A MODEL OF QUASI-STEADY-STATE 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, for quasi-steady-state (QSS) criticalities resulting from slow accumulation of FM in systems with negative 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 potential. 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]. If FM continued to be added to a critical system with negative temperature feedback, the power and temperature would steadily rise to compensate for the addition of reactivity. Such systems are referred to here as QSS criticalities. Rates of accumulation of FM would be very slow under repository conditions. The timescales of a QSS criticality might be long enough that it is important to model radioactive decay of the Pu-239 and burn-up of the Pu 239 and U-235. No transient criticality model was available that could take into account decay and burn-up and it was decided to develop one for QSS transients resulting from the slow accumulation of FM.

The approach taken was to design a relatively simple model – referred to as the 'QSS 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. The model [5] is a development of a quasi-steady-state, analytic model of repository criticality developed in the US by Morris [6].

2. Conceptual Model

The conceptual model consists of a sphere containing fissile and other isotopes of plutonium and uranium and their fission and decay products, NRVB (Nirex Reference Vault Backfill, the reasons for the choice of backfill are discussed in [3]) and water, surrounded by an infinite expanse of NRVB and water. It is assumed that plutonium and/or uranium is being deposited in the NRVB sphere, wholly or partially saturated with water. The FM region is assumed homogeneous and isothermal. The size of this region remains the same, and the plutonium and uranium is deposited uniformly throughout the region, until the

defined supply of material is exhausted. This region is surrounded by an effectively infinite region of NRVB in which any plutonium and/or uranium content is negligible. A specified flux of groundwater is assumed to flow through the NRVB. During the criticality, it is assumed that all conditions remain constant apart from the rise in temperature, due to the power generated by fissions, and the composition of the FM region, due to the arrival, decay and burn up of the plutonium and uranium.

The transient is assumed to progress at a sufficiently slow rate to allow the negative temperature feedback to maintain the reactivity close to zero. Hence, the approximation is made that the reactivity is zero throughout the transient. The calculations begin when the system has just become critical. As further fissile material accumulates, the reactivity increases. This raises the power, which raises the temperature, which in turn decreases the reactivity to compensate for the rise due to the arrival 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. Fissile material would also be lost because of burn-up and radioactive decay and these processes are included in the model. Timescales could be sufficiently long that the effect of radioactive decay would be greater than that of burning. The system tends to a long-term steady state in which the fissile material arrival rate is balanced by the combined rates of burn-up and decay. The system remains in this steady state until the supply of new fissile material is interrupted. In the model, the arrival rate is reduced to zero when the first of two conditions is met: a defined amount of fissile material has been deposited in the FM region; or the pores in the NRVB have become completely filled with fissile material.

3. Mathematical Model and Solution

The model equations are:

maintain zero reactivity -

$$\sum_{i=1}^{N} \frac{\partial \rho}{\partial n_i} \dot{n}_i + \frac{\partial \rho}{\partial T} \dot{T} = 0$$

species conservation for the ith nuclide -

$$\left(E_f N_A \sum_{j \in FM} \sigma_{fj} n_j \right) \left(\dot{n}_i - \frac{d_{fi}}{\omega_i} + \lambda_i n_i - \sum_{j \in \check{I}} \lambda_j n_j \right) = \bar{H} \Delta T \left(\sum_{j \in \tilde{I}} \sigma_{cj} n_j - \sigma_{ci} n_i + \sum_{j \in \bar{I}} \chi_{ij} \sigma_{fj} n_j - \sigma_{fi} n_i \right)$$

where:

N	number of materials	ρ	reactivity
n_i	concentration of the i^{th} material	σ_{fj}	fission cross-section of the j^{th} nuclide
ω_i	atomic weight of the i^{th} nuclide	σ_{cj}	capture cross-section of the j^{th} nuclide
N_A	Avogadro's number	d_{fi}	source of the i^{th} nuclide
T	temperature	λ_i	decay coefficient of the i th nuclide
ΔT	temperature rise of the FM region above ambient	$\check{I}$	set of nuclides that decay to nuclide i
E_f	energy released per fission	$\tilde{I}$	set of nuclides that produce nuclide i upon capture
$\bar{H}$	heat transfer per unit volume	$\bar{I}$	set of nuclides that produce nuclide i upon fission

The dot indicates a time derivative.

