UNDERSTANDING CRITICALITY UNDER REPOSITORY CONDITIONS

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Introduction

The 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 Nirex would be integrated into the NDA. The mission of United Kingdom Nirex Limited (Nirex) is, 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 the PGRC is the consideration of post-closure safety. The wastes include a wide range of materials and radionuclides. The wastes contain fissile materials, the most significant in the waste inventory being Pu-239 and U-235. Engineering measures are available to prevent criticality for such time as the waste packaging affords a high level of containment. In particular, limits are being established on the amount of fissile material (FM) 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 repository. In the long term, however, after deterioration of the physical containment provided by the waste packages, 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 after closure because, for example, of the heat that would be produced affecting the engineered barriers. It is therefore necessary to consider 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 the 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. Subsequent to the reporting of these calculations and model developments, work has continued to build confidence in the results of the models, in the absence of data to fully validate the models. This paper gives an overview of the programme.

It is important to emphasise that the work programme described is not itself an assessment of post-closure criticality safety of the PGRC. The results of the work on understanding criticality will feed into developing an improved methodology for assessing post-closure criticality safety. The direction that the research takes is directed by the need to have an adequate knowledge, on a reasonably short timescale, of how a criticality event would affect the performance of the repository.

1. Nirex Repository Concept

In the PGRC, physical containment of radionuclides would be achieved by immobilisation and packaging of wastes in containers. Geological isolation would be achieved by emplacement of the waste packages in vaults excavated deep underground within a suitable geological environment. Chemical conditioning would result from backfilling the vaults with a cementitious material, called Nirex Reference Vault Backfill, NRVB . After closure of the repository the vaults would become resaturated by the inflow groundwater. The resulting porewater would be characterised by a high pH from the dissolution of the soluble constituents of the NRVB and waste encapsulation grouts, and by a low redox potential resulting from the corrosion of steels. The backfill would also contribute to the sorption capacity of the repository. Together, these characteristics would form a chemical barrier to the migration of many radionuclides from the vaults into the surrounding rock. They would limit the movement and accumulation of FM within and around the repository.

There are a number of standard types of waste container, including 500 l drums and 3m3 boxes. These containers are made of stainless steel. Four 500 l drums would be held in a steel crate. In the current concept, the waste vaults (for the waste packages containing most of the FM) are 300 m long, with a cross-section of approximately 16 x 16 m. The stacks of backfilled waste packages are surrounded by slabs of peripheral backfill approximately a metre thick. The waste stacks, including the peripheral backfill, have a height of approximately 10 m.

A number of aspects of the repository concept are important for criticality modelling studies. The pressure in a repository once it was sealed and it resaturated would be much higher than atmospheric pressure principally because of the overlying column of rock and groundwater. In the reference design, the repository is 650 m deep. These high pressures would have a significant effect on the evolution of some criticalities.

In these studies, it has been assumed that the host rock will be a strong, fractured rock with low porosity of less than 1%. Criticality would be difficult for geometrical reasons in such a rock. In the vaults, the average porosity would be much higher.

A great variety of different wastes would be disposed of in the repository. Typically fissile bearing wastes would contain fissile material at levels of no more than a few hundred grammes per cubic metre of unconditioned waste. Some wastes would contain only Pu contaminated materials, whilst others would contain highly enriched, low enriched or natural U wastes. Radioactive decay would convert Pu-239 to U-235 over repository timescales. Once physical containment of the wastes within packages were lost, chemical processes might also alter the ratios of Pu-239, U-235 and U-238 present at a location in the repository. The mix of isotopes at a location is therefore uncertain.

2. Background

For a criticality to occur in a repository post-closure, FM from a number of waste packages would either have to concentrate within the volume of these waste packages, or migrate and accumulate elsewhere in the repository, which might be in some other waste package or set of packages, the peripheral backfill, or in rock in the immediate vicinity of the waste vaults. A number of conditions and mechanisms would have to be met or act together for a criticality to occur. Studies of the potential for a criticality suggest the probability of a criticality would be low [2]. There is a range of reasons for this low probability, including, for example, the fissile limits on individual package contents, low groundwater fluxes, chemical conditions limiting solubility and sorption, and geometrical constraints. The design of the repository concept would therefore in general reduce the likelihood of a criticality. It has not been possible, however, to quantify fully and hence discount the potential for a criticality, because of 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.

