

**WASTE PACKAGE SPECIFICATION AND
GUIDANCE DOCUMENTATION**

**WPS/903: Guidance on the Immobilisation
of Radionuclides in Wasteforms**

**March 2007
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WASTE PACKAGE SPECIFICATION AND GUIDANCE DOCUMENTATION

GUIDANCE ON THE IMMOBILISATION OF RADIONUCLIDES IN WASTEFORMS

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 disposal 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) process¹.

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 LLW².

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*³.

The diverse physical, chemical and radiological nature of ILW 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 an abstract of each, can be found in *Introduction to Nirex Waste Packaging Guidance Notes, WPS/900*.

¹ Formerly known as the Letter of Comfort process.

² For the purposes of this document, any reference to ILW shall include those categories of LLW that are to be subject to a regime of long-term waste management in accordance with the PGRC

³ Specific references to individual sections of the WPSGD are made in this document in *italic script*, followed by the relevant WPS number.

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Industry practice for the conditioning of ILW in the UK generally involves the intimate mixing or infiltration of the waste with an encapsulating material (usually cementitious) or, when appropriate, by supercompaction and grouting. The waste packages thus produced provide isolation and containment of the waste and renders it consistent with the requirements for passive safety and disposability. The process of conditioning forms one of the engineered barriers that prevents the return of radioactivity to the environment.

Whichever conditioning method is used, in order to ensure the compatibility with all stages of long-term management long-term management defined by the PGRC (i.e. interim surface storage, transport to a phased geological repository, emplacement and final disposal), waste packages must conform with the standards and specifications defined by the GWPS.

The nature of some wastes are such as to include radionuclides in a mobile form that could affect the ability of the chosen method of conditioning to be effective. This could potentially have a significant effect on the risk posed by packaged waste throughout its lifetime, during which it could be exposed to a variety of conditions. It is therefore important that such mobile radionuclides, when present in significant quantities, are explicitly dealt with as part of the conditioning process. It is also important that a demonstration of the effectiveness of the selected conditioning process is presented by the waste packager for assessment as part of the LoC process, in particular in the assessment of wastefrom performance.

This guidance deals with the issues relating to the immobilisation of radionuclides in wastefroms by identifying the forms in which potential mobile activity could exist and discussing the approaches that could be adopted to render them immobile. The guidance should be read in conjunction with the GWPS [2] to which extensive reference is made.

2 BACKGROUND

2.1 Organisation and Aims of this Document

The particular aims of this document are to:

- define 'immobilisation' in the context of the packaging of ILW;
- explain the basis for the requirements of the GWPS with respect to immobilisation;
- identify the different physical and chemical forms in which mobile radionuclides exist in ILW and describe practical means by which their immobilisation can be achieved.

This section provides summaries of the Nirex PGRC, the LoC Assessment Process⁴ and the Nirex approach to setting standards and specifications for the packaging of ILW which form the basis for the LoC process.

Section 3 defines immobilisation, the need for the achievement of immobilisation in the packaging of ILW and a summary of the approaches that may be adopted to achieve effective immobilisation.

Section 4 considers the specific areas of waste package performance that may be affected by the presence of mobile activity and discusses the waste conditioning techniques that can be used as part of a packaging process to help ensure compliance with the requirements of the GWPS.

Section 5 identifies, by reference to the relevant sections in the wasteform specification of the GWPS, the types of wastes that may give rise to mobile activity and the approaches that can be adopted to produce wasteforms in which this activity can be rendered sufficiently immobile.

Appendix A contains the Wasteform Specification from the GWPS to which extensive reference is made in Sections 4 and 5.

2.2 The Nirex PGRC and Letter of Compliance Assessment Process

The PGRC [1] has been developed by Nirex as a viable option for the long-term management of ILW in the UK and, as such, is the basis for the packaging standards and specifications which constitute the GWPS [2]. The PGRC envisages that, following a period of interim surface storage at the site of arising, packaged wastes would be transported to a repository facility. Such a facility would be constructed in stable geology, deep underground, to provide long-term isolation of the radioactivity in the wastes in order to protect human health and the accessible environment. The PGRC allows for the facility to be operated in a phased approach with the ultimate aim of sealing and closure. Each phase would be reversible and time would be available to build confidence at each stage before moving to the next.

The safety philosophy adopted in the PGRC is one of containment of radionuclides by multiple barriers of which that provided by the waste package is a key component. The waste package can actually be considered as two independent but complementary barriers, the waste container and the wasteform, each of which plays an important role in containment. In consequence of this the GWPS sets performance requirements for both of these components, against which the overall performance of the waste package is judged as part of the LoC assessment process.

The LoC assessment process, has been developed as a means of assessing the disposability of packaged wastes, against the requirements of the GWPS. In undertaking

⁴ A full description of the LoC process can be found in *WPS/650*.

LoC assessments Nirex determines whether wastes when packaged will have characteristics compliant with plans for transport to, and operations at the repository facility, and ultimately whether the wastes could be accommodated within the repository long-term post-closure safety case. As described in regulatory guidance [3] this assessment of disposability is required to provide a component of overall safety case for the operator's packaging plant and the waste packages that will ultimately be produced.

Upon completion of an assessment of a packaging proposal, Nirex will provide an Assessment Report relating to the further progression of the proposed packaging route, which may be accompanied by the issue of a LoC endorsing the packaging proposal. The Assessment Report may recommend prior treatment of the waste to deal with specific concerns. These and other particular uncertainties and risks arising from the chosen packaging method(s) will be highlighted, as Action Points. Subsequent to the issue of an Assessment Report, Nirex will continue to monitor progress with the resolution of such Action Points.

2.3 The Generic Waste Package Specification

Since its inception, a major area of Nirex's work has been in the provision of advice to the packagers of ILW in the UK. This has involved the definition of packaging standards and specifications, known as waste package specifications, this process culminated in 2005 with the production of the GWPS [2]. Derived from the PGRC, and its associated generic documentation, which comprise the system specifications and safety assessments that define the PGRC, the GWPS provides the basis for the assessment of proposals for the packaging of ILW in the UK.

The packaging standards and specifications presented in the GWPS are generic in two respects in that they are:

- derived from a full consideration of all future phases of waste management, as defined by the PGRC; and
- independent of the location of the site of a repository facility, which could be implemented at a range of different sites within the UK, representing a range of geological environments.

The GWPS specifies what is to be achieved, but avoids placing undue limitations on the methods by which the requirements may be met.

The format of the GWPS is to define:

- general requirements that are applicable to all waste packages;
- a range of standard waste containers;
- specific requirements for the standard waste packages that are created using the standard waste containers;
- requirements for the conditioned wasteforms that are placed into containers;
- requirements for quality management and for the creation and maintenance of records about each individual waste package.

In the GWPS a key requirement for all wasteforms is the effective immobilisation of activity, be it in particulate, liquid or gaseous form. Additionally the GWPS requires that wasteforms, in which activity was originally rendered immobile, do not evolve in such a manner as to create mobile activity.

Effective immobilisation of radionuclides is necessary if the packages are to satisfy the requirements for minimisation of releases of radioactive materials under normal and accident (e.g. fire and impact) conditions. The longevity of radionuclide immobilisation

must also be considered, because the evolution of a wasteform may result in physical and chemical degradation that could reduce the effectiveness of immobilisation.

As part of the justification for the requirements for the effective immobilisation of activity the GWPS refers to Guidance to Inspectors issued by the HSE [4] which points out the risks of waste dispersion and how they can be reduced through the concept of passive safety. This guidance states:

'Passive safety requires the radioactivity to be immobilised and packaged in a form that is physically and chemically stable..... In many cases, the raw radioactive material or radioactive waste will require conditioning to place it into a passively safe form to immobilise the radioactivity.'

Similarly, IAEA guidance on the conditioning of radioactive wastes and, in particular, the requirements and methods for low and intermediate level waste package acceptability [5] states that:

'...waste forms that promote dispersion in the event of a release (respirable particles)...should be prohibited in the waste package.'

