MIXED WASTE FOCUS AREA DEVELOPMENT OF TECHNOLOGIES TO CHARACTERIZE REMOTE-HANDLED TRANSURANIC WASTES

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ABSTRACT

The Mixed Waste Focus Area (MWFA) is funding the development of technologies to support the treatment and disposal of the Department of Energy's remote-handled transuranic wastes. Three areas of need were identified in support of this objective: 1) characterization of the radionuclide content to meet disposal facility waste acceptance criteria and transportation requirements, 2) determination of the gas generation potential of these wastes to meet shipping requirements, 3) and techniques to physically move the waste through certification and treatment processes to meet site ALARA concerns. The MWFA is currently supporting the development of three technologies to support the non-destructive assay of RH-TRU wastes and two projects to support the data collection and analysis of headspace gases to determine flammable gas generation rates. Work in the material handling area will be initiated in March, with an evaluation of site needs and current technical capabilities (both commercially and laboratory available). Development schedules and program summaries of each MWFA supported activity are given in this paper.

INTRODUCTION

Problem Definition

Approximately 4000 cubic meters of remote-handled transuranic (RH-TRU) waste is planned to be disposed at the Waste Isolation Pilot Plant (WIPP) between 2003 and 2033 (1). Four major sites (Idaho National Engineering and Environmental Laboratory, Hanford reservation, Los Alamos National Laboratory, Oak Ridge National Laboratory) and several small quantity sites will initiate the activities that are required to ready waste for shipment to WIPP within the next 3 – 10 years. As part of those activities, sites must characterize their RH-TRU wastes to verify compliance with transportation and disposal facility requirements. Three primary problems that sites face in their RH-TRU waste certification activities are 1) characterization of the waste's radioactive and hazardous components, 2) characterization of the waste's gas generation potential, and 3) movement of the wastes through the process steps (material handling). Furthermore, even the simplest of tasks are complicated by the high radiation fields.

Characterization

Sites currently characterize contact-handled (CH) wastes using a combination of non-destructive assay (NDA) techniques (e.g. gamma spectroscopy, passive and active neutron measurements), sampling/destructive analysis, and acceptable knowledge (AK). Unfortunately, the background radiation levels in RH-TRU wastes interfere with the detection of neutron and gamma ray signals, when using the same NDA techniques that are used for CH wastes. Sampling and destructive analysis of the RH-TRU waste streams is an alternative, but few analytical laboratories exist that can accept RH wastes. Besides the costs to take the samples and perform

the needed chemical analysis are high. Sites may also use documented information on a waste stream as acceptable knowledge, but for most waste streams additional information is required to meet the current transportation and disposal documentation requirements. Several waste streams at both the Los Alamos National Laboratory and the Oak Ridge National Laboratory were characterized several years ago using combinations of non-destructive techniques and sampling/analysis.

Gas Generation

The gas generation potential of wastes poses a problem during transport from the DOE storage or treatment facility to WIPP. Current transportation requirements limit flammable gases in TRU waste shipping packages to less than 5% for flammable gases and 500 ppm for volatile organics. The gas generation characteristics of CH waste were studied extensively and methodologies are in place to calculate and certify the gas generation potential for each CH waste container. These methodologies may or may not be applicable to RH-TRU wastes. Although the basic waste compositions are similar between CH and many of the RH wastes (job control materials, weapons grade plutonium distributions), the effects of additional gamma radiation on gas generation rates has not been determined.

Material Handling

Movement of wastes through the certification process is a concern for many of the sites, particularly the small quantity sites. RH-TRU wastes are currently packaged and stored in several configurations, for example, bagged wastes sitting in a hot cell, drummed wastes (1, 30, 55-gallon containers) in a storage facility or hot cell, canistered waste in a underground storage. In preparation for transport to WIPP, these wastes must be loaded in a waste canister and the canister loaded into the 72B shipping cask. Characterization may be completed on the small waste containers or on the waste canister, and depending on the technique utilized, through a shielded or unshielded waste package. Therefore, the appropriate equipment must be demonstrated to support the physical movement of the waste from its storage area through the packaging and characterization activities, while always keeping in mind ALARA (as low as reasonably achievable).

