United Kingdom Nirex Limited

WASTE PACKAGE SPECIFICATION AND GUIDANCE DOCUMENTATION

WPS/902: Guidance Note on the Packaging of Radon-generating Wastes

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August 2005

WASTE PACKAGE SPECIFICATION AND GUIDANCE DOCUMENTATION

GUIDANCE NOTE ON THE PACKAGING OF RADON-GENERATING WASTES

This document forms part of a suite of documents prepared and issued by Nirex to assist waste packagers condition and package Intermediate Level and certain Low Level radioactive wastes.

The Waste Package Specification and Guidance Documentation (WPSGD) is based on, and is compatible with the Generic Waste Package Specification (GWPS) and therefore provides specification and guidance on waste packages that meet the transport and disposability requirements derived for the Nirex Phased Geological Repository Concept.

The WPSGD is intended to provide a 'user-level' interpretation of the GWPS to assist waste packagers in the early development of plans and strategies for the management of radioactive wastes. Waste packagers are advised to contact Nirex at an early stage to seek detailed assessment of specific packaging proposals.

The WPSGD will be subject to periodic revision and waste packagers are advised to contact Nirex to confirm that they are in possession of the latest version of documentation.

This document has been compiled on the basis of information obtained by Nirex. The document was verified in accordance with arrangements established by Nirex that meet the requirements of ISO 9001. The document has been fully verified and approved for publication by Nirex.

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1 INTRODUCTION

Nirex was established in 1982 with an objective of assisting producers of intermediate level (ILW) and long-lived low level radioactive waste (LLW) to package those wastes in a form compatible with disposal in an underground repository.

Nirex has fulfilled this objective by developing a long-term management concept, the Phased Geological Repository Concept (PGRC) [1], and by developing standards and specifications for the packaging of waste based on this concept. This is important because radioactive wastes in unconditioned form can pose a significant hazard to people and the environment and Nirex packaging standards have been designed to improve the safety and long-term behaviour of the wastes.

The mission of Nirex was strengthened in 2004 and agreed with Government as follows:

> *'In support of Government policy, develop and advise on safe, environmentally sound and publicly acceptable options for the long-term management of radioactive materials in the UK.'*

Four objectives have been set to determine the scope and manner of implementation of this mission and one of these requires that Nirex set standards and specifications for the packaging of waste, and advise waste packagers on how to treat and package radioactive waste in accordance with those standards and specifications, through the Letter of Compliance (LoC) process[1].

In order to facilitate the safe and efficient packaging, transport and disposal of waste, Nirex has defined packaging standards and specifications based on the requirements of the PGRC, involving transport of waste to a phased geological repository, monitored and retrievable underground storage with the option to seal and close the repository in the long-term.

The PGRC is underpinned by a suite of documents, including the Generic Waste Package Specification (GWPS) [2]. The GWPS defines and describes the packaging standards and specifications that have been derived from the PGRC and is used in the UK as the basis for the packaging of ILW and certain LLW.

The GWPS is the primary document defining Nirex packaging standards and specifications and is supported by the Waste Package Specification and Guidance Documentation (WPSGD). The WPSGD comprises a suite of documentation primarily aimed at waste packagers, its intention being to present the generic packaging standards and specifications at the user level, together with explanatory material and guidance that users will find helpful when it comes to application of the specification to practical packaging projects. For further information on the extent and the role of the WPSGD, reference should be made to the *Introduction to the Nirex Waste Package Specification and Guidance Documentation, WPS/100*[2].

The diverse physical, chemical and radiological nature of Intermediate Level Waste (ILW) and Low Level Waste (LLW) in the UK means that particular challenges arise in the packaging of certain wastes. To assist waste packagers with the preparation of proposals for the packaging of such wastes, Nirex has produced, and continues to add to, a suite of documents known as Guidance Notes. A full list of the Guidance Notes produced by Nirex, together with a abstract of each, can be found in *Introduction to Nirex Waste Packaging Guidance Notes, WPS/900.*

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[1] Formerly known as the Letter of Comfort process.

[2] Specific references to individual sections of the WPSGD are made in this document in *italic script,* followed by the relevant WPS number.

The release of radioactive gases from waste packages poses a challenge, and will need to be defined and addressed to enable safety cases for the different phases of the PGRC to be made. Consequently, for some wastes, restriction of the release of radioactive gases through waste treatment or waste package design may be required.

Radon is one of a number of radioactive gases that can be released from packaged waste and, due to its mobility, radio-toxicity and the manner in which it is continually generated, by the radioactive decay of its parent, it presents particular concerns. Fortunately, the relatively short half-lives of its isotopes mean that its release from waste packages is amenable to mitigation through appropriate packaging. In particular, the rate of radon release can be reduced by radioactive decay during migration through the barriers presented by the packaging. This Guidance Note is intended to assist waste packagers with the presentation of robust arguments regarding the performance of packaging, and to facilitate the safe and efficient packaging of radon-generating wastes.

2 SCOPE OF THE GUIDANCE NOTE

The development of suitable packaging methods for radon-generating wastes, and the demonstration of the suitability of a proposed method, requires attention to particular aspects of waste package and wasteform performance. Therefore, this document identifies the issues arising from the generation of radon by waste packages, their implications to the PGRC, the methods that could be used to develop robust arguments, and the supporting information that could be used in presenting the case to Nirex. Appendix A is a glossary of the terminology adopted in this Guidance Note. A brief summary of available data to support the development of arguments by waste packagers is included as Appendix B.

3 RADON AND ITS SOURCES

Radon is a gaseous element (boiling point -62°C), it is the densest of the elemental gases (density approximately ten times that of air) and an inert gas. It exists as 29 isotopes with half-lives ranging from a fraction of a microsecond to a few days. The predominant isotope in nature is radon-222 which is the progeny of radium-226, a member of the uranium-radium series derived initially from uranium-238. Information on the isotopes of significance in the packaging of ILW and LLW is shown in Table 1.

3.1 Radon-222 (radon)

Radon-222 is the longest lived isotope of radon and is produced by the radioactive decay of radium-226. Wastes that contain radium-226 therefore will generate radon-222 for as long as the parent radium-226 remains; 1TBq of radium-226 produces 7.5GBq of radon-222 per hour. As the half-life of radium-226 is 1600 years, radon-222 will continue to be produced well beyond the operational phase of the PGRC and, in the longer term, radon-222 will also be generated by radium-226 arising from the decay of uranium-238 (half-life 4.5×10^9 years) disposed of in the repository as well as that occurring naturally in the surrounding rock.

Radon-222 is an α-emitter, decaying to polonium-218 and a variety of other radionuclides, before eventually decaying to stable lead-206. The dose consequences of radon progenies also need to be considered during any assessment of the radiological impact of radon generation and release.

Table 1 Information on significant radon isotopes

Isotope	Decay mode(s)	Half-life	Specific Activity (Bq/m^3)	Parent(s)
Rn-219	α,γ	3.96s	4.7×10^{12}	Ra-223[3], At-219
Rn-220	α	55.6s	3.3×10^{11}	Ra-224[4]
Rn-222	α	3.825d	5.6×10^{7}	Ra-226[5]

3.2 Other Isotopes of Radon

Two other isotopes of radon are produced by the decay of radionuclides which are present in some waste in significant quantities.

