OBSERVATION OF VOLATILE CLOUD FORMATION DURING THE EARLY STAGES OF PULVERIZED COAL COMBUSTION

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During the early stages of combustion when bituminous coal particles are in a nearly single particle combustion environment, a surrounding mantle of volatile products is observed. Initially, the cloud is of spherical shape and almost concentric with the particle. As the reaction progresses, more material is ejected in the shape of jet-like tails, This rapid mass release was observed through in-situ high speed photographs of particles in combustion enviroment, and was also confirmed by examining cross sections of the quenched samples of coal particles. The inner structure became porous, while base material softened and some portion of it was ejected due to high pressure build up inside. Direct sampling of the partly burned coal particles revealed that the material in the volatile cloud contained tarry substances and small sized particle fragments in addition to gaseous volatiles. These solid and condensible substances were seen as a luminous envelope which implied that they participated in the radiative transfer process.

Key Words: Pulverized Coal Combustion, Devolatilization, Volatilecloud

1. INTRODUCTION

Understanding of pulverized coal particle behavior during the early stages of combustion has been greatly improved through recent endeavors of both experimental and modeling approaches. Direct in-situ observations of coal particles which are burning while being transported by the surrounding gas, has been made possible by the high speed shadow movie technique (McLean et al., 1980) and by the holographic imaging technique (Seeker et al., 1980). Both experiments showed the progress of pulverized coal combustion immediately after being injected into simulated combustion environments. Typical bituminous coal particles were observed to have very intense luminous 'flames', immediately following the initial heating-up phase. Both the shadowgraphs and holographs showed that the particles at these stages were surrounded by their accompanied mantles of nearly concentric shape. This surrounding mantle configuration is called the formation of the volatile cloud. The cocentric cloud was observed to survive until long and slender tail-like structures were formed. Initially, the tail appeared to extend from the particle, but gradually they separated from the particle.

In an attempt to explain this observation of the volatile cloud formation, several models have been proposed (Timothy et al., 1982; Jost et al., 1984; Choi and Kruger, 1985; Gururajan et al., 1988; Masarra et al., 1985; Phuoc and Durbetaki, 1985). Many of the models resemble those for liquid droplet combustion, in which a spherically symmetrical volume of the mantle is filled with the devolatilized products that will oxidize at a very fast rate forming an infinitely thin flame sheet.

The previous observation and modeling have provided more insight into the behavior of the pulverized coal particles, but major questions still remain to be answered: what is the material present in the volatile cloud?; when and how are the spherical volatile cloud and the jet-like long tail formed?; and what are the dominating mechanisms of luminous radiation from the burning coal particles? The investigation attempts to provide partial answers to the questions raised above, by presenting some recent experimental results in order to construct a plausible scenario of the phenomena.

2. EXPERIMENTAL APPARATUS AND PROCEDURE

2.1 Coal Combustion Burner

A small laboratory-scale burner with a fluidized coal feeder were used. (see Fig. 1. for the schematic of the experimental set-up). The burner is an atmospheric pressure flat flame burmer fed with premixed mixture of methane and air. The flame is formed immediately downstream of a porous metal plate. Approximately 400 holes of 1 mm diameter are drilled with 2 mm spacing. By changing the stoichiometry, the oxygen concentration in the postcombustion gas flow could be adjusted. Typical equivalence ratios selected for the experiments resulted in the excess oxygen concentrations between 0.0 and 4.0%.

The fluidized-bed coal feeder was constructed of acryl tube of 50 mm diameter. Both the top and bottom end walls are made of 10 mm thick sponge and several sheets of filter paper. The fluidizing air was fed from the bottom end while most of the air was blown out through the top end wall exhaust. Minute quantities of particles were carried through the intake tube of diameter 0.6 mm. This coal was introduced into the combustion environment through an injector tube located immediately upstream of the perforated metal plate in the flat flame burner.