Project Prioritization

The three problem areas listed in Section 1.A. are documented in Site Technology Coordination Group (STCG) needs. Each Department of Energy (DOE) field office has established an STCG to work with waste storage and treatment facilities to identify, document, and prioritize their site technology development needs. The prioritized needs are transmitted to the MWFA. The MWFA then organizes similar needs into work areas. The MWFA work areas that address RH-TRU wastes include: Characterization, TRU Transportation, and Material Handling. The work areas and the activities within each work area are prioritized and funded accordingly. Final prioritization of the MWFA supported activities is based on several factors, including:

- End User Commitment – deployment commitment, visible End User Champion
- Likelihood of Success – status of development, time to complete the development activity, developer history
- Impact to Baseline Operations – site schedule, impact to baseline
- Impact to Needs – number and priority of needs

The prioritization is reviewed with the Carlsbad Area Office (WIPP operations) and representatives from each of the major TRU waste sites (MWFA End User Steering Committee and the TRU Waste Steering Committee). Much of the data to support high scorings in the End User Commitment and Impact to Baseline Operations criteria is generated from the DOE Carlsbad Area Office (CAO) and the Steering Committees. Within the MWFA project ranking, the RH-TRU related projects are prioritized in the top half of the list of MWFA supported activities. This is well above the funding line for the current and projected MWFA budget.

Transportation and Disposal Requirements

The transportation and disposal requirements for RH-TRU waste are currently draft. Technology development requirements for the characterization and gas generation activities were established from the draft RH-TRU Waste Acceptance Criteria, Waste Acceptance Plan, and the draft 72B Safety Analysis Report (2, 3, 4). A meeting is scheduled in January 2000 to discuss the draft RH-TRU Waste Acceptance Plan. A schedule for finalizing the WAP will be identified at that time. Below are listed the draft requirements that guide the MWFA funded development activities in the characterization and gas generation areas. Requirements for material handling techniques will be developed as technology's to support certification activities are finalized.

Characterization

Hazardous Components

AK, radiography, or waste inspection will be used to assign EPA hazardous waste numbers to RH-TRU waste streams. Those waste streams with inadequate AK will conservatively be assigned all spent solvent EPA hazardous waste numbers (F001, F002, F004, F005) and all other applicable codes (e.g. metals).

Radioactive Components

The draft WIPP waste acceptance criteria require that the following be met:

- Waste contains $\geq$100 nCi TRU isotopes per gram of waste
- Upper limits for thermal power and/or decay heat limit are 50 watts per canister for cellulosic wastes and 300 watts per canister for metallic wastes to maintain cask integrity
- Limits for thermal power and/or decay heat limit to ensure H_2 generation is <5% during transport. The limiting value for decay heat depends on the H_2 generation g-value for the waste type, and only applies if the radioassay-determined decay heat is chosen as the method for measuring the H_2 generation.
- ^{239}Pu FGE limit of <325 grams per canister
- PE-Ci limit of $\leq$1000 Ci per canister
- Activity $\leq$23 curies/liter averaged over the canister
- Canister surface dose rate $\leq$1000 rem/hr for all waste; $\leq$100 rem/hr for at least 95% of the RH waste; and neutron contribution to surface dose rate $\leq$270 mrem/hr
- Radioisotopic content for shipping manifests, PA, and Land Withdrawal Act RH curie limit.

Gas Generation

Data is needed on the gas generation potential of RH-TRU waste. The effects of high radiation backgrounds may destroy or reduce the organic components of the waste or increase the volatilization rate of the organic components. This would result in either much lower or higher VOC and semi-VOC concentrations in the canister headspace. The additional gamma radiation

may also enhance matrix depletion, which initially increases hydrogen gas generation rates in RH-TRU waste but yields lower gas generation rates for older wastes.

Although requirements for the 72B cask are not yet final, the CAO and MWFA have assumed that the Nuclear Regulatory Commission will impose the same requirements, hydrogen < 5% and VOCs < 500 ppm, that is imposed on the TRUPACT-II container.

Material Handling
Requirements for solutions that address this problem area will be generated in late FY-00 through FY-01. Solution development activities will be identified and selected to meet those requirements.

MWFA PROGRAMS

The MWFA initiated work in FY-98 to develop characterization technologies and evaluate the gas generation potential for RH-TRU wastes. Three technologies were identified as candidates for RH-TRU waste characterization: Gamma Spectrometry Combined with Acceptable Knowledge, Fast Neutron Coincidence Counter, and Multi Detector Analysis System. Two programs, one at Los Alamos National Laboratory and one at the Idaho National Engineering and Environmental Laboratory/Argonne National Laboratory, were funded to perform gas generation evaluations on actual RH-TRU wastes. The MWFA will initiate work in the RH-TRU material handling area in FY-01. Section 2.A summarizes the characterization projects and Section 2.B. summarizes the gas generation activities that the MWFA is currently supporting.

