Fuel 132 (2014) 12-19

Contents lists available at ScienceDirect

Fuel

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

Partial coal pyrolysis and its implication to enhance coalbed methane recovery, Part I: An experimental investigation



Yidong Cai^a, Dameng Liu^{a,*}, Yanbin Yao^a, Zhentao Li^a, Zhejun Pan^b

^a Coal Reservoir Laboratory of National Engineering Research Center of CBM Development & Utilization, China University of Geosciences, Beijing 100083, China ^b CSIRO Earth Science and Resource Engineering, Private Bag 10, Clayton South, Victoria 3169, Australia

HIGHLIGHTS

• Feasibility of partial coal pyrolysis for ECBM was evaluated.

• Improvement of petrophysics for coal reservoir was discussed.

• A suitable temperature for ECBM was selected.

ARTICLE INFO

Article history: Received 11 February 2014 Received in revised form 6 April 2014 Accepted 25 April 2014 Available online 9 May 2014

Keywords: Coal pyrolysis Coalbed methane Pore structure Permeability

ABSTRACT

This paper examines the feasibility of combining a process known as enhanced methane recovery with partial coal pyrolysis to improve the petrophysics of coal seams and ultimately extract higher methane yields with accompanying pyrolysis gases. Partial pyrolysis for coal gas generation changes the pore and fracture structure, which in turn affect the permeability. A series of laboratory experiments on three coal rank samples monitored the changes in pore structure and permeability accompanying coal pyrolysis. Thermogravimetry-mass spectrometry (TG-MS) analysis evaluated mass loss and product composition. The pore and fracture structure evolution was determined by a combination of mercury intrusion porosimetry (MIP), scanning electron microscope (SEM) and methane adsorption capacity measurements on heat-treated coal blocks of ~100 g. The pore volume and methane adsorption capacity of LRC specimen (0.56% R_{o,m}) with 10 °C/min and a hold time of 30 min experienced slight changes during the heating process from 25 °C to 400 °C, but when heated from 400 °C to 800 °C, the pore volume in the LRC specimen greatly increased and the mercury-determined total porosity went from 36% at 400 °C to 43% at 800 °C. The permeability of the specimens at the temperature range of 300-400 °C increased exponentially with temperature due to the generated pore-fracture system. The sample LRC (800 °C) with the highest mercury-determined pore volume possessed the lowest methane capacity $(19.45 \text{ cm}^3/\text{g})$ due to the maximum adsorption volume of pyrolyzed coal obtained from the Langmuir model was related not only to the pore structure but also to the extent of graphitization. Therefore, they may have significant implications for enhanced coalbed methane (CBM) recovery.

© 2014 Elsevier Ltd. All rights reserved.

1. Introduction

Coalbed methane recovery before coal extraction is very important from greenhouse gas emission, assistance in development of gas industry, safety and economic of mining point of view [1–3]. Enhanced coalbed methane (ECBM) recovery from coal seams may consist of a multi-branch horizontal well, with applications of CO_2 sequestration, hydrofracturing, biotechnology and heating (steam injection and coal burning) etc. to store gases or enhance permeability/methane production. These methods have associated environmental and economic challenges. Coal pyrolysis is an alternative approach and is a component of underground coal gasification (UCG) [4–6]. This method of gas production produces CH₄, H₂ and CO₂ from the thermochemical decomposition and gasification of organics at high temperatures in the absence of oxygen, which is similar as kerogen pyrolysis in petroleum field [7–10]. A similar approach where only the pyrolysis stage is performed could be a promising option for enhancing CBM recovery. Different physical and thermochemical transformations occur at elevated temperatures: (1) the first stage (25–300 °C), is the dry gas phase. Here, moisture and a small amount of adsorbed gases (including CH₄



^{*} Corresponding author. Tel.: +86 10 82320892 (O); fax: +86 10 82326850. *E-mail address:* dmliu@cugb.edu.cn (D. Liu).

and N₂, etc.) desorb from the matrix pores and cleat the system until \sim 200 °C. Thus, the coal structure does not change significantly. From 200 °C to 300 °C, thermal decomposition occurs for the low-rank coals such as lignite; (2) the second stage (300-550 °C), the coal pyrolysis phase has decomposition reactions, forming gases and tars. Before 400 °C, coal may soften, and viscous plastic mass can form. In the range from 400 to 550 °C, coal gas evolves and coal tar precipitates from the thermally decomposed products. The residual substance in coal gradually stiffens and solidifies as char. Gas produced in 450–550 °C range contains light aromatic hydrocarbon and long chain fatty mass. The petrophysics of coals (including pore structure, fractures and permeability etc.) changes during this phase [6,11–13]. The condensation reaction during the process of the formation of char is not very obvious; (3) in the third stage (550-1000 °C), polycondensation reactions occur (also called secondary generation phase), as well as the carbocoal transition phase. During this phase, carbocoal aromatic sizes increase, the arrangement of aromatic layers often become more ordered, true density increases and a high degree of aromatization occurs along with an of increasing metallic luster. The gas generation here is associated with the thermal decomposition of coal molecule functional groups. Complex organics are pyrolyzed into liquids and gases, which flow/burst out of the particles. Additionally, this stress and mass loss creates more pores and fractures. Thus, the permeability of the pyrolyzed coal is changed.

