

# Modeling Compositional Compressible Two-Phase Flow in Porous Media by the Concept of the Global Pressure

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## Abstract

We derive a new formulation for the compositional compressible two-phase flow in heterogeneous porous media. We consider a liquid-gas system with two components: water and hydrogen. The formulation considers gravity, capillary effects and diffusivity of each component. The main feature of this formulation is the introduction of the global pressure variable that partially decouples the system equations. To formulate the final system, and in order to avoid primary unknowns changing between saturated and unsaturated zones, an another new variable is introduced: the total hydrogen mass density. The derived system is written in terms of the global pressure and the total hydrogen mass density. The system is capable of modeling the flows in both saturated and unsaturated zones with no changes of the primary unknowns. The mathematical structure is well defined: the system consists of two nonlinear parabolic equations, the global pressure equation and the total hydrogen mass density equation. The derived formulation is fully equivalent to the original equations. Numerical computation of the coefficients introduced in the system of this new formulation are presented.

## 1 Introduction

The modeling of multiphase flow in porous formations is important for both the management of petroleum reservoirs and environmental remediation. More recently, modeling multiphase flow received an increasing attention in connection with the disposal of radioactive waste and  $CO_2$  storage in geological formations.

Multiphase flows in porous media can be modeled by mass balance laws for each of the fluids. The fractional flow formulation can be obtained by summation of these equations and introduction of the total flow and the new coefficients. In order to make the equations'

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coupling less strong, an artificial variable called global pressure can be introduced. This way, the global pressure–saturation fractional flow formulation is obtained. The notion of the global pressure was first introduced by [1, 8] for immiscible incompressible two-phase flow and was then revisited by other authors, see for instance [9]. It has been since used in a wide range of engineering specialties related to numerical simulation in hydrology and petroleum reservoir engineering, see for instance [10] and references therein. It has been proven that this fractional flow approach is far more efficient than the original two-pressure approach from the computational point of view [10]. This formulation was generalized, under the approximation that the volume factors be evaluated at the new global pressure instead of the corresponding phase pressure, to compressible two- and three-phase flows in [8]. This may be the case for models with small capillary pressures. These assumptions have limited the use of the global pressure formulation in numerical simulations codes for compressible flows. However, comparison with other formulations [10] show the computational effectiveness of the global pressure when it can be put to work, which may explain the current revival of interest for the concept of global pressure for numerical modeling of multiphase flow in porous media.

Most recently, the concept of the global pressure was generalized to the study of immiscible, compressible two-phase flow in porous media, without any exchange between the phases, taking into account gravity, capillary effects, and heterogeneity. The derived formulation is more suitable for mathematical and numerical analysis, for more details see [4]. The accuracy and effectiveness of the new formulation is demonstrated through numerical results [5]. Let us also mention that a fully equivalent global pressure formulation for three-phases compressible flows was recently established in [7] and validated by numerical results in [11].

The choice of the formulation has a significant impact on the numerical methods to be used. Application of certain numerical method for multiphase flow problems requires careful investigation of the implications of the chosen formulation. In other words, it is certainly worthwhile to revisit the form of the governing equations and the implications coming from it.

Compositional compressible two-phase flows in porous medium are usually modeled by the mass balance law written for each component and the Darcy-Muscat law for the conservation of momentum. Repartition of the components between the phases is given by local thermodynamical equilibrium hypothesis and governed by equality of component chemical potentials in both phases. The obtained equations represent a set of highly coupled nonlinear partial differential equations. In order to model both saturated and unsaturated zones, a common technique is to change the main unknowns of the system. In the saturated zones, pressure and saturation of one of the phases are commonly chosen as the main unknowns, whereas in the unsaturated zones pressure of the present phase can be chosen and the saturation may be replaced by mass concentration of one of the components in its phase. 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. It is well known that several problems arise due to Newton-Raphson’s flash calculations and the phase appearance and disappearance.

The numerical modeling and analysis of compositional two-phase flow in porous media has been a problem of interest for many years and many methods have been developed. There is an extensive literature on this subject. We will not attempt a literature review here, but merely mention a few references. We refer to the books [8, 10, 15] and the references therein. A recent review of the methods developed can be viewed in [6, 12, 14, 16] and the references

therein.

The aim of this paper is to derive a new fully equivalent fractional flow formulation for compositional compressible two-phase flow in porous media which can be efficiently solved numerically. We consider a liquid-gas system with two components such as immiscible gas injection in oil reservoirs or gas migration through engineered and geological barriers in a deep repository for radioactive waste and  $CO_2$  storage in geological formations. The formulation considers gravity, capillary effects and diffusivity of each component. The main feature of this formulation is the introduction of a new variable called the global pressure. The derived system is written in terms of the global pressure and the total gas mass density that partially decouples the equations and is able to model the flows both in the saturated and unsaturated zones with no changes of the primary unknowns and can be used to determine the composition of the phases in all zones. Both variables are well defined in saturated and unsaturated zones. The mathematical structure is well defined: the system consists of two nonlinear degenerate parabolic equations. The derived formulation is fully equivalent to the original equations and is more suitable for mathematical and numerical analysis. For simplicity of presentation, we derive the new formulation for a water-hydrogen two-phase flow, which gives us simple analytical expressions for the coefficients of the partial differential equations of the system.

The rest of the paper is organized as follows. In the second section we present the basic concepts, laws and governing equations for the two-phase partially miscible compressible flow, with liquid and gas phases. In the third section we generalize the concept of the global pressure to this model, which uses the global pressure as main pressure unknown. In the unsaturated zones the saturation is replaced by the mass concentration of the corresponding component. To avoid these change of the main unknowns, in the fourth section following the idea presented in [6], we introduce a new variable which represent the total hydrogen mass density. The system is then rewritten in the terms of the global pressure and the total hydrogen mass density. The obtained system has well defined mathematical structure and is well suited for the numerical treatment of this model which is our future research. In section 5, we present some numerical computation of the coefficients that appear in the system of PDEs of the new formulation. Additional conclusions are drawn in the last section. In Appendix A, we detail some calculations from section 4.

## 2 Governing system modeling two-phase compositional flow

### 2.1 Basic notation and physical laws

We consider two-phase immiscible compressible flow, liquid and gas phases, in a porous medium under isothermal conditions. For each phase  $\alpha \in \{l, g\}$  we denote by  $p_\alpha$  the corresponding phase pressure, by  $S_\alpha$  the saturation, and by  $\rho_\alpha$  the mass density. Here, the subscript  $l$  denotes the liquid phase and  $g$  denotes the gas phase. For each phase  $\mu_\alpha$  denotes the  $\alpha$ -phase viscosity.

Phase saturations satisfy

$$S_l + S_g = 1, \tag{1}$$

and they follow the capillary pressure law

$$p_c(S_g) = p_g - p_l, \tag{2}$$

where  $p_c(S_g)$  is the capillary pressure function, which can be given by different models, for example by van Genuchten, Brooks and Corey and other models, see for instance [15].

The two fluid phases contain two chemical components: water and hydrogen. Therefore, the liquid phase can contain water and dissolved hydrogen, and gas phase could consist of hydrogen and vaporized water. We will use the superscript  $i \in \{h, w\}$  to denote the quantities which describe certain property related to the component. For example,  $p_\alpha^i$  denotes the partial pressure of the  $i$ -component in the  $\alpha$ -phase. Composition of the two fluid phases will be described by components mass concentrations in the  $\alpha$ -phase, denoted by  $\rho_\alpha^i$  for  $i \in \{h, w\}$  and  $\alpha \in \{g, l\}$ . Each phase mass density,  $\rho_\alpha$  for  $\alpha \in \{l, g\}$ , can be expressed as

$$\rho_l = \rho_l^w + \rho_l^h \quad \rho_g = \rho_g^w + \rho_g^h. \quad (3)$$

Partial pressures of the gas phase satisfy Dalton's law,

$$p_g = p_g^w + p_g^h, \quad (4)$$

and we assume validity of the ideal gas law,

$$\rho_g^w = C_g^w p_g^w, \quad \rho_g^h = C_g^h p_g^h. \quad (5)$$

Here, we have introduced the constants

$$C_g^w = \frac{M^w}{RT}, \quad C_g^h = \frac{M^h}{RT},$$

where  $T$  is the fluid temperature,  $R$  is the universal gas constant and  $M^w, M^h$  are the water and the hydrogen molar masses.

