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MEASUREMENT OF CRITICAL SELF-IGNITION TEMPERATURES OF LOW RANK COAL PILES

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ABSTRACT: Considerable research exists on self-heating or spontaneous combustion of coal stockpiles from various aspects. The equation for critical temperature of self-ignition was derived from the Frank-Kamenetskii model expressing heat balance between heat generation rate in the centre of a coal pile and heat transfer from the outer surface of the pile. However, critical ignition-temperatures of low-rank coals have not been established for safety criteria in storing and transporting these coal. In this study, experimental apparatus and measurement procedures of thermal diffusivity and internal temperature in the coal pile have been presented to evaluate the critical self-ignition temperature of coal samples. The coal samples tested were low-rank lignite and sub-bituminous. Their critical ignition-temperatures were evaluated based on laboratory temperature measurements of coal piles in cube mesh-boxes of three different sizes (25, 50 and 100 mm side length) placed in hot ambient-air at temperatures ranging from 50 to 140 ºC under standard atmospheric pressure. Analysis of the results has enabled the critical ignition-temperature of the coal sample to be presented as a function of stockpile volume.

INTRODUCTION

Coal is a major energy resource for the world, and its production and consumption will increase with increasing demand for electricity in developing countries. Low rank coal, such as sub-bituminous coal and lignite often referred to as brown coal, ~~is~~ are expected to be the predominant thermal coals in the future, because their reserves are abundant and constitute over 50% of total global reserves of coal in the world. Consequently, spontaneous combustion issues will increase with increasing usage of low rank coals, due to their high propensity to spontaneously combust. In particular, thermal drying of these coals to increase the calorific value poses an additional risk as the removal of moisture may enhance the potential for spontaneous ignition and combustion. Other factors on self-heating characteristics of coal stockpiles are wind flow or natural convection flow in the stockpile (Moghtaderi, *et al*., 2000), chemical reactions and the equivalent oxidation exposure time, that was reviewed by Sasaki and Sugai (2011), to understand the temperature behaviour of spontaneous combustion ~~of~~ in a coal stockpile.

Spontaneous combustion or self-heating of coal is a naturally-occurring process caused by many chemical reactions and oxidation of the coal matrix (Nordon, 1979). Gray and Lee (1967) measured the pre-ignition-temperatures of coal samples at different positions in a cylindrical reaction vessel. They used the heat balance model presented by Frank-Kamenetskii (1959) in considering a size of the stockpile, such as radius of the equivalent sphere. Bowes (1984) also presented simplified self-heating models based on the Frank-Kamenetskii model. However, previous studies did not show the methodologies to determine accurate Critical Self-Ignition Temperature (CSIT) and to expand CSIT for larger volume of coal stockpiles.

This paper presents the results of a study using experimental results to estimate CSIT of an actual coal stockpile from theoretical equations for scaling. In the experiments, temperature profiles at the centre of the coal pile set in a cube mesh-box was measured for different ambient air temperatures and three pile volumes of 15, 120 and 960 cm^3. Two lignite samples (one raw and one thermally dried) and one sub-bituminous coal sample were used in the experiments.

EQUATION FOR CRITICAL TEMPERATURE

Based on the Frank-Kamenetskii's model, CSIT can be formulated from heat balance at the temperature between heat generation and heat loss rates of the coal pile. Suppose the heat generation rate is

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expressed by the Arrhenius equation consisting of parameters for activation energy, *E*, frequency factor, *A*, and critical radius of the coal pile, r_c ,

$$\ln\left(\frac{\delta T_C^2}{r_C^2}\right) = \beta - \frac{E}{R}(T_C)^{-1} \quad ; \quad \beta = \ln\left(\frac{EQA\rho}{R\lambda}\right) \tag{1}$$

By plotting $\ln(\delta T_C^2/r^2)$ against $(T_C)^{-1}$, β and activation energy, *E*, can be obtained from the intercept and slope of the linear equation, respectively. Finally, heat generating rate, *Q*, and CSIT or T_C as shown in Equation (1) can be obtained as a function of r_c to predict CSIT for larger volume coal piles.