Radon-220 is generated by the decay of radium-224. However, as the half-life of radium-224 is short (3.66 days), radon-220 may be considered a progeny of thorium-232 (half-life 1.4×10^{10} years), and is accordingly often referred to as thoron. However, the inventory of radon-220's immediate precursors in waste is commonly small and, as the half-life of radon-220 (56 seconds) is significantly shorter than that of radon-222, measures to mitigate the release of radon-222 will be at least as effective in the case of radon-220.

Similar arguments can be applied to radon-219 (half-life 4 seconds), a progeny of radium-223 (half-life 11.2 days) and uranium-235 (half-life 7.0×10^{8} years).

This Guidance Note therefore focuses on radon-222, hereafter referred to as radon.

3.3 Presence of Radium-226 in Wastes

Radium-226 is identified or reported to be present in a wide range of radioactive waste streams, the total quantity identified in the 2001 UK Radioactive Waste Inventory [3] being 17.8TBq as at 2040. Table 2 lists the total quantities of radium-226 in wastes destined for the Nirex repository together with the radon production rates from these wastes.

This shows that the vast majority of the radium-226 inventory is to be found in wastes to be packaged in unshielded ILW (UILW) packages, and that mean radon generation rates from shielded ILW (SILW) and LLW will be several orders of magnitude less than those from UILW.

The mean values for UILW are somewhat misleading and a closer examination of the Inventory data shows that ~95% of the total radium-226 inventory is contained in two waste streams which comprise only ~0.3% of the total conditioned volume of UILW. Figure 1 better illustrates the distribution of radium-226 throughout the Inventory showing that only ~2% of UILW packages contain more than the mean value given in Table 2.

The Inventory currently lists only two radium-226 bearing waste streams currently intended for packaging as SILW; mean radium-226 inventories for these streams are of the order of 10^{-11}TBq/m^3.

The problem of the packaging of radium-226 is, therefore, limited to a relatively small number of high-inventory UILW packages.

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[3] U-235 Series

[4] Th-232 Series

[5] U-238 Series

Table 2 Quantities of Radium-226 and generation rates for Radon-222 in ILW and LLW

Waste Type	Conditioned Volume	Total Radium-226 Inventory at 2040	Total Radon-222 Generation Rate	Mean Radium-226 Concentration	Mean Radon-222 Generation Rate
	m^3	TBq	TBq/hr	TBq/m^3	$TBq/hr/m^3$
UILW[6]	151,000	1.78×10^{1}	1.3×10^{-1}	1.2×10^{-4}	8.6×10^{-7}
SILW[7]	14,300	6.61×10^{-9}	5.0×10^{-11}	4.6×10^{-13}	3.5×10^{-15}
LLW	1,900	5.42×10^{-8}	4.1×10^{-10}	2.9×10^{-11}	2.2×10^{-13}

As radium-226 is produced by the decay of uranium-238, via uranium-234 and thorium-230, the time required to achieve secular equilibrium between the parent and progeny radionuclides and the radium-226 progeny will be governed by the half-lives of the various radionuclides in the chain and is very long (i.e. on a geological timescale). Hence, although ‘natural’ uranium deposits may be in equilibrium with radium-226, processed and purified materials, such as fuel residues and related wastes, from which radium-226 will have been separated, will not be at secular equilibrium. As a result, the maximum radium-226 inventory will occur approximately 10^5 years after repository closure and will amount to ~40TBq [3].

The initial radium-226 inventory of fuel will depend on the efficiency with which radium-226 was separated during the original processing of the uranium. The quantity of radium-226 in some irradiated fuels will be greater than would be estimated from the decay of uranium-238 alone. This is due to the increased uranium-234 content compared with natural uranium (as a consequence of the method used to enrich uranium in uranium-235 prior to fuel fabrication) and the decay of any plutonium-238 produced by neutron irradiation. Nonetheless, the inventory of radium-226 in fuel-related wastes will be small.

Radium-226 also will be present in technological wastes (i.e. medical and industrial sources etc.) that are deliberately enriched in either radium-226 or its direct parent radionuclide, thorium-230. In the case of thorium-rich wastes, the time to reach equilibrium with radium-226 is several thousand years and hence radium-226 will be present at a significant concentration only if the original thorium-rich material was relatively impure.

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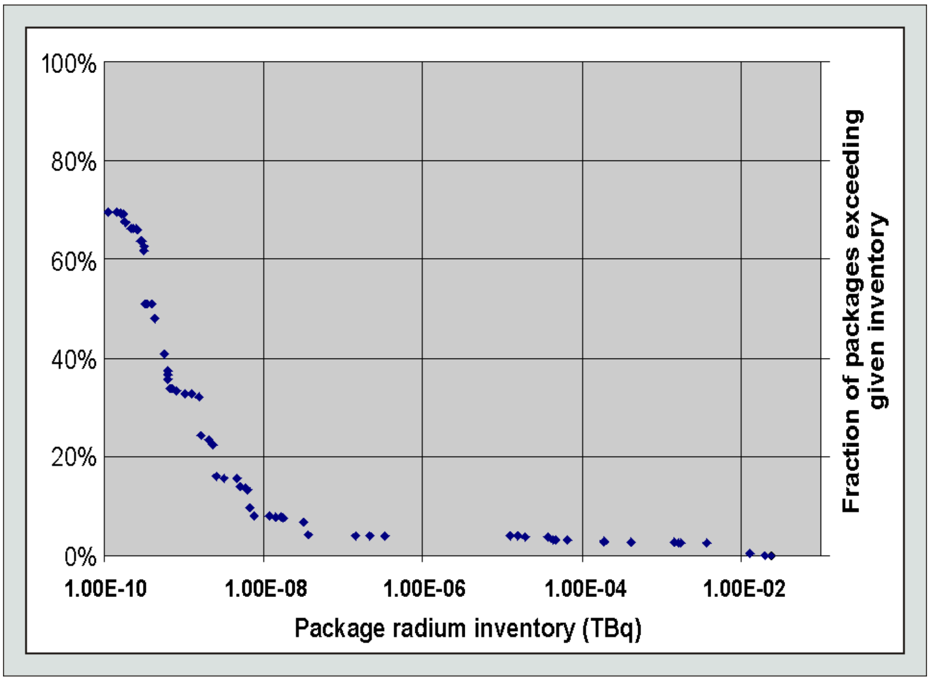
[6] Unshielded ILW to be packaged in Nirex unshielded waste packages (i.e. 500 litre Drums or 3 cubic metre Boxes or Drums)

[7] Shielded ILW to be packaged in Nirex shielded waste packages (i.e. 2 metre or 4 metre Boxes)

Figure 1 Distribution of Radium-226 in UILW



It may be concluded that significant radon generation is likely only for materials enriched in radium-226. It should, however, be noted that for wastes containing a significant inventory of thorium-230, radium-226 in-growth may be significant on the timescale of relevance to the transport and operational phases of the PGRC (i.e. up to a few hundred years). For uranium fuel based wastes, the in-growth of radium-226 from the precursor materials will not be significant on the timescale of relevance (i.e. during the packaging, transport and operational phases of the PGRC); therefore, only relatively impure fuel materials or thorium-230 wastes could contain a substantial inventory of radium-226.

