

**Radioactive Waste Management**

**Gas Generation and Migration  
in Radioactive Waste Disposal  
Safety-relevant Issues**

**Workshop Proceedings  
Reims, France,  
26-28 June 2000**

NUCLEAR ENERGY AGENCY  
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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## FOREWORD

In underground repositories for radioactive waste, significant quantities of gases may be generated as a result of several processes. These gases may migrate through the engineered barrier system and the natural geological barrier. The potential impact of gas generation, accumulation and migration on the performances of the various barriers and, ultimately, on the long-term safety of a repository, should therefore be assessed in the development of safety cases for underground repositories.

At the time of writing, only a few performance assessments had addressed the gas problem in any detail. A substantial amount of experimental and modelling work had nevertheless been carried out in numerous national and international programmes. To draw together the results of this work, a review of the current status of the basic understanding of the gas issue was prepared under the joint auspices of the European Commission (EC) and the OECD Nuclear Energy Agency (NEA). The results were published as European Commission Report EUR 19122 EN at the end of 1999.

Building on these results, a workshop was organised in order to determine which questions still needed to be addressed to ensure an adequate consideration of the gas issue in safety cases for deep repositories. This workshop, entitled “Gas Generation, Accumulation and Migration in Underground Repository Systems for Radioactive Waste: Safety-relevant Issues”, was held under EC and NEA auspices in Reims, France on 26-28 June 2000. It was hosted by ANDRA, the French National Radioactive Waste Management Agency. The workshop began with sessions of invited presentations (covering current understanding, approaches to the management and assessment of the effects of gas generation in repositories, regulatory expectations, etc.). Following this, participants split into five parallel working groups to examine the main issues that need to be addressed in order to properly evaluate the effects of gas generation in repositories.

The present publication includes both the texts of the invited presentations and reports of the working group deliberations. It begins with a synopsis that highlights the main conclusions of the workshop. The opinions, conclusions and recommendations expressed herein are those of the authors only, and do not necessarily reflect those of any OECD Member country or international organisation.

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- Willam Rodwell, AEA Technology, representing Nirex (United Kingdom) and Andy Harris, AEA Technology (United Kingdom) who helped the Secretariat in drafting the workshop synthesis.
- The working group chairmen: Wolfgang Müller, ISTec (Germany); Alan Hooper, NIREX (United Kingdom); Antonio Gens, UPC (Spain); Pierre Berest, École Polytechnique (France); and Karsten Pruess, LBNL (United States), who led and summarised the debates that took place in the five working groups.
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## **PART A**

### **SYNTHESIS OF THE WORKSHOP**



## EXECUTIVE SUMMARY

Following the publication in late 1999 of an EC/NEA status report on Gas Migration and Two-phase Flow through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste\*, it was considered appropriate to hold an international workshop to present the content of the status report and to determine what needed to be done to ensure that it would be possible to address gas issues adequately in future safety cases for deep repositories. Accordingly, such a workshop was organised jointly by the NEA and the EC, and hosted by ANDRA in Reims, France, in June 2000.

The workshop was planned as a working meeting, consisting of two parts: (i) sessions of invited presentations which set the context for considering the way forward, and (ii) deliberations of working groups which were charged with addressing particular issues in gas generation and migration and making recommendations about what needed to be done to address these issues. Thirteen countries were represented at the workshop, together with the NEA and the EC. A synopsis of the workshop was to be published in proceedings that would also contain the reports of the working groups and written versions of the invited papers. The first two of these appear in Part A of these proceedings, and the last in Part B.

The synopsis aims to draw out the overall conclusions and recommendations of the workshop. Many of these are specific to particular repository concepts and host rocks, and as such cannot be satisfactorily summarised in a short executive summary. However, the following general remarks can be made.

- a) The most pronounced specificity of issues was for the unique example of a repository concept located in an unsaturated rock, namely Yucca Mountain (USA). While some of the issues for this site were shared with those for saturated sites, especially ones related to modelling, there were fundamental differences in many of the processes occurring. A thorough presentation of developments at Yucca Mountain was provided, but the bulk of the deliberations of the workshop focused on repositories in saturated host rocks.
- b) There was generally a reasonable level of confidence that, for most types of waste currently under consideration for deep disposal, it would be possible to develop repository concepts for which satisfactorily robust arguments could be deployed to demonstrate that gas generation would not compromise the safety case, albeit that there were still issues to be resolved. It was recognised that processes involving gas in repositories and the geosphere were complex and often coupled. This meant that it may not be practicable to provide quantitative predictions at the field scale based on detailed process modelling. Alternative,

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\* W.R. Rodwell, A.W. Harris, S.T. Horseman, P. Lalieux, W. Müller, L. Ortiz Amaya, and K. Pruess, *Gas Migration and Two-Phase Flow through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste*. A Joint EC/NEA Status Report published by the EC, European Commission Report EUR 19122 EN, 1999.

more empirically based models were expected to deliver the requirements of performance assessment. Suitable representative data at an appropriate scale would be needed to justify such models, but it would also be necessary to demonstrate a proper understanding of fundamental processes to provide confidence in the reasonableness of more empirical models.

- c) Gas generation rates are likely to be substantially higher for repositories for ILW and LLW than for those for HLW or spent fuel, because of the larger volumes of metals and organic materials in the former compared to the latter. The possibility of the production of quantities of radioactive gases that might be sufficient to cause hazards if they reached the surface is largely confined to the former types of wastes.
- d) It was considered that the relevant scientific issues pertaining to gas generation are satisfactorily understood, but that there is a need for additional data and more robust predictive models. For ILW (and LLW), the focus of future work should be on microbial gas generation, building confidence in models and developing statistically based empirical models. For HLW and spent fuel, better characterisation of corrosion processes was the priority, both in terms of factors influencing corrosion and the accuracy of data.
- e) It was recommended that design measures should be sought to minimise gas generation and mitigate its consequences. A number of measures that were currently being employed were indicated and others suggested. There was a view from the regulatory standpoint that such measures might be expected as part of repository design optimisation. Gas issues should be addressed at an early stage in the development of a repository concept.
- f) There is a general need to understand better early transitional phases in repository development, during which resaturation is occurring and the longer term pseudo-equilibrium chemical conditions are being established. Thermal effects could have their greatest significance during this period. Both gas generation and buffer performance (e.g. bentonite or crushed salt) would, for example, be sensitive to the changes taking place during this period.
- g) Gas migration through bentonite-based buffer materials is a key issue for many repository concepts. While it was recognised that there are different views in the scientific community about the mechanism of this migration, there was a consensus at the workshop that gas migration through compacted saturated bentonite, at least, is not likely to occur without microfissuring of the clay. There is nevertheless a need for further careful experiments to elucidate the mechanism of gas migration through buffer materials. The stress tensor would be an important controlling factor on fissure generation. Model development is needed both at the process simulation level to develop understanding and interpret laboratory experiments, and following a more empirical approach to provide field-scale PA tools. The latter would need to be supported by suitable large-scale experiments. Resealing of bentonite buffers was expected after gas migration, but more confirmatory data is needed.
- h) The workshop formed a similar view about gas migration through clay host rocks as for clay buffer materials, with at least the more plastic clays expected to behave in an analogous fashion to bentonite buffers. However, the data on gas migration through clays, particularly indurated mudrocks, is very scarce and there is an urgent need for further data acquisition. Gas migration is likely to be sensitive to rock weaknesses. The resealing capacity of both excavation disturbed zones (EDZ) and pressure-induced fissures is expected to depend on both the bond strength and the mineralogy of the clay. Some indurated mudrocks may behave more like fractured rocks than intact clays.

- i) Overpressurisation caused by geosphere (as opposed to buffer) properties is not likely for fractured crystalline rocks. It could occur for clay or salt host rocks, if the gas generation rate is sufficiently high, which makes it more of an issue for ILW and LLW, than for spent fuel and HLW. For the latter wastes in repositories in intact salt, lack of water should limit gas generation, so overpressurisation is generally only an issue for altered evolution scenarios involving water ingress. Gas transport through an EDZ could provide the preferred route for gas migration in clay and salt host rocks, and a better understanding of gas migration properties of EDZs and their capacity to reseal is required. The possibility should be investigated that if gas pressure build-up was sufficiently slow, then dilatancy and porosity/permeability increase could occur and relieve the gas pressure without the incidence of microfracturing.
- j) It was expected that very little water would be displaced from compacted clay buffers and low permeability argillaceous rocks by migrating gas (or from intact salt). This may limit the opportunity for coupling between gas and water flows within these materials to enhance water-borne contaminant transport (although gas may still enhance water flows by, for example, creating fissures or by forcing water from waste containers). However, for fractured host rocks and crushed salt backfill, in particular, the possible effect of gas on water movement and hence on water-borne radionuclide transport is an important unresolved issue. Several mechanisms by which this could occur were recognised and the issue is a priority area for further work.
- k) While it was believed that the physics of two-phase flow in fractured rock is satisfactorily understood, modelling this at the field scale presents problems because of the difficulty of obtaining consistent hydrogeological parameters and constitutive relationships at the appropriate scale. The applicability of conventional continuum models on a scale much larger than the intrinsic scale of the flow phenomena occurring is also questionable. Work is needed to address these issues, in conjunction with the consideration of gas-water interactions. The “upscaling” required could be supported by field experiments designed to test the aggregated response of a rock mass or, in principle, by natural analogue data.
- l) The possibility should be considered that gas in the geosphere might increase the activity of microbial communities there and that this may affect contaminant transport.

# 1. INTRODUCTION

## 1.1 Workshop Rationale

In underground repositories for radioactive waste, significant quantities of gases may be generated as a result of several processes, principally the interaction of groundwaters and brines with waste and engineered materials present in the disposal system. In some cases, the gases may migrate through the engineered barrier system and the natural geological barrier. The potential impact of gas generation, accumulation and migration on the performances of the various barriers and, ultimately, on the long-term safety of a repository should therefore be addressed and assessed in the development of safety cases for underground repositories for radioactive waste.

Most investigations into gas migration through engineered and geological barriers of deep radioactive waste repositories have only taken place over the past 15 years, a shorter period of investigation than for other issues affecting repository behaviour, in particular groundwater transport. In the development of most repository concepts, the “gas problem” has, for justifiable reasons, also been assigned a lower priority and its investigation has commanded fewer resources than those issues regarded as most central to the safety case; all performance assessments include detailed analyses of the groundwater pathway, but only a few have so far addressed the gas problem in any detail.

Nevertheless, a substantial body of work has been carried out in numerous national and international programmes and significant progress has been made. To draw together the results of this work, a review of the current status of the basic understanding of the relevant topics was prepared under the joint auspices of the European Commission (EC) and the OECD Nuclear Energy Agency (NEA). This work, which was commissioned by the EC, in the framework of its R&D programme on Nuclear Fission Safety, and by several national waste management organisations represented within the NEA Co-ordinating Group for Site Evaluation and Design of Experiments for Radioactive Waste Disposal (SEDE), was documented in a status report published\* by the EC at the end of 1999.

The above report responds to the question “Where do we stand in relation to current understanding of issues relating to the effects of the generation of gas on radioactive waste repositories?” To address this question, it provided a review of:

- a) the available experimental evidence and conceptual and modelling approaches regarding gas generation, accumulation and migration through engineered and natural barriers of most current underground disposal concepts;
- b) the various ways gas is treated in existing performance assessment exercises and safety cases;
- c) the potential impacts of gas on repository long-term safety.

To build on the results of this report, the workshop reported in this volume focused on the complementary question:

***“What is still needed for an adequate consideration of the gas issue in safety cases for deep repositories?”***

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\* W.R. Rodwell, A.W. Harris, S.T. Horseman, P. Lalieux, W. Müller, L. Ortiz Amaya, and K. Pruess, *Gas Migration and Two-phase Flow through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste*. A Joint EC/NEA Status Report published by the EC, European Commission Report EUR 19122 EN, 1999.

## 1.2 Workshop Objectives, Scope and Limitations

The main objectives of the workshop were to:

- a) Present the EC/NEA Status Report “Gas Migration and Two-phase Flow through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste” with particular focus on:
  - the status of knowledge on safety-relevant issues;
  - “closed” questions versus “open” issues;
  - the treatment of gas in performance assessments.
- b) Develop practical ways forward to address open, safety-relevant issues and to deal adequately with gas issues in performance assessments / safety cases.
- c) Assess the overall status of the gas issue for different types of waste to be disposed of in underground repositories.

The workshop remit included consideration of the potential long-term safety impacts of gas generation, accumulation and migration in underground repositories for all types of radioactive waste and sited in all types of potential host rocks. It focused on the definition of safety-relevant issues (e.g. which are the common ones, which ones are concept- or site-specific?) and on how to address them in the future.

As indicated above, the workshop objectives and programme were purposely built upon the outcome of the recent EC/NEA Status Report. Hence, the workshop was not aimed at discussing the details of past or on-going experimental, theoretical or modelling activities. Furthermore, the following topics or issues were explicitly excluded from the remit of the workshop:

- surface disposal systems, operational or pre-closure safety;
- gas from natural geosphere sources (the atmospheric gas trapped in the repository at closure was however included in the remit of the workshop as it may influence saturation times and chemistry at early post-closure times);
- dose conversion factors for gas at the geosphere/biosphere interface (however, surface arrivals of gas were to be considered).

In addition, it was no part of the workshop remit or aims to undertake any comparison of the respective performances of different repository concepts and/or host rocks.

## 1.3 Organisation of Workshop

The workshop consisted of two main parts: sessions of presented papers and working group sessions. In order to ensure the focus of the workshop onto the key issues identified above, the presentations were confined to invited papers covering specific topic areas. Twenty one such presentations were made and written versions of the technical papers are provided in the form of extended abstracts and/or full papers in the Workshop Proceedings included as Part B of this volume.

The introductory session (Session I) was devoted to the presentation of summaries of most of the EC/NEA Gas Status Report mentioned in section 1.1. Not only was this to acquaint participants with the content of the report, but to provide a common, general technical background to all participants to the workshop, and indicate the series of outstanding issues to be debated. It was also

intended help reduce any difficulties encountered due to differences in terminology in the various disciplines covered and waste management programmes represented.

No presentation was provided of the parts of the EC/NEA Gas Status Report relating to the treatment of the gas issue in performance assessment exercises. Instead, Session II of the workshop was devoted to presentations on general aspects of dealing with gas-related issues in several recent performance assessment exercises. Discussion of the unique example of a repository sited in an unsaturated host rock (i.e. Yucca Mountain, USA) was addressed in this Session.

Session III was concerned with the general approaches and expectations of regulatory authorities, and/or their scientific advisory bodies, regarding gas-related issues.

To provide a forward look, Session IV was aimed at providing short insights into strategies being developed to deal with gas-related issues in various waste disposal programmes. Elements of approaches considered included experimental data gathering (e.g. large-scale *in situ* test), theoretical and conceptual developments, modelling, incorporation of the treatments of gas into performance assessment, and optimisation of repository design *vis-à-vis* the gas issue.

A major element of the workshop was an extended session (Session V) in which the workshop participants separated into five working groups for in depth discussion of specific gas-related, safety relevant issues. To help reach one of the objectives of the workshop, these working groups were to attempt to provide concrete recommendations on how to address these issues in the future. The selection of topics to be addressed by the working groups was intended to embrace all unresolved, potentially significant, safety relevant, gas-related issues that had been identified for deep repositories.

To provide a broader perspective for the discussions to be held within the working groups, Session V opened with a presentation from an invited expert from outside the waste disposal community. This included discussion of conceptual aspects of the following issues (for details, see Part B):

- The mechanism of gas flow in low-permeability, plastically deformable barriers, for which it was noted that the displacement of “free” water by gas is not feasible and that gas migration must involve microfracturing. This is consistent with current concepts of gas migration in saturated low-permeability clays (see discussion of working group C).
- The role of transient effects, depending on the stress field and solid-fluid interactions, in controlling two-phase flow phenomena, for example permeability reduction as a consequence of rock convergence.
- The dependence of barrier integrity on the imbalance of radial and tangential stress, which if sufficiently great can lead to rock failure with dilatancy and a consequential increase in permeability and porosity.

The topics addressed by the working groups were:

A) Chemical Effects for Repositories for High-level Waste and Spent Fuel, which was to include:

- Gas generation in high-level waste and spent fuel repositories.
- Geochemical evolution of the near field.
- Effects of gas generation.

B) Chemical Effects for Repositories for Intermediate-level Waste, which was to include:

- Gas generation in intermediate-level waste repositories.
- Geochemical evolution of the near field.
- Effects of gas generation.

C) Gas Migration through Bentonite and Natural Clays, which was to include:

- Consideration of all processes relevant to safety assessment.
- Identification of pressure-induced pathways.
- Effects of pressure-induced pathways on aqueous transport of radionuclides.
- Definition/conceptualisation of breakthrough.
- Consistency (concepts and results) of classical two-phase flow models and new models.
- Contribution of new models; for example models invoking pathway dilatation, fractal theory based models, etc.
- Dependence on models of clay microstructure/macrostructure.

D) Overpressurisation and its Consequences, which was to include:

- Mechanical integrity of the near-field.
- Mechanical integrity of the far-field.
- Induced damage.

E) Effects on Contaminant Transport and Groundwater Flow in the Geosphere, which was to include:

- Whether gas affects water movement.
- Enhanced radionuclide release.
- Two-phase flow in fractured formations.

The outcome of the working group discussions were presented by the working group chairpersons to a plenary workshop session (Session VI). They were also documented by the chairpersons in the reports included in Annexes 1-5 of this synthesis.

## **1.4 Structure of the Synthesis**

The synopsis of the workshop that follows in the next two sections consists of, first, an overview of the content of the invited presentations made in Sessions I-V of the workshop and, secondly, summaries of the conclusions and recommendations of the five working groups.

The NEA/EC status report on gas migration, which was the starting point for the workshop reported here, included, by way of summary, a tabulation of the current status of gas migration issues for a set of reference repository concepts. To relate the outcome of the current workshop to the earlier report, this table has been updated in the light of the outcome of the workshop and the revised table is presented in section 4.

## **2. OVERVIEW OF PRESENTATIONS OF WORKSHOP SESSIONS I-V**

As already noted, Session I of the workshop was devoted to providing a summary of the contents of the recently produced status report on gas migration and two-phase flow in repositories. Summaries, including an overall summary of the document, are provided in Part B of this volume, so reference to these should be made for an account of the presentations of this Session.

The presentations of Sessions II-V were intended to provide authoritative examples of the way issues arising from gas generation in repositories have been treated in past assessments, are being treated in current repository-development strategies, and might be expected to be addressed by regulators. It was not possible in the time available for the presentations to provide an exhaustive coverage of these topics [coverage of a wider range of past assessments can be found in the aforementioned current status report (section 1.1)], but rather to provide informed background and stimulation for the working group sessions. It is however possible to highlight some general themes arising from the presentations. Specific to repositories in saturated host rocks were the following:

- a) Gas generation rates are likely to be substantially higher for repositories for ILW and LLW than for those for HLW or spent fuel, because of the larger volumes of metals and organic materials in the former compared to the latter. In the former cases there was expected to be much more gas produced than could be transported away from the repository in solution in the groundwater (by diffusion or advection) so that a free gas-phase would form. For these wastes, the timescale for gas production could be relatively short compared to the time scales for groundwater transport of radionuclides from the repository to the surface. The possibility of the production of quantities of radioactive gases that might be sufficient to cause hazards if they reached the surface was largely confined to these types of wastes.
- b) In the cases of HLW or spent fuel, if copper were used as the canister material and in some circumstances if stainless steel were used, a free gas might not form. However, the possibility of free gas formation would still need to be considered in many cases because of the necessity to make conservative assumptions and to deal with gas production from canister contents in the event of canister failure allowing water ingress. The use of carbon steel canisters could lead to larger gas production rates which would be expected to lead to the formation of a free gas phase. In some repositories, notably in rock salt, water supply would effectively limit gas production except in altered evolution scenarios involving water ingress through excavation routes.
- c) A number of repository designs relied on the behaviour of bentonite buffer material or an argillaceous host rock to allow gas escape without thereafter compromising the low permeability of these materials to groundwater or creating fast groundwater transport pathways. The available experimental evidence was considered to support such a “resealing” behaviour, although more data was required in this area.
- d) Various design measures aimed at mitigating the effects of gas generation had been considered for some repository concepts. These included the potential use of concrete or stainless steel instead of carbon steel for the fabrication of overpacks, the disposition of access tunnels to prevent displacement of groundwater along the excavation disturbed zone of the tunnel, the use of backfill mortars with a high permeability and porosity to facilitate gas transport, and the reliance on the reaction of carbon dioxide with cementitious backfill to remove potentially hazardous  $^{14}\text{CO}_2$  from the gas phase. The use of such measures to increase the robustness of the safety case with respect to the effect of gas generation may provide a useful component of an integrated safety strategy in developing a safety case to satisfy regulators.
- e) There was generally a reasonable level of confidence that it should be possible to deploy satisfactorily robust arguments that would demonstrate that gas generation would not compromise the safety case for most repositories, albeit that there were at present a number of issues, which varied with repository concept, that still needed to be resolved.

The proposed repository site at Yucca Mountain was unique in being the only example involving an unsaturated host rock. Gas generated from the wastes at this site does not give rise to the same issues as for saturated sites, but there are complex two-phase flow issues to be understood. The comprehensive programme of work being undertaken to achieve this and build a robust safety case for this concept were described (see paper in Part B).

### **3. SUMMARY OF WORKING GROUP REPORTS**

The following subsections provided summaries of the main conclusions of the five working groups. As the working groups themselves produced rather concise accounts of their deliberations that are presented in Part B of this document, the present summaries do not reproduce the detailed justifications of the conclusions; reference to the working group reports should be made for these or for clarification of the summaries.

In two cases, working groups (B and C) drew attention to the advantage of looking for design measures that would minimise the quantities or rates of production of gas generated, or the impact of gas generation. This is mentioned here rather than in the summaries that follow, as this issue is perhaps of wider potential interest than just in relation to the remit of a particular working group. Measures (see working group B report) that could be considered might include:

- use of “passivable” materials or highly corrosion-resistant alloys for waste containers;
- engineered void volumes (to accommodate the gas generated and manage its consequences);
- minimal use of metal in containers (e.g. “thin wall” containers) or elimination of its use (using concrete instead, for example);
- use of specially-designed backfill having one or more desirable, safety-relevant properties e.g. compactable; homogenising (of chemical conditions); cement-based, designed to trap CO<sub>2</sub>; high pH, to limit steel corrosion; capacity to trap hydrogen (speculative at this stage).
- design basis, in particular contain versus disperse.
- emplacement strategy, where for example, if feasible, the wastes that represent the principal source of gas could be emplaced separately from the wastes that contain the most significant radionuclide burden with respect to the groundwater pathway.

It was important to take proper and early account of gas migration issues in repository design.

A further common theme was that it was recognised that processes involving gas in repositories and the geosphere were complex, often involving subtle coupled and non-linear phenomena. A consequence of this was that in considering these processes at the field scale, in heterogeneous media and over a broad range of space and time scales, the prospects for quantitative predictions, based on detailed process modelling were not promising. However, it was considered that alternative, more empirically based models, would deliver the requirements of performance assessment. These would represent, in an appropriate way, the overall response of the system, in safety relevant areas, to gas generation. Collection of data on a suitable scale and with satisfactory statistical representativeness would be needed to justify such models. It would still be necessary to demonstrate that a proper understanding of the fundamental processes involved in gas generation and migration existed to provide confidence that those models used in performance assessments were consistent with the actual processes that were expected to occur (see report of working group E).

Another general point that arose was the possibility that gas effects do not always lend themselves easily to bounding conservative analysis, as larger rates and amounts of gas do not necessarily imply stronger impacts on radionuclide containment. For example, more gas could mean reduced groundwater access to the wastes with reduced gas effects in the long term, or a stronger gas source may imply more stable gas pathways, with less impact on groundwater flow than a weaker gas source, for which gas pathways may be more dynamic (see report of working group E).

### **3.1 Chemical Effects for Repositories for High-level Waste and Spent Fuel (Working Group A)**

This group addressed chemical effects on gas generation in repositories for HLW and spent fuel. The important gas generation mechanisms were corrosion, which generally predominates, and (for thin-walled containers or early container failure) radiolysis. The latter was considered well understood. Corrosion of both canisters and, after canister failure, the canister contents were considered for all host rock types.

HLW and spent fuel wastes will typically either be surrounded by a bentonite based or natural clay buffer in an argillaceous or crystalline host rock, or by salt in a salt host rock.

For concepts involving clay buffers (relatively high pH), low rates of passive corrosion are expected in a well established geochemical environment, and there is reasonable confidence in predictions of upper bounds to gas generation rates, but there is uncertainty in the actual value of corrosion rates. Issues affecting gas production by corrosion in these environments that need further clarification include:

- a) improved accuracy in corrosion rates (which will require clarification of measurement effects);
- b) data on corrosion under unsaturated conditions, in the presence of temperature gradients, and in the transition phase while the long-term pH and Eh are being established (in current assessments, the buffer material is usually assumed to be completely saturated at closure; this is believed to be a conservative assumption);
- c) more data on the effects of salts or impurities;
- d) chemical effects arising from the deterioration of concrete present as a structural material;
- e) determination of whether microbially induced corrosion is relevant to the corrosion of thick-walled containers;
- f) confirmation of whether the recently reported transformation of hydrogen to methane by microbes in clay is significant under repository conditions and whether it is a long term or transient process;
- g) determination of whether there might be a coupling between effects (e) and (f).

To increase confidence, it was considered that improved understanding of corrosion processes is needed, especially (i) for stainless steel in the presence of radiation, and (ii) to establish whether, because of the effect of passivation, long-term corrosion rates might be lower than generally assumed, conservatively, in assessments.

The active corrosion that is predicted in the more hostile saline environment of a repository in salt results in greater uncertainty in gas generation than for a repository in argillaceous or crystalline rocks. Those items of the above list that are not specific to clay-buffered systems also need to be considered for repositories in salt. Additionally for the latter, geochemical conditions are more complex and variable over time, involving, for example, precipitation and resolution, and corrosion shows greater sensitivity to the geochemical conditions. More detailed characterisation of the geochemical evolution in a repository in salt, and the effect of this on corrosion is desirable. For performance assessment purposes, simplified models of the geochemistry in repositories in salt are needed.

Other points to note were that the effects of water-supply limitation on corrosion may need to be considered for some repository concepts, and that the significance of experimental results showing the removal of hydrogen by reactions with radiolytic oxidants ( $\text{H}_2\text{O}_2$  and  $\text{O}_2$ ) or oxidised uranium that are catalysed by the spent fuel surface needs to be assessed.

### **3.2 Chemical Effects for Repositories for Intermediate-level Waste (Working Group B)**

This group believed that all relevant scientific issues pertaining to gas generation in repositories for intermediate-level waste are well understood and the relevant processes well characterised. The main requirements that exist are for improvements in the quantity and precision of certain data, but this was case dependent.

The main challenges in addressing gas generation for intermediate-level waste were in:

- the complexity of the wastes in terms of their physical and chemical characteristics;
- the coupling of events and processes; and
- the relationship of gas generation, accumulation and migration to disposal system design (see introduction to section 3).

It was considered helpful to consider requirements in relation to two phases in the evolution of the repository:

- the pre-closure and saturation phase, in which there would be significant and rapid change in the repository, which would be highly heterogeneous;
- a long-term phase in which conditions are approaching equilibrium and behaviour is one of a more homogeneous system. This phase relates particularly to the availability for release of radionuclides dissolved in groundwater; the transport of these could be influenced by gas pressurisation and migration.

The sensitivity of processes occurring in the latter phase to the chemical conditions established during the earlier phase was an important factor. Relevant information was:

- the chemical and physical state of the near field, in particular the fluid flow properties and key geochemical parameters;
- the quantities of gas generating material remaining;
- the approximate time at which radionuclides dissolved in groundwater would be available to be transported from the repository (this might, for example, be determined by the timing of canister failure).

Although consideration of pre-closure safety fell outside the scope of the workshop, comment was made that control measures were available that could ensure safety during this phase.

The information required to determine the gas generation rates during the resaturation post-closure phase was considered to be:

- The quantities of water available to supply the water demand for gas generation.
- A long-term average corrosion rate to give an upper bound to the quantity of corrodible material available after resaturation.
- The early peak release rate of all gases (to test whether this could cause overpressurisation damage).
- A representative assumed constant temperature (above the long-term value but setting aside possible short term transient peaks).
- In the event that microbially generated gases are significant, release rates that can be shown to be conservative with confidence.

To deliver these requirements, future work should be mainly on gas generation by microbial processes, and should include in particular:

- More testing of gas generation models against results from carefully designed experiments.
- Inter-programme comparisons of generation models.

In the long-term phase the relevant safety issue was the effect of gas on the geosphere transport of radionuclides. Evaluation of this required knowledge of the time-dependence of the gas generation rate and/or the increase in volume of trapped gas. Possible influences on gas generation would include:

- pH/Eh and their evolution with time;
- salinity or solution composition;
- temperature (assumption of a conservative constant value was recommended);
- galvanic coupling;
- nature of corrosion product films and their influence on corrosion rate;
- radiolysis.

The effects of the gas would depend on the extent to which it dissolved or reacted, and hence on the nature of the gas.

It was neither necessary nor practicable to have a detailed description of the combination of processes relevant to the range of materials present in ILW repositories, but some reliable statistically based method of predicting the gas generation rate was required. The accuracy required in estimates of gas generation rates, and hence requirements for information, would be dependent on the degree of uncertainty that was acceptable when considering the effects of the gas; it should be determined by sensitivity studies on a case-by-case basis.

Overall, although it was considered that the issues relevant to gas generation in an ILW repository were generally well understood, there was a need to build confidence in some areas:

- microbial generation of gases;
- the treatment of the transition from “unstable” and heterogeneous to quasi-equilibrium and homogeneous; and
- statistical averaging methods for predicting the rate of gas generation by a range of concomitant corrosion processes.

### **3.3 Gas Migration through Bentonite and Natural Clays (Working Group C)**

The materials considered by this working group covered bentonite engineered barriers and seals and argillaceous host rocks covering the spectrum from plastic clays to indurated mudrocks. Two key, and probably related, issues were uncertainty in the mechanism of gas migration in these materials and the lack of sufficiently comprehensive experimental data. These had implications for modelling and performance assessment. These issues are discussed in the following subsections:

#### **3.3.1 *Gas transport mechanisms***

There was a consensus that there were three possible mechanisms for gas transport in initially saturated clay materials associated with repositories:

- a) Transport of gas dissolved in groundwater by diffusion and advection. This was a well understood process that could be satisfactorily modelled, although in many cases it would be inadequate to transport all the gas generated.
- b) Flow of gas through existing porosity. There is a strong tendency for this flow to localize in the largest connected pores or fissures. Therefore, little displacement of water is expected. A threshold pressure basically related to the apertures of the pathways would have to be exceeded by the gas pressure before gas migration occurred.
- c) Flow of gas via self-created pathways. This mechanism was considered to be controlled basically by the stress tensor, in particular the direction and magnitude of the minimum principal stress component, and this would tend to impose a direction on the gas migration. There would again be a gas pressure threshold value for the mechanism to occur, but this would depend on the stress state (and possibly the stress history) rather than the pore structure. Again there would be little displacement of water.

The mechanism that will occur in a particular instance will be that with the smallest threshold pressure. Therefore, mechanism (c) is likely to be dominant in low-permeability materials. Mechanisms (b) and (c) may appear quite similar when observed macroscopically, although they are fundamentally different. Mixed modes are possible in which both occur simultaneously, or in which features of both are involved (e.g. gas migration through preexisting pores or fissures which dilate in response to an elevated gas pressure).

In unsaturated argillaceous materials, connected gas-filled pathways would already exist and could transport gas as conventionally assumed for porous media.

### **3.3.2 *Experimental observations of gas migration***

In spite of significant efforts to characterise gas migration in the laboratory, which led to the consensus summarised above, there remained significant uncertainties that needed to be resolved. This was no doubt due the particular difficulties in carrying out experiments in this area, including the sensitivity of results to the testing conditions.

Further experimental programmes that seek to elucidate the fundamental issues of gas migration through argillaceous materials are required, to provide a sound basis for further advance.

### **3.3.3 *Modelling approaches for gas migration***

Conventional concepts of two-phase flow (capillary pressure, relative permeability), and the modelling approaches based on these, may not properly represent the mechanisms involved in gas phase invasion of saturated low-permeability clayey material, especially when mechanism (c) above is involved. Alternative approaches, being developed to take account of the physical basis of the process, should be better able to reproduce satisfactorily many of the features of gas migration by mechanism (c) [or even (b)]. There was some evidence that conventional-two phase flow models applied to random heterogeneous media may be able (with appropriate choices of parameters) to simulate satisfactorily gas migration by mechanism (b).

Models designed to represent the development of discrete gas pathways in clays are likely to be too restricted in application for full scale performance assessment calculations, and the development of continuum models that would satisfy the needs of the latter was considered a priority. Factors that need to be considered in developing and applying the required models include:

- coupling with mechanical phenomena;
- experimental data specifically derived for the definition of the model;
- understanding the basic gas migration mechanisms.

In addition to the continuum models, the physically based models will have a role in developing the required understanding of gas transport models. Therefore, a range of models designed to explain and assess the phenomenon of gas migration in low-permeability saturated clays are likely to coexist for the foreseeable future. Gas flow in unsaturated clays could be modelled using conventional two-phase flow approaches.

### **3.3.4 *Safety relevant issues***

The most important safety relevant issues in this area were:

- the possibility of causing permanent damage to the barrier performance of buffer or host rock;
- the amount of water displaced by the gas because this water may be contaminated.

For barriers and seals, it was believed that the evidence suggested that any disruption caused by gas migration was likely to reseal and that only a small amount of water would be displaced by the gas. However more knowledge was needed to confirm this from laboratory and large-scale tests.

For argillaceous host rocks, gas migration through the excavation disturbed zone (EDZ) and the rock matrix was considered.

In both cases it was considered that little water was likely to be displaced, but that resealing capacity would depend on the bonding strength and mineralogy of the barrier or rock. Additionally, for “intact” rock, the presence of rock weakness would play a strong role in determining the pattern of gas migration.

Data on gas migration through argillaceous rocks was extremely scarce and there was an urgent need for a comprehensive laboratory testing programme. The need for large-scale in situ tests would depend on the particular repository design and the preliminary assessment of the importance of gas migration issues.

### **3.4 Overpressurisation and its Consequences (Working Group D)**

The starting point for the discussion of this working group was that gas generation would lead to the formation of a free gas phase in the repository vaults, and that at least partial gas confinement and pressure build-up would occur. Since this was unlikely to be the case for a repository in fractured crystalline rock, attention was confined to repositories in salt and indurated clay. Apart from the properties of the geosphere itself (mainly its permeability), other features of repository design would could affect overpressurisation:

- a) It is well established that an excavation disturbed zone (EDZ) with a thickness of a few metres is likely to develop around rooms, galleries and shafts. In the EDZ the rock permeability is likely to be around 100 times that of the virgin host rock. While significant quantities of data are available about the properties of disturbed zones, a “damage”– versus – permeability relationship has not been determined for indurated clays.
- b) To minimise the effect of the EDZ, which could provide a preferential path for gas or liquid migration, “keys” will be set in an attempt to seal the EDZ. Bentonite would be typically used for these seals.
- c) Void spaces will exist, for example in canisters; these may help to accommodate some of the generated gas.
- d) Resaturation of bentonite buffers and seals will strongly affect their permeability to gas. This resaturation is the normal scenario for repositories in clay; in salt it is expected only in accidental scenarios, as sufficient water would not be available to resaturate bentonite in the normal scenario (or to produce significant quantities of gas).

The working group also gave some consideration to effects of retrievability requirements and to the time scales on which various events in repository evolution might occur. Reference should be made to the working group report for discussion of these issues (Annex D).

#### **3.4.1 *Effects of gas in the near field***

In considering the effects of gas in the near field, the group came to the following conclusions:

- a) It was difficult to argue convincingly that the build-up of gas pressure would make gas production a self-limiting process by preventing water ingress; two-phase behaviour was too complex to allow this. (Note that the view was also expressed in the workshop plenary sessions that gas generation would occur to the same extent in the presence of saturated water vapour as in contact with liquid water.)

- b) The lower thermal conductivity of gas compared to water would probably mean that the presence of gas in vaults would lead to slightly higher vault temperatures.
- c) Overpressurisation was likely to lead to damage to the engineered barriers. For concrete barriers this damage would be irreversible; salt and bentonite backfilling were likely to reseal. However, few experimental results under disposal conditions were available, and further tests to address this issue were needed.
- d) Gas seepage could be a periodic process, involving, for example, repeated sequences of gas pressure build-up, crack opening, gas escape and pressure release, and crack closure.
- e) Modelling these process was difficult and needed to be built on adequate data. It required the identification of elementary mechanisms from laboratory experiments. To transfer this understanding to real systems, large scale laboratory tests followed by in-situ tests will be needed.

### **3.4.2 *Gas migration through the geosphere***

The working group considered the following issues pertaining to gas migration through the geosphere:

- Will the gas migrate through the EDZ or the intact rock?
- Will fracturing induced by high gas pressure develop upwards, which would be the most unfavourable scenario for repository safety?
- What is the influence of the pressure build-up rate?
- Will gas or water be displaced from the repository vaults?

For rock salt and indurated clay it was felt that the EDZ would be the preferred path for gas migration, although in some scenarios there may be pre-existing, but normally closed, discontinuities that could be opened by the gas pressure to provide additional routes through the intact rock itself.

The direction of fracturing would depend on many factors, such as the state of stress, the existence of planes of weakness, etc., but upward development is likely in a perfectly homogeneous, isotropic formation.

When gas pressure is close to the rock stress, dilatancy and porosity/permeability increase even before “true” fracture development occurs. Provided the rate of gas generation is not too fast, this increase in permeability can lead to pressure release and prevent full fracture development. The gas would remain confined in the vicinity of the galleries in a newly created disturbed zone. Further assessment of this scenario is considered to be of the utmost importance. (Note that if the original EDZ is not fully saturated it is expected that this will provide a route for gas to enter the rock).

Finally it was pointed out that in the case of indurated clays, galleries will be slowly filled with groundwater. Gas pressure build-up may then lead predominantly to water expulsion from the vaults. The extent to which this occurs depends on the capillary pressure for gas entry into the rock and the thickness of the gas cushion produced. This process has been modelled, but test data is needed.

### **3.5 Gas Effects on Contaminant Migration and Groundwater Flow in the Geosphere (Working Group E)**

This working group proceeded by:

- a) identifying the safety relevant issues related to presence and migration of gas in the geosphere;
- b) assessing the current understanding of gas-related effects in the geological media of interest;
- c) suggesting activities that could be undertaken to improve understanding and confidence.

Most safety relevant issues related to a potential for enhanced or altered transport of radionuclides as aqueous solutes or volatiles, and included:

- forcing water (potentially contaminated) from the repository;
- interfering with and thereby altering groundwater flow and transport;
- damaging engineered and natural barriers by overpressurisation leading to gas- and hydro-fracing with subsequent faster flow and transport;
- transport of volatiles, not only waste-derived, but potentially also from natural sources (radon);
- enhanced transport through particles (colloids, surfactants, microbes) that attach to gas-water interfaces;
- mobilisation of radionuclides by enhanced microbial activity;
- altered geochemical and microbiological activity.

There was a consensus that an assessment of gas-related issues required consideration of the governing gas migration processes in the rock formations. It was felt helpful to consider crystalline rocks and “barrier rocks” (clay and salt) separately, recognising nevertheless that there may be processes common to both:

#### **3.5.1 Crystalline rocks**

Experience at the laboratory scale suggested that classical continuum models provide an adequate conceptual basis for modelling the behaviour of two-phase gas-water systems with a bulk flowing gas phase in rock fractures. Difficulties arise in field-scale system in the following areas:

- a) obtaining consistent hydrogeological parameters and constitutive relationships (permeability, porosity, relative permeabilities, capillary pressures) for complex fracture networks on large scales;
- b) the applicability of continuum models (including “dual continua” models) when volume averaging is performed on a scale that that may be much larger than the intrinsic scale of the flow phenomena (this problem may be related to that of the preceding point);
- c) characterising the potential effect of migrating gas on groundwater flow and transport;
- d) determining the contribution of bubble flow to gas migration and to transport of contaminants by attachment to the gas-water interfaces of bubbles.

These issues require further research.

Other considerations noted were:

- The presence of gas might increase the activity of microbial communities in rocks, by providing a source of energy and carbon; this may affect contaminant transport.
- The impacts of gas flow may be enhanced by certain hydrogeological conditions, such as faults and flow funnelling structures; gas flowing along such structures may cause some mechanical erosion, thereby altering the hydrogeological properties of the rock mass.

### **3.5.2 *Barrier rocks (salt and clay)***

For clays the conclusions of his working group were in line with those of working group C, and so that discussion will not be repeated here. In both undisturbed clay and salt, gas migration may not be possible because of the extreme “tightness” of the materials. Gas migration would only occur if gas pressure built up enough to cause fractures, although there was the possibility that, with sufficiently slow gas pressure build-up, the materials may respond by mechanical creep rather than fracturing. Fractures, if formed, may allow groundwater to inflow and access the wastes, but experimental data is available to suggest that self-healing of these fractures is likely to occur. These are areas of ongoing research.

Other points noted were that (i) there is evidence that the unique barrier properties of pure salt and clay can become significantly changed when modest impurities of other rock forming minerals are present, and (ii) the behaviour of some clay and salt formations on a large scale may be more similar to that of crystalline rocks.

### **3.5.3 *Confidence building***

There was felt to be a need for field observations and experiments to test conceptual and quantitative models to build confidence in understanding of processes and parameters, especially those designed to bring out the aggregated response of a rock mass on a larger scale, and thereby improve the possibilities of meaningful characterisation, modelling and prediction at such scales.

A number of types of experiments were suggested as examples of possible approaches; these are presented in the working group report. It was also considered that observations of natural systems (analogues) could play an important role in building confidence. Again, some specific examples are included in the working group report.

## **4. SUMMARY TABLE**

The final chapter of the NEA/EC status report on Gas Migration and Two-phase Flow through Engineered and Geological Barriers for a Deep Repository contained a tabulated summary of the status then of gas migration issues for a set of reference repository concepts. To relate the results of the recent workshop to the earlier report and to provide an up-to-date summary of the status of gas issues in repository performance assessment, this table has been revised to take account of the conclusions and recommendations of the workshop. The revised table is shown as table 4.1 below. The revisions made to the table as a result of the workshop are shown in italics. The table is structured round a set of reference repository concepts. These are fully described in the Current Status Report, but, for ease of reference, the set relevant to table 4.1 is tabulated in table 4.2.

