Modelling and Numerical Simulation of Gas Migration in a Nuclear Waste Repository

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Abstract We present a compositional compressible two-phase, liquid and gas, flow model for numerical simulations of hydrogen migration in deep geological radioactive waste repository. This model includes capillary effects and gas diffusivity. The choice of the main variables in this model, total or dissolved hydrogen mass concentration and liquid pressure, leads to a unique and consistent formulation, no matter whether there is a gas phase or not. After introducing and explaining our model, we show computational evidences of its adequacy to simulate gas phase appearance and disappearance in different but typical situations for gas migration in an underground radioactive waste repository.

Keywords Two-phase flow, compositional flow, porous medium, underground nuclear waste management, gas migration

Contents
1 Introduction .................................. 1
2 Modeling Physical Assumptions .............. 2
3 Liquid Saturated/Unsaturated state; a general formulation . 4
4 Numerical experiments ........................ 7
5 Concluding remarks .......................... 15

1 Introduction

The simultaneous flow of immiscible fluids in porous media occurs in a wide variety of applications. The most concentrated research in the field of multiphase flows over the past four decades has focused on unsaturated groundwater flows, and flows in underground petroleum reservoirs. Most recently, multiphase flows have generated serious interest among engineers concerned with deep geological repository for radioactive waste, and for CO₂ capture and storage simulations (CSS). For radioactive waste geological repositories in Europe, which are in Argillaneous or Granitic rocks, there is a growing awareness that the effect of hydrogen gas generation, due to anaerobic corrosion of the radioactive waste packages steel engineered barriers (carbon steel overpacks and stainless steel envelopes), can affect all the functions allocated to the canisters or to the buffers and even to the backfill (see [18], [31], [35]). The host rock safety function may even be threaten by overpressurisation, [7], leading to opening fractures in the host rock and inducing groundwater flow and transport of radionuclides outside of the waste site boundaries. Our ability to understand and predict underground gas migration is crucial in the designing and the performance assessment of any reliable geological nuclear waste storage.

In nuclear waste management, the migration of gas through the near field environment and the host rock, involves two components, water and pure hydrogen H₂; and two phases: “liquid” and “gas”. Due to the inherent complexity of the physics, equations governing this type of flow in porous media are nonlinear and coupled. Moreover the geometries and material properties characterizing many applications can be quite irregular and contrasted. As a result of all these difficulties, numerical simulation often offers the only viable approach to modeling transport and multiphase flows in porous media.
An important consideration, in the modeling of fluid flow with mass exchange between phases, is the choice of the primary variables that define the thermodynamic state of the system. When a phase appears or disappears, the set of appropriate thermodynamic variables may change. There are two different approaches to that problem. The first one, widely used in simulators such as TOUGH2, [34], relies on a primary variable substitution algorithm. This algorithm uses in two-phase conditions the appropriate variables like pressure and saturation, and, when a transition to single-phase conditions occurs, it switches to new variables adapted to the one-phase conditions, like pressure and concentration. This variable substitution is done after each Newton iteration according to some “switching criteria”, see [13,19,32,36]. A different presentation of this approach was done recently in [22,24], where the solubility conditions are formulated as complementary conditions which complement the conservation law equations. The whole system is then solved by a semismooth Newton method as in [16,37], which consists in working on an intermediate active node set (see [24]). The second possibility is to use a set of primary “persistent” variables, such as pressure and component density, which will remain well defined when phase conditions change, so that they can be used throughout the single and two-phase regions, like in [8,3].

This paper addresses the problem of the phase appearance/disappearance through a single set of persistent variables, well adapted to heterogeneous porous media, which does not degenerate and hence could be used, without requiring switching the primary variables, as an unique formulation for both situations: liquid saturated and unsaturated. We will demonstrate through four numerical tests, the ability of this new formulation to actually cope with the appearance or/and disappearance of one phase in simple, but typical and challenging situations like the ones we met in underground radioactive waste repository simulations.

Although the application we had in view for this model was the gas migration in geological radioactive waste repositories; we are aware that the very same problem of phase appearance and disappearance is also crucial in modelling the recently discussed technology of Carbon Capture and Storage (see for instance [14, 15]).

2 Modeling Physical Assumptions

We consider herein a porous medium saturated with a fluid composed of 2 phases, liquid and gas, and according to the application we have in mind, we consider the fluid as a mixture of two components: water (only liquid) and hydrogen (H₂, mostly gas) or any gas with similar thermodynamical properties. In the following, for sake of simplicity we will call hydrogen the non-water component and use indices w and h for the water and the hydrogen components.

According to our goal, which was to focus on the phase appearance and disappearance phenomena, we have done several simplifying assumptions which are not essential for understanding our approach. Not doing these assumptions would not have affected neither our choices of primary variables nor the conclusions, but they would have considerably complicated the present paper.

- The porous medium is assumed to be in thermal equilibrium. This hypothesis can be questionable in the case of application to nuclear waste repository where heat is generated by the nuclear waste, but, as argued in [7], the near field thermal characteristic time is usually smaller than the corrosion time, so that the most of the hydrogen production take place when the system is close to thermal equilibrium. Hence, although thermal flux and energy conservation could be taken easily in account, for simplicity, they will not be discussed herein and we will consider only isothermal flows.

- After restoring thermal equilibrium in the repository and resaturation of the clay engineered barriers (in several hundred years, [7]), the water pressure will be sufficiently high compared to the water vapour pressure and the water vaporization can be safely neglected.

- Although at the depth of some storages, the water density could be affected by the pressure, we suppose for simplicity in our presentation that the water component is incompressible. For the very same reasons, the porous medium is supposed rigid, meaning that the porosity \( \Phi \) is only a function of the space variable \( \Phi = \Phi(x) \).

- We are assuming that the gas flow can be described by the generalized two-phase Darcy’s law, and we are not taking in account the possibility, in clayey rocks of having the gas transported by other mechanism, see [29].

The two phases are denoted by indices, \( l \) for liquid, and \( g \) for gas. Associated to each phase \( \alpha \in \{l, g\} \), we have, in the porous medium, the phase pressures \( p_\alpha \), the phase saturations \( S_\alpha \), the phase mass densities \( \rho_\alpha \) and the phase volumetric fluxes \( q_\alpha \). The phase volumetric fluxes are given by the Darcy-Muskat law (see [33,4])

\[
q_l = -k(x)\lambda_l(S_l)(\nabla p_l - \rho_l g), \\
q_g = -k(x)\lambda_g(S_g)(\nabla p_g - \rho_g g),
\]

where \( k(x) \) is the absolute permeability tensor, \( \lambda_\alpha(S_\alpha) \) is the \( \alpha \)-phase relative mobility function, and \( g \) is the gravity acceleration; \( S_\alpha \) is the effective \( \alpha \)-phase saturation and then satisfies:

\[
S_l + S_g = 1. \tag{2}
\]

Pressures are connected through a given capillary pressure law (see [25,5])

\[
p_c(S_g) = p_g - p_l. \tag{3}
\]
From definition (3) we notice that \( p_c \) is a strictly increasing function of gas saturation, \( p_c' (S_g) > 0 \), leading to a capillary constraint:

\[
p_g > p_l + p_c(0),
\]

where \( p_c(0) \geq 0 \) is the capillary curve entry pressure (see Figure 2).

