

FORGE

Fate Of Repository Gases

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Description of numerical models used by GRS for the analysis of the HG-C experiment

FORGE Report D5.5

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Fate of repository gases (FORGE)

The multiple barrier concept is the cornerstone of all proposed schemes for underground disposal of radioactive wastes. The concept invokes a series of barriers, both engineered and natural, between the waste and the surface. Achieving this concept is the primary objective of all disposal programmes, from site appraisal and characterisation to repository design and construction. However, the performance of the repository as a whole (waste, buffer, engineering disturbed zone, host rock), and in particular its gas transport properties, are still poorly understood. Issues still to be adequately examined that relate to understanding basic processes include: dilational versus visco-capillary flow mechanisms; long-term integrity of seals, in particular gas flow along contacts; role of the EDZ as a conduit for preferential flow; laboratory to field up-scaling. Understanding gas generation and migration is thus vital in the quantitative assessment of repositories and is the focus of the research in this integrated, multi-disciplinary project. The FORGE project is a pan-European project with links to international radioactive waste management organisations, regulators and academia, specifically designed to tackle the key research issues associated with the generation and movement of repository gasses. Of particular importance are the long-term performance of bentonite buffers, plastic clays, indurated mudrocks and crystalline formations. Further experimental data are required to reduce uncertainty relating to the quantitative treatment of gas in performance assessment. FORGE will address these issues through a series of laboratory and field-scale experiments, including the development of new methods for up-scaling allowing the optimisation of concepts through detailed scenario analysis. The FORGE partners are committed to training and CPD through a broad portfolio of training opportunities and initiatives which form a significant part of the project.

Further details on the FORGE project and its outcomes can be accessed at www.FORGEproject.org.

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1 Introduction

In FORGE work package 5, task 5.3.4, GRS contributes to the interpretation of the HG-C experiment by numerical simulation of hydro-mechanically coupled 2-phase-flow processes. The modelling work will be based on the 2-phase flow code TOUGH2 which has been modified by GRS to simulate THM-interactions and micro-crack dilation, which are both relevant to the transport of gas in indurated clay. TOUGH2 is a well validated code which has found wide acceptance in the field of PA partly due to its high accuracy in modelling thermodynamic processes. According to the dominant processes observed in the HG-C experiment, either a coupled TOUGH2/FLAC3D code for the simulation of three dimensional hydro-mechanical interactions will be applied, or a TOUGH2-modification for simulation of pressure-induced dilation of secondary pathways in porous media (TOUGH2-PD). Table 1-1 gives an overview over main features and differences of the models.

The investigations planned for FORGE WP 5 aim at testing and improving the applied code against the experimental results and at enhancing the understanding of gas transport processes in undisturbed indurated clay and the capabilities of the considered physical and numerical models.

In this milestone report both models, TOUGH2-PD and TOUGH2/FLAC3D will be presented.

Table 1-1: Comparison of TOUGH2-PD and TOUGH2/FLAC3D

	TOUGH2-PD	TOUGH2/FLAC3D
Matrix deformation captured by ...	<ul style="list-style-type: none"> ... dilation threshold pressures ... porosity changes ... gas permeability changes ... assumptions on the local mechanical behaviour of the rock (no stress field calculation) 	<ul style="list-style-type: none"> ... coupled HM-Simulation ... porosity change ... change of intrinsic permeability ... capillary pressure change
Does dilation/compaction change phase composition?	Yes	No
Distribution of phases between dilated and non-dilated pores	Unequal (dilated pores have a higher gas phase fraction)	Equal
Can the gas phase flow without displacing water?	Yes	No
Currently not applicable or further code development necessary if ...	<ul style="list-style-type: none"> ... homogenisation of dilation effects is not possible on the element scale ... intermittent gas flow on the element scale is present ... memory effects are important in the considered cases 	<ul style="list-style-type: none"> ... homogenisation of dilation effects is not possible on the element scale ... intermittent gas flow on the element scale is present ... gas enters into saturated rock with immobile liquid phase <p><i>(Mechanical restrictions dependent on chosen constitutive laws)</i></p>

2 Description of TOUGH2/FLAC3D

In order to study coupled thermo-hydrologic-mechanical (THM) processes in repository systems the thermo-hydrological code TOUGH2 (Pruess 1990) was coupled to the thermo-mechanical code FLAC3D (Itasca 2002). The TOUGH2 version used is a modification of GRS originally developed for the investigation of gas, heat and nuclide transport in a repository in rock salt by introducing a porosity and permeability of crushed salt which is dependent on pressure, temperature and rock convergence (Javeri 2006a). Mechanical effects are not included explicitly. The code FLAC3D was developed by Itasca for rock and soil mechanical applications and is capable of THM-simulations for single phase flow but not for two phase flow. On the basis of the work of Javeri (2006b) in which TOUGH2 and FLAC3D were coupled linearly for the simulation of gas and nuclide transport in three dimensional isothermal systems, the two codes are now coupled sequentially according to Rutqvist et al. (2002) in order to analyse three dimensional non-isothermal systems.

