Fuel 193 (2017) 54–64

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

The effect of densification on brown coal physical properties and its spontaneous combustion propensity

Mohammad Reza Parsa [a], Yoshimitsu Tsukasaki [a,b], Emily L. Perkins [a,c], Alan L. Chaffee [a,*]

[a] School of Chemistry, Monash University, Clayton, Victoria, Australia
[b] Nippon Steel and Sumitomo Metal Corporation, 6-1 Marunouchi 2-Chome, Chiyoda-ku, Tokyo 100-8071, Japan
[c] Department of Chemical Engineering, Monash University, Clayton, Victoria, Australia

HIGHLIGHTS

- Densification of brown coal with NaOH reduces its tendency to spontaneously combust.
- Surface area and pore volume of densified products directly correlate with T_{cr}.
- Vickers hardness of densified products increases with NaOH concentration.
- Surface morphology of products densified with NaOH becomes smoother.

ARTICLE INFO

Article history:
Received 24 May 2016
Received in revised form 1 December 2016
Accepted 7 December 2016

Keywords:
Brown coal
Lignite
Densified coal
Self-heating
Surface area
Pore volume

ABSTRACT

The process, where brown coal is extruded after mechanical kneading and then allowed to air dry slowly to form a product known as 'densified coal', was applied to reduce the moisture content of two Victorian brown coals. NaOH at different concentrations (0–1.5 M) was used as an additive in the kneading step. The spontaneous combustion propensity of the densified products was evaluated and compared against multiple physical properties and morphological features of materials.

The densification process reduced the moisture content of the sample from around 60% to around 12%. NaOH addition led to a progressive reduction in the CO_2 surface area, as well as the porosity determined by mercury intrusion, due to the development of a stronger electrostatic network within the coal structure. The reduced micropore volume limits the accessibility of O_2 to internal surfaces of the coal leading to a significant increase in the critical ignition temperature (T_{cr}) measured by the wire basket test method. SEM imaging indicated that the coal particle surface changed from spongy and porous for nitrogen dried raw coal to very smooth and contiguous for densified coal. These trends also correlated with progressive reduction in the CO_2 surface area, as well as the porosity determined by mercury intrusion porosimetry.

1. Introduction

The state of Victoria in Australia is said to possess 25% of the known world reserves of low rank coal. Victorian brown coal is a very significant source of energy because of its potential for open-cut mining (lower mining cost), its high reactivity and low ash content. Although it has the potential to remain a major energy source for the local economy into the future [1], a significant obstacle to further development of Victorian brown coal usage is its high moisture content, typically around 60%. Thus, it would be beneficial to remove the water in order to decrease transportation costs, increase power plant efficiency and reduce net CO_2 emissions.

A further issue is that dried brown coal easily disintegrates into fine dust, which results in an increase of spontaneous combustion propensity [2,3]. Spontaneous combustion takes place when the accumulation of heat within carboniferous materials, resulting from some low temperature chemical and/or physical processes, is faster than the release of heat into the environment [4,5]. Spontaneous combustion of coal is a serious issue in the world's coal industry causing many problems such as the difficulty of transportation of coal over large distances, fire safety concerns, storage issues and long-term environmental problems [6].

Self-heating needs to be prevented and/or controlled, but the processes responsible for self-heating are complex. The tendency

* Corresponding author.
E-mail address: alan.chaffee@monash.edu (A.L. Chaffee).

http://dx.doi.org/10.1016/j.fuel.2016.12.016
0016-2361/

for spontaneous combustion in the field is affected not only by inherent factors, which were the subject of our study, but also by external factors such as ambient temperature and humidity, stockpile size and management protocols, moisture content and precipitation patterns, prevailing winds and, where relevant, transport conditions [7,8]. The complexity of chemical structure, the inhomogeneity of coal seams (and samples) and the variability of experimental methods has led to inconsistent, confusing and sometimes conflicting results in the literature [5]. Although much valuable research has been carried out to describe the spontaneous combustion of coal, studies focused on developing a fundamental scientific understanding of the impact of physical and chemical properties on low temperature reactions are rare [5,9].

Many techniques have been proposed for removing the water from brown coal by evaporative and non-evaporative techniques. Some of these techniques such as hydrothermal dewatering [10], mechanical thermal dewatering [11] and the densification process [12] have advantages that include a reduced energy requirement and less severe conditions to remove the water from the coal [10,13]. The optimum drying process would require minimal energy; and produce a product of sufficient strength such that it can be handled without attrition together with physical properties that provide it with a reduced propensity to spontaneously combustion.

