An experimental investigation of flame propagation in clouds of silicon dust dispersed in air, hydrogen-air mixtures, and hybrid Si-H_2-air mixtures

Trygve Skjold
University of Bergen & GexCon AS
Bergen, Norway

1 Introduction

Dust explosions pose a hazard whenever a sufficient amount of combustible material is present as fine powder, the powder can be dispersed in air to form an explosive dust-air cloud within a sufficiently confined volume, and there is an ignition source present. Industrial dust explosions have evolved with technological changes in agriculture, manufacturing, mining, transportation and storage. Major accidents during the eighteenth century were dominated by coal dust explosions in mines, often combined with hybrid mixtures of methane and coal, and grain dust explosions in mills. Whereas coal and grain dust explosions still represent significant hazards in the twenty-first century, numerous other combustible materials are also processed, transported or stored as powder.

Highly purified silicon (Si) has numerous applications in industry, including the manufacturing of semiconductors and solar panels. Silicon is a combustible material, and the hazardous nature of silicon dust is evident from the devastating dust explosion that took place on 18 October 1972 in the silicon grinding plant at Elkem Bremanger. Five people lost their lives and four people were severely injured in the accident [1]. Since production and use of silicon in certain industrial settings may entail flammable gases such as hydrogen [2], it is also of interest to consider hybrid mixtures. The elements silicon and hydrogen are also relevant for production of semiconductors in the form of gaseous silane (SiH_4), which is a toxic and extremely flammable chemical compound [3], but this aspect will not be investigated in the current paper.

Whereas combustion parameters for most gaseous fuels can be found in the open literature, at least for combustion in air at near ambient initial conditions, this is not the case for dust-air suspensions and hybrid mixtures. Since the rate of combustion in a heterogeneous system depends strongly on particle size, or rather the particle size distribution, it is not straightforward to generalize results. Most methods for estimating the consequences of industrial dust and hybrid explosions rely on empirical correlations, and explosion parameters for a given sample are typically determined in standardized tests performed in constant volume explosion vessels [1]. The primary motivation for the present work has been to gain improved understanding of flame propagation in suspensions of silicon dust in air, as well as hybrid Si-H_2-air mixtures, with a view to develop improved models and methods for assessing and reducing the risk posed by accidental explosions in the process industry. Most of the data for silicon and hydrogen have been presented before [4-5], and similar work has been reported by others [6-8].

Correspondence to: trygve@gexcon.com

2 Experiments

The experiments were performed in two explosion vessels at the University of Bergen [4]: a vessel similar to the standard 20-litre vessel developed by US Bureau of Mines (USBM), and a cubical 20-litre vessel. Both vessels were equipped with the dispersion system developed by Kuhner AG for the 20-litre Siwek sphere. The effect of nominal dust concentration, ignition delay time and particle size on explosion pressure and maximum rate of pressure rise were investigated for nine different lots of jet-milled silicon, or Silgrain™, from Elkem Bremanger. Most of the silicon tests were performed in the 20-litre cubical vessel fitted with a rebound nozzle. The tests with hydrogen and hybrid mixtures were performed in the USBM vessel. The experimental procedure is described in detail elsewhere [4].

3 Results and discussion

Figure 1 shows particle size distributions for nine lots of jet-milled silicon dust, as determined by low angle laser light scattering (LALLS) with a Malvern Instruments SN.22 with focal length 100 mm and beam length 2.2 mm, and particles dispersed in air. Some results are also included from a Malvern Mastersizer X (long bed version 2.18), with focal length 100 mm and beam length 2.4 mm, and particles dispersed in water. Table 1 summarizes selected particle size measures for the various samples. Although the results depend on the measurement technique, the overall trend is reasonably consistent. Figure 2 shows scanning electron microscope (SEM) pictures of silicon dust, as well as residue gathered from the explosion vessels after explosions.