In accordance with the stated objective of the workshop to answer the question “What is still needed for an adequate consideration of the gas issue in safety cases for deep repositories?”, it is intended that table 4.1 should include reference to all recommendations for future work to address the gas issue that resulted from the workshop deliberations.

**Table 4.1 Summary of the Current Status of Gas Migration Issues for the Reference Repository Concepts (Italics represent updates to the table in Rodwell *et al.*, 1999\*)**

Repository concept	Fractured rock L/ILW	Fractured rock spent fuel	Clay/mudrock L/ILW	Clay/mudrock spent fuel/HLW	Salt spent fuel / HLW
Issues	(1)	(2)	(3)	(4)	(5)
<b>Gas generation</b>	<p>Significant gas generation expected; predictive capability still being developed; some process uncertainty. Conservative bounds may be adequate for some Pas.</p> <p><i>Further work needed on microbial gas generation, including: more testing of models against experiments; inter-programme model comparisons, and on: the treatment of the transition from an initial heterogeneous environment to a long-term quasi-equilibrium one; the development of statistical averaging methods to predict gas generation from a range of concomitant corrosion processes.</i></p>	<p>With corrosion resistant canisters, most significant source is from canister contents as a result of water ingress through canister defects.</p> <p><i>Further work on corrosion rates needed to: improve accuracy in rates; provide data for unsaturated conditions, in temperature gradients, and during development of Eh and pH; provide data on effects of salts and impurities and structural concrete degradation products; determine relevance of microbially induced corrosion; Confirm microbial conversion of H<sub>2</sub> to CH<sub>4</sub>.</i></p>	<p>As for concept (1). Conservative (upper bounds) may be more difficult to employ because of lower rock permeability than for concept (1).</p> <p><i>Further work as for concept (1).</i></p>	<p>Limited gas generation from corrosion and radiolysis indicated by experiments. May be no formation of a free gas phase.</p> <p><i>Use of carbon steel canisters could lead to higher gas generation rates and a free gas phase.</i></p> <p><i>Further work as for concept (2).</i></p>	<p>Gas generation from corrosion and radiolysis water-supply limited (relative importance of corrosion and radiolysis depends on nature of canister). Water-inflow scenarios would increase gas production.</p> <p><i>Further work on corrosion rates needed to: Improve accuracy in rates; provide more detailed characterisation of the geochemical evolution of a repository in salt and its effect on corrosion; develop simplified models of the geochemistry in salt for PA.</i></p>
<b>Design measures to mitigate effects</b>	<p><i>Use of “passivable” materials, stainless steel or concrete for waste containers; minimising use of metal in containers; engineering void volumes to accommodate gas; disposition of access tunnels to prevent displacement of water; emplacement strategy to separate principal gas generating wastes from those with highest radionuclide burden; etc.</i></p>				

Repository concept	Fractured rock L/ILW	Fractured rock spent fuel	Clay/mudrock L/ILW	Clay/mudrock spent fuel/HLW	Salt spent fuel / HLW
Issues	(1)	(2)	(3)	(4)	(5)
<b>Overpressurisation from far-field gas migration</b>	Typical fractured host rocks expected to have capacity to allow gas escape without causing overpressurisation due to capillary barriers or low permeability. May not always be the case. Reasonable confidence possible in PA predictions of overpressurisation in some cases.	As for concept (1), but with greater confidence because of low gas generation rate.	Some overpressurisation possible because of gas entry threshold pressure and low permeability of host rock. Effects of EDZ and resealing behaviour need to be understood. <i>Consensus developing on mechanisms of gas migration in clays (cf bentonite buffers), but data is extremely scarce and further experimental work is urgently required to elucidate fundamentals. Need to investigate possibility that if gas pressure build-up is sufficiently slow, permeability/porosity increase may occur before true fracture development occurs. Could prevent full fracture development.</i>	Issues as for concept (3), but mitigated by lower gas generation rate.	Host rock salt “impermeable” to gas. Any gas migration will be through crushed salt backfill (see box below).  <i>Any fractures caused by overpressurisation are likely to reseal. The possibility that a slow pressure build-up could be relieved by mechanical creep rather than fracturing should be considered.</i>

Repository concept	Fractured rock L/ILW	Fractured rock spent fuel	Clay/mudrock L/ILW	Clay/mudrock spent fuel/HLW	Salt spent fuel / HLW
Issues	(1)	(2)	(3)	(4)	(5)
<b>Gas release from near field – overpressurisation and its consequences</b>	<p>Considered that it should be possible to design containers and cementitious backfill to ensure no significant near-field impediment to gas escape. Shortage of data on two-phase flow characteristics of backfill and concrete, and limited understanding of effects of reaction with gases. Not typically a high-priority PA issue.</p>	<p>Water-saturated bentonite buffer material is main barrier to gas escape; may be essentially impermeable to gas without overpressurisation needed to overcome threshold pressure for gas entry. Continued work required to fully understand gas migration and resealing mechanisms for buffer clay, <i>but a general consensus on probable processes now exists. Effective resealing thought likely.</i> Buffer clay performance is an important PA issue.</p> <p><i>Large scale laboratory and then in situ tests will be needed, following elucidation of fundamental mechanisms in small scale experiments.</i></p>	<p>As for concept (1). Cementitious backfill is likely to be a less significant barrier to gas migration than the undisturbed argillaceous host rock.</p>	<p>Buffer material likely to behave as for concept (2). As for concept (2), work to consolidate understanding of near-field gas migration behaviour is needed.</p>	<p>The main gas migration pathway is the crushed salt backfill, whose properties converge over time to those of the host rock salt. Potential problem after compaction of crushed salt backfilling if water is available for gas generation. Analysis depends on understanding coupled processes of gas generation, two-phase flow, rock convergence and phase behaviour.</p> <p><i>Large scale laboratory and then in situ tests will be needed to build on understanding being developed from small scale experiments.</i></p>

Repository concept	Fractured rock L/ILW	Fractured rock spent fuel	Clay/mudrock L/ILW	Clay/mudrock spent fuel/HLW	Salt spent fuel / HLW
Issues	(1)	(2)	(3)	(4)	(5)
<b>Hazards from gas phase-transport to biosphere</b>	<sup>3</sup> H, <sup>14</sup> C, etc in gases may pose significant hazards to be quantified in PAs. Inventory and site specific. Work on two-phase flow in fracture networks needed to characterise distribution of releases to biosphere. <i>Understanding of CO<sub>2</sub> reaction with cementitious grout/backfill needs development.</i>	Not addressed. Quantities of gas considered too small for this to be an issue?	In principle the same potential hazards as for concept (1); fate of gas in argillaceous environments is an issue to be addressed. Likely to be site and repository specific.	As for concept (2)	As for concept (2)
<b>Effects of gas on transport of water and non-volatile radionuclides</b>	Gas expected to induce water-movement (instabilities, bubbles, gas cushions). Topic of current research. Significance and/or magnitude of issue not assessed. An important unresolved issue. <i>Possible effects of gas on geosphere microbial communities and the effect that this might have on groundwater flow and transport need investigation.</i>	Identified as a possible issue, but of secondary importance because of low gas generation rate. <i>Little displacement of water from bentonite buffer expected.</i>	Needs to be considered for this concept as for concept (1). <i>Little displacement of water from host rock expected.</i>	As for concept (3), but of secondary importance because of low gas generation rate.	Addressed through treatment of coupled gas-water flows.
<b>Resaturation</b>	Expected to occur relatively quickly in typical host rocks. Process thought to be satisfactorily understood.	Resaturation of bentonite buffers could be slow. Better understanding desirable, although the assumption of rapid resaturation, although unlikely, may be a satisfactory conservative one.	Resaturation through host rock could be very slow, so that inflows through EDZ and shafts may be significant.	As for concept (2) with more restricted supply of water from host rock.	Depends on scenario envisaged. Necessary to show that all credible scenarios are acceptable. Requires development of modelling tools indicated below.

Repository concept	Fractured rock L/ILW	Fractured rock spent fuel	Clay/mudrock L/ILW	Clay/mudrock spent fuel/HLW	Salt spent fuel / HLW
Issues	(1)	(2)	(3)	(4)	(5)
<b>Thermal effects</b>	Not considered significant for gas migration because of low thermal output of waste.	May be important in influencing buffer behaviour by drying buffer or retarding resaturation.  <i>The presence of free gas will affect thermal conductivity.</i>	As for concept (1)	As for concept (2).	Important factor in phase behaviour of salt-brine-gas system. Thermal gradients can also drive brine flows. Understanding being developed.
<b>Status of conceptual and computational models</b>	For PA, currently rely on conventional continuum porous medium models, which are considered satisfactory for treating gross effects. Alternative models needed to scope effects of flow in fracture networks, gas-water interactions, distributions of gas releases, etc. <i>Issue of “scale of representation” in models needs consideration.</i> Field-scale demonstration of models needed.	Further development of conceptual and computational models of gas migration through bentonite needed, although experimental data suggests that buffer performance is robust for key PA issues (gas escape, resealing)	Further development of conceptual and computational models of gas migration through argillaceous host rocks required. Need to address creation of gas pathways, effects of EDZ, and resealing.  <i>Possibility that some indurated mudrocks may behave like fractured rocks.</i>	As for concept (2) for buffer materials and (3) for host rock.	Development of models coupling two-phase flow, salt compaction, phase behaviour and thermal effects is a priority.
<b>General PA Comments</b>	<i>Complexity of gas processes would limit capability for detailed predictive field-scale modelling. However, alternative more empirically based models would deliver PA requirements. Collection of representative data on a suitable scale needed to underpin such models. Proper understanding of fundamental processes still needed to provide confidence.</i>  <i>Proper and early account should be taken of gas migration issues in repository design</i>				

\* W.R. Rodwell, A.W. Harris, S.T. Horseman, P. Lalieux, W. Müller, L. Ortiz Amaya, and K. Pruess, *Gas Migration and Two-Phase Flow through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste*. A Joint EC/NEA Status Report published by the EC, European Commission Report EUR 19122 EN, 1999

**Table 4.2 Representative repository concepts**

<b>Concept</b>	<b>Host rock</b>	<b>Waste type</b>	<b>Container material</b>	<b>Backfill material</b>	<b>Sealing material</b>
1	fractured rock	L/ILW	carbon steelconcrete	cementitious grout	bentonite bentonite/sand
2	fractured rock	spent fuel	stainless steelcopper	bentonite	unspecified
3	plastic clay/mudrock	L/ILW	carbon steel	cementitious grout	unspecified
4	plastic clay/mudrock	spent fuel/HLW	stainless steel	bentonite	unspecified
5	salt	spent fuel /vitrified HLW	stainless steel	crushed salt	bentonite/ bitumen

## *Annex 1*

### **REPORT OF WORKING GROUP A**

#### **CHEMICAL EFFECTS FOR REPOSITORIES FOR HIGH-LEVEL WASTE AND SPENT FUEL**

*Chairman: Wolfgang Müller, ISTec (Germany)*

*Members: Jacques Gruppa (NRG, Netherlands); Lorenzo Ortiz-Amaya (SCK•CEN, Belgium);  
Frédéric Plas (ANDRA, France); Javier Rodriguez (CSN, Spain); Patrik Sellin (SKB,  
Sweden).*

#### **1. INTRODUCTION**

Working Group A addressed the issue of “Chemical effects for repositories for high-level waste and spent fuel”. The following sections summarise the results of this working group as presented at the end of the workshop in the closing session.

#### **2. SCENARIOS**

The scenarios for gas generation in repositories for high-level waste and spent fuel are in general well understood and characterised. The initial aerobic phase after sealing emplacement areas or the whole repository is considered as negligible since oxygen consumption is limited to very short times (days or months depending on repository design). For bentonite and clay it is assumed that saturation is achieved immediately. In practice, the time span for this process may reach some years. Since this delays water contact and thus gas generation this simplification is believed to overestimate real consequences of gas generation. For all host rocks scenarios are investigated in which the waste containers fail. Direct contact of water with the waste product is treated as a separate scenario for gas generation in this case. For the development of realistic scenarios in clay presently possible effects of concrete deterioration is under investigation since concrete is considered as structural material in the repository.

#### **3. GAS GENERATION MECHANISMS**

For disposal of high-level waste and spent fuel **corrosion** is the dominant gas generation mechanism in most cases. In clay and bentonite rather low corrosion rates are expected ( $< 1 \mu\text{m/a}$ ). Since the corrosion rate is very sensitive for the performance assessment of the closure concept a higher accuracy than presently achieved seems to be desirable for these materials. In the same context, clarification is needed for the discrepancies in measured corrosion rates from different measurement techniques. In all kinds of host rock knowledge about corrosion rates in undersaturated conditions in a temperature field (gradient) is limited. Additional investigations would be extremely helpful to qualify existing models and underlying assumptions.

**Radiolysis** may become relevant as gas generation mechanism for thin-walled containers with minor shielding effect for  $\gamma$  radiation from the waste. In this case its contribution may even be dominant. In addition, radiolysis plays a role after container tightness has been lost. If water enters the container and comes into contact with the waste it depends on the container life time, i.e. the decay time for the contained activity how much gas generation has to be expected from radiolysis. It was agreed that existing information on this mechanism is sufficient for the purpose of performance assessment.

In the highly hostile environment of a container with high-level waste or spent fuel gas generation by **microbial degradation** is not expected to be of relevance. The only effect that was considered to be of interest was the interaction of microbial activity with corrosion, which is discussed below (see ch. 5).

Formation of Helium by  $\alpha$  **decay** or other radioactive or non-radioactive gases by **spontaneous fission** has been investigated by some authors. The rate and amount of gas generation for these mechanisms was, however, negligible in all cases.

#### 4. GEOCHEMISTRY

The geochemical conditions in the post-operational phase seem to be well defined for clay, granite, bentonite and concrete. In salt these conditions are complex and variable in time. Therefore, at present some possibly important processes like salt precipitation and re-solution and its influence on e.g. pH development or brine saturation at container surface can be assessed very roughly only. More detailed characterisation of the geochemistry and its development in time would be extremely helpful in deriving a realistic assessment of the gas generation rates.

Corrosion as the most important gas generation mechanism is not very sensitive to the geochemical conditions in pure clay or bentonite since it appears as passive corrosion in this high pH area. In the transition phase (during which the buffer is being saturated and the long term pH and Eh is being established), however, sensitivity may be higher. Additional information needed in this regard is the influence of salts or other impurities.

Sensitivity of corrosion against geochemical conditions is in general higher in salt brines because active corrosion takes place.

H<sub>2</sub> consumption by microbes has been observed in experiments in clay under inactive conditions. Clarification is needed if this effect appears with relevant rates and amounts under repository conditions and if it is a long-term or transient process.

A decrease in H<sub>2</sub> via reaction of hydrogen atoms, whose production is catalysed by the spent fuel surface, with either radiolytic oxidants (H<sub>2</sub>O<sub>2</sub> or O<sub>2</sub>) or oxidised uranium seems to be established by experiments. How much this effect influences the rate and total amount of gas generation needs further clarification.

#### 5. INTERACTIONS

Safety relevant interactions between different gas generation mechanisms or in combination with other phenomena so far identified are:

The availability of water for gas generation may be mass transport determined to a certain extent for all host rock media. This means an interaction exists between fluid flow and gas generation.

This interaction is considered more or less in most analyses. However, the gas generation under partially saturated conditions in a temperature fields still needs more clarification and quantification (cf. ch. 3).

Microbial induced corrosion (MIC) has received growing attention recently. For high-level waste and spent fuel in thick-walled containers this interaction may be relevant but has not yet been sufficiently addresses in all cases.

If MIC is taken into account this may interact with H<sub>2</sub> consumption by microbes. Since both effects need further clarification this interaction needs to be addressed, too.

Interaction of corrosion and radiolysis has already been mentioned in the context of geochemical questions. Further investigations should clarify if this interaction may have an effect on the stability of the engineered barrier system, too.

## 6. TIME SCALE

No general end point for corrosion induced gas generation can be given because of the rate dependence of this mechanism. Reduction effects by protective layer formation may add to this but cannot yet be quantified with sufficient accuracy and reliability. If credit is to be taken from this reduction further investigation would be necessary.

For radiolysis radionuclides are relevant which all have life times of some ten years. Therefore, the importance of radiolysis if at all is restricted to some hundred years if at all.  $\alpha$  emitters although having higher half life and higher G(H<sub>2</sub>) values will not change this observation because their activity is several orders of magnitude lower than for the  $\beta/\gamma$  emitters.

## 7. MITIGATION

To reduce or avoid gas generation a number of measures have been proposed or tested. **Chemical additives** at present do not offer a viable solution to this issue for high-level waste and spent fuel.

Regarding the dominant role of corrosion, **replacement materials** for containers have been considered, but are not readily available. An option investigated to some extent are geopolymers. But their qualification for use in waste disposal is at a very early stage. Material selection for favoured options (carbon steel, stainless steel, copper) is mainly based on reasons of structural stability and container life time, rather than gas generation potential.

## 8. PREDICTIVE MODELLING

Models for the description of gas generation as a function of time are settled for bentonite and clay. For salt open questions remain due to geochemical uncertainties (see above).

## **9. CONFIDENCE BUILDING**

Experimental and theoretical qualification of gas generation modelling and prediction has already achieved a high level. A further possible measure for confidence building has been identified for increased understanding of corrosion processes, especially in cases where interactions have to be taken into account (see above).

Natural or anthropogenic analogues are mostly limited in scope but may add to confidence incrementally, too.

## **10. PERFORMANCE ASSESSMENT**

At present transfer of gas generation models to PA is considered easy since geochemical conditions are rather stable and corrosion seems to be the only mechanism of relevance. For salt because of the higher geochemical complexity simplifications are necessary, which need to be justified by theoretical and experimental evidence.

## *Annex 2*

### **REPORT OF WORKING GROUP B**

#### **CHEMICAL EFFECTS FOR REPOSITORIES FOR INTERMEDIATE-LEVEL WASTE**

*Chairman: Alan Hooper, Nirex (UK)*

*Members: Ingo Müller-Lyda (GRS, Germany); Sylvie Voinis (ANDRA, France); Erik Frank (HSK, Switzerland); Andy Harris (AEA Technology, UK); Peter Tomse (Agency RAO, Slovenia).*

## **1. INTRODUCTION**

The Group considered that there were three main challenges to be addressed, viz:

- the complexity of the wastes in terms of their physical and chemical characteristics;
- the coupling of events and processes; and
- the relationship of gas generation, accumulation and migration to disposal system design.

In order to provide a framework for the systematic treatment of gas-related issues, the Group favoured an approach based on the definition of phases. The definition should be achieved on the basis of safety-relevant events and processes, and of strategies for the design and operation of the disposal system. A simple framework was viewed as adequate defining just two phases:

- i) “Pre-closure and Saturation” – where conditions in the near field of the disposal system are subject to significant and rapid change (i.e. unstable) and are highly heterogeneous; and
- ii) “Long-term” (Post-saturation) – where conditions in the near field are approaching equilibrium (i.e. relatively stable) and more homogeneous.

The definition of the “Long-term Phase” is related in particular to the availability of radionuclides dissolved in groundwater for release, which could be influenced by the effects of gas pressurisation and migration.

A key feature in such an approach is to determine the sensitivity of processes occurring in the long-term to the chemical conditions in the near field established in the “Pre-closure and Saturation Phase”.

## **2. THE PRE-CLOSURE AND SATURATION PHASE**

This phase was considered to have greatest safety relevance in relation to operation of a repository and to any period of keeping the repository open to monitor the wastes and to have them retrievable in a straightforward manner. In these periods the key safety issues were considered to be explosion hazards (from flammable gas/air mixtures) and radiological consequences of gases containing radionuclides.

The group considered that there was ample evidence that safety could be assured by control measures, viz: filtered ventilation systems and trapping of radioactive materials.

It was considered that gas releases in this phase would be dominated by internal package conditions, and that it would be a reasonable assumption that the water demand for the relevant gas-generating reactions would be met by the initial content of the waste packages.

The information required to determine the rates of gas generation was considered to be:

- a long-term average for corrosion rates established to give an upper bound;
- the early peak release rate of all gases (to test whether this could cause over-pressurisation damage);
- a value for an assumed constant temperature (where it was considered that high initial temperatures are too transient to be of significance and that, conservatively, a temperature rather above that expected in the long term should be assumed); and
- in the event that microbially generated gases are significant, release rates that can be shown to be conservative with confidence.

It was agreed that there were few significant outstanding issues in relation to this phase. The group recommended that any future work should focus on the generation of gases by microbial processes. Noting the availability of gas generation models, the group recommended:

- more testing of gas generation models against results from carefully designed experiments;
- inter-programme comparisons of generation models.

### **3. TRANSITION FROM PRE-CLOSURE AND SATURATION PHASE**

It was considered that key information was required on the transition to the “Long-term Phase”, as follows:

- the chemical and physical state of the near field, in particular its fluid flow properties and key geochemical parameters (defined later);
- residual quantities of gas-generating materials;
- the timing of the transition, where it was considered that this was only required to be a reasonable order of magnitude assumption that should be determined in relation to the availability of radionuclides dissolved in groundwater and/or any stylised approaches to timeframes in assessment calculations such as assumed container failures at a given time (e.g. 10 000 years post-closure).

It would be necessary to check that in the event of adopting conservative values for gas generation rates in the Pre-closure Phase, the residual quantities of gas-generating materials would not be underestimated as a result of assumed consumption. It was also decided that arguments based on the delay of repository saturation by creation of a gas bubble in the near field are not sustainable and should not be used to claim significant delays in the time for water to contact wastes.

#### 4. LONG-TERM PHASE

The Group considered that the overriding issue was the time-dependence of the gas generation rate and/or the increase in volume of trapped gas. This was because in this phase the safety-relevance is the effect on the geosphere transport of radionuclides. The key issues that would require to be taken into account were as follows:

- description of initial corrosion products on metal surfaces and their influence upon corrosion rates;
- pH evolution with time;
- nature of the gases generated e.g. CO<sub>2</sub> can be consumed by subsequent chemical reactions or dissolution in water (which processes themselves are highly pH-dependent).

There was considerable debate on how much further effort, if any, is required to measure the low corrosion rates that pertain in this phase, and in particular whether there is a minimum threshold below which detailed measurement could be demonstrated to be unnecessary. It was recognised that where the rate of transport by diffusion could be shown to exceed the rate of gas generation there would be no useful purpose in obtaining precise generation rates. More generally, it was agreed that the precision required should be related to the level of uncertainty allowable in considering effects in the geosphere. This would have to take account of the coupling of gas generation to characteristics such as water inflow rates, change of permeability or change in void volume.

It was recognised that in the long term, a range of corrosion mechanisms could be relevant, e.g. passivation, linear corrosion or galvanic coupling. It is neither necessary nor practicable to have a detailed description of the combination of the relevant processes for the range of materials typically present in intermediate-level wastes. However some reliable process of statistical averaging would be required and the Group was not aware of a successful case study in this area.

The requirements for information in relation to this phase would always be case-dependent and should be determined by sensitivity studies before investing in major research or data acquisition programmes, which may not be required. The Group was sensitive to the need to consider the degree of physical realism that might be required for a performance assessment to be acceptable.

Possible influences on gas generation would include:

- pH/Eh;
- salinity/solution composition;
- temperature (again recommended to be assumed at a conservative constant value);
- galvanic coupling (as mentioned above);
- nature of corrosion product films;
- radiolysis.

The range of conditions to be studied would be highly dependent upon the assessment basis and scenario.

## 5. DESIGN STRATEGIES

The Group agreed that, while no scientific issues had been identified that were not well-understood and all the relevant processes appear well-characterised, more attention should be given to design strategies in relation to gas generation. This would demonstrate a realistic approach to system optimisation. The areas considered worth further study were as follows:

- use of “passivable” materials (low gas-generation alloys) or stainless steel for waste containers;
- engineered void volumes (to accommodate the gas generated and manage its consequences);
- minimal use of metal in containers (e.g. “thin wall” containers) or elimination of its use (using concrete instead, for example);
- use of specially-designed backfill having one or more desirable, safety-relevant properties e.g. compactable; homogenising (of chemical conditions); cement-based, designed to trap CO<sub>2</sub>; high pH, to limit steel corrosion; capacity to trap hydrogen (speculative at this stage);
- design basis, in particular contain versus disperse (which was recognised as having important implications as to whether the build-up of gas pressure or the rate of gas generation would be of most importance);
- emplacement strategy, where for example, if feasible, the wastes that represent the principal source of gas could be emplaced separately from the wastes that contain the most significant radionuclide burden with respect to the groundwater pathway.

## 6. SUMMARY

In summary, the Group believed that all relevant issues are well-understood and the relevant processes well-characterised. The main potential requirement for further information is in respect of data and the precision of the relevant data, and that requirement is highly case-dependent.

There was a clear need for better integration of the strong knowledge base into performance assessments and design strategy.

Certain areas could be identified where there was a need to build confidence, viz:

- microbial generation of gases;
- the treatment of the transition stage from “unstable” and heterogeneous to quasi-equilibrium and homogeneous; and
- statistical averaging methods for the rate of gas generation by a wide range of concomitant corrosion processes.

Finally, it was viewed as important that the gas generation capacity of the disposal system and the relevant design features of the system should be used to determine the extent of the understanding required of the migration of the gas through the geosphere. A requirement defined in that way might prove to be readily achievable with existing knowledge.

### *Annex 3*

## **REPORT OF WORKING GROUP C**

### **GAS MIGRATION THROUGH BENTONITE AND NATURAL CLAYS**

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*Members: Wim Cool (ONDRAF/NIRAS, Belgium); Miguel Cuñado (ENRESA, Spain); Stephen Horseman (BGS, UK); Jukka-Pekka Salo (POSIVA, Finland); Kenji Tanai (JNC, Japan); Mikihiro Yamamoto (TEC, Japan); Geert Volckaert (SCK•CEN, Belgium).*

#### **1. INTRODUCTION**

Working group C addressed the question of gas migration through bentonite engineered barriers and seals and, also, through argillaceous geological media. Both plastic clays (notably Boom Clay) and indurated mudrocks were considered.

From the examination of the Joint EC/NEA Report on Gas migration and from a number of contributions to the Workshop, it appeared that there was a significant degree of uncertainty and some discrepancies concerning the basic gas migration mechanisms likely to operate in saturated argillaceous materials. It was felt therefore that it should be the first issue to be considered by the group.

A possible source of this uncertainty could be the lack of a sufficiently comprehensive experimental database, the next issue to be examined by the group. In addition, any performance or safety assessment exercise will probably require a simulation of gas migration; the adequate modelling approach to be used and its relationship with the basic gas transport phenomena was discussed. After those basic items had been examined in depth, the safety-relevant issues were addressed by the group.

In consequence, the report of Working Group C is arranged in the following sections:

- Gas transport mechanisms;
- Experimental observation of gas migration;
- Modelling approaches for gas transport;
- Safety-relevant issues.

#### **2. GAS TRANSPORT MECHANISMS**

After a full discussion, a basic consensus emerged on the relevant gas transport mechanisms in initially saturated clayey materials in spite of the fact that many details still remain uncertain. Three possible gas transport mechanisms were envisaged:

- Mechanism A: Diffusion/advection of gas dissolved in the liquid phase.
- Mechanism B: Flow of gas through existing pore space.
- Mechanism C: Flow of gas via self-created pathways.

The main features of each mechanism are as follows:

Mechanism A: *Diffusion/Advection of gas dissolved in the liquid phase*

- It is a generally accepted mechanism with limited carrying capacity.
- There is no gas pressure threshold value for the mechanism to occur.
- Successful modelling simply requires the prescription of adequate boundary conditions and the adoption of the correct diffusion coefficient and gas solubility coefficient.

Mechanism B: *Flow of gas through existing porosity*

- The mechanism depends basically on the pore structure of the material.
- It is an “extreme value” phenomenon. Generally, it affects only the most conductive continuous paths (associated with the largest interconnected pores).
- There is a gas pressure threshold value ( $P_B$ ) for the mechanism to occur, depending on pore structure. It is often called capillary threshold pressure although the phenomena involved are certainly more complex than the simple capillary model.
- There is a strong tendency for the gas flow to localise. The tendency is more marked the more heterogeneous (at pore scale) the material is. Larger scale lithological variability might also lead to flow localization in some host clay formations.
- The amount of water displaced by the gas will depend on the degree of gas migration localisation. Highly localized flow will displace little water.
- The mechanism is more likely to occur in higher permeability materials.
- If it occurs in low permeability materials, experimental observations may require a long testing time due to the need for the gas to displace water from the pores.
- Highly localized flow will create difficulties for conventional two-phase-flow modelling approach. Flow localization associated with pore-scale heterogeneity can be modelled by applying the two-phase-flow approach to random heterogeneous porous media.

Mechanism C: *Flow of gas via self-created and stress-induced pathways*

- This mechanism is basically controlled by the stress tensor.
- The magnitude and direction of the minimum principal stress are the principal factors governing the development of gas pressure induced pathways. Consequently, gas transport will usually be directional (anisotropic). The tensile strength is usually low or zero and, therefore, plays a lesser role.
- There is a gas pressure threshold value ( $P_C$ ) for the mechanism to occur. It depends mainly on the current stress state.
- Extensile rupture under triaxial-extension stress fields (e.g. EDZ) and shear-induced dilatancy under general states of stress (e.g. EDZ and shear zones) may also be important when considering pathway flow of gas in the more indurated mudrocks.
- Gas transport will localize with little (if any) dependence on pore structure.
- The amount of water displaced will probably be small (usually less than 1% of total).

- The mechanism is likely to be dominant in low permeability materials (where  $P_c$  is typically less than  $P_b$ ). Overconsolidation can be an important consideration when making judgements on this issue.
- This mechanism is difficult to model with conventional two-phase-flow models. The validity of basic concepts of relative and intrinsic permeability can be questioned. Discrete pathways models can probably reproduce better the basic phenomena.

On occasions Mechanism B and C may appear quite similar when observed macroscopically, in spite of their differences at a fundamental level. Mixed modes of gas migration in which the two mechanisms occur simultaneously are also possible. For instance gas transport through pores enlarged by the effects of gas pressure is basically a mechanism B case, but it shares some important features (mechanical coupling for instance) with mechanism C. Gas migration via pathways associated with crack dilation are also an intermediate case. An additional example concerns some indurated mudrocks, where gas migration may occur through macropores that are linked by self-created pathways.

In case of gas migration in unsaturated materials with continuous gas phase, no special difficulties arise. Under such conditions, gas flow can take place through the pores of the material (Mechanism B) and the phenomenon can be readily described by a conventional two-phase flow approach.

### **3. EXPERIMENTAL OBSERVATION OF GAS MIGRATION**

In spite of the significant efforts carried out in recent times to characterize gas migration in the laboratory, it is felt that important uncertainties still remain that will require additional efforts in the future. This situation may be due to the special difficulties that arise in the experimental investigation of gas migration through saturated clayey materials. In this respect, it should be recognized that experimental results are sensitive to testing conditions (e.g. sample size, test boundary conditions, pressure control vs. volume control). Breakthrough pressures (both initial and successive peak values) and shut-in pressures are known to be sensitive to experimental boundary conditions.

A potential example of this dependence on testing conditions is the often quoted remark that excess gas pressure at breakthrough has a value similar to the swelling pressure. This may be an artifact of the way in which the tests are carried out. Often, they are performed in an oedometer where the sample of compacted bentonite is saturated under no volume change conditions before the gas injection test. Under those conditions, the stress in the sample will be close to the swelling pressure, and, therefore, breakthrough should be expected when the gas pressure reaches that value (Mechanism C). Carrying the tests under different stress conditions might lead to a different breakthrough pressure.

In any case, experimental programmes should seek to elucidate the fundamental issues outlined above, especially in connection with the various gas migration mechanisms. Only such a fundamental approach will provide a sound basis for further advances.

### **4. MODELLING APPROACHES FOR GAS MIGRATION**

As pointed out above, a conventional two-phase-flow approach will encounter very serious difficulties in modelling gas migration through saturated clayey porous media. In fact concepts such as relative permeability or intrinsic permeability may have doubtful validity in some of the saturated argillaceous materials considered here when subjected to gas migration.

There exist alternative approaches to carry out an adequate process modelling that takes into account the physical bases of the phenomena. Discrete pathway approaches (e.g. capillary bundle, multiple front propagation, and crack propagation models) should be able to reproduce satisfactorily many of the features of gas migration by mechanism C. Two-phase-flow models applied to random heterogeneous media appear to simulate satisfactorily gas migration via mechanism B. Some discrete pathway models can also be used in relation to this mechanism.

However, it is felt that, from a practical point of view, continuum models will have to be used in Performance Assessment exercises. These might draw on the two-phase-flow approach, with modifications to mimic the observed gas transport phenomena. Such models have yet to be fully developed and should be the object of research priority. They will certainly require:

- Coupling with mechanical phenomena.
- Experimental data specifically derived for the definition of the model.
- Understanding of the basic gas migration mechanisms. The use of process models can be very valuable in this regard.

Therefore, it is envisaged that a range of models will coexist in this area for the foreseeable future. Each type of models will have its particular range of applications. In contrast to initially-saturated materials, gas migration through unsaturated materials with continuous gas phase can be modelled using the conventional two-phase-flow approach.

## **5. SAFETY-RELEVANT ISSUES**

The most important safety-relevant issues that were identified in relation to gas migration were:

- The possibility of causing permanent damage.
- The amount of water displaced by the gas.

both for the engineered barrier (or seals) and for the argillaceous rocks.

### **5.1 Barriers and seals**

Basic conceptual differences may arise from the fact of whether gas production (and hence gas migration) occurs when the barrier is saturated or whether, on the contrary, gas production starts with the barrier in unsaturated state. The latter case is more amenable to understanding and analysis.

It was concluded that, in the case of barriers and seals, any damage caused by gas migration was likely to reseal and that the water displaced by the gas would be small. However it was also concluded that more knowledge is needed from:

- Laboratory tests especially for the materials subjected to high temperatures and at high degrees of saturation. Emphasis should be placed on phenomenon/mechanism characterization, It should be noted that properties are likely to be material specific. Therefore, whenever possible, the materials intended for barriers and seals should be used in the experimental programmes, prepared using exactly the same procedure of compaction and conditioning.

- Large-scale tests mainly to observe transport mechanisms and other qualitative features at relevant scales. They should also be used as benchmarks for model validation, if sufficient information is gathered.

## **5.2 Argillaceous rocks (plastic clays and indurate mudrocks)**

Here gas flow through joints or other possible discontinuities was deemed not to be specific of the topics examined by the group. The question addressed was gas migration through the rock matrix. The analysis of the safety-relevant issues is design sensitive. Among other factors, it depends on whether gas migration occurs through the EDZ or through the “intact” host rock.

In the case of gas migration through the EDZ, it is concluded that water displacement will probably be small. Resealing capacity is expected to depend on the bonding strength and clay mineralogy of the rock. Maximum resealing capacity will be associated with plastic clays. This capacity will reduce as the bond strength of the rock increases and the proportion of expansive clay minerals decreases in indurate mudrocks. Sealing completely the EDZ to gas migration is likely to be a very difficult undertaking.

In gas migration through “intact” host rock, the presence of rock weaknesses will play a strong role regarding the patterns of gas migration. In any case, the amount of water displaced is expected to be small and the capacity for resealing will be again associated with the bonding strength and mineralogy of the rock.

In any case it was concluded that there was a great scarcity of data regarding those phenomena. Therefore a comprehensive laboratory testing programme is certainly required to examine the flow of gas through argillaceous rocks. The need for large scale “*in situ*” tests will finally depend on the particular repository design and the importance of gas migration issues to the disposal concept. In any case, the results of tests of this type would probably be rock-specific.

## **5.3 Other points**

It was strongly felt that gas issues should be taken on board at a very early stage of the design. It may affect safety and its relative importance in this respect may be strongly dependent on design features. In conclusions gas migration issues should certainly be a factor in repository design.

In fact, it was felt that the most fruitful approach to deal with those issues is to try to minimize them whenever possible via an adequate repository design. In very rough terms, the magnitude of the problem may be related to the energy product  $P$  (pressure)  $\times$   $V$  (volume). Reducing gas pressure or gas volume automatically leads to a smaller gas problem. Features may be incorporated in repository design that contribute to the goal of gas energy minimisation.



## Annex 4

### REPORT OF WORKING GROUP D

#### OVERPRESSURISATION AND ITS CONSEQUENCES

Chairman: *Pierre Bérest (École Polytechnique, France)*

Members: *Karim Ben Slimane (Andra, France); François Besnus (IPSN, France); Laszlo Kovacs (PURAM, Hungary); Thierry Lassabatère (EDF, France); Günter Pusch (Technical University Clausthal, Germany); Bertrand Ruegger (NEA, France); Jurgen Wollrath (BFS, Germany).*

#### 1. GENERAL

The working group discussed waste disposal in salt (in Germany) or indurated clay (in France and Hungary). The group assumed that advection and diffusion will not be able to accommodate the gas generation rate, leading to at least, partial gas confinement and pressure build-up. The credibility of such an assumption was not the topic to be discussed. Sets of general questions on various topics were discussed and the responses collected. Plus (+) signs indicate that the amount of available knowledge is good; minus (-) signs indicate that further progress is needed.

**Disturbed Zone** – As shown by laboratory and field experiments (++) , a disturbed zone with thickness of a few meters is likely to develop in the vicinity of rooms, galleries and shafts walls, both in indurated clays and salt. In this disturbed zone (EDZ), rock behavior is dilatant (Its apparent volume increases, due to the opening of numerous more-or-less interconnected cracks, according to the level of accumulated damage) and leads to increases in both porosity and permeability. For the case of rock salt, EDZ permeability is of the order of  $K = 10^{-20} \text{ m}^2$ , compared to the permeability of virgin salt,  $K = 10^{-22} \text{ m}^2$ . The corresponding values for Bure clay are, respectively,  $K = 10^{-19}$  to  $10^{-17} \text{ m}^2$  (EDZ) and  $K = 10^{-21}$  to  $10^{-19} \text{ m}^2$  (virgin rock). These figures are indicative rather than exact; they are based on laboratory tests (France), extensive field tests (Germany), and preliminary *in situ* tests (Hungary). Data are also available from the WIPP and Mont Terri sites (+). A “damage” – versus – permeability constitutive relationship is still missing in the case of indurated clay (-).

**Seals** – “Keys” will be set to minimize the effect of the disturbed/permeable zone (Germany, France), which could become a preferential path way for gas/liquid migration. Seal length will be roughly equal to key diameter. It is hoped that no (or only a small) disturbed zone will develop in the vicinity of such keys. Bentonite is considered to be a suitable sealing material in indurated clays and is considered for the case of rock salt.

**Canisters** – The canister “apical” void (France) can accommodate some gas generation.

**Bentonite Re-saturation** – This is included in the “normal” scenario for the case of indurated clay, but it is considered an accidental scenario (the existence of brine pockets making water available) for the case of rock salt.

## 2. NEAR FIELD

- a) Is gas production a self-limiting process (when gas accumulates in the vicinity of a canister, no water is available for further corrosion)?

This statement is widely considered to be arguable. Fingering, capillary phenomena can lead to a complex situation: parts of a canister wall are wet, parts are dry; a definite “front” separating gas and water will not exist in the bentonite.

- b) Does high gas content, or a gas film at the canister/bentonite interface, lower thermal conductivity (or, more generally, heat transport)?

Probably, leading to (slightly?) higher canister temperatures (n.b., after 200-300 years, a temperature of 80°C is expected in the bentonite.)

- c) Does high gas pressure create irreversible damage to the engineered barriers?

Likely. Some results are or will be available (Horseman’s tests, Febex III decommissioning). Few experiments on disposal conditions are available. Bentonite and crushed salt are likely to self-heal; the same cannot be said for concrete(-). Further tests are needed.

- d) Gas seepage could be a periodic process. (Gas accumulates, pressure builds up, cracks open, gas escapes, cracks close, gas accumulates, etc.)

- e) Modeling these phenomena (including boundary conditions analysis, validation, etc.) is not an easy task, as many transient phenomena play a role. “You hardly learn mechanisms from *in situ* tests”. The first steps must be to identify elementary mechanisms and perform laboratory tests. Large-scale laboratory tests will be second; last, in-situ tests will be needed.

## 3. EDZ VERSUS GEOLOGICAL BARRIER

- a) Which will be the weaker point – the EDZ or the geological barrier?

For the cases of rock salt and indurated clay, the EDZ will clearly be become the preferential pathway for gas migration (Germany, Hungary, France). Fractured granite is an exception.

Some scenarios combine the EDZ and the rock mass. A pre-existing (more-or-less closed in virgin conditions) discontinuity can be re-opened by the effect of gas pressure in a large disposal area. (In some scenarios, additional forces may assist gas pressure – e.g., thermal stresses induced by differential thermal expansion.)

- b) Does fracturing induced by high gas pressure develop upward (the most unfavorable scenario)?

It depends on many factors (state of stress, existence of weaker planes, etc.) Upward development is likely in a perfectly homogeneous/isotropic formation (+).

- c) What is the influence of the pressure build-up rate?

When gas pressure is close to the rock stress, dilatancy and porosity/permeability increase – even before “peak pressure” (i.e. frac development) is reached. The increase in permeability can prevent “true” gas fracturing and can lead to pressure release. However, it leads to the

creation of a new disturbed zone in the gas/rock contact area. When gas pressure builds up and reaches hydrostatic figures, two scenarios can be considered:

- i) the ED is not fully saturated -gas penetrates the rock formation; and
- ii) the EDZ is re-saturated. Then, pressure builds to the lithostatic figure, but fracturing does not occur – a dilatant (permeable) zone develops. Gas will remain confined in the vicinity of the galleries in a newly created disturbed zone, modifying water flow patterns.

Further assessment of these scenarios is considered to be of utmost importance.

**Gas or Water Displacement?** – For the case of indurated clays, galleries will be slowly refilled by water flowing from the rock mass. Then, gas pressure build-up will lead to water displacement (and no – or only a small – gas displacement), because, for example, capillary forces will prevent gas migration, provided the EDZ is fully saturated. These phenomena can be modeled, and testing is needed.

#### **4. OVER-PRESSURIZATION AND RETRIEVABILITY**

- a) For a couple of decades, radiolysis and microbial activity effects can be tackled through ventilation. Experience from coal mines is available (+).
- b) When longer periods of time are considered, no clear answer can be given.

#### **5. A TENTATIVE TIME FRAME**

- a) “The EDZ is re-saturated.” – For clay, three centuries is considered a maximum value. This notion does not apply to salt. (No brine is available; healing is likely, due to salt creep). Bentonite swelling is a rapid process.
- b) “Galleries are flooded.” – This scenario is considered to be accidental for the case of salt. For clay, a period of  $10^3$  to  $10^4$  years is assumed in the standard scenario. (Clay permeability is  $K \cong 10^{-20} \text{ m}^2$ ). A much shorter time frame ( $\times 10^{-2}$ ) must be considered when pessimistic scenarios ( $K \cong 10^{-18} \text{ m}^2$ ) are taken into account.
- c) “Gas pressure builds up.” – The time frame is no longer than 10 centuries for the case of vitrified wastes, but this depends on waste type, room size, and available porosity – it could be longer. Over-pressurization (i.e., gas pressure is larger than hydrostatic pressure) occurs 2-3 centuries after corrosion begins. Boundary conditions are of the utmost importance.
- d) “Radionuclide release begins.” – This depends on waste type. The process is not uniform. At the same instant, some canisters are intact, while others are corroded and some may release radionuclides.



