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NDA System Response Modeling and its Application

Savannah River National Laboratory
Material Science and Technology

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Savannah River Nuclear Solutions
Savannah River Site
Aiken, SC 29808

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APPROVALS

______________________________ **Date:** ____________
D.W. Vinson, Author
SRNL-Materials Applications & Process Technology
MATERIALS SCIENCE AND TECHNOLOGY

______________________________ **Date:** ____________
G.K. Becker, Author
Becker Consulting

______________________________ **Date:** ____________
G.T. Chandler, Manager
SRNL-Materials Applications & Process Technology
MATERIALS SCIENCE AND TECHNOLOGY

______________________________ **Date:** ____________
T. Hines, Customer

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1.0 INTRODUCTION

The Portsmouth gaseous diffusion plant (PORTS) is a uranium enrichment facility that was historically used to enrich uranium to levels that range from 2% to greater than 97%. The feed material for PORTS was obtained from the Paducah Gaseous Diffusion Plant (PGDP) that produced uranium in the form of UF_6 that was enriched to about 1 to 2%. The enrichment process involves a multistage process by which gaseous UF_6 passed through a diffusion barrier in each stage. The porous diffusion barrier in each stage retards the rate of the diffusion of the heavier ^{238}U atoms relative to the diffusion of the lighter ^{235}U atoms. By this process the product stream is slightly enriched by each stage of the process. Each stage consists of a compressor, converter and a motor. There are more than 4000 stages that are linked together with piping of various diameters to form the PORTS cascade. The cascade spans three interconnected buildings and comprises miles of piping, thousands of seals, converters, valves, motors, and compressors.

During operation, PORTS process equipment contained UF_6 gas with uranium enrichment that increased in the process stream from the first to the last stage in a known manner. Gaseous UF_6 moving through the PORTS process equipment had potential to form deposits within the process equipment by several mechanisms, including solidification due to incorrect temperature and pressure conditions during the process, inleakage of atmospheric moisture that chemically reacts with UF_6 to form hydrated uranyl fluoride solids, reduction reactions of UF_6 with cascade metals, and UF_6 condensation on the internal equipment surfaces. As a result, the process equipment of the PORTS contains a variable and unknown quantity of uranium with variable enrichment that has been deposited within the equipment during plant operations. The exact chemical form of this uranium is variable, although it is expected that the bulk of the material is of the form of uranyl fluoride that will become hydrated on exposure to moisture in air when the systems are no longer buffered. The deposit geometry and thickness is uncertain and variable. However, a reasonable assessment of the level of material holdup in this equipment is necessary to support decommissioning efforts.

The assessment of nuclear material holdup in process equipment is a complex process that requires integration of process knowledge, nondestructive assay (NDA) measurements, and computer modeling to maximize capabilities and minimize uncertainty. The current report is focused on the use of computer modeling and simulation of NDA measurements.

2.0 PROCESS KNOWLEDGE

Development of computer models to support NDA methods relies considerably on process knowledge. Process knowledge includes a vast volume of information that ranges from specification data on process equipment, e.g., pipe, compressors, traps, ducts, etc., to analytical data on deposit composition. Using process equipment design and specification data is straightforward in terms of replicating the properties and characteristics of an item of interest in the computer model. Data developed through plant operation including online measurement systems, sampling of process or treatment gas, sampling of removed process equipment, and product assays may also provide information on the chemistry and/or isotopics of potential deposits within the cascade. These data should be incorporated into any computer modeling campaign.

3.0 REVIEW OF NDA MEASUREMENT CONSIDERATIONS

The NDA of a specific process component involves the measurement of the radiation emitted by material potentially within the component to determine the amount and composition of the material. Neutron and gamma-based NDA techniques are both useful in the assessment of radioactive material quantities. Gamma-ray spectroscopy techniques provide several advantages over neutron-based NDA techniques in holdup measurements. Gamma-based techniques provide spatial resolution through collimation and strategic shielding geometries. These techniques allow for isolation of deposits of interest from nearby deposits and allow for background reduction. In addition, gamma-spectroscopy allows for the identification of multiple isotopes simultaneously by their photopeaks, providing information useful for quantification of the isotopes present in the deposit. The primary gamma energies for the uranium isotopes are presented in Table 1. Figure 1 presents the gamma spectra for thick samples of U_3O_8 with different enrichment as measured with a sodium iodide detector and with constant ^{238}U content. Figure 2 shows the response of a high-purity germanium detector to similar materials.