The water component and the gas component which are naturally in liquid state and in gas state at standard conditions are also denoted respectively solvent and solute. We will assume herein, for simplicity that the mixture contains only one solvent, the water and one gas component, the hydrogen.

Writing all the quantities relative to one component with the superscript \( i \in \{w, h\} \), we define then \( M_i' \) as the molar mass of the \( i \)--component and \( \rho_{i}' \), \( c_{i}' \), \( X_{i}' \), as, respectively: the dissolved mass and the dissolved molar densities, the molar fraction, of the \( i \)--component in the \( \alpha \)--phase, \( \alpha \in \{l, g\} \). All these quantities satisfy:

\[
\begin{align*}
\rho_{\alpha}^i &= M_i' c_{\alpha}^i, \quad X_{\alpha}^i = \frac{c_{\alpha}^i}{c_{\alpha}}, \\
\rho_{\alpha} &= \sum_{k=w}^{h} \rho_{k}^i, \quad c_{\alpha} = \sum_{k=w}^{h} c_{k \alpha}.
\end{align*}
\]

As said before, in the gas phase, we neglect the water vaporization and we use the ideal gas law (see [17]):

\[
\rho_{g} = C_v p_g,
\]

with \( C_v = M_i^h/(RT) \), where \( T \) is the temperature, \( R \) the universal gas constant.

Mass conservation for each component leads to the following differential equations:

\[
\begin{align*}
\Phi \frac{\partial}{\partial t} (S_l \rho_w^l) + \text{div} (\rho_w^l \mathbf{q}_l + j^l_w) &= \mathcal{F}_w, \\
\Phi \frac{\partial}{\partial t} (S_g \rho_g^l + S_g \rho_g^g) + \text{div} (\rho_g^l \mathbf{q}_l + \rho_g \mathbf{q}_g + j^g) &= \mathcal{F}_g,
\end{align*}
\]

where the phase flow velocities, \( \mathbf{q}_l \) and \( \mathbf{q}_g \), are given by the Darcy-Muskat law (1), \( \mathcal{F}_w^k \) and \( j^l_k \), \( k \in \{w, h\} \), are respectively the \( k \)--component source terms and the diffusive flux in the liquid phase (see eq. (13)).

Assuming water incompressibility and independance of the liquid volume from the dissolved hydrogen concentration, we may assume the water component concentration in the liquid phase to be constant, i.e.:

\[
\rho_w^w = \rho_w^{std},
\]

where \( \rho_w^{std} \) is the standard water mass density. The assumption of hydrogen thermodynamical equilibrium in both phases leads to equal chemical potentials in each phase: \( \mu_h^b(T, p_g, X_h^h) = \mu_l^b(T, p_l, X_l^h) \). Assuming that in the gas phase there is only the hydrogen component and no water, leads to \( X_g^h = 1 \), and then, from the above chemical potentials equality, we have a relationship \( p_g = F(T, p_l, X_l^h) \). Assuming that the liquid pressure influence could be neglected in the pressure range considered herein and using the hydrogen low solubility, \( \rho_g^h \ll \rho_g^l = \rho_g^{std} \), we may then linearise the solubility relation between \( p_g \) and \( X_h^h \), and obtain the Henry’s law \( p_g = K_h X_h^h \), where \( K_h \) is a constant specific to the mixture water/hydrogen and depends on the temperature \( T \) (see [17]). Furthermore, using (9) and the hydrogen low solubility, the molar fraction, \( X_h^h \), reduces to \( \frac{\rho_g^h M_h^w}{\rho_w^{std}} \) (see eqs. (9)-(11) in [8]) and the Henry’s law can be written as

\[
\rho_g^h = C_h p_g,
\]

where \( C_h = HM_h^b = \rho_g^{std} M_h^w/(M_w^w K_h^b) \); with \( H \), the Henry’s law constant.

Remark 1 On the one hand the gas pressure obey the Capillary pressure law (3) with the constraint (4), but on the other hand it should also satisfy the local thermodynamical equilibrium and obey a solubility equation like the Henry’s law (10). More precisely if there are two phases, i.e. if the dissolved hydrogen mass density, \( \rho_g^h \), is sufficiently high to lead to the appearance of a gas phase \( (S_g > 0) \), we have from (10) and (3):

\[
\rho_g^h = C_h (p_l + p_c(S_g)).
\]

Moreover, \( S_g > 0 \) with the capillary constraint (4) and the Henry’s law (10), give the solubility constraint:

\[
\rho_g^h > C_h (p_l + p_c(0)).
\]

But if the dissolved hydrogen mass density, \( \rho_g^h \), is smaller than the concentration threshold (see Figure 1), then there is only a liquid phase \( (S_g = 0) \) and none of all the relationships (3) or (12), connected to capillary equilibrium, applies anymore; we have only \( S_g = 0 \), with \( \rho_g^h \leq C_h p_g \).
The concentration threshold line, \( \rho^h_0 = C_h(p_l + p_c(0)) \) in the phase diagram, is then separating the one phase (liquid saturated) region from the two phase (unsaturated) region.

The existence of a concentration threshold line can also be written as unilateral conditions:

\[
0 \leq S_g \leq 1, \quad 0 \leq \rho_0^h \leq C_h p_g, \quad S_g (C_h p_g - \rho_0^h) = 0;
\]

which could be added to the conservation laws (7)-(8), and solved conjointly at each time step by means of a semi-smooth Newton’s method, as explained in the Introduction and in [22] or [24].

Since hydrogen is highly diffusive we include the dissolved hydrogen diffusion in the liquid phase. The diffusive fluxes in the liquid phase are given by the Fick’s law

\[
j_l = -D_h \frac{\rho_l}{\rho_l + \rho_i} \nabla \rho_l^h,
\]

and

\[
j_i = -D_h \frac{\rho_i}{\rho_i} \nabla \rho_i^h.
\]

drogen component molar fractions (see eqs. (12) and (13) in equations (5), (6), (9), and (13), doesn’t appear,

But in the liquid saturated regions, where the gas phase doesn’t appear, \( S_l = 1 \), the system (14)–(18) degenerates to:

\[
\frac{\partial \rho_l^h}{\partial t} + \text{div}(\rho_l^h \nabla \rho_l^h) = \mathcal{F}_l^h,
\]

\[
\frac{\partial \rho_i^h}{\partial t} + \text{div}(\rho_i^h \nabla \rho_i^h) = \mathcal{F}_i^h,
\]

where

\[
\mathcal{F}_l^h = -\Phi S_l \nabla \rho_l^h, \quad \mathcal{F}_i^h = -\Phi S_i \nabla \rho_i^h
\]

3 Liquid Saturated/Unsaturated state; a general formulation

As recalled in the Introduction a traditional choice for the primary unknowns, in modeling two phase flow and transport process, is the saturation and one of the phases pressure, for example \( S_g \) and \( p_l \). But as seen above, in (19)–(22), saturation is no longer a consistent variable in saturated regions and this set of unknowns cannot describe the flow in a region where there is only one phase (see [36]). In this section, we recall and compare two possible choices of primary variables presented in [8] for circumvent this difficult problem.

