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Experimental study on flames propagating through zirconium particle clouds

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ABSTRACT

To reveal the mechanisms of flame propagation through the hardly volatile metal dust clouds clearly, the flame propagating through zirconium particle clouds has been examined experimentally. A high-speed video camera was used to record the propagation process of the flame. Combustion zone temperature was detected by a fine thermocouple. Based on the experimental results, structure of flame and combustion courses of zirconium particles were analyzed, the combustion propagation in zirconium dust was investigated, and the velocity and temperature characteristics of the combustion zone were also elucidated. The combustion zone propagating through zirconium particle clouds consists of luminous particles. Particle concentration plays an important role in the combustion zone propagation process. With the increase of zirconium particle concentration, the maximum temperature of the combustion zone increases at the lower concentration, takes a maximum value, and then decreases at the higher concentration. It is also found that the propagation velocity of the combustion zone has a linear relationship with its maximum temperature.

1. Introduction

Due to having the characteristic of resisting high temperature, radiation and causticity, zirconium is used in many fields, such as atomic energy industries, chemical plant and weapon industries. However, zirconium dust is flammable and easily explosive. Zirconium dust explosions belong to the researching categories of dust explosions, which have caused many calamities in industries.

The hazards of combustible dust clouds have been well recognized, and many experimental and theoretical researches for preventing losses by such explosions have been performed in the past few decades. In most of these researches [1–5], focus is on the characteristics of dust explosions, such as the maximum explosion pressure, the maximum rate of pressure rise, the explosion temperature, the minimum ignition energy, and the explosion concentration limits. Indeed, these data are useful for evaluating the hazards of combustible powders. Because the combustion in a dust cloud is a very complex phenomenon, it is difficult to prevent accidental dust explosions appropriately and restrain the damages without clear understanding of some basic phenomena including the flame formation and the flame propagation in a combustible dust cloud. Recently, there are some fundamental researches on flame propagation through combustible dust clouds (starch [6], lycopodium [7,8], stearic acid [9,10], metal [11–13]), such as the mechanisms of flame propagation, structure of flame, and the movement of particles around the leading edge of the flame. However, only a few studies have been conducted to clarify the mechanisms of flame propagating through hardly volatile metal dust cloud. Although various characteristics of metal dust explosions have been investigated, the mechanisms of flame propagating through hardly volatile metal dust cloud have not been sufficiently clarified. Hardly volatile metal do not vaporize even at their combustion temperature, so flame propagation characteristics through hardly volatile metal dust cloud are obviously different from that through easily volatile metal, such as aluminum [13]. Therefore, it is necessary for us to reveal the mechanisms of flame propagating through hardly volatile metal particle cloud clearly.

In this study, the experiments on flame propagating through particle clouds of zirconium, a hardly volatile metal, have been conducted. Propagation and temperature characteristics of the combustion zone, structure of flame, and combustion courses of zirconium particles were elucidated.

2. Experimental

2.1. Experimental apparatus

The experimental system is schematically shown in Fig. 1, which is composed of a gas supplying unit, a combustion chamber, a thermocouple, an ignition system, a data recorder, a high-speed video

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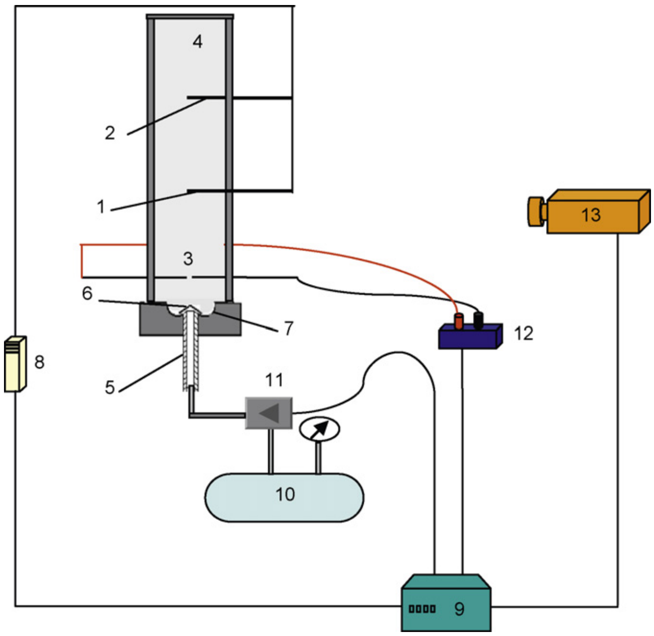

