A MODEL OF RAPID TRANSIENT CRITICALITY UNDER REPOSITORY CONDITIONS

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Introduction

The UK Government announced in October 2006 that higher activity wastes will be managed in the long term through geological disposal. Government also announced that it was giving the Nuclear Decommissioning Authority (NDA) the responsibility for planning and implementing geological disposal and that United Kingdom Nirex Limited (Nirex) would be integrated into the NDA. The mission of Nirex has been, in support of Government policy, to develop and advise on safe, environmentally sound and publicly acceptable options for the long-term management of radioactive materials in the UK. These materials include intermediate-level and some low-level wastes (ILW and LLW) for which currently there is no disposal route. Nirex has developed a Phased Geological Repository Concept (PGRC) for ILW and LLW that makes use of a combination of engineered and natural barriers [1].

An important component of Nirex's research in support of the further development of this PGRC is the consideration of post-closure safety. Some of the wastes will contain fissile material (FM) and hence one aspect of safety that needs to be considered is criticality safety. While package integrity is maintained, the risk of criticality is eliminated by package design and control of package contents. In the post-closure phase, however, after loss of package integrity, there would be the possibility of movement of FM out of the waste packages and subsequent accumulation into new configurations that could in principle lead to a criticality. It is conceivable that a criticality could adversely affect the performance of a repository because of effects on the chemical and physical barriers to the release of radionuclides. It is therefore necessary to assess the post-closure criticality safety of the repository concept. Nirex has been undertaking work on post-closure criticality safety since the early 1990s [2]. Both the potential for a criticality and the consequences if one occurred have been examined.

Nirex is currently undertaking a programme of work with the objective of obtaining a better understanding of the nature of criticality under repository conditions. The aim is to obtain a better understanding of the processes that would control the nature and magnitude of a criticality under the particular conditions of a Nirex repository concept. The programme began in 2001. The main elements are a suite of static criticality calculations and the development of existing or new transient models of criticality under repository conditions. An overview of the programme is given in a separate ICNC 2007 paper [3]. The results of the work will feed into developing an improved methodology for assessing post-closure criticality safety.

This paper describes the development and testing of one of the new transient models, the rapid transient model (RTM) for rapid criticality transients resulting from slow accumulation of FM in systems with positive temperature feedback on reactivity.

The technical work described in this paper was undertaken in the period December 2001 to March 2006 by staff from Serco Assurance on behalf of Nirex.

1. Background

The main scenario for a criticality under repository conditions is the mobilisation of FM from a set of waste packages and its slow accumulation at some location in the repository or in its immediate vicinity in the host rock. For example, dissolved FM might precipitate at a change in chemical conditions. Provided sufficient FM accumulated, a criticality would result. Studies have shown the potential for a criticality to be low [2], but it has not been possible to quantify fully and hence discount the possibility. The reason for this is the uncertainties in the factors and processes that would control whether or not a criticality would occur on the relatively short length scales over which they would need to act. Hence the need to consider the consequences of a criticality, and for transient models of criticalities under repository conditions to estimate the magnitude of the effects of a criticality.

It has been shown that most critical systems under repository conditions would have negative temperature feedback [4]. However, systems containing significant amounts of 239Pu at low concentrations have been found to exhibit positive temperature feedback. In addition, a small range of low concentration 235U based systems has been found to have positive temperature coefficients at higher temperatures. For systems with positive feedback the temperature would rise rapidly until either the temperature feedback turns over and becomes negative, or the system expands to a subcritical configuration. In the former case, which covers the possibility of boiling resulting in a negative feedback coefficient, once the feedback coefficient became negative the system would approach a quasi-steady-state condition. In the latter case the expansion of the system results in termination of the criticality transient. Timescales would be rapid in such cases as they would be controlled by the neutron multiplication time. It is such systems that are considered here.

The approach taken was to design a relatively simple model – referred to as the 'RTM model' – that would be robust and be quick enough to undertake a wide range of calculations across the space of critical systems, to understand trends in the magnitudes of effects and test sensitivities [5]. The model has been compared with more detailed calculations performed by Imperial College, using their 3D, finite element coupled neutronic and thermal-hydraulic code, FETCH [6, 7].

2. Conceptual Model

This model is intended to provide a fast-running, approximate representation of the most energetic rapid transients. The physical model is based on the observed behaviour of underground explosions. In energetic underground explosions, material in the vicinity of the explosion is melted and vaporised, leading to the formation of a spherical cavity, centred on the explosion. The force

of the explosion results in crushing of a shell of material outside the cavity. Beyond the crushed zone is a further spherical shell in which cracking occurs, known as the cracked zone.

