MECHANISM OF SINGLE COAL PARTICLE IGNITION UNDER MICROGRAVITY CONDITION

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In order to simplify the complex coal ignition process, a microgravity condition (10^{-3} g), in which natural convection is negligible, has been employed to observe ignition of single coal particles. The particle was heated and ignited by radiation from spot heaters. Three coals with different volatile matter contents were employed, having particle sizes in a range of 0.8 mm to 1.2 mm.

As expected, under microgravity, a nearly spherical volatile cloud was found to form around the particle. Results clearly show that regardless of the particle size, the ignition is preceded by homogeneous gas-phase volatile ignition, followed by heterogeneous solid-phase char ignition after burn-out of the volatiles. The heterogeneous ignition temperature and time were higher and longer, respectively, under microgravity than under normal gravity condition. The homogeneous ignition also took place at a higher particle temperature under microgravity than under normal gravity. Further, the temperature for heterogeneous ignition increased with the absolute amount of volatiles contained in a coal particle, whereas the particle temperature for the homogeneous ignition decreased.

Introduction

When heated in air up to temperatures higher than ca. 600 K, coal separates into two major product components, *i.e.*, gaseous volatiles and solid char particles, due to the pyrolysis of carbonaceous constituents. Since both components are combustible, either the homogeneous ignition of volatiles or heterogeneous ignition of the char follows the pyrolysis in the course of further heating to higher temperatures. The amount of volatiles released from coal depends on operating variables such as heating rate and temperature as well as coal nature. Moreover, a portion of the volatiles remaining around a char particle is significantly affected by natural and/or forced convections. Thus, it is at present difficult to predict whether ignition of a coal particle commences homogeneously or heterogeneously and to define the ignition temperature as a coal property.

Ignition of single coal particles has so far been widely studied under normal gravity conditions. One of the difficulties in most of the experimental techniques has been how to support the particle. The most frequently used technique of dropping a coal particle into a hot gas results in relative motion between the particle and the surrounding gas. This enhances the separation of volatiles from the vicinity

of particle. The case is also the same when a particle is suspended in an upward flowing gas (Chen *et al.*, 1994). By direct attachment of a particle onto the temperature measuring device (Tomeczek and Wojcik, 1990; Katalambula *et al.*, 1996), the volatiles sweeping-away effect is only confined to that caused by natural convection. However, the technique becomes inapplicable to fine particles. Recent studies on the levitation of char particles (Wong *et al.*, 1995; Cozzani *et al.*, 1995) showed promising results but so far this technique has only been applied to char particles which produce little or no volatiles. Other shortcomings of the levitation technique are that particles lose charge at higher temperatures during heating thus shift from the focus point. In the case of the electrodynamic balance (EDB) (Cozzani *et al.*, 1995), particles do not ignite in air or in the presence of oxygen at lower concentrations. Thus experiments have to be performed in pure oxygen, showing a departure from reality.

Experimental results which seem more or less to depend on the techniques used have been reported relating to the mechanism of coal particle ignition. According to Chen *et al.* (1994), the ignition mechanism depends not only on coal nature but also on oxygen concentration. The volatile matter content of coal, flammability of the volatiles and their transport from the particle determines by which mechanism a coal particle may ignite (Du and Annamalai, 1994). Karcz *et al.* (1980) reported that

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coal particles ignite heterogeneously for smaller particles and homogeneously for larger ones, which was later confirmed by Du and Annamalai (1994). On the other hand, Solomon *et al.* (1990) indicated that particles started to burn heterogeneously at their surface, which was followed by secondary ignition. A major reason leading to these different results is that different amounts of volatiles remains in the vicinity of the char particle after some have been swept away by natural and/or forced convections depending on the employed techniques.

