www.czasopisma.pan.pl PAN www.journals.pan.pl
POLSKA AKADEMIA NAUK

DE GRUYTER OPEN

Arch. Min. Sci., Vol. 60 (2015), No 4, p. 1013–1028

Electronic version (in color) of this paper is available: http://mining.archives.pl

DOI 10.1515/amsc-2015-0067

LEI ZHANG*[1], NAJ AZIZ**, TING REN**, JAN NEMCIK**, SHIHAO TU*[1]

NITROGEN INJECTION TO FLUSH COAL SEAM GAS OUT OF COAL: AN EXPERIMENTAL STUDY

WPROWADZANIE AZOTU DO ZŁÓŻ WĘGLA W CELU WYPŁUKIWANIA GAZÓW Z POKŁADU – BADANIA EKSPERYMENTALNE

Several mines operating in the Bulli seam of the Sydney Basin in NSW, Australia are experiencing difficulties in reducing gas content within the available drainage lead time in various sections of the coal deposit. Increased density of drainage boreholes has proven to be ineffective, particularly in sections of the coal seam rich in CO_2. Plus with the increasing worldwide concern on green house gas reduction and clean energy utilisation, significant attention is paid to develop a more practical and economical method of enhancing the gas recovery from coal seams. A technology based on N_2 injection was proposed to flush the Coal Seam Gas (CSG) out of coal and enhance the gas drainage process. In this study, laboratory tests on CO_2 and CH_4 gas recovery from coal by N_2 injection are described and results show that N_2 flushing has a significant impact on the CO_2 and CH_4 desorption and removal from coal. During the flushing stage, it was found that N_2 flushing plays a more effective role in reducing adsorbed CH_4 than CO_2. Comparatively, during the desorption stage, the study shows gas desorption after N_2 flushing plays a more effective role in reducing adsorbed CO_2 than CH_4.

Keywords: N_2 injection, coal seam gas, coal, gas composition, gas volume

W kilku kopalniach eksploatujących złoże Bulli w zagłębiu węglowym Sydney w Nowej Południowej Walii w Australii pojawił się problem redukcji zawartości gazu kopalnianego w złożach zawartego w różnych częściach złoża, w określonym czasie. Zwiększenie gęstości wykonywania odwiertów drenażowych okazało się być metodą nieskuteczną, zwłaszcza w częściach złoża bogatego w CO_2. Inne kwestie to wzrastająca w świecie świadomość konieczności redukcji gazów cieplarnianych i wykorzystania czystej energii, stąd też podejmowane wysiłki na rzecz opracowania praktycznych i ekonomicznych metod odzyskiwania gazu ze złóż węgla. W pracy przedstawiono technologię opartą na wprowadzaniu azotu do złoża w celu wypłukania gazu zawartego w węglu, poprawiając skuteczność ich odzyskiwania. W prowadzonych pracach badano skuteczność odzysku CO_2 i metanu ze złoża węgla po wprowadzeniu

* SCHOOL OF MINES, CHINA UNIVERSITY OF MINING AND TECHNOLOGY, XUZHOU CITY, JIANGSU 221116, CHINA. KEY LABORATORY OF DEEP COAL RESOURCE MINING, MINISTRY OF EDUCATION OF CHINA, XUZHOU 221116, CHINA.

** SCHOOL OF CIVIL, MINING & ENVIRONMENTAL ENGINEERING, FACULTY OF ENGINEERING AND INFORMATION SCIENCES, UNIVERSITY OF WOLLONGONG, NSW 2522, AUSTRALIA.

[1] CORRESPONDING AUTHORS, E-mail: lz811@uowmail.edu.au; papertsh@126.com

do niego azotu. Wyniki badań wskazują, że wypłukiwanie azotem w poważnym stopniu wpływa na proces desorpcji CO_2 i CH_4 i ich usuwania z węgla. Na etapie wprowadzania azotu, stwierdzono że wypłukiwanie azotem w większym stopniu wspomaga usuwanie adsorbowanego CH_4 niż CO_2. Dla porównania, w trakcie desorpcji, wykazano, że desorpcja gazów po wprowadzeniu do złoża azotu znacznie skuteczniej redukuje ilość adsorbowanego CO_2 niż CH_4.

Słowa kluczowe: wprowadzanie azotu do złoża, gaz zawarty w węglu, węgiel, skład gazu, objętość gazu

1. Introduction

There is growing interest in gas injection to enhance Coal Seam Gas (CSG) recovery. The utilisation of N_2 injection has been found to help CSG recovery (Reeves & Oudinot, 2004, 2005; Florentin et al., 2010; Kiyama et al., 2011; Packham et al., 2012; Zhang, 2013). In the reservoir and economic analysis study of Tiffany unit N_2 – ECBM pilot (Reeves & Oudinot, 2005), it was found that incremental methane recovery of approximately 10-20% of the original gas in place was achieved with N_2 injection. The future N_2 injection was forecast to add another 25-40% to the total recovery of original gas in place. The future N_2 injection at Tiffany was also forecast to be economic.

Packham et al. (2012) reported the results from a field trial conducted with Surface to In-Seam (SIS) pre-drainage wells and concluded that the enhanced drainage could provide the means for both accelerating methane drainage and reducing residual gas content. Packham et al. (2011) provided the background to this field trial including details of the reservoir characteristics, well geometry and installations. They also described how history matching of the reservoir and simulation of the effects of nitrogen injection indicated that accelerated drainage was likely.

In concept, the principle of N_2 injection to Enhance Coalbed Methane recovery (N_2-ECBM) can be described as follows: N_2 is injected into a coal reservoir, it displaces the gaseous CSG from the cleat system, decreasing the CSG partial pressure and creating a compositional disequilibrium between the gaseous and adsorbed phases. These combined influences cause the CO_2 or CH_4 to desorb and diffuse into the cleat system, becoming the "stripped gas" from the matrix. The CSG then migrates to and is produced from production wells (Reeves & Oudinot, 2004).

Gas injection into coal seams can also cause physical changes to coal and hence coal permeability changes. Kiyama et al. (2011) found that the core coal permeability decreases after supercritical CO_2 injection, showing that adsorption-induced swelling has a significant impact on coal permeability. Subsequent N_2 flooding tests following CO_2 injection showed slow strain recovery, suggesting that N_2 displaces the adsorbed CO_2 in the coal matrix and the permeability of the coal core also recovered to a certain degree after N_2 injection. All these indicates that N_2 gas injection can be used to enhance the gas drainage of CSG and gas injection will cause a significant impact on coal behaviour and further influence the gas transport in coal when carrying out gas drainage.

