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DOI
10.1021/acs.energyfuels.7b02402

Publication date
2017

Document Version
Accepted author manuscript

Published in
Energy & Fuels

Citation (APA)
Hadi Mosleh, M., Sedighi, M., Vardon, P. J., & Turner, M. (2017). Efficiency of Carbon Dioxide Storage and Enhanced Methane Recovery in a High Rank Coal. *Energy & Fuels*, *31*(12), 13892-13900. https://doi.org/10.1021/acs.energyfuels.7b02402

Efficiency of carbon dioxide storage and enhanced methane recovery in a high rank coal

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Abstract

High affinity and adsorption capacity of coal to carbon dioxide provides alternative approach for the enhanced recovery of methane from unminable coalfields (CO_2-ECBM) by which a potential solution for long-term CO_2 sequestration in deep geological formations can also be achieved. However, due to chemo-mechanical effects induced by the interactions between CO_2 and coal, the effective methane production and carbon dioxide storage can be degraded which has caused uncertainties about the techno-economic feasibility of the CO_2-ECBM process. This study presents an experimental investigation that aims to address key knowledge gaps related to the efficiency of CO_2 storage and CH_4 recovery in high rank coals for which comprehensive experimental data set and analysis are largely missing. Competitive displacements of CH_4 with N_2 or CO_2 in an anthracite coal sample from South Wales coalfield have been studies, based on a series of core flooding experiments.

The results show that the N_2 breakthrough time (the time at which 1% of the total gas injected was recovered) was almost spontaneous whereas a considerably delayed breakthrough time was observed for the case of CO_2-ECBM experiment. In addition it was observed that for the CO_2-ECBM experiment, the ratios of CH_4 recovery with respect to the total amount of gas injected and gas stored were higher by factors of 10 and 2.4, respectively. The results also show that 90% of the total N_2 injected was produced in the outflow gas, whereas for the case of the CO_2 experiment, only 63% of the total injected CO_2 was produced. Presence of high amount of N_2 in the outflow may lead to additional challenges in order to separate N_2 from CH_4 and thus affects the efficiency of the N_2-ECBM method. Under the conditions of the experiments, the total CH_4 displacement ratio and breakthrough for the case CO_2-ECBM were found to be more favorable compared to those obtained from N_2-ECBM. This study provides new insights into the efficiency of CO_2-ECBM process and offers a comprehensive experimental data set that can be used for testing the accuracy of predictive models.

Keywords: *CO_2 sequestration, enhanced coalbed methane recovery (ECBM), anthracite coal, core flooding, gas permeability, gas sorption, South Wales coalfield.*

1. Introduction

The process of coalbed methane (CBM) production from unminable coal fields usually involves the pressure depletion in the coal reservoir by pumping out the naturally existing formation water. However, the pressure depletion only allows a limited amount of in-place-gas (CH_4) to be produced because the methane is adsorbed on the coal even at low pressures (White et al., 2005). As a result, approximately 30 to 70% of the in-place-gas in coal cannot be recovered using the conventional pressure depletion method (Puri and Yee, 1990). Enhanced coalbed methane (ECBM) recovery is a process by which N_2 or CO_2 (or a mixture of both) is injected into the coal seam to enhance the recovery of coalbed methane (White et al., 2005). In N_2-ECBM, N_2 first displaces the free CH_4 from the seam and reduces the partial pressure of methane in the reservoir that enables further methane in the adsorbed phase (in coal matrix) to be released (Shi and Durucan, 2005). However, rapid breakthrough of N_2 during the production of methane has been reported to be the major issue in the field projects (Perera and Ranjith, 2015). Alternatively, CO_2 has been suggested to enhance the coalbed methane recovery (CO_2-ECBM) due to the higher affinity of coal to adsorb carbon dioxide compared to methane that may result in larger amount of coalbed methane production. In addition, the potential long term sequestration of CO_2 in deep unminable coal seams during the process of CO_2-ECBM is an advantage over the N_2-ECBM process (Shi and Durucan, 2005). Permeability evolution in coal as the result of changes in effective stress and sorption-induced swelling and shrinkage during ECBM process has been extensively studies by researchers, through laboratory investigation and numerical modeling, *e.g.* Feng et al. (2017), Hadi Mosleh et al, (2017), Liu and Harpalani (2013), Ma et al. (2011).

Experimental studies show that coal can exhibit shrinkage or swelling during interaction with different gas species, *e.g.* Mazumder et al. (2006), Mazzotti et al. (2009), Hadi Mosleh (2014). Therefore, the uptake or release of CO_2 and CH_4 is a combination of adsorption/desorption processes and coal swelling/shrinkage that can affect the permeability of the coal, resulting in the overall rate and efficiency of methane recovery and carbon dioxide storage.

Observations from limited pilot tests of carbon sequestration in coal seams that have been reported in the literature (Japan, Poland and China) indicate the reduction in the injectivity of carbon dioxide as the result of reduction in coal permeability induced by swelling (Reeves and Oudinot, 2005; Yamaguchi et al. 2006; van Bergen et al. 2006). However, the outcomes of the field tests reported have largely remained inconclusive (Pan and Connell, 2012). A limited number of lab-scale experimental studies have also been reported on processes related to enhanced coalbed methane recovery nitrogen and carbon dioxide, *e.g.* van Hemert et al. (2012), Wang et al. (2010), Mazumder and Wolf (2008), Yu et al. (2008). The experimental approach adopted in majority of these studies is based on core flooding test in which a core sample of coal is first saturated with CH_4 and then N_2 or CO_2 (or a mixture if gas) is injected into the sample. The composition of the outflow gas is monitored during the test to evaluate the breakthrough time of the injected gas, displacement of gases and the rate of gas storage/recovery. Wang et al. (2010) carried out a series of gas storage and recovery experiments on highly volatile bituminous coal and showed that for the example considered, the amount of the adsorbed CO_2 was two orders of magnitude larger as compared to the amount of desorbed CH_4. Yu et al. (2008) has reported gas storage and recovery experiments on coal samples originated from Qinshui basin in China. The results show that initially the CO_2 fraction in the outflow gas was very small compared to the CH_4 and the initial CH_4 displacement with CO_2 was not associated with the CO_2 release. They have also shown that with an increase of the volume of replaced CH_4, the discharge capacity of CO_2 has slowly increased.

Compared to the extensive experimental investigations carried out on adsorption/desorption characteristics and permeability properties of coal to gas species, the laboratory scale experimental studies on the process of N_2-ECBM and CO_2-ECBM are very limited and comparative assessment of the efficiency of enhanced methane recovery by N_2 and CO_2 is still lacking. In the last decade several conceptual models have been developed to account for the flow of gas in coal and fractured rock (*e.g.* Shi and Durucan, 2003; Salimzadeh and Khalili, 2015; Hosking, 2014). These models are usually based on mechanistic approaches that require appropriate constitutive relationships (*e.g.* gas permeability model) and experimental data for testing. Data sets generated from laboratory scale experiments on N_2-ECBM and CO_2-ECBM are critical bench marks for testing such numerical models that can be used for simulation and design of the process at field scale. However, the experimental studies that contain adequate material properties and provide constitutive relationships for numerical modeling are scarce, especially for high rank coals, *i.e.* anthracite. In this work, we aim to address i) gaps in knowledge related to the response of a high rank coal to gas injection, displacement and storage during ECBM process, and ii) the lack of adequate and comprehensive experimental dataset required for testing the predictive models. In this paper, an experimental investigation on the process of N_2-ECBM and CO_2-ECBM in a high rank coal from South Wales coalfield is presented and comparative assessment of the efficiency based on gas recovery and storage for both N_2-ECBM and CO_2-ECBM is discussed. Core flooding experiments have been conducted in which N_2 and CO_2 were injected into the CH_4-saturated coal sample to evaluate the competitive displacement of CH_4 with N_2 and CO_2 under simulated underground conditions. The displacement process, gas breakthrough, and recovery ratios are also discussed.

2. Material and methods

The anthracite coal sample used in this work was obtained from the Six Feet coal seam, at the Unity coal mine located in South Wales, UK (Hadi Mosleh et al., 2017). Blocks of coal with dimensions of approximately 0.5×0.5×0.5m were collected from the depth of approximately 550m. Prior to core flooding experiments, a series of coal characterisation tests including the Proximate and Ultimate Analyses (BS 1016-104 and BS 1016-106) were conducted on crushed samples in order to determine key properties of moisture content, ash content, and volatile matter as well as elemental composition such as sulphur and carbon contents. Table 1 presents a summary of the physical and chemical properties of the coal used.

