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The consequences of hypothetical post-closure criticality

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ABSTRACT

The Environment Agency *Guidance on Requirements for Authorisation* (GRA) of a geological disposal facility (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." A neutron chain reaction that is just self-sustaining is also known as critical.

Waste packages can be designed to ensure that criticality is impossible during the transport and operational phases of a GDF, and for a significant period post-closure. Over longer times, however, packages may degrade, and groundwater flows could lead to a localized accumulation of fissile material. Hence, even though the initial distribution of materials would need to change substantially, criticality cannot be ruled out completely.

This paper describes how an accumulation of fissile material could, hypothetically, lead to a critical configuration; how such a system could evolve; what the local consequences could be; and how the engineered and geological barriers could be affected. The conclusion from studies to date is that, even for large (and very unlikely) fissile accumulations, the consequences of a post-closure criticality event are not a significant concern.

Keywords: disposal, post-closure, criticality.

Introduction

The Nuclear Decommissioning Authority (NDA) have 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 (Nuclear Decommissioning Authority, 2010*a*). At present a site for a GDF has not been identified, and RWMD has produced an initial 'generic' disposal system safety case (DSSC) (Nuclear Decommissioning Authority, 2010*b*), 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 guidance given in the Environment Agency and Northern Ireland Environment Agency (2009) *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.

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Waste packages can be designed to ensure that criticality is not possible during the transport and operational phases of a GDF, and for a significant period post-closure (Nuclear Decommissioning Authority, 2010*c*). Over longer times, however, packages may degrade, and groundwater flows coupled with geochemical processes such as dissolution and sorption could transport fissile nuclides, leading to a localized accumulation of fissile material so that criticality cannot be ruled out completely.

Approach to understanding 'what-if' post-closure criticality

In the 1998 assessment of the features, events and processes (FEPs) that might lead to a post-closure criticality for intermediate-level waste (ILW) in a higher strength host rock disposal facility, it was concluded that, although criticality would not be expected based on best-estimate data, the possibility of criticality events could not be entirely discounted (Nirex, 1998). This led to the initiation of the 'Understanding Criticality under Repository Conditions' (UCuRC) programme, to obtain a better understanding of the processes that would control the nature and magnitude of a criticality under the particular conditions arising post-closure in a GDF containing ILW. The UCuRC programme has involved the development and benchmarking of models that predict the evolution of 'what-if' critical systems, including the effects on the surroundings. Coupled with knowledge of the transport of radionuclides within and from the vicinity of the disposal facility, the models can be used to estimate the consequences of criticality. An up-to-date summary of the models, the associated verification, validation and benchmarking activities, and further references to detailed technical reports are given in Mason *et al*. (2009*c*).

The UCuRC programme to assess 'what-if' scenarios has followed a staged approach:

(1) How, hypothetically, could a critical system arise?

(2) How could a critical system evolve as a transient?

(3) What would the local consequences be?

(4) How would the engineered and geological barriers be affected?

This paper considers each of these questions in turn. The likelihood of scenarios under which a critical system could arise is the subject of further research. It is expected that some, if not all, scenarios will have a very low likelihood (Nuclear Decommissioning Authority, 2010*c*).

To understand the consequences of 'what-if' criticality events, this paper provides an overview of results from two mathematical models that have been developed in support of the NDA's research strategy (Nuclear Decommissioning Authority, 2009). Studies to build confidence in the models are also introduced.

Analysis: critical configurations

In a system containing fissile material, the fissile nuclides such as uranium-235 (^{235}U) and plutonium-239 (^{239}Pu) emit neutrons. The neutrons released may cause another fissile nuclide to split into two smaller nuclides, accompanied by the release of more neutrons, a process known as nuclear fission. The process of fission also releases energy as radiation and heat. Neutrons may be lost through absorption in non-fissile nuclides, or may leave the fissile part of the system to be absorbed in surrounding materials (leakage).

