

Performance Assessment Methodologies in Application to Guide the Development of the Safety Case

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FOREWORD

The work presented in this report was developed within the Integrated Project PAMINA: **P**erformance **A**ssessment **M**ethodologies **IN A**pplication to Guide the Development of the Safety Case. This project is part of the Sixth Framework Programme of the European Commission. It brings together 25 organisations from ten European countries and one EC Joint Research Centre in order to improve and harmonise methodologies and tools for demonstrating the safety of deep geological disposal of long-lived radioactive waste for different waste types, repository designs and geological environments. The results will be of interest to national waste management organisations, regulators and lay stakeholders.

The work is organised in four Research and Technology Development Components (RTDCs) and one additional component dealing with knowledge management and dissemination of knowledge:

- In RTDC 1 the aim is to evaluate the state of the art of methodologies and approaches needed for assessing the safety of deep geological disposal, on the basis of comprehensive review of international practice. This work includes the identification of any deficiencies in methods and tools.
- In RTDC 2 the aim is to establish a framework and methodology for the treatment of uncertainty during PA and safety case development. Guidance on, and examples of, good practice will be provided on the communication and treatment of different types of uncertainty, spatial variability, the development of probabilistic safety assessment tools, and techniques for sensitivity and uncertainty analysis.
- In RTDC 3 the aim is to develop methodologies and tools for integrated PA for various geological disposal concepts. This work includes the development of PA scenarios, of the PA approach to gas migration processes, of the PA approach to radionuclide source term modelling, and of safety and performance indicators.
- In RTDC 4 the aim is to conduct several benchmark exercises on specific processes, in which quantitative comparisons are made between approaches that rely on simplifying assumptions and models, and those that rely on complex models that take into account a more complete process conceptualization in space and time.

The work presented in this report was performed in the scope of RTDC 3, and more specifically of WP 3.2 "PA approach to gas migration". The objective of SCK•CEN within this workpackage (as was formulated in the technical Annex 1 of the project's contract description and updated on a yearly basis), is threefold:

 Assess the influence of high near field temperatures on the gas generation and migration processes





- Verification of the numerical accuracy of the applied simulation tool TOUGH2 by performing benchmark calculations with another code.
- Assessing the feasibility of modifying the formulation of certain constitutive laws within the TOUGH2 code (tool refinement)

This milestone M3.2.16 constitutes the final report of SCK•CEN dealing with gas-related research conducted in the framework of PAMINA's WP3.2. It presents the selected calculation cases based on the Belgian multi-barrier repository design for disposal of vitrified HLW and the most essential results obtained at interim stages of the project and already reported in M3.2.2, M3.2.7 and M3.2.15. Additionally, it presents some new, yet unpublished, results obtained in the last stage of the project, focussing on building confidence in the numerical tools applied by means of benchmark calculations.

As such, this report should constitutes a stand-alone report, providing a good overview of the modelling progress during the PAMINA project 2007-2009 by SCK•CEN.

All PAMINA reports can be downloaded from http://www.ip-pamina.eu.

1. Introduction

Assessment of gas generation and transport is a necessary part of a total system performance assessment and hence of a safety case. This aspect has to be thoroughly investigated to demonstrate the system's safety and, if deemed necessary, to assess to which degree measures (e.g. facilitating gas evacuation) are to be taken in order to guarantee the safety of a repository system, whether it is a surface disposal facility or a repository in deep geological formations. As such, gas generation and transport may be one of the most important issues left in demonstrating the safety of a repository system. At least it is one of the most challenging issues, because of its complexity and the relatively large uncertainties associated with both the source term and the gas transport processes. The uncertainty related to gas transport is related to the complex, non-linear system's behaviour, largely due to mechanical implications of which the governing constitutive laws are not yet fully established, the difficulty to characterise the considered porous media concerning gas transport and the lack of large-scale experiments validating the process models and tools used in assessing gas transport.

In this report, gas issues in the context of a safety case are assessed by means of a case study, based on disposal of vitrified high-level waste (HLW) in Boom Clay. A stepwise approach will be followed with particular focus on *insight modelling* in view of system understanding and *tool verification*. The focus lies on near-field processes and their interactions. Gas-related processes outside the host rock and in the biosphere are not discussed nor gas issues during the pre-closure (operational) phase.

The report starts with some introductory chapters on the current PA approach to gas migration (chapter 2), some background information on disposal of vitrified high-level waste in Belgium and a short description how to tackle the gas problem for this case (chapter 3). Chapter 4 assesses the timescale for saturation of the EBS materials, to scope to which degree the corrosion reaction and gas transport is subject to an increased in-situ temperature. In addition, the gas source term is quantified. Chapter 5 is the main chapter of this report, which summarizes two basic types of modelling studies: scoping calculations assessing the ability of the host formation to dissipate the generated gas volumes by considering a simplified geometry and conceptual model (transport by diffusion only); and detailed total system evaluations which consider all transport processes (diffusion and advection in liquid and gas phase). Additionally, the effect of an improved temperature dependent gas solubility formulation was assessed, which required some modification to the source code. Chapter 5 concludes with a benchmarking study comparing results of the numerical codes TOUGH2 and CODE_BRIGHT for three cases. Finally, chapter 6 highlights the overall conclusions drawn in the framework of this workpackage.





2. Current PA approach to gas migration

As a starting point for the treatment of gas issues in a safety case, it is obvious that for key waste fluxes within their dedicated designs, an accurate estimation of the source term for gas should be obtained. Recent system-level reports (Johnson, 2006; Weetjens *et al.*, 2006) dealing with disposal of HLW and spent fuel confirmed that anaerobic corrosion, which produces H_2 gas, will be the main contributor to gas production in the repository near field (NF). In case of disposal in a low permeability plastic clay formation, such as Boom Clay, it is important to assess whether or not gas production rates can exceed the capacity of the near field to store and dissipate these gasses.

This kind of assessment is usually performed using numerical tools, implemented in a stepwise manner from simplified models (pure diffusion) to more complex models (two-phase flow possibly coupled to geomechanical models) according to the need. The methodology for these calculations depends on the questions to be answered:

- 1. Will the gas production rate be small enough for the host formation to evacuate the dissolved gas by molecular diffusion through the pore water for the considered waste type?
- 2. If not, will the maximum near field gas pressure be high enough for fractures in the host formation to be created (or in other words: what is the impact on the disturbed/damaged zone?)
- 3. Between these extremes, what is the maximum degree of desaturation and what will be the advective flow component?

These numerical assessments are complemented with sound qualitative arguments (*e.g.* potential impact of EDZ, role of anisotropy in sedimentary host rocks). Furthermore, validation of the models is important to evaluate whether the conceptual models and applied constitutive laws are adequate and comprehensive, (*i.e.* are all processes or at least the most dominant ones properly captured?). Ideally, this requires large-scale experiments conducted in realistic conditions, and these types of experiments are scarce for the time being. Finally, it is clear that there should be confidence in the numerical tools themselves. Therefore, they should be verified for their computational accuracy, which can be done by benchmarking with other codes.

Ideally, this approach should be applied to all waste streams considered within a given safety case. At least, a qualitative judgement should be done based on knowledge of the inventory and surface area for corrosion of the waste stream under consideration and, if necessary, detailed assessments of gas generation and transport for key waste streams.





3. Problem description: vitrified HLW disposal in Boom Clay

3.1 Boom Clay as reference host formation

ONDRAF/NIRAS is assessing the feasibility of disposing vitrified HLW and/or spent fuel in a deep geological repository excavated in the Boom Clay of the Boom Formation. Research for over 30 years on this formation has repeatedly shown its favourable characteristics for hosting a high-level radioactive waste repository and its effective barrier properties to the migration of radionuclides.

Boom Clay is a marine sediment deposited about 30 My ago. At the Mol-Dessel nuclear site, the Boom Clay lies 186 - 289 m below ground surface. The Boom Clay layer is slightly dipping into the north-north-eastern direction with a slope of about 1 - 2% and increasing thickness with water bearing sand layers situated above and below (Wemaere et al., 2008). Owing to its very low hydraulic conductivity (about 10-12 m/s) and the weak hydraulic gradient (about 0.02 m/m), migration of liquids, dissolved gasses and other solutes in the undisturbed Boom Clay formation is dominated by diffusion (Marivoet et al., 1997).

3.2 Short description of the supercontainer concept and design

The envisaged repository for disposal of vitrified waste consists of a series of rectilinear galleries, in which the waste will be disposed, situated around the Boom Clay midplane. Access to the disposal galleries is provided via the centrally located access gallery, to which all disposal galleries are linked. In its turn, the access gallery is connected to the surface via at least two shafts (ONDRAF/NIRAS, 2006). Because the Boom Clay is a plastic clay, a concrete lining is required to limit convergence of the gallery walls. A sketch of the different components in the envisaged repository system is shown in Figure 1.



Figure 1: Parts of a geological disposal system for vitrified HLW

A key component of the engineered barrier system (EBS) for vitrified high-level waste is the so called BSC-1 supercontainer. This design is based on the Contained Environment Concept (ONDRAF/NIRAS, 2004a; Wickham, 2005a). The supercontainer design with a concrete buffer based on Ordinary Portland Cement (OPC) was selected by ONDRAF/NIRAS through a multi-criteria analysis (ONDRAF/NIRAS, 2004a). A cross-section and longitudinal section of the supercontainer concept is depicted respectively in Figure 2 and Figure 3 (dimensions not to scale). The detailed dimensions of the repository components is given in Figure 4.

In this design, every two vitrified waste canisters are enclosed within a 30 mm thick carbon steel overpack. This overpack will be inserted into a prefabricated cementitious cylindrical buffer (Phase 1 concrete). The cylindrical cavity between the overpack and the buffer will be filled with a concrete filler, called Phase 2 concrete (liquid mortar or similar). The top of the buffer is closed by pouring concrete which forms the sealing plug. Eventually, the annular void between the supercontainer and the disposal gallery lining will be backfilled with cementitious material. It is assumed that supercontainers will be placed end-to-end (without a gap) and that the spacing between neighbouring disposal drifts is 50 m.







Figure 2: Cross-section of the supercontainer-OPC design



Figure 3: Longitudinal section of the supercontainer-OPC design.





Boom Clay	↑ ^r o		
Boom only	overpack length 2	2.771 m	า
	overpack diameter ().532 m	ı
	overpack thickness	30 m	۱m
	thickness of the filler	5 ci	m
gallery lining	supercontainer length	3.973 m	า
	supercontainer diameter	l.955 m	า
cementitious backfill	average backfill thickness	52.25 ci	m
	inner gallery diameter	3 m	า
	thickness of the gallery lining	30 ci	m
OPC concrete buffer	outer gallery diameter	3.6 m	า
filler	r +		
waste and overpack	θ×		

Figure 4: Dimensions of the repository components for vitrified HLW (ONDRAF/NIRAS, 2008)

Figure 2 and Figure 3 show that the waste package is enclosed in a stainless steel liner known as the envelope. This envelope has no function apart from keeping the waste package together and its presence introduces uncertainties in the evolution of the EBS. For this reason, ONDRAF/NIRAS is considering to omit this steel liner, if feasible. In the calculations presented in this report, the presence of the liner is not taken into account.

