

# Investigation of Two-Phase Flow Phenomena Associated with Corrosion in an SF/HLW Repository in Opalinus Clay, Switzerland

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## **ABSTRACT**

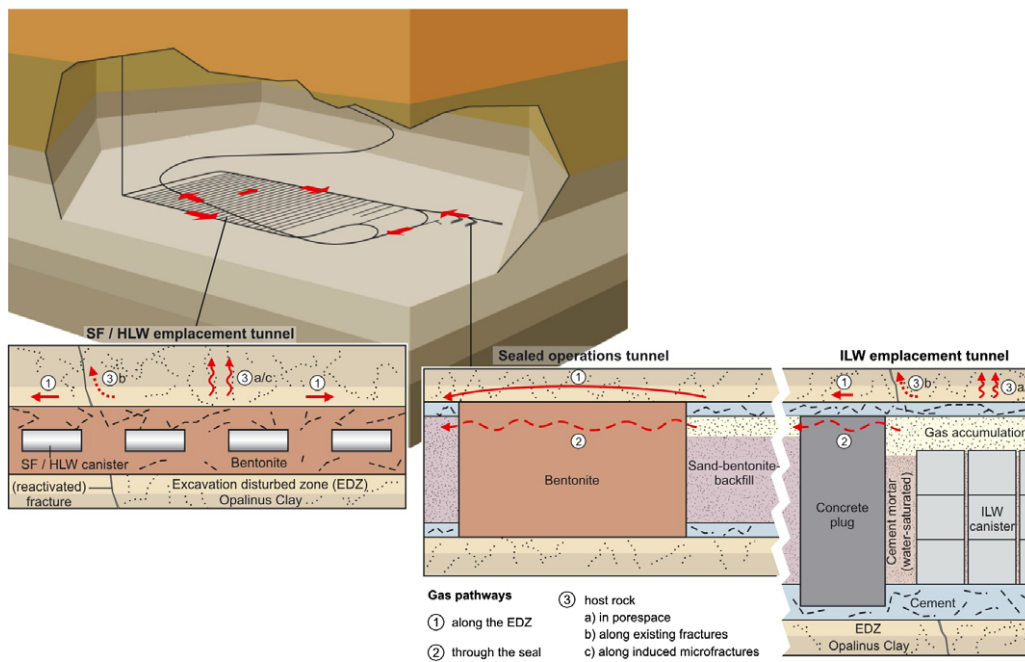
Gas generation from corrosion of the waste canisters and gas accumulation in the backfilled emplacement tunnels is a key issue in the assessment of long-term radiological safety of the proposed repository for spent fuel and high-level waste (SF/HLW) sited in the Opalinus Clay formation of Northern Switzerland. Previous modeling studies indicated a significant pressure buildup in the backfilled emplacement tunnels for those sensitivity runs, where corrosion rates were high and the permeability of the Opalinus Clay was very low. As an extension to those studies, a refined process model of the canister corrosion phenomena has been developed, which accounts not only for the gas generation but also for the water consumption associated with the chemical reaction of corrosion of steel under anaerobic conditions. The simulations with the new process model indicate, that with increasing corrosion rates and decreasing host-rock permeability, pressure buildup increased, as expected. However, the simulations taking into account water consumption show that the pressure buildup is reduced compared to the simulation considering only gas generation. The pressure reduction is enhanced for lower permeability of the Opalinus Clay and for higher corrosion rates, which correspond to higher gas generations rates and higher water consumption rates. Moreover, the simulated two-phase flow patterns in the engineered barrier system (EBS) and surrounding Opalinus Clay show important differences at late time of the gas production phase as the generated gas continues to migrate outward into the surrounding host rock. For the case without water consumption, the water flow indicates overall downward flow due to a change in the overall density of the gas–fluid mixture from that based on the initially prescribed hydrostatic pressure gradient. For the case with water consumption, water flow converges toward the waste canister at a rate corresponding to the water consumption rate associated with the corrosion reaction. The water flow toward the canister is maintained even for very low permeabilities of the Opalinus clay, sustaining the anaerobic corrosion of the steel canister.

## **0. Introduction**

In deep geological repositories for spent fuel and high-level waste (SF/HLW), hydrogen will be produced by anaerobic corrosion of metals. The generation, accumulation, and release of gases from the disposal system may affect a number of processes that influence long-term radiological safety of the repository (NEA, 2001). This issue has become a focus in many national radioactive waste isolation programs, such as in Switzerland (Nagra, 2002a,b,2004), France (Andra, 2005), and Belgium (Ortiz et al., 2002) among others. The safety relevant features, events and processes (FEPs) associated with waste generated gas include (a) excess gas/water pressures could affect the mechanical integrity of the engineered barrier system (EBS), (b) expulsion of contaminated water from the waste package due to the gas buildup, and (c) transport of volatile

radionuclides through the EBS and surrounding geological barriers.

A comprehensive study of gas release was accomplished in the context of Nagra's Geosynthesis study (Nagra, 2002a), comprising a detailed assessment of both the relevant gas transport mechanisms and the gas release paths. Figure 1 shows a schematic representation of Nagra's concept for a SF/HLW/ILW repository in Opalinus Clay with the possible gas pathways from the emplacement tunnels through the EBS and the host rock. Scoping calculations were conducted to simulate gas pressure buildup in a gallery of parallel SF/ HLW tunnels. The assumption was made that the gas flow along the backfilled tunnel can be neglected, which allowed the use of a two-dimensional model geometry in a vertical cross section perpendicular to

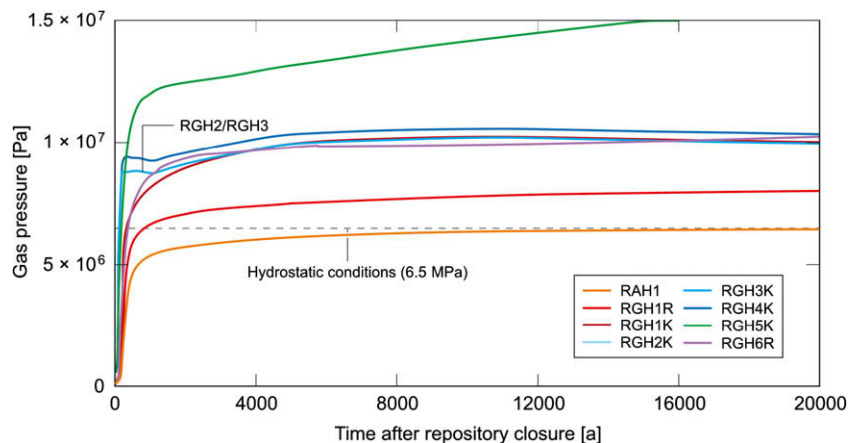


**Fig. 1.** Schematic representation of Nagra's concept for a SF/HLW/ILW repository in Opalinus Clay with the possible gas pathways from the emplacement tunnels through the EBS and the host rock (after Nagra, 2004).

