Preparation of Waste Fingerprints for the Miscellaneous Beta Gamma Waste Feeds to the Box Encapsulation Plant at Sellafield – 16080

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ABSTRACT

There are a number of legacy pond and silo storage facilities on Sellafield Site that contain a broad range of ILW arising from the early Windscale Pile and Magnox Fuel Reprocessing Programmes. The waste comprises of solid Miscellaneous Beta Gamma Waste (MBGW) from operations (including filters, containerised waste, thermocouple wire, support struts, pumps, soft wastes) and associated sludges that have arisen from the corrosion of the fuel, Magnox swarf cladding and the MBGW. It is intended to retrieve and package this waste to meet the requirements of interim storage and final disposal in the Box Encapsulation Plant (BEP) at Sellafield. As part of this process each package must be assigned a realistic and justifiable radionuclide and material inventory. These inventories can either be assigned by radiometric measurements including the associated errors or developed by fundamental data underpinning these assessments and a “chain of custody” approach of knowing where the waste has been and how it has evolved. The subject of this paper is concerned with the development of the waste fingerprints for these legacy waste streams.

As the wastes have arisen over a ~70 year period there have been an assortment of different working practices and developments in equipment design that have led to significant variations in the waste condition and its composition. During storage the waste items have become mixed, corroded and sludge deposits have arisen. Because of this, and that early consignment records are either incomplete or missing, the task of assigning material and radionuclide fingerprints has been very challenging. In addition, to comply with regulatory requirements the inventory assignment has to be sufficiently robust to meet the processing envelope of the encapsulation process i.e. to allow controls to be made on the amounts of reactive materials (e.g. aluminium, Magnox and uranium) that can be processed within each product and allow identification of excluded items from being consigned from the donor plants. Also, as the waste comprises actinides, fission and activation products it was noted that whilst the UK standard inventory code FISPIN10 predicts the actinides and fission products well, there was a significant discrepancy in the prediction of activation products when compared with those predicted by the specialised fusion code FISPACT. It should be noted that the use of FISPACT to predict actinides and fission products is not recommended.

To address these issues,

- position statements were produced for each of the waste streams that would be consigned to BEP;
- gaps within the data were assessed and where possible inactive trial work was identified to address it;
- material and radionuclide fingerprint justification documents were developed from the position statements based upon available data, the experimental work and justified assumptions;
- where FISPIN runs were required, these were run with a modified activation data library based on the European Activation File 2010 (EAF2010) to model the activation products.

This paper summarises the BEP process, discusses examples of how the material and radionuclide waste stream fingerprints have been justified by revisiting the sample data, original consignment records, engineering drawings and recent remotely operated vehicle (ROV) pond surveys and making reasoned assumptions based on operational practices and non-active experimental work. Incorporation of the EAF2010 library within the FISPIN code has offered significant benefits to BEP and indeed other plants processing mixed fission and activated wastes as it allows comparable predictions of activation products compared with that made by FISPACT and for the plant to maintain one data processing stream rather than two if it had to utilise both modelling codes.

This work will underpin the inventory submission in support of the Interim Letter of Compliance (ILoC) to Radioactive Waste Management Limited (RWM). The process of examining the original records, plant operation schedules and engineering drawings has enabled removal of some of the pessimisms that have been made in previous assessments.

INTRODUCTION

Sellafield Ltd (SL) is pursing the packaging of legacy wastes on the Sellafield site by utilising Box Encapsulation Plant (BEP) to package these wastes into grouted BEP $3m^3$ boxes for disposal. Numerous types of waste and materials constitute solid Miscellaneous Beta Gamma Waste (MBGW) and SL needs to confirm that the BEP process is acceptable for all the identified wastes. Some of the waste materials present particular challenges in terms of meeting Radioactive Waste Management Limited (RWM) requirements for the completed waste packages, which will need to be resolved before a Letter of Compliance can be obtained.

One of these challenges is the provision of a realistic and justifiable inventory for the each waste package produced. Such data generation and recording needs to consider both the expectations of Radioactive Waste Management Ltd (RWM)(the company tasked with developing the geological disposal concept in the UK) and the practicability of assigning physical and radionuclide inventories to individual waste packages, which will vary between waste types [a].

