Novel High Pressure Multi-Component Diffusion Cell

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Abstract— **A novel high pressure multi-component diffusion cell (HPMCDC) apparatus has been designed and built to measure single and binary gas diffusion, including co-current and counter-diffusion, from low to high pressures. The apparatus incorporates capability to investigate scale effects in solid coal specimens, up to 25 mm in diameter and 25 mm in thickness. Future experiments will be conducted to measure diffusion and counter-diffusion of CH_4 and CO_2 gases in solid coal, at various temperatures, pressures and for three distinct ranks of coal. The experiments will also address the frequent and controversial literature conclusions that the apparent-diffusion of CH_4, inconsistent with gas diffusion theory.**

Keywords—**Coalbed Methane, carbon dioxide, methane, diffusion.**

I. INTRODUCTION

Coalbed methane (CBM) or coal seam gas is an unconventional gas found in underground coal that formed as a result of: (1) thermogenic processes in high rank coals (bituminous to anthracite) during coalification; (2) biogenic processes, mainly in low rank coals (subbituminous) is generated by the activity of bacteria; and (3) secondary biogenic gas accumulating in high rank coal [1-3].

In recent decades, coal seam gas or coal bed methane (CBM) has become an important source of energy in some countries in the world. Production of coal seam gas is complex and difficult to predict and analyse, especially at the early stages of recovery. Gas production from reservoirs of coal bed methane is governed by the complex interaction of single phase gas diffusion through the micropore system, called the matrix, and two phase gas and water flow through the macropore system, i.e. in the natural fractures called cleats. The gas is desorbed from the micropores (matrix) and then then diffuses through the micropores and mesopores, finally entering the macropore (cleat) system [4].

II. LITERATURE REVIEW

Aminian [4] predicted the initial CBM production rate and the total life production profile as a complex and difficult exercise, as three key fluid transport steps are involved. Accurate models need to include a complete understanding of the combined effect of large scale variations, significant spatial and directional variations, and dynamic changes over the production life. The key production processes are: desorption, diffusion and Darcy flow. Gas diffusion occurs throughout the micropore system, called matrix, and through the macropore fracture system, called cleats, as shown in Fig. 1 below:
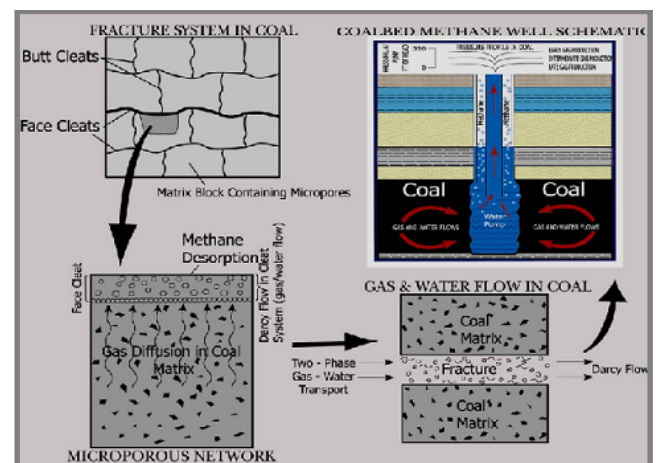

Fig. 1 Gas and water transport flow chart from coal matrix through cleat fractures to borehole during dewatering of a coal reservoir [2]

A. Coal Diffusion

There is an on-going need to improve the understanding of methane gas diffusion throughout the large variation in pore sizes present in coal bed methane reservoirs. Since 1950, CH_4 diffusion in coal has been studied by doing desorption of CH_4 during extraction of coal samples from the sub-surface [5, 6], but it is still poorly understood.

In order to produce gas from CBM reservoirs, there are four steps needed to be controlled, (1) the dewatering process, (2) the desorption of gas from coal surface, (3) diffusion of gas to the fracture systems, and (4) flow of the

gas through the fractures to the wellbores [7, 8]. The measured values of gas desorption rate are generally controlled by two processes: (1) the sorption process (sorption characteristic of the coal) and (2) the diffusion process (diffusion of gas through the coal matrix). These two processes are usually lumped together and described by the parameter of *sorption time* in numerical models [9] and this is the primary characteristic used by some practitioners for evaluation and predictive modelling of coal reservoirs [10, 11].

B. Coal Porosity

In simplified models, coal seams are characterized by a dual porosity system, consisting of micropore and macropores. The micro-pores are contained in the coal matrix, which is highly heterogeneous. The majority of CBM is present in the sorbed state in these micropores, particularly at low reservoir pressures. The macropore system is established by the natural fracture network known as the cleat system [12].