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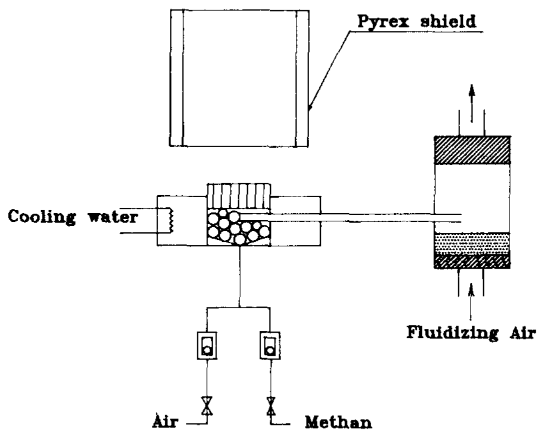


Fig. 1 Schematic of experimental apparatus

Several different types of coal samples were tested. The tests reported here were performd with Australian high volatile bituminous coal (called Palmco following the name of the supplier) used at the Korea Electric Power Corporation. The coal samples from the pulverizer mills were size classified using standard sieves (−100 mesh and +200 mesh) and were air dried in the fluidized bed. Smaller size particles which were inevitably carried along with larger particles were blown away during this preparation process.

2.2 Schilieren Photography

Visualization of the burning coal particles is difficult in the sense that small objects (typically on the order of less than 100μm) are moving at a relatively fast speed (typically on the order of 1~10 m/sec). Considering the sensitivity of the photographic recording devices, back lighting was favored to the direct photographic imaging of the object. A Schilieren photography arrangement was installed with a flash light source of very short duration time. A Nanolite (of Impulspysik) was used typically at 5 kHz repetition with 10 nanoseconds duration.

Typically a 5x magnification ot the a film plane was achieved using an achromatic lens. A rotating drum camera (of Impulsphysik) was used along with Ilford HP5 35 mm film of ASA 400 (however, the film was processed to an equivalent sensitivity of ASA 6400).

2.3 Sampling and Observation

Pulverized coal particles in the post-flame gas flow were collected by inserting cold plates into the flame at a direction normal to the flow. The cold plates used were either standard quartz glass for microscope slides or metalized glass plates with an electrically conductive surface (Corning No. 7059, 1μm Molybdenum coated glass). The position of the plate insertion was carefully adjusted so that the collected particles would represent those at the equivalent residence time. Coal particles, as well as some intermediate products, were quenched upon contact with the cold plate. They adhered to the plate, requiring no additional treatment. The sample plates were examined with the aid of optical and scanning electron microscopes.

In order to observe the internal structures of the coal particles, a cross-section micogaphy technique was used. A sample holder made of epoxy resin with one end of the column partially cured. Coal particles in the flame would then be deposited in-situ. After a careful sample collection period, aditional epoxy resin was poured onto the surface where the quenched coal particles had been deposited. Once the whole sample piece was completely cured, the top surface was ground and polished following the standard procedures of sample preparation used for metallographic observation. The cross-section of the partly reacted coal particles were examined with optical microscopes.