## *Annex 5*

### **REPORT OF WORKING GROUP E**

#### **GAS EFFECTS ON CONTAMINANT MIGRATION AND GROUNDWATER FLOW IN THE GEOSPHERE**

*Chairman: Karsten Pruess, LBNL (USA)*

*Members: Paul Marschall (Nagra, Switzerland); Wernt Brewitz (GRS, Germany); Vijen Javeri (GRS, Germany); Ronald M. Linden (USDOE); Gauthier Vercoûtère (ANDRA, France).*

## **1. INTRODUCTION**

The Working Group (WG) decided to proceed by (1) identifying safety-relevant issues related to presence and migration of gas in the geosphere, (2) assessing the current understanding of gas-related effects in the geologic media of interest, and (3) attempting to identify activities that could be undertaken to improve understanding and confidence. There was consensus in the WG that an assessment of gas-related issues has to consider the governing gas migration processes in the rock formations. We began by addressing fractured crystalline rocks. In this type of rock, gas migration predominantly takes place along a complex network of pre-existing discrete fractures. “Barrier rocks” (salt, clay) were to be considered later; the WG believed that gas issues relating to these media might largely overlap, albeit with some different emphasis, with those for the crystalline rocks. In barrier rocks, due to their low permeability, gas accumulation may create new fractures (micro-fractures, gas-fractures, hydro-fractures). These induced fractures are likely to form the main gas flowpaths. Finally, some attention was to be given also to sedimentary rocks that might be present near repository host formations and thereby be impacted by gas migration.

The WG felt that safety-related issues should be identified in the most comprehensive manner possible, including issues that may appear to be of little concern, on the premise that even unlikely and unconventional issues will have to be addressed when building a safety case through performance assessment (PA). More specifically, any aspect of gas behaviour was deemed “safety-relevant,” and would need to be identified and resolved, if it could impact the procedure or outcome of repository PA. This does not necessarily imply that a great deal of research needs to be expended on all of these issues, or even that a thorough scientific understanding is required for all of them in order to meet engineering objectives.

The WG also felt that it will be necessary to demonstrate a general understanding of gas behaviour in repository host rocks and neighbouring geologic media at a repository site, regardless of specific impacts on formalised PA, in order for a repository safety case to be acceptable to stake holders. In this sense the WG believes that a pervasive safety-relevant issue associated with gas effects is a need to obtain a general understanding of such effects in geologic media. This pertains to all media and repository concepts. Specific issues are listed and discussed below.

## **2. SAFETY-RELEVANT ISSUES**

Most such issues relate to a potential for enhanced or altered transport of radionuclides, as aqueous solutes or volatiles. Effects that could impact radionuclide migration include:

- forcing water (potentially contaminated) from the repository;

- interfering with and thereby altering groundwater flow and transport;
- damaging engineered and natural barriers by overpressurization, leading to gas and hydrofracing with subsequent faster flow and transport;
- transport of volatiles, not only waste-derived, but potentially also from the host rock (radon!);
- enhanced transport through particles (colloids, surfactants, microbes) that attach to gas-water interfaces;
- mobilisation of radionuclides by enhanced microbial activity;
- altered geochemical and microbiological environment.

Another safety-relevant issue is the flammability hazard from combustible gases (hydrogen, methane).

### 3. PROCESS UNDERSTANDING – CRYSTALLINE ROCKS

Fractured crystalline rocks comprise a broad range of hydrogeologic settings, that may range from sparsely fractured to well-connected fracture networks. The individual fractures may or may not contain fill. There is a variety of “two continua” models, developed primarily in petroleum and geothermal reservoir engineering, that would be applicable to describe flow and transport in such systems, including double-porosity, dual permeability, and multiple interacting continua models. In sparsely fractured systems, fractures would have to be represented individually as porous continua with appropriate geometric and hydrogeologic parameters, rather than being lumped into effective continuum descriptions as would be appropriate for well-connected networks.

Based on experience with laboratory-scale experiments, the WG believes that an adequate conceptual basis for modelling the behaviour of two-phase gas-water systems with a bulk gas phase is provided by classical continuum models. These are based on mass and energy conservation, and a generalised version of Darcy’s law with relative permeability and capillary pressure effects. Difficult issues arise in field-scale systems, due to the complexity of fracture networks. A description in terms of a generalised Darcy’s law may still be applicable in principle, but consistent hydrogeologic parameters and constitutive relations (permeability, porosity, relative permeability, capillary pressure) are difficult to obtain on appropriate (large) scales. The problems of obtaining consistent *in situ* parameters for gas flow may be related to a more fundamental issue, namely, the applicability of continuum models when volume averaging is performed on a scale that may be much larger than the intrinsic scale of the flow phenomena. Gas migration in heterogeneous fractures and fracture networks is subject to hydrodynamic instabilities and may be affected by small-scale features, such as heterogeneities of individual fractures and fracture intersections. Gas flow may occur preferentially along localised pathways with dimensions of order dm - m in the horizontal direction. These cannot be practically characterised and resolved at field sites to make deterministic modelling of advective flows (gas displacing water) possible on a “performance assessment scale” (10-1 000 m or more). Field-scale modelling of gas flow would use space-discretized approaches (finite differences, finite elements) with typical resolution (grid block dimensions) of 10-100 m or more. It is not known whether a mechanistic description of flows in terms of driving forces and effective permeabilities, as implied by the generalised Darcy’s law, will be applicable when spatial features are volume-averaged on this scale.

The classical continuum models for two-phase flow have a firm basis for porous sedimentary rocks, but even in these relatively well-understood systems hydrodynamic instabilities from the lower

viscosity and density of gas as compared to water will give rise to complex non-linear phenomena of fingering and intermittent flow. Such phenomena are strongly affected by heterogeneities and small-scale variability, which limits our ability for quantitative deterministic modelling in the field. These issues require more research. Continuum models for multiphase flow may be more useful for demonstrating qualitative and conceptual understanding, and for bounding analysis and sensitivity studies, than as tools for detailed quantitative modelling. However, it may be possible to obtain a large-scale average description by “aggregating” results of high-resolution modelling for different stochastic realisations, rather than by using volume averaging. Detailed quantitative models of gas migration in fracture networks may not be needed for a safety case.

Bubble flow has been suggested as a possible mechanism for gas migration, but it is not clear whether this can be a significant process on a macroscopic scale. It may be possible for bubbles to become stabilised through naturally occurring surfactants. Bubble flow would entail much enhanced gas-water interface areas, and may cause enhanced transport of particles that attach to such interfaces (colloids, microbes, and others). The presence of gas is likely to increase the activity of microbial communities, by providing a source of energy and carbon, which can affect contaminant transport in numerous ways, e.g. by altering geochemical and sorption characteristics, and enhancing stability of gas bubbles. Diffusion of gaseous constituents and dissolved gases may also impact phase distributions and radionuclide migration. Partitioning of gaseous constituents between gas and aqueous phases is well understood, as is molecular diffusion in both phases. Robust and bounding estimates can be made for *in situ* diffusivities, so that this process can be readily modelled.

Gas pressurisation may be sufficient for gas and hydrofracing, as well as opening pre-existing fractures. However, the rate of pressure build-up from gas generation in nuclear waste repositories is likely to be very slow, and it is not known whether this may lead to responses other than fracing (elastic deformation, creep).

The impacts of gas flow may be enhanced by certain hydrogeologic conditions, such as the presence of faults, or by flow-funnelling structures such as fracture intersections and terminations, and permeability breaks in strata of low permeability. Identification and characterisation of such structures may be difficult in practice. Gas flowing along localised pathways may cause some mechanical erosion, and may alter the hydraulic properties of the rock mass.

#### **4. “BARRIER” ROCKS (Salt, Clay)**

Safety-relevant issues are generally similar to those discussed for fractured crystalline rocks, but differences in emphasis arise from the different hydrologic properties and behaviour (bound water, swelling, osmosis, creep). Groundwater flow may not be possible in salt and clay formations, either because there is no continuous aqueous phase (salt), or because water may be tightly held chemically (clay). Gas migration likewise may not be possible in undisturbed barrier rock formations, causing gas pressures to build-up until a gas frac occurs that could allow groundwater to access wastes and mobilise contaminants. Laboratory and field experiments have demonstrated self-healing mechanisms for such fracs by means of swelling from groundwater ingress (clay) and mechanical creep (clay, salt). Because the rate of pressure build-up from repository gas generation would be small, the formations may respond with mechanical deformation and creep rather than by fracing. These processes are complex, will involve a range of space and time scales, and will depend on site-specific conditions. Research using laboratory experiments, mathematical modelling, and observations of natural systems is ongoing. There is evidence that the unique barrier properties of pure salt and clay can become significantly changed when modest impurities of other rock forming minerals are present. The behaviour of clay and salt formations on a large scale may be more similar to that of crystalline rocks.

## 5. CONFIDENCE BUILDING

Field observations and experiments may be used for testing conceptual and quantitative models, and for building confidence in our understanding of processes and parameters. Gas migration in saturated heterogeneous formations is an unstable process that is subject to heterogeneities on multiple scales, so that the prospects for detailed quantitative predictions of gas migration pathways do not seem promising. It is possible, however, to design field experiments that would bring out an aggregated response of a rock mass on a larger scale, thereby improving the possibilities for meaningful characterisation, modelling, and prediction.

A possible test could involve excavation of a chamber of appropriate scale (100 m long, say) in fractured rock, which would be pressurised by gas injection while the leakoff rate into the formation would be monitored. Single-phase liquid pressure transient tests, either water injection or ventilation tests, would be used prior to gas injection to characterise formation permeability and porosity. In addition limited gas injection tests would be conducted from boreholes to measure threshold pressures. Two-phase flow modelling of gas leakoff from the chamber, using standard formulations for relative permeability and capillary pressure functions, should be capable of making reasonably approximate predictions.

Another test design that would probe gas effects relevant to nuclear waste disposal could use a “tracer doublet”: steady flow would be established between an injection and a production borehole completed in saturated conditions, and tracer pulse tests would be conducted in the established steady flow field. Subsequent to probing tracer migration under single-phase conditions, the flow would be disturbed by gas injection through a third borehole from below; changes in tracer breakthrough curves (mean travel time, hydrodynamic dispersion) would be monitored to reveal the impact of gas injection on groundwater flow.

These test designs are not intended as specific recommendations but are meant to illustrate possible approaches. They could be implemented in fractured crystalline rock, to obtain information about gas effects and build confidence in modelling capabilities in repository host rocks. Testing as well as modelling are likely to be simpler in sedimentary formations with intergranular porosity. Although this would not directly address issues of gas effects in fractured rocks, it could nonetheless help to build confidence in the ability of modelling approaches to describe field-scale processes.

Observations and analyses of natural systems (analogues) can play an important role in building understanding and confidence in process understanding and parameters. Initial and boundary conditions in such systems will typically have significant uncertainty, which may limit their utility in the context of quantitative mathematical modelling. However, they can provide rich qualitative information, and do so on large space and time scales that cannot be probed by controlled experimentation. In many cases it may be sufficient to document and evaluate existing knowledge to bring out the repository-relevant issues, rather than to conduct new research. For example, bedded salt formations in north and central Germany have been cut by basalt dikes in the geologic past, and have been subject to CO<sub>2</sub> injection from below. These phenomena have been well studied, and may serve as analogues for salt response to gas pressurisation from a repository.

Borehole-based studies in argillaceous formations at Mt. Terri, Switzerland, have investigated coupled hydro-mechanical processes during gas/water injection tests, in particular the generation of gas- and hydro-fracs, gas migration along such fracs, and self-healing of induced fractures.

*In situ* gas chemistry obtained through borehole sampling can provide information relevant to the flow of gases in the geosphere. The chemical and isotopic signatures of gas extracted from

individual lithologies (either host-rock or overburden) or from discrete intervals within a specific rock type may reveal important information regarding the circulatory properties and history of gas flow within a rock unit. Gas flow depends primarily on the characteristics of fracture networks because fractures are generally much more permeable to the gas phase than the matrix, especially in crystalline rocks. In contrast, rocks such as salt or clay may not contain permeable fractures. Construction of field-scale chemical profiles of long-term gas behaviour may help determine whether gas migration in a specific rock is expected to be a problem or not.

## 6. CONCLUSIONS

Generation of gas in subsurface disposal facilities that comprise a variety of engineered barriers and geologic media will give rise to coupled phenomena of flow, transport, chemical reactions, microbial interactions, and rock mechanical effects. The different processes will be played out in complex heterogeneous media over a broad range of space and time scales. There is a general sense in the technical community that impacts of gas generation on geosphere flow and transport are unlikely to be large. Yet, because of the large space and time scales involved, and the subtlety of many of the gas-related processes, it appears to be very difficult to achieve definitive and quantitative understanding. How should research into these phenomena be prioritised?

It would be easy to enumerate a range of studies that could be scientifically interesting and rewarding. However, the WG feels that priorities should be derived not from scientific curiosity but from the safety-relevance of different issues, as determined from the impacts of uncertain process elements in PA exercises. Confidence in mathematical modelling approaches could certainly be increased by additional research efforts. These could address the mathematical representation of process models, identification of model parameters and calibrations for specific sites, and empirical evidence that models can capture qualitative and/or quantitative aspects of flow and transport behaviour in two-phase gas-water systems. The introduction of different gases into saturated geologic formations may have significant impacts on microbial activity, which in turn may affect groundwater flow, solute transport, and geochemical conditions. These processes have attracted more detailed study only recently; their significance for repository safety is likely to depend on hydrogeologic conditions and disposal concepts at specific sites.

One particularly vexing problem has to do with the fact that gas effects do not seem to lend themselves easily to bounding or conservative analysis. Larger rates and amounts of gas do not necessarily imply stronger impacts on radionuclide containment; in fact, more gas could mean reduced groundwater access to the wastes, with reduced corrosion rates and more limited gas effects in the long term. In the geosphere, a stronger gas source may imply more stable gas pathways, with less impact on groundwater flow than a weaker gas source, for which gas pathways may be more dynamic. These non-linear effects are difficult to model, and generally appear to limit the utility of generic as opposed to site-specific models in addressing gas issues.

Regardless of (site-) specific priorities, it seems clear that careful observation and analysis of natural systems will have to play a significant role in developing and demonstrating an understanding of gas effects in the geosphere, and in increasing confidence in the ability of modelling approaches to capture the behaviour of natural systems.

**PART B**  
**WORKSHOP PROCEEDINGS**

**SESSION I**  
**PRESENTATION OF THE EC/NEA GAS STATUS REPORT**

Chairman: Juan-Luis Santiago (ENRESA, Spain)



# **Introduction to the EC/NEA Status Report: “Gas Migration and Two-phase Flow through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste”**

**William R. Rodwell**

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## **1. BACKGROUND**

It is recognised that in underground repositories for radioactive waste, significant quantities of gases may be generated as a result of several processes, notably the interaction of groundwaters and brines with waste and engineered materials placed in disposal systems. The gases may need to migrate through the engineered barrier system and the natural geological barrier. The potential impact of gas generation, accumulation and migration on the long-term safety of a repository will be dependent upon the waste types, the repository concept, the host geological environment and the scenarios for the long-term evolution of the system. It is therefore recommended that the potential impact of gas accumulation and migration on the performance of the various barriers should be addressed and assessed in the development of safety cases for radioactive waste repositories.

To address this need in the safety assessment of repositories, significant effort has been expended in recent years in numerous national and international programmes in attempts to understand the potential impact of gas migration, and of other two-phase gas-water processes on the performance of underground radioactive waste repositories, and to provide modelling tools or approaches that will allow these impacts to be assessed. These efforts have included the work of several projects carried out in the framework of the European Commission’s Nuclear Fission Safety programme (the PEGASUS project), and have resulted in a significant increase in understanding of the relevant processes and to an important collection of experimental data.

In the light of these efforts, it was, in 1997, considered timely and of potentially widespread benefit to organisations involved in the deep disposal of radioactive waste to undertake a review of the knowledge so far gained of gas migration and two-phase flow processes relevant to repositories and to establish the current status of understanding of these topics. Accordingly, the NEA and the European Commission jointly initiated the work necessary to produce the required review, and this led to the publication in late 1999 of a status report on “Gas Migration and Two-phase Flow through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste” [1]. Funding for the preparation of the report was provided jointly by the European Commission and a consortium of national organisations active in the field of radioactive waste management and represented within the NEA Co-ordinating Group for Site Evaluation and Design of Experiments for Radioactive Waste Disposal [2].

The following paragraphs provide a short summary of the contents of the report; further details of the topics addressed are presented in companion papers. It is hoped that the report will serve

as a useful foundation for the deliberations of this NEA/EC/ANDRA “Gas” Workshop, at least as far as gas migration and two-phase flow issues are concerned. As will be seen below, the report does include a review of gas generation, since gas migration from repositories cannot be adequately addressed without an appreciation of the source of the gas, but the primary focus of the report is on flow processes involving gas.

## **2. CONTENTS OF THE STATUS REPORT**

The status report addresses the following topics:

The safety issues potentially associated with gas in repositories. These include overpressurisation and its consequences, the release of radioactive and flammable gases at the surface, effects on the movement of contaminated groundwater, the transport of attached particles at interfaces, and issues particular to an unsaturated site.

An examination of existing disposal concepts, and the development of a set of representative repository concepts (see Section 3) to be used in subsequent discussions of gas migration. These repository concepts were intended to embrace those appropriate for different waste types and to cover the range of host rocks being considered for repository sites. The latter were categorised as water-saturated low-permeability fractured rock, unsaturated fractured rock, plastic clay, indurated mudrocks and rock salt formations. Unsaturated fractured rock referred specifically to Yucca Mountain as the only site of this type currently envisaged.

Gas generation in repositories. While flow processes involving gas were the central issue of the report, it was also important to understand the gas source term for repositories in saturated sites, and the dependence of this source term on both the waste and the site characteristics, as this affects the nature of the safety issues that might arise.

Fundamental concepts in gas migration and two-phase flow. The basic physical processes and modelling assumptions in two-phase porous-medium flow are summarised to provide the context for the more detailed discussion given in relation to engineered barriers and the different rock types considered.

Gas migration and two-phase flow in engineered barrier systems. The barriers discussed divide into bentonite or sand-bentonite buffers and cementitious materials.

The mechanisms of gas migration and two-phase flow through the geosphere. These are discussed in detail in relation to the available experimental evidence and uncertainties for the defined categories of host rock.

Modelling of gas migration and two-phase flows. Approaches that have been developed for and issues that arise in modelling gas migration are again discussed for the different host rock categories that have been identified.

Potential impacts on the performance of repository systems. The treatments that have been accorded to gas in previous performance assessments are reviewed, and the potential impacts of gas on the performance of the reference repository concepts that have been defined are discussed.

### 3. REPOSITORY CONCEPTS

Table 1 lists the set of reference repository concepts [see point (b) in Section 2] used in the Status Report to provide a framework for discussing gas migration issues.

Table 1. **Representative repository concepts**

<b>Host rock</b>	<b>waste type</b>	<b>container material</b>	<b>backfill material</b>	<b>sealing material</b>
fractured rock	L/ILW	carbon steel concrete	cementitious grout	bentonite sand
fractured rock	spent fuel	stainless steel copper	bentonite	unspecified
plastic clay/ mudrock	L/ILW	carbon steel	cementitious grout	unspecified
plastic clay/ mudrock	spent fuel/ HLW	stainless steel	bentonite	unspecified
salt	spent fuel	stainless steel	crushed salt	bentonite/ bitumen
salt	vitriified HLW	stainless steel	crushed salt	bentonite/ bitumen
fractured tuff*	spent fuel	stainless steel	none	unspecified

\* Tuff refers here to unsaturated, fractured rock as exemplified by the tuff host rock at Yucca Mountain.

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The following organisations, in addition to the European Commission, contributed to the funding of the preparation of reference [1]: ANDRA, France (National Radioactive Waste Management Agency); ENRESA, Spain (National Agency for Radioactive Waste); IPSN, France (Nuclear Protection and Safety Institute); ONDRAF/NIRAS, Belgium (National Organisation for Radioactive Waste and Fissile Materials); Ontario Power Generation, Canada; NAGRA, Switzerland (National Co-operative for the Disposal of Radioactive Waste); NIREX, United Kingdom; SKB (Swedish Nuclear Fuel and Waste Management Company); SKI, (Swedish Nuclear Power Inspectorate); United Kingdom Environment Agency; and USDOE/YMP (United States Department of Energy, Yucca Mountain Project). Furthermore, GRS, Germany (Company for Reactor Safety) and SCK/CEN, Belgium (Nuclear Energy Research Centre) contributed to the status report by providing a thorough review of the draft versions.



# Gas Generation

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## 1. INTRODUCTION

The generation of gases in wastes is a widespread and well-known phenomenon for conventional, as well as radioactive, wastes. At conventional landfills, the gases, if formed in sufficient amounts, may even be collected and used as an energy source. Gas generation in radioactive wastes under final disposal conditions is unavoidable and stems from the nature of certain major components of the wastes. Since the amount and types of gases have been identified as potential safety concerns for a repository in deep geologic formations (e.g. explosive and toxicity hazards and interactions with movement of groundwater), major efforts have been made to characterise the gas generating processes and their generation potential. The state of the art reached has been compiled in a recent status report [1]. This paper summarises the results. Therefore, evaluated literature sources are not repeated here, but are extensively documented in the status report.

## 2. BOUNDARY CONDITIONS

Because a higher gas generation or generation rate not always means higher consequences determination of the gas generation can not be restricted to the assessment of upper bounds but has to provide a realistic source term as far as possible. This means that initial and boundary conditions for gas generation have to be carefully characterised, variations in time and space have to be considered and interactions with other phenomena and processes have to be taken into account.

Except for a short initial period gas generation in a repository takes place in anaerobic conditions. The equilibrium rock temperature depends on the depth of the repository and may vary between about 10 and 50°C. In addition, the waste itself may generate heat. The resulting temperature can be controlled by the waste composition and the interim storage time.

The selected host rock has a major influence on the gas generation, too. This is due to differences in the available humidity, the mineralisation and composition of the groundwater, the geochemical setting (pH, Eh) and the possible or required backfill material. Table 1 gives an overview on some of these parameters for different host rocks.

Table 1. **Composition of groundwaters in different host rocks (in ppm)**

Formation	Hard rock		Clay	Salt		Tuff
	granitic	Oolitic (Konrad)		rock salt	Q brine	
origin			Boom clay			Yucca Mountain
Na <sup>+</sup>	90-4800	62000	330	40000-150000	7000	44
K <sup>+</sup>	0.2-90	240	13	1500-35000	32000	5.1
Ca <sup>++</sup>	30-3300	11300	<5	600-1400		12.5
Mg <sup>++</sup>	0.05-50	2300	3	340-35000	90000	1.9
Cl <sup>-</sup>	180-13000	117000	27	190000-250000	270000	6.9
SO <sub>4</sub> <sup>++</sup>	0.14-1800	600	400	3500-5000	15000	18.7
HCO <sub>3</sub> <sup>-</sup>	<300	60	850	<700	-	125
pH	8-10	6-7.2	9	6-7.4	5	7.6

### 3. MECHANISMS

A number of mechanisms have been identified which may contribute in different degrees to gas generation in repositories. The main mechanisms by which gas could be generated in deep repositories are corrosion of metals in wastes and packaging, giving hydrogen; radiolysis of water and certain organic materials in the packages, yielding mainly hydrogen; and microbial degradation of organic waste components, which gives methane and carbon dioxide as the main products. These processes are reviewed in the following subsections.

#### 3.1 Corrosion

Gas generation by corrosion is a widely investigated mechanism. Since iron is the dominating metallic component in the waste, in the waste packaging and eventually in the structural materials of a repository experimental work has concentrated on corrosion of iron and its alloys. In addition, some measurements on materials like titanium, copper, aluminium, zirconium, lead and their alloys are available.

Corrosion is an electrochemical process by which a metal is transformed into a higher oxidation state, i.e. the metal releases electrons. Therefore an electron acceptor is needed as corroding agent. Under repository conditions mainly oxygen and water are considered for this. The anodic and cathodic half-reactions for these processes are:



Equation (1) and (2) prevail under aerobic conditions, while equation (1) and (3) describe the corrosion process under anaerobic conditions. Obviously, the dominating process under aerobic conditions does not produce gas (hydrogen). Thus hydrogen generation under aerobic conditions is not excluded, but largely reduced by the competing thermodynamically favoured reaction.

Corrosion may further depend on a number of physical and chemical parameters, some of which have been studied for their effect on gas generation extensively. Examples for such *physical*

parameters are temperature, pressure and radiation. The primary *chemical* factor is the waste itself. In addition, the geochemical conditions determined by the groundwater composition and the host rock material have to be considered. Investigations normally use groundwater samples or comparable synthetic solutions to cope with the latter parameter. Further chemical factors that have been checked in experiments have been pH value, Eh value, mineralisation of groundwater (salt content) and impurities contained in the groundwater.

Table 2 summarises some characteristic corrosion rates in different host rock environments. Tuff has been omitted in this compilation, because conditions may remain aerobic for the Yucca Mountain repository concept. Thus gas generation can not be assessed from the measured corrosion rates under these conditions. The values are meant as indicators for the range of corrosion rates that might be expected. For a detailed assessment the individual site and concept specific conditions have to be regarded, which can lead to substantial changes in these rates.

Table 2. **Typical corrosion rates in different geological environments (in  $\mu\text{m/a}$ )**

Material	Hard rock	Clay	Salt
<b>Iron</b>			
low alloy steel			
• ca. 25	1	2	0
• ca. 50	0-6.5	–	0.1
• ca. 90	1-2.5	5-8	0.2-2
• ca. 150	–	9	0.1-0.5
cast iron/steel			
• ca. 25	–	0	0
• ca. 50	–		0
• ca. 90	0-5		0-5
• ca. 150	1.7-13		25
copper/Cu alloys	0	0	–
titanium	0-0.15	0	0-0.5

The determination of a gas source term from metal corrosion can today be performed straightforward, if the waste constituents, the metallic surfaces and the chemical conditions in the waste are well known. This mainly applies to heat generating wastes, like vitrified HLW and spent fuel. In case of low-level mixed waste, this knowledge can be achieved only by very rough estimates. Therefore, alternative methods for the determination of gas generation have to be considered for this waste, if realistic estimates are required.

### 3.2 Radiolysis

For the decomposition of chemical compounds by radiation internal and external radiolysis have to be distinguished. Internal radiolysis means processes taking place in the waste product and its packaging. External radiolysis concerns the backfill material surrounding the waste and possibly the host rock. Inside the waste packages all kinds of radiation ( $\alpha$ ,  $\beta$ ,  $\gamma$ , n) may contribute to gas generation by radiolysis. In the backfill, due to the shielding properties of the packaging,  $\gamma$ -radiation dominates.

Gas generation by radiolysis implies that gases are produced as primary or secondary decomposition products of the radiolytic processes. The leading example for this kind of reaction is the decomposition of water ( $\text{H}_2\text{O}$ ) resulting under idealised conditions in the formation of stoichiometric amounts of hydrogen and oxygen gas. Since oxygen is generated in atomic form, it

normally reacts immediately with its environment. Therefore, gas generation by radiolysis of water is commonly characterised by the amount of hydrogen produced per unit mass of substrate and applied radiation dose. This model implies a linear relationship between gas generation and the applied dose, which is expressed by a constant, the so-called G value:

$$B = G \cdot D \cdot m$$

where

- B = generated gas volume
- D = radiation dose
- m = mass of irradiated material.

G is specific for the generated gas – for example G(H<sub>2</sub>) - and the irradiated material.

Internal radiolysis mainly appears with low and intermediate level waste and may concern the waste, the matrix and eventually the container (if e.g. made from concrete). Typical G(H<sub>2</sub>) values for different waste products can be found in [1,2]. Because of the low activity concentration radiolysis in general plays a secondary role for gas generation in these wastes. It is only for some cemented heat producing wastes (fuel element hardware, dissolver sludge from reprocessing) that the contribution of internal radiolysis to the total gas generation may be relevant.

In contrast, for HLW gas generation by radiolysis happens mainly in the surrounding backfill or host rock material, since the waste contains nearly no material that may be decomposed by radiation. If thin-walled containers are used this contribution may even become dominating for gas generation. Otherwise the shielding effect of the container walls prevents a substantial gas generation. This situation may, however, change in the long term if container degradation by corrosion allows for direct contact between groundwater and the waste. Table 3 gives some characteristic G(H<sub>2</sub>) values for different host rock and backfill materials.

Table 3. G(H<sub>2</sub>) values [μl/kgGy] in host rock and backfill materials

Material	G(H <sub>2</sub> )
salt	0.002
granite	0.005 <sup>1)</sup>
clay	0.05 – 0.26
bentonite (α radiation)	2.3
concrete	0.14 – 0.7
<sup>1)</sup> assuming 0.2 % water content	

### 3.3 Microbial degradation

Since a multitude of micro-organisms exists with different metabolisms, different habitats and different assimilation capabilities, gas generation by microbial degradation is a very heterogeneous phenomenon with a broad range of possible generation rates and time-dependent development. Since the types and amounts of micro-organisms that contribute to gas generation cannot be exactly determined or effectively influenced, starting and boundary conditions for microbial gas generation are vague. Generation rates may be dependent on environmental factors, which are not easy to assess over thousands of years. Their assimilation behaviour helps micro-organisms to overcome unfavourable living conditions, which may alter their gas-generating potential, even in the case of a

constant environmental situation. Nonetheless, there exist some prerequisites for microbial gas generation from which estimates and limiting factors for the gas generation potential can be derived.

An essential prerequisite for microbial activity and thus microbial gas generation is the availability of water. In fractured rock and mudrock this is provided by the water in the geological formation. But even in a rather dry host rock, water is present in the residual humidity of the waste, the backfill material and the repository atmosphere. In the latter case it may be prudent to monitor the water balance, since water-consuming processes like corrosion, radiolysis, hydration and hydrolysis may reduce the available amount of water.

A second condition for microbial activity is the existence of nutrients. This concerns on one hand the availability of organic material containing the carbon that forms the basis of any metabolism. This restricts the microbial gas generation mainly to LLW. Easily degradable and/or soluble substrates like cellulose, acetate or glucose markedly accelerate gas generation. Because of the multitude of metabolisms observed for microbes, a range of other materials may serve as nutrients in addition. Inorganic salts like nitrate or sulphate may enhance microbial activity, as may metals or trace elements. However, even in the apparent absence of these nutrients, microbial gas generation is possible to a limited extent, if the natural environment is not sterile but contains traces of organic materials and nutrients. Further environmental factors of more or less importance are the pH value, temperature, oxygen supply, salinity or impurities in water, radiation and pressure.

Measurements of microbial gas generation are rare. Earlier measurements have been compiled in [2]. The most comprehensive study up to now has recently been performed by *Francis et al.* [3] for a repository in salt (WIPP). Although the study concentrates on saline conditions and the formation of carbon dioxide, some more general conclusions can be drawn from the results. All measurements so far demonstrate a rather swift saturation behaviour of the CO<sub>2</sub> production, which is followed by lower generation rates, with mostly CH<sub>4</sub> and CO<sub>2</sub> as the major gas components. For long-term safety considerations, methane is of particular interest, since its production dominates microbial gas production at relatively long times. These observations are consistent with experience from conventional landfills.

At present, it remains a problematic task to translate observations into gas generation rates in real waste under repository conditions. Measurements on real waste (cf. section 4) may provide some answers. Because of the flexibility of the microbes and their assimilation behaviour, long-term predictions remain difficult. Preferentially assessment of long-term cumulative gas generation therefore should rely on limiting factors or, if appropriate, on natural observations on long-lived reservoirs of organic materials.

### **3.4 Others**

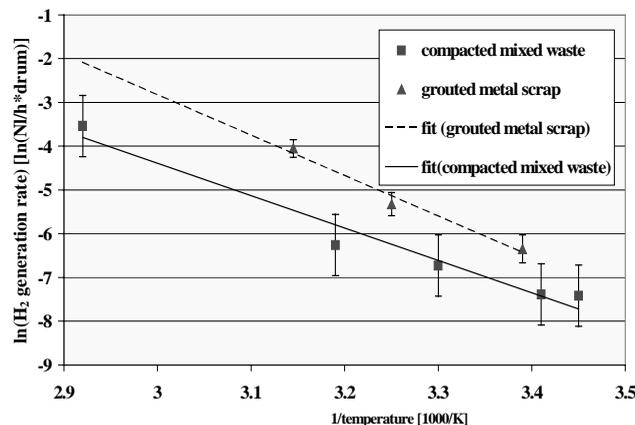
Other possible mechanisms contributing to gas generation may be gas release from the host rock, radioactive decay or other chemical reactions than corrosion. From these only the latter has been identified as a possibly relevant gas generation mechanism. It is the hydrolysis of aluminium present to a certain extent in LLW that may generate larger amounts of gases, partially at high rates. Therefore, it is recommendable to characterise the Al content of the waste carefully to assess the importance of this contribution.

## Measurements on real wastes

So far, characterisation of gas generation has focused mainly on detailed investigations of certain gas generation mechanisms in laboratories and measurements using individual waste containers (e.g. drums) with more or less well-characterised contents. The physico-chemical environment for gas generation in a container in a repository is in general only to a limited extent comparable with laboratory conditions. Furthermore, the limited knowledge about the governing conditions, e.g. waste constituents contributing to or influencing gas generation, hamper transfer of laboratory measurements to real waste. Therefore, it has been considered helpful, especially for low-level waste, to complete existing knowledge about individual effects by measurements on real waste. The experience gained from these measurements can be used to create a validated link between laboratory data and real waste situations, which aids in a prognostic characterisation of gas generation for the post-operational phase of a repository.

First results of a comprehensive evaluation of measurement data from low-level waste have been reported in [4]. Measured gas generation rates were differentiated for the type of waste, waste age and temperature. Figure 1 demonstrates the temperature dependence derived from routine as well as deliberate measurements for two types of waste. Gas generation follows an Arrhenius type temperature function, which is waste type specific.

Figure 1. **Temperature dependence of gas generation in low-level waste types**



## Modelling

A number of codes are available which are intended to model gas generation in various environments for different types of wastes. In [1] the following codes are compared:

### GABI

GABI has been developed for all types of waste. The code is based on the German repository concept in salt with crushed salt backfill. Balances are kept for the corroding agent (brine) and the corroding material. Time dependent gas generation rates are calculated with interactive selection of starting and boundary conditions.

## GAMMON

GAMMON treats gas generation in low- and intermediate-level wastes by corrosion and microbial activity. Other mechanisms were assessed to be negligible for the types of waste considered. Special efforts were made over the modelling of the time dependent change from aerobic to anaerobic conditions so as to understand the pattern of gas generation in the operational period of a repository as well as in the post-closure phase.

## GASFORM

GASFORM deals with high-level waste in granitic host rock. Sand or bentonite is considered as backfill material. GASFORM was intended to be coupled with a gas transport code such as TOUGH2. Together with the GABI code, benchmark calculations were performed for code validation.

## GETAR

The code GETAR consists of several modules developed for the description of  $^{14}\text{C}$  release from a near-surface waste repository to the environment. One of these modules deals with gas generation. It models gas generation from low-level waste mainly by microbial activity and hydrolytic reactions. Corrosion is considered only for its effect on oxygen consumption.

## RADCALC

RADCALC was developed to assess the gas generation by radiolysis in certain types of waste during transport. The main objective was to demonstrate compliance with the requirement to limit pressure build-up in waste containers and to avoid explosive gas mixtures during transport.

## Brush *et al.*

developed a kinetic reaction path model based on thermodynamic equilibria. The model is designed for transuranic waste under repository conditions at the WIPP site. Reaction paths can be influenced by a pre-set of reaction products. Gas generation rates for the thermodynamic equilibrium established by the calculations are taken from experiments.

To describe the details of these codes would be beyond the scope of this presentation. The codes are all built to purpose and qualified to a certain extent.

## Conclusion

In summary, it can be concluded that modelling of gas generation is most advanced for corrosion, although the difficulty remains that the metallic contents of the waste and the available corroding media have to be characterised in some detail. Radiolysis can be predicted with well established models. The accuracy is dependent on the available characterisation of the activity contents in the waste package and the degree of detail implemented in the model. The modelling of microbial gas generation is restricted to well defined scenarios, which are not necessarily comparable with the long-term situation in an underground repository. For an extrapolation beyond their validation range an increased knowledge about the processes, their dominant parameters and their limiting conditions are essential prerequisites. Interactions have to be considered in modelling between the gas generation mechanisms (e.g. by water balance in case of limited supply) as well as with other processes like fluid flow (water saturation), geochemical history and geomechanical forces (vs. pressure build-up).

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# **Gas Migration through Bentonitic Engineered Barrier Systems and through Non-indurated Clays**

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## **INTRODUCTION**

This extended abstract summarises briefly the main conclusions and perspectives of the research work carried out in the frame of gas migration from a radioactive waste disposal through bentonitic engineered barrier systems (EBS) and non-indurated natural clay formations. After a description of the most important experimental results and the conceptual model evolution, we will focus on the safety relevant issues and the way the gas migration through such media is currently treated in performance assessment for different types of waste. Finally, the remaining open questions will be addressed at the end of this paper. Further insights are provided in the EC/NEA status report [Rodwell *et al.*, 1999].

### **1. STATUS OF KNOWLEDGE CONCERNING EXPERIMENTAL AND MODELLING ISSUES**

Many experimental studies have shown that the gas migration phenomena in saturated bentonitic EBS and in non-indurated natural clay formations are similar. Passage of a gas phase is only possible if the gas pressure exceeds some threshold value. The following subsections will describe the respective behaviour of such materials when they are exposed to a gas pressure build-up generated by the radioactive waste.

#### **1.1 Gas migration in bentonitic engineered barrier system**

Materials based on clay have been proposed as components of repository EBS by numerous organisations and are usually called buffer materials. The particular clay materials used are derived from bentonite deposits. Bentonite does not fit the picture of a classic porous medium. Montmorillonite, the principal constituent of bentonite, exists as a stack of extremely thin and flexible sheets separated by narrow spaces containing interlayer cations and water molecules. The aperture of the interlayer spaces varies from 1 to 1.9 nm, depending on the hydration state of the clay. The swelling process is a direct consequence of the movement of water molecules into interlayer positions. Calculations of the average interstack water film thickness suggest an average film thickness in the range of 3 to 6 nm.

The two-phase flow theory, where the retention function (matric suction against water content) is interpreted in terms of the capillary pressure and the pore size distribution of the medium, is inappropriate for gas migration into saturated buffer bentonite. The reason is that a very large proportion of the total water content is adsorbed to mineral surfaces. Results of gas migration experiments carried out on different types of bentonite (Kunigel VI, Fo-Ca clay, Avonlea bentonite and MX-80) are available. The controlled flow rate experiments carried out by *Horseman et al.* [1999] on initially fully saturated MX-80 bentonite showed a peak in gas pressure followed by a spontaneous negative transient. The peak is similar to that observed during hydrofracturing, suggesting that gas moves as a discrete phase through a network of pathways formed by the rupture of the clay fabric under high-applied gas pressures. This hypothesis is supported by more recent work carried out on Fo-Ca clay [*Dereeper and Volckaert*, to be published]. The difference at breakthrough between the gas pressure and the original pore water pressure is more or less equivalent to the swelling pressure of the bentonite, which is a function of its dry density.

Recent model developments include those described by *Nash et al.* [1998] and *Wood* [1998]. The first describes the gas invasion of the clay as a microfracturing process, with the dimensions of the pathways formed allowed to adjust in response to changes in gas pressure. The second follows a capillary bundle approach, with stochastically generated variable aperture capillaries, whose radii can also vary with gas pressure. In this case, gas invasion is modelled as a water displacement process.

The addition of sand to the bentonite buffer material can drastically change its gas migration behaviour [*Rodwell et al.*, 1997; *Dereeper and Volckaert*, to be published]. This makes sense if we consider that the structural charge of the sand particles is neutral and therefore decreases the cohesion forces at macro- and microscopic scale, which makes interstitial water more mobile. No model testing has been carried out with these experimental data.

## 1.2 Gas migration in non-indurated natural clay formations

In the PAGIS report [*Marivoet and Bonne*, 1988], several non-indurated clay formations are mentioned as potential hosts for radioactive waste disposal. The most relevant data sets on gas migration have been acquired on the Boom Clay formation, since it is studied as a potential geological host formation for high- (HLW) and intermediate-level (ILW) radioactive waste storage in Belgium [*Volckaert et al.*, 1995; *Ortiz et al.*, 1997; *Rodwell et al.*, 1999 and *Rodwell*, 2000].

Two gas transport mechanisms have been identified, depending on the gas pressure level. Below a value referred as gas entry pressure, the predominant transport mechanism is the Fickian diffusion in groundwater. Once this gas entry pressure is exceeded, gas start to displace the interstitial pore water or push back the clay particles, or both, to create gas-occupied pathways in the clay. As we can see, we can make a parallel between this description and the gas migration behaviour in bentonitic engineered barrier system. The only difference might be the proportion of mobile water, which can vary according to the clay composition (smectite, illite, kaolinite, quartz, ...): 2 to 5% for Boom Clay and less than 1% for pure bentonite. As in pure bentonite, it appeared that the gas migration properties of Boom Clay might be correlated with the local geomechanical stress conditions. It has been shown that an engineering disturbed zone (EDZ) might form a preferential pathway for gas migration owing to the lower local effective stress level. Also, the influence of the state of stress and of stress history (overconsolidation ratio) on gas transport parameters in Pontida Clay, a non-indurated reconstituted clay, is demonstrated in [*Rodwell*, 2000].

The “self-healing” capacity of such created gas-pathway has been tested for the Boom Clay formation [*Rodwell*, 2000]. A gas pathway was maintained open during one year between two screens located on a multipiezometer that has been installed in the Boom Clay formation. Forty days after

stopping the gas injection, a tritiated water loop was put in contact with the former gas injection screen. The modelled evolution of the activity concentration in the injection and neighbouring screens, considering pre-determined transport properties of undisturbed clay, corresponded well with the experimental measurements. This indicates a complete self-healing of the previously created gas pathway with respect to radionuclide migration.