Measurements of neutrons and/or gammas emitted from a given component provide a means to detect and characterize uranium deposits within individual cascade components. The measurement of neutrons for NDA is primarily useful when the gammas associated with held up uranium materials are sufficiently attenuated or shielded by the larger process components. Neutron-based techniques have the advantage of greater penetrability in bulk media but lack spatial resolution and isotopic specificity. Also, neutron-based NDA techniques are complicated by their sensitivity to reflection, moderation, multiplication, and (α,n) reactions with light matrix materials. The primary neutron production rates for U, UF_6 and UO_2F_2 are presented in Table 2.

Table 2 shows that for lower enrichments, the majority of spontaneous fission neutrons come from the spontaneous fission of ^{238}U. The largest source of neutrons is $^{19}F(\alpha,n)^{22}Na$ reactions that are driven by the ^{234}U decay alphas. The $^{19}F(\alpha,n)^{22}Na$ reaction accounts for approximately 58% of total neutrons from natural UO_2F_2. This fraction of total neutrons due to the $^{19}F(\alpha,n)^{22}Na$ reaction increases rapidly as the uranium is enriched in ^{234}U and, to a lesser extent, ^{235}U. The total neutron production rate of UO_2F_2 increases by a factor of 30 as UO_2F_2 is enriched from 3% to 97% ^{235}U. The sensitivity of this factor to uranium enrichment provides a useful tool to aid the determination of uranium mass and enrichment.

Table 1 Gamma Radiation from Uranium Isotopes

Isotope	Gamma-Ray Energy (keV)	Specific Intensity (γ/s-g of isotope)
^{232}U	129.1	6.5×10^8
	270.5	3.0×10^7
	327.8	2.7×10^7
^{233}U	119.0	3.9×10^4
	120.8	3.2×10^4
	146.4	6.6×10^4
	164.6	6.4×10^4
	245.3	3.8×10^4
	291.3	5.8×10^4
	317.2	8.3×10^4
^{234}U	120.9	5.4×10^5
^{235}U	143.8	7.8×10^3
	163.4	3.7×10^3
	185.7	4.3×10^4
	202.1	8.0×10^2
	205.3	4.0×10^3
^{238}U	742.8	7.1
	766.4	2.6×10^1
	786.3	4.3
	1001.0	7.5×10^1

