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Development of nuclear criticality safety controls on intermediate-level waste packages

T. W. Hicks[1], P. Wood[2], D. Putley[3] and T. D. Baldwin[1]

[1] Galson Sciences Limited, 5 Grosvenor House, Melton Road, Oakham, Rutland LE15 6AX, UK
[2] NDA Radioactive Waste Management Directorate, Building 587, Curie Avenue, Harwell, Didcot, Oxfordshire OX11 ORH, UK
[3] Serco Technical Consulting Services, Thomson House, Warrington, Cheshire WA3 6GA, UK

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ABSTRACT

Intermediate-level wastes (ILW) include substantial quantities of fissile material and controls are required to ensure that its storage, transport and disposal does not present a nuclear criticality hazard. This paper describes the Radioactive Waste Management Directorate's research to develop package fissile material limits (in the form of screening levels) for four different categories of ILW, defined according to uranium or plutonium composition: (1) irradiated natural and slightly enriched uranium (uranium containing up to 1.9 wt.% ^{235}U); (2) low-enriched uranium (uranium containing up to 4 wt.% ^{235}U); (3) high-enriched uranium (uranium containing up to 100 wt.% ^{235}U); and (4) separated plutonium (plutonium containing up to 100 wt.% ^{239}Pu).

The derivation of package screening levels was supported by neutron transport calculations that addressed conditions during waste package transport to a geological disposal facility (GDF), during the GDF operational phase and after GDF closure. The analysis included consideration of combinations of events and processes that could result in fissile material accumulation and concentration after GDF closure, when waste packages have deteriorated sufficiently for fissile material to be mobilized. The results of the calculations have provided input to Radioactive Waste Management Directorate's decision making on setting waste package screening levels.

Keywords: ILW packages, criticality hazard.

Introduction

The Radioactive Waste Management Directorate (RWMD) of the Nuclear Decommissioning Authority (NDA) is responsible for implementing UK Government policy for long-term management of higher activity radioactive wastes. Government policy for geological disposal of radioactive waste, preceded by safe and secure interim storage, is set out in the *Managing Radioactive Waste Safely* (MRWS) White Paper (Department for Environment Fisheries and Rural Affairs *et al*., 2008), along with details of the radioactive waste inventory (the Baseline Inventory) that requires disposal.

The greatest volume of waste in the Baseline Inventory is intermediate-level waste (ILW). The ILW arises from the reprocessing of spent nuclear fuel and from the operation, maintenance and decommissioning of nuclear facilities, including power plants and reprocessing facilities. These wastes include substantial quantities of fissile material (mainly ^{239}Pu and ^{235}U) and the amounts of these materials placed in disposal packages must be controlled to ensure that criticality cannot occur during waste packaging, storage, transport and emplacement in a GDF. Further, to meet the environment agencies' *Guidance on the Requirements for Authorisation* (GRA) for geological disposal facilities (Environment

* E-mail: twh@galson-sciences.co.uk
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Agency and the Northern Ireland Environment Agency, 2009), it must be demonstrated that the possibility of criticality after disposal does not present a significant concern. This paper describes analysis undertaken to support the development of ILW package criticality controls, based on the criticality assessment methodology described by (Hicks *et al*., 2011). The RWMD's overall approach to assessing the criticality safety of the broad range of UK fissile wastes is described in the *Criticality Safety Status Report* (Nuclear Decommissioning Authority, 2010*a*).

Assessment approach

The RWMD has derived ILW package criticality controls in the form of waste package 'screening levels' that are applicable to different categories of waste (Hicks *et al*., 2011). The screening levels provide criteria against which judgments can be made on the suitability of a proposed waste package concept for disposal in a GDF. Derivation of the screening levels involves consideration of the conditions that a waste package may be subjected to during transport to a GDF, during the GDF operational phase and after GDF closure.