3.1 Modeling based on the total hydrogen mass density, \( \rho^h_{\text{tot}} \)

To solve this problem, instead of using the gas saturation \( S_g \) we have proposed, in [8], to use \( \rho^h_{\text{tot}} \), the total hydrogen mass density, defined as:

\[
\rho^h_{\text{tot}} = S_l \rho_l^h + S_g \rho_i^h.
\]

Then, defining

\[
a(S_g) = C_h (1 - S_g) + C_g S_g \in [C_h, C_g];
\]

with

\[
\alpha'(S_g) = C_g - C_h > 0,
\]

since \( C_g > C_h \), from the assumption of weak solubility: we may rewrite the total hydrogen mass density, \( \rho^h_{\text{tot}} \), defined in (23), as:

\[
\rho^h_{\text{tot}} = \begin{cases} \alpha(S_g)(p_l + p_c(S_g)) & \text{if } S_g > 0 \\ \rho^h_0 & \text{if } S_g = 0. \end{cases}
\]

As noticed in the previous section in Remark 1, using the monotonicity of functions \( p_c(S_g) \) and \( a(S_g) \), we see a concentration threshold corresponding to \( C_h (p_l + p_c(0)) \) separating the liquid saturated zone, \( \rho^h_{\text{tot}} \leq C_h (p_l + p_c(0)) \), from the unsaturated zone, \( \rho^h_{\text{tot}} > C_h (p_l + p_c(0)) \).

With this choice of primary variables, \( \rho^h_{\text{tot}} \) and \( p_l \), the two systems of equations (14)–(18) and (19)–(22) reduce to a single system of equations:

\[
\frac{\partial \rho^h_{\text{tot}}}{\partial t} - \text{div} \left( \rho^h_{\text{tot}} \nabla \rho^h_{\text{tot}} \right) = \mathcal{F}_l^h,
\]

\[
\Phi \frac{\partial \rho^h_{\text{tot}}}{\partial t} + \text{div}(\rho^h_{\text{tot}} \nabla \rho^h_{\text{tot}}) = \mathcal{F}_l^h, - \text{div} \left( C_i p_g \nabla \rho^h_{\text{tot}} \right)
\]

\[
\Phi \frac{\partial \rho^h_{\text{tot}}}{\partial t} - \text{div} \left( C_l p_l \nabla \rho^h_{\text{tot}} \right)
\]

\[
- \text{div} \left( C_i p_g \nabla \rho^h_{\text{tot}} \right)
\]

\[
- \text{div} \left( \Phi S_l \nabla \rho^h_{\text{tot}} \right) = \mathcal{F}_l^h.
\]
If we want to study the mathematical properties of the operators in this system of equations, we should develop the above system of equations using first the dependency of the secondary variables \( S_g = S_g(p_l, \rho^h_{\text{tot}}), S_l = 1 - S_g = S_l(p_l, \rho^h_{\text{tot}}), \rho_l = \rho^h(p_l, \rho^h_{\text{tot}}) \), and secondly computing the derivatives of the saturations, from equation (26),

\[
\frac{\partial S_g}{\partial p_l} = -\frac{a(S_g)}{C_A \rho^h_{\text{tot}} + a(S_g)^2 \rho^h_{\text{tot}}}, \tag{29}
\]

\[
\frac{\partial S_l}{\partial p_l} = a(S_l) \frac{\rho^h_{\text{tot}}}{C_A \rho^h_{\text{tot}} + a(S_l)^2 \rho^h_{\text{tot}}}, \tag{30}
\]

where \( \mathbb{1}_{\{\rho^h_{\text{tot}} > C_h(p_l + p_g(0))\}} \) is the characteristic function of the set \( \{\rho^h_{\text{tot}} > C_h(p_l + p_g(0))\} \).

As noted in section 2.5 in [8], we have \( \partial S_g / \partial p_l \leq 0 \) and \( \partial S_l / \partial p_l > 0 \), when the gas phase is present. Then the system (14)–(15) can be written:

\[
-\Phi \rho^h_{\text{tot}} \frac{\partial S_g}{\partial p_l} \frac{\partial p_l}{\partial t} - \text{div} \left( \lambda_{1,1}(\rho_l, \rho^h_{\text{tot}}) \mathbb{1} + \lambda_{2,1} \rho^h_{\text{tot}} + B_1 \mathbb{I} \right) \mathbb{K} g = \mathbb{F}^w
\]

\[
\frac{\partial \rho^h_{\text{tot}}}{\partial t} - \text{div} \left( \lambda_{2,1}(\rho_l, \rho^h_{\text{tot}}) \rho^h_{\text{tot}} + B_2 \mathbb{K} g \right) = \mathbb{F}^h, \tag{32}
\]

where the coefficients are defined by:

\[
\lambda_{1,1}(\rho_l, \rho^h_{\text{tot}}) = \lambda_l(S_g) \rho^h_{\text{tot}} \mathbb{K} \mathbb{I} - \Phi(1 - S_g) DC_h \mathbb{N}^l, \tag{33}
\]

\[
\lambda_{2,1}(\rho_l, \rho^h_{\text{tot}}) = -\Phi(1 - S_g) \frac{1 - N}{a(S_g)} DC_h \mathbb{N}^l, \tag{34}
\]

\[
\lambda_{2,2}(\rho_l, \rho^h_{\text{tot}}) = \lambda_l(S_g) \rho^h_{\text{tot}} + \lambda_g(S_g) \mathbb{I} + \Phi(1 - S_g) DC_h \mathbb{N}^l, \tag{35}
\]

\[
\lambda_{2,2}(\rho_l, \rho^h_{\text{tot}}) = \lambda_l(S_g) \frac{1 - N}{a(S_g)} DC_h \mathbb{N}^l, \tag{36}
\]

\[
B_1(p_l, \rho^h_{\text{tot}}) = -\lambda_l(S_g) \rho^h_{\text{tot}} \mathbb{I} \left[ \rho^h_{\text{tot}} + \rho^h_l \mathbb{I} \right], \tag{37}
\]

\[
B_2(p_l, \rho^h_{\text{tot}}) = -\lambda_l(S_g) \left[ \rho^h_{\text{tot}} \mathbb{I} \left[ \rho^h_{\text{tot}} + \rho^h_l \mathbb{I} \right] + \lambda_g(S_g) \mathbb{I} \left[ \rho^h_{\text{tot}} + \rho^h_l \mathbb{I} \right] \right], \tag{38}
\]

with \( \mathbb{I} \) denoting the identity matrix and with the auxiliary functions

\[
N(p_l, \rho^h_{\text{tot}}) = \frac{C_A \rho^h_{\text{tot}}}{C_A \rho^h_{\text{tot}} + a(S_l)^2 \rho^h_{\text{tot}}}, \quad \mathbb{I} = \mathbb{1}_{\{\rho^h_{\text{tot}} > C_h(p_l + p_g(0))\}} \tag{39}
\]

\[
\rho^h_l(p_l, \rho^h_{\text{tot}}) = \min (C_h p_g(p_l, \rho^h_{\text{tot}}), \rho^h_{\text{tot}}), \tag{40}
\]

\[
p_g(p_l, \rho^h_{\text{tot}}) = p_l + p_c(S_g(p_l, \rho^h_{\text{tot}})), \tag{41}
\]

We should notice first that equation (32) is uniformly parabolic in the presence of capillarity and diffusion; but if capillarity and diffusion are neglected, this same equation becomes a pure hyperbolic transport equation (see sec. 2.6 in [8]). Then, if we sum equations (31) and (32) we obtain a uniformly parabolic/elliptic equation, which is parabolic in the unsaturated (two-phases) region and elliptic in the liquid saturated (one-phase) region.