4 WHY RADON RELEASE NEEDS TO BE CONSIDERED

4.1 Transport Safety

Releases of radon during the transport of waste packages could result in exposure of transport workers and members of the public. In this context the UK has adopted the International Atomic Energy Agency (IAEA) Transport Regulations [4] for the transport of radioactive wastes through the public domain. These regulations impose limits on releases of radionuclides, including those in the form of gases, from transport packages during normal operations and under specified accident conditions. The impact of the IAEA Transport Regulations on the PGRC is explored in the GWPS [2] and this is dealt with in Section 6.1 for the transport of radon-generating packages. In addition to national and international legislation, the overall transport operation should result in releases of activity which are As Low As Reasonably Practicable (ALARP).

4.2 Repository Operational Safety

Exposure to radon and its progenies could occur to workers within a repository (or in a Waste Packager’s surface store prior to transport to the facility) and to members of the public through stack discharges. The Nirex Radiological Protection Policy Manual (RPPM) [5] summarises the statutory dose limits and Nirex Design Targets applicable to the design of a repository; these are listed in Table 3.

The Generic Operational Safety Assessment (GOSA) [6], one of the safety assessments supporting the PGRC, has examined the consequences of the discharge of gaseous activity, including radon, from waste packages during this phase of the PGRC, taking into account the potential exposure routes resulting from on- and off-site releases during the repository operational phase. This safety assessment has been used to derive guidance levels for the radium-226 content and radon release rates of individual waste packages (Section 6.2).

Table 3 Dose targets and limits for on- and off-site exposure

	Dose Target (mSv/y)	Statutory Dose Limit (mSv/y)
Effective dose to workers	2	20
Effective dose to members of the public	0.02	1

4.3 Repository Post-closure Safety

Radon originating from radium-226 in a repository is not expected to present a hazard in the accessible environment once such a facility has been closed. Due to its relatively short half-life, radon originating from radium-226 within the facility is expected to decay before reaching the surface in significant quantities. Over very long timescales a small proportion of the uranium-238 originating from the facility may migrate, dissolved in groundwater, nearer to the accessible environment and decay to radium, through its progenies, and provide a source of radon. This has been considered by Nirex in the Generic Post-closure Assessment (GPA) [7] and is not expected to place requirements on the physical design of waste packaging over and above those required during transport and the operational phase. However, control of materials that could enhance uranium solubility in groundwater may be necessary and is advised by Nirex due to the potential significance of uranium migration and production of progeny radionuclides. This is not the subject of this Guidance Note but will be dealt with in future guidance.

5 GUIDANCE ON RADON RELEASE RATES AND RADIUM INVENTORIES

Control of the rate of radon release can be achieved either through limits on the radium-226 content of waste packages, since this is the source of the radon, and/or by designing the waste package to restrict radon release. The ensuing Section uses data from the PGRC safety assessments and relevant legislation to provide numerical guidance on the allowable levels of radon release from waste packages and the corresponding waste package radium-226 inventories.

5.1 Transport Safety

Two types of transport package exist within the PGRC. Nirex unshielded waste packages (i.e. the 500 litre Drum, the 3 cubic metre Box and the 3 cubic metre Drum) will be transported through the public domain in reusable shielded transport containers (RSTC’s), and these are categorised as Type B transport packages in the IAEA Transport Regulations. Nirex shielded packages (i.e. the 2 metre Box and 4 metre Box) are transport packages in their own right, and are categorised IP-2 transport packages under the IAEA Transport Regulations.

5.1.1 Type B Transport Packages

The IAEA Transport Regulations place a limit of $10^{-6}A_2$[8] per hour on releases of activity from Type B transport packages under normal conditions of transport (NCT). Radon released by waste packages will accumulate in the RSTC cavity and will gradually leak through the lid seal. Under NCT the maximum total gas leakage rate of this seal (expressed as a Standardised Leak Rate (SLR)) is 10^{-2} Pa/m^3/s. This value can be combined with the activity release limit to calculate a maximum allowable radioactive gas hold-up in the RSTC cavity, and a further calculation can determine the maximum rate at which waste packages can release gaseous activity. These calculations assume a build up of pressure within the cavity over the maximum period of transport (28 days) and set package release limits by assuming that the RSTC release limit of $10^{-6}A_2$ per hour is reached at the end of that period.

In the case of radon-222, the relatively short half-life (i.e. 3.825 days) means that significant decay of the gas, to a solid progeny product, will occur during transport and that this will allow a greater rate of release of radon from the waste packages into the cavity.

Table 4 lists the maximum radon release rates that would satisfy the requirements described above for the three Nirex unshielded packages, together with the quantities of radium-226 that would produce such rates. The values quoted do take into account the decay of radon in the RSTC cavity but do not take into account contributory releases from other radioactive gases. The presence and release of other radioactive gases would clearly lead to a reduction in the allowable radon release rates.

5.1.2 Type IP-2 Transport Packages

No explicit permissible release rate for radioactive gas from IP-2 packages is specified in the IAEA Transport Regulations, although Nirex has chosen to interpret the requirement to *'prevent loss or dispersal of the radioactive contents'* as being the same containment requirement as for Type B packages (i.e. $10^{-6}A_2$ per hour). Table 4 lists the maximum radon release rates that would be compatible with this requirement, together with the equivalent maximum allowable radium-226 inventories. No allowance is made for the radioactive decay of radon within the waste package as it is assumed that the gas is not held up within the package and escapes directly via the package vents.

It should be noted, however, that the possibility exists of sealing the waste package vents during transport, and this would permit higher radon generation rates.

5.2 Repository Operational Safety

The release of radon from packages, once emplaced in a repository, will have dose consequences to workers on-site and members of the public off-site, both of which have been assessed in the GOSA [6]. This has shown that the maximum on-site doses due to all radioactive gases, including radon, would be 0.18mSv/y, and that the maximum off-site dose due to all radioactive gases would be 2.1μSv/y, of which the contribution due to radon would be 0.08μSv/y. Both these dose rates are significantly lower (i.e. ~0.01%) than the target values from the Nirex RPPM, as listed in Table 3. However, both calculations assume significant retention of radon in waste packages, and the consequential reduction in effective emanation rates and dose consequences; the factor (known as the emanation coefficient) used was 2×10^{-3}.

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[8] A_2 is a measure of activity linked to possible exposure pathways and defined in the IAEA Transport Regulations. $1A_2$ is currently equal to 4×10^{-3}TBq of radon-222.

Work to estimate the degree of radon retention by waste packages [8] suggests that this value of emanation coefficient may not be readily achievable using conventional packaging techniques (i.e. intimate grouting using a cementitious material or even by enhancement of this technique by the use of an inactive grout annulus), and that additional measures will be needed to achieve it.

Accordingly, for the purposes of this document, a guidance level has been set on the basis that no benefit is claimed for radon retention by the waste package (i.e. the emanation coefficient is assumed to be unity) and using the off-site dose as the limiting factor. The guidance level, given in terms of TBq of radium-226 per cubic metre of conditioned waste, is calculated on the basis that, if it were applied to the entire waste volume emplaced in the repository, this would not result in the off-site dose target (i.e. 0.02mSv/y) being exceeded.