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[a] The term waste “type” as used here may apply to a single design of item, a particular material or a collection of items/materials (e.g. MBGW).

Sellafield's operational requirements, including safety case and Safeguards supporting data, also needs to be taken into consideration and developed in parallel.

This paper summarises the BEP process and the nature of the waste to be processed through BEP, discusses the strategic approach to the development of inventories for the waste and provides examples of how the material and radionuclide waste stream fingerprints have been justified by revisiting the sample data, original consignment records, engineering drawings and recent remotely operated vehicle (ROV) pond surveys and making reasoned assumptions based on operational practices and non-active experimental work.

The incorporation of the EAF2010 library within the FISPIN neutronics modelling code has offered significant benefits to BEP, and indeed other plants processing mixed fission and activated wastes, as it allows comparable predictions of activation products to be made compared with that made by the FISPACT neutronics modelling code and for the plant to maintain one data processing stream rather than two if it had to utilise both modelling codes.

THE BEP PROCESS

BEP will be a solids encapsulation plant, designed to produce a variety of mixed MBGW packages with a minimal amount of pre-treatment. Sludge may be associated with the wastes to be packaged at BEP; therefore, the process is being designed to tolerate its presence. The process will allow for handling of wastes and some simple pre-treatment operations. However, the overall design philosophy is to enable generation of suitable packages with the minimum amount of handling.

In general, waste will be imported to BEP from the donor facilities in pond skips or $3m^3$ box liners. The transfer pond skip will be placed into a box liner in the buffer position of the Waste Treatment Cell (WTC) of BEP prior to being moved to the unloading position next to the Waste Handling Table (Fig. 1).

The waste to be processed through BEP falls in to three broad categories:

- Segregated wastes: comprising largely quantified waste items as received at BEP which will be consigned directly in their entirety to liners;
- Identified wastes: comprising any and all specific waste types of interest that will be quantified where practicable during operations at BEP; and
- Remaining mixed waste: comprising all other items that do not require, or cannot be, specifically quantified during operations within BEP.

Segregated waste types will not be examined at BEP prior to encapsulation with reliance placed on the donor facilities to provide information on the radionuclide and physical/chemical composition of the waste. Identification or quantification of wastes to maintain packaging limits or application of any required additional treatment may be carried out on the Waste Handling Table (Fig. 1). The remaining mixed waste is that which will not require additional treatment or does not pose special inventory considerations for packaging.