Under certain conditions, including a suitable combination of fissile mass, density, volume and shape; fissile concentration and enrichment (the weight fraction of the fissile nuclide ^{235}U in uranium); and the presence of neutron moderators (which slow down neutrons), absorbers (which absorb neutrons) and reflectors (which reduce the leakage of neutrons), a self-sustaining chain reaction of fission can be established.

At the point where the chain reaction becomes self-sustaining the system is said to be critical and there is a balance between the number of neutrons being produced by fission and the number being lost by absorption and leakage. If the number of neutrons produced by fission exceeds the number being lost the system is said to be super-critical. In a sub-critical system neutron losses exceed neutron production so that a chain reaction cannot be sustained.

Mathematically, a measure of how close a system is to being critical is defined as $k_{effective}$, 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 $k_{effective}$ is equal to unity. In super-critical systems $k_{effective}$ is greater than 1; it is less than 1 in sub-critical systems.

Given the number of conditions required for a critical configuration, the presence of fissile materials alone does not mean that such a configuration can occur. Indeed, for any wastes emplaced in a GDF, the initial distribution of fissile, or other, materials, would need to change significantly from the sub-critical emplacement configuration (Nuclear Decommissioning Authority, 2010*c*). Such changes will only be possible once sufficient time has passed that the engineered barriers are significantly degraded.

Under the assumption that fissile materials can relocate to a critical configuration, Fig. 1 shows the masses and concentrations that could lead to a critical system with $k_{effective} = 1$, under optimal configuration conditions within a regular cylindrical geometry, with equal diameter and height. Results are shown for a variety of fissile materials. These curves, reproduced from Cummings *et al.* (2007), are based on accumulation of fissile material in a backfill of water saturated Nirex reference vault backfill (NRVB) (Nirex, 1997), with a porosity of 50%. Any fissile materials displace water from the pore space.

The curves in Fig. 1 are based on the assumption that suitable combinations of fissile mass and accumulation volume (and hence fissile concentration) could arise, without consideration of what could cause such accumulations within a particular sub-volume of the total volume occupied by NRVB. Having determined the conditions needed for a criticality in terms of the mass and concentration of the fissile nuclides, the likelihood of obtaining such configurations by selective transport processes can be considered. Based on the overall package limits and the initial concentration of fissile nuclides, it has been concluded that the likelihood of obtaining a criticality is, in general, low, and even lower for either large fissile masses or concentrations (Nirex, 1998; Nuclear Decommissioning Authority, 2010*c*).

Analysis: criticality transients

Once a neutron chain reaction starts (i.e. the system is critical, corresponding to a point on one of the curves in Fig. 1), heat is released and so, locally, the temperature will increase. The affect of a change in temperature on the value of $k_{effective}$ determines the nature of the criticality transient as summarized in Fig. 2. There are two main 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 $k_{effective}$, but another process, such as the continued arrival of more fissile material can act to increase $k_{effective}$ (Smith *et al.*, 2007*a*). The

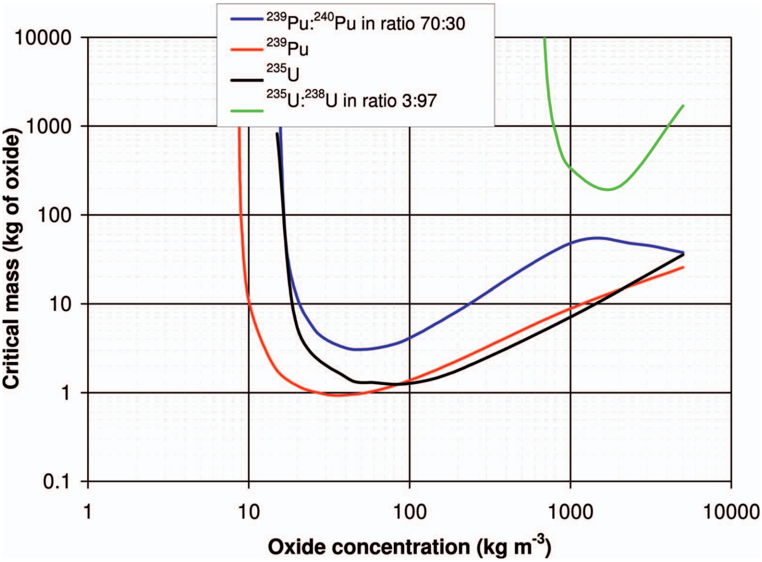

FIG. 1. Critical masses of plutonium and uranium for cylindrical fissile systems in NRVB; fissile material in oxide form.