Table 4 Waste package radon release and radium inventory limits as set by transport constraints

Transport Package Type		Maximum Radon-222 Release Rate	Maximum Radium-226 Package Inventory
		TBq/hr	TBq
Type B	500 litre Drum[9]	3.2×10^{-7}	4.3×10^{-5}
	3 cubic metre Box	4.4×10^{-7}	5.9×10^{-5}
	3 cubic metre Drum	9.5×10^{-7}	1.3×10^{-4}
Type IP-2.	2 metre Box[10]	4×10^{-9}	5.4×10^{-7}
	4 metre Box[10]	4×10^{-9}	5.4×10^{-7}

Using a worst case value for the Dose Release Ratio for members of the public [9] for radon (i.e. 1.8×10^{-2}mSv/TBq) leads to a value of 1.1TBq/y for the total radon release that would lead to an off-site dose equal to the target value. This translates to an average radon release of 6.5×10^{-6}TBq/y per cubic metre of conditioned waste for the Reference Case volume of 168,000m^3. Table 5 uses this value to give guidance values for radon emanation from the five Nirex standard packages, together with the equivalent radium-226 inventories, assuming that no other radioactive gases are released from the waste.

The basis for the values given in Table 5 is very pessimistic in that no allowance is made for radon retention and decay by any mechanism. However, no allowance is made for the effects of other radioactive gas discharges which would contribute to off-site dose. The values given should, therefore, be regarded as coarse guidance levels; waste packages which have radium inventories less than these would not be expected to raise safety concerns, and no specific measures to reduce radon emanation need be considered.

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[9] Values are for a single drum assumed to be contained within a RSTC with three other identical drums.

[10] Assuming package vents are open.

5.3 Repository Post-closure Safety

As noted in Section 4.3, post-closure safety does not impose limits on the radium-226 content of waste packages over and above those required for the transport and operational phases of the PGRC.

Table 5 Guidance levels for Radon-222 generation and Radium-226 inventories for packages following emplacement.

Waste Package Type	Guidance Level for Radon-222 Generation	Guidance Level for Radium-226 Inventory
	TBq/hour	TBq
500 litre Drum	$3.7x10^{-10}$	$5.0x10^{-8}$
3 cubic metre Box	$2.0x10^{-9}$	$2.7x10^{-7}$
3 cubic metre Drum	$1.9x10^{-10}$	$2.5x10^{-7}$
2 metre Box	$6.7x10^{-9}$	$9.0x10^{-7}$
4 metre Box	$1.3x10^{-8}$	$1.8x10^{-6}$

5.4 Application of Limits and Guidance Levels

Comparing the values in Tables 4 and 5 shows that, for UILW waste packages, the guidance levels set by off-site dose considerations are more onerous than the limits placed by transport, the reverse being the case for SILW waste packages. Comparison of these values with the Inventory data in Figure 1 shows that a very small proportion (i.e. <5%) of UILW has radium-226 inventories in excess of the most onerous of the two limits.

It should be emphasised that the values in both Tables 4 and 5 do not necessarily represent firm limits on the inventories of radium-226 in waste packages. The values are conservatively derived and do not claim any benefit from any degree of waste package hold-up of radon that could reasonably be expected from the use of conventional packaging methods.

Accordingly, it may be allowable to package wastes with significantly higher inventories of radium-226 without incorporating specific measures to maximise this hold-up. The levels do, however, provide an indication of the radium-226 inventories above which the waste packager will be required to show to Nirex that specific consideration has been given to the potential for radon hold-up within the waste package as designed.

6 PACKAGING OF WASTES WITH LOW RADIUM INVENTORIES

Waste packages which have radium-226 inventories below the values shown in Table 5 for unshielded packages and Table 4 for shielded packages will not raise safety concerns. In order to ensure that radon release rates and consequent doses are ALARP, however, good practice should still be followed in the design of such waste packages.

Despite it being considered chemically un-reactive, radon can migrate relatively rapidly in some materials. Consequently, careful selection of packaging materials and good practice should be followed in package design to ensure a satisfactory reduction in the release rate. This could entail:

- use of common waste conditioning methods, such as use of cementitious grouts or supercompaction and grouting;
- use of capping grouts on the wasteform;
- use of container materials with good longevity under appropriate storage conditions, such as stainless steels;
- where gas vents are fitted to waste containers, the optimisation of the gas vent size.

7 PACKAGING OF WASTES WITH SIGNIFICANT RADIUM INVENTORIES

Waste packages which exceed the values shown in Tables 4 or 5 will require specific assessment by Nirex. As well as the best practice listed in Section 7, additional features may be required in waste package design, and an explicit case may need to be made by the waste packager to justify the proposed packaging method. Early dialogue with Nirex is, therefore, recommended to establish potential requirements.

The remainder of this guidance document provides information on packaging methods that could be considered and on methods for assessing and demonstrating waste package performance. Where a case needs to be made, consideration and justification of the measures applied to ensure compliance with limits on the rate of radon release advised by Nirex, and based on its detailed assessment of proposals, will be required. The design and implementation of such measures will be the responsibility of the Waste Packager.

7.1 Issues to Consider

A number of different packaging concepts may be applied to the packaging of radioactive wastes. The following issues are likely to be of concern in the development of packaging methods for radon-generating wastes:

- identification of the radium-containing items and, where possible, the estimation or measurement of the effective radon generation rate from those items. This should include a consideration of the evolution of such items following packaging;
- evaluation of the degree of mitigation required of the packaging, such as the encapsulant or waste container and associated features;
- specification of materials and, in particular, provision of robust migration rate data for the packaging materials under relevant conditions (degree of water saturation, age, extent of irradiation etc);
- particular consideration of the sealing of containers and the robustness of the packaging against manufacturing defects;
- consideration of the possibility of cracking or degradation damaging the barrier properties of the materials or design features used, including the evolution of the waste package over time (desiccation, extent of irradiation etc);
- generation of bulk gases in the same container, from degradation of wastes and any added encapsulants, and their effect on sealed or poorly vented containers, and on radon retention;
- provision of data regarding the properties of the packaging materials, obtained under relevant conditions (waste packagers are advised that properties are best measured directly, although available literature data are acceptable if they can be shown to be relevant);

- validation of models and arguments, ideally through measurements on packaged wastes or simulants (Nirex is developing models for assessment purposes, which may be of value to waste packagers).

Although some data on the properties of waste or packaging materials are available from the literature, these are not necessarily relevant to packaged wastes. The value of experimental measurements for the materials to be used is therefore emphasised, as is that of measurements of the rate of radon generation, effective generation rates and the rate of radon release (as necessary).

In all cases, the waste packager will need to provide an argument demonstrating that the performance of the proposed packaging method is suitable and likely to remain so over an appropriate period of time.

The various methods for mitigating the rate of radon release are discussed in more detail below. Further guidance on the presentation and validation of the arguments to support the use of a particular method is provided in Sections 8.3 and 9, respectively.

7.2 Packaging of Waste

7.2.1 Waste Sorting and Identification

In many cases, the radium inventory of a waste is confined to particular items, for example radium sources. The sorting of such items for separate packaging potentially provides a significant reduction in the amount of material for which special packaging measures might be necessary. Furthermore, materials that might give rise to significant bulk inactive gas generation, and hence bulk gas advection, may be segregated from radon-generating items, thereby providing greater flexibility in the packaging concepts available.

Waste packagers are also referred to *Specification for Waste Package Data and Information Recording, WPS/400.* It will be necessary to generate realistic and justifiable records for wastes, and this is likely to require examination of stored wastes to generate such a record where inadequate records exist, before packaging for long-term waste management.

7.2.2 Retention of Radon by the Waste

Radon will be retained within some types of waste, giving an effective radon generation rate that is significantly lower than the rate derived directly from the radium-226 inventory. This reduction can be characterised by the emanation coefficient, the ratio of the observed and expected radon generation rates. For some types of waste, the emanation coefficient may be substantially less than unity due to the physical nature of the waste.

