# Flammability data measurement of aluminium powder and oxide content influence

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## 1 Introduction

Metal dust explosion hazard is of great interest for safety management. Regulations like European ATEX directives imply strong security levels which are still a problem for industrials. In spite of sharp statistics, it seems that inorganic dust explosion number increase. Aluminium powder is of interest because of its great use and the important waste quantities generated [1].
Applying security regulations is expensive for a company, so a good evaluation of hazards is important. Concerning aluminium, a lot of studies show that the ignition of a single particle will occur after the crack of the oxide layer [2]. In this way a decrease of sensitivity of an Al. powder with the oxide content could be expected. An investigation of this assumption is presented here through the evaluation of ignition sensitivity to electric spark and flame velocity. In order to achieve this study, an original process was developed to prepare several samples with a variation of the alumina amount.

## 2 Experimental set up and test procedure

### *Experimental set up*

The dust flammability experiments were conducted in an explosion tube based on the principle of Hartmann apparatus. A specific spark generator was developed in order to improve the estimation of the ignition energies of dust clouds. This system has the distinctive feature of producing a spark at nearly constant power. An auxiliary spark discharge at high voltage is used to initiate the main spark at low voltage. Its energy was estimated to be about 18 mJ. Concerning the main spark, the current is adjustable in the range [2–8] A and residual voltage varied between 50 and 70 V, depending on the properties of the dust cloud itself. In this way, the energy is controlled by the duration of the spark. The energy of the spark is obtained by the relation:

$$\mathrm{E} = P \times t_p + 18\mathrm{mJ}$$ where $P$ and $t_p$ are respectively the power and the duration of the main spark

Some parameters were adjusted on the experimental set up to have a good dispersion of dust. The overpressure ($\Delta$P) in the air tank is discharged in the tube during $\Delta t_0$, then the valve is closed and after $\Delta t_1$ the mixture is ignited. During the series of experiments, the parameters $\Delta$P, $\Delta t_0$ and $\Delta t_1$ were respectively set at: 0.5 bar, 100 ms and 100 ms. The opening between the cup where the dust is deposited and the deflector was also optimized and set at 2.5 mm. Pointed tungsten electrodes were used for all this study (diameter: 2.4 mm and spark gap: 4 mm). An extended description of this system is available in [3].

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The flame propagation was filmed with an high speed video camera (Photron Fascam PCI) at a record rate of 1000 fps. In the same time a piezo electric pressure sensor (Kistler 6001) was used to evaluate the pressure rise in the explosion tube.

## *Samples and test procedure*

The aim of this work is to evaluate the influence of oxide content of an aluminium powder on flammability data. A commercial aluminium powder (purity > 99.7%) with closely spherical particles was used as a raw material. A SEM picture of particles is presented on Fig. 1a. The size distribution (Fig.1b) of this powder was determined with a laser diffraction technique (Spraytech, Malvern). It should be noted that this measure is based on the assumption of spherical particles. The real shape of our sample might lead to a slightly overestimated result.

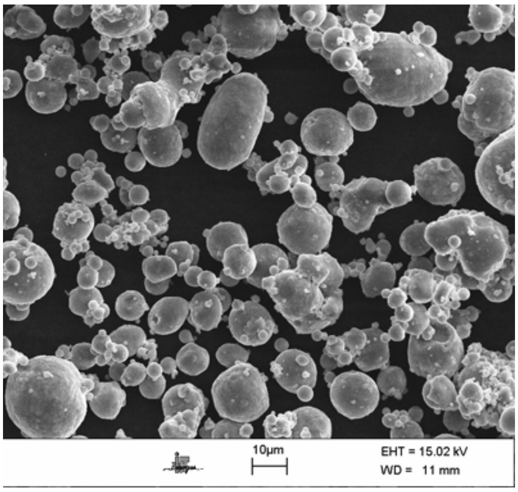


Fig.1a SEM picture of the raw material.

