

Influence of the Particle Size Distribution on Dust Explosion: How to Choose the Right Metrics?

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It is well established that the 'diameter' of a powder and its explosivity are strongly correlated. But, in fact, what diameter should be considered? The mass median diameter d_{50} is often used as an indicator of the particle size distribution (PSD) of a powder, but this choice is not always consistent with the mechanisms involved in a dust explosion. Moreover, authors mainly refer to the initial diameter of the powder, whereas it is more relevant to consider its characteristics at the exact time of the ignition. Many other metrics can be of interest concerning dust explosion: Sauter diameter, d_{10} , solid surface concentration. To highlight these points, tests were performed on raw starch powders of 24 μm d_{50} and 0.49 $\text{m}^2\cdot\text{g}^{-1}$ specific surface area. They were mechanically agglomerated with an Instron press and then coarsely ground by a blade mill. Different size classes up to 283 μm d_{50} and 0.16 $\text{m}^2\cdot\text{g}^{-1}$ specific surface area were selected by sieving. The PSD of the samples were compared before and during their injection in the explosion vessels. The influence of the actual PSD on their explosion severity and minimum ignition energy (MIE) was determined. It has been demonstrated that the evolution of the MIE with regard to PSD is subject to a threshold phenomenon. Moreover, if the mass median diameter can be interesting for kinetics controlled by homogeneous chemical reaction and directly related to the fuel equivalent ratio, the reactive surface is more relevant when the combustion is controlled by surface reaction or diffusion to the surface as it is the case for the various starch samples. Chromatography measurements of the combustion gases confirm these results. Finally, when the agglomerates are partially fragmented by the injection, both the agglomerate and primary diameters must be considered.

1. Introduction

The agglomeration phenomenon results from an inter-particle force called adhesion. The adhesion between two particles is due to effective short-range forces; among them are the Van der Waals forces, the electrostatic forces, the magnetic and capillary forces (Rumpf, 1962). They vary as a function of the particle nature but also depend on the particle size: the smaller the particles, the more they will tend to agglomerate.

A dust explosion can occur when fine particles are dispersed in the air in sufficient concentration and with a source of ignition. The particle size distribution has a major influence on the dust cloud homogeneity, its turbulence level and on its reactivity (Callé et al., 2005). Modifying the particle size distribution (PSD), by granulation or by breaking the agglomerates will then impact these latter parameters and the explosion severity. Choosing the right indicator of the PSD as a function of the phenomenon to be considered is essential (Li et al., 2016; Yuzuriha et al., 2017). The mass median diameter d_{50} of the powder usually plays this role. However, it is often determined by wet dispersion, which does not correspond to the dispersion conditions of the dust cloud. In situ particle size measurement should be preferred, but many other metrics can be of interest concerning dust explosion: the Sauter diameter $d_{3,2}$, the maximum diameter of the 10% finer particles, solid surface concentration. In order to highlight these points, the minimum ignition energy and

explosion severity of various starch samples were determined. Their PSD was modified by dry granulation, which means that the predominant forces will therefore be the Van der Waals and electrostatic forces.

2. Materials and methods

2.1 Powders characteristics and granulation

Experiments were performed on starch whose properties are described in Table 1. The particle size measurements were initially done in ethanol in order to limit a potential swelling effect (Malvern – Mastersizer 2000). In order to study the influence of the powder agglomeration on its explosivity, agglomerates were formed by dry granulation. Starch tablets were made by compression of the raw starch powder, without additive, using a press (Instron 5569). The maximum force applied on the tablet was set at 20 kN after several tests: a weaker force leads to too friable tablets, a stronger one do not bring significant changes in the agglomerates shapes and sizes. The tablets were subsequently ground and sieved. Two ranges of agglomerate diameters were chosen: 112-224 μm and 224-450 μm . Only the second range will be studied here in order to highlight the impact of dust agglomeration on its explosivity.

Table 1: Particle size distribution in ethanol of raw starch and starch agglomerates

d (μm)	d_{10}	d_{50}	d_{90}	Specific surface area ($\text{m}^2 \cdot \text{g}^{-1}$)
Raw starch	11.3	24.2	44.8	0.49
Agglomerates (224-450)	19.2	283.5	579.1	0.16

Table 1 shows that, even after sieving, the particle size distribution of the agglomerates is far from being unimodal. Fine particles adhere to the agglomerates surface, which explains the value of the diameter d_{10} . Scanning electron microscopy (SEM) images are presented in Figure 1. They illustrate the initial agglomeration of the raw powder (Figure 1A) as well as the peculiar shape of an agglomerate formed by dry granulation, grinding and sieving (Figure 2A).