In the following we assume that the local equilibrium thermodynamic behavior of the fluid system can be described by simplified analytical laws, namely the ideal gas law, Henry's and Raoult's laws. Our main interest in these simplified analytical expressions is to make more explicit the analysis of the transformed system of partial differential equations which will be derived by the introduction of new independent variables. We refer for instance to [17] for an analysis of the range of applicability of these laws and a comparison to a real mixture.

The hydrogen mass concentration in the liquid phase is given by Henry's law,

$$\rho_l^h = C_l^h p_g^h, \quad (6)$$

where  $C_l^h = H(T)M^h$ , and  $H(T)$  is Henry's law constant, depending only on the temperature.

The water vapor partial pressure is given by Raoult's law,

$$p_g^w = \hat{p}_g^w(T) \frac{\rho_l^w}{\rho_l^w + (M^w/M^h)\rho_l^h}, \quad (7)$$

where  $\hat{p}_g^w(T)$  is the vapor pressure of pure water, depending only on the temperature. Henry's and Raoult's laws are valid only in the saturated regions, i.e. in the regions where  $S_g \neq 0, 1$ , where the thermodynamical equilibrium applies.

To close the system we need one more equation for the water phase state. In order to simplify further the calculations we use a very simple model for the liquid water density by assuming that the water density in the liquid phase is constant:

$$\rho_l^w = \rho_l^{std} = cte. \quad (8)$$

Note that the dependency of the liquid density on the liquid pressure would not introduce any real difficulty, but it would complicate the calculations. The equations (3), (4), (5), (6), (7) and (8) completely describe the thermodynamical system.

Let us also emphasize that the mass densities and the concentrations depend on the partial gas pressures of the two component. We have:

$$\rho_l = \rho_l^{std} + \rho_l^h(p_g^h) = \rho_l(p_g^h), \quad \rho_g = \rho_g^h(p_g^h) + \rho_g^w(p_g^w) = \rho_g(p_g^h, p_g^w).$$

When both phases are present, we can express  $p_g^h$  (and  $p_g^w$ ) from (7) as a function of  $p_g$  by applying Henry's and Dalton's law. The partial pressure  $p_g^h$  is defined as a function of the gas pressure  $p_g$  by the equation:

$$p_g^h + \hat{p}_g^w(T) \frac{\rho_l^{std}}{\rho_l^{std} + \frac{M^w}{M^h} C_l^h p_g^h} - p_g = 0, \quad (9)$$

which can be solved explicitly, under the condition<sup>1</sup>

$$c = \frac{\hat{p}_g^w(T) M^w C_l^h}{\rho_l^{std} M^h} < \frac{1}{4}, \quad (10)$$

which is obviously satisfied for fluid systems like water and hydrogen. Let us denote this function, defined by (9), as  $\bar{p}_g^h(p_g)$  and note that, under the condition (10), it is a well defined function for all positive values of its argument. It satisfies

$$\frac{d\bar{p}_g^h}{dp_g}(p_g) \geq 1. \quad (11)$$

Therefore, we conclude that both fluid phase densities can be expressed as functions of the gas pressure.<sup>2</sup>

The function  $\bar{p}_g^h$  is useful in writing analytical conditions for appearance and disappearance of phases. For example, at certain liquid pressure  $p_l$  the gas phase cannot appear if a quantity of dissolved hydrogen is not strong enough to produce the gas pressure larger of the medium entry pressure. That is, while it holds

$$\begin{aligned} \rho_l^h / C_l^h + \hat{p}_g^w(T) \frac{\rho_l^{std}}{\rho_l^{std} + (M^w / M^h) \rho_l^h} < p_l + p_c(0) = \bar{p}_g^h(p_l + p_c(0)) \\ + \hat{p}_g^w(T) \frac{\rho_l^{std}}{\rho_l^{std} + \frac{M^w}{M^h} C_l^h \bar{p}_g^h(p_l + p_c(0))} \end{aligned} \quad (12)$$

where the second equality comes from the definition (9) of the function  $\bar{p}_g^h$ . From (10) it follows that the function

$$x \mapsto x + \hat{p}_g^w(T) \frac{\rho_l^{std}}{\rho_l^{std} + (M^w / M^h) C_l^h x}$$

is monotone increasing and therefore (12) is equivalent to

$$\rho_l^h < C_l^h \bar{p}_g^h(p_l + p_c(0)). \quad (13)$$

<sup>1</sup>For hydrogen at 25 °C, with  $\rho_l^{std} = 1000 \text{ kg/m}^3$ , we get  $c = 4.44 \cdot 10^{-7}$ .

<sup>2</sup> Under standard conditions the function  $p_g \mapsto \bar{p}_g^h(p_g)$  is practically identical to  $p_g - \hat{p}_g^w(T)$ .

When the value of  $\bar{p}_g^h(p_l + p_c(0))$  is zero or negative the gas phase will appear even when there is no hydrogen.

The liquid phase cannot exist at certain gas pressure  $p_g$  if the vapor partial pressure is lower than the equilibrium one:

$$p_g - p_g^h < \hat{p}_g^w(T) \frac{\rho_l^{std}}{\rho_l^{std} + \frac{M^w}{M^h} C_l^h \bar{p}_g^h(p_g)}.$$

After multiplying by  $C_g^h$  this can be written as

$$\rho_g^h > C_g^h p_g - C_g^h \hat{p}_g^w(T) \frac{\rho_l^{std}}{\rho_l^{std} + \frac{M^w}{M^h} C_l^h \bar{p}_g^h(p_g)} = C_g^h \bar{p}_g^h(p_g),$$

where in the last step we have used the definition of the function  $\bar{p}_g^h$ . So, the gas saturated media at the pressure  $p_g$  is possible if and only if

$$\rho_g^h > C_g^h \bar{p}_g^h(p_g). \quad (14)$$

## 2.2 Governing equations

Governing equations are obtained from the mass balance law, written for each component. Following the discussion described in [6], for each component, the conservation of the mass is written as follows

$$\Phi \frac{\partial}{\partial t} (S_l \rho_l^{std} + S_g \rho_g^w) + \text{div}(\rho_l^{std} \mathbf{q}_l + \rho_g^w \mathbf{q}_g + \mathbf{j}_l^w + \mathbf{j}_g^w) = \mathcal{F}^w \quad (15)$$

$$\Phi \frac{\partial}{\partial t} (S_l \rho_l^h + S_g \rho_g^h) + \text{div}(\rho_l^h \mathbf{q}_l + \rho_g^h \mathbf{q}_g + \mathbf{j}_l^h + \mathbf{j}_g^h) = \mathcal{F}^h, \quad (16)$$

where  $\Phi$  is the porosity,  $\mathbb{K}$  is the absolute permeability tensor and  $\mathbf{q}_l$  and  $\mathbf{q}_g$  are the phase volumetric velocities given by the Darcy-Muskat law as follows

$$\mathbf{q}_l = -\mathbb{K} \lambda_l(S_g) (\nabla p_l - \rho_l \mathbf{g}) \quad (17)$$

$$\mathbf{q}_g = -\mathbb{K} \lambda_g(S_g) (\nabla p_g - \rho_g \mathbf{g}). \quad (18)$$

Here,  $\lambda_\alpha$  is the  $\alpha$ -phase mobility,  $\mathbf{g}$  is the gravitational, downward-pointing, constant vector and  $\mathcal{F}^i$ ,  $i \in \{w, h\}$  are the source terms.

