WASTE PACKAGE SPECIFICATION AND GUIDANCE DOCUMENTATION

WPS/907: Guidance on the Packaging of Tritium Bearing Wastes

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Corporate Communications Administrator

United Kingdom Nirex Limited

Curie Avenue

Harwell

Didcot

Oxfordshire

OX11 0RH

UK

Or by e-mail to: info@nirex.co.uk

WASTE PACKAGE SPECIFICATION AND GUIDANCE DOCUMENTATION

GUIDANCE ON THE PACKAGING OF TRITIUM BEARING 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.

CONTENTS

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[2].

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

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

[2] For the remainder of this document references to ILW shall include those categories of LLW that are assumed to be subject to geological disposal in the UK.

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 challenging wastes, Nirex has produced, and continues to add to, a suite of thematic 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.*

Tritium is to be found at a wide range of concentrations in ILW streams in the UK, and several of the characteristics of the isotope make the management of tritium-bearing wastes a particular challenge. These include the propensity for tritium to be released from wastes and waste packages as a gas, its mobility and high residence time in the environment, the high isotopic exchange rate of tritium, and its ease of assimilation into living matter.

The chemical form of tritium in wastes varies, and influences the behaviour of tritium after wastes have been packaged. The relatively short half-life of tritium (i.e. 12.3 years) means that it is not a significant issue for the later stages of waste management (i.e. beyond ~100 years). However, the mechanisms for tritium release from wastes and waste packages are complex and not easily understood or predicted. Accordingly specific consideration needs to be given to the features of waste packages that are designed to reduce rates of tritium release following waste conditioning and packaging.

This Guidance Note is intended to assist waste packagers with the presentation of robust arguments regarding the performance of packages containing tritium-bearing wastes, and to facilitate their safe and efficient long-term management.

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

2 BACKGROUND

2.1 Scope of this Guidance Note

The development of suitable packaging methods for tritium-bearing wastes, and the demonstration of the suitability of a proposed method, requires attention to particular aspects of waste package and wasteform performance. Therefore, this guidance identifies the issues arising from the presence of tritium in waste packages, their implications for the PGRC, the methods that could be used to develop robust arguments, and the supporting information that could be used to support the presentation of packaging proposals to Nirex.

This section provides summaries of the Nirex PGRC, the LoC Assessment Process[4] and the Nirex approach to setting standards and specifications for the packaging of ILW which form the basis for the LoC process.

Section 3 summarises the properties of tritium, particularly as they would influence the long-term management of ILW.

Section 4 provides information regarding the sources and uses of tritium and identifies the waste streams in which tritium is present in significant quantities and/or concentrations.

Section 5 derives 'Guidance Quantities' for tritium in waste packages to allow the identification of those waste streams that may warrant specific treatment to deal with the hazards presented by the presence of tritium.

Section 6 discusses strategies that can be adopted in the packaging of waste bearing significant quantities of tritium.

Section 7 outlines how arguments can be presented to show that a chosen packaging approach for a particular tritium-bearing waste stream will provide adequate waste package performance.

A large number of documents were consulted in the production of this Guidance Note. Specific references are, where appropriate, provided throughout and a bibliography of the detailed technical documents consulted is also provided. A glossary of the terminology adopted in this Guidance Note is also included.

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, forms the basis for Nirex waste packaging standards and specifications which form 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

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

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 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 operators 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. The process of the production of waste package specifications 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 the repository, 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;

- the range of Nirex standard waste containers;

- specific requirements for the standard waste package design 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 that the generation and release of radioactive gases should not compromise the ability of a waste package to be the subject of long-term management in accordance with the PGRC. The GWPS defines limits on the allowable release of such gases determined following assessment of all phases of the PGRC.

The GWPS also requires that wasteforms provide effective immobilisation of activity, be it in particulate, liquid or gaseous form and that wasteforms, in which activity was originally rendered immobile, do not evolve in such a manner as to create mobile activity.

Effective immobilisation of gaseous 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.

Additionally, two of the three safety assessments that help underpin the PGRC (i.e. the Generic Transport Safety Assessment (GTSA) [4] and the Generic Operational Safety Assessment (GOSA) [5]) rely on the requirement that radionuclides, in all forms including gases, are effectively immobilised within wasteforms, so that release of activity under normal and 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 limiting the release of radioactive gases.

3 PROPERTIES OF TRITIUM

There are three naturally occurring isotopes of hydrogen:

- 'ordinary' hydrogen, sometimes referred to as protium (symbol ^{1}H or P), is a stable isotope and the most abundant element in the universe;

- deuterium, sometimes called heavy hydrogen (symbol ^{2}H or D), is also a stable isotope;

- tritium (symbol ^{3}H or T) is radioactive.

The natural abundances of the three hydrogen isotopes are given in Table 1.

Table 1   Natural abundances of hydrogen isotopes

Isotope	Symbol	Natural abundance (%)
Hydrogen	^{1}H or P	99.985
Deuterium	^{2}H or D	0.015
Tritium	^{3}H or T	1E-18

3.1 Radiological Properties

Tritium decays to helium-3 (^{3}He) by emitting a low-energy beta particle (electron) and a neutrino. The total decay energy is constant (18.6keV), but is shared between the beta particle and neutrino in varying proportions. Consequently, the energy of the beta particle produced by tritium decay varies from 0 to 18.6keV with an average energy of 5.7keV.

The generally accepted half-life of tritium is 12.3 years which leads to a specific activity of 353TBq/g. The value of the A_2[5] multiplier for tritium is 40TBq.

3.2 Physical and Chemical Properties

In many respects, the chemical and physical properties of compounds of tritium are the same as the analogous compounds of hydrogen. However, the higher atomic mass of tritium compared to hydrogen, and the radioactive nature of tritium, do result in some differences in chemical properties.

The most commonly encountered chemical forms of tritium are tritium gas, tritiated water, metal tritides and organically bound tritium (OBT – see Section 3.2.4). Tritium gas can take the form of molecules containing two atoms of tritium (chemical symbol T_2) or one atom of tritium and one of hydrogen (chemical symbol HT, also referred to as tritiated hydrogen).

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[5] A_2 is a measure of activity defined in the IAEA Transport Regulations, that applies to the entire range of radionuclides and is linked to radiotoxicity and possible exposure pathways.

Similarly, tritiated water can take the form of T_2O (sometimes referred to as tritium oxide) or HTO.

3.2.1 Tritium Gas

The physical properties of tritium gas are very similar to those of hydrogen; it is a colourless, odourless gas with a low density. Like hydrogen, tritium gas has a high diffusivity, and can readily diffuse through many materials considered impermeable to other gases (*e.g.* metals).

The chemical bond in molecular hydrogen is relatively strong, which means that hydrogen tends to be unreactive at room temperature. The reactivity of tritium gas at room temperature is, however, greater than that of hydrogen, due to the reactive chemical species produced by radioactive decay of the tritium. For example, tritium gas will react with the oxygen in air at room temperature to produce tritiated water. Similarly, tritium gas will react with many organic materials that are inert towards hydrogen at room temperature, resulting in the production of OBT. A further consequence of the reactivity of tritium gas is that, when it is brought into contact with ordinary water, tritiated water is formed.

Like hydrogen, tritium gas is readily absorbed by a number of metals. Hydrogen embrittlement can have a deleterious effect on the mechanical properties of metals (most importantly stainless steels) that have been in contact with hydrogen. This problem can be exacerbated with tritium gas due to the additional effects of helium embrittlement (helium being introduced into the metal by either tritium absorption, diffusion and decay, or by implantation during radioactive decay). It should be noted, however, that the concentrations of tritium in ILW are so low as to make such effects insignificant in the context of the packaging of such wastes.

3.2.2 Tritiated Water

The chemical and physical properties of tritiated water are in many respects very similar to those of ordinary water. The production of reactive chemical species as a result of radioactive decay makes tritiated water more reactive than ordinary water, which can result in the transfer of tritium into materials brought into contact with tritiated water. The propensity of water to form hydrogen bonds provides a further mechanism for the transfer of tritium from tritiated water into other materials.

