Evaluating variable switching and flash methods in modeling carbon sequestration in deep geologic formations using PFLOTRAN

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Abstract. PFLOTRAN a massively parallel computer code for modeling coupled hydro-thermal-chemical processes in variably saturated, non-isothermal porous media is applied to sequestration of supercritical CO_2 in deep geologic formations. Two different methods of solution to the governing partial differential equations are implemented referred to as variable switching and the flash approach. Variable switching entails choosing the independent variables according to the set of phases present in a control volume, whereas in the flash approach a persistent set of variables are used through the calculation. The features and performance of the two approaches are described and contrasted in regard to stability and convergence, flexibility of choice of solver, and scaling behavior.

1. Introduction

Application of high performance computing to assess the risks involved in carbon sequestration in deep geologic formations is an important issue in mitigating global warming caused by release of green house gases into the atmosphere. Models simulating multiphase subsurface reactive flows are needed which apply to basin-scale systems with three-dimensional computational domains on the order of 100 km × 100 km × 5 km. The model must account for hundreds of injection wells to accommodate the volume of CO_2 that must be injected requiring localized high spatial resolution around each well. Additional processes involving chemical reactions and mechanical effects increase the complexity and computational resources that are needed. One approach is to use adaptive mesh refinement (AMR) to refine the grid where needed in the domain, such as around wells and interfaces between different stratigraphic units. AMR is based on multilevel solvers and conventionally requires a single set of variables common to all levels. This work investigates use of the flash approach to replace variable switching methods which allows for a persistent set of unknown variables as phase changes take place.

2. Modeling Multiphase Flow

The general governing equations for a multiphase system may be written as

$$\sum_\alpha \left\{ \frac{\partial}{\partial t}(\varphi s_\alpha \eta_\alpha x_{i\alpha}) + \boldsymbol{\nabla}\cdot\boldsymbol{F}_{i\alpha} \right\} \;=\; Q_i, \tag{1}$$

where φ denotes the porosity of the geologic formation, s_α, η_α, refer to the saturation and molar density of phase α, $x_{i\alpha}$ denotes the mole fraction of the ith component in phase α, and Q_i denotes a source/sink term associated with the ith component. The flux $\boldsymbol{F}_{i\alpha}$ contains contributions from advection derived from Darcy's law and diffusion/dispersion

$$\boldsymbol{F}_{i\alpha} = \boldsymbol{q}_\alpha \eta_\alpha x_{i\alpha} + \boldsymbol{j}_{i\alpha}, \tag{2}$$

with Darcy velocity

$$\boldsymbol{q}_\alpha = -\frac{k k_\alpha}{\mu_\alpha} \boldsymbol{\nabla}\big(p_\alpha - W_\alpha \eta_\alpha g \boldsymbol{z}\big), \tag{3}$$

where formation permeability is denoted by k, relative permeability by k_α, fluid viscosity by μ_α, fluid pressure by p_α, acceleration of gravity by g, formula weight by W_α, and diffusive flux by $\boldsymbol{j}_{i\alpha}$ defined as

$$\boldsymbol{j}_{i\alpha} = -\varphi s_\alpha D_\alpha \eta_\alpha \boldsymbol{\nabla} x_{i\alpha}. \tag{4}$$

For an isotropic medium, the dispersion tensor is given by

$$D_\alpha = \big(D_\alpha^0 + \alpha_T v_\alpha\big)\boldsymbol{I} + \big(\alpha_L - \alpha_T\big)\frac{\boldsymbol{v}_\alpha \boldsymbol{v}_\alpha}{v_\alpha}, \tag{5}$$

with species-independent diffusion coefficient D_α^0 and fluid velocity $\boldsymbol{v}_\alpha = \varphi \boldsymbol{q}_\alpha$. The diffusive/dispersive flux satisfies the condition

$$\sum_i \boldsymbol{j}_{i\alpha} = 0. \tag{6}$$

These equations are subject to the constraint conditions

$$\sum_\alpha s_\alpha = 1, \tag{7a}$$

$$\sum_i x_{i\alpha} = 1, \tag{7b}$$

the solubility constraints

$$x_{i\beta} = K_i^{\alpha\beta} x_{i\alpha}, \tag{8}$$

and capillary pressure relations

$$p_{\alpha\beta}^c = p_\alpha - p_\beta. \tag{9}$$

Constitutive relations are needed to relate capillary pressure and phase saturation such as van Genuchten or Brooks-Corey relations.

There are $N_C + 1$ unknowns and an equal number of equations, where N_C refers to the number of independent components in the system in addition to pressure and temperature. Summing Eqn.(1) over all species the diffusive flux disappears yielding the flow equation

$$\sum_\alpha \left\{\frac{\partial}{\partial t}\big(\varphi s_\alpha \eta_\alpha\big) + \boldsymbol{\nabla}\cdot\big(\boldsymbol{q}_\alpha \eta_\alpha\big)\right\} = \sum_i Q_i. \tag{10}$$

This equation may be substituted for one of the equations in Eqn.(1) such as H_2O, for example. The mass conservation equations are coupled to the energy conservation equation providing an equal number of equations as unknowns.