The densification process developed in the late 1980s [12] transforms run-of-mine brown coal into a dense, dry, hard product. The process begins by shear attrition using a batch or continuous kneading process which leads to the formation of a dough of plasticine consistency. Applied shearing stress releases water from the cellular structure of the coal, forming a relatively homogenous smooth plastic dough during the kneading stage. The dough is extruded in order to produce pellets of the desired shape and size. Finally, the pellets are air-dried at room temperature forming hard and dense coal pellets with high compressive strength [12,14]. However, the impact of densification on the spontaneous combustion propensity of the coal has not previously been investigated in any systematic way.

Sujanti and Zhang studied the effect of different inorganic additives on brown coal spontaneous combustion after impregnation from aqueous solution [9,15,16]. Potassium chloride, Montan powder, and sodium chloride were shown to have the highest inhibitory effect, followed by calcium chloride and magnesium acetate, whereas the spontaneous combustion was promoted when calcium carbonate, sodium acetate, potassium acetate, and pyrite were added to the coal. The spontaneous combustion propensity of coal samples was not significantly affected by the addition of sodium nitrate and ammonium chloride [9]. However, it remains unknown just how these additives affect the coal properties so as to modify the spontaneous combustion propensity.

The objective of this study was to develop an understanding of the relationship between densified brown coal's physical properties and the spontaneous combustion behaviour of brown coal. We have systematically modified some individual brown coal properties (surface area, pore size distribution, hardness) in a controlled way which also overcomes the complexity inherent in some prior studies that have focussed on comparisons between different coals. The spontaneous combustion tendency of the products was evaluated using the wire basket method.

2. Experimental

2.1. Coal samples

Two Victorian brown coals, Morwell (MW) and Loy Yang (LY), have been used in this study originating from the Latrobe Valley, Victoria, Australia. Table 1 shows the proximate and ultimate analyses of both coals. The Campbell Microanalytical laboratory at the University of Otago carried out the ultimate analysis. The inorganic compositions of the coal samples, determined as described in Section 2.4, are given in Table 2. Received samples were milled to <3 mm and then homogenised prior to the densification process. All samples were ground and sieved to <0.018 mm before all other analytical tests.

In order to remove the inorganic matter, an acid washing procedure was carried out by placing 175 g wet coal in 2 l side-arm flasks with 1.5 l sulfuric acid (0.1 M concentration) and stirred for 24 h. Acid washed (AW) samples were vacuum filtered and further washed with deionized water, and vacuum filtered to remove acid. This rinsing procedure was repeated until the pH of sample became constant. Water washed (WW) samples were also rinsed in the same way, so as to mimic any effects of further washing on the coal as experienced by acid washing.

2.2. Densification process

The process begins by applying shear attrition to 100 g of wet coal and desired amount of water or NaOH solution, using an IKA HKD-T 0.6 laboratory kneader. The ratio of total water (added, plus the coal moisture) to solid coal (dry basis) employed was 95 ml/40 g, and the amount of NaOH to db coal was 0.7 g (0.5 M), 1.4 g (1 M) and 2.1 g (1.5 M) NaOH to 40 g db coal. These ratios were kept constant for washed samples (WW and AW) with higher initial moisture content.

During the kneading stage, any residual component and cellular structures of the coal collapses under the applied shearing stress, the sample becomes thoroughly homogenised and water within the coal structure is released. After 1 h of kneading with a torque of 25 N cm and a speed of 120 rpm a smooth coal dough which is plastic-like in character, is produced.

Next, the dough is extruded through an 8 mm diameter steel nozzle using a compressed air driven rod (620 kPa). The extruded material was collected in a tray and cut into 5–10 mm length pellets with a scalpel. Finally, the pellets were air-dried at ambient conditions for at least 72 h to form hard and dense coal pellets [12,14].

2.3. The wire basket method

The wire basket method was used to determine the T_{cr} of samples [17]. The wire basket test was established as a standard test method for characterising the inherent tendency of the coal to react in circumstances where heat insulation is minimal (sample size of 1 cubic in.) and air flow is not impeded (fan forced oven). For a given coal sample of known particle size, T_{cr} values are reproducible to within ±2 °C or less [5].

Briefly, 10–13 g of sample, which can vary according to the packing density of the sample, was loaded to fill a 1 in. stainless-steel mesh cube basket. The loaded basket was fixed to a sample holder. Three K-type thermocouples were inserted vertically in the middle of the sample, one at the centre of the basket and the other two half-way between the centre and two opposite outside edges of the cube. A fourth thermocouple was positioned horizontally to measure the actual oven temperature. The wire basket was suspended in a fan forced oven, which could be heated to a pre-set temperature to observe the combustion behaviour of the sample.

At the start of the experiment, the oven was set to the desired temperature. Fan forcing maintained the airflow during the experiment. The temperatures measured by the thermocouples were continuously logged. To obtain T_{cr} for a sample, the wire basket experiments were repeated at different oven temperatures, at 2 °C intervals. The T_{cr} of each sample was determined by averaging