The consequences of a criticality, should one occur, have also been studied because it is not possible to discount entirely the occurrence of a criticality. If there were a criticality within the repository or its vicinity, the rock above would provide sufficient shielding and stop the immediate release of activity and prevent the direct adverse effects to man that are normally of concern for such an event. It is the effects on the barriers to the release of radionuclides that might degrade repository performance that are of potential concern. For example, the heat released by a criticality might degrade the backfill and affect the chemical barrier to the release of radionuclides. Early studies suggested that the effects would not be significant [2], at least for a single event, because a criticality could only affect a small region of the repository. These studies, however, were not comprehensive, being hindered particularly by the lack of transient models of criticalities under repository conditions. Nirex therefore began its programme on understanding criticality under repository conditions. As well as providing the models to calculate the magnitudes of the effects of a particular criticality, adequate understanding of the system and processes being assessed will be demonstrated, an important consideration for the overall safety justification of the concept.

3. Criticality Under Repository Conditions

There are two general mechanisms that might lead to a criticality in the PGRC through the accumulation or concentration of FM:

1. *Once physical containment by the waste packages had been lost, FM might be relocated in the groundwater, either by advection or by diffusion. Various mechanisms, depending on the form of the FM, might lead to an accumulation of the FM. For example, dissolved or colloidal material might precipitate out in a region where chemical conditions changed. For a criticality to occur, sufficient mass of FM would have to accumulate, how much material depending on a number of factors. The accumulation of FM in this way would be a slow process taking many years. This is significant because the rate of concentration can affect the magnitude of the effects of a criticality.*
2. *Another mechanism by which FM might concentrate is through the slumping of materials in a stack of waste packages over time. For this to occur to a great enough extent for a criticality would require most of the other materials in a small region of a waste stack to be dissolved into the groundwater and carried away, leaving the FM behind. This could only occur if locally there were enough FM and a relatively very high rate of groundwater flow (a repository would only be located where there were a low overall groundwater flow) and under conditions that would prevent the FM also being dissolved away. The concentration or accumulation of FM through the slumping of materials would most likely also be a slow process.*

Earlier work [2] suggested that two basic types of criticality might occur under repository conditions. Either type of criticality could result from each of the two mechanisms just described. These two types are quasi-steady-state (QSS) and rapid transient (RT) criticalities.

QSS criticality would result from a critical system with negative temperature feedback when FM were added. The temperature of the system might oscillate initially if there were a step addition of FM, but would settle to a steady temperature unless its temperature exceeded the boiling point of the groundwater under repository pressures. If more FM were added continuously, the temperature would continue to increase to a maximum value. A QSS criticality would eventually shutdown, for example, by burn-up. Based on earlier work, QSS criticalities would be expected to occur if fissile U or high concentrations of Pu accumulated. Work undertaken during the programme has shown, however, that under specific circumstances U systems might have positive feedback.

A RT criticality would result if the temperature feedback of the system were positive when FM were added. It was known that a RT would result for a range of Pu-239 concentrations lower than for QSS criticalities (although still at much higher concentrations than those at which FM would be disposed of in a repository). At some stage, a process would act to shut down the criticality or at least prevent further rises in temperature. There are a number of possibilities for this process in a solid medium. The coefficients controlling temperature feedback are temperature dependent and so the system feedback might turn negative as the temperature rose. Alternatively, the groundwater might boil or radiolytic gas form, which might stabilise or, at least temporarily, shut down the criticality. Increasing pressure might increase the permeability of the local materials allowing gas to form more easily. If temperature continued to rise, other materials would vaporise and an energetic transient would eventually result, dispersing the FM and shutting down the criticality or at least turning it over.

Work within the programme has not altered fundamentally this understanding of the nature of criticalities under repository conditions, although much additional *insight has been gained into, for example, behaviour in QSS systems if boiling temperatures were reached, behaviour in critical systems resulting from the slumping of repository materials, and the parameter space in which an energetic event might occur.*

4. Programme

The programme on understanding criticality under repository conditions was planned based on the understanding set out in the previous sections. During the planning stage, it was decided to develop an existing program, FETCH, to model transient criticalities. FETCH was applied in an earlier work for Nirex [2]. It was shown early in the programme that energetic transients could potentially occur and the decision was taken to develop models for such events.

The technical objectives of the programme were as follows:

1. *undertake a suite of calculations to gain insights into the behaviour of systems under repository conditions, including identifying the boundaries between sub-critical, QSS and RT systems, and provide reactivity coefficients for transient models;*
2. *develop a relatively simple model of QSS systems resulting from slow accumulation of FM, to model behaviour over long timescales and to allow the effects of a large number of systems across parameter space to be calculated quickly;*
3. *adapt FETCH and use it to examine the shut down mechanisms of rapid including energetic transients;*
4. *develop and apply a relatively simple, point kinetics model of energetic transients resulting from slow accumulation (RTM – Rapid Transient Model), to gain additional insights, improve confidence, and allow a wider range of sensitivity studies; and*
5. *apply FETCH to model the slumping scenario.*

Data do not exist to validate complete models of transient criticalities under repository conditions, although partial validation is possible. Obtaining data for validation of complete models cannot be justified on environmental grounds. It was therefore decided to develop two models of energetic events, FETCH and RTM, from different starting points and using different methods, with the intention of building confidence in the results obtained through comparison.

