INEL-96/00295
INEL--96/00295

A PRELIMINARY EVALUATION OF CERTAIN NDA TECHNIQUES FOR RH-TRU CHARACTERIZATION

CONF-970126--16

J. K. Hartwell, W. Y. Yoon, and H. K. Peterson
Idaho National Engineering Laboratory
Idaho Falls, ID 83415

ABSTRACT

This report presents the results of modeling efforts to evaluate selected NDA assay methods for RH-TRU waste characterization. The target waste stream was Content Code 104/107 113-liter waste drums that comprise the majority of the INEL's RH-TRU waste inventory.

Two NDA techniques are treated in detail. One primary NDA technique examined is gamma-ray spectrometry to determine the drum fission and activation product content, and fuel sample inventory calculations using the ORIGEN code to predict the total drum inventory. A heavily shielded and strongly collimated HPGe spectrometer system was designed using MCNP modeling. Detection limits and expected precision of this approach were estimated by a combination of Monte Carlo modeling and synthetic gamma-ray spectrum generation. This technique may allow the radionuclide content of these wastes to be determined with relative standard deviations of 20 to 55% depending on the drum matrix and radionuclide.

The INEL Passive/Active Neutron (PAN) assay system is the second primary technique considered. A shielded overpack for the 113-liter CC104/107 RH-TRU drums was designed to shield the PAN detectors from excessive gamma radiation. MCNP modeling suggests PAN detection limits of about 0.06 g ^{235}U and 0.04 g ^{239}Pu during active assays.

INTRODUCTION

Remote handled transuranic (RH-TRU) waste is presently being retrievably stored at the Idaho National Engineering Laboratory (INEL). In compliance with Department of Energy (DOE) guidelines this waste has been stored in the Intermediate Level Transuranic Storage Facility (ILTSF) at the Radioactive Waste Management Complex (RWMC). This RH-TRU waste will eventually be retrieved and shipped to Waste Isolation Pilot Plant (WIPP) disposal facility, which is the located near Carlsbad, New Mexico. Each drum containing RH-TRU waste must be characterized and certified for shipment to and disposal at the WIPP repository.

This work evaluates selected nondestructive assay (NDA) technologies that may be able to characterize the primary INEL RH-TRU waste stream, Content Code 104/107 (CC104/107), in a cost effective manner. The NDA approaches evaluated are (1) direct gamma-ray spectroscopy for fission product content in combination with the calculation of fissile and other important nuclide contents using the ORIGEN fission product buildup and decay code; and (2) a combined passive-active neutron coincidence and dieaway measurement.

The bulk ((90%) of the INEL's RH-TRU wastes are CC104/107 wastes from Argonne National Laboratory-East (ANL-E) shipped in 30-gal DOT-17H drums. The contents of these are primarily alpha-gamma hot-cell wastes from ANL-E. They are primarily from metallurgical examination of Experimental Breeder Reactor II (EBR-II) irradiated fuel samples sent to ANL-E from ANL-West. The waste packages have a surface gamma radiation reading that does not exceed 30 R/hr. If a package reads greater than 30 R/hr, it is repackaged before shipment. These radiation fields arise from fission products in the irradiated fuel samples. The fuel material can be ^{235}U, ^{239}Pu as metal or ^{239}Pu as oxide. The Pu also contains ^{240}Pu and other Pu isotopes. The wastes are largely materials used in cleanup such as rags, paper, plastics, bits of metal, small tools or other contaminated hardware. Combustible materials are segregated from non-combustibles. No fuel sample materials are discarded as waste. All fuel remnants are returned to ANL-W. The TRU and fission products are simply contaminants in the waste from hot-cell cleanup after an examination campaign. The TRU arises from sawing and grinding operations and metallurgical mount preparation of samples. During the process, special effort is made to collect all grinding filings and saw cuttings. Also, as much material as possible is removed from grinding papers prior to discarding as waste. The cuttings and filings are NOT discarded as waste. For shipment of these wastes to the INEL's Radioactive Waste Management Complex (RWMC) the TRU content in the waste is estimated from process knowledge and historical loss rates (1½% of the TRU mass) with no direct assay or sampling of the waste. Typically these values are from 0.06 to 1.6 grams per 30-gal drum; however, in a few cases they can be as large as 16 grams.

The waste is generally placed into 7.5-gal containers (approximately 1 ft^3) and two of these stacked vertically in the center of a lined 30-gal drum. A given drum is loaded either with combustible or noncombustible wastes. The weight of the waste materials ranges from 11 to 66 kg (24 to 145 lb.), but is typically around 45 kg (100 lb.). Examination of the INEL storage records shows that 56% of the RH-TRU waste drums have surface radiation fields below 5 R/hr, 72% are less than 10 R/hr, and 84% are less than 20 R/hr. Only 27 drums have fields between 20 and 30 R/hr. None of the CC104/107 drums have a field above 30 R/hr.

Actual RH-TRU drum handling is expensive. This report relies on calculational models to evaluate the capability of the proposed NDA techniques, and to select systems for further development and testing. The modeling results are expected to provide a "road map" for future development and assessment measurements, and thus improve the cost effectiveness of the overall RH-TRU NDA characterization effort.

THE MODELING APPROACH

This work used the ORIGEN[1] code for fission and activation product inventory calculations; the MCNP[2] code for neutron and photon transport modeling; and the SYNTH[3] code for modeling of the gamma-ray spectral response. Since nearly 90% of the INEL's RH-TRU inventory is CC104/107 waste from ANL-E, this waste stream was chosen as the design basis case for NDA equipment evaluation.

Generally, waste packages with high surface dose rates present the greatest NDA challenge. Thus, the design basis drum was chosen to be an CC104/107 waste package containing 45 kg of homogeneously contaminated combustible waste with a density of 0.4 g/cm^3. The drum surface dose rate was chosen to be 30 R/hr, the maximum dose rate for this waste stream. The isotopic composition of the radioactive waste was presumed to be that of 15-year aged EBR-II fuel drawn from an ORIGEN inventory calculation computed for Pin DP-81 from a representative EBR-II irradiation.[a] The absolute drum content was that amount determined by MCNP calculations to yield a 30 R/hr gamma-ray dose rate at the exterior drum surface. The design basis drum contents are presented in Table 1.

Table 1. Radionuclide content of the RH-TRU design basis drum

Isotope	*Half-life (Yr)*	*Curies per drum*
^{54}Mn	0.85	3.72E-05
^{60}Co	5.27	1.91E-04
^{106}Ru(^{106}Rh)	1.02	6.69E-04
^{125}Sb	2.76	2.15E-02
^{134}Cs	2.07	3.35E-03
^{137}Cs	30.2	6.11E+00
^{144}Ce(^{144}Pr)	0.78	2.96E-04
^{152}Eu	13.5	3.12E-05
^{154}Eu	8.59	5.61E-03
^{155}Eu	4.71	5.24E-02

PASSIVE AND ACTIVE NEUTRON ASSAY

Neutron detection systems are generally the systems of choice for the direct measurement of fissile material content in bulk samples with a high gamma-ray dose rate. The INEL routinely uses a

[a] Inventory calculations were performed by K. Bunde (ANL-W) using ORIGEN-R, an ANL version of the ORIGEN code, and a proprietary set of EBR-II-specific libraries. Pin DP-81 was part of assembly S/A X-447 which accumulated 17.766 GWD from Nov. 1987 through Nov. 1988.

