

Influence of heating rate and particle size on thermo-swelling properties of heating coal

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Abstract

Two bituminous coal samples with particle size less than 212 μm were heated at three different heating rates 3, 5 and 10 $^{\circ}\text{C}/\text{min}$ under argon atmosphere to investigate the effect of heating rate on swelling, the apparent specific heat and thermal conductivity. Additional studies were undertaken using one sample with a larger particle size of less than 500 μm was pyrolysed at a heating rate of 10 $^{\circ}\text{C}/\text{min}$ to explore the effect of particle size on swelling and thermal properties. The pyrolytic experiments were conducted using novel Computer Aided Thermal Analysis (CATA) technique. The results indicated that the swelling increased with increasing heating rate and particle size in the temperatures between 400 and 600 $^{\circ}\text{C}$. The temperature at which rapid swelling commenced during pyrolysis ascended with increasing heating rate but descended with increasing particle size. The apparent specific heat indicated that exothermic heats of reaction, during the primary devolatilization, increased with heating rate and particle size. Thermal conductivities were almost constant prior to the primary devolatilization for all experiments but underwent significant increase during the primary devolatilization depending on the apparent fluidity.

Keywords: Heating rate, Particle size, Swelling, Specific heat, Thermal conductivity

1. Introduction

During heating under inert conditions, coking coal undergoes transient changes in volume swelling, specific heat and thermal conductivity. The swelling/shrinkage progress history of heated coal is strongly associated with a number of consequent chemical reactions that were determined by coal properties and heating conditions such as heating rate and particle size. It is well established that a rise in heating rate may shift the maximum rate of gas evolution to higher temperature range, raise the softening and solidification temperature, result in a more extensive thermal fragmentation of coal's molecule structure, lower the minimum viscosity, enhance the plastic properties of the coals and suppress the secondary reactions and the loss of fixed carbon [1-4]. In case of the effect of particle size on heating coal, it is also well known that small particles can increase softening temperature, decrease plastic temperature range [4] and lead to a higher extent of secondary reactions, although it did not cause significant difference in total volatile yield during carbonization [5].

A number of techniques have been employed to investigate the specific heat and thermal conductivity, for instance, Differential Scanning Calorimetry (DSC) [6] or Differential Thermal Analysis (DTA) [7] can describe the endothermic or exothermic reactions zones, but these techniques lack of resolution, sensitivity, temperature range or a combination of each. Strezov et al. [8] have

recently developed an advanced Computer Aided Thermal Analysis (CATA) technique for simultaneous measurement of the apparent specific heat and thermal conductivity of heated coal, which resolves most of the disadvantages of the conventional thermo-analytical techniques discussed above, is capable of accurately quantifying the energy requirements for a solid fuel and the heats of coal pyrolysis [9]. In this study, the objective is to simultaneously explore the effect of heating rate and particle size on swelling and heats of reactions as well as thermal conductivity during coking. A novel CATA technique will be modified by introducing a Linear Velocity Displacement Transducer (LVDT), which is capable of measuring the swelling/shrinkage behaviour during a coal plastic transition, to the existing CATA technique to simultaneously evaluate swelling and thermal properties of heated coal.

2. Experimental

2.1 Sample

Two bituminous coal samples in this study were selected for simultaneously investigating the effect of heating rate and particle size on swelling and thermal properties of heating coal. Coal properties analysis was shown in Table 1. Sample C was grounded to -212 μm and -500 μm , and sample E was grounded to -212 μm for investigating the effect of heating rate and particle size on swelling and thermal properties.