Where the emanation coefficient of the waste is substantially less than unity, it may be possible to demonstrate that, although the generation rate calculated from a known radium-226 inventory is greater than limits, the effective generation rate is nonetheless consistent with such limits due to retention of radon by the waste itself. If this is the case, no additional credit need be taken for any mitigation of the rate of radon release by the packaging. The degree of retention by the waste can range from granular materials (low retention) to engineered objects such as sealed sources (high retention)[11]. Examples of emanation coefficients for some common materials are shown in Appendix B.

The presentation of an argument based on emanation coefficient will require waste packagers to demonstrate the degree to which radon is held up in the waste. The review of data has suggested that a reasoned argument is unlikely to be sufficient and, therefore, experimental evidence will be required. This should take the form of measured effective generation rates from the waste.

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[11] If the integrity of such sources can be assured for the operational phase of the repository.

Retention of radon in the waste may be threatened by the evolution of the waste itself and by changes in the environment after packaging. Waste packagers should, therefore, demonstrate an understanding of the evolution of the waste under appropriate conditions and its impact on radon retention. This should include consideration of the conditions that may be experienced during transport; for example increases in temperature.

7.2.3 Conventional Packaging Methods

Conventional waste packaging methods for ILW are based on the intimate grouting of wastes with a cementitious material or the use of an annulus enclosing compacted wastes. It may be possible to generate suitably validated models to show the degree of radon retention by the wasteforms, particularly the encapsulant materials, but this may not be a simple exercise. Nirex has commissioned modelling work to assess the effectiveness of these conventional methods in the mitigation of radon release rates [8]. This work suggests that significant benefits can be achieved by conventional ILW packaging, and that emanation coefficients of as low as 0.015 for intimately grouted wastes and 0.001 for annular grouted wastes could be achieved. However, these values are very dependent on grout porosity, and less porous grouts would yield significantly higher values for emanation coefficient (0.47 and 0.38 respectively). Uncertainties associated with waste heterogeneity and ageing of grout encapsulants also have to be considered.

7.2.4 Use of Engineered Features

In addition to the use of encapsulants, it may be possible to engineer other features into the wasteform or waste container design which will provide barriers to radon migration. These could include reduced container vent sizes, decay tubes or filters.

A gas vent will usually be required to release bulk gases and thus radon releases may also occur. Nevertheless, it will be best practice to minimise the vent size. If this is insufficient to control radon release, and in many cases this is unlikely to be sufficient, consideration could be given to use of decay tubes or absorbers as part of the container or wasteform component design. A decay tube is designed to provide a long pathway for radon diffusion, thus allowing decay to occur rather than release. However, to be successful consideration would need to be given to:

- the effect of gas advection speeding radon migration, due to bulk gas generation and release;
- the physical robustness and longevity of any engineered feature.

7.2.5 Containerisation of Waste in Gas-tight Packaging

The packaging of radium-containing waste in a gas tight container could provide a solution to the issue of radon generation. It provides the clear advantages that the rate of radon release will be (effectively) zero over the time period[12] for which integrity can be guaranteed, and that the justification of the expected performance is likely to be straightforward. However, the GWPS [2] includes ‘sealed containers’ in the list of hazardous materials to be excluded from wasteforms, so if such containerisation was to be considered, its wider implications would need to be assessed.

Of particular concern would be the effects of gas pressure on container integrity, this could be particularly so for very small containers where significant pressurisation could occur over the period of interest, potentially threatening the integrity of the container. Excessive pressure within a container may threaten the integrity of seals. This is particularly the case for polymer-based materials, which are relatively weak. To show that a proposed gas-tight containment method is acceptable, a waste packager would need to demonstrate that all potential gas generation mechanisms have been considered, and that the volumes of gas

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[12] A representative time period would need to at least cover the operation phase of the repository.

that may be generated within the container have been shown to be acceptable. It should also be noted that, if an approach involving the use a gas-tight component is proposed (either the waste container or sealed internal vessels within the wasteform), this would, as a minimum, require a convincing argument of gas-tightness prior to transport. There may also be implications in terms of the maintenance of integrity during the operational phase of the PGRC.

Pressurisation by radon is unlikely to be of concern as it can be shown that the volume of the radon generated from wastes is small. For example, 1TBq of radium-226 produces a volume of $1.2\text{x}10^{-6}\text{m}^3/\text{y}$ of radon at room temperature and pressure although, due to the decay of the radon to non-gaseous progeny, the equilibrium pressure resulting from this generation will be very small. The simultaneous generation of helium (due to the α-decay of radium-226 and its progenies) is also likely to be very small (1TBq of radium-226 will generate ~$6\text{x}10^{-6}\text{m}^3/\text{y}$ of helium in total) but, unless the helium is released, pressure will increase with time. Despite these relatively small volumes, the consequences of a sudden release of a volume of gas containing radon at secular equilibrium would need to be assessed.

Other sources of gas generation could, however, be more significant. These could include wasteform corrosion, microbial decay of organic materials, and/or the radiolysis of water and organic materials. Such mechanisms of gas generation will be greatly reduced if water is excluded from the waste, as it is an important reactant for both of these mechanisms. It is recommended that the exclusion of any source of water be adopted as a specification for such containerised waste. Organic materials may be excluded from the waste itself, but where a polymeric material is used to form the primary container, degradation of the container should be considered. If sterility can be guaranteed, then it may be possible to rule out microbial degradation of organic materials, and the consequential production of carbon dioxide and methane.

Other potential hazards arising from the use of a gas-tight container would be the generation of fine particulates and the formation of hazardous materials (e.g. pyrophoric materials such as uranium hydride).

Two concepts for sealed vessels have been identified; containers fabricated from metal or polymer. The former has precedents in the manufacture of sealed sources, whereas the latter might be considered analogous to food packaging or the use of membranes to exclude radon from dwellings.

The container material should have a well-documented low permeability and diffusion coefficient (or permeation coefficient) for radon through the intact material. However, the performance of such a container will then depend not on the properties of the intact material but on the integrity of the container seals or manufacturing joints. Waste packagers are advised that arguments based upon a sealed container should consider not only the properties of the materials but also the means of sealing the container and how the expected performance can be guaranteed over relevant timescales.

A number of commercial polymeric materials are available that are potentially suitable for use in the packaging of radon-generating wastes. These include aluminised polyethylene terphthalate (PET or Mylar) and other polymeric materials used in food packaging, and membranes intended for use in the construction of radon-proof walls and flooring. Examination of manufacturers’ information often provides details of radon migration rates for the materials, and such data may be cited to demonstrate the expected performance of waste packaging. However, it is recommended that the provenance of such data be examined to ensure that it is of suitable quality.

Overall, even though gas-tight packaging has many advantages and may seem attractive, there will be many potentially onerous considerations with regard to long-term waste management due to the reliability and longevity of sealing arrangements.

7.2.6 Multi-barrier Approaches

Multi-barrier approaches to wasteform or overall waste package design, which progressively reduce radon migration through each barrier without vulnerability to failure of a single barrier, could provide a high degree of confidence in radon retention for long-term waste management. This may also allow for some release of other gases and for reduced sensitivity to evolution of the waste packages during prolonged storage. Such an approach could involve the use of semi-permeable barriers, either in the form of encapsulants or use of other engineered features such as decay tubes. Factors to consider could be:

- the reliability of each barrier as manufactured;
- time periods for which this reliability could be guaranteed;
- evolution of the barriers;
- the ability to disperse other gases due to waste degradation whilst maintaining adequate containment of radon.

