Modelling of Consequences of Hypothetical Criticality:
Post-closure Criticality Consequence Analysis for HLW, Spent Fuel, Plutonium and HEU Disposal

To: Radioactive Waste Management Limited
Date: 20 March 2015
From: AMEC
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\(^\dagger\) When Issue 2 of this report was produced Radioactive Waste Management Limited was known as the Radioactive Waste Management Directorate (RWMD). Since the modifications in Issue 3 of this report are to correct a data error (see page vii) the document refers to RWMD throughout, other than on this page and the title page.
Title
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Prepared for
NDA RWMD

Your Reference
RWM005140

Our Reference
AMEC/SF2409/012 Issue 3

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<table>
<thead>
<tr>
<th>Name</th>
<th>Signature</th>
<th>Date</th>
</tr>
</thead>
<tbody>
<tr>
<td>Author</td>
<td>R.M. Mason</td>
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</tr>
</tbody>
</table>

Transport Flask Photograph courtesy of Magnox Electric Ltd
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## Document History

<table>
<thead>
<tr>
<th>Version</th>
<th>Author</th>
<th>Date</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
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<td>R.M. Mason</td>
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<td>R.M. Mason</td>
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Executive Summary

The Nuclear Decommissioning Authority (NDA) has been charged with implementing the UK Government’s policy for the long-term management of higher activity radioactive waste by planning, building and operating a Geological Disposal Facility (GDF). The Radioactive Waste Management Directorate (RWMD) is in the early stages of planning for implementation. At present a site for a GDF has not been identified, and RWMD has produced an initial ‘generic’ Disposal System Safety Case (DSSC) to put forward the safety arguments for geological disposal using a range of illustrative disposal concepts and host geologies.

Given that a GDF will include the disposal of fissile nuclides, and such nuclides could, under certain conditions, lead to an unplanned neutron chain reaction (criticality), the demonstration of criticality safety forms an important part of the DSSC. In particular the Environment Agency Guidance on Requirements for Authorisation (GRA) for a GDF requires 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.” In addition, the guidance states that the “environmental safety case should also investigate, as a ‘what-if’ scenario, the impact of a postulated criticality event on the performance of the disposal system”. The environmental safety case is a key document in the DSSC.

RWMD has undertaken research over a number of years to ensure criticality safety at all stages of radioactive waste disposal. The criticality safety sub-topic ‘Modelling of Consequences of Hypothetical Criticality’ has a clear focus on understanding the consequences of hypothetical ‘what-if’ post-closure criticality events in a GDF. The sub-topic ‘Likelihood of Criticality’ considers the possibility of post-closure criticality.

For all radioactive wastes disposed of within a GDF, the initial emplacement will be significantly sub-critical by design and/or control of fissile content, so that substantial changes would have to occur to one or more criticality controls (such as fissile mass, fissile concentration, neutron moderation, neutron leakage or geometry) for a critical configuration to be possible. Therefore, the expectation is that any ‘what-if’ criticality scenario will be unlikely. Furthermore, for any scenario it is expected that the greater the changes required for a critical system to develop (e.g. the greater the fissile mass), the less likely the formation of that system becomes.

To understand the potential consequences of hypothetical post-closure criticality events a significant body of research was undertaken within the Understanding Criticality under Repository Conditions (UCuRC) programme from 2002 to 2010. This programme identified that two types of transient criticality event could, hypothetically, develop:

- quasi-steady-state for a system where a just-critical system could be maintained over a period of time; and
- rapid transient, characterised by a short-lived energetic release.

During the UCuRC programme software models of how such hypothetical critical events could evolve were developed. The transient criticality models are known as the QSS (Quasi-Steady-State) model and the RTM (Rapid Transient Model). For the latter a Bounding Approach model is also available as a simpler alternative. During the UCuRC programme the application of these models predominantly focused on the criticality scenario of post-closure accumulation (transport and accumulation of fissile nuclides forming a localised critical volume) from the storage of Intermediate Level Waste (ILW) in a GDF located within a higher strength rock environment. The UCuRC analysis and results were considered sufficient to support the conclusion in the 2010 Criticality Safety Status Report (CSSR) that, for the ILW in higher strength rock illustrative disposal concept, post-closure criticality would be a low consequence event.

Following the publication of the CSSR it was recognised that further research was required to understand both the likelihood and consequences of post-closure criticality scenarios. For consequence analysis a key objective was to extend the application of the transient criticality models to include higher heat generating wastes such as plutonium and Spent Fuel (SF), other illustrative disposal concepts, and further scenarios for hypothetical criticality. A second objective
was to develop (as required) and document the transient criticality models. This has been completed through the publication of user guides for the QSS, RTM and Bounding Approach software models.

This report is one of two reports describing how the transient criticality models have been applied to understand the potential consequences of 'what-if' post-closure criticality scenarios. This report focuses on scenarios following the disposal of High Level Waste (HLW), SF, plutonium and Highly Enriched Uranium (HEU), whereas the second focuses on scenarios following the disposal of Low Level Waste (LLW) and Depleted, Natural and Low Enriched Uranium (DNLEU). The work described in both reports is bounded by the 2007 Radioactive Waste Inventory, and the illustrative disposal concepts of the 2010 DSSC.

One method for identifying criticality scenarios for further investigation is the use of Features, Events and Processes (FEPs) analysis. Combinations of FEPs, each of which represents some change to the original sub-critical configuration, form ‘what-if’ scenarios of interest. Building on an overview of the illustrative disposal concepts, together with information on package and near-field evolution behaviour, this report summarises an alternative, and complementary, approach to FEP analysis to identify ‘what-if’ scenarios for criticality. Through the consideration of criticality controls such as fissile mass and concentration, two ‘what-if’ scenarios are identified:

- Localised accumulation of fissile materials from one or more waste packages in any region of a GDF or the host rocks;
- The in-package scenario for a single waste package. This is applicable to waste packages that may contain sufficient fissile material for a critical configuration given significant package evolution. It could apply to some SF, plutonium and HEU but not HLW. For a critical configuration to develop, partial flooding with water or significant degradation and rearrangement of the package contents would be necessary.

The methodology used in this work to understand the consequences of criticality is:

- Determination of what configurations of fissile material would have to develop for hypothetical criticality events to occur, by undertaking static criticality calculations with criticality software. For the research reported here, analysis has been extended from that achieved during the UCuRC programme by the addition of calculations for in-package scenarios. For SF the analysis focused on the partial flooding of fuel packages containing Pressurised Water Reactor (PWR) fuel. The analysis includes calculations of the axial variation in irradiation along fuel elements for different levels of average fuel irradiation. For plutonium and HEU waste packages the analysis focused on the removal of neutron absorbers and dissolution of packaging materials.
- Determination of how hypothetical critical configurations could evolve as either rapid transient or quasi-steady-state criticality events. This involves determining whether an increase in temperature acts to make a system more or less critical. The RTM or Bounding Approach model is required for the former and the QSS model for the latter.
- Specification of representative calculations for criticality consequence analysis using the transient criticality models, to sufficiently scope parameter space and identify trends.
- Decisions on how to apply the models to the specified calculations, including the construction of model inputs.
- Application of the models to the specified calculations and analysis of the results.

Using the above methodology has produced valuable insight into the consequences of a range of hypothetical criticality events for two ‘what-if’ scenarios for post-closure criticality.

For the accumulation scenario it is concluded from static criticality calculations for idealised spherical accumulations that:

- Critical masses are hypothetically possible in bentonite (used as a buffer around waste packages), granite (higher strength rock) and clay (lower strength sedimentary rock). A critical mass is not possible in evaporite rock for the specific material composition and porosity.
considered, but might be possible for a different chemical composition or a larger porosity. For the granite material composition no critical systems are possible for low enriched uranium.

- Where critical systems are possible, a specified fissile material will have a minimum critical mass, below which criticality is not possible.
  - For any host material the accumulation of $^{239}\text{PuO}_2$ provides the lowest value for the minimum critical mass. Therefore, since the minimum critical mass for a $^{238}\text{UO}_2$ accumulation is larger than for a $^{239}\text{PuO}_2$ accumulation in a given host material, the fissile mass required for criticality will increase as $^{239}\text{Pu}$ decays to $^{235}\text{U}$.
  - For the host materials where the minimum critical mass is the lowest ($^{239}\text{PuO}_2$ accumulation in bentonite) at least 1 kg of $^{239}\text{PuO}_2$ must accumulate. For other host materials the minimum critical mass for $^{239}\text{PuO}_2$ accumulation is larger.
  - If $^{238}\text{UO}_2$ is present, then the minimum critical mass can be much larger. In bentonite, for example, 100% enriched uranium has a minimum critical mass of 2.5 kg of $^{235}\text{UO}_2$, whereas for 3% enriched uranium the minimum mass increases to 630 kg of UO$_2$, of which 19 kg is $^{235}\text{UO}_2$.

- Generally, as the porosity of the host rock decreases, the minimum critical mass for a given fissile material increases. For example, with an assumed porosity of 12% the minimum critical mass for $^{239}\text{PuO}_2$ in clay is 3.1 kg, whereas for granite with an assumed porosity of 1% the minimum critical mass for $^{239}\text{PuO}_2$ is more than ten times larger at 44 kg. In addition to porosity, the chemical composition of the host rock can also have a significant effect on the critical mass.

- In any host material the fissile mass required for a critical configuration would require the fissile contents of multiple HLW packages to accumulate, so that this scenario is not considered credible for HLW.

- The majority of hypothetical accumulation systems would evolve as quasi-steady-states, if maintained by the continued accumulation of fissile materials.

- Rapid transient criticality, which is only possible for systems where increases in temperature make the system more critical, requires sufficient $^{239}\text{Pu}$ to be present. Furthermore, rapid transient criticality is only possible in a narrow range of fissile concentrations (5 kg/m$^3$ to 11 kg/m$^3$ for the accumulation of $^{239}\text{PuO}_2$ in bentonite, for example), and this concentration range as $^{239}\text{Pu}$ decays, with a half life of 24,100 years.

- As $^{239}\text{Pu}$ decays to $^{235}\text{U}$ the mass of fissile material required for rapid transient criticality increases significantly. In bentonite, for example, a rapid transient requires at least 2.6 kg of $^{239}\text{PuO}_2$. After four half-lives at least 20 kg of $^{239}\text{PuO}_2$ and $^{235}\text{UO}_2$, in ratio 6.25:93.75 due to decay, would be required.

- The above trends suggest that the longer it takes for a hypothetical critical accumulation of fissile material to occur, the less credible a rapid transient criticality becomes, with the expectation that rapid transient criticality is not credible for hypothetical critical accumulations that require post-closure timescales of at least 100,000 years to develop.

For the accumulation scenario it is concluded from transient criticality calculations for idealised spherical accumulations that:

- For systems that could evolve as quasi-steady-states the QSS model predicts that for mass arrival rates of the order of 1 g/year, or lower, the power is limited to a few kilowatts, and temperature rises of more than 10 °C above ambient are limited to a localised radius of a few metres. Larger consequences are possible at higher rates of accumulation, but only if it is assumed that fissile accumulation can continue when the pore water boils. Pore water boiling will generally act to not only prevent accumulation of fissile material, but also acts to reduce neutron moderation.

- The rule of thumb for QSS calculations is a powerful tool in understanding how results will change with different fissile material arrival rates. The rule, which has been confirmed for a wide range of calculations, states that, provided the temperature increase is not sufficient for pore water to boil then an increase in the arrival rate by a factor of X:
will scale the peak temperature increase (above ambient) and power by a factor of \(X\); but
those peaks will occur at a time scaled by \(1/X\).

In many cases the rule applies even if the arrival rates are sufficient to boil pore water in the accumulation region.

- For systems that could evolve as rapid transients, considering 10 kg of fissile oxide as an illustrative mass for accumulation, the Bounding Approach model predicts an energy release which could create a local cavity radius of between two to six metres. Local cracking would extend to no more than a few tens of metres in radius. Furthermore, as \(^{239}\text{Pu}\) decays to \(^{235}\text{U}\), the consequences of rapid transient events reduce. Larger mass accumulations, which are considered less likely, and would initially take longer to accumulate, would have larger consequences. However, the relationship is weaker than linear; for example, increasing the mass of \(^{239}\text{PuO}_2\) by a factor of ten scales the cavity radius by a factor of about three. The relationship is weaker still if the decay of \(^{239}\text{Pu}\) is taken into account.

**In-package scenarios** are only credible for waste packages that contain sufficient fissile mass. They are therefore conceivable, but considered unlikely, for spent fuel, plutonium and HEU, but not HLW. For **in-package scenarios** the conclusions of static criticality calculations are:

- For HEU and plutonium wastes, assuming the can-in-canister packaging concept with a ceramic wasteform within cans surrounded by borosilicate glass within an outer canister, it is concluded that:
  - The illustrative wasteform and package design will not become critical even if the neutron absorbers are removed from the ceramic wasteform, and the borosilicate glass within the canister is completely dissolved and replaced by water.
  - There is therefore no requirement for in-package transient criticality analysis for HEU and plutonium wasteforms disposed of using this concept.

- For spent fuel waste, static criticality analysis for fresh and irradiated PWR fuel in an illustrative package design containing four PWR fuel elements, allows the following conclusions to be drawn:
  - Both vertically and horizontally oriented spent PWR fuel packages could, hypothetically, become critical, following partial package flooding with water. However, they would not become critical from flooding if the average irradiation of each fuel element is greater than 35 GWd/Te. The average irradiation assumed for PWR fuel is 55 GWd/Te.
  - For low or zero irradiated fuel, where transient criticality events are hypothetically possible from the flooding of a package, only quasi-steady-state transient criticality could result.

For the **in-package scenario**, assuming a worst-case of flooding within a vertically oriented PWR fuel package, containing four fresh fuel elements, it is concluded from transient criticality calculations with the QSS model that:

- For a large range of assumed rates of flooding of the void space within the package, from hours to thousands of years, the peak temperature and power of the transient do not vary.
- Assuming transients where the heat transfer is dominated by the thermal properties of bentonite for the conduction of heat away from the critical region, the maximum power of the criticality transient is less than two kilowatts. It is noted that this power is similar to that which packages could be subject to from the decay heat of irradiated fuel in the earlier stages of emplacement within a GDF.
- If the thermal properties of steel are used instead, representing heat transfer dominated by the thermal properties of the package insert, then the QSS model predicts a larger maximum power of about ten kilowatts. This is considered less realistic than the bentonite case, but indicates the sensitivity of model outputs to assumed material properties.

These conclusions and the underlying research are an important component in demonstrating that the consequences of hypothetical post-closure criticality events are not a significant concern. The analysis of in-package scenarios is a significant extension to the research undertaken during the UCuRC programme.
## Contents

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Glossary</strong></td>
<td>1</td>
</tr>
<tr>
<td>Glossary of Terminology</td>
<td>1</td>
</tr>
<tr>
<td>Acronyms and Names</td>
<td>1</td>
</tr>
<tr>
<td><strong>1 Introduction</strong></td>
<td>3</td>
</tr>
<tr>
<td>1.1 Background</td>
<td>3</td>
</tr>
<tr>
<td>1.2 Criticality Safety</td>
<td>3</td>
</tr>
<tr>
<td>1.3 Post-closure Criticality</td>
<td>4</td>
</tr>
<tr>
<td>1.4 Report Overview</td>
<td>6</td>
</tr>
<tr>
<td><strong>2 Overview of Waste Packages, Disposal Concepts and Post-closure Evolution</strong></td>
<td>7</td>
</tr>
<tr>
<td>2.1 Waste Packaging Concepts</td>
<td>7</td>
</tr>
<tr>
<td>2.2 Waste Inventory</td>
<td>12</td>
</tr>
<tr>
<td>2.3 Illustrative Disposal Concepts</td>
<td>13</td>
</tr>
<tr>
<td>2.4 Overview of Post-closure Evolution</td>
<td>20</td>
</tr>
<tr>
<td>2.5 FEPs for Hypothetical Post-closure Criticality</td>
<td>24</td>
</tr>
<tr>
<td><strong>3 Scenarios to Scope the Consequences of Hypothetical Criticality</strong></td>
<td>26</td>
</tr>
<tr>
<td>3.1 The UCuRC Programme</td>
<td>26</td>
</tr>
<tr>
<td>3.2 Selection of ‘What-if’ Scenarios for Further Criticality Consequence Analysis</td>
<td>27</td>
</tr>
<tr>
<td>3.3 Methodology for Criticality Consequences Analysis</td>
<td>32</td>
</tr>
<tr>
<td><strong>4 Static Criticality Analysis</strong></td>
<td>35</td>
</tr>
<tr>
<td>4.1 Determining Whether Scenarios could be Critical</td>
<td>35</td>
</tr>
<tr>
<td>4.2 The UCuRC Programme</td>
<td>35</td>
</tr>
<tr>
<td>4.3 Static Criticality Analysis Methodology</td>
<td>36</td>
</tr>
<tr>
<td>4.4 Information Required for Static Analysis</td>
<td>38</td>
</tr>
<tr>
<td>4.5 Static Analysis for the Identified ‘What-if’ Scenarios</td>
<td>40</td>
</tr>
<tr>
<td>4.6 Nuclear Data and Temperature Feedback Coefficients</td>
<td>61</td>
</tr>
<tr>
<td>4.7 Summary</td>
<td>67</td>
</tr>
<tr>
<td><strong>5 Modelling Transient Criticality</strong></td>
<td>69</td>
</tr>
<tr>
<td>5.1 The UCuRC Programme</td>
<td>69</td>
</tr>
<tr>
<td>5.2 Extending the QSS Model for Changing Geometry</td>
<td>71</td>
</tr>
<tr>
<td>5.3 Specification for Further Transient Calculations</td>
<td>72</td>
</tr>
<tr>
<td>5.4 Requirements for Model Inputs</td>
<td>78</td>
</tr>
<tr>
<td>5.5 Summary</td>
<td>79</td>
</tr>
<tr>
<td><strong>6 Criticality Consequence Analysis</strong></td>
<td>81</td>
</tr>
<tr>
<td>6.1 Model Outputs</td>
<td>81</td>
</tr>
<tr>
<td>6.2 The UCuRC Programme</td>
<td>83</td>
</tr>
<tr>
<td>6.3 Further Criticality Consequence Analysis</td>
<td>87</td>
</tr>
<tr>
<td>6.4 Summary</td>
<td>101</td>
</tr>
</tbody>
</table>
Summary and Conclusions 103

7.1 Summary 103
7.2 Conclusions 104

Acknowledgements 106

References 107

Appendices 113

Appendix A Material Properties for Static and Transient Criticality Analysis 115
Appendix B Reactivity Functions for the Transient Criticality Models 125
Appendix C Summary of Assumptions 135
Glossary of Terminology

‘What-if’ criticality scenario: An assumed sequence of events whereby, within a localised volume of a GDF or the surrounding host rock, a critical configuration of fissile materials is reached.

Static criticality calculations: The use of criticality software to determine the neutron multiplication factor $k_{\text{effective}}$.

Hypothetical criticality event: A specific example for a ‘what-if’ scenario whereby a critical configuration is selected for criticality consequence analysis.

Transient criticality models: The QSS model, RTM and Bounding Approach models.

Criticality consequence analysis: Use of the transient criticality models to understand the local consequences of hypothetical transient criticality events.

Acronyms and Names

<table>
<thead>
<tr>
<th>Expression or Name</th>
<th>Definition / Explanation</th>
</tr>
</thead>
<tbody>
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<td>AGR</td>
<td>Advanced Gas-cooled Reactor</td>
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<tr>
<td>Bounding Approach</td>
<td>A simplified version of RTM to understand the consequences of RT criticality</td>
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<tr>
<td>CSSR</td>
<td>Criticality Safety Status Report</td>
</tr>
<tr>
<td>DICE</td>
<td>A nuclear data library for MONK</td>
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<tr>
<td>DNLEU</td>
<td>Depleted, Natural and Low Enriched Uranium</td>
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<tr>
<td>DSSC</td>
<td>Disposal System Safety Case</td>
</tr>
<tr>
<td>DU</td>
<td>Depleted Uranium</td>
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<tr>
<td>ENDF/B</td>
<td>Evaluated nuclear data files used to construct data libraries for criticality software</td>
</tr>
<tr>
<td>EoS</td>
<td>Equation of State. Required for use of RTM</td>
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<td>ESC</td>
<td>Environmental Safety Case</td>
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<td>FEPs</td>
<td>Features, Events and Processes</td>
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<tr>
<td>FETCH</td>
<td>A coupled fluid dynamics and reactor physics code developed by Imperial College</td>
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<tr>
<td>GDF</td>
<td>Geological Disposal Facility</td>
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<tr>
<td>GRA</td>
<td>Guidance on Requirements for Authorisation</td>
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<tr>
<td>GWd/Te</td>
<td>Gigawatt Days per Tonne. Used to measure the burn-up (irradiation) of reactor fuel</td>
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<td>HEU</td>
<td>Highly Enriched Uranium</td>
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<td>HLW</td>
<td>High Level Waste</td>
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<tr>
<td>ICASPA</td>
<td>A goal seeking algorithm used with MONK to generate criticality maps</td>
</tr>
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<td>ILW</td>
<td>Intermediate Level Waste</td>
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<tr>
<td>JEF/TEFF</td>
<td>Evaluated nuclear data files used to construct data libraries for criticality software</td>
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<tr>
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<td>Definition / Explanation</td>
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</tr>
<tr>
<td>$k_{\text{effective}}$</td>
<td>Neutron multiplication factor. The ratio of the number of neutrons resulting from fission in one generation to the number lost by absorption and leakage in the preceding generation. If the value is less than unity a system is sub-critical. A value of unity shows a just-critical system. A value greater than unity means the system is super-critical.</td>
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<tr>
<td>$k_{\text{infinity}}$</td>
<td>For a system of infinite extent, where there is no leakage of neutrons, $k_{\text{infinity}}$ is the ratio of the number of neutrons resulting from fission in one generation to the number absorbed in the preceding generation.</td>
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<td>LEU</td>
<td>Low Enriched Uranium</td>
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<tr>
<td>LLW</td>
<td>Low Level Waste</td>
</tr>
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<td>LLWR</td>
<td>Low Level Waste Repository</td>
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<tr>
<td>mNiles</td>
<td>milliNiles. Another term for pcm.</td>
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<td>MONK®</td>
<td>Criticality software developed and licensed by AMEC</td>
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<tr>
<td>MRWS</td>
<td>Managing Radioactive Waste Safely</td>
</tr>
<tr>
<td>NDA</td>
<td>Nuclear Decommissioning Authority</td>
</tr>
<tr>
<td>NESR</td>
<td>Near-field Evolution Status Report</td>
</tr>
<tr>
<td>NRVB</td>
<td>Nirex Reference Vault Backfill</td>
</tr>
<tr>
<td>NU</td>
<td>Natural Uranium</td>
</tr>
<tr>
<td>pcm</td>
<td>Per cent mille. A unit for measuring the reactivity (or changes in reactivity) of a system</td>
</tr>
<tr>
<td>PCSA</td>
<td>Post-closure Safety Assessment</td>
</tr>
<tr>
<td>PESR</td>
<td>Package Evolution Status Report</td>
</tr>
<tr>
<td>PFR</td>
<td>Prototype Fast Reactor</td>
</tr>
<tr>
<td>PWR</td>
<td>Pressurised Water Reactor</td>
</tr>
<tr>
<td>QSS</td>
<td>Quasi-Steady-State. Used for both quasi-steady-state criticality transients and the QSS model developed to understand the consequences of such transients</td>
</tr>
<tr>
<td>Reactivity</td>
<td>Related to $k_{\text{effective}}$. It is calculated as $10^5 (k_{\text{effective}}^{-1} - 1)/k_{\text{effective}}$ in units of pcm (or mNiles). Super-critical systems have positive reactivity, just-critical systems have zero reactivity, and sub-critical systems have negative reactivity.</td>
</tr>
<tr>
<td>RT</td>
<td>Rapid Transient</td>
</tr>
<tr>
<td>RTM</td>
<td>Rapid Transient Model used to understand the consequences of RT criticality</td>
</tr>
<tr>
<td>RWMD</td>
<td>Radioactive Waste Management Directorate. Formed when Nirex was subsumed into the NDA</td>
</tr>
<tr>
<td>SF</td>
<td>Spent Fuel</td>
</tr>
<tr>
<td>TRU</td>
<td>Transuranic</td>
</tr>
<tr>
<td>UCuRC</td>
<td>Understanding Criticality under Repository Conditions. Repository was a term previously used for a GDF.</td>
</tr>
<tr>
<td>WIMS</td>
<td>Criticality and Reactor Physics software developed and licensed by AMEC</td>
</tr>
<tr>
<td>WIPP</td>
<td>Waste Isolation Pilot Plant (in the United States)</td>
</tr>
<tr>
<td>WVP</td>
<td>Waste Vitrification Plant</td>
</tr>
</tbody>
</table>
1 Introduction

1.1 Background

In 2001 the UK Government and devolved administrations initiated the Managing Radioactive Waste Safely (MRWS) programme with the aim of finding a practicable solution for the UK’s higher activity radioactive wastes.

After a period of consultation, the Government published a White Paper in June 2008 that lays out the framework for inviting communities to express an interest in opening up discussions, without commitment, on the possibility of hosting a geological disposal facility (GDF) at some point in the future [1]. The Nuclear Decommissioning Authority (NDA) is supporting this process, but the Government will be responsible for initial dialogue with potential host communities. The NDA has responsibility for planning, building and operating a GDF in the future.

The NDA programme to implement geological disposal is framed by the voluntarism and partnership approach to site a GDF. To support this approach a long-term project has been developed for implementing geological disposal [2], the phases of which are in line with the stages in the site selection process as set out in the Government’s White paper. The Government is currently (2013) reviewing the siting process for implementing geological disposal. NDA RWMD’s programme will be updated to reflect any changes made.

1.2 Criticality Safety

The UK has accumulated a legacy of radioactive waste from electricity generation, defence activities and other industrial, medical, agricultural and research activities. Radioactive wastes continue to be produced from these activities. Some of these wastes will remain hazardous for hundreds of thousands of years.

The wastes within a GDF will include a wide range of materials and radionuclides. Some radionuclides are fissile, i.e. they can cause a chain reaction (criticality) with slow neutrons, which have thermal energies, assuming that sufficient mass of the radionuclides is present in the right geometry and conditions. The most significant fissile radionuclides in the waste inventory are the plutonium isotope $^{239}$Pu and the uranium isotope $^{235}$U.

The Radioactive Waste Management Directorate (RWMD) within the NDA has undertaken research over a number of years to ensure criticality safety at all stages of radioactive waste disposal in support of the GDF programme. The demonstration of criticality safety is required for the Disposal System Safety Case (DSSC) [3] to meet regulatory requirements for a GDF [4]. Specific drivers for criticality safety research and development [5] are:

- to demonstrate and assure sub-criticality in the design of transport and disposal containers for Spent Fuel (SF), Intermediate Level Waste (ILW), High Level Waste (HLW), plutonium and uranium;
- to inform the design, loading and choice of wasteforms and containers in strategic studies for plutonium and uranium that input to Government policy;
- to provide data and understanding on the likelihood of post-closure criticality events for the DSSC, to demonstrate that criticality safety in a GDF is ensured; and
- to provide data and understanding on the consequences of hypothetical post-closure criticality events for the DSSC, to demonstrate that criticality safety in a GDF is ensured.

Work on the topic of criticality safety is divided into three subtopics: design for criticality safety, likelihood of criticality and consequences of hypothetical criticality, which are aligned with the above drivers [5].
1.3 Post-closure Criticality

1.3.1 Overview

For the final two bullet points in Section 1.2 above, which both refer to the post-closure phase of a GDF, there is specific regulatory guidance. In particular, the Environment Agency Guidance on Requirements for Authorisation (GRA) for a GDF [4] requires 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.” In addition, the guidance states that the “environmental safety case should also investigate, as a ‘what-if’ scenario, the impact of a postulated criticality event on the performance of the disposal system”.

This document refers to research undertaken for the consequences of hypothetical criticality subtopic, covering the final bullet point in Section 1.2. Through analysis of the consequences of hypothetical criticality the aim is not only to satisfy the requirement of undertaking calculations for ‘what-if’ scenarios, but also to demonstrate that even if a criticality event were to occur, the consequences would not be a significant concern for post-closure performance of the GDF. Research into the likelihood of criticality has also been undertaken to address the requirements of the GRA through probabilistic analysis. By demonstrating that post-closure criticality is both low likelihood and low consequence the requirements of the GRA will clearly be met.

The Criticality Safety Status Report (CSSR) [6], produced in 2010, describes the status at that time of research and development on criticality safety and summarises the evidence and understanding supporting the safety arguments made in the generic DSSC [3]. Arguments on the consequences of criticality post-closure are also applied in the Environmental Safety Case (ESC) [7] and Post-closure Safety Assessment (PCSA) [8].

Engineering measures are available to prevent criticality for such time as the waste packaging affords a high level of containment [6]. In particular, limits can be established on the amount of fissile material that can be incorporated into individual waste packages. These limits will ensure that a criticality cannot occur during any credible configuration of waste packages and conditions that could occur during waste storage, transport and emplacement in a GDF.

In the long term, however, after deterioration of the physical containment provided by the waste packages [9], there would be the possibility of either:

- movement of fissile material out of the packages [10] and subsequent accumulation into new configurations; or
- internal changes to waste package contents, that could in principle lead to criticality.

It is conceivable that a criticality could adversely affect the performance of a GDF after closure because, for example, the heat that would be produced could detrimentally affect components of the engineered barrier system.

1.3.2 Modelling the Consequences of Hypothetical Criticality

This report is part of a programme entitled ‘Modelling of Consequences of Hypothetical Criticality’ which builds upon previous research, including a body of work within the Understanding Criticality under Repository Conditions (UCuRC) research programme undertaken from 2002 to 2010 [11-27], where computer models of the evolution of hypothetical critical events were developed.

Whether a system involving fissile materials is critical is determined by the neutron multiplication factor, \(k_{\text{effective}}\). If \(k_{\text{effective}}\) is less than unity the system is sub-critical. If \(k_{\text{effective}}\) is equal to unity the system is just-critical (a neutron chain reaction is just possible), and if greater than unity the system is super-critical. A closely related mathematical quantity is reactivity, defined in units of mNiles to be:
\[ \rho = \frac{10^5 (k_{\text{effective}} - 1)}{k_{\text{effective}}} \]  

(1.1)

Should a hypothetical critical configuration \( k_{\text{effective}} \geq 1 \) or \( \rho \geq 0 \) arise within a GDF, the potential consequences are strongly dependent on how the critical configuration might evolve. The nature of the criticality event can be determined by the effects of temperature on system reactivity (reactivity increases as \( k_{\text{effective}} \) increases). If an increase in temperature leads to a decrease in reactivity then with some compensating mechanism to insert reactivity, such as the continued accumulation of fissile material, a just-critical system may be maintained. This is known as a quasi-steady-state (QSS) criticality, which is typically a long-lived, low-power event. If an increase in temperature leads to an increase in reactivity then a rapid transient (RT) criticality is possible, whereby there is a short duration release of energy followed by a return to sub-critical conditions given sufficient expansion and/or increase in temperature.

Under the UCuRC programme computer models were developed to understand the potential consequences of hypothetical criticality events for both QSS and RT scenarios. Calculations using these models are referred to as criticality consequence calculations. At Imperial College the three dimensional finite element model, FETCH, underwent a number of developments to include some extra physics for application to GDF scenarios, specifically for hypothetical rapid transient events. At AMEC (formerly Serco Technical Services) transient criticality models were also developed. These are intended as quick-running tools suitable for scoping calculations (e.g. parametric surveys) of hypothetical post-closure criticality events. Originally two models were developed; known as the QSS model (for QSS criticality transients) and the RTM (rapid transient model for RT criticality transients). Some calculations using RTM were benchmarked against FETCH. For RT criticality transients a Bounding Approach model was subsequently developed as a simpler alternative to RTM. User guides are available for the QSS model [28] and RTM and the Bounding Approach model [29].

During the UCuRC programme, the application of the models predominantly focused on criticality scenarios for the storage of ILW in a GDF in a higher strength rock disposal concept. The UCuRC studies undertaken to develop, apply and build confidence in the models have been well documented on the RWMD bibliography [11-27]. In addition some of the research has been published more widely [30-34]. Key documents from the UCuRC programme include two suites of criticality consequence calculations using the QSS model and RTM [22,23], a review of the development status of the transient models [25], and calculations using the Bounding Approach model [27].

### Programme Objectives

Overall, the programme to understand the consequences of hypothetical criticality aims to develop, document and apply the transient criticality models to a range of ‘what-if’ criticality scenarios. This effectively provides a ‘toolkit’ that RWMD can use, and if necessary modify, at a later date to cover new package designs and facility designs or concepts as the GDF programme progresses.

Following the UCuRC programme a further research contract was placed with Serco Technical Services (subsequently novated to AMEC) to continue studies into the consequences of hypothetical post-closure criticality events. The specific programme objectives were to:

1. document and supply the QSS model, RTM and the Bounding Approach model, developed under the UCuRC programme so that they may be used by RWMD and its contractors (to investigate the consequences of criticality in other materials and concepts) and by other stakeholders;

2. apply the UCuRC QSS model and Bounding Approach model to scope the consequences of criticality transients in other materials and illustrative concepts (the models need to be developed and applied sufficiently to meet safety case and stakeholder requirements for the current preparatory studies phase).
3. bring the UCuRC programme to a documented conclusion, through the above scoping calculations and review of the previous documentation;
4. recommend how the associated models and understanding should be developed and applied in future, beyond the preparatory studies phase.

The programme roadmap identified how these objectives would be met, and how the research would be reported. For the first objective two user guides have been published [28, 29]. For the remaining objectives three reports have been written:

- Two reports (of which this is one) are detailed technical reports which address objectives 2 and 3. They report selected results and conclusions from the UCuRC programme and the extended application of the transient criticality models for a wider range of scenarios and disposal concepts. This report summarises criticality consequence analysis undertaken for HLW, SF, plutonium and Highly Enriched Uranium (HEU). The second technical report [35] summarises criticality consequence analysis undertaken for ILW, Low Level Waste (LLW) and Depleted, Natural and Low Enriched Uranium (DNLEU);
- The third report is an overall programme synthesis report [36], summarising all activities undertaken for the extended (post-UCuRC) programme, and hence summarising the progress against all four programme objectives.

Overall, the three reports summarise the current analysis of the consequences of hypothetical criticality events, including a significantly extended range of scoping transient criticality calculations compared with the UCuRC programme.

A similar set of documents is available for research undertaken into understanding the likelihood of post-closure criticality [37, 38, 39].

1.4 Report Overview

The contents of this report, summarising the consequences of hypothetical post-closure criticality for the disposal of HLW, SF, plutonium and HEU within a GDF, are as follows:

- Section 2: Overview of waste packages and disposal concepts and post-closure evolution;
- Section 3: Scenarios for hypothetical criticality;
- Section 4: Static criticality analysis;
- Section 5: Modelling options for transient criticality;
- Section 6: Criticality consequence calculations;
- Section 7: Summary and Conclusions.

Appendices are used to provide additional details as required. Selected analysis and results from the UCuRC programme are included in this report to illustrate key results or trends. For further information the relevant references are provided in the text.

1 Where scenarios are described as ‘conceivable’ in this report it is not intended to imply that they could happen, but that the assumed set of events which lead to the scenario have some physical basis, even if unlikely to occur.
2 Overview of Waste Packages, Disposal Concepts and Post-closure Evolution

This section provides an overview of the waste packaging and illustrative disposal concepts within the DSSC [3] relevant to the disposal of HLW, SF, plutonium and HEU, together with an overview of information on the volumes of radioactive waste for disposal and the presence of fissile isotopes within those wastes. The latter is taken from the 2007 Radioactive Waste Inventory [40], together with further development of the derived inventory for the relevant waste types [41,42,43]. All analysis of the consequences of criticality summarised in this report are bounded by the 2007 Radioactive Waste Inventory and the assumptions of the 2010 DSSC [3].

Most potential ‘sources’ of HLW, SF, plutonium and HEU from nuclear reactors and other nuclear facilities have not formally been declared as wastes for geological disposal. However, they may be declared as wastes in the future, and are therefore considered as such for the research reported here.

The emplacement of any waste package into a GDF will be significantly sub-critical (i.e. the neutron multiplication constant $k_{\text{effective}}$ will be much lower than unity), as assured by the package design, transport and operational controls of a GDF [6]. However, given sufficient post-closure evolution of a GDF, it is conceivable that critical systems could develop. This section also provides an overview of post-closure evolution, and how that evolution could, hypothetically, lead to scenarios where critical configurations of fissile material could develop. The ‘what-if’ scenarios considered for consequence analysis are described further in Section 3.

It is emphasised that for any such scenarios to occur there would need to be substantial changes to one or more criticality controls (such as fissile mass, fissile concentration, moderation, or geometry). The expectation is therefore that all ‘what-if’ scenarios will be unlikely. Furthermore, for any scenario it is expected that the greater the changes required for a critical system to develop (e.g. the greater the fissile mass), the less likely that scenario becomes.

Hypothetical criticality scenarios have also been considered for analysis of the likelihood of criticality and are reported in [38,39]. It is noted that in order to undertake calculations to estimate the likelihood of criticality, more detailed information is required on wasteform evolution, package design and evolution, the waste inventory and the Feature, Events and Processes (FEPs) from which the scenarios are constructed. The likelihood of criticality reports [38,39] therefore provide more detailed discussions of the information summarised here.

2.1 Waste Packaging Concepts

2.1.1 Background

HLW is a by-product from the reprocessing of spent nuclear fuel. Most HLW for geological disposal is assumed to be a uniform and consistent solid wasteform, whereby the initial highly radioactive liquid form will have been made ‘passively safe’ by converting the liquid into solid using the vitrification treatment process [1]. The solid HLW is packaged into Waste Vitrification Plant (WVP) canisters. Current Government policy is that vitrified HLW should be stored for at least 50 years before disposal to allow short-lived radionuclides to decay so that the waste will become cooler, which will make its transport and disposal easier.

SF is not currently classified as a waste because it contains large amounts of uranium and some plutonium that can be recovered through reprocessing and used to make new fuel. Most of the UK’s SF from civil reactors has been reprocessed, producing separated plutonium and uranium (with HLW, ILW and LLW as waste by-products). If SF reprocessing was not undertaken, it could be packaged and disposed of in a GDF instead.
Separated plutonium and uranium are both products of spent fuel reprocessing. Enriched uranium is also produced by increasing the fraction of $^{235}$U (and thereby decreasing the fraction of $^{238}$U) in uranium extracted from uranium ore. Neither plutonium nor uranium are currently classified as wastes because they could be used to make new fuel. However, if it was decided that these materials had no further use, they could be packaged and disposed of in a GDF.

All wastes for geological disposal must be packaged in a safe form for storage, transport, and underground emplacement. There have been no definite decisions on the packaging of HLW, SF, plutonium and HEU for geological disposal. Research is ongoing to consider whether packages can be designed for use with several of these wastes, thereby reducing the number of package designs for safety analysis and manufacture. For the research presented here packaging assumptions have been based on RMWD’s three illustrative geological disposal concepts, as discussed in the 2010 DSSC [3], and summarised in Section 2.3.2 below.

For the illustrative packaging concepts presented below it is emphasised that no decisions have been made on actual package designs, and no HLW, SF, plutonium or HEU packages have been produced for geological disposal. As a result, the packaging concepts summarised below are subject to change. For example, while the descriptions and illustrative figures in Section 2.1.2 show an outer copper canister for packages, the material could be different, with options including stainless steel, titanium, carbon steel, and nickel-based alloys, for example. The material used could differ between different waste types and disposal concepts.

### 2.1.2 HLW and Spent Fuel Packaging Concepts

The material and design for the disposal containers has not been selected, although functional and technical requirements have been reported [44]. For the 2010 DSSC [3] HLW and SF are assumed to be disposed of in large cylindrical waste packages with a diameter of 0.9 m for all waste types, but a length of between about 2.5 and 4.5 metres, dependent on the waste type. Copper and steel canister examples were considered in the DSSC, but other designs, such as large multi-purpose canisters, may be possible.

Illustrative package designs for HLW and SF, as considered in the 2010 DSSC, were based on those from [42], which includes dimensional information for copper disposal canisters.

Figure 2.1 shows the package concept for HLW in a copper disposal canister. The package contains two (inner) stainless steel canisters, each of which has an external diameter of 0.43 m and a length of 1.34 m. The inner canisters contain HLW in a solid glass form, from the vitrification process. These two canisters are surrounded by an iron insert of thickness 0.185 m (from radius 0.215 m to 0.4 m) within the outer copper canister.

For spent fuel, two major sources within the UK are Advanced Gas-cooled Reactor (AGR) and Pressurised Water Reactor (PWR) fuel.

It is assumed that AGR fuel assemblies would be dismantled and that the graphite sleeves, support grids, and braces, etc. would be processed separately as ILW [42]. The remaining fuel pins would be consolidated in bundles within a can. A total of eight consolidated fuel bundles, equivalent to the pins from 24 AGR fuel elements, would be packaged inside a copper disposal canister in two layers with four cans in each layer. This is illustrated in Figure 2.2. The assumed disposal canister would have an external diameter of 0.9 m and a length of 2.5 m.

For PWR fuel it is assumed that the fuel assemblies would not be dismantled before packaging, and that four intact fuel assemblies would be packaged within a single disposal canister within four compartments of square cross-section [42]. This is shown in Figure 2.3. The assumed disposal canister would have an external diameter of 0.9 m and a length of 4.5 m.
Figure 2.1: The copper disposal canister, as an example of a HLW packaging concept [42]

![Copper disposal canister diagram](image1)

Figure 2.2: The copper disposal canister, as an example of a packaging concept for spent AGR fuel [42].

![AGR fuel bundle diagram](image2)
AGR and PWR fuels are only two of several radioactive fuels that could require geological disposal. Other fuels, and potential packaging concepts for them, are discussed in [42]. In overview, the concepts for packaging of different spent fuels are [42]:

- For spent Magnox fuel, it is assumed that 17 intact Magnox fuel elements would be packaged inside a WVP type canister, but with different dimensions (1.2 m long x 0.46 m diameter) to that used for HLW. Two such canisters would be placed in a disposal canister having the same dimensions as that used for the packaging of HLW so that the cast iron insert would need an adjusted geometry.

- For spent Prototype Fast Reactor (PFR) fuel from Dounreay the current assumption is that seven PFR sub-assemblies could be accommodated within a disposal canister, whilst allowing a thickness of cast iron around the sub-assemblies comparable to that assumed for AGR and PWR fuels. The canister is assumed to have an external diameter of 0.9 metres and a length of 3 metres.

- For spent submarine fuel it is assumed that the copper canister package design proposed for spent PWR fuel would be used for submarine fuel, with a loading of 1.4 tonnes of uranium per disposal canister. In reality the length of the disposal canister and the geometry of the insert would be designed to accommodate the dimensions of the fuel.

- There are a number of other miscellaneous spent fuels at Sellafield, from decommissioned research reactors, for example. There is currently insufficient detailed information to allow packaging assumptions to be developed for these fuels [42]. For the Derived Inventory, it was assumed that these spent fuels would be packaged using the same concept as that proposed for spent Magnox fuel [42].
2.1.3 Plutonium and Uranium Packaging Concepts

The development of a disposal packaging concept for plutonium and uranium wastes is at an early stage. For the preparation of the Derived Inventory [43], assumptions were made about both the wasteforms and the waste containers. These assumptions were also used in the 2010 DSSC [3], and are summarised below.

Plutonium and uranium (predominantly considered to be HEU) are assumed to be immobilised in a titanium-based ceramic and loaded into stainless steel cans which in turn would be encapsulated in glass within a large steel canister. This is known as the can-in-canister approach. These canisters would then be loaded into disposal canisters of the same design as for HLW and SF.