This research program is a systematic project to investigate the comprehensive gas flow characteristics and hard-to-drain problem in the coal seam, the studies include the coal sorption capacity in terms of the temperature and moisture influences (Zhang et al., 2014b), coal particle size influence (Zhang et al., 2014a), coal sorption theory (Zhang et al., 2014c) and permeability influence (Zhang et al., 2014d) have already been carried out and the relevant results have been published. An experiment was conducted to further understand the mechanism of N_2 gas flushing to enhance the recovery of CSG, such as CO_2 and CH_4. The relationships between flushing time and N_2 as well as CO_2 and CH_4 concentration, N_2 charging volume and CO_2 and CH_4 recovery volume and flushing process were analysed in this experimental study. The term of gas composition means gas concentration or gas percentage of the gas mixture in this paper and the strain behaviour of coal is not discussed in this study, as it is a topic which is beyond the scope of this paper.

2. Geological background

The experimental study of N_2 injection to flush CSG was carried out on coal samples obtained from a typical hard-to-drain area (MG 22, 8-11 c/t) of Metropolitan Colliery in NSW, Australia. The current operating seam is the Bulli seam, which is stratigraphically the uppermost seam in the Illawarra coal measures, of the Sydney Basin, which belong to Permian and Triassic era (Faiz et al., 2007; Aziz et al., 2013). The Bulli coal is high quality coking coal with volatile matters ranging from 18-23% (air dried), the ash level varies between 8-10% (air dried), and the sulphur content is low at around 0.3%, the mineral content averages around 4% (Aziz et al., 2013). The vitrinite content is moderate at 45%, the inertinite content is about 50% and vitrinite reflectance at around 1.3 (Saghafi & Roberts, 2008; Aziz et al., 2013). The permeability of the coal varies between 0.5 to 6.0 mD, determined both *in situ* and in the laboratory (Lingard et al., 1982; Sereshki, 2005; Black, 2012; Zhang, 2013).

In situ gas contents of coal in the Illawarra Coal Measures range from less than 1 to 20 m^3/t with the highest contents occurring at depths between 600 and 800 m. The desorbed gas often comprises CH_4, CO_2, N_2, C_2H_6 and other higher hydrocarbons (Faiz et al., 2007). The two most abundant gases are CO_2 and CH_4, accounting for greater than 90% of the total gas in most areas of the Sydney Basin. Thermal history modelling indicates that most of the hydrocarbon gases were generated as a result of coalification during the Jurassic and Early Cretaceous (Faiz et al., 2003); additional CH_4 was apparently generated from post-Cretaceous microbial activity (Smith & Pallasser, 1996; Faiz et al., 2003).

Faiz et al. (2007) stated that isotope carbon-13 ($\delta^{13}C$) values for CO_2 from coal seams of the Illawarra Coal Measures vary between –25 and +15‰ (IAEA international standard defining Vienna Peedee Belemnite, VPDB), indicating various sources. These sources include thermogenic gas from coal/microbial oxidation of hydrocarbons ($\delta^{13}C$ –25 ±5‰), magmatic activity ($\delta^{13}C$ –7 ±3‰) and residual CO_2 after microbial reduction of CO_2 to CH_4 (0 to +15‰). Most of the $\delta^{13}C$ values ranging between –5 and –10‰, suggesting mainly magmatic sources, which was probably associated with the main episodes of igneous activity in the Permian, Jurassic and Tertiary (Faiz et al., 2007).

The variations of CO_2 and CH_4 are mainly related to the geological structure and depth. The variations in the gas composition have no clear relationship with coal composition or rank but show well-defined relationships with geological structure and stratigraphy. High proportions of CH_4 occur in the synclinal structures, whereas the CO_2 content increases towards structural highs. Extensive areas of pure CO_2 gas occur on anticlines and domes. In structural lows, high CO_2 concentrations are found near some dykes and related faults (Faiz & Hutton, 1995). This feature appears to also exist within the typical hard-to-drain area of this study.

Many Australian underground coal mines are mining in areas that require the use of gas drainage to reduce coal seam gas content to below a prescribed Threshold Limit Value (TLV). The TLV represents the maximum allowable gas content, relative to gas composition, considered safe for mine operations (Black, 2012). Mine operators are required to ensure seam gas content has been reduced below the applicable TLV prior to mining. In a number of cases, these mines encounter areas where the gas is hard to drain from the coal, ahead of mining (Black, 2012). Fig. 1 shows the gas content and composition analysis of the coal within the typical hard-to-drain area (MG 22, 8-11 c/t) of Metropolitan Colliery with 94 sample test results. The scatter of typical hard-to-drain area is concentrated almost entirely in the CO_2 rich area. Among the 94 samples, 63 samples are "Fail" samples, accounting for 67.0%, which directly indicates the area is a typically hard-to-drain area. The average values of CO_2 in both "Pass" and "fail" samples are 87.6% and 84.5% respectively.

Different factors including low permeability, high CO_2 concentration and geological variations have caused the hard-to-drain problems in certain parts of Bulli seam. Results from N_2 injection tests may provide invaluable knowledge for field trials of this innovative technology that could potentially lead to much enhanced gas recovery from hard-to-drain or low permeability seams.

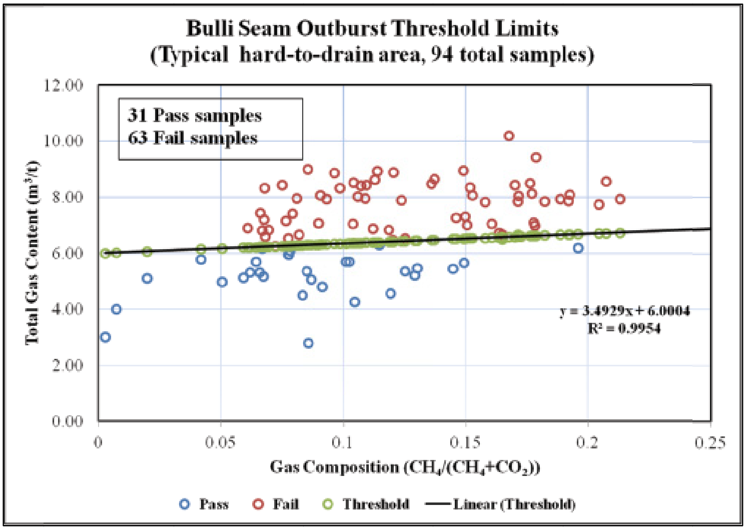


Fig. 1. Bulli seam outburst threshold limits (Typical hard-to-drain area)

3. Methods and experimental procedures

3.1. Testing apparatus and coal samples

The combined set up of a Multi Function Outburst Research Rig (MFORR) and Gas Chromatograph (GC) used in this test is shown in Fig. 2. The MFORR has various key components. These include the main apparatus support frame and a precision drill, a high pressure chamber which contains a load cell for measuring the load applied to the samples of coal, a pressure transducer for measuring the pressure inside the chamber, several flow meters set in series for measuring the gas flow rate, two strain gauges for measuring volumetric changes of the coal sample vertically and horizontally, a universal socket for loading a sample of coal vertically into the gas pressure chamber, a data acquisition system and a GC for the analysis of the gases discharged from the chamber. A four column GC is used to test gas for CO_2, CH_4 and N_2, which is discharged from the gas chamber in the experiment.