Remark 2 Simulations presented in sec. 3.2 in [8] show that this last choice of primary variables, \( \rho^h_{\text{tot}}, \) and \( p_l, \) could easily handle phase transitions (appearance/disappearance of the gas phase, saturated zones, ...) in two-phase partially miscible flows. However, the discontinuity of the characteristic function with respect to the main variable \( p^h_{\text{tot}}, \) on the concentration threshold line, \( \mathbb{1}_{\{\rho^h_{\text{tot}} > C_h(p_l + p_g(0))\}} \) in (29), (30) and (39), have some effect on the conditioning of the Jacobian matrix and hence on the number of Newton iterations and the number of iterations required to solve the Jacobian system. Except if the fraction in front of the characteristic function in (39) tends to zero as \( S_g \to 0, \) which is the case when the van Genuchten's capillary curves are used.

An other variant is presented in [1], where using the total hydrogen concentration,

\[
C^h_{\text{tot}} = \frac{(1 - S_g) \rho^h_{\text{tot}} + S_g p_g}{(1 - S_g) p_l + S_g p_g}, \tag{42}
\]

an extended saturation can be defined from the inverse of (42):

\[
S_g = \frac{C^h_{\text{tot}} p_l - \rho^h_{\text{tot}}}{C^h_{\text{tot}} p_l - \rho^h_{\text{tot}} + (1 - C)p_g}. \tag{43}
\]

This saturation which was initially defined in the two-phase region is then extended outside this region by doing \( p_g = p_l \) in (42). Since, no matter in what region we are, there exists always an "extended" saturation \( S_g \leq 0, \) outside the two-phase region, which can be chosen as primary variable. It is then possible to model both the one-phase flow and the two-phase flow with the same system of equations written with this extended saturation as main unknown, and the gas appearance and disappearance is actually treated through the total hydrogen concentration \( C^h_{\text{tot}} \) expression (see [28]).

3.2 Modeling based on the dissolved hydrogen mass density in the liquid phase, \( \rho^h_l \)

We have seen that the variables \( p_l \) and \( \rho^h_{\text{tot}}, \) introduced in the last section, describe perfectly the flow system, both in the one-phase and in the two-phase regions, independently
of the presence of diffusion or capillary forces. But if we assume moreover that the effects of the capillary forces are not negligible we can choose an other set of primary variables.

Namely, using the retention curve (inverse of the capillary pressure curve), we may define the phase saturation as a function of the dissolved hydrogen mass density in the liquid, \( \rho^h \), and of the liquid pressure, \( p_l \); and hence use them as main unknowns. With these two variables, \( \rho^h \) and \( p_l \), the two systems (14)–(18) and (19)–(22) are transformed in a single system of equations able to describe both liquid saturated and unsaturated flow.

Since the capillary pressure curve \( S_g \rightarrow p_c(S_g) \) is a strictly increasing function we can define an inverse function (retention curve) \( f : \mathbb{R} \rightarrow [0,1] \), (see Fig. 2), by

\[
f(\pi) = \begin{cases} p_c^{-1}(\pi) & \text{if } \pi \geq p_c(0) \\ 0 & \text{otherwise.} \end{cases}
\]

(44)

By definition of the retention function \( f \), using (10) and (12), we have:

\[
f\left( \frac{\rho^h}{C_h} - p_l \right) = S_g,
\]

(45)

and it is then possible to compute the gas saturations, \( S_g \), from \( p_l \) and \( \rho^h \). These two variables being well defined in both the one and two-phase regimes, we will now use them as principal unknowns.

Equations (14)-(18) with unknowns \( p_l \) and \( \rho^h \) can be written as:

\[
- \Phi \rho^{ad}_w \frac{\partial}{\partial t} \left( f \left( \frac{\rho^h}{C_h} - p_l \right) \right)
- \text{div} \left( \bar{k}^{1.1} \nabla p_l + \bar{k}^{1.2} \nabla \rho^h + B_1 \nabla \xi \right) = \mathcal{F}^w,
\]

(46)

\[
\Phi \frac{\partial}{\partial t} (a^* f \left( \frac{\rho^h}{C_h} - p_l \right) \rho^h)
- \text{div} \left( \bar{k}^{2.1} \nabla p_l + \bar{k}^{2.2} \nabla \rho^h + B_2 \nabla \xi \right) = \mathcal{F}^h,
\]

(47)

where the coefficients are given by the following formulas:

\[
\bar{k}^{1.1} = \lambda_l(S_g) \rho^{ad}_w \xi, \quad \bar{k}^{1.2} = -\Phi (1 - S_g) D \xi,
\]

(48)

\[
\bar{k}^{2.1} = \lambda_l(S_g) \rho^{ad}_h \xi,
\]

(49)

\[
\bar{k}^{2.2} = \lambda_g(S_g) \frac{C_v}{C_h} \rho^h \xi + \Phi (1 - S_g) D \xi,
\]

(50)

with \( B_1 \) and \( B_2 \) defined as in (37), (38), and

\[
a^* (S_g) = a(S_g) C_h = 1 + \left( \frac{C_v}{C_h} - 1 \right) S_g.
\]

(51)

If we consider first, equation (47), we may write it as

\[
\Phi \left( a^* (S_g) + \rho^h \frac{\partial a^* (S_g)}{\partial p_l} \right) \frac{\partial \rho^h}{\partial t}
- \text{div} \left( \bar{k}^{2.1} \nabla p_l + \bar{k}^{2.2} \nabla \rho^h + B_2 \nabla \xi \right)
+ \Phi \rho^h \frac{\partial a^* (S_g)}{\partial p_l} \frac{\partial p_l}{\partial t} = \mathcal{F}^h.
\]

Moreover, from (51) and because \( f \) and \( f' \) are positive, we have

\[
a^* (S_g) + \rho^h \frac{\partial a^* (S_g)}{\partial p_l} = 1 + \left( \frac{C_v}{C_h} - 1 \right) \left( f \left( \frac{\rho^h}{C_h} - p_l \right) + \frac{\rho^h}{C_h} f' \left( \frac{\rho^h}{C_h} - p_l \right) \right) \geq 1;
\]

and if the diffusion is not neglected, we have definite positiveness of the quadratic form \( \bar{k}^{2.2} \xi \cdot \xi \), in equation (47); i.e. for any \( \xi \neq 0 \),

\[
\lambda_2(\xi) = \lambda_2(S_g) \frac{C_v}{C_h} \rho^h \xi \cdot \xi + \Phi (1 - S_g) D \xi^2 > 0,
\]

and therefore equation (47) is strictly parabolic in \( \rho^h \).

If we develop, equation (46) as follows:

\[
\Phi \rho^{ad}_w f' \left( \frac{\rho^h}{C_h} - p_l \right) \frac{\partial p_l}{\partial t}
- \text{div} \left( \bar{k}^{1.1} \nabla p_l + \bar{k}^{1.2} \nabla \rho^h + B_1 \nabla \xi \right)
- \lambda_1(S_g) \frac{\rho^{ad}_w \xi \cdot \xi}{C_h} \geq 0,
\]

and therefore positive of \( \bar{k}^{1.1} \xi \cdot \xi \) and of \( \bar{k}^{2.1} \xi \).

