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NON-ISOTHERMAL COMPOSITIONAL GAS FLOW DURING CARBON DIOXIDE STORAGE AND ENHANCED GAS RECOVERY

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Abstract. In this work we present the conceptual modeling and the numerical scheme for carbon dioxide storage into nearly depleted gas reservoirs for enhanced gas recovery reasons. For this we develop non-isothermal compositional gas flow model. We used a combined monolithic / staggered coupling scheme to solve mass balance equation for the gaseous mixture with heat and fractional mass transport equations. Temperature change resulting from fluid expansion and viscous heat dissipation is included in heat transport in addition to advection and conduction. We have used a modified version of the Peng-Robinson equations of state, to determine the density of the real gas mixture along with an empirically extended ideal gas equation. A real behavior of mixture is accounted by using energy and distance parameters.

1 INTRODUCTION

Fossil fuels continue to be the main global energy source. Therefore, squeezing the gas from matured fields will be of positive interest and additionally offering space for injection and storage of CO_2. Sequestration of CO_2 is at present of much importance because CO_2 has been identified to be one of the main greenhouse gases which can potentially lead to global climate change. Nearly depleted natural gas reservoirs are good candidates for storing CO_2 and at the same time recovering the rest gas found in these reservoirs. They offer several advantages for EGR processes namely integrity against gas escape, larger volume capacities, a good reservoir history enhancing its understanding and existing infrastructure. Numerical simulation studies have been performed showing the technical feasibility of carbon sequestration and enhanced gas recovery (CSEGR) with existing commercial codes (Pusch [1], Oldenburg [2]).

According to Holloway [3] underground storage of industrial quantities of carbon dioxide is technologically feasible. However, combustion of coal produces more carbon dioxide than could be stored in the space from which the coal came from [4, 5]. If one wishes to dispose of carbon dioxide permanently, it has to be confined within depleted hydrocarbon reservoirs or trapped in an aquifer directly analogous to a hydrocarbon reservoir [6]. Hydrocarbon reservoirs have many advantages over aquifers. Firstly, the reservoirs have a seal that has proven capable of retaining liquids or gases for thousands to millions of years. Secondly, some of the equipment installed on the surface or underground for oil or gas recovery may be re-used for carbon dioxide disposal. The underground reservoir needs a sealed trap to prevent the carbon dioxide from reaching the earth's surface due to percolation upwards through inhabitant ground water. This is because supercritical or liquid carbon dioxide is lighter than water and tends to force its way up through the water-saturated rocks.

Starting from appropriate local formulations expressing the balance laws of continuum mechanics, we develop the governing equations for non-isothermal miscible displacement of compressible mixture of nonpolar gases through a non-deformable porous medium. The compressibility of the gaseous mixture is again defined according to the Peng and Robinson [7] equation of state. The mass balance equations are then written to describe the macroscopic behavior of the gaseous mixture with heat transport. The energy conservation equation is solved to account for heat loss due to gas expansion and viscous heat dissipation along with advection and conduction. Macroscopic fractional mass transport equations are then obtained for each component of the mixture. Equation of state for density and viscosity is introduced to obtain the general field variables. We select gaseous mixture pressure p, temperature T and mass-fraction of each component x_k as primary field variables.

A numerical module has been developed for compositional gas flow coupled with heat and fractional mass transport. The finite element module is embedded in the object-oriented framework of the scientific open source code OpenGeoSys ([8, 9]). Therefore, allows an easy extension of the numerical model for multi-dimensional problems. A combined, monolithic and staggered coupling scheme is used (i.e., monolithic for the pressure and temperature fields and staggered for the component mass-fraction) with variable time stepping.

