacent to the drum surface contain three ^{3}He tubes to achieve proper efficiency, while the other detector slabs have two ^{3}He tubes. Each detector slab has a protective cover of 1 cm-thick carbon steel to protect the system from gamma radiation and manipulator movement.

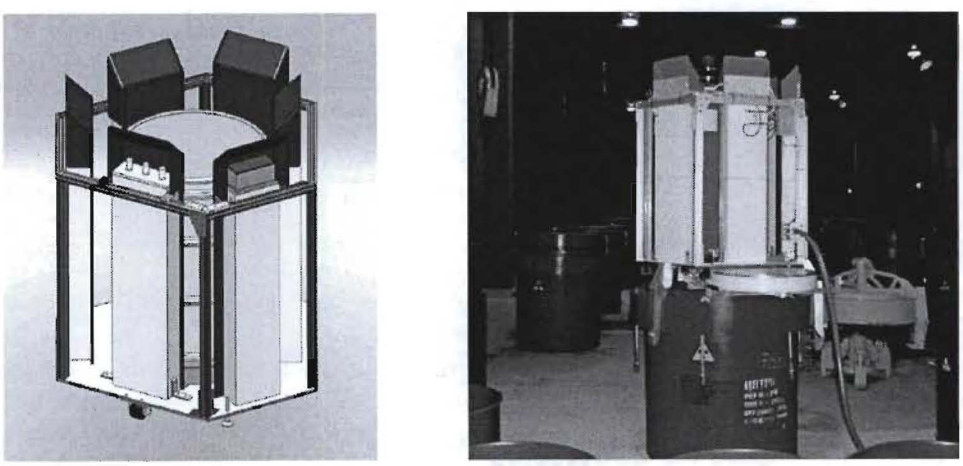


Fig. 4. CDAS on the KTZV-02 waste drum container.

For the AS measurement, the CDAS was designed to have a sliding source tray at the bottom of the system. The source tray is designed to slide in and out of the system, so that operators can pull out the tray to make a space for drum manipulation and push in the tray for the AS measurement. The source tray is made of a 3-in high-density polyethylene and an aluminum cover of 1 cm thickness. A cylindrical ^{252}Cf source is located at the center of the tray using a 1-m stainless steel rod through the hole of 0.5 cm radius at side. To maximize the interrogation neutron flux to the drum, a 1-in thick Nickel reflector is placed underneath of the source.

SYSTEM PERFORMANCE

Detection Efficiency

Among the variables in the point model, the doubles gate fraction *(f_d)*, neutron multiplication *(M)*, and detection efficiency *(ε)* are the variables that are affected by matrices surrounding SNM. Using the AS measurements, the doubles ratio of the empty drum (after source decay correction) to the net loaded drum (D_0/D_{net}) can be used for the matrix correction for efficiency, gate fraction, and multiplication. The detection efficiency calculated with MCNPX is shown in Fig. 5 (left) as a function of matrix perturbation. The matrix perturbations are simulated for sample matrices of air, concrete, and soil with different fill heights.

According to the design requirement by the ChNPP, natural boron might be co-mingled with in the SNM sample and matrix as well. Since the boron concentration does not exceed 0.5% of sample mass, it is reasonable to assume that the boron contribution to the matrix perturbation is dominated by the surrounding matrix rather than SNM itself. The MCNPX simulations extended to investigate the contribution of the boron concentration in matrix to the detection parameters show that there is no significant change in the neutron multiplication and die-away time by the boron contribution, while the changes in detection efficiencies are observable. The extended response of the detection efficiency as a function of matrix perturbation with boron mixture is also shown the Fig. 5 (left).