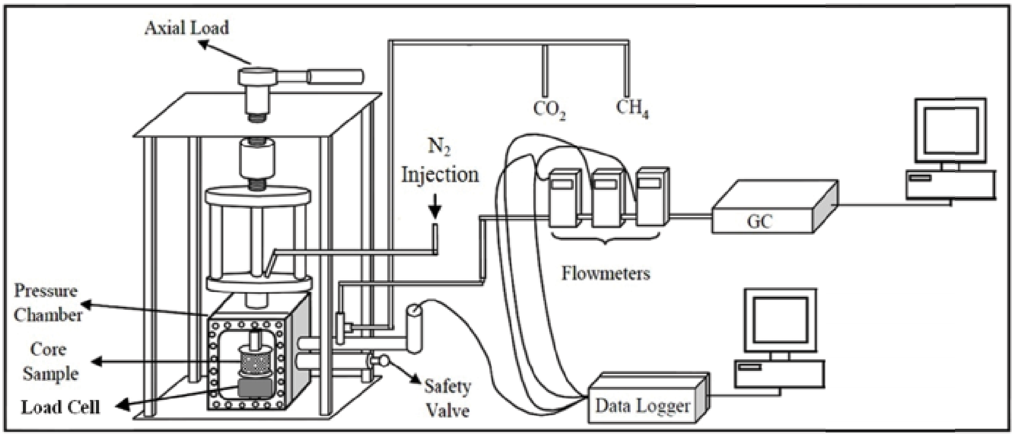


Fig. 2. A combination set up of MFORR and GC (modified from Florentin et al., 2010)

The sample for the flushing test was collected from the prescribed hard-to-drain area of the Bulli seam. The standard core samples were prepared with dimension of 54 mm in diameter and 50 mm in height. A 2 mm diameter hole was drilled in the middle of the cored coal. Prior to testing, two strain gauges were glued horizontally and vertically to the sample and both ends of the prepared specimen were sealed with a rubber layer. Fig. 3 shows a snapshot of the sample.

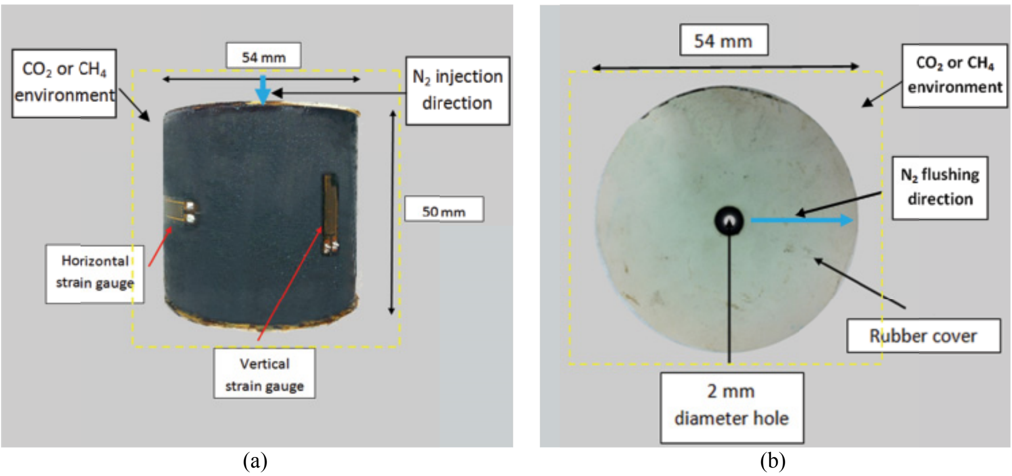


Fig. 3. Coal samples for N_2 flushing test

3.2. Experimental procedures

3.2.1. Stage 1 – Coal sorption process

In stage 1, the gas chamber was sealed with the prepared coal sample inside, before the gas sorption process, the system was vacuumed to –100 kPa (relative pressure) to remove the air inside the chamber and degas the coal samples. The whole system was maintained in a non-leakage condition operated properly by valves through the entire test. During all the three stages of the experiments, the laboratory temperature was kept at 25°C. The coal sample was then loaded axially to 3 MPa (equal to the axial load of 730 kg) initially and then the chamber was injected with CO_2 or CH_4 to 3 MPa. CSG gas was injected to allow the gas to diffuse and adsorbed in coal, until the coal reached gas sorption equilibrium at around 2 MPa pressure.

As the MFORR apparatus could not test the sorption capacity of coal, the sorption capacities with CO_2 and CH_4 were estimated through independent coal isotherm testing. The gravimetric method with only a sample cell, also referred to as the indirect gravimetric method, was first reported by Lama and Bartosiewicz (1982), and later by Aziz and Li (1999) and Sereshki (2005). Actually, coal sorption isotherm apparatus in the University of Wollongong is the combination of the gravimetric and volumetric methods, it utilises the gravimetric principle to calculate the total gas amount in the bomb and the volumetric principle to calculate the gas amount in the void space.

3.2.2. Stage 2 – N_2 injection to flush CO_2 and CH_4 process

Prior to the commencement of the N_2 injection test, the GC was calibrated to allow accurate measuring of the gas composition of CO_2, CH_4 and N_2 from the low to high range. N_2 gas flushing was carried out separately after the coal sample was saturated with CO_2 or CH_4 at the prescribed 2 MPa. The gas inside the chamber was tested by the GC to make sure that gas composition of either CO_2 or CH_4 was pure (99.9%), and the whole system was not contaminated by air.

At 2 MPa pressure, N_2 gas was then introduced to the gas chamber, charged through the central hole of the coal sample to allow N_2 gas to penetrate and permeate the coal sample along the radius and flow into the chamber. The directions of N_2 gas injection and flushing through the coal is indicated by blue arrow as shown in the sample in Fig. 3. The released gas was systematically discharged from the side hole of the chamber at 6 min intervals, going through a measuring system and a line of gas flowmeters (0-2 L/min and 0-15 L/min measurement range). The gas was collected in a 1 L capacity sample bag, which was directly connected to the GC to test gas composition.