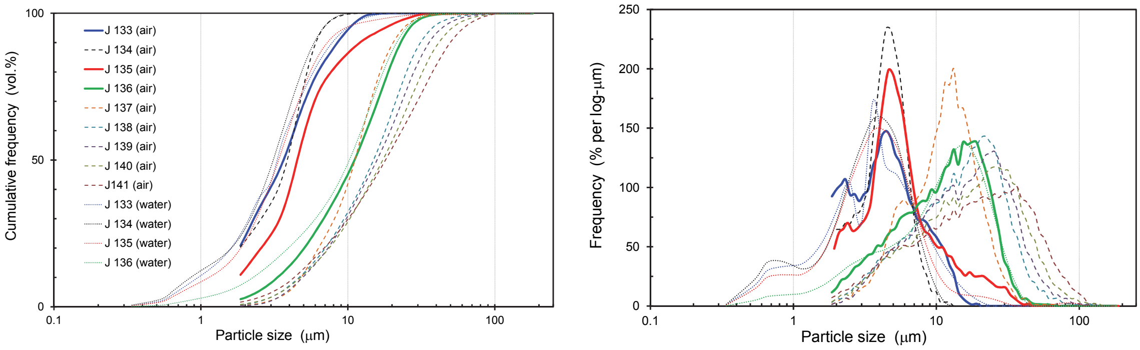

Figure 1. Cumulative particle size distribution $F(x)$ and frequency distribution $dF/dlog(x)$ for nine lots of jet-milled silicon dust (Silgrain™) from Elkem Bremanger; LALLS with samples dispersed in air or water.

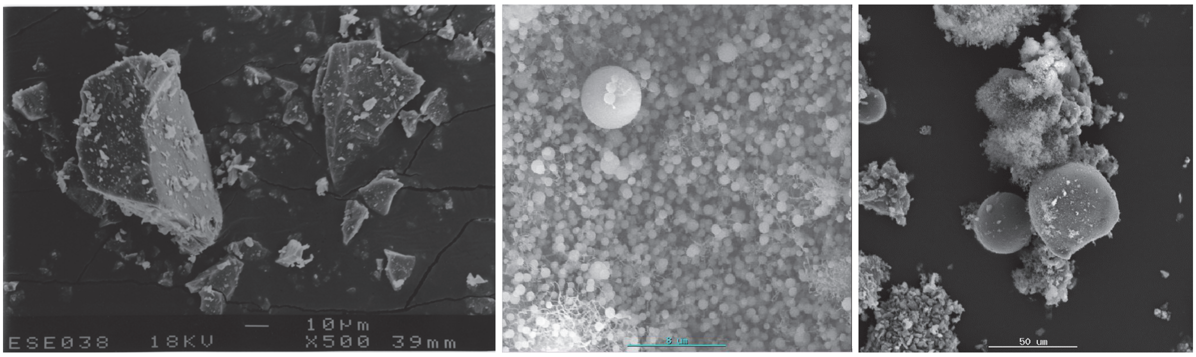

Figure 2. SEM pictures of silicon dust (left), white residue after explosions in a 'fuel lean' silicon-air mixture (centre), and brown residue after explosions in a 'fuel rich' silicon-air mixture (right).

Table 1: Selected particle size measures (μm) for jet milled silicon measures by LALLS.

Lot no.	$d_{4,3}$		$d_{3,2}$		$d_{v,\,0.1}$		$d_{v,\,0.5}$		$d_{v,\,0.9}$	
J 133	4.3	4.4*	2.4	2.5*	1.1	1.4*	3.5	3.8*	8.6	8.8*
J 134	3.9		2.5		1.3		4.0		6.3	
J 135	4.9	6.2*	2.6	3.3*	1.2	1.9*	3.7	4.7*	7.9	12.5*
J 136	12.2	12.8*	5.5	6.9*	2.5	3.4*	11.2	11.5*	23.5	23.7*
J 137	12.5		9.0		5.0		11.6		21.0	
J 138	17.7		10.0		4.8		15.5		32.3	
J 139	19.5		11.0		5.1		16.4		37.0	
J 140	21.9		10.7		4.9		17.9		43,1	
J 141	24.2		9.9		4.5		18.7		51.6	

* Particles dispersed in water

Figure 3 summarizes explosion pressures and maximum rates of pressure rise for three of the nine silicon samples from Table 1. Particle size clearly influences the results, but contrary to expectations the J 135 sample yields higher rates of pressure rise than the finer J 133 sample. Figure 4 illustrates the effect of varying the ignition delay time, measured from onset of dust dispersion, for the same three samples. By varying the ignition delay time, it is possible to achieve different levels of turbulence inside the vessel. However, the fact that the explosion pressure drops quite rapidly, even for quite moderate ignition delays, suggests that a significant fraction of the dust leaves the suspension. Figure 5 illustrates the effect of ignition energy and particle size for all nine samples of jet-milled silicon dust. Whereas the explosion pressure is not significantly influenced by the ignition source, the use of chemical igniters yields a significant increase in the rate of pressure rise. This effect is probably caused by the volumetric nature of the energy release from the chemical igniters, since the flame front reaches a given radius in shorter time, and combustion can proceed at a higher level of turbulence.