The derivation of these equations is described in [5]. The equations form a set of N+1 ODEs in N+1 unknowns (n1,.....,nN, ΔT). Mathematica integrated technical computing software [7] is used to integrate the ODEs numerically, using user-supplied reactivity feedback coefficients.

The following materials are included in the model: U-235, U-236, U-238, Pu-239, Pu-240, Pu 241, Pu-242 and Pb. A simplified decay, transmutation and fission scheme is assumed. Pb is used to represent the end-point of actinide and fission product decay. It was found that Pb has little effect on reactivity and the reactivity dependence of Pb is now neglected. A pre-processor has been implemented that allows for radioactive decay of the shorter-lived nuclides (e.g. Pu-239) in the source of materials before they accumulate in the FM region.

Heat transfer allows for the effects of buoyancy as well as from background groundwater flow. Although there will be a temperature profile across the FM region, only an average temperature is calculated in the model. Therefore, boiling must be spread out over a range of average temperatures, usually of 20 degrees. This is achieved by incorporating the latent heat of the phase change into heat capacity, over the appropriate temperature range.

4. Reactivity Function

One objective was to provide a model quick enough to undertake a wide range of calculations. Solving the model equations requires reactivity feedback coefficients for changes in temperature and material composition. To avoid the need to calculate new feedback coefficients for each new calculation, a reactivity function has been developed to provide coefficients over a large fraction of parameter space. Reactivity coefficients are calculated by interpolation within a matrix of pre-calculated values. The reactivity function currently covers systems in saturated NRVB.

The pre-calculated values are too widely spaced to give sufficient accuracy using linear interpolation. A physical interpolation scheme has been developed, using simple reactor physics methods, to overcome this problem. If the physical interpolation scheme is used directly, run times are very slow. Instead, linear interpolation is used on a pre-calculated set of points in the local region of parameter space. The process of defining the local region of parameter space has been automated.

5. Model Output

The model produces the output required for assessment purposes, including masses and concentrations of materials arrived in the FM region, masses and con-

centrations of fissile materials in the FM region (taking into account decay and fission), average temperature in the FM region and power. The model also calculates the neutron and gamma powers and neutron flux in the FM region, and the temperature profile in the material surrounding the criticality and the resulting strain in the material.

A methodology has also been developed to estimate the resulting fission product inventory. A series of FISPIN [8] calculations are performed as the inventory of plutonium and uranium changes and the neutron flux evolves.

6. Verification and Validation

The QSS 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 source decay pre-processor, main solver, and power and structural response post-processors have been compared with analytical results 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. One opportunity for building confidence in the model that is being considered is comparing results produced by the model with understanding gained in studies of the Oklo reactors [9]. This is not straightforward because of uncertainties in the conditions that initiated and processes that sustained the reactions two billion years ago.

7. Example Calculations

A range of example calculations have been performed with the QSS model [5]. Selected results for $U^{235}O_2$ accumulating at 10^{-10} kgm^{-3}s^{-1} in a saturated NRVB sphere of radius 0.15 m are shown in Table 1. The minimum critical mass in this accumulation volume is close to the absolute minimum critical mass in NRVB.

Table 1.

Time (y)	Conc. Arrived (kgm^{-3})	Fissile Mass Arrived (kg)	Fissile Mass Left (kg)	Temp. (°C)	Power (W)	Neutron Flux (m^{-2}s^{-1})
0	59.7	0.844	0.844	40.0	5.90×10^{-23}	1.66×10^{-11}
0.0317	59.7	0.844	0.844	40.0	1.57×10^{-3}	4.40×10^{8}
0.317	59.7	0.844	0.844	40.0	0.0157	4.40×10^{9}
3.17	59.7	0.845	0.845	40.1	0.156	4.40×10^{10}
31.7	59.8	0.846	0.846	40.8	1.55	4.36×10^{11}
317	60.7	0.859	0.857	47.6	14.3	3.96×10^{12}
3,170	69.7	0.986	0.912	77.3	70.3	1.83×10^{13}
31,700	160	2.26	0.998	83.7	82.3	1.96×10^{13}
317,000	1060	15.0	1.82	82.1	79.5	1.03×10^{13}
1,570,000	5000	70.7	5.33	82.0	79.3	3.53×10^{12}
1,760,000	5000	70.7	4.79	40.0	7.54×10^{-5}	3.74×10^{6}