For  $i \in \{w, h\}$ ,  $\alpha \in \{l, g\}$ , the vectors  $\mathbf{j}_\alpha^i$  represent diffusive mass fluxes of the component  $i$  in the phase  $\alpha$ . They are given by

$$\mathbf{j}_\alpha^i = -\Phi S_\alpha \rho_\alpha D_\alpha^i \nabla c_\alpha^i \quad i \in \{w, h\}, \alpha \in \{l, g\}, \quad (19)$$

where  $c_\alpha^i = \rho_\alpha^i / \rho_\alpha$  are the mass fractions of the  $i$ -th component in the phase  $\alpha$  and  $D_\alpha^i$  (unit  $L^2/T$ ) are the molecular diffusion coefficients of the component  $i$  in the phase  $\alpha \in \{l, g\}$ . In the two-component system we assume that the diffusive fluxes satisfy

$$\mathbf{j}_\alpha^h + \mathbf{j}_\alpha^w = 0, \quad \alpha \in \{l, g\}, \quad (20)$$

or in other words  $D_\alpha^h = D_\alpha^w$ .

By introducing a new notation

$$\mathbf{J}_l := \mathbf{j}_l^h \quad \mathbf{J}_g := \mathbf{j}_g^h,$$

we can write

$$\mathbf{J}_l = -\Phi S_l \frac{\rho_l^{std}}{\rho_l} D_l^h \nabla \rho_l^h \quad (21)$$

$$\mathbf{J}_g = -\Phi S_g \frac{1}{\rho_g} D_g^h (\rho_g^w \nabla \rho_g^h - \rho_g^h \nabla \rho_g^w). \quad (22)$$

In the saturated regions Henry's and Raoult's laws are valid and we can work with the variables  $p_g$  and  $S_g$  in the equations (15),(16). Other choice of phase pressure and saturation variables is possible from (1) and (2). In the unsaturated regions ( $S_g = 0, 1$ ) this choice of primary unknowns is not possible. We will therefore analyze the saturated and the unsaturated regions separately.

In the *saturated regions*, we obtain the following system, written in the primary unknowns  $S_g$  and  $p_g$ :

$$\Phi \frac{\partial}{\partial t} (S_l \rho_l^{std} + S_g \rho_g^w) + \text{div}(\rho_l^{std} \mathbf{q}_l + \rho_g^w \mathbf{q}_g - \mathbf{J}_l - \mathbf{J}_g) = \mathcal{F}^w \quad (23)$$

$$\Phi \frac{\partial}{\partial t} (S_l \rho_l^h + S_g \rho_g^h) + \text{div}(\rho_l^h \mathbf{q}_l + \rho_g^h \mathbf{q}_g + \mathbf{J}_l + \mathbf{J}_g) = \mathcal{F}^h \quad (24)$$

$$\mathbf{J}_l = -\Phi S_l \mathcal{D}_l \nabla p_g \quad \mathbf{J}_g = -\Phi S_g \mathcal{D}_g \nabla p_g \quad (25)$$

where we have introduced the coefficients

$$\mathcal{D}_l = \frac{\rho_l^{std}}{\rho_l} \frac{d\bar{p}_g^h}{dp_g} C_l^h D_l^h \quad \mathcal{D}_g = \frac{1}{\rho_g} \left( p_g \frac{d\bar{p}_g^h}{dp_g} - \bar{p}_g^h \right) C_g^h C_g^w D_g^h \quad (26)$$

and the phase velocities are given by the Darcy-Muskat law as before. System (1), (2), (17), (18), (23), (24), (25), can be written in the terms of the variables  $p_g$  and  $S_g$  since in the saturated region all other variables can be expressed in terms of  $p_g$  and  $S_g$ . Note also that the diffusive terms (26) are strictly positive if the water vapor pressure is strictly positive. If there is no evaporation of the water, then  $\mathbf{J}_g = 0$ , since the gas consists only of the hydrogen.

The situation where the gas phase is missing ( $S_g = 0, S_l = 1$ ) we will refer to as *the unsaturated case 1*. In this case we replace the saturation  $S_g$  by  $\rho_l^h$ , and we will work with  $\rho_l^h$  and  $p_l$  as primary unknowns. The governing equations reduce to

$$\text{div}(\rho_l^{std} \mathbf{q}_l - \mathbf{J}_l) = \mathcal{F}^w, \quad \Phi \frac{\partial \rho_l^h}{\partial t} + \text{div}(\rho_l^h \mathbf{q}_l + \mathbf{J}_l) = \mathcal{F}^h \quad (27)$$

$$\mathbf{q}_l = -\mathbb{K} \lambda_l(0) (\nabla p_l - \rho_l \mathbf{g}), \quad \mathbf{J}_l = -\Phi \frac{\rho_l^{std}}{\rho_l} D_l^h \nabla \rho_l^h. \quad (28)$$

The situation where the liquid phase is missing ( $S_g = 1, S_l = 0$ ) we refer to as *the unsaturated case 2*. In this case the saturation variable  $S_g$  will be replaced by the variable  $\rho_g^h$  and we will choose  $\rho_g^h$  and  $p_g$  as primary unknowns. The vapor mass concentration  $\rho_g^w$  can be expressed as a function of the gas phase pressure and  $\rho_g^h$ :

$$\rho_g^w = C_g^w p_g - \frac{M^w}{M^h} \rho_g^h, \quad \rho_g^w \nabla \rho_g^h - \rho_g^h \nabla \rho_g^w = C_g^w (p_g \nabla \rho_g^h - \rho_g^h \nabla p_g). \quad (29)$$

The governing equations reduce to

$$\Phi \frac{\partial \rho_g^w}{\partial t} + \operatorname{div}(\rho_g^w \mathbf{q}_g - \mathbf{J}_g) = \mathcal{F}^w, \quad \Phi \frac{\partial \rho_g^h}{\partial t} + \operatorname{div}(\rho_g^h \mathbf{q}_g + \mathbf{J}_g) = \mathcal{F}^h \quad (30)$$

$$\mathbf{q}_g = -\mathbb{K} \lambda_g(1) (\nabla p_g - \rho_g \mathbf{g}), \quad \mathbf{J}_g = -\Phi \frac{1}{\rho_g} C_g^w D_g^h (p_g \nabla \rho_g^h - \rho_g^h \nabla p_g). \quad (31)$$

The system (30)-(31) can be written in the terms of the gas pressure  $p_g$  and the hydrogen mass concentration in the gas phase  $\rho_g^h$  since  $\rho_g$  and  $\rho_g^w$  can be expressed in terms of them.

In the next section, in order to avoid change of the pressure variable between the saturated and the unsaturated zones, following the idea developed in [3], we introduce a new variable called the global pressure.

