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SIMULATIONS OF THE HYDROGEN MIGRATION OUT OF INTERMEDIATE-LEVEL RADIOACTIVE WASTE DISPOSAL DRIFTS USING TOUGH2

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ABSTRACT

Corrosion and radiolysis processes cause hydrogen to be generated in a repository for intermediate-level radioactive waste (ILW) for at least 10’000 years after the closure of the facility. Two model configurations have been used for the TOUGH2 simulations to investigate the impact of the hydrogen in terms of the pressure build-up and the gas saturation level in the disposal drift and in the surrounding argillaceous rock formation.

1. Cross-section (2D) through the disposal drift, perpendicular to its axis: The overpressure caused by the hydrogen has been found to be less than the expected frac pressure, for the production rate and scheme considered.
2. Longitudinal vertical section (3D) through the disposal, access and main drifts: The flow of gas/water from the disposal drift through/around the sealing plug in the access drift has been found to have limited impact on the overpressure inside the disposal drift.

INTRODUCTION

The French Agency for the Management of Radioactive Waste (Andra) is currently investigating the feasibility of deep geological disposal of radioactive waste in an argillaceous formation (Andra, 2005). The repository would be built in an indurated clay formation around 500 m bgl. A question related to the long-term performance of the repository concerns the impact of the hydrogen gas generated in the wastes on the pressure and saturation fields in the repository and the host rock. In this paper, these questions are addressed for ILW with TOUGH2 simulations.

The general layout of the ILW disposal set-up is presented in Figure 1. The disposal configuration comprises a disposal drift with waste packages embedded in a concrete backfill, a concrete plug, and an access drift with a bentonite sealing plug and another concrete plug toward the main transport drift (concrete plugs are there for mechanical stability).

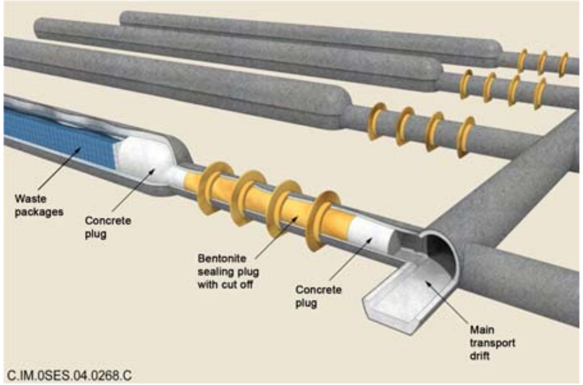

Figure 1. General layout of the ILW part of the repository (Andra, 2005).

GENERAL DISCUSSION OF THE PROCESSES

During the operational phase of the repository, the disposal and access drifts as well as the access and ventilation shafts will be ventilated and the host rock will be progressively depressurized and – to a certain extent – desaturated. As soon as the drifts are backfilled and sealed with bentonite plugs, resaturation of the host rock and the backfill material starts. Due to corrosion of steel components in the waste packages and the radiolysis of bitumen in the packaging, hydrogen will be generated for up to at least 10’000 years.

The processes taking place during and after the hydrogen generation are:

- Dissolution in the porewater (Henry’s law) and transport by diffusion/convection
- Convection of and diffusion in the gas phase

Furthermore, as long as the pressure level remains low as compared to the minimum principal stress, neither micro-fissuration nor macro-fracturation occurs and two-phase flow can be described with the generalized Darcy’s law (Marschall et al., 2005 and Andra, 2005). The flow and transport processes are considered to be isothermal.

The TOUGH2 module EOS5 (water/hydrogen) was used for the computations (Pruess et al., 1999).

2D CONFIGURATION

Geometry And Parameters

The 2D configuration is a half plane through the disposal drift. Outflow from the drift is considered to occur in the radial direction only, i.e., not along the drift axis). The simulation domain has a vertical extension of about 130 m which represents the thickness of the Callovo-Oxfordian argillite at the potential disposal site. The disposal drift has a diameter of approx. 11 m and is backfilled with concrete. It is placed around 500 m below groundlevel. The width of the domain is 50 m and corresponds to half the distance between neighboring disposal drifts. At the upper and lower boundaries of the simulation domain, a constant pressure is imposed which corresponds to the hydraulic heads of the aquifers above and below the argillite. Due to the symmetry of the disposal configuration with a horizontally placed array of disposal drifts, the vertical boundaries are set to no flow conditions. Considerable material details are taken into account in the simulation: a waste package with a high performance cement overpack, emplacement voids, concrete backfill, fractured argillite (thickness ca. 0.5 m), disturbed argillite (i.e. micro-fissured, thickness ca. 4.0 m) and undisturbed argillite. The simulations with this configuration aim at capturing the details of flow close to the waste packages and the surrounding rock.