The times shown in Table 1 are from first criticality. An additional 19,000 years would be required to accumulate the initial critical mass. The FM stops accumulating after 1,570,000 years when the pore space is completely full (no account is taken of the likelihood of FM accumulating in this way over such a long period, it is simply assumed in the model that some accumulation mechanism can act until the pore space is full). The criticality takes about 190,000 years to run down and stop 1,760,000 years after first criticality. It can be seen that most of the U-235 fissions as it arrives. The average temperature in the FM region takes a few thousand years to approach the maximum value about 40°C above ambient temperature. The maximum power is only about 80 W and neutron flux about 2×10^{13} m^{-2}s^{-1}. Temperature as a function of radius when the temperature in the FM region peaks is shown in Figure 1. In this case, calculated temperatures remain within the design specification and are only increased by more than a few degrees in a very small volume in the repository.

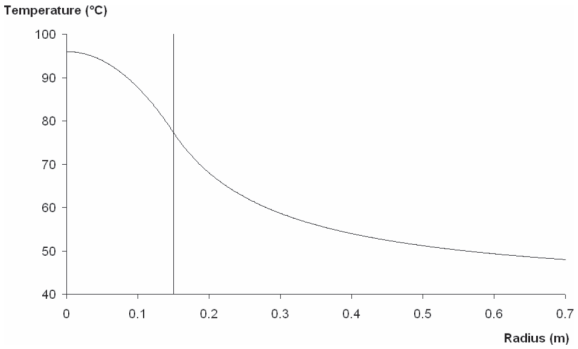

Figure 1. Temperature as a function of radius for accumulation rate of 10^{-10} kgm^{-3}s^{-1}. The vertical line shows the radius of the FM region.

Calculations have also been undertaken for accumulation rates ten times slower and ten times faster than 10^{-10} kgm^{-3}s^{-1}. With the slower accumulation rate, the timescales are all ten times longer, with the exception of the run-down time, which is 100,000 years, and the

peak temperature, power and neutron flux are ten times less. With the faster accumulation rate, the accumulation ceases ten times sooner after 157,000 years and in this case the run-down time is 150,000 years. Temperature increases more quickly, reaching the boiling range of 271 to 291°C after about 10,000 years (ambient pressure is assumed to be 6.5 MPa). The temperature evolution is shown in Figure 2. Once the boiling temperatures have been reached, the temperature rises very slowly, by about 20°C in 140,000 years, before rising rapidly to a peak temperature of 490°C when fresh fissile material stops arriving. (It should be noted that the model assumes that accumulation continues uniformly in the FM region without disruption even if boiling occurs, which may not be realistic.) The temperature then decays rapidly to the boiling range, decreases slowly through the boiling range over approximately 100,000 years, and finally decays to the ambient temperature over approximately 50,000 years. This behaviour can be understood from the variation of reactivity with temperature, shown in Figure 3 for the initial system composition. The slope of the reactivity function (the temperature feedback coefficient) determines by how much the temperature must rise to balance the additional reactivity of arriving fissile material. Since material arrives at a constant rate, the temperature rise will have to be much faster when the slope is gentle than when it is steep. The slope is steep in the boiling range, but much less so outside. It is coincidental that a large temperature rise to the final peak occurs just before fissile material stops arriving.