2 GOVERNING EQUATIONS

The differential form of the mass balance equation for the k$^{\mathrm{th}}$ component of the gaseous mixture is

$$\frac{\partial\left(n\rho_{\mathrm{k}}\right)}{\partial t}+\nabla\cdot\left[\mathbf{J}_{\mathrm{FA}_{\mathrm{k}}}+\mathbf{J}_{\mathrm{FD}_{\mathrm{k}}}\right]=Q_{\rho_{\mathrm{k}}} \tag{1}$$

Here, advective mass-flux (mass rate per unit flow area) of the k^{th} component is $\mathbf{J}_{\mathrm{FA_k}} = \rho_{\mathrm{k}} n\mathbf{v}$. In presence of concentration gradients, diffusive mass-flux (mass rate per unit flow area) of the k^{th} component w.r.t. $\mathbf{v}$ (velocity of center of the gravity) is $\mathbf{J}_{\mathrm{FD_k}} = x_{\mathrm{k}}\rho n(\mathbf{v}_{\mathrm{k}} - \mathbf{v})$ where $\mathbf{v}_{\mathrm{k}}$ is the average velocity of the k^{th} component. It is clear from this relationship that sum of diffusive flux over all components (total diffusive mass-flux) is zero. Density ρ, is defined as mass of gas per unit volume which varies with pressure and temperature. $Q_{\rho_{\mathrm{k}}}$ is an external source/sink term due to the k^{th} component.

The linear momentum balance equation for the mixture can be expressed in the form of the extended Darcy's law. This is a reasonable approximation of slow laminar flow arising in reservoir engineering problems. According to Darcy's law, the advective part of the total mass-flux due to all components is $\mathbf{J}_{\mathrm{FA}} = \rho n\mathbf{v}$ and can be written as

$$\rho n\mathbf{v} = -\rho\frac{\mathbf{k}}{\mu}(\nabla p - \rho\mathbf{g}); \text{where } \mathbf{v} = \sum_{\mathrm{k}} x_{\mathrm{k}}\mathbf{v}_{\mathrm{k}}. \tag{2}$$

Where $\mathbf{k}$ is intrinsic permeability, μ is mixture dynamic viscosity and $\mathbf{g}$ is gravity constant. Summarizing the mass balance Eq. (1) corresponding to each component results in the mass balance equation for the gaseous mixture,

$$\frac{\partial(n\rho)}{\partial t} + \nabla\cdot(\rho n\mathbf{v}) = Q_\rho; \text{where } \ \rho = \sum_{\mathrm{k}} \rho_{\mathrm{k}}. \tag{3}$$

Here, ρ is calculated by the extended ideal gas equation which is given by

$$\rho = \frac{pM}{\mathrm{z_{sc}}RT} \tag{4}$$

where M is the molecular weight of the gaseous mixture, R is the universal gas constant; the mixture super compressibility factor $\mathrm{z_{sc}}$ is a non-dimensional constant which varies with pressure, temperature and composition.

Following the assumption of local thermal equilibrium, the energy balance equation of porous media can be obtained as

$$\begin{aligned} &(\rho c_p)_{\mathrm{eff}}\frac{\partial T}{\partial t} + c_p\rho n\mathbf{v}\cdot\nabla T - \nabla\cdot[\kappa_{\mathrm{eff}}\nabla T] = \\ &\underbrace{n\beta_{\mathrm{T}}T\frac{\partial p}{\partial t} + \beta_{\mathrm{T}}Tn\mathbf{v}\cdot\nabla p}_{JTC} - \underbrace{n\mathbf{v}\cdot\nabla p}_{VHD} + Q_T \end{aligned} \tag{5}$$

The 1^{st} term of the right hand side is related to Jule-Thomson Cooling (JTC) and the 2^{nd} term is related to Viscous Heat Dissipation (VHD). where κ_{eff}, is the effective thermal conductivity tensor of the porous media, with coordinates defined as $(\kappa_{\mathrm{eff}})_{ij} = (1-n)\kappa^s_{ij} + n\kappa_{ij}$. κ is the thermal conductivity of the gaseous mixture. $(\rho c_p)_{\mathrm{eff}}$ is the effective heat capacity of the porous medium defined by $(\rho c_p)_{\text{eff}} = (1-n)\rho^s c_p^s + n\rho c_p$. Here, specific heat capacity and thermal conductivity of the gaseous mixture are approximated by averaging over its components.

