MEASUREMENT OF THE URANIUM CONTENT OF INTERMEDIATE LEVEL WASTE FROM THE SELLAFIELD DRYPAC PLANT BY ACTIVE NEUTRON INTERROGATION

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ABSTRACT

The Sellafield DryPac Plant (SDP) is a new facility for the treatment of Intermediate Level Waste (ILW), prior to long term storage in a future Nirex repository. A current Nirex requirement is that the fissile content of waste packages shall not exceed a predefined post closure case criticality limit. In order to police this limit the plutonium content of each SDP product drum will be measured by Passive Neutron Coincidence Counting (PNCC), using a pair of custom-designed assay chambers. These instruments have already been assembled and subjected to works and customer acceptance tests.

It has recently been decided to incorporate an active neutron interrogation capability into one of the existing passive neutron monitors (PNM). This is intended to allow the fissile uranium content to be measured for selected drums, containing waste from particular streams, which are known to have a significant inventory of enriched uranium. Active neutron interrogation is hence necessary for these drums, to avoid the risk of unknowingly exceeding the criticality limit due to ^{235}U.

This paper describes the modifications which are now being been made to one of the passive neutron monitors, to incorporate a californium shuffler system, with a description of the physics and engineering design of the system. The proposed methodology for the shuffler calibration, using simulated waste matrices, is also described.

INTRODUCTION

The Sellafield DryPac Plant (SDP) is a new facility, which will treat Intermediate Level Waste (ILW) arisings from principally Magnox fuel reprocessing operations at BNFL's Sellafield site in the United Kingdom, and convert it into a form suitable for encapsulation in drums for storage on site and destined for disposal in a future Nirex repository. During the waste sorting process, small items and sludge waste will be collected as "undersize" waste. Physically large items will accumulate as "oversize" waste, fuel – bearing items will be broken down and returned to the undersize stream. Waste will be collected in 200 litre sacrificial drums, which are dried and compacted to form "pucks" (oversize waste is compacted without drying). Pucks are then placed

into 500 litre concrete – lined product drums (roughly 3 and 5 pucks per drum, for undersize and oversize waste respectively).

The total fissile content of each product drum is subject to post closure criticality limits set by Nirex. To this end, knowledge of the fissile mass inventory is required. For undersize waste, Differential Density (DDT)* measurements allow the bulk uranium mass to be estimated. Using plant fingerprint data for the Pu / U ratio, the fissile Pu content can then be derived. However the physical composition of the oversize waste means that DDT measurements cannot be made for this waste. It was therefore decided to use Passive Neutron Coincidence Counting (PNCC) to measure the Pu content of each drum, this being the most important factor in terms of criticality assessment, for oversize waste. Plant fingerprint data for the ^{239}Pu / $^{240}Pu_{equiv}$, ratio or "R – value", then allows the ^{239}Pu mass to be calculated. This accounts for the contribution of all the spontaneously fissile transuranic nuclides as a function of fuel type and burnup history. For oversize waste this provides the only value for the Pu mass, while for undersize waste it will be a confirmatory measurement to support the findings of the DDT measurements.

Two identical PNCC assay chambers have been supplied to British Nuclear Fuels Engineering Ltd (BEL), for measurement of the $^{240}Pu_{equiv}$ content. The design, calibration, performance and testing of the systems have been described elsewhere (1).

Following an examination of the plant data relating to fuel residues in a particular waste stream (known as compartment 11 or "C11"), it became clear that some fuel may be present in the form of enriched uranium oxide (as opposed to the natural uranium Magnox fuel from which the waste in most streams derives). This meant that compliance with the Nirex repository drum fissile content limit could not be demonstrated by DDT / passive neutron assay techniques alone for these wastes. To overcome this problem it was decided to incorporate an active neutron interrogation system into one of the assay chambers, to allow the ^{235}U content to be assayed. The most cost effective, and practical type of active neutron interrogation system for the present purposes is a californium shuffler. It offers sufficient sensitivity and has minimal adverse engineering implications. The optimum chamber wall materials and detector configuration for a shuffler system are broadly compatible with those for a conventional PNCC system, so that the existing PNM chamber and detectors can be left intact with no modifications. A Differential Die-Away system, on the other hand, offers far greater sensitivity, but would be much more costly and would require considerable changes to the detector configuration and chamber design. The combined PNCC and shuffler system will provide the $^{240}Pu_{equiv}$ mass and the $^{235}U_{equiv}$ mass contents.