### 3 Global pressure formulation

Let us consider the saturated region and rewrite the equations (23)-(25) by summation, in the following form:

$$\Phi \frac{\partial}{\partial t} (\rho_l(1 - S_g) + \rho_g S_g) + \operatorname{div}(\rho_l \mathbf{q}_l + \rho_g \mathbf{q}_g) = \mathcal{F}^w + \mathcal{F}^h \quad (32)$$

$$\Phi \frac{\partial}{\partial t} (\rho_l^h(1 - S_g) + \rho_g^h S_g) + \operatorname{div}(\rho_l^h \mathbf{q}_l + \rho_g^h \mathbf{q}_g + \mathbf{J}_l + \mathbf{J}_g) = \mathcal{F}^h. \quad (33)$$

Let us define the total component fluxes as:

$$\mathbf{Q}^h = \rho_l^h \mathbf{q}_l + \rho_g^h \mathbf{q}_g + \mathbf{J}_l + \mathbf{J}_g \quad (34)$$

$$\mathbf{Q}^w = \rho_l^w \mathbf{q}_l + \rho_g^w \mathbf{q}_g - \mathbf{J}_l - \mathbf{J}_g, \quad (35)$$

and the total flow as the sum of the components fluxes:

$$\mathbf{Q}_t = \mathbf{Q}^w + \mathbf{Q}^h = \rho_l \mathbf{q}_l + \rho_g \mathbf{q}_g. \quad (36)$$

Let us introduce the total mobility  $\lambda(p_g, S_g)$ , the fractional flow functions of the gas and liquid phases  $f_g(p_g, S_g)$ ,  $f_l(p_g, S_g)$ , and the mean density function  $\rho(p_g, S_g)$  as follows:

$$\lambda = \rho_l \lambda_l(S_g) + \rho_g \lambda_g(S_g) \quad (37)$$

$$f_g = \frac{\rho_g \lambda_g(S_g)}{\lambda(p_g, S_g)}, \quad f_l = \frac{\rho_l \lambda_l(S_g)}{\lambda(p_g, S_g)} \quad (38)$$

$$\rho = \frac{\rho_l^2 \lambda_l(S_g) + \rho_g^2 \lambda_g(S_g)}{\lambda(p_g, S_g)} \quad (39)$$

Note that  $f_g + f_l = 1$ . In the saturated region, all these functions depend only on the gas pressure and the gas saturation, since all the mass densities and the concentrations depend only on the gas pressure. In the unsaturated regions they depend on the gas/liquid pressure and the corresponding hydrogen mass concentration.

From the Darcy-Muscat law we can rewrite the total flow as

$$\mathbf{Q}_t = -\lambda \mathbb{K} (\nabla p_l + f_g \nabla p_c(S_g) - \rho \mathbf{g}), \quad (40)$$



or

$$\mathbf{Q}_t = -\lambda\mathbb{K}(\nabla p_g - f_l \nabla p_c(S_g) - \rho\mathbf{g}). \quad (41)$$

In order to write the total flow in a form of the Darcy-Muscat law we need to eliminate  $\nabla p_c(S_g)$  from (40) and (41) in the saturated region; in the unsaturated regions the total flow reduces to the gas or the liquid flow. This can be done by introducing a new pressure variable  $p$ , called the global pressure, and by expressing the phase pressures as functions of  $p$ :  $p_g = p_g(p, S_g)$  and  $p_l = p_l(p, S_g)$ . Obviously we will have  $p_l(p, S_g) = p_g(p, S_g) - p_c(S_g)$  and therefore, we need to find only one function, say  $p_g(p, S_g)$ . This functions has to satisfy (see [3])

$$\nabla p_g(p, S_g) - f_l \nabla p_c(S_g) = \omega(p, S_g) \nabla p, \quad (42)$$

for some scalar function  $\omega$ . This problem can be easily solved in the saturated region ( $0 < S_g < 1$ ) where the fractional flow  $f_l$  depends on  $p_g$  and  $S_g$ . It is sufficient to solve the following Cauchy problem

$$\begin{cases} \frac{\partial p_g}{\partial S_g}(p, S_g) = f_l(p_g(p, S_g), S_g) p'_c(S_g) & 0 < S_g < 1 \\ p_g(p, 0) = p + p_c(0) \end{cases} \quad (43)$$

for all values of the global pressure  $p$  (which has a role of a parameter). Note that we set the initial condition  $p_g(p, 0) = p + p_c(0)$ , so in *the unsaturated case 1* ( $S_g = 0$ ) the global pressure equals to the pressure of the liquid phase.

Since in most capillary pressure models the function  $\lambda_l(S_g) p'_c(S_g)$  is a bounded function in the vicinity of  $S_g = 1$ , the problem (43) has a unique solution on the whole segment  $S_g \in (0, 1]$ . It follows that the function  $p \mapsto p_g(p, 1)$  is well defined and invertible, such that the global pressure  $p$  can be used as the pressure unknown also in *the unsaturated case 2*. We will use the notation  $p_g^1(p) = p_g(p, 1)$ .

The above discussion shows that the global pressure can be used in all three regimes as unique pressure unknown. Now, we need to show how to calculate the function  $\omega(p, S_g)$ . From (42) it follows

$$\frac{\partial p_g}{\partial p}(p, S_g) = \frac{\partial p_l}{\partial p}(p, S_g) = \omega(p, S_g), \quad (44)$$

and therefore, the function  $\omega(p, S_g)$  is the solution of the problem

$$\begin{cases} \frac{\partial \omega}{\partial S_g}(p, S_g) = \partial_{p_g} f_l(p_g(p, S_g), S_g) p'_c(S_g) \omega(p, S_g) & 0 < S_g < 1 \\ \omega(p, 0) = 1. \end{cases} \quad (45)$$

Now, it is not difficult to see that the solution is given by

$$\omega(p, S_g) = \exp\left(-\int_0^{S_g} (v_g(s, p) - v_l(s, p)) \frac{\rho_l(p_g(p, s)) \rho_g(p_g(p, s)) \lambda_l(s) \lambda_g(s) p'_c(s)}{(\rho_l(p_g(p, s)) \lambda_l(s) + \rho_g(p_g(p, s)) \lambda_g(s))^2} ds\right) \quad (46)$$

where  $v_g(s, p) = \frac{\rho'_g(p_g(p, S_g))}{\rho_g(p_g(p, S_g))}$ ,  $v_l(s, p) = \frac{\rho'_l(p_g(p, S_g))}{\rho_l(p_g(p, S_g))}$ . The function  $\omega$  is well defined, bounded and strictly positive for  $S_g \in [0, 1]$  and  $p \in \mathbb{R}$ , and therefore the application  $p \mapsto p_g(p, S_g)$  is invertible for all  $S_g \in [0, 1]$ .

The total flux can be expressed as

$$\mathbf{Q}_t = -\lambda \mathbb{K}(\omega \nabla p - \rho \mathbf{g}), \quad (47)$$

where the coefficients  $\lambda$ ,  $\omega$  and  $\rho$  are given by (37), (46) and (39). This representation uses the global pressure in both saturated and unsaturated regions. In the other hand, formulas for expressing the coefficients  $\lambda$ ,  $\omega$  and  $\rho$  change when passing from saturated to unsaturated region and back.

We can also write

$$\mathbf{Q}^h = -\Lambda^h \mathbb{K} \nabla p - (c_g^h - c_l^h) a \mathbb{K} \nabla p_c(S_g) + b_g^h \mathbb{K} \mathbf{g} + \mathbf{J}_l + \mathbf{J}_g \quad (48)$$

where the coefficients are given as

$$\Lambda^h = (\rho_l^h \lambda_l(S_g) + \rho_g^h \lambda_g(S_g)) \omega(p, S_g), \quad (49)$$

$$a = \frac{\rho_l \rho_g \lambda_g(S_g) \lambda_l(S_g)}{\lambda(p, S_g)} \quad (50)$$

$$b_g^h = \rho_l^h \rho_l \lambda_l(S_g) + \rho_g^h \rho_g \lambda_g(S_g). \quad (51)$$

In formulation (48) of the total hydrogen flux the global pressure can be used in both saturated and unsaturated regions. In the other hand, the diffusive fluxes and all the coefficients change their form in different regions.