The GWPS lists the key benefits of radionuclide immobilisation, particularly where cementitious matrices are used, as helping to ensure:

- that releases of radioactivity following accidents are more likely to be low and predictable;
- low and predictable rates of corrosion of waste and waste container materials;
- reduced solubility of many key radionuclides and toxic chemicals;
- compatibility of waste packages (e.g. porosity, permeability and stability) with the backfilled repository environment.

Additionally, two of the three safety assessments that help underpin the PGRC (i.e. the Generic Transport Safety Assessment (GTSA) [6] and the Generic Operational Safety Assessment (GOSA) [7]) rely on the requirement that radionuclides are effectively immobilised within wasteforms, such that release of activity under accident conditions will be small.

Finally, as part of the quality management requirements of the GWPS, waste packagers are required to provide evidence or reasoned argument concerning the effectiveness of the wasteform production process, and its effectiveness for the immobilisation of radionuclides.

3 IMMOBILISATION

3.1 General

This section considers what is meant by ‘immobilisation’ in the context of the packaging of ILW, and the underlying reasons for such a requirement.

The GWPS [2] states that:

‘The wasteform shall be designed to immobilise radionuclides and toxic materials so as to ensure appropriate waste package performance during all phases of the PGRC. For many wastes, this immobilisation requires the use of an encapsulating matrix.’

More specifically it also requires that:

‘All reasonable measures shall be taken to ensure that radionuclides and toxic materials in the waste are immobilised and that loose particulate material is minimised.’

Radionuclides can be said to be immobile if the characteristics of fluidity, dispersibility and freedom of movement within a waste package are eliminated. Immobilisation is therefore any process by which mobile waste fractions are conditioned in such a way that the potential for migration or dispersion of the radioactivity associated with a waste, at any stage during its long-term management, is reduced to an acceptable degree.

Adequate immobilisation is the conditioning of waste in such a way that, as a minimum, the release of radionuclides from the packaged waste, under normal or accident conditions, is within the acceptable range of values defined in the GWPS. In the case of radioactive gases, adequate immobilisation will offer an appropriate degree of hold-up and allow controlled and predictable release of the gases at a rate consistent with the defined release limits. For liquids (aqueous and non-aqueous) and particulates, movement will be prevented by the wasteform matrix into which the previously mobile wastes are incorporated.

The degree of immobilisation afforded by a waste package under the various conditions that may arise during any stage of long-term management is required to be such that the waste package is consistent with the following general requirements or principles:

- The radiological detriment to members of the public should be as low as reasonably achievable (ALARA) [8];
- Best practicable means (BPM) should be used to ensure that radiological detriment is ALARA;
- ILW should be conditioned and stored in accordance with the principle of passive safety set out in NII guidance to its inspectors. This requires that the radioactivity be immobile and the wasteform and container be chemically and physically stable, so that the need for safety systems, maintenance, monitoring and human intervention is minimised [4, 9].

Consequently, waste packagers should aim to maximise the degree of immobilisation afforded by wasteforms in particular and waste packages in general.

Wastes that will require specific conditioning to achieve immobilisation are those that contain radionuclides in particulate, liquid and gaseous forms and those that are volatile or soluble. These characteristics and properties may be associated with wet solids, slurries, sludges, powders, particulate material and bulk solid material that may corrode to produce loose material. Immobilisation for these types of wastes can generally be achieved by intimate encapsulation of wastes with a cementitious or other conditioning agent.

Potentially mobile radionuclides will be either physically trapped or chemically bound within

the immobilising matrix. Physical trapping could be within the pores and/or gel of the immobilising matrix or within isolated pockets closely surrounded by the matrix. Additional conditioning, prior to encapsulation may be required for wastes with particular physical and/or chemical properties.

Both of the approaches to immobilisation described above are commonly referred to as 'encapsulation' although it is recognised that the definition of 'encapsulation' varies, e.g. in the USA. A distinction is therefore drawn in this guidance between microencapsulation - containment of individual physical components of the waste in a matrix (here referred to as 'encapsulation'); and macroencapsulation - the encasement of a mass of waste within a matrix, here referred to as 'enclosing'.

In some cases, adequate immobilisation may be afforded by the waste itself. Those wastes in which the radioactivity is not present in a mobile form (i.e. bulk metals containing neutron-activated radionuclides) and that will not generate mobile radionuclides by their evolution, may not require additional conditioning in order to render them passively safe and acceptable for disposal, although may require measures to reduce voidage.

3.2 Definition of Significant Quantities

It is acknowledged that the complete immobilisation of all activity in a wasteform is an unduly onerous requirement for all waste packages and that, for some waste packages, the total activity associated with potentially mobile waste fractions will be so low such that the release of radioactivity under normal or any credible accident conditions cannot exceed the limits defined in the GWPS.

A consideration of the GWPS requirements for allowable activity release from waste packages shows that, for activity in particulate form, release limits are bounded by those associated with impact and fire accident performance requirements. Accordingly, *de minimus* quantities of mobile activity in a wasteform, below which specific waste conditioning to immobilise such activity would not normally be required, have been defined for the range of standard waste packages. These are shown in Table 1.

Table 1 *De minimus* levels for particulate activity in standard waste packages

Waste Package Type	Particulate Activity
	(A ₂ ⁵)
500 litre Drum	1.5
3 cubic metre Box	3
3 cubic metre Drum	9
2 metre Box	3
4 metre Box	6

For waste packages that contain less than these *de minimus* quantities of particulate activity, the development and adoption of specific processes incurring additional cost and

⁵ A₂ is a measure of activity linked to possible exposure pathways defined in the IAEA Transport Regulations.

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dose uptake in order to demonstrate that complete immobilisation is achieved may not be warranted. The use of simpler approaches to packaging may be adequate to satisfy the requirement of the GWPS. Waste packaging proposals for packages that have radionuclide inventories in excess of these values should address the issue of immobilisation, although such demonstration is likely to be simple, and to rely on reasoned argument and a consideration of other aspects of the waste and wasteform, for mobile activity inventories of less than a few multiples of the values in Table 1.

When considering whether a waste package contains a potentially significant source term, account should be taken of the effects of prolonged storage on the radionuclide inventory. Although the general trend is for the radionuclide inventory of waste packages to decline with time due to radioactive decay, in certain wastes there is the potential for it to increase due to radioactive in-growth. This can lead to pronounced increases in inventory where activity is measured in terms of A_2 and where the daughter has a low A_2 value relative to the parent (e.g. Am-241 produced by the decay of Pu-241). This can also be a significant factor with the formation of radioactive gases from the decay of a parent radionuclide (i.e. Rn-222 from the decay of Ra-226).

3.3 Waste Packaging Options

Current common practices used for the packaging of ILW intended for long-term management in accordance with the PGRC can be categorised as:

- in-drum grouting;
- compaction followed by grouting;
- in-drum mixing.

Wastes may additionally need to be subject to a range of pre-treatments before any of these options can yield a satisfactory product, or in order to improve the performance of the final product. However most waste packaging processes endorsed to date can be seen to have been produced using one of the approaches identified above. In any case, waste packages produced by any process, or combination of processes, should be consistent with the requirements for the immobilisation of radionuclides, and all other requirements, of the GWPS.

In-drum grouting involves the infilling of solid wastes within the disposal container by the addition of the immobilising material in a form with suitable fluidity to infiltrate the waste, e.g. cement grout or other solidifying matrix (e.g. polymeric). The conditioning of wastes by such methods has been used extensively in the UK for the packaging of solid ILW.