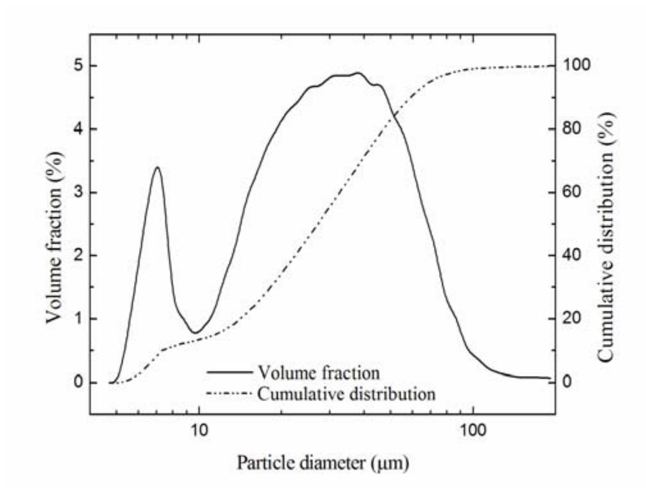


Fig.1b Size distribution of the raw material.

This powder was first separated by sieving into three samples of various size distributions: d < 25 µm, 25 < d < 30 µm and 30 < d < 50 µm. By mean of thermogravimetry, the oxide content of the raw material was evaluated at 0.48 %wt. The oxide content of the initial material was then increased by applying an anodisation process. This method enables preparing three samples with oxide content of 1.44, 2.3, 6.3 and 9.8 %wt. The oxidized powders were sieved under 50 µm.

The Langlie method [4] was used to determine ignition energies. This statistic method is based on the principle of dichotomy. The Langlie test consists of about 20 consecutive ignition tests with a variation of the applied energy. As said before, the spark energy is controlled by the duration the spark. So this procedure enables to evaluate $t_{p50}$ which is the duration of the spark leading to a probability of ignition of 50%. Considering the fact that the power of the spark depends on the dust cloud properties, a control of this value was made thanks to the spark current and voltage measurements. The energy $E_{50}$ is obtained by relation:

$$\mathrm{E}_{50} = \left(t_{p50} \times \int \mathrm{UI}dt\right) + 18\mathrm{mJ}$$

The explosion of the air/dust mixture in our experimental apparatus could be broken down into three stages. First, a spherical flame is observed, until the front flame reach the tube. Then, the flame is propagated upward in the tube. At last, the breaking of the filter due to the overpressure leads to a brusque acceleration of the flame. This study will only be interested in the first stage of the phenomenon in order to avoid wall effects. A program written in Matlab language was used for image processing of flame propagation records. Each frame is coding as a data matrix whose elements represent an intensity level (in the range [0; 255]). In spite of the use of filters during acquisitions the aluminium flame is very luminous. In this way, the part of the picture corresponding to the flame area was selected by keeping matrix elements over 200. The flame velocity is deduced from the flame radius evolution in time. Several methods were investigated for estimation of this radius. One method consists in the determination of the distance between the centre of the flame area and the flame front into a vertical or horizontal direction. The centre of the flame area may be evaluated at each time step or considered fixed and calculated on the first picture. The first assumption is interesting as it take into account the flame displacement due to the confinement in the bottom of the tube. An other method is to calculate the radius of disk with the same area than the flame. The choice of the method will be discussed latter. For all flame velocity measurements, ignition spark duration was set to 5 ms (E ~ 1.2 J). In order to evaluate the flame velocity of a self sustained combustion, image processing will be started at the end of the spark. This moment will be considerate as the initial time t = 0 ms.

# 3 Results

## *Ignition energies*

Fig.2 shows the influence of equivalent ratio and size distribution on ignition energies $E_{50}$.

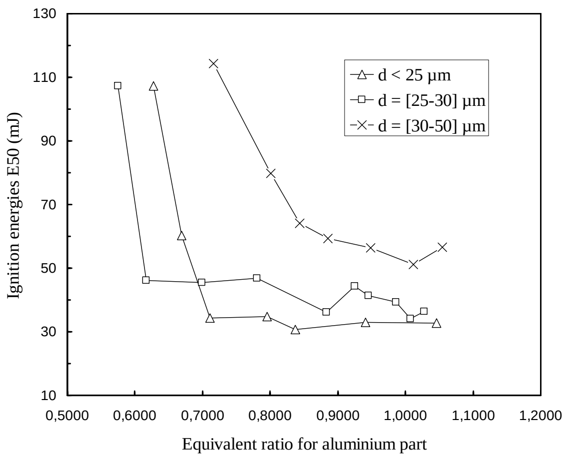


Fig.2 Influence of size distribution and dust concentration on ignition energies.

