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Numerical modeling and experimental validation of anomalous time and space subdiffusion for gas transport in porous coal matrix

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ABSTRACT

Accurate description of gas transport in porous coal matrix is one critical issue for coalbed methane production. However, the adequacy of existing Fickian diffusion-based models for gas transport in heterogeneous coal matrix is debated. In this study, taking into account the basic topological complexity inherent to porous coal matrix and the strong adsorption effect of coal on gas molecules, an anomalous subdiffusion model with fractional time and space derivatives is proposed to characterize the mechanism of gas transport in heterogeneous coal matrix. The fractional diffusion equation is discretized and solved by using an implicit numerical scheme which is based on the generalization of standard finite-difference method. Furthermore, gas adsorption and desorption experiments with two coal samples collected from China were carried out to validate the anomalous subdiffusion model. It is revealed that the anomalous subdiffusion model with merely three parameters can reproduce the dynamic process of gas transport with better accuracy than existing Fickian diffusion-based models, suggesting the anomalous time and space subdiffusion to be the governing process of gas transport in coal matrix. Finally, the parametric sensitivity analysis shows that the introduction of fractional parameters in the present model is essential to accurate description of gas transport in heterogeneous coal matrix.

1. Introduction

Coalbed methane (also known as gas in coal mine) is a kind of clean energy, but also a greenhouse gas with the greenhouse effect being 20 times that of carbon dioxide. Motivated by the coalbed methane exploitation as well as the environmental improvement, gas storage and transport in coal seams has received considerable attention during recent decades, and continues to be a topic of intense research [1]. Coal seams differ significantly from conventional gas reservoirs in that most gas in coal seams is originally adsorbed on the surface area of porous coal matrix rather than stored as a free gas [2]. Accordingly, the dynamic process of gas transport in coal seams occurs at two scales: gas adsorption and desorption within the coal matrix and gas flow through the coal cleats [3]. In the literature [4,2,3], gas adsorption and desorption process in the coal matrix is assumed to be concentration-driven, whereas gas flow through the coal cleats is considered to be pressure-driven. The coal matrix plays a role as the gas source while the coal cleat network provides main paths for gas flow.

In terms of the pore diameter and the mean free path of gas molecules, three different mechanisms have been identified for transport process of an adsorbing gas in the coal matrix: bulk diffusion, Knudsen diffusion and surface diffusion. Bulk diffusion takes place when the pore diameter is greater than 10 times the mean free path of gas molecules. In this case, the gas molecules collide more frequently with other molecules than with the pore walls, and the diffusional resistance comes primarily from intermolecular collisions [5]. On the other hand, Knudsen diffusion prevails when the pore diameter is less than 10 times the mean free path. In this case, the diffusional resistance is not due to the collision between gas molecules, but rather due to gas molecules colliding with pore walls [6]. In the intermediate regime, surface diffusion occurs and both wall collisions and intermolecular collisions contribute to the diffusional resistance [7]. As a whole, there will be a transition from Knudsen flow at lower pressures to bulk diffusion at higher pressures.

In numerical modeling of gas transport in coal matrix, several mathematical models based on Fickian diffusion law have been developed. One of the simplest model used in engineering

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applications is the equilibrium model, which treats the gas diffusion process as an instantaneous diffusion [8]. This kind of diffusion model represents the matrix response to changes in gas pressure in a lumped parameter fashion, neglecting the true spatial variation of gas concentration. Another widely used model is the unipore diffusion model, which is derived from the Fickian second law for spherically symmetric flow. This kind of diffusion model treats the gas diffusion process as one-step diffusion and accounts for the effects of the gas concentration gradient. It has been shown that for the purpose of making a first estimate of the adsorption rates in a specific coal reservoir, the unipore model can describe the diffusion process to some extent [9]. However, the unipore model assumes a homogenous pore structure in the coal matrix, which leads to significant deviations between numerical modeling and experiment data [10]. Thus, the unipore diffusion model is inadequate for modeling gas diffusion in coal matrix.

Historically, some studies reported that coals may have a bimodal pore size distribution, with significant fractions of the total pore volume being found in size greater than 30 nm and less than 1.2 nm [11]. In view of this property, many researchers adopted the bidisperse diffusion model developed by Ruckenstein [12] for pelletized particles to describe the gas transport in coal matrix[10]. This kind of diffusion model conceives the bimodal pore structure as a macroporous particle which consists of microporous particles of uniform size, and assumes two-step diffusion process in a coal matrix [7]. In contrast to the unipore model, the bidisperse diffusion model provides a better description of the adsorption and desorption processes [13]. However, one key issue for bidisperse diffusion model is how to obtain a reasonable representation of the heterogeneous pore structure for numerical modeling [8]. Furthermore, it has been found that multi-modal pore size distributions may exist for some bituminous coals, for which the gas transport may undergo a multi-step diffusion process rather than two-step diffusion process [14]. Accordingly, the adequacy of bidisperse diffusion model for gas transport process in heterogeneous coal matrix is debated.

