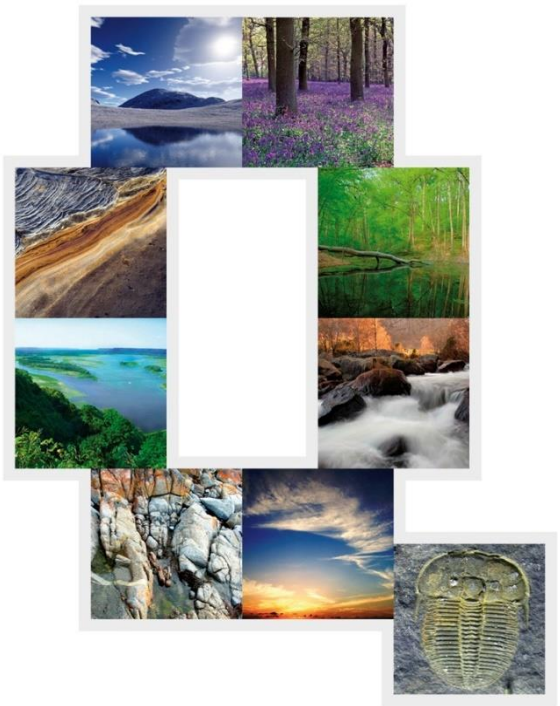
Potential Gas Generation in a Salt Repository

Waste Groups and High-level Conceptualisation



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Summary

COVRA wishes to develop a simple scoping-level model for estimating amounts and timescales associated with gas generation in a repository in a salt host rock. This document defines the waste groups that will be considered in that model. It then reviews the assumptions and models used in the OPERA project and by other waste management organisations to model gas generation in a deep repository. This information is then used to define high-level conceptual models for gas generation for each waste group. These conceptual models are subsequently developed into a functional specification by Benbow et al. (2023a) for use in calculations described in Benbow et al. (2023b).

Table of Contents

1 Introduction 4

2 Approach 4

3 Waste Groups 6

3.1 Groupings and Waste Descriptions used in OPERA 6

3.1.1 Heat-Generating HLW 8

3.1.2 Non Heat-Generating HLW 13

3.1.3 (TE)NORM 15

3.1.4 LILW 16

3.2 Proposed Groupings for this Study 19

3.3 Proposed Disposal Layout 21

4 Review of Gas Generation Processes 23

4.1 Post-closure Phase 24

4.1.1 Environmental Conditions 24

4.1.2 Corrosion Processes and Assumptions 25

4.1.3 Summary 39

4.2 Storage/Operational Phase 39

4.2.1 Environmental Conditions 39

4.2.2 Corrosion Processes and Assumptions 40

4.2.3 Summary 45

5 Processes and Assumptions to Include in this Study 46

5.1 Overall Assumptions 46

5.2 Breakdown by Waste Group 47

5.2.1 Vitrified Waste 47

5.2.2 Research Reactor Spent Fuel 48

5.2.3 Uranium Collection Filters 49

5.2.4 Reprocessing Waste 50

5.2.5 HLW Technical Waste – OPERA Concept 51

5.2.6 HLW Technical Waste – DDS Concept for Legacy Waste 53

5.2.7 HLW Technical Waste – KONRAD II Container Concept for Decommissioning Waste 54

5.2.8 Depleted Uranium 55

5.2.9 Molybdenum Waste 55

5.2.10 Non-compactible LILW 56

5.2.11 Compactible LILW 57

6 References 58

1 Introduction

COVRA wishes to develop a simple scoping-level model for estimating amounts and timescales associated with gas generation in a repository in a salt host rock. This document defines the waste groups that will be considered in that model. It then reviews the assumptions and models used in the OPERA project and by other waste management organisations to model gas generation in a deep repository. This information is then used to define high-level conceptual models for gas generation for each waste group. These conceptual models are subsequently developed into a functional specification by Benbow et al. (2023a) for use in calculations described in Benbow et al. (2023b).

The information that will be available to parameterise the model has some influence on the definition of waste groups and specification of processes to be represented because there is little point in attempting to model detail that cannot be parameterised. Further simplification may be necessary during development of an implementable functional specification, and approximations and assumptions may be needed during development of the model parameters from the available information.

The waste groups defined for OPERA (Verhoef et al., 2017) were used as a starting point for the definition of waste groups for this study. The OPERA waste groups were largely defined with aqueous transport to support dose assessment in mind. Modifications to split some OPERA groups and combine others were made to better reflect the gas generation potential of the different waste types in the inventory.

This report is structured according to the topics that need to be considered and the logical order for assessing them. First some general remarks are made in Section 2 about the overall approach and the 'boundary conditions' for the current work, which is part of a wider programme of work being undertaken by COVRA. Where possible, assumptions that are consistent with these other studies need to be made or the relevant characteristic needs to be identified as a parameter that can be varied in sensitivity studies to allow the results of all of the COVRA studies to be integrated. The report then considers the waste groups that are appropriate for this study (Section 3), the approaches for estimating gas generation that are used by some other waste management organisations (Section 4) and the gas generation processes that might be relevant for each waste group (Section 5).

2 Approach

The OPERA study (Verhoef et al., 2017) is the most recent published safety case for a deep repository in the Netherlands. OPERA focussed on exploring the potential performance of a repository in a clay host rock. OPERA included consideration of gas

generation in and release from the engineered barrier system (Box 6.2 in Verhoef et al., 2017). This study will review the assumptions made in that study and build on it as appropriate, bearing in mind that the OPERA analyses focussed on a clay host rock while this study considers a salt host rock. Two reports in the OPERA document suite (Hart et al., 2015a, b) considered a salt host rock. These reports reviewed the current state of knowledge of salt-based repositories and the potential application of this knowledge to safety cases for a Dutch repository in a salt host rock.

The approach to this study is to develop models for gas generation that are as simple as possible while at the same time representing the most important gas generation processes in order to understand the key controls on system behaviour. Inevitably, some aspects of the calculations will be constrained by the availability of data about the wastes and about conditions in the stores and repository. In some cases, generic information can be used to supplement the waste and site-specific information, but in other cases it will be necessary to make assumptions. It is important that these assumptions are clearly identified and do not become confused with real or generic data.

This project only considers the waste and the storage packaging. For some waste types, the waste is either repackaged or overpacked for disposal. Gas generation from any overpack or subsequent packaging is the subject of a parallel study being carried out for COVRA. An outline design for an overpack for high level waste packages developed by that study is considered in this work but other results were not available at the time of writing. The overpack has an important influence on conditions at the surface of the waste package that is within the scope of the current study because it will determine the access to water (timing and flow rate) and the geochemical conditions for the gas generating processes.

No site has yet been chosen for a repository. The characteristics assumed for the host rock are based on the general characteristics of potentially suitable salt host rocks found in the Netherlands. The key parameters will be the flow rate/availability and composition of water. Initial bounding calculations could assume unlimited water availability during the post-closure period, but more realistic calculations may be needed if these result in unacceptable gas pressures in the near field. These calculations will require assumptions to be made about water availability and flow in the host rock, for which no site-specific data are currently available. Salt formations are unlikely to be completely dry, although one bounding calculation would be to consider only the water that is introduced with the waste. The alternative scenarios that COVRA wish to explore are based on changes in water availability.

Different waste types will produce different total volumes of gas at different rates and times. It is important that the potentially important gas generating waste types are identified and parameterised but at this point the actual number of waste packages of each type is not crucial, provided there are enough for a package type to be considered

as a potentially significant source. The study will therefore focus on gas generation at a single package level.

3 Waste Groups

Radioactive waste in the Netherlands requiring disposal in a deep geological repository comprises:

- A range of waste types from the operation and decommissioning of nuclear power plants. Spent fuel is reprocessed and vitrified high level waste plus intermediate-level waste comprising metallic components of spent fuel assemblies are returned.
- A range of waste types from the operation and decommissioning of research reactors. The intention is to directly dispose of research reactor spent fuel.
- Low and intermediate-level waste from medicine, industry and the operation of nuclear power plants.
- Wastes arising from production of isotopes used in medicine, specifically production of medical molybdenum.
- Depleted uranium from URENCO uranium enrichment operations.

3.1 Groupings and Waste Descriptions used in OPERA

Wastes are grouped into families in OPERA (Verhoef et al., 2016). Families are groups of radioactive waste from the same origin, of similar nature, and having identical or closely related conditioning characteristics, while belonging to the same category of the current waste classification. The groupings reflect both the level of detail required for a safety assessment and the amount of information available about the different waste types. The top-level grouping is into LILW and HLW, using the Dutch waste classification scheme. HLW is then divided into heat-generating and non-heat-generating wastes, which will be placed in different parts of the repository. LILW is divided into LILW and (TE)NORM.

Figure 1 shows the waste families that were considered in OPERA. The characteristics of these families are summarised below (taken from Verhoef et al. 2016, 2017). An alternative waste packaging concept for non-heat generating waste (HLW technical waste) is also summarised. Verhoef et al. (2016) derives standardised description for each of the families, which met the needs of OPERA and is likely to largely meet the needs of this project. The standardised description includes: the origin of the waste (the generation and processing), the number of packages, characteristics of the waste

container (dimensions, steel or concrete type), the waste matrix (chemical composition of the waste), the radionuclides per waste container, how these radionuclides are expected to be present in the waste matrix and, if relevant, the expected heat output in 2130. The expected numbers of containers for each family are shown in Figure 1 and the total masses and volumes assumed in OPERA are given in Table 1.

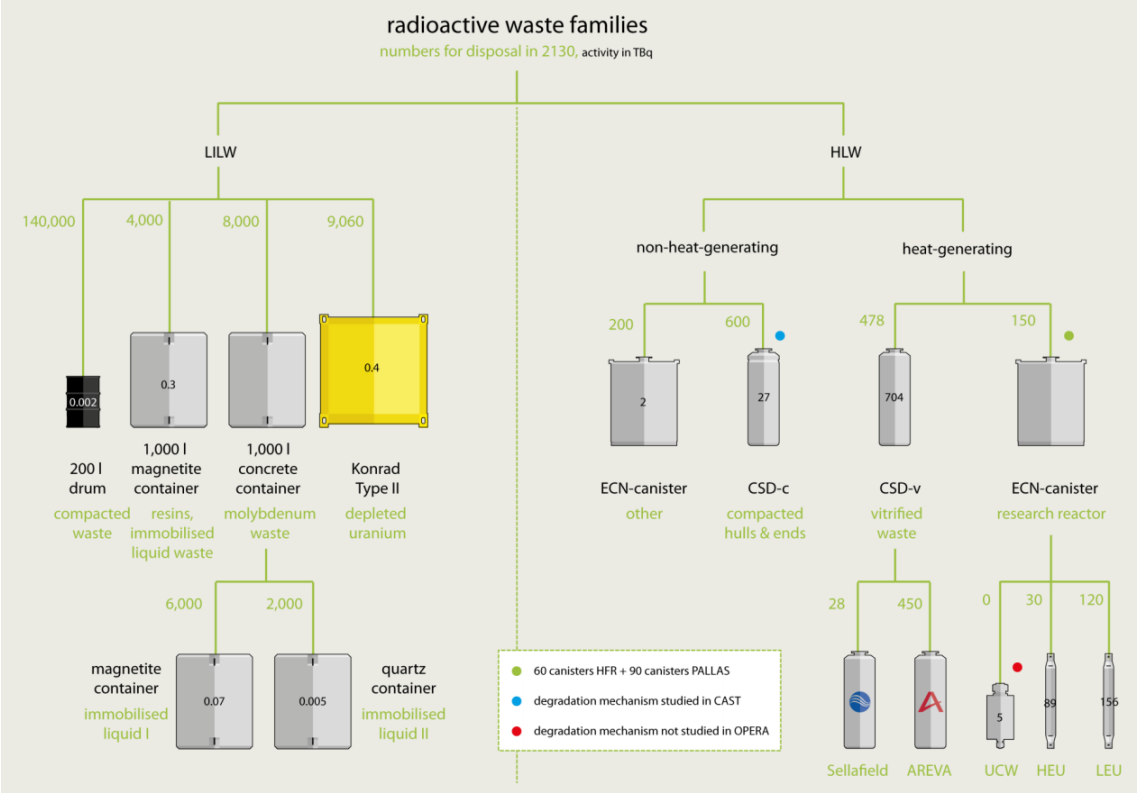

Figure 1: Waste families used in OPERA (from Verhoef et al., 2016)

Table 1: Waste masses and inventories assumed in OPERA (Verhoef et al., 2016)

Waste Category	In storage		Packaged for disposal		
	Volume [m3]	Weight [tonne]	Number of containers	Volume [m3]	Max weight [tonne]
Processed LILW	45000	150000	152000	45000	150000
TENORM	34000	110000	9060	40000	182000
Vitrified HLW	93	191	478	3388	9560
Spent research reactor fuel	104	99	75	638	1800
Other HLW	256	600	700	5104	14400

Verhoef et al. (2017) refers to 200 l and 600 l painted drums and to both 1000 l and 1500 l concrete packages for LILW. There appear to be 500 drums of 600 l and 1500 l capacity which are lumped in with the 1000 l packages in OPERA. Only 200 l drums and 1000 l

packages are shown in Figure 1 and described in Verhoef et al. (2016). This report takes Verhoef et al. (2016) as the primary data source but supplemented by other sources (mostly information supplied directly by COVRA) where information gaps were identified or clarifications needed.

3.1.1 Heat-Generating HLW

Heat-generating HLW consists of the vitrified waste from the reprocessing of spent fuel from the two nuclear power reactors in the Netherlands (Borssele and Dodewaard), the spent fuel of the two research reactors (Petten and Delft), and the spent uranium targets from molybdenum production.

Vitrified Waste

The primary container for vitrified waste is a fully sealed stainless steel container. The waste matrix is a borosilicate glass that immobilises the radionuclide inventory. Figure 2 shows the CSD container that is used for vitrified waste that is returned by Areva/Orano. The same outer container is also used for compacted hulls and ends, which are non heat-generating HLW (see Section 3.1.2). The small amount of vitrified waste from spent fuel that was reprocessed at Sellafield was returned in containers that are sufficiently similar to the Orano CSD container that a single type of vitrified waste container can be assumed.

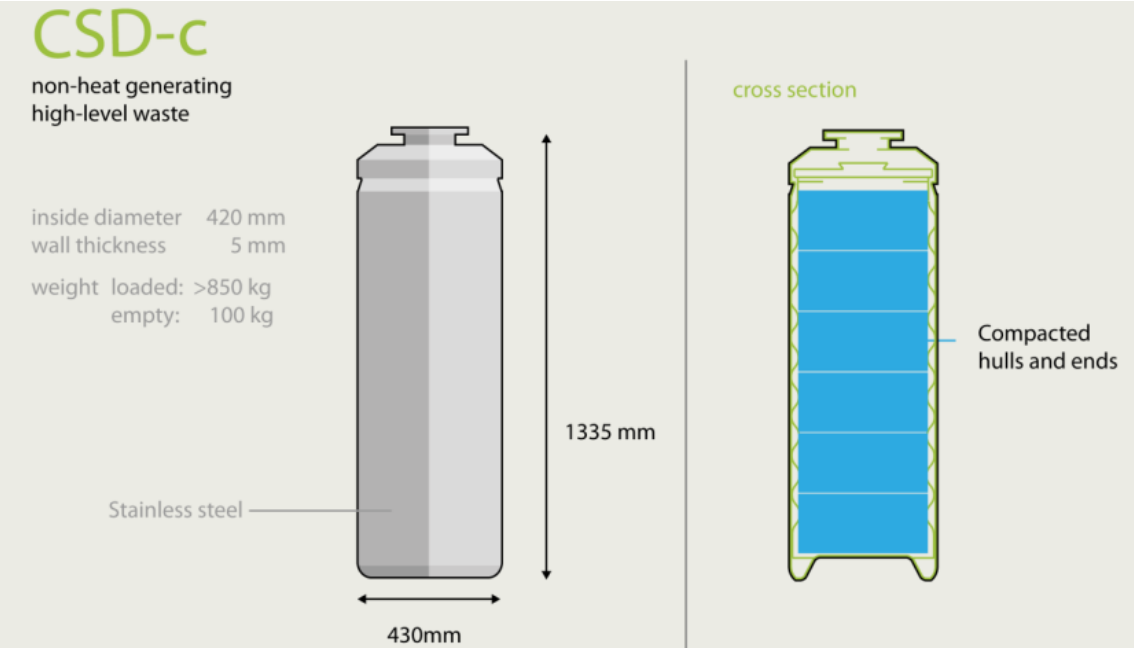

Figure 2: Areva/Orano Colis Standard de Déchets (CSD) waste container used for vitrified waste and compacted hulls and ends (from Verhoef et al., 2016)

These CSD containers will be overpacked for disposal. The overpack is not included in the scope of this project but will be an important control on the evolution of the stainless steel container and the timing/rate of gas generation from it and the waste. The overpack assumed in OPERA comprised a 30 mm thick carbon steel overpack within a supercontainer that incorporates a concrete buffer. For OPERA, there were two CSDs per supercontainer.