The can-in-canister approach to packaging pucks of plutonium and HEU is illustrated schematically in Figure 2.4. The approach was developed in the USA [45] for the packaging of plutonium in ceramic together with HLW in glass. For the packaging of plutonium and HEU in the UK it is assumed that the HLW glass would be replaced with inactive borosilicate glass. A nominal 10 wt% of Pu (~50 g) would be immobilised in a titanate-based ceramic to form a flat cylindrical puck, with a 6.9 cm diameter and thickness 2.5 cm [43].

Twenty pucks would be loaded into stainless steel cans, each 7.6 cm in diameter and 51 cm in length [43]. Multiple cans of pucks would be encapsulated in glass within a large canister as shown in Figure 2.4. The large canister is a steel cylinder 61 cm in diameter and 3.06 m in height and would contain 280 kg of ceramic pucks (equivalent to 33.3 kg of PuO$_2$ or UO$_2$) [43].

The dimensions of the steel canister are such that a single canister would be loaded into an outer canister to complete the waste package for geological disposal. It is assumed to be similar to the disposal canisters considered for HLW and SF (see Section 2.1.2 above).

Figure 2.4: The can-in-canister packaging concept for plutonium and HEU disposal [43,45]. Diagrams a) to c) show different sections of the packaging concept. Diagram a) is a slice through the package showing an arrangement of seven cans surrounded by borosilicate glass (green). In Diagrams b) and c) the borosilicate glass is not shown. Diagram b) shows a cut-away of the package and the different layers of cans. Diagram c) shows the ceramic pucks within the inner cans.
2.2 Waste Inventory

HLW, SF, plutonium and HEU represent a potentially significant amount of radioactive waste for geological disposal. From the 2007 Radioactive Waste Inventory [40], together with further development of the derived inventory for the relevant waste types [41,42,43], the activity of fissile nuclides within the different waste types can be estimated, both currently and in the future. Converting the activity to mass for each nuclide provides an estimate of the mass of fissile materials that could be disposed of in a GDF.

The HLW and SF Derived Inventory reference case is based on assumptions in the 2007 UK radioactive waste inventory [40] and considers UK HLW and spent AGR and PWR (Sizewell B) fuels only [42] - new build wastes, and additional civil and MoD submarine fuels are not included.

The Pu and HEU inventory is subject to greater uncertainty than the HLW and SF inventory, because information about some of the potential waste streams is not made public, and what information is published is generally not as detailed. The uranium and plutonium Derived Inventory reference case [43] is equivalent to the Baseline Inventory in the 2008 White Paper [1], and uses total masses from the 2007 UK Radioactive Waste Inventory [40]. It considers plutonium and uranium from civil programmes only [43] and therefore does not include nuclear materials owned by the MoD or “small users” such as universities and research establishments.

Concentrating on the two fissile isotopes of $^{235}$U and $^{239}$Pu, Table 2.1 summarises the anticipated masses present within different wastes in the year 2150, based on the assumptions of inventory as in [42,43]. In Table 2.1 the conditioned volume is that of the waste itself, and its primary containment. Therefore, the outer copper canister and cast iron insert (for the examples in Figures 2.1 to 2.3) are not included in the conditioned volume.

The two fissile isotopes $^{235}$U and $^{239}$Pu will not be the only isotopes in the waste packages. Neutron absorbing isotopes such as $^{238}$U may also be present, for example, so that the effective enrichment of the fissile nuclides (i.e. the weight fraction of uranium and plutonium isotopes that are fissile) is reduced. For different potential wastes more information on the isotopic composition is summarised in [38]. The summary tables in [38] show that the effective enrichment could be nearly 100% for some plutonium and HEU wastes, whereas spent fuel generally has a low effective enrichment of about 1% on average for both AGR and PWR. This is because for both AGRs and PWRs the fresh fuel, as emplaced into a nuclear reactor, is uranium with a maximum enrichment of about 5%. During residency in the reactor the fissile nuclide $^{235}$U will get burned up and, while there can be in-growth of $^{239}$Pu (from neutron capture in $^{238}$U), the net effect will be to reduce the effective enrichment. Unless a fault develops in the fuel, it is not normally removed from the reactor until the effective enrichment has reached such a level that fission is no longer economical under reactor conditions.

Table 2.1: The inventory of $^{235}$U and $^{239}$Pu for HLW, SF, plutonium and HEU in 2150. Information based on data from [38], which is derived from [42,43]$^2$.

<table>
<thead>
<tr>
<th>Waste</th>
<th>Total conditioned volume ($m^3$)</th>
<th>Total $^{235}$U content (kg)</th>
<th>Total $^{239}$Pu content (kg)</th>
<th>Number of waste packages</th>
</tr>
</thead>
<tbody>
<tr>
<td>AGR SF</td>
<td>1,933</td>
<td>24,248</td>
<td>14,474</td>
<td>5,341</td>
</tr>
<tr>
<td>PWR SF</td>
<td>492</td>
<td>4,788</td>
<td>8,908</td>
<td>655</td>
</tr>
<tr>
<td>HLW</td>
<td>1,422</td>
<td>10.8</td>
<td>88.0</td>
<td>3,656</td>
</tr>
<tr>
<td>Pu</td>
<td>3,049</td>
<td>302</td>
<td>66,043</td>
<td>3,428</td>
</tr>
<tr>
<td>HEU</td>
<td>44</td>
<td>1,395</td>
<td>0</td>
<td>50</td>
</tr>
</tbody>
</table>

$^2$ Tables 3.3 to 3.9 of [38] provide the information used.
Table 2.2 is based on the data in Table 2.1, and derives the average fissile concentration (in the conditioned waste volume), and the average fissile mass per waste package for the different waste types. For HLW the concentration and mass per package are much lower than for the other wastes at 0.07 kg/m$^3$ and 0.03 kg (per package), respectively. Even under idealised conditions of moderation in water in an idealised geometry this mass is not sufficient for a critical configuration. For SF, plutonium and HEU wastes the fissile concentrations and masses are larger. However, given the low effective enrichment for spent fuel, and the presence of added neutron absorbers in the plutonium and HEU wasteforms (together with very little void space in the packaging concept), the packages are, by design, significantly sub-critical. This is demonstrated through criticality calculations in Sections 4.5.2 (for PWR fuel) and 4.5.3 (for plutonium and HEU).

<table>
<thead>
<tr>
<th>Waste</th>
<th>Number of waste packages</th>
<th>Total conditioned volume (m$^3$)</th>
<th>Total $^{235}$U and $^{239}$Pu content (kg)</th>
<th>Average fissile concentration (kg/m$^3$)</th>
<th>Average fissile mass per waste package (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AGR SF</td>
<td>5,341</td>
<td>1,933</td>
<td>38,722</td>
<td>20.0</td>
<td>7.3</td>
</tr>
<tr>
<td>PWR SF</td>
<td>655</td>
<td>492</td>
<td>13,696</td>
<td>27.9</td>
<td>20.9</td>
</tr>
<tr>
<td>HLW</td>
<td>3,656</td>
<td>1,422</td>
<td>98.8</td>
<td>0.07</td>
<td>0.03</td>
</tr>
<tr>
<td>Pu</td>
<td>3,428</td>
<td>3,049</td>
<td>66,344</td>
<td>21.8</td>
<td>19.4</td>
</tr>
<tr>
<td>HEU</td>
<td>50</td>
<td>44</td>
<td>1,395</td>
<td>31.4</td>
<td>27.9</td>
</tr>
</tbody>
</table>

### 2.3 Illustrative Disposal Concepts

Because a site has not been chosen for a GDF the approach in the DSSC, and supporting research, has been to consider three illustrative disposal concepts. An overview of these is given in Section 2.3.2, while Section 2.3.1 below provides an overview of the multi-barrier approach which is common to all of the illustrative disposal concepts.

#### 2.3.1 The Multi-barrier Approach

The multiple barrier approach includes a number of engineered barriers and addresses two objectives with respect to providing safety - the isolation of the wastes and the containment of the radionuclides associated with the wastes [6]:

- **Isolation** means removing the waste from people and the surface environment. Geological disposal at depth in a suitable environment provides isolation by reducing the likelihood of inadvertent and unauthorised human interference and also provides shielding of the natural environment from direct radiation from the waste;
- **Containment** means retaining radionuclides within various parts of the multi-barrier system. For many radionuclides, disposal concepts can provide total containment until the radionuclides and their daughters decay to insignificant levels of radioactivity within the engineered barrier system. However, the engineered barriers in a disposal facility will degrade progressively over time and gradually lose their ability to provide containment. Further containment is provided by the geological barrier, which acts to delay the movement of any small amounts of long-lived radionuclides that are released from the engineered barrier system. Locating the geological disposal facility in a suitably deep and stable environment protects the engineered barriers, helping them to preserve their containment functions for longer times.
Both of these objectives are significant for post-closure criticality. By preventing or reducing the movement of fissile nuclides a criticality event is unlikely. For containment, the multi-barrier approach is significant in that, in the unlikely event of a criticality, it would help to prevent any effects of criticality from reaching the surface environment.

A schematic representation of a multiple barrier system is provided in Figure 2.5. More specific examples are presented in [3]. The main barriers are:

- **The wasteform**: This is the form into which the waste is conditioned to make it suitable for disposal.
- **The waste container**: The conditioned waste is placed in a container (sometimes called a canister), creating what is referred to as the waste package.
- **The buffer or backfill**: The buffer or backfill in this context refers to material that is placed immediately around emplaced waste containers in a disposal facility. In addition, other types of ‘mass backfill’ will be required to fill excavated access tunnels, shafts or drifts, together with sealing systems.
- **The geosphere**: The geosphere refers to the geological environment in which a GDF is constructed and provides a considerable barrier to the movement of radioactivity back to the accessible environment. In addition to providing isolation, this barrier protects the emplaced wastes from extreme changes that may take place at the Earth’s surface, as a result of either natural causes, as in the example of glaciations, or human actions.

In terms of criticality, the wasteform, buffer/backfill and geosphere all represent regions in which critical systems could hypothetically develop given sufficient time for systems to evolve from the conditions at emplacement.

**Figure 2.5: Schematic representation of a multiple barrier system [6]**
2.3.2 Illustrative Geologies

A wide range of geological settings could be suitable for hosting a GDF in the UK. Until a site is identified for a GDF, RWMD has chosen to define a limited number of generic geological settings encompassing typical, potentially suitable, UK geologies. This provides a manageable number of concepts to consider for assessments of safety [3].

The geological settings defined by RWMD are based on consideration of the host rock formation (where the disposal regions will be developed) and the cover rocks (the geological formations that occur between the disposal regions and the ground surface). Further information on these generic geological environments is provided in [2]. In brief they are:

- Higher strength rocks - these are typically crystalline igneous, metamorphic rocks or geologically older sedimentary rocks, where any fluid movement is predominantly through discontinuities in the rock. In the UCuRC studies these were often referred to as hard rocks; granite was used as a representative example.

- Lower strength sedimentary rocks - these are typically geologically younger sedimentary rocks, where any fluid movement is predominantly through the rock mass. In the UCuRC programme mudstone or clay was used as a representative example.

- Evaporites - these are typically anhydrite (anhydrous calcium sulphate), halite (rock salt) or other rocks that result from the evaporation of water from water bodies containing dissolved salts.

The illustrative geological disposal concepts, with examples, are listed in Table 2.3.

Table 2.3: Illustrative disposal concept examples [6].

<table>
<thead>
<tr>
<th>Host rock</th>
<th>Illustrative Geological Disposal Concept Examples$^*$</th>
<th>ILW/LLW</th>
<th>HLW/SF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Higher strength rocks$^*$</td>
<td>UK ILW/LLW Concept (NDA, UK)</td>
<td>KBS-3V Concept (SKB, Sweden)</td>
<td></td>
</tr>
<tr>
<td>Lower strength sedimentary rock$^*$</td>
<td>Opalinus Clay Concept (Nagra, Switzerland)</td>
<td>Opalinus Clay Concept (Nagra, Switzerland)</td>
<td></td>
</tr>
<tr>
<td>Evaporites$^*$</td>
<td>WIPP Bedded Salt Concept (US-DOE, USA)</td>
<td>Gorleben Salt Dome Concept (DBE-Technology, Germany)</td>
<td></td>
</tr>
</tbody>
</table>

Notes
a. Higher strength rocks – the UK ILW/LLW concept and KBS-3V concept for spent fuel were selected due to availability of information on these concepts for the UK context.
b. Lower strength sedimentary rocks – the Opalinus Clay concept for disposal of long-lived ILW, HLW and spent fuel was selected because a recent OECD Nuclear Energy Agency review regarded the Nagra (Switzerland) assessment of the concept as state of the art with respect to the level of knowledge available. However, it should be noted that there is similarly extensive information available for a concept that has been developed for implementation in Callovo-Oxfordian Clay by Andra (France), and which has also been accorded strong endorsement from international peer review. Although we will use the Opalinus Clay concept as the basis of the illustrative example, we will also draw on information from the Andra programme. In addition, we will draw on information from the Belgian super container concept, based on disposal of HLW and spent fuel in Boom Clay.
c. Evaporites – the concept for the disposal of transuranic wastes (TRU) (long-lived ILW) in a bedded salt host rock at the Waste Isolation Pilot Plant (WIPP) in New Mexico was selected because of the wealth of information available from this United States Environmental Protection Agency (EPA) certified, and operating facility. The concept for disposal of HLW and spent fuel in a salt dome host rock developed by DBE Technology (Germany) was selected due to the level of concept information available.
d. For planning purposes the illustrative concept for depleted, natural and low enriched uranium is assumed to be same as for ILW/LLW and for plutonium and highly enriched uranium is assumed to the same as for HLW/SF.
For the purpose of developing the illustrative designs, RWMD has assumed the following geological settings and GDF depths below ground level for the three host rock environments [46]:

- For the higher strength rock example, the disposal facility depth has been assumed to be 650 m and the host geology has been assumed to be overlain by a permeable sedimentary rock.
- For the lower strength sedimentary rock example, the disposal facility depth has been assumed to be 500 m and the host geology has been assumed to be overlain by a permeable sedimentary rock.
- For the evaporite rock example, the disposal facility depth has been assumed to be 650 m and the host geology has been assumed to be overlain by a sedimentary rock.

The layouts of the illustrative GDF designs for the three generic geological host rock environments are described in Section 2.3.3 below.

### 2.3.3 Illustrative GDF Layouts

The illustrative GDF designs are based on the assumption of a single facility to accommodate all of the wastes and materials in the Baseline Inventory [40]. In such a ‘co-located’ disposal facility it is assumed in the 2010 DSSC [3] that there would be two distinct disposal regions, one where a number of packages would be stored within vaults (known as the ILW/LLW region, although this could also include DNLEU) and one where waste packages would be stored in tunnels or individual deposition holes (known as the HLW/SF region, although this could also include plutonium and HEU). These would share surface facilities and access tunnels [46].

The disposal regions would be physically separated to ensure that potential thermal, mechanical, hydrogeological and chemical interactions do not compromise the key safety functions of the different engineered barrier components. A minimum horizontal separation distance of 500 m has been assumed between the two disposal regions [46].

The illustrative layouts for GDFs in higher strength rock, lower strength sedimentary rock and evaporite rock are shown in Figures 2.6 to 2.8. RWMD does not intend that one of the illustrative concepts is necessarily the one that would be used in the relevant geological setting - at this stage no geological disposal concept has been ruled out. In particular, the layout of each illustrative GDF is based on construction of the facility on a single level with idealised spacings between the packages within disposal vaults and tunnels and between disposal regions [46]. Alternative layouts could be developed for specific sites with specific host rock characteristics, potentially resulting in different spacing between waste packages, vaults and tunnels, and different separation distances between disposal regions, which could affect the size of the facility.

*Figure 2.6: Illustrative underground layout for a GDF accommodating the Baseline Inventory in a higher strength host rock [46]*.
Figure 2.7: Illustrative underground layout for a GDF accommodating the Baseline Inventory in a lower strength sedimentary host rock [46].

Figure 2.8: Illustrative underground layout for a GDF accommodating the Baseline Inventory in an evaporite host rock [46].
2.3.4 Illustrative Designs for Waste Package Disposal

In Figures 2.6 to 2.8 above, the deposition regions marked HLW/SF (the right hand side of each figure) are the disposal volumes for HLW, SF, plutonium and HEU. On the assumption that plutonium and uranium would be packaged in similar containers to those for HLW and SF, it is assumed that the illustrative layouts in Figures 2.6 to 2.8, and the further discussion below, apply to any HLW, SF, plutonium or HEU package. Hence, in Sections 2.3.4.1 to 2.3.4.3, references to the HLW/SF disposal region are also assumed to include plutonium and HEU wastes.

2.3.4.1 Higher Strength Rock

The illustrative design for the HLW/SF disposal region in a higher strength rock consists of disposal tunnels designed for vertical emplacement of individual disposal canisters in deposition holes, each surrounded by pre-compacted bentonite blocks and rings, as shown in Figure 2.9. Bentonite is a clay like material which swells when wet.

Each disposal tunnel would be 340 m long with an effective disposal length of 310 m and dimensions of 5.5 m wide by 5.5 m high [46]. Ten disposal tunnels would be grouped together to form a disposal area and the tunnels would be spaced at 25 m between their centres. It is assumed that each disposal tunnel would accommodate 48 deposition holes spaced at 6.5 m between their centres. Different depth holes would be drilled to accommodate the different length disposal canisters.

Before emplacement of any disposal canister, the selected deposition hole would be prepared with bentonite blocks and rings, comprising a base block and a number of rings (depending on the type of disposal canister) [46]. Following emplacement of the disposal canister, three further bentonite blocks would be placed in the deposition hole.

When all of the deposition holes in a tunnel have been filled, the disposal tunnel would be backfilled with a mixture of crushed rock (70%) and bentonite (30%) [46]. The backfill would also be used to infill unused or rejected deposition holes and the top 1.0 m of each deposition hole.

Figure 2.9: Longitudinal section through a HLW, SF, plutonium and uranium disposal tunnel in the higher strength rock disposal concept [46].
2.3.4.2 Lower Strength Sedimentary Rock

The illustrative design for the HLW/SF disposal region in a lower strength sedimentary rock consists of disposal tunnels designed for the horizontal disposal of individual disposal canisters on top of a layer of bentonite blocks. Figure 2.10 provides further details.

It is assumed that each disposal tunnel would be 800 m long and 2.5 m in diameter [46]. The entrance to each disposal tunnel (the first 20 m) would be larger at 5.6 m wide by 3.8 m high, to allow for the transfer of the disposal canister from the transport container onto bentonite blocks that would have been pre-placed onto a disposal trolley. The disposal tunnels would be constructed with 40 m between their centres.

Following emplacement of each disposal canister on a bed of bentonite blocks, a mobile bentonite hopper would be used to place pre-compacted bentonite pellets in the disposal tunnel. This would be repeated after each disposal canister, therefore providing progressive backfilling of the disposal tunnel. It is assumed each canister would be placed 3 m apart from the previous canister [46].

Additional low-permeability seals would be placed in the main disposal facility access ways (shafts and drift). Void space in the remaining tunnels and underground facility areas would be backfilled with a 30% bentonite and 70% sand mixture [46].

Figure 2.10: Schematic of a HLW, SF, plutonium and uranium disposal tunnel in a lower strength sedimentary host rock [46].

2.3.4.3 Evaporite Rock

The illustrative design for the HLW/SF disposal region in an evaporite rock consists of tunnels designed for horizontal emplacement of individual disposal canisters on the tunnel floor. Figure 2.11 provides further details.

It is assumed that each disposal tunnel is 800 m long and 4.5 m wide by 3.5 m high. The entrance to each disposal tunnel (the first 20 m) would be larger (6 m wide by 5.5 m high) to accommodate the transfer of the disposal canister from the transport container onto the disposal trolley. The disposal tunnels would be constructed with 40 m between their centres.

The area around the disposal canister would be filled with crushed rock salt immediately after emplacement of each disposal canister using a mobile hopper [46]. This cycle would be repeated until the disposal tunnel was filled with disposal canisters and crushed rock salt fills the remaining voids. It is assumed each canister would be placed 3 m apart from the previous canister [46].
Once all of the disposal canisters have been placed within a disposal tunnel, a tunnel seal would be constructed. The access tunnels would be in-filled with crushed rock salt, with periodic placement of concrete tunnel seals [46]. Multi-component shaft seals containing concrete, clay, asphalt and/or salt would provide barriers to fluid flow. The remaining tunnels, shafts and drift would be backfilled with crushed rock salt [46].

2.4 Overview of Post-closure Evolution

2.4.1 Package Evolution

Waste packages for HLW, SF, plutonium and HEU will be designed to ensure that their safety functions remain during the transport and operational stages of a GDF, and for a period post-closure. A key aim of the packages is to contain radionuclides for sufficient time to allow some radioactive decay to occur, and therefore for sufficient time that there is a significant reduction in the heat generated. As a result the waste containers are generally required to provide containment of the radionuclides for a period of between 1,000 and 10,000 years [9]. This will require high-integrity, sealed containers.

Whatever choice of material is made for the package containers they will, eventually, evolve due to processes such as corrosion and weld failure. A detailed review of research into package evolution, as of 2010, can be found in the Package Evolution Status Report (PESR) [9]. This includes descriptions of the understanding of both wasteform and package evolution, including a summary of research that has been undertaken for overseas disposal facilities. The PESR generally discusses packaging concepts for HLW and SF. It is assumed, however, that if plutonium and HEU were to be packaged in a similar way (at least for the outer containers) then the timescales for package evolution would be similar.

It is currently envisaged that HLW, SF, plutonium and HEU will be stored in ‘dry’ containers, so that only trace volumes of water may be present within packages, and hence internal corrosion from water would be negligible or have a very small effect. The use of dry wasteforms is significant for the prevention of critical configurations within a package since, for the fissile mass within any package, sufficient moderator (see Section 3.2.1.3) is required for any hypothetical critical configuration, and water acts as an efficient moderator. It is therefore expected that any hypothetical critical configuration could only occur if sufficient water were to enter packages, or the fissile materials were to be transported to a suitably moderated region of the GDF.
For the evolution of a GDF, it is generally assumed for the illustrative disposal concepts (Section 2.3) that the GDF will saturate with groundwater over a period of decades to thousands of years [47].

Although the GDF and buffer around emplaced packages may saturate with groundwater, the illustrative package designs have no direct route (e.g. vents within the container) through which water could enter following saturation. The only possibility for water to enter a package will be following a penetration through the container such as a failed weld, physical damage, or container corrosion. Corrosion rates would depend on the container material and the chemistry of groundwater. Given typical thicknesses of the containers (at least several centimetres), and typical corrosion rates of the order of 0.01 – 10 µm/year [9] (dependent on material), corrosion could act as a potential route for water ingress into a package over thousands of years or much longer. Damage could include that from the transport and operational stages of a GDF, or from post-closure geological activity.

Water ingress into packages leads to the possibilities of:

- Flooding of any void volumes within the package;
- Mobilisation of fissile and other materials within the packages and their subsequent rearrangement;
- Mobilisation of fissile materials out of packages and into the surroundings;
- Mobilisation of non-fissile materials out of packages and into the surroundings, leading to increased water in the package.

Apart from flooding, which could occur relatively quickly (given a sufficient penetration through the package container to any void volumes), the above are expected to be slow processes, requiring long timescales. Even in the disposal packages with larger void volumes that water could occupy (e.g. between the pins of PWR fuel elements), the mobilisation of fissile, or other, materials would require long timescales for significant corrosion and transportation.

In the package concepts for HLW, plutonium and HEU there is very little void space, so that significant internal changes would require cracking and dissolution of glass (the wasteform itself for HLW) and the packaging borosilicate glass for plutonium and HEU. For plutonium and HEU, cracking and dissolution of the ceramic wasteform would also be required if fissile nuclides were to be transported either within packages or to the surroundings. Glass and ceramic materials are chemically stable and can withstand a range of chemical environments without suffering significant degradation. Hence it is expected that the timescales for the ‘release’ of fissile (or other nuclides) is expected to be very slow. An illustrative example in [9] quotes about 70,000-700,000 years to dissolve 10 cm of glass, for example.

Given the saturation of a GDF, failure of one or more packages allowing water ingress, and a means by which the package contents could be re-distributed, it is conceivable (although considered unlikely) that critical configurations could occur. Any such configuration would require sufficient time for the system to evolve and sufficient fissile material to be available. Coupled with information on how the near-field may evolve in Section 2.4.2, Sections 2.5 and 3 discuss how GDF evolution could lead to ‘what-if’ scenarios for criticality.

2.4.2 Near-field Evolution

As discussed in Section 2.4.1, given sufficient time post-closure, the GDF is generally expected to saturate with groundwater. This provides a means for evolution of not only the waste package itself, but the surrounding buffer/backfill to the emplaced wastes. With this evolution comes the possibility for increased mobility of fissile nuclides, and hence the possibility, though unlikely, of a critical system developing.
This section provides a brief overview of how the near-field environment, including the buffer/backfill, could evolve post-closure. More information can be found in the Near-field Evolution Status Report (NESR) [10].

2.4.2.1 Higher Strength Rock

After emplacement of disposal packages into the disposal tunnels the bentonite buffer and tunnel backfill will begin to saturate, causing the bentonite to swell [10]. Saturation is expected to take a few decades to a few centuries [10], although individual deposition holes could saturate in a few years dependent on the local groundwater flow.

On saturation, the bentonite buffer would provide a low-permeability barrier around the disposal packages, hence restricting the groundwater flow in contact with the waste packages. However, if fluid flow through fractures intersecting deposition holes is sufficiently large, then the bentonite may be susceptible to piping and erosion [10]. These processes have the potential to remove significant amounts of bentonite from a deposition hole and, hence, reduce the ability of the buffer to protect the waste package. As far as possible, the intersection of features of such high groundwater flow would be avoided, or if a deposition hole intersected such a feature the deposition hole may not be used.

Heat generation in the initial period after disposal, resulting primarily from the decay of short-lived radionuclides, would increase the temperature of the bentonite buffer and the adjacent host rock by several tens of degrees above ambient. Temperatures would be expected to peak within a few decades after emplacement, but elevated temperatures could persist for about 10,000 years [10]. Elevated temperatures could reduce the capacity of the buffer to swell and could cause increases in groundwater flow in the buffer [10]. Increased temperatures could also impact the solubility of minerals and could influence corrosion rates, for example. However, assuming no defects or damage to a canister, it is expected to remain intact and prevent water from contacting the waste until after temperatures have returned to ambient [10].

Chemical reactions between groundwater and bentonite minerals may occur, including mineral dissolution and precipitation. These reactions are expected to result in chemical buffering of the bentonite pore water to a near-neutral or slightly alkaline pH [10]. In the long term, the capacity for chemical buffering of the bentonite by reaction with groundwater would diminish as the reactants in the bentonite are exhausted. Dependent on the chemical composition of the groundwater, the composition of the bentonite buffer might be altered which could affect the porosity and permeability of the buffer, to either inhibit or enhance groundwater flow.

Assuming a copper canister for the waste packages as in the illustrative figures in Section 2.1.2, the conditions in the near-field following saturation are expected to support long canister lifetimes [48]. While aggressive species such as sulphides and microbes could accelerate corrosion, the transport of these species to disposal canister surfaces will be limited by the low permeability of the bentonite. Similarly, the rate of migration of any corrosion products away from the canister will be slow [10]. Hence, taking the expected evolution of the near-field into account, copper canisters are expected to maintain their integrity and provide full radionuclide containment for the order of 100,000 years. This does not guarantee such a lifetime for all waste packages, however, since a small number of canisters could have undetected manufacturing defects, which could result in a reduced period of radionuclide containment. An assumed containment period of 10,000 years has been suggested for canisters with undetected manufacturing defects [10].

Water entry into the disposal canister could initiate dissolution of the wasteform and release of any radionuclides remaining at the time of canister failure, as discussed in Section 2.4.1 above. Should fissile materials be released from a waste package (or a number of waste packages) into the bentonite buffer then transport is expected to be diffusion dominated. In the higher permeability crushed rock and bentonite backfill region, or the surrounding rock, advective transport dominates.
2.4.2.2 Lower Strength Sedimentary Rock

The expected near-field evolution for the lower strength sedimentary rock illustrative disposal concept is similar to that for higher strength rock, in that the GDF is expected to saturate and that water could come into contact with packages, and ultimately ingress into them. There are a number of differences, however:

- GDF saturation is expected to take longer, with up to a few centuries to saturate the bentonite buffer surrounding the waste packages.
- The illustrative disposal concept assumes carbon steel canisters instead of copper canisters. The carbon steel would begin to corrode after emplacement. Initially, the corrosion rate could be relatively high under aerobic conditions, but occurs at a much slower rate under anaerobic conditions which would develop after saturation. The carbon steel disposal packages are expected to provide complete radionuclide containment for 10,000 years, or at least 1,000 years under worse-case conditions [9,10].
- Steel corrosion generates hydrogen, which could result in the formation of gas. The gas pressure may be sufficient for the gas to break through the bentonite, but such an event is not expected to compromise the buffer’s properties as a barrier to groundwater flow, because the bentonite would reseal after the gas breakthrough [10].
- Should fissile materials be released from a waste package (or a number of waste packages) then transport is expected to be diffusion dominated in the bentonite buffer, tunnel backfill and the host-rock.

2.4.2.3 Evaporite Rock

The near-field environment for a GDF located in an evaporite rock could evolve quite differently to those for higher strength rock and lower strength sedimentary rock. Firstly, evaporite rocks tend to have both low permeability and porosity. Any water tends to be brine trapped in the rock [10], rather than established groundwater flow. If possible, saturation of the salt backfill would be slow, taking several centuries, or more [10].

A key characteristic of many evaporites is the ability of the rock to creep, i.e. to flow plastically at relatively fast rates [10]. As a result of this characteristic, an evaporite host rock (or backfill derived from it, such as crushed salt) has the potential to seal the openings around and within a GDF. Over time this can provide a complete, uninterrupted barrier around waste packages, and ultimately the GDF.

This self-sealing behaviour means that any pathways for groundwater flow into the GDF are typically short-lived. Depending on the prevailing conditions creep would be expected to render the permeability of disturbed evaporite rocks similar to that of the undisturbed rock within approximately 200 years [10]. Where waste such as HLW and SF produce heat from radioactive decay following emplacement, the heat could reduce the time taken for creep to self-seal regions around the waste packages [10]. As a result of creep, an evaporite host rock may be able to provide a high degree of containment over the very long term.

For waste packages that generate significant heat, it is possible that thermal contraction could cause the evaporite to crack [10] as the heat output in the near-field decreases (after sufficient decay of the package contents). This provides a possible, but localised, route for groundwater flow to reach a package. However, based on the expected creep for the GDF as a whole, groundwater movement in the evaporite is expected to be extremely slow or even non-existent.

The illustrative disposal concept for evaporite rocks assumes carbon steel canisters. The canisters are expected to resist the mechanical effects of creep closure and maintain their integrity until processes such as corrosion have reduced their structural strength [10]. Given the lack of water in evaporite rocks, the disposal canisters are not expected to be significantly degraded until at least several thousands of years after disposal. Furthermore, even if the canisters were to corrode, the possibility of transport of fissile (or other) nuclides with groundwater, is expected to be very low.
being limited by the small amounts of pore water. If any materials were to be transported it is expected that diffusive processes would dominate [10].

2.5 FEPs for Hypothetical Post-closure Criticality

As discussed in Section 2.1, any waste package(s) emplaced into a GDF would have to evolve significantly for a critical configuration to develop. The consideration of relevant Features, Events and Processes (FEPs) is an accepted approach for identifying how a system could change due to environmental changes, chemical changes, human interaction, etc. From a combination of FEPs, ‘what-if’ scenarios can then be constructed. In this research programme (and the preceding UCuRC programme) and the Likelihood of Criticality programme, scenarios whereby a critical configuration of fissile materials could conceivably arise, are those of interest.

FEPs which could lead to a hypothetical critical configuration of fissile material are, to a significant extent, related to how the wasteform, waste container and backfill could evolve post-closure. Hence, the effects of saturation with groundwater and the subsequent possible changes to the integrity of the waste package and backfill, as described in Sections 2.4.1 and 2.4.2, are a key part of FEP analysis.

In the sections below a brief overview of FEP analysis from the Nirex and Likelihood of Criticality programmes is given. For research into understanding the consequences of hypothetical criticality, Section 3 describes a further independent and complementary method for defining ‘what-if’ criticality scenarios.

2.5.1 Nirex Programme

Previous consideration of criticality FEPs in relation to geological disposal of radioactive waste in the UK primarily considered ILW disposal in higher strength rock. The Nirex work programme reported in 1998 [49] considered the accumulation of fissile material at such time as the containment of the waste packaging has deteriorated after emplacement in a GDF. This work aimed to identify all FEPs which might influence the potential for, or the effects of, a criticality. Scenario-defining FEPs were identified and qualitative descriptions of GDF evolution for each scenario-defining FEP were developed.

As part of this work a formal discussion of FEPs in relation to post-closure criticality was held and reported [50], and a detailed set of scenario-defining FEPs for a criticality was described [51]. Four mechanisms, by which a criticality could occur were identified [49,51]:

- Relocation and concentration of dissolved fissile material – where material is dissolved in groundwater (and therefore present at the molecular level), relocated and concentrated by for example precipitation, co-precipitation, sorption or mineralisation.
- Relocation and concentration of colloidal fissile material – where colloidal material (colloids are small particles in the size range 1 nm to 1 µm) is dispersed in groundwater or on gas bubbles, relocated and concentrated by for example filtration or deposition.
- Relocation and concentration of particulate fissile material – where particles, of size greater than approximately 1 µm, are mobilised by the groundwater, relocated and concentrated by settling or filtration.
- Bulk movement of solid materials – where fissile material is concentrated through the movement of solid materials in a waste stack, for example, by the gravitational slumping of materials as the waste stack degrades over time.

It was these scenario-defining FEPs that were used for the studies of criticality consequences in the UCuRC programme. The first three, which relate to the slow accumulation of fissile material in a localised region of a GDF, were termed the ‘slow accumulation’ scenario. The final scenario was considered as a ‘stack slumping’ scenario.
2.5.2 Likelihood of Criticality Programme

For research into the Likelihood of Criticality it is important to construct scenarios from sequences of FEPs, and to understand the key physical processes governing those FEPs so that the GDF evolution, and likelihood of a critical system can be calculated. To support this, a detailed and independent review of FEPs has been undertaken for the disposal of HLW, SF, plutonium and HEU wastes [38,39]. In overview, the review in [38] describes two main scenarios for criticality as follows:

- **Within a package.** Following saturation, water enters a package through weld defects or openings caused by package corrosion or damage from displacement on a neighbouring fracture (geological activity). Package materials corrode and some may be removed from the package as corrosion products in the groundwater. Fissile nuclides become mobilised and remain in the package, mixed with water. Weakening of the degrading package and contents may then allow the remaining materials (including fissile) to collapse under gravity.

- **Outside of a package.** Following saturation, water enters one or more damaged waste packages through weld defects or openings caused by corrosion or geological activity. Fissile nuclides are released from the degrading packages in dissolved, colloidal or particulate form due to groundwater flow. The fissile materials migrate through the buffer, or potentially into the host rocks, until sorption, precipitation, mineralisation or settling occur in a localised volume, causing an accumulation of fissile material.

The scenario for accumulation outside of a package (e.g. in the buffer) could conceivably involve the fissile material from a number of degraded packages. In the Likelihood of Criticality research, the distinction between fissile accumulation from one or several packages is significant since the likelihood of package failure for a number of the high-integrity waste packages used for HLW, SF, plutonium and HEU is expected to be lower than that for single package failure [38]. For criticality consequence analysis the source of the fissile materials is less significant.
3 Scenarios to Scope the Consequences of Hypothetical Criticality

For research into understanding the consequences of hypothetical criticality events in a GDF post-closure the following definition is used:

- ‘What-if’ Criticality Scenario – an assumed sequence of events whereby, within a localised volume of a GDF or the surrounding host rock, a critical configuration of fissile materials is reached.

3.1 The UCuRC Programme

The UCuRC programme considered the two main scenarios for post-closure criticality from the Nirex Programme, as discussed in Section 2.5.1; slow accumulation and stack slumping. Three transient criticality models (see Section 1.3.2) were developed by AMEC (formerly Serco Technical Services) to estimate the consequences of hypothetical criticality events from the slow accumulation scenario, whereas the FETCH code, developed by Imperial College, was applied to both the slow accumulation and the stack slumping scenarios [52,53].

For the study of criticality consequences analysis for ‘what-if’ criticality scenarios, as recommended by the GRA [4], it is not essential to restrict attention to hypothetical criticality events with larger (but still expected to be low) likelihood; more extreme events, with very low likelihood, can help to demonstrate trends, for example. This does not, however, mean that ‘impossible’ calculations should be undertaken. For example, should a hypothetical criticality event require more fissile mass than would ever be disposed of, then this is not considered a useful calculation to undertake.

The approach of the UCuRC programme was to consider the consequences of post-closure criticality for a wide range of hypothetical criticality events (for ILW disposal in higher strength rock) [22,23,27]. In the accumulation scenario, for example, accumulations of up to one tonne of fissile material, either $^{235}$U or $^{239}$Pu in oxide form, were modelled. By undertaking calculations for a wide range of fissile masses a goal was to establish which (if any) hypothetical critical configurations could have a significant impact on the performance of a GDF. Undertaking a range of calculations also provided a better understanding of the processes that would control the nature and magnitude of hypothetical criticality events under the particular conditions of the Nirex phased geological repository concept.

Further consideration of the radioactive inventory, together with the disposal concepts and the expected timescales required for any critical system to develop, support arguments that it is not credible that hundreds of kilograms of $^{239}$Pu, or larger, could accumulate, as assumed in the UCuRC programme. Firstly, even though the inventory for HLW, SF, plutonium and HEU (Table 2.2) could include tens of tonnes of fissile materials (particularly from SF and plutonium, if declared as wastes), there would be no more than about 20 kg (on average) of $^{239}$Pu in any individual package. Such wastes, as packaged, would be conditioned to be significantly sub-critical via a number of criticality controls. In addition, the high-integrity packages have a low expected rate of failure over long timescales. For 100 kg of fissile material to be released the entire fissile contents of several (five or more) packages would have to be mobilised in groundwater and accumulate. Given both the expected package integrity and the separation between packages, this is not considered credible. Secondly, since the processes required for plutonium to be released from failed waste packages would act over times comparable to, or greater than, the half-life of $^{239}$Pu (24,100 years), much of the $^{239}$Pu would decay before the expected timescales for accumulation. Accumulations of larger masses of $^{235}$U are considered more credible than for $^{239}$Pu (but still unlikely) because the half-life of $^{235}$U is very large at over 700 million years. However, the contents from several packages would still be required for accumulations of the order of 100 kg of fissile

3 Here, not credible is where the probability is expected to be vanishingly small or zero.
material. Whether hypothetical accumulations include $^{235}$U or $^{239}$Pu is significant in how a criticality transient could develop, as shown in Section 4.6.2.

The above arguments do not render calculations for large masses invalid. Indeed, if it can be shown that the consequences of criticality do not have any significant impact on the performance of a GDF for even extreme criticality events (where the likelihood is expected to be vanishingly small), then this is a significant conclusion in demonstrating that post-closure criticality is not a significant concern. However, given that a number of such calculations were undertaken for the UCuRC programme, the aim for the extended research programme reported here was to focus criticality consequence analysis, as far as is practicable, on scenarios where the expected likelihood will be low but not zero.

### 3.2 Selection of ‘What-if’ Scenarios for Further Criticality Consequence Analysis

This section identifies the ‘what-if’ scenarios for post-closure criticality that were used as the basis for scoping consequence calculations in meeting programme objective 2 from Section 1.3.3.

To ensure the objective was met Section 3.2.1 presents an overview of how, hypothetically, a critical configuration could be reached. This is intended as a first principles consideration of criticality, and is an independent check that adequate scenarios are considered to meet the programme objectives. The overview is not intended as a repetition of the FEP analysis from the Likelihood of Criticality programme [38], but as complementary to it in the interests of completeness.

Based on the summary of how a critical configuration could develop, Section 3.2.2 presents the details of which scenarios were taken forward to scope the consequences of hypothetical post-closure criticality events for HLW, SF, plutonium and HEU wastes. Due to the presence of long-lived fissile nuclides in waste packages, the scenarios consider long timescales of $10^5$ – $10^6$ years.

#### 3.2.1 Reaching a Critical Configuration

Any individual waste package placed in a GDF will be significantly sub-critical, having been designed to meet the criticality safety requirements for both the transport and operational stages of a disposal facility. Hence, for any waste and disposal concept, there will need to be a substantial change from the initial disposal configuration for a critical configuration to be possible. Criticality is influenced by a number of ‘controls’. Given a sub-critical system, one or more of these will have to change for a critical configuration to be possible. Hence an understanding of criticality controls provides a basis for forming ‘what-if’ scenarios for criticality. An overview of the factors influencing criticality, and how these might be important for geological disposal, is given in the following subsections. The criticality controls are often closely linked, so that one cannot change without influencing the others, making it difficult for critical conditions to arise. This is one of the reasons why obtaining a critical configuration is expected to be a low likelihood event for a GDF.

##### 3.2.1.1 Fissile Mass and Concentration

For a critical system to occur fissile materials are required, and need to be present in sufficient concentration and mass. For a system where the geometry is unconstrained (i.e. the size of the system can be adjusted to accommodate a given mass and concentration of fissile material), critical systems typically lie on a handbook curve (or criticality map) which is a curve in concentration and mass parameter space. Such curves will have a minimum critical concentration, below which criticality is impossible, irrespective of the fissile mass (and size) of the system. There will also be a minimum critical mass, below which criticality is impossible, irrespective of the concentration of that mass. For the UCuRC programme a detailed analysis of handbook curves for fissile materials in NRVB (Nirex Reference Vault Backfill) was undertaken and reported in [11].

Under the emplacement conditions for waste within a GDF the fissile mass or concentration would need to change significantly for critical conditions to develop. While packages for SF, plutonium and HEU could contain a fissile mass greater than that required for criticality under optimum
conditions, the packages will have been designed to ensure that the wasteform and emplacement of fissile material within a package have a significant safety margin from optimum conditions.

3.2.1.2 Fissile Decay

Different fissile nuclides may be disposed of in a GDF, with $^{235}$U and $^{239}$Pu being the major contributors to the fissile inventory. Other fissionable nuclides such as $^{238}$U and $^{240}$Pu may also be present, and in general will tend to act as neutron absorbers. These may therefore increase the mass or concentration of fissile materials required for a critical configuration. Given a ‘what-if’ scenario for criticality the mass/concentration required will be different for uranium and plutonium systems, and furthermore the evolution of any transient criticality can be different (see Sections 4.5 and 4.6).

Over long timescales (half-life 24,100 years) $^{239}$Pu will decay to $^{235}$U and hence the timescale of any criticality scenario needs to consider plutonium decay, and how this could affect not only the initial critical configuration, but any transient evolution and consequence analysis. For HLW, SF, plutonium and HEU wastes, where the package integrity is high and package failure could take a very long time (e.g. Section 2.4.1), it is expected that the fraction of remaining $^{239}$Pu will be low on the timescales of any hypothetical criticality event. For example after a period of 100,000 years, only about 6% of any original $^{239}$Pu would remain, the remainder having decayed to $^{235}$U.

3.2.1.3 Moderation

For a system containing fissile materials moderators act to reduce the energy of the neutrons emitted from fission, and can influence the establishment of neutron chain reactions. Water acts as a moderator, for example, as does the presence of organic matter. For an under-moderated system the addition of moderator will act to increase the neutron multiplication factor, $k_{\text{effective}}$, and the addition of sufficient moderator can lead to a critical system if sufficient fissile material is present. For an over-moderated system, where the moderator also acts to absorb neutrons, the removal of moderator will also act to increase $k_{\text{effective}}$.

From the viewpoint of ‘what-if’ scenarios for criticality arising from HLW, SF, plutonium and HEU disposal the considerations of moderation as a criticality control are:

- Waste packages will be emplaced into a GDF while dry, or with low residual water content. Package failure and water ingress into any void space is therefore a means of increasing moderation. On longer timescales any water entering a package could lead to the dissolution and removal of packaging materials, further increasing the moderation of the remaining fissile materials.
- If fissile materials are able to relocate from one or more waste packages then saturation in the buffer or host rock may lead to over-moderation, allowing large (but non-critical) fissile masses to accumulate. Subsequent changes to moderation (due to change in geometry for example) could then expel water, reduce moderation and hence lead to a critical configuration.