Table 1. Possible choices of independent variables used in the variable switching approach. Note that g = supercritical CO_2 = SC, and l = H_2O phase.

Phase	Variables								
	X_1	X_2	X_3	X_1	X_2	X_3	X_1	X_2	X_3
liquid	p_l	T	$X^l_{\mathrm{CO}_2}$	p_l	T	$X^l_{\mathrm{CO}_2}$	p_l	T	$X^l_{\mathrm{CO}_2}$
gas	p_g	T	$X^g_{\mathrm{CO}_2}$	p_g	T	$X^g_{\mathrm{CO}_2}$	p_g	T	$X^g_{\mathrm{CO}_2}$
two-phase	p_g	T	s_g	p_g	T	p_l	p_g	s_g	$X^l_{\mathrm{CO}_2}$

2.1. Methods of Solution

2.1.1. Variable Switching In the variable switching approach the independent variables are chosen based on the phases present. This choice is not unique. Several different possibilities are listed in Table 1. An important drawback of the variable switching approach is that it may be difficult to apply when using multilevel solvers where the independent variables are different at different levels [1].

2.1.2. Flash Method An alternative approach to variable switching is the flash method. Although the variable switching method is often considered stable and efficient [2], it has several shortcomings: 1) it causes perturbations during Newton iterations when phase changes take place; 2) the change in the definition of independent variables affects the structure of the Jacobian matrix; and 3) as a consequence this degrades performance of the preconditioner during the linear solve. Finally, the variable switching approach is not appropriate for use with multilevel solvers because of the possibility for the need to solve for different independent variables on different levels [1].

In the flash approach the primary variables preserved during the solution of the governing equations. The flash method has been implemented in the FLASH2 mode in PFLOTRAN. The primary variables are p, T and the total mole fraction z_i of the ith component summed over all phases, defined as:

$$z_i = \frac{\sum_\alpha n_i^\alpha}{\sum_\alpha \sum_j n_j^\alpha} = \frac{\sum_\alpha s_\alpha \rho_\alpha x_i^\alpha}{\sum_\alpha s_\alpha \rho_\alpha}, \tag{11a}$$

where the latter form is derived from the identity

$$\frac{n_i^\alpha}{V} = \frac{n_i^\alpha}{n_\alpha} \frac{n_\alpha}{V_\alpha} \frac{V_\alpha}{V_p} \frac{V_p}{V} = \varphi s_\alpha \rho_\alpha x_i^\alpha, \tag{12a}$$

where V is the total control volume and V_p denotes the pore volume. Explicitly for a two-phase ($\alpha = w$, SC) system where w designates the phase H_2O and SC designates supercritical CO_2, z_i can be expressed as

$$z_i = \frac{s_w \rho_w x_i^w + s_{\mathrm{SC}} \rho_{\mathrm{SC}} x_i^{\mathrm{SC}}}{s_w \rho_w + s_{\mathrm{SC}} \rho_{\mathrm{SC}}}, \tag{13}$$

for molar fluid densities ρ_w, ρ_{SC} and saturation s_w, $s_{\mathrm{SC}} = 1 - s_w$. The variable z_i is a persistent degree of freedom throughout the simulation.

Let x_i, y_i be the mole fraction of component i in liquid and supercritical phases, respectively, related by the equilibrium constant K_i by the expression

$$y_i = K_i x_i, \tag{14}$$

and let ζ_{SC} represent the supercritical phase mole fraction defined as

$$\zeta_{\mathrm{SC}} = \frac{\sum_i n_i^{\mathrm{SC}}}{\sum_\alpha \sum_i n_i^\alpha}. \tag{15}$$

Under a phase transformation mass conservation implies the relation

$$z_i = \zeta_{\mathrm{SC}} y_i + (1 - \zeta_{\mathrm{SC}}) x_i. \tag{16}$$

Using Eqn.(14) it follows that

$$x_i = \frac{z_i}{1 + (K_i - 1)\zeta_{\mathrm{SC}}}, \quad y_i = \frac{K_i z_i}{1 + (K_i - 1)\zeta_{\mathrm{SC}}}. \tag{17a}$$

The value of ζ_v can be found by solving the flash equation (i.e. the Rachford-Rice equation)

$$F(\zeta_{\mathrm{SC}}) = \sum_i (y_i - x_i) = \sum_i \frac{z_i(K_i - 1)}{1 + (K_i - 1)\zeta_{\mathrm{SC}}} = 0. \tag{18}$$

3. Comparison of Variable Switching and Flash Approaches

Variable switching and flash approaches are compared for a 3D injection problem. Physical properties used in the simulations consist of the Span-Wagner [4] EOS for supercritical CO_2 and the density mixture correlation taken from Duan et al. [3]. The variation in mixture density with temperature calculated using this correlation is shown in Figure 1 for different brine concentrations. At higher temperatures the mixture density becomes less than the brine density. However, at lower temperatures the mixture density is greater than the brine density and the mixture sinks [5]. It should be noted that use of ideal mixing results in a mixture density that is always less than the brine density. The solubility of CO_2 in brine was calculated using the correlations presented in Duan et al. [6].

