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Local diffusion coefficient measurements in shale using dynamic micro-computed tomography



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ABSTRACT

Diffusion is an important mass transport mechanism in ultra-low permeability shale matrix and thus, characterization of shale diffusivity is of practical necessity for shale gas developments. We present a novel method for measuring bulk and local diffusion coefficients of shale core-plugs using dynamic X-ray micro-computed tomography (micro-CT). Liquid diffusion experiments are conducted on a centimeter-scale shale core and a series of time-sequenced 3D micro-CT images are acquired through dynamic imaging. Local diffusion coefficients are measured numerically from the micro-CT data using a new mathematical method that allows us to evaluate the heterogeneity of shale diffusivity at the sub-core scale. The variation of local diffusion coefficients is quantified using the Dykstra Parsons method, which provides a means to quantify core-scale heterogeneity in shale samples. Although the micro-CT image data may be influenced by noise, the presented technique provides reasonable results and our validation studies provide fundamental design parameters for measuring diffusivity values from dynamic micro-CT experiments. In addition the presented method can be applied to other porous materials where diffusion occurs.

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1. Introduction

Shale gas has become one of the most important sources of natural gas in the United States according to the U.S. Energy Information Administration [1]. Statistics show that in the United States shale gas production increased more than 10-fold from 2000 to 2010 [2], about 23% of total gas production was shale gas in 2010 and the percentage is predicted to reach 49% by 2035 [1]. Globally, the shale gas industry is also developing in other countries including China [3], Australia [4], and Canada [5].

There are however many challenges for shale gas development and production. For instance, a better understanding of shale pore structure and flow mechanisms is required because storage and flow in shale are more complex than conventional gas reservoirs due to nanoscale pore structure of shale and its complex composition of mineral and organic matter [7]. Unlike conventional gas reservoirs in which the dominating mass transport mechanism is viscous flow that is governed by Darcy's law, molecular diffusion

and viscous flow are both important and thus contribute to the total gas transport in shale gas reservoirs [8–10].

Molecular diffusion is a significant transport mechanism in shale gas because the matrix permeability is extremely low in the range of nano-Darcy level (1 nano-Darcy equals 10^{-21} m²) [6,8]. As a result, gas displacement by molecular diffusion is comparable to or greater than that by viscous flow in the shale matrix, especially at low pressure conditions [6]. At high Knudsen numbers (Knudsen diffusion flow regime), the correction factor defined as the ratio between apparent permeability and intrinsic permeability is greater than one, which suggests stronger diffusion transport than viscous flow. In fact, molecular diffusion is even more important since over half of the total gas reserve for a shale gas reservoir is stored in the shale matrix and molecular diffusion is the key recovery mechanism [11]. As a result, molecular diffusion is a key factor affecting reserve volume, production profile and ultimately the economic prospect of a shale gas project [8,12].

Numerous researchers have studied molecular diffusion in shale and other tight porous media [6,7,10,13–17]. At the continuum scale, molecular diffusion is governed by Fick's first and second laws denoted in Eqs. (1) and (2), respectively.

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$$J = -D \cdot \nabla \varphi \quad (1)$$

$$\frac{\partial \varphi}{\partial t} = \nabla \cdot (D \nabla \varphi) \quad (2)$$

Here J is diffusive flux, φ is concentration, t is time and D is the diffusion coefficient. Most of the previous studies focused on the measurement of the diffusion coefficient to characterize shale rock [17,18]. Three common methods for measuring diffusion coefficient can be found in the literature: gas adsorption experiment, numerical simulation and X-ray micro-computed tomography (micro-CT) imaging.

The first method is by gas adsorption experiment [18]. A simple unipore diffusion model was developed by Crank [19], which describes diffusion taking place in a spherical particle with uniform-sized pores, assuming a linear adsorption isotherm [14]. By solving Fick's second law, Eq. (2), for spherically symmetric flow, an analytical solution is given by:

$$\frac{M_t}{M_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(-\frac{D_e n^2 \pi^2 t}{R_0^2}\right) \quad (3)$$

where M_t is the amount of gas diffusing into the spherical particle at transient time t , M_∞ is that at equilibrium for a given operation pressure, D_e is the effective diffusion coefficient, and R_0 is the radius of the spherical particle [20]. Gas adsorption rate data, which may be gravimetric or volumetric, are recorded during the experiment [18]. The diffusion coefficient can be obtained by fitting Eq. (3) to experimental data by adjusting D_e [13,18].