Rise in temperature reduces $k_{effective}$

↓

System will become sub-critical ($k_{effective}<1$), unless some mechanism prevents this. Examples include the addition of more fissile material, or a reduction in neutron poisons.

↓

Termed a quasi-steady-state (QSS) transient criticality.

↓

Typically, long duration, low power events.

Rise in temperature increases $k_{effective}$

↓

System will become super-critical ($k_{effective}>1$), until a mechanism acts to reduce $k_{effective}$. Examples include reaching very high temperatures, or system expansion.

↓

Termed a rapid transient (RT) criticality.

↓

Typically, short duration, energetic events.

FIG. 2. Overview and characteristics of quasi-steady state and rapid transient criticality events.

other type is a short-lived, but energetic transient, and can result from a critical system with positive temperature feedback, where $k_{effective}$ increases with a rise in temperature (Smith *et al.*, 2007*b*). These are termed quasi-steady state (QSS) and rapid transient (RT) criticality, respectively, and could potentially have very different impacts on a GDF. Understanding both types of transient criticality is important in addressing the 'what-if' scenario of the GRA (Nuclear Decommissioning Authority, 2010*c*).

Most critical configurations of uranium can only develop as QSS transients, given a sustaining mechanism such as fissile accumulation. Plutonium systems can develop as RT or QSS transients. Figure 3 shows where each type of transient could, hypothetically, develop for an accumulation of $^{239}PuO_2$, with RT events at low concentrations or QSS events at higher concentrations.

Two bespoke transient criticality computer models have been developed to understand QSS and RT criticality events (Smith *et al.*, 2007*a*,*b*;

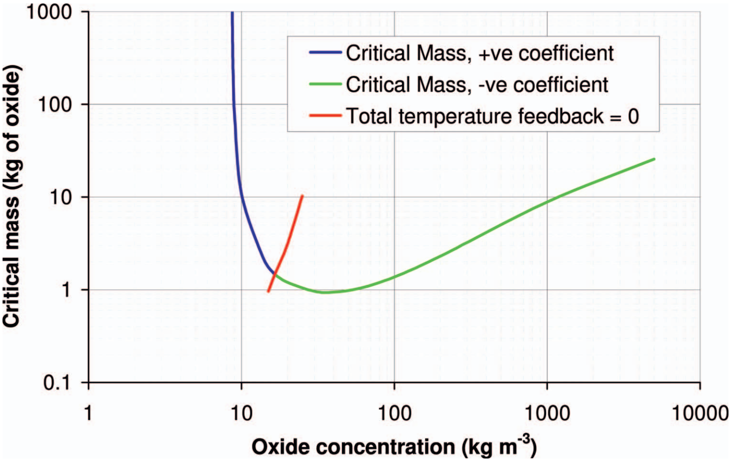

FIG. 3. Critical masses of plutonium oxide for cylindrical fissile systems in NRVB, showing the sign of the temperature feedback coefficient.

Mason *et al.*, 2009*c*). The computer models use calculations of $k_{effective}$ as a function of composition, size of the fissile region and temperature, obtained with a detailed criticality code such as *MONK* (Armishaw and Cooper, 2007). For a QSS event, the heat generated by the criticality can be removed principally by thermal conduction, so this process is modelled. The escalating $k_{effective}$ with temperature in an RT event means that it is not possible to offset the heat generation by thermal conduction (unless the temperature feedback coefficient becomes negative), so there is a need to model the expansion of the critical system. This requires models for the equation of state of the expanding fissile region and a structural response model for the surrounding medium (Smith *et al.*, 2007*b*). Typically an RT event is terminated by the formation of a cavity in the surrounding medium.

