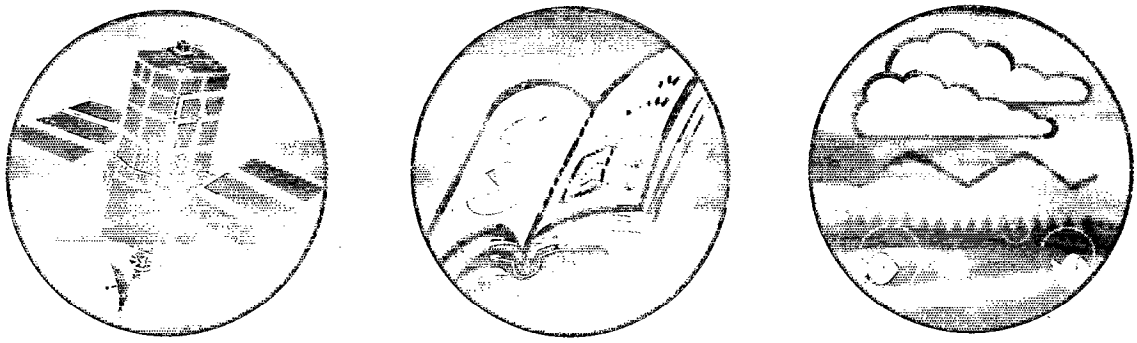
A Review of the Treatment of Criticality in Post-Closure Safety Assessment of Radioactive Waste Disposal



Research and Development

Technical Report
P222

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A Review of the Treatment of Criticality in Post-Closure Safety Assessment of Radioactive Waste Disposal

R&D Technical Report P222

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Publishing Organisation:
Environment Agency
Rio House
Waterside Drive
Aztec West
Almondsbury
Bristol BS32 4UD

Tel: 01454 624400 Fax: 01454 624409

ISBN: 1 85705 005 3

Dissemination status
Internal: Released to Regions
External: Released to the Public Domain

Statement of use
This report provides a review of approaches to making a post-closure safety case in relation to nuclear criticality. This information will enable the Environment Agency, in consultation with other regulatory organizations, to provide an effective input to the process of evaluating proposed fissile contents of waste packages. This information is also for use by the Environment Agency in determining its future research requirements in the area of post-closure criticality.

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FOREWORD

The review of assay techniques reported here was undertaken by one of the authors, Dr Tommy Green (AEA Technology, Winfrith), under subcontract to Galson Sciences Limited. Dr Larry Sanchez (Sandia National Laboratories, Albuquerque, US) also made a significant contribution to this project by providing information on the post-closure criticality analyses being undertaken for the proposed repository at Yucca Mountain, Nevada, US. In addition, discussions with Mr Roger Yearsley (Environment Agency project manager), Dr David Simister (Her Majesty's Nuclear Installations Inspectorate) and Dr David Bennett (Galson Sciences) proved invaluable, and review by Dr Daniel Galson (Galson Sciences) provided many improvements. The diagrams were produced by Miss Chris Taylor (Galson Sciences).

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EXECUTIVE SUMMARY

Any safety case for a radioactive waste disposal facility in England and Wales will include consideration of the potential for post-closure nuclear criticality if the waste includes fissile material. In order to fulfil its responsibility to assess such a safety case, the Environment Agency will need to be able to understand and assess the treatment of post-closure criticality. The project reported here forms the basis for the Environment Agency's development of a capability to analyse post-closure criticality issues. The report will also help the Environment Agency, in consultation with other regulatory organizations, to provide an effective input to the process of evaluating proposed fissile contents of waste packages.

The report presents a review of key post-closure criticality studies undertaken in radioactive waste disposal programmes around the world (including the Waste Isolation Pilot Plant and Yucca Mountain projects in the US, the Konrad repository in Germany, and proposed deep geologic repositories in the UK, Canada, and Sweden), and also examines the treatment of criticality safety in several waste handling and storage facilities. In general, these studies emphasise the need for post-closure criticality to be considered as part of a repository safety case, although most post-closure criticality studies suggest that criticality is a low-probability event with low consequences in terms of overall repository safety.

The review has led to the identification of five post-closure criticality scenarios that are typical of the scenarios studied in post-closure criticality assessments, although the specification of scenarios is not unique. The scenarios relate to the concentration of fissile radionuclides under different conditions at various locations in a disposal system:

- criticality in an intact waste container;
- criticality in a leaking waste container;
- criticality in one or more collapsed waste containers;
- criticality in the near-field; and
- criticality in the far-field.

Analysis of the criticality scenarios requires assumptions to be made in relation to the sequences and combinations of features, events and processes associated with each scenario. The scenarios relating to criticality in waste containers are, to some extent, independent of repository design, in that they do not require detailed information on repository location and layout, or rock properties. Thus, these scenarios may reasonably be included in post-closure criticality assessments in the UK in the absence of an intermediate-level waste (ILW) repository site and design. However, for the scenarios involving leaking or collapsed containers, some assumptions would need to be made concerning post-closure hydraulic and geochemical conditions in the vicinity of the waste containers. These scenarios are also relevant to waste storage facilities and the pre-closure period of a repository, particularly if long periods of operation are envisaged.

The near-field and far-field scenarios are relevant to the repository post-closure period, and may involve concentrations of fissile material from more than one waste container. The analyses undertaken for the Konrad repository in Germany showed that consideration of the near-field criticality scenario can lead to the specification of more restrictive limits on the fissile material content of waste containers than limits derived for criticality safety during waste handling and storage. Assessment of the near-field scenario requires some understanding of disposal room geometry, and the mechanisms and locations for potential radionuclide accumulation in critical geometries. Assessment of the far-field scenario requires an understanding of factors such as fracture properties and groundwater oxidation states, which could only be addressed as part of a site evaluation process. Without a specific ILW repository site or disposal concept, as at present in the UK, bounding assumptions, based on, for example, generic repository data, would need to be made concerning near-field and far-field conditions in order to evaluate these scenarios.

An initial deterministic evaluation of these five criticality scenarios, involving consideration of their credibility for a particular disposal concept and/or evaluation of their consequences, may show that post-closure criticality concerns are insignificant. If post-closure criticality concerns remain after such analysis, then it may be possible to address these concerns by reducing conservatism and/or uncertainty in models and/or parameter values. Alternatively, post-closure criticality concerns may be addressed by undertaking risk-based analysis. Such analysis would require determination of the fission yield from, and probability of, criticality excursions, and would require use of a suite of risk analysis codes and significant computational resources. Using such a risk-based approach, the consequences of criticality could, if necessary, be accounted for in a repository performance assessment (PA). However, operators should aim to ensure that radiological doses or risks are as low as reasonably achievable, and so it may be more desirable to include controls on waste packaging and repository design that minimized the possibility of a criticality excursion occurring, than to account for the potential consequences of post-closure criticality in a full performance assessment. The most direct criticality control is to limit the fissile mass in a waste container, although other controls may be imposed on the waste form, waste container properties, disposal room geometry, and backfill materials.

Waste producers can only demonstrate compliance with limits on the fissile content of waste packages to the extent allowed by assay techniques. The accuracy of the three non-destructive assay (NDA) methods used by the nuclear industry (gamma scanning, passive neutron coincidence counting, and neutron interrogation) is discussed in this report. The accuracy and precision of measurements using these devices should be taken into account when setting criticality limits. NDA measurements may be supported by destructive sampling and analysis of waste package contents, but destructive assay is not a viable alternative to NDA.

The Environment Agency could use this report as a basis for maintaining an in-house capability to review post-closure criticality safety cases for radioactive waste disposal prepared by UK waste producers and disposers.

Key Words: radioactive waste; fissile material; nuclear criticality; geological disposal; repository performance assessment; radionuclide migration; assay.

1. INTRODUCTION

1.1 Background

The criticality safety of radioactive waste is of concern both to the Environment Agency and to the Health and Safety Executive (HSE), who share regulatory responsibility for radioactive waste conditioning and subsequent disposal in any future repository. The Nuclear Installations Inspectorate (NII) of the HSE is responsible for regulating the nuclear safety of waste stored at licensed nuclear sites, and any future licensable activities associated with waste emplaced in a repository in the period up to closure of the facility and while the repository site is managed after closure. HSE's responsibility includes monitoring the adequacy of the nuclear industry's arrangements with respect to the fissile material content of waste packages. The Environment Agency is responsible for authorising the disposal of radioactive waste, such that post-closure safety of the repository is ensured. The Environment Agency has identified nuclear criticality as an important issue to be addressed in repository post-closure safety assessments[1].

Waste producers currently package intermediate-level waste for future disposal according to specifications set out by United Kingdom Nirex Limited (Nirex), although such specifications have no regulatory status. If the regulators determine that the criteria employed by Nirex for accepting waste for disposal are inadequate, and that, for example, lower limits on the fissile material content of waste drums are required, then existing waste may need to be repackaged. Thus, the regulators should assess post-closure safety conditions on waste packages as soon as possible so that the regulators' requirements can be accounted for in waste packaging operations, and the risks and expense of having to repackage waste in the future minimized. Galson Sciences Limited has been contracted by the Environment Agency to provide a review of approaches to making a post-closure safety case in relation to criticality, so that the Environment Agency is able to provide an effective input to the process of evaluating proposed fissile contents of waste packages.

1.2 Project Objectives, Scope, and Report Structure

The main objective of the project is to identify the factors which influence the treatment of nuclear criticality in post-closure safety assessments of radioactive waste disposal facilities. This objective has been achieved by undertaking the tasks set out in the Environment Agency's Technical Specification for the project. The five technical tasks are:

Task 1 Undertake a review of existing literature on approaches to criticality in repository development programmes.

Task 2 Review of assay techniques for fissile material.

[1] The document "Disposal Facilities on Land for Low and Intermediate Level Radioactive Wastes: Guidance on Requirements for Authorisation" (Environment Agency et al., 1997) requires a safety case to include a demonstration that the possibility of a local accumulation of fissile material such as to produce a neutron chain reaction is not a significant concern.

Task 3 Setting of pre-disposal criticality limits.

Task 4 Examination of factors affecting criticality in post-closure safety assessments.

Task 5 Potential implications of a post-closure criticality event.

Task 1 has involved reviewing key studies of post-closure nuclear criticality which have been undertaken in radioactive waste disposal programmes around the world. The results of this review are presented in Chapter 2. The disposal concepts and the key assumptions and techniques underlying these criticality calculations are discussed in detail. In particular, the different approaches available for evaluating fissile content limits of waste packages, taking into account post-closure criticality safety, have been assessed. Under Task 3, the key issues that need to be considered in setting fissile content limits have been highlighted.

The sequences and combinations of different processes necessary for the concentration of fissile radionuclides at various locations in the disposal system have been discussed in Chapter 3 in terms of possible post-closure nuclear criticality scenarios (Task 4). The information required for the Environment Agency to carry out post-closure criticality analysis has been outlined. The safety implications which might need to be addressed in a post-closure safety case require an understanding of the potential consequences of nuclear criticality. Thus, under Task 5, the potential implications of a post-closure criticality event have also been discussed in Chapter 3. Furthermore, controls that could be introduced to minimize the potential for post-closure criticality have been listed.

The review of post-closure criticality analyses provides insight into the calculational techniques that could be used to set fissile content limits on waste packages. However, waste producers can only demonstrate compliance with these limits to the extent allowed by the techniques currently available to measure the specified waste package properties. Thus, under Task 2, a review of assay techniques for fissile material has been carried out, and the implications of different assay techniques for setting fissile content limits of waste packages, derived from considerations of post-closure criticality, have been discussed. The review has considered the practicalities of each technique, which may involve destructive or non-destructive sampling of the waste, and the accuracy, precision and reliability of the measurements. The results of this review are presented in Chapter 4.

Chapter 5 lists conclusions concerning post-closure criticality analyses, criticality controls, and assay techniques, and recommendations for the Environment Agency's involvement in assessing post-closure criticality safety cases.

The remainder of Chapter 1 discusses the concepts of nuclear criticality and the issue of criticality safety in the context of radioactive waste disposal in the UK. A glossary of criticality terminology is provided in Appendix A.

1.3 Nuclear Criticality

Nuclear criticality refers to a sustained nuclear chain reaction, in which neutrons liberated in fission events induce further fission reactions in surrounding materials. For criticality to occur, a sufficient mass of fissionable material, such as ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu, must form a specific shape at a specific concentration, usually in the presence of a specific amount of a neutron moderator, such as water. In the context of the post-closure safety of a radioactive waste repository, there is concern that a fissile mass may assemble in a critical geometry in the repository or in the geosphere as a result of hydrological, geochemical and mechanical processes. Moderated criticality at a slow rate of assembly is known to have occurred under natural conditions at Oklo in Gabon, West Africa (Wickham and Bennett, 1997). Mining and field investigations in the Oklo region have led to the discovery of sixteen reactor zones that are depleted in ^{235}U.

The repository post-closure criticality analysis presented by Rechard et al. (1996b) included a discussion of nuclear criticality concepts and terminology. This discussion forms the basis of the summary description of nuclear criticality provided here.

1.3.1 Chain reactions

In a fission reaction the nucleus of a fissionable material, such as ^{235}U, absorbs a neutron, which causes it to split into other elements and emit a number of neutrons (fission of ^{235}U releases 2 or 3 neutrons). If the neutrons produced by a fission reaction cause further fission reactions, then a chain reaction may begin. A chain reaction will not occur if sufficient neutrons leave the system, are absorbed by the fissile material without causing fission, or are absorbed by other materials (neutron poisons).

1.3.2 Multiplication factor

Whether or not a system is critical is generally expressed in terms of a neutron multiplication factor, which defines the fission rate of a fissionable material. If n neutrons are introduced into a block of fissionable material and there are kn neutrons in the second generation, there will be k^2n in the third, k^3n in the fourth, and so on. The total number of neutrons that would appear in the block due to n initial neutrons is:

$$n\sum_{r=0}^{r=\infty} k^r$$

If $k < 1$, the sum converges to $n/(1-k)$. Usually, in criticality analyses, the effective multiplication factor is defined as:

$$k_{eff} = \frac{Number\ of\ neutrons\ in\ one\ generation}{Number\ of\ neutrons\ in\ the\ preceding\ generation}$$

Situations resulting from different values of k_{eff} are:

$k_{eff} < 1$ the system is subcritical with zero or decreasing fission rate;

$k_{eff} = 1$ the system is critical at a constant fission rate; and

$k_{eff} > 1$ the system is supercritical at an increasing fission rate.

Evaluation of k_{eff} first requires calculation of the infinite multiplication factor, k_{inf}, from measured physical parameters for a uniform homogeneous medium with infinite size. The actual value of k_{eff} can be determined by accounting for the loss of neutrons from a surface of finite size. Loss of neutrons can reduce a system with $k_{inf} > 1$ to a system with $k_{eff} < 1$.

1.3.3 Nuclear reactors

To promote critical conditions in nuclear reactors, materials are placed around the reactor core to reflect back into the core many of the neutrons that would otherwise escape. Common reflector materials are graphite, beryllium, water, and natural or depleted uranium. Neutron moderators are also used directly in the core of thermal reactors to promote critical conditions by slowing down the high-velocity neutrons from the fission process. Slow (thermal) neutrons are less likely to be absorbed by ^{238}U in the reactor and thus have a much greater probability of producing a fission in ^{235}U and ^{239}Pu (fissile materials). Common neutron moderators used in a nuclear reactor are natural ("light") water, "heavy" water (water with significant quantities of the deuterium isotope) and graphite, because the hydrogen and carbon atoms of these materials slow down neutrons effectively through scattering, while absorbing few of them.

1.3.4 Negative feedback

In a chain reaction, the time interval between neutron generations is small, so that if $k_{eff} > 1$ neutron multiplication is rapid and the activity can rise rapidly. During a supercritical excursion the power and temperature will rise until negative feedback becomes large enough to reduce the system to a subcritical condition. The main negative feedback mechanisms associated with temperature increases are:

- density decrease from thermal expansion, which can increase neutron leakage;
- Doppler effects, which increase the probability that neutrons are absorbed without fission; and
- thermal expansion of the moderator, which causes fewer neutrons to be slowed down.

If supercriticality cannot be counteracted by the negative feedback mechanisms, then enough energy may be produced to melt the fissile material, boil the moderator (solutions), or break the assembly apart.

1.3.5 Positive feedback

It is possible for positive feedback mechanisms to cause the reactivity (and power) of a critical system to increase (autocatalytic criticality). For example, a fissionable material may be subcritical because it contains too much neutron moderator (such as water), which behaves, in part, as a neutron poison (an over-moderated system). Removal of some of the moderator may cause the system to become critical with $k_{eff} = 1$. The resulting heat energy produced by fission reactions may cause further expulsion of the moderator, increasing k_{eff} above 1. Such an instability will continue until the system configuration changes. Bowman and Venneri (1996) proposed that underground disposal of fissile material could result in autocatalytic criticality.

1.4 Consideration of Nuclear Criticality in a UK Radioactive Waste Repository

When fission occurs in a fissile mass, whether it is caused by uranium or plutonium, the event results in the production of energy, fission products, neutrons, and various types of radiation (gamma, beta, etc.). The consequences of a critical event in a repository are discussed in Section 3.3 and include increased radiological hazard and mechanical damage caused by kinetic or heat energy. The potential for post-closure criticality, and the possible risks associated with a post-closure criticality excursion, require consideration in assessments of repository safety. Operators should aim to ensure that radiological doses or risks are as low as reasonably achievable. Thus, it is possible that the likelihood or potential consequences of post-closure criticality would need to be limited by placing restrictions on the fissile material content of the waste packages and repository.

1.4.1 Radioactive Waste Disposal in the UK

The methods used, or proposed, to ensure safe disposal of radioactive wastes depend on the level of radioactivity of the waste material. Radioactive wastes in the UK are classified as Very Low-Level Waste (VLLW), Low-Level Waste (LLW), Intermediate-Level Waste (ILW), and High-Level Waste (HLW) (Environment Agency et al., 1997). The activities of these wastes, and the position concerning their disposal in the UK, are as follows:

- **VLLW** contains less than 400 kBq beta/gamma activity per 0.1 m^3 of material (or single items containing less than 40 kBq beta/gamma). VLLW is safely disposed of with ordinary refuse.

- **LLW** contains radioactive materials not exceeding 4 GBq/tonne alpha or 12 GBq/tonne beta/gamma activity. LLW is currently disposed of at the national LLW disposal facility owned and operated by British Nuclear Fuels Limited (BNFL) at Drigg in Cumbria.

- **ILW** has activities above that of LLW but is not significantly heat generating. Such waste includes Plutonium Contaminated Material (**PCM**). Approximately 80,000 m^3 of conditioned ILW currently exists in the UK. Nirex has responsibility to provide and manage a national disposal facility in the form of a deep repository for solid ILW and some LLW that cannot be disposed of at Drigg. Nirex has investigated the potential for siting an ILW repository near Sellafield (west Cumbria), but, in 1997, failed to obtain planning permission to construct a Rock Characterization Facility in the investigation area.

- **HLW** may increase in temperature significantly as a result of its radioactivity and so may require special consideration in the design of storage and disposal facilities. Approximately 900 m^3 of HLW currently exists in the UK and an estimated 2,300 m^3 of HLW may exist by the year 2030. Current UK Government policy on HLW includes a fifty-year storage period prior to disposal. The Department of the Environment, Transport and the Regions is currently considering requirements for a research strategy in relation to HLW disposal.

1.4.2 Nuclear Criticality Concerns

VLLW poses no risk of nuclear criticality and LLW is thought to present no significant risk of criticality, although it remains for BNFL to demonstrate, as part of the safety case for the Drigg disposal facility, that the possibility of a local accumulation of fissile material producing a neutron chain reaction is not a significant concern. Nuclear criticality considerations are required for ILW and HLW storage and disposal. This project focuses on considerations of post-closure nuclear criticality in a deep ILW repository in the UK, because ILW is currently being packaged for deep disposal according to Nirex's repository design concept, despite the current lack of a repository site. However, many of the results of this study are relevant to criticality concerns with respect to LLW and HLW storage and disposal facilities.

2. POST-CLOSURE CRITICALITY ASSESSMENTS

The likelihood and potential consequences of post-closure nuclear criticality have been studied as part of several radioactive waste disposal programmes, and have drawn attention at an international level, as discussed in Section 2.1. Post-closure criticality studies can be categorized in terms of the type of waste intended for disposal and the associated disposal concept. This report is primarily concerned with the potential for post-closure criticality in a deep ILW repository and, thus, this chapter focuses initially on criticality studies related to ILW facilities. However, the criticality issue has received greatest attention outside the UK in the context of disposal facilities for HLW, and has also been addressed in assessments of LLW disposal facilities, and radioactive waste handling and storage facilities. A review of the methods used to address nuclear criticality concerns at this range of facilities, and the results obtained, will support the development of an approach to addressing criticality concerns at an ILW repository in the UK. Reviews of criticality studies are presented in Section 2.2, and review findings are summarized in Section 2.3.

2.1 An International Perspective

The significance of the post-closure criticality issue has been recognized at an international level. For example, the Nuclear Energy Agency (NEA) Working Group on Integrated Performance Assessments of Deep Repositories (IPAG) drew attention to nuclear criticality through its study of the status of performance assessments (PAs) in member countries of the Organization for Economic Co-operation and Development (NEA, 1997). IPAG examined ten PAs from seven countries (Canada, Germany, Switzerland, Japan, Sweden, Finland, and the United States), primarily through the production of a questionnaire completed by representatives from each of the ten organizations that had conducted the PAs. Survey participants were asked whether their respective PA studies mentioned or addressed criticality. NEA (1997) reported that only the USDOE (1996) assessment of the Waste Isolation Pilot Plant (WIPP) in New Mexico, US, addressed post-closure criticality directly, but that the SKI (1996) and Goodwin et al. (1994) assessments of deep repositories in Sweden and Canada respectively, referred to earlier studies that showed post-closure criticality to be unlikely (Behrenz and Hannerz, 1978; McCamis, 1992). The issue of criticality was not addressed in the other projects surveyed.