The C11 waste contains residues from the reprocessing of oxide fuels from various reactor types. As a consequence of the physical nature of the waste handling process, compacted pucks formed in the usual way would not be readily amenable to remote handling. For this reason, a physical bulking agent ($Mg(OH)_2$) will be mixed with the hulls to create pucks which can be handled properly, comprising a new, fixed matrix undersize waste stream. The physical properties of this bulking agent have been chosen, to facilitate the mechanical handling of the resulting pucks, as well as to ensure that the materials do not have an un-necessarily detrimental effect on the active

* The differential density technique uses the fact that fuel bearing material is of a higher density than the rest of the material in the waste. It uses the weight and the volume of each puck to calculate the puck density and from this the amount of fissile material present is estimated.

neutron measurements. The hulls will be mixed thoroughly with the bulking agent, ensuring both a homogeneous matrix, and a uniform radial distribution of fissile material within the pucks. The C11 undersize waste stream therefore essentially comprises a fixed, homogeneous matrix.

SHUFFLER PERFORMANCE REQUIREMENTS

The User Requirements Specification for the active/passive monitor (2) gave details of the required performance of the shuffler system with respect to drum throughput, Lower Limit of Detection (LLD) and Total Measurement Uncertainty (TMU). The maximum shuffler assay time per drum is 40 minutes, including the time for a standardisation measurement. A 3σ LLD of less than 2 g $^{235}U_{equiv}$, is required. The required TMU is 50% or less, quoted at the 3σ level (equivalent to a confidence level of 99.8%); the TMU value must be reported for each individual drum assayed.

The 3σ TMU, $3\sigma_T$, is defined as three times the quadrature sum of the random statistical counting precision, σ_R, and the systematic uncertainty, σ_S, on the measurement, as expressed by Equation 1.

$$3s_T = 3\sqrt{s_R^2 + s_S^2} \qquad \text{(Eq. 1).}$$

The systematic assay uncertainty arises from the residual variation (that is, the bias after any correction factors have been applied by the data analysis algorithms) of response with the position of the fissile material within a drum. This arises because of the inherent physics limitations of this assay technique. The position effects are minimised by ensuring that the equipment design is consistent with a flat axial response profile over the height of the drum. This is important because the fissile loading is likely to vary from one puck to the next. We assume both that the matrix is homogeneous, and that the radial distribution of fissile material is approximately uniform as a consequence of the C11 waste mixing process. These assumptions are important because the assay chamber has no turntable, meaning that rotation cannot be used to "even out" the effect on the response, of non – uniformities. From the nature of the C11 waste process in which the hulls are mixed thoroughly with the bulking agent, the assumption of radial uniformity for both the matrix and the fissile material distribution, is considered to be wholly justified. As we are dealing with a fixed matrix calibration, there is no systematic uncertainty component due to matrix effects alone. However, a matrix interrogation measurement is performed to check that the bulking agent has been prepared correctly.

The 3σ LLD is defined as the mass of material, which would give rise to a net signal which would be measured with a mean fractional standard deviation (precision) of 33.3%. For a ^{235}U mass equal to the LLD, the statistical precision contribution ($3\sigma_R$) to the TMU is therefore, by definition, 100%,. Thus for Pu masses close to the LLD, it is impossible to achieve a 3σ TMU of 50%. However as the ^{235}U mass increases, the statistical precision contribution to the TMU for a fixed assay time decreases. Thus for quantities of ^{235}U which are much greater than the LLD, the statistical precision contributes only a small amount to the overall TMU and can be neglected.

Maintaining a low TMU minimises the risk of false sentencing for drums close to the fissile criticality limit, with subsequent savings in respect of storage of the drums.

