Fuel 112 (2013) 14–22

Contents lists available at SciVerse ScienceDirect

Fuel

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

Investigation of coalbed methane potential in low-rank coal reservoirs – Free and soluble gas contents

Aihua Liu [a,c,*], Xuehai Fu [a,b], Kexin Wang [d], Hui An [e], Geoff Wang [c,e]

[a] School of Resources & Earth Science, China University of Mining & Technology, Xuzhou, Jiangsu 221008, China
[b] School of Geology and Mining Engineering, Xinjiang University, Urumqi 830047, China
[c] Key Laboratory of CBM Resources and Dynamic Accumulation Process (CUMT), Ministry of Education, Xuzhou, Jiangsu 221008, China
[d] Jiangsu Bureau of Coal Geology, Nanjing, Jiangsu 210046, China
[e] School of Chemical Engineering, The University of Queensland, St. Lucia, Brisbane, QLD 4072, Australia

HIGHLIGHTS

- A prediction method of free gas content in low-rank coal reservoirs was proposed.
- The changes of methane solubility with pressure and temperature were analyzed.
- Prediction showed that free and soluble gas contents increase with burial depth.
- A CBM content evaluation model was established for low-rank coal reservoirs.
- Free and soluble gas contents are considerable in low-rank coal reservoirs.

ARTICLE INFO

Article history:
Received 20 August 2012
Received in revised form 4 May 2013
Accepted 7 May 2013
Available online 24 May 2013

Keywords:
Low-rank coal
Coalbed methane
Methane solubility
Free gas content
Soluble gas content

ABSTRACT

In low-rank coal (lignite and subbituminous coal) reservoirs, it is difficult to investigate the potential of free and soluble gases at different burial depths because of the lack of measuring methods available in practice. In this work, Mariotte's law was adopted to predict free gas content and methane solubility in coal seam water was studied to calculate soluble gas content. Coal samples were collected from Chinese typical low-rank coal-bearing basins. This study shows volume of pores occupied by free gas becomes smaller when moisture content and confining pressure are high. Methane dissolving tests in four coal seam water samples under set temperatures and pressures show that methane solubility increases with increasing pressure and temperature. Pressure seems to be a more effective influencing factor than temperature on methane solubility although temperature effect is enhanced at high temperature and pressure. A mathematical model of *in situ* methane content containing adsorbed, free and soluble gases, was established to evaluate the *in situ* gas content of low-rank coal reservoirs at burial depths from 600 m to 1400 m. While the *in situ* gas content of the studied coal reservoirs increases with burial depth, the percentage of the free and soluble gases in the *in situ* contents ranges from 8% to 34%, and hence have to be taken into account in the evaluation of coalbed methane (CBM) potential of low-rank coal reservoirs for CBM recovery.

1. Introduction

Free and soluble gases are usually ignored in the evaluation of coalbed methane (CBM) potential for CBM recovery from coal reservoirs. This is generally acceptable for most middle- and high-rank coal reservoirs due to their limited methane existing at free and/or soluble states [1–5]. However this is not the case for most low-rank coal reservoirs. A recent survey conducted on several low-rank coal reservoirs confirmed the significant content of free and soluble gases in China [4,6,7]. Half of the total coal reserves in China are identified as low-rank coals [8–10]. The low-rank coal reservoirs contain a large amount of CBM in total although the gas content of low-rank coals is usually lower than that of high-rank coals. This represents an important reserve of unconventional natural gas that can be recovered. Thus, it is paramount to accurately evaluate free and soluble gases in low-rank coal reservoirs.

A low-rank coal reservoir is characterized with high permeability, low absorbability but preferable storage capability of free gas [11,12]. Ignoring free gas results in underestimation of the actual gas content [13,14]. Free gas content has relations with factors of

---

* Corresponding author at: School of Resources & Earth Science, China University of Mining & Technology, Xuzhou, Jiangsu 221008, China. Tel.: +86 15162110043.
E-mail address: liuaihua0129@163.com (A. Liu).

0016-2361/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.fuel.2013.05.032

in situ porosity, initial water saturation and gas pressure in original position. However, the measuring methods are not available in practice [15,16]. Soluble gas is methane dissolved in coal seam water. The interspaces of low-rank coal reservoirs are mostly filled with water. The solubility of methane is low at ambient temperature and atmospheric pressure. However under certain temperatures and pressures, the soluble methane takes a large part of the total CBM content [4,6,11]. Solubility of methane in pure water, deionized water and aqueous electrolyte solutions at high temperatures and high pressures was measured in view of natural gas migration and release [17–21]. The experiments of methane dissolving in water revealed that the methane solubility is controlled by temperature, pressure and salinity of coal seam water [16,22,23], mainly described by Henry's law at present [12,15]. Comparatively, coal seam water is a more complex electrolyte solution containing various salts. The solubility of methane in coal seam water has not yet been studied. Therefore, an analysis of its soluble characteristics is highly desirable.

