

## Investigation into the ignition of coal powders in the presence of oxygen and natural gas by means of high-speed cinematography

Nikolai M. Rubtsov,\* Boris S. Seplyarskii, Georgii I. Tsvetkov and Victor I. Chernysh

*Institute of Structural Macrokinetics and Problems of Materials Science, Russian Academy of Sciences, 142432 Chernogolovka, Moscow Region, Russian Federation. Fax: +7 495 962 8025; e-mail: nmrubtss@mtu-net.ru*

DOI: 10.1016/j.mencom.2012.01.019

The mixtures of soot or graphite powder with a stoichiometric mixture of natural gas and oxygen intensely ignite at lower initial temperatures, as compared with that for powder–gas mixtures of coal powders with the above mixture of natural gas and oxygen.

Dust explosions in mining are very dangerous due to the formation of suspensions of combustible particles in air. The probability of explosion of dust powders increases with the concentration of gaseous combustible components.<sup>1</sup> The minimum energy of ignition of a hybrid coal powder–gas mixture (PGM) can be lowered by a factor of 20–30 in the presence of 2–3% methane.<sup>2</sup> Such conditions are characteristic of mines where both combustibles, coal powder and natural gas (NG), can be present simultaneously. Therefore, the investigation into the ignition of hybrid coal PGM containing gaseous fuel and oxidizer is of considerable current interest.

We have shown recently<sup>3</sup> that high volatile coal powder cannot ignite NG (4.5–33%) + oxygen mixtures, though it ignites in pure O<sub>2</sub>; the reactor surface markedly influences the ignition of hybrid PGM; the heated reactor surface covered with coal powder evolves the effective inhibitor of methane combustion, probably, a polycyclic aromatic hydrocarbon; the ignition of hybrid PGM can be promoted with small amounts of a chemically active additive (e.g., dichlorosilane).

It follows from published results<sup>3,4</sup> that the gases evolving from heated coal powder show a considerable inhibiting influence on the spontaneous ignition and combustion of hybrid coal PGM. Note that volatiles in coal contain more than 200 chemical compounds,<sup>1,2</sup> and it complicates identification of the substance inhibiting methane combustion. Therefore, a comparative approach was used, which allows revealing a basic role of the volatiles evolving from coal powders in ignition process. The method is based on a comparison of the ignition and combustion of powders, which do not contain volatiles (soot, graphite), and coal powders with the various volatile contents. Based on the results, we notice that the features of gas combustion cannot be applied to the combustion of hybrid PGM because the ignition of combustible gas and solid powder are in close interrelation.<sup>3</sup>

The aim of this work was to investigate the flammability of coal powders with various volatile contents in oxygen by means of high-speed cinematography. The ignition of soot and graphite

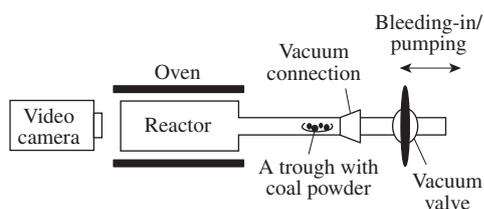
powders (which do not contain volatiles) in oxygen and stoichiometric mixture of NG with oxygen were also investigated.<sup>†</sup>

At the moment of injection of coal particles into the reactor by oxygen stream, bright sparks occur in the reactor, representing ignition of separate coal powder particles in oxygen. The ignition is not accompanied by gaseous combustion. This process lasts for ~0.01 s, and then light emission of the sparks dies away [Figure 2(a)]. In the time interval of several seconds during which light emission in the reactor was not observed, intense luminescence occurs. The luminescence can arise either in the reactor volume or at the walls and then propagates over the reactor volume. Figure 2 shows that the time of occurrence of intense emission, which we treat as ignition time of hybrid coal PGM, is much less for PGM containing HV coal powder than for A coal powder.