7.3 Presentation of Arguments

This section provides guidance on the factors that may need to be considered in the development of a packaging proposal in which credit is required for features of waste packaging. Further background information is provided in Appendix B.

7.3.1 Release of Radon from Waste

As discussed in Section 8.2.2, the retention of radon within the waste material may provide a significantly reduced effective generation rate. Although the potential retardation of radon due to diffusion within solid waste particles can be modelled, the validity of the conclusions is dependent on the validity of the assumed parameters describing the waste. It is, therefore, recommended that any claimed benefit from retention of radon in the waste should be based on experimental measurements of emanation coefficients.

7.3.2 Migration of Radon in Wasteforms

A number of different mechanisms for the migration of radon through the packaging may be identified, as follows:

- diffusive migration through the solid raw waste placed into a container;
- diffusive migration through an intimately-grouted wasteform in the aqueous or gas phases;
- diffusive migration through a grout annulus or capping grout in the aqueous or gas phases;
- advection of a mixture of gases including radon through the wasteform or annulus;
- diffusive migration or advection through cracks or defects in wasteform grouts.

In each case, the rate of migration may be used to derive a time delay before release from the waste package, and the resulting reduction in the rate of radon release estimated.

All arguments should be presented in an appropriate degree of detail. Suitably simplified or conservative arguments will be acceptable, although the following issues may need to be taken into account:

- Arguments may be based on simplified geometries, often one-dimensional. Where this is the case, the validity of the argument for the actual package should be considered. For example, horizontal migration through the sides of a wasteform may represent a shorter path length than vertical migration.
- Factors such as gas compressibility may be neglected in simplified but conservative arguments. However, if such arguments are not sufficient to demonstrate compliance with release limits, more complete descriptions may be appropriate.

- The potentially significant 'transient' period prior to the establishment of a constant pressure and uniform flow may not be included in a conservative argument. However, this period may encompass the entire storage period and hence may be beneficially included in a more complete description.
- The available data for use in the calculations are limited and may need to be validated for a particular system. The relevance of existing literature is always subject to question.
- Models often assume that the barriers presented by the packaging are intact and their properties can be represented by a single-valued parameter (although flawed or cracked wasteforms would also be amenable to modelling). This may be an optimistic over-simplification, and the sensitivity of the calculations should be considered.
- The assumptions upon which arguments are based may change with time. The sensitivity of calculations to such changes should be considered.

In the light of these comments, it is recommended that models should be developed to provide more accurate and realistic representations of the mechanisms.

7.3.3 Evolution of Waste Packaging

Any argument intended to demonstrate the suitability of a packaging proposal for the mitigation of radon release is required to take due account of the possible evolution of the package during extended storage. Of particular significance are any potential changes to the migration properties of packaging materials and the more general degradation of packaging and of containers intended to contain radon. In addition, if containerisation is adopted, the evolution of the container and its contents should be considered against the complete range of criteria in the Nirex specifications for waste packages.

The degree of water saturation in cementitious materials and its evolution may be of particular significance. At high water saturation, the migration of gases can occur only by dissolution of the gas in the pore solution and subsequent diffusion through the barrier. It is relatively straightforward to estimate the rate of radon migration due to such a mechanism. However, arguments based on the barrier presented by saturated cement may be compromised by a number of factors:

- the degree of water saturation may diminish with time as the cement dries and is subject to self-desiccation;
- the generation of bulk gas through, for example, corrosion may cause the wasteform to pressurise, threatening the integrity of the barrier;
- bulk gas pressurisation may also cause two-phase flow or the expulsion of water from the pore structure [10];
- the barrier may crack due to other mechanisms; for example, the expansive corrosion of waste.

Any argument based upon a water-saturated barrier and dissolution-diffusion should consider these possibilities and demonstrate that performance will not be compromised under the conditions that may be encountered. In practice, it is expected that the development of such an argument will be complicated and difficult to validate.

In addition to the above, the following factors are highlighted as potentially requiring consideration:

- changes in the mass-transport properties of cements as the wasteform evolves;
- degradation of any gas-tight metal containers due to corrosion or pressurisation;
- degradation of any engineered features of containers which act as barriers or limit the rate of radon release (such as the containment boundary and lid seals).

7.3.4 Data for Calculations

Appendix B provides a brief review of the availability of published data. This demonstrates that the radon migration properties of the various materials encompass a range. Consequently, the use of a particular value of a parameter selected from the literature needs careful justification. It is, therefore, recommended that a case based on modelling should endeavour to use data for the specific materials to be used in the proposed packaging. It is possible that this will require experimental measurements specific to the proposal, depending on the degree of reliability required.

8 VALIDATION OF ARGUMENTS

8.1 Validation of Radium Inventories

Any case simply based on limitation of the radium-226 inventory to place an upper bound on potential radon releases should be validated using data records of a suitable quality, supported by measurements of radium inventory where practicable.

8.2 Validation of Calculated Release Rates

Calculated rates of radon release from packaged wastes should be validated by experimental measurement wherever possible. This is particularly the case where a large reduction in the rate of radon release is being claimed. It is noted that the combination of modelling and experiments also provides a means for extrapolating the measured performance to take account of the evolution of the packaging.

The simple measurement of the rate of radon release from packaged waste potentially provides an unequivocal demonstration that the required reduction in the release rate is achievable. However, guidance on radon measurement methods is not provided herein. An approach based solely on measurement after packaging has a number of potential drawbacks. Firstly, the use of a measurement on the packaged waste carries the risk that, in the event that the packaging is not well designed, the limit will not be met and expensive re-packaging may be required. Secondly, consideration of the evolution of the packaging will be required, perhaps leading to repeated measurements. It should also be noted that, as part of the pre-transport checks required for waste packages and outlined in *Guidance on Fitness for Transport Quality Checks, WPS/660*, confirmatory measurements will be necessary prior to transport.

9 SUMMARY

The release of radioactive gases from packages of waste will need to be defined and controlled to enable safety cases for transport, storage and disposal to be made. Radon is one of a number of radioactive gases; however the generation of radon presents particular concerns due to its mobility and radio-toxicity. Fortunately, the relatively short half-life of the dominant isotope (radon-222 from the decay of radium-226) means that its generation is amenable to mitigation through appropriate packaging. In particular, the rate of radon release can be reduced by radioactive decay during migration through the barriers presented by the packaging.

Analysis of data from the 2001 Radioactive Waste Inventory shows that the vast majority of the radium-226 in wastes destined for the Nirex repository will be concentrated in a relatively small number of UILW packages. Significant radon generation is likely only for materials enriched in radium-226 (e.g. medical and industrial sources). It should, however, be noted that for wastes containing a significant inventory of thorium-230, radium-226 in-growth may be significant over the timescale of relevance to the transport and operational phases of the repository.

Calculations based on the pessimistic assumption that waste packages provide no mitigation of radon release have been undertaken to derive guidance values for the radium-226 inventory of UILW and SILW packages that would ensure that robust transport and repository operational safety cases could be made. Waste packages with radium-226 inventories below these guidance values will not raise safety concerns. In order to ensure that radon release rates and consequent doses are ALARP, however, good practice should still be followed in the design of such waste packages.