3.2.3. Stage 3 – Desorption process after N_2 injection

In stage 3, desorption test was carried out, following the N_2 injection test, when the CO_2 or CH_4 gas composition was around 3%. The N_2 injection valve was closed. Gas pressure inside the chamber began to gradually drop as the remaining gas volume in the chamber was gradually removed. The released gas was collected in a 1 L storage capacity sample bag and analysed in the GC to test gas composition. The desorption process was suspended when the chamber pressure dropped to atmosphere pressure level. It should be noted that CO_2 and CH_4 flushing tests were carried out separately, but the experimental procedures were kept the same for comparison purposes.

4. Results and discussions

4.1. Stage 1 – Coal sorption process

Stage 1 is basically a coal sorption process, and prior to the N_2 flushing test. The coal samples were initially saturated with CO_2 and CH_4 at 2 MPa. This sorption tests were carried out uniquely by an indirect gravimetric method of determining the gas content of gas in coal. Four hard-to-drain coal samples were tested for the sorption capacity. Fig. 4 shows the comparative results of the adsorption isotherms for both CO_2 and CH_4 gas.

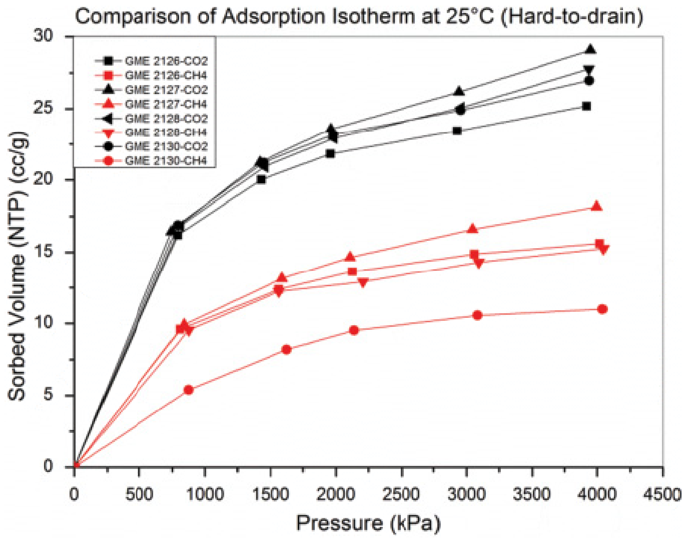


Fig. 4. Coal sorption isotherms of hard-to-drain coal samples

The Langmuir equation shown in Equation 1 was used to model the gas adsorption testing results. Langmuir parameters were calculated for each isotherm and shown in Table 1.

$$n_a = \frac{V_L P}{P + P_L} \tag{1}$$

where n_a is adsorbed gas content (gas volume per unit mass of coal), P is gas pressure, and V_L and P_L are experimental coefficients. The coefficient V_L represents the maximum gas storage capacity of the coal and is termed the 'Langmuir volume'. The coefficient P_L is the 'Langmuir pressure' and represents the gas pressure at which coal adsorbs a volume of gas equal to half of its maximum capacity (Harpalani et al., 2006).

TABLE 1

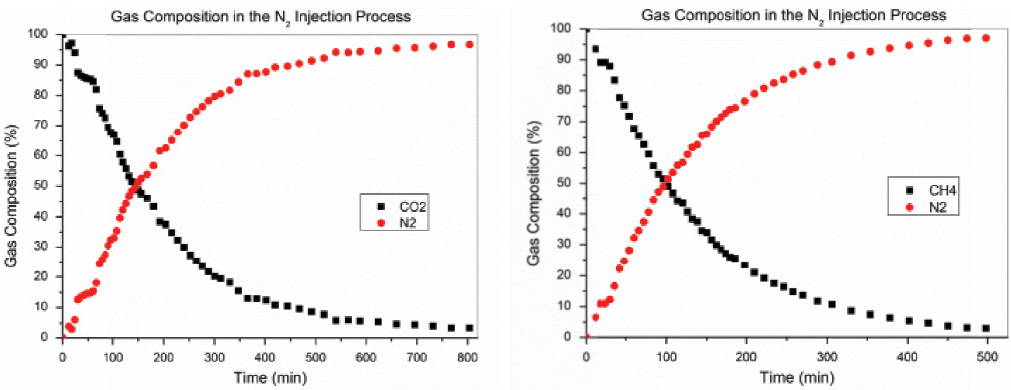
Langmuir parameters for the tested samples in terms of CO_2 and CH_4 (hard-to-drain area)

Langmuir parameters	**GME 2126**	**GME 2127**	**GME 2128**	**GME 2130**
Drainage area	**Hard-to-drain**	**Hard-to-drain**	**Hard-to-drain**	**Hard-to-drain**
Langmuir volume for CO_2 (cc/g)	29.2	35.2	33.1	31.4
Langmuir pressure for CO_2 (kPa)	653.4	992.1	845.0	704.4
Langmuir volume for CH_4 (cc/g)	18.6	23.4	18.2	15.3
Langmuir pressure for CH_4 (kPa)	774.4	1213.5	812.8	1457.5

4.2. Stage 2 – N_2 injection to flush CO_2 and CH_4 process

At 2 MPa pressure, N_2 gas was injected through the central hole of the coal sample to allow N_2 gas to penetrate and permeate the coal sample along the radius and flow into the chamber. The gas composition change inside the chamber was continuously monitored and the chamber pressure was maintained constant at 2 MPa during the whole N_2 injection process.

As shown in Fig. 5, during the N_2 flushing process, the CO_2 and CH_4 binary gas composition in the chamber gradually decreased and N_2 percentage increased, which indicates that CSG continues to be flushed out by N_2. The whole flushing test takes more than 13 h (800 min) for CO_2 shown in Fig. 5 (a) and 8 h (500 min) for CH_4, shown in Fig. 5 (b). At the lower CSG concentration stage, it appears that the flushing process is becoming harder as coal continues to desorb relatively higher CO_2 and CH_4 gas and the injected N_2 gas assists this adsorbed gas to desorb into the chamber. This phenomenon is especially apparent for the CO_2 flushing test, as coal still sorbed more CO_2 at low gas pressure, compared with CH_4. This finding generally agreed with the study of Florentin et al., (2010), who carried out similar test and found CSG can be flushed out with N_2 injection in the experimental test.