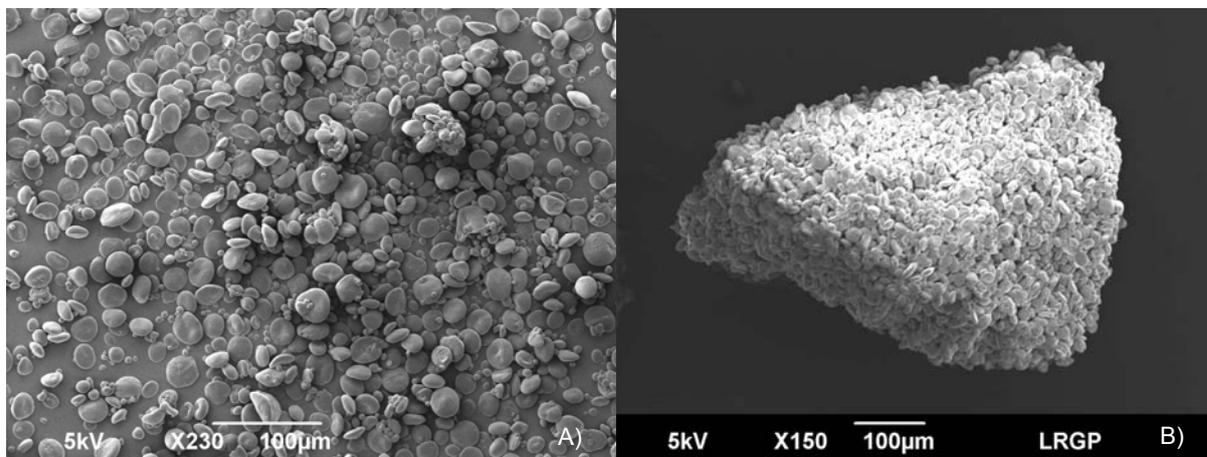


Figure 1: SEM images of A) the raw starch powder, B) a starch agglomerate generated by dry granulation

2.2 Dust dispersion, ignition and explosion

The particle size distributions shown in Table 1 do not represent the characteristics of a dust cloud dispersed in a standard explosion vessel. As tests were performed with a 20L sphere according to EN ISO/IEC 80079-20-2:2016/AC (2017), a similar vessel equipped with windows was designed in order to visualize the dust dispersion (Murillo, 2016). A laser diffraction sensor (Helos Vario - Sympatec) was used to measure the time evolution of the PSD on a 0.5 to 175 μm range at 2000 Hz. Figure 2A demonstrates that the particle size distribution of the raw powder is only slightly modified by the dust injection in the sphere through a rebound nozzle. The mean diameter only decreases from 24 to 18 μm due to the fragmentation of some of the agglomerates shown in Figure 1A. Moreover, the PSD is stable as a function of time, notably between 15 and 165 ms. Only a slight shift towards the biggest particles is visible after 120 ms, which is due to the sedimentation of these structures. Figure 2B shows that the particle size distribution of the agglomerates dispersed within the 20L sphere greatly varies with regard to that determined in ethanol or observed by SEM.

Agglomerates are broken when injected into the sphere and their mean size decreases from 280 to approximately 150 μm . Nevertheless, it should be stressed that the laser diffraction sensor cannot measure particles with diameter greater than 175 μm . In addition, SEM observations were performed by collecting the dust after dispersion into the explosion sphere. They confirm the fragmentation of the agglomerates into finer structures whose dimensions are between 100 and 200 μm . The presence of unagglomerated raw starch particles is also confirmed by Figure 2B and SEM observations. The time evolution of the PSD of the agglomerates shows that the presence of the larger structures is more perceptible 115 ms after the beginning of the dust dispersion. As the PSD is not stable as a function of the time, the choice of the ignition delay time (t_v) can have an impact on the dust explosivity.

