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Full Length Article

CO_2-wettability of low to high rank coal seams: Implications for carbon sequestration and enhanced methane recovery

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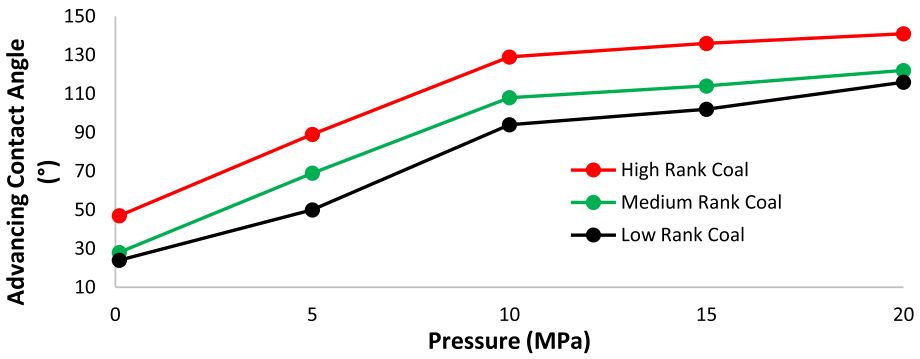
HIGHLIGHTS

- Effect of pressure, temperature and salinity on CO_2-wettability of coals.
- Effect of coal rank on CO_2-wettability of coals.
- Implications of measured data.

GRAPHICAL ABSTRACT



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ABSTRACT

Coal seams offer tremendous potential for carbon geo-sequestration with the dual benefit of enhanced methane recovery. In this context, it is essential to characterize the wettability of the coal–CO_2–water system as it significantly impacts CO_2 storage capacity and methane recovery efficiency. Technically, wettability is influenced by reservoir pressure, coal seam temperature, water salinity and coal rank. Thus a comprehensive investigation of the impact of the aforementioned parameters on CO_2-wettability is crucial in terms of storage site selection and predicting the injectivity behaviour and associated fluid dynamics. To accomplish this, we measured advancing and receding water contact angles using the pendent drop tilted plate technique for coals of low, medium and high ranks as a function of pressure, temperature and salinity and systematically investigated the associated trends. We found that high rank coals are strongly CO_2-wet, medium rank coals are weakly CO_2-wet, and low rank coals are intermediate-wet at typical storage conditions. Further, we found that CO_2-wettability of coal increased with pressure and salinity and decreased with temperature irrespective of coal rank. We conclude that at a given reservoir pressure, high rank coal seams existing at low temperature are potentially more efficient with respect to CO_2-storage and enhanced methane recovery due to increased CO_2-wettability and thus increased adsorption trapping.

1. Introduction

Carbon capture and storage (CCS) is the most promising approach to mitigate anthropogenic CO_2 emissions and thus ensure a cleaner environment [1–5]. The storage of CO_2 in depleted

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oil and gas reservoirs [6–8] or deep saline aquifers [9–11] allows trapping of enormous volumes over a long period of time. Another option is injection of CO_2 into coal seams [12–15] with the dual benefit of enhanced coal bed methane recovery [16–18]. In conventional reservoirs, CO_2 is held trapped by means of four mechanisms which are structural trapping [19–22], capillary or residual trapping [8,23–25], dissolution trapping [26,27] and mineral trapping [28]. In coal seams, however, the dominant storage mechanism is adsorption trapping of CO_2 onto the coal matrix [12,29,30]. Typically the adsorption capacity of CO_2 is higher than that of methane, depending on coal rank [31–33]; consequently, CO_2 displaces methane toward the production well and itself gets sorbed within the micropores of the coal seam and remains trapped. CO_2 storage by means of adsorption in coal seams is strongly influenced by wettability of the CO_2–water–coal system [12,34], which in turn is generally a function of reservoir pressure [34–37], temperature and salinity. Moreover, in coal seams wettability is also a function of coal rank, vitrinite reflectance, fixed carbon and ash content [35,38]. Therefore, it is essential to describe CO_2-wettability of coals of varying ranks, and how reservoir conditions (pressure, temperature and brine salinity) influences this wettability.

In this context, several studies reported CO_2-wettability of coals at ambient conditions [38–41], but only a limited amount of literature data for the more relevant higher pressures have been reported [34–37,42,43]. Table 1 presents a summary of the major experimental variables considered in previous studies, and this work.