Self-radiolysis and corrosion reactions with metals can result in the production of tritium gas from tritiated water.

3.2.3 Metal Tritides

Metal tritides are commonly used as a convenient means of reversibly storing tritium gas. Finely divided forms of some metals react readily with tritium gas to form metal tritides; subsequent heating results in thermal decomposition of the metal tritide and liberation of tritium gas.

Metals that are commonly employed for this purpose are uranium, palladium, titanium and zirconium. Some of these finely divided metals and their tritides are pyrophoric and reactive with water.

3.2.4 Organically Bound Tritium (OBT)

Common organic materials such as plastics, rubbers and oils can become tritiated when brought into contact with tritium gas or tritiated water. In addition, a vast array of tritium-labelled organic compounds are manufactured for use in chemical and biochemical research. Tritium that has become incorporated into organic molecules by bonding to oxygen, sulphur, phosphorus or nitrogen atoms in organic molecules is conventionally termed exchangeable OBT, since it is readily able to participate in isotopic exchange reactions. Tritium that has been introduced into an organic molecule by bonding to the carbon chain of the molecule is known as non-exchangeable OBT.

Self-radiolysis and radiolysis resulting from the presence of other contaminating radionuclides can result in the production of tritium gas and other tritiated gaseous species (*e.g.* tritiated methane) from tritiated organic compounds.

3.3   Biological Properties

Transfer of environmental tritiated water to humans takes place *via* inhalation, diffusion through the skin and ingestion. Inhalation is the only meaningful pathway of tritium gas to humans. The body does not readily absorb tritium gas; a small fraction of that inhaled is absorbed in the blood stream and then exhaled after a few minutes. Tritiated water is readily retained in the body and remains with a biological half-life of approximately 10 days. As a result, exposure to tritiated water in air is up to 25,000 times more hazardous than exposure to gaseous tritium. Immersion of the body in tritiated water, most commonly in the form of tritiated water vapour in air, is also a significant pathway.

OBT has a longer retention time than tritiated water, as it is incorporated into a variety of biochemical compounds. These longer retention times confer a greater radiotoxicity on OBT compared with tritiated water.

3.4   Why Tritium is Significant in the Long-term Management of ILW

3.4.1 Transport Safety

Releases of tritium in a gaseous form (as either T_2/HT, tritiated water vapour or tritiated gaseous compounds such as CH_4) 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 [6] 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 consequences of the IAEA Transport Regulations on the PGRC is explored in the GWPS [2] and this is dealt with in Section 5.1 for the transport of packages containing tritium-bearing wastes. In addition to national and international legislation, the overall transport operation should result in releases of activity that are As Low As Reasonably Practicable (ALARP).

3.4.2 Repository Operational Safety

Exposure to tritium could occur to workers within a repository (or in a Waste Packager's surface store prior to transport to a repository) and to members of the public through discharges from a repository, as a result of the release of tritium in a gaseous form from waste packages. Such exposures could arise as a result of routine operations in a repository and in the event of fire and impact accidents involving waste packages containing tritium-bearing wastes. The GOSA [5], one of the safety assessments

supporting the PGRC, has examined the consequences of the discharge of gaseous activity, including tritium, 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 is dealt with in Section 5.2.

3.4.3 Repository Post-closure Safety

Public exposure to tritium could occur following the closure of a repository *via* three different routes: the release of tritium in the gaseous form (the gas pathway), the release of tritium in the dissolved form (the groundwater pathway), and as a result of human intrusion into the repository (the human intrusion pathway). The risks from these three routes have been assessed in the Generic Post-closure Safety Assessment (GPA) [7] and is dealt with in Section 5.3.

4   PRODUCTION, USES AND QUANTITIES OF TRITIUM

4.1   Worldwide Production and Uses

Tritium is produced naturally by the interaction of high energy cosmic rays with oxygen and nitrogen in the upper atmosphere. The tritium converts into tritiated water and reaches the Earth's surface as rain. The estimated rate of production of tritium in the upper atmosphere is 1.48×10^5TBq/year, resulting in a worldwide steady-state natural tritium inventory of 2.6×10^6TBq [8].

Atmospheric nuclear weapons testing from 1945 to 1975 added approximately 3×10^8TBq of tritium to the environment, much of which has since decayed; however, about 2×10^7TBq remain in the environment, mostly diluted in the oceans [8].

Tritium is 'adventitiously' produced in fission reactors by two main routes: activation of impurities in reactor fuel and structural materials, and ternary fission. In addition, tritium is deliberately produced by the irradiation of targets in fission reactors. The total worldwide rate of adventitious tritium production in fission reactors is approximately 1.4×10^6TBq/year, of which an estimated 3×10^5TBq are released into the environment [8, 9]. Total Worldwide deliberate production of tritium in fission reactors is approximately 1.5×10^4TBq/year [10].

The most significant use of tritium that has been deliberately produced in fission reactors is in nuclear weapons. An increasingly important use of tritium is as a fuel in fusion reactors. Tritium also has a number of commercial applications, including use in self-luminescent devices and biochemical research.

4.2   Production and Uses in the UK

Tritium has been, and continues to be, adventitiously produced in nuclear fission reactors in the UK. Three types of reactor have dominated the commercial production of nuclear energy in the UK: the Magnox reactor, the Advanced Gas Cooled Reactor (AGR) and the Pressurised Water Reactor (PWR). In addition, a variety of other different types of reactor have been operated in the UK for research, development and defence purposes.

Tritium is no longer deliberately produced in UK fission reactors. Until recently, tritium was produced by the irradiation of targets in the Magnox reactors at Chapelcross. Historically, tritium was also deliberately produced in other UK fission reactors.

4.2.1   Tritium Production and Behaviour in the Magnox Fuel Cycle

Tritium is produced adventitiously by two processes in a Magnox reactor core: neutron activation of impurities in the graphite moderator, and ternary fission in the uranium metal fuel.

The neutron activation of lithium impurities in the graphite moderator is the main source of tritium during the early generating life of a Magnox reactor, but declines in importance with time as the impurities are consumed. Tritium production from ternary fission is directly proportional to power loading and, therefore, remains a significant source of tritium throughout the generating life of a Magnox reactor.

The majority of the tritium produced in the graphite moderator of Magnox reactors is retained therein, with only a small fraction being released into the carbon dioxide reactor coolant. Similarly, the majority of the tritium produced in the uranium metal fuel is retained therein; a fraction diffuses into the Magnox fuel cladding and from there into the carbon dioxide coolant.

Moisture levels in the carbon dioxide coolant of Magnox reactors are minimised by the use of desiccant, through which the coolant is passed. Tritiated water, along with inactive water, is captured on the desiccant which is periodically regenerated by heating to drive off its tritiated water burden. The vapour is then condensed and discharged to the environment as an aqueous effluent whilst any residual tritium in the reactor coolant is discharged as a gaseous effluent. This process does not remove all of the tritium from the desiccant which will have to eventually disposed of as ILW.

In the reprocessing of Magnox fuel, the cladding is mechanically removed prior to dissolution of the irradiated uranium metal. The Magnox swarf from the de-cladding process, with its associated tritium inventory, is conditioned for storage as ILW. The great majority of the tritium in the uranium metal fuel appears as tritiated water in the solution produced in the fuel dissolution process, with only a small fraction of the tritium being discharged in a gaseous effluent. The tritiated water in the solution of dissolved fuel is ultimately discharged to the environment in liquid effluents.

4.2.2 Tritium Production and Behaviour in the AGR Fuel Cycle

The production and behaviour of tritium in the AGR fuel cycle are very similar to those in the Magnox fuel cycle, and most of the information in the previous section is equally applicable to the AGR fuel cycle. The only significant difference is that a higher proportion of the tritium produced in AGR fuel is released into the carbon dioxide reactor coolant, and only a small fraction is retained in the stainless steel fuel cladding.

4.2.3 Tritium Production and Behaviour in the PWR Fuel Cycle

The main source of tritium in a PWR is ternary fission in the fuel. Some of the tritium produced is retained in the fuel, and the remainder becomes trapped in the zircaloy fuel cladding; very little tritium is released into the water coolant.

