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Research article

Adsorption and dispersion performance of oxidized sulfomethylated kraft lignin in coal water slurry

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ABSTRACT

In this study, oxidized and sulfomethylated lignin (OSL) with the charge density of −4.4 meq/g was used as a dispersant for coal water slurry (CWS). The impact of the OSL on CWS having different particle sizes was fundamentally investigated. The size of particles greatly influenced the adsorption of OSL on coal particles and the rheology and stability of the CWS. The maximum adsorption of OSL was found to be 0.96 mg/g on coal A having the particle size of 22 μm (coal particles with smallest size). OSL improved the wettability of coal by 5–28%. The interfacial tension between coal particle and OSL was reduced from 17 mN/m to 13 mN/m, and the zeta potential of the suspension dropped from −30 mV to −47 mV when 0.96 mg/g of OSL was adsorbed on coal A. The addition of 64 mg/L of OSL to coal A suspension (50 wt%) reduced its viscosity from 1350 to 520 mPa·s. The stability of coal particles was also improved as the destabilization index of CWS was reduced from 4 to 0.2. These results confirmed that the OSL was an effective dispersant for the coal water slurry.

1. Introduction

Recently, the indiscriminate use of oil and its derived products have led to energy depletion [1,2]. Numerous studies have reported alternative energy sources, such as biodiesel, hydrogen fuel [3], biofuel [4] and coal water slurry (CWS) for use in industry [5]. CWS seems to be a promising fuel due to its low production cost and ease of handling, and thus may be considered as an efficient alternative for partially replacing fossil fuels [6]. For CWS to be an effective fuel, it should have a high coal content for addressing the energy demand and moderately low viscosity for storage and transportation [7,8]. However, coal particles may not remain stable in slurry due to interactions between the coal particles, which would lead to a high apparent viscosity of CWS (30–70 wt%) that may exceed permissible viscosity needed for its storage [9,10]. The addition of dispersants can minimize the interaction of particles and improve the rheological behaviour of the slurries, thus improving the performance of CWS as fuel.

Synthetic polymers were studied as dispersants for coal water slurries in the past [11,12]. However, their non-biodegradable and toxic nature are their main barriers [13]. Recently, the products obtained via modification of kraft lignin found application as dispersants in many areas [14,15]. In one study, sulfomethylated kraft lignin produced via treating alkali lignin with sodium sulfite and formaldehyde improved the dispersion efficiency of red dye by 98% at a dosage of 0.6 wt% [16]. In another study, oxidized sulfomethylated kraft lignin produced via treating softwood kraft lignin with nitric acid, formaldehyde and sodium sulfite successfully improved the fluidity of cement admixture from 65 mm to 200 mm at a dosage of 0.5 wt% [17]. Alternatively, an anionic lignin-based product obtained via modification of hardwood kraft lignin with sodium chloroacetate improved the dispersion of clay suspensions (20 g/L) by 50% at the dosage of 0.2 wt% [18]. Previously, sulfomethylated lignin (0.6 wt%) prepared via treating soda lignin with formaldehyde and sodium sulfite had a similar behaviour to commercial lignosulfonate in dispersing mortar [19]. In another study on the dispersion of cement admixtures, oxidized and sulfomethylated lignosulfonate improved the fluidity of the suspensions from 160 to 190 mm at a 0.3 wt% dosage [20]. However, the application of kraft lignin-based dispersants in coal water slurries has not been studied. It is worth mentioning that, kraft lignin is already used as a fuel in kraft pulping process, thus its modification and use as a dispersant in CWS may be beneficial as it can ultimately contribute to the energy generation when burned along with CWS. The first objective of this work was to study the application of anionically charged kraft lignin as a dispersant for CWS.

Recent findings showed that polymeric dispersants adsorb at coal/liquid interface via hydrophobic or hydrophilic interactions [15,21]; thus affecting the stability of coal particles. The main goal of adding dispersant is to minimize the particle-particle interaction either by increasing the electronegativity of the coal surface or by increasing the wettability of coal particles [22]. Factors, such as surface tension of the

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dispersants and functional groups, associated with dispersants found to affect the interactions between the coal particles [23]. Hong and co-authors reported that polycarboxylic acid (0.5 wt%) reduced the viscosity of coal suspensions from 1200 to 580 mPa·s via generating electrostatic repulsion between the coal particles [24]. In another study on the stabilization of CWS (50 wt%) using polystyrene sulfonate (PSS) as a dispersant, the viscosity of CWS was reduced to 150 mPa·s with an increase in concentration of PSS from 1.0 to 1.8 wt%. This behaviour was attributed to the adsorption of PSS on coal particles and thus to the increase in wettability of coal particles [25]. The second objective of this work was to study the influence of OSL on the viscosity and wettability of coal particles having different sizes. For the first time, the interactions between OSL and coal particles in an aqueous solution were studied fundamentally and the impact of OSL on stabilizing coal water slurries containing particles with different sizes was determined.