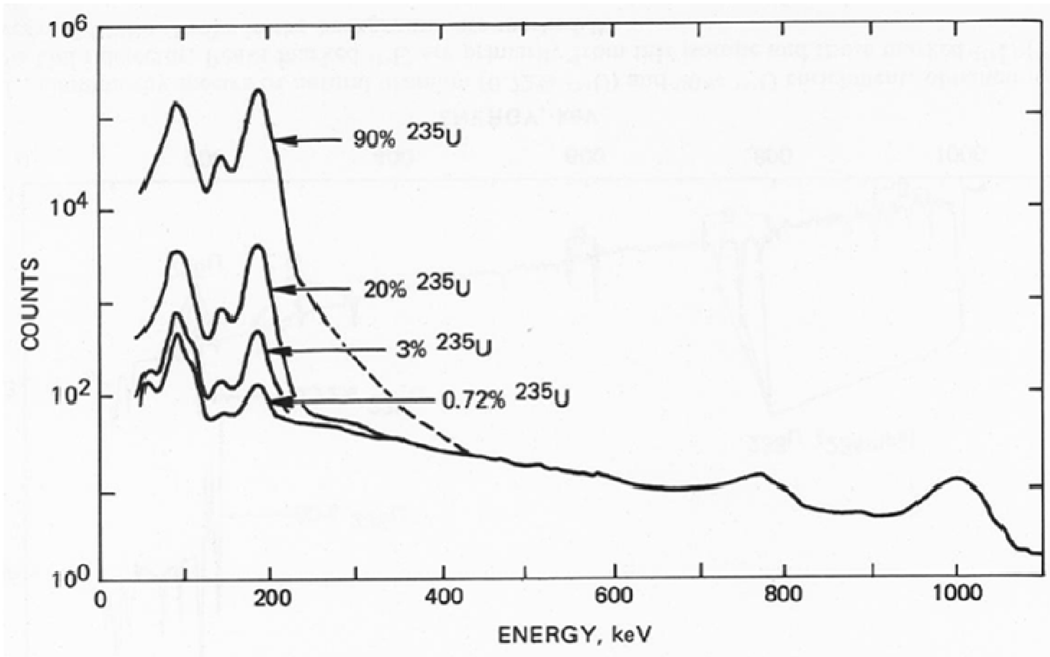

Figure 1 Response of a NaI detector to uranium in U_3O_8 with constant ^{238}U at various enrichments.[1]

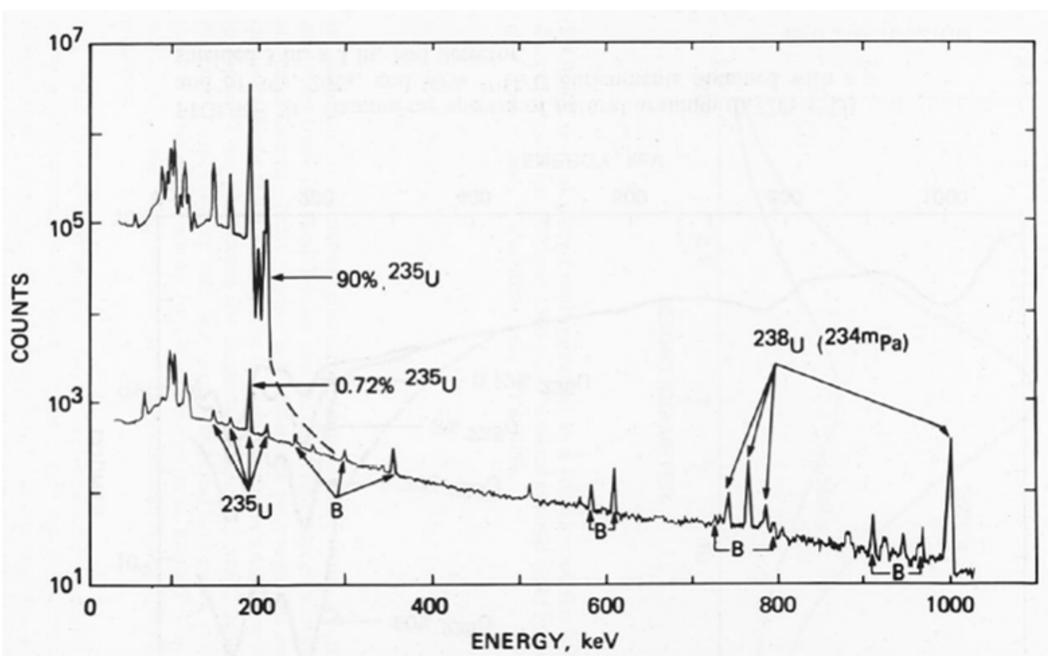

Figure 2 HPGe response to natural and enriched uranium in U_3O_8 with constant ^{238}U.[1]

Table 2 Primary neutron production rates in UO_2F_2 and UF_6 for several uranium isotopic compositions[2]

Isotope	Amount (wt%)	Neutron Production Rate for 10-kg of U (n/s)		
		Metal (spontaneous fission)	UO_2F_2 (α,n)	UF_6 (α,n)
^{234}U	0.0005	0	9	29
^{235}U	0.1977	0	1	2
^{236}U	0.0036	0	0	1
^{238}U	99.8	136	175	279
	Totals	136	185	311
^{234}U	0.0049	0	90	284
^{235}U	0.7108	0	3	6
^{236}U	-	-	-	-
^{238}U	99.28	135	174	278
	Totals	135	267	568
^{234}U	0.0244	0	449	1415
^{235}U	3.001	0	11	24
^{236}U	0.0184	0	2	5
^{238}U	96.96	132	170	271
	Totals	132	632	1715
^{234}U	0.0865	0	1592	5017
^{235}U	18.15	1	65	145
^{236}U	0.2313	0	28	67
^{238}U	81.53	111	143	228
	Totals	112	1828	5457
^{234}U	0.1404	0	2583	8143
^{235}U	31.71	1	114	254
^{236}U	0.3506	0	42	102
^{238}U	67.80	92	119	190
	Totals	93	2858	8689
^{234}U	0.2632	0	4843	15265
^{235}U	57.38	2	207	459
^{236}U	0.5010	0	60	145
^{238}U	41.86	57	73	117
	Totals	59	5184	15986
^{234}U	0.3338	0	6142	19360
^{235}U	69.58	2	250	557
^{236}U	0.5358	0	64	155
^{238}U	29.55	40	52	83
	Totals	42	6508	20155
^{234}U	1.032	1	18989	59856
^{235}U	97.65	3	352	781
^{236}U	0.2523	0	30	73
^{238}U	1.07	1	2	3
	Totals	5	19373	60713