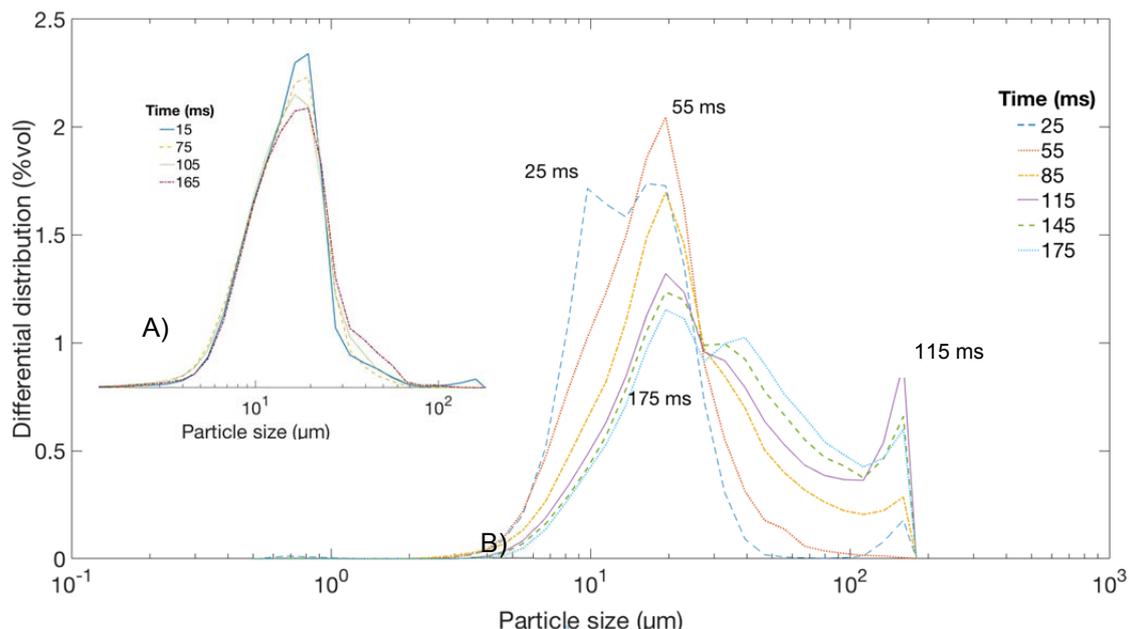


Figure 2: Time evolution of the particle size distribution of A) the raw starch powder, B) the starch agglomerates generated by dry granulation

The minimum ignition energy (MIE) of raw starch and agglomerates was also determined using a modified Hartmann tube for different size ranges of agglomerates prepared as previously described. It appears that agglomerates greater than 315 μm cannot be ignited by energies lower than 1 J. However, samples with a lower PSD, notably between 224 and 315 μm , were characterized by a MIE of 67 mJ. The same MIE was obtained for raw starch. Once again, it appears that fine particles were still present onto the agglomerates and that their presence, even at low concentration, has probably an impact on the ignition sensitivity. In this case, the most relevant parameter to consider could be the d_{10} diameter. The explosion severity was characterized for raw starch and for the agglomerates described in Table 1. A 20 L explosion sphere equipped with a standard rebound nozzle was used to determine the maximum overpressure P_m and the maximum rate of pressure rise $(dP/dt)_m$. Ignition was ensured at different ignition delay times t_v by 100 J chemical igniters. The pyrolysis gases were collected in a collapsible Tedlar bag and analyzed thanks to a micro gas chromatography (GC Varian, CP 4900).

3. Results and discussion

3.1 Mass concentration as a key parameter of dust explosivity

The representation of the severity of the explosions is very commonly done as a function of the mass concentration in dust. This concentration has the advantage of being easily understandable in an industrial environment and can be converted into preventive measures. Figure 3A shows the evolution of the maximum explosion pressure as a function of the dust concentration for a t_v parameter of 60 ms. P_m reaches a maximum at 750 $\text{g}\cdot\text{m}^{-3}$ for the raw starch, whereas this maximum is increased to 1000 $\text{g}\cdot\text{m}^{-3}$ for the agglomerates. Nevertheless, it should be noted that the two graphs are rather similar. The explosion pressure being related to the thermodynamic of the reactive system, it is therefore coherent that, for the same powder concentration, the evolution of P_m is nearly identical for both reactive systems. Figure 3B corresponds to the

variation of the maximum rate of pressure rise as a function of the concentration. An offset of the two curves is clearly visible: the rate of pressure rise increases less rapidly with the concentration for the agglomerated powder than for the raw starch; but the maximum $(dP/dt)_m$ is almost identical for both curves. It should also be underlined that the minimum explosive concentration is increased for the larger particles.