On theoretical analyses, numerous models have been developed for the ignition of single coal particles. They assume symmetrical distribution of volatiles around the particle or negligible natural convection (Gururajan *et al.*, 1990; Yang and Wang, 1990; Zhang and Wall, 1993; Phuoc *et al.*, 1993; Du and Annamalai, 1994). The assumption may be valid for some particular cases, but actual situations are quite different in experiments under normal gravity conditions.

In order to get fundamental information on the ignition behavior of single coal particles, it is thus worthwhile to simplify the complicated ignition process. If natural and forced convections are eliminated, most volatiles will burn in the vicinity of the particle from which they were released and this will lead to a fair conclusion of which ignition mechanism prevails. On the basis of Grashof number, Gr ;

$$Gr = g\rho^2\beta\Delta Td_p^3/\mu^2 \quad (1)$$

the influence of natural convection is negligible for Gr of the order of 10^4 whereas natural convection significantly disturbs the flow around the particle when $Gr > 10^6$ (Dordain and Lockwood, 1987). Among the parameters contained in the equation, the thermal expansion coefficient, β , the characteristic flame temperature, ΔT , and the gas viscosity, μ , are principally related to the nature of the reactants and therefore can not be adjusted to reduce Gr . This leaves the gas density, ρ , the particle diameter, d_p , and the gravitational acceleration, g , to be adjusted. Among them, the reduction of g is an effective method to reduce Gr . At $g=0$ there is no natural convection and together with the absence of forced convection the ignition process is thoroughly governed by diffusion processes. A microgravity environment also gives rise to another avenue to evaluate accumulation of volatiles released and subsequent reactions since they are not swept away from the particle.

1. Experimental

1.1 Apparatus and Procedure

The equipment used is basically the same as employed in our previous study (Katalambula *et al.*

Fig. 1a Schematic diagram of experimental apparatus

Fig. 1b Microgravity quality as a function of time for HNIRI drop tower

1996) which used radiant energy from spot heaters to ignite a coal particle. A schematic diagram of the current equipment is shown in Fig. 1a. A single coal particle was supported on the coiled thermocouple (material Pt/PtRh 13%, wire diameter 0.1 mm, tip diameter 0.4 mm, response rate 32.5 K/ms, error < 0.01), which was also used to measure the particle temperature. In microgravity experiments, the whole system is placed in a chamber and a board computer is connected via an AD converter and a multiplexer for data acquisition. The system is activated by a zero-gravity sensor switch which turns on the delay timer D at the start of the fall. The delay timer then turns on the board computer after a given delay time expires. The delay time enables the system to transgress from normal gravity to microgravity before the initiation of particle heating. Once turned on, the board computer immediately sends a signal to the heater timer H to turn the spot heaters on, thus starting the ignition process. The heating rate of particles ranges from 770 to 800 K/s in the present experiments.

The 10 m-high drop tower at the Hokkaido National Industrial Research Institute (HNIRI) was used to create the microgravity environment. In addition, the drop tower at the Japan Microgravity Center (JAMIC) was used to provide additional

Table 1 Properties of coals used

Coal	Proximate analysis				Ultimate analysis				
	Mo ^a	Ash	V.M. ^b	F.C. ^c	C	H	O	N	S
		[wt%]					[d.a.f. ^d wt%]		
Taiheiyo	5.9	20.1	40.2	33.8	77.5	6.4	14.9	1.1	0.3
Coal valley	8.0	9.9	32.5	49.6	79.0	5.0	14.8	1.1	0.3
Datong	4.3	10.1	26.6	59.0	84.6	4.6	9.1	0.9	0.9

a: moisture, b: volatile matter, c: fixed carbon, d: dry-ash-free

information for longer experimental durations. The drop tower at HNIRI provides a microgravity period of about 1.2 s while that at JAMIC provides about 10 s. Figure 1b shows the quality of microgravity as a function of time for the HNIRI drop tower. The top curve indicates the capsule movement in the left-right (x-axis) direction and the middle and the bottom curves indicate those in the front-back (y-axis) and in the top-down (z-axis) directions, respectively. The variations in the x- and y-axes are centered around the zero value, whereas those in the z-axis direction around a value of 10^{-3} g. The total drop time is 1.41 s and the effective microgravity time is about 1.37 s leaving 0.04 s for transgressing from normal gravity to 10^{-3} g. Further, the drop time with a good quality of microgravity is about 1.2 s.

