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Study of the Susceptibility of Coal for Spontaneous Combustion using Adiabatic Oxidation Method

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Largest contributor for fires in a coal mill is due to presence of combustible materials in pulverisers. Dry coal that is accumulating or settling in pulveriser components can spontaneously ignite. Aim of this study was to investigate the effect of moisture content, relative humidity of airflow, airflow rates, coal size fractions, nitrogen suppression and prior oxidation on the susceptibility of coal for spontaneous combustion. Adiabatic oxidation method was adopted whereby spontaneous heating potential was measured according to total temperature rise (TTR) while considering temperature rise versus time. Results demonstrated that humidity of air is an important factor in deciding whether heating will progress or not. Particle size affected the TTR values whereby oxidation rate of coal increased with decrease in particle size up to a critical diameter below which dependence ceases. Total temperature rise recorded in the test varied with storage time. Findings of this study have identified factors responsible for spontaneous heating and could be incorporated in design of coal mills to prevent damage to equipment.

1. Introduction

Occurrence of pulveriser fires is a major concern for coal mills. Accumulation and settling in pulveriser components allows coal to dry. Such an accumulation can spontaneously ignite. Without adequate steps, a fire may cause extensive damage requiring long down times and great expense. Spontaneous combustion of coal is initiated by low temperature oxidation of coal that takes place whenever it is in contact with atmosphere (Arisoy and Beamish, 2015). This process is an exothermic reaction in which heat generated is dissipated by conduction and convection. If heat generated from the process is greater than heat lost, spontaneous combustion is likely to occur. Coal oxidation obeys an Arrhenius type rate law, whereby the reaction rate increases exponentially with increased temperature (Zhu et al., 2013). This type of reaction accounts for the runaway phenomenon. Oxidation of coal that is barely warm can accelerate to the point where spontaneous combustion occurs, and visible fire breaks out.

Past researchers work has mainly focussed on study of inherent properties of coal with a correlation to the self-heating tendency (Sahu et al, 2009). Studies have mainly focussed on external factors relating to mining such as particle size, geological condition, and mining methods (Morris and Atkinson, 1988). Limited number of studies has been conducted on the study of conditions related to a coal pulveriser mill and their relationship to the self-heating tendency of coal. Previous experimental investigations have been carried out with a heating system used to initiate the self-heating process (Mohalik et al, 2016). On the other hand, adiabatic oxidation test allows the coal sample to exhibit its self-heating behaviour without any external heating system (Beamish et al, 2001). This method can be modified to closely replicate in situ conditions in a coal mill and this allows for a variable study. Effects of air flow rate, air moisture content, nitrogen suppression and particle size could be integrated into adiabatic oxidation tests.

Aim of this study is to identify conditions that are favorable for spontaneous combustion of coal to occur. The intention is also to provide required information and correlation of variables with self-heating behavior of coal. In the following sections the methodology used in conducting adiabatic oxidation tests is described. Results of

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moisture content, heat of wetting effect, size fraction, nitrogen suppression and ageing of coal sample tests are presented and discussed.

2. Methodology

Adiabatic oxidation test apparatus developed in Nottingham University to study spontaneous combustion liability of pulverized coal samples (Ren et al, 1999) was used in this study. Samples of coal, pulverized to grind size of -75 µm to +300 µm, and weighing 200 g, were placed in the reaction vessel encased in a calorimeter (Figure 1) for drying at a preset temperature. The reaction vessel consisted of thermostat cabinet with thermocouples connected to a temperature control unit.

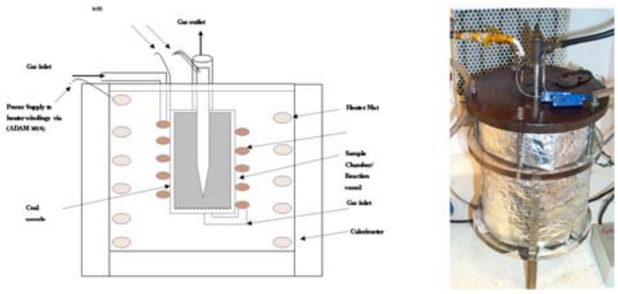

Figure 1: Schematic diagram of reaction vessel within the calorimeter. Picture of the calorimeter installed with air and nitrogen flow inlet and outlet, thermocouples connected to data acquisition and temperature control unit.

Tests were carried out to determine the most suitable airflow rate by changing the flow rate entering the reaction vessel. Airflow rates of 150, 200 and 250 cc/min were used under the conditions of dried coal and saturated air. Tests with 200 cc/min showed the maximum oxidation potential of the coal sample and this was chosen as the standard flow rate for tests throughout this study.

