Observation of laser ignition and combustion of pulverized coals

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Direct observation of pulsed-laser ignition and combustion of pulverized coals with a high speed video has been made. A dilute stream of particles was dropped into a laminar, upward-flow wind tunnel with a quartz test section. The gas stream was not preheated. A single pulse from a Nd:YAG laser was focused through the tunnel and ignited several particles. The transparent test section and cool walls allowed for optical detection of the reaction sequence. The ignition and combustion behaviours of amorphous carbon spheres and an anthracite were as expected for heterogeneous ignition and reaction. Surprisingly, the reaction of a high volatile bituminous and a subbituminous coal exhibited two broad emissions, as detected with a photomultiplier tube. For these coals, high speed videos revealed that two successive clouds of volatiles surrounded the particles, and each cloud was associated with one of the broad emissions. It is hypothesized that the first cloud was due to the evolution and reaction of tar, and the second resulted from low molecular weight, combustible gases. The subbituminous and bituminous coals apparently ignited heterogeneously, and possible explanations for this are discussed.

(Keywords: combustion; laser ignition; coal)

In a previous study¹ a novel experiment to study the ignition of pulverized coals was presented. By detecting the emission from the laser-ignited particles, it was concluded that the ignition and combustion of carbon spheres and an anthracite occurred heterogeneously. The emission from a high volatile (hv) bituminous coal, surprisingly, exhibited two broad peaks, and its ignition mechanism could not be determined. Here, the previous study is extended by observing the reaction sequence with high-speed video. The goals are to determine the ignition mechanism of these fuels by direct observation, and to examine the behaviour of coals containing a high volatile matter content. In addition to the three previous samples, a subbituminous coal is included.

EXPERIMENTAL

Figure 1 presents a schematic of the laser ignition experiment; the inset shows the details around the test section. Sieve-sized particles were dropped through a tube into a laminar, upward-flow wind tunnel with a quartz test section (5 cm square cross-section). The gas was not preheated. The gas flow rate was set so that the particles emerged from the feeder tube, fell approximately 3 cm, then turned and travelled upward out of the tunnel. This ensured that the particles were moving slowly downward at the ignition point, chosen to be 1.5 cm below the feeder-tube exit. It is obvious that bigger particles need a higher gas velocity in this configuration, but different coals of the same size-cut also require different gas flow

rates because of density variations. The average gas velocities used for the samples in this study (described later) are listed in *Table 1*. A single pulse from a Nd:YAG laser was focused and deflected at an angle down through the test section. The beam was defocused upon exiting the test section, and two additional prisms folded the beam back through the ignition point. For nearly every case, one to five particles were contained in the volume formed by the two intersecting beams, as determined by observation with a high speed video.

The feeder was a capped cylinder (12 mm diameter) with a tapered bottom connected to a 4 mm tube. Within the feeder a wire mesh was suspended, and supported a mound of particles. A jolt, provided by directing the exit of a pressurized solenoid valve at the feeder, caused particles to fall through the mesh and into the feeder tube. For coal particles, it was found that the optimum mesh in the feeder was two mesh sizes larger than the finest through which the particles were sized. For spherical carbon particles, the finest mesh through which the particles were sized was used.

The YAG laser operated at 5 Hz and emitted a nearly collimated beam (8 mm diameter) in the near-infrared (at a wavelength, $\lambda = 1.06 \mu\text{m}$). The laser pulse duration was 150 μs and the energy was variable up to 720 mJ per pulse, with pulse-to-pulse energy fluctuations of less than 3%. At the ignition point the beam diameter normal to its propagation direction was ~ 2.5 mm on each pass of the beam. A laser 'gate' (see *Figure 1*) permitted the passage of a single pulse to the test section. The gate was formed by cutting a wide slot in a piece of aluminum and mounting it on a controlled stepper motor. Prior to each experiment one edge of the gate blocked the laser pulses until the motor controller was triggered, causing