Some relatively small amounts of tritium is produced from activation of boron added to the water coolant of PWR's to control the reactivity of the reactors. This boron is ultimately discharged to the environment as tritiated water in liquid effluents.

In the reprocessing of PWR fuel, the tritium in the fuel cladding is effectively retained therein; the cladding waste is conditioned for storage as ILW. The great majority of the tritium in the fuel appears as tritiated water in the solution produced in the fuel dissolution process, with only a small fraction of the tritium being discharged in a gaseous effluent. The tritiated water in the solution of dissolved fuel is ultimately discharged to the environment in liquid effluents.

4.2.4 Tritium Production and Behaviour in other UK Reactor Fuel Cycles

A variety of other different types of reactor have been operated in the UK for research, development and defence purposes. Tritium production and behaviour in some of these reactors is significant.

Relatively large amounts of tritium are produced in reactors that are cooled or moderated with heavy water, due to thermal neutron capture by deuterium atoms in the heavy water. Most of this tritium accumulates in the coolant or moderator over the operational reactor lifetime. Notable examples of heavy water reactors operated in the UK are the Steam Generating Heavy Water Reactor (SGHWR) at Winfrith, and the Materials Test Reactors (MTR's) at Dounreay and Harwell.

4.2.5 Deliberate UK Tritium Production and Processing

Tritium has been deliberately produced in the UK by the irradiation of lithium targets for many years, most recently in the Chapelcross Magnox reactors. Historically, tritium was also produced by the irradiation of targets in the Windscale piles, the BEPO reactor and the MTR's at Harwell. A processing plant was until recently operated at the Chapelcross site to recover the tritium from irradiated targets, prior to its transfer to Aldermaston. Significant tritium processing facilities have been operated at the Aldermaston site for many years.

The Joint European Torus (JET) is an experimental fusion device operated at the Culham site, which is fuelled by a mixture of deuterium and tritium.

The GE Healthcare Maynard Centre near Cardiff produces tritiated organic compounds for use in medical and pharmaceutical research.

4.3   Quantities of Tritium Requiring Disposal in the UK

Information derived from the 2004 UK Radioactive Waste Inventory [11] shows that the total undecayed tritium inventory in ILW requiring deep geological disposal is 7.59×10^5 TBq. Table 2 provides detailed information on the ten waste streams that contain the highest undecayed tritium inventories (these account for over 98% of the total undecayed tritium inventory in ILW requiring deep geological disposal), while Table 3 provides details on the ten waste streams that contain the highest undecayed conditioned waste tritium concentrations in the 2004 UK Radioactive Waste Inventory.

The information in Tables 2 and 3 illustrates the diversity in the origin, and chemical and physical characteristics, of the major tritium-bearing waste streams in the UK inventory of ILW requiring deep geological disposal. The information also shows that a high proportion of the total undecayed tritium inventory in ILW requiring deep geological disposal will be concentrated in a relatively small number of waste packages, the majority of which will come within the category of Unshielded ILW (UILW)[6].

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[6] 'Shielded' ILW (SILW) is so called because it is to be packaged in Nirex shielded waste packages (2 metre and 4 metre Boxes) as distinct from 'Unshielded' ILW (UILW) which is to be packaged in Nirex unshielded waste packages (500-litre Drums, 3 cubic metre Boxes and Drums).

Table 2   ILW waste streams with highest total tritium inventories

Waste Stream Identifier	Waste Stream title and waste description	Waste category	Anticipated Packaging Approach		Undecayed tritium inventory (TBq)	Proportion of total undecayed ILW tritium inventory (%)
			Waste Package Type	Total Number of Waste Packages		
7A31	Operational ILW tritiated (water) -Tritiated water absorbed on zeolite.	UILW	500 litre Drum	17	6.12×10^5	80.8
5H301	JET decommissioning non-activated - ILW Wastes (mostly steels) from dismantling plant and process equipment in experimental fusion device.	SILW	2 metre Box	15	9.7×10^4	12.8
2F04/C	Encapsulated LWR cladding - Zircaloy cladding hulls from fuel reprocessing.	UILW	500 litre Drum	3700	1.1×10^4	1.5
2D24	Magnox cladding and misc. solid waste - swarf etc. from fuel reprocessing.	UILW	3 cubic metre Drum	500	6.0×10^3	0.8
2F03/C	Encapsulated AGR cladding - Stainless steel cladding from fuel reprocessing.	UILW	500 litre Drum	3900	5.0×10^3	0.7
2C06	Ceramic pellets - Scrap lithium aluminate targets used for tritium production.	UILW	500 litre Drum	20	3.9×10^3	0.5
2D09	Magnox cladding and miscellaneous solid waste - swarf etc. from fuel reprocessing.	UILW	3 cubic metre Box	1100	3.7×10^3	0.5
2D22	Magnox cladding and miscellaneous solid waste - swarf etc. from fuel reprocessing.	UILW	3 cubic metre Box	400	2.6×10^3	0.4
1B04	ILW containing tritium - Tritium contaminated trash and tritiated water from the production of tritiated organic compounds for medical and pharmaceutical research.	UILW	500 litre Drum	400	2.5×10^3	0.3
5B302	Prototype fast reactor ILW Wastes - (mostly steels) from dismantling fast breeder reactor.	UILW	3 cubic metre Box	600	1.7×10^3	0.2

Table 3   ILW waste streams with highest tritium concentrations

Waste stream identifier	Waste Stream title and waste description	Waste category	Anticipated Waste Package Type	Undecayed conditioned waste tritium concentration (TBq/m^3)	Average waste package inventory (TBq)
7A31	Operational ILW tritiated (water) - Tritiated water absorbed on zeolite.	UILW	500 litre Drum	72,000	36,000
2C15	Rotary pump oil - Tritiated oil absorbed on vermiculite.	UILW	500 litre Drum	836[7]	800
5H301	JET decommissioning non-activated ILW - Wastes (mostly steels) from dismantling plant and process equipment in experimental fusion device.	SILW	2 metre Box	701	6,500
5B356	PFR Absorbers - Scrap neutron absorber rods containing boron carbide.	UILW	500 litre Drum	467	250
2C06	Ceramic pellets - Scrap lithium aluminate targets used for tritium production.	UILW	500 litre Drum	400	200
7J27	Intermediate Level Tritium Waste - Scrap tritium luminising equipment.	UILW	500 litre Drum	30	15
2D37	Spent Cartridges - Scrap lithium/magnesium alloy targets used for tritium production.	UILW	500 litre Drum	29	15
5G04	Miscellaneous ILW - Miscellaneous cell-line wastes contaminated with tritiated water.	UILW	500 litre Drum	18	9
1B04	ILW containing tritium - Tritium contaminated trash and tritiated water from the production of tritiated organic compounds for medical and pharmaceutical research.	UILW	500 litre Drum	14	6
6C36	NDS Solid Tritiated ILW - Scrap lithium targets used for tritium production, swabs contaminated with tritiated heavy water, and miscellaneous tritium-bearing wastes (sealed sources, luminising equipment *etc.*).	UILW	500 litre Drum	13	6

[7] This is a very small volume waste stream (i.e. <0.5m^3), hence the apparent conflict between these two values

5   DERIVATION OF GUIDANCE LEVELS FOR TRITIUM WASTE PACKAGE INVENTORIES

The ensuing Sections use data from the PGRC safety assessments and relevant legislation to provide numerical guidance on the waste package inventories for tritium-bearing wastes above which the waste packager will be required to show to Nirex that specific consideration has been given to the ability of the waste package to provide retention of tritium.

