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Geological Disposal of Graphite Wastes

George Towler, James Penfold, Laura Limer[1] and Elizabeth Arter

[1]Limer Scientific Consulting Ltd

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Prepared by:	George Towler1Limer Scientific Consulting Ltd		
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Quintessa Limited
The Hub, 14 Station Road
Henley-on-Thames
Oxfordshire RG9 1AY
United Kingdom

Tel: +44 (0) 1491 636246
Fax: +44 (0) 1491 636247
info@quintessa.org
www.quintessa.org
www.quintessa-online.com

Summary

The European Commission (EC) CARBOWASTE project, in which NDA RWMD is participating, is currently exploring options for the management and disposal of irradiated graphite. RWMD's contribution to the CARBOWASTE project concerns the post-closure aspects of disposal of irradiated graphite wastes.

NDA RWMD has commissioned a study to support their contributions to CARBOWASTE. This report is the output from the second phase of work in that study (Phase 2). The objective of Phase 2 of the study (i.e. this report) is to examine the suitability of irradiated graphite wastes for geological disposal. This is achieved by testing whether graphite can be safely disposed in a wide range of disposal systems, where a disposal system comprises the near-field and geosphere barriers.

The analysis is not tied to a specific disposal concept, geosphere environment, or facility depth. Instead the performance of the different barriers that may be included in a disposal concept and can contribute to the post-closure safety is explored. This report is therefore relevant to all organisations considering geological disposal of graphite (RWMD, other participants within CARBOWASTE and other waste management organisations, including those considering shallow geological and near-surface disposal).

Calculations in support of the analysis have been undertaken for the UK national inventory of irradiated graphite wastes, which comprises a significant volume of waste and a significant associated radionuclide inventory. Therefore conclusions that are drawn regarding the suitability of irradiated graphite wastes for geological disposal are relevant to large waste inventories, not only small quantities of irradiated graphite.

The analysis presented in this report has demonstrated that it should be possible to safely dispose of irradiated graphite wastes in isolation (i.e. in vaults containing only packages of graphite wastes) in a wide range of disposal systems (i.e. combination of disposal concept / Engineered Barrier System (EBS) and geosphere); including near-surface, shallow and deep geological disposal and a wide range of host rocks. Assessment calculations show that regulatory guidelines can be satisfied even given conservative assessment assumptions. A broader range of systems might be suitable given less conservative calculation assumptions. One particular issue that potentially requires careful management is the potential impacts associated with disruption of, or large scale intrusion into, near-surface facilities.

It may also be possible to safely dispose of irradiated graphite wastes in the same vaults as other ILW in a wide range of disposal systems. However, a broader range of processes become important, behaviour becomes more site / design specific and the

important scenarios and behaviours may change as the system evolves. This makes it difficult to generically explore the suitability of graphite for geological disposal with other ILW.

Specific waste types of concern are those that give rise to bulk gas generation (i.e. metals, organics, strongly irradiating wastes) and that might lead to incorporation of C-14 in methane gas (i.e. organics), and therefore increase the potential for generation and transport of C-14 labelled gases. The potential for transport of C-14 labelled gases is particularly important for fractured host rocks and potentially for lower permeability host rocks where the concept includes an Engineered Gas Transport System (EGTS) to control the peak gas pressure.

Therefore, although it may not be necessary in all cases, there are advantages to disposing graphite wastes in isolation compared with co-disposal in the same vaults as other ILW. These include:

- simpler, more predictable behaviour;
- improved performance, e.g. transport in gas not likely to be an issue; and
- simplified safety arguments and safety case.

In the UK, the majority of the graphite waste (by volume and radionuclide inventory) is classified as Shielded ILW (SILW). SILW contains relatively little quantities of organic and reactive metals, which are largely associated with Unshielded ILW (UILW) wastes. Further segregation of graphite from other SILW should be relatively straightforward and might offer performance benefits, for example through the ability to optimise the disposal concept for graphite wastes.

If transport of C-14 in gas is of concern for segregated graphite waste packages, e.g. potentially in a fractured host rock, it is likely that further performance benefits would be obtained from disposing graphite in concrete containers rather than steel containers, thereby reducing bulk gas generation to a very low level. This would also reduce the potential for reduction of inorganic C-14 to $^{14}CH_{4(g)}$ by autotrophic bacteria, using H_2 derived from anaerobic corrosion of steel, to a very low level. The potential significance of this process for isolated packages of graphite waste in steel containers is uncertain, although the available information and expert judgements indicate that it is unlikely to be significant.

At the generic level of this report, factors that may limit the range of disposal systems suitable for geological disposal of graphite wastes include assumptions regarding the nature of the biosphere into which releases may occur, which tend to be more conservative for generic assessments such as those undertaken in this study; uncertainties regarding radionuclide release from graphite wastes; and cautious assumptions in assessment calculations.

Although significant work has been undertaken within CARBOWASTE to better understand radionuclide release from graphite, uncertainties remain regarding:

- release mechanism(s), the form(s) and release rate(s) of C-14 labelled gas(es) from dry, damp and wet graphite wastes;
- the mechanism(s) for long-term release of C-14 to water;
- the fraction of the C-14 inventory that is released; and
- the potential for release of C-14 and Cl-36 in aqueous organic compounds, and their subsequent fate and transport.

Due to these uncertainties, safety assessment calculations often include cautious assumptions that are likely to overestimate the amount of C-14 released and potentially the rate of C-14 release. It is difficult to represent the complex geochemical behaviour of C-14 in the near-field in simple safety assessment calculations. Consequently, such calculations tend to include cautious assumptions that underestimate the chemical barrier performance for concepts including a cementitious EBS.

These uncertainties do not preclude safe disposal of graphite wastes in a wide range of disposal systems, but as stated above a wider range of disposal systems might be suitable if these uncertainties were further reduced.

A second consequence of these remaining uncertainties, and how they are accounted for in assessments, is that it may be difficult to demonstrate that a disposal concept is optimised. This may mean that a cautious approach to engineering design may be required, potentially involving higher-specification near-field barriers than might be appropriate if waste guideline performance were better understood.

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1 Introduction

The European Commission (EC) CARBOWASTE project, in which NDA RWMD is participating, is currently exploring options for the management and disposal of irradiated graphite (Banford et al., 2008; EC, 2008). RWMD's contribution to the CARBOWASTE project concerns the post-closure aspects of disposal of irradiated graphite wastes.

NDA RWMD has commissioned a study to support their contributions to CARBOWASTE. This report is the output from the second phase of work in that study (Phase 2). It builds on work undertaken during the first phase (Phase 1), as reported by Towler et al. (2011).

The current UK reference assumption for the disposal of irradiated graphite wastes, particularly large volumes of reactor core graphite, is disposal in a Geological Disposal Facility (GDF). Other countries are adopting similar approaches, considering a range of different disposal facility concepts and depths, including near-surface and shallow[1] geological disposal (e.g. Ozanam, 2009).

In the UK, the base assumption is that irradiated graphite wastes will be cement encapsulated in stainless steel containers, which will be stacked in the ILW vaults of a GDF. However, for the majority of graphite wastes final decisions have yet to be made concerning conditioning and packaging, and therefore there is the opportunity to compare different options, which might have important advantages and disadvantages given the large volume of irradiated graphite wastes and the radionuclide inventory associated with them.

The objective of Phase 1 of the study was to better understand the potential behaviour of graphite wastes in a GDF, how the post-closure performance of the GDF is influenced by different disposal options (including different choices of host rock), and the potential impact of our current uncertainties in process understanding and parameterisation. In particular the study focussed on the behaviour of C-14, which potentially displays complex behaviour in the GDF meaning that there was scope to better understand the implications of different disposal concepts. The outputs from Phase 1 will inform RWMD's consideration of an optimised graphite disposal strategy.

The objective of Phase 2 of the study (i.e. this report) is to examine the suitability of irradiated graphite wastes (herein simply referred to as graphite) for geological

[1]For example, shallow geological disposal is the reference scenario in France, but Andra have not yet decided whether irradiated graphite will be disposed to a shallow or deep facility, or treated.

disposal. This is achieved by testing whether graphite can be disposed in a wide range of disposal systems, where a disposal system comprises the near-field and geosphere barriers.

The combined outputs from Phase 1 and Phase 2 will inform RWMD's development of any future research programme, if additional research is required to address any residual uncertainties following completion of the CARBOWASTE project.

For Phase 2, 'disposability' is considered in the context of safety assessment calculations, and therefore the outputs from this report can be used to inform concept selection, for example to build confidence that a concept will provide the required performance before undertaking a full safety assessment. However, this report does not directly consider issues such as storage, transport and operational safety, or optimisation of the disposal system in terms of post-closure performance (e.g. complexity versus simplicity and reliability, homogeneity and predictability versus flexibility, etc) or indeed wider socio-economic issues.

The focus of this report is therefore on the behaviour of, and potential impacts associated with, the key radionuclides for post-closure safety. Previous studies (e.g. Limer et al., 2010) have established that the key radionuclides are C 14 (which may be released in radiolabelled gases during both the GDF operational and post-closure phases, and water during the post-closure phase), and Cl-36 (which may also be released in water during the post-closure phase).

Although optimisation of the disposal system is not directly considered in this report, analysis of the requirements, role and performance of individual barriers is relevant to demonstrating that the design has been optimised. The analysis presented in this report can also be used to identify the key safety features / functions and develop safety arguments for the performance of multiple barriers and defence in depth of the design. Understanding of the disposal system and identification of key safety features / functions and their optimisation are key components of the overall safety case.

Some of the performance related issues are complex and it is not possible to draw conclusions without examining a site specific disposal system. Examples of such issues are multi-phase flow and transport through vault and tunnel seals, or the consequences of human intrusion and disruptive events. Such issues are identified and considered in this report, but the treatment is naturally more generalised. The report therefore highlights issues that will need to be considered in detail in a full safety assessment.

The following are not considered: non-radiological (i.e. chemical) contaminants that could be associated with graphite; 'exotic' graphite wastes, such as those that may be

highly contaminated with fission products[2]; non-human biota; and maximum acceptable environmental concentrations.

The analysis is not tied to a specific disposal concept, geosphere environment, or facility depth. The findings are therefore relevant to all organisations considering geological disposal of graphite (RWMD, other participants within CARBOWASTE and other waste management organisations, including those considering near-surface and shallow geological disposal).

1.1 Approach

In order to examine the disposal system performance required for post-closure safety this study builds on the approach of Limer et al. (2010), who examined acceptable radionuclide release rates to the biosphere, for the key radionuclides associated with graphite, i.e. C 14 and Cl-36. Limer et al. (2010) examined a range of relevant assessment biospheres, and determined the required system (i.e. near-field and geosphere) performance. However, Limer et al. (2010) did not fully consider how the required near-field and geosphere performance might be achieved. This is further explored in this study, which therefore builds on the work of Limer et al. (2010).

The first step of the approach is to define the barrier functions that control the potential impacts from graphite. These barrier functions reflect the nature of irradiated graphite wastes, radionuclide release and transport, and regulatory guidance regarding required disposal system performance (Section 2). As part of the background context, it is also useful to consider the approaches typically used to calculate barrier performance, and any associated key assumptions and simplifications.

The next step is to examine the near-field and geosphere properties required to provide the necessary barrier functions. The analysis considers Normal Evolution Scenarios (NES, Section 3) and Human Intrusion and Disruptive Event Variant Scenarios (HI and DE, Section 4).

Analysis of the NES is undertaken in two steps. Firstly disposal of packages of graphite wastes independent of other waste types is considered. This is the simplest possible system. Then disposal of packages of graphite wastes with packages of other ILW (co-disposal) is considered. These are much more complex systems. These other ILW wastes may also be important sources of the key radionuclides associated with graphite (C-14 and Cl-36). However, the analyses do not include these additional

[2]The UK waste inventory does not include 'exotic' graphite wastes. However, it is recognised that other countries do have such wastes.

sources, or the consequences of the different timings and rates of radionuclide releases from the different wastes.

In order to examine the near-field and geosphere properties required to achieve regulatory guidance levels, it is necessary to make some assumptions about the biosphere (see Section 2.4). Different biosphere assumptions will lead to more or less stringent functional requirements. Therefore the analysis presented in this report is not intended to replace site specific safety assessments. However, it is appropriate to make representative assumptions about the biosphere in order to demonstrate the disposability of graphite wastes, i.e. appropriate barrier properties can readily be achieved.

Finally, conclusions are drawn regarding the suitability of graphite wastes for geological disposal (Section 5). Residual uncertainties subsequent to the work that has been undertaken within CARBOWASTE are identified, and the consequences are discussed.

2 Background

2.1 Irradiated Graphite Wastes

Graphite has low chemical reactivity. There are not expected to be any significant chemical reactions between graphite and water or gases in the GDF; between graphite and waste package / container and EBS materials; or between graphite and the host rock. Enhanced corrosion of metal containers might be a concern due to galvanic coupling, however experiments undertaken by Smart et al. (2004) showed that in cementitious environments, galvanic coupling between graphite and stainless steel does not significantly alter corrosion rates.

Graphite is also a structurally strong material, such that it will support repository stability for concepts where the mechanical performance is important, for example the concepts being considered by Andra (2005a,b) and Nagra (2008) for deep geological disposal of ILW in indurated mudstone host rocks. For graphite that has experienced high mass loss in the reactor environment, and consequently has a high porosity and reduced strength, the implications for mechanical performance of the GDF might need to be considered.

Given its strength and low reactivity, graphite is therefore suitable for geological disposal. Further information on the properties of the bulk material and its suitability for geological disposal is provided in the Phase 1 report (Towler et al., 2011). Challenges to the disposal of irradiated graphite wastes arise due to the inventory and behaviour of the associated radionuclides, particularly Cl-36 and C-14.

For large quantities of irradiated graphite wastes, e.g. Magnox and AGR reactor cores in the UK context, the inventories of Cl-36 and C-14 may be substantial. A significant fraction of the Cl-36 inventory may be released rapidly, as inorganic chlorine, upon contact with water. Chloride ions are highly soluble and not significantly chemically reactive therefore Cl-36 is mobile in water. Due to the mobility and long half-life of Cl-36, it has the potential to migrate from the GDF to the biosphere. C-14 is released more slowly upon contact with water, and it has a shorter half-life and is more chemically reactive than Cl-36. It exhibits complex behaviour partitioning between water and gas, inorganic and organic phases. The potential for incorporation of C-14 in gas, and its subsequent transport is of particular concern.

Radionuclide release and transport is further described in the subsequent sections.

2.2 Radionuclide Release from Irradiated Graphite

It is beyond the scope of this report to summarise, discuss and integrate all the available information on the formation and distribution of radionuclides in irradiated graphite, and radionuclide release from irradiated graphite. There is significant debate surrounding the key mechanisms and processes, and significant variability in findings, for example depending on the initial characteristics of the non-irradiated graphite, the conditions the graphite has been exposed to both within the reactor and subsequently, and experimental set-up (e.g. for leaching experiments).

The Phase 1 report (Towler et al., 2011) provides a summary of the current understanding of the forms in which radionuclides are present in graphite and the distribution between these forms. The distribution is important because it affects the availability and hence release of radionuclides from graphite. There is ongoing debate surrounding the relative importance of different formation processes for C-14, and ongoing research into the forms and distribution of C-14 and Cl-36. However, the Phase 1 report remains relatively up-to-date and therefore the reader is referred to that report for more information on generation processes and radionuclide distribution.

The Phase 1 report also discusses release mechanisms and rates. While the Phase 1 reported noted that C-14 could be released in gas, only limited information was available at that time and the focus was on radionuclide release by leaching. In particular empirical information available from leaching tests was reviewed and best estimate radionuclide release rates we proposed for generic performance assessment modelling (Table 2-1). It was noted that most leaching studies have been concerned with releases associated with core dismantling under water and therefore have been short duration experiments circa 100 days. Only a limited number of longer-term studies have been undertaken, for example Takahashi et al. (1999) reported leaching rates (in alkaline solution) after 720 days. Actual release rates are sensitive to factors such as the graphite origin, reactor operating conditions and experimental conditions. For performance assessment calculations it was cautiously assumed that the entire inventory of C-14 and Cl-36 is available for release on post-closure timescales.

Table 2-1. Radionuclide leach rates for generic performance assessment

Radionuclide	Fractional Leaching Rate (y^{-1})	
	Very Early Period (≤ 30 days)	Later Period (> 30 days)
C-14	2.0E-02	1.7E-02 to 1.83E-5
Cl-36	1.1E+01	3.7E-01

Work undertaken as part of CARBOWASTE provides significant further information regarding leaching mechanisms for C-14 and Cl-36, and release of C-14 in gas. This new information is discussed below and is related to some of the existing literature considered in the Phase 1 report.

Cl-36

Vaudey et al. (2010) examined chemical speciation of chlorine in virgin (i.e. non-irradiated) graphite, and evolution with temperature. They found that chlorine is present in two forms: c.30% inorganic chlorine, which probably comprises ClO_2^- and ClO_3^- compounds; and c.70% organic chlorine bound to carbon in graphite. It was found that Cl was leached as HCl, indicating that the inorganic fraction is more labile. The release rate of inorganic Cl increases with temperature, while the release rate of organic Cl is less temperature sensitive.

Cl-36 is formed through activation of stable chlorine. Gray and Morgan (1988) found that a small fraction (<0.2%) of Cl-36 in irradiated Hanford graphite is labile and rapidly leached. Once this labile fraction had been leached, the cumulative amount of Cl-36 leached did not appreciably increase. The leach rate increased slightly with increasing leachate temperature. The low release fraction was confirmed by CEA leach tests on sub-samples of the same material (Comte and Petit, 2011).

Gray and Morgan (1989) found similar results for irradiated French graphite, but the labile fraction was up to 39%. Sensitivity to leachate temperature was not examined. CEA tests confirmed that the labile fraction is higher in French graphite, but they measured a labile fraction of only 1.7% for a sub-sample of the same material that had a labile fraction of 39% in Gray and Morgan's tests (Comte and Petit, 2011). Subsequent work by CEA has demonstrated that the difference was due to the size of the sub-samples, and occurred because the release of Cl-36 is diffusion controlled (Comte and Petit, 2011).

Further leaching tests by CEA have demonstrated labile fractions up to 90%, with release occurring during the first 2 months of leaching (Comte and Petit, 2011). The labile fraction is correlated to the thermal history of the graphite; there is no significant correlation with the sample Cl-36 inventory, irradiation history, leachate chemistry or temperature within the bounds studied. Also, release is not dependent on water impregnation into the graphite (Comte and Guy, 2008). The labile fraction decreases with increasing operating temperature (Comte and Petit, 2011).

Comte and Petit (2011) fitted a bi-modal diffusion model to the results of a 450 day leaching experiment. Effective diffusivities are derived for the labile fraction of the inventory, and the fraction that diffuses more slowly. The fitted diffusivities correspond with measurements for diffusion of Cl-36 through non-irradiated graphite. Comte and Guy (2010) examined the chemical form of Cl in the leachate using ion chromatography. The majority of Cl was present as chloride and a small amount as chlorite.

From the available evidence it is uncertain whether the fraction of Cl-36 that Comte and Petit (2011) observed to diffuse more slowly might correlate with Cl-36 in organic form in the graphite, whether chlorine was also released in an organic form, and whether hydrolysis of organic chlorine to inorganic chlorine occurred (either in solution or as the release mechanism).

It is concluded that release of Cl-36 from graphite will be sensitive to the thermal history of the graphite and the size of the pieces of waste graphite that are disposed. The majority of the Cl-36 inventory may be released slowly or rapidly by diffusion. For the fraction that diffuses more slowly, the effective diffusivity may be of the order of 2E-14 $m^2 s^{-1}$ (Comte and Petit, 2011). The time for diffusion of Cl-36 out of the waste can be estimated:

$X_{RMS} = \sqrt{(2.n.D_e.t)}$

Where

X_{RMS} is the root mean squared distance diffused (m)

n is 1 for linear, 2 for cylindrical and 3 for radial (-)

D_e is the effective diffusivity ($m^2 s^{-1}$)

t is the time (s)

Figure 2-1 shows that for large pieces of graphite waste, that have been exposed to high operating temperatures, the waste itself may be a significant barrier to radionuclide release.

The Phase 1 report (Towler et al., 2011) notes various lines of evidence as to the volatility of Cl-36, and that significant release is possible prior to disposal. The evidence for volatility is generally associated with graphite dust and powder and is therefore consistent with the new information described above.

For the purposes of this study it is cautious to assume that there is little volatile release of Cl-36 prior to disposal and a significant proportion of the Cl-36 inventory is labile and is released instantaneously compared with post-closure timescales and the half-life

of Cl-36. Since risk scales linearly with radionuclide flux, and hence the labile inventory, in the context of the objectives of this report it is not overly cautious to make the bounding assumption that all Cl-36 is released instantaneously upon contact with water, unless the overall labile fraction is less than circa 5% to 10%.

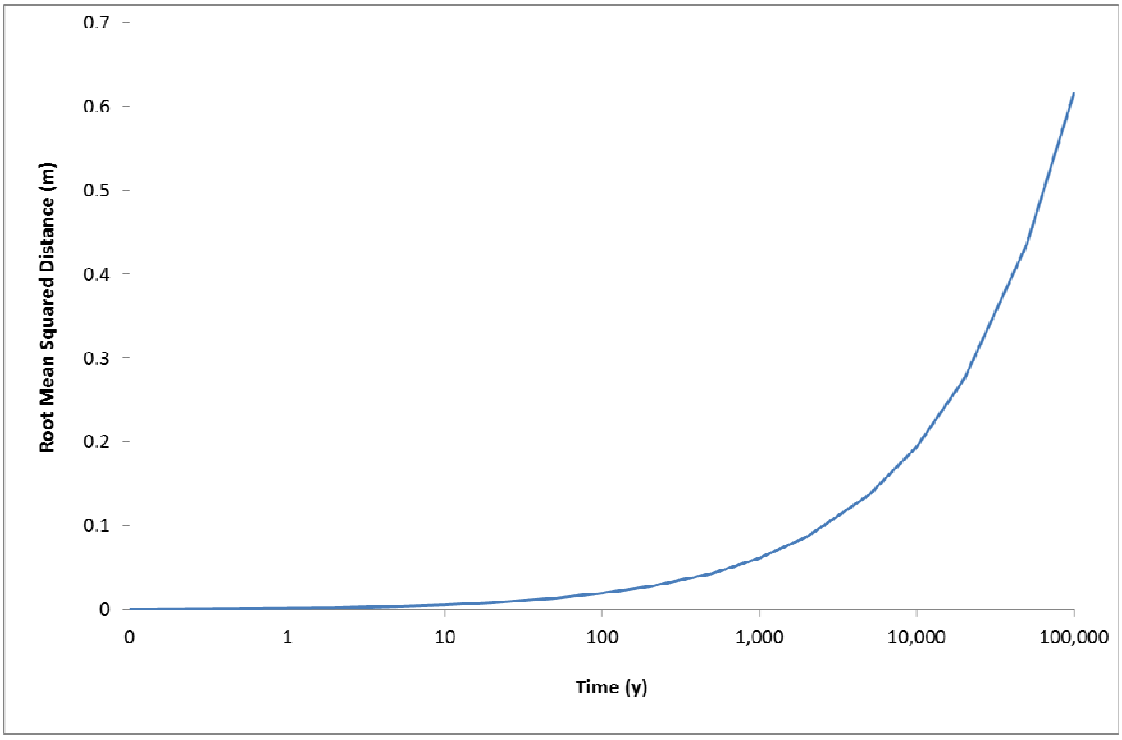

Figure 2-1. Radial diffusion from graphite of the fraction of Cl-36 that diffuses more slowly

C-14

The majority of C-14 is formed by activation of ^{13}C in the graphite, and ^{14}N impurities in the graphite originating from manufacture. C-14 is displaced from the graphite lattice upon formation, but is progressively re-integrated into the lattice. Therefore C-14 may be present in carbon in the graphite lattice and in a variety of interstitial positions: as discreet atoms, lines and small new 'rings', and bridging positions between the individual layer places in the graphite crystallites. C-14 may also be present in carbonaceous deposits in the open porosity and on the geometrical surface due to activation of N impurities in the coolant gas, and O in CO_2 and air cooled reactors.

The distribution of C-14 between different forms and locations implies that initial release rates may be high as C-14 is released from geometrical surfaces and open pores,

but then will decrease because the remaining C-14 is in more stable forms.The fraction of C-14 released from Hanford graphite and French (UNGG) graphite in leaching experiments is low (<1%) (Comte and Petit, 2008).The form and distribution of C-14, and the release mechanisms associated with each form, are not fully characterised. This means that, in the absence of long-term empirical data, the long-term release rates are uncertain.

As a result, for safety assessment calculations, a conservative approach is often taken whereby release rates measured at the end of leaching experiments are applied throughout the post-closure assessment period to the entire C-14 inventory, thereby potentially overestimating the rate of release and the total fraction released. For example, there is evidence that the majority of C-14 in the graphite might not be released.

- Under harsh acidic environments no more than 30% of C-14 is leached (Banford et al., 2012).
- Workers at Hanford (Gray, 1982) deduced that release of C-14 from the matrix is by oxidation, not by the water but from oxygen dissolved in it. This implies that release from the matrix will decrease as anoxic conditions are established in the GDF, which is anticipated to occur on a timescale of a few years, to a few decades for vaults with a low chemical oxygen demand. Therefore it is possible that there may be negligible long-term release of C-14 from the graphite matrix in the GDF environment, only short term release from the matrix concurrent with release of C-14 in labile forms.
- Grambow (2010) concluded that C-14 is not likely to be released from the matrix of French graphite due to radiolytic damage of the graphite structure, once the inventory of Co-60 has decayed. Therefore radiolytic release is likely to cease before the disposal facility is closed and Grambow (2010) concluded that radiolytic release is unlikely to result in significant C-14 release from the matrix.

The results of long-term (431 day) leaching experiments have recently been published by NDA RWMD (Marshall et al., 2011). These experiments used BEPO reactor graphite and were undertaken under high pH conditions. Previous experiments had noted that in addition to leaching there was some direct release of C-14 to gas. Marshall et al. (2011) focused on measuring the chemical forms of C-14 released to water and gas, and characterising the gas release rates.

Marshall et al. (2011) reported that C-14 was released as inorganic carbon in solution (presumably carbonate ions under the high pH experimental conditions), CO gas and

organic gas (assumed to be CH_4). It has been assumed that inorganic C-14 gas was in the form of CO because this is the radiolytic oxidation product formed in the reactor.

Since generation of CO by oxidation under leaching conditions is only possible at an exceedingly slow rate, the implication is that this was release of adsorbed CO, presumably initiated by exchange with water. However, the experimental method was unable to distinguish C-14 released as CO gas 'directly' from the graphite, from C-14 leached from the graphite and subsequently released as CO_2 gas due to carbonate equilibria. An alternative explanation is that the experimental method might have resulted in some degassing of CO_2 (and hence $^{14}CO_2$) from solution, because CO_2 was 'stripped' from the air entering the experimental vessel, such that the partial pressure of CO_2 was very low, and potentially out of equilibrium with the solution. Conversely, release of $^{14}CO_2$ would have been limited by the low concentration of carbonate in solution due to the high pH conditions.

The release rate to gas was found to be small compared with the leaching rate to water (Table 2-2). However, the gas release rate exhibited characteristic leaching behaviour, with a high initial release rate that fell rapidly with time. After 431 days the cumulative release of C-14 in gas had nearly plateaued. The 'apparent' labile fraction of the inventory was small: less than the uncertainty in the total C-14 inventory.

This behaviour[3] raises some important issues. Firstly, the evolving and rapidly declining gas release rate implies that there was no significant release prior to contact with water, which indicates a geochemical reaction requiring the presence of water. As described previously release may have been due to carbonate reactions or release of adsorbed CO by exchange with water. If gas release was associated with exchange of adsorbed CO with water, there may have been slow release prior to the experiment due to exchange with water vapour in the atmosphere. The release rate then accelerated rapidly upon contact with liquid water and release rapidly completed. Since release of C-14 in organic gas exhibited the same behaviour, this may also have been release of adsorbed gas.

Release of C-14 in gas has also been observed from shut-down reactors waiting to be decommissioned. For example, the UK is adopting a 'SafeStore' strategy whereby reactor cores are subject to a period of decay storage lasting several decades prior to dismantling and disposal of the wastes. A number of UK reactors are now defueled and are passively vented. There is direct release of C-14 labelled gases from these 'dry' (atmospheric humidity), passively vented cores, e.g. Magnox (2008). The most likely

[3]The experiment was undertaken using BEPO reactor graphite. BEPO is an air cooled reactor so the amount of C-14 coming from activation of $N_{2(g)}$ may higher than in CO_2 cooled reactors, resulting in different C-14 distribution, e.g. an increased fraction of C-14 in deposits on geometrical surfaces and in open pores. However, the general pattern of leaching behaviour is consistent with experiments using graphite from a range of other reactor types.

source of C-14 labelled gases is the graphite reactor core. The chemical form of the C-14 gas and release mechanism have not been reported, however release might be due to desorption of gases in response to exchange with water vapour in the atmosphere.

It is anticipated that the flux of gas will have decreased significantly by the end of the SafeStore period. It is possible that there may be a period of enhanced gas release during and following encapsulation with cement, due to contact with bleed water. This might significantly reduce the potential for gas release in the GDF. Additional information on the form and rate of direct release to gas would be beneficial since this may be important for understanding disposal facility performance, particularly where a SafeStore strategy is not being adopted and for disposal concepts where the wastes are not encapsulated. In the absence of a mechanistic understanding of direct releases to gas, it will be cautious to assume a small long-term direct release of $^{14}CH_4$ gas in the safety assessment, e.g. 1E-5 y-1 (Table 2 2).

Table 2-2. Amounts of C-14 released from BEPO reactor graphite during leaching experiment conducted by Marshall et al. (2011), (initial inventory 2.1E6 Bq +/- 0.2 MBq).)

Species	Amount released after 431 days (Bq)	Average Release Rate (y^{-1})
$CO_3^{2-}{}_{(aq)}$	2145	8.7E-4
$CO_{(g)}$ and $CO_{2(g)}$*	87.5	3.5E-5
$CH_{4(g)}$	24.8	1.0E-5

* CO and CO_2 gas cannot be distinguished by the experimental method. Potentially some CO_2 may be released from water depending on changes in/to carbonate equilibria, but this would have been limited by the low concentration of carbonate in solution at the high pH of the experiment.

Similar to many other leaching experiments, Marshall et al. (2011) used a batch method. They fitted an exponential function to their measured gas release rates. We note that the form of this function potentially suggests a diffusive release mechanism, with the release rate decreasing as the system evolved towards equilibrium. (This implies release of $^{14}CO_2$ gas due to changes in carbonate equilibria rather than desorption of CO gas).

Other authors have noted that leaching of C-14 may be diffusion controlled. For example, Gray and Morgan (1988) observed that for Hanford graphite cumulative C-14 release is linear with the square root of time, which suggests release is controlled by a diffusion mechanism. Conversely, Gray and Morgan (1989) found that this relationship was only true for one of three samples of French graphite. However, release rates were generally higher for the French graphite than the Hanford graphite, so potentially the

different release curves reflect differences in the contaminant distributions between different forms and hence the dominant release mechanisms.

Gray and Morgan (1988) show that the (aqueous) C-14 leach rate increases with increasing temperature for DIW, and Hanford groundwater, although the effect is less pronounced for the latter. This result is consistent with diffusion control since the rate of diffusion is expected to increase with temperature.

If leaching of C-14 is diffusion controlled (at least for some fraction of the labile C-14, e.g. that which is not located on the geometrical surfaces), for C-14 released as bicarbonate / carbonate ions, this implies that there must also be ion exchange with stable carbon because the total amount of carbon in solution should be constant at a fixed pH / pCO_2 (controlled by carbonate equilibria).

This alternative interpretation of diffusion controlled release would cast doubt on validity of applying release rates measured towards the end of leaching experiments to long-term post-closure release of C-14, because release rates could be overestimated or underestimated, depending on conditions in the GDF. It also implies that it might be possible to develop a simple mechanistic release model that could be directly incorporated into safety assessment calculations, for example a diffusive exchange release model. Such a release model might reduce uncertainty in C-14 release rates on post-closure timescales.

Evidence from leaching experiments indicates that C-14 will dominantly be leached from graphite as aqueous inorganic carbon ions (predominantly carbonate under high pH conditions). A small amount of C-14 may also be released in organic phases such as methanoic (formic) acid and oxalite generated through radiolysis (Grambow, 2010).

The majority of leaching experiments have been undertaken under oxic conditions. It is possible that under reducing conditions a higher proportion of C-14 could be released in organic form(s). However, French experiments have indicated that this is not the case (Grambow, B. 2012, pers. comm.). RWMD is currently undertaking experiments under reducing conditions.

In summary, a small fraction of the C-14 inventory is labile. Some of the labile C-14, potentially that associated with geometrical surfaces, is released rapidly upon contact with water, while the remainder of the labile fraction is released more slowly. This slower release process may be diffusion controlled, potentially involving ion exchange with stable carbon in the water. C-14 can also be released to gas, both under conditions of atmospheric storage, and in response to contact with water. The long-term release rates (to water and gas) and the fraction of the C-14 inventory that can be released are uncertain (on post-closure timescales). Assuming that the entire inventory is released is likely to be significantly cautious. If graphite wastes are cement encapsulated, the most

labile inorganic C-14 will tend to be incorporated in carbonate minerals. Similarly the most labile gas may be released on encapsulation. Therefore only a fraction of the labile inventory may be available for release post-closure.

2.3 Transport Behaviour of Radionuclides Released from Irradiated Graphite

Near-field

Cl-36 will be present as chloride ions in water. Many inorganic chloride compounds are highly soluble and not significantly chemically reactive, therefore Cl-36 is mobile in water. Transport will be by advection and/or diffusion, depending on the hydraulic properties of the Engineered Barrier Systems (EBS) and host rock, and hydraulic gradients. Some Cl-36 may also be released in organic form which may subsequently be hydrolysed to chloride.