Passive/Active Neutron (PAN) assay unit to quantify the TRU content of contact-handled (CH)-TRU waste drums. A preliminary assessment suggested that the INEL's PAN system could possibly be used for the assay of RH-TRU waste drums.[4]

The INEL's PAN system is designed to assay TRU in 55-gal steel drums. It consists of an enclosed assay chamber, neutron shielding and moderator materials, ^{3}He neutron detectors, and a 14 MeV (D,T) neutron generator. The enclosure incorporates a motor-driven turntable that rotates a drum during assay. The system can be operated with the neutron generator running (Active Mode), or without the generator (Passive Mode).

Although the existing PAN system, since it uses ^{3}He neutron detectors, is quite tolerant of high gamma-ray backgrounds, the RH-TRU waste drums may require shielding in order to avoid an excessive accidental rate due to gamma-ray pile up events in the neutron detectors. Experience[4] suggests that the surface gamma-ray dose rate of a drum to be assayed should be below about 50 mR/hour. Since the PAN assay chamber is sized for 208-liter (55-gallon) waste drums, a shielded overpack was designed to reduce the dose rate of the 113-liter (30-gallon) RH-TRU drums to less than 0.05 R/hr. The thickness of lead shielding required to reduce the dose rate of the design basis RH-TRU drum (30 R/hr surface dose rate) to no more than 50 mR/hr was determined using the MCNP code. This model predicts that a 4.4-cm (1.75-in) thick lead shield provides the required dose rate reduction. A shield of this thickness can be accommodated within the PAN assay chamber. The PAN system turntable may require replacement to support the increase weight of drum and shield.

INEL researchers have developed a detailed MCNP model to predict the response of the PAN system.[5] This model was employed to estimate the detection sensitivity of the PAN system for assay of RH-TRU waste drums in a 4.4 cm lead overpack. Both active and passive assay modes were modeled; although, it was expected that only the active mode would be useful.

MCNP modeling estimated the response of the passive-mode PAN system to the design basis combustible matrix RH-TRU drum. The model assumed a waste density of 0.4 g/cm^{3} containing homogeneously distributed fissile material masses of 0.459 g ^{235}U, 0.068 g ^{239}Pu, and 0.008 g ^{240}Pu. These masses were determined from the typical CC104/107 RH-TRU drum inventories. The expected passive counting rate in the shielded detectors was 0.0157 counts per source neutron and 0.0955 counts per source neutron in the shielded plus unshielded (system) detectors. Converting these results to counts per gram of ^{240}Pu (using the neutron emission rate of 1.02E+03 n/s/g ^{240}Pu) and total Pu (using the assumed ^{240}Pu to Pu mass ratio of 11.7%) the expected shielded detector counting rates are 16 c/s/g ^{240}Pu and 1.9 c/s/ g Pu. In the system total detectors these values are 97 c/s/g ^{240}Pu and 11.4 c/s/g Pu. These

are total counting rates not coincidence counting rates. Only the neutron response was modeled. Photon interactions were not modeled.

The expected passive detection limit can be estimated from published values and the RH-TRU-specific MCNP modeling. For "benign" matrices, the existing PAN passive system has a detection limit of about 1g of weapons grade Pu in a 200 second count.[5] MCNP modeling of the existing system predicts a systems total counting efficiency of 0.127 counts per source neutron.[b] Assuming the same sample-induced background and a systems total counting efficiency of 0.0955 counts per source neutron, an estimated passive detection limit for the EBR-II-grade Pu would be about 0.8 grams.

The passive detection limit for Pu in the RH-TRU waste drums is greater than the usual estimated content. Additionally, this detection technique will not detect ^{235}U, often the highest concentration fissile isotope in these waste drums. To assay these drums will require active interrogation.

MCNP modeling of the overpack-shielded design basis RH-TRU drum predicts active mode counting rates in the time-correlated window of 0.38 c/pulse/g ^{235}U and 0.64 c/pulse/g ^{239}Pu. Thus, in a typical 2000 pulse assay, one expects 765 counts/g ^{235}U and 1280 counts/g ^{239}Pu. For comparison, similar sensitivity values for the 208-liter PAN assays, without the shielded overpack, are 1.6 to 1.9 c/pulse/g ^{239}Pu. The decrease of roughly a factor of three in detection sensitivity is believed due to neutron scattering in the shield, and the smaller geometry of the interrogated sample.

During the SWEPP PAN system performance demonstration testing, background rates in the accidentals gate were recorded for active assay of Pu-containing drums.[b] Six replicate runs were recorded on both a 208-liter waste drum empty except for the test sources, and on a drum containing Ethafoam©, a combustible matrix surrogate with a density of about 0.4 g/cm^3. Each run was for 40 second (2000 generator pulses). The mean counts in the accidentals gate were 502±34 counts for the empty matrix drum and 580±24 counts for the Ethafoam matrix drum. Scaling these to correct for the factor of five difference in gate time, suggests that a typical active assay background in the time-correlated channel would be about 110 counts in an assay period. The resultant detection limit[6] [(LD)] would be about 52 counts per assay period. Given the assay sensitivities calculated for the CC104/107 RH-TRU drums this would translate into reliable (95% confidence) detection of 0.06 g ^{235}U or 0.04 g ^{239}Pu. CC104/107 RH-TRU drums are typically said to contain between 0.06 to 1.6 g of TRU. These quantities should be reliably detected by PAN system assays in a shielded overpack. However, some caveats are in order.

---

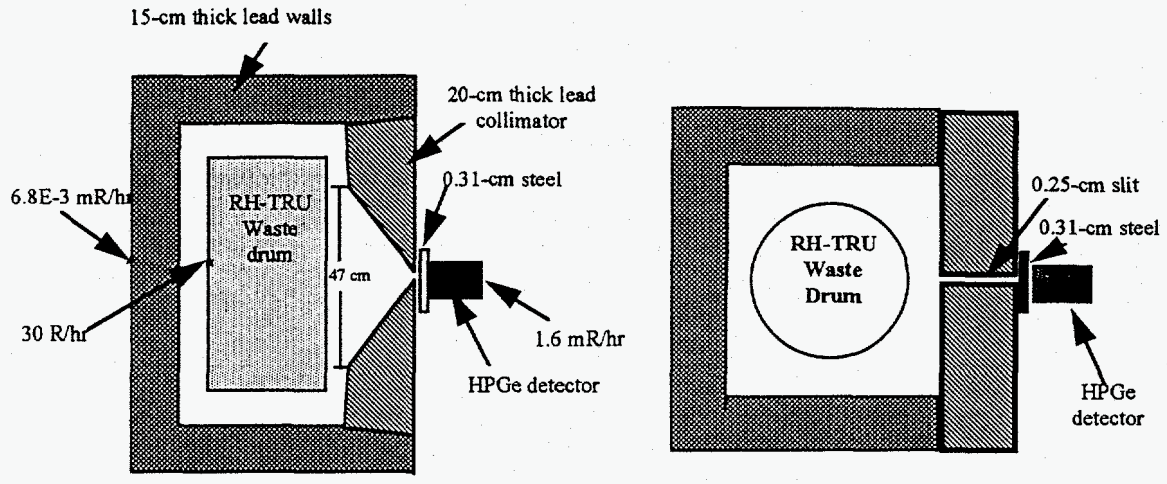
[b] Y. D. Harker, *Private Communication*, September 9, 1996.

PAN system active assays are sensitive to a number of waste-form-specific parameters. Among these are sample self shielding (if the fissile material is in "hunks"), matrix absorption, and matrix moderation. From our understanding of the CC104/107 wastes, sample self absorption should not be a problem; however, little is know about the other two parameters. These parameters cannot be well modeled. Careful calibration of the PAN system for the specific waste matrix will be required.