The model consists of a sphere containing a mixture of fissile material (FM), NRVB (Nirex Repository Vault Backfill) or rock and water, known as the Fissile Material (FM) region, surrounded by an infinite region of saturated NRVB or rock. The FM region is assumed to be isothermal and homogeneous and to contain a critical mass of FM with a positive temperature feedback coefficient on reactivity. The positive feedback results in a rapid increase in neutron flux, leading to heat generation in the FM region. The heat up and consequent pressurisation causes the FM region to expand, which tends to reduce the reactivity of the system. At higher temperatures the temperature feedback coefficient becomes negative and may also contribute to the termination of the criticality event.

The aim of the modelling is to identify the temperatures and pressures achieved during a criticality transient and the size of the resulting cavity. A simple experimental correlation can then be used to estimate the extent of cracking in the vicinity of the explosion. The neutron and gamma powers in the FM region are estimated, as they may effect the chemistry and hence retention of radionuclides in the neighbourhood of the criticality.

3. Mathematical Model and Solution

The neutronics of the criticality transient are represented by the point kinetics equation [8]:

$$\dot{P} = \frac{10^{-5}}{\Lambda}(\rho_{mN} - 210)P \qquad (1)$$

where ρmN *= reactivity in mNiles,*

P *= power,*

Λ *= neutron generation time.*

Due to the positive temperature feedback the system will rapidly become prompt critical and the neutron flux will increase faster than exponentially. Consequently delayed neutrons can be neglected in equation 1.

The power generated from the fissions goes into heating up the FM region, expanding the FM region, increasing the kinetic energy of the FM region and vaporising surrounding material, as represented by the four terms on the right-hand-side of the following equation, respectively [5]:

$$P = NC_p\dot{T} - N\alpha T v_m \dot{p} + \frac{3}{5}M\dot{R}(t)\ddot{R}(t) + \dot{N}\left[h(T,p) - h(T_b,p) + \left(f_b^{-1} - 1\right)L_h\right] \qquad (2)$$

where $N/T/p$ *= number of moles/temperature/pressure of the FM region,*

Cp/h *= specific heat capacity/enthalpy of the FM region material,*

α/Lh *= thermal expansion coefficient/latent heat of vaporisation of structural material,*

fb *= fraction of heat transferred to the surrounding material that goes into vaporisation.*

Once the temperature, T, of the FM region exceeds the boiling point of the surrounding material (SM), Tb, it will cause boiling of the SM. This will increase the number of moles of fluid, N, in the FM region according to [5]:

$$\dot{N} = \frac{4\pi f_b \sigma R^2 \alpha_s}{L_h}\left(T^4 - T_b^4\right) \qquad (3)$$

where σ *= Stephan – Boltzmann constant =* 5.67×10^{-8} *W m*^{2} *K*$^{-4}$*,*

R *= radius of FM region,*

Lh *= latent heat of vaporisation of the SM.*

The radius of the FM region increases due to the structural response of the surrounding material to the rising pressure and due to the vaporisation of material at the boundary of the FM region. This is represented by the two terms on the right of the structural response equation [5]:

$$\ddot{R} = \frac{1}{\sqrt{E\rho}}\dot{p} + \frac{v_{mSM}}{4\pi R^3}\left[R\ddot{N} - 2\dot{N}\dot{R}\right]. \qquad (4)$$

where $E/\rho/vmSM$ *= Young's modulus/density/molar volume of the SM.*

These equations are supplemented by the definition of the molar volume:

$$v_m = 4\pi R^3 / 3N \qquad (5)$$

and the equation of state for the material in the FM region:

$$\xi(p, T, v_m) = 0. \qquad (6)$$

Equations 1 to 6 form a complete set of equations for the six unknowns: N, R, T, P, p, vm. They are solved by using the ODE numerical solver package in Mathematica [9].