3. EXPERIMENTAL RESULTS

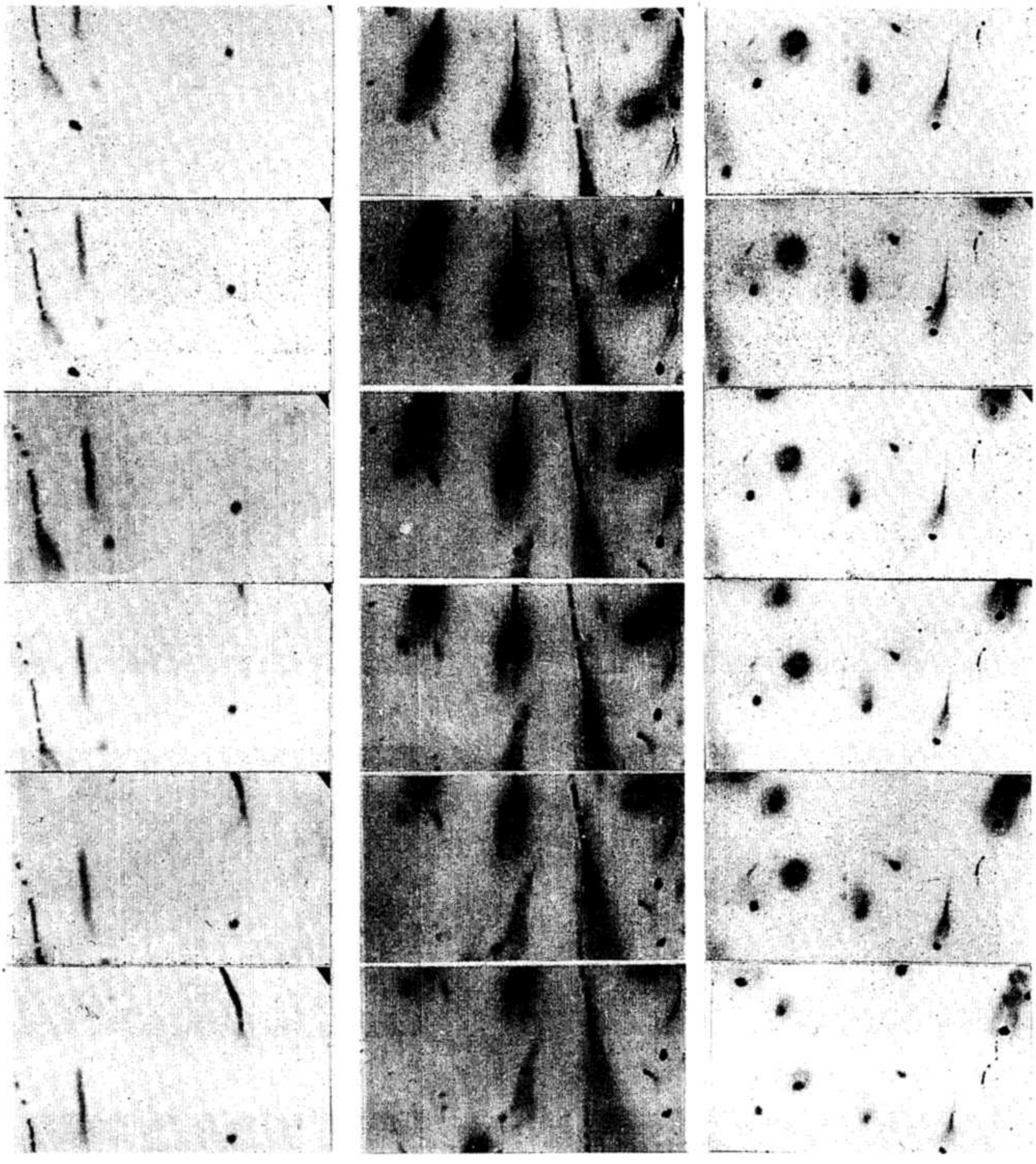
Coal particles under simultaneous devolatilization and combustion were visualized by Schilieren photogrphy. Figure 2 shows a series of high speed movie frames. The time interval between each frame is 200 μs and the scale magnification is 5x on the film plane. Figure 2(a) shows a set of typical movie frames taken from a burner location equivalent to 10 ms after injection. The gas environment contains 3.6 % of excess oxygen (equivalence ratio of $\phi = 0.84$) and the temperature is 1750±150 K. Figure 2(b) and (c) represent particles at residence times of 20, and 40 ms, respectively. Though Figs. 2(a), (b) and (c) do not represent identical particles, they are deemed to represent typical behavior of the particles at the corresponding residence times.

The particles in Fig. 2(a) are shown with more or less concentric volatile clouds. There is, obviously, particle-to-particle variation. The volatile cloud should be of higher density material, since the cloud blocks the light coming from behind, but direct photographs also indicate that the total cloud is illuminating. It is not clear whether the radiation originates from the cloud as a whole, or from the flame front where the volatile gases burn according to the oxygen diffusion rate controlled reaction hypothesis.

The jet-like tails shown from the Fig. 2 (b) represent partcles at a slightly later time than the time of the concentrical volatile cloud formation. The observations are similar to those of previous investigators (McLean et al., 1980), who reasoned that the 'wakes' were mainly due to velocity slip between the particle and the surrounding gas. The formation of these 'tails' can also be interpreted as jetting of volatile matter from the particle through macro pores leading to the outer surface.

Figure 2(c) shows coal particles observed without the surrounding volatile cloud. Volatile products in the tail remains unburned (at least not completely oxidized) in much the same way as the solid coal particles. At different stoichiometry (no excess oxygen but temperature of 1850±150 K), a similar sequence of events was observed. One major difference was that the volatile cloud survived for a longer period before being swept away from the particle.