One of the key arguments in favour of the Contained Environment Concept is that the long-term uniform corrosion rate of the carbon steel overpack is very low in an alkaline environment and localised corrosion phenomena are extremely unlikely. Carbon steel forms a protective passive film in a high-pH environment, such as in OPC-based concrete. This layer consists of ferric oxides which make the steel less prone to local corrosion phenomena and corrosion occurs more or less in a uniform manner.

As such, the supercontainer design is optimised to provide containment of contaminants for as long as possible, but at least during the thermal phase (ONDRAF/NIRAS, 2008). The containment (C) safety function is to be fulfilled by the overpack, while the buffer provides optimal geochemical conditions to keep the steel passivated.

3.3 Approach

As a first step, an assessment of the gas source term is made, using literature data. In several publications (Ortiz, 2002; Johnson, 2006), anaerobic corrosion is reported to be the largest contributor to the gas source term. As there is no gas generation associated with the corrosion reaction in aerobic conditions, it is first of importance to have an idea of the timescale during which oxygen (oxidising conditions) is present in the vicinity of the overpack. Hydraulic calculations estimating the time until all EBS materials are water saturated will help in indicating





the onset of H_2 generation¹. Complementary, other sources of gas generation will be reassessed, using the most up-to-date literature data, in order to have the complete picture of gas production mechanisms and their relative importance.

The second step consists of evaluating whether the diffusive gas migration through the various near field materials is fast enough to prevent the build-up of large gas pressures. First, it will be evaluated whether a build-up of a free gas phase will occur, by comparing gas production rates with the rate of its diffusive removal from the near field. Should there be a clear indication that the gas production exceeds the diffusive gas dissipation capacity of the near field, more detailed system calculations are needed to evaluate the consequences in terms of maximum near field gas pressures and degree of local desaturation. As these are complex calculations involving a number of poorly known input parameters and material characteristics, the emphasis is on insight modelling. Varying parameter values makes us familiar with the sensitivities of the system and using different numerical tools allows us to build confidence in these tools and evaluate the consequences of some conceptual uncertainty. However, there is still a need for model validation against experimental results. These are expected to become available in the framework of the EC FORGE project, which is entirely dedicated to the problem of gas generation in geological disposal facilities and which runs from 2009 until 2013.

The third step involves two-phase calculations with the actual hydrogen generation rate. Anaerobic corrosion of steel assumes Arrhenius behaviour, which introduces the effect of temperature to chemical reaction rates. Therefore the link between gas and heat generation rates will be established. Heat generation affects not only gas production rate, but also the general behaviour of the system through temperature dependent parameters. The calculations will provide a useful insight into a system performance when exposed to coupled gas-heat generation processes.

Since some of the engineered barriers (and even Boom Clay) are sometimes poorly characterised in view of the governing processes in this study, broad ranges for parameter values are used as much as possible in order to enhance the validity of these exploratory calculations, especially in view of the Belgian research programme on geological disposal. Previously reported sensitivity calculations (Weetjens and Sillen, 2006) identified, apart from the corrosion rate, the following parameters as having a large influence on the evolution of the gas pressure field: the diffusion coefficient of H_2 in the aqueous phase, the hydraulic conductivity K of the buffer and its gas entry pressure. Bounding values covering a reasonable range will be applied for these parameters.

¹ Please note that the saturation time is only roughly indicative for the duration of oxidising conditions: O_2 could be consumed earlier due to oxidation of clayey materials and other NF materials leaving only N_2 . On the other hand, it might be possible that oxidising conditions continue to exist after complete saturation, due to remnants of dissolved oxygen, or additional production of oxidising species due to radiolysis.





4. Estimation of the gas source term

4.1 Anaerobic corrosion of the overpack

4.1.1 Estimation of timescale of transition from oxidising to reducing conditions

After supercontainer emplacement, the annular void spaces between the supercontainer and gallery lining will be backfilled with a cementitious material. A fraction of the pore volume in the EBS materials contains trapped air. Hence, redox conditions will initially be oxidising.

Water provided by the Boom Clay will gradually flow into the backfilled galleries and start to saturate the EBS materials. Trapped air will gradually dissolve into the pore water. When all pores are water saturated and the dissolved oxygen concentration (as well as the concentration of other oxidising species possibly present) is small enough, the redox condition in the vicinity of the overpack will become reducing, marking as such the onset of the anaerobic corrosion process. The oxidising conditions can be prolonged due to radiolysis of H₂O in the buffer concrete, producing additional oxidising species. However, it is expected that these species are very reactive. Relying on the prospect that oxygen (oxidising species) will soon be depleted once in saturated conditions, the timescale of the transition from oxidising to reducing conditions can be assessed by computing the time needed for full saturation. This was done by performing variably saturated flow calculations.

These hydraulic calculations are performed with the numerical flow and transport code PORFLOW v3.07 (Runchal, 1997) in a 1D radial geometry and the multiphysics code COMSOL Multiphysics (COMSOL AB, 2005) in a 2D radial geometry. The dimensions of a disposal cell is based on Figure 4 and the applied material properties are listed in Table I in the next chapter.

Obviously the saturation time depends on the (currently uncharacterised) initial saturation degree of the EBS materials as well. To assess the impact of initial conditions on the timescale of saturation, calculations were performed using an initial saturation of 40%, 60% and 80%.

The time until saturation depends on the hydraulic properties of the materials and the prevailing hydraulic gradient in the clay around a disposal gallery. In these calculations, the time between gallery excavation and backfilling is arbitrarily assumed to be two years. However, a pore water pressure profile corresponding to three years pure drainage is implemented, because this can be considered as the in-situ pressure situation after two years, since the pressure drop due to instantaneous mechanical convergence upon excavation corresponds to about one year drainage.

Concerning the hydraulic properties of the buffer, an intrinsic permeability (k) value of $4 \times 10^{-21} \text{ m}^2$ was reported in Wickham (2005b). This corresponds approximately to a saturated hydraulic conductivity K_s of 4×10^{-14} m/s, which is very low. As an alternative value, a K_s around 4×10^{-12}





m/s was selected, which is approximately the same as the hydraulic conductivity of Boom Clay. Both values are applied in the calculations.

Results in terms of pressure head and saturation profiles for different initial conditions are depicted in Figure 6 for the case of a very low permeability buffer ($K_s=4\times10^{-14}$ m/s) denoted as "low-K buffer" and in Figure 9 for the case of a buffer permeability comparable to Boom Clay, denoted further as "clay-K" buffer ($K_s=4\times10^{-12}$ m/s).

The evolution in time of pressure head and saturation close to the overpack is shown in Figure 7 and Figure 8 respectively for the low-K buffer and in Figure 10 and Figure 11 for the clay-K buffer.

These results indicate that it could take approximately 4 to 16 years, depending on initial conditions, to reach full saturation close to the overpack. Should the permeability of the buffer be very low, it may take 20 to 80 years. The 2D radial model calculated with COMSOL agrees very well with the simplified 1D radial PORFLOW model, which means that the additional time needed to saturate the inner concrete buffer part at the supercontainer axis is negligible.

Coupled thermohydraulic calculations performed in the framework of the EC sponsored NF-PRO project (Weetjens *et al.*, 2006) show that the computed saturation time is about two times smaller for a simulation that takes the thermal evolution into account, in comparison to purely hydraulic calculations. However, the coupling was limited to the effect of temperature on viscosity, which enhances the permeability. The temperature influences the surface tension as well, which has an effect on the capillary suction characteristics (effect on the α parameter in the van Genuchten relationship). This effect counteracts the viscosity effect somewhat but not to the extent that they compensate each other, based on the small variation in surface tension with temperature.

The dissolution of air or oxygen into the pore water (as prescribed by Henry's law) was not taken into account in these calculations, but sensitivity calculations performed for the BENIPA project showed that air dissolution and diffusion rates are high enough not to prolong the saturation time. (BENIPA, 2003)



Figure 5: COMSOL 2D and PORFLOW 1D model geometries applied in the resaturation calculations. Red dots show the location of the 'observation' nodes where computed saturation and pressure head are followed-up.







Figure 6: Pressure head and saturation profiles along the midplane of the supercontainer (1D radial PORFLOW calculation). Case with a low-permeability buffer ($K_s=4\times10^{-14}$ m/s). Top: 80% initial saturation, mid: 60% initial saturation and bottom: 40% initial saturation.







Figure 7: Evolution of the pressure head at the overpack (buffer $K_s=4\times10^{-14}$ m/s) for different initial saturation degrees (PORFLOW 1D radial versus COMSOL 2D radial geometry)



Figure 8: Evolution of the saturation at the overpack (buffer $K_s=4\times10^{-14}$ m/s) for different initial saturation degrees (PORFLOW 1D radial versus COMSOL 2D radial geometry)







Figure 9: Pressure head and saturation profiles along the midplane of the supercontainer (1D radial PORFLOW calculation). Case with a clay-permeability buffer ($K_s=4\times10^{-12}$ m/s). Top: 80% initial saturation, mid: 60% initial saturation and bottom: 40% initial saturation.







Figure 10: Evolution of the pressure head at the overpack (buffer $K_s=4\times10^{-12}$ m/s) for different initial saturation degrees (PORFLOW 1D radial versus COMSOL 2D radial geometry)



Figure 11: Evolution of the saturation at the overpack (buffer $K_s=4\times10^{-12}$ m/s) for different initial saturation degrees (PORFLOW 1D radial versus COMSOL 2D radial geometry).





The propagation of the waterfront is influenced by the way the hydraulic conductivity is averaged at the interface between computational cells. In PORFLOW 3.07, upwind averaging of the conductivity was imposed explicitly. Differences in results between PORFLOW 3.07 and COMSOL could be a consequence of different averaging methods.