There are two main transport mechanisms which control gas flow in coal: viscous laminar flow through the cleats, which follows Darcy's law; and diffusion through the coal matrix bounded by the cleats, which follows Fick's law [13]. Cleats consist of the more continuous face cleats and less continuous butt cleats. Usually, the cleat system is the primary water and gas pathway during production. On the other hand, other fracture systems often occur at a micrometre scale to form micro cleats in the coal matrix, although its size, shape and continuity are also affected by coal lithotypes [14, 15]. Based on the literature, pore structure in coal matrix is highly heterogeneous. It is commonly divided into three size categories: micropores (<2 nm in diameter), mesopores (>2 and <50 nm) and macropores (>50 nm), [16, 17]. More than 95% of the coal matrix pores across these pore sizes act as an adsorption surface in coal [18].

The different coal porosities make a large contribution to the swelling and shrinkage of coal during adsorption and desorption processes [16, 19].

In subbituminous coal samples, micropores could contribute 50 to 60% of the total matrix porosity of 13 to 25% [20, 21]. Significantly, Levine in 1993 [22] mentioned that the dimensions of the micropores and fine mesopores found in the coal matrix material imply that the porosity is intermolecular, as opposed to interparticulate, and is determined by molecular interactions [21]. These molecular interactions imply the accessibility of the pore system will depend on the fluid used for its determination (such as; He versus N_2 versus CO_2) and therefore that the porosity of coal is not a fixed value, but is a function of the fluid used to measure porosity..

C. Coal Shrinkage & Swelling

One of the potential problems during carbon dioxide sequestration is shrinkage and swelling that occurs in coal [23-29].

Shrinkage and swelling of coal the matrix is a function of pressure, type of gas adsorbed and the coal rank [30]. The components of dependency of swelling are shown in the experimental study done by Cui et al. (2007) [31]. The swelling effect of CO_2 is greater than those of CH_4 and N_2 at the same pressure. Then, when the pressure of each gas component increases, the swelling effect also increases. Therefore, injection of CO_2 at high pressures is expected to lead to increased swelling and decreased injection capacity.

Coal shrinkage can have a significant impact on increased cleat permeability in CBM production. Though there are now quite a number of laboratory studies confirming this behaviour, the literature reports only limited field examples of this (San Juan basin, USA, and Scotia field, Australia). The molecular and micro-structure lattice effects are still poorly understood. It is important for enhanced coalbed methane (ECBM) production that via geologic sequestration of carbon dioxide (CO_2), that multi-component diffusion be lab-evaluated, prior to undertaking large scale sequestration projects. Anderson (1965) and Walker (1956) noted that the micropore system in coal is not readily accessible to the N_2 molecule at 77 K because of an activated diffusion process and/or shrinkage of pores. Bybee (2007) [32] summarized that the influence of swelling and shrinkage in coal needs to be investigated and may cause significant changes in permeability that result in significant reservoir pressure effects. The swelling and shrinkage will depend on gas type as well as the state of adsorption and desorption.

D. Moisture Content

In his paper Gauger (1932) concluded that the water content in coals originates from the following sources: (1) decomposition of organic molecules (called combined water), (2) surface-adsorbed water, (3) capillary-condensed water, (4) dissolved water, and (5) water of hydration of inorganic constituent of the coal. Brown (1953) also mentioned that the moisture content in coal is divided into constituent categories: (1) free or adherent moisture (essentially surface adsorbed) possessing the physical properties of ordinary water; (2) physically bound or inherent moisture with vapour pressure lowered by the small diameter of the pores of the coal structure in which it is absorbed; and (3) chemically bound water of hydration or combined water.

Since the moisture content in the matrix occupies pore volume and part of the moisture adsorbs on the pore surface, it definitely has an impact on the mechanisms in gas diffusion behaviour [33].

III. AIM AND METHODOLOGY

A further motivation for this study was to investigate the various experimental methodologies used in the literature to measure coal diffusion. This study reviews and comments on their advantages and disadvantages, providing guidance for the design of improved measurement techniques. We note the surprising conclusion regarding the greater *relative* diffusivity of CO_2 compared to CH_4 in coal seams reported in the literature [10, 34-37]. For these reasons, it is important to understand the nature of diffusion in microporous solids, to differentiate between diffusion and other transport phenomena and develop more accurate model.