A new overpack has been designed in the parallel study (Wunderlich et al., 2023) being carried out for CVORA by BGE. A schematic of this overpack is shown in Figure 3. The carbon steel overpack has a wall thickness of 300 mm and the lid would be welded before final disposal. Each overpack would hold either 6 CSD containers (Figure 2) or 2 ECN canisters (Figure 5, top). This overpack might be expected to prevent water from coming into contact with the waste packages for at least several tens of thousands of years.

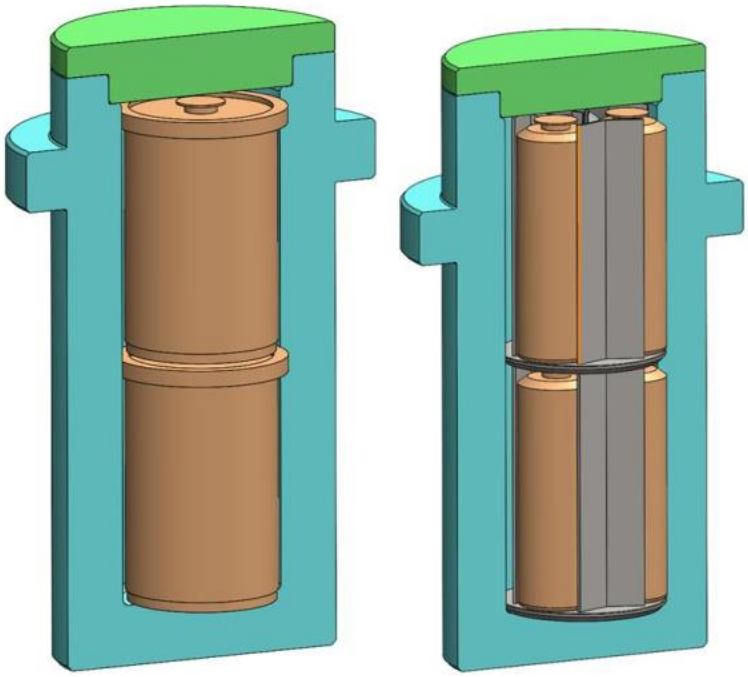

Figure 3: Conceptual design of new overpack for use in a salt repository (from Wunderlich et al., 2023)

Research Reactor Spent Fuel

There are three research reactors that produce or have produced spent fuel that will need to be disposed of in the repository; research reactor spent fuel is not reprocessed.

- The High-Flux Reactor (HFR) 45 MW_{th} in Petten. The HFR is a tank-in-pool type research reactor. The core is composed of 33 fuel assemblies and 6 control assemblies.
- The Low-Flux Reactor (LFR) 30kW_{th} in Petten. The LFR started operations in 1960 and was shut down in 2010.

- The Hoger Onderwijs Reactor (HOR) 2 MWth in Delft. The HOR is an open-pool type research reactor, using MTR-fuel assemblies and low-enriched Uranium-235 (< 20%) as fuel. The core is composed of 20 fuel assemblies and 4 control assemblies.

These reactors originally used highly enriched uranium (HEU) fuel but have now either been converted to low-enriched uranium (LEU) fuel or closed. The conversion of radioisotope targets from HEU to LEU was still ongoing in 2016. The research reactor spent fuel is placed in ECN waste containers for storage and disposal (Figure 5). The ECN container has a wall thickness of 5 mm and a welded lid.

OPERA assumes that all research reactor fuel has the characteristics of HFR fuel. However, the number of plates per assembly varies and this may affect the gas generation rate. The spent fuel assemblies are 40 – 150 µm particles of either UAl_x (HEU) or U_3Si_2 (LEU) particles dispersed in an Al matrix with an Al cladding (Figure 4). Figure 5 and Table 3-3 plus accompanying text in Verhoef et al., (2016, 2017) and Deissmann et al. (2016) provide additional geometric details.

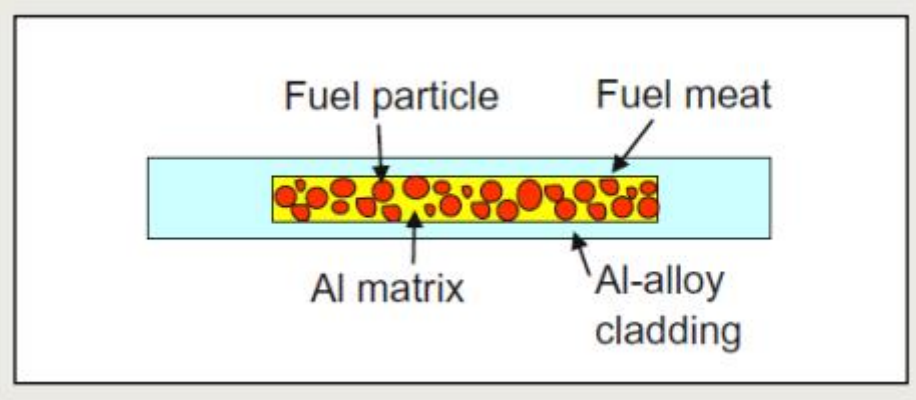

Figure 4: Schematic cross section through a dispersion type plate fuel used in research and test reactors. From Deissmann et al. (2016).

It appears from Figure 5 that the spent fuel assemblies are contained within an outer skin for which no material details have been found but, based on other waste types, it seems likely that this is aluminium.

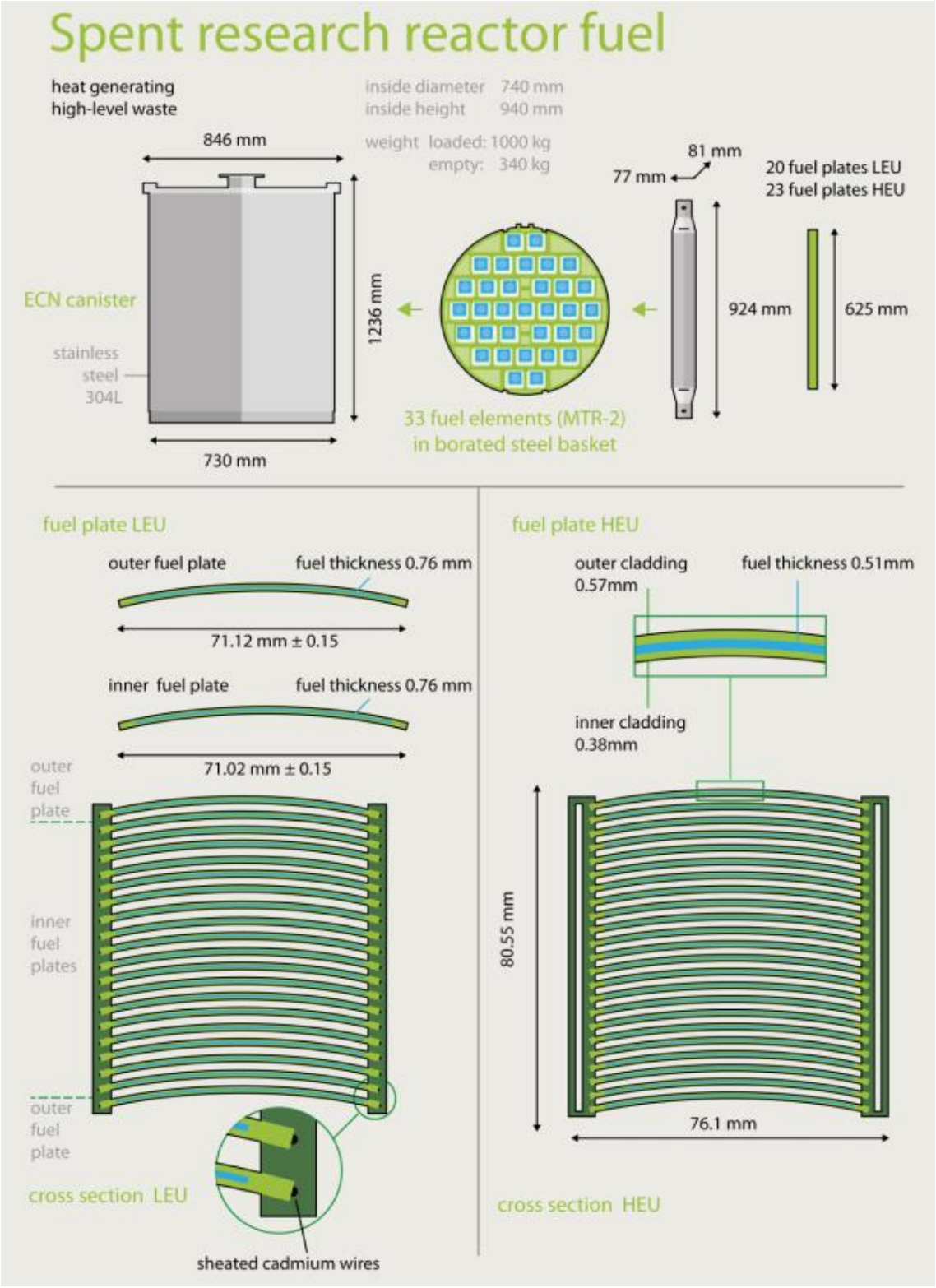

Figure 5: Schematics of research reactor spent fuel and ECN container (from Verhoef et al., 2016)

The ECN containers that contain the spent fuel will be loaded into overpacks. These overpacks will determine the evolution of water availability and composition at the

container surface but are outside the scope of the current project and so provide a boundary condition. The most likely overpack design for use in a salt repository is shown in Figure 3.

Uranium Collection Filters

The isotope ^{99m}Tc is a widely used for medical diagnostics. With a half-life of only 6 hours, ^{99m}Tc must be produced near to where it is used. In hospitals, the technetium is produced by the decay of ^{99}Mo. Mallinckrodt in Petten is the second largest producer of molybdenum for medical uses in the world. Production of medical molybdenum involves irradiation of an HEU target followed by dissolution to produce an alkali solution and a solid residue. The filtrate is collected in a precipitation vessel. Uranium precipitates in the pores of a stainless steel filter (sinter metal) as $Na_2U_2O_7.6H_2O$. The residue from a number of batches is collected in a collection filter (UCW filter). These filters are dried and subsequently packaged for storage at COVRA.

Three filters are placed in an aluminium casing, which is welded shut. Up to 33 of these aluminium capsules are then placed into an ECN container. The number of containers expected is small so these wastes were not considered explicitly in OPERA. It is assumed that these ECN containers would be overpacked in the same way as the ECN containers holding research reactor spent fuel.

Figure 6: Schematic showing Uranium Collection Filters and ECN container (from Verhoef et al., 2016)

3.1.2 Non Heat-Generating HLW

Non heat-generating HLW arises from the reprocessing of spent fuel and from activities such as research and the decommissioning of reactors. It is similar to wastes that are classified as intermediate level waste in many other countries.

Compacted Hulls and Ends

This waste arises from the reprocessing of spent fuel from Borssele and comprises the fuel assembly components that are not dissolved during the reprocessing. The equivalent waste from Dodewaard was not returned to the Netherlands as it was exchanged for a small amount of vitrified waste.

The waste comprises zircaloy hulls and various inconel parts. The end pieces and various operational wastes are stainless steel. These components are placed in 90 l cans and compacted to form pucks with a residual porosity of about 20%. An average of 7 or 8 pucks is then placed in a CSD container (see Figure 2). Verhoef et al. (2017) assume that these containers are placed in the same type of overpack as the vitrified waste.

Other HLW

This waste family includes legacy waste from a storage facility that was located at Petten. It comprises waste from four decades of nuclear research, including material residues (spent uranium targets and irradiated fuel) and fission and activation products. It will also include some waste from the dismantling and decommissioning of current nuclear facilities.

For the purposes of OPERA, this waste stream was assumed to comprise neutron activated metals from dismantled experiments, fuel cladding, and organic materials. The waste appears to be supercompacted into pucks before being placed in a concrete lined ECN container (Figure 7). These ECN containers ware likely to be overpacked in the same way as the ECN containers for research reactor spent fuel.

Figure 7: ECN container for 'other' non-heat generating waste (from Verhoef et al., 2016)

An alternative packaging solution has been proposed for this waste group, which is also considered in this study. As they are non-heat generating and largely ILW rather than HLW, it is proposed that they would be stored in the MOG and disposed of in the upper part of the repository with the LILW (see Section 3.3). The waste stream is divided into two parts (email from COVRA 28/9/22): legacy and decommissioning. The legacy waste would be placed in 'crinkle' drums, which are overpacked in DDS drums, four of which would then be placed into a concrete overpack (Amber), and decommissioning waste would be placed in KONRAD II containers (Figure 8). In both cases, the waste will be loose (i.e. not grouted). The DDS drum is fabricated from 304 grade stainless steel and the KONRAD II from carbon steel. Both container types have bolted lids so will not be fully sealed at the start of the post-closure period.

The DDS drum is within the scope of this project but the concrete overpack and KONRAD II container are not.

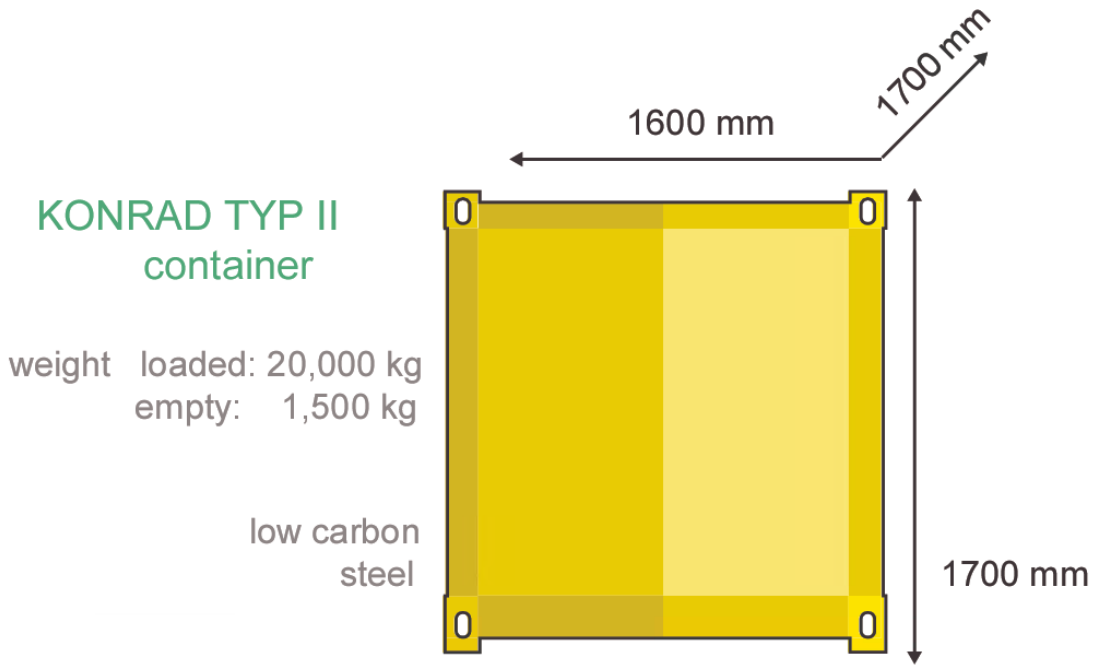

Figure 8: KONRAD container for non-heat generating HLW (from drawing supplied by COVRA).