the axis of the emplacement tunnels. The calculations were based on robust assumptions with regard to the gas generation rates (termed “pessimistic” and “over-conservative” gas generation rates of  $0.04 \text{ m}^3/\text{a}$  and  $0.4 \text{ m}^3/\text{a}$ , respectively, corresponding to corrosion rates of  $3.6$  and  $36 \text{ } \mu\text{m}/\text{a}$ ), gas generation scenarios (immediate/delayed onset of gas generation), and the site-specific conditions (variation of intrinsic permeability and gas entry pressure). Furthermore, the simulations included both isothermal and non-isothermal modeling runs (heat production by the SF/HLW). A multitude of sensitivity runs confirmed that it is very unlikely that excessive gas pressures are reached in the emplacement tunnels. In this context, the term “excessive” means that gas pressure reaches the magnitude of minimum stress (around  $15 \text{ MPa}$  at repository level), which is seen as indicative for the onset of irreversible deformation of the intact host rock (Fig. 2; cf. Nagra, 2002a, 2004). Only in the extreme case RGH5K with “over-conservative” gas generation rate, horizontal intrinsic permeability as low

as  $2.0\text{E}-21 \text{ m}^2$  and with a gas entry pressure of  $21 \text{ MPa}$ , did the simulated gas pressure in the emplacement tunnel reach  $15 \text{ MPa}$  after about  $16,000 \text{ yrs}$ . In all other cases the gas pressure did not exceed  $10 \text{ MPa}$ , corresponding to an overpressure of  $3.5 \text{ MPa}$  above hydrostatic conditions.

Even though most of the aforementioned sensitivity runs revealed moderate gas overpressures in the backfilled SF/HLW emplacement tunnels, there was yet a low likelihood of producing excessive pressures, when unfavorable input parameters were assumed. It is obvious that such rare, yet possible parameter combinations could occur in probabilistic safety assessments. Hence, the need was seen for refined process and system models of gas release, which are able to better constrain the degree of conservatism in the basic conceptual assumptions. Among other aspects, the following conservatisms of the previous modeling approach were further investigated in recent studies: (i) the 2-D model geometry does not account for gas release along the backfilled tunnels



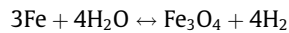
**Fig. 2.** Two-phase flow simulations of pressure buildup in the SF/HLW emplacement tunnels after Nagra (2002a). Modelling runs indicated with “R” (e.g. RGL2R, RGH1R) are assuming gas generation rates, which correspond to a corrosion rate of  $3.6 \text{ } \mu\text{m}/\text{a}$ . The runs marked with “K” (RGL1K, RGH1K) correspond to a corrosion rate of  $36 \text{ } \mu\text{m}/\text{a}$ . Runs RGH3K – RGH6R are non-isothermal simulations. A detailed description of the model parameters is given in Nagra (2002a).

through the excavation damage zone, (ii) previous model approaches assumed a homogeneous host rock, resulting in significant overestimation of the effective gas entry pressure, and (iii) the water consumption for maintaining the corrosion reaction was not considered. The current paper focuses on the effect of water consumption on the gas pressure buildup. Few other studies have considered both gas generation and water consumption associated with the corrosion of the metal waste canister (i.e., Lassabatère et al., 2004).

In the current study the different processes associated with generation and migration of waste-generated gas from a SF/HLW repository were investigated. For this, numerical simulations were performed using a 2-D model geometry to describe the relevant two-phase flow phenomena for the case considering only gas generation compared to the case considering both gas generation and water consumption.

## 1. Corrosion processes and gas transport phenomena

The resaturation of the initially partially saturated repository and thermal perturbation associated with heat generation from radioactive decay of the waste material is expected to last only a few hundred years, after which anaerobic conditions prevail. The corrosion of the steel canisters under anaerobic conditions produces hydrogen gas ( $H_2$ ) and a solid corrosion product magnetite ( $Fe_3O_4$ ) as the more likely chemical reaction, which is described by the following redox reaction:



This chemical reaction indicates that for each mole of  $H_2$  generated 1 Mol of  $H_2O$  is consumed. That is, a sufficient amount of water in the form of liquid water or water vapor is required to maintain the corrosion. Laboratory experiments on corrosion of iron metal under atmospheric conditions indicate that the corrosion rate decreases drastically for a relative humidity decreasing from 90% to 60% (Brown and Masters, 1982). On the other hand, thermodynamic considerations predict that the gas production could continue theoretically at very low rates until the  $H_2$  partial pressure increases above 40 MPa (Neretnieks, 1985).

The reference design for the SF and HLW canisters involves a cast steel body with 15 cm and 25 cm wall thickness, respectively (NAGRA, 2002b). Both canister types are about 1 m in diameter and are surrounded by a 0.75 m thick bentonite buffer in emplacement tunnels which are 2.5 m in diameter (NAGRA, 2002b). In the present study it is assumed that the corrosion and associated  $H_2$  gas generation rate is maintained at constant rates, which range between 0.0112 m<sup>3</sup>/a/tm (tunnel meter) and 0.112 m<sup>3</sup>/a/tm, corresponding to steel corrosion rates of 1 and 10  $\mu$ m/a, respectively. These rates represent the so-called realistic and conservative rates, respectively, with regard to the potential pressure buildup associated with gas generation. The corresponding water consumption rates range from 9 g/a/tm to 90 g/a/tm.

The conversion of Fe (steel) to  $Fe_3O_4$  results in an overall volume increase by a factor of 2.1 (Johnson and King, 2003), which is partly accounted for by an increase in porosity of the magnetite that develops on the surface of the canister. At the upper range of the realistic corrosion rate of 1  $\mu$ m/a (NAGRA, 2004), the corrosion depth is 10 cm after 100,000 yrs which could exert a significant compressive stress on the surrounding bentonite. The migration of  $H_2$  through the bentonite buffer likely occurs much earlier as described below, and the potential geomechanical effects on the surrounding bentonite can be assumed to be small in a first approximation and are not considered in this study. Similarly, the conversion of Fe metal to  $Fe_3O_4$  and corresponding changes in hydraulic properties is not considered in the model. A very small

porosity was assumed for the canister hull to account for the development of potential porosity associated with the conversion of Fe (steel) to  $Fe_3O_4$  (Table 1). Also, a finite permeability was assumed for the waste container to account for potential flow into and out of the container. For the purposes of these calculations, it is arbitrarily assumed, in order to explore the phenomena, that the canisters are “breached” at  $t = 0$ .