Recently, an increasing number of diffusion phenomena in nature were recognized not to fit into the relatively simple description of diffusion governed by Fickian law, which were commonly termed “anomalous diffusion” in the literature [15]. Examples of anomalous diffusion include the contaminant transport in groundwater [16], tracer transport in nonhomogeneous media [17] and particle dynamics in disordered systems [18]. In fact, although termed “anomalous”, anomalous diffusion processes are ubiquitous and have grown so compelling as to prompt punchlines such as “anomalous is normal” [19]. In modeling anomalous diffusion processes, the so-called fractional diffusion equations play an important role with the advantage that they take into account the initial-boundary value problems in a straightforward way [20].

The starting point of the fractional derivative model is usually a classical differential equation which is modified by replacing derivatives of an integer order in the constitutive equations by derivatives of fractional order [21]. In the last decades, the theory of fractional derivatives has been successfully applied to problems in physics, chemistry, biology, medicine, finance etc. [22]. Especially, much success of fractional diffusion equations has been achieved in the description of anomalous diffusion, such as the calcium spark in cardiac myocytes [23], non-self-similar infiltration in porous media [24], neutron motion in nuclear reactors [25] and the infection pathway of the virus in the cytoplasm [26], to mention just a few. In general, anomalous diffusion processes are encountered in complex systems with geometric constraints like porous media and doped crystals, or coherent structures like turbulence and chaotic dynamics.

From an overview of the references, it can be found that (i) the existing diffusion models have obvious disadvantages in modeling the mechanism of gas transport in heterogeneous coal matrix; (ii) due to the basic topological complexity inherent to porous media, gas transport in coal matrix may no longer obey Fickian diffusion law, but the anomalous diffusion theory; (iii) fractional diffusion equation has gained much success in the description of anomalous diffusion phenomena. Motivated by these facts, this study develops an anomalous subdiffusion model with fractional time and space derivatives to describe gas transport in heterogeneous coal matrix. The accuracy of anomalous diffusion model is verified by comparing the numerically simulated results with those of existing Fickian diffusion-based models and experimental data. In addition, the effects of model parameters on the gas transport are also discussed.

2. Mathematical formulation of anomalous subdiffusion model

2.1. Fickian diffusion-based models

Two different Fickian diffusion-based models have been successfully used in prior studies to describe the process of gas transport in coal matrix, namely unipore diffusion model and bidisperse diffusion model.

The unipore diffusion model assumes a homogeneous and uniform pore structure in coal matrix and the diffusion equation is given as

$$\frac{\partial C}{\partial t}=\frac{D_0}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial C}{\partial r}\right) \tag{1}$$

where C is the gas concentration, r the particle radius, t the diffusion time and D_0 the diffusivity. For a constant surface concentration and isothermal conditions, the fraction of gas desorbed can be calculated as

$$\frac{V_t}{V_\infty}=1-\frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-\frac{n^2\pi^2D_0t}{r_p^2}\right) \tag{2}$$

where V_t is the total volume of gas desorbed at time t, V_∞ the total desorbed mass in infinite time and r_p the diffusion path length. Due to the assumption that all pores are of the same size, the unipore model is inadequate for materials with heterogeneous pore structures and would overestimate the quantity of gas desorbed [10].

The bidisperse model takes into account both the diffusion in macropore and micropore of coal matrix using a simplified approach, in which the coal matrix is assumed as a sphere comprised of microporous spheres and the macropores are the volume around these microspheres [12]. The governing equations describing the diffusion in macropores and microporous spheres are written respectively as

$$\frac{D_a\epsilon_a}{r_a^2}\frac{\partial}{\partial r_a}\left(r_a^2\frac{\partial C_a}{\partial r_a}\right)=\epsilon_a\frac{\partial C_a}{\partial t}+S_a\frac{\partial C_{sa}}{\partial t}+4\pi nR_i^2\epsilon_iD_i\left(\frac{\partial C_i}{\partial r_i}\right)_{r_i=R_i}, \tag{3}$$

$$\frac{D_i\epsilon_i}{r_i^2}\frac{\partial}{\partial r_i}\left(r_i^2\frac{\partial C_i}{\partial r_i}\right)=\epsilon_i\frac{\partial C_i}{\partial t}+S_i\frac{\partial C_{si}}{\partial t} \tag{4}$$

where the terms on the left hand side are due to the diffusional flux based on Fickian second law in the macropore and micropore, the first and second terms on the right hand side are the concentration variation due to accumulation in the pore volume and on the pore surface respectively, and the third term on the right hand side of Eq. (3) is due to the diffusional flux at the surface of the microspheres. Here, the subscripts ‘*a*’ and ‘*i*’ denote the variables for macrosphere and microsphere, respectively. Assuming the linear isotherms and equilibrium between each step, the relationship between free gas concentration and the adsorbed phase concentration satisfies