Carbon-14 Project Team Phase 2: Irradiated Steel Wastes

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Preface

This report has been prepared by Amec Foster Wheeler under contract to Radioactive Waste Management Limited (RWM) and forms part of an ongoing programme of research commissioned by RWM to underpin the long-term safety of a geological disposal facility for higher-activity radioactive wastes.

The report has been reviewed by RWM. The views expressed and conclusions drawn in this report are those of Amec Foster Wheeler and do not necessarily represent those of the RWM.

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Abstract

Carbon-14 is a key radionuclide in the assessment of the safety of a geological disposal facility for radioactive waste because of the potential impact of gaseous carbon-14 bearing species. Radioactive Waste Management has established a project team, in which the partners work together to develop an holistic approach to carbon-14 management in a geological disposal system. This report is part of a suite of reports produced as part of the project.

This report summarises the work related to irradiated steel wastes that has been completed as part of Phase 2 of the project. The focus of the work described in this report has been:

- To improve the understanding of the inventory of carbon-14 in stainless steel wastes, and in particular the inventory associated with steel wastes that would arise from new build reactors;
- To improve the understanding of the thickness of steel wastes;
- To review the corrosion rates of steels, which control the rate at which carbon-14 present in steels may potentially be released;
- To review current understanding of carbon-14 speciation in irradiated steels and the likely speciation of carbon-14 releases;
- To perform a generic assessment of the potential impacts of carbon-14 releases from waste steels in both the operational period of a geological disposal facility and after its closure; and
- To consider alternative options for management and disposal of steel wastes.
Executive Summary

Carbon-14 (half-life 5,730 years) is a key radionuclide in the assessment of the safety of a geological disposal facility (GDF) for radioactive waste. In particular, the radiological impact of gaseous carbon-14 bearing species has been recognised as a potential issue. Carbon-14 is expected to be released from a GDF over a timescale of several thousand years. A number of radioactive gases will be generated from waste materials within a GDF, with carbon-14 bearing methane ($^{14}$CH$_4$) likely to be the dominant carbon-14 species transported in the gas phase, potentially reaching the biosphere at low activity concentrations.

The main sources of carbon-14 are: irradiated graphite, irradiated steels, irradiated reactive metals and spent fuel. There are 17,700 TBq of carbon-14 in the 2013 Derived Inventory of which 7,060 TBq is associated with irradiated steel wastes.

This is one of a suite of reports that present the results of Phase 2 of RWM’s carbon-14 project. The project was established to develop an holistic approach to carbon-14 management in a GDF. The technical approach is informed by the key generation and migration processes that affect the fate of carbon-14 in the Derived Inventory. This report presents the work that has been completed during Phase 2 related to irradiated steel wastes containing carbon-14.

Carbon-14 is produced in irradiated steel components of nuclear reactors by the thermal neutron activation of nitrogen-14 impurities in the steel. In the majority of cases, the estimated carbon-14 activities of irradiated wastes in the Derived Inventory are based on neutron activation calculations using assumed nitrogen impurity concentrations for the steel materials concerned. The inventory of carbon-14 in stainless steel has increased in the 2013 Derived Inventory, with the inclusion of an illustrative fleet of new build reactors. However, the carbon-14 inventories of a number of important waste streams from legacy reactors have decreased as a result of improved understanding of the nitrogen impurity concentrations in these steels. This has resulted in the carbon-14 inventory in legacy steel wastes being reduced by over 50%.

In the calculations of gas generation performed in this work, carbon-14 is assumed to be released from the steels at a uniform rate as the wastes corrode. This leads to modest release rates of carbon-14 bearing gases from these wastes during both the operational and post-closure periods. Releases are sensitive to the thickness of the steel; the understanding of waste component dimensions has improved for some of the legacy waste streams and thickness data have been refined as part of the Phase 2 work.

As part of the international CAST project, a detailed review of steel corrosion rates under disposal conditions has been undertaken. The results of that review are summarised in this report. Although much of the information collected has supported the corrosion rates used in earlier work, additional data have become available on the anaerobic corrosion of stainless steel, and the corrosion rate has been reduced by over an order of magnitude. This has implications for the rates of generation of bulk gas and for the rates of release of carbon-14 from activated steels.

Based on a simple congruent source term model for carbon-14 release, release rates of carbon-14 bearing gases have been calculated through the various stages of a GDF from waste emplacement during the operational phase through backfilling (with a cementitious material) and closure of the vaults, to the long-term post-closure period.

The potential consequences of these releases to a locally resident exposed group of humans have been assessed. The pathway considered for release from the GDF during the operational period is gaseous discharge from a 15m high ventilation stack. For the post-closure period, a number of migration cases have been developed that are illustrative of potential gas migration behaviour through the geosphere to a release area at the surface. The effective doses have then been calculated by multiplying the carbon-14 discharges by effective dose factors (which depend on the assumed speciation of the carbon-14 and the mechanism of carbon-14 exposure (inhalation or ingestion from crops)).
In the absence of suitable experimental data, there is considerable uncertainty about the chemical form(s) in which the carbon-14 will be released from irradiated steels. It is possible that the carbon-14 may be released as an aqueous species, be retained in solid phases or be released to a gas phase. Therefore in the gas generation calculations, no speciation is assigned to the carbon-14 released from steel wastes. Different assumptions about the gas phase speciation are scoped in the biosphere to assess the potential consequences of the carbon-14 releases:

- in the operational phase assessment, it is cautiously assumed that all of the carbon-14 release occurs to the gas phase; two cases are considered in which carbon-14 is released as either 100% methane or 100% carbon monoxide;
- in the post-closure Reference Case, it is assumed that 10% of the carbon-14 is released to the gas phase as methane; two variant cases have also been developed where either 100% or 1% of the carbon-14 is released from steels as methane gas.

There is a lack of corrosion rate data for irradiated steels. Therefore, the Reference Case calculations performed in this study have used corrosion rate data measured for unirradiated materials. However, it is known that some stainless (but not mild) steels subjected to neutron irradiation may undergo sensitisation due to radiation-induced segregation, which may increase their susceptibility to intergranular attack. Intergranular corrosion may lead to stress corrosion cracking of the steel matrix along grain boundaries, which may increase the surface area of steel available for general corrosion. Therefore, a variant case has been developed for irradiated stainless steel wastes in which higher effective corrosion rates (by a factor of ten) are assumed under all conditions, to account for uncertainties in the corrosion rates of irradiated materials and the potential effects of increased surface area due to intergranular stress corrosion cracking (IGSCC) on the general corrosion of these wastes and consequental rates of carbon-14 release. This case is considered to be a cautious treatment of these potential effects, because not all stainless steel wastes will be sensitised and the extent of intergranular corrosion and IGSCC of sensitised materials prior to packaging will depend on their storage history. There is also uncertainty concerning whether intergranular attack and IGSCC of sensitised materials will continue under anaerobic alkaline conditions in the longer term.

The results of the assessment calculations for the operational and post-closure Reference Cases can be summarised as follows:

- In the operational period, effective doses from carbon-14 gases released from steels are assessed to be below the source-related dose constraint for members of the public for a new facility of 0.15 mSv yr⁻¹.
- In the post-closure period, the radiological impact of any release of carbon-14 as gas from irradiated steels will be site-specific. Steel wastes are the major contributor to the risks via the gas pathway at times greater than five hundred years post-closure. The calculated risks arising from steel wastes are highest (~3.6x10⁻⁷), but remain below the risk guidance level (10⁻⁶) where all of the gas-phase release is focused to a small area (10⁴ m⁻²) at the surface. Even for a focused surface release, the risks arising from steel wastes will only be above the risk guidance level if a significant proportion (>30%) of the carbon-14 is released as gas.
- In the early post-closure period, the rates of carbon-14 generation from steels are higher for a short period of about 5 years. However, these releases are not expected to give rise to higher doses to a resident family group due to spreading of the releases in time to the soil zone in some geological environments. It is noted that the generation rates from steel wastes are lower than those from Magnox wastes in the same period.

For the variant case accounting for potential IGSCC of stainless steel wastes:

- the effective doses from carbon-14 gases released from steels during the operational period are assessed to be above the design target level only if the pessimistic assumption is made that the carbon-14 is released predominantly as carbon monoxide. Releases as 100% methane would be well below the design target level.
- In the post-closure period, the medium- and long-term post-closure risks are calculated to be above the risk guidance level only where all the release is focused to a small area (assuming that 10% of the carbon-14 is released as methane).
A clear implication of these results is that it will be important to understand the gas migration characteristics of any site proposed for the disposal of these wastes. It is also important to understand the speciation of carbon-14 released from irradiated steel wastes.

Alternative treatment, packaging, design and disposal options are not particularly beneficial for these wastes, but could be considered in future for specific waste streams, should they prove to be problematic.

Nevertheless, for irradiated steel wastes, consideration should be given to the following aspects:

- Whether the distribution of carbon-14 might not be uniform for any particular waste streams (as is assumed in the model), e.g. due to self-shielding of thick or peripheral activated components.
- Whether a different corrosion rate might be applicable to specific waste streams (taking into account the possible effects of irradiation and sensitisation of stainless steel wastes); this may be informed by current work under the NDA’s Direct Research Portfolio (DRP) concerned with corrosion behaviour of spent AGR fuel and fuel cladding during interim storage.
- Whether the carbon-14 inventory could be overestimated, or particularly for new build wastes, for which the inventory is indicative only at this stage, whether material specifications could be chosen to reduce the expected inventory.
- Whether there could be additional surface precipitation of carbon-14 from coolant gas onto reactor steel components during reactor operations and whether this additional carbon-14 could remain following packaging. This is not currently included in the carbon-14 inventory.

It is noted also that the UK Radioactive Waste Inventory is updated every three years and future improvements to the carbon-14 inventory are expected as further understanding is gained of UK legacy wastes including steel wastes from reactor decommissioning programmes and of NNB wastes as the new build programme proceeds.

Ongoing work on irradiated steel wastes is focussing on:

- Experiments to measure the release of carbon-14 from irradiated stainless steel in anaerobic environments, which are in progress under the international CAST project. The implications of the findings of CAST for the assessments presented in this report will be considered as part of the CAST project.

Possible future work includes:

- Consideration could be given to extending measurements of carbon-14 release to the aerobic period and to other steels (although this is not currently a priority).
- Work on the corrosion rates for irradiated stainless steel, and in particular to what extent sensitisation of the steel affects the metal (i.e. intergranular attack and IGSCC) and the general corrosion of the bulk material (potentially building on the DRP AGR fuel work). However, given the difficulties associated with testing of irradiated materials, this process may be better studied using ‘what-if’ calculations, as done in this work.

Our improved understanding also has implications for the assessment of the groundwater pathway in terms of the inventory available for release and the speciation of carbon-14 in the aqueous phase. Account could be taken of the long timescales over which carbon-14 would be released from steel wastes.
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1 Introduction

Carbon-14 (half-life 5,730 years) is a key radionuclide in the assessment of the safety of a geological disposal facility (GDF) for radioactive waste [1]. In particular, the radiological impact of gaseous carbon-14 bearing species is a potential issue and has been recognised as such in Nirex Report N/122 [2], in the 2010 generic Disposal System Safety Case (DSSC) [3], and in the issues register [4]. Carbon-14 is expected to be released from a GDF over a timescale of several thousand years. A number of radioactive gases will be generated from waste materials within a GDF, with carbon-14 bearing methane ($^{14}$CH$_4$) likely to be the dominant carbon-14 species transported in the gas phase, potentially reaching the biosphere at low activity concentrations.

Radioactive Waste Management (RWM) has been carrying out a range of research and assessment tasks on carbon-14 to improve the understanding of the expected evolution of the disposal system and the consequences for the calculated annual risk. Much of this work is summarised in the 2010 gas status report [5], the 2010 radionuclide behaviour status report [6], the 2010 biosphere status report [7] and in the 2014 Science & Technology Plan [8]. However, following publication of the 2010 DSSC it was decided to adopt a collaborative approach between RWM, its technology suppliers and academia to tackle issues related to carbon-14 by establishing a project team in which the partners work together to develop an holistic approach to carbon-14 management in a GDF.

1.1 Carbon-14: Key Processes

The main sources of carbon-14 in waste materials that will be emplaced in a GDF include activated metals, graphite and spent fuel. As these materials are corroded, leached or otherwise degraded, carbon-14 could be released in aqueous or gaseous form. An overview of the key processes affecting the fate of carbon-14 in the geological disposal system is shown in Figure 1.

![Figure 1: Key generation and migration processes affecting the fate of carbon-14 in the disposal system](image-url)
At the same time, corrosion of metals, radiolysis and degradation of organic materials will generate ‘bulk gas’, mostly hydrogen. Intermediate-level waste (ILW) containers are generally vented and any gas generated will be released into the surrounding engineered barrier system (EBS). Depending on the quantity of bulk gas generated, some species containing carbon-14 will dissolve in the near-field pore water and some will remain in the gas phase. Carbon dioxide is likely to be retained within the EBS due to carbonation of cementitious materials. However, methane and carbon monoxide are likely to be either dissolved or entrained within a bulk gas phase.

The geosphere may contain features that delay or prevent the migration of gas but, if gas is able to reach the biosphere, it could be released directly as methane or converted to carbon dioxide by microbes in the soil zone. This carbon dioxide could then be taken up by plants, and so enter the food chain, leading to a consequent dose to any exposed groups or potentially exposed groups.

1.2 Aims and Objectives of the Carbon-14 Project

The overall aim of the carbon-14 project is:

*To support geological disposal of UK wastes containing carbon-14, by integrating our evolving understanding from current and pre-existing projects, in order to develop an holistic approach to carbon-14 management in the disposal system.*

This includes the following sub-objectives:

- To inform strategic decisions on disposal options for specific waste types and inform packaging decisions for specific waste streams;
- To provide data and understanding and, if necessary, develop conceptual and component-level models to support the transport, operational and environmental safety cases;
- To develop options for an integrated waste management approach to disposal of these wastes;
- To establish whether mitigation measures of design solutions are required;
- To develop data and understanding required to inform the siting process; and
- To inform the requirements for site characterisation

At the outset, it was established that a successful outcome from the project would be that:

*RWM understands the envelope of conditions within which the disposal of the UK’s wastes containing carbon-14 can be managed.*

1.3 The ‘AND’ Approach

The project team has developed an integrated technical approach to the work, based on an understanding of the key processes affecting the fate of carbon-14. The approach is known as the ‘AND’ approach and is described in Figure 2. The project team has used this approach as a way of considering the problem comprehensively and to underpin the prioritisation of the work. The ‘AND’ approach has been used for each of the key waste streams to break the problem down in a manageable way.
For the radiological impact of gaseous carbon-14 to be an issue:

- There must be a significant inventory of carbon-14;
  AND
- That waste has to generate carbon-14 bearing gas;
  AND
- A bulk gas has to entrain the carbon-14 bearing gas;
  AND
- These gases must migrate through the engineered barriers in significant quantities;
  AND
- These gases must migrate through the overlying geological environment (either as a distinct gas phase or as dissolved gas);
  AND
- These gases must interact with materials in the biosphere (i.e. plants) in a manner that leads to significant doses and risks to exposed groups or potentially exposed groups.

Figure 2 The integrated technical approach (the ‘AND’ approach)

1.4 Overview of the Project

The project has consisted of two phases. In Phase 1, the project team carried out a six month programme of work to summarise the current understanding and to set out a roadmap for Phase 2 [1]. The work concluded that the calculated release of carbon-14 from a GDF will be dominated by: corrosion of reactive metals (in the operational and early post-closure time frame); corrosion of irradiated stainless steel and leaching of irradiated graphite. However, there was scope for improving the models used to calculate the radiological consequences for these wastes. The work also concluded that there was scope for considering approaches to alternative management strategies that would mitigating the consequences, and for considering how site-specific issues might affect the consequences once sites are identified.

The roadmap identified a number of specific tasks that combined desk-based, numerical modelling and experimental activities. The scope of some tasks was focused on particular groups of carbon-14 bearing wastes, such as graphite, irradiated steels and irradiated reactive metals; these tasks also included consideration of alternative treatment, packaging, design and disposal options for these wastes. Other tasks related to aspects of the disposal system that are not specific to particular wastes: these included migration through engineered barriers, the geosphere and release to the biosphere. Finally, there were tasks in a number of overarching areas, such as modelling in support of safety assessments. Some tasks were already ongoing and were transferred into the project.

Phase 2 of the project has been completed. This report is one of a suite of reports issued by the project. An overview of all of the work undertaken, and the conclusions reached, is provided in Reference [9]. The main reports produced as part of the project are given in Table 1.
Table 1  Key reports for Phase 2

<table>
<thead>
<tr>
<th>Title</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon-14 Project Phase 2: Inventory</td>
<td>AMEC/200047/003 [10]</td>
</tr>
<tr>
<td>Carbon-14 Project Phase 2: Irradiated Steel Wastes</td>
<td>This report</td>
</tr>
<tr>
<td>Carbon-14 Project Phase 2: Irradiated Reactive Metals Wastes</td>
<td>AMEC/200047/006 [12]</td>
</tr>
<tr>
<td>The Oxidation of Methane in Soil as a factor in Determining the Radiological Impact of a Geological Disposal Facility</td>
<td>AMEC/200047/001 [14]</td>
</tr>
<tr>
<td>Operational Impacts from Aerial Discharges of C-14 Labelled Gases</td>
<td>AMEC/200047/002 [15]</td>
</tr>
<tr>
<td>Carbon-14 Project Phase 2: Modelling</td>
<td>AMEC/200047/008 [16]</td>
</tr>
</tbody>
</table>

1.5  This Report

The focus of this report is the work undertaken on irradiated steel wastes.

In the Phase 1 calculations, the release of carbon-14 from irradiated steels dominates the longer term post-closure release in the unshielded ILW vaults and the release rate leads to a calculated risk that exceeds the regulatory risk guidance level by a factor of about seven [1]. However, this is based on a number of simplifications that need to be better understood.

In Phase 2, our approach to applying the ‘AND’ approach for steel wastes was to seek new evidence and to improve the available data, rather than developing new models for the release of carbon-14 from irradiated steels. The following uncertainties related to steel wastes were the focus of work on steels during Phase 2:

- The inventory is based on assumed nitrogen concentrations in steels, which may be overestimated. Work has been undertaken to improve understanding of steel compositions and the basis of waste stream activity calculations performed by waste producers.

- There are only very limited data on carbon-14 release from irradiated stainless steels. An experimental programme to gather these data is planned through the collaborative international CAST project, which started in October 2013 [17]. CAST is not expected to deliver results during the timeframe of Phase 2; however, a detailed review of current understanding has been undertaken as part of the CAST project [18] and forms part of the Phase 2 work.

- The rate of bulk gas generation and carbon-14 release from the corrosion of stainless steel wastes under post-closure conditions is based on an assumed upper limit for the stainless steel corrosion rate. A literature review of steel corrosion rates has been undertaken as part of the CAST project [18] and forms part of the Phase 2 work.

- The modelling approach could take into account more appropriate surface area data to calculate the gas produced by corrosion of specific steel waste types (e.g. AGR fuel cladding). Further information on the geometries of waste steels has been obtained.

This work is now broadly complete and is reported here. The experimental work on the measurement of the rate and speciation of the release of carbon-14 from irradiated steel wastes is still ongoing, and the results will be updated once that work is complete.
This report is structured as follows.

- Information on the projected carbon-14 inventory of steel wastes requiring disposal in a UK GDF for ILW/LLW is presented in Section 2. This section draws on the work outlined in the Inventory Report [10] and presents updated information on the grades and compositions of the waste steels and the geometries of these materials.

- Experimental evidence concerning the measured corrosion rates of steels under geological disposal conditions and the speciation of carbon-14 releases from irradiated steels, which have recently been reviewed under the CAST project [18], are summarised in Section 3.

- The modelling basis for the calculation of carbon-14 and bulk hydrogen gas generation from steels due to corrosion is presented in Section 4; this section also details how the steel corrosion model is parameterised and how different categories of steel wastes are treated.

- Other aspects of the revised modelling basis used in the Reference Case assessment are presented in Section 5.

- The results of the Reference Case calculations are presented in Section 6 with emphasis of the potential contribution of waste steels. The rates of carbon-14 gas generation and the potential consequences of the carbon-14 releases from steel wastes during both the operational and post-closure periods of a GDF are presented.

- The results of variant calculations to scope the potential impact of higher stainless steel corrosion rates in the assessment are presented in Section 7.

- Alternative management approaches for irradiated steel wastes are considered briefly in Section 8.

- Finally, the key results and conclusions of the Phase 2 work on steel waste are presented in Section 9.

An Appendix provides additional information on work to study the distribution of carbon-14 in an irradiated steel specimen.
2 Inventory

This section presents the currently projected carbon-14 inventory of waste steels that will be disposed in a UK GDF. It also presents information on the grades and compositions of the waste steels and the geometries of these materials. This information is required to enable models for the treatment of waste steels in the safety case to be parameterised.

2.1 Overview of improvements to the carbon-14 inventory

In Phase 1 of the project, contributions to the generation rates of carbon-14-containing gas and bulk gas from different types of waste were analysed by processing the results from the 2012 gas generation assessment [19]. This was done to identify the most important contributions to carbon-14 gas generation. The 2012 gas generation assessment was based on the carbon-14 and bulk materials inventories developed as part of the 2007 Derived Inventory (DI) [20]. During Phase 1 [1] and Phase 2 [10] of the project, the carbon-14 disposal inventory has been progressively updated and improved as new and updated information has become available.

In Phase 1 of the project, a carbon-14 inventory for wastes destined for disposal in a UK GDF was developed based on information contained in RWM’s 2010 Derived Inventory [21]. A number of data enhancements were applied to address some limitations of the 2010 DI. The Phase 1 report [1] summarises the then current understanding of the carbon-14 inventory and presents further information on those waste streams that are the major contributors to the carbon-14 inventory. This included waste steels. Gaps in our knowledge and key uncertainties in the carbon-14 inventory were also identified. The understanding of the carbon-14 inventory of waste steels developed during Phase 1 is summarised in sub-section 2.1.1, below.

Since the Inventory work under Phase 1 was completed, a Government White Paper on geological disposal [22] and the 2013 UK Radioactive Waste Inventory [23] have been published, both of which have had an impact on the carbon-14 inventory for geological disposal. During Phase 2 of the project, the carbon-14 inventory has been updated to include the latest information available from the 2013 UK RWI [23] and to address the changes in wastes to be included in the disposal inventory, as laid out in the White Paper. Further information about specific waste materials has been obtained through interactions with the key waste producers (Magnox, Sellafield Limited (SL) and EDF Energy (EDFE)) and with Springfields Fuels Limited (SFL), the makers of the UK’s nuclear fuel, to address knowledge gaps identified during Phase 1. This has enabled a number of additional improvements to be made to the carbon-14 inventory [10]. The resulting updated carbon-14 inventory forms part of RWM’s 2013 Derived Inventory for geological disposal [24]. It is this updated inventory that has been used as the basis of the carbon-14 gas generation assessment calculations presented in later sections of this report and in companion reports arising from the Carbon-14 Project [11, 12, 16].

2.1.1 Understanding of the carbon-14 inventory of steel wastes developed during Phase 1

Carbon-14 is produced in reactor steels by the interaction of thermal neutrons with nitrogen impurities in the steel via the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction. Owing to the low natural abundance and thermal neutron cross-sections of carbon-13, the $^{13}\text{C}(n,\gamma)^{14}\text{C}$ reaction is not a significant source of carbon-14 in the low-carbon content stainless and mild steels that are used in reactor applications.

In the majority of cases, the estimated carbon-14 activities of irradiated steel wastes in the UK RWI are based on neutron activation calculations using assumed nitrogen impurity concentrations for the steel materials concerned.

The carbon-14 inventory for steel wastes produced during Phase 1 is shown in Table 2. Several major types of waste steels contributing to the carbon-14 inventory were identified. These waste types were distributed into five waste stream groups, as indicated in the table. These groups distinguish between operational and decommissioning waste streams and between predominantly stainless steel or other ferrous metal (primarily mild steels) wastes. One waste stream, 2F03/C,
stainless steel AGR fuel cladding was treated separately, because it was identified as contributing significantly to the carbon-14 source term arising from steel wastes. For each waste stream group the best estimate for the total carbon-14 activity is presented in Table 2.

### Table 2 Phase 1 carbon-14 inventory for steel wastes at 2150 based on the 2010 Derived Inventory

<table>
<thead>
<tr>
<th>Material category – Waste stream group</th>
<th>Total C-14 radioactivity at 2150 (TBq) (^{(1,2)})</th>
<th>Contribution to total radioactivity (%) (^{(3,4)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Steels - ILW AGR stainless steel fuel cladding (2F03/C)</td>
<td>28 ((+143))</td>
<td>1.7</td>
</tr>
<tr>
<td>Steels - ILW AGR stainless steel fuel assembly components</td>
<td>356</td>
<td>3.6</td>
</tr>
<tr>
<td>Steels - ILW stainless steels from reactor decommissioning</td>
<td>226</td>
<td>2.3</td>
</tr>
<tr>
<td>Steels - ILW other ferrous metal decommissioning wastes</td>
<td>197</td>
<td>2.0</td>
</tr>
<tr>
<td>Steels - ILW other ferrous metal reactor wastes</td>
<td>28</td>
<td>0.3</td>
</tr>
<tr>
<td>Total</td>
<td>835 ((+143))</td>
<td>9.9</td>
</tr>
</tbody>
</table>

Notes:  
(1) Red figures show the impact of enhancement of the 2010 Derived Inventory. These enhancements were carried forward from the 2007 Derived Inventory.  
(2) Includes Scottish Policy wastes.  
(3) The total Phase 1 carbon-14 inventory amounted to 9,680 (\(+193\)) TBq.  
(4) Includes enhancements.

The nature of these waste stream groups, which have undergone some refinement and division during Phase 2, and the basis of their carbon-14 activities, will be discussed in more detail in Section 2.2.

Knowledge gaps concerning the carbon-14 inventory of steel wastes that were identified during Phase 1 and which have been the focus of Phase 2 inventory investigations of steel wastes are listed in Table 3. Essentially, these concern: the grades and compositions of the reactor steels; the concentrations of nitrogen impurity levels assumed to be present as the basis for activation calculations; and details of the geometries and thicknesses of the materials. During Phase 2, the project team has engaged with waste producers and the manufacturers of the UK’s nuclear fuel to address these knowledge gaps.
### Table 3  Phase 1 key knowledge gaps identified for waste steels

<table>
<thead>
<tr>
<th>Waste stream group</th>
<th>Knowledge gap</th>
</tr>
</thead>
<tbody>
<tr>
<td>ILW AGR stainless steel fuel cladding</td>
<td>Details of the metal thicknesses.</td>
</tr>
<tr>
<td>ILW AGR stainless steel fuel assembly components</td>
<td>Details of the metal thicknesses. Nitrogen impurity levels in the cladding.</td>
</tr>
<tr>
<td>ILW stainless steels from reactor decommissioning</td>
<td>Details of the metal thicknesses. The stainless steel composition/grade used in the AGR reactors and the PWR reactor at Sizewell, and associated data on nitrogen concentration.</td>
</tr>
<tr>
<td>ILW other ferrous metal decommissioning wastes</td>
<td>Details of the metal thicknesses. The steel composition/grade used in the AGR reactors and the PWR reactor at Sizewell, and associated data on nitrogen concentration.</td>
</tr>
<tr>
<td>ILW other ferrous metal reactor wastes</td>
<td>Details of the metal composition/grades.</td>
</tr>
</tbody>
</table>

#### 2.1.2 Changes in the 2013 Derived Inventory

The 2013 Derived Inventory differs in three significant ways from the 2010 Derived Inventory:

- It includes the waste and spent fuel arising from a 16 GW(e) nuclear new build programme (NNB) [22];
- It includes 95% of the plutonium inventory, which is assumed to be disposed as mixed oxide (MOX) spent fuel in line with Government’s preferred policy for long term management [25]; and
- It excludes wastes that are managed under the Scottish Government's policy for higher activity radioactive wastes [26].

In addition, the latest data from the 2013 UK RWI [23] have been integrated into the 2013 DI and the inventory of legacy spent fuel (not included in the 2013 UK RWI) has been revised for the 2013 DI. A number of improvements to the inventory have also resulted from work carried out during Phase 2 of this project. Finally, a revised reference date of 2200 AD has been used for the 2013 DI, taking into account a later closure date for a GDF that would accommodate NNB wastes; all waste stream activities have been decay corrected to 2200 AD.

The net effect of these changes for the 2013 DI is an overall increase in the carbon-14 activity of 7890 TBq compared to the 2010 DI developed during Phase 1 of the project. A significant proportion of this increase, a net 6,100 TBq, is associated with waste steels, primarily from the waste stainless steels that would arise from NNB reactors on decommissioning.

Further details of how these changes have affected the carbon-14 inventory are discussed in the Inventory Report [10].

#### 2.2 Important waste streams containing steel wastes

Waste steels contributing to the carbon-14 disposal inventory are derived from a combination of:

- legacy wastes, the historic and future arisings from currently operating and closed nuclear reactors; and
- NNB wastes, future arisings from a 16 GW(e) nuclear new build programme.

The legacy waste streams contributing to carbon-14 in steels are grouped as follows:

- ILW AGR stainless steel fuel cladding;
- ILW AGR stainless steel fuel assembly components;
- ILW AGR fuel stringer debris¹;
- ILW stainless steel from reactor decommissioning;
- ILW other stainless steel reactor wastes (operational wastes)²;
- ILW other ferrous metals from reactor decommissioning; and
- ILW other ferrous metal reactor wastes (operational wastes).

NNB steel waste streams contributing to the carbon-14 inventory are grouped as:

- ILW AP1000 stainless steel from reactor decommissioning;
- ILW EPR stainless steel from reactor decommissioning;
- ILW other ferrous metals from reactor decommissioning; and
- ILW other stainless steel reactor wastes (operational wastes)²

Appendix Tables A2(a) – A2(g) in the Inventory Report [10] list the waste streams in each group and their contributions to carbon 14 activity in the 2013 Derived Inventory³. The tables identify whether the waste stream will be packaged in shielded or unshielded ILW (SILW or UILW) containers or in ductile cast iron containers (DCICs). In addition, the tables provide the upper and lower bound uncertainty factors for the carbon-14 activity in each waste stream, a description of the types of steel and other materials present in the waste stream and the method used to calculate the carbon-14 inventory in each case⁴. Data on the total material mass (in tonnes), the predominant geometry of the wastes (plate-like or sphere-like) and the mean thickness or diameter of the steels in each waste stream are provided in Appendix Tables B2(a) – B2(j) (for stainless steels) and Tables B3(a) – B3(i) (for other ferrous metals) of the Inventory Report [10].