Figure 1 Schematic of the laser ignition experiment

Table 1 Sample analyses^a

Sample	Gas velocity (cm s ⁻¹)	Volatile matter (wt%, db)	Ash (wt%, db)	C (wt%, daf)	H (wt%, daf)	N (wt%, daf)	S (wt%, daf)	O (diff.)
Unibeads C	38.0	N.m. ^b	N.m.	96.0	1.2	1.7	N.m.	N.m.
Newlands hv bituminous	59.3	26.3	14.7	84.5	5.1	3.1	0.5	6.8
Sanxi anthracite	66.7	6.3	12.4	94.6	1.3	1.7	0.0	2.4
Skyline subbituminous	49.3	41.8	8.6	75.3	5.7	3.0	0.5	15.5

^a Analyses for coals performed on bulk, unsized sample

^b Not measured

the gate to rotate to the position shown in Figure 1. After a set dwell time, which permitted the passage of a single pulse, the controller was triggered again and rotated to block the succeeding pulses. By synchronizing both the solenoid valve and the stepper motor controller with the laser, the delay time could be controlled between the firing of the feeder and the passage of the laser pulse. This control was necessary since different samples (in coal type or size) arrived at the ignition point at different times after the feeder was fired. Heating the particles from two sides achieved more spatial uniformity and allowed for higher energy input than a single laser pass. By measuring the laser energy after each pass through the test section, it was estimated that 60 and 40% of the laser energy (measured at the laser output) arrived at the ignition point after the first and second passes, respectively.

Emission from the igniting/burning particles was detected with three photomultiplier tubes (PMT) and, concurrently, the events were recorded with a high speed

video. The emission system viewed the particles along the direction in which the laser beam propagated, and the video camera was positioned at 90° to this direction. The video system consisted of a gated, intensified camera mounted with a 105 mm macro lens and associated equipment for camera control, video control and monitor. The scene was not illuminated; the light source was simply the hot, emitting particles and volatiles. In all cases the camera's gain was set to 27% of the maximum and the gate time (the exposure duration) to 50 μs; the recording speed was 1000 frames s⁻¹. The field of view was 2 × 2 cm, giving a pixel resolution of ~100 μm square.

To align the emission system, a simple lens was used to image the common end (5.5 mm) of a trifurcated optical fibre bundle to a 25 mm circle centred about the ignition point. Since the fibre bundle consists of thousands of 100 μm glass fibres, it is possible that individual particles may be imaged onto individual fibres, leading to different legs of the trifurcated bundle. This is undesirable, since

the aim is for all three detectors to see the same emission so that two-colour pyrometry can be used to measure temperatures. To avoid this problem, the fibre bundle was moved forward 25% of the distance to the lens, as shown in *Figure 1*. Moving the bundle off the imaging point in this manner ensured that the entire bundle was uniformly illuminated by the emission from all particles. The resulting field of view for the emission system was ~ 45 mm in diameter. The light transmitted through each leg of the bundle was directed into an assembly consisting of a collimating lens and a band-pass filter, and was then detected by a PMT. The centre wavelengths, λ_c , and bandwidths, $\Delta\lambda$, of the filters were: $\lambda_c = 799$, 631 and 504 nm, and $\Delta\lambda = 11$, 10 and 20 nm, respectively. A microcomputer-based data acquisition system, with simultaneous sample-and-hold, recorded the signals at 20 kHz.

Reported here are the ignition and combustion behaviours of spherical, amorphous carbon particles and three coals. The carbon, known as Unibeads C, is a commercially available product used as column packing material for gas chromatography. It is believed to be similar to the Sphero carb particles widely used in other studies. The coals are an Australian high volatile (hv) bituminous (Newlands), a Chinese anthracite (Sanxi) and an American subbituminous (Skyline). All samples were sized to -100 to $+120$ mesh ($136 \mu\text{m}$ average) and dried at 70°C under vacuum. The proximate analyses for the coals are listed in *Table 1*, along with ultimate analyses for all samples.