Another NEA working group oversaw the development of an international database of features, events and processes (FEPs) potentially relevant to the performance of a repository for radioactive waste (NEA, 1998). The international database contains FEP lists and, in most cases, FEP descriptions from seven repository programmes (Goodwin et al., 1994; Andersson, 1989; Miller and Chapman, 1993; Nagra, 1994; USDOE, 1996; SKI, 1996; NEA, 1992). Each project FEP list includes nuclear criticality, and any associated discussions generally make reference to supporting studies that show post-closure criticality is unlikely to occur (Behrenz and Hannerz, 1978; McCamis, 1992; Rechard et al., 1996a).

Recently, Bowman and Venneri (1996) suggested that the potential exists for an atomic explosion in the proposed HLW repository at Yucca Mountain, Nevada, US. This work

prompted several further studies in the US that strongly criticized the Bowman and Venneri (1996) analysis, and concluded that the likelihood of an atomic explosion in a repository is low (Van Konynenburg, 1996; Kastenberg et al., 1996; Rechard et al., 1997).

In summary, several key evaluations of the potential for nuclear criticality in a deep radioactive waste repository have received international attention, including the pioneering analysis of Behrenz and Hannerz (1978) and the controversial study by Bowman and Venneri (1996). Reviews of these key studies are included in Section 2.2.

2.2 A Review of Post-Closure Criticality Studies

Reviews of criticality studies have been summarized in terms of:

- the aims of the studies;
- the key features and assumptions associated with the disposal concept and criticality analysis, including waste-form, waste packaging and waste emplacement configuration;
- the scenarios and factors considered to have the potential to result in a critical event;
- the approaches and models used in quantifying the probability and/or consequences of criticality; and
- the key results of the work regarding the probability and consequences of criticality, and any implications of the work for setting waste acceptance criteria.

The following criticality studies have been reviewed and are summarized here in tabular form, with related studies discussed together; more detailed reviews are provided in Hicks (1998):

Proposed deep ILW repositories

Table 2.1	Sellafield, UK	Wise et al. (1997)
Table 2.2	Waste Isolation Pilot Plant, New Mexico, US	Rechard et al. (1996a)
Table 2.3	Konrad Repository, Germany	Berg and Gmal (1993)

Proposed spent nuclear fuel/HLW repositories

Table 2.4	Sweden	Behrenz and Hannerz (1978) Oversby (1996)
Table 2.5	Canada	McCamis (1992) Johnson et al. (1994)
Table 2.6	Yucca Mountain, US	Bowman and Venneri (1996)
Table 2.7	Yucca Mountain, US	Kastenberg et al. (1996)
Table 2.8	Yucca Mountain, US	Rechard et al. (1995) Rechard et al. (1996b) Rechard et al. (1997)
Table 2.9	Finland	Anttila (1996)

Other criticality studies

Table 2.10	Envirocare LLW facility, Utah, US	Hopper and Parks (1997) Toran et al. (1997)
Table 2.11	Babcock and Wilcox uranium processing facility, Virginia, US	Alcorn (1997)
Table 2.12	Deep-hole disposal, Russia	Kouzmine et al. (1997)

Table 2.1 Criticality analysis for a proposed LLW/ILW repository in the UK (Wise et al., 1997)

Sellafield, UK

Aims of the analysis

Wise et al. (1997) undertook a criticality analysis as part of Nirex's development of a general criticality safety case (GCSC) for ILW and LLW disposal in a proposed deep repository at Sellafield. The analysis aimed to derive fissile mass limits for individual waste packages that contain low levels of fissile material, which would demonstrate the criticality safety of these containers in a final repository. Nirex will require waste producers either to demonstrate compliance with the criticality limits derived in the GCSC, or to make a criticality safety case for packages containing higher levels of fissile material.

Key features and assumptions

- Nirex's repository design involves a multi-barrier containment system, comprising engineered and natural barriers.
- Waste will be packaged in grout in 500-litre stainless steel containers, and the repository will be backfilled with a cement-based material.
- ILW packages will collectively contain up to 5 tonnes of ^{239}Pu and 12 tonnes of ^{235}U.
- The criticality analysis assumed that ^{239}Pu is homogenized with polyethylene (neutron moderator).
- The steel drum wall was the only neutron absorber assumed in the analysis. The rest of the material in each drum was assumed to have no effect on the neutronics calculations.

Scenarios assessed

- Criticality in a single intact drum.
- Criticality as a result of neutron interaction between neighbouring intact drums.

Methodology

Wise et al. (1997) determined the mass and concentration of ^{239}Pu in a drum for which $k_{eff} = 0.95$ (with a standard deviation of less than 0.003). An infinite array of drums was assumed, and the drum wall thickness, drum material, fissile mass shapes and locations, and backfill properties were varied. The Monte Carlo neutronics code MONK6B was used to calculate k_{eff}.

Results

- The selected general case assumed 2.5-mm thick, 316L steel containers, void between the drums, and four fissile quarter spheres in the corners of four drums in closest proximity. The calculated maximum allowable fissile mass for the GCSC was **50 g ^{239}Pu per 500-litre drum** at a concentration of 45 g ^{239}Pu per litre of polyethylene.
- For a less pessimistic case in which the ^{239}Pu was assumed to be homogeneously dispersed in a water-filled drum, the minimum critical mass was calculated to be about **3.5 kg ^{239}Pu per 500-litre drum** at a concentration of 7 g ^{239}Pu/litre.

Sellafield, UK

Additional findings

- The location and shape of fissile units is a significant factor in controlling the allowable fissile mass.
- The drum wall acts as a neutron absorber and the allowable fissile mass is sensitive to the drum wall steel specification. Any reduction in drum wall thickness would reduce the allowable fissile mass.
- The addition of backfill between the drums increases the allowable fissile mass per drum, because the presence of hydrogen in the backfill material ($Ca(OH)_2$) prevents neutron interaction between the drums. Wise et al. (1997) noted that timing of backfilling of the repository has yet to be decided by Nirex. Thus, the assumption for the GCSC that there is no backfill is conservative with respect to the criticality analysis.
- The reactivity of a waste drum would be increased if a neutron reflector material was assumed to be present in the drum. Thus, the general case value applies for containers that do not contain neutron reflectors or moderators, such as beryllium or graphite.

Other relevant analysis

Plutonium contaminated material in decommissioning waste produced at Sellafield is packed into 200-litre storage drums. A fissile mass limit of 230 g Pu for a 200-litre drum has been determined for arrangements of stacked drums under normal operating conditions (Ogilvie and Harris, 1997). The reactivity of a stack of drums was determined by comparison with criticality calculations for an optimally moderated plutonium metal/water slab with water reflection on one side and concrete reflection on the other. The 200-litre drums are supercompacted and the compact is loaded into 500-litre drums and grouted for disposal, with a criticality safety limit of 260 g Pu per 500-litre drum.

Miller and Chapman (1993) discussed nuclear criticality as part of their consideration of the features, events and processes that could affect the performance of a LLW/ILW repository in the UK. This analysis formed part of an assessment of the Sellafield site undertaken on behalf of Her Majesty's Inspectorate of Pollution. Miller and Chapman (1993) remarked that ^{235}U would not be emplaced in sufficient quantities in a repository for a fissile mass to accumulate, and that criticality could only occur if a large mass of ^{239}Pu formed by some concentration process. They considered that the accumulation of a critical mass of ^{239}Pu was improbable in a repository and recommended that criticality should not be included in repository performance assessments. However, these considerations were not based on any quantitative analysis of post-closure criticality.

Table 2.2 Criticality analysis for the WIPP repository in the US (Rechard et al., 1996a)

WIPP, US

Aims of the analysis

The Waste Isolation Pilot Plant (WIPP) in New Mexico, US, is planned to receive transuranic radioactive wastes generated by US Department of Energy (DOE) defence programmes. Rechard et al. (1996a) analysed the potential for nuclear criticality at the WIPP, with the aim of demonstrating that post-closure criticality is of low probability and low consequence.

Key features and assumptions

- The repository will contain about 21 Mg ^{239}Pu fissile gramme equivalent.
- Waste will be emplaced in disposal rooms backfilled with MgO.
- The repository is located 655-m deep in low-permeability, low-porosity bedded salt (the Salado Formation).
- The Culebra dolomite is the most transmissive unit above the repository host-rock (in the far field).
- The oxidation state of the repository and the Culebra will remain reducing.
- The WIPP Waste Acceptance Criteria (USDOE, 1991) include requirements that the fissionable radionuclide content of a 0.21-m^3 (208-litre) drum is less than 200 g ^{239}Pu fissile-gram equivalent (FGE), and the fissionable radionuclide content of a 1.8-m^3 (1,800-litre) waste box is less than 350 g ^{239}Pu FGE.

Scenarios assessed

- Criticality in the repository near field.
- Criticality in the host rock.
- Criticality in the far field (the Culebra dolomite) following radionuclide transport through a repository intrusion borehole.

Methodology

Rechard et al. (1996a) calculated the minimum mass required for criticality in the Culebra and the repository using the Monte Carlo neutron transport code, MCNP. The results of the 1996 WIPP PA were used to assess whether such masses could accumulate by dissolution, compaction, adsorption, colloid filtration, or precipitation. The likelihood and potential consequences of criticality were assessed by analogy with aqueous accidents and observations of the Oklo natural reactors.

WIPP, US

Results

- A minimum of about **2.2 kg ^{239}Pu** is required for criticality in the Culebra or repository (assuming Culebra brine is present) at a concentration of **3 kg ^{239}Pu m^{-3}**.
- A minimum of about **50 kg ^{235}U** is required for criticality in the Culebra or repository (assuming Culebra brine is present) at a concentration of about **10 kg m^{-3}**.
- A minimum of about **700 kg ^{239}Pu** is required for criticality in the Salado at a concentration of about **50 kg m^{-3}**.

Probability

- Fracture apertures of at least 0.45 m are needed to accommodate a fissile mass at the required concentration for criticality, but fracture apertures of such size are unlikely to exist near the repository or in the Culebra.
- Critical concentrations of ^{239}Pu and ^{235}U are unlikely to form in the repository or the host-rock by dissolution, compaction, adsorption, colloid filtration, or precipitation.
- PA calculations show that critical masses of ^{239}Pu and ^{235}U are unlikely to reach the Culebra or other strata as a result of human intrusion into the repository.

Consequences

- The number of fissions represented by the waste in the repository (10^{27} fissions) is a factor of three more than the maximum number of criticality fission products possible in the repository.
- The maximum 110 kg ^{239}Pu that could potentially reach the Culebra could be represented by about 10^{25} fissions, which is two orders of magnitude less than the number of fissions represented by the repository.
- Sudden assembly of fissile material in the repository is unlikely to cause significant damage. A void of less than 1.3 m radius would be generated in the salt at repository depth.
- The small amount of rapidly produced energy from sudden assembly of fissile material is unlikely to cause significant damage in the Culebra dolomite. A void of less than 0.8 m radius would be formed.
- By comparison with Oklo, the thermal power produced in the Culebra as a result of criticality would be 3.7 kW, much less than the initial heat load in the repository (137 kW).

Table 2.3 Criticality analysis for the Konrad Repository in Germany (Berg and Gmal, 1993)

Konrad Repository, Germany

Aims of the analysis

The radioactive waste destined for the Konrad Repository in Germany must be non-heat generating, but may contain residual quantities of ^{239}Pu and ^{235}U in solid form. Berg and Gmal (1993) analysed the criticality safety of the repository to derive limits on fissile material in waste containers.

Key features and assumptions

- The emplacement room is 7.0 m wide and has a height of 6.0 m (with a 1.15-m gap at the top).
- There are 11 types of waste container with volumes ranging from between 0.7 m^3 (700 litres) to 10.9 m^3 (10,900 litres). Waste is either conditioned directly in the container or is first placed in 200 litre (0.2 m^3) drums.
- The container stacking sections are 2.5 x 10^4 m long in total and each stack is 1.24 m long.
- Ground water enters the emplacement area in the post-operational phase, but not in the operational phase.
- The fissile content of waste packages is limited by the most restrictive of the following:
  - The concentration of fissile material in a waste package must be less than **50 g per 100 litres of the waste form** (0.5 kg m^{-3}),.which is a safety requirement during handling and emplacement.
  - The mass of fissile material in a container must be less than 45% of the smallest calculated critical mass, which is a safety requirement during handling and emplacement.
  - The mass of fissile material in a stacking section of an emplacement room must be restricted to the smallest calculated critical spherical mass of U/Pu-oxide mixtures with water and a concrete reflector, which is a post-operational phase safety requirement.

Scenarios assessed

Leaching of fissile material from several waste packages results in accumulation of fissile material at high concentrations in a hole at the base of the emplacement room.

Methodology

Calculations were performed for stacked assemblies of different waste packages. Critical masses, k_{inf} or k_{eff} were determined for different spherical systems using XSDRNPM, a discrete ordinate transport code. More complicated stacking assemblies were analysed using the Monte Carlo code, KENO IV. k_{inf} was calculated for a typical emplacement room based on the fissile mass concentration limit. To determine the mass limit in a stacking cross-section, a spherical geometry of fissile material was assumed and separate calculations were made with k_{eff} = 1 for ^{239}Pu, ^{241}Pu, ^{233}U (low [< 5%] and high [> 5%] enrichment), and ^{235}U (low and high enrichment). The permissible mass of fissile material per waste package was calculated based on the total permissible mass in a cross-section of a room and the number of waste packages that can fit in the cross-section.

Konrad Repository, Germany

Results

- For an assembly of eight containers with a maximum mass concentration of 50 g ^{239}Pu per 100 l of the waste form, $k_{inf} < 0.6$. For a maximum concentration of ^{241}Pu in the same configuration $k_{inf} < 1$.
- For one stack of containers in a cross-section of a room, with $k_{eff} = 1$, the fissile mass limit is: 13 kg for ^{235}U (low enrichment); 2 kg for ^{235}U (high enrichment); 4 kg for ^{233}U (low enrichment); 1.1 kg for ^{233}U (high enrichment); 0.55 kg for ^{241}Pu; and 1.1 kg for ^{239}Pu.
- The calculated admissible mass of fissile material for waste packages (700 litre to 10,900 litre) is: **120 - 850 g ^{235}U (low enrichment); 50 - 350 g ^{235}U (high enrichment); 70 - 500 g/ ^{233}U (low enrichment); 28 - 220 g ^{233}U (high enrichment); 14 - 110 g ^{241}Pu; and 28 - 220 g/ ^{239}Pu.**
- The 700-litre container will be packed with one 200-litre drum. The calculated admissible mass of 120 g ^{235}U (low enrichment) for the 700-litre container correlates to a concentration of 0.6 g ^{235}U (low enrichment)/litre in the waste drum. This concentration exceeds the critical mass concentration limit of 0.5 g/l.
- The maximum admissible mass of fissile material in the repository is: 2.22×10^4 kg (5×10^{16} Bq) ^{239}Pu; 1.109×10^4 kg (4×10^{19} Bq) ^{241}Pu; 2.2×10^4 kg (8×10^{15} Bq) ^{233}U (high enrichment); 8.1×10^4 kg (3×10^{16} Bq) ^{233}U (low enrichment); 4.0×10^4 kg (3×10^{12} Bq) ^{235}U (high enrichment); 2.6×10^5 kg (2×10^{13} Bq) ^{235}U (low enrichment).

Additional findings

- The activities at the end of the operational phase are expected to be up to two orders of magnitude less than those calculated for a criticality-safe layout.
- A summation formula was developed for a radionuclide mixture (^{235}U, ^{233}U, ^{241}Pu, and ^{239}Pu) in a waste package.

Table 2.4 Criticality analysis for a spent fuel repository in Sweden (Behrenz and Hannerz, 1978; Oversby, 1996)

Spent fuel repository, Sweden

Aims of the analysis

Behrenz and Hannerz (1978) and Oversby (1996) evaluated the potential for criticality in a spent-fuel repository in Sweden. Their analysis assumed that a repository would be excavated deep in wet crystalline rock and would contain large amounts of fissile material (primarily ^{239}Pu and ^{235}U) from Boiling-Water Reactor (BWR) and Pressurized-Water Reactor (PWR) spent fuel rods.

Key features and assumptions

- The Behrenz and Hannerz (1978) analysis assumed 200-mm thick copper-walled canisters (2.19 m^3) with the free space in the canisters filled with lead cast into place (KBS concept). Oversby (1996) assumed combined copper/steel canisters (2.95 m^3) (KBS-3 concept).
- The repository is at a depth of 500 m in low-permeability crystalline rock.
- The canisters are placed in vertical holes drilled from horizontal tunnels.
- Canisters are surrounded with a buffer material (compacted bentonite).
- The tunnels above the deposition hole are filled with a mixture of quartz sand and bentonite.
- KBS canister inventory 100 years after disposal will include about 14 kg ^{235}U and 9 kg ^{239}Pu.
- Fracture apertures are about 0.1 mm with fracture spacings of a few metres.
- Water entering the deposition holes and tunnels will cause the bentonite to swell.

Scenarios assessed

- Plutonium criticality inside a canister after early canister failure (within the first 10^5 years), removal of neutron absorbing uranium, and inflow of moderating water.
- Plutonium criticality in a deposition hole after early canister failure and selective precipitation of plutonium.
- Uranium criticality in a deposition hole after canister failure. In-growth from decay of ^{239}Pu and ^{243}Am will increase ^{235}U to a level where a criticality incident is theoretically possible, but uranium from many canisters would need to accumulate in one location to form a critical assembly.
- Uranium criticality in a tunnel after canister failure, uranium transport, and uranium precipitation in the tunnel.

Methodology

Reactor physics calculations to determine the infinite neutron multiplication factor were carried out using the ASEA-ATOM code MICØ. Critical masses and geometries were determined for each scenario and the potential for these critical masses to form was evaluated based on expected diffusion rates through the different repository materials and dissolution, precipitation, and absorption rates.

Spent fuel repository, Sweden

Results

- Insufficient uranium can be removed from the canister for plutonium criticality to occur in the canister.
- A minimum of about **2.2 kg of plutonium** are required for criticality in water-saturated bentonite at a concentration of about **40 kg/m^3**, but insufficient plutonium (maximum of about 1 kg) will be available in the buffer at any time for a critical mass to be formed. Criticality in the bentonite without water would require more than 100 kg of plutonium.
- The radiological impact of plutonium criticality is more than an order of magnitude less than the pre-existing radiological impact of the canister.
- Uranium from several canisters would need to accumulate in one deposition hole for a critical mass to form, but this was considered unlikely.
- A minimum critical mass of **4,400 kg of uranium** (1.66% enriched) is required for criticality in a tunnel, requiring uranium from at least 4 canisters. Insufficient uranium (maximum of about 200 kg) will be available in the tunnel at any time for a critical mass to be formed.
- Uranium criticality would not result in a sudden release of energy, and the reactor would have a strongly negative temperature and power coefficient. If all uranium (227,000 kg) from all 160 holes in one tunnel formed a critical configuration at a uranium density of 780 kgU/m^3, the power of the reactor would be less than 130 kW. The reactor would generate no more than 3×10^6 MWd of energy over 10,000 years before it shuts down due to depletion of fissile material. The amount of long-lived fission products and radionuclides formed would be about 10% of the original repository inventory.
- Based on analogy with the Oklo natural uranium reactors (Gabon, Africa), Oversby (1996) determined that uranium criticality could not occur in a canister. Further, a volume of 120 m^3, containing 143,000 kg of uranium (more than the total uranium from 71 canisters), would be required to achieve an Oklo-type criticality in the tunnel. Oversby (1996) considered that such migration and reconcentration of uranium would be unlikely.

Additional findings

- The potential for uranium criticality in the tunnels could be minimized by adding 7% neutron absorbing magnetite to the quartz and sand tunnel fill.
- Neither plutonium nor uranium criticality would be possible in fractures in the host rock because the fracture apertures are too small for a critical assembly to form.
- The reducing conditions of the deep ground water of the Fennoscandian Shield would prevent long-range transport of uranium, because of the low solubility of uranium (IV).
- If a large proportion of the bentonite was removed from the deposition hole, then the potential for criticality would be increased in all scenarios owing to increased transport rates and the reduction in the neutron absorption capacity of the bentonite.

Table 2.5 Criticality analysis for a spent fuel repository in Canada (McCamis, 1992; Johnson et al., 1994)

Spent fuel repository, Canada

Aims of the analysis

McCamis (1992) undertook criticality safety calculations in support of an evaluation of spent nuclear fuel disposal in deep underground facilities in Canada. This analysis was extended by Johnson et al. (1994) as part of a post-closure performance assessment.

Key features and assumptions

- The repository is at a depth of between 500 m and 1,000 m in the plutonic rock of the Canadian Shield.
- Titanium containers will hold immobilized spent fuel and interior voids will be filled with small glass spheres.
- Containers will be placed in boreholes drilled in the floor of emplacement rooms and surrounded by a clay-based buffer (a clay-sand mixture).
- The excavations will be backfilled with low-permeability material comprising mixtures of clay and crushed granite and mixtures of clay and sand.
- Each container will hold about 1,363 kg U and about 3.73 kg of ^{239}Pu.

Scenarios assessed

- Plutonium criticality in a container after container failure and water inflow.
- Plutonium criticality in the water-saturated near field (borehole buffer or tunnel backfill) following container failure and migration and accumulation of plutonium.

Methodology

The Monte Carlo criticality safety code KENO V.a was used in the analysis. The multiplication factor, k_{eff}, was calculated for the canister criticality scenarios assuming the materials were water saturated. The critical volume ($k_{eff} = 1$) of an aqueous plutonium solution in the near field as a function of plutonium concentration was determined for mixtures of water-saturated sand and buffer or backfill. Estimates of ^{239}Pu dissolution and diffusion rates were used to assess the potential for achieving these critical volumes in the near field.