C-14 exhibits complex behaviour partitioning between water and gas, inorganic and organic phases. Therefore C-14 can potentially be transported in water and gas. Partitioning and transport of C-14 will be consistent with the behaviour of bulk carbon in the GDF, in particular carbonate equilibria reactions and reaction of aqueous carbon species with cement minerals (cementitious EBS, overpacks, seals, etc as applicable to disposal concept). However deviations from the behaviour of bulk carbon may occur due to heterogeneous conditions in the vaults (this is discussed in Section 2.5).

Figure 2-2 illustrates the conceptual model of the behaviour of bulk carbon in the GDF, considering the types of materials that are likely to be present and the associated key reactions. The materials present, relevant reactions, and the magnitudes of fluxes will be concept specific. The key processes are:

- generation of CO_2 and CH_4 gas from microbial degradation of organic wastes by organotrophic bacteria;
- generation of H_2 gas from anaerobic corrosion of metals and radiolysis;
- generation of CH_4 gas by reduction of CO_2 with H_2 by autotrophic bacteria;
- reaction of cement minerals with CO_2 generated from degradation of organic wastes and bi/carbonate and other ions present in the host rock porewaters;
- dissolution of CH_4 gas in accordance with Henry's law; and

- partitioning of carbon between gaseous (CO_2), aqueous (HCO_3^-/CO_3^{2-}) and solid ($CaCO_3$) species, in response to carbonate equilibria and the reactions identified above.

Release of C-14 in CO gas, with dissolution in accordance with Henry's law, might also be important.

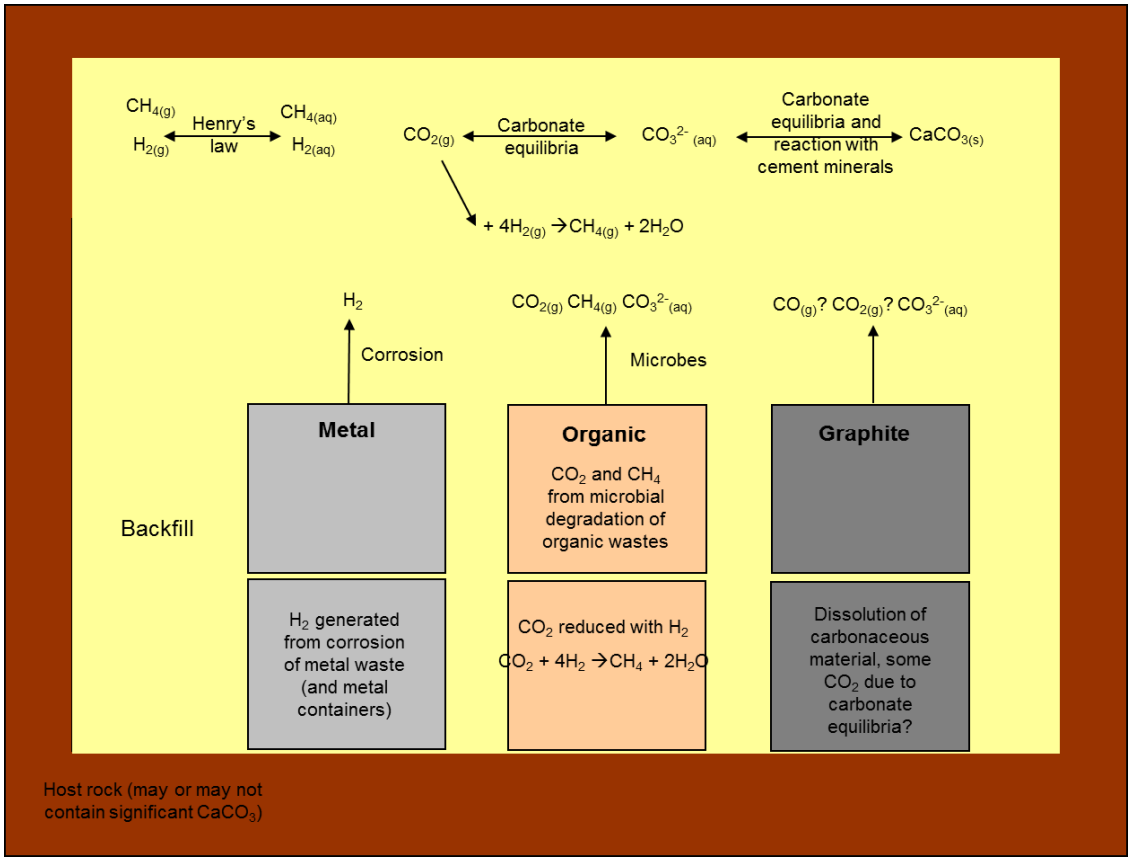

Figure 2-2. Conceptual model of the behaviour of bulk carbon in the GDF

Generation of methane gas by organotrophic bacteria is not expected to be a significant process for waste packages comprising graphite wastes cement grouted into steel or concrete containers, because there is very little material to support microbial processes. CO_2 (carbonate) can be reduced to methane by autotrophic bacteria using H_2 generated from anaerobic corrosion of metals. However, in recent work considering C-14 release from isolated packages of cement encapsulated graphite waste in steel containers, a group of experts considered that this process would not be significant (LLWR, 2012). This expectation is consistent with evidence from the French programme (Grambow, B., 2012, pers. comm; Andra, 2005c).

Generation of methane by autotrophic bacteria is known to be significant for mixed I/LLW containing organic wastes where methane generation by organotrophic bacteria is occurring. Therefore these methane generating processes may be important

where packages of graphite waste are co-disposed in the same vault as packages of I/LLW containing organic wastes (Section 3.2).

Evidence from leaching experiments (Section 2.2) indicates that C-14 will dominantly be leached from graphite as aqueous inorganic carbon ions (predominantly carbonate under high pH conditions). A small amount of C-14 may also be released in organic phases such as methanoic (formic) acid and oxalite generated through radiolysis (Grambow, 2010).

C-14 will therefore be incorporated into gaseous and aqueous phases by the processes described above. It is anticipated that C-14 will partition between gaseous, aqueous and solid phases in the same proportions as bulk carbon. This is termed a specific activity model. C-14 may be preferentially associated with the aqueous / solid phases rather than gaseous phases due to isotope fraction effects, but these will be minor and can be ignored (Appendix 2 in Jackson and Yates, 2011). Recent leaching experiments (Marshall et al., 2011) indicate that there may be a small direct release of C-14 from graphite as radiolabelled gas. This was discussed in Section 2.2, and includes the potential for release as CO. CO is unlikely to be hydrolysed, but it may be degraded radiolytically.

In summary, C-14 and Cl-36 will dominantly be present in the following chemical forms:

- $Cl^-_{(aq)}$
- HCO_3^- / $CO_3^{2-}{}_{(aq)}$ (predominantly the latter under high pH conditions)
- $CO_{2(g)}$
- $CO_{(g)}$
- $CH_{4(g)}$
- Dissolved $CH_{4(g)}$

Radionuclides can be transported by advection and / or diffusion in water (aqueous ions and dissolved gases), and in a free gas phase. C-14 may be transported in organic or inorganic forms. Cl-36 ($^{36}Cl^-_{(aq)}$) may be weakly chemically retarded through sorption onto minerals, or more strongly attenuated by co-precipitation in minerals such as Friedel's salt under certain geochemical conditions (Ondraf / Niras, 2009). C-14 ($^{14}CO_3^{2-}{}_{(aq)}$) may be attenuated by carbonate equilibria, ion exchange and co-precipitation in carbonate minerals at the carbonation front in a cementitious EBS. These mechanisms for chemical retardation of C-14 are illustrated in Figure 2-3 and further discussed in Section 3.1.1.

The minor component of aqueous C-14 that is released in organic form may not be attenuated by inorganic geochemical processes. However depending on the geochemical conditions, and hence available oxidants, these organic molecules may be metabolised to CO_2 and CH_4 (plus a small fraction retained in biomass) by microbial processes.

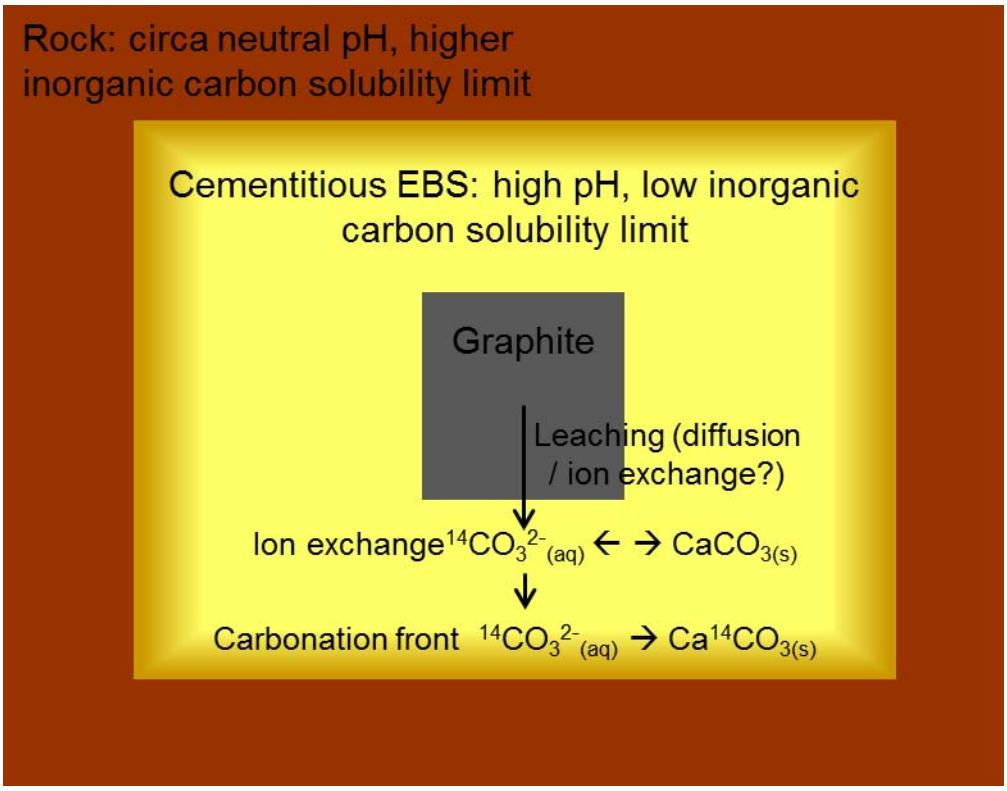

Figure 2-3. Conceptual model of chemical attenuation of C-14 in a cementitious near-field

Geosphere

Similar to the near-field, radionuclides can be transported by advection and / or diffusion in water (aqueous ions and dissolved gases), and in a free gas phase. C-14 may be transported in organic or inorganic forms. C-14 ($H^{14}CO_3^-{}_{(aq)}$) and Cl-36 ($^{36}Cl^-{}_{(aq)}$) ions are not expected to be significantly retarded by geochemical processes in the geosphere, unless for example, C-14 is co-precipitated in carbonate minerals in response to a reduction in the partial pressure of CO_2.

The behaviour of gas will significantly depend on the hydraulic properties of the host rock, and for very low permeability (gas tight) host rocks, the performance of tunnel seals and the Excavation Damaged Zone (EDZ). Transport in gas will be affected by the

total amount of bulk gas generated (from anaerobic corrosion of metals, degradation of organics and radiolysis, etc.), since bulk gas will act as a carrier for C-14 labelled gases; and migration of bulk and C-14 labelled gases will increase as bulk gas generation increases.

Migration of C-14 in gas will also significantly depend on:

- the biogeochemical conditions in the GDF, incorporation and partitioning of C-14 into the gas phase; and
- dissolution of gases in water in the GDF and geosphere.

For very low permeability (gas tight) host rocks, although there may be some migration of free gas into the host rock, the primary mechanism for migration of gas will be dissolution and subsequent diffusion (Andra, 2005a). So long as the diffusion pathlength is sufficiently long, C-14 in dissolved gas may decay to insignificant concentrations within the geosphere.

Gas can readily migrate through fractured host rocks, and the capacity for dissolution is low due to the low porosity. Dissolution may primarily occur in overlying permeable formations, where flow is primarily through the rock matrix porosity. Therefore gas can potentially migrate through the host rock and significantly dissolve in overlying near-surface geological formations. This may provide a 'short-circuit', bypassing the geosphere barrier.

Such short circuits might also be possible in very low permeability (gas tight) host rocks due to migration through backfilled shafts, drifts and their EDZs, depending on the performance of the seals and the EDZ. This potential pathway has been considered in safety assessments, e.g. Quintessa (2011). Similarly some conceptssuch as those for GDFs at shallower depths may include gas permeable drift seals to limit the peak gas pressure and prevent the risk of hydrofracking (Nagra, 2008).

Shallow relatively permeable formations will not be present at all sites, and in some instances low permeability formations may extend to the biosphere. Concepts considering shallow geological disposal of graphite in low permeability formations have been developed (e.g. Ozanam, 2009).

2.4 Guidance Level Barrier Performance

The disposability of irradiated graphite wastes is primarily dependent on the potential post-closure impacts of the associated radionuclides, noting that these impacts are influenced by the properties of the material itself (Section 2.1). In this study, the approach to examining the disposability of graphite is to test whether graphite can be

disposed in a wide range of systems, where a disposal system comprises the near-field and geosphere barriers.

In support of safety arguments and evidence, it is helpful to determine the properties of geosphere and near-field barriers in terms of providing containment, and to test the understanding of those properties against a quantitative metric of performance based upon impacts being of the order of regulatory guidance levels. If these required properties can easily be achieved, it can be concluded that graphite is generally suitable for geological disposal.

The relevant metric chosen for this study concerns the radionuclide flux to the biosphere that could give rise to impacts around regulatory guidance levels. In order to calculate this metric it is necessary to specify the assessment criteria, and to make some assumptions about the biosphere. The resultant metric is naturally subject to the assumptions underpinning the biosphere model, and alternative assumptions could result in a more or less stringent metric. For example, recent work undertaken by LLWR (2012) identified that their assessment model had very conservative parameterisation based on the results of the BIOPROTA international comparison of biosphere models (Limer et al., 2012). Further development of LLWR's assessment model led to two orders of magnitude reduction in the dose per unit $^{14}CH_4$ gas flux. Therefore, while this approach is consistent with exploring the range of suitable disposal systems for graphite wastes, the suitability of those systems approaching or slightly exceeding the metric will be uncertain.

Assessment Criteria

The performance metric we have chosen for this study is a target maximum annual[4] dose of 17 μSv. This equates to a conditional risk[5] of 1E-6 y^{-1} to a human receptor, and is based on the risk guidance level described in the UK regulatory guidance (Environment Agency and Northern Ireland Environment Agency, 2009; Environment Agency et al., 2009). This is not to say that a safety case for a system meeting this target would be accepted, but indicates that it could be possible to make a successful safety case, given demonstration of detailed system understanding including identification of key safety features / functions underpinning key safety arguments, and demonstration of optimisation.

[4]The annual dose is the sum of effective doses received in a year plus the sum of committed effective doses from intakes (ingestion and inhalation, within the same year). The committed doses are not all received in the year of intake, so the dose is not a dose per year or dose rate.

[5] This is the increment to lifetime risk that an individual will suffer a serious radiation-induced health effect accumulated via exposure over a year, as opposed to the risk of such a health effect occurring in any one year.

Other countries will have different assessment criteria, but an annual dose target of 17 μSv is anticipated to be appropriate to explore geological disposal of graphite in a way that is of interest to all participants in CARBOWASTE.

Biosphere Model

The RWMD generic biosphere model has been taken as representative of a typical biosphere modelling approach. The generic biosphere considers doses associated with:

- natural discharge of Cl-36 and C-14 in water to a generic terrestrial biosphere;
- consumption and use (e.g. for irrigation) of well water containing Cl-36 and C-14; and
- foodchain uptake of C-14 labelled gases in the soil zone.

For this assessment we have excluded doses associated with natural discharge to the biosphere. This is because the doses may be significantly influenced by the geography of the biosphere, e.g. discharges may be concentrated into incised river valleys, or more widely distributed across flat lowlands. We have also excluded doses associated with inhalation of C-14 labelled gases because the dose per unit flux is much smaller than the dose associated with foodchain uptake (Limer et al., 2010). Doses associated with use of well water (for drinking and irrigation) and foodchain uptake of C-14 labelled gases are less sensitive and more generic, and are therefore included.

The generic biosphere model is described by Thorne (2007), and has been used to calculate biosphere flux to dose conversion factors associated with the use of well water. Foodchain uptake of C-14 labelled gases in the soil zone is analysed using a separate model (the Enhanced RIMERS model) described by Thorne (2006). The BIOPROTA international model comparison has shown this to be a cautious model.

The biosphere model makes the assumption that the host rock, and overlying low permeability geological formations are overlain by relatively permeable shallow geological formations from which well water can be abstracted. An assumption is made regarding the amount of dilution in the shallow geological formations, but in reality this will vary as a function of the site specific GDF footprint and layout, direction and dominant mechanism for radionuclide transport through the geosphere, and the shallow hydrogeological environment.

Foodchain uptake of C-14 labelled methane gas is not independent of the discharge area to the biosphere. For methane gas to be taken up into the foodchain it needs to be metabolised into CO_2 by the microbial population in the soil. The microbial population

has a maximum capacity (per unit area) for metabolism of methane gas. Thus, the C-14 concentration in soil (and hence biomass) will be a function of the flux of C-14 labelled methane and the total bulk methane gas flux (which 'competes' with C-14 labelled methane to be metabolised).

For a bulk methane gas flux of 1E3 m^3 y^{-1} (at Standard Temperature and Pressure, STP), and a $^{14}CH_4$ flux of 1 TBq y^{-1}, Thorne (2006) calculated that that for releases to an area of 1E6 m^2 the fraction of the total methane flux which is metabolised by the microbial population in the soil will be less than 1 (best estimate 0.15). For smaller discharge areas, the metabolic capacity of the microbial population in the soil will be exceeded and the C-14 concentration in soil / biomass will be independent of the size of the area of release.

For areas of 1E7 m^2 and larger, the fraction of methane that is metabolised will increase to 1, as the mass flux of methane will be reduced and the capacity of the microbial system will not be exceeded. Thus for areas of 1E7 m^2 or greater, the concentration of C-14 in soil / biomass will decrease in proportion to the increase in the area of the discharge zone. This is summarised in Table 2-3.

We assume the gas discharge area will be similar to the footprint of the ILW/LLW area of the GDF which may be of the order 1 km^2 to several km^2, depending on the volume of graphite that is to be disposed, the host rock and hence GDF concept, and whether the discharge zone relates to graphite wastes only or all ILW/LLW (segregation Vs co-disposal). Therefore, it is assumed that all C-14 labelled gases enter the foodchain and there is no significant dilution of the C-14 concentration in soil / biomass due to a large discharge zone (Table 2-3). This may be a very conservative assumption for a GDF constructed in a gas tight host rock (e.g. indurated mudstone) since migration of gas is most likely to occur past shaft seals and therefore the discharge zone may be very localised and limit potential exposures.

Table 2-3: Concentration of C-14 in standing biomass for various release areas for a discharge of 1 TBq y^{-1} of $^{14}CH_4$and a bulk methane release rate of1E3 m^3 y^{-1} (at STP)

Area of Release, A (m^2)	Concentration in Standing Biomass (Bq kg[C]$^{-1}$)
1E4	5.2E4
1E5	5.2E4
1E6	5.2E4
1E7	3.5E4
1E8	3.5E3

2.4.1 Calculation of a Representative Metric for the Guidance Level Barrier Performance

The radionuclide flux to the biosphere that gives rise to impacts around regulatory guidance levels can be calculated from the dose criterion using the biosphere dose factor (Sv y^{-1}) per unit radionuclide flux to the biosphere (Bq y^{-1}). The flux-dose factor can be determined by undertaking biosphere model calculations for a unit input radionuclide flux (Bq y^{-1}).

It is also useful to define transfer rate metrics, for comparison against experimental results reported as rates (y^{-1}) and to provide a common basis for comparison of barrier performance. The limitation of transfer rate metrics is that they only apply to a specific radionuclide inventory.

We define two metrics:

$$\lambda_1 = d / ib$$

Where,

λ_1 is the transfer rate that gives rise to impacts around regulatory guidance levels (y^{-1})

d is the maximum acceptable dose (Sv y^{-1})

i is the radionuclide inventory (Bq)

b is the biosphere dose factor (Sv y^{-1}) per unit radionuclide flux to the biosphere (Bq y^{-1})

This metric is applicable where radionuclides are homogeneously distributed throughout a material, and are slowly being released from that material, for example to examine the barrier function of graphite wastes themselves based on experimental measurements of release rate.

λ_2 (y^{-1}) is applicable where radionuclides are being transported from a source through a barrier, subject to decay and dispersion. While λ_1 can simply be calculated analytically,λ_2 was derived from modelling, as described below.

For both the radionuclide flux and transfer rate metrics, it should be noted that the metrics only consider each radionuclide in turn, i.e. it is assumed that there are no doses from other radionuclides. It is also assumed that graphite is the only source of C-14 and Cl-36 in the GDF. These radionuclides may also be present in other wastes. Whilst relevant to a safety assessment for a GDF, these other sources are not relevant when exploring the suitability of graphite for geological disposal.

Metric Fluxes and Transfer Rates

Metric fluxes were calculated using RWMD's generic biosphere model for transport in water, and RWMD's enhanced RIMERS model for C-14 labelled methane (see above) and are reported in Table 2-4.

Transfer rates that give rise to impacts around regulatory guidance levels were calculated for the radionuclide inventory associated with UK graphite wastes classified as Shielded ILW (SILW). SILW includes the majority of the graphite waste (by volume and radionuclide inventory): 6.04E3 TBq C-14 and 25.23 TBq Cl-36 (Appendix A). (SILW is further described in Section 3.1.1).

λ_1 was calculated analytically. A simple compartment model was used to calculate λ_2. The compartment model was implemented using the GoldSim code and is described in Appendix B. Metric transfer rates are given in Table 2-4.

Table 2-4. Radionuclide fluxes and transfer rates (for the UK SILW graphite inventory) that give rise to impacts around regulatory guidance levels

Case	Max radionuclide flux (Bq y^{-1})	λ_1(y^{-1})	λ_2(y^{-1})
C-14 only in water	6.3E7	1E-8	7E-6
Cl-36 only in water	7.8E8	3E-5	4E-5
C-14 only in gas[1]	8.4E9 (5.7E9)	1E-6 (7E-7)	3E-5

[1] Assuming that all C-14 labelled gases enter the foodchain and there is no significant dilution of the C-14 concentration in soil / biomass due to a large discharge area. Values are given for a discharge area of 1E7 m^2, values in parentheses are for discharge area of 1E6 m^2 or less.

2.5 Safety Assessment Calculation Approaches

In the previous section it was stated that a quantitative metric is helpful to determine the properties of geosphere and near-field barriers that would limit impacts to around regulatory guidance levels. This section considers how the properties of geosphere and near-field barriers consistent with that metric can be calculated.

For a specific system with associated barrier properties, safety assessment calculations are used to calculate radionuclide fluxes to the biosphere. Since the metric for this study is the radionuclide flux to the biosphere that is consistent with regulatory

guidance levels, the calculations can be reversed in order to calculate the required barrier properties.

It is useful to consider the typical approaches to such calculations, and common key assumptions, to place the calculation results in context, when using them to support safety arguments and evidence. Not least, because the analysis presented in the subsequent sections demonstrates that the simplifications that might be made in typical performance assessment models may overestimate the rate of C-14 release from graphite, and underestimate the performance of the near-field chemical barrier for cementitious GDF concepts.

Safety assessment calculations may be undertaken using simplified compartmental models, more detailed coupled models, or analytical calculations. Waste management organisations may adopt differing calculation approaches depending on the specific characteristics of their wastes, GDF design and host geological environment, and therefore the key Features, Events and Processes (FEPs) and process couplings (phenomenology) that need to be included in the calculations.

Regardless of the detailed modelling approach or code, radionuclide release and transport models typically include: a function that represents radionuclide release from the waste; and solubility limits and sorption distribution coefficients that provide simplified representations of the effects of the controlling chemical reactions.

Limitations

For graphite wastes, typical approaches would include assuming that the entire radionuclide inventory is instantly released from graphite at the calculation start time, or to represent gradual release of the entire inventory using empirical rates from leaching experiments. Assuming that the entire radionuclide inventory is released may be very cautious, particularly for C-14. For some radionuclides, i.e. Cl-36, the empirical rates are sufficiently fast (for the fraction of the inventory that is 'available') that they are equivalent to instantaneous release on post-closure timescales, while for other radionuclides, i.e. C-14, it is assumed that leaching rates measured on timescales of months, to a few years, can be applied over much longer post-closure timescales.

For C-14 it may be appropriate to specify an initial higher leaching rate, followed by a lower long-term rate. However, for concepts which include cementitious encapsulation of the graphite, this initial higher release will occur upon encapsulation, and aqueous inorganic C-14 will likely be incorporated in calcite. Although disposal strategies differ in the order and scheduling of encapsulation, transport, emplacement and closure, this

high early release and incorporation of C-14 in calcite is likely to be complete before the start of the post-closure period.

An invariant long-term leach rate for inorganic C-14 may result in the modelled concentration of aqueous C-14 in the waste or EBS reaching the solubility limit, particularly for cementitious concepts where the solubility limit is low. Stable isotopes of carbon, C-12 and C-13, will also be present in the GDF. It is anticipated that C-14 released from graphite will isotopically exchange with C-12/13 in other phases. For a simple safety assessment model, the equilibrium C-14 concentration might be represented by an effective solubility limit that is less than the solubility limit for total inorganic carbon (IC). Further, as C-14 is leached from graphite and equilibrium ratios of C-12/C-13/C-14 are reached, it is anticipated that the C-14 leach rate will decrease.

Therefore, the assumption of instant C-14 release or an invariant long-term leaching rate will tend to overestimate release of C-14. If the presence of stable carbon is not accounted for, aqueous transport of C-14 will be overestimated. This is particularly important when modelling cementitious concepts because although the IC solubility limit is low in absolute terms, it is still significant in terms of the potential flux of C-14.

The impact of 'background' C-12/13 on C-14 solubility cannot simply be achieved by multiplying the IC solubility limit by the ratio C-14/Total C. This is because C is present in water, gas and many inorganic minerals and (where relevant) organic materials. The different mineral phases have different stabilities and kinetics, and simply applying the ratio C-14/Total C implies that C-14 is always homogenously distributed throughout all phases which is unrealistic, e.g. while C-14 might exchange with C-12/13 on the surfaces of $CaCO_3$ minerals grains, it will not exchange with C-12/13 in the middle of the grains.

It is therefore difficult to accurately model the distribution of C-14 without using detailed reactive transport models. For simple safety assessment calculations the cautious approach is to use the solubility limit of IC for C-14. The simple calculations presented in the subsequent sections of this report further explore this issue and demonstrate that using the solubility limit of IC for C-14 is very cautious.

3 Analysis of Normal Evolution Scenarios

In this section we analyse the properties of near-field and geosphere barriers, testing their individual and combined performance using simple models, to identify properties that would limit impacts to around regulatory guidance levels for the normal evolution scenario. The disposability of graphite is then discussed, considering whether these barrier properties can readily be achieved.

The analysis is undertaken in two stages. Firstly we consider a GDF only containing graphite wastes: this is the simplest possible system. We then consider the implications of co-disposal of graphite with other ILW. These are more complex systems and interactions between waste types may affect the performance of the GDF for graphite wastes (for example see the potential interactions illustrated in Figure 2-2).

For both graphite wastes disposed in isolation and co-disposal of graphite with other ILW, radionuclide transport in water and gas is considered. We consider the ability of the near-field and the geosphere barriers to achieve the guideline level performance on their own, before considering the combined system performance.

The properties of near-field and geosphere barriers that would limit impacts to around regulatory guidance levels are a function of the total radionuclide inventory. Radionuclide concentrations in the waste are also an important factor, for example in relation to solubility limitation of C-14 in the near-field, but also human intrusion and disruptive event scenarios (Section 4).

The suitability of graphite wastes for geological disposal is therefore most appropriately explored using a real inventory, which contains a large amount of graphite and a significant associated radionuclide inventory. If a large inventory can safely be disposed in a wide range of systems, then smaller inventories (both in terms of volume, and radionuclide inventory) will be even more readily disposed. Therefore, the assessment is undertaken for the UK national radionuclide inventory associated with graphite wastes (Appendix B).

3.1 Graphite Wastes Disposed in Isolation

3.1.1 Near-Field Barriers

Table 2-4 describes radionuclide fluxes and transfer rates for the UK SILW graphite inventory that give rise to impacts around regulatory guidance levels. While it is possible, for example, to calculate maximum water flow rates through the near-field

and then relate these flow rates to the barrier hydraulic properties, complexities arise because of issues such as solubility limitation, interactions between different near-field barriers, and the boundary conditions imposed by the geosphere. It was therefore considered that better insight would be gained by analysing an example concept in detail and extrapolating the findings to other concepts.

Example Near-Field

Hicks et al. (2008) provides an overview of different concepts for geological disposal of ILW that have been, or are being considered by various waste management organisations. These concepts may include multiple barriers, and these may be physical and / or chemical barriers.

We have chosen to analyse the UK ILW/LLW concept for the following reasons:

- information on design and number of waste packages, vaults, etc is available that is consistent with the UK inventory information;
- it is a cementitious concept, and therefore has a strong chemical barrier for inorganic C-14. It is therefore an appropriate concept for geological disposal of graphite and it enables some of the assessment issues identified in Section 2.5 to be explored; and
- at the current stage of the UK Managing Radioactive Waste Safely (MRWS) programme, possible candidate sites for the GDF have not been selected. Therefore RWMD are considering a range of potential host rocks that is consistent with exploring the suitability of graphite wastes for geological disposal.

In the UK ILW/LLW concept the graphite wastes are assumed to be encapsulated in stainless steel containers using cementitious encapsulant. The wastes are divided into those that require remote handling due to the high dose rates from the packages (Unshielded ILW, UILW), and those that can be handled using a stacker truck (Shielded ILW, SILW). Different containers are assumed for UILW (NDA 3 m^3 boxes) and SILW (NDA 4 m boxes), and the SILW containers can include concrete shielding if required to reduce the package surface dose rate. The focus is on SILW since this contains the majority of the graphite inventory by volume and activity (Appendix A).

The potential host rocks being considered by RWMD are:

- higher strength host rock (e.g. fractured crystalline rock);
- lower strength sedimentary host rocks (e.g. indurated mudstone); and

- evaporite host rock (e.g. anhydrite or halite).

The same waste packages are assumed for each host rock, however the GDF designs, vault dimensions, vault stack arrangements and backfilling / sealing strategies are different. For higher strength host rock, and lower strength sedimentary host rock, the waste packages will be surrounded with cementitious backfill in the vaults (Figure 3-1). For evaporite host rock, there will not be any backfill, since the void space will be closed by creep of the host rock. However, sacks of MgO may be placed in the vaults to absorb moisture and CO_2 gas.

The near-field barriers included in the UK ILW/LLW concept are the encapsulant, container and backfill. The wastes themselves may also be a barrier to radionuclide release. This captures the major near-field barriers of many disposal concepts, although naturally there are significant differences in geometry and material specifications between concepts. Features that are not included in the UK ILW/LLW concept, but are included in other concepts are (concrete) monolithic wasteforms and structural tunnel linings. However, the results of the analysis for the UK ILW/LLW concept are used to inform discussion of the barrier function of these other features.

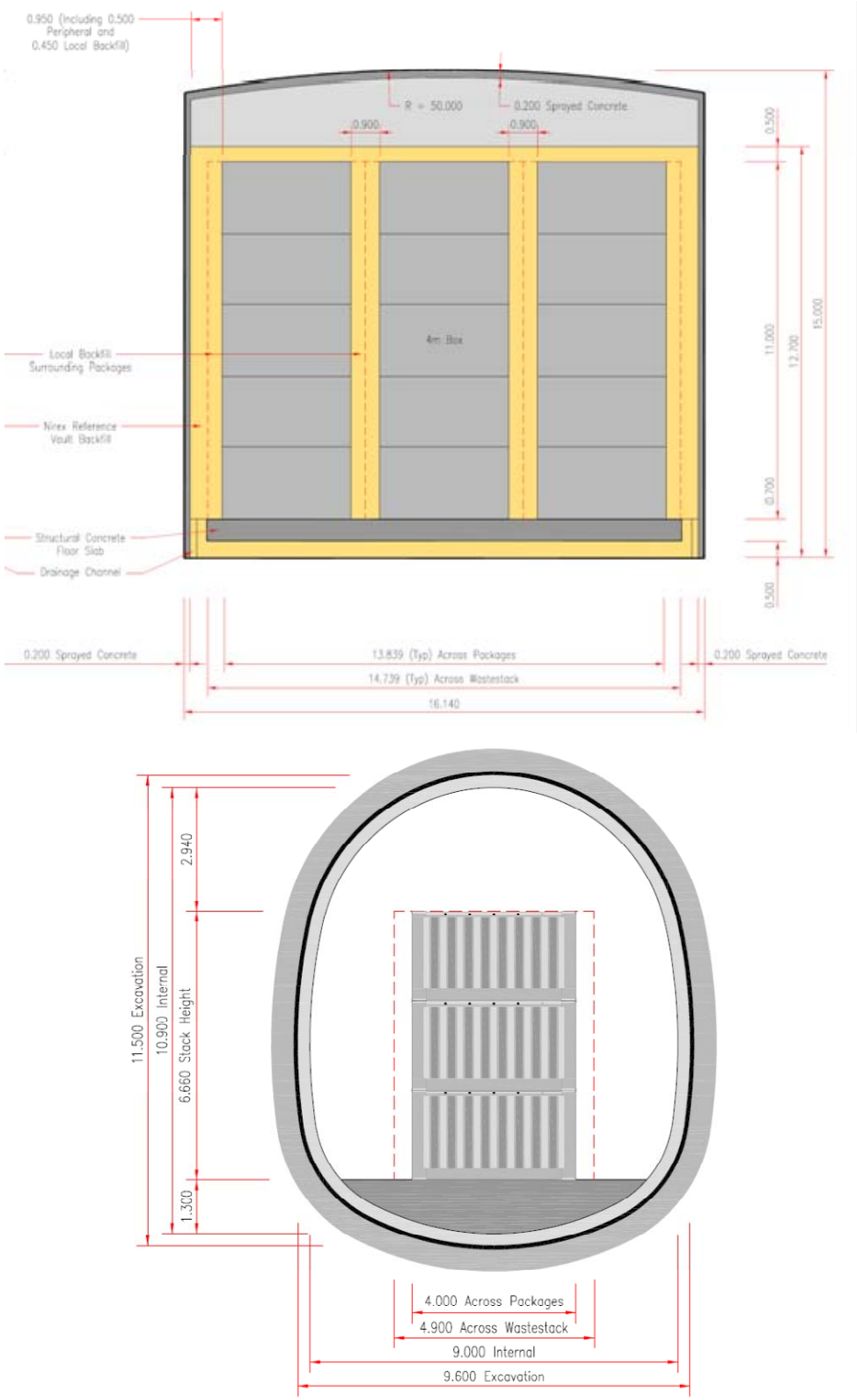

Figure 3-1. UK ILW/LLW concept for SILW in higher strength host rock (top) and lower strength sedimentary host rock (bottom)

Assessment Model for Transport in Water in the Near-Field

As described in the previous section, the approach to understanding the performance and interaction of the near-field barriers is to analyse an example concept in detail. It was decided to consider the UK ILW/LLW concept for the UK SILW graphite inventory, considering a range of different potential host rocks. To support the analysis a simple model has been developed to help analyse the performance of the different near-field barriers (Appendix C).

