Radioactive Waste Management

# Ponds Solids Legacy Isotope Cartridges: Encapsulation, CVD and pre-treatment options (Conceptual stage)

## Summary of Assessment Report

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### *Background*

Isotope cartridge wastes originating from the Windscale Pile and Calder Hall reactors are currently stored in the First Generation Magnox Storage Pond (FGMSP) and the Pile Fuel Storage Pond (PFSP) at Sellafield, facilities of deteriorating infrastructure representing some of the highest hazards in the UK nuclear industry liabilities portfolio. Other wastes held in these facilities are not considered here.

Isotope cartridges consist of a target material which when irradiated produces the isotope of interest surrounded cladding.

The isotope cartridge waste is currently stored in skips in the open ponds. It is likely to be in variable condition due to corrosion over time, and to be associated with sludge (arising from corrosion of Magnox swarf, legacy fuel and steel, together with biological matter, sand, etc.). There may be items of beta gamma waste or fuel mixed in some skips, which would need to be identified and removed prior to processing.

Sellafield Ltd held a technology screening workshop in 2011, from which they selected processing options for conditioning the isotope cartridge wastes, namely encapsulation (using inorganic cement or organic polymer) and cold vacuum drying (CVD) as described in this Assessment Report.

In a separate assessment completed in 2013, proposals for conditioning of the isotope cartridge wastes with other Miscellaneous Beta Gamma Waste (MBGW) through the Sellafield Box Encapsulation Plant (BEP) gained Conceptual stage endorsement with the exclusion of aluminium nitride (AlN) cartridges.

### *RWMD Basis for Assessment and Endorsement*

This conceptual stage assessment has considered the compatibility of the proposed packages with RWMD requirements for safe long-term management, including storage, transport, emplacement and potentially extended storage underground in a geeolical disposal facility, and geological disposal. The reference basis for this assessment is the generic Disposal System Safety Case (DSSC) and the requirements placed on waste packages by the level 2 Generic Specification for low heat-generating wastes. Further information on the disposability assessment process is available elsewhere.

### *Scope of the Assessment*

This Assessment Report is based on information submitted by Sellafield Ltd in a proposal for conditioning isotope cartridge wastes using encapsulation and Cold Vacuum Drying (CVD) techniques. The proposal identified that aluminium nitride (AlN), lithium/magnesium alloy (AM) and potassium chloride (KCl) cartridges may have properties resulting in unacceptable safety issues if not appropriately conditioned. The Sellafield Ltd proposal contained methods for identifying cartridges by type allowing separation and pre-treatment of such cartridges. Pre-treatment by placing cartridges in long-lived containment vessels or chemical conversion of carbon-14 in AlN cartridges to low solubility carbonate were proposed. The options for pre-treatment and conditioning were assessed separately, with separate disposability conclusions presented here.

Issues for consideration included:

- the CVD treatment method is novel, uses a non standard waste container and so may result in disposal concept compatibility issues;
- magnesium phosphate cement is proposed as a potential encapsulant and magnesium phosphate cement wasteforms have not previously been endorsed for disposal; and
- the carbon-14 inventory associated with AlN cartridges is of potential environmental safety significance in the post closure phase following GDF closure with Sellafield Ltd taking a novel approach to management of this issue.

On this basis a Nuclear Safety and Environmental Category B was assigned to this proposal. The assessment findings were therefore presented to RWMD's Nuclear Safety and Environment Committee (NSEC). This report incorporates the views of this Committee.

### *Waste Packaging Proposals*

Several proposals were assessed for conditioning the isotope cartridge wastes including 3 encapsulation options and a cold vacuum drying option. Two pre-treatment options were also assessed.

### *Nature of the waste*

Although the precise number is not known, Sellafield Ltd estimate there is $13m^3$ or 6,600 kg, of raw waste comprising 1500-1800 cartridges, including about 2,000kg to 2,400 kg total target material mass. 8 cartridge types are present, dominated by cobalt and bismuth oxide (LM cartridges):

- 842 to1122 cobalt cartridges
- 493 to 498 LM (bismuth oxide) cartridges
- 70 AlN (aluminium nitride) cartridges
- 55 AM (lithium/magnesium alloy) cartridges
- 14 KCl (potassium chloride) cartridges,
- 6 to17 Sb/Be (antimony/beryllium) cartridges
- 6 $Tm_2O_3$ (thulium oxide) cartridges
- 6 Th (thorium metal) cartridges

In addition to the isotope cartridge target material about 70 kg aluminium cladding is associated with Windscale Pile cartridges and approximately 3660 kg Magnox cladding with Calder Hall cartridges.

