

Gaseous flame suppression mechanisms

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Abstract

Suppression is used in industrial situations where other mitigations means are inadequate. A number of suppression techniques exist but, today, most of them use an extinguishing powder like sodium bicarbonate. In this paper, a physical analysis of the action of this particular suppressant is proposed and compared to that of radically different inert powders like magnesium oxide. The incidence of the particle size is discussed. The results of the analysis are compared to specific experimental data produced on purpose using a 1 m³ vessel and methane-air mixtures.

Keywords: *gas explosions, flame propagation, extinction, suppression*

1. Introduction

Suppression is used in industrial situations where other mitigations means are inadequate. The technique is not recent and may even be one of the oldest. Is it remembered that “neutralisation” of coal dust and “passive barriers” using either limestone dust or water have been used since the beginning of the twentieth century in coal mines throughout Europe (Cybulski, 1973). Note that the question of the particle size and chemical nature of the extinguishing powder was discussed since the beginning. This mitigation method is also available for the other sectors of the industry although using radically different techniques to inject and mix the extinguishing powder with the explosible mixture. In vessels, the blast of the explosion is not large enough to disperse the dust and the powder is injected under pressure from a pressurised reservoir. To do this, a pressure sensor can be used to detect the explosion, triggering a control unit which releases a valve or breaks a diaphragm (Hattwig and Steen, 2008). In Europe, according to the ATEX directives, any “suppression technique” should be certified. The relevant standard is EN14373. In this particular standard, the detailed physics of the extinction mechanism is not clarified so that the influencing parameters are not categorised by order of importance. If the flame propagating speed (hidden in the Kst coefficient) is certainly of importance, the minimum quantity of extinguishing powder to inject in order to quench the flame is at least as important. But, this important aspect is not explicitly addressed. And the quenching efficiency may depend on radically different explosion parameters (minimum ignition parameters for instance) but also on the characteristics of the extinguishing powder itself (particle size, chemical nature).

It is the aim of the present paper to investigate further the quenching aspects. The first part deals with the available theories describing the interactions between an inert powder and a gaseous flame for which some key parameters can be extracted. The second part is devoted to an experimental verification and the last one pertains to the implications about suppression techniques.

2. Physical analysis

The theoretical approach of “matched asymptotics” (Cole, 1968) provided a number of qualitative and quantitative insights into the flame propagation mechanisms. In particular, the influence of inert particles (Joulin, 1980) even vaporizing (Mitani, 1981) was addressed a few decades ago. It was extended 10 years later (Ju and Law, 2000).

It is worthwhile recalling the saillant features. These authors propose a description of a laminar flame submitted to a volumetric heat loss $L(T)$ where T is the local temperature. Using a one step global reaction kinetics assuming large activation energy, they demonstrated that flame quenching occurs as soon as the power density extracted in the flame front reaches about 18% of the volumetric power released throughout the flame front (figure 1). The adimensional volumetric heat release parameter is given by :

$$q \approx \frac{\lambda.R.E}{(\rho_0.Cp.S_{lad}.R.T_{ad})^2} * \left\{ \int_{T_u}^{T_{ad}} L(T).dT + L(T_{ad}) \right\} \quad (1)$$

Where :

- S_{lad} is the adiabatic laminar burning velocity
- ρ_0 the specific mass of the gaseous reactants at the ambient temperature T_0
- Cp the specific heat of the gaseous mixture
- R the perfect gas constant
- E the activation energy of the reaction
- T_{ad} the adiabatic combustion temperature
- λ the thermal conductivity of the mixture

At the quenching point, the burning velocity of the mixture (S_1) has dropped by 40% (figure 1). It was further established that the quenching criterion is reached when the heat losses were so intense that the temperature in the reaction zone has dropped by the following quantity :

$$\Delta T_{crit} = R.T_{ad}^2 / E \quad (2)$$

Some kinetic parameters were tabulated some time ago (Westbrook and Dryer, 1981). Typical values for the Zeldovitch number (here E/RT_{ad}) are on the order 7 for a number of hydrocarbons burning in air so that the temperature drop in the reaction zone remains very limited (less than 15%).