The following description of the coupling of TOUGH2 and FLAC3D uses the following symbols:

k:	permeability [m^2]
p_{liq} :	liquid pressure [Pa]
p_{gas} :	gas pressure [Pa]
p_{eff} :	effective pressure [Pa]
p_{cap} :	capillary pressure [Pa]
$p_{\text{cap},0}$:	capillary pressure at time t_0 [Pa]
S:	liquid saturation
t:	time [s]
T:	temperature [K]
V:	volume [m^3]
β :	rock compressibility [$1/\text{Pa}$]
ϵ :	strain
ϕ :	porosity
σ :	total stress (tensile: > 0 , compressive: < 0) [Pa]
σ_j :	principal total stress (FLAC3D: $\sigma_1 \leq \sigma_2 \leq \sigma_3$) [Pa]
σ_{mean} :	mean total stress [Pa]
$\sigma_{\text{mean,eff}}$:	mean effective stress [Pa]
ψ :	Bishop factor

Subscripts:

i:	TOUGH2 element index
k:	index of a FLAC3D connected zone
n:	FLAC3D grid point index

2.1 THERMO-HYDROLOGICAL ANALYSIS WITH TOUGH2

To perform analyses with TOUGH2, which is a recognized tool to study three dimensional coupled fluid and heat transport of two-phase multi-component fluid mixtures in porous media, one of the several equation-of-state (EOS) modules must be linked with the main code of TOUGH2. The number and the properties of the fluid components are determined by the EOS module selected. The mass balance equation of each phase component includes advection and

dispersion in the gas and liquid phases. In the heat balance equation, convection and conduction are considered. The velocities of the gas and liquid phases are determined by a multiphase extension of Darcy's law considering a relative permeability of each phase which depends on the liquid saturation (Pruess, 1990). In the equation of state module EOS7, which is used here, the following three fluid components are considered (Pruess, 1991): groundwater, brine and a soluble gas (air or hydrogen). The complete steam table is included to calculate the liquid properties as a function of pressure, temperature and mass fraction of solute or salt water. The gas phase is treated as a mixture of ideal gases such as air or hydrogen and vapour. The model domain has to be discretised into volume elements. The conservation equations are solved simultaneously with the integral finite difference method. Time is discretised fully implicitly with a first order backward finite difference scheme. Scalar quantities like pressure and temperature are determined at the centre of an element and the vector quantities such as velocities, mass and heat fluxes at the element interfaces.

2.2 MECHANICAL ANALYSIS WITH FLAC3D

FLAC3D is an established three dimensional explicit finite difference code for engineering mechanics which is able to describe the behavior of structures composed of soil, rock or materials that undergo plastic flow when their yield limits are reached. Materials are represented by polyhedral elements within a three dimensional grid that is adjusted by the user to fit the shape of the object to be modeled. Each element behaves according to a prescribed linear or non linear stress/strain law in response to applied forces or boundary restrains. To study coupled processes, fluid pressure and temperature can be prescribed by invoking the fluid and thermal configuration and the mean effective normal stress can be calculated as (Fredlund and Rahardjo, 1993; Rutqvist et al., 2002)

$$\sigma_{\text{mean}} = (1/3)(\sigma_1 + \sigma_2 + \sigma_3), \quad p_{\text{eff}} = p_{\text{gas}} - \psi p_{\text{cap}}, \quad \sigma_{\text{mean,eff}} = \sigma_{\text{mean}} + p_{\text{eff}}$$

The Bishop factor ψ which should be derived from site specific rock properties approaches unity for saturated soil and zero for dry soil. For simplicity ψ is substituted by the liquid saturation ($\psi = S$). To study THM processes, it has to be noted that the total stress, liquid pressure and temperature are computed at the corner grid points of a zone and the effective stress and effective pressure at the center of a zone.

2.3 SEQUENTIAL COUPLING OF TOUGH2 AND FLAC3D

The original version of TOUGH2 with EOS7 was coupled to FLAC3D sequentially. For the coupling, an identical numerical grid with equal numbers of FLAC3D zones and TOUGH2 elements is required (Figure 2-1). TOUGH2 is executed as a 'main program' to perform thermo-hydrological analyses and FLAC3D is executed as a 'subroutine' to conduct quasi static mechanical analyses (Figure 2-2). In the beginning, the initial liquid pressure and temperature distributions are computed by TOUGH2 and transferred to FLAC3D. Since in TOUGH2, the pressure and the temperature are calculated at the centre point of an element and since in FLAC3D the temperature and pressure are allocated to the corner grid points of an element, data have to be interpolated from the mid-element of TOUGH2 to the corner grid points of FLAC3D. The volume averaged pressure and temperature at all grid points of FLAC3D can be derived as:

$$p_{\text{eff},n} = [\sum p_{\text{eff},k} V_k] / \sum V_k, \quad T_n = [\sum T_k V_k] / \sum V_k.$$

This is one possible scheme which has been used as a first approach to compute the pressure and temperature at the grid points. Depending on the problem other schemes should also be considered. Subsequently, the initial stress distribution is calculated and a restart file is created. During the transient analysis, at the end of each time step, TOUGH2 calls FLAC3D and the current fluid and temperature distributions are transferred to FLAC3D. In FLAC3D, the new strain and stress distributions are computed by employing the previous restart file and the current fluid and temperature distributions of TOUGH2. At the end of the FLAC3D run, a new restart file is created and the current stress and strain distributions are transferred back to TOUGH2. Since the effective stresses and strains are determined at the centre of a zone, they can directly be allocated to the corresponding elements of TOUGH2. To continue the two phase flow, heat and nuclide transport analysis with TOUGH2, the hydrological properties, such as porosity, permeability and capillary pressure can be determined basically as a function of stress or strain at each time step. Usually, these coupling functions, which are very crucial for the safety analysis, can be non-linear and should be obtained from site specific data.

Porosity change is implemented in TOUGH2 by means of the rock compressibility β :

$$\Delta\phi := \Delta\varepsilon_{\text{volume}}, \quad \beta = (1/\phi)[\Delta\phi/\Delta p_{\text{eff}}]$$

Thus, it is assumed that the solid fraction of the porous medium is incompressible and the porosity can be directly related to volumetric strain (Fredlund and Rahardjo, 1993).