SHUFFLER SYSTEM DESIGN

The existing neutron counting chamber walls comprise two layers of high density polyethylene (HDPE) with a central cavity in which the ^{3}He proportional counters are mounted horizontally. The counters are spaced more closely towards the top and bottom of the drum (see Figure 1), in order to flatten the axial response profile. The chamber cavity is lined on its inner surface with a lead shield, to protect the detectors from the gamma ray pileup, which would otherwise result from the high gamma ray dose emanating from the ILW waste drums. A standard Matrix Interrogation Source (MIS – also known as add-a-source) system is included (1), to facilitate matrix compensation in the passive mode.

The shuffler system (see Figure 1) comprises a separate source store for the ^{252}Cf shuffler source, a flight tube assembly which passes centrally up the inner surface of one chamber wall, and a servo motor / gearbox assembly to control the motion of the source. The source store is constructed from alternate layers of HDPE and borated HDPE, the source home position being located within a lead block. It is located beneath the bottom of the assay chamber, below removable floor plates. The main role of the source store is to reduce the external dose rate to acceptable levels, but it also acts to minimise the background count rate in the chamber ^{3}He counters, from the ^{252}Cf source in its home position. The short section of flight tube between the source store and the assay chamber, is also surrounded by lead and HDPE to control the flash dose as the source travels between the store and the chamber.

The chamber does not include a turntable as it was originally designed as a near 4π PNCC. This inevitably leads to an asymmetry in the magnitude of the interrogating thermal neutron flux. The flux falls off rapidly as one moves across the drum diameter, away from an irradiation position. This effect leads to a large azimuthal variation of response, and means that the assumption of a uniform matrix and fissile material distribution across the drum, is important. In practice, the system is designed so that there are three separate, fixed irradiation positions within the chamber, spaced uniformly up the height of the drum. This means that the entire volume of the drum is interrogated. In addition, by analysing the responses from the 3 irradiation heights separately, one can compensate for axial non-uniformities in the fissile material distribution (that is, different total fissile loadings in different pucks). However, the analysis assumes both that the matrix is uniform within the drum, and that the fissile material distribution is uniform across the drum diameter, at a particular height. That is, we assume that fissile material may be considered to be present as a series of uniform discs (at different heights).

In a shuffler counting cycle the source is first driven rapidly from the source store to the first of the three fixed irradiation positions, where it irradiates the drum for a period of approximately 4 seconds. This is followed by a 1 second delay during which the source is rapidly withdrawn, after which delayed fission neutrons are counted in the ^{3}He detectors for a further period of approximately 4 seconds. This process is repeated cyclically, allowing an equilibrium distribution of delayed neutron precursors to establish. In practice, a complete shuffler assay will comprise a fixed number of irradiation cycles at each of the three positions, which will correspond to 1/6, 1/2 and 5/6 of the drum height. There will be approximately 30 cycles (of approximately 10 seconds each) at each position, making a total of 900 seconds. The measured count rate is then proportional to the "total fissile" mass, which will be expressed as a ^{235}U equivalent mass.

Sensors are included in the system to monitor the positions of the shuffler ^{252}Cf source. Optical sensors monitor the position of the tail end of the Teleflex^{TM} cable (to which the ^{252}Cf source is attached) and a small ^{3}He proportional counter ("source home monitor") linked to an alarm ratemeter provides an additional safety measure. If either of the sensors indicates that either the source is not stored safely in its source store, then the doors to the measurement area remain locked, preventing personnel access.