Spent fuel repository, Canada

Results

- For realistic fuel geometries, with fresh fuel bundles, k_{eff}'s were calculated to be about 0.4.
- For a uniform mixture of fissionable material and container materials, k_{eff} remained below 0.5.
- The smallest calculated radius of a critical spherical mass was 19 cm, involving **2.9 kg ^{239}Pu** at a concentration of **10^{-1} g/cm^3** (100 kg/m^3) in buffer host material.
- Within the first 500 years radiolysis will keep the redox conditions oxidizing and the dissolution rate of UO_2 (hexavalent) will be high. After 500 years the redox state is expected to be reducing and the dissolution rate of UO_2 (tetravalent) will be low. Based on estimated fuel dissolution rates assuming early canister failure, a total mass of 106 g ^{239}Pu would be released in 3.5×10^4 years (the time of maximum ^{239}Pu inventory). Criticality can not occur because the mass of ^{239}Pu required is greater than the maximum estimated release of ^{239}Pu.
- The amount of ^{239}Pu released from 27,468 containers in a single vault of the repository was calculated, based on dissolution rates and the rate of diffusion through the buffer in the boreholes. A maximum of about 1.5 g of ^{239}Pu from all 27,468 containers was calculated to accumulate in the buffer after about 3.5×10^4 years, with about 5×10^{-6} g of ^{239}Pu around each container. Thus, criticality cannot be achieved in the buffer, backfill or host rock, even if plutonium from many containers accumulates in one location.

Additional findings

The materials placed around a waste container should be highly compacted to minimize pore space and, thus, to minimize the probability that a solution containing fissionable materials might collect in a critical geometry.

Table 2.6 Criticality analysis for a spent fuel/HLW repository in the US (Bowman and Venneri, 1996)

Spent fuel/HLW repository, US

Aims of the analysis

Bowman and Venneri (1996) conducted an analysis that aimed to show that underground disposal of thermally fissile material (TFM) could lead to underground supercriticality.

Key features and assumptions

- The disposal concept involves vitrification of plutonium followed by storage in deep boreholes.
- A typical 60-cm diameter, 3-m long borosilicate glass log would contain about 90 kg of plutonium.
- Rock may be represented by SiO_2 with a density of 2.2 g cm^{-3}.

Scenarios assessed

- After some time the emplacement canister loses its integrity, water enters the canister, the vitrified material begins to dissolve and the TFM in the canister becomes critical.
- The TFM is dispersed as a colloidal suspension, or in solution, into a region containing moderators, resulting in criticality. Dry criticality with positive feed back (autocatalytic criticality) occurs if enough plutonium is present at the emplacement site.

Methodology

Criticality calculations were made using the MCNP code. Critical compositions of pure ^{239}Pu, water, and SiO_2, in spherical geometries, surrounded by an SiO_2 reflector, were calculated with $k_{eff} = 1$, and trends in criticality excursions were examined qualitatively. Upon reaching criticality, energy generation will cause water to be expelled from the system. If a system reaches criticality from an over-moderated condition, the system will exhibit positive feedback and be autocatalytic.

Spent fuel/HLW repository, US

Results

- Criticality resulting from water entering a system will exhibit negative feedback, because the system will shut down as fission energy causes water to be expelled.

- Critical systems resulting from plutonium migration to wet rock could result in positive or negative feedback mechanisms. Positive feedback can be achieved for quite small plutonium masses (e.g., a 30-cm radius sphere with a mass of 1 to 2 kg ^{239}Pu), before water expulsion eventually leads to subcritical conditions. Feedback is always negative for spheres of radius less than about 25 cm.

- Drying out could result in supercriticality with positive feedback for systems containing large ^{239}Pu masses at low plutonium mass fractions. Such systems are initially over-moderated because the neutron absorbing capacity of water exceeds its moderating capacity and, thus, removal of water increases reactivity.

- Repeated criticality caused by water entry and expulsion could lead to redistribution of fissile material into an increasingly larger radius sphere containing an increasing amount of moderating rock. If this involves large quantities of ^{239}Pu (over 50 kg) dispersed into a sphere of large radius (over 200 cm), then criticality with positive feedback could eventually occur. As the temperature of the dry supercritical system increased (up to about 5,000 K), the water density would decrease, which would result in increased reactivity in the prompt critical stage.

- Criticality with large positive feedback could release significant energy before the excursion was terminated by system expansion and/or temperature increase. The energy produced could vaporize up to 2×10^5 kg of rock (a spherical volume of 2.8 m radius). The prompt kinetic yield could affect neighbouring waste packages or could contribute to dose if the gas reached the atmosphere.

Other relevant analysis

The study by Bowman and Venneri (1996) was the subject of extensive discussion and publication in national newspapers in the US, and raised public concern regarding the potential for nuclear criticality at the proposed radioactive waste repository at Yucca Mountain in the US. This work prompted several further studies that strongly criticized the Bowman and Venneri (1996) analysis, and showed that the likelihood of an atomic explosion in a repository was extremely low (Van Konynenburg, 1996; Kastenberg et al., 1996; Rechard et al., 1997). For example, Van Konynenburg (1996) noted that Bowman and Venneri (1996) had made unrealistic assumptions concerning the properties and behaviour of the waste, rock, and water, and that plausible explanations of processes that could result in the rapid formation of critical configurations had not been presented. In conclusion, concerning the Bowman and Venneri (1996) study, Van Konynenburg (1996) asserted that "we do not believe it would make a useful contribution to the literature in the field of criticality safety in geologic disposal of fissile materials".

Table 2.7 Criticality analysis for a spent fuel/HLW repository in the US (Kastenberg et al., 1996)

Spent fuel/HLW repository, US

Aims of the analysis

Kastenberg et al. (1996) undertook further analysis of the potential for autocatalytic criticality of fissile materials in geologic repositories, in particular with respect to the proposed spent nuclear fuel/HLW repository at Yucca Mountain, Nevada, US.

Key features and assumptions

- The repository is located in unsaturated, highly-fractured welded tuff, a minimum of 200 m below the surface and 300 m above the water table.
- Metal canisters of vitrified waste are emplaced horizontally in mined drifts.
- Four classes of materials are being considered for potential geologic disposal: commercial spent fuel with typical effective ^{239}Pu and ^{235}U enrichments of 2%; vitrified military reprocessing wastes containing trace quantities of plutonium and uranium; separated excess weapons plutonium (about 50,000 kg), which may be immobilized in glass or ceramic; and highly enriched uranium from research and naval reactors (about 210,000 kg). A typical canister of commercial spent fuel would contain around 60 kg of ^{239}Pu and 40 kg of ^{240}Pu.

Scenarios assessed

Autocatalytic criticality with venting of radioactivity to the atmosphere, which requires that:

- waste packages degrade before significant decay of fissile material;
- chemical processes separate neutron absorbing poisons (such as boron and ^{238}U) from the fissile material;
- the fissile material is dispersed into a moderating material around the original emplacement or carried away (in solution or in colloid form);
- a sufficient quantity of fissile material is available for transport;
- the fissile material is deposited in a fissile configuration;
- positive reactivity feedback occurs as the system heats; and
- the dynamic response of the system keeps k_{eff} above unity until sufficient energy has been released to cause venting of radioactivity to the atmosphere.

Methodology

Reactivity feedback mechanisms (water removal, TFM temperature increase, rock temperature increase, homogenization of TFM and rock, expansion, and buildup of fission and transmutation products) were studied for over-moderated accumulations of TFM in fractures and spherical masses away from the emplacement location. The BONAMI-NITAWL-XSDRN code and MCNP, a 3-D Monte Carlo code, were used in the analysis to calculate the neutron multiplication factor. The amount of uranium that can be sorbed in the tuff was calculated based on estimated solubilities and diffusion-advection-sorption analysis.

Spent fuel/HLW repository, US

Results

- Dry criticality in tuff rock would require **85.6 kg of ^{239}Pu** under optimum conditions. The mass of ^{239}Pu (60 kg) in one canister would be insufficient to result in dry criticality.

- Water removal, TFM temperature increase, rock temperature increase, and homogenization of TFM and rock have a positive reactivity feedback. Expansion reduces neutron leakage and provides the primary mechanism for reducing the multiplication factor. The build-up of fission products and other neutron absorbers has a negligible effect. Positive feedback from pore water removal disappears if the rock neutron absorber concentration is sufficiently large.

- The amount of energy release for a homogeneous 2-m radius sphere of 254 kg ^{239}Pu in tuff was examined. Homogenization led to large reactivity insertion and the power increased rapidly at 5.8 msec. The system became subcritical at 6.8 msec, on reaching a core radius of 3.1 m. The energy release was 1.3 10^{12} J. The overburden is sufficient to contain this energy release, but the potential for venting depends on the proximity of the critical excursion to, and the properties of, the engineered repository.

- Plutonium will not travel large distances to form critical configurations at Yucca Mountain. Plutonium is more likely to be mineralized on glass surfaces, to form particulates on the drift floor, or to be deposited (by colloid filtration or sorption) in fractures near the emplacement area. The probability of a critical mass of plutonium forming is small because of the small travel distances involved and the relatively rapid decay rate of ^{239}Pu compared to the rate of degradation of the waste form. The primary role of ^{239}Pu in criticality is in producing ^{235}U by radioactive decay.

- Water chemistry at Yucca Mountain is conducive to high uranium solubility. Once uranium is in solution it is unlikely to precipitate into critical configurations due to lack of reducing agents. Based on water infiltration rates, solubilities, and sorption coefficients, a calculated sorbed concentration of 0.02 kg U/m^3 could accumulate in fractures, which is too small for a critical assembly.

Additional findings

Criticality concerns could be alleviated by:

- providing low solubility neutron absorbers (poisons) in the waste form to reduce the probability of criticality in emplacement area;

- diluting ^{239}Pu or highly enriched uranium with depleted uranium (99.7% ^{238}U); and

- using backfill to immobilize plutonium colloids.

Table 2.8 Criticality analysis for a spent fuel/HLW repository in the US (Rechard et al., 1995; Rechard et al., 1996b; Rechard et al., 1997)

Spent fuel/HLW repository, US

Aims of the analysis

Rechard et al. (1995), Rechard et al. (1996b) and Rechard et al. (1997) assessed the potential for criticality in a spent nuclear fuel repository (containing highly enriched uranium) in unsaturated tuff.

Key features and assumptions

- The repository is located in unsaturated volcanic tuff, similar to Yucca Mountain, lying 286 m below the surface and 333 m above an aquifer.
- The stainless steel canisters include borated stainless steel to absorb neutrons.
- The repository will contain 70,000 metric tons of heavy metal (MTHM) including 12,060 MTHM of US DOE spent fuel or equivalent high-level waste in 2,643 containers. About 210 MTHM (210 Mg) of the US DOE spent fuel will be originally highly enriched, of which more than 20% will be ^{235}U.
- The waste containers are emplaced horizontally, and the container spacing is small enough to allow heat from radioactive decay to keep the containers dry for several hundreds of years.

Scenarios assessed

Criticality requires container failure and removal of neutron-absorbing boron from the container. Criticality scenarios involving slow, fast or explosive assembly were considered:

- A moderated slow rate of assembly is represented by the natural reactors in uranium ore deposits at Oklo in Gabon, Africa. This is the most likely type of criticality that could occur in or near a repository because the media (SiO_2 content, porosity, and fracturing) are similar.
- A moderated fast rate of assembly corresponds to the critical aqueous-type accidents that have occurred in nuclear fuel processing plants. This type of criticality excursion could result from a human intrusion that generated a slurry of fissile material and water.
- A moderated explosive assembly was hypothesized by Bowman and Venneri (1996) (see Table 2.6).

Methodology

The probability of assembling a critical configuration, and the consequence of the continued cyclic operation of a nuclear chain reaction were assessed. The MCNP code was used to calculate k_{eff} for different amounts of water and uranium in various mineral forms uniformly distributed within tuff pores. Rechard et al. (1995) estimated the amount of ^{235}U that can be removed from a container based on container failure, dissolution, fluid flow, transport, and precipitation rates.

Results

- The probability of uranium criticality in a container within 10,000 years was estimated to be 3.8 x 10^{-3}. The rate of formation of critical masses was calculated based on estimates of the probability of a container being placed under a dripping fracture (20% based on a fracture spacing of 25 m and a container length of 5 m), the amount of fissile material in the repository (210 Mg ^{235}U), and analogy with the conditions required for the formation of the Oklo reactors. About 133 Mg of uranium were estimated to be associated with each Oklo reactor zone, and each reactor was estimated to have had a minimum operating life of about 2 x 10^5 years. Thus, one Oklo-like criticality was assumed to occur per 133 Mg of uranium per 2 x 10^5 years (3.75 x 10^{-8} events/Mg ^{235}U/year). The rate of formation of critical masses in the repository is thus 1.6 x 10^{-6} events/year (0.2 x 210 Mg ^{235}U x 3.75 x 10^{-8} events/Mg ^{235}U/year). The probability of uranium criticality was calculated by integrating a failure-rate function over 7,300 years, based on the time for container failure by corrosion (300 years), and the subsequent time for complete boron removal from the container (7,000 years).

- The probability of uranium migrating from the repository and forming a critical mass in the tuff was estimated to be 2.5 x 10^{-3} in 10,000 years, based on the conditions required for formation of the Oklo reactors, estimated uranium solubility, and flow rates of water through the repository. Based on similar analysis, the probability of a critical event involving plutonium is less than 10^{-4} in 10,000 years, because plutonium has a lower solubility than uranium.

- In both moderated and unmoderated fast rates of assembly (critical aqueous-type accidents), the fission yield is 10^{15} to 10^{20} fissions, which is negligible compared to the 10^{30} fissions in the repository.

- Slow rates of assembly could be considered continuous and so would produce more total fissions. One critical event (assuming 10^{20} fissions) each day for 1,000,000 years would amount to 10^{28} fissions, which is only 1 % of the fission inventory represented by the repository.

- The thermal power from criticality in a container at one event per day was estimated to be less than 21 kW, which is less than 0.5% of the thermal power produced from radioactive decay at the time the waste is placed in the repository.

- The thermal power associated with criticality in the volcanic tuff was estimated to be 7 kW.

- Fast aqueous and unmoderated criticality, associated with, for example, the sudden collapse of a container, could release energy over a short time (about 1.87 10^7J). The extent of the resulting potential damage to the host rock would be no greater than the damage caused by the excavation of the repository tunnel.

- Rechard et al. (1995) calculated that, with sufficient precipitation of soddyite within a sphere (about 30 cm radius), k_{eff} is greater than 1 with less than **7 kg of ^{235}U**. Calculations indicated that an average of 4.8 kg/container of ^{235}U could be removed per 10,000 years and so criticality would be possible with selective precipitation of uranium minerals.

Spent fuel/HLW repository, US

Additional findings

- A moderated explosive assembly with positive feedback is unlikely to occur because:
  - volcanic tuff contains several elements that can significantly absorb neutrons;
  - a critical assembly would be a heterogeneous mixture that can shield the fissile mass from neutrons;
  - removal of water from an over-moderated system by natural processes (such as evaporation) would take much longer than the microseconds required for a nuclear explosion;
  - no mechanism has been identified or observed that could cause a system to go from a near-critical state to a state in which a nuclear explosion occurred solely as a result of positive feedback;
  - over-moderated systems in which fissile material is being deposited would reach a negative-feedback, Oklo-like criticality configuration, before the initial conditions required for criticality with positive feedback could be achieved. Negative feedback criticality would occur first because it requires less fissile mass, albeit at a greater density of fissile material, than positive feedback criticality; and
  - Silica-moderated "dry" criticality would first require fissile material to be transported by water and deposited before drying out, and water-moderated critical conditions would occur before conditions for dry criticality could be achieved. Water-moderated criticality would require less fissile mass, but at a higher density, than dry criticality.
- The probability of criticality occurring in 10^6 years cannot be dismissed easily (except for explosive assembly, and plutonium criticality away from the repository), and detailed site-specific calculations are required to explore the underlying processes leading up to a self-sustaining nuclear reaction.
- Repository design options that could preclude criticality are:
  - use of neutron poisons;
  - limitation of fissile mass in each container;
  - use of a salt repository;
  - surface storage of spent fuel assemblies;
  - reprocessing and reuse of fissile material in a nuclear reactor or transmutation; and
  - mixing spent fuel with depleted uranium.

Table 2.9 Criticality analysis for a spent fuel repository in Finland (Antilla, 1996)

Spent fuel repository, Finland

Aims of the analysis

Anttila (1996) undertook criticality safety calculations for nuclear waste canisters. The canisters will be disposed of in a repository 300 m to 700 m deep in Finnish crystalline bedrock.

Key features and assumptions

- Spent fuel will be placed in copper/iron canisters for final deep disposal.
- A total of 1.873×10^6 kgU from the TVO reactors (Olkiluoto) will be disposed of in 969 canisters at an average 1,933 kgU/canister, and a total of 7.4×10^5 kgU from the IVO reactors (Loviisa) will be disposed of in 561 canisters at an average 1,319 kgU/canister.

Scenarios assessed

A canister was assumed to be damaged such that water could fill it and result in criticality.

Methodology

The work aimed to find the minimum discharge burnup of fuel bundles at which the criticality safety of disposal canisters is assured according to the safety requirement that the effective multiplication factor, k_{eff}, for canisters is less than 0.95. The reactivity of a canister was calculated using MCNP4A, a neutron-photon-electron transport code for a generalized geometry, based on the Monte Carlo technique. The CASMO-4 and CASMO-HEX fuel assembly burn-up codes were used to calculate the nuclide composition of each fuel rod and the average nuclide composition of a fuel bundle as a function of burn-up.

Results

- Canisters are subcritical when dry, with k_{eff} about 0.22.
- The maximum value of k_{eff} for the canisters is approximately 1 for fresh fuel assemblies (about 3.5% enrichment).
- An increase in burnup of one MWd/kgU decreases the reactivity (k_{eff}) of a canister by 0.006. The criticality safety criteria would be fulfilled if:
  - all bundles in an IVO canister have a burnup higher than 4 MWd/kgU (with an initial enrichment of 3.6% or less); and
  - all bundles in a TVO canister have a burnup higher than 10 MWd/kgU (with an initial enrichment of 3.5% or less).
- The canisters are safe if they have been loaded with fuel that has been in the reactor for at least one normal annual cycle. An increase in enrichment by 0.1 % may be compensated for both fuel types by increasing the minimum discharge burn-up by about one MWd/kgU.

Table 2.10 Criticality analysis for a LLW facility in Utah, US (Hopper and Parks, 1997; Toran et al., 1997)

Envirocare LLW facility, Utah, US

Aims of the analysis

Hopper and Parks (1997) and Toran et al. (1997) assessed the potential for and consequences of criticality resulting from hydrogeochemically concentrated fissile uranium blended with soil in low-level waste disposal facilities. Their analysis was undertaken in the context of the Envirocare LLW facility in Clive, Utah.

Key features and assumptions

- Waste is buried in a 10-m deep trench.
- The soil (waste matrix) has a dry density of 1.6 g cm^{-3} and is either a SiO_2 soil matrix with a minimal neutron absorption capacity or a world average "nominal soil" matrix.
- State of Utah Licence conditions limit the activity of waste to 770 x 10^{-12} Ci of ^{235}U per gram of waste (equivalent to about **0.6 kg ^{235}U m^{-3}** of waste).
- The waste was conservatively assumed to contain 100% enriched ^{235}U.

Scenarios assessed

Under wet conditions, uranium concentrates within the disposal trench after burial by hydrogeochemical processes. Critical conditions are initiated during a drying-out period and continue until terminated because of water evaporation. Key factors in determining the potential for criticality are: the composition of the waste matrix; the enrichment of ^{235}U mass compared with the total uranium mass; the waste matrix density; the density of ^{235}U in the matrix; the degree of neutron moderation in the matrix; the degree of neutron reflection; the geometry or distribution of ^{235}U in the matrix; and the neutronic interaction of one deposit with another deposit.

Methodology

- Simple finite media geometries (spheres) were assumed and their dimensions were calculated for $k_{eff} = 0.95$ with various densities of ^{235}U and water. For infinite media (infinitely-long cylinders and infinite slabs), k_{inf} was calculated. A 2 m-thick neutron reflector was assumed at the surface of each geometry. XSDRNPM, a 1-D discrete ordinate, neutron transport theory deterministic code was used to determine k_{inf} and k_{eff}.
- The density of ^{235}U was assumed to increase for particular geometries according to a concentration factor given by the calculated critical density of ^{235}U for a finite-media geometry divided by the maximum authorized density of ^{235}U allowed for burial. Concentration factors up to 10.5 were considered.
- The consequences of criticality were estimated assuming 10^{17} fissions are required to boil one litre of water and that all water is boiled and evaporated as a result of criticality.

Envirocare LLW facility, Utah, US

Results

- For an average soil with a concentration factor of 10 (0.006 g ^{235}U cm^{-3}), a water content of 0.04 g cm^{-3}, and k_{inf} of 1.3: the infinite slab thickness is 88 cm, with an areal density of 5.3 kg ^{235}U m^{-2}; the infinite cylinder diameter is 165 cm, with a linear density of 12.9 kg ^{235}U m^{-1}; and the sphere diameter is 233 cm, with a mass of 40 kg ^{235}U.
- The critical thickness of the slab and the critical diameters of the cylinder and sphere decrease as the water content increases until the systems become over-moderated.
- The consequences of criticality in a slab geometry of 18-m diameter were estimated. For a concentration factor of 10.5, the critical mass was calculated to contain 4.1 x 10^4 litres of water, which would produce 4.1 x 10^{21} fissions before drying out.
- The fission yields were calculated to result in doses (neutron and gamma) of up to 1.6 Sv for an individual positioned 1 m above the trench.

Additional findings

- Concentration of uranium would be a slow process (taking thousands of years) and this would mitigate rapid approaches to critical or supercritical conditions.
- Reviews of disposal burial records from Envirocare revealed that actual concentrations of ^{235}U are more than a factor of 10 less than maximum permitted concentrations of ^{235}U, and that average enrichment is below the minimum 1% required to achieve nuclear criticality. Thus, for historical operations, uranium concentrations increasing to levels of concern are not expected.
- The potential for uranium concentration could be reduced by: reducing water infiltration; limiting enrichment of ^{235}U; minimizing the potential for reducing conditions (under which uranium precipitates); and limiting the areal density of uranium by limiting the depth of disposal cells.

