International Journal of Coal Geology 159 (2016) 120–134

Contents lists available at ScienceDirect

International Journal of Coal Geology

journal homepage: www.elsevier.com/locate/ijcoalgeo

Direct simulation of supercritical gas flow in complex nanoporous media and prediction of apparent permeability

Christopher J. Landry [a,*], Maša Prodanović [b,1], Peter Eichhubl [c,2]

[a] Center for Petroleum and Geosystems Engineering, The University of Texas at Austin, Austin, TX 78712, USA
[b] Department of Petroleum and Geosystems Engineering, The University of Texas at Austin, Austin, TX 78712, USA
[c] Bureau of Economic Geology, Jackson School of Geosciences, The University of Texas at Austin, Austin, TX 78757, USA

ARTICLE INFO

Article history:
Received 7 January 2016
Received in revised form 22 March 2016
Accepted 23 March 2016
Available online 3 April 2016

Keywords:
Slip-flow
Lattice Boltzmann
Mudrock
Shale
Pore-scale
Mesoscopic model

ABSTRACT

Mudrocks and shales contain pores within the size range of 2–100 nm. Flow of supercritical gas in these pores at reservoir pressure–temperature conditions falls within the slip-flow and early transition-flow regime ($0.001 < Kn < 1.0$). Currently, the description of supercritical gas flow in these flow regimes is mostly limited to simple tube and channel geometries that are of limited applicability to the sponge-like or platy nanoporous geometry in organic matter or clays. Here, we present a local-effective-viscosity multi-relaxation-time lattice Boltzmann model (LEV-LBM) designed to simulate gas flow in the slip- and early-transition-flow regimes in complex geometries. The LEV-LBM is informed with local effective viscosities at each node to capture the variance of the mean free path of gas molecules in a bounded system. The corrected mean free path for each lattice node is determined using a three-dimensional wall function adaptable to complex pore geometries. To enforce a non-zero slip velocity at solid boundaries, a combined diffusive bounce-back scheme is applied to the pore-walls. The LEV-LBM is first validated in simple tube geometries, where good agreement is found for Knudsen numbers up to 1.0. We then use the LEV-LBM to quantify the finite tube length effect and comment on the implications for pore-network models. We finally demonstrate the utility of the LEV-LBM by simulating pure methane flow in digital reconstructions of nanoporous organic matter at reservoir conditions, and compare the results to bundle of tubes models. We show that the bundle of tubes models overestimate apparent permeability by factors between 1.52 and 153, due to the non-trivial dependence of flow on pore space connectivity and shape.

1. Introduction

Solid bitumen/pyrobitumen, and organic components of thermally mature mudrocks host nanometer-scale pores (nanopores) that provide pathways for gas flow through mudrocks (Chalmers et al., 2012; Heath et al., 2011; Jiao et al., 2014; Klaver et al., 2015; Loucks et al., 2012; Milliken et al., 2013; Peng et al., 2015; Wood et al., 2015). At reservoir pressure–temperature conditions the mean free paths of supercritical gases, such as methane and carbon dioxide, are at or near the same order in length as the size of the pores. This causes the flow of these gases to significantly deviate from classic Navier–Stokes predictions, resulting in an increase in the apparent permeability of the porous media with decreasing pore pressure. Although this rarefaction effect on, or non-Darcy behavior of, the apparent permeability of porous media is well-known (Klinkenberg, 1941), the prediction of apparent permeability as a function of pore space geometry and gas properties remains elusive.

The deviation of gas flow from the classic Navier–Stokes description is qualitatively described by the flow-regime, determined by the Knudsen number, $Kn = \lambda_o / L_{ref}$, where λ_o is the unbound mean free path of the gas and L_{ref} is a reference length (i.e. the radius of a tube or aperture of a slit). A $Kn > 0.001$ indicates the onset of non-negligible deviation from Navier–Stokes predictions, with flow regimes being divided up into slip, transition and free-molecular for Knudsen numbers between, 0.001 to 0.1, 0.1 to 10, and greater than 10, respectively. These flow regimes are roughly divided up in this manner to reflect the changing physics as the Knudsen number increases (Colin, 2005; Zhang et al., 2012a). In the slip-flow regime the gas–gas collisions are far greater in number than the gas–wall collisions, however, the gas–wall collisions result in a significant non-zero flow velocity at the walls in comparison to the mean flow velocity, hence the term "slip". In the transition-flow regime there is a transition from gas–gas collision to gas–wall collision dominance, with both having a significant effect on the overall flow. In the free-molecular-flow regime the gas–wall collisions become increasingly dominant to the extent that the gas–gas collisions can be ignored, approaching flow that is described by Knudsen diffusion. It is somewhat

