NON-DESTRUCTIVE ASSAY OF TRANSURANIC (TRU) RAD-WASTE SLUDGES USING THE IMAGING PASSIVE ACTIVE NEUTRON (IPAN) TECHNIQUE

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ABSTRACT

A new technique has been developed for assay of transuranic radioactive sludge waste in 208 liter drums. The method uses the high sensitivity of the Imaging Passive Active Neutron (IPAN) system to provide regulatory acceptable measurements on sludges. The technique has been field tested on IPAN drum counters at Rocky Flats and Hanford. In one application, assays are performed on Transuranic (TRU) and Low Level Waste (LLW) sludge waste that originated from an aqueous precipitation process. In a second application, the waste came from spent nuclear fuel (SNF) storage and will therefore be contaminated with fuel corrosion by-products including TRU isotopes and fission/activation products. Prior to assay, the SNF sludge is grouted with a cement mixture. A new calibration technique was developed specifically for these two challenging waste streams that uses a radial weighted average method to define the imaging response matrix. This method provides the required sensitivity to the height distribution of special nuclear material within the 208 liter drum, and makes use of the uniform radial distribution that will occur for a distribution of a large population of small particles in a homogeneous matrix. The active and passive modes of the IPAN provides a wide dynamic range of assay: from below the TRU/LLW sorting threshold of 100 nCi/g (3700 Bq/g) up to several hundred grams of Weapons Grade Pu Equivalent. For mixtures of uranium and plutonium, isotopic categorization can be performed by analysis of the ratio of the active and passive effective masses. Extensive validation and testing with specially designed surrogate sludge drums and radioactive standards have resulted in regulatory acceptance of this technique, permitting ultimate disposal of the sludge drums at the Waste Isolation Pilot Plant.

INTRODUCTION

The assay of homogeneous waste forms such as sludges, soils and concrete presents a unique challenge for radiometric instrumentation. These waste forms tend to have a relatively high density and often a high hydrogen content causing significant attenuation of characteristic radiation such as neutrons and gamma rays. A new method has been developed using a combined passive neutron and active neutron assay for reliable characterization of homogeneous waste in drums and larger containers. The key development is the application of imaging algorithms to reduce the impact of positional effects of source material within the waste matrix. An imaging passive active neutron (IPAN) system has been used to demonstrate the efficacy of the technique with sludge waste at Rocky Flats Environmental Technology Site (RFETS). The technique will also be applied to characterize SNF contaminated sludge at Hanford.

SLUDGE PROPERTIES

Globally, a large volume of sludge rad-waste has been produced as a by-product of the nuclear fuel cycle and weapons production programs including:

- residues from chemical processing for recovery of plutonium or other TRU elements,
- corrosion of spent nuclear fuel stored for long periods underwater,
- waste treatment processes such as aqueous precipitation.

After the sludge has been retrieved, packaged and, where necessary, stabilized (e.g. by addition of grout), radioassay is usually required to characterize the radioactive contents prior to intermediate storage or ultimate disposal.

Most sludge waste forms will comprise an inorganic precipitate suspended in aqueous solvent. Often, the sludge will be treated to remove large particles of transuranic debris, resulting in a distribution of radioactive material is the form of fine particulates. The actual distribution of source material will depend on the type and consistency of the sludge, but in most cases a uniform radial distribution can be expected, with the axial distribution dependent on whether sufficient time has elapsed for settling of particulates to occur.

Table I provides two examples of sludge waste form properties from the US Department of Energy (DOE) operations at Hanford and RFETS. In each case, the final composition reflects a mixture of cement and sludge required to stabilize the final waste form in 208 liter drums. These examples illustrate a wide diversity in the quantity of cement used. For the Hanford case, only a small quantity of sludge is mixed with the grout whereas, at RFETS, only a small quantity of grout was required.

Table I. Example of US DOE sludge physical properties .

	Hanford Sludge	**RFETS Sludge**
Production Process	Corrosion of spent nuclear fuel	Aqueous precipitation
Final waste form density (g/cm^3)	1.9	1.3
Dry Sludge wt%	10%	18%
Dry Cement wt%	61%	12%
Water wt%	29%	70%

Sludge surrogate drums have been constructed using a simulated sludge generation process with inactive materials (i.e. no radioactivity added). The process of preparing the sludge/cement mixture and curing the final assembly was carefully controlled in order to produce a surrogate with physical properties as close as possible to actual DOE sludge waste. Insert tubes (diameter of 5.1cm) were built into these drums are various radii, in order to allow radioactive standards to be located within the drum for the purpose of calibration and testing. Small tubes known as "spacers" containing the grout/cement mixture are provided with the drum to back-fill the insert tubes.