5.1   Transport Safety

Two types of transport package exist within the PGRC. Nirex unshielded waste packages (i.e. 500 litre Drum, 3 cubic metre Box and 3 cubic metre Drum) will be transported through the public domain in reusable shielded transport containers[8], and this combination is categorised as a Type B transport package in the IAEA Transport Regulations [6]. Nirex shielded waste packages (i.e. 2 metre Box and 4 metre Boxes) 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 $10^{-6}A_2$ per hour on releases of activity from Type B transport packages under normal conditions of transport (NCT). During transport, tritium could escape from waste packages in the gaseous form and accumulate in the cavity of the sealed transport container. The rate of activity release from the transport container will then depend on the rate of leakage of tritium through the lid seal. Using assumed rates of leakage of tritium through the transport container lid seal it is possible to calculate the maximum permissible concentration of tritium within the transport container cavity during transport (these calculations assume a build-up of pressure within the cavity over the maximum 28 day period of transport and assume that the release limit of $10^{-6}A_2$ per hour is reached at the end of that period). Knowledge of the cavity volume (which will be different for the various waste packages due to their different displacement volumes) allows an allowable cavity tritium inventory to be calculated. This methodology is described in full in the GWPS [2].

Relating this allowable cavity inventory limit to waste package inventory limits is problematic. To do this rigorously, it would be necessary to have information on the fractional release of tritium during the period of transport, and such release properties are wasteform-dependent. Adopting a pessimistic approach, it can be assumed that the entire tritium inventory of the waste package(s) within the transport container is released into the transport container cavity during transport. In this case, the limit on the waste package tritium inventory will equal the maximum allowable transport container cavity inventory. Using this approach, it is possible to arrive at the waste package tritium inventory Guidance Levels for unshielded waste packages, as set by transport constraints, listed in Table 4. It should be noted that these values do not take account of contributory releases from other

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[8] The transport container assumed to be used for all Nirex standard unshielded waste packages is the Standard Waste Transport Container (SWTC).

radioactive gases. The presence and release of other radioactive gases would obviously lead to a reduction in the tritium inventory Guidance Levels.

The Transport Regulations also place limits on the release of activity under accident conditions of transport (ACT) and these can be similarly used to derive waste package Guidance Levels for tritium. These values are considerably higher than those derived from the NCT release limits, as discussed in the GWPS [2] and are not included in this guidance.

Table 4   Waste package tritium inventory Guidance Levels for unshielded waste packages, as set by transport constraints

Waste Package Type	Tritium Inventory Guidance Level (TBq)
500 litre Drum[9]	0.12
3 cubic metre Box[10]	0.13 – 0.23
3 cubic metre Drum	0.40

These Guidance Levels indicate waste package inventories for which the case (in this instance, the case for a Type B transport package) can readily be made. Inventories exceeding these Guidance Levels may also be acceptable but, as indicated in the opening paragraph of this Section, the waste packager will be required to give specific consideration to the means by which the waste package will retain tritium.

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).

Because IP-2 packages are designed to be transport packages in their own right, no credit can be taken for tritium retention by any overpack during transport. The design concepts for the 2 metre and 4 metre Boxes incorporate package vents to allow the escape of gases and prevent pressurisation of the containers. The rate of tritium release from IP-2 packages during transport will, therefore, depend on the tritium retention characteristics of the wasteform. The definition of *de minimis* Guidance Levels for the tritium inventory of IP-2 package is not, therefore, possible.

[9] The value given is for a single 500 litre Drum assumed to be contained within an SWTC with three other identical drums.

[10] A number of variants of the 3 cubic metre Box exist, the values stated correspond to the range of displacement volumes for these variants.

5.2 Repository Operational Safety

The release of tritium in the gaseous form from waste packages, once emplaced in a repository, will have dose consequences to workers on-site and the public off-site, both of which have been assessed in the GOSA [5].

For the Reference Case repository inventory, the GOSA estimates the peak off-site dose resulting from releases of gaseous radioactivity during the operational phase of a repository to be 2.1μSv/y, the dominant contributor to which is dose arising from tritium discharges. This is approximately a factor of ten lower than the off-site dose target of 20μSv/y specified in the Nirex Radiological Protection Policy Manual (RPPM). It does not follow, however, that an almost ten-fold increase in the total tritium inventory of the repository could necessarily be accommodated without the off-site dose target being exceeded, since the chemical form of the tritium-bearing waste is significant.

The main contributor to the peak rate of release of gaseous tritium during the repository operational phase has been predicted to be the production of tritiated hydrogen from Magnox corrosion. This is despite the fact that the total inventory of tritium in Magnox represents less than 0.03% of the total tritium inventory in the repository. It follows that an increase in the total inventory of tritium in Magnox could have a significant effect on the predicted peak off-site dose during the operational phase, despite the fact that such an increase could represent a very small change in the tritium inventory of the repository as a whole.

In summary, the GOSA has concluded that the Reference Case repository tritium inventory is acceptable in terms of its implications for off-site dose during the operational phase, but the acceptability of any increase in the Reference Case tritium inventory depends on the chemical form of the tritium-bearing waste. On that basis, waste package tritium inventory Guidance Levels can be defined that are consistent with the average concentration of tritium in the conditioned waste in the Reference Case repository inventory ($0.57 TBq/m^3$ for a total tritium inventory of approximately $9.6 \times 10^4 TBq$ at 2040 and a total conditioned waste volume of $1.68 \times 10^5 m^3$). These Guidance Levels, being the product of the average tritium concentration and the maximum payload volumes for the standard waste packages, are listed in Table 5.

Table 5   Waste Package Tritium Inventory Guidance Levels for Nirex Standard Waste Packages, as set by Operational Off-site Dose Constraints

Waste Package Type	Tritium Inventory Guidance Level (TBq)
500 litre Drum	0.3
3 cubic metre Box	1.8 – 1.9
3 cubic metre Drum	1.5
2 metre Box	5.6
4 metre Box	10.9

The GOSA assessment of on-site doses to repository workers concludes that there is confidence that the dose targets specified in the Nirex RPPM could be met. There is, therefore, no requirement to set more restrictive waste package tritium inventory Guidance Levels than those listed in Table 5 from considerations of on-site dose.

5.3 Repository Post-closure Safety

The post-closure safety issues that could arise from the presence of tritium in ILW are considered in general terms in the GPA [7] and more specifically in [12]. The time period required for establishment of the gas pathway has been assessed as being very long in relation to the half-life of tritium. Calculations, based on a generic geology and the 2001 National Inventory, have suggested that the timescale for the gas pathway to form was about 6,000 years. Although this value is very dependent on the assumed geological context etc., it is sufficiently large to indicate that, even in a much less suitable geology than that assumed, the radiological impact of tritium by inhalation will be insignificant.

Similarly, the GPA assessment of a groundwater travel time through the geosphere of approximately 30,000 to 300,000 years would indicate that the risk of the exposure to tritium as a result of this route is also insignificant.

As a consequence of the above, the GPA has concluded that the tritium inventory of a repository at closure will almost entirely decay within the engineered system and the geosphere, with very little being released to the accessible environment.

The GPA has also assessed tritium as being an insignificant contributor to the risks associated with the human intrusion pathway in part due to the relatively long periods of repository operation and institutional control, potentially extending to several centuries in total, assumed to precede any inadvertent human intrusion.

5.4 Application of Guidance Levels

Comparing the values in Tables 4 and 5 shows that, for UILW waste packages, the Guidance Levels set by transport constraints (Table 4) are more onerous than those set by considerations of repository operational safety. Comparison of these values with the information contained in the 2004 UK Radioactive Waste Inventory indicates that approximately 80% of the total conditioned volume of UILW requiring deep geological disposal will be contained in waste packages with tritium inventories less than the Guidance Levels listed in Table 4. Although it is possible to set Guidance Levels for SILW packages from consideration of repository operational safety, transport constraints are more onerous to the extent that it is not possible to define *de minimis* Guidance Levels for SILW packages.

It should be emphasised that the Guidance Levels set out for UILW packages in Table 4 do not necessarily represent firm limits on the tritium inventory of the packages, and that the inability to define a *de minimis* Guidance Level for SILW packages does not mean that these cannot be used to package wastes bearing any tritium. The derivation of Guidance Levels has been undertaken in a conservative manner, and takes no credit for any degree of tritium retention that could reasonably be expected from the use of conventional packaging methods. Accordingly, it may be permissible to package wastes with significantly higher tritium inventories without incorporating specific measures to maximise this retention. The Guidance Levels do, however, provide an indication of the tritium inventories above which the waste packager will be required to demonstrate to Nirex that specific consideration has been given to the potential for tritium retention within the waste package as-designed.

