A DUAL CONTINUUM COUPLED MULTIPHASE FLOW MODEL WITH MIXED SECOND ORDER WATER TRANSFER TERM FOR STRUCTURED SOILS: I THEORY

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ABSTRACT. Compacted bentonites are being used in many countries as backfill and sealing material in high-level radioactive waste disposal. Experimental studies indicate that bentonite exhibits two types of porosity: A macro-porous domain which is characterized by free water and a micro-porous domain mainly composed of interlayer water. The microscopic heterogeneity of bentonite and the interactions of thermal, hydrodynamic and geochemical processes call for a fully coupled dual-domain flow model to describe the hydrodynamic behaviour of bentonite. In this series of two papers we present a dual domain model. In the first part we present the mathematical formulation of non-isothermal multiphase flow considering chemical and thermal osmosis. A second order water transfer term which accounts for water transfer between the two domains induced by both hydraulic pressure and chemical osmosis pressure gradient is proposed. Their implementation in a finite element code are discussed. Testing of the mathematical formulation with synthetic and real data is presented in the second paper of the series.

RESUMEN. Varios paises contemplan la utilización de bentonita compactada como material de relleno y sellado de un almacenamiento profundo de residuos radiactivos. Los estudios experimentales indican que la bentonita muestra dos tipos de porosidad: Los macroporos con agua libre y los microporos con agua interlaminar. La heterogeneidad microscópica de la bentonita y las interacciones entre los procesos térrmicos, hidrodinámicos y geoquímicos requieren utilizar un modelo de doble porosidad. En esta serie de dos trabajos se presenta un modelo de doble porosidad. En este primer trabajo se describe la formulación matemática del flujo noisotermo multifásico considerando ósmosis térmica y química. Se utiliza un término de segundo orden para el flujo de agua entre subdominios producido por los gradientes de presión intersticial y osmótica. Se describe además la implementación de esta formulación en un programa de elementos finitos. En el segundo trabajo se describe la aplicación del modelo a datos sintéticos y a un experimento real.

1. Introduction

'Dual continuum', 'dual domain' or 'double porosity' are terms generally used to denote a conceptual model, in which the medium is divided into two domains which are coupled by an interaction term for modeling flow and solute transport. In these models the porous medium is assumed to consist of two separate but connected continua. One continuum is associated with a network of fractures, fissures, macropores, or interggregate pores, while the second continuum involves the porous matrix blocks or soil aggregates. This kind of conceptual model can be called 'dual-domain', 'dual region', 'multi-region', etc., and the sub-regions can be two (e.g. Gerke and van Genuchten, 1993a) or more mobile regions (Gwo et al., 1995; Wu et al., 2004a) or one mobile region and one immobile region (Šimunek et al., 2003; Zimmerman et al., 1996).

The dual continuum concept has been commonly used to describe the preferential movement of water and solute at the macroscopic scale, a phenomenon that is widely believed to occur in most natural (undisturbed) media. Dual continuum models were introduced to simulate single-phase saturated (e.g. Barenblatt et al., 1960), variable saturated (Gerke and van Genuchten, 1993a; Gerke and van Genuchten, 1993b; Gerke and van Genuchten, 1996; Jarvis et al., 1991a; Jarvis et al., 1991b; Larsson and Jarvis, 1999; Lewandowska et al., 2004; Ray et al., 1997) and multiphase flow (Wu et al., 2004b; Zimmerman et al., 1996) in fractured rocks and structured soils. Most double scale models can be grouped into two types of Dual Continuum Models (DCM): the Dual Continuum Connected Matrix (DCCM) model and the Dual Continuum Disconnected Matrix (DCDM) model (Lichtner, 2000). A DCCM represents the original concept proposed by Barenblatt et al., (1960): the fracture network and matrix are distinct coexisting continua, and a coupling term provides the exchange of mass and heat between them. A DCDM refers to the conceptual model used by Warren and Root (1963) and Pruess and Narisimhan (1985), the matrix continuum is partially disconnected, with each matrix block connected to surrounding fractures but not to other matrix blocks. Liquid flow in the macro-porous domain is calculated with various calculation approaches involving different equations such as the kinematic wave equation (Germann and Beven, 1985; Greco, 2002), Richards equation (Gerke and van Genuchten, 1993a; 1993b; 1996; Jarvis et al., 1991a; 1991b; Larsson and Jarvis, 1999; Lewandowska et al., 2004; Ray et al., 1997) and the multiphase flow equation (Wu et al., 2004b; Zimmerman et al., 1996). Water flow in the micro-porous domain in DCCM can be described with an equation similar to that of the macro-porous domain. Sometimes, it can also be described with other equations. For example, Greco (2002) described water flow in macro-pores with a kinematic wave equation while Darcy equation was used to describe water flow in the soil matrix. In DCDM concept, a simple mass balance equation was used to describe moisture dynamics (Navarro, 1997; Zimmerman et al., 1996; Šimunek et al., 2003).