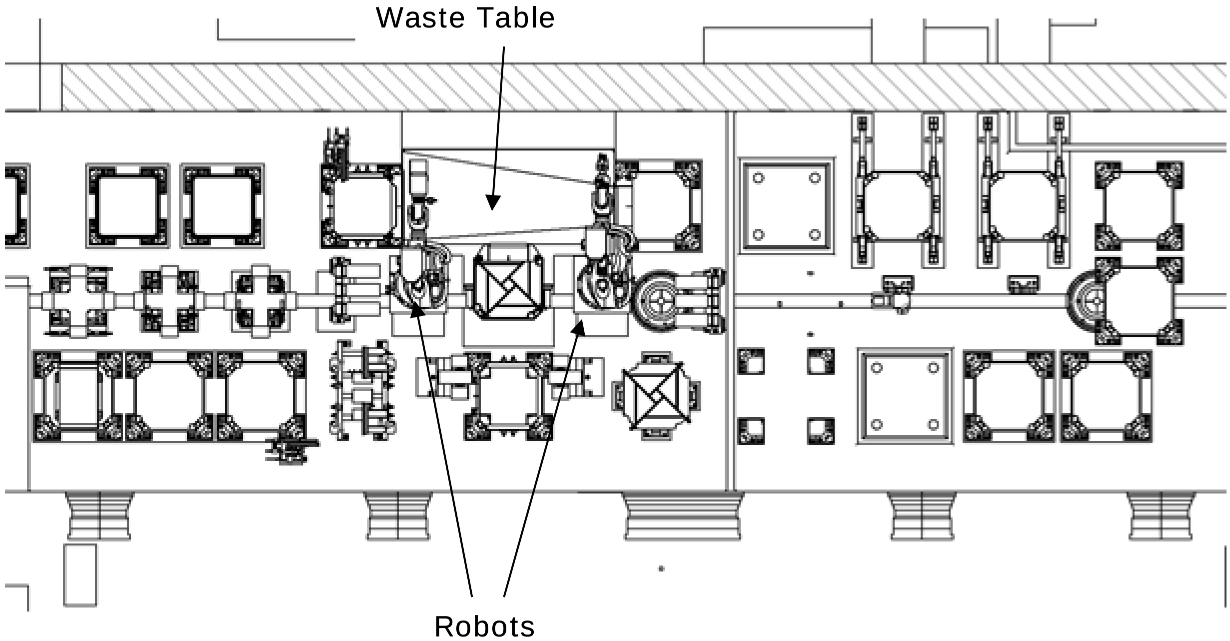

Fig. 1. Plan View of the Waste Treatment Cell of BEP

The contents of a single pond skip will be packed into one 3m^3 box liner. It is the intention that skips will have undergone some form of pre-sorting at the donor plant. Skips that are suitable for direct encapsulation will be visually inspected on arrival at BEP to ensure that they match with the consignment records and confirm that they are suitable for grouting. In general, the skip will not be unpacked and will be loaded directly into a 3m^3 box liner in the WTC.

Where it has not been possible to pre-sort this waste, the contents of the skip will be removed by robots and placed into a 3m^3 box liner, taking account of any material or item content limits for packages. Where appropriate, waste will be placed on the Waste Handling Table (Fig. 1) to facilitate any additional treatment and liner filling. The process aim will be to pack at least two skips into a single liner.

Any sludge and liquor present with the waste, or generated during handling operations, will be collected into a settling liner. This will be a standard 3m^3 box liner positioned at a station next to the table (Fig. 1). Irrigation of the table will direct mobile sludge and liquor into this liner where sludge will be allowed to settle for a period of time before the supernatant liquid will be decanted off. It is currently assumed that the settling liner would have a maximum of 160 litres sludge when full with 2,100 litres liquor. Once the supernatant has been decanted, the liner, with a heel of sludge, will be used as the next fill liner to receive wastes. The settling liner may also have a nominally emptied pond skip present. When the fill liner is deemed full, an anti-flotation plate (AFP) will be fitted to the 3m^3 box liner in most cases, before the liner is moved to the grouting station.

The liner will be flood grouted from the top using a 5:1 ratio of ground granulated blast furnace slag (GGBS) and Portland cement (CEM I[b]). During interim storage, the annulus between the liner and the 3 m^3 box walls will be left un-grouted allowing a certain degree of expansion due to corrosion to be accommodated by the waste package. This allows greater flexibility in the choice of fill material to maximise the performance required at the time of transport and disposal.

GENERAL NATURE OF THE WASTE

The majority of the BEP solid ILW feeds are described as MBGW, a term that encompasses a wide variety of materials arising predominantly arising from the Windscale Piles and Magnox reprocessing programmes. This includes fuel furniture, irradiated components from reactors, failed mechanical items from different plants and cans/bags of operational and PIE wastes from a number of facilities on the Sellafield site. These wastes have historically been placed within a number of wet and dry stores, from where they will be retrieved for packaging in BEP.

Although described as “beta/gamma” wastes, i.e. requiring remote handling, alpha activity may also be present, particularly in the form of fuel-derived contamination or discrete fuel pieces. The waste items may be activated, i.e. irradiated in-reactor, or simply contaminated.

The items vary in dimension from material only a few inches long, to large/long items that will require size reduction to enable transfer to BEP. The average composition of the MBGW varies, but is typically dominated by stainless steel and mild steel metallic items.

Some additional discrete challenging waste types may also be packaged through BEP, and the data recording methodologies have been developed to encompass the full range of waste types that are expected to be encountered. These waste types are:

- Ionsiv Cartridges; cartridges of ion exchange material used for removing activity from pond water.
- Zeolite skips, ion exchange material used for removing activity from storage ponds water.
- Zircaloy and stainless steel fuel hulls (opportunistic waste feed).
- Isotope cartridges of various designs from the ponds, which will be sorted prior to consignment to BEP.
- Bulk Magnox swarf and aluminium doughnuts (opportunistic waste feed).
- Dependent on the effluent system design – any settled sludge may be managed with other wastes, the nature of which will vary significantly between consigning plants.