The hydrogen diffusion flux (in the water and gas phase) is a function of the degree of saturation. In materials such as argillite this dependency is not well known. A conservative choice was made for this study by defining the diffusive flux as a function of the relative permeability function. This minimizes the capacity of the argillite to evacuate the hydrogen and leads to an overestimate of the gas pressure in the drift.

Characteristic flow parameters for a selection of materials are presented in Table 1. The two-phase flow parametrization of the relative permeability capillary pressure saturation constitutive relationship after Van Genuchten – Mualem is used, which is justified also in the case of argillaceous rocks (Marschall et al., 2005 and Croisé et al., 2006).

Initial And Boundary Conditions

The rock mass is assumed to be fully water saturated at the start of the simulation, i.e. the impact of the ventilation in the operational phase of the repository is neglected. This is a conservative assumption when evaluating the pressure build-up resulting from the hydrogen generation inside the cavern because the desaturation would lead to additional gas filled volume and the pressure build-up would be lower.

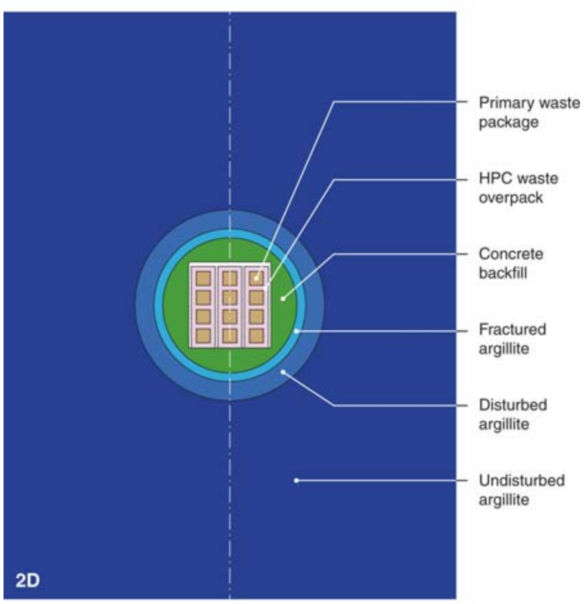

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Figure 2. Schematic of the geometry of the 2D configuration; zoom on the interior of the disposal drift and its near field.

Table 1. Selection of characteristic material parameters for the simulations (2D configuration).

	Waste	High performance cement (waste overpack)	Fractured argillite	Undisturbed Callovo-Oxfordian argillite	Unit
Porosity ϕ	0.25	0.15	0.16	0.15	-
Permeability k	10^{-15}	10^{-19}	$1 \cdot 10^{-16}$	$k_v=5 \cdot 10^{-21}$ $k_h=5 \cdot 10^{-20}$	m^2
Van Genuchten coefficient n	1.5	1.54	1.5	1.49	-
Van Genuchten pseudo gas entry pressure P_0	$3 \cdot 10^3$	$2 \cdot 10^6$	$2 \cdot 10^6$	$1.5 \cdot 10^7$	Pa

The initial hydraulic head is imposed as extracted from the natural gradient between the overlying and underlying aquifers. The materials inside the disposal drift present different degrees of water saturation: 20% in the waste package, 100% for the concrete backfill and the high performance cement overpack. Again, full saturation is a rather unrealistic but conservative assumption with respect to the pressure build-up (tendency to over-estimate the pressure build-up). The emplacement voids between waste packages are considered to be unsaturated initially.

The simulation period is 0 to 10’000 years. The hydrogen gas generation from the waste is imposed in

2 periods with constant rates: from 0 to 500 years, 100 mol/year/m drift length and from 500 years to 10’000 years, 8 mol/year/m drift length.

2D-Mesh And Numerical Challenges

The mesh is unstructured and made of 5’504 volumes. The problem was numerically demanding as lots of different materials were taken into account at scales between 10 cm and several tens of meters. Not surprisingly, because of the high contrasts in permeability (up to 8 orders of magnitude) and capillarity properties of the different materials, several runs ended with convergence problems of the numerical schemes. In most cases they could be solved by adapting the spatial discretization. A TOUGH2 internal adaptive scheme was used for the time discretization. Some cases had run times of several days (or weeks) on high-performance PCs in a Linux cluster, which was felt to be large with respect to the relatively small number of volumes.

