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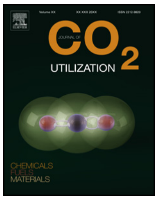

In-situ disposal of CO_2: Liquid and supercritical CO_2 permeability in coal at multiple down-hole stress conditions

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ABSTRACT

Geological CO_2 sequestration is one of the most effective methods to counter global climate change. Coal matrix shrinkage, swelling, gas diffusion and permeability are the key phenomena associated with geological CO_2 disposal in coal. In this study, permeability experiments were approached using the supercritical and liquid phases of CO_2 (less understood; most likely insitu phases) for naturally fractured bituminous coal. Experiments were performed under triaxial conditions using four sets of various confinement conditions corresponding to variable depths. Injection pressure was varied gradually and the permeability changes were calculated using the Darcy's equation for subcritical and supercritical CO_2 exclusively. Changes in CO_2 phases were obtained by changing the system temperature from 26° C for liquid CO_2 to 34 °C for supercritical CO_2. N_2 was alternatively injected at the start and between the injections of CO_2 to analyze the changes in the permeability from a relatively very less sorptive medium's perspective. It was observed that the supercritical CO_2 flow reduced the permeability significantly and this behavior was greatly attributed to the highly viscous nature of the supercritical phase and the high volumetric deformation or the swelling of coal under supercritical CO_2 as compared to liquid CO_2. The injection pressures were observed to reduce the effective stress behavior, which in turn pushed the permeability evolution at each confinement to a positive trend. However, the permeability of CO_2 reduced exponentially with increasing effective stresses. Two different empirical equations were proposed for permeability of both the phases of CO_2 with effective stresses.

1. Introduction

Anthropogenic contribution resulting in the rise of CO_2 has exponentially risen, especially in the past three decades, with atmospheric CO_2 levels going above 400 ppm in April 2014 at all World Meteorological Organization (WMO) stations in the northern hemisphere [1]. Geological CO_2 sequestration is one of the most effective and readily available methods to keep the rise in CO_2 levels in control and achieve climatic targets. In India, which is the world's third largest emitter of CO_2 after China and U.S.A respectively, the unmineable deep coal seams are among the most promising candidates to physically sequester CO_2, with or without coalbed methane recovery [2,3]. According to a survey in 2009, coal's contribution via power and steel plants to the Indian carbon emissions was 72% [4]. Thus, in context of operational proximity and availability of storage reservoirs, coalbed sequestration in India is a more favorable option when compared to mineral trapping in basalt formations, offshore sequestration in sedimentary basins, and depleted oil and gas reservoirs. In coal, CO_2 may be sequestered in three states: free-space (gas, liquid or supercritical fluid), in solution in mine water, and adsorbed to the remaining coal in the mine [5,6]. Geological media suitable for CO_2 storage must have the capacity to store the intended volume of CO_2. This enables them to accept the injectant at the intended flow rate and confinement potential for safe storage in geological time scales [7]. The structure of coal makes it one of the prime options for CO_2 storage in the above context. Also, CO_2 has greater affinity to adsorb onto the coal's microstructure among CO_2, CH_4 and N_2 [8] making it a feasible injectant for enhanced coalbed methane (ECBM). Potential for CO_2 storage in coal reservoirs with/without CBM recovery has been justified in a diverse range of reservoirs throughout the world [9–19].

Previous permeability studies on Indian coal have been carried out with the sub-critical phase of CO_2 at varying confinements for both intact and naturally fractured coal specimens [2,20]. However, in case of deep seam sequestration, the pressure temperature (P-T) conditions could lead to either liquid or supercritical stage of CO_2. CO_2 exists in supercritical state above

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the pressure of 7.38 MPa and temperature of 31.1° C, which is likely to be encountered at depths more than 800 m below the surface. The critical point of CO_2 is the transition zone for CO_2 in the P-T regime, where significant changes in the hydro-dynamic properties of CO_2 occur for relatively small fluctuations in the P-T conditions near the critical point. In the Indian context, typical depths for underground coal mining range between 300 m and 1200 m below ground level. Confining seam conditions in this range can induce phase transitions in CO_2, which further affects the coal-fluid interactions. According to Krooss et al. [21], CO_2 has higher adsorption affinity into the coal matrix under super-critical conditions as compared to sub-critical conditions whereas White et al. [10] mentioned that the supercritical CO_2 has a higher potential to displace the existing gases from coal. Thus, it is vital to investigate the influence of liquid and supercritical CO_2 flow at varying confinements, corresponding to different depths of coal seam.