The volatile cloud and the remaining jet-like tails were more closely observed by direct sampling. The cold plate insertion method collects material not only from the particles but also from the intermediate products such as the volatile cloud and the long tails. Figures 3(a) ad 3(b) show scanning electron micrographs of typical particles and their accompanying volatile products. Figures 3(c) and 3(d) show enlarged views of the tails shown in Fig. 3(b). The sponge-like structure along with smaller particles of diameter on the order of 1 μm are evident. It is believed that these material must have been ejected from the particle through holes on the outer surface of the particles, some of which are shown in Fig. 3(a). The diameter of these long tails can be estimated to

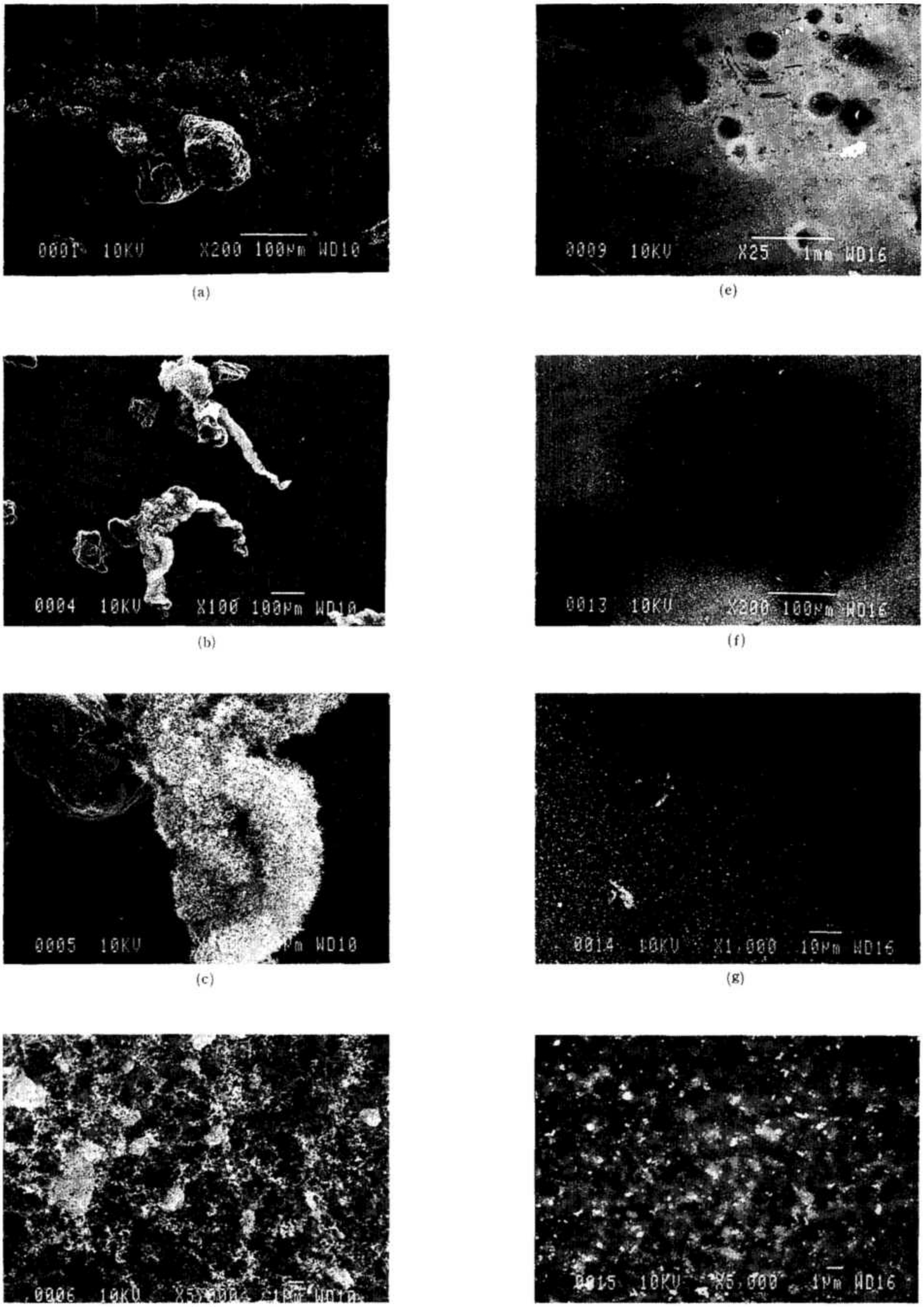


(a) represents residence time of 10 msec after injection, (b) 20 msec, (c) 40 msec