3D CONFIGURATION

Geometry And Parameters

The 3D configuration represents an axially symmetric simplification of the actual disposal set-up. It approximates the drifts as a series of concentric “pipes” with the same horizontal axis (see Figure 3). The main transportation drift is tagged on as an additional backfill element even though it runs perpendicular to the access drift. This configuration is used to investigate the impact of flow around the sealing bentonite plug in the access drift on the pressure build-up. Details of the waste material are not taken into account. Instead, the waste and its packaging etc. in the disposal drift is considered as a homogenized material.

The parameters for the undisturbed Callovo-Oxfordian argillite are identical to those listed in Table 1, only the permeability is considered to be isotropic with a value of $5 \cdot 10^{-21}$ m^2. Characteristic flow parameters for the waste, bentonite, concrete and backfill are specified in Table 2. Note that in this configuration, it was assumed that the fractured and disturbed (micro-fissured) argillite no longer exists, which may be the result of self healing of the argillite over time. In terms of pressure build-up in the disposal drift, it is a conservative assumption, as it leads to an overestimation of the pressure in comparison to cases taking into account a more permeable fractured argillite zone.

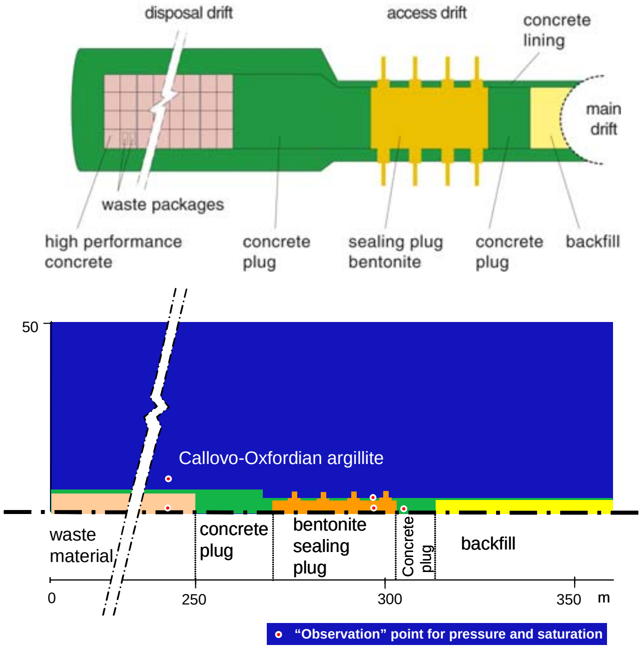

Figure 3. Vertical longitudinal section through the disposal, access and main drifts and the corresponding axially symmetric 3D model.

Initial And Boundary Conditions

As in the 2D configuration, the rock mass initially is fully water saturated. The impact of the vertical gradient is neglected and an initial pressure of 5 MPa is imposed along the upper horizontal boundary. The saturation conditions in the drifts are: 20% in the waste material, 70% in bentonite, concrete and backfill.

The hydrogen gas generation in the waste is imposed in 3 constant rate periods: from 0 to 500 years, 100 mol/year/m drift length, from 500 years to 10’000 years, 7 mol/year/m drift length, and from 10’000 years to 140’000 years a linear decrease from 7 mol/year/m drift length to zero.

3D-Mesh And Numerical Challenges

The rectangular mesh consists of 11’481 volumes, i.e. horizontal hollow cylinders. The same challenging issues were encountered as in the 2D configuration. Furthermore it was necessary to introduce a small amount of initial gas (saturation of 0.001) to achieve convergence of the numerical scheme. Checks of the effect of this “numerical artifact” on the results show that the effects are small and lead to a slight overestimate of the pressure build-up in the waste disposal drift. This is reasonable as the amount of gas in the system is larger with an initial saturation in the full domain. Therefore the dissolved gas is degassing in the vicinity of the drift. Some cases required run times of several days (or weeks) on high-performance PCs in a Linux cluster.

Table 2. Characteristic material parameters for the simulations (3D configuration).