4.1.2 Estimation of the gas production rates

Once anaerobic conditions prevail in the near field environment, anaerobic corrosion of the steel components in the EBS will be a dominant source of hydrogen gas. Anaerobic corrosion can be described by the following reaction, which involves production of magnetite:

$$3 \operatorname{Fe}_{(s)} + 4 \operatorname{H}_2O \leftrightarrow \operatorname{Fe}_3O_{4(s)} + 4 \operatorname{H}_{2(g)}$$
(1)

The evolution of hydrogen generation due to corrosion is very concept dependent. Obviously, the corrosion rate is dependent on the local chemical conditions. Furthermore, the corrosion rate, and hence the rate of hydrogen production is directly proportional to the exposed surface area of the steel EBS components. The steel barrier thickness determines the duration of the gas generation process. Moreover, the surface area between the steel components and the surrounding porous media also determines how effectively the hydrogen can be dissipated by diffusion. Thus, the interplay of both phenomena determines whether gas will be accumulated in the near field or not. For a thin steel plate, the hydrogen gas generation rate can be estimated by (Volckaert and Mallants, 1999):

$$v_{H_2} = S \times R \times 1000 \times \rho \times m \times \frac{f_{Fe}}{M_{Fe}},$$
(2)

with:

 v_{H2} = gas generation rate (mole/year)

S = surface area (m²)

R = corrosion rate (m/year)

 ρ = density of steel (kg/m³) = 7900 kg/m³

m = stoechiometric factor = 4/3 for corrosion reaction [1]

 M_{Fe} = molecular weight of iron = 55.85

 f_{Fe} = fraction of iron in the steel (0.99 for C-steel)

In this study, only the corrosion of the 30 mm thick carbon steel overpack is considered, as it is still unclear if an envelope is required. Recent calculations (Weetjens *et al.*, 2006; Weetjens and Sillen, 2006) taking the presence of the envelope into account showed that, despite its larger surface area, gas production due to corrosion of the envelope is not significant. This can be





explained by the value of the best estimate corrosion rate of stainless steel, which is fairly small (0.01 μ m/year; Kursten *et al.*, 2004) and the larger surface area for diffusion, which makes that H₂ gas due to corrosion of the envelope can be evacuated by diffusion as a dissolved species.

In general, one of the key arguments in favour of the supercontainer concept is that corrosion rates of steel are very small in a highly alkaline environment. For the corrosion of the C-steel overpack, the longterm uniform corrosion rate is believed to be < 0.1 μ m/year (Kursten *et al.*, 2008; Kursten *et al.*, 2004; ONDRAF/NIRAS, 2004) based on *a.o.* passive current density measurements (Azizi *et al.*, 2008; Macdonald *et al.*, 2006). This value is currently considered as an "upper limit". Other techniques, *e.g.* gas volume measurements (N. Smart *et al.*, 2004) indicate that the long-term uniform corrosion rate could even be as low as 0.01 μ m/year. Based on the saturation calculations discussed in paragraph 4.1.1, full hydraulic saturation could be reached in a couple of years. At that time, near field temperatures are at their maximum close to the overpack (between 90 and 100°C) due to radiogenic heat generation in the waste. It is reasonable to assume that the corrosion rates are temperature dependent. For carbon steel in alkaline media, values of 1 μ m/year at 80°C and 0.1 μ m/year at 30°C were found in the literature (Kursten *et al.*, 2004; ONDRAF/NIRAS, 2004). Based on these data and application of the Arrhenius law, temperature dependent corrosion rates can be implemented for the relevant temperature range in a vitrified HLW disposal gallery. This will be treated as a variant case.

Applying the formerly mentioned corrosion rates, the maximum hydrogen generation rate can be calculated. Initially, about 1.7 moles/year H₂ would be produced per supercontainer in the temperature dependent case and 0.1 moles/year in case of a constant uniform corrosion rate of 0.1 μ m/year (see Figure 12). The theoretical total amount of gas produced would be 594 m³ (STP 0°C, 1 atm) and gas generation would be spread over at least 300 000 years.







Figure 12: Hydrogen source term from anaerobic corrosion of the overpack as a function of time for different corrosion rates. The data correspond to one supercontainer.

4.2 Other sources of gas

In general, anaerobic corrosion of steel EBS components is found to be the most significant source of gas production within the near field of a repository for vitrified HLW. This is, amongst others, reported in publications of Volckaert and Mallants (1999) Ortiz *et al.* (2002) and Johnson (2006). A short overview of other potential sources for gas and their relative importance in case of vitrified HLW disposal in supercontainers is listed below.

4.2.1 Microbial degradation

Gas generation from microbial degradation of organic wastes is only an issue for certain types of ILW and LLW (Rodwell *et al.*, 2003). This mechanism for gas generation is considered to be irrelevant for HLW disposal.

4.2.2 Radiolysis

Radiolysis is a process which causes dissociation of molecules due to ionising radiation. Gas generation due to radiolysis can be due to α , β or γ radiation and depends on the absorbed energy. As α and β radiation are stopped by the overpack, only radiolytic gas generation due to γ radiation is considered. Bouniol (2005) studied the gas generation due to γ radiolysis in the buffer of a supercontainer. The H₂ concentration in the buffer pore water was estimated to 8×10⁻⁴ mol/dm³ after 100 years in a closed and saturated system. Since the pore water volume in the buffer is about 1.34 m³ (using a porosity of 10.4%), the amount of H₂ produced by radiolysis in the buffer could be estimated to about 1 mol after 100 years, or an average production rate of





 1×10^{-2} mole/year. This corresponds to 10% of the hydrogen production due to anaerobic corrosion of the overpack, assuming a corrosion rate of 0.1μ m/year.

4.2.3 Radioactive Decay

Radioactive decay will produce helium gas (owing to alpha decay) and radon gas (mainly owing to ²²⁶Ra, itself a decay product of the ²³⁸U chain), (Mallants *et al.*, 2004; Ortiz *et al.*, 2001). Both gases will be generated inside the waste matrix. It is assumed that there is some gas storage capacity inside the canister and that the tensile strength of the overpack is sufficient to accommodate a potential internal gas pressure increase. As such, gas generation *inside* the waste is assumed not to cause safety problems. However, this gas is instantaneously released upon perforation and could constitute an additional gas source term at a certain point in time.

The importance of He gas production can be easily evaluated considering the amount of α emitters present in the vitrified HLW inventory. This was done in the framework of the NF-PRO project and reported in Weetjens *et al.* (2006). The amount of helium produced by decay was estimated to 0.5×10^{-2} moles/year per canister or 1×10^{-2} mole/year per supercontainer, which corresponds to 10% of the best estimate hydrogen production due to anaerobic corrosion.

Although constantly generated in the waste due to decay of Ra isotopes, the gaseous Rn isotopes will decay very quickly into metallic elements (half-lives of ²²²Rn, ²²⁰Rn and ²¹⁹Rn are 3.8 days, 55.6 seconds and 4.0 seconds respectively). Hence, the Rn gas volume generated in the waste is negligible.





5. Gas generation and dissipation modelling studies

5.1 Assessment of the diffusive removal of dissolved H₂ gas

One possible way for the generated hydrogen to move away from the source is to dissolve into the near field pore water and be transported away from the source by diffusion into the Boom Clay pore water. As the water flow rate through the low-permeability Boom Clay is very small, advection is negligible compared to diffusion. The gas solubility is assumed to be linearly dependent on the corresponding partial pressure of the gas, following Henry's law:

$$X_{aq} = k_H(H_2) \times P(H_2), \tag{3}$$

with $k_H(H_2)$, Henry's constant for hydrogen, 7.8×10^{-4} mole/(kg×bar) (at 298.15 K), and P(H₂) the partial pressure of hydrogen. The geological repository is situated at the Boom Clay midplane, where the in-situ hydrostatic pressure is 22 bar (23 bar absolute). The amount of hydrogen that can dissolve in the pore water at that pressure is then 0.018 mole/l.

Currently, a new research programme is running (2009) to accurately determine the diffusion of hydrogen in Boom Clay. In this research programme, a sophisticated experimental set-up is being developed to measure diffusive H₂ transport in-situ. In addition, old measurements executed in the framework of the MEGAS project (Volckaert *et al.*, 1995) are being reinterpreted. Until new data become available, we assume an apparent diffusion coefficient for dissolved H₂ of 5×10^{-10} m² s⁻¹. In order to assess the sensitivity to this parameter, a conservative value of 5×10^{-11} m² s⁻¹ was also tested.

A series of 1D radial transport simulations have been carried out with the numerical code PORFLOW v3.07 (Runchal, 1997) to calculate the largest possible diffusive hydrogen fluxes from the near field into the Boom Clay. To that aim, we set a constant concentration boundary condition at different positions, starting from the overpack radius and moving outwards through the EBS and host rock. The concentration was set equal to the hydrogen solubility at in-situ pressure i.e. 0.018 mole/I, and we calculate the diffusive fluxes in function of time at the selected radius. Results in terms of cumulative flux are compared to the cumulative H₂ production in Figure 13 for a H₂ diffusion coefficient (D_{H2}) of 5×10^{-10} m² s⁻¹ and in Figure 14 for a D_{H2} of 5×10^{-11} m² s⁻¹.



Figure 13: Maximal cumulative diffusive migration (with $D_{H2}=5\times10^{-10}$ m² s⁻¹) of dissolved H₂ versus cumulative H₂ production.



Figure 14: Maximal cumulative diffusive migration (with $D_{H2}=5\times10^{-11}$ m² s⁻¹) of dissolved H₂ versus cumulative H₂ production.





Figure 13 shows that only in case of a temperature dependent gas production, the cumulative diffusive migration is not sufficient, for a relatively short period of time, to evacuate all the gas within the EBS (at the overpack and in the supercontainer buffer). Likewise, it can be seen from Figure 14 that, in the short term, only the EBS will be subject to increased gas pressures in case of a temperature dependent gas production. In this case however, also in the longer term, increased gas pressures will dominate the EBS, because the concentration gradient in the Boom Clay has flattened and the guasi-steady-state diffusive migration is not sufficient to evacuate the produced gas within the EBS (up to the interface with Boom Clay). In general, these figures show that the cumulative hydrogen production does not exceed the capacity of the Boom Clay to evacuate the dissolved gas by diffusion in pore water for both diffusion coefficient values. However, due to the lower cumulative flux in the long term, the required surface area for diffusion increases up to the radius of r=1.8 m, which is at the EBS/Boom Clay interface. Furthermore, it is possible that a free gas phase is generated soon after repository closure within the EBS. It should be noted that this is a oversimplified and conservative calculation at constant pressure, while in reality gradually more and more hydrogen could dissolve in the pore water if the pressure increases. To evaluate the possible gas pressures attained in the near field and the corresponding degree of desaturation, a complementary two-phase-flow analysis is necessary.

5.2 Multiphase flow analysis

5.2.1 Phenomenological description

Due to anaerobic corrosion of the steel EBS materials, hydrogen will be released in the near field. In the beginning, this hydrogen will dissolve in the pore water and diffuse through the EBS materials towards the host formation. Once the gas concentration in the pore water reaches the gas solubility, a free gas phase will appear. It is important to note that the existence of a two-phase system, *i.e.* water-gas, does not necessarily imply two-phase flow. Gas flow will only happen when the gas saturation is large enough to displace pore water, or equivalently, when the gas pressure exceeds the gas entry pressure of the saturated porous medium. These values depend on the characteristics of the capillary pressure / liquid saturation function, also called 'water retention curve', of the considered porous material.