Characterization

Gamma Spectrometry Combined with Acceptable Knowledge (GSAK)
Technology Description
The GSAK concept uses conventional NDA gamma spectrometry instrumentation combined with knowledge of the waste contents to characterize an RH waste drum (5). The gamma-ray spectrometer consists of a high purity Germanium detector, a detector shield and cart, and a self-contained electronics and multichannel analyzer system. The spectroscopy system is called *ISOCS* by the manufacturer (Canberra). RH waste streams that the GSAK technique can accommodate primarily consist of heterogeneous hot cell debris that is contaminated with test residue from destructive fuel examinations. The chemical and radiological properties of the fuels that produced the radioactive test residues were documented as part of the fuel examination program. This information can be used as the AK for the GSAK concept. Approximately 90% of the Idaho National Engineering and Environmental Laboratory's (INEEL) remote-handled waste inventory consists of this heterogeneous hot cell debris and is a candidate for characterization using this technique.

The GSAK measurement consists of acquiring a gamma-ray spectrum of a rotating shielded RH-TRU waste drum. The gamma-ray spectrum is analyzed to determine the activity of certain fission and activation product nuclides within the waste debris. Facility records from the fuel examinations were used to provide the input required for content calculations using a buildup and depletion code (ORIGEN or REBUS). Fission product isotope ratios measured by the gamma-ray spectrometer confirmed the burnup and/or last date of irradiation given in the facility

records. The total drum contents are then determined by applying the calculated isotopic ratios from the inventory calculation along with the total measured activity of certain key nuclides from the gamma-ray spectrometry results.

The objective of the MWFA funded program was to test the GSAK concept on a well-known surrogate matrix and thereby demonstrating the concept. This program was an opportunity for the MWFA to support a relatively inexpensive demonstration using modified commercially available technology within a 1 – 2 year time frame. Once demonstrated, the GSAK hardware will be moved to support real waste measurements of INEEL RH wastes.

Test Results

The GSAK System incorporates an *ISOCS*-characterized detector and can therefore, be calibrated by calculation of the detector response, using one of a set of templates that specify the source and detector geometry. The ability of the ISOCS software to correctly determine the detector efficiency in the heavily shielded GSAK geometry was tested using a standard prepared from a calibrated ^{152}Eu stock solution. The calibrated solution was diluted and added to a plastic-lined 7 gallon RH-TRU waste "pail." The standard-containing 7 gallon pail was placed within a 30-gallon drum positioned within a prototype steel overpack. The prototype overpack design was later used to support the surrogate and real waste measurements. The ISOCS-calculated and the measured efficiency curves agreed within ±10% from 300 keV to 1000 keV and within ±30% above 1000 keV.

After the calibration check, the system was moved to the INEEL's TREAT facility for the surrogate waste tests. The waste surrogate assemblies incorporate matrix materials and source positioning ports analogous to those used for CH-TRU wastes. The sources used were 0.63-cm segments of a spent EBRII driver element for which segment-specific nuclide inventories had been calculated. The three surrogates – C (combustibles), NC (metals), and Z (metal) – were loaded with 8, 6, and 3 encapsulated rod segments respectively. Surrogate C and NC were loaded with a symmetric source pattern while Z was loaded asymmetrically. Five additional segments from the same element were submitted for radiochemical analysis to confirm the calculated inventory results. The surrogate assemblies were loaded with the source segments in an ANL-W hot cell and moved to the TREAT reactor building, where the loaded assemblies were lowered into shielded overpacks previously configured with 30-gallon drums and spacer materials.

Each surrogate-containing overpack was positioned on the GSAK rotary table and a number of spectra acquired on each rotating assembly. Measurements were performed both at 1-meter and at 2-meter detector to overpack distances. Replicate measurements were performed at each distance and the detector and shield arrangement was moved between each measurement in order to incorporate any positioning error into the measurement reproducibility. On each surrogate assembly, at least one long (usually overnight) count was acquired to optimize detection and precision. The isotopes detected and quantified were ^{54}Mn, ^{60}Co, ^{134}Cs, ^{125}Sb, ^{137}Cs, ^{144}Ce(^{144}Pr), and ^{154}Eu. These measurements were completed in September 1999.

The radiochemical analysis of three of the five fuel segments was completed at the time of this report. The comparison of the radiochemical analysis results and the calculated inventories for the GSAK-detected nuclides in a representative fuel segment is presented in Table I. The reason for the inconsistent calculational results in Table I is under review.

Table I. Calculated and radiochemically measured content of fuel segment SADY33.