### *Initial processing of the waste*

The first stage of the process involves deluge washing of the pond skips at the donor facilities to remove sludge and facilitate pre-treatment and processing.

Following this washing step the contents would be inspected and cartridges identified by type. The proposal details the features of the cartridges to be used for visual identification and physical properties to be used such as dimensions, mass, radiation emitted and internal structure as identified by X radiography. Decision trees are presented in the proposal which demonstrate the Sellafield Ltd logic behind cartridge identification by type. Isotope cartridges would be separated to allow pre-treatments, and compliance with any package loading limits and exclusions.

### *Encapsulation processes*

Packaging would occur at a Sellafield solids treatment plant, such as the proposed Box Encapsulation Plant (BEP).

Three types of encapsulant were proposed: firstly a formulation containing ordinary Portland cement with ground granulated blast furnace slag (referred to as OPC grout); secondly a formulation with magnesium oxide, potassium dihydrogen phosphate and pulverised fly ash producing a magnesium phosphate grout (referred to as MPC grout) and; thirdly an organic epoxy resin polymer.

Washed sorted cartridges would be imported to the plant in a pond skip. The skip and waste would be placed into a 8mm walled stainless steel box liner and the chosen encapsulant applied. After curing a capping layer could be added. The liner would then be placed into a 3 cubic metre stainless steel box which would be lidded and decontaminated. The annulus would remain unfilled to accommodate the potential for wasteform expansion. The encapsulation proposals are expected to result in 20 x 3$m^3$ box packages.

Following a period of storage in the Box Encapsulation Plant Product Store, during which time any expansive corrosion would result in wasteform evolution, and before transport the remaining space between the liner and the box would be filled with encapsulant, creating a clean protective annulus layer.

### *Cold Vacuum Drying*

Packaging of the isotope cartridges would occur at a CVD plant associated with fuel drying operations from the Sellafield ponds.

The proposed technique is based on the method used by the United States Department of Energy at Hanford to treat fuel wastes for interim storage.

Skips of isotope cartridge waste would be recovered from the ponds and bulk sludge washed off. At the plant waste would be removed from the skip and a further low volume high pressure wash step performed prior to drying. After draining it would be loaded into a stainless steel drying can, placed in a basket and dried. The drying cycle would involve heating the waste material to 50$^{o}$C in an inert atmosphere and then cooling under vacuum (50mbar absolute pressure). The cycle would be repeated until the dried waste did not result in pressure change when held under vacuum (5-10mbar absolute pressure) for 15 minutes.

Once the isotope cartridge material is dry enough the baskets would be loaded into a 5mm thickness, cylindrical stainless steel canister (1400mm high x 610mm diameter), inerted, and welded shut. The canisters are designed to withstand a pressure of 30 bar absolute at 100°C, to accommodate gas production from radiolysis of any remaining water. After weld testing and decontamination, canisters would interim stored 4 to a stillage in the Box Encapsulation Plant Product Store. The CVD proposal is anticipated to result in about 81 canisters, contained in 21 stillages.

### *Pre-treatments*

Pre-treatment of AM, KCl and AlN cartridges is proposed to allow their disposal, if it is concluded that additional disposability challenges prevent them being conditioned with the other cartridges.

One option is to place dried cartridges into a thick walled stainless steel or copper vessel which would be welded shut to prevent radionuclide release for an extended period allowing radionuclides to decay to a safe level. These vessels could be included in either the encapsulated or CVD products.

The alternative pre-treatment option proposed for AlN cartridges is to extract carbon-14 from the AlN cartridges and chemically convert it to a low solubility carbonate for inclusion in an OPC grout wasteform.

