



The effect of saline water on the critical degree of coal surface oxidation for coal flotation

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ABSTRACT

In this work, the effects of water with different salinities on the flotation of oxidized coals were studied and the underlying mechanisms were revealed. Flotation tests were conducted using coal samples with different degrees of surface oxidation and in water of different salinities. A critical degree of coal surface oxidation, above which the coal particles cannot be recovered by true flotation, was observed in flotation with all types of water. It is interesting to find that the critical degree of surface oxidation increased with water salinity, suggesting that coal flotation in saline water had a better tolerance for surface oxidation. The effects of water salinity on the flotation of oxidized coals were studied by investigating the change of coal surface properties and froth stability. On one hand, the surface hydrophobicity of oxidized coals decreased in saline water, which was unfavorable for coal flotation. On the other hand, the compression of the electric double layer in saline water could increase the flotation efficiency by reducing the electrostatic repulsion between particle and bubble. In addition, smaller bubbles and a higher stability froth were generated in saline water, which could also increase the flotation efficiency.

1. Introduction

Coal oxidation is a major problem confronting the coal industry due to its deleterious effects on coal flotation. With the depletion of high-quality coals, more and more oxidized coals have to be processed. Coal oxidation generates oxygenated groups including the aliphatic or aromatic hydroxyl groups ($-OH$), carbonyl groups ($-C=O$) and carboxyl groups ($-COO^-$), which make coal surfaces hydrophilic and repulsive from oily collectors, and thus exacerbate coal flotation. In the previous studies, we have defined the degree of coal surface oxidation as the percentage of oxidized carbon and identified the existence of a critical degree of surface oxidation for coal flotation using fresh water, above which the oxidized coals cannot be recovered by true flotation with diesel as the only collector (Chang et al., 2017).

Limited studies have been carried out to understand the effect of saline water on the flotation of oxidized coals despite the wide application of saline water in coal preparation plants. Due to the scarcity of fresh water and increasingly stringent regulations on the quality of discharged water, flotation plants have to use groundwater or seawater with a high salinity. In coal preparation plants, water is reused, leading to an increase in the salinity as a result of evaporation and ongoing salt inputs from groundwater and runoff from spoil. A survey of Australian coal preparation plants on the quality of process water revealed a wide

variation in water quality in terms of salinity, type and concentration of ions from plant to plant, and minimum, medium and maximum values of the concentration of major ions were given as an indication of variation in composition of process water (Ofori et al., 2010). Since flotation is sensitive to its environment, the oxidized coals may display a completely different flotation behavior in saline water and fresh water.

A number of studies have investigated the influence of saline water on the flotation of inherently hydrophobic or un-oxidized coals. It has been reported that the flotation of hydrophobic coals can be improved in saline water (Yoon, 1982; Yoon and Sabey, 1982; Kurniawan et al., 2011; Liu et al., 2013; Wang and Peng, 2014; Laskowski and Castro, 2015). Some researchers proposed that the hydrophobicity of coal surfaces could be increased in a highly concentrated electrolyte solution, which was attributed to the reduction of hydration layers surrounding coal particles by the adsorbed ions by lowering the electrical surface potential (Klassen and Mokrousov, 1963; Muller, 1988, 1990). However, other researchers reported that only the inherently hydrophobic coals could be floated very well in saline water, while the less hydrophobic coals were poorly recovered by flotation in saline water (Fuerstenau et al., 1983). Therefore, they argued that the inorganic ions could not make coal surfaces more hydrophobic (Laskowski and Castro, 2015). In addition, the presence of metal ions can also affect the electric double layers surrounding coal particles. The surface charge may

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introduce an energy barrier between particles and air bubbles when the particles collide with the air bubbles. This energy barrier can be reduced in saline water due to the compression of the electric double layer to increase the particle-bubble collision and attachment in flotation (Laskowski et al., 1992). Furthermore, the presence of metal ions in saline water can reduce the bubble size and increase the froth stability in flotation due to the inhibition of bubble coalescence (Marrucci and Nicodemo, 1967; Marrucci, 1969; Laskowski et al., 2003). The reduced bubble size increases the particle-bubble collision efficiency while the increased froth stability can increase the probability of the attached particles retained in the froth until being recovered as the concentrate (Farrokhpay, 2011), both increasing the overall flotation efficiency. However, most of these previous studies were based on fresh coal samples with minimum surface oxidation and therefore the conclusions may not be applicable to the oxidized coals.

In this paper, the effect of saline water on the flotation of oxidized coals was studied. In particular, the effect of different strengths of saline water on the critical degree of surface oxidation for coal flotation was examined. The mechanism underpinning the effect of saline water on the flotation of oxidized coals was also studied by determining the change in surface wettability and electrical surface property on oxidized coals, bubble size and froth stability in saline water, which govern the overall flotation efficiency.