Table 2.11 Criticality analysis for the Babcock and Wilcox Uranium Processing Facility, US (Alcorn, 1997)

Babcock and Wilcox Uranium Processing Facility, US

Aims of the analysis

Alcorn (1997) considered the nuclear criticality safety of waste drums stored at the Babcock and Wilcox Company Uranium Processing Facility in Virginia, US. The facility processes uranium over the full range of possible enrichments, from depleted to 97.65 wt% ^{235}U.

Key features and assumptions

- The facility generates about 200 55-gal drums of radioactive waste each month. About 2,000 drums are stored on site, along with 64 drums of uranium scrap (from which uranium is recovered).
- Nuclear criticality safety for arrays of 55-gal drums depends on establishing a ^{235}U mass limit for each drum, and measuring accurately or bounding conservatively the mass of ^{235}U in each drum.
- The Babcock and Wilcox base nuclear criticality limit is **350 g ^{235}U per drum.**

Scenarios assessed

Criticality in a stored array of 55-gal waste drums.

Methodology

Alcorn (1997) verified the criticality safety of an array of drums assuming the criticality limit on drums. Calculations were performed using the KENO code, assuming uranium to be present at full enrichment with no credit for neutron poisons. The ^{235}U was assumed to be homogeneously distributed at various concentrations, or as spheres of different degrees of moderation and at different locations in the drums, including at corners adjacent to other drums. The waste drums were assumed to be stacked two high, resting on a concrete floor, fully reflected by water, and with optimum interspersed moderation between the drums.

Results

Calculations showed that a close-packed infinite array of drums, with 350 g ^{235}U per drum, is safe.

Additional findings

- Exceptions to the 350 g ^{235}U limit per drum are:
  - If the mass in the waste drum is uncertain and is based only on a non-destructive analysis (NDA) measuring technique (drum counter) after loading the drum, the nuclear criticality limit is usually reduced to **100 g ^{235}U** maximum per drum to account for uncertainty in NDA measuring techniques and appropriateness of calibration standards.
  - If waste in the drum contains beryllium, the limit is set to **100 g ^{235}U** per drum, which assumes pure U-Be. If the U-Be waste is first packaged into polyethylene bottles of 2.5 l or less the drum limit is **350 g ^{235}U.**
- Old drums of uncertain content are isolated by 1 m and, if necessary, opened and remeasured.
- Currently, the Babcock and Wilcox Company ships waste drums for burial with a drum limit of **100 g ^{235}U.** Few drums require repackaging to meet this limit.

Table 2.12 Criticality analysis for deep-hole disposal of liquid HLW in Russia (Kouzmine et al., 1997)

Deep-hole disposal, Russia

Aims of the analysis

Kouzmine et al. (1997) undertook a criticality evaluation of deep-hole disposal of fissile-containing liquid radioactive waste in Russia.

Key features and assumptions

The rock contains major components of Si, Ca, Al, and Fe in different compositions, corresponding to geologies in which experimental deep-hole disposal of high-level liquid radioactive waste has been undertaken.

Scenarios assessed

Criticality during disposal of radioactive wastes through boreholes in sandstones.

Methodology

- Calculations were made of the criticality potential of the systems that can be formed during deep hole disposal of radioactive wastes. An infinite homogeneous mixture of ^{239}Pu, water and rock components was assumed. The one-dimensional CRAB-1 code was used to calculate k_{inf}.
- Data on the maximum accumulation ability of fissile nuclei from laboratory experiments and deep hole injection tests were used. In the 1950's, large-scale trial experiments were undertaken to dispose of highly radioactive wastes in sandstones at a depth of over 250 m.

Results

- The expected plutonium concentration in a unit stratum volume was determined as a function of the plutonium concentration in a solution, rock density, the distribution factor and the dynamic void fraction of the rock. The expected plutonium concentration was calculated to be less than **2.3 mg ^{239}Pu/l**.
- Measurements showed that the plutonium from borehole injections was uniformly distributed over a volume of rock 20-m thick to a distance 20 - 25 m from each borehole. Criticality calculations showed that a minimum plutonium concentration of **2.84 g ^{239}Pu/l** is required for criticality in such a geometry with an optimum rock-to-water ratio. Therefore, the ^{239}Pu concentration required for criticality is three orders of magnitude greater than the expected ^{239}Pu concentration in rocks resulting from the injection of radioactive waste.

2.3 Summary of Criticality Studies

A wide range of criticality studies related to radioactive waste disposal, storage and reprocessing have been reviewed under this project. The review has included the key studies of the potential for post-closure criticality undertaken as part of radioactive waste disposal programmes. This section summarizes the aims and approaches of the various criticality analyses reviewed, and the different criticality safety criteria used or developed. Criticality assessment methodologies, including criticality scenario development and consequence analyses, are discussed in detail in Chapter 3.

2.3.1 The objectives and approaches of criticality studies

The criticality studies discussed in this chapter had a range of different objectives, but the evaluation of the neutron multiplication factor, k_{eff} (or k_{inf}), for specific configurations of fissile material formed a fundamental part of each study. The criticality studies reviewed had one or more of the following objectives:

- **To demonstrate that a system would remain subcritical**, based on a specific (known, expected, or regulation-limited) fissile material inventory for a particular facility (Berg and Gmal, 1993; Anttila, 1996; and Alcorn, 1997). The analyses generally aimed to demonstrate criticality safety ($k_{eff} < 0.95$) of individual, or arrays of, intact waste containers.

- **To derive criticality-safety criteria** for the acceptance of waste for storage or disposal, such as limits on fissile material inventory, waste container materials, and spent fuel burn-up. The criticality analysis generally involved making a combination of realistic assumptions concerning container or disposal room materials and geometries, and conservative assumptions concerning moderating conditions and the shape and concentration of fissile material. A value of $k_{eff} = 0.95$ was used by Wise et al. (1997) and Anttila (1996) to analyse various waste container configurations. Using $k_{inf} = 1$, Berg and Gmal (1993), uniquely, determined limits on the fissile material contents of different waste containers, assuming the post-closure accumulation of fissile material from several failed containers in a disposal room.

- **To assess the probability of a criticality excursion** based on considerations of the potential for fissile material to accumulate in a critical configuration for a specific criticality scenario. This type of analysis was applied to post-closure criticality assessments, and generally involved conceiving of criticality scenarios (for which $k_{eff} = 1$) and showing that such scenarios can not occur or have a very low probability of occurring (Rechard et al., 1996a; Behrenz and Hannerz, 1978; Johnson et al., 1994; Kastenberg et al., 1996; Rechard et al., 1997; Hopper and Parks, 1997; and Kouzmine et al., 1997). Determinations of the potential for post-closure criticality at various locations in disposal systems were based on considerations of radionuclide inventories (or regulatory limits on fissile material), hydrogeological and hydrogeochemical conditions, and radionuclide migration rates and reconcentration mechanisms. Rechard et al. (1997) uniquely estimated the probabilities of post-closure criticality in a disposal

system, based on considerations of the operational time of the Oklo natural reactor zones.

- **To assess the consequences of a criticality excursion** for specific criticality scenarios. The consequence analyses generally involved determining the fission yield, energy, and/or mechanical damage associated with criticality, and in some cases drew analogies with the Oklo natural reactors and aqueous-type criticality accidents (Rechard et al., 1996a; Behrenz and Hannerz, 1978; Oversby, 1996; Kastenberg et al., 1996; and Rechard et al., 1997). Bowman and Venneri (1996) specified scenarios which could lead to supercritical conditions (i.e., positive-feedback systems with $k_{eff} > 1$) as a result of the accumulation of fissile material in over-moderated systems, and described the potential consequences of such scenarios.

Most of the post-closure criticality studies suggested that criticality is a low-probability event with low consequences in terms of overall repository safety.

2.3.2 Criticality safety criteria

Several of the studies reviewed in this chapter used or derived criticality safety limits on waste package contents. These limits on the fissile material content of waste containers are listed in Table 2.13, and are discussed in the following subsections. To facilitate comparison of limits, Table 2.13 also lists these limits converted to consistent units.

Proposed ILW repository, Sellafield, UK

Plutonium contaminated material (PCM) in decommissioning waste produced at Sellafield is packaged into 200-litre storage drums. A fissile mass limit of 230 g Pu for a 200-litre drum has been determined for arrangements of stacked drums under normal operating conditions (Ogilvie and Harris, 1997). The 200-litre drums are supercompacted and the compact is loaded into 500-litre drums and grouted for storage, with a criticality safety limit of 260 g Pu per 500-litre drum. Other miscellaneous ILW is packaged in storage boxes subject to a plutonium mass limit of 370 g.

Wise et al. (1997) assessed the potential for post-closure criticality at the then proposed repository at Sellafield in the UK. A limit on the fissile mass content of a waste drum containing low levels of fissile material was derived. The safety limit of 50 g ^{239}Pu per 500-litre waste drum was determined on the basis of criticality calculations for a particular drum material (2.5-mm-thick 316L steel) and waste material. Waste producers will be required either to demonstrate compliance with this criticality limit, or to make a criticality safety case for packages containing higher levels of fissile material.

The safety limit may not be appropriate for waste packaged in containers made from other materials or for wastes containing significant quantities of neutron reflectors. Also, Wise et al. (1997) did not consider the potential for criticality as a result of accumulations of fissile material elsewhere in the disposal system. Such considerations may have an impact on the criticality safety limit.

Table 2.13 World-wide limits on the mass of fissile material in a waste drum and the concentration of fissile material in the waste form.

Site	Container type	Container limit	Unit conversion
PCM storage, Sellafield, UK (Ogilvie and Harris, 1997)	200-litre waste drum	230 g Pu	1.15 g Pu/l
PCM storage, Sellafield, UK	500-litre waste drum	260 g Pu	0.52 g Pu/l
ILW repository, Sellfield, UK (Wise et al., 1997)	500-litre waste drum	50 g ^{239}Pu	0.1 g ^{239}Pu/l
WIPP transuranic-waste repository, New Mexico, US (USDOE, 1991)	208-litre waste drum	200 g ^{239}Pu FGE[1]	0.96 g ^{239}Pu FGE/l
	1,800-litre waste box	350 g ^{239}Pu FGE	0.2 g ^{239}Pu FGE/l
Konrad repository, Germany (Berg and Gmal, 1993)	700-litre waste package containing one 200-litre waste drum	120 g ^{235}U (LE[2])	0.6 g ^{235}U (LE)/l
		50 g ^{235}U (HE)	0.25 g ^{235}U (HE)/l
		28 g ^{239}Pu	0.14 g ^{239}Pu/l
	5,400-litre waste package containing ten 200-litre waste drums	425 g ^{235}U (LE)	0.21 g ^{235}U (LE)/l
		175 g ^{235}U (HE)	0.09 g ^{235}U (HE)/l
		110 g ^{239}Pu	0.06 g ^{239}Pu/l
	Waste directly conditioned into a 10,900-litre waste package	850 g ^{235}U (LE)	0.08 g ^{235}U (LE)/l
		350 g ^{235}U (HE)	0.03 g ^{235}U (HE)/l
		220 g ^{239}Pu	0.02 g ^{239}Pu/l
Babcock and Wilcox processing facility, US (Alcorn, 1997)	208-litre waste drum (storage)	350 g ^{235}U	1.7 g ^{235}U/l
	208-litre waste drum (storage when contents are uncertain or waste contains beryllium)	100 g ^{235}U	0.5 g ^{235}U/l
	208-litre waste drum (for disposal)	100 g ^{235}U	0.5 g ^{235}U/l

Site	Concentration limit	Unit conversion
LLW Drigg, UK (see Section 4.1.1)	0.1 GBq per tonne in total of ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{242}Pu	0.000044 g ^{239}Pu/l
Konrad repository, Germany (Berg and Gmal, 1993)	50 g FM[3] per 100 litres of the waste form	0.5 g FM/l
Envirocare LLW facility, US (Hopper and Parks, 1997)	0.6 kg ^{235}U/m^3 of waste form	0.6 g ^{235}U/l

[1] FGE = Fissile Gram Equivalent.
[2] LE = Low Enrichment; HE = High Enrichment.
[3] FM = Fissionable Material

WIPP ILW repository, US

Criticality safety criteria have been defined as part of the Waste Acceptance Criteria (WAC) for the WIPP (USDOE, 1991), although these criteria were not used directly by Rechard et al. (1996a) in their post-closure criticality analysis for the WIPP. The WIPP WAC include requirements that the fissionable radionuclide content of a 0.21-m^3 (208-litre) drum is less than 200 g ^{239}Pu fissile-gram equivalent (FGE), and the fissionable radionuclide content of a 1.8-m^3 (1,800-litre) waste box is less than 350 g ^{239}Pu FGE.

Konrad ILW Repository, Germany

Berg and Gmal (1993) demonstrated criticality safety for a regulation-limited fissile material inventory at the Konrad repository in Germany. Criticality safety during waste handling operations require the concentration of fissile material in a waste package to be less than 50 g per 100 litres of the waste form.

Berg and Gmal (1993) also calculated admissible fissile masses for various types of waste container destined for the Konrad repository. Some of these waste containers will be packaged with 200-litre waste drums, such that considerable waste-free volume will exist in the containers. For example, a 700-litre container holds only one 200-litre drum, so that the container criticality safety limit of 120 g ^{235}U correlates to a fissile material concentration of 0.6 g ^{235}U/litre in the drum. Such a concentration exceeds the waste handling and emplacement mass concentration limit (0.5 g/litre). Similarly, the container criticality safety limit of 128 g ^{239}Pu in a 700-litre container correlates to a fissile material concentration of 0.14 g ^{239}Pu/litre in a 200-litre drum.

Spent fuel repository, Finland

Anttila (1996) calculated the criticality safety of different Finnish spent fuel containers as a function of burn-up, rather than directly as a function of fissile material content of waste containers. An increase in burnup of one MWd/kgU was shown to decrease the reactivity (k_{eff}) of a canister by 0.006. This type of limit is not relevant to ILW disposal requirements and is discussed no further in this report.

LLW repository, Utah, US

Hopper and Parks (1997) assessed the potential for nuclear criticality based on a regulatory limit of 0.6 kg ^{235}U/m^3 of waste for a LLW disposal facility in the US. The disposal concept involved waste buried in a 10-m deep trench rather than in waste containers.

Uranium processing facility, Virginia, US

Alcorn (1997) demonstrated criticality safety for a regulation-limited fissile material inventory at the Babcock and Wilcox uranium processing facility in the US. The Babcock and Wilcox base nuclear criticality limit is 350 g ^{235}U per 55-gal waste drum.

2.3.3 Discussion

The criticality studies undertaken by Wise et al. (1997) and Berg and Gmal (1993) show the importance of considering the geometric and material properties of waste containers, and their distribution in a repository, in deriving fissile material limits for waste packages. In particular, the analysis carried out by Berg and Gmal (1993) for the Konrad repository showed that there may be significant variation in fissile content limits for different types and volumes of waste container stacked in a disposal room. The allowable concentration of fissile material in a 200-litre drum (within a 700-litre waste package) is nearly an order of magnitude greater than the allowable concentration of fissile material in waste conditioned directly into a 10,900-litre waste package.

Berg and Gmal (1993) also showed that considerations of post-closure criticality can lead to more restrictive limits on the fissile material content of waste containers than waste handling and storage limits. The limits for waste packages at the Konrad repository were determined based on a conservative assumption that all the fissile material in waste packages in a cross-section of the disposal room is selectively leached out and accumulates in a hole at the bottom of the waste room. In most cases the calculated waste package limits were found to be more restrictive than the waste handling and emplacement mass concentration limit. More realistic assumptions concerning the potential for radionuclide migration and reconcentration in a disposal room would, most likely, lead to less restrictive controls on waste container contents.

3. ASSESSING POST-CLOSURE CRITICALITY

The criticality studies reviewed in Chapter 2 emphasised the need for post-closure criticality to be considered as part of a repository safety case. This chapter discusses the techniques that may be used to assess post-closure criticality. These techniques usually involve defining several criticality scenarios, and may involve assessing the potential consequences of post-closure criticality. Criticality scenarios and consequences are described in this chapter. Finally, a number of controls are discussed, which could be imposed to mitigate the likelihood or consequences of post-closure criticality.

3.1 Post-Closure Criticality Assessment Methods

The criticality studies reviewed in Chapter 2 aimed to:

- demonstrate that a system would remain subcritical;
- derive criticality-safety criteria;
- assess the probability of a criticality excursion; and/or
- assess the consequences of a criticality excursion.

In general, the studies were based on deterministic approaches, which involved criticality analyses for specific waste storage, disposal room, near-field and/or far-field configurations. For example, Berg and Gmal (1993) determined criticality limits for waste packages at the Konrad repository, in Germany, assuming that the contents of many waste containers could concentrate in a disposal room after closure. However, in response to Bowman and Venneri's (1996) study of underground supercriticality at the proposed HLW repository at Yucca Mountain, several researchers concluded that the issue of post-closure nuclear criticality should be addressed though a risk-based analysis of possible criticality scenarios (e.g., Van Konynenburg, 1996; Scott and Doering, 1997). These different approaches to assessing post-closure criticality are discussed in the following subsections.

3.1.1 Deterministic approaches

Several of the repository post-closure studies reviewed in Chapter 2 aimed to show that specific criticality scenarios were unlikely for the facilities in question, because the necessary concentrations, masses, and geometries of fissile material required for criticality could not form. Such an approach requires knowledge of how radionuclides migrate and accumulate in the disposal system. This information can be derived in a semi-qualitative sense if pre-determined values of key parameters, such as solubilities, advection and diffusion rates, precipitation rates, compaction rates, and oxidation states, are available from previous studies and experiments.

This type of deterministic approach was adopted by Rechard et al. (1996a) in their analysis of the potential for post-closure criticality at the planned WIPP repository. The masses and concentrations of ^{239}Pu and ^{235}U required to achieve criticality in the near-field and the far-field of the disposal system were calculated. Rechard et al. (1996a) then made use of available performance assessment (PA) calculations that predicted the migration of radionuclides in the disposal system. In most cases they were able to show that insufficient fissile material could accumulate for a critical mass to form. Otherwise they showed that either the porosity, or the radionuclide reconcentration potential, was insufficient for criticality to occur at locations of concern. Similar approaches were adopted by Behrenz and Hannerz (1978) and Johnson et al. (1994). Others took more qualitative approaches, based on, for example, estimates of radionuclide dissolution, advection, diffusion and precipitation rates, to conclude that conjectured critical conditions could not be achieved.

One potential problem with the use of existing PA calculations in deriving the key factors used in subsequent criticality analyses, is that modelling assumptions may have been made that are not conservative with respect to the potential for nuclear criticality. For example, performance assessments may have included assumptions that minimize radionuclide retardation in a disposal system in order to make conservative calculations of dose to a critical group. Evaluations of the potential for nuclear criticality would, more likely, make assumptions that maximize radionuclide retardation in parts of the disposal system in order to make conservative estimates of the potential for reconcentration of fissile material. Thus, it may be appropriate to perform radionuclide migration calculations specifically for nuclear criticality assessments.

3.1.2 Risk-based approaches

The methodology currently adopted to address criticality concerns at the Yucca Mountain repository involves separate consideration of pre-closure and post-closure periods (Doering et al., 1997). Criticality safety analyses are being undertaken for the time that humans would be in the immediate vicinity of the waste. This involves calculating k_{eff}, taking account of bias and uncertainty, for waste handling and emplacement designs, and showing criticality safety to a required safety margin (see Section 3.1.4). However, risk analysis calculations are being performed for the post-closure period, aimed at identifying the long-term risk of criticality excursions to future generations.

Recently, Sanchez et al. (1998) developed criticality scenarios by using probabilistic methods to determine the probability of the events that lead to potentially critical configurations. The criticality potential of the configurations identified to be of concern were evaluated using deterministic methods, involving comparison with the subcritical limit of k_{eff}, and consequence analyses were performed for the configurations that exceeded the criticality limit. The frequency of nuclear excursions and their impact on the repository radionuclide inventory, their thermal effects, and their effects on groundwater flow were determined. Risk calculations were undertaken that estimated fission-yield products generated by possible future criticality excursions. The results could be used in the overall Yucca Mountain repository performance assessment. Such an approach requires the use of a suite of PA codes and significant computational resources, but the method could be simplified by modelling only the processes directly related to criticality (i.e., the factors listed for the scenario described in Section 3.2).

3.1.3 Neutron transport codes

Evaluation of the neutron multiplication factor, k_{eff}, for specific configurations of fissile material is a fundamental part of any pre-closure or post-closure criticality analysis. Various neutron transport codes were used to calculate k_{eff} in the studies reviewed (see Chapter 2). In most cases, a Monte Carlo neutronics computer code, and associated cross-section library, was used to determine the neutron multiplication factor. Wise et al. (1997) used the MONK6B code for the Sellafield repository criticality analysis. The MONK code is used throughout the UK nuclear industry to assess criticality safety, and Smith and Thorne (1997) discussed the recent development of MONK7. Outside the UK, particularly in the US (Cabrilla, 1997), the MCNP, KENO and VIM Monte Carlo codes are more commonly used.

In three of the studies reviewed, one-dimensional deterministic neutron transport codes were used. Also, a deterministic neutronics code, WIMS, is frequently used in the UK nuclear industry for criticality safety analysis as an alternative to the MONK Monte Carlo code (e.g. Farrington and Willock, 1997). Nouri and Smith (1997) presented results of an ongoing neutronics code inter-comparison exercise, which includes the application of both Monte Carlo codes (including MONK) and deterministic codes (including WIMS), and reported good agreement between the various codes used in the exercise.

Any quantitative evaluation of post-closure criticality in the context of setting fissile material limits on waste packages would require calculations using a neutron transport computer code.

3.1.4 Neutron multiplication factor safety margins

Calculations of k_{eff} in the studies reviewed in Chapter 2 assumed different margins of safety depending on the objectives of the study. Demonstrations of criticality safety and calculations of criticality safety limits generally required k_{eff} to be less than 0.95, whereas post-closure criticality analyses typically assessed systems with k_{eff}= 1.0.