RESULTS

The thermal behaviour of a carbon sphere heated by a laser pulse¹ has been previously analysed. The analysis showed that a particle can be rapidly heated by the laser pulse, but a large temperature gradient was sustained within the particle. The internal temperature equilibrated rapidly, however, because the rate of heat conduction within the particle was much higher than the rate of heat loss to the surroundings. To illustrate, for a $136 \mu\text{m}$ diameter particle uniformly irradiated by a $150 \mu\text{s}$ laser pulse with an energy density of $3.6 \times 10^4 \text{ J m}^{-2}$ (the maximum value for this experiment), the surface attained a temperature of 2000 K at the end of the pulse, while the centre was at 440 K. In this case the temperature equilibrated within the particle about $450 \mu\text{s}$ after the laser pulse ended. Furthermore, the calculations showed that the temperature gradient at the end of the laser heating period was less steep as particle size or laser energy was decreased, and the time needed for temperature equilibration decreased with decreasing particle size. The heating rate, defined as the equilibrated temperature divided by the equilibration time, was in the range of 10^6 – 10^7 K s^{-1} .

Given that the time scales for laser heating and temperature equilibration are short, relative to typical combustion times of tens of milliseconds in this experiment, the neglect of chemical reactions in this short time is justified. The particle behaviour after the temperature equilibrates throughout the particle is determined by the balance between heat generation (due to chemical reactions) and heat loss. An excess in heat generation leads to particle ignition.

In a previous study PMT traces were presented from the ignition and combustion of amorphous carbon spheres, the hv bituminous coal and the anthracite; several differences between their behaviours were also noted. These traces are shown in *Figure 2*. In all three signal traces the first short emission (corresponding to the periods of laser heating and temperature equilibration) varied from ~ 1 ms for the carbon sphere and the hv bituminous coal to ~ 3 ms for the anthracite. This difference was attributed to variations in thermal conductivity (or the prevalence of pores which decreases the effective conductivity), density or specific heat between the samples, which led to slower temperature equilibration within the anthracite particles. A second difference was the period of reaction. On average, the reaction period in pure oxygen for the carbon spheres (50–80 ms) was longer than that of the hv bituminous coal (40–70 ms), which in turn was longer than that of the anthracite (15–30 ms). The final difference in the ignition and combustion behaviours was the temporal emission profiles after the first spike due to the laser: The Unibeads C and the anthracite exhibited a single broad peak, while the hv bituminous coal showed two broad

Figure 2 PMT traces showing the ignition and combustion behaviours of (a) Unibeads C particles, (b) anthracite and (c) hv bituminous coal in pure oxygen

Figure 3 Reaction sequence of the carbon spheres: (a) corresponds to the first frame seen, (b) 1 ms later, (c) 6 ms later, and (d) 47 ms later. Figure 3e is the PMT trace, and the labelled, dashed lines mark the events shown in Figures 3b–d. The oxygen concentration is 100%, and the laser energy is 720 mJ

peaks. In order to determine the causes for these differences, the processes have been observed with high-speed video.

The ignition and combustion of the carbon spheres is shown in Figures 3a–d, and Figure 3e shows the PMT trace from the channel with $\lambda_c = 0.631 \mu\text{m}$. In all photographs, the laser travelled from near the upper left and the lower right corners on the first and second passes, respectively. Figure 3a shows that eight to 10 particles were intercepted by the two laser passes. However, since the beams did not travel along the exact same path, only the particles near the centre of the photograph were heated by both laser passes. This is clearly seen in Figure 3b, in which the particles near the top and bottom (heated to a relatively lower temperature by a single laser pass) have cooled off and can no longer be seen, and only high temperature particles remain visible. Six milliseconds after the laser pulse, Figure 3c, all particles except one had extinguished; and clearly this particle ignited and was responsible for the emission of Figure 3e. In the time between Figures 3c and d, the particle left the field of view of the camera, but remained in the emission system's view. Soon after leaving the camera view the particle reversed direction and travelled