There has been an evolution in the models elaborated to simulate gas migration behaviours in non-indurated clays. The first approach was to use a simple two-phase flow model based on an extended form of Darcy's law, taking the a linear variation of the rock porosity and permeability with the gas injection pressure. Since accumulated experimental results provided evidence that gas migrates by the formation of preferential pathways instead of by the advancement of a de-saturation front, alternative models have been developed. At first, a capillary bundle model with non-intersecting capillaries of fixed length and uniform radii was built. A first modification was brought to that model, including connections between capillaries, and later further modifications allowed the pore radii to be functions of gas pressure, time, and even pore length for non-intersecting capillaries. The last step in the model development was to take the influence of the geomechanical factors into account in order to build a gas front propagation model and to check the influence of soil heterogeneity on gas transport.

## **2. TREATMENT IN PA ANALYSES AND SAFETY RELEVANT ISSUES**

This section will first focus on the state of the art on treatment in PA analyses in bentonitic EBS (near field). The treatment of gas generation and migration in PA analyses for non-indurated clay are more thoroughly described in [Volckaert and Mallants, 2000]. Then, the safety relevant issues for non-indurated clay formations (far field) will be described.

### **2.1 Current PA treatment in bentonitic EBS**

The effects that gas generation might have on the integrity of this type of near-field will mostly be of a physical nature. The potential physical perturbations are the disturbance of the hydrogeological regime around the repository, the creation of pathways that might facilitate the transport of radionuclides and a faster transport of contaminants in the interstitial water expelled by the gas phase. No specific treatment of the bentonitic barriers towards the gas migration issue has been used until now, since the domain covered by the PA analyses corresponds usually to a regional scale. However, several post-closure safety analyses of repository sites using bentonite as a buffer material or as a barrier and involving the problem of gas generation and migration on the near-field issues are available for crystalline rock repositories. These are:

- The Forsmark repository for low- and intermediate level waste.
- The Posiva spent fuel repository (TILA-99 safety assessment).
- The Nagra's HLW repository (Kristallin-I project).
- The AECL's concept for disposal of spent fuel.

We must be aware that the PA treatment for the bentonitic EBS largely depends on the concept that will be chosen. First, we could try to prevent any gas release by using pure saturated bentonite material as EBS. This possibility is difficult to evaluate right now in PA analyses, since both the pressure threshold of gas pathway formation and the spatial distribution of pathways cannot be predicted with the required accuracy. The other alternative is that we could decide to ease the release of the gas produced by using mixtures of sand and bentonite as EBS, or even by installing a supplementary material layer between the bentonite-based EBS and the waste canister that would act as a gas vent while preventing any preferential radionuclide release. Such a material is currently under development in some national waste management programmes.

## 2.2 Safety relevant issues for the non-indurated clay host rock reference case

The comments of this subsection principally refer to disposal repository concepts specific to the Boom Clay formation. First, a distinction must be made between two types of radioactive waste concepts: the spent fuel and HLW repository concepts on the one hand, and the ILW repository concept on the other hand.

The issues to be considered for a spent fuel or HLW repository concept should be the following:

- The gas generation source term, considering the corrosion rates and the possible transformation of the produced gas (e.g. by present bacteria, if relevant in the considered repository conditions).
- The temperature effects, considering that HLW and spent fuel are heat-emitting waste, and that the temperature present at the waste canister interface might be larger than 100°C. This level of temperature might be responsible for the formation of vapour and for the modification of the clay-based buffer material properties, which both could affect the concept safety.
- The radiolysis.

For the ILW repository, the issues to be considered are:

- The gas generation source term, that would depend on the waste inventory (amount of carbon steel to be corroded) and on the waste matrix (concrete or bitumen).
- The microbial activity, bound with the near-field conditions (presence of concrete) and the radioactivity level.
- The radiolysis.

## 3. REMAINING OPEN ISSUES AND RECOMMENDATIONS

There is evidently a need for advances in the conceptual understanding of the mechanisms of gas migration associated with bentonitic EBS and non-indurated clay formations. This may include the need to understand clay-specific coupled processes involving thermal, hydraulic, mechanical and chemical processes.

From the modelling point of view, complex dependencies arise from the strong interactions between water and the solid particles making up the clay fabric. Gas entry into the clay, which requires a threshold pressure to be exceeded, appears to require some, probably microscopic, deformation of the clay fabric and not just the overcoming of a conventional capillary pressure. The current models should be regarded as tools for interpreting gas migration experiments and for elucidating gas migration mechanisms. The development of suitable macroscale models from these prototypes, when they have been suitably validated, is an issue for future work.

Gas generation from repositories located in very low permeability non-indurated clay formations are likely to encounter problems of gas pressure build-up and possible fracturing in the case of waste producing significant quantities of gas. Further research and development focused on gas should be devoted to this problem.

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## Gas Migration through Indurated Clays (Mudrocks)

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The main features that distinguish the gas transport properties of mudrocks from those of all other rock-types are the sub-microscopic dimensions of the interparticle spaces, the very large specific surface of the mineral phases, the strong physico-chemical interactions between water molecules and surfaces, the exceptionally low permeability and the very pronounced coupling between the hydraulic and mechanical responses.

Gas migration by the diffusion of gas molecules in aqueous solution is a slow background process in all mudrock formations [Krooss, 1989]. The diffusion coefficient and the solubility coefficient are two important parameters in the quantitative treatment of diffusion. The diffusion coefficient is essentially a property of the water-saturated rock. If the concentration of gas in water at the source remains constant with time, the magnitude of the steady-state diffusive flux away from the source is regulated solely by the rock. The steady-state flux can never exceed the upper bound set by the diffusion coefficient and solubility in water.

In many practical situations, the actual gas flux through the mudrock formation is determined by the rate of production of the gas and not by the rate at which a particular transport mechanism can accommodate the flux. We can think of the gas flux as being imposed on the formation, rather than being determined by it. When the imposed flux exceeds the upper bound for diffusion, the gas must be transported through the rock as a separate gas phase [Rodwell *et al.*, 1999].

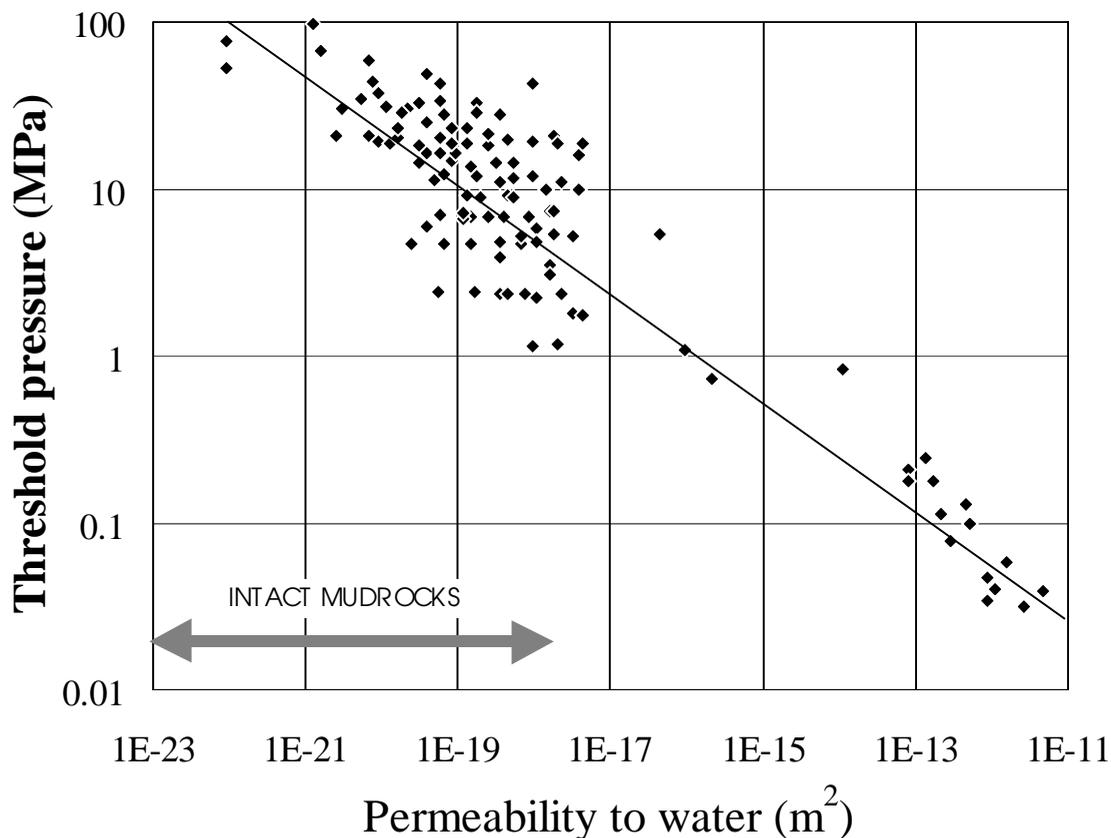
In intact mudrocks it is expected that groundwater flow velocities will be too low for advection of gas dissolved in water to contribute significantly to gas transport.

The maximum pressure of the gas at the source (i.e. in the repository near-field) is capped by the state of stress acting in the rock. The stress at any point in the subsurface can be quantified in terms of three mutually perpendicular principal stresses [Amadei and Stephansson, 1997]. When the rock mass has negligible tensile strength, the sensible upper limit for the gas pressure at the source is set by the minor principal stress. In a gradually subsiding sedimentary basin, the minor principal stress usually acts in a horizontal direction. The orientation of minor stress in a tectonically active region is less easily predicted. If the gas pressure at the source approaches the minor principal stress, gas will usually migrate along preferential pathways. In many cases, the incipient pathways of gas movement are present naturally as dilatant microcracks, crack networks, fissures and faults [Rodwell *et al.*, 1999].

Experience of gas movement in other porous materials suggests that there should be another mechanism of gas migration in mudrocks. This involves the displacement of water from the pore space, desaturation and the development of an interconnected network of gas-filled pore channels.

These are the processes conventionally considered in the mathematical theory of two-phase flow [Aziz and Settari, 1979].

Figure 1. Plot of threshold capillary displacement pressure against intrinsic permeability for a wide range of undifferentiated lithologies including shale, limestone and anhydrite, based on *in situ* measurements [from Ibrahim *et al.*, 1970, Ingram *et al.*, 1997].



There are some very real doubts concerning the applicability of standard two-phase flow theory to mudrocks [Tissot and Pelet, 1971; Hedberg, 1974; Harrington and Horseman, 1999; Rodwell *et al.*, 1999]. In order for gas to penetrate the original pores of a water-saturated mudrock, the gas pressure must be large enough to overcome the restrictions imposed by capillarity. Compiled oil industry data on the threshold capillary displacement pressure of low permeability rocks (see Figure 1) suggest that the required gas pressure is often so large that it can approach the lithostatic stress [Ibrahim *et al.*, 1970; Ingram *et al.*, 1997; Rodwell *et al.*, 1999]. Furthermore, most of the porewater present in a clay-rich mudrock can be shown to be present as very thin interparticle films. Water in thin films has properties that can be quite different from those of bulk water [Clifford, 1975]. Water molecules can be strongly adsorbed to mineral surfaces [Newman, 1987]. Thin films of water are also capable of supporting the full lithostatic stress. It seems unlikely that water molecules can be desorbed from the thin films by a mobile gas phase at a pressure significantly less than total stress. If gas cannot penetrate the original pores of a mudrock and cannot displace water from these pores, then it is

impossible for the clay matrix to become desaturated [Harrington and Horseman, 1999]. Hinch [1978] makes the observation “Porewater and shale grains should not be considered as a two-phase system, as in the conventional view of water in sandstone. They must be considered as interactive through adsorption, that is, as a single shale-water system in which the behaviour of porewater, oil and gas are essentially controlled by intermolecular forces.”

Hedberg [1974] quotes a passage from Tissot and Pelet [1971] which states: “The displacement of an oil or gas phase from the centre of a finely-divided argillaceous matrix goes against the laws of capillarity and is in principle impossible. This barrier can however be broken in one way. The pressure within the fluids formed in the pores of the source-rock increases constantly as the products of the evolution of the kerogen are formed. If this pressure comes to exceed the mechanical resistance of the rock, microfissures will be produced which are many orders of size greater than the natural (pore) channels of the rock, and will permit the escape of an oil or gas phase, until the pressure has fallen below the threshold which allows the fissures to be filled and a new cycle commences.”

The processes of uplift, erosion and exhumation of a mudrock lead to overconsolidation and associated stress-relief. These are important considerations when examining the probable routes of gas migration through a mudrock [Rodwell *et al.*, 1999]. All mudrocks currently proposed as host rocks for radioactive waste repositories are overconsolidated, in so far as the current stress is less than the maximum burial stress. Since the threshold capillary displacement pressure of two-phase flow theory (see Figure 1) is determined by the size of the pore channels, we might suppose that its value becomes established at the time of maximum burial. As the overburden is removed by erosion, the ratio of gas entry pressure to total stress becomes increasing unfavourable for normal gas entry. Clayton and Hay [1992] define two basic mechanisms of hydrocarbon (i.e. liquid and gas) leakage across shale caprocks, capillary failure and hydraulic fracturing. According to these researchers, shale caprocks at current depths below surface less than about 500 m are always breached by fracturing. Somewhat controversially, they suggest that capillary failure is the principal mechanism of gas leakage at current depths greater than 1 500 m.

When examining gas migration from a radioactive waste repository three categories of fracture can be considered: (a) fractures which are natural features of the host formation, (b) fractures formed during repository engineering activities, largely within the engineering disturbed zone (EDZ), and (c) pressure-induced fractures which might develop during overpressuring by repository gases.

In simple terms, fractures can be classed in terms of the stress conditions that produced them. By definition, faults always exhibit some wall-rock displacement and can be considered to be large-scale shear fractures or zones of shear fracturing. Smaller scale shear fractures are sometimes observed. Other fractures may be either tensile or extensile in their origin. They exhibit no significant wall-rock displacement, but are capable of opening and closing. For a tensile crack to form, at least one of the principal stresses must be tensile. In contrast, an extensile fracture is the product of extensile straining of the fabric and is it possible for such fractures to develop even when all three of the principal stresses is compressive.

The development of fractures and fissures during stress-relief (unloading) is well known in overconsolidated mudrocks. Such fractures develop when the magnitude of one of the principal stresses becomes substantially less than the other two. These fractures often tend to be planar and are generally orientated so that the normal to the plane is coincident with the axis of the minor principal stress. They may be caused by rupture of the rock fabric under extensile strain. Extensional fractures are also developed during tunnelling operations in fairly-indurated mudrocks, where they tend to be orientated sub-parallel to the tunnel walls [Boisson *et al.*, 1998; Bossart and Adler, 1999; Mori and Bossart, 1999]. Because of the low stress acting normal to these discontinuities, extensile fractures are likely to act as preferential pathways for the movement of repository gases [Rodwell *et al.*, 1999].

Extensile fractures in the engineering disturbed zone (EDZ) around underground repository excavations are usually dilated, leading to enhanced gas permeability along the general line of these excavations. The capillary threshold for gas entry into these dilated fractures will also be lower than for entry into the undisturbed fractures of the host rock. It therefore seems likely that gas flow will be focused in the EDZ. Indirect evidence for enhanced air permeability of the tunnel EDZ is provided by the observed surface oxidation of fracture surfaces (gypsum spots) at the Mont Terri Rock Laboratory in Switzerland [Bossart and Adler, 1999].

If gas transport through indurated mudrocks centres around fractures, then it is inevitable that our attention is focused on the formation, spatial arrangement, density, aperture, and connectivity of these important discontinuities [Rodwell *et al.*, 1999]. These are largely matters for the structural geologists and the practitioners of rock mechanics to elucidate.

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# Gas Migration through Crystalline Rocks

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## 1. INTRODUCTION

Fractured rocks have been considered as potential host rocks for the deep disposal of radioactive waste in a number of countries. The representative repository concepts which were identified in reference [1] as associated with fractured rock involved:

- a) Low- and intermediate-level waste in water-saturated fractured rock.
- b) Spent fuel (or HLW) in water-saturated fractured rock.
- c) Spent fuel in unsaturated fractured tuff (Yucca Mountain).

The key gas-related issues are likely to be different for these three repository concepts. Concept (a) typically involves the emplacement of packaged wastes in caverns or tunnels, probably backfilled with a cement grout, and perhaps involving structural concrete lining. The quantities of gas produced for a given volume of waste are expected to be larger than for spent fuel or high-level waste and may include radioactive gases whose release at the surface requires assessment for its potential radiological consequences. For this concept, understanding the mechanisms and effects of gas migration through the geosphere is important in repository performance assessment.

For concept (b), the waste is typically contained in long-lasting canisters emplaced in holes lined with compacted bentonite. The bentonite barriers are intended to provide the main barrier to groundwater access to the waste, and the quantities of gas expected to be produced are predicted to be sufficiently small that the host rock is not expected to provide a serious obstacle to gas escape from the region of the canister. In this concept, the main barrier to gas migration is considered to be the bentonite buffer; gas migration through this is discussed in a companion paper.

Concept (c) is unique in involving emplacement of wastes in unsaturated rock, well above the water table, in a semi-arid region at Yucca Mountain in Nevada. Here the two-phase flow issues relate primarily to the infiltration of water through the fractured rock from the surface, which may involve flow channelling and intermittent flow, and the generation of strongly heat-driven flows, with extensive vaporisation and condensation phenomena, caused by the emplacement of heat-generating wastes.

The sources for the material presented in this abstract are too numerous to cite in the space available, but can be found in reference [1].

## 2. GAS MIGRATION IN SATURATED FRACTURED ROCK

In the low-permeability water-saturated rock typically considered as potential repository host rocks, most of the permeability is provided by networks of fractures and fissures, with very low

permeability associated with the rock matrix. This characteristic of the rock permeability may have a significant influence on the way gas migrates through the rock.

A number of studies have been carried out to assess whether gas can satisfactorily escape from the near field into the fractured host rock. This depends first on the capillary entry pressure for the fractures, and the consensus for most rocks considered is that this will be small compared with the hydrostatic pressure prevailing at the repository depth. The advance into the geosphere of gas escaping from the repository near field will then depend on the effective permeability of the host rock. The effective permeability required to support a steady-steady flow of gas that is commensurate with the gas generation rate is likely to be typically significantly less than the rock permeability; however, transient pressure rises could be associated with the period during which water, with its higher viscosity and density compared with gas, is displaced ahead of the migrating gas. The extent of any pressure build-up from gas generation could depend on the condition in the repository at the start of gas generation, particularly the degree of water saturation.

The above conceptual model of migration through a water-saturated repository host rock is what emerges from a conventional continuum model of two-phase flow in porous media. It involves a number of assumptions which should be examined, and there are additional aspects of gas migration in fractured rocks that need to be understood.

There is an implicit assumption in conventional concepts of two-phase flow in porous media, that flowing phases are connected phases. Over the substantial distances involved in gas migration through a typical fractured host rock, it can be questioned whether the gas phase would occupy stable connected pathways over extended periods. It has been suggested that transport as bubbles might contribute to underground gas migration. It is also possible that gas pathways might continuously collapse and reform, perhaps with changes to the positions of the pathways in the process. Such occurrences may have the potential to affect water-borne contaminant transport: the collapsing and reforming of gas pathways necessarily implies some movement of the groundwater; groundwater could be entrained in bubble flow thereby accelerating its vertical movement; and contaminants could also attach to bubble surfaces. Viscous and gravitational instabilities (gas rising through water) and the heterogeneity in fracture flow channels are likely to serve to promote instabilities in gas migration.

Assessment of the radiological hazard from the release of radioactive gases at the surface requires some understanding of how the release is likely to be distributed across the surface.

A number of experimental programmes have been instituted to address various of these issues. Programmes of field investigations of gas migration through initially water-saturated fractured rock have included:

- a) injection into an isolated zone in a borehole to establish gas entry pressures;
- b) investigation of gas migration along fractures connecting a pair of boreholes;
- c) monitoring the appearance at the surface of gas injected into fractured rock from an isolated zone of a borehole.

Various laboratory experiments have been carried out on single natural or artificial fractures in attempts to provide a better understanding of two-phase flow in fractures or to measure two-phase flow parameters.

### **3. TWO-PHASE FLOW AT YUCCA MOUNTAIN**

At the unsaturated site of Yucca Mountain, important two-phase flow issues for repository performance under ambient conditions include the percolation flux at the repository level, seepage into

drifts that could contact waste packages, seepage behaviour below the repository, flow partitioning between fractures and rock matrix, and the scale dependence of hydrogeologic parameters

Net infiltration at Yucca Mountain is low, and has been estimated to be of the order of  $5 \text{ mm yr}^{-1}$  on average at current climatic conditions. Thick layers of fractured, welded and non-welded tuffs provide large-scale permeability which easily accommodates natural water percolation rates, causing development of a very thick (approximately 600 m) unsaturated zone. However, water seepage in this zone is complex and difficult to characterise, and it has become clear that low average values for infiltration do not necessarily translate into slow migration of aqueous solutes. Seepage behaviour is highly variable both temporally and spatially. There is now a large body of field data, particularly those provided by man-made environmental tracers, that demonstrate the presence of fast preferential flow paths, where aqueous solutes can travel downward with average flow velocities of  $10 \text{ m yr}^{-1}$  or more. At the same time, slowly moving “old” waters, with ages of several thousand years or more, have also been documented.

Early work at Yucca Mountain had to rely on sampling and observation from boreholes. The recent completion of the Exploratory Studies Facility (ESF) has provided direct access to the proposed repository host formations. Ongoing programmes of ESF-based sampling and testing (including gas-phase flow and water percolation tests that are valuable in determining connectivity of the fracture system) have led to rapid progress in developing understanding of fluid flow and mass transport at the site.

At the repository horizon, the effect of fast preferential flow paths would appear to have the potential to compromise the waste isolation capacity of the repository by providing fast solute transport, but this possibility seems to be balanced by the reduced probability, compared to uniform slow infiltration, of waste packages being contacted by water (episodic flow in localised channels would bypass most waste packages).

Emplacement of a strong heat source in a partially saturated fractured medium gives rise to boiling and condensation phenomena with a potential for large-scale redistribution of moisture. Water will be vaporised in the rock matrix, the vapour will be pressurised and flow towards the fractures where it will condense upon encountering cooler wall rocks. The condensate may drain downward or may in part return towards the heat source under capillary and gravity forces. An intriguing possibility is the development of water-vapour counterflow systems, generally referred to as “heat pipes” in the heat transfer and geothermal literature. Because of their very efficient heat transfer characteristics, heat pipes would keep rock temperatures lower than in purely conductive regimes, and would also cause liquid water to persist in regions that otherwise would be subject to dry-out. This could have considerable impacts on waste package and repository design.

Heat-driven flow at a Yucca Mountain repository has been extensively studied through computer simulation, employing standard multiphase flow and heat transfer concepts borrowed from geothermal reservoir and petroleum engineering. More recently, useful insight was gained through laboratory flow visualisation experiments, and through field tests in the ESF facility at Yucca Mountain.

Laboratory flow visualisation experiments have allowed direct observation and confirmation of many of the multi-phase fluid and heat flow phenomena mentioned above that had been hypothesised in modelling studies. An important recent development is that of heater testing in the ESF facility for examining thermo-hydrologic behaviour over a range of space and time scales.

#### **4. MODELLING GAS MIGRATION IN FRACTURED ROCK**

Two-phase flow models in which the porous medium is treated as a continuum have been the most widely used tools for the numerical simulation gas migration from repositories. The computational models are essentially discretised versions of Darcy's law in which the two-phase aspects of the flow are represented through the introduction of relative permeability and capillary pressure functions of the gas saturation. Determination of these functions for flow in fracture networks presents one of the difficulties of the application of these models to two-phase flow in fractured rocks. Where there is flow in both the fractures and the matrix (for example water infiltrating at an unsaturated site where it will flow rapidly through fractures but can also soak into the matrix), computational models in which the porosity and/or permeability of fractures and matrix are treated as separate interacting subsystems (dual porosity and dual permeability models) can provide additional functionality. These continuum models present the only computationally practicable approach available at present for reasonably detailed field-scale two-phase flow calculations. Heat flow and transport can be included in these models, as well as heat-driven phase behaviour. These extensions can be important for studies of the disposal of HLW and spent fuel.

Despite their widespread use, it has become recognised, as the gas migration and two-phase flow processes associated with deep repositories become better understood, that conventional continuum two-phase flow models have their limitations.

The continuum approach can become inadequate when the scale on which a process can be satisfactorily averaged (using the conventional framework) is smaller than the scale on which a model is discretised in a numerical model (and there is always a practical lower limit to this scale for simulations on a domain of a given overall size). This may occur in water infiltration into unsaturated rock or the development of gas pathways through saturated rock. It does not necessarily follow that a satisfactory macro-scale model cannot be developed to describe such processes, but this will be a different (possibly phenomenologically based) model from the conventional two-phase flow model (or will at least use suitably modified input data designed to produce results on the required scale).

The conventional two-phase approach may also fail when processes are involved that are not encapsulated within the conceptual model. Processes involving entrainment of water in gas flow that exhibits instability would fall into this category.

To meet these difficulties other modelling approaches have been explored to elucidate processes involved in various aspects of gas migration and two-phase flow in fractured rock. These include fracture network models, numerical studies of two-phase flows in single fractures, and the "weeps" model of the rapid flow of infiltrating water through fractures in unsaturated rock. Some of these models may be intended to assist understanding and perhaps in interpreting experiments rather than for direct use in performance assessment.

The development of confidence in models of gas migration and two-phase flow is an issue for all host rock types that needs further attention.

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# Gas Migration through Salt Rocks

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## 1. INTRODUCTION

Salt as a host rock for a repository for radioactive waste may appear as a layered formation as observed at the WIPP site in the USA or as domed salt, which is abundant in the northern part of central Europe. Planned or actual repository sites like Gorleben, Morsleben or Asse in Germany are located in such salt domes. They have risen up in geological time from Permian salt beds until their upward movement has come to an end.

Rock salt exists under geological conditions as an extremely dry material with a residual moisture content well below 1%. Due to its crystalline nature, its permeability and porosity are very low. In addition, because of its plastic behaviour under stress salt has a high self-healing capacity. In fact, under undisturbed conditions, rock salt is considered as impermeable (permeability less than  $10^{-22}$  m<sup>2</sup>). This is demonstrated impressively by brine inclusions which have been included millions of years ago and are kept in place until today. Thus, in considering conditions for two phase flow, undisturbed salt neither offers sufficient water nor appropriate hydraulic properties for scenarios involving normal two-phase flow to occur. Therefore, there is a fundamental difference to other host rock material, in that long term safety analyses for waste repositories in salt have, in general, to assume accident scenarios or some kind of faulted conditions to produce a scenario where gas production and two-phase flow become relevant.

The main focus of those safety analyses is on compacted crushed salt as backfill material, possibly on seals and plugs for emplacement rooms or borehole closures and on the engineering disturbed zone (EDZ).

For completeness, mention should be made of the specifics of the WIPP site in this regard. The Salado formation, in which this repository is situated, is only a few meters distant from intersecting water bearing anhydrite layers. It is therefore assumed that the EDZ extends across the complete salt layer overlying the disposal area. Thus no credit is taken for the sealing properties of undisturbed salt in this case.

Because of the circumstances described above, the following paragraphs do not deal with two-phase flow phenomena in undisturbed rock salt but only in compacted crushed salt and the disturbed rock zone adjacent to the disposal areas.

## 2. CONCEPTUAL MODELS

Modelling of two-phase flow in salt relies on the generalised Darcy approach. Regarding the possible flow regime in the backfill and the EDZ these approach is justified. For application, however, some supplementary model developments are necessary.

### 2.1 Solubility

The solubility of salt requires that the equation of states (EOS) are expressed as a three component system (gas, water, salt) instead of only two components for other host rocks. This means that for thermodynamic properties, vapour pressure, viscosity, gas solubility etc. the dependency from the brine saturation has to be implemented in addition. In a more precise manner, even the composition of the brine would have to be considered. Because of the dominating proportion of NaCl in solution this can often be neglected. [1] gives an overview to which extent common physical laws can be applied for this purpose. More recently an updated relationship for the brine enthalpy enables to enlarge the range of applicability for the thermodynamic properties from 100°C to a lower bound of 10°C [2].

The brine composition may play an important role, if the influence of solubility on the hydraulic parameters for two-phase flow in the solid medium is concerned. Precipitation and re-solution can cause changes in porosity and permeability. Especially in a temperature gradient this process is dependent on brine composition. Its relevance for the fluid flow therefore should be checked and considered in detail where appropriate.

### 2.2 Compaction

The plastic behaviour of salt under stress implies that salt has a high self-healing capacity. Although this effect is highly desirable in terms of isolation of the waste against the biosphere, it may strongly influence fluid flow in the repository by different kinds of interaction. First, compaction of crushed salt reduces the void volume. This in turn may lead to higher gas pressures. Because of the volume reduction porosity and permeability decrease and gas migration is slowed down. This causes an additional pressure increase. Higher pressure creates a counteracting force, which balances the compaction process.

To describe this interaction, several different approaches have been developed. Zhang et al. [3] assume that the influence of temperature, stress and compaction can be treated separately. Based on this assumption they developed the following formulation, which relates the compaction rate  $\dot{\epsilon}$  directly to the geomechanical stress:

$$\dot{\epsilon} = A \cdot e^{-\frac{Q}{RT}} \cdot \left(\frac{\sigma}{\sigma_0}\right)^n \cdot \left(\ln\left(\frac{\phi_0(1-\phi)}{\phi(1-\phi_0)}\right)\right)^{-m} \quad (1)$$

where

- $R$  is the universal gas constant (8.314 10<sup>-3</sup> kJ mol<sup>-1</sup> K<sup>-1</sup>),
- $Q$  is the activation energy (kJ mol<sup>-1</sup>),
- $\sigma_0$  is the reference stress (1 MPa),
- $\phi_0$  is the reference porosity,
- $\phi$  is the porosity, and
- $A, n, m$  are material specific parameters.

In [1] further examples for such relations are given. Since a direct coupling between the geomechanical stress and fluid dynamic processes is an ambitious task which requires considerable computational resources *constitutive relationships* have been sought, which represent the relationship between stress and the fluid dynamic parameters in a way, that is easier to handle numerically. A common approach is to use the fluid pressure instead of the hydrostatic stress. Several conceptual models have been developed based on this assumption [1].

Another approach used in the performance assessment for the WIPP repository is the *boundary backstress method*. Salt is considered to behave as a fluid phase with high viscosity. The fluid properties are calibrated in advance by geomechanical calculations to approximate the expected salt creep and room closure.

Finally, the *pressure-time-porosity (PTP) line interpolation method* is based on extensive precalculated compaction histories which are used to adjust the porosity as a function of gas pressure and time during two-phase flow calculations.

All these approaches need further constitutive relationships for modelling like the relation between porosity and permeability or the capillary pressure as a function of saturation. For the first relationship, Kozeny's equation or empirical functions of the type

$$k = a \cdot \phi^n, \quad (2)$$

are common, where  $a$  and  $n$  are material specific parameters. The second relationship is mostly based on a dimensionless function introduced by Leverett [4] although limitations exist with regard to the dependency of the capillary pressure on the permeability.

### 2.3 Flow type

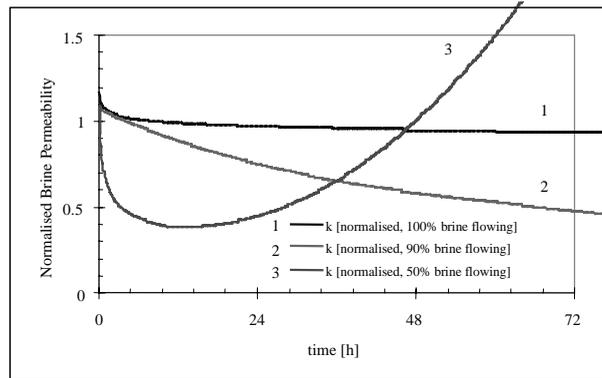
Two-phase fluid flow in compacted crushed salt or the EDZ of a repository can adequately be described by Darcy's law. Flow mechanisms other than Darcy flow, however, have to be considered for brine supply from rock salt for the calculation of a proper balance of the available humidity. Especially for temperature driven brine migration in rock salt Knudsen diffusion may be more appropriate.

## 3. EXPERIMENTAL FOUNDATION

Basic salt properties like salt solubilities or enthalpy of salt solutions are well established, but some supplementary work is necessary for the characterisation of the thermodynamic properties of the three-component system brine-gas-salt.

Modelling of salt compaction has increasingly been validated by experimental work, but a comprehensive material law is still lacking. For two-phase flow modelling, existing compaction models may be considered as sufficiently detailed to be implemented in numerical models for isothermal conditions, i.e. for waste with negligible heat generation. Two-phase flow parameters have to be determined experimentally for specific sites. First measurements of these parameters show, that requirements for the measurement technique are demanding and results may not conform with common expectations from other materials [5]. Figure 1 demonstrates this by measurement results for permeability history in salt cores flooded with undersaturated brine. Surprisingly, permeability initially decreases in spite of salt dissolution.

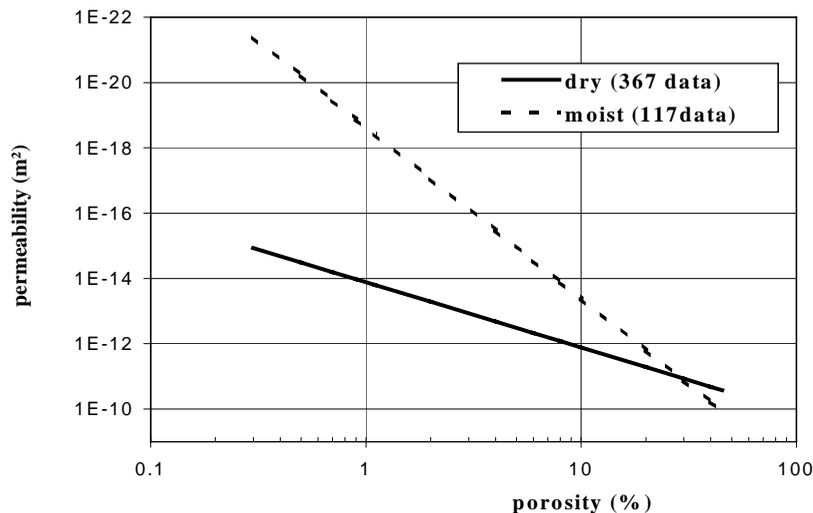
Figure 1. Effect of solution process on the brine permeability



The observed decrease in permeability is probably caused by occlusion of small salt crystals occurring in the flowing brine. However, due to the relatively high effective flow velocities in the experiment the dissolution as well as the occlusion are considered as local effects which do not influence remote flow area. Therefore, for realistic flow processes just a local dissolution of the refill material is expected, but no effects in more distant flow areas.

Numerous experimental investigations of the permeability and porosity of compacted crushed salt exist. A comprehensive review of literature up to 1991 is given by [6] and up to 1995, for measurements concerning the WIPP site, by [7]. A comparison of different investigations shows large scattering caused by different imposed conditions, experimental setup and salt composition. Figure 2 demonstrates this effects by differentiating between measurements on dry ( $\leq 0,1\text{wt}\%$ ) and wet ( $\leq 1\text{wt}\%$ ) samples. Accordingly, different parameters for equation (2) have been derived [8].

Figure 2. Permeability-porosity relationship [8]



#### 4. NUMERICAL MODELLING APPROACHES

Salt specific model implementations in two-phase flow codes have mainly involved the codes ECLIPSE, CODE\_BRIGHT, BRAGFLOW and TOUGH2. Comparable implementations of compaction models exist for single-phase flow codes, but are omitted here. Since a complete comparison of the four codes mentioned would be out of the range of this paper only the salt specific features are discussed here.

*Solubility effects* are treated only in CODE-BRIGHT and TOUGH2. CODE\_BRIGHT considers some solubility effects for the salt dissolved in the brine, but no dissolution and precipitation of the host rock. For the TOUGH2 there are two EOS modules addressing solubility effects and thermodynamic properties of salt, EOS7 and EWASG. The EOS7 module describes the fluid properties of brine and does not take solubility effects into account. The EWASG module is much more general and includes dissolution and precipitation as well as porosity related changes of permeability. In the latest release of TOUGH2 (version 2.0) both features are incorporated.

For the ECLIPSE code, modelling applications to *compaction* using the boundary backstress method exist. The modifications for consideration of compaction were applied in a safety analysis of the WIPP site [9]. The parameters for creep were determined using the SANCHO finite element code. The results indicate that, up to a period of approximately 100–200 years, porosity of the disposal room decreases with time due to creep, then it increases due to pressure increase caused by gas generation.

CODE-BRIGHT is the only code with implementation of compaction in its standard version. Compaction is modelled in terms of geomechanical parameters according to [10]. For the permeability-porosity relationship, Kozeny's equation is used.

Freeze *et al.* [7] implemented both the PTP line interpolation method and the boundary backstress method into TOUGH2 for a safety analysis of the WIPP site. The PTP relationship for the WIPP site was established by numerous simulations of convergence using SANCHO. For the boundary backstress method, the same procedure as in the ECLIPSE code was used. Lorenz and Müller [11] and Javeri [12] implemented compaction by constitutive relationships in TOUGH2 based on the semi-empirical evaluation of extensive experimental work.

The two-phase flow modelling part of BRAGFLOW relies on the same physical models as TOUGH2. Since BRAGFLOW was used in performance assessment for numerous calculations to establish cumulative complementary frequency distributions for possible consequences and their probability of occurrence its numerical approach is simplified compared to ECLIPSE or TOUGH2. Therefore, the extensive two-phase calculations for WIPP performed with TOUGH2 can be considered as a kind of indirect confidence building based on the comprehensive testing and application experience for TOUGH2.

Compaction modelling was based on the PTP relationship. The same procedure as for ECLIPSE was used in BRAGFLOW with the geomechanical code SANTOS for establishing the PTP relationship.

Although compaction is always connected to changes in porosity and permeability, the *additional constitutive relationships* needed to describe these were not implemented in all cases when compaction was modelled; the reasons for this are not always apparent. In particular, the additional constitutive relationship for capillary pressure is only implemented in TOUGH2 version 2.0, which was just recently released. In this version the constitutive relationship for porosity and permeability due to precipitation and resolution according to Verma and Pruess [13] is implemented. Earlier versions use the exponential expression outlined above with varying parameters [11,12]. The

calculations were performed for the Morsleben repository and the Gorleben site. Table 1 summarises the main features of the four codes treated.

Table 1. **Two-phase flow codes with salt specific modules**

<b>code/effect</b>	<b>ECLIPSE</b>	<b>CODE-BRIGHT</b>	<b>BRAGFLOW</b>	<b>TOUGH2</b>
solubility	–	(+) <sup>1</sup>	–	+ (EWASG)
compaction	boundary backstress	geomechanical	boundary backstress +PTP line	constitutive relationship
permeability-porosity	–	Kozeny	related to brine pressure	Verma & Pruess + exponential
capillary pressure	–	–	–	Leverett
<sup>1</sup> . only for salt in brine				

## 5. CONCLUSIONS AND RECOMMENDATIONS

Although a good deal of progress has been reached with regard to two-phase flow modelling in salt in the past ten years has been achieved there is still theoretical and experimental work needed for complete understanding and satisfactory modelling of two-phase flow in repositories in salt formations.

From the theoretical aspect, modelling of compaction requires additional work. The present models may be considered appropriate for isothermal situations. In HLW disposal, however, neither constitutive relationships nor the boundary backstress method or the pressure-time-porosity approach are sufficiently qualified. The first method at present implies that effective stress and fluid pressure are linearly coupled. If this assumption holds if a heat source is introduced providing an additional stress field has still to be demonstrated. At present it seems more promising to go for a direct coupling of geomechanical parameters with fluid flow, even if this means a higher effort for implementation. The two other methods are based on isothermal geomechanical calculations. For HLW disposal this would require calculations including temperature histories. If this can be managed with reasonable efforts and if the calculations can be validated are still open questions.

Constitutive porosity-permeability relationships fail at low porosities. Here theoretical work is required to identify essential physical mechanisms to provide the framework for better parameterisation in this range.

From the experimental point of view, data on capillary pressure and relative permeability are needed urgently for appropriate simulations. In repositories in rock salt, safety analyses require scenarios as a flooded and an inflow scenario for brine, which correspond to imbibition and drainage. Therefore, hysteresis effects also have to be taken into account. Since these effects lack sufficient theoretical foundation as well as experimental characterisation both areas should be covered by further research work.

The presently available models and the incorporated features have all been developed on the basis of experimental evidence. However, this evidence was always restricted to the individual effect that had to be described. For a qualified application of the models and the related codes it seems helpful to perform integrated experiments, which cover not only the individual effects but also the possible interactions.

Finally, increasing modelling sophistication requires correspondingly higher computational resources. The existing codes that are applied in this field are already at their limit with regard to computing time and storage demand. Therefore additional efforts are necessary to improve their computing speed and to provide adequate data handling features.

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# **The EC/NEA Status Report: Conclusions and Outstanding Issues**

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Companion papers to this will have specifically addressed gas migration and two-phase flow in both the engineered barriers of deep underground radioactive waste repositories and the various types of host rock in which such repositories might be located. In doing so both experimental data that has been collected about gas migration and two-phase flow in these materials and the ways in which these processes have been modelled will have been discussed. Here an attempt is made to summarise the conclusions that were reached about which issues relating to gas production in repositories are important for different repository concepts, the current capability to assess the effects of gas produced in repositories, and the issues that need to be resolved.

It is important to note that the main findings of the status report [1] summarised below are intended to provide only a generic overview of the status of the main gas issues for various, reference disposal concepts. They should not be used to support definite conclusions for a specific repository and/or site; indeed, the importance of issues can be highly dependent on the particular circumstances and should be assessed in relation to these. Such a short summary also necessarily represents a gross oversimplification of the results of the report, and reference should be made to the original for more complete details.

Amongst the repository concepts that are reviewed in the report, that for Yucca Mountain is unique in that disposal is envisaged in unsaturated rock in an arid region. The consequence of this is that for this concept two-phase flow issues are integral and central to repository performance analysis, and have therefore commanded more resources and attention than gas migration in repository concepts developed for saturated locations. It is now evident that seepage behaviour at Yucca Mountain is highly variable both temporally and spatially, and that fast preferential flow paths exist. Ongoing investigations include the interplay between fast preferential flow and perched water bodies, the manner in which localised infiltration pulses may be dampened and slowed in the less fractured non-welded units, the role of fracture-matrix interaction in flow and transport, hydrogeologic properties and behaviour of fault zones, capillary barrier effects at sloping lithologic contacts, and heat-driven two-phase flow processes. Conventional approaches to modelling these systems using large finite-difference models are subject to practical limitations arising from the complex heterogeneous nature of the fractured and faulted tuff formations, and from preferential flow effects on small scales that may not be adequately described by large-scale volume averaging. Serious questions and uncertainties remain about the conceptual basis and validity of these large-scale volume-averaged models. To meet these difficulties alternative, more phenomenologically oriented modelling approaches for performance assessment have also been developed. These approaches avoid the complexities and conceptual difficulties of the mechanistic process models; however, their calibration may not be any easier than it is for the detailed process models.