Analogous to the derivation of the mass balance equation, the transport equation results from the balancing of each component's mass. The characteristic quantity for mass transport is $n\rho x_{\text{k}}$ which is conserved by definition. With macroscopic dispersion, we obtain the mass transport equation for each component in the porous medium,

$$\frac{\partial \left[n\rho x_{\text{k}}\right]}{\partial t} + \nabla \cdot \mathbf{J}_{\text{MA}_{\text{k}}} + \nabla \cdot \mathbf{J}_{\text{MD}_{\text{k}}} = Q_{\text{x}_{\text{k}}}. \tag{6}$$

where x_{k} represents the mass-fraction of the k^{th} component of the gaseous mixture. $Q_{\text{x}_{\text{k}}}$ is the mass source/ sink term. Advective and diffusive mass-fluxes of the k^{th} components are $\mathbf{J}_{\text{MA}_{\text{k}}} = -(x_{\text{k}}\rho\mathbf{k}/\mu)(\nabla p - \rho\mathbf{g})$ and $\mathbf{J}_{\text{MD}_{\text{k}}} = -n\tau\mathbf{D}_{\text{k}}\nabla\left(\rho x_{\text{k}}\right)$, respectively.

Using the componential mass balance Eq. (1) with mass transport Eq. (6), the divergent form of the fractional-mass transport equation is given by

$$n\rho\frac{\partial x_{\text{k}}}{\partial t} + \rho n\mathbf{v} \cdot \nabla x_{\text{k}} + x_{\text{k}}Q_{\rho_{\text{k}}} - \nabla \cdot \left[n\rho\mathbb{D}\nabla x_{\text{k}}\right] = Q_{\text{x}_{\text{k}}}. \tag{7}$$

3 CONSTITUTIVE RELATIONS

Density and viscosity of the carbon dioxide change abruptly as it passes through critical pressure near critical temperature. Here, we describe the procedure which we have used to calculate the mixture density and viscosity for the necessary range of pressure and temperature.

In reservoir engineering, the empirically extended ideal gas law is well known. Oldenburg et al.[2] have used this in their numerical model for CSEGR. Here, the degree of non-ideality shown by a gaseous mixture can be expressed by the super compressibility factor z_{sc}. For the gaseous mixture z_{sc} is obtained by solving constitutive equations, which are, e.g., derived by inserting $\text{z}_{\text{sc}} = pV/(N_{\text{n}}RT)$ in the modified Peng-Robinson equation of state (PR-EOS). For the gaseous mixture, PR-EOS needs critical constants and the acentric factor to get the attraction $a(T)$ and repulsion b parameters values. The critical pressure and temperature of the mixture are approximated as

$$p^{\text{cr}} = M\sum_{\text{k}} x_{\text{k}}M_{\text{k}}^{-1}p_{\text{k}}^{cr}; \quad T^{\text{cr}} = M\sum_{\text{k}} x_{\text{k}}M_{\text{k}}^{-1}T_{\text{k}}^{cr} \tag{8}$$

As we have values of $a(T)$ and b, we can solve the following cubic equation analytically.

$$\text{z}_{\text{sc}}^3 + \text{z}_{\text{sc}}^2(B-1) + \text{z}_{\text{sc}}(A - 3B^2 - 2B) + (B^3 - B^2 - AB) = 0 \tag{9}$$

where, $A = p\,a(T)/(RT)^2$ and $B = p\,b/(RT)$. The parameters $a(T)$ and b are calculated from empirical relations

$$a(T) = 0.4572\frac{R^2T^{\text{cr}2}}{p^{\text{cr}}}\left[1 + a_0\left(1 - \sqrt{\frac{T}{T^{\text{cr}}}}\right)\right]^2; \quad b = 0.07779\frac{RT^{\text{cr}}}{p^{\text{cr}}} \tag{10}$$

where, $a_0 = 0.3746 + 1.5422\omega - 0.2699\omega^2$; ω is the acentric factor of the gaseous mixture. $a(T)$ accounts for attractive forces between the molecules of the mixture and b takes into account the reduction in free volume of the particular molecule.

