

A NON-ISOTHERMAL TWO-PHASE FLOW MODEL FOR COMPRESSIBLE, SUPERCRITICAL FLUIDS IN POROUS MEDIA

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Summary. This work presents a two-phase flow model for compressible fluids coupled with heat transport simulation. The coupling is realized via equations of state, which are used to determine the relation between temperature, pressure and density of a fluid. The density correlation is necessary to describe the compressibility of a gas or a liquid, furthermore, density together with temperature affects fluid viscosity and thermal conductivity. Using this coupled H²T-model, we developed a test scenario based on an enhanced gas recovery application. This test case consists of a 1D cutout from a natural gas reservoir in the vicinity of the CO₂ injection well. The simulation shows the displacement of natural gas by carbon dioxide at the first stage of injection. A simplified simulation with constant fluid properties but similar conditions was compared to an analytical solution to prove the validity of the governing equations.

1 INTRODUCTION

Simulation of non-isothermal two-phase flow (H²T) has a wide range of applications in different disciplines like soil science or chemical, environmental and petroleum industries. Today, numerical implementation of multiphase flow processes is essential in nuclear waste

disposal, geothermal, or groundwater remediation applications. Typical two-phase flow problems are liquid-gas or water-NAPL systems.

If pressure and temperature within a system are high enough (typical in geological formations), the fluids in the system may exist in a supercritical state of aggregation. In this state, the fluid properties show both liquid and gaseous behaviour: Fluids may be immiscible and are compressible due to pressure changes. So, simulation of supercritical fluid flow is not only important for geological applications such as carbon dioxide (CO₂)-capture and storage (CCS), enhanced oil or gas recovery (EOR, EGR) or enhanced coalbed methane recovery (ECMR), but also for industrial applications related to supercritical fluid extraction.

The critical point of a fluid specifies the temperature and pressure conditions, when its state of aggregation cannot be distinguished between liquid or gaseous. At this state, the fluid has properties from both liquids and gases.¹

The motive for developing an H²T model for supercritical fluids was to simulate the CO₂ storage process within an EGR application. In this project, a natural gas which is assumed to be pure methane is going to be displaced by carbon dioxide. Both fluids are considered to be immiscible and to be in supercritical state. Because injected and residing fluids have different temperatures, the process is considered to be non-isothermal. Due to cooling and heating processes, the fluid volumina (and fluid mobilities) are changing. Unlike liquids, the volume change of a supercritical fluid due to pressure variations is not negligible. This compressibility of a fluid can be determined by equations of state (EOS). In this work, we use EOS based on the *Helmholtz* free energy.^{2,3} Fluid transport properties as function of density and temperature are determined by highly accurate correlations designed for carbon dioxide^{4,5} and for methane⁶ individually.

Several works have been published dealing with supercritical fluid flow in porous media. However, this works are constrained to monophasic and isothermal flow^{7,8} or have experimental emphasis.^{9,10,11}

2 GOVERNING EQUATIONS

2.1 Balance equations

Multiphase heat and mass transfer in porous media can be computed by solving the balances of heat and mass at each point of the investigated domain. These continuity equations are partial differential equations (PDEs), which can only be solved analytically for very simplified conditions. In complex geometries, PDEs can be solved numerically by partitioning the regarded system into a finite number of representative elementary volumes (REV). In this work, pore space is considered to be constant due to neglect of mechanical deformation or precipitation of solid matter. When source or sink terms are avoided, the content of a phase α within an REV can be expressed as the change of its

saturation S_α and its density ρ_α over time t :

$$n \frac{\partial S_\alpha \rho_\alpha}{\partial t} = \text{div} (\rho_\alpha \lambda_\alpha \mathbf{K} \{ \nabla p_\alpha - \rho \mathbf{g} \}) \quad (1)$$