A parameter space was defined covering factors such as fissile materials, the properties of other repository materials, maximum fissile masses, accumulation rates of fissile materials, and thermodynamic conditions (related to repository depth).

5. Static Calculations

The approach taken was first to investigate systems of Pu-239 in NRVB, by calculating the minimum critical mass of Pu-239 for different concentrations of the FM and the temperature feedback coefficient in each case. Calculating the former showed the minimum mass of Pu-239 that would need to accumulate for criticality in a given volume of repository material. Calculating the latter allowed the boundary between positive and negative feedback systems (RT and QSS systems, respectively) to be found. NRVB was selected because it is the most common material by volume in the repository vaults. It also has a relatively high porosity, of approximately 50%, leading to a high water content when saturated and lower minimum critical masses than most other bulk materials.

Using results for Pu-239 in backfill as a base case, the effects of varying a number of parameters were investigated: Pu-240 concentration; U-235 and U-238 concentrations; ambient temperature and pressure; saturation; repository material/porosity; and iron concentration.

U systems were also investigated. Of particular interest was confirming whether or not critical U systems would always have negative feedback under PGRC conditions. Earlier work for Nirex suggested that most U systems would, although there are counter examples in the literature on waste repositories of U systems having positive feedback where the distribution of U is heterogeneous [3,4]. As already mentioned, homogeneous U systems with positive feedback have been found. Also of particular interest were systems with low concentrations of U-235 in U-238, because of the low average ratio of FM to U 238 in the disposal inventory and decay of Pu-239 to U-235 over repository timescales.

There was also some investigation of the effect of a heterogeneous distribution of FM on reactivity, and on FM being at a different temperature from the other repository materials.

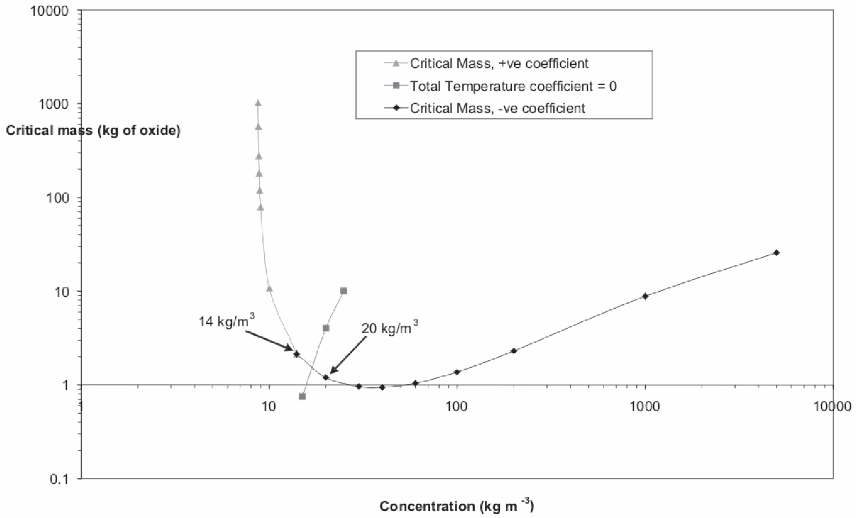

Figure 1. Example results for the base case of Pu-239 in NRVB. The boundary between systems with positive and negative feedback initially is shown.

6. QSS Model

The timescales of a QSS transient could be long enough that it is important to model decay of Pu-239 and burn-up of Pu-239 and U-235. No transient criticality model was available that could take into account decay and burn-up. A relatively simple approach was taken to developing a model for QSS transients from slow accumulation, the 'QSS model', that would have quick enough run times to undertake a range of calculations across the space of critical systems, to understand trends in the magnitudes of effects and test sensitivities.