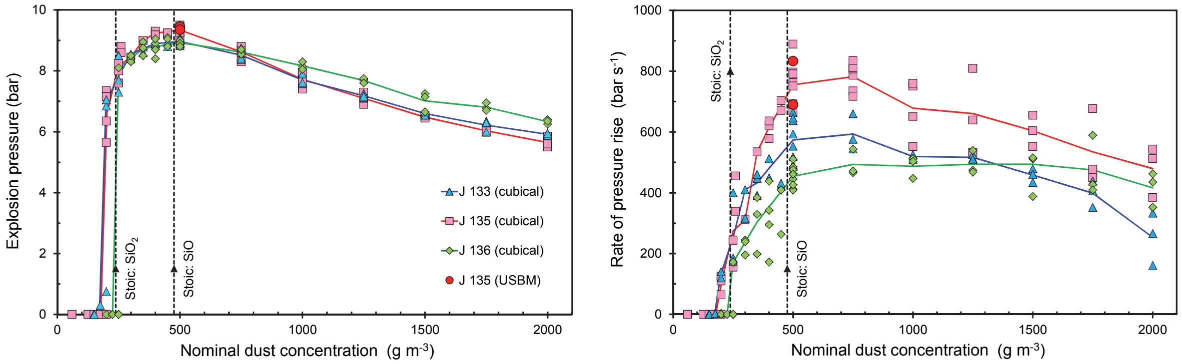

Figure 3. Effect of nominal dust concentration on explosion pressure and maximum rate of pressure rise.

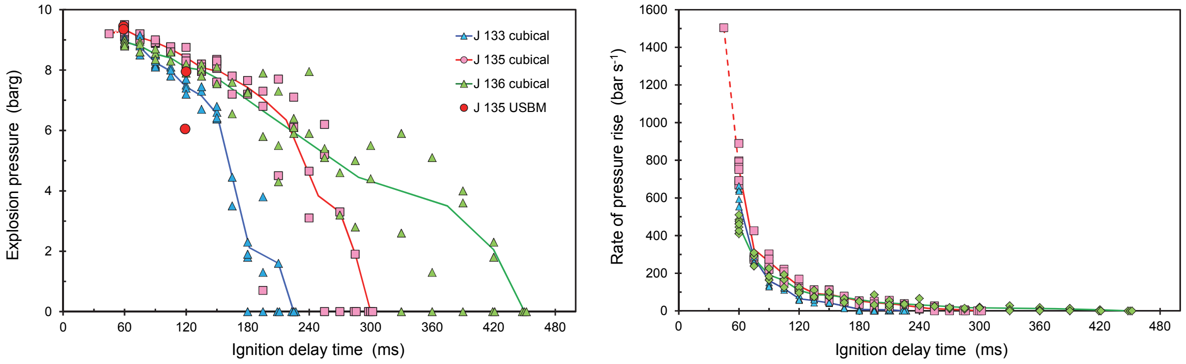

Figure 4. Effect of ignition delay time on explosion pressure and maximum rate of pressure rise for three samples of jet-milled silicon – nominal dust concentrations 500 g m^{-3} and ignition by 6 J arc discharges.