The capillary pressure is modified according to Leverett function (Rutqvist et al., 2002):

$$p_{\text{cap}} = p_{\text{gas}} - p_{\text{liq}} = p_{\text{cap},0}(S)[(k_0\phi)/(k\phi_0)]^{1/2} \quad \text{with} \quad \phi_0 = \phi(t_0), \quad k_0 = k(t_0).$$

Two options for the permeability change are suggested. In the first option, the permeability is determined as a power function of porosity (Fredlund and Rahardjo, 1993):

$$k/k_0 = (\phi/\phi_0)^4.$$

In the second option, the permeability is described as a function of σ_3 (Popp et al., 2007):

$$\log(k) \sim f[\tanh(\sigma_{3,\text{eff}})] \quad \text{with} \quad \sigma_{3,\text{eff}} = \sigma_3 + p_{\text{eff}}.$$

Thus, the main state variables, pressure, temperature and stress are exchanged between the codes at each time step. Depending on the problem other coupling functions can also be considered. For instance, coupling functions depending upon the mean effective stress were applied in (Javeri, 2006b). The present sequential method in which the two codes are coupled at the end of each time step can be improved and verified by an implicit iterative sequential coupling, in which the two codes are coupled at every Newtonian (physical) iteration within a time step. But, the iterative sequential coupling can increase the computational work quite significantly.

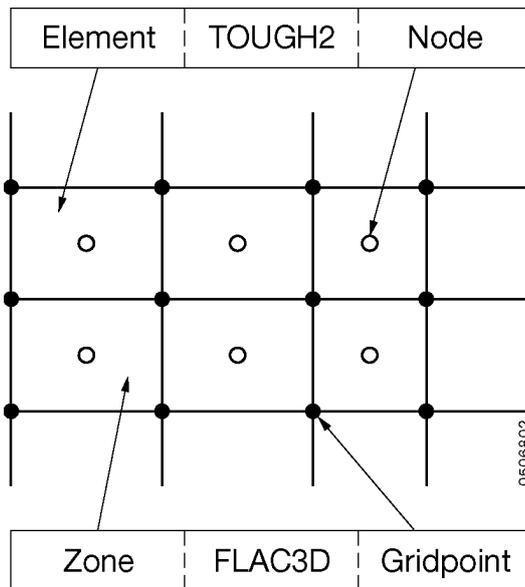


Figure 2-1: Identical mesh for TOUGH2 and FLAC3D

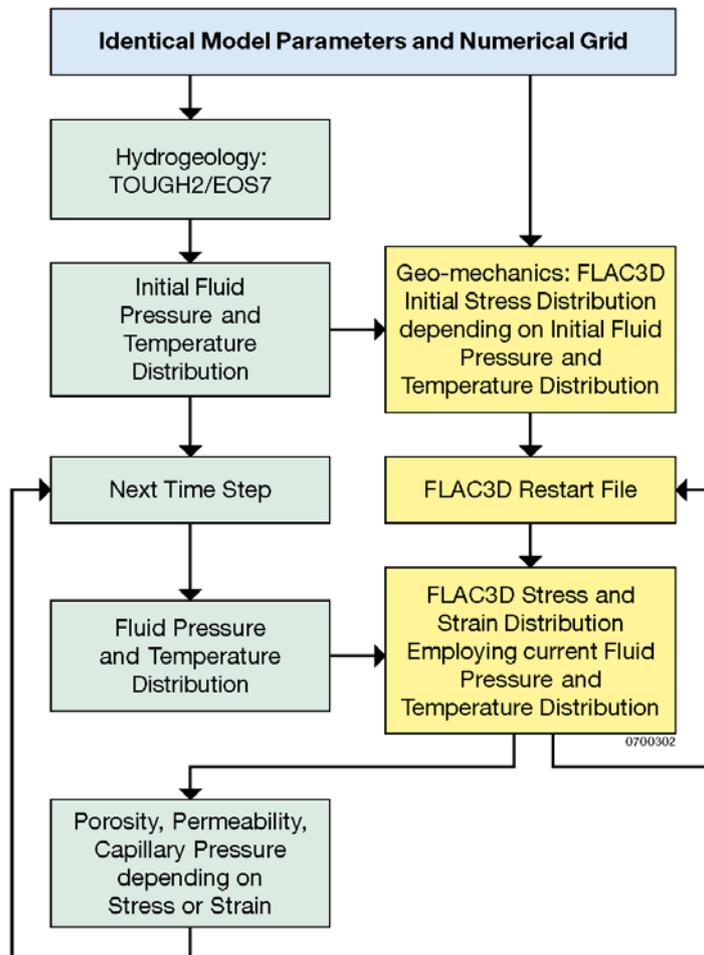


Figure 2-2: Sequential coupling of TOUGH2 and FLAC3D.

3 Description of TOUGH2-PD

The code TOUGH2/EOS7 has been modified to account for the mechanism of pathway dilation in clay stones with very low permeability (Navarro 2009). This code was named TOUGH2-PD (TOUGH2 with pathway dilation). A pressure-dependent porosity has been introduced in order to allow a gas phase to flow without need to displace the liquid phase. Whether desaturation and water displacement takes place or not is strongly influenced by the constitutive two-phase flow relations which are used. The TOUGH2-PD code does not exclude displacement of water by the gas phase but it is able to handle cases where such a displacement is difficult e.g. due to no-flow boundary conditions or very rigorous gas entry thresholds.