It is clear from the results that the accumulation rate would have a significant influence on the temperatures reached in a QSS criticality. The accumulation rates assumed above are high for a repository. For example, the accumulation rate of 10^{-10} kgm^{-3}s^{-1} implies a concentration of uranium in groundwater of 0.02 kgm^{-3}, for the groundwater velocity assumed of 10^{-9} ms^{-1}. This flow rate would result on average if 300 m^3 per year of groundwater flowed along the waste stacks in the current concept design [1]. 300 m^3 per year is the assumed central value for total flow through a repository in the current performance assessment [10]. A uranium concentration of 0.02 kgm^{-3} is the central value assumed in the current performance assessment for the solubility limit of uranium in groundwater conditioned by the repository NRVB, enhanced by a factor of 900 to account for the effect on solubility of the degradation of organic materials. 900 is the best estimate value for the average loading of organic materials in the vaults containing the most FM. Organic enhancement of solubility would not be expected to last over the very long timescales of the example calculations above, although it might be higher for a limited period if there were locally an above average concentration of organic waste materials in a waste stack.

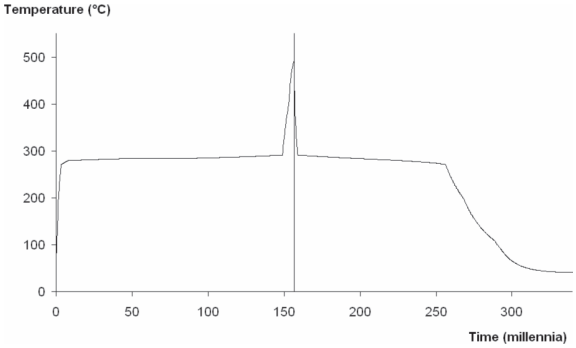

Figure 2. Evolution of average temperature in FM region for accumulation rate of 10-9 kgm-3s-1. The vertical line shows when FM stops accumulating.

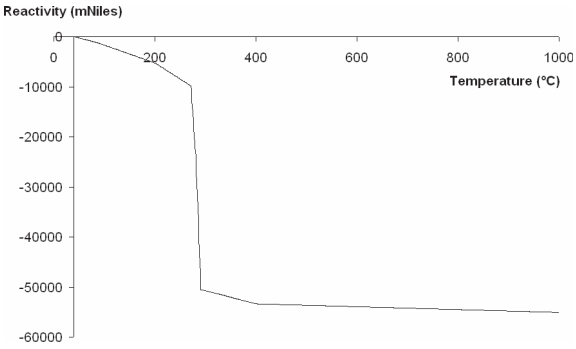

Figure 3. Reactivity as a Function of Temperature for the Initial Composition.

Similar temperature rises can be calculated with lower accumulation rates if a larger critical volume is assumed, but much more FM would have to accumulate. For example, for a FM region with a radius of 1.5 m, an accumulation rate of U-235O_2 of 10-12 kgm-3s-1 produces a temperature rise of about 45°C, similar to that for the smaller volume with an accumulation rate of 10^{-10} kgm^{-3}s^{-1}. An accumulation rate of 10^{-12} kgm^{-3}s^{-1} still implies a concentration of U in conditioned groundwater ten times higher than the central value for the unenhanced solubility limit for U, and a large amount of FM would be required to achieve these temperatures, more than 200 kg. It is unlikely that such a large amount of U-235 would collect on its own for a number of reasons, one being that the average concentration of U-235 plus Pu 239 (which decays to U-235) in the disposal inventory is less than 1%. Repeating the calculation for a FM region with radius 1.5 m with 3% enriched U accumulating at the higher rate of 10^{-10} kgm^{-3}s^{-1} give a maximum temperature rise of about 140°C and maximum power of about 3 kW. With an accumulation rate of 10-11 kgm-3s-1, the maximum temperature rise was 13°C and maximum power about 250 W. Again, a large amount of FM would be required for criticality and even with the larger accumulation volume only a small volume of the repository would be affected.

Calculations have also been undertaken for Pu-239O2 and a 70%:30% mix of Pu-239O_2 and Pu 240O_2 accumulating in small volumes. The results were similar to those for U described above, as would partly be expected from the decay of Pu-239 into U-235 over the long timescales of the calculations.