The following sections describe example calculations using the mathematical models to illustrate the consequences of hypothetical, 'what-if' scenarios.

Local consequences: quasi-steady-state model

The *Quasi-steady-state* (*QSS*) model (Smith *et al.* 2007*a*) has been developed to understand the evolution of QSS transient criticality events where, under suitable conditions, such as the continued accumulation of fissile material, the system can remain just critical for a significant period of time. The model solves a system of equations relating the system temperature, power, $k_{effective}$, and the concentrations of key nuclides within the area undergoing fission.

Figure 4 shows the results of example *QSS* model calculations for the continued accumulation of $^{235}UO_2$ in NRVB for a critical radius of 0.15 m; the effect of different accumulation rates is shown. The results for this and other calculations (e.g. Mason *et al.* 2009*a*,*b*) have demonstrated a 'rule of thumb' to scale the results for different arrival rates which is consistent with the understanding of the physical processes involved. The results in Fig. 4 are typical, in that for most accumulation rates, there is a local temperature rise of order 10ºC or less, rising to a few hundred degrees only for very large (and very unlikely) accumulation rates.

Sensitivity and uncertainty calculations show that the parameter having the largest impact on the results is the fissile accumulation rate (Mason *et al.*, 2007*a*), which depends on both groundwater flow rates, and sorption/solubility parameters. A review of the *QSS* model has shown it to be a powerful tool in understanding transient criticality (Mason *et al.* 2009*c*). In addition to a wide range of scoping and sensitivity calculations, the model has been successfully applied to data from the Oklo natural reactors (Mason *et al.*, 2011, 2012), where neutron poison burn-up is believed to have sustained natural nuclear reactors about 2 billion

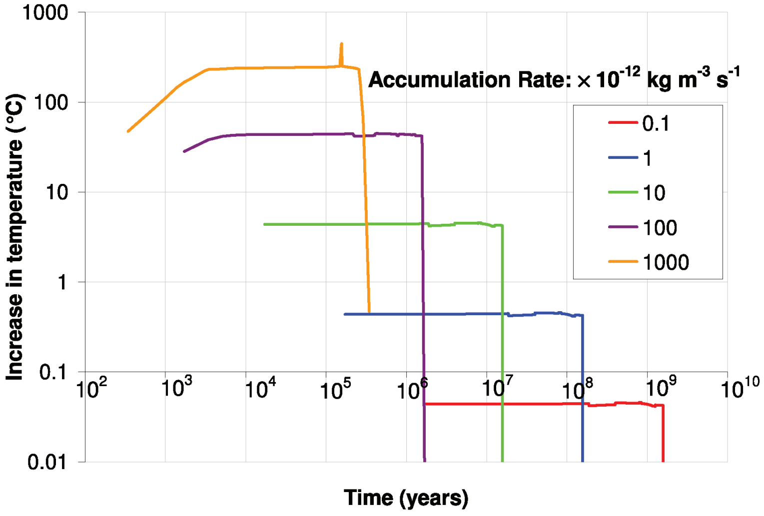

FIG. 4. Results of *QSS* calculations for the accumulation of $^{235}UO_2$ in NRVB for a critical radius of 0.15 m. The effect of different accumulation rates is shown.

years ago. Neutron poisons are good absorbers of neutrons, but in absorbing them, become less absorptive, and hence provide a mechanism for increasing $k_{effective}$ as an alternative to the continued arrival of fissile material.

Local consequences: rapid transient model

The *rapid transient model* (*RTM*) (Smith *et al*., 2007*b*) has been developed to understand the evolution of RT transient criticality events, where the systems can become super-critical, potentially releasing significant amounts of energy, before becoming sub-critical due to expansion or reaching a high temperature. The model solves a system of equations relating the system temperature, pressure, expansion, density, power and $k_{effective}$.