Isotope	Calculated Content	Measured Content		Measured/Calc Ratio
	(milliCi)	(milliCi)	2σ Error	
Mn-54	0.13	0.16	33%	1.26
Co-60	0.85	0.21	0.19	0.25
Sb-125	4.82	3.9	0.19	0.81
Cs-134	3.48	1.5	0.15	0.43
Cs-137	307.66	130	0.15	0.42
Eu-154	0.96	0.91	0.19	0.94

The preliminary GSAK measurement results (corrected for gamma-ray attenuation in the fuel segments, the fuel segment cans, and the surrogate matrix) are compared in Table II with the expected activity for each surrogate. The expected activity for each nuclide was the calculated segment inventory multiplied by the average measured-to-calculated ratio for that nuclide from the radiochemically analyzed segments.

Table II. Measured and expected values for the GSAK surrogate measurements.

Surrogate	Isotope	GSAK Measured		Expected Inventory		Measured/Expected Ratio	
		Ave Curies	% Error (2σ)	Curies	% Error (2σ)	Ratio as %	± Error (2σ)
C	Mn-54	7.96E-04	26.4%	1.33E-03	27.6%	60.0%	22.9%
	Co-60	7.32E-04	21.7%	1.57E-03	20.7%	46.6%	14.0%
	Sb-125	1.75E-02	22.1%	3.18E-02	22.4%	54.8%	17.2%
	Cs-134	1.18E-02	22.1%	1.36E-02	21.9%	87.3%	27.2%
	Cs-137	9.20E-01	22.6%	1.09E+00	20.7%	84.7%	26.0%
	CePr-144	2.23E-02	22.1%	4.90E-02	23.4%	45.5%	14.6%
	Eu-154	2.20E-03	23.2%	4.00E-03	24.3%	54.9%	18.5%
NC	Isotope	GSAK Measured		Expected Inventory		Measured/Expected Ratio	
		Ave Curies	% Error (2σ)	Curies	% Error (2σ)	Ratio as %	± Error (2σ)
	Mn-54	6.27E-04	25.0%	9.77E-04	27.6%	64.2%	23.9%
	Co-60	6.14E-04	21.6%	1.18E-03	20.7%	52.1%	15.6%
	Sb-125	1.34E-02	23.0%	2.39E-02	22.4%	56.2%	18.0%
	Cs-134	8.13E-03	22.2%	1.03E-02	21.9%	78.8%	24.6%
	Cs-137	6.89E-01	24.9%	8.14E-01	20.7%	84.6%	27.4%
	CePr-144	1.72E-02	22.1%	3.67E-02	23.4%	46.7%	15.0%
	Eu-154	1.72E-03	22.9%	3.05E-03	24.3%	56.3%	18.8%
Z	Isotope	GSAK Measured		Expected Inventory		Measured/Expected Ratio	
		Ave Curies	% Error (2σ)	Curies	% Error (2σ)	Ratio as %	± Error (2σ)
	Mn-54	3.05E-04	29.9%	5.00E-04	27.6%	61.1%	24.9%
	Co-60	2.95E-04	22.4%	5.94E-04	20.7%	49.7%	15.2%
	Sb-125	7.09E-03	24.3%	1.20E-02	22.4%	58.9%	19.4%
	Cs-134	4.07E-03	22.2%	5.19E-03	21.9%	78.4%	24.5%
	Cs-137	3.70E-01	35.5%	4.11E-01	20.7%	90.0%	37.0%
	CePr-144	8.79E-03	27.9%	1.85E-02	23.4%	47.5%	17.3%
	Eu-154	7.79E-04	24.1%	1.53E-03	24.3%	50.9%	17.4%

Based on initial review of the data, the system is scheduled to move to the RWMC in FY-01 to perform measurements on actual RH waste that is scheduled for retrieval and storage in overpacks. Total MWFA cost to complete the surrogate demonstration was $484 K. (Note additional funds on the order of $ K was provided by the INEEL.) The RH-TRU surrogates developed through this program are available for use in other system development programs. The system is now available to support real waste measurements at the INEEL's RWMC.

