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The Effect of Coal Rank on the Wettability Behavior of Wet Coal System with Injection of Carbon Dioxide and Flue Gas

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SUMMARY

The injection of carbon dioxide (CO₂) or flue gas into coal layers enhances the coal bed methane production (ECBM) and is also an option for CO₂-storage. The success of this combined process depends strongly on the wetting behavior of the coal, which is a function of coal rank, ash content, pressure, temperature and composition of the gas.

Two coal samples have been used for this study representing different ranks: hvBb and semi-anthracite rank. The wettability behaviour of the wet coal samples upon injection of synthetic flue gas and pure CO₂ was investigated in a modified pendant drop cell at a constant temperature of 318 K and pressures varying between 0.1- 16 MPa.

For the hvBb sample, the wettability of the coal surface changed from intermediate-wet to CO₂-wet at pressures above 8.5 MPa. When injecting synthetic flue gas, only a change from water-wet to intermediate-wet was observed. For the semi-anthracite rank Selar Cornish sample and CO₂ injection this alteration was observed at about 5.3 MPa. Experimental results with synthetic flue gas revealed that the wettability of Selar Cornish coal is intermediate wet at all pressures and the contact angle only slightly increases with increasing pressure.

Introduction

Carbon dioxide (CO₂) emission from burning fossil fuels has been identified as the major contributor to the increase in atmospheric CO₂ levels. Sequestration is regarded as a remedy for carbon dioxide emissions including strategies such as CO₂ injection into deep saline aquifers, depleted oil and gas reservoirs, and unmineable coal seams. Additionally, the injection of CO₂ or flue gas, into coal layers enhances the coal bed methane production (ECBM).

Power plants are one of the major sources for CO₂ emission. The emitted flue gas consists mainly of CO₂ and nitrogen and traces of NO_x and SO_x. Commonly, CO₂ is separated from the flue gases prior to its injection for sequestration. The latter makes the whole process less efficient. Because of the chromatographic behavior of coal, direct injection of flue gas eliminates the necessity of flue gas purification prior to the injection into the coal bed. The success of this combined process depends strongly on the wetting behavior of the coal, which is a function of coal rank, ash content, heterogeneity of the coal surface, pressure, temperature and composition of the gas (Shojai Kaveh et al., 2011).

CO₂ sequestration and coal bed methane (CBM) production are largely controlled by the interactions among CO₂, the reservoir fluid, the coal matrix and the ash. The wettability of coal is however believed to change with the coal rank (chemical composition) (Klassen, 1963; Laskowski, 1994; Brady and Gauger, 1940; Sun, 1954), mineral matter content (He and Laskowski, 1992; Aplan, 1993), moisture level (Elyashevitch, 1941), porosity (Drelich et al., 2000; Keller, 1987), degree of oxidation (Laskowski, 1994; Sun, 1954; Gayle et al., 1965), and the pressure and temperature of the reservoir and the reservoir fluids. Thus these parameters are essential determining the success of the process (Gutierrez-Rodriguez et al., 1984; Orumwense, 2001).

For a hydrophobic coal it is expected that the small cleats are filled with gas, leading to a much faster transport of the injected gas to the coal particulates than for a hydrophilic coal. For hydrophilic coal the small cleats are filled with water slowing down the mass transfer of the gas from the main cleat system to the particulates (Shojai Kaveh et al., 2011). Arnold and Aplan (1989) found the biggest difference in the wetting behavior between hydrophobic organic macerals and hydrophilic inorganic mineral inclusions. Therefore, the wettability of the coal is an important factor for the evaluation of ECBM applications.

The wettability of dry and wet coal systems has been experimentally determined using contact angle measurements (Keller, 1987). However, only a few experimental data on the wettability at high pressures and elevated temperatures exist, particularly in the presence of flue gas (Shojai Kaveh et al., 2011; Siemons, 2007; Chi et al., 1987). From the data of Siemons it can be concluded that for high-rank coal wetting alteration from water-wet to CO₂-wet occurs at a pressure as low as 0.27 MPa. For a medium-rank coal this wetting alteration was observed for pressures above 8.7 MPa. It needs to be mentioned that during these studies the water was not fully saturated with CO₂ so that at each experimental datum the composition of the aqueous phase was different. In the work presented here, the experiments were repeated but now with water fully saturated with CO₂ to exclude the effect of changing composition of the aqueous phase and dissolution effects.

Two coal samples have been used for this study representing different ranks, a Warndt Luisenthal (high volatile bituminous rank- hvBb) and a Selar Cornish (semi-anthracite rank) coal. The wettability behaviour of the wet coal samples upon injection of a synthetic flue gas (20% CO₂/ 80% N₂) and pure CO₂ was investigated by means of contact angle measurements. The experiments were done in a modified pendant drop cell at a constant temperature of 318 K and pressures varying between 0.1- 16 MPa.