When the gas pressure approaches the in-situ lithostatic pressure, the solid skeleton will experience increasingly large mechanical deformations and eventually, fractures may appear. The gas may then escape through a preferential flowpath, which will most likely be situated along the Excavation Disturbed Zone (EDZ).

It has been observed in the MEGAS experiment that, when the pressure in the gas phase has dropped below the lithostatic pressure (after a gas breakthrough), the preferential flowpaths close again and the original flow properties of the Boom Clay seem to be restored (Volckaert *et al.*, 1995).





5.2.2 Simulation tool: TOUGH2

The gas calculations are performed with the TOUGH2 code (Pruess, 1999; Pruess, 2004) using PetraSim as graphical interface. TOUGH2 is a numerical simulator for nonisothermal flows of multicomponent, multiphase fluids in one, two, and three-dimensional porous and fractured media. The chief applications for which TOUGH2 is designed are situated in geothermal reservoir engineering, nuclear waste disposal, environmental assessment and remediation, and unsaturated and saturated zone hydrology.

PetraSim is a graphical interface for TOUGH2, T2VOC, TMVOC, TOUGHREACT, TOUGH-Fx/HYDRATE, and TETRAD.

5.2.3 Conceptual model

5.2.3.1 Model abstraction and geometry

The considered repository lies 220 m deep in Boom clay and consists of several components as depicted in Figure 5. The supercontainer and galleries are shaped cylindrically and could be thus represented in a quasi 3D axi-symmetrical computational domain. By this simplification we neglect the effect of gravity which in reality makes the problem solution non axi-symmetrical and forces hydrogen towards the surface. In addition, potential material anisotropy, especially relevant for the Boom Clay, could also not be modelled in radial symmetry.

In the z-direction only half of the supercontainer is modelled due to geometrical symmetry (Figure 15). The axi-symmetrical rotational axis is at r=0. The modelled domain is taken to be $0 \text{ m} \le r \le 40 \text{ m}$ and $0 \text{ m} \le z \le 1.9865 \text{ m}$.







Figure 15: Axi-symmetrical model discretization with locations where the calculated primary values are recorded (here denoted as 'observation' nodes).

The computational domain is discretized by 40 elements in the z-direction and 79 elements in the r-direction.

5.2.3.2 Boundary and initial conditions

The model is bounded by four boundaries. The assumption is, that 40 m vertical distance will provide sufficient length so that the primary variables could be fixed to constant value at the outer radial (r=40 m) boundary. Pressure and temperature are therefore fixed to $P(t_0)=2.2$ MPa and $T(t_0)=15.7$ °C, respectively. All other boundaries are defined as no flow boundaries due to symmetry properties of the model as shown in Figure 16.

TOUGH2 defines Dirichlet boundary conditions through application of a large volume at the fixed boundary elements (arbitrary value 10⁵⁰ m³ used). Since the volume is very large, changes of state variables are equivalently very small. No flow boundaries are defined very simply by no





connection between elements. Hence any non-connected element represents a no-flow boundary condition.

The simplified initial pressure conditions are divided into two parts. In the Boom clay a constant initial pressure of 2.2 MPa is assumed (denoted by red colour in Figure 16), whereas all engineered barriers (lining, backfill, buffer, waste and overpack) are at normal atmospheric pressure 0.101325 MPa (denoted by blue colour in Figure 16). The temperature and initial saturation degree are constant: 15.7 $^{\circ}$ C and 1 [-], respectively.



Figure 16: Boundary and initial conditions for two-phase flow calculations.





5.2.3.3 Material properties

In the absence of experimental data almost all parameter values in Table I are based on "best guesses" from literature data (for references: see Weetjens, 2007).

Table I: Material properties used in calculations.

		Unit	Waste	Boom Clay	Lining	Backfill	Buffer	Second phase concrete
solid density	$ ho_{s}$	kg/m ³	7850	2650	2650	2650	2650	2650
porosity	η	-	0	0.391	0.104	0.3	0.104	0.3
permeability	kı	m²	0	4.6E-19	4.6E-17	4.6E-17	4.6E-19*	4.6E-17
Klinkenberg parameter	b	MPa	0	7.0	7.0	7.0	7.0	7.0
pore compressibility	$eta_{\scriptscriptstyle P}$	MPa⁻¹	0	7.50E-3	3.83E-5	3.83E-5	3.83E-5	3.83E-5
			relative	permeability	curve			
shape parameter	λ	-		0.355	0.430	0.430	0.430	0.430
residual w saturation	S_{lr}	-		0.20	0.25	0.25	0.25	0.25
maximum w saturation	S_{ls}	-		1.00	1.00	1.00	1.00	1.00
residual g saturation	S_{gr}	-		0.174	0.20	0.20	0.20	0.20
			capillar	y pressure fur	nction			
shape parameter	λ	-		0.355	0.430	0.430	0.430	0.430
residual w saturation	S_{lr}	-		0.012	0.20	0.20	0.20	0.20
maximum w saturation	S_{ls}	-		1	1	1	1	1
shape parameter	α	m⁻¹		3.47E-3	1.96E-2	1.96E-2	1.96E-2	1.96E-2
air entry pressure	P_0	MPa		2.83	0.50	0.50	0.50	0.50
	heat transport							
thermal conductivity	$\lambda_{_T}$	$\operatorname{Wm}_{1}^{-1}K^{-}$		1.7	1.7	1.0	1.7	1.0
specific heat	C_p	Jkg⁻¹K⁻		1100	800	800	800	800

* Alternative set of calculations with two orders of magnitude lower permeability is considered in some calculations (see Table IV).





5.2.3.4 Gas source term

The dominant source of pressure is due to anaerobic corrosion of steel components and consequent hydrogen production as described in Chapter 4. In our model the gas source is located in the second phase concrete filler. In fact, hydrogen is generated at the boundary between overpack and second phase concrete. For practical reasons, however, it is assigned to the second phase concrete as it is much more convenient to enter a volumetric source into PetraSim (TOUGH2). Differences between the two approaches are assumed not to have any significant effect since the permeability of the second phase concrete is two orders of magnitude larger as the concrete buffer. The location of the gas source is shown in Figure 17.



Figure 17: Location of the gas source (in red).

Three approaches how to treat source term are considered:

- A constant hydrogen production based on a fixed corrosion rate. Preliminary calculations have shown that no gas phase is formed using the upper limit for long-term uniform corrosion rate of 0.1 μm/year, assuming no effect of temperature on the corrosion rate. Therefore, as a conservative test case, a constant corrosion rate of 1 μm/y was assumed.
- The second approach assumes again constant values over the three time periods. The first time period extends from the beginning time of gas production until 100 years. The corrosion rate of the steel overpack during the first period is 1 µm/y since in this period higher temperatures are present. The second period extends until 300000 years using a





long-term corrosion rate of 0.1 μ m/y. After that, 30 mm of overpack is gone and therefore the corrosion rate becomes 0.

 By application of the Arrhenius law, the corrosion rate becomes temperature dependent and hence time dependent through knowledge of the temperature evolution at the source location. From thermal calculations done separately we are able to extract temperatures in the second phase concrete. Unfortunately a direct link between thermal calculations and the gas source is not possible within TOUGH2 for now.

All types of sources are graphically presented in Figure 18.



H2 gas production due to anaerobic corrosion of the overpack

Figure 18: Three types of sources considered in the two-phase flow calculations.

5.2.3.5 Thermal source

The heat formed in the vitrified HLW is well approximated by a formula derived by Put (Put and Henrion, 1992, p. 153). It can be written as

$$Q = \sum_{i} A_{i} e^{-\lambda_{i} t} \quad (Q \text{ in W/tHM}), \tag{4}$$

with the coefficients A_i and λ_i defined in Table II and t the time after waste production (i.e. vitrification). As can be seen from Figure 19, Put's formula is in good agreement with calculations made using the ORIGEN code and independent results from NAGRA and JNC (H-12 report, JNC, 2000). In the following calculations, a cooling time of 60 years was considered.





It is assumed that per initial tonne Uranium metal, 0.75 tonnes of vitrified HLW are produced in the reprocessing facility.

Table II: Coefficients for equation [1] in case of vitrified HLW.

A_1	A_2	A_3	A_4	A_5
5021	1205	27.04	0.7576	0.1
λ_{I}	λ_2	λ_3	λ_4	λ_5
3.894E-01	2.458E-02	1.63E-03	6.546E-05	0.00E+00



Figure 19: Vitrified HLW heat production (from Sillen and Marivoet, 2007).

In our numerical model the thermal source is located in a waste region as shown denoted in red colour in Figure 20.






Figure 20: Location of the thermal source (in red).

5.2.4 TOUGH2 Mathematical model

5.2.5.1 Two-phase modelling

The two-phase model is described by two sets of mass conservation equations i.e. for wetting (subscript w) and non-wetting phase (subscript nw).

$$\theta_{s} \frac{\partial Se_{w}}{\partial t} + \nabla \cdot \left(-\frac{\kappa_{int}k_{r,w}}{\eta_{w}} \nabla \left(p_{w} + \rho_{w}g \cdot \vec{k} \right) \right) = \dot{S}_{w}$$
(5)

$$\theta_{s} \frac{\partial Se_{nw}}{\partial t} + \nabla \cdot \left(-\frac{\kappa_{int}k_{r,nw}}{\eta_{nw}} \nabla \left(p_{nw} + \rho_{nw}g \cdot \vec{k} \right) \right) = \dot{S}_{nw}$$
(6)

 θ_s represents saturated water content, which is equal to porosity in our case. *Se* is the effective saturation function. κ_{int} is the intristic permeability [m²], k_r is the relative permeability function for a given fluid, η is dynamic viscosity of the fluid [kg m⁻¹ s⁻¹], p is the pressure [kg m⁻¹ s⁻²], ρ is the fluid density [kg m⁻³], g is acceleration of gravity [m s⁻²], \vec{k} is a vector pointing to the direction of gravity and t is time [s]. Gravitation in our case is omitted due to radial symmetry of the problem.





Each phase is characterized by different retention relationships. In our case van Genuchten relations are used. For the wetting phase the relations read

$$S_{l} = \begin{cases} S_{lr} + Se_{w} \cdot (S_{ls} - S_{lr}) & H_{c} > 0 \\ S_{ls} & H_{c} \le 0 \end{cases}$$

$$Se_{w} = \begin{cases} \frac{1}{\left(1 + \left|\alpha \cdot H_{c}\right|^{n}\right)^{\lambda}} & H_{c} > 0 \\ 1 & H_{c} \le 0 \end{cases}$$

$$k_{r,w} = \begin{cases} Se_{w}^{L} \left(1 - \left(1 - Se_{w}^{1/\lambda}\right)^{\lambda}\right)^{2} & H_{c} > 0 \\ 1 & H_{c} \le 0 \end{cases}$$
(7)

 α , n, λ and L are van Genuchten shape parameters which define soil properties where $n = \frac{1}{1-\lambda}$. Normally the assumption is that L=0.5. Effective water saturation S_{ew} is calculated from residual water content S_{tr} and saturated water content S_{ts} . Effective water saturation and capillary pressure are linked through capillary head H_c [m] in equation (7). Capillary pressure p_c could be then calculated as

$$p_{c} = -\frac{\rho_{w} \cdot g}{\alpha} \left(Se_{w}^{-1/\lambda} - 1 \right)^{1/n}, \tag{8}$$

where $\frac{\rho_w \cdot g}{\alpha}$ represents air entry pressure in the TOUGH2 code.