### *Parameters for Assessment of Disposability*

### *Assessment inventory and number of packages*

A number of ‘assessment inventory’ waste packages were defined in order to assess properties and performance. Inventories representing maximum package dose rate, maximum activity (in $A_2$ multiples), maximum carbon-14 and maximum tritium (hydrogen-3) inventory were used in safety analyses. Average package inventory cases for the encapsulation and CVD options were produced by composing a summated inventory for all isotope cartridge types, and dividing by the total number of packages expected.

The isotope cartridges contain significant quantities of cobalt-60, tritium and carbon-14. Radiation from cobalt-60 (half life 5.7yr) will provide the greatest contribution to external dose rates and so is important for transport and operations. Tritium (half life 12.3yr) if released in gaseous form has the potential to contribute to radiation dose to the public during Geological Disposal Facility operations and transport. Carbon-14 (half life 5730yr) if released from the AlN cartridges as carbon-14 containing methane has the potential to contribute to the radiation dose to the public during the disposal facility operation, transport and in the post-closure period. The chlorine-36 radionuclide inventory in the KCl cartridges was assessed not to be sufficient to pose a significant risk to disposability.

### *Sorting, loading and exclusions*

Sellafield Ltd provided sufficient detail about the visual features they would use to identify and separate the different isotope cartridges included in the proposal, and the physical criteria which would be used if visual separation were not possible, for RWMD to conclude the proposal to be feasible. The decision trees presented in the proposal demonstrate that cartridge type can be determined reasonably reliably from such measurements. Identification by cartridge type would allow any excluded cartridge types to be separated and compliance with any package loading limits prior to waste conditioning. Significant further development work would be required to demonstrate the process as the proposals progress.

### *Wasteform properties – OPC grout*

Due to the protection offered from the pond skip, liner, grout and capping layers, the assessment concluded that some incomplete infiltration of the waste with grout could be tolerated. Reactive metal such as Magnox and aluminium in cartridge cladding would be expected to undergo acute corrosion in the highly alkaline grout which might result in fissures and internal voidage in the wasteform. However, since the wasteform would be capped and a clean annulus grout formed to finish the package the assessment concluded that the effect of a weakened wasteform on safety performance was lessened. It was concluded that so long as loadings were appropriately controlled, the production of wasteforms with acceptable properties was feasible.

In the longer term, expansive corrosion would occur with some expansion of the wasteform expected before the annulus grout would be applied. Further work is expected on the extent and implication of wasteform evolution on package performance.

Release fractions for the packages under fault conditions, derived from finite element models, were similar to those of other $3m^3$ box grouted wastes and no cliff-edge effects were predicted and so were concluded to have acceptable properties.

Assessment of KCl cartridges concluded that, in the quantities proposed, the hydroxyl ion to chloride concentration was high enough to prevent the chloride causing pitting corrosion. They could therefore be excluded from the need for separate pre-treatment.

### *Wasteform properties – MPC grout*

MPC grouted waste pH and ionic composition differs significantly from OPC (buffering at pH 8 compared to OPC at pH 13; and magnesium ions and phosphate ions compared to calcium ions and silicate ions), and might affect the function of RWMD reference case vault backfill. Scoping calculations indicated that the overall pH effects are likely to be small due to the limited number of packages proposed. The proposal was concluded to be acceptable at Conceptual stage, although further work to better understand the extent and implications of potential MPC/OPC interactions is required at Interim stage.

Development work by Sellafield Ltd was concluded to have addressed a number of Conceptual stage issues arising from previous assessments of this cement system with control of reactivity/set time, infiltration and waste compatibility demonstrated at the scale required. Development work has shown that MPC grout also has comparable strength and radiation stability to OPC grout.

Since the MPC grout fluid phase pH is between 5 and 6, corrosion of aluminium cladding would be expected to be significantly lower compared to the acute phase for highly alkaline grouts. However the lower pH may increase Magnox cladding corrosion rate compared to OPC which may lead to a requirement for Magnox cladding loading controls. Magnesium ions from corroded Magnox cladding could affect the MPC grout set rate, which would require further development and appropriate control. However the assessment concluded that so long as Magnox cladding loadings were appropriately controlled production of wasteforms with no cliff-edge effects and acceptable properties under fault conditions was feasible.