The QSS model is a development of an analytic model of repository criticality developed in the US by Morris [5]. The conceptual model consists of a sphere containing fissile and other isotopes of Pu and U and their fission and decay products, and backfill and porewater. Pu and/or U is deposited uniformly in the sphere. The FM region is assumed homogeneous and isothermal. This region is surrounded by an effectively infinite region of backfill. A specified flux of groundwater is assumed to flow through the backfill. The transient is assumed to progress at a sufficiently slow rate that the approximation can be made that the reactivity is zero throughout the transient. As FM accumulates, power and temperature increase. FM is also lost because of burn-up and radioactive decay 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 FM 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 FM is interrupted or the pores fill. In order to accommodate the non-linear temperature feedback if the water boils, and allow for the possibility of non-constant coefficients, the model was implemented in Mathematica [6]. To enable calculations to be performed across parameter space whilst avoiding the need to calculate new reactivity coefficients for each calculation, a Reactivity Coefficient Function using a physically-based interpolation scheme was developed.

Calculations have been made with the model for a range of test cases that demonstrate its successful application.

An over-moderated system results in positive feedback if the saturation temperature is reached. The QSS model combined with FETCH have been used to investigate such systems.

7. RTM

The approach taken to developing RTM was similar to that for the QSS model, of implementing in Mathematica a relatively simple, point kinetics reactor physics formulation coupled to control-volume thermal-hydraulics. The model is based on the known phenomenology of underground explosions [7,8].

The model consists of a sphere containing a mixture of Pu-239 and saturated backfill or granite, surrounded by an infinite region of saturated backfill or rock. The FM region is assumed isothermal and homogeneous. The power generated by the criticality heats the FM region, quickly leading to vaporisation of the materials. The high pressures generated force an expansion of the FM region. Sufficient expansion shuts down the reaction. The FM region is assumed to be prompt-critical at the start. Due to the exponential growth of the neutron flux, delayed neutrons were initially neglected. A test case with a modified form of the model has showed the effect of delayed neutrons to be negligible. A Van der Waal's EoS was developed for the backfill and the Tillotson equation [9] used for the granite. Appropriate enthalpy functions have also been constructed. Two different structural response models have been implemented. Calculations have been made for test cases for backfill and granite. Empirical evidence from underground explosions can be used to predict the radius out to which fracturing would occur based on the final cavity radius. The fractured zone typically extends to about 3 to 6 times the radius of the crushed zone.

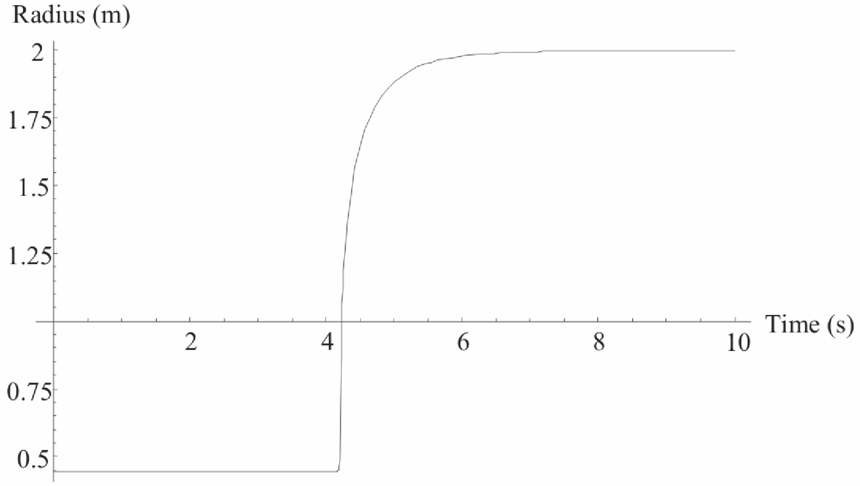

Figure 2. Example RTM results for cavity radius. 3.9 kg 239PuO2 at 10 kgm-3 in a NRVB sphere of radius 0.44 m.

8. FETCH – Slow Accumulation

FETCH [10] was developed to fully couple an arbitrary geometry radiation transport code (EVENT) with a finite element, two-phase fluid dynamics code (FLUIDITY), with options for porous media, fluidised granular material and bubbly solutions. It provides a framework for modelling spatial effects in a transient criticality, including material flows and structural response, through a symmetry in the *form of the equations solved. Earlier work [2] showed that at least one rapid transient in backfill would not 'turn over' under repository pressures just through gas generation. Early on in the programme it was also shown, using FETCH, that increasing permeability resulting from the pressure wave caused by the transient would not allow sufficient gas generation to allow the transient to turn over. It was clear, for a range of Pu 239 concentrations in backfill, that for the transient to at least turn over, materials would have to vaporise and be displaced. The decision was therefore taken to develop FETCH, and the RTM, to model such energetic events. Extensive development of FETCH was required, both within and outside Nirex programme, to adapt FETCH to model rapid transients resulting from either the slow accumulation of Pu in backfill or rock, or from the concentration of FM in a slumped region. FETCH has been applied to test cases for accumulations in backfill and granite. An axi-symmetric geometry has been used, with a sphere or cylinder of either backfill or granite containing Pu-239 surrounded by backfill or rock.*