Finally we obtain the following system of equations which relies on the global pressure variable as one unknown, and the gas saturation/hydrogen mass concentration as another unknown:

$$\Phi \frac{\partial}{\partial t} (\rho_l (1 - S_g) + \rho_g S_g) - \text{div} (\lambda \mathbb{K} (\omega \nabla p - \rho \mathbf{g})) = \mathcal{F}^w + \mathcal{F}^h \quad (52)$$

$$\begin{aligned} \Phi \frac{\partial}{\partial t} (\rho_l^h (1 - S_g) + \rho_g^h S_g) - \text{div} \left( \Lambda^h \mathbb{K} \nabla p + (c_g^h - c_l^h) a \mathbb{K} \nabla p_c(S_g) - b_g^h \mathbb{K} \mathbf{g} \right) \\ - \text{div} \left( \Phi \frac{S_l \rho_l^{std}}{\rho_l} D_l^h \nabla \rho_l^h + \Phi \frac{S_g}{\rho_g} D_g^h (\rho_g^w \nabla \rho_g^h - \rho_g^h \nabla \rho_g^w) \right) = \mathcal{F}^h. \end{aligned} \quad (53)$$

## 4 Saturated-Unsaturated case formulation

The system of equations (52), (53) has different form in the saturated and the unsaturated regions. In the saturated region all the mass concentrations,  $\rho_l^h$ ,  $\rho_g^h$  and  $\rho_g^w$ , are functions of the gas pressure, which can be expressed as functions of the global pressure and the saturation. In the unsaturated region 1 ( $S_g = 0$ ) the convective and diffusive gas fluxes are zero and  $\rho_l^h$  becomes an independent unknown. In the unsaturated region 2 ( $S_g = 1$ ) the convective and diffusive liquid fluxes are zero and  $\rho_g^h$  becomes an independent unknown. Our goal in this section is to introduce one persistent unknown to avoid the change of unknowns in different regions.

Following the idea explained in [6] we introduce a new variable  $X$  which represents the total hydrogen mass density and is defined as follows:

$$X = (1 - S_g) \rho_l^h + S_g \rho_g^h. \quad (54)$$

More precisely, we can write

$$X = \begin{cases} \rho_l^h & \text{if } S_g = 0 \\ (C_l^h(1 - S_g) + C_g^h S_g) \bar{p}_g^h(p, S_g) & \text{if } 0 < S_g < 1 \\ \rho_g^h & \text{if } S_g = 1 \end{cases} . \quad (55)$$

Here we use the fact that the function  $\bar{p}_g^h$  depends on the gas pressure  $p_g$ , which can be expressed as a function of the global pressure and the saturation,  $p_g = p_g(p, S_g)$ , and therefore we write  $p_g^h = \bar{p}_g^h(p, S_g)$  instead of  $p_g^h = \bar{p}_g^h(p_g(p, S_g))$ . This convention will be used for other functions too.

In order to express the gas saturation as a function of the new variable  $X$  we must show that the relation (55) is invertible in the saturated region. For this, we will use the fact that for the hydrogen, we have

$$C_g^h - C_l^h > 0. \quad (56)$$

Now we can show that the relation (55) can be inverted by calculating the derivative  $\partial X / \partial S_g$ . To simplify the calculation we use the following notation:

$$\alpha(p, S_g) = \left( (C_g^h - C_l^h) S_g + C_l^h \right) \frac{d\bar{p}_g^h}{dp_g}(p_g(p, S_g)), \quad (57)$$

$$\beta(p, S_g) = (C_g^h - C_l^h) \bar{p}_g^h(p, S_g). \quad (58)$$

Using (11) and (43) it is easy to see that for all  $0 < S_g < 1$

$$\frac{\partial X}{\partial S_g} = \beta(p, S_g) + \alpha(p, S_g) \frac{\partial p_g}{\partial S_g} > 0, \quad (59)$$

and therefore, for each  $p$  we can find the inverse function  $S_g = S_g(p, X)$  which satisfies

$$\frac{\partial S_g}{\partial X} > 0, \quad 0 < S_g < 1. \quad (60)$$

This function can be expressed as follows (see the discussion on the phase transition criteria in Section 2.1)

$$S_g = \begin{cases} 0 & \text{if } X \leq C_l^h \bar{p}_g^h(p, 0) \\ X_{S_g}^{-1}(p, X) & \text{if } C_l^h \bar{p}_g^h(p, 0) < X < C_g^h \bar{p}_g^h(p, 1) \\ 1 & \text{if } X \geq C_g^h \bar{p}_g^h(p, 1) \end{cases} . \quad (61)$$

By using that  $C_l^h \leq C_l^h(1 - S_g) + C_g^h S_g \leq C_g^h$ , which follows from (56), we can now express all mass concentrations through  $X$  and  $p$ :

$$\rho_l^h(p, X) = \min(C_l^h p_g^h(p, S_g(p, X)), X), \quad \text{for } S_g < 1, \quad (62)$$

$$\rho_g^h(p, X) = \max(C_g^h p_g^h(p, S_g(p, X)), X), \quad \text{for } S_g > 0, \quad (63)$$

$$\rho_g^w(p, X) = C_g^w p_g(p, S_g(p, X)) - \frac{C_g^w}{C_g^h} \rho_g^h(p, X), \quad \text{for } S_g > 0. \quad (64)$$

Note that the expression for  $\rho_l^h(p, X)$  is not valid for  $S_g = 1$ , since the concentration becomes meaningless, but this is not important since  $\rho_l^h$  appears in the differential equations always

multiplied by  $S_l$  or a quantity that is equal to zero at  $S_l = 0$ . Similar statement holds for  $\rho_g^h$  and  $\rho_g^w$ . Therefore, we can take that (62), (63) and (64) hold, by extension, for all  $S_g \in [0, 1]$ .

We can now rewrite the system (52), (53) in the variables  $X$  and  $p$  by using (61), (62), (63) and (64) to express all coefficients as functions of  $X$  and  $p$ . To finish the transformation we need to calculate the partial derivatives of  $S_g$ ,  $\rho_l^h$ ,  $\rho_g^h$  and  $\rho_g^w$  with respect to  $p$  and  $X$ .

Now, a direct calculus gives us the partial derivatives of  $S_g(p, X)$ :

$$\frac{\partial S_g}{\partial p} = -\frac{\omega(p, S_g)\alpha(p, S_g)\chi(p, X)}{\beta(p, S_g) + \alpha(p, S_g)\frac{\partial p_g}{\partial S_g}(p, S_g)}, \quad (65)$$

$$\frac{\partial S_g}{\partial X} = \frac{\chi(p, X)}{\beta(p, S_g) + \alpha(p, S_g)\frac{\partial p_g}{\partial S_g}(p, S_g)}, \quad (66)$$

where for every  $p$  we denote by  $\chi(p, X)$  the characteristic function of the set

$$A_p = \left\{ (p, X), C_l^h \bar{p}_g^h(p, 0) < X < C_g^h \bar{p}_g^h(p, 1) \right\}, \quad (67)$$

which describes the saturated region in the  $p, X$ -space. It is useful to introduce the following functions

$$\Omega = \omega + \frac{\partial p_g}{\partial S_g} \frac{\partial S_g}{\partial p}, \quad M = \frac{\partial p_g}{\partial S_g} \frac{\partial S_g}{\partial X}. \quad (68)$$