Experiments

The experiments were conducted in a modified pendant drop (PD) cell (Shojai Kaveh et al., 2011), which was adapted to allow captive-bubble contact-angle measurements on real coal surfaces. The determination of the contact angle is based on visual observation i.e. image capture and the

subsequent image analysis. A schematic drawing of the experimental set-up is given in Figure 1. The cell can be used for pressures up to 60 MPa. Both sides of the cell consist of a steel cap with glass windows. These glass windows allow the visual observation of the bubble and the coal substrate inside the pendant drop cell.

Figure 1 Schematic diagram of experimental set-up (Pendant drop cell)

The Warndt Luisenthal (WL) and the Selar Cornish (SC) coal samples were mined in the intramountain Saar basin in Western Germany and Selar colliery in South Wales Coalfield, respectively. CO₂ has been purchased from Linde Gas Benelux and has a purity of 99.7 vol%. As a synthetic flue gas a mixture of 20 vol% CO₂ and 80 vol% N₂ from Linde Gas Benelux was used. The results of the ultimate and proximate analysis of the coal samples are given in Table 1.

Table 1 Proximate, ultimate analysis and coal petrology of used coal samples

Sample	Warndt Luisenthal	Selar Cornish
Rank	<i>hVbB</i>	<i>Semi anthracite</i>
<i>R_{max}</i> (%)	0.71	2.41
<i>Vitrinite</i> (%)	74.4	73.6
<i>Liptinite</i> (%)	15.6	0
<i>Inertinite</i> (%)	9	24.6
<i>Minerals</i> (%)	1.0	1.8
<i>Volatile Matter (w.f.)</i> (%)	40.5	10.4
<i>Carbon (wt%)</i>	81.3	85.7
<i>Hydrogen</i> (%)	5.58	3.36
<i>Nitrogen</i> (%)	1.88	1.56
<i>Sulfur</i> (%)	0.69	0.68
<i>Oxygen</i> (%)	5.47	5.58
<i>Ash (w.f.)</i> [%]	2.77	3.94-5.5
<i>Fixed Carbon (d.a.f.)</i> [%]	58.36	89.27

Image analysis

Visualization of the bubbles and the contact angle is possible because of the use of an endoscope and the windows at either side of the pendant drop cell. A Nikon D90 Spiegel reflex digital camera with a resolution of 12.3 MP was used to take pictures from the pending gas bubbles. Several pictures were taken from each bubble under the same conditions, in order to check the reproducibility of the determined contact angles. Contact angles were determined using an improved in-house MATLAB routine (Shojai Kaveh et al., 2011).

Results and Conclusions

For the hvBb sample, when injecting pure CO₂ the wettability of the coal surface changed from intermediate-wet to CO₂-wet at 8.5 MPa. This means that the coal surface becomes hydrophobic at pressures above 8.5 MPa at 318 K. This would mean that for CO₂ storage in hvBb coal the injection pressure of CO₂ has to overcome a pressure threshold of 8.5 MPa to wet the surface and thus to enhance the storage capacities. When injecting synthetic flue gas only a change from water-wet to intermediate-wet was observed for pressures above 10.5 MPa (Shojai Kaveh et al., 2011). For the semi-anthracite rank Selar Cornish sample it was found that the wettability alteration with pure CO₂ injection occurs at about 5.3 MPa and that the contact angle increases with increasing pressure. Experimental results with synthetic flue gas revealed that the wettability of Selar Cornish coal is intermediate wet at all pressures and the contact angle only slightly increases with increasing pressure. The results are summarized in Figure 2.

In general, on both coal samples the contact angles of flue gas bubbles are smaller than of the CO₂ bubbles. The hydrophobicity of the coal samples increases with coal rank and pressure. When injecting CO₂ the wettability alteration occurs at lower pressure for the semi-anthracite sample than for the hvBb sample. In addition, when injecting flue gas, the contact angles on the semi-anthracite Selar Cornish sample are higher than for the Warndt Luisenthal coal, thus is more hydrophobic. Additionally, for injection of either CO₂ or flue gas the pressure shows less effect on the contact angles for the semi-anthracite sample than for the Warndt Luisenthal. Thus for these samples the process cannot be controlled by the pressure.

Figure 2 Contact angle as function of the pressures for the different investigated systems. The abbreviations SC and WL are used to describe the experimental data for the Selar Cornish and the Warndt Luisenthal coal samples: (a) CO₂ injection, (b) synthetic flue gas injection, (c) CO₂ and synthetic flue gas injection on Selar Cornish sample, (d) CO₂ and synthetic flue gas injection on Warndt Luisenthal sample.

The mechanisms causing the observed wettability alterations have not yet been identified. Possible causes are the sorption of the CO₂ on the coal, change of aggregation state of CO₂ from gaseous to supercritical state, or the increased CO₂ solubility in water at higher pressures. Interestingly, the maximum sorption of CO₂ on wet WL and SC coal samples was found in the same pressure range as the wettability alteration (Siemons, 2007).

The alteration of the coal wettability from water-wet to intermediate or gas-wet is of particular interest as it changes the efficiency of the CO₂ storage. It is expected that the behavior found in this study is generally applicable to coals with the same rank and with similar compositions, not considering the ash content.

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