Regulations concerning criticality safety during storage and transportation of nuclear waste and during repository operations are essentially deterministic. Usually, an arbitrary subcritical margin of safety is introduced for a criticality safety analysis that demonstrates that criticality will not occur for a particular system. The US Nuclear Regulatory Commission has generally required the maximum value of k_{eff} to be less than 0.95 for handling, storage and transportation of radioactive material, provided that the statistical and modelling uncertainty associated with the calculation of k_{eff}, and the bias and uncertainty associated with benchmark experiments, are taken into account (Bhatnagar, 1997; Wright and Thomas, 1997). This implies that a minimum subcritical margin of 0.05 must be applied to licensing criticality calculations in the US. However, Bhatnagar (1997) noted a case in which the US Department of Energy accepted a value of 0.98 for k_{eff} (Hanford), and other US nuclear installations (Paducah and Savannah River) have used a value of 0.9596 as an upper limit.

Currently, no subcritical margins have been defined that apply to the long-term disposal of radioactive waste in deep geologic repositories in the US (Wright and Thomas, 1997). Proposed regulations applicable to disposal of HLW at Yucca Mountain (proposed rule 10

CFR 63) require the probability and consequences of criticality to be examined and, potentially, the effects of criticality to be included in a risk-based post-closure performance assessment. Typically, a probabilistic analysis does not employ an arbitrary additional conservatism to the end result, but accounts for biases and uncertainties in the probabilities and consequences of events. Thus, introduction of an arbitrary margin of safety is unlikely to be necessary in analyses aimed at evaluating the probability and consequences of post-closure criticality. Scott and Doering (1997) concluded that post-closure criticality analysis should account for bias and uncertainty and should be based on a value of $k_{eff} = 1$.

3.2 Post-Closure Criticality Scenarios

As highlighted in the criticality review reported in Chapter 2, different sequences and combinations of factors could result in critical concentrations of fissile radionuclides at different times and at different locations within a disposal system. The different types of criticality scenarios that can be envisaged primarily depend on the waste type and disposal concept under consideration. This chapter focuses on the features, events and processes that could influence long-term criticality safety and might need to be considered in a post-closure safety case for a deep LLW/ILW repository.

Repository designs for the disposal of radioactive waste generally involve multi-barrier containment systems, comprising engineered and natural components. ILW disposal systems typically require that ^{239}Pu- and ^{235}U-contaminated waste is packaged in containers (possibly in grout), which are emplaced in the tunnels and vaults of the repository. The disposal regions are backfilled (in most designs) and sealed before the repository is closed. For such a disposal concept, criticality scenarios may be defined based on the integrity of the waste containers and the location of the envisaged critical excursion:

Scenario 1: criticality in an intact waste container;

Scenario 2: criticality in a leaking waste container;

Scenario 3: criticality in one or more collapsed waste containers;

Scenario 4: criticality in the near-field (seals, shafts and excavation damage zones, and backfilled disposal rooms, boreholes, and tunnels); and

Scenario 5: criticality in the far-field (rock matrix and fractures).

These scenarios represent those typically examined in post-closure criticality assessments, but the specification of scenarios is not unique; for example, scenarios could be defined in terms of radionuclide concentration mechanisms. Also, further sub-division of scenarios may be convenient for quantitative analysis based on, for example, the location of the critical excursion in the near-field. Whichever approach to scenario development is taken, it is important that all relevant features events and processes are taken into account.

A criticality incident could involve either ^{239}Pu or ^{235}U, but criticality is not possible after the fissile material has decayed significantly[2]. The key factors that influence the likelihood and possible consequences of criticality are provided in the following subsections for each of the five scenarios listed above. These factors have been derived from the various criticality scenarios considered in the studies reviewed in Chapter 2. The potential for each criticality scenario could be controlled by setting constraints on appropriate factors (see Section 3.4.2).

3.2.1 Scenario 1: criticality in an intact waste container

While the containers remain intact, the potential for criticality depends primarily on the mass and geometry of fissile material and the amount of any neutron moderators, reflectors and poisons in each container. Criticality may occur in a single container or as a result of neutron interaction between two or more neighbouring containers stacked in a disposal room. The key factors affecting the potential for and consequences of criticality in an intact waste container, as illustrated in Figure 3.1, are:

- the mass of fissile material in each container (which will change as a result of decay and ingrowth);
- the initial location and distribution of fissile material in each container;
- the formation of a critical mass of fissile material as a concentrated solution (which depends on radionuclide solubilities, the liquid content of the waste, and waste porosity), as a result of adsorption on mineral surfaces in the container (e.g., by ion exchange or surface complexation), as a result of precipitation in the container; or as a result of generation, destabilization and precipitation of colloids;
- the amount of neutron moderators (e.g., water, polyethylene), reflectors (e.g., beryllium, graphite, grout) and poisons (e.g., boron, ^{238}U) in the waste form;
- the neutron absorption capacity and reflecting properties of the container, which depend on the container wall material (e.g., the type of steel), thickness, and dimensions;
- the geometry of the disposal rooms and the distribution of waste containers;
- the parasitic neutron absorption capacity and reflecting properties of the material between the waste containers (e.g., air, water, backfill); and
- the leaching of backfill (such as $Ca(OH)_2$) and the associated reduction in neutron-absorbing hydrogen in the backfill as a results of fluid flow in the disposal area.

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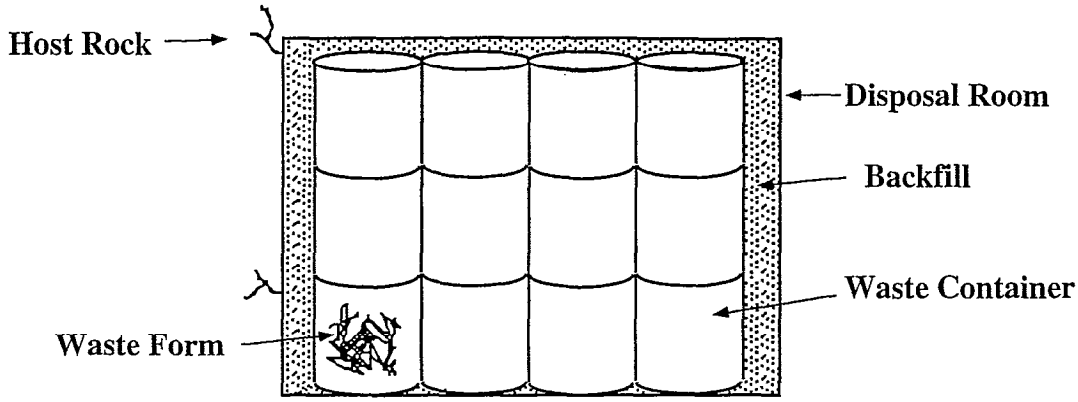
[2]The half life of ^{239}Pu is 2.41×10^4 years and the half life of ^{235}U (and plutonium degraded to uranium) is 7.04×10^8 years.



	Key Factors	Related Criticality Controls
Scenario 1	**Waste Form**	
	Fissile mass	Limit fissile mass and concentration
	Neutron moderation	Limit amount of neutron moderators
	Neutron reflection	Limit amount of neutron reflectors
	Neutron absorption	Add neutron poisons/depleted uranium
	Distribution of materials	Distribute materials uniformly
	Reconcentration of fissile material	Compact waste to reduce porosity
	- in a concentrated solution	Control oxidation states
	- by adsorption	-
	- by precipitation	Control oxidation states
	Decay and ingrowth	-
	Waste Container	
	Neutron absorption/reflection	Change wall thickness/material specification
	Disposal Room	
	Container distribution	Reduce density of containers
	Backfill	
	Neutron absorption/reflection	Add neutron poisons
	Leaching of backfill	-
	Host Rock	
	Fluid flow conditions	-
Scenario 2	**Waste Form**	
	Reconcentration of fissile material	
	- by colloid filtration	-
	Waste Container	
	Container failure	Change material specification
	Separation/removal of poisons	-
	Water entry	-

Figure 3.1 Scenario 1: criticality in an intact waste container and Scenario 2: criticality in a leaking waste container. The factors affecting the potential for criticality are listed for each component of the disposal room. Possible criticality controls related to these factors are also shown.

3.2.2 Scenario 2: criticality in a leaking waste container

A container may be vented, may have a material defect at the time of emplacement, or may fail such that an opening occurs in the container wall but the container maintains its structure at its original location. Criticality may occur in the container depending on the different rates of removal of neutron poisons and fissile material from the container, and the rate of flow of water into the container. This scenario depends on the factors involved in Scenario 1 (see Figure 3.1) and:

- the time of container failure, which depends on the initial integrity of the container, the container corrosion rate (if failure is by corrosion), or the timing of seismic/tectonic events (if mechanical failure occurs);

- the rate of separation of neutron-absorbing poisons from the fissile material in the waste by chemical processes (involving differential dissolution rates);

- the rates of diffusion and advection of poisons from the container into the surrounding materials and fractures in the host rock, which depend on the hydraulic properties of the various components involved, and may change with time as a result of, for example, backfill leaching;

- the rates of diffusion and advection of fissile material from the container into the surrounding materials and fractures in the host rock;

- the rate of entry of water (neutron moderator) into the failed container; and

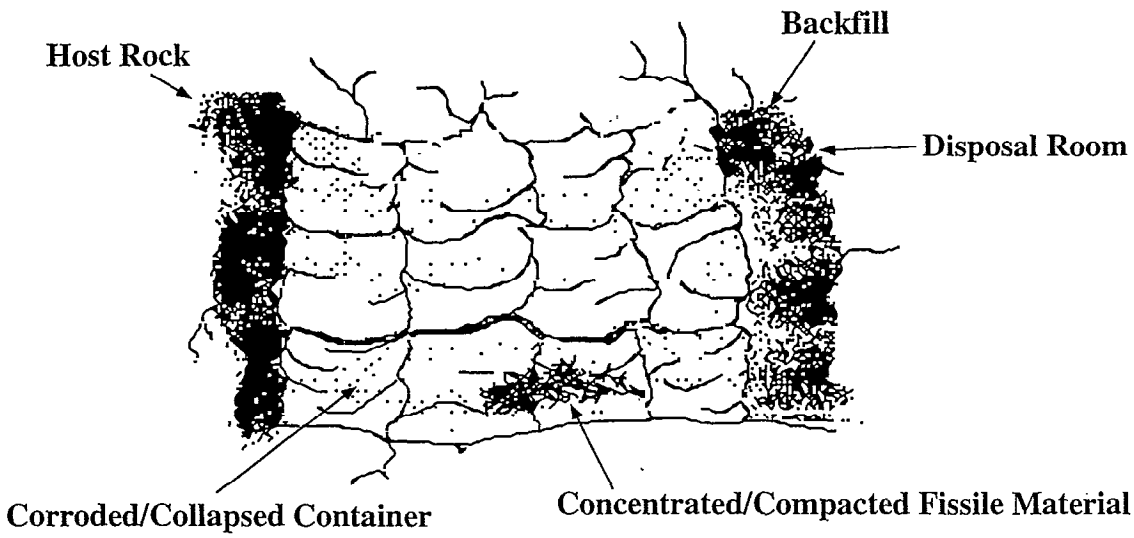
- the extent of concentration of fissile material as a result of filtration of colloidal material (as well as by the concentration processes described for Scenario 1).

3.2.3 Scenario 3: criticality in one or more collapsed waste containers

Large-scale container failure and collapse may occur as a result of container weakening by corrosion and/or stress loading. Extensive corrosion of containers could result in a slump of waste materials, container corrosion products, and backfill material in a critical geometry. Compaction of containers could result in the formation of a critical concentration of fissile material under moderated or unmoderated conditions (see Section 3.3.2). This criticality scenario is illustrated in Figure 3.2, and depends on the factors involved in Scenarios 1 and 2, which relate to the canister, waste, and backfill conditions, as well as:

- the time of corrosion-induced collapse of the container and/or the time and magnitude of seismic or tectonic events; and

- the extent of concentration of fissile material as a result of container collapse and/or compaction.



	Key Factors	Related Criticality Controls
Scenario 3	**Waste Form** Reconcentration of fissile material - by collapse/compaction	Compact waste to increase strength
	Waste Container Time of collapse	Change material specification

Figure 3.2 Scenario 3: criticality in one or more collapsed waste containers. The factors affecting the potential for criticality, in addition to those shown for Scenarios 1 and 2, are listed for different components of the disposal room. Possible criticality controls related to these factors are also shown.

3.2.4 Scenario 4: criticality in the near-field

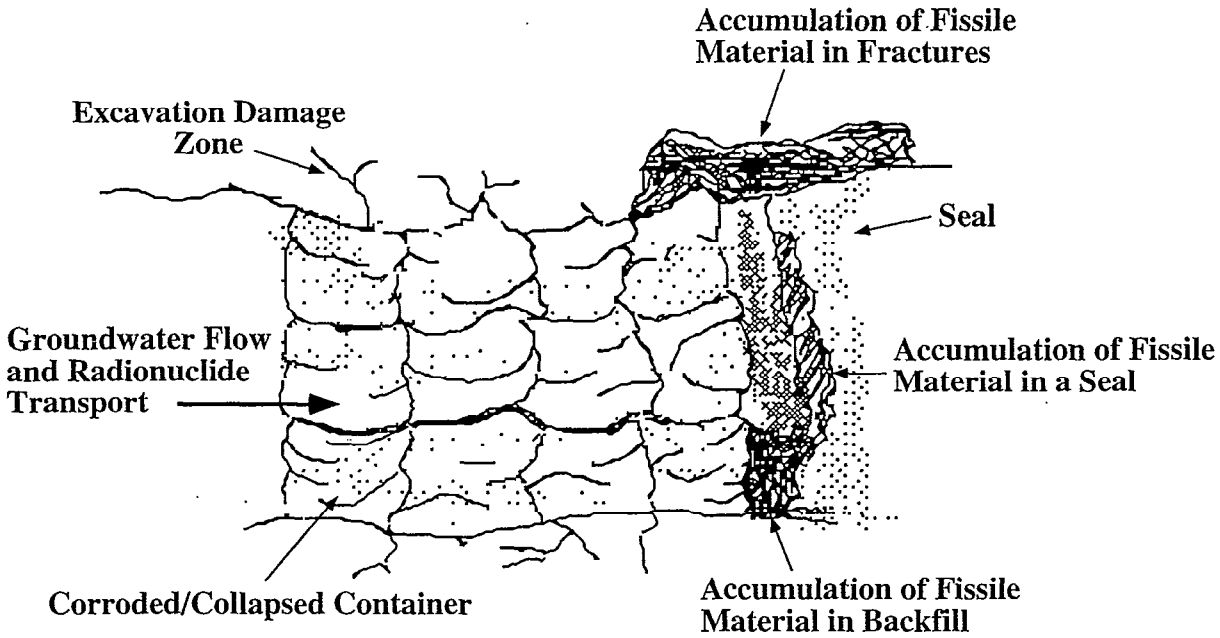
Criticality could result from the accumulation of fissile material from one or more failed containers in a critical geometry in the near-field. This scenario involves leaching of waste material from failed containers and the subsequent redeposition of fissile material in a critical geometry in, for example, the backfill, seals or excavation damage zone. Near-field criticality is illustrated in Figure 3.3. Near-field criticality depends on the factors listed for Scenarios 1, 2, and 3, which relate to the container, waste, and backfill conditions, and:

- the rate of diffusion and advection of fissile material in the near-field;
- the extent of preferential re-concentration of fissile material from a transported mixture into a critical mass as a concentrated solution, or as a result of adsorption, precipitation (which requires a change from oxidizing conditions to reducing conditions), or filtration of colloidal material; and
- the amount of neutron moderating, absorbing, and reflecting material at the site of fissile material accumulation.
- Sufficient porosity for fissile material to concentrate in a critical configuration.

3.2.5 Scenario 5: criticality in the far-field

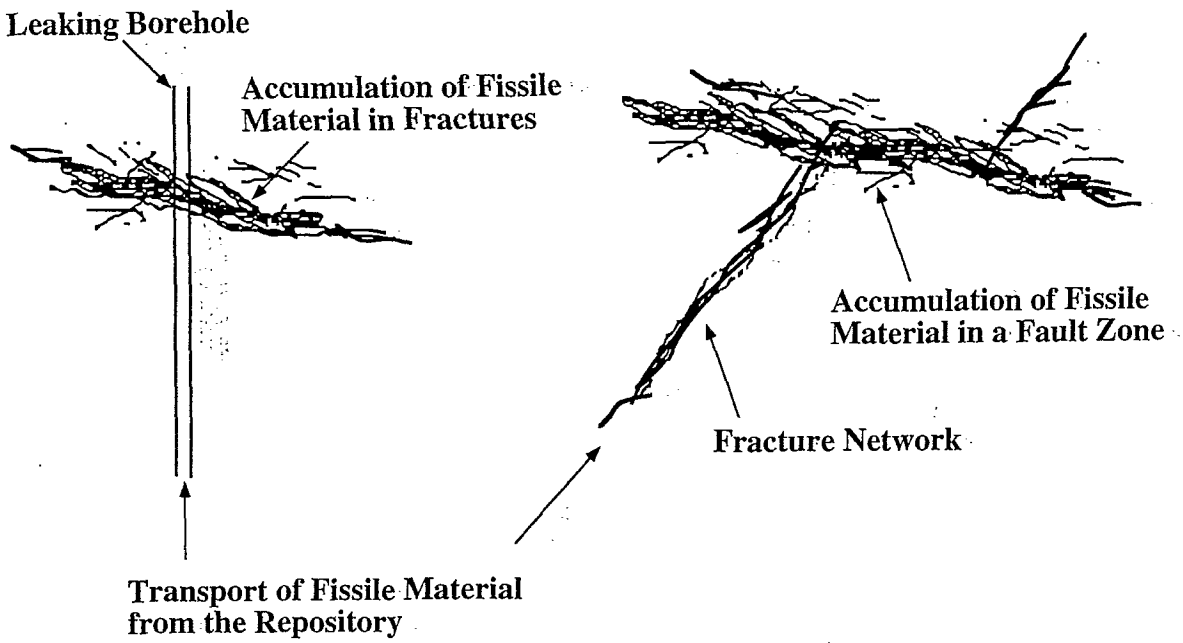
Criticality could result from the failure of one or more containers followed by dissolution and redeposition of fissile material in the far-field (host-rock or other units). A key requirement of this type of criticality is the accumulation of the fissile material at a distant location. Far-field criticality is illustrated in Figure 3.4, and depends on the factors listed for Scenarios 1, 2, and 3, which relate to the canister, waste, and backfill conditions, and:

- the existence of flow paths (e.g., rock matrix permeability, fractures, and boreholes) from the repository to distant locations;
- the rate of diffusion and advection of fissile material (preferably from several containers) along the flow paths;
- the extent of preferential re-concentration of fissile material from the transported mixture as a concentrated solution, or as a result of adsorption, precipitation (which requires a change from oxidizing conditions to reducing conditions), or the filtration of colloidal material;
- the amount of neutron moderating and absorbing material at the site of fissile material accumulation; and
- the existence of sufficient matrix porosity or fracture void space for fissile material to concentrate in a critical configuration.



	Key Factors	Related Criticality Controls
Scenario 4	**Near-Field**	
	Transport of fissile material	-
	Preferential reconcentration	
	- in a concentrated solution	Control oxidation states
	- by adsorption	-
	- by precipitation	Control oxidation states
	- by colloid filtration	Use compacted backfill to immobilize colloids
	Neutron moderation	-
	Neutron absorption	-
	Neutron reflection	-
	Porosity	Compact backfill to reduce porosity

Figure 3.3 Scenario 4: criticality in the near-field. The factors affecting the potential for near-field criticality, in addition to those shown for Scenarios 1, 2 and 3, are listed. Possible criticality controls related to these factors are also shown.



	Key Factors	Related Criticality Controls
Scenario 5	**Far-Field**	
	Flow paths	-
	Transport of fissile material	-
	Preferential reconcentration	
	- in a concentrated solution	-
	- by adsorption	-
	- by precipitation	-
	- by colloid filtration	-
	Neutron moderation	-
	Neutron absorption	-
	Porosity	-

Figure 3.4 Scenario 5: criticality in the far-field. The factors affecting the potential for far-field criticality, in addition to those shown for Scenarios 1, 2, 3 and 4, are listed. Migration of fissile material via a borehole and via a fracture network is illustrated.

3.3 The Consequences of Post-Closure Criticality

This section describes the potential consequences of the post-closure criticality scenarios outlined in Section 3.2. When fission occurs in a fissile mass, whether it is caused by uranium or plutonium, the event results in the production of energy, fission products, neutrons, and various types of radiation (gamma, beta, etc.). The consequences of a critical event in a repository include increased radiological hazard and mechanical damage caused by kinetic or heat energy. The extent to which the increased radionuclide inventory would affect the release of radioactive materials from the repository is of greatest concern. Several repository studies have made estimates of the possible consequences of criticality, including considerations of the fission yield, the induced thermal load, and material damage. In particular, Rechard et al. (1997) considered the possible consequences of slow, fast and explosive rates of critical assembly under unmoderated and moderated conditions.

3.3.1 Slow assembly of a critical mass

A moderated slow rate of assembly of fissile material is represented by the natural reactors in uranium ore deposits at Oklo in Gabon, West Africa. In 1972, samples of uranium ore from a mine at Oklo were found to have $^{235}U/^{238}U$ ratios significantly less than the present day global ratio (Oversby, 1996; Wickham and Bennett, 1997). The depletions in ^{235}U have been attributed to past nuclear fission of ^{235}U under natural conditions. Based on natural processes and repository conditions, moderated criticality at a slow rate of assembly represents the type of excursion most likely to occur in a disposal system. The unmoderated slow assembly of a critical mass has similarities to the conditions that are created in a faster breeder reactor. However, slow assembly of fissile material without water is not credible in a repository under natural geologic conditions.