High-force compaction ('supercompaction'), is a process by which wastes are reduced in volume by mechanical compaction, using forces of the order of 10^6N . Compaction offers beneficial volume reduction of wastes, thereby offering a potentially significant reduction in the costs associated with their subsequent management. Compaction also generates, in many cases, a coherent solid waste that acts to immobilise radionuclides and prevent release. Compaction operations in the UK have routinely been applied to achieve significant volume reduction of compactable LLW in 200 litre drums. Supercompaction of drummed plutonium contaminated wastes has also been applied, notably at the Sellafield Waste Treatment Complex (WTC), where a 500 litre Drum waste package contains an average of $\sim 1\text{m}^3$ of waste reduced in volume by a factor of ~ 2.5 . Supercompacted 'pucks' are typically loaded into standard 500 litre Drums and grouted to form what is known as an 'annular grouted' wasteform

In-drum mixing involves the mixing of liquid or slurry wastes with an immobilising agent, e.g. cement powder, within a cylindrical waste container (i.e. 500 litre Drum or 3 cubic metre Drum) using a mixing paddle which remains within the wasteform after mixing (the

so-called 'lost' paddle). This process has been used extensively in the UK for the packaging of fluid wastes, including liquors, sludges and flocs.

Some wastes may be suitable for packaging without the need of a conditioning matrix. In such cases (i.e. for irradiated metal wastes with little or no loose contamination), the immobilisation of radionuclides is provided by the form of the waste. The lack of intimate encapsulation may, however, raise wasteform issues (i.e. voidage) that will need to be considered and addressed if the packaged waste is to be shown to be compliant with the GWPS.

4 IMMOBILISATION REQUIREMENTS OF THE GWPS

Section 3 has addressed the general packaging issues that may arise as a result of the presence of potentially mobile activity in a particular waste, in particular the properties of packaged waste that may influence the ability of a waste package to afford adequate immobilisation of such activity. The GWPS identifies a number of wasteform and waste package properties that will be of relevance in assessing the continuing ability of packages to contain radionuclides over the timescales envisaged for long-term management in accordance with the PGRC and, in particular under defined impact and fire accident conditions.

This section provides a summary of some key issues that will be considered as part of the LoC assessment process and that may have significance when the immobilisation of activity is considered. This does not represent the full range of wasteform and waste package issues, but discusses those which are likely to be particularly relevant in respect of mobile activity.

Reference is made to the wasteform specification of the GWPS and, for convenience, this is included in this guidance as Appendix A.

4.1 Impact performance

Waste packages are required to perform adequately in response to a number of defined impact accidents that may occur during handling, transport and repository operations. These requirements are expressed in the GWPS in terms of allowable limits on the release of activity following drops from specified heights which have been derived from the IAEA Transport Regulations [10] (0.3m and 10m) and the Design Basis Accident (DBA) analysis that forms part of the GOSA [7] (25m).

The allowable limits for activity release following these impact accidents are listed for the five Nirex standard waste packages in Table 2.

In addition to these release limits, the GWPS also states that:

'The wasteform and container should control the production and release of particulate material in the event of an impact accident, such that the waste package shall exhibit progressive and predictable release of material with increasing impact energy'

'The [impact] releases should not depend unduly on the maintenance of the waste container integrity'.

It is not necessarily the case that wasteforms in which radionuclides are adequately immobilised at the time of packaging will also perform adequately under impact accident conditions, since other wasteform and container properties such as mechanical strength will be relevant and because particulates may be generated by the impact. Further, impact release limits are relevant to the transport and operational phases of waste management, such that package components may have undergone degradation during interim storage, and that any effects arising may have altered the degree of immobilisation afforded by the wasteform.

If a waste package has adequate mechanical properties⁶ and has undergone no significant degradation during interim storage (Section 4.3), then packages in which radionuclides are immobilised at time of packaging are likely to show progressive and predictable release following impact.

Table 2 Impact and Fire Release Limits for Standard Waste Packages

Accident Type	'Normal handling' Impact	Transport Impact Accident	Repository Impact Accident ⁷	Repository Fire Accident
Accident Criterion	0.3m drop on to flat unyielding surface	10m drop on to flat unyielding surface	25m drop on to unyielding surface	1 hour duration fire, 1000°C flame temperature
Maximum Particle Diameter	N/A	40µm	100µm	N/A
Waste Package Type	Release limit (A₂)			
500 litre Drum	No loss of contents	3	3	1.5
3 cubic metre Box		3	12	4
3 cubic metre Drum		11	12	9
2 metre Box	No loss of contents. Loss of shielding integrity must not result in >20% increase in surface dose rate	N/A	4	3
4 metre Box		N/A	8	6

⁶ Wasteforms with compressive strengths of between 4 and 40 MPa have been shown to perform adequately under impact accident conditions, although wasteforms whose properties fall outside of this range may also meet the requirements.

⁷ A number of aspects of these DBAs are currently under review. Waste packagers should contact Nirex to ascertain the current situation.

4.2 Fire performance

The GWPS includes criteria for the release of activity from waste packages following exposure to a specified fire accident. As was the case of the 25m impact accident, such an event is considered a DBA in the GOSA and the allowable releases of activity have been derived using a similar methodology. The allowable release limits for standard waste packages exposed to a fire accident are listed in Table 2.

Factors relevant to the assessment of the fire accident performance of waste packages include:

- radionuclide inventory and speciation; gaseous, volatile, and entrained radionuclides in soluble and particulate form may be released;
- wasteform specific activity;
- distribution of the wastes, e.g. they may be close to the drum wall and therefore subject to a more pronounced temperature excursion;
- distribution of radioactivity and voidage within the wasteform; retention of activity by the waste, short circuit pathways for steam-entrained or volatilised radionuclides through voids;
- response of encapsulating agents to temperature increase; for example, cracking would allow for more rapid release of entrained or gaseous radionuclides;
- amount and form of water in the package; heat removal by evaporation;
- thermal properties of the wasteform and its components.

Where releases are predicted to exceed, or be close to, the specified release criterion, thermal performance will be improved by the provision of a protective grout annulus surrounding the waste, so that radionuclides are essentially insulated against excessive heating. This could be achieved by the use of a pre-cast grout annulus or container furniture to position the waste away from the drum walls. The addition of a grout cap to the wasteform will also act to limit the radionuclide release from packages in the event of fire.

4.3 Waste Package Evolution

Waste packages are assessed in terms of their long-term performance under the timescales and conditions pertaining to interim storage, transport and emplacement within a phased geological repository facility. Consideration is given to the ability of the package to continue to meet the requirements of the GWPS.

As discussed above the waste package can be considered as two independent but complementary barriers, the waste container and the wasteform, each of which plays an important role in containment. When considering waste package evolution it is therefore necessary to consider the potential for the degradation of both of these barriers with time. The waste container provides the primary barrier between the conditioned waste (i.e. the wasteform) and the environment (e.g. in the surface store, transport container cavity and repository vaults). The GWPS sets an integrity target of 500 years for the integrity of the waste container body and the surety of its lifting features in order to facilitate a period of repository operations whereby waste packages are maintained under a regime of monitoring and retrievability. This target is deemed readily achievable by the selection of suitable container materials, fabrication techniques and storage conditions. The durability of other components of the waste container (i.e. the lid seal and the filtered vent) are also addressed by the GWPS.

The potential outcome of wasteform evolution could be the loss of required beneficial wasteform properties, or a loss of any beneficial property or properties of a waste for which credit has been taken. For example, the ability of a sorber to retain free aqueous or non-

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aqueous liquids could support the adequate performance of a waste package at the time of production, but any such beneficial property could be lost through the degradation of the sorber during storage. Waste packagers should therefore demonstrate that potential modes of ageing and evolution and the effects on the properties of the waste and wasteform have been adequately addressed.

Factors relevant to assessment and likely to be of relevance when considering the effects of evolution on the continuing ability of the wasteform to offer adequate immobilisation include:

- The generation of particulates; this may be as a result of, for example, small impacts coincident with handling, corrosion of metallic wastes or the formation of expansive phases within the wasteform matrix;
- The generation of free liquids; for example, following loss of sorbing capacity or coalescence of unstable emulsions;
- The generation of NAPLs, e.g. by the radiolysis of polymers;
- The formation of voids following wasteform degradation and migration of the products of evolution;
- The generation of complexants by the degradation of organic wastes (see Section 5.6);
- The generation and liberation of gaseous radionuclides followed by release; as a function of radioactive decay and waste evolution, respectively;
- The modification of the mass transport properties of the wasteform matrix; the permeability of cementitious matrices would be expected to fall as hydration continues with time in the presence of water. Reduced permeability would slow the migration of radionuclides, including gases, through the wasteform. Such a retardation of movement is beneficial in terms of the retention of radionuclides, but may have implications for the ability of the matrix to release gases in cases where the rates of generation are high;
- Radionuclide migration; particularly for gaseous radionuclides, some migration is expected to occur during storage. If wasteforms are homogeneous with respect to the distribution of sources of radioactive gases, this should not pose a threat to wasteform performance, provided that the long-term release rate is consistent with the relevant limits. It is conceivable, however, that gases migrating from a point source could be released over a relatively short period during transport.