Waste package fissile material limits for the transport phase have been identified based on consideration of credible accident conditions during the transport of an ILW package to a GDF. The supporting analysis considered detailed waste package and transport overpack designs.

Assessment of GDF operational and post-closure phase criticality safety requires assumptions to be made about the disposal system design and geological environment. At the present stage in the process of implementing geological disposal in the UK, no disposal sites have been identified. Therefore, RWMD has developed a programme that considers disposal of higher activity wastes in different geological settings to provide a basis for planning and assessment. The derivation of ILW package criticality controls is based on a reference concept for the disposal of ILW in a facility located in a higher-strength fractured host rock. This paper focuses on the derivation of ILW package screening levels for the GDF operational and post-closure phases.

The GDF operational phase

In the concept for a higher strength fractured host rock, ILW packages are stacked in a series of disposal vaults. The waste packages are expected to remain intact during the operational phase, such that containment of the waste materials is maintained. The disposal facility would be kept dry but, in the event of a flooding accident, the packages could become immersed in water. Eventually, the disposal vaults would be backfilled with Nirex Reference Vault Backfill (NRVB), which is a cement-based grout. In the operational phase assessment, conditions under which the waste packages are surrounded by air, water or NRVB have been evaluated.

The GDF post-closure phase

Fissile material may be redistributed after GDF closure as hydrological and geochemical conditions in the GDF evolve, which is likely to lead to a reduction in the neutron multiplication factor, although it is possible that this factor could increase.

The expected evolution of GDF near-field conditions is described in the NDA's *Near-field Evolution Status Report* (Nuclear Decommissioning Authority, 2010*b*). After backfilling, the disposal area will start to resaturate, with full resaturation expected in a few decades to a few centuries. The groundwater would rapidly equilibrate with the cement-based backfill resulting in the development of alkaline conditions, and reducing conditions would soon be established as oxygen is consumed by corrosion reactions and microbial activity. Once the vaults have resaturated, small concentrations of uranium and plutonium could be released into the backfill through the vents in the containers. The rate of radionuclide release is likely to increase as the waste packages corrode and degrade.

The alkaline and reducing conditions would limit uranium and plutonium solubilities, and any dissolved species would sorb strongly to corrosion products, encapsulant material and backfill. However, organic complexing agents derived from the degradation of cellulosic materials in the wastes and colloidal materials may increase radionuclide mobility.

The relocation and accumulation of plutonium and uranium under evolving GDF conditions could lead to increases in the neutron multiplication factor (although it is more likely that reductions would occur). For the purpose of deriving package limits, post-closure criticality scenarios can be constructed by considering combinations of events

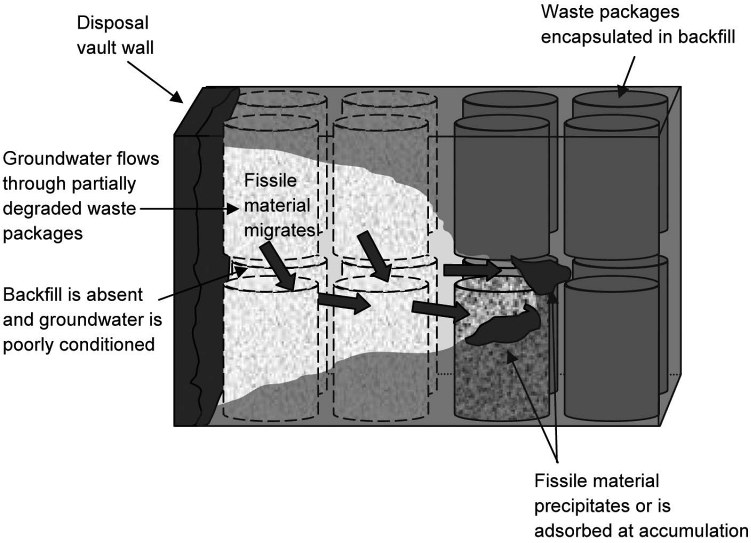

FIG. 1. Processes involved in the precipitation/sorption scenario for a section of a disposal vault containing ILW in 500 litre drums.

and processes that could lead to such accumulations. Hicks (2007*a*) identified the following scenarios:

(1) *Precipitation/adsorption* of dissolved fissile material following release and migration from several degrading waste packages (Fig. 1). Precipitation could occur following transfer from an oxidizing to a reducing environment or from a low-pH to a high-pH environment.