IV. EXPERIMENTAL

A. New Design of Diffusivity Apparatus

In order to address the above issues, we have developed an advanced design of a high pressure diffusion cell, capable of measuring diffusion in solid coal samples at varying temperatures and pressures and with a counter-diffusion capability. This High Pressure Multi-Component Diffusion Cell (HPMCDC) is shown in **Error! Reference source not found.**. The pressure can be set between 1 and 10 MPa. It is designed to be used in single gas diffusion or in counter or co-current diffusion of a binary gas mixture. The cylindrical coal samples can be up to 25 mm thick and are generally 25 mm in diameter. The CH_4 and CO_2 are introduced into the cell, and the temperature as well as pressure can be varied. A computerised data recording system will capture experimental data, including time. Each experiment is influenced by the variation of each parameter. The composition of gas(es) diffusing through the coal sample can be measured by collecting the gas and passing it into a Gas Chromatograph.
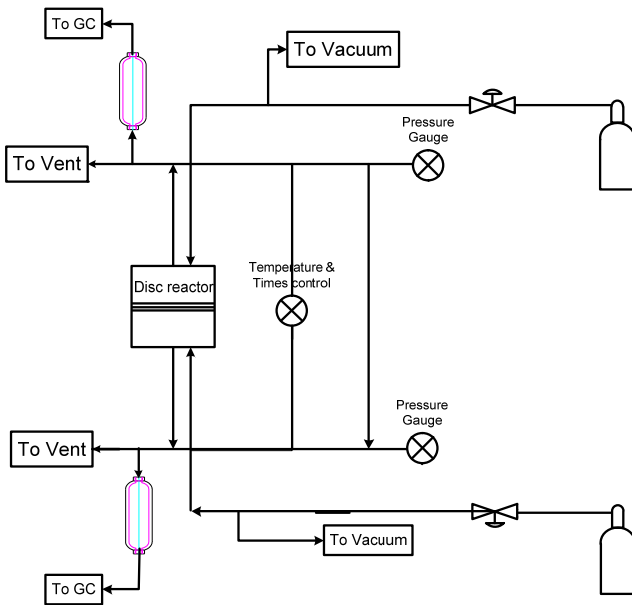

Fig. 2 Schematic of High Presure Multi Component Diffusivity Cell (HPMCDC)

The figure 3 below shows the flexibility of the cell reactor of HPMCDC. This is very useful for analysing coal samples in various thicknesses by only using one cell reactor.
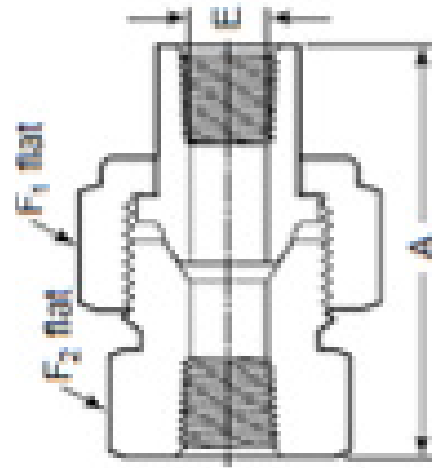

Fig. 3 Flexibility in thickness of diffusion cell of HPMCDC

V. GAPS IN KNOWLEDGE

In order to have practical limits on the many permutations of experiments that could be conducted using the HPMCDC, several gaps in knowledge have been identified as focus areas for investigation. They are: (1) Develop methods to examine diffusivity in coal in both counter and co-current configurations; (2) Review literature arguments and conclusions on the relative diffusivities between CH_4 and CO_2 in coal; (3) Examine the effect of the size of the coal matrix by using different thickness the coal samples ; (4) Measure relative diffusivity based on varying moisture contents in the coal matrix; (5) Examine the effect of swelling and shrinkage in coal on diffusivity; and (6) Perform other supportive measurements, such as: petrography of coal, proximate and ultimate analysis, coal rank, chemical and physical analysis; and pore size distribution studies with various test fluids and apparatus.

VI. CONCLUSIONS

Further studies are much recommended to start the experiments of gas diffusivities in coal, to address the controversial conclusions from many historical experiments, whereby the apparent-diffusion of CO_2 in coal is larger by an order of magnitude than the apparent diffusion of CH_4.

The new HPMCDC is adequately designed to investigate leading-edge effects of solid coal sample size, diffusion, binary co-diffusion and binary counter-diffusion, as well as temperature and pressure effects, in the range required by the CBM & ECBM industry.

NOMENCLATURE

CBM   Coal Bed Methane
ECBM   Enhanced Coal Bed Methane
CH_4 Methane
CO_2 Carbon dioxide

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