4. Reactivity Function

In order to solve equation 1 above it is necessary to specify the reactivity of the system as a function of temperature and radius of the FM region. This is done by pre-calculating the reactivity at a number of discrete temperature and radius pairs. The solver interpolates within the matrix of calculated values to estimate the reactivity at a particular temperature and radius. An example of a pre-calculated reactivity function, for an extreme case of 1 tonne of $^{239}PuO_2$ *accumulated within a sphere of saturated NRVB of radius 2.67 m is shown in Figure 1:*

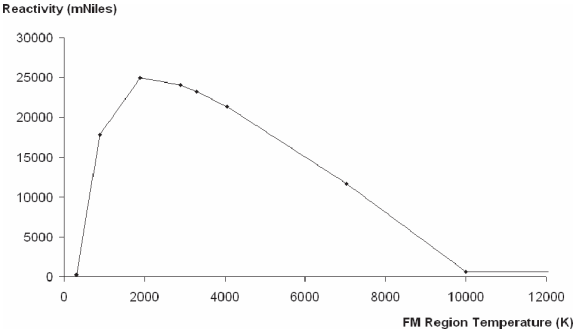

Figure 1. Variation of Reactivity with Temperature

In order to estimate the reactivities at elevated temperatures, a series of WIMS8 [10] nuclear data libraries were generated containing data at temperatures of 293K, 900K, 1,900K, 2,900K, 3,300K, 4,050K, 7,025 K, 10,000K, 100,000K and 1,000,000K for a number of nuclides (H, O, Na, Al, Si, K, Ca, Ti, Fe and ^{239}Pu*) [5]. The effect of the 239Pu resonance at about 3300 K on the reactivity can be seen in Figure 1.*

5. Model Output

The model produces the output required for assessment purposes, including the power history, total energy released, temperature and pressure histories, cavity radius, extent fracturing of the surrounding material, neutron and gamma powers and neutron flux. A methodology

has also been developed to estimate the resulting fission product inventory. A series of FISPIN [11] calculations are performed as the inventory of plutonium changes and the neutron flux evolves.

6. Verification and Validation

The RTM model has been extensively verified. The coding of the governing equations within Mathematica has been independently checked, confirming that the Mathematica implementation faithfully follows the documented model. The results from the RTM code have been compared with analytical results or alternative calculational methods and satisfactory agreement found.

It is not possible to validate directly full models of transient criticalities under repository conditions because of lack of experimental data. However, results have been compared with those from the more detailed FETCH code and satisfactory agreement obtained [7].

7. Example Calculations

A range of example calculations have been performed with the RTM model [5]. Selected results for $^{239}PuO_2$ in a saturated NRVB or granite sphere are shown in Table 1. Two examples of critical systems in NRVB are considered; in both cases the fissile material is present at a concentration of 10 kg m^{-3}. An example is also provided of a system with almost 1 tonne of fissile material in granite (where minimum critical masses are much higher than in NRVB) with a concentration of 3.62 kg m^{-3}.

Table 1: RTM Results for Spherical NRVB and Granite Systems with 239PuO2

	System 1	System 2	System 3
Host material	NRVB	NRVB	Granite
$^{239}PuO_2$ concentration (kg m^{-3})	10	10	3.62
Initial Radius (m)	0.4447	0.6842	4.025
$^{239}PuO_2$ mass (kg)	3.684	13.4	988.7
Initial reactivity (mNiles)	243	6899	227
Total Energy Released (kt TNT equivalent)	$1.7x10^{-4}$	0.0156	3.63
Maximum Power (W)	$1.43x10^{12}$	$6.57x10^{13}$	$3.58x10^{15}$
Maximum Temperature (K)	2890	9620	9990
Maximum Pressure (MPa)	380	1990	2190
Maximum Radius (m)	2.0	4.55	4.70

First consider systems 1 and 2 of fissile material accumulated in NRVB. System 1 contains just over the minimum critical mass of $^{239}PuO_2$ for a concentration of 10 kg m^{-3}. This can be seen from Table 1, which shows that the initial reactivity of the system is just above the prompt critical value of 210 mNiles. System 2 has a radius that is 54% greater than system 1 and therefore contains nearly 10 kg more Pu. This leads to a system with an initial reactivity of 6899 mNiles. Such a configuration could in theory be attained by accumulating the 13.4 kg of fissile material in a heterogeneous system. It is shown in [4] that, for the type of system considered here, heterogeneous Pu systems are less reactive than homogeneous systems. Therefore the required mass could be accumulated in a heterogeneous system, which homogenizes upon going critical, resulting in a large reactivity insertion. Such scenarios were hypothesized and studied by Kastenberg et al. [12].

The pre-evaluated reactivities for system 2 are displayed in Table 2, where the bold values indicate critical systems and the other values represent sub-critical system. The system is seen to become sub-critical if the temperature approaches or exceeds 10,000 K, or the radius approaches or exceeds roughly 1m. If either of these conditions is satisfied the criticality will quickly terminate and the power generation will fall to zero.