(2) *Filtration/settling* of fissile material in colloidal form (1 nm to 1 μm) or particulate form (greater than 1 μm) following migration to environments of smaller pore space. Gravitational settling of particulates could occur following a reduction in flow rate along a flow path.

(3) *Wasteform degradation/settling* in which particulate fissile material settles and accumulates in a degrading waste package.

(4) *Slumping* of solid fissile material through a stack of degrading waste packages (Fig. 2).

(5) *Collapse/compaction* of stacks of waste packages as a result of corrosion-related weakening, backfill loss and/or stress loading (natural events).

If a sufficiently large amount of fissile material accumulates and concentrates by these mechanisms a criticality could occur. The mass and concentration required for criticality depend on the fissile nuclides involved, the geometry of the accumulation region, and the amount, type and distribution of neutron moderators, absorbers and reflectors.

The criticality scenarios may be distinguished according to the location of the fissile material accumulation site and the extent of fissile material transport and concentration compared to the initial distribution, as follows:

(1) Package-scale scenarios in which fissile material concentrates or accumulates within or near to the source waste package.

(2) Stack-scale scenarios in which there is localized degradation and fissile material from several degraded waste packages concentrates or accumulates in the GDF.

(3) Vault-scale scenarios in which there is material degradation on a large scale and fissile material from many waste packages concentrates or accumulates.

In the evaluation of package-scale scenarios, such as the waste degradation/settling scenario, it was assumed that the overall mechanical structure of each waste package is maintained. A timescale of 15,000 years was judged to be pessimistic for evaluation of package-scale screening levels (Hicks, 2007*a*). For the stack-scale configuration the assessments focussed on the slumping scenario. Stack-scale screening levels were evaluated at 60,000 years (Hicks, 2007*a*).

No attempt was made to constrain the sources of fissile material contributing to the vault-scale scenario owing to the large uncertainties involved. Instead, in order to derive a measure of criticality safety margins, consideration was given to the

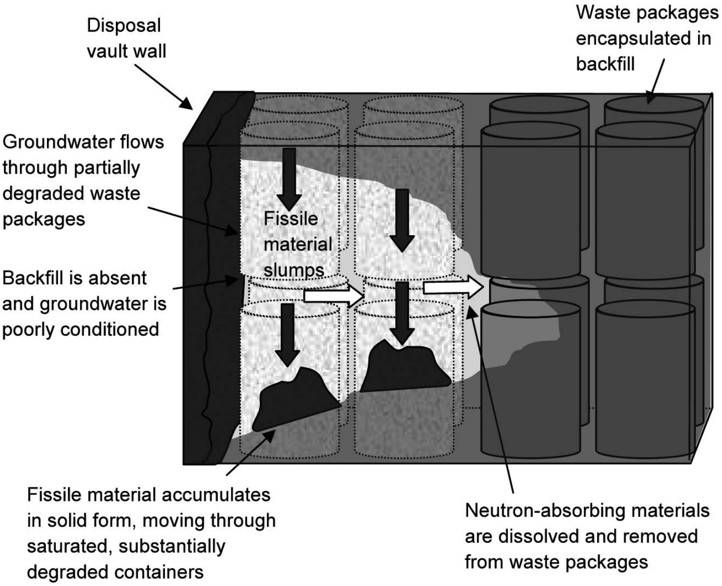

FIG. 2. Processes involved in the slumping scenario for a section of a disposal vault containing ILW in 500 litre drums.

extent to which fissile material must be concentrated compared to some initial average distribution in a vault to achieve a critical concentration.