upward, and re-entered the camera field in Figure 3d. The reason it appeared larger in Figure 3d than in c is unclear, but two explanations are possible. First, the apparent enlargement may be due to a volatile flame burning around the particle, but this is unlikely for this carbon sphere with little volatile matter, and a previous combustion study² on a similar sample did not report the observation of a volatile flame. The second possibility is that the enlargement is caused by particle movement out of the camera's focus. The ignition frequency of Unibeads C was very low, even in 100% oxygen and at maximum laser energy, so it could not be observed enough times to establish whether the enlargement was a general behaviour.

Figure 4 shows the ignition and combustion sequence, and the PMT trace of the anthracite. The sequence from Figures 4a–c shows that the particle was shattered by the laser pulse. The many particles seen could not be due to fine particles in the sample, since all coals were treated in the same manner; and the presence of so many particles was seen only for the anthracite. Nearly all fragments extinguished within 10 ms after the laser pulse; however, in approximately 50% of the cases one to three of the particles ignited. It was not clear whether these were large

Figure 4 Reaction sequence of the anthracite. The times after the first frame, (a), are: (b) 2 ms, (c) 3 ms, (d) 11 ms, and (e) 18 ms. Figure 4f is the associated PMT trace, and the dashed lines mark the events shown in Figures 4b–e. The oxygen concentration is 100%, and the laser energy is 720 mJ

fragments of shattered particles or whole particles. Although the gas velocity used for the anthracite was the highest of all samples (see *Table 1*), extinction always occurred within the camera's field of view. The short combustion duration of this sample, at the same laser energy and oxygen concentration as for the carbon, suggests that the ignited particles were fragments of larger particles, with fragmentation possibly resulting from stresses in the particles caused by the large temperature gradients. The Unibeads were never observed to shatter, which means that either the temperature gradients were less severe because of faster internal heat conduction, or the tensile strength of this material was higher.

The ignition and combustion behaviour of samples with high volatile contents is remarkably different, as seen first in *Figures 5a–f* for the hv bituminous coal in 75% O₂; the associated emission and temperature profiles are shown in *Figures 5g and h*. In this case two particles were ignited by the laser, *Figure 5b*, and the point of minimum temperature corresponded to the first minimum in the emission. This temperature minimum is defined as the ignition point. At the peak of the first broad emission, *Figure 5c*, a volatile cloud could be seen surrounding each of the particles. Note that these clouds must be at high temperatures and emitting visible radiation in order to be recorded on video. At the point corresponding to the valley between the two broad emissions, *Figure 5d*, the clouds were no longer visible and the temperature had decreased. The second broad emission was several times stronger than the first, and *Figure 5e* shows that the particles were surrounded by strongly emitting volatile clouds. The apparent temperature during this time, however, was not significantly different from that measured at the peak of the first emission. Finally, some 20 ms after the second emission peak, the volatile clouds disappeared and only char combustion was seen.

Both the early and late volatile clouds appeared

spherical, but were rarely concentric about the particle. The sphericity attests to the relatively quiescent atmosphere in the reactor, and is in contrast with the wake trails observed in other studies using much higher gas velocities^{3,4}. The random eccentricity of the volatile clouds about the particles is similar to the observed random orientation of the wake trails in previous studies^{3,4}, and is attributed to preferential evolution of volatiles from certain sites on the surface.

The sequence of events just described was observed for all conditions under which the hv bituminous coal was ignited. *Figure 6* shows the same major events for particles ignited in pure oxygen. Since the laser energy was higher than the previous case (460 mJ compared to 390 mJ), the equilibrated temperature attained by the particle was higher, and thus the ignition temperature was also higher (1060 K compared to 1000 K), even though the oxygen concentration was increased. The previous description for the events shown in *Figures 5c–f* applies exactly to *Figures 6c–f*.