Table 2. Reactivity versus Temperature and Radius for System 2

Temp. (K) → / Radius (m) ↓	313.16	900	1900	2900	3300	4050	7025	10000
0.6842	6899	20114	24715	23708	22886	21011	11171	-753
0.71	6146	19436	24027	22955	22098	20142	9847	-2688
0.74	5169	18564	23135	21979	21075	19011	8120	-5215
0.8	2876	16516	21040	19682	18661	16339	4007	-11265
0.875	-679	13329	17781	16085	14873	12129	-2554	-20981
0.95	-5089	9371	13723	11572	10107	6802	-10978	-33550
1.2	-27035	-10472	-6704	-11603	-14568	-21160	-56932	-103389
1.7	-110105	-86124	-85603	-105864	-116895	-140894	-268490	-431734

The power history for the System 2 transient is shown in Figure 2. The power rise is seen to be very rapid and the shutdown is even quicker. The visible power transient lasts only a few milliseconds. From Table 1, the total energy released is seen to be equivalent to that released by 0.0156 kilotons (kt) of TNT.

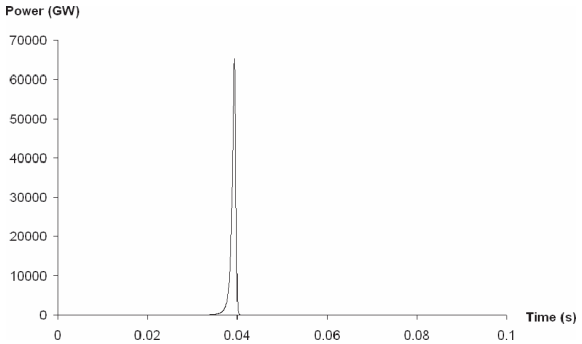

Figure 2. Variation of Power with Time for System 2

The resulting temperature history is depicted in Figure 3. The temperature turns over at 9620 K (see Table 1), below 10,000 K, as anticipated from the reactivities given in Table 2. Once the power transient is terminated the temperature of the FM regions falls back towards ambient, as a result of heat transfer to the surrounding material.

The pressure history is displayed in Figure 4. The peak value of over 1.2 GPa is reached very quickly. The pressure is then relieved by the expansion and cooling of the FM region.

The radius of the FM region as a function of time is shown in Figure 5. The expansion is seen to take several seconds, which is three orders of magnitude slower than the power excursion. This is due to the inertia of the surrounding material, which keeps it moving long after the power production has fallen to zero. Data from underground explosions in rock indicate that the size of the cavity produced is roughly proportional to the cube root of the yield for a wide range of conditions. The constant of proportionality is found to be of order 10 m kt -1/3 for the rock types studied [13]. From Table 1 the constant of proportionality is found to be 18 kt-1/3 for System 2, in line with the experimental data. From measured data on underground blasting it is found that cracking of the surrounding material extends out to roughly six times the cavity radius [14], i.e. about 27 m in this case.

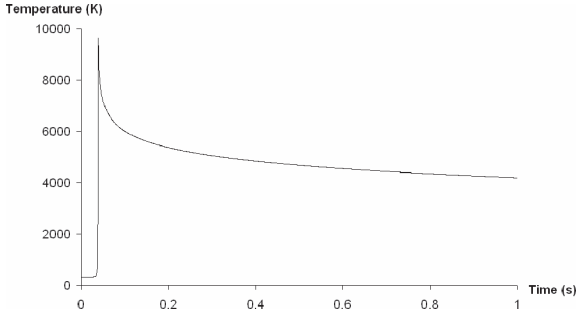

Figure 3. Cavity Temperature as a Function of Time for System 2

Once the temperature of the FM region has risen above the boiling point of the host material, the surrounding material is vaporised by heat transfer from the FM region. Though negligible vaporisation of the surrounding material occurs during the brief power excursion, the sustained high temperatures beyond the termination of the criticality allows time for vaporisation to occur in the longer term. It is predicted that over 1.5 kmoles of surrounding material is vaporised over the thirty seconds following the criticality transient.