Due to the fact that a high gravitational force is exerted on the particle during its initial fall period, it has to be securely fixed at the focal point of the spot heaters to make the particle ignition possible. For this purpose, the coiled thermocouple was supported on a quartz glass rod. The particle is held in place by the top support of a 0.2 mm spring loaded wire.

The experiment is started by placing the coiled thermocouple at the focal point by adjusting the manipulator knobs. A coal particle is then placed on the thermocouple, and the top support is brought down to hold the particle in place. The front window of the chamber is then closed and the chamber is placed in the capsule ready for drop. When the capsule is released, the zero-gravity switch activates the system as described above and the particle is ignited. The capsule is retrieved back to the launching platform and the data are transferred from the board computer for processing. The data from the computer provides a temperature history for the particle and the video camera records the ignition process. All temperatures in this work refers to the coal particle surface.

1.2 Sample materials

Three different coals were used, namely Taiheiyo (Japan), Coal Valley (Canada) and Datong (China). Relevant properties of the coals are listed in Table 1. To examine the effects of the volatile matter content (V.M.) on ignition characteristics, the particle

size was fixed at 1 mm for all coals. Also, the effects of particle size were examined employing Coal Valley coal the sizes of 0.80, 1.00 and 1.20 mm.

2. Results

2.1 Ignition phenomena

From video observation, it was found that homogeneous ignition preceded heterogeneous ignition as found in our previous work under normal gravity (Katalambula *et al.*, 1996). The major difference was that most volatiles released from the particle accumulated around the particle under microgravity conditions. As a typical example, a series of still photographs are shown in Fig. 2a which were taken from video frames of a 1 mm dia. Coal Valley coal particle. In these photographs volatiles are seen in white color. Photo A indicates a very early stage of volatile release from the particle. In this particular case, initial volatiles are released in a form of jets and accumulate surrounding the particle as a cloud. As time and temperature increase, the volatile cloud keeps growing and becomes a near-spherical ball, as shown in Photo B. After some time, a yellow spot is detected somewhere within the cloud on the right-hand side, indicating volatile ignition to form a gaseous flame (Photo C). The yellow spot increases in size until finally the color of the whole cloud turns yellow (Photo D). The yellow ball disappears within a very short period, signifying burn-out of the volatiles, which is soon thereafter followed by the formation of char flame (Photo E). The char flame grows rapidly (Photo F) and then gradually shrinks as the char is burnt out.

Figure 2b quantifies variations with time, t , of the particle temperature, T_p , and the volume of the volatile cloud, V_v , or of the flame, V_f , which was calculated by approximating the cloud shape as being spherical. Here, the points A to E correspond to the photograph numbers in Fig. 2a. It is seen that the volatile formation starts at $T_p = \text{ca. } 450 \text{ K}$ within 0.3 s after the spot heaters are turned on ($t=0$ s). At point B where the volatile ignition is observed in the photograph, no discontinuous increase is seen in both T_p and V_v . Between points B and C, V_v keeps

Fig. 2a Typical photographs of coal ignition phenomenon under microgravity conditions

Fig. 2b Variation of volatile cloud volume or flame volume and particle temperature with time under microgravity condition

increasing because more volatiles are being released from the particle as well as the gas expansion due to temperature increase. At the same time, an induction period starts in the T_p curve. At point C, V_v reaches a maximum suggesting the completion of volatile release from the particle. The cloud then shrinks slightly to point D. Then, during the rapid decrease of V_v between points D and E, a rapid increase occurs in T_p as marked by point G. This point represents the heterogeneous ignition point as defined in our previous paper (Katalambula *et al.* 1996). In the case of $d_p \geq 1$ mm, the char ignition is accompanied by the formation of char flame, which is

Fig. 3 Effect of particle temperature on volume of volatiles released from particles of different sizes under microgravity condition

represented by a small peak in the V_f curve around point F. A similar phenomenon was also observed under normal gravity conditions. The only difference is that under normal gravity, the size of the char flame remained rather constant for a while and then decreased to extinction. Also, similar results were obtained from the other coals used here.