3.1.3 (TE)NORM

The largest waste family by volume is depleted uranium generated as a result of URENCO's uranium enrichment activities. The uranium tails are in the form of U_3O_8 and are currently stored in standard 3.5 m^3 DV70 containers. It is intended that the depleted uranium will be conditioned with concrete in carbon steel KONRAD II containers for disposal (Figure 9). Verhoef et al. (2017) describes these containers as having a total volume of 4.6 m^3 and a payload of 4.2 m^3. It is noted that the dimensions of the KONRAD container given in Verhoef et al. (2016) appear to be incorrect and the dimensions given in Verhoef et al. (2017) and additional drawings supplied by COVRA (Figure 8) are used.

The KONRAD II containers are not in scope for the gas generation calculations. It is anticipated that an alternative design of container may be developed for the disposal of (TE)NORM in salt.

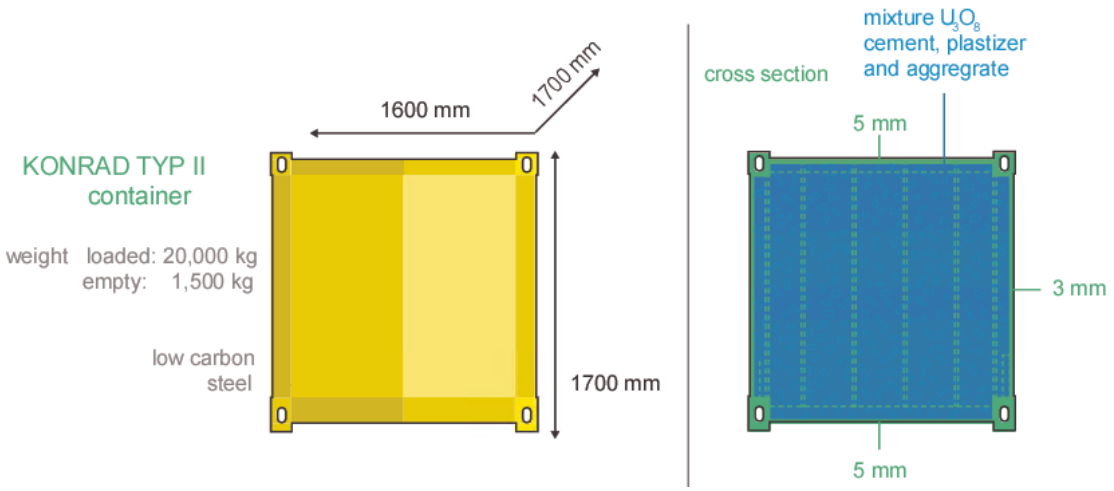

Figure 9: Schematic of KONRAD II disposal container and depleted uranium wasteform. Figure supplied by COVRA.

3.1.4 LILW

Compacted LILW Waste

Some two hundred organisations in the Netherlands ranging from nuclear power plants and research establishments to industrial users and hospitals produce LILW. Most of them generate only small volumes of low- and intermediate-level waste but there is a wide range of waste forms: solids, liquids of all natures, slurries, animal carcasses, machines, equipment, sealed sources, etc. LILW, mostly comprising concrete and metals, also arises from the dismantling of nuclear and other installations.

The majority of the LILW is solid compactable waste and is compacted into pucks before being transferred into concrete-lined 200 l drums (Figure 10). The drums are made from galvanised steel with a wall thickness of 1mm thick and are painted. They do not have a lid so any gas that is generated in the waste package can escape easily. There are generally 4-7 pucks per drum and the residual space is filled with concrete. There are concrete discs at the top and bottom of the drum. The waste comprises cellulosics, plastic, metal, glass concrete etc. There are also some dried sludges. OPERA assumed that all waste in 200 l drums is compactible, although this will not actually be the case. The estimated amount of each type of material is given in Table 2.

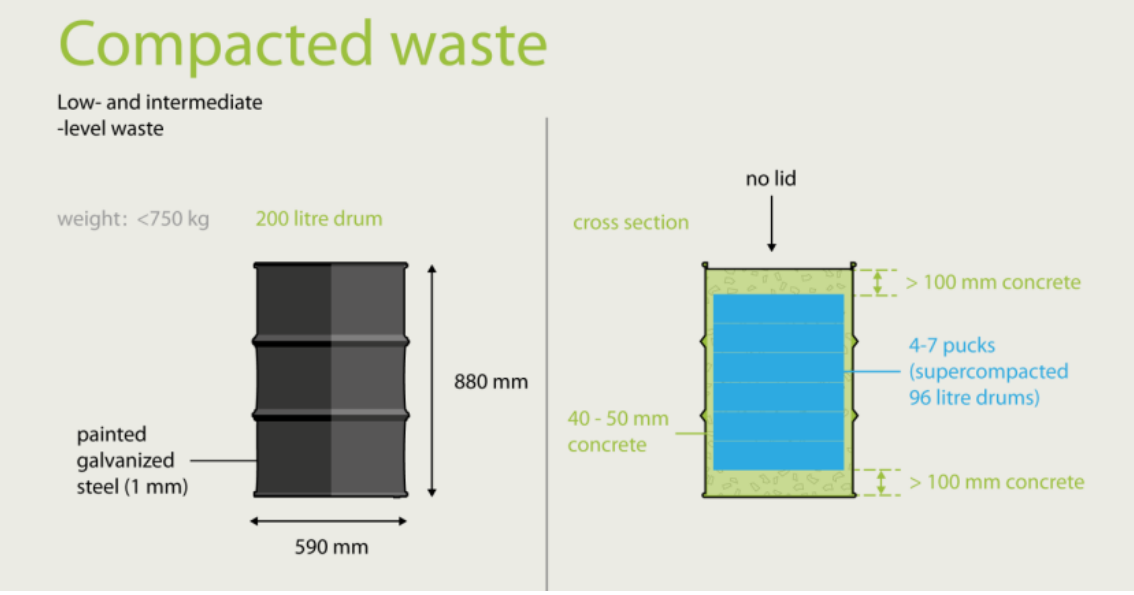

Figure 10: Schematic of 200 l concrete lined drums containing compacted LILW.. (from Verhoef et al., 2016)

Table 2: Estimated average composition of the compacted waste family (from Verhoef et al., 2016).

Matrix	Specification	Fraction of matrix	Volume [%]	Margin [%]
Organic materials	Cellulosics (cloth, paper, tissue) organic sludge etcetera	1	35	20
Metals	Steel	0.75	30	20
	Aluminium	0.25		
Plastics	Halogenated	0.10	25	20
	Non-halogenated	0.90		
Others	Glass, rubber, concrete, inorganic adsorption materials, salts et cetera	1	10	10

Processed Liquid Molybdenum Waste

This waste arises from the conditioning of the two alkaline waste streams arising from the production of medical molybdenum. The uranium collection filters used in the process are considered in Section 3.1.1. OPERA only considered the waste from processing HEU targets; a new process is being developed for LEU targets.

The two liquid waste stream are cemented into 200 l drums, which are then grouted into 1000 l reinforced concrete containers (Figure 11). The aggregate in these containers is magnetite for waste stream 1 and quartz for waste stream 2.

It is not clear how/whether these waste packages are vented. The 200 l drums have lids, but it is assumed that these lids are bolted and not gas or water tight in the long term.

The concrete containers are designed to have sufficient gas permeability to allow gas to escape before the waste package becomes over-pressurised.

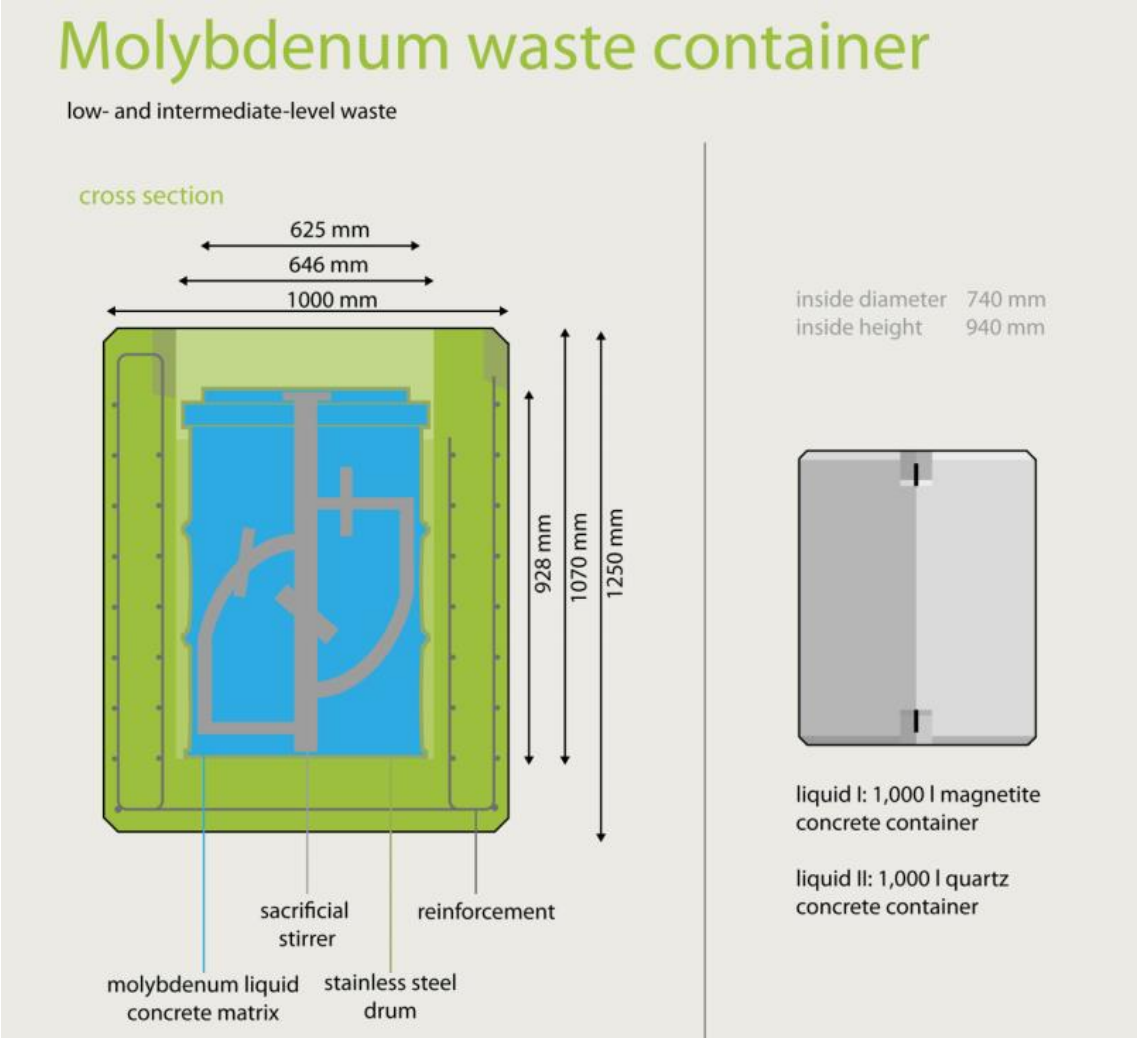

Figure 11: Schematic of waste package for processed molybdenum waste (from Verhoef et al., 2016)

Ion Exchange Resins and Sludges

Ion exchange resins are generated as operational waste from all of the reactors. It appears that the only resins that are conditioned with cement are those from Borssele. It is not clear how the other resins are conditioned and packaged, but the working assumption is that they will be conditioned in the same way as the resins from Borssele.

The Borssele PWR uses bead and powdered resins. These are conditioned in cement, together with the sludge from the ponds in a similar manner to the molybdenum wastes. The outer concrete container is the magnetite variant (Figure 11). As for the molybdenum wastes, it is assumed that the waste packages are designed to have sufficient permeability to prevent pressurisation.

3.2 Proposed Groupings for this Study

The waste families used in OPERA provide a good basis for the gas generation work, not least because it makes sense to maintain compatibility with earlier work wherever possible. They reflect the different types of proposed waste packaging and characteristics in terms of radionuclide inventory and release characteristics but are not optimised from the point of view of assessing gas generation. This section describes some proposed modifications and the reasons for proposing them.

Table 3 shows the waste grouping that are proposed for the gas generation work.

Table 3: Proposed waste groups for gas generation

Group	OPERA Waste Families	Notes
Vitrified Waste	Heat generating HLW CSD-v	Vitrified waste from La Hague (majority) and Sellafield considered together. Will be overpacked but scope only extends out to the outer surface of the stainless steel CSD container.
Research Reactor Spent Fuel HEU	Heat generating HLW ECN	This covers the HEU research reactor fuel. Will be overpacked but scope only extends out to the outer surface of the stainless steel ECN container.
Research Reactor Spent Fuel LEU	Heat generating HLW ECN	This covers the LEU research reactor fuel. Will be overpacked but scope only extends out to the outer surface of the stainless steel ECN container.
Uranium Collection Filters	Heat generating HLW ECN	This covers the collection filters for both HEU and LEU. It is a small number of packages but they may produce significant gas. Will be overpacked but scope only extends out to the outer surface of the stainless steel ECN container.
Reprocessing waste	Non-heat generating HLW CSD-c	These packages contain cladding and hulls returned from La Hague. Will be overpacked but scope only extends out to the outer surface of the stainless steel CSD-c container.

Group	OPERA Waste Families	Notes
HLW Technical Waste (OPERA concept)	Non-heat generating HLW ECN	These packages mostly contain high/long-lived research and decommissioning waste with a variety of material types. Will be overpacked but scope only extends out to the outer surface of the stainless steel ECN container.
HLW Technical Waste (Alternative concept)	Fraction of non-heat generating HLW ECN	Placed in DDS containers with no compaction or grouting. Will be overpacked but scope only extends to outer surface of the stainless steel DDS container.
HLW Technical Waste (Alternative concept)	Fraction of non-heat generating HLW ECN	Placed in KONRAD II containers with no compaction or grouting. Scope only includes waste, not KONRAD II container.
Depleted Uranium	LILW Konrad Type II	Grouted uranium oxide. KONRAD II container not in scope. Currently stored in DV70 containers.
Molybdenum Waste	LILW 1000 l concrete container (both magnetite and quartz)	All cemented Molybdenum processing waste regardless of containers aggregate. The two different waste streams should be close enough for gas generation purposes. Note reinforcement and mixing paddle.
Non-compactable LILW	LILW 1000 l magnetite container	This stream includes resins and other non-compactable waste including metals. May include metal waste or mixing paddle and concrete has reinforcement
Compactible LILW	LILW 200 l drum compactible waste	This waste includes a range of materials that may provide different source terms so a range of cases may need to be considered to allow impacts to be bounded.
		'Average' package, which includes a mix of waste types

Group	OPERA Waste Families	Notes
		'Organics' package, which only includes compacted organic waste
		'Metals' package, which only includes compacted metal waste
		'Plastics' package, which only includes compacted plastic. May be possible to combine with inert depending on parameters
		'Inert' package, which only includes non-gas-generating waste so the only source is the various metal containers

Some notes about the table are provided below:

- It currently appears that the gas generation profiles for HEU and LEU research reactor spent fuel may be slightly different as a result of different surface areas and package loadings for cases where gas generation is not water limited. Therefore, the research reactor fuel is split into two waste groups.
- It is proposed to combine the different Molybdenum waste streams as the differences seem to relate to radionuclide fingerprint rather than gas generation potential.
- It is proposed to consider the compactible waste at the level of a waste puck and then combine the waste pucks in different ways with a 200 l steel container to generate waste packages. It will be important to understand the likely variability in the gas generation profile for these waste packages in order to understand whether, for example, selective emplacement might be useful to manage gas and whether it would be wise to ensure that the generation by particular types of package are minimised in the future.
- It may be informative to consider bounding as well as average packages for HLW technical waste and non-compactible LILW owing to the variety of waste materials that are expected.