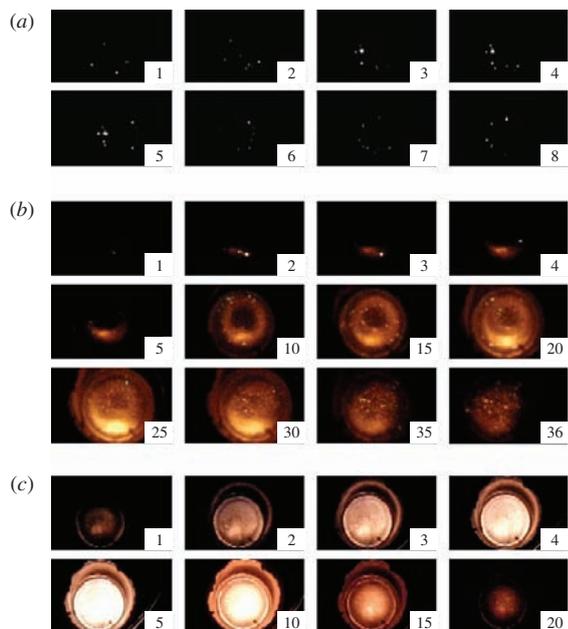
We assumed that the emission shown in Figure 2 is caused by the combustion of volatiles evolved from heated coal powder in oxygen. The time interval  $\tau_L$  between the injection of PGM into the reactor and ignition is comprised of the time of heating of powder particles  $t_1$ , the time of evolving volatiles from heated

<sup>†</sup> The experiments were performed at a total pressure of 85 Torr and initial temperatures of 600–750 °C in a heated cylindrical quartz reactor 3.6 cm in diameter and 25 cm in length. Hybrid PGM in the reactor was produced in the way described elsewhere.<sup>3</sup> Coal, soot or graphite powder (0.2 g, particle size ~60 μm) was poured into the quartz trough. Then, the trough was placed in the reactor at its inlet (Figure 1) and the reactor was pumped to 10<sup>-2</sup> Torr. The coal powder was injected into the evacuated reactor with a sharp stream of test gaseous mixture from a buffer volume so that the total pressure in the reactor reached a necessary value (85 Torr O<sub>2</sub> and 1 atm air). At achievement of the pressure the reactor was shut off the buffer volume. The ignition was recorded by means of a Casio Exilim F1 Pro colour high-speed digital camera (1200 frames s<sup>-1</sup>); each set of frames was stored in computer memory; the resulting frame sequence was analyzed. The hybrid PGM obtained as described above occurred in the reactor volume at 680 K for no less than 10 s. To be certain that coal particles remain in PGM the particle cloud was imaged using a horizontal sheet of laser light ( $\lambda = 524$  nm) formed by means of a cylindrical lens along with camera recording (not shown in Figure 1), as described previously.<sup>5</sup> Therefore, in our experiments, coal particles remained in the mixture at ignition delays  $\tau < 10$  s. NG contained 98% methane and 2% propane and butane. Chemically pure O<sub>2</sub> was used.

The following types of coal with a particle size of ~60 μm were used: high volatile steam coal (HV, ~38% volatiles, GOST 10101-79), coking coal (C, ~17% volatiles, GOST 25543-88), anthracite (A, ~8% volatiles, GOST 25543-88; average particle size, 55 μm), and also powders of soot P840T GOST 38154-88 (particles contained nearly equal contents of fractions: 0.2, 0.5, 1.1, 5 and 10 μm) and graphite GOST 7478-75 (average particle size, 55 μm). The analysis of the particle size was performed using an Analyzette 22 laser diffraction analyzer (Germany).



**Figure 1** Experimental setup for creating hybrid PGM in the reactor.



**Figure 2** Sequences of video images of hybrid coal PGM ignition in oxygen at 85 Torr (1200 frames  $s^{-1}$ ). Numbers in the picture correspond to consecutive number of the video image. The first shot number corresponds to the beginning of process registration. (a) Ignition of coal powder particles after injection (coal A, 654 °C); (b) ignition of volatiles evolved from coal A (654 °C) in  $\tau_L$ ; (c) ignition of volatiles evolved from coal HV (649 °C) in  $\tau_L$ .