High Speed Neutron Detector Evaluation

Technology Description

A fast neutron counting concept is being evaluated to characterize RH-TRU and high activity CH-TRU wastes. Initial work focused on evaluating several developmental high-speed neutron detectors. One detector, a boron-loaded scintillator (BC454) coupled to a bismuth germanate (BGO) inorganic scintillator (phoswich detector), was selected for surrogate and real waste testing in FY-99. Neutrons enter the phoswich detector, scatter in the BC454 plastic, and are captured on the boron. Neutron capture by the boron-10 results in a 2 Mev release of kinetic energy and 2 charged particles, helium-4(α-particle) and lithium-7. The charged particles deposit their energy in the BC454 scintillator, which results in a measurable energy equivalent to 93 keV. The lithium-7also decays via a 478 keV gamma ray, which may be detected by the BGO. A neutron is identified as an event in which signals from both the BC454 and the BGO detector elements meet the respective energy requirements. For each detected event, an energy is recorded for both the BC454 and BGO along with the event time. Filtering of the event stream is accomplished by selecting only those events that occur within a given energy range (corresponding to a neutron capture) and subsequently analyzing the event times using time-correlation techniques appropriate for fission sources. Results of the analysis yield the plutonium-240 equivalent mass in the sample. High resolution gamma-ray spectroscopy, mass spectroscopy, or other acceptable facility knowledge is required to convert the neutron measurement to total plutonium content.

To support the experimental portion of this work, ten detectors were incorporated into an existing FNCC. The sample cavity for this well counter was 20-cm in diameter by 25-cm high. Graphite between the detectors and in the endcaps increased neutron capture efficiency without increasing the mean lifetime of the neutrons in the systems. The well was big enough to support the measurement of the sources and the cans/pails of CH and RH-TRU waste used in the experiments.

The results from the surrogate and real waste measurements were used to benchmark Figure-of-Merit (FOM) calculations of the expected performance of a fast neutron assay system for RH-TRU wastes. By varying parameters in the FOM calculations to match other detector characteristics, the performance of additional neutron counter designs can be predicted. Two other high speed neutron detectors were evaluated using the FOM code; high pressure helium-3 tubes and BC501 liquid scintillators.

Test Results

Work in FY-99 focused on fabrication of the experimental FNCC prototype and testing the current hardware and software configuration (6). A primary area of need within the DOE is the assay of very high-alpha wastes with moderate gamma doses of 1-2 R/hr outside of the shielded

or unshielded container. The existing prototype FNCC was tested to see if fast neutron counting can meet this need by assaying Molten Salt Extraction (MSE) salts and other high-alpha scrap and waste at the Los Alamos Plutonium Facility. The key measurement parameters were neutron detection efficiency, average neutron lifetime (die-away time), gamma-ray leakage, coincidence leakage, and background count rates. These parameters made it possible to refine the assay variance estimators in the FOM code and the detection sensitivity calculations based on Excel spreadsheets.

Through November 1999, approximately 200 measurements were taken at the Los Alamos Plutonium Facility. Background and californium source measurements were made every day that the counter was used for plutonium measurements, to verify detection efficiency, stability, and energy regions of interest for the plutonium data. The materials measured during the campaign included plutonium oxide standards, impure plutonium oxide, impure plutonium salts, impure molten salt extraction salts and plutonium tetrafluoride. The α values ranged from 1 to 130, with the lower values being typical of CH-TRU waste, and the higher values being typical of RH-TRU waste. These materials were each measured multiple times to provide a rough estimate of assay uncertainty for comparison with the FOM code calculations. Table III summarizes the wastes and number of measurements for the FNCC experiments.

Table III. Summary of FNCC measurements

Item ID	Material Type	Sample Mass (g Pu)	Predicted α	Actual α	No. of Measurements	240 [a] Pueff. (g)	240 Pu [b] assay (g)
A1-91	PuF4 powder	10	130		1	0.56	
A1-86	Pu oxide std.	10	0.90		1	0.56	0.77
LAO250c10	Pu oxide std.	60	0.40	2.0	1	10.1	10.4
LAO251c10	Pu oxide std.	172	0.41	1.4	1	29.3	29.1
LAO252c10	Pu oxide std.	322	0.40	0.8	1	54.4	53.3
LAO256c10	Pu oxide std.	385	0.40	0.8	1	65.2	62.9
Calex-2	Pu oxide std.	398	0.90	1.6	5	23.6	25.4
STD-40	Pu oxide std.	874	0.75	1.3	3	107.4	103.4
MSSTD-1	Pu oxide std.	2436	0.92	1.2	33	156	118.5
Sp9802	Impure Pu oxide	648	1 - 2	2.2	1	38.8	38.6
XBLSCL25	Impure Pu salt	166	6.0	21	6	10	31.3
XBLSCL28	Impure MSE salt	167	9.0	14	5	10	40.8
XBS9413	Impure Pu salt	273	9.5	19	7	16.4	71.5
GVB17C1	Impure dioxide	314	29.4	26	19	24.6	154.8
ARF876595	Impure MSE salt	263	34.2	32	29	16.5	77.3
PUF4-1	PuF4 powder	281	110	92	52	16.9	75.4

[a]Based primarily on calorimetry values
[b]Experimental FNCC measurement

The data in Table III shows that a high bias is seen when the ratio of the assay value (column 8) to the reference value (column 7) is taken. The bias increases with an increase in α ratios. These test data provided the baseline performance data needed to evaluate use of an FNCC, an Epithermal Neutron Counter (ENC), and a Liquid Scintillator Counter (LSC). The ENC design

uses 10-atm helium-3 tubes and the LSC design BC501 liquid scintillator cells. The helium-3 and scintillator detectors were both developed and tested under separate programs.