2.2 Homogeneous ignition

Figure 3 compares the increase of V_v for Coal Valley coal particles with different diameters, d_p . It can be seen that V_v is higher for bigger particles. In addition, V_v increases with T_p but the increase is not

Fig. 4 Effect of volatile matter content on initial volatile release temperature and volatile ignition temperature under normal and microgravity conditions

Fig. 5a Effect of particle size on initial volatile release temperature and volatile ignition temperature under normal and microgravity conditions

linear as was the case of the variation with time shown in Fig. 2b. From 700 to 800 K, the rate of volatile release increases rapidly and this range coincides with the volatiles ignition temperature. This is considered to be brought about by increasing volatile release with additional heat from the burning volatiles and by thermal expansion of gas at elevated temperatures.

Figure 4 shows the effect of the volatile matter content, w_{VM} , on the temperature for the initial volatile release, T_{v0} , and the temperature for volatile ignition, T_{vi} , and compares these temperatures obtained under microgravity with those under normal gravity. T_{v0} was determined by playing back the video recording frame by frame (30 frame/s) until the initial jets or a cloud of volatiles was seen to come out of the particle. At this point, the particle temperature, T_{v0} , and the time taken, t_{v0} , were obtained from the temperature history curve. The particle temperature and the time for the volatile ignition T_{vi} and t_{vi} , respectively, were also determined by proceeding with the frame-by-frame inspection until a yellow spot was first detected within the bulk of the accumulated volatiles. It can be seen that T_{v0} is much the same

Fig. 5b Effect of particle size on time for initial volatile release and volatile ignition time under normal and microgravity conditions

Fig. 6 Effect of volatile matter content on char ignition temperature under normal and microgravity conditions

under both gravity conditions for coals with different w_{VM} 's and decreases with increasing w_{VM} . On the other hand, T_{vi} is 40 to 60 K higher under microgravity than under normal gravity for all coals and decreases with w_{VM} . The effect of d_p on T_{v0} , T_{vi} , t_{v0} and t_{vi} was examined for Coal Valley coal. Results are shown in Figs. 5a and 5b. Again, T_{v0} is almost independent of the gravity condition and slightly decreases with d_p . The difference in T_{vi} 's under the two gravity conditions is also seen for different coal sizes and T_{vi} decreases with d_p (Fig. 5a). The variations of t_{v0} and t_{vi} with d_p are as shown in Fig. 5b. The time, t_{v0} required for the initial volatile release does not depend on the gravity condition while the difference in t_{vi} 's suggests that volatile ignition takes place earlier under normal conditions than under microgravity conditions. However, the effect of d_p is negligible on t_{vi} under both gravity conditions.

2.3 Heterogeneous ignition

The temperature and time for the heterogeneous ignition, T_c and t_c , respectively, were determined from the temperature history curves. Figure 6 represents the effect of w_{VM} on T_c under both gravity conditions,

Fig. 7a Effect of particle size on char ignition temperature under normal and microgravity conditions

Fig. 7b Effect of particle size on char ignition time under normal and microgravity conditions

where T_c is shown to increase with w_{VM} . It is further noted that the rate of increase is higher under microgravity than under normal gravity and that the difference between the two conditions increases with w_{VM} . In **Figs. 7a** and **7b**, effects of d_p on T_c and t_c are shown in both gravity conditions. It is clearly seen that both T_c and t_c increase with d_p and are appreciably higher under microgravity than under normal gravity.

