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GAS WETTABILITY OF COAL AND IMPLICATIONS FOR GAS DESORPTION AND DRAINAGE

Abouna Saghafi¹, Kaydy Pinetown² and Hoda Javanmard¹

ABSTRACT: A key parameter affecting the flow of gas in coal is the wetting potential of gas, in comparison to water, to spread over the wall of coal micropores and microfissures. Wettability is quantified in terms of the contact angle of the fluid interface with the solid surface. A fluid with a small angle of contact would spread over the pore walls and eventually displace the non-wetting fluid. Depending on the nature of the coal, gas type and environmental conditions in coal reservoirs, either water or the gas phase could wet coal more strongly. Furthermore, in mixed gas conditions, one gas may be more strongly attached to coal than the other gases. In water-saturated coal, gas desorption in small pores -where most adsorbed gas is stored - can be totally inhibited by water if it is a strong wetting phase. Reducing the hydraulic head (drawdown to achieve the gas desorption pressure) should allow desorption of gas in larger fractures, whereas in small pores, gas desorption could be inhibited by capillary pressure due to the effect of interfacial tension and gas-wetting properties of coal. In this study, we built a new system to quantify the wettability of coal by gas. The contact angle of the water-gas interface with the coal surface inside the gas phase was measured using a captive gas bubble technique. The contact angles of CH₄ and CO₂ bubbles in water with a coal from the Sydney Basin were measured at different gas-water pressures of up to 15 MPa for CH₄ and 6.1 MPa for CO₂. The results show that as gas bubbles dissolve in water, the contact angle of the bubble with the coal surface reduces. The contact angle values were smaller for CO₂ gas than CH₄, and in general, the contact angle value decreases as gas-water pressure increases.

INTRODUCTION

In coal-bearing sedimentary sequences, the fluid phases consist of gas and water. For effective coal seam gas drainage in deep coal mines or gas recovery, removal of formation water eases the desorption of gas from the coal matrix and its flow into the larger cleat and crack system. The resistance to gas desorption by the medium could be partly due to coal being preferentially wet by water rather than by gas. The release of gas from coal can be inhibited by a water-wet coal, which does not allow the diffusion of gas to the surface of the cracks and desorption from coal matrix. This is experienced in gas adsorption experiments using moist coals or gas recovery operations, which require discharging sufficient volumes of water from the borehole before gas flow is established. In a water-wet coal, water is generally in contact with the surface of pores, and gas movement and desorption from the coal matrix is resisted by molecules of water on the pore wall. In a gas-wet coal, gas molecules – not water molecules – are in contact with the pore wall, and gas molecules readily spread over the surface of the pore walls. Hence, the movement of gas in and out of the coal matrix and along the connecting microfissures into macrofissures and large cracks is largely facilitated. Since wettability plays such an important role in the migration and drainage of gas from coal, its quantification and relative ranking with respect to gas and coal types are vital to optimise gas recovery and drainage from coal seams.

Quantification of coal wettability by water and other liquids has been an area of significant interest in the coal cleaning and flotation industry. Numerous studies have aimed to evaluate coal floatability (see for example Keller, 1987; Arnold and Aplan, 1989; Drelich, *et al.*, 2000; Drelich, 2001; Gosiewska, *et al.*, 2002). These authors studied the effect of mineral matter on the water wettability of coal.

To date, limited research has been undertaken to study the relative wettability of coal by gas and water (Siemons, *et al.*, 2006; Sakurovs and Lavrencic, 2011; Shojai Kaveh, *et al.*, 2011). All of these studies have been in the context of CO₂ sequestration in coal seams and dealt with CO₂ wettability of coal. As far as is known no work has been published in the context of CH₄ drainage and flow in coal seams, and our study is the first on CH₄ wettability of coal. The data can be used to evaluate the effect of wettability properties on gas drainage efficiency in water-saturated coals.

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After presenting some background to surface interaction phenomena, this paper discusses the results of recent experimental research on gas wetting of a coal collected from a Sydney Basin coal mine.