Two-phase flow phenomena associated with gas generation from corrosion of waste canisters include initial dissolution of  $H_2$  in the pore water at the corrosion surface of the waste canister and in the surrounding buffer. As pore water becomes fully gas saturated and no more gas can be dissolved, a free gas phase develops causing a significant increase in the gas pressure. Gas starts to migrate into the surrounding bentonite buffer and farther into the Opalinus Clay driven by the pressure buildup. The corrosion rate is maintained as long as sufficient water is available for the redox reaction. The processes associated with the corrosion of the waste canister are focused on the surface of the canister that is in contact with the surrounding bentonite buffer.

The modeling of  $H_2$  gas generation associated with corrosion of the waste canister and the potential impact of  $H_2O$  consumption on gas pressure buildup is implemented using the two-phase flow code TOUGH2 (Pruess, 1991), accounting for the different mass transfer processes which are depicted schematically in Fig. 3. For this study, the  $H_2$  migration assuming initially fully water-saturated conditions is simulated using the Equation-of-State module EOS5 for two-phase flow of hydrogen and water. Advective flow is described with a multiphase extension of Darcy's law. Diffusive mass transport is considered only in the gas phase. Interference

**Table 1**

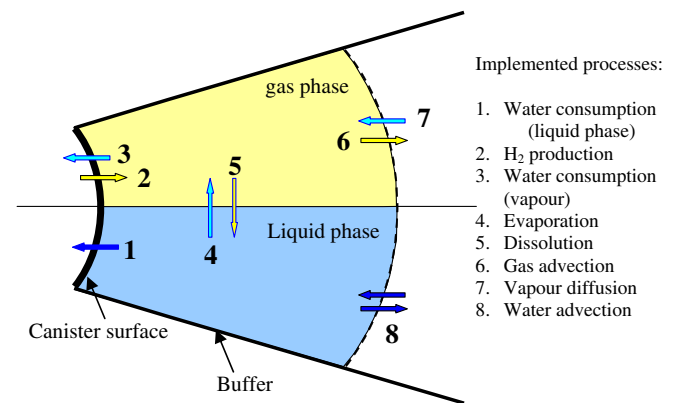
Input parameters for reference case, modified from Nagra (2002a).

	Container <sup>a</sup>	Bentonite	Opalinus Clay
Porosity (–)	0.17 <sup>b</sup> , 1.E–4 <sup>c</sup>	0.40	0.12
Permeability (m <sup>2</sup> )	$1 \times 10^{-19}$	$1 \times 10^{-19}$	$1 \times 10^{-20}$
Pore compressibility ( $\alpha/\phi$ ) (Pa <sup>-1</sup> )	$2.14 \times 10^{-9}$	$3.58 \times 10^{-9}$	$1.83 \times 10^{-9}$
Residual liquid saturation (–)	0.0	0.3	0.5
Initial liquid saturation (–)	1.0	1.0	1.0
Residual gas saturation (–)	0.0	0.0	0.0
Van Genuchten parameter $n$ (–)	2.0	1.82	1.67
Gas entry pressure (Pa)	1.0	$1.8 \times 10^7$	$1.8 \times 10^7$

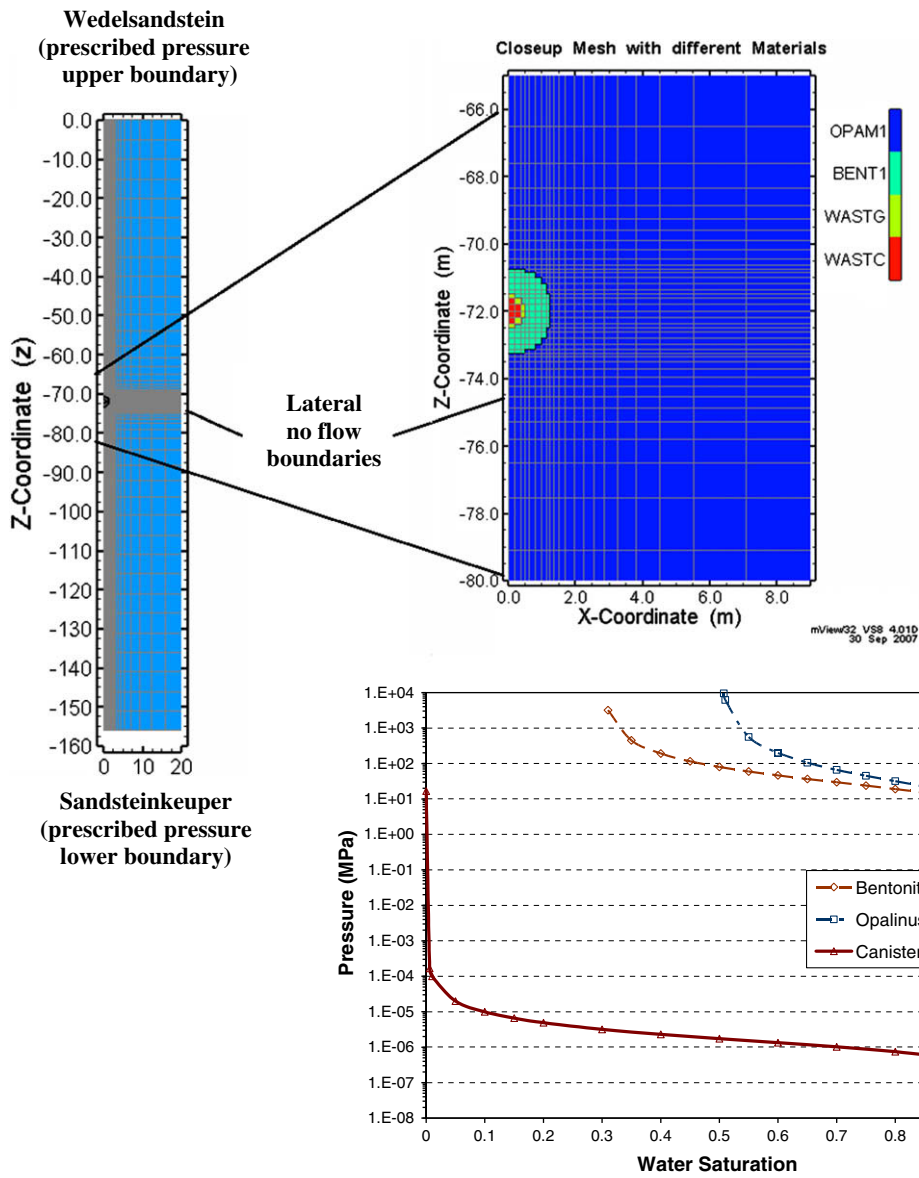
<sup>a</sup> Container represented by a equivalent porous medium with fictitious values for porosity, permeability and gas entry pressure.

