ASSAY OF URANIUM CONTAMINATED WASTE IN INTERMODAL CONTAINERS BY PASSIVE NEUTRON MEASUREMENT AT THE OAK RIDGE 3 BUILDING D&D PROJECT

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ABSTRACT

Waste consignments in Intermodal and SeaLand containers from the BNFL Inc. 3 Building D&D project at Oak Ridge, TN are successfully assayed for U235 content prior to shipment for disposal – these measurements being necessary to meet nuclear safety and transportation regulations, and disposal acceptance criteria. The decommissioning operations at this former gaseous diffusion plant generate large quantities of metallic waste and each Intermodal contains upto 20 tons of material which comprises components from converters, compressors, valves, motors, pipework etc. The assay station for these filled Intermodal containers is a large shielded chamber incorporating an extensive array of neutron detection modules.

This report describes the radiometric features of the BNFL Inc 'Uranium Waste Monitor' chamber and the performance of this system during commissioning and through the early program of waste measurements. Of particular interest is the approach taken to remove the component of neutron background resulting from cosmic ray generated spallation events in the high masses of steel in each container.

INTRODUCTION

On August 25, 1997 the US Department of Energy (DOE) and BNFL Inc signed a contract for the East Tennessee Technology Park (ETTP) Three Building Decontamination, Decommissioning and Recycle Project at the former K25 site, Oak Ridge TN. This is a fixed-price contract to dismantle, remove and decontaminate and/or dispose of the process equipment and support systems within the three gaseous diffusion plant buildings – K29, K31 and K33. [Figure 1 shows an aerial view of this facility.] Some recovered metals will be recycled whilst the remainder will be consigned as low level radioactive waste (LLW) and transported in large waste containers to licensed disposal sites.

Fig. 1. Aerial view of the former K25 site

In response to the need for characterization of high density metallic wastes from the project BNFL Inc. has received a passive neutron based Uranium Waste Monitor (UWM) supplied by BNFL Instruments. The UWM was ordered in late 1998, developed and fabricated in the BNFL Instruments workshops in the UK, assembled in the US and delivered in April 1999. Set-up and system commissioning was completed in Summer 2000.

Use of the UWM provides two important benefits to the project – first, reduced sampling and analysis costs for the wastes and second, a total U estimate for each LLW consignment that is more accurate than that attainable from in-situ gamma NDA. Using this system, the data to support compliance with recycle facilities and disposal site acceptance criteria for SNM from this project are greatly improved

WASTE CHARACTERISTICS AND REASONS FOR MEASUREMENT

Decommissioning the gaseous diffusion plants in K29, K31 and K33 will, in total, result in over 100,000 tons of waste steel. Transportation of this waste to licensed disposal sites will be in Intermodal, SeaLand, ST90 or other large waste containers - each LLW consignment largely consisting of stainless steel components and small amounts of other metals, size reduced as necessary to achieve an optimum packing fraction in each container.

The dimensions of an Intermodal are approximately 20' x 8' x 8' with each container likely to hold up to 45,000 lb (20 tons) waste.

For the purposes of this project the distribution and degree of dispersion of low enriched uranium (LEU) contaminated waste within any individual Intermodal is not assumed to be uniform or homogenous in order to truly represent the real world situation.

Strict regulations and limits exist for both the transportation and disposal of LLW. In particular, nuclear safety considerations place a 350g U-235 limit on the total inventory of some disposal and recycle facilities. Combined with overestimation of gram quantities in waste shipments this limit places a constraint on the throughput of material in these facilities. A more accurate methodology for assay of U-235 is therefore required to assist with meeting throughput needs.

LLW CHARACTERIZATION STRATEGY

The waste characterization strategy for LLW arising from the ETTP 3 Building D&D Project is built up from the following components:

- Collection of historical data from sample analyses obtained during K33 operation and maintenance phases of each building has been a key initial stage of this strategy. This information was used to build a profile of U-235 enrichment, U-234 enrichment and Tc-99:U ratio across all units and cells. Additional radiochemical analysis of samples as part of the decommissioning program was instigated where existing plant data was uncertain or unavailable.

Fig. 2. Dismantling a Converter

During the early decommissioning stages a high proportion of size reduced waste items or process vessel components have been surveyed for their uranium contamination levels using radiological hand held instruments (HP&S probes). In addition, the physical dimensions and weight of each item was recorded and a database of values developed. This data was then used to determine 'activity/unit waste mass' values for defined waste components from which estimates of the total uranium content of filled transport containers has been established. It is important to note, however, that some wastes are not suitable for surface contamination survey by hand held probes. These will usually be characterized as large quantities of small items, items with inaccessible surfaces or bulk granular or powdered waste that essentially constitutes a volume source.