It should be noted that some of the above may also impact the effectiveness of the containment barrier (i.e. the waste container). Particular examples are the expansive corrosion of wasteform materials leading to stresses in the waste container or the radiolytic generation of aggressive species leading to accelerated corrosion of the waste container.

4.4 Waste Product Specification

As part of each Interim stage LoC submission, waste packagers are required to produce a Waste Product Specification (WPrS) which defines the specification of the waste package product that a packaging process is setting out to produce. Guidance on the content of a WPrS is available in *Waste Product Specification: Guide to Structure and Format, WPS/620*.

There is a clear need to ensure that wastes as presented for treatment and packaging, and when packaged, are compliant with the WPrS developed during packaging proposal development. If the WPrS fails to adequately identify the factors of relevance, then

assessment of the packages against any future Waste Acceptance Criteria (WAC)⁸ for a phased geological repository facility may be unnecessarily challenging.

The WPrS will need to embody the features that have been identified as important to the immobilisation of the waste. The key features for packages containing potentially mobile wastes, that is particulates, aqueous and non-aqueous liquids, or significant inventories of gaseous or short-lived soluble radiotoxic radionuclides, relate to:

- Waste envelope, including acceptable solids loading, maximum NAPL loading, etc;
- Mixing process and required order of addition of powders and other wastefrom constituents;
- Fluidity of grouts used to encapsulate the wastes;
- The expected modes and rate of waste and wastefrom ageing and degradation;
- Research and development work used to demonstrate the expected performance and evolution of the waste packages.

5 GUIDANCE ON IMMOBILISATION OF ACTIVITY IN WASTEFORMS

This section provides guidance on the key criteria of relevance to immobilisation of activity in wastefroms by examining each of the following potential sources of mobile radionuclides;

- non-aqueous phase liquids (NAPLs);
- loose particulates;
- free liquids;
- gaseous radionuclides;
- soluble short-lived radionuclides, and;
- complexants.

and considers the wastes with which they may arise, and ways to treat and package them in order to achieve adequate immobilisation.

Each sub-section is headed by a list of the relevant sections from the Wastefrom Specification of the GWPS, which is reproduced in full in Appendix A.

5.1 Non-aqueous phase liquids

Relevant GWPS Criteria	
6.1.1	Immobilisation of radionuclides and particulates
6.1.5	Non-aqueous phase liquids
6.2.4	Homogeneity/uniformity

Examples of NAPLs that may arise as ILW are oils (e.g. scintillation, lubricating and cutting oils), tributyl phosphate (TBP), odourless kerosene (OK), toluene and other solvents, e.g. cleaning solvents. NAPLs may arise as a waste stream in their own right, or be mixed with

⁸ When radioactive waste is disposed of in an operational waste repository in the UK, waste packages are required to meet WAC, produced and issued by the facility operator, having been prepared in conjunction with the relevant regulatory authorities.

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other wastes as part of a slurry, or as a contaminant on solid wastes such as scrap machinery. Some NAPLs are stored with sorbers such as vermiculite. NAPLs may also be formed as a result of the evolution of the waste, e.g. by the radiolysis of plastics.

The GWPS states that:

'non-aqueous phase liquids such as oils, shall be removed from wastes prior to packaging wherever practicable or immobilised'.

In some cases removal may be effected by draining, removal of NAPL-bearing fractions, liquid-liquid extraction or volatilisation.

NAPLs that cannot be removed from wastes are required to be immobilised because:

- They can increase the mobility of radionuclides, and therefore constitute a readily mobile source of radioactivity in the event of a loss of packaging integrity;
- They are subject to more rapid microbial degradation if not conditioned;
- They potentially represent a separate pathway for the return of radionuclides from a deep repository to the human environment as migration might be more rapid than for radionuclides in the groundwater pathway.

NAPLs pose a particular packaging challenge because they are not miscible with water, which can result in their separation from the remainder of the wastefrom following packaging. The immobilisation of NAPLs has been demonstrated, however, using conventional cementitious grouts in which the NAPL is dispersed throughout the matrix and physically trapped within the cement pore structure [11]. Experience to date suggests that this method may result in low NAPL concentrations (i.e. <10% v/v) which suggests that, wherever practicable, the volume of NAPLs requiring packaging should be minimised. In cases where NAPLs are present only as a minor constituent of a waste, this may prove unnecessary, since the total waste loading may not be unduly constrained by the presence of oils.

Work carried out to investigate the suitability of polymer based absorption systems for oils has suggested that effective immobilisation of loadings of greater than 10% v/v may be possible [12]. The efficacy of such a processes would, however, have to be demonstrated for each particular oil type as part of supporting R&D work for a LoC submission.

5.1.1 In-drum Grouting

NAPLs that are present as a contaminant on solid waste, but cannot be removed will need to be immobilised by the conditioning agent.

Wastefroms containing NAPLs within voids that are surrounded by the conditioning matrix, rather than infiltrated, may not be acceptable because of the potential for migration and release from the surface of the wastefrom. Wastes that may include isolated regions containing NAPLs are gearboxes, hydraulic cylinders and pumps. In general, such isolated regions should be opened or treated in such a way that they can be drained and infiltrated. The actual approach will depend on the volume of the voids and/or the nature and quantity of the enclosed NAPLs.

Some oily ILW is stored with sorbers such as vermiculite. It will need to be demonstrated that for such wastes that the oil is retained within the product following conditioning. Experience to date suggests that grout-infilling such wastes is unlikely to achieve the degree of infilling required for such retention.

5.1.2 Compaction and Grouting

Wastes contaminated with trace quantities of NAPLs may be suitable for supercompaction if it can be demonstrated that the liquids will be adequately immobilised by the compacted waste. NAPLs present in wastes presented for supercompaction may be displaced during

compaction. Any NAPLs remaining in the compacted puck would be present within the residual voidage in the puck or sorbed into solids and may not, therefore, be immobilised. However, residual quantities would be small, and it may be possible to demonstrate that the form of the waste in the pucks is adequate to provide containment. It should be noted that the migration pathway for oil through cement will be relatively short for annular grouted wasteforms, so that retention within the pucks may be particularly important.

Wastes associated with larger quantities of oil are unlikely to be suitable for compaction unless a process is available for the collection and treatment of oil displaced from the waste during compaction. For this reason some compaction facilities may impose limits on the presence of free liquids, including NAPLs.

5.1.3 In-drum Mixing

It has been shown that oils and solvents can be immobilised using in-drum mixing with cementitious conditioning agents [11]. Waste loadings at which satisfactory products have been achieved are low, typically less than 8-10 wt%. At higher loadings, seepage of free oil from the product, as well as increased viscosity and extended setting times, have been observed.

For NAPLs arising as a separate stream or as part of a nominally liquid waste (i.e. sludge, slurry, liquor or floc), the addition of an appropriate emulsifier may allow the formation of a stable water-oil emulsion and encapsulation by in-drum mixing with cementitious materials will provide adequate immobilisation of NAPLs (e.g. [13]). The method used to form the emulsion appears to be critical, as there is the potential for near-immediate separation of the aqueous and non-aqueous phases following cessation of mixing. If emulsifiers are used, their effect and that of their degradation products on the chemical containment of the waste package under repository conditions should be considered. The persistence of the beneficial effects of the additives must also be considered, as loss of emulsion stability could result in coalescence and increased NAPL mobility. However, it should be noted that emulsifiers are expected to act by dispersing small droplets of NAPL more uniformly throughout the wasteform, in which state the mobility of NAPLs will be restricted by the physical properties of the matrix.

