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A model of dynamic adsorption–diffusion for modeling gas transport and storage in shale

Zehao Yang [a], Weihong Wang [b], Mingzhe Dong [c,*], Jinjie Wang [a], Yajun Li [a], Houjian Gong [a], Qian Sang [a]

[a] College of Petroleum Engineering, China University of Petroleum, Qingdao 266555, China
[b] State Key Laboratory of Shale Oil and Gas Enrichment Mechanisms and Effective Development, Sinopec Research Institute of Petroleum Exploration and Production, Beijing 100083, China
[c] Department of Chemical and Petroleum Engineering, University of Calgary, Calgary T2N 1N4, Canada

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ABSTRACT

Understanding gas transport and storage processes is essential for analysis of reservoir accumulation mechanisms and evaluation of a shale formation. Because organic matter such as kerogen is widely distributed in shale, it plays an important role in gas diffusion and adsorption. Although many mathematical models considering gas diffusion and adsorption have been proposed and evaluated, very few models consider the effect of organic matter on the dynamic adsorption process, and research studies systematically combining a mathematical model history-matched to experimental data is also rare. In this study, a dynamic, approaching equilibrium (hereinafter referred to as "delayed") adsorption–diffusion (DAD) method is presented to analyze gas transport and storage processes in crushed particles of three different sizes. The delayed effect for adsorption results from the dynamic mechanisms of gas dissolution and adsorption in the semi-liquid layer of organic matter. The mathematical model for this phenomenon is based on dynamic adsorption experiments with a constant pressure condition. The general and approximate solutions for the DAD model are obtained to estimate the physical parameters through a multilevel single-linkage method. From the fitting results of the DAD method, it is found that the absolute permeability begins to decrease when particles are crushed into smaller sizes and observed that particle size plays more important role than permeability in the diffusion process for crushed shale samples. Isotherm measurements for total gas and free gas reveal that the difference in gas content between total gas and adsorbed gas become greater as pressure increases. Sensitivity analyses for the model disclose that the apparent diffusion coefficient and adsorption/desorption rate coefficient determine gas transport and storage processes together. Analysis and comparison of a diffusion model, an instantaneous adsorption–diffusion (IAD) model, and the DAD model reveal that the former two models are special forms of the DAD model, and the sequence of equilibrium times required for the gas transport and storage processes is: DAD > IAD > diffusion model.

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1. Introduction

Gas production from organic rich shale has become a very important source of fossil energy in the United States [1]. Many other countries such as China, Canada, and Germany are currently investigating the potential of this promising unconventional resource in both theory and practice. However, a shale gas reservoir is a very complex system with unique storage characteristics, making it more challenging to analyze gas transport and storage processes as compared to a conventional gas reservoir. The natural gas in shale includes not only free gas stored in the pore structure, but also the condensed phase gas storage, including adsorbed gas on the surface of pores within the organic matter and clay minerals, as well as dissolved gas in organic materials such as kerosene and bitumen [2–4]. This requires a method to analyze a shale gas system, which is quite different from a traditional gas reservoir. Understanding gas transport and storage in a shale matrix is important for analyzing reservoir accumulation mechanisms and evaluating the characteristics of a shale formation. It also important for optimizing a production strategy [5–7]. A new and improved method therefore needs to be developed to better analyze a shale gas system.

Understanding the pore structure in shale is critical to characterizing its storage phenomena, which can affect selection of

* Corresponding author at: University of Calgary, Calgary, Alberta T2N 1N4, Canada. Tel.: +1 403 210 7642; fax: +1 403 284 4852.
E-mail address: mingzhe.dong@ucalgary.ca (M. Dong).

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simulation techniques for modeling gas transport and storage processes in shale. Many methods are available to detect the pore structure in shale gas samples, including X-ray powder diffraction (XRD) [8], transmission electron microscopy (TEM) [8], scanning electron microscopy (SEM) [1,9–11], atomic force microscopy (AFM) [12], nuclear magnetic resonance (NMR) [13], N_2 sorption test [9,14], and ultra-high pressure mercury injection [15,16]. These methods have found that organic matter plays a very important role in gas storage in shale. Many nano-scale pores exist in shale, which provide the adsorption sites for CH_4 due to the attendant large surface area. At the same time, gas dissolves into the organic matter just as natural gas dissolves into heavy oil. It has also been found that a large number of natural gas molecules might adsorb onto the surface of inorganic matter such as clay [14,17–19]. It can be seen from Fig. 1a that many micro- and nano-pores exist in inorganic matter. Fig. 1b is a magnified SEM image for the observation point in Fig. 1a. It displays that most of the pores in organic matter are as small as nanometer scale, and that the number of these pores is very large. Finally, it also demonstrates that organic matter is widely distributed in the shale given that the observation point is arbitrary.

Many researchers have recently studied the diffusion mechanism in shale [12,15,20–23]. The importance of adsorption characteristics of a shale system has also been realized [2,24–30]. Some systematic gas transport and storage models, which consider both the processes of free gas diffusion and gas adsorption, have also been proposed [7,31–35]. However, very few research studies employ models that focus on the effect of organic matter on the dynamic adsorption process.