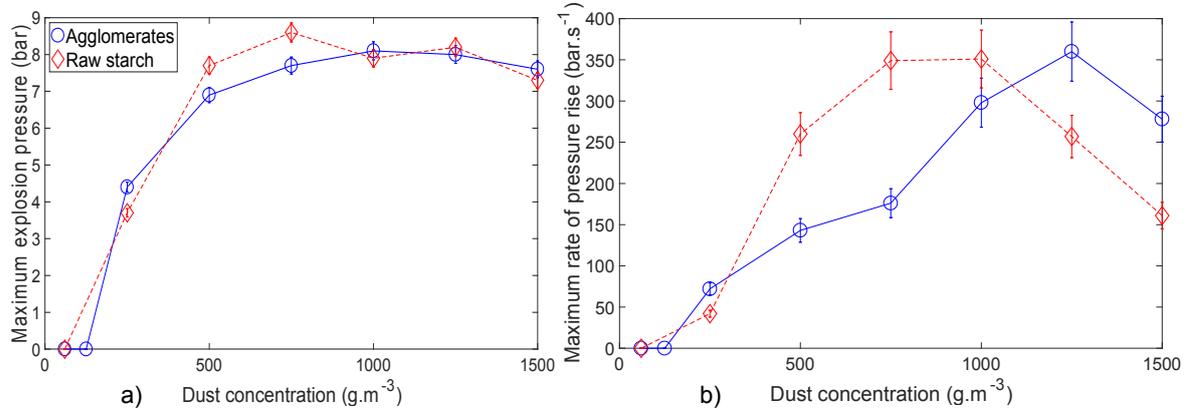


Figure 3: Influence of the dust concentration on A-left) the maximum explosion pressure and B-right) on the maximum rate of pressure rise for the raw starch and starch agglomerates

3.2 Which metrics to determine the rate-limiting step?

As previously stated, the mass concentration is useful for the transposition of experimental data on an industrial scale. However, this representation gives very little information about the mechanisms involved in the dust explosion. Various limitations are encountered during the explosion of an organic powder. The devolatilization step of the particle can be controlled by internal or external heat transfer. In the case of starch particles or agglomerates, as in the case of many organic particles smaller than approximately 300 μm , both Biot and Damköhler numbers are lower than unity, which excludes the possibility of a heat transfer limitation (Di Benedetto et al., 2010). The explosion can also be controlled by the reactions occurring in heterogeneous phase or between the pyrolysis gases and oxygen. By considering the formalism of the shrinking core model, the rate-controlling step will depend on one of the following three phenomena: diffusion through the gas film, diffusion through the ashes/porous shell, surface reaction. By considering the specific surface area of the powder, and more exactly, the total reactive surface of the fuel exposed to the oxidizing agent, a first selection of the rate-limiting steps can be made.

3.3 Correlation between specific surface area and dust explosivity

The agglomeration level of a powder has a significant impact on its specific surface and on its reactivity. To highlight this point, the results presented in Figure 3B are plotted as a function of the specific surface area calculated from the PSD obtained 60 ms after the beginning of the dust dispersion (Figure 2).

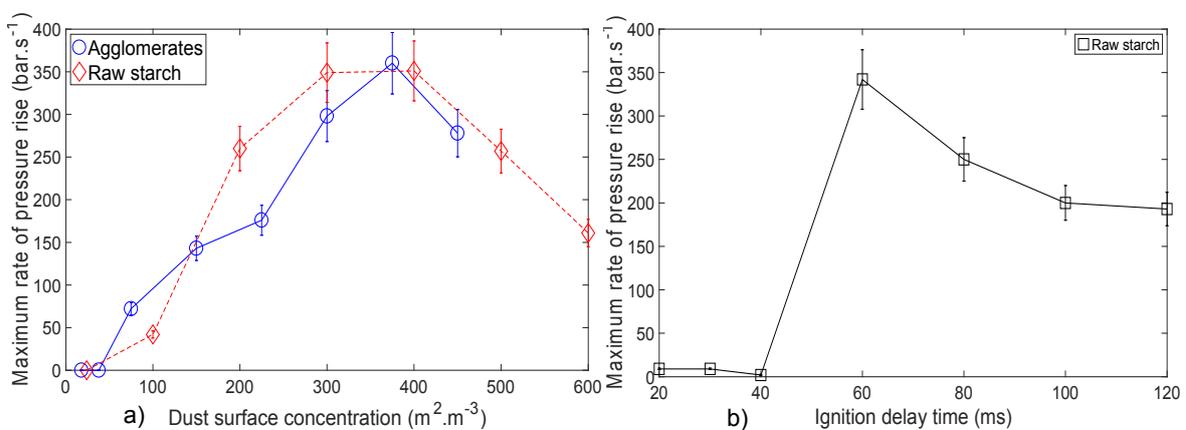


Figure 4: Evolution of the maximum rate of pressure rise of starch as a function of A-left) the fuel surface concentration and B-right) the ignition delay time (at 750 g.m⁻³)