2.1. Preparation of core samples

The core sample used was drilled from a large block of coal using a diamond core drill bit with 70mm internal diameter. The core sample was then cut into the required length using a diamond saw. Special care was taken during the coring and cutting processes to minimise breakage or damage to the coal structure. Any small breakage especially around the edges could potentially damage or puncture the rubber membrane during triaxial core flooding tests and under the high confining pressures and therefore had to be removed. In order to prevent breakage of the coal samples under high stress conditions, the ends of the specimens were ground and made parallel to each other using a fine sand paper. This allowed a uniform distribution of the axial stresses to both ends of the sample. In order to remove any residual moisture, the core sample was then air-dried for 24hr before it was placed in the triaxial cell for the tests.

2.2. Triaxial core flooding setup

Triaxial core flooding setup was designed and developed by Hadi Mosleh (2014). The experimental setup comprises i) a high pressure triaxial core flooding system, ii) a pressure control system, iii) a temperature control system, and iv) an ancillary system including gas supply and analysing units. A schematic diagram of the developed laboratory facility is presented in Figure 1 (Hadi Mosleh et al., 2017).

The core sample sits within a 1.5mm thick silicone rubber sleeve and the gas passes through a porous plate at the bottom of the sample, then it leaves the cell through a similar arrangement at the top after having passed through the test core. A Mass Flow Meter capable of measuring high flow rates up to $17\times10^{-6}m^3/s$ (1L/min) is connected to the outlet which is capable of working under both subcritical and supercritical conditions, with pressures up to 20MPa.

The pressure control system includes a pressure/volume controller to control the confining pressure and a high pressure regulator with a needle valve to control the gas pore pressure. Two 32MPa in-line pore pressure transducers were selected to measure the inlet and the outlet gas pressures. The confining system consists of a 32MPa pressure/volume controller with a $2\times10^{-4}m^3$ (200mL) oil reservoir. Volume changes can be measured and displayed to $1\times10^{-9}m^3$ (0.001mL). The confining pressure is provided by silicone oil 350 (Polydimethylsiloxane) and a hydraulic pump. The composition of the outflow gases was determined using an Emerson X-Stream general purpose gas analyser (standard 19"/3HU version) with the optimum gas flow rate of $1.7\times1^{-5}m^3/s$ (1L/min) and ±0.01% accuracy of Full Range Output (FRO). More details of the design and development of the laboratory setup can be found in Hadi Mosleh et al. (2017).

2.3. Experimental procedure

The core sample of 70mm diameter and 100mm length was carefully wrapped with a thick PTFE tape before being placed in the silicone rubber sleeve. The displacement transducers, two axial and one radial, and three thermocouples were then attached to the sample. The top cap was placed on the base pedestal and the cell was filled with the silicone oil. The temperature of the system was adjusted to 25°C (the corresponding temperature of the depth at which the sample was taken), using four heating elements attached to the cell's body and a programmable controller. The temperature was kept constant throughout the test. A confining pressure of 1MPa was applied and the sample was subjected to vacuum for 24 hours to remove the residual moisture and gases from the pore space. Prior to each core flooding test, the core sample was saturated with the chosen gas.

The steady-state method has been used to estimate permeability of the coal sample (Carles et al., 2007). For the initial permeability measurements, the confining pressure was maintained at the desired pressure and increased stepwise. Once the steady-state flow rate was achieved at each step, the differential gas pressures and gas flow rates were recorded. The permeability of the coal sample was calculated using Darcy's law (Carman, 1956), given as:

$$k_g = \frac{2Q_0\mu_g LP_0}{A(P_{up}^2 - P_{down}^2)} \tag{1}$$

where, k_g is the gas permeability coefficient (m^2), Q_0 is the volumetric rate of flow (m^3/s), μ_g is the viscosity of the gas (Pa.s), L is the sample length (m) and P_0 refers to the reference pressure (Pa) which in this study was atmospheric pressure, *i.e.* 1×10^{-5} Pa. A is the cross-sectional area of the sample (m^2), P_{up} is the upstream gas pressure (Pa), and P_{down} is the downstream gas pressure (Pa). The viscosity of gases (μ_g) was calculated as function of temperature, using the Sutherland formula (Smits and Dussauge, 2006).

The experimental tests carried out based on the steps summarised in Figure 2:

Stage 1: Flow characterisation of the coal sample

Since helium is a non-adsorptive/non-reactive gas it was used to characterise the coal sample for its intrinsic permeability and to investigate the mechanical effect of stresses on permeability of coal during gas injection and depletion processes. The intrinsic permeability to helium has been used as a key material property to compare gas flow behaviour of the coal sample used in this study with those from previous studies, *i.e.* Hadi Mosleh (2014), and also to evaluate the changes in permeability of the same coal sample with respect to gas species. The permeability of the coal sample to helium was estimated for a range of gas injection pressures (up to 5.5MPa) and at several confining pressures (up to 6MPa). In order to evaluate the effects of confining pressure on the gas flow properties of the coal sample, the confining pressure was first increased stepwise up to 6MPa while gas injection pressure was kept constant at 3.5MPa. To assess the effect of pore pressure changes on gas flow and permeability of the coal sample, gas pressure was increased gradually to 5.5MPa while the confining pressure was kept constant (6MPa). The coal sample was then subjected to vacuum for 24 hours and saturated with CH_4 at 5MPa injection pressure and 6MPa confining pressure. The permeability of the coal to CH_4 was measured by performing a CH_4 flooding experiment under a range of gas injection pressures from 3.5 to 5.5MPa.

Stage 2: N_2- and CO_2-ECBM experiments

The coal sample was re-saturated with CH_4 at 5MPa injection pressure and 6MPa confining pressure. N_2 gas was then injected into the CH_4-saturated at 5MPa injection pressure while the downstream valve was at atmospheric pressure, *i.e.* 0.1MPa. The composition of the outflow gas was analysed during the experiment using the gas analyser. After the N_2-ECBM experiment, the residual gas was removed from the sample by applying vacuum. The sample was purged with CH_4 While the composition of outflow gas was monitored using the gas analyser until no residual N_2 was present in the outflow. The sample was then re-saturated with CH_4, and CO_2 subsequently was injected at 5 MPa. The confining pressure was kept constant throughout at 6MPa. The composition of the outflow gas was monitored continuously and the test was continued until CH_4 was mostly displaced with CO_2. Figure 3 presents a schematic diagram of the experimental conditions applied for the N_2- and CO_2-ECBM experiments.

3. Results and discussions

3.1. Helium flooding experiment

For low permeability coals, the flow behaviour is highly dependent on the effective stress (Huy et al., 2010). For incompressible fluid such as water, the effective stress is defined as the difference between the confining pressure and linear mean gas pressure across the sample (Harpalani and Chen, 1997):

$$\sigma_{eff} = P_c - \frac{P_{up} + P_{down}}{2} \tag{2}$$

where, σ_{eff} is the effective stress and P_c is the confining pressure.

Since gas is compressible, its bulk density varies greatly which in turn has a significant effect on gas transport within the porous medium. Therefore unlike incompressible fluids, variation of gas pore pressure across sample length is not expected to be linear. In this study, the analytical solution presented by Wu et al. (1998) has been used to estimate the changes in gas pore pressure across the sample at steady-state flow conditions:

$$P(x) = -b + \sqrt{b^2 + P_L^2 + 2bP_L + 2q_m\mu(L-x)/k_\infty\beta} \tag{3}$$

where, $P(x)$ is the gas pressure (Pa) at linear distance x (m), b is the Klinkenberg coefficient, P_L is the gas pressure at outlet boundaries of linear flow systems (Pa), q_m is the gas mass injection or pumping flux (kg/s.m^2), L is the length of linear flow systems or thickness of unsaturated zone (m), k_∞ is the absolute permeability (m^2), and β is the compressibility factor; μ viscosity (Pa.s).

The length of the sample was divided into 7 sections of 0.02m long, and for each section the average pore pressure was estimated using Eq. (3). In general, the average pore pressure within the coal sample obtained from Eq. (3) was found to be approximately 26-28% higher than average pore pressure obtained from linear approximation method. The mean gas pore pressure obtained from Eq. (3) was then used to estimate the effective stress, using Eq. (2).