Relations for the non-wetting phase are

$$S_{g} = S_{ls} - S_{l}$$

$$Se_{nw} = 1 - Se_{w}$$

$$k_{r,nw} = \begin{cases} \left(1 - \hat{S}e_{w}\right)^{2} \left(1 - \hat{S}e_{w}^{2}\right) & H_{c} > 0 \\ 0 & H_{c} \le 0 \end{cases}$$

$$\hat{S}e_{w} = \frac{S_{l} - S_{lr}}{1 - S_{lr} - S_{gr}}$$
(9)





5.2.5.2 Variables

Variables used for this type of calculations can be divided into primary and secondary ones. All secondary variables can be derived from the primary variables. The choice of the primary variables that define the thermodynamic state of the system is an important consideration in the modelling of fluid and heat flow processes with phase changes. When a phase appears or disappears, the set of appropriate thermodynamic variables may change. In single-component flows involving water, for example, appropriate thermodynamic variables for describing singlephase conditions (subcooled liquid or superheated steam) are temperature T and pressure P. However, in two-phase conditions pressure and temperature are not independent, but are related by the vapour-pressure relationship $P = P_{sat}(T)$. There are two alternative ways for dealing with this problem. One possibility is to use a set of "persistent" variables such as (pressure, enthalpy) or (density, internal energy), which remain independent even as phase conditions change, so that they can be used throughout the single- and two-phase regions. A drawback of this approach is that parametric relationships for thermophysical properties are usually formulated in terms of the "natural" variables pressure and temperature, so that their computation as functions of "persistent" variables becomes either more difficult (requiring solution of implicit equations) or entails some sacrifice in accuracy. An alternative possibility is to use the variables (pressure in the liquid phase P, the mass fraction of hydrogen in the liquid phase X_{HydL} and the temperature T) only for single-phase conditions, and to "switch" to variables (pressure in the gas phase P_{g} , the gas saturation S_{g} and the temperature T) when a transition to two-phase conditions occurs. Experience has proven variable switching to be a very robust method for treating multiphase systems, and this is the approach used in most TOUGH2 modules (TOUGH2, 1999).

5.2.5.3 Computation of the hydrogen mass fraction in the liquid phase

The primary variable P_g is composed of two partial pressures, namely the hydrogen pressure P_{hyd} and the pressure of water vapour P_{vap} . Both partial pressures are temperature dependent and treated as an ideal gas. The pressure of water vapour P_{vap} is calculated internally in TOUGH2 from water tables. After subtraction of P_{vap} from P_g calculation of X_{HydL} is derived from P_{hyd} as

$$X_{HydL} = \frac{X_{HydL}^{m}}{X_{HydL}^{m} \cdot M_{H_{2}} + (1 - X_{HydL}^{m} \cdot M_{H_{2}}) \cdot M_{H_{2}O}} \text{ [g H_{2}/g liquid],}$$
(10)

where M_{H_2} and M_{H_2O} are the molar mass of hydrogen and water respectively, and mole fraction in the liquid phase $X_{H_2H}^m$ is calculated from

$$X_{HydL}^{m} = K_{H} \cdot P_{hyd} \text{ [mole H}_{2}/\text{mole liquid]}.$$
(11)

Henry's constant K_H is a temperature dependent parameter which is interpolated linearly with temperature as





$$K_{H} = 1.697 \cdot 10^{-10} - 1.272 \cdot 10^{-12} \cdot T[^{0}C] \text{ [mole hydrogen/(mole liquid \times Pa)]}, \tag{12}$$

Equation (12) is valid until the temperature of 25° C. For higher temperatures a constant value for 25° C is assumed.

5.2.5.4 Computation of the hydrogen mass fraction in the gas phase

The calculation of hydrogen mass fraction in the gas phase is straightforward by dividing the density of the hydrogen by the total density of water vapour and hydrogen

$$X_{HydG} = \frac{\rho_{hyd}}{\rho_{hyd} + \rho_{vap}},$$
(13)

where the density of water vapour [kg m⁻³] is calculated internally in TOUGH2 and the density of hydrogen obeys the ideal gas law which gives

$$\rho_{hyd} = \frac{P_{hyd} \cdot M_{H_2}}{R \cdot T}, \qquad (14)$$

with temperature T [K] and universal gas constant R [Pa m³ mol⁻¹ K⁻¹].

5.2.5.5 Temperature and pressure dependence of parameters

State variables are normally dependent on pressure and temperature. Part of the calculations in this report are carried out with a thermal source due to radioactive decay which heats the surrounding material considerably. Therefore both, pressure and temperature dependence are taken into account.

The dependence of the diffusion coefficient in the gas phase on P and T takes the following form

$$D_i(P,T) = D_i(P_0,T_0) \frac{P_0}{P} \left[\frac{T + 273.15}{273.15} \right]^{\omega},$$
(15)

where $D_i(P_0, T_0)$ is the diffusion coefficient at standard conditions $P_0 = 1.01325$ bar and $T_0 = 0^{\circ}$ C. ω is taken to be 1.8 as this is default value in the TOUGH2.

The hydrogen density is calculated through the ideal gas law as described in equation (14). The viscosity of hydrogen is linearly interpolated between values given in Table III.





Table III: Viscosity of hydrogen [in Pas].

	T=0°C	T=100 ⁰ C	
P=1 bar	$8.40 \cdot 10^{-6}$	$10.33 \cdot 10^{-6}$	
P=100 bar	$8.57 \cdot 10^{-6}$	$10.44 \cdot 10^{-6}$	

The hydrogen solubility in water is determined by Henry's constant (11) which at 1 bar gives

T=0°C	$1.92 \cdot 10^{-6} \text{ g H}_2 / \text{g H}_2 \text{O}$
T=25°C	$1.54 \cdot 10^{-6} \text{ g H}_2 / \text{g H}_2 \text{O}$

As mentioned previously, the hydrogen solubility remains $1.54 \cdot 10^{-6} \text{ g H}_2/\text{ g H}_2\text{O}$ for temperatures above 25[°]C, which is not conservative in terms of free hydrogen gas appearance. The effect of this simplification within TOUGH2 is assessed and discussed in paragraph 5.3.

Water characteristics are intrinsically calculated by TOUGH2 from tables.

5.2.5 Calculation cases

The calculation cases are divided according to their complexity and physical phenomena involved. The basic calculation involves only two-phase mass transport with constant hydrogen production. The calculation is then extended to the pressure analysis of the system exposed to hydrogen production with a stepwise constant source term at different time periods. This stepwise constant source term is the first approximation to the temperature dependent hydrogen production. To increase the level of realism, the model is then expanded by a more realistic time-dependent source term to show the difference between models used in previous studies and a new Arrhenius law gas production. Most comparisons are made only for two extreme cases, that is for the constant and Arrhenius law based case.

The pressure analysis models are expanded by coupling with thermal transport. Heat is generated by radioactive decay in the vitrified HLW embedded in the supercontainer. A comparison will show the effect of temperature increase on the system.

Finally, one sensitivity analysis is made regarding the concrete buffer permeability. As for the resaturation calculations of Chapter 4.1.1, the effect of a (very) low permeability buffer was also tested.

In total, 7 cases are considered and denoted as shown in Table IV.





Table IV:	Calculation	cases	considered.

Gas source	Heat source	Permeability	
H=Const	Q=0	high	
		low	
	Q=0		
H=Step	Q=f(t)	high	
	Q=0	high	
H=f(t)	Q=f(t)	high	
		low	

5.2.6 Results

Different cases shown in Table IV will be elaborated depending on the source formulation, introduction of heat source and the permeability of the concrete buffer. Cases with a simplified source will be presented just for comparison, whereas cases with more realistic pressure and thermal source will be analyzed in more detail.

Further division is made in terms of primary variables i.e. pressure, temperature and water saturation. Pressure analysis is important from the point of view of mechanical stability of the engineered barriers and the host formation. Should the pressure approach or exceed the lithostatic pressure of the host formation, cracks could form and further analysis and development of a more complex model would be required. The temperature is also an important factor which influences material properties and overall behaviour of the system e.g. dilatation. We are interested in saturation because of its relation to corrosion effects. For now, gas generation is based on the assumption that the corrosion rate is constant regardless the saturation. Another variant might be to change the corrosion rate might be dependent on the partial pressure of H₂. These aspects, however, exceed the scope of the present report.

5.2.6.1 Pressure evolution

As a result from TOUGH2 calculation we get only one pressure record, which is in line with the switching strategy between one-phase and two-phase regime. When the porous medium is fully saturated the recorded pressure is pressure of water P_{w} . If both phases exist the recorded





pressure is the gas pressure, which is always higher than the water pressure, the difference being the capillary pressure. Therefore, if we are interested in knowing the water pressure in two-phase conditions we have to use the following relation

$$P_c = P_w - P_g . aga{16}$$

The capillary pressure P_c could be calculated following equation (8). The equivalent water saturation Se_w in equation (8) can be calculated from the gas saturation S_g which is given as a TOUGH2 output

$$Se_{w} = \frac{1 - S_{g} - S_{lr}}{1 - S_{lr}},$$
(17)

with residual saturation S_{lr} .

Cases with H=Const (corrosion rate 1 µm/year)

The basic model assumes a constant gas source based on the corrosion rate 1 μ m/y. Two cases are compared, namely one with the concrete buffer permeability of k=4.6×10⁻²¹ m² denoted by low permeability) in Figure 21 and the second with two orders higher permeability. *i.e.* k=4.6×10⁻¹⁹ m² (denoted by high permeability) in Figure 22. Initially the pressure is at the state defined by the simplified initial conditions *i.e.* all engineered barriers are at atmospheric pressure and the pressure in Boom clay is 2.2 MPa. The primary variables are recorded at the locations as described in Figure 15. From the initial state, pressure starts to rise in 'observation' elements 1, 2, 3 and 4 due to the gas source and pressure equilibration between the Boom Clay pressure and the EBS pore pressure. One can observe that in the first time period (roughly until t<10⁻³ y) equilibration has larger contribution to pressure rise than the source itself. Therefore pressure will rise earlier in the elements closer to the Boom Clay. In reality, however, the process might be somewhat slower, because the pressure during the construction of galleries is distributed continuously and not as a step function like the initial conditions defined in the present work. Please note that the gas source applied is corresponding to a corrosion rate of 1 μ m/y which is ten times larger than the currently considered upper limit of 0.1 μ m/y.