ACTIVE / PASSIVE NEUTRON IMAGING METHOD

The IPAN system hardware (Fig. 1.) has been specifically designed to enable imaging of source material within waste containers. Imaging produces a significant improvement in accuracy over traditional techniques by reducing the sensitivity to variation in source position between actual waste containers and the calibration configuration. The IPAN's data acquisition system records the active and passive signal for localized banks of He-3 detectors which are arranged in the walls, floor and roof of the assay chamber. A pulsed Deuterium – Tritium (D-T) neutron generator is used to generate 14 MeV neutrons for the active mode assay. The generator is located externally to the assay chamber within a moderating assembly comprised of lead, graphite and polythene. This design is intended to provide a planar interrogating neutron flux, which together with the carefully selected position and orientation of the detector banks provides optimum imaging performance.

Fig. 1. IPAN™ system

IPAN calibration proceeds by placement of radioactive standards at various height and radii within one or more surrogate waste containers. For drum assay, the standard IPAN measurement consists of 16 separate angular measurements for both active and passive portions. Data for each of the 16 "grabs" is summed by the data analysis software. The effect of the drum rotation is to effectively simulate an annulus source volume element (known as a "voxel") for each source/tube placement. The imaging algorithm performs a matrix reconstruction analysis that compares observed signal data with processed calibration response sets. This step determines the most probable fissile (and passive source) signal distributions within the drum that are also compatible with the indicated matrix material properties. This image reconstruction algorithm is an iterative "least-squares" fitting process.

The imaging technique works well with light to medium density heterogeneous debris waste. The algorithm makes use of the fact that a source at any given voxel within the drum will produce a unique response vector in the detector banks. However, for highly interfering matrices, imaging performance suffers from the fact that the sensitivity to sources near the centre of the drum may differ by more than an order of magnitude compared to the outside of the drum.

For matrices that are both homogeneous and highly interfering we can take advantage of the fact that the source distribution will be radially homogeneous. The calibration libraries are pre-corrected by performing a volume average across all radii and thus the imaging process is effectively limited to the axial dimension.

COMBINING IPAN DATA WITH GAMMA SPECTROSCOPY

The combined active/passive technique provides a highly sensitive method for assay of uranium and plutonium in sludges. The active method provides a direct measure of the fissile content (including Pu-239, U-233 and U-235) and the passive method is sensitive to spontaneous fission emitters (such as Pu-240, Pu-238, Cm-244). For waste streams where the isotopic mixture is well characterized (e.g. through an acceptable knowledge program), the active and passive response may be converted directly to parameters of interest such as radionuclide inventory, total Pu, total alpha activity, total Watts etc.

For waste streams where a wide diversity of isotopics can be encountered, or where no prior knowledge is available, gamma spectroscopy can often be used to delineate the isotopics and thus provide an acceptable degree of accuracy in the measurement without the need to make gross assumptions.

For sludges contaminated with fission / activation products, the gamma spectroscopy results may have a more limited role to play. This is because Compton scattering of the high energy photons typically associated with these nuclides (e.g 662 keV from Cs-137) will often wash out the low energy photopeaks associated with uranium and plutonium nuclides. However, the combined assay results can still be used to extract important information on the radiological characteristics of the waste. For example the detection of key nuclides by gamma spectroscopy may provide a means of categorizing a particular waste container against a population of known waste streams, which is particularly useful for historic waste where containers from different streams may have been mixed together during storage.

The multiple technique approach also offers the capability of verifying acceptable knowledge. This can be done by comparing the activity of two characteristic nuclides in the waste by gamma spectroscopy or by using the ratio of active neutron signal to passive neutron signal, with the caveat that such ratios are only meaningful where good counting statistics are available in both data sets.

The combined active neutron/passive neutron/gamma spectroscopy method therefore provides a highly versatile method for characterization of radioactive sludge waste.

PERFORMANCE EVALUATION CASE STUDY: ROCKY FLATS IPAN

An IPAN system at Rocky Flats was used to develop and test the sludge assay technique. This system had previously been calibrated for 208 liter drums of heterogeneous debris waste and certified to meet waste characterization requirements for disposal at the Waste Isolation Pilot Plant (WIPP) [1]. In November 2002, a development program was undertaken to extend the range of WIPP certification to include the site's sludge waste.

Calibration measurements were performed with the IPAN using a surrogate sludge drum. Two calibration sources were used: a Cf-252 spontaneous fission source with an effective Pu-240 mass of approximately 2.45g (at 11/15/2002) equivalent to 40.8g of weapons grade Pu (WG Pu) and a depleted uranium source with an effective Pu-239 mass of 1.159 g equivalent to 1.237 g WG Pu. Measurements were performed with each source positioned at twelve reference points in the drum. Software and checking procedures were used to confirm correct operation of the system. Measurement control runs were made each day in accordance with the operating procedure.