### *Wasteform properties – epoxy polymer encapsulant*

Recent development work by Sellafield Ltd indicated that epoxy polymer wasteforms, using the system chosen here, are currently not practicable at full scale, with relatively small scale development work resulting in excess heating and charring of epoxy polymer pours. The work did not progress to include waste simulants. The assessment concluded that this epoxy polymer system is not currently practicable at this scale of use and so did not assess the option further.

The radiation stability of epoxy polymer may not be sufficient for use with cobalt cartridges and further development work would be required to demonstrate compatibility.

### *Properties of CVD waste packages*

The CVD wasteform is comprised of waste which is not fully physically constrained, the geometry and corrosion state of which may be variable, from whole cartridges to corroded pieces. Loose particles are likely to be present or form during handling or as a result of accidents.

The assessment concluded that the proposal is not fully compliant with the requirements of the Generic Waste Package Specification (GWPS), the Level 2 Generic Specification for packages containing low heat generating waste and the geological disposal concept. The key conceptual stage issues are set out below.

The assessment identified a very substantial shortfall in the canister performance under accident conditions. Only a small fraction of the available kinetic energy (<10%) is required to cause loss of containment in aggressive impacts for a 15m drop. Since the dried waste is unconstrained and un-encapsulated, significant release of activity could result from impact accidents with implications for Geological Disposal Facility operations. Redesign to provide adequate safety performance would be required. Possible development options include strengthening the canister and/or providing secondary containment.

The CVD canister design height is not compliant with the current designs for the standard waste transport containers (SWTCs), such a transport container would be required for the transport of CVD canisters, and the canister handling features are non-compliant with the current designs for the Geological Disposal Facility related package receipt and handling systems. These would either require design changes to the canisters or changes to the disposal concept through formal change control. The latter option would only be considered if provision of appropriate canister performance was feasible.

### *Assessment of Disposability – Encapsulation Options*

Where appropriate the OPC grout wasteform was assessed providing a high bulk gas production case for assessment of the encapsulation options: and having similar physical properties to other inorganic grouts and providing high corrosion and gas generation rates. MPC wasteforms were not assessed quantitatively but qualitative differences are discussed. Since polymer was concluded not practicable at full scale it is not discussed further.

### *Transport safety assessment*

All of the $3m^3$ waste packages would be transported to the Geological Disposal Facility within standard waste transport containers. Radioactive gas releases from AlN and AM cartridges, based on pessimistic assumptions for rate of gas release, result in a breach of the containment requirements under normal conditions for a SWTC with 285mm of shielding (by factors of 5 and 110 times respectively). On this basis they would need to be excluded until further development work demonstrated their inclusion was acceptable for safe transport. Sellafield Ltd has recognised this providing pre-treatment options.

The bulk gas generation rate in OPC grout was estimated at up to 20 litres a day, which is within SWTC pressurisation limits and control of flammable gas concentration could be achieved by venting and purging with nitrogen before transport.

Bulk gas production and release from MPC grouted aluminium bearing wastes would be expected to be significantly lower reflecting lower rate of corrosion. Magnox reaction rate in MPC may be somewhat higher than in OPC and require further investigation and development of appropriate loading rates. Purging with nitrogen could be used to manage flammable gas releases during transport.

The $3m^3$ boxes proposed for the encapsulation option is based on the Silo Direct Encapsulation Plant (SDP) variant of the box. At the time of this assessment disposability issues concerning the box twistlock pocket design and box finish were awaiting resolution and were caveated in the assessment.

### *Operational safety assessment*

Using design basis accident analysis (DBA) for the operational phase of the Geological Disposal Facility estimated radiation doses to operatives and the public arising from fire and impact accidents are acceptable with doses from most scenarios within the basic safety objectives (BSO) and all below the lowest basic safety levels (BSL).

### *Post-closure safety assessment*

The assessment reviewed the effect of the isotope cartridge inventory on post-closure safety, concluding that the radioisotope inventory was already accounted for in the United Kingdom Radioactive Waste Inventory (UKRWI) and the current disposal system safety case and that the packaging proposals were consistent with those assumed in the UKRWI data sheets.