4.0 CALIBRATION OF NDA SYSTEM WITH WORKING REFERENCE MATERIALS

Calibration of NDA measurement systems requires calibration through the use of traceable standards, hereafter referred to as Working Reference Materials (WRM). Calibration of the NDA system is required to establish a known and traceable relationship between the response of the NDA instrument and a WRM configuration. This is necessary to demonstrate a traceable calibration and to establish the uncertainty in the measurement result at a given level of confidence.

It is important that NDA calibrations are representative of the actual process items being measured. Deviation of the physical properties and characteristics of actual measurement items from the calibration configurations increase uncertainty in the measured results. Considering the diversity of chemical and physical characteristics of the holdup material in the PORTS cascade, it is impractical to maintain sufficient quantities and diversity of calibration standards representative of all probable deposit conditions to be assayed. This issue can be mitigated by developing an understanding of the effects of chemical and physical deviation of the potential deposits to be assayed from the configuration of the limited number of WRMs. This can be achieved by developing sufficient WRMs in a variety of physical and chemical configurations that bound probable configurations expected within the facility. A more practical solution is to use numerical simulations of NDA measurement techniques and configurations to develop “correction factors” that can be applied to the NDA system response for configurations that have no physical standards. The utilization of numerical simulation of NDA measurements provides for the reduction in the necessary number of NDA standards to a manageable number.

5.0 MODEL DEVELOPMENT FOR GAMMA-BASED APPLICATIONS

NDA measurement of the WRMs is used to provide confidence that the NDA measurement system is performing acceptably in correctly quantifying the known WRM. However, utilization of the measuring system on other geometries and/or source configurations introduces uncertainty in the measured results. Computer models are developed to predict detector response changes for holdup measurements of process items. Changes in detector response, relative to the response for measurement of WRMs are introduced by changes in the characteristics of holdup deposits or wastes.

Although straightforward in concept, development of an accurate computer model is not a one step procedure. First the computer model must be “benchmarked” to the response of the NDA detector through the use of applicable traceable WRMs in known configurations. The term benchmarked is used to indicate that the computer model can reproduce the NDA system response at a specified confidence level. Secondly, the limits of modeling in terms of the magnitude of deviation from benchmarked configuration(s) to other measurement item configurations of interest must be determined and bounded.

Theoretically once the model is benchmarked, any configuration could be modeled and the results expected to be accurate. Practically speaking though, the model needs to be verified as the measurement item geometry and composition begins to significantly depart from the base benchmarked configurations. What “significantly departs” means must be determined to ensure the model is not being used in a manner where the confidence in the result is degraded. There are also additional factors affecting NDA system response including; internal detector signal noise, detector count rate, environmental conditions, detector efficiency, etc. that should be considered.

5.1 NDA System Computer Model Benchmarking

The first step is to determine the response of the NDA system to a traceable WRM. A computer model is developed of the detector, the measurement item, and their geometrical relationship. The detector is modeled in great detail using manufacturer data and specifications. The first benchmark case is typically something simple such as a point source or cylinder of radioactive material. Comparison of the simulated detector response results with results from physical measurements using the NDA measuring system provides the basis for benchmarking the computer model of the detector system. The computer model simulation results and the physical measurements should agree within statistical uncertainty of the data. Several iterations of the model may be required to get good statistical agreement between the physical detector and the modeled detector response. Differences that may affect detector efficiency and require consideration are differences due to manufacturers' specifications and dimensions that are not exact, dead regions in the detector, signal processing losses, background, etc.