One of the most critical components of DCM is the water exchange term. In early papers the water transfer term was of linear first-order 'Warren-Root' type (Warren and Root, 1963). After that, modifications were made to 'Warren-Root' type water exchange term. The modified first order water transfer term could be driven by pressure head gradient such as that proposed by Gerke and Van Genuchten (1993a; 1993b) and subsequently used by Gwo et al., (1995) and Ray et al., (1997), or by fluid saturation as that indicated by Jarvis et al., (1991b), Larson and Jarvis (1999) and Saxena et al. (1994). As pointed by Zimmerman et al. (1996) and Köhne et al., (2004), the fracture/matrix flux predicted by the first-order transfer term is not accurate over all time scales and tends to underestimate the flux for early time. An improvement of this coupling term was suggested by Dykkuizen (1990) which consisted of two different equations applicable to early and later times. Zimmerman et al., (1993) integrated these two equations into one nonlinear ordinary differential equation and subsequently Zimmerman et al. (1996) modified it for variably saturated conditions. The second order water transfer term proposed by Zimmerman et al. (1996) was further modified by Köhne et al. (2004). The hydraulic conductivity in the water transfer term was calculated as a weighted average function of pressure head in the matrix and fracture. However, for the fine texture medium, the second order water transfer term tends to overestimate the flux at early time and underestimate it at later time (Köhne et al., 2004). The applicability of the second order water transfer term to the bentonite barrier needs further study.

The porous system of compacted bentonite exhibits different scales: Interlayer pores (2-10 Å), Intraaggregate pores (micropores 50 Å), Interaggregate pores (macropores 50-200 Å) (Bradbury and Baeyens, 2003; Fernández, 2003; Fernández et al., 2004). Double-porosity models (double-structure) has been used extentively for the study of Thermo-hydro-mechanical behaviour for compacted bentonite (Alonso et al., 1991; Gens et al., 1993; Guimarães, 2002; Sánchez et al., 2005). However, in these models, hydrodynamic equilibrium between macro-porous and micro-porous domains is assumed and the water transfer between two domains is neglected. Although bentonite can be assumed to be hydrodynamically similar to fractured rock in the sense of DCM, it has its own features. The most obvious is its extremely small scale, i.e. the characteristic length of both macro-pores and micro-pores is one or two orders of magnitude smaller than that of fractured rocks. Osmosis pressure plays a more important role in water transfer in clays than in fractured rock.

In this paper, the multiphase THG model of Zheng and Samper (2004) is extended to DCM. Two types of DCMs, the DCCM (dual continuum connected matrix) and DCDM (dual continuum disconnected matrix), are implemented. Liquid flow in macro-pores is described with a mass conservation equation which is composed of a Darcian flow term, terms accounting for chemical and thermal osmosis. In DCCM, the water flux in micro-pores is calculated by the same equation as that for macro-porous domain. In DCDM, a simple mass balance equation which consists of the mass variation term and the water exchange term is utilized to describe the moisture dynamics in micro-pores. A mixed type water exchange term is adopted, which includes a second order term accounting for water transfer due to the difference of liquid pressure and a second order term accounting for water transfer due to the gradient of chemical osmosis pressure. The hydraulic conductivity in water transfer term is evaluated as a weighted average function of pressure heads in the macro-porous and micro-porous domains. The scaling factor γ_w (see Gerke and van Genuchten, 1993a) and the weighting factor (see Köhne et al., 2004) are estimated with an inverse algorithm. Three mass conservation equations (for liquid, gas and heat) in macro-pores and one mass conservation equation for the liquid in micro-pores are solved wtih a Newton-Raphson method. All these features are implemented in a finite element code INVERS-FADES-CORE (Zheng and Samper 2004).