Table II. illustrates the effect of highly interfering nature of the sludge matrix in active and passive neutron mode for the IPAN. Similar statistics are provided for a sample of heterogeneous debris surrogate drums (light, medium and heavy debris and metals) for comparison. The "matrix perturbation factor" is defined as the volume average efficiency for a particular matrix divided by the corresponding efficiency for a non-interfering matrix (i.e. the empty drum). The detection limit in active mode is given in terms of milligrams of Pu239. As expected, this is a function of both net weight and matrix perturbation factor. When considering the minimum detectable concentration (MDC), i.e. detection limit expressed as alpha activity normalized to net drum weight, the light debris matrix shows abnormally high MDC because of the small denominator (net weight). The final estimate of MDC for sludge shows that the IPAN can reliably achieve a MDC less than 100nCi/g and thus the instrument meets the WIPP disposal criteria and enables TRU/LLW sorting of sludge.

Table II. Comparison of Detection Limits for RFETS IPAN for debris waste and sludge.

Matrix	**Light Debris**	**Medium Debris**	**Heavy Debris**	**Metals**	**Sludge**
Net Matrix Mass (kg)	14.6	72.5	122.5	156.6	190.0
Active Mode Matrix Perturbation Factor	1.02	1.16	2.26	3.27	16.33
Passive Mode Matrix Perturbation Factor	1.13	1.09	N/A	1.01	16.75
Detection Limit (mg Pu239)	9.4	13.6	17.6	47.3	152.1
Minimum Detectable Conc. (alpha nCi/g)	51.7	15.1	11.54	24.3	64.3
Minimum Detectable Conc. (alpha Bq/g)	1913	559	427	899	2379

A series of calibration verification measurements were performed using the new sludge calibration in order to demonstrate instrument performance. In addition to providing verification that the calibration is appropriate for plutonium, these measurements yield an estimate of the accuracy (bias) and precision of the measurement method under normal operating conditions.

The sludge calibration verification involved loading a surrogate sludge matrix with plutonium standards with certified Pu mass. The placement of each standard in the surrogate matrix was carefully controlled to ensure that the source weight distribution represents an even radial distribution of plutonium within the surrogate drum. In order to achieve this, the matrix volume is notionally divided into four concentric horizontal rings placed over the vertical source loading tubes. The horizontal cross sectional area of each ring expressed as a percentage of total cross sectional area of the drum is given in Table III. These percentages define the ideal distribution of source to simulate an even distribution of plutonium within the sludge. Clearly it is not possible to achieve a perfect match to this ideal case; however, by careful selection of standards it was possible to closely approximate the ideal case for most plutonium loadings.

Table III. Target loading of standards in the surrogate drum to replicate radial homogeneity.

Tube Number	Radius (cm)	Target source loading (%)
1	0	2.45%
2	8.6	15.13%
3	14.5	25.84%
4	23.1	56.58%

The accuracy and precision of the assay results were required to meet the interfering matrix quality assurance objectives used in WIPP DOE cross-site evaluation program known as the Performance Demonstration Program (PDP) [2]. System accuracy was assessed by determining the percent recovery (%R). This is defined as the average measured plutonium mass divided by the know plutonium mass, expressed as a percent. Thus 100 %R reflects the ideal measurement. The PDP requirement for interfering drums is %R in the range 40% to 160%. Precision is defined as percent relative standard deviation (%RSD). This is the standard deviation in the measured Pu mass (for a set of six replicates) divided by the known mass. The PDP requirement for precision depends on the activity loading. For example, for tests where the drums were loaded with between 0.02 and 2.0 alpha-Curies of activity (0.25 to 25g WG Pu), %RSD must be less than 12 %.

The results of the sludge calibration verification measurements are given in Table IV. From this data it can be seen that from 0.5g WG Pu to 169g WG Pu, all verification criteria were met. The active mode calibration yields satisfactory results below 25g WG Pu. The passive calibration yields satisfactory results from 25g to 169 g WG Pu. Thus 25g WG Pu was therefore selected as the threshold for use of active / passive mode.

Table IV. Summary of the RFETS IPAN sludge calibration verification results.

Pu Certified Mass Used (g WG Pu)	Mode Selected	Accuracy (%R)		Precision (%RSD)	
		Required Range	Measured Result	Required Range	Measured Result
0.53	Active	40%-160%	117%	<12%	11%
1.07	Active	40%-160%	86%	<12%	6%
6.98	Active	40%-160%	86%	<12%	3%
10.03	Active	40%-160%	93%	<12%	2%
24.95	Passive	40%-160%	89%	<12%	5%
169.03	Passive	40%-160%	104%	<6%	3%

FUTURE APPLICATION: HANFORD IPANs

The Waste Receiving and Processing (WRAP) facility at the Hanford Federal Nuclear Reservation intends to characterize the transuranic (TRU) waste content of 208 liter drums containing grouted sludge using its dual IPAN drum assay systems. As with the RFETS application, these measurements must meet Waste Isolation Pilot Plant (WIPP) characterization requirements.