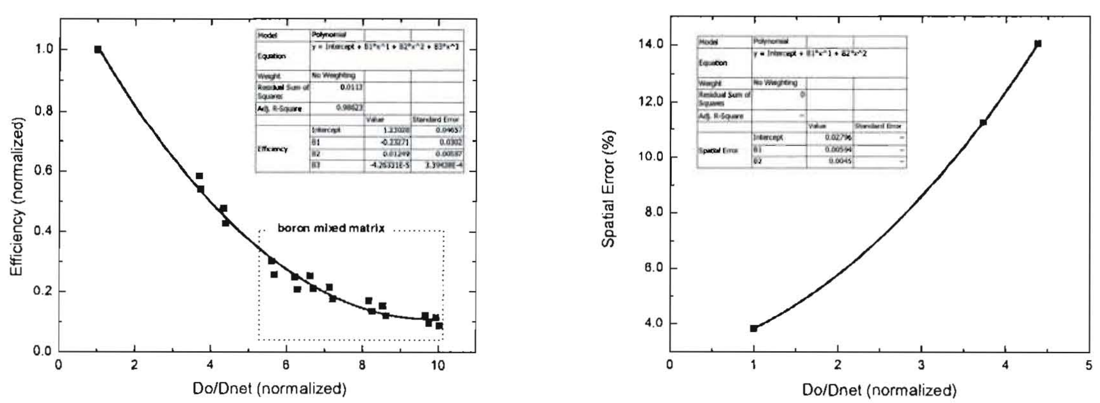


Fig. 5. Detection efficiency (left) and spatial error (right) as a function of matrix perturbation.

Spatial Efficiency Profile

Because the location of the SNM in a waste drum would be random in nature, the assay system is designed to have constant detection efficiency regardless of the geometrical source location in a suspected matrix. Even with the design approach to minimize the geometrical variance in detection efficiency, the deviation of the efficiency for the variable source location in the waste drum needs to be determined and combined in overall measurement uncertainty. To determine sample location effects, MCNPX simulations have been performed for ^{252}Cf point sources at various vertical and radial positions in waste drums filled with different matrices.

The vertical efficiency profile measurements were made for ^{252}Cf neutron sources at radial distances of 0 - 15 cm from the center of the 200-L drum. The outside edge of the drum has a radius of 28 cm and the 20-cm radius is approximately the volume-averaged mean radius. That is, the drum volume inside 20 cm equals the volume outside 20 cm. Because of the asymmetric configuration of the detector slabs in the CDAS, the detection efficiencies for several source positions at r = 0 - 15 cm in x-y direction were calculated and averaged. The standard deviations of the averaged efficiencies for the air, concrete, and soil are 3.72%, 4.38%, and 5.13% respectively.

For the radial efficiency profile, ^{252}Cf sources were positioned at four different radial positions and seven different vertical positions. The vertical positions were from -30 cm to 25 cm in regards of the drum center. The standard deviations of the averaged efficiencies for the air, concrete, and soil are 0.97%, 13.33%, and 10.05% respectively. As shown in Fig. 5 (right), the overall spatial error ranges 4% - 14% as a function of matrix.

Matrix Characterization

With an assumption that the intrinsic neutron multiplication of SNM itself is negligible (M=1) and net neutron multiplication is mostly affected by surrounding matrix, the variable *M* also can be a function of surrounding matrix. The neutron net multiplication was calculated with MCNPX as a function of matrix perturbation. The matrix perturbations are simulated for sample matrices of air, concrete, and soil with different fill heights as shown in Fig. 6 (left). According to the information provided by the ChNPP, natural boron can be contained in the SNM sample and matrix as well. Since the boron concentration does not exceed 0.5% of sample mass, it is reasonable to assume that the boron contribution to the matrix perturbation is dominated by the surrounding matrix rather than source inside. The MCNPX simulations were extended to investigate the contribution of the boron concentration in matrix to the detection parameters show that there is no significant change in the neutron multiplication and die-away time by the boron contribution.

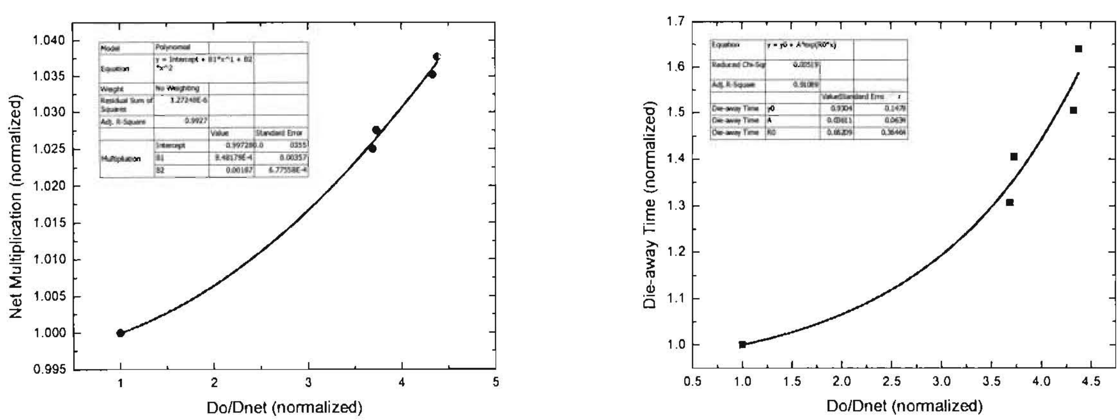


Fig. 6. Neutron multiplication (left) and die-away time (right) as function of matrix perturbation.