When the gaseous flame is seeded with monodisperse inert particles $L(T)$ reads :

$$L(T) = N_p.Nu.\lambda.\pi.d.(T - T_p) \quad (3)$$

Where :

- d stands for the particle diameter, Nu the Nusselt number ($Nu = 2$ for small particles),
- T_p the temperature of the particles (different from that of the surrounding gas T),
- N_p particle number per unit volume.

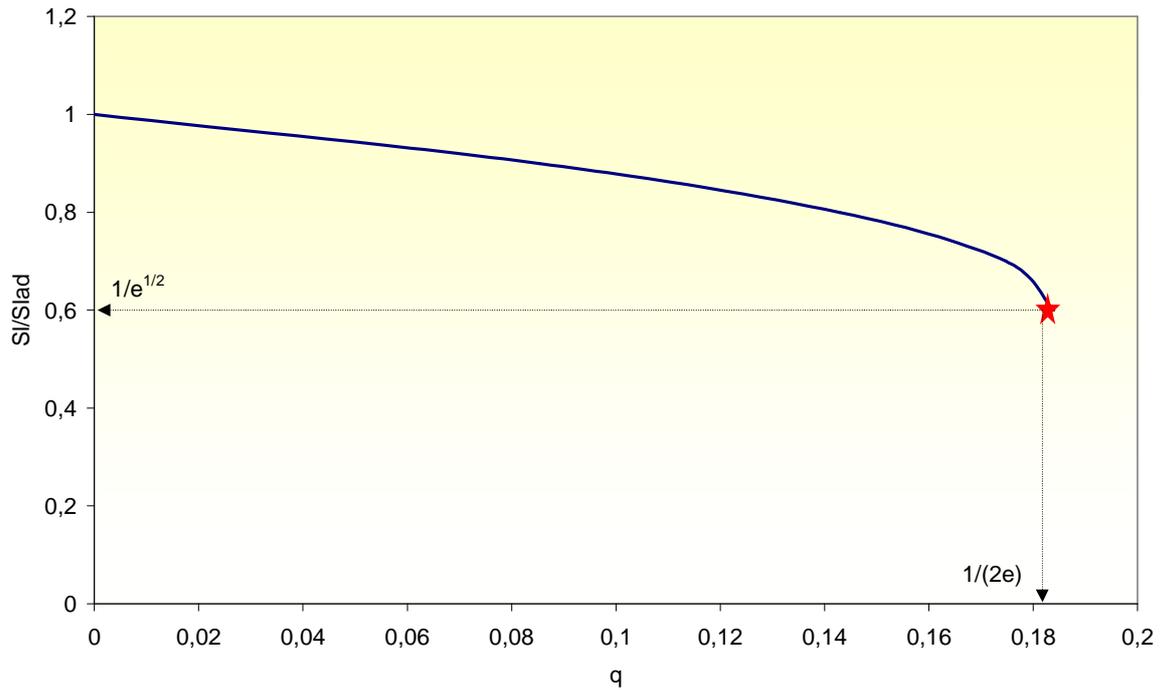


Figure 1: evolution of the laminar burning velocity of a non adiabatic gaseous flame as function of the reduced volumetric heat loss.