The uranium ore deposits at Oklo derived from erosion of the Archaean crystalline basement and deposition of sediments as sandstone and conglomerate units. Ore concentration was achieved by dissolution in circulating oxidizing fluids that encountered reducing conditions in overlying black shales. Uranium precipitation occurred in a narrow sandstone layer (a few metres thick) below the base of the black shales. Mining in a region where the uranium ore bed intersects the ground surface has led to the discovery of nine "reactor zones" which are depleted in ^{235}U. Another six reactor zones have been found at deeper locations in the Oklo region, and one has been found in a shallow location 30 km east of Oklo. The reactor zones are typically lenticular in shape, 10 to 50 cm thick, with average lengths and widths of 10 m. The uranium ore at Oklo has typical concentrations of 0.2-1% UO_2, but where the reactor zones are found the ore is rich in uraninite with concentrations of 20-60% UO_2.

Based on considerations of the geometry and uranium inventory of the Oklo reactors, Oversby (1996) determined that uranium criticality could not occur in a canister in a Swedish spent fuel repository (see Table 2.4). Furthermore, Oversby (1996) estimated that the uranium from more than 71 canisters would be required to achieve an Oklo-type criticality in a repository tunnel.

Rechard et al. (1997) estimated the probability of uranium criticality at the proposed Yucca Mountain repository based on a consideration of the conditions required for the formation of the Oklo reactors. About 133 Mg of uranium were estimated to have been associated with each Oklo reactor zone, and each reactor was estimated to have had a minimum operating life of about 2×10^5 years (see Table 2.8).

Other researchers assessed the potential thermal effects of an Oklo-like criticality. The amount of heat generated in the Oklo natural reactors has been estimated as 0.1 kW/m^3 (see Table 2.2). By comparison, Rechard et al. (1996a) estimated that an Oklo-like criticality excursion in the far-field at the WIPP could generate a thermal power of 3.7 kW. Rechard et al. (1997) estimated the thermal power from criticality at the Yucca Mountain repository would be between 7 and 21 kW (Table 2.8). These criticality-induced thermal loads are a small percentage of the expected initial thermal load of the repositories.

3.3.2 Fast assembly of a critical mass

A moderated fast rate of assembly corresponds to the critical aqueous-type accidents that have occurred in, for example, nuclear fuel processing plants (^{235}U and ^{239}Pu aqueous solutions). The yield from such accidents is often limited by the disassembly of the system. Rechard et al. (1996b) provided a list of such excursions, which have generated fewer than 10^{20} total fissions (1.25×10^9 J), with an average of about 10^{18} fissions (1.87×10^7 J). This type of criticality excursion could result from a human intrusion into a repository that generated a slurry of fissile material and water.

Unmoderated (e.g., involving little water) fast rates of assembly could occur as a result of the sudden collapse of a waste container. Accidents and experiments involving unmoderated fast rates of assembly have yielded between 10^{16} and 10^{17} fissions. Yields of between 10^{18} and 10^{19} fissions have been calculated for a hypothetical criticality accident in a waste supercompactor. These prompt fission yields are comparable to the number of prompt fissions that have been produced in aqueous accidents (Rechard et al., 1996b).

Prompt radiation exposure (mainly neutron and gamma) for a typical aqueous excursion, generating approximately 10^{17} prompt fissions, would cause human fatalities at distances of less than 3 m from an unshielded critical assembly. The spatial dependency of the exposure on distance is due mainly to the geometric spread of the radiation. Since there would be ample shielding between post-closure fissile material in an underground geologic repository and the accessible environment, prompt exposures to humans are not of concern. Hopper and Parks (1997) estimated the potential consequences of criticality in a shallow LLW disposal facility (a 10-m deep trench), as a result of the concentration of fissile material (see Table 2.10). A moderated critical mass was calculated to contain 4.1×10^4 litres of water, which would produce 4.1×10^{21} fissions before drying out. The fission yield was calculated to result in doses (neutron and gamma) of up to 1.6 Sv for an individual positioned 1 m above the trench[3].

[3] For the purposes of discharge authorization in the UK, a dose limit of 1 mSv/y to members of the public from all man-made sources of radioactivity (other than medical applications) is applied (Environment Agency et al., 1997).

The fission yield associated with the moderated or unmoderated fast rates of assembly (10^{15} to 10^{20} fissions by analogy with aqueous excursions) is likely to be negligible compared to the number of fissions represented by a repository. For example, Rechard et al. (1996a) estimated that the WIPP transuranic waste repository would be represented by 10^{27} fissions (Table 2.2), and Rechard et al. (1997) estimated that the Yucca Mountain HLW repository represented 10^{30} fissions (Table 2.8).

The small amount of rapidly produced energy from sudden assembly of fissile material below the surface is unlikely to cause significant damage. For example, Rechard et al. (1996a) estimated that voids of no more than 1.3 m radius would occur in the salt host-rock of the WIPP repository (Table 2.2), and voids of less than 0.8 m radius would be formed in the Culebra unit overlying the repository host rock.

3.3.3 Assembly of a supercritical mass

Bowman and Venneri (1996) suggested that underground supercriticality could occur and that the resulting prompt kinetic yield could contribute to dose if the gas produced reached the atmosphere (see Table 2.6). However, Bowman's and Venneri's (1996) analysis was much criticized for having adopted hypothetical, idealized configurations. For example, the study assumed homogeneous rather than heterogeneous mixtures, pure SiO_2 rock rather than rock with a realistic distribution of elements (including neutron absorbers), and concentration of ^{239}Pu but not even isotopes such as ^{240}Pu. Also, the study took no account of whether or not the distributions of fissile material required for autocatalytic criticality could be achieved.

Low moderation with explosive assembly occurs in a nuclear weapon, and requires rapid creation of a fissile assembly under conditions that cannot be achieved in a repository under natural geologic conditions.

3.4 Specifying Criticality Controls

3.4.1 Methodologies

Deterministic or risk-based post-closure criticality analyses of the type described in Section 3.1 could be undertaken to determine regulatory requirements concerning criticality. Also, waste disposers may want to determine measures that could be taken to reduce the potential for criticality based on post-closure criticality assessments. However, such criticality controls are likely to have an impact on other aspects of repository performance. Consideration should be given to the implications of criticality controls in the context of overall disposal system behaviour.

Although several projects have undertaken analyses to set criticality safety limits for repositories, considerations have been restricted to criticality in the waste containers and/or the near-field for assumed concentrations, geometries, masses, etc., of fissile material. None of the studies reviewed in Chapter 2 set waste acceptance limits based on considerations of far-field criticality (i.e., Scenario 5 as described in Section 3.2.5). However, Berg and Gmal (1993)

(see Table 2.3) determined the limits for waste packages at the Konrad repository, in Germany, based on a conservative assumption that all the fissile material in waste packages in a cross-section of a disposal room is selectively leached out and accumulates in a hole at the bottom of the waste room (Scenario 4 as described in Section 3.2.4). Their analysis showed that considerations of post-closure criticality can lead to more restrictive limits on the fissile material content of waste containers than waste handling and storage limits.

Deterministic methods have been used to show that post-closure criticality is unlikely to occur in the far-field of specific disposal systems. However, if significant post-closure criticality concerns remain, then such techniques could be extended to set criticality controls. That is, analysis could be undertaken to calculate the rate of accumulation of fissile material at specific locations in the disposal system. If a critical configuration was found to be achievable based on reasonable modelling and data assumptions, considering uncertainties, then a limit on the fissile content of waste packages (or any other appropriate limit) could be set that minimized the potential for far-field criticality.

3.4.2 Criticality controls

Several of the studies reviewed in Chapter 2 describe the types of criticality controls that could be specified as a result of post-closure criticality analyses. The most direct criticality control is to limit the fissile mass in a waste container. For spent fuel, this may be achieved most effectively by increasing burn-up or reprocessing and reusing fissile material. Other criticality measures that could be imposed for spent fuel/HLW or ILW disposal in a deep geologic repository are summarized in Table 3.1 in terms of controls on the waste form, waste container properties, disposal room geometry, and backfill materials. These controls are also listed in Figures 3.1 to 3.4 in the context of the five criticality scenarios discussed in Section 3.2.

Table 3.1 Summary of possible criticality controls

Component	Control
Waste form	Limit the fissile mass in each waste container.
	Distribute the fissile material in the waste form uniformly.
	Add low-solubility neutron absorbers (poisons) to the waste form.
	Dilute ^{239}Pu or enriched uranium with depleted uranium (99.7% ^{238}U).
	Limit the amount of neutron reflector and moderator materials in the waste.
	Compact the waste to minimize pore space and, thus, minimize the probability that a solution containing fissionable materials might collect in a critical geometry.
	Compact the waste to maximise the strength of the waste form and, thus, limit the potential for criticality as a result of post-closure compaction.

Component	Control
Waste containers	Optimize container wall thickness or container material specification to increase the neutron absorption capacity or decrease the neutron reflecting capacity of each container.
	Optimize the container material specifications to reduce the corrosion rate or increase container strength and thus increase expected container lifetime.
Disposal room	Limit the density of fissile material by reducing the density of waste containers in the repository.
Backfill	Use backfill to prevent neutron interaction between the containers.
	Add neutron poisons to backfill (such as magnetite).
	Use backfill to immobilize contaminated colloids.
	Use backfill materials to minimize pore space and, thus, reduce the probability that a solution containing fissionable materials might collect in a critical geometry.

Also, by minimizing the potential for different oxidation states occurring, or oxidation states changing, in the disposal rooms and groundwaters the potential for dissolution and precipitation of fissile material would be minimized. However, long-term conditioning of the near-field by backfill would need to be justified in a criticality safety case.

3.5 Discussion

In general, the studies reviewed in Chapter 2 emphasise the need for post-closure criticality to be considered as part of a repository safety case, although most post-closure criticality analyses suggest that criticality is a low-probability event with low consequences in terms of overall repository safety.

3.5.1 The neutron multiplication factor

Evaluation of the neutron multiplication factor, k_{eff}, for specific configurations of fissile material is a key part of any criticality analysis, and requires calculations using a neutron transport computer code; assessment of the respective merits of different neutron transport codes was outside the scope of this project. A system is subcritical if $k_{eff} < 1.0$, although demonstrations of criticality safety and calculations of criticality safety limits usually include an arbitrary margin of safety. Analyses aimed at evaluating the probability and consequences of post-closure criticality are likely to involve greater uncertainties and more conservative modelling assumptions than criticality safety calculations. Thus, it is reasonable to base such post-closure analyses on a value of $k_{eff} = 1.0$, and to account for bias and uncertainty in the modelling.

3.5.2 Post-closure criticality analysis

A consistent approach to assessing post-closure criticality in deep geologic repositories has been taken in disposal programmes, since the pioneering analyses undertaken by Behrenz and Hannerz (1978) for a proposed spent-fuel repository in Sweden. The studies reviewed were generally based on deterministic approaches, which involved assessing the criticality potential of specific waste, container, disposal room, near-field and/or far-field configurations. Consideration of the sequences and combinations of the processes and events necessary for the concentration of fissile radionuclides at various locations in a disposal system has led to the identification of five distinct post-closure criticality scenarios, although this specification of scenarios is not unique.

Scenario 1: criticality in an intact waste container;

Scenario 2: criticality in a leaking waste container;

Scenario 3: criticality in one or more collapsed waste containers;

Scenario 4: criticality in the near-field; and

Scenario 5: criticality in the far-field.

Analysis of each criticality scenario requires parameter values to be set and/or assumptions to be made in relation to each of the relevant factors. Scenarios 1, 2, and 3, are, to some extent, independent of repository design, in that they do not require detailed information on repository location and layout, or rock properties. Thus, it is reasonable for these scenarios to be included in post-closure criticality assessments in the UK in the absence of an ILW repository site and design. However, assumptions would need to be made concerning post-closure hydraulic and geochemical conditions in the vicinity of the waste containers. Factors such as initial saturation levels, natural groundwater conditions, thermal effects (heat from decay and exothermic reactions), gas generation, and any assumed borehole intrusion-related effects, may need to be considered in making assumptions concerning hydrogeochemical conditions in the disposal rooms.

Criticality Scenarios 1, 2, and 3, which describe criticality in waste containers, are also relevant to waste storage facilities (with the exclusion of backfill) and the pre-closure period of a repository (in which the timing of backfilling may be significant). In particular, Scenarios 2 and 3, involving damaged containers, may require consideration if long periods are envisaged for waste storage or the repository pre-closure phase.

The near-field and far-field scenarios are relevant only to a repository post-closure period, and may involve critical concentrations of fissile material emanating from more than one waste container. Assessment of these scenarios requires evaluation of radionuclide migration and accumulation rates for potentially critical geometries. With regard to far-field criticality, this may require an understanding of factors such as fracture properties and groundwater oxidation states, which could only be addressed as part of a site evaluation process. Without a specific ILW repository site or disposal concept, as at present in the UK, any post-closure criticality

safety assessment would need to make bounding assumptions concerning near-field and far-field conditions, based on, for example, generic repository data, in order to evaluate Scenarios 4 and 5.

An initial deterministic evaluation of these five criticality scenarios, involving consideration of their credibility for a particular disposal concept and/or evaluation of their consequences, may show that post-closure criticality concerns are insignificant. However, if post-closure criticality concerns remain after such analysis, then it may be possible to address these concerns by reducing conservatism and/or uncertainty in models and/or parameter values.

Alternatively, a risk-based analysis could be undertaken in order to evaluate the probability and consequences of post-closure criticality and, if necessary, account for criticality consequences in the repository PA. Risk-based analysis would require determination of the fission yield from criticality excursions and determination of the probability of criticality scenarios. The risk calculations would involve estimating the doses that could result from the additional radionuclides (fission-yield products) generated by possible future excursions. Such an approach would require use of a suite of PA codes and significant computational resources, but the method could be simplified by modelling only the processes directly related to criticality (i.e., the factors listed for each scenario in Section 3.2).

3.5.3 Criticality controls

Operators should aim to ensure that radiological doses or risks are as low as reasonably achievable, and so it may be more desirable to include controls on waste packaging and repository design that minimized the possibility of a criticality excursion occurring, than to account for the potential consequences of post-closure criticality in a full performance assessment. The implications of any criticality controls on all aspects of disposal system performance should be considered. The most direct criticality control is to limit the fissile mass in a waste container, although other controls may be imposed on the waste form, waste container properties, disposal room geometry, and backfill materials (see Section 3.4.2).

Extremely conservative assumptions, concerning the potential for the accumulation of fissile material in the near-field or far-field, could result in the specification of unnecessarily low criticality safety limits on the fissile content of waste containers. However, if the near-field and far-field criticality scenarios are assessed only once a repository site has been selected and characterized, then there is a possibility that, to alleviate post-closure criticality concerns, waste would have to be repackaged to reduce the fissile material content of waste packages, alternative criticality controls would have to be incorporated into the repository design, or the consequences of criticality scenarios would have to be included in repository PAs.

4. REVIEW OF ASSAY TECHNIQUES

This chapter presents the results of a review of assay techniques used in the UK for determination of the fissile material content of radioactive waste packages. The review provides an overview of the methodologies used for fissile material assay, including comments on factors which could affect measurement accuracy. The review also aims to set out the implications of the different assay techniques for setting pre-disposal criticality limits. Details of the review are provided in Green (1998). The following subsections provide:

- descriptions of the types of waste which could contain fissile material within the UK nuclear industry with an overview of the packaging used;

- descriptions of the non-destructive techniques used for assay of fissile material in packages containing intermediate-level waste (ILW), plutonium contaminated material (PCM), and/or low-level waste (LLW); and

- a summary of the destructive sampling and analysis methodologies used directly in, or in support of, assay of fissile material in packages containing PCM and/or LLW.

Lists of factors which could affect the accuracy of assay techniques are summarized in several tables in this chapter. These factors are not of equal importance and some are easier to mitigate against than others. However, it is not within the scope of this project to assess the means by which assay measurement errors and inaccuracies can be avoided.

4.1 Waste Types and Packaging

4.1.1 Low-level waste

LLW consists of general rubbish (such as used paper towels and discarded laboratory clothing) and other lightly contaminated plant items and equipment, as well as some materials that have been irradiated, arising predominantly from the operation of nuclear facilities. Building materials and larger items of plant and equipment are also produced from the decommissioning of facilities (Nirex, 1996).

BNFL's conditions for acceptance of LLW for disposal at the Drigg facility require that the waste should contain less than 0.1 GBq per tonne in total of ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{242}Pu. This is well below levels of concern relating to criticality. For example, 0.1 GBq per tonne of ^{239}Pu equates to about 44×10^{-3} g ^{239}Pu per tonne of waste. For a waste package with a bulk density of 1 kg per litre, the ^{239}Pu content would be equivalent to 44×10^{-6} g/litre of waste. In practice, this category of waste would contain less ^{239}Pu, because the total Pu alpha content must be less than 0.1 GBq per tonne. Ogilvie and Harris (1997) noted that normal plutonium assay techniques cannot achieve the accuracy necessary to demonstrate compliance with this limit. One method used to resolve this problem involves defining materials and items for disposal which are considered to have little plutonium content. These materials are monitored by surface contamination checks.

LLW produced at Sellafield is normally placed in large containers (e.g. skips). The wastes are volume-reduced and packaged into 1-m^3 boxes, and are assayed by gamma scanning, passive neutron counting and neutron interrogation. Other LLW is normally packaged inside 200-litre drums. The drums are assayed for gamma emitters (including fissile material content) by dedicated drum scanners. The 1-m^3 boxes and 200-litre drums are supercompacted and loaded into half height ISO containers, which are grouted and sent to the Drigg vaults for disposal.

4.1.2 Intermediate-level waste

ILW consists principally of materials that have been irradiated in a nuclear reactor (e.g. fuel cladding and reactor components), and equipment and materials that have been used in the processing of radioactive materials (e.g. ion exchange resins and filters). Most ILW, including PCM, is currently stored at the producing site and is packaged into a variety of containers, including 25-litre, 100-litre and 200-litre drums. PCM is commonly packaged, assayed and stored inside 100-litre or 200-litre drums. For example, at Dounreay, PCM inside 25-litre La Calhene containers is repackaged after assay into nominal 200-litre drums (Sellers, 1994). Fissile content measurements can be undertaken directly on large items of waste, such as decommissioned gloveboxes and redundant filters.

ILW will be packaged in steel or concrete containers for disposal, and will be either stored prior to disposal or sent directly to a deep repository. The ILW for underground disposal falls into two categories depending on packaging requirements (Nirex, 1997):

- Unshielded ILW, which requires the use of re-usable shielded containers for transport and remote handling underground at the repository, and will be disposed of in a range of containers, including 500-litre drums and 3-m^3 boxes; and

- Shielded ILW, which is contained in shielded disposal packages, such as 4-m boxes, and does not require additional external shielding.

Generally, all ILW will be grouted within the containers. Conditioning of some ILW streams commenced in 1990 (Nirex, 1996).

4.2 Non-Destructive Assay Methods

The principal, commercially available, non-destructive techniques used to assay the fissile material content of waste packages are:

- gamma spectroscopy;

- passive neutron counting; and

- neutron interrogation.

Calorimetric techniques (which rely on heat measurements) have also been explored, but these are generally used for more concentrated forms of fissile material, such as those found in reactor fuel.

Instrument manufacturers, operators and researchers usually quote the precision and, sometimes, the accuracy of non-destructive assay equipment, based on experiments with wastes and sources of known composition. Table 4.1 provides a list of factors which could affect the accuracy of non-destructive assay techniques. This list can be applied to gamma spectrometers, passive neutron counters and neutron interrogation systems. Other factors unique to each technique are listed in subsequent sections of this chapter.

Table 4.1 Factors relating to the verification and accuracy of non-destructive assay techniques

Factor	Comments
Container characteristics	
Package size	Measurement accuracy is likely to be greatest for small packages because variations in matrix properties will be less significant.
Package geometry	A non-standard package geometry may not have been accounted for during calibration.
Instrument calibration	
Accuracy of source calibration	Inaccurate assay results will be obtained if the assumed fissile material content of the calibration drums is erroneous.
Calibration assumptions	The calibration factors resulting from the assumption that the fissile material is present as a point source could be different from those resulting from the assumption that the fissile material is evenly distributed. If the assay system is calibrated using a small test source (e.g. 1 gram of plutonium), but is used to measure larger quantities of plutonium (e.g. 100 grams of plutonium), the accuracy of the measurement could be better than claimed (e.g. because of the increased signal to noise ratio) or worse than claimed. If a ^{252}Cf source is used for calibration, the differences between the neutron properties of ^{252}Cf and uranium and plutonium could lead to incorrect assumptions and hence measurement bias. These include differences in the neutron energy spectra and in the average number of neutrons per prompt fission (Rogers and Wells, 1987).
Instrument calibration operations	Erroneous results will be obtained if the instrument is not properly calibrated.

Factor	Comments
Operator errors	
Operator input errors	Erroneous results could be obtained if the operator enters incorrect details (e.g. wrong drum identifier) and this is not detected by another system (e.g. bar coding).
Analysis errors	
Fundamental constants	The fundamental constants used in the analysis may be wrong. For example, there may be software errors in the abundances of plutonium or uranium isotopes or daughter products.
Computer analysis equations	The equations used in the computer analysis software may be erroneous.
Computer software algorithms	Some events may be misinterpreted if the computer software algorithms are not understood by operators.

4.2.1 Gamma spectroscopy

Description of gamma scanners

The measurement of the gamma emissions from fissile material inside waste packages is undertaken using segmented gamma scanners (SGS). SGS technology has been applied routinely in the UK and world-wide for LLW and ILW packaged in 100- or 200-litre drums. The waste is not normally immobilized (e.g. by cementation) prior to determination of the gamma content. The non-intrusive passive measurement of fissile materials in waste drums is carried out using either sodium-iodide detectors or high-purity germanium detectors. The sodium-iodide detection systems have high relative efficiencies and are relatively cheap, but have largely been replaced by higher resolution intrinsic germanium systems. The high purity germanium detectors are cooled to liquid nitrogen temperatures to reduce the leakage current to the point that the associated noise does not spoil their energy resolution. These low temperatures are achieved in practice by adopting conventional liquid nitrogen cooling techniques. LLW SGSs also use electrically cooled detectors.

The performance of passive gamma scanners is affected by variations in the matrix and waste densities, and by variations in activity distribution within the waste drum. These effects are normally reduced by rotating the drum and by scanning different segments of the drum, hence the term segmented gamma scanner. SGSs are particularly applicable to waste containing heavy, high density objects (hard waste), because they correct for variations in waste density.