It is argued that the simultaneous pursuit of several alternative modelling approaches and concepts to describe unsaturated flow and transport behaviour on different scales, together with numerical, field, and laboratory studies, should lead to the degree of robustness in the engineering analysis and design needed to establish confidence in a repository project for this environment.

For other repository concepts, those involving emplacement in saturated rocks, the scenarios involving substantive gas migration issues vary with repository concept. Important in the evaluation of gas migration effects is of course the rate and period of gas generation.

For some waste types, notably low- and intermediate-level wastes, particularly when there is the ready availability of water, gas is expected to be generated at significant rates and its effects in migrating from the repository have to be addressed. For these wastes it is probable that in most circumstances a free gas phase would form and migrate from the repository.

For repositories for LLW/ILW in fractured crystalline rock, the consensus appears to be that there will in most cases be sufficient transport capacity in the rock to accommodate the expected flux of gas from such wastes without mechanical disruption, although some issues remain in demonstrating that the arguments on which this view is premised are applicable at the field scale. There remains a need for a proper range and quality of field data on gas migration through both fractured crystalline and other rock types to build confidence in predictions of gas migration at the large scale. For some waste inventories, radioactive gases will be amongst the gases produced and will be transported along with the larger volumes of non-radioactive gases. Further understanding of the way radioactive gases might be delivered to the surface may be required to support performance assessment in some cases. A significant source of uncertainty at present is the possibility that migrating gas might affect the transport of water-borne radionuclides. This could occur through a number of mechanisms: (i) instabilities in gas flows or the transport of gas as bubbles could enhance the vertical component of the local groundwater flow; (ii) adsorption of contaminants at gas-interfaces could contribute to the transport.

For repositories in very low-permeability mudrocks or clays for wastes producing significant quantities of gas (e.g. LLW/ILW), conventional porous-medium flow concepts would suggest that substantial gas pressure build-up in the repository to levels of potential concern could occur. However, alternative mechanisms of gas migration that would alleviate this situation are postulated to occur in mudrocks and other argillaceous materials (including bentonite buffers). The amount of data available on mudrocks is very limited, and further data acquisition is needed to rectify this situation (to a lesser extent for all argillaceous materials). In such impermeable host rocks, the role of backfilled and sealed shafts or addits, and engineering damaged zones in providing a gas migration route is an issue for investigation. Such evidence as is available suggests that, after gas generation has ceased, water ingress into the gas pathways will cause them to reseal, restoring the original low host rock permeability and preventing the gas pathways from providing fast transport routes for dissolved radionuclides, but this needs to be confirmed.

In some waste concepts, notably for high-level waste or spent fuel in canisters made of non-corroding materials, or in a repository in salt, where water supply constraints will eventually inhibit gas production, very little gas production is predicted in the planned repository evolution scenario, and a free gas phase is predicted not to be formed at all in some cases. However, scenarios involving accidents or based on conservative assumptions can be envisaged in which free gas could be formed in these concepts also, and thus gas migration needs to be addressed for them as well. For these wastes types the typical primary gas migration barrier is either compacted bentonite or converging crushed salt, depending on host rock type. The development of understanding of gas migration in these materials is an ongoing research programme involving investigation of complex processes, usually involving coupled thermo-hydro-mechanical effects.

For gas migration in bentonite buffers the precise mechanism of gas migration and the factors which control it remain controversial, and require further elucidation, although the empirical evidence that gas can escape through a saturated buffer satisfactorily, and that the buffer subsequently reseals is encouraging. The resaturation of bentonite buffers in the presence of heat from wastes, and the way that this might influence the state of the buffer when gas migration starts are issues for further clarification.

For a repository in salt, gas migration issues have greatest importance for scenarios giving rise to the possible inflow of brine into the repository, when gas generation may no longer be dependent on water availability. Understanding potential counter-current flow regimes with water inflow and gas outflow is complex, and behaviour is likely to be design and scenario dependent. The complexity includes possible coupling of the convergence behaviour of the salt (because of its plasticity) with two-phase flow and gas generation, and the effect of salt solubility (which is affected by both temperature and pressure gradients). For the assessment of such scenarios, models that integrate these complex coupled processes are needed and are still under development.

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## **SESSION II**

### **PRESENT APPROACHES TO DEALING WITH GAS-RELATED ISSUES IN PERFORMANCE ASSESSMENT EXERCISES**

Chairman: Paul Marschall (Nagra, Switzerland)



# **The Gas Pathway in a Site-specific Performance Assessment: Nirex 97**

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## **1. INTRODUCTION**

The presentation relates to work on the assessment of the post-closure performance of a repository at Sellafield conducted by Nirex in support of the development of a deep geological disposal facility for the United Kingdom's intermediate-level and certain low-level radioactive wastes. A significant proportion of these wastes are produced as a result of the reprocessing of spent nuclear fuel and in consequence contain significant quantities of long-lived radionuclides (actinides and long-lived fission and activation products). The Nirex repository concept is that wastes would be immobilised, typically in a cement-based matrix in a steel or concrete container and the resulting waste package placed in excavated vaults deep underground. The spaces around the emplaced packages would eventually be backfilled with a specially-formulated, cement-based material (Nirex Reference Vault Backfill) before sealing the access to the repository.

Volume 4 of Nirex 97 [1] describes the generation of gases within the modelled repository at Sellafield, the reaction of gases with cementitious components of the repository, the migration of gas through the near field geosphere, and the radiological impact of gas in the biosphere. Modelling techniques to assess the performance of the repository for the gas pathway are also described, and details of the treatments of repository resaturation by groundwater after closure and of gas flammability hazard are presented. A description of the overall approach to the assessment of the gas pathway is given in Nirex Science Report S/04/003 [2].

The hydrogeological conceptual model of the Sellafield Site for the gas pathway assessment is the same as that used in the groundwater pathway assessment. However, the time scales of relevance to the gas pathway are rather shorter than those of relevance to the groundwater pathway. Whereas the most significant consequences of the release of radionuclides to the biosphere as a consequence of the groundwater pathway arise on time scales in excess of thousands of years, the most significant consequences of the gas pathway arise on time scales of order of a hundred years, within the likely control period for a repository. Interactions between gas and groundwater have not received a detailed treatment in Nirex 97; research on this topic is proceeding in the Nirex Safety Assessment Research Programme [3].

## **2. GAS GENERATION**

A range of gases may be generated within the repository in significant quantities [3]:

- (a) hydrogen may be generated by the corrosion of metals in wastes and packaging;

- (b) methane and carbon dioxide may be produced by microbial degradation of cellulosic material in wastes, mainly wood and paper;
- (c) hydrogen sulphide may be generated in small quantities from microbial action on sulphate ions in waste and groundwater; and
- (d) nitrogen may be generated by microbial action on nitrate ions in wastes and groundwater, although the quantities generated are small.

Traces of three other types of gas may also form:

- (a) radioactive gases might form, in particular tritiated hydrogen ( $^3\text{HH}$ ), methane containing carbon-14 ( $^{14}\text{CH}_4$ ) and carbon dioxide containing carbon-14 ( $^{14}\text{CO}_2$ );
- (b) radon could be formed by the radioactive decay of uranium, thorium and radium;
- (c) other toxic non-radioactive gases might form as the result of microbial processes.

The quantity of gas generated in the repository will vary as a function of time, and the generation of these gases could give rise to a number of potential hazards. Some of these potential hazards can be excluded from consideration on the basis of simple arguments. A treatment of other hazards, such as gas-driven groundwater flow, requires further data or information that is not currently available and would require *in situ* experiments in an underground laboratory. Such hazards are being investigated, as discussed in Nirex Science Report S/96/002 [3]. The key hazards discussed in Nirex 97 are:

- (d) the radiological consequences (risks or doses) of exposure in the biosphere to the radioactive forms of the gases generated within the repository (particularly  $^{14}\text{CO}_2$ ,  $^{14}\text{CH}_4$ ,  $^3\text{HH}$ , radon-222 and radon 220) or in the case of radon-222 and radon-220, “stripped” from the geosphere;
- (e) the extent of overpressurisation within the repository vaults, as this might create new pathways and driving forces for the migration of gas and groundwater;
- (f) the toxicity of trace gases such as hydrogen sulphide;
- (g) the potential for the occurrence of flammable mixtures of hydrogen and methane in man-made surface buildings.

The quantity of gas generated in the repository is estimated as a function of time. The program GAMMON [4] has been used in Nirex 97 to model the generation of hydrogen, carbon dioxide, methane and hydrogen sulphide by corrosion of metals in wastes and packaging, and by microbial degradation of organic wastes. Solid-state diffusion may result in the release of  $^3\text{HH}$  from waste steels and this process is also considered.

Repository performance is also affected by the extent to which generated gases react with the cementitious components of the repository. Significant reactions are expected in the case of carbon dioxide and hydrogen sulphide.

### 3. GAS MIGRATION

In order to understand the consequences of gas release to the biosphere, it is also necessary to develop models of the transport of gas through the repository and the geosphere. In Nirex 97, the program techSIM [5] was utilised to model two-phase flow of gas and groundwater. A model of the domain around the repository was constructed and used to evaluate the degree of overpressurisation

that would occur in vaults, the times taken for gas to be released from the vaults and to migrate to the surface, and the fluxes of gas to the biosphere. The influence of gas generation on the resaturation of the vaults was also examined. The generation rates, estimated using GAMMON, are supplied as input data to techSIM.

Additionally, a separate steady-state model of radon emanation, transport in a carrier gas and decay has been developed for Nirex 97. This model addresses:

- (a) the transport of repository-derived radon from the vaults as a minor component of the gas released from the repository;
- (b) the transport of naturally occurring radon, 'stripped' from the geosphere by migrating gases.

The migration calculations are based on the same hydrogeological data that are used in the assessment of the groundwater pathway.

#### **4. SUMMARY OF REPOSITORY PERFORMANCE FOR GAS PATHWAY**

On the basis of the models and data used in the assessment of the gas pathway, most of the potential hazards from the gas pathway have been assessed to be insignificant.

$^3\text{HH}$  is expected to be generated in a number of ways: by attack of  $^3\text{HHO}$  on metals and packaging; by release of embedded  $^3\text{HH}$  from metals by solid-state diffusion. Tritium has a comparatively short half-life (12.35 years), and therefore any radiological consequences occur at relatively short times after closure, during the period of control.

Calculations of annual individual doses for releases of  $^3\text{HH}$  to the soil have been undertaken. These have taken account of the decay during the period up to the first breakthrough of gas in the biosphere and some spreading of the peak in the appearance of gas at the surface compared with the generation profile. For discharge to soils, the estimate of annual individual dose for a discharge area of  $10^4\text{m}^2$  is calculated to be  $0.25\text{ mSv yr}^{-1}$ . This dose would only arise for a highly focused discharge; this is judged unlikely<sup>1</sup>. For a larger discharge area of  $3.5 \times 10^5\text{m}^2$  (corresponding to an area occupied by a small farm) the calculated dose is  $7.2 \times 10^{-3}\text{ mSv yr}^{-1}$ . The time of the maximum dose is 48 years; it falls off exponentially thereafter, because of radioactive decay. Doses for discharges to buildings are around a factor of 20 lower than those for discharge to soil.

The calculated doses and risks from  $^{14}\text{CH}_4$  from the soil pathway and from inhalation are not significant. One waste stream has been identified to date that contains microbially accessible carbon-14 making it a potential source of  $^{14}\text{CO}_2$ . Numerical modelling indicates that this trace gas would be generated over a comparatively short time scale (within  $10^2$  years of closure). However, it is concluded that the  $^{14}\text{CO}_2$  would react with the cement matrix inside the container to produce calcite with a negligible quantity remaining as free gas. This is based on the assumption that the waste containers would be engineered such that equilibration between carbon dioxide and in-drum grout is achieved. The Nirex Reference Vault Backfill would in any case afford additional containment by reacting with any  $\text{CO}_2$  released from the waste containers.

Radon will be produced from the radioactive decay of uranium, thorium and radium present in the wastes in the repository and in naturally occurring minerals in the host rocks. Radon from these sources might enter the stream of gas migrating from the repository to the surface. The half-lives of the main radon isotopes are short, radon-222 being the longest lived (half-life 3.82 days), and this is

the primary isotope requiring consideration in performance assessments. It is found in this assessment that radon generated in the repository is insignificant but that higher calculated doses result from radon-222 “stripped” from the Quaternary rocks by the bulk, non-active gases generated in the repository.

The possibility of repository overpressurisation was considered in Nirex 97. The calculated overpressures are assessed to be too low to give rise to significant deleterious effects on the repository system in terms of cracking. Although a number of toxic gases, such as hydrogen sulphide may be generated in the repository, it is considered that they will react with the materials in the repository, and therefore no toxic hazards will arise in the biosphere. The possibility that flammable mixtures will form in surface engineered structures has been considered. It is assessed that flammable mixtures of gases are extremely unlikely to form in engineered structures at the surface, above a repository, even in the event of a highly focused discharge.

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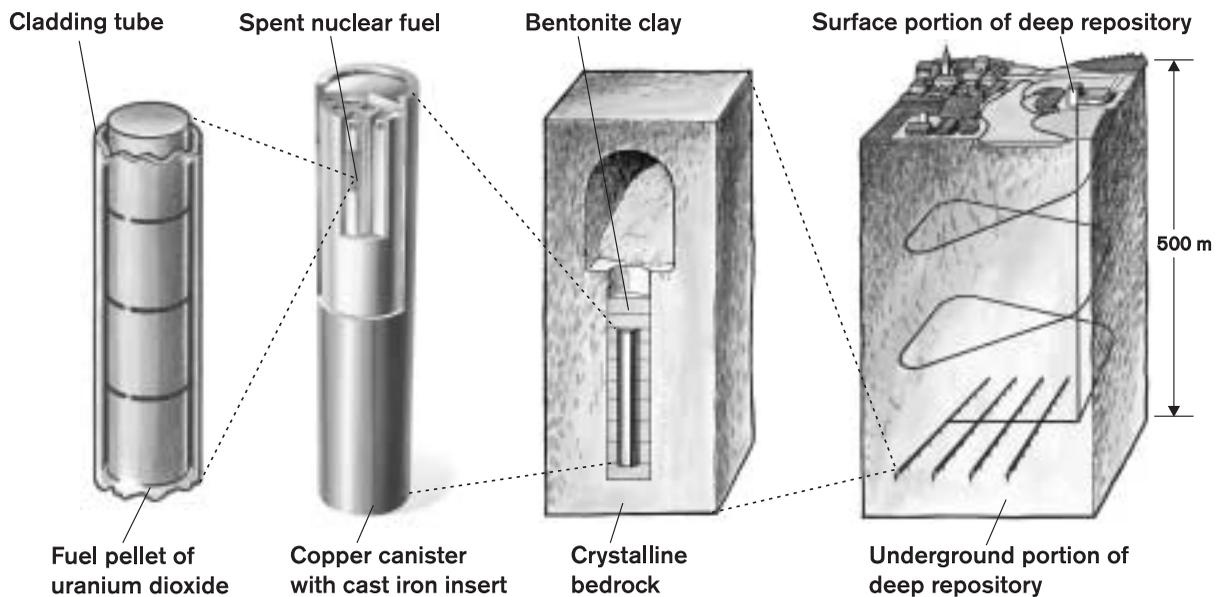
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# The Treatment of Gas in the SR 97 Performance Assessment for a Spent-fuel Repository in the Swedish Crystalline Basement

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In preparation for coming site investigations for siting of a deep repository for spent nuclear fuel, SKB has carried out the long-term safety assessment SR 97[1], requested by the Swedish Government. The repository is of the KBS-3 type, where the fuel is placed in isolating copper canisters with a high-strength cast iron insert. The canisters are surrounded by bentonite clay in individual deposition holes at a depth of 500 m in granitic bedrock (figure 1). Geological data are taken from three sites in Sweden to shed light on different conditions in Swedish granitic bedrock.

Figure 1. The KBS-3 system



## 1. GAS ISSUES

The processes that are related to gas in SR 97 can be divided up into five groups:

- 1) Gas inside an intact canister.
- 2) Water intrusion and gas generation.
- 3) Gas escape through buffer and geosphere.
- 4) Effects of hydrogen gas on spent fuel alteration.
- 5) Radionuclide transport in gas phase.

This paper gives a brief overview of the treatment of these processes in SR 97.

## 2. GAS INSIDE AN INTACT CANISTER

Air and water in an intact canister can be decomposed by means of radiolysis. The products can then be converted to corrosive gases such as nitric acid and nitrous acid. These gases can be of importance for stress corrosion on the canister insert. Calculations show that with 50 g of water, just under 160 g of nitric acid can be formed. This will have no effect on the general corrosion of the insert. In order for SCC to occur, not only a corrosive environment but also tensile stresses in the material are required. In the deep repository, the canister insert is under external pressure and tensile stresses occur on the cast insert only locally and in small areas. It is therefore highly improbable that SCC could lead to penetrating cracks in the canister, and above all that this would jeopardize the integrity of the canister.

$\alpha$  particles (helium nuclei) from  $\alpha$  decay in the fuel form gaseous helium after they have been slowed down in the fuel matrix. Around a fuel rod with an intact cladding tube, this leads to a pressure build-up inside the tube, which can in turn lead to mechanical tube rupture. The pressure increase lies in the range 10 to 20 MPa in 100,000 years, based on a void of 20 cm<sup>3</sup>. If this pressure should lead to rupture of the cladding tubes, the pressure increase in the void in the canister would be in the order of 0.1 MPa, which is completely negligible.

## 3. WATER INTRUSION AND GAS GENERATION

If the copper canister is penetrated, water can enter the canister cavity as liquid or water vapour. Transport of water, water vapour and other gases in the canister is then determined by the detailed geometry of the canister cavities, the presence of water/vapour in the cavities, and temperature and pressure. The process is strongly coupled to several other processes, for example corrosion of the canister insert, where water is consumed and hydrogen is formed.

Water/gas transport in the canister is of fundamental importance for a number of other processes that are dependent on the presence of water in the canister, such as fuel dissolution, radionuclide transport and corrosion of the iron insert.

If the canister is penetrated, water will enter. The process is controlled by the pressure difference between buffer and canister cavity (initially 5-7 MPa) and the hydraulic conductivity in the bentonite. Water ingress can be expected to proceed very slowly. In the presence of water in the canister, gas will be generated both by radiolysis and by corrosion of the cast iron insert. The build-up of gas pressure in the canister will lead to a gradual decrease in the inflow of water and, when the pressure is sufficiently high, gas transport through the canister and the bentonite.

Qualitatively, the course of events can be described as follows: water enters through the hole in the copper canister and causes anaerobic corrosion of the iron surfaces. This leads to hydrogen gas formation, which gradually increases the pressure inside the cavity in the canister and thereby reduces the ingress of water. The corrosion leads to consumption of water. If the surface area available for corrosion is constant, the rate of water consumption will also be constant and the water level in the canister will reach a peak and then sink. Even after all liquid water has been consumed, iron corrosion will continue since water vapour can diffuse into the canister.

#### **4. GAS ESCAPE THROUGH BUFFER AND GEOSPHERE**

Transport of gas in the buffer can occur in two phases of the repository's evolution:

- When the repository is sealed, air will be trapped in the buffer. When the buffer becomes saturated with water, the air must escape.
- If a canister should have a through-wall defect and water should penetrate through the copper shell, the cast iron insert is expected to corrode and release hydrogen. If more hydrogen is produced than can be dissolved in the water in the canister, a gas phase will form.

Gas which is trapped in or by the buffer and backfill can escape in two ways:

- If the production rate is low or the gas quantity is small, the gas can be dissolved in the pore water and be removed by diffusion.
- If the production rate is high or the gas quantity is large, a gas phase will form, the pressure will rise, and a flow path will be formed through the buffer at a critical pressure.

Gas in conjunction with wetting of the buffer and backfill is expected to be dissolved in the buffer's pore water and transported by diffusion.

If the copper canister is defect, hydrogen gas formed by corrosion of the cast iron insert can dissolve in the pore water and migrate from the canister by diffusion. However, the diffusive transport capacity is considerably lower than the hydrogen gas production from corrosion if it is assumed that the entire surface of the cast iron insert is accessible for corrosion and the water supply does not limit corrosion. Under such conditions it is probable that a gas phase will be formed inside the canister and that the gas must escape by gas-phase flow. In an initial phase after canister breakthrough, water is expected to enter the canister, resulting in corrosion and hydrogen gas generation. The gas pressure in the canister will thereby rise. At a critical pressure, the buffer is expected to allow the gas to pass through. A large number of experiments have shown that bentonite does not allow gas to pass until the pressure exceeds the sum of the swelling pressure and the water pressure. When the pressure reaches this value, a transport pathway is formed through the buffer and gas is released. The pressure falls, and if the gas production is low enough, the transport pathway is expected to close. This takes place at a so-called "shut-in pressure", which is dependent on the swelling pressure.

#### **5. EFFECTS OF HYDROGEN GAS ON SPENT FUEL ALTERATION**

In SR 97, a fuel alteration model is used that assumes that the fuel matrix is dissolved as a consequence both of its "own" solubility and of the oxidants produced by radiolysis of water. The model quantifies:

- Radiolysis processes in the water between fuel and cladding.

- A series of reactions between different radiolysis products in the water and between radiolysis products and dissolved hydrogen from corrosion of the insert.
- Reactions between oxidants and the uranium dioxide, i.e. the direct cause of fuel dissolution.

The hydrogen concentration is of crucial importance, since hydrogen reacts with the OH radical to form atomic hydrogen:  $OH + H_2 \rightarrow H_2O + H$ . Atomic hydrogen in turn reacts with  $H_2O_2$  and  $O_2$ , i.e. the oxidants are consumed by atomic hydrogen instead of reacting only with the fuel.

## 6. RADIONUCLIDE TRANSPORT IN GAS PHASE

A defective canister is expected to contain hydrogen gas for a very long time. Radionuclides could thereby be transported to the surface in the gas phase if hydrogen were released. Carbon-14 and radon-222 are the only remaining radionuclides that could conceivably be transported in the gas phase at the times such transport is possible.

C-14 could occur as carbon dioxide or methane, which means that it could be transported in the gas phase. A large portion of the gas in a canister is expected to be released as a pulse. The transport resistances in both the near and far field are small for gas transport. The importance of gas-phase transport is illustrated with an extreme case:

- 2.5 per cent of the inventory of C-14 is released at the time of deposition.
- The gas is released directly to the biosphere without retardation.
- The dose is calculated in the same way as for gas releases from reprocessing plants.

With these premises, the collective dose was 480  $\mu$ manSv for C-14 locally and regionally.

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# **The Treatment of Gas in the H12 Performance Assessment for HLW Disposal in Japan**

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## **1. INTRODUCTION**

The Japan Nuclear Cycle Development Institute (JNC) has published the second progress report (entitled H12) describing research and development for geological disposal of high-level radioactive waste (HLW) in Japan. The purpose of the H12 project was based on the guidelines in April 1997 by the Advisory Committee on Nuclear Fuel Cycle Backend Policy of the Atomic Energy Commission (AEC) [1]. The main goal of the H12 is to demonstrate the technical feasibility and reliability of the specified concept of geological disposal in Japan, prior to site selection and decisions on other key factors such as safety criteria. Accordingly, a number of geological environments, repository and engineered barrier designs and safety measures are described in the H12 safety assessment.

The potential effects of gas generation and transport on the integrity of the engineered barriers and release of radionuclides on the H12 safety assessment are:

- damaging of the engineered barriers due to a gas over-pressure; and
- drainage (or release) of contaminated water from the repository.

This paper presents a description of the gas-related impacts on the H12 safety assessment.

## **2. GEOLOGICAL DISPOSAL CONCEPT IN JAPAN**

The concept of geological disposal in Japan is similar to that in other countries, being based on a multiple barrier system which combines robust engineered barriers with the natural barrier present in the geological environment. The present approach for development of the repository concept has targeted neither a particular type of rock nor a specific site. In establishing the feasibility of this concept, particular consideration is given to the long-term stability of the geological environment, taking into account the fact that Japan is located in a tectonically active zone. The wide range of geological environment present in Japan is also considered. Given this context and Japan's complex geology, a design for an engineered barrier system (EBS) with margins of safety in its isolation functions sufficient to accommodate a range of geological environments has been developed. The major role in overall barrier performance of the disposal system is expected by the near-field, while the remainder of the geosphere serves to reinforce and complement the performance of the EBS.

The reference layout of the EBS involves either axial, horizontal emplacement in the tunnel or vertical emplacement of vitrified waste encapsulated in a steel overpack which are surrounded by highly compacted bentonite. This massive EBS was introduced to ensure long-term performance of the disposal system for a wide range of geological environments.

The geological disposal system in H12 safety assessment was defined by combining the reference-case geological environment and design. Alternative systems were also considered in the process of evaluating potential scenarios and carrying out various sensitivity analyses. The reference-case geological environment and design is shown in Table 1 [2].

Table 1. **The reference-case geological environment and design**

Topography	plains (lowlands)
Groundwater	fresh-high pH groundwater
Rock type	crystalline rock (silisic)
Vitrified waste	model vitrified waste inventory referring to JNFL specifications (assuming 40 000 containers)
Overpack	carbon steel: 0.19m thick
Buffer material	bentonite 70 wt% + sand 30 wt% dry density: 1.6 Mg m <sup>-3</sup> , thickness: 0.7 m
Backfill material	mixture of bentonite with crushed rock
Depth of repository	1 000 m
Waste emplacement	horizontal emplacement

### 3. **GAS-RELATED IMPACTS IN H12 SAFETY ASSESSMENT**

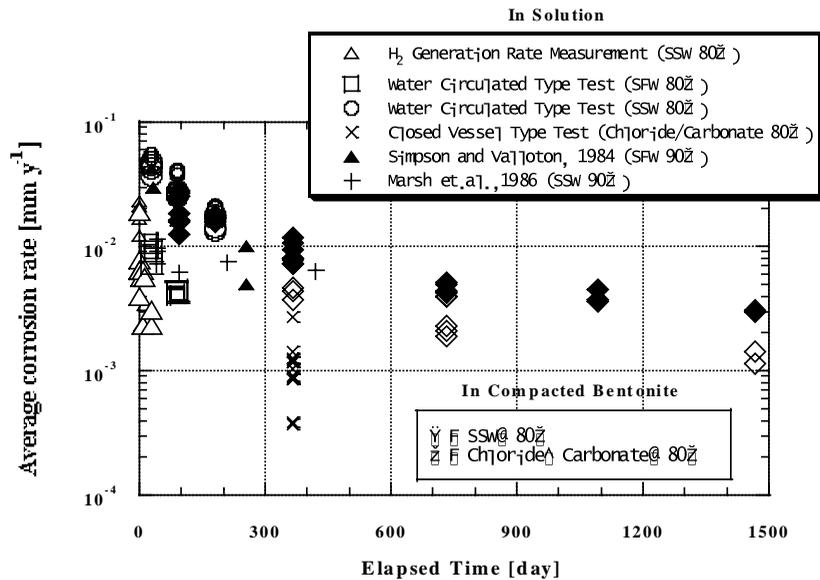
As a possible scenario, after an initial period of aqueous aerobic corrosion of a carbon steel overpack, the long-term corrosion mechanism is assumed to be anaerobic corrosion, with water as an oxidant. This leads to generation of molecular hydrogen.

The key engineering concern is that if hydrogen continues to be generated at the overpack surface without escaping, then concentration of dissolved hydrogen will exceed the solubility and a free gas phase of hydrogen will form. The subsequent concern is that if a gas phase forms, the pressure in the gas phase will continue to increase causing possible deformation to the buffer/host rock and possible episodic flow through the buffer that may also damage the buffer or expel radionuclide-bearing porewater.

Figure 1 shows the corrosion rates of carbon steel in the environments simulating reducing groundwaters. The figure also involves corrosion rates measured by Simpson and Valloton [3] and Marsh et al. [4]. As can be seen, the corrosion rate tends to decrease with time and reach a value of 0.005 mm y<sup>-1</sup> or less after 2 years [5].

Average corrosion depth for 1 000 years would be 5 mm, assuming the constant corrosion rate of 0.005 mm y<sup>-1</sup>. However, these experiments do not cover all of conditions expected under repository environment. Therefore, a corrosion rate of 0.01 mm y<sup>-1</sup> was assumed as average corrosion rate of the carbon steel overpack for the corrosion life.

Figure 1. Time dependency of average corrosion rate of carbon steel under anaerobic conditions in solution and in compacted bentonite



(SSW: Synthetic Sea Water, SFW: Synthetic Fresh Water)

A comparison of the rate of diffusive transport of dissolved hydrogen with the gas generation rate indicates that the formation of a gas phase cannot be neglected out when a conservative value of the corrosion rate ( $0.01 \text{ mm y}^{-1}$ ) is used. However, the corrosion rate tends to decrease with time and reaches a value of  $0.005 \text{ mm y}^{-1}$  or less after 2 years. When this value is used, the possibility of gas build-up is small.

Other gas generation mechanisms in the buffer include hydrogen gas generation due to radiolysis and methane gas generation following the decomposition of organic substances. However the amount of gas generated from these phenomena is relatively small.

It has been experimentally shown that even if hydrogen gas builds up, it will be released by transport through preferential pathways in the buffer material [6], and that these pathways are sealed by swelling of bentonite (Figure 2) [7].

In order to evaluate the migration of the accumulated hydrogen gas, a two-phase flow analysis (TOUGH-2 code) was carried out using a conservative corrosion rate ( $0.01 \text{ mm y}^{-1}$ ) for carbon steel overpack [8]. It was assumed in this calculation that the buffer layer and surrounding rock is initially saturated by groundwater and the corrosion of carbon steel overpack occurs immediately after repository closure. Results of this analysis indicate:

- Pore pressure, quickly returns to a value close to the underground water pressure at 30 years after closure, even if heat expansion of water is considered. The contribution of gas generation to pore pressure is minimal and the increase rate of pore pressure is 10% or less of initial pore pressure (Figure 3).
- The time for gas to reach the buffer is 3~10 years, and the starting time to penetrate into the sides of the rock mass is roughly 30~50 years. The degree of saturation after

penetration varies 96.0-98.0 % in all barriers and thus, it is found that the gas migrate through the bentonite/sand mixture replace very little amount of porewater.

Figure 2. **Repeatability of the breakthrough pressure**

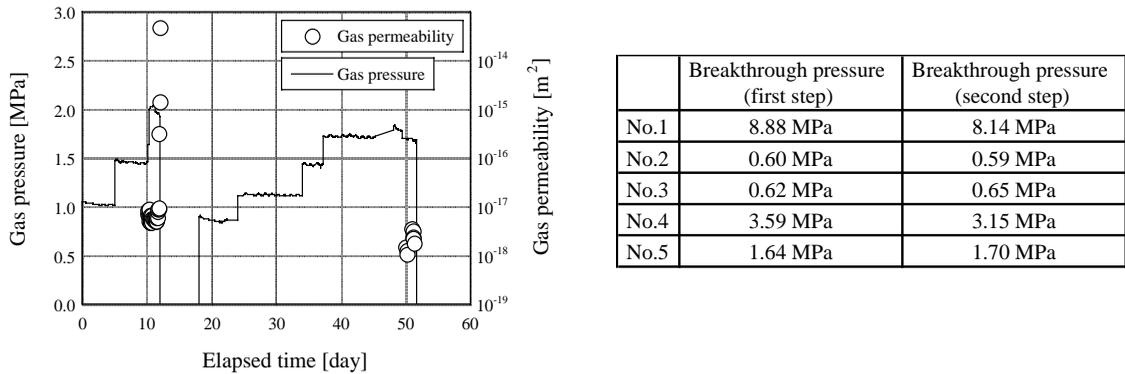
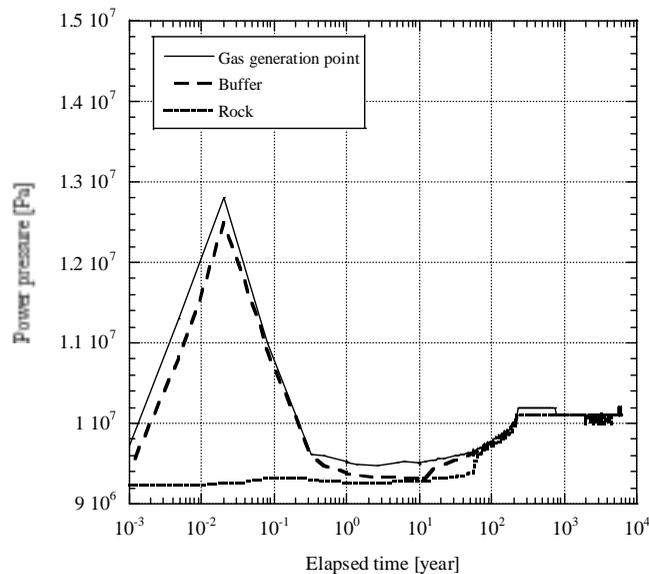


Figure 3. **Change of pore pressure with time (corrosion rate: 0.01 mm y<sup>-1</sup>)**



In view of the above, it is considered that the influence of gas generation and migration on the EBS is minimal with respect to structural mechanics, and further, that effect on radionuclides migration influenced by the discharge of porewater is negligible. However, if hydrogen gas were to build-up around the overpack, this may decrease corrosion rate, which would be beneficial for long-term safety.

#### 4. CONCLUSIONS AND FUTURE WORKS

Anaerobic corrosion of the carbon steel overpack produces hydrogen gas. The production rate may exceed the maximum diffusive flux of dissolved hydrogen in the buffer porewater and a free gas phase may be formed. However, if the corrosion rate decreases sufficiently with time and

comparable amount of dissolved hydrogen diffuses into the buffer porewater, the pressure of the free gas phase is unlikely to exceed the swelling pressure of the buffer material and to migrate through the buffer material. In addition, it has been observed in experiments that, even if gas is transported through preferential paths, the paths are sealed by swelling of bentonite.

In order to increase the confidence in gas transport analysis, new mechanistic models are under development to calculate gas migration in the buffer and fractured rock media. For this purpose, more reliable data base for the gas permeability in buffer and rocks is also planned to develop for more realistic estimation of gas accumulation and migration pathways.

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# Treatment of Two-phase Flow in Repository Performance Assessment at Yucca Mountain

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## 1. HYDROGEOLOGY OF YUCCA MOUNTAIN

The site under consideration by the U.S. Department of Energy (DOE) as a repository for high-level nuclear waste from civilian U.S. reactors is located in a semi-arid region of the southwestern U.S., near the Nevada-California border. Yucca Mountain is an unusual hydrogeologic environment. Net infiltration is low and has been estimated as being on the order of 5 mm/yr on average, resulting in an unsaturated zone of approximately 600 m thickness. Infiltration is now believed to be highly variable spatially and temporally. Potential evapotranspiration is large, far larger than the typical annual precipitation of 150-200 mm/yr. Deep percolation may occur only episodically, perhaps only once or twice every few years.

The unsaturated zone at Yucca Mountain consists of thick alternating layers of welded and non-welded tuffs, which tilt eastward at 5-7.5° and are cut by a series of primarily north-south trending faults. The welded tuffs have very low matrix permeability of order 1  $\mu\text{d}$  ( $10^{-18} \text{ m}^2$ ), corresponding to a hydraulic conductivity of  $10^{-11} \text{ m/s}$ , or 0.3 mm/yr; they have considerable fracture permeability of order 1 d ( $10^{-12} \text{ m}^2$ ). Water saturation in the welded units is in the range of 60-90 %, with matric potential in the range of 1-3 bar. The non-welded units have higher matrix permeability, of order 100 md ( $10^{-13} \text{ m}^2$ ) and much lower degree of fracturing. Average net infiltration is considerably larger than matrix permeability in the welded units, indicating that water percolation must proceed through the welded units primarily by way of fracture flow. Fast water flow through fractures, with velocities of 10 m/yr or more, is also indicated by the observation of environmental tracers that originated from atmospheric testing of nuclear weapons in the 1950s and 60s ( $^{36}\text{Cl}$ ,  $^3\text{H}$ ) at several hundred meter depth.

In early studies of the unsaturated zone at Yucca Mountain it had been suggested that due to capillary forces water should be confined to the small pores in the rock matrix. As evidence for fracture flow accumulated researchers were puzzled “how fractures could remain sufficiently saturated to act as fast paths in the face of high matrix suction” (Cook *et al.*, 1991). It has now become clear that much flow in the welded units occurs along localized preferential pathways and may also be episodic rather than steady. This limits fracture-matrix interaction, and makes it possible for water to flow relatively freely in the fractures in the presence of an unsaturated rock matrix (Ho, 1997; Pruess, 1999).

## 2. Modeling of Two-phase Processes

From the standpoint of repository design and performance assessment, the main hydrogeologic issues in the unsaturated zone of Yucca Mountain include: rates and space-time distribution of water flow, fluid residence time distributions, seepage into underground openings (drifts), fracture-matrix interaction for flow and transport, sorption behavior, redistribution of moisture and rock alteration from thermal loading, and gas flow effects.

Modeling of water and gas flow, solute transport, and thermal effects has used “macroscale continuum” concepts and methods that were originally developed in petroleum engineering and geothermal reservoir engineering. Integral equations or partial differential equations (PDEs) are formulated to express mass balances for water (liquid and vapor), air (gaseous and dissolved), and chemical species, which may be dissolved, volatilized, and sorbed. The heat balance equation includes heat transfer by conduction and convection, with sensible and latent heat effects. Mass fluxes are expressed through a multiphase version of Darcy’s law, with relative permeability and capillary pressure effects. Some modeling studies include multiphase diffusion and hydrodynamic dispersion, and migration of colloids. Different approaches have been used to model fracture-matrix interactions, including effective continuum, double porosity, dual permeability, and multiple interacting continua models. In some cases simplifying approximations have been employed, such as Richards’ equation for unsaturated seepage and a single-phase gas flow equation for pneumatic tests at ambient conditions.

Under most circumstances of interest the PDEs for multiphase fluid and heat flow in unsaturated zones are highly nonlinear and must be solved by numerical techniques. This requires discretization of the continuous space and time variables, implying limited space and time resolution and averaging. Current flow and transport models for the unsaturated zone at Yucca Mountain have been summarized in a special issue of the *Journal of Contaminant Hydrology* (Bodvarsson and Tsang, 1999). These models have become very elaborate, employing on the order of 50 000 grid blocks or more to represent nonisothermal multiphase flow in three dimensions with spatial resolution of order 100 m. Calibration of the models uses comprehensive data sets and constitutive relations obtained from field and laboratory measurements, including water saturations and matric potentials, matrix and fracture permeabilities and porosities, characteristic curves (relative permeabilities and capillary pressures), temperatures, gas pressures, locations and amounts of perched water, chemical composition and isotopic abundance of groundwaters and minerals, fault locations, and density and orientation of fracturing. The calibration process has been partially automated through the use of inverse models (Bodvarsson and Tsang, 1999).

In spite of the sophistication of the large-scale volume-averaged models, serious questions remain about their conceptual basis and validity. The volume averaging implied by finite difference techniques has a firm basis when applied to diffusive processes described by parabolic PDEs, in which averaging occurs by means of physical mechanisms within the flow system. Examples include gas flow in fracture networks, molecular diffusion of solutes, and heat conduction. However, water flow in unsaturated fracture networks of high permeability is described by a PDE that is essentially hyperbolic in character. In this case, intrinsic averaging mechanisms are weak, and water flow may occur predominantly along localized preferential pathways. Volume-averaged descriptions may produce erroneous results; for example, it was shown that downward seepage of water under gravity force may proceed faster in media with lower “average” permeability (Pruess, 1999).

Alternative conceptualizations have been proposed to model water seepage in thick unsaturated zones of fractured rock, including the “weeps” model of Gauthier *et al.* (1992), and transit time and transfer function models (Chesnut, 1992).

The Yucca Mountain project entered a new phase with the completion of the Exploratory Studies Facility (ESF) in 1997. A tunnel of almost 8 km length, the ESF has made possible a comprehensive *in situ* testing program, including water infiltration, solute transport, pneumatic tests, and heater experiments. Design and analysis of these tests is presenting challenges to mathematical modeling, and has led to rapid progress in understanding of two-phase flow phenomena on different scales.

### 3. TOTAL SYSTEM PERFORMANCE ASSESSMENT

In December 1998, DOE delivered to the U.S. Congress and the President the so-called "Viability Assessment" (VA), which in five volumes presents a comprehensive summary of current understanding of the hydrogeology of Yucca Mountain, as well as detailed information on repository design and performance assessment (CRWMS M&O, 1998). The VA concludes: "Based on the scientific study of Yucca Mountain, DOE believes that the site remains promising for development as a geologic repository. However, uncertainties remain about key natural processes, the preliminary design, and how the site and design would interact." The key safety attributes of a Yucca Mountain repository emphasized by DOE include (1) limited water contacting waste packages, (2) long waste package lifetime, (3) low rate of release of radionuclides from breached waste packages, and (4) radionuclide concentration reduction during transport from the waste packages (CRWMS M&O, 1998). Quantitative evaluation of future repository behavior, and demonstration of compliance with regulatory standards, is accomplished through a process known as "Total System Performance Assessment" (TSPA). The regulatory standards as promulgated by the Nuclear Regulatory Commission (NRC) and the Environmental Protection Agency (EPA) themselves are in flux. An earlier release-based standard was remanded in 1992; a dose- or risk-based standard is in preparation but has not been officially promulgated.

The interconnected processes to be addressed by TSPA have been succinctly stated in a report on the Viability Assessment that was prepared by U.S. Geological Survey (USGS) scientists to the Director of the USGS (Hanks *et al.*, 1999): "Climate and climate change in the vicinity of Yucca Mountain determine precipitation on the mountain, some of which infiltrates into it. This infiltration drives percolation of water in the unsaturated zone, that part of the mountain mass above the water table, to greater depth. If the rate of percolation is sufficiently high at the repository level (approximately 300 m beneath the Yucca Mountain crest), water seeps into the emplacement drifts (waste-storage tunnels). This seepage accelerates corrosion of the containment canisters and then the interior cladding about the radioactive wastes, exposing them to the seeping water. This water now becomes the vehicle for dissolving and transporting exposed radionuclides out of the emplacement drifts into the unsaturated zone below and, finally, to the water table, which is currently 300 m beneath the repository. Flow in the saturated zone dilutes the concentration of dissolved and colloid-bound radionuclides first reaching the water table, but may allow them access to the biosphere downstream from Yucca Mountain, either by natural processes that bring the contaminated groundwater to the surface or because of human activities, such as ground-water pumping. Radioactive materials emanate harmful radiation, and the "dose rates" of this radiation and, perhaps, the probabilities of exceeding these dose rates must be less than certain amounts at certain distances and times yet to be specified by the U.S. Environmental Protection Agency."