DIRECT GAMMA-RAY SPECTROSCOPY (DGS)

Since the irradiation history, but not necessarily the total radionuclide content of the INEL's CC104/107 RH-TRU drums is well known, their radionuclide content might be sufficiently determined by a gamma-ray spectrometric assay of the major fission and activation product emitters in the drum along with calculations using the ORIGEN code to determine the total content. One uncertainty in the present knowledge of the CC104/107 RH-TRU drums is the fraction of the total activity that is due to activation (^{60}Co, for example) rather than fission products (^{137}Cs and ^{125}Sb are examples). By defining the relative amounts of the detected gamma-ray emitters and providing an estimate of the total content of a few radionuclides, a DGS system could improve the accuracy with which ORIGEN can calculate the total drum inventory. Additionally, certain fission product activity ratios and total activity estimates would be useful for confirmation of the declared burn-up and decay history on which the ORIGEN inventory values rely.

A cutaway drawing of the preliminary design for a direct gamma-ray spectroscopy (DGS) system for assay of RH-TRU drums is presented in Figure 1. The DGS incorporates a high-purity germanium (HPGe) detector with standard pulse height analysis electronics and a relatively common coaxial detector design. In order to allow the assay of the high activity waste drums, the system includes massive shielding and strong detector collimation. Since the collimator defines a source area that covers a thin strip from the top to the bottom of the drum, the system will require drum rotation to provide a full view of the drum.



a. Vertical cross section

b. Horizontal cross section

Figure 1. Cutaway drawings of the proposed DGS shield and collimator

The shielding and collimation design features were derived from MCNP modeling.[c] A system requirement was that the total counting rate in the HPGe detector be less than 50,000 counts per second (c/s) when a design-basis combustible matrix RH-TRU drum with a surface dose rate reading of 30 R/hr was assayed. This insures that the total system input rate is compatible with standard gamma-ray spectrometer component rate-handling capabilities. Figure 1 is annotated with the MCNP-calculated dose rates at critical points.

The DGS collimator defines the viewed source area and limits the HPGe detector counting rate to levels that can be reliably processed by readily available nuclear spectroscopy equipment. As depicted in Fig.1, a fan-shaped collimator with an entrance height of 47 cm was chosen to provide a view of a full vertical slice of the rotating RH-TRU drum. The collimator width and exit height were adjusted such that the MCNP model predicted a detector counting rate of 50,000 counts per second (c/s) for this highest activity drum. The collimator width and exit height that provided this modeled counting rate were both 0.25 cm (0.1 in), and a 0.31-cm (0.12-in) thick steel absorber was used to block low-energy gamma- and X-rays.

In any strongly collimated system, shield penetration can be a problem. If the detector counting rate from degraded photons that have passed through the shield is a significant fraction of the total system counting rate, then the signal-to-background ratio can be untenably small. Shield leakage was assessed by

---

[c] Private communication, W. Yoon to Y. D. Harker, J. K. Hartwell, and H. K. Peterson, Notegram of 4/23/96 "Preliminary HPGe Detection System for the R-H Waste Drum," and additional calculations performed 7/19/96.

comparing the calculated dose rate at the detector face for the collimated DGS with identical calculations performed for a solid shielding wall (these are normally termed "collimator open" and "collimator closed" results). For the proposed DGS design, the open-to-closed ratio was greater than 70,000, a fully acceptable result.

Accurate modeling of gamma-ray spectra requires that the spectral continuum be correctly predicted. In the DGS, the continuum will have two primary components 1) the Compton continuum from scattering events in the detector, and 2) photons scattered (and thus degraded in energy) during transport to the detector. MCNP has the capability to predict the complete gamma-ray spectrum for this DGS problem, and the modeled results would include both of the continuum components in addition to the full energy peaks. However, the computer time required for this calculation would be excessive. Consequently, an abbreviated MCNP model was used to predict the general shape of the scattered photon component reaching the detector and the total detector counting rate, and a recently developed code, SYNTH, [3] modeled the expected spectrum.

SYNTH requires that a source configuration, external absorbers, source radionuclide concentrations, detector configuration, and electronic settings be defined as input. From these input parameters SYNTH uses accepted parametric calculational techniques to predict the expected gamma-ray spectrum. Comparisons of measured and SYNTH-calculated spectra have been published.[3,7]

MCNP modeling predicted that the total detector counting rate would be 50,000 counts per second for the design basis 30 R/hr drum and the chosen collimator design. About half of the photon flux reaching the detector was predicted to be full energy (uncollided) source gamma-rays, and the remaining half degraded in energy (collided). Additionally, the MCNP model predicted the energy spectrum (152 bins) of the collided and the uncollided portion of the photon flux reaching the detector.

The isotopic distribution of the source used for the SYNTH calculations was drawn from the EBR-II inventory data provided. In order to emulate the expected attenuation in the RH-TRU waste, the source was described to SYNTH as a 10 cm^2 disk source with a thickness of 10 cm and a mass of 40 grams. (This description is not meant to reproduce the RH-TRU counting geometry, but rather to force a proper shape to the SYNTH-calculated detector efficiency curve.) The source composition was chosen to be 96% C_2H_2, 2% iron, and 2% uranium. The source-to-detector distance was arbitrarily set to 12 cm. To account for attenuation through the container walls and steel absorber plate, an external absorber of 0.65 cm of iron was selected. The detector was described as a standard HPGe detector with a diameter of 5 cm, a depth of 6 cm, and a relative efficiency of about 20%. The spectral gain and zero were chosen to be 0.36

keV/channel and 0 keV respectively. This provides an upper energy limit of 2950 keV in an 8192 channel spectrum.

From these parameters, SYNTH was used to calculate an absolute photopeak efficiency curve. As a check on the accuracy of this SYNTH prediction, an MCNP calculation of photopeak counts in the detector per source photon was performed. Since the SYNTH calculation was provided with an arbitrary source concentration (to be normalized later) the absolute photopeak efficiency values cannot be directly compared; however, the correctness of the SYNTH-calculated efficiency curve as a function of energy can be evaluated by comparing the relative efficiency curve determined by MCNP with the relative efficiency curve calculated by SYNTH. Relative photopeak efficiencies were computed in both cases by normalizing each curve to an efficiency of 1.0 at the 1332 keV line of ^{60}Co. The results are compared in Table 2. Throughout the energy range of interest, the SYNTH-calculated curve agrees very well with the MCNP values.