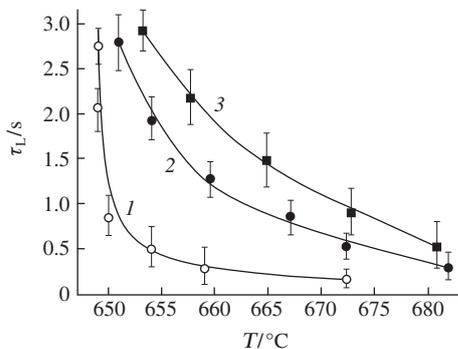
powder to gas phase  $t_2$  and the ignition delay of volatiles in oxygen  $t_3$ :

$$\tau_L = t_1 + t_2 + t_3.$$

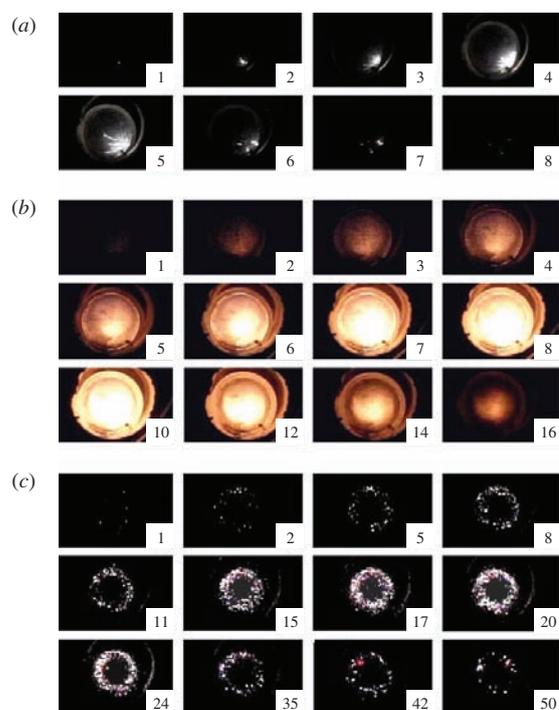
It was established that  $\tau_L$  decreases with the reactor temperature and depends on the type of coal powder; thus, minimal  $\tau_L$  values relate to PGM evolved from HV coal powder with the greatest content of volatiles (Figure 3). The ignition of this PGM in the test range of temperatures causes both quartz trough being propelled across reactor and a characteristic sound; *i.e.*, the ignition of the volatiles causes a pressure jump in the reactor, which is capable to move a thing in the reactor. PGM containing coal powders C and A ignite less intensely and with longer  $\tau_L$  (Figure 3).

Additional experiments have been performed with the injection of PGM with air to atmospheric pressure. The ignition of HV coal powder at 530 °C also causes both quartz trough being propelled across the reactor and a characteristic sound; PGM containing coal powders C and A do not ignite at all at this temperature.

To verify the assumption that volatiles evolving from heated coal powder provide the main contribution into the flammability of coal powders, a series of experiments with soot and graphite



**Figure 3** Dependence of time interval  $\tau_L$  between the injection of PGM into the reactor and ignition on temperature and type of coal powder in hybrid PGM: (1) coal HV, (2) coal C and (3) coal A.



**Figure 4** Sequences of video images of combustion of hybrid PGM containing soot or graphite (85 Torr; 1200 frames  $s^{-1}$ ). Numbers in the picture correspond to consecutive number of the video image. The first shot number corresponds to the beginning of process registration. (a) Ignition of graphite powder particles after injection (622 °C); (b) ignition of a stoichiometric mixture of NG+O<sub>2</sub> (graphite powder, 690 °C) after delay time of ignition and (c) simultaneous ignition of soot powder and a stoichiometric mixture of NG+O<sub>2</sub> (690 °C).

powders was performed.<sup>1</sup> These powders were injected into a heated reactor with oxygen stream to a total pressure of 85 Torr as described above. The first ignition of the powders, as well as coal powders, takes place right after the injection and it is accompanied by the occurrence of sparks in the reactor (Figure 4), graphite particles in an oxygen atmosphere even explode [Figure 4(a)]. Unlike coal powders, the second ignition of hybrid PGM containing soot or graphite powders and oxygen was not observed. It testifies that the second ignition of hybrid coal PGM is associated with the ignition of volatiles.