3. Discussion

3.1 Homogeneous ignition

As was shown in **Figs. 4** and **5a**, T_{v0} is much the same under both gravity conditions for coals with different w_{VM} , as well as for particles with different sizes. This is because factors affecting the volatile release apart from the heat supplied from an external source are chemical reactions within the particle itself. External factors such as presence or absence of natural convection around the particle would have no appreciable effect. However, it is seen that T_{v0} decreases with increasing w_{VM} , suggesting that a coal particle with higher w_{VM} releases volatiles more readily than the one with lower w_{VM} . This observed

trend agrees with Howard (1981), who reported that the temperature at which a given fraction of the final weight loss is achieved decreases with an increase in w_{VM} . In the case of the present experiment, the weight loss of coal particles is mainly due to the initial volatile release, and the fraction of the final weight loss should be almost the same for particles of the same size. Also, T_{vi} is higher under microgravity than under normal gravity conditions for all coals and decreases with increasing w_{VM} . These results can be explained by a plausible fact that, under microgravity, volatiles released from the coal particle tend to displace oxygen from the vicinity of the particle thus creating an oxygen concentration gradient between the surfaces of the cloud and the particle. At the same time, a temperature gradient would be produced in the opposite direction from the particle to the cloud surface since the ambient is at lower temperature. As time elapses, oxygen diffuses towards the particle surface and temperature keeps rising until a point is reached when a flammable mixture is formed at a certain location within the cloud. If the temperature at the location is high enough to ignite the mixture, the volatiles are ignited. During the oxygen diffusion process, the particle temperature continues to rise and becomes fairly high when the volatile ignition takes place. Under normal gravity, volatiles also tend to displace oxygen from the vicinity of the particle but natural convection sweeps some of them away and supplies oxygen. This leaves only a thin layer of volatiles surrounding the particle, which would ignite much earlier upon forming a flammable mixture and hence at lower temperature. Natural convection may also slightly cool the particle under normal gravity.

The decrease of T_{vi} with increasing d_p , depicted in **Fig. 5a** under both gravity conditions can be explained by the fact that a smaller particle requires a higher temperature than a larger one to release a given amount of volatiles. If a certain amount of volatiles is required to form a flammable mixture, it will be achieved at higher temperatures for smaller particles than for larger ones.

As far as the t_{v0} is concerned (**Fig. 5b**), there is not much difference between the two gravity conditions. However, there is a remarkable difference in t_{vi} compared to that in T_{vi} . The earlier ignition under normal gravity than under microgravity is attributed to a diffusion controlled transport of oxygen to the reaction zone assisted by natural convection. Further, the negligible effect of d_p on T_{v0} under both conditions and the longer t_{vi} under microgravity are due to volatile accumulation around the particle as explained above.

3.2 Heterogeneous ignition

Dependencies of T_c on w_{VM} and d_p have been shown to differ under normal gravity and micro-

gravity conditions (Figs. 6 and 7a). The difference can be explained as follows: For a particle to ignite, all the volatiles surrounding it must ignite first so that oxygen may reach the particle surface to initiate the heterogeneous reaction. Under normal gravity, natural convection sweeps some of the released volatiles away from the vicinity of the particle, thus only a portion is left. On the contrary, under microgravity all the released volatiles remain in the vicinity of the particle. Hence, a longer period is needed for them to ignite and burn out. During this period, the particle continues to receive radiant heat from the spot heaters, and also absorbs a portion of heat from the burning volatiles thus causing heat accumulation in the particle. By the time oxygen becomes available on the particle surface, the particle temperature is already much higher, thus resulting in higher T_c . In addition, natural convection may cool the particle slightly under normal gravity but this cooling effect is eliminated under microgravity thus causing higher T_c .