Figure 5 shows the results of an example *RTM* calculation, with an initial critical configuration of 100 kg of $^{239}PuO_2$ in NRVB. This mass is for illustration purposes, and does not indicate that such an accumulation is possible. The calculations were started assuming that a super-critical system with $k_{effective}$ slightly larger than unity had formed at time, $t = 0$. There is then a period of time (here about 2.9 s) where the temperature and $k_{effective}$ gradually increase, leading to escalating heat generation sufficient to cause expansion of the fissile region. Figure 5 shows that the system exhibits a rapid release of energy before shutting down through both high temperature and system expansion. Most of the expansion occurs after the energy release (power spike). Table 1 shows results for other masses of $^{239}PuO_2$, showing that even for the largest, and extremely unlikely, mass of 200 kg the maximum radius (cavity size) is about 30 m, which is small compared with the likely depth of the GDF. The size of the cavity can be used to assess the damage to the surrounding rocks, and thus the possible reduction in confinement of the radioactive inventory of the GDF. In general, it is concluded that only local damage will occur, without opening pathways to the environment.

A review of *RTM* has concluded that it is a powerful tool providing valuable insight into the consequences of hypothetical rapid transient criticality events (Mason *et al*., 2009*c*). It is, however, difficult to benchmark. Sensitivity and uncertainty analysis (Mason *et al*., 2007*b*), and some comparison with the Imperial College *FETCH* code (Smith *et al*., 2008), show that the dominant uncertainties arise from the equation of state, and the structural response model, which are required to solve the governing equations.

Recently, a simpler 'bounding approach' has also been developed to model RT criticality events. For scoping calculations this is simpler to apply than *RTM*, but (by design) will calculate a larger energy release. If the consequences of this bounding energy are not a concern, detailed *RTM* calculations may not be required. The bounding approach is therefore a very useful tool to scope the consequences of RT criticality events for a wider range of GDF geologies and hypothetical scenarios, with the *RTM* providing a means of more detailed analysis, if required.

Geological disposal facility consequence assessment

The results from the transient calculations, including the examples above, have demonstrated that, even for the largest (and most improbable) fissile masses, the local consequences of a post-closure criticality event are not a significant concern. In particular, the analysis undertaken concludes that even for the largest, most energetic rapid transient criticality events, there would not be sufficient energy release to disturb all of the geological barriers.

TABLE 1. Results of *RTM* calculations for different accumulations of $^{239}PuO_2$ in NRVB.

Fissile mass (kg)	3.684	10	100	200
Fissile concentration (kg m^{-3})	10.0	10.1	8.99	8.94
Initial radius (m)	0.4447	0.619	1.385	1.748
Total energy released ($\times 10^9$ J)	4.39	182	824	2160
Maximum power ($\times 10^{12}$ W)	1.54	9.77	680	1470
Maximum temperature (K)	2918	3029	16850	22030
Maximum pressure (MPa)	386	506	2970	4090
Maximum radius (m)	2.012	4.72	21.7	30.6