6   PACKAGING OF TRITIUM-BEARING WASTES

As discussed above, in assessing the adequacy of packaging arrangements for tritium-bearing wastes, particular attention needs to be paid to the rate of emission of gaseous tritium from waste packages during their transport to a repository. It is expected that UILW packages containing tritium inventories below the Guidance Levels set out in Table 4 will meet the relevant limits on gaseous tritium emissions at the time of transport and will not raise any other safety concerns for Nirex. UILW packages with a tritium inventory in excess of the Guidance Levels set out in Table 4 and all SILW packages containing tritium-bearing wastes will require specific assessment. In such cases, 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.

In addition to these Nirex-specific requirements, UK health and safety and environmental legislation places requirements on the operators of nuclear licensed sites to ensure that doses to workers are ALARP and that doses to members of the public and the population as a whole arising from discharges of radioactivity to the environment are As Low As Reasonably Achievable (ALARA). These requirements are relevant to doses arising from gaseous emissions of tritium from waste packages during all phases in their management (interim surface storage, transport to and storage in a repository *etc.*), and are applicable regardless of the tritium inventory of the waste packages or their gaseous tritium emission rates.

The remainder of this section provides advice on the packaging of tritium-bearing wastes in order to minimise rates of emission of gaseous tritium. In almost all cases, gaseous tritium emissions from waste packages will be dominated by two chemical forms: tritiated water and tritiated hydrogen. The advice is principally intended to assist waste packagers in ensuring that limits of emission of gaseous tritium at the time of transport of waste packages to a repository will be met, but will hopefully also be of use when considering the implications of gaseous tritium emissions from waste packages in their broader regulatory context.

6.1   Pre-processing of Tritium-bearing Wastes

Before making any choice on packaging methods, waste packagers should consider whether pre-processing of a tritium-bearing waste could be advantageous. Beneficial pre-processing could include sorting and segregating wastes in order to reduce the quantity requiring 'special' packaging arrangements, processing to reduce the tritium inventory of the waste (de-tritiation) and perhaps recovery of the tritium for reuse or recycle, and processing the waste to convert it into a form more amenable to packaging.

De-tritiation has been extensively investigated as a means of reducing the tritium inventory of wastes arising from fusion devices; processes involving chemical, physical and thermal de-tritiation have been researched. Incineration has often been suggested as a means of simplifying the packaging of organic tritium-bearing wastes such as lubricating oils although other issues, particularly the discharge of gaseous tritium following incineration, may preclude such an approach.

In cases where the tritiated waste is amenable to segregation from other, particularly long lived, wastes the potential exists for decay storage. Given a sufficient period of storage the activity of the tritium will decay to ultimately permit management as LLW.

6.2   General Considerations in Selection of Packaging Method

As discussed earlier, tritium-bearing wastes arise in the UK in a diverse range of chemical and physical forms. The concentrations, distributions and chemical forms of tritium in the wastes also vary greatly. Given these diversities, there is no single preferred approach to the packaging of tritium-bearing wastes; each case needs to be considered individually.

For any tritium-bearing waste, the issue of gaseous tritium emissions from waste packages is likely to be just one factor that influences the choice of packaging method. Other characteristics of the waste will also have an influence and where there are conflicting influences these will need to be balanced appropriately.

There is a substantial body of published research and development on the packaging of tritium-bearing wastes, much of it aimed at minimising the loss of tritium from the packages. In other countries, R&D work has been shaped by differing regulatory requirements on waste package storage, transport and disposal with respect to tritium loss and many other issues. As such, the details of much of the published work are not always directly applicable to the management of tritium-bearing wastes in the UK, but are certainly of general relevance.

Typically, a package containing tritium-bearing wastes contains a number of barriers to tritium loss: the waste itself, the waste conditioning matrix (if present), and the container. Environmental conditions during storage and transport of waste packages can also affect gaseous tritium emission rates.

6.3   The Waste as a Barrier to Tritium Emission

The waste itself may form the first barrier to tritium release from waste packages, either by accident or design. For example, zircaloy cladding from Light Water Reactor (LWR) fuel happens to contain tritium in the form of a stable chemical compound (zirconium hydride) present within the metallic matrix of the waste, while tritiated water is often deliberately immobilised by adsorption onto drying agents (*e.g.* molecular sieves). If a particular tritium-bearing waste stream has yet to arise, consideration should be given to how the waste producing process might be designed to optimise the tritium retention characteristics of the waste.

A detailed understanding of the chemical form and behaviour of the tritium within a conditioned waste, and at the interface between the waste and the surrounding conditioning matrix, is required in order to predict the identity and flux of tritiated species emitted from the waste. Obtaining such an understanding is often difficult, given the complexity of the processes involved, and it is worth noting that reality does not always accord with simple intuitive assumptions. For example, it has been shown that the vast majority of the tritium emitted into an atmosphere containing water vapour from samples of stainless steel containing dissolved tritiated hydrogen occurs in the form of tritiated water, possibly as a result of isotopic exchange reactions catalysed by the metal surface.

6.4 The Conditioning Matrix as a Barrier to Tritium Emission

Ideally, the conditioning matrix should complement, or at least not unduly undermine, the tritium retention characteristics of the waste itself. As well as influencing the choice of conditioning matrix, this consideration can raise the question of whether a waste should be conditioned at all. This question is discussed further in Section 6.4.1.

Where tritium is present in a mobile form, or where there are other sufficiently good reasons for choosing to do so, tritium-bearing wastes should be conditioned. By far the most commonly used and proposed materials for the conditioning of radioactive wastes in the UK are blended Portland cements, and the use of these for conditioning tritium-bearing wastes is discussed further in Section 6.4.2. The GWPS does not, however, rule out the use of conditioning matrices other than Portland cements, and the use of several alternative matrices is discussed in the remainder of Sections 6.4.2 to 6.4.5.

6.4.1 Unconditioned Tritium-bearing Wastes

Conditioning can provide a mechanism by which tritium can be liberated from wastes (*e.g.* heating caused by the curing of a conditioning matrix, isotopic exchange of tritium into the water content of a conditioning matrix, reactions between the waste and the conditioning matrix resulting in the physical or chemical degradation of the waste) and in some cases the benefits of not conditioning with respect to tritium retention could outweigh other disadvantages of such an approach. For example, at some Magnox reactor sites in the UK, consideration has been given to packaging tritiated reactor desiccant in an unconditioned form; conditioning the desiccant with a cement grout would liberate some of the tritium from the waste, resulting in environmental discharges during the waste packaging process and, potentially, undermining the ability of the waste packages to meet radioactive gas generation limits at the time of transport.

There are many advantages to the conditioning of wastes, potentially including the provision of a further barrier to tritium emission in the form of the conditioning matrix, and these benefits may outweigh the advantages of not conditioning with respect to tritium retention within the waste itself. There will also be instances where other characteristics of tritium-bearing wastes will dictate that they should be conditioned even if not conditioning would be beneficial with respect to tritium retention. An example is zircaloy hulls from LWR fuel reprocessing, where the presence of residual irradiated fuel associated with the hulls dictates that they should be conditioned, even though conditioning (in a cement grout) will enhance the rate of release of tritium from the waste.

In all cases, it is likely that the decision on whether or not to condition a tritium-bearing waste will require the balancing of several, often contradictory, influences in order to identify an optimum solution.

6.4.2 Blended Portland Cements as Conditioning Matrices for Tritium-bearing Wastes

In addition to being relatively cheap and readily available, Portland cements have many desirable properties as conditioning matrices. They have excellent mechanical, physical and thermal stability and are tolerant of a wide range of incorporated materials. The high pH buffering capacity of Portland cements allows them to contribute to the chemical containment of radioactivity in a repository, and they are compatible with the proposed repository backfill.

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Portland cements have received somewhat of a bad press in the published literature with respect to their performance in conditioning tritium-bearing wastes. Although this is partly justified, it is largely based on a specific measure of performance in conditioning a certain type of tritium-bearing waste.