The variations of permeability of the coal sample to helium with effective stress are presented in Figure 4a. Based on the results, the permeability of the coal sample ranges between 0.15 and 0.45×10^{-15}m^2 over the range of applied pressures and confining stresses applied. Variations in confining pressure and gas pore pressure showed slightly different effects on permeability evolution of the coal sample, however the overall trends of permeability behaviour as a result of changes in confining pressure and gas pore pressure were similar, *i.e.* the coal permeability to helium was reduced with increase in effective stress. These results are consistent with the results of another series of core flooding experiments performed by Hadi Mosleh (2014) on a similar coal sample (sister sample) obtained from same block of coal (Figure 4b). Coal permeability variations with effective stress can be attributed to the expansion or closure of the internal fractures and microfractures (Vishal et al., 2013). Non-linear evolution of coal permeability to gases with effective stress has been reported by other researchers (*e.g.* Feng et al., 2016; Mitra et al., 2012).

3.2. CH_4 flooding experiment

The CH_4 core flooding experiment was performed to evaluate the initial permeability of the coal sample to CH_4 before introducing CO_2 during the gas storage and recovery experiments and its consequent swelling effect on gas flow properties of coal. The results of the coal permeability to CH_4 versus effective stress are presented in Figure 5. The coal permeability to CH_4 was found to vary between 0.03×10^{-15}m^2 and 0.14×10^{-15}m^2 under the applied stresses. In general, the permeability of the coal sample to CH_4 was found to be one order of magnitude lower than that to helium which can be mainly related to their differences in molecular size. The effect of gas sorption-induced permeability evolution of coal should also be taken into account when interpreting such results. The range of permeability evolution of coal reported in the literature as the result of CH_4 interaction with coal matrix varies greatly. For instance, Harpalani and Mitra (2010) have reported a CH_4 permeability reduction of approximately 25% compared to the original value, whereas in a study conducted by by Milewska-Duda et al. (2000), the effect of CH_4 on coal matrix swelling and permeability reduction was found to be negligible, compared to highly reactive gases such as CO_2.

Once the evaluation of the initial properties of the coal sample was completed, the coal sample was re-saturated with CH_4 at injection pressure of 5MPa and confining pressure of 6MPa.

3.3. N_2 - ECBM recovery

The results of the gas displacement process during the N_2-ECBM experiment are presented in Figure 6. The results present the variation of the gas composition in the outflow with time. It can be observed that an early breakthrough of the injected N_2 has occurred shortly after the injection process was started. The definition of the breakthrough time of the injected gas varies in the literature. A common definition for the breakthrough in work related to gas storage and recovery processes has been considered here which is described as the time at which 1% of the total gas injected is recovered (van Hemert et al., 2012; Mazumder and Wolf, 2008; Ross, 2007). In this case, the breakthrough time was less than 100s.

The breakthrough time reported in the literature might vary greatly ranging from several minutes to several days depending on the coal type, sample size, permeability, gas injection pressure, injection rate and effective stresses, *e.g.* Connell et al. (2011), Shi et al. (2008), Yu et al. (2008). The early breakthrough observed in the N_2-ECBM experiment and relatively fast displacements of CH4 are primarily related to the displacement of the free gas existing within the coal cleats/microfractures rather than the free gas in the coal matrix. This is related to the experimental conditions in which a relatively high injection pressure has been applied under low effective stress on the sample.

The results presented in Figure 6 show that within the first half an hour of the experiment, more than 95% of the production gas consisted of N_2. Rapid changes in N_2 and CH_4 fractions at this stage of the experiment can be attributed to the displacement of the free or weakly adsorbed CH_4 molecules with N_2. Therefore, the dominant gas exchange process at this stage was the differences in the partial pressures of N_2 and CH_4 within the coal cleats and microfractures. As the experiment continued, the rate of changes in the fraction of CH_4 in the production gas became very slow and remained steady and continuous. This can be related to the slow diffusion of N_2 and CH_4 in the coal matrix (Cui et al., 2004), which is the dominant gas exchange process at this stage of the experiment. As a result, the diffusion of N_2 into the coal matrix gradually reduces the partial pressure of CH_4 in the coal matrix and eventually leads to CH_4 desorption.

3.4. CO_2 - ECBM recovery

The experimental results of the gas displacement process during the CO_2-ECBM experiment are presented in Figure 7 which shows the variations of gas composition in the production gas with time as a result of CH_4 displacement with CO_2. The small spikes on the graph are related to the minor fluctuations in the injection pressure generated as a result of simultaneous operation of the gas booster.

From the results presented, it is apparent that the breakthrough time of CO_2 is two times slower than the N_2-ECBM experiment that can be attributed to a combination of gas diffusion process in the coal and the effects of coal matrix swelling induced by the CO_2 adsorption. Although, the volumetric deformation has not been directly measured in the experiments presented here, reduction in the mass flow rate of outflow gas during the CO_2-ECBM experiment demonstrate the significance of the effect of coal swelling on the flow process. The mass flow rates recorded at downstream of the sample reduced from 14g/h to 1g/h during the course of CO_2-ECBM experiment (more than 5 hours), whereas for the case of N_2-ECBM experiment, the recorded mass-flow rates remained relatively steady throughout the experiment, *i.e.* 28-34g/h.

With its relatively smaller kinetic diameter, CO_2 molecules can penetrate into the micro-pores of coal which are inaccessible or less accessible to CH_4 and N_2 molecules that have larger kinetic diameters. This can result in one or two orders of magnitude higher diffusivity of CO_2 in the coal matrix compared with N_2 and CH_4 (Cui et al., 2004). Hence, the higher diffusion of CO_2 than that for CH_4 and N_2 may limit its breakthrough in the production gas through faster diffusion into the coal matrix. Moreover, the effect of the coal swelling induced by CO_2 adsorption can also result in slower rate of gas flow in the cleat and consequently increases the time of the breakthrough.

The results of the gas storage and recovery experiments show that, the overall rate of CO_2 displacement with CH_4 is much slower compared to case for N_2. For the N_2 experiment, more than 95% of CH_4 has been displaced within less than 30 minutes, whereas for the case of CO_2 experiment, the fraction of displaced CH_4 has almost reached 90% after 3 hours of continuous CO_2 injection. As stated earlier, higher diffusion rate of CO_2 and the effect of sorption-induced swelling on coal matrix during the CO_2-ECBM process can together govern the slower gas displacement rates observed. Whereas, in the N_2-ECBM process, the slower rate of N_2 diffusion into the coal matrix leads to relatively higher partial pressure of N_2 gas within the coal cleats and therefore faster gas displacement rate and N_2 breakthrough is observed.

3.5. Gas storage and recovery in coal

Based on the experimental data of gas flow rates at upstream and downstream obtained from two mass flow meters, and the composition of the outflow gas obtained from gas analyser, the upstream and downstream mass flow rates were estimated and converted to mole per second (mol/s). The cumulative amounts of gas injected, recovered and stored in the coal sample were then calculated for each experiment. It should be mentioned that the gas storage here implies the total amount of gas adsorbed to the coal matrix as well as the free gas stored in cleats/microfractures and matrix pore volume. Figures 8 and 9 present the cumulative amounts of gas injected, recovered and stored in the coal sample during N_2- and CO_2-ECBM experiments, respectively.

The results presented in Figures 8 and 9 show that for the same duration of the experiment and under similar experimental conditions, the total amount of injected N_2 was 11 times higher than that for CO_2 injection. In addition, the results show that 90% of the total injected N_2 was in the production gas, whereas in the case of the CO_2 experiment, only 63% of the total injected CO_2 was in the production gas. Therefore 36% of the injected CO_2 has been retained within the coal sample.