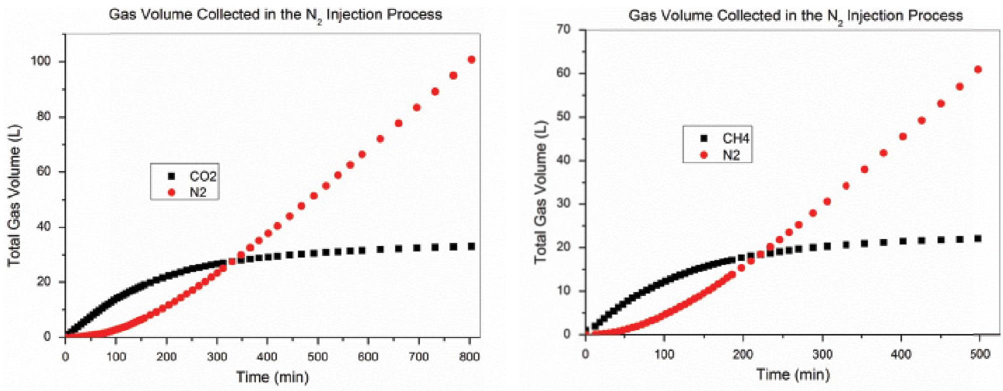


(a) (b)

Fig. 5. Gas composition during N_2 injection

As each step of the test, gas was discharged through the sample bag of 1L capacity, hence the volume of discharged gas in the chamber can be calculated Fig. 6 shows the volume of the various gases being discharged out of the pressure chamber over the whole test period.

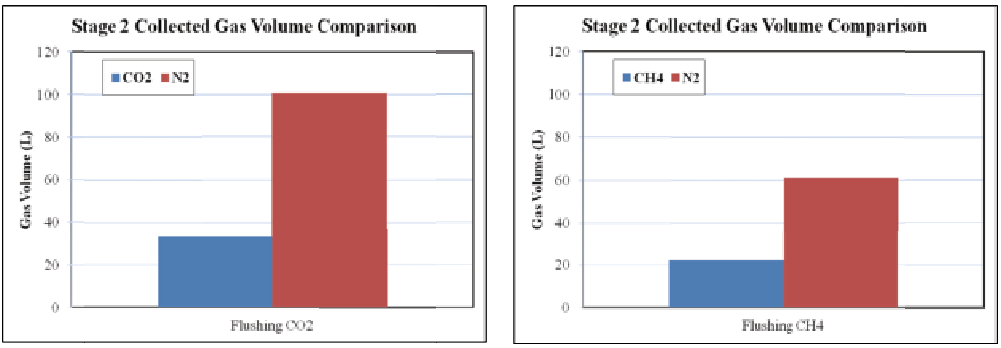
With the volume of N_2 gas injected into the chamber increasing, the total volume of CO_2 and CH_4 flushed out of system was accumulating. In the end, the total gases consumed during the flushing stage was estimated to be 100.9 L of N_2, liberating 33.1 L of CO_2 out of the system (Fig. 6 (a)). While, it was estimated that 61.0 L of N_2 were consumed in the flushing test, liberating 22.0 L of CH_4 (Fig. 6 (b)). Test results indicate that a greater volume of N_2 gas is needed to flush CO_2 than CH_4 gas out of coal, especially during the later stage of flushing. The total gas volume here includes both free gas in the chamber and adsorbed gas by the coal.



(a) (b)

Fig. 6. Gas volume during N_2 injection

Fig. 7 shows a comparison of collected gas volume in the flushing stage for CO_2 and CH_4. It can be observed that more N_2 is consumed than the recovered CO_2 or CH_4. The ratio of collected volume of N_2:CO_2 is around 3.05 and the ratio of collected volume of N_2:CH_4 is around 2.77. It indicates that more N_2 is needed to flush the same amount of CO_2 than CH_4.



(a) (b)

Fig. 7. Comparison of collected gas volume in Stage 2

According to the tested coal sorption isotherm of this typical hard-to-drain coal in stage 1, the average values of Langmuir parameters are followed, V_L = 32.2 cc/g, P_L = 798.5 kPa for CO_2 and V_L = 18.9 cc/g, P_L = 1064.55 kPa for CH_4. Thus, by combining all the parameters and using the Langmuir equation, when coal is saturated at 2 MPa, the adsorbed gas content is 23.01 cc/g for CO_2 and 12.33 cc/g for CH_4. It should be noted that this calculation is based on the assumption that the coal sample was fully saturated. For the flushed 160 g of coal sample, the adsorbed volume of CO_2 was 3.68 *L* and 1.97 *L* for CH_4. It is believed that all the adsorbed gas is flushed out during Stage 2 and Stage 3. As the gas composition of CO_2 or CH_4 was very low at the end of the flushing stage, all the gas coming out in the next desorption stage (Stage 3) is assumed to be adsorbed gas, which is 2.3 *L* for CO_2 and 1.1 *L* for CH_4. Hence, the total adsorbed gas volume flushed in Stage 2 is 1.38 *L* for CO_2 and 0.87 *L* for CH_4.

Based on the experimental data the following equation is adopted to calculate the gas content in coal during the flushing stage:

$$v_t = v_0 - \sum_{i=1}^{t} \left(c_{t-1} - c_t\right) \times \frac{\Delta v}{\Delta c} \tag{2}$$

where:

v_t is the gas content in coal during the flushing stage;
v_0 is the gas content in coal at the time 0 (starting point of flushing stage);
c_t and c_{t-1} are the gas composition in the chamber at the time t and $t-1$ during the flushing stage;
Δv is the total gas content drop in coal in the flushing stage;
Δc is the total gas composition drop in the chamber in the flushing stage, all the gas referred here is CO_2 or CH_4.

This above proposed calculation model is based on that the value of gas content in coal changes simultaneously with the change of gas composition or gas partial pressure, and the changing relationship between them is linear.

Fig. 8 shows the gas content change in coal during the flushing stage based on the above calculation, in total 1.38 *L* adsorbed CO_2 and 0.87 *L* of adsorbed CH_4 are flushed out of coal, helping reduce coal gas content of CO_2 from 23.01 cc/g to 14.385 cc/g and from 12.33 cc/g to 6.89 cc/g for CH_4. The reduction of 8.625 cc/g CO_2 gas content accounts for 37.5% of the total adsorbed CO_2 gas content while the reduction of 5.44 cc/g accounts for 44.1% of the total adsorbed CH_4 gas content, which indicates N_2 flushing plays a more effective role in reducing adsorbed CH_4 than CO_2. Hence, it is obvious that longer flushing time is needed to flush out CO_2 than CH_4 at the same equilibrium pressure (2 MPa) level.

