Fuel 182 (2016) 314–322

Contents lists available at ScienceDirect

Fuel

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

Full Length Article

Fractal characteristics of coal samples utilizing image analysis and gas adsorption

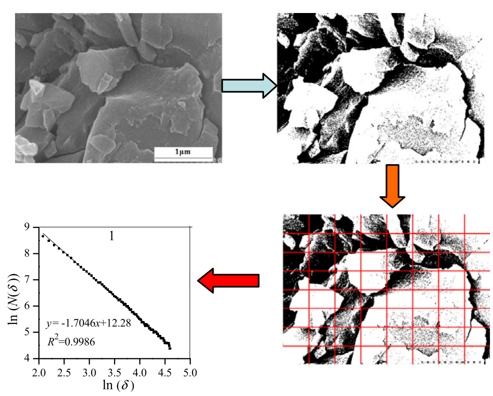
Liu Xianfeng *, Nie Baisheng *

State Key Lab of Coal Resources and Safe Mining, China University of Mining & Technology (Beijing), Beijing 10083, China
School of Resource and Safety Engineering, China University of Mining & Technology (Beijing), Beijing 100083, China

HIGHLIGHTS

- Fractal characteristics of coals were analyzed using SEM and gas adsorption.
- Both V_L and P_L were considered when assessing the adsorption capacity of coals.
- Coalification has significant effect on the fractal dimensions of coal samples.
- D_1 and D_2 have different impacts on CH_4 adsorption.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 12 October 2015
Received in revised form 22 May 2016
Accepted 23 May 2016

Keywords:
Image analysis
LP-N_2GA
Fractal dimension
Methane adsorption
Coalification

ABSTRACT

Coal is a porous medium with fractal characteristics. In order to investigate the effect of fractal dimensions on methane adsorption capacity, fractal characteristics of 11 coal samples were analyzed, using scanning electron microscopy (SEM) and low-pressure nitrogen gas adsorption (LP-N_2GA). Data from SEM image analysis and LP-N_2GA experiments were applied to assess the heterogeneities of pore distribution (D_1) and the irregularities of coal surface (D_2) on the basis of box-counting method and Frenkel–Halsey–Hill (FHH) theory, respectively. The relationship between fractal dimensions and coalification was investigated. Based on the physical description of fractal surfaces and pore distributions, the influence of fractal dimensions (both D_1 and D_2) on CH_4 adsorption characteristics was also discussed. The results indicate that these coal samples have different CH_4 adsorption characteristics and fractal geometries, with D_1 ranging between 1.5380 and 1.8267, and D_2 varying from 2.2656 to 2.6541. The U-shaped curve relationship between D values (including D_1 and D_2) and volatile matter (V_{daf}) is observed, demonstrating that coalification makes coal surfaces and pore networks comparatively smoother and more regular for lower rank coals (V_{daf} > 15%), but rougher and more complex for higher rank coals (V_{daf} < 15%). The Langmuir volume (V_L) shows a positive linear correlation with the fractal dimension D_2 values, but little correlation with D_1 values. While, the Langmuir pressure (P_L) is affected by both D_1 and D_2. Fractal dimensions comprehensively reflect the difference in physical properties of coal, which can be used to evaluate CH_4 adsorption capacity. Fractal analysis is of great significance for better understanding of the surface irregularity and methane storage capacity of a coal reservoir.

* Address: School of Resource and Safety Engineering, China University of Mining & Technology (Beijing), Beijing 100083, China.
E-mail addresses: cumtblxf@163.com (X. Liu), bshnie@cumtb.edu.cn (B. Nie).

http://dx.doi.org/10.1016/j.fuel.2016.05.110
0016-2361/

1. Introduction

With the increasing demands on clean energy and natural resources, the enhanced production of gas from the unconventional reservoirs, such as coalbeds, shales and mudstones, is attracting the growing interest all over the world [1–5]. As a potential reservoir for coalbed methane (CBM), in contrast to the conventional petroleum reservoir, coal seams normally contain large amounts of adsorbed gases [6–8]. The adsorption capacity of CBM gases depends heavily on the pore structure in coals, which have a great number of pores and cleats [9,10]. The complicated pore system in coals to a great extent affects not only the storage of gas, but also the gas diffusion in coal seams [11–14]. Thus, a good knowledge of the nature of porosity in coals can provide valuable information about the gas storage and flow mechanisms.

Coal is a natural polymeric material with complex porous structures. It is widely, though not universally, accepted that the pores in coals have a broad size distribution and form a constricted, interconnected network [15–17]. The pore size distribution of coals commonly ranges from micropore (pore width < 2 nm) to mesopore (2 nm $\leqslant$ pore width $\leqslant$ 50 nm) and macropore (pore width > 50 nm), according to the International Union of Pure and Applied Chemistry (IUPAC) classification standard [18,19]. Previous studies have shown the relationship between pore structure and adsorption capacity of coals [20–23]. Coal porosity and pore size distribution vary with the maturity of coals [24,25]. Micropores play a dominant role in gas adsorption and a positive correlation between micropore volumes and methane capacity of coals is observed [26–30]. Clarkson and Bustin [31] investigated the influence of composition on micropore capacity and size distribution in Canadian bituminous coal, proving that the number of micropores is positively correlated with vitrinite content and that micropore heterogeneity increases with an increase in inertinite and mineral matter content. Pan et al. [32] and Deng et al. [33] also suggested that coal rank, mineral composition and deformation structures have a significant effect on microstructures of coals. Wang and co-workers [34] found that coalification makes pore structure more complex and pore surface rougher and that the fractal dimensions of seepage pores and adsorption pores gradually increase with increasing coal rank for middle-high rank coals.