Calculations have been undertaken for two different example host rocks: higher strength host rock and lower strength sedimentary host rock. The results can be extrapolated to inform discussion of barrier performance in evaporite host rocks.

These simple models only consider transport in water, and assume that the near-field is fully saturated. This may not be true for lower strength sedimentary host rocks where gas may be trapped in the vaults. The near-field models will therefore tend to overestimate diffusive transport of radionuclides in water for these host rocks. Release of C-14 from the near-field in gas (dissolved and free gas) is discussed subsequently.

Transport in Water in the Near-Field

Waste

Radionuclide release from irradiated graphite has already been discussed in Section 2.2. For the calculations presented in the subsequent sections it is cautiously assumed that the entire inventory is available for release upon contact with water.

Cl-36 is rapidly leached from graphite upon contact with water, and as such the wasteform is not a significant barrier for Cl-36. C-14 is released more slowly. Table 3-1 compares the disposal system transfer rate to achieve guideline performance for C-14 transport in water and long-term release rates from leaching experiments. With the exception of relatively rapid early release of C-14, which in any case may be significantly incorporated into carbonate precipitates upon encapsulation, the waste itself may be an important barrier, and contribute to the overall disposal system performance.

However, in Section 2.2 we noted that the C-14 release rate measured in leaching experiments tends to decrease with increasing duration of the experiment. This may actually reflect batch experiments reaching equilibrium rather than the 'availability' of C-14 in the waste. Therefore the barrier function of the waste may be lower than

indicated by long-term batch leaching experiments, if the waste package does not isolate the waste to the extent required for equilibrium to be reached.

In reality, it is more likely that the encapsulant and container will act to isolate the wastes such that equilibrium conditions may be established in individual waste packages, and C-14 release from the waste might fall to very low rates. This demonstrates the need to analyse how the near-field barriers work in combination, but also that an improved understanding of mechanisms of radionuclide release from graphite might lead to more robust safety assessment calculations. However, this would only be necessary if there was doubt that the required performance could be provided by other near-field and geosphere barriers (analysed below), or if required to demonstrate that the system is optimised.

Table 3-1. Comparison of disposal system transfer rate to achieve guideline performance for C-14 transport in water and long-term release rates from leaching experiments

λ_1: Waste release transfer rate (y^{-1}) to achieve guideline performance for C-14 transport in water* (i.e. no near-field or geosphere barriers)	**λ_2: Disposal system transfer rate (y^{-1}) to achieve guideline performance for C-14 transport in water***	**Long-term release rates from leaching experiments (y^{-1})**
1E-8	7E-6	1.7E-02 to 1.83E-5

* For the UK SILW graphite inventory

Cementitious Encapsulant

Large blocks of waste may be surrounded by an annulus of encapsulant, while small pieces of waste may be intimately mixed with the encapsulant. Cementitious encapsulant contains free water. Therefore leaching of radionuclides from graphite will begin as soon as the wastes are encapsulated. C-14 can be incorporated into carbonate minerals, while Cl-36 will remain in the encapsulant pore water unless conditions are such that Cl-36 co-precipitates in minerals such as Friedel's salts (discussed below). (If the packaged wastes are kept in low humidity stores for a period of years or decades prior to transfer to the GDF, the packages may dry and radionuclides may be incorporated in meta-stable mineral phases. These meta-stable phases will re-dissolve as the package resaturates in the GDF).

The encapsulant may be a physical and / or chemical barrier. In the UK, wastes are encapsulated in containers using a two stage process. The first stage is to encapsulate the wastes using cementitious grout. Once the wastes have been encapsulated a second, more fluidic, capping grout is used to fill the container as close to the vent as possible, and minimise the ullage space in the container. Different grout formulations

are used for different wastes. Typical encapsulant porosities are of the order of 20% (Towler et al., 2012). Hydraulic conductivities have not been measured but are anticipated to be higher than for structural concrete, but lower than for high permeability cementitious vault backfill, such as Nirex Reference Vault Backfill (NRVB), which is designed to be a chemical barrier (Table 3-2).

It is not just the permeability of the encapsulant that is important, but also its interaction with the container. Standard RWMD container designs are fully sealed except for a gas vent on the top of the container. Such containers will prevent flow of water through the encapsulant until the containers are perforated by corrosion. There will only be diffusive transport of aqueous radionuclides through the container vent until the container is perforated.

Table 3-2. Hydraulic conductivity of 'concretes'

Media	**Porosity (-)**	**Hydraulic Conductivity (m s^{-1})**	**Reference**
Undegraded structural concrete	0.125	1E-10	Savage and Stenhouse (2002)
	0.15	1E-12	Hoch and Swift (2010)
Undegraded NRVB	0.55	1E-8	Towler and Bond (2011)

Cl-36 is highly soluble and only weakly sorbs onto cements (Table 3-3). Therefore the encapsulant may be a physical barrier for Cl-36, but only a weak chemical barrier. The sorption distribution coefficient for Cl-36 is sensitive to the total chloride concentration (i.e. Cl-36 plus background chloride) (Ondraf/Niras, 2009; Blin et al., 2010). Approximately linear, but different, relationships are observed between the sorption distribution coefficient and total chlorine concentration above and below a total chloride concentration of a few millimolal (Ondraf/Niras, 2009). At total chloride concentrations of a few millimolal Friedel's salts or calcium oxychlorides may form (Ondraf/Niras, 2009), and Cl-36 may be retarded through co-precipitation. A sorption distribution coefficient of 0.05 m^3 kg^{-1} has been selected for the illustrative calculations, which is consistent with a low background chloride concentration.

Table 3-3. Sorption distribution coefficient for Cl-36 onto cements

Media	Kd (m^3 kg^{-1})[1]	Reference
Fresh cement, pH controlled by portlandite (stage II)	Upper: 0.2 (0.01) Best estimate: 0.05 (0.001) Lower: 0.001 (0.0002)	Ondraf/Niras (2009) - recommended values based on extensive literature review

[1] Values are for total (Cl-36 plus background) chloride concentrations ≤ 1 millimolal. Values in parenthesis are for total chloride concentrations > 1 millimolal.

The behaviour of C-14 is more complex than that of Cl-36, since it can partition between water and gas, and chemically react with cementitious encapsulant. Therefore C-14 requires further consideration.

In a system containing only graphite, water and CO_2 gas, C-14 is expected to partition between water and gas according to carbonate equilibria. C-14 will partition in the same ratio as bulk carbon (C-12 and C-13) that is also present in the water and gas. This equilibrium will be modified by the presence of other minerals and solutes. In particular calcite and cement minerals. Dissolved CO_2 (present as carbonic acid, carbonate and bicarbonate ions) will react with cement minerals forming calcite, e.g.:

$Ca(OH)_2$ (Portlandite) + H_2CO_3 = $CaCO_3$ + $2H_2O$

Incorporation of C-14 in calcite will act as a significant chemical barrier. This process will not remove all carbon (including C-14) in water and gas, since calcite will be in equilibrium with carbonate /bicarbonate ions and CO_2 gas. However, the equilibrium concentrations in the presence of cement are much lower than would be anticipated for natural groundwater conditions. Therefore the fluxes of C-14 in water and gas will be reduced. Table 3-4 compares the compositions of a range of natural groundwaters, and how they are modified by the presence of cement minerals (based on PHREEQC calculations). The precise composition will be affected by the cement formulation.

Table 3-4. Groundwater compositions with and without the presence of cement (Towler et al., 2010)

Water	Groundwater composition	Composition in the presence of cement
	Dissolved IC (mol/l)	Dissolved IC (mol/l)
Sellafield BH9B, SPFT3	9.5E-3	8.61E-6
Dounreay BH1, DET8	1.73E-4	8.67E-6
Sellafield BH3, DET1	1.07E-2	3.13E-5
Ocean Standard Water	4.65E-4	1.03E-5

Sorption distribution coefficients are available for C-14. These data are from batch experiments and show that C-14 is very strongly sorbed. However, from the preceding discussion it is clear that this apparent sorption may actually reflect carbonate equilibria and reaction with cement minerals. Adsorption of CO_2 onto the surface of cement minerals and processes such as ion-exchange may occur, but are only likely to be of secondary importance.

Performance assessment calculations (and associated models) normally assume linear, equilibrium (i.e. reversible) sorption, based on the assumption that processes such as adsorption and ion-exchange, dominate. Therefore, application of 'standard' sorption distribution coefficients is inappropriate for C-14, unless the distribution coefficients only reflect ion-exchange and other reversible processes.

Solubility limits can be calculated for inorganic carbon under disposal facility conditions including the presence of cements (Table 3-4). However, as discussed in Section 2.5, it is not simple to calculate an effective solubility limit for C-14 that takes into account equilibrium with background C-12/13 that is present in gas, aqueous ions, and associated with cement minerals, organic[6] and other wastes, and the host rock. This is because carbon is distributed between different phases and minerals, and different solid phases will have different solubilities, and due to reaction kinetics. For example C-14 would only interact with a small fraction of the solid minerals present (i.e. mineral surfaces).

The key process controlling the concentration of inorganic C-14 in water is isotope exchange with stable carbon present in solid minerals. Retardation of C-14 by isotope exchange is explored by Small et al. in Appendix 2 of Jackson and Yates (2011).Small et al. proposed a model for representing isotope exchange as a linear reversible sorption process. This would enable isotope change to be included in safety assessment calculations using standard modelling tools and approaches. However, data were not available to calculate the fraction of the mineral inventory C-14 could interact with and therefore Small et al. could not parameterise the model. Andra (2005b) proposed the same model as Small et al., but Andra have further developed the model to account for the amount of carbonate that C-14 can interact with:

$$R_d = \frac{\alpha A_{CO3}}{[CO_3^{2-}]}$$

Where

R_d is the distribution coefficient ($m^3\,kg^{-1}$)

[6] Organic wastes can be microbially degraded forming inorganic carbon (CO_2).

A_{CO3} is the concentration of carbonate in the cementitious encapsulant / backfill (mol kg^{-1})

$[CO_3^{2-}]$ is the concentration of carbon in solution (mol m^{-3})

α is an accessibility factor (-)

$\alpha \approx 0.17.(1 - \log \Phi)$

Where

Φ is the diameter of the calcareous aggregates in the cement (m).

Andra (2005b) calculate R_d values for a range of different concrete formulations and degradation states. R_d values were generally in the range 0.1 to 1 m^3 kg^{-1}.

The encapsulant will degrade over time, in response to reaction with CO_2, sulphate, etc in the water from the host rock, and from the wastes. The lifetime of the encapsulant as a physico-chemical barrier will be affected by the choice of backfill, for example cementitious backfill will act to condition inflowing water and therefore extend the lifetime of cementitious encapsulant. The container will also offer protection against solutes from the rock that would react with, and degrade, the encapsulant.

As the encapsulant degrades, it is anticipated that the porosity and permeability will increase with time (Table 3-5). However, if carbonation is the dominant process, and the container prevents significant leaching of solutes from the encapsulant, then the porosity and permeability could actually decrease with time if pores become occluded with calcite.

Table 3-5. Properties of fresh and degraded cement (Savage and Stenhouse, 2002)

	Porosity (-)			**Hydraulic Conductivity (m s^{-1})**		
Evolution Stage	**I**	**II**	**III**	**I**	**II**	**III**
Porous concrete	0.5	0.5	0.6	1E-8	1E-8	1E-8
Structural concrete	0.125	0.125	0.25	1E-10	1E-9	1E-8

As the encapsulant degrades the chemical barrier will also weaken. Eventually, all the cement minerals will have degraded and the carbonate equilibria will return to the conditions that would be expected in the absence of cement. Some of the carbonate precipitates may dissolve, releasing C-14.

The timescales for degradation of the chemical barrier are uncertain. However, for the UK ILW/LLW concept, even in a more permeable host rock the chemical barrier

would be maintained for several hundred thousand years given the total cement inventory in the encapsulant and backfill (NDA RWMD, 2010a), i.e. much greater than ten C-14 half-lives. Banford et al. (2012) reached similar conclusions for a cement barrier in a diffusive system, even though the barrier was only 20 cm thick. Therefore degradation of the barrier, and release of C-14 from carbonate precipitates, is not likely to be important because the backfill will condition water flowing into the GDF, and therefore help to 'protect' the encapsulant from alteration, and the backfill will also act as a chemical barrier (see below).

There may be cracks in cementitious encapsulant, and there could be preferential fluid flow (and hence also contaminant migration) though these cracks. Alteration of the crack surfaces (armouring) may limit the chemical barrier function of the encapsulant. Interfaces between the waste and the encapsulant and between the encapsulant and the container may have similar effects. However, the effects will likely be limited while the container is intact and advective flow of water through the encapsulant is not possible. Cracks may be more significant once the container has been perforated. The effects of cracks and armouring are further discussed below, in the context of the backfill.

Container

The previous discussion has highlighted the role of the containers as a physical barrier to radionuclide transport, but also the role of the container in protecting the encapsulant and maintaining the barrier performance of the encapsulant.

In the UK ILW/LLW concept, the stainless steel construction of the containers, and the anticipated low corrosion rates under high pH conditions in the disposal facility, means that the containers may function as a barrier for a significant period of time. The anticipated corrosion behaviour and potential lifetime (excluding mechanical effects) are described by Towler et al. (2010), and may be summarised as follows.

Initially conditions in the vault will be oxic and pit corrosion of metal containers could occur. It might be important to ensure that the container is not punctured by pit corrosion during the oxic phase since this introduces the potential for advective flow though the container. As conditions in the disposal facility become anoxic, the corrosion mechanism is anticipated to change from pit corrosion to uniform corrosion.

Watson et al. (2012) identified that it might take several decades for anoxic conditions to develop in SILW vaults. The timescales could be even longer in vaults containing only packages of graphite wastes because the chemical oxygen demand will be lower (due to the lower total quantities of metals, and the absence of the small quantities of reactive metals and organics present in other SILW wastes). This increases the potential

extent of pit corrosion of the containers during the early post-closure phase. However, Towler et al. (2010) calculated that the amount of oxygen trapped in the vault at closure would be small; ~3 mol per m^2 of metal container surface. Therefore, although pit and crevice corrosion might occur during the early oxic phase, the extent of perforation would be limited.

For uniform corrosion, the corrosion time can be estimated from the container thickness and corrosion rate under the prevailing chemical conditions. Some examples are provided in Table 3-6. It should be noted that these container corrosion times do not consider the potential for galvanic corrosion; or breaching of the containers due to mechanical loading and expansion stresses (i.e. formation of high specific volume minerals such as metal corrosion products, or ettringite in cementitious encapsulant), especially as the containers weaken through corrosion.

Initial, bounding scenarios of GDF mechanical evolution in a higher strength host rock are described by Towler et al. (2012) for the UK context. Considering the potential for galvanic corrosion, swelling stresses and mechanical evolution scenarios, for the full range of L/ILW wastes that might disposed in a GDF, container lifetimes could range from several thousand years to the times given in Table 3-6. For graphite wastes where galvanic coupling is not significant, and expansion stresses are limited to those arising due to alteration of cementitious encapsulant and surrounding backfill, container lifetimes may be substantial.

In contrast Andra (2005a) considered that high integrity concrete disposal containers would provide a barrier function for 10,000 y, but their 2009 assessment was more pessimistic. Container disruption (cracking) is initiated by failure of the concrete vault liner, resulting in transfer of lithostatic load onto the containers. Steel containers might be expected to be similarly mechanically disrupted.

Mechanical disruption might occur after tens to a few hundred years in an evaporite host rock in response to creep and lithostatic loading.

Table 3-6. Estimated stainless steel container corrosion times under high pH conditions

Type	Thickness (m)	Corrosion Rate (m y^{-1})	Corrosion Time (y)*
RWMD 4 m box	3E-3	1E-8	150,000
RWMD 3 m^3 box	6E-3	1E-8	300,000

* Uniform corrosion from outside and inside container.

In the next sub-section we examine the performance of the waste package as a whole.

Waste package (waste + cementitious encapsulant + container)

The simple model described in Appendix C was used to explore the performance of the waste package as a barrier. The release and geochemical behaviour of C-14 and Cl-36 were represented as follows:

- the entire radionuclide inventory is available to be released at the rates given in Table 2-1;
- the total inorganic carbon solubility limit is applied for C-14, therefore the rate of release of C-14 does not slow if the C-12/13/14 ratio approaches equilibrium;
- there is no ion-exchange with solid mineral phases; and
- Cl-36 is not solubility limited but can take part in sorption reactions (K_d = 0.05 m^3 kg^{-1}).

Retardation of C-14 by ion exchange was considered as a variant calculation assuming a K_d of 0.1 m^3 kg^{-1}(and no anion exclusion). Co-precipitation of C-14 at the carbonation front was not considered because the carbonation front is assumed to migrate from the backfill-rock interface, and is located within the backfill.

The barrier function of the waste package has been calculated from the ratio of the modelled aqueous radionuclide fluxes out of the waste package to the amounts of the radionuclides in the encapsulant (Figure 3-2 and Figure 3-3). The calculations have been undertaken considering the presence and absence of the container. With the container present there is only diffusive transport through the container vent until the container is perforated. Once the container has been perforated advective transport and radial diffusion out of the encapsulant is possible. With no container, advective transport and radial diffusion out of the encapsulant are always possible.

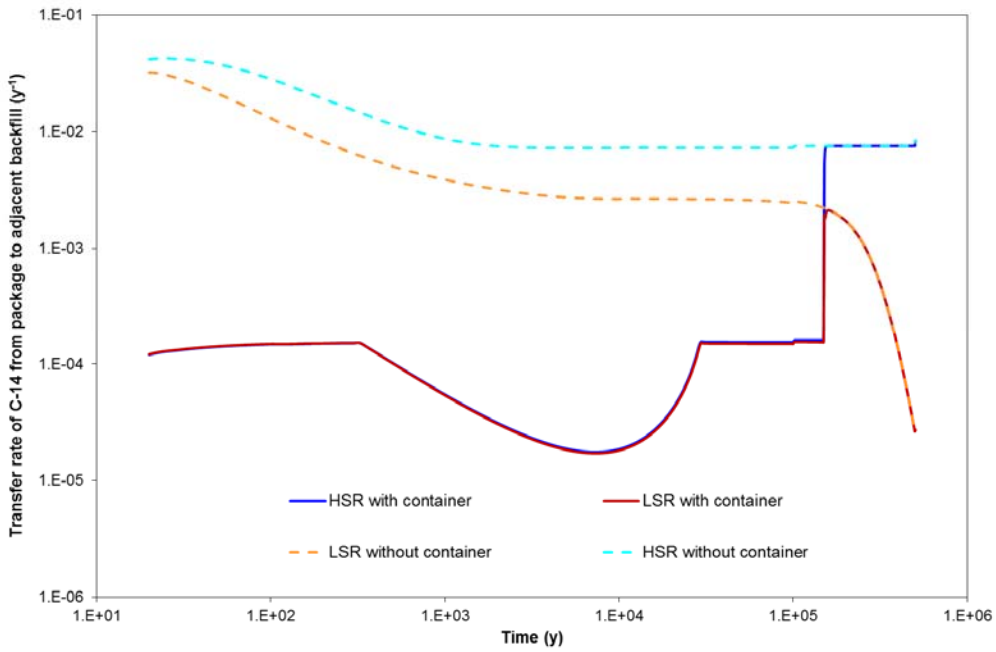

Figure 3-2.Barrier function of the waste package for release of inorganic C-14 in water from packages of UK SILW graphite. For cases with the container, the container is completely perforated by uniform corrosion at 150,000 y. (HSR = Higher Strength Host Rock, LSR = Lower Strength Sedimentary Host Rock).

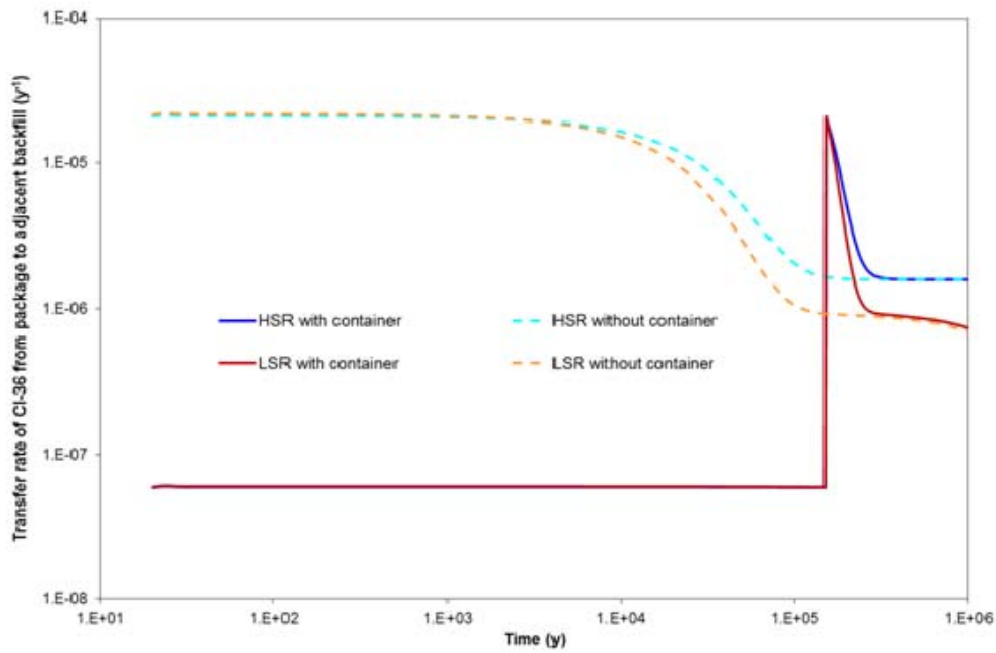

Figure 3-3.Barrier function of the waste package for release of Cl-36 in water from packages of UK SILW graphite. For cases with the container, the container is completely perforated by uniform corrosion at 150,000 y. (HSR = Higher Strength Host Rock, LSR = Lower Strength Sedimentary Host Rock).

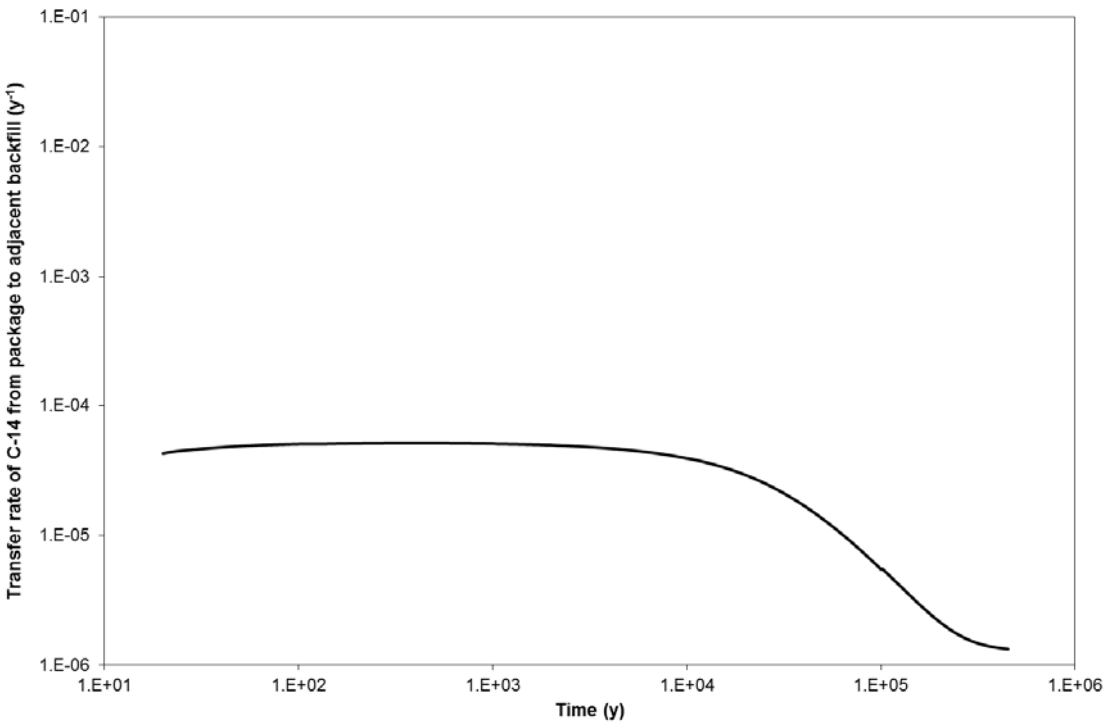

Figure 3-4.Barrier function of the waste package for release of inorganic C-14 in water from packages of UK SILW graphite. Higher strength host rock, excluding the container, with retardation through ion exchange (K_d= 0.1 m^3 kg^{-1}).

The barrier function provided by the container is significant. For Cl-36 there is a 'build up' of dissolved radionuclides in the container, with a spike in the transfer rate when the container is perforated by uniform corrosion (at 150,000 y). Although there is a corresponding spike in the C-14 transfer rate at 150,000 y, the amount of C-14 remaining in the package at 150,000 y is a small fraction of the initial inventory due to migration and, in particular, radioactive decay.

Dissolved C-14 concentrations reach the solubility limit for inorganic carbon inside the container because the graphite release model (specified long-term release rate of 1.83E-5 y^{-1}) does not consider the potential decrease in the release rate as the C-14 / bulk C concentrations reach equilibrium. Therefore, the C-14 release rate is likely to be overestimated. The period for which C-14 release is solubility limited is indicated by the 'dip' in the transfer rate from ~300 y to ~30,000 y for the "with container" results: the flux remains constant (solubility limited), but the amount of C-14 available continues to increase, so the transfer rate decreases.

It is useful to note that the model includes the backfill and that C-14 concentrations in the backfill are much less than the inorganic carbon solubility limit. Similarly, the

inorganic carbon solubility limit is not reached in the encapsulant for cases with no container because C-14 is more readily able to migrate into the backfill (see specific discussion below for more details on backfill properties).

The case where the container is not considered has been re-run for higher strength host rock, and including ion exchange of C-14 in the encapsulant. Despite the limited thickness of the encapsulant, ion exchange has a significant impact on the barrier function (Figure 3-4).

The model results indicate that for Cl-36 the encapsulant on its own is sufficient to achieve containment consistent with the performance metric because the water flow rate through the encapsulant is small and Cl-36 is considered to be sorbed onto the encapsulant (Table 3-3). The performance for Cl-36 is sensitive to the amount of sorption that is assumed. The container improves physical containment of Cl-36 until it is perforated by corrosion, but the container lifetime is less than one half-life for Cl-36, so it does not significantly reduce the peak flux.

For C-14 the waste package (waste, encapsulant and container) is not sufficient to achieve the performance metric, except for the case where the container is represented and ion exchange is also considered (results not shown). However, this result is sensitive to the longevity of the container.

We have not quantitatively analysed concepts where a number of waste packages are grouted into (high integrity) concrete disposal containers, e.g. Andra (2005a) and Nagra (2008). However, based on the above analysis, we would expect the performance for graphite wastes to be very good due to the low permeability of the disposal container, and the chemical barrier provided by the disposal container and internal cementitiousgrout. If a concept with concrete disposal containers was used in a higher strength host rock in which there was advective water flow in fractures in the rock, a potential performance issue would be cracking of the concrete disposal containers in response to lithostatic loading. However, both the Andra and Nagra concepts are for a lower strength sedimentary host rock, where transport is diffusion dominated. Thus, the presence of cracks might not significantly reduce the barrier function of the disposal container, although cautiously Andra consider no barrier function once the container is cracked.

Backfill

The UK ILW/LLW concept assumes different types of cementitious backfill for higher strength host rock, and lower strength sedimentary host rock. For higher strength host rock, a non-structural, high porosity backfill (NRVB) is designed to form a chemical, but not a physical barrier. There is the potential for mechanical movement of blocks of host rock into the vault, resulting in compaction of the backfill. This could initially lead

to increased fracturing of the backfill, and a reduction in barrier performance. However, upon further compaction, the backfill barrier performance would likely recover, and potentially improve compared with the initial condition (Towler et al., 2012).

For lower strength sedimentary host rock, it is envisaged that a structural backfill such as Nagra structural mortar (Nagra, 2008) would be used. The structural function of the backfill is required to support the rock, and promote self-healing of the EDZ. Without support, there would be further development of the EDZ as rock moves into the vaults, and this might provide a pathway for migration of fluids (i.e. water and gas) and associated radionuclides. The structural backfill is also designed to be a chemical barrier, but has lower porosity than NRVB. Structural backfill may act as a physical barrier, but in any case transport through this backfill will be by diffusion due to the very low permeability of the surrounding rock.

The simple model described in Appendix C, has also been used to examine the barrier function of the backfill for the Disposal System Safety Case (DSSC) illustrative concept examples (Figure 3-1). The barrier function of the cementitious backfill has been examined by calculating the radionuclide transfer rates out of the backfill for comparison with the performance metric. The transfer rate has been calculated from the radionuclide fluxes out of the backfill compared with the amounts in the backfill (Figure 3-5).

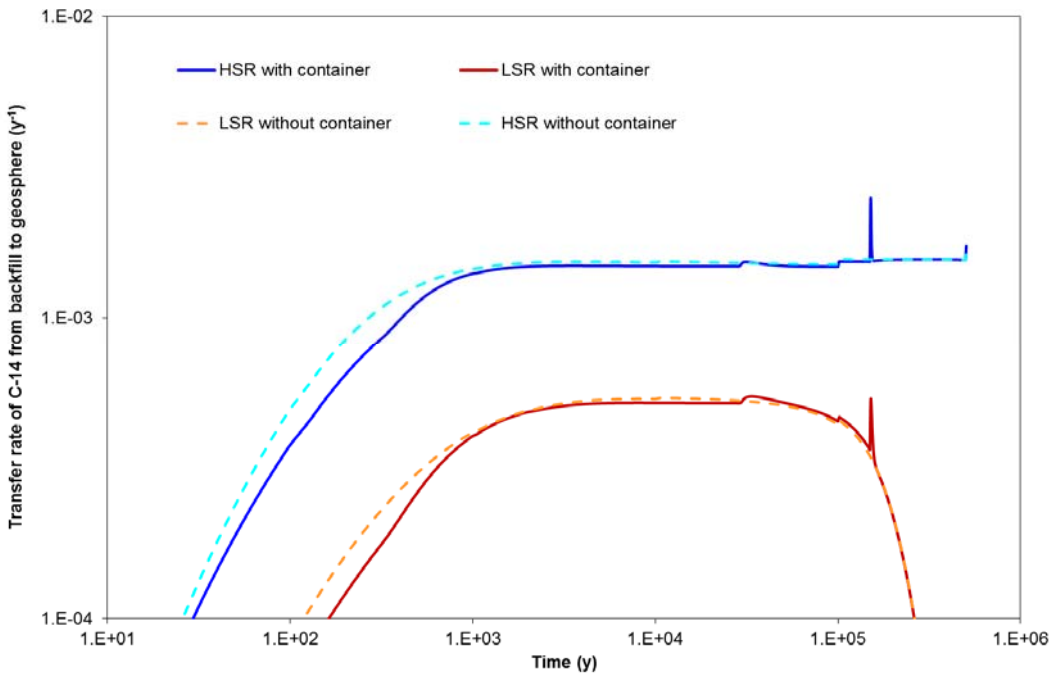

Figure 3-5.Barrier function of cementitious backfill for release of inorganic C-14 in water from packages of UK SILW graphite. For cases with the container, the container is completely perforated by uniform corrosion at 150,000 y.

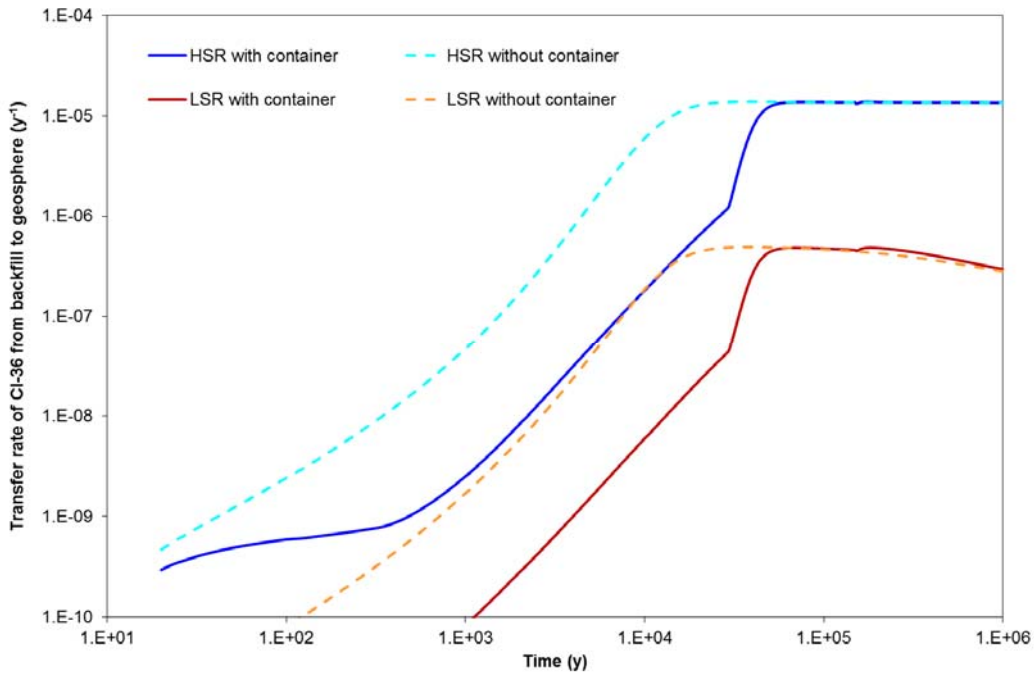

Figure 3-6.Barrier function of cementitious backfill for release of Cl-36 in water from packages of UK SILW graphite. For cases with the container, the container is completely perforated by uniform corrosion at 150,000 y.