Passive gamma scanners can be used to quantify the fissile material content of waste packages, providing their concentrations are above the limit of detection of the system. Intrinsic germanium detectors are capable of measuring gamma emissions over the range 35 keV to about 10 MeV. The range of interest relevant to assay of all types of wastes is about 35 keV to about 3 MeV. The low end of this range is relevant to measurement of plutonium and uranium isotopes. The total amount of gamma activity in a 100- or 200-litre drum will be variable, depending on its origin. PCM waste should contain predominantly transuranics but

LLW could contain several kilobecquerels to several thousand Megabecquerels of activity. The limit of detection of the lower energy gamma emitters (e.g. uranium and plutonium isotopes) in LLW will depend on the amount of other gamma emitters present, because some peaks could be difficult to detect if the Compton Scattering contribution is high.

Accuracy of gamma scanners

Information on the accuracy of segmented gamma scanners has been obtained through discussion with instrument manufacturers. The indicative accuracy of a segmented gamma scanner used for assay of 200-litre (or smaller) drums ranges from +/- 15 % to 20 %, based on 1 gram of plutonium or uranium of known isotopic composition.

Rogers and Wells (1987) discussed the precision and accuracy of a segmented gamma scanner used to determine the plutonium content in 15- to 25-litre containers of low density waste, and concluded that, for large plutonium masses, the overall error was between 15 % and 20 %.

Table 4.2 provides a list of factors which could affect the accuracy of gamma spectroscopy. The information contained in this table is supplementary to that shown in Table 4.1.

Table 4.2 Factors relating to the verification and accuracy of gamma spectroscopy

Factor	Comments
Waste matrix characteristics	
Waste density	Attenuation of gamma radiation increases as a function of density. The presence of dense matter in the waste could cause measurement bias. The gamma emissions from plutonium shielded by high density material (e.g. lead) could lead to negative measurement bias.
Waste atomic numbers	Attenuation of gamma radiation increases as a function of atomic number. The presence of high atomic number materials in the waste could cause negative measurement bias.
Waste hardness	The harder the waste, the greater the necessity to ensure that the density corrections are accurate.
Concentrated plutonium or uranium	Attenuation of the gamma energies by discrete concentrations of uranium and/or plutonium could result in underestimates of fissile material content. (This limitation is recognized to have been one of the driving forces which led to the development of neutron assay systems.)
Waste immobilization	The gamma-emitting content of wastes is normally measured prior to immobilization. Results will less be accurate if the waste is immobilized prior to assay.
Isotopic composition of waste	Measurements may be inaccurate if the waste contains a mixture of materials with different isotopic compositions (e.g. a mixture of depleted and enriched uranium or plutonium with different burn-up rates).

Factor	Comments
Void space in drum	If the determination of drum density is not applicable to the voidage at the top of the drum, the assumption could be made that the top of the drum contains waste, leading to measurement bias.
Inhomogeneities within a drum	Measurement could be biased if the determination of drum density does not identify inhomogeneities that exist within the drum.
Other gamma emitters inside a drum	Other gamma emitters (e.g. ^{137}Cs, ^{60}Co) inside the waste could lead to high Compton Scattering and could make it difficult to quantify the low energy uranium and plutonium peaks.
Instrument characteristics	
Counting statistics	Statistical errors decrease as the counting period is increased.
Package density measurements	If the technique used to estimate the package density is not valid for the particular waste package, the measurements could be in error. This could arise if, for example, the density correction technique compares gamma peaks from the same isotope, and only a small amount of this isotope is present.
Instrument drift	Errors could arise if the gamma spectrometer channels drift away from the set values and are not corrected.
Detector cross talk	If two or more SGS detectors measure the gamma emissions from the same waste drum segment, or if one detector measures the gamma emissions from more than one segment, an overestimate of fissile material could be obtained.
Transmission correction source decay	Plutonium gamma scanners use ^{75}Se to determine the package density. The half life of ^{75}Se is 120 days. If the source decays and the ^{75}Se signals are barely measurable, package densities could be in error and measurement inaccuracies could arise.
Operator errors	
Background measurement errors	Inaccuracies can arise if there are background variations (e.g. other sources inadvertently brought close to the gamma spectrometer) or if the duration of background measurement is different to that used for package measurement.
Analysis errors	
Computer isotope library errors and omissions	Erroneous results could be obtained if the computer database does not contain the isotopes of interest (e.g. the ^{238}U daughter product ^{234}Pa energies).

4.2.2 Passive neutron counting

Description of passive neutron counting

Radionuclides that decay through spontaneous neutron emission can be quantified by direct measurement of their neutron emissions by a technique referred to as passive neutron counting. Total neutrons and/or the coincidence neutrons from radioactive decay can be measured. The latter type of measurement is referred to as passive neutron coincidence counting (PNCC) and is used to compensate for neutron emissions from alpha/neutron reactions. Passive neutron counting is used in the UK and world-wide to assay loose and packaged PCM and LLW, including drums of 25-, 100- and 200-litre capacity and crates. The waste is not normally immobilized (e.g. by cementation) prior to determination of the passive neutron emitter content.

Nearly all heavy nuclides with mass numbers greater than 230 atomic mass units undergo spontaneous fission and emit fast neutrons. The most important of these are ^{238}U, ^{238}Pu, ^{240}Pu, ^{242}Pu, ^{242}Cm, ^{244}Cm and ^{252}Cf, which can all be present in waste. The radionuclides are characterized by their even atomic numbers, even mass numbers and, in some cases, a long spontaneous fission half-life. Spontaneous fission of the nucleus results in the emission of, on average, two to three neutrons.

Waste matrices may contain light elements such as oxygen, carbon and nitrogen, which can interact with alpha particles (emitted by the actinides) in the waste matrix to produce fast neutrons. The neutron output from alpha/n reactions is therefore dependent not only on the amount of alpha activity, but also on the amount and types of light elements present in the waste. Simultaneous detection of pairs or triplets of neutrons allows discrimination between spontaneous neutron emitters and neutrons from alpha/n reactions.

Neutrons must be transformed into a charged particle within the counter in order to be detected. The most common technique is to slow down (thermalize) the fast neutrons to energies of about 0.025 eV. This can be achieved by surrounding the waste drum with a moderator material, which is typically a substance containing atoms of a similar mass to the neutron, such as water or polythene. The neutron detectors are embedded in the moderator materials. The detectors most commonly used in neutron waste assay systems are boron trifluoride (BF_3) and helium (^3He) gas proportional counters.

Accurate assay of neutron emitters requires coincidence electronics to separate the time-correlated spontaneous fission neutrons from the random alpha/n events. Spontaneous fission emits a number of neutrons simultaneously and, when a neutron is detected, it triggers an electronic device (gate) which opens for a finite time. If during this time a further neutron is detected then a coincident event is recorded (coincidence counting). If a neutron from a random event is detected, the probability of further neutrons being recorded while the gate is open is small. The system can also be used to count the total number of emitted neutrons.

If the isotopic composition of the waste is known, the concentrations of odd numbered (fissile) isotopes can be inferred. This is normally referred to as fingerprinting. When the waste contains mixtures of isotopes (e.g. U, Pu and Cm isotopes), prior knowledge of the isotopic

composition and/or other measurements may be necessary before the results can be interpreted.

Accuracy of passive neutron counters

Information on the accuracy of passive neutron coincidence counters has been obtained through discussion with instrument manufacturers. The indicative accuracy of a passive neutron coincidence counter used for assay of 200-litre (or smaller) drums ranges from +/- 5 % to 10 %, based on 1 gram of plutonium or uranium of known isotopic composition. Accuracies of +/- 15 % have been suggested for assay of 500-litre drums.

Sher et al. (1979) discussed the accuracy of coincident counting techniques tested with well characterized PuO_2 samples up to about 2 kg in mass. The experimental results indicated that it is possible to determine the effective ^{240}Pu mass to an overall precision of the order of +/- 2 % or better for samples ranging from a few grams to about 2 kg of total plutonium, which contains up to 20 % ^{240}Pu.

An assessment of overall PNCC errors was provided by Rogers and Hooton (1985), who concluded that, for small and medium samples with Pu effective less than 300 grams, the overall error should be less than 5 %. In many cases where the samples are well characterized, an accuracy of better than 3 % may be achieved. Measurements on large samples are susceptible to larger errors due to dead time corrections and significant neutron multiplication effects, and the overall error may rise to 10 %. In cases where the samples are not well characterized, the overall error could be substantially higher than 10 %.

Smith et al. (1985) reported the results of high performance neutron coincidence counting on nine high burn-up plutonium samples. The accuracy for a given batch of PuO_2 was within 0.5 - 1 %, and was 2.17 % for a wide range of batches.

Orr et al. (1981) examined systematic effects in the neutron coincidence counting of large PuO_2 samples. Measurements on PuO_2 in a variety of container types indicated observed real coincidence rate variations of up to 20 % for a 290 gram ^{240}Pu equivalent sample. Large errors could be introduced if accurate isotopic information (particularly for the isotopes ^{238}Pu and ^{241}Am) was not available.

Table 4.3 provides a list of factors which could affect the accuracy of passive neutron counting techniques. The information contained in this table is supplementary to that shown in Table 4.1.

Table 4.3 Factors relating to the verification and accuracy of passive neutron counting techniques

Factor	Comments
Waste Matrix Characteristics	
Waste atomic numbers	Moderation and absorption of neutrons generally decreases as a function of atomic number. The presence of low atomic number materials in the waste will increase the ability of the waste to moderate and stop the neutrons. This could lead to negative measurement bias.
Waste hardness	If the waste contains soft materials (e.g. hydrogenous materials and plastics) the moderating and absorption ability of the waste will increase, which could lead to negative measurement bias.
Alpha/neutron reactions	Interactions of the alpha particles from plutonium with light elements such as fluorine and oxygen will produce random neutrons. If these neutrons are not measured using time-correlated methods, they could be misinterpreted as spontaneous fission neutrons, which would lead to positive measurement bias.
Waste immobilization	The fissile material content of wastes is normally measured prior to immobilization. Less accurate results could be obtained if the waste is immobilized prior to assay and the moderating effect of the water in the cement formulations is not taken into account.
Isotopic composition of waste	Because passive neutron systems measure the even numbered neutron-emitting isotopes (e.g. ^{240}Pu) and infer the odd numbered isotopes (e.g. ^{239}Pu), it is essential to know the isotopic composition of the waste. Also, if the waste contains a mixture of fissile materials with different isotopic compositions (e.g. a mixture of depleted and enriched uranium or plutonium with different burn-up rates), measurement inaccuracies could result.
Passive neutron emitters inside a drum other than those required to be measured	Other sources of passive neutrons (e.g. spontaneous fissions of ^{242}Cm and/or ^{244}Cm) inside the waste package could lead to positive measurement bias.
Neutron poisons in the waste	Cadmium and gadolinium have high neutron capture cross-sections. If the waste contains materials such as these, and no account is taken of their presence, negative measurement bias will result.
Strong gamma fields from materials present in the waste	Strong gamma fields in the waste could be detected by the ^{3}He or BF_3 neutron detectors, leading to a positive measurement bias.
Moderating materials in the waste	Any materials inside the waste that would increase the moderating and absorption capability of the spontaneous neutrons could cause decreased response and possibly measurement inaccuracies if not taken into account.
Uranium and plutonium in the waste	The even numbered isotopes of uranium and plutonium emit spontaneous neutrons. Errors would occur if the relative contributions from plutonium and uranium were not taken into account.

Factor	Comments
Age of plutonium	^{241}Pu decays to ^{241}Am, which is an alpha emitter. This could cause an increase in the number of neutrons produced from alpha/n reactions, which could cause an increase in random neutrons and, if not correctly measured, will lead to positive measurement bias. The burn-up and age of the sample and the calibration source should, ideally, be similar.
Neutron multiplication	Spontaneous fission and alpha/n reaction neutrons emitted by the sample can cause further secondary fissions. The neutrons from the secondary fissions could be identified as primary fission neutrons and result in positive measurement bias. The secondary neutrons could also produce coincidence responses from single neutron events, leading to positive measurement bias.
Instrument Characteristics	
Counting statistics	Statistical errors decrease as the counting period is increased.
Saturation of detectors	Errors may occur if high neutron fields are present and the detector is saturated.
Detector wall effects	If neutrons strike the BF_3 or H-3 detector chamber walls, a smaller pulse is produced compared to when the neutrons deposit their energy in the detector gas. If this is not accounted for, negative measurement bias will result.
Detector malfunctions	If a small number of detectors are used in the assay system and problems arise (e.g. detector operating outside the plateau voltage), measurement errors would result.
Operator Errors	
Background measurements	At lower limits of detection it is necessary to shield combined passive and active assay systems from cosmic neutrons. Inaccuracies can arise if there are background variations (e.g. inadvertent neutron sources brought into the neighbourhood) or if the duration of background measurement is different to that used for package measurement.
Analysis Errors	
Time correlation methodology	If the time correlation methodology has errors, this will impinge on the accuracy of the final measurement.

4.2.3 Neutron interrogation

Description of neutron interrogation techniques

Fissile materials, such as ^{239}Pu and ^{235}U, which are present in either PCM or LLW can be made to fission if subjected to a source of thermalized neutrons. Neutrons released from the fissioning process can be detected and measured, thus providing a means of quantifying the amount of fissile material present. This measurement technique is referred to as active neutron interrogation. Active neutron interrogation in the UK has so far been restricted to the assay of

wastes packaged in 200-litre drums or smaller containers. In the US, the technique has been applied to both LLW and PCM packaged in crates.

Fissions can be induced in actinides with odd mass numbers (e.g. ^{235}U, ^{239}Pu) by neutron interrogation of the waste package using an external neutron source. Neutrons produced by the fissioning process, like spontaneous fission neutrons, possess a broad energy spectrum and are time-correlated. Most of the resultant neutrons are produced in less than 0.2 seconds after fission and are referred to as prompt neutrons. Prompt neutrons result from the direct fissioning of the fissile material present in the waste.

Delayed neutrons are also produced as a result of the spontaneous neutron emissions from the resultant fission products. These delayed neutrons are produced typically between 0.2 to 55 seconds after fission with an abundance of approximately 1 % of the prompt neutrons. Active neutron interrogation techniques have been developed where either the delayed or prompt neutrons are measured, thereby allowing the fissile material present in the waste to be quantified. Systems which measure delayed neutrons are based on the Californium Shuffler principle, and prompt neutron measurements are carried out using the Differential Die-Away (DDA) technique. If the isotopic composition of the waste is known, the concentrations of even numbered isotopes can be inferred using fingerprinting techniques.

Californium 252 Shuffler

The isotope ^{252}Cf has a half-life of 2.6 years and emits $2.6x10^{12}$ neutrons per gram per second by spontaneous fission. It is used in a Californium 252 Shuffler to interrogate a waste drum for fissionable nuclides. After a few seconds of interrogation, the ^{252}Cf source is quickly removed to a shielded position and the delayed neutrons emitted by the fission fragments in the waste drum are counted. The number of delayed neutrons emitted is proportional to the amount of fissionable material present in the waste drum. The cycle of interrogation and delayed neutron counting can be repeated many times to obtain good statistical precision.

Differential Die-Away

Active neutron interrogation, employing DDA techniques, can be used to provide a measure of the quantity of fissile material within a waste package. DDA uses a primary source of pulsed fast neutrons, which are thermalized before reaching the waste package. The thermalized neutrons induce fissions in any fissile material present, and the prompt neutrons which are emitted are detected and counted. The high yield of prompt fission neutrons results in a better detection efficiency than that achieved for the ^{252}Cf Shuffler, which detects a lower yield of delayed neutrons. The delayed neutron signal is also detected and the time separation between the prompt and delayed fission neutrons allows on-line corrections to be made for matrix absorption effects and self-shielding in hard wastes.

Accuracy of neutron interrogation techniques

Information on the accuracy of differential die away systems has been obtained through discussion with instrument manufacturers. The indicative accuracy of an active differential die away system (which also includes passive neutron coincidence counting) used for assay of 200-litre (or 500-litre) drums, ranges from +/- 15 % to 20 % based on 1 gram of plutonium or uranium of known isotopic composition.

Sprinkle et al. (1990) discussed the accuracy of an active/passive neutron counter based on a ^{252}Cf shuffler for 208-litre drums, installed in the Portsmouth Gaseous Diffusion Plant in Ohio, US. The results of a calibration exercise for an iron matrix were quoted as meeting the accuracy goal of 10 % and the precision goal of 1 % for 100 grams of ^{235}U.

Sampson et al. (1985) discussed matrix effects in the assay of fissile material in 200-litre drums by the neutron die-away technique. One conclusion relating to self-shielding of thermal neutrons was that a 1 gram sphere of highly enriched uranium will produce a response equivalent to about 0.1 gram of dilute materials, whereas there will be negligible self-shielding for 1 kg of 3 % enriched UO_2 at a density of 5 g/cm^3. The standard deviation for matrix corrected results was 15 %.

Table 4.4 provides a list of factors which could affect the accuracy of neutron interrogation. The information contained in this table is supplementary to that shown in Table 4.1.

Table 4.4 Factors relating to the verification and accuracy of neutron interrogation techniques

Factor	Comment
Waste Matrix Characteristics	
Waste atomic numbers	Attenuation of neutrons decreases as a function of atomic number. The presence of low atomic number materials in the waste will increase the ability of the waste to moderate and stop the neutrons from the fission process. This could lead to negative measurement bias.
Waste hardness	If the waste contains soft materials (e.g. hydrogenous materials and plastics), the neutron moderating and absorption ability of the waste will increase, which could lead to negative measurement bias.
Neutron poisons present in the waste	Cadmium and gadolinium have high neutron capture cross-sections. If the waste contains materials such as these, and no account is taken of their presence, negative measurement bias will result.
Strong gamma fields from materials present in the waste	Strong gamma fields in the waste could be detected by the ^3He or BF_3 neutron detectors, leading to a positive measurement bias.
Waste immobilization	The fissile content of wastes is normally measured prior to immobilization. Less accurate results could be obtained if the waste is immobilized prior to assay and the moderating effect of the water in the cement formulations is not taken into account.

Factor	Comment
Isotopic composition of waste	Active neutron interrogation systems directly measure odd numbered neutron emitting isotopes (e.g. ^{239}Pu) and the associated PNCC normally measures the even numbered isotopes (e.g. ^{240}Pu). Because a direct measurement of fissile material content is provided, these systems are less susceptible to variations in the isotopic composition of the waste. However, if assumptions on the isotopic composition of the waste have to be made (e.g. for mixtures of uranium and plutonium present in the waste) and if these are not correct, then the final result could be inaccurate.
Moderating materials inside waste	Any materials inside the waste that would increase the moderating and absorption capability of the fission neutrons could cause decreased response and possibly measurement inaccuracies if not taken into account.
Neutron multiplication	Neutrons emitted by the interrogated sample can cause further secondary fissions. The neutrons from the secondary fissions could be identified as primary interrogation fission neutrons and result in positive measurement bias. The secondary neutrons could also produce co-incidence responses from single neutron events, leading to positive measurement bias.
Instrument Characteristics	
Counting statistics	Statistical errors decrease as the counting period is increased.
Saturation of detectors	Errors may occur if high neutron fields are present and the detector is saturated.
Detector wall effects	If neutrons strike the detector chamber walls a smaller pulse is produced compared to when the neutrons deposit their energy in the detector gas. If this is not accounted for, negative measurement bias will result.
Detector malfunctions	If a small number of detectors are used in the assay system and problems arise (e.g. detectors operating outside the plateau voltage), measurement errors would result.
Dead time corrections	If accurate dead time coefficients are not determined, this could lead to measurement bias.
Prompt and delayed neutrons	The prompt neutron emissions are far more abundant than the delayed signals. An instrument which can measure prompt neutron signals (e.g. DDA) will have increased signal to noise ratios over other systems (e.g. Californium Shufflers), which could lead to increased accuracies.
^{252}Cf decay	The ^{252}Cf source (in a Californium Shuffler) has a half life of 2.65 years. Errors could arise if the ^{252}Cf decays beyond its design basis.
Faulty neutron generator	Errors could occur if faults in the neutron generator (in a differential die-away system) are not recognized.

4.3 Destructive Sampling and Analysis

4.3.1 Description of destructive sampling

Green et al. (1990) and Green et al. (1992) discussed package sampling techniques, mainly in relation to LLW, but the methodology is also applicable to PCM. For example, waste packages can be sampled for fissile material content by coupling them to a glove-box and opening them up in a controlled atmosphere. Samples can then be removed. Preliminary measurements can be made using hand-held instrumentation to assist in the choice of samples for further analysis.

A waste package may be opened to provide samples to measure the isotopic composition of the fissile material, or to provide reassurance of the absence of materials which could affect the NDA measurements (e.g. lead shielding, cadmium and gadolinium). The main problem associated with destructive sampling is the ability to take representative samples. It is not often feasible to sample the total contents of drums. Sampling errors can therefore be indeterminate.

In destructive analysis, the waste sampled from a selected package is dissolved in a suitable solvent to release the radioactive components from the waste matrix. Once the radioactivity is in solution, it is often necessary to separate the isotopes of interest (e.g. uranium and plutonium isotopes) by specific analytical methods before measurement for the radioisotope of interest. These separations can be lengthy and rely heavily on operator experience. Interference by other chemicals present in the waste can lead to method bias.

Once the sample is in solution, and the necessary chemical separations undertaken, measurements can be made using a variety of techniques including:

- gamma spectroscopy (for gamma-emitters, e.g. ^{239}Pu, ^{238}U);
- alpha spectrometry (for alpha emitters, e.g. isotopes of U and Pu);
- liquid scintillation counting (for alpha and beta emitters);
- mass spectrometry (for isotopic ratios, usually of Pu or U);
- atomic absorption spectrometry (for non-active elements such as Cd or Gd);
- emission spectroscopy (for non-active elements such as Cd or Gd);
- infrared spectroscopy (for organics); and
- gas chromatography (for organics).