It is clear that, to address all of these phenomena, TSPA must integrate a complex system of models for all of the different natural and engineered repository components and aspects, including the unsaturated and saturated zone, mined openings, waste packages, and "external" conditions, including present and future climates, and various potential disruption scenarios, such as seismicity, volcanic hazards, human interference, and nuclear criticality. Probabilistic Monte-Carlo techniques and alternative conceptual models are employed to deal with uncertainty. Unsaturated zone process models

used in TSPA are implemented through several different simulation codes developed by DOE National Laboratories, as follows. Ambient mountain-scale unsaturated flow is modeled with the TOUGH2 code, employing a 3-D dual permeability model and an approximation of steady flow. The inverse code iTOUGH2 is used to aid in model calibration. Solute transport in the TOUGH2-generated flow fields is modeled with the FEHM code, using particle tracking. Ambient drift-scale flow is represented with 3-D heterogeneous models, also implemented through TOUGH2. Mountain-scale thermally-driven flows are modeled with TOUGH2, using 2-D sections extracted from the 3-D ambient flow model. Thermally-driven flows on the drift scale are modeled with the NUFT code, using 1-D, 2-D, and 3-D models. Near-field geochemistry in the heated regions is modeled by zero-dimensional equilibrium batch calculations, using the EQ3/6 speciation and reaction path code.

#### **4. CURRENT ISSUES IN TWO-PHASE FLOW**

In commenting on DOE's Viability Assessment (1998), the staff of the Nuclear Regulatory Commission (NRC) noted that "The data and models used in the VA to calculate the quantity and chemistry of water dripping on WPs [waste packages] are inadequate to describe the process and extent of potential dripping under ambient and thermally-altered conditions." The question of seepage into drifts, a precondition for water to contact waste packages, was identified as the most important issue in a USGS report (Hanks *et al.*, 1999), and was also raised as an important topic by the Nuclear Waste Technical Review Board (NWTRB). In assessing the uncertainty of various factors on postclosure repository performance, DOE's Viability Assessment rated seepage into drifts as having the highest significance. A large number of additional two-phase flow issues are identified in the VA, including infiltration rate under present and future climates, localized flow channeling, role of perched water, stability of seepage locations, scale dependence of parameters, episodic percolation, retardation effects from sorption and matrix diffusion, colloid transport, condensate buildup in fractures, coupled processes, hysteresis of hydrologic properties, thermal alteration of hydrologic properties, and geochemical effects on flow and transport.

To address these issues, DOE is currently pursuing an intensive program of hypothesis-based testing, mostly using *in situ* experiments in the ESF. Field studies are focusing on water flux, solute transport, heater tests, pneumatic tests, and geochemical sampling. An important objective is to obtain property measurements for fractures, rock matrix, and faults. Natural features are being sampled and characterized to explore and confirm conceptual models. Experimental studies are closely integrated with 3-D modeling to investigate seepage exclusion from drifts, water flow and solute transport in fractures, fracture-matrix interactions, hydrologic behavior of faults, and hydrothermal alteration.

#### **5. CONCLUDING REMARKS**

Yucca Mountain represents an unusual hydrogeologic environment, with many attributes that are favorable to waste isolation. Hydrogeologic processes are subtle and complex, and are difficult to characterize and quantify, as had been anticipated by USGS scientists who originally championed the site. A comprehensive program of site characterization, testing, and modeling has led to a much improved understanding of two-phase flow in the unsaturated zone of Yucca Mountain. Mechanistic modeling of flow and transport remains difficult because processes occur on a broad range of space and time scales. Uncertainties in processes and parameters are being reduced through a vigorous program of *in situ* testing, that is integrated with flow, transport, and geochemical modeling. Alternative conceptual models may be employed to achieve a more robust performance assessment (Pruess *et al.*, 1999).

## 6. ACKNOWLEDGEMENT

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# **The Treatment of Gas in the Performance Assessment for the Disposal of HLW and MLW in Boom Clay**

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## **1. INTRODUCTION**

In Belgium the Boom Clay is studied as potential host rock for the geological disposal of high level (HLW) and intermediate level radioactive waste (ILW). The Boom Clay has been chosen because of its very low hydraulic conductivity ( $2 \cdot 10^{-12}$  m/s). Consequently, transport of contaminants in pore water of the Boom Clay is diffusion-controlled whereas advection has a negligible contribution to the overall migration. Also the transport of dissolved gas is very limited. Therefore, when gas is generated this can easily lead to a gas pressure build-up and thus to a safety concern. In the following sections the experimental evidence about gas generation and transport in Boom Clay are briefly summarised, then the approach used to treat the gas issue, followed by the assessment of the gas generation and the assessment of its potential consequences.

## **2. EXPERIMENTAL EVIDENCE**

### Gas generation by anaerobic corrosion

The gas generation experiments performed in the framework of MEGAS (Ortiz 1997, Volckaert 1995) and PROGRESS (Rodwell 2000) projects have shown that in Boom Clay slurries carbon steel corrodes at a rate probably lower than  $1 \mu\text{m}/\text{year}$ , while stainless steel of the AISI 316 L type shows hardly any corrosion. Initially the produced gas is hydrogen, but after a few days up to a few months the hydrogen is converted to methane by microbial activity. The above corrosion rates are within the range obtained from independent laboratory and in situ corrosion experiments. The microbial conversion of hydrogen to methane was also confirmed by independent experiments.

### Gas diffusion through Boom Clay

Gas diffusion experiments on saturated Boom Clay cores have been performed with hydrogen and methane. The obtained apparent diffusion coefficients for hydrogen were lower than expected, i.e. close to the value for tritiated water. Their spread was rather large, which could be due to the conversion to methane by bacteria. For methane a diffusion coefficient of about  $10^{-10} \text{ m}^2/\text{s}$  was obtained.

### Gas flow through Boom Clay

Both in situ and laboratory tests have shown that there is a strong coupling between the geomechanical stress distribution and the gas breakthrough pressure: gas breakthrough occurs when the gas pressure reaches the value of the minor component of the local total stress tensor and in the direction orthogonal to it. The degree of desaturation at breakthrough is very small (maximum a few

percent of total porosity). The relation between gas flow and gas pressure is highly non-linear and cannot be represented by the extended Darcy's law. After stopping the gas injection and removal of the gas in the piezometer, the pathway created by the gas breakthrough seals quickly and the original permeability is restored. This "self-healing" capacity of the Boom Clay was also confirmed by a HTO migration test performed after a gas breakthrough test.

### **3. METHODOLOGY/APPROACH FOR TREATING THE GAS ISSUE IN PA**

The methodology applied for the evaluation of possible gas effects on the performance of the repository system consists in the following steps:

- Assessment of the total gas generation potential and the gas generation rate as function of time for a given type of waste and disposal concept.
- Assessment of the possible transport of dissolved gas through the Boom Clay at a gas partial pressure ( $P_g$ ) equal to the local hydrostatic pressure (2.2 MPa) and to the total pressure (4.4 MPa) at 225 m depth.
- Determination of possible "gas scenarios" on the basis of a comparison of cumulative gas generation and cumulative gas transport:
  - If gas generation is lower than diffusive gas transport at  $P_g = 2.2$  MPa then only dissolved gas is present.
  - If gas generation is larger than diffusive gas transport at  $P_g = 2.2$  MPa then gas bubbles form and expulsion of water occurs with a rate equal to the grow rate of the bubble.
  - If gas generation is larger than diffusive gas transport at  $P_g = 4.4$  MPa then gas breakthrough occurs.
- Description of the expected radionuclide transport in each of the above cases.
- Assessment of possible consequences and comparison with normal evolution scenario.

### **4. COMPARISON OF GAS GENERATION AND TRANSPORT OF DISSOLVED GAS: SOME EXAMPLES**

Fig. 1 compares the estimated amount of gas produced by anaerobic corrosion of the disposal tube, the overpack and the vitrified HLW canister in a "PRACLAY" type design (Verstricht and De Bruyn 2000) and the cumulative diffusive gas transport for a realistic range of diffusion coefficients and for a gas partial pressure of respectively 2.2 MPa and 4.4 MPa. In this calculation a conservative corrosion rate, for 316 L stainless steel, of 0.05  $\mu\text{m}/\text{year}$  was supposed. It is clear that if hydrogen is completely converted to methane all the methane can be transported by diffusion and no gas bubbles will be formed (Fig. 1). In case the hydrogen is not transformed to methane, gas bubbles could form if the diffusion coefficient is lower than  $10^{-10}$   $\text{m}^2/\text{s}$ . However, the gas pressure (Fig. 1) will not reach the lithostatic stress.

In Fig. 2 a similar comparison is given for a specific type of intermediate level waste. For this waste type both hydrogen and methane generation will lead to the formation of gas bubbles but gas breakthrough is more probable in case of hydrogen than in case of methane generation.

Figure 1. Cumulative hydrogen (left) and methane (right) production and transport for vitrified HLW disposed of in a “PRACLAY” type repository in Boom Clay

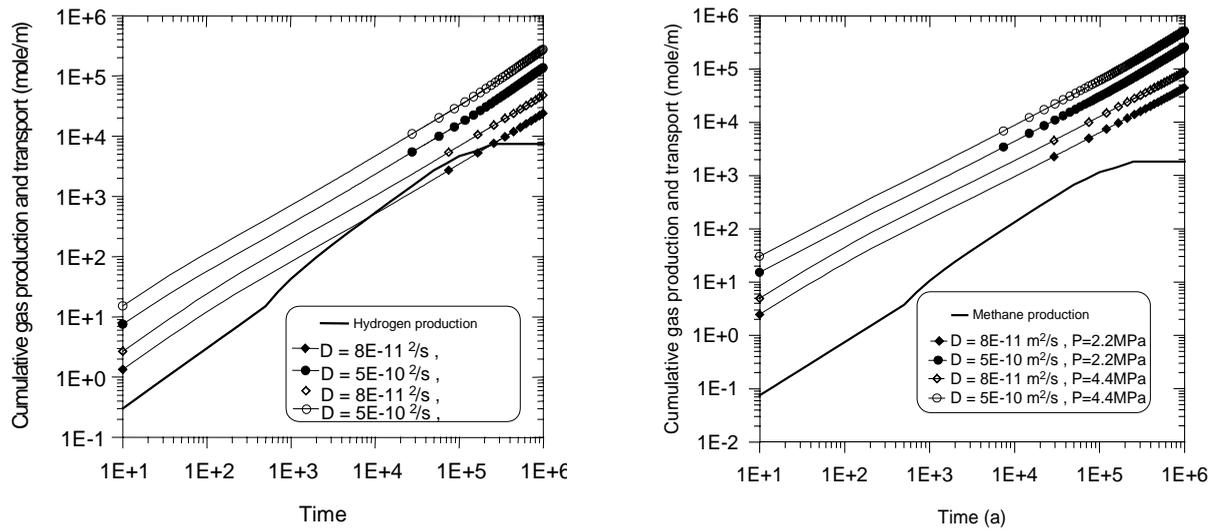
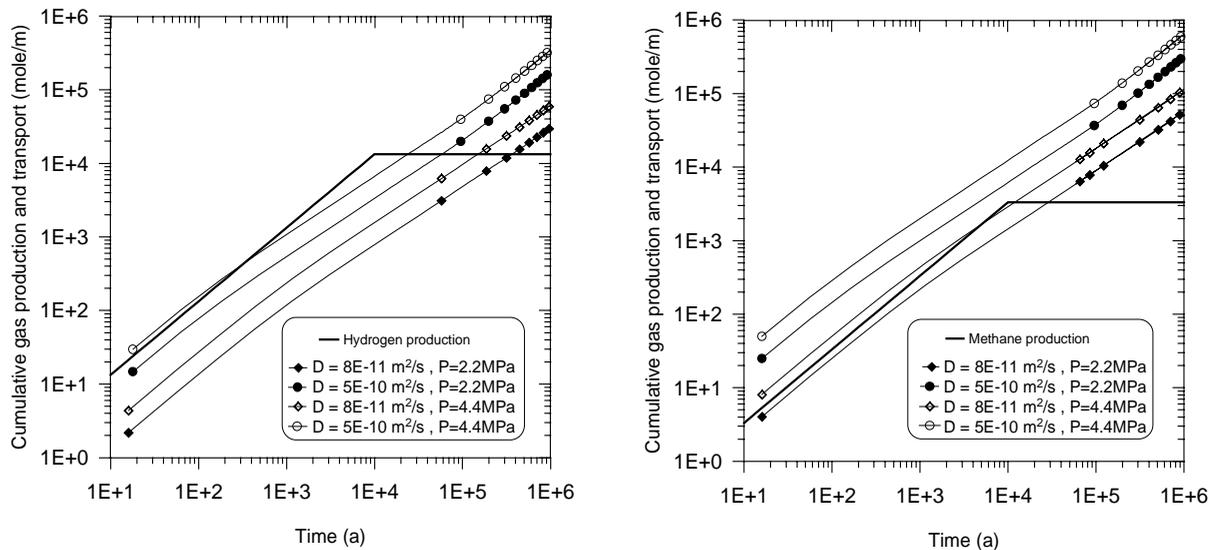


Figure 2. Cumulative hydrogen (left) and methane (right) production and transport for the disposal of a ILW type in a repository in Boom Clay



### Consequence analysis

The main radiological risk of gas generation is not due to the contamination of the gas itself but due to the advective transport of radionuclides in the pore water expelled by the gas. Gas bubbles formed in the near field of the waste may displaced contaminated water at a rate equal to the grow rate of the gas bubbles. Two cases are possible:

- the gas bubbles grow slowly, water is displaced at the same rate and flows in the Boom Clay according to Darcy’s law;
- the gas bubbles grow quickly, the pore water cannot be transported by Darcian flow only and the pore water is pressurised so quickly that a creation of preferential pathways occurs.

It is conservatively supposed that gas bubbles will grow until their pressure reaches 4.4 MPa, because then gas breakthrough through the Boom Clay will occur. Probably gas breakthrough will occur already at a lower pressure through the excavation disturbed zone or in the near field.

While in the first case contaminated water will be transported through the full effective porosity of the Boom Clay, in the second case it will only be transported through the effective porosity of the created pathways. The latter is probably only a very small fraction of the porosity used in the first case. The only experimental information we have about the effective porosity in case of creation of preferential pathways is the very low desaturation measured in the gas breakthrough experiments.

In the radionuclide transport simulations we assumed that the near field pore water is in equilibrium with the waste and thus with maximal radionuclide concentrations. The simulations show that, for the scenario with water expulsion by Darcian flow, the increase in radionuclide flux at the Boom Clay-aquifer interface is only important for I-129 (non-retarded). In the case of water expulsion through preferential pathways with an effective porosity of 1%, the increase is also important for retarded radionuclides. However, the creation of preferential pathways would occur very soon after closure of the repository when the near field pore water is probably not yet saturated with radionuclides.

## 5. CONCLUSIONS

Gas generation most probably is not an important issue in case of the current "PRACLAY" concept for vitrified HLW disposal in the Boom Clay owing to the very low corrosion rate of the selected stainless steel. In case of ILW, gas pressure build-up is very probable, especially when the waste is packed in carbon steel or contains important quantities of carbon steel. In this case an enhanced radionuclide transport through the Boom Clay is possible. The simulations of this enhanced transport show that only in case of transport through preferential pathways, created by an increase in hydraulic pressure, the increase in radionuclide flux is considerable. However, many conservative assumptions were made in this assessment. For a more realistic assessment it would be necessary to couple the gas generation process directly with the gas, water and radionuclide transport processes in the calculations. Also the determination of the properties of these preferential pathways would be needed. An other possibility is the development of a concept that allows for a controlled gas release with minimal water displacement.

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## **SESSION III**

### **REGULATORY POINT OF VIEW AND EXPECTATIONS**

Chairman: Erik Frank (HSK, Switzerland)



# **Gas Generation in High Level Waste Geological Disposal: Elements for Integration in a Safety Strategy**

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## **1. INTRODUCTION**

Gas generation in high level nuclear waste repositories has become a subject of concern during the past decade. Important work has been done in evaluating the possible sources and quantities of gas likely to be produced in the lifetime of the repository, together with the development of experimental and modeling capabilities for assessing the possible consequences of such generation [1,2]. As a general picture of the evaluations that have been carried out so far, it can be drawn out that a gas pressure build-up is possible in the infrastructures of a repository. Though the gas production rates are generally low, the isolation capacity of the engineered barriers and host rock that are looked for in order to confine radionuclides are sufficiently high to prevent gas from freely dissolve and diffuse in the media. The gas formed is mainly hydrogen from metal corrosion and water radiolysis, and possibly carbon dioxide from bacterial degradation of organic matter contained in the wastes. The formation of a gaseous phase in a repository is therefore bearing potential risks, of “classical” nature (such as for example fire hazards in mined structures) or of radioactive nature (contamination of operated vaults by volatile activity, gas being a vector of transport). It also bears a potential for damaging engineered systems in the long term and create pathways of possible faster transport characteristics for released activity. There is consequently a need for thoroughly assessing the potential effects of gas generation in view of building the safety case of a repository, and of implementing a safety strategy that accounts for a minimisation of these effects. Elements for establishing a framework in which such a strategy may be developed is given hereafter. This framework is an illustration, focused on gas generation problematic, of the incremental process needed to progressively achieve confidence in the safety of geological repositories.

## **2. FRAMEWORK FOR ESTABLISHING A SAFETY STRATEGY WITH REGARD TO GAS GENERATION IN GEOLOGICAL DISPOSAL**

As it was just mentioned, assessing the potential effects of gas generation in a repository and implementing a sound strategy that accounts for the minimisation of these effects is part of an incremental process. It is indeed recognised that achieving an optimised treatment of the problem will presumably require several iterations between steps of knowledge acquisition, steps of evaluation and steps of repository conception. It is however believed that these iterations can be organised around a scheme that can be repeated as many times as necessary. Starting from a given concept, it may therefore be proposed that the operator addresses the question of gas generation as follows:

- for each potential risk associated to gas generation, assemble the necessary knowledge and perform an evaluation enabling a clear identification of the situations for which the considered risk is effective, and quantify this risk;

- implement a strategy (ies) allowing an elimination or a reduction of the risks based on the application of a “defence in depth principle”;
- evaluate, through integrated performance assessments, the pros and cons of the strategy (ies) proposed and verify that the selected strategy does not lead to unacceptable consequences.

The first step of work constitutes the background analysis necessary for carrying out the safety assessment of the repository, with regard to gas generation. During this step must first be established a list of the potential risks associated to gas production in the repository, during the successive phases or transitory states that it will encounter (operational phase, reversibility phase, post closure thermal phase, unsaturated state, saturated state). These risks, which could be defined as the occurrences of phenomena that would affect the safety of the repository, may be of direct nature (i.e. due to the gas phase itself such as risks of explosion or contamination by transport of volatile nuclides) or indirect nature (i.e. due to the effects of gas on the other barriers, such as for example damages on waste packages and engineered barriers, fracture opening or enhancement of temperature leading to barrier performance weakening).

An evaluation of the amplitude of each of the risks identified must then be conducted, using appropriate models and data. The types of evaluation required may broadly follow two goals on which emphasis is successively given as the repository project develops. At first, the evaluations are to provide a quantified estimation of the risks linked to gas generation in repositories, so as to assess the effectiveness of the proposed design against these risks. The evaluations carried out for this purpose should be “reasonably conservative”, in the sense that the parameter values should be chosen on the «conservative side» when the range of possible values is large, but also that the mechanisms accounted for in the evaluation should be possibly described without oversimplification. This is meant to allow a pertinent comparison of various dispositions of conception when several design strategies can logically be applied. At a later stage, when a strategy has finally been adopted, a demonstration of the conservative nature of the evaluations carried out will have to be demonstrated for dimensioning purposes. As for other processes important for safety, the gas production and its effects will then have to be described through the use of models and parameter values that are proven to give an “envelope” estimation of the scenarios concerned.

It is of course necessary that the above mentioned evaluations be supported by all necessary experimental tests, for pertinent data acquisition and model validation. Again, this support work may focus on different aspects of the problem along the course of project development. In a first step, production mechanisms and parameters controlling the fluxes of gas in the repository will have to be quantified through experiments at sample or mock up scale. At a later stage, together with the research work needed to reduce uncertainties on the data and mechanisms to which evaluations are sensitive, tests aiming at representing more accurately processes and events occurring at “industrial” scale, such as for example the estimation of barrier defects which require a practice of underground works, will be necessary.

This step of evaluation done, the operator should make benefit of the necessary background information to implement a strategy for minimising the risks associated to gas production. This strategy should be based on the application of a defence in depth principle, implying that all necessary efforts should be made to prevent the gas phase from forming, control and limit the occurrence of two phase flow and pressurisation if prevention cannot be achieved, and possibly limit the effects and consequences of gas pressure build-up and flow. This may be implemented by the definition and application of “lines” of defence, which may be “active” or “passive” barriers, aiming at preventing gas build-up and flow or limiting their consequences on safety, but also procedures and corrective actions, aiming at controlling the phenomena and implement necessary actions so as to get back to

normal functioning conditions. The types of lines of defence to adopt depends on the phase considered and the accessibility of the repository infrastructures. It is also necessary to implement a strategy that accounts for a possible failure of the primary lines of defence and proposes dispositions for limiting the consequences of such failure, since the repository should not rely on lines of defence of which single loss of function may jeopardise the safety of the facility.

The task of the operator is thus to clearly define the «lines» of defence that may be implemented against gas production effects, with due regard to all plausible situations that the repository may encounter during the phases of its life, and assess their efficiency in a new safety assessment. It is however important to recognise that some dispositions may have counterparts on other aspects of the safety of the repository, in particular since minimisation of a pressure build-up would tend to require a facilitated dilution and diffusion of gas, as opposed to favourable confinement properties generally looked for in order to prevent radionuclide dissemination. A clear description of such counterparts, for each of the lines of defence proposed against each of the risks associated to gas production, must therefore be given by the operator and also quantified in a new safety assessment. It is probable in the end that several strategies may be proposed, giving emphasis on different aspects of the repository safety.

The third step of the iteration consists mainly in comparing the efficiency of the various designs that can be derived from the safety strategies implemented by the operator, by means of integrated performance assessments. This step of evaluation is required to estimate the fluxes of radionuclides (in activity or mass) and toxic substances that may be transported through the successive barriers up to the outlets of the repository. Such evaluation, that must consider normal evolutions of the repository together with altered scenarios, should enable a clear identification of the advantages procured by the various elements of design and for which type of situations these advantages are effective. These are important elements of decision for finally achieving a choice for implementing a strategy likely to be “optimised” on the important aspects of the repository safety. An integrated performance assessment will also be needed to verify that the radiological and toxic impact of the repository does not lead to unacceptable consequences on human health and environment, and may also ultimately be used to check whether additional safety margins should or may be obtained through reasonably feasible adjustments of the chosen strategy.

Some short examples are given hereafter to illustrate some aspects of the approach that has just been described.

### **3. SOME EXAMPLES OF STRATEGY IMPLEMENTATION PROCESS AGAINST GAS POTENTIAL EFFECTS IN REPOSITORIES**

#### **3.1 Operational and reversibility phases**

During operation of the repository and a phase during which an easy retrieval of the waste packages placed in the vaults is to be ensured, implying that all infrastructures of the repository, except possibly vaults, are not yet backfilled and can be visited, the risks of explosion, due to a burst of hydrogen in air filled galleries, and the dissemination of radioactivity in the mined infrastructures must be strictly controlled and avoided.

The first phase of assessment and quantification of these risks requires primarily a thorough evaluation of the gas production rates. During the considered phases, water radiolysis is expected to be largely the dominant process. The rates of hydrogen production, due to radiolysis of water initially contained in the wastes (mainly cemented IL wastes of type B, according to the French nomenclature), must be estimated. Additional sources must also be quantified, such as hydrogen produced by

radiolysis of water in materials surrounding irradiating waste packages (type C and spent fuel) as well as hydrogen produced by radiolysis of bitumen matrixes. Though radiolysis may be a complex phenomenon to describe, the estimations of gas production can be expressed through a radiolytic ratio of production, of which consistency can fairly well be verified by measurements carried out on waste packages already produced. Uncertainties on production can therefore probably be estimated with rather good confidence, at least for the main sources of gas that are presumably cemented wastes. However, this might not be the case for the bacterial degradation of organic wastes, in particular bituminised effluents. This mechanism has indeed a high potential for carbon dioxide production, at rates of even higher value than radiolytic production of hydrogen, but the conditions of bacterial development and breeding are very uncertain, as observed in laboratory experiments. Whether this potential for producing large amounts of gas remains a question that must be addressed in safety assessments of geological repositories, and deserves probably additional research work.

Once the rates of production have been established with reasonably conservative data, the possible pressure rise in the waste packages and on the vault plugs and/or the fluxes of gas reaching the galleries must be estimated. The amount of radionuclides likely to be carried by the gas vector, in particular the volatile or labile content of the waste packages must also be assessed. Such assessment necessitates a good knowledge of the amount of void spaces available in the waste package and in the vault, as well as the leakage rates of the waste containers and the vault plug that possibly depends on the pressure rise. If data from the experience acquired on container tightness and from sample tests on plug material performances can be used in early iterations, confirmation of such performances at "industrial scale" will be necessary, especially for real size plugs that might behave quite differently than at sample scale. If a confinement strategy during the operational and reversibility phases is chosen and that the evaluations show that a significant pressure rise might occur, such confirmation would be required rather early in project development since it would condition important elements of design.

This first step of safety assessment will draw a first picture of design adaptation to the effects of gas production and will allow to focus on the areas of uncertainties remaining as well as on the weak points for which new elements of a safety strategy must be developed. Some preliminary remarks on this subject can however be given here.

A first line of defence against gas production hazards would concern the prevention of gas production. Attended the fact that internal water radiolysis is probably the main source of hydrogen produced by B wastes, a favourable factor would be to limit or avoid the amount of free water contained in the waste or in its embedding matrix. This however would only apply for wastes to be produced, and the absence of cemented matrix, though favourable with regard to gas production, would raise questions on other aspects of safety (unblocked wastes of poor mechanical resistance, high void ratios, ...). Concerning gas production from external water radiolysis, additional shields could be proposed (if this mechanism is proved to generate significant amount of gas) so as to lower the g field, but such shields, probably of metallic nature may induce several disadvantages (weight, manipulation, vault over sizing, chemical perturbation, other source of gas by corrosion, ...). Finally, the prevention of bacterial gas production from organic wastes, if practicable, requires to act on parameters that are not yet well established and need further research work. It is therefore doubtful at the time being that advantageous solutions of prevention may be developed for radiolytic gas potentially produced by the wastes existing at present time. Only a period of preliminary storage could significantly reduce the amount of gas produced in cemented wastes since radiolytic ratios may decrease of a factor 20 during a 50 year period.

Concerning the limitation of the fluxes of gas and associated activity released outside the vaults, one may consider two opposite strategies: either confine the gas in wastes and vaults, either evacuate the gas produced in a controlled manner. The first strategy is manageable if sufficient

confidence can be obtained that gas will not be produced at such rates that it could make containers and vault plug fail, and that former equipment can be realised with very low probability of defects. The soundness of such strategy requires therefore a validation through industrial practices. The second strategy, that can be achieved through the realisation of vents and filters in containers or plugs, offers the advantage of avoiding sudden bursts of gas that may possibly occur after plug breaking, but requires a strict control of the amount of gas and activity released as well as operations of maintenance. However, whatever strategy is chosen, it is clear that control and dilution of gas through an adequately designed ventilation system are probably most necessary lines of defence to implement against gas production hazards during operational and reversibility phase. In particular, a monitoring of gas pressure in the vault should be considered so as to avoid incidental releases in galleries by means of possible corrective actions aiming at recovering conditions of “normal functioning” of the vault. The conditions of ventilation should be studied so that a possible accidental release of radioactive gas does not spread to other sectors of the repository. The possibility of sectorising the repository and nuclearise the ventilation should be studied. One should however recognise that such dispositions may lead to oversize the ventilation system which may induce drawbacks for the long term safety of the repository (multiplication of the number of shafts and galleries, position of the shafts, size of the shafts and galleries, that may induce faster pathways for migration of radionuclides...). Such choices need therefore to be carefully balanced by a reasonable weighing of the likeliness and amplitude of the risks associated to all scenarios concerned.

A phase of integrated performance assessment may help to compare the advantages of adopting a strategy in favour of a limitation of operational risks, against the possible drawbacks of such a strategy with regard to the long term consequences of the repository (the “operation orientated” design may be not optimised with regard to long term radionuclide transport processes). It is however doubtful, if the operational hazards mentioned above are proved to be likely, that the long term drawbacks induce a choice of strategy that are not in favour of dealing with operational hazards. An adequate quantification of the risks occurring during operational and reversibility phase is therefore of prime importance since that the elements of design required by operational safety will have a significant weight in choosing a strategy.

### **3.2 Post closure phase**

One of the main effects of gas generation after closure of the repository, against which efforts of prevention and limitation should be possibly made, is the occurrence of damages on the near field components of the repository, due to overpressures induced by gas build-up. In particular, the degradation of engineered barriers (surrounding the waste packages) and seals as well as the possible enhancement of excavation damage zone (EDZ) conductivity must be assessed, in order to estimate whether preferential pathways can be created.

The assessment of such effects requires first to have a rather precise estimation of the corrosion rates of the metallic components (iron based) of the repository, since gas build-up after closure of the repository mainly depends on this mechanism. If orders of magnitude are now currently available for corrosion of metals in anoxic conditions, representative of various underground water compositions, one should however pay attention to the possible synergetic effects (temperature, presence of carbon dioxide, microbial induced corrosion) that may induce significant increases in the rates of hydrogen production. This phase of assessment must therefore lead to a “reasonably” conservative estimate of the production rates. A modelling of the pressure rise and consequent flow of gas in the near field must then be performed, accounting for the main factors that may govern gas evolution in the repository. According to the host rock considered, these factors may have different weight: hydro-mechanical parameters and diffusion should presumably be of prime importance in clay based formation as mechanical parameters and convective flow processes are likely to play a major

role in fractured crystalline rocks. Whatever the case is, a precise modelling of such effects of gas production is a complex task and calculations will face problems of approximation due to processes that are not yet fully understood. If orders of magnitude of pressure rise can be obtained, the credibility of the assessments for mechanical damages induced and the subsequent flow of gas in near field certainly require experimental evidence. The characterisation of engineered barriers and seals tightness, the hydro-mechanical properties of EDZ (fracturation, flow, self healing...) must therefore strongly support the assessment. In particular, scale effects deserve attention since the extrapolation of laboratory or mock up investigations to industrial scale are not straight forward. *In situ* testing of (“close to”) real size components of the repository must therefore be carried out in appropriate time during the course of development of a repository project.

This step(s) of assessment made for a given concept, it is expected from the operator to possibly adjust its strategy so as to increase safety with regard to gas production production. In the long term time frames considered for a repository, lines of defence to be opposed to this phenomena can only be passive since safety, cannot rely on the possible maintenance or intervention of man on the repository system. Considering the prevention of gas build-up, the main action that can be carried out on the source of gas is to limit the amount of metallic components introduced in the repository, or to modify the nature of these components (replace carbon steel by stainless steel or “passive” alloys). Besides the economical consequences of such choices, their pertinence must be assessed against the possible negative effects of metal limitation or modification. In particular, the feasibility of supporting underground works without metallic equipment (or with removable one), or its replacement by concrete supporting equipment, possibly inducing complex chemical interactions with host rock, must be assessed. The removal of overpacks, protecting wastes against early releases during the thermal phase, or the occurrence of pitting corrosion in alloys that may also induce faster inlet of water in the waste, must also be assessed. It is expected from the operator to provide best estimate results in doing so, so as to allow optimisation to be made on clear, well informed bases. If a gas build-up cannot be avoided, another aspect of defence in depth is to provide a design that enable a limitation of the pressure rise, below threshold pressures corresponding to the breakthrough of gas in the various components of the repository. This implies to provide for gas, inside the vaults or galleries of the repository, sufficient porous space so as to enhance its dilution and diffusion in the infrastructures and to allow its expansion between engineered barriers. Such strategy implies, as was said before, a good knowledge of the threshold pressures of the components of the repository so as to adjust the design. It also requires to clearly define the time frames over which the engineered barriers and seals must keep their initial function of confinement so as to define the volumes made available for gas to expand in vaults and in galleries, and provide a correct location of seals. According to the function given to each component and the likeliness of their failure due to gas production, the level of redundancy of engineered features must also be defined. It is of course obvious that the limitation of gas pressure in a repository requires technical solutions that are not likely to be preferred for the confinement of radionuclides (enhancement of porous, connected space for gas expansion against tight material to oppose underground water flow). In particular, the possible enhancement underground infrastructures sizes may, in some formations, induce enhanced damage zones that are potential preferential pathways, and the location of seals might preferably be governed by the need to treat site fracturation rather than the need to allow sufficient void volume between seals. Such clear possible disadvantages for safety need to be thoroughly assessed so as to optimise the design of the repository. A good knowledge of site structural features and underground testing of the mechanical behaviour of the host formation is a pre requisite to find this optimum.

Provided the fact that the previous studies will probably lead to propose several strategies for designing a repository, each having pros and cons with regard to the various phenomena that must be accounted for in a safety assessment, an integrated performance assessment will have to be carried out, so as to compare, in term of fluxes of radionuclides in near and far field or doses, for which situation and during which time frame a given strategy procures a significant benefit. Regarding the problem of

gas production after closure of the repository, this comparison will in particular concern the consequences of early releases during the thermal phase (due to a limitation of metallic protection of the waste form so as to avoid gas production) against the consequences of possible damages in near field leading to the occurrence of preferential pathways (if gas production is not preferably limited).

The implementation of the proposed approach will in the end allow the operator to make a clear, well balanced choice of strategy for designing a repository, and to assemble all elements for dimensioning the repository upon conservative assumptions. In doing so, the operator will show that all efforts have been made to keep the consequences of the repository “as low as reasonably achievable” and to verify that the chosen strategy enables to meet given protection standards.

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# **Licensing Oriented Scoping Analyses of Gas and Nuclide Transport in Underground Repositories**

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## **1. INTRODUCTION**

To assess the long-term safety of an underground repository for the radioactive waste, generally water or brine inflow into the repository is postulated. The water or brine can react with the radioactive waste or with its containers and can gradually disassemble them. The nuclides after being dissolved in liquid phase can be transported out of the repository by the natural pressure gradient and, in case of salt formation, by rock convergence. The liquid flow and the nuclide transport can be enhanced by gas generation, mainly by hydrogen due to corrosion of containers and metallic materials. To investigate these effects, analyses were performed for a simplified two dimensional model with data and parameters taken from the Konrad iron ore mine in Germany, currently in the licensing procedure to serve as a repository for nuclear waste of negligible or low heat generation. Further in this paper, combined gas and nuclide transport in a repository in a deep salt formation is also analysed considering variable liquid density and rock convergence.

## **2. GAS RELEASE FROM A REPOSITORY IN AN ORE MINE**

To verify the applicant analysis of gas migration from the repository in Konrad facility employing the computer code ECLIPSE 100 [Resele 1991], a comparative analysis using code TOUGH2/EOS5 with two fluid components water and hydrogen was performed [Javeri 1992]. These two independent studies assuming a hydrogen source of 3 780 kg/year in the repository for  $t \leq 5\,000$  years show a substantial difference in the distribution of gas saturation (Fig. 1 and 2), which, even after a detailed discussion with the applicant, could not be clarified. However, since the broad parameter variations did not show any substantial influence of gas generation on groundwater flow, no relevant negative impact on the long term safety of the repository was concluded.

Figure 1. Gas saturation at 5 000 years in the simplified Konrad model (Resele 1991)

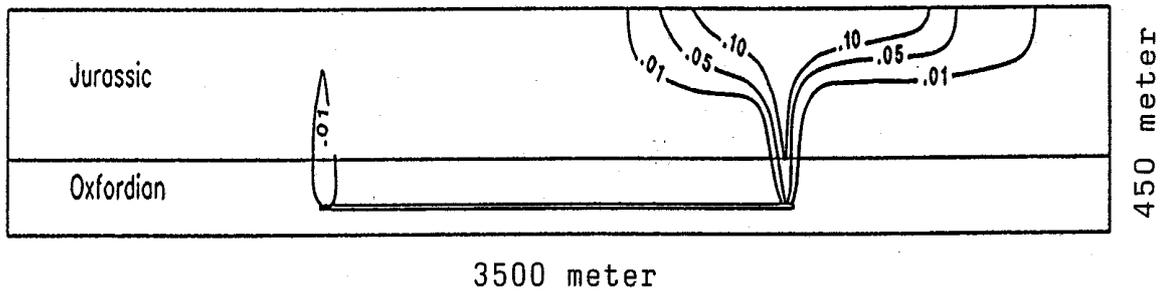
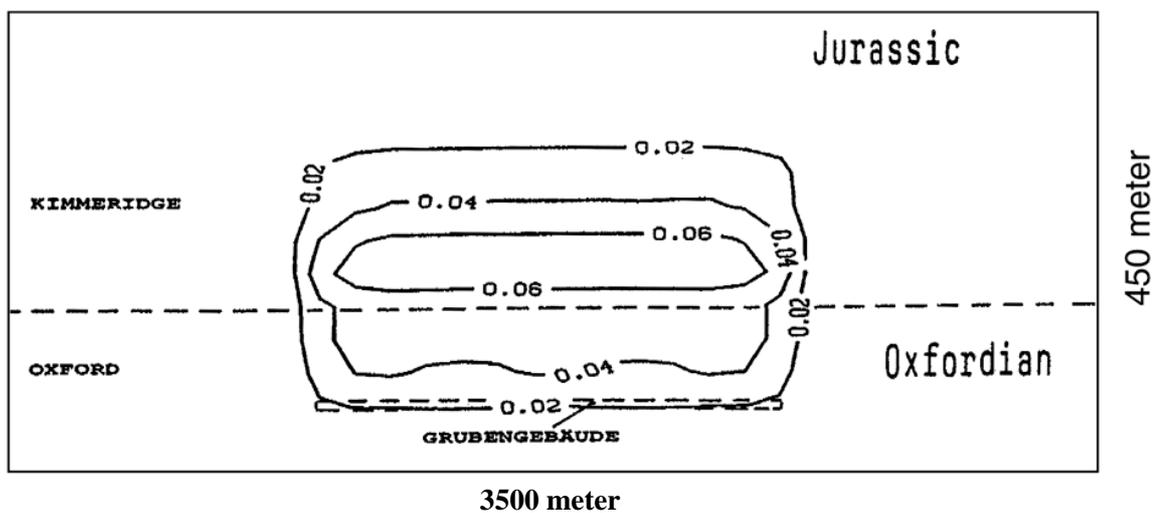


Figure 2. Gas saturation at 5 000 years in the simplified Konrad model (Javeri 1992)



### 3. GAS AND NUCLIDE TRANSPORT IN A REPOSITORY IN ROCK SALT

In the following two dimensional scoping analysis, a network of repository with two drift levels in rock salt is postulated (Fig. 3). A storage chamber with radioactive materials in the left corner of the bottom drift at a depth of around 500 m is filled with compactible crushed salt and is connected to the shaft via two drifts. Both lower and upper drift are filled with a non-compactible sealing material and the shaft with filling material. Since the sealing material has a low permeability of  $1E-16 \text{ m}^2$ , the excavation disturbed zone with the similar permeability along the drifts is also considered. Initially, the repository, except the storage chamber, is fully flooded with a low density (NaCl) brine entering from the top of the shaft. For the chamber, an initial liquid saturation of  $S_F = 0.2$  is assumed so that a required amount of liquid is available to generate hydrogen with a rate of  $Q_5 = 178 \text{ kg/year}$  or  $2\ 000 \text{ STP m}^3/\text{year}$  for  $0 \leq t \leq 2\ 000$  years in the chamber. Simultaneously, the crushed salt is compacted by the variable rock convergence reducing the void volume of the chamber. The radioactive substances ( $10^4 \text{ kg}$ ) in the chamber are simulated with a parent nuclide (half life:  $10^4$  years; daughter nuclide: no decay) in the liquid phase and can be driven out of the chamber by the pressurisation due to gas production and by porosity reduction in the chamber. With a source of high-density brine ( $Q_2 = 100 \text{ kg/year}$ ) at the bottom of the shaft, a possible inflow of Mg-rich brine is also simulated. Hence, the fluid system consists of four liquid components (low density brine, high density brine, parent nuclide, daughter nuclide) and one gas component hydrogen. To calculate a variable

porosity  $n$  and a permeability  $k$  of a compactible filling material, TOUGH2 is extended to consider a variable rock convergence depending upon pressure  $p$  and temperature  $T$  and to consider the induced fluid flow  $Q$  due to porosity change [Javeri 1996; 1998]:

$$n = n(p, T, t) = V_{\text{Void}}/V_{\text{total}}, \quad n \geq n_{\text{min}}, \quad n_t = (1/n) (dn/dt) = C_L/n, \quad k = k(n),$$

$$C_L = C_{\text{ref}} f_1(p) f_2(n) f_3(T), \quad C_{\text{ref}}: \text{Reference convergence rate of salt rock} = \text{constant},$$

$$f_1 = (1 - p_F / p_{\text{Rock}})^m, \quad f_1 = 0 \text{ if } p_F \geq p_{\text{Rock}}, \quad n_1 = 1 - n / n_{\text{Ref}}, \quad n_2 = (nn_1)^{(1/m)}, \quad f_3 = 1 \text{ for } T = \text{constant},$$

$$f_2 = 1 \text{ for } n \geq n_{\text{Ref}}, \quad f_2 = nn_1 (n_1^2 + n_2)^{-m} \text{ for } n < n_{\text{Ref}}, \quad C_L = 0 \text{ for } n < n_{\text{min}},$$

$$S_F = V_F/V_{\text{Void}}, \quad S_G = V_G/V_{\text{Void}}, \quad V_{\text{total}} = \text{constant}, \quad X_i = m_i/m_F,$$

$$Q_{F,i,\text{por}} = |nV_{\text{total}} n_t \rho_F X_i|, \quad Q_{G,\text{por}} = |nV_{\text{total}} n_t \rho_G S_G|.$$

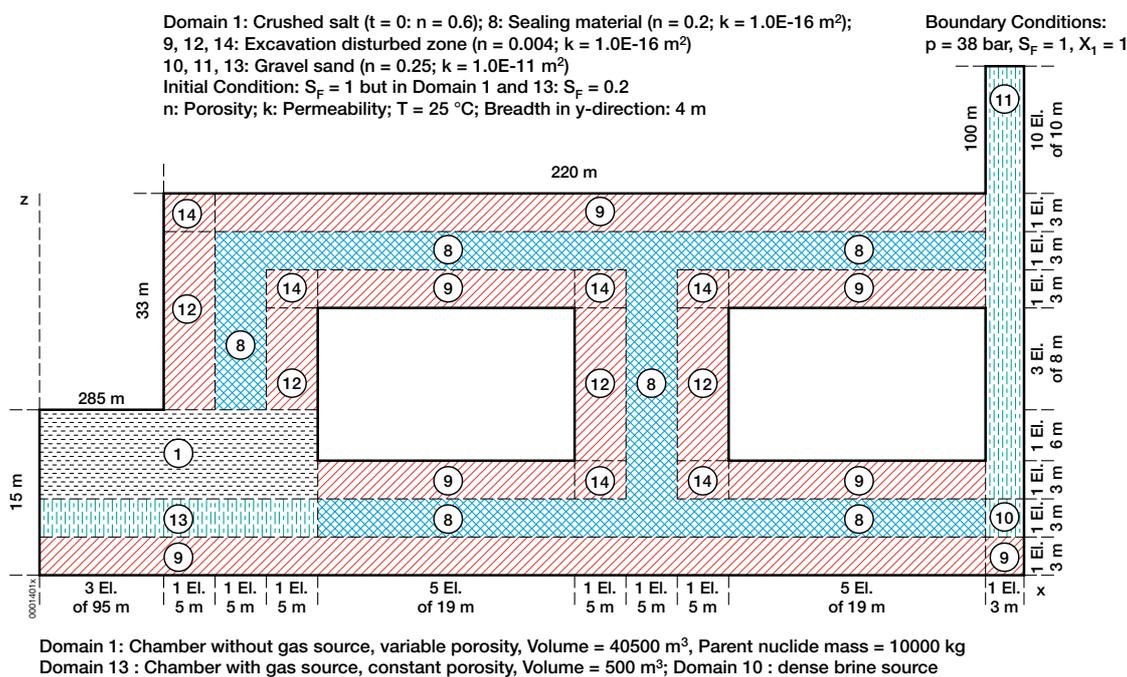
The important parameters are [Javeri 2000]:

- Variable rock convergence and crushed salt properties:  $C_{\text{ref}} = -5 \cdot 10^{-5}$  / year,  
 $n_{\text{Ref}} = 0.3$ ,  $n_{\text{min}} = 0.005$ ,  $m = 4$ ,  $p_{\text{Rock}} = 107$  bar,  $k = (2 \cdot 10^{-9}) n^{4.5} \text{ m}^2$ .
- Liquid phase:  $1/\rho_F = (1 - X_2) / \rho_1 + X_2 / \rho_2$ ,  $\mu_F = 6.29 \cdot \mu_{\text{Water}}(p, T)$ ,  
 $\rho_1 = 1.2 \cdot \rho_{\text{Water}}(p, T)$ ,  $\rho_2 = 1.292 \cdot \rho_{\text{Water}}(p, T)$ .
- Gas Phase (hydrogen): gas constant = 4124 J / (kg·K),  $\mu_G = 8.95 \cdot 10^{-6}$  Pa sec.
- Brooks-Corey relations for two phase flow:  
 $S_{F,\text{eff}} = (S_F - S_{\text{Fr}}) / (1 - S_{\text{Gr}} - S_{\text{Fr}})$ ,  $p_{\text{cap}} = p_{\text{cap,min}} / (S_{F,\text{eff}})^{0.5}$ ,  $S_{\text{Gr}} = 0.02$ ;  $S_{\text{Fr}} = 0.1$ ;  
 $k_{F,\text{rel}} = (S_{F,\text{eff}})^4$ ,  $k_{G,\text{rel}} = (1 - S_{F,\text{eff}})^2 (1 - S_{F,\text{eff}}^2)$ ,  $k_F = kk_{F,\text{rel}}$ ,  $k_G = kk_{G,\text{rel}}$ ,  
 $p_{\text{cap,min}} = 1$  bar for domain 8, 9, 12, 14 and 0 for domain 1, 10, 11, 13.