4.3. Stage 3 – Desorption test after N_2 injection

In stage 3, a desorption test was carried out following the N_2 injection test when the CO_2 or CH_4 gas composition was around 3%. The N_2 injection valve was closed. Gas pressure inside the chamber began to drop as the remaining gas volume in the chamber was gradually removed. Fig. 9 shows the pressure drop (relative pressure) linearly in the desorption process, Fig. 9 (a) for the CO_2 test and Fig. 9 (b) for the CH_4 test.

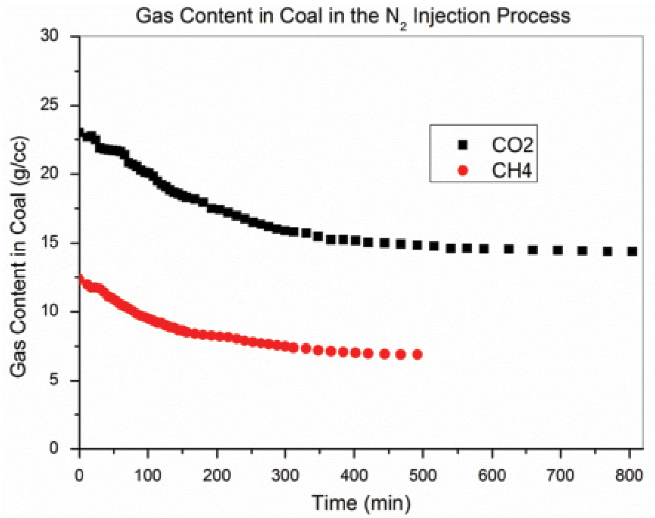
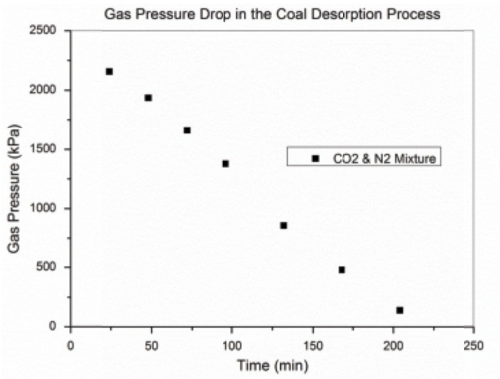
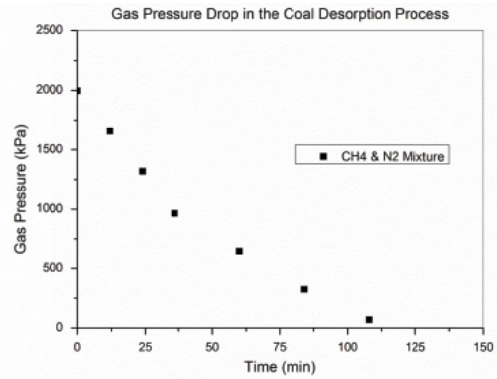


Fig. 8. Comparison of gas content in coal in Stage 2



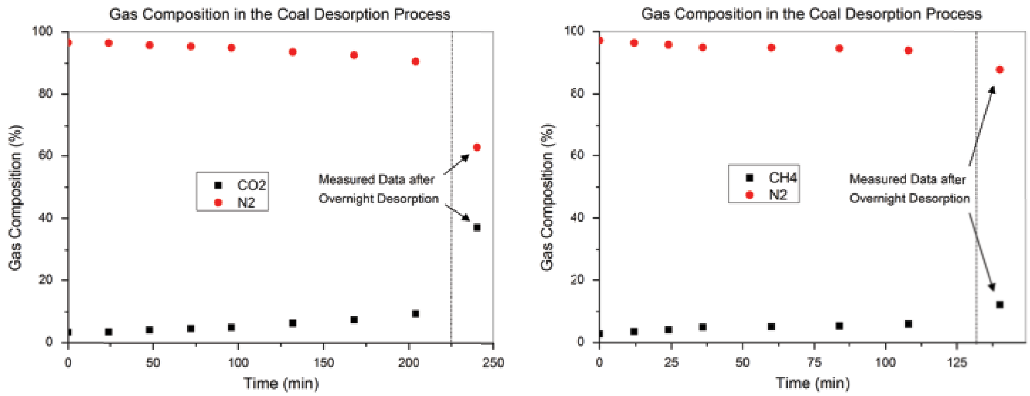
(a)



(b)

Fig. 9. Gas pressure drop during desorption

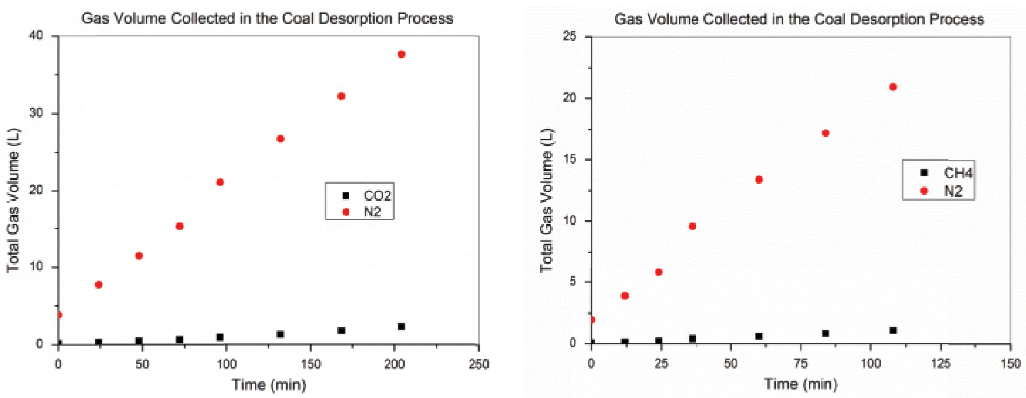
Fig. 10 shows the change of gas composition in the desorption process, the gas composition of CO_2 or CH_4 increases and at the same time the N_2 gas composition decreases. Specifically, in the CO_2 test, the CO_2 percentage starts to increase from 3.4% to 9.4% over a period of around 3 h (200 min) (Fig. 10 (a)), while in the CH_4 test, the CH_4 percentage starts to increase from 2.8% to 6.0% over a period of around 2 h (110 min) (Fig. 10 (b)). More CO_2 or CH_4 gas desorbs from the coal than N_2 in this process indicating greater sorption capacity of CO_2 or CH_4 than N_2. Further measured data after overnight desorption pointed out in Fig. 10 also confirm this conclusion, with CO_2 reaching 37.2% and CH_4 reaching 12.2%, N_2 decreasing to 62.8% and 87.8%, respectively. It should be noted that the pressure in the chamber was reduced to normal atmospheric level (101.320 kPa, absolute pressure).