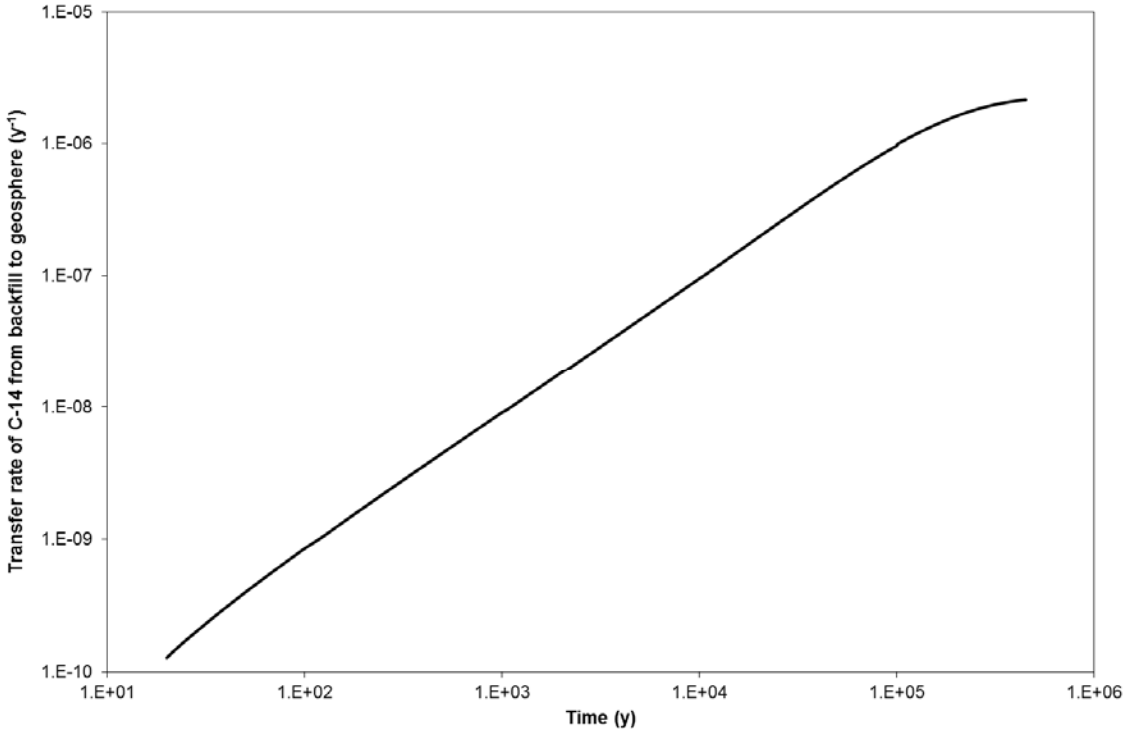

Figure 3-7.Barrier function of cementitious backfill for release of inorganic C-14 in water from packages of UK SILW graphite. Higher strength host rock, excluding the container, with retardation through ion exchange (K_d= 0.1 m^3 kg^{-1}).

The calculations indicate that in higher strength host rock the backfill is only a weak barrier due to advective flow of water through the barrier, and because the chemical barrier effect has already been provided by the encapsulant (i.e. solubility limitation), which is assumed not to degrade chemically in these calculations. However, in reality the backfill has an important role: to protect the encapsulant from chemical degradation by conditioning water flowing into the vaults, and to provide a second chemical barrier that will become important as the barrier function of the waste package decreases in response to chemical and mechanical processes.

The backfill is a stronger barrier in lower strength sedimentary host rock, however the smaller vault dimensions compared with higher strength host rock, and consequently greater number of vaults, means that there is more surface area for diffusive transport compared with higher strength host rock. The backfill is not sufficient to provide the required barrier performance for C-14, but is sufficient to provide the required barrier performance for Cl-36 due to slow diffusion through the backfill, but also because there is some sorption onto the backfill (Table 3-3). The performance of the backfill for Cl-36 is sensitive to the distribution coefficient.

The calculations likely underestimate the effect of the chemical barrier provided by the backfill because two important processes are not included in the calculations. The first of these concerns isotope exchange between C-14 in the water and stable C in the backfill minerals, as the 'C-14 front' migrates through the backfill. Figure 3-7 shows that assuming C-14 is able to interact with all the backfill (i.e. the effects of transport in fractures are not significant – see below), accounting for ion exchange using the model described previously, the backfill alone is able to provide the required barrier performance for C-14.

Secondly, a potentially very important retardation mechanism, which is largely irreversible over the timescales relevant to C-14, is co-precipitation of C-14 in calcite at the backfill carbonation front (Figure 2-3). For example calculations presented in Banford et al. (2012) show that c.99.5% of inorganic C-14 released from graphite may be retained at the carbonation front.

The presence of fractures in the backfill, and chemical alteration (armouring) of fracture surfaces, could significantly reduce the chemical barrier performance particularly where advective flow through the backfill is possible (i.e. a higher strength host rock environment). Interfaces between the containers and the backfill may have similar effects. Towler et al. (2012) considers the effects of backfill fracturing for bounding cases of vault mechanical evolution. The available evidence indicates that armouring has the greatest potential to be significant where: flow rates are relatively high; the number of fractures (and hence total fracture surface area) is low; and generation of voids in the backfill is possible (e.g. following failure of waste containers if the wastes are not encapsulated). This is an area of ongoing research.

Instead of structural backfill, in their design for a deep disposal facility, Andra proposes using structural concrete tunnel liners, which form stillages into which concrete disposal containers can be placed. Andra anticipates that the liner will be cracked by lithostaticloading, however the mechanical performance of the liner will be sufficient to allow the EDZ to heal (Andra, 2005a). Although the liner will crack, due to the low permeability host rock, radionuclide transport through the liner will be by diffusion. The presence of cracks will only have a limited impact on the barrier function of the liner: although radionuclides may diffuse more rapidly through the cracks than through the liner, the area of the cracks is very small compared with the area of the liner. Also diffusion through the rock at the end of the cracks will be slow, thereby limiting the rate of diffusion through the cracks. However, cautiously Andra (2005a) assumed that the liner is not a barrier to diffusion, i.e. the effective diffusivity was set to that for diffusion in water.

The structural concrete lining proposed by Andra will provide a similar chemical barrier function to structural cementitious backfill, and it will act to protect the

monolithic disposal containers from chemical degradation. However, due to the low solute flux from the rock into the vault in lower strength sedimentary host rock, this function is less important than in higher strength host rock.

Sum of Near-field Barriers

The simple model can also be used to calculate the barrier function for the entire near-field (Table 3-7). In the context of radionuclide transport in water, the near-field alone could potentially provide the guideline barrier performance for geological disposal of the UK inventory of irradiated graphite wastes. However, the performance for Cl-36 is sensitive to the amount of sorption, and the guideline barrier performance is for C-14 is only achieved if the model representation of the chemical barrier is not overly cautious.

Table 3-7. Total near-field barrier performance for transport in water

Transfer Rate (y^{-1})	**C-14**	**Cl-36**
Performance metric (λ_2)	7E-6	4E-5
Higher strength host rock including container	4E-5*	2E-6
Higher strength host rock excluding container, including ion exchange	1E-6	N/A
Lower strength sedimentary host rock including container	3E-5*	1E-7
Flux (Bq y^{-1})		
Performance metric	6.3E7	7.8E8
Higher strength host rock including container	4.7E9	1.5E7
Higher strength host rock excluding container, including ion exchange	9.7E6	N/A
Lower strength sedimentary host rock including container	4.0E9	7.6E5

*Results are very similar because the limiting factor is solubility limitation in the waste package (see Figure 3-9).

Transport in Gas in the Near-Field

C-14 can potentially be released from the vaults as C-14 labelled CO, CO_2, or CH_4 gas. The Phase 1 work demonstrated that the potential flux of CO_2 gas (and hence $^{14}CO_2$) is low due to reaction with the cementitious encapsulant and backfill, but also microbial reduction of CO_2 to CH_4 if methanogenic conditions are established and if sufficient

hydrogen is available, e.g. from anaerobic corrosion of steels. Therefore generation and migration of C-14 labelled methane is the key process.

Watson et al. (2012) modelled gas generation in SILW vaults containing graphite and other wastes. The gas generation model used by Watson et al. (2012) was more detailed than the simple gas generation model used in the Phase 1 work. The detailed model results indicated that very little methane would be generated in SILW vaults containing graphite and other SILW, and methane generation would not start until 3,500 y post-closure.The detailed model used by Watson et al. (2012) also demonstrated that if methanogenic conditions are established, then a significant proportion of C-14 labelled CO_2 can be reduced to CH_4 gas, noting the competing reaction of CO_2 with cement minerals.

If only packages of graphite wastes were disposed in the vault, then it is unlikely that there would be significant methane generation. Therefore the major C-14 gas flux would probably be direct release of C-14 labelled methane from the graphite wastes (Table 2-2). This flux would likely not exceed the required disposal system performance (λ_2) for transport in gas unless bulk gas containing C-14 methane was able to rapidly migrate through the near-field and geosphere to be biosphere (Table 3-8).

Table 3-8. Direct release rate of $^{14}CH_4$ from graphite compared with performance metric

	Rate (y^{-1})
Direct release rate of $^{14}CH_4$ from graphite	1E-5
λ_1: Waste release transfer rate (y^{-1}) to achieve guideline performance for C-14 transport in gas(i.e. no near-field or geosphere barriers)	1E-6
λ_2: Disposal system transfer rate (y^{-1}) to achieve guideline performance for C-14 transport in gas	3E-5

The total gas generation rate would also be low in a vault containing only packages of graphite waste, with generation of hydrogen from anaerobic corrosion of steel being the major source. Therefore, there would be limited bulk gas to act as a 'carrier' for C-14 labelled methane, and it is anticipated that much of the gas, including C-14 labelled methane will dissolve in water.

Gas-water interaction, and dissolution of gas, will occur as gas rises buoyantly through the near-field and geosphere. Migration of gas will be significantly influenced by the properties of the host rock, and potentially also the near-field barriers. The cementitious encapsulant, vented containers and cementitious backfill included in the

UK ILW/LLW concept are all gas permeable. Therefore, release of gas from the near-field will be controlled by the properties of the rock.

If the rock is fractured, e.g. a higher strength host rock, gas will readily migrate out of the near-field and through the geosphere. If the rock is of very low permeability and unfractured, e.g. lower strength sedimentary host rock, gas will be trapped in the near-field and pressurisation can occur. The rock gas entry pressure will control migration of gas from the near-field into the geosphere. Gas (and water, driven by the gas pressure) may preferentially migrate through the EDZ and tunnel seals rather than the rock. This is discussed in the next section. Migration of gas through the geosphere is discussed in Section 3.1.2.

For other concepts the near-field EBS may act as a physical barrier, limiting migration of gas into the geosphere, for example a clay backfill. Structural concrete linings of the type proposed by Andra (2005a) might also be a barrier to migration of gas, until they degrade and crack in response to lithostatic loading. However, cracking of the concrete liner is not important for migration of gas in the Andra concept since the main barrier to migration of gas from the near-field is the host rock (lower strength sedimentary type) and the shaft / drift / tunnel seals. Consequently migration of gas is primarily by dissolution and subsequent diffusion in the rock.

Transport in Water and Gas through the Seals and EDZ

Higher Strength Host Rock

The permeability of the EDZ is expected to be enhanced compared with the host rock. Therefore the EDZ can potentially form a preferential pathway for fluid migration. The host rock and the EDZ are both anticipated to contain fractures, and have very low gas entry pressures.

Vaults containing only packages of graphite wastes are expected to resaturate rapidly (Towler et al., 2011). Gas generation rates will be low and gas will readily migrate vertically from the vaults, through fractures in the EDZ and rock.Since free gas can readily migrate via the rock, the EDZ is unlikely to form a preferential pathway for migration of gas.

Pressurisation of the vaults is also unlikely, so water will not be driven through the EDZ due to the gas pressure. Bate et al. (2012) identified that some pressurisation could potentially occur if thick structural concrete vault linings were required. However, their calculations assumed UILW vaults with a much higher bulk gas generation rate than is relevant for vaults containing only packages of graphite wastes.

The EDZ might form a preferential pathway for radionuclide transport in water depending on the orientation of the vaults and tunnels, etc with respect to the background regional groundwater flow field. In terms of the overall disposal system performance, this could be described as a reduction in the performance of the geosphere barrier because it is bypassed.

NDA RWMD's illustrative GDF design for higher strength host rock (NDA RWMD, 2010b) assumes that the vaults will be closed with concrete seals in the access tunnels. The access drift and shafts will be sealed using compacted bentonite. Facility designs developed by other waste management organisations use similar sealing materials, although the specifications and performance requirements vary.

While the seals act to minimise the potential for flow and transport through the shafts and drift in higher strength host rock, their safety function is not as critical as in lower strength sedimentary host rock (described below). The performance requirement is that the seals are at least as impermeable as the host rock. In practice all the seals in the GDF should be relatively impermeable compared with the rock and EDZ, and there should not be significant fluid flow through the seals.

Lower Strength Sedimentary Host Rock

For lower strength rock, the permeability of the EDZ is also expected to be enhanced compared with the host rock. Therefore the EDZ can potentially form a preferential pathway for fluid migration, especially for gas since the EDZ gas entry pressure is expected to be lower than for the host rock. Gas generated in the vaults is expected to be trapped, with the resultant pressure build up potentially driving fluid (water and gas) flow along the EDZ, including the EDZs associated with the shafts / drift (Geofirma and Quintessa, 2011).

For vaults containing only packages of graphite wastes the gas generation rates are anticipated to be very low. It may take a very long time for gas to build up in the vaults and for the pressure to rise above hydrostatic. Indeed, gas could potentially dissolve and diffuse away from the vaults at a rate comparable to the generation rate, preventing pressurisation.

Watson et al. (2012) modelled gas generation in SILW vaults containing graphite and other wastes. The models considered a single (half) vault, but did not include the EDZ. The calculated saturations with time are shown in Figure 3-8. The timescale is the date (year AD), and the model includes the operational and post-closure phases. (The water saturation is zero during the operational phase). Although water is slowly driven out of the vault, even after 100,000 years insufficient gas has been generated to drive all the water out of the vault and there is limited pressurisation. However, the pressure could be sufficient to drive some fluid flow through the EDZ and any poorly performing

seals. Again this would be equivalent to bypassing at least part of the geosphere barrier.

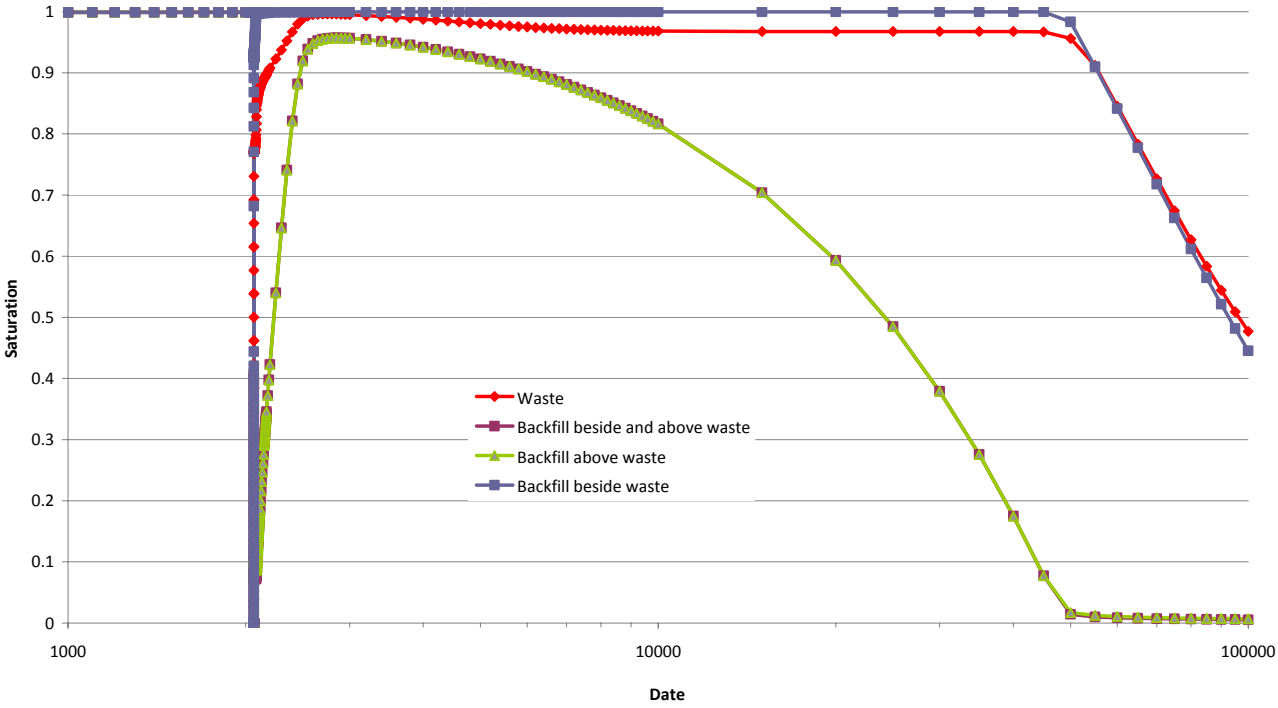

Figure 3-8. Evolution of water saturation in a SILW vault in lower strength sedimentary host rock (Watson et al. 2012)

Summary of Key Points from Section 3.1.1 (Near-Field Barriers)

- For the UK irradiated graphite waste inventory, the performance of the near-field barriers included in the UK ILW/LLW concept is consistent with guideline performance requirements for Cl-36 so long as there is a small amount of sorption. The near-field barriers significantly contribute to the guideline performance requirements for C-14 given cautious assumptions, and provide the guideline performance requirements given more realistic assumptions.
- Since the UK has a large inventory of irradiated graphite waste (by volume and radionuclide content), the calculations provide confidence that irradiated graphite wastes can be demonstrated to be suitable for geological disposal in isolation, once the additional barrier provided by the geosphere (next section) and conservatisms in the calculations are taken into consideration.
- Cl-36 is only weakly sorbed therefore the low water flow rate through the near-field is important (physical barrier). However, the small amount of sorption that

does occur can be important, and in addition under certain conditions Cl-36 may be retarded by co-precipitation. Doses from Cl-36 are also sensitive to the amount of dilution in shallow geosphere formations that is assumed in the biosphere model (i.e. derived guideline performance metric). For larger Cl-36 inventories low permeability EBS or long-lived containers may be required to reduce the rate of release of Cl-36 from the near-field, or a very low permeability geosphere may be required to provide a geosphere travel time that is significant compared with the half-life of Cl-36.

- It is anticipated that C-14 release rates from graphite will decrease as C-14 / C-12/13 concentrations in the container reach equilibrium. The assumption of instantaneous release or application of empirical C-14 release rates likely overestimates the flux of C-14 from graphite, as does the assumption that the entire C-14 inventory can be released.
- As a result of the above, the barrier performance of cementitious waste packages and backfill is likely underestimated in simple safety assessment calculations. Barrier performance may also be underestimated by conservatively excluding the processes of ion-exchange and co-precipitation at the carbonation front.
- The container is an important barrier. Containers could readily be engineered that are expected to have a lifetime that is significant compared with the half-life of C-14. In the simple model, the flux of C-14 from the near-field is controlled by diffusion out of the containers at the inorganic carbon solubility limit.
- Transport of C-14 in gas is unlikely to be a significant issue for graphite disposed in isolation, although direct release of gas from dry, damp and wet graphite, is poorly characterised.

3.1.2 Geosphere Barrier

The geosphere acts to isolate the wastes, and is therefore the primary barrier preventing inadvertent human intrusion, disruption / dispersion of the wastes by surface geomorphological processes, and impacts due to climate change. These aspects of the geosphere barrier performance are discussed in Section 4. The host rock also forms a barrier to radionuclide migration to the biosphere, which is the focus of this section. In this section we assume that the geosphere is the only barrier present. We explore the geosphere properties required to achieve the guideline performance.

C-14 has a half-life of 5.70E3 y (ICRP, 2007). It is therefore possible that C-14 will decay to negligible amounts before it is transported through the geosphere to the biosphere. Cl-36 has a half-life of 3.01E5 y (ICRP, 2007). It is therefore possible that Cl-36 may be subject to several half-lives of decay in the geosphere, but is only likely to decay to

negligible amounts in fairly extreme situations, i.e. a deep facility in which transport is diffusion dominated through a large thickness of very impermeable rock.

The permeability of the host rock and the magnitude of regional pressure gradients will control whether advective transport in the geosphere is possible. If advective transport in the geosphere is possible, there may also be advective transport in the near-field, depending on the properties of the near-field barriers. If advection is not possible, then radionuclide transport in water will be by diffusion.

C-14 and Cl-36 will not be significantly chemically retarded, e.g. through sorption, in the geosphere, e.g. Table 3-9 and Table 3-10. Therefore the key controls on transport will be hydraulic (including transport in water and gas for C-14).

Table 3-9. Sorption distribution coefficients for C-14

Rock	Kd (m^3 kg^{-1})	Reference
Marl	0.001	Stenhouse (1995)
Opalinus Clay	0.00013-0.006	Bradbury and Baeyens (2010)
Bentonite	0	Stenhouse (1995)
MX-80 Bentonite	0.000027-0.00032	Bradbury and Baeyens (2010)
Sandstone	0.000016	NDA RWMD (2010a)

Table 3-10. Sorption distribution coefficients for Cl-36

Rock	Kd(m^3 kg^{-1})	Reference
Marl	0	Stenhouse (1995)
Opalinus Clay	0	Bradbury and Baeyens (2010)
London Clay	0.0003-0.0038	Linklater et al. (2003)
Bentonite	0	Stenhouse (1995)
MX-80 Bentonite	0	Bradbury and Baeyens (2010)
Sandstone	0	NDA RWMD (2010a)

Gas will readily migrate from the GDF through any fractures in the host rock, but may be trapped in the GDF in unfractured very low permeability rocks, such as those being studied by Andra, Nagra and NWMO (Andra, 2005a; Nagra, 2008; Quintessa, 2011). Fractured rocks could extend to the biosphere, or may be overlain by sedimentary formations in which dissolution of gas is more likely to occur. In unfractured very low

permeability rocks there will be little transport of gas until a gas 'pathway'[7] is established, e.g. Nagra (2008), Geofirma and Quintessa (2011).

In the following section we consider the geosphere properties required to achieve the guideline performance for radionuclide transport in water. Radionuclide transport in gas is considered subsequently.

Advective Transport in Water in the Geosphere

Advective transport is most likely to occur in relatively permeable host rocks, although such host rocks may still be of low permeability in absolute terms. Advective transport can only occur if there is a regional hydraulic gradient driving flow. This hydraulic gradient may be topographically driven, or as a consequence of disequilibrium pressures at depth, i.e. pressures that are out of equilibrium with present day surface conditions, for example pressures are still recovering following melting of ice sheets after the last glacial maximum.

There is a relationship between the scale of topography and the depth to which it can influence flow. Only large, regional scale topography will influence flows at depths of several hundred metres. Disequilibrium pressures can only be maintained in very low permeability rocks. The lower the permeability of the rock the higher the disequilibrium pressure that can be maintained and the longer the time it can be maintained for.

For example, Nirex's Sellafield (UK) investigations revealed small excess pressures of 10 m water head at depth; and under-pressures of ~300 m water head and over-pressures of ~150 m water head have been measured at OPG's Bruce site in Canada (Quintessa and Geofirma, 2011). The permeability of the rock at the Bruce site is exceedingly low and despite the very high hydraulic gradients there is no advective flow: only diffusive transport is possible.

Assuming that the geosphere is the only barrier, for an advective system, the geosphere properties that are consistent with the guideline performance can be calculated. The calculations assume that the near-field is not a barrier, but do not ignore the presence of the near-field. The near-field is considered as a simple source volume, from which radionuclides are released (without decay or retardation) to the geosphere at a rate that is consistent with the advective flow rate in the geosphere. This is more realistic than considering that the entire radionuclide inventory is instantly released to the geosphere as a pulse source. A pulse source is not possible in reality

[7]A continuous free gas phase is present between the GDF and a permeable horizon / the ground surface.

and would require a stronger geosphere barrier to achieve the guideline performance. The calculations are fully described in Appendix D. (Note the calculations assume a near-field : geosphere porosity ratio of 5 : 1. Calculations assuming a ratio of 50 : 1 are also presented in Appendix D).

Figure 3-9 and Figure 3-10 show the calculation results for C-14 and Cl-36 respectively. Calculations are undertaken for different values of the hydraulic gradient. The maximum hydraulic gradient that has been considered is 0.1 since very high hydraulic gradients can only be sustained by very low permeability rocks, in which transport is likely to be diffusion rather than advection dominated, despite the high hydraulic gradient.

For a given set of rock properties and hydraulic gradient, Figure 3-9 and Figure 3-10 can be used to calculate the required geosphere pathlength to achieve the performance metric. It should be noted that the geosphere properties derived from the charts are consistent with a number of underpinning assumptions: C-14 or Cl-36 from graphite is the only radionuclide contributing to risk; the C-14 or Cl-36 inventory is the UK national inventory in graphite (classified as SILW); all C-14 or Cl-36 is immediately available for transport in water; the geosphere is the only pathway; and doses arise due to abstraction and use of well water.

For example, for a geosphere with a hydraulic conductivity of 1E-9 m s^{-1}, a porosity of 0.01, and a hydraulic gradient of 0.001, the pathlength from the GDF to the biosphere would need to be circa 200 m to achieve the guideline performance for C-14. For a hydraulic gradient of 0.01, the pathlength would need to be greater than 1000 m.

The same analysis can be applied to Figure 3-10 for Cl-36. The geosphere is required to be a better hydraulic barrier to achieve the guideline performance when compared with the requirements for C-14. This is because the half-life of Cl-36 (3.01E5 y) is significantly longer than for C-14. The half-life is also long compared with the anticipated geosphere travel time in a system where transport is dominantly by advection. (Cl-36 is not anticipated to be significantly retarded through sorption). Therefore, radionuclide decay in the geosphere is a relatively minor factor limiting the flux to the biosphere.

The shape of the Cl-36 is different to the C-14 curves. This is because the inventory of Cl-36 is small compared with C-14 (in terms of activity), and at low geosphere flow rates, the release rate from the near-field source is sufficiently low that the performance metric is achieved without the requirement for a geosphere barrier.

In a geosphere where transport is dominated by advection, the performance for Cl-36 is dominantly controlled by the slow flow rate through the GDF and weak sorption on

cements in the near-field (as applicable to the concept) and the amount of dilution assumed to occur in the shallow geological formations from which well water is abstracted.

Table 3-11 presents the hydraulic properties of a range of rock types that have been, or are being considered in radioactive waste disposal programmes. For rocks in which advective transport may be dominant, e.g. fractured crystalline rocks and sandstones, the geosphere alone may not be sufficient to provide the required disposal system performance. However, for deep geological disposal, the geosphere may significantly contribute to the overall disposal system performance.

Some of the programmes consider lower strength sedimentary type host rocks. Diffusion is anticipated to be the dominant transport processes in these rocks, and is considered next.

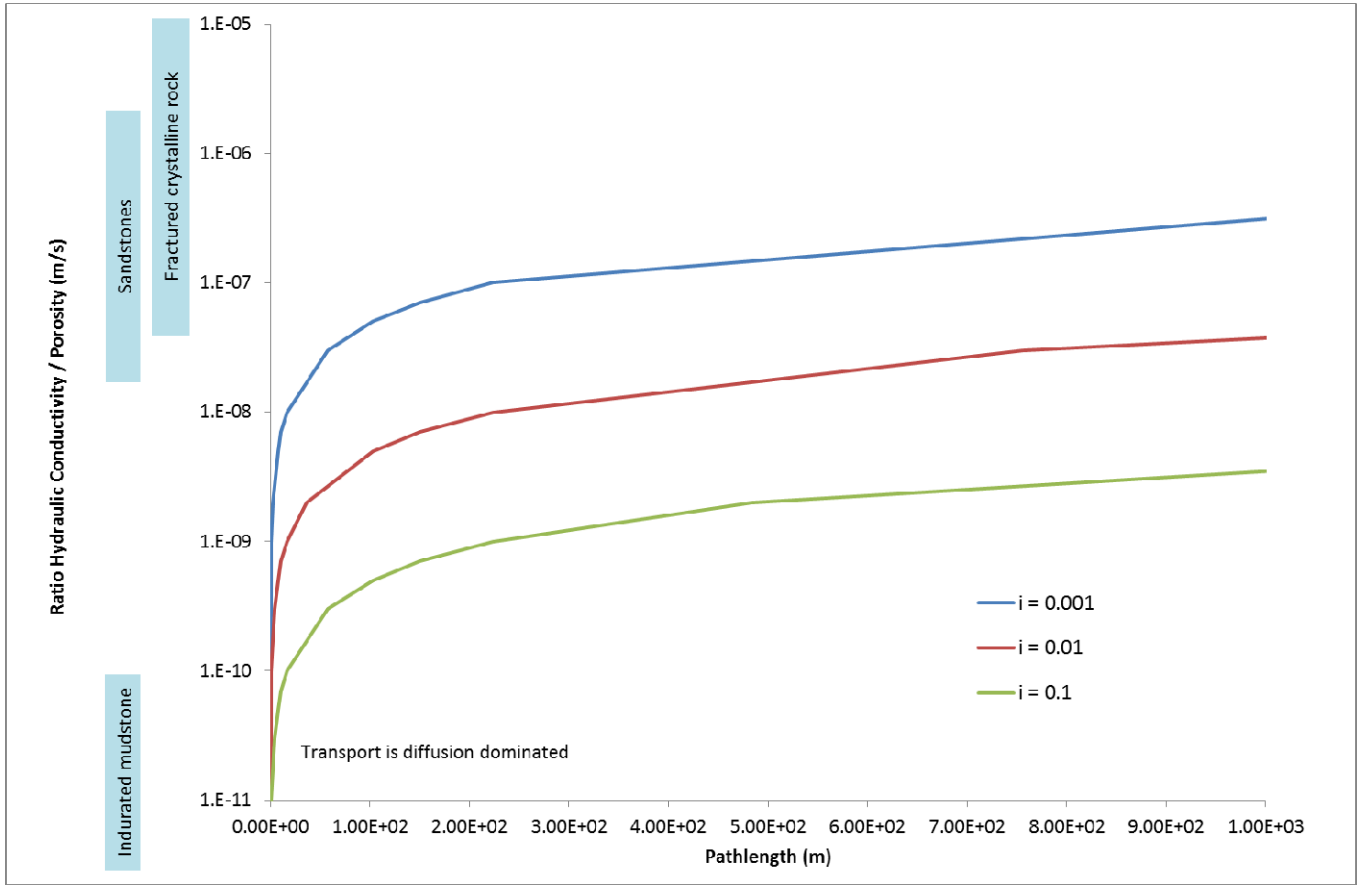

Figure 3-9. Geosphere properties to achieve performance metric for C-14 where transport is advection dominated (based on radionuclide inventory associated with UK SILW graphite wastes). i = hydraulic gradient.

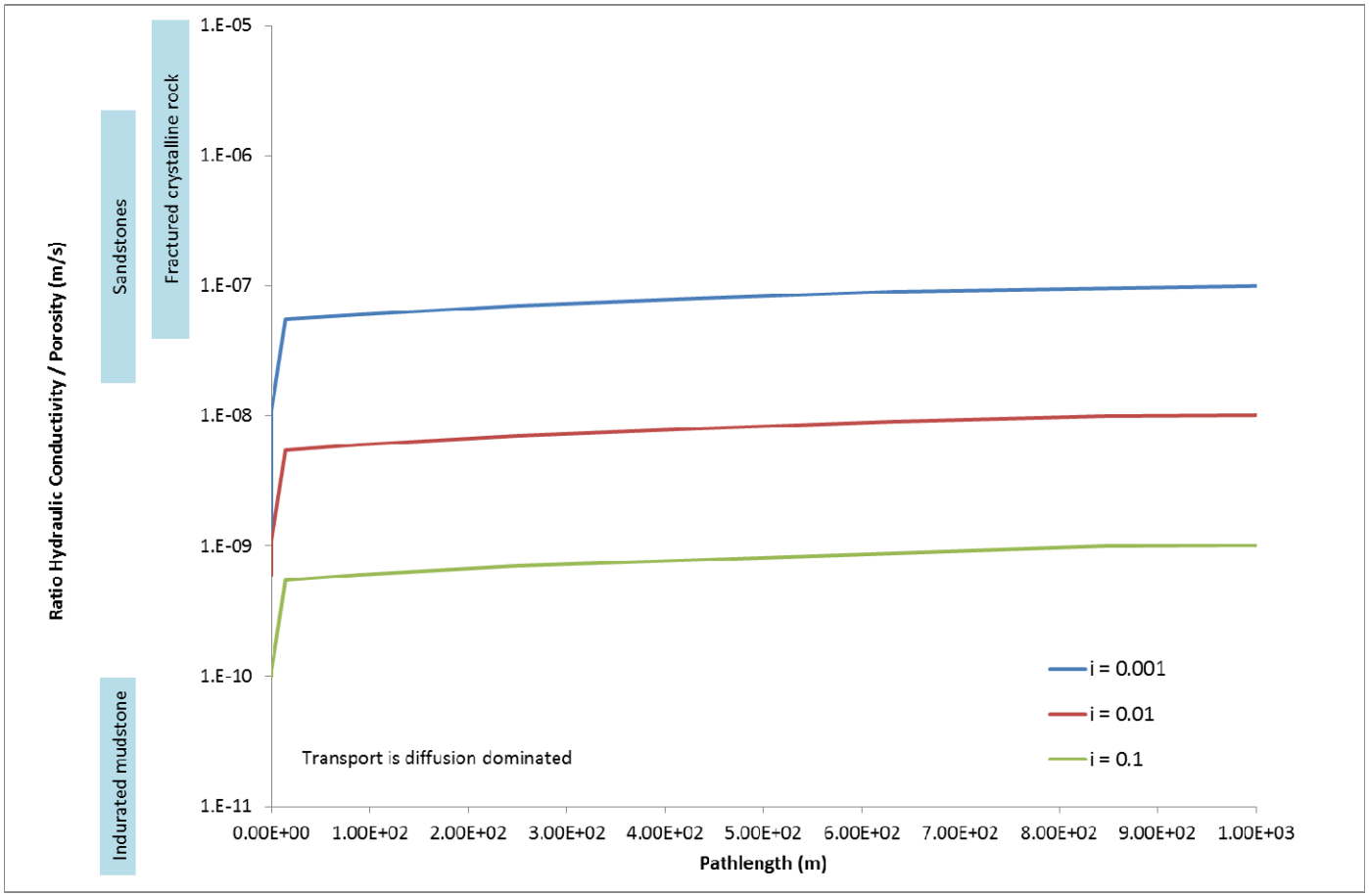

Figure 3-10. Geosphere properties to achieve performance metric for Cl-36where transport is advection dominated (based on radionuclide inventory associated with UK SILW graphite wastes).i = hydraulic gradient.