3.3 Proposed Disposal Layout

The repository is assumed to be constructed on two levels with waste that is overpacked in the steel overpacks (Figure 3) on the lower level and other waste on the upper level. This subdivision places all of the heat generating waste and much of the ILW at the lower level. Figure 12 and Figure 13 show illustrative layouts supplied by COVRA. The location of the HLW technical waste depends on the packaging concept. If the OPERA concept is used it will be placed in the lower level and if the DDS/KONRAD packaging is used it will be in the upper level.

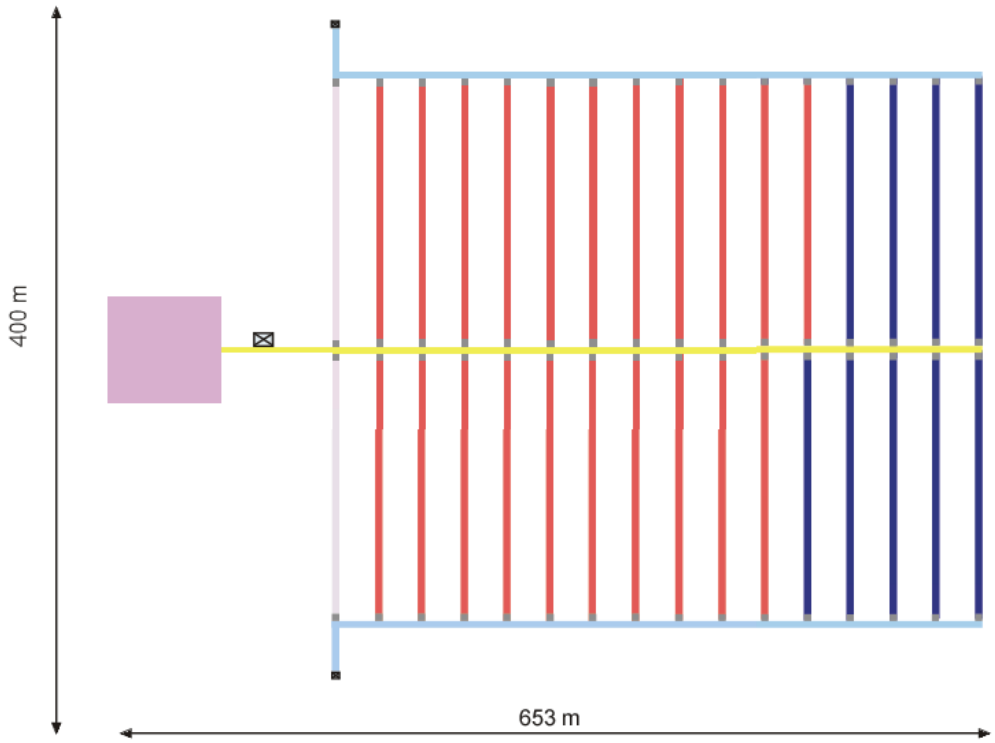

Figure 12: Illustrative layout for the lower level of the repository, assuming the new concepts for HLW technical waste (from COVRA). Red tunnels contain heat-generating packages and blue tunnels contain non-heat generating packages.

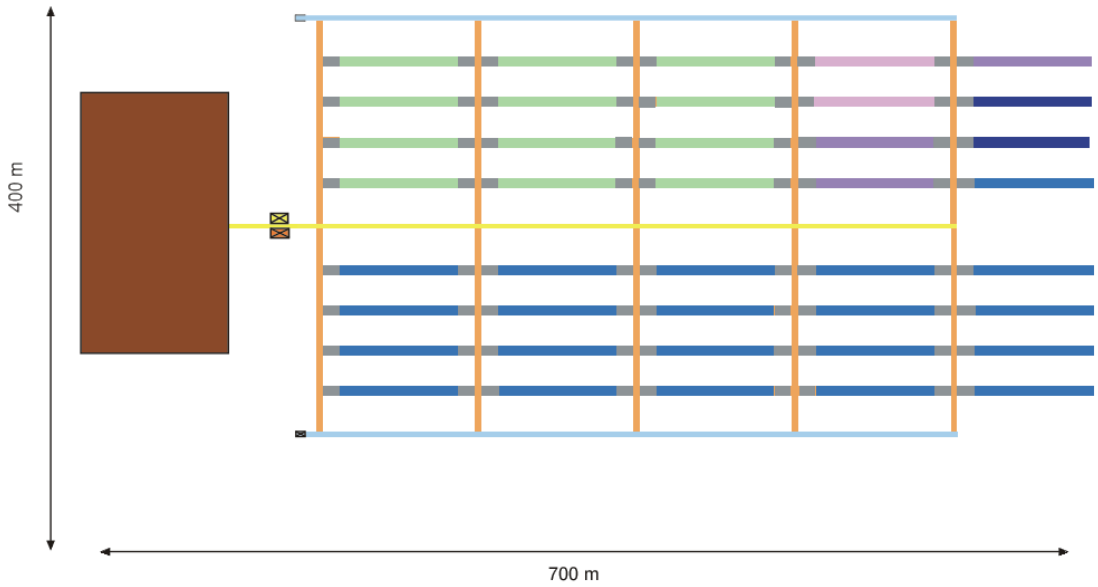

Figure 13: Illustrative layout for the upper level of the repository assuming the new concepts for HLW technical waste (from COVRA). Different colours indicate disposal of different types of waste.

4 Review of Gas Generation Processes

This section considers the gas generation processes that may be relevant for the COVRA waste. It considers processes that have been considered in other studies, including OPERA (Verhoef et al., 2017). Section 5 provides recommendations for the processes that are likely to be significant and merit inclusion in this study.

The gas generation processes that may be important are determined by the combination of the source materials and environmental conditions, including the availability of water.

The major gas generating processes are expected to be (RWM, 2016):

- Corrosion
- Microbial degradation of organic matter
- Radiolysis
- Radioactive decay.

However, as discussed below, these are not the only processes that might affect the amount of gas that is present. For example, processes such as dissolution may reduce the volume of free gas that is generated and processes such as methanogenesis may alter the amount and composition of the gas.

It is assumed for this work that processes such as dissolution/exsolution and methanogenesis that alter the volume and composition of the gas after it has been generated are out of scope. However, it is noted that there may be no significant time between gas being generated and it taking part in other such reactions. Methanogenesis ($CO_2 + 4H_2 = CH_4 + 2H_2O$) may proceed simultaneously with production of H_2 and CO_2 and dissolution of CO_2 and carbonation of cement would likely be fast. The simplifications assumed are therefore conservative as they would tend to reduce the volume of gas in the near field.

For the expected wastes the principal potential source materials for gas generation are:

- Metals both in the wastes and in the waste packaging
- Organic materials that are subject to microbial degradation
- Organic materials that are susceptible to radiolysis
- Water that is incorporated into the waste and encapsulants that may be subject to radiolysis.

Overpacks, structures associated with waste emplacement (e.g. stillages) and structural materials and equipment left in the disposal vaults and tunnels are outside the scope of this study.

The storage/operational and post-closure phases are considered separately because the environmental conditions and water availability are likely to be significantly different for these two periods. It is also important to account for the impact of the operational phase on the potential for gas generation during the post-closure phase, for example as a result of gas generating material that is consumed during storage. A long storage period for a vented waste package has the potential to reduce the total volume of gas that must be managed during the post-closure period.

The difference in environmental conditions between different phases (e.g. oxic, low-salinity water present in storage phases, but anoxic, hypersaline water post-closure) means that different processes will dominate for the two periods. Most studies consider the storage / operational and post-closure periods separately and few studies actually consider both. Most reports focus on the post-closure phase so that is considered first in the material presented below.

4.1 Post-closure Phase

4.1.1 Environmental Conditions

It is generally assumed that environmental conditions cannot be controlled during the post-closure phase but that it is possible to predict the likely conditions at any given site. Conditions vary between repository concepts and sites and will evolve most rapidly during the early part of the post-closure period while the disturbance to the geosphere caused by repository excavation and operations diminishes towards the natural background. Thereafter, the evolution of environmental conditions will be much slower and reflect long-term processes such as climate change. The environmental conditions seen by the COVRA waste packages that are overpacked will largely be defined by the design of the overpack; one of the functions of the overpack is to control environmental conditions at the surface of the waste packages. Key features of the environmental conditions expected in a COVRA repository constructed in salt are:

- Conditions will become reducing once any oxygen that is trapped at closure has been used up by corrosion and other degradation processes.
- Water will be more available than during the storage period but is not expected to be abundant for a repository in salt. Resaturation is expected to be slow, and may not complete. Degradation processes are likely to be water limited for a normal evolution scenario in salt but scenarios where water is more abundant can be conceived. Water that is disposed of as part of the waste packages/overpacks is likely to be important.

- Any water that enters the repository from the host rock is expected to be highly saline. For overpacked waste packages, the composition of the water in contact with the waste containers will be strongly influenced by the composition of the overpack. The overpacks proposed for the HLW waste packages are carbon steel so will not influence the pH of the incoming water at the surface of the waste package (Figure 3). However, a cementitious overpack, such as is proposed for the DDS drums (Section 3.1.2) or the cementitious overpack assumed in OPERA for HLW would be expected to result in the water in contact with the waste package being alkaline.
- The temperature will be determined by factors such as the waste package spacing and the depth of the repository. Salt has a relatively high thermal conductivity but waste package temperatures are likely to be higher than during storage.
- The waste packages may be subjected to significant loading as the salt creeps. The loading may result in mechanical failure of the waste package, which may affect the access of water to the waste. COVRA's reference assumption is that HLW waste packages would remain intact for at least 500 years but most likely much longer.

4.1.2 Corrosion Processes and Assumptions

RWM's SMOGG Model

Radioactive Waste Management in the UK uses a simple model called SMOGG (Simplified Model of Gas Generation) to assess the gas generation rate (Swift, 2016). SMOGG is implemented in C++ with a spreadsheet front end. It is intended to be used to assess gas generation during all phases of the repository programme. SMOGG was originally developed when RWM's focus was on a cementitious disposal concept in a higher strength fractured host rock and many of the assumptions reflect this environment, which has a high water availability and assumes highly alkaline porewater for the majority of the assessment period.

The model specified focuses on gas generation on a waste package or, equivalently, a waste stream basis. This maintains the required consistency of approach for different circumstances (even after closure, containers are expected to retain their integrity for the time during which most gas is generated for the conditions for which SMOGG was originally developed). Repository or vault scale gas production is obtained by summing over representative sets of characteristic packages. The model represents the following gas generation processes:

a) Corrosion of stainless steel, carbon steel, Zircaloy, uranium, Magnox and aluminium to produce hydrogen;

b) Release of radionuclides (H-3, C-14, Kr-81, Kr-85, Ar-39, Ar-42) in gaseous form (i.e. because they are in themselves gases under relevant conditions (Kr and Ar) or incorporated into gaseous molecules, CH_4 or H_2 in the case of 3-H and CH_4 or CO_2 in the case of 14-C) from stainless steel, carbon steel, Zircaloy, uranium, Magnox and aluminium due to corrosion and diffusion;
c) Degradation of cellulose to produce glucose or ISA, and subsequently microbial degradation of glucose or ISA to produce carbon dioxide and methane;
d) Release of 14-C containing gases from microbial degradation of glucose or ISA;
e) Radon production from radioactive decay of 226-Ra and its parents;
f) Production of gas by radiolysis of cementitious materials containing water, cellulose and a number of polymeric materials;
g) Release of 14-C-containing gases by radiolysis of small molecules;
h) Release in gaseous molecules of "trapped" 3-H and 14-C from graphite;
i) Reaction between carbon dioxide and hydrogen generated by other processes to produce methane.

SMOGG also provides a facility to estimate the volume expansion in the wasteform as a result of corrosion to inform estimates of container lifetime. The gas generation processes included in SMOGG are summarised in Figure 14.

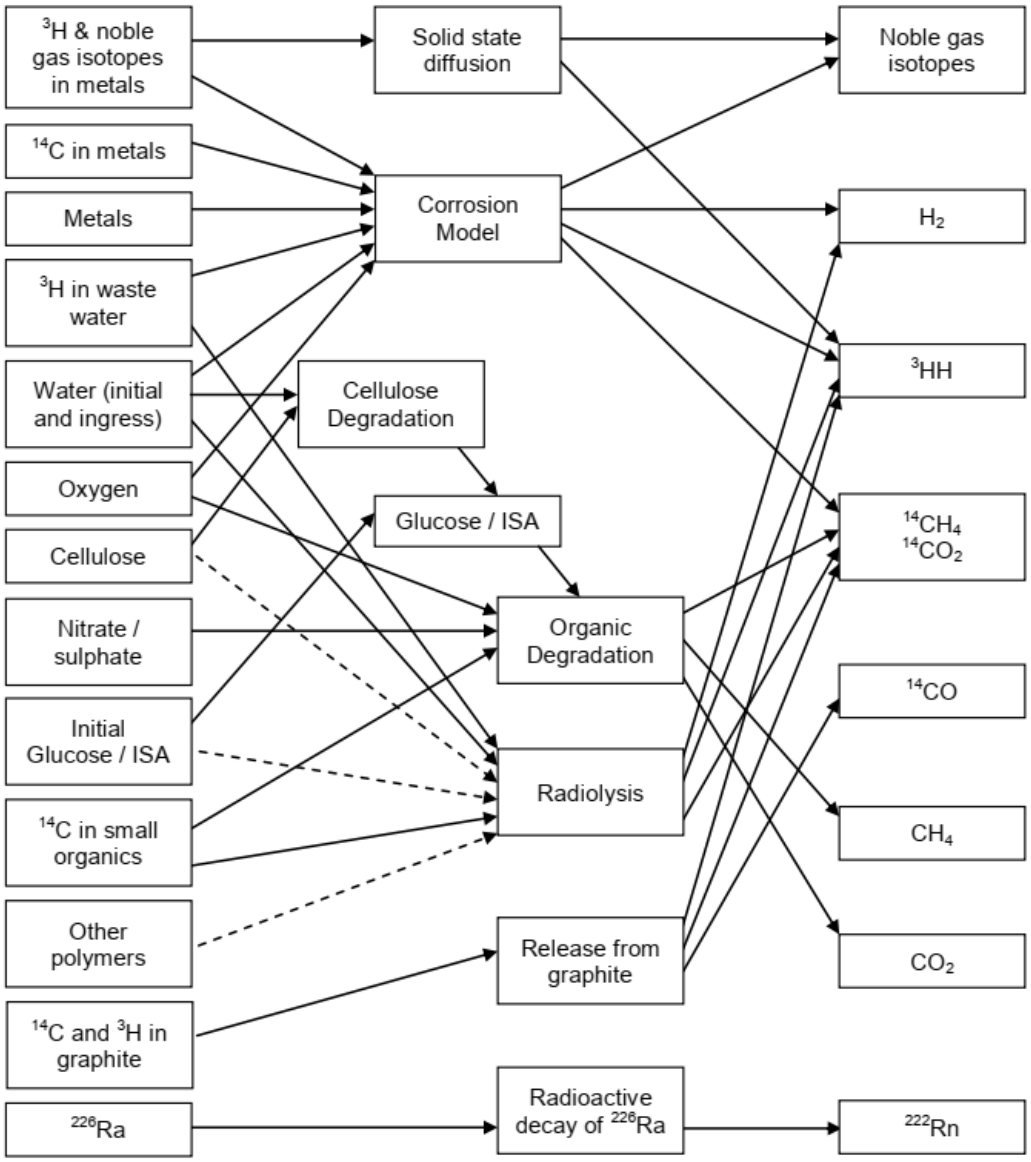

Figure 14: Gas generation processes included in SMOGG. Dashed lines indicate that consumption of the substrate is not modelled. From Swift (2016).

The SMOGG model allows for emplacement of different waste streams at different times or for a waste stream to be split into several emplacements. The gas generation calculations require information on water availability, oxygen availability, temperature, pH (SMOGG assumes LLW and ILW will always be cement conditioned and that the backfill is always cementitious) and chemical conditions (e.g. presence of certain reactants and groundwater composition, notably chloride content). Container walls have inner and outer surfaces, allowing for different corrosion behaviour inside and outside, and there is a distinction between metallic fuel cladding and metallic fuel.