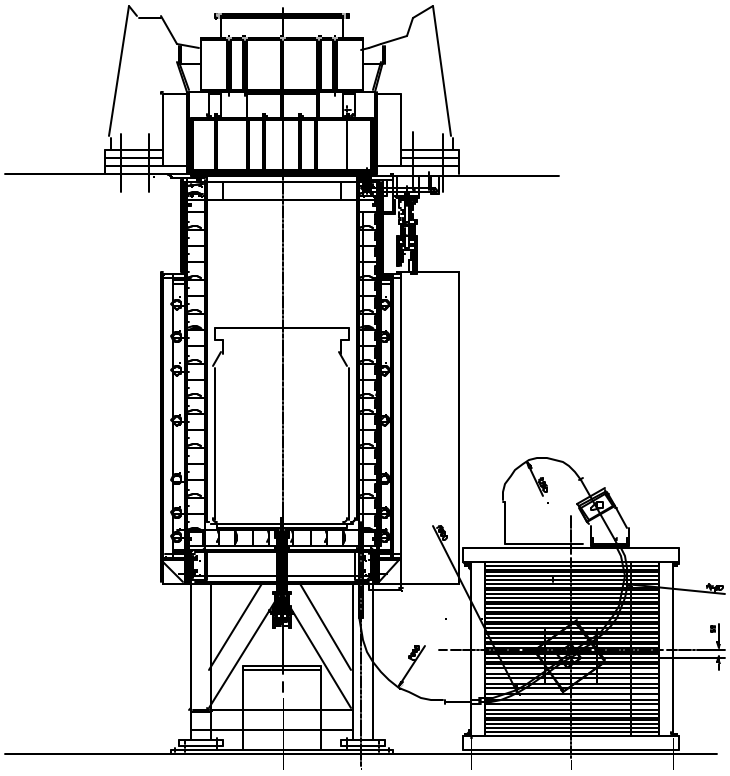

Fig. 1: Neutron counting chamber. The source store for the shuffler ^{252}Cf source is shown to the right of the main chamber, while the smaller MIS store is shown underneath the chamber. A 500 litre product drum is visible inside the chamber. The unevenly spaced horizontal ^{3}He counters are visible within the chamber walls.

The ^{252}Cf source strength was selected using a Monte Carlo model of the chamber using the MCNP(4B) code (3), coupled with a predictive code to calculate the LLD given the characteristic count rate per unit mass (from the MCNP modelling), the background count rate as measured during the PNM calibration and testing, and the known count time. The approach described in (4), was used to calculate the specific shuffler response (cnt/s/g dilute ^{235}U). Modelling a typical SDP undersize waste matrix, with a uniform distribution of ^{235}U, a source strength of 10^{8} n.s^{-1} was found to be adequate to achieve an LLD comfortably below the required 2 g ^{235}U level. A source of strength (when new) of approximately 2×10^{8} n.s^{-1} was therefore chosen, to allow for the decay of the source during its operational life.

CALIBRATION METHODOLOGY

Matrix Effects

In order to assist in defining a suitable reference calibration matrix, an MCNP study has been performed to investigate the effects of various materials, on the shuffler performance. This study can be used to assess a suitable calibration drum, which can be constructed from readily available inactive materials. The approach described in (4), was used to calculate the specific shuffler response (cnt/s/g dilute ^{235}U) at various radial, axial and azimuthal locations within 500 litre product drums filled with the different matrices. This simulated response data was then used as input for trial data analysis algorithms, in order to deduce the assay response under specific conditions. The drum radius was split conceptually into 4 azimuthal quadrants. This is necessary because the drum will not be rotated during the measurement, and irradiation will be performed only along a single chamber wall. It allows the fall-off of response on moving further away from the irradiation position, to be considered. A statistical analysis of these results then enabled the TMU values to be estimated. The specific response results, combined with the known chamber background count rate, also enabled the LLD to be predicted for each matrix.

The materials studied included $Mg(OH)_2$, $CaCO_3$ (Calcium Carbonate), and "typical" SDP undersize waste mixtures. $Mg(OH)_2$ material is identical in chemical composition to fully corroded Magnox swarf, which represents a large proportion of the SDP waste and has a similar bulk density to that of a typical undersize puck. Calcium carbonate is potentially useful as it contains no hydrogen and therefore has a modest effect on the shuffler response. Both of these materials are readily available and it would be easy to define a homogeneous, reproducible calibration matrix drum using either of these. For each matrix, the effect of absorbed moisture was determined by modelling the matrix both in dry form, and with 5% by weight of water added.