A summary of the system design, and performance of the three fast neutron detector options is given in Table IV. The performance is evaluated in terms of two requirements for RH-TRU characterization: the measurement uncertainty for assay of a plutonium sample with total Pu mass close to the Fissile Gram Equivalent limit of 325 grams, and the Minimum Detection Limit. The relative cost and schedule to design and fabricate a prototype system are also included in Table IV.

Table IV. Summary of design features and performance for three fast neutron counter options for RH-TRU waste drums.

Detector System	BC454/NaI Scintillators	Epithermal Neutron Counter	Liquid Scintillator Counter
Detector type	Decoupled BC454/NaI	10-atm 3He tubes	BC501 cells
Number of detectors	80	324	48
Total neutron efficiency	13%	54%	25%
Neutron die-away (ms)	4 μs	20 μs	100 ns
Predelay (μs)	1 μs	1.5 μs	0
Gate Width (μs)	6 μs	24 μs	100 ns
Background cps	1000	500	500
Precision at FGE Limit	11 - 32%	2 - 6%	0.4 - 1.0%
MDL for RH-TRU	0.26 - 10.3 g 240-Pu	0.005 - 0.20 g 240-Pu	0.008 - 0.31 g 240-Pu
Operational Complexity	High	Low	Moderate to High
Design & Fabrication Time Estimate	24 months	18 months	24 months
Cost of System	Most Expensive	Least Expensive	

Total MWFA cost to complete the experimental FNCC measurements and FOM calculations was $545 K. The experimental data was generated to support the design of an optimized neutron counter and the performance expected of the system. The path forward for this program is the topic at a February 2000 meeting. Options include fabrication of an optimized prototype (funding source TBD), additional evaluation of several recently developed detectors for application to RH-TRU, or placing the program on hold until the MDAS concept is demonstrated and a decision to go forward is made.

Multi Detector Analysis System (MDAS)

Technology Description

The MDAS concept incorporates a new understanding of the physics of radiation sources. This is based on the correlation of the prompt radiations (gamma ray and neutrons) that occur in the fission process and in addition the multiple gamma rays that are produced by the prompt de-excitation of the isotopes produced in fission. Until the mid- to late-1980s, fission fragments

were never studied directly because of instrumental limitations. Experiments conducted at ANL's Intense Neutron Pulse Source showed that immediately following fission, the resulting daughters produced by the fission process emit characteristic gamma emissions (7). This gamma emission can be used by a coincidence system to identify the fissioning element. Based on knowledge of the fission process, the daughter element concentrations can be used to determine the element responsible for the fission and the amount of the element present in the material. The fact that the gamma emission occurs immediately following fission can be used to discriminate between the gamma emission occurring normally from the radioactivity of the material.

MDAS uses a pulse of neutrons generated by a neutron generator similar to those currently used in differential die away methods. By gating the gamma detectors with the neutron generator and monitoring only for approximately 100 nanoseconds following the neutron pulse, the contribution to the measured gamma spectrum by the natural gamma emission associated with radioactive waste is almost nonexistent. MDAS is expected to be able to measure waste without prior significant knowledge of the waste matrix and history.

The experimental MDAS system incorporates state-of-the-art electronics and specially designed fast detectors. The prototype MDAS has 68 detectors; 20 high-purity germanium detectors and 48 liquid scintillator detectors. The system also uses fast coincidence methods, list-mode data collection and storage, gamma-ray coincidence, neutron coincidence, pulse-shape discrimination. Funding in FY-99 from the MWFA and the Spent Fuel Program went to support the refurbishment and installation of a radiofrequency quadrupole (RFQ) electron accelerator neutron generator. Present NDA systems use commercially available sealed-tube pulsing neutron generators that provide a neutron flux up to 10E8 n/s/4pi. This flux was inadequate for the assay of RH-TRU waste having neutron emission rates ranging from 10E6 to 10E8 n/s.