Each waste group is discussed in turn below, highlighting the new information that has been gained during Phase 2 of the project but noting some of the remaining knowledge gaps. It is noted here that:

- It has proved difficult to find accurate data for the compositions of the various grades of steel used in Magnox reactors because many of them are old grades and the information is not readily available. As a consequence, calculated carbon-14 activities for irradiated steels across the Magnox fleet have been based on generic values for nitrogen contents of the steels, which appear to have been cautiously selected as likely upper limits.
- Very limited information has been obtained to date on steel grades, component thicknesses and nitrogen precursor levels in AGR decommissioning steels (i.e. structural steels installed during reactor build), because this information is also not readily available. However, significantly more information has been obtained concerning fuel cladding and fuel assembly steels from the manufacturing records held by SFL. This information has enabled improvements to be made to the carbon-14 inventory for these wastes.
- There are significant uncertainties concerning the carbon-14 inventory of steel wastes that will arise from new build reactors, which have yet to be built. The inventory is based on illustrative fleets of six European Pressurised [water] Reactors (EPR) and six Advanced Passive pressurised water (AP1000) reactors that will be operated in the UK. Activation calculations have assumed operational lifetimes of sixty years and the carbon-14 inventories are based on generic values for nitrogen contents of steel grades, which have been cautiously selected as likely upper limits.
- The uncertainty factors estimated for the carbon-14 inventories (i.e. multiplication factors for lower and upper bounding inventories) of individual waste streams given in the sections below are taken directly from the 2013 UK RWI, and are based on waste producer submissions.

The inventory information for waste steels is summarised in Section 2.4 (and Table 8 therein).

¹ Treated as a separate waste group in Phase 2; fuel stringer debris was included in the stainless steel fuel assembly components group in Phase 1.
² A new group introduced in Phase 2 for legacy operational wastes identified in the 2013 UK RWI; operational wastes will also arise from NNB reactors.
³ In Appendix A of the Inventory Report [10], NNB steel decommissioning and operational wastes are grouped with similar legacy wastes.
⁴ This information is provided in the 2013 UK RWI [23].
2.2.1 Legacy wastes

2.2.1.1 ILW AGR stainless steel fuel cladding

This waste group comprises a single waste stream, 2F03/C, the encapsulated chopped and nitric acid-leached cladding from AGR fuel pins. AGR fuel cladding comprises austenitic stainless steel type 20Cr/25Ni/Nb and varies in thickness from 0.37mm to 0.46mm. The waste stream also includes the ceramic Sintox end discs of the AGR fuel as a minor component; in addition, some carry-over of fuel with the cladding is allowed for in the inventory calculations. This waste stream is projected to contribute 29.4 TBq of carbon-14 in the 2013 Derived Inventory.

The carbon-14 inventory of this waste stream was re-evaluated for the 2013 DI. The activation calculations (performed with the FISPIN code in 2004) were based on a nitrogen precursor concentration of 100 ppm in the stainless steel. This is consistent with mean values of about 100 ppm measured in cladding materials used in fuel manufacture between 1996 and 2000. It is noted that more recent nitrogen measurements in the period 2008 to 2013 gave lower mean concentrations in the range 53 to 70 ppm. However, no account of these lower values has been taken in the 2013 DI because they were derived from a limited number of measurements on materials used in only a portion of the AGR programme. Nevertheless, these measurements suggest that estimates of the carbon-14 inventory, particularly for future arisings, may have some in-built conservatism.

The revised information for the nitrogen content of this waste stream has shown that a previous enhancement for the carbon-14 inventory of 143 TBq that was included in the Phase 1 inventory (see Table 2), can be discounted.

The uncertainty factors estimated for the carbon-14 activity of this waste stream in the UK RWI are 0.3 and 3 for the lower and upper bounds, respectively (i.e. 8.8-88 TBq. carbon-14).

2.2.1.2 ILW AGR stainless steel fuel assembly components

This waste group comprises a single waste stream, 2F08, stainless steel components (grids, braces and supporting guide tube) removed from AGR fuel assemblies during initial processing at the Sellafield plant. A cutaway illustration of a typical AGR fuel element showing the location of the principal components is given in Figure 3.

Fuel element grids and braces are reported to be made from 20Cr/25Ni/Ti stainless steel and vary in thickness from 0.4 to 1.4 mm, with an average thickness of 0.8 mm.
This waste stream is projected to contribute 38.3 TBq of carbon-14 in the 2013 DI. The specific activity of this waste stream was re-evaluated during the project, based on a nitrogen precursor concentration of 100 ppm and using a conservative assumption for average fuel burn-up\(^5\). This has led to a 170 TBq reduction in the total carbon-14 activity of this stream compared to the 2013 UK RWI.

The uncertainty factors estimated for the carbon-14 activity of this waste stream in the 2013 UK RWI are 0.1 and 10 for the lower and upper bounds, respectively. These uncertainty factors have not been re-evaluated by the project.

2.2.1.3 **ILW AGR fuel stringer debris**

This waste group consists of nine waste streams comprising miscellaneous activated components (MAC) and/or fuel stringer debris, which are removed from AGR fuel stringers during their dismantling at each power station, before the spent fuel assemblies are transported to Sellafield. These wastes are currently stored in MAC vaults at the five AGR stations in England. New waste streams based on individual MAC vaults have been introduced in the 2013 UK RWI, superceding the MAC/fuel stringer debris waste streams considered in the 2010 DI and in Phase 1 of the carbon-14 project.

\(^5\) It is thought that previous estimates of the carbon-14 inventory for this waste stream were based on the upper limit for nitrogen in the specifications for AGR fuel assembly steels of 200 ppm.
In general, these wastes comprise mixtures of stainless steel and graphite in varying proportions with, in some cases, mild steels and/or smaller amounts of Nimonic alloy. Limited information is available on the steel compositions, but in some cases, the stainless steel is stated to be 18Cr/9Ni/Nb; tie bars comprise Nimonic PE16 (a nickel-iron-chromium alloy containing up to 2% by weight of cobalt).

Limited data are available on the geometries of the stainless steel components of 3N38 from Hinkley Point B, which are assigned an average plate thickness of 10 mm.

Historically, the inventories of fuel stringer debris waste streams at each AGR station have combined the activities of the graphite and steel components. In the 2013 UK RWI, new waste streams have been introduced based on individual storage vaults. However, some inconsistencies have been found, in particular for Hinkley Point B. EDFE is currently re-evaluating the activities for all its AGR ILW vaults in preparation for the 2016 UK RWI.

The current best estimate for the total carbon-14 activity of these waste streams in the 2013 DI is 99.5 TBq of which about 30.2 TBq arises from stainless steel components, 32.2 TBq from mild steels and 37.1 TBq from graphite. The uncertainty factors estimated for the carbon-14 activity of waste streams in this group in the 2013 UK RWI vary between 0.01 and 0.1 for the lower bounds and between 10 and 100 for the upper bounds, respectively.

2.2.1.4 **ILW stainless steel from reactor decommissioning**

This waste group comprises 19 waste streams that will arise principally from the final stage clearance of the UK’s commercial Magnox (34.4 TBq) and AGR (11.1 TBq) reactors and the Sizewell B Pressurised Water Reactor (PWR, 35.3 TBq). It also includes waste streams from submarine reactors (7.1 TBq) and the Vulcan Naval Test Reactor at Dounreay (9.6 TBq) plus two waste streams of reactor neutron sources from the Hinkley Point A reactors (5.9 TBq).

The predominant grade of stainless steel in Magnox decommissioning wastes is EN58B (equivalent to AISI Type 321, a titanium stabilised grade); other steels identified across the Magnox fleet include BS970-EN56; BS970-EN59; BS970-EN59F; BS970-321S12; BS980-CDS20; BS1631/1950 and BS1508-82. Information in the 2013 UK RWI states that component thicknesses vary between a few mm and 25 mm.

For activation calculations, a nitrogen impurity concentration of 1,000 ppm was assumed for all grades of Magnox stainless steels (the nitrogen specification upper limit for EN58B) with the exception of Bradwell, where 1,500 ppm was assumed. These values are considered to be upper limits and are likely to overestimate the carbon-14 inventory in the Magnox steel wastes. The uncertainty factors estimated for the carbon-14 activities of these waste streams in the 2013 UK RWI are 0.1 and 10 for the lower and upper bounds, respectively.

No details have been obtained concerning the stainless steel grades, component thicknesses or the nitrogen precursor levels used in activation calculations for AGR decommissioning wastes. However, the uncertainty factors estimated for the carbon-14 activities of these AGR waste streams in the 2013 UK RWI are 0.1 and 10 for the lower and upper bounds, respectively.

Stainless steel wastes from the Sizewell B PWR (waste stream 3S306) will comprise components of the radial shield, the upper and lower core plates and the lower support columns. A breakdown of stainless steel component masses, thicknesses and carbon-14 inventory has been obtained (see Table 15 in [10]), although no information has been obtained concerning the stainless steel grades. A revised estimate for the carbon-14 activity of 3S306 of ~35 TBq comes from a recent review of carbon-14 activity data by EDFE and is based on a nitrogen impurity concentration of 500 ppm. This new estimate is significantly lower than the total carbon-14 activity of ~120 TBq for the waste stream in the 2013 UK RWI. The uncertainty factors assigned to this waste stream in the 2013 UK RWI were 0.001 and 1000 for the lower and upper bounds; these values have yet to be revised but should be reduced by at least an order of magnitude.

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6 It has proved difficult to find accurate data for the compositions of these various grades of steel, because many of them are old grades and the information is not readily available.
No specific information was sought for the remaining waste streams comprising this waste group. The following information was available from the 2013 UK RWI; no details are available concerning the evaluation of carbon-14 activities.

Stainless steels from nuclear submarines, including Vulcan, are reported to comprise austenitic and low-alloy steels. It is notable that the submarine steels, with mean thickness 20mm, have a narrow uncertainty range for their carbon-14 content in the 2013 UK RWI, with uncertainty factors of 0.3 and 1.5 for lower and upper bounds, respectively. In contrast, stainless steels from the Vulcan reactor have a significantly broader uncertainty range for carbon-14, with factors of 0.01 and 100 for the lower and upper bounds, respectively.

The reactor neutron sources from Hinkley Point A comprise sets of 10 irradiated stainless steel (EN58B) tubes (37% by mass) containing slugs of antimony (49% by mass) and beryllium (14% by mass) from each reactor. The uncertainty factors assigned to this waste stream in the 2013 UK RWI were 0.1 and 10 for the lower and upper bounds, respectively.

2.2.1.5 ILW other stainless steel reactor wastes

This group consists of 11 waste streams that comprise debris and redundant components from two Magnox stations (Berkeley and Dungeness A), plus spent cartridge filters from the Sizewell B PWR (stream 3S03), which contribute 4.4 TBq to the carbon-14 inventory in total.

The Magnox components comprise stainless steel fuel element debris from Berkeley and miscellaneous activated components (such as absorber bars, fuelling machine equipment, thermocouple wires and burst can detector equipment) from Dungeness A, which contain a mix of stainless and mild steels. In common with other Magnox stainless steel wastes, component activities were calculated by assuming a nitrogen concentration of 1,000 ppm. The uncertainty factors estimated for the carbon-14 activity in the 2013 UK RWI are 0.01 or 0.1 for the lower limit and a factor of 10 for the upper limit. No information has been obtained on component geometries. The PWR filters comprise 85% type 304 stainless steel. There is no information concerning the component thicknesses or the nitrogen precursor levels used in activation calculations for this waste stream. The uncertainty factors estimated for the carbon-14 activity in the 2013 UK RWI are 0.1 and 10 for the lower and upper bounds, respectively.

2.2.1.6 ILW other ferrous metals from reactor decommissioning

Other ferrous metal waste streams comprise predominantly mild steels that will arise from final dismantling and site clearance (FDSC) of reactor structures, principally from Magnox (90 TBq) and AGR reactors (18 TBq), but with sizeable contributions from other UK reactors: Windscale Piles (3.08 TBq), Dragon (0.76 TBq), WAGR (3.6 TBq) and SGHWR7 (8.4 TBq).

For Magnox reactors, the 2013 UK RWI reports a range of standard steel grades; it is reported in the 2013 UK RWI that metal thicknesses are likely to vary from a few mm to about 100 mm. Table 4 below presents grades and material thicknesses obtained for mild steel components from the Calder Hall Magnox reactor (waste stream 2A311). The nitrogen impurity concentration assumed for all mild steel grades in activation calculations is 200 ppm.

No details are available concerning the types of other ferrous metal wastes, component thicknesses or the nitrogen precursor levels used in activation calculations for AGR FDSC, SGHWR or Dragon wastes. However, detailed composition and geometric information has been obtained for the mild steel components from the Windscale Piles (see Table 17 in [10]), although no details have been obtained concerning nitrogen concentrations. The mild steel thermal shield plates from WAGR, which make up a significant mass fraction of waste stream 2S308/C are reported to be 50 mm thick in the 2013 UK RWI, although the weighted average thickness of all mild steel in the waste package has been calculated to be 20 mm8.

7 Windcale Advanced Gas-cooled Reactor (WAGR); and Steam Generating Heavy Water Reactor (SGHWR)
8 The typical thickness of this waste stream is incorrectly reported as 2 mm in the 2013 DI.
Table 4  Mild steel reactor components from the Calder Hall Magnox reactors

<table>
<thead>
<tr>
<th>Component</th>
<th>Grade</th>
<th>Thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Main pressure vessel shell</td>
<td>COHLO 1 (or LOWTEM)</td>
<td>51</td>
</tr>
<tr>
<td>Vessel reinforcing plates</td>
<td>BS 1501-157B</td>
<td>114</td>
</tr>
<tr>
<td>Inlet/outlet ducts</td>
<td>Plate</td>
<td>13</td>
</tr>
<tr>
<td>Charge tubes</td>
<td>BS1507-151B</td>
<td>6</td>
</tr>
<tr>
<td>Diagrid</td>
<td>BS 15</td>
<td>10 - 32</td>
</tr>
<tr>
<td>Core support plates</td>
<td>BS 15</td>
<td>102</td>
</tr>
<tr>
<td>Gas flow fairings</td>
<td>Plate</td>
<td>6</td>
</tr>
<tr>
<td>Vessel support A frames</td>
<td>-</td>
<td>19 - 25</td>
</tr>
<tr>
<td>Thermal Shield Plates (on inside of bioshield)</td>
<td>-</td>
<td>152</td>
</tr>
</tbody>
</table>

The uncertainty factors estimated for the carbon-14 activity in all Magnox and AGR decommissioning mild steels in the 2013 UK RWI are 0.1 and 10 for the lower and upper bounds, respectively. It is notable that significantly narrower uncertainty ranges are assigned for the Windscale Piles (factors of 0.3 and 3) and prototype reactors (0.3 and 1 for Dragon; 0.7 and 1.5 for WAGR; 0.3 and 3 for SGHWR).

2.2.1.7 ILW other ferrous metal reactor wastes

This waste group comprises miscellaneous activated equipment made principally from mild steels, arising predominantly from Magnox reactors. No specific information is available concerning steel grades. The activities of these components are calculated using neutron activation codes, with an assumed nitrogen concentration of 200 ppm for mild steel and 1,000 ppm for stainless steel.

The uncertainty factors estimated for the carbon-14 activity in these waste steels in the 2013 UK RWI are 0.1 or 0.01 for lower bounds and 10 for the upper bounds, respectively.

2.2.2 Nuclear new build wastes

2.2.2.1 ILW stainless steel and other ferrous metals from the decommissioning of new build reactors

The five waste streams contributing to carbon-14 activity in decommissioning steels from NNB reactors in the 2013 DI are described in Table 5. The 2013 DI assumes that illustrative fleets of six EPR and six AP1000 reactors will be operated in the UK. The activities of these waste streams were calculated in RWM's disposability assessments for the EPR [28] and AP1000 [29] reactors as part of the Generic Design Assessment process, assuming an operational lifetime of 60 years for both reactor types.

Owing to the differing thicknesses and compositions of these waste streams, they have been divided into three waste stream groups for the gas generation calculations:

- ILW AP1000 stainless steel from reactor decommissioning (AP301);
- ILW EPR stainless steel from reactor decommissioning (EP302 and EP303);
- ILW other ferrous metals from reactor decommissioning (AP302 and EP301).

Further details of the decommissioning waste streams from each reactor type are given below.
**EPR**

The largest contributor to the carbon-14 inventory in waste steels is waste stream EP303, the EPR heavy reflector (or shield), contributing 4,870 TBq of carbon-14 to the disposal inventory. The heavy reflector, illustrated in Figure 4, is a large stainless steel structure designed to reduce fast neutron leakage and flatten the core power distribution. The steel specification is a French grade of austenitic stainless steel equivalent to Type 304L.

The calculation of carbon-14 activity in the EPR decommissioning waste streams assumes a nitrogen precursor concentration of 800 ppm to be representative of the reactor steels [28]. In the absence of other information, this value was selected as a conservative assumption. For neutron activation calculations, upper neutron flux values were adopted for EP303 (4,870 TBq). However, for streams EP301 (0.2 TBq) and EP302 (610 TBq), the neutron fluxes were scaled down by applying flux ratios [28]. Note that EP301, the mid-height section of the reactor pressure vessel (RPV) is specified to be ferritic (i.e. mild) steel, with an internal lining, “a few millimeters thick”, of stainless steel for improved corrosion resistance [30].

There is some conflicting information in the available literature concerning the mean steel thickness for waste streams EP302 and EP303. In the absence of further information, the values recommended for use in gas generation assessment calculations are given in Table 5.

**AP1000 reactor**

The carbon-14 activity of ILW steel from the decommissioning of AP1000 reactors is 1,180 TBq. The nature of the activity calculations is similar to those used for the EPR. The calculation of the carbon-14 activity for waste streams AP301 and AP302 assumed stainless steel grade type 304 with a nitrogen concentration of 1000 ppm [29]. In the absence of further information, this value was selected as a conservative assumption. Average neutron flux values were used for the irradiation of AP1000 reactor components, in contrast to the upper estimates used in the EPR calculations. This central estimate of activity and the lower electrical power output of an AP1000 (1.14 GW(e)) in comparison to an EPR (1.6 GW(e)) result in significantly lower activities of carbon-14 in the decommissioning steels.

Waste stream AP301 contains various reactor internal components such as: the radial shield baffle, core barrel, neutron pads and formers, upper and lower axial shield, loop pipes, radial shield installation, and core liner [29]. The steels are expected to be in plate form with a typical thickness of ~10 mm [29].

Waste stream AP302 contains the mid-height section of the RPV and internal vessel cladding. The bulk of the pressure vessel is specified to be a ferritic (i.e. mild) steel, with a thickness of ~200 mm [29, 30], with a internal lining of stainless steel a few millimetres thick.

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9 The RPV stainless steel linings for both EP301 and AP302 have been assumed to have a thickness of 2 mm in the Inventory Report [10]. However, no carbon-14 has been assigned to the linings; all carbon-14 in these waste streams is assigned to the mild steel bulk of the RPVs.
<table>
<thead>
<tr>
<th>Waste stream ID</th>
<th>Waste stream name</th>
<th>Physical description</th>
<th>Steel grade</th>
<th>Mass (t)</th>
<th>Average thickness (mm)</th>
<th>C-14 activity (TBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>EPR:</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>EP301</td>
<td>Decommissioning: reactor vessel</td>
<td>Ferritic (mild) steel associated with the mid-height section of the pressure vessel with an inner lining of stainless steel a few mm thick.</td>
<td>Ferritic (other ferrous metal)</td>
<td>1080 (+57 lining)</td>
<td>200</td>
<td>0.22</td>
</tr>
<tr>
<td>EP302</td>
<td>Decommissioning: upper and lower reactor internals</td>
<td>Low cobalt stainless steel in the form of plates with thickness of the order of 10 mm.</td>
<td>Low cobalt stainless steel</td>
<td>459</td>
<td>275 (1)</td>
<td>610</td>
</tr>
<tr>
<td>EP303</td>
<td>Decommissioning: lower reactor internals including heavy reflector (or shield)</td>
<td>Low cobalt stainless steel used for components closest to the core receiving the highest irradiation, and a grade containing higher concentrations of cobalt for more peripheral components.</td>
<td>Z2 CN19-10 + N2 (2)</td>
<td>833</td>
<td>150 (3)</td>
<td>4,870</td>
</tr>
<tr>
<td>AP1000:</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>AP301</td>
<td>Decommissioning: ILW steel</td>
<td>Stainless steel associated with pressure vessel internals.</td>
<td>Stainless steel (4)</td>
<td>867</td>
<td>10</td>
<td>1,180</td>
</tr>
<tr>
<td>AP302</td>
<td>Decommissioning: pressure vessel</td>
<td>Ferritic (mild) steel associated with the mid-height section of the pressure vessel with an inner lining of stainless steel a few mm thick.</td>
<td>Ferritic (other ferrous metal) (4)</td>
<td>1840 (+97 lining)</td>
<td>200</td>
<td>1.14</td>
</tr>
</tbody>
</table>

Notes:
(1) This value has been selected from reference [31] and is the average thickness for reactor components with plate thicknesses that range from 60 to 415 mm. Another value has also been reported ~10 mm [28].
(2) French specification – equivalent to 304L with controlled nitrogen content.
(3) This value has been selected from reference [32] and is an effective thickness that takes account of cooling holes drilled vertically through the reflector that occupy about 5% of the total volume (these can be clearly seen in the photograph in Figure 4). Another value has also been reported ~10 mm [28].
(4) Assumed to be type 304 (an austenitic steel) in activation calculations.
Figure 4  EPR heavy reflector (EP303) [33]
2.2.2.2 ILW other stainless steel reactor waste from new build reactors

This waste stream group comprises two operational waste streams from the EPR and one from the AP1000 reactor. The waste streams, which comprise spent filters, are described in Table 6. The activities of these waste streams were calculated in NDA’s GDA disposability assessments for the EPR [28] and AP1000 reactors [29]; their total carbon-14 activity in the 2013 Derived Inventory is 3.39 TBq.

There is no information available concerning the likely compositions of these wastes; for this reason they are assumed to have the same composition as waste stream 3S03, spent cartridge filters (ILW) from the Sizewell PWR, which comprises 85% 304-type stainless steel with glass fibre filter element and some organic materials plus retained water (~7%).

Table 6 New build operational stainless steel wastes

<table>
<thead>
<tr>
<th>Stream identifier</th>
<th>Stream name</th>
<th>Physical description</th>
<th>Mass (t)</th>
<th>Average thickness (mm)</th>
<th>C-14 activity (TBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>EP02</td>
<td>Spent Cartridge Filters (ILW)</td>
<td>Filters consisting of a stainless steel support with a glass fibre or organic filter media</td>
<td>230</td>
<td>5</td>
<td>2.57</td>
</tr>
<tr>
<td>EP03</td>
<td>Spent Cartridge Filters (ILW+LLW)</td>
<td></td>
<td>230</td>
<td>5</td>
<td>0.75</td>
</tr>
<tr>
<td>AP01</td>
<td>Primary Circuit Filters</td>
<td>Filters used in: Chemical and Volume Control System (CVCS), Spent Fuel pond cooling System (SFS), Liquid Radwaste System (WLS), Solid Radwaste System (WSS)</td>
<td>33.0</td>
<td>5</td>
<td>0.066</td>
</tr>
</tbody>
</table>

2.2.3 Packaging of waste steels

For most ILW, including reactor steel wastes, packaging will consist of encapsulation in a cement-based material within 500 litre stainless steel drums or 3m³ stainless steel boxes; some larger items may be packaged in higher capacity stainless steel or concrete boxes [34]. These containers will contain vents to allow the escape of gas arising from waste degradation processes, in particular steel corrosion.

Two basic categories of waste package for ILW have been defined [35]:

- For ILW with lower specific activity, that would not generally require the use of remote handling techniques, ‘shielded’ waste packages can be used. The containers used for such waste packages would generally be fabricated using relatively thin (i.e. a few mm) stainless steel and can incorporate integral concrete shielding, if this is needed, to ensure that external radiation dose rates do not exceed the regulatory limits for transport.

- For higher activity ILW, that would require the use of remote handling techniques, ‘unshielded’ waste packages have typically been used. Such waste packages are also fabricated using thin-walled containers but do not usually incorporate any integral shielding. Because of their high external radiation dose rate, or requirements for the containment of their contents, unshielded waste packages would require additional external shielding to permit them to be transported, which would normally be provided by a reusable shielded transport container.

Owing to their different handling requirements, these different categories of waste package will be placed in separate disposal vaults within a GDF.

Owing to the high specific activities of reactor steels, the majority of carbon-14 containing steel wastes (including most NNB steel wastes) will be designated as unshielded ILW. A higher proportion of waste mild steels will be disposed as shielded packages, however.
Some ILW from nuclear power stations is being packaged in thick-walled ductile cast iron containers (DCICs) without encapsulation of the wastes. These containers do not require a store with significant levels of shielding. A number of legacy other stainless steel and other ferrous metal waste streams may be packaged in DCICs.

Further information on the distribution of steel waste streams between waste package categories is given in Appendices A and B of the Inventory Report [10]. The distribution of the carbon-14 in steels inventory between these package categories for use in the assessment calculations is summarised in Section 4.2.

2.3 Knowledge gained of carbon-14 inventory in steel wastes

Work on steel wastes during Phase 2 of the project has focused on those material categories and waste stream groups making the largest contributions to the carbon-14 inventory. Table 7 provides a summary of the knowledge gained concerning legacy steel wastes during Phase 2. NNB steel wastes have been included in the carbon-14 inventory for the first time during Phase 2, and at this stage their carbon-14 inventory is illustrative. Significant improvement to the NNB component to the carbon-14 inventory is expected as the new build programme proceeds. In addition, the UK RWI is updated every three years and future improvements are expected as further understanding is gained of UK wastes including waste steels.

<table>
<thead>
<tr>
<th>Waste stream group</th>
<th>Knowledge gap identified in Phase 1</th>
<th>Knowledge gained during study</th>
</tr>
</thead>
<tbody>
<tr>
<td>All groups</td>
<td>Details of activation calculations that underpin the 2013 UK RWI activities.</td>
<td>Basis of activation calculations has been obtained. For Magnox reactors, carbon-14 activities are based on a zonal MCBEND neutron flux model. EDFE has used an average Westcott neutron flux for activation calculations in AGRs.</td>
</tr>
<tr>
<td></td>
<td>How the uncertainties have been allocated in the 2013 UK RWI activities.</td>
<td>Uncertainties in neutron flux, etc. have been taken into account in activity uncertainty bands.</td>
</tr>
<tr>
<td></td>
<td>How representative are activities of waste streams as a whole (includes variability).</td>
<td>AGR activities are likely overestimates, yet lie within quoted uncertainty bands.</td>
</tr>
<tr>
<td>ILW AGR stainless steel fuel cladding (2F03/C)</td>
<td>Details of the metal thicknesses, surface area, size and shape (geometry).</td>
<td>Metal thicknesses are known.</td>
</tr>
<tr>
<td></td>
<td>Nitrogen impurity levels in the cladding.</td>
<td>Information from SFL supports 100 ppm N basis of existing calculation, enhancement used in the 2007 DI and applied to 2010 DI can be discounted. Propose that historic N measurements are investigated to confirm 100 ppm level.</td>
</tr>
<tr>
<td>Waste stream group</td>
<td>Knowledge gap identified in Phase 1</td>
<td>Knowledge gained during study</td>
</tr>
<tr>
<td>--------------------</td>
<td>-----------------------------------</td>
<td>-------------------------------</td>
</tr>
<tr>
<td>ILW AGR stainless steel fuel assembly components (2F08)</td>
<td>Details of the metal thicknesses, surface area, size and shape (geometry).</td>
<td>SFL have provided thickness data for assembly components fabricated post-1990.</td>
</tr>
<tr>
<td></td>
<td>Nitrogen impurity levels in the grids and braces.</td>
<td>SFL have provided nitrogen specification limits and nitrogen levels measured over a number of years.</td>
</tr>
<tr>
<td>ILW AGR fuel stringer debris</td>
<td>Considered with 2F08 in Phase 1</td>
<td>Discrepancies have been identified in the assignment of carbon-14 activities for these mixed waste streams in the 2013 RWI. EDFE is undertaking a review of activities of AGR ILW storage vaults</td>
</tr>
<tr>
<td>ILW stainless steels from reactor decommissioning</td>
<td>Details of the metal thicknesses, surface area, size and shape (geometry).</td>
<td>Some thickness, surface area and geometry data have been obtained for three Magnox stations. Information for PWR decommissioning ILW stainless steel typical thicknesses has been obtained. No data have been obtained for AGR stations.</td>
</tr>
<tr>
<td></td>
<td>The stainless steel composition/grade used in the AGR reactors and the PWR reactor at Sizewell, and associated data on nitrogen concentration.</td>
<td>A nitrogen precursor value of 500 ppm has been taken for all stainless steel decommissioning waste from Sizewell B PWR. No further information on grades for AGR/PWR stainless steels has been obtained. Sizewell B carbon-14 activity overestimated in 2013 UK RWI.</td>
</tr>
<tr>
<td>ILW other stainless steel reactor wastes</td>
<td>Not considered separately in Phase 1</td>
<td>No further information has been sought.</td>
</tr>
<tr>
<td>ILW other ferrous metal decommissioning wastes</td>
<td>Details of the metal thicknesses.</td>
<td>Some thickness, surface area and geometry data have been obtained for three Magnox stations.</td>
</tr>
<tr>
<td></td>
<td>The steel composition/grade used in the AGR reactors and the PWR reactor at Sizewell, and associated data on nitrogen concentration.</td>
<td>No further information has been obtained.</td>
</tr>
<tr>
<td>ILW other ferrous metal reactor wastes</td>
<td>Details of the metal composition/grades.</td>
<td>All Magnox reactor decommissioning mild steels have an assumed nitrogen precursor value of 200 ppm in calculations.</td>
</tr>
</tbody>
</table>

### 2.4 Summary

The total carbon-14 activities for each steel waste stream group in the 2013 Derived Inventory are given in Table 8, together with the projected masses, range of thicknesses and packaged volumes of the wastes. These inventories represent the position at the end of Phase 2 of the Carbon-14 Project.
Table 8  Summary of packaged volume, mass, thickness and carbon-14 activity of waste steels (at 2200 AD) in the 2013 Derived Inventory by waste stream group

<table>
<thead>
<tr>
<th>ILW Waste stream group</th>
<th>No. of waste streams</th>
<th>Packaged volume (m³)</th>
<th>Mass of steel (t)</th>
<th>Thickness range (mm)</th>
<th>Carbon-14 activity (TBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Legacy - AGR stainless steel fuel cladding (2F03/C)</td>
<td>1</td>
<td>2,070</td>
<td>934</td>
<td>0.37-0.46</td>
<td>29.4</td>
</tr>
<tr>
<td>Legacy - AGR stainless steel fuel assembly components (2F08)</td>
<td>1</td>
<td>1,250</td>
<td>235</td>
<td>0.4-1.4</td>
<td>38.3</td>
</tr>
<tr>
<td>Legacy - AGR fuel stringer debris</td>
<td>9</td>
<td>3,310</td>
<td>927</td>
<td>10 (1)</td>
<td>62.4 (2)</td>
</tr>
<tr>
<td>Legacy - stainless steels from reactor decommissioning</td>
<td>19</td>
<td>4,560</td>
<td>3,620</td>
<td>2.0-25</td>
<td>103</td>
</tr>
<tr>
<td>Legacy - other stainless steel reactor wastes</td>
<td>11</td>
<td>320</td>
<td>109</td>
<td>na</td>
<td>4.39</td>
</tr>
<tr>
<td>Legacy - other ferrous metal decommissioning wastes</td>
<td>28</td>
<td>21,800</td>
<td>17,410</td>
<td>2-152</td>
<td>124</td>
</tr>
<tr>
<td>Legacy - other ferrous metal reactor wastes</td>
<td>29</td>
<td>2,570</td>
<td>1,630</td>
<td>5 (3)</td>
<td>29.4</td>
</tr>
<tr>
<td>NNB – AP1000 stainless steels from reactor decommissioning (AP301)</td>
<td>1</td>
<td>432</td>
<td>867</td>
<td>10</td>
<td>1,180</td>
</tr>
<tr>
<td>NNB – EPR stainless steels from reactor decommissioning (EP302, EP303)</td>
<td>2</td>
<td>1,390</td>
<td>1,290</td>
<td>60-415</td>
<td>5,480</td>
</tr>
<tr>
<td>NNB – other ferrous metal decommissioning wastes (EP301, AP302)</td>
<td>2</td>
<td>2,040</td>
<td>2,920 (4)</td>
<td>200</td>
<td>1.36</td>
</tr>
<tr>
<td>NNB - other stainless steel reactor wastes</td>
<td>3</td>
<td>8,790</td>
<td>493</td>
<td>5</td>
<td>3.39</td>
</tr>
<tr>
<td>Totals</td>
<td>106</td>
<td>48,600</td>
<td>30,400</td>
<td></td>
<td>7,060</td>
</tr>
</tbody>
</table>

Notes:  na = no data available
(1) Data available for one waste stream only, with mean thickness 10mm.
(2) The total carbon-14 activity associated with this waste stream group is 99.5 TBq of which about 37 TBq of carbon-14 arises from irradiated graphite.
(3) Data available for three Sizewell A and three Wylfa streams only, each with mean thickness 5mm.
(4) This figure excludes 154 t of stainless steel liner.
3 Experimental Evidence

This section gives an overview of the experimental evidence concerning:

- the corrosion rates of mild steel and stainless steel under conditions that will arise during both operational phase and the post-closure phase of a GDF;
- the speciation of carbon-14 released from irradiated steels.