Moreover,

\[
\Phi \rho^{ad}_w f' \left( \frac{\rho^h}{C_h} - p_l \right) \frac{\partial p_l}{\partial t} \geq 0.
\]

However, equations in system (46)-(47) are not uniformly parabolic/elliptic for the pressure \( p_l \), because the coefficients, \( \bar{k}^{1.1} \), \( \bar{k}^{2.1} \), in front of \( \nabla p_l \) in (46)–(47) tend to zero as \( S_g \rightarrow 1 \).

**Remark 3** It is worth noticing that this system (46)-(47), with variables \( p_l \) and \( \rho^h \), has interesting properties for numerical simulations in strongly heterogeneous porous media. These two variables are continuous through interfaces separating different porous media with different rock types.
(different absolute permeability, different capillary and permeability curves), as we will see in 4.3; which is not the case for the total hydrogen density \( \rho_{tot} \). An other advantage is the continuity, in the neighbourhood of the concentration threshold line, of all the coefficients \( \kappa^{1/3} \), in (46)-(47) and of \( f \) in (47). However, the choice of total hydrogen mass density, \( \rho_{tot} \), for primary variable does not require capillary effects, making it useful when the capillary effects are negligible; which is not the case with the choice of the dissolved hydrogen density, \( \rho_h \), as primary variable, which is relying on an invertible capillary pressure curve (see equation (45)). Moreover, with this choice \( \rho_h \), as primary variable, steepest or infinite slope in the capillary curve or in the retention curve have effect on the conditioning of the Jacobian and make problem for computing back the secondary variables.

**Remark 4** From the solubility equation given by the Henry’s law (10), it is possible to define an “extended” gas pressure by \( \bar{p}_g = \rho_h / C_h \) even inside the liquid saturated region. Obviously, this “extended” gas pressure coincide with the true gas pressure \( p_g \) in the two-phase region. In [3] and [30], this “extended” gas pressure and the liquid pressure are chosen for primary variables and the gas appearance and disappearance is treated through the retention curve.

4 Numerical experiments

In this last section, we present four numerical tests specially designed for illustrating the ability of the model described by equations (46)-(47) to deal with gas phase appearance and disappearance. All the computations were done using the variables, \( p_l \) and \( \rho_h \), we are also displaying, for each test, the saturation and pressure level curves. These two last quantities are obtained after a post processing step using the Capillary Pressure law (3), equations (45), Henry’s law (10), and the constraints (4) and (12) (see Figure 1).

The first test focuses on the gas phase appearance produced by injecting pure hydrogen in a 2-D homogeneous porous domain \( \Omega \) (see Figure 3), which is initially liquid saturated by pure water. Because the main goal of all these numerical experiments is to test the model efficiency, for describing the phase appearance or disappearance, the porous domain geometry does not really matter and we will use a porous domain with a simple geometry. Consequently, we choose a simple, quasi-1D, porous domain (see Figure 4) for the following three tests.

The test case number 2 is more complex, it shows local disappearance of the gas phase created by injecting pure hydrogen in a homogeneous unsaturated porous medium (initially both phases, liquid and gas, are present everywhere).

The two last tests aim is to focus on the main challenges in simulating the flow crossing the engineered barriers, located around the waste packages. In the test case number 3, the porous medium domain is split in two parts with different and highly contrasted rock types, and like in the first one, the gas phase appearance is produced by injecting pure hydrogen in an initially water saturated porous domain. The test case number 4 addresses the evolution of the phases, from an initial phase disequilibrium to a stabilized stationary state, in a closed porous domain (no flux boundary conditions).

### Fluid Parameters: Phases and Components Characteristics

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \theta )</td>
<td>303</td>
</tr>
<tr>
<td>( D^0 )</td>
<td>3 ( 10^{-9} ) m(^2)/s</td>
</tr>
<tr>
<td>( \mu_l )</td>
<td>1 ( 10^{-3} ) Pa.s</td>
</tr>
<tr>
<td>( \mu_g )</td>
<td>9 ( 10^{-6} ) Pa.s</td>
</tr>
<tr>
<td>( H(\theta = 303K) )</td>
<td>7.65 ( 10^{-6} ) mol/(Pa.m(^3))</td>
</tr>
<tr>
<td>( M^w )</td>
<td>18 g/mol</td>
</tr>
<tr>
<td>( M_h )</td>
<td>2 ( 10^{-3} ) kg/mol</td>
</tr>
<tr>
<td>( \rho_{tot} )</td>
<td>10(^3) kg/m(^3)</td>
</tr>
</tbody>
</table>

**Table 1** Fluid parameters: phases and components characteristics.

In all these four test cases, for simplicity, the porous medium is assumed to be isotropic, such that \( K = kI \) with \( k \) a positive scalar; and the source terms are assumed to be null: \( \mathcal{F}_w = 0 \) and \( \mathcal{F}_h = 0 \). As usual in hydrogeology, the van Genuchten-Mualem model for the capillary pressure law and the relative permeability functions (see [20,27]) are used for underground nuclear waste modeling, i.e.:

\[
\begin{align*}
\rho_c &= p_f \left( S_{le}^{1/m} - 1 \right)^{1/n} \\
\lambda_l &= \frac{1}{\mu_l} \sqrt{S_{le} \left( 1 - (1 - S_{le}^{1/m})^m \right)^2} \\
\lambda_g &= \frac{1}{\mu_g} \sqrt{1 - S_{le}} \left( 1 - S_{le}^{1/m} \right)^{2m/n}
\end{align*}
\]

with \( S_{le} = \frac{S_f - S_{l,res}}{1 - S_{l,res} - S_{g,res}} \) and \( m = 1 - \frac{1}{n} \).

Note that in the van Genuchten-Mualem model, there is no Capillary pressure jump at 0, \( p_c(0) = 0 \), but the presence of a jump, like in the Brooks-Corey model (see [10]), would not lead to any difficulty, neither from the mathematical point

<table>
<thead>
<tr>
<th>Mesh size range</th>
<th>Time step range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Test number 1</td>
<td>2 m – 6 m (*)</td>
</tr>
<tr>
<td>Test number 2</td>
<td>1 m (*)</td>
</tr>
<tr>
<td>Test number 3</td>
<td>1 m (+)</td>
</tr>
<tr>
<td>Test number 4</td>
<td>2 ( 10^{-3}) m (+)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Mesh sizes and time steps used in the different Numerical Test</th>
<th>(*) Unstructured triangular mesh</th>
<th>(**) Regular quadrangular mesh</th>
</tr>
</thead>
</table>

**Table 2** Mesh sizes and time steps used in the different numerical tests.
The initial conditions, $p_l = p_{l, out}$ on $\Gamma_{out}$, $\rho_l^h = 0$ on $\Gamma_{out}$ are chosen, on $\Gamma_{out}$, such that $\rho_l^h > C_h p_l$, in order to keep the gas phase (according to the phase

We observe in the beginning (see time $t = 1200$ years in Figure 5) that all the injected hydrogen through $\Gamma_{in}$ is totally dissolved in the liquid phase, the gas saturation stay null on all the domain (there is no gas phase). During that same period of time: the increasing in liquid pressure is relatively small and the liquid phase flux originates slowly (they are both hard to see on the figures); then the hydrogen is transported mainly by diffusion of the dissolved hydrogen in the liquid phase.