Fig. 1. Experimental system.

camera and a synchronization controller. The combustion chamber with square cross-section of 80 mm × 80 mm is 500 mm height, and its top end is opened. To observe the flame propagation process conveniently, two sides of the chamber were made of glass and the other two sides were made of stainless steel. The combustion chamber was also provided with a gas nozzle, a dispersing cone and a sample dish. The ignition system consists of a high voltage transformer and a pair of tungsten wire electrodes with 0.4 mm diameter. The distance between the tips of the two electrodes is approximately 5 mm. A high voltage transformer with the output of 30 kV was used to produce an ignition spark. The spark duration is 0.01 s. The electrodes were set in the middle of the chamber, which locates 5 cm above the bottom of the chamber.

2.2. Experimental procedure and conditions

In this study, the pure zirconium dust was used. The diameters of zirconium particles are all less than 42 μm. Certain mass of zirconium dust weighed by a balance was put on the sample dish evenly. Then the zirconium dust was dispersed into the chamber by pressurized air and ignited by an electric spark after 100 ms delay. The flame propagation process was recorded by the high-speed video camera.

In the test, the startup time of the high-speed video camera, the data recorder and the high voltage igniter and on–off time of the electromagnetic valve was controlled by a synchronization controller. The detailed experimental conditions were given as follows:

Ignition voltage: 30,000 V; discharge period: 0.01 s; framing rate of high-speed video camera: 2000 frames/s or 6000 frames/s; injection time: 0.5 s, pressure in the tank: 0.4 MPa.

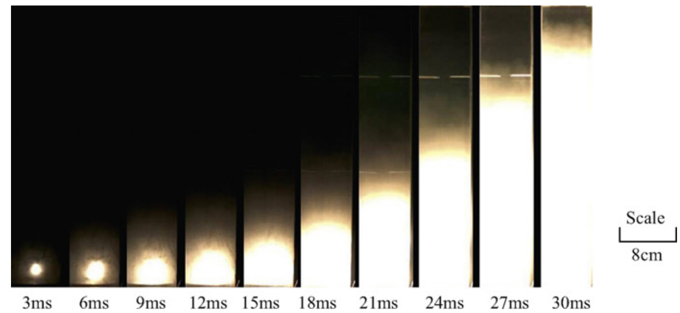

Fig. 2. High-speed photographs of zirconium particles cloud flame. Zirconium particle concentration: 0.51 kg/m^3, framing rate: 2000 frames/s.

3. Results and discussion

3.1. Structure of flame and combustion course of zirconium particle

Fig. 2 shows a series of typical high-speed video photographs recorded by a high-speed video camera with a normal lens, which represent the flame propagation processes through the zirconium dust cloud in the vertical chamber. As shown in Fig. 2, after ignition, flame kernel grows with a white luminous zone, the outline of which is a regular shape. First, flame propagates along all the directions. After the flame reaches the bottom of the chamber, it propagates only upwards because of the closed bottom of the chamber. From 12 ms to 27 ms, the flame propagates upwards with a large velocity. Because the combustion of zirconium particles needs a few time, the luminous zone becomes longer during the propagation process and it exists in a larger zone.

Fig. 3 shows a typical series of high-speed photographs with a microscopic lens. It can be found that the combustion zone consists of many burning zirconium particles. According to the experimental results mentioned above and chemical and physical properties of zirconium and its oxide, the combustion process of a zirconium particle can be explained as the following interpretation. The solid zirconium particle is heated when it approaches the combustion zone front. Then as the zirconium particle temperature rises, an oxidation reaction would happen at the surface of zirconium particle, which forms an oxide coating on the particle surface. Because the boiling point (4650 K) [14] of zirconium is much higher than the adiabatic flame temperature (2870 K) of zirconium particle cloud combustion, there is not zirconium vapor produced from the zirconium particle in the zirconium particle combustion process. Thus, the zirconium particle combustion only takes place at the surface of zirconium particle.

3.2. Propagation velocities of combustion zone

The leading edge of the combustion zone is observed to be smooth and clear. The propagation velocity of the combustion zone can be determined by examining the movement of its leading edge. Fig. 4 shows the relationship between the distance from the ignition point and the propagation velocity of the combustion zone with

Fig. 3. High-speed photographs with a microscopic lens of zirconium particles cloud flame. Zirconium particle concentration: 0.34 kg/m^3, framing rate: 6000 frames/s, t: time from ignition.

