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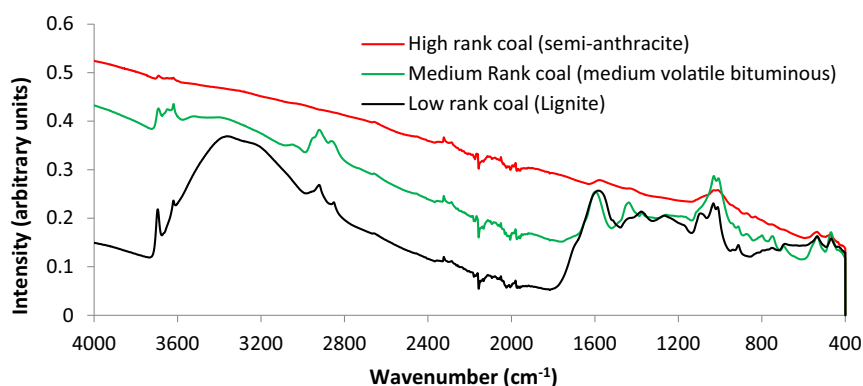
Influence of surface chemistry on interfacial properties of low to high rank coal seams

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HIGHLIGHTS

- Zeta potential of coals as function of temperature, salinity, salt type and coal rank.
- Contact angle as function of temperature, salinity, salt type and coal rank.
- Infrared spectroscopy of coals.
- Implications of measured data.

GRAPHICAL ABSTRACT



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ABSTRACT

Wettability of CO₂/water/coal systems is a fundamental petro-physical parameter, which governs the fluid flow and distribution in coal seams and thus directly affects CO₂-storage and methane recovery from unmineable coal seams. The recognition of wettability of coal/CO₂/brine systems help to de-risk CO₂-storage and enhanced methane recovery projects in coal seams. To understand the factors influencing the wetting characteristics of coals, a detailed examination and characterization of coal surface chemistry is essential and literature data in this context is missing. We thus measured zeta potentials as a function of temperature (298–343 K), brine salinity (0 wt% NaCl–5 wt% NaCl) and salt type (NaCl, CaCl₂ and MgCl₂) for coals of low, medium and high ranks. Further, we measured water advancing and receding contact angles as a function of temperature and salinity for the same experimental matrix in order to associate wettability changes to the surface charge at the coal/brine interface.

Moreover, coal surfaces were investigated by Fourier transformed infrared (FTIR) spectroscopy and the surface functional groups responsible for a particular wetting behaviour were identified. We found that zeta potential increased with temperature, salinity and cation valency. Both advancing and receding contact angles decreased with temperature, and increased with salinity and cation valency irrespective of the coal rank. Finally the XRD measurements and infrared spectra revealed that the presence of polar surface functional groups (e.g. Si–OH and carboxylic acid groups) which is responsible for the hydrophilic behaviour of low rank coals and the absence of these groups in high rank coal is responsible for their

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hydrophobic behaviour even at lower pressure. The high rank coal seams at high pressure are better for CO₂ storage and methane recovery.

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1. Introduction

The injection of CO₂ in depleted oil and gas reservoirs or deep saline aquifers is capable of trapping tremendous amounts of CO₂ and thus reduce anthropogenic CO₂ emissions [1–3]. Certain trapping mechanisms render CO₂ immobile in the porous medium, and these are structural trapping [4–7], residual or capillary trapping [8,9], dissolution or solubility trapping [10] and mineral trapping [11]. Coal seams, too, offer enormous potential for CO₂ storage and enhanced methane recovery by means of preferential adsorption of CO₂ [12,13]. Adsorption is the major CO₂ trapping mechanism in unminable coal seams and the wettability of the specific CO₂/brine/coal system plays a significant role in this context [12,14]. Typically, the adsorption capacity of CO₂ is higher than that of methane, consequently, CO₂ displaces methane toward the production well and itself gets sorbed within the micropores of the coal seam and remains trapped [12]. Moreover, Arif et al. [12] pointed out that CO₂-adsorption may be higher when coals are more CO₂-wet which thus leads to larger storage volumes [12].