The aspect of permeability evolution in coal upon CO_2 injection in subcritical phases have been studied in the past [22–30] and there have been a few studies on the behavior of supercritical CO_2 flow upon injection into fractured and intact coals. Toribio et al. [31] and Perera et al. [32] observed that there was an increase in sorption when the CO_2 phase condition changed from subcritical to supercritical state. This is due to the fact that supercritical CO_2 has greater affinity for coal sorption when compared to subcritical CO_2. Also, Perera et al. [33] described that the high improvement in viscosity of the fluid at high downstream pressure could be the reason for the sudden decline in the permeability apart from the usually induced swelling of coal. While many studies have gone into injection of liquid/supercritical CO_2 for CBM extraction [34,10,35], very limited studies have been carried out in context of observational permeability evolution with respect to varied experimental conditions. Also, till date, no experimental, analytical or modeling study has been carried out in the area of permeability behavior upon injection of liquid and supercritical CO_2 in context to Indian Gondwana coals which form a large share of coalfields worldwide, except one recent study on the role of saturation cycles on permeability of coal by this research group recently [36]. In this study, a detailed experimental scheme was followed to study the permeability of coal under deep in-situ conditions at multiple injection pressures and effective stresses.

2. Methodology

Bituminous coal samples from Damodar valley coalfields, in the state of Jharkhand, India were used in this study. The samples used for CO_2 flow studies were vitrinite and inertinite rich and contained 86.23% by mass of pure coal. The samples were obtained from drill cores and were polished in the laboratory and stored in a humidity chamber to prevent the loss of moisture. As the objective of the study was to study the flow of CO_2 through the coal sample under in-situ conditions, a high pressure triaxial setup with the capability of fluid flow through the sample under confinement conditions and axial loading was used. The detailed experimental procedure used in this study is described in the scientific articles published earlier [37]. A simple schematic of the experimental setup is shown in Fig. 1. As two different high pressure phases of CO_2 were being used in the experimental procedure, the system was thoroughly examined for safety. High values of confining stresses were used keeping the variations same for tests carried out using both phases of CO_2. Constant temperature was maintained at 26° C for liquid CO_2 and 34° C for supercritical CO_2 flow.

During experiments, CO_2 was injected through the ports on one end of the sample. The coal specimen was 54 mm in diameter and

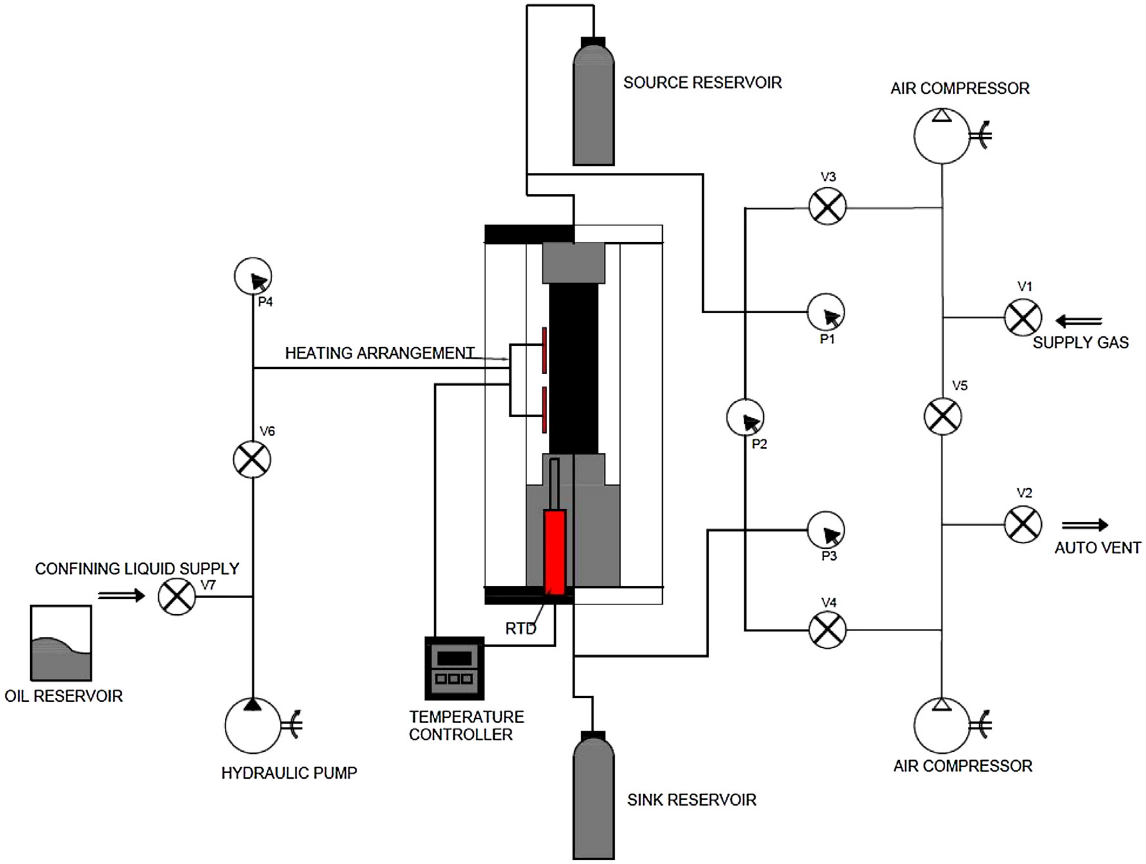

Fig. 1. Simple schematic of the triaxial permeameter setup.