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Carbon dioxide flow and interactions in a high rank coal: Permeability evolution and reversibility of reactive processes

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ABSTRACT

Uncertainties exist on the efficiency of CO_2 injection and storage in deep unminable coal seems due to potential reduction in the permeability of coal that is induced by CO_2 adsorption into the coal matrix. In addition, there is a limited knowledge about the stability of CO_2 stored in coal due to changes in gas partial pressure caused by potential leakage. This paper presents an experimental study on permeability evolution in a high rank coal from South Wales coalfield due to interaction with different types of gases. The reversibility of the processes and stability of the stored CO_2 in coal are investigated *via* a series of core flooding experiments in a bespoke triaxial flooding setup. A comprehensive and new set of high-resolution data on the permeability evolution of anthracite coal is presented.

The results show a considerable reduction of permeability above 1.5 MPa CO_2 pressure that is correlated with the coal matrix swelling induced by CO_2 adsorption. Notably studied in this work, the chemically-induced strain due to gas sorption into coal, that has been isolated and quantified from the mechanically-induced strain as a result of changes in effective stress conditions. The results of post-CO_2 core flooding tests using helium (He), nitrogen (N_2) and methane (CH_4) demonstrated a degree of restoration of the initial permeability. The injection of N_2 showed no significant changes in the coal permeability and reversibility of matrix swelling. The initial permeability of the coal sample was partially restored after replacing N_2 by CH_4. Observation of permeability evolution indicates that the stored CO_2 has remained stable in coal under the conditions of the experiments.

1. Introduction

Emerging interest in deep subsurface energy applications related to geological carbon sequestration has highlighted the importance of an in-depth understanding of the complex physical and chemical phenomena that can occur during gas-rock interactions. Among those are the processes related to gas flow in coal, which are relevant to applications such as CO_2 sequestration in unminable coal seams and coalbed methane recovery. Complex and coupled physical, chemical and mechanical processes can occur during the flow of gas species in coal, affecting the key flow property of the coal, *i.e.* permeability. This is highlighted for the case of CO_2 interaction with coal due to the chemical and physical changes in the coal microstructure during adsorption and desorption (White et al., 2005).

It has been shown that the permeability of coal to gas species is dependent on several factors, including cleat and fracture systems (Harpalani and Chen, 1997; Olson et al., 2009), porosity, type of gas and pressure and mechanical stresses (Somerton et al., 1975; Palmer and Mansoori, 1998; Sasaki et al., 2004), fracture orientation (Laubach et al., 1998), and the effects of matrix swelling/shrinkage induced by gas sorption. The permeability of coal can decrease with an increase in the effective stress (*e.g.*, McKee et al., 1988; Jasinge et al., 2011). An increase in the effective stress can cause compression of the pore space available for gas flow, resulting in permeability reduction (Ranjith and Perera, 2011). It has been shown that the uptake or release of CO_2 and CH_4 is a combination of adsorption or desorption processes together with matrix swelling and shrinkage (Mazzotti et al., 2009). The amount of swelling depends on a number of parameters, including the structure and properties of the coal, gas composition, confining stress, pore pressure, temperature, fracture geometry and moisture content (Wang et al., 2013).

Compared to the extensive reported studies related to the adsorption and desorption of gases in coal (mostly on powdered samples), a limited number of experimental investigations have been reported on

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gas transport and reactions in intact coal samples based on core flooding experiments. Tsotsis et al. (2004) reported core flooding experiments to study the mechanisms involved in CO_2 sequestration in a highly volatile bituminous coal. Mazumder and Wolf (2008) conducted core flooding experiments on dry and wet coal samples from the Beringen coal mines in Belgium, the Silesian coalfield in Poland, and the Tupton coalfields in the UK. Yu et al. (2008) performed gas storage and displacement experiments on coal samples originated from the Jincheng and Luan mines, Qinshui basin, North China. Wang et al. (2010) have reported core flooding experiments on high volatile bituminous coal from the Bowen Basin, Australia, and van Hemert et al. (2012) conducted a series of gas storage and recovery experiments (ECBM) on coal samples from Nottinghamshire by injecting N_2, CO_2 and mixtures of these two gases. Similarly, Connell et al. (2011) studied CH_4 displacement experiment with N_2 on a coal sample from The Bowen Basin, Australia at low and high gas injection pressures up to 10 MPa. Gas adsorption and desorption in the coal matrix has been shown to be an influential factor in permeability evolution by inducing swelling and shrinkage in coal matrix. Massarotto et al. (2007) observed permeability increases between 100 to 1200% during CH_4 desorption, compared to permeability decreases of 60–80% during CO_2 adsorption. In a study by Harpalani and Mitra (2010), the reduction of permeability to CH_4 was found to be approximately 25% of the original value, whereas the permeability to CO_2 was found to be 40% less than that to CH_4. It was reported that at elevated gas pressures, the swelling increased nearly linearly with the amount of CO_2 adsorbed (van Bergen et al., 2009). At pressures higher than 8 MPa, the gas adsorption continued to increase but the coal matrix volume remained constant, *i.e.* no coal matrix swelling occurred (Harpalani and Mitra, 2010; Kelemen et al., 2006; Gensterblum et al., 2010). Harpalani and Mitra (2010) showed that the volumetric strain of coal due to CO_2 or CH_4 adsorption followed a Langmuir-type model.