COAL SAMPLES AND EXPERIMENTAL APPARATUS

The characteristics of two lignite coals (sample #1, thermally dried lignite and sample #2, raw lignite) and one sub-bituminous coal sample (sample #3) are listed in Table 1. Their density, specific heat capacity and results of proximate analyses are quite different from each other.

Table 1 - Density, heat capacity and proximate analyses of coal samples

Sample No. and Coal Rank		Density ρ(g/m^3)	Heat Capacity *Cp* (kJ/kg/°C)	Fixed Carbon (%)	Ash (%)	Volatile Matter (%)	Moisture (%)
#1	Lignite	0.740	1.270	30.7	10.5	55.8	3.0
#2	Lignite	1.028	3.825	16.3	0.62	24.1	59.0
#3	Sub-bituminous	1.290	2.181	54.0	6.98	33.62	5.5

The coal samples were crushed into the size range of 0.25 to 0.75 mm. The coal piles were formed by filling them in the three different sized cube boxes (25, 50 and 100 mm) composed of one open top and five screened (270 mesh) faces so that air can permeate into the coal pile. Their net stock volumes were 15, 120 and 960 cm^3, respectively.

As shown in Figure 1, the cube mesh-boxes were set in the constant temperature chamber where ambient air temperature was controlled to be a constant temperature ranging from 50 to 150 ºC. Temperature sensors (thermocouple) were used to detect internal temperatures at 2 to 5 positions including the centre of the coal pile. They were installed from the open top into the coal pile.

The coal was loaded into the constant temperature chamber at room temperature (around 23°C). Each of the coal heating experiments using the cube mesh-box was done with separate constant temperature conditions to establish the self-heating characteristics of coal samples to ignition if it occurred.

MEASUREMENT RESULTS AND DISCUSSIONS

Prior to measurements of CSIT, apparent density and thermal conductivity of the coal pile, λ, was evaluated. The samples have significantly different densities and heat capacities as shown in Table 1.

Figure 2 shows the difference in temperature profiles between the three samples after sitting in the hot ambient-air of 140ºC. During the first 20 minutes from the start of the test, two samples #1 and #3 indicated similar temperature profiles, while sample #2 showed a smaller temperature rise rate than the others. The reason for this difference can be attributed to sample #2 having the highest moisture content and heat capacity of the three coal samples. After this initial stage of the heat conduction process, sample #2 showed a gradual temperature rise to reach a maximum temperature of 150 ºC after 300 min, before decreasing to the set ambient air temperature. In contrast, samples #1 and #2 showed self-heating curves reaching ignition. Thus, it is clear that the CSIT value of samples #1 and #3 is lower than 140 ºC, while that of sample #2 is over 140 ºC.

Sample #1 had the shortest time to reach ignition. As such it is used to show the process of obtaining the CSIT value for the coal and the subsequent kinetic analysis derived from this value. Figure 3 shows test results for Sample #1 at different ambient air temperatures, to find the critical point of self-ignition. In this case the value is determined to be 123 ºC (396 ºK).