[b] CEM I (Cement I) was formerly known as Ordinary Portland Cement (OPC). It is manufactured to conform to BS197-1

- Tokai Mura End Crops, both cemented and uncemented (opportunistic waste feed).
- Bins of uranium pieces known as U-bits, both cemented and uncemented (opportunistic waste feed).

DATA RECORDING PROCESS

The BEP approach to data recording for wastes and waste packages is based upon the detailed assessment of underpinning data and a "chain of custody" approach of knowing where the waste has been and how it has evolved over time.

The processes adopted in the plant to deliver this approach include a combination of sorting (as being able to be performed at the donor plants), segregation, visual inspection, box packing controls, mass, assay and scaling of fingerprints (as performed at both the donor plants and BEP). The process is described in more detail in the bullet points below:

- Sorting – The waste will be sorted either within the donor plants or at BEP so individual items that need pre-treatment or those whose quantity is limited in a box can be segregated. This will also assist the estimation of material volumes i.e. quantity of steel, graphite, soft wastes etc.
- Segregation – Waste items or materials requiring further investigation, or those whose content is limited in a box, will be segregated. The quantities of each controlled item per box will be recorded and form part of the package records. In the context of BEP, segregation refers only to the transfer of the waste item to a region of the table to undertake any necessary processing or for ensuring box packing controls e.g. distribution of items across a number of packages. The waste item will ultimately be transferred into a container for grouting.
- Visual inspection – As the waste is added to the skips within the donor facilities, upon receipt at BEP or as waste is removed and transferred into liners at BEP the operators will use visual inspection to crudely estimate the volume percent of the different materials in order to calculate the box total (although it has to be recognised this may be somewhat subjective). The operator will also record the actual number of items with specified controls added to a liner e.g. numbers of filters or grabs worth of swarf. Each liner will have a unique number.
- Box packing controls – The operator will control the quantity of any controlled items added to a liner e.g. reactive metal quantities. These will be recorded on the box package records.
- Mass – The mass of the waste (as measured by the cell handlers) will be used in combination with the volume estimate of the material components, any records of the waste and a pre-determined relative density for these items to determine the mass of the different component/material types in a basket. This operation is performed by the technical support team and waste tracking system.

- Gamma measurements – Gamma dose measurements or spectrometry performed at the donor plants may be used to determine and/or confirm inventory assignments.
- Radionuclide/Materials Fingerprints – These fingerprints will be applied to the calculated masses of the materials or measured gamma dose rates (at the donor plants) to calculate the radionuclide and physical/chemical inventories of individual packages to confirm compliance with the applicable limits.

Radionuclide / Materials Fingerprint Development

Key to the delivery of the approach detailed above is the development of the radionuclide and materials fingerprints. This is achieved through the preparation of Fingerprint Justification Documents (FJDs). The FJDs captures in one document the material and radionuclide fingerprints for use in the implementation of data recording on BEP and the data/assumptions upon which these fingerprints are based.

A review of the historic inventory information has been undertaken for waste originating from the donor plants where wastes are to be routed to BEP for treatment. These detailed reviews of waste records and radionuclide inventories have supported the development of the understanding the physical, radiological and chemical inventories of the wastes. This information has been captured in a suite of Position Statements.

The information in the Position Statements has then been used as the underpinning data to develop the fingerprint justification documents for the individual waste streams. As part of this fingerprint development work a gap analysis has been performed to identify what information and fingerprints are available and either justifying why those were adequate to represent the wastes under consideration, or further work has been carried out (e.g. FISPIN/FISPACT modelling) to provide a justifiable set of fingerprints.

Early work on the neutronics modelling of fingerprints has identified a need to develop a new data library for use with the UK standard inventory code FISPIN10 as significant discrepancies have been identified in the prediction of activation products when compared with those predicted by the specialised fusion code FISPACT. Therefore, where FISPIN runs were required, these were run with a modified activation data library based on the European Activation File 2010 (EAF2010) to model the activation products and this is discussed further below. Further information on the development of these fingerprints is provided in this paper.