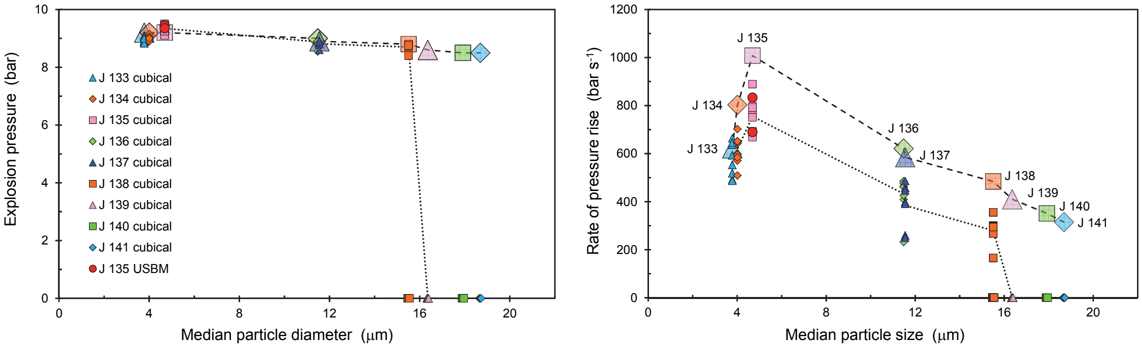

Figure 5. Effect of particle size and ignition energy on explosion pressure and maximum rate of pressure rise for three lots of jet-milled silicon dust, nominal dust concentrations 500 g m^{-3} – the smaller symbols represent ignition by 6 J electrical discharges and the large symbols represent ignition by 2 x 5 kJ chemical igniters.

The observation that the explosion pressure decreases quite rapidly with increasing ignition delay time triggered further investigations, and it was found that the colour of the residue, primarily silicon oxides, was a function of nominal dust concentration for a given ignition delay time, as well as a function of ignition delay time for a given nominal dust concentration. Figure 6 summarizes these observations for three lots of jet-milled silicon dust. The results are consistent with the observations in Figure 4, and the primary explanation appeared to be rapid depositing of dust on the vessel walls [4].

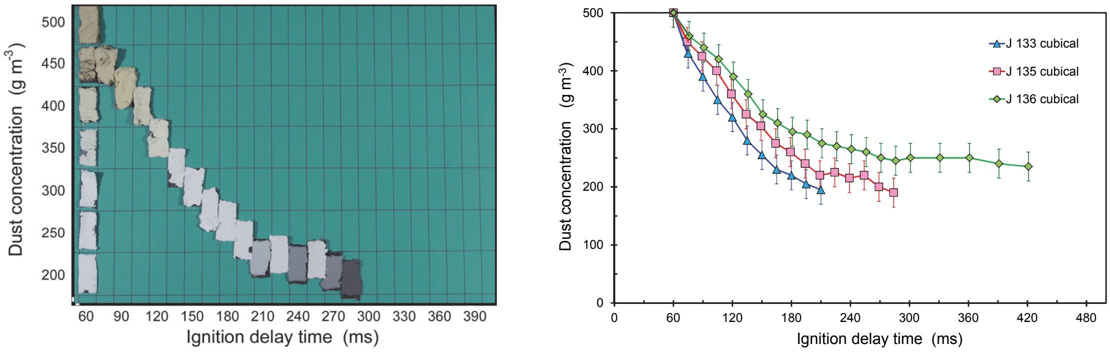

Figure 6. Effect of ignition delay time on oxide colour for a nominal dust concentration of 500 g m^{-3} (left; the samples placed vertically represent tests with varying nominal dust concentration for J 135 and a fixed ignition delay time), and summary of results from visual observation of residue in the combustion chamber (right).

Figure 7 summarizes the results obtained for hydrogen-air mixtures. The measured explosion pressures are in fairly good agreement with thermodynamic calculations, and not significantly influenced by the level of turbulence. The maximum rate of pressure rise, on the other hand, varies significantly with for different ignition delay times.

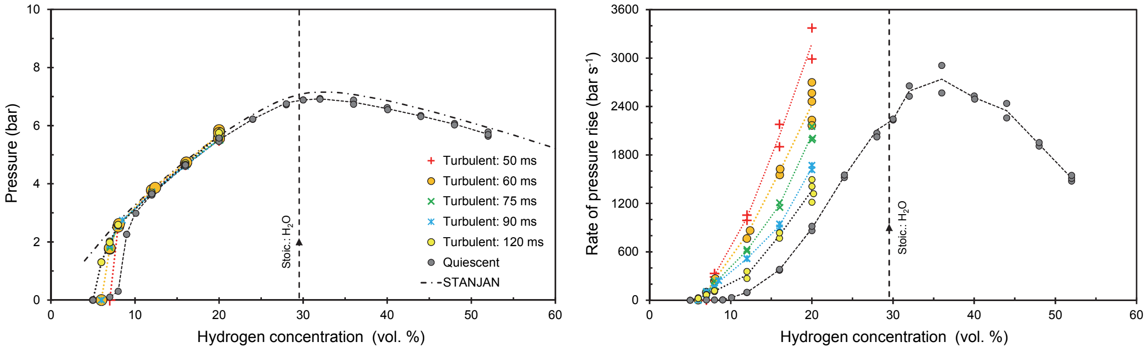

Figure 7. Effect of hydrogen concentration and ignition delay time on the explosion pressure and maximum rate of pressure rise measured in the 20-litre USBM vessel.