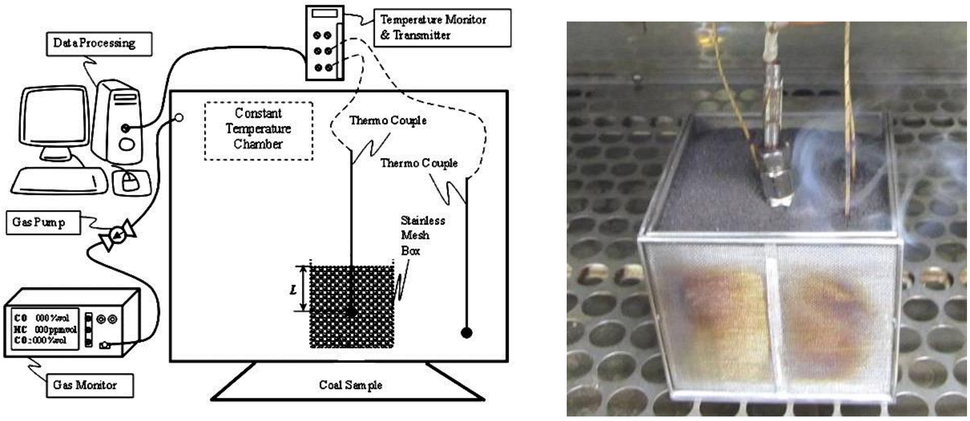

Figure 1 - Schematic figure and a photo of experimental apparatus

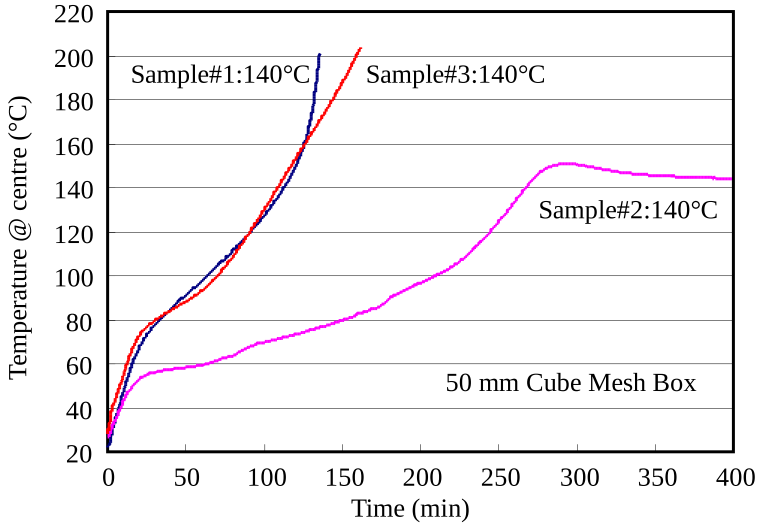

Figure 2 - Examples of temperature profiles at centre of 50 mm cube mesh-box for 140°C ambient air)

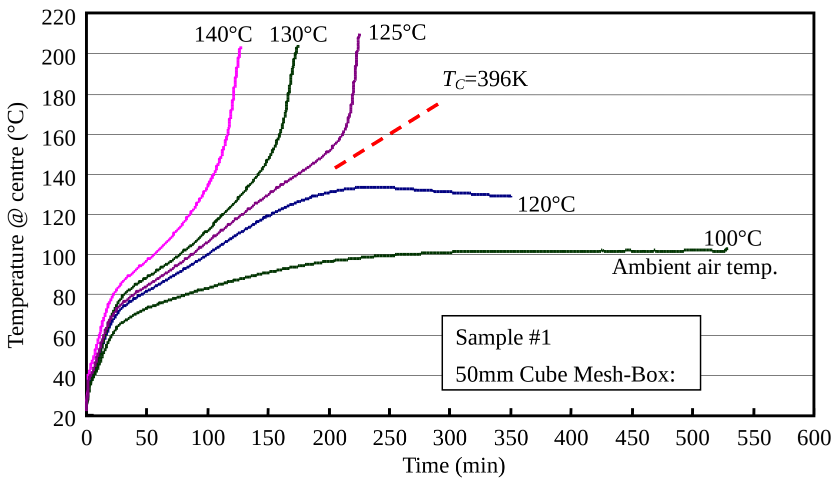

Figure 3 - Examples of temperature profiles for sample #1 at different ambient air temperatures at centre of 50mm cube mesh-box)

The data for the different mesh-box sizes are shown in Figure 4. Sample #1 shows a distinct linear relationship exists between CSIT and mesh-box size. The measured CSIT was lower for the large coal pile size and higher for the small coal pile size. This data can therefore be used to examine CSIT values for larger stockpile volumes.