The above cubic equation can have three positive roots, with the largest positive root corresponding to the gaseous mixture. To guarantee the single phase i.e., gas phase we choose the reservoir temperature 393.15 K, which is greater than the critical temperature of any component.

The dynamic viscosity of the gaseous mixture is calculated from a temperature and density dependent empirically correlated function. This predicts the viscosity for non-polar gases with an error of $\leq 2\%$.

$$\mu(\rho, T) = d_0\left(\frac{1}{G_2} + A_6Y\right) + f_0A_7Y^2G_2\exp\left(A_8 + \frac{A_9\epsilon}{kT} + \frac{A_{10}\epsilon^2}{k^2T^2}\right) \tag{11}$$

where

$$G_2 = \frac{\frac{A_1}{Y}(1 - e^{-A_4Y}) + A_2G_1e^{A_5Y} + A_3G_1}{A_1A_4 + A_2 + A_3};\; G_1 = \frac{1 - 0.5Y}{(1-Y)^3}\; Y = 1.0\times 10^{-3}\frac{\rho V^{\mathrm{cr}}}{M};$$

$$d_0 = 4.0785\times 10^{-7}\frac{(MT)^{\frac{1}{2}}}{V^{\mathrm{cr}\frac{2}{3}}\Omega}(1 - 0.2756\omega);\; f_0 = 3.6344\times 10^{-7}\frac{(MT)^{\frac{1}{2}}}{V^{\mathrm{cr}\frac{2}{3}}}$$

where k is the Boltzmann constant and the reduced collision integral Ω depends on the intermolecular potential. Empirical equation for Ω and constants $A_0 - A_{10}$ are given in [10]. The acentric factor and the molecular weight of the mixture are calculated by

$$\omega = \frac{1}{\sigma^3}\sum_i\sum_j x_ix_j\left(\omega_i\omega_j\right)^{\frac{1}{2}}\left(\sigma_i\sigma_j\right)^{\frac{3}{2}} \tag{12}$$

and

$$M^{\frac{1}{2}} = \frac{1}{\epsilon\sigma^2}\sum_i\sum_j x_ix_j\sigma_i\sigma_j\left(\epsilon_i\epsilon_j\right)^{\frac{1}{2}}\left(\frac{2M_iM_j}{M_i + M_j}\right)^{\frac{1}{2}}. \tag{13}$$

Here, i and j stand for the i^{th} and j^{th} components.

The parameters of the intermolecular potential for the gaseous mixture, σ and ϵ, are approximated by averaging over its components.

$$\sigma^3 = \sum_i\sum_j x_ix_j\left(\sigma_i\sigma_j\right)^{\frac{3}{2}};\quad \epsilon = \frac{1}{\sigma^3}\sum_i\sum_j x_ix_j\left(\sigma_i\sigma_j\right)^{\frac{3}{2}}\left(\epsilon_i\epsilon_j\right)^{\frac{1}{2}} \tag{14}$$

σ and ϵ are the parameters of the intermolecular potential describing the interaction between molecules of components of the mixture and are expressed, respectively, in Angstrom units and degrees Kelvin. An approximation of these two constants for a respective component is provided by $\epsilon_{\mathrm{k}} = 0.77kT_{\mathrm{k}}^{\mathrm{cr}}$ and $\sigma_{\mathrm{k}} = 0.0833\left(\frac{V_{\mathrm{k}}^{\mathrm{cr}}}{M}\right)^{\frac{1}{3}}$.

To obtain the mixture viscosity at any temperature and any pressure, we first calculate the corresponding density of the mixture by using Eq. (4). This requires the values of M, σ, ϵ, and ω, which can be obtained from Eqs. (12)-(14) by using the critical constants and acentric factor of the components. Finally the mixture viscosity is calculated using Eq. (11), where the mixture critical volume V^{cr} is estimated from the relation $V^{\text{cr}} = M\left(\frac{\sigma}{0.0833}\right)^3$. Martial parameters used in this simulation are given in Table 1.