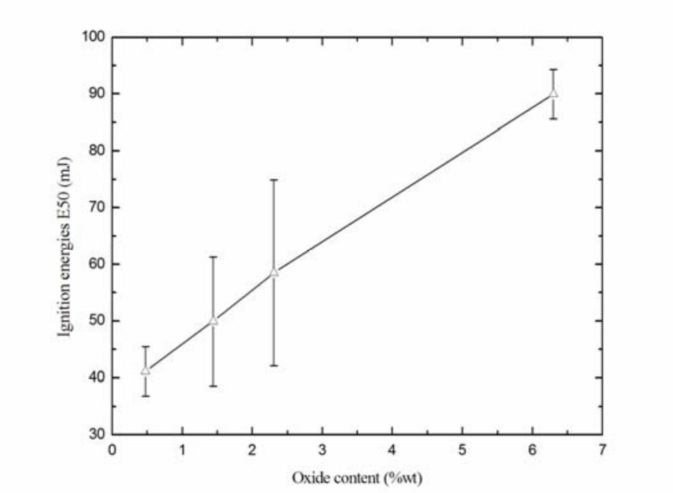


Fig.3 Influence of oxide content on ignition energies (equivalent ratio ~1).

The ignition energies clearly increase for small concentrations. This limit depends on size distribution: 0.7 for d<25 µm, 0.6 for 25<d<30 and 0.85 for 30<d<50. A decrease of the lean dust concentration limit with particles size was expected. So, this typical evolution of ignition energies is not obtained here. An explanation may be the fact that the fluidization parameters were the same for all results presented on Fig.2. Improvements to this point should be carried out for the sample with smaller particles (d<25 µm).

An equivalent ratio giving a minimum value of $E_{50}$ can't be observed. It appears that minimum ignition energies are obtained in the range [0.8; 1.05]. This phenomenon is attributed to residual turbulence in the dust cloud at the time of ignition. The ignition energies of samples decrease rapidly when reducing particles size. Experiments for rich mixture don't give good results. It seems to be very difficult to adjust fluidization parameter ($\Delta P$, $\Delta t_0$…) to generate an homogeneous dust cloud for high mass of powder. When increasing $\Delta P$, the resulting turbulence intensity increase too so that the ignition become impossible.

The evolution of the ignition energies with the oxide content of aluminium powders (equivalent ratio ~1) is presented on Fig.3. For oxide contents from 0.48 (raw material) to 6.3, energies $E_{50}$ increase in a quite linear way. From those results, an empirical relation could be expressed.

$E_{50}$ (mJ) = 8.13 x + 41 where: x = oxide content of the aluminium powder (%wt)

For the two points with 1.44 and 2.3%wt of $Al_2O_3$, larger standard deviations were obtained. Physico chemical analyses pointed out that these samples were less homogeneous in term of oxide content. A better control in preparation of the powder with 6.3%wt $Al_2O_3$ leads to standard deviation in the same order as for raw material.

Ignition of the sample with oxide content of 9.8%wt was not possible in spite of high spark energy values (up to 7 J). For some tests, the beginning of a combustion was obtained but without further self propagation.

## *Flame velocity*

An example of flame radius evolution resulting from different estimation methods is given on Fig.4.

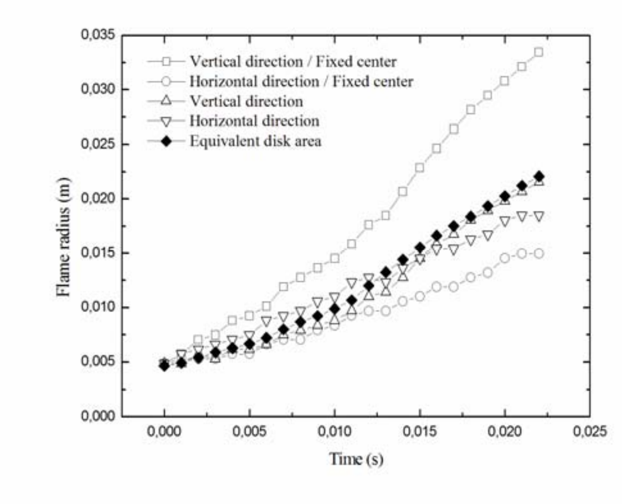


Fig.4 Comparison of methods for evaluation of radius time evolution
(Unsieved powder, equivalent ratio ~1)

The shape of the flame is generally close to a sphere. A vertical movement of the flame is sometimes observed. This is attributed to the fact that the tube is confined at the bottom and partially confined by the filter at the top. That leads to an overestimation of the flame radius when using the method with fixed centre in a vertical direction. As can be seen on Fig.4, the method considering the flame area seems to give an average evolution of the radius in time and will be kept to evaluate the flame velocity. Two additional reasons confirm the choice of this method. On the one hand, the flame front is not really smooth, so this method results in a more regular evolution of the radius than other ones. On the other hand it is interesting, for our future studies, to assume a spherical flame evolution.