Porosity increase was modelled separately from the porosity-change-feature of TOUGH2 which affects the fluxes but not the storage capacity of the rock. Gas permeability was defined by adding a pressure-dependent gas flux within the secondary porosity (created by dilation) to the gas flux in the primary porosity. Liquid permeability is not affected by the dilation process because the dilated pore space is thought to be created and used mainly by the gas phase. This also aims at reproducing the observation of low water displacement in gas migration experiments. The approach of separate gas fluxes in the primary and secondary pore space also allows decoupling of weighting schemes for these two flows and thus a realisation of the intended easy propagation of the dilation front according to the assumption of quick equilibration.

3.1 SUMMARY OF CODE CHANGES

Pressure-dependent porosity. Due to pathway dilation the gas phase is thought to be mobile even if water can not be displaced (e.g. due to rigorous gas entry pressures or due to no-flow boundaries). To achieve this, additional pore space has to be created to accommodate the mobile gas phase or otherwise the gas phase would propagate with infinite pore velocity. For this reason, a pressure-dependent porosity has been implemented into the TOUGH2 code.

In fact, a change of porosity is not of major importance in itself because the porosity changes considered here are very small compared to the total porosity of the rock. However, in media which are almost fully saturated, even a small addition of pore space occupied by a gas phase may significantly change the mobility of gas and the capacity of the rock to store a gas phase.

Porosity increase was modelled separately from the porosity-change-feature of TOUGH2 which affects the fluxes but not the storage capacity of the rock. Due to the strong non-linearity of the relationships describing dilation behaviour, porosity and permeability are changed after completion of a time step in order to avoid convergence problems. To comply with the user-defined relationships between porosity and pressure and to guarantee mass conservation, porosities, pressures and saturations are changed after each time step. This is done by simulating isothermal compression or expansion of the pore space according to the thermodynamic processes implemented in the EOS7 module. It required an inverse formulation of some EOS-algorithms.

Pressure-dependent gas permeability. An additional flux of the gas phase in the dilated pores is added to the flux in the non-deformed matrix. The additional flux follows Darcy's law using a pressure-dependent gas permeability for the dilated pores. This allows to use independent weighting schemes for the permeability of the dilated and of the unaffected pore space. Upstream weighting has been chosen for the permeability of the dilated pore space in order to attain an easy propagation of the dilation front.

Thresholds. A scalar field for the minimal principal stress has been introduced to determine pressure thresholds for dilation relatively to the minimal principal stress. Different pressure thresholds may be defined for pore opening and closure.

Anisotropy. A simple anisotropy model has been implemented into the TOUGH2 code. The user may define a number of micro-crack networks each with a specific dilation threshold, permeability-pressure and porosity-pressure-relationship and tensor for the anisotropy of permeability (Which does not imply that the dilated pores have to be cracks in reality).

Gas component. The gas component of the EOS7 module has been changed from air to H₂. This includes changes to the molar mass, specific enthalpy, Henry-Isotherm and viscosity of the H₂-vapour mixture. Molar mass, specific enthalpy and Henry-Isotherm were taken from the EOS5-module which is why the Henry-Isotherm does not consider salinity yet. Viscosity of the gas phase is of minor importance in systems where a gas phase and a more viscous liquid phase are displaced simultaneously. Flow is then usually governed by the viscosity of the liquid. In the model presented here, the gas phase may migrate without displacing the liquid phase, so that more attention has to be paid to the viscosity of the gas phase. A new viscosity function for H₂-vapour mixtures has been implemented according to kinetic gas theory with data from /LAN 69/ and a pressure correction derived from the viscosity data used in the EOS5-module (note that viscosity depends only slightly on pressure).

Automatic time step control. Using the automatic time step control of TOUGH2, time step widths can exceed ranges that have to be met to simulate the dilation-controlled transport processes appropriately. The automatic time stepping procedure provided by TOUGH2 has therefore been modified to achieve better results in this respect.

Input routines. Input routines have been implemented so that all dilation parameters can be defined using external files.

3.2 CONVENTIONAL TWO-PHASE FLOW

The governing equations of TOUGH2 and EOS7 will be delineated according to Pruess et al. (1999). Heat transport will not be considered which means that no energy balance equations will be presented in the following although they can be solved by TOUGH2. It also means that all processes are assumed to be approximately isothermal.

Three components, water, brine and air, are considered by the equation-of-state module EOS7. Using a component index k ($=1$ for water, $=2$ for brine, $=3$ for the gas component), the following mass balance equations are solved by TOUGH2

$$\frac{\partial M^\kappa}{\partial t} = -\text{div} (F_{\text{adv}}^\kappa + F_{\text{dif}}^\kappa) + q^\kappa \quad (1)$$

where M^κ are local mass densities. F_{adv}^κ and F_{dif}^κ are advective and diffusive flow densities, respectively, and q^κ are source terms for each component. Note that hydrodynamic dispersion is not part of the standard TOUGH2 code.

Diffusive flow density of component k in phase β ($\beta=1$: gas phase, $\beta=2$: liquid phase) is given by

$$F_\beta^\kappa = -\phi\tau_0 \sum_\beta \tau_\beta (S_\beta) \rho_\beta d_\beta^\kappa \nabla X_\beta^\kappa \quad (2)$$

where ϕ denotes porosity, ρ_β the density of phase β , X_β^κ the mass fraction and d_β^κ the molecular diffusion coefficient of component κ in phase β . $\tau_0 \tau_\beta(S_\beta)$ is the tortuosity, which itself is split up into a rock dependent factor τ_0 and a saturation dependent factor $\tau_\beta(S_\beta)$. Using effective diffusion coefficients equation (2) can be rewritten as

$$F_\beta^\kappa = -\sum_{\beta} D_{\text{eff},\beta}^\kappa \nabla(\rho_\beta X_\beta^\kappa) \quad (3)$$

with effective diffusion coefficient

$$D_{\text{eff},\beta}^\kappa = \phi \tau_0 \tau_\beta(S_\beta) d_\beta^\kappa. \quad (4)$$

This equation will be needed later on to determine $d_{\text{liq}}^{\text{H}_2}$ from effective diffusion coefficients.