WETTABILITY

Wettability is measured in terms of the contact angle of the interface between the two immiscible phases (gas and water) against the surface of the solid phase (pore walls in coal capillaries). In this work, contact angle is measured in the phase for which the wettability is to be estimated (gas phase). Using this convention, a contact angle of less than 90° indicates that the targeted phase (gas) is the wetting phase, and a contact angle larger than 90° indicates that the other phase (water) is the wetting phase. Figure 1 shows a pore in coal that contains water and gas. In this example, water is the wetting phase, because the contact angle of gas phase with solid surface (θ) is larger than 90° . This means that water is attached more strongly to the pore walls and has low mobility, and that the interfacial forces draw water into small pores and inhibit the non-wetting phase (gas) from penetrating or escaping from these pores

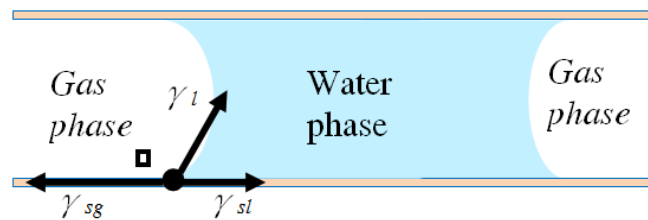


Figure 1 - Gas and water in a pore: the gas wettability of the solid (coal) is measured in terms of the contact angle of the gas–water interface with the solid surface inside the gas phase (θ)

The magnitude of the contact angle is a function of the interfacial tension, which is the energy required to separate a unit area of the interface between two phases; it is expressed in terms of energy per unit area or force per unit length (J/m^2 or N/m). An equilibrium of the cohesive and adhesive forces at the line of contact between the three phases (solid, liquid and gas) is reached when the interface between the liquid and gas forms a certain angle θ (contact angle) with the solid. Assuming equilibrium of forces on the line of the intersection of the three phases (gas–water interface and solid in Figure 1) it can be shown that the magnitude of the contact angle between the gas–water interface and the solid should follow the Young (1850) and Laplace (1806) equation:

$$\gamma_{lg} \cos \theta = \gamma_{sl} - \gamma_{sg} \quad (1)$$

where the subscripts *s*, *l* and *g* denote solid, liquid and gas. For example, γ_{sg} is the interfacial tension between the solid and gas phases.

Since the interfacial tensions are properties of materials, the contact angle is also considered as a material property of the fluid and solid system independent of any particular configuration. Note that if $\gamma_{sg} > \gamma_{sl}$, then $\theta > 90^\circ$ and water is the wetting phase (Figure 1). The magnitude of the interfacial tension is a function of the difference between the cohesive and adhesive forces acting on the interface of the two phases. Strong adhesive forces produce contact angles of less than 90° , which is a characteristic of the wetting phase. Similarly, strong cohesive forces produce contact angles larger than 90° (non-wetting phase).

The curvature of interface between the two fluid phases (gas and water) produces a discontinuity in fluid pressure across the interface. For the non-wetting phase (gas in Figure 1) to desorb and move through the capillary, the free gas pressure needs to be more than this pressure difference. The pressure difference across the interface of gas and water can be estimated using the geometrical form of the Young–Laplace Equation. If the effect of gravity is neglected, the difference in fluid pressure across the interface is (see for example Bear, 1988; Butt, *et al.*, 2006):

$$\delta p = p_{nw} - p_w = \gamma \left(\frac{1}{R_1} + \frac{1}{R_2} \right) = \gamma C \quad (2)$$

where p_{nw} and p_w are fluid pressures in the non-wetting and wetting phases, γ is the interfacial tension between the two fluid phases, and R_1 and R_2 are the two principal radii of curvature on the fluids'

interface. The principal radii are determined in two orthogonal directions at the point of interest on the surface. The quantity $C = 1/R_1 + 1/R_2$ is called the mean curvature. It is a geometrical property that determines the shape of the interface and does not depend on the orientation.

Measurement of wettability

Evaluation of the wettability of a solid by a fluid has been the subject of research and applications in various disciplines, and various methods have been developed and used (see for example Neumann and Good, 1979). The two main methods for directly measuring a contact angle from the profile of a fluid drop or bubble on a flat solid surface are the sessile drop and captive gas bubble techniques.