The resolution is proposed using the following additional parameters. K is the ratio between the transit time (of a particle) through the flame front and the characteristic heating time of a particle, whereas h is the ratio between the energy extracted by the particles and the heat release by the combustion. Note h is directly proportional to the numerical concentration of particles.

$$K = \frac{6 \cdot \lambda^2 \cdot Nu}{\rho_0 \cdot C_p \cdot S_{lad}^2 \cdot \rho_s \cdot C_s \cdot d^2} \quad h = \frac{\lambda^2 \cdot Nu}{(\rho_0 \cdot C_p)^2 \cdot S_{lad}^2} \cdot \frac{N_p \cdot \pi \cdot d \cdot E}{R \cdot T_{ad}} \quad (4)$$

Where :

- ρ_s is the specific mass of the condensed phase
- C_s is the specific heat of the condensed phase

A graphical representation is proposed on figure 2. It is shown that only when the particles are sufficiently large ($K < K_c = 0.0767$) quenching can be obtained. With smaller particles, the temperature lag remains very small and $L(T)$ is never sufficiently large to quench the flame. As the mass particle concentration increases, the laminar burning velocity decreases steadily. The particles act as a diluent.

The solutions proposed by Joulin were used to investigate the quenching of stoichiometric methane-air flames (figure 3 with the data from table 1). Only with particles larger than 40 μm quenching can be observed with a mass particle concentrations increasing (quadratically) with the particle size.

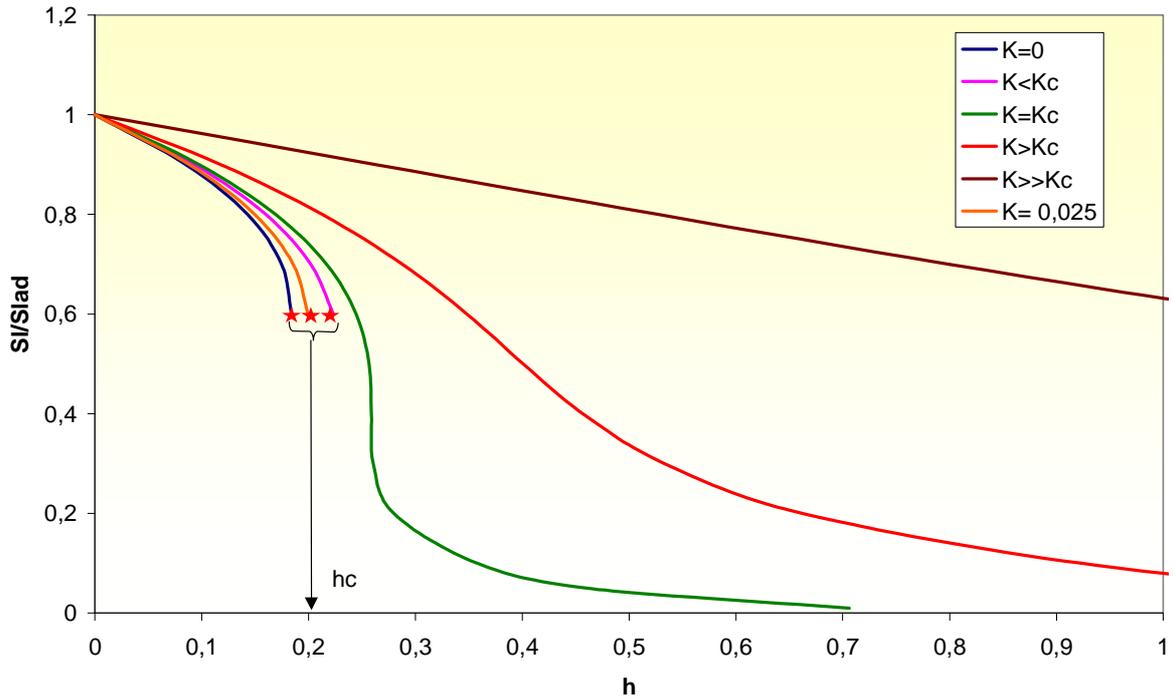


Figure 2 : Evolution of the laminar burning velocity of premixed gaseous flames seeded with inert particles