Categories of intermediate level waste

Based on this approach to assessing GDF operational phase and post-closure conditions, ILW package screening levels were derived that are applicable to different categories of ILW. The RWMD produced a series of four generic *Criticality Safety Assessments* (CSAs) for common categories of fissile ILW (Hicks, 2007*b*): irradiated natural and slightly enriched uranium (NU) (Hicks, 2007*c*), low-enriched uranium (LEU) (Hicks, 2007*d*), highly enriched uranium (HEU) (Hicks, 2007*e*) and separated plutonium (Pu)[1] (Hicks, 2007*f*):

(1) The irradiated NU category refers to ILW that contains uranium containing up to 1.9 wt.% ^{235}U.

(2) The LEU category refers to ILW that contains uranium containing up to 4 wt.% ^{235}U.

(3) The HEU category refers to ILW that contains uranium containing up to 100 wt.% ^{235}U.

(4) The separated Pu category refers to ILW that contains plutonium containing up to 100 wt.% ^{239}Pu.

The irradiated NU and LEU categories were defined such that credit can be taken for the presence of the neutron absorber ^{238}U. The HEU and separated Pu categories are intended to be applicable to wastes characterized by high ^{235}U and ^{239}Pu enrichments, respectively. The four generic ILW categories encompass packaging concepts for most of the UK's ILW. Package-specific CSAs would be required for ILW packages that fall outside the scope of the generic CSAs.

For each generic CSA, two screening levels were calculated; a lower screening level (LSL) intended to represent a conservative view of possible criticality scenarios and an upper screening level (USL) that adopts a more credible, yet still conservative representation of scenarios, in which credit is taken for specific measurable characteristics of the wasteform. The four generic CSAs provide less restrictive screening levels than the general screening level (GSL) of 50 g ^{239}Pu (or the amount of another radionuclide that will produce an equivalent contribution to the neutron multiplication factor under similar condi-

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[1] The general high enriched uranium and separated plutonium ILW categories refer to wastes that are contaminated with these radionuclides. They do not refer to the UK's uranium and separated plutonium stocks that are not currently classified as wastes.

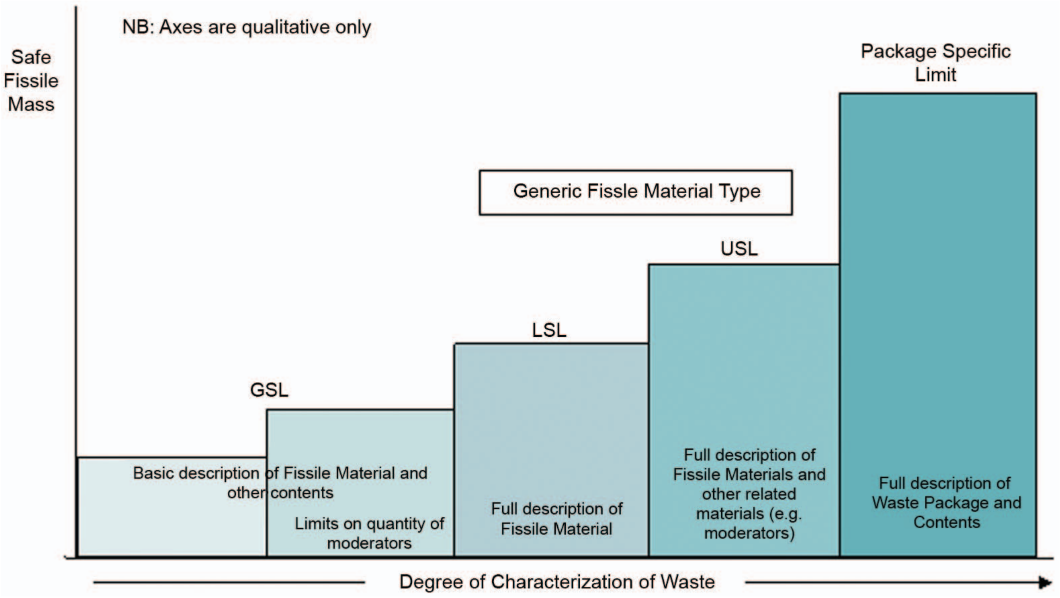

FIG. 3. Hierarchy of safe fissile masses for waste packages (Nuclear Decommissioning Authority, 2010*a*).