Table 1: Material parameters

Symbol	Meaning	Value	Unit
h_p	Layer thickness	$4, 6, 19$	m
l_p	Reservoir column length	1000	m
z_{sc}	Super compressibility factor	Max root of Eq. (9)	–
μ	Dynamic viscosity	Eq. (11)	Pas
ρ	Density	Eq. (4), 2460, 2700, 2940	kg m^{-3}
c_p	Heat capacity	$1839.3, 960, 920, 880$	$\text{Jkg}^{-1}\text{K}^{-1}$
κ	Thermal conductivity	$0.0466, 3.0, 2.5, 2.2$	$\text{Wm}^{-1}\text{K}^{-1}$
M	Mixture molecular weight	Eq. (13)	kg kmol^{-1}
$\mathbf{k}$	Intrinsic permeability	$1.0 \times 10^{-17,-18,-20}$	m^2
n	Porosity	0.085, 0.014, 0.01	–
τ	Tortuosity	$n^{\frac{1}{3}}$	-
D_0	Self diffusion coefficient	0.965, 2.0, 1.78×10^{-5}	m s^{-2}
α_L, α_T	Dispersivity	$0.1, 0.01$	m
ϵ	Energy parameter	$0.77kT^{\text{cr}}$	$\text{m}^2\text{kgs}^{-2}$
σ	Distance parameter	$0.0833\left(\frac{V^{\text{cr}}}{M}\right)^{\frac{1}{3}}$	A
ω	Acentric factor	0.228, 0.0114, 0.04	–

4 NUMERICAL SCHEME

The numerical treatment of the coupled problem of gas flow in undeformable porous media is based on the governing field equations together with discretization methods in the space and time domains. The method of weighted residuals is applied to derive the weak formulations of all the governing equations given. Within the framework of a standard Galerkin procedure, a spatially discretized formulation of the weak forms is defined in the finite element space.

For the required time discretization of the global system of governing equations we use the generalized first order difference scheme. The nonlinear coupled boundary value problem is solved iteratively using the Picard linearization within the context of the finite element method [9]. We adopt a staggered scheme without losing the accuracy of the solutions. Within this context, at first the gas flow problem is solved, then heat and mass transport problem are solved separately considering the results from the gas flow. This iteration cycle is repeated until the converged solution of all primary variables is achieved. For the sake of simplicity, at the current stage, gas flow to transport coupling is conducted in one way only, i.e. pore pressure changes influence transport, without the inverse feedback.

5 BENCHMARK DEFINITION

In the present study of non-isothermal compressible flow, we consider three main components (carbon dioxide, nitrogen and methane), which appear in the natural gas mixture. The accurate calculation of density of nonpolar light hydrocarbon mixtures is done by using a modified version of the Peng-Robinson equations of state. The pressure provided by carbon dioxide during expansion upon natural gas, enhances gas recovery. Concentration gradients in the gas phase lead to mixing by molecular diffusion-dispersion. The extent to which carbon dioxide could mix with natural gas is a primary concern for technical and economic feasibility of carbon sequestration with enhanced gas recovery. Pressure gradients arise due to mixing and introduce minor advective flow at rates depending on the permeability and porosity of the porous reservoir medium.

5.1 System geometry and conditions

- Geometry: We used middle and bottom layers as reservoir and top one for seal. Injection and production well are 500 m apart. 2-dimensional depleted gas reservoir model is taken in $r-z$ plane which is 19 m thick in z-direction and 1000 m long in r-direction (see Fig. 1).

- IC: We assume that caprock-reservoir system is filled with 75% methane and 25% nitrogen at pressure of 4.0×10^6 Pa and temperature 393.15 K.

- BC: Through injection point I, we perform injection of CO_2 gas with pressure 6.0×10^6 Pa and temperature 353.15 K. At production running well at pressure 3.0×10^6 Pa, we are producing natural gas which is a mixture of 75% methane and 25% nitrogen at temperature 393.15 K.