By analysing the responses from the 3 separate irradiation heights, it is possible to derive a compensation for a non-uniform axial distribution of fissile material. After determining the specific response from fissile material in each of the three height segments, from irradiation at the three separate heights, the specific response can be expressed as a 3 x 3 matrix. Expressing the measured shuffler response as a vector for the 3 drum segments, the fissile mass vector (for the three axial segments) can be obtained by matrix inversion. The total mass is then obtained by summation.

Alternatively, by averaging the count rates over the three irradiation heights, an uncompensated calibration can be made, assuming that the ^{235}U is distributed uniformly throughout the entire drum. This is known as a "Volume Weighted Average" (VWA) calibration. The VWA calibration parameter (cnt/s/g ^{235}U) is obtained by taking the weighted average of the results for the various positions in the drum, assigning a weight to each position proportional to the volume of the region, which it represents. For the present MCNP modelling, the drum is divided conceptually into 72 zones (6 axial segments, 3 radial segments and 4 azimuthal quadrants) into each of which a unit source is placed in order to estimate the statistical variation. These zones allow the spatial variability of response to be calculated in the axial, radial and azimuthal senses. The azimuthal response variation is of interest as the drum will not be rotated during the measurement period.

The LLD results are summarised in Table I. Clearly there is a comfortable safety margin against the required value of 2 g ^{235}U, for all materials considered here. A Totals background rate of 60 cnt/s was assumed, based on the ambient levels measured during testing of the passive assay systems at Harwell. In practice, the background at the SDP site is expected to be a factor of a few lower than this, due to the shielded location of the instruments at the SDP site. The Pu content of the fuel will elevate the effective background for the active measurement slightly. For example, the presence of 1 gram of $^{total}Pu$ (with 20% ^{240}Pu equivalent) with 1 gram of ^{235}U (a pessimistically high Pu/U ratio) will increase the background count rate by approximately 10-20 cps. However, after combining the effects on the background of the Pu neutron emission, with the additional shielding at SDP, the LLD figures quoted in Table I are still considered to be upper limits.

Table I. LLD results expected (g ^{235}U for a 900 second shuffler assay, for a 1×10^8 n.s^{-1} ^{252}Cf source) at Harwell, based on VWA calibrations. A totals background count rate of 60 counts per second is assumed.

$CaCO_3$		$Mg(OH)_2$		Undersize waste	
Dry	Wet	Dry	Wet	Dry	Wet
1.20	1.15	1.60	1.83	1.38	1.51

The spatial response variability results are given in Table II. Data is given for both the uncorrected (that is, VWA results) and axially corrected results. The benefit of the axial compensation is clear, as the standard deviation reduces significantly. For each matrix, the axially compensated value is comfortably below the required 1σ TMU value of 16.7%. Without axial compensation, the values are significantly greater, and in particular for the $CaCO_3$ matrix the value is considerably greater than 16.7%. Note that the effect of including radial variations, is to increase the σ value to approximately 25 – 50%, highlighting the importance of ensuring a homogeneous hulls – matrix mixing scheme.

Table II. Statistical analysis of the axial and radial response variability for the candidate matrices, derived from MCNP calculations. σ_{ax} includes axial variations only, while σ_{rad} also includes radial variations.

σ (%)		$CaCO_3$		$Mg(OH)_2$		Undersize waste	
		Dry	Wet	Dry	Wet	Dry	Wet
VWA calibration	σ_{ax}	23.5	21.1	9.8	8.0	14.1	12.2
	σ_{rad}	29.8	30.2	51.5	58.5	39.3	45.3
Axially corrected	σ_{ax}	11.7	12.0	8.0	6.5	9.1	8.3
	σ_{rad}	23.3	25.8	51.3	58.3	37.5	44.4

Effects of Fissile Material Lumps

Self multiplication gives rise to a tendency for a positive bias in the ^{240}Pu equivalent mass in passive neutron counting systems. When lumps of fissile material are assayed by active means, however, self shielding must also be considered. This leads to an underestimation of the fissile mass due to absorption of the interrogating flux in the outer layers, and is generally the dominant effect in active neutron waste assay systems.

