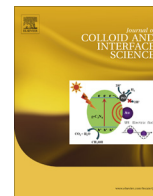




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On wettability of shale rocks

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H I G H L I G H T S

- Increase in pressure and temperature increases the contact angle of brine/shale system.
- Bivalent ions exhibit higher contact angle compared to monovalent ions.
- Effect of pressure and temperature on contact angle is more pronounced at high ionic concentrations.
- A physical model was developed to get insight into variation of contact angle.

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The low recovery of hydraulic fracturing fluid in unconventional shale reservoirs has been in the centre of attention from both technical and environmental perspectives in the last decade. One explanation for the loss of hydraulic fracturing fluid is fluid uptake by the shale matrix; where capillarity is the dominant process controlling this uptake. Detailed understanding of the rock wettability is thus an essential step in analysis of loss of the hydraulic fracturing fluid in shale reservoirs, especially at reservoir conditions. We therefore performed a suit of contact angle measurements on a shale sample with oil and aqueous ionic solutions, and tested the influence of different ion types (NaCl, KCl, MgCl₂, CaCl₂), concentrations (0.1, 0.5 and 1 M), pressures (0.1, 10 and 20 MPa) and temperatures (35 and 70 °C). Furthermore, a physical model was developed based on the diffuse double layer theory to provide a framework for the observed experimental data.

Our results show that the water contact angle for bivalent ions is larger than for monovalent ions; and that the contact angle (of both oil and different aqueous ionic solutions) increases with increase in pressure and/or temperature; these increases are more pronounced at higher ionic concentrations. Finally, the developed model correctly predicted the influence of each tested variable on contact angle. Knowing contact angle and therefore wettability, the contribution of the capillary process in terms of water uptake into shale rocks and the possible impairment of hydrocarbon production due to such uptake can be quantified.

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1. Introduction

With the decline of conventional oil and gas resources, gravitation towards unconventional resource development has increased significantly [1]. This is particularly true for oil and gas shales which have significantly contributed to the energy economy recently, in particular in the US [2]. Gas and oil shales typically have a very low permeability [3,4], and production is therefore usually associated with hydraulic fracturing operations, which increases the reservoir permeability through activating pre-existing fractures and creating new fractures thereby enhancing

connectivity [5]. However, one key technical-environmental issue associated with hydraulic fracturing is the very low recovery of hydraulic fracturing fluid; several hypotheses have been suggested to explain this disappearance of hydraulic fracturing fluids: hydration of clay minerals [6–8]; capillary forces [9–14]; creation of micro-fractures [3,9,15]; storage of the fluid within the fracture network [11,16]; and osmotic flow [7,17].

Water uptake is a complex function of these physico-chemical processes [7,10,14]. It is thus of vital importance to quantify the contribution of all processes in particular capillary forces [10,18] with respect to water uptake. Therefore the contact angle between shale and the introduced fluids needs to be thoroughly investigated.

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In this context, Fakcharoenphol et al. [17] showed that water-wet shale samples cannot significantly imbibe water of higher salinity than that of the in-situ pore water. In contrast, Xu and Dehghanpour [19] showed that organic-rich shale samples can absorb a considerable amount of water even when their surface is lipophilic. Moreover, Engelder et al. [16] investigated the wettability of organic shale samples and concluded that the samples are strongly oil-wet in the presence of air. They further mentioned that deionised (DI) water causes the highest contact angle on the sample surface and that highly saline solutions lead to the lowest contact angles, whereas other studies [7,20] suggest that the (water) contact angle (θ) increases with salinity.

However, all above mentioned studies have been carried out under atmospheric conditions, although it is well known that pressure and temperature can have a dramatic impact on θ [21–28]. In addition, there is a lack of systematic measurements of contact angles for brines containing major exchangeable cations (Mg^{2+} , Ca^{2+} , K^+ , Na^+) at different concentrations. We thus measured contact angles on a shale sample as a function of salinity and salt type at realistic reservoir conditions. Finally we proposed a physical model to qualitatively describe the physical processes responsible for variation in contact angle due to pressure, temperature, ion type and ionic concentration.

2. Experimental procedure

The shale sample was extracted from 1547 m depth of a nominated CO_2 storage site in New South Wales, Australia. The geological and chemical properties of this sample including X-ray diffraction (XRD), scanning electron microscopy (SEM), total organic carbon (TOC), and pore throat size distribution measurements using mercury intrusion were reported previously [28] and are therefore only briefly mentioned in this study. In the work presented here, an additional elemental analysis via X-ray fluorescence (instrument PW2400 XRF) was carried out to further deepen the understanding of the shale sample's chemical composition.