Later on, the dissolved hydrogen accumulates around $\Gamma_{in}$ until the dissolved hydrogen mass density $\rho_l^h$ reaches the threshold $\rho_l^h = C_h p_l$ (according to Figure 1 and $p_c(0) = 0$ in 1), at time $t = 1600$ years, when the gas phase appears in the vicinity of $\Gamma_{in}$. Then this unsaturated region progressively expands. The gas phase volume expansion creates a gradient of the liquid pressure in the porous domain, causing the liquid phase to flow from $\Gamma_{in}$ to $\Gamma_{out}$. Consequently, after this time, $t = 1600$ years: the hydrogen is transported by convection in the gas phase and the dissolved hydrogen is transported by both convection and diffusion in the liquid phase. The liquid phase pressure increases globally in the whole domain until time $t = 260,000$ years (see Figure 5), and it starts to decrease in the whole domain until reaching a uniform and stationary state at $t = 10^6$ years, in which the water component flux is null everywhere.

### 4.1 Numerical Test number 1

The geometry of this test case is given in Figure 3; and the related data are given in Table 3. A constant flux of hydrogen is imposed on the input boundary, $\Gamma_{in}$, while Dirichlet conditions $p_l = p_{l, out}$, $\rho_l^h = 0$ are given on $\Gamma_{out}$ in order to have only the water component on this part of the boundary. The initial conditions, $p_l = p_{l, out}$ and $\rho_l^h = 0$, are uniform on all the domain, and correspond to a porous domain initially saturated with pure water.

The main steps of the corresponding simulation are presented in Figure 5.

---

**Table 3** Numerical Test case number 1: Boundary and Initial Conditions; porous medium characteristics and domain geometry; $\phi^w$ and $\phi^h$ are denoting respectively the water and hydrogen flux.

<table>
<thead>
<tr>
<th>Boundary conditions</th>
<th>Porous medium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial condition</td>
<td></td>
</tr>
<tr>
<td>$\phi^w \cdot v = 0$ on $\Gamma_{imp}$</td>
<td>$k$</td>
</tr>
<tr>
<td>$\phi^h \cdot v = 0$ on $\Gamma_{imp}$</td>
<td>$\Phi$</td>
</tr>
<tr>
<td>$\phi^w \cdot v = 0$ on $\Gamma_{in}$</td>
<td>$P_l$</td>
</tr>
<tr>
<td>$\phi^h \cdot v = 2 \Phi$ on $\Gamma_{in}$</td>
<td>$n$</td>
</tr>
<tr>
<td>$p_l = p_{l, out}$ on $\Gamma_{out}$</td>
<td>$S_{l, res}$</td>
</tr>
<tr>
<td>$\rho_l^h = 0$ on $\Gamma_{out}$</td>
<td>$S_{h, res}$</td>
</tr>
<tr>
<td>$p_l(t = 0) = P_{l, out}$ in $\Omega$</td>
<td>Others</td>
</tr>
<tr>
<td>$\rho_l^h(t = 0) = 0$ in $\Omega$</td>
<td></td>
</tr>
<tr>
<td>$p_{l, out} = 10^6$ Pa</td>
<td></td>
</tr>
</tbody>
</table>

We have the following symbols:

- $\Omega$: the whole domain.
- $\Gamma_{in}$: the input boundary; $\Gamma_{out}$: the output boundary.
- $\Gamma_{imp}$: interface between saturated and unsaturated regions.
- $\Gamma_{reg}$: interface between liquid and gas phases.
- $\phi^w$ and $\phi^h$: water and hydrogen flux.
- $k$: hydraulic conductivity.
- $\Phi$: potential.
- $P_l$: liquid phase pressure.
- $\rho_l^h$: dissolved hydrogen mass density in the liquid phase.
- $S_{l, res}$ and $S_{h, res}$: residual saturation of liquid and gas phases, respectively.

---

**Fig. 3** Test case number 1: Geometry a the 2-D porous domain, $\Omega$.

**Fig. 4** Test cases number 2, 3 and 4: Geometry of the quasi-1D porous domain, $\Omega = \Omega_1 \cup \Omega_2$. 

---

**Fig. 5** That all the injected hydrogen through $\Gamma_{in}$ is totally dissolved in the liquid phase, the gas saturation stay null on all the domain (there is no gas phase). During that same period of time: the increasing in liquid pressure is relatively small and the liquid phase flux originates slowly (they are both hard to see on the figures); then the hydrogen is transported mainly by diffusion of the dissolved hydrogen in the liquid phase.

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**Table 3** Numerical Test case number 1: Boundary and Initial Conditions; porous medium characteristics and domain geometry; $\phi^w$ and $\phi^h$ are denoting respectively the water and hydrogen flux.  

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**Table 3** Numerical Test case number 1: Boundary and Initial Conditions; porous medium characteristics and domain geometry; $\phi^w$ and $\phi^h$ are denoting respectively the water and hydrogen flux.  

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**Fig. 5** That all the injected hydrogen through $\Gamma_{in}$ is totally dissolved in the liquid phase, the gas saturation stay null on all the domain (there is no gas phase). During that same period of time: the increasing in liquid pressure is relatively small and the liquid phase flux originates slowly (they are both hard to see on the figures); then the hydrogen is transported mainly by diffusion of the dissolved hydrogen in the liquid phase.
The main steps of the corresponding simulation are presented in Figures 6 and 7 where we show the liquid pressure $p_l$, the dissolved hydrogen molar density $c_h^l$ (equal to $\rho_h^l / M_h$) and the gas saturation $S_g$ profiles, at different times. At the beginning, up to $t < 1400$ years, the two phases are present in the whole domain (see time $t = 500$ years on Figure 6). The permanent injection of hydrogen increases the gas saturation in the vicinity of $\Gamma_m$. The local gas saturation drop is due to the difference in relative mobilities $\lambda_{\alpha}(S_{\alpha})$ between the two phases: the lower liquid mobility leads to a bigger liquid pressure increase, compared to the gas pressure increase; which is finally producing a capillary pressure drop (according to definitions (1) and (3); see Figure 2), and creating a liquid saturated zone. At time $t = 1400$ years, the gas phase starts to disappear in some region of the porous domain (see time $t = 1500$ years, in Figure 7).
Porous medium