Experimental data specifying that the gases evolving from heated coal powder show considerable inhibiting action on spontaneous ignition of hybrid coal PGM containing NG+O<sub>2</sub> mixture with high methane content were obtained previously.<sup>3</sup> For example, hybrid coal PGM containing coal powder of any of the used types and stoichiometric NG+O<sub>2</sub> mixture does not ignite in the reactor, whose walls are coated with A coal powder, at temperatures up to 700 °C. Note that the inhibiting action of volatiles on the combustion of poor methane mixtures (<9%) with oxygen is less pronounced.<sup>3</sup>

Since soot and graphite powders do not contain volatiles, it can be expected that the particles of these powders deposited on the reactor by injection do not have an inhibiting influence on the spontaneous ignition and combustion of hybrid PGM containing soot or graphite and NG+O<sub>2</sub> and will hence ignite at lower temperatures than coal PGM. The following series of experiments were aimed at the investigation of the flammability of hybrid PGM containing soot or graphite powders and a stoichiometric NG+O<sub>2</sub> mixture at a total pressure of 85 Torr,  $T_0 = 685$  and 690 °C.

The measured delay time  $\tau$  of the spontaneous ignition of 33% NG in O<sub>2</sub> in the absence of PGM was considered as a control point. The value of  $\tau$  made up  $48 \pm 2$  s at 685 °C and  $43 \pm 4$  s at 690 °C over quartz surface freshly treated with 30% HF was in good agreement with previous experimental data.<sup>6</sup>

Further ignition of hybrid PGM, arising at injection of graphite powder into the reactor with 33% NG in O<sub>2</sub> was investigated. The occurrence of sparks in the reactor, representing the ignition and explosion of separate particles of graphite [Figure 4(a)], was observed after the injection. Then, the light emission of particles wiped out; however, after 48±2 s at 685 °C and 43±4 s at 690 °C, intense ignition [Figure 4(b)] was observed. Obviously, it was NG ignition in oxygen; in this case, the time interval between the injection and ignition is almost equal to the delay time of ignition of NG in O<sub>2</sub> without graphite powder. Thus, hybrid PGM containing graphite powder, does not affect the ignition of 33% NG in O<sub>2</sub>.

Then, the ignition of hybrid PGM arising at injection of soot powder into the reactor with 33% NG in O<sub>2</sub> was investigated. The intense ignition of NG was observed after soot injection along with the occurrence of sparks in the reactor [Figure 4(c)]. Thus, the presence of soot particles in hybrid PGM promotes the ignition. The promoting effect is caused by the fact that the soot powder contains easily flammable fractions with characteristic sizes of 0.2 and 0.5 μm according to particle size analysis (see above). Thus, the powders that do not contain volatiles either have no influence on the ignition of NG (graphite) or exhibit a promoting effect on the ignition at the expense of particles with a characteristic size of <1 μm (soot).

In special experiments, we found that the first ignition of soot powder in oxygen occurs at the lowest ignition temperature of 225 °C among all types of powders used. It is possible to assume that this temperature of soot ignition is low due to the presence of submicron particles.

The experimental results suggest the following mechanism of ignition in mines caused by the formation of hybrid coal PGM.

At mining of coals with high content of volatiles (HV, C), the ignition of volatiles in locally heated volumes (for example, at contact of a cutting surface to rock) is possible even in the absence of methane (Figure 2). In the presence of small quantities of methane (poor mixtures), the ignition of volatiles evolved from coal can provide the subsequent methane ignition. On the other hand, the presence of submicron coal particles in a locally heated volume can directly lead to methane ignition [Figure 3(c)]. This mechanism can occur at anthracite mining in which the content of volatiles is low.

This work was supported by the Russian Foundation for Basic Research (project no. 09-03-00622-a).

## References

- 1 R. K. Eckhoff, *Dust Explosions in the Process Industries*, 2<sup>nd</sup> edn., Butterworth–Heinemann, Oxford, 1997.
- 2 H. Franke, *Bestimmung der Minderstzudenergie von Kohlenstaub/Methan/Luft Gemisches (hybride Gemische)*, VDI-Berichte, 1978, N 304, pp. 69–72.
- 3 N. M. Rubtsov, B. S. Seplyarskii, G. I. Tsvetkov and V. I. Chernysh, *Mendeleev Commun.*, 2010, **20**, 98.
- 4 N. M. Rubtsov, B. S. Seplyarskii, G. I. Tsvetkov and V. I. Chernysh, *Mendeleev Commun.*, 2008, **18**, 340.
- 5 N. M. Rubtsov, G. I. Tsvetkov and V. I. Chernysh, *Mendeleev Commun.*, 2006, 38.
- 6 M. B. Neiman and L. N. Egorov, *Zh. Fiz. Khim.*, 1932, **3**, 61 (in Russian).

Received: 4th July 2011; Com. 11/3756