The response of neutron die-away time was also calculated with MCNPX as a function of matrix perturbation. The matrix perturbations are simulated for sample matrices of air, concrete, and soil with different fill heights as shown in Fig. 6 (right). The die-away time responses can be converted the doubles gate fraction with known pre-delay and gate width of the counter.

Measurement Uncertainty

The measurement uncertainty of the SNM mass evaluation using CDAS was evaluated based on the uncertainty components related to the measurement. For the isotopic composition estimation, which is based on the burnup estimation using gamma spectrometry and Origen-ARP code value, it is reasonable to apply maximum 15% of systematic error. The uncertainty of the gamma spectrometry for the burnup estimation depends on the instrumentation and measurements environment at the excavation site; it would be reasonable to assume maximum 10% of systematic error and 5% of random error considering the tough measurement environment at the site.

There is no reference waste drum for calibration in ChNPP, so efficiency calibration for the CDAS needs to be conducted using a ^{252}Cf source and MCNPX simulations. Therefore, maximum 5% of systematic error and 5% of random error are assigned for the efficiency calibration. As evaluated in previous, the detection efficiency has variations according to the spatial location in the drum, so it is reasonable to add maximum 15% of systematic error in detection efficiency.

The response functions for the matrix dependent parameters (efficiency, die-away time, multiplication) have uncertainty variances of 5%, 10%, and 5% respectively. The response curves of the neutron multiplicity values as a function of fuel burnup have maximum 5% of uncertainty. Maximum 1% of error is anticipated for Singles and Doubles measurements using CDAS. Because the waste drum is to be pulled up from the KTZV-02 container and be hold by a drum manipulator during measurement, the measurement position of the drum in the system can be different for each measurement. Even with the level sensor adopted to stop the manipulator at same position, 5% of error is applied to accommodate the error in drum positioning. Based on the error components analysis, it is anticipated that the overall uncertainty in SNM mass evaluation using the CDAS measurement would be less than 30%.

SUMMARY

This paper describes the Chernobyl Drum Assay System (CDAS) that was developed by Los Alamos National Laboratory in cooperation with Sonalysts Inc. and Chernobyl Nuclear Power Plant (ChNPP). The system is for non-destructive assay of waste drums from the New Safe Confinement (NSC) excavation site at ChNPP. The design of the system has to be tailored to the unique circumstances and work processes at the NSC construction site and the ChNPP.

In order to accomplish the design requirement of the system; which is quantitative measurement of U, Pu, and ^{235}U contents in a sample with an uncertainty less than 50%, a series of assay procedure has been developed based on the Add-a-Source neutron coincidence counting technique. The approach is to measure the matrix perturbation to the counting rate from a small ^{252}Cf source on the outside of the sample and use the information to correct for the matrix perturbation on the inside of the sample. For the procedure development, a series of MCNPX simulations was performed to simulate the counter responses for doubles and triples in active and passive modes. To investigate the reliability of the gamma measurement data for sample burnup estimation, the gamma measurement data from the site were analyzed with Origen-ARP code. The validation of the Origen-ARP code is also established by comparing the results with isotopic composition data provided by the site for several spent fuel assemblies; the reliability of the isotopic data were evaluated with Russian originated FUEL code and confirmed by IAEA.

The system was shipped to ChNPP and installed at the Temporary Storage Facility (Building 12) in November 2009. During the cold test operation with standard ^{252}Cf, the system showed proper performance as design specifications. The result of preliminary test operation showed that the CDAS assay approach is appropriate for the ChNPP waste drum application and meets the design requirement from the facility. The performance of the system will be confirmed during the hot test operation scheduled in June 2010.

ACKNOWLEDGEMENTS

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