The spontaneous combustion tendency of blended coal

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Abstract – The spontaneous combustion behaviour of several coals and of their blend was studied with the crossing point method. Particular attention is given to the effect of particle size and the influence of the lower rank coal in the blend on the kinetic parameters of oxidation. It is found that the particle size greatly affects the reactivities, the activation energy of the overall reaction, and consequently the critical ambient temperature for the ignition of the coal. The temperature histories and plots of dT/dt for the blends reflect those of the individual components. The potential for spontaneous combustion of blended coal is greater following the increased percentage of the more reactive coal. Experimental findings are discussed and the key parameters determining the spontaneous combustion propensity are indicated.

Keyword : spontaneous combustion, coal blends, effect of particle size.

1. Introduction

Spontaneous combustion of stored coals cause a serious problem for coal producers and users⁽¹⁾. Improvements to low rank coal are made by either thermal drying or through blend with higher rank coals. This is of significant industrial interest. Thermal drying of moist lower rank coals could increase the calorific value of a product whilst blending of coals of different types offers a greater flexibility and economic benefit. However, the problem of spontaneous combustion assumes even greater significance, since the removal of moisture can enhance the potential for spontaneous ignition and combustion. The risk of spontaneous combustion is also made greater during blending and when storage of such lower rank coals takes place. This is particularly the case with low sulphur sub-bituminous coals which are now used to meet emission limits.

The primary source of heat generation within coal stockpiles is the exothermic low-temperature oxidation reaction⁽¹⁻³⁾, while mass and heat transport play a major role in determining the magnitude of the temperature rise in a given situation. Despite the extensive previous works on spontaneous ignition of coal using various techniques⁽⁴⁻⁶⁾, the effect of particle size in the case of single type coals on the rate of low temperature oxidation, remains controversial and poorly understood.

The spontaneous combustion propensity of coals of different rank is studied using an oven heating test where a basket of the coal sample is heated at a constant oven temperature ranging from 350 - 500 K. The crossing point method (reported in the 27^{th} International Symposium on Combustion⁽¹⁾) is used to establish kinetic rate parameters and in particular to trace the effect of particle size and coal blending on the critical ambient temperature.

Results of the self-heating test of a single coal using the new method confirmed the value of the new method compared to the classical Frank-Kamanetskii approach $^{(1-3)}$. The crossing point method postulates that when the profile of temperatures at centre part of the basket is locally flat, the one-dimensional energy equation can be simplified without the diffusion term as follows:

$$\rho_{s}C_{ps}\frac{\partial T}{\partial t} \approx \rho_{s}AQ\exp\left(\frac{E_{a}}{RT}\right)$$
(1)

thus

$$\ln\left[\frac{dT}{dt}\right]_{T_c} = \ln\left(\frac{QA}{C_{ps}}\right) - \left(\frac{E_a}{RT_c}\right)$$
(2)

where ρ_s is the bulk density (kgm⁻³), C_{ps} is the specific heat capacity (J kg⁻¹ K⁻¹), A is reactivity (s⁻¹), Q is the heat of reaction (J kg⁻¹), E_a is activation energy (Jmol⁻¹), R is the Universal gas constant (J K⁻¹ mol⁻¹), t is time(s), and T_c is the crossing point temperature (K). The kinetic plot for the determination of the kinetic oxidation parameters is deduced from the transient data (T_c and dT/dt at T_c) of the sample inside the basket. Therefore the

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repetitive process and the uncertainty of the oven temperature which may occur in the classical approach is avoided. The reader is referred to the earlier paper⁽¹⁾ and that of Chong *et. al.*⁽²⁾ to see the justification of this method. The aim of this paper is to show the effect of particle size and blending on the activation energy and other kinetic parameters of mixed coals.

2. Experimental

Three Indonesian coals, the South Bangko coal (lower rank coal), the Pinang and Prima coals (higher rank coals) were used in this work. The as-received coal samples were ground and sieved into samples of average particle diameter $d_p = 0.18$ mm, $d_p = 0.68$ mm, $d_p = 1.5$ mm. The samples were then air-dried for more than 24 hours. Proximate and elemental analysis of these coals are given in Table 1. All experiments were carried out using a 50 mm basket and the detailed experimental set-up and procedure were given in our previous work ⁽¹⁾.

	Coal types (location)		
Properties	South Bangko	Prima	Pinang
	(South Sumatera)	(East Kalimantan)	(East Kalimantan)
Calorific value (MJ/kg)	23	32	29
Density (kg m ⁻³)	635	725	725
Proximate analysis (wt %)			
Moisture :	14.2	5.2	8.8
Volatile matter :	43.7	46.8	39.2
Fixed carbon :	37.5	49.1	47.5
Ash :	4.6	2.9	4.5
Ultimate analysis (daf. wt %)			
Nitrogen :	1.25	1.76	1.55
Carbon :	62.54	76.78	72.02
Hydrogen :	4.20	5.50	4.99
Oxygen (by difference):	32.01	15.96	21.44
Combustible sulphur:	0.39	0.30	0.24

Table 1 Properties of the coal samples

3. Results and Discussion

Effect of particle size for a single coal

The rate of reaction between coal and oxygen is affected mainly by two factors: one involves the chemistry such as carbon type, active sites, etc., and the other one covers physical characteristics, such as the specific surface area, pore or surface diffusion, etc. For different coals, the physical properties differ, so that E_a and A certainly change with the type of coal. There is evidence that activation energy E_a depends on the chemical structure of the coal and the individual particle temperature. The reactivity A, which represents the effective number of collisions between carbon and oxygen molecules, depends heavily on the coal's properties⁽⁷⁾. Thus, E_a and A are affected by both chemical and physical factors. The aim of this work, referred to in the introduction, is to show experimentally the effect of the physical structure (such as particle size and surface area) on E_a and A under self-heating conditions.