To accurately and continuously acquire the suitable maximum temperature for enhancing CBM recovery, thermogravimetry coupled with a mass spectrometry (TG–MS) was adopted to analyze the organic decomposition products from different rank coals at elevated temperatures (25–1200 °C) to obtain structural information. Secondly, methane adsorption profiles were determined by a programmed gas adsorption analysis rig. At the same time, the pore structures including pore volume, pore size distribution, porosity and permeability of coals during pyrolysis are investigated with increased temperatures from 25 °C to 800 °C. The feasibility of partial coal pyrolysis for low rank coal (LRC) will be focused due to its low gas content and shallow burial depth.

2. Experimental methods

2.1. Coal sampling

Table 1

Three coal samples with a volume of approximately $40 \times 40 \times 40 \text{ cm}^3$ were directly collected from three different rank coals including one low-rank coal (LRC, 0.56% $R_{o,m}$), one medium rank coal (MRC, 1.68% $R_{o,m}$) and one high rank coal (HRC, 2.59% $R_{o,m}$). LRC originated from the southern Junggar basin, NW China; MRC is collected from Ordos basin, North China; and HRC comes from Qinshui basin, North China. All samples were carefully packed and taken to the laboratory for experiments. Each sample was carefully preserved in an intact form.

2.2. TG-MS during coal pyrolysis

Thermogravimetry–mass spectrometry (TG–MS) analysis was conducted by a Rigaku TG–DTA coupled with an Omnistar MS.

Vitrinite reflectance, proximate and ultimate analysis of different rank coals.

TG was linked to the programmed furnace, which can collect the continuous weights of samples and execute the data analysis. The TG has a temperature range of 25–1200 °C. The MS has a Nier type enclosed ion source, two detectors and a triple mass filter, which was controlled by computer. The two rigs were coupled by a transfer line leading from the TG to the MS. Generated gases in the TG rig can flow into the MS rig through the transfer line in a few seconds later. The MS need just very small fractions of the gases [14]. The TG conditions used to study the relationship between the structure and gas compositions were heating rate of 10 °C/min at the temperatures of 25-1200 °C; sweep nitrogen, 60 cm³/min; retention time, 30 mins; constant sample volume weighing 25-50 mg. The MS was scanned over a range of 0-100 amu with measurement intervals of approximately 19 s. The characteristics of the products of coal pyrolysis were determined vs. the elevated temperature in the multiple ion detection modes.

2.3. Microscopy and MIP analysis

Vitrinite reflectance $(R_{o,m%})$ and fracture analyses were conducted as previous research [15]. Coal compositions, proximate and ultimate analyses of used coals are documented in Table 1. The pore morphology was acquired using scanning electron microscope (SEM) and pore structure analysis for different rank coals (LRC, MRC and HRC) at different temperatures (25 °C, 200 °C, 400 °C, 600 °C and 800 °C) was performed with a PoreMasterGT60 which automatically registers pressure, pore radius and mercury injection volume. The SEM pictures of LRC sample was selected to be a representative due to the same trend for all three rank coals. Previous research [16,17] found that coal compressibility has an effect on mercury intrusion porosimetry (MIP) results especially when pressure is greater than 20 MPa. Data processing and the coal compressibility correction procedures are the same as in our previous work [18,19]. Assuming that pores are composed of a variety of cylindrical pores, the relationship between the pore radius and pressure can be acquired by the Washburn Equation [20]. On the basis that cumulative mercury injection volume, pore radius and space distribution can be inferred from the measured mercury injection curves.

2.4. Methane adsorption measurement

Methane adsorption on coal samples was conducted at 25 °C with the pressure range from 0 MPa to 10 MPa. Before the adsorption experiment, the crushed sample (60–80 mesh) was dried overnight at 50 °C and then degassed at 25 °C for 1 h under a vacuum. Approximately 100 g were used for each measurement. Methane adsorption isotherms were measured using the Isotherm Measurement System (KT100-40HT). The experimental procedure and the helium calibration for pore volume in coals were described in the previous research [15,21]. Adsorbed methane at a given pressure was similar to the measurement of the pore volume, which was finished when the pressure approached 10 MPa. Each process was repeated two or three times for data accuracy [22,23] and was analyzed using the Langmuir model to obtain adsorption parameters for each sample [24].

Samples No.	$R_{o,m}$ (%)	Proximate	Proximate analysis wt.% (ad)				Ultimate analysis wt.% (daf)			
		М	V	А	FC	С	Н	Ν	St	
LRC	0.56	0.74	9.58	12.71	76.97	74.80	4.49	3.07	1.08	
MRC	1.68	1.19	11.64	6.22	80.95	81.92	4.40	4.03	0.23	
HRC	2.59	2.24	5.34	3.50	88.92	90.96	2.76	1.67	0.13	

Note: M-Moistures; V-Volatiles; A-Ash; FC-Fixed Carbon; C-Carbon; H-Hydrogen; N-Nitrogen; St-Total sulfur; ad-air dry basis; daf-dry ash for free basis.

Download English Version:

https://daneshyari.com/en/article/206184

Download Persian Version:

https://daneshyari.com/article/206184

Daneshyari.com