The 'residual oil content' of a solid is considered to be that oil that cannot be removed unless the properties of the oil or the matrix/fluid interactions are altered. Increasing the residual oil content of a cementitious matrix will, therefore, allow for higher oil loadings without seepage. Residual oil content is dependent on a number of factors such as matrix porosity, matrix permeability, oil droplet size and the properties of the oil. Some of these factors are controllable. It may be practicable to optimise the residual oil content of wasteforms, such that the potential for loss of oil is eliminated. For example, the mobility of NAPLs in the matrix can be limited by increasing viscosity, which can be achieved by heating to remove volatile components. The efficacy of such an approach would need to be assessed on a case-by-case basis.

For oils held on sorbers, in-drum mixing may constitute a practical packaging option. The majority of oil sorbed by vermiculite would be released on mixing, and it should be demonstrated that all oil is adequately trapped within the wasteform matrix. The effects of the degradation of sorbers that are shown to retain oil following mixing should be considered, since release could result in unimmobilised NAPLs. For sorbed oils arising as a relatively small volume component of a larger waste stream, it may be appropriate for the oil and any associated sorber to be segregated for separate treatment, e.g. mixed with a cement in smaller containers, and for these to be grouted as for other solid wastes.

5.2 Loose particulates

Relevant GWPS Criteria	
6.1.1	Immobilisation of radionuclides and particulates
6.2.4	Homogeneity/uniformity

Loose particulate material can be defined as unimmobilised finely divided solids which could, under certain circumstances, constitute a mobile source term. The GWPS defines 'suspendible' particulates as those with aerodynamic diameters of up to 100µm⁹.

IAEA guidance on the requirements and methods for low and intermediate level waste package acceptability [14] states that:

'...waste forms¹⁰ that promote dispersion in the event of a release (respirable particles)....should be prohibited in the waste package.'

Specifically, it states that particles with diameters less than 200µm must be 'stabilised' or immobilised.

The presence of unimmobilised loose particulate in a wastefrom potentially provides a source term for the release of activity under both normal and accident conditions. Furthermore the presence of regions of such material in a wastefrom is also inconsistent with the requirement for wastefrom homogeneity and could also adversely affect other properties of the wastefrom, such as mechanical strength, voidage and mass transport properties.

Examples of wastes that may be contaminated with or contain particulate material are:

- Bulk particulate waste streams, e.g. powders;
- Metals contaminated with associated corrosion products in the form of solid particulates or sludges;
- Mixed wastes associated with particulate or other mobile contaminants; for example, plutonium contaminated materials (PCM) and filters;
- Cans, tins, packets, closed sources¹¹ or other closed containers containing particulate deposits;
- Sludges or other nominally 'liquid' wastes containing particulates in suspension.

Wherever practicable, and regardless of size, all the individual components of wastes including radionuclides in particular form should be intimately encapsulated by the conditioning agent to ensure that the waste package is consistent with the requirement for immobilisation unless a case can be made for non-encapsulation.

A particular example might be particulates contained within cans or similar closed containers. If such containers can be shown to be of sufficiently robust construction, and arguments made to demonstrate that the release of activity in suspendible form will be less

⁹ It should be noted that not all material deemed to be 'suspendible' is also 'respirable' and as such would contribute to internal dose to exposed persons. Respirable particles are defined by the GWPS as having a diameters of up to 40µm.

¹⁰ In this quotation the term 'waste forms' (two words) refers to waste in its original or 'raw' state. As such it does not refer to conditioned 'wastefroms' (one word) as used in this guidance.

¹¹ See also *Guidance on the Packaging of Closed Sources, WPS/906*.

than the accident release criteria specified in the GWPS (see Table 2), it may be possible to make a case for leaving such particulates in unconditioned form.

5.2.1 In-drum Grouting

Grouting is expected to achieve immobilisation of particulates so long as the grout is sufficiently fluid to infiltrate the individual components of the waste, including any intricacies inherent in the waste, e.g. pleats in filter media, closely packing wastes, and narrow tubes and pipes.

Inadequate infiltration of intricate solid wastes could leave voids adjacent to mobile waste fractions, or unconditioned pockets of bulk particulate. Such regions are inconsistent with the requirement for the minimisation of loose particulate material, as well as the requirements for:

- the release of material from wasteforms to be progressive and predictable under accident conditions;
- minimisation of voidage;
- wasteform homogeneity and uniformity.

In the case of metal wastes contaminated with particulate material, including particulates arising from the degradation of the waste during storage, adequate immobilisation will be achieved if the waste is sufficiently infiltrated by the encapsulating grout such that voidage is minimised and there are no unfiltered pockets of bulk particulate. This will usually require that coherent bodies of loose particulate are not present on the surface of a solid waste.

The use of a suitably fluid grout formulation and of processes such as vibro-grouting have been adopted previously to aid the infiltration of intricate solid wastes by cementitious grouts. A range of options for obtaining suitably fluid cementitious grouts are available including, for example:

- correct selection of grout water/cement ratio;
- controlling the particle size of the cement powders;
- the use of ternary formulations (blends of ordinary Portland cement/pulverised fuel ash/ground granulated blast furnace slag (OPC/PFA/BFS)).

Particulates that are less dense than the fresh grout will tend to float and could therefore create a waste-rich region at the wasteform surface. The effect of such a lack of homogeneity on the performance of the wasteform will depend on the quantity and characteristics of the particulates. If the particulates are bound by the encapsulating matrix, then the addition of a capping grout would be expected to ensure adequate immobilisation of the fines. However, if fines are left free at the wasteform surface, then they may float to the top of the capping grout. This could have a significant effect on the accident performance of the waste packages where the material released from the package tends to come from the upper region of the wasteform.

The addition of a pre-mixed grout to bulk particulate waste streams is considered unlikely to result in an acceptable product as the grout may not penetrate the bulk solids. Consequently, other options are more likely to yield a product that is consistent with the requirements of the GWPS, e.g. slurring and in-drum mixing (see Section 5.2.3).

Grouting is not applicable to wastes that include isolated regions containing particulates, unless the voids are rendered infiltratable. Wastes incorporating closed regions that may contain particulates include cans, tins, packets, sealed sources and vacuum cleaner bags. The potential for making a case to leave the contents of such containers unimmobilised was discussed in Section 5.2, however if the activity associated with such particulates is too high for such a case to be made, some method of pre-treatment will be required.

The pre-treatment processes adopted to ensure the immobilisation of particulates within isolated regions will depend on the nature and quantity of particulates present, and the properties of the structure in which they are held. One option is for isolated regions, e.g. cans, to be opened or punctured in such a way as to allow them to be grouted as for other intricate solid wastes. The orientation of the can may be critical in ensuring adequate infiltration, such that encapsulation trials should establish whether this is the case and what measures are required to ensure that appropriate orientation is achieved and maintained throughout filling operations. Alternatively, shredding may be adopted to destroy the cans.

For isolated regions containing larger quantities of particulate, the addition of a grout may simply cover the particulates, rather than ensuring their immobility by infiltrating the waste and incorporating the particles within the solid matrix. In this case, it may be more appropriate for the particulates to be segregated from the bulk waste and mixed with a suitable conditioning agent, prior to the can being returned to the bulk waste feed and grouted as for other solid wastes.

For intricate, particulate filled regions, such as pleated filter membranes or Magnox swarf, with corrosion products and other particulate contamination, cementitious grouts will need to offer adequate fluidity to ensure infiltration, recognising that very fluid grouts may be subject to separation. In such cases, other encapsulants and processes may be appropriate, e.g. polymer injection. For soft wastes, such as sealed packets or vacuum cleaner bags, associated with a particulate source-term, pre-treatment options such as shredding or cutting may render a waste more suitable for grouting, in-drum mixing or supercompaction. As in all cases, if grouting of shredded soft wastes is pursued, adequate immobilisation of particulates would need to be demonstrated through encapsulation trials or by reference to examples of similar practises that resulted in acceptable wastefrom properties.