Table 3-11. Hydraulic properties for a range of rock types

Rock	Hydraulic Conductivity (m s^{-1})	Porosity (-)	Ratio Hydraulic Conductivity / Porosity (m s^{-1})	Reference
St Bees Sandstone, UK	Circa 1E-8*	0.10	1E-7	Nirex (1997)
BVG, UK	Circa 1E-9*	0.011 / 1E-5	>1E-7	Nirex (1997) / Hoch et al. (2008)
Fractured Crystalline, Sweden	1E-10 to 1E-8	Not available	>1E-8	Rhen and Hartley (2009)
Boom Clay, Mol, Belgium	1.3E-12 to 2.7E-10	0.34 to 0.41	3E-12 to 8E-10	Figure 7.3 of NWMO (2011)
Callovo-Oxfordian, Bure, France	6.1E-14 to 5.1E-13	0.15 to 0.20	3E-13 to 3E-12	Figure 7.3 of NWMO (2011)
Opalinus Clay, Mont Teri, Switzerland	1.8E-14 to 8.5E-13	0.15 to 0.17	1E-13 to 6E-12	Figure 7.3 of NWMO (2011)
Cobourg Fm, Bruce Site, Canada	1E-14 to 2E-14	0.01 to 0.03	3E-13 to 2E-12	Figure 7.3 of NWMO (2011)

For the cells shaded green, transport is anticipated to be diffusion dominated.

* Indicative values based on historic data, not intended to reflect a specific location.

Diffusive Transport in Water in the Geosphere

Charts of the geosphere properties required to achieve the desired barrier performance can also be developed considering diffusive transport in water. The key controls on the barrier performance are the pathlength and effective diffusivity.

Due to the planar geometry of the GDF, diffusion through the host rock will be approximately 1-D (Towler et al., 2008). The diffusive flux (J, mol s^{-1}) is equal to:

$$J = D_e \Delta / R$$

Where,

D_e is the effective diffusivity (m^2 s^{-1})

Δ is the concentration gradient (mol m^{-1})

R is the retardation factor due to sorption (-)

The effective diffusivity is equal to:

$$D_e = D\theta t$$

Where,

D is the free water diffusivity (1E-9 m^2 s^{-1})

θ is the porosity (-)

t is the tortuosity (-)

The concentration gradient in the geosphere will be non-linear and will evolve with time. Therefore the required geosphere properties were calculated using a simple GoldSim model which is described in Appendix E.

The calculations assume that the near-field is not a barrier, but do not ignore the presence of the near-field. The near-field is considered as a simple source volume, from which radionuclides are released by diffusion (without retardation) to the geosphere. Consistent with the basis for the performance metric, the calculations assume the host rock is overlain by more permeable geological formations from which well water can be abstracted.

The results are presented in Figure 3-11 and Figure 3-12. For relevant host rock properties (Table 3-12), the geosphere pathlength (and hence rock thickness) only needs to be of the order of tens of metres to achieve the guideline performance. The geosphere is required to be a stronger diffusive barrier to achieve the guideline performance for C-14 than for Cl-36. This is because in a diffusive system, the rate of diffusion out of the near-field source volume is almost sufficient to achieve the guideline performance for Cl-36 without the requirement for a geosphere barrier.

In summary, for a low permeability host rock in which transport is diffusion dominated, the geosphere alone can provide the required barrier performance for radionuclides associated with UK SILW graphite wastes, including for shallow geological disposal concepts.

Table 3-12. Diffuse host rock properties (Figure 7.4 of NWMO, 2011)

Rock	Effective Diffusivity of HTO (m^2 s^{-1})	Porosity (-)
Cobourg Fm, Bruce Site, Canada	2.0E-13	0.01
Palfris Fm, Wellenberg, Switzerland	2.0E-12	0.02
Queenston, Bruce Site, Canada	2.0E-12	0.08
Opalinus Clay, Benken, Switzerland	6.0E-12	0.12
Toarcian-Domerian, Tourmemire, France	6.5E-12	0.10
Opalinus Clay, Mont Teri, Switzerland	1.5E-11	0.16
Callovo-Oxfordian, Bure, France	2.6E-11	0.18
Boom Clay, Mol, Belgium	7.6E-11	0.38
London Clay, UK	7.5E-11	0.54

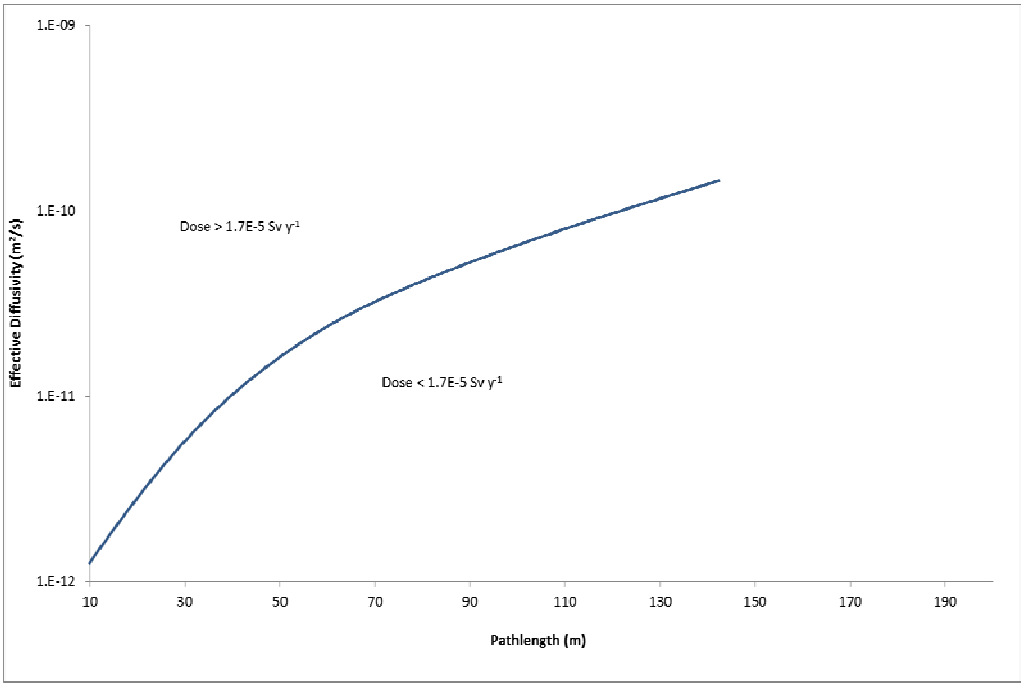

Figure 3-11. Geosphere properties to achieve performance metric for C-14 where transport is diffusion dominated (based on radionuclide inventory associated with UK SILW graphite wastes)

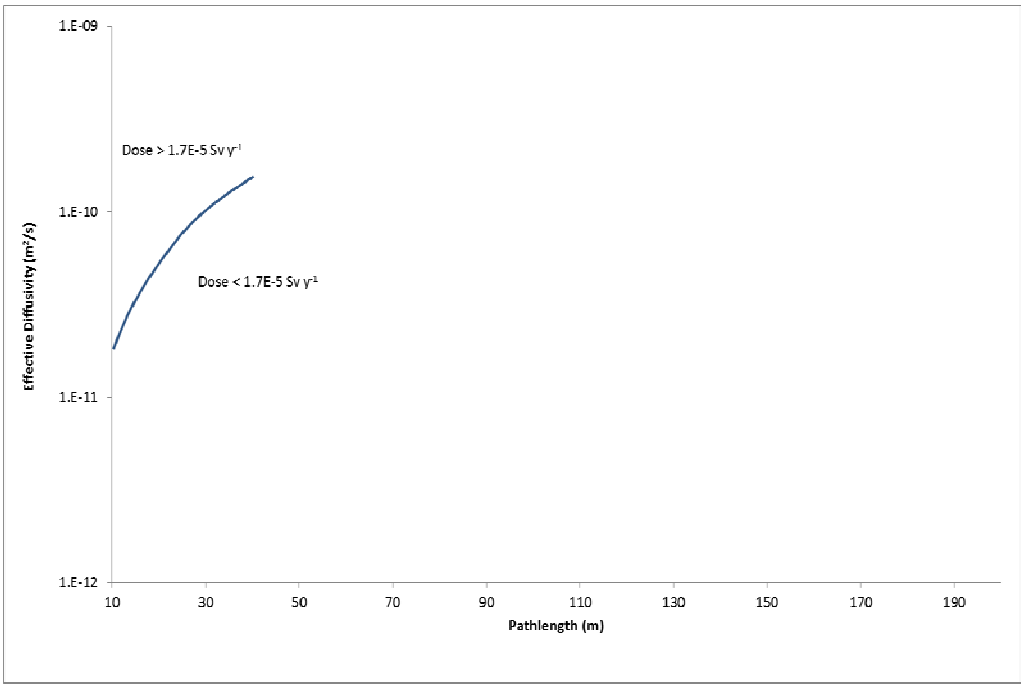

Figure 3-12.Geosphere properties to achieve performance metric for Cl-36 where transport is diffusion dominated (based on radionuclide inventory associated with UK SILW graphite wastes)

Transport in Gas in the Geosphere

In Table 2-4 a metric transfer rate of 3E-5 y^{-1} was calculated for C-14 in gas, given the C-14 inventory associated with UK's SILW graphite wastes. This correlates to a flux of 8.4E9 Bq y^{-1}. It is not possible to simply correlate this metric transfer rate with a simple set of geosphere properties. Although gas flow rates can be calculated as Darcy flow, the presence of water needs to be taken into account. Therefore, determination of the gas flow rate is a multi-phase flow problem, at a minimum considering water and gas, for example see the Phase 1 report (Towler et al. 2011). As a consequence the behaviour of gas is complex and non-linear.

The gas velocity will be equal to:

$$V_g = kk_r \rho g i / \mu \theta$$

Where,

V_g is the gas velocity (m s^{-1})

k is the intrinsic permeability of the rock (m^2)

k_r is the relative permeability, which varies, typically non-linearly, between 0 and 1 as a function of the gas saturation (-)

ρ is the gas density (kg m^{-3})

i is the gas pressure gradient, which includes both pressure and buoyancy forces (-)

g is acceleration due to gravity (m s^{-2})

θ is the porosity (-)

μ is the gas viscosity (Pa s)

We assume that there is not a 'background' free gas phase present in the geosphere. Therefore, initially (i.e. at closure) the relative permeability for gas will be zero (see equation above). Gas in the GDF will have to displace water in the geosphere in order to migrate into the rock. To displace water the gas pressure must rise above the geosphere hydrostatic pressure, plus the capillary suction pressure (for water), which increases as the water saturation in the rock decreases. (If 'background' free gas is present in the geosphere it will flow into the GDF because the background gas pressure will be high – greater than hydrostatic pressure).

Depending on the properties of the geosphere, gas may only migrate slowly from the GDF into the geosphere, gradually forming a 'plume' in which free gas and water are present in the rock. Some of the gas will dissolve in the water. If there is sufficient gas for this plume to reach a more permeable horizon a gas 'pathway' will be established and gas will migrate relatively rapidly via the pathway until the gas pressure drops and the pathway can no longer be maintained. Examples of gas pathway development are provided by Nagra (2008) and Geofirma and Quintessa (2011).

The key controls on migration of gas are therefore:

- the total gas generation rate and the total amount of gas (i.e. C-14 labelled gases and other bulk gases);
- the intrinsic permeability of the host rock, plus anisotropy which may drive lateral movement of gas in preference to vertical movement;
- GDF depth, and hence the hydrostatic pressure and pathlength;
- the host rock capillary suction curve (capillary suction as a function of water saturation);
- gas buoyancy; and
- host rock porosity and gas solubility, which control the amount of dissolution.

The total gas generation rate and the total amount of bulk gas are very important because this affects the gas pressure, the potential for a gas pathway to be established,

and the fraction of total gas which dissolves in water in the geosphere, i.e. bulk gas acts as a 'carrier' for C-14 labelled gas, for example see the Phase 1 modelling results. If the total gas generation rate and total amount of bulk gas are low then it is possible that all the gas will dissolve, the water will not become gas saturated, and a free gas phase will not form.

The chemical form of C-14, as either C-14 labelled CO_2 or CH_4 is also important, as is the ratio of C-14 labelled methane to stable methane, since there is only finite capacity for uptake of methane in biosphere soils (Section 2.4).

A key distinction between different host rocks is whether they are fractured or not, i.e. higher strength host rocks in which fractures may be present, e.g. fractured crystalline rock, and host rocks in which fractures may not be present, i.e. lower strength sedimentary and evaporite host rocks.

Fractured Rock

Gas is expected to be more mobile in fractured rock than unfractured rock: fractured rock is expected to be more permeable than unfractured rock, and the gas entry pressure of the fractures is low. The porosity of the fractures is also low. Consequently the pressure required for gas to enter the rock is relatively low, there is minimal pore volume in which gas can be 'stored' in the rock thereby minimising the pressure drop as gas migrates (expands) into the rock, and there is minimal volume of water for gas to dissolve in.

There is not anticipated to be release of bulk gas from graphite, only C-14 labelled trace gases. Therefore, in a vault only containing packages of graphite wastes, the only sources of bulk gas are corrosion of the stainless steel containers, and any metal vault furniture. This situation was examined for higher strength host rock in Case 2 of the Phase 1 report. The bulk gas generation rate was low, such that the gas saturation in the vault was low, and the majority of gas dissolved in the geosphere. Gas breakthrough occurred at the top of the host rock after ~6,000 years, but the flux was very small. Overlying formations were not included in the model, but this small gas flux would likely dissolve in water in the overlying formations.

Watson et al. (2012) explored generation and release of bulk gas from SILW vaults containing packages of many different types of waste in higher strength host rock. Bulk gas generation rates were higher than for vaults containing only packages of graphite wastes, but the results were comparable to the Phase 1 results.

The potential for dissolution in higher strength host rock depends on whether gas can dissolve in water in all the dead-end fractures and pores in the rock, or if gas migration is confined to the fracture porosity only. For example, the Phase 1 report and Watson et

al. (2012) assumed a total porosity of 1%, while Hoch et al. (2008) assumed a flowing fracture porosity of ~1E-3%.

In Section 3.1.1 direct release of C-14 labelled methane from graphite was discussed. Comparison of the measured release rate and performance metrics (Table 3-8) indicated that direct release of C-14 labelled methane might exceed the performance metric if the gas could rapidly be transported to the biosphere. Rapid transport may be possible in fractured host rock, but for graphite wastes disposed in isolation where bulk gas generation is low, gas would likely dissolve before reaching the biosphere.

If the small amount of bulk gas, and associated C-14 labelled methane was able to migrate through the fractured rock with no dissolution, and subsequently dissolve in shallow geosphere formations, from which well water is abstracted, the peak flux to the biosphere could be 6.1E10 Bq y^{-1} (Table 3-13). This is significantly higher than the maximum acceptable flux of C-14 to the biosphere in water (Table 3-13).

However, in reality this situation is unlikely to be important because it would require no dissolution in the near-field and deep geosphere and it is unlikely that more than a small fraction of the dissolved $^{14}CH_4$ will ever be captured by wells. Further, once the abstracted well water is exposed to the atmosphere, almost all $^{14}CH_4$ will exsolve (i.e. come out of solution and be released as gas) and might not lead to any exposures via the foodchain. Conversely, some $^{14}CH_4$ may be oxidised to $^{14}CO_2$ in oxic shallow geosphere formations, which would not exsolve to the same extent as $^{14}CH_4$ and might be more available for foodchain uptake.

Table 3-13. Consequences of dissolution of $^{14}CH_4$ gas (released directly from UK graphite) in shallow groundwater

Flux to shallow water from direct release of $^{14}CH_4$ from graphite	1E-5 (y^{-1}) x 6136 TBq = 6.1E10 Bq y^{-1}
Maximum acceptable flux of C-14 to the biosphere in water	6.3E7 (Bq y^{-1})

Unfractured Rock

Low permeability, unfractured rocks minimise the mobility of gas, and if such rocks form the host rock, gas will tend to be trapped in the disposal facility. Migration of bulk and trace gases will dominantly be via dissolution and subsequent diffusion (so long as the seals are effective). So long as the host rock and any overlying low permeability formations are of reasonable thickness, the diffusive travel time will be sufficient for C-14 to decay to negligible amounts before it diffuses to the shallow geosphere or biosphere.

These contrasting behaviours are illustrated by the multi-phase flow models presented in the Phase 1 report for higher strength host rock and lower strength sedimentary host rock. In higher strength host rock gas was rapidly released from the vaults and rapidly migrated through the host rock. In lower strength sedimentary host rock gas was largely trapped in the vaults, although there was some migration of gas into the EDZ.

Therefore, for graphite wastes in isolation, in lower strength sedimentary and evaporate host rocks there is not anticipated to be any significant migration of gas from the near-field (Section 3.1.1).

Summary of Key Points from Section 3.1.2 (Geosphere Barrier)

- Only very low permeability rocks in which transport is diffusion dominated are potentially important barriers for Cl-36.
- Gas is mobile in fractured rocks, but immobile in unfractured very low permeability rocks. Bulk gas generation is important as a carrier for trace gases, including $^{14}CH_4$.
- For fractured host rocks, the potential for gas discharge at the surface is affected by the presence / absence of permeable sedimentary formations overlying the fractured rocks. However, even if fractured rocks extend to the surface, near-surface the rocks will likely be significantly more fractured and hence permeable and porous, thereby allowing more gas to dissolve.
- Bulk gas generation is expected to be low for graphite disposed in isolation. Any gas that does not dissolve in deep water and reaches the shallow geosphere is likely to dissolve in shallow groundwater rather than discharge at the ground surface.
- Impacts from abstraction of shallow groundwater contaminated with dissolved C-14 gases will be sensitive to the amount of gas that dissolves in shallow groundwater, the amount of C-14 captured by the well, oxidation of $^{14}CH_4$ to $^{14}CO_2$ in oxic shallow groundwaters and exsolution of gas during abstraction. This is unlikely to be a significant issue for graphite wastes disposed in isolation but may be more significant for graphite wastes co-disposed with other gas generating ILW (Section 3.2).

3.1.3 Overall System Performance (Near-field and Geosphere)

In the previous sections it has been demonstrated that for graphite wastes disposed in isolation, guideline performance can be provided for transport of C-14 and Cl-36 in water and C-14 in gas by the near-field and/or geosphere barriers. Figure 3-13 shows the geosphere travel time required to achieve guideline performance for C-14 in water given the radionuclide fluxes from the near-field calculated by the simple near-field model, when ion exchange and co-precipitation at the carbonation front are not considered. The required travel time could easily be achieved in a geosphere where transport is diffusion dominated, and for many geospheres where transport is advection dominated.

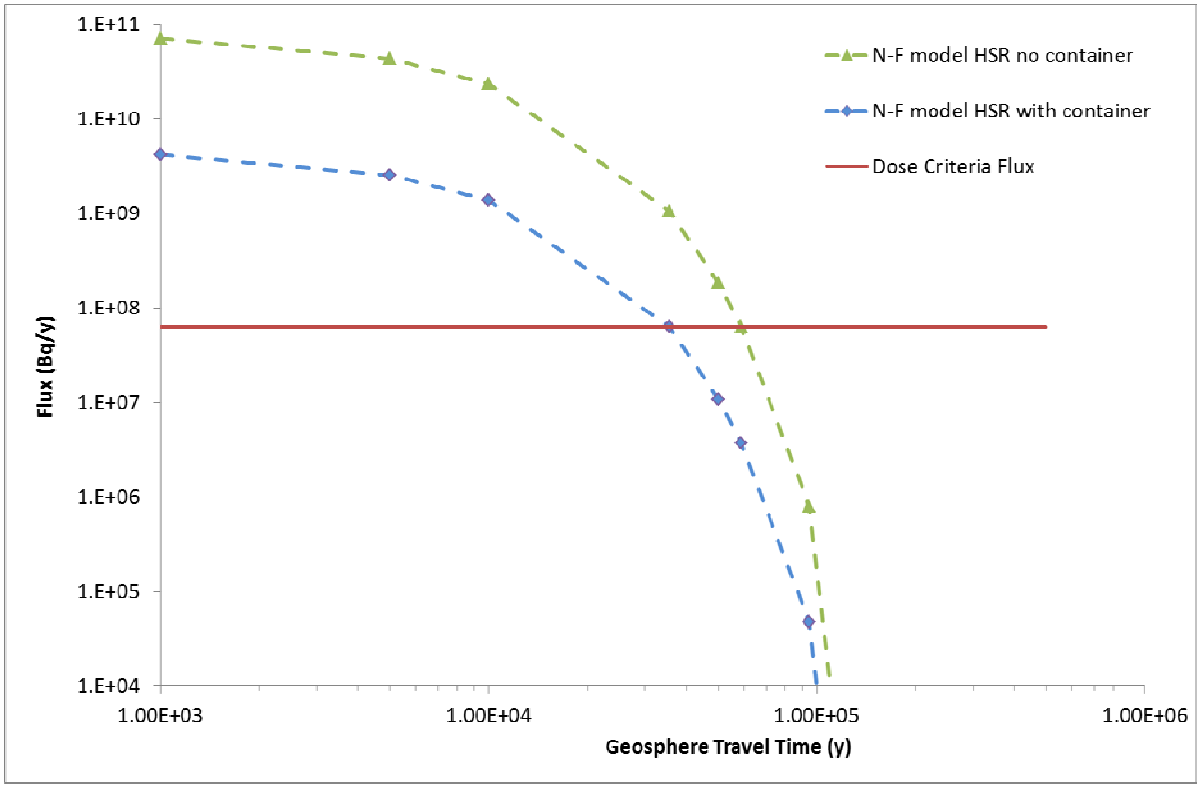

Figure 3-13. Required geosphere travel time in water to achieve guideline performance for C-14 given the radionuclides fluxes calculated by the near-field model for the UK SILW graphite inventory

3.1.4 Summary of Findings for Graphite Wastes Disposed in Isolation

This sub-section summarises the key conclusions from Sections 3.1.1 to 3.1.3.

The UK has a large inventory of irradiated graphite wastes, both in terms of volume and associated radionuclide inventory. It has been shown that it should be possible to safely dispose these graphite wastes in isolation in a wide range of host rocks. Therefore it can be concluded that graphite wastes are suitable for geological disposal in isolation.

Guideline performance can be provided by near-field and/or geosphere barriers, but the near-field chemical barrier provided by a cementitious concept is particularly important for C-14. This implies the need for a cementitious concept, however other concepts such as low permeability clay backfill may provide the required performance and may be optimal for higher Cl-36 inventories.

Graphite is a chemically unreactive material, and there is confidence in its long-term Thermo-Hydro-Mechanical-Chemical (THMC) evolution, i.e. there is not expected to be significant evolution. The key uncertainties relate to long-term C-14 release rate and fraction, and the behaviour of C-14 in the near-field. Simple near-field models, such as those used in this study, likely underestimate the chemical barrier performance of the cementitious near-field for C-14. Similarly, conservatisms in the biosphere models assumed for these calculations could result in the potential impacts being overestimates. Even with these conservatisms it is possible to safely dispose of irradiated graphite, but the conservatisms may affect siting and design decisions, or optimisation of the facility.

The uncertainties in C-14 release rate and behaviour in the near-field are potentially directly related, i.e. does the distribution of C-12/13/14 approach equilibrium and does the C-14 release rate decrease in response? Any future research to address residual uncertainties beyond CARBOWASTE should focus on developing a mechanistic release model for C-14, not just undertaking longer duration leaching experiments; and improved modelling of the behaviour of C-14 in the near-field.

3.2 Co-disposal of Graphite with Other ILW

In the previous section disposal of graphite wastes in isolation (i.e. vaults only containing packages of graphite wastes) was considered. However, packages of graphite wastes could be disposed in vaults containing packages of other ILW. For example, this is the assumption in RWMD's generic Disposal System Safety Case (gDSSC). Therefore the implications of co-disposal of packages of graphite wastes alongside packages of other ILW is considered in this section.

The other ILW wastes of particular interest are those that can influence the physico-chemical evolution of the vaults, and hence transport of radionuclides released from graphite. Processes influencing facility evolution can be categorised as being Thermo, Hydro, Mechanical or Chemical (THMC) processes. Analysis of the key THMC evolution processes for facility performance for co-disposal of graphite with other ILW wastes is presented in the subsequent sections. For Cl-36, there is only one additional potentially important process compared with disposal of graphite wastes in isolation – pressure driven flow in low permeability (gas tight) host rocks. Therefore, conclusions regarding barrier performance for Cl-36 are unchanged for most situations. This is not the case for C-14, which is therefore the focus of the analysis.

For the UK national inventory other wastes of interest include metal and organic wastes (Figure 2-2). The presence of these wastes will increase bulk gas generation, and hence the potential for transport in gas; and increase the potential for formation of methane, and hence the potential for formation of C-14 labelled methane. In very low permeability host rocks where gas tends to be trapped in the host rock, bulk gas generation will also influence the peak gas pressure, and hence the potential to drive flow of gas, water, and associated dissolved radionuclides, particularly via the EDZ and seals. Interactions between different wastes types and heterogeneity in the vaults also need to be considered since they may influence incorporation of C-14 in methane.

The majority of the UK graphite wastes and the associated radionuclide inventory are classified as SILW. There are only limited quantities of reactive metal and organic wastes in SILW, such that the consequences of disposal of packages of graphite wastes with other SILW wastes will be limited. The consequences will be more significant for UILW, since UILW wastes include large quantities of reactive metals and organics.

3.2.1 Higher Strength Host Rock

Table 3-14 summarises the key THMC evolution processes for facility performance for disposal of graphite, together with other ILW wastes, in higher strength host rock. For Cl-36, there are no additional important processes compared with those considered in Section 3.1. Therefore, conclusions regarding barrier performance for Cl-36 are unchanged. This is not the case for C-14.

Figure 2-2 describes the behaviour of bulk carbon. Methane generation is most likely to occur within waste packages containing organic wastes. Methane generation might also occur in the backfill. This depends on the effect of the containers as barriers to mixing and homogenisation of conditions in the vault. Since the graphite (source of C-14) and organic wastes are in distinct packages, the containers will act to inhibit incorporation of C-14 in methane (Figure 3-14).

Table 3-14. Summary of the key near-field THMC processes for facility performance for disposal of graphite in higher strength host rock

Thermo	There will be a thermal pulse associated with curing of cementitious backfill. This will increase the corrosion rate of some metals, in particular Magnox in the UK national inventory. This can result in a pulse of gas generation from Magnox waste, which can act as a carrier gas for C-14 labelled gases. Magnox corrosion is exothermic, so design temperature limits are required to prevent runaway corrosion, assuming sufficient water is available to support the reaction. This is also important because Magnox metal is another source of C-14. Longer-term, there is not anticipated to be significant heat generation from degradation of ILW/LLW.
Hydro	Materials in the facility will be partially saturated at closure. The vaults will rapidly substantially resaturate post-closure, and the pressure will recover over similar timescales. However, complete resaturation will not occur until gas generation ceases. Once the pressure in the vault recovers, there may be advective flow of water through the vault depending on the background regional hydraulic gradient. If there is no driving hydraulic gradient, then contaminant transport in water will be by diffusion. There will not be significant pressurisation of the vault. Gas will readily escape through fractures in the host rock, and dissolve in water in the disposal facility and host rock.
Mechanical	The backfill is not designed to be structural. There may be some movement of rock into the vault compacting the backfill and other materials in the vault, for example movement of blocks of rock that are bounded by fractures or weaker volumes of rock once rockbolts fail. This will result in enhancement of the EDZ. Mechanical compaction will alter the porosity of the backfill, and could damage the containers, especially once they have been weakened by corrosion.
Chemical	Even for pessimistic assumptions regarding water flow through the vaults, the backfill cement mineral inventory is sufficient to act as a chemical barrier to inorganic C-14 migration for hundreds of thousands to more than one million years (NDA RWMD, 2010a). Phase 1 model results indicate that armouring of fractures in the backfill will not denude the chemical barrier before the potential peak C-14 flux has passed. Initially radionuclides will only be able to diffuse out of the container vents. Once the containers have been perforated through corrosion, they will no longer prevent flow of water past the wastes.

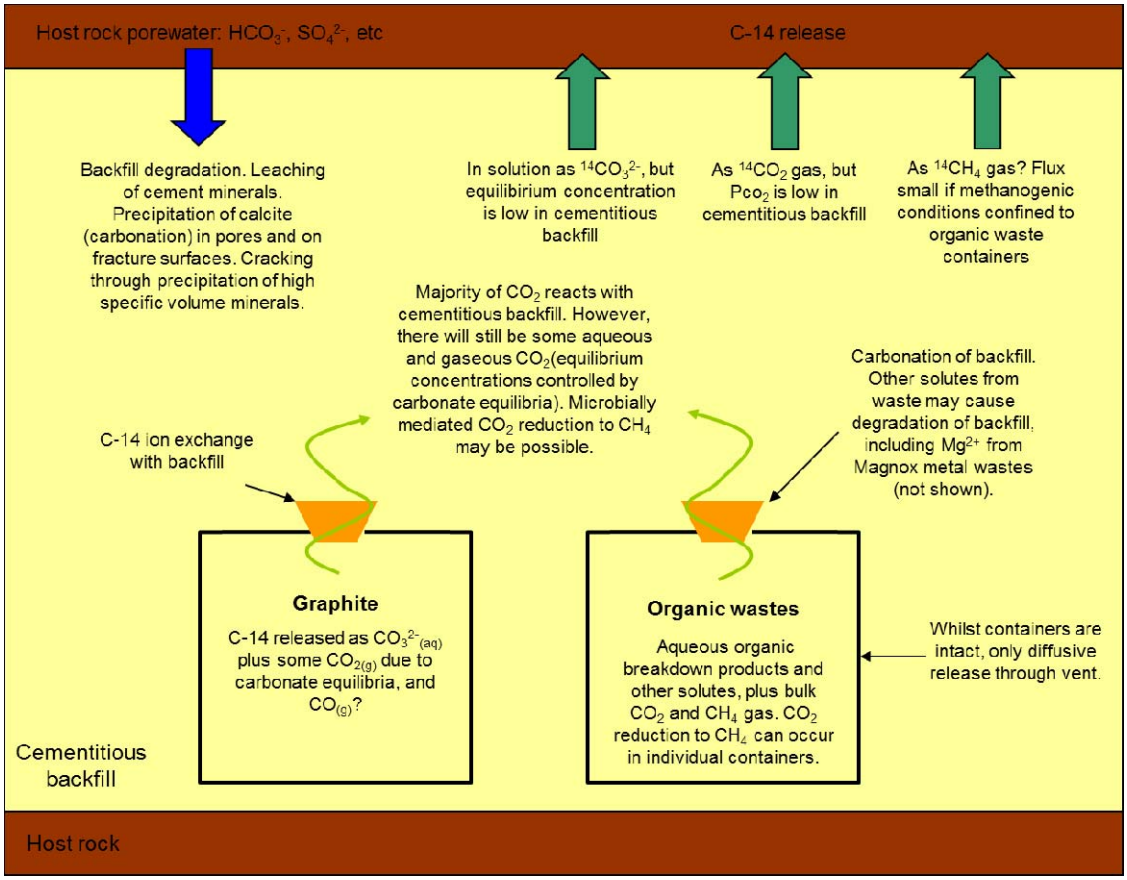

Figure 3-14. Conceptual model of the behaviour of inorganic C-14 in the GDF (for clarity, the following related processes that are not shown: generation of H_2 from anaerobic steel corrosion and radiolysis of water, polymers, oils and cellulose; generation of CH_4 and CO_2 by radiolysis of ISA – see Watson et al. (2012))

Table 3-15 compares the performance metric for C-14 release to the biosphere in methane and water with:

- the assumed direct release rate of C-14 labelled methane from graphite (Marshall et al., 2011);
- near-field release rates for C-14 in gas and water (dominantly C-14 labelled methane in gas phase and as dissolved gas) from the Phase 1 models (Case 1 in Towler et al., 2011).

The Phase 1 model results assume homogeneous conditions in the vault, that all the graphite wastes (i.e. UILW and SILW) are exposed to UILW vault conditions, and that CO_2 reduction to CH_4 is only limited by the available H_2. The values in parenthesis in Table 3-15 describe the model results for UILW graphite only.

For the conditions assumed in the Phase 1 model (Case 1), even if only UILW graphite was disposed, in the absence of any attenuation of C-14 labelled methane gas through dissolution in the geosphere, the peak C-14 gas flux to the biosphere would exceed the performance metric. The mobility and dissolution of gas is considered below.