Published work on the use of Portland cements for conditioning tritium-bearing wastes is dominated by studies on the conditioning of tritiated water, and by far the most commonly employed measure of the effectiveness of the immobilisation achieved has been the performance of the wasteforms produced in water-immersion leach tests. In this specific application, using that particular measure of performance, it is undeniable that Portland cements show poor tritium retention characteristics. However, as discussed above, this Guidance Note is concerned with the conditioning of a diverse range of tritium-bearing wastes, and gaseous tritium emissions from waste packages under conditions of storage and transport are of greater potential concern to Nirex than losses from packages in the post-closure phase of a repository. In this broader context, Portland cements can be seen to have very many good properties with respect to tritium retention and retardation.

Blended Portland cements can be very effective in retarding the emission of tritiated water vapour, which is expected to be a major contributor to gaseous tritium emissions for many waste packages. This is illustrated by reported observations that concrete structures contaminated with tritium (*e.g.* through contact with spilled tritiated water or airborne tritiated water vapour) outgas tritium very slowly and can retain appreciable tritium inventories for many years. There are several key features of blended Portland cements that contribute to this performance: they are able to chemically bind water within the hydrated cement matrix; although porous, water transport within the hydrated cement matrix is restricted; and the large inactive water content of the cement provides a high degree of isotopic dilution of tritiated water.

Any free tritiated water present at the time of waste conditioning, including any released from the waste on contact with the uncured conditioning matrix, will be incorporated into the cement as it hydrates. The tritiated water will be bound with varying degrees of firmness within the hydrated cement matrix, from water chemically combined into cement hydration products to free water in the capillary pores of the hydrated cement. As the chemical properties of tritiated water are essentially the same as those of inactive water, the proportions of tritiated water held in different forms in a hydrated cement matrix will be the same as those of the inactive water in the matrix. Any tritiated water released from a conditioned waste after the cement has cured will undergo isotopic exchange with all forms of inactive water in the cement.

There are three main routes for the emission of tritiated water vapour from blended Portland cement wasteforms: in evaporative water losses from exposed wasteform surfaces; in the water vapour content of gases generated within, and released from, the wasteform; and *via* isotopic exchange with water vapour in the atmosphere in contact with exposed wasteform surfaces.

The internal relative humidity (RH) of cured blended Portland cements is typically high, and will usually be higher than that in the atmosphere in which waste packages are stored and transported. Such a difference in RH provides a driving force for the loss of water from the wasteform *via* evaporation from wasteform surfaces in contact with the external environment. However, many of the features of typical waste packaging arrangements (the massive size of the wasteform, the use of an inactive capping grout, the casting of the wasteform within a steel container) will minimise the contact of the wasteform with the external environment, and it would be expected that evaporative water loss from most wasteforms would be extremely slow.

The emission of tritiated water in evaporative water losses from wasteforms can be minimised by reducing their internal RH (thereby reducing the driving force for evaporative water loss) and by reducing the mobility of water within the wasteforms (thereby restricting the rate of evaporative water loss). A low water-to-cement ratio in the cement formulation will tend to reduce both the internal RH of the wasteform (by reducing the amount of free water within the hydrated cement), and the rate of transport of water within the wasteform (by producing a finer and less continuous pore structure within the hydrated cement). The use of a low water-to-cement ratio also has the undesirable effect of reducing the degree of isotopic dilution of tritiated water, which would tend to increase the concentration of HTO in any evaporative water loss, but published information indicates that this is outweighed by the benefits of the measure in reducing the rate of evaporative water loss. The same effects of reduced internal RH and rate of water transport can also be achieved by the use of Portland cement blends containing Blast Furnace Slag (BFS) and, especially, Condensed Silica Fume (CSF).

Any internally generated gas will pick up water vapour in its passage through and out of a wasteform. Since the internal RH of blended Portland cement wasteforms is typically high, water loss *via* this route could be significant if the rate of internal gas generation is high. Also, since any approach to moisture equilibrium between a wasteform isolated within a package and the external environment is likely to be extremely slow, water loss through internally generated gas could theoretically proceed until essentially all the evaporable water in the wasteform has been lost.

The emission of tritiated water *via* this route can be minimised by reducing the rate of internal gas generation. For most wasteforms, gas generation will be dominated by the processes of radiolysis and chemical reactions involving the waste. Internal gas generation rates can, therefore, be minimised by a number of measures: reducing the inventory of other radionuclides in packages of tritium-bearing wastes; not storing packages of tritium-bearing wastes in proximity to other high dose rate waste packages; reducing the quantity of 'reactive' materials (*e.g.* aluminium) within packages of tritium-bearing wastes; and controlling the conditions within the wasteform in order to reduce the rate of reaction of any 'reactive' materials. The latter measure can have an influence on the blended Portland cement formulation; for example, the use of cement with a high proportion of BFS and a low water-cement-ratio can reduce the internal pH and water availability in the wasteform, thereby minimising the rate of reaction of metals such as aluminium. The use of a low water-to-cement ratio can also help to reduce water losses *via* internally generated gas by reducing the internal wasteform RH. Care must be taken, however, to ensure that the wasteform remains sufficiently porous to allow the internally generated gas to dissipate without causing any undue cracking of the wasteform.

Even if there is no net loss of water from a wasteform, tritiated water emission can still occur *via* isotopic exchange of tritiated water with atmospheric water vapour at exposed surfaces of the wasteform. The typical packaging approach of casting wasteforms within steel containers will minimise the surface area of the wasteform exposed to the external atmosphere. The use of a layer of inactive capping grout will also initially isolate the wasteform from the external atmosphere, although this effect will be temporary; over time, tritiated water will diffuse out of the wasteform and into the capping grout. The rate of tritiated water emission from wasteforms *via* isotopic exchange can be minimised by reducing the rate of water transport within the wasteform. As discussed above, this can be achieved by the use of blended Portland cements with a low water-to-cement ratio and containing BFS and CSF.

The use of organic polymers for coating the surfaces of the wasteform, and for infiltrating the porosity of the wasteform, has been studied as a means of reducing emissions of tritiated water. Significant short-term improvements in tritium retention have been reported in some cases but overall results are contradictory, perhaps due to problems with ensuring the production and long-term integrity of defect-free polymer coatings or impregnations.

Blended Portland cement wasteforms would be expected to offer essentially no retention or retardation of any internally generated tritiated hydrogen. At low tritium concentrations, the rate of radiolytically induced reactions of tritiated hydrogen with oxygen and airborne water vapour to produce tritiated water are very slow, and would not be expected to occur to any significant extent in the time taken for the tritiated hydrogen to diffuse through and out of the wasteform.

The dominant processes contributing to hydrogen generation in most packages are radiolysis and the chemical reaction of 'reactive' materials. Many of the measures discussed above that would minimise the rate of generation of bulk gases within the wasteform *via* these processes would also be effective in minimising the production of tritiated hydrogen.

6.4.3 Organic Polymers as Conditioning Matrices for Tritium-bearing Wastes

To date, organic polymers have not been widely employed in the conditioning of radioactive wastes in the UK[11]. Increasingly, however, in response to requirements to package wastes that do not lend themselves to effective conditioning with blended Portland cements, waste producers are considering the use of organic polymers. In general, the performance and properties of blended Portland cements in radioactive waste conditioning applications are better understood than those of organic polymers, and there is likely to remain, therefore, a preference for the use of inorganic cements wherever they can be shown to be practicable and adequately effective. Organic polymers do, however, have the potential to fill niches where inorganic cements are impractical or ineffective.

The use of organic polymers for conditioning tritium-bearing wastes has been studied to a limited extent. The consensus has tended to be that organic polymers offer little improvement over Portland cements in terms of tritium retention, although this conclusion has been based on the results of tests involving a limited variety of tritium-bearing wastes in water-immersion leach tests. When considered in a broader context, organic polymers could offer some advantages.

Tritiated water is not bound chemically in organic polymers but it can be physically immobilised to some degree. Although organic polymers are non-porous matrices, many still have appreciable permeability to tritiated water.