Figure 8 summarizes the results obtained for hybrid mixtures of Si and H_2, including results for pure Si-air suspensions and H_2-air mixtures, for two ignition delay times: 60 and 120 ms. Note that the nominal silicon concentrations increases as the hydrogen concentrations increases from left to right in both plots. The mixture compositions were selected to cover parts of the concentration range where both the explosion pressure and the maximum rate of pressure rise change rapidly for both fuels (ref. Figures 3 and 7). For nominal silicon concentrations in excess of 250-300 g m^{-3}, neither the explosion pressure nor the maximum rate of pressure rise seems to be significantly influenced by the hydrogen.

When a modest amount of hydrogen is 'replaced' with silicon, both the explosion pressure and the rate of pressure rise are reduced. The reduction in pressure is consistent with the results for pure hydrogen mixtures, but the results for the maximum rate of pressure rise indicate a certain inhibiting effect of the silicon. Figure 9 shows selected pressure-time curves for some of the tests. The results obtained for 200 g m^{-3} silicon and 12 vol.% hydrogen indicate that the initial rapid pressure rise is caused primarily by combustion of hydrogen. The slower pressure rise that follows is most likely a result of delayed combustion of silicon particles.

Future work should involve a more thorough investigation of hybrid Si-H_2 mixtures. The interpretation of the results presented in Figures 8 and 9 could benefit from a separate series of experiments aimed at investigating the effect of gradually adding more silicon dust to H_2-air mixtures with a fixed concentration. It would also be interesting to experimentally verify whether the secondary pressure rise, as observed for mixtures of 200 g m^{-3} silicon and 12 vol.% hydrogen, is a result of purely volumetric combustion, or if it is the result of a secondary flame front propagating through the dust cloud. The tests in constant volume explosion vessels should be complemented with experiments at constant pressure in burners, vertical tubes, or transparent balloons [10].

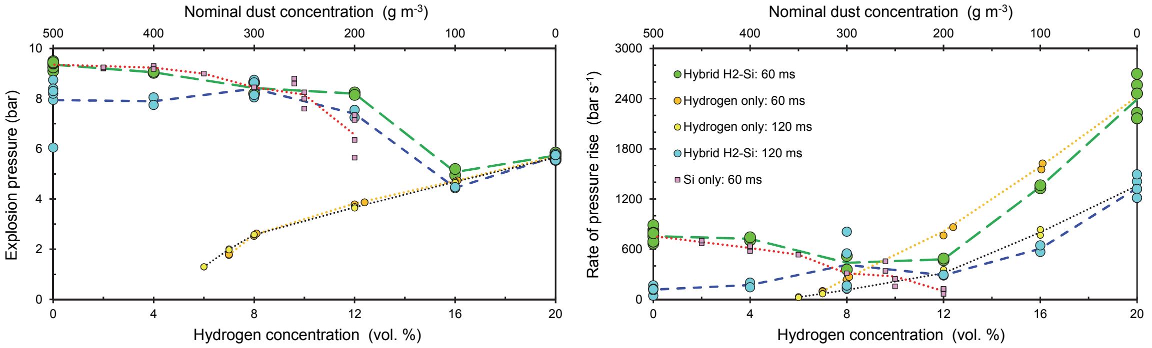

Figure 8. Explosion pressure and maximum rate of pressure rise for Si-H_2 hybrid mixtures.