Figure 7 shows the ignition and combustion sequence for the subbituminous coal in 75% O₂. As with the hv bituminous coal, the PMT trace showed two broad emissions after the initial spike due to laser heating, *Figure 7f*, and the video showed a strongly emitting volatile cloud at the peak of the second broad emission, *Figure 7e*. The difference, however, was in *Figure 7c*, which corresponds to the peak of the first broad emission; no volatile cloud surrounded the particle during this time. However, the lack of an obvious cloud about the particle does not mean the absence of a volatile flame. Closer inspection of the ignition point, the first peak and the valley, *Figures 7b–d*, revealed that the emitting area increased then decreased in size slightly. This was observed in every case for the subbituminous coal, and strongly suggested that a volatile flame burned close to the particle surface.

It was noted that, as with the hv bituminous coal, the

Figure 5 Reaction sequence of the hv bituminous coal ignited by a 390 mJ laser pulse in 75% oxygen. The times after the first frame, (a), are: (b) 2 ms, (c) 8 ms, (d) 13 ms, (e) 24 ms, and (f) 44 ms. *Figures g and h* show the corresponding PMT trace and the temperature measured by two-colour pyrometry. Events shown in *Figures 5b–f* are marked in *Figure 5g* by dashed lines

Figure 6 Reaction sequence of the hv bituminous coal ignited by a 460 mJ laser pulse in pure oxygen. The times after the first frame, (a), are: (b) 2 ms, (c) 6 ms, (d) 10 ms, (e) 26 ms, and (f) 46 ms. Figures 6g and h show the corresponding PMT trace and the temperature measured by two-colour pyrometry. Events shown in Figures 6b–f are marked in Figure 6g by dashed lines

Figure 7 Reaction sequence of the subbituminous coal ignited by a 310 mJ laser pulse in 75% oxygen. The times after the first frame, (a), are: (b) 2 ms, (c) 8 ms, (d) 13 ms, and (e) 21 ms. Figure 7f is the associated PMT trace, and the events shown in Figures 7b–e are marked by dashed lines

sequence of events just described occurred at all conditions in which the subbituminous coal ignited. Thus it can be generalized that, while the combustion behaviour of the samples depended on coal type, oxygen concentration and laser energy did not affect the visually observable events of each sample. Of course, oxygen level and laser energy influenced measurable quantities, such as the apparent temperature and combustion duration.

For the hv bituminous and subbituminous samples of this study, a phenomenon called ‘cloud ignition’ was sometimes observed and is demonstrated by the sequence in Figure 8. Figures 8a and b show that, at most, three closely spaced particles were ignited by the laser pulse. However, the volatiles from the particles interacted and formed a much more extensive cloud, which grew rapidly in size as more particles were fed into this cloud, Figure 8c. The region of intense burning increased until a cloud containing tens of particles was formed, Figure 8d. Driven by buoyancy the flame rose and was finally stabilized on the end of the feeder tube. This phenomenon occurred most frequently at high oxygen concentration and high laser energy, and more readily for the subbituminous than the bituminous coal. Note from Figure 8e that it was still possible to measure the ignition temperature in this circumstance.

In closing, one final observation regarding the video images is made. For the three coals in this study the first video frame nearly always showed an intensely bright region (see Figures 4a, 5a, 6a and 7a); this was not observed with the carbon spheres, Figure 3a. The source of this emission is unclear. One possibility is the formation of a plasma, but this may be ruled out as the laser energy needed to ignite the hv bituminous and subbituminous samples was low, compared to that used with the carbon and anthracite. The surface temperature of the carbon spheres must be much higher than the bituminous and subbituminous coals, but no bright region was observed in this case. Another explanation is that the large