Table 2. Comparison of MCNP and SYNTH-calculated relative detector efficiencies as a function of energy

Energy (MeV)	*Rel Eff (MCNP)*	*Rel Eff (SYNTH)*	*SYNTH/MCNP Ratio*
1.3325	1.00	1.00	1.00
1.1173	1.13	1.08	0.95
0.6616	1.66	1.77	1.07
0.6359	1.71	1.79	1.05
0.6006	1.78	1.80	1.01
0.4634	2.17	2.15	0.99
0.4435	2.25	2.24	1.00
0.4279	2.31	2.40	1.04
0.3804	2.52	2.44	0.97

Since the shape of the SYNTH-calculated efficiency curve with energy seems correct, the next step was to define the total activity of the SYNTH-specified source. The relative radionuclide distributions were as specified for 15-year decayed material (Table 1). Again, the MCNP modeling results guided the total activity specification. MCNP calculations predicted a total detector counting rate of 50,000 counts/second with about half of these counts coming from uncollided photons and about half from collided photons. Thus, the SYNTH-specified source activity was adjusted to yield a spectrum containing about 25,000 total counts/second, simulating the "uncollided" portion of the modeled spectrum. The collided photon spectrum, an important component of the spectral background, was also determined from MCNP calculations. The MCNP results provided a binned energy spectrum of the collided photon flux impinging on the detector end cap. To conserve calculation time, the calculation used only 152 energy

bins. The MCNP results were smoothed, converted from counts/keV to counts per channel using the known spectral energy calibration of 0.36 keV/channel, and then interpolated into 0 to 4096 channels. (There is no significant collided photon flux above 1408 keV or channel 3920.) The channel contents of this interpolated spectrum were further smoothed and adjusted by a constant factor to yield a total of 25,000 counts/second. The SYNTH-generated source photon spectrum and the estimated collided photon spectrum were summed using the "background addition" function of SYNTH. (This procedure ignores the effect of the detector relative efficiency curve on the collided photon spectrum, thus the synthetic continuum will be over estimated at the higher energies.) Statistical fluctuations were then imposed on the channel contents (another SYNTH function) to produce a best estimate of the expected RH-TRU assay spectrum. The resultant spectrum with the two summed components identified is presented in Figure 2.

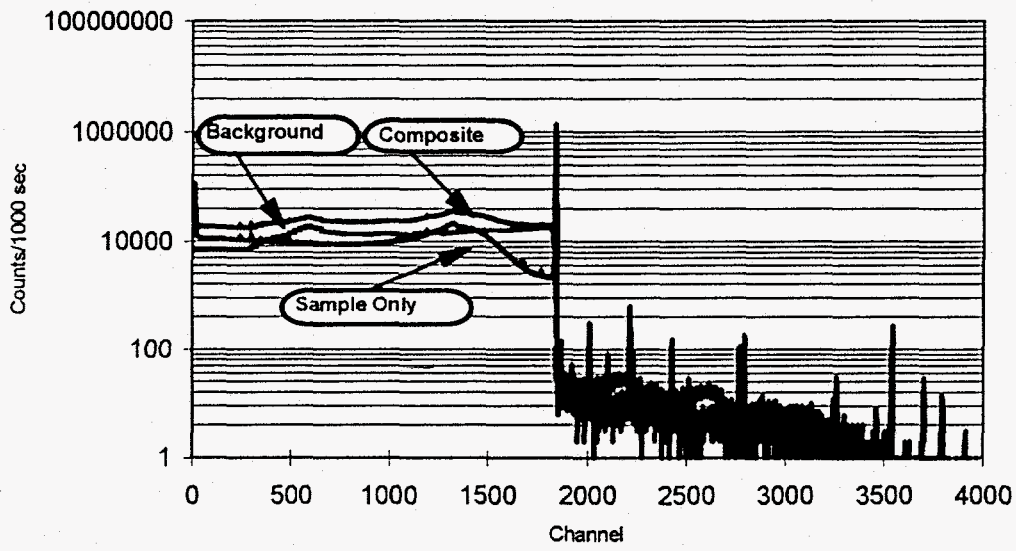


Figure 2. The simulated DGS spectrum detailing its components

The simulated DGS spectrum for the design basis RH-TRU drum was submitted to the commercial gamma-ray spectral analysis program *GammaVision.*© The analysis results provide the expected peak areas in a 1000 second count time spectrum, the uncertainties in these peak areas, and the total number of background (continuum) counts underlying each peak. These values were then used to determine the uncertainty in the spectral analysis results, and the expected system detection limits. Detection limits were computed following the technique of Currie.[6] Currie defines a critical level (L_C) and a detection limit (L_D). The critical level is the response that must be measured in a given spectrum in order to conclude "detected," while the detection limit is the response level that will result in a detected decision 95% of the time.

The expected peak areas, peak area uncertainties (as one relative standard deviation from counting statistics only), the spectral critical levels (L_C), and estimated detection limits at the 95% confidence level (L_D) are presented in Table 4. The detection limits are expressed both as peak area counts (in a 1000 second count time spectrum) and in terms of Curies of drum activity. Those spectral peaks for which the expected peak area exceeds the calculated L_D limit, and thus should be detectable 95% of the time, are identified as "Easily" detected.

It is important to note that the calculated detection limits are only correct for the design basis RH-TRU drum reading 30 R/hr at the drum surface and for a system assay time of 1000 seconds. The

Table 4. Spectral analysis results for the simulated DGS spectrum

Isotope	line energy (keV)	Est bkg (counts)	Est critical level LC (counts)	Est detection limit LD (counts)	Expected peak area (counts)	Relative Stand Dev	Detected?	Curies/ sample	Expected Counts/Sec per Ci	Detectable limit (Ci)
Co-60	1173	37	14	31	153	10%	Easily	1.9E-04	8.01E+02	3.9E-05
	1332	6	6	14	147	9%	Easily	1.9E-04	7.70E+02	1.8E-05
Mn-54	834	379	45	94	86	34%	Yes	3.7E-05	2.31E+03	4.0E-05
Rh-106	622	311675	1303	2608	621	127%	No	6.7E-04	9.28E+02	2.8E-03
Sb-125	176	229027	1117	2236	6577	10%	Easily	2.2E-02	3.06E+02	7.3E-03
	381	237029	1136	2275	914	75%	No	2.2E-02	4.25E+01	5.4E-02
	428	389703	1457	2916	12559	7%	Easily	2.2E-02	5.84E+02	5.0E-03
	463	421897	1516	3034	3801	24%	Easily	2.2E-02	1.77E+02	1.7E-02
	601	561290	1748	3499	4073	26%	Easily	2.2E-02	1.89E+02	1.8E-02
	636	293341	1264	2530	3098	25%	Easily	2.2E-02	1.44E+02	1.8E-02
Cs-134	563	608976	1821	3645	2038	54%	Yes	3.4E-03	6.08E+02	6.0E-03
	569	355548	1391	2785	3125	27%	Easily	3.4E-03	9.33E+02	3.0E-03
	605	338785	1358	2719	2628	31%	Yes	3.4E-03	7.84E+02	3.5E-03
	796	552	55	112	2992	2%	Easily	3.4E-03	8.93E+02	1.3E-04
	802	600	57	117	181	21%	Easily	3.4E-03	5.40E+01	2.2E-03
	1168	218	34	72	55	40%	Yes	3.4E-03	1.64E+01	4.4E-03
Cs-137	662	107790	766	1535	6.00E+06	0%	Easily	6.1E+00	9.81E+02	1.6E-03
Ce-144	133	205688	1058	2119	346	185%	No	3.0E-04	1.17E+03	1.8E-03
Pr-144	696	90	22	47	4	339%	No	3.0E-04	1.35E+01	3.5E-03
Eu-152	122	380290	1439	2881	1269	69%	No	3.1E-05	4.07E+04	7.1E-05
	344	252400	1172	2347	187	380%	No	3.1E-05	5.99E+03	3.9E-04
	779	83	21	45	5	262%	No	3.1E-05	1.60E+02	2.8E-04
	1408	6	6	14	8	56%	Yes	3.1E-05	2.56E+02	5.5E-05
Eu-154	123	365752	1411	2825	5217	16%	Easily	5.6E-03	9.30E+02	3.0E-03
	248	445642	1558	3118	613	154%	No	5.6E-03	1.09E+02	2.9E-02
	723	411	47	97	1174	4%	Easily	5.6E-03	2.09E+02	4.7E-04
	757	468	50	104	305	12%	Easily	5.6E-03	5.44E+01	1.9E-03
	873	226	35	73	673	5%	Easily	5.6E-03	1.20E+02	6.1E-04
	996	75	20	43	518	5%	Easily	5.6E-03	9.23E+01	4.7E-04
	1005	36	14	31	917	3%	Easily	5.6E-03	1.63E+02	1.9E-04
	1274	7	6	15	1468	3%	Easily	5.6E-03	2.62E+02	5.8E-05
	1596	5	5	13	60	14%	Easily	5.6E-03	1.07E+01	1.2E-03
Eu-155	86	177335	983	1968	7159	8%	Easily	5.2E-02	1.37E+02	1.4E-02
	105	178808	987	1976	11512	5%	Easily	5.2E-02	2.20E+02	9.0E-03