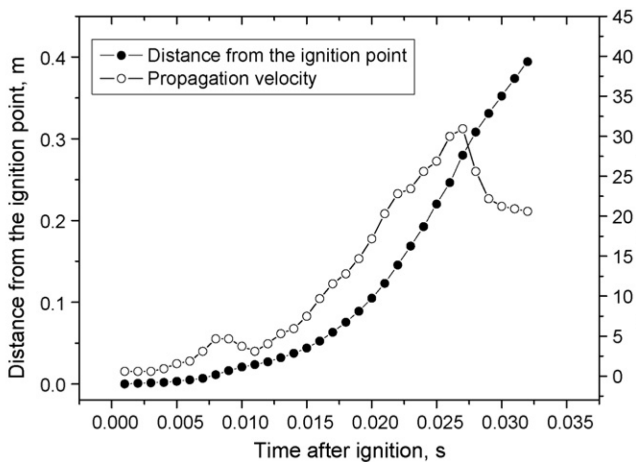

Fig. 4. Relationship between the distance from the ignition point and the propagation velocity of the combustion zone with time from the ignition. Zirconium particle concentration: 0.51 kg/m^3.

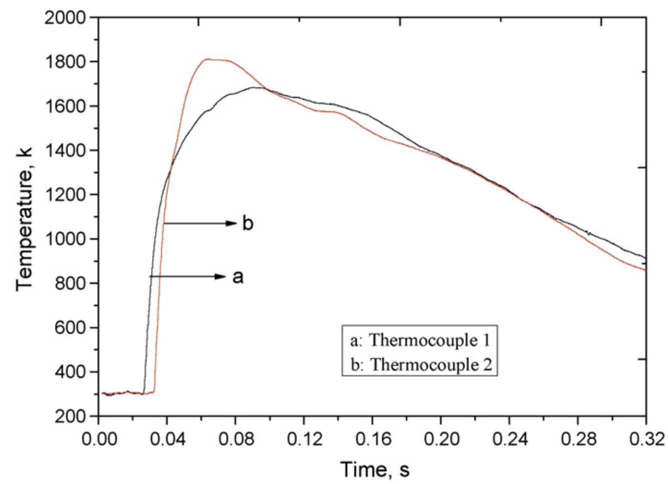

Fig. 6. Temperature profiles of the combustion zone propagating through a zirconium particles cloud. Zirconium particle concentration: 0.51 kg/m^3.

time after ignition. It is seen that the propagation velocity of the combustion zone is not constant. The combustion zone propagates at a constant velocity in the first 3 ms after ignition and then it has a slight acceleration process from 3 ms to 9 ms. Because of the influence of the chamber wall, the propagation of the combustion zone has a slight deceleration process from 9 ms to 11 ms. Then it has an obvious acceleration process from 11 ms to 27 ms, and the combustion zone has a maximum flame propagation velocity of 30.9 m/s at 27 ms. Then it decreases from 27 ms to 33 ms.

Fig. 5 shows the relationship between the measured propagation velocity of the combustion zone and the time from the ignition for various concentrations of the zirconium particle cloud. It is found that the propagation velocities of the combustion zone with different concentration of the zirconium particle cloud are not the same, which must be caused by the difference of the heat produced by the combustion of the different concentration zirconium particle.

3.3. Temperature characteristic of combustion zone

In this study, two thermocouples, made of 25 μm diameter wires of Pt/Pt (13%) Rh, were used to detect the combustion zone temperature. They were fixed in the middle of the stainless steel side and located 20 cm and 35 cm above the bottom of the chamber respectively.

Fig. 6 shows the typical temperature profiles of the combustion zone propagating through a zirconium particle cloud (0.51 kg/m^3). When the combustion zone front approaches the thermocouples, both temperature profiles of thermocouples 1 and 2 increase to the maximum value in tens of milliseconds, which indicates that the combustion reaction of zirconium particles is very violently and releases much heat in short time. The temperatures measured by thermocouples 1 and 2 increase at about 27 ms and 31 ms respectively, which both increase slowly during the initial 1.5 ms approximately. After 1.5 ms, they both increase quickly, and reach the maximum 1684k and 1813k at 94 ms and 64 ms respectively. Compared with temperature profile, (a) the increase rate of temperature profile (b) is larger than that of temperature profile (a) in the incipient stage. The maximum temperature measured by thermocouple 2 is also larger than that measured by thermocouple 1. This phenomenon must be caused by much richer oxygen supplied on the top of chamber.