First, the conceptual DCM is discussed. After that, governing equations and their implementation in INVERS-FADES-CORE are presented.

2. Conceptual model

As pointed by Wilson et al. (1992) dividing soil pores into multiple pore regions is somewhat arbitrary. The division of porous medium into dual continua can either be intuitive like the fracture and matrix in fractured rocks or more quantitative such as the general concept adopted by Luxmoore et al. (1990) and Gwo, et al., (1995) in which macro-, meso- and micro-pores are distinguished by the equivalent pore diameter. In terms of numerical modelling, A medium can be double porosity or single porosity depending on the processes one wants to model and the physical heterogeneities. In order to facilitate the numerical modelling of thermal (T), hydraulic (H) and geochemical (G) behaviour of bentonite, we propose a dual continuum model in which the micro-porous domain includes interlayer space and intraaggregate pores and the the macro-porous includes the interaggregate pores (Figure 1), based on the following facts of: 1) Interlayer water has properties different with those of free water due to its more structured nature (Bradbury and Baeyens, 2003), 2) Local hydraulic and transport non-equilibrium between the interaggregate pores and interlayer spaces leads to transient transfer of water between these two domains (Alonso et al., 1991; Gens et al., 1993; Navarro, 1997), 3) Chemical non-equilibrium between double layer water and free water, (Fernández, 2003; Fernández et al., 2004), and 4) Interlayer space and intraaggregate pores are both inside aggregates.

In this conceptual model of dual porosity, the macro-porous domain holds 'free water' while the micro-porous domain contains interlayer water. The external surface of aggregates is the interface between macro-pores and micro-pores.

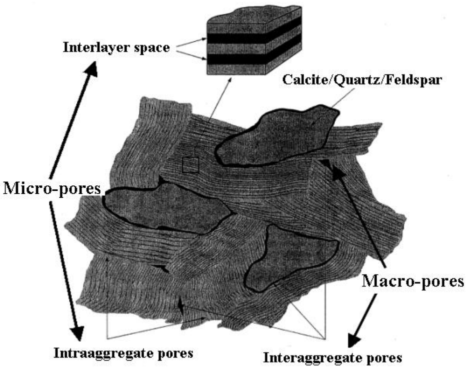

Fig. 1. Schematic representation of 'macro-pores' and 'micro-pores' in a dual continuum model (Bradbury and Baeyens, 2003).

3. Mathematical formulations

3.1. Description of processes

The processes considered for multiphase flow in dual continuum model include:

1) Coupled flow of liquid water under the hydraulic gradient, chemical and thermal osmosis gradient.
2) Flow of vapor (advection and diffusion)
3) Flow of gaseous species other than steam ('dry' air) (advection and diffusion)
4) Flow of air dissolved in the water (advection)
5) Transport of heat through the liquid and gaseous phases in both domains (convection) and the solid skeleton (conduction)

Here the dispersion of vapor and gaseous species are assumed to be negligible. The flux of water induced by chemical osmosis can be expressed by applying an analogue of Darcy's law modified by a reflection coefficient σ (Keijzer et al., 1999; Soler, 1999; Keijzer, 2000; Keijzer and Loch, 2001; Soler, 2001):

$$J_{co} = \rho\sigma K \frac{\partial \pi_h}{\partial x} \tag{1}$$

where J_{co} is the water flux induced by chemical osmosis (kg/m^2/s), ρ is the water density (kg/m^3), K is the hydraulic conductivity (m/s), and π_h is osmotic pressure head (m).