It appears that the offset between the raw starch and its agglomerates disappears or, at least, is less perceptible (Figure 4A). In the case of a kinetic control of the gas reaction, the surface will not have an influence on the dust explosivity as demonstrated here. Moreover, both when diffusion through the gas film or surface reaction are rate-limiting steps, the consumption rate is proportional to the particle surface; which is not the case when the limitation is due to the diffusion through the ashes or porous shell. Figure 4A shows that the latter limitation should not be considered for the tested starch particles, which was expected as the high turbulence level during the explosion propagation certainly tends to tear off the char/ashes potentially created from the burning particles.

3.4 The numerous effects of turbulence

By modifying the gas flow velocity, the mass transfer coefficient will be affected but not the surface reaction rate. A change in the ignition delay time can thus help to identify if the rate limiting step is related to the diffusion through the gas film. Explosion tests were performed on raw starch at 750 g.m^{-3} for various ignition delay times, i.e. various turbulence levels of the dust cloud (Figure 4B). The flame kernel, generated by the explosion of the 100 J chemical igniter, is quenched when the turbulence is great, i.e. at 20, 30 and 40 ms. For ignition delay times equal or greater than 60 ms, the maximum rate of pressure rise decreases when the turbulence decreases, which is due to lower flame stretching but can also be related to the evolution of the mass transfer coefficient at the vicinity of the particles. Figure 2 also underlines the possibility of a change in the PSD as a function of the ignition delay time, notably when agglomerates fragmentation occurs. With regard to the magnitude of the decrease observed in Figure 4B, the influence of the turbulence on the mass transfer coefficient and on the dust explosivity can be nor neglected, nor confirmed with a great level of confidence.

3.5 Which indicator for the combustion kinetics?

For each explosion tests, chromatographic analyzes were performed on the burnt gases in order to identify the main permanent gases generated such as carbon dioxide, carbon monoxide, oxygen, nitrogen, hydrogen, methane, ethylene and some BTX vapors. Water vapor is also created but is not quantified. Figure 5 shows the evolution of the CO/CO₂ ratio, hydrogen and oxygen contents. The oxygen content decreases rapidly and reaches a minimum at 750 g.m^{-3} , which is the concentration at which the maximum $(dP/dt)_m$ is reached. Concurrently, the CO/CO₂ ratio increases significantly when the fuel equivalent ratio increases, then it drops for dust concentration is greater than 1000 g.m^{-3} . When the oxygen content is low, the oxidation of starch lead to carbon monoxide formation. This trend is amplified by Boudouard reaction which promotes carbon monoxide at the expense of carbon dioxide when the temperature grows. When the concentration at which the maximum $(dP/dt)_m$ is exceeded, the flame temperature decreases, leading to a CO/CO₂ ratio diminution. Hydrogen content follows a very similar trend.

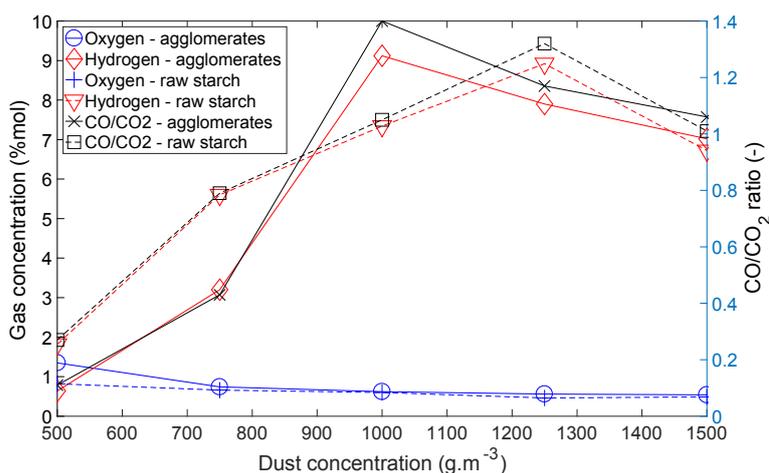


Figure 5: Influence of the dust mass concentration on the composition of the burnt gases for raw starch and starch agglomerates

Comparable trends are observed for starch agglomerates. However, an offset between raw starch and its agglomerates is clearly visible: for instance, the maximum CO/CO₂ ratio is reached more rapidly, i.e. for a lower dust concentration, when agglomerates explode. It can also be observed that the oxygen content

decreases less rapidly for the agglomerates, meaning that oxygen conversion is more difficult for the agglomerates than for raw starch. The same set of data has been represented in Figure 6, but as a function of the dust surface concentration, i.e. the fuel surface area available per cubic meter.