Compton continuum level and the scattered photon components will decrease as the source activity, and thus the total detector counting rate, decreases. A lowered continuum level will result in more sensitive detection of minor isotopic components. However, the expected peak areas will also decrease as the source activity declines. The measurement sensitivity increases with total counting time. For the design basis 30 R/hr drum the system detection limits would drop by factors of 7 to 10 with counting times of 50,000 seconds (14 hours) and 100,000 seconds (24 hours) respectively. For the limited number of RH-TRU drums to be processed, counting times of a day or so are probably not prohibitive.

In any NDA technique that uses gamma-ray spectral measurements to assay bulk samples, proper correction for gamma-ray attenuation in the assayed matrix can be critical to the success of the method, and can contribute strongly to the overall uncertainty of the assay. This is particularly true when the energy of the gamma-rays to be assayed is low (less than about 300 keV). Several approaches have been used successfully in similar situations,[8] including transmission source corrections, Monte Carlo modeling, relative efficiency calculations based on multiple detected gamma-ray lines, and geometric calculations. Since the actual waste packages for the CC104/107 RH-TRU wastes are small (1 ft^3), the contents relatively well defined, and the gamma-ray energies of primary interest are above about 600 keV, the contribution of the assumed attenuation corrections to the total DGS measurement uncertainty is expected to be acceptably low. The validity of this expectation is supported by the following scoping calculation.

For a different gamma-ray system for assay of TRU isotopic ratios in contact-handled (CH) drums, a review was conducted of the CH-waste drum contents, and a set of representative tables of gamma-ray mass attenuation coefficients calculated as a function of energy.[9] The waste matrix descriptions for the CH-handled combustible and mixed metals match closely our understanding of the CC104/107 RH-TRU waste. Thus the energy-dependent mass attenuation coefficients for these matrices were chosen for scoping calculations to evaluate the total magnitude and overall uncertainty contribution of the matrix attenuation correction for the design basis RH-TRU case.

The waste pails used to package CC104/107 wastes are 18.75 cm in radius. For an isotropic angular source distribution, this seems to be an appropriate attenuation pathlenght to use for correction calculations; however, the fan-shaped collimator suggested for the DGS allows a maximum path length through the matrix of 22 cm.

Although there are a number of techniques that could be employed to measure the effect of matrix attenuation on the DGS results, some initial calculations that assumed that no measured corrections to the DGS data were performed to guide the development of improved attenuation correction techniques. For these calculations it was presumed that each spectral result would be "blindly" corrected

using the mass attenuation coefficients of reference 9, the overall average waste density computed from INEL storage records, and a transmission path equal to the radius of the pails used to package waste in the 30-gal RH-TRU drums. The first set of calculations computed the appropriate attenuation correction for a set of gamma-ray lines ranging in energy from 600 keV (^{125}Sb) to 1332 keV (^{60}Co). Correction factors were calculated for the "average" set of waste densities and the assumed attenuation length of 18.75 cm, for a minimum waste density and an attenuation length of 18.5 cm, and for a maximum waste density and a maximum attenuation length of 22 cm. Attenuation correction factors were calculated both for the absolute correction of each gamma-ray to provide a total activity estimate, and for the correction of isotopic ratios relative to the 1332 keV line of ^{60}Co. The results are presented in Table 5.

Table 5. Calculation of matrix attenuation corrections for minimum, average, and maximum attenuation conditions for both combustible and noncombustible waste matrices

Combustible Matrix			*At Average Density*		*At Max Density*		*At Min Density*	
Isotope	*E (keV)*	*Att Coef*	*Corr Factor*	*Relative to 1332*	*Corr Factor*	*Relative to 1332*	*Corr Factor*	*Relative to 1332*
Sb-125	600	0.0919	1.24	1.06	2.27	1.22	1.07	1.02
	636	0.0879	1.23	1.05	2.20	1.18	1.07	1.02
Cs-137	662	0.087	1.23	1.05	2.19	1.18	1.07	1.02
Eu-154	723	0.0818	1.21	1.04	2.10	1.13	1.06	1.01
	1005	0.0726	1.19	1.01	1.96	1.05	1.06	1.00
	1274	0.0663	1.17	1.00	1.86	1.00	1.05	1.00
Co-60	1173	0.0663	1.17	1.00	1.86	1.00	1.05	1.00
	1332	0.0663	1.17	1.00	1.86	1.00	1.05	1.00

Noncombustible Matrix			*At Average Density*		*At Max Density*		*At Min Density*	
Isotope	*E (keV)*	*Att Coef*	*Corr Factor*	*Relative to 1332*	*Corr Factor*	*Relative to 1332*	*Corr Factor*	*Relative to 1332*
Sb-125	600	0.0758	1.49	1.12	3.16	1.34	1.15	1.04
	636	0.0739	1.47	1.11	3.09	1.31	1.14	1.04
Cs-137	662	0.0726	1.46	1.11	3.05	1.29	1.14	1.04
Eu-154	723	0.0696	1.44	1.09	2.94	1.24	1.14	1.03
	1005	0.0595	1.37	1.04	2.60	1.10	1.12	1.01
	1274	0.0528	1.32	1.00	2.38	1.01	1.10	1.00
Co-60	1173	0.055	1.34	1.01	2.46	1.04	1.11	1.01
	1332	0.0522	1.32	1.00	2.37	1.00	1.10	1.00

As expected, the differences between the assumed average correction and those at the minimum and the maximum attenuation conditions are greatest for the noncombustible matrix drums, and for the lower energy gamma-rays. For the maximum attenuation conditions the absolute attenuation correction factor for the ^{125}Sb 600 keV line would be underestimated by the average correction by a factor of about 2 (3.16/1.49) while at the minimum density conditions the absolute correction factor relative to the assumed

average would be overestimated by about 30% (1.15/1.49). (An overestimate of the attenuation correction factor would result in an overestimate of the absolute activity.) However, note that for this worst case scoping calculation, the correction for the ^{125}Sb to ^{60}Co activity ratios could be determined to within a factor of about 20%. For the combustible matrix drums the over and under estimates are of similar magnitude.