Although coal wettability has been characterized as a function of pressure, temperature, salinity and coal rank [12,15], the factors responsible for a particular wetting behaviour are so far unclear. Specifically, the knowledge and characterization of coal surface chemistry is essential to understand the factors that influence its wettability [16,17]. In this context, zeta potential is used to probe the electric double layer at the surface of the coal. The nature of the electric double layer affects the repulsion/attraction of system's components, and depends upon the physicochemical properties of all components in the overall system [18]. Thus, what occurs at the brine/mineral or brine/rock interface (at a particular temperature and brine salinity) is strongly affected by the electrical double layer and studies have shown that this double layer is closely related to wettability [19–21].

We thus examined the electrochemical behaviour of coal by measuring zeta potentials as a function of temperature (298–343 K), salinity (0 wt% NaCl–5 wt% NaCl), salt type (NaCl, CaCl₂ and MgCl₂) and coal rank (low, medium and high rank) to allow surface characterization for broad range of conditions. In addition, following the same experimental matrix, we measured advancing and receding contact angles for air/coal/brine systems (at ambient pressure). The associated trends were analysed systematically and relationships were developed between zeta potential and wettability. We found that the zeta potential increased with increasing rank and increasing salinity and contact angles also increased with increasing rank and salinity, implying a positive correlation between zeta potential and wettability. However, with increasing temperature zeta potential increased but contact angle decreased, implying that correlation with respect to temperature is inconsistent. Moreover, the zeta potential results were also compared to our published contact angle data at high pressure, and we hypothesized that zeta potential at high pressure may follow similar trends as those at ambient pressure.

Further, in order to evaluate the impact of coal rank on wettability of the coal/CO₂/brine systems, we conducted Fourier transformed infrared spectroscopy (FTIR) measurements on the three coal samples (low, medium and high rank) and thoroughly characterized the surface functional groups. The results demonstrated that the abundance of OH and carboxylic acid groups on the low rank

coal is responsible for the hydrophilic nature of lignite and the lack of these groups explains the hydrophobic nature of high rank coal (semi-anthracite).

2. Experimental methodology

2.1. Fluid/sample preparation

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 1)] were used in this research. The samples were cut to cuboid shape (~1 cm × 1 cm × 0.5 cm) or crushed into fine powder (particle size ~30 μm) using milling equipment (Labtechnics Adelaide, Model TP-4/5, and Oscillation: 50 Hz). The powdered samples were then placed in an oven at 90 °C for 12 h until the weight became constant.

Coal/brine composite samples for all coal ranks were prepared by adding 2 wt% coal powder to aqueous salt solutions composed of DI water (0 M), 1 wt% NaCl (0.17 M), 5 wt% NaCl (0.855 M), 1 wt% CaCl₂ (0.27 M), and 1 wt% MgCl₂ (0.305 M). Note: the numbers in brackets represent the equivalent ionic strength of electrolyte which is typically preferred while comparing different types of salt of same strength. In this paper, we mainly used wt% to express salinity, however, for salts comparison we used ionic

Table 1
Properties of coal samples used.

Sample Rank	Semi-Anthracite	Medium-volatile Bituminous	Lignite
Geological location	Hazelton, Pennsylvania	Morgantown, West Virginia	North Dakota
Overall rank (used in this work)	High rank	Medium rank	Low rank
<i>Petrology</i>			
Vitrinite reflectance (R _o , % ^a)	3.92	0.82	0.35
Vitrinite (%)	89.6	73.1	83
Liptinite (%)	0	3.4	4
Inertinite (%)	7.6	18.8	10.8
Minerals (%)	2.8	4.7	2.1
<i>Proximate analysis</i>			
Moisture (air dried, %)	2.6	2	16.3
Ash (%)	9.7	6.4	7.8
Volatile matter (%)	2.9	32.4	34.8
Fixed carbon (%)	84.9	59.2	41.1
<i>Ultimate analysis</i>			
Ash (%)	9.7	6.4	7.8
Carbon (%)	82.6	78.6	54.6
Total Hydrogen (%)	2.35	5.07	5.27
Hydrogen (%)	2.06	4.85	3.45
Nitrogen (%)	1.16	1.54	0.62
Total Sulphur (%)	0.8	0.99	0.66
Oxygen by difference (%)	3.68	7.62	2.87
<i>Properties</i>			
Bulk density (g/cc)	1.30	1.28	1.44
Dry sample volume (cc)	16.55	12.77	4.26
Dry mass (g)	21.17	16.194	6.159

^a All percentages in above table represent weight percent.

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