In the sessile drop method, the solid sample is ground to form two horizontal surfaces and then mounted on a horizontal stage. A drop of liquid for which the contact angle against the solid surface is required is placed on the solid surface using a device (such as a pipette with small tip) to form the drop. The solid sample and fluid drop may be placed in a water-filled cell. The profile of the fluid drop on the solid surface and the contact angle are evaluated using a telescope and a protractor. A camera may also be used to take frequent pictures of the drop profile. The image data are logged and stored digitally. Image analysis software is then used to calculate the drop geometry and contact angle.

In the captive gas bubble technique, the solid sample is placed at the top of a water cell with the sample facing down. A gas bubble is introduced from the bottom of the water cell. The gas bubble rises and comes into contact with the solid surface. As in the sessile drop technique, measurement of contact angle is undertaken from the profile of the bubble adhered to the surface. Similarly, a camera is used to record images of the profile and calculate the contact angle from the bubble profile using an image processing method.

GAS FLOW IN COAL AND WETTABILITY

Coal seam reservoirs are generally water saturated, which largely restricts the flow of gas. The retaining effect of water is significantly larger in small pores, where most adsorbed gas is stored. A main indicator of the resistance to gas movement is the relative wettability of coal by water and gas. Hence, quantifying the wettability of coal in terms of the contact angle of the gas–water interface with the coal surface will improve understanding of gas flow and drainage in coal seams.

The δp in Eq (2) can be seen as a pressure barrier for the movement of the non-wetting fluid through the medium. In larger cracks and fractures, the mean curvature (C in Eq 2) is small and hence δp remains small. However, in smaller micropores, the curvature of the fluid interface can be significant, and very large δp can develop across the interface, preventing the movement of gas through the coal. If the pores are modelled as tubular capillaries, it can be shown that the curvature of the fluid interface in the tubular pore is:

$$C = \frac{-4 \cos \theta}{d} \quad (3)$$

where d is the diameter of the tubular pore. Substituting C from Eq (3) in Eq (1) yields:

$$\delta p = \frac{-4\gamma \cos \theta}{d} \quad (4)$$

Therefore, if gas is the non-wetting phase in coal, then to allow gas desorption from pore walls, it is not enough to reduce water hydrostatic pressure to below the gas desorption pressure by pumping water out of gas drainage boreholes. Instead, the total water pressure, including the capillary pressure (δp), should be reduced. In other words, to allow desorption and flow of gas bubbles from micropores into larger cracks and cleats, the desorption pressure must be larger than the sum of the hydrostatic and capillary pressure:

$$p_g > p_w + \delta p \quad (5)$$

where p_g is gas desorption pressure and p_w is the hydrostatic pressure (hydraulic head). Note that when d is large enough (e.g. in fractures and larger-aperture cleats) δp becomes quite small relative to the hydrostatic pressure, and gas desorbs from coal as soon as the hydrostatic pressure falls below the gas

desorption pressure. Therefore, for larger fractures, reducing hydraulic head alone can allow gas desorption. This is usually achieved by pumping out a certain volume of water from gas drainage boreholes.

Numeric example of the effect of wettability on gas desorption and drainage

To illustrate the effect of wettability (in terms of contact angle), a parametric example is presented.

Assume a water-saturated coal seam at depth of 500 m, and assume that gas content is $10 \text{ m}^3/\text{t}$ and gas adsorption capacity of coal in terms of Langmuir parameters is $V_L = 27 \text{ m}^3/\text{t}$ and $P_L = 3.1 \text{ MPa}$. Based on these data, the hydrostatic pressure at this depth would be about 4.9 MPa, whereas the gas desorption pressure is about 1.8 MPa. Therefore, to allow the onset of gas desorption, water (hydrostatic) pressure should be reduced from 4.9 MPa to 1.8 MPa (3.1 MPa of water drawdown). This reduction in water pressure would allow gas desorption to take place in larger fractures and cleats (e.g. millimetre-aperture cracks). For small pores (micrometre-sized and smaller), where most adsorbed gas is stored, the effect of capillary pressure should be taken into account, because it can prevent gas desorption and migration toward drainage pathways. For example, for pores of $1 \mu\text{m}$ diameter, assuming a contact angle of 170° , the capillary pressure is 275 kPa. However, for pores of $0.1 \mu\text{m}$, the capillary pressure will be much larger at 2.8 MPa. For very small pores (nanometer-sized) the capillary pressure might become a full barrier to desorption and movement of gas in pore voids. For a pore of 1 nm diameter, capillary pressure is 27.6 MPa (contact angle of 170°).