- As decommissioning operations continue (from unit to unit) confidence has been developed in an ability to predict the residual activity levels from each identified component of the size reduced vessels based on previous survey data. This increased confidence allows a reduced frequency of contamination surveys and waste dimension measurements to be carried out.
- In parallel, the frequency of NDA measurements using the UWM on filled transport containers has been increased as the surface survey frequency decreased. NDA measurements allow confidence to build in the fact that large quantities of uranium are not being overlooked during LLW size reduction and cleaning processes prior to the loading of transport containers. In this way NDA measurements provide confidence in, and confirmation of, the radionuclide activity values quoted on each shipping manifest.

Ultimately, the frequency of NDA measurements using the UWM will be reduced to that of a 'customs' check. Alternatively, NDA measurements can remain at a high level where facility records or radiochemistry sampling suggests that problems may have occurred at specific locations during plant operations, resulting in non-typical contamination conditions.

URANIUM WASTE MONITOR (UWM) – SELECTED ASSAY TECHNIQUE AND SYSTEM DESCRIPTION

During the development phase of the UWM a number of radiometric measurement methods were considered against criteria which included practicability, assay accuracy and lifetime costs. Uranium has limited passive radiation emissions. Foremost amongst these is the abundant gamma emission of U-235 at 185.7 keV, routinely used for assay of radwaste bags and drums from the operational or decommissioning phases of uranium enrichment/fuel fabrication facilities. However the utilisation of this radiation was not selected due to the ready attenuation by even modest quantities of metallic waste. Although of generally higher energies, similar problems exist for the gamma radiation from U-238 and its daughters, especially Pa-234m. Because of the low penetration of uranium gamma radiation accurate (external) measurement of LEU concentrations at the center of an Intermodal containing approximately 20 tons of metallic waste was not considered to be reliable.

By contrast fast neutrons are highly penetrating - especially in the metallic and low moderator matrices of LLW from the ETTP 3 Building Project. Although the spontaneous fission generated neutron emission of LEU is relatively small, the (α, n) interactions between the alphas emitted by uranium isotopes (predominantly U-234) and surrounding light nuclei such as fluorine do generate a measurable neutron signal. This penetrating neutron radiation from the (α, n) interactions of residual UO_2F_2 and UF_6 therefore forms the basis of uranium measurements by the UWM for LLW transport containers destined for disposal.

Fig. 3. Uranium Waste Monitor Chamber

The UWM shown in Figure 3 is a unique passive neutron counter (PNC) capable of accurate uranium assay for a range of freight containers including Intermodals, ISO (SeaLand) containers, ST90 boxes, and drum packs. To achieve this the system employs 80 polyethylene clad neutron counting modules [known as DISPIM®] together with electronics that provides regulated high voltage distribution, signal conditioning and amplification for 12 independent neutron counting channels. The UWM container movement controller and readout computer are located in an adjacent operator area, with data logging and remote readout capabilities. The system utilizes 160 pressurized He-3 thermal neutron detectors of standard design; two detectors per DISPIM® neutron counting module.

All DISPIM® modules are deployed on the walls, floor and ceiling of the assay chamber to provide a '4π' counting geometry for large waste containers. Using a mathematical model (MCNP) of the counting arrangement, optimum locations were found for each of the 80 DISPIM® modules within an assay chamber capable of accommodating the largest waste container.
For this optimized arrangement an average neutron detection efficiency of 5-6% was predicted. In addition, the uniformity of response for a uranium concentration in this chamber was expected to be better than +/-20%, making the chamber measurements quite independent of the location or degree of dispersion of uranium within expected waste loads.

As with the measurement of any small radiation signal it was vitally important to control ambient background levels such that they are as low as possible and remain steady during the complete measurement cycle of each waste container. To identify the best measurement location available close to the decommissioning operations a neutron background survey of the site was carried out. Building K-761 at ETTP was found to be an appropriate location with low ambient background and sufficient space around the proposed monitor location to construct additional neutron shielding if required.

A final challenge for the project concerned the additional source of background 'spallation' neutrons caused by cosmic ray generated interactions with the waste carbon-steel matrix of each filled Intermodal during measurement. These neutrons must be accounted for as they contribute to the measured count rate yet are not related to the amount of uranium in the chamber. Test neutron measurements carried out in the UK of steel masses up to 15 tons showed a valuable correlation between the waste mass and additional neutron fluxes – detected as either as single neutron events or as multiple events. Of particular interest were the multiple events that are generated as a result of cosmic ray interactions occurring as "showers" producing many time correlated neutrons in large metal masses. Although the use of neutron multiplicity counting electronics was considered as a means of removing this 'background' component a rather simpler approach was finally selected. The intensity of spallation neutrons was readily demonstated to be linearly related to the mass of waste steel in a consignment. This relationship therefore presented a useful means of removing the spallation background component given that the weight of steel could be readily established for each LLW consignment. To minimize both local and cosmic ray generated spallation neutron backgrounds while also improving the sensitivity and stability of the UWM system, the entire detector array at Oak Ridge is housed in a 24" thick concrete shield constructed by O'Brien & Gere Engineers, Inc. (OBG).