Although there are many examples where the unipore diffusion model are successfully applied [13,16,18], the model itself is limited when applied to shale because it assumes uniform-sized pores where diffusion and adsorption occur [18]. It has been proven that shales are heterogeneous with pore sizes ranging from nanometers to micrometers and as a result, diffusion occurs simultaneously in each type of pore and thus at different rates [7]. To account for pore size variation, a bidisperse model was developed by Ruckenstein et al. [20]. The model assumes a spherical porous particle that consists of a certain number of smaller, equal-sized and porous spherical particles, so there are two types of pores, macro-pores between smaller particles and micro-pores within smaller particles. Under these assumptions, diffusion is modeled as a two-stage process with the first step being fast diffusion into macro-pores and the second step being slower diffusion into micro-pores. Consequently, both macro-pore and micro-pore diffusion coefficients can be evaluated with this model using an analytical solution [20]. By analyzing the shape of the $\frac{M_t}{M_\infty}$ vs. \sqrt{t} plot, three types of the two-stage process can be identified; (1) macro-pore diffusion dominating, (2) macro-pore diffusion much faster than micro-pore or (3) equally important micro-pore and macro-pore diffusion. Comparison of this model to the unipore model shows differences up to 5 orders of magnitude between diffusion coefficients of methane in macro-pores and micro-pores [12]. Although Clarkson and Bustin [19] improved the bidisperse model to enable its use for high-pressure volumetric adsorption/desorption experiments, only average micro-pore and macro-pore diffusion coefficients can be estimated, with no information on the spatial variation, which is important for smaller scale heterogeneity studies.

The second type of method is by numerical simulation, e.g. Lattice Boltzmann Method (LBM) and pore network modeling. Chen et al. [6] used LBM to simulate simultaneous Darcy flow and Knudsen diffusion in pore structures that are reconstructed from scanning electron microscopy (SEM) images of shale samples. An effective Knudsen diffusion coefficient was determined from the resulting concentration fields. This method also provides a way to quantify the contributions of viscous and diffusive flow to total

flow and enables visualization of the diffusion coefficient variation from the concentration fields. However, when using this method, the reconstructed shale samples are usually very small, hundred nanometers length scale, which may have limited representativeness of real shales. In addition, LBM simulations are also limited by their own inherent problems, e.g. numerical instability, computational efficiency and etc. [21,22]. For the pore network modeling method, previous studies on shale matrix diffusion can be found in the works by Mehmani et al. [23] and by Huang et al. [24].

The third commonly used method is micro-CT imaging. Micro-CT imaging can distinguish between solid phases and pore space in reservoir rock. In addition, with the benefit of attenuating fluids, known as tracers, the spatial distribution of different phases of the fluids in the pore space can also be visualized. Micro-CT is an ideal tool to non-destructively study porous media over representative volumes at high resolution and it has been used extensively to study reservoir rock porosity [25,26], fracture characterization [27,28], morphology and topology [29,30], and multi-phase flow [31–35]. A review of micro-CT imaging techniques and imaging-based mass transport modeling can be found in Bultreys et al. [36] and [37].

A few publications have used micro-CT to quantitatively study the dynamics of diffusion processes [15,17,38–42]. The basic concept of these applications is to monitor the diffusion process by acquiring a series of time-sequenced micro-CT images that show linear attenuation coefficients changing over time, which are then used to calculate concentration and density data using linear absorption theory [40]. An X-ray attenuating fluid, e.g. potassium iodide (KI) or sodium iodide (NaI) solution, is used as a tracer and its concentration variation due to diffusion can be tracked in 4 dimensions using micro-CT, where the 4th dimension is time. For bulk diffusion coefficient measurements the sample is often considered to be homogeneous and a curve-fitting procedure is adopted. A concentration profile generated from either numerical simulation or analytical solution is fitted to concentration profiles extracted from the micro-CT data. Cavé et al. (2009) measured the one-dimensional bulk diffusion coefficient of shale for liquid using this method and also compared results with experimental measurements. Tidwell et al. (2000) investigated the effects of spatially heterogeneous porosity on matrix diffusion of liquid in centimeter-scale dolomite using a similar approach. However, the mathematical model they used was one-dimensional and thus diffusion in other dimensions during the experiment could not be accounted for.

Measurements of local diffusion coefficients are more complex. The rock sample must be considered as heterogeneous and thus a spatially-varying diffusion coefficient must be considered in Eq. (2). Image quality is a precondition to guarantee the accuracy and reliability of the results, which implies that experimental design, image acquisition and image processing are of utmost importance. Only a few publications on local diffusion coefficient measurement using micro-CT can be found in the literature; however, there exists no comprehensive analysis of the method for reservoir rocks. Liu et al. [39] calculated the local diffusion coefficients of one-dimensional CO₂ diffusion through n-decane in an artificial porous medium (glass-bead pack, with porosity of 36.8%) using a non-iterative finite volume method. However, due to the nanometer length scale pores in shale and heterogeneity, more considerations are required for experiment design, image processing and mathematical model validation to ensure the reliability of results. Also, the measurement of diffusion coefficients from micro-CT data requires a comprehensive validation of the method and testing of parameters that may influence the results.

In this paper, a 3D liquid diffusion experiment is conducted and visualized using 4D dynamic micro-CT imaging. We develop a novel method to measure local diffusion coefficients from the

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