Fig. 2 High speed schlieren photographs, 5000 frames per second

be of the same order of magnitude as the particles, and length can range up to 1 mm. The particular tails are shown to be connected to the respective particles.

The volatile cloud can also be observed from the cold plate samples. Figures 3(e), 3(f), 3(g), 3(h) show volatile clouds at different magnifications. At the magnification of Fig. 3(e), several volatile clouds are observed along with original coal particles. It is believed that the material from the volatile cloud is directly deposited on the plate. Some of the coal particles are not shown on this micrograph, but the corresponding sites are clearly identifiable. The volatile clouds observed at higher magnifcations show that material in the form of solid or condensible phase does exist inside the apparent volatile cloud. It may be of fine soot-like structure



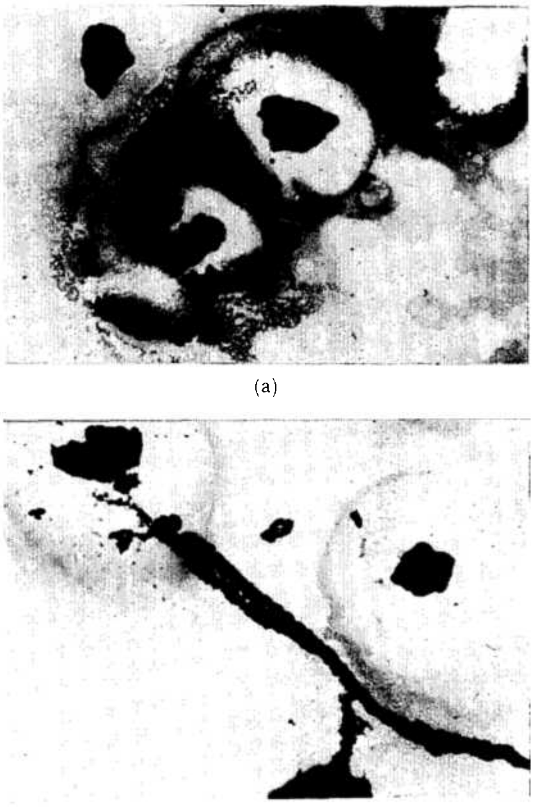
(a) (e)

(b) (f)

(c) (g)

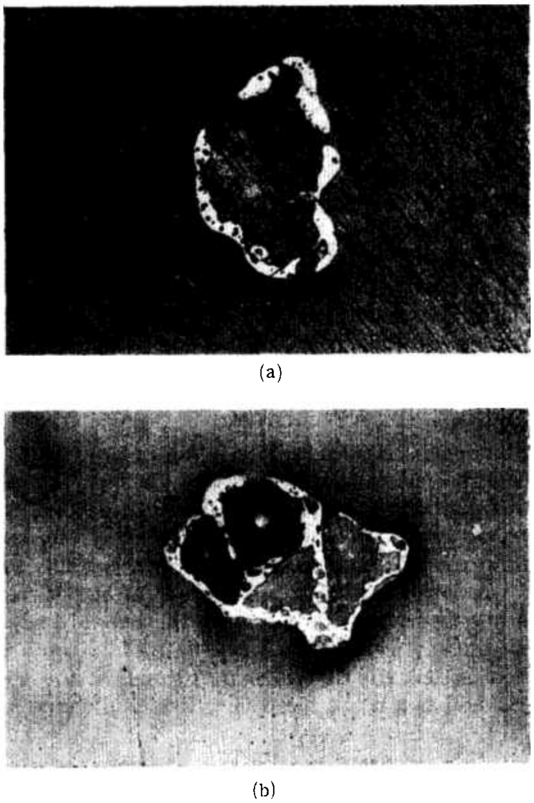
(d) (h)

Fig. 3 Scanning electron micrographs of volatile products (a), (b), (c), (d), (e), (f), (g), (h)


(a)

(b)

Fig. 4 Optical micrographs of volatile cloud and tails


(a)

(b)

Fig. 5 Cross-section micrographs of partly reacted coal particles (a), (b)

as shown in Fig. 3(f) or of sub-micron particles as shown in Fig. 3(h). It is not clear, however, whether the exactly same material is present inside the volatile cloud at the time of combustion in the hot environment.