<sup>b</sup> Average porosity assumed for waste material.

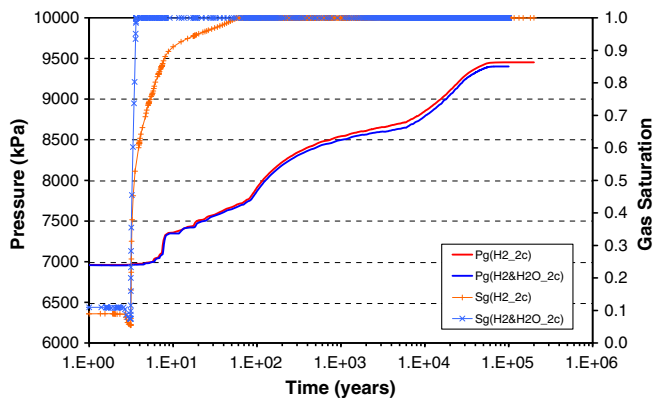
<sup>c</sup> Reduced porosity assumed for container hull.



**Fig. 3.** Schematic representation of the mass transfer phenomena, which were implemented in the two-phase flow model for simulating the water consumption associated with the corrosion processes on the canister hull.



**Fig. 4.** Modell geometry with the 2D mesh, showing the entire vertical cross section (left) and a close-up in the vicinity of the repository (right). The different materials are colour coded: container with waste material (red) and surrounding canister wall (yellow), bentonite (green), and Opalinus Clay (blue). The capillary pressure – saturation relationships of the different materials are shown in the lower diagram. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 5.** Simulated gas pressure and gas saturation in the canister hull for case with reference permeability and pessimistic gas generation rates.

between the phases is represented by means of relative permeability and capillary pressure functions. Solubility of hydrogen gas in water is represented by Henry's law.

The static conditions for maintaining the corrosion reaction in terms of relative humidity can be assessed based on the potential desaturation of the bentonite, as described below. However, it is assumed that water is transported toward the canister either as liquid water or water vapor, which is consumed in the canister producing  $H_2$  and  $Fe_3O_4$ . Water vapor can be transported toward the canister by diffusion. For vapor diffusion, the diffusion coefficient given by Vargaftik (1975) and Walker et al. (1981) is used, which depends on pressure, saturation, and tortuosity. In this study, a tortuosity of one was used for all materials. Net vapor flow toward the canister is limited due to the gas pressure buildup around the canister, and vapor will be more likely transported by advection together with the generated  $H_2$  gas away from the canister.

The detailed physico-chemical processes at the interface between the canister surface and the surrounding bentonite are



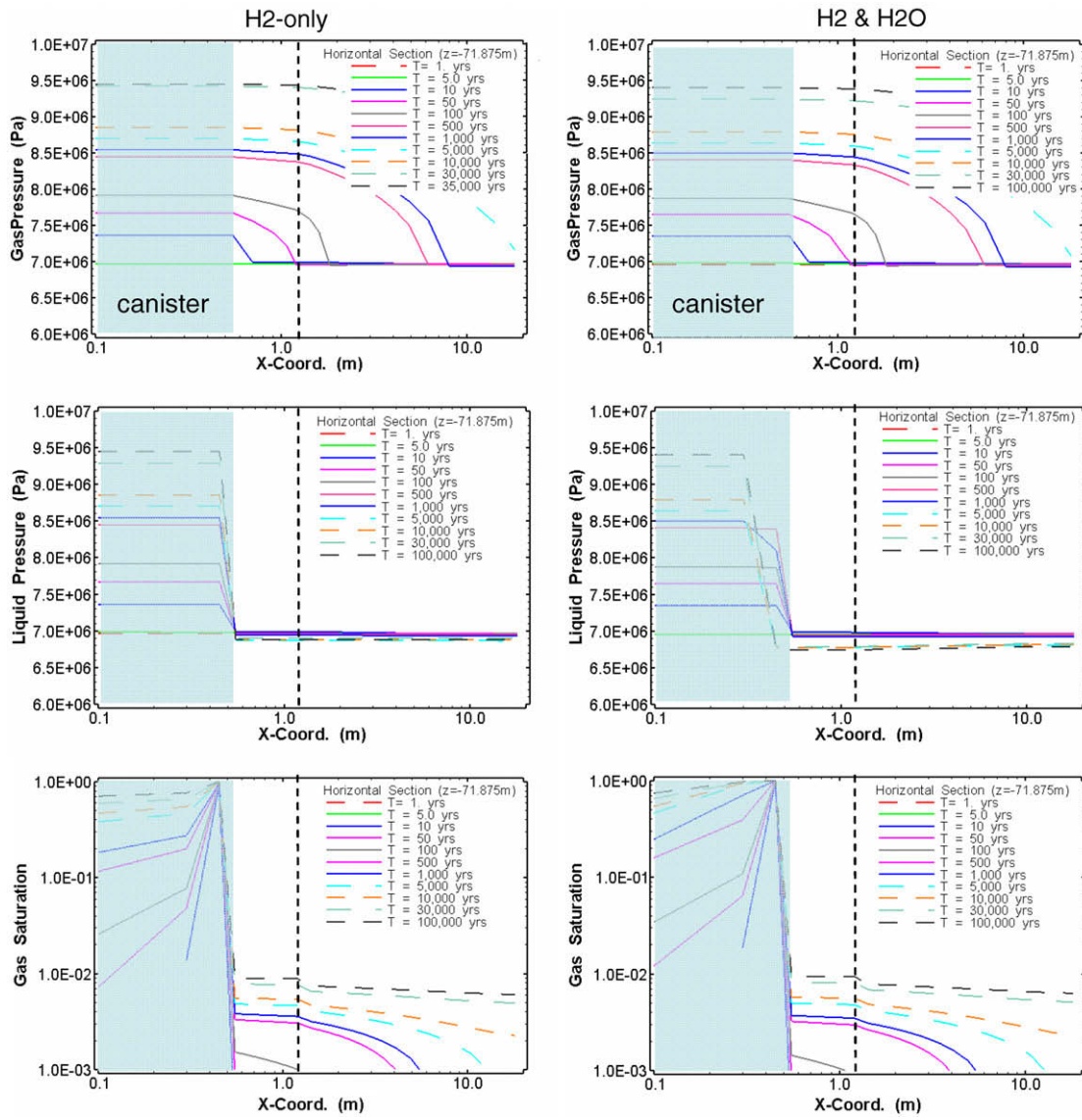


Fig. 6. Profiles of gas pressures, liquid pressures, and gas saturations with and without water consumption at different characteristic time.

complex and are approximated in the numerical model by prescribing the reaction products in terms of gas generation rates and water-consumption rates to the elements representing the canister hull (Fig. 3). In order to consider water withdrawal under two-phase conditions, a code change was necessary in the TOUGH2 code to allow only the liquid phase to be extracted. In the standard TOUGH2 version, the withdrawal rate represents a mass withdrawal of the fluid mixture, which can consist of both gas and liquid, depending on either the phase condition in the element or the mobility of the element connection. The potential effect of removal of dissolved  $H_2$  associated with the prescribed water consumption was considered small as indicated in sensitivity simulations not presented here.