To check accuracy and sensitivity of the apparatus and verify the self-heating curve is representative of coal sample, repeatability tests were carried out. At least three repeatability tests were conducted within 3 to 4 days for given coal sample to ensure no pre-oxidation and ageing effect. Tests were conducted with a high volatile bituminous coal sample throughout this study. Proximate analysis of the coal sample was fixed carbon content of 40.7 %, volatile matter of 29.4 %, ash content of 12 % and calorific value of 17,282 kcal/kg. Up to four tests were conducted for a given sample. Variation in total temperature rise between each test was within ±0.5 ℃.

Calorimeter and reaction vessel were set to the adiabatic oven mode at an initial temperature of 40 °C using the temperature control unit. Coal samples were dried inside the reaction vessel under nitrogen flow at 40 °C for approximately 18 hrs to ensure sample was moisture free. After sample temperature had stabilized, oven was switched to remote monitoring mode and gas selection switch turned to oxygen with a constant flow rate of 200 cc/min. Temperature change with time during oxidation process was recorded by a data logging system.

Adiabatic oxidation tests were carried out on coal samples at an initial temperature of 40 °C to mimic typical conditions in a coal milling plant. Self-heating rate was monitored for approximately 8 hours. Initially self-heating increased at constant rate. Approximately after 4 hrs of testing there was no increase in temperature rise. Total temperature rise (TTR) was considered as the difference between the initial temperature and the maximum temperature observed expressed in °C. Results obtained in this study have been presented as self-heating curves to enable comparison under varying conditions.

3. Results

3.1 Moisture Content Test

The following tests were carried out:

Test A: Dry coals at initial temperatures of 40°C (equilibrated with dry nitrogen gas flow), with a saturated airflow at 200 cc/min at 100 % relative humidity (r.h.).

Test B: Wet coals (equilibrated at 40°C with wet nitrogen) at moisture content of 100 % r.h., with dry airflow at 200 cc/min of 0 % r.h.

Test C: Wet coals (equilibrated at 40°C with wet nitrogen) at moisture content of 100 % r.h., with wet airflow at 200 cc/min of 100 % r.h.

Results indicated that when moisture content of the coal is in excess of that of the airflow, temperature of coal falls below 40 °C after an initial temperature rise (figure 2- curve C). Wet coal sample (relative humidity of air in contact with coal was measured 100 %) reacted at 40 °C with dry air (at 0 % r.h.).

Curve B of figure 2 shows the heat generated by the adsorption of moisture onto the coal surface. In this case a wet sample (100 % r.h. with wet nitrogen) reacted at 40 °C with air of 100 % r.h. When air containing moisture was used under same conditions a temperature rise up to 4.2 °C was recorded. This effect is further illustrated by curve A of figure 1. In this case a dry coal sample (5 % r.h. of air in contact) reacted with airflow of 100 % r.h. Heat generated has increased up to 14 °C for given coal sample. These results demonstrated that dry coal when exposed to saturated air shows the greatest susceptibility for spontaneous combustion to occur.

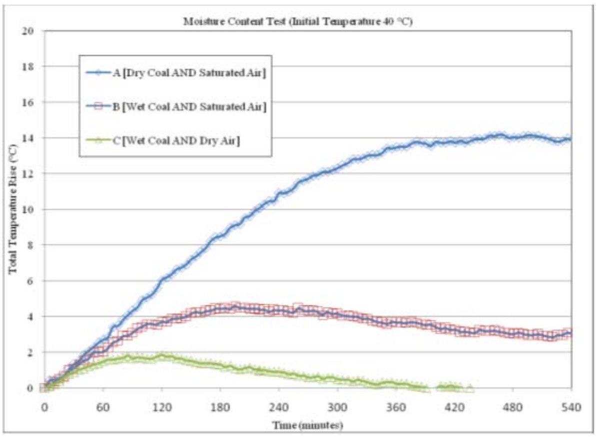

Figure 2: Self-heating curves for coal sample tested under Dry coal & Saturated air [curve A]; Wet coal & Saturated air [curve B]; Wet coal & Dry air [curve C].

Self-heating of coal is most dominant when dry coal was exposed to humidified airflow. Results demonstrated the moisture effect on early stages of self-heating. Therefore, humidity of air in contact with coal is an important factor in deciding whether heating will progress rapidly or not. In a coal mill it is likely that pulverised coal dust will accumulate at some ‘dead’ corners and gradually dry out. When saturated air is introduced, this ‘settled’ coal dust could absorb moisture and release heat, which may provide initial energy for heating. On the other hand, when moisture content of coal is in excess of surrounding atmosphere, temperature of coal falls due to latent heat.