The near-field release rate of dissolved C-14 labelled methane gas is also notable, since for UILW only it is approaching the performance metric for C-14 in water to the biosphere, and exceeds the metric for all graphite. However, the C-14 flux associated with transport of dissolved $^{14}CH_4$ will be attenuated by decay in the geosphere.

The higher the bulk gas generation rate the larger the plume of free gas that develops in the geosphere and the less water there is for dissolution of gas. Hoch et al. (2008) showed that for fractured crystalline rock with a fracture porosity of ~1E-5, the total GDF gas generation rate was sufficient for free gas to migrate to the top of the host rock. In that example the host rock was overlain by thick sedimentary formations which had sufficient capacity for all the gas to dissolve. The formations were also sufficiently thick that dissolved C-14 decayed while it was transported through the sedimentary formations to the biosphere. However, geosphere systems can be envisaged where fluxes to the biosphere in water or gas could exceed the required disposal system performance, e.g. fractured rock to surface, or fractured rock only overlain by thin sedimentary formations.

Table 3-15. Release of C-14 from the GDF for graphite wastes co-disposed with other ILW in higher strength host rock

Measure	Release Rate (y^{-1})	Flux (Bq y^{-1})	Notes
Direct release to methane from graphite	1E-5	6.1E10	UILW and SILW graphite Peak flux at time = 0 y is 1E-5 (y^{-1}) x 6.1E15 Bq = 6.1E10 Bq y^{-1}
Release of C-14 in gas from Phase 1 model results (Case 1 in Towler et al., 2011)	2E-2	1.0E11 (1.2E9)	Peak rate from Phase 1 model results (Case 1 therein with realistic C-14 leaching rate). Model assumes homogeneous UILW vault conditions applied to the total UK C-14 inventory in graphite. Model excludes contribution from direct release to methane from graphite.
Gas flux and rate metrics: λ_1: Waste release transfer rate (y^{-1}) to achieve guideline performance for C-14 transport in gas(i.e. no near-field or geosphere barriers) λ_2: Disposal system transfer rate (y^{-1}) to achieve guideline performance for C-14 transport in gas	1E-6 3E-5	7.8E8	
Release rate for C-14 in water from Phase 1 model results (Case 1)	4E-5	1.6E9 (1.8E7)	Dominated by dissolved methane.
Water flux and rate metrics: λ_2: Maximum acceptable C-14 release rate in water	7E-6	6.3E7	

*Value in parenthesis describes results for UILW graphite only.

3.2.2 Lower Strength Sedimentary Host Rock

Table 3-16 summarises the key near-field THMC evolution processes for facility performance for disposal of graphite, together with other ILW wastes, in lower strength sedimentary host rock. For Cl-36, there is only one significant change to the important processes compared with those considered in Section 3.1. This is the increased potential for water containing dissolved Cl-36 to be driven through the

seals/EDZ due to the gas pressure. This is discussed in the next sub-section. Otherwise the conclusions regarding barrier performance for Cl-36 are unchanged. This is not the case for C-14.

Conditions in the near-field may be more homogeneous than with higher strength host rock, because release from the near-field will be very slow, and there will be time for interaction and homogenisation via diffusion, especially as diffusion through the vault materials may be rapid compared with diffusion into the rock. This increases the potential for interaction between different waste types and incorporation of C-14 into methane generated from degradation of organic wastes.

Table 3-16. Summary of the key near-field THMC processes for facility performance for disposal of graphite in lower strength sedimentary host rock

Thermo	There will be a thermal pulse associated with curing of cementitious backfill. This will increase the corrosion rate of some metals, in particular Magnox in the UK national inventory. This can result in a pulse of gas generation from Magnox waste. Magnox corrosion is exothermic, so design temperature limits are required to prevent runaway corrosion, assuming sufficient water is available to support the reaction. This is also important because Magnox metal is another source of C-14. Longer-term, there is not anticipated to be significant heat generation from degradation of ILW/LLW.
Hydro	Materials in the facility will be partially saturated at closure. There may be some gas release as the seals resaturate, because the relative permeability for gas will be high in partially saturated seals. However, gas generated in the vaults will largely be trapped in the vaults and the pressure will increase, decreasing the rate of inflow of water into the vaults. The vaults will resaturate slowly post-closure. The gas pressure may rise above hydrostatic pressure, pushing water out of the vaults. The vaults may remain partially saturated, or largely desaturated for hundreds of thousands of years or more. Aqueous transport will be by diffusion. Gas will dissolve in water in the rock and will slowly diffuse away from the vault. The vaults will only fully resaturate once gas generation ceases and as gas dissolves and diffuses away from the vault.
Mechanical	The backfill is structural. The backfill will resist movement of rock into the vault, and promote self-healing of the EDZ. As the load on the backfill increases, due to creep and swelling of the rock, and the waste packages and backfill chemically degrade, the backfill will crack (e.g. Andra, 2005a). Steel containers will be distorted and damaged. The backfill will still provide a chemical barrier because there will not be advective flows through the backfill.
Chemical	There will only be diffusion of solutes into the vault. The backfill chemical barrier will therefore last for longer than in the higher strength host rock example. Fractures in the backfill are unlikely to decrease performance due to armouring of fracture surfaces (no advective flow through the fractures).

Table 3-17 compares the performance metric for C-14 release rate to the biosphere in methane and water with:

- the assumed direct release rate of C-14 labelled methane from graphite (Marshall et al., 2011);
- near-field release rates for C-14 in gas and water (dominantly C-14 labelled methane in gas phase and as dissolved gas) from the Phase 1 models (Case 5 in Towler et al., 2011).

Again, the Phase 1 model results assume homogeneous conditions in the vault, that all the graphite wastes (i.e. UILW and SILW) are exposed to UILW vault conditions, and that CO_2 reduction to CH_4 is only limited by the available H_2. The values in parenthesis in Table 3-17 describe the model results for UILW graphite only.

The model results show that the near-field alone provides the guideline barrier performance for UILW graphite, and provides a significant proportion of the guideline barrier performance for all graphite (i.e. SILW and UILW). Given the very low permeability of the host rock, the combined performance of the near-field and geosphere barriers should achieve the guideline barrier performance for all graphite. However, the model does not consider migration of gas through the seals / EDZ, which is a potentially important transport route. This is discussed in the next sub-section.

Table 3-17. Release of C-14 from the GDF for graphite wastes co-disposed with other ILW in lower strength sedimentary host rock

Measure	Release Rate (y^{-1})	Flux (Bq y^{-1})	Notes
Direct release to methane from graphite	1E-5	6.1E10	UILW and SILW graphite Peak flux at time = 0 y is 1E-5 (y^{-1}) x 6.1E15 Bq = 6.1E10 Bq y^{-1}
Release of C-14 in gas from Phase 1 model results (Case 5 in Towler et al., 2011)	5E-5	1.4E10 (1.5E8)	Peak rate from Phase 1 model results (Case 5 therein with realistic C-14 leaching rate). Model assumes homogeneous UILW vault conditions applied to the total UK C-14 inventory in graphite. Model excludes contribution from direct release to methane from graphite. Ignores higher release rate to EDZ only during the first few years post-closure.
Gas flux and rate metrics: λ_1: Waste release transfer rate (y^{-1}) to achieve guideline performance for C-14 transport in gas(i.e. no near-field or geosphere barriers) λ_2: Disposal system transfer rate (y^{-1}) to achieve guideline performance for C-14 transport in gas	1E-6 3E-5	7.8E8	
Release rate for C-14 in water from Phase 1 model results (Case 5)	7E-6	6.5E8 (7.3E6)	Ignores higher release rate to EDZ only during the first few years post-closure.
Water flux and rate metrics: λ_2: Maximum acceptable C-14 release rate in water	7E-6	6.3E7	

*Value in parenthesis describes results for UILW graphite only.

Transport in Water and Gas through the Seals and EDZ

The greater the bulk gas generation rate in the vaults containing packages of graphite wastes, the greater the potential for transport of radionuclides from graphite, in water and gas, through the seals and EDZ due to pressure driven flow. Different gas management strategies are available, and the chosen strategy will need to consider a

range of issues in addition to transport of radionuclides released from graphite, e.g. peak gas pressure and the potential for hydrofracking.

In a very low permeability host rock, the shafts / drift and associated EDZs may be important potential preferential pathways. For the most impermeable host rocks it may not be possible to achieve seals that are as impermeable as the rock (Table 3-18). In this instance there may be some migration of gas through the seals even though they are not intended to form an Engineered Gas Transport System (EGTS) (see below).

Table 3-18.NWMO rock and seal material hydraulic properties (Quintessa and Geofirma, 2011)

Material	Hydraulic Conductivity (for Water) (m s^{-1})
Undegraded structural concrete	1E-10
Undegraded Low Heat High Performance Cement (LHHPC)	2E-12
Asphalt	1E-12
Cobourg (host formation)	2E-14 horizontal 2E-15 vertical
Queenston (an overlying formation)	2E-14 horizontal 2E-15 vertical

Therefore, NWMO and Andra's designs, which aim to contain gas in the near-field, include strategies to disrupt transport through the seals and EDZ:

- mechanical removal of the EDZ, although complete mechanical removal may not be possible since undamaged rock will relax into the excavation as it is exposed (NWMO);
- penetration (NWMO) / interruption (Andra) of the EDZ and sealing of interfaces by swelling clay seals; and
- structural support to promote self-healing of the EDZ in argillaceous rocks (Andra).

Geofirma and Quintessa (2011) examined the performance of NWMO's proposed shaft seals, and the shaft EDZ. They found that gas transport to permeable shallow geological formations was only possible for a severe shaft seal failure scenario, and then only if extreme assumptions were made about the properties of the degraded shaft materials.

In contrast Nagra's Engineered Gas Transport System (EGTS) is designed to permit gas transport with consequent pressure relief, while still being impermeable to water. The

objective of the EGTS is to prevent hydrofracking of the host rock, which is more likely to occur than for the NWMO and Andra concepts, due to the shallower depth of the facility, and hence lower lithostatic pressure. If an EGTS is used, the consequences of dissolution of gas in the tunnels / shafts / drift and overlying formations will potentially become more significant.

If the gas pressure is sufficient to drive water up the shaft / drift and the associated EDZ, dissolved gases will be transported with the water. There will be little dilution as the water migrates up the shaft / EDZ, such that dissolved gases may come out of solution as the pressure drops.

Exsolution is not expected to be a major transport pathway for gas. CH_4 is not very soluble such that the amount of gas that can dissolve and subsequently exsolve is anticipated to be small. Exsolution of dissolved CO_2 will also result in precipitation of calcite and trapping of some C-14 in calcite. As calcite precipitates it will act to reduce the permeability of the shaft / drift seals and EDZ.

Finally it is noted that there may be background free gas present in the host rock. If this is the case the gas must have been trapped for geological timescales, and therefore the presence of free gas does not imply an established gas pathway. It is assumed that site selection criteria will have excluded potential resources such as shale gas. Permeable reservoir rocks containing gas (including 'sour' gas) will also be excluded because of their resource potential and because construction will not be possible. Therefore, the only free gas that potentially may be present is small amounts (i.e. not commercially exploitable) in very low permeability formations.

In such circumstances the geosphere gas pressure will be high, and gas will tend to migrate from the rock into the disposal facility. This might increase the peak gas pressure in the repository and may increase the potential for migration of gas and water via the shaft / draft and associated EDZ. However, if gas is trapped because of the inherent low permeability of the host rock, the timescales for migration of gas into the repository will be very long (Geofirma and Quintessa, 20011).

3.2.3 Evaporite Host Rock

Evaporite host rock is highly impermeable and there is not likely to be significant radionuclide transport in gas or water via advection or diffusion. Creep of the host rock will cause the EDZ to heal and will reconstitute crushed evaporite backfill in the access, service, etc tunnels. It is possible that there could be some gas release at early times before the EDZ and tunnel fill reconstitutes. This will be sensitive to the amount of water present in the waste packages when they are disposed, and whether any water is available from the host rock (Watson et al., 2012). Microbial gas generation may be limited by the highly saline conditions.

3.2.4 Summary of Findings for Co-disposal of Graphite Wastes with Other ILW

A broader range of processes become important for co-disposal of graphite with other ILW compared with graphite wastes isolation. Behaviour becomes more site / design specific and the important behaviours may change as the system evolves. This makes it difficult to explore generically the suitability of graphite for geological disposal with other ILW.

Detailed models of example systems indicate that it should be possible to achieve the guidance performance for lower strength sedimentary (and by analogy, evaporate) host rocks. It may also be possible to achieve guideline performance in higher strength host rock environments, but model results indicate that migration of C-14 in (free and dissolved) gas may be an issue. However, the models tend to overestimate the significance of migration of C-14 in gas because they do not take into account the role of the waste packages as a barrier to migration (e.g. of dissolved gas), or as a barrier to interactions between graphite wastes and other ILW.

Overall, it is concluded that it is preferable to segregate packages of graphite wastes from packages containing organic wastes, reactive metals and any other significant gas generating wastes. Where this is not possible / desirable, the waste package is an important barrier to prevent interaction between different waste types. For vaults containing different waste types, complex models (e.g. Towler et al., 2011; Quintessa, 2011) can be used to help understand the behaviour and partitioning of C-14, sensitivity to carrier gas generation and peak gas pressure, etc. The bulk gas management strategy (e.g. containment or engineered gas transport system, EGTS) needs to consider the behaviour and transport of C-14 in addition to issues such as the peak gas pressure.

4 Analysis of Human Intrusion and Disruptive Event Variant Scenarios

The previous sections have explored the suitability of graphite wastes for geological disposal considering the normal evolution of the GDF. Taking the UK national inventory as an example, it has been demonstrated that it should be possible to safely dispose of a large volume of irradiated graphite waste under a wide range of geological conditions.

Safety is provided by engineered and natural (geological) barriers. Human intrusion and disruptive events have the potential to bypass these barriers. Therefore this section considers whether there are specific issues associated with graphite wastes for human intrusion and disruptive events:

- What is the magnitude of the potential impacts associated with human intrusion and disruptive scenarios for graphite wastes?
- How does the magnitude of potential impacts associated with intrusion and disruptive scenarios for graphite wastes compare with other wastes that may be present in the disposal facility?
- Which scenarios are most important for graphite? Are these different to the important scenarios for other wastes, or the scenarios typically considered in safety assessments?

It is assumed that radionuclide migration in water and gas will eventually occur. However, the probability of human intrusion and disruptive events is low and uncertain. It is therefore necessary to firstly consider how this uncertainty affects the criteria and approach used to assess these variant scenarios (Section 4.1).

A wide range of potentially relevant human intrusion and disruptive event scenarios can be envisaged for different sites and GDF designs (in particular depending on the depth of the facility). It is not possible to consider all potential scenarios of interest in this report. Therefore, having discussed the assessment criteria and approach, we review approaches to identifying scenarios (Section 4.2) before exploring the consequences of graphite wastes for some of the more prominent scenarios (Sections 4.3 and 4.4). The consequences are again explored for the UK national inventory in order to identify and examine any issues that may challenge the suitability of irradiated graphite wastes for geological disposal.

Further international guidance on assessment of human intrusion and disruptive event scenarios is provided by IAEA (2004, 2012).

4.1 Assessment Criteria and Approach

4.1.1 Human Intrusion

The radiological consequences associated with human intrusion into a deep geological disposal facility are typically considered in a different way to other scenarios. This is because the probability of human intrusion is considered to be very low. Nevertheless, there is a possibility of occurrence, and the scenario can involve bypassing natural and engineered barriers resulting in direct exposure to the waste (and consequently potentially significant impacts). Therefore international guidance and national waste management regulations, e.g. Guidance on Requirements for Authorisation (GRA) (Environment Agency and Northern Ireland Environment Agency, 2009; Environment Agency et al., 2009) require the potential consequences of human intrusion after the period of authorisation to be assessed.

The UK regulator's recommended approach for assessment of the consequences of human intrusion into a deep GDF after the period of authorisation is shown in Figure 4-1.

Human intrusion is taken to include intrusion directly into a disposal facility as well as actions that degrade barriers. Human intrusion assessments should consider both those involved in the intrusion and those who might receive "secondary" exposures afterwards as well as non-human biota. Consequences beyond this (such as sinking a well into an aquifer contaminated by radionuclides) are also relevant. We do not explore the consequences for non-human biota in this report.

It is assumed that a period of institutional control will be maintained following closure of the disposal facility. Inadvertent human intrusion will not be possible during the period of institutional control. Many concepts assume an institutional control period of 100 y, however longer timescales up to 300 y may be considered depending on the nature of the wastes, the change in hazard with time and other considerations. A specific institutional control period has not been assumed in the following sections.

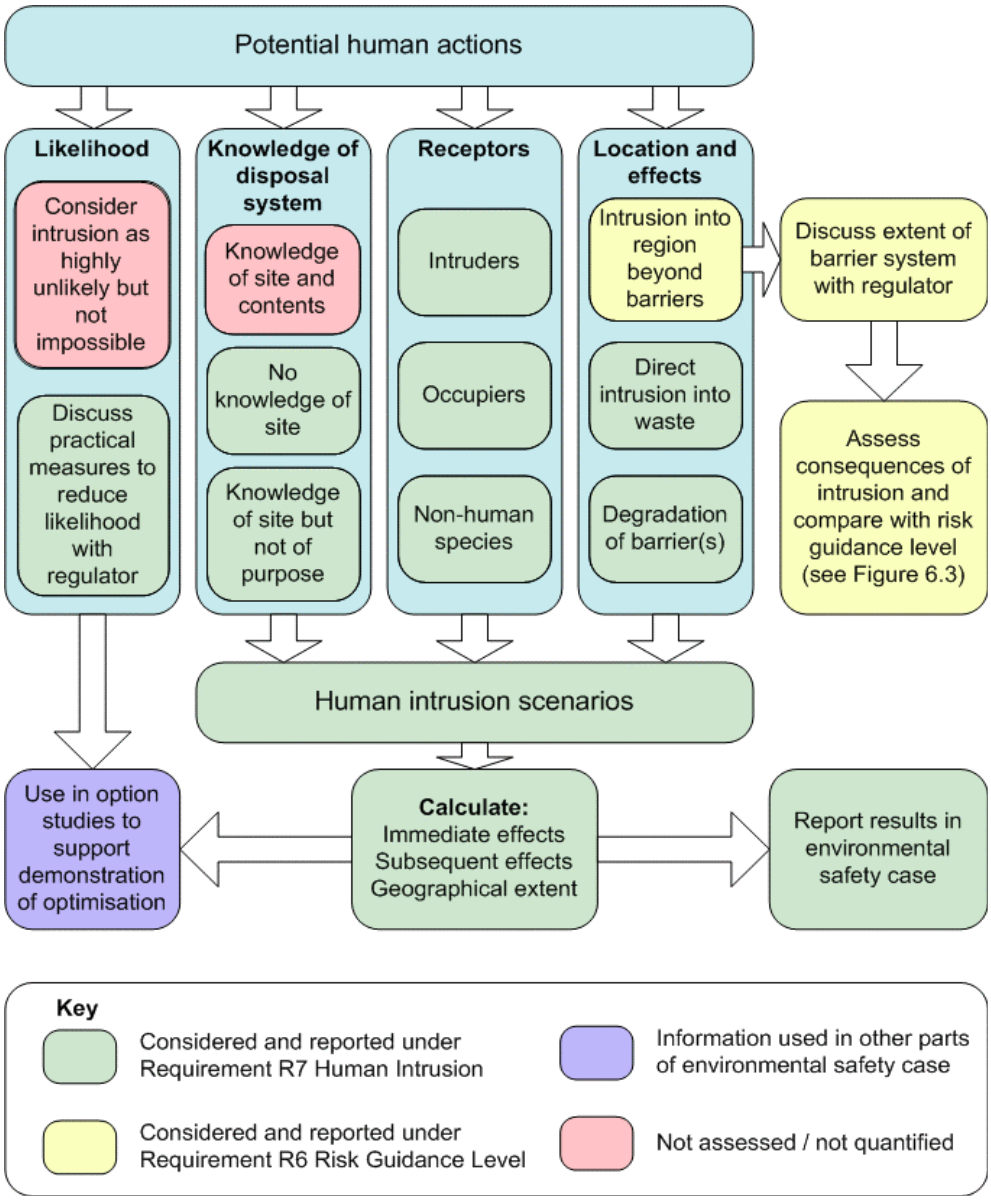

Figure 4-1: Process for the assessment of human intrusion into a GDF, after the period of authorisation (in the UK: Environment Agency and Northern Ireland Environment Agency, 2009)

It is noted that human intrusion into near-surface disposal facilities may be treated significantly differently. This is important for concepts that consider near-surface disposal of graphite. The difference in approach reflects the much greater human impact on the near-surface environment. This, in turn, leads to a greater range of potential situations in which intrusion may occur, and a greater likelihood of occurrence. Indeed, regulatory guidance in the UK (Environment Agency et al., 2009) requires that for near-surface facilities human intrusion be considered "likely to occur" (over the whole period that is assessed after the period of authorisation).

Scenarios can be identified from international and site specific FEP lists, for example IAEA (2004). An example of the range of possible intrusion events that have been considered for a near-surface LLW disposal facility, and measures that could be undertaken to reduce the likelihood or consequence of each intrusion event is provided by Hicks and Baldwin (2011). Examples of the events considered by Hicks and Baldwin (2011) include: housing development, leisure development, farm / smallholding, geotechnical investigations, large infrastructure development, etc. This is further discussed in Section 4.2.1.

For near-surface disposal, in the UK the assessed effective dose is compared to a dose guidance level in the range of around 3 mSv y^{-1} to around 20 mSv y^{-1}. Values towards the lower end of this range are applicable to assessed exposures continuing over a period of years (prolonged exposures), while values towards the upper end of the range are applicable to assessed exposures that are only short term (transitory exposures). A dose criterion is used because it is problematic to assign probability estimates (from which risks can be derived) over long timescales to events that are highly dependent on human society and its activities.

For deep disposal in the UK, guidance indicates that the dose guidance level is not applicable for scenarios considering human intrusion into the facility; however the developer / operator is expected to present material to help judge whether the disposal facility is properly optimised (Environment Agency and Northern Ireland Environment Agency, 2009). Nevertheless for the purposes of this study we apply a dose guidance level of 3 mSv $y^{-1}$to 20 mSv y^{-1} to both shallow and deep geological disposal, to provide a common basis to understand the potential importance of irradiated graphite wastes in the context of human intrusion, and identify particularly significant scenarios.

It is recognised that assessment criteria may vary in other countries, however the UK dose criteria is considered to be appropriate to exploring the suitability of graphite wastes for geological disposal. ICRP (2007) provides international guidance on doses associated with intervention situations.

4.1.2 Disruptive Events

Disruptive events other than those characterised by human actions (discussed above) also require assessment. These events are uncertain, but have the potential to reduce the effectiveness of the natural and/or engineered barriers. Fault reactivation and glacial denudation (significant erosion of the geosphere) are examples of disruptive event scenarios.

If the probability of an event occurring can reasonably be determined (and in some cases, such as glacial perturbation, this may be unity), it can be explicitly introduced into a calculation of risk. This is because the assessed radiological risk can be calculated as the product of:

- the calculated effective dose;
- the estimated probability of health detriment per unit dose; and
- the estimated probability that the exposure would occur within the assessment timeframe.

The first part of the calculation is a conventional radiological assessment calculation for the given scenario. It is this aspect that any specific properties of graphite wastes may influence, such as a significant concentration of a particular key radionuclide. International guidance (ICRP, 2007) provides the basis for the "risk factor" that is the second element of the calculation. The final part is scenario-specific and requires a basis of evidence for its quantification. Typically, for natural events, historic evidence (as obtained from geological records) provides the basis for the estimation of the probability of occurrence. Information on earthquakes, for example, may be determined with a degree of reliability in this way. However, other potentially important disruptive effects may be difficult to predict because historic conditions do not provide a good analogue for the future (e.g. because of climate change) or because other less predictable factors influence them (e.g. human activity).

In the UK, consistent with the normal evolution scenario, risks calculated for disruptive events are compared against a risk guidance level of 1E-6 y^{-1}. Again, although assessment criteria may vary in other countries, this risk guideline is considered to be appropriate to explore the suitability of graphite wastes for geological disposal.

It is noted that the probability of occurrence may be unquantifiable. In this instance it can remain useful to investigate consequences provided due allowance is given for the anticipated likelihood. Therefore, even where events are highly uncertain, "what-if" calculations can be informative.

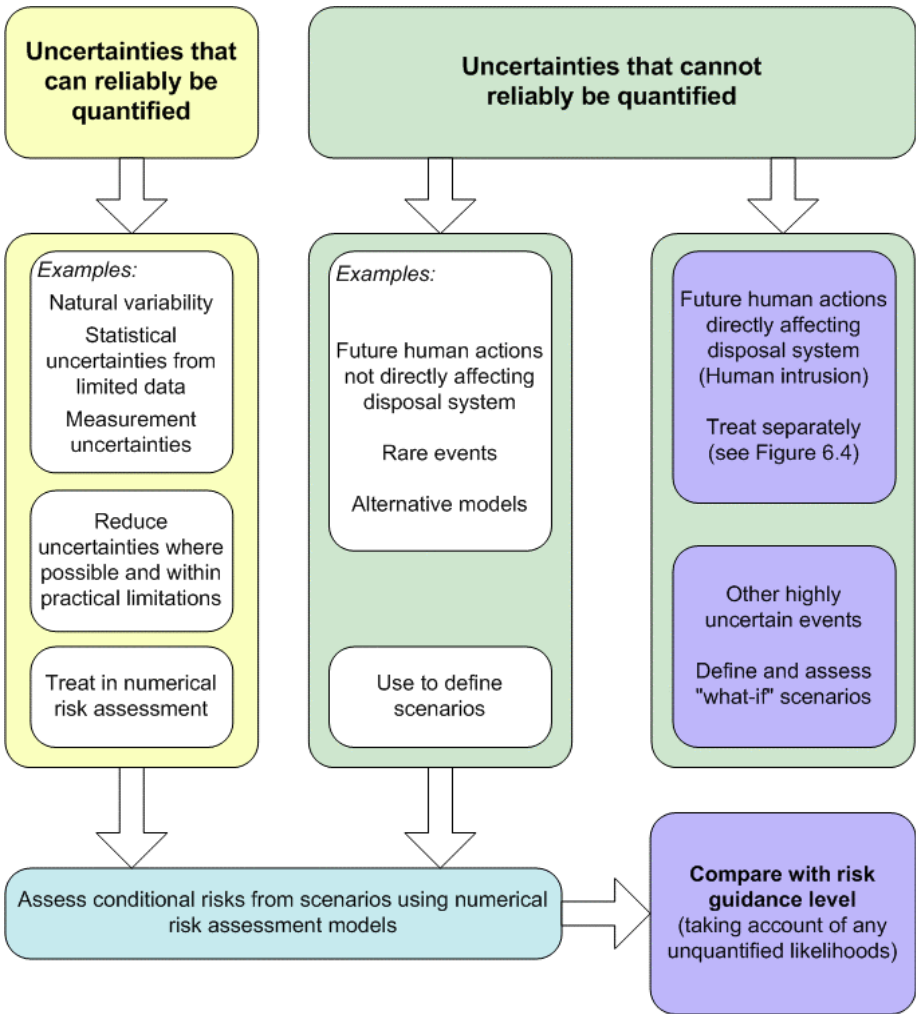

Figure 4-2: Approach to assessing events that are uncertain (in the UK: Environment Agency and Northern Ireland Environment Agency, 2009)

4.1.3 Importance of Graphite Wastes

Graphite represents about 33% of the total mass of ILW/LLW in the UK (Pöyry, 2010) and therefore represents, in its own right, a significant proportion of the national inventory. A general indication of the importance of graphite wastes to the outcome of human intrusion and natural disruptive event assessments can be gained by comparing the key radionuclides identified in post-closure safety assessments with those present in graphite wastes.

Pöyry (2010) identified the most important radionuclides in the UK derived inventory of ILW based on potential post closure impacts (Table 4-1). In discussing the importance of specific materials, Pöyry (2010) notes that graphite is one of the most important materials due to its contribution to the total UK inventory of C-14 and Cl-36. Table 4-1 shows that C-14 is a key radionuclide in respect of gas and Cl-36 in respect of

groundwater, but that other radionuclides dominate the human intrusion scenario. The radionuclides dominating human intrusion are long-lived alpha-emitters (e.g. actinides) and gamma-emitters (e.g. Nb-94).

In the calculations summarised in Table 4-1 pathways other than human intrusion are associated with the peak risks for C-14 and Cl-36. Nevertheless it is possible that impacts from the human intrusion pathway could also be important. These aspects require specific consideration because:

- Human intrusion is a key mechanism by which C-14 bearing gas release can occur from the disposal system;
- Human intrusion and natural disruptive events have the potential to degrade engineered and natural barriers and hence increase the rate at which contaminants can migrate via groundwater, potentially increasing the overall impact of the pathway for which Cl-36 is a key radionuclide.
- Both Cl-36 and C-14 are radionuclides of moderate radiotoxicity. Both radionuclides are long-lived beta-emitters and therefore the main exposure mode is by internal exposure. Hence scenarios of relevance are those that might lead to internal exposure via radionuclide transport in water and gas. The following subsections consider in further detail the scenarios of relevance and their assessment by RWMD and others, and provide a more detailed analysis of the importance of graphite wastes in these scenarios.

Table 4-1: Key radionuclides in the UK ILW/LLW Inventory

Radionuclide	Contribution of graphite to total inventory*	Peak conditional risk (y^{-1})^	Exposure
C-14	79%	1.0E-06	Gas
Cl-36	72%	3.0E-08	Groundwater
Se-79	-	1.0E-08	Groundwater
Nb-94	-	1.1E-09	Intrusion
Tc-99	-	1.1E-09	Groundwater
Sn-126	-	1.1E-08	Groundwater
I-129	-	5.1E-08	Groundwater
Cs-135	-	3.1E-09	Groundwater
Th-232	-	3.5E-09	Groundwater
U-234	-	2.9E-08	Groundwater
U-235	-	1.6E-08	Groundwater
U-236	-	4.7E-09	Groundwater
U-238	-	9.3E-07	Intrusion
Np-237	-	2.6E-07	Groundwater
Pu-239	-	6.5E-09	Intrusion
Pu-240	-	4.9E-09	Intrusion
Am-241	-	3.1E-09	Intrusion

Note: * The only key radionuclides that occur with any significance in the graphite are C-14 and Cl-36. ^The peak risk is that cited by Pöyry (2010), these values are only indicative but illustrate the magnitude of risks and key pathways for particular radionuclides.

4.2 Scenarios

4.2.1 Scenario Identification Methods

For a given disposal system, both human intrusion and natural disruptive event scenarios can be determined by considering the processes and events that could bypass the natural and engineered barriers. There are various methods for identifying scenario-initiating events and/or processes (e.g. as discussed by IAEA, 2004) but most make use of a FEP (Features, Events and Process) list. FEPs are screened for inclusion or exclusion from the scope of the assessment by consideration of their likelihood and

consequence. The location of the disposal system will tend to determine the likelihood, whereas the properties of the wastes (e.g. radionuclide concentrations or physical form) will tend to influence the consequence. Whilst it is the latter that is of interest in reviewing the significance of graphite wastes, it is necessary to constrain the scope of scenarios to be considered with reference to the likelihood of their occurrence. This is most readily done by reviewing the types of scenarios usually assessed for geological disposal systems.

4.2.2 Scenarios Considered in NDA RWMD's gDSSC

In RWMD's recent generic Disposal System Safety Case (gDSSC) (NDA RWMD, 2010a) it is noted that human intrusion and disruptive events can be dependent on the specific location and detailed GDF design (particularly facility depth and footprint). As the assessment is generic, it is only possible to include illustrative calculations.

In relation to human intrusion, a situation is considered in which exploratory drilling for natural resources penetrates the repository. Material is assumed to be retrieved from the repository (i.e. in borehole core). A geotechnical worker is assumed to be exposed when examining the material. The release of gas to the surface is also considered by NDA RWMD (2010a). Both a "slow" release (the same rate as gas is being generated), and a "rapid" release (of pressurised gas) are considered. The results are preliminary, and indicate that the primary radionuclide of importance is Rn-222. In their assessment of the impact of gas released as a result of human intrusion, Hoch and Thorne (2008) state that:

> *"...for some source region characteristics and times of intrusion, the radiological impact of ^{14}C-methane release in over-pressurised gas cannot be neglected. Nevertheless, it is of limited significance compared with the potential radiological impact of releases of ^{222}Rn at the rate of production."*

Disruptive events are not directly assessed in the gDSSC as phenomenological "scenarios". However, an investigation is made into the sensitivity of the performance of the GDF in terms of "travel time" through the geosphere. Figure 4-3shows the calculated risks for a variety of combinations of groundwater travel time and flow velocity through the GDF. The clear grouping of results into three sets is a result of the dominant effect of the three different groundwater travel time values included in the analysis.

A shorter travel time can be considered to be representative of degradation of the geosphere that could occur due to some major, unspecified, disruptive event. Therefore the magnitude of potential consequences of the event for radionuclide transport can be indicated by a transition from an initial set of curves, to a second set of curves which are associated with a shorter geosphere travel time.