where n is porosity, λ_α is mobility (5), p is phase pressure, \mathbf{g} is vector of gravity, and \mathbf{K} is the intrinsic permeability tensor. The use of this formulation of the continuity equation has the advantage that capillary pressure is easy to handle since it is no primary variable.¹² Capillary pressure is the pressure difference at the interphase of both phases and is defined by (8). In (1), primary variables are saturation S and pressure p . The changing of density of a fluid can be forced by variation of pressure or temperature conditions according to

$$\frac{\partial \rho_\alpha}{\partial t} = \frac{\partial \rho_\alpha}{\partial p_\alpha} \frac{\partial p_\alpha}{\partial t} + \frac{\partial \rho_\alpha}{\partial T} \frac{\partial T}{\partial t} \quad (2)$$

In case of a liquid, the pressure based compressibility is negligible. But, fluids in supercritical state of aggregation behave more like gases, so pressure changes play an important role on the fluid density. In this case, an equation of state has to be used to describe the relation between pressure, temperature and density.

2.2 Equation of state

The EOS used in this work are semi-empirical fundamental equations based on the *Helmholtz* free energy. This kind of EOS uses a theoretical approach, which is correlated by a multitude of measurement data to a specific fluid. This correlation was developed by *Span and Wagner* (1996)² for carbon dioxide and by *Setzmann and Wagner* (1991)³ for methane. By the use of precise EOS, it is possible to determine the compressibility of a fluid due to pressure and temperature changes. The *Helmholtz* free energy in its dimensionless formulation,

$$\phi(\delta, \tau) = \phi^o(\delta, \tau) + \phi^r(\delta, \tau), \quad (3)$$

consists of an ideal part, ϕ^o and a residual part, ϕ^r , which describes the deviations of the real gas from ideal gas behaviour. In its dimensionless form, the function arguments density and temperature are given as normalized quantities. The normalization is performed with respect to critical values, so that $\delta = \rho/\rho_c$ and $\tau = T_c/T$. From (3), all thermodynamic properties of a fluid can be determined. The relation between pressure and *Helmholtz* free energy is given by

$$p(\delta, \tau) = \left(1 + \delta \frac{\partial \phi^r}{\partial \delta} \right) \rho RT \quad (4)$$

(4) can not be solved algebraically for density, because the term $\partial \phi^r / \partial \delta$ is a function of density as well. So, it has to be solved by an iterative approximation. The *van Wijngaarden-Dekker-Brent*¹³ method turned out to be the most applicable approach. This root-finding approach guarantees convergence as long as the values of the function are computable within a given region containing a root.

2.3 Constitutive equations

The mobility λ_α of phase α is the relation between phase-permeability k_α and phase-viscosity μ_α . Considering non-isothermal, compressible multiphase flow problems, λ_α is a function of phase saturation, temperature and pressure:

$$\lambda_\alpha = -\frac{k_{r\alpha}(S_\alpha)}{\mu_\alpha(p, T)} \quad (5)$$

In general, the phases of a two-phase system are indicated as wetting phase $\alpha = w$ and non-wetting phase $\alpha = n$. In this paper, the polar carbon dioxide is assumed to be the wetting phase, while nonpolar methane is regarded as non-wetting phase. Relative permeabilities for wetting and non-wetting phase are described by the *Brooks&Corey*¹⁴ model:

$$k_{r,w}(S_w) = S_e^{2+3\lambda^*}, \quad k_{r,nw}(S_w) = (1 - S_e)^2(1 - S_e^{2+\lambda^*}) \quad (6)$$

where S_e is effective saturation and λ^* is a soil distribution parameter. In both cases, the wetting-phase saturation represents the independent variable of (6). The effective saturation S_e is depending on maximal saturation S_{\max} and residual saturation $S_{r,w}$ of the wetting phase and is defined as

$$S_e = \frac{S_w - S_{r,w}}{S_{\max} - S_{r,w}} \quad (7)$$

At the interphase between wetting and non-wetting phase, a pressure difference known as capillary pressure p_c can also be estimated by the *Brooks&Corey* model:

$$p_c = S_e^{-1/\lambda^*} \cdot p_0 \quad (8)$$

where p_0 is the characteristic entry pressure. The transport properties (viscosity and thermal conductivity) of fluids can be determined by correlation functions depending on temperature and density. These empirical functions were adjusted by measurement data for each fluid individually and they consist of three terms, describing the respective fluid property at different density regions:

$$X(\rho, T) = X_0(T) + X_1(\rho, T) + \Delta X_c(\rho, T) \quad (9)$$

where X is either viscosity η or thermal conductivity λ . $X_0(T)$ is valid for the dilute gas region, $X_1(\rho, T)$ for the moderately dense gas region, and $\Delta X_c(\rho, T)$ is an enhancement term needed close to the critical point. A correlation function for viscosity of carbon dioxide was developed by *Fenghour* (1998).⁵ This viscosity representation covers the temperature range $200 \text{ K} \leq T \leq 1500 \text{ K}$ and pressures up to 300 MPa. The thermal conductivity of CO_2 was determined by the contribution of *Vesovic* (1990).⁶ This function provides valid thermal conductivities between $200 \text{ K} \leq T \leq 1500 \text{ K}$ and for pressures up to 100 MPa. For methane, *Friend* (1989)⁴ presented a correlation for viscosity and thermal conductivity in the range from about 91 K to 600 K and for pressures below 100 MPa.

3 NUMERICAL MODELLING

The developed H²T model, equations of state for CO₂ and CH₄ as well as the respective constitutive equations were implemented into the scientific open source code OpenGeoSys (OGS).¹⁵ Using OGS, the model setup was tested by a simplified enhanced gas recovery scenario simulation. In this case, the model domain was reduced to a 1D cutout of the natural gas reservoir in a depth of about 3500 m. This cutout consists of homogeneous material and has a length of 5 m, starting from the injection well on the left side (see Figure 1). The wetting behaviour of the sandstone material is described by the *Brooks&Corey* model using a soil distribution index of $\lambda^* = 2$. Relative permeability and capillary pressure are given by (6) and (8). An overview of all used soil parameters is given in Table 1.

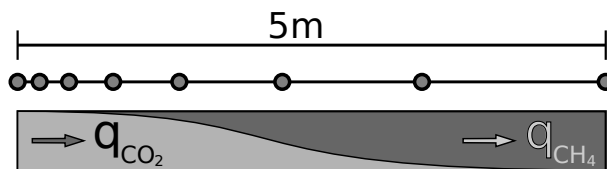


Figure 1: Model domain of homogeneous 1D test scenario

Table 1: Solid matrix properties and constitutive relationships

parameter	value	parameter	value
absolute permeability \mathbf{K}	10^{-10} m^2	residual saturation S_{CH_4}	0 [-]
porosity n	0.03 [-]	relative permeability k_{r,CO_2}	eq. (6)
distribution index λ^*	2 [-]	relative permeability k_{r,CH_4}	eq. (6)
entry pressure p_0	5000 Pa	capillary pressure p_c	eq. (8)
residual saturation S_{CO_2}	0 [-]		

The size of the model domain is chosen such that the right boundary will not be influenced by CO₂ injection within the simulation time. This scenario shows the very beginning of an EGR process: at time $t = 0$, the reservoir is filled completely with a natural gas mixture, represented in this case by an amount of 99% of methane in supercritical state and 1% carbon dioxide. As mentioned in sec. 2.1, there are no source terms controlling the CO₂ storage process. When the injection starts, the CO₂ inflow at the left model boundary is forced through *Dirichlet* boundary conditions ($S_{\text{CH}_4} = 0$, $p_{\text{CO}_2} = 7.5 \text{ MPa}$ and $T = 300 \text{ K}$). Due to the constant injection, carbon dioxide displaces the residing natural gas from

left to right. At the injection point, the gradient of the CO_2 saturation curve is very strong at the beginning of the simulation, so the spatial discretisation in this region needs to be very fine.