3.2.1.4 Reflection

The presence of neutron reflecting material, such as steel and graphite, around a region containing fissile material can affect the possibility of criticality. Reflectors will act to reduce the leakage of neutrons from the system. Hence the replacement of non-reflective material around a fissile region with reflective material could assist in the establishment of a critical system. Since water acts as a reflector then this is potentially significant for wastes in a GDF following saturation. For all wastes placed in a GDF, reflection following backfilling, flooding and re-saturation will have been considered to ensure criticality safety for the operational phase of the GDF. Hence, coupled with the initial sub-critical configuration, it is assumed that changes in reflection alone could not lead directly to a criticality scenario. However, the influence of reflection is part of any analysis of critical configurations arising from changes to other criticality controls.
3.2.1.5 Removal of Neutron Absorbing Materials

Neutron absorbing materials act to absorb the neutrons released from fission, and hence act to lower the neutron multiplication constant, $k_{\text{effective}}$. In most waste types some neutron absorbing materials will be present, but will not be a primary control on the criticality of the system. In such cases the removal of the neutron absorbing materials could not lead to a critical system. Waste packages for plutonium and HEU disposal are different, in that the ceramic wasteform specifically includes the neutron absorbing materials of gadolinium and hafnium. Furthermore, the borosilicate glass surrounding the waste cans within a package includes $^{10}$B, a significant neutron absorber. Removal of one or more of the neutron absorbers could, hypothetically, cause or contribute to, a critical configuration.

In SF, $^{238}$U acts as a significant neutron absorber. For reactor fuels at the end of life the effective fuel enrichment will be of order 1%, so that $^{238}$U (and other absorbing actinides) form the majority of the remaining fuel. However, the removal of these absorbers – for example the separation and removal of $^{238}$U from uranium – leaving behind the fissile isotopes of uranium is not considered credible.

3.2.1.6 Change in Geometry

Geometry is a key criticality control and is often closely related to other criticality controls, since a change in geometry is likely to cause a change in fissile concentration, moderation or leakage.

Changes in geometry leading to the relocation of waste packages is also a potential scenario, although this would require significant seismic or geological activity to occur. Minor seismic activity is quite common in the UK, and the “ten thousand year earthquake” is often used in probabilistic safety analysis for nuclear plant severe accident scenarios, for example. However, for post-closure criticality consequence analysis, the possibility of criticality from the relocation of HLW, SF, plutonium or HEU waste packages is not considered credible, given the separation of several metres between packages in the illustrative disposal concepts.

Further potential scenarios for criticality from geometrical change exist for HLW, SF, plutonium or HEU waste packages, although these could only lead to ‘in-package’ criticality events for those waste packages containing sufficient fissile mass (hence, excluding the possibility for HLW). Scenarios can be hypothesised under both dry conditions (i.e. no water penetration into a waste package) and wet conditions (with water penetration into the package). Dry scenarios could include the gradual degradation/embrittlement and relocation of spent fuel from repeating heating/pressurisation of a package for example. The ‘bowing’ of fuel pins within a fuel element (under compression of a waste package) is a further possibility. Generally, due to the requirement for sufficient moderator to be present for a critical system to be possible, dry in-package scenarios will be much less likely than wet scenarios.

3.2.2 Scenarios for Criticality Consequence Analysis

Having considered the scenarios from the UCuRC programme and Likelihood of Criticality programme [38] in Section 2.5, along with an independent consideration of criticality controls in Section 3.2.1, this section summarises the ‘what-if’ scenarios that have been considered for criticality consequence analysis to extend the understanding from the UCuRC. It is noted that the identification of a ‘what-if’ scenario at this stage does not necessarily imply that transient consequence calculations were required for that scenario. For example, where it can be shown that a scenario cannot lead to a critical configuration, then no transient analysis is required. The selected scenarios illustrate the establishment of critical systems resulting from changes to all of the criticality controls described in Sections 3.2.1.1–3.2.1.6.

3.2.2.1 Out-of-package Accumulation Scenario

In this ‘what-if’ scenario one or more waste packages must be subject to water ingress following package degradation or failure. Through package flow must be established, so that materials are transported from the package(s). The fissile materials could be transported as colloids, particulate, or dissolved in the water. Dependent on the disposal concept, the fissile materials may then be
transported by fluid flow, and could accumulate elsewhere in the GDF or beyond. Potential accumulation sites could include the buffer, system seals, backfilled tunnels/shafts, or the host rock for the GDF, dependent on waste type and disposal concept.

For any accumulation site a balance of fissile mass and concentration, moderation, reflection and geometry will be required for hypothetical critical configurations to be possible.

This scenario is applicable to SF, HLW, plutonium and HEU disposal, in each of the illustrative disposal concepts. For HLW the accumulation from multiple waste packages would be required for a critical configuration to be possible since the fissile content of each package is low (Table 2.2). For SF, plutonium and HEU critical accumulations can be considered for the transport of fissile materials from single or multiple packages. Understanding of this scenario is well developed for criticality consequence analysis, having been the main focus of the UCuRC programme and the development of transient criticality models. A number of criticality consequence calculations for fissile accumulations in NRVB, granite (higher strength rock) and mudstone (lower strength sedimentary rock) have previously been undertaken (e.g. [22,23]).

For calculations of the consequences of criticality from out-of-package accumulation scenarios in a given disposal concept, it is not essential to make a distinction between accumulations from single or multiple waste packages, especially for static criticality calculations. Nor is it essential to make distinctions between the waste types the accumulated material may have come from. Suppose, for example, that a hypothetical critical configuration of $^{235}\text{U}$ is possible in the host rock for a given disposal concept. From a consequences viewpoint, it does not matter whether the $^{235}\text{U}$ originated from HEU, SF, or plutonium having undergone decay, but simply that the configuration is, hypothetically, possible and consequence calculations can be undertaken. Hence, while the likelihood of the accumulation for a given scenario could vary considerably based on the originating wastes [37,38,39], the consequences will not. On this basis, some of the criticality consequence research reported here is equally applicable to the ‘what-if’ accumulation scenario for ILW, LLW and DNLEU disposal [35].

3.2.2.2 In-package Scenarios from Water Ingress

Should a package contain sufficient fissile mass, and the package fail in a saturated disposal facility, then water ingress into the waste package may be sufficient for a critical configuration to develop through increased moderation. Water can have further effects on the package contents, including degradation of the package contents through chemical reactions, or acting as a mechanism to transport structural (or other) materials out of the waste package. This could lead to a variety of ‘what-if’ scenarios for criticality.

Figure 3.1 illustrates possible scenarios for a spent fuel waste package positioned vertically in a GDF. Similar scenarios would apply for a package with a horizontal orientation. In the initial stages it is assumed that water penetrates the package and fills any voidage in the package. Given sufficient void space for the water to occupy, and a suitable combination of fissile mass, concentration and geometry, it is conceivable that a critical configuration could be possible from flooding alone.

If a critical configuration is not possible from the ingress of water alone, then further in-package evolution can be considered, as below:

- Illustration i) in Figure 3.1 shows a scenario where the water ingress degrades the fuel at a greater rate than the structural materials, leading to a ‘slump’ of fissile material to the base of the fuel compartments.
- Illustration ii) shows a scenario where the structural materials degrade at a greater rate and ‘slump’ leaving the fuel in its original location.

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4 For a rapid transient, the source of the fissile material leading to the initial critical configuration will not affect the consequences. For a quasi-steady-state transient the source of the fissile material does not matter for a given arrival rate. If the arrival rate differs dependent on the fissile material source, then the consequences will change.
In Illustration iii) both the fuel and the structural material degrade at similar rates, and ‘slump’ together.

All cases could evolve further (possibly leading to increased concentrations of fissile material, greater neutron moderation or lower neutron absorption) as structural materials are transported out of the package.

Similar scenarios to those in Figure 3.1 can be envisaged for plutonium and HEU packages, although the initial fuel configuration will differ, and the possibility of water ingress is very limited due to low voidage of the waste packages. With a low void space for water, the dissolution of packaging and waste materials (particularly those containing neutron absorbing materials) was deemed an important scenario to consider.

**Figure 3.1: Illustration of different scenarios for in-package evolution of a vertically oriented spent fuel canister following water ingress.** Light orange represents the package canister, light grey the spent fuel, dark grey structural materials, green where water has entered voidage in the structural materials, and blue water. In i) the spent fuel degrades and ‘slumps’ at the greatest rate, in ii) the structural materials slump at the greatest rate, and in iii) all package contents degrade and slump together.

### 3.2.2.3 Summary of ‘What-if’ Scenarios for Criticality Consequence Analysis

The sections above present two general ‘what-if’ scenarios for post-closure criticality from the disposal of HLW, SF, plutonium and HEU wastes within a GDF:

- The **accumulation scenario** which could hypothetically occur in a localised region of a GDF or the surroundings, such as the buffer or host rock.
- The **in-package scenario** for individual waste packages. This assumes that water is able to enter the package. Dependent on the waste type the scenarios taken forward for analysis were different:
- For SF, flooding of the void space in the package.
- For plutonium and HEU, flooding of the void space coupled with the removal of neutron absorbing materials and structural materials.

The study of these scenarios formed the basis of criticality consequence analysis described in this report.

### 3.3 Methodology for Criticality Consequences Analysis

With a defined set of scenarios for post-closure criticality, this section summarises how criticality software is used to understand the consequences of transient criticality events which could, hypothetically, develop from the identified scenarios.

Figure 3.2 summarises the methodology used for understanding the consequences of criticality for the ‘what-if’ scenarios. The method follows a staged approach of firstly selecting a scenario for consideration and then determining whether critical configurations are possible. If critical configurations are possible (and there may be more than one), then the next stage is to determine whether the hypothetical critical configurations would evolve as quasi-steady-state transients or rapid transients, through analysis of the temperature feedback coefficient for reactivity. This determines which of the transient criticality models is appropriate. In either case specific calculations can then be selected and analysed using the transient criticality models. The results of these models return the local consequences. Sections 4 to 6 describe the process in detail. The sections below provide a brief overview of how each stage in Figure 3.2 is approached.

**Figure 3.2: Flow diagram of the methodology for understanding the consequences of criticality for ‘what-if’ criticality scenarios.** For analysis of rapid transient calculations (positive temperature feedback on reactivity) only the Bounding Approach was used for additional calculations during the Modelling of Consequences of Hypothetical Criticality programme. RTM calculations were undertaken during the UCuRC programme\(^5\).

#### 3.3.1 Selection of a Scenario

The scenario of accumulation can apply to any HLW, SF, plutonium or HEU wastes in any illustrative disposal concept\(^6\). To undertake criticality consequence analysis, the first step is to select a specific ‘what-if’ scenario for study. For example, this could be the accumulation of fissile materials in bentonite.

To scope the potential consequences of criticality from accumulation scenarios for all waste types and illustrative disposal concepts it is necessary to consider possible combinations of:

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\(^5\) The Bounding Approach model will, through design, calculate a larger energy release than a comparable calculation using the RTM, and hence larger consequences to the surrounding material. It therefore has built in conservatism. The advantage of using the Bounding Approach model compared with the RTM is that the Bounding Approach model requires fewer complex model inputs to describe the host geology. It is therefore more readily applied for scoping analysis in different geologies.

\(^6\) It is noted, however, that the accumulation from multiple HLW packages would be required for a critical configuration, given the low fissile content of the packages.
• accumulation sites, including buffer, GDF tunnels and host rock; and

• mixes of fissile and fissionable materials, including a $^{235}$U case and a $^{239}$Pu case as a minimum. Selected cases with $^{235}$U and $^{238}$U at different enrichment levels are also of interest. Furthermore different ratios of $^{239}$Pu and $^{235}$U should be considered to recognise the effects of $^{239}$Pu decay to $^{235}$U.

For the in-package scenarios, different variants apply for SF and for plutonium and HEU. To scope the potential consequences of criticality it is important to consider a suitable number of these, such as:

• For SF: different spent fuel types, levels of irradiation, orientation, and levels of flooding and degradation

• For plutonium and HEU: the removal of different neutron absorbing materials and package materials in different combinations following package failure and flooding.

### 3.3.2 Are Critical Configurations Possible?

Given a 'what-if' scenario, static criticality analysis using criticality software such as MONK® or WIMS is required to understand whether critical configurations are possible. Through construction and analysis of a representative model for the ‘what-if’ scenario it is possible to understand the conditions, if any, under which a critical configuration is hypothetically possible.

For the accumulation scenario there will be more than one critical configuration, and often a continuum of varying conditions for criticality [e.g. 11]. This is termed a ‘handbook curve’ or ‘criticality map’. The criticality maps show, for a given fissile composition:

• a minimum critical mass, below which no critical configurations are possible, irrespective of fissile concentration; and

• a minimum concentration, below which no critical configurations are possible, irrespective of how much fissile mass is present.

To derive the criticality maps presented in this report the modelling approach is as follows. For a given host material with porous fraction, $\phi$, the solid host material occupies a fraction $1-\phi$ of any volume. In an inner spherical region of radius, $R$ (metres), it is assumed that fissile material (as oxide) and water occupy all of the pore space. The concentration, $C$ (kg/m$^3$), of fissile oxide is the mass, $M$ (kg), of fissile oxide in the sphere, divided by the volume of the sphere, so that:

$$ M = \frac{4}{3} \pi R^3 C. \quad (3.1) $$

Surrounding the spherical fissile region is the host material in which the pore space is fully saturated with water and any fissile material is neglected. Both regions are modelled as homogeneous, and in criticality calculations the outer region is usually a large sphere to essentially represent an infinite extent for the saturated host rock.

The minimum critical mass can help to inform whether any of the critical configurations could arise from single packages, or otherwise whether multiple packages are required. Further details and examples of criticality maps are given in Section 4.5.

For in-package scenarios it is necessary to model how the package contents could evolve. This will depend on the specific scenario under consideration. Using suitable representations of the geometry and materials for a ‘what-if’ scenario, it is possible to undertake studies of whether the proposed package evolution could lead to critical configurations. For example, with a dry spent fuel

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7 The same general method was used in the UCuRC programme to produce criticality maps. The only difference for some analysis was the use of a cylinder (with height equal to its diameter) in place of a sphere [11]. This means the equation relating mass and concentration requires 2 in place of $4/3$. 

package being sub-critical (as it must be by design), static criticality calculations can be undertaken to model partial and complete flooding of the fuel compartments to establish if criticality is possible and, if so, at what volume of flooding.

3.3.3 How Could a Criticality System Evolve?

If critical configurations are hypothetically possible, then understanding how a critical system could evolve is a natural extension of static criticality analysis. By undertaking further criticality calculations for elevated temperature it is possible to determine whether the system reactivity increases or decreases with temperature. There are two possible contributions to the change in reactivity which are nuclear data\(^8\) effects and changes in the density of water. If the reactivity increases with temperature the system has positive temperature feedback and criticality consequence analysis for rapid transient events is relevant. If the reactivity decreases with temperature the system has negative temperature feedback and criticality consequence analysis for quasi-steady-state transient events is relevant. In this case some mechanism such as continued accumulation or in-package flooding will be required to sustain the quasi-steady-state transient.

For criticality maps there is sometimes a fissile concentration where the system ‘switches’ from positive to negative temperature feedback on reactivity as the concentration increases. Further details on calculating temperature feedback coefficients, with examples, are given in Section 4.6.2

3.3.4 Selection of Calculations

Given an understanding of whether hypothetical criticality events are possible, and how they could evolve, specific criticality consequences calculations can be specified. In doing so, the aim is to undertake a range of calculations. To do so requires that the relevant model inputs are available or can be calculated, and that the models can be applied to the scenario under consideration. Section 5 describes how calculations were specified, model inputs were defined, and model developments were undertaken, to extend the work of the UCoRC programme to include calculations for hypothetical criticality events relevant to the disposal of HLW, SF, plutonium and HEU.

\(^8\) Nuclear data includes information on the behaviour of systems with free neutrons and includes absorption, scattering and fission cross-sections.
4 Static Criticality Analysis

This section provides an overview of why static criticality analysis is required to understand whether ‘what-if’ scenarios could, in theory, lead to a critical system – i.e. the establishment of a sustained neutron chain reaction. The section also provides an overview of software and methods that can be used to determine whether a particular arrangement of materials could be critical. This includes the ICASPA method, which is an automated algorithm for seeking critical configurations of material and the production of criticality maps.

The section also includes a brief overview of static criticality analysis undertaken during the UCuRC programme, and extended analysis relevant to the disposal of HLW, SF, plutonium and HEU. The extended analysis acts both to confirm previous analysis, and to extend it to further host materials (for the accumulation scenario) and a number of different in-package scenarios for SF, plutonium and HEU.

4.1 Determining Whether Scenarios could be Critical

As described in Section 3.2.2 there are two main categories of ‘what-if’ scenarios for criticality from the disposal of HLW, SF, plutonium and HEU, which are:

- Accumulation – fissile materials from one or more packages accumulate in a localised region of the GDF or beyond.
- In-package – following the failure of a package, water is able to enter. Package flooding, or subsequent internal changes to the package contents are ‘what-if’ scenarios for criticality.

The proposition of such scenarios does not mean that criticality events would result from them. Even without considering whether there are physical mechanisms which could lead to the development of the ‘what-if’ scenarios, nor the likelihood that those mechanisms could occur, some of the scenarios might never lead to a critical system. This is because there are a number of factors which control whether a system can be critical. If an appropriate balance of these factors cannot be found, then a critical configuration cannot occur. Furthermore, any emplacement of waste packages into a GDF will be significantly sub-critical, so that substantial changes would have to occur for a critical configuration to be possible.

For static criticality analysis the objective was to undertake a range of criticality calculations for the identified ‘what-if’ scenarios to understand:

1. Whether critical configurations could occur and, if so, under what range of conditions; and
2. How those critical configurations could evolve as transient criticality events – determined by the temperature feedback coefficient for $k_{\text{effective}}$.

4.2 The UCuRC Programme

During the UCuRC programme, accumulation scenarios were considered in detail for a range of materials [11,22,23], so it is already known that critical configurations of material are theoretically possible within a GDF under certain modelling assumptions. While the majority of the analysis was for NRVB, which is a backfill material specific to the disposal of ILW, LLW and DNLEU, some of the materials considered in the UCuRC programme are equally applicable to HLW, SF, plutonium and HEU disposal. In particular the host rocks are common to all wastes for a given illustrative geology. Furthermore, the general trends and observations for NRVB provide valuable background information to how critical configurations could be affected by different conditions, such as the presence of neutron absorbing materials.

For slow accumulation, a number of conclusions were drawn from the UCuRC static analysis [11], including:

- The range of concentrations over which rapid transients are hypothetically possible was established for $^{239}\text{PuO}_2$ in fully saturated NRVB as being between about 8 and 20 kg/m$^3$. 

Much greater masses, although not necessarily concentrations, of fissile material would need to accumulate for a criticality to occur in low porosity rock; however, rapid transient criticality is hypothetically possible if $^{239}$Pu accumulated in the right ranges of mass and concentration.

If neutron absorbers such as iron were to accumulate alongside any fissile materials, then this would not change the qualitative behaviour of critical systems, but could significantly increase the amount of fissile material needed for a criticality to occur.

Low-enriched uranium systems have much larger minimum critical concentrations and masses than other higher enrichment systems.

It is noted that under the Likelihood of Criticality Programme a detailed review of static criticality analysis from former programmes, including material from the UCuRC studies was undertaken [38]. This provides useful information to compare with the additional analysis presented here.

The UCuRC programme included no analysis for in-package scenarios, nor consideration of the specific package designs for HLW, SF, plutonium and HEU. However, this does not mean that no analysis has been undertaken. For the transport and operational stages of a GDF, the criticality safety of all waste packages must be ensured, and hence suitable models of HLW, SF, plutonium and HEU packages have been constructed and analysed for potential criticality risks. An example is [54]. Models from [54] for SF, plutonium and HEU packages are discussed in Section 4.4.2.

4.3 Static Criticality Analysis Methodology

Understanding whether a configuration of fissile and non-fissile materials within a given geometry is critical requires the calculation of $k_{\text{effective}}$, the neutron multiplication constant. The calculation of $k_{\text{effective}}$ for complex geometrical arrangements which can involve many different materials requires criticality software, and various packages are available. During the UCuRC programme several tools were used [25]. For this programme, the software used was MONK, which includes a complex geometry package and looping options suitable for undertaking ranges of scoping calculations. Furthermore, the ICASPA method (see below) has been developed specifically for MONK.

In order to undertake calculations to determine $k_{\text{effective}}$ three key items and sets of information are required:

1. Software to undertake the calculation (i.e. MONK for this study), and a compatible nuclear data library.
2. A geometrical model understood by the software, which needs to adequately represent all key geometrical components. For MONK a three-dimensional model is required.
3. Material specifications for each region of the geometrical model.

For the software, there are clear advantages in using MONK, since ICASPA can be employed for the accumulation scenarios. ICASPA is an algorithm which automates the production of criticality maps for simple geometrical arrangements [55]. It was developed for calculations similar to the accumulation scenario. The ICASPA method has been applied to produce a number of criticality maps, including in the Criticality Assessment for the Low Level Waste Repository (LLWR) Environmental Safety Case [56].

Figure 4.1 provides an overview of how the application of ICASPA and MONK fit into the methodology used to undertake scoping transient criticality analyses and shows that the identification of critical systems is a key step in the process used to scope the consequences of hypothetical criticality.
For nuclear data all of the calculations described in this report use JEF2.2 DICE libraries for MONK\(^9\). The majority of the static criticality calculations are undertaken using the density of water at either room temperature or 40 °C. In either case the closest temperature in the standard DICE library is at room temperature (20 °C) so this value is assumed for the nuclear data. For certain calculations extended versions of the standard library are required which includes additional temperature data for a set of nuclides. This is relevant to the calculations in Section 4.6, and the very high temperature libraries discussed in Section 4.6.3.

**Figure 4.1: Illustration of how static criticality calculations support the programme to scope the consequences of hypothetical criticality.** Unless stated otherwise all of the calculations of \(k_{\text{effective}}\) in this document have been undertaken using MONK 9A. ICASPA can be used for static calculations for the accumulation scenario.

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\(^9\) While there are more recent evaluated nuclear data files available, including JEFF3.1 and ENDF/B-VII, it is not expected that these would have a significant effect on the analysis undertaken. This is supported by analysis in [24] which showed that allowing for uncertainties in cross-section data did not significantly effect the transient criticality calculations undertaken using the QSS model and RTM.
4.4 Information Required for Static Analysis

4.4.1 Accumulation Scenario

For the accumulation scenario the geometrical representation is assumed to be simple – the accumulation of materials in a spherical region. A simple geometry is also required for ICASPA. A sphere is chosen conservatively, since it minimises the amount of fissile material required for a critical configuration at a given fissile concentration.

The remainder of this section (supported by Appendix A, Section A.1) summarises the sources of information that have been used to enable static criticality analysis to be undertaken, including any assumptions that have been made.

For water, at an assumed GDF pressure and temperature of 6.5 MPa and 40 °C (chosen as typical for conditions at a depth of 650 m) the density is 995.01 kg m\(^{-3}\) [57]. For the fissile material, a solid density of 10,000 kg m\(^{-3}\) has been assumed, and that the fissile material (whether uranium or plutonium or a mixture of both) is present as an oxide. While this solid density is slightly lower than the maximum theoretical density quoted in textbooks, it is consistent with the value that was assumed throughout the UCuRC programme [11], and is also considered as ‘typical’ for the fissile materials, which may be present in an oxide form other than the dioxide form used in the calculations. For all static and transient calculations fractions of nuclides in the fissile material have also been taken as atom percentages\(^{10}\).

A material specification (density, porosity and chemical composition) is required for the solid host material (i.e. with any pore space removed). Hence, for example, if a dry material had a density of 2,500 kg m\(^{-3}\) with a porosity of 20%, so the solid fraction is 0.8, then the solid (pore-free) material would have a density of 2,500/0.8 = 3,125 kg m\(^{-3}\). This is significant since when ‘dry’ density is quoted in the literature it is not always clear whether this is with or without the allowance for porosity. In such cases an assumption is required, which has an increasing significance for higher porosity materials. Generally for porosity of less than about 10% the difference is comparable to the uncertainty in density from samples.

There are a number of potential sites at which accumulation of fissile materials could occur within a GDF. The complete list of materials identified was:

- higher strength rock (i.e. granite),
- lower strength sedimentary rock (i.e. clay/mudstone),
- evaporite rock,
- NRVB (Nirex Reference Vault Backfill),
- grout (as a vault backfill),
- grout (as used within ILW packages),
- concrete,
- bentonite,
- compacted bentonite,
- crushed rock (rock type unspecified),
- sand,
- gravel,
- 70% crushed rock (rock type unspecified) and 30% bentonite,
- 70% sand and 30% bentonite,
- crushed rock salt,
- MgO bags,
- clay,
- asphalt.

\(^{10}\)Hence, for example the ratio of 1:3 for \(^{239}\)Pu:\(^{235}\)U would indicate that for every 1 atom of \(^{239}\)Pu there are 3 of \(^{235}\)U. Likewise, 3% enriched uranium would be modelled as 3 \(^{235}\)U atoms to every 97 \(^{238}\)U atoms. This is a subtle difference from ratios by weight percent, but either representation is suitable for the scoping analysis that has been undertaken.
This list includes all materials listed for the three illustrative disposal concepts in any part of the GDF, including both the ILW, LLW and DNLEU region and the HLW, SF, plutonium and HEU region. To reduce this list to a manageable number for both static and transient criticality analyses, a subset of materials was chosen as representative for the illustrative disposal concepts in general:

- higher strength rock (i.e. granite),
- lower strength sedimentary rock (i.e. clay/mudstone),
- evaporite rock,
- NRVB,
- grout (as used within ILW packages),
- bentonite,
- crushed rock (granite) and bentonite in a 70:30 ratio.

This set was chosen on the following basis:

- The three illustrative host rocks are included (higher strength, lower strength sedimentary and evaporite).
- Bentonite is a commonly proposed backfill material surrounding individual waste packages for HLW, SF, plutonium and HEU.
- Crushed rock is potentially present in large volumes within the GDF tunnels, shafts, etc., and may have a larger total porosity than the equivalent solid rock due to porosity (voidage) between the crushed granules.
- NRVB represents a high porosity backfill material, and is a material for which static and transient calculations have previously been undertaken [11,22,23].
- Grout is present in significant quantities for ILW, LLW and DNLEU waste packages. It is therefore a sensible choice for the stack slumping scenario and for the accumulation of fissile materials from one or more waste packages within a single package.

For studies of the HLW, SF, plutonium and HEU region of the GDF this list was reduced further to bentonite, granite, clay, evaporite, and crushed rock which represent the buffer for the wastes, the host rocks and backfill for the tunnels and shafts. While tunnels are present in all regions of the disposal facility, there will be a larger number in the HLW, SF, plutonium and HEU region. Hence the tunnel backfills of crushed rocks have been considered for this report, not [35] for ILW, LLW and DNLEU disposal. Likewise, bentonite could be used in the ILW, LLW and DNLEU region of a GDF and hence be a potential accumulation site. However, since bentonite is used directly as a buffer for waste packages in the HLW, SF, plutonium and HEU region (in contrast to tunnel backfill in the ILW, LLW and DNLU region), it is considered in this report, and not [35].

Appendix A summarises the material specification of the chosen materials for static criticality analysis, where the information has been derived from a number of sources [11,49,58-63]. These are an essential input for static criticality analysis (and also in the production of reactivity functions where required for transient criticality calculations – see Section 5.4.2 and Appendix B).

4.4.2 In-Package Scenarios

To undertake criticality analysis for in-package scenarios, the representation of geometry and materials must be sufficiently detailed to enable a reliable calculation of whether the system could be critical. Spent fuel disposal canisters will contain a number of fuel elements, each of which can have a complex arrangement of fuel rods/pins (see Section 2.1.2 and [54]). For plutonium and HEU, the current illustrative disposal concept is for the fissile material to be stored in ceramic form, with a set of ‘pucks’ in a single can, and a number of cans within the disposal canister (see Section 2.1.3 and [54]). The concept includes neutron absorbing materials in both the ceramic fuel and the surrounding packaging, which are significant for any criticality calculations.

Complex geometries such as ‘pucks within cans within a canister’ and ‘fuel elements within compartments within a canister’ will not be realistically modelled in criticality software packages if the total package contents are ‘smeread’ or averaged into a homogeneous representation over the
package volume. As a minimum the volumes containing fissile material and those not containing fissile material need to be known, along with the correct chemical composition of those regions and their relative locations. To accurately model in-package criticality scenarios for these packages the preferred option was to construct a three-dimensional representation with as much detail for both geometry and material composition as possible.

Because criticality safety is a key component of the safety case for the transport and operational phases of a GDF, the possibility of criticality accidents must be considered from scenarios such as flooding, fire and impact. Inputs for criticality software tools therefore exist for a number of waste packages for the purposes of demonstrating that, even if the packages are subject to an accident during transport or operations, the accident will not develop into a criticality accident (an unplanned neutron chain reaction). International Nuclear Services (INS), with Sellafield Ltd as subcontractors, has undertaken such analysis for PWR fuel packages and plutonium/HEU packages using MONK as the criticality software [54]. Two of the inputs from this work were supplied as follows:

- pwr_inf_package_10128.dat. This includes sets of looping calculations for PWR fuel elements in a disposal canister. The PWR fuel has zero irradiation (i.e. is ‘fresh’ fuel), although different enrichments are included in looping options within the model.

- puck_inf_package_17486.dat. This includes sets of looping calculations for plutonium or highly enriched uranium pucks within cans arranged in a disposal canister. This model is a slice through the canister with the height of one can of pucks, and is modelled with reflective boundary conditions at the top and bottom to represent an infinite array of such cans. This is a simplification of the finite arrangement of cans in a disposal canister, but is a method often used since if an infinite array can be shown to be safe, then a finite subset will be too.

These models include all key volumetric and material components of the waste packages for PWR SF and plutonium/HEU, respectively. While they required some modifications to represent post-closure conditions (mainly the removal of the transportation containers beyond the disposal canister and the addition of a suitable GDF host material such as saturated bentonite), the supply of these models was very helpful in formulating and undertaking a range of calculations for ‘what-if’ post-closure criticality scenarios. The adaption of the models for ‘what-if’ post-closure criticality scenarios, and results from their use with MONK are described in Section 4.5.2

4.5 Static Analysis for the Identified ‘What-if’ Scenarios

This section summarises a number of static criticality calculations, undertaken specifically for the scenarios identified during the programme. The underlying aim was to provide a basis from which to select critical configurations for further analysis using the transient criticality models developed during the UCuRC programme.

For the research programme described in this report the consideration of in-package scenarios was one of the key extensions of transient criticality analysis compared with the UCuRC programme. It was therefore important that sufficient static criticality calculations be undertaken to determine whether critical configurations might, theoretically, be possible for in-package scenarios. To restrict the analysis to a manageable scope, the decision was taken to concentrate on calculations for one type of spent fuel, and the plutonium or highly enriched uranium concept of ceramic pucks. For spent fuel, consideration of PWR fuel was considered sensible. This was on the basis that:

1. Transport safety case calculations have been undertaken previously for PWR fuel [54];
2. The enrichment of uranium in fresh PWR fuel could be relatively high\(^1\) at up to 5%;
3. PWR fuel is designed to be critical in the presence of water (unlike fuel for gas reactors), so that water ingress might lead to the development of critical configurations. The results for

\(^1\) Currently no PWR fuel is enriched above 5%. The calculations for PWR fuel in [54] allow for the possibility of higher enrichment in the future of up to 6%. Calculations in Section 4.5.2.1 have also been undertaken at 6% for a) consistency with [54], and b) to illustrate the trends with fuel enrichment. For any subsequent analysis, a maximum enrichment of 5% was considered.
transport arrangements [54] support this possibility, albeit with different surrounding materials to the waste canister.

4. While the mass of AGR fuel is larger than that for PWR fuel (Table 2.1), the analysis in [54] shows that for fresh AGR fuel at 4% enrichment, the maximum value of $k_{\text{effective}}$ (including three standard deviations) for a fully flooded package, with a water density of 1,000 kg/m$^3$ at atmospheric pressure, is 1.04. This compares to values of 1.19 and 1.25 for $k_{\text{effective}}$ (again including three standard deviations) for 4% and 5% enriched PWR fuel, using the same water density and pressure. The larger values of $k_{\text{effective}}$ for PWR fuel provide the potential for greater reactivity insertion from in-package flooding, and hence larger consequences from a hypothetical criticality event.

4.5.1 The Accumulation Scenario

This section summarises calculations using the ICASPA method with MONK to produce detailed criticality maps for a range of host materials. Due to the savings that ICASPA offers over ‘manually’ searching for critical configurations (as was used in previous studies [11]), it was possible to consider a range of fissile compositions. One of the key factors which determines the nature of a transient criticality is the temperature feedback on the reactivity (see Section 4.6.2). At low fissile oxide concentrations the presence of $^{239}$Pu in the fissile material has a strong influence on this [11]. However, over very long timescales, $^{239}$Pu decays to $^{235}$U (with a half-life of approximately 24,100 years) so that if accumulations take a long time to occur (comparable to the $^{239}$Pu half-life), then a pure $^{239}$Pu system is not possible and a mix of $^{239}$Pu and $^{235}$U would be more representative. To represent this, different ratios of $^{239}$Pu and $^{235}$U were considered in the static analysis, representing one, two and four half-lives for $^{239}$Pu. With calculations for 100% $^{239}$Pu, 100% $^{235}$U, and 3% and 10% enriched uranium this represents a maximum of seven criticality maps for each host material. For each host material the seven criticality maps (or fewer if no critical systems are found for some of the fissile materials) are generally presented in a single figure.

The inputs to MONK, and the use of ICASPA, were independently verified, as was the subsequent analysis of the MONK results to produce the criticality maps (handbook curves) summarised below. It was assumed that all fissile and fissionable materials are in oxide form. Some of the calculations are ‘common’ to those considered for ILW, LLW and DNLEU disposal [35]. This is because accumulation in the host rock is a ‘what-if’ criticality scenario applicable to any fissile waste. In [35] the comparison of results with those from previous studies is also included, giving confidence that the searching algorithm within ICASPA is correctly identifying critical systems.

In the criticality maps below, the critical mass of oxide is plotted on the vertical axis, against the concentration of oxide on the horizontal axis. All curves are derived by searching for concentration and mass combinations that have $k_{\text{effective}}=1$. The figure legends the ratio Pu:U means $^{239}$Pu:$^{235}$U. 100U means 100% $^{235}$U. 100U10 means 100% of 10% enriched uranium, and 100U3 means 100% of 3% enriched uranium. Uranium enrichments and Pu:U ratios are based on the number of atoms. For a given fissile composition the region below the curve is sub-critical. Above the curve is super-critical. On the right hand side (maximum concentration) for any criticality map it is assumed that the fissile oxide completely fills any porosity in the host material. In reality this could not occur due to partial blocking of pore space, for example. However, it represents a convenient upper bound for these theoretical calculations.

4.5.1.1 Bentonite

Figure 4.2 shows a number of criticality maps for bentonite. The minimum critical mass is approximately 1.5 kg (of oxide) for a $^{239}$Pu system, which increases to about 2.5 kg (of oxide) for a $^{235}$U system. There is a clear trend in the curves as $^{239}$Pu decays to $^{235}$U. For the calculations with 10% and 3% enriched uranium, large mass and concentration combinations are required for critical

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12 This means that for the 3% and 10% enriched uranium systems, both the mass and concentration include the totals of $^{235}$UO$_2$ and $^{238}$UO$_2$. The mass and concentration of just $^{235}$UO$_2$, or just $^{238}$U (i.e. the pure metal) can be deduced by scaling the results in the figures.
configurations. Nearly 50 kg (of oxide) is required for a critical configuration with 10% enriched uranium, increasing to about 600 kg for 3% enriched uranium. These correspond to approximately 5 kg and 18 kg of $^{235}\text{UO}_2$ respectively.

Figure 4.2: Criticality maps for bentonite with a porosity of 41%.

![Criticality maps for bentonite](image)

4.5.1.2 Granite

Figure 4.3 shows a number of criticality maps for granite. The minimum critical mass is approximately 44 kg (of oxide) for a $^{239}\text{Pu}$ system, which increases to about 90 kg for a $^{235}\text{U}$ system. There is a clear trend in the curves as $^{239}\text{Pu}$ decays to $^{235}\text{U}$. For the calculations with 10% and 3% enriched uranium no critical configurations were found.

Figure 4.3: Criticality maps for granite with a porosity of 1%.

![Criticality maps for granite](image)
In previous research reporting calculations for accumulation in granite, questions were raised as to whether a homogeneous representation of the accumulation region is reasonable? This is based on the expectation that the porosity in granite will tend to be concentrated in a number of distinct fractures, rather than uniformly distributed.

Formulating a heterogeneous model would be complex if a random fracture network were to be modelled. However, to scope the effects of fractures a simpler representation has been considered through the use of horizontal fractures of given width at the required spacing to simulate 1% porosity. The results show that heterogeneity (of the form assumed) acts to increase the minimum critical mass, and suggests that the use of a homogeneous representation is sensible to bound the minimum critical mass for plutonium systems in granite. Further description of the calculations and results can be found in [35].

4.5.1.3 Clay

Figure 4.4 shows a number of criticality maps for clay (mudstone). The minimum critical mass is approximately 3 kg (of oxide) for a $^{239}$Pu system, which increases to about 5 kg for a $^{235}$U system. There is a clear trend in the curves as $^{239}$Pu decays to $^{235}$U. For the calculations with 10% and 3% enriched uranium, large mass and concentration combinations are required for critical configurations.

Figure 4.4: Criticality maps for clay (mudstone) with a porosity of 12%.

4.5.1.4 Evaporite

Using the evaporite specification in Table A.4 with 1% porosity did not produce any critical systems and so no criticality maps were produced. Analysis of the MONK results show that this is because the chlorine in the evaporite is an effective neutron absorber, and therefore acts to prevent a critical configuration from being possible.

To understand whether a different porosity might, in theory, allow a critical configuration, a further set of calculations were undertaken where the ICASPA method was used repeatedly for a range of porosities from 1.5% to 10%. Figure 4.5 shows that while some critical configurations were found for $^{239}$Pu systems, they were only possible at concentrations of greater than 100 kg m$^{-3}$ of oxide,
and masses greater than 300 kg of oxide. Greater concentrations and masses would be expected to be required for systems with $^{235}$U.

The absence of critical configurations for evaporite with 1% porosity is important in selecting calculations for transient criticality analysis, as is the observation that large masses of fissile material would be required for increased porosity of the evaporite. It is emphasised, however, that should a different evaporite composition be considered (particularly with a reduced level of chlorine) then the same conclusions should not be drawn without undertaking suitable static criticality analysis.

**Figure 4.5: Criticality maps for evaporite with varying porosity from 1.5% to 10%.** These results are for 100% $^{239}$PuO$_2$.

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### 4.5.1.5 Crushed Rock

Detailed information specifying crushed rock is sparse. For detailed calculations it is necessary to understand the porosity (also termed voidage) in the rock particles, what mix of crushed rocks was to be used, the inter-particle voidage (related to the packing fraction of the particles), and a size distribution for the particles. Most references to the use of crushed rock for GDFs come from the SKB literature and suggest a 70:30 ratio of crushed rock and crushed bentonite [64,65], although crushed rock alone is suggested in some cases [66]. In all cases the porosity between the grains of the crushed rock(s) is about 30%.

If detailed information were available on particle size distributions then consideration of a suitable MONK model, including the heterogeneity of crushed rock could be considered. In the absence of such information, and with the expectation that porosity has a significant influence on criticality calculations, simpler homogeneous models were considered instead.

To represent different crushed rocks that could be used in the illustrative disposal concepts two systems were considered:

- The first used the granite composition from Table A.2, but with an increased porosity of 30%. This is representative of crushed host rock – in this instance for the higher strength rock disposal concept.
- The second used a mix of 70% granite and 30% bentonite, with an assumed inter particle voidage of 30% (being representative of the values quoted in [64,65]). Since granite and
bentonite are themselves porous, this gives a total porosity$^{13}$ in the crushed rock of 0.391. The solid composition can be calculated from an appropriate weighting of the compositions in Tables A.1 and A.2.

Figure 4.6 summarises the results from MONK calculations for $^{239}$Pu systems using the two specifications of crushed rock. The results show that for $^{239}$PuO$_2$, the minimum critical mass (of oxide) is just below 3 kg for the porous granite calculations, and just below 2 kg for the granite and bentonite mix. This shows that the granite and bentonite mix, at the assumed porosity, has a very similar behaviour to bentonite alone (where the lowermost curve from Figure 4.2 is for $^{239}$PuO$_2$).

Figure 4.6: Criticality maps for calculations representing crushed rock with $^{239}$PuO$_2$ accumulation.

4.5.1.6 Comparison of Materials

Based on the observation that the criticality maps for $^{239}$PuO$_2$ in bentonite and a granite/bentonite mix are similar, it is interesting to compare the different materials for which criticality maps have been produced.

Figure 4.7 shows this comparison for all of the materials considered above for critical systems with $^{239}$PuO$_2$. From Figure 4.7 it would be possible to deduce that porosity is the dominant control on where critical configurations could hypothetically occur, since there is a clear trend that as the porosity increases the minimum critical mass decreases. However, it is known from the analysis of further materials (specifically grout and NRVB) for the accumulation scenario for ILW, LLW and DNLEU that this deduction is not correct, since the material specification (chemical composition) can also be important [35]. Furthermore, if chemical composition was not important, then there should be a criticality map for accumulation in evaporite matching that of granite since both have 1% porosity. In summary therefore, Figure 4.7 does demonstrate that porosity (and hence the availability of volume for moderator) is a key factor in determining criticality maps, but it must be recognised that the chemical composition of the solid can be important too.

$^{13}$ The total porosity is the sum of three components: 1) the inter-particle voidage 0.3; 2) the small contribution from granite (fraction of rock*fraction of granite*porosity of granite), 0.7*0.7*0.01 = 0.0049; and 3) the contribution from bentonite (fraction of rock*fraction of bentonite*porosity of bentonite), 0.7*0.3*0.41=0.0861.
4.5.2 In-package Scenarios for PWR Fuel

This section describes static criticality calculations for PWR fuel packages, subject to flooding after package failure in a saturated GDF. The approach was initially to modify the MONK input supplied by Sellafield Ltd, through INS (see Section 4.4.2), to represent a package in a GDF instead of a transportation package. This was followed by consideration of how to refine the model to include an irradiated (burned up) fuel specification, since in reality most (or possibly all) fuel that could be disposed of in a GDF would have been subject to some irradiation whilst resident in a nuclear reactor. The effects of irradiated fuel compared with fresh fuel could then be considered.

4.5.2.1 Static Calculations Using Fresh PWR Fuel

The MONK input pwr_inf_package_10128.dat supplied by Sellafield Ltd, through INS, (and reported in [54]) was modified to remove the transport container and add a 50 cm thick water saturated bentonite reflector. Furthermore, the calculations were restricted to 17x17 PWR fuel elements at 1.26 cm pin pitch (the distance between fuel rod centres).

The premise for initial calculations was that the canister is stored vertically and a breach of the copper canister has occurred to allow water ingress, which might then lead to further canister or fuel degradation. Flooding of the canister can affect the value of $k_{\text{effective}}$, since the addition of water can increase neutron moderation and therefore insert reactivity.

Calculations were undertaken for three $^{235}$U enrichments; 6.0, 5.0 and 4.0 wt%. The mass of water present in each fuel element compartment was varied from zero to 30 kg. Figure 4.8 shows a horizontal slice through the disposal canister. The light purple is saturated bentonite, orange is the copper canister, and green the cast iron insert. Figure 4.9 shows a vertical slice through the disposal canister; where in the right diagram the fuel pins in each fuel element compartment have been flooded by 30 kg of water. The figure shows that 30 kg of water floods about 20% of the height of each element, so approximately 150 kg of water per element would be required for complete flooding.