In this work, a dynamic, approaching equilibrium (hereinafter referred to as "delayed") adsorption–diffusion (DAD) method is presented to analyze shale gas storage and transport processes in crushed spherical shale samples. It considers that gas expansion, gas adsorption, and gas dissolution take place at the same time when gas diffuses into the shale pore structure. Because the thickness of organic matter is as small as nano-scale [1] and therefore very hard to determine precisely, one assumes that the effect for organic matter is that there is a delay in the adsorption process attaining equilibrium, and that the gas dissolution process in organic matter is not considered alone. So, the adsorbed gas mentioned in this study contains both the gas adsorbing on the surface as well as dissolved gas. And, because the organic matter is widely distributed in the shale, one also assumes that this delay effect takes place on all the adsorption surfaces in the pore (the reason for the delay effect on adsorption will be further discussed at the beginning of Section 2). The mathematical model for gas transport and storage processes considering the delay effect is then formulated and assembled. The general and approximate solutions for the DAD model are obtained and used to estimate physical parameters of shale through combining experimental results and a global optimization method. The DAD model is compared with the diffusion model as well as the instantaneous diffusion–adsorption (IAD) model to make the characteristics of our model clear. The DAD model is robust and easy to use, saving time and expense for laboratory-based shale gas evaluation and characterization.

2. Theory

Before presenting the mathematical model in detail, we discuss some basic concepts for the DAD method.

The process of physical adsorption on a surface often attains equilibrium very fast [36–39]. When gas diffuses into a spherical shale particle, the adsorption sites of the inner surface will immediately reach their corresponding equilibrium adsorption capacity [7,40]. And when the gas diffusion process reaches equilibrium, the adsorption capacity of all adsorption sites have attained and are equal to the overall equilibrium adsorption capacity. Most previous methods for analysis of shale gas storage and transport are based on this presumption of an instantaneous adsorption–diffusion (IAD) physical process. This is because the influence of organic materials such as kerogen on the adsorption process is often not considered in their models [7,31–34]. In the DAD model, it is assumed that gas begins to dissolve into the organic material as soon as gas adsorbs onto the adsorption sites. The dissolution process may therefore delay the adsorption process from instantly attaining the respective equilibrium adsorption capacity [6,41,42]. Do and Wang [43] also found this delayed phenomenon in their study on activated carbon through both experimental observations using electron microscopy along with the analysis of the adsorption and desorption equilibrium data. They argued that the adsorption in the semi-liquid layer on the surface of organic matter is quite heterogeneous, resulting in adsorption not occurring instantaneously; i.e., it may take some time to attain equilibrium. This means the time scales for the gas adsorption process and the gas diffusion process are comparable [6,43]. Therefore, the mechanisms of dissolution and adsorption in the semi-liquid layer of organic matter result in the delayed effect of the gas transport and storage processes.

The physical adsorption process is reversible [36–39], which means that gas molecules dynamically adsorb and desorb on the adsorption sites simultaneously. When the number of "forward" (i.e., adsorbing) molecules is greater than that of "backward" (i.e., desorbing) molecules in a given time, the net adsorption capacity will increase; this phenomenon occurs in the gas transport and storage processes [44]. In the DAD model, physical adsorption is considered to be reversible, and the rate of the forward adsorption process is proportional to the concentration of free gas available to

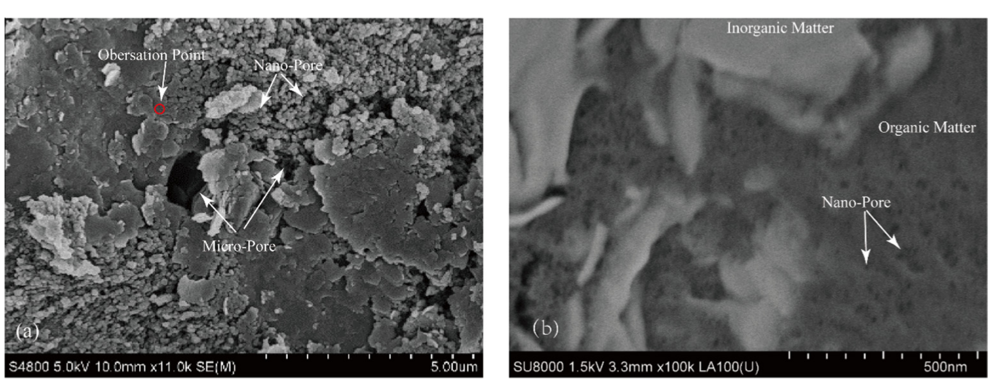

Fig. 1. (a) An SEM image displaying distribution of inorganic micro- and nanopores in inorganic matter. (b) A magnified SEM image for the observation point in (a) revealing the distribution of organic and inorganic matter, and depicting nanometer scale pores in organic matter.

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