* Corresponding author at: Center for Petroleum and Geosystems Engineering, The University of Texas at Austin, 1 University Station C0304, Austin, TX 78712, USA.
E-mail addresses: christopher.landry@utexas.edu (C.J. Landry), masha@utexas.edu (M. Prodanović), peter.eichhubl@beg.utexas.edu (P. Eichhubl).
[1] Department of Petroleum and Geosystems Engineering, The University of Texas at Austin, 200 E. Dean Keeton, Stop C0300, Austin, TX 78712-1585, USA.
[2] Bureau of Economic Geology, The University of Texas at Austin, University Station, Box X, Austin, Texas 78713-8924, USA.

http://dx.doi.org/10.1016/j.coal.2016.03.015
0166-5162/

common to refer to these flow regimes as non-continuum, however, it is important to note that although these flows deviate from classic Navier–Stokes description, there is no failure of the continuum assumption (Hadjiconstantinou, 2006; Guo et al., 2007). For these flows "the hydrodynamic fields and the associated conservation laws remain well-defined" (Hadjiconstantinou, 2006). The molecular velocities are far greater than the flow velocities of interest here, and subsequently the time scale of individual intermolecular collisions is far smaller than the characteristic time scale of steady-state flow. For steady-state flow the time-averaged microscopic behavior of the fluid in any volume of pore space results in well-defined velocity moments (flow velocity), this suggests that a continuum treatment is valid. Therefore, the deviation of supercritical gas flow in nanoporous media from the classic Navier–Stokes description can be accounted for using an "extended" Navier–Stokes constitution (Guo et al., 2007). Considering the gas flow in nanopores at reservoir pressure–temperature conditions is not actually 'rarefied', includes flow outside the slip-flow regime, and the term non-continuum is misleading, we refer to this type of flow as mixed viscous-ballistic (MVB).

Physical measurement of MVB flow in simple engineered geometries are typically conducted under rarefied conditions, as in, at low pressure, often with large inlet over outlet pressure ratios (e.g. Tison, 1993; Arkilic et al., 1997; Roy et al., 2003; Colin, 2005; Marino, 2009). These pressure conditions are significantly different from those experienced during hydrocarbon production. The prediction of apparent permeability as a function of pore space geometry in complex nanoporous media requires an investigation at the pore-scale. For the investigation presented here this results in a spatial scale of interest on the order of 100–1000 nm (thousands to millions of ~5 nm radius pores). At this length scale the difference between the upper and lower pressure bounds is very small in comparison to typical pore pressures at reservoir conditions. For example, a 100 MPa/m pressure gradient would result in an upper and lower pressure difference for a 100 nm volume of only 10 Pa. The pore-scale flow of supercritical gases in nanoporous media at the spatial scale of interest here is subject to minimal compressibility effects. Direct comparison of pore-scale modeling results, such as the one presented here, to these physical measurements would require an investigation into these compressibility related effects in the model. The flow conditions of interest of this investigation are limited to theoretical work (e.g. Loyalka and Hamoodi, 1990; Sharipov and Seleznev, 1994), due to the difficulty of physically measuring either pressure difference or flow rate while minimizing compressibility effects (Marino, 2009).

Predicting the apparent permeability of mudrocks as a function of pore space geometry, pressure–temperature conditions and gas composition for MVB flows has recently received ample attention in the hydrocarbon recovery literature (e.g. Javadpour, 2009; Civan, 2010; Darabi et al., 2012; Sakhaee-Pour and Bryant, 2012; Mehmani et al., 2013; Rahmanian et al., 2013; Yao et al., 2013; Islam and Patzek, 2014; Ma et al., 2014; Singh et al., 2014; Kazemi and Takbiri-Borujeni, 2015; Naraghi and Javadpour, 2015). The over-arching goal is predicting long-term production outcomes from the characterization of mudrock pore space geometry. However, all of these models simplify these pore spaces into a collection of constant cross-section tubes or slits, by use of bundle of tubes models which treat pores as a collection of isolated tortuous tubes (e.g. Civan, 2010; Darabi et al., 2012; Naraghi and Javadpour, 2015) or by use of pore network models which treat pores as a network of constant cross-section conduits (Sakhaee-Pour and Bryant, 2012; Mehmani et al., 2013; Ma et al., 2014). Unlike the bundle of tubes models, pore network models allow for the investigation of the dependence of flow on pore space connectivity, including interconnectivity in heterogeneous networks with bimodal pore size distributions (Mehmani and Prodanović, 2014). The tube/slit models employed by bundle of tubes models and pore network studies were developed for very long tubes, with lengths approximately 20 times larger than the radius of the tube, and often far larger. Unlike for viscous flow, the permeability of a tube for MVB flows is dependent on the length of the tube and pressure gradient as a result of significant end effects (Beskok and Karniadakis, 1999; Colin, 2005; Marino, 2009). None of the above referenced studies considers the effect of finite length on flow at reservoir pressure temperature conditions and we address that issue in this work.