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It was also necessary to change some specific Sellafield Ltd entries in the input file (for their Codemore system) to standard MONK inputs in order to use the supplied input file.
Figure 4.8: Horizontal slice through copper canister, showing portion of saturated bentonite reflector. As in the Sellafield Ltd model [54] the fuel elements are pushed to their closest approach, constrained by the size of the cast iron (green) compartment areas.

Figure 4.9: Vertical slice through copper canister, showing the PWR fuel elements in the canister channels. The left diagram shows the dry arrangement. In the right diagram water (pink) occupies volume around and between the fuel element pins.

| No water in the compartments | 30 kg of water in each compartment |

The results from MONK calculations in a vertical canister following water ingress to each fuel element compartment are summarised in Figure 4.10. For each uranium enrichment considered, the system was critical with between 10 and 15 kg of water present in each compartment\(^{15}\). This

\(^{15}\) Should flooding occur in fewer than four fuel element compartments (vertical configuration) or two fuel element compartments (horizontal configuration – see below), then it is expected that more water would be required per flooded channel for a critical system to arise. Due to the number of combinations that could be considered, and the fact that flooding of four (two) channels represents a worst case for the vertical (horizontal) configuration then calculations with fewer channels have not been considered.
equates to a height up the fuel rods of only 28.75 cm – 43.13 cm (or 6.4% – 9.6% of the total fuel height modelled).

Because flooding of the package alone could lead to a critical configuration, no further degradation studies were conducted. If a system could, hypothetically, become critical from flooding alone, then subsequent degradation leading to criticality is secondary to the initial criticality from flooding. The use of fresh fuel is, however, a ‘worst case’ in terms of the expected insertion of reactivity from flooding. This is demonstrated in Section 4.5.2.3 below.

**Figure 4.10: Results for the addition of water to each fuel element compartment of a spent fuel canister.** The results are for a vertically orientated copper disposal canister, as illustrated in Figures 4.8 and 4.9.

![Graph](image)

It is noted that the observation that critical systems can result from water ingress to the fuel element volumes is consistent with the results of transport safety case calculations in [54]. Furthermore, from a literature review [36] undertaken as part of the research programme to extend the understanding of hypothetical criticality events, it was noted for the Swedish geological disposal programme that recent studies [67] on the long-term safety at the Forsmark disposal site conclude that burn-up credit still has to be assumed to demonstrate criticality safety:

“Credit for burn-up has to be taken to demonstrate that the canister remains subcritical in the repository for all reasonably conceivable scenarios…”.

This is consistent with the results here that with fresh fuel, a scenario with water ingress could lead to a critical configuration.

Consideration was also given to the scenario of water ingress with the canister arranged horizontally. Figure 4.11 illustrates the geometrical arrangement with a vertical slice. The upper two fuel elements are pushed to their closest approach (horizontally) and lowest vertical position, constrained by the size of the cast iron (green) compartment areas. Likewise for the lower two fuel elements. Water (pink) occupies volume around and between the fuel element pins in the lower two elements before rising into the top two elements.
The results from MONK calculations in a horizontal canister following water ingress to the fuel element compartments (the lower two must fill before the upper two) are summarised in Figure 4.12. The water height was varied until a $k_{\text{effective}}$ of $>1$ was achieved.

Figure 4.12 shows that (as for the vertically oriented canister) the system can become critical with just the addition of water for each of the uranium enrichments considered. Since each compartment can contain about 150 kg of water when fully flooded, the results also show that water would only need to flood the lower two fuel elements for a critical configuration to be reached (i.e. the upper two fuel elements would remain dry). However, the mass of water required to achieve criticality is larger than for the vertically oriented canister. For example, with 5% enriched fuel, approximately 100 kg of water per compartment is required (200 kg in total) for a critical configuration in the horizontal canister, compared with about 11 kg per compartment (44 kg in total) in the vertical canister.

**Figure 4.11: Vertical slice through copper canister when placed horizontally.**

**Figure 4.12: Results for the addition of water to the fuel element compartments of a spent fuel canister.** The results are for a horizontally orientated copper disposal canister as illustrated in Figure 4.11. Water would need to flood the lower two elements before reaching the upper two elements.
As for the vertically oriented package, because flooding of the package alone could lead to a critical configuration, no further degradation studies were conducted. If a system could, hypothetically, become critical from flooding alone, then subsequent degradation leading to criticality is secondary to the initial criticality from flooding. However, the use of fresh fuel is, again, a ‘worst case’ in terms of the expected insertion of reactivity from flooding.

Since these static criticality calculations for in-package flooding of PWR fuel show that, on the worst case assumption of fresh fuel, water ingress alone into a fuel canister could be sufficient for a hypothetical critical configuration to occur, this provided potential criticality events for which the transient criticality models could be applied. The analysis also clearly demonstrates that a critical system could be possible with only partial flooding of the waste packages, providing the possibility that further flooding could sustain a transient criticality event.

Before considering the modelling of any transient criticality events, however, it was considered important to undertake further static analysis for PWR fuel, with added realism for the fuel composition. To do so required an understanding of the typical irradiation history of PWR fuel while resident in a reactor, and how this would effect the non-irradiated enriched uranium composition. The modelling of fuel irradiation, together with its use to understand how the results in Figure 4.10 would change if irradiated fuel was modelled, are described in the sections below.

4.5.2.2 Allowance for Irradiation of PWR Fuel

The burn-up of spent fuel could vary widely from zero (for fresh fuel), to high levels where the fuel elements can no longer be arranged to become critical within a reactor configuration. Any level in between could also be present, for example from a failed fuel element removed from a reactor core while at relatively low burn-up. An average burn-up of 55 GWd/Te (Gigawatt days per tonne of uranium) has been used for other studies of spent PWR fuel undertaken by the NDA [68]. Using this value, a WIMS calculation has been undertaken to calculate the typical burned-up composition of a PWR fuel element.

The burn-up of a fuel element will not be constant along its length. The neutron flux profile in a reactor core will tend to be lower at the core edges and fuel element ends. Hence the burn-up of fissile material at the ends of a fuel element will be lower than in the centre. For disposal packages in a GDF the different burn-up at the fuel element ends is significant. In fresh fuel the static criticality calculations in Section 4.5.2.1 showed that a hypothetical critical configuration could arise following water ingress into a disposal canister containing between 4 and 6% enriched uranium fuel\(^{16}\), and to a relatively small fraction of the available height within the fuel element compartments. Hence, if the burned-up fuel still retained an effective enrichment of this level at its ends then a reduced enrichment (higher burn-up) in the mid-regions of the length may not be significant in terms of whether a hypothetical critical configuration can be achieved from flooding of a spent fuel package.

Detailed studies of axial burn-up have previously been undertaken for PWR fuel, a selection of which are summarised in [69,70,71,72]. The aim of the calculation summarised here was to undertake a simple analysis for a PWR fuel element with an average burn-up of 55 GWd/Te to enable the production of a burned-up fuel composition which varies along the length of a fuel element. This composition could then be used to determine whether critical configurations are hypothetically possible for the burned-up specification (Section 4.5.2.3).

A WIMS model was constructed using the assumptions in Table 4.1 in addition to the fuel element specification from [54] and the accompanying MONK input, as used for the fresh fuel static criticality analysis above. The values in Table 4.1 are typical parameters for PWR calculations based on modelling experience. This includes reducing the active length to 3.6 m, compared with the assumed length of 4.5 m assumed in [54] (and for the static criticality calculations above). The fuel was modelled in WIMS as a pincell (a single fuel pin) and was burned up to a total irradiation of 55 GWd/Te. A diffusion theory model was used to generate the axial flux profile. Axially the

\(^{16}\) 6% is greater than the 5% maximum currently used for PWR fuel, and was included to illustrate trends should higher enrichment fuel be considered in the future.
element length was divided into twenty sections of equal length, and a plane of symmetry is assumed between the 10th and 11th sections. This means that the WIMS model only had to calculate the burn-up and fuel compositions for 10 regions.

In addition to a number of burn-up steps to model an average 55 GWd/Te burn-up, some additional cooling stages were added at the end of the calculation to show how the final (55 GWd/Te average burn-up) composition would continue to evolve 10, 100 and 1000 years post-irradiation. Over such timescales many fission products, and some fissile actinides would decay to low levels due to radioactive decay.

A selection of results from the WIMS calculation is presented below. The burned up compositions from each of the twenty axial regions provide an irradiated material specification along the length of a fuel element. This can be used to enable further static criticality analysis to investigate whether the change from fresh fuel to a typically burned up fuel will increase the height of water flooding required for a hypothetical in-package spent fuel criticality, or possibly lead to a system which cannot become critical from package flooding (assuming no degradation of the fuel element structure). These calculations are described in Section 4.5.2.3.

Table 4.1: Parameters for a WIMS burn-up calculation for a PWR fuel element.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel material</td>
<td>UO₂ 5% enriched</td>
<td></td>
</tr>
<tr>
<td>Active length</td>
<td>cm</td>
<td>366.0</td>
</tr>
<tr>
<td>Pin pitch</td>
<td>cm</td>
<td>1.26</td>
</tr>
<tr>
<td>Fuel temperature</td>
<td>K</td>
<td>850</td>
</tr>
<tr>
<td>Clad temperature</td>
<td>K</td>
<td>580</td>
</tr>
<tr>
<td>Water temperature</td>
<td>K</td>
<td>580</td>
</tr>
<tr>
<td>Coolant</td>
<td>Water at 15 MPa pressure</td>
<td></td>
</tr>
<tr>
<td>Water density</td>
<td>g/cm³</td>
<td>0.71</td>
</tr>
<tr>
<td>Dissolved boron</td>
<td>ppm by mass</td>
<td>600.0</td>
</tr>
<tr>
<td>Rating</td>
<td>MW/Te</td>
<td>40.0</td>
</tr>
</tbody>
</table>

In each of the Figures 4.13 to 4.19 results from the WIMS burn-up calculation are presented for the 10 axial divisions of the fuel element, where axial region 1 is nearest the end of an element and axial region 10 near the middle of element. The results in regions 11 to 20 are assumed to be symmetrical to those in 10 to 1. It is noted that the legend orders have been selected to match the order of the curves where possible, so that the legend order is reversed for some figures compared with others. The values in GWd/Te in the legends are the average burn-up for the fuel element. In Figures 4.14 to 4.17 the units on the vertical axis are the number of atoms per 10⁻²⁴ cm³. For ²³⁵U and ²³⁸U the initial (zero burn-up) values are 0.001238 and 0.02321 respectively, so that the total number of uranium atoms per 10⁻²⁴ cm³ is 0.02445. Therefore, the initial fraction of uranium atoms which are fissile is 0.0506, as confirmed in Figure 4.18. This is consistent with the initial 5% uranium enrichment by weight, since ²³⁵U has a slightly smaller atomic mass than ²³⁸U.

Figure 4.13 shows the burn-up for each of the 10 axial distributed regions. For low levels of burn-up (e.g. 5 GWd/Te) the distribution has a cosine-like profile, but as the burn-up increases the profile develops to one which is flat across the mid-element regions (4 to 10) and decreases towards the end(s) of the fuel element. The burn-up in the endmost region is about 70% of that for the majority of the axial regions. The axial burn-up profile is in good agreement with previous studies [72].

Figure 4.14 shows the burn-up of ²³⁵U for the different axial regions. It is clear that while less ²³⁵U is burned up at the end(s) of the fuel element, a significant fraction of the original ²³⁵U has burned up
for a mean overall burn-up of 55 GWd/Te. Figure 4.15 shows similar behaviour for $^{238}$U, with lower, but non-trivial burn-up at the element ends.

Figure 4.16 shows how the burn-up of the original 5% enriched uranium fuel leads to the production of $^{239}$Pu, and how the quantity of $^{239}$Pu increases with burn-up. Apart from at the element end(s) there is little increase in the quantity of $^{239}$Pu beyond an average burn-up of 45 GWd/Te. At the element end(s) a small amount of $^{239}$Pu is gained between the average burn-up of 45 and 55 GWd/Te.

Figure 4.17 shows the variation in the quantity of $^{239}$Pu in the burned up fuel during a cooling period following irradiation. At 10 years the amount of $^{239}$Pu has increased for all axial regions from the decay of higher actinides. By 100 years, the amount of $^{239}$Pu is in decline, and this trend continues at 1,000 years. This is expected since $^{239}$Pu has a half-life of about 24,100 years.

**Figure 4.13: Variation of burn-up along the axis of a PWR fuel element.** The legend shows the mean burn-up for the fuel element.

**Figure 4.14: Variation of $^{235}$U along a fuel element at different levels of average burn-up.**
Figure 4.15: Variation of $^{238}$U along a fuel element at different levels of average burn-up.

![Graph showing variation of $^{238}$U](image)

Figure 4.16: Variation of $^{239}$Pu along a fuel element at different levels of average burn-up.

$^{239}$Pu is an actinide that builds up during the irradiation of the fuel element.

![Graph showing variation of $^{239}$Pu](image)
Figure 4.17: Variation of $^{239}\text{Pu}$ along a fuel element after different levels of post-irradiation cooling. $^{239}\text{Pu}$ has a half-life of about 24,100 years, which is significant over the longest cooling timescale considered.

Figure 4.18 shows the fraction of uranium and plutonium isotopes which are fissile. The figure shows how this ratio reduces from the original 5% (uranium enrichment) value to a value of about 2% along most of the element length and about 2.7% at the element end(s). This shows that there is a significant reduction in the fissile isotope fraction along the whole fuel element length, including at the element end(s), compared with the original 5% enrichment.

Figure 4.19 shows that allowing for some post-irradiation cooling, the fraction of fissile isotopes of uranium and plutonium reduces to just over 2.5% at the element end(s), and about 1.8% for most of the element length. There is little difference between the curves for 100 and 1000 years of cooling. This is because the dominant decay terms are from either one fissile nuclide to another fissile nuclide (e.g. $^{239}\text{Pu}$ to $^{235}\text{U}$) or from a non-fissile nuclide to another non-fissile nuclide (e.g. $^{240}\text{Pu}$ to $^{236}\text{U}$).

The reduced fraction of fissile nuclides along the whole length of the element is an indication that more flooding would be required into a spent fuel disposal canister for a hypothetical criticality to occur. Furthermore, at fissile fractions of about 2% it is possible that no critical configurations could exist following flooding of a spent fuel canister (assuming no degradation to the fuel). Section 4.5.2.3 presents static criticality analysis using the burned up fuel composition in place of fresh fuel.
Figure 4.18: Variation of the fraction of fissile uranium and plutonium nuclides at different levels of average burn-up. The fraction is calculated as the ratio of the number of atoms of fissile U and Pu isotopes to the total number of atoms of all U and Pu isotopes. The initial fraction is slightly larger than 5% since the ratio is atom, not weight, percentage.

Figure 4.19: Variation of the fraction of fissile uranium and plutonium nuclides at different levels of post-irradiation cooling. The fraction is calculated as the ratio of the number of atoms of fissile U and Pu isotopes to the total number of atoms of all U and Pu isotopes. The curves for 100 and 1000 years of cooling are very similar.
4.5.2.3 Static Calculations Using Irradiated PWR Fuel

Having undertaken a WIMS calculation for the burn-up of PWR fuel, including the use of axial subsections to capture the lower burn-up at fuel element ends, it is possible to extract fuel composition data from any burn-up step in the WIMS model. These were set intervals of 5 GWd/Te for the average fuel burn-up.

Taking these data, the burned-up spent fuel compositions have been used in a vertically oriented waste package. At a given level of average burn-up the method used was to extract the full compositional data in each axial element from the WIMS model, and then use this in a modified MONK model where the fuel element length was separated into twenty divisions of equal length. For a conservative approach only the uranium and plutonium isotopes from the WIMS model were used in the MONK specification, thereby neglecting other actinides and fission products that would tend to act as neutron absorbers. Using the WIMS data for composition allowed MONK calculations to be undertaken to show what level of burn-up would be sufficient to prevent the onset of criticality from package flooding (assuming no internal arrangement/degradation of the fuel). The results of MONK calculations using the burned-up fuel compositions are summarised in Figure 4.20. Table 4.2 shows the peak values of $k_{\text{effective}}$ for each curve in Figure 4.20.

Figure 4.20 shows that for fresh fuel (zero burn-up), the system can become critical with water ingress of approximately 11 kg per compartment which is consistent with Figure 4.10. For further water ingress the system becomes super-critical with a peak $k_{\text{effective}}$ of about 1.15.

Figure 4.20 also demonstrates that as the irradiation of the fuel increases, the possibility of a critical configuration reduces. At an average fuel irradiation of 35 GWd/Te (and assuming no subsequent cooling period) it is just about possible to reach a critical configuration, while at higher irradiation levels the system remains sub-critical, even if the compartments containing the fuel elements are completely flooded (just over 150 kg of water per compartment). Furthermore, at an average irradiation of 55 GWd/Te, calculations allowing for different cooling periods (which can be compared with the time from taking the fuel from a reactor to the onset of flooding of a canister in a GDF), it can be seen that cooling acts to make the system more sub-critical. The general conclusions from these calculations are that, for the modelling assumptions employed, the formation of a critical configuration in a PWR spent fuel canister would not be possible, provided the irradiation of all fuel elements in the canister was above 35 GWd/Te. For irradiation levels close to this it may also be possible to demonstrate that the passage of sufficient time post-irradiation would be sufficient for all levels of flooding to be sub-critical.

Table 4.2: Peak $k_{\text{effective}}$ values for each curve in Figure 4.20. The standard deviation on $k_{\text{effective}}$ is 0.001 in each case.

<table>
<thead>
<tr>
<th>Burn-up (GWd/Te)</th>
<th>Cooling Period , $T_i$ (years)</th>
<th>Maximum $k_{\text{effective}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>1.1496</td>
</tr>
<tr>
<td>35</td>
<td>0</td>
<td>0.9988</td>
</tr>
<tr>
<td>40</td>
<td>0</td>
<td>0.9790</td>
</tr>
<tr>
<td>55</td>
<td>0</td>
<td>0.9238</td>
</tr>
<tr>
<td>55</td>
<td>10</td>
<td>0.9055</td>
</tr>
<tr>
<td>55</td>
<td>100</td>
<td>0.8742</td>
</tr>
<tr>
<td>55</td>
<td>1000</td>
<td>0.8783</td>
</tr>
</tbody>
</table>
Figure 4.20: Calculations of $k_{\text{effective}}$ as a function of water ingress into a spent PWR fuel disposal package (copper canister) for 5% enriched uranium dioxide fuel (initially) at different levels of irradiation. The average irradiation is given in GWD/Te, and the cooling period, $T$, is given in years where applicable. 10 kg of water per compartment is equivalent to a flooded height of 0.289 m (156.5 kg = fully flooded).

4.5.3 In-package Scenarios for Plutonium and HEU

The MONK input puck_inf_package_17486.dat supplied by Sellafield Ltd, through INS, (and reported in [54]) was modified to remove the transport container and add a 50 cm thick saturated bentonite reflector\textsuperscript{17}.

The basic arrangement of a radial slice through the model is shown in the left diagram in Figure 4.21. The fissile pucks (red) are within a thin stainless steel can. The cans of pucks are surrounded by borosilicate glass (cream), within a stainless steel canister (light purple). This is placed within a further stainless steel liner (green) to an outer copper canister (orange). The copper canister is surrounded by bentonite (brown-green). There are seven stacks of cans within the inner package, all surrounded by borosilicate glass.

The model supplied by INS (and used for the calculations in [54]) actually models an infinite arrangement by using appropriate reflection data on all sides. A section with the height of one can of pucks is modelled with a hexagonal boundary, which is then reflected to give a model that is effectively axially and radially infinite. For the calculations reported here a finite representation of a single disposal canister was developed.

The description in [54] states that a single package would contain four layers of cans. However, Figure 4 of that report shows five layers, and this is compatible with the package dimensions specified. Hence, as a pessimistic assumption, the single canister model used in the MONK calculations reported below used five layers of cans. Each can contains 20 pucks, each of diameter 6.9 cm and height 2.5 cm. The total can height is 51 cm. The right hand diagram in Figure 4.21 shows this arrangement. The material colours are the same as for the left diagram, but the glass (cream) has been excluded so that the cans can be visualised.

\textsuperscript{17} It was also necessary to change some specific Sellafield Ltd entries in the input file (for their Codemore system) to standard MONK inputs in order to use the supplied input file.
MONK calculations were undertaken for plutonium (90 and 100 wt% $^{239}$Pu, where any remaining plutonium is $^{240}$Pu) and HEU (90 and 100 wt% $^{235}$U, where any remaining uranium is $^{238}$U) with the 0.01 cm gap between the pucks within the cans filled with void or water. The water cases would only be possible if some mechanism had formed for water to penetrate the multiple barriers in the canister/cans arrangement. In all calculations the value of $k_{\text{effective}}$ was <0.2, representing a significantly sub-critical arrangement. The largest value calculated was for 100 wt% $^{239}$Pu with flooding, which gave a value of 0.1554 for $k_{\text{effective}}$. For 90 wt% $^{235}$Pu the result was 0.1479, and for 100 wt% $^{235}$U and 90 wt% $^{235}$U the results were 0.1360 and 0.1271, respectively.

This observation that $k_{\text{effective}}$ is significantly sub-critical for all arrangements considered is consistent with the analysis in [54].

Further calculations were then undertaken to see whether changes to the material compositions could increase $k_{\text{effective}}$ to a critical value. An identified ‘what-if’ scenario for this is the removal of neutron absorbing materials (Section 3.2.2.3), which are included in both the ceramic pucks and the glass in the disposal package concept.

The first step was to degrade the boron in the borosilicate glass – achieved by changing the natural boron input in the material specification to $^{11}$B (the largely non-absorbing component of natural boron). With this modification the MONK calculations were again undertaken for plutonium (90 and 100 wt% $^{239}$Pu) and highly enriched uranium (90 and 100 wt% $^{235}$U) and for the 0.01 cm gap between the pucks within the cans filled with void or water. The largest value of $k_{\text{effective}}$ calculated was for 100 wt% $^{239}$Pu with flooding in the cans, with $k_{\text{effective}} = 0.3232$. This is still significantly sub-critical.

The next step was to degrade the gadolinium in the ceramic puck material – achieved by changing the natural gadolinium input in the material specification to $^{162}$Gd (the component of natural gadolinium with the smallest absorption cross-section). This degradation was coupled with that from the previous boron degradation case, i.e. no $^{10}$B in the borosilicate glass and only $^{162}$Gd in the ceramic pucks. With this modification the MONK calculations were repeated, and the most critical system was again for 100 wt% $^{239}$Pu with flooding in the cans, with $k_{\text{effective}} = 0.3710$.

The next step was to degrade the hafnium in the ceramic puck material – achieved by changing the natural hafnium input in the material specification to $^{176}$Hf (the component of natural hafnium with the smallest absorption cross-section). This degradation was coupled with that from the previous boron and gadolinium degradation cases, i.e. no $^{10}$B in the borosilicate glass, only $^{162}$Gd in the ceramic gadolinium and only $^{176}$Hf in the ceramic hafnium. With this modification the MONK calculations were repeated, and the most critical system was again for 100 wt% $^{239}$Pu with flooding in the cans, with $k_{\text{effective}} = 0.4187$.

These differences indicate how the results for $k_{\text{effective}}$ change between Pu and HEU systems, and on whether the enrichment is 100% or lower.
These results suggest that even the removal of all the dominant neutron absorbing materials from the ceramic fuel form and the glass infill around the cans could not lead to the development of a critical configuration within a package. Hence, further degradation of the package contents was considered.

The next set of calculations took the last modification from above (i.e. no $^{10}$B in the borosilicate glass, only $^{162}$Gd in the ceramic gadolinium and only $^{176}$Hf in the ceramic hafnium) as a starting point and introduced the concept of cracked, dissolving, and flooded glass. Here a porosity variable was introduced for the glass region, and water was added to the volume created by this parameter. This was achieved using the HOM Hole in MONK and example visualisations are shown in Figure 4.22.

Using this modification to the model the maximum $k_{\text{effective}}$ was achieved when all of the glass had been replaced by water. Even in this case the maximum value of $k_{\text{effective}}$ was only 0.5227, again for 100wt% $^{239}$Pu.

The analysis undertaken strongly suggests that the ceramic form for the plutonium and HEU, and its packaging according to the proposed geometrical arrangement of pucks in cans in canisters, is inherently safe with regards to the possibility of criticality. Even with the removal of several criticality controls such as neutron absorbing materials and the ingress of water into cracked glass, the calculations do not indicate that a critical configuration is possible.

Figure 4.22: Arrangement for Pu or HEU ceramic pucks in a disposal canister where the glass is modelled as cracked, with the pore space filled with water.

In order to assess whether it could, in fact, be possible to obtain a criticality with this ceramic material a number of further MONK calculations were carried out\(^{19}\). A calculation containing an infinite array of pucks (100% $^{239}$Pu in the plutonium component) was performed using the simple puck model in Figure 4.23. Initially the puck compositions were as specified in [54]. Undertaking calculations for systems of infinite extent (achieved via the use of reflecting boundary conditions) enables the calculation of $K_{\text{efffinity}}$, the ratio of the number of neutrons resulting from fission in one generation to the number absorbed in the preceding generation.

\(^{19}\) The aim was to consider highly idealised arrangements of the ceramic pucks to establish 1) whether an infinite arrangement could be critical (if not, then a finite critical system is very unlikely), and 2) whether, under idealised configurations, there is sufficient fissile mass (in the ceramic form) within a disposal canister for a critical system.
Figure 4.23: A single 6.9 cm diameter puck surrounded by a hexagon to allow an infinite array with variable spacing. In calculations the space was either filled with void or water and gap size was varied between 0 and 1 cm.

The single 6.9 cm diameter puck was surrounded by a hexagon to allow an infinite array with variable spacing. The space was either filled with void or water and gap size was varied between zero and 1 cm. The void cases all yielded a $k_{\infty}$ of ~1.125. The lack of leakage in the model means that a neutron leaving one ceramic puck enters another with no change in energy or direction so it is effectively as if the gap is not there. Interestingly, the maximum $k_{\infty}$ observed for the flooded cases was 0.9109, when the pucks were touching. This reduction is due to the moderation causing the thermal absorbing Gd and Hf elements in the ceramic to become more effective.

Further calculations were performed for this same model but with the gadolinium and hafnium elements changed as above to simulate the removal of neutron absorbing materials (only $^{162}$Gd in the ceramic gadolinium and only $^{176}$Hf in the ceramic hafnium). The maximum value of $k_{\infty}$ from this set of calculations was 1.5189 where a 0.7 cm gap (both radially and axially) between pucks was adopted. This suggests that under an optimum configuration of pucks with the neutron absorbing materials removed, a finite critical configuration would be possible.

A calculation was then undertaken where the infinite geometry was constrained to a sphere of variable radius, surrounded by a 50 cm thick saturated bentonite reflector. The puck specification included the changes to simulate the removal of neutron absorbing materials as above. The value of $k_{\text{effective}}$ became greater than unity at a radius just in excess of 23 cm. This equates to 321 pucks or the total quantity of 16 cans of pucks. While this is less than the total number of pucks in a single disposal canister and illustrates that following significant geometric re-arrangement of the pucks and poison removal, a critical system might be possible, the above is not considered to be a credible scenario for in-package evolution. It should, therefore, not be viewed as a suggestion that such an arrangement is possible. Instead it supports the conclusion that, for the range of in-package MONK calculations considered for plutonium and HEU canisters, it has not been possible to find a credible critical configuration.

Further possible analysis could be undertaken to consider the cracking of (and water ingress into) the ceramic pucks for example. However, given the large number of criticality controls that would have to have been removed or changed, the credibility of such analysis needs to be considered. Furthermore, if the package degradation was so extensive that all of the materials were cracked/broken, then it could be argued that the transport of fissile material with groundwater would be occurring. In this case the accumulation scenario could apply instead.
4.6 Nuclear Data and Temperature Feedback Coefficients

Once a neutron chain reaction starts (i.e. the system is critical) heat is released and so, locally, the temperature will increase. The effect of a change in temperature on the value of $k_{\text{effective}}$ determines the nature of the criticality transient. There are two ways in which a criticality event may evolve. One type of transient criticality is a potentially long-lived, but low-power transient, sustained by competing processes maintaining a just-critical system. Such a transient can result from a critical system with negative temperature feedback, where the temperature increase acts to reduce $k_{\text{effective}}$, but another process, such as the continued arrival of more fissile material can act to increase $k_{\text{effective}}$. The other type is a short-lived, but energetic transient, and can result from a critical system with positive temperature feedback, where $k_{\text{effective}}$ increases with a rise in temperature. These are termed Quasi-Steady State (QSS) and Rapid Transient (RT) criticality, respectively, and could potentially have very different impacts on a GDF. A significant part of the UCuRC programme was spent developing software models to understand the evolution of QSS and RT criticality events.

The calculation of temperature feedback requires the calculation of how much $k_{\text{effective}}$ changes with temperature. The temperature feedback coefficient is the rate of change of $k_{\text{effective}}$ with temperature. There are two influences of temperature that are relevant. Firstly the density of water in the pores will change with temperature, and secondly nuclear data can vary with temperature. Nuclear data includes cross-section data for neutron absorption, neutron scattering, and the fission of fissile nuclides, for example. Density is simple to change through the MONK material specifications, but changes in nuclear data can require the addition of information to the nuclear data libraries used by MONK (or other criticality software). The remainder of this section considers the need for such data, and its application in calculating temperature feedback coefficients.

4.6.1 Higher Temperature Nuclear Data

The standard JEF2.2 DICE library used for MONK has data for a large number of nuclides (and natural elements) at room temperature (20 °C). For a smaller set of nuclides/elements including hydrogen, oxygen, and some uranium isotopes, the standard library also includes additional nuclear data at 100, 200, 300, 400, 500 and 600 °C.

Higher temperature nuclear data libraries were used extensively in a study into the sensitivity of the QSS and RT models to nuclear data [24] during the UCuRC programme. During that study the standard MONK JEF2.2 DICE library was extended to include additional higher temperature data for nuclides/elements considered significant for GDF transient criticality applications. Furthermore, for the application of RTM (or indeed the Bounding Approach [27]), much higher temperature nuclear data libraries are required, including temperatures up to the order of 100,000 °C. A specific high temperature DICE library was therefore created for the nuclear data sensitivity study including a range of nuclides/elements relevant to the application of the transient models for rapid transient events [24].

For this study, the initial requirements for higher temperature data are in the standard MONK DICE library. By undertaking calculations at the elevated temperature of 100 °C, the results can be compared with those at a lower temperature to establish if the temperature feedback is positive or negative.

Based on the material specifications of the different solids for the accumulation scenarios (Appendix A) any nuclide/element with a weight fraction greater than 0.01 in any material was considered as a requirement for higher temperature nuclear data. Together with a consideration of what other nuclides/elements may be important for ‘what-if’ criticality scenarios for ILW, LLW and DNLEU [35], it was concluded that the addition of higher temperature data (i.e. at 100, 200, 300, 400, 500 and 600 °C) to the standard DICE library would be beneficial for $^{27}$Al, Ca, $^{23}$Na, Si, Cu, $^{10}$B, $^{11}$B, $^{238}$Pu, $^{239}$Pu and $^{241}$Pu. This is in addition to nuclides/elements with elevated temperatures in the standard library and those added for the nuclear data sensitivity study [24].
An extended library to include all of these nuclides/elements was created, using the same methodology as for the studies in [24]. Its application to calculate temperature feedback coefficients is described below.

4.6.2 Calculation of Temperature Feedback Coefficients

All of the static analysis that has been undertaken uses a temperature of 40 °C for calculating the water density (995 kg/m$^3$ at an assumed pressure of 6.5 MPa). In all cases the nuclear data used by MONK will be that at 20 °C. Using the extended DICE nuclear data library, temperature feedback coefficients can be calculated by repeating each calculation at 100 °C. The change in $k_{\text{effective}}$, divided by the temperature change gives the temperature feedback coefficient. The value is expressed in pcm per °C which is calculated from converting $k_{\text{effective}}$ to reactivity in pcm (reactivity = $10^5$($k_{\text{effective}}$ - 1)/$k_{\text{effective}}$), then calculating $20$(change in reactivity)/(change in temperature). Note that pcm is sometimes referred to as mNiles for the units of reactivity.

4.6.2.1 Accumulation Scenario

For the accumulation scenario, feedback coefficients were calculated for an increase in temperature of the fissile material region. For a given criticality map the method used was as follows:

- For each concentration and mass pair from the criticality maps (from which the radius can be calculated) four further MONK calculations are undertaken:
  - A re-run of the MONK calculation to confirm that the value of $k_{\text{effective}}$ is close to unity, and to obtain a reference calculation with a small standard deviation (0.0005).
  - A re-run of the MONK calculation with the density of water in the inner fissile region reduced to 961.33 kg m$^3$, which is the value at 100 °C and 6.5 MPa used in [24], but using the standard DICE library with nuclear data at 20 °C.
  - A re-run of the MONK calculation using the 100 °C nuclear data from the extended DICE library, but the water density at the value for 40 °C and 6.5 MPa (995.01 kg m$^3$).
  - A re-run of the MONK calculation using both the 100 °C density for water, and the 100 °C nuclear data from the extended DICE library.

The second and third steps are included since, to within statistical variations, the change in $k_{\text{effective}}$ for the final variation (both density and nuclear data changes), as compared with the reference calculation, should be approximately equal to the sum of the differences of each of the other two calculations relative to the reference calculation. The method for calculating the feedback coefficients is in good agreement with previous analysis [11].

Table 4.3 shows results for temperature feedback coefficients in bentonite for the accumulation of critical configurations for $^{239}$PuO$_2$. The second column confirms that the ‘base’ values of $k_{\text{effective}}$ are close to unity for all of the concentrations considered. The third column shows that the feedback coefficient for the change in water density is positive for low oxide concentrations, and negative from above about 8.5 kg m$^3$, although at very high concentrations the feedback coefficient is only marginally negative. The fourth column shows that the nuclear data feedback coefficient is also positive at lower oxide concentrations, and remains positive to above 10.5 kg m$^3$. Generally, since the MONK calculations were undertaken to a standard deviation of 0.0005 for $k_{\text{effective}}$, the standard deviation in any reactivity value is about 50 pcm, and hence in the difference of two reactivity values, the standard deviation is about 70 pcm. This means that the standard deviation in any temperature feedback coefficient is approximately 1.2 pcm per degree Celsius. Bearing this in mind, the final column of Table 4.3 shows that the total feedback coefficient is in agreement with

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$^{23}$ For calculations where the base temperature is 40°C for water density, but 20°C is used for nuclear data, the temperature change at 100°C is either 60°C for density or 80°C for nuclear data. For the calculation of the feedback coefficient, where it is the sign (positive or negative) which is of greatest interest, the temperature change is assumed to be 60°C.
the sum of the feedback coefficients for density and nuclear data effects to within three standard deviations\(^\text{21}\).

All of the general trends are in good agreement with similar analysis undertaken during the UCuRC programme [11].

Table 4.3: Temperature feedback coefficients for \(^{239}\text{PuO}_2\) critical systems in bentonite.

<table>
<thead>
<tr>
<th>Oxide Concentration (kg m(^{-3}))</th>
<th>(k_{\text{effective}}) (base case)</th>
<th>Temperature feedback coefficient (density only) pcm/(°)C</th>
<th>Temperature feedback coefficient (nuclear data only) pcm/(°)C</th>
<th>Temperature feedback coefficient (density and nuclear data) pcm/(°)C</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.3</td>
<td>1.0012</td>
<td>13.9</td>
<td>25.5</td>
<td>43.6</td>
</tr>
<tr>
<td>6.2</td>
<td>0.9990</td>
<td>12.4</td>
<td>19.5</td>
<td>30.5</td>
</tr>
<tr>
<td>6.6</td>
<td>1.0020</td>
<td>10.7</td>
<td>17.6</td>
<td>26.8</td>
</tr>
<tr>
<td>7.4</td>
<td>1.0000</td>
<td>8.0</td>
<td>16.3</td>
<td>19.6</td>
</tr>
<tr>
<td>8.2</td>
<td>0.9987</td>
<td>0.3</td>
<td>10.1</td>
<td>9.5</td>
</tr>
<tr>
<td>10.3</td>
<td>1.0007</td>
<td>-5.5</td>
<td>0.5</td>
<td>-2.3</td>
</tr>
<tr>
<td>14.4</td>
<td>0.9985</td>
<td>-10.8</td>
<td>-3.5</td>
<td>-15.5</td>
</tr>
<tr>
<td>20.5</td>
<td>1.0003</td>
<td>-17.5</td>
<td>-9.0</td>
<td>-26.1</td>
</tr>
<tr>
<td>28.7</td>
<td>0.9993</td>
<td>-19.4</td>
<td>-11.9</td>
<td>-32.0</td>
</tr>
<tr>
<td>41.0</td>
<td>1.0005</td>
<td>-22.4</td>
<td>-9.9</td>
<td>-34.2</td>
</tr>
<tr>
<td>71.8</td>
<td>1.0002</td>
<td>-24.2</td>
<td>-8.4</td>
<td>-35.2</td>
</tr>
<tr>
<td>123</td>
<td>1.0000</td>
<td>-22.3</td>
<td>-6.5</td>
<td>-31.6</td>
</tr>
<tr>
<td>369</td>
<td>0.9997</td>
<td>-22.0</td>
<td>-2.7</td>
<td>-25.9</td>
</tr>
<tr>
<td>615</td>
<td>0.9983</td>
<td>-17.4</td>
<td>-2.5</td>
<td>-22.4</td>
</tr>
<tr>
<td>820</td>
<td>0.9974</td>
<td>-18.3</td>
<td>-3.0</td>
<td>-21.7</td>
</tr>
<tr>
<td>1025</td>
<td>0.9992</td>
<td>-18.1</td>
<td>-2.8</td>
<td>-20.4</td>
</tr>
<tr>
<td>1640</td>
<td>0.9990</td>
<td>-15.2</td>
<td>-3.8</td>
<td>-15.5</td>
</tr>
<tr>
<td>2460</td>
<td>0.9973</td>
<td>-7.1</td>
<td>-0.2</td>
<td>-9.6</td>
</tr>
<tr>
<td>4100</td>
<td>0.9998</td>
<td>-1.0</td>
<td>-0.7</td>
<td>-0.8</td>
</tr>
</tbody>
</table>

Figures 4.24 to 4.26 summarise the total temperature coefficients for bentonite, clay and granite. Calculations in evaporite were not considered since no critical configurations were found at the specified porosity for the evaporite composition considered. Calculations in crushed rock were not undertaken, although would be expected to show similar trends. In each of Figures 4.24 to 4.26 the temperature feedback coefficients have been calculated over the range of oxide concentrations where critical configurations were found in the static (criticality map) analysis.

All of the results show similar trends, which are in agreement with previous analysis [11]. The main observations are:

\(^{21}\) The standard deviation on the sum of the feedback coefficients for density and nuclear data is approximately 1.7 pcm per degree Celsius.
1. Positive temperature feedback is only possible for low concentration systems (typically less than about 10 kg m\(^{-3}\) of oxide in the fissile region), and only if \(^{239}\)Pu is present in sufficient quantity. This means that rapid transient criticality events could only develop if sufficient \(^{239}\)Pu is present. Furthermore, the passage of time, and associated decay of \(^{239}\)Pu to \(^{235}\)U, narrows the range of fissile concentrations from which a hypothetical rapid transient criticality could result.

2. Most configurations have negative temperature feedback so could only result in a QSS transient criticality, given a suitable sustaining mechanism such as continued accumulation.

3. As the concentration increases, the feedback coefficient tends to reduce to a minimum (or largest negative value), before increasing (but remaining negative) as the oxide concentration is increased further.

4. Total temperature feedback coefficients are generally constrained to between plus or minus order tens of pcm per degree Celsius.

5. Where the temperature feedback coefficients are smaller (i.e. Figure 4.26 for granite) the disadvantage of using a Monte Carlo method such as MONK can be seen, since if variations are small the statistical uncertainty in the calculations can be significant. This appears as ‘oscillations’ in the feedback coefficient summaries. For most of the materials these statistical uncertainties are not significant.

Together with the criticality maps these summaries of temperature feedback coefficients enabled scoping calculations to be selected for a wider range of host materials than considered in the UCuRC programme for the ‘what-if’ accumulation scenario.

**Figure 4.24: Total temperature feedback coefficients for critical systems in bentonite with 41% porosity.**
Figure 4.25: Total temperature feedback coefficients for critical systems in clay.

Figure 4.26: Total temperature feedback coefficients for critical systems in granite.
Using the temperature feedback information together with criticality maps provides an enhancement to the maps, whereby the ‘switch’ from positive temperature feedback coefficient at lower concentrations to negative temperature feedback at larger concentrations can be seen. This is shown for bentonite in Figure 4.27.

Figure 4.27: Criticality maps for bentonite, with estimates (black diamonds) of where the switch from positive to negative temperature feedback occurs as the fissile concentration increases. No marker means that positive feedback is not possible for the given fissile composition.

Figure 4.27 is produced as follows. From Figure 4.24 it is possible to estimate where the feedback coefficient is zero for each of the different fissile compositions considered. Superimposing this information on the criticality maps (from Figure 4.2), as shown with the black diamonds in Figure 4.27, shows the approximate location of the switch in sign for the temperature feedback coefficient. For curves with no black diamond the temperature feedback is always negative, which is the case for the all of the curves where the fissile material does not contain plutonium.

Figure 4.27 clearly shows that, for fissile accumulations in bentonite, positive temperature feedback coefficients are only possible in a narrow range of fissile concentrations from just over 5 kg/m$^3$ to just over 10 kg/m$^3$, with the range decreasing as $^{239}$PuO$_2$ decays to $^{235}$UO$_2$. Furthermore, as the fraction of $^{235}$UO$_2$ increases in the mixed $^{239}$PuO$_2$: $^{235}$UO$_2$ systems, the total fissile mass (of both $^{239}$UO$_2$ and $^{235}$PuO$_2$) required for a positive feedback system increases significantly. These trends suggest that the longer it takes for a hypothetical critical accumulation of fissile material to occur, the less credible a rapid transient criticality becomes, with the expectation that rapid transient

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22 It is noted that since the calculation of temperature feedback coefficients is subject to a standard deviation of approximately 1.2 pcm per degree Celsius, the concentration value at which the temperature feedback coefficient is zero is an estimated value. Where the temperature feedback coefficients are small (i.e. a few pcm per degree Celsius) this means there is uncertainty about the sign of the feedback coefficient (positive or negative). Hence, for example, the statement that hypothetical positive feedback systems can be found for plutonium systems after four half-lives of decay (albeit within a very narrow range of fissile concentrations) could be due to uncertainty alone. This is because in such instances the positive feedback coefficients are usually no more than a few pcm, but the uncertainty (one standard deviation) is ±1.2 pcm. Therefore, at less than two standard deviations of uncertainty the feedback coefficient could be negative.
criticality is not credible for hypothetical critical accumulations that require post-closure timescales of at least 100,000 years to develop.

4.6.2.2 In-Package Scenario for PWR Fuel

Similar analysis to that for the accumulation scenario was undertaken for the in-package scenario of flooding within a PWR fuel package. For those systems where critical configurations were found (i.e. sufficient flooding and lower levels of irradiation) the expectation was that all temperature feedback coefficients would be negative. This is on the basis of both the high fraction of $^{238}$U in any PWR fuel (even when irradiated), and that, by design for use in water reactors, the temperature feedback coefficient should always be negative. Formal calculations of temperature feedback confirmed the values to be negative.