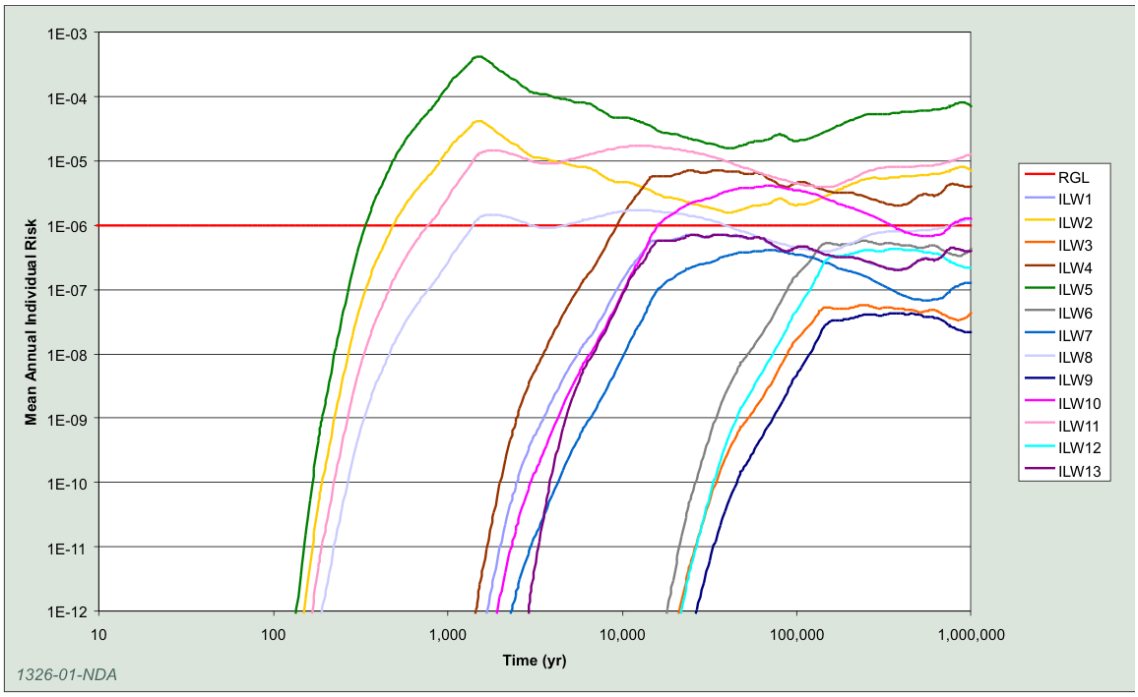

Figure 4-3: Total radiological risks from groundwater releases for a suite of calculations exploring sensitivity to travel time and flow velocity through the GDF (All ILW and LLW including graphite) (NDA RWMD, 2010a)

4.2.3 Scenarios in Other Safety Assessments

An example of systematically identified scenarios for a geological disposal system is the recent assessment of the Deep Geologic Repository (DGR) in Canada (Quintessa and SENES, 2011). The screening argument for a human intrusion FEP is shown in Figure 4-4, and the screening argument for a disruptive event is shown in Figure 4-5.

Figure 4-4: Example of screening the FEP "Future Human Actions: Drilling Activities" in relation to the proposed DGR at Bruce, Canada (Quintessa et al., 2011)

Screening Analysis

Water wells can be expected to be drilled into the Shallow Bedrock Groundwater Zone in the future (such wells currently exist in the region around the site). They are unlikely to penetrate beyond 80 m at the site since the water is not potable at greater depths…

Exploration boreholes could be drilled down into the [Deep Geological Repository]. However, the drilling of such deep boreholes at the site is very unlikely. The depth (about 680 m), small footprint of the DGR's emplacement rooms (about 0.065 km^2)…, and lack of commercially viable natural resources … in the area mean that the annual probability of such a borehole intruding into the DGR would be very low. Furthermore, over a million year time frame, drilling would also be limited by the presence of ice-sheets.

It is possible that the repository might be detected by remote measurement methods, and be deliberately targeted for study. Again, the uniformity of the sediments and lack of interesting minerals or geologic features in the area would argue against deliberate surveys of the area. Furthermore, if the repository were detected as an anomaly and deliberately targeted, then the nature of the contact with the repository would likely be more carefully managed.

Such wells and boreholes would only be drilled on the site once controls were no longer effective.

FEP Screening

Shallow water wells are included in all scenarios. Deep drilling is screened out from all scenarios other than the Human Intrusion Scenario. An exploration borehole penetrating the DGR is considered in the Human Intrusion Scenario.

Figure 4-5: Example of screening the FEP "Geological Processes and Effects: Seismicity" in relation to the proposed DGR at Bruce, Canada (Quintessa et al., 2011)

Screening Analysis

Southwestern Ontario and the Bruce region lie within the tectonically stable interior of the North American continent, which is characterized by low rates of seismicity. A recent study of seismicity in the Canadian craton suggests a seismic recurrence rate for **M**> 6 events of <0.001 per annum per 10^6 km^2 with a variability (standard deviation) of about a factor of three ... To put this into context, an event of **M** ≥ 6 would be expected somewhere within a 20-km radius of the Bruce nuclear site roughly once in 800,000 years with an uncertainty factor of 3 on this return period.

Records of earthquakes in the region since the late 1800s up to 2009, and monitoring results from the seismograph stations around the Bruce nuclear site, show that the Bruce region experiences sparse seismic activity… Most recorded events have a magnitude less than **M**3…

Dynamic mechanical modelling studies have been used to investigate the impacts of seismic events on cavern stability… The model results show that seismic shaking does not cause any additional damage or fracturing of the rock mass. … The seismic shaking does promote unravelling of already fractured and loose rock mass...Geomechanical modelling has demonstrated that the caverns and pillars will remain stable for seismic events of **M** ≤ 5.0 at 15 km radius and of **M** ≤ 6.5 at 50 km from the site (Section 6.5, AECOM 2011).

Seismic reactivation of existing faults is a remote possibility as it would require a very large event to occur right at the repository site. Furthermore, since the repository is sited in an area where no faults have been observed, it would require fracturing of previously unfaulted rock.

FEP Screening

Include rockfall effect in all scenarios. Severe seismic event considered as a potential cause for the activation of the fault in the Vertical Fault Scenario and the degradation of the shaft seals in the Severe Shaft Seal Failure Scenario.

The comprehensive FEP analysis undertaken for the Canadian DGR for L/ILW identified the following human intrusion and disruptive event scenarios (Quintessa and SENES, 2011):

- Human Intrusion - unintentional intrusion into the repository as a result of an exploration borehole, leading to release of contaminated materials (gas, water and solid and particulate) from the repository to the surface environment and to the shallow groundwater;
- Severe Shaft Seal Failure Scenario – a "what if" scenario considering the unexpected poor performance of the shaft seals which leads to enhanced flows of groundwater (and potentially gas) up the shaft;
- Poorly Sealed Borehole Scenario - the potential for a poorly sealed site investigation/monitoring borehole near the repository, resulting in an enhanced permeability path through the rock towards the surface; and
- Vertical Fault Scenario - an extreme earthquake is assumed to reactivate a hypothetical fault in the vicinity of the DGR, providing a more permeable connection to overlying geological formations.

Another example of the scenarios identified for a deep geological repository is those identified in the SR-Site assessment for a deep geological repository for Spent Fuel (SF) in Sweden (SKB, 2011). The scenarios considered in the assessment are:

- Human intrusion into the repository through drilling/boring;
- Human intrusion into rock nearby the disposal facility (e.g. construction of a nearby rock facility);
- Unsealed repository;
- Performance degradation of the buffer around the spent fuel canisters, for example due to poor emplacement or freezing; and
- Performance degradation of the disposal container (canister), for example due to isostatic load, shear or unexpected corrosion.

The general themes under investigation in both the DGR and SR-Site assessment are similar. Human intrusion is assessed, even if it is unlikely, but boreholes are the only reasonably plausible manner by which a deep geological disposal system may be affected. Scenarios related to other disruptive events tend to focus on the degradation of key barriers in order to understand their overall contribution to the safety function of the disposal facility. These include "what if" cases such as the unsealed repository scenario.

A dominant factor in determining the events that are relevant to assess is the location of the repository at depth, in a geological environment. A much wider range of situations is relevant for a facility nearer the surface. For example, HPA identifies a wide range of stylised human intrusion scenarios for a near-surface facility (Oatway et al., 2011). The guidance considered a facility located at between 15 and 65 m depth, and the suggested scenarios included tunnelling into the facility, open cast mining, and direct intrusion (with man access) into the disposal facility. The greater proximity of the disposal facility to the surface means that larger-scale excavations are conceivable. The greater proximity to the surface also broadens the range of disruptive events that require assessment. For example, coastal erosion can potentially affect the surface to a depth of tens of metres, and glacial scouring potentially deeper still.

4.3 Qualitative Assessment

The importance of graphite wastes to the potential impacts of human intrusion and disruptive events is dependent on their particular characteristics in the context of the scenarios discussed in Section 4.2. In this respect, it is first useful to note the main physical, chemical and radiological characteristics of graphite wastes (Table 4-2). The key features of graphite wastes, from the perspective of human intrusion and disruptive events, are:

- the homogeneity of the material;
- the potential for direct release of radioactive gas, water and solid; and
- the environmental mobility of the radionuclides present (especially Cl-36).

Table 4-2: Comparison of physical, chemical and radiological properties of interest to human intrusion and disruptive event scenarios

Property	Graphite Wastes	The Whole Inventory
Physical	Graphite is a highly homogeneous material. Easily identified. It has moderate strength and might, for example, be retrieved intact or in large pieces.	Throughout the disposal system there is a considerable degree of heterogeneity in wastes. The dominant types of material are likely to include metals and concreted wastes.
Chemical	Both C-14 and Cl-36 can be leached from graphite in water. As discussed in Section 2.2, C-14 may be released from graphite both in solution and gas, and may also be present in gas evolved in the repository. Both radionuclides have the potential to be mobile in the geological environment.	The GDF chemistry is complex, but likely to be dominated by the influence of grout (alkaline conditions – where relevant), metal corrosion and organic degradation. These influence the rate at which radionuclides are released and the chemical conditions in the disposal facility (including gas generation).
Radiological	The dominant radionuclides in graphite wastes are C-14 and Cl-36. Both radionuclides decay by beta decay, and consequently the most significant exposure mode is generally internal exposure rather than external irradiation.	There is a wide range of radionuclides, including long-lived alpha and beta/gamma emitters present in high concentrations, which can dominate human intrusion and disruptive events.

Physical Properties and Homogeneity

In respect of human intrusion and disruptive event scenarios, the physical properties and homogeneity of graphite wastes is important because the material is reasonably predictable, identifiable, and reasonably robust. In an anoxic environment, where oxidation is not possible, graphite might still be identifiable on geological timescales, even if its physical structure had been 'damaged' through lithostatic loading, etc.

Considering a human intrusion scenario, it is plausible that, should a graphite block be intercepted by a borehole, the material would be retrieved substantially intact in the borehole core. While its radioactivity might not be determined by a geotechnical worker, it would probably be identified as an unusual, and likely to be man-made, material. This could influence the attention given to the sample, potentially affecting the exposure duration.

In respect of disruptive events, the events are expected to primarily impact on GDF performance remotely, by degrading barriers. Therefore the physical properties of graphite are not important.

Potential for Direct Release of Gas, Water and Solid

Although graphite is not expected to result in any significant quantities of bulk gas, the partitioning of carbon between solid, liquid and aqueous phases, and the potentially large source of C-14 in graphite (80% of the total UK ILW/LLW C-14 inventory, Table 4-3), means that the wastes may have an important influence on the generation of C-14 labelled CO_2 and CH_4. As noted in Section 3.2 the generation of bulk CO_2 is likely to be low for cementitious disposal system concepts and interaction with organic wastes is likely required for C-14 from graphite to be significantly incorporated into methane.

Bulk gas provides a medium by which contaminants may be rapidly transported under certain conditions. Migration of radionuclides in gas is potentially important for human intrusion and disruptive event situations, especially if pressurisation can occur (Hoch and Thorne, 2008; Quintessa and SENES, 2011). Consequently, graphite wastes may be significant for human intrusion and disruptive event scenarios in which gas is a key medium.

It is notable that in the assessment of the Canadian DGR concept (Quintessa and SENES, 2011) it was found that direct release to the surface of water and solid from the repository could occur. Specific conditions, in terms of repository pressurisation and saturation, are necessary. If these occur, it was found that the transport of C-14 in aqueous form to the surface provided an important pathway, especially for long-term residents of the former drilling site. Indeed, this mode of release led to higher calculated exposures from C-14 than release from a borehole in gas.

Table 4-3: Radioactivity of C-14 associated with materials in wastes at 2090 AD for the Reference Case (Pöyry, 2010)

Material	Shielded wastes (TBq)	Unshielded wastes (TBq)	Total (TBq)
Graphite	6.04E+03	6.88E+01	6.10E+03
Stainless steel	1.89E+01	7.71E+02	7.90E+02
Other ferrous based alloys	1.09E+02	6.35E+01	1.72E+02
Zircaloy& zirconium		3.16E+01	3.16E+01
Nimonic (nickel based)alloys	-	2.38E+01	2.38E+01
Magnox alloys	7.84E-04	7.49E+01	7.49E+01
Uranium metal 80A	-	1.97E+01	1.97E+01
Magnox alloy corrosion products	-	2.14E+01	2.14E+01
Uranium metal corrosion products	-	9.62E+00	9.62E+00
Desiccant & ion exchange	-	8.90E+01	8.90E+01
Barium carbonate etc	1.10E-02	8.17E+00	8.18E+00
Total	6.16E+03	1.48E+03	7.64E+03

Mobility of Radionuclides

Both human intrusion and disruptive events have the potential to circumvent or degrade the natural and engineered barriers that are in place at the closure of the disposal vaults. Degraded barriers permit more rapid release and transport of contaminants through the barriers to environmental receptors. The more mobile contaminants are, the more significant they will be for such scenarios.

Cl-36 is long-lived and mobile. More than 70% of Cl-36 in the UK national ILW/LLW inventory is in graphite wastes (Pöyry, 2010). Furthermore, this radionuclide can be one of the more significant radionuclides under normal evolution conditions (see groundwater results in Table 4-1). It can therefore be anticipated that Cl-36 derived from graphite wastes could be an important determinant of the disposal system's performance should disruptive events result in the degradation of the barriers.

Aqueous C-14 can be mobile in geological environments. However, for cementitious concepts, the chemical conditioning of the disposal facility and its immediate environment acts to retain the radionuclide (see discussion in Section 3.1.1). The chemical conditions are unlikely to be strongly affected by any physical disruption to the repository or surrounding geology. This is because the alkaline conditions resulting from cementitious materials are not confined to the materials themselves, but will

result in a general raising of the pH in the vicinity of the material. On this basis, the retardation of the migration of C-14 in aqueous phase will be expected to remain, and the radionuclide is unlikely to be a significant contributor to radiological impacts.

C-14 in gas form may, however, be important under conditions in which the disposal system engineering is degraded. In particular, if pressurisation of the repository is possible, C-14 in gas could migrate via either a degraded shaft or an intrusion borehole. Such situations were found to be important in the assessment undertaken by Quintessa and SENES (2011) of the DGR in Canada and the scoping calculations by Hoch and Thorne (2008).

4.4 Quantitative Assessment

Consistent with the overall objectives of the study, the analysis of the consequences of human intrusion is based on existing assessment studies and reflects the current reference radionuclide inventory of graphite in the UK. The results can be scaled for other inventories (either total activity of disposed graphite, or the activity concentration of the material itself). The discussion in Section 4.2 and 4.3 indicates that intrusion as a result of drilling is likely to be the only credible scenario for most geological disposal facilities, but that the release of radionuclides may occur in various forms:

- waste retrieved as borehole core;
- gas released from a pressurised repository; and
- slurry (water and suspended solids) ejected from a pressurised repository.

Potential exposures to each of these media are discussed in the following sections.

4.4.1 Human Intrusion: Borehole Core

Limer et al. (2010) estimated the potential radiation exposure that may occur from exposure to graphite as a result of human intrusion. The assessment calculations made use of stylised exposure situations, based on assumptions made in previous assessments. The person intruding into the wastes was assumed to be exposed to drill core which contains raw wastes. Since Limer et al. (2010) was published the reference inventory of irradiated graphite in the UK has been updated, therefore the results calculated by Limer et al. (2010) have been scaled to the values presented in Appendix A, and are shown in Table 4-4. In this table, the radiation doses are calculated at times after emplacement (assumed to be 2090 AD). It can be expected that any repository would be subject to a period of institutional control of 100 – 300 y,

during which time inadvertent intrusion is not possible. Therefore doses are calculated for a number of different times after emplacement, noting that the peak dose would occur immediately following the end of the period of institutional control.

The calculated dose from graphite for this scenario is approximately an order of magnitude below the UK guidance level.

Table 4-4: Calculated radiation doses to an intruder in mSv y^{-1}

Radionuclide	100 y after emplacement*	300 y after emplacement*	500 y after emplacement*
H-3	0.0E+00	0.0E+00	0.0E+00
Be-10	1.1E-09	1.1E-09	1.1E-09
C-14	1.3E-01	1.3E-01	1.3E-01
Cl-36	1.1E-03	1.1E-03	1.1E-03
Ca-41	1.5E-07	1.5E-07	1.5E-07
Co-60	6.3E-07	1.2E-16	1.3E-16
Ni-59	6.3E-07	6.3E-07	6.4E-07
Ni-63	5.9E-05	1.4E-05	3.6E-06
Nb-93m	7.3E-10	1.5E-13	3.3E-17
Mo-93	5.1E-06	4.9E-06	4.7E-06
Ag-108m	1.1E-01	7.7E-02	5.5E-02
Sn-121m	1.4E-05	8.8E-07	5.5E-08
Ba-133	6.7E-08	1.3E-13	2.8E-19
Eu-152	1.5E-04	4.2E-09	1.3E-13
Eu-154	4.9E-07	4.9E-14	5.3E-19
Total	**2.4E-01**	**2.1E-01**	**1.8E-01**

* Time after facility backfilling, sealing and closure, will be less than time after emplacement.

4.4.2 Human Intrusion: Gas Release

The amount of C-14 potentially released in gas is highly dependent on the specific conditions in a disposal facility. The chemical conditions and materials present will determine the rate of evolution of gas, and the properties of the geosphere will also be relevant in determining the gas saturation and pressure that is reached. It is not, therefore, possible to generalise the scenario and quantitative estimates of impacts should be considered as being indicative of the possible consequences. However, based on the analysis presented in Section 3, it is possible to identify in which situations this

may be an important scenario. From least likely to be important to most likely to be important:

- Gas permeable EBS and fractured host rock – gas is readily released from the GDF, little free gas in the GDF, little or no pressurisation.
- Graphite wastes in isolation in (gas tight) lower strength sedimentary host rock – gas generation rate is low, but a free gas phase slowly develops and is trapped in the vault. Gas is at a significant pressure (hydrostatic or higher).
- Graphite wastes co-disposed with other ILW in (gas tight) lower strength sedimentary host rock. High bulk gas generation rate, gas is trapped in the vaults and there may be significant pressurisation (pressures greater than hydrostatic).
- As above, but concept with no backfill – there is no backfill to restrict the rate of gas flow into the intruding borehole.

It follows that the most appropriate basis for a conservative quantitative estimate is the situation assessed by NWMO, which is equivalent to the final situation indicated above (Quintessa et al., 2011). In this case, the disposal facility included a significant inventory of unconditioned wastes and was located in a very low permeability geological formation.

In the NWMO assessment, the total inventory of C-14 as disposed is 6.1 10^{15}Bq with the dominant source of C-14 being ion exchange resins (86%). Repository conditions are expected to lead to gas pressures that exceed hydrostatic pressure after a few tens of thousands of years. The peak concentration of C-14 in gas is about 6 10^{9}Bq m^{-3} after 3000 y.

Consistent with standard practices during drilling, the NWMO scenario assesses the managed release of the gas by the drillers, using a blowout preventer. This limits the rate of gas release to 1 m^3 s^{-1} and also means that the gas release could be for an extended period (in the NWMO case this could be more than a year). The drill crew can inhale released gas while working on the site and the exposure of nearby residents to dispersed gas is also assessed. The drill crew is assumed to work for 12 hours a day over 30 days. A nearby resident is assumed to live permanently 100 m from the point of gas release. Atmospheric dispersion is accounted for. The calculated radiation dose for the nearby resident is 0.1 mSv y^{-1} from gas release. For the drill crew the dose is 0.2 mSv y^{-1}.

The inventory of C-14 derived in Table 4-3 is the same (to two significant figures) as that considered by NWMO, and NWMO assume that C-14 is rapidly released from ion-exchange resins upon contact with highly saline water from the host rock. As noted

by Hoch and Thorne (2008), the dominant C-14 labelled gas anticipated to be produced from the disposal of the UK inventory of ILW/LLW wastes is methane, which is also similar to the DGR case. Given these similarities, the calculated doses may be indicative of those associated with UK graphite wastes under NWMO facility conditions.

Hoch and Thorne (2008) estimate that for the entire UK ILW/LLW inventory, including all sources of C-14, the total activity available for release as a gas after a few thousands of years is 30 TBq. This compares to a peak amount of C-14 in gas of around 3000 TBq for the DGR case. The much greater amount of C-14 in gas in the DGR case is a reflection that:

- The majority of the C-14 is found in resins in the NWMO inventory, rather than graphite in the NDA inventory. C-14 is released instantaneously from the unconditioned resins upon contact with highly saline water from the host rock.
- Ion-exchange resins are in the same vaults as organic wastes which drive methane generation and formation of $^{14}CH_4$, while for the UK case the majority of graphite and organic wastes are in separate vaults (SILW and UILW vaults respectively).

The much smaller amount of contaminated gas in the facility suggests that, for the UK graphite inventory the consequences could be of the order of micro Sieverts per year, more than two orders of magnitude below the lower end of the range of the performance guideline.

Hoch and Thorne (2008) also examined the consequences of release of $^{14}CH_4$ gas due to borehole intrusion into a pressurised GDF. They concluded that doses from an old site investigation borehole leaking $^{14}CH_4$ gas into a house would be approximately two orders of magnitude below the performance guideline.

4.4.3 Human Intrusion: Slurry and Solids Release

In addition to providing a pathway for the release of contaminated gas, a borehole that penetrates a geological disposal facility could also enable liquids and particulate material to be released from a disposal facility. This would occur if there were sufficient pressurisation at the repository horizon to permit a flow of fluid. A significant discharge of material would be most likely in a situation in which a facility was partially saturated and there is a significant over-pressurisation due to gas generation. Entrainment of particulate material would be possible for a GDF concept where there is no backfill, but would be less significant for a concept which includes backfill since particulate material would be filtered by the backfill.

A generally applicable set of exposure scenario assumptions (and also those likely to be most cautious) are described by Limer et al. (2010). These are derived from human intrusion calculations for near surface disposal facilities, in which considerable amounts of waste could be brought to the surface and ultimately become mixed with soil (e.g. excavation of a cutting for a road). Limer et al. (2010) examine an exposure scenario in which a person resides on contaminated soil containing 10% waste from the repository. It is assumed that 500 m^2 is contaminated, and the person ingests soil, contaminated crops and animal products, and is subject to external exposure and inhalation of dust.

The scenario assessed by Limer et al. (2010) leads to very significant doses (20 mSv y^{-1} or significantly greater) from C-14 and Cl-36, but this is a function of the amount of material assumed to be disposed of in the surface, and the amount of dilution in the soil. The most notable conservatism is the volume of waste assumed to be involved. Limer et al. (2010) assume volumes of 10 – 2000 m^3.

NWMO (Quintessa et al. 2010) conclude that the retrieval of large volumes of material is not feasible for a deep geological disposal facility, and adopt a value of 0.1 m^3 as a conservative value for waste (entrained in other media) released to the surface. This value assumes that there is no backfill, although the vaults and wastes may have been affected by rockfall.

The NWMO value has been used here, with the inventory data presented in Appendix A. It is assumed that the discharged material is spread over an area of 500 m^2 that is used for agriculture. The calculated doses that result are shown in Table 4-5. It is notable that the peak dose of 17 mSv y^{-1} is close to the performance guideline, and is dominated by Cl-36 and C-14.

The calculation presented above extrapolates the results of Limer et al. (2010) to the situation of a deep GDF. However, we also note the original context of the calculation, and the calculation results, which show that for a near-surface facility exposure to a relatively small volume of waste can lead to significant doses to a subsequent site occupier.

Table 4-5: Calculated radiation doses to a site residentresulting from ejection of waste (entrained in other media) from an intruding borehole, in mSv y^{-1}

Radionuclide	100 y after emplacement*	300 y after emplacement*	500 y after emplacement*
H-3	5.0E-06	6.3E-11	8.0E-16
Be-10	1.1E-08	1.1E-08	1.1E-08
C-14	5.3E+00	5.1E+00	5.0E+00
Cl-36	1.2E+01	1.2E+01	1.2E+01
Ca-41	3.4E-04	3.4E-04	3.3E-04
Co-60	5.5E-10	1.0E-19	1.1E-19
Ni-59	6.5E-05	6.5E-05	6.6E-05
Ni-63	4.9E-03	1.2E-03	3.0E-04
Nb-93m	8.9E-11	1.9E-14	4.1E-18
Mo-93	4.0E-04	3.8E-04	3.7E-04
Ag-108m	1.8E-04	1.3E-04	9.3E-05
Sn-121m	1.6E-06	1.0E-07	6.3E-09
Ba-133	6.6E-11	1.3E-16	2.7E-22
Eu-152	1.0E-07	2.9E-12	8.8E-17
Eu-154	3.4E-10	3.4E-17	3.7E-22
Total	**1.7E+01**	**1.7E+01**	**1.7E+01**

* Time after facility backfilling, sealing and closure, will be less than time after emplacement.

4.4.4 Human Intrusion: Summary of Quantitative Results

The quantitative results for human intrusion scenarios as summarised in Table 4-6. For many situations graphite wastes will not lead to doses in excess of the 3 mSv y^{-1} to 20 mSv y^{-1} guidance level, although the calculation results will be sensitive to the scenario assumptions. Doses may be more significant for some specific situations, for example occupation of a contaminated site following significant intrusion into a near-surface facility or ejection of contaminated water and solids from a pressurised facility.

Table 4-6. Summary of Quantitative Results for Human Intrusion Scenarios

Scenario	Release Form	Impact
Inspection of borehole core	Solid	Medium – doses approximately 1 OM below 3 mSv y^{-1}guidance level
Gas release from a pressurised facility over a 30 day period as a result of borehole intrusion (driller and local resident)	Gas	Low to Medium – doses 1 OM to 2 OM or more below 3 mSv y^{-1} guidance level
Chronic gas release from a leaky borehole into a house	Gas	Low – doses approximately 2 OM below 3 mSv y^{-1} guidance level
Site occupancy following significant intrusion into a near-surface facility (e.g. road cutting)	Solid	Very high – 20 mSv y^{-1} guidance level or significantly greater
Site occupancy following ejection of contaminated water and solids from an intruding site investigation borehole	Solid and water	High – approximately 20 mSv y^{-1} guidance level

4.4.5 Natural Disruption

As discussed in Sections 4.1.2 and 4.2.2, natural disruptive events are only likely to affect contaminant behaviour by decreasing the containment of contaminants by the geologic barriers. For example, the formation of a conductive fault as a result of an earthquake was considered by NWMO. Such a situation is highly dependent on both the specific characteristics of the geosphere and the nature of the disruptive event, and is therefore very difficult to generalise.

The required geosphere performance has previously been discussed in Section 3.1.2. All host rocks may be significant barriers for C-14, but only lower strength sedimentary and evaporite host rocks are likely to be significant barriers for Cl-36. Only in situations where the geosphere is a significant barrier will a more conductive feature be important. Therefore a disruptive event that leads to a more conductive feature, e.g. fault reactivation, may be significant for C-14 in any host rock, but is only likely to be significant for Cl-36 in lower strength sedimentary or evaporite host rocks.

It should be noted that disruptive events may only affect transport in water for a fraction of the radionuclide inventory, and may lead to localised releases where the probability of exposure is small compared with general transport from the whole footprint of the facility. Conversely, localised releases may lead to less dilution, and higher environmental radionuclide concentrations.

5 Conclusions

Key Messages

This report has examined the suitability of irradiated graphite wastes for geological disposal. Calculations in support of the analysis have been undertaken for the UK national inventory of irradiated graphite wastes, which comprises a significant volume of waste and a significant associated radionuclide inventory. Therefore conclusions that are drawn regarding the suitability of irradiated graphite wastes for geological disposal are relevant to large waste inventories, not only small quantities of irradiated graphite.

The analysis presented in this report has demonstrated that it should be possible to safely dispose of irradiated graphite wastes in isolation (i.e. in vaults containing only packages of graphite wastes) in a wide range of disposal systems (i.e. combination of disposal concept / EBS and geosphere); including near-surface, shallow and deep geological disposal and a wide range of host rocks. Assessment calculations show that regulatory guidelines can be satisfied even given conservative assessment assumptions. A broader range of systems might be suitable given less conservative calculation assumptions. One particular issue that potentially requires careful consideration, taking cognisance of national regulatory requirements, is the potential impacts associated with disruption of, or large scale intrusion into, near-surface facilities.

It may also be possible to safely dispose of irradiated graphite wastes in the same vaults as other ILW in a wide range of disposal systems. However, a broader range of processes become important, behaviour becomes more site / design specific and the important scenarios and behaviours may change as the system evolves. This makes it difficult to generically explore the suitability of graphite for geological disposal with other ILW. Specific waste types of concern are those that give rise to bulk gas generation (i.e. metals, organics, strongly irradiating wastes) and that might lead to incorporation of C-14 in methane gas (i.e. organics) and therefore increase the potential for generation and transport of C-14 labelled gases. The potential for transport of C-14 labelled gases is particularly important for fractured host rocks and potentially for lower permeability host rocks where the concept includes an Engineered Gas Transport System (EGTS) to limit the peak gas pressure.

Therefore, although it may not be necessary in all cases, there are advantages to disposing graphite wastes in isolation compared with co-disposal in the same vaults as other ILW. These include:

- simpler, more predictable behaviour;
- improved performance, e.g. transport in gas not likely to be an issue; and
- simplified safety arguments and safety case.

In the UK, the majority of the graphite waste (by volume and radionuclide inventory) is classified as Shielded ILW (SILW). SILW contains relatively little quantities of organic and reactive metals, which are largely associated with Unshielded ILW (UILW) wastes. Further segregation of graphite from other SILW should be relatively straightforward and might offer performance benefits, for example through the ability to optimise the disposal concept for graphite wastes.

If transport of C-14 in gas is of concern for segregated graphite waste packages, e.g. potentially in a fractured host rock, it is likely that further performance benefits would be obtained from disposing graphite in concrete containers rather than steel containers, thereby reducing bulk gas generation to a very low level. This would also reduce the potential for reduction of inorganic C-14 to $^{14}CH_{4(g)}$ by autotrophic bacteria, using H_2 derived from anaerobic corrosion of steel, to a very low level. The potential significance of this process for isolated packages of graphite waste in steel containers is uncertain, although the available information and expert judgements indicate that it is unlikely to be significant.

Residual Uncertainties

At the generic level of this report, factors that may limit the range of disposal systems suitable for geological disposal of graphite wastes include assumptions regarding the nature of the biosphere into which releases may occur, which tend to be more conservative for generic assessments such as those undertaken in this study; uncertainties regarding radionuclide release from graphite wastes; and cautious assumptions in assessment calculations.

Although significant work has been undertaken within CARBOWASTE to better understand radionuclide release from graphite, uncertainties remain regarding:

- release mechanism(s), the form(s) and release rate(s) of C-14 labelled gas(es) from dry, damp and wet graphite wastes;
- the mechanism(s) for long-term release of C-14 to water; and

- the potential for release of C-14 and Cl-36 in aqueous organic compounds, and their subsequent fate and transport.

Due to these uncertainties safety assessment calculations often include cautious assumptions that are likely to overestimate the amount of C-14 releasedand potentially the rate of C-14 release. It is difficult to represent the complex geochemical behaviour of C-14 in the near-field in simple safety assessment calculations. Consequently, such calculations tend to include cautious assumptions that underestimate the chemical barrier performance for concepts including a cementitious EBS.

These factors do not preclude safe disposal of graphite wastes in a wide range of disposal systems, but as stated above a wider range of disposal systems might be suitable given improved understanding of C-14 release and assessment models.

A second consequence of these remaining uncertainties, and how they are accounted for in assessments, is that it may be difficult to demonstrate that a disposal concept is optimised. This may mean that a cautious approach to engineering design may be required, potentially involving higher-specification near-field barriers than might be appropriate if waste guideline performance were better understood.

Summary of System Performance

A comprehensive analysis of the potential impacts arising from geological disposal of graphite wastes has been undertaken. The findings are summarised as follows.

The key radionuclides associated with irradiated graphite are C-14 and Cl-36. Transport in water (C-14 and Cl-36) and gas (C-14) are the key processes of concern in relation to the normal evolution of the geological disposal facility. Transport in gas is unlikely to be a significant issue for graphite wastes that are disposed in isolation from other wastes, and near-field and geosphere barrier properties that are consistent with performance for transport in water have been calculated.

Potential doses associated with human intrusion into a deep geological disposal facility containing graphite wastes are not insignificant. However doses associated with graphite wastes are likely to be lower than those associated with other waste types likely to be consigned to such a facility. This is because the dominant radionuclides for such scenarios (strong gamma emitters and alpha-emitters) are not present at significant concentrations in graphite. Therefore, external doses from examining borehole cores and associated situations are lower than for other ILW types.

Potential doses associated with human intrusion into graphite wastes are significantly greater for a near-surface facility where large scale excavation of solid material is

possible. Similarly, large scale disruption of a surface facility by natural processes and events could lead to significant doses to subsequent site occupiers, compared with regulatory guidelines. In both cases doses are due to uptake via the foodchain. Therefore for a near-surface facility, the significant potential for large scale human intrusion and the fate of exposed material could be important considerations in determining the acceptability of the concept.

If packages of graphite wastes are disposed in the same vault as packages of other ILW, the system behaviour may be more complex and hence less readily predictable than for a vault containing only packages of graphite wastes. For example, the presence of organic wastes may lead to methanogenic conditions being established upon their degradation, thus increasing the potential for incorporation of C-14 in methane gas and thereby increasing the potential mobility of C-14 and potential C-14 fluxes to the biosphere. In such cases, the individual waste packages may be important barriers to prevent interaction between graphite and organic wastes and incorporation of C-14 in methane. (It should be possible to maintain container integrity until methane generation from organic wastes has reduced to low levels).

With increasing organic and metal (particularly reactive metal) waste inventories in the disposal vaults, the potential total bulk gas generation rate will also increase (given certain assumptions, in particular assuming sufficient water is available to support corrosion). This has the potential to significantly increase the mobility of C-14 via a number of complex processes:

- in higher strength host rock, bulk gas acts as a carrier gas for C-14 labelled trace gases, e.g. by establishing a gas pathway to the surface or near-surface environment, including a shallow aquifer;
- in lower strength sedimentary host rock, bulk gas may lead to pressurisation and potential for fluids to be driven through the EDZ and any poorly performing seals; and
- potential for ejection of C-14 in water, slurry and particulates following intrusion into a pressurised GDF (see below).

