HIGH SENSITIVITY ASSAY OF CEMENT ENCAPSULATED SPENT NUCLEAR FUEL SLUDGE USING THE IMAGING PASSIVE ACTIVE NEUTRON (IPAN™) SYSTEM

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ABSTRACT

A new technique has been developed for high sensitivity assay of grouted spent nuclear fuel (SNF) sludge waste in 208 liter drums. The method uses the Imaging Passive Active Neutron (IPAN™) system to provide regulatory acceptable measurements. At the Waste Receiving and Processing (WRAP) Facility in Hanford, two IPAN™ systems have been successfully calibrated and validated for assay of SNF grouted sludge drums (encapsulated with a cement mixture). The systems have been demonstrated to be capable of performing low level waste (LLW) / transuranic (TRU) waste sorting even in the presence of high gamma radiation fields emitted by the fission and activation products associated with SNF. The active and passive modes of the IPAN™ provide a wide dynamic range of assay: from below the TRU/LLW sorting threshold (100nCi/g or 3700 Bq/g) up to several hundred grams of Weapons Grade Pu Equivalent. A new calibration technique was developed 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. Extensive validation and testing with specially designed surrogate grouted sludge drums and radioactive standards have resulted in regulatory acceptance of this technique, permitting ultimate disposal of the SNF sludge drums at the Waste Isolation Pilot Plant.

INTRODUCTION

The Hanford Sludge Retrieval and Disposition Project (SRDP) disposal of North Load Out Pit (NLOP) waste will produce 208 liter (55 US gallon) grouted sludge drums bearing Spent Nuclear Fuel (SNF). The drums are required to be assayed with a detection of less than 100 nCi (3700 Bq) of transuranic (TRU) alpha activity per gram of waste matrix in order to correctly sentence the final waste form. At the Waste Receiving and Processing (WRAP), two identical IPAN™ systems which had previously demonstrated this low detection limit capability for debris waste were selected as the appropriate technology for assay of this challenging waste stream.

In order to meet the required measurement objectives including demonstration of precision and accuracy requirements for the disposal of the waste at the Waste Isolation Pilot Plant (WIPP) in New Mexico, the following program of work was undertaken:

(i) Development of suitably representative non-radioactive sludge surrogate drums,
(ii) Calibration of the IPAN™ systems using a novel imaging technique,
(iii) Testing of the systems using special nuclear materials and high energy gamma emitting fission product standards and
(iv) Demonstration of the performance requirements under strict a Quality Assurance program.

The implications of the successful completion of this work have far reaching consequences for the radioassay of remote handled waste streams. The new method offers a viable alternative measurement approach over the “dose to Curie” method, without the latter method’s reliance on operator records and sampling regimes.

OVERVIEW OF THE IPAN™ TECHNIQUE

An IPAN™ System is depicted in Figure 1. The measurement technique comprises active neutron interrogation and passive neutron counting in a single assay chamber. The results may be combined with isotopic information from either acceptable knowledge or a gamma spectroscopy measurement in order to determine the TRU alpha activity of the waste.

The active neutron mode measures neutron-induced fission events in fissile material such as Pu-239 within the waste. Neutrons from an interrogating source are introduced into a measurement chamber consisting of moderating and shielding materials. These neutrons induce fission events in fissile material, giving rise to the emission of secondary fast neutrons. The IPAN™ chamber design maximizes the sensitivity to secondary neutrons, while minimizing the signal from the interrogating source [1]. This method is known as differential die-away (DDA). Short pulses of fast neutrons from a neutron generator are injected into the measurement chamber. This gives rise to a thermal neutron flux, which persists for a few milliseconds. Fast neutrons arising from the induced fission events are then counted using fast neutron detector packages embedded in the chamber walls. Using data derived in system calibration, the measured signal is converted into the effective Pu239 mass (Pu-239eff).

Fig. 1 IPAN™ system

Passive neutron counting consists of measuring the intrinsic fast neutron emission from the waste. The passive neutron coincidence counting technique exploits the fact that neutrons from spontaneous fission are emitted essentially simultaneously, whereas background neutrons and neutrons emitted from (alpha,n) reactions are non-coincident. Passive neutron measurements are used to quantify even isotopes of plutonium and other spontaneous neutron emitters. The system calibration converts the coincidence signal to Pu-240eff mass.

The passive and active neutron measurement data are processed through a mathematical imaging algorithm. The purpose of the imaging algorithm is to reduce the measurement uncertainty due to source positioning variation within the waste containers. The result is improved sensitivity and regulatory compliant precision and accuracy of results [2].