4.6.3 Requirements for Very High Temperature Nuclear Data

For critical systems with a positive temperature feedback coefficient, transient criticality analysis for either the RTM or Bounding Approach transient criticality models requires the input of system reactivity as a function of temperature (and radius for RTM). These reactivity calculations may require nuclear data libraries with data for temperatures up to the order of 100,000 °C. A specific high temperature DICE library was created for a nuclear data sensitivity study [24] to include the species H (free gas), C, O, Na, Mg, Al, Si, S, K, Ca, Ti, Fe, $^{235}$U, $^{236}$U, $^{238}$U and $^{239}$Pu. H in H$_2$O is also included at room temperature and 300 °C (the highest temperature available) in the library. All data are from JEF2.2 evaluated data files.

This nuclear data library was already suitable for most of the material specifications in the accumulation scenario, as required for the material compositions in Appendix A (and also for material specifications for host materials in [35]). However, bentonite includes the element P (phosphorus) which was not in the nuclear data library used in [24], so the library was modified to include this. It was not considered necessary to add Cl for evaporite, as no critical configurations were identified for that model. For the in-package scenario for PWR fuel no further nuclides/elements were required. The requirement for additional nuclides/elements for in-package scenarios for plutonium and HEU was not considered, since no critical systems were found for in-package plutonium or HEU scenarios (see Section 4.5.3).

4.7 Summary

Sections 4.1 to 4.6 above summarise how static criticality analysis was undertaken to establish whether critical configurations are possible for the ‘what-if’ criticality scenarios, and how critical systems could develop as either QSS or rapid transients.

Using suitable compositional data for the required materials, and the ICASPA tool, criticality maps have been produced for a range of host materials for the accumulation scenario, including materials that could be present in the three illustrative disposal concepts for a GDF. A key observation is that for the evaporite material specification considered no critical configurations are possible.

Static criticality analysis for in-package scenarios provide valuable insight into whether the ‘what-if’ scenarios could, hypothetically, result in critical configurations. For a study of a PWR fuel package the static analysis shows that critical configurations could result from package flooding alone, but only if the fuel is fresh or has an average irradiation of less than 35 GWd/Te. Because critical systems can be found from package flooding these provide a basis for undertaking criticality consequence analysis using the transient criticality models. Scenarios of further package degradation following flooding have not been undertaken, but could be considered as extensions to the analysis. For the in-package flooding scenario only QSS calculations are required since no systems with positive temperature feedback were found.

The requirements to extend nuclear data libraries to include higher temperature data have also been considered, and the libraries amended as required. From an extension of the JEF 2.2 DICE data library for MONK, temperature feedback coefficients have been calculated for a range of
materials for the accumulation scenario criticality maps, and for critical systems from the in-package scenario for PWR fuel. The accumulation scenario results are in good agreement with previous analysis, with the key observation that positive temperature feedback is only possible for low concentration systems (typically less than about 10 kg m\(^{-3}\) of oxide in the fissile region), and only if \(^{239}\)Pu is present in sufficient quantity. Due to the decay of \(^{239}\)Pu to \(^{235}\)U, sufficient quantity can be related to the elapsed time post-closure. The static criticality analyses show that, for the different host materials considered for the accumulation scenario, positive temperature coefficients (and hence rapid transient criticality events) are not considered credible after about four half-lives of \(^{239}\)Pu decay, or 100,000 years, given an initial composition of \(^{239}\)PuO\(_2\) for the fissile material.

The analysis undertaken provided sufficiently detailed results to guide the selection of initial conditions (i.e. systems which are critical or super-critical) for a range of transient calculations using the QSS and Bounding Approach models. This is discussed further in Section 5.
5 Modelling Transient Criticality

Section 1.3.2 briefly described how different models were developed during the UCuRC programme to understand the potential consequences of hypothetical post-closure criticality events in a GDF. This section provides some more details on the models including the key assumptions. The requirements to extend the QSS model to enable scoping calculations for an in-package scenario for spent fuel are also described. A specification for transient criticality calculations is then given, followed by a description of the preparations required in order to undertake those calculations.

5.1 The UCuRC Programme

As described in Section 1.3.2, different transient criticality models were developed during the UCuRC programme, and their development and application for a range of hypothetical criticality events is well documented. In follow on work user guides for the models have been produced [28,29]. The model development during the UCuRC programme predominantly focused on the slow accumulation scenario. A schematic diagram of the scenario, which motivated the model development, is shown in Figure 5.1.

**Figure 5.1: Schematic diagram of the slow accumulation scenario.** In the inset schematic graph, $t$ and $T$ are time and temperature, respectively.

In Figure 5.1 the assumption was that fissile materials would be deposited in a spherical region until such time as it became critical. Dependent on the temperature feedback coefficient, the criticality would either

1. end (if negative temperature feedback and there is no means to sustain the criticality);
2. continue as a quasi-steady-state under continued accumulation of fissile material, whereby the mass and concentration within the spherical fissile region increase as further fissile material accumulates; or
3. evolve as a short-lived rapid transient criticality event.

The QSS model was developed for option 2, and the RTM and Bounding Approach models for option 3. Sections 5.1.1 to 5.1.3 provide an overview of the models, as developed for the UCuRC programme, including the assumptions made when using the models.

5.1.1 The QSS Model

The QSS model was developed to gain an understanding of hypothetical QSS criticality events for the scenario of slow accumulation of fissile material. The model is idealised, in that it aims to
capture the essential physics of a QSS criticality under slow accumulation. Simplifying assumptions are made about the geometry, composition and heterogeneity that would be present should a QSS criticality occur in a GDF, post-closure. The aim of developing the QSS model was to implement a relatively simple model that would run quickly enough to undertake a range of calculations across the space of critical systems and to understand trends in the magnitudes of effects and to test sensitivities [15].

The QSS model is a development of a quasi-steady-state, analytic model of GDF criticality developed in the US by Morris [73]. The conceptual model consists of a sphere of host material and water in which fissile and other isotopes of plutonium and uranium can accumulate, together with their fission and decay products. The volume of the sphere remains constant and it is surrounded by an infinite expanse of a host material and water, which contains no uranium or plutonium. It is assumed that the sphere and the surrounding backfill are wholly saturated with water, that the fissile material in the sphere is homogeneous and that the plutonium and/or uranium are deposited uniformly in the sphere. During the criticality, it is assumed that conditions remain constant apart from the rise in temperature (due to the power generated by fissions), and the composition of the plutonium, uranium and (fission, decay and capture) products derived from them.

The transient is assumed to progress at a sufficiently slow rate to allow the negative temperature feedback to maintain the reactivity close to zero. As further fissile material accumulates, the positive insertion of reactivity from the accumulating fissile material (at a specified arrival rate) is balanced by the negative temperature feedback and burn-up or decay of the fissile materials. Hence, the approximation is made that the reactivity is zero throughout the transient. This balance of competing contributions to reactivity is physically explained as follows. Because the system is just-critical some energy will be released from fission. The heat produced increases the temperature, and hence acts to decrease the reactivity, thereby compensating for the reactivity insertion from the addition of the fissile material. Thus, as more fissile material accumulates, the temperature increases to maintain zero reactivity. This progression continues as long as the mass of fissile material keeps increasing, and as long as the system reactivity increases with added fissile material. In the model, the arrival of plutonium/uranium is usually stopped when the pores in the critical region have become completely filled with fissile material or the arrival rate becomes zero as specified by user input.

The QSS model is implemented in Mathematica [74], and a full mathematical description of the model can be found in [28].

5.1.2 The RT Model (RTM)

The Rapid Transient Model (RTM) was developed to gain an understanding of hypothetical RT criticality events under the scenario of slow accumulation. The model is idealised, in that it aims to capture the essential physics of an RT criticality under slow accumulation. Simplifying assumptions are made about the geometry, composition and heterogeneity that would be present in the unlikely event that an RT criticality occurs in a GDF, post-closure. The aim of developing the RTM was to implement a relatively simple model that would have quick enough run-times to undertake a range of calculations for different critical systems, help to understand trends in the magnitudes of effects, and enable sensitivities to be tested [16]. The RTM was originally designed to undertake a subset of the calculations possible using FETCH, a computer code developed at Imperial College, as part of a ‘twin-track approach’ including comparisons of RTM and FETCH calculations [14,20].

\[\text{For the calculation of reactivity it is also assumed that the fissile region is isothermal.}\]

\[\text{As the concentration of fissile material increases towards fully occupying the pore space of the backfill it is possible that the system becomes under moderated so that the increase in reactivity associated with additional fissile material can be negated by a reduction in reactivity due to the displacement of moderator, and the net insertion of reactivity is negative. In such cases the QSS model will predict unphysical negative temperature rises, and the calculation should be ended by the user when the temperature rise reduces to zero [28].}\]
The RTM models energetic rapid transients using a relatively simple, point kinetics reactor physics formulation coupled to control-volume thermal-hydraulics, a full description of which is given in [29]. The model is based on the known phenomenology of underground explosions resulting from the detonation of chemical high explosives [75] and nuclear devices [76]. Development of the model included the construction of appropriate equations of state (EoS) and structural response models (how the surrounding material responds to a release of energy) for a number of different host materials. The model consists of a sphere containing a mixture of fissile and a host material, referred to as the fissile material region, surrounded by an infinite region of the saturated host material. The fissile material region is assumed isothermal and homogeneous. The power generated by the criticality heats the fissile material region, quickly leading to vaporisation of the materials there. The high pressures generated force an expansion of the fissile material region. Sufficient expansion or sufficient increase in temperature changes the sign of the temperature feedback (from positive to negative) and shuts down the reaction. The fissile material region is assumed to be prompt-critical at the start, i.e. the criticality can be sustained by the ‘prompt’ neutrons directly released by the fissile material alone and does not require the ‘delayed’ neutrons released on a slower timescale by the fission products.

RTM is implemented in Mathematica [74], and a full mathematical description of the model can be found in [29].

5.1.3 The Bounding Approach

The Bounding Approach was developed to gain an understanding of hypothetical RT criticality events under the scenario of slow accumulation. It is a simplified version of RTM. Following a detailed review of the status of the transient criticality models (including verification, validation, benchmarking, uncertainty and sensitivity analysis) [25], it was concluded that the results of RTM calculations were subject to significant uncertainty due to uncertainty in supplying a realistic equation of state and structural response parameters to the model. It also proved time consuming to attempt to define these model inputs for different geologies. A simpler Bounding Approach model was therefore proposed, and the idea was subsequently developed and applied to a range of hypothetical RT criticality events [27]. By constraining the energy release of a rapid transient event to a fissile material region of fixed size, the model provides an upper bound on the energy release. Empirical relationships can then be used to estimate the radius of the cavity that could form, and the extent of cracking to the host material. The Bounding Approach model will, through its governing equations, bound the energy release compared with an RTM calculation for the same hypothetical criticality event. For comparisons undertaken in [27] the energy release was between a factor of two to ten greater for the Bounding Approach model than for the RTM.

While the Bounding Approach requires fewer inputs than RTM, it does require:

- A reactivity function (dependent on temperature only), and also the neutron generation time and delayed neutron fraction [27,29].
- The specific heat capacity of the host material as a function of temperature.
- Empirical relationships, based on underground tests with explosives [75,76].

The Bounding Approach is implemented in Mathematica [74], and a full mathematical description of the model can be found in [29].

5.2 Extending the QSS Model for Changing Geometry

Based on static criticality calculations for the flooding of PWR spent fuel packages, it was shown in Section 4.5.2.3 that, given sufficient flooding within a package where the average irradiation (burn-up) is less than about 35 GWd/Te, hypothetical critical configurations can be found. For PWR fuel with zero or low burn-up these critical configurations require only partial flooding of a waste package, and further flooding could act to insert reactivity and hence maintain a critical system. In Section 4.6.2.2 it was discussed that only negative temperature feedback systems would be possible, should a critical configuration be reached. Hence, only QSS transient criticality events
need to be modelled to understand the possible consequences of hypothetical criticality events for this scenario.

The QSS model, as developed during the UCuRC programme, was not readily applicable to the in-package flooding scenario for hypothetical systems with negative temperature feedback as it applied only to systems of fixed size and geometry. For the flooding scenario, the volume of the region undergoing fissions clearly evolves, as water floods the void space within the fuel compartments. This can be visualised as the increase in the water height in Figure 4.9 (for a vertically oriented canister), for example. Development was therefore undertaken to allow the QSS model to be applied to systems of variable (time dependent) size.

In conjunction with the above scenario requiring modifications to the QSS model, the scenario of ‘stack slumping’ which is relevant to ILW, LLW and DNLEU disposal [35], required similar model developments, but for a fissile region of reducing volume. The QSS model, initially being constructed for a single volume containing fissile material, was more readily adapted to the stack slumping scenario than the in-package scenario which has multiple fuel compartments. Hence, development to the QSS model to enable transient criticality calculations for the stack slumping scenario was implemented initially, while considering whether further developments would be required for the in-package flooding scenario.

In fact, having developed and tested the extended model [35], it was concluded that it could be applied directly to the in-package flooding scenario, provided a number of sensible assumptions were taken, including:

- How to represent the multi-compartment fissile region for the in-package scenario within the QSS model;
- How to use a combination of geometrical change and the arrival rate to correctly simulate the addition of fissile material to the (increasing volume) flooded region;
- How to construct a reactivity function compatible with the above.

Full details of the model developments undertaken to add variable volume functionality to the QSS model, including verification and further testing, are described in Appendix B of [35]. Each of the verification tests, including a previous set from [12] and new tests for the additional functionality, was passed successfully. Examples using the variable volume QSS model were also supplied with the QSS user guide [28].

### 5.3 Specification for Further Transient Calculations

#### 5.3.1 Overview

The static criticality analysis described in Section 4 established what critical configurations are hypothetically possible and how they could evolve.

Based on the results of the static criticality analysis a number of hypothetical criticality events were selected for criticality consequence analysis. These included both QSS and RT criticality events for the accumulation scenario and QSS criticality events for the in-package scenario of flooding within a PWR fuel package.

Extreme transient criticality calculations were kept to a small number by limiting the mass of fissile material in the system at the onset of criticality to the order of 1 to 10 kilograms in most cases. The hypothetical criticality events for criticality consequence analysis were chosen to be compatible with the Likelihood of Criticality programme where possible; for example by including calculations at the minimum critical mass for the accumulation scenario. In the case of the analysis for spent fuel, however, the aim was to consider a ‘worst case’ of flooding within a package containing fresh fuel. This is considered a worst case, since fresh fuel provides the largest possible insertion of reactivity. It is also consistent with modelling studies for the transportation stage of a GDF [54].
Based on the wide range of static calculations that were undertaken which could lead to many transient calculations the aim was to specify a practicable and achievable range of transient calculations to be undertaken using either the QSS model or the Bounding Approach. Although both types of transient criticality can be hypothesised for the accumulation scenario the static criticality analysis shows that rapid transient criticality is only possible if there is a large enough fraction of the isotope $^{239}\text{Pu}$ in the fissile material.

Formal specifications for calculations for the accumulation and in-package flooding scenarios are presented in the following sections, along with the justification for the selections made.

5.3.2 Accumulation Scenario

For this scenario it is hypothesised that some of the fissile mass from one or more waste packages has been transported via groundwater flows (either advective or diffusive), to accumulate in some localised region of the GDF or surrounding rock, and that a critical configuration can arise. For critically consequences analysis it is not important what the ‘source’ of the fissile materials is, only that a critical configuration is, hypothetically, possible. While the source of fissile materials from waste packages is not important, changes resulting from nuclide decay (the most significant being $^{239}\text{Pu}$ decay to $^{235}\text{U}$) can be very important, so calculations considering how the consequences might change due to fissile decay were included in the specification.

For the accumulation scenario the aim was to apply the QSS and Bounding Approach models to scope the consequences of hypothetical criticality events in host materials which have not been considered previously. Based on the results of statics calculations, and previous transient analysis, it was decided that the calculations should focus on accumulations of the fissile isotopes $^{239}\text{Pu}$ and $^{235}\text{U}$. Non-fissile isotopes such as $^{238}\text{U}$ act to increase the mass of fissile/fissionable materials that needs to accumulate for a critical system, and also act to reduce the reactivity of critical systems if more is added.

For QSS calculations it is noted that irrespective of whether the initial configuration is $^{239}\text{PuO}_2$, $^{235}\text{UO}_2$ (or a mixture of the two) then, given sufficient time, the system will tend to evolve as for a $^{235}\text{UO}_2$ system of equivalent initial fissile mass due to radioactive decay. The amount of $^{239}\text{PuO}_2$ that has decayed is determined by the duration of the transient. Lower arrival rates for $^{239}\text{PuO}_2$ would have longer duration (since it takes longer to fill the pores of the host material), and hence will have a closer resemblance to a $^{235}\text{UO}_2$ system.

Taking the materials that have been considered as possible accumulation sites in the static criticality analysis the following specification was agreed:

- Bentonite: This material has not been considered in previous criticality analysis, but could be present in large quantities in a GDF, particularly around waste packages containing HLW, SF, plutonium or HEU. Since bentonite could be present in large quantities in a GDF, and in the vicinity of (potentially) larger masses of fissile material, scoping calculations in bentonite were specified for both RT and QSS systems.

25 The decay of $^{239}\text{Pu}$ can influence calculations in two ways. In any calculation where $^{239}\text{PuO}_2$ is present at the initialisation of a QSS transient, the radioactive decay of $^{239}\text{Pu}$ will be accounted for in both the contents of the fissile region and the arriving fissile material. During the time taken for material to originally accumulate into a hypothetical critical configuration, the decay of $^{239}\text{Pu}$ would also be important, and the greater the time to the establishment of a critical configuration the lower the initial $^{239}\text{Pu}$ content would be. Initial conditions of purely $^{239}\text{Pu}$ are therefore bounding, and not credible unless a critical system could accumulate in a time much less than the half-life of $^{239}\text{Pu}$ (24,100 years).

26 It is noted that the porosity of 41% for bentonite appears quite large, although it comes from a published document. The explanation is believed to be that the value is correct for the total water content of the material, which is important for criticality calculations. However, not all of the ‘porosity’ would necessarily contain water that could be displaced from pore space, since it could be bound to the material. Hence criticality maps at high fissile concentrations might not be achievable in practice, but the calculations of minimum critical mass and minimum critical concentration accurately account for the water content.
Granite: Some RTM calculations and Bounding Approach calculations have been undertaken previously [22,23,27]. A QSS calculation for $^{235}$U was also undertaken [22]. Since the mass of fissile material required for a critical configuration in granite is large compared with that for other materials, and some results are available already, no further scoping calculations were specified for granite systems.

Clay (Mudstone): An RTM calculation for a large mass of 500 kg of $^{239}$PuO$_2$ had limited success due to difficulties in constructing an equation of state [21]. A QSS calculation for $^{235}$U was also undertaken [23]. Since the mass transfer mechanism will be diffusion dominated in clay the credibility of significant accumulations is considered low. On this basis further calculations that required additional model inputs were not undertaken. A Bounding Approach calculation based on the inputs to the RTM calculation considered previously was included in the specification (for comparison with similar fissile masses in NRVB and granite), with the understanding that this was one of only a few extreme, and highly unlikely, rapid transient criticality calculations to be considered.

Evaporite: For the specified material composition and porosity of 1% for a generic evaporite no critical configurations were found. By increasing the porosity it was shown that critical configurations might be possible, but only at large masses (100’s or 1000’s of kg of oxide). Hence no transient calculations were specified for evaporite.

Crushed rock: Static calculations were undertaken for a higher porosity crushed granite (30% porosity), and a bentonite/granite mix (total porosity 39.1%) as described in Section 4.5.1.5. For the second the total porosity assumes a voidage of 30% in the crushed rock/bentonite mix. Intuitively, it might be expected that the crushed bentonite would fill some of the voidage in the crushed rock, which suggests that the 30% voidage is too large. However, no alternative values have been found in the literature. Due to this uncertainty on the porosity, a decision was taken to focus transient criticality calculations on those for bentonite.

In summary, scoping calculations for the accumulation scenario focused on QSS and Bounding Approach calculations in bentonite. Considering the two types of transient criticality in turn, a specification for scoping transient calculations for the accumulation scenario is given in the following two subsections. It is noted that the specification of criticality consequence calculations for only bentonite as the host material for the accumulation scenario does not indicate that different host rocks have been overlooked. In the broader programme including UCURC and additional studies for ILW, LLW and DNLEU disposal, the accumulation scenario has been analysed extensively for NRVB [22,23,27], grout and degraded grout [35], and through additional calculations for granite [23,27] and mudstone [21,23].

5.3.2.1 Rapid Transient Criticality: Bounding Approach Calculations

For the Bounding Approach to be applied a critical configuration is identified and the mass and geometry for the critical region are held constant in order to calculate the system reactivity as a function of temperature. Because the mass and geometry of the critical region do not change, the density of water does not change. Hence, for a rapid transient to be possible, the temperature feedback coefficient must be positive for the effects of temperature on nuclear data alone$^{27}$. In regions of positive total temperature feedback (as shown in Figures 4.24 to 4.26, for example), the nuclear data feedback coefficient is usually smaller than the total temperature feedback coefficient (which includes the water density effect). The requirement to know the feedback coefficient for nuclear data alone causes no difficulty since this contribution to the total temperature feedback coefficient is readily calculated (e.g. Table 4.3 for bentonite).

$^{27}$This also applies to RTM calculations. While in the early stages of a rapid transient the response might be sufficiently ‘slow’ that the density of water in the pore space could change (only possible if mass can be expelled to the surrounding region), for the main part of the energetic event there would not be time for the density of the water to change. Hence, the change in reactivity due to temperature would be dominated by the effects of temperature on nuclear data. For this reason, RTM calculations undertaken previously begin with a system where the temperature feedback from nuclear data is positive. The temperature feedback from the change in water density will usually be positive too, but is not used in the reactivity function for the RTM calculation.
For this programme the specification of calculations to be undertaken was:

- A Bounding Approach calculation for clay (mudstone), based on information for the previous RTM calculation in this material [21]. This calculation was one of a few extreme transient criticality calculations proposed. It was undertaken to allow comparison with similar large mass calculations for NRVB and granite that have been undertaken previously [27].

- To scope the consequences of RT criticality in other materials the focus was on bentonite. For this systems needed to be identified which are prompt critical (the criticality maps produced are for just-critical), and where the nuclear data temperature feedback is positive. These were sought for different ratios of $^{239}$Pu and $^{235}$U. Accumulation masses of 10 kg were specified in most instances, with only one calculation at the more extreme mass of 100 kg for comparison with other large mass Bounding Approach calculations.

Four Bounding Approach calculations were specified for bentonite, as summarised in Table 5.1. Three systems were specified with 10 kg of fissile material. These were restricted to no more than two half-lives of $^{239}$Pu (where the ratio of $^{233}$Pu: $^{235}$U will be 25:75), since at higher fractions of $^{235}$U positive temperature feedback coefficients are unlikely. The calculations covered a range of fissile materials so that trends could be established. The calculation at 100 kg mass was only specified for $^{239}$Pu, to demonstrate how the consequences of extreme transient criticality calculations compare against the more credible (but still unlikely) 10 kg calculations.

<table>
<thead>
<tr>
<th>Material</th>
<th>$^{239}$PuO$_2$</th>
<th>$^{239}$PuO$_2$: $^{235}$UO$_2$ in ratio 50:50</th>
<th>$^{239}$PuO$_2$: $^{235}$UO$_2$ in ratio 25:75</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bentonite</td>
<td>100 kg &amp; 10 kg</td>
<td>10 kg</td>
<td>10 kg</td>
</tr>
</tbody>
</table>

5.3.2.2 QSS Transient Criticality

As in the UCuRC programme, the aim was to apply the QSS model to a range of accumulation scenarios. For each calculation specified a reactivity function must be constructed for supply to the QSS model. This typically requires dependence on temperature and the concentrations of a number of nuclides requiring hundreds or thousands of reactor physics calculations. Hence, the range of calculations to be undertaken needs to be achievable and focused on key areas of interest.

Taking account of calculations that were undertaken for the UCuRC programme [22,23] and for ILW, LLW and DNLEU disposal [35], the focus of QSS calculations for the accumulation scenario was for accumulations in bentonite. A range of initial fissile compositions (mixes of $^{239}$PuO$_2$ and $^{238}$UO$_2$) was specified although it is noted that given sufficient duration, calculations can tend to those for $^{239}$UO$_2$ accumulation due to the radioactive decay of $^{239}$PuO$_2$.

Choosing sets of calculations for comparison within a given material has some challenges. For example, consider the criticality maps for bentonite in Figure 4.2. If calculations were required for 100% $^{239}$PuO$_2$, 50:50 $^{239}$PuO$_2$: $^{238}$UO$_2$ and 100% $^{235}$UO$_2$ for example, and a fixed mass were required, then this could be no lower than the minimum mass for 100% $^{235}$UO$_2$ of about 3 kg (concentration about 40 kg/m$^3$). Taking this mass for 100% $^{239}$PuO$_2$ has a much lower concentration of about 9 kg/m$^3$ (or alternatively a higher concentration of about 90 kg/m$^3$). At the lower concentration the system might lead to a rapid transient (Figure 4.27), so would not be

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28 It was not guaranteed that a Bounding Approach calculation could be undertaken in each case. If the nuclear data temperature feedback coefficient is negative for any of the proposed mass/fissile material/host material combinations then a Bounding Approach calculation cannot be undertaken.

29 The larger mass calculations are undertaken with $^{239}$Pu as the fissile material as extreme accumulations. The use of $^{239}$Pu is considered to be bounding in terms of the expected energy release. In reality, any hypothetical accumulation would take a timescale sufficiently long that a significant fraction of the fissile material $^{239}$Pu would have decayed to $^{235}$U.
suitable for a QSS calculation. Alternatively, if a fixed concentration is specified, then the initial critical masses could be quite different.

The specification, therefore, was to undertake comparison calculations for 100% $^{239}$PuO$_2$, 50:50 $^{239}$PuO$_2$:235UO$_2$ and 100% $^{235}$UO$_2$, starting from an initial condition close to the minimum critical mass for each curve. While the initial masses and concentrations are different, it is something that can be applied consistently. Furthermore, at the minimum critical mass, the size of the system is fairly independent of the Pu:U ratio. This can be seen in Figure 5.2, which presents the criticality map data for radius against mass. It is seen that for all of the Pu:U curves (the five curves grouped closely together), the minimum critical mass occurs at a radius of about 0.25 m.

Figure 5.2: Criticality maps (radius versus mass) for bentonite

During discussions with RMWD it was noted that there is interest in a range of initial masses for QSS calculations. This does not mean that lots of calculations are required, just a suitable range. During further programme alignment discussions with the Likelihood of Criticality programme it was thought that systems where the fissile material(s) fill all of the pore space in a material are not realistic. In reality there would be a point where the pores are effectively blocked for further deposition. It was therefore decided that, for a given material, larger initial masses of about three and ten times the minimum critical mass be considered. It is noted that in this region (e.g. for oxide concentrations of several hundreds of kg/m$^3$ and masses of up to a few tens of kg in Figure 4.2 for bentonite), the criticality maps are fairly independent of the $^{239}$Pu:$^{235}$U ratio. Since it is assumed that all cases decay to $^{235}$UO$_2$ systems given sufficient time, it was not considered necessary to undertake calculations for different ratios of $^{239}$PuO$_2$:235UO$_2$ for the larger mass/concentration initial conditions.

Table 5.2 summarises the specified QSS calculations in bentonite. Five calculations were specified covering different ratios of $^{239}$PuO$_2$ and $^{235}$UO$_2$, and different initial fissile masses. For any QSS calculation a period of time would have to pass before the initial conditions in Table 5.2 could be established. For a given arrival rate of fissile material to the region which eventually becomes critical, it would take longer for the larger mass initial conditions to occur. This initiation time is not considered in the analysis of QSS calculations, but could be significant, and is the reason why initial conditions with a large fraction of $^{239}$PuO$_2$ are unlikely. Once a QSS transient is initiated, the continued arrival of fissile material increases the fissile mass in the fissile material region at a given rate.
Table 5.2: Summary of specified QSS accumulation calculations

<table>
<thead>
<tr>
<th>Material</th>
<th>$^{239}$PuO$_2$</th>
<th>$^{239}$PuO$_2$: $^{235}$UO$_2$ in ratio 50:50</th>
<th>$^{235}$UO$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bentonite</td>
<td>Minimum critical mass</td>
<td>Minimum critical mass</td>
<td>~3 times minimum critical mass</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>~10 times minimum critical mass</td>
</tr>
</tbody>
</table>

5.3.3 In-Package Scenario for PWR fuel

The modelling of ‘what-if’ in-package scenarios was not considered during the UCuRC programme. However, the analysis of ‘what-if’ scenarios for criticality in Sections 4.5.2 and 4.5.3 for different waste packages has shown that the establishment of critical configurations is conceivable for some waste packages, under specific conditions. In particular, the flooding of a waste package containing PWR spent fuel of sufficiently low (or zero) average burn-up, is a scenario where a hypothetical criticality event could develop.

For criticality studies involving spent reactor fuel, it is common to consider ‘fresh’ or non-irradiated fuel as a conservative approach. This approximation is used in transport [54] and dry-storage studies, for example. This is because fresh fuel provides the largest possible insertion of reactivity (increase in $k_{\text{effective}}$) following accidents where the possibility of criticality from flooding must be considered. In reality many, if not all, of the fuel elements would have been subject to some irradiation and burn-up of the original fissile materials during residence of the fuel element within an operating reactor. Hence by demonstrating criticality safety for fresh fuel, it will be ensured for irradiated fuel too.

For the scenario of flooding within a waste package containing PWR fuel, only QSS calculations were required, since the temperature feedback coefficient on reactivity is negative. Based on the static criticality analysis, the use of fresh fuel in other criticality safety analysis, and the requirement for the calculation to be practicable with the available model$^{30}$ QSS calculations were specified as follows:

- Start with a critical configuration for fresh non-irradiated PWR fuel in a vertically oriented waste package, while limiting the uranium enrichment to 5%. The uppermost curve in Figure 4.20 shows that this becomes critical with about 11 kg of water in each of the four fuel compartments. This corresponds to a flooded height of 0.336 m.
  - A decision was taken to consider the vertically oriented canister, since this requires less water ingress for the onset of criticality (compare Figures 4.10 and 4.12) and is also more uniform in flooding since all four fuel elements are assumed to flood simultaneously, whereas in a horizontal configuration the lower compartments would flood first.

- Establish a mechanism to sustain the transient. Figure 4.20 shows that the addition of further water to the compartments acts as a mechanism for increasing reactivity. When fully flooded $k_{\text{effective}}$ reaches a maximum of 1.15 (Figure 4.20 and Table 4.2).

- Construct an appropriate reactivity function to allow for changes in the volume of the flooded region, temperature rise, and changes in nuclide composition of the fuel due to a sustained QSS criticality event.
  - For the reactivity function, assume a cylindrical geometry for the fissile region as an approximation to the four fuel element compartments of square cross-section.

---

$^{30}$ The QSS model [28] requires a simple burn-up model of nuclide decay, fission and capture for key nuclides. The reactivity function also needs to be a function of these nuclides. If the initial specification was for burned up fuel, then it may be necessary to include a greater number of actinides and fission products in the QSS burn-up model and reactivity function. The use of fresh fuel helps to limit the number of nuclides required.
Use the QSS model (as modified to enable variable volume calculations) with increasing volume for the fissile region (the flooded region) to investigate the consequences of this hypothetical criticality event.

- Vary the rate of increase for the flooded volume (the rate of flooding) to determine its effect on the QSS results. Dependent on the mechanism for flooding the rate could be relatively fast (e.g. a sudden weld failure following geological activity) to very slow (e.g. slow water ingress due to pin-hole corrosion through the package canister and insert).
- Consider, where possible, the model approximation of assuming a single fissile region (as required by the QSS model) by varying how this is represented for heat transfer away from the critical region in the QSS model.

To enable the QSS model to be applied for these calculations an assumption is that the pressure is constant. In reality, for a hypothetical flooding package (assumed to be atmospheric pressure when emplaced) surrounded by bentonite (assumed saturated at a pressure of 6.5 MPa corresponding to a GDF depth of 650 m) the pressure in the canister would evolve as a function of time. For the scoping calculations reported in Section 6.3.2 an in-package pressure of 6.5 MPa has been assumed. This is required for the reactivity function for the model (Appendix B), where the density of water varies with temperature at the specified pressure.

5.4 Requirements for Model Inputs

Two major data inputs are required to undertake criticality consequence calculations using the transient criticality models. The first of these is to ensure that suitable values or functions are defined for material properties that are required by the transient criticality models. The second is to ensure that each of the required calculations has a suitably defined, calculated and constructed reactivity function. The reactivity function is usually constructed from the linear interpolation of calculated values of $k_{\text{effective}}$, dependent on one or more variables.

5.4.1 Material Properties

The QSS and Bounding Approach models both require some information on material properties, mainly for the host material(s) in which the hypothetical criticality event is assumed to occur. The inputs required range from scalar quantities to those which are supplied to the models as functions of temperature. The required properties can usually be determined via literature searches, or if that is not possible, through the use of approximation methods. Appendix A (Section A.2) describes the sources of information [57,58,60,66,77,78] and approximations undertaken, in defining material properties for the calculations specified in Section 5.3.

5.4.2 Reactivity Functions

The QSS and Bounding Approach models both require reactivity functions to be supplied. These are specific to each calculation to be undertaken. In the case of a Bounding Approach calculation the reactivity function can be constructed from the interpolation of a set of temperature and reactivity (or $k_{\text{effective}}$) pairs. For a QSS calculation a more complicated function is required, typically requiring the dependence of system reactivity on temperature and the concentration (or mass) of a number of different fissile nuclides, decay, fission and capture products. For the variable volume QSS model, dependence on height and radius are also possible for cylindrical geometry, and dependence on radius for a spherical geometry. For the calculations specified above, only the dependence on height, for an assumed cylindrical geometry was required for the in-package flooding scenario for PWR fuel. All accumulation calculations assume a sphere of constant radius.

Appendix B describes the steps taken to construct reactivity functions in general. For the accumulation scenario, specific examples are included for both the Bounding Approach and QSS models. For the QSS model, the reactivity function must be accompanied by a representative burn-up model, including nuclear data from sources such as [79,80]. The construction of reactivity functions builds on the experience from the UCuRC programme where a number of reactivity...
functions were constructed, including a complex ‘generic’ function for NRVB slow accumulation calculations [12,22].

For the in-package flooding scenario for PWR fuel, the construction of a reactivity function requires a different approach to that for accumulation. The function must ensure that the increase in the fissile volume from flooding, and subsequent burn-up of nuclides, is represented in a physically sensible way. Appendix B (Section B.2.2) provides further details on how the function was constructed.

Other than specific modelling issues for specific scenarios, the only significant difference in the reactivity functions constructed for the calculations specified in Section 5.3, when compared with previous work, is for QSS calculations, where fewer temperature interpolation points have been used. In previous calculations [12,22,23] reactivity functions for the QSS model were constructed for temperatures up to several thousand degrees Celsius, whereas the examples in Appendix B are restricted to a few hundred degrees Celsius.

The reasons for this are twofold. Firstly, with a large number of reactivity functions to generate (including those for criticality consequences calculations for ILW, LLW and DNLEU disposal [35]), the number of MONK calculations had to be kept to a manageable level. Secondly, and with physical justification, the relevance of calculating reactivity functions to higher temperatures can be questioned on the grounds of credibility for accumulation scenarios. At a pressure of 6.5 MPa (corresponding to an assumed depth of 650 m) pore water will boil at 281 °C. If the fissile material region exceeds this temperature then the evaporating water (requiring more volume as steam) will act to displace surrounding water away from the fissile region, and being a long duration event, there will be time for this to occur. In doing so, the capacity for further fissile material to accumulate will be reduced, or even stopped, since the assumption is that there is some underlying flow of groundwater to transport fissile materials.

On this basis the detailed extension of QSS reactivity functions to larger temperatures was considered a lower priority than ensuring that a wide range of calculations for different accumulation materials and fissile masses could be undertaken. For higher temperatures the expectation was that trends established in the UCuRC programme could be assumed. This trend is discussed further in Section 6.2.

5.5 Summary

Sections 5.1 to 5.4 above provide the information required to undertake further consequences of criticality calculations, thereby extending the range of scoping calculations undertaken when compared with those from the UCuRC programme. The extension of the QSS model to enable calculations in systems with variable volume (for the fissile material region) is a significant development, allowing not only for calculations for scenarios such as in-package flooding, but also the ‘what-if’ stack slumping scenario, an understanding of which is considered important for ILW, LLW and DNLEU disposal [35].

Compared with the number of calculations undertaken for ILW, LLW and DNLEU disposal [22,23,27,35], the specification of calculations for HLW, SF, plutonium and HEU disposal is relatively small. This is because:

- For the accumulation scenario, the analysis in [22,23,27,35] covers a wide range of host materials relevant to GDFs, including grout, degraded grout, NRVB, granite and clay. These already cover a representative range of material compositions and porosity values, and the materials include encapsulants, backfill and the surrounding host rocks. Bentonite, as used extensively as a buffer for HLW, SF, plutonium and HEU packages in the illustrative geological concepts, is the only significant omission to other analysis.

- For in-package scenarios, the static analysis in Section 4.5.3 demonstrates that for plutonium and uranium waste packages, criticality within a package is not credible, even if a number of criticality controls are removed from the package. For HLW, the packages do not contain sufficient fissile mass for an in-package criticality to be considered credible. It is only for spent
fuel packages that the analysis in Section 4.5.2 indicates that in-package criticality is conceivable, and only then as quasi-steady-state transients for fuel with sufficiently low burn-up. Criticality consequence analysis can only be undertaken where critical configurations can be demonstrated, and hence the requirements for application of the transient criticality models for in-package scenarios is restricted to analysis of flooding spent fuel packages.
6 Criticality Consequence Analysis

This section summarises the results from a subset of scoping transient criticality calculations undertaken during the UCuRC programme, and all of those specified in Section 5.3. Before summarising the results from the scoping criticality calculations the following section provides an overview of the output from the transient criticality models and how it is used to infer the localised consequences of hypothetical criticality events.

6.1 Model Outputs

The aim of transient criticality calculations is to understand the possible consequences of a range of hypothetical transient criticality events, including the length scales of the effects. Undertaking these calculations in no way indicates that the criticality events would occur. Indeed, supporting analysis of the likelihood of criticality has shown that all of the scenarios are unlikely. Furthermore, some of the calculations are more unlikely on the grounds of either:

- Mass and concentration – intuitively if a large mass is required for a critical configuration, then it is expected that the likelihood will be lower than for a smaller mass. The same is true for concentration. For example, as the concentration required for a critical configuration approaches the maximum theoretical concentration (i.e. displacement of all pore water), then the likelihood will be very low.

- Time – given time, the likelihood of a critical system involving a significant fraction of $^{239}\text{Pu}$ will reduce due to the decay of $^{239}\text{Pu}$. The results in Section 4.6.2.1 also show that a rapid transient criticality event is not expected to be credible after about 100,000 years, due to trends for the fissile concentration range to narrow, and the fissile mass to increase, as $^{239}\text{Pu}$ decays.

Analysis of the Likelihood of Criticality for HLW, SF, plutonium and HEU disposal is reported in [38,39].

For all transient criticality calculations it is emphasised that the models are designed as scoping tools. The models include a number of assumptions and approximations (see Sections 5.1.1 to 5.1.3) so that absolute values of the results produced need to be treated appropriately and with consideration of uncertainty [25]. The models do, however, provide a means to analyse trends in the results such as the ‘rule-of-thumb’ for QSS accumulation scenarios (see Section 6.2), and how energy release depends on the initial fissile mass for Bounding Approach calculations, for example.

6.1.1 Results from Bounding Approach Calculations for Rapid Transient Events

For Bounding Approach calculations the key result of interest is the energy release in joules$^{31}$. From the energy release an estimate of the cavity radius formed by the energy release can be calculated from the empirical relationship [27]:

$$R_c = c \left( \frac{W}{4.18 \times 10^{12}} \right)^{1/3},$$  \hspace{1cm} (6.1)

where $R_c$ is the cavity radius (the radius that vaporises following an energetic release) in metres, $W$ the energy release in joules, and $c$ is an empirical coefficient, dependent on material type and with units m kt$^{-1/3}$. This relationship was established from underground explosive tests [75,76], although it also has a sound physical basis in that taking the cube of the equation, the relationship is essentially that the vaporised volume is proportional to the energy release. The coefficient $c$ is a (dimensional) number of the order of 10 although this varies between different materials, with quoted values of 7.6 m kt$^{-1/3}$ for dense carbonate rocks (e.g. dolomite or limestone) and 11 m kt$^{-1/3}$ for dense silicate rocks (e.g. granite). For NRVB, as a ‘soft’ material it was proposed in [27] that

$^{31}$ The energy in joules can be related to kilotons of TNT equivalent via the conversion 1 kiloton (kt) of TNT = $4.18 \times 10^{12}$ J. This unit of energy is used in the empirical relationships based on underground test data [75,76].
the value could be as large as $20 \text{ m kt}^{-1/3}$ since NRVB is weaker and less dense than carbonate or silicate rocks. With these representative values it is useful to apply each of these factors to the energy release to understand the range of cavity radii that could result from a hypothetical rapid transient event\textsuperscript{32}.

Further length scales of interest are the radii to which cracking may occur. Using data from conventional and nuclear underground tests, the general view \cite{27} is that there is:

- A high permeability (crushed) region of about 2 to 3 $R_c$;
- A fracture / shear failure zone region of about 4 to 5 $R_c$;
- A radial cracking region of about 10 to 14 $R_c$, although it is noted that beyond about 3$R_c$ there is very little change to the permeability of the rocks, even though some fracturing and cracking may have occurred.

The radii of these regions can therefore be estimated from the cavity radius for each calculation.

### 6.1.2 Results from QSS Calculations

The QSS model returns a number of outputs as functions of time, including the average temperature of the fissile material region, the concentrations of each nuclide in the burn-up model (see Appendix B, Figure B.1 and Table B.4 for an example), the power, neutron flux, and radial and tangential strains \cite{28}.

To estimate the length scales of the consequences from QSS calculations an analysis of strains is one option. However, as noted in Appendix A (Section A.2.1.2), the strains are negative for values of Poisson’s ratio less than 3/7 ($\approx 0.43$), and hence for most typical rock/clay like materials.

An alternative way to estimate the length scales of QSS transient criticality events is to use the temperature distribution around the fissile material region. The QSS model calculates the average temperature rise of the fissile material region, $\Delta T$, which is the temperature increase of the fissile material above the ambient temperature $T_\infty$. For a spherical system or constant radius a more realistic temperature distribution can be calculated \cite{12} from:

\[
T = \begin{cases} 
T_\infty + \frac{5\Delta T}{4} \left(1 - \frac{\bar{r}^2}{3}\right) : \bar{r} \leq 1, \\
T_\infty + \frac{5\Delta T}{6\bar{r}} : \bar{r} > 1.
\end{cases}
\]  

(6.2)

where $\bar{r}$ is the non-dimensional radius, $\bar{r} = r/R$, $R$ being the radius of the fissile material region.

This expression, therefore, provides a radial temperature profile for a given average temperature rise for the fissile material region. From this, the distance to which the temperature rise exceeds a given level can be calculated, and hence length scales for the consequences of hypothetical QSS criticality events. This approach has been used for all of the accumulation calculations presented in the sections below, by providing (for the maximum average temperature increase in the fissile material region) the radius within which temperatures would be more than 10 °C above ambient, and another radius within which temperatures would be more than 1 °C above ambient.

\textsuperscript{32} The value of 20 m kt$^{-1/3}$ for the empirical constant can essentially be considered as an additional bound. Not only is the energy bounded (through the model itself), but the cavity radius is then further bounded by the use of an estimated upper bound for the empirical constant. For most rock types it is believed that a value close to 10 m kt$^{-1/3}$ (as opposed to the estimated bound of 20 m kt$^{-1/3}$) is more realistic.
Generally for each QSS calculation and each variation of arrival rate (or rate of flooding for in-package calculations) it is useful to summarise, where possible:

- The time of the maximum temperature rise.
- The maximum temperature rise.
- The maximum power.
- For the maximum temperature rise, the radius within which temperatures would be at least 10 °C above ambient.
- For the maximum temperature rise, the radius within which temperatures would be at least 1 °C above ambient.
- The time at which the arrival of material (or in-package flooding) ends, or the end-time of the calculation if it ends earlier for any reason.
- The temperature rise at the end of the calculation.
- The power at the end of the calculation.

