



Chromatographic separation and liquid drop-out in unconventional gas reservoirs



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ABSTRACT

A study is presented on the impact of heavier components on gas production in unconventional reservoirs. The competing effects of Knudsen and molecular diffusion are incorporated by using Maxwell-Stefan equations and the Dusty-Gas approach. The total flux of gas considers contribution of viscous flow, molecular and Knudsen diffusion, adsorption/desorption and surface diffusion. The model is used to perform numerical experiments demonstrating the effects of transport mechanisms, medium permeability and initial gas composition on production performance. We evaluate pressure decline and chromatographic separation effects in the presence of a wider range of components in flow through both adsorbing and poorly-adsorbing porous media.

During gas production, the flow of different chemical species will result in chromatographic separation. Therefore, gas composition will change over time, causing shifts in the phase envelope. Depending on the degree of chromatographic separation, liquid drop-out may occur in the near-wellbore.

In this study, we demonstrate that for poorly adsorbing media, the degree of chromatographic separation is small due to the balance between Knudsen and molecular diffusion fluxes. In this case, chromatographic separation is intensified with decreasing permeability, since molecular sieve mechanism becomes dominant with smaller pore sizes at lower pressures. However, when adsorption is present, species separation is more relevant, resulting in significant shifts in the phase envelope. Production behavior is affected by the higher affinity of hydrocarbon molecules with adsorption sites. As a result, condensate formation in the reservoir is likely to occur as production progresses.

Many analytical/semi-analytical models available for gas transport in unconventional reservoirs are based on single component or binary mixtures. In this work, we demonstrate the effect of including heavier fractions in modelling gas transport when they are present, which is relevant in prediction of pressure decline, produced gas composition and natural gas liquids (NGLs) yields. Moreover, we demonstrate that gas composition at the wellbore can be significantly influenced by a balance between viscous and diffusion fluxes, and we highlight the relevant transport mechanisms to be considered when modelling multicomponent gas flow in tight reservoirs.

1. Introduction

Modelling multicomponent gas flow in porous media is relevant in many fields, such as chemical reaction engineering (Levenspiel, 1999), solid-oxide fuel cells (Yuan and Sunden, 2014; He et al., 2013), large-scale separation processes (Wankat, 1986), among others. The transport models used in those processes have been increasingly applied to the modelling of unconventional tight gas reservoirs. Although those processes are distinct macroscopically, the physics involved are similar at the microscopic level: momentum and mass transfer of chemical species through a porous medium.

Pore systems in tight/shale reservoirs vary from micro to nanometer scales (King et al., 2015). Under operational conditions, all flow regimes may occur in a tortuous pathway in the same porous medium (from continuum to Knudsen flow regime) (Gad-el-Hak, 1999), and different mechanisms will manifest as pressure decreases (due to production) or increases (due to injection) in the reservoir. The total flux in tight reservoirs can be quantified as a sum of fluxes due to diffusion and viscous mechanisms. In addition to that, in both operation modes, different molecules are being transported, and species separation and affinity with adsorption sites will start to play a role on fluid transport.

In shale reservoirs, chromatographic separation effects become

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relevant whenever adsorption is present, for smaller pore sizes and for smaller pressures. As a result, a composition gradient is generated in the porous medium, which can be significant, depending on fluid composition, operating conditions and reservoir characteristics (e.g., permeability, adsorption properties). Changing fluid composition along the reservoir can greatly impact gas and condensate production, as well as pressure decline. Multicomponent gas flow and storage in tight formations is also relevant for gas in place calculations (Hartman et al., 2011; Das et al., 2012), CO₂ injection and storage (Schaefer et al., 2014; Middleton et al., 2015), fluid composition in production data analysis (Clarkson et al., 2016) and forecasting of condensate production. Therefore, it is important to properly model this phenomenon.

A plethora of analytical and semi-analytical methods were developed recently to evaluate pressure response and apparent permeability in unconventional gas reservoirs. Several methods, however, address solely single component gas flow (Ziarani and Aguilera, 2012; Darabi et al., 2012; Singh et al., 2014; Wu et al., 2016; Yang et al., 2016; Behrang and Kantzas, 2017). When heavier fractions are present, simplified single component models can no longer be used. Even in some dry gas reservoirs, heavier hydrocarbon fractions may be present, although at low composition. The issue is then to evaluate how reliable are single component models when lesser amounts of heavy hydrocarbons fractions are present.