First, it is appropriate to give a brief overview of the range of conditions that will arise during the operational and post-closure phases of a GDF, under which corrosion of both unirradiated steels present in the EBS (e.g. as containers) and irradiated waste steels may occur. The corrosion rate of the irradiated steels is expected to control the rate at which releases of carbon-14 may occur from the steel. Under anaerobic conditions, the corrosion rate of the steel will also control the release of bulk hydrogen gas.

3.1 Chemical conditions

In the current modelling basis, the consequence of carbon-14 bearing gas is assessed from emplacement in the GDF during the operational phase through the backfilling of the waste vaults and tunnels, and their resaturation with water, to the long-term post-closure phase of a GDF. The range of chemical environments that need to be considered in the assessment, and thus the range of corrosion parameters that need to be defined, are outlined in the following sub-sections. A distinction is drawn between aqueous and atmospheric exposure, because corrosion is different for these two environments. Under aqueous exposure, the key parameters affecting the corrosion rate are: the availability of oxygen and redox potential; the solution pH, the temperature; and the concentration of chloride (and other corrosive anions) in the water. Under atmospheric exposure, the key parameters are the humidity of the atmosphere and the potential for deposition of corrosive salts (in particular chlorides) onto the steel surface.

For the purposes of the assessment calculations undertaken in Phase 2 of the project, an illustrative GDF concept for ILW/LLW disposal in a higher strength host rock has been selected as the Reference Case. In this illustrative disposal concept [36], ILW packages will be placed in engineered vaults, which will be backfilled with a cementitious material, such as the Nirex Reference Vault Backfill, that is designed to maintain a high near-field pH for a very long time.

3.1.1 Conditions during the operational period

As noted in sub-section 2.2.2.2, waste steels are anticipated to be packaged in vented stainless steel ILW containers that will be grouted with a cementitious material. Thus, the waste steels and internal surfaces of waste containers will be exposed to an alkaline, cementitious environment throughout their handling, interim storage, transport, emplacement and disposal in a GDF.

During the operational period of the GDF, conditions in the waste packages are expected to remain aerobic, assuming that air can enter through the gas vent. It is conceivable, however, that some isolated pockets of anaerobic conditions could develop in closed voids as a result of corrosion or microbial activity\(^1\)\(^2\) [18]. For waste packages containing segregated steel wastes, chloride concentrations in the porewater would be expected to be low.

Although the cement grout will be fully saturated initially, water losses are expected to occur due to evaporation through the container vent and its consumption by corrosion. As a result, the steel wastes may be exposed to both the atmosphere within the waste package and any free solution.

The internal temperature of the waste package will be determined by the external temperature and the small amount of heat generated by the wastes. Prior to emplacement in the GDF, the ambient temperature of the atmosphere in contact with the waste container during fabrication, filling, above

\(^{10}\) Steel corrosion rates are lower under anaerobic conditions, so this would have the beneficial effect of lowering the rates of carbon-14 release in these isolated regions.
ground storage and transport will probably be in the range 5°C to 35°C. Temperature cycling during storage may cause condensation / evaporation cycles, but radiation induced heating will tend to counteract this effect. The relative humidity inside the package will be a function of the water content of the cement grout and the external relative humidity. The temperature in a disposal vault during the operational phase may be as high as 35°C depending on the depth of the facility and the effects of radiogenic heating (see discussion in reference [13]).

The external surfaces of steel waste containers will be exposed to an indoor atmosphere during interim storage and emplacement in a GDF up until backfilling of the ILW vaults. Under these sheltered conditions, the deposition of air-borne salt particles onto the container surfaces is likely to be minimised.

For a GDF located in a higher strength rock, there will be a natural tendency for groundwater to enter the void space of the engineered vaults. Inflowing water will be removed by pumping. Although humidity in the vaults will be controlled to levels less than 100%, it is unlikely that the humidity will be controlled to sufficiently low levels to prevent atmospheric corrosion.

### 3.1.2 Conditions during the closure and post-closure phases

Once the GDF is full, the waste vaults will be backfilled with a cement-based backfill material, the access tunnels backfilled and the facility closed. In the early years after closure, the temperature within some parts of the facility will increase, mainly due to chemical reactions such as curing of the backfill, radioactive decay, corrosion and microbial degradation. Several recent studies [37, 38] provide information on the expected temperature variation in a GDF over time.

After closure, groundwater will flow back into the engineered facility, and there will be a period of intermediate water saturation before full re-saturation of the backfilled waste vaults is achieved. The rate of re-saturation will depend primarily on the permeability of the host rock and the regional head gradient. This intermediate period may last anything from a few years in a fractured higher strength host rock to a thousand or more years in a low-permeability sedimentary rock. During this period the humidity in the vault atmosphere may be 100%, but there is uncertainty whether gas generation processes will be inhibited in those packages into which water has yet to percolate.

The groundwater is expected to contain very low concentrations of oxygen and therefore will impose anaerobic conditions within the GDF. Corrosion and microbial activity will also reduce the oxygen concentration in the near field; once the residual oxygen has been consumed, conditions will become anaerobic.

The inflowing groundwater may contain significant concentrations of chloride ions (up to tens of thousands of ppm in coastal locations), a particularly important aggressive anion with regard to the corrosion behaviour of metals (especially stainless steels), and/or other anionic species (such as sulphide) that may affect corrosion rates. However, the pH of the near-field porewater will be controlled by the cementitious backfill materials and would be expected to be >12.5 initially. In general, once re-saturation has been achieved, these alkaline, anaerobic aqueous conditions, with chloride (and other ion) concentrations imposed by the groundwater composition, are expected to persist throughout the period relevant to the post-closure assessment of risk associated with releases of carbon-14 (i.e. up to 100,000 years).

### 3.1.3 Summary of the chemical conditions

A summary of the temperature variation, water availability and oxygen availability assumed for the GDF during different periods and used as the modelling basis for the assessment calculations, is given in Table 9. The same set of assumptions was used in the 2012 gas assessment [19].
Table 9  Timing of stages, temperature and water and oxygen availability in the Reference Case

<table>
<thead>
<tr>
<th>Stage</th>
<th>Time (AD)</th>
<th>Temperature</th>
<th>Water availability</th>
<th>Oxygen availability</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emplacement</td>
<td>For Legacy vaults: 2040 – 2190</td>
<td>35°C</td>
<td>85% saturation of voidage within waste packages initially</td>
<td>Aerobic (except for uranium waste)</td>
</tr>
<tr>
<td></td>
<td>For NNB vaults: 2100 – 2190</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Backfilling and closure</td>
<td>2190 – 2200</td>
<td>45°C</td>
<td>As for Emplacement</td>
<td>As for Emplacement</td>
</tr>
<tr>
<td>Post-closure (resaturation)</td>
<td>2200 – 2205</td>
<td>45°C</td>
<td>95% saturation of pore space within backfill initially. All pore space resaturates over 5 years.</td>
<td>Oxygen present at closure is consumed (or is released after 5 years)</td>
</tr>
<tr>
<td>Post-closure (long term)</td>
<td>2205 onwards</td>
<td>35°C</td>
<td>Unlimited</td>
<td>Anaerobic</td>
</tr>
</tbody>
</table>

¹ HSR = higher strength rock

Therefore, corrosion rate data are required under the following conditions:

- indoor atmospheric corrosion - for the external surfaces of steel containers up to and during the resaturation period post-closure;
- aerobic corrosion under high-pH conditions - for steel wastes up to and during the resaturation period post-closure; and
- anaerobic corrosion under high-pH conditions - for steel wastes and containers during the long-term post-closure period.

3.2 Corrosion of steel

Data on the corrosion rates of carbon steels and stainless steels under disposal conditions were reviewed in 2007 [39]. Based on that review, recommendations were made regarding the corrosion rates to be used in the SMOGG¹¹ gas generation model [40]. The recommended corrosion rates are summarised in Sections 4.4, 6.3 and 8.3 of reference [39]. The same values were used in the 2012 gas assessment [19].

The review of steel corrosion rates has been updated as a task during the first year of the CAST project [18]. The main findings of the CAST review are summarised in the following sub-sections. First, the mechanisms of corrosion affecting waste steels will be outlined.

3.2.1 Mechanisms of corrosion

Corrosion processes can generally be divided into two main categories, namely general corrosion and localised corrosion. General (or uniform) corrosion results in an even distribution of metal removal across the complete surface of the metal, whereas localised corrosion results in corrosion at very small sites, while the main part of the surface remains uncorroded. Carbon steel is not a corrosion resistant material and it is mainly affected by general corrosion, whereas stainless steel and nickel alloys (such as nimonic) are corrosion resistant materials that experience very low

¹¹ The Simplified Model of Gas Generation, a spreadsheet tool developed for RWM to model gas generation from radioactive wastes in the UK Waste Inventory for use in packaging proposal and safety assessment studies.
uniform corrosion rates, due to the presence of a protective passive film. However, they can be subject to various forms of localised corrosion in the presence of aggressive species such as chloride under oxic conditions. This localised corrosion can take the form of pitting, in which small hemispherical depressions develop at sites of breaks in the passive film, crevice corrosion which develops at mating surfaces, where locally aggressive conditions can develop, and stress corrosion cracking, which can develop in the presence of an applied stress, aggressive species or a susceptible material.

General corrosion tends to be the most significant corrosion mechanism for carbon steels, but for stainless steels, localised corrosion (e.g. pitting) may be a more important mechanism for release of a significant corroded volume of material under oxic conditions, due to the low uniform corrosion rates.

If microbially influenced corrosion (MIC) were to occur, the corrosion rates could be significantly higher than the abiotic corrosion rate. MIC of various materials that may arise in nuclear waste has been reviewed by King et al. [41], who carried out a detailed review of the potential for microbial activity in a range of different disposal concepts. In all concepts, microbes will be present in the groundwater, but the backfill material will tend to reduce microbial activity in the vicinity of the waste package. In the case of a cement-based near-field, of principal interest here, the alkaline environment will prevent microbial activity so that microbial activity at the surface of the steel wastes will be severely restricted.

3.2.1.1 Corrosion processes affecting carbon steel

The following reactions [42] are assumed to occur in oxygenated conditions:

\[
\begin{align*}
4\text{Fe} + 6\text{H}_2\text{O} + 3\text{O}_2 & \rightarrow 4\text{Fe(OH)}_3 \\
4\text{Fe} + 3\text{O}_2 & \rightarrow 2\text{Fe}_2\text{O}_3
\end{align*}
\]

(1)

These reactions result in the consumption of oxygen.

In anaerobic conditions, corrosion is thought to occur according to the reactions [43, 44]:

\[
\begin{align*}
\text{Fe} + 2\text{H}_2\text{O} & \rightarrow \text{Fe(OH)}_2 + \text{H}_2 \\
3\text{Fe} + 4\text{H}_2\text{O} & \rightarrow \text{Fe}_3\text{O}_4 + 4\text{H}_2 \\
3\text{Fe(OH)}_2 & \rightarrow \text{Fe}_3\text{O}_4 + 2\text{H}_2\text{O} + \text{H}_2 & \text{(at higher temperatures)}
\end{align*}
\]

(2)

In these reactions water is reduced with the release of hydrogen gas.

The equations above provide a simplified view of the complex array of reactions that are likely to occur in practice. A range of corrosion products would be formed under the many combinations of pH, chloride concentration and temperature that could occur during waste management. The composition of the corrosion products and passive films that are likely to form on steel surfaces during the various stages of radioactive waste management are discussed in Section 3 of reference [18].

3.2.1.2 Corrosion processes affecting stainless steel

The corrosion behaviour of stainless steels is dominated by the presence of a thin passive film on the surface of the metal [50], which is commonly composed of a hydrated chromium-rich oxy-hydroxide [45]. The exact composition and structure depend on the material and the environment. The passive film may be only of the order of a few nm thick, but it is sufficient to confer a high degree of corrosion resistance, so that the uniform corrosion rates are normally very low, unless the environment is highly acidic, which is not likely to occur in the context of waste

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12 There are some alkaline-tolerant microbial species, but the upper pH limit for microbial activity is thought to be around pH 11. It is possible that lower pH niches could develop that would allow greater microbial activity.

13 Nickel-chromium alloys, such as Inconel, are also protected by a passive film which confers high corrosion resistance [46,47].
disposal in normal circumstances. There has been much experimental research and theoretical modelling of the structure and properties of the passive film (e.g. [48]).

If stainless steel does suffer from corrosion under aerated conditions, it is normally in the form of localised corrosion and this is most commonly caused by the presence of chloride ions which are ubiquitous in natural environments. Chloride ions are able to disrupt the stability of the passive film, although the exact mechanism of this process is still not well understood and this is the subject of ongoing research. The two main types of mechanism for the breakdown of passivity leading to pit initiation are the ‘ion penetration’ and ‘film breakdown’ mechanisms [49]. The pit propagation process involves acidification inside the pit due to hydrolysis of the metal ions released during the anodic dissolution step, supported by external cathodic reactions (e.g. oxygen reduction) on the surface of the surrounding metal around the pit mouths. The higher the concentration of chloride the higher the risk of various types of localised corrosion, such as pitting and crevice corrosion.

Under de-aerated conditions the risk of localised corrosion is very low because the corrosion potential is below the breakdown potentials for localised corrosion to occur, and there is a lack of cathodic reactant, but in the presence of oxygen the risk of localised corrosion increases as the corrosion potential becomes more positive. For example, de-aeration is used in industrial plant to eliminate the risk of localised corrosion of stainless steels, for example in desalination equipment. In aerated conditions, the risk of localised corrosion increases with increasing temperature, increasing chloride concentration and decreasing pH.

The corrosion properties of stainless steel in general are thoroughly described and summarised by Sedriks [50]; the corrosion behaviour of austenitic and duplex stainless steels specifically during radioactive waste management has been reviewed by King et al. for the UK NDA [51, 52], with a focus on the 304 and 316L austenitic stainless steels and the 2304 and 2205 duplex stainless steels. Under the reducing alkaline conditions that will develop in waste packages in a GDF post-closure, the likelihood of localised corrosion of stainless steels is minimised. However, in a GDF the effects of irradiation on the corrosion of stainless steel wastes also needs to be considered and this is discussed further in Section 3.2.5.

### 3.2.2 Treatment of steel corrosion in SMOGG

In general, experimental studies of the corrosion of steels have been performed using materials that have been treated with acid to remove the passivating oxide layer on the steel surface. Typically, there is an initial short-term period of rapid (acute) corrosion followed by a long-term period of slower (chronic) corrosion. This is represented in the current gas generation model, as implemented in SMOGG [40], by the sum of two terms, each of which decays exponentially with time. Each contribution is the product of a rate constant, a time-varying exponential factor and an exponential (Arrhenius) factor describing the temperature dependence. The model implemented specifies the corrosion rate of steels, \( r \) [m yr\(^{-1}\)] as:

\[
    r = \frac{ds}{dt} = -k_a e^{-\frac{t}{t_a}(T)} - k_c e^{-\frac{t}{t_c}(T)}
\]  

(3)

where

- \( s \) is the position of the surface of the steel [m];
- \( t \) is the time during which corrosion is occurring [yr];
- \( k_a \) is the initial acute corrosion rate (defined at a reference temperature, \( T_{ref} \) for which \( t_a = 1 \) [m yr\(^{-1}\)];
- \( t_a \) is a characteristic time for the acute corrosion [yr];
- \( f_a \) is a factor that specifies the dependence of the acute corrosion rate on temperature [\(-\)];
- \( T \) is the temperature [K];
- \( k_c \) is the initial chronic corrosion rate (defined at a reference temperature, \( T_{ref} \) for which \( t_c = 1 \) [m yr\(^{-1}\)];
- \( t_c \) is a characteristic time for the chronic corrosion [yr];
\[ f_c \] is a factor that specifies the dependence of the chronic corrosion rate on temperature [\text{-}].

This formulation allows separate corrosion rates to be specified for an acute phase and a chronic phase of corrosion.

In the current implementation of the model in SMOGG, the assumption is made that the corrosion rates increase with temperature according to an Arrhenius relationship (although alternative relationships can be applied). For each corrosion phase, \( p \) (acute or chronic):

\[
f_p(T) = \exp \left( - \frac{E_p}{R} \left( \frac{1}{T} - \frac{1}{T_{\text{ref}}} \right) \right)
\]

where

- \( E_p \) is the activation energy for the phase of corrosion [J mol\(^{-1}\)];
- \( R \) is the gas constant [J mol\(^{-1}\)K\(^{-1}\)]; and
- \( T_{\text{ref}} \) is a reference temperature at which \( k_a \) and \( k_c \) are defined [K].

The same model can be applied for both carbon steels and stainless steel, although there is a variation in the model parameters used for each steel type. Hence, a total of six parameters must be specified to define the corrosion rate of each steel type for a particular corrosive environment:

- two rate constants (\( k_a \) and \( k_c \) giving the base corrosion rates);
- two characteristic times (\( t_a \) and \( t_c \) defining the decrease in corrosion rates with time); and
- two activation energies (\( E_a \) and \( E_c \) defining the effect of temperature).

### 3.2.3 Corrosion rates of carbon steel during waste management

Literature data on the corrosion rates of carbon steels under different environmental conditions that may arise during waste management have been collated in a number of summary tables in Section 5 of reference [18]. In particular, this has included more recent data on the anaerobic corrosion of carbon steel carried out in support of the Belgian supercontainer project [53-57], for steel bar corrosion in concrete [e.g. 58] and for long-term atmospheric corrosion under a variety of atmospheric conditions [59], that has been published since 2007. In general, these data have not changed the view of the likely corrosion rates for carbon steel in the conditions expected during waste storage and disposal given in reference [39], with the exception of atmospheric corrosion for which corrosion rate data were not selected previously.

The most relevant data have been taken into account when selecting representative values for carbon steel corrosion rates and their temperature dependence under each set of conditions for use in gas assessment calculations. In reality, it is expected that carbon steel will experience a range of corrosion rates at each stage of waste management, depending on the exact nature of the chemical environment (e.g. that produced by the specific waste stream). The selected corrosion rates for use in the assessment are discussed below.

#### 3.2.3.1 Atmospheric corrosion

During storage and subsequently during the operational phase of a GDF, the carbon steels used in the packaging for some types of wastes will be exposed to the ambient indoor atmosphere, rather than being immersed in an aqueous phase. In this situation, corrosion may occur due to the presence of an adsorbed layer of moisture on the metal surface, which may form under conditions of higher humidity; a thin film of liquid water may condense if the temperature falls below the dew point. The corrosion rate is highly dependent on the composition of the moisture layer. The film may contain chloride deposited from the air if the steel is exposed in a marine environment, and/or sulphur oxides in an industrial environment. In humid marine atmospheres, corrosion rates up to 1200 \( \mu \text{m yr}^{-1} \) may be experienced. However, corrosion rates are much lower in unpolluted, rural atmospheres and are typically in the range 2.5-50 \( \mu \text{m yr}^{-1} \) depending on the exposure time, degree of shelter (extent of exposure to sun, wind and rain) and type of climate. Even lower rates may be expected under controlled indoor conditions typical of a GDF where temperature and humidity can...
be expected to be stable, thereby avoiding periodic condensation on and drying of the steel surfaces. In general, atmospheric corrosion rates measured in longer-term studies are observed to decrease with time due to passivation.

An atmospheric corrosion rate of 10 \(\mu\text{m}\text{ yr}^{-1}\) has been selected as a representative long-term atmospheric corrosion rate for carbon steels under storage conditions based on longer-term corrosion rates measured in rural locations [e.g. 59]. This is considered to be a conservative bounding value for corrosion in an indoor location.

There is a lack of atmospheric corrosion rate data for temperatures greater than about 30°C. However, it is considered unlikely that temperature would have a significant direct effect on the atmospheric corrosion rate (in comparison to other factors discussed above) within the temperature range expected during ILW storage or GDF operation (e.g. 0-50°C).

Atmospheric corrosion may also affect the surface condition of irradiated carbon steel wastes before they are packaged and prior to emplacement in a GDF. However, no account is taken of prior corrosion in the gas assessment.

### 3.2.3.2 Aerobic corrosion in grout

There are few data relating to the short-term corrosion rate of carbon steel in cements. However, Arya and Xu [60] have reported electrochemical data showing an exponential decrease in corrosion rate with time for a number of different cement mixes containing 1% or 3% chloride. The initial peak corrosion rate was of the order of 200 \(\mu\text{m}\text{ yr}^{-1}\). The decay in corrosion rate was considered to reflect both the reduction in the availability of water during curing and the passivation of the steel surface. Passivation typically took about 20 days in 1% chloride tests compared to 80 days in 3% chloride tests. In the absence of chloride, it is probable that passivation would be complete within a few hours.

The precise corrosion rate will depend on the chemistry of the specific cement type and the waste stream, however, an acute initial corrosion rate of 200 \(\mu\text{m}\text{ yr}^{-1}\) at ambient temperature and a characteristic time of 0.003 years (~1 day) were selected as representative values.

In the absence of chloride and carbonation effects, the corrosion rate of carbon steel in cementitious environments is very low owing to the protection offered by the passive film formed in alkaline conditions. A long-term corrosion rate for carbon steel in grout of 0.1 \(\mu\text{m}\text{ yr}^{-1}\) at ambient temperature was selected for use in SMOGG on the assumption that the chloride concentrations and effects of carbonation would be insignificant during storage and GDF operational phases. There is a limited amount of data relating to the effect of temperature on the corrosion rate of carbon steel in concrete because most experimental studies have been performed at ambient temperature. However, Liu and Weyers [61] carried out a systematic 5-year study of the corrosion rate of steel in concrete as a function of temperature (0-35°C). The corrosion rate was found to vary with temperature according to an Arrhenius relationship with an activation energy of 19 KJ mol\(^{-1}\). In the absence of any information on the temperature dependence of the acute corrosion rate of carbon steel in cements, this activation energy has been selected for both acute and long-term stages of carbon steel corrosion in grout [39].

### 3.2.3.3 Anaerobic corrosion alkaline conditions

Following backfilling and closure of a GDF, the disposal vaults will resaturate and conditions will change from aerobic to anaerobic as the available oxygen is consumed. Steel wastes and containers will have undergone corrosion under aerobic conditions and it is recognised that there is likely to be an induction period before anaerobic corrosion initiates, as shown by Smart et al. [62] and Fujisawa et al. [63]. For pre-corroded material it is reasonable to assume that anaerobic corrosion will initiate at a low level; it is unlikely that there will be an initial peak in the anaerobic corrosion rate, characteristic of an uncorroded surface.

Studies of uncorroded carbon steel exposed to anaerobic alkaline solutions have shown that the initial corrosion rate depends on temperature and follows an Arrhenius type relationship [62]. The data give an acute corrosion rate of approximately 0.5 \(\mu\text{m}\text{ yr}^{-1}\) at a temperature of 30°C and an
activation energy of 56 KJ mol\(^{-1}\), which have been selected as representative values. A conservative value for the characteristic time of 0.5 years was selected on the basis of carbon steel corrosion data in calcium hydroxide solution at 50°C [39].

The most relevant data for the long-term anaerobic corrosion rate of carbon steel is the gas evolution data obtained in the UK [62], in Japanese work [63, 64, 65] and in recent work for the Belgium national programme on the anaerobic corrosion of the carbon steel overpack in the concrete buffer of their Supercontainer concept [53-57]. Work on the Belgian programme has examined the corrosion behavior of carbon steel at pH 13.4 in concrete and in aqueous environments as a function of temperature, chloride concentration, irradiation (up to 25 Gy hr\(^{-1}\)) and the presence of sulphur species (specifically sulphide and thiosulphate), to reflect the operating conditions in the proposed Belgian disposal concept. This programme has found little effect of radiation, chloride or sulphur species on the anaerobic corrosion rate; and the long-term corrosion rates have been found to be <0.1 µm yr\(^{-1}\).

Corrosion rates for carbon steel measured for durations in excess of 9 years [62], found a slow decrease in anaerobic corrosion rate with time due to the build-up of a corrosion product layer composed predominantly of magnetite. The anaerobic corrosion rate appears to be only weakly affected by the presence of chloride under alkaline conditions [62]. On the basis of calibrations carried out using these data with the SMOGG corrosion model, a long-term corrosion rate of 0.005 µm yr\(^{-1}\) was derived for alkaline, anaerobic conditions. It was not possible to distinguish whether the corrosion rate would fall below a value of 0.005 µm yr\(^{-1}\) with time, owing to the sensitivity limits of the techniques used to measure the corrosion rates. No effect of temperature on the long-term corrosion rate was discernible from the experimental data.

### 3.2.4 Corrosion rates of stainless steel during waste management

Stainless steel repassivates rapidly if the passive film is disrupted [66], so it is anticipated that there will not be a distinct period of rapid corrosion and that the long-term general corrosion rate will be low under the various environmental conditions identified above.

#### 3.2.4.1 Atmospheric corrosion

In general, there is a lack of atmospheric corrosion data for stainless steel indoors because the general corrosion rate is so low. On the basis of the data collated in reference [18] and the likelihood that stainless steel will experience an indoor environment where the time of wetness will be short, a uniform corrosion rate of 0.03 µm yr\(^{-1}\) was selected as being representative [18, 39]. This is probably conservatively high.

There are no relevant data relating to the effect of temperature on the atmospheric corrosion rate of stainless steel, but there is unlikely to be any significant effect of temperature on the atmospheric corrosion rate within the range expected during ILW storage or GDF operation (e.g. 0-50°C).

It is likely that some minor superficial pitting corrosion of stainless steel containers will occur during interim storage and the operational phase of the GDF before backfill emplacement. However, this process is not accounted for in the gas generation model because the rate of gas generation is considered to be very low. Some pitting corrosion of irradiated stainless steels wastes may also occur prior to packaging if the steels are exposed to an atmospheric environment. Any such pits would be present on the surface of the waste steels on packaging, however, the alkaline conditions in grout would passivate the stainless steels towards further localised corrosion. Any further propagation of pits would be expected to cease after the conditions become anaerobic, post-closure, as the corrosion potential of the stainless steel falls below the repassivation potential.