The AlN cartridges represent a concentrated form of carbon-14 compared to other wastes. This inventory could present a challenge to post-closure safety, particularly under pessimistic assumptions of instantaneous release. The assessment therefore concluded that AlN cartridges needed to be excluded from endorsement for direct encapsulation in OPC grout, requiring pre-treatment before disposal.

Inclusion of superplasticiser (SP) in the OPC grout formulation could increase the mobility of radionuclides in the waste within the groundwater pathway. Currently RWMD is undertaking work to identify an acceptable SP and until this is complete SP is excluded from use.

There is some uncertainty about the potential for interactions between MPC and OPC wasteforms in the long term and the potential for these to affect vault performance. Scoping calculations indicated the effects from the limited volume of MPC encapsulated waste would be likely to be small and so acceptable at this stage of the LoC process although further work would be required at Interim stage to fully understand the implication of MPC use on post closure evolution of the reference backfill system. Accordingly the assessment provides conclusions for this limited proposal for 20 MPC packages.

### *Criticality safety*

All the cartridges except the 6 irradiated thorium cartridges contain no fissile material and provide no criticality safety risk. The total fissile mass in the thorium cartridges is low and since the fissile content is intimately mixed with the thorium metal it is effectively poisoned. However since generic criticality safety arguments do not cover this material and so a package specific case and safe fissile mass would be required as the proposal progressed.

### *Assessment of Disposability – CVD Option*

### *Transport safety assessment*

Since the CVD canister could potentially become pressurised, it is important for Sellafield Ltd to establish whether it could be transported in compliance with the requirements of the transport regulations.

CVD packages are relatively and so high loadings of irradiated cobalt cartridges could result in a significant external dose rate. Loading rates of cobalt cartridges would need to be controlled to meet transport regulations requirements.

### *Operational safety assessment*

Operational safety was assessed using design basis accident analysis (DBA) for the operational phase of the Geological Disposal Facility. Doses from DBA fire accidents result in doses near to or below the BSO and are therefore acceptable. However the lack of robustness of the CVD canister results in unacceptable operational safety consequences to operatives with doses above the highest basic safety level (BSL). Doses to the public from the same DBA impact accidents are close to the BSO and therefore are largely acceptable.

### *Post-closure phase*

Following a period of container corrosion (up to 5,000 years) loss of canister integrity would lead to rapid release of radionuclides to the gas and groundwater pathways affecting the ability of the chemical barrier to work efficiently. The consequence on environmental risk could be significant for carbon-14 releases (as discussed previously in post closure safety for encapsulated waste) the assessment concluding that AlN cartridges needed to be excluded.

### *Criticality safety*

Most of the cartridges contain no fissile material, with the exception the 6 thorium cartridges. The total fissile mass in these cartridges is low. Since the fissile content is intimately mixed with the thorium metal it is considered to be effectively poisoned. This is acceptable at conceptual stage but any further development of the CVD option would need to include a package specific argument and derived safe fissile mass.

### *Assessment of Pre-treatment Options*

Following assessment of KCl cartridges as not requiring pre-treatment options were assessed for use with the remaining AM and AlN cartridge types.

### *Thick walled sealed vessels*

The AlN cartridges represent a reactive chemical form of carbon-14 in a concentration not found in other wastes. Since there is no GDF site and the effect of geology on gas hold up in the geosphere is uncertain pessimistic assumptions of instantaneous release were used resulting in a challenge to post-closure safety. It is estimated that the full AlN cartridge carbon-14 inventory of 0.8 terrabequerels would decay sufficiently to meet Geological Disposal Facility post-closure radiological requirements (against the $10^{-6}$ risk guidance level) in about 10 half lives or ~60,000 years; and approximately 3 half lives ~18,000 years would render releases from a single cartridge below the risk guidance level. The vessels would therefore be required to provide physical protection and containment to AlN cartridges and to withstand the geological forces which may be generated over a time period of 60,000 years.

Design and fabrication of stainless steel or copper vessels, up to 30mm thickness, was concluded as feasible. Similarly design of a closure mechanism for the vessels would be feasible. On this basis the proposals for welding “difficult” cartridge types (principally AlN cartridges) into 30mm thick walled sealed vessels were concluded to be technically feasible.