RWM assumes that resaturation will occur over a period of between a few years and 10,000 years, with most work having focussed on short resaturation times. It is possible

to define the resaturation time and the time at which conditions change from oxic to anoxic. Where the gas generation mechanisms are well understood, properly representative gas release models are implemented, but where there is less understanding more empirical models are used.

Corrosion of Aluminium and Magnox produces hydrogen under both oxic and anoxic conditions. Corrosion of steel, zircaloy and uranium under anoxic conditions produces hydrogen. Corrosion rates are pH and time dependent to match observed behaviour. Two phases of corrosion, acute and chronic, with associated characteristic times are included in the model:

$$\frac{ds}{dt} = -k_a e^{-\frac{t}{t_a}} - -k_c e^{-\frac{t}{t_c}}$$

where s (m) is the location of the corroding surface at time t (y), k_a and k_c (m/y) are the initial and chronic corrosion rates and t_a and t_c (y) are the characteristic times for acute and chronic corrosion. The corrosion rates vary with chemical conditions (oxic or anoxic, pH and high/low chloride) and temperature. The rate of gas generation can then be calculated from the volume of metal that is corroded and the stoichiometry of the corrosion reaction. Metals are represented as either plates or spheres with the appropriate thickness/radius and total mass (to allow surface area to be determined).

SMOGG also calculates the release of radioactive gases, including gaseous C-14, from the corroding metal.

Radiolysis of pure water and water in cementitious materials produces hydrogen with the production rate being material/water composition dependent. Radiolysis of organic compounds present in wastes and of polymeric encapsulants can lead to the generation of a variety of gases. SMOGG assumes that the main gas produced is hydrogen but also includes the release of carbon dioxide and methane. A wide range of organic materials and resins are included in the radiolysis model. Radiolysis is represented using a standard model:

$$Q_R = \sum_{v=\alpha,\beta,\gamma} G_v D_v$$

where Q_R (mol) is the cumulative quantity of gas, G_v (mol/J) is the G value for each radiation type and D_v is the cumulative absorbed energy for each radiation type. It is assumed that all α and β energy is absorbed within the waste package and a user-specified fraction of the γ energy escapes from the package.

SMOGG considers the production of gas by microbially mediated degradation of cellulose and small organic molecules. It uses empirically-derived rate constants to describe the gas generation rates. The rate constants depend on the amount and type of microbes present with certain conditions, such as very high pH assumed to inhibit microbial activity. It is conservative to assume that microbes might be present. Cellulose

is initially hydrolysed to small organic molecules, which are then degraded to produce carbon dioxide and methane. Production of methane requires anoxic conditions and the absence of nitrate and sulphate. Prior to exhaustion of nitrate and sulphate, only carbon dioxide is produced.

The only gas produced by radioactive decay that is included is 222-Rn. The short half life (3.8 days) means this is unlikely to be significant during the post-closure period as the majority will be retained in the waste packages or repository. It may however be important during the storage/operational period (see Section 4.2).

SMOGG includes the release of radioactive gases from metals by solid state diffusion but notes that the underpinning data is limited. The large inventory of irradiated graphite in the UK inventory means that SMOGG also includes an empirical model for the release of radio-labelled gases from graphite. It is assumed that no bulk gas is released from graphite.

Swift (2016) discusses the coupling between different waste streams under conditions where the amounts of oxygen and water are limited. For oxygen, the user must specify how the oxygen present at closure is partitioned between the waste streams and conditions become anoxic on a waste stream (or package) by waste stream basis as this oxygen is consumed. For water availability, two options are available:

- For vented containers, resaturation can be specified as occurring between closure and a user-specified time. SMOGG calculates the inflow rate required to achieve this resaturation rate in the absence of any water consuming reactions and if there is insufficient water available scales the rates of corrosion, hydrolysis of cellulose and radiolysis to the available amount of water. After the user-specified resaturation time, there is assumed to be no water limitation.
- The rate at which water enters the package and its associated backfill is specified. If there is insufficient water available to support all the water-consuming reactions, these are scaled back. SMOGG keeps track of the degree of saturation and once full saturation has been achieved assumes that there will be no further water limitation.

RWM's 2016 Gas Status Report

RWM (2016) provides an overview of gas generation processes and includes a brief discussion of the likely gas generation rates for a repository in an evaporite host rock. It considers that the dry environment means that gas generation is likely to be very limited but the water present in the waste packages will be important. It notes that the gas generating materials include waste, waste packaging (containers and encapsulants) and materials associated with the construction and operation of the repository. It considers the main mechanisms of gas generation to be corrosion, radiolysis and microbial

degradation. The bulk of the gas will be hydrogen but there will also be some carbon dioxide and methane. Gases may be tritiated or carbon-14 labelled and it may be necessary to consider Radon-222. It also notes that some radioactive gases may be released by diffusion or leaching and some gas may be generated by radioactive decay.

RWM (2016) makes the following high level statements about gas generation processes and includes further details about rates and reactions.

- Hydrogen is produced from the corrosion of iron, steels, Zircaloys and metallic uranium exclusively under anaerobic conditions. Hydrogen is also produced from the corrosion of Magnox and aluminium under aerobic and anaerobic conditions. Under highly alkaline conditions Magnox, aluminium and uranium will corrode rapidly. Corrosion depends on the environment surrounding the metal. The presence of water is an important prerequisite for gas generation by corrosion. During transport and operations, this will be provided by the water associated with the wasteform (for example, the grout porewater in cement-encapsulated LLW and ILW) and the relative humidity of the environment around and within the package. If the host rock has a low permeability the restricted supply of groundwater may limit corrosion. Evaporite rock (halite), if the far field remains undisturbed, is a practically dry environment, very small amounts of water that is present being located within non-connected fluid inclusions. They also note that pore/groundwater composition, temperature and presence of oxygen affect the nature and rate of corrosion.
- Microbial action requires the presence of water. The most important process is the degradation of cellulose. Gases such as carbon dioxide and methane will be generated as a result of microbial degradation of organic materials in ILW and LLW. Gas production from microbial action within the repository after closure will be very heterogeneous, with broad ranges of possible generation rates and time dependencies. The reactions and rates depend on a range of factors including nutrient, oxygen and water availability, ground/pore water composition and temperature.
- Radiolysis is defined as the decomposition of chemical compounds by ionising radiation. Radiolysis can occur both within a waste package (from α-, β-, and γ-irradiation) and external to a waste package in the buffer or backfill and possibly in the host rock (due to γ-irradiation). Gases can be produced as products of the radiolysis process. The archetype is the decomposition of water, resulting in the production of hydrogen. In addition to hydrogen generated by radiolysis of water, gases such as hydrogen, carbon dioxide and methane can be generated from the radiolytic degradation of organic materials (for example cellulosic wastes, synthetic polymers, oils and small organic molecules).
- The amount of helium generated by radioactive decay will be small in terms of total bulk gas volume.

- The radiotoxic gases of potential importance are: tritium; gaseous molecules containing carbon-14, such as methane, carbon dioxide and carbon monoxide; and radon-222. Of these, carbon-14 is the only one with a sufficiently long half-life to be of post-closure interest from its presence in waste packages.

It should also be noted that many evaporite formations contain gypsum ($CaSO_4.2H_2O$) or other hydrous phases, which may evolve free water if temperatures become sufficiently high to cause thermal decomposition. In the case of gypsum this thermal decomposition can start at temperatures around 100°C. However, this process is unlikely to be important for the COVRA repository where it is anticipated that the maximum temperature will be around 70°C.

Consideration of gas generation during backfilling and after closure focusses on a cementitious repository where resaturation is rapid and water is freely available so is not relevant to a repository in salt. It is noted that repository environments with a low groundwater flow rate will limit the potential for gas generation. In an evaporite host rock, the gas generation rate will be more limited than for the other host rocks and the water contents of the waste packages at closure constrain the amount of gas that can be generated. This may be less than the water content at the time of packaging because of ongoing water-consuming reactions during waste package storage prior to transport to the repository.

RWM assume that if cementitious materials are present carbon dioxide reacts with them and is removed from the bulk gas. Carbonation may be relevant for COVRA because the wastes that are most likely to generate carbon dioxide are cement-encapsulated.

RWM's Carbon-14 Integrated Project

Lever et al. (2016) includes a description of modelling of gas generation from waste packages containing Magnox. Gas generation from these waste packages is water limited even for the 'wet' higher strength rock environment considered by RWM. The study also considers the effect of the low chloride water in the grout and backfill being displaced by chloride rich groundwater. The Quintessa model developed as part of this study is described in Section 4.2. A key feature of the model results is the influence of the assumptions about the behaviour of the ullage space and filter on the drying and then the later resaturation behaviour of the waste package.

For some assumptions, the waste package dries out to the extent that Magnox corrosion ceases (Figure 15), even for the wet environment assumed by RWM. For the case on the left, there is no suction in the ullage or filter, so the water pressure in the ullage / filter is equal to the gas pressure. The gas pressure in the ullage is higher than in the backfill because gas is generated within the package. The gas pressure in the ullage will build up until it exceeds the hydrostatic pressure in the backfill, at which point the water

pressure gradient would drive water out of the ullage and into the backfill. The ullage will therefore dry out and water will not be able to flow into the package. Corrosion consumes water in the package and eventually stops when the saturation reaches the residual saturation. For the case on the right there is a capillary suction in the ullage and filter so the water pressure is always less than the gas pressure and there will be a water pressure gradient that drives water flow into the package, even though this flow is small for the parameters considered. Gas generation continues until all the Magnox has corroded.

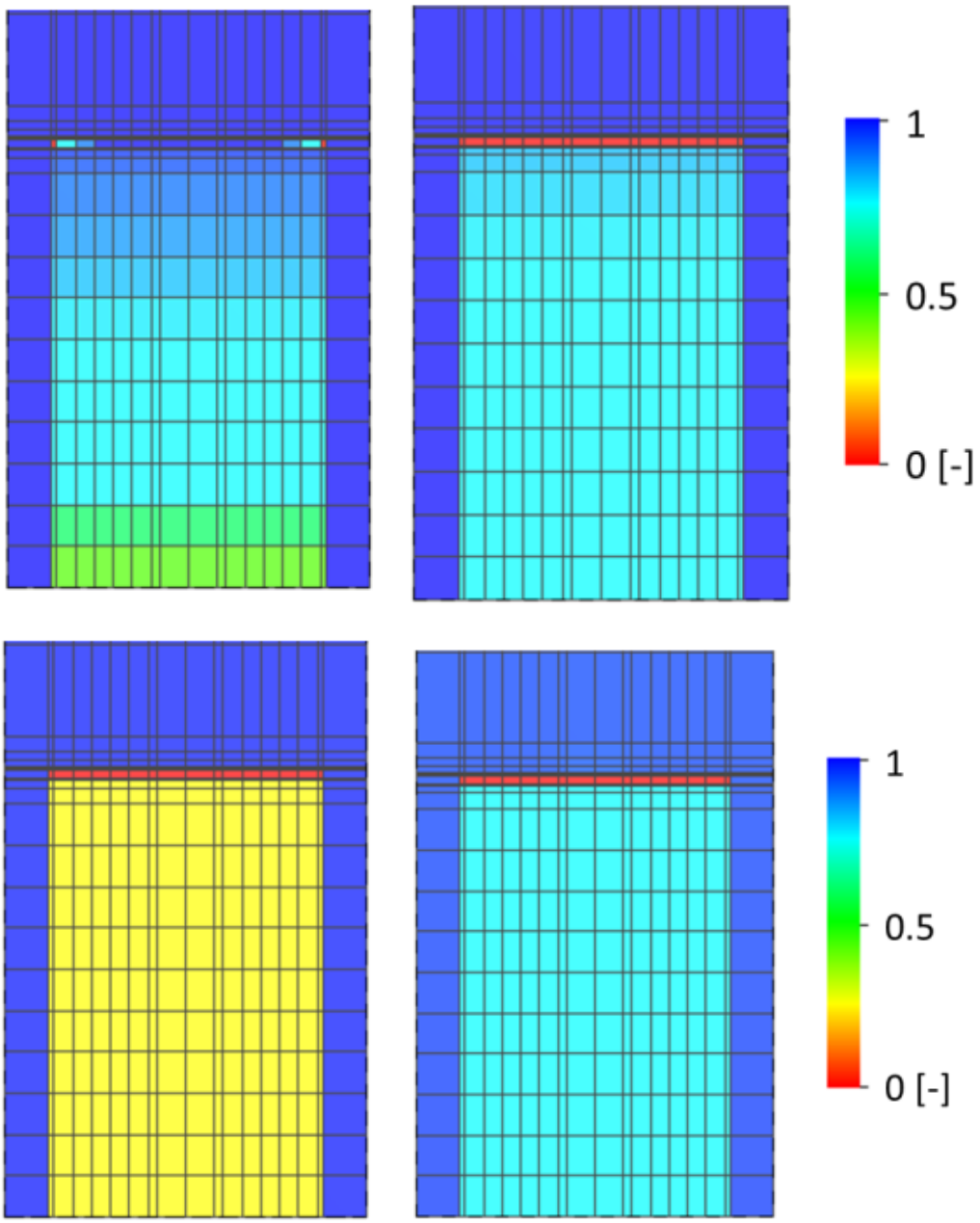

Figure 15: Water saturation 2.5 years (top) and 610 years (bottom) after backfilling and repository closure. No suction is assumed for the case on the left and weak suction is assumed for the case on the right. See also Figure 19 for evolution during

storage, evolution during operations continues the same trends. From Lever et al. (2016).

The study also considered the impact of saline groundwater entering the disposal vault and then the waste package. The timing and rate of gas generation was found to be sensitive to the parameterisation, and in many cases the corrosion became water limited for a period of time. Figure 16 shows results for a case with weak suction in the ullage and saline water entering the vault after backfilling.

AMEC carried out a parallel modelling study that provided similar conclusions to the Quintessa study.

While these results are for a wet environment, they illustrate some of the effects that can occur when the supply of water is not sufficient to support all the gas generating processes.