#### 3.2.4.2 Aerobic corrosion in grout

The aerobic corrosion rate of stainless steel in concrete has been measured to be very low, and available data show no significant impact of chloride loading of the grout on the steel corrosion rate. On the basis of long-term weight loss data reported by Treadaway et al. [67] a corrosion rate for stainless steel in aerated grout of 0.02 µm yr\(^{-1}\) at 20°C was selected as being representative. The corrosion rate is likely to remain constant with time.
There is evidence from passive current density measurements for stainless steel in alkaline simulated porewaters [68] for a temperature dependence of the aerobic corrosion rate under alkaline conditions. The data were found to follow an Arrhenius relationship with an activation energy of 50 kJ mol⁻¹.

3.2.4.3 Anaerobic corrosion in alkaline solutions

Anaerobic corrosion rates of stainless steel in alkaline solutions are very low and have proved difficult to measure in the past. On the basis of UK measurements in anaerobic alkaline conditions [62] (which provided only an upper limit for the corrosion rate) and comparable, independent Japanese work [69], a corrosion rate of 0.01 µm yr⁻¹ was selected as being representative in a previous review [39]. It was also concluded that there is no initial high corrosion-rate phase and that the corrosion rate is likely to remain constant in the long term.

Recently, RWMC in Japan have reported some experimental measurements [70, 71] on the corrosion rate of 18Cr-8Ni stainless steel under anoxic conditions in pH 12.5 solution at 30 to 80°C over test periods up to 720 days (Figure 5). The corrosion rate was measured using the hydrogen generation technique (the amount of gas released was measured using gas chromatography (GC)). The measured corrosion rates for 720 days exposure at 30, 50, 80°C were 0.00077 µm yr⁻¹, 0.0026 µm yr⁻¹, and 0.0075 µm yr⁻¹ respectively (i.e. the corrosion rate increases with temperature). The new data indicate a mean anaerobic corrosion rate of 0.0008 µm yr⁻¹ for 18/8 stainless steel at 30°C after two years exposure, about one order of magnitude lower than the value used in the 2012 gas assessment [19]. The Japanese data also show a decrease of the anaerobic corrosion rate under alkaline conditions with time, and a temperature dependence, neither of which could be discriminated previously. As a first approximation, the two-year data have been selected as being representative of the long-term corrosion rate, with linear interpolation of the effect of temperature.

By analogy with corrosion under grouted aerobic conditions, and in common with carbon steels, the presence of chloride is considered unlikely to affect the corrosion rate of stainless steels under anaerobic conditions.

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¹⁴ Recently published work has supported this very low corrosion rate for stainless steels under alkaline anoxic conditions [72]; a lower value of 0.0004 µm yr⁻¹ has been reported for a 18Cr-8Ni stainless steel at pH 12.5 and 30°C, determined by continuous measurement of hydrogen gas release over a 6.5 year period using a gas flow system.
Anaerobic corrosion of 304 (18-8) stainless steel in pH 12.5 solution under at 30 to 80°C: a) equivalent corrosion thickness; and b) average corrosion rate with time; a simple power law of the form \( \Delta W = a \times t^b \) was used to fit the data where \( \Delta W \) is the equivalent corrosion thickness (\( \mu m \)), \( a \) is the initial corrosion rate (\( \mu m \) day\(^{-1} \)), \( t \) is time (day) and \( b \) is an exponent to account for the decrease in corrosion rate with time (from reference [70] with permission from RWMC).
3.2.5 Effects of irradiation on corrosion

In the preceding sections, the corrosion of unirradiated materials has been considered. The potential impact of neutron irradiation on reactor steels and their resulting structural damage and activation also need to be considered.

Irradiation of reactor steels can potentially affect corrosion processes in two main ways. Neutron irradiation can change the properties of the steels themselves, by for example inducing segregation of trace elements in the matrix [73, 74]. This can alter the corrosion susceptibility of particular locations in the microstructure of the material. In the context of stainless steel AGR cladding material, for example, neutron irradiation can lead to the development of radiation-induced depletion of chromium at grain boundaries, which renders the material more susceptible to intergranular corrosion (IGC) [75], a process known as sensitisation.

Secondly, as a result of neutron activation, the waste steels themselves become a source of γ-radiation; the resulting γ-irradiation fields can cause radiolysis of an aqueous phase, generating reactive species such as radicals and peroxide. These reactive species can affect the chemical and electrochemical processes that are involved in the corrosion reactions and potentially change the corrosion rate of the material.

The current understanding of each of these processes on the corrosion of steels is discussed in the sub-sections below.

3.2.5.1 Sensitisation

Sensitisation of stainless steel AGR fuel cladding has been studied because it is recognised as a potential failure mode for spent AGR fuel during handling and storage [75]. The mechanism of radiation-induced segregation (RIS) and the in-service conditions that promote sensitisation are well understood [see e.g. 76]. However, the effects of sensitisation on the corrosion of irradiated steels are understood only in broad terms.

The composition of the AGR fuel cladding was designed to provide a stainless steel with high temperature resistance through the addition of 0.7% niobium. The niobium is designed to capture carbon to form carbides in the steel and prevent the formation of chromium carbides at grain boundaries, which can give rise to thermal sensitisation. As a result, thermal sensitisation is not known to be an issue with AGR fuel cladding [76].

However, irradiation of the cladding by fast neutrons creates vacancies in the steel lattice which diffuse towards the grain boundaries [76]; a corresponding diffusion of elements away from the grain boundaries is induced. Segregation results from the differential rate of diffusion of alloying elements; chromium is the most rapidly diffusive species and so becomes depleted at the grain boundaries, whereas nickel, a less mobile element, becomes more concentrated. This process may lead to a reduction in chromium concentrations at grain boundaries below the threshold for the formation of a chromium oxy-hydroxide passive film. The depth into the grain over which segregation occurs may be only a few nanometers. Nevertheless, disruption of the passive film makes the steel more susceptible to IGC and to intergranular stress corrosion cracking (IGSCC) in certain environments. IGSCC can lead to the perforation of fuel cladding and the release of fission products from irradiated fuel, i.e. fuel failure.

The process of sensitisation occurs at cladding temperatures between about 350 and 520°C with a peak effect at about 420°C [77]; at higher temperatures the rate of chromium solid-state diffusion becomes sufficiently high to anneal out the inhomogeneities arising from segregation. The significance of this is that steels in the lowest two fuel elements in an AGR fuel stringer are most susceptible to sensitisation because, being close to the cooling gas inlets, they are subjected to the lowest operating temperatures, which happen to fall in a range below 520°C [76]. The higher fuel elements in the stringer (3-8, or 3-7 at Dungeness B), which are subjected to higher temperatures are less affected.
The spent AGR fuel is routinely discharged into reactor cooling ponds before transfer to Sellafield for reprocessing in THORP\textsuperscript{15}. Prior to reprocessing, the fuel stringers are dismantled and the fuel elements are placed into interim storage ponds.

The failure of sensitised fuel cladding has been found to occur by atmospheric corrosion or due to storage in water in the absence of corrosion inhibitors [75]. In particular, sensitised fuel is particularly sensitive to excursions in chloride concentration in storage ponds (occasional excursions are unavoidable in a coastal location), which may lead to fuel failure. However, the dosing of two of the three AGR fuel storage ponds at Sellafield\textsuperscript{16} with sodium hydroxide to pH 11.4 has been successful in effectively stopping fuel failure therein, presumably due to the much lower rates of stainless steel corrosion under alkaline conditions. Nevertheless, even with alkaline inhibition, corrosion will be continuing at a low rate; but there is some uncertainty as to whether the rate is faster for sensitised material than for unsensitised material. The actual corrosion rate of AGR cladding during storage under alkaline conditions has not been determined.

Based on data available up to 2007, Walters \textit{et al.} [78] estimated that of the spent AGR fuel then in pond storage, some 22\% was considered to be sensitised and 3\% was in a failed state. However, failed fuel is prioritised for reprocessing and operations since 2008 are thought to have significantly reduced the inventory of failed fuel in pond storage at Sellafield [78].

The focus of the studies to date has been on the sensitisation of cladding, but it is noted by Hill and Tice [76] that the braces of some fuel elements (from elements 3 and 4), which comprise part of the AGR fuel assembly component waste steam (2F08), may also be sensitised. In addition, other steel components of AGR reactors (and other reactor types) may be sensitised depending on their in-service temperature and irradiation history, however, there is no information available on this topic. It is noted that the extent of chromium depletion from grain boundaries needs to be significant for the steel to become sensitised. Thus, not all irradiated steels (and not all AGR cladding) will be affected.

In the absence of measured corrosion rates for irradiated or sensitised steels, the impact of sensitisation is difficult to assess. Sensitisation promotes localised corrosion by intergranular attack. This can occur due to faster rates of corrosion initially over very small areas of the surface at grain boundaries. As faster corrosion proceeds along sensitised grain boundaries, this is likely to result in the opening up of intergranular porosity and, potentially, to IGSCC of the steel matrix. These processes would result in additional, intergranular surface area becoming accessible for general corrosion.

Where thin materials, such as AGR cladding, are sensitised, it would be expected that the stainless steel would be sensitised through its full thickness. As a result, over time, some intergranular corrosion and IGSCC may be expected to develop through the full thickness of the material, as observed with failed fuel.

While corrosion may be accelerated along sensitised grain boundaries, the effect of sensitisation on the corrosion of the stainless steel grains would be much more limited. A sensitised surface that is depleted in chromium may undergo more rapid corrosion than an unsensitised surface initially. But as corrosion proceeds, and the depleted layer is corroded, the concentration of chromium at the corroding surface will increase and eventually exceed the threshold for the chromium oxy-hydroxide passive film to become re-established. Thereafter, the corrosion rates at grain surfaces might be expected to return to very low levels typical of unirradiated stainless steels, albeit over a wider exposed surface area.

\textbf{3.2.5.2 Radiolysis}

In a recent review of the effects of γ-irradiation on corrosion of nuclear fuel and container materials, Čuba \textit{et al.} [79] have stated that “\textit{... there is [only] a small effect of irradiation on corrosion of...}”

\textsuperscript{15} Thermal Oxide Reprocessing Plant.

\textsuperscript{16} The third pond also contains light water reactor fuel, which is stored in aluminium containers. Because these containers would corrode rapidly under alkaline conditions, the pond contains demineralised water only [78].
stainless steel, especially under unaerated conditions”. In contrast, carbon steels and cast irons are more susceptible. Accelerated corrosion of mild steel due to radiolysis has been observed in neutral solutions under aerobic and anaerobic conditions, but there is little evidence that there are significant effects under anaerobic alkaline conditions typical of post-closure conditions in cement-based disposal concepts. Under near-neutral conditions for example, Reda et al. [80] observed intergranular attack of mild steel irradiated in brine solutions, and Marsh and Taylor [81] report accelerated corrosion of 0.2% carbon steel in argon-purged synthetic granitic groundwater (pH 9.4) and seawater exposed to γ-radiation.

A review of potential effects of γ-radiolysis on the carbon steel overpack proposed for use in the Belgian Supercontainer concept was undertaken by Bennett [82]. The review concluded that the potential effects would be small, but recommended further work under concept-specific experimental conditions. Subsequent work on the Belgian programme [54, 55] has confirmed that there is little effect of radiation (at up to 25 Gy hr⁻¹) on the anaerobic corrosion rate of carbon steel at pH 13.4; long-term corrosion rates were found to be <0.1 μm yr⁻¹.

3.3 Release of carbon and carbon-14

The review of carbon-14 releases from steels in reference [18] sought to include all available information on the releases of stable carbon (carbon-12 and carbon-13) from irons and steels to supplement the very limited information available on carbon-14 releases from irradiated steels. In fact, the review has found that information on the chemical forms of carbon released from steels by corrosion is also limited and the processes involved are not well understood.

The speciation of carbon releases from steels upon corrosion is likely to be determined both by the chemical form of the carbon in the steel and the corrosion conditions, in particular the availability of oxygen. In the CAST literature review [18], the range of possible carbon-14 species in irradiated steels has been considered to evaluate the range of carbon-14 species that could be released from irradiated steels by corrosion.

3.3.1 Chemical forms of carbon and nitrogen in steels

Carbon and nitrogen are soluble to some extent (as a solid solution) in the iron phases that comprise the bulk of a steel: austenite in 300-series stainless steels and ferrite in mild steel. Nitrogen may also be present in distinct phases as nitrides of iron and the alloying metals (e.g. niobium, manganese); carbon may also be present as metal carbides. Carbonitrides may also form with iron and many alloying elements. In general, the stainless and mild steels used in nuclear reactor applications have low carbon contents, generally <0.3wt% carbon, and do not contain carbon in graphite form [83]. The initial distributions of nitrogen and carbon between different species in a steel will depend on the steel composition and its production process.

The distribution of carbon between different phases (metal carbide phases, cementite, etc.) in steels was not included within the scope of the CAST literature review. However, work has been undertaken at Loughborough University for RWM on the distribution of carbon and nitrogen in unirradiated samples of steels used in the UK nuclear programme [84]. In particular, it was found that two titanium-stabilised stainless steels (including the 20/25 Cr/Ni steel used in AGR fuel assemblies) contained carbon and nitrogen predominantly in the form of titanium carbonitride particles.

There is considerable uncertainty whether carbon-14 produced by activation of nitrogen-14 will be present in the same chemical forms in irradiated steels as the carbon present in the steels at the time of their production; the effects of nuclear displacement processes, thermal diffusion and radiation-induced segregation of alloying components of steel may all contribute to a redistribution of the carbon species.

3.3.2 Potential reactions of carbon species in steels with water

The reactivity of the different carbon species that may be present in a steel on contact with water varies considerably [85]. Some alloying components (e.g. niobium, titanium) form very stable interstitial carbide phases that react only slowly with acids, releasing methane and hydrogen. In
contrast, the electropositive metals (e.g. aluminium) form reactive carbides that are essentially ionic in character, rapidly releasing hydrocarbons related to the nature of the carbide ion present (acetylene in the case of calcium carbide, CaC$_2$) on contact with water. The smaller-sized transition metal elements (e.g. iron, manganese) form so-called intermediate carbides in which carbon atoms may form chains through a distorted metal lattice; multiple carbide stoichiometries are possible and these carbides may be hydrolysed by water and dilute acids, releasing a range of hydrocarbon species and hydrogen. In general, carbonitride phases tend to be more resistant to hydrolysis than the equivalent carbide phase.

On the basis of the known information on the reactions of metal carbides and carbonitrides with water, the following carbon-14 release behaviour might be expected on leaching an irradiated steel.

- As the steel slowly corrodes, the carbon-14 in its different chemical forms will be exposed to the waste disposal environment; different forms of carbon-14 may be expected to be released in different ways.
- Dispersed ionic and intermediate carbide phases and interstitial carbon present in the iron phase may be hydrolysed over relatively short time periods to form methane, and other hydrocarbons.
- Intermediate carbonitrides may be hydrolysed at a much slower rate than the equivalent carbides to yield a range of products that may include hydrocarbons and compounds containing C-N bonds such as amines and cyanides.
- Stable carbides and carbonitride phases may react only very slowly (if at all) and may be released in particulate form.

3.3.3 Inactive studies of carbon release from irons and steels

There is relatively little published information on the release of carbon from irons and steels and the processes involved; the possible range of carbon species released is not well understood. Current understanding can be summarised as follows:

- Experiments on inactive iron-water systems have shown clear evidence for the release of carbon as gaseous hydrocarbon species as a result of the hydrolysis of carbide species in the iron. A variety of species ranging from C1 to C5 hydrocarbons have been identified in the gas phase in separate studies [86, 87], and quantitative conversion of carbide carbon to hydrocarbons has been reported [87]. The mechanism of hydrocarbon formation is proposed to occur through processes similar to those that occur in Fischer-Tropsch synthesis [87]; it is suggested by Deng et al. [87] that the carbide species exposed at the iron surface may be similar to the intermediates formed on the surfaces of metal catalysts in the Fischer-Tropsch synthesis.
- In contrast, in Japanese experiments on inactive carbon steel and iron carbide, which focused on releases to the solution phase, carbon releases were presented as arising primarily as water soluble organic species, although some of the carbon in solution was found to be inorganic [88]. A range of low-molecular weight organic species were identified on the basis of high performance liquid chromatography, although these identifications must be treated as tentative. Carbon releases from iron carbide were identified as both inorganic and organic, which appears to be at odds with the results of iron-water studies.

The CAST literature review has highlighted the lack of well-documented inactive studies of the release of stable carbon from low-carbon and stainless steels that include measurements of carbon releases to both the gas phase and to the solution phase under anaerobic alkaline conditions. Further inactive studies of carbon release from irons and steels are in progress under CAST Work Package 2 (WP2), and in supporting in-house work by CAST participants [70]. These studies are designed to inform the development of advanced methods for determining the speciation of the much lower concentrations of carbon-14 released from irradiated steels in the corrosion experiments planned under CAST WP2.

Recent work carried out by the Paul Scherrer Institute in Switzerland [70], has sought to repeat the types of experiments performed by Deng et al. [87] but include both gas phase and liquid phase analysis of the carbon species released from cast irons. These studies have detected both...
gas-phase hydrocarbons and more oxidised dissolved organic species (carboxylic acids) when iron filings are immersed in water and in high-pH, cementitious solutions. However, the release of the carboxylic acids was found to arise as an initial fast release on immersion in solution, with little further release detectable beyond 3 days. In addition, higher concentrations of carboxylic acids were detected in leachates from an iron powder that had been pre-treated by acid washing (to remove residues from manufacture) compared to an untreated iron. Two possible explanations for the fast, transient release of dissolved organic species during the initial stages of iron corrosion have been proposed:

- that the carboxylic acids may be present as contaminants on the iron samples, arising in particular during the pre-treatment process, which are released rapidly on immersion in aqueous solutions; or

- that the carboxylic acids are formed rapidly and simultaneously with the reduced carbon (hydrocarbon) species due to the presence of residual oxygen in solution, until the oxygen is consumed.

Further work is in progress at PSI to distinguish between these two hypotheses.

These observations provide a possible explanation as to why carbon releases were reported to occur primarily as water-soluble organic species in the Japanese experiments.

### 3.3.4 Studies of carbon-14 release from irradiated steels

The information on carbon-14 release from irradiated steels is sparse; only three experimental studies have been performed to date, all in Japan. Unfortunately, insufficient information is available to allow a full evaluation of these studies. The findings are outlined below, but require corroboration:

- In an experiment in which a sample of irradiated stainless steel from the upper grid of a Boiling Water Reactor (BWR) was leached in a pH 10 cement-equilibrated water [88, 89], carbon-14 was reported to be released to the solution phase as a mixture of inorganic (25-34%) and organic (66-75%) species. No measurements were made of gas phase releases, and no information is available concerning the amount of carbon-14 released.

- In experiments with irradiated metals where the sample was first acid-cleaned to remove the passivating oxide film, carbon-14 is reported to be released from irradiated stainless steel, nickel alloy and zirconium alloy into solution at rates that are consistent with metal corrosion rates [90].

- In the most recent experiment reported [91], a small amount of carbon-14 was released to the gas phase on leaching irradiated stainless steel in alkaline solution for 42 months; the distribution of the 4.76 Bq released was reported to be 25% to the gas phase with a ratio of organic to inorganic carbon-14 of nearly one in the solution phase. Organic species in solution were reported to be predominantly in anionic forms (i.e. carboxylates) rather than neutral species (e.g. alcohols).

The findings are consistent with the inactive Japanese studies of carbon release from steels, but in common with the inactive studies there is no information on the time-dependence of the releases. Nevertheless, the results highlight the possibility that some of the carbon-14 may be released as water soluble organic and inorganic species rather than as hydrocarbons.

Experiments planned under CAST WP2 include a programme of work being undertaken by Amec Foster Wheeler/NRG to study the rates and speciation of release of carbon-14 to the gas phase from irradiated specimens of 316L(N) austenitic stainless steel under anaerobic alkaline (pH 13) conditions. Other experiments planned under CAST WP2 are designed to study the speciation of carbon-14 releases to both the gas and solution phase from irradiated steels; highly sensitive accelerator mass spectrometry techniques are being developed for speciation analysis [70].
3.4 Distribution of carbon-14 in irradiated steels

It is noted that the carbon-14 inventories of waste streams are derived from activation calculations that are based on average neutron fluxes for specific regions of the reactor structure and yield average specific activities for the irradiated wastes. This provides no information on the spatial distribution of carbon-14 within the waste steels, however.

The distribution of carbon-14 in irradiated steels will be dependent on the location of the steel components in a reactor. This is due to spatial variations in the neutron flux within the reactor and its periphery. Inhomogeneities in the distribution of carbon-14 may arise over varying length scales. For the assessment of carbon-14 release from irradiated steels, the most significant form of inhomogeneity is that which may arise normal to the corroding surfaces of the metal.

It can be envisaged that steel components present in the core of a reactor (such as fuel assembly components) will be subjected to a high neutron flux that may be similar in all directions. For relatively thin materials, this might be expected to result in a locally uniform distribution of carbon-14 through the material. Some large components (such as control rods) or small but abundant components (e.g. fuel assembly braces) may be subjected to variations in neutron flux with radial or axial position in the reactor; this may lead to variations in the extent of activation along the length of a component or a distribution of activities for abundant component types. Such variations may not be significant in terms of the overall carbon-14 release, where the specific activity is locally homogeneous perpendicular to the corroding surface. This is because the average specific activity as a function of depth remains constant.

However, components on the periphery of the reactor (such as thermal shield plates) will receive a directional neutron flux. Neutron absorption in the steel (i.e. self-shielding), will result in a reduction in the neutron flux with depth into the steel away from the irradiated face. This will result in a reduction in the extent of nitrogen-14 activation with depth into the steel and a non-uniform distribution of carbon-14 on a macroscopic scale. This may have implications for the rates of carbon-14 release from some types of steel waste.

Atomic displacements resulting from nuclear interactions and solid state diffusion of trace elements in the steel will tend to reduce inhomogeneities at the microscopic scale. The temperature of reactor operations will control the rate of solid-state diffusion of trace elements, in particular carbon and nitrogen. However, these processes are unlikely to remove inhomogeneities at the macroscopic scale.

Evidence for the inhomogeneity of activation of peripheral steel components has come from direct measurements of the carbon-14 content of a sample of steel from the thermal shield plates of Windscale Pile No. 2. Analysis of a series of sub-samples taken at increasing depths into the steel found that both cobalt-60 and carbon-14 were concentrated at the irradiated surface of the steel and decreased exponentially with distance from the irradiated face. A description of the work and the results obtained are provided in Appendix 1.

All calculations of carbon-14 release from irradiated metals to date [e.g. 19] have assumed a uniform distribution of carbon-14 through the steel. This assumption seems to be appropriate for the key waste streams that contribute most to the generation of carbon-14 bearing gases (e.g. 2F03/C and 2F08), which comprise the thinnest steel components from the reactor core (Table 8). Although options are available in SMOGG to specify the distribution of carbon-14 through a bulk metal, there is insufficient information on carbon-14 distributions in irradiated steels to develop an appropriate model for those waste streams for which non-uniform distributions might be important. However, the resulting additional contribution to the rates of carbon-14 bearing gas generation is expected to be modest.
3.5 Data Summary

3.5.1 Corrosion data – carbon steel

The standard set of parameters for mild steel corrosion selected for use in the assessment calculations is summarised in Table 10. These parameters are the same as those recommended previously [39] with the addition of parameters for atmospheric corrosion, which were not selected in the previous review. These parameters have been used in the SMOGG model [40] and in recent RWM gas assessments [19].

Table 10 Parameter values for treatment of carbon steel corrosion in the assessment of bulk gas generation and carbon-14 release

<table>
<thead>
<tr>
<th>Stage in SMOGG model</th>
<th>Corrosion parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Atmospheric</strong></td>
<td></td>
</tr>
<tr>
<td>acute</td>
<td>No acute stage</td>
</tr>
<tr>
<td>chronic</td>
<td>( k_c = 10 , \mu\text{m yr}^{-1} ) at 20°C, ( t_c \sim \infty ) \text{ years}</td>
</tr>
<tr>
<td></td>
<td>No temperature dependence: activation energy ( E_c = 0 , \text{kJ mol}^{-1} )</td>
</tr>
<tr>
<td><strong>Aerobic, grout, low chloride</strong></td>
<td></td>
</tr>
<tr>
<td>acute</td>
<td>( k_a = 200 , \mu\text{m yr}^{-1} ) at 20°C, ( t_a = 0.003 ) years</td>
</tr>
<tr>
<td></td>
<td>Arrhenius variation of ( k_a ): activation energy ( E_a = 19 , \text{kJ mol}^{-1} )</td>
</tr>
<tr>
<td>chronic</td>
<td>( k_c = 0.1 , \mu\text{m yr}^{-1} ) at 20°C, ( t_c \sim \infty ) \text{ years}</td>
</tr>
<tr>
<td></td>
<td>Arrhenius variation of ( k_c ): activation energy ( E_c = 19 , \text{kJ mol}^{-1} )</td>
</tr>
<tr>
<td><strong>Anaerobic, low chloride</strong></td>
<td></td>
</tr>
<tr>
<td>acute</td>
<td>( k_a = 0.5 , \mu\text{m yr}^{-1} ) at 30°C, ( t_a = 0.5 ) years</td>
</tr>
<tr>
<td></td>
<td>Arrhenius variation of ( k_a ): activation energy ( E_a = 56 , \text{kJ mol}^{-1} )</td>
</tr>
<tr>
<td>chronic</td>
<td>( k_c = 0.005 , \mu\text{m yr}^{-1} )</td>
</tr>
<tr>
<td></td>
<td>( t_c \sim (1) ) \text{ years}</td>
</tr>
<tr>
<td></td>
<td>No temperature dependence: activation energy ( E_c = 0 , \text{kJ mol}^{-1} )</td>
</tr>
<tr>
<td><strong>Anaerobic, high chloride</strong></td>
<td></td>
</tr>
<tr>
<td>acute</td>
<td>( k_a = 0.5 , \mu\text{m yr}^{-1} ) at 30°C, ( t_a = 0.5 ) years</td>
</tr>
<tr>
<td></td>
<td>Arrhenius variation of ( k_a ): activation energy ( E_a = 56 , \text{kJ mol}^{-1} )</td>
</tr>
<tr>
<td>chronic</td>
<td>( k_c = 0.005 , \mu\text{m yr}^{-1} )</td>
</tr>
<tr>
<td></td>
<td>( t_c \sim (1) ) \text{ years}</td>
</tr>
<tr>
<td></td>
<td>No temperature dependence: activation energy ( E_c = 0 , \text{kJ mol}^{-1} )</td>
</tr>
</tbody>
</table>

Notes: (1) In using the SMOGG model a finite value is required; a value much greater than the duration of the calculation should be used (e.g. \( 10^8 \) years).

3.5.2 Corrosion data stainless steel

The standard set of parameters for stainless steel corrosion that should be used in the assessment calculations is summarised in Table 11. These parameters for atmospheric and aerobic conditions are the same as those recommended previously [39], which have been used in the SMOGG model [40] and in recent RWM gas assessments [19]. The corrosion rate for stainless steel under anaerobic alkaline conditions has been reduced in the light of the new Japanese corrosion data [70] described in sub-section 3.2.4.3. In particular, the new data have shown a dependence of the anaerobic alkaline corrosion rate on temperature. There are insufficient data to determine an activation energy for stainless steel corrosion under these conditions. Therefore, the temperature dependence is included in the standard set of parameters, using the average corrosion rates measured after two years at 30°C and 50°C and assuming a simple linear interpolation between them.
### Table 11  Parameter values for treatment of stainless steel corrosion in the assessment of bulk gas generation and carbon-14 release

<table>
<thead>
<tr>
<th>Stage in SMOGG model</th>
<th>Corrosion data</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Atmospheric</strong></td>
<td></td>
</tr>
<tr>
<td>acute</td>
<td>No acute stage</td>
</tr>
<tr>
<td>chronic</td>
<td>$k_c = 0.03 , \mu m , yr^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$t_c = \infty , yr^{-1}$</td>
</tr>
<tr>
<td></td>
<td>No temperature dependence: activation energy $E_c = 0 , kJ , mol^{-1}$</td>
</tr>
<tr>
<td><strong>Aerobic, grout</strong></td>
<td></td>
</tr>
<tr>
<td>acute</td>
<td>No acute stage</td>
</tr>
<tr>
<td>chronic</td>
<td>$k_c = 0.02 , \mu m , yr^{-1}$ at 20°C</td>
</tr>
<tr>
<td></td>
<td>$t_c = \infty , yr^{-1}$</td>
</tr>
<tr>
<td></td>
<td>Arrhenius variation of $k_c$: activation energy $E_c = 50 , kJ , mol^{-1}$</td>
</tr>
<tr>
<td><strong>Anaerobic, alkaline</strong></td>
<td></td>
</tr>
<tr>
<td>acute</td>
<td>No acute stage</td>
</tr>
<tr>
<td>chronic</td>
<td>$k_c = 0.00077 , \mu m , yr^{-1}$ at 30°C</td>
</tr>
<tr>
<td></td>
<td>$k_c = 0.0026 , \mu m , yr^{-1}$ at 50°C</td>
</tr>
<tr>
<td></td>
<td>$t_c = \infty , yr^{-1}$</td>
</tr>
<tr>
<td></td>
<td>Temperature dependence of $k_c$ based on linear interpolation between data points</td>
</tr>
</tbody>
</table>

Notes: (1) In using the SMOGG model a finite value is required; a value much greater than the duration of the calculation should be used (e.g. $10^6$ years).

#### 3.5.3 Effects of irradiation

A significant proportion of stainless steel AGR fuel cladding (currently estimated as of the order of 25%) may be sensitised as a result of RIS and thus susceptible to intergranular attack. Other stainless steel reactor components, such as fuel stringer and fuel assembly components, may also be affected, depending on their irradiation history and in-service temperatures. However, there are no data available on the extent of sensitisation for these other reactor steels, nor are there any general corrosion rate data for irradiated or sensitised materials.

In terms of the impact of sensitisation on stainless steel corrosion, two potential effects can be distinguished: an accelerated rate of corrosion along sensitised grain boundaries and associated carbon-14 release along sensitised grain boundaries; and an increase in the exposed surface area of the steel available for general corrosion as a result of intergranular corrosion.