In another study conducted by Hadi Mosleh (2014), the maximum adsorption capacity of the same coal to CO_2 was estimated 1.21mol/kg. By assuming the total amount of stored CO_2 during CO_2-ECBM experiment as adsorbed gas, it can be postulated that only 20% of the adsorption capacity of coal sample to CO_2 was utilised. This can be related to several factors:

- Effect of gas diffusion rate: the gas adsorption isotherms were measured on powdered coal samples and over 24 to 48 hours, when the equilibrium state was observed (Hadi Mosleh, 2014), whereas the experiments of this study have been conducted on intact 70mm core samples, and over shorter durations (~4hr). Therefore, much longer time was needed for the gas to diffuse into the coal matrix and to achieve equilibrium state.
- Effect of coal swelling: coal swells when it comes into contact with CO_2, this resulted in reduction of coal permeability under confined conditions and therefore a reduction in the accessibility of the coal matrix to more CO_2 gas.
- Effect of effective stress on coal permeability: in gas adsorption measurements by Hadi Mosleh (2014), powdered coal was placed in a high pressure cell without confining pressure applied to coal sample. Whereas in this study, the results of helium and CH_4 flooding experiments have shown that permeability of the coal decreases as effective stress increases. This is related to compression of fractures and microfractures and therefore reduction in accessibility of coal matrix to gas.

3.6. Efficiency of CH_4 recovery

342 Figure 10 shows the cumulative amounts of CH_4 recovered during the N_2-ECBM and CO_2-ECBM experiments. From the results, it may be suggested that the total amounts of CH_4 recovered in N_2- and CO_2-ECBM experiments are almost equal (0.187mol and 183mol, respectively) and therefore both methods can be equally effective in terms of CH_4 recovery. However the rate of CH_4 recovery, which is an important factor affecting the success of ECBM application, should be taken into account when interpreting such results. The results presented in Figure 10 show that the rate of CH_4 recovery for the case of N_2 injection was significantly higher at the early stages of the experiment and reduced considerably for much of the experiment's duration; whereas for the case of CO_2 injection, the rate of CH_4 was found to be more or less steady throughout the experiment. Similar observations have been reported by Yu et al. (2008) based on the core flooding experiments conducted on coal samples from Qinshui basin in North China.

In addition, the efficiency of CH_4 recovery from coal can be related to other factors including: i) the amount of gas required for the injection, ii) the fraction of injected gas in the production gas which needs to be separated from CH_4, and iii) the amount of gas that can be stored in coal which is particularly important for the case of CO_2 sequestration process. Figures 11 and 12 present the ratio of CH_4 recovery with respect to the amount of gas injected and stored, respectively, during both N_2 and CO_2 experiments.

The results of CH_4 recovery versus injected gas (Figure 11) indicate that the CO_2 injection lead to a higher ratio of CH_4 recovery throughout the experiment (up to 10 times higher than that obtained by N_2 injection). This is mainly related to the higher amounts of N_2 injected under similar experimental conditions, *i.e.* gas injection pressure and confining pressure. The amount of the CH_4 recovery for both experiments decreased sharply in the first hour of the experiments. The rate of CH_4 recovery, however, remained almost steady for the N_2 experiment after the first hour, whereas for the CO_2 experiment, it decreased gradually over time. This behaviour can be attributed to the effect of coal matrix swelling on gas permeability and flow as described previously.

The results of CH_4 recovery versus stored gas (Figure 12) also show that storage of CO_2 lead to higher ratio of CH_4 recovery (on average 2.4 times higher than that obtained by N_2 injection). In other words, for every mole of CO_2 stored in the coal sample 1.2mol of CH_4 was recovered. Higher ratios have also been reported in the literature, *e.g.* a ratio of 2 to 1 (Tsotsis et al., 2004) which can be related to the differences in coal type (different sorption capacities and gas flow and mechanical properties) and the experimental conditions.

4. Conclusions

The results of gas storage and recovery from a series of experiments conducted on the CH_4 saturated coal sample were presented on a high rank coal from South Wales coalfield. N_2 and CO_2 were injected at the upstream of the sample and composition of outflow gas was monitored. The results of both experiments were assessed and compared in terms of breakthrough time, gas displacement rate, and efficiency of gas storage and recovery.

The results showed that the N_2 breakthrough time was almost spontaneous whereas for the case of CO_2, the breakthrough time was delayed by a factor of two. Similarly, the gas displacement rate observed in both experiments varied greatly which was mostly related to differences between diffusivity of N_2 and CO_2 as well as higher affinity of the coal to CO_2. The latter effect also resulted in storage of 36% of injected CO_2 in coal. The results of CH_4 recovery showed that with respect to both injected and stored gas, the ratios of CH_4 recovered during CO_2 experiment were higher than those for N_2 experiment.

This study has shown that early N_2 breakthrough and higher rate of N_2 production may lead to additional challenges in order to separate N_2 from CH_4 and thus affects the efficiency of the N_2-ECBM method. For the case of CO_2, the total CH_4 recovery, displacement ratio, breakthrough and CO_2 storage are more favourable. In general, the displacement ratio, CO_2 breakthrough time and CO_2 storage are important parameters affecting the success of carbon sequestration application and

392 the results of this study provide new insights into the efficiency of gas recovery and storage in
393 anthracite coal using carbon dioxide and under the experimental conditions applied.

394 Acknowledgements

395 The financial support received from the Welsh-European Funding Office as part of the
396 Geoenvironmental Research Centre's SEREN project is gratefully acknowledged. The authors
397 would like to thank Dr Snehasis Tripathy for his support. We also would like to thank GDS
398 Instruments for their contribution for construction and commissioning of laboratory equipment.
399 Technical support from the technicians and staff of the Engineering Workshop at Cardiff
400 University is also gratefully acknowledged.

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Table 1. Physical and chemical properties of the coal sample.

Moisture (wt%)	1.2	Carbon (%)	86.4
Sample diameter (*mm*)	70	Volatile matter (%)	9.7
Sample length (*mm*)	120	Fixed carbon (%)	84.4
Bulk density (kg/m^3)	1398	Sulphur (%)	0.8
Porosity (-)	0.05	Ash (%)	4.9

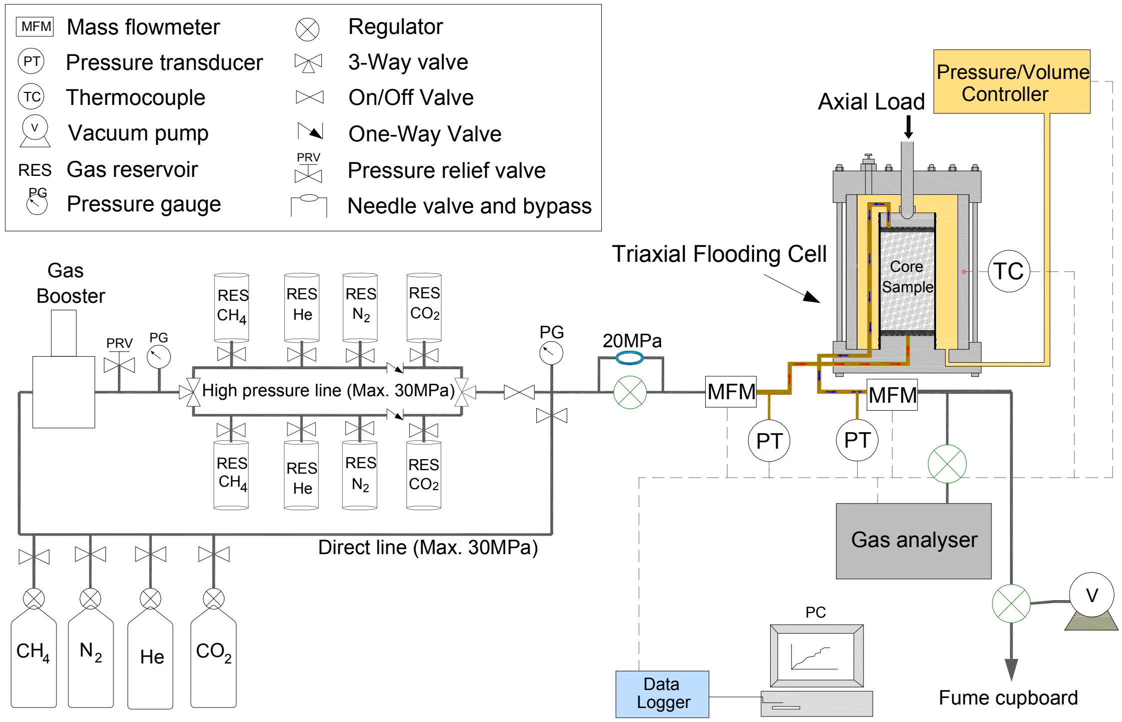

Figure 1. A schematic diagram of the developed laboratory facility (Hadi Mosleh et al., 2017).