To obtain accurate data, the effect of thermal inertia for the thermocouple is taken into account. It is assumed that convective heat transfers to the junction of the thermocouple predominates over radiative heat transfer, thus the temperature value measured using

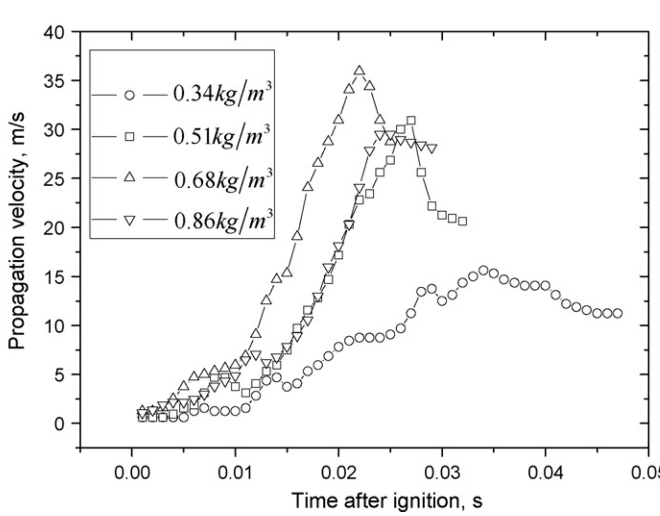

Fig. 5. Variation of propagation velocity of the combustion zone with the time after ignition for different particle concentrations.

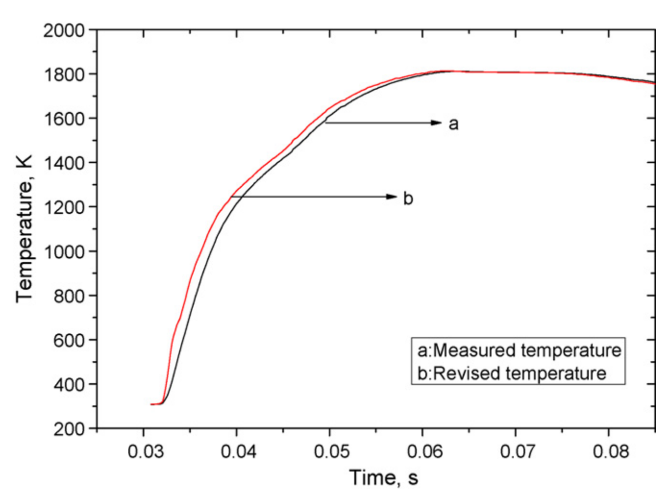

Fig. 7. Temperature profiles of the combustion zone propagating through a zirconium particles cloud (thermocouple 2). Zirconium particle concentration: 0.51 kg/m^3.

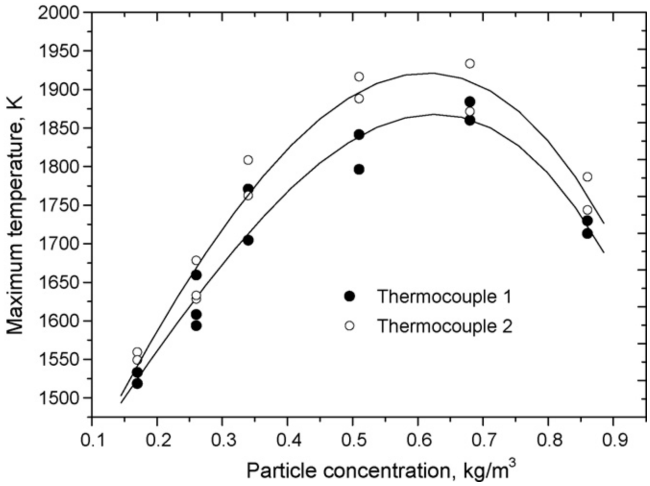

Fig. 8. Relationship between the maximum temperatures of the combustion zone and zirconium particle concentration.