Then we have

$$\Omega\chi = \omega(p, X) \frac{\beta(p, S_g)\chi(p, X)}{\beta(p, S_g) + \alpha(p, S_g)\frac{\partial p_g}{\partial S_g}(p, S_g)}, \quad \Omega(1 - \chi) = \omega(p, X) \quad (69)$$

$$M = \frac{\chi(p, X)\frac{\partial p_g}{\partial S_g}(p, S_g)}{\beta(p, S_g) + \alpha(p, S_g)\frac{\partial p_g}{\partial S_g}(p, S_g)}, \quad (70)$$

and the derivatives of the mass concentrations can be expressed in the following way (further details may be found in Appendix A, formulas (81) and (82)):

$$\frac{\partial \rho_l^h}{\partial p} = C_l^h \frac{d\bar{p}_g^h}{dp_g} \Omega\chi, \quad \frac{\partial \rho_l^h}{\partial X} = 1 - \chi + C_l^h \frac{d\bar{p}_g^h}{dp_g} M\chi. \quad (71)$$

The derivatives are possibly not correct in the region where  $S_g = 1$ , but in the regions saturated by the gas these terms are multiplied by zero. In the same way we have

$$\frac{\partial \rho_g^h}{\partial p} = C_g^h \frac{d\bar{p}_g^h}{dp_g} \Omega\chi, \quad \frac{\partial \rho_g^h}{\partial X} = 1 - \chi + C_g^h \frac{d\bar{p}_g^h}{dp_g} M\chi \quad (72)$$

As well, the derivatives are possibly not correct in the region where  $S_g = 0$ , but in the liquid saturated region these term are multiplied by zero.

The diffusive fluxes can be written as (further details may be found in Appendix A, formula (83)):

$$\mathbf{J}_l + \mathbf{J}_g = -\Phi\{\mathcal{A}_p \nabla p + \mathcal{A}_X \nabla X\}, \quad (73)$$

where

$$\mathcal{A}_p = \left( \frac{S_l \rho_l^{std}}{\rho_l} D_l^h C_l^h \frac{d\bar{p}_g^h}{dp_g} + \frac{S_g}{\rho_g} D_g^h [C_g^h \rho_g^w \frac{d\bar{p}_g^h}{dp_g} + \rho_g^h C_g^w (\frac{d\bar{p}_g^h}{dp_g} - 1)] \right) \Omega_\chi \quad (74)$$

$$- \frac{S_g}{\rho_g} D_g^h \rho_g^h C_g^w \omega (1 - \chi),$$

$$\mathcal{A}_X = \left\{ \frac{S_l \rho_l^{std}}{\rho_l} D_l^h + \frac{S_g}{\rho_g} D_g^h [\rho_g^w + \rho_g^h \frac{C_g^w}{C_g^h}] \right\} [1 - \chi] \quad (75)$$

$$+ \left[ \frac{S_l}{\rho_l} D_l^h C_l^h + \frac{S_g}{\rho_g} D_g^h C_g^h \rho_g^w \right] \frac{dp_g^h}{dp_g} M\chi + \frac{S_g}{\rho_g} D_g^h \rho_g^h C_g^w \left[ \frac{dp_g^h}{dp_g} - 1 \right] M\chi.$$

Finally, the system (52), (52) can be written as

$$\Phi \frac{\partial \mathcal{N}(p, X)}{\partial t} - \text{div}(\lambda \mathbb{K}(\omega \nabla p - \rho \mathbf{g})) = \mathcal{F}^w + \mathcal{F}^h, \quad (76)$$

$$\Phi \frac{\partial X}{\partial t} - \text{div} \left( (\mathcal{B}_p \mathbb{K} + \mathcal{A}_p \Phi) \nabla p + (\mathcal{B}_X \mathbb{K} + \mathcal{A}_X \Phi) \nabla X - b_g^h \mathbb{K} \mathbf{g} \right) = \mathcal{F}^h, \quad (77)$$

where the coefficients are given by

$$\mathcal{N}(p, X) = X + \rho_l^{std} - (\rho_l^{std} - \rho_g^w) S_g, \quad (78)$$

$$\mathcal{B}_p(p, X) = [\Lambda^h + (c_g^h - c_l^h) a(p, S_g) p'_c(S_g) \frac{\partial S_g}{\partial p}], \quad (79)$$

$$\mathcal{B}_X(p, X) = (c_g^h - c_l^h) a(p, S_g) p'_c(S_g) \frac{\partial S_g}{\partial X}. \quad (80)$$

The derivative (65) is negative and therefore the derivative of  $\mathcal{N}$  with respect to  $p$  is positive. This, together with the fact that the coefficient  $\lambda \omega$  is strictly positive makes the partial differential equation (76) parabolic as an equation for the global pressure  $p$ . However, it is degenerate parabolic equation since the derivative  $\partial \mathcal{N} / \partial p$  is equal to zero in the unsaturated regions. This type of degeneracy is already found in compressible immiscible flows.

The equation (77) is parabolic, of convection-diffusion type in the variable  $X$ . The capillary diffusion coefficient  $\mathcal{B}_X$  is non negative since in flows modeled by Henry's and Raoult's laws one has  $c_g^h - c_l^h > 0$ . This coefficient degenerates to zero in the unsaturated regions, as well as in immiscible flow. The diffusion coefficient  $\mathcal{A}_X$  stays strictly positive in both saturated and unsaturated regions (see (11)). Note also that the coefficients  $\mathcal{B}_X$  and  $\mathcal{B}_p$  are continuous on the boundary between the saturated and the unsaturated regions. The coefficients  $\mathcal{A}_X$  and  $\mathcal{A}_p$ , in the other hand, are generally discontinuous and this discontinuity is solution dependent.

There are several different convection terms in (77): convection governed by  $\mathbb{K} \nabla p$ ,  $\Phi \nabla p$  and  $\mathbb{K} \mathbf{g}$ . Nonlinear functions in this convection terms are strictly positive as  $b_g^h$ , non negative as  $\mathcal{B}_p$ , or it can turn to negative in the unsaturated regions as  $\mathcal{A}_p$ .

We have shown that the global pressure and the total hydrogen mass can be used as the unknowns in the two-phase partially miscible flow equations which can describe the flow in both saturated and unsaturated regions. The system of equations written in these variables shows the same kind of degeneracies as the immiscible two-phase flow. The presence of the diffusion in the gas and liquid phases removes the degeneracy in the convection-diffusion equation for  $X$ . In the other hand, the convective part of equation (77) is complicated by the presence of the diffusive fluxes.

Temperature	298.15 K
$M^w$	$18.0153 \cdot 10^{-3}$ kg/mol
$M^h$	$2.0159 \cdot 10^{-3}$ kg/mol
$\rho_w^{std}$	$10^3$ kg/m <sup>3</sup>
$H(T = 298.15 \text{ K})$	$7.8 \cdot 10^{-6}$ mol/(m <sup>3</sup> Pa)
$\hat{p}_g^w(T = 298.15 \text{ K})$	2310 Pa
$D_l^h$	$5.6 \cdot 10^{-9}$ m <sup>2</sup> /s
$D_g^h$	$7.6 \cdot 10^{-5}$ m <sup>2</sup> /s
$\mu_l$	$10^{-3}$ Pa·s
$\mu_g$	$10^{-5}$ Pa·s

Table 1: Fluid properties

## 5 An example of numerical computation of the coefficients

In this section, we illustrate our new model for the global pressure–total mass formulation by computing different coefficients that appear in the system of PDEs (76)–(77). We consider a liquid-gas system with two components: water and hydrogen. For simplicity we will assume that the phase viscosities are independent of the composition. The fluid properties used in the coefficients are given in Table 1. With these parameters for the hydrogen, the function  $\bar{p}_g^h(p_g)$  is almost affine.