5.2.2 Compaction and Grouting

Successful compaction of wastes offers beneficial volume reduction, as well as effectively fixing particulates within the compacted waste mass and minimising voidage. The appropriateness of compaction for achieving immobilisation will depend heavily on the nature and quantity of the particulates and the properties of the resultant pucks. In addition, the structure and materials of construction of the container¹² in which the particulates are held may have an effect, e.g. if they are subject to rupture following compaction or rapid corrosion following grouting of the pucks. Additional constraints may be imposed by the availability of compaction facilities and the operational limits of the plant. Examples of typical restrictions on the wastes that are suitable for compaction are physical size, as limited by the dimensions of the compaction facility; and massive metallic items, which have the potential to jam inside the compactor, causing damage to the press and drum. Actual limits placed on wastes will depend on the specification of individual compaction facilities.

Compaction may be aided by the pre-treatment of wastes, e.g. cutting or shredding, such that the resultant pucks contain a more coherent waste.

The use of smaller-scale compaction may be appropriate for individual items of waste that are particulate-bearing and arise as a small volume component of mixed waste streams for which another packaging process is adopted.

If compaction is pursued, the expected volume of residual voidage will have to be quantified to ensure compliance with the GWPS requirements for the minimisation of voidage (see Section 5.2.1).

¹² Often referred to as a 'sacrificial container'

5.2.3 In-drum mixing

Nominally liquid wastes containing particulates, e.g. sludges, are typically encapsulated by in-drum mixing, whereby the waste solids are suspended by some process of agitation (i.e. stirring) and cement powders added to achieve a homogeneous solid waste. Wasteforms produced by this method are expected to be acceptable with respect to the immobilisation of particulates, given a suitable formulation and appropriate process parameters such as mixing time and rate. Bulk particulate waste streams packaged in this way are expected to be acceptable with respect to the immobilisation of particulates given an appropriate formulation and process.

Some bulk particulate wastes may be resistant to wetting, with the result that wasteforms may incorporate regions of unimmobilised particulate agglomerations. This undesirable wasteform characteristic can typically be avoided by vigorous mixing of the waste and encapsulant. In some cases, settled or compacted particulates may not be readily mobilised because of their rheological properties. There is the potential for unmixed particulates to settle near the base of wasteforms during the mixing phase in regions of low turbulence, i.e. near the base of the paddle.

The adequacy of mixing processes in terms of the incorporation of particulates within the encapsulating matrix should be demonstrated. This is usually achieved by simulant development work and physical sectioning of the product.

5.3 Free liquids

Relevant GWPS Criteria	
6.1.1	Immobilisation of radionuclides and particulates
6.1.4	Free liquids
6.2.4	Homogeneity/uniformity
6.6	Gas generation

Free liquids can be defined as aqueous liquids that are not bound by the solid matrix of a wasteform. Examples of wastes that may include or be contaminated with free liquids are:

- nominally liquid wastes including sludges, flocs and liquors;
- solid, absorbent wastes bearing liquids;
- solid wastes with free liquids held in isolated or closed regions, or;
- intricate solid wastes with free liquids trapped in interstices.

Free liquids could also be generated during waste packaging as bleed water from cementitious grouts and would typically be observed at the surface of the wasteform. Following completion of the packaging process water may enter the waste container via the lid joint or filter during decontamination of the external surfaces using high pressure water washing.

The presence of free liquids in wasteforms is specifically excluded by the GWPS, which states that:

'liquids shall be removed from wastes or immobilised by a suitable conditioning process'.

Free liquids within a wasteform are unacceptable because of the reduced predictability of the wasteform under normal and accident conditions, which is a result of:

- increased mobility of radionuclides by solution or suspension, resulting in a readily mobile source of radioactivity in the event of a loss of packaging integrity
- increased potential for deleterious evolution of the wasteform, e.g. enhanced microbial activity and corrosion.

Cementitious materials have a significant capacity for water and may continue to absorb water after the product has set and aged. It is recognised that the demonstration of the absence of small volumes of free liquids within the waste packages is not a simple exercise. Mechanisms such as wasteform shrinkage and condensation may provide a mechanism for the accumulation of water at the base edges of waste packages, albeit in small quantities. Inorganic cements, if well-formulated, can be used to ensure that any water is absorbed during hydration. Free liquids that could accumulate as a result of condensation during storage will therefore be limited in volume and would be expected to be reabsorbed into the wasteform as a result of continuing hydration.

5.3.1 In-drum Grouting

Free liquids that are present in intricate solid wastes may not be contacted by the grout and would, therefore, remain free. Quantities of free liquid associated with intricate solid wastes are likely to be small, as bulk liquids would be largely free-draining or removable using simple means such as a dewatering tube. The packaging process and formulation envelope should therefore provide adequate infiltration of the waste and be robust to variations in the volume of residual free liquid present, such that all free liquids are effectively immobilised by the grout. Infiltration by the encapsulating grout can be increased using a variety of measures, as discussed in Section 5.2.1. The waste packager would need to demonstrate that the packaging process consistently achieves infiltration of the waste and, therefore, the immobilisation of free liquids.

The presence of free liquids held within closed regions in the waste, for example cans, is not consistent with the requirements of the GWPS. It will therefore be necessary for any such regions to be opened and the liquid either immobilised in situ or removed from the container and treated separately. In situ immobilisation could be, for example, in-can mixing with cementitious powders. Emptied cans could contain residual liquid which would require immobilisation. Appropriate techniques for their treatment are as for other intricate solid wastes associated with free liquids.

5.3.2 In-drum mixing

Liquid and slurry wastes are typically encapsulated by in-drum mixing, whereby the waste solids are suspended by mixing and cement powders added to achieve a homogeneous solid waste. Wasteforms produced by this method are expected to be acceptable with respect to the immobilisation of free liquids, given a suitable formulation. Although wet wastes can be packaged in this way to give an acceptable wasteform, it is noted that reducing the quantity of free liquid by dewatering may offer a beneficial reduction in the volume of waste to be encapsulated as ILW.

The guidance presented in Section 5.2.3 regarding the in-drum mixing of bulk particulate wastes is also of relevance to nominally liquid wastes containing solids.

5.3.3 Compaction and Grouting

Some compaction facilities place restrictions on the presence of liquids, as compaction will result in the displacement of liquids from waste items. The tolerance of compaction facilities to liquids can be increased by the installation of liquid collection and treatment facilities.

Supercompaction may be appropriate for wastes associated with limited quantities of free liquids, notwithstanding the limitations imposed by some compaction facilities. It should be demonstrable that the pucks contain minimal voidage in which free liquids could collect and that, instead, any liquids are effectively displaced from the compacted waste or absorbed by the waste, and are therefore immobile. It may be appropriate to dry sludges and slurries and for the resulting dry waste to be compacted. The issues associated with the compaction of dried wastes are considered in Section 5.2.2.

5.4 Gaseous release

Relevant GWPS Criteria	
6.1.1	Immobilisation of radionuclides and particulates
6.6	Gas generation

Radioactive gases may be present in waste at the time of packaging, for example radioisotopes of Ar, Kr and Xe may be present in spent fuel and fuel cladding. As well as being present in the as-packaged waste, radioactive gases may also be generated by the waste as a function of radioactive decay, i.e. Rn-220 and Rn-222, which occur in the decay series of Th-232 and U-238. Other radiotoxic gases that may be released are labelled gases, in which radionuclides are incorporated into gaseous molecules, e.g. CO₂ and CH₄ labelled with C-14.

The GWPS states that:

‘radioactive gases shall be removed from wastes prior to packaging or immobilised by a suitable waste conditioning process’

and that:

‘gases generated after packaging should not compromise the ability to meet radioactivity release restrictions (or other aspects of the GWPS) for future stages of waste management’.

The GWPS sets limits on the release of radioactive gases from transport packages¹³ derived from the IAEA Transport Regulations [10] and by the transport arrangements which are part of the PGRC. Further limits are set on the quantity of bulk gases that can safely be released from packages based on the limits on pressure build-up in a shielded transport container. The limits for radioactive gases will be more restrictive, than the bulk gas release limits.