8. Systems that Convert to Positive Temperature Feedback

Temperature feedback coefficients are temperature dependent and static calculations [4,5] have shown that uranium systems in NRVB can change from having negative to positive feedback as temperatures increase, for a limited range of FM concentrations and in systems fully or close to fully saturated. This is true for both the U-235 and 3% enriched U cases where accumulation occurs in a 1.5 m radius sphere, discussed in the previous section, in the former case when temperatures exceed approximately 90°C and in the latter 200°C. Once feedback becomes positive, the QSS model does not apply. These positive feedback systems could, however, be investigated using the models developed in Nirex's programme for rapid transients resulting from positive feedback, RTM [3,11] or FETCH [3].

Positive feedback would also occur when boiling temperatures were reached in over-moderated systems. Calculations with FETCH [12] suggest that, provided the accumulation continued and the rate were sufficiently high, a short transient would occur that carried the system to a higher temperature and lower saturation where the feedback again became negative. The FETCH calculations suggest that the transient would only be a few hundred seconds in duration, a very short period compared with a QSS criticality, and that the system composition would not change significantly during the period of positive temperature feedback. The reactivity function can be used to locate the higher temperature at which the reactivity returns to zero and the QSS model calculation restarted from this temperature. The QSS model can therefore be applied to such systems despite the positive feedback transient. Interestingly, there is a small range of accumulation rates great enough to ensure boiling temperatures would be reached, but not sufficient to sustain the system in a negative feedback regime once the short positive feedback transient has occurred. The temperature would start to fall back towards where positive feedback would occur. This suggests oscillatory behaviour would result.

9. Peer Review

A peer review of Nirex's programme on understanding criticality under repository conditions was performed during Summer 2006 [13,14]. 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 benchmarking and building confidence in the material response models is recognised by Nirex and is part of the ongoing programme.

10. Sensitivity Study

A large number of sensitivity calculations have been performed as part of the process of building confidence for the application of the QSS model to repository criticality safety assessment studies [15]. 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 study confirms that the reactivity function gives similar results to directly calculated reactivities. The study also shows that results are fairly insensitive to the user-input parameters for interpolating within the reactivity function, with a minor exception for high densities of Pu 240. The investigation indicates that errors of order a few Niles may be present in the reactivities calculated with the function; however, the model results are generally insensitive to reactivity variations of this size. The burn-up model is robust to the simplifying approximations. It has been found to be insensitive to the nuclear data (fission cross-sections, absorption cross-sections and half-lives), to the treatment of the fission products, and to the representation of the stable end-products of the fission product decay chains. Heat transfer is dominated by conduction and therefore the thermal conductivity of NRVB can strongly influence the temperature rise of the FM region. The composition of materials in the FM region is affected to a lesser extent, unless the change in thermal conductivity is sufficient to cause boiling of the pore water. The groundwater flow rate only contributes significantly to the heat transfer coefficient for high rates above 10^{-7} ms^{-1}. The permeability usually has very little effect, unless the temperature rise is enough to boil the pore water. Then, a change from 'pristine' NRVB (permeability 10^{-16} m^2) to fully degraded NRVB (permeability 10^{-13} m^2) could be enough to prevent the water in the fissile region from boiling. Results are insensitive to the temperature range over which boiling is assumed to occur. NRVB cracking is unlikely to occur for the chosen values of Poisson's ratio and Young's modulus; however, there is uncertainty in the relevant structural properties of NRVB. Changing the ambient pressure and temperature of the repository has a small effect on the results. Results are insensitive to the user-input accuracy options for the ODE solver.

Concluding Comments

A description has been given of the development and testing of a model of transient criticalities under repository conditions with negative temperature feedback and resulting from the slow accumulation of FM. A more detailed description of the development and testing can be found in references [5,9,12,15]. 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. Test results suggest that, assuming a criticality did occur, only relatively fast accumulation rates would lead to significantly elevated temperatures and that there would be not be enough FM available to heat up a significant fraction of the repository.

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