This information is provided for the calculations summarised below, with the exception of the radius of given temperature rises for the in-package QSS calculations. This is because equation 6.2 is derived for heat conduction surrounding a spherical region. While it would be a reasonable approximation for cylindrical geometries where the height and diameter are comparable, this is not the case for the in-package scenarios, where the fully flooded length of the fuel compartments (assumed to be 4.5 m) is much larger than the effective diameter of the fissile region (which will be less than the package diameter of 0.9 m).

### 6.2 The UCuRC Programme

As described in Section 1.3.2, the UCuRC programme extended over nearly ten years, and included model development, verification and confidence building, alongside suites of scoping calculations for hypothetical criticality events. The majority of the calculations were focused on hypothetical events from the disposal of ILW in a higher strength rock. For applications of the QSS, RTM and Bounding Approach models all calculations were for the slow-accumulation criticality scenario. This section provides a brief overview of the calculations undertaken in [22,23,27], with selected results for comparison with the additional calculations reported in Section 6.3.

#### 6.2.1 Results from RTM and Bounding Approach Calculations

In the UCuRC studies of hypothetical rapid transient events, no account was taken of the credibility of the hypothetical criticality events considered. As a result, transient criticality calculations with up to one tonne of $^{239}$Pu were considered for RTM and Bounding Approach calculations. Further discussion of the motivation for this, and why the large mass transient criticality calculations are not considered credible, was given in Section 3.1. While these calculations are not considered credible, they can serve a purpose in demonstrating low consequences for ‘extreme’ hypothetical criticality events.

Tables 6.1 and 6.2 summarise some results from [27], for RTM and Bounding Approach calculations in granite (higher strength rock). Table 6.1, for RTM calculations, shows that even for the extreme assumption of 500 kg of fissile plutonium oxide in granite, the calculated cavity would only be about seven metres. However, RTM results, such as those in Table 6.1, should be

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33 Since Equation 6.2 is for spherical systems, and flooded PWR fuel packages are modelled as cylinders where the height can be several times larger than the diameter, it is not sensible to use Equation 6.2 for the QSS in-package flooding calculations.

34 The selected results from the UCuRC programme only include those relevant to HLW, SF, plutonium and HEU disposal, such as accumulation in granite (higher strength rock) and clay (lower strength sedimentary rock). An overview of results for NRVB, which is relevant to ILW, LLW and DNLEU disposal, is given in [35].
considered as indicative due to uncertainties in how to model the structural response and equation of state, leading to an uncertainty of a factor of two or more in calculated results [25].

Table 6.2 shows the same hypothetical accumulation of fissile material, but modelled with the Bounding Approach model. By comparison with Table 6.1 it is seen that the energy release from the Bounding Approach calculation is a factor of about 2.3 larger than for the RTM calculation. However, through the application of the empirical relationship for the cavity radius (Equation 6.1), it is possible to bound the range of the cavity radius (and hence dependent quantities such as the extent of cracking), without having to consider the effects of model approximations in structural response or equation of state parameters.

An important observation from Table 6.2 is that even for the extreme 500 kg accumulation, which is not considered a credible event (requiring the failure of tens of high-integrity waste packages in close proximity and the accumulation of their fissile contents within a few half-lives of $^{239}\text{Pu}$), the calculated cavity radius is of the order of tens of metres, and damage (cracking/enhanced permeability) would therefore be below 100 m. This demonstrates, that even for an unrealistic and extreme hypothetical criticality event, the damage would be on a scale below that of the GDF depth.

Table 6.1: Energy release and calculated cavity radius for an RTM calculation from accumulation of fissile material in granite. (taken from [27] – RTM version using variable neutron generation time and delayed neutron fraction).

<table>
<thead>
<tr>
<th>Host Material(s)</th>
<th>Fissile Material(s)</th>
<th>Mass of Fissile Oxide (kg)</th>
<th>Radius of Fissile Region (m)</th>
<th>Energy Release (GJ)</th>
<th>RTM Cavity radius (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Granite</td>
<td>$^{239}\text{PuO}_2$</td>
<td>500</td>
<td>3.15</td>
<td>3,550</td>
<td>6.76</td>
</tr>
</tbody>
</table>

Table 6.2: Energy releases and estimates of cavity radius for different empirical constants for a bounding approach calculation from accumulation of fissile material in granite (taken from [27]).

<table>
<thead>
<tr>
<th>Host Material(s)</th>
<th>Fissile Material(s)</th>
<th>Mass of Fissile Oxide (kg)</th>
<th>Radius of Fissile Region (m)</th>
<th>Energy Release (GJ)</th>
<th>Cavity radius (m) assuming empirical factor</th>
<th>Cavity radius (m) assuming empirical factor</th>
<th>Cavity radius (m) assuming empirical factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Granite</td>
<td>$^{239}\text{PuO}_2$</td>
<td>500</td>
<td>3.15</td>
<td>8,190</td>
<td>7.6 m kt$^{-1/3}$</td>
<td>11 m kt$^{-1/3}$</td>
<td>20 m kt$^{-1/3}$</td>
</tr>
</tbody>
</table>

6.2.2 Results from QSS Calculations

During the UCuRC programme the QSS model was applied extensively for the slow accumulation scenario, with a detailed suite of calculations for accumulation in NRVB [22], and further calculations in granite [22] and mudstone [23]. For NRVB the calculations provided wide coverage of different variants for accumulation. This included different rates of arrival (accumulation), different fissile compositions and examples including non-fissile nuclides such as $^{238}\text{U}$ and $^{240}\text{Pu}$, as well as investigations into behaviours at higher temperature including the hypothetical possibility of a temporary switch to a positive temperature feedback system if the pore water boiled. It was from these detailed studies, and supporting analysis of sensitivity that the QSS model was considered fit-for-purpose in [25]. A successful comparison of the QSS model with the Oklo natural reactors [26] has subsequently added further confidence in the modelling methodology.

A key observation of sets of QSS calculations is the ‘rule of thumb’, a phrase first summarised in [22]. The rule was also reported in the CSSR [6]. The rule is summarised below (from [22]):
Consider a system with a given arrival rate, which is not large enough for the system to reach temperatures where the pore water in the NRVB will boil. Suppose that at time \( t \), while material is still arriving, this system has a temperature rise of \( \Delta T \) above the ambient temperature. Also suppose that the power of the system is \( P \), the neutron flux \( \phi \), and the composition of the fissile/fissionable materials is the vector \( C \). Now suppose that in a new calculation, the arrival rate of material is increased by a factor \( X \). Provided \( X \) is not large enough for the system to reach boiling temperatures (or temperatures where positive temperature feedback is reached) the following will be approximately satisfied: at time \( \frac{t}{X} \) the temperature rise will be \( X\Delta T \), the power will be \( XP \), the neutron flux will be \( X\phi \), but the composition will still be \( C \). This general ‘rule of thumb’ can apply over several orders of magnitude for \( X \), and even applies to an extent if the system does undergo pore water boiling. Things are more complicated in this case, however, since there is often a large negative temperature feedback coefficient over the boiling range, requiring a large amount of material to arrive to raise the temperature. It can therefore take a long time for the pore water to boil.

As with any ‘rule’ such as this care should be taken to ensure that it is only applied over the parameter range for which it has been demonstrated, and that any special cases be considered carefully. The transition through pore water boiling is an important example of where caution should be exercised.

Figure 6.1 shows an illustration of the rule of thumb for the temperature rise of the fissile material region for a system where the transient criticality starts from a critical mass of 112 kg of \(^{235}\text{UO}_2\) in granite with a radius of 1.1025 m, and further \(^{235}\text{UO}_2\) accumulates at specific illustrative rates. The figure clearly shows the trend in temperature rise with increasing rates of arrival, including at the largest arrival rate, where the pore water boils. For low arrival rates, which are consistent with the low groundwater flows expected in a GDF, the predicted temperature rise is small. In Figure 6.1 the largest arrival rate, \( 1\times10^{-10} \) kg/m\(^3\)/s, is equivalent to 18 g/year. The smallest, \( 1\times10^{-13} \) kg/m\(^3\)/s, is equivalent to 18 mg/year.

**Figure 6.1: Results of QSS transient calculations for the accumulation of \(^{235}\text{UO}_2\) in granite within a radius of 1.1025 m; the effect of different arrival rates is shown [22].**
Tables 6.3 and 6.4 summarise the results of QSS calculations for the accumulation of $^{235}$UO$_2$ in granite and clay (mudstone), respectively, where additional information has been added (to that in [22,23]) for the peak temperature and radial extent of the temperature rise, as discussed in Section 6.1.2. The arrival rates of fissile material (as oxide) have also been included in units of g/year, calculated by multiplying the rate in kg/m$^3$/s by the volume of the fissile region, then converting from kg/s to g/year. The QSS calculations in both granite and mudstone assumed the same initial radius of 1.1025 m. The initial fissile masses were 112 kg and 63.3 kg of oxide, respectively. By comparison with Figures 4.3 and 4.4, these masses are just above the minimum critical mass of $^{235}$UO$_2$ in granite and about ten times the minimum critical mass of $^{235}$UO$_2$ in clay.

The results for QSS calculations in granite in Table 6.3 agree with the rule of thumb trends, including for large arrival rates where the pore water would boil. If the arrival rate were no larger than about 1 g/year, then the maximum power predicted would be of the order of one kilowatt, and temperature rises above 10 °C would be limited to a radius of a few metres. Larger consequences are only calculated for larger and less likely arrival rates. Table 6.4, for mudstone, is consistent with this, although for the same size (radius) system and same arrival rate, the consequences are slightly larger in mudstone than in granite.

Table 6.3: Summary of QSS outputs for the accumulation of $^{235}$UO$_2$ in granite within a critical radius of 1.1025 m. Results are shown for a range of fissile material arrival rates.

<table>
<thead>
<tr>
<th>Arrival rate (kg/m$^3$/s)</th>
<th>Arrival rate (g/year)</th>
<th>Time of maximum temp. (millennia)</th>
<th>Maximum average temp. rise (°C)</th>
<th>Maximum power (W)</th>
<th>Radius (m) of 10° C temp. rise at maximum average temp.</th>
<th>Radius (m) of 1° C temp. rise at maximum average temp.</th>
<th>End of calculation (millennia)</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 \times 10^{-10}$</td>
<td>18</td>
<td>25</td>
<td>1,067</td>
<td>30,800</td>
<td>98</td>
<td>980</td>
<td>25</td>
<td>1,067</td>
<td>30,800</td>
</tr>
<tr>
<td>$1 \times 10^{-11}$</td>
<td>1.8</td>
<td>154</td>
<td>98</td>
<td>2,830</td>
<td>90</td>
<td>90</td>
<td>254</td>
<td>95</td>
<td>2,730</td>
</tr>
<tr>
<td>$1 \times 10^{-12}$</td>
<td>0.18</td>
<td>1,110</td>
<td>9.7</td>
<td>281</td>
<td>0.80</td>
<td>8.9</td>
<td>2,540</td>
<td>9.4</td>
<td>270</td>
</tr>
<tr>
<td>$1 \times 10^{-13}$</td>
<td>0.018</td>
<td>11,000</td>
<td>0.97</td>
<td>28</td>
<td>-</td>
<td>0.80</td>
<td>15,400</td>
<td>0.9</td>
<td>27</td>
</tr>
</tbody>
</table>

Table 6.4: Summary of QSS outputs for the accumulation of $^{235}$UO$_2$ in clay (mudstone) within a critical radius of 1.1025 m. Results are shown for a range of fissile material arrival rates.

<table>
<thead>
<tr>
<th>Arrival rate (kg/m$^3$/s)</th>
<th>Arrival rate (g/year)</th>
<th>Time of maximum temp. (millennia)</th>
<th>Maximum average temp. rise (°C)</th>
<th>Maximum power (W)</th>
<th>Radius (m) of 10° C temp. rise at maximum average temp.</th>
<th>Radius (m) of 1° C temp. rise at maximum average temp.</th>
<th>End of calculation (millennia)</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 \times 10^{-11}$</td>
<td>1.8</td>
<td>529</td>
<td>145</td>
<td>3,440</td>
<td>13</td>
<td>133</td>
<td>529</td>
<td>145</td>
<td>3,440</td>
</tr>
<tr>
<td>$1 \times 10^{-12}$</td>
<td>0.18</td>
<td>5,290</td>
<td>14.5</td>
<td>344</td>
<td>1.3</td>
<td>13</td>
<td>5,290</td>
<td>14.5</td>
<td>344</td>
</tr>
<tr>
<td>$1 \times 10^{-13}$</td>
<td>0.018</td>
<td>52,900</td>
<td>1.5</td>
<td>34</td>
<td>-</td>
<td>1.38</td>
<td>52,900</td>
<td>1.5</td>
<td>34</td>
</tr>
</tbody>
</table>
6.3 Further Criticality Consequence Analysis

To extend the range of transient criticality calculations undertaken from those of the UCuRC programme, further calculations were specified (Section 5.3). This section summarises the results from the calculations. The calculations use the QSS model and Bounding Approach.

6.3.1 Scoping Calculations for the Accumulation Scenario

6.3.1.1 Bounding Approach Calculations for Rapid Transient Events

This section summarises the results from four Bounding Approach calculations for hypothetical rapid transient events from the accumulation of fissile materials. The calculations cover different masses of fissile material and some different ratios of $^{239}\text{PuO}_2$ to $^{235}\text{UO}_2$ for accumulation in bentonite, and one large mass accumulation in clay (mudstone). The two calculations with accumulated masses of 100 kg or larger are included as extreme transient criticality calculations, where the likelihood is expected to be vanishingly small. The mudstone calculation with 500 kg of fissile material is also included as an extreme calculation for comparison with similar calculations considered previously [27], and in particular the granite calculation in Table 6.2.

A typical result from a Bounding Approach calculation is shown in Figure 6.2. The figure shows that there is an initial ‘delay’ of about 2.45 seconds while the system reactivity increases from its initially just-prompt-critical value, to a highly super-critical value. There is then a period of about ten milliseconds where the power ‘spikes’ and the energy from the rapid transient event is released. The energy release of the rapid transient can be calculated from the integral of the power curve.

Figure 6.2: Power as a function of time for a typical Bounding Approach calculation. This example is for 10 kg of $^{239}\text{PuO}_2$ in bentonite.

A summary of the energy releases from all of the Bounding Approach calculations undertaken for the accumulation scenario are given in Table 6.5, alongside the application of Equation 6.1 with three different values of the coefficient, $c$, used to calculate the cavity radius. For Table 6.5 it is noted that since all of the fissile materials include $^{239}\text{PuO}_2$, then the original ‘source’ of the material must be a waste package (or packages) containing $^{239}\text{PuO}_2$, so that the rapid transient events could not originate from waste packages containing only uranium.

The results in Table 6.5 clearly show that, putting the extreme transient criticality calculations with 100 kg or more of fissile materials to one side, the energy releases are limited to less than 125 GJ, and that even for the largest scaling factor of 20 m kt$^{-1/3}$ for Equation 6.1, the cavity radius is limited to the order of metres, and hence the extent of cracking would be of the order of tens of metres at most.
Another important observation from Table 6.5 is that for equivalent initial fissile masses in the same material, an increasing fraction of $^{235}\text{UO}_2$ reduces the energy release, and generally in ratio to the proportion of $^{239}\text{PuO}_2$ – e.g. in bentonite the energy release for 10 kg of $^{239}\text{PuO}_2$ is 125 GJ. At 50% $^{238}\text{UO}_2$ this is approximately halved.

For the 100 kg fissile mass in bentonite calculation, the energy release is 4.3 TJ, which is consistent with analysis in other materials for similar masses (e.g. in Table 6.2 the larger 500 kg mass in granite has an energy release of about 8 TJ). For the very large mass calculation in clay the energy release is 12.5 TJ. Even for such extreme events the cavity radii would be bounded at tens of metres. Hence, with significant changes in permeability at about three times the cavity radius, this would be less than 100 m. Comparison of the two extreme calculations for 500 kg of fissile oxide shows that the energy release and consequences are larger for the clay calculation. This is a useful observation, since if a given mass were required for a critical configuration, it is expected that the time for that mass to accumulate could be greater in clay than in granite due to diffusive dominated mass transfer in clay compared with advective transfer in granite. Hence, while the presence of a given mass of plutonium oxide might lead to higher consequences in mudstone, the longer time for accumulation could compensate by reducing the fraction of $^{239}\text{Pu}$ (through radioactive decay), and hence the consequences.

Table 6.5 allows an understanding of the effects of the accumulated mass on the consequences of criticality. Calculations with 10 kg and 100 kg of $^{239}\text{PuO}_2$ for bentonite show that the energy release is about 35 times greater for 100 kg of fissile oxide than for 10 kg. This indicates that, for the large mass calculations, the energy release increases at a rate greater than linear. However, this does not mean that the consequences of criticality scale in such a way. Because the radius of the cavity formed (and hence the radius of cracking) scales as the cube root of energy release (Equation 6.1), a factor of 10 increase in mass from 10 kg to 100 kg, scales the length scales of hypothetical criticality events by a factor of about 3 ($\approx 35^{1/3}$).

### Table 6.5: Energy releases and estimates of cavity radius for different empirical constants for Bounding Approach calculations from accumulation.

<table>
<thead>
<tr>
<th>Host Material(s)</th>
<th>Fissile Material(s)</th>
<th>Mass of Fissile Materials (kg)</th>
<th>Radius of Fissile Region (m)</th>
<th>Energy Release (GJ)</th>
<th>Cavity radius (m) assuming empirical factor 7.6 m kt$^{-1/3}$</th>
<th>Cavity radius (m) assuming empirical factor 11 m kt$^{-1/3}$</th>
<th>Cavity radius (m) assuming empirical factor 20 m kt$^{-1/3}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bentonite</td>
<td>$^{239}\text{PuO}_2$</td>
<td>100</td>
<td>1.63</td>
<td>4,300</td>
<td>7.7</td>
<td>11</td>
<td>20</td>
</tr>
<tr>
<td>Bentonite</td>
<td>$^{239}\text{PuO}_2$</td>
<td>10</td>
<td>0.71</td>
<td>125</td>
<td>2.4</td>
<td>3.4</td>
<td>6.2</td>
</tr>
<tr>
<td>Bentonite</td>
<td>$^{239}\text{PuO}_2$, $^{235}\text{UO}_2$ in ratio 50:50</td>
<td>10</td>
<td>0.65</td>
<td>62.7</td>
<td>1.9</td>
<td>2.7</td>
<td>4.9</td>
</tr>
<tr>
<td>Clay (mudstone)</td>
<td>$^{239}\text{PuO}_2$</td>
<td>500</td>
<td>2.79</td>
<td>12,500</td>
<td>11</td>
<td>16</td>
<td>29</td>
</tr>
</tbody>
</table>

### 6.3.1.2 QSS Calculations

A number of QSS calculations were undertaken for the accumulation scenario in bentonite, covering different fissile materials, and different masses. The model was applied to five different accumulation cases (one for each of the initial conditions from Table B.3, and each using a different reactivity function). In most cases at least three different arrival rates were considered.
Example results from QSS calculations are shown in Figures 6.3 to 6.8 for the accumulation of $^{235}\text{UO}_2$ in a sphere of radius 0.244 m in bentonite (initially containing a minimum critical mass of $^{238}\text{UO}_2$). The observations from these figures\(^{35}\) are:

- In each of the figures an increase in arrival rate by a factor of ten reduces the duration of the calculation by a factor of ten.
- Figure 6.3 shows that increasing the arrival rate by a factor of 10 increases the temperature rise by a factor of ten.
- In Figures 6.4 to 6.6 the evolution of the concentrations of $^{235}\text{UO}_2$, $^{236}\text{UO}_2$ and PbO\(_2\) is similar for all arrival rates\(^{36}\), but ‘stretched’ over the duration of the transient.
- Figures 6.5 and 6.6 show that the ratio of $^{236}\text{UO}_2$ to PbO\(_2\) is constant, in agreement with the simple burn-up model used by the QSS model.
- Figure 6.7 shows that increasing the arrival rate by a factor of 10 increases the power by a factor of ten.
- Figure 6.8 shows that increasing the arrival rate by a factor of 10 increases the neutron flux by a factor of ten.

This QSS calculation, and all others for accumulation in bentonite, demonstrate the expected behaviour based on similar calculations in NRVB, granite and clay during the UCuRC programme [22,23]; selected results from which are summarised in Tables 6.3 and 6.4 above. In particular, the rule of thumb (Section 6.2.2) can be seen in the trends above.

**Figure 6.3:** Average temperature increase of the fissile material region as a function of time for the accumulation of $^{235}\text{UO}_2$ in a sphere of radius 0.244 m in bentonite - initially containing a minimum critical mass of $^{238}\text{UO}_2$. The largest arrival rate of $10^{-10}$ kg/m\(^3\)/s is equivalent to 0.19 g/year.

\(^{35}\) In Figures such as 6.3 and 6.7 the apparent ‘oscillations’ are an artefact of the way in which the reactivity functions are constructed using interpolation between calculated values. The linear interpolation can lead to step changes in gradient (reactivity feedback coefficient) at points in the interpolation matrix, which lead to changes in the model results. Previous sensitivity calculations [15] have shown that increasing the refinement of the interpolation grid does not change the nature of the results so that the general trend from the results is accurate.

\(^{36}\) The role of Pb (lead) in the calculations is as a representation of the fission products. This is discussed further in the example burn-up model description in Appendix B (Section B.2.1.2.4). $^{236}\text{UO}_2$ is produced from neutron capture in $^{235}\text{UO}_2$. 
Figure 6.4: concentration of $^{235}$UO$_2$ in the fissile material region as a function of time for the accumulation of $^{235}$UO$_2$ in a sphere of radius 0.244 m in bentonite - initially containing a minimum critical mass of $^{235}$UO$_2$. The largest arrival rate of $10^{-10}$ kg/m$^3$/s is equivalent to 0.19 g/year.

Figure 6.5: Concentration of $^{236}$UO$_2$ in the fissile material region as a function of time for the accumulation of $^{235}$UO$_2$ in a sphere of radius 0.244 m in bentonite - initially containing a minimum critical mass of $^{235}$UO$_2$. The largest arrival rate of $10^{-10}$ kg/m$^3$/s is equivalent to 0.19 g/year.
Figure 6.6: Concentration of PbO$_2$ in the fissile material region as a function of time for the accumulation of $^{235}$UO$_2$ in a sphere of radius 0.244 m in bentonite - initially containing a minimum critical mass of $^{235}$UO$_2$. The largest arrival rate of $10^{-10}$ kg/m$^3$/s is equivalent to 0.19 g/year.

Figure 6.7: Power as a function of time for the accumulation of $^{235}$UO$_2$ in a sphere of radius 0.244 m in bentonite - initially containing a minimum critical mass of $^{235}$UO$_2$. The largest arrival rate of $10^{-10}$ kg/m$^3$/s is equivalent to 0.19 g/year.
Detailed results of QSS calculations for accumulation in bentonite are given in Tables 6.6 to 6.10. These relate, in turn, to each of the initial conditions in Table B.3 (Appendix B). If the initial system is identified as $^{235}\text{UO}_2$, then the accumulating material was $^{235}\text{UO}_2$ at all times. If the initial system has any $^{239}\text{PuO}_2$, then the arriving material (and that initially in the fissile material region) decays with time to have a larger fraction of $^{235}\text{UO}_2$. The ‘sourcedecay’ pre-processor to the QSS model [28] is used to determine how the arriving material changes with time. For very long duration events (compared with the 24,100 year half-life of $^{239}\text{Pu}$), the arriving material is close to 100% $^{235}\text{UO}_2$ for most of the duration of the transient.

In line with the approach of the UCuRC programme, arrival rates were chosen to be as wide ranging as possible, while limiting the temperature rise to be within the limits of the generated reactivity functions (Appendix B). Generally the arrival rates used were between $10^{-12}$ kg/m$^3$/s and $10^{-9}$ kg/m$^3$/s of oxide into the fissile material region.

For comparison, The Likelihood of Criticality programme has produced a model where fissile materials are removed from a plutonium or HEU waste package, and are transported with groundwater$^{37}$ into the surrounding bentonite [38]. For a specific example of HEU in higher strength rock, Figure 4.26 of [38] shows the results from a number of calculations where a maximum of about 25 kg of $^{235}\text{U}$ accumulates in a ring of bentonite around a failed waste package over a period of $10^8$ years. Differentiating this curve with respect to time shows that the peak mass transfer rate of $^{235}\text{U}$ to the bentonite region is about $6\times10^{-4}$ g/year. Converting this to units of kg/m$^3$/s for comparison with the QSS model is more difficult. The volume of the bentonite region used in the Likelihood of Criticality calculations is 5.66 m$^3$ (a ring of inner radius 0.45 m, outer radius 0.875 m, and height 3.2 m). Assuming the $^{235}\text{U}$ is uniformly distributed over this volume, the peak arrival rate would be less than $10^{-14}$ kg/m$^3$/s. Alternatively, assuming that all the mass were to accumulate in a region of minimum radius for a critical configuration (about 0.25 m from Figure 5.2 of this report) the peak arrival rate would be about $10^{-12}$ kg/m$^3$/s. Hence, this specific case

$^{37}$ Because of the long times involved both the plutonium and HEU packages are expected to exhibit similar behaviour since any $^{239}\text{Pu}$ initially present would have decayed to $^{235}\text{U}$. 
suggests that the arrival rates used in the QSS model are physically sensible, but generally larger than those from the analysis in [38].

Each of the tables of results (Tables 6.6 to 6.10) shows the general trends of the rule of thumb in maximum temperature, maximum power, and final temperature and power.

There are several further trends that can be identified from the tables of results. The key trends and observations are given below:

- In terms of the length scales of the results, the radius at which the temperature is raised by 10 °C or more is limited to 2.9 m for the largest maximum temperatures, and is much lower for many of the calculations undertaken. The radius to which the temperature rise is more than 1 °C can be tens of metres for the larger maximum temperatures.

- For all of the calculations in bentonite (i.e. the different fissile materials and arrival rates) the maximum power is no larger than 1 kW. This is consistent with results from the UCuRC programme, where for comparable mass arrival rates of 1.8 g/year, the maximum power for QSS calculations in granite and mudstone was a few kW.

- For a given host material, a QSS initiated from a mass greater than the minimum critical mass of $^{235}\text{UO}_2$ generally has lower consequences (temperature, power, and radial extent of temperature increases) than that for the minimum critical mass at comparable arrival rates. For example, in Tables 6.6 to 6.8, the maximum temperature rises for an arrival rate of $10^{-10}$ kg/m$^3$/s are 107 °C, 16 °C and 7.9 °C for initial $^{235}\text{UO}_2$ masses of 2.5 kg, 7.0 kg and 23.9 kg respectively. It is noted that the mass arrival rate for these calculations is 0.19, 0.077 and 0.073 g/year respectively. Hence, comparing on mass arrival rate, a scaling in the rate by about 0.4 reduces the peak temperature rise by a factor of about ten.

- For a given host material, a QSS initiated from a minimum critical mass with the presence of $^{239}\text{PuO}_2$ generally has larger consequences than that for the minimum critical mass of $^{235}\text{UO}_2$ alone. For example, comparing Tables 6.6 and 6.9, the temperatures and powers are larger in Table 6.9, which is an initially $^{239}\text{PuO}_2$ accumulation.

- Generally the peak temperature rise occurs early in the QSS transient (assuming that it could continue to the theoretical maximum duration). There are a few exceptions to this, but in general the observation is that should a QSS transient end earlier than its theoretical maximum duration then the peak temperature rise is likely to have passed.

Table 6.6: Summary of QSS outputs for a system starting from the minimum critical mass of $^{235}\text{UO}_2$ in bentonite for a range of fissile material arrival rates.

<table>
<thead>
<tr>
<th>Arrival rate (kg/m$^3$/s)</th>
<th>Arrival rate (g/year)</th>
<th>Time of maximum temp. (millennia)</th>
<th>Maximum average temp. rise (°C)</th>
<th>Maximum power (W)</th>
<th>Radius (m) of 10°C temp. rise at maximum average temp.</th>
<th>Radius (m) of 1°C temp. rise at maximum average temp.</th>
<th>End of calculation (millennia)</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1\times10^{-10}$</td>
<td>0.19</td>
<td>174</td>
<td>107</td>
<td>357</td>
<td>2.18</td>
<td>21.8</td>
<td>1,290</td>
<td>11.1</td>
<td>36.8</td>
</tr>
<tr>
<td>$1\times10^{-11}$</td>
<td>0.019</td>
<td>2,970</td>
<td>10.7</td>
<td>35.6</td>
<td>0.211</td>
<td>2.17</td>
<td>12,900</td>
<td>1.07</td>
<td>3.55</td>
</tr>
<tr>
<td>$1\times10^{-12}$</td>
<td>0.0019</td>
<td>30,300</td>
<td>1.04</td>
<td>3.5</td>
<td>0</td>
<td>0.203</td>
<td>129,000</td>
<td>0.11</td>
<td>0.35</td>
</tr>
</tbody>
</table>
Table 6.7: Summary of QSS outputs for a system starting from about three times the minimum critical mass of $^{235}$UO$_2$ in bentonite for a range of fissile material arrival rates.

<table>
<thead>
<tr>
<th>Arrival rate (kg/m$^3$/s)</th>
<th>Arrival rate (g/year)</th>
<th>Time of maximum temp. (millennia)</th>
<th>Maximum average temp. rise (°C)</th>
<th>Maximum power (W)</th>
<th>Radius (m) of 10°C temp. rise at maximum average temp.</th>
<th>Radius (m) of 1°C temp. rise at maximum average temp.</th>
<th>End of calculation (millennia)</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 \times 10^{-9}$</td>
<td>0.77</td>
<td>120</td>
<td>117</td>
<td>288</td>
<td>1.77</td>
<td>17.7</td>
<td>121</td>
<td>100</td>
<td>246</td>
</tr>
<tr>
<td>$1 \times 10^{-10}$</td>
<td>0.077</td>
<td>43</td>
<td>16.0</td>
<td>39.3</td>
<td>0.240</td>
<td>2.40</td>
<td>1,210</td>
<td>6.22</td>
<td>15.3</td>
</tr>
<tr>
<td>$1 \times 10^{-11}$</td>
<td>0.0077</td>
<td>502</td>
<td>1.90</td>
<td>4.71</td>
<td>0</td>
<td>0.288</td>
<td>12,100</td>
<td>0.60</td>
<td>1.48</td>
</tr>
<tr>
<td>$1 \times 10^{-12}$</td>
<td>0.00077</td>
<td>5,100</td>
<td>0.18</td>
<td>0.46</td>
<td>0</td>
<td>0</td>
<td>121,000</td>
<td>0.06</td>
<td>0.15</td>
</tr>
</tbody>
</table>

Table 6.8: Summary of QSS outputs for a system starting from about ten times the minimum critical mass of $^{235}$UO$_2$ in bentonite for a range of fissile material arrival rates.

<table>
<thead>
<tr>
<th>Arrival rate (kg/m$^3$/s)</th>
<th>Arrival rate (g/year)</th>
<th>Time of maximum temp. (millennia)</th>
<th>Maximum average temp. rise (°C)</th>
<th>Maximum power (W)</th>
<th>Radius (m) of 10°C temp. rise at maximum average temp.</th>
<th>Radius (m) of 1°C temp. rise at maximum average temp.</th>
<th>End of calculation (millennia)</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 \times 10^{-9}$</td>
<td>0.73</td>
<td>97</td>
<td>93.3</td>
<td>225</td>
<td>1.38</td>
<td>13.8</td>
<td>97</td>
<td>93.2</td>
<td>225</td>
</tr>
<tr>
<td>$1 \times 10^{-10}$</td>
<td>0.073</td>
<td>331</td>
<td>7.92</td>
<td>19.1</td>
<td>0</td>
<td>1.17</td>
<td>974</td>
<td>5.24</td>
<td>12.7</td>
</tr>
<tr>
<td>$1 \times 10^{-11}$</td>
<td>0.0073</td>
<td>3,350</td>
<td>0.91</td>
<td>2.21</td>
<td>0</td>
<td>0.109</td>
<td>9,740</td>
<td>0.50</td>
<td>1.21</td>
</tr>
<tr>
<td>$1 \times 10^{-12}$</td>
<td>0.00073</td>
<td>33,500</td>
<td>0.09</td>
<td>0.22</td>
<td>0</td>
<td>0</td>
<td>97,400</td>
<td>0.05</td>
<td>0.12</td>
</tr>
</tbody>
</table>

Table 6.9: Summary of QSS outputs for a system starting from the minimum critical mass of $^{239}$PuO$_2$ in bentonite for a range of fissile material arrival rates.

<table>
<thead>
<tr>
<th>Arrival rate (kg/m$^3$/s)</th>
<th>Arrival rate (g/year)</th>
<th>Time of maximum temp. (millennia)</th>
<th>Maximum average temp. rise (°C)</th>
<th>Maximum power (W)</th>
<th>Radius (m) of 10°C temp. rise at maximum average temp.</th>
<th>Radius (m) of 1°C temp. rise at maximum average temp.</th>
<th>End of calculation (millennia)</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 \times 10^{-10}$</td>
<td>0.21</td>
<td>43</td>
<td>138</td>
<td>463</td>
<td>2.91</td>
<td>29.1</td>
<td>1,290</td>
<td>53.5</td>
<td>185</td>
</tr>
<tr>
<td>$1 \times 10^{-11}$</td>
<td>0.021</td>
<td>59</td>
<td>23.1</td>
<td>79.8</td>
<td>0.488</td>
<td>4.88</td>
<td>12,900</td>
<td>5.13</td>
<td>17.7</td>
</tr>
<tr>
<td>$1 \times 10^{-12}$</td>
<td>0.0021</td>
<td>63</td>
<td>11.1</td>
<td>38.2</td>
<td>0</td>
<td>2.34</td>
<td>129,000</td>
<td>0.50</td>
<td>1.74</td>
</tr>
</tbody>
</table>
Table 6.10: Summary of QSS outputs for a system starting from the minimum critical mass of $^{239}$PuO$_2$: $^{235}$UO$_2$ in ratio 50:50 in bentonite for a range of fissile material arrival rates.

<table>
<thead>
<tr>
<th>Arrival rate (kg/m$^3$/s)</th>
<th>Arrival rate (g/year)</th>
<th>Time of maximum temp. (millennia)</th>
<th>Maximum average temp. rise (°C)</th>
<th>Maximum power (W)</th>
<th>Radius (m) of 10°C temp. rise at maximum average temp.</th>
<th>Radius (m) of 1°C temp. rise at maximum average temp.</th>
<th>End of calculation (millennia)</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 \times 10^{-10}$</td>
<td>0.21</td>
<td>39</td>
<td>119</td>
<td>408.</td>
<td>2.50</td>
<td>25.0</td>
<td>1,290</td>
<td>52.7</td>
<td>180</td>
</tr>
<tr>
<td>$1 \times 10^{-11}$</td>
<td>0.021</td>
<td>78</td>
<td>12.5</td>
<td>42.7</td>
<td>0.262</td>
<td>2.62</td>
<td>12,900</td>
<td>1.70</td>
<td>16.4</td>
</tr>
<tr>
<td>$1 \times 10^{-12}$</td>
<td>0.0021</td>
<td>87</td>
<td>1.70</td>
<td>5.82</td>
<td>0</td>
<td>0.357</td>
<td>129,000</td>
<td>0.47</td>
<td>1.60</td>
</tr>
</tbody>
</table>

6.3.2 Scoping Calculations for the In-Package Scenario for PWR Fuel

QSS calculations (using the QSS model with varying volume) for an in-package scenario for PWR fuel were completed using a reactivity function constructed using the methods described in Appendix B (Section B.2.2) and consistent rates for geometrical change (the rate of increase of the flooded region of the disposal package) and material arrival$^{38}$. The observation that all of the $k_{\text{effective}}$ values in the reactivity function are significantly sub-critical at 300 °C (since by this temperature the water would have vaporised, greatly reducing the density of H$_2$O in the void space surrounding the fuel pins) is noted in Appendix B (Section B.2.2) suggesting that a temperature approaching 300 °C would not be possible in any QSS calculation.

A range of rates of change for the height of the flooded region of the disposal canister were considered, leading to temperature increases of a few °C to just above 200 °C. Since the QSS model was designed for geometries where the fissile material region is either a homogeneous sphere or cylinder$^{39}$, and neither of these applies for the in-package flooding scenario, there are different ways in which heat transfer terms could be implemented.

Two different cases were considered, both for a cylindrical geometry, but with different assumptions for heat transfer. In the first case the radius of the fissile material region (i.e. the region which heats up during a QSS transient criticality event) was taken to be the outer radius of the copper canister (0.45 m), while the height was that of the flooded region. The thermal conductivity of bentonite was then used for the surrounding region. In the second case an effective radius for the four fuel elements was used$^{39}$, the height was that of the flooded region, and the thermal conductivity of steel was used for the surrounding material, steel being another possibility for the outer canister (instead of copper), and also assumed to have similar properties to the cast iron insert which surrounds the four fuel compartments in a package. The thermal conductivity values used are given in Appendix A (Section A.2.2). In reality the heat conduction would be through cast iron, copper and bentonite, which is beyond the design of the QSS model. The two cases of bentonite and steel are considered sensible candidates for lower and upper bounds on thermal conductivity. However, since the bentonite would limit the heat transfer (it has the lowest thermal conductivity) the calculations using the thermal conductivity of bentonite are considered to be the most realistic.

The results of key outputs from QSS calculations for different rates of flooding are given in Table 6.11 (for the approximation using the thermal conductivity of bentonite) and Table 6.12 (using the thermal conductivity of steel). A wide range of flooding rates from $10^{-14}$ m/s (~14.3 million years to flood the 4.5 m fuel compartments) to $10^{-3}$ m/s (1.25 hours to flood the compartments) were considered since, dependent on the mechanism for flooding, the rate could be relatively fast (e.g. a

$^{38}$ In the QSS model the rate of addition (or arrival) of fuel (5% enriched uranium) to the reacting region is proportional to the rate at which the height of the flooded region increases.

$^{39}$ Each of the four compartments has a square cross-section, of side length 0.234 m. Hence four compartments have an area of 0.22 m$^2$. This is equivalent to a circle of radius 0.264 m.
sudden weld failure following geological activity) to very slow (e.g. slow water ingress due to pin-hole corrosion through the package canister and insert) and it is difficult to judge which of the rates (if any) would be the most realistic.

Table 6.11: Summary of QSS outputs for a PWR disposal canister system starting from a just-critical configuration, and with different rates of flooding. Thermal conduction of bentonite assumed for heat transfer.

<table>
<thead>
<tr>
<th>Rate of increase of water height (m/s)</th>
<th>Time to flood the canister length of 4.5 m</th>
<th>Time of maximum temp.</th>
<th>Max. average temp. rise (°C)</th>
<th>Max. power (W)</th>
<th>End of calculation</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1×10⁻³</td>
<td>1.25 hours</td>
<td>1.16 hours</td>
<td>165</td>
<td>1,460</td>
<td>1.16 hours</td>
<td>165</td>
<td>1,460</td>
</tr>
<tr>
<td>1×10⁻⁴</td>
<td>12.5 hours</td>
<td>11.6 hours</td>
<td>165</td>
<td>1,460</td>
<td>11.6 hours</td>
<td>165</td>
<td>1,460</td>
</tr>
<tr>
<td>1×10⁻⁵</td>
<td>5.2 days</td>
<td>4.8 days</td>
<td>165</td>
<td>1,460</td>
<td>4.8 days</td>
<td>165</td>
<td>1,460</td>
</tr>
<tr>
<td>1×10⁻⁶</td>
<td>52 days</td>
<td>48 days</td>
<td>165</td>
<td>1,460</td>
<td>48 days</td>
<td>165</td>
<td>1,460</td>
</tr>
<tr>
<td>1×10⁻⁷</td>
<td>1.4 years</td>
<td>1.32 years</td>
<td>165</td>
<td>1,460</td>
<td>1.32 years</td>
<td>165</td>
<td>1,460</td>
</tr>
<tr>
<td>1×10⁻⁸</td>
<td>14 years</td>
<td>13.2 years</td>
<td>165</td>
<td>1,460</td>
<td>13.2 years</td>
<td>165</td>
<td>1,460</td>
</tr>
<tr>
<td>1×10⁻⁹</td>
<td>140 years</td>
<td>132 years</td>
<td>165</td>
<td>1,460</td>
<td>132 years</td>
<td>165</td>
<td>1,460</td>
</tr>
<tr>
<td>1×10⁻¹⁰</td>
<td>1.4 millennia</td>
<td>1.32 millennia</td>
<td>165</td>
<td>1,459</td>
<td>1.32 millennia</td>
<td>165</td>
<td>1,459</td>
</tr>
<tr>
<td>1×10⁻¹¹</td>
<td>14 millennia</td>
<td>13.2 millennia</td>
<td>164</td>
<td>1,450</td>
<td>13.2 millennia</td>
<td>164</td>
<td>1,450</td>
</tr>
<tr>
<td>1×10⁻¹²</td>
<td>140 millennia</td>
<td>13.1 millennia</td>
<td>160</td>
<td>1,190</td>
<td>132 millennia</td>
<td>109</td>
<td>959</td>
</tr>
<tr>
<td>1×10⁻¹³</td>
<td>1,400 millennia</td>
<td>103 millennia</td>
<td>32.6</td>
<td>219</td>
<td>1320 millennia</td>
<td>12.8</td>
<td>113</td>
</tr>
<tr>
<td>1×10⁻¹⁴</td>
<td>14,000 millennia</td>
<td>957 millennia</td>
<td>3.39</td>
<td>22.5</td>
<td>13200 millennia</td>
<td>1.30</td>
<td>11.5</td>
</tr>
</tbody>
</table>

Table 6.12: Summary of QSS outputs for a PWR disposal canister system starting from a just-critical configuration, and with different rates of flooding. Thermal conduction of steel assumed for heat transfer.

<table>
<thead>
<tr>
<th>Rate of increase of water height (m/s)</th>
<th>Time to flood the canister length of 4.5 m</th>
<th>Time of maximum temp.</th>
<th>Max. average temp. rise (°C)</th>
<th>Max. power (W)</th>
<th>End of calculation</th>
<th>Average temp. rise at end (°C)</th>
<th>Power at end (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1×10⁻³</td>
<td>1.25 hours</td>
<td>1.16 hours</td>
<td>165</td>
<td>10,800</td>
<td>1.16 hours</td>
<td>165</td>
<td>10,800</td>
</tr>
<tr>
<td>1×10⁻⁴</td>
<td>12.5 hours</td>
<td>11.6 hours</td>
<td>165</td>
<td>10,800</td>
<td>11.6 hours</td>
<td>165</td>
<td>10,800</td>
</tr>
<tr>
<td>1×10⁻⁵</td>
<td>5.2 days</td>
<td>4.8 days</td>
<td>165</td>
<td>10,800</td>
<td>4.8 days</td>
<td>165</td>
<td>10,800</td>
</tr>
<tr>
<td>1×10⁻⁶</td>
<td>52 days</td>
<td>48 days</td>
<td>165</td>
<td>10,800</td>
<td>48 days</td>
<td>165</td>
<td>10,800</td>
</tr>
<tr>
<td>1×10⁻⁷</td>
<td>1.4 years</td>
<td>1.32 years</td>
<td>165</td>
<td>10,800</td>
<td>1.32 years</td>
<td>165</td>
<td>10,800</td>
</tr>
<tr>
<td>1×10⁻⁸</td>
<td>14 years</td>
<td>13.2 years</td>
<td>165</td>
<td>10,800</td>
<td>13.2 years</td>
<td>165</td>
<td>10,800</td>
</tr>
<tr>
<td>1×10⁻⁹</td>
<td>140 years</td>
<td>132 years</td>
<td>165</td>
<td>10,800</td>
<td>132 years</td>
<td>165</td>
<td>10,800</td>
</tr>
<tr>
<td>1×10⁻¹⁰</td>
<td>1.4 millennia</td>
<td>1.32 millennia</td>
<td>165</td>
<td>10,700</td>
<td>1.32 millennia</td>
<td>165</td>
<td>10,700</td>
</tr>
<tr>
<td>1×10⁻¹¹</td>
<td>14 millennia</td>
<td>3.7 millennia</td>
<td>161</td>
<td>10,000</td>
<td>13.2 millennia</td>
<td>137</td>
<td>8,920</td>
</tr>
<tr>
<td>1×10⁻¹²</td>
<td>140 millennia</td>
<td>10 millennia</td>
<td>38.4</td>
<td>2,110</td>
<td>132 millennia</td>
<td>17.2</td>
<td>1,120</td>
</tr>
<tr>
<td>1×10⁻¹³</td>
<td>1,400 millennia</td>
<td>92 millennia</td>
<td>4.09</td>
<td>222</td>
<td>1320 millennia</td>
<td>1.76</td>
<td>115</td>
</tr>
</tbody>
</table>

For the lowest flooding rates (i.e. 10⁻¹² m/s and below) the high thermal conductivity case (Table 6.12) has a temperature rise for a given flooding rate that is an order of magnitude lower than that for the low thermal conductivity case (Table 6.11), while the powers are similar. This is consistent with the higher thermal conductivity being able to conduct more heat away from the fissile region.
At the lowest flooding rates the results in Tables 6.11 and 6.12 show that an increase in the rate by ten increases the power and temperature by ten. At higher flooding rates the behaviour is different, and consistent with the observation that all of the $k_{\text{effective}}$ values in the reactivity function are significantly sub-critical at 300 °C. For all flooding rates of $10^{-11}$ m/s and above an increase in the flooding rate by ten reduces the time (but not the values) of the peak temperature and power by a factor of ten.