The effect of self shielding on the overall TMU, has been investigated by using MCNP to calculate the values of the self shielding factor (SSF) (5) for specific bounding cases. The calculational method also takes into account any self multiplication in the sample, although the net effect is generally for SSF values less than unity and net underestimation. The self shielding factor is defined as the ratio of the reported to the true total fissile mass, if the calibration is based on dilute fissile material. In practice, the most extreme form of fissile material lumping, in the present application, is thought to arise from AGR fuel hull cropping operations, in which case a crimped end could give rise to a cylindrical lump of UO_2 (2.68% ^{235}U) of length up to approximately 50 mm, and diameter approximately 12 mm. The calculated SSF in this case corresponds to an underestimation of 26%. The effect of the matrix on this SSF value is negligible, to first order. If we take a calibration midway between SSF values of 1.0 and 0.74, and treat the two extremes as 3 sigma cases, then the worst case under and over estimation would each be 13% (1σ approx. 4.3%). This is considerably lower than the standard deviations from position effects (Table II) and so will have a small effect when added in quadrature to the Table II values, to obtain the overall TMU. The ^{239}Pu in irradiated fuel is dispersed throughout the fuel and therefore does not alter the SSF much due to the fact that as Pu builds up the ^{235}U content is depleted. Natural uranium and depleted uranium (which by definition is the form that uranium in spent fuel is in) is less important to the Post Closure criticality safety case of the eventual repository because it is self poisoning (whereas enriched uranium is not)

Planned Calibration Procedure

The MCNP calculations described above, showed that there is a modest difference between the specific shuffler count rates, for the various matrices considered. In the light of these results, it is planned to construct a calibration drum containing $Mg(OH)_2$. This material is readily available in the form of small spheres (dimensions of typically 3 mm), ensuring that a homogeneous bulk matrix can be obtained. MCNP calculations will later be used to determine a correction factor so that the real physical bulking material matrix can be cross – calibrated against the calibration drum. This correction factor is expected to amount to no more than 10 – 20%. This level of agreement provides confidence in the accuracy of the calibrations performed.

Measurements will be made using the $Mg(OH)_2$ calibration drum, using a set of 93% enriched uranium metal standards of known ^{235}U mass. The self shielding factor for each sample is known from MCNP modelling. This means that a correction can be made so that the calibration is applicable for dispersed ^{235}U (that is, with no self shielding). The samples are approximately 1.5 mm thick, and approximately 25 mm square. The ^{235}U effective mass of each sample is about 1.7 g. This is equal to the product of the true ^{235}U mass and the known self shielding factor, and is therefore represents the mass if the material were present in dispersed form. By combining up to 6

samples, a total effective ^{235}U mass of about 10 g can be achieved. This fissile mass, which is of the order of 10 times greater than the LLD, will ensure that good counting statistics are achieved for the calibration measurements.

A sample frame will be constructed, comprising 4 vertical tubes, placed at selected positions within the calibration drum (see Figure 2). Three equal area radial zones are thus defined, as shown (zones 1, 2 and 3), with a tube positioned at a point in each, which further bisects the zone into two equal areas. An additional tube is placed on the drum axis (0), in order to define a reference point, which will also coincide with the position of lowest response in the matrix. By measuring tubes 1, 2 and 3 at each of 4 azimuthal positions (NEAR, FAR, RHS and LHS[*] - achieved by rotating the drum) and taking the average over the resulting 12 readings, a spatially averaged average response can be obtained at a fixed measurement height. By taking measurements at different heights, the drum can be further divided into 3 axial segments, commensurate with the analysis algorithms described above. A bounding case analysis (that is, extreme axial positions for radially uniform material) can then be defined for use with the data analysis algorithm, in order to determine the 3σ TMU performance for the calibration matrix.