Using these model assumptions, following cases are defined:

- GS1: loading phase without heavy brine and without gas generation ( $Q_2 = 0$ ,  $Q_5 = 0$ );
- GS2: as GS1, but with  $Q_5 = 178$  kg/year for  $t \leq 2000$  years and  $Q_2 = 100$  kg/year;
- GS3: as GS2 but  $k$  (sealing material) =  $1\text{E-}13 \text{ m}^2$  and  $k$  (excavation disturbed zone) =  $1\text{E-}15 \text{ m}^2$ ;
- GS4: as GS2, but the upper drift is removed, i.e. no direct connection between the chamber and the upper drift available.

Figure 3. Two dimensional network of a repository in rock salt



These cases were computed with TOUGH2/EOS7R [Pruess 1991, 1995] including variable rock convergence and five fluid components up to  $t = 10\,000$  years [Javeri 2000]. The Fig. 4 and 5 show the pressure and porosity in chamber. In case GS1 without hydrogen source, the pressure increases slowly, the porosity decreases and around  $t = 7\,800$  years the flooding of the chamber ends. In other cases with a hydrogen source, depending upon the flow paths and the permeability of the sealing material, the pressure rises up to 110 bar, which can affect the mechanical stability of the filling or sealing material and repository substantially. The Fig. 6 to 8 reveal the liquid, gas and parent nuclide flow out of the chamber. In case GS1 without hydrogen source, the outflow starts after 7 800 years and is negligible compared to the cases with a hydrogen source. In Fig. 6, the case GS23 is not depicted, as the values are far greater than the plotting range. In case GS2 with a direct flow path between chamber and upper drift, the gas escapes faster and the contaminated brine flow out of the chamber is slower compared to case GS4 without upper drift. As expected, in case GS23 with a more permeable sealing material the gas escapes very early and the nuclide release is also significant. Without gas generation the nuclide removal from chamber is negligible, since brine outflow starts after 7 800 years. A maximum nuclide removal of 26.6% within 10 000 years occurs in case GS4 compared to 3.6% in case GS2 (Fig. 9). Additional calculations are reported in [Javeri 2000].

Summarising, one can conclude from the scoping analyses, that the gas generation can enhance pressure build-up in storage chamber affecting the mechanical stability of the repository and nuclide removal substantially, which however can be limited, if a significant gas release from the top of the chamber is allowed. The independent scoping analyses with reasonable assumptions by the reviewers in the early stage of the project can help to define the specifications to develop a decommissioning concept for a repository. For the licensing procedure, comprehensive studies including an appropriate arrangement of relevant flow paths and a broad variation of key parameters (gas generation, phase permeability) are very essential.

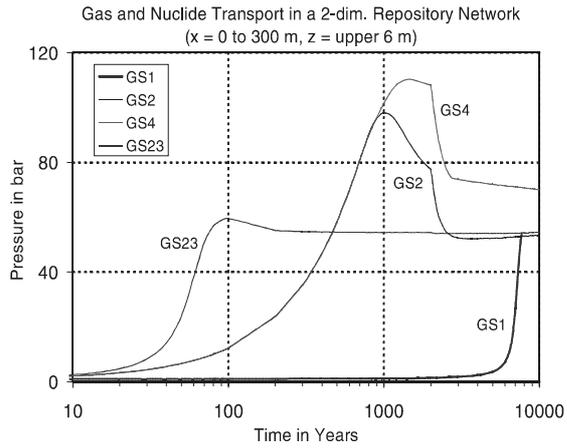


Figure 4: Pressure in the Chamber

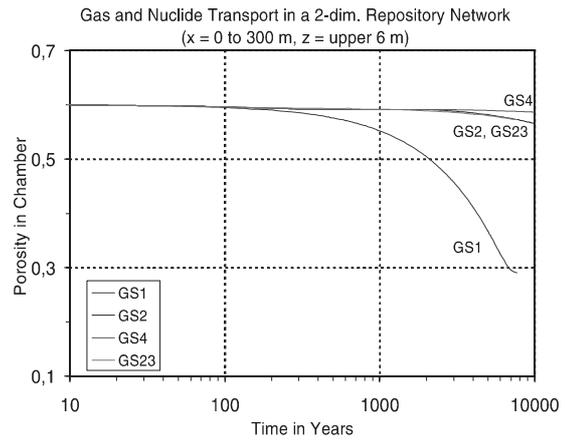


Figure 5: Porosity of Crushed Salt in chamber

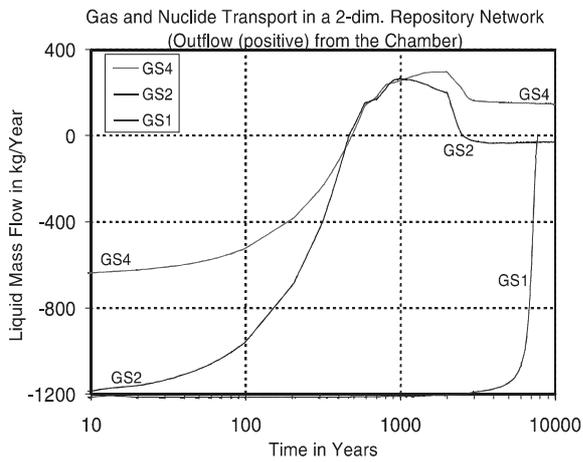


Figure 6: Liquid Mass Flow out of Chamber

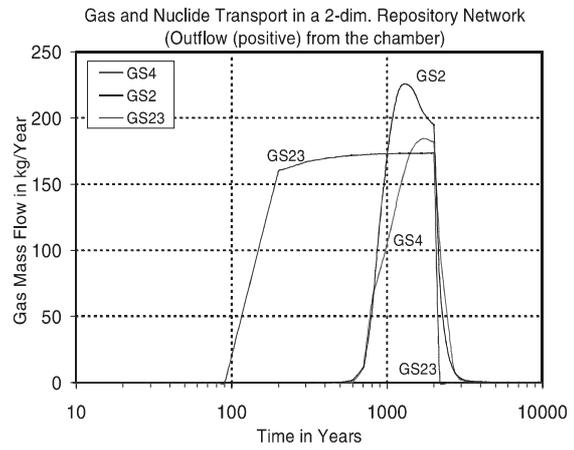


Figure 7: Gas Mass Flow out of Chamber

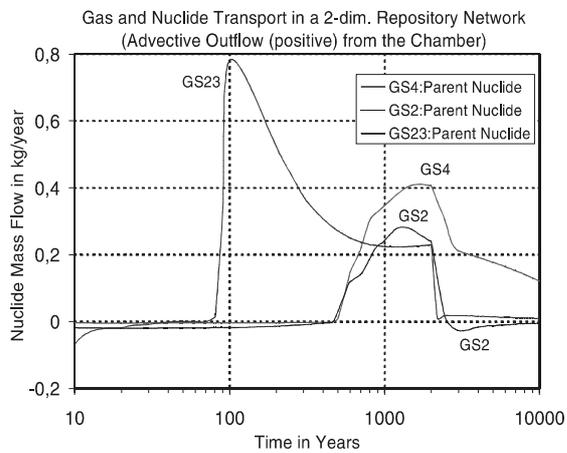


Figure 8: Parent Nuclide Mass Flow out of Chamber

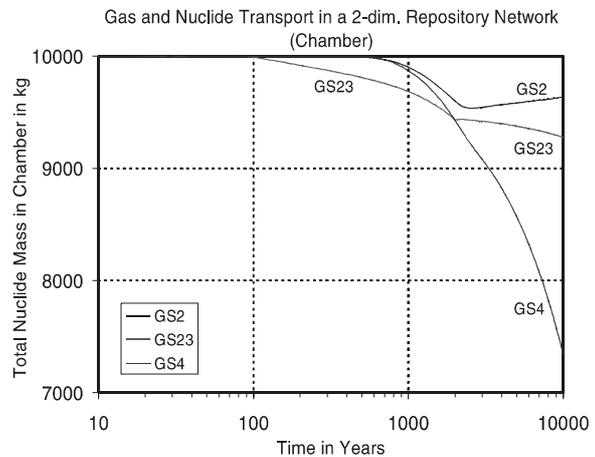


Figure 9: Total Mass of Parent and Daughter Nuclide in Chamber

## Symbols

m: mass [kg];  $\alpha$ : stress parameter; n: porosity; u: Darcy velocity; V: volume [m<sup>3</sup>];  
S: phase saturation; X: mass fraction;  $\mu$ : dynamic viscosity [Pa·s];  $\rho$ : density [kg/m<sup>3</sup>]

## Subscripts

G: gas phase; F: liquid phase; 1: low density brine; 2: high density brine;  
3: parent nuclide; 4: daughter nuclide; 5: hydrogen; i: fluid components 1 to 5

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## **SESSION IV**

### **PLANNED STRATEGIES FOR DEALING WITH GAS-RELATED ISSUES**

Chairman: Geert Volckaert (SCK•CEN, Belgium)



# **ANDRA's Strategy for the Treatment of Gas-related Issues**

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## **1. INTRODUCTION**

This paper briefly describes how gas is taken into account by ANDRA in its process of design of a deep underground repository. In the first part the context of ANDRA is reminded and the objectives of ANDRA with the associated schedule are precised. In a second part, the main questions associated to gas issue in a repository are listed. The methodology adopted by ANDRA in its design process is also presented. In the third part, a focus is made on the period between 1996 to 1998 to show how the studies carried out about gas issue during this period were taken into account in the design. Finally, some research and development axes are identified.

## **2. CONTEXT AND OBJECTIVES OF ANDRA**

### **2.1 Legal context**

The French law of 1991 concerning the management of high level/long lived radioactive wastes includes a programme with three axes of research:

- Partitioning and transmutation.
- Conditioning process and long term surface storage.
- Deep geological disposal.

ANDRA is in charge of the deep geological disposal project.

The law sets a fifteen-year period for research ending in 2006, after which the Government will submit an overall report evaluating the results of the research for all three axes to Parliament.

Within this framework, ANDRA is expected to assess by the year 2005 the feasibility of a deep geological radwaste disposal. A range of pertinent disposal concepts will be proposed in 2005, as well as related hypothesis of waste production and acceptance criteria. This goal implies to provide an answer to the major scientific and technical issues related to this feasibility. This item concerns in particular the gas issue in radioactive waste repository.

## 2.2 Meuse – Haute-Marne Site (“East Site”)

By the decree of August 1999, the French government authorised ANDRA to carry on its research programme in the Meuse – Haute-Marne site with an underground laboratory, and requests ANDRA to find a new granite site.

In all this paper, the discussion and results are focused on the “East” site, with its own characteristic regarding gas issue (permeabilities, porosity, diffusion coefficient, ...).

## 2.3 Radwastes types

ANDRA has to deal with 3 major types of radwastes: “B” (Intermediate Level Long Lived Wastes), vitrified wastes and spent fuel. Each radwaste type is characterised by specific physico-chemical properties and radionuclide inventory and brings its own gas source; for each waste type, the source of gas associated to the design must also to be taken into account (*cf. tables 1, 2, 3, 4*).

Table 1. **Characteristics of French ILLW wastes**

<b>French “ B ” Waste package types</b>	<b>Main composition (initial design concept base)</b>
Cemented Hulls and End-caps	Zircaloy, Ni alloy, stainless steel, carbon steel, ...
Cemented technological wastes	Stainless steel, carbon steel, plastic, ...
Compacted wastes (CSD-C)	Zircaloy, Ni alloy, stainless steel, carbon steel, ...
Bituminized sludges	Bitumen + Sludges (Salts : NaNO <sub>3</sub> , BaSO <sub>4</sub> , ...), ...

Table 2 **Gas source terms for ILLW wastes**

<b>Origin</b>	<b>Gas type</b>	<b>Mechanism</b>
Waste	H <sub>2</sub> , CO <sub>2</sub> , CH <sub>4</sub> , ... Radioactive gas	Corrosion Biodegradation Radiolysis
Package	H <sub>2</sub>	Corrosion
Overpack	H <sub>2</sub>	Corrosion
EBS	H <sub>2</sub> , CO <sub>2</sub> , CH <sub>4</sub> , ...	Radiolysis Bioactivity
Infrastructure Liner Supporting Wall Armature	H <sub>2</sub>	Corrosion

Remark: the four last lines of this table correspond to gas source term that can be optimised by the designer whereas the waste composition can't be modified.

Table 3. **Characteristics of Vitrified Wastes and Spent Fuel**

Waste type	Main composition (initial design concept base)
Vitrified Wastes	Glass Matrix : Si, Na, Al, B, ... ; Package : Ni alloy
Spent Fuel	UO <sub>2</sub> / PuO <sub>2</sub> + Zircaloy, Ni alloy, stainless steel, carbon steel, ...

Table 4. **Gas source terms for vitrified wastes and spent fuel**

Origin	Gas type	
Waste	H <sub>2</sub> , He Radioactive gas	Mechanism
Package	H <sub>2</sub>	Radiolysis Alpha decay
Overpack	H <sub>2</sub>	Corrosion
EBS	H <sub>2</sub> , CO <sub>2</sub> , CH <sub>4</sub>	Corrosion
Infrastructure Liner Supporting Wall Armatures	H <sub>2</sub>	Radiolysis Bioactivity

Remark: the four last lines of this table correspond to gas source term that can be optimised by the designer whereas the waste composition can't be modified.

### 3. GAS ISSUE AND DESIGN METHODOLOGY

The main objective concerning gas issue is to understand in what extent gas generation could influence radionuclide (RN) migration from repository to the biosphere for safety assessment. This main question can be split into the following sub-questions:

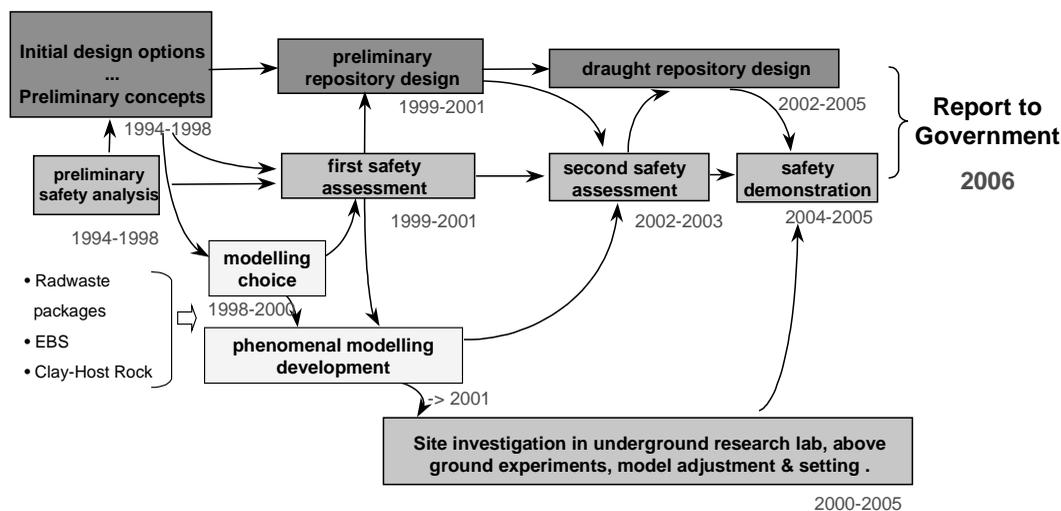
- Could a potential gas pressure build-up alter the performances of the Engineered Barrier System (EBS), by fracturing in particular?
- Could a potential gas pressure build-up alter the confinement performances of the geological media, by fracturing in particular?
- Could gas transfer modify the phenomenology of RN migration from the repository up to the biosphere?
- Could gas generation modify the chemistry in the EBS?
- How gas generation will modify the Thermo-Hydro-Mechanical (THM) transitory of a repository?

To cope with all these questions and to take them into account in the design process, ANDRA has adopted a general methodology based on formal iterations between design, safety assessment as well as modelling and experimental programmes (*cf. figure 1*). This methodology is

expected to ensure consistency between the advancement of the design, the assessment of the repository safety and the advancement of the geological knowledge of the site.

To illustrate this methodology we propose to present how studies carried out from 1996 to 1998 about gas issue were taken into account in the design.

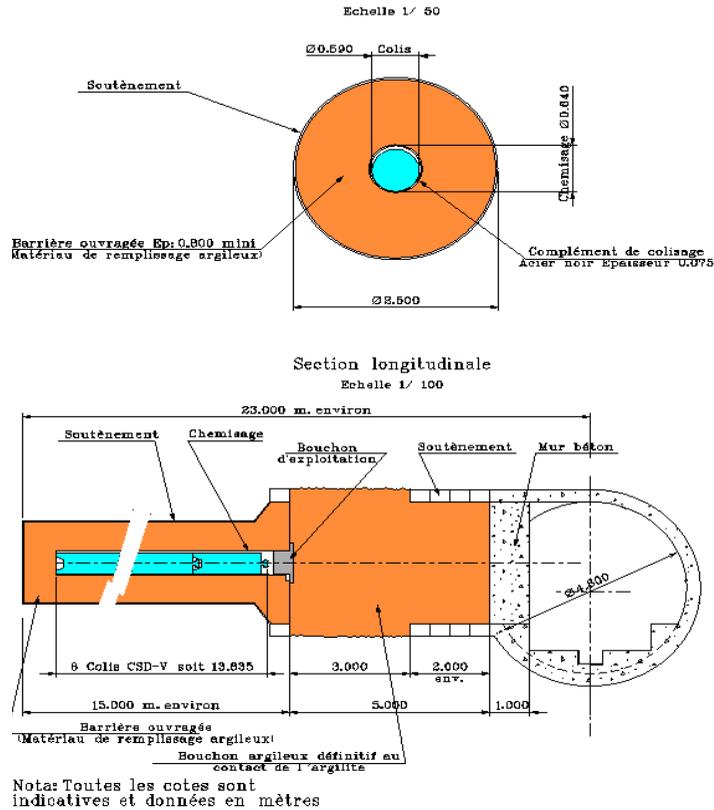
**Figure 1.** The French development plan for the deep geological disposal project



#### 4. DESIGN EVOLUTION FROM 1996 TO 1998 REGARDING GAS ISSUE

In 1996, ANDRA identified design options for each type of radwastes; that means specific geometries and engineering material options (in particular carbon steel overpacks; *cf. figure 2*). At this stage, gas issue had not been clearly taken into account. A set of studies about gas generation processes and gas migration was then initiated to understand in what extent gas generation could be a (safety) problem. A particular attention was also paid on the uncertainties associated to gas generation and transfer processes.

Figure 2. Initial design option for vitrified wastes



#### 4.1 Main results concerning gas generation

A systematic assessment of nature and amount of gas that might be produced in a repository was made for each design. A Kinetic of gas generation was also assessed for each gas generation mechanism (cf. table 5, figure 3).

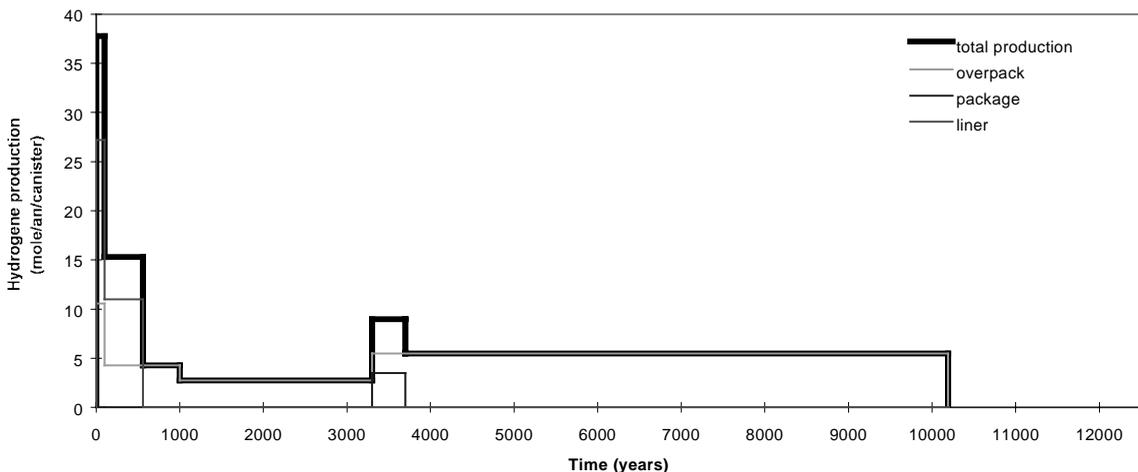
The main conclusions of these studies are the following:

- Hydrogen is be the main gas produced.
- Corrosion is the main gas source term considering both total amount that might be produced and the associated kinetic. Nevertheless, internal radiolysis and biodegradation may be not negligible for bituminized waste and compacted and cemented hulls and caps.
- Carbon steel overpacks of vitrified wastes appear as the main source of gas.

Table 5. Main gas source mechanism for different radwastes

WASTE TYPE	MAIN SOURCE INTEGRATE)	KINETIC (mole/year/canister) & ASSOCIATED DURATION
<b>Vitrified</b>	<i>corrosion</i> : 58 kmol H <sub>2</sub> / canister	<i>corrosion</i> (after oxidant phase) ≈ 40 from 0 to 100 years ≈ 20 from 100 to 500 years ≈ 5 from 500 to 10 000 years 0 >
<b>Bituminized</b>	<i>corrosion</i> : 20-30 kmol H <sub>2</sub> / canister  <i>internal radiolysis</i> : mainly H <sub>2</sub> 6 kmol/colis for bitumen For water?	<i>corrosion</i> : (after oxidant phase) ≈ 1 from 0 to 20 000 years 0 > <i>biodegradation</i> (after water income): 0,35 over 1 600 years 0 > <i>internal radiolysis</i> (after manufacturing): between 0,8 et 0,4 from 0 to 10 years between 0,4 et 0,1 from 10 to 3 000 years 0,03 >...
<b>CSDC (hulls and caps)</b>	<i>corrosion</i> : 20 kmol H <sub>2</sub> / canister  <i>internal radiolysis</i> (H <sub>2</sub> ) : ?	<i>corrosion</i> : (after oxidant phase) ≈ 1 from 0 à 20 000 years 0 > <i>internal radiolysis</i> : 1.7 from 1 000 years to ...?
<b>CBFC2 CEA CAC</b>	<i>corrosion</i> : 40 - 70 kmol H <sub>2</sub> / canister	<i>corrosion</i> : (after oxidant phase) between 1 et 3 from 0 à 20 000 years

Figure 3. Assessment of gas generation rate by corrosion for Vitrified Wastes



Remark: The hypothesis in temperature is the following: 120°C from 0 to 100 years; 70°C from 100 to 1 000 years and 50°C afterwards. Therefore, the steps in corrosion rates corresponds whether to a step in temperature or to water income in an initially impermeable package

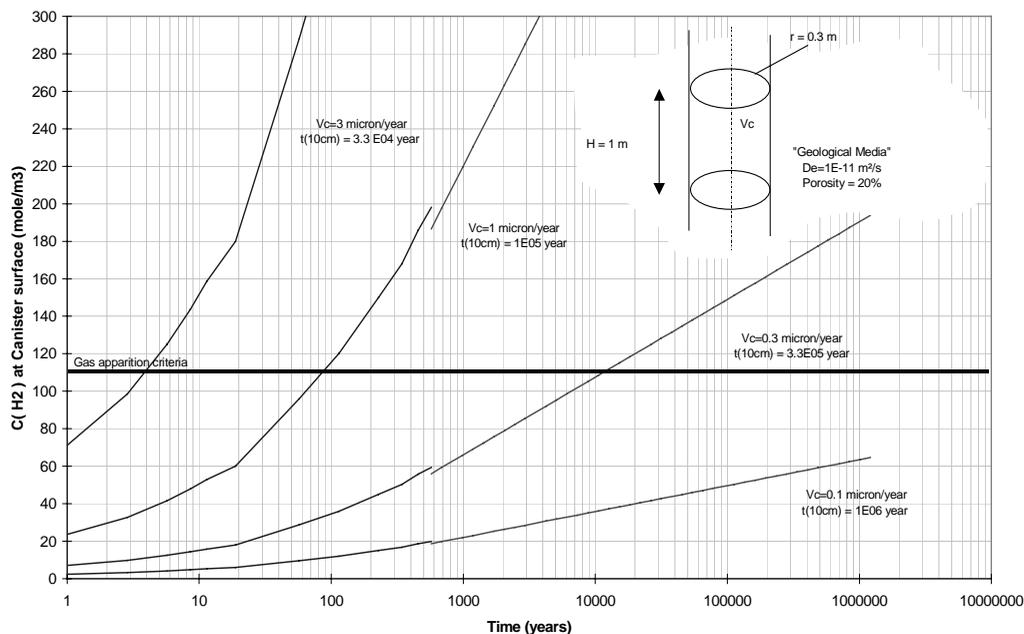
## 4.2 Main results about gas migration

ANDRA has carried out studies to understand in what extent gas generation could be a problem for its first design options: for example, diffusion of hydrogen was assessed to check if this sole mechanism could be able to evacuate the gas produced (*cf. figure 4*); the amount of gas was compared to the voids available in the EBS.

The main conclusions of these studies are the following:

- Diffusion of dissolved gas would not avoid gas phase rising.
- Voids available in the near field and the galleries are not able to deal with the total amount of gas, at hydrostatic pressure.
- Results obtained by considering gas generation and migration as decoupled are too rough to get conclusions about gas issue. Gas issue appears fundamentally as a dynamic problem for which it is required to take into account both the kinetic of gas production and the migration mechanisms.

Figure 4. Evaluation of hydrogen concentration at canister surface as a function of time for different corrosion rates



Remarks:  $t_c$  (10cm) corresponds to the time needed to corrode 10 cm of carbon steel; the gas apparition concentration criteria is assessed by taking into account hydrogen solubility and capillary effects

### 4.3 Main uncertainties

A particular attention was paid to the uncertainties associated to the assessment of gas generation and transfer processes. This attention led to identify three main uncertainties:

- In the assessment of the gas source term, the infrastructure of the repository, like metal liners and reinforcement of concrete structures, was not taken into account; well its contribution to the source of gas could be of major importance, and could consequently reduce the relative contribution of overpacks.
- The kinetics of corrosion adopted in the analysis were chosen to stay conservative. Nevertheless, it is felt that kinetic of corrosion could be smaller if the effects of expanding corrosion products, the availability of water in the EBS taking into account the argillite properties, or the retroaction of gas itself, were considered.
- Experimental gas transfer data show that Darcy's law can't be used to describe gas transfer in high compacted swelling clays; that's why ANDRA has decided to get involved in the Gambit Club project which aims at developing new physical and numerical models to deal with gas transfer mechanisms. Nevertheless, there are still uncertainties today on the phenomena since, on the one hand, the model developed in the Gambit Club does not totally fit the experimental results, and on the other hand, experimental data available today are not homogeneous. The Gas Workshop should be an opportunity to discuss this point.

### 4.4 New design

These results as a whole led ANDRA to consider that gas generation in a repository can't be neglected but also that there are uncertainties on gas generation and transfer processes. That's why it is difficult today to say if gas is a major safety issue for a repository. Therefore ANDRA has decided to keep the first design selected in 1996 but also to choose new designs integrating precautionary measures: at first, the amount of steel present in the repository will be limited as much as possible, by the choice of concrete containers for "B" wastes for example, and secondly, gas generation rates will be limited by the using of specific materials, like stainless steel or nickel alloys, for vitrified wastes and spent fuel overpacks / containers.

## 5. FUTURE R&D ACTIVITIES ON GAS GENERATION AND RELEASE

In agreement with the iterative methodology of design presented in part 3 and on figure 1, ANDRA has today chosen the preliminary designs and start a new iteration. The next milestones are 2001 and 2004 corresponding to safety assessment exercises.

Within this schedule, the main subjects that will be treated concerning gas issue are presented hereafter. They concerns both gas generation, to deal with the origin of the problem as a deep defence principle, and gas migration, to assess the consequences of gas generation on the safety of a repository.

The main axes of studies are presented here for each waste type:

### 5.1 Vitrified wastes and spent fuel

- reassessment of the amount of gas that might be produced;
- reassessment of the associated kinetics;

- R&D programme about corrosion mechanisms for carbon steel (modelling) and stainless steel (modelling / experiment);
- Description of THM behaviour of the near field with available models (Darcy), taking into account gas generation (study in progress);
- Development of new gas transfer models by participating to international projects (Gambit Club);
- Experimental data acquisition concerning gas transfer in bentonite (Hydro-Mechanical understanding);

## **5.2 “B” wastes**

- Reassessment of the nature and amount of gas that might be produced by radiolysis, biodegradation and corrosion of infrastructure;
- Reassessment of the associated kinetics;
- Assessment of pressure built up (modelling);

## **5.3 General topics**

- Data acquisition on gas transfer in the Argilite (Diffusion coefficient, permeabilities, ...);
- Up to now, gas issue was considered as a near field problem. Today, it appears of major importance to study the consequences of gas generation on RN migration, from the repository up to the biosphere.

# NAGRA Strategy for the Treatment of Gas-related Issues for Disposal of Long-lived Waste and Low and Intermediate Level Waste in Sedimentary Host Rocks

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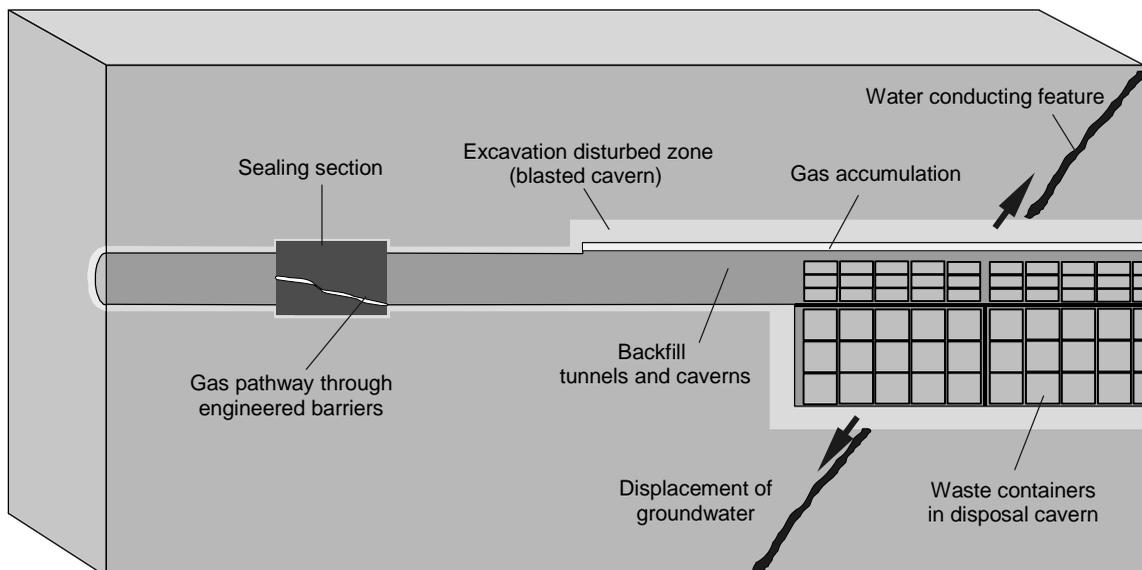
The Swiss waste management strategy foresees two different repositories; one for low and short lived intermediate level waste (L/ILW) and one for high level and long lived intermediate level waste (HLW/ILW). Both repositories are planned to be sited in tight, stable host rocks. The proposed site for the L/ILW repository is Wellenberg, a fractured marl formation located in central Switzerland. Two different host rock formations are under investigation for the HLW/ILW repository, the crystalline basement and the Opalinus clay formation, both of them situated in Northern Switzerland.

In the post closure phase, gas production in the disposal area may affect the performance of the repository due to the build-up of gas over-pressures (cf. Figure 1):

- formation of gas pathway (“channels”) through the engineered barriers and the geosphere;
- displacement of groundwater from the disposal area into the geosphere.

The release of volatile radionuclides is expected to be less important in the Swiss disposal concept.

Figure 1. Schematic representation of an emplacement cavern for low level waste



The relevance of gas generation and its treatment in performance assessment strongly depends on the properties of the different categories of radioactive waste. In the proposed L/ILW repository, significant amounts of gas will be produced due to corrosion of iron and steel and degradation of organic material. A free gas phase will accumulate in the upper part of the emplacement tunnels, because the low permeability of the host rock formation restricts gas release into the geosphere. An adequate description of gas flow through the fractured host rock formation is a key issue in estimating the magnitude of the pressure build-up in the emplacement tunnels of the L/ILW repository.

In Nagra's HLW/ILW programme, the amounts of gas produced by the intermediate level waste are expected to be considerably lower than for low level waste, but nevertheless significant. Formation of a free gas phase in the ceiling of the emplacement tunnels followed by pressure build-up is a likely scenario. The magnitude of the pressure build-up – in particular for the proposed Opalinus clay option - is governed by the rock properties and of the variables of state in the disturbed rock zone around the emplacement tunnels. Special focus is given to the duration of the resaturation period and to the mechanical convergence of the backfilled emplacement tunnels.

The gas generation rates of the high level wastes (vitrified wastes and spent fuel) are rather small and are due to corrosion of the steel overpacks. Arranged in a panel of parallel emplacement tunnels, the canisters are embedded in a bentonite buffer system. Currently, it cannot be ruled out that also for these wastes a free gas phase will develop. Release of gas through the bentonite buffer system is expected to be followed by formation of preferential gas 'channels' in the adjacent rock formation.

Nagra has been investigating the impact of gas accumulation and gas release on repository performance for more than a decade. The key issues under investigation were:

- Modifications of cavern layout and engineered barrier system design.
- Numerical simulations of pressure build-up due to gas accumulation and quantification of groundwater displacement from emplacement caverns of the repository.
- *In situ* determination of two-phase flow properties as part of the site characterisation programmes.
- Supplementary research programmes at the Grimsel Test Site (fractured hardrock) and at the Mont Terri Rock Laboratory (Opalinus clay).

Modifications of the cavern layout were focused on the arrangement of access tunnels and emplacement caverns of an L/ILW repository. In the modified layout, the access tunnels are connected to the upper part of the emplacement caverns rather than to the bottom part to prevent displacement of groundwater through the access tunnel along the excavation disturbed zone of the sealing section (cf. Figure 1).

Special backfill materials for the emplacement caverns were developed, thereby reducing the risk of fracturing the engineered barriers due to localised pressure build-up in the immediate vicinity of the waste containers. The developed backfill mortar is characterised by high porosity and high permeability [Jacobs *et al.*, 1993]. Extensive laboratory experiments were carried out to determine the material properties of the mortar [Mayer *et al.*, 1999a&b].

Numerous numerical studies were conducted to improve the understanding of two-phase flow processes in the vicinity of an emplacement cavern of the L/ILW repository [Mishra & Zuidema 1992, Senger *et al.* 1994, Senger *et al.* 1999a]. The aspects under investigation were (i) the estimation of maximum pressure build-up in the emplacement caverns also as a design criterion for engineering the sealing sections and (ii) the evolution of the pressure build-up with time as a determining factor for

the displacement of groundwater from the caverns. The studies included development of upscaling procedures for two-phase flow properties of fractured rock and sensitivity studies of gas migration in the host rock formation for a wide range of effective two-phase flow properties.

As part of the Wellenberg site investigation programme, a total of 10 gas threshold pressure tests were carried out in the deep investigation boreholes for *in situ* determination of two-phase flow properties of the host rock formation. These tests were analysed using a numerical two-phase flow simulator [Senger *et al.* 1999b]. Within the scope of the HLW/ILW programme, additional *in situ* and laboratory testing on two-phase flow properties have been carried out.

At the Grimsel rock laboratory, located in the central Swiss Alps in a crystalline geological setting, supplementary research programmes on gas migration have been conducted for more than 10 years. Improved methodologies for *in situ* determination of two-phase flow properties were developed, like the so-called “Extended Gas Threshold Pressure Test” [Croise *et al.* 1999]. Upscaling techniques and models were proposed to describe gas migration in fracture networks [Marschall *et al.* 1999]. As part of the current investigation phase, two new experiments have been established, which address gas migration processes in heterogeneous shear zones [Marschall *et al.* 1998] and gas migration through engineered barrier systems [Fujiwara *et al.* 2000].

As part of the Mont Terri investigation programme, various studies on gas migration in clay formations have been conducted since 1996. The research programme consists of in-situ determination of gas permeability [Wyss *et al.* 1999], laboratory tests for determination of two-phase flow parameters and *in situ* gas fracturing experiments.

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# **GRS/ISTec Strategy for the Treatment of Gas-related Issues for Repositories Located in Rock Salt**

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## **1. INTRODUCTION**

Performance assessments (PA) for underground repositories for all types of radioactive waste and the development of the necessary state-of-the-art PA-tools have been and still are an important domain of activity for GRS and its subsidiary ISTec. In the last decade the importance of gas-related issues to the operational and long-term safety of underground repositories was realised, and thus the gas generation processes and the resulting effects like gas migration, two-phase-flow, and pressure build-up have been included into the models for the prediction of potential radionuclide releases. Because rock salt was favoured as host rock for a HLW repository in Germany, emphasis was laid on the effects, the special boundary conditions and the interactions occurring in salt. Although operational issues of gas generation may play an important role in safety analyses, too, the following outline of the GRS/ISTec strategy is confined to long-term safety aspects.

## **2. GRS/ISTEC STRATEGY**

The strategy of GRS and ISTec for the treatment of gas-related issues in safety analyses for repositories located in rock salt comprises the following main stages, which will be partially discussed more detailed in the following sections:

- adequate geological characterisation of the repository area and the over- and underlying strata with regard to hydraulic properties and geochemical setting;
- comprehensive scenario analysis of the repository system;
- characterisation of the gas source term;
- physical understanding and modelling of the relevant fluid dynamical processes;
- coupling of fluid dynamics models with geomechanics, solubility effects and geochemistry;
- provision of necessary input parameters and qualification of models by experimental investigations;
- development of adequate numerical models and codes for the description of the multi-component multiphase flow processes;

- interactive assistance in the development of an adjusted repository design to cope with the gas issues;
- development of simplified representative models for implementation in uncertainty analyses and corresponding PA codes;
- application of the models and their results for safety analyses and PA for repository systems.

These activities are controlled by the requirements from the licensing. With this strategy, an adapted set of instruments for an adequate description of the gas-related issues and to give guidance for a repository design that will control all gas impacts to repository safety is made available in due time.

In a more detailed discussion of these topics it must be kept in mind that the gas related issues for wastes with negligible heat production (roughly low and intermediate level waste, L/ILW) heat producing waste (high level waste, HLW) principally differ. Therefore in the following sections the strategy for the individual stages will be outlined by contrasting both waste categories.

### **3. SCENARIO ANALYSIS**

For HLW in salt, gas generation is low because of the small quantities of water present in the waste, the host rock, and the backfill. It was demonstrated even for extended time periods that neither temperature, nor pressure, nor concentration-driven transport can provide amounts of water sufficient for the development of two-phase flow or a safety-relevant pressure build-up [1]. For this reason long-term safety analyses for HLW repositories in salt have to assume accident scenarios or some kind of faulted conditions to produce scenarios in which gas generation is a relevant issue.