In the low permeability case (Figure 21) the pressure rises close to the overpack (element 1). The hydrostatic pressure of the host formation is reached after 4 years. Thereafter, the peak of 3.2 MPa is reached at approximately 90 years. At the same time, the capillary pressure reaches a value of 0.72 MPa. At the time of peak pressure breakthrough is observed and the pressure is high enough to push water out of the system. This reflects in the fast pressure rise in the concrete buffer (element 2). The peak pressure at that location is roughly 3 MPa. An increase of the capillary pressure is later only observed in the cementitious backfill (element 3) after almost 2000 y. The gallery lining and Boom Clay are not affected by the pressure rise in the source.





In the case of a high permeability buffer, the results are slightly different (Figure 22). Due to the lower resistance to flow, the pressure rises slowly up to the end of simulation time (3000 y) with no peak value. Naturally the pressure is lower (2.76 MPa at the end), since water can be pushed out more easily. Nevertheless, the formation of capillary pressure in the buffer (element 2) occurs at the same time as in the case with a low permeability concrete buffer. The pressure in element 2 in a high permeability case (2.75 MPa) is only little lower than in element 1 (0.01 MPa), as opposed to the one observed at the low permeability case, where the difference is 0.2 MPa.







Figure 21: Pressure history for the constant hydrogen production source and **low buffer** concrete permeability.







Figure 22: Pressure history for the constant hydrogen production source and **high buffer concrete permeability.**





Case with H=f(t)(step function) and Q=0

General observations for the pressure history with a transient hydrogen production (Figure 23) are similar as in the previous chapter for the pressure history with constant source (Figure 22). Capillary pressure close to the overpack (element 1) forms and the peak time occurs at approximately the same time as in the case with constant source. However, after 20 years the source in the case of H=f(t) decreases and thus the peak pressure is also lower (2.65 MPa). Due to the slower hydrogen production after 100 years, the increase of pressure in the buffer (element 2) is not significantly increased (decrease of pressure after peak value is not due to breakthrough, but due to decrease of the gas source term).







Figure 23: Pressure history for transient case without thermal source (low permeability buffer).





<u>Case with H=f(t) (smooth) and Q=f(t)</u>

In addition to the previous case, a time dependent heat source term Q=f(t) is assumed here. Due to this heat source term the temperature in the waste region starts to rise and diffuses to surrounding materials. The gas source term is the same as in previous section: a transient hydrogen production H=f(t).

After 2.5 years, desaturation begins inside the second phase concrete, where the source is located. Gas pressure reaches the peak at 20 years. Thereafter, gas pressure diminishes due to decrease of the corrosion rate. The end of the presence of a gas phase is at approximately 300 years. A capillary pressure is not observed at any observation element further from the source. This indicates the very localized effect of gas formation.

The maximal pressure is observed in the second phase concrete with value 2.85 MPa.







Figure 24: Pressure history for the transient case with a thermal source (high permeability buffer)





It should be stressed that the hydrostatic pressure indicated in Figure 24 is just the initial water pressure. When temperature rises, water pressure rises as well due to thermal expansion of water. This can be clearly seen in Figure 24, when water pressure exceeds initial hydrostatic pressure, but a gas phase still does not form. The dependence of water pressure on temperature is presented in Figure 25 for element 1. One can observe the same tendency of higher water pressure with higher temperature.



Figure 25: Relationship between water pressure and temperature.

An interesting comparison can be done by using a low permeability buffer concrete in combination with a heat source. In this case it is expected that water can not be evacuated fast enough. Therefore water expansion will result in high water pressure as the temperature increases as indicated in

Figure 26. It can be seen that very high water pressures exceeding 9 MPa are calculated in the vicinity of the thermal source using this model. This pressure would damage engineered barriers after which our present model is no longer valid (*i.e.* cracks formed in the concrete). The pressure in the backfill (element 3) and further on is similar to the high permeability case in Figure 24. Actually, assuming our conditions, the effect of the heat source on the resulting pressure is much larger than the hydrogen production itself. In this case, the heat source is applied in a saturated near field environment. However, in case of a low-permeability buffer, it takes a longer time for the EBS to become saturated (from section 4.1.1: 20-80 years depending on initial saturation level). Upon saturation, the heat source term will already be considerably lower and the water pressure increase due to water expansion is less significant.







Figure 26: Pressure history for transient H_2 production case with a thermal source and low buffer permeability.





Pressure profiles for the high permeability case H=f(t) and Q=f(t) at different times are shown in Figure 27. The gas is formed in the second phase concrete causing a pressure increase which is relatively quickly transferred throughout all engineered barriers. This is well observed from the flat profiles in the EBS region. This indicates that, under the model assumptions, the water can be pushed out of the engineered barriers relatively easily. This is also the reason that no gas phase appears in the model results further away from the source.



Figure 27: Pressure profiles at different times.

5.2.6.2 Saturation time history

Saturation history corresponds to the increase of capillary pressure shown in the pressure time history results. Two cases are elaborated:

- Basic case with constant hydrogen production: comparison in terms of low and high buffer concrete permeability,
- Case with H=f(t) source: comparison in terms of presence of heat source.





In the first case desaturation occurs in three observation elements, while in the second case desaturation takes place only at the location where gas is formed, hence only at the first element.



Figure 28: Saturation time history for basic case with low and high permeability of the concrete buffer.

Figure 28 shows gas saturation of nearly 30% in the case of low permeability buffer concrete and a much lower gas saturation of 23% in the case of high permeability concrete. Three periods of gas saturation can be recognised. The first period is when the whole system is water saturated. No gas fraction exists. Close to the overpack (element 1) gas phase formation (Sg>0) is observed at 4 years. In the second period water desaturation increases due to the gas





source. In this phase the influence of the concrete buffer permeability is not significant (both curves are aligned). After 70 years the third phase begins, and the influence of buffer permeability becomes apparent. Similar observations can be made for elements 2 and 3.



Figure 29: Saturation time history for the case H=f(t) with and without thermal source.

In the second case with H=f(t) the gas phase stays very localized in both cases regardless the thermal source being present or not (Figure 29). There is a small difference in the magnitude of gas saturation between the two cases. In the case of a thermal source, the gas saturation is approximately 2% higher than in the case without thermal source. This is probably the consequence of a higher gas partial pressure due to the higher temperature, similar to the higher water pressure described before. In other words, due to thermal expansion of the gas, desaturation is higher. However, this is only a 'mild' effect as, unlike water, gas is compressible.

Desaturation close to the overpack (element 1) starts at approximately 3 years when gas partial pressure reaches the level of hydrostatic pressure. In both cases the time of gas phase formation is nearly the same although water pressure is somewhat higher in the case with thermal source. Therefore the temperature increase reflects more in overall pressure increase, but not in (de)saturation conditions to a large extent.





Comparison of pressures using different source formulations

As explained earlier, one reason for these calculations is also to compare results obtained by a different source term formulation. In previous section, the results were presented in terms of thermal source presence. Here, also the comparison is made between a constant, simplified step gas source and a more realistic gas source.

From Figure 30 it can be observed, that the differences between results based on different gas source terms are not large. Results in Figure 30 are shown for observation element 1. The difference between the simplified step source and Arrhenius curve gas source does not play a crucial role. The simplified source is assumed to be high up to 300 y. For this reason, the curve of peak pressure is prolonged in comparison to Arrhenius type source, where the hydrogen production diminishes continuously with time. From 100 y on, the curve is prolonged only for the case with the constant source. On the other hand, desaturation starts earlier with the Arrhenius type source, because gas formation is much more intensive in the first period (until approximately 20 y).

Figure 30 also reveals that the source does not have a big influence on the pressure until the equilibration with surrounding pressure is finished (assuming initially saturated conditions). Hence the pressure increase rate due to the gas source is much lower than the pressure increase rate due to equilibration in our case. The results would be different with a larger gas source or the presence of materials with lower permeability close to the source as shown in Figure 26. Similar observations are seen in the other elements.



Figure 30: Comparison between pressure history for different source terms.





5.2.6.3 Temperature time history

The heat source causes the temperature in the waste region to increase. Heat is transported mainly by conduction through the surrounding material. Temperature histories in selected observation nodes (Figure 31) correspond roughly with the temperature histories calculated in (Weetjens *et al.*, 2006). There is a minor difference due to different geometry of the model since the supercontainer dimensions were recently updated. The conceptual model is updated by introducing advective heat transfer in addition to conductive heat transfer due to water being expelled from the system. Furthermore, the temperature here is coupled to a larger number of phenomena, namely pressure increase and other non-isothermal processes involved. The oscillatory behaviour is not due to the numerical instabilities, but due to the source interpolation within TOUGH2.



Figure 31: Temperature history.

Naturally, the highest temperature is inside the thermal source (waste region). There, the peak temperature is achieved at 10 years after waste emplacement. The time of the peak temperature in different layers (materials) corresponds to the distance from the source. In the Boom Clay (element 5) the peak occurs approximately at 20 y after waste emplacement.

Temperature profiles over the first 5 m from the axis are presented in Figure 32.







Figure 32: Temperature profiles.

5.2.6.4 Comparison of dissolved hydrogen concentration profiles

Hydrogen dissolves in water up to the solubility dictated by Henry's constant (11). Unfortunately, the temperature dependence of solubility in TOUGH2 is limited to 25°C, hence the difference is negligible between solubility at 15.7°C (at isothermal conditions) and 25°C at the highest temperature. Furthermore, there are other differences that influence the dissolved hydrogen transport. The first, and probably the most important, is diffusion, which increases with higher temperature. Another phenomenon relates to advection caused by larger pressure gradients due to thermal expansion of the pore water and consequently water velocities in the outward radial direction as shown in Figure 30. Due to both phenomena, we expect transport to be faster at higher temperatures, which is proven by the results presented in Figure 33. The dissolved hydrogen concentration is the highest at the source and decreases with the distance from the source. The results are presented for three time outputs, namely at 1, 10 and 50 years.







Figure 33: Dissolved hydrogen profile.

From the comparison between the solid and dashed lines in Figure 33 we can see that the dissolved hydrogen is removed faster with increased temperature involved (dotted lines in comparison to full lines), but the effect of the thermally enhanced diffusion and advection is not particularly large.





5.3 Effect of the H₂ gas solubility formulation in TOUGH2

5.3.1 Problem description

As mentioned previously, the solubility of hydrogen in the pore water is defined only until 25^oC and a constant value is assigned for higher temperatures in the two-phase flow code TOUGH2. The aim of this paragraph is to assess the influence of a more realistic temperature dependent solubility. The implementation of this requires a modification in the source code.