We use the Van Genuchten-Mualem relative permeability functions,

$$kr_w(S_g) = \sqrt{1 - S_g} \left[ 1 - \left( 1 - (1 - S_g)^{1/m} \right)^m \right]^2, \quad kr_g(S_g) = \sqrt{S_g} \left[ 1 - (1 - S_g)^{1/m} \right]^{2m}.$$

and Van Genuchten’s capillary pressure function,

$$p_c(S_g) = P_e \left( (1 - S_g)^{-1/m} - 1 \right)^{1/n},$$

where  $m = 1 - 1/n$  and it is assumed that the water and the gas residual saturation are equal to zero. The coefficients taken in this calculations are  $n = 2$  and  $P_e = 2.0$  MPa.

For the calculation of the coefficients, a code is created which uses GSL library [13] for solution of the Cauchy problem (43). The function  $p_g(p, S_g)$  is thus obtained in tabular form, for different values of the global pressure  $p$ ; an interpolation procedure is used to define  $p_g(p, S_g)$  for all values of  $p$  and  $S_g$ . The function  $S_g(p, X)$  is also calculated in tabular form from the table of values  $X(S_g, p)$ .

$\omega(p, S)$  and  $p_g(p, S)$  are presented as functions of the gas phase saturation  $S = S_g$  for several fixed global pressure values  $p$  in Figure 1.

In Figure 2 we show the function  $S_g(p, X)$  for different values of  $p$  in certain range of the total hydrogen mass density (between zero and 6 kg/m<sup>3</sup>). As it can be seen in Figure 2, in the largest part of the two-phase flow the function  $X \mapsto S_g(p, X)$  is linear; we also show in Figure 2 the dissolved hydrogen mass concentration  $\rho_l^h(p, X)$ .

From Figure 2 we can find the extent of the saturated zone for given global pressure  $p$ . For example, for the value  $p = 0.1$  MPa the saturated zone is between  $X_1 = 3.1 \cdot 10^{-3}$  and  $X_2 = 2.09$  kg/m<sup>3</sup>, for the value  $p = 1.0$  MPa is between  $X_1 = 1.6 \cdot 10^{-2}$  and  $X_2 = 2.63$  kg/m<sup>3</sup> and for the value  $p = 5.0$  MPa is between  $X_1 = 7.9 \cdot 10^{-2}$  and  $X_2 = 5.56$  kg/m<sup>3</sup>

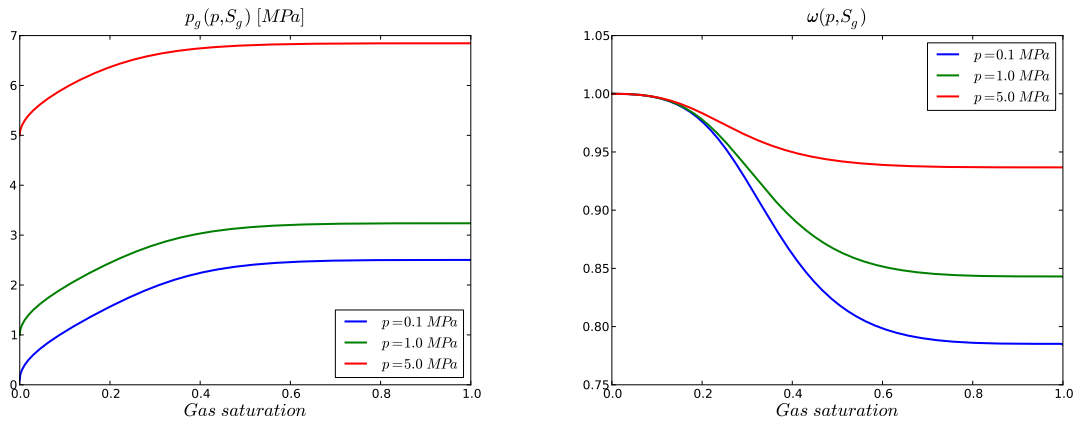


Figure 1: Gas pressure  $p_g(p, S)$  and  $\omega(p, S)$  as functions of the gas saturation  $S$ , for different values of the global pressure  $p$ .

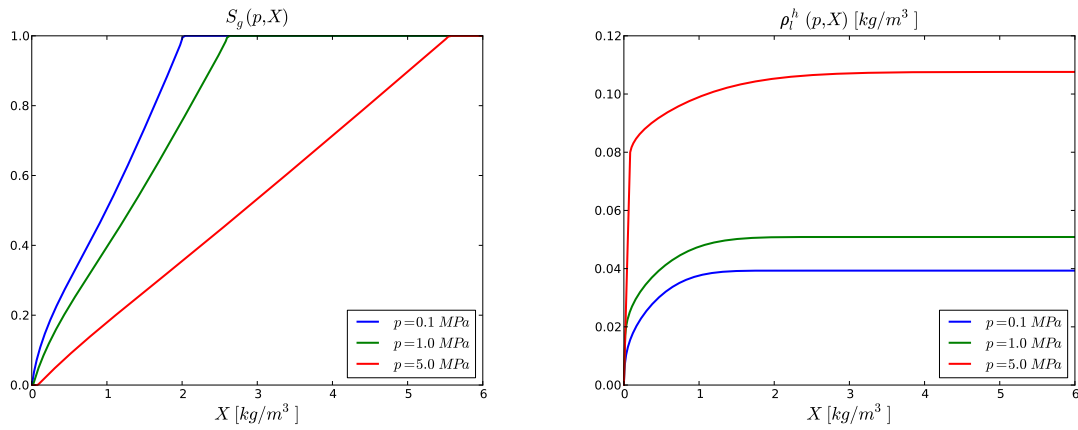


Figure 2: Gas saturation  $S_g(p, X)$  and dissolved hydrogen concentration  $\rho_i^h(p, X)$  as a function of the total hydrogen density  $X$ , for different values of the global pressure  $p$ .

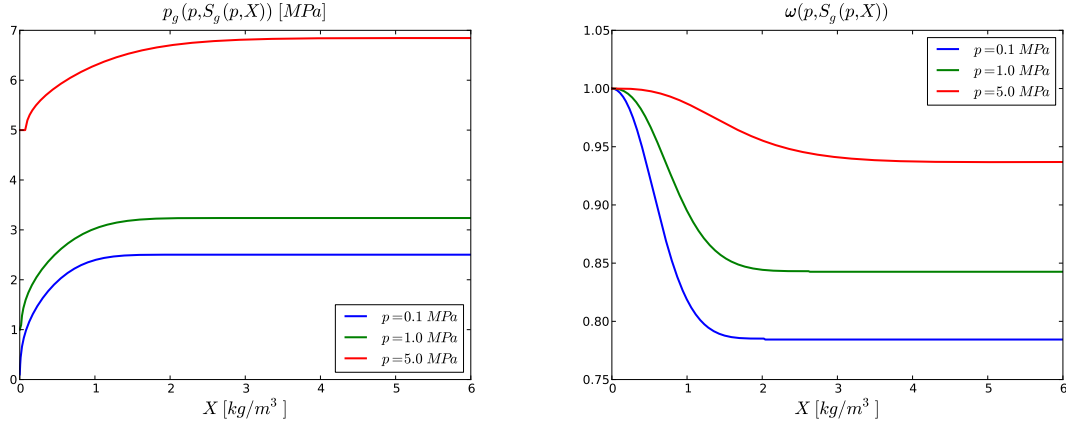


Figure 3: Gas pressure  $p_g(p, X)$  and  $\omega(p, X)$  as functions of the total hydrogen density  $X$ , for different values of the global pressure  $p$ .