Due to the fact that higher gas solubility in the liquid phase makes the interpretation of contact angle data more complex, air (with low solubility in liquid phases: water and oil, especially with pressure) was chosen as the gas phase in this study. The shale sample was exposed to air plasma for 5 min to remove organic surface contaminations [29]. Subsequently the substrate was placed in a high pressure-high temperature cell (Fig. 1), and droplets of different ionic solutions or oil (n-decane) were dispensed onto the tilted substrate [30] at different pressures and temperatures. This process was videoed and advancing and receding contact angles were measured simultaneously following Al-Yaseri et al.'s [21] procedure.

A broad range of salt types (KCl, NaCl, $MgCl_2$ and $CaCl_2$ covering major exchangeable cations) and salinities (0.1, 0.5 and 1 M concentrations) were tested at 0.1, 10 and 20 MPa pressures at 70 °C temperature. Additional measurements were conducted at 35 °C and 20 MPa to investigate the effect of temperature.

3. Model development

In order to describe the physical basis for variation of contact angle with pressure, temperature, ion type and ionic concentration on minerals carrying negative charges such as clays, we developed a constitutive equation. We begin with a formulation for contact angles on mineral surfaces without electric charges. This formulation is then extended to minerals with surface charges, assuming that the electrically charged mineral surface has a constant electric potential, and that this electric potential reduces exponentially

towards the bulk solution. The zone where the electrical potential of the charged surface is active is the so-called diffuse double layer and its thickness is given by the Debye length [31]. This thickness coupled with the chemistry of the charged surfaces will be the basis for our model development for contact angle of charge surfaces.

3.1. Mineral without surface charges

When a drop of a liquid, in the presence of another fluid (liquid or gas), is placed on a solid surface with no electric potential, the droplet spreads across the solid surface, until the minimum free energy is reached [32]. This is related to the cohesion forces in the fluids and adhesion forces between the fluids and the solid surface. If contact angle hysteresis due to energy dissipation is neglected, and the free energy change due to an infinitesimal increase in base area of the droplet on the solid surface (surrounded by another fluid) is considered, the free energy of the system is written [32,33]:

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

where γ is the interfacial tension or surface free energy, F is total free energy of the system, θ is contact angle of the droplet and dA is the infinitesimal surface area (s, g and l represent the solid, fluid (gas or liquid) and liquid phases). When the minimum energy is reached and equilibrium is established, Eq. (1) turns in a special case, the Young's equation [33].

3.2. Mineral with electrically charged surfaces

We now use the concept of altering the contact angle by applying an electric field between a solid surface and an electrolyte [32,34]. Recall that the electric field is created by negative electrical charges present on the surface of the mineral [35] and there is no external electric field. Thus the solid-liquid free energy in Eq. (1) can be extended:

$$dF = \gamma_{lg} \cos \theta dA - \gamma_{sg} dA + (\gamma_{sl} dA - dW_c) \quad (2)$$

where dW_c is the work performed by the electric potential of the solid surface to re-arrange the positions of the cations and anions in the diffuse double layer in the presence of the electric field and therefore adjusts the γ_{sl} . By using the concept of a charged parallel-plate, dW_c can be related to the electrical energy stored in the system [32,34]:

$$dW_c = \frac{1}{2} dC \psi_s^2 \quad (3)$$

where ψ is the electric potential of the solid surface and C is the capacitance per unit area, defined as $dC = D \epsilon_0 \frac{dA}{k}$ according to Gauss's law: D is the dielectric constant of the medium (solid surface and surrounding liquid), ϵ_0 is the permittivity of vacuum and k is the distance between the charged plates where the electric potential is active. By combining Gauss's law with Eqs. (2) and (3) we obtain:

$$dF = \gamma_{lg} \cos \theta(\psi_s) dA - \gamma_{sg} dA + \left(\gamma_{sl} dA - \frac{1}{2} D \epsilon_0 \frac{dA}{k} \psi_s^2 \right) \quad (4)$$

In order to take into account the effect of liquid-surface interaction on the dielectric properties, the medium dielectric constant was employed in the formulation. There is no general equation to evaluate the dielectric constant of the medium but the experimental data for different rocks are often available in the literature [36]. Further, we write the contact angle of a neutral surface at equilibrium ($\cos \theta = (\gamma_{sg} - \gamma_{sl}) / \gamma_{lg}$; [33]) as [37]:

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