	Waste	Bentonite	Concrete	Backfill	Unit
Porosity ϕ	0.4	0.35	0.15	0.35	-
Permeability k	10^{-14}	$1\cdot10^{-20}$	10^{-18}	$6\cdot10^{-16}$	m^2
Van Genuchten coefficient n	1.5	1.6	1.54	1.4	-
Van Genuchten pseudo gas entry pressure P_0	$1\cdot10^4$	$1.8\cdot10^7$	$2\cdot10^6$	$6\cdot10^5$	Pa

MODELLING RESULTS

Results Of The 2D Configuration

Evolution of the pressure

The evolution of the pressure at selected points is shown in Figure 4.

During the first period of hydrogen generation (0-500 years: 100 mol/year/m), the pressure in the disposal drift increases and reaches a value of 9 MPa. The pressure drive is not sufficient to transport the generated gas away from the waste and through the argillite.

During the second period of hydrogen generation (500-10’000 years: 8 mol/year/m), the pressure decreases at first because the high pressure causes more gas to be transported away than is generated. The pressure then increases again, albeit much slower than in the first period as the gas generation rate is lower. At the end of the simulation period of 10’000 years, the pressure reaches 7.8 MPa.

The (gas) pressures in the waste, the concrete backfill, the fractured argillite and the disturbed argillite are quasi-identical. The water pressure in the undisturbed argillite decreases for the first about 100 years until it hits a minimum at 3.2 MPa. After that, the water pressure increases up to a first maximum of 7.2 MPa at 500 years. At 10’000 years, the pressure in the undisturbed argillite is still around 7.8 MPa.

From the pressure distribution at 500 years (Figure 5), we observe that the water pressure in the undisturbed argillite is also increased several tens of meters away from the disposal drift. The diffusion of the pressure is larger in the horizontal direction due to the assumed anisotropy in permeability.

Evolution of the gas saturation

The time evolution presented in Figure 4 shows that: i) the waste material maintains the high gas saturation (more than 80%), ii) the initially water saturated concrete backfill and the fractured argillite reach a gas saturation of approx. 15%, iii) the gas saturation in the disturbed argillite is low (less than 6%) and iv) the gas saturation in the argillite at around 30 m from the symmetry axis is almost negligible.

The spatial distribution of the gas saturation (Figure 5) in and around the disposal drift presents the following characteristics: The discontinuous saturation levels in the different materials are due to the different parameters of the capillary pressure saturation functions. The main changes in the gas saturation are taking place in the concrete backfill, the HPC overpack and the fractured and disturbed argillite (steady increase over time). In the waste material the changes are small. The changes in the undisturbed argillite are very small (less than 1%). As a result of gravity, the gas saturation in the waste packages along the bottom is slightly smaller than that of those placed along the top.

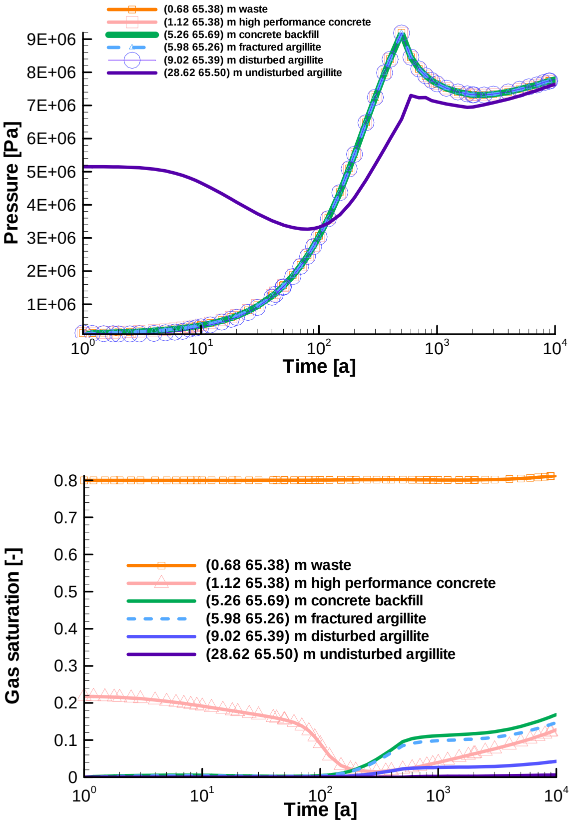

Figure 4. Time evolution of the pressure, and gas saturation in waste, concrete backfill, fractured, disturbed and undisturbed argillite in the 2D configuration.