Figure 2 plots the capillary pressure developed in tubular pores of 1.0 , 0.1 and $0.01 \mu\text{m}$ diameter as a function of contact angle of the gas–water interface with pore walls.

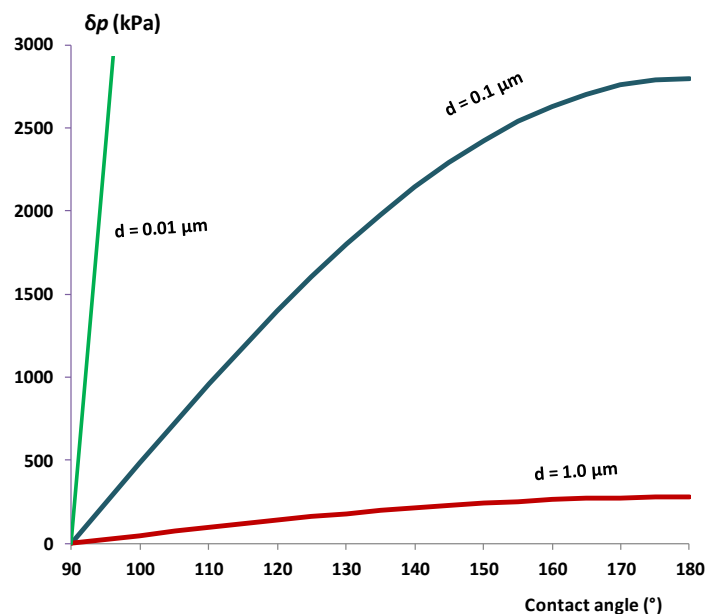


Figure 2 - Capillary pressure (δp) developed in tubular pores of coal, the magnitude of this pressure depends on pore diameter and the contact angle of the gas–water interface with pore walls (water is the wetting phase)

MEASUREMENT METHOD USED IN THIS STUDY

The CSIRO system for measurement of gas wettability of coal is a variant of the captive gas bubble technique. In this system, a coal disc is prepared from a lump or core coal sample. The disc is then fixed to the end of a sample holder in the form of a rod, which is introduced into a high-pressure water-filled cell with the coal surface facing down. Pressurised gas is slowly released from a nozzle below the coal sample to form a gas bubble, which rises and comes into contact with the coal surface. The formation of bubbles at very high pressures and fixation of bubbles on the coal surface is a delicate operation, and requires the presence of an experienced operator and sufficient time (hours) to obtain suitable results. The gas pressure is set using an ISCO-260D syringe pump, while water is pressurised using a

Shimadzu LC-9A water pump. Distilled water (pH 5 to 6) is used to fill and pressurise the cell. The water pressure is set slightly below the gas pressure so that gas bubbles can form.

A camera is used to capture images at a pre-determined rate. These settings are adjusted in the software prior to the start of measurement. The image data are then processed using an image analysis code to deliver the right and left contact angle values as a function of time. Figure 3 shows a photograph of a gas bubble in contact with the coal surface where the contact angle is larger than 90° .

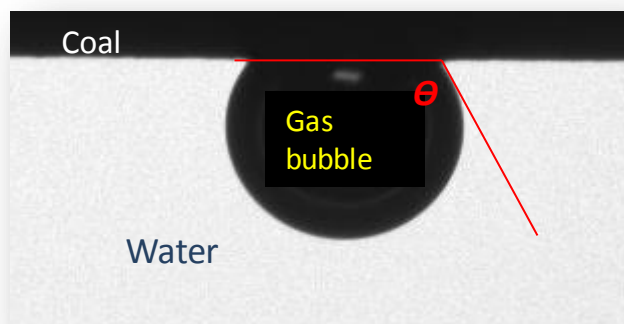


Figure 3 - Image of a gas bubble in contact with a solid coal surface. The coal and gas bubble are immersed in pressurised water; in this instance, gas is the non-wetting phase ($\theta > 90^\circ$)