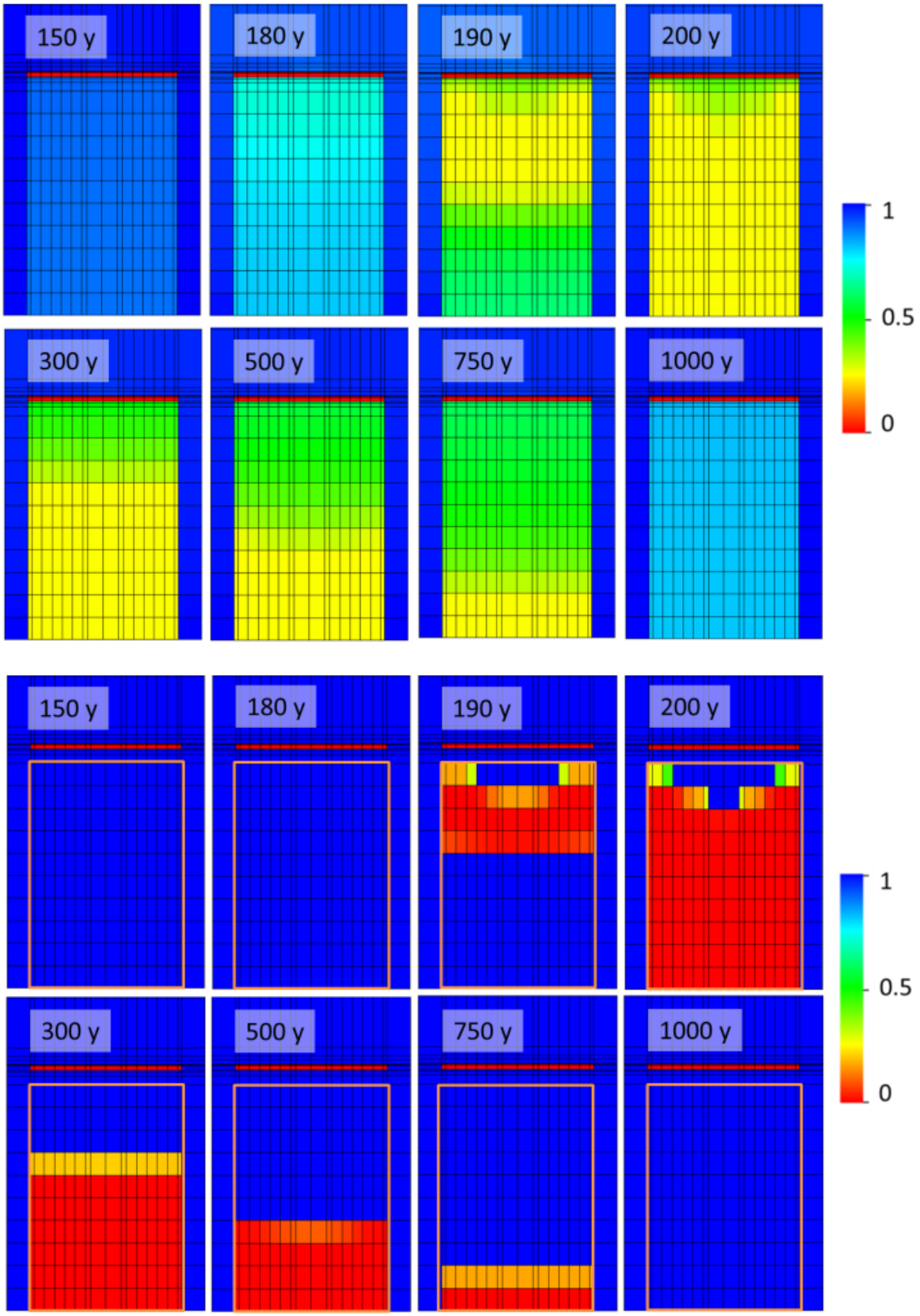

Figure 16: Water saturation (top) and fraction of potential corrosion rate (bottom) for a case in which there is weak suction in the ullage and saline water enters the vault. The repository is closed at 140 years.

Quintessa's Coupled Gas Generation and Migration Study

Watson et al. (2012) reproduced a model implemented in an earlier version of SMOGG than that described in Swift (2016) and used it to investigate gas generation in a range of geological environments, including an evaporite host rock. The study included the coupling of groundwater flow and gas generation/migration, which is not possible in SMOGG, to ensure that water limitation was taken into account in low permeability environments.

The study focused on gas generation as a result of corrosion, microbial degradation and radiolysis. It considered RWM's full 2010 waste inventory comprising L/ILW in a range of waste package types (all vented), depleted low and natural uranium (DNLEU in vented waste packages and high-level waste/spent fuel in fully sealed waste packages. The inventory did not include metallic spent fuel. The study considered gas generation from both waste and packaging materials. The key gas generating processes considered for each waste type were:

- L/ILW: Corrosion of metals, degradation of organics (cellulose and small organic molecules), methanogenesis, radiolysis of water, organics and polymers, release of radioactive gases including leaching from graphite, corrosion of waste containers, reaction of carbon dioxide with backfill.
- DNLEU: Radiolysis, Radon-222 from radioactive decay and corrosion of waste containers
- HLW/SF: Radiolysis, corrosion of overpacks. Gas generation from the wasteform was not considered.

It was assumed that L/ILW and DNLEU waste packages were 75% water-saturated when emplaced in the repository. Emplacement extended over a period of 100 years. The report notes that the time at which individual vaults are sealed to prevent ingress of oxygen and control of environmental conditions is lost is a key parameter for gas generation, and may be more important for gas generation than the actual repository closure date.

Calculations considered a single 'average' vault or tunnel and ignored any potential interactions between adjacent vaults/tunnels. Gas generation rates were scaled when the amount of water flowing into the vault or tunnel was not sufficient to support all the possible gas generation. All reactions were scaled equally i.e. no process was given priority for receiving water. Waste containers were not represented explicitly, but were instead applied as a flow resistance between backfill and waste.

It was assumed that the evaporite host rock was completely dry with only gas being able to move through the host rock. As a result, the total amount of gas that could be generated was limited by the amount of water that is present in the waste packages at closure or released by reactions in the waste packages. It was assumed that MgO backfill

is used in the L/ILW and DNLEU vaults and that this backfill can remove water from the system via hydration as well as absorbing carbon dioxide.

Creep closure was represented using a time and pressure dependent porosity in the buffer/backfill to simulate creep closure and the potential for creep to be reversed if gas pressures become sufficiently high. In the absence of gas generation, creep was assumed to take 700 years to complete for a L/ILW vault.

Volumes of gas were relatively small and the pressures did not exceed lithostatic. Figure 17 shows illustrative results for an ILW vault. The maximum over-pressure was about 4MPa, meaning that the maximum gas pressure was significantly less than lithostatic pressure. Hydrogen dominates the bulk gas but methane is also significant at early times. Carbon dioxide is more significant than for other host rocks because the MgO backfill must hydrate before it can start to react with carbon dioxide and remove it from the gas phase.

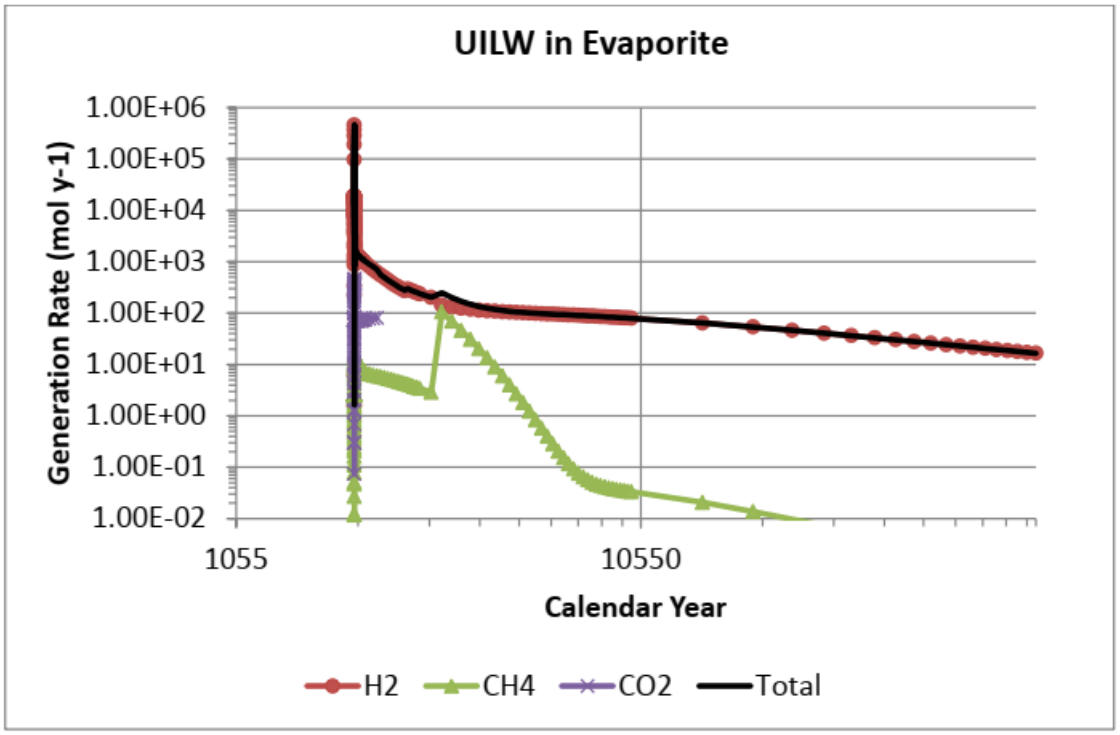

Figure 17: Rate of bulk gas generation for an ILW vault in evaporite for the coupled processes study (Watson et al., 2012).

Figure 18 shows results for a vault containing DNLEU in stainless steel drums. Bulk gas is generated as a result of radiolysis and corrosion of the inner surfaces of the drums. The gas generation rate increases with time as ingrowth increases the activity of the waste. The maximum overpressure for this case was less than 1 MPa and occurred just before creep ended.

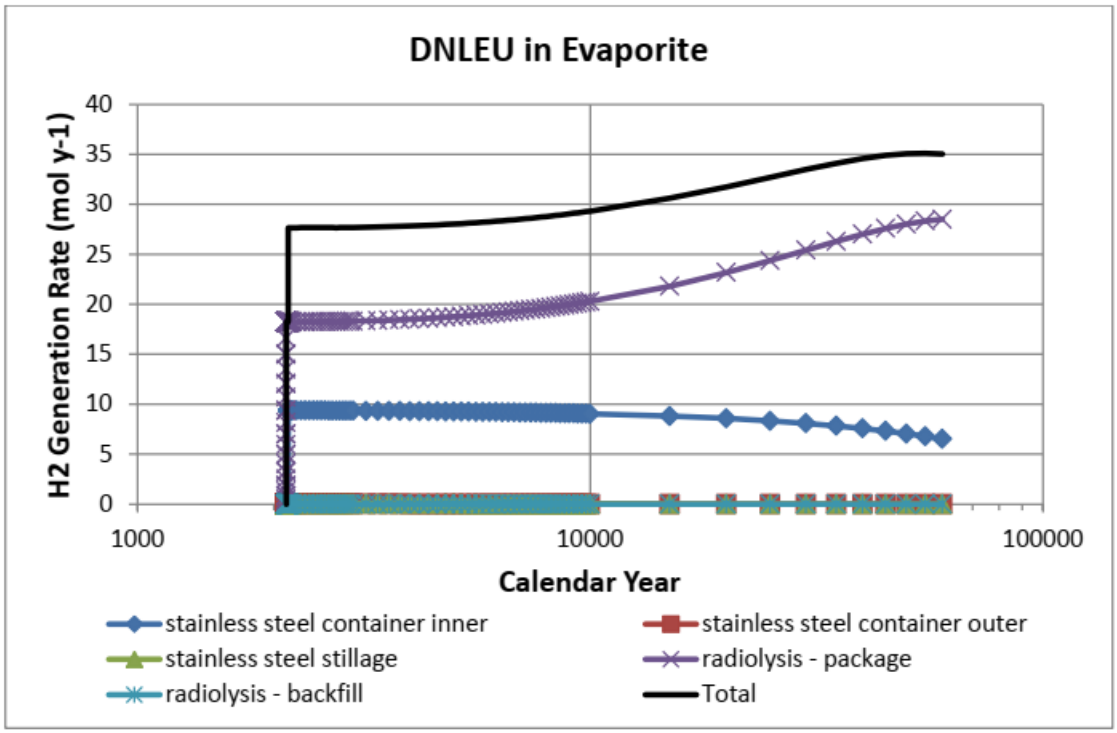

Figure 18: Bulk gas generation rate for a DNLEU vault in an evaporite for the coupled processes study (Watson et al., 2012).

Nagra

Nagra intends to construct a deep repository in the Opalinus Clay host rock. While, not an evaporite, this is expected to be a low flow environment and a key concern is the potential for over-pressurisation of the near field. It is therefore of some relevance to COVRA. The most recent major review of gas generation processes was published in 2016 and is summarised in Diomidis et al. (2016). The study considered both the production and the consumption of gas in the near field. It considered both L/ILW in a cementitious environment and HLW/SF surrounded by a bentonite buffer.

The gas generation processes considered were degradation of organic materials and corrosion of metals. Radiolysis was not included as it was judged that the volume of gas generated would be significantly smaller than for the other two processes. Gas consumption processes were chemical reactions such as carbonation and microbial processes such as methanogenesis. It is also assumed that conditions are anoxic.

Organic material is considered to degrade by a combination of hydrolysis to break down large molecules and microbial degradation of smaller organic molecules to generate carbon dioxide and methane. The production of other gases such as NH_3 and H_2S from materials containing nitrogen or sulphur (e.g. bitumen) was also taken into account. Organic materials are divided into low (ethanol, cellulose etc) and high (resins, PVC etc)

molecular weight groups, which are assigned different degradation rates. Low molecular weight organics degrade quickly, using a rate based on data for cellulose, and high organic weight organics degrade more slowly, using a rate based on data for bitumen. The microbial degradation part of the process stops when the water activity drops below a specified value.

The corrosion model considers general corrosion of iron and carbon steel, stainless steel and nickel alloys, Zircaloy, Aluminium, copper, lead, zinc and magnesium. Copper only corrodes in the presence of sulphide. All of these reactions generate hydrogen gas. Corrosion rates for iron-based materials are selected for high- and low- pH conditions to reflect the differences between L/ILW and HLW disposal concepts and the potential for carbonation of cementitious grouts and backfills.

The local gas consumption mechanisms that are considered are consumption of hydrogen by microbes, production of H_2S, methanogenesis, reaction of H_2S with iron and carbonation. The most significant processes are considered to be microbially mediated oxidation of hydrogen by sulphate (as the Opalinus Clay porewater has a high sulphate content) and methanogenesis. It is noted that the gas sinks may not be at the same locations as the gas sources.

The inventory for gas generation comprises the waste materials, the waste containers and construction materials that remain after closure. For the inventory considered by Nagra, hydrogen from metal corrosion is the dominant gas. All carbon dioxide is assumed to be consumed by reactions and a small amount of methane is produced. Other gases are negligible.

OPERA

OPERA (Box 6-2 of Verhoef et al., 2017) considered the following gas generation processes:

- Alpha decay leading to helium production
- Radiolysis of porewaters leading to hydrogen and oxygen production
- Degradation of organic materials generating carbon dioxide and methane
- Corrosion of metals generating hydrogen.

Corrosion was considered to be the most significant post-closure gas generation process for the OPERA system. Alpha decay and radiolysis were considered to produce negligible amounts of gas and microbial activity was expected to be limited by inhospitable near-field conditions. Conditions in the near field were assumed to be reducing and alkaline. Alkaline conditions result from grouting and use of cementitious liners in some waste packages, some concrete waste packages and cementitious material in overpacks.

4.1.3 Summary

The studies that are summarised in Section 4.1.2 cover the UK and Swiss programmes, but can be considered to be wider reviews since these two programmes draw on wider knowledge and participation in international programmes. There is general agreement that

- The most important gas generation processes are general corrosion and microbial degradation of organic materials.
- Radiolysis may be important for higher activity materials.
- The dominant gas is hydrogen, with smaller amounts of methane. Carbon dioxide is generally assumed to react in the near field.
- Anoxic conditions are established soon after closure so it is often conservatively assumed that there is no oxic period post-closure.
- In low permeability environments or where water access to the waste is restricted for some reason, gas generation may be limited by the amount of water available. The amount of water that is present in the waste packages at closure is very important for these cases.
- Additional processes such as diffusive release may need to be considered if the release of radioactive gases from the waste packages is to be characterised. The data required to parameterise many of these processes is poorly known.

4.2 Storage/Operational Phase

4.2.1 Environmental Conditions

It is generally assumed that environmental conditions can be controlled during the storage and operational phases, although the degree of control depends on the type of store and the environmental controls in place prior to repository closure. The key features of the expected environmental conditions during storage/operations are:

- Oxygen present so while conditions are not strongly oxidising, it is unlikely that reducing conditions will be present on the outsides of the waste packages. Reducing conditions may develop within individual waste packages if processes inside the packages consume oxygen faster than it is able to enter the package.
- Water availability is limited/controlled. It is assumed that the waste is stored under cover and pond storage is not considered. In the majority of cases, the waste packages are stored in humidity-controlled conditions that are designed to prevent or minimise deliquescence onto the waste packages. Some waste packages contain free water, for example as a side effect of the encapsulation process, which may be consumed during this period, but it is unlikely that significant water would enter a waste package. Vented packages may lose water via evaporation during storage.

- Water composition within waste packages will be determined by the waste/encapsulant. The composition of any water that deliquesces on the outside of waste packages will be determined by local conditions (e.g. how close the store is to the coast) but may have high concentrations of salts such as chlorides, nitrates and sulphates.
- Temperature is controlled. The environmental controls in the majority of stores ensure that temperatures are kept within the range that limits/prevents deliquescence and potentially also reactions within the waste packages. The temperature control may involve either heating or cooling depending on the waste involved and the weather.
- Pressure is atmospheric. Waste packages are stored at atmospheric pressure, which allows gas to vent freely if the packages are designed to allow gas to escape. The ventilation system may include measures to control the gas composition within the store or vault, for example to ensure that the hydrogen content remains below the explosive limit.