The solubility is normally defined by Henry's law which states "At a constant temperature, the amount of a given gas dissolved in a given type and volume of liquid is directly proportional to the partial pressure of that gas in equilibrium with that liquid".

In case of hydrogen production, the solubility defines the amount of hydrogen stored in water before gas phase starts to form. The solubility is temperature dependent. In the TOUGH2 code, this is implemented in a simplified manner:

$$H(T) = \begin{cases} 1.379 \cdot 10^{-10} & T \ge 25^{\circ}C \\ 1.697 \cdot 10^{-10} - 1.272 \cdot 10^{-12} \times T[{}^{\circ}C] & T < 25^{\circ}C \end{cases}$$
(18)

Experimental values, obtained from Perry's chemical engineers' handbook (Perry and Green, 1984) are presented in tabulated form in Table V.





temperature	Solubility
[⁰ C]	[mol H ₂ /(mol H ₂ O*Pa)]
0	1.70E-10
5	1.62E-10
10	1.55E-10
15	1.49E-10
20	1.44E-10
25	1.40E-10
30	1.35E-10
35	1.33E-10
40	1.31E-10
45	1.30E-10
50	1.29E-10
60	1.29E-10
70	1.30E-10
80	1.31E-10
90	1.31E-10
100	1.32E-10

Table V: Experimental values from (Perry and Green, 1984)

Comparison between experimental and TOUGH2 values can be seen in Figure 34, where the agreement is good until a temperature of 25^oC, but less accurate for higher temperatures.



Figure 34: Comparison between solubility values in TOUGH2 and experimental values.

In order to implement the experimental points into the TOUGH2 code, the experimental points have to be expressed as a function. The function chosen to fit the experimental points is as follows





 $H(T)=a+b\cdot T+c\cdot T^2+d\cdot \exp(-e\cdot T)$

(19)

with parameters a,b,c,d and e defined as

 $a = -6.88401 \cdot 10^{-10}$ $b = 7.35462 \cdot 10^{-12}$ $c = -2.08518 \cdot 10^{-14}$ $d = 8.59034 \cdot 10^{-10}$ $e = 1.07349 \cdot 10^{-2}$

The goodness of fit between the fitted function and experimental points also shown in Figure 34.

5.3.2 Results

Numerical results will be presented for two examples with a variable source temperature. The first example is with a high buffer permeability (Figure 35) and the second with a low buffer permeability (Figure 36). The results from the simplified linear relation with cut-off (equation (18)), taken from Figures 24 and 26 are plotted with dashed line, while the new results from the fitted function (equation (19)) are represented by solid lines.

In the example with a high permeability buffer material Figure 35, differences are negligible. Small differences are observed in the Element 1 (overpack), while no difference is noticeable farther from the source.

Larger differences are observed if a lower permeability buffer is simulated (Figure 36). The peak pressure, which is in this case due to thermal expansion of the pore water is slightly higher and occurs a little later. The formation of a gas phase (after about 3 years) occurs earlier compared to the original TOUGH2 implementation. This can be explained by the lower solubility of gasses with higher temperatures in the fitted equation (19). The difference between the original (simplified) and the improved solubility formulation diminishes with the distance from the source.







Figure 35: Pressure history for transient H_2 production case with a thermal source and high buffer permeability.







Figure 36: Pressure history for transient H_2 production case with a thermal source and low buffer permeability.





Because the effect of the improved solubility formulation is best visible in element 1 i.e. overpack, the results are magnified and presented in Figure 37.



Figure 37: Pressure history at the overpack (element 1) for transient H_2 production case with a heat source - low buffer permeability (top) and high buffer permeability (bottom).





5.3.3 Discussion

Although the original equation that expresses the temperature dependency of the hydrogen solubility in water implemented in TOUGH2 is very simplified, this simplification does not considerably influence the results. Differences, albeit small, are more pronounced for the low permeability buffer case, where the peak **water** pressure (due to thermal expansion of water) close to the overpack reaches 9.09 MPa for the simplified formulation and 9.16 MPa in the improved solubility formulation. Concerning the **gas** pressures reached in the near field: the peak pressure reached 2.95 MPa using the simplified function and 2.96 MPa using the improved solubility formulation. For the high permeability case, the difference is also limited to 0.01 MPa: the gas peak pressures reach 2.85 MPa for the simplified and 2.86 MPa for the improved solubility formulation.





5.4 Benchmarking of TOUGH2 by comparison with CODE_BRIGHT

5.4.1 Introduction

In a safety assessment, the importance of building confidence in the applied models, codes and input data is paramount, whether it concerns detailed process modelling or performance assessment calculations. Indeed for these type of calculations, the involved timescales are extremely long. To that aim, quality procedures are introduced, often demanded by regulatory bodies, which insist the application of model validation (whenever possible), model qualification and code verification.

- Qualification is the process of ensuring that a conceptual model and its associated data takes account of all relevant features and processes and that the various simplifying assumptions can be justified in terms of the intended use.
- Verification of a computer model, that is, verification of the representation of a conceptual model in terms of parameter values, mathematical equations, boundary conditions and the computer codes used to solve them, is the process of checking that the simplifications and abstractions made in order to solve the governing equations of the model have acceptably low influence on the calculated results. Computer codes must further be shown to be accurate and error free. The latter is typically done by performing benchmarking analyses in which numerical codes are compared with others or, in case of simplified systems, with analytical solutions.
- *Validation* in the classical scientific sense of comparing the predictions of computer simulations with observations on a real system is in most cases not possible over the temporal and spatial scales that are relevant for safety assessments, but it may be possible for process models.

The theoretical relation between conceptual model, computer model, qualification, verification and validation is shown schematically in Figure 38.







Figure 38: Schematic illustration of the concepts of qualification, verification and validation in theory, taken from ONDRAF/NIRAS (to be published) after Schelsinger (1979)

In this paragraph, benchmark calculations comparing TOUGH2 and CODE_BRIGHT are presented and differences in the results obtained by both modeling tools are analyzed. This assures proper application of the software and increased understanding of its kernel features.

5.4.2 CODE_BRIGHT: features and mathematical formulations

Both TOUGH2 and CODE_BRIGHT are extensively used in the context of evaluating nuclear waste disposal facilities. TOUGH2 was developed primarily at Lawrence Berkeley National Laboratory (LBNL), and its features and capabilities were already discussed in 5.2.2. CODE_BRIGHT is a finite element code developed by the Technical University of Cataluña (UPC), Spain, for calculating displacements, liquid pressure, gas pressure, temperature and salt content for boundary value problems in saturated or unsaturated soil (Olivella et al., 1996).

All the important constitutive laws used in solving the mass and energy balance equations within CODE_BRIGHT are essentially the same as for TOUGH2, with a few exceptions:

- Dissolution of hydrogen in liquid phase: Henry's constant varies linearly with temperature in TOUGH2, while it is constant in CODE_BRIGHT.
- Diffusion of dissolved gas in liquid phase : the diffusion coefficient for dissolved gas in the liquid phase, D^g_l, in both codes are set to be constant in the benchmark cases. In CODE_BRIGHT, the tortuosity τ is also set to be constant, while in TOUGH2, it is allowed to vary according to the relative permeability model
- In CODE_BRIGHT, the temperature dependency of the viscosity is expressed as:





$$\mu(gas/liquid) = A \exp\left(\frac{B}{273.15 + T}\right)$$
(20)

• The water density in CODE_BRIGHT is temperature and pressure dependent and expressed as:

$$\rho_{l} = \rho_{l0} \exp(\beta (P_{l} - P_{l0}) + \alpha (T - T_{0}))$$
(21)

Where ρ_{l_0} is the reference liquid density at reference pressure P_{I0} (0.1MPa), T₀ is the reference temperature (0°C); β is the compressibility coefficient; and α is the volumetric thermal expansion coefficient for the liquid phase.

5.4.3 Benchmark case 1: HG MODELING in (1D)

5.4.3.1 Benchmark case 1 definition

In benchmark 1, a simple HG coupled model is carried out for a 1D problem to simulate the desaturation process due to gas pressure build up (see Figure 39). The whole domain is separated into two regions. Region I (x=0-0.316m) represents the engineered barrier system (EBS) and region II represents the host formation (Boom Clay). In order to simplify the problem, the properties of the EBS materials are set to be the same as those of Boom Clay.

There is a fixed hydrogen injection rate at the left boundary of the system to simulate gas production as result of anaerobic corrosion of the carbon steel overpack (the corrosion rate is assumed to be 1 μ m/y). Fixed gas and liquid pressures are prescribed at the right boundary to represent undisturbed far field conditions. The whole domain is initially saturated and at a gas pressure of 0.1 MPa. Due to the excavation and backfilling of the tunnel, initial liquid pressure is set to 0.1 MPa in region I to reflect near field depressurization during construction, while hydrostatic pressure conditions of 2.3 MPa are assumed (220 m in depth) in region II. The temperature is assumed to be 25°C in the whole domain. Five observation points are selected within the domain and their positions are listed in Figure 39.

In CODE_BRIGHT, gas and liquid pressures are outputted at each node, while liquid saturation is outputted for each element which is averaged from the element nodal values. In TOUGH2, all the results are outputted at the centre of the volume element. In this benchmark, observation points are chosen to be node positions in CODE_BRIGHT, while the centre of volume elements were chosen in TOUGH2.







Figure 39: Benchmark case 1: model geometry and position of observation points: 1) x=0.007 m 2) x=0.36 m 3) x=3.0 m 4) x=5.7 m 5)x=9.54 m

5.4.3.2 Results of benchmark case 1

Time evolutions of gas/liquid pressure and gas saturation up to 100,000 years at five observation points are compared for the two codes in Figure 40. Because TOUGH2 does not output gas pressure explicitly for saturated materials, comparisons between gas pressures are carried out only under unsaturated conditions. As illustrated in this figure, the results from the two codes are in good agreement before 3,000 years when the gas pressure front reaches the right boundary (marked by B). Both codes describe the process of resaturation and desaturation in a very similar manner.

When two-phase flow starts, gas pressure and gas saturation obtained by CODE_BRIGHT start to exhibit oscillations (after the transitional point A). This may due to the steep decrease in relative liquid permeability when desaturation starts. As soon as the gas pressure front reaches the right boundary, results from CODE_BRIGHT show an abrupt increase of gas pressure due to the sharp transition between an unsaturated element and the saturated boundary condition prescribed at the right boundary. The application of volume elements and the finite difference method makes results from TOUGH2 much more stable in such a situation.





Figure 40: Comparison of time evolutions of gas, liquid pressure and gas saturation at five observation points between two numerical tools in benchmark case 1.

It is not surprising that gas saturation, and hence gas pressure at point 5 is quite different for two codes. In CODE_BRIGHT, the saturation is averaged from nodal values within the element of 8.81m < x < 9.542m, while in TOUGH2, the output value is the central value of the element of 9.084m < x < 10m. Obviously, the latter has a higher liquid degree than the former, which corresponds to a lower gas pressure/saturation.