To address species transport, methods such as Advective-Dispersive Model and Dusty-Gas Model have been used to combine momentum and mass transfer (Webb and Pruess, 2003). Among those, the Dusty-Gas model is popular and has been successfully applied in many processes. Nevertheless, research is still undergoing on how to properly model gas transport in slip and transitional flow regimes. In this sense, the Dusty-Gas model is treated with some skepticism, because it is claimed that it over-estimates flow in these regimes (Landry et al., 2016; Bravo, 2007). While molecular simulation has been used to address some of these issues in modelling (Niu et al., 2014; He et al., 2016), given the heterogeneity and scale span of the flow, its wider application becomes impractical with the computational resources currently available.

Essentially, it can be considered that, at the core and reservoir scales the mechanisms of gas transport in micro and nanopores remain unresolved. Ultimately, models applied at those flow regimes can reproduce physical experiments by using matching parameters or correction factors that lump some of the uncertainties related to interactions between molecules and pore walls (e.g., Tangential Momentum Accommodation – TMAC, Klinkenberg parameter, surface diffusivity, etc.). In particular, the Dusty-Gas model has been able to reproduce experimental data for many processes with a high degree of success, as demonstrated by data-model comparisons in Krishna and Wesselingh (1997) and references therein.

Recent attempts to model multicomponent flow in shale reservoirs have also been performed using compositional models from commercial simulators (Alharthy et al., 2016; Li et al., 2016). Reservoir simulators are advantageous for being able to include some of the heterogeneities present at reservoir-scale. Nevertheless, the physics involved in gas transport in shale reservoirs is still not well understood at the pore scale, and commercial simulators do not give the flexibility to evaluate how different physics affect production.

The work developed in this paper was inspired by a sensitivity analysis performed by Rezaveisi et al. (2014) on the investigation of chromatographic separation in shale gas wells. In their work, a model that accounts for bulk (Darcy) flow, slip flow and Knudsen diffusion was used. Although the authors perform relevant sensitivity studies on the effect of several parameters on outlet gas composition, the model lacks the effects of adsorption and molecular diffusion.

Freeman (2013) also investigated the effect of changing gas composition by using the Dusty-Gas model. They used hypothetical adsorption parameters to evaluate its impact on produced gas composition. Also, stimulated reservoir volume (SRV) was incorporated in their model,

which complicated the deconvolution of reservoir response. This is because composition changes are partially induced by production operations, and may confound analysis of transport phenomena through porous media and *in-situ* gas composition (Gensterblum et al., 2015).

In this work, single phase, multicomponent gas flow is evaluated. The Dusty-Gas model is selected due to its successful history of application, its use of Maxwell-Stefan equation to address momentum transfer between molecules and the relatively straight-forward numerical implementation when compared to more complex molecular models. Adsorption and surface diffusion effects are included, and the effect of commingling transport mechanisms is evaluated. We demonstrate that both adsorption and molecular diffusion are crucial for dictating outlet gas composition, and they cannot be ignored if organic matter sites are present. This work goes beyond superficial analysis by evaluating the effect of transport mechanisms in different reservoir types (dry-gas, wet-gas), the potential of liquid drop-out in the near-wellbore due to adsorption, and condensate production. This application is particularly relevant for wet gas tight reservoirs.

2. Statement of theory

2.1. Non-adsorbing porous medium

For non-adsorbing 1D porous medium, the total flux is the sum of viscous and diffusion fluxes, which incorporates molecular (bulk) and Knudsen diffusion.

$$\mathbf{N} = \mathbf{N}^{vis} + \mathbf{N}^{dif} \quad (1)$$

The viscous flux for multicomponent flow is obtained by using a modified form of Darcy's equation:

$$N_i^{vis} = -\frac{x_i P k_{\infty}}{RT \mu} \frac{dP}{dz} \quad (2)$$

In this equation, $i = 1, 2, \dots, n$, where n is the number of components. k_{∞} represents the absolute permeability of the system.

Molecular diffusion is the random motion of molecules. The most commonly used method to model this phenomena in petroleum upstream processes is the classical Fick's Law. From classical Fick's law, the diffusion flux of a component in a binary mixture is defined as:

$$J_1 = -cD_{12}\nabla x_1 \quad (3)$$

For a negative concentration gradient, the flux will monotonically increase. A natural extension of the classical Fick's law to a multicomponent mixture is:

$$J_i = -cD_i^{eff}\nabla x_i \quad (4)$$

D_i^{eff} is an effective diffusion coefficient, and corresponds to the diffusivity of component i in the mixture. Equation (4) postulates that each flux is proportional to its own composition gradient and the components do not interact with diffusing and non-diffusing species in the mixture. This condition of independent diffusion is strictly met in three cases:

1. For binary diffusion;
2. For diffusion of dilute species;
3. When the diffusion coefficients of all components in the mixture can be regarded as equal.

However, those conditions will not be met in most practical applications. For processes where components have a high concentration of varied species with widely differing properties, the assumption of independent diffusion breaks down.

Krishna and Standart (1979) demonstrated that classical Fick's law for

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