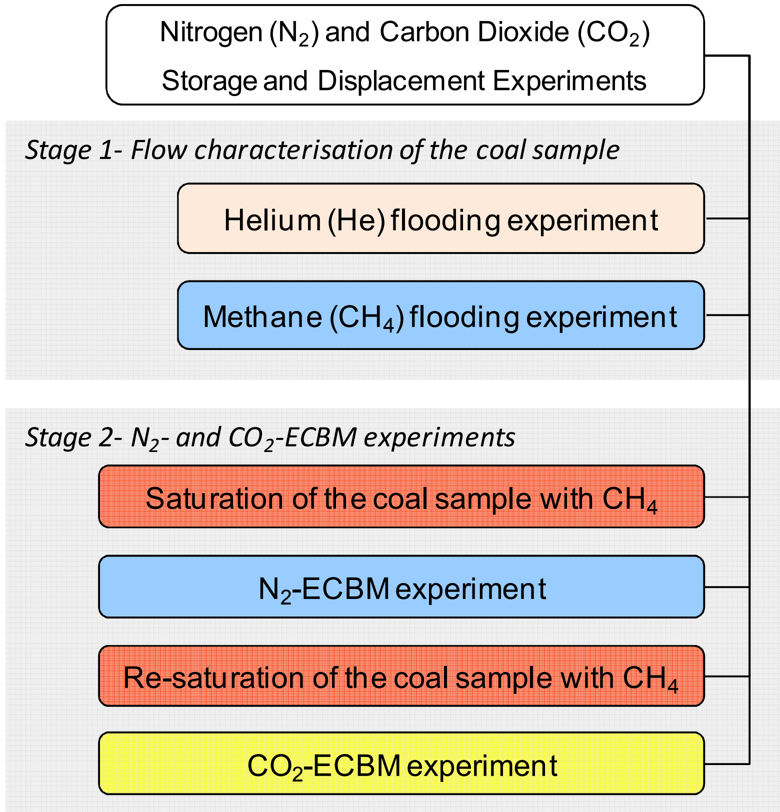

Figure 2. The order of experimental studies carried out.

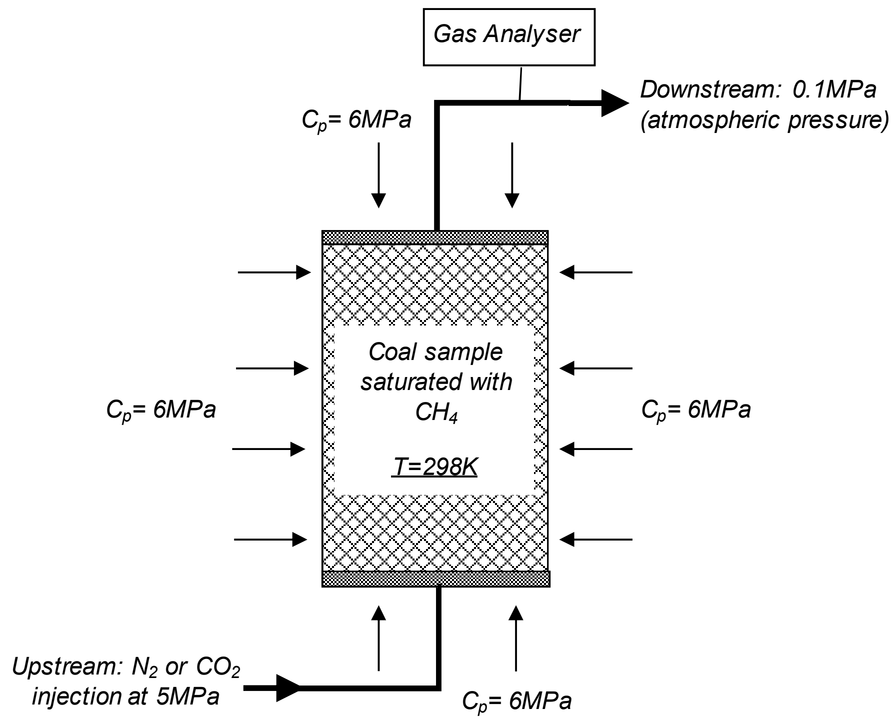

Figure 3. Schematic diagram of the experimental conditions applied for the N_2 and CO_2 ECBM experiments, C_p is the confining pressure.

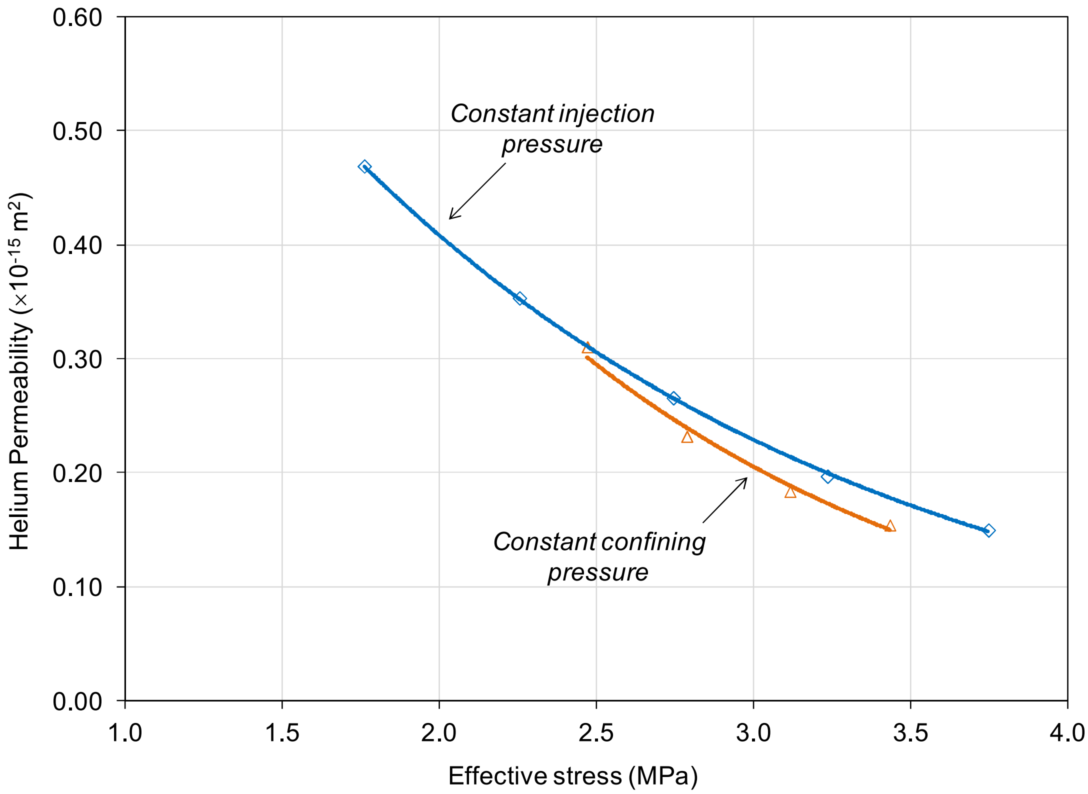

Figure 4a. Variations of the coal permeability to helium with effective stress under constant injection pressure and constant confining pressure.