SLUDGE WASTE CHARACTISTICS

The final waste form will comprise NLOP sludge encapsulated in a cement and bentolite matrix within a 208 liter drum. The sludge comprises corrosion products from underwater storage of spent nuclear fuel and will therefore contain particles of uranium and embedded fission / activation products and transuranics including plutonium. In order to simulate the sludge, two non-radioactive surrogates were built to the specification given in Table 1. Note that after the specified amount of water was added to the dry sludge, the supernatant was decanted and the precipitate was filtered (but not dried) and mixed with cement. The mixture was poured into a 55 gallon drum with a polyethylene liner and allowed to cure. The drum contains 3 polypropylene insert tubes that allow radioactive standards (up to 2 inches in diameter) to be loaded with the matrix at various radii and heights. Spacer inserts were provided with each drum. These are small aluminum cylindrical containers containing the same sludge / cement matrix used in the drum. The spacers are used to fill dead space in the source insert tubes in order to prevent neutron streaming through the insert tubes.

	Sludge “M1”	**Sludge “M2”**
Description	Dry inorganic sludge mixture based on carrier precipitation process mixed with water and cement.	Dry sludge mixture representative of NLOP stream mixed with water, cement and bentolite.
Fill Height (cm)	68.6	73.7
Sludge : Cement mass ratio	7.4 : 1	1 : 38.5
Bentolite added (kg)	0	17.6
Water added (liters)	720*	106
Spacers (kg)	4.8	6.5
Final Matrix Mass (kg)	183.9	241.9

Table 1- Sludge Surrogate characteristics

The primary physical differences between the surrogate drums is the amount of water added to the sludge recipe. Hydrogen is a very effective neutron moderator and also has a high absorption cross section for thermal neutrons. Thus water content is the most important physical characteristic of the sludge drums with regard to neutron measurements because the drum's final hydrogen concentration is dominated by the amount of water in the sludge.

CALIBRATION TECHNIQUE

Calibration measurements were acquired at the WRAP facility in February - March 2006, using the two surrogate sludge matrices in 55 gallon drums. A 50 g Weapons-Grade (WG) Pu standard with a Pu-240eff coincidence mass of 3.03 g was used for passive mode calibration measurements and a 1.0g WG Pu standard with a Pu-239eff mass of 0.9408g was used for active mode calibrations. Measurements were performed with each source positioned at 12 different reference points in the 55 gallon surrogate drums.

The sludge calibration library files were constructed using a radial averaging method. For a given source height, the response factors from each tube were averaged together using the weighting factors given in Table 2 in order to simulate the effect of source material uniformly distributed across the drum radius. This averaging is done to simulate the expected spatial distribution of source material in homogeneous sludge. As a consequence the imaging matrix term has 3 volume elements (Height 0, 20 and 40 cm) for each matrix, in contrast to the 12 volume elements used in the standard debris calibration.

Tube Number	Radius (cm)	Uniform Distribution Weighting Factor (%)
1	0	2.45%
2	8.6	15.13%
3	14.5	25.84%
4	23.1	56.58%

Table 2 Sludge Calibration Tube Weighting Factors

DETECTION LIMITS

The lower limit of detection (LLD) in active and passive mode was determined by performing replicate blank measurements on the surrogate sludge drums with and without a 113mCi (4.18 GBq) Cs-137 source present in the center of the drum. The method for calculating detection limit is based on the Currie formula [3]. The LLD values are based on the assumption of a uniform distribution of material across drum radius and height.

The active mode detection limit has been converted to a nCi/g equivalent minimum detectable concentration (MDC) based on WG Pu isotopics (6% Pu-240/Pu), 12% Pu-240/Pu, 18% Pu-240/Pu and for K-Basins Container Sludge average isotopics [6, 7] and NLOP Grouted Sludge AK isotopics [8]. The latter two grades represent typical SNF sludge for the Hanford site.

For SNF sludge isotopics, the nCi/g MDC calculation includes a correction for the presence of U-235 in these streams. For these cases MDC is calculated as follows:

$$MDC(nCi/g) = K1(g\,Pu/\,fiss\,sig)K2(/\,g-matrix)K_{TRU\alpha}(nCi/g\,Pu)FS_{LD}$$

where,

FS_{LD} is the fissile signal detection limit,
$K_{TRU\alpha}(nCi/g\,Pu)$ is the TRU alpha activity per gram of Pu,
$K2(/\,g-matrix)$ is the inverse of the surrogate drum net weight and
$K1(g\,Pu/\,fiss\,sig)$ is defined as follows:

$$K1(g\,Pu/\,fiss\,sig) = \frac{m_{CAL}(g\,Pu-239)}{\left(\overline{FS_{CAL}} - \overline{FS_0}\right) r_{Pu-239eff}}$$

where $r_{Pu-239eff}$ is the mass fraction of Pu-239eff to total plutonium defined as follows:

$$r_{Pu-239eff} = r_{Pu-239} + 0.652 r_{U-235}$$

where,
r_{Pu-239} is the mass fraction of Pu-239 relative to total Pu and
r_{U-235} is the mass fraction of U-235 relative to total Pu.