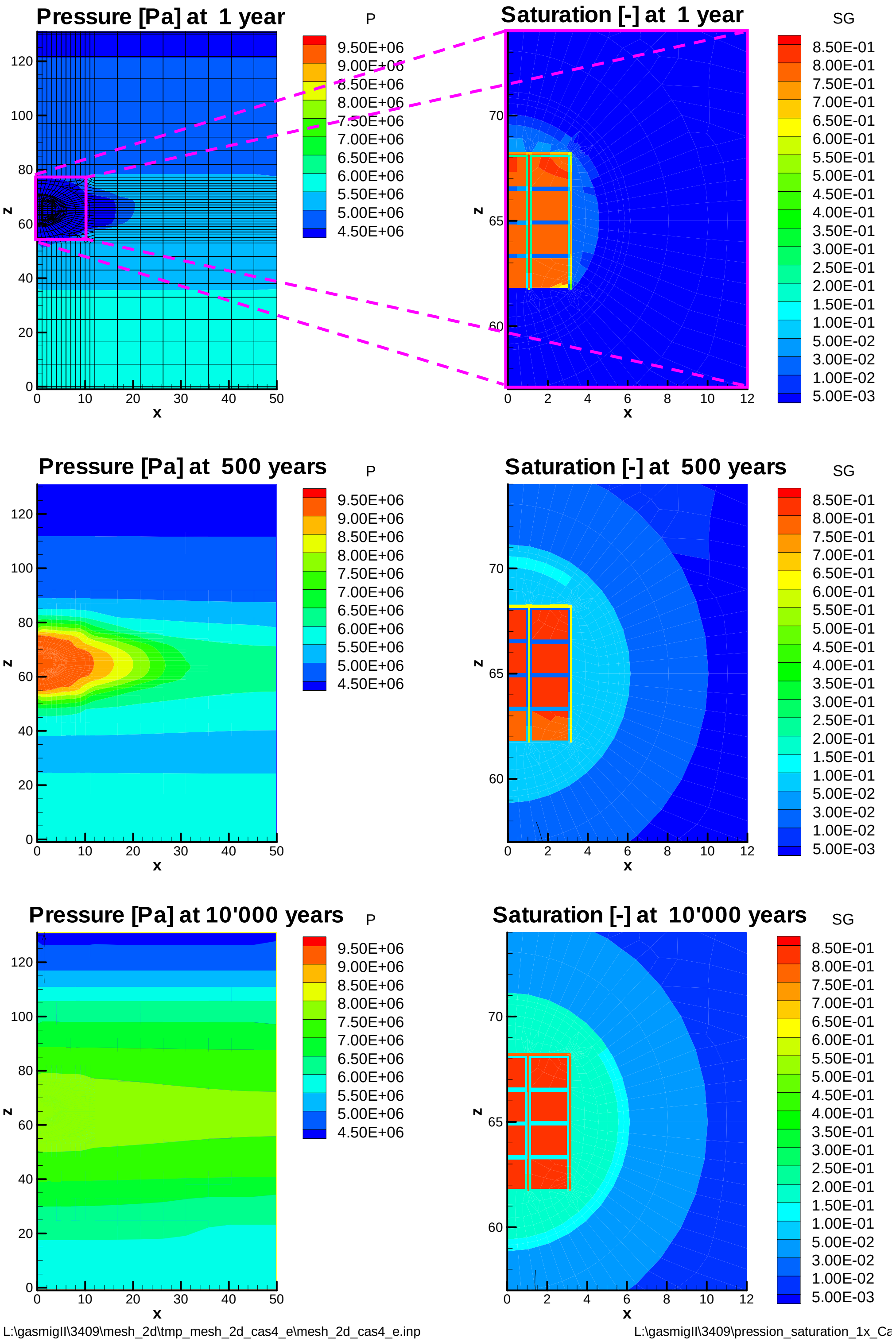

Figure 5. Spatial distribution of the pressure (left) and gas saturation (right) for the 2D configuration; gas saturation presented in a close-up around the disposal drift.

Results Of The 3D Configuration

Evolution of the pressure

During the first period of hydrogen generation (0-500 years: 100 mol/year/m), the pressure in the disposal drift increases and peaks at approx. 5 MPa. This value is much lower than in the 2D configuration. The reason is the higher pore volumes of the waste material and the concrete backfill (see Tables 1 and 2). At 10’000 years the pressure reaches 7.2 MPa which is again lower than the 7.8 MPa of the 2D configuration. This shows that, in the long run, the transport capacity of the surrounding rock is the limiting process for the pressure evolution inside the disposal drift.