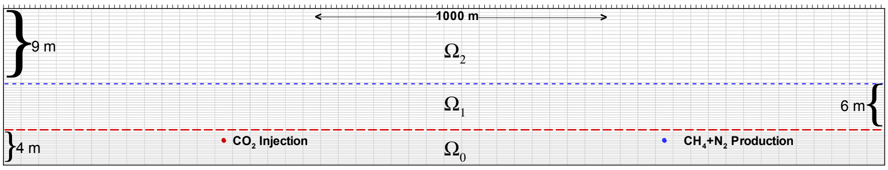
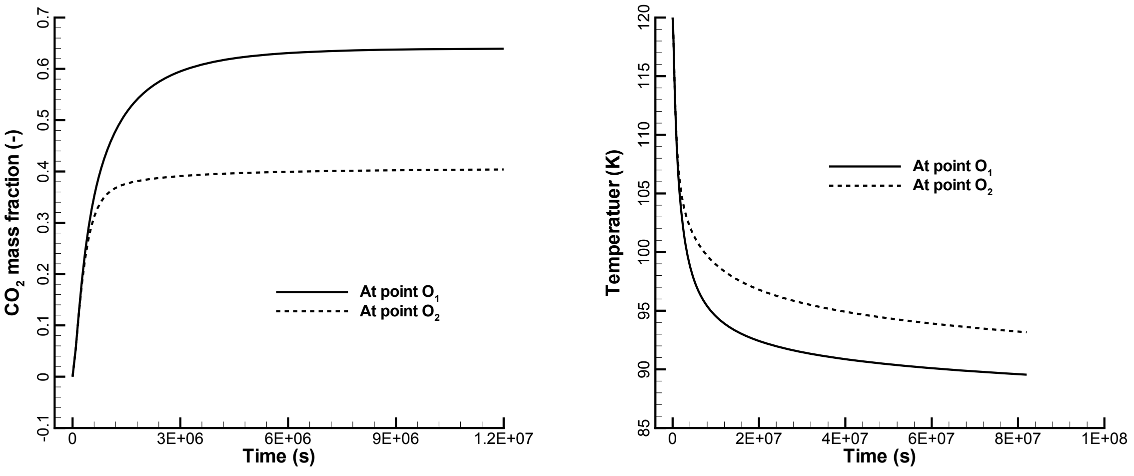


Figure 1: Model setup for CSEGR and grid arrangement

5.2 Results

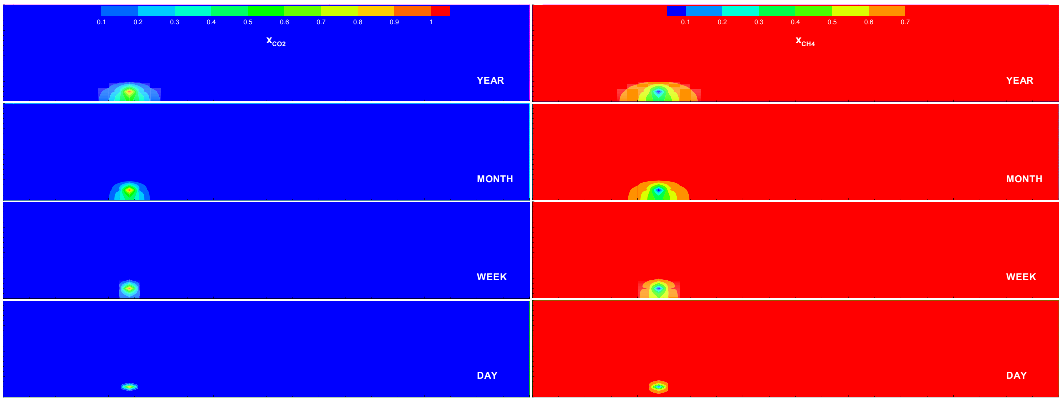
In Figs. 2a, b we have shown the time evolution of carbon dioxide mass fraction and temperature at certain observation points. The observation points are O_1 and O_2, which are at the perimeter of circle around the injection point I. Larger value of x_{CO_2} and temperature at observation point below to injection point than above is shown by Figs. 2a, b, and is due to advection enhanced by gravity force. At beginning of the simulation rate of gas expansion is high due to relatively low formation pressure. Hence advective transport plays role to make steep rise of mass fraction and temperature in Figs. 2a, b. As injection and formation pressure gets in equilibrium then advection is not so extensive, hence after steep rise temperature and mass fraction versus time curve showing steady nature for both observation points. During this time transport by diffusion-dispersion is dominated over advection.