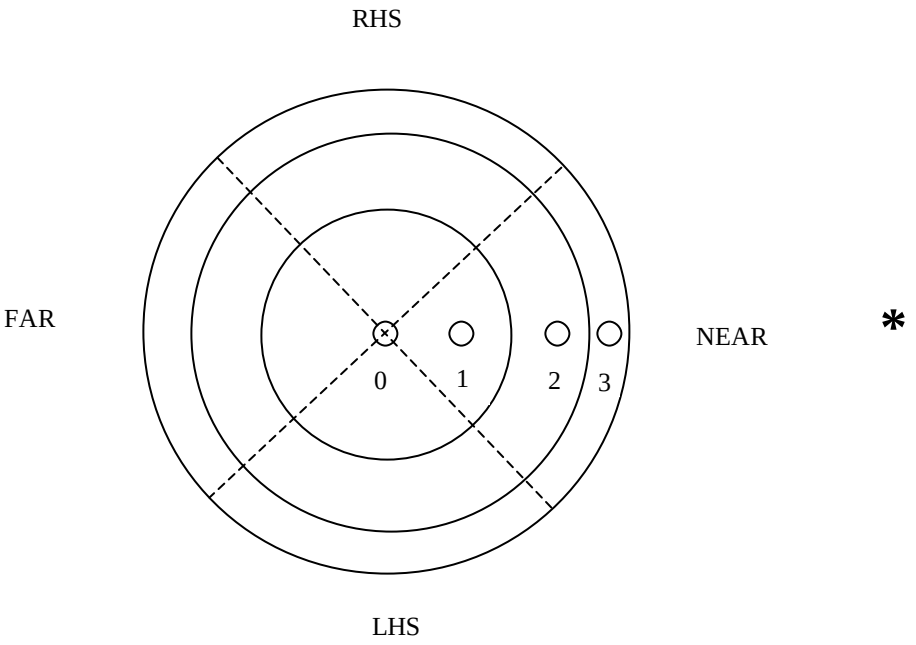

Figure 2: Locations of the 4 measurement tubes, which are used to measure the response in 3 radial zones of equal volume. The ^{252}Cf source position is marked (*), with the measurement tubes oriented parallel to the guide tube for the ^{252}Cf source

[*] Note that due to symmetry, the results for RHS and LHS should be essentially the same.

SYSTEM OPERATION AND SELF CHECKING

Operation of the system is normally carried out by the SDP plant control system. A passive measurement sequence consists of a standardisation measurement with an empty chamber, a full PNCC assay with the drum loaded (including an MIS sequence), followed by a second standardisation measurement. A standardisation measurement consists of an MIS measurement sequence with an empty chamber. This is intended to confirm the correct functionality of the system; the decay corrected count rates in each individual detector being checked for consistency, to within a specified tolerance.

An active measurement sequence comprises a full passive mode assay, followed by a shuffler interrogation. The passive assay gross Totals count rate is used as the background subtraction for the shuffler measurement. At the end of the shuffler measurement the fissile uranium mass is determined by subtracting the masses of the fissile Pu isotopes (determined from the PNCC measured ^{240}Pu equivalent mass combined with the known Pu isotopics) from the shuffler measured ^{235}U equivalent mass, using Equation 2 (obtained from reference 6).

$$m\left(^{235}U_{equiv}\right) = m\left(^{235}U\right) + 0.643m\left(^{239}Pu\right) + 1.907m\left(^{241}Pu\right) + 0.429m\left(^{233}U\right) \qquad \text{(Eq. 2).}$$

The detector count rates from an assay are checked in order to detect a malfunction. Checks are made that the count in each detector is both non zero, and less than a defined upper limit.

The system can also be operated in maintenance mode, in which various system maintenance functions can be selected, for example measurement of detector High Voltage plateaux, coincidence die-away time and predelay measurements, and adjusting the values of the various system parameters. The MIS and shuffler sources can be moved to specified positions, and counting can be initiated in each channel, for diagnostic purposes. Routine assay of a standard, check calibration drum, can also be initiated.

SUMMARY

Because significant quantities of enriched uranium are likely to be present in a particular waste stream at the SDP plant at Sellafield, the decision has been taken to fit a californium shuffler active neutron interrogation system to one of the existing PNCC chambers. This paper describes the system design, performance requirements and operating methodology for the shuffler. It also describes the planned calibration methodology, showing how the required performance will be demonstrated.

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