The principal routes for the emission of tritiated water vapour from organic polymer wasteforms are the same as those for blended Portland cements: in evaporative water losses from exposed wasteform surfaces; in the water vapour content of gases generated

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[11] Nirex has produced *Guidance Note on the Use of Organic Polymers for the Encapsulation of Intermediate Level Waste, WPS/901* to provide waste packagers with information on the types and range of applicable of such encapsulants.

within, and released from, the wasteform; and *via* isotopic exchange with water vapour in the atmosphere in contact with exposed wasteform surfaces.

Organic polymers themselves contain no inactive water, so that in the absence of any appreciable amount of inactive water in the waste itself, there is no great driving force for evaporative water loss from organic polymer wasteforms. The rate of any evaporative water loss can be minimised by selecting an organic polymer with low water permeability. Diffusion coefficients for water in organic polymers can be several orders of magnitude lower than in blended Portland cements.

Internal gas generation could be an important route for the loss of tritiated water from organic polymer wasteforms. Radiolytic degradation of the polymer itself is a potential source of internal gas generation, which could be minimised by the choice of polymer and by reducing the inventory of radionuclides in the wasteform. One of the attractive features of organic polymers is their minimal chemical interaction with wastes, which would be beneficial in minimising internal gas generation rates in wasteforms containing materials that are 'reactive' with blended Portland cements.

Isotopic exchange of tritiated water with atmospheric water vapour will occur at exposed surfaces of organic polymer wasteforms. The rate of emission of tritiated water *via* this route can be minimised by selecting a polymer with low water permeability.

6.4.4 Alternative Cements as Conditioning Matrices for Tritium-bearing Wastes

Recent years have seen an increasing interest in the potential use of alternative inorganic cements for conditioning certain wastes – chiefly those for which conditioning with blended Portland cements is problematic. A number of alternative cements are being actively researched, *e.g.* calcium aluminate cements (or high alumina cements), calcium sulfoaluminate cements, phosphate ceramics and inorganic geopolymers.

Of these examples, only calcium aluminate cements (or high alumina cements) have been investigated to any extent for conditioning tritium-bearing wastes, mainly tritiated water. The principal potential advantages of calcium aluminate cements in conditioning tritiated water have been perceived to be their higher water loading and shorter curing time relative to Portland cements. Published studies suggest, however, that calcium aluminate cements offer no significant advantages over Portland cements with respect to long-term tritium retention. The relatively low internal pH in cured calcium aluminate cements, compared with Portland cements, could reduce rates of bulk gas generation in wasteforms containing reactive metals such as aluminium, which could help to minimise the rate of emission of tritiated water vapour from such wasteforms. A significant concern with the use of calcium aluminate cements for conditioning tritium-bearing wastes could be the process known as 'conversion'. The cement hydrates formed after the curing of calcium aluminate cements are unstable at temperatures in excess of 20°C; the higher the temperature, the faster their rate of conversion. During the conversion reactions, free water is released and there is a substantial increase in the porosity and permeability of the cement, which would be a disadvantage with respect to tritium retention.

In the absence of any published information on which to base firm conclusions, it is only possible to speculate on the merits of some other alternative inorganic cements for conditioning tritium-bearing wastes. Calcium sulfoaluminate cements offer some of the same potential advantages as calcium aluminate cements: short curing times and a low internal pH. In addition, research has suggested that water availability in calcium sulfoaluminate cements may be significantly lower than Portland cement systems, which

could have advantages with respect to minimising tritium emission rates. Phosphate ceramics are often reported to have extremely low porosity, which could help to reduce tritium emissions. Inorganic geopolymers have a relatively low internal pH, which could help to reduce bulk gas generation and, therefore, tritium emission rates.

6.4.5 'Special' Conditioning Matrices

In some instances, perhaps when a waste contains tritium in a particularly high concentration and radiochemical purity, there may be merit in considering 'special' conditioning matrices. For example, consideration has been given to immobilising high concentration, high purity tritiated water at Aldermaston using molecular sieve. The rate of emission of gaseous tritium species from molecular sieve containing absorbed tritiated water have been shown to be very low.

Another example might be high concentration, high purity tritiated hydrogen gas, which can be effectively immobilised as metal hydrides (*e.g.* titanium or zirconium).

6.5 The Waste Package as a Barrier to Tritium Emission

Retention of gaseous tritium within waste packages could be achieved by sealing them (*e.g.* welding on a lid with no vent), and such an approach to packaging has, for example, been suggested for decommissioning wastes from the JET at Culham. Sealed metal containers are certainly an effective means of retaining gaseous tritium, but pressurisation of sealed ILW packages as a result of internal bulk gas generation could be a cause for concern. In the case of the JET decommissioning wastes, the use of a 'getter' has been suggested as a means of trapping tritium and reducing the effective gas generation rate within the sealed container and prevent significant pressurisation.

Temporary sealing of shielded waste packages during transport, in order to ensure compliance with the relevant gaseous tritium release rates, could be an option if it could be demonstrated that no significant pressurisation of the waste packages would occur during that period, and provided that the subsequent tritium release on removing the seals was acceptable.

Minimisation of the size of the container vent, which is generally good practice, would restrict the access of atmospheric water vapour into the waste package, thereby helping to minimise the loss of tritiated water from the wasteform *via* isotopic exchange.

6.6 The Effect of Storage and Transport Conditions

The rates of many of the processes involved in the emission of gaseous tritium from wastes packages are increased at elevated temperature, *e.g.* tritiated water diffusion in wasteforms and gas generation from chemical reactions in wasteforms. Some advantages could, therefore, be gained by minimising the temperature to which waste packages containing tritium-bearing wastes are exposed during their storage and transport. Also, maintaining a low RH in the waste package storage environment could be beneficial in reducing tritiated water losses *via* isotopic exchange. It must be noted, however, that the control of the temperature and humidity of the waste package storage environment is an important consideration in minimising container corrosion rates, and this must be considered in identifying an optimum set of storage conditions.

7 PRESENTATION OF ARGUMENTS FOR PACKAGING PROPOSALS

Regulatory guidance [3] requires that the waste packager (the site licensee) produces an ILW Conditioning Proposal to detail proposals for the conditioning of ILW together with the associated justification in safety and environmental terms. Proposals which involve segregation of wastes and, for example, decay storage to allow management of tritiated wastes as LLW, should also be addressed by the Conditioning Proposal.

In the case of tritiated wastes that are proposed for long term management in accordance with the PGRC, information would need to be supplied on the wastes for assessment under the LoC process.

UILW packages with a tritium inventory in excess of the Guidance Levels set out in Table 4 and all SILW packages containing tritium-bearing wastes will require specific assessment by Nirex. In such cases, an explicit case may need to be made by the waste packager to justify the proposed packaging method and demonstrate how it will ensure compliance with gaseous tritium emission rates at the time of waste package transport to a repository. In making such a case, a waste producer will need to present arguments supporting the effectiveness of the barriers to tritium emission in the proposed waste package.

There exists a substantial body of published information on the characteristics and behaviour of tritium in a variety of wastes and a limited number of conditioning matrices (mostly Portland cements). While it is unlikely to be able to use this information alone to construct definitive arguments for the adequacy of proposed packaging arrangements for tritium-bearing wastes, the information is likely to be useful in developing and supporting such arguments.

There are an appreciable number of published studies involving the experimental measurement of gaseous tritium emission rates from unconditioned wastes, and a few equivalent studies for conditioned wastes. This published information will, in some cases, be of use to waste producers in supporting arguments for the adequacy of packaging arrangements for tritium-bearing wastes. Waste producers should also consider undertaking their own experiments to measure gaseous tritium emission rates from wastes and wasteforms. The results of such studies could make an important contribution to an overall argument.

Mathematical modelling has been used to predict the behaviour of tritium in some wastes and conditioning matrices, particularly Portland cements. There are several examples of mathematical models being well validated by experimental observations, but these have been for relatively simple systems (*e.g.* tritiated water conditioned with Portland cement). Many real examples of wasteforms containing tritium-bearing wastes are likely to be more complex and, therefore, less amenable to modelling. In addition, uncertainty in the evolution of wasteform properties with age is likely to make long-term modelling of tritium behaviour difficult. In most cases, it is likely that relatively simple mathematical models could be of use in supporting arguments, rather than providing definitive evidence.