Radioactive gases represent a potentially mobile source of radionuclides such that their generation and release should be minimised, and within the defined release limits. Achievement of these requirements should not compromise the performance of the package in terms of its ability to release gases at such a rate that pressurisation of the conditioning matrix and consequent physical damage is prevented. Migration will be in two stages, where the first is the release of gases from the waste, and the second the movement of gases through the wasteform and out of the container.

Kr-85 is used in some sealed sources, which typically comprise capsules with a thin (25µm) stainless steel window and Kr-85 inventories in the range 37MBq to 37GBq. Ra-226

¹³ These comprise ‘unshielded’ waste packages (i.e. 500 litre Drums, 3 cubic metre Boxes and Drums) when carried in a transport overpack or ‘shielded’ waste packages (i.e. 2 metre and 4 metre Boxes) which are transport packages in their own right.

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sources, typically comprising a high integrity TIG welded stainless steel capsule with a Ra-226 inventory in the range 4MBq to 4GBq, will generate Rn-222 as it undergoes radioactive decay. Release from sources may occur as a result of breach of the source containment, or as a function of diffusion. Rates of diffusion through the solid are expected to be slow. Breach of the source will depend upon the environmental conditions and the properties of the containment. Consideration should be given to the period for which the source is expected to afford containment, based on the expected rate of degradation. The corrosion of stainless steel in intimate contact with an inorganic cement is expected to be slow; $<0.01\mu\text{myr}^{-1}$. After the containment is breached, the inventory of radioactive gas in the source and the properties of the encapsulating matrix will determine the rate of release from the package. Reference should be made to *Guidance on the Packaging of Sealed Sources*, WPS/906 for further information of this type of waste.

Tritium and other gaseous fission products such as krypton may be present in metallic wastes such as fuel cladding. These gases may be released following corrosion of the metal or through diffusion. The rate of release into the wasteform will therefore be dependent on the rate of corrosion and on the diffusivity of the solid waste as well as the wasteform.

Tritiated water present in the waste may be involved in the corrosion of metallic wastes and released as tritiated hydrogen or be subject to evaporation. Both tritiated hydrogen and tritiated water may be involved in the microbial degradation of organic wastes to produce tritium-labelled methane or hydrogen sulphide. Similarly, C-14 may be released from wastes as labelled carbon dioxide or methane as a function of microbial degradation.

Accordingly, the rate of release of H-3 and C-14 labelled gases, and of other radioactive gases present in solid wastes, into the wasteform will depend on the rate of degradation of wastes in which they are present. Gas generation as a result of corrosion and microbial degradation is slowed by intimate contact between the waste and inorganic cement and by minimising the availability of water. *Guidance on the Packaging of Tritium Bearing Wastes: WPS/907* and *Guidance on the Packaging of Carbon-14 Bearing Wastes: WPS/910* are of relevance here.

Once released into the wasteform matrix, migration would be controlled by the physical properties of the matrix and by the rate of release of bulk (inactive) gases. Gases may migrate through a wasteform as a function of advection; driven by a pressure gradient resulting from the generation of bulk gases; or diffusion; driven by a concentration gradient. Diffusion may be through a continuous gas or liquid phase, or solid-state. The rate of release of gases from a wasteform will depend on:

- The degree of hold-up afforded by the waste itself
- The distribution of the waste within the wasteform
- The mass transport properties of the wasteform, including any capping grout; e.g. permeability, porosity, interconnectivity of pores, extent of saturation
- The properties of the container, including vent and seal
- The rates of generation and release of bulk (non-radioactive) gases
- Environmental conditions (e.g. temperature and pressure)
- The properties of the gas (e.g. viscosity, compressibility, mean free path).

The half lives of a number of relevant gaseous radionuclides (i.e. Rn-220, Rn-222, Ar-37 and the Xe isotopes) are relatively short (i.e. up to a few tens of days) and, as a result, the release of these gases from waste packages will be significantly reduced if the degree of hold-up afforded by the wasteform and container is sufficient to allow the inventory to undergo radioactive decay. As the half-life of radioactive gases increases, the degree of hold-up offered by the wasteform will be less significant in reducing the releases of these

radionuclides, and the waste package inventory is likely to become the controlling factor. *Guidance of the Packaging of Radon-generating Wastes, WPS/901*, has been produced to address this particular issue.

For the purposes of assessment, therefore, information will be required on the typical and maximum package inventories of radioactive gases (where these exceed the threshold guidance quantities [15]) together with an estimate of their rate of release from the waste and wasteform. For H-3 and C-14, an indication of the mole ratio of active:inactive isotopes (e.g. C-14:C-12) and a quantification of the expected rate of bulk gas generation may be required for assessment.

Arguments seeking to demonstrate the performance of a proposed waste package in terms of limiting the release of radioactive gases should consider the following issues:

- provision of data regarding the properties of the packaging materials, obtained under relevant conditions;
- the generation of bulk gases;
- the evolution of waste and wasteform;
- the performance and evolution of the filtered vent;
- validation of models and arguments using experimental measurements of the release of gases from packages containing simulated waste.

5.5 Soluble, short-lived radionuclides

Relevant GWPS Criteria	
6.1.1	Immobilisation of radionuclides and particulates
6.2.3	Mass transport properties
6.3	Chemical containment

The GWPS states that BPM should be applied to the physical containment of soluble radionuclides. The treatment and packaging of waste, and in particular encapsulation in a matrix such as cement, provides a barrier to the release of radionuclides by controlling their mobility.

Containment can be quantified by the radiotoxicity of the repository pore solution arising from the soluble radionuclides released from the waste package. It is generally accepted that BPM has been applied if the physical form of the waste package provides containment of soluble, short-lived radionuclides until the radiotoxicity attributable to them has decayed to below the long-term average value of radiotoxicity expected within the repository.

Sr-90 and Cs-137 commonly dominate the estimated radiotoxicity of fresh wastes because of their solubility, abundance and biological impact. However, other radionuclides also may be important, depending on their specific activity in the waste, biological impact and chemical behaviour in the disposal environment. Advice may be obtained from Nirex on the radionuclides of importance for a particular waste. Where the inventories of soluble species are such that the radiotoxicity cannot exceed the long-term background value for a repository, performance will be acceptable even if no additional credit is taken for retention in the container. In this case, no further argument may be necessary to demonstrate that waste packages are consistent with BPM.

In general, where the radiotoxicity of a waste is relevant, immobilisation by intimate grouting or in-drum mixing will be beneficial.

5.6 Complexants

Relevant GWPS Criteria	
6.1.1	Immobilisation of radionuclides and particulates
6.3	Chemical containment

The mobility (solubility) of some radionuclides may be increased by forming complexes with other components of the waste. Compounds that are known to be complexants include ethylenediaminetetracetic acid (EDTA), and citric and oxalic acids. The degradation products of other waste or packaging components may also act to increase radionuclide solubility by complexation, e.g. cellulose. The addition of known or suspected complexants at the point of waste generation (e.g. decontamination chemicals), and during waste treatment and waste packaging (e.g. superplasticisers or other cement additives) should be avoided. Complexants within the waste should be removed wherever practicable, or packaged in such a way that the effects are minimised. The biodegradation of organic materials is slowed if the quantity of water available is minimised and if the waste is in intimate contact with inorganic cement. As well as biodegradation, alkaline hydrolysis is also expected to result in the formation of complexing degradation products from cellulose and, to a lesser extent condensation polymers such as nylon and epoxy resins. The wasteform loading of these materials may need to be limited in order to ensure adequate performance. This will be assessed on a case-by-case basis by Nirex. Release of radionuclides will be minimised if the mass transport properties of the wasteform and container reduce the flux of groundwater through the package in a repository. This can be achieved by intimate encapsulation.

5.7 Demonstration of Immobilisation

The demonstration of the adequacy of an approach to the immobilisation of activity in a waste will be an important part of demonstrating the overall compliance of a waste package with the requirements of the GWPS in general. Such demonstration is likely to be underpinned by the results of R&D work involving small and/or large scale inactive and/or active wasteform trials. The demonstration of the adequacy of wasteform characteristics in this way is discussed in *Guidance on the Characteristics and Demonstration of Robust Formulation Envelopes for Cementitious Wasteforms, WPS/904*. Such R&D work may be supported by evidence obtained during the non-active commissioning of the as-built packaging plant.