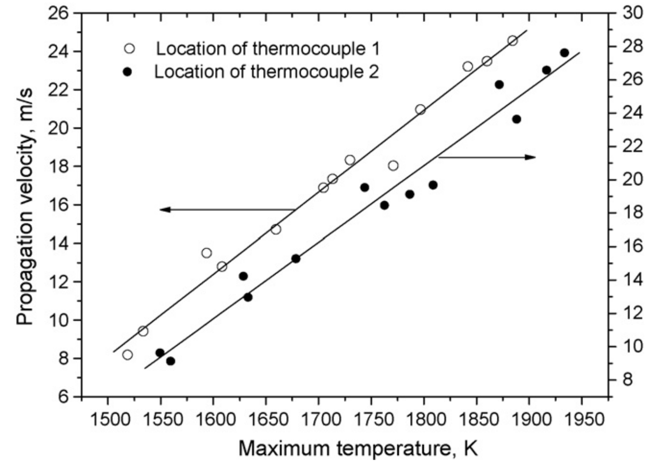

Fig. 10. Relationship between the maximum temperature and the propagation velocity of the combustion zone.

the thermocouple can be compensated as follows [15]:

$$T = T_m + \tau \frac{dT_m}{dt} \quad (1)$$

Where T_m is the temperature measured by the thermocouple, τ is a time constant of the thermocouple. In our experiments, the value of τ is 6.7×10^{-3} s approximately. Take the temperature profile (b) in Fig. 6 for an example, the temperature revised by Eq. (1) is shown in Fig. 7. It shows that there are a few milliseconds delay approximately between the measured temperature and revised temperature.

The maximum temperature of the combustion zone varies with zirconium particle concentration. Fig. 8 shows the maximum temperature measured by thermocouples 1 and 2 for several concentrations of zirconium particles. It can be seen that the maximum temperatures measured by thermocouples 1 and 2 increase with the concentration of zirconium particles at the lower concentration, reach the maximum value at the concentration 0.62 kg/m^3 approximately, and then decrease at the higher concentration.

Fig. 9 shows the variations of the propagation velocity of the combustion zone and the maximum temperature versus the zirconium particle concentration. The combustion zone propagation velocity means the instantaneous propagation velocity of combustion zone which reaches the location of thermocouple. As shown in Fig. 9, the tendencies of the combustion zone propagation velocity and the maximum temperature with the zirconium particle concentration are almost the same. They both take their maximum values at a concentration of 0.60–0.65 kg/m^3 zirconium particles. Fig. 10 shows the relationship between the maximum temperature of combustion zone and the instantaneous propagation velocity of combustion zone, which reaches the location of thermocouple. The result indicates that the propagation velocity of the combustion zone has a linear relationship with its maximum temperature.

4. Conclusions

Flame propagating through zirconium particle cloud has been examined experimentally, and the temperature profile of the combustion zone propagating through a zirconium particle cloud was also measured experimentally by a thermocouple, the following results were obtained.

Combustion zone propagating through a zirconium particle cloud consists of many burning zirconium particles. The combustion of zirconium particle only takes place at the surface of the zirconium particle.

In a vertical chamber, the propagation velocity of the combustion zone propagating through a zirconium particle cloud is not constant; combustion zone propagation velocities with different concentration of the zirconium particle cloud are not the same.

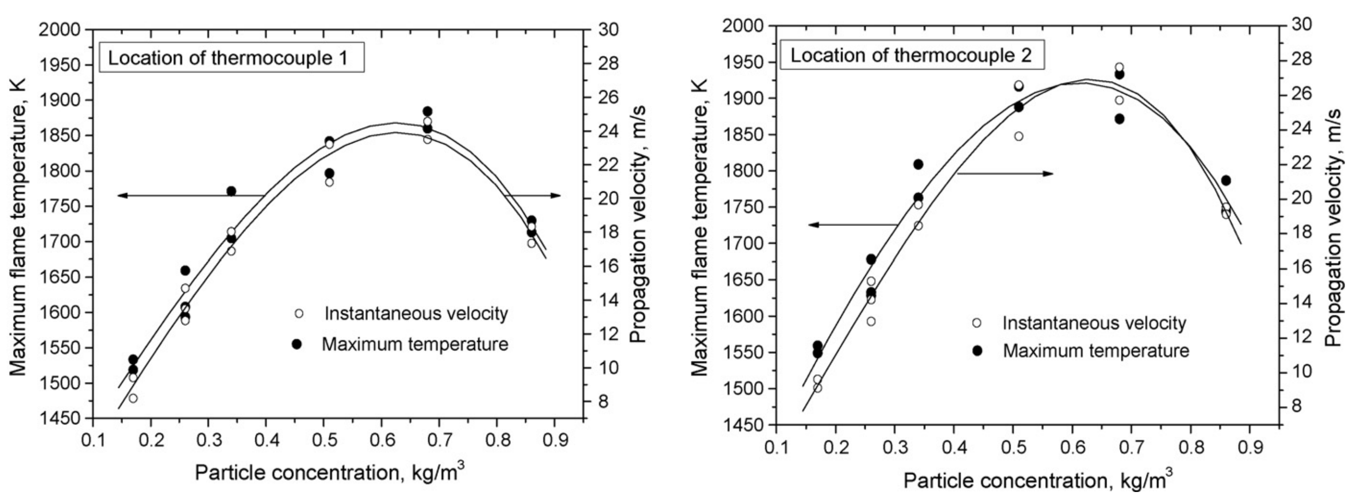

Fig. 9. Variations of the propagation velocity of the combustion zone and the maximum temperature with the concentration of zirconium particles.