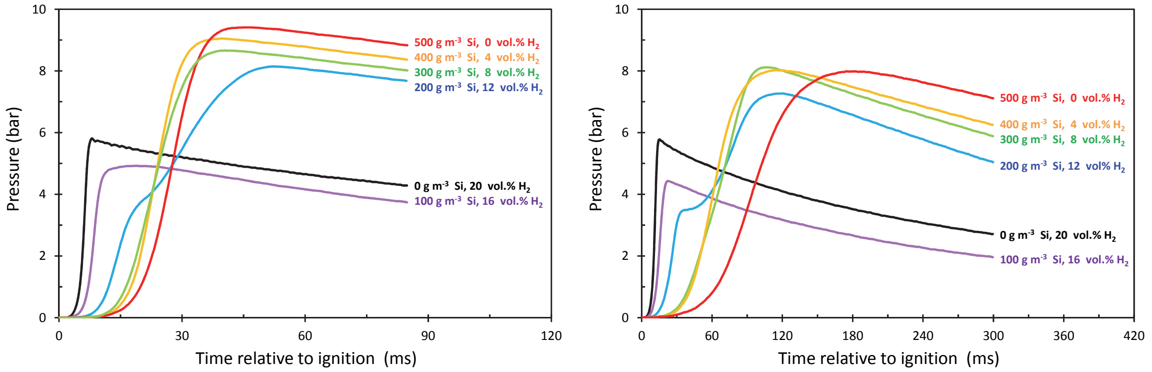

Figure 9. Characteristic pressure-time curves for hybrid mixtures of jet-milled silicon dust (J 135) and hydrogen, ignited to deflagration by 6 J electrical discharges in the 20-litre USBM vessel; ignition delay 60 ms (left) and 120 ms (right).

4 Conclusions

Flame propagation in dust clouds entails ‘premixed combustion with non-premixed substructures’ [9], and detailed modelling from first principles is not straightforward. The process of generating mechanical suspensions in constant volume explosion vessels entails transient turbulent flow conditions that complicate the task of determining fundamental combustion parameters, such as burning velocity and flame thickness, as well as actual dust concentration and turbulence parameters.

The results obtained for silicon dust is consistent with the findings in previous studies [6-8]. However, the rapid decrease in dust concentration with increasing ignition delay time represents a severe limitation with respect to the applicability of results from constant volume explosion vessels at laboratory scale. The rate of dust settling on the vessel walls appears to be strongly related to both type of dust and particle size distribution.

The results obtained for hybrid Si-H_2 mixtures demonstrate that typical parameters used for design of explosion protection systems, such as explosion pressure and maximum rate of pressure rise, can vary significantly depending on the relative composition of the hybrid mixture. It is not obvious that design based on parameters for the more reactive component will be sufficient, since the specific energy release per unit air may be higher for the other component.

Testing of dusts and hybrid mixtures in standardized 20-litre explosion vessels will continue to play an important role in practical process safety for years to come. However, such tests come with several inherent limitations with respect to both control of experimental conditions and accuracy in results, and it is foreseen that alternative, or at least complementary, test methods will evolve.

References

[1] Eckhoff, R.K. (2003). Dust explosions in the process industries. Third edition. Gulf Professional Publishing, Amsterdam.

[2] Fthenakis, V.M. (2003). Overview of potential hazards. Chapter VII-2 in: Practical handbook of photovoltaics, T. Markvart & L. Castaner (Eds.), Elsevier, Amsterdam (ISBN 1-856-17390-9).

[3] Koda, S. (1992). Kinetic aspects of oxidation and combustion of silane and related compounds. Prog. Energy Combust. Sci. 18: 513-528.

[4] Skjold, T. (2003). Selected aspects of turbulence and combustion in 20-litre explosion vessels. Candidatus Scientiarum thesis, University of Bergen. Available (accessed 4 May 2013): https://bora.uib.no/bitstream/handle/1956/1631/Masterthesis_Skjold.pdf

[5] Middha, P., Skjold, T. & Dahoe, A.E. (2006). Turbulent and laminar burning velocities of hydrogen-air mixtures from constant volume explosions in a 20-litre vessel. Poster Thirty-first Symposium (Int.) on Combustion, 6-11 August 2006, Heidelberg, Germany.

[6] Eckhoff, R.K., Parker, S.J., Gruvin, B., Hatcher, M. & Johansson, T. (1986). Ignitability and Explosibility of Silicon Dust Clouds. Journal of the Electrochemical Society, 133: 2631-2637.

[7] van der Wel, P., Lemkowitz, S., Scarlett, B. & van Wingerden, K. (1991). A Study of Particle Factors Affecting Dust Explosions. Particle & Particle Systems Characterization, 8, pp. 90-94.

[8] Matsuda, T., Yashima, M., Nifuku, M & Enomoto, H. (2001). Some aspects in testing and assessment of metal dust explosions. J. Loss Prevention Process Ind., 14: 449-453.

[9] Williams, F.A. (1996). Lectures on applied mathematics in combustion: Past contributions and future problems in laminar and turbulent combustion. Physica, 20D: 21-34.

[10] Skjold, T. (2013). A constant pressure dust explosion experiment. J. Loss Prevention Process Ind., 26: 562-570.