Because it is not practical to sample the total content of waste packages, the package is often assayed using non-destructive techniques and the total package content calculated from both destructive and non-destructive results.

Destructive sampling and analysis techniques are not viable alternatives to NDA methods. Opening up waste packages can be time consuming and expensive and can produce secondary wastes. It can also result in increased radiation dose to analytical staff. For these reasons, destructive sampling and analysis should be used either to supply information which will assist in the interpretation of NDA results, or as confirmation that the NDA results are accurate.

4.3.2 Accuracy of destructive sampling

The accuracy of destructive sampling and analysis errors is dominated by the often large and unknown sampling errors. These errors could lead to overestimates or underestimates by factors which are difficult to quantify. Table 4.5 provides a list of factors which could affect the accuracy of destructive sampling and analysis.

Table 4.5 Factors relating to the verification and accuracy of destructive sampling and analysis techniques

Factor	Comments
Non-representative waste sampling	It is extremely difficult to take representative samples of heterogeneous waste packages. If the total package contents are not sampled, errors could arise from the deficiencies and limitations of representative sampling. This could lead to inaccurate destructive sampling and analysis results.
Package NDA measurements in support of destructive sampling and analysis	If the waste package is only partially sampled and non-destructive assay techniques (e.g. gamma spectroscopy) are used to assay the drum before and after sampling and derive a "destructive assay" result, NDA errors could influence the accuracy of the exercise. At any rate any interdependencies between the two methodologies should be recognized.
Sample dissolution	If the sample (e.g. concrete) is not totally dissolved and residues remain, errors could arise if the amount of activity in the residues is not accurately determined.
Sample separation	The sample separation technique could be a large source of error. These errors could arise from components of the waste that interfere with the chemistry of the separation.
Sample measurement	The sample measurement technique will have associated errors that will contribute to the overall error of the destructive analysis technique.

4.4 Discussion

The information contained in this chapter relates mainly to characterization of PCM and LLW in the UK and the US. The NDA systems for measurement of fissile material have been described mainly as stand-alone techniques, although passive neutron counting can be combined with gamma spectroscopy and neutron interrogation in the same instrument. In some instances, there are advantages in using more than one NDA technique. Also, nuclear wastes (such as spent fuel) often contain neutron poisons, which require consideration when measuring fissile material. These issues are discussed in the following subsections.

4.4.1 Gamma scanning and passive neutron counting

Gamma scanning combined with passive neutron counting could be used for confirmation measurements, or if the isotopic composition of the plutonium was not known. The gamma scanner would quantify the ^{239}Pu content and the passive neutron counter the ^{240}Pu content. The measurement principles, analysis equations and nuclear constants are sufficiently different to allow gamma scanning and passive neutron counting to be regarded as independent of one another.

The two techniques have complementary features which reinforce their independence. For example, the gamma scanner measures the ^{239}Pu content directly, whereas passive neutron counters measure the ^{240}Pu content and allow the ^{239}Pu content to be inferred. Also, the accuracy of gamma scanners can decrease with waste hardness, whereas the opposite applies to passive neutron counters.

4.4.2 Passive neutron counting and neutron interrogation

Passive neutron counting combined with neutron interrogation could be used for confirmation measurements, or if the isotopic composition of the plutonium was not known and a lower limit of detection than that provided by gamma scanning was required. The neutron interrogation system would quantify the ^{239}Pu content, and the passive neutron counter the ^{240}Pu content. The measurement principles and analysis equations are sufficiently different to allow these two methods to be regarded as independent of one another.

The two techniques have features which reinforce their independence. For example, as discussed above, passive neutron counters measure the ^{240}Pu content and infer the ^{239}Pu content, whereas neutron interrogation systems measure the ^{239}Pu content directly. The accuracy of both systems can increase with waste hardness, because of the absence of moderating and absorbing materials.

Increased confidence in non-destructive assay measurements may be obtained if from time to time selected packages are sampled and the waste samples are subjected to destructive chemical analysis. Results obtained by such destructive analysis should be consistent with the passive and active neutron measurements.

4.4.3 Assay of waste containing neutron poisons

Compounds of cadmium, gadolinium and boron (neutron poisons) can be found inside waste packages. Cadmium is used by the nuclear industry as a neutron absorber and gadolinium is used as a burnable poison in, for example, pressurized water reactor (PWR) and boiling water reactor (BWR) fuel. Boron is found in pyrex glass and is used as a burnable poison in PWRs. Wastes containing these neutron poisons can be assayed accurately by SGS, as their presence will not interfere with this type of measurement.

The neutron capture cross-section of these poisons has, to a first approximation, an inverse relationship to the neutron energy. The neutron energy spectrum is therefore the dominant property that will define the accuracy of the NDA method. PNCC measures spontaneous neutrons which have a spectrum containing both thermal and fast energies. Neutron poisons in the waste will cause some attenuation, which will increase as a function of the moderating ability of the waste matrix. The neutrons utilized by DDA are generally thermalized before they enter the waste chamber and, therefore, this technique is sensitive to neutron poisons. The ^{252}Cf shuffler neutrons have a relatively hard spectrum and are therefore less susceptible to the presence of neutron poisons.

One way to overcome these effects is to use a matrix correction method known as the "add a source" technique (Menlove and Ecclestone, 1992). The basis of this method is to measure the matrix perturbation to the counting rate from a small ^{252}Cf source on the outside of a sample. The information is then used to correct for the matrix perturbation on the inside of the sample.

4.5 Summary

This chapter provides summaries of the methodologies used to determine the fissile material content of packaged LLW and PCM. The three non-destructive methods used by the nuclear industry are:

- gamma scanning;
- passive neutron coincidence counting; and
- neutron interrogation.

Information on the accuracy of non-destructive assay devices has been obtained from instrument manufacturers.

- The indicative accuracy of a segmented gamma scanner used for assay of 200-litre (or smaller) drums under laboratory conditions is approximately +/- 15 % to 20 % based on 1 gram of plutonium or uranium of known isotopic composition. Thus, assay of a true 100-gram plutonium source inside a waste drum could give results ranging from 80 grams to 120 grams. Assay of a true kilogram of uranium could give results ranging from 800 grams to 1,200 grams.

- The indicative accuracy of a passive neutron coincidence counter used for assay of 200-litre (or smaller) drums under laboratory conditions is approximately +/- 5 % to 10 % based on 1 gram of plutonium or uranium of known isotopic composition. Thus, assay of a true 100-gram plutonium source inside a waste drum could give results ranging from 90 grams to 110 grams. Assay of a true kilogram of uranium could give results ranging from 900 grams to 1,100 grams.

- The indicative accuracy of an active differential die away system, which also includes passive neutron coincidence counting, used for assay of 200-litre (or 500-litre) drums under laboratory conditions is approximately +/- 15 % to 20 % based on 1 gram of plutonium or uranium of known isotopic composition. Thus, assay of a true 100-gram plutonium source inside a waste drum could give results ranging from 80 grams to 120 grams. Assay of a true kilogram of uranium could give results ranging from 800 grams to 1,200 grams.

These indicative accuracies are based on measurements on simulated waste packages containing fissile material as discrete metal oxide pellets. Measurements on drums of real waste may be less accurate, depending on the complexity of the waste form (as discussed in Section 4.2).

The accuracy and precision of measurements should be taken into account when setting criticality limits. Suppose, for example, the maximum permissible plutonium content of a 200-litre drum is 300 grams. If the accuracy of the NDA method is +/- 30 % (at three standard deviations), a working limit of 230 grams of plutonium per drum would be set. This ensures that the maximum permissible plutonium content and, hence, the criticality limit can not be exceeded.

Destructive sampling and analysis methods are not viable alternatives to non-destructive assay methods. Opening waste packages can be time consuming and expensive, can produce secondary wastes, and can result in increased radiation doses to analytical staff. Also, the accuracy of destructive sampling and analysis is often dominated by large and unknown sampling errors. However, if sampling errors are known, destructive sampling and analysis may be used to assist in the interpretation of NDA results, or to confirm that NDA measurements are accurate.

5. CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

1. Post-closure criticality should be considered as part of a repository safety case because of the possible consequences in terms of increased radiological hazard and mechanical damage to the engineered and natural barriers of the disposal system, and because of the public perception of potential for loss of management control. (Section 3.3.)

2. Following a literature survey, a review of methods used to address nuclear criticality concerns has been undertaken, including studies of ten post-closure criticality assessments carried out in radioactive waste disposal programmes around the world. The review covers a range of disposal concepts, including deep disposal of ILW and spent fuel, and shallow disposal of LLW. Although many of these disposal concepts have limited direct relevance to a future ILW repository in the UK, the criticality assessment methodologies used are generally applicable. (Section 2.2.)

3. Most post-closure criticality studies suggest that criticality is a low-probability event with low consequences in terms of overall repository safety. (Section 3.5.)

4. Evaluation of the neutron multiplication factor, k_{eff}, for specific configurations of fissile material is necessary for any criticality analysis, and requires calculations using a neutron transport computer code. Existing computer codes are probably adequate for such analysis. (Section 3.5.1.)

5. Analyses aimed at evaluating the probability and consequences of post-closure criticality may reasonably be based on a value of k_{eff} = 1.0, provided bias and uncertainty is accounted for in the modelling. A lower values of k_{eff}, which incorporates a criticality safety margin, is usually required to ensure operational safety. (Section 3.5.1.)

6. The analysis undertaken for the Konrad repository in Germany showed that considerations of post-closure criticality can lead to more restrictive limits on the fissile material content of waste containers than waste handling and storage limits (Section 2.3.3.)

7. It is difficult to draw any firm conclusions relating to numerical limits for disposal criteria, but the review has summarized limits on the fissile material content of waste containers used in several countries. (Table 2.13.)

8. Consideration of the sequences and combinations of the processes and events necessary for the concentration of fissile radionuclides at various locations in a disposal system has led to the identification of five distinct post-closure criticality scenarios, although the specification of scenarios is not unique. These scenarios describe: criticality in an intact waste container; criticality in a leaking waste container; criticality in one or more collapsed waste containers; criticality in the near-field; and criticality in the far-field. (Section 3.5.2.)

9. A deterministic evaluation of criticality scenarios, involving consideration of their credibility for a particular disposal concept and/or evaluation of their consequences, may show that post-closure criticality concerns are insignificant. Alternatively, a risk-based analysis could be undertaken in order to evaluate the probability and consequences of post-closure criticality and, if necessary, account for criticality consequences in a repository PA. Such an approach would require use of a suite of PA codes and significant computational resources. (Section 3.5.2.)

10. Operators should aim to ensure that radiological doses or risks are as low as reasonably achievable, and so it may be more desirable to include controls on waste packaging and repository design that minimized the possibility of a criticality excursion occurring, than to account for the potential consequences of post-closure criticality in a full performance assessment. Typical criticality measures that might be imposed for ILW disposal in a deep geologic repository are as follows. (Section 3.5.3.)

    10.1 Limit the fissile mass in each waste container.

    10.2 Distribute the fissile material in the waste form uniformly.

    10.3 Add low-solubility neutron absorbers (poisons) to the waste form.

    10.4 Dilute ^{239}Pu or enriched uranium with depleted uranium (99.7% ^{238}U).

    10.5 Limit the amount of neutron reflector and moderator materials in the waste.

    10.6 Compact the waste to minimize pore space and, thus, minimize the probability that a solution containing fissionable materials might collect in a critical geometry.

    10.7 Compact the waste to maximise the strength of the waste form and, thus, limit the potential for criticality as a result of post-closure compaction.

    10.8 Optimize container wall thickness or container material specification to increase the neutron absorption capacity or decrease the neutron reflecting capacity of each container.

    10.9 Optimize the container material specifications to reduce the corrosion rate or increase container strength and thus increase expected container lifetime.

    10.10 Limit the density of fissile material by reducing the density of waste containers in the repository.

    10.11 Use backfill to prevent neutron interaction between the containers.

    10.12 Add neutron poisons to backfill (such as magnetite).

    10.13 Use backfill to immobilize contaminated colloids.

10.14 Use backfill materials to minimize pore space and, thus, reduce the probability that a solution containing fissionable materials might collect in a critical geometry.

11. A review of assay techniques for fissile material has been carried out. A range of techniques exists which provides confidence that compliance with fissile material criticality limits can be achieved. (Section 4.5.)

5.2 Recommendations

With regard to involvement in the assessment of post-closure criticality safety cases, the Environment Agency could:

1. Develop and maintain an in-house capability to review criticality safety cases for radioactive waste disposal prepared by UK waste producers and disposers. The material provided in this report serves to form the foundations for this capability.

2. In collaboration with other regulators, review criticality safety assessments prepared by UK waste producers and disposers for ILW packages destined for deep geologic disposal. Such reviews should assess:

   - whether pre-closure and post-closure criticality scenarios have been addressed adequately;

   - whether reasonable assumptions have been made with regard to modelled features, events, and processes and associated parameter values;

   - whether a reasonable criticality assessment methodology has been used, involving an appropriate neutron transport code and, if necessary, suitable radionuclide transport codes; and

   - whether the issues of assay measurement accuracy, model bias and uncertainty, and criticality safety margins have been addressed adequately.

Concerns with regard to the accuracy of assay techniques could be further addressed by:

3. Assessing the means to mitigate those factors that affect the accuracy of assay techniques for measuring fissile material contents of waste. This assessment could be carried out in conjunction with technical auditing of the instruments, analyses, and software used by waste producers and packagers, and could be undertaken by the Environment Agency in collaboration with the NII.

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Appendix A

Glossary

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Glossary

Alpha/n reactions
Low atomic mass elements, such as oxygen, carbon and nitrogen, can interact with alpha particles to produce fast neutrons. The neutron output from these alpha/n reactions is dependent on the amount of alpha activity and the amount and types of light elements present.

Burn-up
Burn-up refers to the significant reduction in the quantity of one or more nuclides arising from neutron absorption in a nuclear reactor. The term can be applied to fuel or other reactors.

Chain reaction
A chain reaction is a series of nuclear transformations initiated by a single nuclear fission. For example, the fission of a ^{235}U nucleus is accompanied by the emission of one, two, or three neutrons, each of which is capable of causing further fission of ^{235}U nuclei. When each transformation causes an average of one further transformation, the reaction is said to be critical. If the average number of further transformations is less than one, the reaction is subcritical; if it exceeds one, it is supercritical.

Coincidence counting
Some processes, such as spontaneous fission, emit a number of neutrons simultaneously. When a neutron is detected, it triggers an electronic device or gate which opens for a finite time. If during this finite time a further neutron is detected, then a coincident event is recorded. If a neutron from a random event, such as reaction of alpha particles with fluorine atoms, is detected, the electronic gate is opened for the same time but the probability of further neutrons being recorded during the set time is greatly reduced. This method of neutron counting is referred to as coincidence counting and is used to differentiate between spontaneous fission and alpha/n reaction neutrons.

Coincident neutrons
On average between two and three neutrons are simultaneously emitted during a fission event. These simultaneous, or coincident neutrons, are detected by coincidence counting techniques.

Compton Scattering
Compton scattering refers to the transfer of incident gamma photon energy to an electron in the absorbing material. The incident photon loses energy and is detected by a gamma spectrometer as a low-energy continuum.

Coulombic forces
Atomic electrons move around the nucleus as a result of the attractive electrostatic forces between the positive nucleus and the negative charge on the electron. The protons inside the nucleus tend to repel each other as a result of the repulsive electrostatic forces between the positive charges. This electrostatic repulsion can contribute to spontaneous fission of heavy elements. These electrostatic forces are referred to as Coulombic forces.

Criticality
Criticality refers to the condition under which the number of neutrons propagated within the mass of a fissile material at least equals the number of neutrons escaping from the system plus those required to sustain neutron production.

Critical mass
The quantity of fissile material necessary to generate a self-sustaining neutron reaction is referred to as the critical mass.

Electronic gate
A coincidence unit is an electronic device that accepts pulses (events) in two or more input channels and provides an output signal only if the input pulses arrive within a prescribed period. An electronic gate of this type enables discrimination between coincident neutrons arising from fission and random neutrons, such as alpha/n neutrons, from other sources.

Enrichment
An element is enriched by increasing the abundance of a particular isotope (e.g., a fissile isotope) in a mixture of the isotopes of the element. The enrichment is the proportion of atoms of a specified isotope present in a mixture of isotopes of the same element, where the proportion is greater than that in a natural mixture.

Even numbered actinides
Nearly all heavy nuclides with mass numbers greater than 230 atomic mass units undergo spontaneous fission and emit fast neutrons. The most important of these are ^{238}U, ^{238}Pu, ^{240}Pu, ^{242}Pu, ^{242}Cm, ^{244}Cm and ^{252}Cf. These radionuclides are characterized by their even atomic numbers and even mass numbers.

Fissile material
A fissile material can sustain a nuclear chain reaction with low-energy (slow) neutrons given adequate mass or concentration.

Fission
Fission refers to the splitting of a heavy nucleus of an atom into two or more fragments of comparable size, usually as the result of the impact of a neutron on the nucleus. Fission is normally accompanied by the emission of neutrons or gamma rays. Plutonium, uranium, and thorium are the principal fissionable elements.

Fissile gram equivalent
The number of grams of fissile material of a given fissile species that it takes, on an equivalent basis comparison to ^{235}U, to form a critical mass is referred to as a fissile gram equivalent.

Gamma emissions

In gamma decay, when a nucleus moves from an excited state to a state of lower energy, the energy difference between the two states is released in the form of a photon. Some decay schemes are complex, resulting in a number of gamma photons per nuclide decay. For example, decay of ^{239}Pu produces 160 gamma photons, rays or lines. The decay scheme of each radionuclide shows probabilities associated with the de-excitation transitions, known as branching ratios. These are used to deduce the number of gamma photons produced per disintegration of a nucleus. These gamma emissions are measured by gamma spectroscopy. For example, for every 100 atoms of ^{60}Co which decay to ^{60}Ni, 99.88 atoms produce a 1.17 MeV gamma line and 99.98 produce a 1.33 MeV gamma line. The two branching ratios are quoted as 99.88 % and 99.98 % respectively.

Leakage Current

Suppose that a semiconducting crystal is exposed to ionizing radiation, at a reduced temperature such that the conduction band is devoid of free electrons. The radiation imparts energy to the semiconductor and free electrons are excited to the conduction band. For every electron raised to the conduction band, a vacancy or hole is created in the valence band in the form of a missing valence electron. If a voltage is applied across the crystal, the electrons will be collected at the anode and the holes at the cathode, and a current will flow in the external circuit. The semiconductor acts as a radiation detector. If a high-purity crystal has a low-energy gap between the valence and conduction bands, electrons can be excited by thermal energy available in the lattice in the conduction band and, on application of an electric field, a small leakage current will flow in the crystal. These leakage currents mitigate against the use of semiconductors as high-efficiency absorbers of radiation energy. Gamma detector designers take steps to reduce leakage currents to a minimum.

Negative feedback

During a supercritical chain reaction the power and temperature will rise until negative feedback becomes large enough to reduce the system to a subcritical condition. The main negative feedback mechanisms associated with temperature increases are: density decrease from thermal expansion, which can increase neutron leakage; Doppler effects, which increase the probability that neutrons are absorbed without fission; and thermal expansion of the moderator, which causes fewer neutrons to be slowed down.

Neutron capture cross-section

The target area or cross-section presented by a nucleus to an approaching neutron is referred to as the neutron capture cross-section. Cross-sections are measured in barns (10^{-28} m^2) and give a measure of the probability of a neutron-induced reaction occurring.

Neutron detectors
Neutrons are uncharged particles and are detected by interactions which produce energetically-charged particles, such as protons and/or alpha particles. The ideal neutron detector is one which has a large cross-section for the reaction, thus allowing efficient detectors to be built with small dimensions. The detectors most commonly used in neutron waste assay systems are either boron trifluoride (BF_3) or helium (^3He) gas proportional counters. In both of these, the neutron energy is deposited in the target material which is a gas. The boron in the BF_3 counters is enriched with ^{10}B (to about 70 %) because of its high neutron capture cross-section relative to ^{11}B.

Neutron moderators
Neutron moderators are used in the core of thermal reactors to promote critical conditions by slowing down the high-velocity neutrons from the fission process. Slow (thermal) neutrons are less likely to be absorbed by ^{238}U in the reactor and thus have a much greater probability of producing a fission in ^{235}U and ^{239}Pu (fissile materials). Common neutron moderators used in a nuclear reactor are natural water and graphite, because the hydrogen and carbon atoms of these materials slow down neutrons effectively through scattering, while absorbing few of them.

Neutron multiplication factor
The neutron multiplication factor, k_{eff}, is the ratio of neutrons produced from one generation to the next; values less than one indicate a system that will be subcritical.

Neutron reflectors
To promote critical conditions in nuclear reactors, materials are placed around the reactor core to reflect back into the core many of the neutrons that would otherwise escape. Common neutron reflector materials are graphite, beryllium, water, and natural or depleted uranium.

Odd numbered actinides
The isotopes with odd atomic and mass numbers tend to be fissile and have very long spontaneous fission half lives. The most important fissile radionuclides are ^{233}U, ^{235}U and ^{239}Pu.

Positive feedback
It is possible for positive feedback mechanisms to cause the reactivity (and power) of a critical system to increase (autocatalytic criticality). For example, a fissionable material may be subcritical because it contains too much neutron moderator (such as water), which behaves, in part, as a neutron poison (an over-moderated system). Removal of some of the moderator may cause the system to become critical. The resulting heat energy produced by fission reactions may cause further expulsion of the moderator, and further increase in reactivity. Such an instability will continue until the system configuration changes.

Random neutrons
Some processes, such as the interaction of alpha particles with low atomic number materials, emit neutrons which appear one at a time. These neutrons are termed random neutrons.

Transmission source
A transmission source is a radioactive source that is positioned at one side of a waste package such that the signals transmitted through waste package can be measured. The stronger the signals, the less dense the material inside the package. ^{75}Se is used for plutonium scanners and ^{152}Eu for low-level waste scanners.

Waste form
The materials comprising the radioactive components of waste and any encapsulating or stabilizing matrix.

Waste package
The waste form and its container.