3.2 Heat of wetting effect on Self-heating of Coal

To examine role of moisture as an oxidation catalyst, coal sample was tested under both oxidizing (O_2) and non-oxidizing (N_2) conditions at a moisture content of 100 % r.h. and flow rate of 200 cc/min. Figure 3 shows results of the heat of wetting test. Temperature rise with saturated N_2 (heat of wetting) and temperature rise with saturated air as test gas at initial temperature of 40°C is shown. The sample reached a maximum temperature rise of 14 °C with saturated air, whereas with saturated N_2 maximum temperature rise was up to 11.8 °C.

Despite the absence of oxygen, self-heating due to saturated N_2 accounted for approximately 80 % of total temperature rise. Shape of the curve indicates that the oxidation process for this coal is initially very rapid due to adsorption of moisture. Coal becomes less reactive at later stages, as oxidation process cannot be sustained for longer periods.

In the test with dry air, coal sample reached a maximum temperature rise of just 1.8 °C. Shape of the curve indicates that oxidation can sustain self-heating in the absence of moisture during initial stages. But, at later stages, temperature of coal falls due to latent heat of moisture as it evaporates.

While comparing temperature histories in these tests with saturated N_2, and saturated air, it was demonstrated that heat of wetting is a significant factor in self-heating of coal at low temperatures, although it is unlikely heat generated by wetting alone will be sustained at later stages because of limited pore surface area available for moisture desorption. This phenomenon is demonstrated by levelling of temperatures and subsequent dip in the profiles.

3.3 Size Fraction Test

Effect of increasing particle size on the self-heating of coal was examined in this study. Three samples of coal of increasing particle diameter were used. Proximate analysis of coal sample was fixed carbon content of 45.3 %, volatile matter of 30.7 %, ash content of 8.9 % and calorific value of 21,118 kcal/kg. Tests were conducted under standard conditions and results are shown in figure 4.

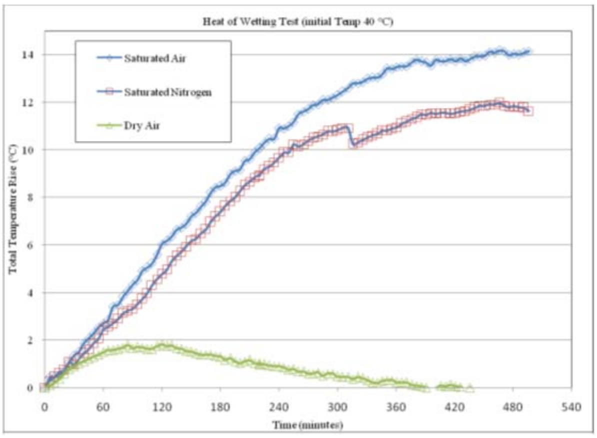

Figure 3: Self-heating curves for the combined effect of moisture and O_2; saturated N_2 (heat of wetting); and dry O_2.

These results indicate a dependence of temperature rise on particle size, down to a particle diameter of 75 to 150 µm. Oxidation rate of coal increases with a decrease in particle size, up to a critical diameter of 75 µm. Beyond this particle size dependence ceases. Increased reactivity is due to increased accessibility of oxygen to internal surfaces of coal sample with decreasing particle sizes.

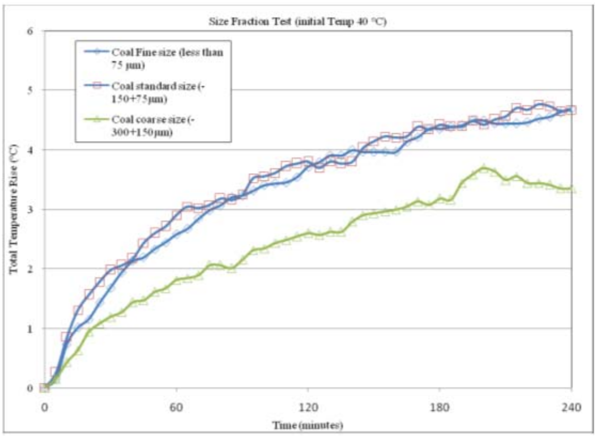

Figure 4: Self-heating curves for size fraction test on coal samples for Fines [-75mm]; Standard size [-75+150mm] and Coarse size [-150 +300mm].

However, based on the above statement, one may have expected a greater temperature rise with fine coal sample (less than 75 µm) when compared with standard size fractions (-150+75 µm). Oxidation of coal increased with decrease in particle size, down to a critical size up to 150 µm below which dependence ceased. Total surface area available is independent of particle size diameter below this critical size. Also, ultra-fine size fractions may behave as larger particles due to agglomeration, suppressing reactivity of coal.

3.4 Nitrogen Suppression Test

To investigate effect of O_2 availability on self-heating of coal, a coal sample was exposed to flows containing 21 %, 18 % and 14 % O_2 by volume. Coal sample had fixed carbon content of 53.1 %, volatile matter of 24.7

%, ash content of 13.5 % and calorific value of 19,735 kcal/kg. Tests were conducted under standard conditions to determine total temperature rise. Results are shown in figure 5.