Because the combustion reaction of zirconium particles is very violently and releases much heat in a short time, the temperature measured by thermocouples 1 and 2 reaches much high temperature quickly. With the concentration of zirconium particles, the maximum temperatures of combustion zone increase at the lower concentration, take the maximum value at a certain concentration, and then decrease at the higher concentration.

The tendencies of the combustion zone propagation velocity and the maximum temperature with the zirconium particle concentration are almost the same. They both take their maximum values at a concentration of 0.60–0.65 kg/m^3 zirconium particles. The propagation velocity of the combustion zone has a linear relationship with its maximum temperature.

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References

[1] K.L. Cashdollar, Coal dust explosibility, J. Loss Prev. Process Ind. 9 (1) (1996) 65–76.
[2] A.E. Dahoe, J.F. Zevenbergen, S.M. Lemkowitz, B. Scartetl, Dust explosions in spherical vessels: the role of flame thickness in the validity of the cube-root law, J. Loss Prev. Process Ind. 9 (1) (1996) 33–44.
[3] M. Hertzberg, I.A. Zlochower, K.L. Cashdollar, Metal dust combustion, explosion limits, pressures, and temperatures, in: 24th Symposium (International) on Combustion, Pittsburgh, 1992, pp. 1827–1835.
[4] M. Hertzberg, K.L. Cashdollar, I.A. Zlochower, Flammability limit measurements for dust and gases, in: 21st Symposium (International) on Combustion, The Combustion Institute, Pittsburgh, PA, 1986, pp. 303–313.
[5] N. Chawla, P.R. Amyotte, M.J. Pegg, A comparison of experimental methods to determine the minimum explosible concentration of dusts, Fuel 75 (6) (1996) 654–658.
[6] C. Proust, B. Veyssiere, Fundamental properties of flames propagating in starch dust-air mixtures, Combust. Sci. Technol. 62 (4–6) (1988) 149–172.
[7] O.S. Han, M. Yashima, T. Matsuda, H. Matsui, A. Miyake, T. Ogawa, Behavior of flames propagating through lycopodium dust clouds in a vertical duct, J. Loss Prev. Process Ind. 13 (6) (2000) 449–457.
[8] O.S. Han, M. Yashima, T. Matsuda, H. Matsui, A. Miyake, T. Ogawa, A study of flame propagation mechanisms in lycopodium dust clouds based on dust particles behavior, J. Loss Prev. Process Ind. 14 (6) (2001) 153–160.
[9] J.L. Chen, R. Dobashi, T. Hirano, Mechanisms of flame propagation through combustible particle clouds, J. Loss Prev. Process Ind. 9 (3) (1996) 225–229.
[10] W.J. Ju, R. Dobashi, T. Hirano, Reaction zone structures and propagation mechanisms of flames in stearic acid particle clouds, J. Loss Prev. Process Ind. 11 (6) (1998) 423–430.
[11] J.H. Sun, R. Dobashi, T. Hirano, Structure of flames propagating through metal particle clouds and behavior of particles, in: 27th Symposium (International) on Combustion, Pittsburgh, 1998, pp. 2405–2411.
[12] J.H. Sun, R. Dobashi, T. Hirano, Concentration profile of particles across a flame propagating through an iron particle cloud, Combust. Flame 134 (4) (2003) 381–387.
[13] J.H. Sun, R. Dobashi, T. Hirano, Structure of flames propagating through aluminum particles cloud and combustion process of particles, J. Loss Prev. Process Ind. 19 (2006) 769–773.
[14] R.C. Weast, CRC Handbook of Chemistry and Physics, 59th ed., CRC Press Inc., Florida, 1978–1979, B-65.
[15] A. Ballantyne, J.B. Moss, Fine wire thermocouple measurements of fluctuating temperature, Combust. Sci. Technol. 17 (1977) 63–72.