Considering the first potential effect, intergranular corrosion may occur over a sensitised grain boundary width of up to 10 nm compared to a typical grain size of about 10 μm. For intergranular corrosion occurring during alkaline storage conditions, if it is assumed that sensitised grain boundaries corrode at the same rate as carbon steel ($k_c = 0.1 \, \mu m \, yr^{-1}$ at 20°C, aerobic conditions in grout) then the net increase in the corrosion rate over this small proportion of the surface area would be about a factor of five compared to stainless steel ($k_c = 0.02 \, \mu m \, yr^{-1}$, aerobic conditions in grout). Thus, the net effect would be a very small increase in the total rate of carbon-14 release compared to general corrosion across the whole surface area.

However, as intergranular corrosion proceeds along sensitised grain boundaries it will result in more intergranular surface area becoming exposed. For thin components, such as AGR fuel cladding, where sensitisation may arise across the wall thickness, intergranular corrosion and IGSCC may eventually result in some cracks developing through the full thickness of the material...
(as observed for failed AGR fuel). This increase in accessible surface area for general corrosion, which would be cumulative, may be more significant in terms of the overall rate of carbon-14 release from the stainless steel than the intergranular corrosion itself. This overall effect may be scoped by either decreasing the effective thickness parameters for the stainless steel components (which increases the surface area per unit mass of material) or increasing the effective corrosion rates for the irradiated stainless steels by a scaling factor.

There may be an initial period of faster corrosion of sensitised material at grain surfaces depleted in chromium until the chromium oxyhydroxide passive film is re-established. However, as the thickness of the steel grains affected in this way may only be a few nanometers, most of this accelerated corrosion may be associated with the intergranular corrosion. Therefore, it is not considered necessary to include this transient, faster corrosion phase in an assessment model.

Results from the literature suggest that the effects of γ-irradiation, which leads to the formation of hydrogen peroxide and radicals in the aqueous phase, has a minimal effect on the corrosion of both mild steel and stainless steel under anaerobic alkaline conditions.

3.5.4 Carbon-14 release

There is currently a lack of well-documented studies on the rate and speciation of release of carbon-14 from irradiated steels under alkaline disposal conditions. There are also relatively few studies of the releases of carbon species from inactive steels and irons in aqueous solutions. However, there is evidence that some of the carbon present as carbide species within carbon steels and cast irons may be released as gaseous hydrocarbon species.

There is significant uncertainty concerning the chemical forms of carbon-14 in irradiated reactor steels, and whether carbon-14 derived from the activation of nitrogen-14 may be present in the same chemical forms as carbon present in the steels at the time of their production. It is also unclear to what extent carbon-14 may be present in releasable forms and the speciation of its release.

It has been noted also that the carbon-14 distribution in thick, irradiated steel reactor components subjected to directional neutron fluxes will not necessarily be uniform, and indeed an inhomogeneous distribution of activation products has been measured in a sample of thermal shield plate from Windscale Pile No. 2 (Appendix 1). Further information on the carbon-14 distributions in irradiated steel wastes from commercial Magnox and AGR reactors may be available from activation calculations that have been used to evaluate the carbon-14 inventory.
4 Modelling Carbon-14 Generation

This section outlines how the release of carbon-14 from steel wastes is treated in the source term model and how the model is parameterised.

The input data required include:

- The inventory of carbon-14 associated with different categories of steel wastes;
- The characteristics of the steel wastes (type of steel, mass, geometry and dimensions);
- The corrosion rates of steels under varying conditions over time in a GDF; and
- The chemical form of carbon-14 released from the steel.

In Section 4.1, the simple model for the release of carbon-14 from steel wastes is described. In the subsequent sections, the way in which the gas generation model is parameterised, based on the information collated in Sections 2 and 3, is outlined.

4.1 Modelling of carbon-14 release from steels

The mathematical model for the release of carbon-14 from steel wastes is a simple congruent source-term model. The overall inventory in the steel wastes is divided amongst a number of waste groups, as discussed in the sub-sections below. The typical shapes of the steel wastes have been considered, and the inventory assigned as either ‘plates’ or ‘spheres’ as appropriate.

The rate of release of carbon-14 bearing methane, $R_{i^{14}CH_4}$, is given by:

$$R_{i^{14}CH_4} = \sum_i \frac{\chi_i r_i F_i I_i(t)}{s_i}$$

where:
- $I_i(t)$ is the inventory of carbon-14 in waste group $i$ (TBq) at time $t$;
- $s_i$ is the mean (plate) thickness of waste type $i$ (m) – for wastes classified as spheres it is the radius;
- $F_i$ is a dimensionless shape factor for waste group $i$. It is 2 for plates (recognising they corrode from both sides). It is initially 3 for spheres, and decreases with time as the spheres corrode;
- $r_i$ is the corrosion rate of the type of steel in waste group $i$ (m yr$^{-1}$) given by equation (3); and
- $\chi_i$ is the fraction of carbon-14 released as $^{14}CH_4$ in waste group $i$ (-).

This is the model included in SMOGG [40]. It assumes the carbon-14 is uniformly distributed throughout the steel and that it is released as the metal corrodes. There is no information provided on the uniformity of carbon-14 distribution in the wastes in the inventory. As noted in Section 3.4, this may be a better approximation for certain waste streams than for others. For relatively thin wastes that have been subjected to a relatively high radiation fields, it is expected to be a reasonable approximation. For thicker materials, that have been subjected to higher radiation fields on one side (e.g. reactor pressure vessels), it may well be a less satisfactory approximation. Further consideration could be given to this as the inventory is refined further.
4.2 Treatment of the waste steels inventory

4.2.1 Treatment of the waste inventory

For the purposes of the gas assessment calculations, the inventory of all carbon-14 containing ILW/LLW wastes, including steel wastes, is considered. Before discussing how the inventory of carbon-14 associated with waste steels is treated in the assessment, it is first necessary to explain how the carbon-14 inventory is treated as a whole.

For the purposes of the modelling work, the ILW/LLW waste streams can be divided into five disposal categories as follows\(^{17}\), based on the type of waste package and thus the disposal vault in which they will be disposed:

- Legacy unshielded intermediate and low level waste (UILW / ULLW);
- Legacy shielded intermediate and low level waste (SILW / SLLW);
- Waste in DCICs;
- Nuclear new build (NNB) unshielded intermediate level waste;
- NNB shielded intermediate level waste.

The legacy wastes and the new build wastes are grouped separately because, although some waste from each group could potentially be emplaced in the same vault, the wastes will be emplaced at different times, so in practice will generally be emplaced in different vaults. The waste emplacement strategy assumed in the assessment calculations is outlined in the next sub-section.

Of these five disposal categories, modelling work has been carried out under the current project for Legacy UILW / ULLW, Legacy SILW / SLLW, and NNB UILW. Modelling was not carried out for waste in DCICs or for NNB SILW because the carbon-14 activities of the various materials are much smaller than the activities of the same materials in the other ILW, and the evolution of these packages is likely to be similar to other ILW packages\(^{18}\). Therefore, it is not expected that the rate of carbon-14 release from these wastes will contribute significantly to the carbon-14 release rate from the GDF overall.

The inventory data that are important for modelling the generation of carbon-14 containing and bulk gases include:

- The carbon-14 activity of each material from which it may be released;
- Mass and geometry data for the waste metals;
- Mass and geometry data for the container metals;
- Masses of the organic waste materials – which may also be a source of bulk gases; and
- Total radionuclide inventories – required to estimate the formation of gas by radiolysis.

In later sub-sections the data used in the assessment of gas generation for the waste steels and container steels will be summarised. For full details of the input data used in the modelling calculations, please refer to the modelling report [16].

\(^{17}\) This excludes spent fuel, which is included in the 2013 DI, and which will include irradiated steel components. Spent fuel is expected to be packaged in long-lived, unvented containers, that will isolate the wastes from the GDF environment for a sufficiently long time (i.e. ~50,000 years) to allow carbon-14 (and other radionuclides) to decay to much lower levels.

\(^{18}\) Together, the DCIC and NNB SILW disposal categories account for about 5 TBq of the carbon-14 inventory of steel wastes in the 2013 DI (see Appendix B in the Inventory Report [10]).
4.2.2 Waste emplacement strategy

The emplacement of wastes advised by RWM [92] differs depending on the waste group:

- For legacy UILW / ULLW emplacement starts at 2040 AD and occurs over 66 years. The first eight vaults are filled during the first 24 years of emplacement, and the remaining nine vaults are filled during the final 42 years. For simplicity this is represented in the model by assuming a 3 year emplacement period for each of the first eight vaults and a 4.5 year emplacement period for each of the remaining nine vaults.

- For legacy SILW / SLLW emplacement starts at 2040 AD and occurs over 73 years. Five vaults will be filled during the emplacement period. For simplicity this is represented in the model by assuming a 14.5 year emplacement period for each of the five vaults.

- For NNB UILW emplacement starts at 2100 AD and occurs over 40 years. Two vaults will be filled during the emplacement period. For simplicity this is represented in the model by assuming a 20 year emplacement period for each of the two vaults.

In practice, the emplacement will continue on a more or less continuous basis. For computational purposes the waste is emplaced in batches, with each batch representing the total emplacement during a half-year of the emplacement period.

GDF operations continue, after emplacement in the vaults for the above waste groups has been completed, until 2200 AD. During the final 10 years of operations (2190 – 2200 AD), backfilling and closure of all the vaults and the rest of the GDF occurs.

The radioactive component of the inventory is decay-corrected to 2040 AD for the legacy waste groups and to 2100 AD for the new build waste group. This means that 2040 AD or 2100 AD is assumed to be the time at which the waste is generated, with the inventory at 2040 AD or 2100 AD calculated so that radioactive decay results in the correct inventory being present at later times, when the waste actually arises. It is assumed, conservatively, that no degradation of the bulk waste (non-radioactive) materials occurs prior to its emplacement in the GDF, e.g. corrosion does not occur prior to emplacement of the waste. and there is no loss of carbon-14 (apart from radioactive decay).

The waste emplacement and backfilling strategy outlined here is the current baseline used in the UK GDF programme\textsuperscript{19}. This strategy provides the basis for the division of the assessment into separate time periods to represent the different stages of the GDF, as outlined in Table 9.

4.2.3 Data for gas generation calculations

4.2.3.1 Waste steel inventory data

To estimate the rate of gas generation due to corrosion of the waste steels, the mass and geometries of the steels are required. The mass data are obtained from the 2013 DI [24]. The approximated geometry data of the steel wastes (i.e. whether they are plate / sphere, and their effective dimensions) along with the division of masses between different geometries are provided in the Inventory Report [10].

It has been noted that there are a wide range of geometries of steel wastes. Analyses carried out during the project found that using an effective geometry for all the stainless steel and mild steel wastes resulted in substantial inaccuracies in the gas generation results calculated. Therefore, steel waste is modelled as several separate groups. Stainless and mild steels containing carbon-14 are modelled separately from other stainless and mild steels. Some waste streams comprising stainless steel containing carbon-14 are further separated because their shapes and thicknesses are significantly different.

\textsuperscript{19} It is noted here that changes in the timings of emplacement and backfilling would have negligible effect on the impact of carbon-14 from steel wastes due to the slow rates of steel corrosion under all conditions in a GDF. The impact of an alternative backfilling strategy has been considered as the scenario for a variant calculation, which is described in the Modelling Report [16].
The inventory data for waste steels\(^ {20} \) are summarised for each of the three disposal categories in Table 12, Table 13 and Table 14.

It will be noted that the steel waste material groups adopted for the modelling calculations are different from those used in characterising the steel waste carbon-14 inventory in the Inventory Report [10] outlined in Section 2. This has arisen because of the need to split the legacy waste streams between shielded and unshielded wastes for the assessment and to treat waste streams with significantly different geometries and thicknesses separately. Thus, for the legacy wastes, the two key carbon-14 containing stainless steel waste streams, 2F03C and 2F08, have been treated separately due to their smaller thicknesses, but all other stainless steel waste streams are grouped together as either UILW / ULLW or SILW / SLLW, and assigned an effective thickness. The effective thickness was calculated as a weighted average thickness for all contributing waste streams, based on the information collated in the Inventory Report [10]. In a similar way, three NNB UILW waste stream groups are treated separately in the assessment based on their different mean thicknesses (Table 14).

All of the stainless steel wastes are best represented by a plate-like geometry. However, some of the mild steel wastes are better represented by a sphere-like geometry with an effective radius.

### Table 12  Steel waste mass and geometry data for Legacy UILW / ULLW based on the 2013 Derived Inventory

<table>
<thead>
<tr>
<th>Waste material group</th>
<th>Total mass in plate (kg)</th>
<th>Effective plate thickness (m)</th>
<th>Total mass in sphere (kg)</th>
<th>Effective sphere radius (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless steel: 2F03/C</td>
<td>9.34\times10^5</td>
<td>4.00\times10^{-4}</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Stainless steel: 2F08</td>
<td>2.35\times10^5</td>
<td>8.00\times10^{-4}</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Stainless steel: containing carbon-14 (excluding 2F03/C and 2F08)(^ {11} )</td>
<td>2.61\times10^6</td>
<td>1.60\times10^{-2}</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Stainless steel: not containing carbon-14</td>
<td>2.87\times10^7</td>
<td>6.75\times10^{-3}</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Mild steel: containing carbon-14</td>
<td>8.35\times10^6</td>
<td>1.34\times10^{-2}</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Mild steel: not containing carbon-14</td>
<td>2.98\times10^7</td>
<td>1.48\times10^{-3}</td>
<td>1.05\times10^5</td>
<td>1.25\times10^{-2}</td>
</tr>
</tbody>
</table>

\(^ {1} \) Includes Nimonic.

\(^ {20} \) Nimonic (which has a significant carbon-14 inventory) is also modelled as stainless steel assuming the same geometry. The mass and carbon-14 inventories are included in the stainless steel waste material groups for UILW / ULLW and SILW / SLLW with appropriate adjustments to account for the difference in density and reaction stoichiometry.
Table 13  Steel waste mass and geometry data for Legacy SILW / SLLW based on the 2013 Derived Inventory

<table>
<thead>
<tr>
<th>Waste material group</th>
<th>Total mass in plate (kg)</th>
<th>Effective plate thickness (m)</th>
<th>Total mass in sphere (kg)</th>
<th>Effective sphere radius (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless steel: containing carbon-14 (1)</td>
<td>2.29×10^6</td>
<td>1.00×10^{-2}</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Stainless steel: not containing carbon-14</td>
<td>6.20×10^5</td>
<td>8.14×10^{-3}</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Mild steel: containing carbon-14</td>
<td>1.08×10^7</td>
<td>1.25×10^{-2}</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Mild steel: not containing carbon-14</td>
<td>1.01×10^4</td>
<td>5.00×10^{-3}</td>
<td>3.62×10^6</td>
<td>5.00×10^{-3}</td>
</tr>
</tbody>
</table>

(1) Includes Nimonic.

Table 14  Steel waste mass and geometry data for NNB UILW based on the 2013 Derived Inventory

<table>
<thead>
<tr>
<th>Waste material group</th>
<th>Total mass in plate (kg)</th>
<th>Effective plate thickness (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless steel: AP301</td>
<td>8.67×10^5</td>
<td>1.00×10^{-2}</td>
</tr>
<tr>
<td>Stainless steel: EP302, EP303</td>
<td>1.29×10^6</td>
<td>1.79×10^{-1}</td>
</tr>
<tr>
<td>Stainless steel: AP01</td>
<td>3.30×10^4</td>
<td>5.00×10^{-3}</td>
</tr>
<tr>
<td>Stainless steel: not containing carbon-14</td>
<td>9.67×10^4</td>
<td>2.00×10^{-3}</td>
</tr>
<tr>
<td>Mild steel: AP302</td>
<td>1.84×10^6</td>
<td>2.00×10^{-1}</td>
</tr>
</tbody>
</table>

4.2.3.2  Container steels inventory data

The mass data for the metal in waste containers are obtained from the 2013 DI [24]. In common with the waste steels, to estimate the generation rate of gas due to corrosion of the container metals, the geometries of the various metal containers are required. These are approximated as plates and the effective thicknesses are provided in the 2013 DI [24].

The inventory data for container metals are summarised for each of the three waste categories in Table 15, Table 16 and Table 17.

Table 15  Metal container mass and geometry data for Legacy UILW / ULLW based on the 2013 Derived Inventory

<table>
<thead>
<tr>
<th>Material</th>
<th>Total mass (kg)</th>
<th>Effective plate thickness (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless steel container and stillage (1)</td>
<td>9.87×10^7</td>
<td>5.78×10^{-3}</td>
</tr>
<tr>
<td>Mild steel container (2)</td>
<td>3.01×10^6</td>
<td>6.00×10^{-3}</td>
</tr>
</tbody>
</table>

Notes:  
(1) A large fraction of the legacy UILW is expected to be packaged in 500 litre drums. These cannot be emplaced directly in the GDF, so will be emplaced in steel stillages, which will allow the packages to be stacked.  
(2) These comprise miscellaneous beta gamma waste store containers.
Table 16  Metal container mass and geometry data for Legacy SILW / SLLW based on the 2013 Derived Inventory

<table>
<thead>
<tr>
<th>Material</th>
<th>Total mass (kg)</th>
<th>Effective plate thickness (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless steel container</td>
<td>2.20×10^7</td>
<td>3.02×10^-3</td>
</tr>
<tr>
<td>Mild steel container (1)</td>
<td>2.96×10^5</td>
<td>2.50×10^-3</td>
</tr>
</tbody>
</table>

Notes:  (1) These comprise 6 m³ reinforced concrete boxes with mild steel collars.

Table 17  Metal container mass and geometry data for NNB UILW based on the 2013 Derived Inventory

<table>
<thead>
<tr>
<th>Material</th>
<th>Total mass (kg)</th>
<th>Effective plate thickness (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless steel container</td>
<td>3.63×10^6</td>
<td>5.17×10^-3</td>
</tr>
</tbody>
</table>

4.2.3.3  Carbon-14 inventory data

To model the release of carbon-14 from irradiated steels requires the distribution of carbon-14 between the waste material groups and the disposal vault type. The carbon-14 inventory present in each waste material group at 2200 AD has been provided in Section 2 (Table 8). For the assessment calculations, the waste group carbon-14 data presented in Section 2 have been distributed by material, geometry and disposal vault type, as shown in Table 18. These data are decay corrected to the earlier start time of the calculations to provide the input data required. Note that these corrections are minor given that timescales involved, 100 or 160 years, are small compared to the 5,730 year half-life of carbon-14.

Table 18  Effective assignment of carbon-14 activity by material type at the start of emplacement in disposal vaults

<table>
<thead>
<tr>
<th>Waste material group</th>
<th>C-14 Activity (TBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Legacy UILW / ULLW</td>
</tr>
<tr>
<td></td>
<td>Legacy SILW / SLLW</td>
</tr>
<tr>
<td></td>
<td>NNB UILW at 2100 AD</td>
</tr>
<tr>
<td>Stainless steel: 2F03/C</td>
<td>30.0</td>
</tr>
<tr>
<td>Stainless steel: 2F08</td>
<td>39.1</td>
</tr>
<tr>
<td>Stainless steel: AP301</td>
<td>–</td>
</tr>
<tr>
<td>Stainless steel: Other waste streams</td>
<td>85.2</td>
</tr>
<tr>
<td>Mild steel</td>
<td>42.8</td>
</tr>
</tbody>
</table>

4.3  Corrosion rate data

Recommended data for the corrosion of steels under the varying conditions experienced in a GDF, which are required for use in the assessment calculations, were presented in Table 10 and Table 11. Based on the selected temperature dependencies of the corrosion rates, the values
used in the assessment calculations are summarised in Table 19. The data are for high pH conditions (appropriate for grouted waste) at illustrative GDF temperatures of 35°C and 45°C, with separate data for aerobic and anaerobic conditions for mild (carbon) and stainless steels. Data for atmospheric conditions are also provided, as these are required for corrosion of the external surfaces of containers during GDF operations. Compared to Phase 1, lower corrosion rates are being used for stainless steel under anaerobic high-pH conditions; the resulting values are shown in Bold in Table 19.

4.3.1 Effect of irradiation history for stainless steels

A source of uncertainty in the corrosion rate data for irradiated stainless steel wastes concerns the potential effect of irradiation history on their corrosion behaviour and the potential for higher rates of corrosion for irradiated, waste materials compared to unirradiated materials. In particular, it is known that under certain irradiation conditions, sensitisation of some stainless steel reactor components may occur, which has the potential to cause accelerated intergranular corrosion of the steel and IGSCC (see sub-section 3.5.3). At the time of writing, there are no corrosion rate data available for irradiated stainless steel materials. Therefore, the corrosion rates selected for irradiated stainless steel wastes in the Reference Case assessment calculations are based on data for unirradiated materials. However, the potential effect of higher effective corrosion rates for irradiated stainless steel wastes is considered in a suite of variant calculations for steel wastes, which are presented in Section 7.

For the purposes of the variant calculations, an increase in the effective corrosion rates for all irradiated stainless steel wastes by a factor of ten is considered.

Table 19 Corrosion rates of steels used in the assessment of gas generation and carbon-14 release

<table>
<thead>
<tr>
<th>Metal</th>
<th>Conditions</th>
<th>Time scale</th>
<th>Corrosion rate (m yr(^{-1}))</th>
<th>Characteristic time (yr) (^{(1)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless Steel</td>
<td>Atmospheric</td>
<td>Chronic</td>
<td>3.00x10(^{-8})</td>
<td>3.00x10(^{-8})</td>
</tr>
<tr>
<td></td>
<td>Aerobic, high pH</td>
<td>Chronic</td>
<td>5.43x10(^{-8})</td>
<td>1.00x10(^{-7})</td>
</tr>
<tr>
<td></td>
<td>Anaerobic, high pH</td>
<td>Chronic</td>
<td>1.23x10(^{-9})</td>
<td>2.14x10(^{-9})</td>
</tr>
<tr>
<td>Mild Steel</td>
<td>Atmospheric</td>
<td>Chronic</td>
<td>1.00x10(^{-5})</td>
<td>1.00x10(^{-5})</td>
</tr>
<tr>
<td></td>
<td>Aerobic, high pH</td>
<td>Acute</td>
<td>2.92x10(^{-4})</td>
<td>3.69x10(^{-4})</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Chronic</td>
<td>1.46x10(^{-7})</td>
<td>1.85x10(^{-7})</td>
</tr>
<tr>
<td></td>
<td>Anaerobic, high pH</td>
<td>Chronic</td>
<td>5.00x10(^{-9})</td>
<td>5.00x10(^{-9})</td>
</tr>
</tbody>
</table>

Notes: (1) Chronic corrosion rates are assumed not to reduce with time, however, for modelling purposes this is approximated using a characteristic time of 10\(^8\) years so that the reduction is insignificant over the time scale of the calculations.

(2) This accounts for a potentially very short period (days) of acute corrosion of mild steel wastes and the inside surfaces of mild steel waste containers on emplacement. This is included for consistency with the treatment of reactive metal wastes. Please see the Modelling Report for further details [16].

4.4 Release of carbon-14

The carbon-14 in the steel wastes is assumed to be evenly distributed throughout the metal and to be released as the steel corrodes. Although options are available in SMOGG to specify the distribution of carbon-14 through a bulk metal, in the absence of information on the carbon-14 distribution in irradiated steel wastes, the distribution is assumed to be homogeneous.
As noted from the information collated in Section 3.3, there is considerable uncertainty about the chemical form(s) in which the carbon-14 will be released from irradiated steels. It is possible that the carbon-14 may be released as an aqueous species, be retained in solid phases or be released to a gas phase. Therefore in the gas generation calculations, no speciation is assigned to the carbon-14 released from steel wastes. Different assumptions about the gas phase speciation are scoped in the biosphere to assess the potential consequences of the carbon-14 releases:

- the release of carbon-14 to the gas phase as either 100% methane or 100% carbon monoxide is considered in the operational phase assessment;
- in the post-closure Reference Case, it is assumed that 10% of the carbon-14 is released to the gas phase as methane; two variant cases have also been developed where either 100% or 1% of the carbon-14 is released from steels as methane gas.

Experiments to provide information on the speciation of carbon-14 releases from irradiated steels under GDF conditions are underway on the CAST project. When the data from the experiments are available the assessment results below will be refined.
5 Revised Assessment Basis

In the assessment, a Reference Case is defined for calculating the rates of release of carbon-14 as gaseous species and bulk gases from a GDF. The Reference Case is based on an illustrative design for a GDF, and the way in which the facility will be operated (including the emplacement of wastes and the backfilling and closure of the facility), to provide a basis for assessing the potential consequences of the carbon-14 released as gas from a GDF over time. The Reference Case provides a baseline against which alternative design and operational scenarios for the GDF can be compared.

The Reference Case uses the best estimates for the inventory of carbon-14 containing wastes and of the various parameter values, such as the corrosion rates of waste metals, which are key inputs to the calculation of rates of carbon-14 gas generation. It also assumes all the carbon-14 is released as a gas, although variants with a lower proportion released as a gas are considered within the Reference Case.

The Reference Case is divided into a number of time frames to assess the potential consequences of carbon-14 gaseous release during the different stages of a GDF, from the operational phase through to the long-term post-closure period. The chemical conditions, in particular the temperature, redox conditions and availability of water, vary over the different stages of the GDF life cycle (Table 9) and these affect both the rates of bulk gas and carbon-14 containing gas generation from different waste materials (e.g. steels, uranium metal) over time.

The methodology outlined in Section 4 provides the basis for calculating the rates of carbon-14 release as gaseous species from irradiated steel wastes in the Reference Case. To understand the potential consequences of such carbon-14 containing gas generation, it is necessary to consider a range of potential scenarios by which the carbon-14 generated from all wastes may migrate to the biosphere over the different stages of the GDF life cycle and give rise to doses to potentially-affected groups (receptor groups) of the local human population.

This section outlines the models of the near field, a range of carbon-14 release and migration scenarios and the treatment of the biosphere, which together form the basis for the Reference Case assessment of the potential consequences of carbon-14 containing gases from a GDF.

5.1 Near-field model

The model used for the near field in the gas generation calculations is based on that for an illustrative disposal concept in a higher strength host rock [36]. This is characterised by a rapid re-saturation of the GDF near field following backfilling (with a permeable cementitious material) and closure\(^\text{21}\).

The waste emplacement strategy and the stages of a GDF considered in the assessment have been outlined in sub-section 4.2.2 (see also Table 9). Key parameters controlling the corrosion of metals in a GDF are the availability of water and oxygen, given in Table 9, which are briefly discussed in the sub-section below.

For releases of carbon-14 from a GDF to pose a risk to man, the carbon-14 containing gases released from wastes must first be entrained in a bulk gas phase and then migrate through the near field and reach the biosphere (Figure 2). During the operational period of the GDF, gases released from waste packages into the vault atmosphere will be vented to the surface via the GDF ventilation system (see sub-section 5.2.1). However, after closure the GDF will re-saturate with in-flowing groundwater from the surrounding host rock. For a bulk gas phase to be maintained in the GDF vaults, the bulk gas must be produced at a sufficient rate that the solubility of the gas in the groundwater that flows through the near field is exceeded. This will depend on a number of

\(^{21}\) In host rock environments with lower permeabilities the resaturation time may be significantly longer (e.g. 10,000 years in a lower strength sedimentary rock) or may not occur at all (e.g. in an evaporite). See reference [13] for further details.
factors, including the design and depth below surface of the GDF vaults, the permeability and head gradient of the host rock, the permeability of the engineered barrier system (in particular the backfill) and the in-situ temperature.

For the purposes of the assessment calculations described herein, the worst case assumption is made that a gas phase is formed in the GDF vaults post-closure and that entrained carbon-14 containing gases are able to migrate in a bulk gas phase into the geosphere. The exception to this is that all carbon dioxide released from the wastes is assumed to react with the cementitious grout and backfill materials within the near field and so does not migrate into the geosphere. As a result, carbon-14 that is released in the form of carbon dioxide is not considered in the calculations of post-closure release and migration to the biosphere.

A full discussion of the behaviour of bulk gases in the near field, including the retention of carbon dioxide due to reactions with the cementitious grout and backfill materials, is provided in the Gas Formation and Migration Report [13].

5.1.1 Near-field data

In addition to the waste inventory and container data (provided for steels in sub-section 4.2.3), SMOGG calculations require input data for the amounts of other materials in GDF vaults, in particular:

- The total mass of the wasteform;
- The mass of grout in the packages;
- The waste package void volume; and
- The backfill void volume.

The total mass of the wasteform and the mass of the grout in the package are required for the radiolysis calculations. The package void volume is used to calculate the amount of water in the packages available for consumption by gas generation reactions during the operational period and early post-closure period; the amount of water in the backfill is required to calculate the amount of oxygen available at closure.

The availability of water is particularly important for Magnox wastes, which consume water rapidly and for which corrosion may become limited by water availability during the operational period. However, for all other waste types the assumption is made that the voidage in the waste packages is 85% saturated with water on emplacement. For steel wastes, under aerobic conditions, no consumption of water due to corrosion would be expected during the operational period.

The amount of water present in the backfill at closure of the GDF was calculated assuming a 95% saturation of the backfill pore space. The oxygen present at closure was calculated assuming all of the void space in the GDF that is not water-filled is air filled, at 101 kPa (atmospheric pressure) and 35°C (background temperature in the vaults). For the case of a higher strength host rock, the re-saturation period is expected to be short; a duration of five years was selected to represent this period (consistent with the previous assessment [22] [19]). In addition, it is expected that most of the gas in the GDF would be driven out during re-saturation.

To allow all of the key features of the waste behaviour to be represented (i.e. so as not to be restricted by the model), the calculations for each disposal category were performed in a number of parts, according to the constituent waste material groups.

For some waste metal groups, the oxygen was calculated to be fully consumed by corrosive processes by the time of full re-saturation, such that corrosion had become anaerobic within the five-year re-saturation period. This included the stainless steel waste streams 2F03/C and 2F08. However, for other waste groups this would not be the case. Therefore, for these waste groups, the oxygen content at closure was adjusted so that it reduced to zero at full re-saturation to mimic

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22 For further information on the range of resaturation times in different host rock environments, please refer to the Gas Phase Formation and Migration Report [13].
the oxygen being driven out by the incoming water. This ensured that oxygen was no longer present in the GDF after re-saturation.

Further details are provided in the Modelling Report [16].