The results in Tables 5 and additional calculations using a Monte Carlo simulation[7] suggest that proper correction for matrix attenuation is not an overwhelming problem in the DGS measurements. If no attempt is made to calculate drum-specific attenuation corrections, other than to provide an estimated waste matrix density, the absolute correction for ^{137}Cs in the combustible matrix drums would have about a 1% high bias and a relative standard deviation of about 6%. The 95% confidence limits of this distribution are at about ±11%. As expected because of its higher gamma-ray energy, the results are somewhat better for ^{60}Co. The absolute correction for ^{60}Co in the combustible matrix drums would have about a 1% high bias (1.18/1.17) and a relative standard deviation of about 4%. The 95% confidence limits of the ^{60}Co distribution are at about ±9%. These results suggest that the ^{137}Cs/^{60}Co ratio can be determined with a bias of less than 1% and a relative standard deviation of about ±7%. The estimated results for the noncombustible matrix drums are somewhat less well predicted; however, the Monte Carlo simulation suggests that the attenuation correction for the137Cs/60Co ratio in these drums would have a high bias of about 2% and a relative standard deviation of about ±23%.[7]

The previous scoping calculations assumed that drum weight (and thus calculated matrix density) was the only drum-specific parameter used to predict the matrix attenuation correction factors. Although the results of these scoping calculations suggest adequate corrections can be made without additional measured data, there are techniques that can improve the estimated attenuation corrections for specific drums. Since the SYTH simulation predicts that a number of multiple lines from multi-gamma-ray emitting isotopes will be detected (for example the 123, 723, 873, 1005, and 1274 keV lines of ^{154}Eu) in the baseline drum, these results can be used to calculate a drum-specific attenuation correction directly. Since the gamma-ray emission probabilities for ^{154}Eu are well known, and the unattenuated detection efficiency will be determined, the multiple line results can be used to solve (probably most efficiently by iteration) the standard matrix attenuation equation for the product $\mu_i x_{eff}$ and the estimated standard deviation on this product. (To solve for both the value and its error requires a minimum of three points.) These measured results can then provide improved attenuation corrections; however, if the required multiple lines cannot be detected in certain drums then the previous scoping calculation defines the applicability of default corrections.

The uncertainty in the radionuclide results can now be estimated. The activity (A) in bequerels (d/s) in a waste barrel of isotope j computed from a gamma-ray line of energy i can be written as:

$$A_{ij} = \frac{(N_i)(C_{ai})}{(\varepsilon_i)(BR_{ij})}$$

where:

N_i = the measured photopeak counting rate at energy i (c/s)
C_{ai}= the matrix self absorption correction factor at energy i
ε_i = the detection efficiency of the system for a nonattenuating matrix (c/γ)
BR_{ij} = the gamma-ray emission probability for (i from isotope j

Using standard error propagation techniques, the standard deviation in A_{ij} can be estimated as (deleting the i and j subscripts for convenience):

$$\frac{\sigma_A}{A} = \sqrt{\left(\frac{\sigma_N}{N}\right)^2 + \left(\frac{\sigma_{C_a}}{C_a}\right)^2 + \left(\frac{\sigma_\varepsilon}{\varepsilon}\right)^2 + \left(\frac{\sigma_{BR}}{BR}\right)^2}$$

where the σ_i values are the estimated standard deviations of the parameters.

Estimated relative standard deviations and systematic errors for these parameters are available from experience, modeling, and nuclear data. Analysis of the SYNTH-generated spectrum provides estimates of N_i and σ_N (see Table 4). The matrix self-absorption correction values and their errors are available from the previous section. Experience with similar systems suggests that, in the energy range of interest, absolute nonattenuated detection efficiency values can be determined to an overall precision of less than 15%; while relative efficiency values (for the $^{60}Co/^{137}Cs$ ratio) can be much better determined, probably to ±3%. Gamma-ray emission probabilities and their associated errors are available from the nuclear literature. Their associated errors are generally 0.1% or less.

Similarly, for the isotopic activity ratios (R_{jk}) of isotope j to isotope k (using gamma-ray lines at energies of i and l respectively):

$$R_{jk} = \left(\frac{N_i}{N_l}\right)\left(\frac{C_{a_l}}{C_{a_i}}\right)\left(\frac{BR_{kl}}{BR_{ij}}\right)\left(\frac{\varepsilon_l}{\varepsilon_i}\right)$$ or, replacing $\frac{\varepsilon_l}{\varepsilon_i}$ with the relative efficiency $R\varepsilon_{il}$ and the ratio $\frac{C_{a_l}}{C_{a_i}}$ with the differential self absorption correction ratio DA_{il} then:

$$R_{jk} = \left(\frac{N_i}{N_l}\right)\left(\frac{BR_{kl}}{BR_{ij}}\right)(R_{\varepsilon il})(DA_{il})$$ and the estimated standard deviation computed as:

$$\frac{\sigma_{R_{jk}}}{R_{jk}} = \left[\left(\frac{\sigma_{N_i}}{N_i} \right)^2 + \left(\frac{\sigma_{N_l}}{N_l} \right)^2 + \left(\frac{\sigma_{DA_{il}}}{DA_{il}} \right)^2 + \left(\frac{\sigma_{R\varepsilon_{il}}}{R\varepsilon_{il}} \right)^2 + \left(\frac{\sigma_{BR\,kl}}{BR_{kl}} \right)^2 + \left(\frac{\sigma_{BR\,ij}}{BR_{ij}} \right)^2 \right]^{1/2}$$

Computing the estimated standard deviations for the absolute concentration of ^{137}Cs, ^{60}Co, and the activity ratio of $^{60}Co/^{137}Cs$ using the equations and data above yields an estimated relative standard deviation for the combustible matrix drums for the absolute concentrations of ^{137}Cs and ^{60}Co respectively of 18% and 17%, and of 12% for the activity ratio. For the noncombustible matrix drums these values are 24%, 20%, and 23% respectively.

ORIGEN INVENTORY CALCULATIONS

Whether the NDA technique of direct gamma-ray spectroscopy to quantify the fission and activation product content, or active neutron assay to determine the fissile material content are employed to characterize CC104/107 RH-TRU wastes, inventory calculations based on the known irradiation history of the fuel pins processed to produce the wastes in a particular drum will be required to specify total drum content. The expected uncertainty of these inventory calculations can be an important uncertainty contributor to the overall drum content determinations.

There is a paucity of published comparisons of calculated-versus-measured inventory results for the EBR-II reactor. While it is likely that such comparisons have been done, they do not appear in the open literature. Thus, all assumptions about the adequacy of ORIGEN fuel inventory predictions for EBR-II must be drawn by parallel to published data on light water reactors (LWRs). There can be a number of hazards in this approach. Just about every thing that one can think of that might affect inventory calculations -- fuel materials, fuel matrix, core size, neutron spectrum, flux and power densities, etc. -- of EBR-II are completely different from those of LWRs. However, the basic calculational techniques remain the same. Thus, *if the core neutronics of EBR-II are known at least as well as those for the LWRs studied, and this knowledge is translated into a proper cross-section library*, then inventory calculations for EBR-II should have uncertainties similar to those in the published set of LWR comparisons. Certain LWR comparison results are presented in this section. From the LWR data set uncertainty contributions to the characterization of CC104/107 RH-TRU wastes are estimated.

Inventory calculations performed using the ORIGEN-S code have been compared with radiochemically measured results for medium to high burnup fuel samples (nominally 30 GWD/MTU) from the US Yankee Rowe, Turkey Point, and H. B. Robinson LWRs.[10] In each case, the modeled power

level was adjusted to predict the radiochemically-measured atom percent burnup as determined from $^{148}Nd/^{235}U$ ratios. Comparison values for TRU inventories are reported for all three test cases; however, fission product comparisons are only reported for the H. B. Robinson assemblies. No inventory comparison results are reported for activation products. For all three reactors, the calculated ^{239}Pu concentrations were 3% to 6% greater than the measured. Conversely, the minor Pu isotopes (240-242) were generally under predicted 5% to 16%. The H. B. Robinson fission product comparison data is complicated by an analysis problem that biased the measured fission product concentration low by 15% to 25%. When corrected for a -20% analytical bias, the ORIGEN predictions agreed with the measured results to within -5.2% (for ^{137}Cs) to +14.9% (for ^{129}I). No measurement precisions were specified.