Optical micrographs of the volatile cloud and long jet-like tail are also shown in Fig. 4(a) and 4(b). It appears that the volatile clouds in these micrographs have a clear outer edge. The main difference in sample preparation between Fig. 3(e) and Fig. 4(a) is that a quartz slide glass was used for Fig. 4(a), and a metalized glass was used for Fig. 3(e). It is not clear upto this point if these two different kinds of volatile cloud pictures are due to the difference in sample preparation, or if the material and configuration of the cloud are different in two cases.

Micrographs of the cross sections of the partly reacted coal particles are shown in Fig. 5(a) and 5(b) (additional micrographs and discussions were reported previously in Han and Choi, 1989). The internal structure of the particles becomes more porous as reaction progresses. Some of the holes observed on the outer surface can be understood as the gate area of the particle perimeter in the cross-section micrographs. It is clear that the major portion of the internal material has disappeared, and it can happen by jetting of the molten (or softened) material through the holes on the surface of the particle.

4. VOLATILE CLOUD FORMATION (A SCENARIO)

As discussed in the previous sections, main idea behind the explanation of experimental observations as well as the theoretical modeling is that the mass release from the particle is so vigorous that oxidation or oxygen diffusion is rate-limiting. In other words, the ejected pyrolysis products can oxidize only at the flame region which is located further outside of the particle. And this causes the formation of the volatile cloud.

One point that is not clear is what is the composition of the released products in the volatile cloud; is it gaseous phase combustibles or fragmented solid particles or condensible hydrocarbons? Researchers examining the devolatilization processes have found that a significant amount of mass is evolved in the form of tar. The tar is not a precisely defined terminology. Suuberg (1978), for example, reported that up to 60 % of mass is released as tar or liquids, for the case of high volatile bituminous coal at the temperature of 1300 K. It is well known that the volatile release rate is significantly enhanced as temperature increases. However, very little information is available about the composition of the released volatiles at higher temperatures, say 1800~2000 K, which is the typical combustion environment where most of the coal combustors are operating.

The present and previous (McLean et al., 1980; Seeker et al., 1980) observations indicate that the released volatiles in the cloud are higher density materials. They include solid phase material of very small size and condensible phase pyrolysis products. These heavy volatile products can be called as sooty pyrolysis products. Significant amounts of these non-gas phase volatile products can be ejected from the particle during the very early stages of the volatile cloud formation. Simultaneously, the inner structure of the particle changes in nearly the same manner as the well-known softening and bubbling of bituminous coals at relatively lower