When the detector response and the model output are statistically the same the computer model can then be said to be "benchmarked" to the detector for that particular measurement configuration. At this point there is a degree of confidence in the computer model for the benchmarked measurement configuration. Once the computer model is benchmarked to a simple configuration, the computer model is adapted to more practical and useful configurations. The configurations expected to be encountered at PORTS includes waste drums, boxes, process equipment and a variety of in-situ configurations.

WRMs in various combinations and geometries are used to mimic actual in-process deposit holdup configurations. Each new model configuration, i.e., detector/standard system, is benchmarked against physical measurement results as previously discussed. Each new WRM configuration is measured by the NDA detection system at a known distance and the ^{235}U mass is determined. Should the NDA system produce a ^{235}U mass that differs from the known traceable ^{235}U mass of the WRM(s), adjustments or correction factors can be applied to the NDA data reduction routine to achieve the proper value. At this point the NDA system is calibrated for the particular WRM configuration and its known ^{235}U content.

Once satisfied the measurement system is yielding correct results for the known WRM, comparison is made between the measured and calculated response functions. The computer simulations utilize detailed geometric models to minimize any NDA system/model differences. Adjustments to the computer model geometry may be necessary to reduce uncertainty and/or error in the results that may be caused by simplifying or incorrect assumptions in the model geometry. The NDA measurement results and the results calculated by the computer model are compared to ensure that the computer model accurately simulates the calibrated measurement system response. Statistically similar results yield a benchmarked computer model measurement configuration, represented by the WRM.

The NDA measurement system that is calibrated to a given set of measurement configurations may be expected to accurately interpret the radiation signature of other components of the same or similar configurations. The value of this calibration may be limited for actual measurements in the PORTS process where measurement configurations vary over several important parameters, including deposit geometry, thickness, enrichment, etc. Also there is a large number and variety of components of vastly different geometries and shielding characteristics. Though a number of various WRMs are being developed for use in assessing holdup in the PORTS cascade, they can not effectively represent all configurations. Computer models are developed and simulations are run to fill this need.

Computer models that have been benchmarked for a specific NDA measurement system and configuration as described previously can be modified to estimate the NDA system response to differing measurement items, i.e., other cascade components. As an example, we consider the case of a NDA detection system that has been calibrated to a WRM that represents a full waste drum of known configuration and radioactive material content. This detection system can be expected to yield correct results for full waste drums with similar contents. There are several types of deviations from this configuration that will decrease confidence in system response to other drums. These deviations include differences in density of the drum contents and the geometry of the material within the drum. For each of these deviations, computer models may be developed to simulate system response to these new configurations and to develop correction factors to be applied to the calibrated NDA measurement system to account for the deviations. For example, we develop a model for a drum that is only half full to estimate the measurement system response and determine the correction factor required to produce correct results for a NDA measurement system that is calibrated to a full drum. In this example, the benchmarked model was utilized to produce a correction factor for the NDA system for which there was no half full drum calibration standard.

5.2 Statistical Bounding Calculations

Once a computer model has been benchmarked for a given configuration, say a large I.D. pipe, variation in the configuration of the pipe can be performed and the associated effect on NDA system response evaluated. Such variations could include deposit location, deposit geometry, uranium compound, enrichment, pipe wall thickness, pipe diameter, etc. A set of models is then specified that statistically represents and bounds all probable configurations. This requires a good deal of process knowledge to ensure all probable configurations are considered or bounded in the study.

Having specified a set of source/pipe configurations, a computer based NDA response determination can be made for the entire set. The mean bias and variance of the result set can then be determined relative to the benchmarked case for which the NDA system response is known. In this manner an overall bias correction factor and uncertainty for the entire set could be applied to the NDA measurements of such items.

Should the overall uncertainty associated with the entire set exceed that required for disposition or other management activities, the total set can be broken into logical subsets. For instance, variation in pipe diameter, wall thickness and enrichment could be removed from the set such that the only variables are deposit location, deposit geometry and uranium compound. This would then allow determination of a bias correction factor and uncertainty that is limited to one pipe diameter and wall thickness. The uncertainty in the NDA bias correction factor would be reduced but only applicable to pipe of that diameter/wall for which the calculations were performed.