Radionuclide Fingerprint Modelling

The radionuclide inventory in ILW waste processes are dominated by heavy nuclides (e.g. Pu, Am etc.) and fission products and the fingerprint predictions of these are well characterised by the FISPIN10 computer code [1] which has an extensive validation database of these against radiochemical measurements. However, BEP is primarily processing MBGW and as a consequence certain activated waste items can be significant contributors to the radionuclide inventory of individual packages e.g. thermocouple wires and cladding materials.

One drawback of using the FISPIN10 code is that the database it uses for activation calculations is not particularly extensive. This can be seen when the FISPIN10 predictions of the nuclides in the RWM list of 112 significant radionuclides [2] are compared with those of the specialised activation computer code FISPACT [3] for the same neutron spectrum. The FISPIN10/FISPACT ratios of a significant number of nuclides deviate from unity by factors greater than 2 or less than 0.5.

FISPACT is a modification of FISPIN primarily used in fusion applications to calculate the activation of materials in the wall of a fusion reactor and as such uses a very much larger database of activation product cross-sections. This database, EAF2010 [4], is the most complete database of reactions leading to activation currently available and was the result of over 20 man-years of effort. Since FISPACT is not recommended for fission product calculations [3] and rather than using FISPIN10 for fission products and FISPACT for activation products, a more cost effective solution has been adopted by incorporating relevant EAF2010 data as the activation database for FISPIN10.

A re-calculation of the activation products in the RWM 112 significant radionuclide list compared with FISPACT then showed agreement to less than 3%, see Fig. 2.

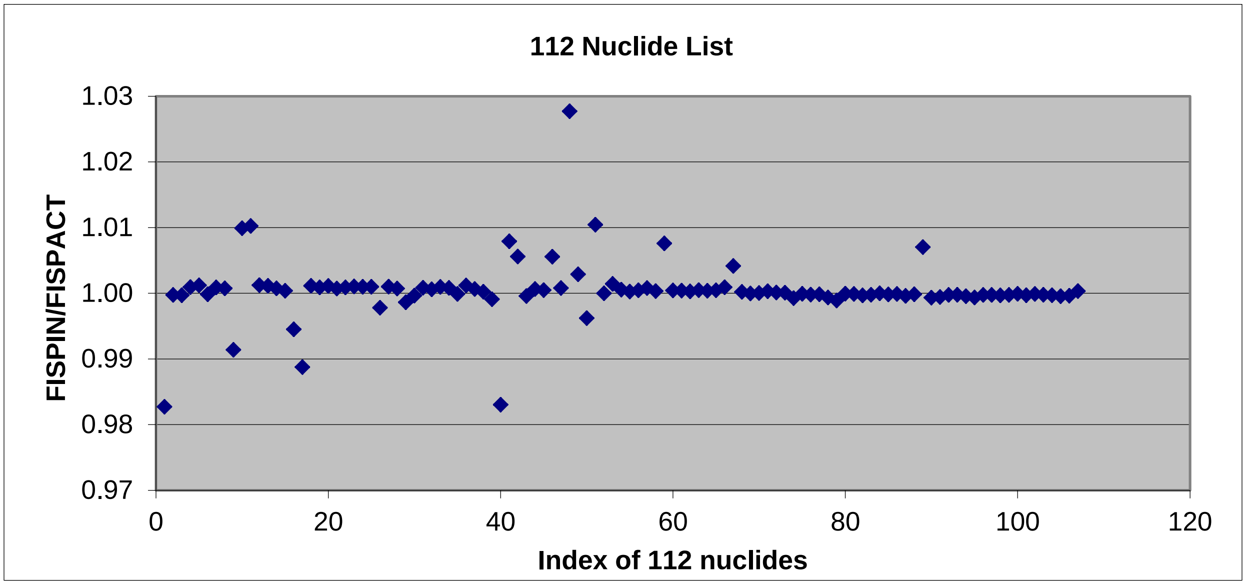

Fig. 2. FISPIN FISPACT Comparison for the 112 Nuclide List

EXAMPLES OF FINGERPRINT DEVELOPMENT

The following sections provide some examples of how the radionuclide and materials fingerprints have been implemented for specific waste streams. Due to sensitivity of these materials some details have had to be omitted.

Development of the Fingerprints for Tokai Mura End Crops

The Tokai Mura Magnox nuclear power station was Japan's first nuclear power plant. Housing two reactors built to the British Magnox design, initial criticality was achieved on November 1965; it ceased operation in March 1998.