Waste packages with radium-226 inventories that exceed the guidance values will require specific assessment by Nirex. This Guidance Note provides advice on the measures that could be taken during waste packaging to optimise the mitigation of radon release from such packages. These measures include appropriate sorting and segregation of wastes prior to their conditioning, and the introduction into the waste packages of barriers to radon release. Multi-barrier approaches to waste package design, which progressively reduce radon migration through each barrier without being vulnerable to the failure of a single barrier, could provide a high degree of confidence in radon retention for long-term waste management.

This Guidance Note also provides advice on the presentation of robust arguments to Nirex, as part of packaging proposals, regarding the performance of waste packages with respect to radon release and mitigation thereof. All such arguments should be presented in an appropriate degree of detail (suitably simplified or conservative arguments will be acceptable), and should take due account of the possible evolution of the waste package during extended storage. Arguments may be based on experimental measurements (e.g. emanation coefficients of raw wastes) and/or models of radon migration in waste packages, although calculated rates of radon release from packaged wastes should be validated by experimental measurement wherever possible.

Overall, this Guidance Note provides information to assist with the design of waste packages and the presentation of robust arguments regarding the performance of packaging, in order to facilitate the safe and efficient packaging of radon-generating wastes. However, due to the wide variety of forms of wastes and potential packaging methods, waste packagers are encouraged to discuss their detailed waste packaging plans with Nirex at an early stage, in order to obtain independent advice on particular packaging proposals. Nirex is prepared to give advice on specific applications, based on its knowledge of waste package behaviour and performance requirements under storage, transport, handling and potential disposal conditions and from its experience obtained during the research and development of transport and disposal systems for radioactive waste.

APPENDIX A GLOSSARY

advective migration	Non-segregative flow of a gas through a permeable medium, driven by a pressure gradient. Commonly described by the Darcy equation. Radon may be advected in a stream of bulk gases or due to the pressure of radon alone.
barrier	Any component of a waste package that prevents or hinders the migration of radon from the package.
bulk gases	Inactive gases generated within wastes due to chemical processes, such as corrosion, and radiolysis. Commonly dominated by hydrogen.
containerisation	The packaging of radon-generating waste into an engineered gas-tight container designed to prevent the release of radon.
diffusion coefficient	(denoted D, units m^2/s). The property of a material that characterises diffusive migration. In Fick's laws of diffusion, relates the molecular flux of a diffusant and the applied concentration gradient.
diffusive migration	Segregative flow of a gas through a medium driven by a concentration gradient. Commonly described by Fick's laws of diffusion. In a waste package, this may take place in the aqueous or gaseous phase, depending on the water saturation of the pore space.
dose release ratio	To allow the results of computer generated models of the likely dose uptake from the release of activity the GOSA defines Dose Release Ratios for individual radionuclides. These provide numerical relationships for the dose received from 1TBq of a radionuclide allowing for its behaviour in the environment.
effective generation rate	(units as desired, e.g. Bq/s) The observable rate at which radon is produced by an item or waste containing radium-226. The product of the rate of radon generation and the emanation coefficient.
emanation coefficient	(no units) The ratio of effective generation rate and the rate of radon generation. Characterises the mitigation offered by radon decay during migration through an item of waste to a free surface.
Henry's law coefficient	(denoted H, units mol/m/N). The coefficient describing the linear relationship between the concentration of dissolved gas in a condensed phase and the partial pressure of that gas adjacent to the condensed phase. Applicable to relatively low solubilities where a linear relationship holds.
mitigation (of radon release)	The reduction in the observed or expected rate of radon release from a waste package, or other containment, when compared with the rate of radon generation or the effective generation rate. Mitigation arises from decay of radon during migration through the barriers provided by the package.

permeability	(denoted k, units m^2). The property of a permeable material that characterises advective migration. In the Darcy equation, relates the volumetric flow rate of a fluid and the applied pressure gradient.
permeation coefficient	A poorly-defined parameter used to characterise the diffusive flux of radon through a solid. May be applied to the product of the radon diffusion coefficient and solubility coefficient (1) (units of m^2/s), or the product of the radon diffusion coefficient and solubility coefficient (2) or Henry's law coefficient (units of mol m/N/s). To be distinguished from the permeability.
radon	The isotope radon-222, a product of the uranium-238 decay series and the direct progeny of radium-226.
(rate of) radon release	The net rate at which radon is released to the external environment from an engineered system such as a waste package. The product of the rate of radon generation and the mitigation offered by the packaging, allowing for any retention in the waste itself.
(rate of) radon generation	The rate at which radon is generated by the decay of radium-226. Equal to the activity of radium-226 if secular equilibrium is achieved.
secular equilibrium	The equilibrium ultimately achieved between a radioactive isotope and its direct progeny isotope(s), at which point the activity of parent and progeny isotope(s) will be equal (for cases where the parent has a longer half-life than the progeny isotope). Achievement of secular equilibrium is approached after a period equal to several half-lives of the progeny isotope.
solubility coefficient (1)	(denoted S, dimensionless). The parameter used by some workers to relate the concentration of gas dissolved in a solid to the concentration in the gas phase immediately adjacent to the solid.
solubility coefficient (2)	(denoted S, units mol/m/N). The parameter used by some workers to relate the concentration of dissolved gas in a condensed phase to the partial pressure of that gas adjacent to the condensed phase. Equal to the Henry's law coefficient if the solubility is low.
thoron	The isotope radon-220, a product of the thorium-232 decay series and the direct progeny of radium-224.

APPENDIX B BACKGROUND INFORMATION

B1. Radon Migration in Waste

The rate of release of radon from an item of waste or a wasteform may be reduced compared with the rate of its generation due to decay during migration through the waste material. Such a retardation of radon is demonstrated by measurements of the emanation coefficient. Measurements of emanation coefficients suggest that the effective reduction in the radon generation rate in solid items of waste could lie between 3 and 500. The actual value for the emanation coefficient will depend strongly on the physical and chemical form of the waste and wasteform.

The radium could be present in the following types of waste:

- radiation sources containing radium compounds;
- medical items such as radium needles;
- luminised (i.e. painted) items;
- fuel residues;
- scrap material produced during the manufacture of radiation sources;
- laboratory glassware and other materials contaminated with radium-bearing solutions (precipitated radium salts);
- tailings from uranium extraction from ores.

Radiation sources and scrap are assumed to be essentially metallic with typical dimensions of the order of 0.01-0.1m. In contrast, precipitated radium salts will be crystalline or amorphous particles with uncertain dimensions, perhaps as small as 10-100μm.

The radon migration properties of wastes are not reported in the literature. Furthermore, comparison of diffusion data for possible analogues with measured emanation rates suggests that the actual rate of release is not readily predicted from simple material properties. It is likely that radon migration through solids may be affected by migration mechanisms other than simple solid-state diffusion, for example diffusion along grain boundaries.

B2. Radon Migration in Cementitious Materials

B2.1 Mechanisms of radon migration

The migration of radon in cementitious materials may take place by a number of mechanisms [11]. The majority of the published data refers to the diffusion of radon through construction materials and is related to reducing the rate of radon entering buildings. The migration of radon is commonly characterised by an effective diffusion coefficient, determined from observed radon ingress and a known concentration gradient.

Advection of both gas and aqueous phases is described by the Darcy equation and characterised by the permeability to the relevant gas or liquid. The advection of radon has not been extensively studied. However, the advection of a bulk mixture of gases containing radon, or an aqueous phase, will be relatively unaffected by the presence of radon and more general data may be assumed to apply [12].