(a) (b)

Fig. 10. Gas composition during desorption

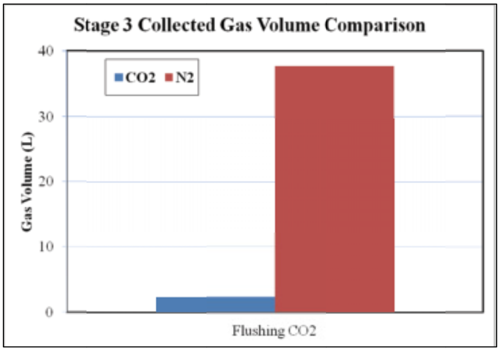
Fig. 11 shows the collected gas volume for each gas in the desorption process, as time proceeded, the total amount of gas volume for each gas increased. As there is a high composition of CSG (CO_2 or CH_4) in the chamber after the flushing test, much more N_2 is collected than CO_2 or CH_4. At the end of the CO_2 flushing test a total of 37.7 L of N_2 and 2.3 L of CO_2 were collected, while a total 20.9 L of N_2 and 1.1 L of CH_4 were collected in the CH_4 flushing test.



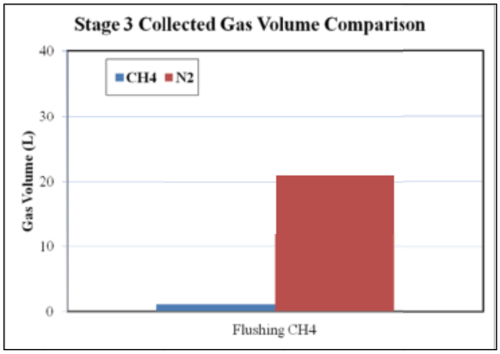
(a) (b)

Fig. 11. Gas volume during coal desorption

Fig. 12 shows the comparison of collected gas volume in the desorption stage for CO_2 and CH_4. It was found that more N_2 volume is collected than CO_2 or CH_4 was recovered. The ratio of collected volume of N_2:CO_2 is around 16.40 and the ratio of collected volume of N_2:CH_4 is around 19.0, which is relatively larger than the CO_2 flushing test.



(a)



(b)

Fig. 12. Comparison of collected gas volume in Stage 3

All the adsorbed gas is flushed out during the Stage 2 and Stage 3 and as the gas composition of CO_2 or CH_4 is very low at the end of the flushing stage, all the gas coming out in the stage 3 is assumed to be adsorbed gas, which is 2.30 L for CO_2 and 1.10 L for CH_4.

Based on the experimental data the following equation is adopted to calculate the gas content during the desorption stage:

$$v_t = v_0 - \sum_{i=1}^{t} \left(c_t - c_{t-1} \right) \times \frac{\Delta v}{\Delta c} \tag{3}$$

where:

v_t is the gas content in coal during the desorption stage;

v_0 is the gas content in coal at the time 0 (starting point of desorption stage);

c_t and c_{t-1} are the gas composition in the chamber at the time t and $t-1$ during the desorption stage;

Δv is the total gas content drop in coal in the desorption stage;

Δc is the total gas composition increase in the chamber in the desorption stage, all the gas referred here is CO_2 or CH_4.

This calculation model is also proposed based on the principles claimed in the Stage 2.

Packham et al. (2012) reported the continued injection of nitrogen would create conditions where the methane content of the coal could be reduced to negligible levels. Fig. 13 shows the gas content change in coal during the desorption stage. A total of 2.30 L of adsorbed CO_2 and 1.10 L of adsorbed CH_4 are desorbed from coal, to help reduce the remaining coal gas content, which is 14.385 cc/g for CO_2 and 6.89 cc/g for CH_4.The reduction accounts for 62.5% of the total adsorbed CO_2 gas content and 55.8% of the total adsorbed CH_4 gas content, respectively. It indicates gas desorption with gas pressure drop after N_2 flushing plays a more effective role in reducing adsorbed CO_2 than CH_4.

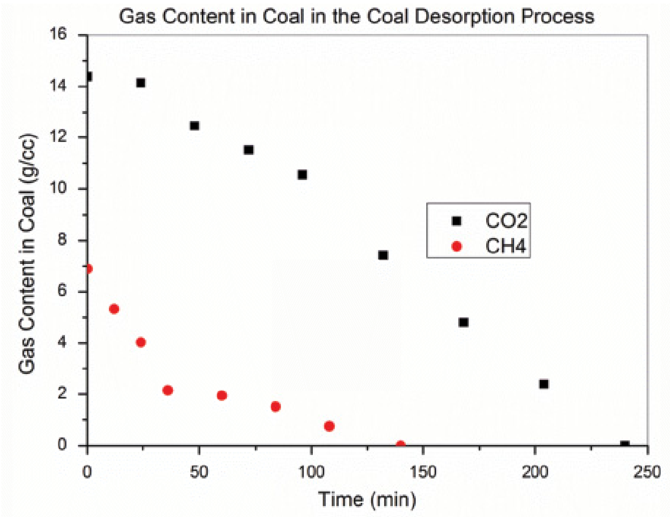


Fig. 13. Comparison of gas content in coal in Stage 3

5. Conclusions

Laboratory N_2 injection tests show that CSG (CO_2 and CH_4) can be flushed out by N_2 injection. During the N_2 flushing process, the CO_2 and CH_4 percentage of the chamber gas gradually decreases and the N_2 percentage increases, and with the N_2 flushing test approaching, the collected total gas volume of both CSG and N_2 increases. It is found that at low CO_2 or CH_4 composition stage, it is hard to use N_2 to achieve effective flushing.

After the flushing test, a certain amount of CO_2 or CH_4 is still adsorbed inside the coal. In the desorption process, the CO_2 or CH_4 percentage change starts to increase, indicating more CO_2 and CH_4 gas desorbs from the coal than N_2.

In the N_2 injection stage, the ratio of N_2:CO_2 collected volume is around 3.05 and the ratio is around 2.77 for N_2:CH_4. In the gas desorption stage, the ratio of N_2:CO_2 collected volume is around 16.40 and the ratio is around 19.0 for N_2:CH_4. During the flushing stage, N_2 injection helps to reduce the adsorbed gas content. The reduction of 8.625 cc/g CO_2 gas content accounts for 37.5% of the total adsorbed CO_2 gas content while the reduction of 5.44 cc/g accounts for 44.1% of the total adsorbed CH_4 gas content, which indicates N_2 flushing plays a more effective role in reducing adsorbed CH_4 than CO_2.