The diffusion coefficients of gases is a function of pressure and temperature

$$d_\beta^\kappa(p, T) = d_\beta^\kappa(p_0, T_0) \frac{p_0}{p} \left(\frac{T + 273.15^\circ\text{C}}{273.15^\circ\text{C}} \right)^\theta \quad (5)$$

with $\theta=1.80$.

TOUGH2 offers three tortuosity models

$$\tau_0 \tau_\beta = \tau_0 k_{r,\beta}(S_\beta) \quad (\text{relative permeability model}) \quad (6)$$

$$\tau_0 \tau_\beta = \phi^{1/3} S_\beta^{10/3} \quad (\text{Millington-Quirk-Model}) \quad (7)$$

$$\tau_0 \tau_\beta = \tau_0 S_\beta \quad (\text{constant diffusivity}) \quad (8)$$

of which the Millington-Quirk-Model will be applied. Assuming this model and using equations (4) and (5), the diffusion coefficient $d_{\text{liq}}^{\text{H}_2}$ at reference state (p_0, T_0) can be determined from the effective gas diffusion coefficient under full saturation $D_{\text{eff,liq}}^{\text{air}}$ by

$$d_{\text{liq}}^{\text{air}}(p_0, T_0) = \frac{D_{\text{eff,liq}}^{\text{air}}}{\phi^{4/3}} \frac{p}{p_0} \left(\frac{273.15^\circ\text{C}}{T + 273.15^\circ\text{C}} \right)^\theta \quad (9)$$

where p and T specify the pressure and temperature applied in the diffusion experiment.

The advective flow density of a component in the mass balance equation (1) is defined by the flow densities F_β in the respective phases:

$$F_{\text{adv}}^\kappa = \sum_{\beta} X_\beta^\kappa F_\beta, \quad (10)$$

where X_β^κ is the mass fraction of component κ in phase β . The phase flows F_β in equation (10) are determined by a generalised Darcy law

$$F_{\beta} = -k_{\beta} \frac{k_{r,\beta} \rho_{\beta}}{\mu_{\beta}} (\nabla p_{\beta} - \rho_{\beta} g). \quad (11)$$

Here, for phase β , k_{β} is the absolute and $k_{r,\beta}$ the relative permeability, ρ_{β} the density and μ_{β} the dynamic viscosity. g is the vector of gravitational acceleration. In the language of TOUGH2 the term

$$k_{\beta} \frac{k_{r,\beta}}{\mu_{\beta}}$$

is called “mobility”.

As a default, absolute gas permeability in equation (11) is set equal to the absolute liquid permeability, but TOUGH2 can account for a lowering of the absolute gas permeability at low pressures p by means of the Klinkenberg relation with Klinkenberg parameter b (Klinkenberg 1941)

$$k_{\text{gas}} = k_{\text{liq}} \left(1 + \frac{b}{p} \right). \quad (12)$$

Phase pressure p_{β} in the definition of the phase flows (11) is the sum of the gas pressure p_{gas} and the (negative) capillary pressure $p_{c,\beta}$ of phase β :

$$p_{\beta} = p_{\text{gas}} + p_{c,\beta}. \quad (13)$$

There are several options in TOUGH2 to define capillary pressure functions $p_{c,\beta}(S)$. Most simulations presented here will make usage of the van Genuchten function (van Genuchten 1980, Pruess et al. 1999)

$$p_{c,\text{liq}} = -p_{\text{entry}} \left(S_{\text{eff}}^{-\lambda-1} - 1 \right)^{1-\lambda} \quad (14)$$

where p_{entry} is the apparent gas entry pressure and S_{eff} the effective saturation

$$S_{\text{eff}} = \frac{S_l - S_{\text{lr}}}{S_{\text{ls}} - S_{\text{lr}}} \quad (15)$$

with liquid saturation S_l , residual liquid saturation S_{lr} and maximum liquid saturation S_{ls} . Negative capillary pressure is held constant below a certain pressure limit $p_{c,\text{min}}$.

For equation (11) TOUGH2 offers several relative permeability functions. In the simulations presented here, relative permeability of the pore space unaffected by pathway dilation will be modelled using Corey’s curves (Corey 1954, Pruess et al. 1999)

$$k_{r,\text{liq}} = S_{\text{eff}}^4 \quad (16)$$

$$k_{r,\text{gas}} = (1 - S_{\text{eff}})^2 (1 - S_{\text{eff}}^2) \quad (17)$$

with effective saturation

$$S_{\text{eff}} = \frac{S_1 - S_{\text{lr}}}{1 - S_{\text{lr}} - S_{\text{gr}}}. \quad (18)$$

Here, S_1 is the liquid saturation, S_{lr} the residual liquid saturation, and S_{gr} the residual gas saturation, with $S_{\text{lr}} + S_{\text{gr}} < 1$.