4 RESULTS

Non-isothermal, compressible multi-phase flow in porous media can not be calculated algebraically, so there is no exact solution which could be compared directly to the simulation results of the presented work. Available analytical solutions are constrained to isothermal conditions and handle with incompressible fluids. One of those approaches is given by *McWhorther*, (1990)^{16, 17} where exact integral solutions for horizontal, steady-state flow of immiscible, viscous fluids in porous media is presented. A direct comparison between *McWhorthers* solution and our simulation results is possible, if all thermodynamic and transport properties of the involved fluids are set to constant values. Such an comparison is shown in Figure 2(a). Both analytical and numerical results are based on identical material properties shown in Table 1. The plot shows the CO_2 saturation profile along the model coordinate x at three points in time. At each time, the numerical simulation result shows excellent agreement to the exact solutions, which indicates that the chosen pressure-saturation scheme of governing equations is applicable for this purpose.

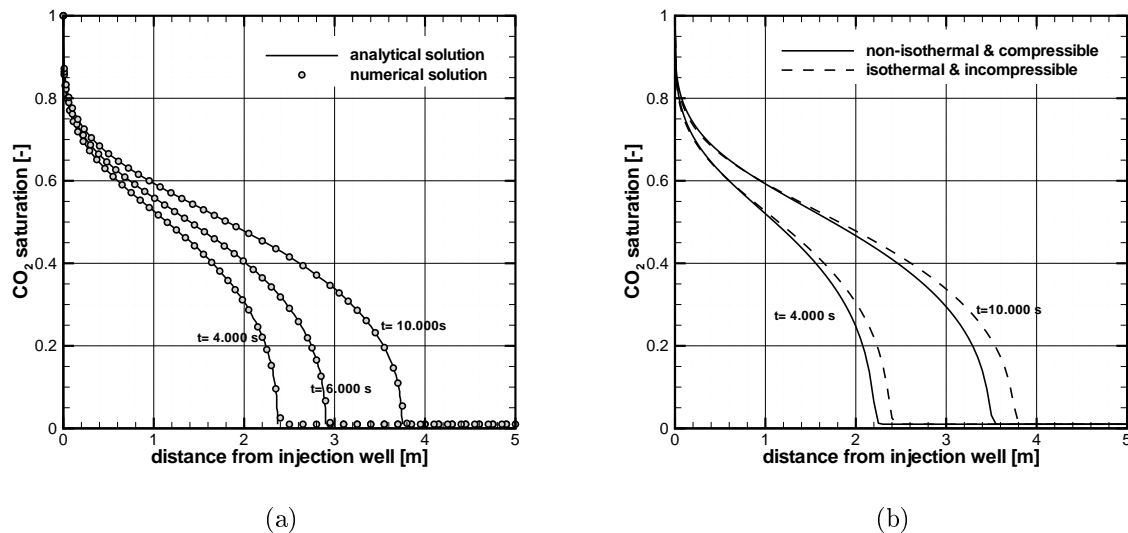


Figure 2: Comparison of (a) numerical simulation and analytical solution of isothermal, incompressible two phase flow of CO_2 and CH_4 and (b) results for isothermal, incompressible (dashed lines) as well as non-isothermal, compressible (solid lines) behaviour.

5 CONCLUSION

In comparison to isothermal and incompressible fluid flow, the developed model for the flow of supercritical fluids shows a slower CO₂ migration within the natural gas reservoir, see Fig. 2(b). The difference between both model approaches has two reasons. Due to the considered compressibility, CO₂ volume is decreasing during the injection, which needs additional energy. The other reason is the decreasing CO₂ mobility due to growing temperatures. Unlike gases, the viscosity of liquids and supercritical fluids increases with growing temperatures. In the isothermal case (dashed lines in Fig. 2(b)), the fluid mobility does not change with growing temperature. So, the CO₂-migration velocity (and the CO₂ storage capacity of the reservoir) is overestimated. This shows the importance of considering non-isothermal effects and compressibility terms in multiphase flow simulations of supercritical fluids.

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