Prior to 500 years, the gas pressures inside the drifts are below the hydrostatic level of 5 MPa and the drifts act as a drainage system. After 10’000 years the gas pressure in the waste decreases steadily to a value of approx. 5.2 MPa at 140’000 years.

At 500 years, the pressure in the downstream concrete plug is much lower (approx. 3 MPa) and a significant gas pressure gradient exists between disposal drift and backfilled access and main drift. At 10’000 years, the absolute maximum of 5 MPa is reached in the downstream concrete plug.

Sensitivity runs were performed by adding an interface zone of higher permeability and small thickness (5 cm) between the undisturbed argillite and the bentonite backfill. The results show a larger pressure increase in the backfilled access and main drifts (maximum of 6.8 MPa) and a slightly reduced pressure maximum in the waste (7 MPa).

Evolution of the gas saturation

The resaturation downstream concrete plug starts at approx. 20 years. The resaturation of the bentonite sealing plug is continuous points situated at the center of the bentonite plug start to resaturate at around 200 years (see Figure 6). After 2000-3000 years, the bentonite sealing plug is almost completely resaturated (gas saturation less than a few 0.001).

In the waste, the saturation remains stable at its initial value for approx. 20’000 years, and then a steady but small increase of the gas saturation is observed. This is related to the pressure decrease in the waste package and the degassing of the hydrogen from the water. The gas saturation of the concrete lining increases to 50% until approx. 40 years, as it acts as a preferential path for the gas between the disposal drift and the backfilled access and main drift. It then remains stable for around 500 years and then drops off again due to progressing resaturation.

Around the sealing plug and the disposal drift, the argillite slightly desaturates over time (0-10’000 years) due to the combined effects of the suction of the bentonite and the gas pressure from the disposal drift. A certain amount of gas flow through the argillite and along the sealing plug can be sustained.

Furthermore, the gas progressively penetrates the undisturbed argillite radially outward from the waste drift. However, at a distance less than a few meters from the drift wall, the gas saturation is less than 1%.

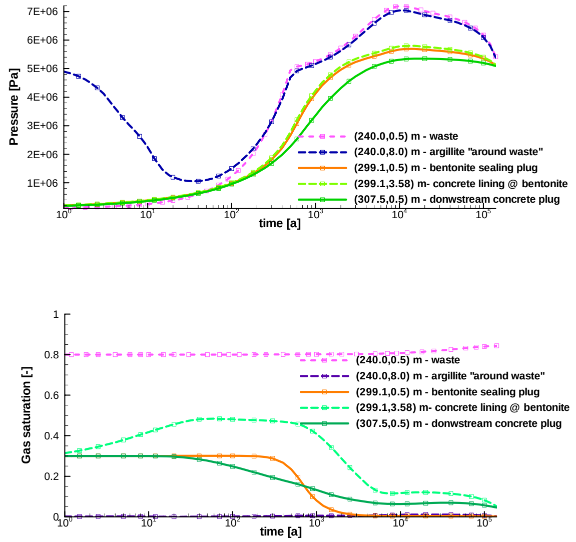

Figure 6. Time evolution of the water pressure, gas pressure and gas saturation at selected observation points (see red dots in Figure 3) in the 3D configuration.