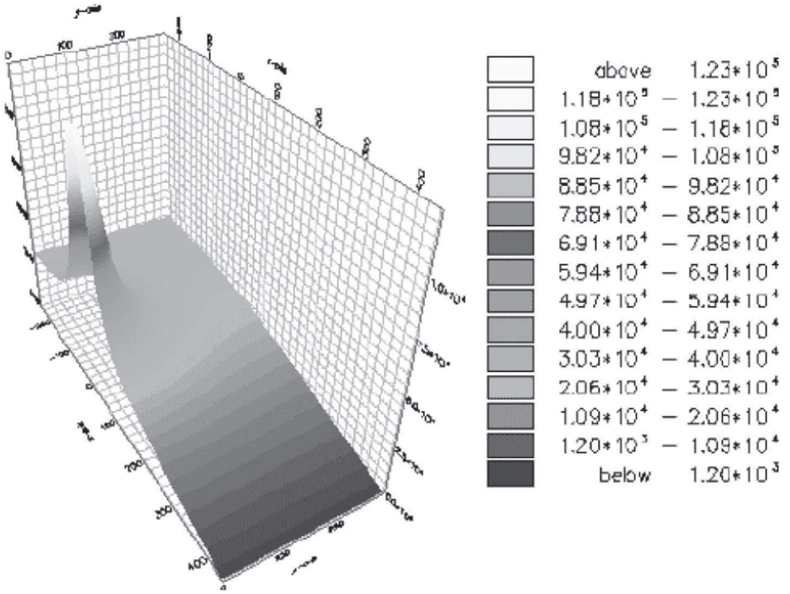

Figure 3. Example FETCH results for pressure (Pascals). 4.5 kg 239PuO2 at 10 kgm-3 in a NRVB cylinder of radius 0.416 m.

9. FETCH – Slumping

The second basic mechanism by which a criticality might occur under repository conditions is through the dissolution of repository materials, leaving a slumped critical concentration of fissile and other materials. FETCH has also been applied to modelling such criticalities. At first, a version of FETCH designed for modelling time dependent criticality of fluidised granular FM was applied, but the focus switched to developing a single tool for modelling transient criticalities under repository conditions, as described in the previous section. This has allowed the modelling of energetic events that might disrupt the solid materials surrounding the slumped volume in which the criticality occurs. Again, an axi-symmetric geometry has been assumed, in which a water-filled, cylindrical slumping region is surrounded by backfill. Silica particles containing FM settle through the water until criticality is initiated. This is a simulation model using a representative material for which the relevant parameters are known. The concentration of FM is set such that with the particles fully slumped the system would be supercritical. Calculations for test cases for both positive and negative feedback systems (when fully slumped) have been performed. A range of cases have been considered, exploring the effects of different diameter slumped volumes, open and closed conditions on the top surface of the slumped volume, different boundary conditions on the other boundaries corresponding to walls with different permeabilities, and different particle sizes and excess reactivities.

10. Comparison of RT Models

An important feature of the programme has been to develop two models of energetic transients under repository conditions – RTM and FETCH – with the objective of helping to build confidence in the results of the models, in the absence of data to fully validate the models. A comparison has been made of calculated temperature rises and energy releases (although not yet extent of fracturing) by the two models for a number of test cases, including one designed to represent a system in which Pu 239 accumulates inhomogeneously until criticality is reached, leading to rapid homogenisation and injection of reactivity [3]. Agreement to within a factor of three or four has been found for important parameters, adequate for assessment purposes.

11. Peer Review

A peer review of the programme was performed during Summer 2006. The peer review was undertaken by independent experts in criticality and rock mechanics, from the US and UK. The report will appear in the Nirex bibliography (see below) in the near future. 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. Given the lack *of direct benchmarking information, the approach of developing two separate models for energetic transients was thought sensible, and the reasonable agreement between them was thought to give added confidence to the overall results. It was thought important to understand the significance of fracturing in real materials, which 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.*

Concluding Comments

The background, scope and peer review of a Nirex programme developing a better understanding of criticality under repository conditions have been described. More detailed descriptions of the work can be found in other ICNC 2007 papers and Nirex reports. A bibliography of Nirex reports can be found on the Nirex website: www.nirex.co.uk. Reports can be requested from the website, by email or post.

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