5.2 Release and migration scenarios

In this section the mechanisms by which carbon-14 bearing gases may migrate to the biosphere are outlined and a set of release and migration cases are defined for which effective dose rates and risks will be calculated using the generation rates of carbon-14 bearing gases. This covers both the operational and the post-closure periods.

In the absence of (i) a site for geological disposal of radioactive wastes and (ii) detailed information on GDF design, it is not possible to develop a single conceptual model for how carbon-14 bearing gases might migrate from a GDF to the biosphere. Rather, at this generic stage of the process for implementing geological disposal, it is appropriate to consider a range of gas migration behaviours, which will depend on hydrogeological details of the selected site. A number of illustrative geological settings are considered.

For calculations of doses during the operation period of the GDF, a similar approach is used for all geological environments.

This section first outlines the mechanisms by which carbon-14 bearing gases may migrate during the various stages of evolution of GDF. It then summarises the range of gas migration behaviours, and defines a set of cases that have been used to link the generation rates of carbon-14 bearing gases to an effective dose rate in the different stages of the evolution of the GDF.

5.2.1 Operational discharges

During the operational phase of the GDF the vaults and tunnels will be ventilated. Thus the GDF ventilation system will be the main pathway for aerial discharges from the underground facilities into the atmosphere through a discharge stack. The discharge stack height and location will be designed to meet the requirements specified in generic GDF technical specifications [93]. For the purposes of the assessment calculations described in this report, a stack height of 15 m has been assumed for consistency with previous assessments [94].

5.2.2 Post-closure release and migration of gas

Bulk gas that is formed in a GDF may dissolve in water, undergo chemical reactions or form a free gas phase. In the post-closure period, the amount of free gas and its migration through the engineered barrier system (EBS) and the geosphere depend strongly on site-specific conditions, and may also depend on the design of the GDF. The bulk gas could act as a carrier for carbon-14 bearing gases. Two important aspects of the geological environment are the amount of water available to dissolve the gas and the permeability and porosity of the host rock.

A small amount of gas could dissolve in groundwater in the EBS. This dissolved gas will undergo advection, along with the groundwater, and diffusion due to concentration gradients in the groundwater. The majority of the gas is expected to form a free gas phase in the EBS (although this may not be retained at longer times in certain environments). To avoid pressurisation, the EBS will be designed to allow migration of gases out of the vented waste packages and through any backfill.

One of the gases that will be generated within a GDF is carbon dioxide. However, the cementitious materials present in a GDF react with carbon dioxide. There is a substantial body of evidence supporting the view that this carbonation will occur [13]. The wastes that will contain significant amounts of carbon-14 (i.e. graphite, steels and reactive metals) are expected to generate only small amounts of carbon dioxide compared with the carbonation capacities of the cement materials that will be present in a GDF [13]. Therefore, it is reasonable to assume that the carbonation reactions will occur, and any carbon-14 bearing carbon dioxide that is generated will be immobilised as solid carbonate minerals. This carbon-14 will not be released at the surface and is
not included in the calculation of risk. Other gases such as carbon monoxide and methane have the potential to migrate into the geosphere, although this depends on site specific aspects of the host rock.

5.2.3 Gas migration through the geosphere

An important characteristic of a site will be the type of host rock for the GDF. The host rocks that have been, or are being, considered for the disposal of radioactive wastes include:

- Higher strength rocks (HSR; these are rocks, for example granite, where fluid would flow through discontinuities, i.e. fractures, in the rock);
- Lower strength sedimentary rocks (LSSR; these are low permeability sedimentary rocks, for example clays, where fluid would flow through the rock matrix); and
- Evaporites (examples include halite; these are formed when water evaporates from a trapped body of water containing dissolved salts).

The potential migration behaviour, in the geosphere, of gas generated from a GDF located in each of these host rock types is discussed in detail in the Gas Formation and Migration Report [13]. A brief summary is given in the sub-sections below.

5.2.3.1 Higher strength rocks

The illustrative disposal concept for a HSR anticipates that ILW/LLW will be emplaced in large vaults (with dimensions about 16m × 16m × 300m), which at a later time will be backfilled with a cementitious material. The use of a cementitious backfill implies that once a vault has been closed, any carbon-dioxide that is formed will react with the backfill (in a carbonation reaction) and will not be released into the surrounding geosphere. The other gases of interest (i.e. hydrogen, as well as carbon-14 bearing methane and carbon monoxide) could migrate into the geosphere.

The way in which gas may migrate away from a GDF will depend on details of the site. Typically, gas will migrate away from the GDF through fractures in the host rock. Buoyancy means that the gas will migrate vertically upwards, and the time taken for the gas to traverse a fracture network will be short. Then, the hydrogeological characteristics of the overlying rock will determine whether the gas is delayed or prevented from reaching the surface and will affect the area over which the gas could be released. As the gas migrates it may:

- be dispersed through a network of pathways to the surface;
- become trapped by overlying rocks of low permeability (e.g. a sedimentary cap rock);
- be channelled to the surface through pathways of high permeability (e.g. open faults); or
- dissolve in the groundwater (e.g. in an overlying aquifer) after migration.

A GDF located in an HSR environment would potentially have the shortest resaturation time, depending on the hydraulic conductivity of the host rock. Reference [13] estimates a resaturation time of 1 year for a hydraulic conductivity of $10^{-9}$ m s$^{-1}$ and 1,000 years for a hydraulic conductivity of $10^{-12}$ m s$^{-1}$. The SMOGG calculations reported later use a resaturation time of 5 years.

5.2.3.2 Lower strength sedimentary rocks

The illustrative disposal concept for a LSSR anticipates that ILW/LLW will be emplaced in vaults (with dimensions about 10m × 10m × 100m), which at a later time will be backfilled with a cementitious material. Similarly to a HSR, the use of a cementitious backfill implies that once a vault has been closed, any carbon-dioxide that is formed will react with the backfill (in a carbonation reaction) and will not be released.

In contrast to a HSR, a LSSR would have a very limited density of fractures, and therefore gas would be unable to migrate into the geosphere so easily. Indeed, a major concern is to avoid high gas pressures in the GDF. In this disposal concept, the gas would migrate along the access shaft(s) of the GDF, before either being released to the surface as a gas phase or dissolving in the groundwater of an overlying aquifer. Numerical simulations of gas migration around a GDF
situated in a LSSR typically show some of the gas migrating through the host rock. The time scale for the gas to cross the host rock will depend on the rate of gas generation and the thickness of LSSR above the GDF, but most of the simulations suggest a time scale from thousands to tens of thousands of years. In a similar way to a HSR, the host rock could be overlain by a cap rock or an aquifer.

Resaturation will be slower in an LSSR environment and water ingress may be reduced or prevented by pressurisation of the GDF. Reference [13] estimates a resaturation time of 10,000 years.

5.2.3.3 Evaporite rocks

The illustrative disposal concept for an evaporite assumes that the ILW/LLW vaults will be about 10m wide × 5m high × 100m long. The vaults will be backfilled with crushed rock. A key feature of evaporite deposits is that they are dry and the GDF will not resaturate with water. Thus only the water present in the waste packages on emplacement is available for corrosion processes. This may limit the amount of gas generated in the EBS.

In general, evaporite deposits are gas tight, with very high entry pressures for gas migration. In this geological environment, the safety argument is that the host rock will become more permeable to gas as the pressure increases in the EBS to the lithostatic pressure. Gas will migrate a small distance into the surrounding rock, sufficient to reduce the pressure in the vaults, but would not migrate further. A zone of gas-pressurised rock will develop with gas becoming stored within the salt matrix. Thus, gas generated in a GDF within an evaporite host rock would be confined to the host rock immediately surrounding the GDF and would not reach the biosphere.

5.2.4 Post-closure cases considered

The calculated post-closure consequences of carbon-14 are dependent on the migration time relative to the half-life of carbon-14 and the area over which the carbon-14 is released to the deep soil zone. Based on the range of possible gas migration behaviours away from a GDF outlined above, six illustrative cases have been developed to enable the implications of the release of carbon-14 to be assessed; they include a case where there is no release of carbon-14. These cases are not exhaustive, but they span the geological environments that may be considered in the future to host a GDF. The six post-closure cases are described in Table 20. Each case considers a delay time and release area for carbon-14. Although single values of these parameters are presented, there will be uncertainty in the values. This broad-brush treatment is considered appropriate at this generic stage of the programme to develop a GDF. Given that there is no specific site currently being investigated, none of these cases is given more weight than any other.
<table>
<thead>
<tr>
<th>I.D.</th>
<th>Description</th>
<th>Delay Time ($^{14}$C $\frac{1}{2}$-lives)</th>
<th>Release Area ($m^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Higher-strength host rock where gas is released over an area similar in size to the footprint of the GDF ILW / LLW vaults</td>
<td>0</td>
<td>$10^6$</td>
</tr>
<tr>
<td>B</td>
<td>Environments where there is a low permeability formation limiting gas migration</td>
<td>No release</td>
<td>No release</td>
</tr>
<tr>
<td>C</td>
<td>Higher-strength host rock where there are features that focus the release of gas to an area much less than the footprint of the GDF ILW / LLW vaults</td>
<td>0</td>
<td>$10^4$</td>
</tr>
<tr>
<td>D</td>
<td>Higher-strength host rock where there are features that distribute the release of gas to an area greater than the footprint of the GDF, e.g. as a result of dissolution</td>
<td>0</td>
<td>$10^7$</td>
</tr>
<tr>
<td>E</td>
<td>Lower-strength host rock where gas migrates slowly across the lower strength sedimentary host rock and is released over the footprint of the GDF ILW / LLW vaults</td>
<td>1</td>
<td>$10^6$</td>
</tr>
<tr>
<td>F</td>
<td>Lower-strength host rock where gas migrates slowly across the lower strength sedimentary host rock and there are features that distribute the release of gas to an area greater than the footprint of the GDF, e.g. as a result of dissolution</td>
<td>1</td>
<td>$10^7$</td>
</tr>
</tbody>
</table>
5.3 Treatment of the biosphere

5.3.1 Operational period

In Phase 1 of the project, the approach to assessing the impact of operational discharges was to use the assessment methodology adopted in the 2010 Generic Operational Environmental Safety Assessment (OESA) [94]. In the light of the results, an updated methodology has been developed [15]. This methodology addresses some of the simplifications in the initial methodology that are not well tailored to assessing the particular nature of the release of carbon-14 being considered, and leads to lower calculated doses. In this improved methodology, $^{14}$CO$_2$, $^{14}$CO and $^{14}$CH$_4$ are treated differently.

The main pathway for aerial discharges from the underground facilities will be through the GDF ventilation system and into the atmosphere through a discharge stack. In Phase 2 of the project, a methodology has been developed to calculate the effective doses rates arising from a continuous discharge of carbon-14 containing gases from a 15m high stack$^{23}$ to a local resident family receptor group [15], which is cautiously assumed$^{24}$:

- to be located 100 metres from a discharge point of a GDF; and
- to consume food grown at 500 metres from a GDF.

This work has provided multiplication factors to convert releases of carbon-14 to dose, as shown in Table 21. In contrast to the factors used in the calculations presented in Phase 1, this work provides separate factors for carbon-14 as methane, carbon monoxide and carbon dioxide, because these have been identified as the three main carbon-14 bearing gases that could be released. Compared with the value of $1.8 \times 10^{-17}$ Sv yr$^{-1}$ per Bq yr$^{-1}$ used in Phase 1 for all carbon-14 containing gases, the value for methane is substantially lower, but the value for carbon monoxide is significantly higher.

Table 21 Effective carbon-14 release rate to dose rate conversion factors for off-site discharge to the local resident family receptor group during the GDF operational period (Sv yr$^{-1}$ per Bq yr$^{-1}$ for a release at a height of 15m)

<table>
<thead>
<tr>
<th>Age Group</th>
<th>Carbon dioxide</th>
<th>Carbon monoxide</th>
<th>Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Infant</td>
<td>$9.0 \times 10^{18}$</td>
<td>$1.2 \times 10^{16}$</td>
<td>$1.5 \times 10^{20}$</td>
</tr>
<tr>
<td>Child</td>
<td>$5.5 \times 10^{18}$</td>
<td>$7.1 \times 10^{17}$</td>
<td>$8.7 \times 10^{21}$</td>
</tr>
<tr>
<td>Adult</td>
<td>$5.5 \times 10^{18}$</td>
<td>$7.2 \times 10^{17}$</td>
<td>$9.0 \times 10^{21}$</td>
</tr>
</tbody>
</table>

The effective dose factors for carbon dioxide, carbon monoxide and methane listed in Table 21 exhibit approximate ratios of 1:13:0.003. Thus, if even a small fraction of the release of carbon-14 bearing gas is as carbon dioxide or carbon monoxide rather than as methane, this fraction will determine the radiological impact of the release. However, this result arises from a cautious assumption concerning the assimilation of carbon monoxide by plants at low atmospheric concentrations of the gas. The values given in Table 21 are for the case that the carbon-14 discharge is entirely one or other of the three carbon-14 bearing gases. In practice, carbon monoxide is likely to comprise at most a small fraction of the discharge, so the larger effective dose rates given for carbon-14 bearing carbon monoxide in Table 21 are likely to be of limited radiological significance.

$^{23}$ Design tasks are in hand that are giving further consideration to the appropriate stack height to adopt. The 15m value was selected for the 2010 OESA [94] and has been retained here for consistency, but the height to be adopted will be reassessed in the future and may be changed.

$^{24}$ These parameters were chosen to be consistent with the Environment Agency guidance [95, 96] and the 2010 OESA [94].
In addition, it is not expected that carbon-14 containing carbon dioxide would be released from waste packages during operations. Given the assumption that the steel wastes will be encapsulated in cementitious grout, it is expected that carbon dioxide will react with the grout to form calcium carbonate, preventing carbon-14 in carbon dioxide from being released.

The release rate to dose rate conversion factors given in Table 21 are combined with the gas generation rates calculated from SMOGG for the operational period. The calculated dose rates are compared with the radiological protection criteria for normal conditions of operation. The effective dose rate for a member of the public should be below the source-related dose constraint for members of the public for a new facility of 0.15 mSv yr⁻¹ (Requirement R5 in [97]). If this constraint is met then the requirement that the effective dose rate for a member of the public should be below the legal dose limit of 1 mSv yr⁻¹ will also be met.

The methodology for assessing operational discharges is applied to the pre-backfilling period. For releases following backfilling, there is some uncertainty as to whether they will be discharged through the ventilation system or whether they will be retained within the EBS until after closure (when the post-closure methodology would be applicable). Given the uncertainties for this stage, it is considered appropriate to apply both the updated OESA methodology (outlined above) and the post-closure methodology (outlined below).

5.3.2 Post-closure period

For the post-closure period, a single multiplying factor is used to convert the flux of carbon-14 containing gases at the surface to the dose rate received by a local family receptor group. The conversion factor, given in Table 22, is based on recent work on carbon-14 in the biosphere [98], and has been derived for carbon-14 released into the soil zone as methane. This value is slightly higher than the value used in Phase 1. As a value for carbon-14 containing carbon monoxide is not provided, the value for carbon-14 containing methane has been applied to the sum of the release rates of carbon-14 as methane and carbon monoxide. This is expected to be a cautious approach.

It is assumed that the carbon-14 containing carbon dioxide generated after closure carbonates the backfill in the vaults. Therefore this carbon-14 will not be released at the surface, so is not included in the calculation of risk.

The dose to risk conversion factor is 0.06 Sv⁻¹ [97].

<table>
<thead>
<tr>
<th>Carbon-14 containing gas</th>
<th>Conversion factor (Sv yr⁻¹ per (Bq m⁻²s⁻¹))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Methane</td>
<td>0.0225</td>
</tr>
</tbody>
</table>

The Environment Agency Guidance on Requirements for Authorisation (GRA) [97] has two requirements that are particularly relevant to the post-closure period:

- If there is a period of institutional control after closure, then the source-related dose constraint for members of the public for a new facility of 0.15 mSv yr⁻¹ applies (Requirement R5; see Section 5.3.1);
- After any period of institutional control, risks should be consistent with the risk guidance level of 10⁻⁶ (Requirement R6 [97]).
At this generic stage of the programme, there is uncertainty over the period of institutional control, and so for simplicity the risk guidance level requirement is applied throughout the post-closure period; this is more stringent than the source-related dose constraint.

In the modelling results presented in Sections 6 and 7, quantities described as ‘risks’ are calculated. It is recognised that this term is a simplification. These quantities are really assessed ‘conditional risks’ in which the occurrence of the scenario and exposure have been assumed to occur with a probability of 1.0. In addition the assessed ‘risk’ is just from carbon-14. There may be risks from other radionuclides, e.g. tritium and radon-222. In a full assessment it would be necessary to take account of any contributions to risk from other radionuclides.

5.4 Modelling calculations

5.4.1 Modelling gas generation

The overall modelling of the generation of bulk gases and carbon-14 bearing gases was undertaken with SMOGG v7.0 [40]. This version of SMOGG includes the most recent updates. SMOGG has been used for gas generation calculations over a number of years, most recently for the 2012 gas assessment [19] and the Phase 1 work of the Carbon-14 Integrated Project [1].

SMOGG takes into account:

- The generation of gas from a range of metals, present either as waste metal or packaging or other metals in the GDF (including the release of carbon-14 in neutron-irradiated metals as these corrode);
- Radiolysis, in particular of water and some organic materials;
- Microbial degradation of organic materials, including the prior hydrolysis of cellulose to smaller organic compounds;
- Diffusion, notably the release of tritium by solid-state diffusion from metals;
- Radioactive decay of radium, which leads to the generation of radon-222; and
- The release of radioactive gases containing tritium or carbon-14 by leaching of irradiated graphite.

Carbon-14 is the focus of the current study, and so the release of tritium and radium are outwith the scope of this work.

SMOGG also takes into account the availability of water and oxygen, and the evolution of temperature. The approach to the operation of the GDF, including the waste emplacement strategy, provides further important inputs.

The modelling basis of gas generation calculations from steel wastes presented in this report is documented in Table 24. A full description of the modelling basis in SMOGG is provided in the Modelling Report [16].

5.4.2 Modelling the consequences of carbon-14 releases

As outlined in Section 5.3, the way in which the potential consequences of the carbon-14 release are determined is different for the operational and post-closure periods. Therefore, the two periods are considered separately in the assessment. In particular it is noted that the GDF ventilation system will continue to operate during the backfilling period and during closure so that some carbon-14 releases may occur from unbackfilled and/or unsealed vaults into ventilated areas during these periods. Once the vaults have been backfilled and sealed, releases would occur, primarily, into the surrounding geological environment25.

25 It is possible also, that there may be a delay in the release of some gases from waste packages, so that some gas generated during the operational period may not be released until after GDF closure.
For the purposes of the assessment, a number of distinct time frames are considered that capture the major changes that occur in the GDF (cf. Table 9) and in the rates of carbon-14 release from the wastes with time, as summarised in Table 23. The table also identified the biosphere model (i.e. the flux to dose conversion factors) applied in each time frame.

**Table 23** Phases of GDF evolution, the time frames for gas generation and the carbon-14 exposure pathways considered in the Reference Case assessment

<table>
<thead>
<tr>
<th>Stage</th>
<th>Time (AD)</th>
<th>Exposure pathways</th>
<th>Biosphere model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emplacement</td>
<td>2040 – 2190</td>
<td>Atmospheric discharge via ventilation stack</td>
<td>Operational</td>
</tr>
<tr>
<td>Backfilling</td>
<td>2190 – 2210 (1)</td>
<td>Atmospheric discharge via ventilation stack</td>
<td>Operational</td>
</tr>
<tr>
<td>Early post-closure</td>
<td>2190 – 2230 (1)</td>
<td>Not established</td>
<td>Post-closure (2)</td>
</tr>
<tr>
<td>Medium-term post-closure</td>
<td>2230 – 3000</td>
<td>Migration to the soil zone</td>
<td>Post-closure</td>
</tr>
<tr>
<td>Long-term post-closure</td>
<td>3000 onwards</td>
<td>Migration to the soil zone</td>
<td>Post-closure</td>
</tr>
</tbody>
</table>

Notes: (1) The time frames for gas generation considered during the backfilling and early post-closure assessments overlap because post-closure processes (e.g. resaturation) will start during the backfilling period as individual vaults are backfilled and sealed and to account for the possibility that gas generated during backfilling is retained in the waste packages and vaults and so does not undergo immediate release via the ventilation system. Thus, the potential impact of gas generated during backfilling is also considered in the early post-closure assessment and vice versa.

(2) The potential implications of releases via the gas migration pathway are considered qualitatively given the short duration of the period of maximum gas generation from steels and noting that migration through both the EBS and the geosphere will act to smooth out short-term variations in gas generation rate.

In the operational assessment calculations, the generation rates of carbon-14 bearing gases determined using SMOGG are multiplied by the effective release rate to dose rate conversion factors (see sub-section 5.3.1) to calculate effective dose rates for releases of carbon-14 as carbon dioxide, carbon monoxide and methane.

In the post-closure assessment calculations, the generation rates of carbon-14 bearing gases are multiplied by an exponential decay term appropriate to the delay time, and then converted to fluxes to the biosphere by dividing by the release area (see sub-section 5.2.3). Finally the fluxes to the biosphere are multiplied by a surface flux to dose rate conversion factor (see sub-section 5.3.2) to calculate the effective dose rates, assuming that carbon-14 is released as methane.

The maximum rates of corrosion and thus the highest rates of carbon-14 bearing gas generation will be experienced from waste steels will arise for a short time during the early post-closure period, before conditions in the waste packages become anaerobic. Given the short duration of this period, and noting that migration through both the EBS and the geosphere will act to smooth out short-term variations in gas generation rate, risks calculated directly from the peak generation rates are not expected to be realised. Therefore the potential implications of carbon-14 bearing gas generation during the early post-closure period are considered only qualitatively in the assessment.
Table 24  Modelling basis – gas generation from steel wastes

<table>
<thead>
<tr>
<th>Model component</th>
<th>Aspect</th>
<th>Conceptual understanding</th>
<th>Model implementation</th>
<th>Data (including references)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Generation</td>
<td>Stainless steel and other ferrous metals</td>
<td>Corrosion of stainless steel and other ferrous metals</td>
<td>2013 DI [A2] divides between spheres and plates</td>
<td>Quantities of waste metals and shape data from 2013 DI [24] and project Inventory Report [10]</td>
<td>Assumes high pH Certain stainless steel waste streams modelled separately to represent the geometry better (e.g. 2F03/C and 2F08)</td>
</tr>
<tr>
<td>Generation</td>
<td>Stainless steel and other ferrous metals</td>
<td>Release of carbon-14</td>
<td>Assume 10% release as CH₄ (might be something else that is not CO₂), variants of 100% and 1% Uniformly distributed throughout metal</td>
<td>Carbon-14 data from 2013 DI [24] and project Inventory Report [10]</td>
<td>There is evidence that significant proportion of the release is to the solution phase, as a range of organic species and carbonate</td>
</tr>
<tr>
<td>Generation</td>
<td>Stainless steel and other ferrous metals</td>
<td>Bulk gas from wastes</td>
<td>Aerobic gives metal oxide (no bulk gas)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Generation</td>
<td>Stainless steel and other ferrous metals</td>
<td>Bulk gas from corrosion of stainless steel containers</td>
<td>Aerobic chronic corrosion rate (non-gas generating)</td>
<td>2013 DI [24] and project Inventory Report [10] give amounts of packaging and shape data</td>
<td></td>
</tr>
<tr>
<td>Generation</td>
<td>Stainless steel and other ferrous metals</td>
<td>Bulk gas from corrosion of mild steel components of containers</td>
<td>Aerobic acute and chronic corrosion rates (non-gas generating)</td>
<td>2013 DI [24] and project Inventory Report [10] give amount of packaging and shape data</td>
<td></td>
</tr>
</tbody>
</table>
6 Reference Case Calculations

In this section, the results from the Reference Case gas generation calculations are presented to evaluate the potential contribution of irradiated steel wastes to the generation of carbon-14 containing gases from a GDF over time. This section also provides an indication of the potential consequences of the gas generation from steel wastes in the GDF as a whole, in both the operational and post-closure periods.

6.1 Generation of carbon-14 bearing gases

In this section, the results from the Reference Case gas generation calculations are presented for the three waste groups from which the majority of the carbon-14 will be generated:

- Legacy UILW / ULLW,
- Legacy SILW / SLLW,
- NNB UILW.

All three waste groups contain significant quantities of irradiated steel wastes that may act as sources of carbon-14 containing gases.

For each waste group, a summary of the gas generation results over the different stages of a GDF is presented. These results include contributions to carbon-14 gas generation from reactive metals, graphite, steel wastes and from other sources of carbon-14, and bulk gas generation from metal wastes, steel waste containers and other sources of bulk gases present in a GDF. This gives an indication of the overall carbon-14 and bulk gas generation rates over time. To give an understanding of the contribution of steel wastes to carbon-14 gas generation from each waste group, the contributions from steel wastes are compared with other sources of carbon-14 at different times; an explanation is provided of the main features of the results.

For each waste group, a full breakdown of the carbon-14 and bulk generation rates over time, considering all contributing waste types, is discussed in the Modelling Report [16].

As noted in Section 4.4, there is considerable uncertainty about the chemical forms in which carbon-14 will be released from steel wastes; it is possible that the carbon-14 may be released as an aqueous species, be retained in solid phases or be released to the gas phase. Therefore in the gas generation calculations, it is assumed, conservatively, that all of the carbon-14 is released as gas, which is nominally labelled as methane. A similar approach is applied to the reactive metal wastes, where there are similar uncertainties concerning the chemical forms of carbon-14 releases.

In the modelling approach, the generation of gaseous carbon-14 from the degradation of graphite is presented as a combination of organic species (nominally labelled as methane), carbon monoxide and carbon dioxide. It is assumed also that all carbon dioxide generated will be immediately removed from the gas phase by carbonation of the cementitious materials in the waste packages and backfill. Therefore, in the results presented here, the generation of carbon-14 as carbon dioxide from graphite wastes is excluded, and only that proportion of the carbon-14 assigned as carbon monoxide or methane is assumed to be released as gas.

Using these approaches, the gas generation results presented in the following sub-sections represent the maximum potential generation of gaseous carbon-14 from the waste groups, and are therefore cautious.
6.1.1 Legacy UILW/LLW

To gain an understanding of the contribution of steel wastes to the generation of gaseous carbon-14 from the Legacy UILW / ULLW, a breakdown of the carbon-14 containing gas generation rates for different parts of the waste steels inventory is compared with other sources of carbon-14 in Figure 6 and Figure 7, for shorter and longer time scales, respectively. Here, the contributions from waste streams 2F08 and 2F03/C (AGR fuel assembly components and AGR fuel cladding) are separated from the other stainless steel wastes within this waste group.

In summary, using the modelling basis developed within the project, the main sources of carbon-14 containing gas generation for Legacy UILW / ULLW are [16]:

- Corrosion of uranium during the operational period;
- Corrosion of Magnox during the operational and the early and medium-term post-closure periods;
- Corrosion of stainless steels in waste streams 2F08 and 2F03/C (AGR assembly components and AGR fuel cladding) in the long term post-closure.

All of the gaseous carbon-14 generation is assumed to be as organic species (nominally labelled as methane) except for the degradation of graphite, which is assumed to also generate gaseous carbon-14 as CO. In practice only a fraction of the carbon-14 released from metals may be released as gas, so the plotted results represent an upper bound on the generation rate (for a given inventory and corrosion rate).

The following sequence of events explains how the carbon-14 containing gas generation rates from steel wastes vary with time:

- There is an initial rapid increase in the carbon-14 generation from uranium corrosion, reaching a constant value after a few years. This is due to a combination of rapid corrosion of emplaced uranium and emplacement of further uranium. At about 2065 AD the rate from uranium corrosion reduces as a result of a change in emplacement rate.
- There is a steady increase in the rate of carbon-14 generation from all other waste types between 2040 and 2105 AD due to waste emplacement. After 2110, but prior to backfilling, steel wastes contribute about 17% of the total rate of carbon-14 gas generation.
- Backfilling starts at 2190 AD. At this time the temperature is assumed to increase from 35°C to 45°C, which increases the corrosion rates of the metals. This results in a step increase in the rate of carbon-14 containing gas generation from corrosion processes, with a small increase for steel wastes.
- Within a few years of closure (at 2200 AD), the vaults become anaerobic as oxygen is consumed by corrosion and organic degradation processes or is flushed out of the near field at full resaturation. Carbon-14 generation rates from waste steels decrease under anaerobic conditions due to reduced corrosion rates.
- Carbon-14 gas generation from the anaerobic corrosion of steel wastes continues in the long term at a relatively low rate; the rate reduces with time due to radioactive decay.
- During the medium-term post-closure period, until about 2700 AD, the corrosion of steel wastes contributes less than 0.5% of the total rate of carbon-14 gas generation. By about 2700 AD, other major sources of carbon-14 gas generation in UILW/ULLW have corroded away leaving steels wastes to be the major sources of carbon-14 gas generation in the long term.
- Through all stages of the evolution of the GDF, waste streams 2F03/C and 2F08 are assessed to be the major sources of carbon-14 from irradiated steels in UILW/ULLW.
Figure 6  Breakdown by waste group of calculated generation rates for carbon-14 containing gases from Legacy UILW / ULLW during GDF operations and early post-closure; the major contributors to the other wastes category are the corrosion of uranium and Magnox
Figure 7  Breakdown by waste group of calculated generation rates for carbon-14 containing gases from Legacy UILW / ULLW in the long term; the major contributors to the other wastes category are the corrosion of uranium (in the short term) and Magnox wastes.
6.1.2 Legacy SILW/SLLW

To gain an understanding of the contribution of steel wastes to the generation of gaseous carbon-14 from the Legacy SILW / SLLW, a breakdown of the carbon-14 containing gas generation rates for mild steel and stainless steel wastes is compared with other sources of carbon-14 in Figure 8 and Figure 9, for shorter and longer time scales respectively.

In summary, for the Legacy SILW / SLLW, the results indicate that the main sources of carbon-14 containing gas generation are [16]:

- Degradation of graphite during the operational and the early and medium-term post-closure periods;
- Corrosion of steels (mainly mild steel) in the long term post-closure.