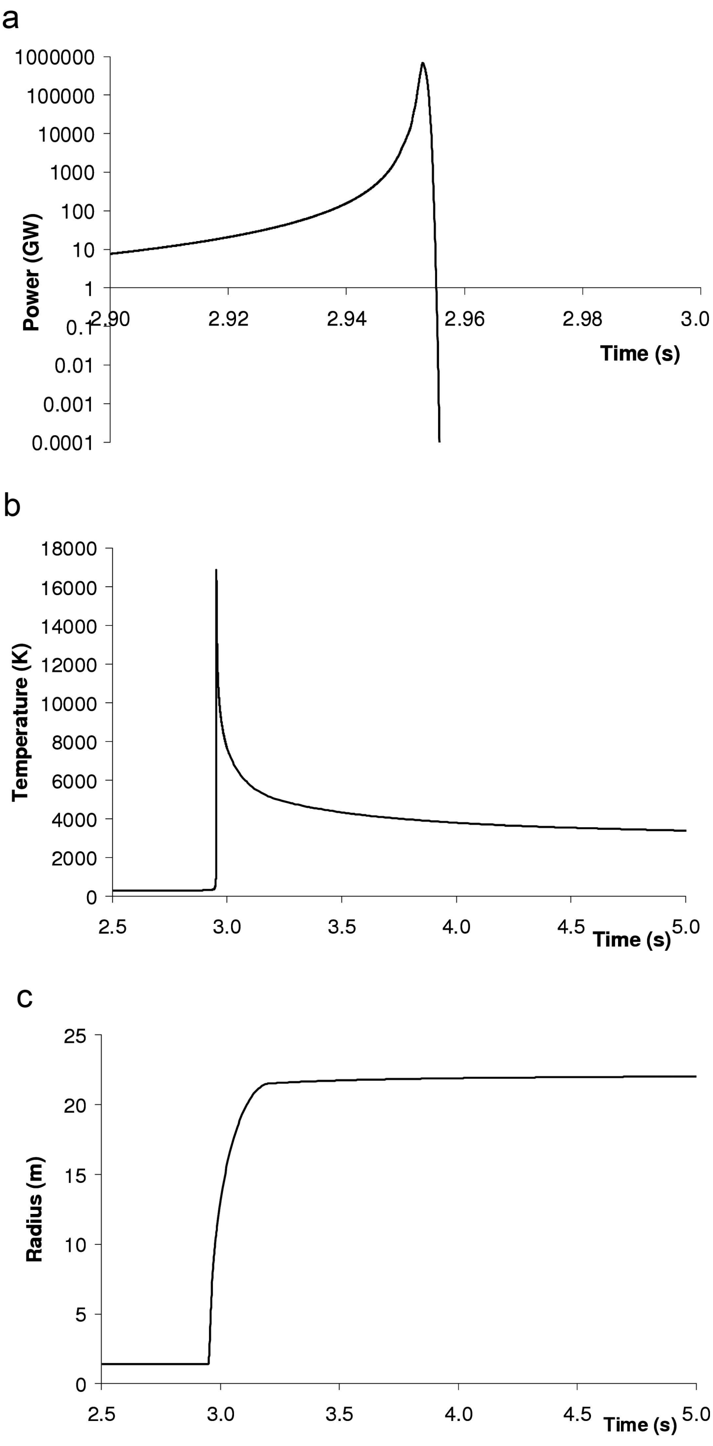

FIG. 5. Results of an *RTM* calculation for 100 kg of $^{239}PuO_2$ in NRVB including (*a*) power, (*b*) temperature and (*c*) radius of the fissile material region, as functions of time.

Furthermore, in 2008 a generic post-closure criticality consequence assessment (PCCCA) was undertaken by Cummings *et al*. (2008) using the results from the *QSS* and *RTM* models. The assessment included analysis of how large a criticality event (or multiple criticality events) would have to be to adversely affect GDF performance. The PCCCA showed that under the assumption of one or more criticality events (each itself unrealistically large) there would only be a slight increase in radiological risk via the groundwater pathway. Also, although the risks from GDF-derived radioactive gases may increase at long times, the peak risk, which arises at early times, would not change.

Conclusions

This paper provides an overview of the research undertaken to understand the potential consequences, of hypothetical, 'what-if', post-closure criticality scenarios, and how they support the DSSC for a GDF. By understanding how criticality could arise, and developing models to estimate the transient evolution of hypothetical critical systems, the research to date has concluded that the consequences of criticality are low for the following reasons:

(1) Even if they did occur, criticality events are likely to affect only a limited part of the GDF storage volume.

(2) Criticality events involving large amounts of fissile material might have a significant effect on a GDF and the near-field environment (defined to be the engineered barrier system and those parts of the host rock whose characteristics have been or could be altered by the GDF or its contents), but these events are very unlikely and could only occur a long time after closure.

(3) The backfill/buffer and geological environment will still act to isolate the radioactive waste from the surface environment.

In addition, direct radiation from the criticality event would be shielded by the surrounding rocks and materials and so is not a significant safety concern post-closure.

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