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Table.1. Coal samples analysis

Proximate analysis (ad)	C	E
Fixed carbon (%)	72.6	61.6
Volatile matter (%)	19.1	27.2
Ash (% ad)	6.9	8.3
Air dried moisture (%)	1.4	2.9
Petrographic analysis		
Romax (%)	1.46	0.98
Vitrinite (mmf %)	52.4	69.1
Liptinite (mmf %)		1.5
Gieseler Plastometer (AS 1038.12.4.1)		
Crucible Swelling Number		8.0
Initial Softening Temperature, °C	435	420
Maximum Fluidity Temperature, °C	470	450
Maximum Fluidity, ddpn	45	60
Solidification Temperature, °C	495	480
Plastic Range, °C	60	60
Dilatation (AS 10.38.12.3)		
Initial Softening Temperature, °C		385
Maximum Contraction Temperature, °C	455	435
Maximum Dilatation Temperature, °C	490	460
Maximum Contraction, %	25	18
Maximum Dilatation, %	23	8

2.2 Swelling and thermal property measurement

The experimental setup uses the principle of the Computer Aided Thermal Analysis (CATA) technique which has been used in literature [8] and specifically modified in this work to measure volumetric change in the sample along with the temperature profile. The heating chamber is shown in Fig. 1. The sample, approximately 2.8 g, in mass was packed in a quartz tube to the length of 30 mm with a diameter of 11.80 mm. The pyrolytic experiments were investigated from room temperature to 1000 °C under an inert atmosphere with a flow of 30 ml/min. The temperatures of the graphite heating element, surface and centre of the samples were measured for calculating the apparent specific heat and thermal conductivity. The transient swelling/shrinkage of sample was recorded on a linear velocity displacement transducer (LVDT).

2.3 Calculation of swelling of samples

The coal bed was restrained on one side by the alumina rod and allowed to expand on the other side through a Linear Velocity Displacement Transducer (LVDT). The LVDT readings as well as temperatures were monitored each second. All experiments were started under same compression of the well calibrated spring. The swelling of samples was determined by the transient value of the LVDT measured during heating. It can be calculated by the transient compressed value ΔL of the spring and the original length L_0 of the sample,

$$\text{Swelling} = \Delta L / L_0 \times 100\%.$$

The calculation of high temperature contraction is based on the length of heating coal measured by LVDT at the maximum swelling and the pyrolysis terminal temperature of 1000 °C.

3. Results and Discussion

3.1 Effect of heating rate on swelling

Figs. 2-3 showed that higher heating rate can shift the initial swelling to higher temperature and increase the swelling at the primary devolatilization. This may be related to the increase in temperature of maximum fusion and the maximum extent to fusion of heating coal with increasing heating rate [10]. An increase in the heating rate leads to an increase in the heat flow, so that more coal becomes plastic per unit time and the plastic layer becomes thicker [11]. The fluidity of plastic mass and the maximum thickness of plastic layer therefore increase with increasing heating rate [10, 11]. During the secondary devolatilization, coal sample C showed a large contraction of 50.7 % from 650 °C to 1000 °C at a heating rate of 10 °C/min, although this contraction was not observed at the heating rates of 3 and 5 °C/min. Marzec et al. [12] suggested that the contraction depended on two factors: (1) the weight percent of coal material converted to the high molecular weight decomposition products on heating coal up to the temperature of resolidification, and (2) the composition of the decomposed products.

3.2 Effect of heating rate on the apparent specific heat and thermal conductivity

Figs.4-5 showed the apparent specific heat for two coal samples at the heating rates of 3, 5 and 10. Coal sample C exhibited the same specific heat at about 350 °C and 200 °C for coal sample E with various heating rates. Over this point, the apparent specific heat slowly increased with temperature until the primary devolatilization. The endothermic peaks related to the primary devolatilization appeared at higher temperature in the temperature range of 420 to 470 °C with increasing heating rate, as in particular shown in coal sample E. The maximum exothermic troughs for both two samples were observed at the highest heating rate of 10 °C/min at the primary devolatilization from 400 and 600 °C, which corresponded to the swelling progress of heating coal at various heating rates. The greatest different in the specific heat with various heating rates was coal sample C, there