(*a*) (*b*)

Figure 2: Time evolution of CO_2 mass fraction and temperature at points O_1 and O_2 located equidistantly (1 m) above and below to injection point, respectively

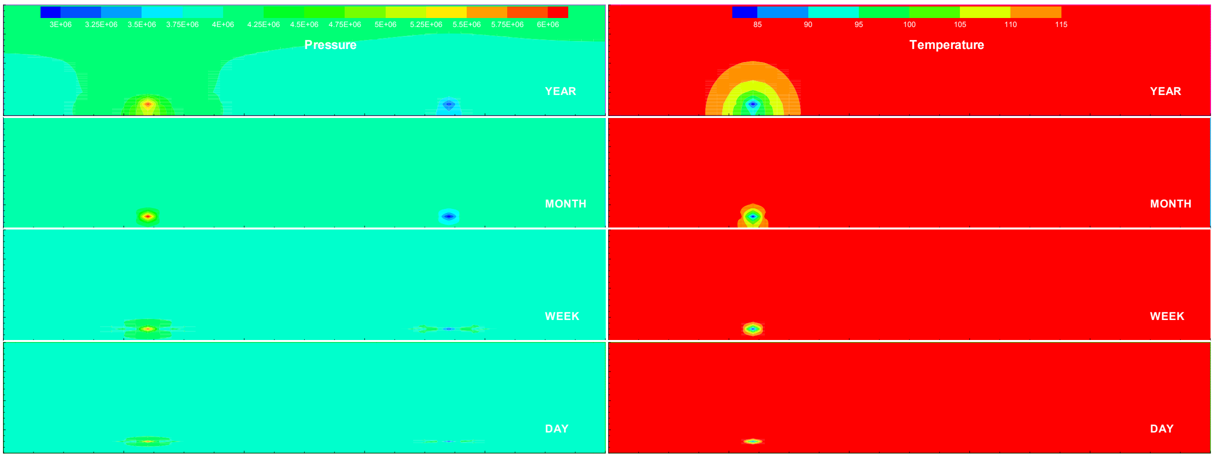
Figs. 3a, b show the distribution of carbon dioxide and methane mass-fractions for four different times. Multi-component transport in the gas mixture is mainly by advection and diffusion whereas dispersion is due to random motion of molecules. Advection is driven by pressure gradients as well as buoyancy forces (in the case when fluid density differences are significant), and diffusion depends on concentration gradients. From Fig. 3a it can be seen that near the injection point advective mass-fluxes are dominating over diffusive ones because of high pressure gradients for compressible flow. At the same time, due to the larger density of injected carbon dioxide than natural gas, gravity makes safe disposal of carbon dioxide in such reservoir caprock system.



(*a*) (*b*)

Figure 3: Mass fraction distribution of (*a*) CO_2; (*b*) CH_4 in the permeable layers of a gas reservoir

Figs. 4a, b depict the evolution of pressure and temperature distribution in the reservoir layer in the sequence of CO_2 injection through point I. As gas is compressible, transient flow is developing at the beginning; simulation shows a symmetry pattern of pressure and temperature distribution, respectively around injection point I (see Figs. 4a, b). Distribution pattern for long duration showing that gravity making the pressure distribution asymmetric and a positive pressure gradient is developed in negative z-direction. Therefore, mass and heat transport in bottom reservoir layer is dominated over the middle one due to gravity (see Figs. 3a, b). This supports the strategy to store carbon dioxide along the lower surface of the bottom reservoir layer and producing natural gases from above reservoir layers