Figure 8 Reaction sequence demonstrating 'cloud ignition' of the hv bituminous coal. The oxygen concentration is 100%, and the laser energy is 300 mJ. The times after the first frame, (a), are: (b) 6 ms, (c) 18 ms, and (d) 36 ms. Figure 8e is the associated PMT trace, and the events shown in Figures 8b–d are marked by dashed lines. The PMT is saturated at a signal level of 10 units

temperature gradient caused by the laser pulse resulted in devolatilization of the outer region of the coal particles. The pyrolysis products, in turn, either burned rapidly in the vicinity of the particles or simply emitted visible radiation due to their high temperature. The only doubt in this hypothesis is the anthracite's behaviour. The extent of the bright region was similar for all three coals, yet the very low volatile content of the anthracite suggested that, if the emission was due to pyrolysis products, its behaviour should be more similar to the carbon spheres. To resolve this issue, pyrolysis modelling of pulverized coals with imposed temperature gradients and on submillisecond time scales should be conducted.

DISCUSSION

The high speed video has resolved many questions raised in a previous study¹ regarding the ignition and combustion behaviours of various fuels. Regarding the anthracite's relatively long emission due to laser heating and temperature equilibration, the video showed that the anthracite's behaviour was anomalous because of particle fragmentation. Furthermore, differences in the reaction

time of various samples are clarified. The carbon spheres exhibited the longest reaction time because the low gas velocity used for this fuel resulted in a long residence time in the detection system. Conversely, the higher gas velocity used for the hv bituminous coal led to a shorter residence time. Since the anthracite was always observed to extinguish before leaving the camera's view and its gas velocity was the highest, its short reaction period was due to the short combustion duration of fragments of the original sample.

The most remarkable finding in this study was the observation of two distinct volatile clouds during the combustion of the hv bituminous coal. Furthermore, each cloud was associated with a broad emission peak detected by the PMT. The combustion behaviour of the subbituminous coal followed the same sequence, it is believed, but the first volatile clouds remained close to the particle surface. The observation of two distinct volatile clouds during combustion in a quiescent atmosphere is not terribly surprising if one considers previous results from pyrolysis studies. Experiments in which secondary pyrolysis, i.e. decomposition of the devolatilization products in hot gas environments, is eliminated have shown that product distribution is strongly dependent on coal type^{5–7}. In the early stage of pyrolysis of hv bituminous coals, weight loss is dominated by tar evolution, with little hydrocarbons and relatively more CO₂ and H₂O, while early subbituminous pyrolysis is due to approximately equal parts of tar and gases, again mainly as CO₂ and H₂O. The late stage of pyrolysis for both coals is similar, and is characterized by the cessation of tar evolution and the production of light hydrocarbon gases, H₂, and CO. It is reasonable, therefore, to presume that the first broad emission peak and volatile cloud of the subbituminous and bituminous coals are due to tar evolution, and the second emission peak and cloud are due to light, combustible gases. Furthermore, the low tar yield of subbituminous coals⁵ would explain why the first volatile flame of this coal remained close to the particle surface.

These experimental observations differ dramatically from previous observations of coals burning in post-flame gases containing excess oxygen. Using high speed shadowgraphy, McLean *et al.*³ showed that the ignition of a bituminous coal was coincident with the appearance of a condensed phase material surrounding each particle. Due to the relative motion between particle and gas, this material was quickly swept into long trails and was eventually oxidized. Seeker *et al.*⁴ made similar observations using holography. Thus, both previous studies reported only the formation of a condensed phase, attributed to soot formed from tar and hydrocarbons, which appeared coincidentally with the ignition point. New hypotheses, for the presence of a second volatile cloud and for the delay in appearance of the first cloud until well after ignition, are described in the remainder of this section.

As noted earlier, this experiment presented a more quiescent atmosphere compared to those of previous studies with direct observation^{3,4}; these gas velocities were five to 10 times lower. The existence of large relative motion between particle and gas in those experiments may be the reason why a second volatile flame was never observed: the late volatiles, mainly combustible gases, were swept away by the bulk gas flow. Under the dilute conditions, the volatiles would be oxidized far from the

particle surface, rather than in a spherical region surrounding each particle.