In the wake of the reactor accident at the Three Mile Island Unit 2 (TMI-2) power station a number of inventory calculations were performed to determine the core inventory.[11] TMI-2 shutdown occurred after very little fuel burnup (core average 3175 MWD/MTU). Inventory calculations were initially performed for the full core average, for each of three initial fuel enrichments, and for a set of multiple (1239) fuel zones grouped according to burnup and enrichment. When compared to measured concentration data on eight specific fuel pellets drawn from known positions on the core periphery (the core center having been destroyed), even the most detailed of these calculations differed from measurements by as much as 100%.[12] To better validate the inventory calculations, additional ORIGEN2 calculations were performed with the modeled power levels adjusted to match the radiochemically-measured burnup of each pellet. These results are reproduced in Table 6.

Table 6. Measured-to-calculated inventory ratios on eight specific fuel pellets from the TMI-2 reactor[12]

Isotope	*Measured/ Calculated*	*Stand Dev*[a]	*Isotope*	*Measured/ Calculated*	*Stand Dev*[a]
^{238}Pu	0.86	0.17	^{90}Sr	0.99	0.02
^{239}Pu	0.90	0.09	^{106}Ru	0.96	0.08
^{240}Pu	0.89	0.09	^{125}Sb	0.43	0.04
^{241}Pu	1.05	0.20	^{129}I	0.72	0.04
Total Pu	0.90	0.09	^{134}Cs	0.86	0.27
Total Kr	0.954	0.014	^{137}Cs	1.021	0.009
Total Xe	0.95	0.04	^{144}Ce	1.07	0.02
			^{154}Eu	0.63	0.06
			^{155}Eu	0.61	0.06

[a] The standard deviations reported are the standard deviations of the eight sets of analytical measurements.

For this set of fuel pellets, the actinides and the direct fission products (^{90}Sr, ^{106}Ru, ^{137}Cs, and ^{144}Ce) are generally well predicted when the measured pellet burnup is matched. The exceptions are ^{129}I and ^{125}Sb. Iodine-129 is extremely long-lived and was thus present in very small concentrations in these low burnup samples. The reason for the poor agreement on ^{125}Sb is not understood. Those nuclides with strong radiative capture coupling (^{154}Eu, ^{155}Eu, and to a lesser extent ^{134}Cs) are less well predicted,

probably due to differences between the modeled and actual neutron spectrum, since the neutron spectral shape is more important in capture than in fission.

Through unpublished communications, we are aware that the version of ORIGEN (ORIGEN-R) in use at ANL-E for calculation of EBR-II subassembly inventories has been tested against experimental results, and "compared favorably." Additionally, when used to predict inventories and decay heat generation rates for standard PWR and LMFBR problems, the ANL-E values agreed within about 5% with those published by ORNL using ORIGEN2. Thus it appears reasonable to assume that ORIGEN-based inventory calculations for EBR-II can be computed with uncertainties on the order of those published for LWRs. However, the apparent lack of open, peer-reviewed literature to support this view is disturbing.

Proper inventory calculations require careful attention to core neutronics and exposure considerations. Assuming that care is taken, reasonable estimates of the uncertainty contribution of ORIGEN inventory calculations to the overall uncertainty in RH-TRU characterization, drawn from published data on PWR comparisons are as follows. Direct fission product nuclides (such as ^{137}Cs) can probably be predicted to within ±5%. Specific TRU nuclides are generally predicted to ±20%, with ^{239}Pu and total Pu somewhat better predicted at ±10%. Strongly radiative capture coupled nuclides (such as ^{154}Eu and ^{134}Cs) may be less well predicted. Uncertainties of ±30% appear reasonable.

Although there is little direct comparison data to evaluate the uncertainty in the prediction of activation product inventories (^{60}Co for example), they will probably be determined less accurately than the strongly capture-coupled nuclides. An uncertainty estimate for activation product inventory calculations of ±50% will be used in this scoping work.

UNCERTAINTY ESTIMATES

Direct Gamma-ray Spectroscopy and ORIGEN

From estimates of the expected standard deviation for determinations by DGS and expected uncertainty estimates for the ORIGEN inventory calculations, an uncertainty in the determined contents of the design basis drum can be estimated. It is assumed that an ORIGEN inventory calculation has been performed that matches the measured $^{137}Cs/^{60}Co$ ratios, and further matches, within the expected calculational uncertainties, the ratios of other detected fission products to ^{137}Cs. These adjusted calculations then predict the fission and activation product content per gram of ^{235}U (or other fissile nuclide depending on fuel type). The ORIGEN calculations then predict the concentration of all the fission products and transuranics per gram of ^{235}U. Additionally calculated is the specific activity of ^{60}Co

per gram of activated stainless steel (SS) cladding. The DGS measurements provide the total activity of ^{137}Cs and ^{60}Co in the waste drum. The total drum contents are then determined from the calculated amount of ^{235}U and irradiated SS in the assayed drum. The relevant equations are:

$$G_{U-235} = \frac{(A_{Cs-137})}{(SA_{Cs-137})} \quad \text{and} \quad \frac{\sigma_{G_{U-235}}}{G_{U-235}} = \sqrt{\left(\frac{\sigma_{A_{Cs-137}}}{A_{Cs-137}}\right)^2 + \left(\frac{\sigma_{SA_{Cs-137}}}{SA_{Cs-137}}\right)^2}$$

and $$G_{SS} = \frac{(A_{Co-60})}{(SA_{Co-60})} \quad \text{and} \quad \frac{\sigma_{G_{SS}}}{G_{SS}} = \sqrt{\left(\frac{\sigma_{A_{Co-60}}}{A_{Co-60}}\right)^2 + \left(\frac{\sigma_{SA_{Co-60}}}{SA_{Co-60}}\right)^2}$$

where: G_i = grams of i in assayed drum
A_j = DGS-measured activity of j in Curies
SA_j = ORIGEN-calculated specific activity of j per gram of i
and the associated uncertainties are self explanatory.

Substitution of the uncertainty estimates from the previous sections into the simple error prorogation equations suggests the drum inventories of ^{235}U and SS can be predicted to about ±22% and ±55% respectively. These values are for noncombustible matrix drums; however, comparable values for combustible matrix drums are similar (±21% and ±53% respectively). To estimate the uncertainty for the determination of other unmeasureable nuclides the estimated relative error in the ^{235}U content would be propagated with the estimated error in the ORIGEN-calculated specific activity. For example, the estimated uncertainty in the waste drum content of ^{63}Ni, an unmeasureable beta-emitting nuclide induced in activated SS, would be about ±75%.

PAN Active Assays and ORIGEN Calculations

To characterize fully the content of RH-TRU drums using the PAN system will require DGS results for the $^{60}Co/^{137}Cs$ activity ratios, and active PAN assay, and an ORIGEN calculation that matches the DGS-predicted ^{137}Cs and ^{60}Co activity ratios. PAN system active assays provide the total fissile content of a waste drum in grams. ORIGEN inventory calculations then provide the fissile material ratios and the estimated fission and activation product concentration ratios. The total amounts of activated SS in a waste drum will rely on the measured $^{60}Co/^{137}Cs$ ratio.