Complex assessment models are required to help understand these coupled processes and behaviour of the system. The consequences of such complexity and associated uncertainty for design optimisation and requirements on barrier performance are as previously described for uncertainty in the radionuclide release mechanisms from the waste.

For graphite wastes, the highest potential doses from an intrusive event are associated with site occupancy following surface release of C-14 and Cl-36 from a borehole in

water, entrained particulates or slurry. Ejection of such material is more likely where there is significant gas generation leading to pressurisation in the vaults.

For deep geological disposal, natural disruptive events are very unlikely to significantly enhance the potential dose from graphite wastes. An event could potentially result in a more conductive groundwater pathway to the surface. However, the key radionuclide in graphite wastes for such a pathway, Cl-36, is sufficiently long-lived and mobile that, for many host rocks, an enhanced pathway is unlikely to significantly affect the peak dose or risk, just the time at which it may arise.

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Appendix A: UK Graphite Wastes and Associated Radionuclide Inventories

Table 6.16 of Adeogun et al. (2010) gives the C-14 activity in graphite at 2090 as 6.04E+03 TBq in SILW and 6.88E+01 TBq in UILW. Adeogun et al. (2010) does not give inventories for other radionuclides in graphite.

Limer et al. (2010) gives the inventories of C-14 and Cl-36 as 5960.63 and 24.79 TBq respectively, in 76,820.73 tonnes of graphite. Comparing against the data in Adeogun et al. (2010), the C-14 inventory has increased by the same factor (1.0295) as the mass of graphite. Applying this factor graphite inventory data of Limer et al. (2010) gives the inventories shown in Table A-1. Table A-2 gives the estimated radionuclide inventories in SILW graphite, assuming that the proportion in SILW is the same as C-14 for all radionuclides.

Table 7.15 of Adeogun et al. (2010) gives the total mass of graphite as 9,524.2 tonnes in UILW, 69,561.7 tonnes in SILW and 15,901.9 tonnes in LLW.

Adeogun (2010) gives the total waste and packaged volume of core graphite as 65,146m^3 and 97,186m^3 respectively. Adeogun (2010) states that there are 4,853 packages of core graphite, each containing 16.62 t of graphite, which gives a total mass of 80,655 t. Core graphite is mostly classified as SILW and is assumed to be disposed in RWMD 4 m boxes.

69,561.7 t SILW / 16.62 t per package = 4,186 SILW packages.

For the illustrative GDF designs considered in the DSSC (NDA RWMD, 2010a):

- In higher strength host rock, RWMD 4 m boxes will be stacked 3 wide by 5 high. There will be 4,186 / 15 = 279 rows in the vaults. The 4 m box width is 2.438 m, and row spacing is 0.9 m, so 3.338 m per row (NDA RWMD, 2010b).Each vault is 300 m long (usable disposal space), so 279 rows x 3.338 m per row / 300 m = 3.1 vaults.
- In lower strength sedimentary host rock, RWMD 4 m boxes will be stacked 1 wide by 3 high. There will be 4,186 / 3 = 1395 rows. Each vault is 100 m long (usable disposal space), and the row spacing is 0.45 m.1395 x (2.438 + 0.45) / 100 = 41 vaults.

Table A-1. Radionuclide inventories in UK graphite wastes

Radionuclide	Current Inventory (Bq) (Limer et al. 2010)	Revised Current Inventory (Bq)	Half-life (y)	Future Inventories (Bq at year AD)				
				2090	2190	2390	2590	3010
H-3	1.70E+15	1.75E+15	1.23E+01	1.94E+13	7.00E+10	9.08E+05	1.18E+01	6.44E-10
Be-10	4.40E+09	4.53E+09	1.51E+06	4.53E+09	4.53E+09	4.53E+09	4.53E+09	4.53E+09
C-14	N/A	N/A	5.70E+03	6.11E+15*	6.03E+15	5.89E+15	5.75E+15	5.46E+15
Cl-36	2.48E+13	2.55E+13	3.01E+05	2.55E+13	2.55E+13	2.55E+13	2.55E+13	2.55E+13
Ca-41	1.40E+13	1.44E+13	1.02E+05	1.44E+13	1.44E+13	1.44E+13	1.44E+13	1.43E+13
Co-60	3.80E+16	3.91E+16	5.27E+00	1.06E+12	2.05E+06	7.77E-06	2.94E-17	3.03E-41
Ni-59	2.50E+13	2.57E+13	1.01E+05	2.57E+13	2.57E+13	2.57E+13	2.56E+13	2.56E+13
Ni-63	2.70E+15	2.78E+15	1.00E+02	1.60E+15	7.99E+14	2.00E+14	5.01E+13	2.73E+12
Nb-93m	7.40E+11	7.62E+11	1.61E+01	2.45E+10	3.33E+08	6.17E+04	1.14E+01	1.66E-07
Mo-93	4.60E+11	4.74E+11	4.00E+03	4.67E+11	4.59E+11	4.43E+11	4.28E+11	3.98E+11
Ag-108m	7.70E+11	7.93E+11	4.18E+02	6.94E+11	5.88E+11	4.22E+11	3.03E+11	1.51E+11
Sn-121m	4.90E+12	5.04E+12	4.39E+01	1.43E+12	2.94E+11	1.25E+10	5.32E+08	7.01E+05
Ba-133	2.70E+11	2.78E+11	1.05E+01	1.43E+09	1.96E+06	3.72E+00	7.04E-06	6.75E-18
Eu-152	1.20E+13	1.24E+13	1.35E+01	2.06E+11	1.23E+09	4.40E+04	1.57E+00	7.23E-10
Eu-154	6.50E+12	6.69E+12	8.59E+00	1.05E+10	3.31E+06	3.26E-01	3.21E-08	6.22E-23

* Adeogun et al. (2010).

Table A-2. Estimated radionuclide inventories in UK SILW graphite wastes

Radionuclide	Revised Current Inventory (Bq)	Future Inventories (Bq at year AD)				
		2090	2190	2390	2590	3010
H-3	7.13E+15	1.92E+13	6.92E+10	8.98E+05	1.16E+01	6.37E-10
Be-10	4.48E+09	4.48E+09	4.48E+09	4.48E+09	4.48E+09	4.48E+09
C-14	6.04E+15	6.04E+15*	5.97E+15	5.82E+15	5.68E+15	5.40E+15
Cl-36	2.52E+13	2.52E+13	2.52E+13	2.52E+13	2.52E+13	2.52E+13
Ca-41	1.43E+13	1.42E+13	1.42E+13	1.42E+13	1.42E+13	1.42E+13
Co-60	3.87E+16	1.04E+12	2.03E+06	7.68E-06	2.91E-17	3.00E-41
Ni-59	2.54E+13	2.54E+13	2.54E+13	2.54E+13	2.53E+13	2.53E+13
Ni-63	2.75E+15	1.58E+15	7.90E+14	1.98E+14	4.95E+13	2.70E+12
Nb-93m	7.53E+11	2.42E+10	3.29E+08	6.10E+04	1.13E+01	1.64E-07
Mo-93	4.68E+11	4.62E+11	4.54E+11	4.38E+11	4.23E+11	3.94E+11
Ag-108m	7.84E+11	6.86E+11	5.82E+11	4.17E+11	3.00E+11	1.49E+11
Sn-121m	4.99E+12	1.41E+12	2.91E+11	1.24E+10	5.26E+08	6.93E+05
Ba-133	2.75E+11	1.41E+09	1.94E+06	3.68E+00	6.96E-06	6.67E-18
Eu-152	1.22E+13	2.03E+11	1.22E+09	4.35E+04	1.56E+00	7.15E-10
Eu-154	6.62E+12	1.04E+10	3.27E+06	3.22E-01	3.18E-08	6.15E-23

* Adeogun et al. (2010).

A.1 References

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Appendix B: Model for Calculation of Guideline Performance Metric (λ_2)

A GoldSim model was used to calculate the disposal system performance such that the peak dose from each radionuclide is equal to 17 µSv y^{-1} (peak conditional risk of 1E-6 y^{-1}). The disposal system performance is expressed in terms of the transfer rate which results in a peak dose of 17 µSv y^{-1}.

The model structure comprises ten compartments (Figure B-1), which represent both the GDF and the geosphere, and a sink compartment (the biosphere). The near-field and geosphere are represented in the model so combinations of near-field and geosphere barrier performance can be explored. Five compartments are used to represent each of the near-field and geosphere in order for the numerical dispersion to be similar to the amount of dispersion that might be expected in reality (Appendix F in Quintessa, 2011).

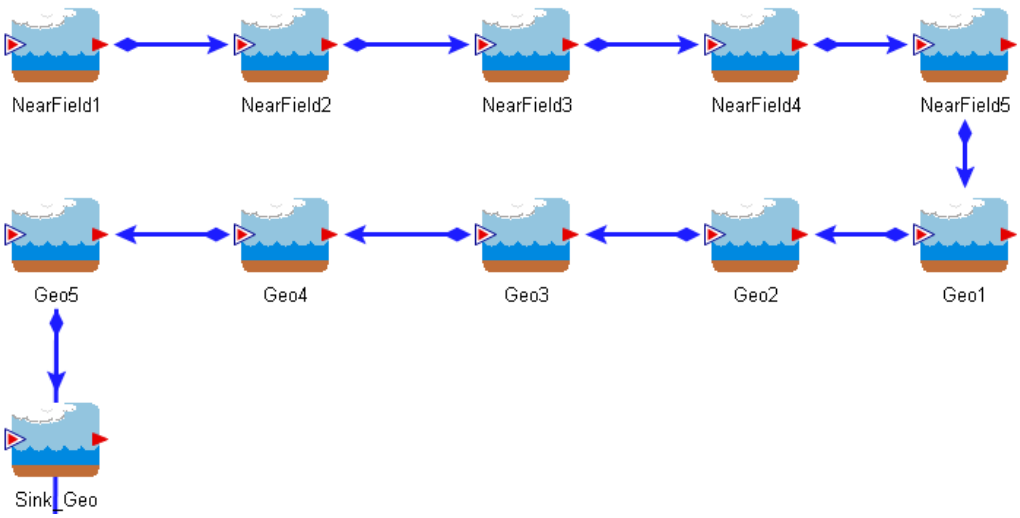

Figure B-1. Structure of the GoldSim model to calculate the required disposal system performance

Calculations were undertaken for the radionuclide inventory associated with UK SILW graphite. The inventory was assigned to the first compartment (NearField1). Radionuclides are transferred between compartments using fractional rates (y^{-1}), with one rate used for the near field compartments (λ_{NF}) and a different rate used for the geosphere compartments, including the transfer to Sink_Geo (λ_{Geo}).

The transfer rates are specified as log uniform Probability Density Functions (PDFs) with rates from 1E-9 y^{-1} to 10 y^{-1}, i.e. very good barrier performance to very poor barrier performance. (A transfer rate of 10 y^{-1} means that the travel time through the barrier is much less than one year). The model was run stochastically in order to

calculate doses for different combinations for near-field and geosphere barrier performance.

Doses were calculated using flux-dose conversion factors from the RWMD generic biosphere model for transport in water (Thorne, 2007), assuming exposure pathways associated with abstraction of contaminated well water only; and flux-dose conversion factors from the Enhanced RIMERS model (Thorne, 2006) for transport in gas.

Results are of the form shown in Figure B-2 and are given in Table 2-4. The transfer rate which results in a peak dose of 17 μSv y^{-1} is the same regardless of whether the barrier function is provided by the near-field, geosphere or a combination of the two barriers.

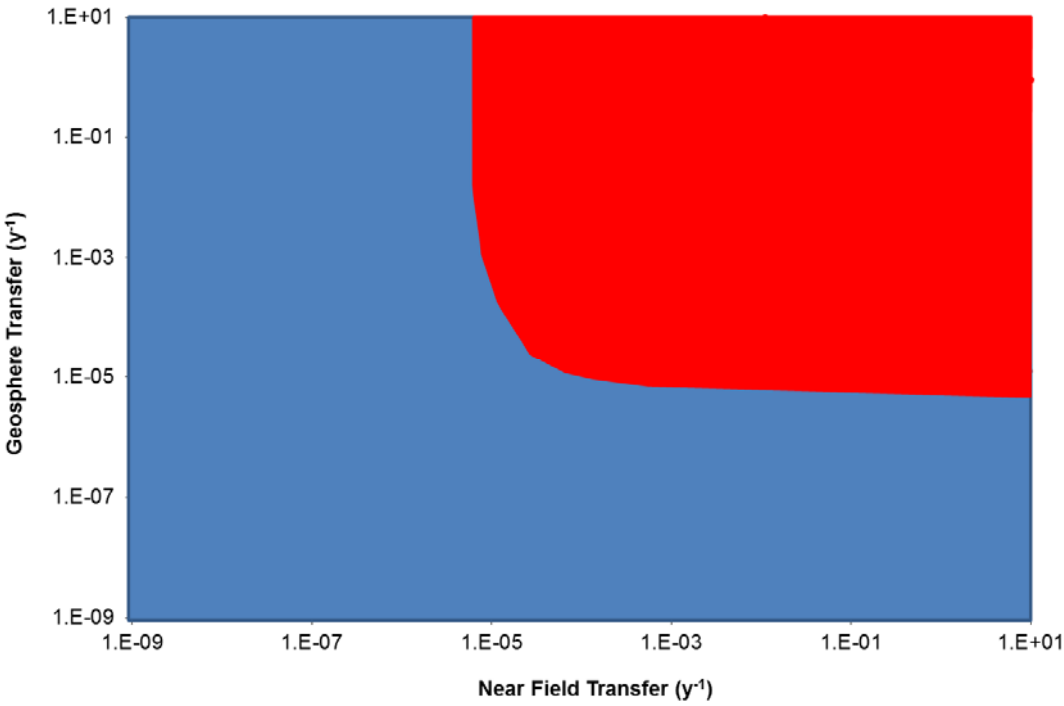

Figure B-2. Combinations of near-field and geosphere transfer rates that give rise to doses >17 μSv y^{-1} (red) and <17 μSv y^{-1} (blue) for transport of C-14 in water from the UK SILW graphite inventory

B.1 References

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Appendix C: Near-Field Barrier Model

A GoldSim model was used to examine the near-field barrier performance. The calculation cases to be explored using the model are described in Table C-1. The model calculation cases consider higher strength host rock and lower strength sedimentary host rock, and the associated illustrative disposal concept examples described in the gDSSC (NDA RWMD, 2010a).

Calculations were undertaken for the radionuclide inventory associated with UK SILW graphite.

Table C-1. Near-field barrier performance calculation cases

Case	Waste	Encapsulant	Container		Backfill	Host Rock*		Notes
	SILW	Cement	No	Yes		HSR	LSR	
1a	X	X		X	NRVB	X		Advective flow through backfill and geosphere. Advective flow through waste packages once container has failed.
1b – No container	X	X	X		NRVB	X		
2a	X	X		X	Nagra Mortar		X	Diffusive transport only.
2b – No container	X	X	X		Nagra Mortar		X	

HSR – Higher Strength Host Rock, LSR – Lower Strength Sedimentary Host Rock.

C.1 Model Options

Two option switches are used to implement the calculation cases within a single model.

Select_HostRock

0 = Higher Strength Rock

1 = Lower Strength Sedimentary Rock

Select_Container

0 = No Container

1 = Includes container (but container is assumed to fail by uniform corroded)

C.2 Model Configuration

The objective of the model is to assess the performance of each of the near-field barriers. Each of the barriers therefore needs to be represented explicitly in the model (Figure C-1).

Performance is calculated in terms of the radionuclide transfer rate through each barrier, i.e. the radionuclide flux from the compartment representing the barrier, divided by the amount of that radionuclide in the compartment. This transfer rate can then be compared against the required system performance calculated using the model described in Appendix B.

Figure C-1. Configuration of the GoldSim near-field model

Waste_Source

The inventory is assigned to the Waste_Source compartment. Radionuclides are transferred to the Wasteform compartment through leaching. Empirical rates from leaching experiments are used in the model.

Wasteform

The wasteform comprises the cement encapsulant and the container. Releases of radionuclides from the wasteform to the immediately adjacent backfill (Adj_Backfill compartment) are diffusive through the container vent until the container is perforated by uniform corrosion. Once the container has been perforated, or if the container is not represented in the model, radial diffusion from the wasteform and flow of water through the encapsulant are possible. (Flow of water through the near-field is only considered for higher strength host rock).

It is assumed that there is no pit corrosion (associated with oxic conditions during the operational phase and early post-closure phase) of the container that would permit small advective flows through the container. Under anaerobic conditions there is uniform corrosion of the containers and therefore the containers are assumed to be completely perforated instantly at a specific time. Corrosion is assumed to proceed at

equal rates from the inside and outside of the vented containers, i.e. the water required for anaerobic corrosion to proceed is able to enter the container through the vent.

Flow through the GDF is partitioned between the wasteform and backfill, accounting for the relatively low permeability of wasteform compared with the backfill.

Adjacent Backfill (Adj_Backfill)

The backfill is discretised into adjacent backfill and peripheral backfill in order provide a better reflection of the geometry of the system, i.e. explicit representation of the diffusion length through the waste stacks to the peripheral backfill.

Adjacent backfill is the immediately adjacent backfill around the containers. Transport to the peripheral backfill (Perip_Backfill container) is advective and diffusive. NRVB backfill is relatively permeable compared with the host rock, and its permeability is not considered to change with time.

Peripheral Backfill (Perip_Backfill)

This is the vault floor, backfill between the waste stacks and vault walls, and backfill in the crown space. Similar considerations to the adjacent backfill apply.

Surrounding host rock (Surrounding_Rock)

The host rock is a sink for radionuclide transport from the near-field.

C.3 Mathematical Models

C.3.1 Contaminant Transport

Solubility limitation, sorption, advective and diffusive transport are modelled using the inbuilt functionality within the GoldSim Contaminant Transport (CT) module. Advective flow rates are calculated using the mathematical model detailed in Section C.3.2.

C.3.2 Flows

Flow Through the Vaults

The total flow rate through the vaults is given by:

$$H_v(W_v + W_r)N_v qY$$

Where :

H_v [m]	Is the height of the vault;
W_v [m]	Is the width of the vault;
W_r[m]	Is the width of the rock pillar between adjacent vaults;
N [-]	Is the number of adjacent graphite vaults along a line perpendicular to the groundwater flow direction;
q [ms^{-1}]	Is the Darcy velocity in the host rock; and
Y [-]	Is the flow focussing factor. This describes the effect of the hydraulic conductivity contrast between the GDF and host rock on the pattern of groundwater flows.

For all cases it is assumed that the vaults are fully saturated.

Flow through the Waste Packages and Backfill

There will be no flow through the waste packages until the containers are perforated. All the flow through the vaults will be in the backfill. Once the containers have been perforated there will be flow through the waste packages. The waste packages and backfill may have different hydraulic conductivities. The flow through the waste packages will be lower than through the backfill. The proportion of the flow in the backfill that passes through the waste packages is given by:

$$A_w K_w / (A_w K_w + A_B K_B)$$

where :

A_w [m^2]	Is the cross-sectional area of the waste packages perpendicular to the flow direction;
K_w [ms^{-1}]	Is the hydraulic conductivity of the waste packages;
A_B[m^2]	Is the cross-sectional area of the backfill perpendicular to the flow direction; and
K_B[ms^{-1}]	Is the hydraulic conductivity of the backfill.

Similarly the flow through the adjacent and peripheral backfill is proportioned based on the relative areas.

C.4 Data

Raw data are presented below. Derived data, for example waste package surface area, are not presented. N.B. Where data are referenced to NDA RWMD (2010c) they were taken directly from RWMD's DSSC GoldSim model.

C.4.1 Radionuclide Leaching Rates

Radionuclide	Value	Notes
C-14	See Table 2-1	Long term leach rate of 1.83E-5 y^{-1} used.
Cl-36	See Table 2-1	-

C.4.2 Container Data (NDA RWMD 4 m box)

Parameter	Value	Reference
Length	4.013 (m)	NDA RWMD (2010b)
Width	2.438 (m)	NDA RWMD (2010b)
Height	2.200 (m)	NDA RWMD (2010b)
Thickness	3 (mm)	S3.1 of Nirex (2005)
Spacing	0.9 (m) in HSR 0.45 (m) in LSR	NDA RWMD (2010a) Towler et al. (2012)
Payload volume (with no shielding)	18.9 (m^3)	Adeogun et al. (2010)
External (displacement) volume	20.0 (m^3)	Adeogun et al. (2010)
Encapsulant conditioning fraction	0.5	Estimated value assuming graphite blocks surrounded by an annulus of encapsulant
Vent area	0.48 (m^2)	Cautiously assumed to be 1% of total surface area

C.4.3 Corrosion Rate

Backfill	Corrosion Rate (m/y)	Reference
Cementitious	1E-8	Table 4-7 of Towler et al. (2010)

C.4.4 Vault Dimensions Higher Strength Rock

Parameter	Value	Reference
Vault height (SILW vault)	15 (m)	NDA RWMD (2010a)
Vault width	16 (m)	NDA RWMD (2010a)

Vault length	300 (m)	NDA RWMD (2010a)
Number of vaults (4 m boxes)	3.1	Appendix A
Vault spacing	75 (m)	Towler and Bond (2011)
HHISOs in stacks	3 wide x 5 high	Appendix A

C.4.5 Vault Dimensions LSR

Parameter	Value	Reference
Vault height	11.5 (m)	NDA RWMD (2010a)
Vault width	9.6 (m)	NDA RWMD (2010a)
Vault length	100 (m)	NDA RWMD (2010a)
Number of vaults (4 m boxes)	41	Appendix A
Vault spacing	30 (m)	Towler and Bond (2011)
HHISOs in stacks	1 wide x 3 high	Appendix A

C.4.6 Encapsulant Physical Properties

Porosity (-)	Tortuosity (-)	Density (kg m^{-3})	Reference
0.2	0.0127	2100	Porosity and density assumed. Tortuosity to give De for StII structural concrete in Savage and Stenhouse (2002).

C.4.7 Backfill Physical Properties

Backfill	Density (kg/m^3)	Porosity (-)	Tortuosity (-)	Reference
NRVB	1200	0.55	0.15	Density and porosity from NDA RWMD (2010c). Tortuosity from Towler and Bond (2011).
Nagra Mortar	1650	0.3	0.15	Nagra (2008), assume tortuosity as NRVB.

C.4.8 Encapsulant and Backfill Transport Properties

Element	Kd cement (m^3/kg)	Anion exclusion factor	Reference
C	0	0.2	Kd from Table 3-3, exclusion factors from NDA RWMD (2010c).
Cl	0.05	0.2	

C.4.9 Near-Field Solubility Limits

Element	Solubility limit cement (mol/l)	Reference
C	8.61E-6	Table 3-4
Cl	Not solubility limited	-

C.4.10 Host Rock Physical Properties

Host Rock	Density (kg/m³)	Porosity (-)	Tortuosity (-)	Reference
Higher Strength	2700	0.01	0.5	NDA RWMD (2010c), tortuosity - Towler and Bone (2011)
Lower Strength Sedimentary	2450	0.15	0.1	Density Fig. 3-4 of Nagra (2008). Porosity and Tortuosity, Towler and Bond (2011).

C.4.11 Host Rock Transport Properties

Sorption distribution coefficients

Element	Kd HSR (m³/kg)	Kd LSR (m³/kg)
C	5.99E-4 (PDF mean value from NDA RWMD, 2010c)	6.7E-5 (Table 5.6 Bradbury and Baeyens, 2010)
Cl	0 (NDA RWMD, 2010c)	0 (Table 5.6 Bradbury and Baeyens, 2010)

Anion exclusion factors

Element	Anion exclusion factor HSR	Anion exclusionfactor LSR
C	1 (NDA RWMD, 2010c)	0.4 (NDA RWMD, 2010d)
Cl	1 (NDA RWMD, 2010c)	0.4 (NDA RWMD, 2010d)

C.4.12 Flow Properties

Parameter	Value	Reference
q	6E-4 (m/y)	NDA RWMD (2010c)

K_{NRVB}	1E-8 (m/s)	Towler and Bond (2011)
$K_{WASTE_PACKAGE}$	1E-10 (m/s)	Assumed
Y_{NRVB}	31.3	NDA RWMD (2010c)

C.5 References

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Appendix D: Calculation of Geosphere Properties Consistent with Guideline Performance for Advective Transport in Water

Calculations were undertaken using an Excel spreadsheet for the radionuclide inventory associated with UK SILW graphite. The peak radionuclide flux from the GDF was calculated as:

$J = kiI/H\varnothing_{nf}$

Where

J is the radionuclide flux (Bq y^{-1})

k is the hydraulic conductivity of the geosphere (m y^{-1})

i is the hydraulic gradient in the geosphere (-)

I is the radionuclide inventory (Bq)

H is the height of the waste stacks in the GDF (m): taken to be 11 m (Figure 3-1)

$\varnothing_{nf}$ is the porosity of the near-field

This calculation assumes that flow through the vaults is vertical, which maximises the release rate, consistent with the calculation assumption of minimal credible influence of the near-field on radionuclide transport. It was assumed that there is not any significant enhancement of flow through the vaults by flow focussing.

The ratio of the radionuclide flux consistent with the guideline performance (Table 2-4) to the peak flux from the near-field (calculated using the equation above) was calculated. From this ratio and the radionuclide half-life, the travel time required for the flux to decay to that to that consistent with the guideline performance, using the formula:

$A/A_0 = e^{-\lambda t}$

Where

A is the radionuclide flux consistent with the guideline performance (Bq y^{-1})

A_0 is the peak radionuclide flux from the near-field (Bq y^{-1})

λ is the radionuclide decay constant (y^{-1})

t is the time for decay (geosphere travel time) (y)

The required geosphere pathlength was then calculated from the travel time:

$p = tki/\varnothing_{geo}$

p is the pathlength (m)

$ø_{geo}$ is the porosity of the geosphere

The calculations are undertaken in terms of the geosphere hydraulic conductivity to porosity ratio (see results presented below). It is therefore necessary to make an assumption about the ratio of the porosity in the near-field to the porosity in the geosphere. This may vary significantly with the host rock, but consistent with the indicative nature of the calculations, for the figures presented in the main text of the report it was assumed that the porosity of the near-field is five times that of the geosphere; which for NRVB backfill (porosity = 0.55) implies a geosphere porosity of 0.11. As the ratio increases the required geosphere pathlength decreases, so a factor of five is conservative for fractured rock. This is illustrated in the following figures (D-1 and D-2).

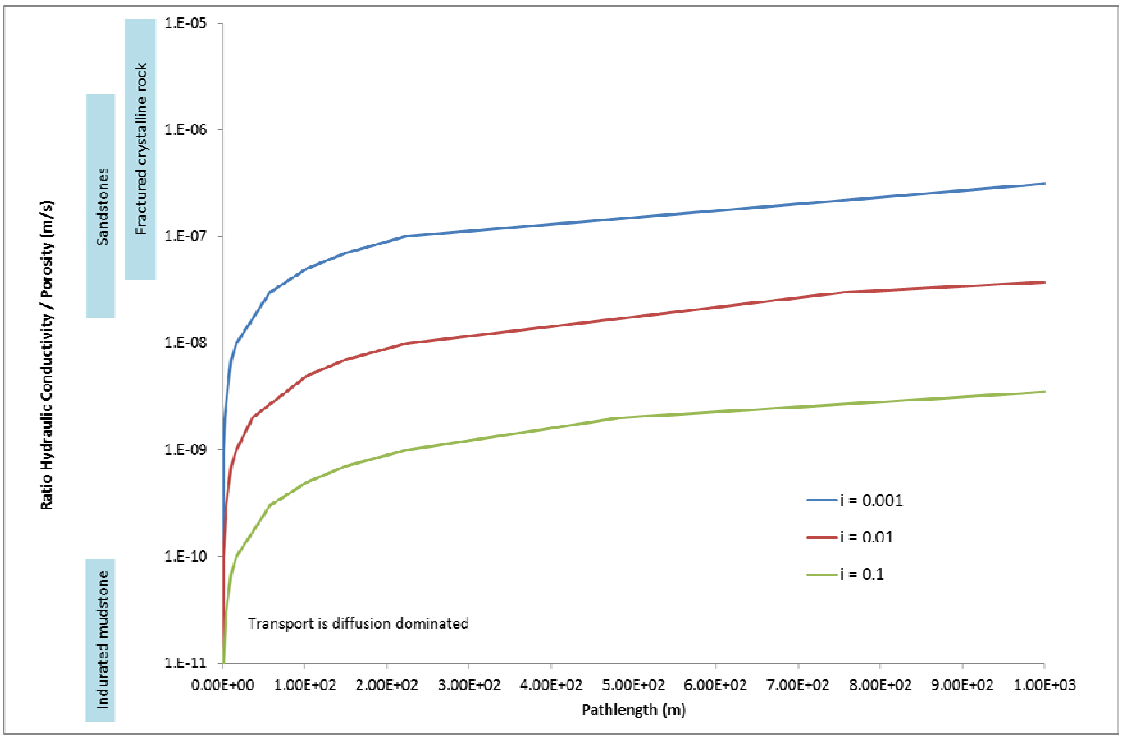

Figure D-1. Geosphere properties to achieve performance metric for C-14 where transport is advection dominated (based on radionuclide inventory associated with UK SILW graphite wastes, near-field/geosphere porosity ratio = 5). i = hydraulic gradient.

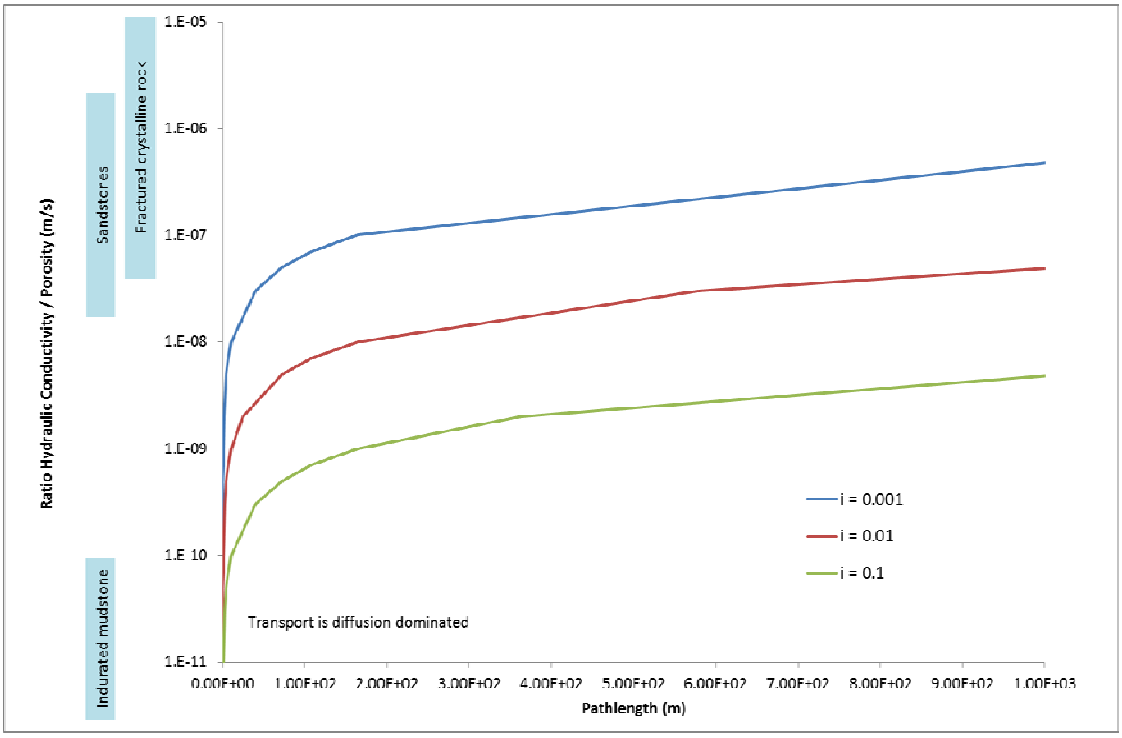

Figure D-2. Geosphere properties to achieve performance metric for C-14 where transport is advection dominated (based on radionuclide inventory associated with UK SILW graphite wastes, near-field/geosphere porosity ratio = 50). i = hydraulic gradient.

Appendix E: Calculation of Geosphere Properties Consistent with Guideline Performance for Diffusive Transport in Water

A simple GoldSim model comprising 7 cells (Figure E-1) was used to calculate 1D diffusive fluxes. The first cell represents the GDF, the next 5 cells represent the geosphere, and the final cell is a large volume sink cell, which represents the biosphere or overlying permeable formations, in which radionuclides are rapidly transported and dispersed compared with the underlying rock, i.e. a zero concentration boundary. Each geosphere cell is of the same volume and represents one fifth of the total geosphere pathlength.

The model was run for the UK national inventory associated with SILW graphite, and was parameterised based on the RWMD GDF concept example for lower strength sedimentary host rock. The model parameters are detailed in Table E-1.

The model was run for 2000 realisations in order to calculate the flux from the geosphere to the sink cell for varying pathlength and tortuosity. Tortuosity was varied between 1 and 1E-3 (-) in order to give effective diffusivities typical of the values measured in international disposal programmes (Table 3-12). The pathlength is equal to the thickness of low permeability host rock overlying the GDF. Conservatively, downwards diffusion into the rock below the GDF was not considered.

TableE-1. Diffusion model parameters

Parameter	Value	Reference
GDF Footprint	0.33 (km^2)	Estimated from Figure 34 of NDA RWMD (2010) for SILW area
Vault Height	11.5 (m)	NDA RWMD (2010)
GDF Volume	8.5E5 (m^3)	9.6 m x 11.5 m x 100 m x 77 vaults (Appendix C of NDA RWMD 2010)
GDF porosity	0.3 (-)	Representative value, e.g. for Nagra structural mortar (Nagra, 2008)
Rock porosity	0.15 (-)	Representative value
Geosphere pathlength (m)	10 m to 300 m	Uniform PDF
Rock Tortuosity (-)	1E-3 to 1 (-)	Uniform PDF

FigureE-1. Configuration of the GoldSim 1-D diffusion model

E.1 References

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