Along with previous researchers^{3,4} it is believed the volatile trails observed were due to soot formed from secondary pyrolysis of tar. This was also supported by recent pyrolysis studies⁸. Furthermore, it is believed the first volatile cloud observed in this experiment was due to tar and/or soot, but one could not distinguish between the two. That this volatile was observed on video means it must have been at high temperature and emitting visible radiation, and this could be attributed to the combustion of tar or soot, or to the absorption and emission of radiation from the coal particle by soot. (The scattering of radiation by very young soot is insignificant⁹.) These uncertainties further complicate temperature measurements by two-colour pyrometry, as shown in numerical studies^{9,10}. The presence of a soot cloud surrounding the particles introduces great uncertainties as to which temperature — the particle's or the soot's — is being measured, and the existence of a flame emitting in various spectral bands would obviously distort pyrometry measurements. The primary objective for this experiment is to examine the ignition of pulverized fuels; therefore, so long as ignition occurs heterogeneously, two-colour pyrometry can accurately measure the ignition temperature.

Examination of *Figures 5b, 6b and 7b* shows that, apparently, ignition of the bituminous and subbituminous coals in this experiment occurred heterogeneously, and volatiles evolution was delayed until well after ignition. McLean *et al.*³ observed ignition and volatile cloud formation to be coincident to within 1 ms for a bituminous coal. Two possible explanations for the apparent delay in volatiles evolution in this experiment are offered. The first has been discussed previously: the temperature gradient caused by the laser pyrolysed the outer region of the particle. Oxidation of the small amount of volatiles produced did not generate sufficient heat to ignite the particle, and the particle's outer layer became devoid of volatile matter, leading to the observed heterogeneous ignition. Again, this hypothesis must be confirmed or denied through detailed modelling.

The second explanation derives from studies of primary pyrolysis in wire-mesh reactors with controlled heating rates^{11,12}. In these studies a monolayer of coal was heated at a fixed rate to various temperatures and then cooled naturally or by forced convection. The sample was next weighed to determine the volatiles yield. The results showed conclusively that increases in heating rate shifted weight loss to higher temperatures. For example, Niksa *et al.*¹² showed that, for two bituminous coals, increasing the heating rate from 10^2 to 10^3 K s⁻¹ resulted in a shift in the temperature at which, say, 10% weight loss was attained from 800 to 1000 K. Thus, it is possible that in this experiment, in which the heating rate is greater than 10^6 K s⁻¹, the onset of devolatilization is shifted to temperatures above which the solid particle ignites. This concept of a transition in ignition mechanism due to heating rate is previously unrecognized, and deserves further examination in experimental and modelling studies.

CONCLUSIONS

From the results and discussion presented here, many questions involved with this new experiment have been answered, and also some new issues have been raised. The videos showed that the amorphous carbon spheres ignited heterogeneously and, based on the emission characteristics, reacted heterogeneously. The videos also demonstrated that the anthracite was not suitable for this experiment because of particle fragmentation by the laser pulse.

The reaction of the subbituminous and hv bituminous coals was characterized by two broad emissions, each associated with a combusting volatile cloud surrounding the particles. It is believed the first cloud consisted of tar and/or soot, and the second was due to the evolution and reaction of low molecular weight, combustible gases. For the subbituminous coal, the first cloud remained close to the particle surface because of the low tar yield of subbituminous coals. The second volatile cloud was conspicuously absent in previous studies because the relative motion between particle and gas in those studies swept away the volatiles. Finally, it can be generalized that, while the combustion behaviour of these fuels strongly depended on coal type, oxygen concentration and laser energy did not affect the visually observable events of each sample.

The videos also revealed that ignition of the bituminous and subbituminous coals occurred heterogeneously, in contrast with previous observations showing simultaneous ignition and volatiles evolution, at least for a bituminous coal. This observation may result from the laser pulse pyrolysing the outer region of the particles, leaving behind a charred surface which ignited heterogeneously. Another plausible explanation is that, at the extremely high heating rate in this experiment, devolatilization was shifted to temperatures above which the solid particle ignites. The second possibility merits further study.

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