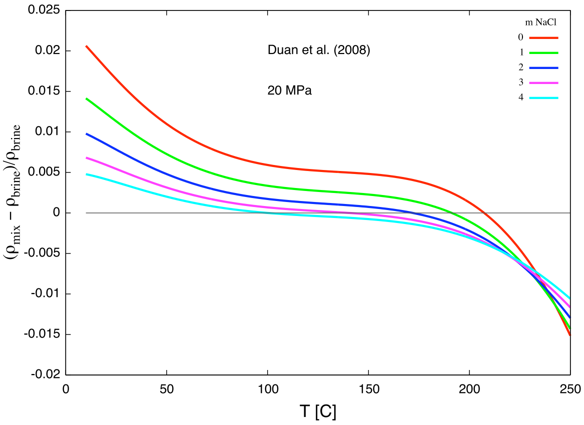

Figure 1. Comparison of mixture density for different brine concentrations for CO_2-brine mixtures of 0, 1, 2, 3, 4 m NaCl with the pure brine density as a function of temperature using the correlation derived in [3].

Performance results for a 3D test problem with domain size 7 km × 7 km × 250 m with grid spacings of $\Delta x = \Delta y = 43.75$ m and $\Delta z = 10$ m are listed in Table 2 with scaling results shown in Figure 2. In these simulations the presence of NaCl was not considered. A formation permeability of 2 Darcy and porosity of 0.38 was used. Injection of supercritical CO_2 at the center of the domain at a rate of 0.2487 kg/s over a period of 25 years. The problem was run on Jaguar XT5 at ORNL using 240 processor cores. For variable switching the first set of variables listed in Table 1 were used. The flash approach shows significant improvement compared to variable switching.

Table 2. Comparison of performance results of variable switching and flash methods for 3D CO_2 injection problem.

Mode	Steps	Newton	Linear	Time-Step Cuts	Time [s]
Var. Switch.	3467	3467	605701	0	4526.2
Flash	2389	2389	408406	0	2973.9

4. Conclusion

The flash and variable switching methods were compared for the two-phase problem of injection of supercritical CO_2 into a brine reservoir fluid. It was found the the flash approach gave better performance compared to variable switching.

Acknowledgments

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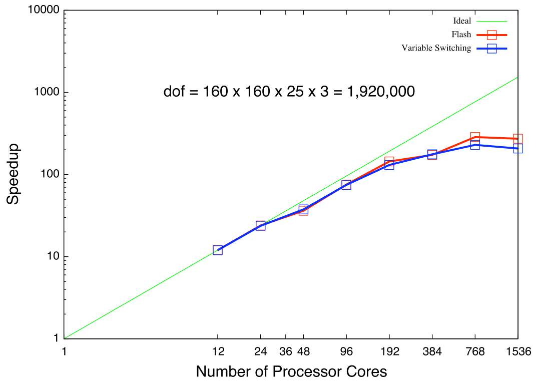

Figure 2. Scaling results for variable switching and flash methods.

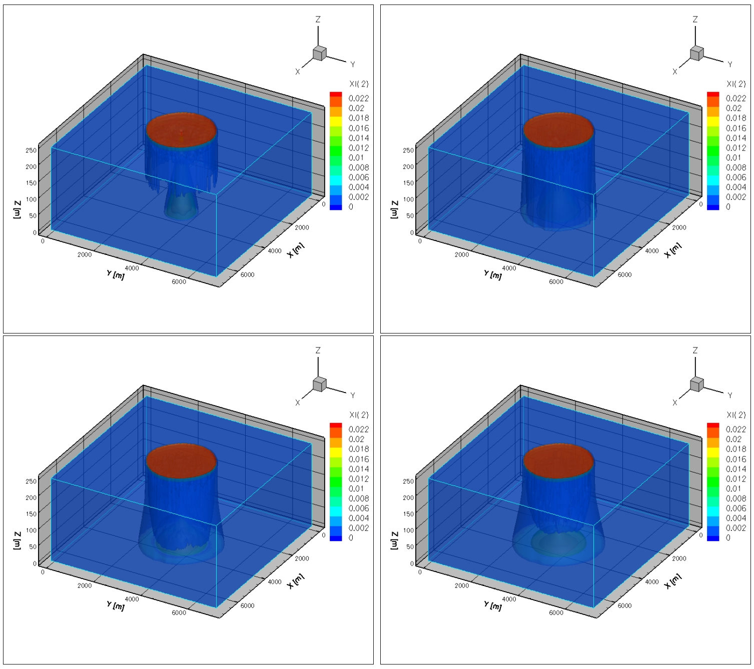

Figure 3. Dissolved CO_2 plotted at times 50, 100, 200 and 300 years.