3.3 THERMODYNAMIC PROCESSES

Thermodynamic processes will be modelled using a modified version of the equation-of-state module EOS7, which in its original version considers two phases (gas phase and liquid phase) and three components (water, brine and air). For the application to a HLW repository with dominant production of H_2 it was necessary to change the gas component from air to H_2 . This includes a redefinition of the molar mass, the specific enthalpy, the viscosity of the gas-vapour mixture and the Henry-Isotherm. Redefining the viscosity was important with regard to the conceptual model for the gas flow due to pathway dilation (see next chapter) since in this model the gas phase does not need to displace water in order to migrate, and this implies that the physical properties of the gas phase become very important for its flow. Using EOS7 instead of EOS5 makes it possible to consider saline groundwater or the transport of a tracer by using the brine component. However, no brine will be addressed in the FORGE project. This simplifies the description of the thermodynamic model to the case of pure water in the following.

H_2 is assumed to behave like an ideal gas and gas pressure p_{gas} is assumed to be the sum

$$p_{\text{gas}} = p_{\text{H}_2} + p_{\text{vapour}} \quad (19)$$

of partial H_2 and vapour pressures p_{H_2} and p_{vapour} , respectively. EOS7 does not account for any vapour lowering effects owing to capillary and phase adsorption effects. For this reason, vapour partial pressure p_{vapour} is set equal to the saturated vapour pressure. The latter is calculated by means of steam table equations given by the International Formulation Committee (IFC 1967).

H_2 partial pressure in (19) is calculated according to Henry's law

$$p_{\text{air}} = K_{\text{h}} x_{\text{liq}}^{\text{H}_2}, \quad (20)$$

where K_{h} is the inverse of Henry's constant and $x_{\text{liq}}^{\text{H}_2}$ is the mol fraction of H_2 in the liquid phase. In EOS5, the coefficient K_{h} is set to

$$K_{\text{h}} = \max(1.379 \cdot 10^{-10} \text{ Pa}, 1.697 \cdot 10^{-10} \text{ Pa} - 1.272 \cdot 10^{-12} \text{ Pa K}^{-1} \cdot T)$$

which has been transferred to the EOS7 module. The mol fraction $x_{\text{liq}}^{\text{H}_2}$ in equation (20) is calculated from the mass fractions $X_{\text{liq}}^{\text{H}_2}$ and $X_{\text{liq}}^{\text{water}}$ of H_2 and water in the liquid phase, respectively, by

$$x_{\text{liq}}^{\text{air}} = \frac{X_{\text{liq}}^{\text{air}} \mu_{\text{air}}^{-1}}{X_{\text{liq}}^{\text{air}} \mu_{\text{air}}^{-1} + X_{\text{liq}}^{\text{water}} \mu_{\text{water}}^{-1}}, \quad (21)$$

where μ_{H_2} is the molar mass of H_2 and μ_{water} is the molar mass of water. Liquid water density and vapour density are calculated using the steam table equations mentioned above.

A new viscosity function for H_2 -vapour mixtures was introduced by GRS according to kinetic gas theory with data from (Landolt-Börnstein 1969) and some pressure correction derived from the viscosity data used in the EOS5-module. Pressure has only minor effect on viscosity.

3.4 GAS FLOW DUE TO PATHWAY DILATION

3.4.1 Conceptual model

Due to the dependency of the capillary forces on the pore size, it is assumed that a gas phase will first enter the largest pores of a saturated clay and thus it is assumed that dilation will first affect these large pores. Postulating that dilation tends to locate at pores which are already dilated, only a small pore fraction will be subjected to pathway dilation. Dilating pores are partially filled with water. However, this water, which only presents a very small amount of the water content of the clay, is supposed to be irrelevant in terms of liquid phase and gas phase flow and in terms of capillary pressure for the following reasons:

- The main water content of the clay will only be able to flow inside the non-deformed pore space due to the elevated gas pressure in dilated pores.
- It is assumed that the gas phase will be able to move by deforming the rock matrix without needing to displace pore water.
- If water filled pores are dilated by the gas phase, the capillary pressure will drop due to the increased pore radii. However, the amount of water within dilated pores is very small and can probably be displaced from these pores easily. As soon as this small quantity of water has been removed the dilated pores, capillary pressure will be controlled by the properties of the non-dilated pores. Dilation therefore should not have any major influence on capillary pressures within the assumptions mentioned above (if capillary pressure is expressed as a function of liquid content).

This means that the liquid phase will not take any major advantage from the dilated pores. Thus, a special feature of the proposed conceptual model is that dilation does not affect the flow of the liquid and gas phase equally.

The original porosity in the non-dilated state will be called *primary porosity* in the following, whereas the porosity gained by dilation will be called *secondary porosity*. It has to be noted that dilated pores contribute to both, primary and secondary porosity. However, the pore fraction that belongs to the primary porosity is supposed to be of minor relevance for the flow processes for the mentioned reasons.

At the moment it is assumed that there is no significant hydro-mechanical interaction between dilated and non-dilated pores. However, hydro-mechanical coupling may be introduced into the code in the future.

The consequence of all these assumptions is that water will be present only in the primary porosity and will not notice any dilation effects. Capillary pressures thus remain constant in relation to the water content and water flow will be enhanced only by the pressure of the intruding gas phase but not by dilation of water filled pores. Experimental data indeed indicate that the amount of water displaced by creation of additional gas pathways is very small for both, bentonite (Hoch et al. 2004) and natural clays (Rodwell et al. 2003). In the model presented here, the gas phase is expected to be able to flow through the clay without needing to displace water, i.e. it will be able to move even if the pore water is completely immobile (which might e.g. be forced by boundary conditions or by very rigorous gas entry pressures).

It is assumed that dilation processes are fast in relation to the pressure build up (quick equilibration of pressure and porosity) and that there aren't any memory effects. It shall be possible to describe flows at element scale by Darcy's law (no intermittent gas flow), which

implies that a homogeneous representation of dilation effects must be possible. It is assumed that the hydraulic effects of pathway dilation can be described adequately by a pressure-dependent porosity and a pressure-dependent anisotropic gas permeability.