Experimental parameters

The image processing software reports the contact angle of the bubble (Θ) with the solid surface on the right and the left side of the image frame. The software matches the boundary of bubble against the solution of the Young–Laplace equation to yield the best fit. The values of the right and left contact angles are then calculated based on the best fit. Other values reported by the software include the length of contact (L), which is the diameter of the circle the bubble makes where it intersects the surface of the coal sample (the line of contact between the three phases), and height (H) of the bubble. The software also calculates the bubble's volume and total surface area from the fitted curve.

RESULTS OF MEASUREMENT OF GAS–WATER CONTACT ANGLE WITH COAL

Coal lump samples were obtained from a coal mine in the Sydney Basin. The studied coal was of medium volatile rank with a vitrinite reflectance of $\sim 1.3\%$ and a dry-ash-free volatile matter content of $\sim 24\%$. Suitable samples were prepared by drilling to prepare discs with a diameter of 16 mm and a thickness of 10 mm.

Two sets of measurements of contact angles were undertaken using pure CH_4 and pure CO_2 gases. All measurements were conducted at a constant temperature of 22°C .

Contact angle of CH_4 with coal

For CH_4 gas, measurements were undertaken at gas–water pressures of 1.8, 5.0, 7.7, 9.6, 12.9 and 15.2 MPa.

The contact angle and the length and height of the bubble were monitored for 1–2 h following the formation of the bubble and its fixation on the coal surface. Figure 4 shows the evolution of the contact angle of CH_4 bubbles with the coal surface at various gas–water pressures. The contact evolution follows a similar pattern for all pressures; at the start of the experiment, the contact angle is larger, and then reduces to reach a relatively constant value.

Further analysis shows that the evolution of the contact angle is influenced by the size of the gas bubbles. As the experiment progresses, the gas bubble slowly dissolves in water. The reduction in bubble size is, however, constrained by resistance from the three-phase contact line to shrink and gas–coal contact area to reduce. Hence, the reduction of gas bubble volume is manifested by a larger reduction in the height compared with the area of contact of gas bubble with coal, leading to smaller contact angle. As the bubble size reduces, the contact angle also reduces, to reach a minimal value after which it stays relatively constant. The pattern of evolution of the contact angle suggests that the

contact angle curve can be characterised by a maximum value (θ_{max}) at the start of the experiment and a minimum value (θ_{min}) at the plateau section of the curve.

Overall, θ_{max} varied between 130° to 140° and θ_{min} between 78° and 96°. The contact angle values were in general smaller under higher pressures.

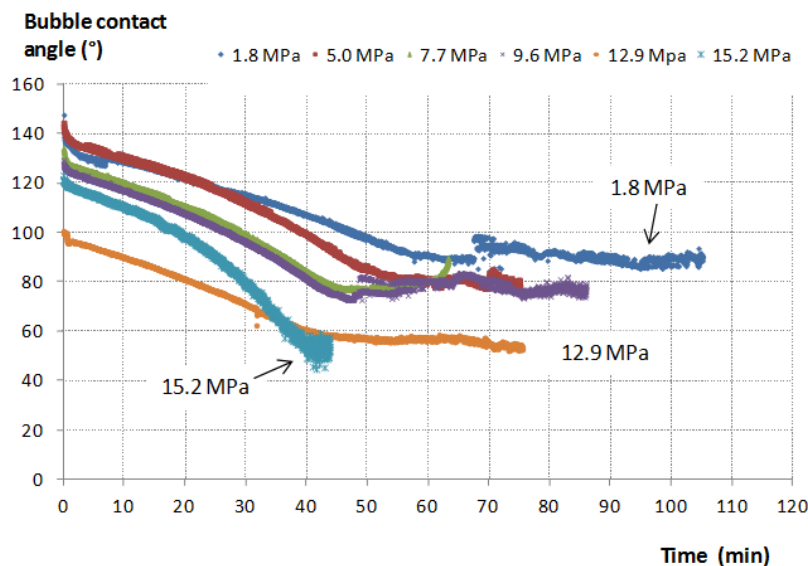


Figure 4 - Evolution of contact angle of CH₄ gas bubbles with coal at various gas–water pressures

Contact angle of CO₂ with coal

For CO₂ gas, contact angle measurements were undertaken at gas–water pressures of 1.8, 5.2 and 6.1 MPa. To maintain the sub-critical conditions for CO₂ gas, larger pressures were not applied.