## 2. Modeling approach

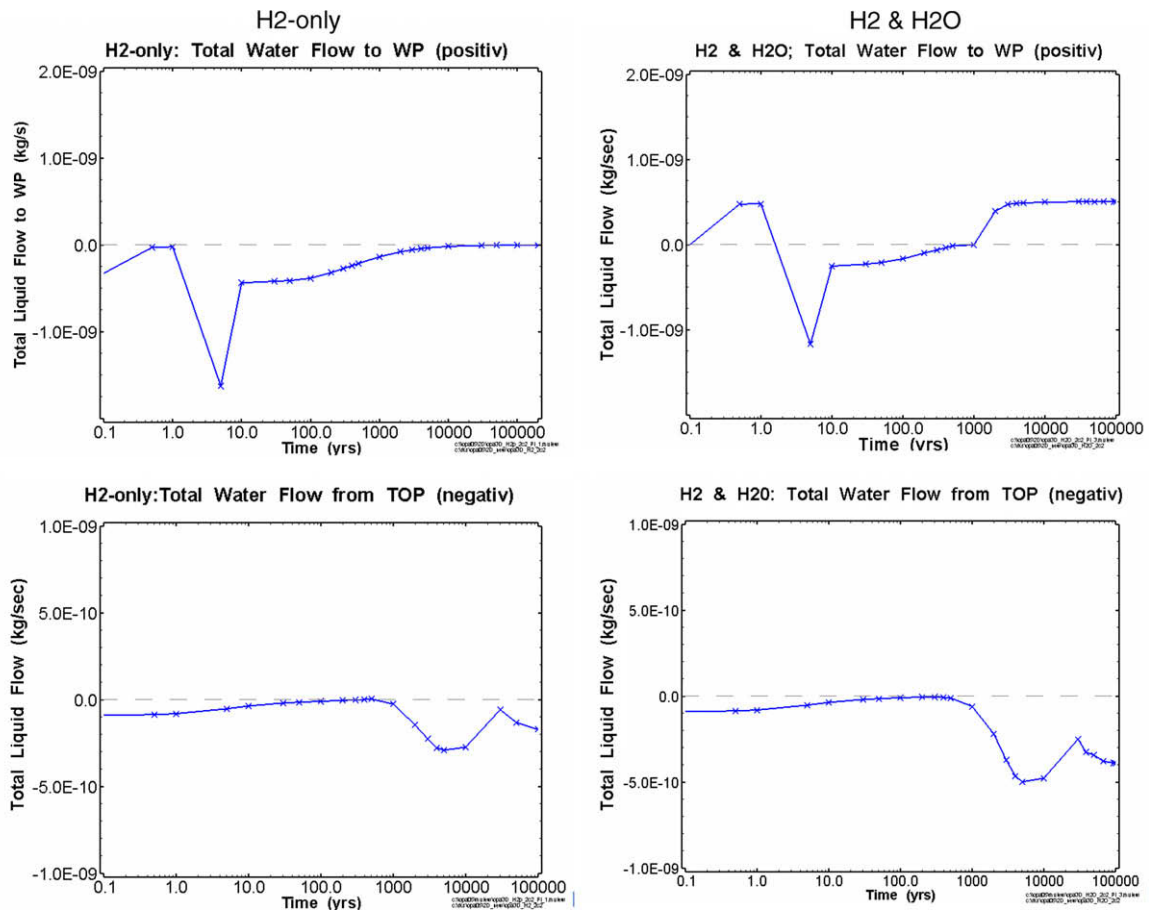
The model geometry is represented by a vertical cross section perpendicular to the array of parallel emplacement tunnels. The radial geometry of the canister and the emplacement tunnel is approximated by discrete rectangular elements (Fig. 4a). The top and bottom boundaries of the model represent the contact of the

Opalinus Clay with the overlying Wedelsandstein and underlying Lias; both formations are assumed to represent local aquifers with hydrostatic head. The left boundary depicts the vertical symmetry through the emplacement tunnel; the right boundary exhibits the symmetry between two adjacent emplacement tunnels which are 40 m apart.

The properties for the different materials – waste canister, bentonite buffer, and Opalinus Clay – are summarized in Table 1.

The capillary pressure – saturation relationships of the different materials are shown in Fig. 4b. The container is represented by an equivalent porous medium with fictitious values for porosity, permeability and gas entry pressure. The capillary pressure curve of the container is relatively low and exhibits a significant increase only when the water saturation goes to zero (i.e., at completely dry conditions). This allows approximation of capillary suction pressure development associated with a reduction in relative humidity at the canister surface due to  $H_2$  gas accumulation from anaerobic corrosion.

At the same time, the canister is in contact with the surrounding bentonite which is characterized by relatively high capillary pressures (Fig. 5). Assuming that the saturation conditions of the



**Fig. 7.** Computed water flow across the canister-bentonite interface (top) and across the top boundary (bottom) for the simulation considering only  $H_2$  generation (left) and  $H_2$  generation &  $H_2O$  consumption (right).

bentonite adjacent to the canister surface controls the relative humidity, the corresponding capillary pressure is given by Kelvin's equation (Edlefsen and Anderson, 1943):

$$P_c = \ln(RH) \cdot \rho_w \frac{RT}{M_w}$$

where  $RH$  is the relative humidity,  $\rho_w$  is the density of water,  $R$  is the gas constant,  $T$  is the absolute temperature (degree Kelvin), and  $M_w$  is the molecular weight of water. The minimum relative humidity of 60%, required to maintain the corrosion reaction of the waste canister, amounts to a capillary suction pressure of 74 MPa. Based on the capillary pressure curve for bentonite (Fig. 4), a capillary pressure of 74 MPa corresponds to a water saturation of about 50%. It is unlikely that the bentonite can be desaturated by the generated  $H_2$  displacing that much pore water from the bentonite requiring a gas pressure of more than 80 MPa. This pressure would significantly exceed the minimum principal stress of the surrounding Opalinus Clay at the repository depth of about 650 m. The gas flow would be enhanced by pathway dilation and macro-fracturing and such a high gas pressure buildup could not be maintained. The threshold pressure for dilatancy-controlled gas flow through the Opalinus Clays is in the order of 13–15 MPa (Nagra, 2004).