The time at which individual vaults/tunnels are closed meaning that control of environmental conditions is lost and oxygen is excluded, rather than the repository closure time, is key for gas generation. The operational schedule is therefore a key input to an assessment of gas generation. The term closure needs to be defined carefully for such studies.

4.2.2 Corrosion Processes and Assumptions

RWM's SMOGG Model

RWM's SMOGG (Swift, 2016) can be used to represent the behaviour of ILW and LLW during the period before repository closure. The following phases of waste management are discussed:

- Encapsulation. Some wastes may degrade prior to encapsulation, reducing the potential for gas generation. High temperatures, the availability of water and changes in chemical environment during the encapsulation process may cause a spike in corrosion and other processes.
- Interim storage: RWM consider that there will not be significant gas generation during interim storage owing to the controlled conditions in the store but do note that the inventory of potentially gas generating materials may be reduced during this period, which should be accounted for in later phases. Conditions will be oxic and the humidity will be controlled meaning that the water content of the packages is likely to decrease with time as a result of radiolysis, corrosion and cement hydration.
- Transport: Transport conditions are assumed to be similar to storage conditions.

- Operational period: Conditions are assumed to be oxic and temperature and humidity will be controlled. Packages are expected to continue to lose rather than gain moisture.
- Backfilling and immediately after closure: This is a key period for gas generation for RWM's concept due to water availability and the temperature increase associated with backfill curing. Curing of a cementitious backfill is unlikely to be relevant for a COVRA repository in salt as any backfill is likely to be MgO and/or crushed salt. The presence or otherwise of backfill may however be important for determining the near-field volume that is available to accommodate any gas that is generated.

The assumption of oxic conditions significantly reduces the potential for gas generation, even under conditions where water is available.

For the processes included in SMOGG, corrosion of Magnox and Aluminium, some microbially mediated degradation and the production of Radon are likely to be the most important.

During storage/operations, SMOGG assumes that gas can only be generated in vented waste packages. It is assumed that sufficient oxygen is always available. There is no coupling between different waste packages in terms of water availability. Three options are available:

- The amount of water available for gas generating processes is specified and these processes stop once it has been used up.
- Additional water can be added at a specified time and is used up in the same way as the initial water.
- Once a package has dried out, water can be made available at a specified maximum rate. If this rate is not sufficient to support all the water-consuming processes, these are scaled. All the water that is introduced during a timestep must be consumed during the timestep.

RWM's 2016 Gas Status Report

RWM (2016) provides a fuller description of the potential for gas generation prior to repository closure. It notes that tritium can be released from wastes as tritiated hydrogen, tritiated water or tritiated hydrocarbons/organic compounds. Much of the inventory of tritium will decay during the period of surface interim storage and during the operational phase of the repository.

The report considers gas generation during transport and operations, but not during interim storage, which is outside RWM's scope. Gas generation during transport is negligible in terms of overall gas production because the analysis considers a time period of 28 days. During operations, there is a need to manage the gas that is generated and released from vented waste packages. The gases that need to be managed are mainly

flammable (for example hydrogen), non-radiological chemotoxic (for example amines) and radiotoxic gases (for example those containing carbon-14). These gases are the same as those requiring management during interim surface storage and as for interim storage are managed by controlling environmental conditions and appropriate ventilation. RWM note that grouted depleted and low enriched uranium is likely to generate hydrogen through radiolysis of water and will be a major source of Radon-222.

RWM (2016) provides an illustration of the gas generation rate for RWM's L/ILW inventory during the operational period, which is relevant to COVRA since it is the period before backfilling and emplacement of backfill. Gas generation is dominated by corrosion of reactive metals and becomes water limited in the relevant waste packages once the water in the encapsulant has been consumed. Oxic corrosion of other metals does not generate bulk hydrogen gas and does not exhaust the available water but does release Carbon-14 labelled methane. Radon-222 becomes increasingly important as the volume of radium-containing wastes increases during the emplacement period.

RWM's Carbon-14 Integrated Project.

One component of the work reported in Lever et al. (2016) considered the evolution of waste packages containing Magnox swarf. Water availability is a key concern for these packages owing to the high corrosion rate and hence high gas generation rate, specially when the waste packages are subjected to elevated temperatures associated with backfilling and water that has a high pH and is potentially saline. The work considered the evolution of the water content in the waste package during a 60-year period of interim storage and 80 years of repository operations. It was important to estimate the water content and inventory of Magnox remaining at repository closure to generate a realistic inventory at the start of the post-closure period. Quintessa's modelling is described below.

At the start of the surface storage period, the encapsulant and capping grout within the package will be nearly saturated. During storage of the waste package in a surface store and underground in a disposal vault, the contents of the package will dry as water vapour moves out of the package through the filter. Experiments on 4 m box waste packages containing LLW indicate that the relative humidity in the ullage is likely to be similar to the relative humidity in the store or vault. During the storage period, gas will be generated within the package and flow out of the package via the ullage and filter.

The model considered two phase flow of gas and water, including the transport of dissolved gas and water vapour, and the corrosion of the Magnox metal. The corrosion rate was coupled to the availability of water and to temperature and chloride content. As corrosion requires free liquid water to be available, the rate was coupled to the saturation of the grout, not the relative humidity of the storage environment. Consumption of water by corrosion meant that it was possible for the grout saturation

to be lower than that implied by the relative humidity. The irreducible saturation of the encapsulant was 0.25, whereas the relative humidity of the storage environment was 70%.

The initial saturation of the grouts was taken to be 0.95, which takes into account mineral formation consuming some of the free water as the cement cures. The initial water content of the encapsulant is sufficient to corrode 35-40% of the Magnox if it is all available to participate in the corrosion reaction.

The model is sensitive to the way in which the ullage and filter are parameterised and several variants were considered ranging from fixing the relative humidity at 70% outside the package to fixing it at 70% in the ullage. Figure 19 illustrates the range of results and indicates that for some assumptions there will be significant drying of the encapsulant during storage and for other cases there is very little drying. Both cases include water consumption due to corrosion of the Magnox waste. These differences are further accentuated during the operational period and result in different initial conditions for the post-closure period. The impact of the different assumptions results in very different resaturation and gas generation behaviours even in the high water availability environment assumed in the study.

A key conclusion from this work is that storage for 140 years at a relative humidity of 70% does not result in sufficient drying to inhibit corrosion for these vented waste drums.

AMEC carried out a parallel modelling study that provided similar conclusions to the Quintessa study.

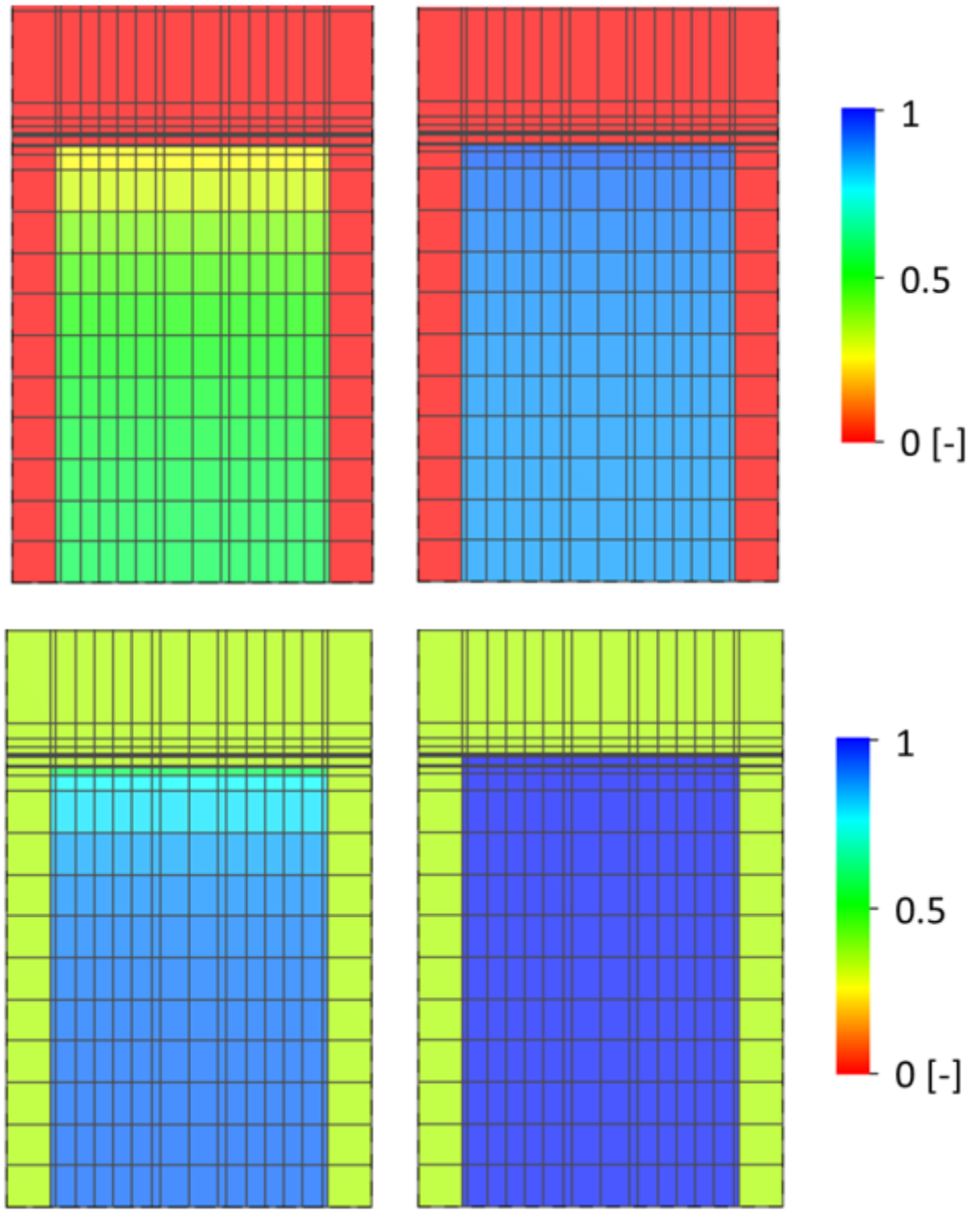

Figure 19: Water saturation (top) and relative humidity (bottom) after 60 years of storage for cases with relative humidity in the ullage and filter fixed and no suction (left) and relative humidity fixed outside the package and calculated in the ullage and filter assuming weak suction. The initial water saturation is 0.95 and the humidity of the store is 0.7. From Lever et al. (2016).

Quintessa's Coupled Gas Generation and Migration Study

Watson et al. (2012) considered gas generation during repository operations but did not include the prior period of interim storage. It was assumed that the water saturation in

encapsulants was 0.75 at the start of the calculations to take account of drying during long-term storage but the inventory of gas-generating materials was not reduced to account for consumption during the storage period. The end of the operational period for each vault was taken as being the time at which the vault was sealed and control over environmental conditions was lost (i.e. ventilation system closed down). Any spike in gas generation associated with backfilling the vault was included. The results were calculated on a vault by vault basis and then summed, taking into account the different operational periods of the different vaults. The summation approach worked well for gas generation processes where radioactive decay does not contribute but required corrections to be applied for processes such as radiolysis where radioactive decay has to be taken into account and the relevant components of the inventory changed on a timescale that was comparable to or shorter than the operational period.

It has been observed that moving a waste package often results in a brief period of increased gas generation. This is interpreted as a resetting of the acute corrosion step (see the description of SMOGG in Section 4.1.2) and was included in the modelling. The precise mechanism is not clear but it is likely to be associated with either disruption of passivating layers on metal surfaces or movement of water within the encapsulant as a result of shaking the package etc.

Nagra

The Nagra work (Diomidis et al., 2016) does not consider gas generation during the storage and operational phases. Instead, it conservatively assumes that all the gas generating inventory is available at the start of the anoxic part of the post-closure phase.

OPERA

OPERA (Verhoef et al., 2017) does not consider gas generation during the storage and operational periods. It is however noted that the outer surfaces of the cement grout in the 200 l LILW drums may become carbonated.

4.2.3 Summary

The studies described in Section 4.2.2 indicate that less attention has generally been paid to gas generation during storage and operations than to post-closure. In most cases, it is conservatively assumed that all the gas source material is present at repository closure.

- Conditions during storage and operations are oxic, which reduces the potential for gas generation.
- The controlled environmental conditions during storage and repository operations are designed to minimise waste package degradation so the only degradation expected is for wasteforms that contain water and container surfaces in direct contact with them.

- The dominant bulk gas is hydrogen.
- It is expected that vented waste packages will dry out to some extent during storage but it seems likely that they will not dry out to an extent that stops the degradation reactions due to lack of water. The extent of drying in the models is very sensitive to assumptions about the interface between the wasteform and the store/repository atmosphere.

5 Processes and Assumptions to Include in this Study

This section provides recommendations for the gas generation processes and assumptions that should be considered in the simple calculations to be carried out for the COVRA inventory in a salt host rock. First, a summary of processes and overall assumptions is provided and then each of the waste groups defined in Section 3.2 is considered in turn.

The functional specification described by Benbow et al. (2023a) is based on these recommendations. The functional specification makes some simplifications and approximations to reflect the level of data available to parameterise the model and the practicalities of implementing it in a simple tool. Additional detail can be added at a later date if the results indicate that it would be useful.

5.1 Overall Assumptions

The information summarised in Section 4 indicates that the following degradation processes that have the potential to generate bulk gas need to be considered:

- General corrosion of metals;
- Degradation of organic materials;
- Radiolysis for the higher activity waste groups (high beta/gamma activity).

If calculation of gas generated by radiolysis turns out to be time-consuming, some scoping calculations may be required to determine which waste groups might generate sufficient gas by radiolysis to mean that this process should be included.

During the storage/operational period the following assumptions are recommended:

- Conditions are oxic.
- Relative humidity is 60% (Verhoef et al., 2016, states < 60%) and temperature as specified in environmental controls.
- No degradation of outer surfaces of waste packages.

- Degradation occurs for water-containing wasteforms and metal surfaces in contact with them. Gas is released for vented packages and fully sealed packages become slightly pressurised.
- Generation of radioactive gases, including Radon-222 is not included in this initial scoping study.

During the post-closure period the following assumptions are recommended:

- The post-closure period is taken to start at the time the vault or tunnel is closed and environmental control ceases, which may not be the same as the date of repository closure.
- Conditions are anoxic. The amount of oxygen trapped at closure is likely to be small and consumed rapidly.
- Relative humidity is 100% meaning that the external surfaces of all waste packages can corrode, provided sufficient water is available
- Temperature equal to rock temperature at repository depth. Salt is a good conductor of heat and the heat output of heat-generating HLW is expected to decay significantly before the containers fail. Alternatively, a predefined variation of temperature with time could be imposed.
- Wasteforms in sealed containers continue to degrade until the water is exhausted. Once the container fails this gas is released and degradation restarts.
- The voidage of the excavations should be able to be varied with time to reflect compaction of any backfill and convergence of the host rock. For the initial scoping calculations carried out in this project, a constant void volume for gas could be considered as pressure should vary linearly with void volume if gas is not allowed to escape from the near field.
- Generation of radioactive gases, including Radon-222 is not included, at least in the first round of calculations.

5.2 Breakdown by Waste Group

This section considers the different waste groups proposed in Table 3. It is expected that gas generation will be determined on a waste package basis. Determining the overall gas generation rate will require the number of waste packages in each group and information on the length of the storage period and closure times for each waste group.

5.2.1 Vitrified Waste

Storage/Operations

Vitrified waste is stored in CSD-v stainless steel containers. No degradation or gas generation is expected during the storage period as the containers are sealed, the wasteform is dry and the external environment is controlled to prevent corrosion.