Thermal osmosis is also a coupled process that can produce a fluid flux. Zhou et al. (1999) showed that additional coupled flow terms due to a temperature gradient had significant effects on the distribution of capillary pressure and saturation degree in a THM model of a thick cylinder heating test. The flux of fluid caused by thermal osmosis v_{to} can be written as (Dirksen, 1969):

$$v_{to} = -k_T \frac{\partial T}{\partial x} \tag{2}$$

where T is temperature and k_T is the thermo-osmosis coefficient (m^2/K/s). Liquid flux caused by thermal osmosis term can be added to Darcian terms (Ghassemi and Diek, 2002; Zhou et al., 1999).

When both thermal and chemical osmosis are taken into account, the total flux of liquid has the classical Darcian terms plus a the chemical osmosis term and a thermal osmosis term:

$$\underline{q}_l = -\frac{k_{il}k_{rl}}{\mu_l}(\nabla P_l + \rho_l g \nabla z) + \sigma \frac{k_{il}k_{rl}}{\mu_l}\nabla \pi_h - k_T \nabla T \tag{3}$$

where k_{il} is the intrinsic permeability for liquid (m^2), k_{rl} is relative permeability, μ_l is the viscosity of liquid, P_l is liquid pressure (Pa).

The flux of gas q_g is expressed by Darcy's law:

$$\underline{q}_g = -\frac{k_{ig}k_{rg}}{\mu_g}(\nabla P_g + \rho_g g \nabla z) \tag{4}$$

where k_{ig} is the intrinsic permeability for gas (m^2), k_{rg} is the relative permeability for gas, μ_g is the viscosity of gas, P_g is the gas pressure (Pa).

The flow of vapor in the gaseous phase j_g^v is expressed by the generalized Fick's law:

$$\underline{\mathbf{j}}_{\mathbf{g}}^{\mathrm{v}} = -\underline{\underline{\mathbf{D}}}_{\mathbf{a}}^{\mathrm{v}} \nabla X_g^{\mathrm{v}} \tag{5}$$

where $\underline{\underline{\mathbf{D}}}_{\mathbf{a}}^{\mathrm{v}}$ is the diffusion coefficient and X_g^{v} is the mass fraction of vapor in the gaseous phase.

3.2. Mass balance equations

Our formulation is based on the THM formulation of Navarro (1997) in which the porous medium can be represented by a liquid phase (consisting of water and dissolved aqueous and gaseous species), a gaseous phase (which, in turn, is made of water vapor and 'dry' air, i.e. a fictitious species which encompasses all the gaseous species except water vapor) and the bulk solid. When the flow model is extended to dual continuum model, it is assumed that:

1) There is heat equilibrium between micro-porous and macro-porous domains.
2) There is gaseous phase equilibrium between micro-porous and macro-porous domains. As pointed by Zimmerman (1996), the air pressure gradients have a negligible effect on liquid imbibition.
3) Water in the micro-porous and macro-porous domains consists of water in liquid phase and vapor.

It should be noticed that the conceptual model of dual-domain (dual-porosity) proposed by Gerke and van Genuchten (1993a) is adopted in this paper. Total porosity is defined as the ratio of total pore volume V_p^{tot} to the total volume of medium V^{tot} i.e. $\phi^{tot} = V_p^{tot} / V^{tot}$. Local porosities for macro-porous and micro-porous domains are defined as $\phi^{ma} = V_p^{ma} / V^{ma}$ and $\phi^{mi} = V_p^{mi} / V^{mi}$ where V_p^{ma} and V_p^{mi} are the pore volumes of each domain and V^{ma} and V^{mi} are the total volumes of each domain. These porosity are related through:

$$\phi^{tot} = W^{ma}\phi^{ma} + (1 - W^{ma})\phi^{mi} \tag{6}$$

where W^{ma} is a volumetric weighting factor given by:

$$W^{ma} = {V^{ma}} / {V^{tot}} \tag{7}$$

The mass balance equation of water in the macro-porous domain is given by:

$$\frac{\partial\left(\phi^{ma} S_L^{ma} \rho_L^{ma} X_{wL}^{ma} + \phi^{ma} S_g^{ma} \rho_g^{ma} X_{vg}^{ma}\right)}{\partial t} = -\nabla\cdot\left(\rho_L^{ma} X_{wL}^{ma} \underline{\mathrm{q}}_L^{ma} + \rho_g^{ma} X_{vg}^{\mathrm{ma}} \underline{\mathbf{q}}_g^{ma} + \underline{\mathbf{j}}_{\mathbf{vg}}^{ma}\right) + r_w^{ma} - \frac{\rho_L^{ma}\Gamma_w}{W^{ma}} \tag{8}$$

where superscript '*ma*' denotes the macro-porous domain, S_L^{ma} is the degree of liquid saturation in macro-pores, ρ_L^{ma} is the liquid density, S_g^{ma} is the degree of gas saturation in macro-pores, ρ_g^{ma} is the gas density, X_{vg}^{ma} is the mass fraction of vapour in the gas in macro-pores and X_{wl}^{ma} is the mass fraction of water in the liquid. q_L^{ma} is the water flux given by Equation (3), q_g^{ma} is gas flux calculated by Equation (4), j_{vg}^{ma} the flow of vapor in the gaseous phase expressed by equation (5), r_w^{ma} is the source/sink term of water liquid phase and Γ_w is the water exchange terms between macro-porous and micro-porous domains.

The mass balance equation of water in micro-porous domain for DCCM model is given by:

$$\frac{\partial\left(\phi^{mi} S_L^{mi} \rho_L^{mi} X_{wL}^{mi} + \phi^{mi} S_g^{mi} \rho_g^{mi} X_{vg}^{mi}\right)}{\partial t} = -\nabla\cdot\left(\rho_L^{mi} \underline{\mathrm{q}}_L^{mi} + \rho_g^{mi} X_{vg}^{\mathrm{mi}} \underline{\mathbf{q}}_g^{\mathrm{mi}} + \underline{\mathbf{j}}_{\mathbf{vg}}^{mi}\right) + r_w^{mi} - \frac{\rho_L^{mi}\Gamma_w}{1 - W^{ma}} \tag{9}$$

The mass balance of water in the micro-porous domain for DCDM is given by:

$$\frac{\partial\left(\phi^{mi} S_L^{mi} \rho_L^{mi} X_{wL}^{mi}\right)}{\partial t} = \frac{\rho_L^{mi}\Gamma_w}{1 - W^{ma}} \tag{10}$$

where superscript '*mi*' denotes the micro-porous domain, S_L^{mi} is the degree of liquid saturation in micro-porous domain, ρ_L^{mi} is the liquid density in micro-porous domain, X_{wl}^{mi} is the mass fraction of water in the liquid. q_L^{mi} is the water flux given by Equation (3), r_w^{mi} is the source/sink term of water liquid phase.

The mass balance equation of air is given by:

$$\frac{\partial\left(\phi^{ma} S_L^{ma} \rho_l X_{al}^{ma} W^{ma} + \phi^{ma} S_g^{ma} \rho_g^{ma} X_{ag} W^{ma} + \phi^{mi} S_g^{mi} \rho_g^{mi} X_{ag} W^{mi}\right)}{\partial t} = -\nabla\cdot\left(\rho_L^{ma} X_{al}^{ma} \mathrm{q}_L^{ma} W^{ma} + \rho_g^{ma} X_{ag}^{ma} \mathrm{q}_g^{ma} W^{ma} + \rho_g^{mi} X_{ag}^{mi} \mathrm{q}_g^{mi} W^{mi} + \underline{\mathrm{j}}_{\mathrm{ag}}^{ma} + \underline{\mathrm{j}}_{\mathrm{ag}}^{mi}\right) + r^a \tag{11}$$

where X_{al}^{ma} is the mass fraction of air in the liquid phase (dissolved air), X_{ag}^{ma} and X_{ag}^{mi} are the mass fractions of air in the gaseous phase in the two domains respectively, S_g^{mi} is the gas saturation in micro-porous domain which is defined as $1 - S_L^{mi}$, S_g^{ma} is the gas saturation in macro-porous domain which is defined as $1 - S_L^{ma}$, j_{ag}^{ma} and j_{ag}^{mi} are the diffusive fluxes of air in gaseous phase in macro--porous and micro-porous domains respectively.