A relative humidity of 90% above which the corrosion is at a maximum (Brown and Masters (1982) amounts to a capillary suction pressure of about 15 MPa, which corresponds to a water saturation of 85%. This amount of desaturation of the bentonite may be achieved by the  $H_2$ -induced pressure buildup, although at these pressures gas transport would likely be enhanced by pathway dilation (Marschall et al., 2005). Potential pathway dilation or fractur-

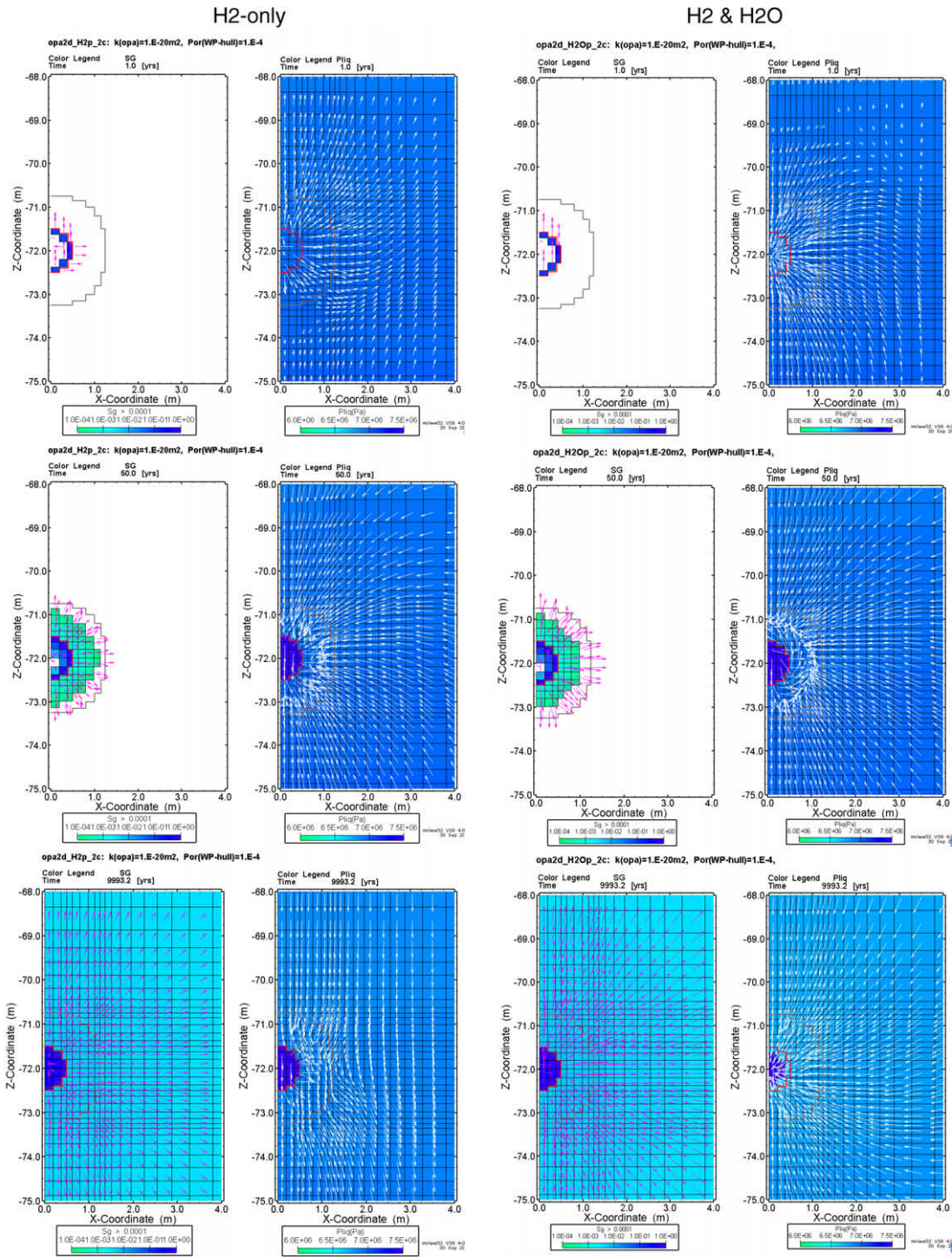
ing at high pressures and the corresponding increase in permeability may significantly affect the capillary pressure – saturation relationship which is not considered. The current study assumes isothermal conditions and focuses on the principal two-phase flow phenomena associated with gas generation from anaerobic corrosion of the waste canister.

For this study the different simulations were performed in two implementations for comparison, which included (a) considering only  $H_2$  generation (designated as “ $H_2$ -only”), and (b) accounting for both  $H_2$  generation and  $H_2O$  consumption (designated as “ $H_2$ & $H_2O$ ”). In a first step, a base-case simulation was performed using the input parameters given in Table 1, and prescribing the pessimistic corrosion rate of 3.6  $\mu\text{m/a}$  at the canister in terms of  $H_2$  generation and  $H_2O$  consumption.

In a second step, sensitivity simulations were performed using (a) the pessimistic corrosion rate of 3.6  $\mu\text{m/a}$  and varying the permeability of the Opalinus Clay between  $1\text{E}-20$  and  $1\text{E}-22\text{ m}^2$ , and (b) using a low permeability of the Opalinus Clay of  $1\text{E}-21\text{ m}^2$  and varying the corrosion rate between 1 and 36  $\mu\text{m/a}$ . Further parameter sensitivities were investigated but are not discussed in this paper.