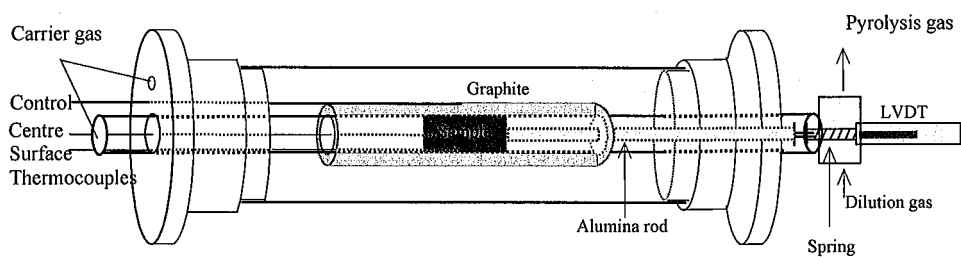


Fig.1. Schematic of the experimental setup with spring loaded on LVDT

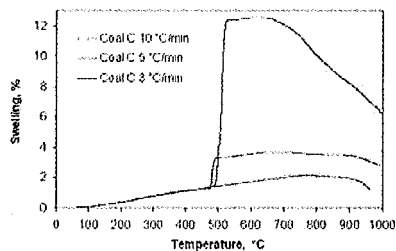


Fig.2. Effect of heating rate on swelling of coal C

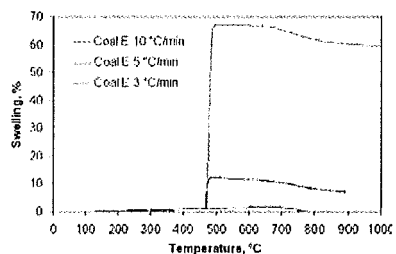


Fig.3. Effect of heating rate on swelling of coal E

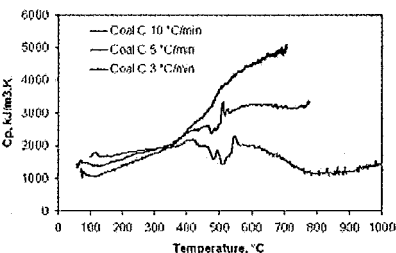


Fig.4. Effect of heating rate on the apparent Cp of coal C

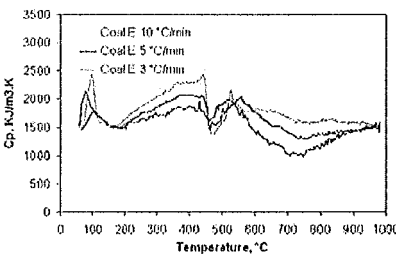


Fig.5. Effect of heating rate on the apparent Cp of coal E

was even no exothermic reactions trough observed at a heating rate of 3 °C/min during the primary devolatilization corresponding with its little increase in swelling at this stage. Figs. 6-7 showed the thermal conductivities for three different heating rates were almost constant until reaching the devolatilization

point for all experiments. To combine the results from all experiments, it can be observed that the thermal conductivity showed a peak at a heating rate of 10 °C/min during the primary devolatilization but this phenomenon was not observed at 3 and 5 °C/min, which is related to the development of fluidity.