The generation of gaseous carbon-14 from degradation of graphite is as a combination of organic species (nominally labelled as methane) and carbon monoxide. However, in the modelling it is assumed that gaseous carbon-14 generation from metal corrosion is all as organic species. In practice only a fraction of the carbon-14 released from metals may be released as gas, so the plotted results represent an upper bound on the generation rate (for a given inventory and corrosion rate).

The following sequence of events explains how the carbon-14 containing gas generation rates from steel wastes vary with time:

- There is a steady increase in the rate of carbon-14 gas generation from all processes between 2040 and 2112 AD due to waste emplacement. This is followed, from 2112 to 2190 AD, by a gradual reduction in the generation rate from graphite wastes, while generation rates from steel wastes remain steady. During the emplacement period, steels contribute up to 14% of the total rate of carbon-14 gas generation from SILW/SLLW. This fraction increases to 37% immediately prior to backfilling.
- Backfilling starts at 2190 AD. At this time the temperature is assumed to increase from 35°C to 45°C. This causes an increase in the corrosion rate of the steels, resulting in small step increases in the corresponding carbon-14 containing gas generation rates.
- On full resaturation at 2205 AD, the vaults become anaerobic and there is a decrease in the rate of carbon-14 generation from steel wastes due to reduced corrosion rates.
- Initially after GDF closure the main source of carbon-14 containing gas is degradation of graphite. However, most of the releasable carbon-14 from this source is released within the first 1000 years. Therefore, by around 2800 AD corrosion of mild steel waste becomes the dominant source of carbon-14 containing gas generation from SILW/SLLW. Generation from this source continues in the long term, with the carbon-14 containing gas generation rate reducing steadily due to radioactive decay of the carbon-14.
Figure 8  Breakdown by waste group of calculated generation rates for carbon-14 containing gases from Legacy SILW / SLLW during GDF operations and early post-closure; the major contributor to the other wastes category is the degradation of graphite
Figure 9   Breakdown by waste group of calculated generation rates for carbon-14 containing gases from Legacy SILW / SLLW in the long term; the major contributor to the other wastes category is the degradation of graphite
6.1.3 NNB UILW

Stainless steel wastes are the main contributors to the carbon-14 inventory for NNB UILW and thus their corrosion provides the main source of carbon-14 containing gas during all periods for this category of wastes. A breakdown of the carbon-14 containing gas generation rates for different steel waste stream groups in NNB UILW is shown in Figure 10 and Figure 11, for shorter and longer time scales respectively.

In the modelling it is assumed that gaseous carbon-14 generation from metal corrosion is all as organic species. In practice only a fraction of the carbon-14 released from metals may be released as gas, so the plotted results represent an upper bound on the generation rate (for a given inventory and corrosion rate).

The following sequence of events explains how the main components of the carbon-14 containing gas generation rates vary with time:

- There is a steady increase in the carbon-14 generation from the corrosion of stainless steels between 2100 and 2140 AD due to waste emplacement. The carbon-14 gas generation then remains steady from 2140 to 2190 AD.

- Backfilling starts in 2190 AD and the temperature is assumed to increase from 35°C to 45°C. This increases the corrosion rates of the steels, resulting in a step increase in all contributions to the carbon-14 containing gas generation.

- On full resaturation at 2205 AD the vaults become anaerobic. As a result, the corrosion rates of the steels decrease, and there are corresponding decreases in the contributions to the carbon-14 containing gas generation rates.

- Generation from corrosion of stainless steel continues in the long term, with the carbon-14 containing gas generation rate reducing steadily due to decay of the carbon-14.

- Through all stages of the evolution of the GDF, waste stream AP301 (AP1000 stainless steel reactor internal decommissioning wastes) is assessed to be the major source of carbon-14 from NNB UILW. It should be noted that apart from AP301, EP302 and EP303, the other NNB UILW steel waste streams, AP302 and AP01, contribute less than $10^5$ TBq yr$^{-1}$ to the carbon-14 generation through all stages of GDF evolution, and less than 0.025% of the total carbon-14 release from NNB UILW.
Figure 10  Breakdown by waste group of calculated generation rates for carbon-14 containing gases from NNB UILW during GDF operations and early post-closure
6.1.4 Summary of carbon-14 containing gas generation from steels

A summary of the maximum rates of carbon-14 gas generation from steel wastes during the five stages of GDF evolution based on SMOGG calculations is presented in Table 25. These maximum rates have been derived on the assumption that all carbon-14 released from the steel wastes is releasable as gaseous species. In the table, the maximum rates of carbon-14 containing gas generation from the steel waste groups are compared with the overall maximum rate of carbon-14 gas generation from all waste types in the GDF during each stage.

The total rate of carbon-14 containing gas generation in the GDF is highest during the operational period, when rapid, anaerobic corrosion of uranium wastes occurs. However, the uranium wastes are expected to corrode away by about 2110 before the GDF is closed.
Table 25  Summary of the maximum rates of carbon-14 containing gas generation (TBq yr⁻¹) during each stage of GDF evolution; the results for steel wastes are compared with total rates of carbon-14 containing gas generation from all sources

<table>
<thead>
<tr>
<th>Waste group</th>
<th>Gas</th>
<th>Emplacement (2040 to 2190 AD)</th>
<th>Backfilling (2190 to 2210 AD)</th>
<th>Early post-closure (2190 to 2230 AD)</th>
<th>Medium-term post-closure (2230 to 3000 AD)</th>
<th>Long-term post-closure (from 3000 AD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Legacy UILW</td>
<td>Stainless steel – 2F03/C</td>
<td>¹⁴CH₄ 8.08×10⁻³</td>
<td>1.48×10⁻²</td>
<td>1.48×10⁻²</td>
<td>1.80×10⁻⁴</td>
<td>1.62×10⁻⁴</td>
</tr>
<tr>
<td></td>
<td>Stainless steel – 2F08</td>
<td>¹⁴CH₄ 5.26×10⁻³</td>
<td>9.62×10⁻³</td>
<td>9.62×10⁻³</td>
<td>1.17×10⁻⁴</td>
<td>1.06×10⁻⁴</td>
</tr>
<tr>
<td></td>
<td>Stainless steel – other</td>
<td>¹⁴CH₄ 7.61×10⁻⁴</td>
<td>1.39×10⁻³</td>
<td>1.39×10⁻³</td>
<td>1.69×10⁻⁵</td>
<td>1.53×10⁻⁵</td>
</tr>
<tr>
<td></td>
<td>Mild steel</td>
<td>¹⁴CH₄ 9.25×10⁻⁴</td>
<td>1.16×10⁻³</td>
<td>1.16×10⁻³</td>
<td>3.12×10⁻⁵</td>
<td>2.81×10⁻⁵</td>
</tr>
<tr>
<td>Legacy SLLW</td>
<td>Stainless steel</td>
<td>¹⁴CH₄ 7.03×10⁻⁴</td>
<td>1.28×10⁻³</td>
<td>1.28×10⁻³</td>
<td>1.56×10⁻⁵</td>
<td>1.41×10⁻⁵</td>
</tr>
<tr>
<td></td>
<td>Mild steel</td>
<td>¹⁴CH₄ 3.24×10⁻³</td>
<td>4.07×10⁻³</td>
<td>4.07×10⁻³</td>
<td>1.09×10⁻⁴</td>
<td>9.85×10⁻⁵</td>
</tr>
<tr>
<td></td>
<td>Stainless steel – AP301</td>
<td>¹⁴CH₄ 1.30×10⁻²</td>
<td>2.38×10⁻²</td>
<td>2.38×10⁻²</td>
<td>2.90×10⁻⁴</td>
<td>2.61×10⁻⁴</td>
</tr>
<tr>
<td></td>
<td>Stainless steel – EP302, EP303</td>
<td>¹⁴CH₄ 3.35×10⁻³</td>
<td>6.15×10⁻³</td>
<td>6.15×10⁻³</td>
<td>7.49×10⁻⁵</td>
<td>6.74×10⁻⁵</td>
</tr>
<tr>
<td></td>
<td>Stainless steel – AP01</td>
<td>¹⁴CH₄ 1.45×10⁻⁶</td>
<td>2.66×10⁻⁶</td>
<td>2.66×10⁻⁶</td>
<td>3.24×10⁻⁸</td>
<td>2.91×10⁻⁸</td>
</tr>
<tr>
<td></td>
<td>Mild steel – AP302</td>
<td>¹⁴CH₄ 1.67×10⁻⁶</td>
<td>2.10×10⁻⁶</td>
<td>2.10×10⁻⁶</td>
<td>5.66×10⁻⁸</td>
<td>5.09×10⁻⁸</td>
</tr>
<tr>
<td>Total from steel wastes</td>
<td>¹⁴CH₄ 3.52×10⁻²</td>
<td>6.22×10⁻²</td>
<td>6.22×10⁻²</td>
<td>8.35×10⁻⁴</td>
<td>7.52×10⁻⁴</td>
<td></td>
</tr>
<tr>
<td>Totals by gas species from all wastes(1)</td>
<td>¹⁴CH₄ 4.26×10⁻¹</td>
<td>2.91×10⁻¹</td>
<td>2.91×10⁻¹</td>
<td>1.24×10⁻¹</td>
<td>8.39×10⁻⁴</td>
<td></td>
</tr>
<tr>
<td></td>
<td>¹⁴CO 1.23×10⁻²</td>
<td>5.61×10⁻³</td>
<td>5.61×10⁻³</td>
<td>2.46×10⁻³</td>
<td>3.72×10⁻⁷</td>
<td></td>
</tr>
<tr>
<td>Total from all wastes(1)</td>
<td>4.39×10⁻¹</td>
<td>2.96×10⁻¹</td>
<td>2.96×10⁻¹</td>
<td>1.25×10⁻¹</td>
<td>8.39×10⁻⁴</td>
<td></td>
</tr>
</tbody>
</table>

Notes  
(1)¹⁴CO₂ releases from irradiated graphite are assumed to be consumed by carbonation reactions in the near field after closure and are not included in the post-closure assessment calculations.
During the remainder of the operational period, through backfilling and the medium-term post-closure period up to about 2700 AD, carbon-14 containing gas generation is dominated by corrosion of Magnox wastes. Once the Magnox wastes are fully corroded, carbon-14 containing gas generation continues at a lower rate due predominantly to the corrosion of irradiated steels.

The rates of carbon-14 generation from steel wastes are highest during the backfilling period when the conditions in the GDF are aerobic and the temperature is 45°C. The maximum long-term rates (at 3000 AD) are slightly lower than the medium-term rates (at 2230 AD), because the radioactive decay that takes place over 770 years is taken into account.

Four groups of steel waste streams give a maximum generation rate above $10^{-4}$ TBq yr$^{-1}$ in the medium and long-term post-closure periods. These are:

- 2F03/C (Encapsulated AGR Cladding);
- 2F08 (AGR Stainless Steel Fuel Assembly Components);
- Legacy mild steel shielded ILW; and
- AP301 (NNB AP1000 stainless steel reactor internal decommissioning wastes).

The first two of these are unshielded ILW. The last is a waste stream arising from the decommissioning of reactors that have not yet been built, and there is significant uncertainty over the characteristics of this and other NNB waste streams. Based on current inventory information, NNB steel wastes contribute about 44-48% of the total carbon-14 containing gas generation from irradiated steels through all stages of GDF evolution.

A comparison of the results for steel wastes with the results from Phase 1 is shown in Figure 12. The generation rates during the operating and backfilling periods are similar to, or slightly higher, in Phase 2. There has been an increase in the inventory of carbon 14 associated with steels, and this would be expected to lead to an increase in generation rates. However, this is offset by the more detailed treatment of important waste streams and the geometrical data that describes them. Thereafter the generation rates are lower. This arises because of the lower value for the stainless steel corrosion rate under anaerobic conditions that is used in the Phase 2 work.

**Figure 12** Comparison of the maximum generation rates (TBq yr$^{-1}$) estimated in Phase 1 and Phase 2 of this project for irradiated steel wastes
6.1.5 Bulk Gas Generation

Bulk gas generation is required post-closure to provide a gas phase within which any gaseous carbon-14 generated can migrate. During the early post-closure period, as corrosion of reactive metals occurs, bulk gas generation is expected to be relatively rapid. At longer times radiolysis may provide some of the bulk gas generation, reducing as the radionuclide inventory decays. Otherwise bulk gas generation is provided by corrosion of steel containers and to some extent steel wastes, and this continues at a relatively lower rate for a very long time. Given this, it is likely to be the long-term bulk gas generation rate that is of interest in terms of whether a gas phase is maintained in vaults for each waste group.

The maximum post-closure bulk gas generation rates per vault for the three waste groups are given in Table 26. The maximum rates are reasonably representative of the rates throughout the specified periods with the exception of the long-term rate for the Legacy UILW / ULLW vaults. This continues to reduce from the value given (applicable at 3000 AD) to 31.3 m$^3$ at STP yr$^{-1}$ by 4000 AD and to 12.0 m$^3$ at STP yr$^{-1}$ by 50,000 AD.

<table>
<thead>
<tr>
<th>Waste group</th>
<th>Maximum bulk gas generation rate per vault (m$^3$ at STP yr$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Early post-closure (2190 to 2230 AD)</td>
</tr>
<tr>
<td>Legacy UILW / ULLW</td>
<td>5085</td>
</tr>
<tr>
<td>Legacy SILW / SLLW</td>
<td>193</td>
</tr>
<tr>
<td>NNB UILW</td>
<td>14.7</td>
</tr>
</tbody>
</table>

The corrosion of irradiated steel wastes is a minor source of bulk gas, providing a constant source of hydrogen at a rate of 0.42 m$^3$ at STP yr$^{-1}$ per vault from legacy UILW / ULLW, 1.00 m$^3$ at STP yr$^{-1}$ from legacy SILW / SLLW (primarily from mild steels) and 0.092 m$^3$ at STP yr$^{-1}$ from NNB UILW throughout the post-closure period.

The rate of generation of bulk gas required for a gas phase to exist is discussed in the Carbon-14 project Gas Formation and Migration Report [13]. Whether or not a free gas phase will form is a site-specific question; it depends on the capacity of the groundwater flowing through the GDF to dissolve the gas generated within the EBS. The capacity of the groundwater for gas is dependent on the hydrogeological properties of the site and the host rock (which control the groundwater flux through the EBS) and the depth of the GDF (which controls the in situ pressure and gas solubility). In general, a higher strength host rock is expected to be the most permeable of the three illustrative host rock types considered in this work, and will give the highest rates of groundwater flow through the EBS into which gas may dissolve and be transported away from the GDF.

The Gas Formation and Migration Report concludes that a gas phase will exist for a vault in most higher strength host rocks at a depth of 650 m if the bulk gas generation rate in the vault is ~10 m$^3$ at STP yr$^{-1}$ or greater. Depending on the host rock properties, a generation rate one or two orders of magnitude lower may well be sufficient. This means that for both the Legacy UILW / ULLW vaults and the Legacy SILW / SLLW vaults a gas phase is likely to exist at all times of relevance to the release of gaseous carbon-14. However, for the SILW / SLLW vaults, a moderate reduction to the bulk gas generation rate from 30 years post-closure could prevent formation of a gas phase from this time for some higher strength host rocks. This would also be the case at longer times for the UILW / ULLW vaults.
For the NNB UILW vaults, there is greater uncertainty concerning the existence of a bulk gas phase for some higher strength host rocks due to the lower rates of bulk gas generation both immediately after closure and in the longer term. The main source of bulk gas during the first 700 years post-closure is radiolysis, with corrosion of stainless steel containers becoming the main source thereafter. Although the current assessment suggests that a gas phase would exist at least during the first few hundred years after closure, this analysis is based on estimated radionuclide inventories for wastes from reactors which have yet to be built, and which are likely to be conservative. Whether a bulk gas phase will exist in the NNB ULW vaults in the longer term in a higher strength host rock will depend on the properties of the host rock.

In the case of LSSR or evaporite host rock types, where there is either limited or no water flow through the EBS, respectively, it is expected that a free gas phase will form in all three types of waste vault and will persist in the long-term throughout the period of carbon-14 generation from steel wastes considered in the current assessment. However, it may not migrate through the host rock to the biosphere.

### 6.2 Understanding the potential consequences of carbon-14 containing gas generation

In this section, the potential consequences of the release of the carbon-14 gas generated from the steel wastes, described in the previous sub-section, are assessed using the methods and the release and migration scenarios outlined in Section 5. For the post-closure cases, the assumption is made that a bulk gas phase, which can transport carbon-14 containing gases, is formed and persists throughout the assessment period.

#### 6.2.1 Operational period

The updated OESA methodology outlined in sub-section 5.3.1 was applied to the emplacement and backfilling stages of the GDF.

The pathway considered for release of carbon-14 from the GDF during the operational phase is via aerial discharge from a 15m high stack. The potential impact of the aerial discharges is assessed by calculating the effective dose rates to a local resident family receptor group. The effective doses can be calculated by combining the generation rates given in Table 25 with the recommended effective dose factors given in Table 21. The resulting effective doses for the emplacement and backfilling stages are shown in Table 27 and Table 28 respectively. These two tables have to be read with care. There is uncertainty concerning both the fraction of carbon-14 that will be released as gas from irradiated steels and the chemical form in which it will be released. Therefore, effective doses have been calculated for three cases in which all of the carbon-14 is assumed to be released as one gaseous species: carbon monoxide or methane. Clearly they cannot all be 100% and the sum cannot exceed 100%. The values for carbon dioxide are not included in the Table, as it would be expected to react with the cementitious grout in the packages.

<table>
<thead>
<tr>
<th>Age Group</th>
<th>Carbon monoxide</th>
<th>Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Infant</td>
<td>$4.2 \times 10^{-3}$</td>
<td>$5.3 \times 10^{-7}$</td>
</tr>
<tr>
<td>Child</td>
<td>$2.5 \times 10^{-3}$</td>
<td>$3.1 \times 10^{-7}$</td>
</tr>
<tr>
<td>Adult</td>
<td>$2.5 \times 10^{-3}$</td>
<td>$3.2 \times 10^{-7}$</td>
</tr>
</tbody>
</table>
6.2.2 Post-closure period

The medium-term and long-term post-closure risks arising from the corrosion of irradiated steels are shown in Table 29 and Table 30, respectively. These are obtained by combining:

- The maximum carbon-14 containing gas generation rates in each stage of the assessment given in Table 25;
- The migration cases described in Table 20;
- The biosphere factor given in Table 22;
- The bases for the proportion of carbon-14 released as methane discussed in sub-section 3.5.4; and
- The dose to risk conversion factor of 0.06 Sv⁻¹ [97].

It will be noted from the tables that the maximum risks during the long-term post-closure period (Table 30) are very similar to those in the medium-term period (Table 29). This is the result of radioactive decay; the risks continue to decrease progressively with time.

The Environment Agencies’ risk guidance level is 10⁻⁶ [97], and risks above this guidance level are shown in Bold in the two tables. Only one of the cases in each table is above the risk guidance level. This is for the variant case of 100% of the carbon-14 being released as methane, with a focused release to an area of 10⁴ m² (Case C). Even with a focused release, the risks for the Reference Case, where 10% of the carbon-14 is released as methane, are below the risk guidance level in the medium and long term periods.

The carbon-14 generation rates are highest in the early post-closure period (Table 25). These rates occur in the short period (5 years after closure) while the conditions in the GDF remain aerobic and the temperature is elevated. However, this release is over a very short period and, in certain geological environments, the release to the biosphere may be spread in time. This would reduce the flux of carbon-14 into the soil zone to no more than that in the medium-term post-closure period. It should be noted that the generation rates from steel wastes are lower than those from Magnox wastes in the same period (see Figure 6).
Table 29  Medium-term post-closure risks for the different steels release cases – combining the migration cases (see Table 20) with the cases for the proportion of carbon-14 released as methane

<table>
<thead>
<tr>
<th>ID</th>
<th>Delay Time ($^{14}$C half-lives)</th>
<th>Release Area ($m^2$)</th>
<th>100% Methane</th>
<th>10% Methane $^{(1)}$</th>
<th>1% Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0</td>
<td>$10^6$</td>
<td>$3.6 \times 10^9$</td>
<td>$3.6 \times 10^9$</td>
<td>$3.6 \times 10^{10}$</td>
</tr>
<tr>
<td>B</td>
<td>No release</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>0</td>
<td>$10^4$</td>
<td>$3.6 \times 10^9$</td>
<td>$3.6 \times 10^7$</td>
<td>$3.6 \times 10^8$</td>
</tr>
<tr>
<td>D</td>
<td>0</td>
<td>$10^7$</td>
<td>$3.6 \times 10^9$</td>
<td>$3.6 \times 10^{10}$</td>
<td>$3.6 \times 10^{11}$</td>
</tr>
<tr>
<td>E</td>
<td>1</td>
<td>$10^6$</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>F</td>
<td>$^{2}$</td>
<td>$10^7$</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Notes:  
$^{(1)}$ The Reference Case

(2) For cases E and F, due to the delay time in the geosphere, carbon-14 released from the GDF does not reach the biosphere during the medium-term post-closure period.

Table 30  Long-term post-closure risks for the different steels release cases – combining the migration cases (see Table 20) with the cases for the proportion of carbon-14 released as methane

<table>
<thead>
<tr>
<th>ID</th>
<th>Delay Time ($^{14}$C half-lives)</th>
<th>Release Area ($m^2$)</th>
<th>100% Methane</th>
<th>10% Methane $^{(1)}$</th>
<th>1% Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0</td>
<td>$10^6$</td>
<td>$3.2 \times 10^9$</td>
<td>$3.2 \times 10^9$</td>
<td>$3.2 \times 10^{10}$</td>
</tr>
<tr>
<td>B</td>
<td>No release</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>0</td>
<td>$10^4$</td>
<td>$3.2 \times 10^9$</td>
<td>$3.2 \times 10^7$</td>
<td>$3.2 \times 10^8$</td>
</tr>
<tr>
<td>D</td>
<td>0</td>
<td>$10^7$</td>
<td>$3.2 \times 10^9$</td>
<td>$3.2 \times 10^{10}$</td>
<td>$3.2 \times 10^{11}$</td>
</tr>
<tr>
<td>E</td>
<td>1</td>
<td>$10^6$</td>
<td>$1.8 \times 10^9$</td>
<td>$1.8 \times 10^9$</td>
<td>$1.8 \times 10^{10}$</td>
</tr>
<tr>
<td>F</td>
<td>$^{2}$</td>
<td>$10^7$</td>
<td>$1.8 \times 10^9$</td>
<td>$1.8 \times 10^9$</td>
<td>$1.8 \times 10^{10}$</td>
</tr>
</tbody>
</table>

Notes:  
$^{(1)}$ The Reference Case.
7 Irradiated Stainless Steel Variant Case

In addition to the Reference Case calculations described in Section 6, a number of alternative scenarios are considered in the carbon-14 assessment calculations performed by the project. These additional calculations are used to investigate some of the major sources of uncertainty in the GDF design, operations and waste behaviour that may affect the generation of carbon-14 containing gases. The results for all alternative scenarios are outlined in the Modelling Report [16]. This section describes the results of one set of variant calculations for an alternative scenario considered for irradiated steel wastes.

7.1 Scenario identification

A source of uncertainty for the irradiated steel wastes concerns the potential impact of irradiation on the corrosion rate of stainless steel wastes and thus on the rate of carbon-14 containing gas generation. As noted in sub-section 3.2.5.1, under certain irradiation conditions, radiation-induced sensitisation of stainless steel reactor components may occur; this may lead to increased intergranular corrosion and IGSCC of some irradiated stainless steel wastes. Although the direct impact of intergranular corrosion on rates of carbon-14 release is considered to be small, the indirect effects are considered to be potentially more significant. In particular, intergranular attack results in separation of grain boundaries and cracking of the steel matrix, which may lead to a progressive increase in the exposed surface area over which general corrosion of the steel surface may operate. However, there is no information available on the rates of corrosion of irradiated or sensitised steel wastes and how these compare with those for unirradiated materials. In addition, there is limited information concerning the extent to which different stainless steel reactor wastes may be sensitised, which will depend on both the thermal and irradiation history of the steel components during service lifetimes. Furthermore, the extent of intergranular corrosion and cracking of sensitised materials prior to packaging will depend on their storage history. There is also uncertainty concerning whether intergranular attack and IGSCC of sensitised materials will continue under anaerobic alkaline conditions in the longer term.

Given these uncertainties, an alternative scenario was considered for irradiated stainless steel wastes to scope the potential impact of higher exposed surface area (leading to higher net rates of mass loss) than for bulk unirradiated materials on the rates of carbon-14 release:

- Case IS – effective rates of irradiated stainless steel corrosion under all conditions are increased by a factor of 10 compared to values for unirradiated stainless steels (as used in the Reference Case calculations).

This is the scaling factor approach outlined in Section 3.5.3.

In this scenario, it is assumed that the corrosion rates of unirradiated steel wastes and irradiated mild steel wastes are unaffected. The increased effective corrosion rates are applied to all irradiated stainless steels from both legacy and NNB waste streams.

7.2 Description of calculations

Variant calculations have been set up to demonstrate the effects of applying these increased effective rates of irradiated stainless steel corrosion to the Reference Case models. The only change in the assessment basis for the Irradiated Steels Variant Case compared to the Reference Case is a change in the corrosion rate parameters for the irradiated stainless steel wastes. The revised corrosion rate parameters for irradiated stainless steel used for the variant case are given in Table 31; the corrosion rate parameters for mild steel wastes and unirradiated stainless steels (i.e. containers) are unchanged as per Table 19.

In common with the gas generation calculations presented for the Reference Case in Section 6.1, the calculations presented here give the maximum potential generation rates of gaseous carbon-14 from steel wastes (for a given inventory and corrosion rates), and, therefore, are cautious.
practice, only a fraction of the carbon-14 released from metals may be released as gas and there is uncertainty concerning the gaseous speciation.

Table 31  Effective corrosion rates of irradiated stainless steels used in the assessment of gas generation and carbon-14 release in the Irradiated Steel Variant Case

<table>
<thead>
<tr>
<th>Metal</th>
<th>Conditions</th>
<th>Time scale</th>
<th>Corrosion rate (m yr⁻¹)</th>
<th>Characteristic time (yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>35°C</td>
<td>45°C</td>
</tr>
<tr>
<td>Stainless Steel</td>
<td>Atmospheric</td>
<td>Chronic</td>
<td>3.00×10⁻⁷</td>
<td>3.00×10⁻⁷</td>
</tr>
<tr>
<td></td>
<td>Aerobic, high pH</td>
<td>Chronic</td>
<td>5.43×10⁻⁷</td>
<td>1.00×10⁻⁶</td>
</tr>
<tr>
<td></td>
<td>Anaerobic, high pH</td>
<td>Chronic</td>
<td>1.23×10⁻⁸</td>
<td>2.14×10⁻⁸</td>
</tr>
</tbody>
</table>

7.3  C-14 gas generation calculations

7.3.1  Gas generation results for Legacy UILW / ULLW

For the legacy UILW / ULLW wastes, the majority of the carbon-14 containing gas generation arising from waste steels arises from the stainless steel waste streams 2F03/C and 2F08, which, in the Reference Case, are the major contributing streams to carbon-14 generation during the post-closure period beyond 2700 AD (see Figure 7). Thus, one impact of the increased rates of irradiated steel corrosion in Case IS, compared to the Reference Case, is to increase the rate of carbon-14 containing gas generation in the long term by a factor of nearly 10. In addition, the stainless steel waste streams become a major source of carbon-14 generation during the operational period once the uranium wastes have corroded away (2110 to 2190 AD), contributing about 66% of the carbon-14 release, and increasing the release rate by about a factor of 2.5 compared to the Reference Case during this period. This increase in the rate of carbon-14 generation continues during the first ten years of the backfilling period (2190 to 2200 AD), when the temperature is elevated but before conditions become anaerobic and the stainless steel corrosion rate drops significantly. These increased rates of carbon-14 generation are illustrated in Figure 13, which shows a breakdown of the carbon-14 containing gas generation rates for the waste steels from emplacement through to the start of the long-term post-closure period (3000 AD) for the Irradiated Steel Variant Case.
7.3.2 Gas generation results for Legacy SILW / SLLW

For the Legacy SILW / SLLW, the majority of the carbon-14 containing gas generation arising from waste steels arises from mild steels, which are the major contributor to carbon-14 generation during the post-closure period beyond about 2550 AD. Thus, the increased rates of irradiated stainless steel corrosion considered in Case IS has only a relatively minor effect on the overall rates of carbon-14 generation as gas during both the operational and post-closure periods, when compared to the Reference Case. Stainless steel wastes become the largest source of carbon-14 from these legacy wastes briefly during the backfilling period (2190 to 2200 AD), when the temperature is elevated but before conditions become anaerobic. The main effect post-closure is an increase in the rate of carbon-14 containing gas generation in the long term by a factor of about 2. This is illustrated in Figure 14, which shows a breakdown of the carbon-14 containing gas generation rates for the waste steels from emplacement through to the start of the long-term post-closure period (3000 AD).
7.3.3 Gas generation results for NNB UILW

Stainless steel wastes are the main contributors to the carbon-14 inventory for NNB UILW and their corrosion provides the major source (>99.9%) of carbon-14 containing gas during all periods for this category of wastes. The effect of the increased effective corrosion rates for irradiated stainless steels for Case IS is to increase the rate of carbon-14 containing gas generation by a factor of 10 throughout all stages of GDF evolution.