CO₂ gas bubbles were rapidly dissolved in water, and hence measurements could be conducted only for a few minutes as the bubbles' volume quickly reached the system's limit of detection for image analysis (less than 0.2 μL). Figure 5 plots the evolution of the contact angle. A similar pattern to that seen with CH₄ occurs for CO₂. The contact angle with coal reduces as the CO₂ bubbles dissolve in water and the size of a bubble reduces.

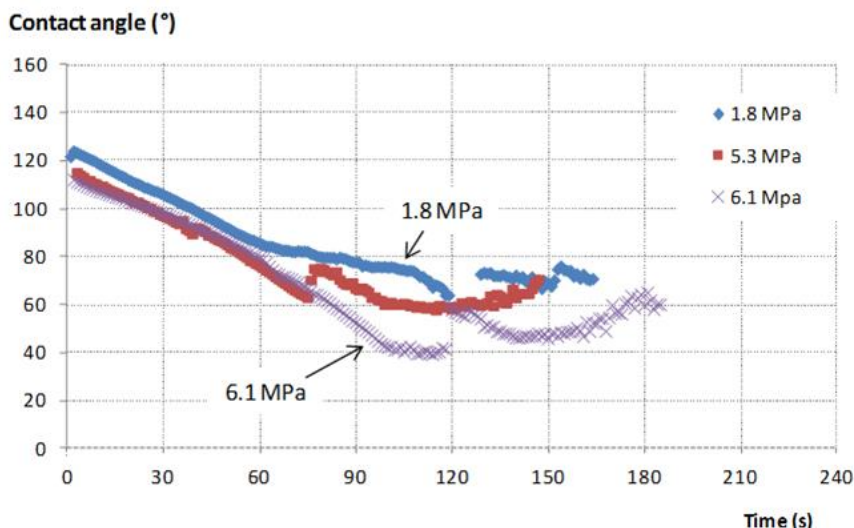


Figure5 - Evolution of contact angle of CO₂ bubble with coal surface at various gas–water pressures

For the three CO₂ experiments, θ_{max} varied between 112° to 122° and θ_{min} between 40° and 68°. The contact angle values were smaller for the higher pressures.

CONCLUSIONS

We have developed a new system for the quantification of gas wettability of coal in terms of the contact angle of gas with the coal surface in the presence of water. The contact angles of CH₄ and CO₂ gas bubbles with a coal from the Sydney Basin were measured at various gas–water pressures and a constant temperature of 22°C.

The data from this study show that contact angle evolution follows a specific pattern; the angle reduces from a maximum value at the start of the experiment to reach a relatively minimum value at the plateau section of the curve. Hence, the contact angle pattern can be characterised by a maximum value (θ_{max}) at the start of the experiment and a minimum (θ_{min}) at the plateau section of the curve.

The reduction in bubble size affects the height of the bubble more strongly than it affects the area of contact between the bubble and the coal surface. This results in an apparent reduction in contact angle as the size of the bubble reduces. However, after a certain period of time, the height of gas bubbles and the contact surface reduce simultaneously, resulting in a relatively constant value of the contact angle (in the plateau section of the curve).

For both gases, the contact angle reduces as the applied gas–water pressure is increased. Moreover, the contact angle is generally higher for CH₄ than for CO₂, indicating that coal is more CO₂-wet than CH₄-wet.

Contact angle data can assist in evaluating the effect of gas wettability of a particular coal on gas desorption and drainage in various pore sizes of that coal. For calculation of capillary pressures, it is recommended to use θ_{max} values to ensure that these pressures are not underestimated. However, note that the data generated in this study is specific to the coal sample measured and should not be used for other coals. We did not investigate the effect of coal rank, coal composition, ash yield or nature of minerals on gas wettability. To characterise a given coal, specific experiments for that coal should be undertaken.

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