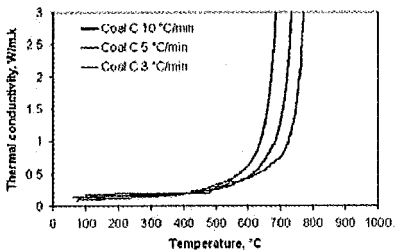


Fig.6. Effect of heating rate on thermal conductivity of coal C

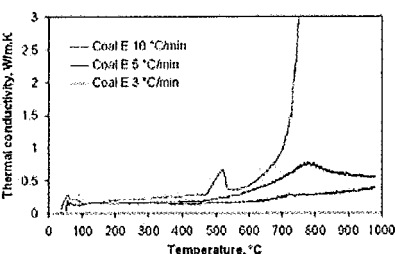


Fig.7. Effect of heating rate on thermal conductivity of coal E

3.3 Effect of particle size on swelling and thermal properties

Fig. 8 showed the initial temperatures as rapid swelling occurred were 471 °C for -500 µm particles and 481 °C for -212 µm particles, respectively. But the temperatures at which maximum swelling was observed for two different particles samples both were 512 °C, indicating large particles finished the primary swelling at a wider temperature range. The maximum swelling for large particles was 74.4 % comparing to only 11.8 % for small particles at the primary devolatilization. A possible explanation for this is that a large particle size increases a particle volume/surface area ratio, which favors the coalescent of coal particles and restricts the release of volatiles.

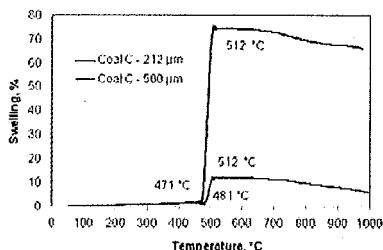


Fig.8. Effect of particles size on swelling for coal C

Figs. 9-10 presented the variations of the apparent specific heat and thermal conductivity with particles size, respectively. For temperatures up to the decomposition point the specific heat showed similar values, apart from the endothermic release of the moisture. The temperature at which the apparent specific heat commenced to decrease due to thermal devolatilization for small particles was higher than that of large particles but almost same temperatures for the first reactions end were observed. This shift of temperature in exothermic reactions for different particles was quite consistent with that in case of swelling in Fig.8. Large particles that showed the swelling to a higher extent at the first devolatilization showed a larger exothermic trough. The thermal conductivities for heating coal with two different particles both increased from 0.20 W/m. K at 480 °C to 0.38 W/m. K at 545 °C due to the increase of fluidity. A slight decrease in thermal conductivity occurred between 545 and 565 °C due to the decrease of fluidity by tar vaporization and volume swelling. The extent of thermal conductivity decrease for large particles was higher than in case of small particles. The reason is that large particles resulted in an increase in pores of semi-coke due to large swelling, and the increase of pores caused a high extent of decrease in fluidity. The increase of thermal conductivities from 565 to 700 °C is attributed to the combination of fluidity, micropores and change in intrinsic conductivity of heating samples. Over 700 °C, the temperatures between sample surface and centre were in a very narrow range, hence the rapid increase of thermal conductivity was observed.

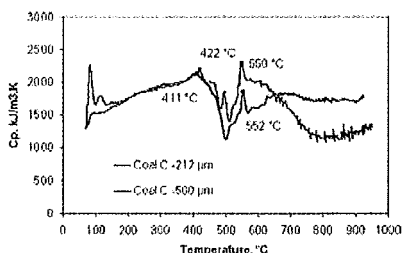


Fig.9. Effect of particle size on apparent Cp for coal C

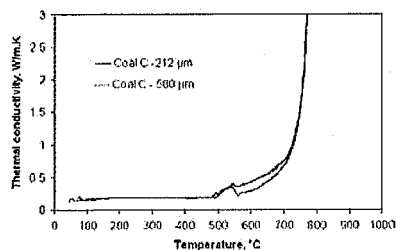


Fig.10. Effect of particle size on thermal conductivity for coal C

4. Conclusion

1. During the primary devolatilization between 400 and 600 °C, higher heating rate can shift the initial endo/exothermic reactions and the initial swelling to higher temperature, increase the exothermic trough and the maximum swelling as well as the thermal conductivity. During the secondary devolatilization between 600 and 1000 °C, higher heating rate can shift the initial endothermic reactions and the initial rapid increase in thermal conductivity to higher temperature.
2. Large particles commenced the exothermic reactions and swelling at lower temperature but finished this stage at almost the same temperature with small particles. Maximum swelling and exothermic trough increased with increasing particles size during the primary devolatilization. Small particles had larger exothermic trough and showed greater contraction at the secondary devolatilization than large particles.

5. References

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