The assumed quick equilibration implies that the propagation of the front of the dilation zone does not show any kinetic effects and does not give any resistance to the flow of the gas phase. This means that the dilation front can propagate easily through the rock as long as there is sufficient gas supply.

3.4.2 Physical and mathematical model

A pressure-dependent porosity has been introduced into the code TOUGH2/EOS7 in order to create additional storage capacity for the gas phase that allows the gas phase to flow without needing to displace water. The increase of porosity was modelled independently from the porosity-change-feature of TOUGH2 (invoked by prescribing a pore compressibility or expansivity) which just affects the fluxes but not the pore volume. Since porosity changes affect the local physical state (in terms of liquid saturation, capillary pressure, pressure and density of the liquid and gas phase, pressure and density of gas phase, water density, vapour density, density of dissolved H₂, and internal energy), state changes due to the expansion or compression of the pore space were calculated along the thermodynamic processes implemented in the used EOS7-module. Thus, the pressure-dependent change of porosity of TOUGH2-PD does not only alter hydraulic properties but also considers the thermodynamic processes triggered by pore expansion or compression.

An anisotropy of gas flow due to dilation is likely with regard to the textural anisotropy of clay stone. However, the sources of anisotropy is not easy to identify. Anisotropy might e.g. be caused by an anisotropic stress field or rock fabric which favours the growth, opening and interconnection of more or less planar pores with specific orientation and shape. Another reason for anisotropy might be that dilation thresholds vary with the orientation of planar pores due to an anisotropic stress state. It is not clear whether tensile strength, which is different normal and parallel to the bedding plane, significantly influences dilation thresholds if gas pressure increases very slowly. Marschall et al. (Mazureck et al. 2003) assume that clay-rich rock cannot withstand long-term gas pressures with a magnitude greater than the minimum principal stress acting on the rock mass. Thus, the mechanical processes causing an anisotropy of gas flow are complicated and hard to predict without further experimental substantiation and hydro-mechanically coupled simulations. For this reason, a simple anisotropy model was introduced into TOUGH2 by giving the user of the code the opportunity to define a set of micro-crack networks, each with its own dilation threshold, anisotropic permeability-pressure relationship, and porosity-pressure relationship.

Equation (11), which TOUGH2 uses for the calculation of the gas phase flow F_{gas} , was redefined by adding a term describing the sum of the gas phase fluxes in n micro-crack networks

$$F_{\text{gas}} = - \left(\underbrace{\frac{\rho_{\text{gas}}}{\mu_{\text{gas}}} k k_{r,\text{gas}} \mathbf{I}}_{\text{determines the gas flux in the primary porosity}} + \underbrace{\frac{\rho_{\text{gas}}}{\mu_{\text{gas}}} \sum_{i=1}^n \mathbf{K}_{\text{dil},i} k_{\text{dil},i}(p_{\text{gas}})}_{\text{determines the gas flux in the secondary porosity of } n \text{ micro-crack networks}} \right) (\nabla p_{\text{gas}} - \rho_{\text{gas}} \mathbf{g}), \quad (22)$$

where k is the absolute and $k_{r,\text{gas}}$ the relative gas permeability, ρ_{gas} the density, μ_{gas} the dynamic viscosity of the gas phase, and \mathbf{g} the vector of gravitational acceleration. $k_{\text{dil},i}$ is the pressure-

dependent gas permeability of micro-crack network i and tensor $\mathbf{K}_{\text{dil},i}$ is introduced in order to make gas permeability dependent on flow direction.

There are several possibilities to introduce a pressure-dependency of the gas flux e.g. by defining a pressure-dependent relative gas-permeability or intrinsic permeability. Modelling the gas flux through the primary and secondary porosity independently, as it was done here, implies that these fluxes are physically independent which is in accordance with the conceptual model. This approach also allows a decoupling of weighting schemes for the flow in dilated and non-dilated pores. By defining an upwind weighting scheme for the gas flux in the secondary porosity the intended easy propagation of the dilation front according to the assumption of quick equilibration can be achieved.

For $k_{\text{dil},i}$ a power law

$$k_{\text{dil},i}(p_g) \stackrel{\text{def}}{=} \begin{cases} 0 & \text{for } p_g \leq p_{\text{thr},i} \\ \left(\frac{p_g - p_{\text{thr},i}}{\Delta p} \right)^\alpha \Delta k_{\text{dil},i} & \text{for } p_g > p_{\text{thr},i} \end{cases} \quad (23)$$

was used, where $\alpha > 1$ is an arbitrary exponent and Δp defines a pressure interval $[p_{\text{thr},i}, p_{\text{thr},i} + \Delta p]$ over which gas permeability changes by $\Delta k_{\text{dil},i}$.

The porosity ϕ is defined as the sum of the initial porosity ϕ_0 and the porosity of each micro-crack network ϕ_i :

$$\phi = \phi_0 + \sum_{i=1}^n \phi_i(p_g). \quad (24)$$

Assuming that micro-cracks are parallel plates which open according to a linear elastic law a linear relation between crack porosity ϕ_i and gas pressure

$$\phi_i(p_g) \stackrel{\text{def}}{=} \begin{cases} 0 & \text{for } p_g \leq p_{\text{thr},i} \\ \frac{p_g - p_{\text{thr},i}}{\Delta p} \Delta \phi & \text{for } p_g > p_{\text{thr},i} \end{cases} \quad (25)$$

is postulated which shall be called “*dilation relationship*” in the following. As above, p_g is the gas pressure and p_{thr} a threshold pressure for the onset of dilation. Δp defines a pressure interval $[p_{\text{thr},i}, p_{\text{thr},i} + \Delta p]$ over which porosity changes by $\Delta \phi$. Other, e.g. non-linear, dilation relationships may be defined by the user with the obligation that porosity has to increase monotonously with pressure.