A key observation is that for all of the higher flooding rates ($10^{-11}$ m/s and above), the temperature rise is bounded by 165 °C, and the power is less than 2 kW for the calculations assuming thermal conduction in bentonite. The power is less than 11 kW if the thermal conductivity of steel is used, although, as noted above, the bentonite conduction case is considered more realistic. It is interesting to note that these power levels and temperature rises are comparable with those expected during the earlier stages of spent fuel disposal where the radioactive decay of activated materials would produce heat within the waste packages which would lead to temperature increases in the canister and the surrounding buffer. Figure 21 in [9], for example, shows a temperature rise of up to about 80 °C over a period of a few thousand years post-closure for a spent fuel canister based on a Canadian spent fuel disposal concept.

Figures 6.9 to 6.15 provide more details on the evolution of the QSS transient criticality calculations for different flooding rates. The results shown are for the case using the thermal conductivity of bentonite for heat transfer. In general Figures 6.9 to 6.15 show that (for flooding rates of $10^{-12}$ m/s or lower) the behaviour of the in-package calculations follows similar behaviour to QSS accumulation calculations where the arrival rate is replaced by the rate of increase in flooded height. For these low flooding rates the results show that increasing the flooding rate by a factor of ten:

- reduces the duration of the QSS transient by a factor of ten (Figures 6.9 to 6.15);
- increases the temperature rise by a factor of about ten (Figure 6.9);
- stretches the evolution of the mass of $^{235}$UO$_2$ (Figure 6.10) and $^{238}$UO$_2$ (Figure 6.11) over the duration of the transient. The same is true for the capture product $^{239}$UO$_2$ and the representative fission product PbO$_2$.
- increases the power (Figure 6.14) and neutron flux (Figure 6.15) by a factor of about ten.

For flooding rates of $10^{-11}$ m/s and above the behaviour is different. Not only does the temperature rise above ambient plateau at about 165 °C, but the burn-up of $^{235}$UO$_2$ is much reduced compared with calculations at lower flooding rates (if there was no burn-up at all the final mass of $^{235}$UO$_2$ would be 150.3 kg). Furthermore, the peak temperature, power and flux are broadly similar with those for a flooding rate of $10^{-12}$ m/s, so do not follow the trends at lower flooding rates - otherwise they would increase ten times for each factor of ten increase in the flooding rate.

The observation of a temperature plateau is consistent with the observation that there are no critical configurations in the reactivity function at 300 °C (Appendix B, Section B.2.2) and demonstrate a ‘ceiling’ on the temperature to somewhere between 200 °C and 300 °C. Assuming that the pressure has reached equilibrium with the surrounding bentonite (assumed to be saturated at a pressure of 6.5 MPa at a GDF depth of 650 m) the ‘ceiling’ temperature would be more like 281 °C, which is the vapourisation temperature of water at this pressure. The reactivity function constructed and used for the QSS calculations does not have enough temperature interpolation points to capture this. If the in-package pressure has not equalised with that of the surrounding saturated bentonite then the vapourisation temperature would be lower.

Figures 6.12 and 6.13 demonstrate that at the lower flooding rates ($10^{-12}$ m/s and lower) the slower the flooding rate (and hence the longer the duration of the transient) and the lower the build up of $^{239}$PuO$_2$ and $^{240}$PuO$_2$. This is entirely consistent with the longer duration events having more time for radioactive decay to affect any plutonium produced from capture in $^{238}$UO$_2$. At the higher flooding rates ($10^{-11}$ m/s and above) very little plutonium is produced since, as the flooding rate
increases (and the duration decreases) with the same maximum neutron flux, less of the uranium is burned up and fewer neutrons are captured to produce plutonium. For all of the calculations the mass of $^{239}$PuO$_2$ is less than 0.05% of the mass of $^{238}$UO$_2$.

**Figure 6.9:** Average temperature increase of the fissile material region as a function of time for an in-package scenario for a PWR fuel disposal canister. Thermal conduction of bentonite assumed for heat transfer. For the flooding rates $1 \text{ pm/s} = 10^{-12} \text{ m/s}$. The lowest flooding rate is equivalent to 14.3 million years to flood a 4.5 m canister. The highest flooding rate is equivalent to 1.25 hours for flooding.

**Figure 6.10:** Mass of $^{235}$UO$_2$ in the flooded region as a function of time for an in-package scenario for a PWR fuel disposal canister. Thermal conduction of bentonite assumed for heat transfer. For the flooding rates $1 \text{ pm/s} = 10^{-12} \text{ m/s}$. 
Figure 6.11: Mass of $^{238}\text{UO}_2$ in the flooded region as a function of time for an in-package scenario for a PWR fuel disposal canister. Thermal conduction of bentonite assumed for heat transfer. For the flooding rates $1\text{ pm/s} = 10^{-12}\text{ m/s}$.

Figure 6.12: Mass of $^{239}\text{PuO}_2$ in the flooded region as a function of time for an in-package scenario for a PWR fuel disposal canister. Thermal conduction of bentonite assumed for heat transfer. For the flooding rates $1\text{ pm/s} = 10^{-12}\text{ m/s}$.
Figure 6.13: Mass of $^{240}\text{PuO}_2$ in the flooded region as a function of time for an in-package scenario for a PWR fuel disposal canister. Thermal conduction of bentonite assumed for heat transfer. For the flooding rates $1 \text{ pm/s} = 10^{-12} \text{ m/s}$.

Figure 6.14: Power as a function of time for an in-package flooding scenario for a PWR fuel disposal canister. Thermal conduction of bentonite assumed for heat transfer. For the flooding rates $1 \text{ pm/s} = 10^{-12} \text{ m/s}$.
Should a hypothetical in-package criticality event reach temperatures where the water in the voids around the fuel vaporises, then there are some important questions to raise about the modelling required. One consideration is that the production of steam may stop the continued flooding of the fuel compartments. Steam trying to escape from the disposal canister could pressurise it relative to the surroundings, slowing or preventing the ingress of water. If this happened then a situation could be envisaged where flooding is temporarily paused. The nuclear reaction would therefore pause, and since no further heat would be produced the fissile region of the model would cool. After sufficient cooling, flooding could resume, providing the possibility that the system may establish another QSS. Theoretically this could lead to oscillatory behaviour.

Another important consideration is possible chemical reactions in the fuel elements, should the water in the void space vaporise. Reactions could include steam reacting with the fuel clad, for example, with the potential of producing gases or degrading the clad and allowing fuel pins to distort and degrade.

Such potential effects are beyond the capability of the QSS model, but may be of interest in quantifying secondary effects from hypothetical criticality events.

6.4 Summary

The results from criticality consequence calculations summarised above provide an overview of the possible consequences of post-closure criticality for hypothetical criticality events for the disposal of HLW, SF, plutonium and HEU. The completion of additional calculations (as specified in Section 5.3), including the analysis of the consequences of hypothetical criticality events for the ‘what-if’ in-package scenario, are an important addition to calculations from the UCoRC programme, and for ILW, LLW and DNLEU disposal. The results provide valuable trends and observations for hypothetical criticality events, and show limits on the extent of cavity radius and cracking for rapid transient events, and on the temperature and power for quasi-steady-state events. A small number of extreme cases are included in the calculations where, while the likelihood of criticality is expected to be vanishingly small, a demonstration of low localised consequences can also be considered valuable.
Together with static analysis to understand whether ‘what-if’ scenarios for HLW, SF, plutonium and HEU disposal could conceivably result in critical configurations, the range of criticality consequences calculations undertaken is considered to satisfy the programme objective to:

- apply the UCuRC QSS model and Bounding Approach model to scope the consequences of criticality transients in other materials and illustrative concepts (the models need to be developed and applied sufficiently to meet safety case and stakeholder requirements for the current preparatory studies phase).
7 Summary and Conclusions

7.1 Summary

This report is one of two reports describing how the transient criticality models have been applied to understand the potential consequences of ‘what-if’ post-closure criticality scenarios. This report focuses on scenarios following the disposal of HLW, SF, plutonium and HEU, whereas the second report focuses on scenarios following the disposal of ILW, LLW and DNLEU. The work described in both reports is bounded by the 2007 Radioactive Waste Inventory, and the illustrative disposal concepts of the 2010 DSSC.

One method for identifying criticality scenarios for further investigation is the use of Features, Events and Processes (FEPs) analysis. Combinations of FEPs, each of which represents some change to the original sub-critical configuration, form ‘what-if’ scenarios of interest. Building on an overview of the illustrative disposal concepts, together with information on package and near-field evolution behaviour, this report summarises an alternative, and complementary, approach to FEP analysis to identify ‘what-if’ scenarios for criticality. Through the consideration of criticality controls such as fissile mass and concentration, two ‘what-if’ scenarios are identified:

- Localised accumulation of fissile materials from one or more waste packages in any region of a GDF or the host rocks;
- The in-package scenario for a single waste package. This is applicable to waste packages that may contain sufficient fissile material for a critical configuration given significant package evolution. It could apply to some SF, plutonium and HEU but not HLW. For a critical configuration to develop, partial flooding with water or significant degradation and rearrangement of the package contents would be necessary.

The methodology used in this work to understand the consequences of criticality is:

- Determination of what configurations of fissile material would have to develop for hypothetical criticality events to occur, by undertaking static criticality calculations with criticality software. For the research reported here, analysis has been extended from that achieved during the UCuRC programme by the addition of calculations for in-package scenarios. For SF the analysis focused on the partial flooding of fuel packages containing Pressurised Water Reactor (PWR) fuel. The analysis includes calculations of the axial variation in irradiation along fuel elements for different levels of average fuel irradiation. For plutonium and HEU waste packages the analysis focused on the removal of neutron absorbers and dissolution of packaging materials.
- Determination of how hypothetical critical configurations could evolve as either rapid transient or quasi-steady-state criticality events. This involves determining whether an increase in temperature acts to make a system more or less critical. The RTM or Bounding Approach model is required for the former and the QSS model for the latter.
- Specification of representative calculations for criticality consequence analysis using the transient criticality models, to sufficiently scope parameter space and identify trends.
- Decisions on how to apply the models to the specified calculations, including the construction of model inputs.
- Application of the models to the specified calculations and analysis of the results.

Using the above methodology has produced valuable insight into the consequences of a range of hypothetical criticality events for two ‘what-if’ scenarios for post-closure criticality. Section 7.2 describes the main conclusions from the research undertaken. To reach these conclusions it is emphasised that a number of modelling assumptions are required for the underlying criticality analysis; Appendix C provides an overview of the assumptions.
7.2 Conclusions

For the accumulation scenario it is concluded from static criticality calculations for idealised spherical accumulations that:

- Critical masses are hypothetically possible in bentonite (used as a buffer around waste packages), granite (higher strength rock) and clay (lower strength sedimentary rock). A critical mass is not possible in evaporite rock for the specific material composition and porosity considered, but might be possible for a different chemical composition or a larger porosity. For the granite material composition no critical systems are possible for low enriched uranium.

- Where critical systems are possible, a specified fissile material will have a minimum critical mass, below which criticality is not possible.
  - For any host material the accumulation of $^{239}\text{PuO}_2$ provides the lowest value for the minimum critical mass. Therefore, since the minimum critical mass for a $^{239}\text{UO}_2$ accumulation is larger than for a $^{239}\text{PuO}_2$ accumulation in a given host material, the fissile mass required for criticality will increase as $^{239}\text{Pu}$ decays to $^{235}\text{U}$.
  - For the host materials where the minimum critical mass is the lowest ($^{239}\text{PuO}_2$ accumulation in bentonite) at least 1 kg of $^{239}\text{PuO}_2$ must accumulate. For other host materials the minimum critical mass for $^{239}\text{PuO}_2$ accumulation is larger.
  - If $^{238}\text{UO}_2$ is present, then the minimum critical mass can be much larger. In bentonite, for example, 100% enriched uranium has a minimum critical mass of 2.5 kg of $^{235}\text{UO}_2$, whereas for 3% enriched uranium the minimum mass increases to 630 kg of $\text{UO}_2$, of which 19 kg is $^{235}\text{UO}_2$.
  - Generally, as the porosity of the host rock decreases, the minimum critical mass for a given fissile material increases. For example, with an assumed porosity of 12% the minimum critical mass for $^{239}\text{PuO}_2$ in clay is 3.1 kg, whereas for granite with an assumed porosity of 1% the minimum critical mass for $^{239}\text{PuO}_2$ is more than ten times larger at 44 kg. In addition to porosity, the chemical composition of the host rock can also have a significant effect on the critical mass.

- In any host material the fissile mass required for a critical configuration would require the fissile contents of multiple HLW packages to accumulate, so that this scenario is not considered credible for HLW.

- The majority of hypothetical accumulation systems would evolve as quasi-steady-states, if maintained by the continued accumulation of fissile materials.

- Rapid transient criticality, which is only possible for systems where increases in temperature make the system more critical, requires sufficient $^{239}\text{Pu}$ to be present. Furthermore, rapid transient criticality is only possible in a narrow range of fissile concentrations (5 kg/m$^3$ for the accumulation of $^{239}\text{PuO}_2$ in bentonite, for example), and this concentration range as $^{239}\text{Pu}$ decays, with a half life of 24,100 years.

- As $^{239}\text{Pu}$ decays to $^{235}\text{U}$ the mass of fissile material required for rapid transient criticality increases significantly. For example, in bentonite, for a rapid transient requires at least 2.6 kg of $^{239}\text{PuO}_2$. After four half-lives at least 20 kg of $^{239}\text{PuO}_2$ and $^{235}\text{UO}_2$, in ratio 6.25:93.75 due to decay, would be required.

- The above trends suggest that the longer it takes for a hypothetical critical accumulation of fissile material to occur, the less credible a rapid transient criticality becomes, with the expectation that rapid transient criticality is not credible for hypothetical critical accumulations that require post-closure timescales of at least 100,000 years to develop.

For the accumulation scenario it is concluded from transient criticality calculations for idealised spherical accumulations that:

- For systems that could evolve as quasi-steady-states the QSS model predicts that for mass arrival rates of the order of 1 g/year, or lower, the power is limited to a few kilowatts, and temperature rises of more than 10 °C above ambient are limited to a localised radius of a few metres. Larger consequences are possible at higher rates of accumulation, but only if it is
assumed that fissile accumulation can continue when the pore water boils. Pore water boiling will generally act to not only prevent accumulation of fissile material, but also acts to reduce neutron moderation.

- The rule of thumb for QSS calculations is a powerful tool in understanding how results will change with different fissile material arrival rates. The rule, which has been confirmed for a wide range of calculations, states that, provided the temperature increase is not sufficient for pore water to boil then an increase in the arrival rate by a factor of \( X \):
  - will scale the peak temperature increase (above ambient) and power by a factor of \( X \); but
  - those peaks will occur at a time scaled by \( 1/X \).

In many cases the rule applies even if the arrival rates are sufficient to boil pore water in the accumulation region.

- For systems that could evolve as rapid transients, considering 10 kg of fissile oxide as an illustrative mass for accumulation, the Bounding Approach model predicts an energy release which could create a local cavity radius of between two to six metres. Local cracking would extend to no more than a few tens of metres in radius. Furthermore, as \(^{239}\text{Pu}\) decays to \(^{235}\text{U}\), the consequences of rapid transient events reduce. Larger mass accumulations, which are considered less likely, and would initially take longer to accumulate, would have larger consequences. However, the relationship is weaker than linear; for example, increasing the mass of \(^{239}\text{PuO}_2\) by a factor of ten scales the cavity radius by a factor of about three. The relationship is weaker still if the decay of \(^{239}\text{Pu}\) is taken into account.

In-package scenarios are only credible for waste packages that contain sufficient fissile mass. They are therefore conceivable, but considered unlikely, for spent fuel, plutonium and HEU, but not HLW. For in-package scenarios the conclusions of static criticality calculations are:

- For HEU and plutonium wastes, assuming the can-in-canister packaging concept with a ceramic wasteform within cans surrounded by borosilicate glass within an outer canister, it is concluded that:
  - The illustrative wasteform and package design will not become critical even if the neutron absorbers are removed from the ceramic wasteform, and the borosilicate glass within the canister is completely dissolved and replaced by water.
  - There is therefore no requirement for in-package transient criticality analysis for HEU and plutonium wasteforms disposed of using this concept.

- For spent fuel waste, static criticality analysis for fresh and irradiated PWR fuel in an illustrative package design containing four PWR fuel elements, allows the following conclusions to be drawn:
  - Both vertically and horizontally oriented spent PWR fuel packages could, hypothetically, become critical, following partial package flooding with water. However, they would not become critical from flooding if the average irradiation of each fuel element is greater than 35 GWd/Te. The average irradiation assumed for PWR fuel is 55 GWd/Te.
  - For low or zero irradiated fuel, where transient criticality events are hypothetically possible from the flooding of a package, only quasi-steady-state transient criticality could result.

For the in-package scenario, assuming a worst-case of flooding within a vertically oriented PWR fuel package containing four fresh fuel elements, it is concluded from transient criticality calculations with the QSS model that:

- For a large range of assumed rates of flooding of the void space within the package, from hours to thousands of years, the peak temperature and power of the transient do not vary.

- Assuming transients where the heat transfer is dominated by the thermal properties of bentonite for the conduction of heat away from the critical region, the maximum power of the criticality transient is less than two kilowatts. It is noted that this power is similar to that which packages could be subject to from the decay heat of irradiated fuel in the earlier stages of emplacement within a GDF.
If the thermal properties of steel are used instead, representing heat transfer dominated by the thermal properties of the package insert, then the QSS model predicts a larger maximum power of about ten kilowatts. This is considered less realistic than the bentonite case, but indicates the sensitivity of model outputs to assumed material properties.

These conclusions and the underlying research are an important component in demonstrating that the consequences of hypothetical post-closure criticality events are not a significant concern. The analysis of in-package scenarios is a significant extension to the research undertaken during the UCuRC programme.

Acknowledgements

The authors would like to thank NDA RWMD for their investment in the development of the transient criticality models and, in particular, Peter Wood for championing the post-closure criticality programme. They would also like to thank RWMD staff and Colin Zimmerman for guidance on the structure of this document and for providing useful feedback and comments. For the analysis and calculations presented in this report, the authors gratefully acknowledge work undertaken by David Hanlon, Tara Hanlon, David Powney and verification by Julie Martin. Thanks are also given to Galson Sciences for some of the Tables reproduced in Section 2 of this report, and to International Nuclear Services (INS) and Sellafield Ltd for supply of MONK input models for PWR and Plutonium/HEU waste packages.
References


|---|---|


## Appendices

### Contents

<table>
<thead>
<tr>
<th>Appendix</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Appendix A</td>
<td>Material Properties for Static and Transient Criticality Analysis</td>
</tr>
<tr>
<td>Appendix B</td>
<td>Reactivity Functions for the Transient Criticality Models</td>
</tr>
<tr>
<td>Appendix C</td>
<td>Summary of Assumptions</td>
</tr>
</tbody>
</table>
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Appendix A
Material Properties for Static and Transient Criticality Analysis

This appendix summarises information required for different materials to facilitate either static criticality calculations (of whether, or under what conditions a system can be critical), or transient criticality calculations, from which the consequences of hypothetical criticality events are calculated.

A.1 Composition Data for Materials

This section summarises information on the chemical composition of materials within the different illustrative disposal concepts for a GDF.

A.1.1 Bentonite

Criticality calculations were not undertaken for bentonite during the UCuRC programme, so this was an additional material for the research programme reported here. Table A.1 summarises suitable information for bentonite.

Table A.1: Composition details for dry bentonite.

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Weight fraction</th>
<th>Element</th>
<th>Weight fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO$_2$</td>
<td>0.6740</td>
<td>Al</td>
<td>0.1116</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>0.2120</td>
<td>C</td>
<td>0.0036</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
<td>0.0414</td>
<td>Ca</td>
<td>0.0104</td>
</tr>
<tr>
<td>MgO</td>
<td>0.0261</td>
<td>Fe</td>
<td>0.0288</td>
</tr>
<tr>
<td>CaO</td>
<td>0.0146</td>
<td>K</td>
<td>0.0045</td>
</tr>
<tr>
<td>Na$_2$O</td>
<td>0.0225</td>
<td>Mg</td>
<td>0.0157</td>
</tr>
<tr>
<td>K$_2$O</td>
<td>0.0055</td>
<td>Na</td>
<td>0.0166</td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>0.0017</td>
<td>O</td>
<td>0.4908</td>
</tr>
<tr>
<td>P$_2$O$_5$</td>
<td>0.0005</td>
<td>P</td>
<td>0.0002</td>
</tr>
<tr>
<td>C</td>
<td>0.0036</td>
<td>S</td>
<td>0.0034</td>
</tr>
<tr>
<td>S</td>
<td>0.0034</td>
<td>Si</td>
<td>0.3134</td>
</tr>
<tr>
<td>Ti</td>
<td>0.0010</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

| Solid Density | 2780 kg m$^{-3}$ |
| Porosity      | 41%             |
| Density of water in the pores | 995 kg m$^{-3}$ |
In Table A.1 the solid density and porosity in the left columns are taken from [58] (Table 2.2 for bentonite blocks\(^{40}\)). The mineral composition is taken from [59] (the Wy mean row of Table 4.2). Using information on the molecular weights of the required elements, the right hand columns were calculated and provide an elemental composition suitable for use in MONK. Note that the molecular weight fractions sum to slightly more than unity, so these were normalised before calculating the elemental composition.

### A.1.2 Granite

Granite is a high strength igneous rock, and is therefore a suitable choice for calculations in the illustrative higher strength rock disposal concept. Some static criticality calculations were undertaken in granite during the UCuRC programme [11], but to a lesser extent than for NRVB. Table A.2 summarises the composition of granite. The left hand columns present the composition in terms of the weight fractions of the dominant molecules, and are reproduced from [22]. The quoted density is taken to be that of the solid with no porosity. Using information on the molecular weights of the required elements, the right hand columns were calculated and provide an elemental composition suitable for use in MONK. The porosity of 1\% is rounded from 0.72\%, given as a best estimate in [49].

#### Table A.2: Composition details for dry granite.

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Weight fraction</th>
<th>Element</th>
<th>Weight fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO(_2)</td>
<td>0.7357</td>
<td>Si</td>
<td>0.34389</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td>0.1300</td>
<td>Al</td>
<td>0.06880</td>
</tr>
<tr>
<td>K(_2)O</td>
<td>0.0400</td>
<td>K</td>
<td>0.03318</td>
</tr>
<tr>
<td>Na(_2)O</td>
<td>0.0400</td>
<td>Na</td>
<td>0.02967</td>
</tr>
<tr>
<td>CaO</td>
<td>0.0100</td>
<td>Ca</td>
<td>0.00713</td>
</tr>
<tr>
<td>Fe(_2)O(_3)</td>
<td>0.0200</td>
<td>Fe</td>
<td>0.02957</td>
</tr>
<tr>
<td>FeO</td>
<td>0.0200</td>
<td>Ti</td>
<td>0.00240</td>
</tr>
<tr>
<td>TiO(_2)</td>
<td>0.0040</td>
<td>H</td>
<td>0.00003</td>
</tr>
<tr>
<td>H(_2)O in rock</td>
<td>0.0003</td>
<td>O</td>
<td>0.48532</td>
</tr>
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<table>
<thead>
<tr>
<th>Solid Density</th>
<th>2673 kg m(^{-3})</th>
</tr>
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<tr>
<td>Porosity</td>
<td>1%</td>
</tr>
<tr>
<td>Density of water in the pores</td>
<td>995 kg m(^{-3})</td>
</tr>
</tbody>
</table>

### A.1.3 Clay

Clay is a lower strength sedimentary rock, and is therefore a suitable choice for calculations in the illustrative lower strength sedimentary rock disposal concept. A small number of criticality calculations were undertaken for lower strength sedimentary rock during the UCuRC programme [23], using a material specification for mudstone, or Opalinus clay [60]. The left hand columns in Table A.3 present the composition in terms of the weight fractions of the dominant molecules. This is reproduced from [60]. The quoted density is taken to be that of the solid with no porosity. Using information on the molecular weights of the required elements, the right hand columns can be calculated and provide an elemental composition suitable for use in MONK. Note that the

\(^{40}\) It is noted that while the porosity for dry bentonite is quoted at 41\% in [58], it is also stated that the bentonite will initially contain some water, so the porosity for groundwater flow may be lower. However, to specify inputs for criticality calculations it is the total maximum water content which is relevant.
molecular weight fractions sum to slightly less than unity, so these were normalised before calculating the elemental composition.

Table A.3: Composition details for dry clay (mudstone).

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Weight fraction</th>
<th>Element</th>
<th>Weight fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>0.581</td>
<td>Si</td>
<td>0.2766</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>0.154</td>
<td>Al</td>
<td>0.0830</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>0.040</td>
<td>Fe</td>
<td>0.0483</td>
</tr>
<tr>
<td>FeO</td>
<td>0.025</td>
<td>Mg</td>
<td>0.0147</td>
</tr>
<tr>
<td>MgO</td>
<td>0.024</td>
<td>Ca</td>
<td>0.0225</td>
</tr>
<tr>
<td>CaO</td>
<td>0.031</td>
<td>Na</td>
<td>0.0098</td>
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<tr>
<td>Na₂O</td>
<td>0.013</td>
<td>K</td>
<td>0.0270</td>
</tr>
<tr>
<td>K₂O</td>
<td>0.032</td>
<td>C</td>
<td>0.0133</td>
</tr>
<tr>
<td>CO₂</td>
<td>0.026</td>
<td>H</td>
<td>0.0057</td>
</tr>
<tr>
<td>C</td>
<td>0.006</td>
<td>O</td>
<td>0.4991</td>
</tr>
<tr>
<td>H₂O in rock</td>
<td>0.050</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Solid Density | 2720 kg m⁻³  
Porosity       | 12%           
Density of water in the pores | 995 kg m⁻³  

A.1.4 Evaporite

Criticality calculations were not undertaken for evaporite rock during the UCuRC programme, so this was an additional material for the research programme reported here. Table A.4 summarises suitable information for an evaporite rock.

In Table A.4 the composition and density in the left columns are taken from [61], which gives a specification in terms of minerals such as halite and anhydrite. The chemical form of these minerals was taken from [62]. Finally the porosity was taken from [63]. While [61] is not clear whether the density is for a material containing pore space, the low porosity means that there would be little difference, so 2,180 kg m⁻³ was taken to be the density of a pore-free solid. Using information on the molecular weights of the required elements, the right hand columns were calculated and provide an elemental composition suitable for use in MONK.
Table A.4: Composition details for dry evaporite.

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Weight fraction</th>
<th>Element</th>
<th>Weight fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Halite (NaCl)</td>
<td>0.932</td>
<td>C</td>
<td>0.0024</td>
</tr>
<tr>
<td>Anhydrite (CaSO₄)</td>
<td>0.017</td>
<td>Ca</td>
<td>0.0112</td>
</tr>
<tr>
<td>Gypsum (CaSO₄·2(H₂O))</td>
<td>0.017</td>
<td>Cl</td>
<td>0.5654</td>
</tr>
<tr>
<td>Magnesite (MgCO₃)</td>
<td>0.017</td>
<td>H</td>
<td>0.0005</td>
</tr>
<tr>
<td>Polyhalite (K₂Ca₂Mg(SO₄)₄·2(H₂O))</td>
<td>0.017</td>
<td>K</td>
<td>0.0022</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mg</td>
<td>0.0056</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Na</td>
<td>0.3666</td>
</tr>
<tr>
<td></td>
<td></td>
<td>O</td>
<td>0.0353</td>
</tr>
<tr>
<td></td>
<td></td>
<td>S</td>
<td>0.0108</td>
</tr>
</tbody>
</table>

| Solid Density                                       | 2180 kg m⁻³    |
| Porosity                                            | 1%              |
| Density of water in the pores                       | 995 kg m⁻³      |

A.2 Information for Transient Criticality Calculations

To undertake the scoping transient calculations identified in Section 5.3 suitable inputs are required for either the QSS or Bounding Approach models, dependent on which is required.

One of the key model inputs for either model is a reactivity function. The construction of these is based on a series of static criticality calculations from which interpolation functions can be constructed. Further information, with examples, is given in the user guides for the transient models [28,29]. The information for constructing these is generally similar to that for other static criticality analyses. For example, much of the required material composition data can be taken from Section A.1 above.

This section considers other information that is required for the specified scoping calculations (Section 5.3). For the accumulation scenario both Bounding and QSS calculations are specified, so material property requirements for both are considered. For the in-package scenario of flooding within a PWR spent fuel canister only QSS calculations were specified.

A.2.1 Accumulation Scenario

Calculations have previously been undertaken in NRVB, granite and clay (mudstone) for the accumulation scenario. Details of the material properties required for such calculations are described in [22,23], for example. The main focus for obtaining information for accumulation scenarios (for the specification of calculations in Section 5.3) was therefore determining suitable information for bentonite material properties[41]. The requirements for the different types of calculations are discussed below.

[41] Properties for grout were also required for a wider set of calculations for the accumulation scenario. Grout is used as the encapsulant for wastes in the ILW/LLW region of the illustrative disposal concepts [35].
A.2.1.1 Bounding Approach Calculations

The main advantage of the Bounding Approach over RTM for rapid transient criticality calculations is that the number of material properties required is greatly reduced. In particular the requirement to specify a structural response model (with associated parameters) and an equation of state is removed if the Bounding Approach is used. Other than a reactivity function (and possibly functions for the neutron generation time and delayed neutron fraction), the only other material property required is the specific heat capacity of the fissile material region as a function of temperature [29]. This can be derived from the enthalpy function for the fissile material region by taking the derivative with respect to temperature. Using the enthalpy function is recommended since latent heats for phase changes can be included; once suitably smeared over a temperature range these appear as effective specific heat capacities over those temperature ranges, and ensure that the energy required for phase changes is not overlooked.

Finding data in open literature sources to enable the derivation of enthalpy functions for materials such as naturally occurring rocks is not straightforward, especially since the information is typically required over temperature ranges of up to 100,000 K. However, during the UCuRC programme a method was developed which allows functions to be calculated. This method has been used to calculate enthalpy functions for use in Bounding Approach calculations in bentonite and grout and is described below.

An overview of the method is described in the steps below:

- Determine a simple representation of the solid rock, specified in terms of molecules that are as simple as possible (e.g. CaO, AlO, Al₂O₃, etc). Choose the key components of this specification (e.g. anything that has a weight fraction above 10%), and re-normalise the material specification.
- Taking the porosity of the material into account, and assuming the pores are saturated with water, calculate the weight fractions of the key solid components and water in the saturated mixture.
- For each of the identified components (including water) seek a means to determine enthalpy as a function of temperature:
  - Seek data from the NIST Chemistry Webbook [57], for example, where many ‘simple’ molecules have data in the form of ‘Shomate coefficients’ which enable enthalpy to be calculated as a function of temperature.
  - Using the NIST data, find Shomate coefficients for the largest temperature range possible. This is often between room temperature (298 K) and 6000 K. To find these data requires the consideration of different phases (solid, liquid and gas) of the molecule.
- Use the coefficients to calculate the enthalpy as a function of temperature for each phase. At this stage the units will be J kmole⁻¹.
- Visualise the enthalpy as a function of temperature and identify key temperatures that would enable it to be approximated as a piecewise linear function. This must include room temperature, temperatures either side of any phase changes (typically 100 K either side) and 6000 K as a minimum, and calculate the enthalpy at these temperatures.
- Set up a new simpler linear interpolation representation of the enthalpy as a function of temperature using the temperature-enthalpy pairs from the previous step. This function is required since:
  - it is simpler in form (piecewise linear) and easier for the Bounding Approach tool to use;
  - it includes smeared representations of phase changes as effective gradients in enthalpy from which the specific heat approximation can be calculated;
  - the function can easily be extrapolated beyond the upper temperature limit without the risk that a more complex functional form (e.g. quadratic and above terms) will give unphysical behaviour.
Use the molecular weight of the molecule to convert the enthalpy function to units of J kg\(^{-1}\).

Once this has been repeated for each of the required molecules, the functions can be added in the required weight fractions to determine an approximate enthalpy function for the original rock.

This method is not guaranteed to produce an enthalpy function, and includes a number of approximations as identified above. An additional assumption is that as the temperature of the material increases then the key components, including water, are chemically independent. This may not be true, but in the absence of the detailed chemical evolution of the system, it provides an approach that can be used and should capture the magnitude of the energy required to heat the material, if not the fine details.

To use the method in practice can require further assumptions. For example, for the molecule \(\text{Al}_2\text{O}_3\) (a component of bentonite), the NIST database has no available data for Shomate coefficients (or any other means to calculate enthalpy) above 4000 K. This either indicates that no data are available, or that the molecule does not exist above the upper temperature limit. The latter is quite possible if, at a phase change, the molecule dissociates into different molecules. If this is assumed, and enthalpy data are available for the assumed dissociated form, then it is possible to extend the temperature range further. This approximation was taken for \(\text{Al}_2\text{O}_3\) whereby, on the assumption that \(2\text{Al}_2\text{O}_3 \rightarrow 4\text{AlO} + \text{O}_2\), it is possible to extend the temperature range to 6000 K, since \(\text{AlO}\) and \(\text{O}_2\) have Shomate coefficients available for a wider temperature range\(^{42}\).

Using the above approximations it was possible to calculate approximate enthalpy functions (and hence specific heat functions) for bentonite as required for the specified Bounding Approach calculations. The assumption is that each kilogram of saturated material is 19.92% \(\text{H}_2\text{O}\), 60.92% \(\text{SiO}_2\) and 19.16% \(\text{Al}_2\text{O}_3\).

Figure A.1 summarises the calculated enthalpy function for bentonite. It also includes a comparison with other functions that have been constructed and used during the UCUcRC programme, and for grout at different porosities, reported in [35]. With the exception of the curve for clay (mudstone) which used a combination of Shomate data for \(\text{H}_2\text{O}\) and other data [60] for the solid, all of the functions were constructed using the methods outlined above. For Bounding Approach calculations the typical slope of the curves is the main interest, and this is seen to be similar for all of the materials.

Figure A.2 compares the calculated specific heat capacities as functions of temperature for the materials in Figure A.1, including the approximated specific heat capacities used to represent phase changes in component materials. Figure A.3 is similar, but concentrates on the underlying specific heat capacity, by restricting the vertical axis.

These summary figures confirm that the different materials have specific heat capacities that are similar in magnitude at typically a few thousand J kg\(^{-1}\) K\(^{-1}\), and with a general (but fairly weak) trend for lower specific heat capacity at higher temperature (although by design the upper value for specific heat will be independent of temperature from extrapolation of the enthalpy function).

The calculated functions for specific heat capacity were considered suitable for use in Bounding Approach calculations. It is noted that the functions, as given, have units of J kg\(^{-1}\) K\(^{-1}\). The user guide [29] specifies J kmole\(^{-1}\) K\(^{-1}\) for the user input ‘cp’ for the specific heat capacity. This is not an issue since either the functions can be converted using an averaged molecular weight for the material in the fissile material region, or the specific heat capacity function can be supplied in units of J kg\(^{-1}\) K\(^{-1}\) to the Bounding Approach model, provided that the amount of material in the fissile material region (the input ‘nmoles’ to the Bounding Approach model [29]) is given in units of kg instead of kmole. This is acceptable since the product of ‘nmoles’ and the specific heat ‘cp’ is the

\(^{42}\) This approximation for \(\text{Al}_2\text{O}_3\) (and similar for \(\text{Fe}_2\text{O}_3\)) were tested through simple sensitivity analysis for an example Bounding Approach calculation for accumulation in grout (a material considered for the disposal of ILW, LLW and DNLEU [35]) by varying the width and position of the assumed phase change. No significant changes in the results were observed.
actual quantity used in the system of equations solved by the model, and has units of J K$^{-1}$ as required.

**Figure A.1: Comparison of enthalpy as a function of temperature for different materials.**

![Figure A.1: Comparison of enthalpy as a function of temperature for different materials.](image1)

**Figure A.2: Comparison of specific heat capacity as a function of temperature for different materials.**

![Figure A.2: Comparison of specific heat capacity as a function of temperature for different materials.](image2)
Figure A.3: Comparison of specific heat capacity as a function of temperature for different materials (restricted vertical axis).

A.2.1.2 Quasi-steady-state Calculations

The QSS model requires a number of inputs for material properties [28]. These fall into one of a few categories:

- ‘nuclear data’ including averaged cross-sections and half-lives, together with which nuclides capture, fission or decay to others. The arrival rate of fissile material to the accumulation region is also part of this input;
- water/steam properties;
- material properties for the calculation of radial and tangential strain terms;
- material properties for the calculation of the heat transfer term.

Most nuclear data inputs are straightforward to specify, and are closely related to the initial conditions and the reactivity function for the calculation to be undertaken (See Appendix B).

For QSS accumulation scenarios the arrival rate of fissile materials to the fissile region is a key input which, together with the reactivity function and heat transfer, determines the temperature rise and evolution of the nuclide composition in the fissile region. It was by undertaking calculations over a wide range of arrival rates for given calculations that the ‘rule of thumb’ (see Section 6.2.2) was first established [22]. The same approach was taken for the additional calculations reported here, where for each QSS accumulation scenario a range of arrival rates were considered (in powers of 10) to produce QSS results where the average temperature rise in the fissile region varies from order 1°C to the upper limit allowed by the reactivity function without significant extrapolation.

Water/steam properties, originally from [57], were taken from previously undertaken QSS calculations [22]. For the calculation of strain terms, values are required for the thermal expansion coefficient and Poisson’s ratio. Since it has been shown that the tangential elastic strain can only be positive (and hence induce possible cracking) in the materials surrounding the fissile region of a spherical QSS calculation if Poisson’s ratio is greater than $3/7 \approx 0.43$ [28,12], and most rock/clay-like materials have ratio’s lower than this, the calculated strains are not of great interest for understanding the potential consequences of QSS criticality events. On this basis, sensible values of the thermal conductivity for concrete and estimates of Poisson’s ratio were supplied to the QSS model. However, none of the model results summarised in this report depend on those inputs.
Several of the inputs to the QSS model are used in constructing the heat transfer term, which determines how the heat produced from nuclear reactions can be used to heat the fissile material region and the surroundings. The heat transfer term has three components, which are:

1. The conduction of heat to the surrounding material, represented by a constant thermal conductivity for the surrounding material.
2. The transfer of heat to accumulating fissile material(s), if any.
3. The transfer of heat to groundwater flow passing through the fissile material region.

Sensitivity studies for QSS calculations in [15] showed that of all the parameters that can be supplied to the QSS model and are used in the heat transfer term, the results are usually only sensitive to the thermal conductivity. The groundwater flow rate could only affect the results if increased by a factor of 1000 from the best estimate value, which was based on flow for fractured higher strength rock geology. For other geologies it is expected that the groundwater flow rate would be lower, and hence the effect is likely to be smaller still.

A mathematical analysis of the components in the heat transfer formula confirms the results from the sensitivity calculations. For general material properties and dimensions for the QSS calculations it can be shown that the heat conduction component (the first item in the list above) is no smaller than a number of the order of 10. The second component in the list is of the order of $10^{-6}$ for even the largest arrival rates that could be considered, so can safely be neglected. The third component can be shown to be no larger than $10^7$ times the groundwater flow rate. Hence the groundwater flow rate needs to be at least $10^6$ m/s for this term to be comparable to thermal conduction. The best estimate for higher strength rock used in previous analysis was $10^{-9}$ m/s, so it is concluded that the third component can also be neglected. On this basis, the focus for the QSS heat transfer term was in identifying a suitable estimate for the thermal conductivity.

For the accumulation calculations specified, a value was required for the thermal conductivity of bentonite. A range of values are quoted in [58,66,77], with a value of 1.3 W/m/K being typical for saturated bentonite, although there is some variation dependent on samples, temperature and saturation. The sensitivity of QSS results to the assumed thermal conductivity can be inferred from [15], where it was generally seen that increasing the thermal conductivity by a factor of $x$, scales the temperature rise by a factor of $1/x$.

### A.2.2 In-package Scenario for PWR Fuel

For this scenario only QSS calculations were specified, since RT criticality events are not possible. While the scenario is different to the accumulation scenario, the input requirements for the QSS model are similar to those described in Section A.2.1.2 above. One requirement specific to the scenario of in-package flooding is the rate at which a package could flood. This rate could vary considerably, dependent on the cause of flooding. A sudden break to a package through geological activity might allow flooding to occur quickly, while pin-hole corrosion at isolated locations on a package would allow minimal flow of water into the package. Because the rate is not well known, it was decided to undertake a range of QSS calculations to understand the consequences of different flooding rates. This is analogous to the variation of arrival rate of fissile material in the accumulation scenario calculations.

The only other information required to enable QSS calculations to be undertaken were thermal conductivity properties. Two variants were considered for this; one using the thermal conductivity of bentonite at 1.3 W/m/K (see Section A.2.1.2 above), and another using the thermal conductivity of steel as 16 W/m/K [78].
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Appendix B
Reactivity Functions for the Transient Criticality Models

This appendix provides an overview of the requirements for reactivity functions in the QSS and Bounding Approach models, as required for the application of these transient criticality models to the calculations specified in Section 5.3. Example reactivity functions are also included with some discussion of what insight they provide (prior to use of the transient models). Reactivity functions are also required for use of RTM. Examples can be found in [22,23].

B.1 System Reactivity

Mathematically, a measure of how close a system is to being critical is defined as \( k_{\text{effective}} \), the ratio of the rate of neutron production (by fission) to the rate of neutron losses (by absorption plus leakage). At the point of criticality \( k_{\text{effective}} \) is equal to unity. For super–critical systems \( k_{\text{effective}} \) is greater than 1, and is less than 1 in sub-critical systems. A closely related mathematical quantity is reactivity, defined in units of mNiles to be:

\[
\rho = \frac{10^5 (k_{\text{effective}} - 1)}{k_{\text{effective}}}
\]

A reactivity function, as required by the transient criticality models, is a suitable function which describes how the system reactivity changes with appropriate physical quantities such as temperature and nuclide composition.

B.2 Overview of Reactivity Function Construction

Irrespective of whether an identified calculation requires application of the QSS model or the Bounding Approach a reactivity function is required for the model, and examples of such functions are well documented from the UCuRC programme [12,13,22,23,27] and the user guides [28,29].