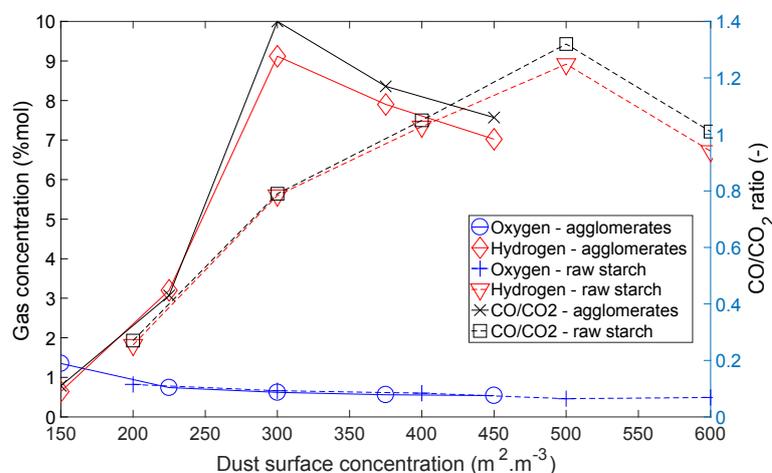


Figure 6: Influence of the dust surface concentration on the composition of the burnt gases for raw starch and starch agglomerates

The oxygen consumption is well correlated to the dust surface area (Figure 6). However, for a same dust surface available in the sphere, different burnt gases compositions are recorded, which means that the chemical reaction is not directly correlated to the fuel surface. By combining the previous results, the influence of oxygen accessibility on the surface of the particles appears clearly. Then, the assumption of the diffusion through gas film being the rate controlling step seems very likely for such powders.

4. Conclusions

If the mass median diameter can be interesting for kinetics controlled by homogeneous chemical reaction and directly related to the fuel equivalent ratio, the reactive surface is more relevant when the combustion is controlled by surface reaction or diffusion to the surface, as it is the case for the tested starch samples. But a dust explosion is a complex phenomenon which not only depends on the combustion kinetics, but also on the radiative transfer and on turbulence/combustion interactions. Therefore, the use of a single characteristic diameter is not sufficient and a multi-metric approach based on the whole PSD, characterized by its skewness (Tascón, 2018), size dispersity (Li et al., 2016) or specific surface area, is recommended.

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References

- Callé S., Klabá L., Thomas D., Perrin L., Dufaud O., 2005, Influence of the size distribution and concentration on wood dust explosion: Experiments and reaction modelling, *Powder Technol.*, 157, 144-148.
- Di Benedetto A., Russo P., Amyotte P., Marchand N., 2010, Modelling the effect of particle size on dust explosions, *Chemical Engineering Science*, 65, 772-779.
- EN ISO/IEC 80079-20-2:2016/AC, 2017, Explosive atmospheres - Part 20-2: Material characteristics - Combustible dusts test methods - Technical Corrigendum 1, European Committee for Standardization.
- Li Q., Wang K., Zheng Y., Ruan M., Mei X., Lin B., 2016, Experimental research of particle size and size dispersity on the explosibility characteristics of coal dust, *Powder Technol.*, 292, 290-297
- Murillo C., 2016, Experimental and numerical approaches to particles dispersion in a turbulent flow: application to dust explosions, PhD Thesis, University of Lorraine, Nancy, France.
- Rumpf H., 1962, The strength of granules and agglomerates, *Agglomeration*, 379-413.
- Tascón A., 2018, Influence of particle size distribution skewness on dust explosibility, *Powder Technology*, 338, 438-445.
- Yuzuriha Y., Gao W., Mogi T., Dobashi R., 2017, Effects of particle size distributions on flame propagation behavior through dust clouds of PMMA, *J. Loss Prev. Process Ind.*, 49, 852-858.