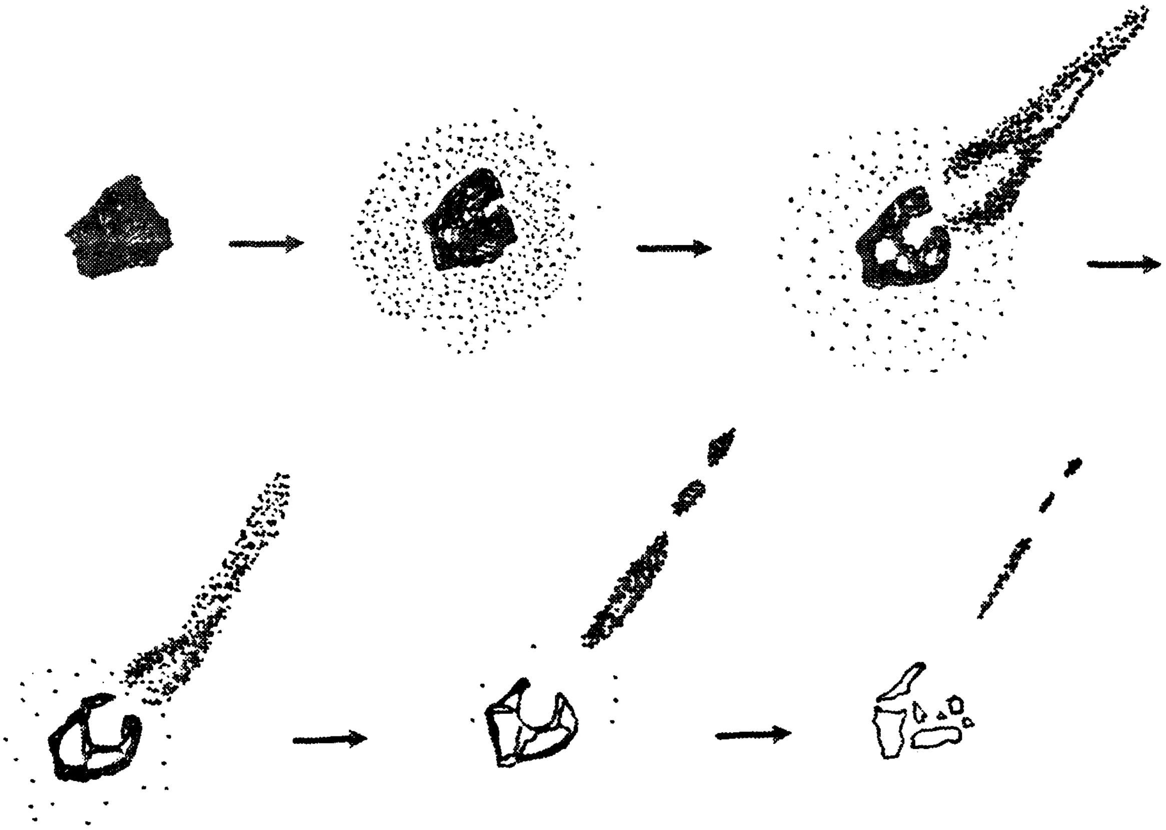


Fig. 6 Conceptual diagram of coal particle combustion behavior.

temperatures of around 1000K.

The softened inner core material (the major portion must be soft vitrinite) can escape the original particle through smooth volatilization, but it can also be ejected through a hole in the coal particle surface. A tail-like long structure may form. The long tails are not immediately oxidized. They tend to retain their shape, while break-up and surface degradation gradually change their overall appearance.

These processes are represented schematically in Fig. 6. When the coal particles are heated up in the combustion environment, volatile products are produced and a nearly concentric mantle of the volatile can be formed. Later, some of inner core material is rapidly ejected through one of the opening pores, and a long tail can be formed. The inner structure changes due to the hot environment and more micro-scale pores are generated. As reaction progresses the particles may break-up and shrink in size. The original particle can no-longer maintain its original structure and it breaks-up. As a result, small size ashes are generated.

5. CONCLUSIONS

(1) Pulverized coal combustion has been investigated experimentally with emphasis on the physical mechanisms of early stages of combustion when devolatilization and combustion take place simulataneously. High speed Schlieren movies show the formation of the volatile colud and the long tails of pyrolysis products. The volatile cloud is initially nearly concentric in shape, and it is filled with condensible material and very fine sized paricles. Some of the pyrolysis products are ejected from the particle as a volatile jet, thereby forming a long and slender tail. These are believed to be ejected from the original particle from a very violent mass ejection. These tails retain their shape for a relatively long period which indicates that the oxidation of these material is not immediate, even in a highly oxidizing and high temperature environment.

(2) During the early stages of devolatilzation, released mass from coal particles can be in gaseous, solid or condensible phases. The small particles found from the volatile cloud are larger in size than the soot precursors that can be observed from the gaseous fuel flames. The internal structures as well as the surface of the particles show evidences of rapid mass ejection.

(3) The coal particles radiate in hot environment. During the simultaneous devolatilization and combustion phase, the illuminating body is larger in size than the original particle. When most of the ejected mass is burned away, the size of the radiation source decreases to that of the particle. It is not clear whether radiation originates from the surrounding flame of gaseous volatiles, or from the overall body of the volatile colud. It is plausible, however, that the sooty volatile products in the volatile cloud participate in the radiative process, and as a result the whole cloud is illuminating.

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