2. Materials and experimental methods

2.1. Raw materials

The coal sample used in this study was obtained from Bulli Mine, New South Wales, Australia. It was a high volatile bituminous coal with a combustible matter content of 89.95 wt%, a moisture content of 2.12 wt% and an ash content of 7.93 wt%. The major mineral matter contained in this coal sample was kaolinite and quartz with a small amount of calcite and siderite. The sample was crushed, dry ground from about 10 cm in diameter to 80 wt% smaller than 200 μm which was the same particle size used in most coal preparation plants in Australia. To obtain coal samples with different degrees of surface oxidation, coal samples were oxidized at 150 °C for different times, based on the previous studies (Winmill, 1914; Schmidt et al., 1940). The degree of coal surface oxidation was determined using XPS and represented by the atomic percentage (at.%) of oxidized carbon on the coal surface. The details were reported elsewhere (Chang et al., 2017).

Both de-ionized water and saline water were used in this study. Saline water was prepared in the laboratory by adding certain amounts of different salts into the de-ionized water and it was classified as medium saline water and high saline water based on the corresponding ion strengths to simulate the medium and maximum saline water used in Australian coal preparation plants (Ofori et al., 2010). These two types of water were used in the previous studies to understand how saline water affected the flotation of un-oxidized coals (Wang and Peng, 2013, 2014). The chemical components of each saline water are shown in Table 1. The saline water was sufficiently mixed before any test to ensure a similar concentration of salts in each test. The conductivity of medium saline water and high saline water was 5.8 mS/cm and 14.5 mS/cm, respectively. The deionized water had a resistivity of 35 Ωm . The industrial grade diesel and MIBC (Methyl Isobutyl Carbinol, $\text{C}_6\text{H}_{12}\text{O}$) were used as the collector and frother, respectively.

Table 1
The composition of the saline water (mg/L).

	Ca^{2+}	Mg^{2+}	Na^+	K^+	HCO_3^-	SO_4^{2-}	Cl^-
Medium saline water	93.0	126.8	1177.4	20.9	835.1	1176.8	1020.3
High saline water	360.5	470.7	3050.8	58.1	2338.3	4505.8	2101.0

2.2. Flotation tests

Flotation was conducted in a JK batch Flotation Cell. For each test, 100 g coal sample was added in 1.5 dm^3 water to achieve a solids percentage of 6.5 wt% in the flotation cell. For each test, the agitation speed was fixed at 900 rpm and the air flow rate was fixed at 3.0 L/min. The chosen water, either de-ionized water or saline water, was used in all stages of the whole test. When saline water was used, the natural pH of the coal slurry was constant at about 8.0 due to a buffer effect. When de-ionized water was used, the natural pH of the slurry of the fresh coal was about 8.8. The natural pH of the coal slurry was observed to decrease with the degree of surface oxidation, probably due to the generation of the acidic groups, such as the phenolic hydroxyl groups and carboxyl groups, on coal surfaces. In this study, a small amount of 1 wt % HCl or NaOH solution was used to keep the pH at 8.0 in all the flotation tests. The dosage of diesel and MIBC used in all the flotation tests was kept at 240 g/t and 100 g/t, respectively, which was selected to achieve more than 90% flotation yield for the fresh coal in deionized water. Flotation concentrates and tailings were filtered, dried at 80 °C and weighed to calculate the flotation yield.

2.3. Contact angle measurements

The surface wettability of oxidized coals was measured using Sigma 700/701 based on the Washburn method that applies the theory of capillary penetration of liquid driven through a packed bed of particles in a capillary tube. The rate of the particles being wetted is determined by its wettability as shown in Eq. (1) (Washburn, 1921; Labajos-Broncano et al., 2001).

$$W^2 = C \cdot \frac{\rho^2 \gamma \cdot \cos \theta}{2\eta} \cdot t \quad (1)$$

where W is the weight of water, C is the material constant, ρ is the density of water, γ is the surface tension of water, θ is the contact angle of solids, η is the viscosity of water, and t is the immersion time. In this study, hexane was used as the reference liquid to determine the capillary constant. The measurement process was systematically controlled by a computer, and the mass of water adsorbed by wetted coal particles was recorded with time. The penetration rate of water increased with the hydrophilicity of the tested particles. To measure the contact angle of oxidized coal particles in de-ionized water, medium saline water and high saline water, coal particles were conditioned in the three types of water. The surface tension of the de-ionized water, the medium saline water and the high saline water was 72.7 mN/m, 73.1 mN/m and 73.7 mN/m, respectively. Each measurement was repeated three times and an average value was reported.

2.4. Zeta potential measurements

The zeta potential of coal particles was measured using Zeta-Probe Analyser developed by Colloidal Dynamics. This technique is based on the measurement of ultrasound waves generated by the moving charged particles. An MHz voltage is applied to a pair of electrodes in contact with the suspension. The electrically charged particles move back and forth between the electrodes and thus generate ultrasound. The zeta potential of coal particles can thus be obtained by measuring the ultrasonic wave. For each measurement, 5 g coal particles were added into 250 ml water with a solids concentration of 2.0 wt%. The slurry was conditioned at 300 rpm for 5 min before the measurement. The pH of coal suspensions was adjusted to the desired value using 1 wt% HCl or NaOH solution. Each measurement was repeated 7 times and the average value was reported.

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