The 0.652 factor is the Pu-239eff factor for U-235. Thus the presence of -U235 in the waste stream has the effect of decreasing the nCi/g MDC.

For sludge measurements, a lower limit of detection of less 0.061 g Pu-239e (less than 100nCi/g) in active mode has been successfully demonstrated. In the passive mode, the detection limit is approximately 0.04g Pu-240e (0.6g WG Pu).

In active mode, the LLD with and without the Cs-137 source is shown in Table 3 for various isotopic grades. It can be seen that the presence of the Cs-137 leads to only a small increase in the system’s active LLD. The passive mode LLD was not effected by the presence of the Cs-137 source.

Isotopic Grade	**Active mode MDC with no Cs-137 (nCi/g)**	**Active mode MDC with 113 mCi Cs-137 (nCi/g)**
Hanford 6% Pu-240 / Pu	20	25
Hanford K-Basins Container SNF	9	11
Hanford NLOP Grout SNF	12	16

Table 3 Sludge detection limits for various isotopic grades

SATURATION EFFECTS

For sludge measurements, an increased total interrogation flux is required to yield acceptable lower levels of detection. Consequently the number of pulses per grab was increased from 625 (standard value for debris waste) to 1875 for sludge active measurements. In addition, passive count time was increased from 20s to 60s per grab for sludge. The reason for increasing the number of pulses per grab (i.e. effectively increasing count time), rather than operating the tube at higher voltage, is that if the neutron generator output is set too high, the matrix correction algorithm may be biased. This is because the algorithm uses the epithermal neutron signal in the 200-250 microsecond interval after the neutron pulse. With too high a neutron generator output, the detectors may not recover sufficiently quickly and some of the neutron pulse will leak into the epithermal gate. A bounding range of neutron generator output is therefore required for operations. The lower limit is dictated by detection limit requirements and the upper limit is dictated by the matrix selection method. It is also preferable to maintain a relatively low neutron generator output in order to extend the tube’s operational lifetime.

VALIDATION

Checks were performed for both systems with sludge surrogates with a range of known Pu source loadings. The results of the tests are provided in Table 4 through Table 6. The %R (average measured result divided by tag mass) and %RSD (standard deviation divided by tag mass) are provided. It can be seen from this data that the validity of the calibration is confirmed in active mode and passive mode up to 198 g WG Pu for both sludge matrix surrogates.

Matrix	**Active Mode**		**Passive Mode**	
	%R	*%RSD*	*%R*	*%RSD*
Sludge M1	93.3%	5.7%	N/A	N/A
Sludge M2	87.6%	3.0%	N/A	N/A

Table 4- Sludge Active Mode Validation at 1.9 g WG Pu

Matrix	**Active Mode**		**Passive Mode**	
	%R	*%RSD*	*%R*	*%RSD*
Sludge M1	83.4%	1.5%	64.5%	2.8%
Sludge M2	83.2%	3.1%	57.4%	0.9%

Table 5- Sludge Active / Passive Mode Validation at 9.9g WG Pu

Matrix	**Active Mode**		**Passive Mode**	
	%R	*%RSD*	*%R*	*%RSD*
Sludge M1	N/A	N/A	108.3%	2.2%
Sludge M2	N/A	N/A	74.7%	1.5%

Table 6- Sludge Passive Mode Validation at 198 g WG PU

SLUDGE WASTE ACTIVE / PASSIVE SELECTION

Generally the active mode is most suited for lower Pu-239eff mass drums were self-shielding effects are minimal. Conversely passive mode is best suited to the high Pu-240eff mass because statistical precision is poor for low mass cases. For sludge measurements it is recommended that passive mode is used when Pu-240eff exceeds 1.2g (the equivalent of 20g WG Pu).

Note that with homogeneous sludge waste, the effect of self-shielding in active mode is likely to be less severe than for debris measurements because (i) the average particle size in sludge is generally smaller than for debris, (ii) the source material is likely to be distributed over the volume of the drum and (iii) large pieces of SNF are removed from the sludge. Therefore the active mode is applicable for sludge up to a higher mass of Pu-239eff than for debris waste.

TOTAL MEASUREMENT UNCERTAINTY

Systematic uncertainty due to matrix effects arises due to residual differences between the neutronic properties of the real waste and those of the materials used to represent them in the calibration. The WRAP IPAN™ systems were calibrated with surrogate sludge matrices possessing neutronic properties that span the range of the expected sludge matrix types.