### 3. Modeling results

The results of the simulation for the reference case permeability of the Opalinus Clay ( $k = 1\text{E}-20\text{ m}^2$ ), the  $H_2$  generation rate of  $0.04\text{ m}^3/\text{a/tm}$  and corresponding  $H_2O$  consumption rate of  $9\text{ g/a/tm}$  are shown in terms of the pressure buildup and gas saturation



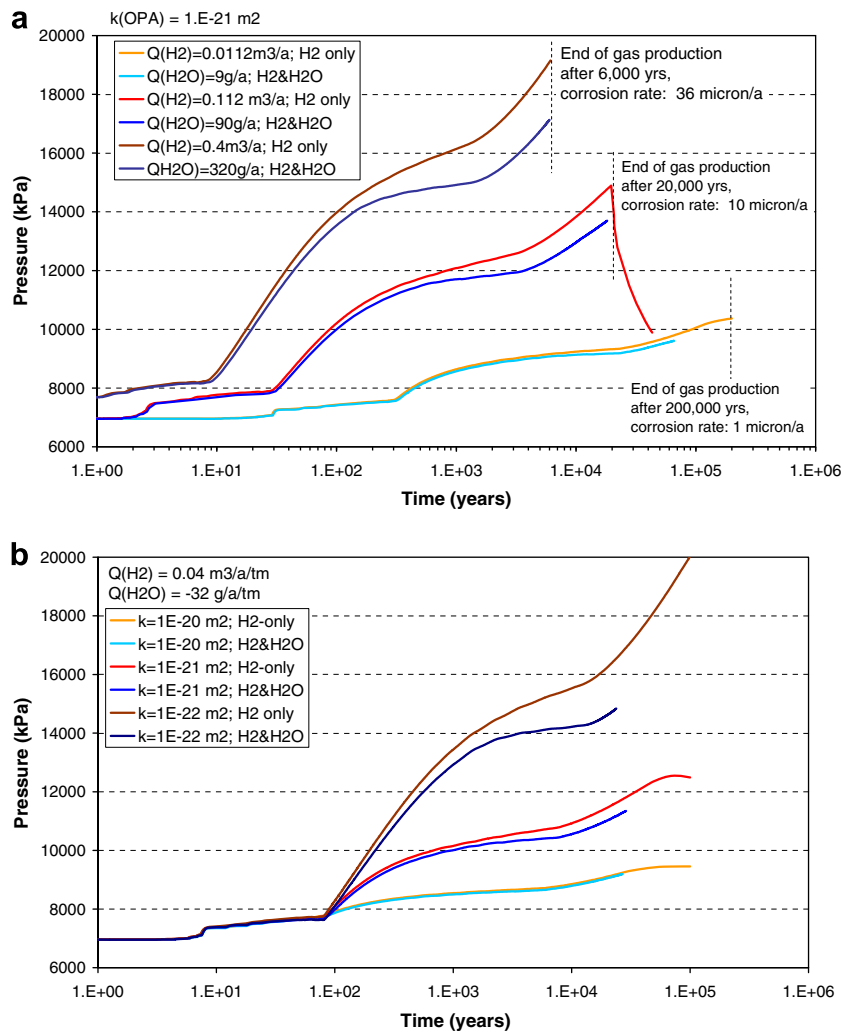
**Fig. 8.** Simulated distribution of gas saturation (greater than 0.001) and gas flow (left) and liquid pressure and water flow (right) for the H<sub>2</sub>-only and the H<sub>2</sub>&H<sub>2</sub>O simulation after (a) 1 yr, (b) 50 yrs, and (c) 10,000 yrs.

in the canister hull in Fig. 5. The gas pressure shows a step increase at early time followed by a near linear increase over a period between 10 and 100 yrs (Fig. 5). The simulated gas saturation indicates that a free gas phase develops very early because of the very small porosity assumed for the canister hull. The early pressure increase at 10 yrs follows a rapid increase in gas saturation. The gas saturation in the canister hull for the H<sub>2</sub>&H<sub>2</sub>O case increase rapidly to 99.95% within 4 yrs and reaches 100% after 10,000 yrs, whereas the case with H<sub>2</sub> generation only shows a delayed increase in gas saturation reaching 99.95% after 50 yrs followed by

a continued increase in to a maximum gas saturation of 99.98% after 100,000 yrs.

The gas pressure response for the two simulations are similar, with the H<sub>2</sub>-only case showing slightly higher pressures than the H<sub>2</sub>&H<sub>2</sub>O case. After about 100 yrs the gas phase reaches the low-permeability host rock and the gas pressure increase steepens. The pressure increase after about 10,000 yrs (Fig. 5) is caused by the lateral no-flow boundary, representing the symmetry axis between two adjacent repository drifts. The pressure increases for both simulations start to level off after the gas front reaches the





**Fig. 9.** Results of sensitivity analyses on gas pressure buildup for: (a) of gas generation rate (over-conservative, conservative and realistic rates with  $k = 1\text{E}-21 \text{ m}^2$ , and (b) intrinsic permeabilities ranging between  $1\text{E}-20$  and  $1\text{E}-22 \text{ m}^2$  with the pessimistic corrosion rate of  $3.6 \mu\text{m/a}$  (corresponding to a gas generation rate of  $0.04 \text{ m}^3/\text{a/tm}$ ).

upper boundary at about 30,000 yrs and becomes constant after about 70,000 yrs when the gas front reaches the lower model boundary.

The horizontal profiles of gas pressure, liquid pressure, and gas saturation at the repository level for the H<sub>2</sub>-only case and the H<sub>2</sub>&H<sub>2</sub>O case are shown in Fig. 6 at different times. The gas pressures increase continuously from the waste canister, to the bentonite, and into the surrounding Opalinus Clay, whereas the liquid pressures increase noticeably only in the waste canister for the H<sub>2</sub>-only case. In the surrounding bentonite and Opalinus Clay, the pressure remains nearly constant at the initial pressure.

As mentioned above, the porosity of the canister hull is assumed to be very small ( $\phi = 1\text{E}-4$ ) compared to that of canister fill ( $\phi = 0.17$ ) representing the waste material in the center. The gas saturation in the canister hull shows early increases for both simulations due to the small porosity, which results in a free H<sub>2</sub> gas phase prior to any noticeable increase in the gas pressure (Fig. 5). The gas saturations increase to near 100% for both cases, but reach only a maximum of 1% in the adjacent bentonite and in the surrounding clay after 100,000 yrs.

For the H<sub>2</sub>&H<sub>2</sub>O case, the liquid pressure in the canister hull indicates a decline after about 500 yrs (Fig. 6) compared to the pressure in the canister center which essentially corresponds to the gas pressure. As indicated in Fig. 4b, the capillary pressure

curve of the canister material increases drastically as the liquid saturation goes to zero (i.e.,  $S_l < 0.0001$ ). The small differences in saturation and corresponding changes in capillary pressure cause significant changes in the liquid pressure and, in turn, in the water flow gradient. In the H<sub>2</sub>-only case, there is only liquid flow from the Opalinus Clay toward the bentonite but not into the canister. For the H<sub>2</sub>&H<sub>2</sub>O case, the liquid pressure in the canister hull decreases to the level of the surrounding bentonite, producing a gradient toward the canister for liquid flow at the prescribed water consumption rate (the time steps at which water flow is into the canister are identified by dashed lines in Fig. 6).