The heat equation is given by:

$$\frac{\partial}{\partial t}\begin{pmatrix} \rho_L^{ma}\phi^{ma} S_L^{ma} h_L W^{ma} + \rho_L^{mi}\phi^{mi} S_L^{mi} h_L W^{mi} + \\ \rho_v^{ma}\phi^{ma}(1 - S_L^{ma}) h_v W^{ma} + \rho_v^{mi}\phi^{mi}(1 - S_L^{mi}) h_v W^{mi} + \\ \rho_a\phi^{ma}(1 - S_L^{ma}) h_a W^{ma} + \rho_a\phi^{mi}(1 - S_L^{mi}) h_a W^{mi} + \\ \rho_a H\phi^{ma} S_L^{ma} h_a W^{ma} + \rho_s(1 - \phi^{ma} W^{ma} - \phi^{mi} W^{mi}) h_s \end{pmatrix} = -\nabla\cdot\left(\Lambda\cdot\nabla T\right) - \nabla\cdot\begin{pmatrix} \rho_L^{ma}\phi^{ma} S_L^{ma} h_L q_L^{ma} W^{ma} + \rho_L^{mi}\phi^{mi} S_L^{mi} h_L q_L^{mi} W^{mi} \\ +\rho_v^{ma}\phi^{ma}(1 - S_L^{ma}) h_v j_{vg}^{ma} W^{ma} \\ +\rho_v^{mi}\phi^{mi}(1 - S_L^{mi}) h_v j_{vg}^{mi} W^{mi} + \rho_a\phi^{ma}(1 - S_L^{ma}) h_a q_g^{mi} W^{ma} \\ +\rho_a\phi^{mi}(1 - S_L^{mi}) h_a q_g^{mi} W^{mi} + \rho_a H\phi^{ma} S_L^{ma} h_a q_L^{ma} \end{pmatrix} \tag{12}$$

where h is enthalpy, subscripts *L*, *v*, *a* and *s* denote liquid, vapour, air and solid, respectively, and *H* is the volumetric coefficient of solubility of dissolved air. The definitions of the rest of variables is that given for Equations (8) to (11).

Horseman et al. (1996) proposed that highly compacted bentonite can function as an efficient semi-permeable membrane. Due to that macro-micro mass transfer will be largely determined by micro-structure (Wu et al., 2004b). Transport of solvent (water) by osmosis may play an important role in water transfer if significant geochemical non-equilibrium exists.

Thus, in this paper, based on the theory of membrane process and the second order water transfer terms proposed by Kohne et al. (2004), we propose the following expression of macro-micro water transfer Γ_w (s^{-1}):

$$\Gamma_w = \frac{a_w^*\bar{K}_{il}\bar{K}_{rl}}{\mu}\frac{(P_L^{ma} - P_L^{mi})\left[\left|P_L^{mi} - P_L^{ini}\right| + \left|P_L^{ma} - P_L^{ini}\right|\right]}{\left|P_L^{mi} - P_L^{ini}\right|} - \frac{a_w^*\bar{\sigma}\bar{K}_{il}\bar{K}_{rl}}{\mu}\frac{(\pi_h^{ma} - \pi_h^{mi})\left[\left|\pi_h^{mi} - \pi_h^{ini}\right| + \left|\pi_h^{ma} - \pi_h^{ini}\right|\right]}{\left|\pi_h^{mi} - \pi_h^{ini}\right|} \tag{13}$$

where P_L^{ma} and P_L^{mi} are the liquid pressures in macro-porous and micro-porous domains, respectively, P_L^{ini} is the initial (time is equal to zero) liquid pressure in the micro-porous domain, π_h^{ma} and π_h^{mi} are the osmotic pressure heads in macro-porous and micro-porous domains, respectively, π_h^{ini} is the initial osmotic pressure head in micro-porous domain. $\overline{\sigma}$ is the effective reflection coefficient; $\overline{K}_{il}$ and $\overline{K}_{rl}$ are the effective relative liquid permeability and intrinsic liquid permeability, respectively. Coefficient a_w^* is given by Gerke and van Genuchten (1993a):