tions), which is supported by the *General Criticality Safety Assessment* (GCSA) (Hicks, 2009). The application of the hierarchy for setting safe fissile masses for waste packages is illustrated in Fig. 3 (Nuclear Decommissioning Authority, 2010*a*).

Modelling assumptions

A pessimistic approach was taken for the operational phase LSL calculations in order to account for the uncertainty associated with the distribution of materials in the waste packages. The approach involved selecting the material configurations that result in the greatest neutron multiplication factor. The fissile material in each waste package was assumed to be in the form of a near-quarter sphere at a top or bottom edge of the package, such that the aggregate material from each two-by-two vertical array of packages forms an approximate sphere (as indicated in Fig. 4 for an array of 500 litre drums). The moderator was

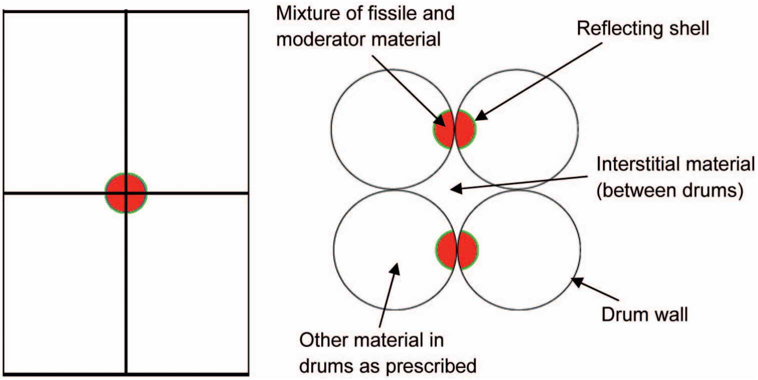

FIG. 4. Elevation view (left) and plan view (right) of fissile material mixture represented as near quarter-spheres in 500 litre drums for the operational phase LSL calculations. The drum array is repeated throughout the vault model.

TABLE 1. Selected screening levels for 500 litre drums derived in the generic CSAs.