A model for pressure evolution in our explosion tube is being tested and validated by comparison with overpressure measurements. This mathematical solution is based on the assumption of smooth spherical flame propagation.

Fig.5a and 5b show flame speed evolution of selected samples. It is to notice that the samples presented on Fig.5a are produced by the same process. In this way, the oxide content of these powders is expected to be identical.

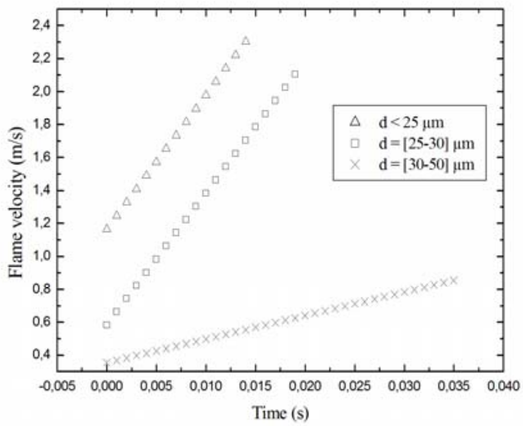


Fig.5a Influence of size distribution on flame velocity (t=0 at the end of ignition spark)

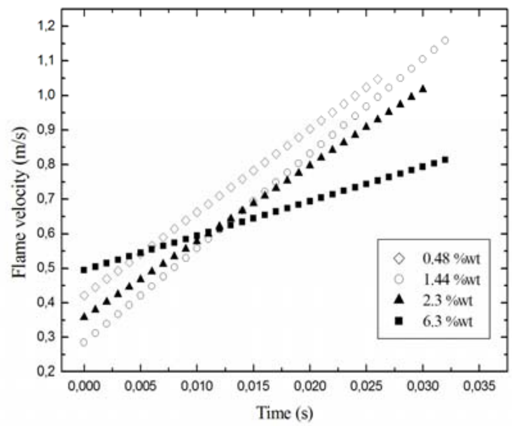


Fig.5b Influence of oxide content on flame velocity

The velocity during the first stage of explosion increases in a linear way. This evolution of flame velocity was soon observed for a flame propagation in an open field and is attributed to combustion process of aluminium particle [5]. It appears that the velocity is more sensitive to size distribution than oxide content. When decreasing particles diameters the flame acceleration increases rapidly. At the end of the first stage, the flame for particles size lower than 25 µm is three times higher than for the sample with sizes 30-50 µm. The influence of alumina part on velocity is different from one on ignition energies. This influence is limited for oxide content lower than 2.3%wt but more important for 6.3% of $Al_2O_3$ in weight.

## 4 Conclusion

Flammability data measurements of aluminium powder with various oxide contents are presented. A new methodology was developed to evaluate the ignition energies $E_{50}$ of dust clouds. On the one hand, a specific spark generating system enables a good control of the applied energy, by controlling the duration of the spark produced at nearly constant power. The Langlie method then was fitted to the assessment of ignitability of dust clouds. It was so possible to link rapidly an energy with a probability of 50% of ignition. On the other hand, an anodisation process was used to increase, under control, the oxide coating of particles. This original method gives us the possibility to evaluate the influence of the oxide thickness on explosion properties of an aluminium dust cloud.

The ignition energies clearly increase with the alumina part of the sample. For an oxide content of 9.8%wt, the $Al_2O_3$ thickness seems too high to obtain an explosion. Further tests with oxide content around 7.5 and 8.5%wt would be necessary to evaluate the critical alumina part.

A method leading to evaluation of flame velocity evolution was presented. Oxide content seems to have a lower influence on the velocity than size distribution of particles. The flame speed will be used to estimate, with a mathematical solution, the pressure time evolution.

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