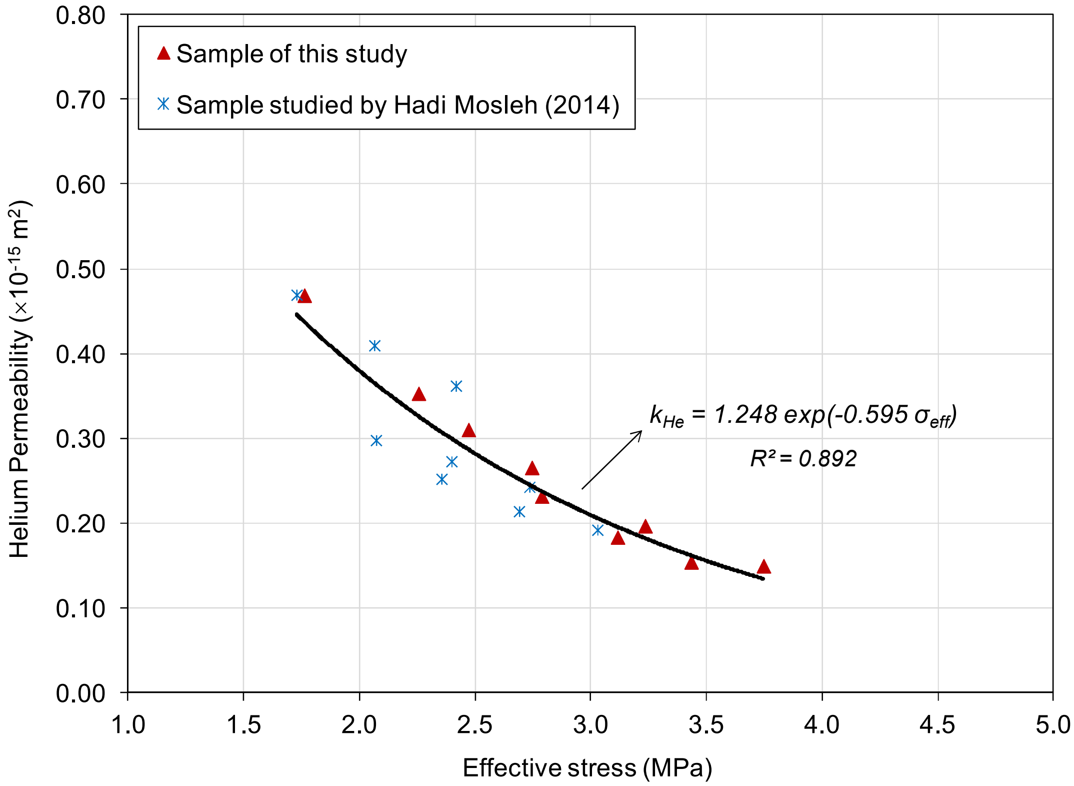

Figure 4b. Variation of coal permeability to helium with effective stress for the sample of this study and a sister sample obtained from same block of coal (Hadi Mosleh, 2014).

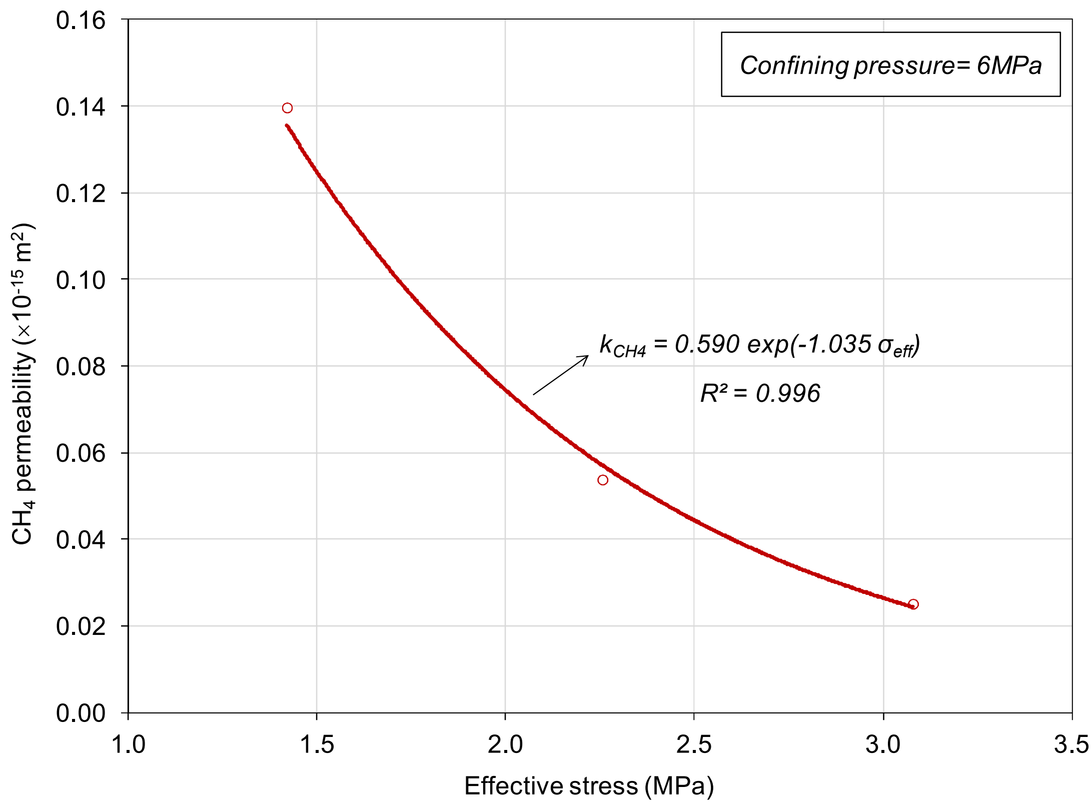

Figure 5. The relationship between permeability of the coal sample to CH_4 and effective stress.

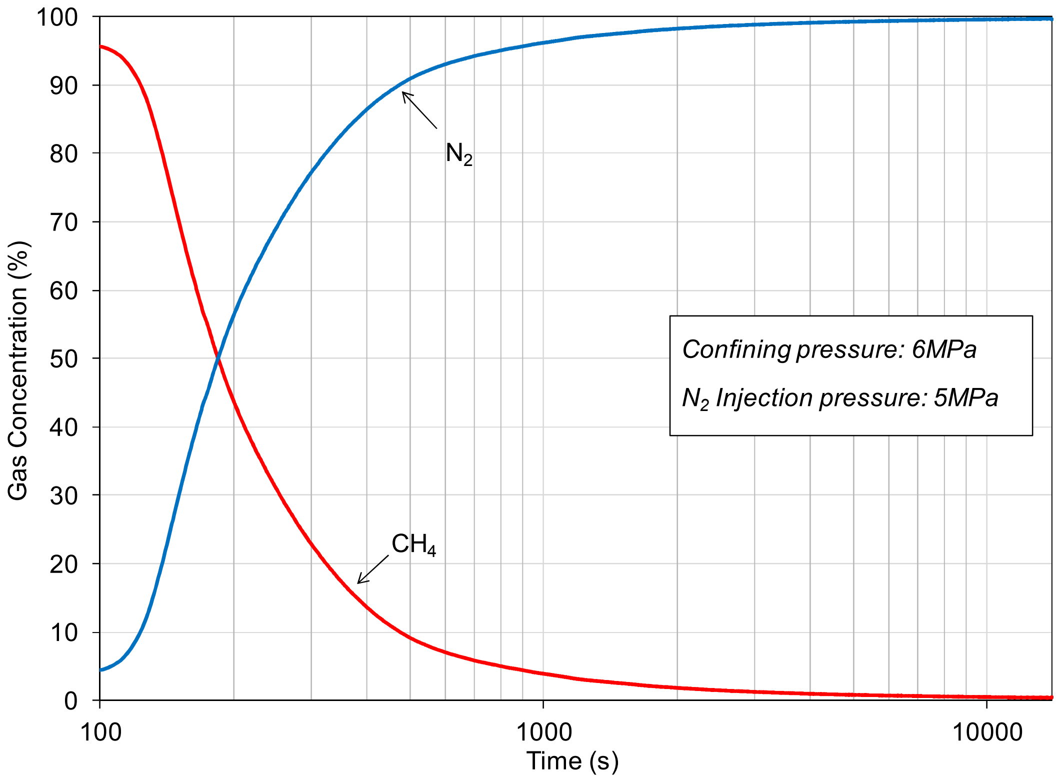

Figure 6. The composition of the production gas with time during the N_2 experiment.