Tokai Mura End Crops (TMECs) are a waste product resulting from the reprocessing of Tokai Mura spent fuel at Sellafield. Tokai Mura fuel is a modified form of Magnox fuel, being a hollow uranium bar fitted with zirconium end plugs with Magnox and Sintox (alumina). The end plugs are incompatible with Magnox reprocessing as they are insoluble in nitric acid and this meant that they had to be removed by "cropping" the fuel prior to decanning. The cropped fuel was then decanned and reprocessed and the TMECs that were generated were placed into pond storage whilst awaiting final treatment.

Historically, two methods of cropping Tokai Mura fuel elements were implemented at different times, i.e. Type A crops resulted from decanning prior to cropping that was used until about 1975 and Type B involving cropping prior to decanning that was adopted from about 1975. Taking the Type B endcrops as an example, the two end plugs or TMECs of the fuel assembly are different, as shown in Fig. 3 and are described as "saddles" (hook and eye) or as "lifting rings".

Fig. 3. Type B Tokai Mura End Crops with End Fittings in Place (Saddle, left and Lifting Ring, right)

End cropping took place underwater with the TMECs being collected in baskets. The baskets were placed into a skip within a container where they were caustic dosed to high pH and then ullaged in order to isolate the container from the bulk pond water.

To assess the inventory and condition of these, the BEP project have considered:

- Inventory data including quantities, burn-up and storage consignment dates supplied by Nuclear Materials Accountancy Safeguards (NMAS).
- Technical reports written at the time of processing giving details of the process and material compositions.

- Engineering drawings to calculate mass and surface areas of Magnox and uranium. These are particularly important as these metals will react with the grout and limits on the number of these items will need to be defined.
- Current condition and uranium carryover based on visual examination and assessment of uranium and Magnox corrosion under storage conditions.
- Material compositions of components, burn-ups, cooling periods and physical characteristics of the TMECs have been used within the FISPIN model to predict the radionuclide inventory (RNI).
- 5% of rods had to be re-cropped (i.e. generation of one re-crop per rod, occasionally the zirconium end cap was not removed by the first cropping operation and therefore a further 50 mm length of bar had to be cropped, these were known as re-crops) which resulted in a greater quantity of uranium carryover. Based upon examination of the operating instructions, these ranged from 50 to 57 mm in length.
- Based upon observed TMEC size distributions and re-crops, average masses were calculated to allow for 100% standard crops and 2.5% re-crops.
- As the TMECs were isolated from the main pond, any sludge deposits that maybe present are assumed to have arisen from the corrosion of the TMECs themselves and that the combined RNI of the TMECs and sludge deposits would be the same as the initial inventory.

Using this approach it has been possible to evaluate the size distribution, number of TMECs per basket, uranium and Magnox carryover, their current condition and radionuclide inventory for each basket of TMECs. The calculated data from these assessments are consistent with plant observations and operational experience. The detailed estimates of uranium and Magnox carryover based upon the corrosion studies also allow better understanding of the amounts that can be encapsulated within each product.

Application of Inventory Strategy to Isotope Cartridges

Isotope cartridges essentially comprise a range of materials that, when subject to neutron irradiation, produced specific radionuclides. For irradiation, the source materials were typically clad in aluminium or Magnox “cans”. Post-irradiation, the cladding was normally removed and the nuclide extracted from the source material. Some of the product nuclides are of particular concern with regard to disposability, e.g. C-14, and require exclusion from or limiting in the BEP waste packages. As a minimum, it is desirable to identify and assign a specific radionuclide inventory to any cartridges (other than those where the product nuclides were all short-lived) as this will be significantly different from typical MBGW.

The vast majority of the isotope cartridges arose from irradiation in either the Windscale Pile reactor or the Calder Hall/Chapelcross reactors, with a small number potentially arising from the British Experimental Pile 0 (BEP0). In general, the cartridge design used in each of the reactor types was different, enabling the reactor type to be easily distinguished from one another during retrieval operations in the ponds or processing in BEP. Distinguishing between the different cartridge types is potentially more complicated. Pile isotope cartridges had etching codes entered onto the end of the aluminium outer can plus a banding code applied on the can (although this did fade under irradiation). Generally discharges from the reactors were carried out on a campaign basis for different cartridge types, and skip content records management.