Comparatively, during the desorption stage, a total of 2.30 *L* of adsorbed CO_2 and 1.10 *L* of adsorbed CH_4 are desorbed from coal. The reduction accounts for 62.5% of the total adsorbed CO_2 gas content and 55.8% of the total adsorbed CH_4 gas content, respectively. It indicates gas desorption after N_2 flushing plays a more effective role in reducing adsorbed CO_2 than CH_4.

The result clearly shows that N_2 gas flushing has a significant effect on the CO_2 and CH_4 desorption and removal from coal. Thus it is important to develop a nitrogen injection technique in field trials, to enhance gas recovery in tight (hard-to-drain) and low permeable seams in future.

Acknowledgment

This research is supported by the Fundamental Research Funds for the Central Universities (2015QNA42), the Scholarship from University of Wollongong and China Scholarship Council, the National Natural Science Foundation of China (Grant No. 51374200), and The Priority Academic Programme Development of Higher Education Institutions in Jiangsu Province (Grant No. SZBF2011-6-B35). The authors wish to thank the staff and management of BHP Billiton-Illawarra Coal for providing coal samples used in this study. Thanks are also due to the technical staff at the University of Wollongong especially Col Devenish for experiment assistance.

References

Aziz N., Li W.M., 1999. *The effect of sorbed gas on the strength of coal – an experimental study*. Geotechnical and Geological Engineering, 17(3), 387-402.

Aziz N., Ren T., Nemcik J., Zhang L., 2013. *Permeability and volumetric changes in coal under different test environment*. Acta Geodyn. Geomater., 10(2), 163-171.

Black D., 2012. *Factors affecting the drainage of gas from coal and methods to improve drainage effectiveness*. PhD thesis (University of Wollongong).

Faiz M., Hutton A.C., 1995. *Geological controls on the distribution of CH_4 and CO_2 in coal seams of the southern coalfield, NSW, Australia*. International Symposium-CUM-Workshop on Management and Control of High Gas Emissions and Outbursts in Underground Coal Mines, Wollongong, NSW, Australia, 375-383.

Faiz M., Saghafi A., Sherwood N., Wang I., 2007. *The influence of petrological properties and burial history on coal seam methane reservoir characterisation, Sydney basin, Australia*. International Journal of Coal Geology, 70(1-3), 193-208.

Faiz M.M., Saghafi A., Barclay S.A., Stalker L., Sherwood N. R., Whitford D.J., 2007. *Evaluating geological sequestration of CO_2 in bituminous coals: The southern Sydney basin, Australia as a natural analogue*. International Journal of Greenhouse Gas Control, 1(2), 223-235.

Faiz M.M., Stalker L., Sherwood N., Saghafi A., Wold M., Barclay S., Choudhury J., Barker W., Wang I., 2003. *Bioenhancement of coal bed methane resources in the southern Sydney basin*. J. Aust. Petrol. Prod. Explor. Assoc., 43, 595-610.

Florentin R., Aziz N., Black D., Nghiem L., Baris K., 2010. *Recovery of stored gas in coal by nitrogen injection – a laboratory study*. 10th Underground Coal Operators' Conference. (Eds: Naj, A. and Jan, N.), University of Wollongong, NSW, Australia: 223-234.

Harpalani S., Prusty B.K., Dutta P., 2006. *Methane/CO_2 Sorption Modeling for Coalbed Methane Production and CO_2 Sequestration*. Energy & Fuels, 20(4), 1591-1599.

Kiyama T., Nishimoto S., Fujioka M., Xue Z., Ishijima Y., Pan Z., Connell L.D., 2011. *Coal swelling strain and permeability change with injecting liquid/supercritical CO_2 and N_2 at stress-constrained conditions*. International Journal of Coal Geology, 85(1), 56-64.

Lama R.D., Bartosiewicz H., 1982. *Determination of gas content of coal seams*. Seam Gas Drainage with Particular Reference to the Working Seam. University of Wollongong, NSW, Australia, 36-52.

Lingard P.S., Phillips H.R., Doig I.D., 1982. *The permeability of some Australian coals*. Seam gas drainage with particular reference to the working seam, (Ed.: Hargraves, A.J.), University of Wollongong, Wollongong, Australia, 70-80.

Packham R., Cinar Y., Moreby R., 2011. *Simulation of an enhanced gas recovery field trial for coal mine gas management*. International Journal of Coal Geology, 85(3-4), 247-256.

Packham R., Connell L., Cinar Y., Moreby R., 2012. *Observations from an enhanced gas recovery field trial for coal mine gas management*. International Journal of Coal Geology, 100, 82-92.

Reeves S., Oudinot A., 2004. *The Tiffany unit N_2 – ECBM pilot: A reservoir modelling study*. Topical Report U.S. Department of Energy**:** DE-FC26-20NT40924.

Reeves S., Oudinot A., 2005. *The Tiffany unit N_2 – ECBM pilot – a reservoir and economic analysis*. 2005 International Coalbed Methane Symposium. Tuscaloosa, Alabama, USA: Paper 0523.

Saghafi A., Roberts D., 2008. *Measurement of CO_2 and CH_4 reservoir properties of coals from Westcliff mine*. CSIRO Investigation Report ET/IR 1033R.

Sereshki F., 2005. *Improving coal mine safety by identifying factors that influence the sudden release of gases in outburst prone zones*. PhD thesis (University of Wollongong).

Smith J.W., Pallasser R.J., 1996. *Microbial origin of Australian coalbed methane*. Am. Assoc. Petrol. Geologists Bull, 80, 807-891.

Zhang L., 2013. *Study of coal sorption characteristics and gas drainage in hard-to-drain coal seams*. PhD thesis (University of Wollongong).

Zhang L., Aziz N., Ren T., Nemcik J., Tu S., 2014a. *Influence of coal particle size on coal adsorption and desorption characteristics*. Archives of Mining Sciences, Vol. 59, No. 3, p. 807-820.

Zhang L., Ren T., Aziz N., 2014b. *Influences of temperature and moisture on coal sorption characteristics of a bituminous coal from the Sydney Basin, Australia*. International Journal of Oil, Gas and Coal Technology, Vol. 8, No. 1, p. 62-78.

Zhang L., Ren T., Aziz N., Tu S., 2014c. *Coal sorption characteristics and coal surface tension*. International Journal of Oil, Gas and Coal Technology, Vol. 8, No. 3, p. 336-352.

Zhang L., Ren T., Aziz N., Tu S., 2014d. *Triaxial permeability testing and microstructure study of hard-to-drain coal from Sydney Basin, Australia*. International Journal of Oil, Gas and Coal Technology, Vol. 8, No. 4, p. 432-448.

Received: 18 December 2013