The total water flow to the canister (indicated by a positive flow rate) is shown in Fig. 7 for both H<sub>2</sub>-only and H<sub>2</sub>&H<sub>2</sub>O cases, respectively. The H<sub>2</sub>-only case indicates only water flow from the canister, representing displaced pore water initially from the canister hull and subsequently from the canister center (at about 5 yrs). At late time water flow from the canister decreases to near zero when the gas saturation approaches 100% (Fig. 5). The H<sub>2</sub>&H<sub>2</sub>O case shows a similar pattern of displaced pore water out of the canister, but indicates positive flow rates at about 1 yr and at late times which corresponds to the water consumption rate prescribed in the canister hull (Fig. 6).

Both the H<sub>2</sub>-only and H<sub>2</sub>&H<sub>2</sub>O cases indicate overall downward flow which is represented by a total inflow across the upper model



boundary (Fig. 6). For the H<sub>2</sub>-only case, the inflow into the top of the model at late time corresponds approximately to the outflow at the base of the model. For the H<sub>2</sub>&H<sub>2</sub>O case the total inflow into the top at late time approaches the H<sub>2</sub>O consumption rate at the waste canister. The variation in total inflow at the top (Fig. 6) varies associated with the gas breakthrough at the top boundary (after about 30,000) and bottom boundary (after about 70,000 yrs). Furthermore, in the H<sub>2</sub>&H<sub>2</sub>O case there is also some water inflow from the bottom boundary of the model.

The spatial distributions of gas and water flow associated with the pressure buildup of the H<sub>2</sub>-only case and H<sub>2</sub>&H<sub>2</sub>O case is shown in Fig. 8 at specific times. As indicated in Fig. 5, a free gas phase developed early in the canister hull because little pore water is available for dissolving the generated H<sub>2</sub>. Pore water is displaced initially from the canister hull (Fig. 8a) but the outflow rate decreases after 1 yr (Fig. 7). More water is then displaced from the center of the waste canister before the gas starts to migrate into the surrounding bentonite shown by the water fluxes in Fig. 8b. As the gas starts to migrate into the bentonite causing an increase in capillary pressure and a concomitant decrease in liquid pressure, the water flow is reversed toward the bentonite (Fig. 8b bottom). With continued gas generation, the gas front migrates into the surrounding Opalinus Clay at which time the gas flow is outward (Fig. 8c). For the H<sub>2</sub>-only case, the water flow in the vicinity of the repository is predominantly downward, whereby the water flow is diverted around the canister (Fig. 9c) with no water flow into the canister as indicated in Fig. 7. For the H<sub>2</sub>&H<sub>2</sub>O case the water flow near the repository is toward the canister (Fig. 8c), whereby the water flow to the canister corresponds to the water-consumption rate associated with the corrosion.

The simulations results for the second step, evaluating the sensitivity of different corrosion rates and permeability of the Opalinus Clay on the pressure buildup are summarized in Fig. 9. Varying the corrosion rates also affects the total duration of the corrosion until the entire 0.2 m thick hull of the Fe canister is converted into Fe<sub>3</sub>O<sub>4</sub>. That is, for the realistic rate for gas generation (0.0112 m<sup>3</sup>/a/tm) the corresponding corrosion rate is 1 µm/a for 200,000 yrs. The conservative rate (0.112 m<sup>3</sup>/a/tm) corresponds to a corrosion rate of 10 µm/a for 20,000 yrs, and the over-conservative rate (0.4 m<sup>3</sup>/a/tm) corresponds to a corrosion rate of 36 µm/a for 6000 yrs.

For the reference permeability for the Opalinus Clay of 1E–21 m<sup>2</sup>, the simulated gas pressures at the canister for the different corrosion rates are shown in Fig. 9a. The results indicate that with increasing corrosion rates the difference in the simulated pressure buildup for the H<sub>2</sub>-only and H<sub>2</sub>&H<sub>2</sub>O increases. That is, the effect of water consumption results in significantly lower pressure compared to the simulations considering H<sub>2</sub> generation (Fig. 9a).

The simulations for a pessimistic corrosion rate of 3.6 µm/a (i.e. gas generate rate 0.04 m<sup>3</sup>/a/tm) indicate that the pressure buildup significantly increases with decreasing permeability. More importantly, the effect of the water consumption results in significantly lower pressure for the simulations with lower permeabilities (Fig. 9b).

#### 4. Conclusions

A refined process model has been developed and applied for the simulation of gas release from a SF/HLW repository. The processes associated with the corrosion of the canisters were implemented in a two-phase flow model by prescribing the corresponding hydrogen gas generation rate together with the water consumption rates. Previous scoping calculations on gas release considered only the source term (gas generation) but neglected the sink term (water consumption).

The comparison of the refined process model with the classical approach reveals some distinct differences, which are relevant for constraining the degree of conservatism in probabilistic assessments of the impact of gas release on repository safety. The following conclusions can be drawn with regard to the concept of a SF/HLW repository in the Opalinus Clay formation:

- For the base-case simulation in this study (i.e., pessimistic gas generation rate and intrinsic rock permeability of 1E–20 m<sup>2</sup>), the pressure buildup in the emplacement tunnels is moderate (gas overpressures of 2–3 MPa above hydrostatic) and the impact of water consumption on pressure buildup is small.
- Significant gas pressures are expected for high gas generation rates, corresponding to corrosion rates ≥ 10 µm/a and for very low host rock permeabilities ≤ 1E–21 m<sup>2</sup>. The simulations taking into account water consumption show that the pressure buildup is reduced compared to the simulation considering only gas generation. The reduction in pressures is enhanced for lower permeabilities of the Opalinus Clay and for higher corrosion rates, which correspond to higher gas generations rates and higher water consumption rates.
- At late time when flow conditions in the near-field have stabilised, water flow is toward the canister opposing the outward H<sub>2</sub> gas migration, with the water flow rate corresponding to the prescribed water consumption rate. For both the H<sub>2</sub>-only and the H<sub>2</sub>&H<sub>2</sub>O cases, the gas migration from the repository indicates only little pore-water displacement from the EBS at early time; at late time overall water flow is downward and toward the canister.

In the present study, simplifying assumptions were made in terms of the representation of corrosion and of the waste package in the numerical model. Further studies are being conducted to include explicit representation of the chemical reaction associated with the anaerobic corrosion of metal, and couple the chemical reaction model to the two-phase flow model to comprehensively investigate the detailed chemical and two-phase flow processes.

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