Furthermore, the application of tube/slit models for accurate prediction of flow in the complex pore geometries of mudrocks is still an open research question. From scanning electron microscopy (SEM) and transmission electron microscopy (TEM) studies it has been shown that the pore spaces of mudrocks are best described as "spongey" for organic matter hosted pores or as a complex collection of variable cross-sectional shape for clay hosted pores. It is also evident from imaging studies that the porosity of organic matter varies widely, and is often far greater than the bulk rock porosity (Chalmers et al., 2012; Heath et al., 2011; Jiao et al., 2014; Klaver et al., 2015; Loucks et al., 2012; Milliken et al., 2013; Wood et al., 2015). The tools for simulating supercritical gas flows in nanoporous media in anything but very long tubes, or very long/wide slits have been limited for the most part to molecular dynamics modelling (e.g. Cao et al., 2006; Falk et al., 2015; Firouzi and Wilcox, 2012). The computational requirement for molecular dynamics model prediction of flow increases with the number of molecules being tracked. Molecular dynamics flow simulations are generally limited to very small physical simulation volumes, on the order of $10 \times 10 \times 10\ nm^3$ (Firouzi and Wilcox, 2012; Falk et al., 2015). The complex and often heterogeneous pore spaces of organic matter pore systems in mudrocks may require flow simulations on volumes orders of magnitude larger than this to determine relations between pore space geometry and macroscopic transport properties. However, molecular dynamics simulations of small volumes remain very useful, particularly when surface–gas interaction forces dominate mass flux. Under such scenarios, molecular dynamics simulations offer insight into, and estimations of, surface transport properties (Firouzi and Wilcox, 2012; Falk et al., 2015), and can be used in the validation of mesoscopic modeling methods.

For MVB flows both local variation of momentum diffusivity and microscopic gas–wall interactions (slip velocity at the wall) must be accounted for, and the mesoscopic approach of lattice Boltzmann (LB) methods has long been considered a natural candidate for the prediction of such flows, at minimal computational cost in comparison to molecular dynamics simulation. Also, LB methods for single phase flows calculate velocity fields explicitly and locally, and are therefore highly scalable on parallel computational resources. A large body of work has been generated attempting to construct LB methods for prediction of MVB flows and great success has been found using a variety of approaches (e.g. Ansumali and Karlin, 2002; Chai et al., 2010; Guo et al., 2006; Guo et al., 2008; Kim et al., 2008; Li et al., 2011; Lim et al., 2002; Liou and Lin, 2014; Liu and Guo, 2013; Nie et al., 2002; Niu et al., 2007; Sbragaglia and Succi, 2005; Shen et al., 2004; Succi, 2002; Suga and Ito, 2011; Suga et al., 2010; Verhaeghe et al., 2009; Zheng et al., 2012). The vast majority of these studies have focused on simple geometries (tubes/slits), with a limited number investigating more complex geometries. Notably, the work of Suga and Ito (2011) showed promising results for three dimensional flows involving obstacles in ducts, and bumpy channels. Similar success was described in the work of Chai et al. (2010) for two-dimensional flow through square arrays of circular cylinders, using a LB model that is partly the basis of the model described here. However, in both of these investigations the methods used treated the Knudsen number as a constant. For the porous systems studied by Suga and Ito (2011) and Chai et al. (2010), assuming a constant Knudsen number was acceptable, because the pore spaces, although complex in shape, were similar in size throughout the simulated volumes. Flow in the nanoscale pore geometry of mudrocks presents an issue that has not yet been resolved, construction of a three-dimensional LB model for MVB flows through complex geometries with locally varying pore sizes, and subsequently a locally varying Knudsen number.