Test Results

Installation of the neutron generator was initiated in 1st Quarter FY-00. Safety reviews and testing of the neutron generator are scheduled for 2nd Quarter with integrated testing beginning in 3rd Quarter. A demonstration of the technology on one surrogate RH-TRU waste matrix will be completed in 4th Quarter FY-00 to benchmark the system's capabilities. If successful, software development and testing will continue through FY-01. It is expected that this technology will be transferred to the commercial sector and a prototype built for final demonstration and deployment. A commercialization strategy is being developed and a commercial partner selected in FY-00.

Total MWFA cost to support this program to date was $660 K. $760 K is required in FY-00 to support the surrogate assay measurement, data analysis, and algorithm development. Future funding for this program will be decided once initial demonstration and data analysis is complete.

Gas Generation Testing

Gas Sampling of RH-TRU Waste Containers - LANL

Technology Description

LANL presently has seventeen shafts that contain characterized RH-TRU waste that is packaged (to WIPP WAC Revision 3) for eventual shipment to WIPP (8). Nine of the seventeen canisters exceed the allowable decay heat limits for the 72-B Cask shipping package. Another option to demonstrate compliance with the flammable gas requirements is to establish the hydrogen gas generation rates for these canisters. LANL selected ten waste canisters, including the nine that exceed the decay heat limit, for sampling and analysis of the canister headspace gases. The data is needed to evaluate the potential for flammable gas generation and to evaluate compliance with the transportation requirements specified in the draft 72-B SARP.

Test Results

A shielded sampling apparatus was fabricated and installed over the shaft openings. Headspace gas samples from the 10 selected waste canisters were pulled at least 5 times over a period of 11 weeks at TA-54, between June and August 1999. The results of the sampling performed through August, with a detailed analysis of gas generation measurements for four of the RH canisters, are documented in Table V. Hydrogen was detected in all ten of the sampled canisters with concentrations ranging from 0.10 volume percent to as high as 1.7 volume percent. Oxygen concentrations are below those in air and indicate that oxygen is being consumed, which is expected to occur through radiolysis of the hydrogenous materials present in the waste and recombination with hydrogen in a radioactive environment. There is good agreement between predicted hydrogen gas generation rates derived from a canister diffusivity characteristic model and a dose-dependent G-value model. However, there is insufficient data at this time to establish a definitive hydrogen gas generation rate for each of the nine canisters that exceed the decay heat limit. Canisters will continue to be sampled and the headspace gases analyzed throughout FY-00.

Table V. Gas generation calculations for four RH-TRU canisters.

Canister	Decay Heat (W)	Average Apparent Hydrogen Generation Rate Based On Gas Flow Rate (mol/s)	Average Apparent Hydrogen Generation Rate Based on Canister Diffusivity Characteristic (mol/s)	Hydrogen Gas Generation Rate Based on G Value of 3.4 (mol/s)	Hydrogen Gas Generation Rate Based on G Value of 1.09 (mol/s) (MDP Results)	% Hydrogenous Materials	Hydrogen Gas Generation Rate Based on G Value of 3.4 x % Hydrogenous (mol/s)	Hydrogen Gas Generation Rate Based on G Value of 1.09 x % Hydrogenous (mol/s) (MDP Results)
LA07	5.546	2.4E-08	7.3E-07	2.0E-06	6.3E-07	2.7	5.3E-08	1.7E-08
LA08	5.559	2.1E-08	4.9E-07	2.0E-06	6.3E-07	15.9	3.1E-07	1.0E-07
LA10	4.806	2.2E-08	1.0E-06	1.7E-06	5.4E-07	39.8	6.7E-07	2.2E-07
LA15	3.295	1.7E-08	4.2E-07	1.2E-06	3.7E-07	50.0	5.8E-07	1.9E-07

FY-00 testing is composed of the following tasks:

- Sample and analyze for all gases and vapors that may be present in each canister including methane, carbon dioxide, carbon monoxide, VOCs, relative humidity or moisture content, and argon
- Develop a methodology for an independent validation of the measured hydrogen gas generation rate. This may include for example an analysis of the ratio of the concentration of

an inert gas such as nitrogen, neon, or argon to that of the hydrogen concentration or the placement of a calibrated hydrogen leak source inside a newly packaged canister

- Prepare a contingency plan and feasibility study for eventual RH canister repackaging if the measured hydrogen gas generation rates are found to exceed the limits.

Total MWFA cost to support this program to date was $635 K. $250 K is required in FY-00 to complete the planned work scope.