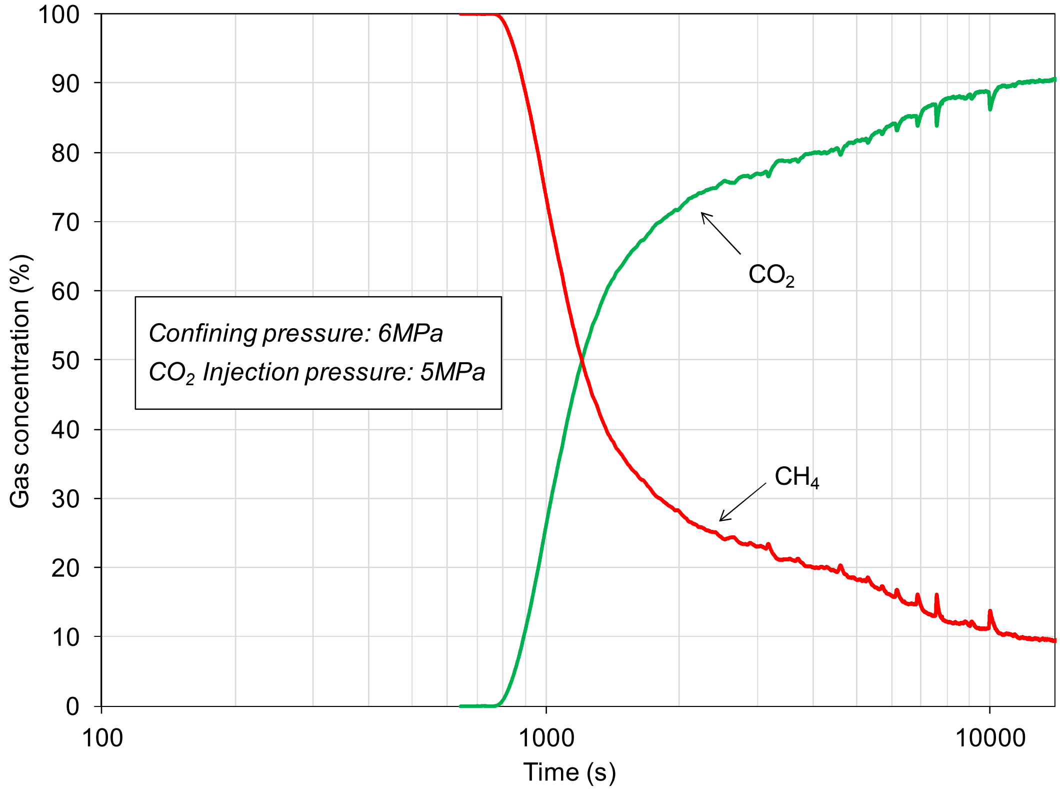

Figure 7. The composition of the production gas with time during the CO_2 experiment.

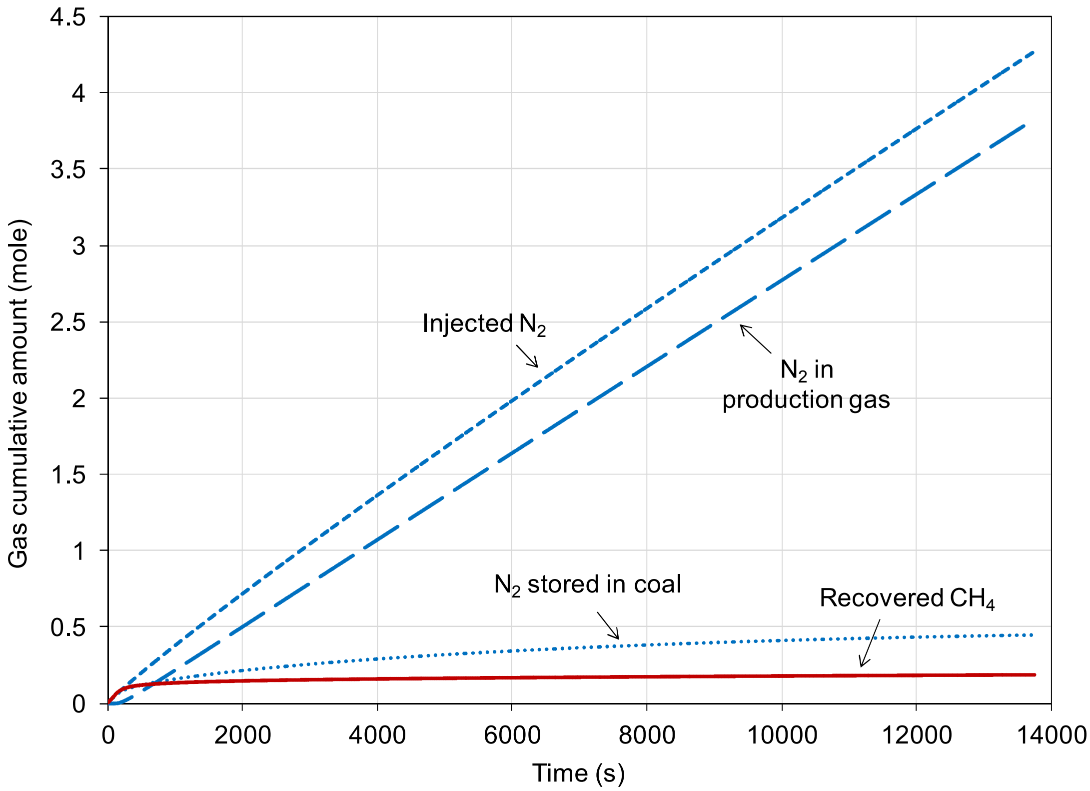

Figure 8. Cumulative amounts of N_2 injected, recovered and stored with time during the N_2-ECBM experiment.

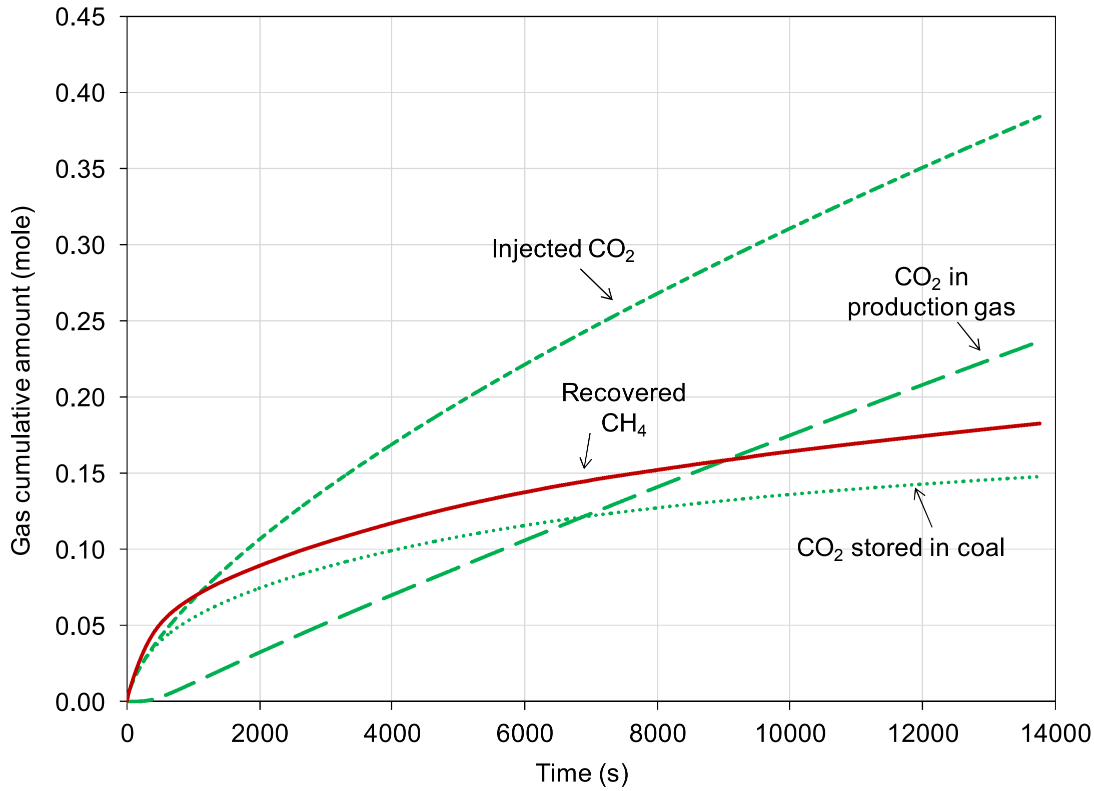

Figure 9. Cumulative amounts of CO_2 injected, recovered and stored with time during the CO_2-ECBM experiments.