3.4.3 Numerical implementation

3.4.3.1 SEMI-EXPLICIT SOLUTION OF FLOW EQUATIONS

Due to the strong non-linearity of the relationships describing the dilation behaviour, porosity and permeability are changed after completion of a time step in order to avoid convergence problems. This implies that the flow equations are solved with constant porosities and

permeabilities which belong to the previous time step. With regard to this, the flow equations are now solved semi-explicitly by TOUGH2.

3.4.3.2 POROSITY CHANGE

To comply with the user-defined relationships between porosity and pressure and to guarantee mass conservation, porosities, pressures and saturations are changed after each time step. This is done by simulating isothermal compression or expansion of the pore space according to the thermodynamic processes implemented in the EOS7 module (see explanation below). Although TOUGH2 allows the definition of a pressure-dependent pore compressibility it was not possible to use this feature to simulate pore expansion and compression since TOUGH2's pore compressibility feature does not affect the physical porosity but only the calculation of mass flows by assuming that all mass concentrations are linearly increased by the given porosity increase. Yet, a physical increase of porosity is necessary in order to let a gas phase flow through a rock with immobile liquid phase. Therefore, a real porosity change was implemented into the code.

3.4.3.3 EQUILIBRATION

After the flow equations have been solved with constant porosities and permeabilities, local pressures and porosities usually do not satisfy the dilation relationship (25). Thus, after a time step, compression or expansion of the pore space has to take place by adapting the TOUGH2 primary variables in order to satisfy the dilation relationship. Since the user may define arbitrary dilation relationships (with the obligation that porosity increases monotonously with pressure) an iterative method is needed to equilibrate gas pressure and porosity. A slightly modified root-finding algorithm of (Press et al. 1996) is used to find a gas pressure change which would lead to a porosity–pressure pair that satisfies the dilation relationship (25) with sufficient accuracy. This is done for each grid element. The primary variables are updated in advance.

Calculating the porosity change from a given pressure change is not straightforward. Each porosity change in a grid element affects its hydraulic and thermodynamic state. In particular such changes influence liquid saturation, capillary pressure, the density of water, vapour and dissolved H₂, the pressure and density of the liquid and gas phase, and the internal energy. This implies that thermodynamic processes have to be considered in order to calculate the porosity change that belongs to a certain pressure change without violating mass conservation. For this purpose an “inverse” EOS routine has been derived from the standard EOS7 routine in this study (temperatures are held constant). The inverse EOS routine has been implemented with high accuracy with regard to the conservation of the mass of the gas component and to the sum of the masses of water and brine (Navarro 2009). To avoid iterative determination of primary variables in the inverse EOS routine, the brine mass is calculated using the brine mass fraction before (not after) porosity change. This causes some minor errors in the water-brine mass ratio. However, for considerations of gas transport in deep geological repositories small errors in the mass conservation of the brine component should be of minor importance.

The component masses which have to be conserved cannot be derived directly from the primary variables with accuracy. The reason for this is that the primary variable ‘gas saturation’ has the meaning of a gas saturation *after* evaporation of water and dissolution of the gas component has taken place. Thus, thermodynamic processes have to be considered to calculate component masses from the primary variables (this is done by the EOS module) or, vice versa, to determine primary variables for given component masses.

3.4.3.4 DECOUPLING OF FLOWS

The weighting procedure for the gas permeability has been completely decoupled from TOUGH2's weighting procedure for the relative permeabilities and "mobilities" (*relative permeability* \times *absolute permeability* / *viscosity*) of the undisturbed matrix. This means that both terms

$$\frac{\rho_{\beta}}{\mu_{\beta}} k_{\beta} k_{r,\beta} \mathbf{I} + \frac{\rho_{\beta}}{\mu_{\beta}} \sum_{i=1}^n \mathbf{K}_{\text{dil},i} k_{\text{dil},i} (p_g)$$

of equation (22) can be weighted independently. The weighting procedure for the first term can be controlled using the TOUGH2 input. Upstream weighting has been chosen for the second term to allow an easy propagation of the dilation front according to the assumptions made.

3.4.3.5 TIME STEP CONTROL

Initial code tests showed that the automatic time step control provided by TOUGH2 is not sufficient to simulate the dilation-controlled transport process appropriately. Therefore, an additional criterion for the reduction of time step widths was introduced. A time step is now repeated with a reduced time step width if

$$\exists i: (p_t^i \geq p_{\text{thr}}^i \wedge |p_t^i - p_{t+\Delta t}^i| > p_{\text{limit}}) \vee (p_t^i < p_{\text{thr}}^i \wedge p_{t+\Delta t}^i \geq p_{\text{thr}}^i \wedge p_{t+\Delta t}^i - p_{\text{thr}}^i > p_{\text{limit}})$$

where p_t^i and $p_{t+\Delta t}^i$ is the pressure in element i before and after a time step, respectively, p_{thr}^i is the minimal dilation threshold at element i , and p_{limit} is the positive maximum pressure change per time step.

In order to avoid oscillations, p_{limit} should be chosen small enough to prevent the first dilation event in element i to cause a pressure drop below the dilation threshold p_{thr}^i . The proposed time step control also limits the amount by which the dilation relationship (25) is violated.

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