$$a_w^* = \frac{\beta}{\alpha^2}\gamma_w \tag{14}$$

where β is a geometry-dependent coefficient and α is the radius for spheres (or cylinders) or the half width for cubes (m), and γ_w is a dimensionless scaling term. Gerke and van Genuchten (1993a) indicated that Equation (14) was derived by comparing Laplace transforms of first order water transfer term with those of the linearized horizontal flow equation. However, as mentioned by Gerke and van Genuchten (1993b), one could consider the geometry coefficient β or better perhaps the entire term a_w^* as an essentially empirical quantity which must be calibrated from observed field data.

Kohne et al. (2004) evaluated different expressions of the effective hydraulic conductivity, such as arithmetic, geometric, integral and weighted arithmetic averages, and proved that the weighted arithmetic average led to the best approximation for the water transfer. Therefore, the weighted arithmetic average is adopted for $\overline{\sigma}$, $\overline{K}_{il}$ and $\overline{K}_{rl}$:

$$\sigma = (w\sigma^{mi} + \sigma^{ma})/(w+1) \tag{15}$$

$$\overline{K}_{il} = (\overline{w}K_{il}^{mi}(P_l^{mi}) + K_{il}^{ma}(P_l^{ma}))/(w+1) \tag{16}$$

$$\overline{K}_{rl} = (wK_{rl}^{mi}(P_l^{mi}) + K_{rl}^{ma}(P_l^{ma}))/(w+1) \qquad 17)$$

where W is an emperical weighting factor. As pointed by Kohne et al. (2004), 'the value of the empirical weighting factor *w* most likely cannot be derived theoretically'. They obtained a value of 17 for *w* for silty clay and silt, but obviously this value is not applicable to bentonite.

It should be mentioned that the code INVERSE-FADES-CORE is capable of solving both inverse and forward non-isothermal multiphase flow problems. The inverse problem is solved by minimizing a generalized least-squares criterion with a Gauss-Newton-Levenberg-Marquardt method (Zheng and Samper, 2004). For details on the inverse algorithm the reader should check Dai (2000) and Dai and Samper (2004). Applications of this inverse algorithm to non-isothermal multiphase flow are presented by Zheng and Samper (2004). The empirical coefficient a_w^* and weighting factor in Equations (15) to (17) can be obtained by inverse modelling of a well-controlled experiment for bentonite such as the 'mock up' test (ENRESA, 2000) .

4. Numerical implementation

The previouss mathematical formulation has implemented in the a finite element code INVERS-FADES-CORE which enables the simultaneous inverse and forward modeling of non-isothermal multiphase flow, reactive solutes transport in one, two dimensions and three-dimensional with axial symmetry in both single and dual continuum. In INVERS-FADES-CORE, the state variables are liquid pressure in macro-porous P_L^{ma} , gas pressure P_g, temperature *T*, liquid pressure in macro-porous P_L^{mi}, concentration of species *j* in macro-porous C_j^{ma} and concentration of species *j* in micro-porous C_j^{mi} . The non-linear equation system (Equations (8) to (12)) are solved simultaneously with the Newton Raphson method to obtain P_L^{ma} , P_g, *T* and P_L^{mi} . Then reactive transport equations are solved by the sequential iteration method to obtain C_j^{ma} and C_j^{mi} . In order to simplify the algorithm, when chemical osmosis is considered, the chemical osmotic pressure is calculated using concentration at the previous time step.

The methodology for solving the inverse problem of non-isothermal multiphase flow and multicomponent reactive solute transport is described in Zheng and Samper (2004). The formulation presented here is tested with synthetic cases and applied to a real experiment in the second part of this series of two papers (see Zheng and Samper, 2005).

5. Conclusions

Experimental studies indicate that bentonite exhibits different porosity scales, which calls for the necessity of using dual-domain model. A second order water transfer term which accounts for both water transfer by hydraulic pressure gradient and osmotic pressure gradient has been implemented in a dual-domain coupled non-isothermal multiphase flow model.

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