5.4.4 Benchmark case 2: HG MODELING (2D)

5.4.4.1 Benchmark case 2 definition

Benchmark 2 is defined according to the geometry of the calculations reported in paragraph 5.2. This case involves 2D axisymmetric modelling of two-phase (liquid and gas) flow with constant hydrogen production in isothermal conditions. A high buffer permeability ($k=4.6 \times 10^{-19} m^2$) was assumed.





The gas influx generated by corrosion of the overpack is applied uniformly on the interface of waste and 2nd phase concrete in CODE_BRIGHT, and distributed uniformly among all the volume elements of the 2nd phase concrete in TOUGH2.

The whole domain is meshed with a total of 2997 linear quadrilateral elements and 3116 nodes in CODE_BRIGHT, and discretized with 3160 volume elements in TOUGH2. Five observation points are selected in the whole domain and their positions are listed in the table below.

Point No.	material	X(m)	Y(m)
1	2 nd phase concrete	0.675	0.291
2	Concrete buffer	0.675	0.491
3	Cementitious backfill	0.675	1.2462
4	Gallery liner	0.675	1.632
5	Boom Clay	0.675	2.4752

Table VI: Position of observation points in benchmark case 2

5.4.4.2 Results of benchmark case 2

Time evolutions of gas/liquid pressure and gas saturation up to 20,000 years at five observation points are compared for the two codes. As illustrated in Figure 41, both codes give quite similar time evolutions of gas and liquid pressures. Two-phase flow is limited in the region within concrete buffer by the end of 10,000 years. When two-phase flow starts, the same phenomenon as in benchmark 1 appears, namely that gas pressure and gas saturation obtained by CODE_BRIGHT show oscillations at the transitional point, while they are quite smooth in the results obtained by TOUGH2. This is especially apparent in the gas saturation evolution plot.








Figure 41: Comparison of time evolutions of gas, liquid pressure and gas saturation at five observation points between two numerical tools in benchmark case 2





5.4.5 Benchmark case 3: THG MODELING (2D)

5.4.5.1 Benchmark case 3: definition

Benchmark 3 is defined using the same conceptual model as in benchmark 2, but extended by coupling with heat transport. In this benchmark case, hydrogen and heat influx are simplified as piecewise linear functions (Figure 42) and applied uniformly along the interface of waste and the 2nd phase concrete in CODE_BRIGHT. In TOUGH2, the thermal source is distributed uniformly in the waste matrix which is given an extremely higher thermal conductivity of 40 Wm⁻¹K⁻¹ to assume a fast homogeneous heat spreading.



Figure 42: Hydrogen and heat influx in benchmark case 3

5.4.5.2 Results of benchmark case 3

Time evolutions of gas/liquid pressure and temperature up to 10 000 years at five observations points are compared for the two codes. As shown in Figure 43, both codes produce comparable results. Increase of liquid pressure in the first five years is due to the thermal expansion of water. Its timescale is consistent with time evolution of temperature. The differences between liquid pressures mainly come from a different definition of the dependency of liquid density on temperature. From the liquid pressure curves of CODE_BRIGHT, it can be clearly seen that there are a lot of oscillations before the appearance of the hump which may be due to calculation instability. There is a good correspondence between temperature results, but CODE_BRIGHT gives a much smoother curve compared to TOUGH2.



Figure 43: Comparison of time evolutions of gas, liquid pressure and gas saturation at five observation points between two numerical tools in benchmark case 3





5.4.6 Conclusions of the benchmarking study

Comparisons between numerical results in terms of the primary variables pressure, saturation and temperature, demonstrate that the tested numerical tools TOUGH2 and CODE_BRIGHT produce similar results in all three benchmarks. The minor differences between results can be attributed to different discretizing methods and numerical techniques, and to several different constitutive laws.

Results from CODE_BRIGHT are very sensitive to the choice of convergence parameters. Convergence problems have been encountered in some occasions. The results reflect sharp oscillations at some critical points, while results from TOUGH2 are more stable.

Although both numerical tools seem to be "fit-for-purpose", meaning that they can be used to simulate the type of coupled multiphase flow problems described here, the results show that TOUGH2 is more stable and efficient in modelling two-phase flow problems. The advantage of CODE_BRIGHT is that it has provisions for solving mechanically coupled problems, and is easier to be implemented with self-defined constitutive laws.





6. Conclusions

In this study, several aspects in the assessment of the impact of gas generation were examined by means of a case study focussing on deep disposal of supercontainers containing vitrified HLW in Boom clay. In general, anaerobic corrosion of steel EBS components is found to be the main source of gas generation (in this case hydrogen) in the near field of a radwaste repository.

The evolution of the EBS in terms of its water saturation, pressure and temperature is quite complex and it is of importance to know the prevailing conditions at the on-set of gas generation. As a first step, resaturation calculations were performed, to find out whether or not near field temperatures are still significantly increased at the start of anaerobic gas production. Note that full saturation is only roughly indicative for the transition of oxidising to reducing conditions, which could be considered as the start of the anaerobic corrosion reaction. Furthermore, since there is still substantial uncertainty on the hydraulic properties of the EBS materials and their initial saturation degree, these resaturation calculations considered a broad range of possibilities. In the most likely case, combining a hydraulic conductivity comparable to the one of Boom clay with a high initial saturation (80%), the whole gallery would be saturated with pore water within a couple of years. This means that temperatures are at their maximum when corrosion gas production starts.

The corrosion gas source term was implemented using different assumptions; namely two constant corrosion rates of 0.1 μ m/year and 1 μ m/year and a transient case where the influence of temperature on the corrosion process was assessed through application of the Arrhenius law. Next, it was assessed whether the generated hydrogen could be evacuated by diffusion as dissolved species, by comparison of cumulative gas production rates and the maximum rate at which dissolved hydrogen can diffuse away from the source. In these simplified transport simulations two values of diffusion coefficients were tested: a D_p of 5x10⁻¹⁰ m² s⁻¹ and a D_p of 5x10⁻¹¹ m² s⁻¹. The results showed that for some calculation cases, diffusion alone is not enough to dissipate the gas produced within the EBS. However, there was no indication that a free gas phase could extend into the Boom Clay, *i.e* free gas, if any, should only be found within the EBS.

A detailed multiphase flow analysis comprised the next step in this study. The aim of these fully coupled two-phase flow calculations was to assess the evolution of pressure, saturation and temperature in the repository and its environment. However, the emphasis was on gaining insight in the possible behaviour of the system, and in particular testing the robustness of the system by using a variety of source term formulations and bounding values of the buffer permeability. In addition, the impact of heat generation was examined. Results of these calculations show that the influence of temperature on the gas *production* process could be substantial (Arrhenius law) but the overall influence of temperature on the gas *transport* process is small. The implementation of a heat source results in a slightly increased total pressure, mainly due to thermal expansion of both water and gas phase. The degree of gas saturation is not significantly higher compared to the isothermal case. In case of a high-





permeability buffer, thermal expansion of the pore water causes slightly higher water pressures, whereas the pressure increase was considerably higher in the low-permeability case. However, this could be a consequence of the sequential modelling of the resaturation process and the gas generation and transport in a heated saturated environment: in reality, with a low-permeability buffer, the resaturation process will take longer (estimated here at 20 to 80 years depending on initial saturation degree), and the temperatures will already be much lower. Besides, the behaviour of the solid phase (concrete, clay) in this model is greatly simplified through the use of a storage coefficient approach. In other words, only one-way fluid to solid coupling is considered, under an implicit constant total stress assumption.

In all considered cases, the presence of a gas phase remains very local, *i.e.* within the EBS, and the Boom clay is not subject to a pressure increase (which was already indicated by the simplified diffusive mass-balance calculations). In the most realistic case, the maximum gas pressure reaches 2.85 MPa in the concrete filler after 20 years of gas production. The corresponding gas saturation is 20%. The tensile strength of concrete of reasonable quality should be larger than the expected gas pressure.

Briefly summarised, the conclusions of this case study could be formulated as follows:

- Under the current assumptions, disposal of vitrified HLW in Boom Clay using a supercontainer as waste package is not likely to pose a hydrogen gas problem due to anaerobic corrosion (which confirms the results obtained in the framework of EC project NF-PRO).
- A free gas phase may develop inside the concrete buffer, but the tensile strength of concrete should normally be larger that the expected gas pressure. Hence, gas-induced fracturing should be unlikely.
- The mechanical and hydraulic integrity of the Boom clay should thus not be threatened.

Moreover, some conservative assumptions to the conceptual model are worth mentioning:

- the corrosion rate was neither dependent on the degree of saturation nor on H₂ pressure
- consumption of H₂O by the anaerobic corrosion reaction was not taken into account, although this is not believed to have a large influence on the results.

It is to be noted, however, that other waste types, particularly intermediate-level wastes, might be more critical with respect to gas production and especially gas production rate than the vitrified high-level waste considered in these exploratory calculations.





These calculations have further shown that it is feasible to improve certain formulations in the constitutive laws of TOUGH2. An example was discussed in which results of a more accurate temperature dependency of the hydrogen solubility were compared to the standard simplified formulation. However, the nature of the curve is such that the influence on the final timing and amplitude of pressure build up is negligible.

Finally, two numerical tools, TOUGH2 and CODE_BRIGHT, are mutually verified through three benchmark cases based on the considered case study: 1) HG coupled model for a 1D problem; 2) HG coupled model for a 2D axisymmetrical problem; 3) THG coupled model for a 2D axisymmetrical problem; 3) THG coupled model for a 2D axisymmetrical problem. Comparisons between numerical results demonstrate that these two numerical tools produce similar results in all three benchmarks. The minor differences between results obtained from the two numerical tools are in part due to the different discretizing method and numerical techniques, and in part due to several different constitutive laws. CODE_BRIGHT seems to be quite sensitive to convergence parameters. During the calculation, convergence problems have been encountered occasionally. The results reflect sharp oscillations at some critical points, while results from TOUGH2 seem to be more stable. However, the advantage of CODE_BRIGHT is that it has provisions for solving mechanically coupled problems, and is easier to be implemented with self-defined constitutive laws.

As an overall conclusion, the achievements within this workpackage have shown that the tools applied are adequate (*selected* processes of concern in gas generation and dissipation – dissolution, diffusion and two-phase flow, if necessary coupled to heat transport – can be implemented), accurate (numerical results of both codes in good agreement) and versatile. However, the challenging task of proving that the conceptual model is *comprehensive* still remains. Substantial efforts are still necessary in the domains of model qualification and, if possible, validation. In this respect, much is expected from the recently started EU-FP7 FORGE project ("Fate of Repository Gases"), in which the various models for the gas generation and migration will be benchmarked to experiments and in which process level knowledge will be further developed.





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