During testing of the PAN system active assay on Pu-containing drums the total fissile contents were determined to relative standard deviations of about ±1.5%.[b] However, the effect of the waste matrix on these results can be substantial. For well-behaved waste matrices total uncertainties of ±10% have been determined.[5] For more difficult matrices disagreements between passive and active assay result of nearly a factor of 10 were noted. It seems reasonable that over the limited set of drums in inventory, and

the relatively benign matrices, PAN active assays could be performed to estimated uncertainties of ±15%. Using standard error propagation techniques the estimated uncertainties (1σ) in the various drum contents using this approach of ORIGEN, PAN, and DGS would be about ±18% in the total Pu and ^{235}U contents, and ±53% in the total SS content.

SUMMARY

A variety of modeling techniques were employed to estimate the expected performance of two NDA approaches to the characterization of the INEL's RH-TRU wastes. These techniques are 1) direct assay of the drum fissile material content using a Passive/Active Neutron (PAN) assay unit, and 2) determination of the fission and activation product content of the waste drums [direct gamma-ray spectrometry (DGS)], coupled with calculation of the fissile material content using the known irradiation history and any of several available fission product inventory codes (e.g. ORIGEN).

Passive/Active Neutron (PAN) Assay

MCNP modeling techniques investigated the usefulness of the INEL's passive/active neutron (PAN) assay system for the RH-TRU waste characterization. Both active and passive assay modes were evaluated. In order to avoid gamma-ray pile up events from interfering with the PAN system neutron counts, a shield was designed to limit the surface dose rate of the samples to ≤ 50mR/hr. This required a lead overpack for the 113-liter RH-TRU drums with a wall thickness of about 4.5 cm. This size of overpack can be accommodated within the existing PAN cavity; however, the existing drum rotator may need to be replaced to accommodate the increased weight of drum and shield.

The PAN passive mode assays lack the sensitivity required for RH-TRU characterization. Active interrogation modeling predicts detection limits of 0.06 g ^{235}U and 0.04 g ^{239}Pu. These appear adequate for the vast majority of the INEL's RH-TRU drums. However, variations in waste matrix and loading in actual rather than modeled drums can have an important effect on these estimated detection limits.

Direct Gamma-ray Spectrometry (DGS)

MCNP modeling aided in the design of a heavily shielded and collimated gamma-ray spectrometer for direct assay of the fission and activation product content of RH-TRU drums. The collimator was sized to yield a detector counting rate of about 50,000 counts/second when a design-basis RH-TRU drum with a surface dose rate of 30 R/hr was assayed.

Detection limit estimates, developed from a synthetic spectrum generated to reflect the modeled measurement conditions, suggest that ^{60}Co, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{154}Eu, and ^{155}Eu should be detected reliably (95% CL) in RH-TRU drums. The analysis predicts that multiple gamma-ray lines from ^{154}Eu should be reliably detected. These lines could be used to determine drum-specific matrix attenuation corrections.

Even without the presence of sufficient gamma-ray lines to allow drum-specific attenuation corrections to be determined, attenuation corrections can be calculated. With no more information than the matrix type (combustible of noncombustible) and the gross drum weight (from which the average matrix density is estimated), attenuation corrections can be calculated with a relative standard deviations of about 10% to 20% in combustible matrix drums, and 20% to 25% in noncombustible matrix drums.

The DGS modeling further predicted relative standard deviation expectations for the absolute concentrations of ^{137}Cs and ^{60}Co, and for the ratio of the $^{60}Co/^{137}Cs$ activity ratio. In a combustible matrix drum these respective values are 20%, 18%, and 16%. Comparable values for a noncombustible matrix drum are 24%, 21%, and 16%.

ORIGEN Inventory Calculations

There are no published data comparing ORIGEN inventory estimates with measurements for EBR-II problems. Assuming the EBR-II case can be calculated at least as well as those for LWRs, it seems likely that burnup-matched ORIGEN calculations can predict total TRU-to-uranium ratios to about ±10%, direct fission product-to-uranium ratios to about ±5%, and the activation product content of stainless steels to about ±50%.

CONCLUSIONS

The analysis in this report estimates that the content of a design basis RH-TRU drum could be determined by a combination of DGS, PAN system active assays, and ORIGEN inventory calculations to estimated relative standard deviations of ±18 to 25% in the total Pu and total ^{235}U content, ±20 to 25% in the direct fission product activities, and ±55 to 75% in the activation product content. This work suggests that both Direct Gamma-ray Spectrometry and Passive/Active Neutron Assay when coupled with ORIGEN inventory calculations show promise for the characterization of the radionuclide content of the INEL's CC104/107 RH-TRU wastes.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the assistance, support and attention provided this project by C. R. Tyler. This work was supported by the U.S. Department of Energy, Assistant Secretary for Environmental Management under DOE Idaho Operations Office Contract DE-AC07-94ID13223.

REFERENCES

1. M. J. Bell, "ORIGEN -- The ORNL Isotope Generation and Depletion Code," *ORNL-4626* (May 1973).
2. J. F. Briesmeiser, "MCNP -- A General Purpose Monte Carlo Code for Neutron and Photon Transport, Version 3A," *LA-7396-M Rev. 2* (1986).
3. W. K. Hensley, A. D. McKinnon, H. S. Miley, M. E. Panisko, and R. M. Savard, "SYNTH: A Spectrum Synthesizer," *Proceedings of the 35th Annual Meeting of the Institute of Nuclear Materials Management*, Naples, FL (July 17-20, 1994).
4. H. D. Killian and P. D. Randolph, "Stored Waste Examination Pilot Plant (SWEPP) Nondestructive Examination/ Assay (NDE/NDA) Capability Study for RH-TRU Waste Stored at the RWMC," *INEL-95/255*, October 1995.
5. W. Y. Yoon, G. K. Becker, and Y. D. Harker, "Monte Carlo Neutron and Photon Modeling and Verification Calculations for the Stored Waste Examination Pilot Plant Passive/Active Neutron Assay System," *CONF-940216, Proceedings of the NonDestructive Assay and NonDestructive Examination Waste Characterization Conference*, Pocatello, Idaho, February 14-16, 1994.
6. L. A. Currie, "Limits for Qualitative Detection and Quantitative Determination," *Analytical Chemistry, 25*, #3, March 1963.
7. J. K. Hartwell, W. Y. Yoon, H. K. Peterson, and C. R. Tyler, "A Preliminary Evaluation of Certain Candidate NDE/NDA Techniques for RH-TRU Characterization," *INEL-96/0233*, in preparation.
8. D. Reilly, N. Ensslin, H. Smith, Jr., and S. Kreiner, "Passive Nondestructive Assay of Nuclear Materials," *NUREG/CR-5550*, March 1991.
9. W. Serrano, "Drum Matrix Mass Attenuation Coefficients," *INEL Engineering Design File RWMC-EDF-722*, March 14, 1994.
10. J. C. Ryman et al., "Fuel Inventory and Afterheat Power Studies of Uranium-fueled Pressurized Water Reactor fuel Assemblies Using the SAS-2 & ORIGEN-S Modules of SCALE with an ENDF/B-V Updated Cross Section Library," *NUREG/CR-2397*, September 1982.
11. B. G. Schnitzler and J. B. Briggs, "TMI-2 Isotopic Inventory Calculations," *EGG-PBS-6798*, August 1985.
12. D. W. Akers and B. G. Schnitzler, "Verification of the ORIGEN2 Code Analysis for the TMI-2 Reactor Core," *EGG-34487*, 1982.

DISCLAIMER