The systems use the measured response from flux monitors and specific detector banks to select the calibration matrix with bulk neutron absorption and moderation properties similar to those of the measured sludge waste drum. These selection factors apply to both active neutron and passive neutron analyses. Because the neutron moderation and absorption properties are measured, knowledge of the elemental composition of the waste matrix is not required.

The systematic uncertainty terms for sludge assay have been estimated by reviewing data acquired in linearity testing. As expected, the matrix term is the dominant uncertainty component. Uncertainty due to source heterogeneity uncertainty is relatively low because the sludge is expected to have a uniform distribution across the drum radius and approximate uniform distribution with height. Estimates of uncertainty are given in Table 7 which presents a comparison on the uncertainty terms for heterogeneous debris waste and sludge. In the final calculation of systematic uncertainty term for the IPAN™ systems, these uncertainty components are added in quadrature. The sludge overall systematic uncertainty is lower than that for debris waste. This is as expected because the sludge will have less variability in terms of matrix properties and source distribution than the debris waste.

Waste Type	Active Matrix Uncertainty	Active Source Distribution Uncertainty	Passive Matrix Uncertainty	Passive Source Distribution Uncertainty
Debris Waste	31.5%	12.9%	15.0%	4.4%
Sludge	15.8%	4.2%	11.7%	6.3%

Table 7- Comparison of Matrix and Source Distribution One Sigma Uncertainty Terms

IMPLICATIONS FOR ASSAY OF REMOTE HANDLED WASTE

Measurements with the WRAP IPAN™ systems have proven that Cs-137 content up to 113.1 mCi does not have a significant impact on the measurement. With this source embedded, the surrogate sludge drum sludge drum M1 had a dose rate of 3.7 mSv/hr (370 mR/hr) at contact and 0.75 mSv/hr at 30 cm. No statistically significant increase in passive totals count rates was observed in the cadmium shielded He-3 detectors when Cs-137 is present. This indicates that the IPAN™’s neutron detection chain is capable of rejecting gamma ray induced events for containers with contact dose rates of up to 3.7 mSv/hr.

These results are particularly encouraging given that the WRAP systems were developed for assay of contact handled TRU waste where gamma ray emission are of lower energy and lower activity than remote handled waste. For neutron assay equipment that is built specifically for high gamma radwaste, the detectors are shielded by lead or other high atomic number materials. In addition, the gamma rejection capability of the neutron detectors may be improved by use of advanced electronics and careful selection of the neutron detectors. For example it has been demonstrated [9] that, aluminum wall He-3 detectors with CO_2 quench gas, produce excellent performance in high gamma radiation fields of up to 0.2 Sv/hr (20R/hr) from a Co-60 source. We would therefore expect that by use of shielded, gamma ruggedized neutron detectors and advanced electronics, the IPAN system will be capable of achieving a detection limit of less than100 nCi /g for waste containers with dose rates up to 10 Sv/hr (1000R/hr).

Traditionally NDA measurements are considered to be of limited applicability to RH waste measurements because of the twin problems of Compton scattering (in relation to gamma spectroscopy) and gamma pile-up (in relation to neutron assay). The development of an assay system capable of TRU/LLW sorting on RH waste represents a major breakthrough in this field. The IPAN technique offers an alternative measurement approach over the “dose to Curie” method (i.e. estimation of the TRU activity content by measuring the gross gamma count or dose rate of the drum and applying scaling factors based on an assumed drum isotopic mixture). The NDA approach offers improved accuracy without the need to assume a fixed relationship between fission product activity and the TRU alpha activity for each waste container. This reduces the burden on sites in terms of compiling defensible operational records for individual containers and the need to perform extensive sampling programs on individual waste streams.

SUMMARY

It has been demonstrated that the IPAN™ measurement technique is suitable for high sensitivity assay of cement encapsulated sludge TRU waste bearing SNF debris with high gamma activity. Detection limits of less than 100 nCi/g (3700 Bq/g) have been achieved in the active mode. A new imaging method has been developed that utilizes a radial averaging approach tailored for the homogeneous sludge waste form. An investigation of the impact of high energy gamma radiation on the system detection limits indicated was performed using a 113mCi Cs137 source that produced contact dose rates up to 370 mrem/hr at the drum surface. There was no statistically significant impact on passive mode LLD and only a relatively small increase in the active mode LLD. These results are particularly encouraging given that the WRAP systems were not specifically tailored to deal with intense gamma radiation. The addition of shielding materials and gamma hardened neutron detectors to the IPAN™ design will further improve the applicability of this method to the measurement of high gamma emitting radwastes. This leads us to conclude that the IPAN™ method will be well suited to TRU/LLW sorting of RH TRU waste.

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