The Calder Hall, Chapelcross, Windscale and BEP0 reactors irradiated numerous different isotopes for various purposes e.g. cobalt to produce Co-60 and aluminium nitride (AlN) to produce C-14 for industrial and medical purposes. It is intended to use a combination of visual identification, low resolution gamma spectroscopy, weighing and measurement to sort and identify the various different types.

Visual: Assuming the cartridges are degraded to such an extent that no external features or markings can be discerned, visual identification will be limited to the length of the cartridges. However, this is enough to enable separation of the Calder Hall cartridges from the Windscale Pile cartridges.

Low Resolution Gamma Spectroscopy: To segregate cobalt containing cartridges. This can be achieved within the storage pond and negates the need to transport cobalt cartridges to a sorting facility.

Weighing, Measurement and X-Radiography: Is primarily used to distinguish AlN cartridges as these are excluded from the BEP process as the C-14 content may migrate into the biosphere during the Geological Disposal Facility (GDF) post closure period as methane.

RNI for each of these cartridges has then been derived by examining the process log books, design specifications, material compositions and using FISPIN data as appropriate. For example, Thorium Cartridges which were used as a flux flattener in the core, but also produced small amounts of U-233 for reactor research purposes. The RNI for this cartridge was derived by considering the following:

- Operation and Inspection and Inspection Requirements for Thorium Cartridges giving details of construction and design.
- Impurities in Thorium 1958.
- The location of where they were placed, the flux in that region and irradiation time within the reactor were acquired from the Pile Process Log Book.

Advantages / Disadvantages of the Planned Approach to Data Recording

The BEP planned approach to data recording is a simple and fit for purpose method by which waste package inventories assignment can be performed for such a chemically and physically diverse range of historical waste items.

Work undertaken by BEP has demonstrated that the use of assay equipment such as High Resolution Gamma Spectrometry and Active/Passive Neutron counting systems would require the waste to be size reduced and placed within standard assay drums. This would have a significant impact on throughput. The number of individual radionuclides that could be measured and the uncertainties associated with these measurements means that the approach using fingerprints and mass will give an inventory for the waste packages which is sufficiently accurate to meet the needs of BEP and RWM.

The approach of using historical information to develop fingerprints for the waste and converting the fingerprint into relative masses for each of the inventory radionuclides has a number of advantages and disadvantages. These are summarised below.

Advantages:

- The identification of the historical information and development of the fingerprints can be undertaken in advance of waste processing operations;
- Can be used for the large variety of heterogeneous wastes to be processed in BEP;
- Can be used to provide both radiological and physical/chemical characterisation data;
- No impact on throughput;
- Can be done off-line;
- No maintenance requirements;
- Used as part of the Quality Assurance (QA) and waste acceptance process for receiving waste into BEP;
- Produces inventories with sufficient accuracy to demonstrate compliance with limits and RWM requirements.

Disadvantages:

- Significant upfront work has to be undertaken to review historical records;
- Requires a clear strategy for data recording from the early stages of inception of the project;
- Requires the Waste Compliance Team at BEP to continually assess and confirm data requirements.

Applicability:

This process is planned to be implemented at BEP. It is simple and inexpensive and has no detrimental impact on throughput whilst producing data of sufficient accuracy to demonstrate compliance with limits and meet RWM requirements.

CONCLUSION

Using historical information to develop fingerprints for the waste and converting the fingerprint (with inventory confirmation from donor plant inspection surveys combined with washing, sorting and segregation) via mass measurements to provide waste package inventories is the most effective method by which inventory assignment for the broad range of MBGW and identified waste items can be performed. The functionality within BEP allows identified items of interest to be counted as they are placed within the liner and the relative masses of these items and their fingerprints can be applied.

Incorporation of EAF2010, (the most complete database of reactions leading to activation currently available) as the activation database for FISPIN10 has enabled calculation of the activation products of the NIREX-112 list to be within 3% of that calculated by FISPACT.

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ACKNOWLEDGEMENTS

The authors would like to thank Sellafield Ltd. and the Nuclear Decommissioning Authority for funding this work.