Therefore there is a clear lack of data available on CO_2-wettability of coals as a function of coal rank, coal formation pressure, and particularly temperature and salinity (cp. Table 1). Thus there exists a gap in terms of proper understanding of CO_2-wettability of coal seams of different ranks at reservoir conditions. Moreover, although it is well established that coal seams offer enormous potential for enhanced methane recovery and CO_2 sequestration, yet certain important questions need to be addressed which are: (1) Which type of coal (low rank, medium rank, or high rank) are most suitable for CO_2 storage and enhanced coalbed methane recovery under the prevailing geothermal and reservoir pressure conditions? (2) Is the suitability of CO_2 sequestration in coal seams of a particular rank valid for a wide range of reservoir pressures, temperatures and salinity conditions? (3) What mechanisms are responsible for long term CO_2-storage in coals? To answer these questions and to generally improve the characterization of CO_2-wettability of coals, we experimentally measured water advancing and receding contact angles on three coal samples as a function of coal rank (low, medium and high ranks), vitrinite reflectance and fixed carbon at different CO_2 pressures (0.1–20 MPa), temperatures (ranging from 308 K to 343 K), and brine salinities (0–10 wt% NaCl) using the pendent drop technique. The results of the study lead to a broad characterization of CO_2-wettability of coals and thus help optimize CO_2-storage and enhanced coal bed methane recovery operations. Our results indicate that CO_2-wettability of coals is strongly influenced by coal rank such that the high rank coals are more CO_2-wet and low rank coals are least CO_2-wet at a given reservoir pressure, temperature and salinity.

2. Materials and methods

2.1. Coal samples

Three coal samples [high rank (semi anthracite; from Hazelton, Pennsylvania, USA), medium rank (medium volatile bituminous; from Morgantown, West Virginia, USA), and low rank (lignite; from North Dakota, USA; Table 2)] were used in this research. The samples were cut to cuboid shape ($\sim$1 cm $\times$ 1 cm $\times$ 0.5 cm) and the surface roughness of each substrate was measured with an atomic force microscope (AFM instrument model DSE 95-200); note that typically surface roughness significantly affects contact angle measurements [44,45]. The RMS surface roughness of the specific coal substrates used were 840 nm, 880 nm and 280 nm for high, medium and low rank coals respectively.

2.2. Petrology, ultimate and proximate analysis

The results of the proximate, ultimate and petrological analysis and the internal properties (density and volume) of the coal samples are listed in Table 2. Note that coal samples of different rank differ mainly in volatile matter, moisture, fixed carbon and vitrinite reflectance [46,47]. Petrology was analysed in accordance with Australian Standard AS2856 and ISO7404; proximate analysis were conducted using AS1038.3, ISO11722 and ASTM D3172-07a, and ultimate analysis were performed using AS1038.6 and ISO 609.

2.3. Fluids

99.9 wt% CO_2 (from BOC, gas code – 082), de-ionized water (Conductivity: 0.02 mS/cm), and 5 wt% and 10 wt% NaCl brine (NaCl Source: Scharlab s.l., Spain, Purity: $\geqslant$0.995 mass%) were used in the study. Acetone (99.9 wt%) was used to wash the coal samples.

2.4. Contact angle measurements

CO_2-brine wettability was measured using the pendent drop tilted plate technique [48]. The experimental setup is shown in Fig. 1; it consists of a high pressure cell (which holds the sample on a tilted plate), a CO_2 cylinder, two high precision syringe pumps (Teledyne D-500, pressure accuracy of 0.1%FS) for water and CO_2 and a video camera. Prior to each experiment, the coal substrates were washed with acetone and then cleaned in air plasma (Diemer Yocto instrument) for 2 min to ensure that no organic contaminants are deposited on the sample, which would introduce a significant bias [49]. We limited plasma treatment to 2 min to avoid changes in surface chemistry which may be induced by the plasma. We, however, note that Shojai Kaveh et al. [43], used wet polish with a series of abrasive papers with a grid ranging from 60 to 1200, followed by polishing with 0.5 μm abrasive alumina

Table 1
CO_2-wettability of coals: Summary of experimental conditions used.

Reference	Pressure (MPa)	Temperature (K)	Salinity	Coal type	Overall coal rank
Chi et al. [42]	Up to 6.2	298	DI water	Not mentioned	Not mentioned
Siemons et al. [37]	Up to 14	318	DI water	Anthracite	High
Sakurovs and Lavrencic [36]	Up to 15	313	DI water	Bituminous	Medium
Shojai Kaveh et al. [43]	Up to 16	318	DI water	High volatile bituminous	Medium
Shojai Kaveh et al. [35]	Up to 16	318	DI water	Semi anthracite, High volatile bituminous	High and medium
Saghafi et al. [34]	Up to 6	295	DI water	Medium volatile bituminous	Medium
This study	Up to 20	308, 323 and 343	0–10 wt% NaCl	Semi-anthracite, medium volatile bituminous, lignite	High, medium and low