As shown in Fig. 7b, t_c is longer under microgravity than under normal gravity and increases with d_p at the same rate under both gravity conditions. This is again due to the different amounts of volatiles surrounding the particle under both gravity conditions as described above. In addition, larger particles have more volatiles around them, resulting in longer t_c and higher T_c . However, as d_p increases, the difference in T_c between the two gravity conditions also increases. A similar trend was observed on the effect of w_{VM} on T_c , suggesting that it is the amount of volatiles available which affects T_c . Under microgravity, a bigger layer of volatiles accumulates around the particle, thus taking a longer time to burn as explained above.

3.3 Simplification of the phenomenon by microgravity

As briefly explained in the introduction, the use of microgravity condition enables reduction of Gr in Eq. (1) and consequently eliminates or reduces the effect of natural convection. Gr can also be reduced by decreasing the particle size, d_p , or the density difference between hot volatiles and surrounding air. In this experiment, d_p is a parameters of interest and the observed ignition behavior of a 1 mm particle under microgravity of $10^{-3} g$ would be equivalent to that of a 0.021 mm particle under normal gravity. According to the reported mechanisms of ignition under normal gravity conditions, however, such a small particle of 0.021 mm should ignite heterogeneously (Du and Annamalai, 1994; Karcz *et al.*, 1980) because it is smaller than the size of 0.40 mm which is the transition of the ignition phase from heterogeneous to homogeneous. This is apparently opposite to what would be expected on the basis of Gr that the smaller the particle size, the less the effect of natural convection. In that case, volatiles are

expected to stay in the vicinity of the particle and hence minimize chances of heterogeneous ignition. On the other hand, Du and Annamalai (1994) reported that since smaller particles have less volatiles, they stand a bigger chance of heterogeneous ignition. Both are true in certain conditions but cannot be generalized without evaluation of the effect of natural or forced convection which sweeps volatiles away from the particle and is more or less dependent on experimental conditions.

Conclusion

Ignition mechanisms of single coal particles have been studied under microgravity condition where combustion of all volatiles released from the particle was possible. Effects of the volatile matter content of coal and particle size have been investigated on the volatile release, the homogeneous volatile ignition and the heterogeneous char ignition. Within the limits of the present experimental conditions, the following conclusions have been drawn:

- 1) Combustion of a coal particle starts with homogeneous ignition, regardless of the particle size if all the volatiles are involved in the reaction. This is then followed by heterogeneous ignition after burn-out of the volatiles. The departure from this is only due to partial involvement of volatiles caused by volatile jets, and natural and/or forced convections which sweeps volatiles away from the vicinity of the particle.

- 2) The homogeneous ignition temperature and time are respectively higher and longer under microgravity than under normal gravity conditions, probably due to temperature and oxygen concentration gradients within the volatile cloud around the particle.

- 3) The heterogeneous ignition temperature and time are respectively higher and longer under microgravity than under normal gravity and increase with the volatile matter content and the particle size. The rate of increase is appreciably higher under microgravity than under normal gravity. These results are caused by significant differences in the amounts of volatiles in the vicinity of particle depending on experimental conditions as well as on coal properties.

- 4) Microgravity has no effect on the particle temperature for the initial volatile release, but increasing volatile matter content and particle size lower this temperature.

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Nomenclature

d_p	= particle diameter	[mm]
g	= gravitational acceleration	[m·s ⁻²]
Gr	= Grashof number defined by Eq. (1)	[—]
t	= time	[s]
T	= particle temperature	[K]
ΔT	= characteristic flame temperature relative to the surrounding	[K]
V	= volume	[mm ³]
w_{VM}	= volatile matter content of coal	[wt%]
β	= thermal volume expansion coefficient	[K ⁻¹]
μ	= viscosity	[kg·m ⁻¹ s ⁻¹]
ρ	= density	[kg·m ⁻³]

<Subscript>

c	= char
f	= flame
v	= volatiles
v_0	= initial volatile release
vi	= volatile ignition

<Others>

(0)	= under microgravity
(1)	= under normal gravity

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