A series of experiments were conducted and comprehensive analyses of field data were performed to investigate the influence of free and soluble gases on CBM potential of low-rank coals. Coal porosity features and pore volume changes under stress were analyzed. Mathematical models were proposed to predict contents of free and soluble gases in low-rank coal reservoirs. Free gas content was calculated by Mariotte's equation at different burial depths. Water samples from a lignite coal seam in the Hailar Basin were obtained and their methane solubility was examined to predict soluble gas content. Gas contents of 16 drilling samples were measured and compared with predicted values to check the rationality of this new predicting method.

2. Experimental methodology

2.1. Sampling

In total 12 coal samples are investigated in this study. These include four coal samples collected from Hailar Basin in Inner Mongolia, 6 from Turpan-Hami Basin and 2 from Junggar Basin in Xinjiang autonomous region (see Table 1). All coal samples were collected in accordance with China National Standards GB/T 482-2008. All samples were collected from active mine faces, and approximately 5 kg of coal was collected for each channel sample. All three regions are typical representatives of low-rank coal reservoirs in the north of China. The selected coal samples and corresponding four water samples shared the same sample numbers HLR-02, HLR-05, HLR-06 and HLR-07, and were taken from a lignite coal seam in the Yimin Formation as well as its roof and floor under different burial depths of Yimin depression in the Hailar Basin as shown in Fig. 1. Hailar Basin formed in Cretaceous Mesozoic time. Yimin Formation is one of the main strata with low-rank coal and bituminous coal deposits. The coal measures are on average 33 m thick in the Yimin Formation, with reported maximum vitrinite reflectance (% $R_{o,max}$) lower than 0.5% [7–9]. The depths of the coal beds range from essentially zero at the subcrop to over 1000 m in most parts of the basin. The 8 coal samples from Turpan-Hami Basin and Junggar Basin are collected to characterize the low-rank coal porosities together with the four from Hailar Basin that will be further examined in the free/soluble gas experiment. In addition, these eight coals provide evaluation foundation for free/soluble gas proportion of the two basins.

$R_{o,max}$ (maximum vitrinite reflectance) measurements of all 12 coal samples were performed on polished surfaces of the coal samples using an Axio Imager M1mLeitz MPV-3 photometer microscope, following the conventional method according to China National Standard GB/6948-1998. It can be seen in Table 1 that the maximum vitrinite reflectance values ($R_{o,max}$) lies between 0.26% and 0.59%.

2.2. Measurement of coal porosity and its pore volume change

Porosity of coal (ϕ) can be calculated by means of true density and apparent density of coal, giving

$$\phi = \frac{\rho_{ct} - \rho_{ca}}{\rho_{ct}} \times 100\%, \tag{1}$$

where ρ_{ct} is the true density which can be calculated by applying Archimedes principle to the measurement with the pycnometry method; and ρ_{ca} is the apparent density that can be determined using the waxing method.

In order to estimate the porosity of *in situ* low-rank coal, porosity tests were further carried out with four lignite coal samples (HLR-02, HLR-05, HLR-06 and HLR-07) in coal tri-axial mechanics experiment. Coal sample cylinders with a diameter of 50 mm and a pillar height of 70–100 mm were dried in a 50 °C oven under vacuum for more than a week to remove the pre-existing moisture. Then the samples were soaked in coalseam water for 48 h to ensure the samples were uniformly saturated with water. The samples were then kept in a sealed chamber to control the water vapor partial pressure within the air at a relative humidity of 97%. Finally, the samples' moisture contents were allowed to equilibrate with this relative humidity over a period of about 50 days. This process ensured moisture in the coal samples reached equilibrium. Rock mechanics experiment system adopting an electro-hydraulic servo (see Fig. 2a) was utilized to test the porosity changes under confining pressure. Considering the low strength of the coal cylinder, experiments had to be performed by adopting

Table 1
Properties of coal samples used in experiments.

Sampling site	No.	$R_{o,max}$ (%)	ρ_{ct} (%)	ρ_{ca} (%)	φ (%)
Hailar Basin	HLR-02	0.26	1.57	1.22	22.29
	HLR-05	0.42	1.39	1.04	25.18
	HLR-06	0.42	1.41	0.96	31.91
	HLR-07	0.42	1.61	1.01	37.21
Turpan-Hami Basin	TH-01	0.50	1.31	1.28	2.29
	TH-02	0.53	1.43	1.39	2.80
	TH-03	0.54	1.34	1.30	2.99
	TH-04	0.54	1.42	1.36	4.23
	TH-05	0.55	1.36	1.30	4.41
	TH-06	0.56	1.35	1.28	5.19
Jung-gar Basin	ZGR-01	0.38	2.14	1.93	9.81
	ZGR-05	0.59	1.27	1.26	0.79

Note: $R_{o,max}$ is maximum reflectance of vitrinite.