2. Materials & methods

2.1. Materials

Hardwood lignin was produced by the Lignoforce technology of FPInnovations located in Thunder Bay, ON [26]. Sodium sulfite (99%), potassium nitrate (99.99 wt%), potassium hydroxide, *Para*-hydroxybenzoic acid (PHBA) and dimethyl sulfoxide-d_6 were all obtained from Sigma Aldrich company and used as received. Bituminous coal was obtained from Fisher Scientific. Nitric acid was obtained from Sigma Aldrich company and diluted to 30 wt% prior to use. Standard hydrochloric acid (HCl) solution (30–35%) was obtained from Fluka analytical and used as received. Sulfuric acid (95–98%) was obtained from Sigma Aldrich company and diluted to 0.1 M prior to use. Cellulose acetate dialysis membrane (molecular weight cut off of 1000 g/mol) was obtained from Spectrum Labs. Inc., USA. Polydiallyldimethylammonium chloride (PDADMAC) was obtained from Sigma Aldrich company and diluted to 0.005 M prior to use. Potassium polyvinyl sulfate (PVSK) was obtained from Wako Pure Chemical Industries Ltd., Japan and diluted to 0.005 M prior to use.

2.2. Preparation of coal samples and CWS

Bituminous coal was ground using a laboratory Micro mill, Bell Art Products (NJ, USA). The samples were then categorized into three fractions (coal A, coal B and coal C) via screening crushed coal powder with sieve meshes (American standard testing sieves) of three different pore openings (60, 200 and 400 mesh). The fractionated coal samples were then used to prepare coal water slurries (CWS) by mixing them in distilled water at a 50 wt% solid concentration for 24 h and 1000 rpm. The pH of the slurry was maintained at pH 7 in all the experiments.

2.3. Synthesis of oxidized and sulfomethylated kraft lignin

Oxidation and sulfomethylation of hardwood kraft lignin was performed as described previously [17]. In this experiment, 5 g of hardwood kraft lignin was mixed with 40 mL of nitric acid solution (25 wt%) in 250 mL three neck flasks. The reaction was carried out at desired temperature (100 °C) for 1 h with constant stirring at 200 rpm (i.e., optimized conditions based on a previous study) [17]. After the reaction, the mixture was cooled to room temperature using tap water and its pH was then adjusted to 7 with 0.1 M NaOH. To this reaction mixture, 1/1 M ratio of lignin/formaldehyde and 0.6/1 M ratio of sodium metabisulfite/lignin were added and sulfomethylation reaction was carried out at 100 °C for 2 h and 200 rpm [17]. Upon completion of the reaction, the mixture was cooled to room temperature and its pH was then adjusted to 7 using 0.1 M H_2SO_4. The reaction contents were dialysed using the membrane dialysis for 2 days by changing water for every 12 h time. After dialysis, the samples were dried at 105 °C and stored at room temperature for future use.

2.4. Charge density and sulfonated group analyses

The charge density of unmodified lignin (UL) and oxidized sulfomethylated lignin samples (OSL) was determined using a Particle Charge Detector (Mutek, PCD 04, Germany). The samples were initially dried at 105 °C overnight in an oven. In this set of experiments, UL or OSL sample (0.2 g) was dissolved in 20 mL of deionized water and incubated in a water bath shaker (Classic C76, New Brunswick Scientific, Edison, NJ, USA) for 2 h at 30 °C and 150 rpm. After the incubation, the samples were centrifuged at 1000 rpm for 10 min and supernatants were collected for the charge density analysis. A 1 mL of supernatant was titrated against PDADMAC solution (0.005 M) to measure the charge density of the samples.

The surface charge density of coal particles was determined via a back-titration method with a Mütek, PCD 04, Particle Charge Detector (Germany). Approximately, 0.2 g of coal powder was suspended in 50 mL of PDADMAC (0.005 M) solution and the suspension was incubated at 30 °C for 2 h at 150 rpm. After the incubation, the samples were filtered using Whatman#1 filter papers and the filtrates were titrated against PVSK solution (0.0055 M). Similarly, the titration analysis was conducted for control sample (i.e., PDADMAC solution) and the difference was considered to quantify the surface charge density of coal particles.

The sulfonated group content of UL and OSL samples was measured using an automatic potentiometer, Metrohm, 905 Titrado, Switzerland. In this experiment, a 1 g of dried sample was suspended in 100 mL of deionized water and its pH was adjusted to 7.0 with 1 M H_2SO_4. The solution was then titrated against a cationic polymer, TEGO trant A 100, in order to determine its sulfonate group content according to Eq. (1):

$$\text{Sulfonated group (SG)} = \frac{V \times C}{S} \quad (1)$$

in which V is volume (mL) of titrant (TEGO trant) consumed, C is the concentration of TEGO trant (mol/L) and S is the dried weight of lignin (g) samples used in this analysis.

2.5. Surface area analysis

The surface area of coal particles was determined using Quantachrome surface area analyzer, Nova2200e (USA). In this set of experiments, the samples were initially dried in an oven at 105 °C overnight and approximately 0.05 g of sample was pretreated for 4 h at 250 °C prior to analysis. The specific surface area of the samples was then analyzed according to Branuer-Emmett-Teller (BET) method via adsorption-desorption isotherms using nitrogen gas at −180 °C in the relative pressure range of 0.01 to 0.99 [27].

2.6. Particle size distribution analysis

In this study, 1 g of dried coal powder was added to 100 mL of deionized water and incubated in a water bath shaker at room temperature and 150 rpm. After the incubation, the samples were transferred to a MasterSizer 2000, Malvern, particle size analyzer (UK) equipped with a light scattering detector to measure the particle size distribution of the coal particles. All the measurements were carried out at room temperature. Three measurements were carried out for each sample and average values were reported.

2.7. Elemental analysis

The elemental analysis of UL or OSL and coal samples was performed using an Elementar Vario EL Cube, Germany, elemental analyzer by following the combustion analysis method. The samples were first dried in a 105 °C oven overnight in order to remove any moisture. Approximately, 2 mg of sample was weighed in silver vessels and

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