Post-closure

The CSD-v containers are expected to be overpacked for disposal using the carbon steel overpack shown in Figure 3. This overpack is initially fully sealed and will prevent water from coming into contact with the outer surface of the CSD-v container until such time as the overpack fails due to corrosion and/or mechanical overload. Conditions are expected to be anoxic by this time so gas will start to be generated when the overpack fails. The radiation field is likely to be sufficient to mean that radiolysis of water around the package needs to be considered. Once the CSD-v container fails, corrosion of its inner surface will start but degradation of the vitrified waste will not generate gas unless the CSD-v container fails sufficiently early that radiolysis is still a concern.

Key Waste Package Data

Design of overpack and time at which it fails

Surface area and thickness of CSD-v container

Corrosion rate for stainless steel

G Values for water

Radionuclide inventory

5.2.2 Research Reactor Spent Fuel

The research reactor spent fuel has been divided into two groups (HEU and LEU) because these groups will have different parameters. However, the processes will be the same so they are treated together in this section.

Storage/Operations

Research reactor spent fuel is stored in ECN stainless steel containers. No degradation or gas generation is expected during the storage period as the containers are sealed and the external environment is controlled to prevent corrosion. It is assumed that there is no carried over water associated with the spent fuel so the waste is dry.

Post-closure

The ECN containers are expected to be overpacked for disposal using the carbon steel overpack shown in Figure 3. This overpack is initially fully sealed and will prevent water from coming into contact with the outer surface of the ECN container until such time as the overpack fails due to corrosion and/or mechanical overload. Conditions are expected to be anoxic by this time so gas will start to be generated when the overpack fails. The radiation field is likely to be sufficient to mean that radiolysis of water around the package needs to be considered. Once the ECN container fails, corrosion of first the capsule and then the dominantly aluminium spent fuel is expected and corrosion of the

inner surface of the ECN container will start. Corrosion of the Aluminium components will be rapid if the porewater is alkaline. If the ECN container fails sufficiently early radiolysis may still be a concern.

Key Waste Package Data

Design of overpack and time of failure

Surface area and thickness of ECN container

Corrosion rate for stainless steel

Material, surface area and thickness for capsule surrounding spent fuel

Corrosion rate for capsule

Surface area for spent fuel

Mass/average thickness for spent fuel

Corrosion rate for spent fuel (dominantly Al).

G Values for water

Radionuclide inventory

5.2.3 Uranium Collection Filters

Storage/Operations

Uranium collection filters are dried and then stored in aluminium tubes which are placed in ECN stainless steel containers. No degradation or gas generation is expected during the storage period as the waste is dried, the containers are sealed and the external environment is controlled to prevent corrosion. Any gas that is generated by corrosion caused by residual water due to incomplete drying would be retained inside the ECN container.

Post-closure

The ECN containers are expected to be overpacked for disposal using the carbon steel overpack shown in Figure 3. This overpack is initially fully sealed and will prevent water from coming into contact with the outer surface of the ECN container until such time as the overpack fails due to corrosion and/or mechanical overload. Conditions are expected to be anoxic by this time so gas will start to be generated when the overpack fails. The radiation field is likely to be sufficient to mean that radiolysis of water around the package needs to be considered. Gas generation inside the ECN container will continue until all of the carried over water has been consumed. Once the ECN container fails, any gas that is inside it will be released. Corrosion of the aluminium cartridge is expected to begin, which will be rapid if conditions are alkaline, and corrosion of the

inner surface of the ECN container will start. Once the cartridge fails, the filter will corrode and this may be rapid due to the high surface area. The filter body will corrode more slowly. If the ECN container fails sufficiently early, radiolysis may still be a concern.

Key Waste Package Data

Design of overpack and time of failure

Surface area and thickness of ECN container

Corrosion rate for stainless steel

Initial water content (carried over water)

Surface area and thickness for cartridge containing filters

Corrosion rate for cartridge (Al)

Surface area and thickness for filter body

Corrosion rate for filter body (assumed stainless steel)

Surface area for filter

Mass/average thickness for filter

Corrosion rate for filter (porous stainless steel structure).

G Values for water

Radionuclide inventory

5.2.4 Reprocessing Waste

Storage/Operations

Reprocessing waste is stored in CSD-c stainless steel containers. No degradation or gas generation is expected during the storage period as the containers are sealed and the external environment is controlled to prevent corrosion. It is assumed that there is no carried over water.

Post-closure

The CSD-c containers are expected to be overpacked for disposal using the carbon steel overpack shown in Figure 3. This overpack is initially fully sealed and will prevent water from coming into contact with the outer surface of the CSD-c canister until such time as the overpack fails due to corrosion and/or mechanical overload. Conditions are expected to be anoxic by this time so gas will start to be generated when the overpack fails. The radiation field is likely to be sufficient to mean that radiolysis of water around the package needs to be considered. Once the CSD-c container fails, corrosion of the

inner surface of the CSD-c container and the reprocessing waste will start. If the CSD-c container fails sufficiently early, radiolysis may still be a concern.

Key Waste Package Data

Design of overpack and time of failure

Surface area and thickness of CSD-c container

Corrosion rate for stainless steel

Surface area and thickness of reprocessing waste (assumed to be dominantly Zircaloy)

Corrosion rate for reprocessing waste

G Values for water

Radionuclide inventory

5.2.5 HLW Technical Waste – OPERA Concept

Storage/Operations

HLW technical waste is stored in ECN stainless steel containers. If it is assumed that the ECN container is welded shut for storage, then no gas can escape during storage. There is potential for gas generation in the sealed ECN container because of the concrete lining and the potential for water to be associated with wastes such as organic material. However, it is assumed that this gas cannot escape and the ECN container becomes pressurised.

Post-closure

The ECN containers are expected to be overpacked for disposal using the carbon steel overpack shown in Figure 3. This overpack is initially fully sealed and will prevent water from coming into contact with the outer surface of the ECN container until such time as the overpack fails due to corrosion and/or mechanical overload. Conditions are expected to be anoxic by this time so gas will start to be generated when the overpack fails. The radiation field is likely to be sufficient to mean that radiolysis of water around the package needs to be considered. Once the ECN container fails, any gas generated prior to ECN container failure will be released, and corrosion of its inner surface and degradation (corrosion and microbial degradation) of the remaining waste begins. The inner liner and the mild steel drums that are compacted to form the waste pucks will also corrode and generate gas. If the ECN container fails sufficiently early radiolysis may still be a concern.

The ECN container is concrete lined and the waste pucks are grouted (Figure 7), presumably for shielding, so the porewater in the failed ECN container should be conditioned to be at least mildly alkaline. Prior to failure of the ECN container, gas

generation will continue until the water has been exhausted but this gas will not be released.

The waste in the ECN container is mixed waste including neutron activated metals from dismantled experiments, fuel cladding (probably aluminium if it relates to research reactors), and organic materials. These will generate gas as a result of corrosion of the metal inventory and degradation of organics. The organic material consists of plastic foils, tissues (paper), cloths used to clean hot-cell filters and so-called 'table-cloths' previously made from PVC, to collect the rubble and debris during cutting, sawing and other mechanical operations in the hot-cell. Given the neutron activation etc, radiolysis may still be a concern at the time that the ECN container fails.

It may be sensible to consider waste packages with a single type of waste (e.g. only metals or only organics) in addition to generating an 'average' waste package.

Key Waste Package Data

Design of overpack and time of failure

Surface area and thickness of ECN container

Corrosion rate for stainless steel

Initial water content for ECN container concrete lining

Material type (probably stainless steel), surface area and thickness for activated metal waste

Material type (probably aluminium), surface area and thickness for cladding waste

Corrosion rates for metallic wastes

Composition of organic waste

Rate constant for degradation of organic waste

G Values for water, concrete, organic materials and overpack material

Surface area and thickness for liner and mild steel drum that was compacted to form puck

Corrosion rate for mild steel

Initial water content for technical wastes

Radionuclide inventory

5.2.6 HLW Technical Waste – DDS Concept for Legacy Waste

Storage/Operations

This portion of the HLW technical waste will be placed in stainless steel DDS drums without compaction. There is potential for gas generation in the DDS container because of the potential for water to be associated with wastes such as organic material. These containers have bolted lids and so will not be completely gas tight, although slight pressurisation may be possible prior to failure of the rubber seals. However, it is assumed that the seals fail during the storage period and this gas is able to escape prior to emplacement in the repository.

Post-closure

The DDS drums are expected to be overpacked in concrete boxes for disposal. These overpacks mean that the water that comes into contact with the DDS drums and their contents will be alkaline. Gas will start to be generated once conditions become anoxic. The radiation field is likely to be sufficient to mean that radiolysis of water in the overpack and in the DDS drum may also need to be considered. The DDS drums are not sealed so the waste and internal 'crinkle drum' will degrade and the drums will corrode from both sides as soon as water becomes available.

The waste in the DDS drum is mixed waste including neutron activated metals from dismantled experiments, fuel cladding (probably aluminium if it relates to research reactors), and organic materials. These will generate gas as a result of corrosion of the metal inventory and degradation of organics, The organic material consists of plastic foils, tissues (paper), cloths used to clean hot-cell filters and so-called 'table-cloths' previously made from PVC, to collect the rubble and debris during cutting, sawing and other mechanical operations in the hot-cell. Given the neutron activation etc, radiolysis may need to be considered.

It may be sensible to consider waste packages with a single type of waste (e.g. only metals or only organics) in addition to generating an 'average' waste package.

Key Waste Package Data

Design of concrete overpack if it to be considered when deriving void space associated with package

Surface area and thickness of DDS drum

Corrosion rate for stainless steel

Material type (probably stainless steel), surface area and thickness for activated metal waste

Material type (probably aluminium), surface area and thickness for cladding waste

Corrosion rates for metallic wastes

Composition of organic waste

Rate constant for degradation of organic waste

G Values for water, concrete, organic materials and overpack material

Initial water content for technical wastes

Radionuclide inventory

5.2.7 HLW Technical Waste – KONRAD II Container Concept for Decommissioning Waste

Storage/Operations

This portion of the HLW technical waste will be stored the KONRAD II containers, but it is not clear whether these will be used for disposal or whether an alternative container will be used. Regardless of container type, the storage container is assumed to be vented but is out of scope for the gas generation calculations. There is likely to be some water associated with the uncompacted waste so corrosion may proceed to a limited extent during the storage period.

Post-closure

The carbon steel KONRAD II container is out of scope for this study. It is vented so water is able to access the waste immediately. The waste in the KONRAD II container is mixed waste including neutron activated metals from dismantled experiments, fuel cladding (probably aluminium if it relates to research reactors) and decommissioning concrete. These will generate gas as a result of corrosion of the metal inventory. Given the neutron activation etc, radiolysis may need to be considered. The volume of concrete is not specified by COVRA but the waste stream description and associated volume suggest that there may be more concrete waste present than in the OPERA case. In the absence of confirmation of the mis of waste materials, conditions are conservatively assumed to be neutral.

It may be sensible to consider waste packages with a single type of waste (e.g. only metals or only concrete) in addition to generating an 'average' waste package.

Key Waste Package Data

Material type (probably stainless steel), surface area and thickness for activated metal waste

Material type (probably aluminium), surface area and thickness for cladding waste

Corrosion rates for metallic wastes

G Values for water and concrete

Initial water content for technical wastes

Radionuclide inventory

5.2.8 Depleted Uranium

Storage/Operations

The depleted uranium is assumed to be stored as a dry powder in DV70 containers. The only gas generation that is expected is production of Radon-222, which is not a bulk gas.

Post-closure

The depleted uranium will be conditioned with concrete and placed in carbon steel containers for disposal. These KONRAD II containers are not in scope. Hydrogen will be generated in the encapsulated wasteform as a result of radiolysis.

Key Waste Package Data

Encapsulation ratio (cement to depleted uranium ratio)

Grainsize of depleted uranium powder

Wasteform porosity

Initial saturation of wasteform

G values for encapsulated waste

Radionuclide inventory

5.2.9 Molybdenum Waste

Storage/Operations

The inner 200 l drum, mixing paddle and the steel reinforcement are expected to corrode very slowly during storage but it will not generate gas as the conditions are assumed to be oxic. It is likely that the volume of metal that corrodes under the expected alkaline conditions will be negligible so the full inventory of metal can probably be carried forward to the post-closure calculation. There may be some gas generated by radiolysis of water in concrete and grouted waste. The reinforced concrete container and grout encapsulated waste are likely to dry out during storage but not to the extent that corrosion ceases.

Post-closure

The metallic components of the waste package will corrode so long as there is sufficient water to support the corrosion reaction. Radiolysis will continue as long as there is water present and sufficient radiation field. The hydrogen gas that is generated should be able to escape through the walls of the concrete container. The supply of water to the inner parts of the waste package may be diffusion controlled due to the low permeability of the concrete 1000 l overpack, even if there is ample water available outside the container. As the waste package evolves, the concrete may crack as a result of expansive corrosion of the metal components, which will increase access of water to the metal components.

Key Waste Package Data

Surface area and thickness of steel reinforcement

Initial water content of reinforced concrete container

Surface area and thickness of 200 l drum

Surface area and thickness of mixing paddle

Corrosion rates for metals (galvanised/carbon and stainless steel)

Initial water content of grouted waste

G Values for cementitious materials

Radionuclide inventory

5.2.10 Non-compactible LILW

Storage/Operations

The inner 200 l drum, the steel reinforcement and any metal waste are expected to corrode very slowly during storage but it will not generate gas as the conditions are assumed to be oxic. Ion exchange resins may generate hydrogen as a result of radiolysis, depending on the inventory. The reinforced concrete container and grout encapsulated waste are likely to dry out during storage.

Post-closure

The metallic components of the waste package will corrode and resins degrade so long as there is sufficient water to support the reactions. although it is assumed that no gas results from degradation of resins. Radiolysis will continue as long as there is water present and a sufficient radiation field. The gas that is generated should be able to escape through the walls of the concrete container, which has sufficient permeability to prevent pressurisation. Carbon dioxide may react with the cementitious materials in the waste package. At early times when the waste package is intact, the supply of water to the

inner parts of the waste package may be diffusion controlled, even if there is ample water available. As the waste package evolves, the concrete may crack as a result of expansive corrosion of the metal components, which will increase access of water.

Radiolysis is considered to be the only gas generating process for the ion exchange resins.

It is noted that COVRA includes other LILW that is non-compactible in the 'compactible LILW' waste stream. Thus the non-compactible LILW waste stream considered in this study only includes ion exchange resins.

Key Waste Package Data

Surface area and thickness of steel reinforcement

Initial water content of reinforced concrete container

Surface area and thickness of 200 l drum

Surface area and thickness of mixing paddle

Material type, surface area and thickness for metal waste

Corrosion rates for metals (galvanised/mild and stainless steel)

Type and mass of resin

Degradation rate for resin

Initial water content of grouted waste.

G Values for cementitious material and resins

Radionuclide inventory

5.2.11 Compactible LILW

Storage/Operations

The contents and inner surfaces of the drums will degrade during storage. The amount of gas generated may be limited as a result of oxic conditions but the inventory of gas generating materials will be depleted. The inner surface of the drums and any metal waste will corrode and organic material will degrade. The grout will dry out but there is likely to always be sufficient water to support degradation reactions, despite the drums having no lids. Conditions will be alkaline. Radiolysis is unlikely to be important because this is LILW in simple packages.

Post-closure

Degradation, now including corrosion of the outer surface of the 200 l drum will continue so long as sufficient water is available. The grout will resaturate provided

sufficient water is available. More gas is likely to be generated than during the storage period as conditions are now anoxic. Radiolysis is unlikely to be significant.

These waste packages contain a variety of different types of waste, which will degrade at different rates. It may be sensible to calculate gas generation for drums containing a single type of waste as well as for an average drum.

Key Waste Package Data

Surface area and thickness of 200 l drum

Initial water content for concrete lining

Material type, surface area and thickness for metal waste

Corrosion rates for metallic wastes and packaging

Type and mass of organics

Degradation rate for organics

Type and mass of plastics

Degradation rate for plastics

Surface area and thickness for mild steel drum that was compacted to form puck

Corrosion rate for mild steel

Initial water content of waste pucks.

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