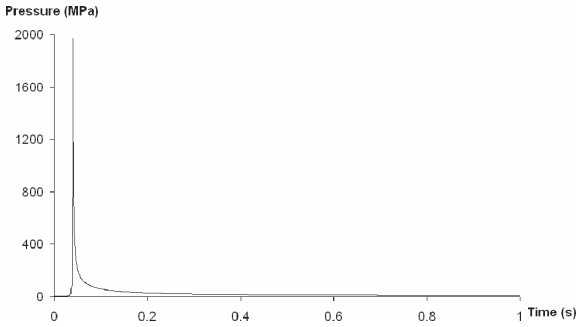

Figure 4. Cavity Pressure as a Function of Time for System 2

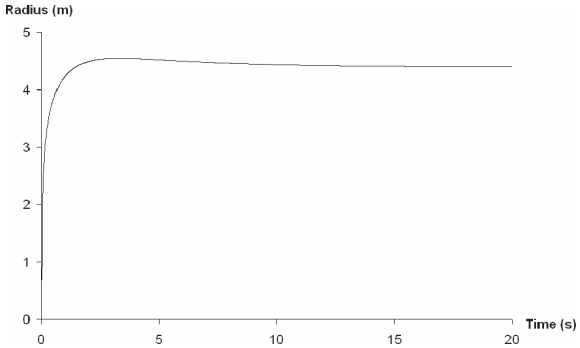

Figure 5. Variation of the Cavity radius with Time for System 2

From Table 1 it is seen that for System 1, the smaller mass of fissile material leads to a yield that is only 1% of that for system 2. This leads to a maximum temperature just under 2900 K. This is insufficient to vaporise all of the material in the FM region. Consequently, the pressure rise is less than 20% of that estimated for System 2. This leads to a cavity radius of only 2 m for System 1.

For System 3 there is almost 100 times the fissile mass of System 2. The resulting energy release is more than twenty times that obtained from System 2. This is partly due to the increase in the amount of fissile material and partly due to the increased confinement provided by the stronger host material. The maximum temperature is estimated to be just under 10,000 K, similar to System 2. This is a consequence of the system becoming subcritical at a temperature several thousand K and the reactivity being very negative (~ -20 Niles) by the time a temperature of 10,000 K is approached. Thus the transient is strongly quenched as a temperature of 10,000 K is approached. The increase in radius is very small in this system, due to the high Young's modulus of granite. In this case the radius of the cracked zone is estimated to be about 28 m. The peak pressure is determined by the peak temperature, the specific volume (i.e. the radius) and the equation of state. For System 3 the peak pressure is a little over 2 GPa, about 10% greater than that for System 2.

9. Peer Review

A peer review of Nirex's programme on understanding criticality under repository conditions was performed during Summer 2006 [15, 16]. The peer review was undertaken by independent experts in criticality and rock mechanics, from the US and UK. The extensive and careful nature of the work was noted by the reviewers. The difficulty in obtaining data for direct benchmarking was recognised, but further benchmarking against any existing

data was considered important. It was thought important to understand the significance of fracturing in the repository situation, where the arrangement of materials would be heterogeneous and materials might already be fractured. The need for further benchmarking and building confidence in the material response models is recognised by Nirex and is part of the ongoing programme.

10. Sensitivity Studies

A number of sensitivity calculations have been performed as part of the process of building confidence for the application of the RTM model to repository criticality safety assessment studies [17]. These calculations have built a comprehensive picture of which input parameters, functions and modelling assumptions can have a significant effect on the results of a calculation.

The results indicate that the estimated parameters are insensitive to the thermal properties of the system, including the enthalpy function, thermal expansion coefficient, absorbtivity, density, molecular weight, melting and boiling points and associated latent heats. The model was also found to be insensitive to number of evaluated reactivities used for interpolation above a minimum threshold (e.g. evaluation at four well-chosen radii was found to be adequate for the cases studied).

However, the results were found to be sensitive to the structural response modelling and associated material properties, such as Young's modulus of the host material. The structural response model is still undergoing development. The results were also found to be sensitive to the equation of state (EoS) used for the host material. Under the estimated conditions of temperature and pressure for the most energetic transients there is some uncertainty in the EoS of likely repository materials.

11. Concluding Comments

A description has been given of the development and testing of a model of transient criticalities under repository conditions with positive temperature feedback and resulting from the slow accumulation of FM. A more detailed description of the development and testing can be found in references [5, 6, 15, 16]. The model will be applied to estimating the magnitude of the effects of a criticality under repository conditions, in support of assessments of the post-closure criticality safety of the Nirex repository concept. In the example calculations for energetic rapid transient events in the NRVB inside a repository, cracking is estimated over lengthscales of up to a few tens of metres. For an extreme case in the rock, outside of the repository vaults, it is estimated that cracking could occur on a lengthscale also of order a few tens of metres. Cracking on such lengthscales would be unlikely to affect repository performance.

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