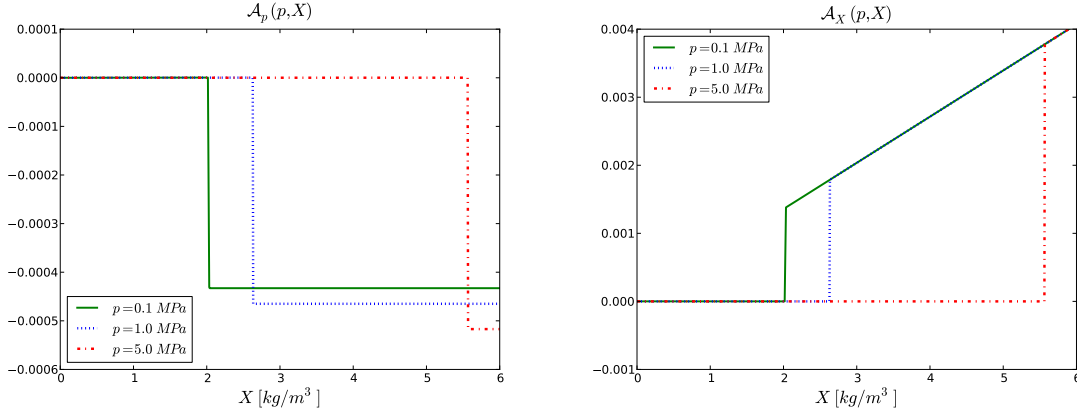


Figure 4: Diffusive coefficients  $\mathcal{A}_p$  and  $\mathcal{A}_X$  as functions of the total hydrogen density  $X$ , for different values of the global pressure  $p$ .

Due to the fact that the function  $X \mapsto S_g(p, X)$  is linear in large part of its domain we see in Figure 3 that the coefficients  $\omega(p, S)$  and  $p_g(p, S)$  presented as functions of  $X$  and  $p$ , do not change significantly.

These figures show that introduction of the variable  $X$  instead of  $S_g$  does not change much the shape of the functions of the saturation. Finally, we show the coefficients  $\mathcal{A}_p$  and  $\mathcal{A}_X$  that come from the diffusive fluxes. We can see that these coefficients are discontinuous between saturated and unsaturated zone 2 where  $\mathcal{A}_p$  becomes negative. In the unsaturated zone 1, the value of these coefficients is very small.

## 6 Conclusion

We have developed a new formulation for a compositional compressible two-phase flow model based on the concept of the global pressure which is fully equivalent to the original phase equations formulation. Our approach consists first of introduction of the global pressure which



eliminates the need to switch the pressure variable when passing from the saturated to the unsaturated states. Then we replace the saturation variable, usually taken as an unknown in saturated state, with a new variable that can describe, together with the global pressure, the saturated and the unsaturated states. The mathematical structure of the coupled system is well defined and its properties are close to those of immiscible two-phase flow equations. The behavior of the coefficients appearing in the system is illustrated for a liquid-gas system with two components: water and hydrogen. We present numerical computations for a simple compositional two-phase flow model with two chemical components and we show how to construct a mathematical model capable of representing fluid state and evolution without changing the variables between the saturated and the unsaturated states. The model is presented as an application for water– $H_2$  system, but it could be extended to more complicated systems, such as water– $CO_2$  system. Our future work will be concentrated on the numerical simulations based on this model using a finite volume method, including the cases of heterogeneous porous media with multiple rock types.

## A Appendix

In this appendix, for the reader's convenience, we detail some calculations from Section 4. First, we will develop the computation of the derivatives with respect to  $p$  and  $X$  for  $\rho_l^h$ . The derivatives of  $\rho_g^h$  are calculated similarly. From definition (62) we have

$$\begin{aligned}
\frac{\partial \rho_l^h}{\partial p} &= \frac{\partial}{\partial p} \begin{cases} C_l^h p_g^h(p, S_g) & \text{if } C_l^h p_g^h(p, S_g) < X \Leftrightarrow S_g > 0 \\ X & \text{if } C_l^h p_g^h(p, S_g) \geq X \Leftrightarrow S_g = 0 \end{cases} \\
&= \begin{cases} C_l^h \frac{d\bar{p}_g^h}{dp_g} \left( \frac{\partial p_g}{\partial p} + \frac{\partial p_g}{\partial S_g} \frac{\partial S_g}{\partial p} \right) & \text{if } C_l^h p_g^h(p, S_g) < X \Leftrightarrow S_g > 0 \\ 0 & \text{if } C_l^h p_g^h(p, S_g) \geq X \Leftrightarrow S_g = 0 \end{cases} \\
&= C_l^h \frac{d\bar{p}_g^h}{dp_g} \Omega \chi
\end{aligned} \tag{81}$$

The final formula may not be true in  $S_g = 1$  ( $S_l = 0$ ), but since this term appears in the diffusive fluxes always multiplied by  $S_l$ , we can simply set it to zero when  $S_g = 1$ . In the same way we have,

$$\begin{aligned}
\frac{\partial \rho_l^h}{\partial X} &= \frac{\partial}{\partial X} \begin{cases} C_l^h p_g^h(p, S_g) & \text{if } C_l^h p_g^h(p, S_g) < X \Leftrightarrow S_g > 0 \\ X & \text{if } C_l^h p_g^h(p, S_g) \geq X \Leftrightarrow S_g = 0 \end{cases} \\
&= \begin{cases} C_l^h \frac{d\bar{p}_g^h}{dp_g} \left( \frac{\partial p_g}{\partial S_g} \frac{\partial S_g}{\partial X} \right) & \text{if } C_l^h p_g^h(p, S_g) < X \Leftrightarrow S_g > 0 \\ 1 & \text{if } C_l^h p_g^h(p, S_g) \geq X \Leftrightarrow S_g = 0 \end{cases} \\
&= 1 - \chi + C_l^h \frac{d\bar{p}_g^h}{dp_g} M \chi.
\end{aligned} \tag{82}$$

where we have applied the same logic at  $S_g = 1$ . Formulas (72) are proved in the same way.

Next we present some details of calculation of the coefficients  $\mathcal{A}_p$  and  $\mathcal{A}_X$ . The diffusive

fluxes can be written as,

$$\begin{aligned}
\mathbf{J}_l + \mathbf{J}_g &= -\Phi S_l \frac{\rho_l^{std}}{\rho_l} D_l^h \nabla \rho_l^h - \Phi S_g \frac{1}{\rho_g} D_g^h (\rho_g^w \nabla \rho_g^h - \rho_g^h \nabla \rho_g^w) \\
&= -\Phi \left\{ \frac{S_l \rho_l^{std}}{\rho_l} D_l^h \frac{\partial \rho_l^h}{\partial p} + \frac{S_g}{\rho_g} D_g^h [\rho_g^w + \rho_g^h \frac{C_g^w}{C_g^h}] \frac{\partial \rho_g^h}{\partial p} - \frac{S_g}{\rho_g} D_g^h \rho_g^h C_g^w \left( \frac{\partial p_g}{\partial p} + \frac{\partial p_g}{\partial S_g} \frac{\partial S_g}{\partial p} \right) \right\} \nabla p \\
&\quad - \Phi \left\{ \frac{S_l \rho_l^{std}}{\rho_l} D_l^h \frac{\partial \rho_l^h}{\partial X} + \frac{S_g}{\rho_g} D_g^h [\rho_g^w + \rho_g^h \frac{C_g^w}{C_g^h}] \frac{\partial \rho_g^h}{\partial X} - \frac{S_g}{\rho_g} D_g^h \rho_g^h C_g^w \frac{\partial p_g}{\partial X} \right\} \nabla X \\
&= -\Phi \{ \mathcal{A}_p \nabla p + \mathcal{A}_X \nabla X \}
\end{aligned} \tag{83}$$

Using expressions (71), (72) and taking care of saturated and unsaturated regions one can easily deduce (74) and (75).

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