The crossing point method provides fast assessment of kinetic parameters of oxidation – activation energy E_a (kJ mol⁻¹), reactivity A (s⁻¹) and critical ambient temperature $T_{a \, crit}$ (K) for coals. Results of the tests for a single coal – the Pinang coal (Indonesia) for different particle sizes are given in Fig. 1A, where ln (dT/dt) at T_c is plotted as a function of $1/T_c$. From the regressions it is clear that the effect of particle size on A is more significant than that on E_a . The increase of reactivity and activation energy with the decrease of particle size is probably due to the larger surface area for smaller coal particles.

These observations are the consequence of the interplay between particle size, activation energy, coal reactivity and critical ambient temperature and can be examined qualitatively through the heat release rate q (W m⁻³) as shown as the right-hand side in Equation (1). For smaller particle sizes the coal becomes much more reactive (higher A), although the activation energy is also higher. By setting q at the same level and solving for $T_{\rm a \, crit}$, it is shown that to have an equal q, the smaller particle size requires a lower $T_{\rm a \, crit}$. This argument is confirmed in Fig. 1B, where particle size gives a trade-off effect on coal oxidation by decreasing A but increasing $T_{\rm a \, crit}$. For smaller particles, the rate of heat generation is higher for two reasons. First, the effectiveness factor is closer to 1, leading to greater oxidation rates per unit volume. Second, the mass transfer coefficient increases with the decrease in particle size and, in conjunction with higher specific surface area, leads to greater rates of deposition. The higher effectiveness factors and mass transfer coefficients for smaller particles are reflected by the higher reactivities. At a low temperature of this experimental set-up (say 375K) the smaller particles generate more heat (q) as a consequence of higher reactivities. However, a decrease in particle size also increases the heat loss due to convection, since the heat transfer coefficient increases with decrease in particle size as does the specific surface area (^{5,6)}.



Fig. 1 (A) Plot of ln (d*T*/dt) $_{Tc}$ against (1/ T_c) for Pinang coal, $d_p = 0.18$ mm, 0.68 mm, 1.50 mm and (B) plot of reactivities and critical ambient temperatures at different particle sizes. Density of the bed is constant at $\rho = 725$ kgm⁻³. Reactivity *A* is calculated using the intercept and the heat capacity C_{ps} is 1260 Jkg⁻¹K⁻¹ (4).

Effect of coal blending

The significance of particle size effects on the spontaneous combustibility of coals is not only apparent for the case of hard coals such as Pinang coal as mentioned earlier, but also for lower rank coals as indicated in our on going study⁽⁸⁾. Therefore, in attempts to investigate the effect of coal blending, it is extremely important to choose an appropriate particle size of the coals. In this study an average particle diameter of 1.5 mm was used as at $d_p = 1.5$ mm the reactivity, the activation energy and the critical ambient temperature of the lower rank coal (e.g. the South Bangko coal $A = 2 \times 10^5$ s⁻¹, $E_a = 89 \pm 3$ kJ mol⁻¹, $T_{a \text{ crit}} = 402$ K) are very different to those of higher rank coals (such as the Prima coal $A = 3.5 \times 10^{-1}$ s⁻¹, $E_a = 51 \pm 3$ kJ mol⁻¹, $T_{a \text{ crit}} = 470$ K and the Pinang coal A = 1.5 s⁻¹, $E_a = 53 \pm 2$ kJ mol⁻¹, $T_{a \text{ crit}} = 425$ K).

Results of the oven heating tests of the individual coal and of the related blends show that the temperature-time (measured at the centre of the samples) of the latter reflect those of the single coals (Fig. 2A). In fact, the curve representing the corresponding dT/dt of a blend is located in between those pertaining to the two individual components of the blend. For a 1:1 blend, the curve closely follows the mid points of the parent coals (Fig. 2B).

Fig. 3A and 3B show the activation energies and reactivities obtained for blends of Prima and Pinang coals with South Bangko coal. Both activation energies and reactivities increase with the percentage of the more reactive coal (the South Bangko coal) in the blend. It is clear that the increase of E_a is not linear and Fig 3B shows that the reactivities increase almost exponentially (notice log scale on vertical axis). Therefore, in terms of the heat release rate q, the influence of the reactivities are more prominent than the activation energies (compare Fig. 3B – graph of log A to Fig. 3A – graph of E_a).

The presence of South Bangko coal also decreases the critical ambient temperatures as shown in Fig. 3B. Meanwhile, higher reactivities lead to a sharper increase in particle temperature and this shortens the ignition delay time⁽⁶⁾. These compounding factors suggest that the presence of a more reactive coal enhances the potential for spontaneous combustion of blended coals by increasing the rate of heat generation and lowering the critical ambient temperature.



Fig. 2 Comparison of temperature-time curve (A) and dT/dt curve (B) obtained with South Bangko coal, Pinang coal and their blend at ambient temperature, $T_0 = 122^\circ$ C and $d_p = 1.5$ mm.



Fig. 3 Activation energies of the blended coals (A) and (B) reactivities and critical ambient temperatures at different blending ratio. Particle diameter $d_p = 1.5$ mm.

4. Conclusions

Early work on the crossing point method has been extended to investigate the effect of particle size and of blending on the spontaneous combustibility of coal. It is found that the activation energies and reactivities of coal decrease with increasing particle size. However the critical ambient temperature for spontaneous combustion to occur behaves inversely as it increases with particle size. The potential for spontaneous combustion of blended coal is greater due to higher reactivities and activation energies, coupled with lower critical ambient temperatures following the increased percentage of the more reactive coal.

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