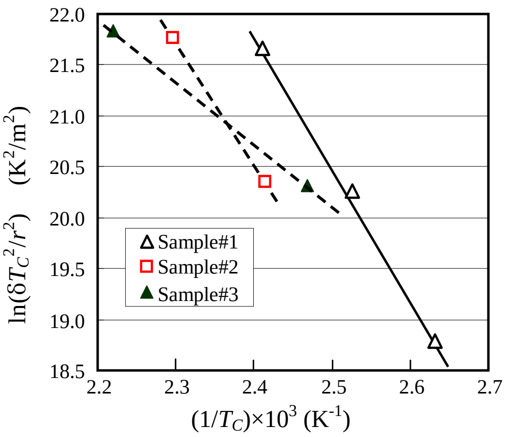

Figure 4 - Plotting ln($\delta T_C^2/r^2$) against $(T_C)^{-1}$ based on critical self-ignition temperature (CSIT), showing results for different mesh-box sizes

EVALUATION OF CRITICAL SELF IGNITION-TEMPERATURE FOR STOCKPILE VOLUMES

By using actual data from the experimental heating test results direct knowledge of the individual values of molar heat of reaction, apparent activation energy, frequency factor, thermal conductivity or specific heat of the coal samples is not needed, so far as the calculation of CSIT is concerned. By analysing the coal inside temperature and application of Fourier equation boundary conditions the self-heating data of the coal sample can be calculated. For example, the data for sample #1 in Figure 4 produces the linear relationship shown in Equation (2) and the slope of the line is equivalent to -*E*/*R*, which yields the activation energy for the coal as shown in Equation (3).

$$\ln\left(\frac{\delta T_C^2}{r_C^2}\right) = 52.8 - 1.29\times10^4\,\frac{1}{T_C} \quad (2)$$

$$E = 107.4\,\text{kJ/mol} \quad (3)$$

The relation of (T_C -273 = -7.0·ln(V) + 65], between CSIT (=T_C – 273) (°C) and stockpile volume V has been obtained from Equations (2) and (3) for stockpile volumes extrapolated over 0.1 m^3. Figure 5 shows this relationship. The CSIT decreases with stockpile volume, while the elapsed time to get to CSIT increases with stockpile volume. Thus, preserving a large volume of coal stock without upsetting inside and outside has a large possibility inducing spontaneous combustion but takes longer elapsed time to get it.

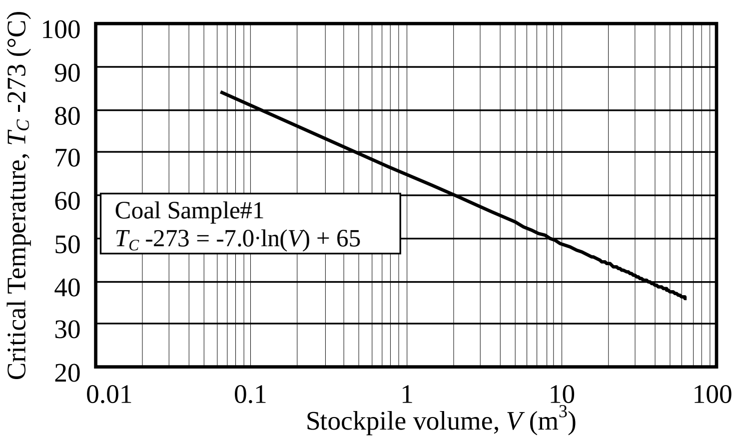

Figure 5 - A typical estimation of critical self-ignition temperature (CSIT) vs. corresponding to stockpile volume (Sample #1)

CONCLUSIONS

The Critical Self-Ignition Temperature (CSIT) of coal is important to evaluate the possibility of spontaneous combustion of coal stockpiles. In this study, the profiles of temperature at the centre of a coal pile were measured for different ambient air temperatures from 50 to 140 ºC, to calculate CSIT values for coal piles in cube mesh-boxes 25, 50, and 100 mm in length. Based on the Frank-Kamenetskii model, CSIT as function of pile size or volume was determined. Finally, the function of critical temperature has been applied to predict CSIT of larger stockpile volumes less than 50 m^3.

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