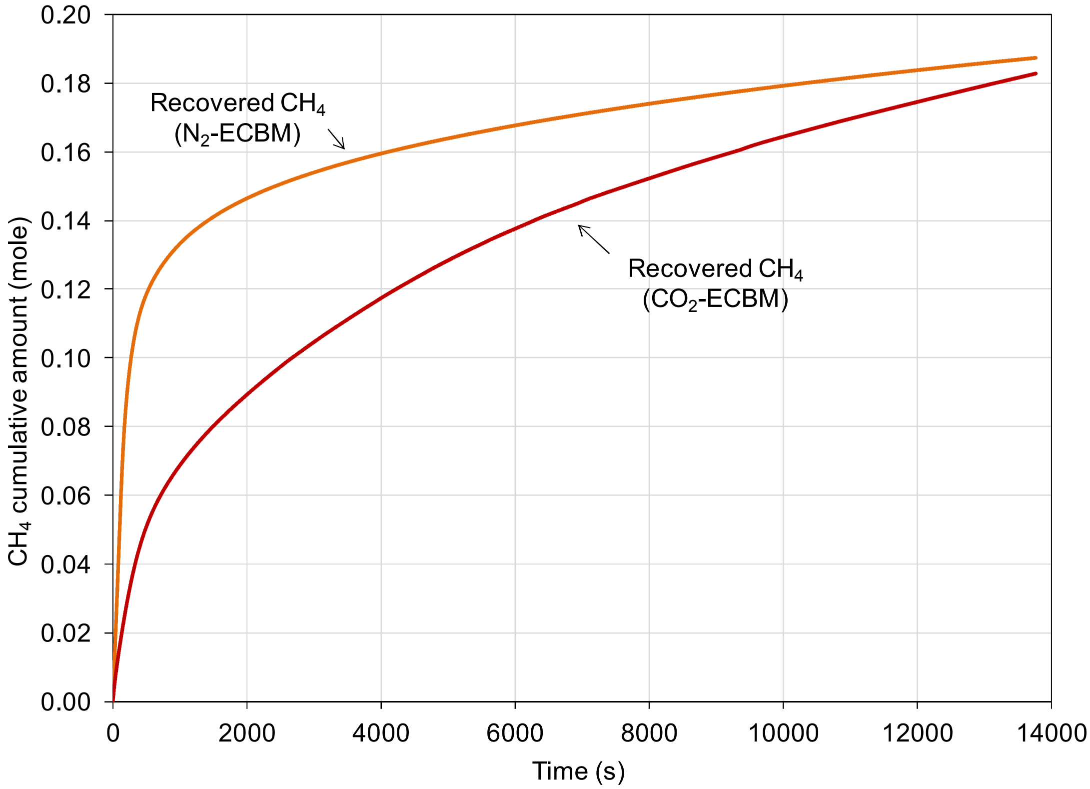

Figure 10. Cumulative amounts of CH_4 recovery during the N_2-ECBM and CO_2-ECBM experiments.

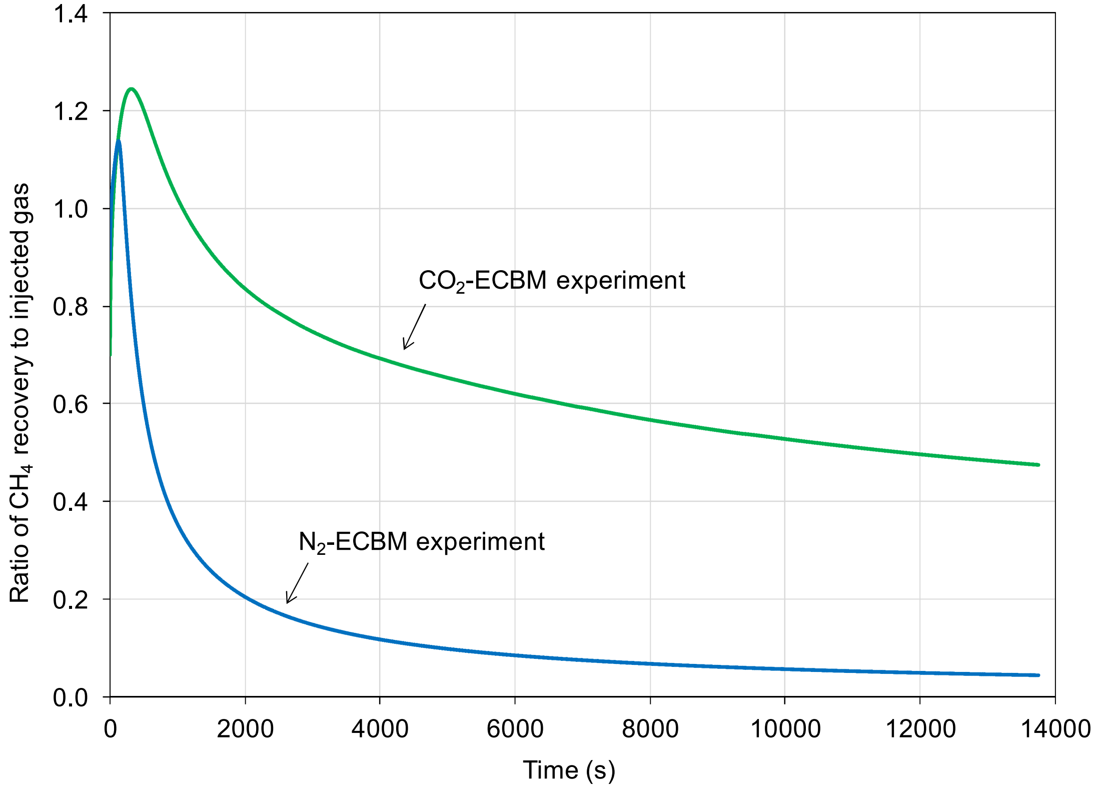

Figure 11. The ratio of CH_4 recovery to injected gas and its variation with time.

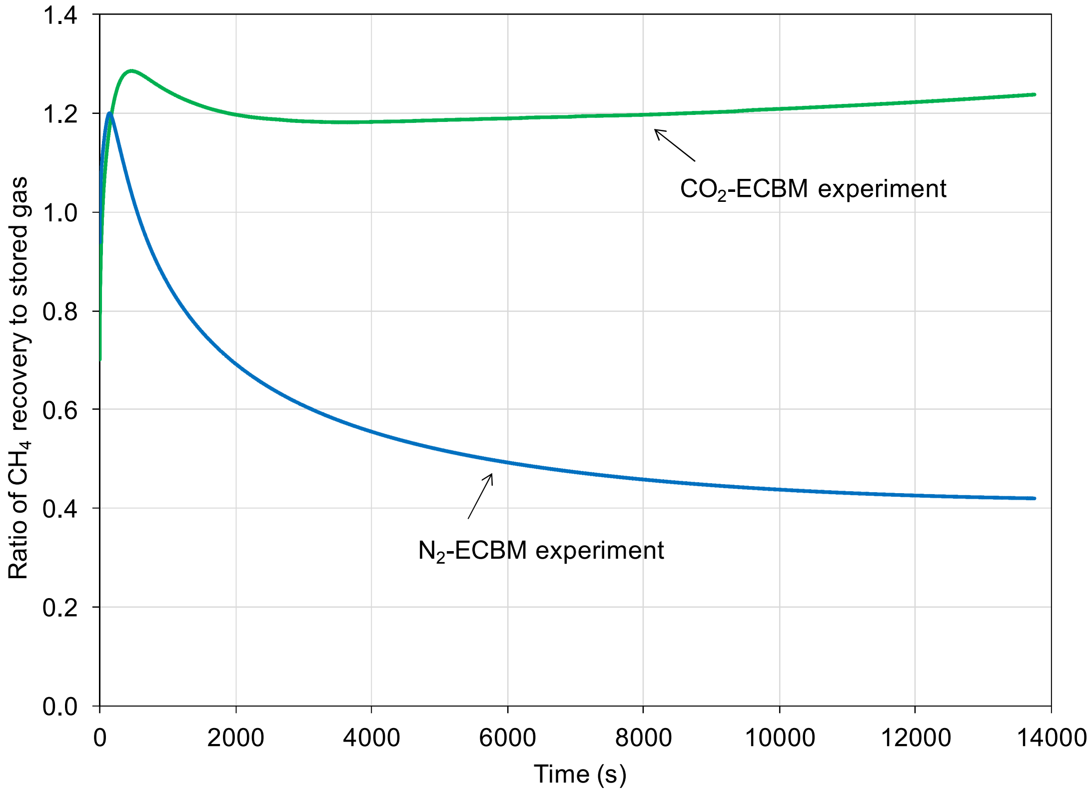

Figure 12. The ratio of CH_4 recovery to stored gas and its variation with time.