Stainless steel in high pH anoxic conditions (which could be assumed for OPC grout systems) would have a slow rate of corrosion (typically 0.1µm annually) and in such conditions the 30mm thick walls could last for ~300,000 years, providing the required containment time. Since environmental conditions required for a low rate of copper corrosion could not be presumed in the disposal concept considered here, further work would be required to justify use of copper vessels. Detailed design justification of vessel performance and longevity would be required as the proposals progress. Evidence of resilience of the vessel designs to the range of physical forces which could be generated over the post-closure period from geological processes would also be required.

Since the high pH conditions required for a low rate of stainless steel corrosion would not be provided by MPC grout the assessment concluded that MPC would not be suitable for encapsulating the long lived vessels.

The assessment concludes that the assumptions for AM cartridges, no loss of tritium during storage but full releases during transport, require these items to be excluded. They could be disposed of in the vessels to prevent releases of tritium exceeding transport containment limits, however the use of such designs may be excessive for this short term problem and Sellafield Ltd may wish to consider other options for controlling transport phase tritium gas releases from AM cartridges such as small scale use of polymer.

As a fallback cartridges which prove indistinguishable from AlNs could also be pre-treated in this way.

### Carbon-14 carbonate

Chemical conversion of carbon-14 in AlN cartridges to a low solubility carbonate would result in a product analogous to barium carbonate slurry from THORP off-gas treatment. A similar process has been used commercially by Amersham International Plc to extract and convert carbon-14 from similar AlN cartridges, so it was concluded feasible. The carbonate from THORP has been successfully packaged at the Sellafield Waste Encapsulation Plant (WEP) producing packages endorsed as compatible with the disposal concept and RWMD safety requirements. Therefore this proposal is meets RWMD requirements at Conceptual stage.

### Conclusions

The proposals from Sellafield Ltd for packaging of legacy isotope cartridge wastes from Sellafield ponds have been assessed. The assessment has concluded that proposals to encapsulate the wastes with OPC grout or MPC grout are consistent with plans for a Geological Disposal Facility and so can be endorsed at the Conceptual stage although there are some exceptions. The exclusions relate to direct encapsulation of tritium bearing AM cartridges and carbon-14 bearing aluminium nitride cartridges. A proposed option to encapsulate with polymer was not endorsed.

The proposals include pre-treatment options for cartridges which may otherwise not be disposable. The first option uses thick walled vessels capable of providing long term containment and physical protection to cartridges allowing the inventory to decay to a safe level. A stainless steel vessel was endorsed at Conceptual stage for disposal within OPC grouted packages. However the copper vessel option was not endorsed requiring further work before it meets RWMD requirements. The second option was chemical transformation of carbon-14 in AlN cartridges to carbonate for incorporation in OPC grouted packages. This pre-treatment was also endorsed at Conceptual stage.

Ongoing issues with the detailed design of the SDP box, use of superplasticiser and restriction to the scope of the MPC endorsement are addressed through an appropriate qualification to the CLoC.

The initial isotope cartridge identification step was concluded to be critical to facilitating the effective implementation of the packaging proposals for cartridges. The assessment concluded that sufficient information was provided for RWMD to support its feasibility and identify actions for further technical development of the identification process.

The conclusions and the implication for endorsement are summarised in the following table.

***Summary of Assessment Conclusions***

| Option | Conclusion | Exclusions / qualifications |
|---|---|---|
| OPC grout encapsulant | Conceptual stage endorsement | 1/ Exclude AlN & AM cartridges.<br>2/ Resolve issues with SDP box design.<br>3/ No use of superplasticiser until RWMD report produced. |
| MPC grout encapsulant | Conceptual stage endorsement | 1/ Exclude AlN & AM cartridges.<br>2/ Resolve issues with SDP box design.<br>3/ Endorsement limited to 20 MPC packages. |
| Epoxy polymer encapsulant | Not endorsed | - |
| Cold Vacuum Drying | Not endorsed | - |
| Pre-treatment: thick walled sealed vessel | Conceptual stage endorsement | Endorsement limited to stainless steel vessels in OPC grout wasteform |
| Pre-treatment: C-14 transformation to carbonate | Conceptual stage endorsement | Endorsement limited to disposal in OPC grout wasteform |