Despite extensive efforts to explore the complex and coupled phenomena involved in gas-coal interactions, understanding of the processes that can occur when CO_2 is injected into the coal and stability of the adsorbed gas in coal is incomplete. In particular, there is limited experimental knowledge related to the behaviour of high rank coals, *i.e.* anthracite, during flow and interaction with different gases. Modelling concepts have been developed in the last two decades to simulate the flow of gas in fractured rock including coal (*e.g.* Shi and Durucan, 2003; Salimzadeh and Khalili, 2015; Hosking, 2014) that are usually based on single or double porosity approaches. These models are usually based on mechanistic approaches that require appropriate constitutive relationships (*e.g.* gas permeability model) and experimental data for testing. Appropriate models/constitutive relationships for coal permeability should reflect the chemo-mechanics of the carbon sequestration and/or enhanced coalbed methane recovery problem that require experimental dataset for testing and evaluation.

The investigation presented in this paper aims to address two key phenomena related to flow of gases in a high rank coal: i) the permeability evolution of coal to different gas species under a range of gas pressures and stress conditions, with particular focus on the adsorption induced coal matrix swelling and permeability degradation during CO_2 injection, and ii) the reversibility of reactive transport processes and stability of CO_2 adsorbed in coal based on indirect observations of permeability evolution. The latter has been achieved by altering the partial gas pressure in coal *via* a sequence of core flooding experiments using different types of gases. These are important aspects related to i) the efficiency of CO_2 storage and potential changes in the storage capacity due to permeability evolution, and ii) the stability of stored CO_2 within the reservoir in case of any changes in gas partial pressure due to potential leakage events.

A novel sequence of core flooding experiments has been designed and conducted in two stages (Fig. 1). In Stage 1, permeability evolution and deformation of the coal sample by exposure to He, N_2 and CO_2 were studied for a range of gas injection pressures and confining stresses, and in Stage 2, the same coal sample (after interactions with CO_2) was subjected to He, N_2, and CH_4 injections and due to the reduction of CO_2 partial pressure in the cleats, changes in intrinsic permeability was used as an indication of CO_2 desorption.

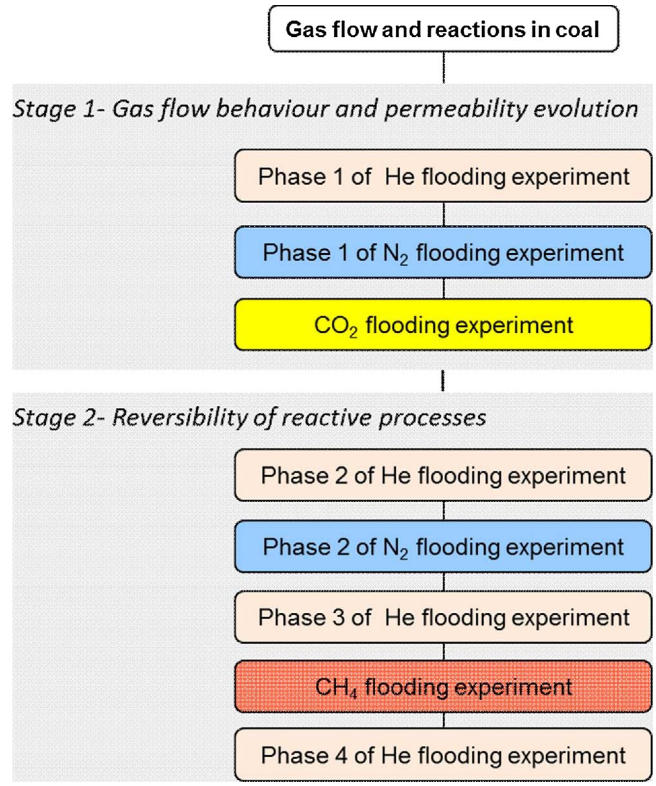

Fig. 1. The flow diagram of the experimental studies on gas flow behaviour in coal and permeability evolution.

2. Materials and methods

2.1. Triaxial core flooding setup

The experimental facility developed and used consists of i) a high pressure triaxial core flooding system by which the transport and deformation properties can be measured and studied, ii) a pressure control system, iii) a temperature control system, and iv) the ancillary system including pure and mixed gas supply and analysis units (Hadi Mosleh et al., 2017b). A schematic diagram of the developed laboratory facility is presented in Fig. 2.

The triaxial cell includes a base pedestal, a top-cap, an internal submersible load cell, and local strain transducers. The core sample sits within a rubber sleeve (Fig. 3a), 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. Two axial and one radial local strain transducers (Linear Variable Differential Transformer (LVDT) from GDS Instruments) are attached to the sleeve (Fig. 3a) in order to measure the volumetric deformation of the sample under axial and radial strain conditions. In addition, a ± 0.025 m displacement transducer with an accuracy of 0.25% has been used to measure the axial displacement of the sample. A Mass Flow Meter capable of measuring high flow rates up to 17×10^{-6} m^3/s (1 L/min) was used that is capable of working under both subcritical and supercritical conditions, with pressures up to 20 MPa.

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 32 MPa in-line pore pressure transducers were selected to measure the inlet and the outlet

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