In the case of a Bounding Approach calculation [27,29], the reactivity function can be constructed from the interpolation of a set of temperature and reactivity pairs. For a QSS calculation a more complicated function is required, typically requiring the dependence of system reactivity on temperature and the concentration (or mass) of a number of different fissile nuclides, decay, fission and capture products [12,22,23,28].

The sections below provide an overview of how a reactivity function can be specified. Some examples are included, although it is noted that many of the reactivity functions require the interpolation of approximately 1,000 data points in multi-dimensional space, so that including full details of all reactivity functions is not practical.

In constructing the reactivity functions for both the Bounding Approach and QSS calculations the MONK criticality software package has been used for all functions described in this Appendix. This choice was made since all static criticality calculations were undertaken in MONK on the basis that the ICASPA method (for MONK) was an efficient means of producing detailed criticality maps for a wide range of accumulation scenarios. It is preferable to use the same software for both static...
criticality calculations and the construction of reactivity functions so that the initial condition (from static analysis) and reactivity function for transient analysis are consistent.

The experience gained from the UCURC programme was used extensively in determining how to specify and define reactivity functions, particularly for the multi-dimensional functions required for the QSS model. Particular relevant experience for the construction of QSS reactivity functions is:

- How to distribute interpolation points in a given dimension. This includes how to estimate the range of values for a parameter, and how to divide that interval into a grid which can capture the main variations in system reactivity without requiring an impractical number of calculations [12,28].

- How the approximate burn-up representation used in the QSS model can be used to reduce the number of independent dimensions for a reactivity function in some instances [26,28].

- How to convert a reactivity function to temperature feedback coefficients for use in the QSS model [12,28], and how to decide whether a function is required for the constant volume or variable volume QSS model [28].

The sections below provide an overview of reactivity function construction for different scenarios for hypothetical criticality and the different transient criticality models.

B.2.1 Accumulation Scenario

B.2.1.1 Rapid Transient Criticality: Bounding Calculations

To construct a reactivity function for a Bounding Approach calculation, the following steps are required:

- Identify an initial configuration where the system is prompt-critical:
  - Prompt-critical means that the initial reactivity of the system must be greater than the delayed neutron fraction (for more details see [13,27,29]).
  - For MONK calculations, it is not possible to derive the delayed neutron fraction explicitly, so a target $k_{\text{eff}}$ of 1.003 (or reactivity of 299 mNiles) was used. This is larger than the typical delayed neutron fraction of 210 mNiles for a $^{239}$Pu system. Since the statistical uncertainty on calculating $k_{\text{eff}}$ using MONK is of the same magnitude as differences in the delayed neutron fraction for systems with different fissile materials the approximation of $k_{\text{eff}}$ of 1.003 for the initial condition was considered sensible.

- For that initial prompt-critical configuration verify that the temperature feedback coefficient for nuclear data effects is positive (i.e. increasing the temperature of the fissile region increases $k_{\text{eff}}$ and hence the reactivity).

- Assuming the temperature feedback coefficient is positive, repeat the MONK calculation, but using successively larger temperatures for the fissile material region, to obtain the dependence of reactivity on temperature. The largest temperatures need to be sufficiently large that the system becomes sub-critical.

- Use the temperature, reactivity pairs to form an interpolation function for use in the Bounding Approach model.

Using ICASPA with MONK initial conditions were determined for prompt-critical systems as specified in Table 5.1. The initial configurations are summarised in Table B.1, which also includes a calculation for clay (an example of a lower strength sedimentary rock), taken from [21]. It is noted that in Table B.1 there are no initial conditions for one of the calculations proposed in Table 5.1. This is because for the system with 10 kg of $^{239}$PuO$_2$-$^{235}$UO$_2$ in ratio 25:75 in bentonite, while

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43 For MONK, which is a Monte Carlo code, the grid must also be suitable so that the key trends are captured, without being dominated by statistical variation in the calculated reactivity values.
an initially prompt-critical configuration can be found, the temperature feedback coefficient is negative so that a Bounding Approach calculation was not possible.

Table B.1: Initial prompt-critical systems with positive temperature feedback coefficients.

<table>
<thead>
<tr>
<th>Host Material</th>
<th>Fissile Material(s)</th>
<th>Mass of Fissile Materials (kg)</th>
<th>Radius of Fissile Region (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bentonite</td>
<td>$^{239}$PuO$_2$</td>
<td>100</td>
<td>1.63</td>
</tr>
<tr>
<td>Bentonite</td>
<td>$^{239}$PuO$_2$</td>
<td>10</td>
<td>0.71</td>
</tr>
<tr>
<td>Bentonite</td>
<td>$^{239}$PuO$_2$-$^{235}$UO$_2$ in ratio 50:50</td>
<td>10</td>
<td>0.65</td>
</tr>
<tr>
<td>Clay (mudstone)</td>
<td>$^{239}$PuO$_2$</td>
<td>500</td>
<td>2.79</td>
</tr>
</tbody>
</table>

From these initial conditions, the dependency of $k_{\text{effective}}$ on temperature was calculated, using the very high temperature nuclear data libraries (Section 4.6.3). The results are shown in Table B.2. For each case a reactivity function can be constructed by interpolation of the temperature and $k_{\text{effective}}$ data, where the function must be converted to units of mNiles for use in the Bounding Approach model.

Table B.2: $k_{\text{effective}}$ as a function of temperature for accumulation systems with initially positive temperature feedback.

<table>
<thead>
<tr>
<th>Case</th>
<th>Temperature (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>293</td>
</tr>
<tr>
<td>100 kg $^{239}$PuO$_2$</td>
<td>1.0039</td>
</tr>
<tr>
<td>10.0 kg $^{239}$PuO$_2$</td>
<td>1.0031</td>
</tr>
<tr>
<td>10.0 kg $^{239}$PuO$_2$-$^{235}$UO$_2$ (50:50 mixture)</td>
<td>1.0026</td>
</tr>
</tbody>
</table>

Some general observations can be made from Table B.2, which provide some useful insight into how the energy release from a calculation may behave. The observations are that:

- All of the systems become sub-critical at sufficiently high temperatures (from Doppler broadening of the nuclear data cross-sections) so that Bounding Approach calculations should be possible.
- For a given material the systems with larger mass become more super-critical than those with lower mass (the peak value of $k_{\text{effective}}$ is larger).
- An increasing fraction of $^{235}$UO$_2$ in the fissile material generally acts to lower the values of $k_{\text{effective}}$.

Since lower values of $k_{\text{effective}}$ (and hence lower reactivity values) will tend to reduce the energy release, it was expected that lower mass, or increased fractions of $^{235}$UO$_2$ will release less energy from a hypothetical rapid transient criticality event. These are significant observations since if only small mass systems are considered credible, and the passage of time leads to increased fractions of $^{235}$UO$_2$ in any initially $^{239}$PuO$_2$ system, then only smaller energy releases will be considered credible.

In addition to a reactivity function the Bounding Approach model also requires values, or functions for the delayed neutron fraction and neutron generation time [29,27]. Using MONK as the criticality software it was not possible to calculate these, although sensible values are required. For the
delayed neutron fraction a value of 210 mNiles is usually used for $^{239}$Pu systems, and this value has been used for all calculations in Section 6.3 of this report. This is on the basis that the variation of this value with the fissile composition is within the statistical uncertainty on $k_{\text{effective}}$ values, and that Bounding Approach calculations are not very sensitive to changes in the value [27]. For the neutron generation time bounding Approach calculations are more sensitive. Historically a value of $10^{-4}$ s was used for all RTM and bounding calculations. However, for greater accuracy the values were calculated explicitly using WIMS (at the ambient temperature only) for each of the bentonite systems, so that realistic values could be used in each bounding Approach calculation. The values varied between $10^{-4}$ and $1.5 \times 10^{-4}$ s.

B.2.1.2 QSS Transient Criticality

B.2.1.2.1 Process for Reactivity Function Generation

Constructing a reactivity function for a QSS accumulation calculation is much more involved than for the bounding Approach. Generally the approach to constructing a reactivity function is as follows:

- Identify a system for analysis which is just-critical. Such initial conditions can be taken directly from criticality maps, such as those in Section 4.5.

- Verify that the temperature feedback coefficient (including both nuclear data and water density effects) is negative.

- Decide on an upper bound for the temperature for QSS calculations, typically several hundred degrees Celsius.

- Estimate how the composition of the fissile material region will vary during a transient criticality event from continued accumulation. Contributions from fission, capture, decay and the arrival of more fissile material need to be considered. A simple representation of nuclear data burn-up will be required for this purpose.

- For the key nuclides identified, estimate an upper and lower bound for the concentration of each nuclide.
  - For many nuclides the lower bound will be zero.
  - The upper bound can be estimated from the theoretical maximum concentration of all accumulating materials (based on the displacement of 100% of the water in the pores of the material in which accumulation occurs) together with some knowledge of how the burn-up model is used in the QSS model.

- Draw on any simplifications that can be made from the burn-up representation for the QSS model. For example, it might be possible to make an approximation that two nuclide concentrations are always in ratio, hence reducing the number of independent variables.

- For each nuclide required, and temperature, divide the required parameter range into a number of intervals.
  - For temperature this may be based on the availability of nuclear data and knowledge of how the system reactivity is likely to vary with temperature.
  - For nuclide concentrations a non-linear division of the parameter range is usually required, since at low concentrations of a nuclide, the addition of a little extra mass will typically have a larger effect on reactivity than the same amount added at a larger concentration.

- Calculate how many criticality calculations will be required if all combinations of all parameters are considered. For example 4 temperatures and 5 different concentrations for each of 5 different nuclides would require $4 \times 5^5 = 12,500$ calculations.

- Consider whether the full matrix of calculations is practical. Generally the aim is to restrict the number to less than 1,000 where possible. If it is not practical then consider whether further approximations are possible. These could include:
- Deciding to neglect reactivity dependence on one or more nuclide concentrations, if the maximum concentration is expected to be small
- Reducing the number of divisions of one or more parameters (nuclide concentrations or temperature) or revising the range.
- Not considering a full matrix of calculations, and instead constructing a series of perturbations about a ‘core’ reactivity function. For example, a core function dependent on temperature and two key nuclide concentrations may be constructed. For every other nuclide a perturbation function is calculated around this core, specific to that nuclide. For combinations of other nuclides the perturbations for each of the individual nuclides are added together to give a total perturbation to the core reactivity.

- Undertake the identified criticality calculations and construct the reactivity interpolation function. Confirm that all behaviours are sensible: adding fissile material should normally increase the reactivity while adding absorbing nuclides or increasing the temperature should reduce it.
- If the behaviours are not sensible investigate why, and refine the reactivity function construction if required.

B.2.1.2.2 Deciding on the Temperature Range

For all of the reactivity functions constructed for this study the temperature has been restricted to either three or four values, covering from ambient conditions (40 °C) to either 200 °C or 300 °C, which enables the number of MONK calculations required for any one reactivity function to be kept to a practicable level.

B.2.1.2.3 QSS Initial Conditions and Concentration Range

Using the criticality maps from Section 4.5, the initial conditions in Table B.3 match the calculations specified in Section 5.3. Table B.3 shows that the radius of the accumulation regions is typically 0.25 m or less, which is a factor of at least two lower than for the proposed Bounding Approach calculations for rapid transient criticality events (Table B.1).

Table B.3: Initial just-critical systems with negative temperature feedback coefficients for QSS accumulation calculations.

<table>
<thead>
<tr>
<th>Calculation Details</th>
<th>Accumulation radius (m)</th>
<th>Initial fissile concentration (kg/m³)</th>
<th>Initial fissile mass (kg)</th>
<th>Theoretical maximum fissile mass (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Minimum critical mass of ²³⁵UO₂ in bentonite</td>
<td>0.244</td>
<td>41.0</td>
<td>2.5</td>
<td>249.4</td>
</tr>
<tr>
<td>About 3 times minimum critical mass of ²³⁵UO₂ in bentonite</td>
<td>0.180</td>
<td>287</td>
<td>7.0</td>
<td>100.3</td>
</tr>
<tr>
<td>About 10 times minimum critical mass of ²³⁵UO₂ in bentonite</td>
<td>0.177</td>
<td>1025</td>
<td>23.9</td>
<td>95.7</td>
</tr>
<tr>
<td>Minimum critical mass of ²³⁹PuO₂ in bentonite</td>
<td>0.253</td>
<td>22.6</td>
<td>1.5</td>
<td>279.4</td>
</tr>
<tr>
<td>Minimum critical mass of ²³⁹PuO₂:²³⁵UO₂ in ratio 50:50 in bentonite</td>
<td>0.251</td>
<td>28.7</td>
<td>1.9</td>
<td>271.5</td>
</tr>
</tbody>
</table>

For the initial conditions based on minimum critical mass, that mass is between 1.5 kg and 2.5 kg, and the fissile concentration is between 22.6 kg/m³ and 41.0 kg/m³. For the larger initial masses...

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The assumption is that for each of the perturbation nuclides the change in reactivity from a change in concentration of one nuclide is independent of the concentration of the other perturbation nuclides. Such an approximation is usually acceptable for non-fissile nuclides.
the concentrations can be much larger, with the ten times minimum critical mass case having an initial concentration of 1025 kg/m$^3$. On the assumption that such a critical configuration could occur, higher initial concentrations would be more difficult, or take longer, to accumulate.

Table B.3 also includes the theoretical maximum fissile mass for the fissile region. This theoretical value is calculated on the basis that all of the pore water within the fissile region is displaced by fissile material of solid density 10,000 kg/m$^3$. So, for example, in bentonite with an assumed porosity of 41% the theoretical maximum concentration of fissile material is 4,100 kg/m$^3$, and hence for a given volume the theoretical maximum mass can be calculated. This value is useful, since for a given arrival rate in a QSS calculation (the rate at which fissile material is assumed to accumulate), the theoretical maximum duration of a transient calculation can be calculated.

The theoretical maximum mass and duration were used for all QSS slow accumulation calculations reported in Section 6.3. While consistent with the methodology of the UCuRC programme [22,23], it is noted that there are reasons why the calculation could end before this, including that the complete blockage of the total porosity is not credible, and that a lower fraction should be considered. By undertaking calculations to the theoretical limit, however, it is a simple exercise to truncate the results at earlier duration in any subsequent analysis, as required.

### B.2.1.2.4 Reactivity Functions for $^{235}$UO$_2$ Systems

Most (three of the five) QSS calculations for bentonite are for systems where the initial fissile accumulation and the material which continues to accumulate is $^{235}$UO$_2$. Under the general simplified burn-up model developed for QSS accumulation calculations during the UCuRC programme [12], and included here as Figure B.1 and Table B.4, the burn-up model is simplified further if the accumulating material is only $^{235}$UO$_2$. In this case, the only capture product in the model is $^{236}$UO$_2$ and the only fission product is $^{239}$Pu. Furthermore, since the cross-sections for capture and fission are constant, then any capture and fission products produced will be in ratio; that ratio being 98.3:528.6, or 14.4% and 85.6% for $^{236}$UO$_2$ and PbO$_2$ concentrations respectively.

**Figure B.1:** Reaction chains for nuclear capture, fission and decay. This simplified example is typical of the burn-up representation used in the QSS model.

For $^{241}$Pu, the decay route could be more realistically represented as $^{241}$Pu to $^{241}$Am to $^{237}$Np. This would introduce two more nuclides, and hence more complexity, to the burn-up model. If $^{241}$Pu is only expected in very small quantities (much smaller than $^{236}$U) then assuming the decay of $^{241}$Pu to $^{236}$U is considered a reasonable approximation.

For fission products the QSS model requires a material to represent the stable end-point for fission product decay chains. Lead (Pb) is used since, while very little of the fission products would actually be lead, it is reasonably inert in terms of neutron interactions (i.e. it does not have a significant effect on system reactivity) and hence is a pessimistic assumption compared with fission products that could act as neutron absorbers. The representation of fission products in the QSS model (including sensitivity studies), was considered in the UCuRC programme [15]. PbO$_2$ is given an artificial low atomic weight in the QSS burn-up model (final row of Table B.4) to ensure the QSS model conserves mass following fission.
Table B.4: Cross-sections (at 2200 m s⁻¹) and half-lives of simplified burn-up model.
Cross-section data from [79], and decay data from [80]. Note that 1 barn = 10⁻²⁴ cm² = 10⁻²⁸ m².

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Atomic weight (dioxide)</th>
<th>Fission</th>
<th>Capture</th>
<th>Decay</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>σσσσ (barns)</td>
<td>Product</td>
<td>σσσσ (barns)</td>
<td>Product</td>
</tr>
<tr>
<td>²³⁵U</td>
<td>267</td>
<td>582.6</td>
<td>Pb</td>
<td>98.3</td>
</tr>
<tr>
<td>²³⁶U</td>
<td>268</td>
<td>0</td>
<td>N/A</td>
<td>0</td>
</tr>
<tr>
<td>²³⁸U</td>
<td>270</td>
<td>0</td>
<td>N/A</td>
<td>2.68</td>
</tr>
<tr>
<td>²³⁹Pu</td>
<td>271</td>
<td>748.1</td>
<td>Pb</td>
<td>269.3</td>
</tr>
<tr>
<td>²⁴⁰Pu</td>
<td>272</td>
<td>0</td>
<td>N/A</td>
<td>289.5</td>
</tr>
<tr>
<td>²⁴¹Pu</td>
<td>273</td>
<td>1011.1</td>
<td>Pb</td>
<td>358.2</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>274</td>
<td>0</td>
<td>N/A</td>
<td>0</td>
</tr>
<tr>
<td>Pb</td>
<td>135</td>
<td>0</td>
<td>N/A</td>
<td>0</td>
</tr>
</tbody>
</table>

The approximation of a fixed ratio of ²³⁶UO₂ and PbO₂ reduces the number of independent parameters to three for ²³⁵UO₂ accumulation reactivity functions: temperature, ²³⁵UO₂ concentration and total concentration of ²³⁶UO₂ and PbO₂.

Table B.5 summarises the results from a set of such MONK calculations for a system which initially has a concentration of 41 kg/m³ of ²³⁵UO₂ in bentonite, corresponding to the initial condition from the first row in Table B.3. From these results a reactivity function can be constructed. It is noted that the numerical values in red are approximate, extrapolated values. Some combinations of the concentrations are not physically sensible (since the total concentration would be more than could be accommodated in the available pore space), but numerical values are required for the interpolation scheme, so sensible values are extrapolated from neighbouring values.

Table B.5: k_{effective} calculations for varying temperature and nuclide concentrations to enable construction of a reactivity function for a system initially at the minimum critical mass of ²³⁵UO₂ in bentonite.

<table>
<thead>
<tr>
<th>Concentration of ²³⁵UO₂ and PbO₂ combined</th>
<th>Temperature 40°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 kg/m³</td>
<td></td>
</tr>
<tr>
<td>100 kg/m³</td>
<td></td>
</tr>
<tr>
<td>200 kg/m³</td>
<td></td>
</tr>
<tr>
<td>500 kg/m³</td>
<td></td>
</tr>
<tr>
<td>1500 kg/m³</td>
<td></td>
</tr>
<tr>
<td>4000 kg/m³</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Concentration of ²³⁵UO₂</th>
<th>0.9994</th>
<th>0.9798</th>
<th>0.9627</th>
<th>0.9209</th>
<th>0.7917</th>
<th>0.5578</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 kg/m³</td>
<td>1.1487</td>
<td>1.1274</td>
<td>1.1088</td>
<td>1.0624</td>
<td>0.9189</td>
<td>0.6630</td>
</tr>
<tr>
<td>200 kg/m³</td>
<td>1.2028</td>
<td>1.1807</td>
<td>1.1622</td>
<td>1.1172</td>
<td>0.9729</td>
<td>0.7230</td>
</tr>
<tr>
<td>500 kg/m³</td>
<td>1.2149</td>
<td>1.1958</td>
<td>1.1789</td>
<td>1.1371</td>
<td>1.0066</td>
<td>0.7882</td>
</tr>
<tr>
<td>1500 kg/m³</td>
<td>1.2036</td>
<td>1.1916</td>
<td>1.1806</td>
<td>1.1504</td>
<td>1.0543</td>
<td>0.8359</td>
</tr>
<tr>
<td>4000 kg/m³</td>
<td>1.2413</td>
<td>1.2338</td>
<td>1.2228</td>
<td>1.1926</td>
<td>1.0965</td>
<td>0.8781</td>
</tr>
</tbody>
</table>
Concentration of $^{236}\text{UO}_2$ and PbO$_2$ combined $\downarrow$

Concentration of $^{235}\text{UO}_2$ $\downarrow$

<table>
<thead>
<tr>
<th>Concentration $\downarrow$</th>
<th>Temperature 100°C</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>0 kg/m$^3$</td>
<td>41 kg/m$^3$</td>
<td>100 kg/m$^3$</td>
<td>200 kg/m$^3$</td>
<td>500 kg/m$^3$</td>
<td>1500 kg/m$^3$</td>
<td>3000 kg/m$^3$</td>
</tr>
<tr>
<td>0.9783</td>
<td>1.1315</td>
<td>1.1864</td>
<td>1.1986</td>
<td>1.1929</td>
<td>1.2420</td>
<td>0.9579</td>
</tr>
<tr>
<td>0.9579</td>
<td>1.1095</td>
<td>1.1632</td>
<td>1.1797</td>
<td>1.1812</td>
<td>1.2331</td>
<td>0.9419</td>
</tr>
<tr>
<td>0.9419</td>
<td>1.0918</td>
<td>1.1454</td>
<td>1.1645</td>
<td>1.1696</td>
<td>1.2215</td>
<td>0.9010</td>
</tr>
<tr>
<td>0.9010</td>
<td>1.0438</td>
<td>1.0968</td>
<td>1.1219</td>
<td>1.1433</td>
<td>1.1952</td>
<td>0.7722</td>
</tr>
<tr>
<td>0.7722</td>
<td>0.9025</td>
<td>0.9573</td>
<td>0.9955</td>
<td>1.0485</td>
<td>1.1004</td>
<td>0.5456</td>
</tr>
<tr>
<td>0.5456</td>
<td>0.6534</td>
<td>0.7136</td>
<td>0.7830</td>
<td>0.8360</td>
<td>0.8879</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Concentration $\downarrow$</th>
<th>Temperature 200°C</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>0 kg/m$^3$</td>
<td>41 kg/m$^3$</td>
<td>100 kg/m$^3$</td>
<td>200 kg/m$^3$</td>
<td>500 kg/m$^3$</td>
<td>1500 kg/m$^3$</td>
<td>4000 kg/m$^3$</td>
</tr>
<tr>
<td>0.9323</td>
<td>1.0832</td>
<td>1.1384</td>
<td>1.1583</td>
<td>1.1671</td>
<td>1.2400</td>
<td>0.9128</td>
</tr>
<tr>
<td>0.9128</td>
<td>1.0620</td>
<td>1.1176</td>
<td>1.1395</td>
<td>1.1569</td>
<td>1.2328</td>
<td>0.8962</td>
</tr>
<tr>
<td>0.8962</td>
<td>1.0439</td>
<td>1.1003</td>
<td>1.1232</td>
<td>1.1462</td>
<td>1.2221</td>
<td>0.8547</td>
</tr>
<tr>
<td>0.8547</td>
<td>0.9983</td>
<td>1.0548</td>
<td>1.0841</td>
<td>1.1209</td>
<td>1.1968</td>
<td>0.7313</td>
</tr>
<tr>
<td>0.7313</td>
<td>0.8607</td>
<td>0.9182</td>
<td>0.9652</td>
<td>1.0347</td>
<td>1.1106</td>
<td>0.5228</td>
</tr>
<tr>
<td>0.5228</td>
<td>0.6314</td>
<td>0.6948</td>
<td>0.7722</td>
<td>0.8417</td>
<td>0.9176</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Concentration $\downarrow$</th>
<th>Temperature 300°C</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>0 kg/m$^3$</td>
<td>41 kg/m$^3$</td>
<td>100 kg/m$^3$</td>
<td>200 kg/m$^3$</td>
<td>500 kg/m$^3$</td>
<td>1500 kg/m$^3$</td>
<td>4000 kg/m$^3$</td>
</tr>
<tr>
<td>0.4196</td>
<td>0.5057</td>
<td>0.5672</td>
<td>0.6655</td>
<td>0.8725</td>
<td>1.2314</td>
<td>0.4152</td>
</tr>
<tr>
<td>0.4152</td>
<td>0.5008</td>
<td>0.5641</td>
<td>0.6656</td>
<td>0.8727</td>
<td>1.2330</td>
<td>0.4122</td>
</tr>
<tr>
<td>0.4122</td>
<td>0.4995</td>
<td>0.5627</td>
<td>0.6645</td>
<td>0.8746</td>
<td>1.2349</td>
<td>0.4074</td>
</tr>
<tr>
<td>0.4074</td>
<td>0.4958</td>
<td>0.5611</td>
<td>0.6659</td>
<td>0.8772</td>
<td>1.2375</td>
<td>0.3976</td>
</tr>
<tr>
<td>0.3976</td>
<td>0.4905</td>
<td>0.5595</td>
<td>0.6706</td>
<td>0.8928</td>
<td>1.2531</td>
<td>0.3864</td>
</tr>
<tr>
<td>0.3864</td>
<td>0.4834</td>
<td>0.5585</td>
<td>0.6784</td>
<td>0.9006</td>
<td>1.2609</td>
<td></td>
</tr>
</tbody>
</table>

Some general observations confirm that the behaviour of the values is sensible. For example:

- Increasing the concentration of $^{235}\text{UO}_2$ increases $k_{\text{effective}}$.
- Increasing the concentration of $^{236}\text{UO}_2$ and PbO$_2$ decreases $k_{\text{effective}}$.
- Increasing the temperature decreases $k_{\text{effective}}$.

Similar reactivity functions were constructed for the other $^{235}\text{UO}_2$ systems for which QSS calculations were required.

**B.2.1.2.5 Reactivity Functions for $^{239}\text{PuO}_2$ Systems**

The construction of reactivity functions for systems containing $^{239}\text{PuO}_2$ or a mixture of $^{239}\text{PuO}_2$ and $^{235}\text{UO}_2$ is much more complicated. Due to the radioactive decay of $^{238}\text{PuO}_2$ to $^{235}\text{UO}_2$ with a half-life of 24,100 years, the ratio of $^{239}\text{PuO}_2$ to $^{235}\text{UO}_2$ in the accumulating material will change as the transient criticality continues [28]. Since the arriving material will change (and any $^{239}\text{Pu}$ at the initial condition will decay) it is more involved to construct a reactivity function that will enable a range of arrival rates to be considered. Furthermore, with $^{239}\text{PuO}_2$ present, capture to $^{240}\text{PuO}_2$ and possibly higher plutonium isotopes must be considered. Finally, since the capture to fission cross-section ratios are different for $^{239}\text{Pu}$ and $^{235}\text{U}$ (Table B.4), it cannot be assumed that the ratio of capture and fission product concentrations is fixed. This greatly increases the number of...
independent parameters on which the reactivity function can depend, and has the potential to require a very large number of criticality calculations to construct a reactivity function.

Two such reactivity functions are required for the calculations specified in Table B.3. For each of these the ‘practical steps’ to reduce the number of calculations required to construct a reactivity function to a reasonable level, as described in Section B.2.1.2.1, need to be invoked. The method used for this study has been to:

- Construct a base reactivity function dependent on the base set of variables - temperature and the concentrations of $^{239}\text{PuO}_2$ and $^{235}\text{UO}_2$.
- For all calculations in the base set, repeat them with different concentrations of $^{236}\text{UO}_2$ added. Using these results, and the base reactivity function, construct a perturbation reactivity function which depends on temperature and the concentrations of $^{239}\text{PuO}_2$, $^{236}\text{UO}_2$ and $^{239}\text{PuO}_2$. For zero $^{236}\text{UO}_2$ the perturbation reactivity function must be zero.
- Do likewise for different concentrations of PbO$_2$ added, and then $^{240}\text{PuO}_2$ added to create two further perturbation reactivity functions.
- Construct the full reactivity function by summing the base reactivity function and the three perturbation reactivity functions.

This method was successfully employed to produce reactivity functions for the $^{239}\text{PuO}_2$ and mixed $^{239}\text{PuO}_2$ and $^{235}\text{UO}_2$ systems identified in Table B.3. Each of them demonstrates physically sensible behaviours as the temperature and nuclide concentration(s) are varied. For these reactivity functions the temperature was limited to three interpolation values of 40 °C, 100 °C and 200 °C, requiring 1,170 MONK calculations per reactivity function.

### B.2.2 In-package Scenario for PWR Fuel

To enable a QSS calculation to be undertaken for the in-package scenario of flooding within a PWR fuel package, the construction of a suitable reactivity function was a key requirement. To enable use of the variable volume QSS model [28,35(Appendix B)] a reactivity function dependent on height\(^47\), temperature and the masses of each nuclide considered important for the QSS burn-up model, was required. Each parameter on which the function depends must be independent, so that the feedback coefficients (which are calculated as partial derivatives) can be calculated accurately.

Constructing such a function presented some challenges. For example, at a given height, if the mass of $^{235}\text{UO}_2$ is reduced, then some other nuclide concentration must vary to occupy the lost volume. Furthermore, if for a given fuel composition some further $^{235}\text{UO}_2$ is added, then this can only be done by either removing some of another material, or increasing the height. This issue stems from the fact that the fuel elements are solids, whereby in the accumulation scenario for which the model was developed (and the variable volume ‘stack slumping’ scenario for ILW, LLW and DNLEU disposal to which the model was extended in [35]) changes in composition through either accumulation or slumping can be accommodated by changing the fraction of pore space occupied by water.

The solution was to define the reactivity function in a different way. Given the mass of all nuclides of interest (in the assumed burn-up representation for the QSS model – e.g. Figure B.1) that are present in the flooded region of the package, then the height of the flooded region can be established. Under the model assumptions for the package there are initially 668 kg of 5% enriched UO$_2$ per metre height of the four fuel elements. Furthermore, given the masses of each nuclide considered important, the fractional composition of the fuel can be determined. It is then possible to define a reactivity function for the spent fuel as a function of height, fractional composition (weight percentage) of the nuclides and temperature. To this function a mapping can be produced to provide a function suitable for use in the QSS model.

\(^{47}\)The PWR fuel package is closest in geometry to a cylinder than a sphere since the fuel element compartments in the canister are long compartments of square cross-section within a cylindrical package.
The initial condition for the just-critical configuration is given in Table B.6, where the fuel is 5\% enriched UO\(_2\).

**Table B.6: Initial just-critical system with a negative temperature feedback coefficient for a QSS calculation in a vertically oriented PWR fuel canister.**

<table>
<thead>
<tr>
<th>Calculation Details</th>
<th>Flooded height in all four fuel compartments (m)</th>
<th>Initial UO(_2) mass in the flooded region (all four fuel compartments) (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Critical volume of 5% enriched UO(_2) in a vertically oriented disposal canister</td>
<td>0.336</td>
<td>224.4</td>
</tr>
</tbody>
</table>

Starting from this initial condition a reactivity function was constructed using four heights, four temperatures, three weight percents for \(^{235}\text{UO}_2\), two weight percents for \(^{236}\text{UO}_2\), three weight percents for \(^{238}\text{UO}_2\), two weight percents for \(^{239}\text{PuO}_2\) and two more for \(^{240}\text{PuO}_2\). Any remaining weight percent was assigned to PbO\(_2\) as the representative fission product. If the total weight percent was greater than 100\% in any case the weight percents were normalised. The ranges for the weight percentages of each nuclide were based on an analysis of the evolution of PWR fuel burn-up, as investigated for the production of an axial burn-up profile (Section 4.5.2.2). While this was generated for a PWR fuel element under typical operational (reactor) conditions, it provided good insight into how the initially 5\% enriched UO\(_2\) evolves during burn-up.

This specification required \(4^2 \times 3^2 \times 2^3 = 1152\) MONK calculations for the full specification in Table B.7. The resulting reactivity function for use with the QSS model has no explicit dependence on the flooded height, since this is in-built implicitly to the mapping to the interpolated function via the conversion from masses to height and weight fractions. Again this displayed sensible behaviours as the temperature and masses of each nuclide are varied, so that the feedback coefficients have the expected signs (negative for increasing temperature or the mass of any non-fissile nuclide, and positive for increasing mass of any fissile nuclide).

**Table B.7: Specification of MONK calculations to construct a reactivity function for the PWR in-package flooding QSS calculation.**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Values for the Interpolation Matrix</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flooded Height (m)</td>
<td>0.33604, 0.75, 1.5, 4.5</td>
</tr>
<tr>
<td>Temperature (° C)</td>
<td>40, 100, 200, 300</td>
</tr>
<tr>
<td>(^{235}\text{UO}_2) weight percent</td>
<td>2, 3.5, 5</td>
</tr>
<tr>
<td>(^{236}\text{UO}_2) weight percent</td>
<td>0, 0.6</td>
</tr>
<tr>
<td>(^{238}\text{UO}_2) weight percent</td>
<td>92, 93.5, 95</td>
</tr>
<tr>
<td>(^{239}\text{PuO}_2) weight percent</td>
<td>0, 0.7</td>
</tr>
<tr>
<td>(^{240}\text{PuO}_2) weight percent</td>
<td>0, 0.2</td>
</tr>
</tbody>
</table>

The temperatures for the reactivity function included 100 °C, 200 °C and 300 °C. The interpolated reactivity function displayed the expected behaviours with temperature, flooded height and nuclide composition. The function consists entirely of sub-critical values (irrespective of composition) at 300 °C. This is expected, since at 300 °C the water will have vaporised, and the density will be low (30.7 kg/m\(^3\) steam density at an assumed pressure of 6.5 MPa [57]). From the calculated values at 300 °C it was expected that any transient criticality event from flooding of a PWR disposal package would therefore be 'limited' to a maximum temperature between 200 °C and 300 °C using the interpolated reactivity function.
Appendix C
Summary of Assumptions

To undertake the analysis presented in this report, including the summary of research from the UCuRC programme, it has been necessary to make a number of assumptions. While the assumptions are stated throughout the report and relevant references (such as the user guides for the transient criticality software models) provide more details, this Appendix provides an overview of the assumptions that have been made during the research programmes. A summary of these assumptions is an important part of the toolkit (including the models and their application) which could support further model development and application, should this be required.

C.1 General Assumptions

Criticality consequences analysis undertaken does not consider, in any detail, the likelihood of the hypothetical critical systems modelled. Any hypothetical critical configuration is expected to be unlikely, and some systems will be much less likely than others.

All of the research, calculations, and analysis undertaken are bounded by:

- The 2007 Radioactive Waste Inventory [40];
- The illustrative geological disposal concepts and waste packaging concepts of the 2010 DSSC [3].

Furthermore:

- For HEU, the same packaging assumptions are made as for plutonium;
- Where required, water properties are taken as those at a pressure of 6.5 MPa, considered to represent a GDF depth of 650 m. The ambient temperature is assumed to be 40 °C.

C.2 Assumptions for Static Criticality Analysis

Static criticality calculations for the accumulation and in-package ‘what-if’ scenarios are presented in this report. A summary of the modelling assumptions is given below.

C.2.1 Accumulation Scenario

For static criticality calculations for the accumulation scenario the following apply:

- Simple geometry models are assumed for all accumulation calculations. The accumulation model comprises two volumes: an inner volume where fissile material is present, and an outer volume where no fissile material is present. The outer volume is large enough to prevent any neutron leakage and hence represents an infinite expanse. New analysis for this report uses spheres. Some previous work used cylinders.
- It is assumed that the composition information used for materials, including density and porosity, are representative of the actual materials.
The system is assumed to be fully saturated with groundwater. It is also assumed that in the fissile material region, the fissile material displaces water from the pore space. The fissile material is assumed to be in oxide form with a solid density of 10,000 kg/m$^3$.

In most calculations all volumes are modelled as homogeneous mixtures of the materials (up to three materials are required for the homogeneous mixture – the host material, water and fissile material). For granite some heterogeneous calculations (representative of a simple fractured rock) have been undertaken, however [35].

No account is taken for other materials present in the waste packages. For example, potential neutron absorbing materials such as iron are neglected.

For criticality calculations it is assumed that nuclear data (cross-section data, etc.) at room temperature (20 °C) can be used when the material compositions (particularly the water density) have been defined at ambient conditions of 40 °C. This is because the JEF2.2 DICE nuclear data library used with MONK does not include data at 40 °C. When temperature feedback coefficients are calculated by analysing the change in reactivity from ambient conditions to 100 °C, it is assumed that the temperature change is 60 °C, even though it is actually 80 °C for the nuclear data.

**C.2.2 In-package Scenarios**

For static criticality calculations for the accumulation scenario the following apply:

- The MONK models supplied by Sellafield Ltd via INS (see Sections 4.5.2 and 4.5.3) have been used as the basis for all static criticality calculations for packages containing PWR fuel, plutonium and HEU. The modifications to the models included replacing the transport flasks with saturated bentonite in all models, and further changes to represent flooding (PWR fuel case) and removal of neutron absorbers and glass (plutonium and HEU case). While all changes to the models were verified, the basic model information including dimensions and material compositions were assumed to accurately represent the relevant wastes and packages.
- Where changes have been modelled within packages, they are implemented in a fairly ‘simple’ way in the models. For example:
  - Flooding of the PWR fuel package has each compartment containing an equal height of water in a vertically oriented package, and an equal volume in the lower two compartments for a horizontally oriented package.
  - Flooding of the PWR package is modelled without any corrosion to the PWR fuel elements or the package.
  - Removal of neutron absorbers has been implemented by replacing highly absorbing isotopes with the least absorptive isotope for that element.
  - Removal of glass from the plutonium and HEU packages has been implemented along the whole length of the package through use of small volumes occupied by water.
- For analysis of the burn-up profile of a PWR fuel element, the model details from the MONK model have been used to create a pincell model in WIMS. A small number of additional assumptions were required for typical reactor temperatures and temperatures based on modelling experience of WIMS experts.

**C.3 Assumptions for Transient Criticality Calculations**

The transient criticality models are described in detail in the user guides [28,29], which include the equations solved and the modelling assumptions. The following sub-sections provide an overview of the key assumptions. In all cases a key assumption is that the equations are representative of reality and do not neglect key physical processes. Validation and benchmarking have been undertaken where possible to build confidence in the models [25,26]. These have been documented during the model development.
C.3.1 The QSS Model

The QSS model is a software model [28] for hypothetical quasi-steady-state transient criticality events. It was used during the UCuRC programme and for additional calculations summarised in this report. For application of the model:

- The system is assumed to evolve sufficiently slowly that a quasi-steady-state is realistic – i.e. competing processes can maintain a just-critical system. For accumulation calculations the continued addition (arrival) of fissile material provides the reactivity insertion mechanism. For the in-package flooding calculations for a package containing PWR fuel, the flooding of the fissile material region provides the reactivity insertion.

- It is assumed that only the fissile material region heats up during the transient (for the model equations and inputs). Furthermore, the composition of the fissile material region can only evolve for nuclides defined in the supplied burn-up model.

- All volumes in the model are assumed isothermal – i.e. the fissile material region has a single temperature, and the surrounding material is at ambient temperature. A temperature profile for the fissile material region and surrounding material is calculated from the average temperature of the fissile material region using Equation 6.2, after application of the QSS model, although at present this is only applicable to spherical systems.

- It is assumed that the burn-up model (describing how neutrons can cause fission or be captured and how nuclides decay) includes sufficient detail to capture the behaviour of the system in terms of nuclide composition. This includes the assumption of a representative fission product (or products).

- It is assumed that the supplied reactivity function (a key input to the model) is representative of how the system reactivity will change with temperature, the concentrations of nuclides in the burn-up model, and (where required), the changing geometry. The in-package flooding calculations require a reactivity function which is dependent on the height of the fissile material region, which increases as the package floods. For accumulation calculations the reactivity function does not have any geometrical dependence since the fissile material region is assumed to have a fixed volume. The reactivity function is an interpolation function calculated from sets of static criticality calculations.

- For the in-package QSS calculations a homogeneous mixture is assumed for the fissile region, even though a burn-up profile could develop as the package floods. Furthermore, the equalisation of the package pressure to that of the saturated surrounding material is neglected, so that the in-package pressure is assumed to match that of the surrounding material (6.5 MPa) at all times during flooding.

- In addition to the burn-up model and reactivity function, other material properties are required for the QSS model such as heat conduction parameters, which includes the specific heat of the fissile material, groundwater flow rate, buoyancy terms and thermal conductivity. It is assumed the values supplied are suitable for the calculations being undertaken.

  - For the QSS model of a flooding PWR fuel package, (which in reality has four separate, but interacting fissile regions), assumptions are required for how to simulate this for heat transfer processes. Two different approximations have been used to understand how the QSS results are sensitive to the modelling of heat transfer. These are 1) a model where the heat transfer is dominated by conduction through bentonite, from the outer radius of the package, and 2) a model where heat transfer is dominated by conduction through steel from an effective radius for the four fuel compartments.

- QSS calculations have been undertaken for a range of arrival rates (accumulation scenario) or flooding rates (in-package flooding scenario), to identify trends. It is assumed that the rates used span or bound realistic rates, or that the identified trends could be used to estimate behaviour at different rates. In some QSS accumulation calculations the arrival of fissile material is assumed to continue after the pore water in the fissile region has turned to steam.
C.3.2 The RT Model

The RT Model (RTM) is a software model [29] for hypothetical rapid transient criticality events. It was used during the UCuRC programme but not for the latest calculations summarised in this report. For application of the model:

- It is assumed that only the fissile material region heats up during the transient (for the model equations and inputs). All volumes in the model are assumed isothermal – i.e. the fissile material region has a single temperature, and the surrounding material is at ambient temperature.

- It is assumed that the supplied reactivity function (a key input to the model) is representative of how the system reactivity will change with temperature and radius (from expansion due to high temperatures and pressures). The reactivity function is an interpolation function calculated from a set of static criticality calculations. Many of the assumptions from Section C.2 above also apply, such as simple geometry, material compositions and homogenous mixtures. Furthermore, it is assumed that nuclear data libraries for elevated temperatures (up to 100,000 K) capture the correct neutronic behaviour.

- In addition to the reactivity function, other modelling and material properties are required. For RTM, these include complex functional models, such as the structural response model and an equation of state. A specific heat capacity function is also required. It is assumed that these are consistent and defined over the required parameter ranges. The difficulty in defining structural response models/parameters and equations of state was the original motivation for the Bounding Approach model [25].

C.3.2 The Bounding Approach Model

The Bounding Approach model is a software model [29] for hypothetical rapid transient criticality events. It is essentially a simplified version of RTM, which does not require a structural response or an equation of state. The model was used during the UCuRC programme and for additional calculations summarised in this report. For application of the model:

- It is assumed that only the fissile material region heats up during the transient (for the model equations and inputs). All volumes in the model are assumed isothermal – i.e. the fissile material region has a single temperature, and the surrounding material is at ambient temperature. Furthermore, the volume of the fissile material region remains constant in the model, essentially constraining the energy release to a fixed volume.

- It is assumed that the supplied reactivity function (a key input to the model) is representative of how the system reactivity will change with temperature. This assumes that the nuclear data libraries for elevated temperatures (up to 100,000 K) capture the correct neutronic behaviour of high temperature systems. The reactivity function is an interpolation function calculated from a set of static criticality calculations. Many of the assumptions from Section C.2 above also apply, such as simple geometry, material compositions and homogenous mixtures.

- Other than the reactivity function, the only model input required is the specific heat of the host material as a function of temperature. To define this function over a large temperature range requires a number of approximations, which are described in detail in Appendix A, Section A.2.1.1.

- Once a Bounding Approach Calculation has been undertaken, the energy release (calculated by the model) can be used to estimate bounds on the cavity radius and cracked region. These require empirical formulae/relationships, including suitable empirical constants. The values used are assumed to be appropriate to the host materials. Otherwise, if a range of values are used, they are assumed to bound the behaviour (and hence consequences of the energy release). Where bounding values are taken in the empirical relationships this is essentially applying a further conservative estimate in addition to the conservatism of the Bounding Approach model itself.
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