Fractal geometry, first coined by Benoit Mandelbrot in 1982 [35], has proven to be a powerful analytical tool, which can effectively characterize the pore irregularity and surface roughness for porous materials, including coal and shale [36–38]. The use of a fractal method can help gain clearer insights into the relationship between the pore/surface structures and the methane adsorption capacity, because the storage and transportation of CBM are closely associated with the pore and surface self-similarity of coal. Work by Sun et al. [39] indicated that shallow-deposited coal usually displays low pore surface fractal dimensions (*D*s), whereas coal collected from deep layers exhibits large *D*s values. Methane adsorption capacity is determined by the *D*s value on a large scale and a higher *D*s value could correspond to an increased capacity of methane adsorption. Wang et al. [40] studied the fractal geometry of lacustrine shale from the Songliao Basin in China. They stated that small pores are more homogeneous than larger pores, and fractal dimensions increase with increasing BET surface areas and total pore volumes, and increase with decreasing average pore diameters. Based on the physical description of the fractal surfaces, pore structure and fractal dimension were investigated by Correa et al. [41] and Yang et al. [42], indicating that micropores have a greater effect on fractal dimension than mesopores and macropores, and fractal dimension is negatively correlated with permeability. Previous research has demonstrated that the fractal dimension of coal can be acquired by various methods, including mercury intrusion [34,43,44], transmission electron microscopy (TEM) [45,46], scanning electron microscopy (SEM) [47,48], nuclear magnetic resonance (NMR) [49,50], field-mission SEM [40,51], small-angle X-ray scattering (SAXS) [52–54], and gas adsorption [39,42,55]. Among these fractal methods, low-pressure N_2 gas adsorption (LP-N_2GA) analysis has been shown to be an effective method of characterizing fractal dimensions of coal [34,40,56]. In addition, the SEM images of coal microstructure can also provide a great deal of information about pore shapes, pore size distribution and inhomogeneity of pore distribution. However, only a few measurements have been reported in the literature for the fractal characterization from the SEM images of coal pore structures, and little attention has been paid to the relationship between methane adsorption features and fractal characteristics of coal [48,57].

The major goals of this work are to investigate the fractal characteristics of different rank coal samples. The fractal dimension used to represent the fractal characterization was determined from the both LP-N_2GA and SEM image analysis. Subsequently, based on the physical significance of the fractal dimension, the impact of fractal dimension on the methane adsorption features is also discussed. These data and research results could be useful for better understanding the pore systems and CBM storage and diffusion in coalbeds.

2. Coal sample preparation and experiments

Eleven samples with various coal maturity were taken from working faces of different coalfields in the northern China mining area. These coalfields, such as Qinshui Coalfield and Pingdingshan Coalfield, contain good CBM generation potential and reservoir conditions. Proximate analysis of these samples was conducted following the China National Standard GB/T 212-2008. Detailed information including coal rank, moisture (M_{ad}), ash (A_{ad}), volatile (V_{daf}) and fixed carbon (FC_{ad}) content of collected samples can be obtained in the Ref. [24].

The methane adsorption measurements were carried out using the high-pressure volumetric method, following the MT/T 752-1997 method of China for the determination of the methane adsorption capacity in coal [58]. Our experimental method was described previously [10,13]. Approximately 100 g of coal samples with 60–80 mesh (0.177–0.250 mm in size fraction) was weighed and placed in a vacuum drying oven. These coal samples were dried at 373 K for 6 h under vacuum (<4 Pa) prior to test. After the samples were cooled to the room temperature and placed in a coal sample tank for evacuation, the methane adsorption experiments were performed in the equilibrium pressure range of 0–5.5 MPa at ambient temperature (298 K).

According to the China National Standard GB/T 20307-2006, the surface morphological characteristics of samples were determined by SEM. In the laboratory, the coal rock samples were cracked into 1–2 cm^3 small cubic blocks, and a relatively smooth natural fracture surface was chosen as the observation face. These samples were observed using the HITACHI S-4800 SEM equipment. Images of eleven coal samples were shown in the Ref. [24].

The low-pressure N_2 gas adsorption (LP-N_2GA) experiments were performed on 60–80 mesh samples (ASTM standard) at 77 K with an Autosorb-6B/3B instrument, produced by Quantachrome Instruments Co., Florida, USA. Detailed description of the LP-N_2GA experiments and N_2 adsorption/desorption isotherms for the coal samples were demonstrated in the Ref. [24].

3. Theory

3.1. Evaluation of methane adsorption capacity

It has been confirmed that at low equilibrium pressure, methane adsorption on the coal surface follows the monolayer