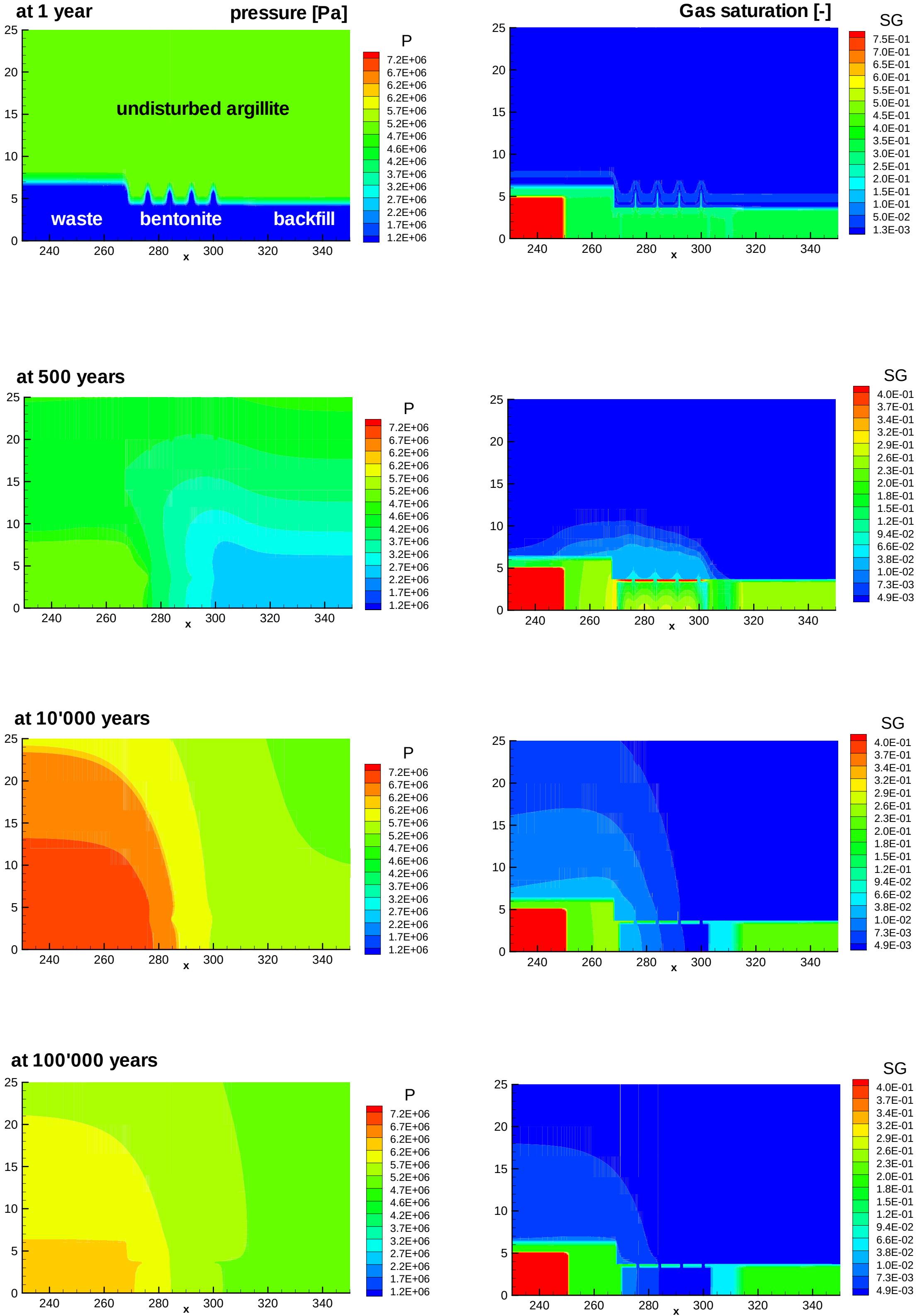

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Figure 7. *Spatial distribution of the pressure (left) and gas saturation (right) for the 3D configuration.*

CONCLUSIONS

To recall, the objective of this study was to perform 2D and 3D simulations to investigate the potential impact of hydrogen gas from radioactive waste on the pressure field in and around a geological repository in the Callovo-Oxfordian argillite.

The 2D simulation performed for a cross section of the waste disposal drift allowed to investigate in detail the gas-water flow in the disposal drift. The problem was numerically challenging as lots of different materials were taken into account at scales between 10 cm and several tens of meters. Numerical convergence issues were solved by adapting the spatial discretization. The results of simulation show that a significant pressure build-up maximum of several MPa above the formation pressure (absolute pressure of 9 MPa) might be reached inside the disposal drift after 500 years, which is assumed to be the end of the period with the high hydrogen generation rate. At the assumed end of gas generation after 10’000 years, the pressure reaches 7.8 MPa. The pressure maximum at 500 years is related to the available gas-filled pore volume. The predicted gas saturations in the saturated argillite are low.

3D simulations were performed in an axi-symmetric configuration (horizontal symmetry axis) and focused on the flow through and around the bentonite sealing plug. They show that a non-negligible gas flow is likely to take place around the sealing plug (e.g. through the concrete lining and the near drift argillite) even though the assumed permeability of the argillite is very low. The simulation results show the interplay of the different materials with strongly differing permeability and two-phase flow constitutive relationships (e.g. suction and mobility). The absolute pressure maximum of 7.1 MPa is reached in the waste at 10’000 years.

ACKNOWLEDGMENTS

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