CSA waste category	Assessment phase	Screening level	Waste packaging requirements	Fissile material limit (g)
Irradiated NU (up to 1.9 wt.% ^{235}U)				
	Transport	N/A	<1 kg beryllium <10 kg graphite	$\mathrm{U + Pu} \leqslant 5650$
	Operational	LSL	<0.1 kg beryllium <1 kg graphite	$\mathrm{U + Pu} \leqslant 34{,}000$
		LSL	<0.1 kg beryllium No limit on graphite	$\mathrm{U + Pu} \leqslant 33{,}000$
		USL	Well mixed wasteform	$\mathrm{U + Pu} \leqslant 155{,}000$
	Post-closure package-scale	LSL	<1 kg graphite	$^{239}\mathrm{Pu} \leqslant 150$
		LSL	No limit on graphite	$^{239}\mathrm{Pu} \leqslant 100$
		USL	Well mixed wasteform	$\mathrm{U+ Pu} \leqslant 200{,}000$
	Post-closure stack-scale	LSL	No graphite present	$^{239}\mathrm{Pu} \leqslant 325$
		LSL	No limit on graphite	$^{239}\mathrm{Pu} \leqslant 160$
		USL	Well mixed wasteform	$\mathrm{U + Pu} \leqslant 28{,}500$
LEU (up to 4 wt% ^{235}U)				
	Transport	N/A	<1 kg beryllium <10 kg graphite	$\mathrm{U + Pu} \leqslant 5650$
	Operational	LSL	<0.1 kg beryllium <1 kg graphite	$\mathrm{U + Pu} \leqslant 10{,}000$
		USL	Well mixed wasteform	$\mathrm{U + Pu} \leqslant 50{,}000$
	Post-closure package-scale	LSL	<1 kg graphite	$\mathrm{U + Pu} \leqslant 8100$
		USL	Well mixed wasteform	$\mathrm{U + Pu} \leqslant 65{,}000$
	Post-closure stack-scale	LSL	None specified	$\mathrm{U + Pu} \leqslant 4350$
		USL	Well mixed wasteform	$\mathrm{U + Pu} \leqslant 9300$
HEU				
	Transport	N/A	<0.1 kg beryllium <5 kg graphite	$^{235}\mathrm{U} + {}^{239}\mathrm{Pu} + {}^{241}\mathrm{Pu} \leqslant 105$
	Operational	LSL	<0.1 kg beryllium <1 kg graphite	$^{235}\mathrm{U} + 1.6 \times ({}^{239}\mathrm{Pu} + {}^{241}\mathrm{Pu}) \leqslant 160$
		USL	Well mixed wasteform	$^{235}\mathrm{U} + 1.6 \times ({}^{239}\mathrm{Pu} + {}^{241}\mathrm{Pu}) \leqslant 1400$
	Post-closure package-scale	LSL	<1 kg graphite	$^{235}\mathrm{U} + 1.39 \times {}^{239}\mathrm{Pu} \leqslant 150$
		USL	Well mixed wasteform	$^{235}\mathrm{U} + 1.39 \times {}^{239}\mathrm{Pu} \leqslant 1600$
	Post-closure stack-scale	LSL	None specified	$^{235}\mathrm{U} + 1.11 \times {}^{239}\mathrm{Pu} \leqslant 94$
		USL	Well mixed wasteform	$^{235}\mathrm{U} + 1.11 \times {}^{239}\mathrm{Pu} \leqslant 240$
Separated Pu				
	Transport	N/A	<0.1 kg beryllium <5 kg graphite	$^{235}\mathrm{U} + {}^{239}\mathrm{Pu} + {}^{241}\mathrm{Pu} \leqslant 105$
	Operational	LSL	<0.1 kg beryllium <1 kg graphite	$0.65 \times {}^{235}\mathrm{U} + {}^{239}\mathrm{Pu} + {}^{241}\mathrm{Pu} \leqslant 95$
		LSL	<0.1 kg beryllium <1 kg graphite >78 mm annulus	$0.65 \times {}^{235}\mathrm{U} + {}^{239}\mathrm{Pu} + {}^{241}\mathrm{Pu} \leqslant 240$
		USL	>80 kg steel >78 mm annulus	$0.65 \times {}^{235}\mathrm{U} + {}^{239}\mathrm{Pu} + {}^{241}\mathrm{Pu} \leqslant 2150$
	Post-closure package-scale	LSL	<1 kg graphite	$0.65 \times {}^{235}\mathrm{U} + 0.88 \times {}^{239}\mathrm{Pu} \leqslant 97.5$
		USL	Well mixed wasteform	$0.65 \times {}^{235}\mathrm{U} + 0.88 \times {}^{239}\mathrm{Pu} \leqslant 1000$
	Post-closure stack-scale	LSL	None specified	$^{235}\mathrm{U} + 1.11 \times {}^{239}\mathrm{Pu} \leqslant 94$
		USL	Well mixed wasteform	$^{235}\mathrm{U} + 1.11 \times {}^{239}\mathrm{Pu} \leqslant 240$

assumed to be water or polythene, depending on the composition of the wasteform in each ILW category. Wasteform materials such as grout, which might absorb neutrons, were ignored (modelled as air), but nominal quantities of beryllium (0.1 kg) and graphite (1 kg) were assumed to provide some neutron reflection. The cautiousness of this approach is supported by sensitivity calculations for similar configurations (e.g. with grout, water, polythene or steel in the drum) that were reported in the GCSA (Hicks, 2009). Optimum conditions for criticality were determined by varying the mass and concentration of U or Pu for different configurations.