Fig. 4. Shielded Chamber Door

LLW transport containers enter and leave the assay chamber through doors at one end. (See Figure 4) These doors also support a number of DISPIM® neutron detection modules together with a composite layer of polyethylene/concrete to complete the background shielding. Filled waste containers that are ready for assay are loaded onto a rail-guided trolley located outside the chamber. Once loaded the trolley is mechanically driven into the chamber and the shielded doors are closed. Assay times for full waste containers are usually between 30 and 60 minutes (depending on the required minimum detectable activity) after which the container is removed and the consignment manifest completed in preparation for onward transportation of the waste to the disposal or recycle facility.

Fig. 5. Intermodal Calibration Container

URANIUM WASTE MONITOR - PERFORMANCE

Neutron detection efficiency measurements carried out during commissioning in building K-761 at ETTP show that the system has an average efficiency of 5.2%. Figure 5 shows the lattice structure of the calibration container that allowed accurate positioning of both calibration sources and simulated waste matrices during commissioning. The efficiency variation with source position within the volume envelope of a standard Intermodal or SeaLand container was found to be 10 - 15% of this mean value. This was in good agreement with our MCNP modeling of the assay chamber.

Measurement of uranium by this technique requires that the system accurately determine the count rate, which is attributable only to the (α, n) neutron emissions of UO_2F_2 or UF_6. In the absence of all background contributions the measured signal from the UWM is typically 2 cps per 10 grams U-235 in the form of UO_2F_2. The background count rate, which is typically 25 cps is determined both immediately before and after each waste container measurement and is established with the chamber empty. However, as cosmic ray spallation is a source of neutron background that is most apparent during the waste container measurement BNFL Instruments implemented the following correction method. As a result of development work in the UK and commissioning trials at ETTP a relationship was established between the mass of carbon steel in the chamber and the enhanced total neutron count rate introduced by spallation neutrons. This source of background neutrons was found to introduce approximately 0.5 cps into the system per 1000 kg of carbon-steel in the chamber (or about 0.25 cps per 1000 lbs). Figure 6 shows the extra neutrons counted as a function of the mass of material introduced into the chamber. The net weight of waste in each transport container can therefore be used to evaluate and remove the background contribution due to spallation neutrons generated in the waste from the UWM total count rate.

Stability of the system and accuracy of the spallation correction enables the UWM to deliver a total measurement uncertainty at the 1 sigma level of 10 grams U-235 for UO_2F_2 and 5 grams U-235 for UF_6 at enrichments typical of that present at ETTP. Count times are typically 30 to 60 minutes.

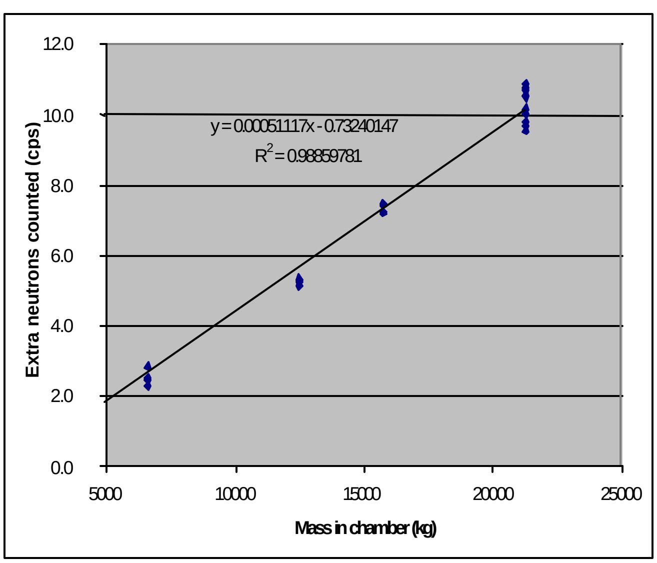

Fig. 6. Spallation effect on Carbon Steel

URANIUM WASTE MONITOR – OPERATIONAL EXPERIENCE

The UWM system has been operated since August, 2000 for measurement of SeaLand Containers, Intermodals and ST-90 containers. Since inception of operations, the system has measured over 150 units of waste.

The data from these measurements is summarized in Figure 7 which shows the proportion of U-235 assayed from the various components of the plant. As expected the converter and compressor parts, being subject to some of the most energetic contact with UF_6 during plant operations, also have the highest uranium content.