(*a*) (*b*)

Figure 4: Evolution of (*a*) gas pressure and (*b*) temperature distribution through the permeable layers of a depleted gas reservoir

In Fig. 5 we present carbon dioxide mass fraction profile along vertical line through injection point I. Figure shows that at lower surface of the bottom reservoir layers (where injection is performed) most of carbon dioxide is disposed. This figure shows some sharp discontinuity along interface of reservoir layers Ω_0 and Ω_1. This is due to lower permeability of layer Ω_1, and carbon dioxide can not penetrate into it easily. We have found that in the caprock layer (least permeable), i.e. Ω_2, after one year maximum carbon dioxide mass fraction is 0.001 and in the reservoir layer Ω_0 (most permeable) is 0.366. This shows present caprock-reservoir system is safe for carbon dioxide disposal.

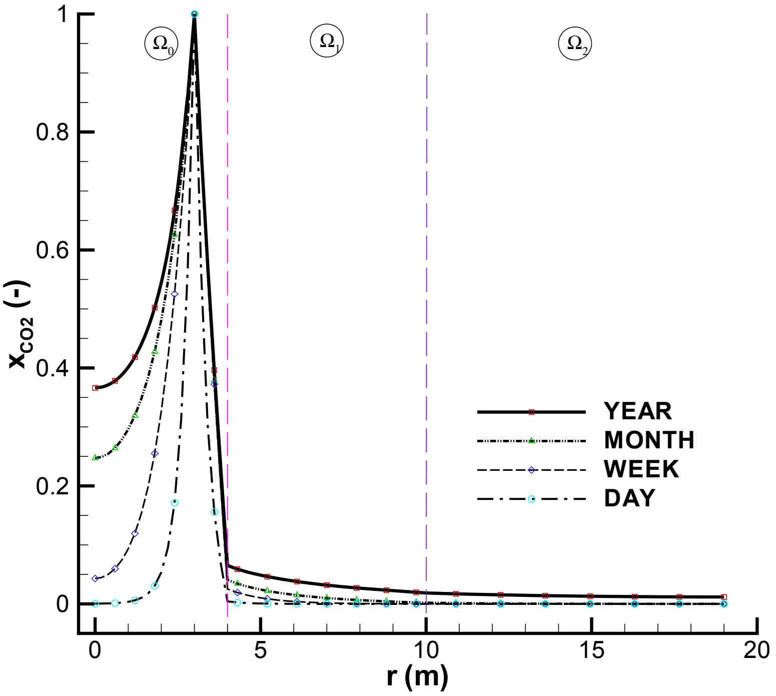


Figure 5: Profile of the CO_2 mass fraction along the vertical line passing through injection point

6 CONCLUSIONS

In this paper, we have demonstrated that the benchmark was carefully defined to analyze carbon dioxide disposal coupled with enhanced natural gas recovery into depleted gas reservoir system. We provide the following conclusions.

- The governing equations for the compositional gas flow model presented here include the mass balance Eq. (3) for the gaseous mixture along with heat and mass transport equation according to Eq. (5) and Eq. (7), respectively. Equation of state for considered mixture of nonpolar gases at high pressure has been used.

- We used a combined monolithic / staggered coupling scheme, i.e. monolithic for the gas flow and staggered for transport processes.
- Non-isothermal compositional gas flow model developed here works for a pT range up to 7.0×10^6 Pa and 393.15 K. This allows model applications for the calculation of gaseous mixture properties in the vicinity of heat loss due to expansion of injected carbon dioxide.
- Properties of CO_2 and CH_4 are favorable for repressurization without extensive mixing over time scales of practical interest. Simulations of the process of CO_2 injection, into a depleted gas reservoir carried out with OGS confirm the plausibility of CSEGR as a way to CO_2 sequestration while enhancing natural gas recovery.

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