8   SUMMARY

Tritium is to be found at a wide range of concentrations in ILW streams in the UK, and several of the characteristics of the isotope make the management of tritium-bearing wastes a particular challenge: the propensity for tritium to be released from wastes and waste packages as a gas, its mobility and long residence time in the environment, the high isotopic exchange rate of tritium, and its ease of assimilation into living matter.

The chemical form of tritium in wastes varies, and influences the behaviour of tritium after wastes have been packaged. The mechanisms for tritium release from wastes and waste packages are complex and not easily understood or predicted. The relatively long half-life of tritium means that features of waste packages designed to reduce rates of tritium release need to maintain their performance for long periods of time. 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 tritium-bearing wastes.

In assessing the adequacy of packaging arrangements for tritium-bearing wastes, Nirex will be particularly concerned with the rate of emission of gaseous tritium from waste packages during their transport to a repository. It is expected that UILW packages containing tritium inventories below the Guidance Levels set out in Table 4 of this Guidance Note will meet the relevant limits on gaseous tritium emissions at the time of transport and will not raise any other safety concerns for Nirex. UILW packages with a tritium inventory in excess of the Guidance Levels set out in Table 4 and all SILW packages containing tritium-bearing wastes will require specific assessment by Nirex. In such cases, 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.

Typically, a package containing tritium-bearing wastes contains a number of barriers to tritium loss: the waste itself, the waste conditioning matrix (if present), and the container. Environmental conditions during storage and transport of waste packages can also affect gaseous tritium emission rates.

The waste itself may form the first barrier to tritium release from waste packages, either by accident or design. Ideally, the conditioning matrix should complement, or at least not unduly undermine, the tritium retention characteristics of the waste itself. As well as influencing the choice of conditioning matrix, this consideration can raise the question of whether a waste should be conditioned at all. Conditioning can provide a mechanism by which tritium can be liberated from wastes and in some cases the benefits of not conditioning with respect to tritium retention could outweigh other disadvantages of such an approach.

Portland cements have received somewhat of a bad press in the published literature with respect to their performance in conditioning tritium-bearing wastes. Although this is partly justified, it is largely based on a specific measure of performance in conditioning a certain type of tritium-bearing waste. When viewed in a broader context, Portland cements can be seen to have very many good properties with respect to tritium retention and retardation.

Blended Portland cements can be very effective in retarding the emission of tritiated water vapour, which is expected to be a major contributor to gaseous tritium emissions for many waste packages. There are several key features of blended Portland cements that contribute to this performance: they are able to chemically bind water within the hydrated cement matrix; although porous, water transport within the hydrated cement matrix is

restricted; and the large inactive water content of the cement provides a high degree of isotopic dilution of tritiated water. Blended Portland cement wasteforms would, however, be expected to be offer essentially no retention or retardation of any internally generated tritiated hydrogen.

The use of organic polymers for conditioning tritium-bearing wastes has been studied to a limited extent. The consensus has tended to be that organic polymers offer little improvement over Portland cements in terms of tritium retention, although this conclusion has been based on the results of tests involving a limited variety of tritium-bearing wastes in water-immersion leach tests. When considered in a broader context, organic polymers could offer some advantages.

Recent years have seen an increasing interest in the potential use of alternative inorganic cements for conditioning certain wastes – chiefly those for which conditioning with blended Portland cements is problematic. Of these alternative cements, only calcium aluminate cements (or high alumina cements) have been investigated to any extent for conditioning tritium-bearing wastes, mainly tritiated water. The principal potential advantages of calcium aluminate cements in conditioning tritiated water have been perceived to be their higher water loading and shorter curing time relative to Portland cements. Published studies suggest, however, that calcium aluminate cements offer no significant advantages over Portland cements with respect to long-term tritium retention.

Retention of gaseous tritium within waste packages could be achieved by sealing them. Sealed metal containers are certainly an effective means of retaining gaseous tritium, but pressurisation of sealed ILW packages as a result of internal bulk gas generation would be a cause for concern. Temporary sealing of waste packages during transport, in order to ensure compliance with the relevant gaseous tritium release rates, could be an option if it could be demonstrated that no significant pressurisation of the waste packages would occur during that period, and provided that the subsequent tritium release on removing the seals was acceptable.

Minimisation of the size of the container vent, which is generally good practice, would restrict the access of atmospheric water vapour into the waste package, thereby helping to minimise the loss of tritiated water from the wasteform *via* isotopic exchange.

Unshielded waste packages with a tritium inventory in excess of the Guidance Levels set out in Table 4 and all shielded waste packages containing tritium-bearing wastes will require specific assessment by Nirex. In such cases, an explicit case may need to be made by the waste packager to justify the proposed packaging method and demonstrate how it will ensure compliance with gaseous tritium emission rates at the time of waste package transport to a repository. In making such a case, a waste producer will need to present arguments supporting the effectiveness of the barriers to tritium emission in the proposed waste package. Such arguments are likely to be based on relevant published information, mathematical modelling, and experimental measurements of tritium emission rates.

9   GLOSSARY

A_2	A_2 is a measure of activity linked to possible exposure pathways defined in the IAEA Transport Regulations [6]. It is used in those Regulations to set contents limits for transport packages and to limit the maximum allowable activity releases under normal and accident transport conditions.
advective migration	Non-segregative flow of a gas through a permeable medium, driven by a pressure gradient. Commonly described by the Darcy equation. Tritium may be advected in a stream of bulk gases or due to the pressure of tritium alone.
barrier	Any component of a waste package that prevents or hinders the migration of tritium from the package.
bulk gases	Inactive gases generated within wastes due to chemical processes, such as corrosion, and radiolysis. Commonly dominated by 'ordinary' hydrogen.
conditioning	The treatment of radioactive waste to create a wasteform that has passive safety.
containerisation	The packaging of tritium-generating waste into an engineered gas-tight container designed to prevent the release of tritium.
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.
Immobilisation	Radioactivity present in waste is generally immobilised by converting the waste to a solid form that confers passive safety. This reduces the potential for migration or dispersion of the radioactivity by natural processes during storage, transport, handling and potential disposal.
organically bound tritium	Tritium which has substituted for ordinary hydrogen in the molecular structure of an organic material (i.e. plastics, rubbers, oils etc)
packaging	The preparation of radioactive waste for safe storage, transportation, handling and potential disposal by means of enclosing a conditioned wasteform in a suitable container.

passive safety	A passively safe wasteform is one in which the waste is chemically and physically stable, and is stored in containment and a manner that minimises the need for safety mechanisms, maintenance, monitoring and human intervention, and that facilitates retrieval for final disposal.
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.
(rate of) tritium release	The net rate at which tritium is released to the external environment from an engineered system such as a waste package. The product of the physical/chemical form of the tritium and the mitigation offered by the packaging, allowing for any retention in the waste itself.
shielded waste package	A shielded waste package is one that either has in-built shielding or contains low activity materials, and thus may be handled by conventional techniques. In most cases, shielded waste packages are also designed to qualify as transport packages in their own right.
transport container	In the context of this Glossary: a reusable container into which waste packages are placed for transport, the whole then qualifying as a Transport Package under the IAEA Transport Regulations.
transport package	As defined in the IAEA Transport Regulations: the complete assembly of the radioactive material and its outer packaging, as presented for transport.
unshielded waste package	An unshielded waste package is one that, owing either to radiation levels or containment requirements, requires remote handling and must be transported in a reusable transport container.
waste container	The vessel into which the wasteform is placed during manufacture of the waste package. In the PGRC the waste container remains in direct contact with the wasteform, and is subsequently managed as part of the complete waste package.
wasteform	The waste in the physical and chemical form in which it will be disposed of, including any conditioning media and container furniture (i.e. in-drum mixing devices, dewatering tubes etc) but not including the waste container itself or any added inactive capping material.
waste package	The wasteform and its waste container, as manufactured and prepared for all future aspects of waste management.

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