For the USL analysis, some of the pessimistic assumptions about the wasteform properties were relaxed. The fissile material was assumed to be uniformly mixed with other materials in the container (cementitious grout or steel), rather than located in the waste package in an optimum geometry and concentration for criticality.

To evaluate the post-closure package-scale LSLs, the waste packages were assumed to be water saturated, but the fissile material geometry in each waste package was assumed to be consistent with that adopted for the operational phase LSL analysis (i.e. the near-quarter sphere arrangement). Graphite was assumed to remain as a neutron-reflecting shell around each fissile assembly.

For the USL analysis, it was assumed that the fissile material migrates and collects at the base of the waste package by settling. The fissile material was assumed to accumulate with water to form a thin slab (approximated by an infinite slab) at the base of the container. Water was assumed to provide full neutron reflection above and below the modelled slab.

For the stack-scale LSL, all of the fissile material from a single stack of seven waste packages was assumed to accumulate in the form of a spherical mass mixed with water on the vault floor. The presence of backfill materials, corrosion products and other waste materials was ignored. The fissile material accumulation was assumed to be surrounded by water or graphite in sufficient mass to provide maximum neutron reflection.

For the USL analysis, particulate fissile material was assumed to migrate and accumulate in a thin slab-shaped geometry (approximated by an infinite slab) at the vault floor. Materials such as backfill and corrosion products, which would have a negative effect on the neutron multiplication factor, were ignored. Water was assumed to provide neutron moderation and full neutron reflection above and below the modelled slab.

In most cases, the screening levels were evaluated using the *MONK* computer code, although in some cases, published data on critical masses for relevant configurations were adopted.

Screening levels

Table 1 summarizes selected screening levels from the four generic CSAs for ILW packaged in 500 litre drums. Screening levels were also calculated for ILW in 3 m^3 boxes and 3 m^3 drums. Note that the limits for packages containing irradiated NU or LEU are presented in terms of U + Pu, and include the large fractions of ^{238}U that would be present at the enrichments indicated.

The RWMD is currently considering limiting the analysis to derive waste package screening levels to assessment of conditions during waste package transport and GDF operations. That is, the assessment of post-closure criticality scenarios would be excluded from the evaluation of waste package screening levels. The assessment of post-closure criticality would then rely on analyses of the likelihood and consequences of criticality in a GDF.

The analysis of vault scale scenarios found that fissile material concentrations in the disposal vaults would need to increase by at least two orders of magnitude compared to initial conditions defined by the LSLs or by one order of magnitude compared to initial conditions defined by the USLs to achieve minimum concentrations for criticality based on pure water-moderated systems that lack neutron absorbing materials other than ^{238}U where appropriate.

Conclusions

Waste package screening levels have been evaluated for each of four categories of ILW (irradiated NU, LEU, HEU and separated Pu) based on consideration of conditions during waste package transport to a GDF, during GDF operations and following GDF closure. LSLs have been derived on the basis of conservative assumptions (in terms of effects on the neutron multiplication factor) about wasteform properties. The USLs have been derived for the GDF operational and post-closure phases by taking credit for specific measurable characteristics of the wasteform. These LSLs and USLs support judgments on the criticality safety of waste packaging proposals. Packaging up to a USL might be accepted provided it can be assured that all requirements regarding the nature and variability of the wasteform can be met and that overall risks across the breadth of waste management would not be as low as reasonably practicable (ALARP) if wastes were packaged to the LSL.

Acknowledgements

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