Extending this concept, bias correction factors and uncertainties could be determined for any subset of the overall probable set initially specified. For example, say it is known for a certain size pipe that the enrichment and uranium compound are fixed or well known. For this case then a bias correction factor and uncertainty could be computed that considers only variations in deposit geometry. This again produces another bias correction factor that is applicable to specific configurations. Bias correction factors for numerous configurations could be modeled and applied as appropriate to NDA measurement results. The key to this approach is that all probable configurations and bounds are technically justifiable and defensible.

6.0 MODEL DEVELOPMENT FOR NEUTRON-BASED APPLICATIONS

The use of neutron-based NDA measurement methods may be used when necessary to measure holdup in the larger equipment of the PORTS cascade, including the converters, compressors, coolers, and large valves. These materials may be too dense to allow decay gammas to sufficiently penetrate to utilize gamma-based measurement technology. Neutrons penetrate metal and large holdup deposits better than gammas but require more nuclear material to produce a statistically significant signal.

Utilization of computer modeling and simulation of neutron-based NDA measuring systems requires benchmarking of calibrated detectors and WRMs. This benchmarking is performed in a similar fashion as described for gamma-based detection techniques. However, the modeling and measurement of neutrons introduces some additional complexity over the gamma modeling and measurement techniques. Generally neutron measurements are more difficult to interpret due to lack of a unique energy. Due to the penetration ability of neutrons, neutron detectors are difficult to collimate. Neutrons are subject to multiplication and moderation by even trace impurities. The primary isotope of concern, ^{235}U, produces very few neutrons, so knowledge of the relative concentrations of isotopes present is an important parameter that must be considered when estimating ^{235}U mass, U mass and the uncertainty of each. In addition, the form of deposit and its hydration affect the number of neutrons produced per unit time per unit mass of material. This also affects uncertainty estimates.

In performing neutron-based NDA, it is imperative that comprehensive computer models be developed that includes accurate cross-section data for all nuclear reactions for all materials in the deposit, the cascade component, and in the neutron detectors. The models must account for neutrons generated by spontaneous fission of uranium isotopes and neutrons generated by the $^{19}F(\alpha,n)^{22}Na$ reaction. The computer model must also provide adequate handling of these reactions as well as good data libraries for addressing temperature sensitive lattice effects of the detector moderators at thermal neutron energies.

7.0 COMBINATION OF NDA MODALITIES

The ideal NDA measuring system will utilize a combination of gamma- and neutron-based signals to provide the most comprehensive understanding of the deposit material being evaluated. The combined response from both neutron and gamma modalities will yield more information and measurement result confidence than either separately.

This moves the utility of modeling up in sophistication in that now there would be a benchmarked model for both a gamma and neutron type NDA system for a given configuration. The first step is to determine what configurations would benefit from combining gamma and neutron measurements. Clearly neutron measurements yield more information for thick deposits and massive process components than do gamma measurements. Conversely, gamma measurements are much more sensitive and accurate to lower mass deposits in configurations where there is relatively little attenuation.

One possible scenario for determination of when to use gamma, neutron, or a combination of the two would be to run neutron transport calculations for all of the models that have been developed for gamma NDA. An algorithm or Figure of Merit could be devised indicating when modalities should be combined to lower the NDA measurement uncertainty.

8.0 CONCLUSION

Computer modeling and simulation of detector response provides a prolific tool for decreasing the uncertainty associated with NDA measurements. Computer modeling allows for the adaptation of existing models that have been benchmarked against NDA standards to any number of configurations for which there are no physical standards. For configurations that are bounded by several existing standards, modeling allows for development of correction factors that are applied to physical detector responses from measurements of these configurations. Computer modeling provides the means for extrapolating the boundaries of existing WRM configurations thereby decreasing overall measurement uncertainty.

Computer modeling and simulation is an important tool useful during the development and use of NDA calibration configurations. Modeling allows optimization for material selection, source material design, and source utilization. Parametric computer modeling studies of physical systems provides a relatively inexpensive tool for evaluating optimal detector positioning to maximize detector response to minimize uncertainty.

9.0 REFERENCES

[1] MLM-3009

[2] Nureg/CR-5550