For wastes containing NAPLs the success of an approach to immobilisation may be argued to be a demonstration that NAPLs present in the waste are uniformly distributed throughout the wasteform matrix. A range of techniques may be applicable to the determination of the distribution of NAPLs within a matrix and the ability of that matrix to ensure its immobility. Simple inspection or swabbing of the outer surfaces of NAPL wasteform simulants would indicate whether there was gross leakage or separation. Depending on the properties of the NAPL, techniques based on the use of UV fluorescence may be applied to external or cut surfaces of samples to show the presence and distribution of NAPLs within the matrix; mineral oils fluoresce under UV light. High-pressure elution tests as used in the oil industry can be applied to demonstrate the ability of the matrix to retain NAPLs. Such tests require the injection of water into a core, and the collection and analysis of any eluted fluids. Previous experience has shown that suitably formulated samples will not release oil after 6

month water injection at pressures of around 5MPa. Oil retention can also been investigated using a pore water press, whereby samples are subjected to a triaxial compressive force and the pore fluids extracted for analysis.

In cases where the immobilisation of particulates is dependant on the infiltration of intricate wastes, and therefore on the elimination of voidage, the volume of air-filled voidage can be calculated from the known weight of waste present in the specimen, the density of the waste and the density of the grout. This approach to the measurement of voidage relies on a knowledge of waste density, and may therefore have limited applicability for mixed wastes. Grout density can be readily measured using a deaerated standard specimen. Such a technique eliminates the effect of any other, pore-related voidage within the cement phase that is a function of, for example, cement hydration reactions. In all cases, simulant waste packages can be sectioned to examine for gross voidage. Regardless of which technique is used, full-scale demonstration is likely to be required where there is any doubt surrounding the ability of the chosen grout to completely infill the waste, and particularly where processes such as vibro-grouting are to be adopted.

There are non-intrusive techniques available for inspecting waste packages for gross voidage. Techniques such as high energy radiography together with computer tomography or real time radiography can be used to inspect the waste package for evidence of defects such as gross voidage, gross cracking, particulate, evidence for corrosion of metallic wastes and evidence of free liquids. These techniques are, however, relatively expensive.

APPENDIX A WASTEFORM SPECIFICATION OF THE GWPS

The ensuing is taken directly from Volume 1 of the GWPS [2] and contains the wasteform specification criteria, to which extensive reference is made in Sections 4 and 5 of this guidance. For convenience, the numbering of the sub-sections given here is the same as that in the GWPS.

6 WASTEFORM SPECIFICATION

The production of the wasteform is the means by which the original 'raw' waste is rendered passively safe, so its design can have a significant influence on waste package performance under both normal and accident conditions. The parameters that could affect the quality of the wasteform, and thus its ability to meet any aspect of the GWPS, should be identified and limits for their control established. This Section lists the requirements that comprise the wasteform specification.

The wasteform requirements are grouped under six headings:

- physical immobilisation;
- mechanical and physical properties;
- chemical containment;
- hazardous materials;
- wasteform evolution;
- gas generation.

The detailed requirements given in this Section are developed from the high-level wasteform criteria defined in Section 4.7 of the GWPS.

These requirements apply to all phases of the PGRC but shall be applied at the time of transport from the waste packager's site to the repository, unless otherwise stated.

The rationale and justification for these requirements are given in Volume 2 of the GWPS.

6.1 Physical Immobilisation

The wasteform shall be designed to immobilise radionuclides and toxic materials so as to ensure appropriate waste package performance during all phases of the PGRC. For many wastes, this immobilisation requires the use of an encapsulating matrix.

6.1.1 Immobilisation of Radionuclides and Particulates

All reasonable measures shall be taken to ensure that radionuclides and toxic materials in the waste are immobilised and that loose particulate material is minimised.

6.1.2 Response to an Impact Accident

All reasonable measures shall be taken to ensure that, in the event of an impact accident, the quantity of potentially mobile radionuclides present within the waste package, including those mobilised as a result of the impact accident, is commensurate with the waste package meeting the relevant activity release limits specified in Section 5 of the GWPS.

6.1.3 Response to a Fire Accident

All reasonable measures shall be taken to ensure that, in the event of a fire accident, the quantity of potentially mobile radionuclides present within the waste package, including those mobilised as a result of the fire accident, is commensurate with the waste package meeting the relevant activity release limits specified in Section 5 of the GWPS. In addition, the wasteform should not readily burn or otherwise support combustion.

6.1.4 Free Liquids

All reasonable measures shall be taken to exclude free liquids from the wasteform. This should include consideration of materials that may degrade to generate liquids. Free liquids not removed from wastes prior to waste packaging should be immobilised by a suitable waste conditioning process.

6.2 Mechanical and Physical Properties

The wasteform shall be designed to provide the mechanical and physical properties necessary to ensure appropriate performance of the waste package during all phases of the PGRC.

6.2.1 Mechanical Strength

The wasteform shall provide sufficient mechanical strength to allow the waste package to be transported and handled without compromising the ability of the waste package to meet any aspect of the GWPS.

6.2.2 Voidage

All reasonable measures shall be taken to ensure that the volume of voidage within the waste package (such as ullage space and other holes or spaces) is minimised.

6.2.3 Mass-transport Properties

The wasteform shall be sufficiently permeable to allow gases generated within the wasteform to be released without compromising the ability of the waste package to meet any aspect of the GWPS.

The mass transport properties of the wasteform (e.g. diffusivity and permeability) shall provide best practicable means for containment of water-soluble radionuclides within the waste package.

6.2.4 Homogeneity

Local concentrations of materials within the wasteform that may compromise the ability of the waste package to meet any aspect of the GWPS should be avoided.

6.2.5 Thermal Conductivity

The thermal conductivity of the wasteform shall be sufficient to dissipate any heat generated within the waste package, when emplaced in the repository, without unacceptable temperature rise. The minimum value of thermal conductivity should be $0.5\text{Wm}^{-1}\text{K}^{-1}$.

6.2.6 Leachability

Wasteforms categorised as LSA-III material shall be sufficiently insoluble as to satisfy the requirements of Paragraph 226 (c) (iii) of the IAEA Transport Regulations.

6.3 Chemical Containment

The wasteform shall not be incompatible with the chemical containment of radionuclides and hazardous materials as embodied in the PGRC.

Where they may inappropriately affect chemical containment, the following items should not be introduced through waste conditioning or packaging, and their presence in wastes should be minimised wherever practicable:

- Oxidising agents;
- Acids and/or materials that degrade to generate acids;
- Cellulose and other organic materials;

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- Complexants and chelating agents, and/or materials that degrade to generate such compounds;
- Non Aqueous Phase Liquids (NAPLs) and/or materials that degrade to generate them;
- Any other materials that could detrimentally affect chemical containment.

6.4 Hazardous Materials

The wasteform shall not contain hazardous materials, or have the potential to generate such materials, unless the conditioning of such materials or items makes them safe. The means by which any of these materials is made safe shall be demonstrable for all relevant phases of the PGRC.

6.5 Gas Generation

Gases generated by the wasteform shall not compromise the ability of the waste package to meet any aspect of the GWPS.

6.6 Wasteform Evolution

Changes in the characteristics of the wasteform as it evolves shall not result in degradation that will compromise the ability of the waste package to meet any aspect of the GWPS.

The deleterious effects of the following processes should be considered:

- dimensional changes, e.g. shrinkage;
- corrosion including, but not limited to, the production of gases and particulate material, and wasteform expansion resulting from the formation of lower density solid corrosion products;
- microbial activity;
- self-irradiation and irradiation by surrounding waste packages;
- heat generation by the wasteform and its surroundings including, but not limited to, localised heat sources within the wasteform, the effects on the curing of the encapsulant material and the consequential effects on longer-term performance.

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