B2.2 Radon diffusion in cement and concrete

No data have been identified relating to radon diffusion in cementitious grouts of the types commonly used in the packaging of radioactive wastes. However, a few summaries of radon diffusion measurements in similar material are available [13, 14, 15]. These indicate that intrinsic diffusion coefficients for radon lie in the range 7.5×10^{-9} to 3×10^{-7}m^2/s. These data are also consistent with the rather limited literature on gas diffusion in cements and concretes published elsewhere [11, 16].

The retardation of radon may also be quantified by the radon emanation coefficient [17]. The emanation coefficient is defined as the ratio of the number of radon atoms released to the number of radon atoms formed by the decay of radium. Typical data are:

- uranium mill tailings, 0.05 to 0.30 [18];
- cement, 0.008 to 0.085 [17];
- brick, 0.008 to 0.16 [17];
- particulate materials, for example pulverised fuel ash (PFA), blast furnace slag (BFS) and gypsum, 0.002 to 0.21 [17].

B2.3 Other properties of cementitious materials

The migration of gases through cementitious materials is strongly affected by the presence of cracks [11]. Consequently, the use of materials (or manufacture of a wasteform) of a suitable quality is important in controlling the containment of radon.

B3. Radon Migration in Polymer Membranes

B3.1 Reporting of radon migration data

The migration of radon through a polymeric membrane occurs by a dissolution-diffusion mechanism governed by Fick's laws of diffusion. The migration rate of dissolved radon under steady-state conditions, where a linear concentration gradient applies, is given by:

$$J = -D\frac{\partial C_p}{\partial x} \tag{1}$$

where J is the molecular flux of radon (mol/m^2/s),

D is the diffusion coefficient (m^2/s),

Cp is the concentration of radon dissolved in the polymer (mol/m^3),

x is the spatial co-ordinate in the direction of migration (m).

In practice, the concentration of radon in the polymer itself is not measured and modified forms of Equation (1) are commonly used [19, 20]:

$$J = -DS\frac{\partial C}{\partial x} \tag{2a}$$

$$J = -DH\frac{\partial p}{\partial x} \tag{2b}$$

where S is a dimensionless solubility coefficient,

H is a Henry's law coefficient relating the dissolved concentration of a gas in a substance to the partial pressure of the gas above the substance (mol/m/N),

p is the partial pressure of radon (Pa),

C is the concentration of radon in the gas phase (mol/m^3).

The solubility coefficient is a dimensionless parameter and is not the same as the molar solubility of the gas in the polymer. Reported values of the solubility coefficient for radon are in the range 1-10, with the lower values being associated with 'harder' materials [19].

The presentation of data in the literature is potentially confusing due to the use of inappropriate terminology. The products DS and DH, although not identical, are both referred to as the 'permeation coefficient' or the 'permeability'. Furthermore, neither product is equal to the permeability coefficient that defines the volumetric flow rate due to advection under a pressure gradient (commonly denoted k, with units of m^2). Equations (2a) and (2b) describe the molecular flux not the volumetric flow rate.

The product DS has the same dimensions as a diffusion coefficient (units of m^2/s). However, the product is not numerically equal to the diffusion coefficient.

Although Equation (2b) effectively describes the flux as being due to a pressure gradient, the product DH does not have the same dimensions as the permeability coefficient (k), as the equation describes the molecular flux not the volumetric flow rate.

B3.2 Radon migration data

There is an extensive literature reporting measurements of the diffusion of radon through polymer membranes [e.g. 19, 20, 21, 22, 23, 24, 25, 26]. A summary of typical measured permeation coefficients (DS values) for some polymers is presented in Table B1. The radon permeation coefficients for the polymeric materials quoted are typically in the range 10^{-13} to $10^{-11} m^2/s$. There is considerable overlap in the reported permeation coefficients of the different materials.

The source of the polymer may be significant, with materials from different manufacturers exhibiting different permeation coefficients. Where materials may be manufactured in hard (more extensively cross-linked) or soft forms, the hard form generally has a lower permeation coefficient. An example of such a material is PVC [19].

Table B1 Radon permeation coefficients for some common polymer materials

Material	Permeation Coefficient (m^2/s)	Reference
Polyester	0.2×10^{-12}	[24] [25]
PVC	$(0.6\text{-}40)\times10^{-12}$	[19] [24] [25] [26]
Polythene	$(0.3\text{-}20)\times10^{-12}$	[21] [23] [24] [26]
Polypropylene	$(0.5\text{-}0.6)\times10^{-12}$	[20] [23]

The radon permeation coefficient is strongly temperature dependent for some polymers. For example, the permeation coefficient for polypropylene increases by an order of magnitude from $5\times10^{-13} m^2/s$ to $5\times10^{-12} m^2/s$ as the temperature is increased from 20°C to 40°C [20]. It has been reported that the gamma irradiation of polyester and polypropylene decreases the permeation coefficient by a factor of about two for doses up to 0.1MGy, although the reasons for this decrease have not been elucidated [B27].

B4. Experimental Methods

The experimental methods currently used in the determination of radon migration properties of materials may be broadly divided into three categories:

- measurement of radon ingress into existing structures and buildings;
- small-scale ‘membrane’ experiments;
- direct measurements of radon emanation from waste, or from simulated or actual packages.

Measurements of radon ingress into buildings are mainly concerned with characterising construction materials and supporting models. This experimental method is otherwise of little relevance to the packaging of wastes.

Membrane experiments involve the measurement of the rate of migration of radon through a relatively thin specimen of the material of interest, driven by a concentration gradient established by placing a radon source on one side of the membrane. The migration rate may be determined by monitoring the rate at which radon appears on the other side of the membrane [13, 17, 28]. A variant of this method is the ‘can’ technique, in which migration is characterised by monitoring the rate of radon loss from a vessel sealed by a membrane [26].

Gas diffusion and permeabilities for cementitious materials may be obtained by a membrane method, although this has not been widely applied to the study of radon migration [29, 30]. As the majority of reported data have been obtained for dried materials, and the migration of gases in cementitious materials is sensitive to water saturation, the use of any such data should be approached with caution [11].

The measurement of emanation rates is potentially straightforward but requires care to ensure that the radon is collected efficiently for measurement. This will be easier for small specimens, and measurements on full-scale waste packages require careful consideration.

All of the above methods may be readily applied to the characterisation of materials that may be used in the packaging of wastes. However, it should be noted that methods such as those based on membranes are based on the use of a small specimen of material, often specially prepared. Migration may be dominated by flaws, for example cracks or joints in polymer membranes, and, therefore, attention should be paid to characterising representative samples of materials in the form in which they will be used, or selecting materials that do not contain these flaws.

B5. Selection of Data for Use in Modelling

A wide range of radon migration data have been reported for a number of different materials. The selection of data for use in modelling to support a waste packaging proposal will depend on the nature of the material and the basis of the arguments used. In principle, the data presented above, or similar, could be used as the basis for a packaging proposal. However, data are material specific and generalisations are difficult, particularly for cements.

As discussed earlier it is expected that the performance of waste packages in retaining radon will be sensitive to the condition of the packaging material as manufactured and as it evolves. It is recommended that waste packagers should provide experimental data relevant to the particular materials to be used in packaging, rather than rely upon citing literature data, unless it can be clearly shown to be applicable.

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