Headspace Sampling of RH-TRU Waste Containers – INEEL/ANL

Technology Description

The purpose of this task was to identify initial hydrogen gas generation data for newly packaged RH-TRU waste (9). Two 30-gallon waste drums were prepared in the ANL-E Alpha-Gamma Hot Cell Facility for the gas generation tests using RH-TRU waste. One waste drum was filled with combustibles (neoprene rubber, polyethylene, and rags) and some metals. The other waste drum was filled with primarily noncombustible waste consisting of metals (iron, tin, aluminum) and glass. The drum configuration for each nominal 30-gallon waste drum was identical to that used in the packaging of RH-TRU wastes stored at the INEEL.

Test Results

Gas sampling was initiated 40 days after drum closure. Three more sampling and analysis campaigns over an 8-month period were completed on each of the test drums. The samples were analyzed for hydrogen, oxygen, nitrogen, and argon, with carbon monoxide, carbon dioxide, and methane included in the last two samples. Table VI lists the data collected on the two drums.

Table VI. Gas volume percent measurements on the RH-TRU waste drums.

Combustible Waste							
Sample Day	H_2	O_2	CO_2	CO	CH_4	Moles H_2in drum (gmole)	H_2 rate (mol/s)
41	0.065	7.37	-	-	-	0.00279	7.88E-10
71	0.076	7.81	-	-	-	0.00326	1.81E-10
153	0.194	6.93	0.150	0.184	0.012	0.00833	7.16E-10
197	0.227	6.90	0.230	0.268	0.014	0.00974	3.71E-10
Noncombustible Waste							
41	0.067	9.17	-	-	-	0.00322	9.09E-10
71	0.093	8.33	-	-	-	0.00399	2.97E-10
153	0.156	10.3	<0.024	0.006	<0.004	0.00670	3.83E-10
197	0.174	9.90	<0.024	0.035	<0.004	0.00748	2.05E-10

Total MWFA cost to support this program to date was $160 K. Additional data to support the determination of "G" values is required. The work scope to accomplish the calculations is currently being defined. Funds are available in FY-2000 to complete this work.

OUT-YEAR ACTIVITIES

The MWFA will continue working with CAO and the sites to identify and fund programs that support the disposal of RH-TRU wastes at WIPP. Funds were requested in FY-00 and FY-01 to support demonstration activities associated with currently funded projects. These projects include deployment of the GSAK technology, potential fabrication/demonstration/deployment of a mobile/portable fast neutron coincidence counter, demonstration/technology transfer of the MDAS technology, and evaluation of the gas generation potential of RH-TRU wastes.

Funds in FY-01 were also identified to address the characterization of small quantity problematic wastes, the sizing/sorting of large equipment into TRU/non-TRU components, and technologies to support the repackaging of RH-TRU wastes. These projects may fall into the basic science, applied research, or development/ demonstration phases of development. Potential solutions will be identified in FY-00 and initiated in FY-01. Depending on the maturity of the selected project(s), systems will be ready for implementation in the FY-02 through FY-05 timeframe.

REFERENCES

1. The National TRU Waste Management Plan, DOE Carlsbad Area Office, DOE/NTP-97-1204, Revision 1 (1997).
2. A. M. MEEKS, J. A. CHAPMAN, “Development of the Remote-Handled Transuranic Waste Radioassay Data Quality Objectives”, ORNL/TM-13362 (1997).
3. Draft Issue Paper, “Proposed Quality Assurance Objectives (QAOs) and Data Quality Objectives (DQOs) for Characterization of Remote-Handled (RH) Transuranic (TRU) Waste Going to the Waste Isolation Pilot Plant (WIPP) (1999).
4. Benchmark Environmental Corporation, “DRAFT Annotated Outline for the WIPP Remote Handled Transuranic Waste Analysis Plan”, January 2000.
5. J. K. HARTWELL, M. E. MCILWAIN, ”Quick Look Report for FY1999 Gamma-Ray Spectroscopy Combined With Acceptable Knowledge (GSAK) Development and Evaluation (ID-7-9-MW-13B)” (1999).
6. N. ENSSLIN, D. MAYO, M. BROWNE, L. CARRILLO, October PTS Report (1999).
7. J.D. COLE, M.W. DRIGERT, R. ARYAEINEJAD, E.L. REBER, J.K. JEWELL, “Multi-Detector Analysis System for Spent Nuclear Fuel Characterization, 1999.
8. L. R. FIELD, R. VILLARREAL, “Headspace Gas Sampling of Remote-Handled Transuranic Waste Canisters at Los Alamos National Laboratory, Fiscal Year 1999 Year-End Status Report (1999).
9. K. J. LIEKHUS, “Gas Generation in Remote-Handled Transuranic (RH-TRU) Waste Drums”, INEEL/EXT-98-01282 (1999).