<table>
<thead>
<tr>
<th>Boundary conditions</th>
<th>Porous medium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial condition</td>
<td>Value</td>
</tr>
<tr>
<td>( \phi^w \cdot v = 0 ) on ( \Gamma_{imp} )</td>
<td>( k ) 5 ( 10^{-20} ) m²</td>
</tr>
<tr>
<td>( \phi^w \cdot v = 0 ) on ( \Gamma_{imp} )</td>
<td>( \Phi ) 0.15</td>
</tr>
<tr>
<td>( \phi^h \cdot v = 0 ) on ( \Gamma_{imp} )</td>
<td>( n ) 1.49</td>
</tr>
<tr>
<td>( p_1 = p_{l,\text{out}} ) on ( \Gamma_{out} )</td>
<td>( S_{l,\text{res}} ) 0.4</td>
</tr>
<tr>
<td>( p_{h,\text{out}} ) on ( \Gamma_{out} )</td>
<td>( S_{g,\text{res}} ) 0.15</td>
</tr>
<tr>
<td>( p_1(t = 0) = p_{l,\text{out}} ) in ( \Omega )</td>
<td>( p_{h,\text{out}}(t = 0) ) in ( \Omega )</td>
</tr>
<tr>
<td>( p_{l,\text{out}} = 10^6 ) Pa</td>
<td>( L_4 ) 200 m</td>
</tr>
<tr>
<td>( p_{g,\text{out}} = 1.1 \times 10^5 ) Pa</td>
<td>( L_5 ) 20 m</td>
</tr>
<tr>
<td>( \phi^w \cdot v = 0 ) on ( \Gamma_{imp} )</td>
<td>( \Phi ) 0.15</td>
</tr>
<tr>
<td>( \phi^h \cdot v = 0 ) on ( \Gamma_{imp} )</td>
<td>( n ) 1.49</td>
</tr>
<tr>
<td>( p_1 = p_{l,\text{out}} ) on ( \Gamma_{out} )</td>
<td>( S_{l,\text{res}} ) 0.4</td>
</tr>
<tr>
<td>( p_{h,\text{out}} ) on ( \Gamma_{out} )</td>
<td>( S_{g,\text{res}} ) 0.15</td>
</tr>
<tr>
<td>( p_1(t = 0) = p_{l,\text{out}} ) in ( \Omega )</td>
<td>( p_{h,\text{out}}(t = 0) ) in ( \Omega )</td>
</tr>
<tr>
<td>( p_{l,\text{out}} = 10^6 ) Pa</td>
<td>( L_4 ) 200 m</td>
</tr>
<tr>
<td>( p_{g,\text{out}} = 1.1 \times 10^5 ) Pa</td>
<td>( L_5 ) 20 m</td>
</tr>
</tbody>
</table>

Table 4 Numerical Test case number 2: Boundary and Initial Conditions; porous medium characteristics and domain geometry. \( \phi^w \) and \( \phi^h \) are denoting respectively the water and hydrogen flux.

Then, a saturated liquid region (\( S_g = 0 \)) will exist until time \( t = 17 \times 10^3 \) years (see Figure 6); and during this period of time, the saturated region is pushed by the injected Hydrogen, from \( \Gamma_{in} \) to \( \Gamma_{out} \).

After the time \( t = 17 \times 10^3 \) years, due to the Dirichlet conditions imposed on \( \Gamma_{out} \), the liquid saturated region disappears and all together the phases pressure and the gas saturation are growing in the whole domain (see the time \( t = 20 \times 10^3 \) years in Figure 7).

Finally the liquid pressure reaches its maximum at time \( t = 20 \times 10^3 \) years and then decreases in the whole domain (see the Figure 7). This is caused, like in the numerical test case number 1, by the evolution of the system towards a stationary state which is characterized by a zero water component flow.

4.3 Numerical Test number 3

The geometry and the data of this numerical test are given in Figure 4, Table 5 and Table 6. Like in the Numerical Test number 2, a constant flux of hydrogen is imposed on the input boundary, \( \Gamma_{in} \), while Dirichlet conditions \( p_1 = p_{l,\text{out}} \), \( p_{h,\text{out}} = 0 \) are given on \( \Gamma_{out} \), in order to have only the liquid phase on this part of the boundary. The initial conditions, \( p_1 = p_{l,\text{out}} \) and \( p_{h,\text{out}} = 0 \), are uniform on all the domain, and correspond to a porous domain initially saturated with pure water. Contrary to the two first numerical tests, the porous domain is non homogeneous, there are two different porous subdomains \( \Omega_1 \) and \( \Omega_2 \); \( L_4 = 200 \) m, \( L_1 = 20 \) m and \( L_2 = 180 \) m.

The simulation time of this test case is \( T = 10^6 \) years; the discretization space mesh is 1 meter; the time step is \( 10^2 \) years at the beginning and grows up to \( 2 \cdot 10^4 \) years in the end of the simulation (see Table 2).

Figures 9 and 10 represent the liquid pressure \( p_1 \), the dissolved hydrogen molar density (equal to \( p_1^h / \Phi^h \)) and the gas saturation \( S_g \) profiles at different times.

The main difference from the previous simulations (which were in a homogeneous porous domain) is the gas saturation discontinuity, staying on the porous domain interface \( x = 20 \) m. Due to the height of this saturation jump, we had to use a logarithm scale for presenting the gas saturation \( S_g \) profiles; but as a consequence although all the \( S_g \) curves go to zero this cannot be seen with a logarithmic scale in Figures 9 and 10.

There are four main steps:

– From 0 to \( 3.8 \cdot 10^4 \) years the increasing of both the gas saturation and the liquid pressure are small and slow in the whole domain and hard to see on the figures; while the hydrogen injection on the left side \( \Gamma_{in} \) of the domain increases notably the hydrogen density level.
From $3.8 \cdot 10^4$ to $5.4 \cdot 10^4$ years both the liquid pressure and the hydrogen density are increasing in the whole domain. The gas start to expanding from the left side of the domain $I_m$. The saturation front is moved towards the porous media discontinuity, at $x = 20 \text{ m}$, which is reached at $t = 5.4 \cdot 10^4$ years; see Figures 9.

From $5.4 \cdot 10^4$ years to $1.3 \cdot 10^5$ years, see Figures 10, the saturation front has crossed the medium discontinuity at $x = 20 \text{ m}$ and, from now, all the saturation profiles will have a discontinuity at $x = 20 \text{ m}$.

From $1.3 \cdot 10^5$ years to $10^6$ years, see Figures 10, both the hydrogen density and the gas saturation keep growing while the liquid pressure decreases towards zero on the entire domain. The gas saturation front keeps moving to the right, pushed by the injected gas, up to $x \approx 150 \text{ m}$ at $10^6$ years.

Until the saturation front reaches the interface between the two porous media, for ($t = 5.4 \cdot 10^4$ years), appearance and evolution of both the gas phase and the unsaturated zone are identical to what was happening in the test case 1 (with a homogeneous porous domain) during the period of gas injection: the dissolved hydrogen is accumulating at the entrance until the liquid phase becomes saturated, at time ($t > 3.8 \cdot 10^4$ years), letting the gas phase to appear.

When the saturation front crosses the interface between the two porous subdomains (at $x = 20 \text{ m}$ and $t = 5.4 \cdot 10^4$ years), the gas saturation is strictly positive on both sides of this interface and the capillary pressure curves is different on each side (see Table 6). The capillary pressure continuity at the interface imposes to $p_c^{(1)}$, the Capillary Pressure in $\Omega_1$, and to $p_c^{(2)}$, the Capillary Pressure in $\Omega_2$, to be equal on this interface. Then $p_c^{(1)} = p_c^{(2)}$ is satisfied only if there are two different saturations, on each interface side $S_g^{(1)}$, and $S_g^{(2)}$: $p_c^{(1)}(S_g^{(1)}) = p_c^{(2)}(S_g^{(2)})$ ; see Figure 8.

In the same way as in the numerical test case number 1, the system tends to a stationary state.
4.4 Numerical Test number 4

This last numerical test is different from all the precedent ones; it intends to be a simplified representation of what happens when an unsaturated porous block is placed within a water saturated porous structure. The challenge is then: how the mechanical balance will be restored in a homogeneous porous domain, which was initially out of equilibrium, i.e. with a jump in the initial phase pressures?