If an event has to be postulated as starting point, neither time nor initial or boundary conditions are certain. Therefore in establishing a safety case calculations cannot be performed straightforward, but must be repeated with parameter variations in order to produce a conservative scenario or a set of scenarios covering all possible safety-relevant consequences. These variations also have to include the geological boundary conditions, e.g. location and nature of the fault, where brine inflow is postulated.

For L/ILW substantial gas generation may be an intrinsic property of the waste itself. In this case a repository in salt has to cope with amounts of gas, which constitute a potential hazard to the geological and engineered barriers. In this case either buffer space to control pressure build-up or a permeable closure concept controlling fluid flow and nuclide transport into the biosphere or combinations of both may be considered as design options. Since water or brine inflow into the repository area cannot be totally excluded under these conditions, two-phase flow modelling including counter-current flow of brine and gas through dams and seals is required a priori.

### **4. GAS GENERATION**

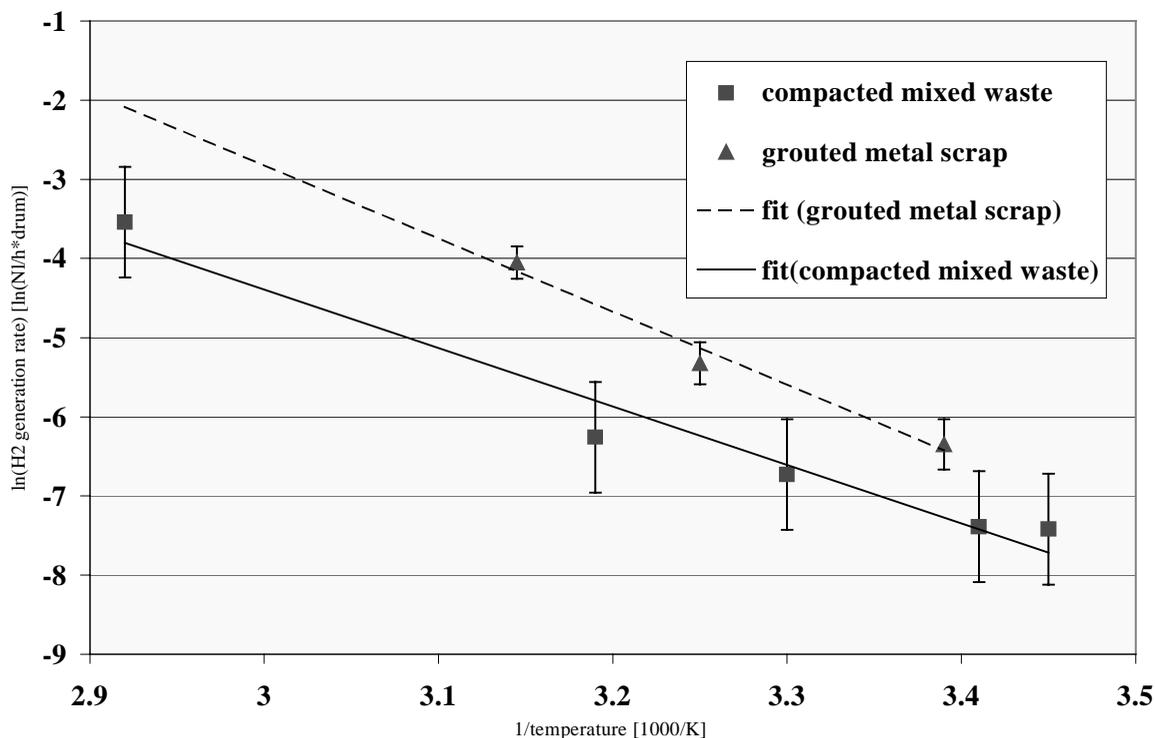
Two-phase flow, especially if coupled with geomechanics is a highly non-linear process. This means that the consequences of changes in gas generation cannot easily be predicted in terms of nuclide transport into the environment. Increasing gas generation may for example in one situation enhance fluid flow and thus the radiological consequences, while in a counter-current flow situation this may lead to a decrease in brine inflow with an associated reduction of contaminated brine outflow. Under these circumstances the GRS/ISTec strategy relies more on a realistic source term determination instead on a conservative gas generation assessment, which is difficult to define [8].

For HLW gas generation can be determined rather precisely from the contributing processes: mainly corrosion and radiolysis. For the expected or postulated situations in salt, intensive investigations for these generation mechanisms have been performed, allowing a sufficient accuracy of prediction. Additional information may be helpful with regard to gas generation from reactions between nuclear fuel and brine in case of direct emplacement of fuel elements and the long-term behaviour of corrosion.

For L/ILW gas generation is more waste specific rather than dependent on the host rock. Less well characterised gas generation processes as microbial degradation and chemical reactions besides corrosion have to be considered here as well. Here the waste constituents are not known in detail. Therefore, GRS/ISTec decided to address to the evaluation of measured gas generation data from real wastes instead of modelling all single gas generation processes. Since the available several thousand measurement results evaluated by ISTec [9, [11] have been achieved under interim storage rather than final disposal conditions, extrapolation is necessary. This is performed by deliberate additional measurements providing relationships e.g. for the gas generation rates of different waste types as a function of environmental temperature, humidity or pH value. Figure 1 gives an example for the temperature dependence.

For long-term predictions, gas generation from microbial degradation may become most relevant. Modelling of such processes is even difficult for short time periods. Therefore, GRS/ISTec have chosen an approach to derive limiting factors from natural analogues such as the degradation in coal or oil reservoirs within geological time scales. This work has just started and will hopefully provide means for a more realistic assessment of long-term gas generation for L/ILW.

Figure 1. **Temperature dependence of gas generation in LLW**



## 5. PHYSICAL MODELLING OF TWO-PHASE FLOW

Physical models for fluid flow in a repository in salt are based on the generalised Darcy law. Applicability is warranted as long as fluid transport takes place in the porous backfill material or the excavation damaged zone (EDZ). For sealing materials like bentonite this approach is not always adequate and other flow types might be more appropriate. At the moment such materials are not considered as primary options for repositories in salt and were therefore omitted. However, modelling has to take into account that an interaction exists between the geomechanically driven convergence of open spaces and the hydraulic properties of the backfill and the EDZ because of the plastic behaviour of salt. Pressure build-up may thus be enhanced but also constitutes reagent against the lithostatic pressure controlling the convergence. Furthermore salt is soluble and may either precipitate in colder regions or go into solutions in unsaturated brines. This may interact with the fluid flow because it reduces porosity and permeability in turn. Finally, gas generation is coupled with geochemical conditions (e.g. pH value, Eh value) and the availability of water or brine, which is a direct function of the liquid saturation in the repository area and/or in the waste.

Thus physical modelling has to describe complex highly non-linear interactions, which are challenging in theoretical, experimental and numerical terms. The GRS/ISTec strategy tries to solve this task by stepwise increasing complexity of the models assisted by experimental confirmation. For example, experiments have been performed to establish the two-phase flow parameters in compacted crushed salt by Technical University Clausthal under contract to ISTec, which provide not only necessary input data for modelling but also insights into the basic phenomena of two-phase flow under conditions of a soluble solid matrix [19].

The necessary degree of detailing and complexity may vary for different types of waste. For L/ILW e.g. it is considered sufficient to express the coupling between fluid flow and the geomechanical stress field by a simple relation between the lithostatic pressure and the convergence rate, because heat generation does not affect the stress field in the rock salt. In addition, solubility effects are modelled with a simple precipitation model [16], which does not consider re-solution processes. This is justified because under isothermal conditions no major influence of solubility effects is expected. For HLW, however, the coupling with geomechanics should rather be considered by means of a parallel modelling of stress history in the host rock and fluid flow, which is still a task for the future. Similarly, salt precipitation modelling should take into account time-dependent brine composition because of interactive solution/precipitation processes between different mineralogical salt constituents.

## 6. NUMERICAL MODELLING

The description of 1- to 3-D coupled transient brine and gas two-phase flow in porous fractured media is based on the mass balances for the components and the extension of Darcy's Law to two phases. Here the permeability and porosity of the porous medium and the saturation-dependent relative permeabilities of the phases and the capillary pressure are essential input parameters, for which reliable data are required. The mathematical model consists of a coupled system of highly-nonlinear partial differential equations and requires a high degree of stability and robustness of the numerical solvers. Within GRS and ISTec, adapted versions of the efficient general-purpose simulators TOUGH2 [14] and MUFTE [2] are used for two-phase flow calculations. As the application ranges of both codes overlap, they may be used as complementary tools as well as for mutual control and qualification.

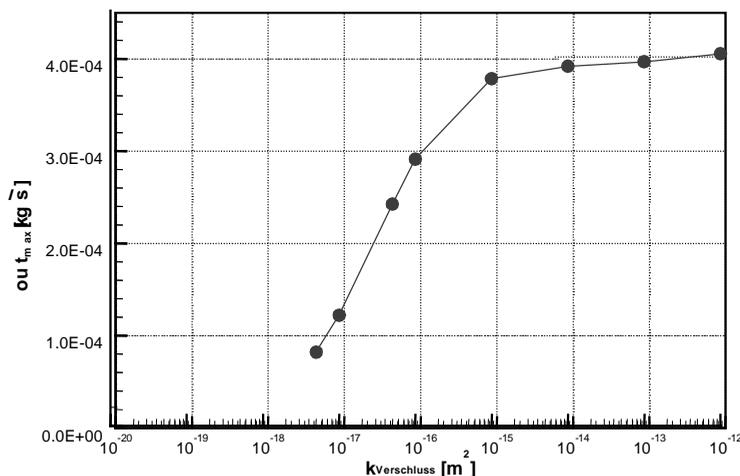
The TOUGH2 simulator is a 3-D numerical model for simulating the coupled transport of water, vapour, non-condensable gas, radionuclides and heat in fractured porous media. Moreover,

TOUGH2 offers capabilities and features as the flexibility to handle different fluid mixtures. Therefore, additional fluid property modules (EOS-Modules) have to be implemented. Since 1992 many enhancements have been appended to the basic version of TOUGH2 to cope with the special requirements for the modelling for repositories in rock salt. These were e.g. the inclusion of anisotropic diffusion and dispersion [4], rock convergence [6], [11], non-linear nuclide absorption and inhomogeneous porosity distributions [7], and solubility effects [15, chapt. 6].

2-D analyses for various conditions and benchmark calculations in comparison with other codes [13] indicate that TOUGH2 provides satisfying results and may be considered suitable for analyses of gas and radionuclide transport in porous media of any configuration. It has been widely used in different scientific areas and international studies and safety analyses including the WIPP licensing procedure.

Figure 2 illustrates the results of a number of TOUGH2 calculations [12] starting from a fully brine saturated repository for HLW in thick-walled POLLUX containers. Modelling included temperature and convergence effects. It shows that the permeability  $k$  of the dam sealing the emplacement gallery becomes relevant for the brine outflow rate only if  $k > 10^{-15} \text{ m}^2$ .

Figure 2. **Maximum brine outflow rate vs. dam permeability**

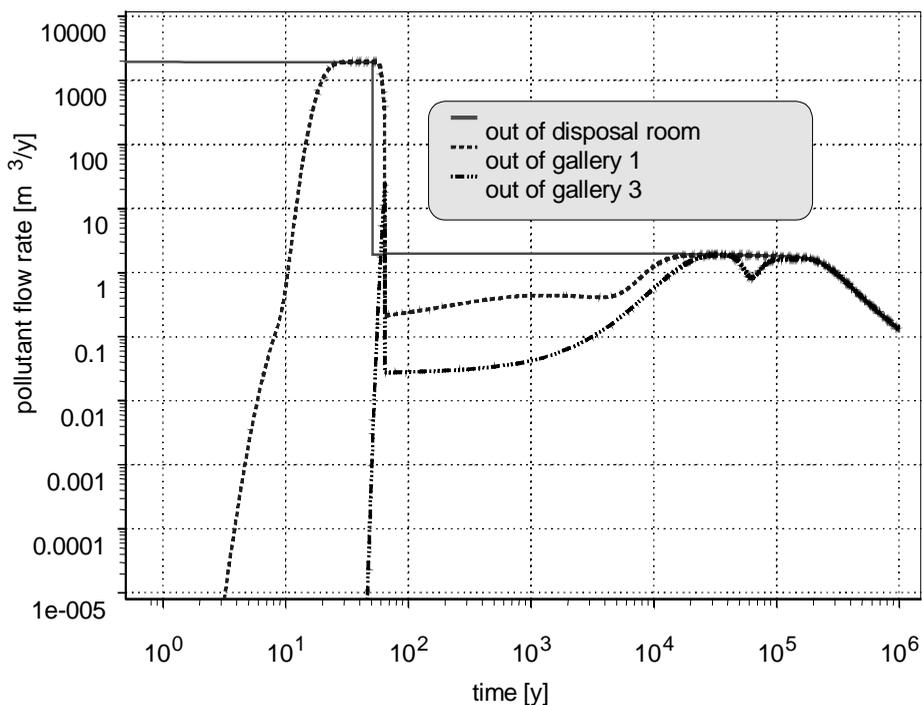


The MUFTE two-phase flow simulator is under development since 1993. For applications in the modelling of TPF processes in fractured porous media different versions are available, such as one version for any combination of liquid and gaseous fluids at constant temperature, and one version capable to model two-phase non-isothermal flow processes including changes in systems with a gas and a liquid phase [2]. The program version MUFTE-UG, which is still under development, is the most advanced [3]. It merges the physical basis of its predecessors with some unique features to model 3-D TPF in discrete fracture-matrix systems, includes very efficient techniques concerning the discretisation scheme and numerical solution methods, and allows the handling of very fine grid resolutions even for 3-D problems.

Two-dimensional sample calculations performed with MUFTE-UG revealed that a partial phase separation may occur at the top of an emplacement chamber and that the gas accumulated there will subsequently migrate along the top of the chamber and the connected galleries. So a separate gas

phase is formed that expands until stationary gas flow is achieved. With the formation of a separate gas phase a corresponding brine volume is expelled from the chamber and the gallery [10]. These model calculations delivered estimations about the extension of the phase and the quantity of brine displacement that allow the development of simplified models of the gas impacts in the near-field module REPOS of the EMOS PA-code. Figure 3 gives an example of the gas- and convergence-driven pollutant flow out of a sealed disposal chamber and within a connected gallery following a brine intrusion calculated with REPOS. Model calculations performed with MUFTE for a repository subsystem consisting of a gallery and a sealed deep backfilled HLW-borehole demonstrate that complex displacement processes between the phases take place which cannot be captured with a single-phase flow model [16]. These calculations also show that the special, partly anomalous hydraulic properties of crushed salt backfill introduce a considerable sensitivity of the results to the material parameters, the initial and boundary conditions and the repository structure in general.

Figure 3. Pollutant flow from gas-driven brine displacement



## 7. APPLICATION IN PERFORMANCE ASSESSMENTS

In the past GRS and ISTec have carried out many PA exercises treating gas issues. Most of this work was directed to an enhancement of the basic understanding of two-phase flow phenomena and to derive insights and requirements for the optimisation of repository designs. The forthcoming shut-down of the Morsleben repository will direct attention to demonstrate the long-term safety for an existing repository and its closure concept in a licensing procedure. GRS/ISTec has taken up work in this regard adhering to the strategy outlined above [5], [17]. With that, the models and codes to deal with the gas-related issues as well as the database have been and will be continuously developed and updated.

## 8. SUMMARY

The treatment of gas-related issues for repositories located in rock salt by GRS and ISTec has followed a strategy which has been developed with increasing complexity and degree of detail in the past. The strategy today clearly indicates the direction to establish a comprehensive safety case and the work that remains to be done. For gas generation mainly long-term aspects are an issue to increase accuracy of predictions. Physical modelling especially for HLW is still incomplete with regard to the coupling of fluid flow with geomechanics, solution/precipitation effects and geochemistry. The appropriate tools to transform the physical models into numerical solutions are at hand in principle but have to be further developed collaterally to the physical modelling. The first full-scale demonstration of safety regarding gas issues in rock salt will have to be provided for the licensing of the Morsleben repository shut-down in the near future.

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# **ENRESA Strategy for the Treatment of Gas-related Issues for HLW Repositories Sited in Granitic and Argillaceous Settings**

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## **1. INTRODUCTION**

Gas generation in HLW repositories is an unavoidable process whose consequences related with the global safety of the system will depend on many issues. Design aspects, materials used, waste form, rock type, behaviour of engineered barriers, etc. will condition the gas generated, amounts and generation rates, generation mechanisms, accumulation or dissipation from the waste form through the near field or transport in the rock. ENRESA has focused the research on gas related issues on those aspects which are relevant to the disposal concept foreseen in the Spanish program for high level radioactive waste management, which is a deep repository in granite, in which carbon steel canisters containing spent fuel assemblies will be placed in horizontal galleries surrounded by a bentonite buffer.

According with this repository concept, several research projects have been initiated by ENRESA, most of them within an international cooperation framework.

## **2. PAST DEVELOPMENTS AND PREVIOUS EXPERIENCE**

Generic projects like Pegase (EU) (1991-1993, ANDRA-GRS-ENRESA), aimed at studying the consequences related to gas production in different types of repositories, host rock and waste form, served to identify the main gas generating processes for several repository concepts, and to recognize the difficulties and needs in the modelling of the transport of gas through different media.

Benchmarking in the Evegase (EU) project (UPC-ENRESA) and the participation in Progress (EU) were good international exercises to test numerical tools developed in the context of previous projects, like Code-Bright.

Participation in the Reseal EU Project (1996-1998, SCK•CEN, ANDRA (CEA), ENRESA (Ciemat,UPC)), contributed to improve our experience on setting experimental devices to measure gas transport properties like permeability and on interpreting tests results. It also gave our research groups an opportunity to improve bentonite and other clay samples characterization oriented towards gas related issues. On the modelling field, expertise was gained through the interpretation of experimental results, through a better understanding of the gas transport processes in argillaceous media and through the improvement and verification of numerical tools with the results of in situ experiments.

The project Gasben (1998, ENRESA) was a short experience where a further review of the gas generation processes was made, resulting in a new version of the code developed in the Pegase project (Gasben). A first exercise to integrate the acquired knowledge and to evaluate the performance of the engineered barriers with respect to gas transport in a repository was also made in this project. A treatment of the coupled processes which affect the behaviour of the bentonite and the gas migration was performed, using a simplified one dimensional geometry. Finally, the implementation of new equipment for the experimental work was made together with an identification of the most adequate experimental procedures, both to investigate classical transport properties and to show transport mechanisms not identified or well understood at present.

ENRESA has contributed to the development of the EC/NEA Status Report on gas migration, which summarizes the state of the art on this subject and contributes to the identification of open issues.

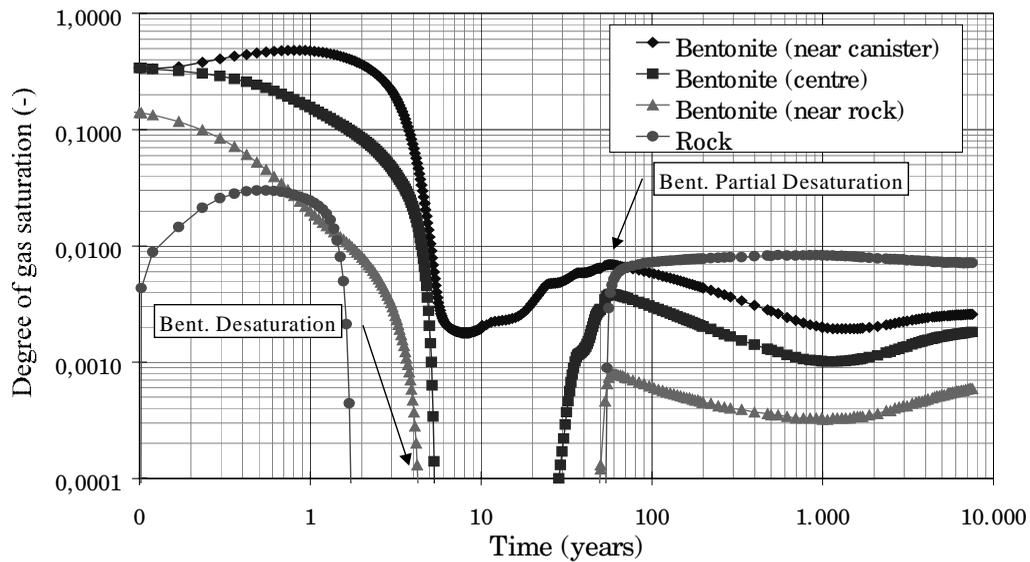
### **3. TREATMENT OF GAS-RELATED ISSUES IN PERFORMANCE ASSESSMENT AND OPEN QUESTIONS**

After the first preliminary exercise made in the Gasben project, a new analysis of the gas migration has recently been made within the context of ENRESA 2000, the integrated performance assessment exercise which is being carried out by ENRESA. This exercise is aimed at integrating the most of the results achieved by the ENRESA R&D Program, the gas migration through the bentonite buffer being one of them, and to test the global performance of the deep disposal concept. The analysis has been performed with Code-Bright, a finite element numerical tool able to deal with the coupled processes which affect the gas migration, based on the energy, mass and momentum balance equations. A two dimensional approach was followed with a classical continuum two-phase flow transport model, and some simplifying assumptions on the initial and boundary conditions. The properties of the materials correspond to those defined in the Spanish disposal concept and obtained in our R&D Program, mainly concerning bentonite properties.

The results obtained confirmed what it should be expected from the classical theory, the assumptions made, and what it has been observed in some experiments, that is, the gas initially present in the bentonite pores is expelled while the liquid saturation of the bentonite is increasing. Later on, once the saturation of the bentonite is complete and after the drying out transient, the amount of gas generated on the surface of the canister is enough to overcome the liquid pressure and to flow through the bentonite pores in two-phase flow and escape to the rock. A sample of this behaviour is shown on Figure 1, in terms of gas saturation at different positions in the bentonite.

Although this is a useful concept and model, many laboratory experimental results still remain to be understood, explained and correctly modelled. The available experiments performed until now show a strong dependency of the gas migration on the size of the clay sample, material type, density, pore size distribution, gas injection rate and pressure, duration of the experiment, etc. The dynamic change of the system while the experiment is being carried out or the performance under non-isothermal conditions has not been clarified yet. This situation may lead to different interpretations for the transport process and different models might apply like continuum two-phase flow, flow on discrete connected cracks, dilatation pathways models, etc. none of them giving full explanation to known experiments.

Figure 1. Gas saturation at different positions in the bentonite buffer



#### 4. FUTURE DEVELOPMENTS

Based on the previous and the international experience, ENRESA is currently involved in several projects, in an international cooperation framework, that will contribute to the solution of the gas related issues in radioactive waste management.

In the framework of Grimsel Phase V, the Gam Project (1998-2001, NAGRA, ENRESA, ANDRA, SANDIA) will produce a deeper insight of the gas transport through discrete features in granitic rock. It will try to clarify the influence of the heterogeneity of the fractures on the fluid flow in two-phase regime, and the role and values of mass exchange coefficients of rock, liquid and gas through different mechanisms (diffusion, sorption, dissolution, ..). The activities comprise field gas tracer tests, solute and particle tracer tests, geophysics measurements, laboratory investigations and a big modelling effort with numerical tools.

Under the auspices of RWMC, the GMT project (1997-2003, RWMC, NAGRA, GRS, ENRESA) is being conducted in the Grimsel Test Site with the aim to study the gas migration through the engineered barrier and the interface to geosphere (granite). The barrier system performance will be assessed with respect to gas migration, checking the response of different materials and concepts (bentonite-sand mixture, cement, presence of vent, ...). Numerical models will also be evaluated and applied in a real scale system as well as in the laboratory or mock-up scale.

Gambit project (2<sup>nd</sup> phase) (1999-2000, ANDRA, NAGRA, JNC, POSIVA, SKB, ENRESA, AEAT) will provide a new computational tool intended to represent the principal features observed recently in laboratory experiments in highly compacted bentonite. It will be used to interpret further experimental results. The model will try to represent gas pathway propagation as well as a dilation pathway model linked to the pressure induced by gas flow.

In the context of a third phase of the FEBEX project is also foreseen to continue with the gas sampling and analysis similar to what it is currently being done, and probably some gas permeability test will also be performed on this full scale experiment.

## **SESSION V**

### **IN-DEPTH DISCUSSION BY WORKING GROUPS**

Chairman: Wernt Brewitz (GRS, Germany)



# **Gas Flow Anomalies in Natural and Engineered Barriers**

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Gas generation and gas migration through natural and engineered barriers of underground repositories for nuclear waste is one of the most important safety relevant issues in the long term safety analysis. From analogy with flow processes in sedimentary rocks and from a relatively broad data basis on specific physical properties of barrier rocks, built up through basic research in the last decade, we have a sound basis to understand, to analyse and predict the migration of gaseous and liquid phases in the environment of an underground repository. However, there are distinct and essential anomalies of fluid flow through barriers which so far cannot be interpreted on the basis of classical flow theories. I want to demonstrate this with three statements, for which I will give experimental and theoretical evidence. Clear indications of the nature of this phenomena and proofs by experiments are missing in an acknowledgeable form.

1. Gas/Water Co-current Flow is Unrealistic for Plastically Deformable Barriers.
2. Gas Flow Mechanisms Depend on the Stress Field and Solid/Fluid Interactions.
3. Mechanical Disintegration/Integrity of Barrier Rocks can be Predicted from “Dilatancy Threshold”.

## **1. CO-CURRENT AND PERCOLATION TWO-PHASE FLOW THEORIES**

The classical concept of two-phase flow of differently wetting phases is explained by the concept of effective permeabilities, which is based on a continuous distribution of gas and water phase in a rigid and accessible pore network (Figure 1). The magnitude of the effective phase permeability is controlled by the pore fluid displacement pressure and can therefore be related to pore size distribution and wettability of the rock. The pore fluid displacement pressure must exceed the capillary pore entry pressure and friction pressure losses.

This concept explains why a certain fraction of the wetting phase remains in small pores, that cannot be drained by the non-wetting phase and explains why a certain amount of non-wetting phase may be entrapped by the wetting phase during the imbibition stage. In the drainage form of the effective permeability curve a critical saturation of the non-wetting phase may exist, which is necessary to built up a continuous gas phase before gas flow starts.

In a barrier rock, porosity can be so small and capillary entrance pressure so high that fluid injection pressure may exceed the micro shear strength of the rock and the non-wetting phase may create new flow paths in the migration process.

Figure 1. “Effective” gas/water permeabilities in a water wet porous medium

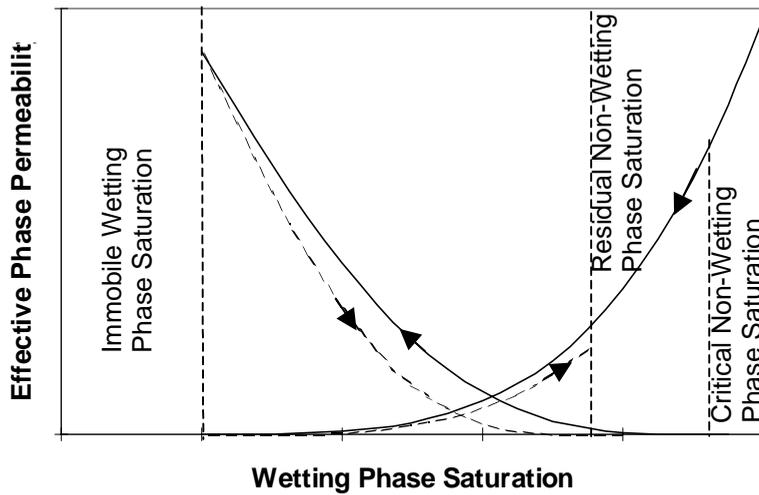
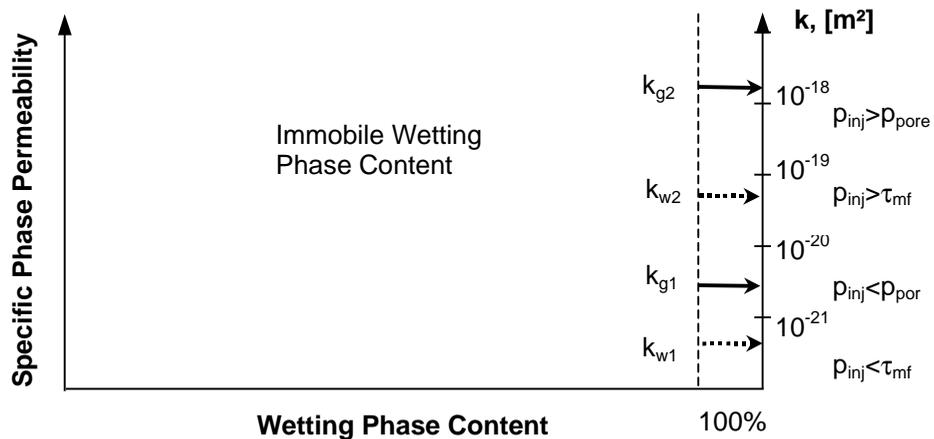
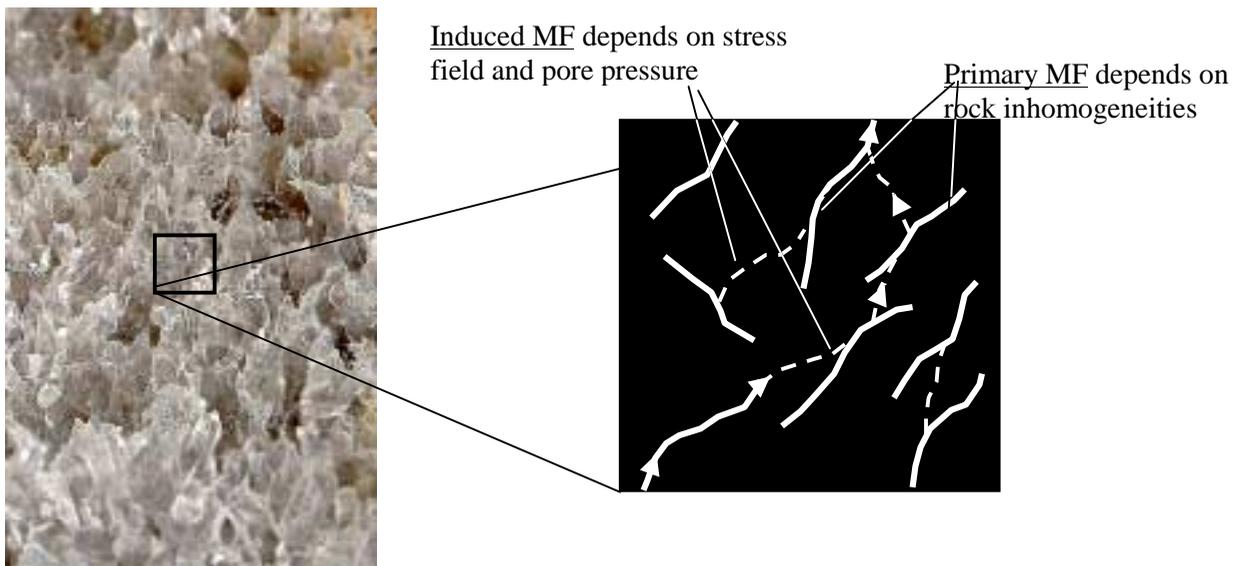


Figure 2. Concept of the percolation type two-phase flow



In some barrier rock types the internal surface area is tremendous and the total micro-porosity is also very large, however it can be hardly determined without destroying the rock: New analytical tools like nuclear spin resonance tomography may help to understand the issue of total and effective porosity in clay rocks, but temporary we can only rely on water content and should therefore forget the term saturation with respect to clay type rocks. In Figure 2 the amount of physically bound water in clays covers nearly the whole pore volume and leaves only little room for the displacement of free-water by a non-wetting phase like gas. The limiting entrance conditions for fluid flow are given by pore pressure for the wetting phase and microscopic shear strength for the non-wetting phase, where permeability of the phase is no longer dependent on saturation and may therefore be called “specific phase permeability” instead of “effective”. The level of specific permeabilities varies exponentially with the injection pressure related to pore pressure and microscopic shear strength, as it can be seen in Figure 2. Following this hypothesis, who have no physical rule to explain, how the network of voids is generated, interconnected and may lead to gas flow in a dense barrier rock (Figure 3).

Figure 3. **Interconnection of primary voids in the percolation flow concept**



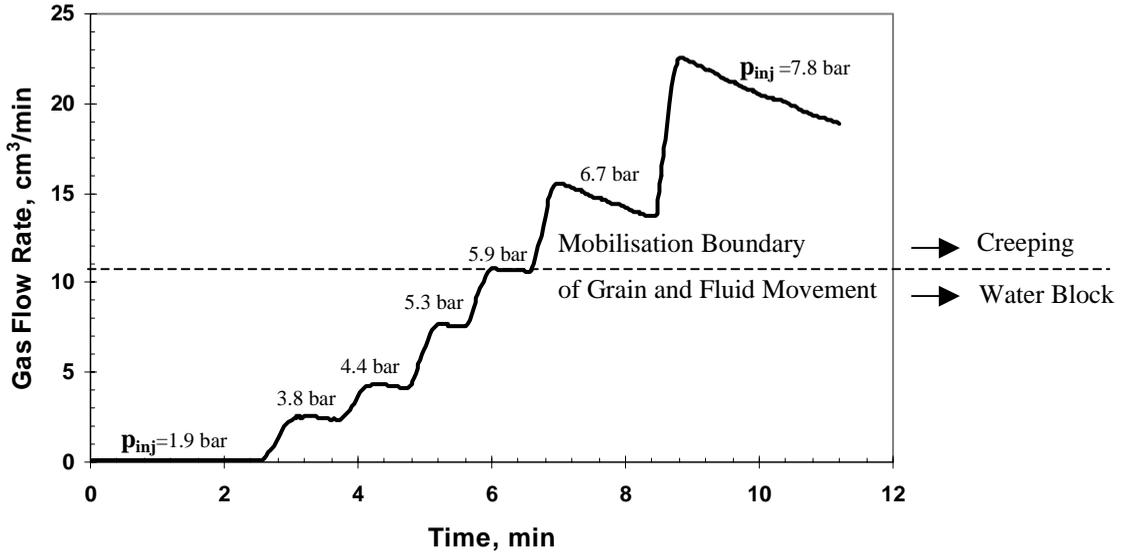
The term microfracture in this respect is used for any type of inter- and intracrystalline voids, which may exist in a tight barrier rock. Figure 3 shows, how the primary microfractures, which depend on rock inhomogeneity may be interconnected by induced microfractures, depending on stress field and pore pressure. Mathematical percolation theories are existing, which can be even improved by neuronal path finding network concepts.

## 2. **GAS FLOW MECHANISMS DEPEND ON STRESS FIELD AND SOLID/FLUID INTERACTIONS**

It is well known, that in ductile rocks the interaction between rock aggregates and fluids may lead to special hydraulic and mechanic effects like creeping, water block, mineral solution, swelling and precipitation. From our own experimental results on engineered barriers (compacted salt granulate), we know that transient effects are controlling the two-phase flow phenomena.

The gas flow in a compacted salt granulate of a final porosity of 5 per cent and hydraulic permeability of several millidarcies was investigated at irreducible brine saturation (Figure 4). This irreducible brine saturation was adjusted by the standard restored – state method of capillary equilibrium. In a long term procedure of stepwise increasing injection pressure, the gas flow rate was measured by a highly sensitive mass flow rate detector. Exceeding a certain level of injection pressure, which can be defined as a mobilisation boundary for grain and fluid movement, transient effects of decreasing gas flow can be observed, which may be attributed to creeping or remobilisation effects of brine. Extrapolating these effects to long term flow mechanisms in engineered barriers and speculating on flow mechanisms in natural salt barriers, this convergence effect may cease the gas flow in plastically deformable rocks and may lead to gas inclusions. Similar observations have been made in caprocks of deep gas carrier rocks, the carboniferous sandstone.

Figure 4. Transient effects on gas flow in compacted salt granulates (2)



### 3. MECHANICAL DISINTEGRATION/INTEGRITY OF BARRIER ROCKS DEPENDS ON THE DILATANCY THRESHOLD CONCEPT

In the vicinity of underground constructions, like shafts and tunnels for repositories the stress field conditions are characterised by an unbalance of tangential and radial stress, which is called a deviatoric stress situation. If this deviatoric stress situation is simulated in laboratory experiments the disintegration of the rock may be observed as a continuous process represented in Figure 5.

When the axial stress exceeds a certain level of the radial pressure on the cylindrical core, dilatancy effects may increase the pore volume the rock specimen by generating new microfractures, which can be deduced from the increase in cumulative acoustic count corresponding to the performance of the porosity curve and to the increase in permeability. This disintegration process is not a reversible type process and may lead to an increase in the residual permeability of the rock after the convergence phase in the repository, as it is shown in Figure 6.

The disintegration performance of a rock may be described by superposition of compressional and dilatant stress effects as explained in the following formula.

$$\frac{k_1}{k_3} = \left( a \frac{\sigma_1}{\sigma_2} \right)^{-n} \exp \left[ A \left\{ \left( \frac{\sigma_1}{\sigma_2} \right)^m - 1 \right\} \right] \quad (1)$$

The critical stress ratio, where the disintegration of the rock begins, corresponds to a critical Poisson ratio and is defined as the dilatancy threshold (compare to Figures 5, 7).

Figure 5. Compressional test on a salt rock core (4)

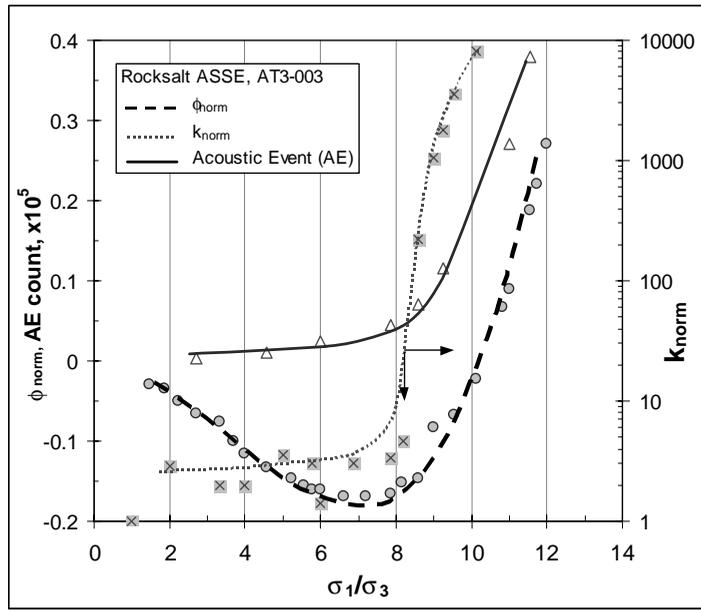
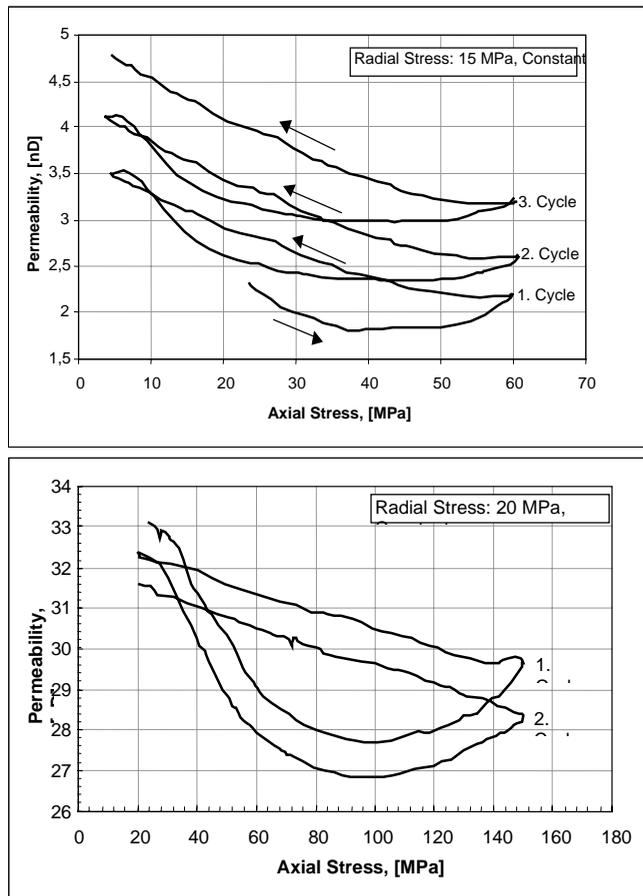


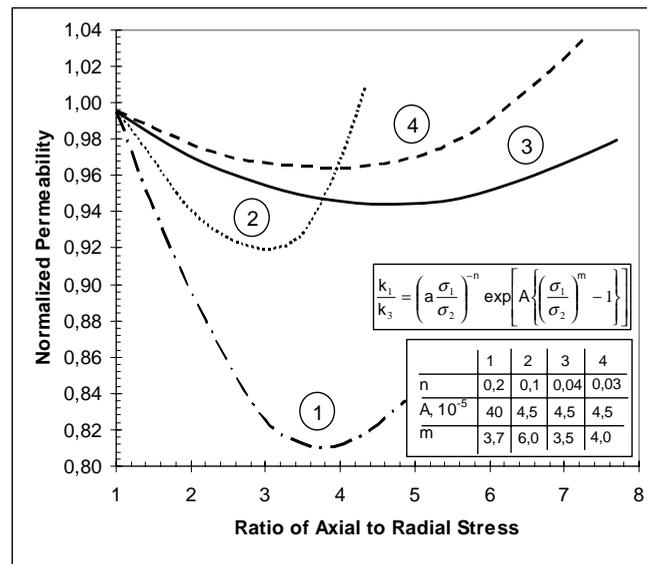
Figure 6. Explanation of stress hardening and stress softening phenomena in hard rock (1)



## CONCLUSION

These three statements derived from experimental evidence have shown, that we have a remarkable deficit in physical understanding of some of the effects occurring during fluid migration from underground repositories of nuclear waste. Comparing these deficits with the uncertainties of geological and physical scenarios in long term safety analysis, we may come up with the conclusion, that it is more straight forward to introduce stochastic modelling concepts, using neuronal network techniques and/or optimisation strategies like evolutionary strategies, simulated annealing methods or simplex strategies as “Path Finder for Fluid Migration”. We have evidence, that these strategies are very successful in many technological areas. Doing so, we might be able to predict flow events with respect to “when” and “where” they might occur, however we would never be able to explain “why” they occur and this in my opinion, gives legitimation to continue experimental research on gas issues for long term safety analysis on nuclear waste repositories.

Figure 7. **Simulation of stress dependent permeability under deviatoric conditions (3)**



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