After large pieces of SNF have been removed from the sludge, it will treated by mixing with Portland Cement and bentonite to form grouted sludge which will be placed in 208 liter drums with 0.23 cm thick liners.

The WRAP IPAN systems have a similar design and function to the RFETS IPAN system, with the additional capability to assaying 320 liter overpack containers.

Two sludge surrogate drums have been constructed: an inorganic sludge surrogate based on the aqueous precipitation process, and a sludge / grout mixture representing the NLOP waste stream for aqueous wastes employed at a number of DOE sites. The former surrogate was filled to 68.6 cm relative to the liner base, and the latter was filled to 73.7 cm.

The effect of gamma radiation is an important design consideration for assay of SNF waste. For most DOE contact handled (CH) waste streams, i.e. those with contact dose rate less than 2mSv/hr (200 mR/hr), the gamma emission is dominated by low energy photons (<200 keV) due to plutonium, uranium and their associated daughter products. Gamma radiation will not normally be counted by the IPAN's neutron detectors because (i) gamma ray interactions have a low pulse height and are usually rejected by careful selection of the pulse height discriminator in the amplifier, and (ii) the IPAN chamber walls provide shielding against low energy gamma rays.

The SNF sludge waste stream presents a unique challenge due to the high gamma radiation fields that will be generated by isotopes such as Cs-137 and Co-60. The high rate of gamma emission from these isotopes combined with the higher energies involved (e.g. 662 keV photons are emitted by Cs-137 decay) can cause interference with the neutron measurement and degrade the operational lifetime of the He-3 detectors. Interference can result at dose rates in excess of 2mSv/hr, due to pile up of low pulse height events. These events do not result in systematic bias in the neutron assay results because the active mode (late gate correction) and passive mode (coincidence counting) algorithms correct for these "noise" events. However the reduction in signal to noise ratio impacts the system precision and detection limit. Long term effects will also accumulate from degradation of the detector's quench gases as a result of exposure to high gamma radiation fields.

The IPAN technique can be readily adapted for assay of remote handled (RH) wastes, including SNF sludges. Several steps need to be taken to harden the system performance against gamma radiation associated with SNF. This includes adding internal gamma shielding, replacing the detectors with specialized gamma hardened detectors, optimizing discriminator settings and use of advanced counting electronics. The drawback of gamma hardening the IPAN system is that it results in under-optimization for the assay of contact handled waste stream. The WRAP IPANs are required for dual purpose assay of both SNF sludge and CH heterogeneous debris waste, therefore the settings will be optimized to enable the systems to assay both streams.

The NLOP treatment process removes large particles of SNF from the sludge waste stream. The maximum size of any SNF particle is 0.64 cm, however the actual particle sizes are assumed to be much smaller. The transuranic content will comprise a uniform spatial distribution of small particles with no significant self-shielding properties with regard to thermal neutron interrogation. At the 95% confidence level, the reduction in the active signal due to self shielding averaged over the transuranic content of the drum is estimated to be less than 40% compared to signal from the same quantity of material where there is no self shielding.

CONCLUSIONS

A new method has been developed for radioassay of sludge rad-waste using the IPAN system. The applicable waste streams that this technique can be applied to include TRU sludge waste originating from an aqueous precipitation process and waste from spent nuclear fuel (SNF) storage contaminated with fuel corrosion by-products including TRU isotopes and fission/activation products. In both cases, measurements are required after the waste drum has been stabilized by addition of a cement mixture.

This technique has been demonstrated to provide regulatory acceptable measurements on sludges at Rocky Flats. At Hanford, development work is being undertaken to adapt the IPAN system for combined assay of debris waste and SNF sludge waste. The high sensitivity and imaging capability of the IPAN are key factors underpinning this innovation.

The IPAN's active and passive operating modes provide a wide dynamic range of sludge assay: from milligram levels of Pu up to several hundred grams of Weapons Grade Pu Equivalent. The detection limits have been demonstrated to be below the TRU/LLW sorting threshold of 100 nCi/g (3700 Bq/g). For mixtures of uranium and plutonium, isotopic categorization can be performed by analysis of the ratio of the active and passive effective mass. Extensive validation and testing with surrogate sludge drums and radioactive standards have resulted in regulatory acceptance of this technique, permitting ultimate disposal of the sludge drums at the Waste Isolation Pilot Plant.

REFERENCES

1. Contact Handled Waste Acceptance Criteria for the Waste Isolation Pilot Plant (Appendix A), DOE/WIPP-02-3122, rev 2.
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