The initial liquid pressure is the same in the entire porous domain: \( \Omega \), \( p_{l,1} = p_{l,2} \), and in the subdomain \( \Omega_1 \) the initial condition, \( p_{l,1} = p_{g,1} \) in Table 7, corresponds to a liquid fully saturated state with a hydrogen concentration reaching the gas appearance concentration threshold \( p_g = p_l \) and \( \rho_h = C_h p_g \) (Figure 1). In the subdomain \( \Omega_2 \) the initial condition \( (p_{l,2} \neq p_{g,2} \text{ and } p_{g,2} \neq p_{g,1}) \) corresponds to a non saturated state (see Table 7).

The porous block initial state is said out of equilibrium, because: if this initial state was in equilibrium, in the two subdomains \( \Omega_1 \) and \( \Omega_2 \), the local mechanical balance would have made the pressures, of both the liquid and the gas phase, continuous in the entire domain \( \Omega \).

For simplicity, we assume the porous medium domain \( \Omega \) is homogeneous and all the porous medium characteristics are the same in the two subdomains \( \Omega_1 \) and \( \Omega_2 \), and corresponding to concrete. The system is then expected to evolve from this initial out of equilibrium state towards a stationary state.

We should notice that, in order to see appearing the final stationary state, in a reasonable period of time, we have shortened the domain \( \Omega \) (\( L_x = 1 \)m), taken the porous media characteristics, and set the final time of this simulation \( T_f \) in at \( T_f = 10^6 \text{ s} \approx 11.6 \text{ days} \). The complete set of data of this test case is given in Table 7.

The space discretization step was taken constant equal to \( 2 \cdot 10^{-3} \text{ m} \) and the time step was variable, going from 0.33 s in the beginning of the simulation to \( 16.7 \cdot 10^{3} \text{ s} \) at the end of the simulation (see Table 2). Figures 11 and 12 represent the liquid pressure \( p_l \), the dissolved hydrogen molar density \( c_h \) and the gas saturation \( S_g \).
concentration $c_h^l$ and the gas saturation $S_g$ profiles at different times.

There are essentially two steps:

- For $0 < t < 1.92 \cdot 10^5$ s (see Figure 11), the initial gas saturation jump moves from $x = 0.5$ m, at $t = 0$ and reaches $T_{in}$, the left domain boundary, at $t = 1.92 \cdot 10^5$ s. During this movement, the saturation jump height (initially $\approx 0.16$) decreases, until approximately 0.03, when it reaches the left boundary $T_{in}$. In front of this discontinuity there is a liquid saturated zone, $S_g = 0$, and in this zone both the liquid pressure and the hydrogen density are spatially uniform (see Figure 11, top). But, while the hydrogen density remains constant and equal to its initial value, the liquid pressure becomes immediately continuous and starts growing quickly (for instance, $p_l(t = 10^3$ s) $\approx 1.6 \cdot 10^6$ Pa), and then more slowly until $t = 1.3 \cdot 10^5$ s, when it starts to slightly decrease.

In Figure 11, located on the gas saturation discontinuity, there are both a high contrast in the dissolved hydrogen concentration (this concentration stays however continuous, but with a strong gradient, as seen in the top right of Figure 11), and a discontinuity in the liquid pressure gradient (see the top left of Figure 11).
For $1.92 \cdot 10^5 \text{s} < t < 10^6 \text{s} = T_{\text{fin}}$ (see Figure 12), all the entire domain is now unsaturated ($S_g > 0$). The liquid pressure, the hydrogen density and the gas saturation profiles are all strictly monotonous and continuous, going towards a spatially uniform distribution, corresponding to the stationary state (see Figure 12).

As expected, the system initially out of equilibrium (discontinuity of the gas pressure), comes back immediately to equilibrium (the gas pressure is continuous) and evolves towards a uniform stationary state (due to the no mass inflow and outflow boundary conditions). Although the liquid pressure and the dissolved hydrogen density are immediately again continuous for $t > 0$, the hydrogen density still have a locally very strong gradient until $t = 1.92 \cdot 10^5 \text{s}$.

At first, and at the very beginning ($\approx 10^2 \text{s}$), see top left of Figure 11, only the liquid pressure evolves in the liquid saturated zone. Due to a gas pressure in the unsaturated zone higher than in the liquid saturated zone ($S_g = 0$; $p_g = 2.5 \text{MPa} > p_l = 1 \text{MPa}$, for the initial state in Table 7), and due to the no flow condition imposed on $\Gamma_{\text{in}}$, the liquid in the saturated zone is compressed by the gas from the unsaturated zone. Then, a liquid gradient pressure appears around the saturation front and makes the liquid to flow from the liquid saturated zone towards the unsaturated one, and then the gas saturation front to move in the opposite direction.

The very strong hydrogen density gradient (until $t = 1.92 \cdot 10^5 \text{s}$), located on the saturation front, is due to the competition between the diffusion and the convective flux of the dissolved hydrogen around the saturation front: the water flow convecting the dissolved hydrogen, from left to right, cancels the smoothing effect of the gas diffusion propagation in the opposite direction. On the one hand the diffusion is supposed to reduce the hydrogen concentration contrast, by creating a flux going from strong concentrations (in the unsaturated zone) towards the low concentrations (in the liquid saturated zone), and on the other hand the flow of the liquid phase goes in the opposite direction (left to right, from $S_g = 0$ to $S_g > 0$). Once the disequilibrium has disappeared, the system tends to reach a uniform stationary state determined by the mass conservation of each component present...
in the initial state (the system is isolated, with no flow on any of the boundaries).

5 Concluding remarks

From balance equations, constitutive relations and equations of state, assuming thermodynamical equilibrium, we have derived a model for describing underground gas migration in water saturated or unsaturated porous media, including diffusion of components in phases and capillary effects. In the second part, we have presented a group of numerical test cases synthesizing the main challenges concerning the gas migration in a deep geological repository. These numerical simulations, are based on simplified but typical situations in underground nuclear waste management; they show evidence of the model ability to describe the gas (hydrogen) migration, and to treat the difficult problem of correctly following the saturated and unsaturated regions created by the gas generation. The selection of primary variables depends in general on the ability to chose a set of variables to represent the system state in saturated as well as in unsaturated regime. The optimal selection of primary variables depends, in general, on the particular problem being simulated. In some circumstances, the selection of primary variables has a large effect on the conditioning of the Jacobian matrix and hence on the number of Newton iterations and the number of iterations required to solve the Jacobian system. The simplicity in evaluating other secondary variables may have also large impact on efficiency of the chosen set of primary variables, and may require further solving of nonlinear algebraic equation as for the method in Section 3.1. In heterogeneous media it would be preferable to chose the primary variables which are continuous through material interfaces (as a phase pressures, and dissolved hydrogen mass $\rho_h$ as in Section 3.2), than variables like saturations or total hydrogen mass which are discontinuous through the interface. We have shown in the numerical tests that the primary variables chosen in Section 3.2 can efficiently treat the gas appearance/disappearance in presence of heterogeneities; although, the formulation based on total energy states the overall mass and energy conservation, which is consistent with the initial state and the system dynamics.
hydrogen mass is more suitable when the capillary effects are negligible.

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