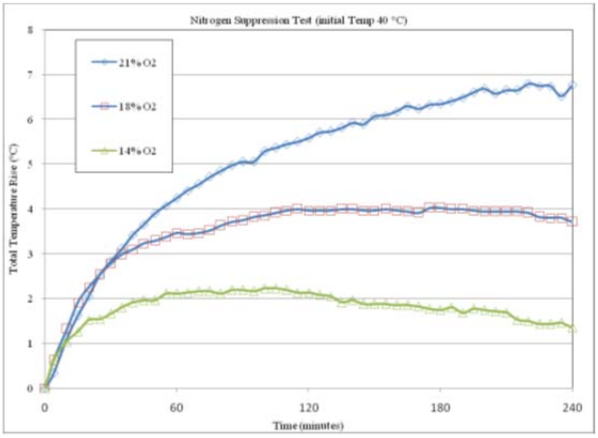

Figure 5: Self-heating curves for coal at 21 %; 18 % and 14 % O_2 by volume.

Tests with 21 % O_2 showed total temperature rise of 6.7 °C. Reducing O_2 concentration to 18 % decreased total temperature rise to 4 °C. At an O_2 concentration of 14 %, temperature rise was just 2.2 °C. Hence a significant decrease of 67 % in temperature rise and coal reactivity was recorded due to O_2 deficiency. It was found that effect on rate of oxidation in terms of total temperature rise differs little regardless of type of coal. Following exponential relationship was derived between the total temperature rise and corresponding O_2 concentration given in Eq (5).

$$T_t = 0.236\, e^{(15.85*O_2)} \tag{5}$$

Where, T_t – total temperature rise of coal, O_2 – oxygen concentration.

Nitrogen suppression test has indicated measured rate of temperature rise could be expressed as exponential function of oxygen concentration in contact with coal. Self-heating temperature rise in coal could be reduced by 67% with reduction in oxygen concentration to 14%. Further tests would be required with coals of varying types to formulate a correlation.

3.5 Ageing Test

Partial oxidation occurs due to exposure of coal surface to air during sample handling and storage. Total temperature rise recorded over a period of 60 days is shown in figure 6.

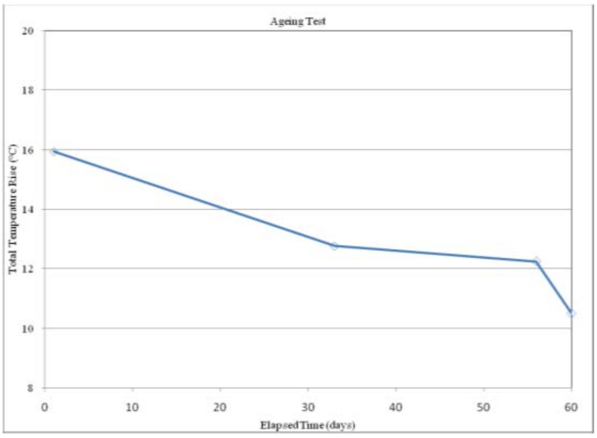

Figure 6: Decreasing trend in total temperature rise with sample age.

These results show that for a stored sample of particle size of – 212 μm, total temperature rise dropped by 5.2 °C. It is clear that an oxidation effect due to ageing has taken place even though samples were stored under controlled conditions. Internal pores of coal are blocked by prior oxidation during preparation and drying of samples. Since oxidation is dependent on diffusion of O_2 into the pores, this has caused rate of oxidation to drop.

Rapid drop in total temperature rise from 12.2 ºC to 10.5 ºC from day 56 to day 60 was recorded. The decay plot for total temperature rise can be expressed by Eq (6):

$$T_R = 15.58 - 0.064\,t \tag{6}$$

Where, T_R is maximum temperature rise, t is time in days, the constant -0.064 was obtained from the line of best fit for these results. Self-heating potential of the coal sample decreased by 50% due to pre-oxidation effects over the period of two months. The above relationship could be used to extrapolate total temperature rise and assess the spontaneous combustion susceptibility of coal over time. Further tests would be required with coals of varying types to formulate a correlation.

4. Conclusions

This study has shown that many factors, other than coal rank, must be considered to define the risk of coal to spontaneous combustion in coal mills such as flow rates, relative humidity, particle size distribution, nitrogen suppression, and ageing of coal. Choice of coal tested was predominantly sub-bituminous. Further study would be of value to investigate the self-heating propensity of high rank anthracite and low rank lignite coals to be able to obtain statistical correlations. Many of the factors that can be controlled in the adiabatic calorimeter may not be applied without problems in a coal mill. These results have provided information to identify factors responsible for spontaneous combustion and thus enable appropriate design of a coal mill to minimize risk and consequent loss of calorific value.

Acknowledgments

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