7.3.4 Summary of carbon-14 containing gas generation from steels

The rates of carbon-14 containing gas generation from a GDF from all sources, and from irradiated steel wastes only, for the irradiated Steel Variant Case, IS, are compared with the Reference Case results in Figure 15. The rates of carbon-14 gas generation are compared from emplacement through to the start of the long-term post-closure period (3000 AD).
Figure 15  Comparison of rates of carbon-14 gas generation from a GDF from all sources and from irradiated steel wastes only for the Irradiated Steel (IS) Variant Case with the Reference Case; rates are compared from emplacement through to the start of the long-term post-closure period (3000 AD)

It will be apparent from the figure that the ten-fold increase in the effective corrosion rates of irradiated stainless steels has three main impacts on the total rate of carbon-14 containing gas generation from a GDF. During the operational period between 2110 and 2190 AD, once the uranium wastes have corroded away, there is a factor of about 3 increase in the total carbon-14 generation compared to the Reference Case. The steel wastes contribute up to 79% of the total carbon-14 generation during this period. However, the total carbon-14 generation remains comparable to that from the uranium wastes prior to 2110 AD. During the first 10 years of the backfilling period (from 2190 to 2200 AD), the total rate of carbon-14 generation from the GDF reaches a peak value of ~0.8 TBq yr\(^{-1}\) (nearly a factor of 3 higher than in the Reference Case) due to the higher temperature of the EBS, before conditions become anaerobic reducing the corrosion rate of stainless steels significantly. Then in the long-term post closure period, once the more reactive Magnox wastes have corroded away (by ~2700 AD), carbon-14 generation rates are dominated by the corrosion of stainless steel wastes, with the overall rates increasing by a factor of nearly 8 compared to the Reference Case.
A summary of the maximum rates of carbon-14 gas generation from steel wastes during the five stages of GDF evolution for the Irradiated Steels Variant Case is presented in Table 32, (cf. Table 25). In each table, the maximum rates of carbon-14 containing gas generation from the steel waste groups are compared with the overall maximum rate of carbon-14 gas generation from all waste types in the GDF during each stage.

The scenario for Case IS includes the conservative assumption that all irradiated stainless steel wastes will be sensitised and susceptible to intergranular corrosion. However, there is significant uncertainty concerning the extent and degree of sensitisation for reactor stainless steels, in particular from new build reactors that have yet to be built. Not all reactor stainless steels will be sensitised. It is known, for example, that the fuel stringer components and cladding in AGRs vary in their susceptibility to sensitisation depending on their location in the reactor (see sub-section 3.2.5.1). Therefore, the higher carbon-14 generation rates from irradiated stainless steels calculated for Case IS are considered to be cautiously high.

It is also noted that the carbon-14 gas generation rates shown in Figure 15 and Table 32 are based on the conservative assumption that the entire carbon-14 content of the irradiated steels is releasable as gaseous species. There remains considerable uncertainty concerning the fraction of carbon-14 that will be released as gas from irradiated steel wastes.

### 7.3.5 Bulk gas generation

It was noted in sub-section 6.1.5, that the corrosion of irradiated steel wastes is a minor source of bulk gas in the post-closure period in comparison to the corrosion of steel waste containers. The use of a higher effective corrosion rate for the irradiated stainless steel wastes in the variant case leads to only small increases in the overall estimates of bulk gas generation rates from the waste vaults in the post-closure period. These amount to up to 3.5% for Legacy UILW / ULLW and up to 12% for Legacy SILW / SLLW. In the case of NNB UILW vaults, where the rates of bulk gas generation are lower (c.f. Table 26), the overall estimate of bulk gas generation increases from 1.32 to 1.92 m³ at STP yr⁻¹ in the long-term due to the higher effective corrosion rates.
Table 32  Summary of the maximum rates of carbon-14 containing gas generation (TBq yr\(^{-1}\)) during each stage of GDF evolution for the Irradiated Steel Variant Case; the results for steel wastes are compared with total rates of carbon-14 containing gas generation from all sources

<table>
<thead>
<tr>
<th>Waste group</th>
<th>Gas</th>
<th>Emplacement</th>
<th>Backfilling</th>
<th>Early post-closure</th>
<th>Medium-term post-closure</th>
<th>Long-term post-closure</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>(2040 to 2190 AD)</td>
<td>(2190 to 2210 AD)</td>
<td>(2190 to 2230 AD)</td>
<td>(2230 to 3000 AD)</td>
<td>(from 3000 AD)</td>
</tr>
<tr>
<td><strong>Legacy UILW</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stainless steel – 2F03/C</td>
<td>(^{14})CH(_{4})</td>
<td>8.08×10(^{2})</td>
<td>1.48×10(^{1})</td>
<td>1.48×10(^{1})</td>
<td>1.80×10(^{-3})</td>
<td>1.62×10(^{3})</td>
</tr>
<tr>
<td>Stainless steel – 2F08</td>
<td>(^{14})CH(_{4})</td>
<td>5.26×10(^{2})</td>
<td>9.62×10(^{2})</td>
<td>9.62×10(^{2})</td>
<td>1.17×10(^{-3})</td>
<td>1.06×10(^{3})</td>
</tr>
<tr>
<td>Stainless steel – other</td>
<td>(^{14})CH(_{4})</td>
<td>7.61×10(^{2})</td>
<td>1.39×10(^{2})</td>
<td>1.39×10(^{-2})</td>
<td>1.69×10(^{-4})</td>
<td>1.53×10(^{-4})</td>
</tr>
<tr>
<td>Mild steel</td>
<td>(^{14})CH(_{4})</td>
<td>9.25×10(^{4})</td>
<td>1.16×10(^{0})</td>
<td>1.16×10(^{-3})</td>
<td>3.12×10(^{-5})</td>
<td>2.81×10(^{-5})</td>
</tr>
<tr>
<td><strong>Legacy SILW</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stainless steel</td>
<td>(^{14})CH(_{4})</td>
<td>7.02×10(^{3})</td>
<td>1.28×10(^{2})</td>
<td>1.28×10(^{-2})</td>
<td>1.56×10(^{-4})</td>
<td>1.41×10(^{-4})</td>
</tr>
<tr>
<td>Mild steel</td>
<td>(^{14})CH(_{4})</td>
<td>3.24×10(^{3})</td>
<td>4.07×10(^{3})</td>
<td>4.07×10(^{-3})</td>
<td>1.09×10(^{-4})</td>
<td>9.85×10(^{-5})</td>
</tr>
<tr>
<td><strong>NNB UILW</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stainless steel – AP301</td>
<td>(^{14})CH(_{4})</td>
<td>1.30×10(^{1})</td>
<td>2.38×10(^{1})</td>
<td>2.38×10(^{1})</td>
<td>2.90×10(^{-3})</td>
<td>2.61×10(^{3})</td>
</tr>
<tr>
<td>Stainless steel – EP302, EP303</td>
<td>(^{14})CH(_{4})</td>
<td>3.35×10(^{2})</td>
<td>6.15×10(^{2})</td>
<td>6.15×10(^{2})</td>
<td>7.49×10(^{-4})</td>
<td>6.74×10(^{-4})</td>
</tr>
<tr>
<td>Stainless steel – AP01</td>
<td>(^{14})CH(_{4})</td>
<td>1.45×10(^{5})</td>
<td>2.66×10(^{5})</td>
<td>2.66×10(^{5})</td>
<td>3.24×10(^{-7})</td>
<td>2.91×10(^{-7})</td>
</tr>
<tr>
<td>Mild steel – AP301</td>
<td>(^{14})CH(_{4})</td>
<td>1.67×10(^{6})</td>
<td>2.10×10(^{6})</td>
<td>2.10×10(^{6})</td>
<td>5.66×10(^{-8})</td>
<td>5.09×10(^{-8})</td>
</tr>
<tr>
<td><strong>Total from steel wastes</strong></td>
<td>(^{14})CH(_{4})</td>
<td>3.15×10(^{1})</td>
<td>5.75×10(^{1})</td>
<td>5.75×10(^{1})</td>
<td>7.09×10(^{-3})</td>
<td>6.38×10(^{-3})</td>
</tr>
<tr>
<td><strong>Totals by gas species from all wastes(^{(1)})</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{14})CH(_{4})</td>
<td>5.08×10(^{1})</td>
<td>8.04×10(^{1})</td>
<td>8.04×10(^{1})</td>
<td>1.30×10(^{-1})</td>
<td>6.47×10(^{-3})</td>
<td></td>
</tr>
<tr>
<td>(^{14})CO</td>
<td>1.23×10(^{2})</td>
<td>5.61×10(^{3})</td>
<td>5.61×10(^{3})</td>
<td>2.46×10(^{-3})</td>
<td>3.72×10(^{-7})</td>
<td></td>
</tr>
<tr>
<td><strong>Total from all wastes(^{(1)})</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5.20×10(^{1})</td>
<td>8.09×10(^{1})</td>
<td>8.09×10(^{1})</td>
<td>1.32×10(^{-1})</td>
<td>6.47×10(^{-3})</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Notes  
\(^{(1)}\) \(^{14}\)CO\(_{2}\) releases from irradiated graphite are assumed to be consumed by carbonation reactions in the near field after closure and are not included in the post-closure assessment calculations.
7.4 Understanding the potential consequences

In this section, the potential consequences of the release of carbon-14 gas generated from the irradiated stainless steel wastes in the variant scenario described in the previous sub-section are assessed using the methods, release and migration scenarios and underlying assumptions used for the Reference Case in Section 6.2.

7.4.1 Operational period

The effective doses to a local resident family group arising from waste steels calculated by combining the gas generation rates for the Irradiated Steels Variant Case with the recommended dose factors (Table 21) are presented in Table 33 and Table 34 for the emplacement and backfilling stages, respectively. These data should be compared with those presented in Table 27 and Table 28.

The effects of increasing the effective corrosion rates of irradiated stainless steels are to increase the effective doses arising from the steel wastes by factors of about nine during both the emplacement and backfilling periods. The highest contribution to effective dose rate to members of the public from carbon-14 containing gas generation for the operational period is calculated to be that for 100% carbon monoxide, namely \(6.9 \times 10^{-2}\) mSv yr\(^{-1}\), during the backfilling period (Table 34). This remains slightly below the maximum effective dose rate constraint to members of the public from a new facility of 0.15 mSv yr\(^{-1}\).

### Table 33  Calculated Effective Doses from carbon-14 (mSv yr\(^{-1}\)) from irradiated steel wastes via aerial discharge during the operational period for the Irradiated Steel Variant Case, assuming 100% generation of each of either carbon monoxide or methane; the doses are based on the maximum carbon-14 containing gas generation rates during the operational period

<table>
<thead>
<tr>
<th>Age Group</th>
<th>Carbon monoxide</th>
<th>Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Infant</td>
<td>(3.8 \times 10^{-2})</td>
<td>(4.7 \times 10^{-6})</td>
</tr>
<tr>
<td>Child</td>
<td>(2.2 \times 10^{-2})</td>
<td>(2.7 \times 10^{-7})</td>
</tr>
<tr>
<td>Adult</td>
<td>(2.3 \times 10^{-2})</td>
<td>(2.8 \times 10^{-7})</td>
</tr>
</tbody>
</table>

### Table 34  Calculated Effective Doses from carbon-14 (mSv yr\(^{-1}\)) from irradiated steel wastes via aerial discharges during backfilling for the Irradiated Steel Variant Case, assuming 100% generation of each of either carbon monoxide or methane; the doses are based on the maximum carbon-14 containing gas generation rates during the temperature peak for irradiated steel wastes

<table>
<thead>
<tr>
<th>Age Group</th>
<th>Carbon monoxide</th>
<th>Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Infant</td>
<td>(6.9 \times 10^{-2})</td>
<td>(8.6 \times 10^{-6})</td>
</tr>
<tr>
<td>Child</td>
<td>(4.1 \times 10^{-2})</td>
<td>(5.0 \times 10^{-6})</td>
</tr>
<tr>
<td>Adult</td>
<td>(4.1 \times 10^{-2})</td>
<td>(5.2 \times 10^{-6})</td>
</tr>
</tbody>
</table>

7.4.2 Post-closure period

The medium- and long-term post-closure risks arising from the corrosion of irradiated steels for the steel variant cases are shown in Table 35 and Table 36, respectively. The risks presented in these tables should be compared with those for the Reference Case in Table 29 and Table 30. In each of these tables, the risks above the Environment Agencies’ risk guidance level of \(10^{-6}\) [97] are shown in Bold.
The effects of increasing the effective corrosion rates of irradiated stainless steels by a factor of 10 is to increase the calculated post-closure radiological risks due to carbon-14 arising from the steel wastes by nearly an order of magnitude in comparison to the Reference Case, respectively for each migration and carbon-14 speciation scenario. Only two of the eighteen cases give calculated risks that are above the risk guidance level. These are for the migration cases with focused release to an area of $10^4\text{m}^2$ (Case C) with either 100% or 10% of the carbon-14 being released as methane. For all other migration and speciation scenarios, the medium- and long-term post-closure risks associated with irradiated steel wastes are below the risk guidance level.

The carbon-14 generation rates are highest in the early post-closure period (Table 32). As noted in sub-section 6.2.2, these rates occur in the short period (5 years after closure) while the conditions in the GDF remain aerobic and the temperature is elevated. However, these releases are not expected to give rise to higher doses to a resident family group due to spreading of the releases in time to the soil zone in some geological environments.

Table 35  Medium-term post-closure risks from irradiated steel wastes for the different release cases – combining the Irradiated Steels Variant Case with the migration cases (see Table 20) and with the cases for the proportion of carbon-14 released as methane

<table>
<thead>
<tr>
<th>ID</th>
<th>Delay Time ($^{14}$C half-lives)</th>
<th>Release Area ($\text{m}^2$)</th>
<th>100% Methane</th>
<th>10% Methane ($^{(1)}$)</th>
<th>1% Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0</td>
<td>$10^6$</td>
<td>$3.0 \times 10^{-7}$</td>
<td>$3.0 \times 10^{-8}$</td>
<td>$3.0 \times 10^{-9}$</td>
</tr>
<tr>
<td>B</td>
<td>No release</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>0</td>
<td>$10^4$</td>
<td>$3.0 \times 10^{-6}$</td>
<td>$3.0 \times 10^{-6}$</td>
<td>$3.0 \times 10^{-7}$</td>
</tr>
<tr>
<td>D</td>
<td>0</td>
<td>$10^7$</td>
<td>$3.0 \times 10^{-8}$</td>
<td>$3.0 \times 10^{-9}$</td>
<td>$3.0 \times 10^{-10}$</td>
</tr>
<tr>
<td>E</td>
<td>$^{(2)}$</td>
<td>$10^6$</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>F</td>
<td>$^{(2)}$</td>
<td>$10^7$</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Notes:  
(1) The Reference Case for speciation.  
(2) For cases E and F, due to the delay time in the geosphere, carbon-14 released from the GDF does not reach the biosphere during the medium-term post-closure period.

Table 36  Long-term post-closure risks from irradiated steel wastes for the different release cases – combining the Irradiated Steels Variant Case with the migration cases (see Table 20) and the cases for the proportion of carbon-14 released as methane

<table>
<thead>
<tr>
<th>ID</th>
<th>Delay Time ($^{14}$C half-lives)</th>
<th>Release Area ($\text{m}^2$)</th>
<th>100% Methane</th>
<th>10% Methane ($^{(1)}$)</th>
<th>1% Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0</td>
<td>$10^6$</td>
<td>$2.7 \times 10^{-7}$</td>
<td>$2.7 \times 10^{-8}$</td>
<td>$2.7 \times 10^{-9}$</td>
</tr>
<tr>
<td>B</td>
<td>No release</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>0</td>
<td>$10^4$</td>
<td>$2.7 \times 10^{-6}$</td>
<td>$2.7 \times 10^{-6}$</td>
<td>$2.7 \times 10^{-7}$</td>
</tr>
<tr>
<td>D</td>
<td>0</td>
<td>$10^7$</td>
<td>$2.7 \times 10^{-8}$</td>
<td>$2.7 \times 10^{-9}$</td>
<td>$2.7 \times 10^{-10}$</td>
</tr>
<tr>
<td>E</td>
<td>1</td>
<td>$10^6$</td>
<td>$1.5 \times 10^{-7}$</td>
<td>$1.5 \times 10^{-8}$</td>
<td>$1.5 \times 10^{-9}$</td>
</tr>
<tr>
<td>F</td>
<td>1</td>
<td>$10^7$</td>
<td>$1.5 \times 10^{-8}$</td>
<td>$1.5 \times 10^{-9}$</td>
<td>$1.5 \times 10^{-10}$</td>
</tr>
</tbody>
</table>

Notes:  
(1) The Reference Case for speciation.
8 Alternative Management Approaches

In the Phase 1 report [1], consideration was given to whether alternative treatment, packaging, design or disposal options would be practicable for the management of irradiated steel wastes. The options considered were:

- Segregation of the steel wastes from other wastes;
- Spread the steel wastes over many vaults;
- Surface treatments of the wastes;
- Melting or thermal treatment; or
- Disposal of the wastes in a long-lived container.

Some of these options may be practicable for certain wastes; some of these options may be beneficial for certain wastes.

Considering the steel waste groups that lead to the highest generation rates of carbon-14 in Table 25, some of them have very large masses and volumes (e.g. SILW Mild Steel), and alternative approaches may well not be appropriate for such large volume waste streams. Some have already been packaged (e.g. 2F03/C), and so an alternative approach may be less appropriate. However, other waste groups leading to the highest generation rates (e.g. 2F08 and AP301) have lower masses and volumes, and so an alternative approach may be practicable if required.

However, on the basis of the results presented in Section 6, and in comparison to other sources of carbon-14, irradiated steels are not the highest priority for such consideration on the basis of the current understanding. It is noted also that it may be possible to reduce the contribution from reactor components that are not yet built (NNB steel wastes) by using steels with lower nitrogen contents than assumed here.
9 Summary and Conclusions

There are about 7,060 TBq of carbon-14 associated with irradiated steel wastes. The overall inventory has increased in the 2013 Derived Inventory as a result of including waste associated with new build reactors, but the inventory of certain key legacy waste streams has decreased as a result of improved understanding of the nitrogen impurity concentrations in these steels. The inventory of carbon-14 in legacy steel wastes has been reduced by over 50%.

Post-closure release of carbon-14 depends particularly on the anaerobic corrosion rate of stainless steel. As part of the international CAST project, a detailed review of corrosion rates has been undertaken and, as a consequence, the long-term corrosion rate of irradiated stainless steels has been reduced by over an order of magnitude.

The overall gas generation rate is also dependent on the thickness of the steel plates and the diameter of more bulky items. The understanding and data for waste dimensions have been improved for some of the important waste streams.

Not all of the carbon-14 released may be released to the gas phase; some of it may dissolve in the aqueous phase or remain in solid phases. Experimental measurements are starting under the CAST project to address this remaining uncertainty concerning the speciation of the carbon-14 released.

The potential consequences of carbon-14 releases from irradiated steel wastes through the various stages of a GDF have been assessed for as follows:

- In the operational period (when all of the carbon-14 is assumed to be released as either 100% methane of 100% carbon monoxide) effective doses from gases released from irradiated steel wastes are assessed to be below the source-related dose constraint for members of the public for a new facility of 0.15 mSv yr\(^{-1}\).

- In the post-closure period, the radiological impact of any release of carbon-14 as gas from irradiated steels will be site specific. Steel wastes are the major contributor to risks via the gas pathway at times greater than five hundred years post-closure. For the post-closure Reference Case (assuming that 10% of the carbon-14 is released as methane), the calculated risks arising from steel wastes are highest (~3.6x10\(^{-5}\)), but remain below the risk guidance level (10\(^{-5}\)) where all of the gas-phase release is focused to a small area (10\(^4\) m\(^2\)) at the surface. Even for a focused surface release, the risks arising from steel wastes will only be above the risk guidance level if a significant proportion (>30%) of the carbon-14 is released as gas.

- In the early post-closure period, the rates of carbon-14 generation from irradiated steels are higher for a short period of about 5 years. However, these releases are not expected to give rise to higher doses to a resident family group due to spreading of the releases in time to the soil zone in some geological environments. It should be noted that the generation rates from steel wastes are lower than those from Magnox wastes in the same period.

For the variant case, with effective corrosion rates increased by a factor of ten to account for potential IGSCC of stainless steel wastes:

- The effective doses from carbon-14 gases released from steels during the operational period are increased, but are assessed to be slightly below the source-related dose constraint for members of the public for a new facility of 0.15 mSv yr\(^{-1}\) even if the pessimistic assumption is made that the carbon-14 is released predominantly as carbon monoxide. Releases as 100% methane would be well below the design target level.

- In the post-closure period, the medium- and long-term post-closure risks are calculated to be above the risk guidance level only where all the release is focused to a small area (assuming that 10% of the carbon-14 is released as methane).
A clear implication of these results is that it will be important to understand the gas migration characteristics of any site proposed for the disposal of these wastes. It is also important to understand the speciation of carbon-14 released from irradiated steel wastes.

Alternative treatment, packaging, design and disposal options are not particularly beneficial for these wastes, but could be considered in future for specific waste streams, should they prove to be problematic.

Nevertheless, for irradiated steel wastes, consideration should be given to the following aspects:

- Whether the distribution of carbon-14 might not be uniform for any particular waste streams (as is assumed in the model), e.g. due to self-shielding of thick or peripheral activated components.
- Whether a different corrosion rate might be applicable to specific waste streams (taking into account the possible effects of irradiation and sensitisation of stainless steel wastes); this may be informed by current work under the NDA’s Direct Research Portfolio (DRP) concerned with corrosion behaviour of spent AGR fuel and fuel cladding during interim storage [99].
- Whether the carbon-14 inventory could be overestimated, or particularly for new build wastes, for which the inventory is indicative only at this stage, whether material specifications could be chosen to reduce the expected inventory.
- Whether there could be additional surface precipitation of carbon-14 from coolant gas onto reactor steel components during reactor operations and whether this additional carbon-14 could remain following packaging. This is not currently included in the carbon-14 inventory.

It is noted also that the UK RWI is updated every three years and future improvements to the carbon-14 inventory are expected as further understanding is gained of UK legacy wastes including steel wastes from reactor decommissioning programmes and of NNB wastes as the new build programme proceeds.

Ongoing work on irradiated steel wastes is focussing on:

- Experiments to measure the release of carbon-14 from irradiated stainless steel in anaerobic environments, which are in progress under the international CAST project. The implications of the findings of CAST for the assessments presented in this report will be considered as part of the CAST project.

Possible future work includes:

- Consideration could be given to the applicability of these measurements to the aerobic period and to other steels (although this is not currently a priority).
- Work on the corrosion rate of irradiated stainless steel, and in particular to what extent sensitisation of the steel affects the metal (i.e. intergranular attack and IGSCC) and the general corrosion of the bulk material (potentially building on the DRP AGR fuel work). However, given the difficulties associated with testing irradiated materials, these effects may be better studied using ‘what-if’ calculations, as has been done in this work.

Our improved understanding also has implications for the assessment of the groundwater pathway in terms of the inventory available for release and the speciation of carbon-14 in the aqueous phase. Account could be taken of the long timescales over which carbon-14 would be released from steel wastes.

A summary of the current position is provided in Table 37. The status is described in the context of the ‘AND’ questions identified in Section 1.3. Certain next steps are also identified in the Table.
Table 37  Summary of the current position on irradiated steel wastes

<table>
<thead>
<tr>
<th>‘AND’ Question</th>
<th>Current Status</th>
</tr>
</thead>
<tbody>
<tr>
<td>Is there a significant inventory?</td>
<td>Yes. There is a significant inventory of carbon-14 in irradiated steels (7,060 TBq, of which 6,660 TBq is associated with NNB). The focus has been on a few legacy waste streams where a significant inventory is in irradiated steels that have relatively small thickness.</td>
</tr>
<tr>
<td>Does the waste stream generate carbon-14 bearing gas?</td>
<td>Yes, but it is not certain what proportion of the carbon-14 is released as gas. It is likely that there is some methane released as the metal corrodes; some carbon-14 may be released as other species, some of which may partition into solution. Experiments are starting to examine how much carbon-14 is released as a gas, and the assessments will be updated when these are complete.</td>
</tr>
<tr>
<td>Is there a bulk gas phase to entrain the carbon-14 bearing gas?</td>
<td>These wastes generate hydrogen, very slowly, when they corrode anaerobically. Corrosion is not expected to be limited by water availability. Hydrogen will also be generated from corrosion of inactive steel waste containers. In the longest time frame, there may not be a bulk gas phase.</td>
</tr>
<tr>
<td>Do these gases migrate through the near field in significant quantities?</td>
<td>Any trace $^{14}\text{CO}_2$ in the gas phase is expected to react with cementitious materials (in package grout or surrounding backfill) – there is no bulk $\text{CO}_2$ from these wastes. $^{14}\text{CH}_4$ and $^{14}\text{CO}$ would migrate through the near field in most concepts.</td>
</tr>
<tr>
<td>Do these gases migrate through the geological environment in significant quantities</td>
<td>This is a site specific question. Some geological environments (e.g. LSSR and evaporite) would provide a significant barrier to gas migration or may allow the gas time to dissolve in groundwater. Important factors are the time taken for the carbon-14 to migrate to the biosphere and the area over which any release is spread.</td>
</tr>
<tr>
<td>Do these gases lead to significant doses in the operational phase?</td>
<td>No.</td>
</tr>
<tr>
<td>Do these gases lead to significant risks to potentially exposed groups in the post-closure phase?</td>
<td>Impact is site specific. The radiological risk is tolerable in most environments. Even where there is a focused release, provided it is found that 10% or less of the carbon-14 from these wastes is released in gaseous form, then the release is expected to be acceptable.</td>
</tr>
</tbody>
</table>

10  Acknowledgements

Colin Leung of Amec Foster Wheeler is thanked for undertaking some of the modelling calculations described in this report. We also thank Arvic Harms of the National Physical Laboratory for performing the experiments described in Appendix 1.
11 References


32 AREVA, *Design Control Document Rev.5 - Tier 2 Chapter 04 - Reactor - Section 4.3 Nuclear Design*, ML13220A6782013, 2013.


99 Nuclear Decommissioning Authority, Research and Development. 5 year Research and Development Plan, Issue 1, 2013.
Appendix 1
Analysis of the distribution of carbon-14 in mild steel from Windscale Pile no. 2
Introduction

A mild steel sample which originated from Windscale Pile 2 and was held in storage by the National Physical Laboratory (NPL), was identified as being potentially suitable for use in experiments to study the release of carbon-14 from irradiated mild steel. To evaluate the suitability of the sample for use in leaching experiments an analysis was made of its radionuclide content by NPL. The analytical methods used and the results obtained are described below.

Irradiated mild steel

A mild steel sample (ID IM06070860) weighing 1.609 kg, 15 cm in length and 4 cm diameter, which originated from Windscale Pile 2 Corehole 1, was held in storage at NPL.

No information was available on the initial composition of the steel batch from which the sample had originated. Semi-quantitative analysis by energy dispersive X-ray analysis in a scanning electron microscope of a low specific activity coupon of the steel found that the material contains about 0.6% manganese with the balance iron. The carbon or nitrogen content could not be detected above background.

Initial measurements indicated the following activities for the sample (with no other radionuclides detected).

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Activity (kBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cobalt-60</td>
<td>45</td>
</tr>
<tr>
<td>Nickel-63</td>
<td>19</td>
</tr>
<tr>
<td>Iron-55</td>
<td>45</td>
</tr>
</tbody>
</table>

The initial cobalt-60 values were estimated by taking the total activity for the 'active' (i.e. reactor side) end and the activity for the 'not-so-active' (i.e. bioshield side), assuming that the profile across the bar is exponential and integrating this profile by calculating the activity at 1 cm intervals and summing these values. The iron-55 and nickel-63 values were estimated by taking the measurement ratio for a Pile 1 steel sample and using these as a fingerprint. The uncertainties (at $k=1$) were estimated as: cobalt-60: +100%, −50%, nickel-63: +110%, −60% and iron-55: +110%, −70%.

Experimental

The carbon-14 and gamma-emitting radionuclide activities in Pile 2 steel sample IM06070860 were determined in February 2009 by NPL. Six sub-samples were taken at regular intervals along the length of the bar and analysed for their carbon-14 content by destructive radiochemical analysis; gamma emitting nuclide activities were measured by gamma spectrometry. The sub-samples, which ranged from 1.7 to 2.4 g in weight, were obtained by drilling six holes with a diameter of 0.9 cm along the length of the bar. The six sub-samples were dissolved in 6 mol dm$^{-3}$ hydrochloric acid and the resulting gases were combusted in a 'Pyrolyser', containing a Pt/Al$_2$O$_3$ catalyst, in an oxygen (50%) rich atmosphere at 800°C. The off-gases were led through bubblers containing water to collect any tritium and subsequent bubblers containing 3-methoxypropylamine to collect any $^{14}$CO$_2$. After 24 hours operation, aliquots of each 3-methoxypropylamine solution were mixed with liquid scintillation cocktail (Readysafe) and the six samples were counted by LSC to determine the $^{14}$C activities. The six residual 6 mol dm$^{-3}$ hydrochloric acid solutions were each collected and analysed with gamma spectrometry.

Results

The results of the mild steel analysis are presented in Table 38 below. The only gamma-emitting radionuclide identified was cobalt-60.
Table 38  Results of the carbon-14 and cobalt-60 analysis of the irradiated mild steel sample

<table>
<thead>
<tr>
<th>Sub-sample</th>
<th>Distance (cm)*</th>
<th>$^{14}$C specific activity (Bq g$^{-1}$)</th>
<th>$^{60}$Co specific activity (Bq g$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.75</td>
<td>5.1</td>
<td>149</td>
</tr>
<tr>
<td>2</td>
<td>3.5</td>
<td>1.2</td>
<td>32</td>
</tr>
<tr>
<td>3</td>
<td>6.2</td>
<td>0.51</td>
<td>6.5</td>
</tr>
<tr>
<td>4</td>
<td>8.7</td>
<td>0.53</td>
<td>2.0</td>
</tr>
<tr>
<td>5</td>
<td>11.4</td>
<td>0.63</td>
<td>&lt;0.6</td>
</tr>
<tr>
<td>6</td>
<td>13.85</td>
<td>0.70</td>
<td>&lt;0.6</td>
</tr>
</tbody>
</table>

*The distance of the centre of the hole from the reactor side

The total activities have been estimated assuming that the profile across the bar is exponential (except between 0 and 2.2 cm from the reactor side, where it is assumed to be constant) and integrating this profile by calculating the activity at specific intervals (depending on the distance between the sample points) and summing these values. The calculation has been independently checked.

The total activity of carbon-14 in IM06070860 is estimated to be 2.3 kBq (inhomogeneously distributed). The total activity of cobalt-60 in IM06070860 is estimated to be 48 kBq (inhomogeneously distributed), which agrees well with the initial estimate of 45 kBq.

**Conclusion**

The estimated carbon-14 content of the mild steel is significantly less than was envisaged at the start of the project and is too low for the sample to be suitable for use in carbon-14 release experiments, as originally planned. Given the results obtained, leaching experiments with this sample of mild steel were cancelled.