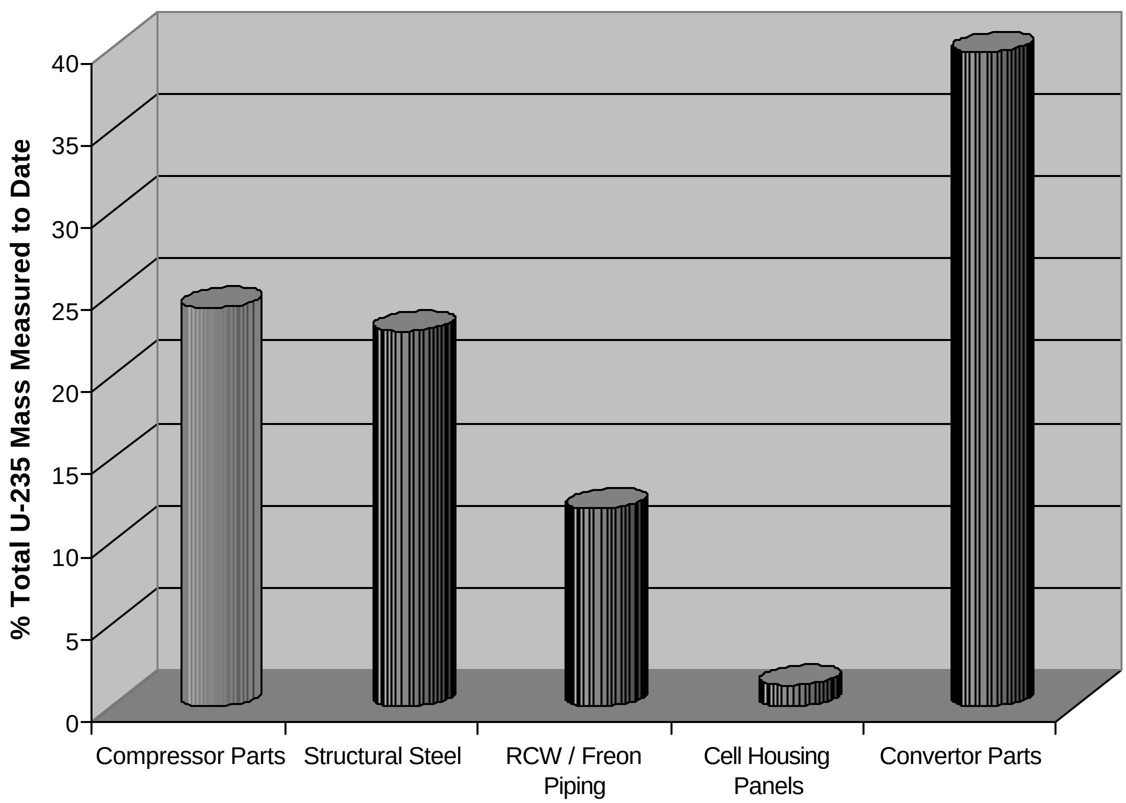

Fig. 7. Percentage of Total U-235 Measured for each Waste Material Type

Table I: Summary of Results from UWM Measurements to date.

ITEM	Percentage Total Waste Mass	Percentage Total U-235	Grams (U-235) / lb of Waste Material
Compressor Parts	6.10%	24.20%	0.0025
Structural Steel	41.90%	22.80%	0.0003
RCW / Freon Piping	18.50%	12.00%	0.0004
Cell Housing Panels	2.70%	1.30%	0.0003
Converter Parts	30.80%	39.70%	0.0008

CONCLUSIONS

The characterization of gaseous diffusion plant low level radioactive wastes from the Three Building D&D operations at ETTP have been simplified by applying NDA measurement of large waste containers filled with metallic scrap weighing upto 20 tons. The relatively consistent radiological characteristics of the gaseous diffusion plant waste and the accurate characterization of the waste using sampling and analytical measurements can be applied to a bulk waste NDA program to reduce sampling and analytical costs, and increase shipment frequencies.

The BNFL Instruments Uranium Waste Monitor has now measured over 2 million pounds of this waste since the Fall of 2000. As a result of these measurements, the reported levels of U-235 and the effort required to characterize these wastes have been reduced. Further, the reduction in reported levels of U-235 has resulted in high throughput rates for 350g U-235 limited facilities such as disposal and recycle facilities.

A key component of the success of the Uranium Waste Monitor project has been the selection of a low neutron background area for measurement together with the implementation of effective methods for the shielding and correction of the spallation neutron background. This background is an inevitable consequence of this measurement approach and is caused by the interaction of cosmic ray generated particles with the high assayed masses of steel.

REFERENCE DOCUMENTATION

1. "Passive Nondestructive Assay of Nuclear Materials" by Reilly, Ensslin, Smith Jr. NUREG/CR-5550, LA-UR-90-732.
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3. "Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM)" NUREG-1575, EPA 402-R-96-018, NTIS-PB97-117659.