Table 1 : data used for the implementation of the model proposed by Joulin

Flame (CH ₄ -air stoich.)	Particles
E = 100000 J/mol and A = 2.5 · 10 ⁶	ρ _{part} = 2000 kg/m ³
T ₀ = 293 K	C _{part} = 1000 J/kg/K
C _p = 1140 J/kg/K (mean value)	Nu = 2
λ = 0,074 W/m/K (mean value)	

This theory can also help to investigate the incidence of the particle size distribution. Note that the value of h for which quenching happens (the stars on figure 2) varies only very little with the particle size (or K). If N_{pd} is the number of particles of diameter d (per unit volume in the cloud), then quenching would be obtained in the following situation¹ :

$$hc \cdot \frac{(\rho_0 \cdot C_p)^2 \cdot S_{lad}^2}{\lambda^2 \cdot Nu \cdot \pi} \cdot \frac{R \cdot T_{ad}}{E} = \sum_0^\infty N_{pd} \cdot d = N_p \cdot d_{10} \quad (5)$$

Recalling that the mass particle concentration reads:

$$\sigma = \rho_s \cdot \frac{\pi}{6} \cdot \sum_0^\infty N_{pd} \cdot d^3 = N_p \cdot d_{30}^3 \quad (6)$$

¹ d₁₀ is the arithmetic mean diameter and d₃₀ the volum mean diameter. d₃₁ is the volum/length mean diameter or mean evaporative diameter and Sauter diameter d₃₂ is the volum/surface mean diameter

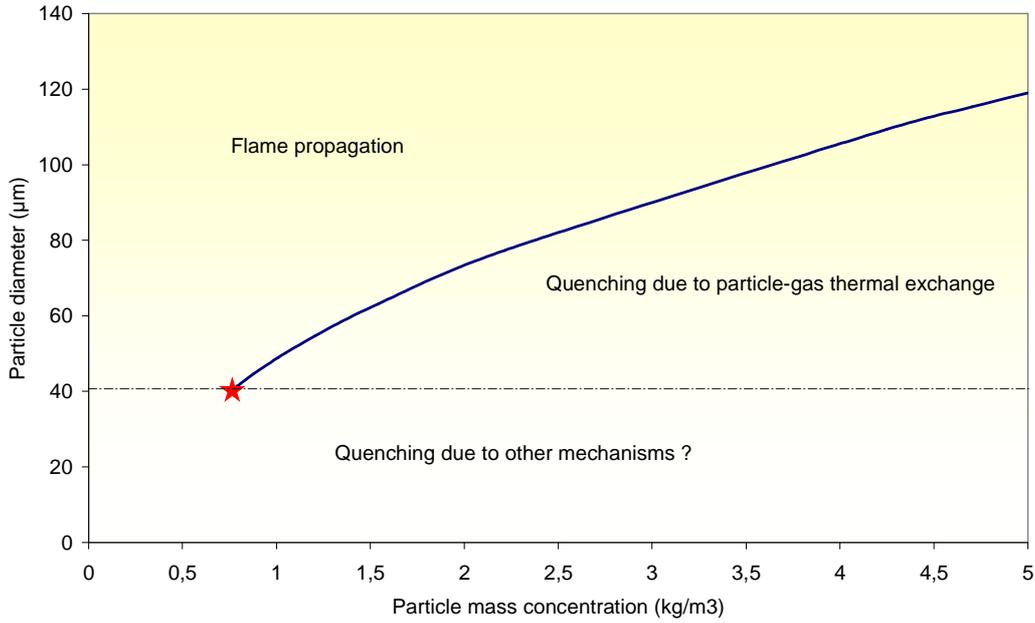


Figure 3 : quenching of stoichiometric methane-air flames by monodispersed inert particles

The relationship between the quenching condition and the particle size distribution is given by the following expression, suggesting the characteristic particle diameter is neither the Sauter mean diameter (d_{32}) nor the VMD (volum median diameter). Taking this into account the curve of figure 3 applies provided d is replaced by d_{31} AND the particles are all larger than 40 μm .

$$hc \cdot \frac{(\rho_0 \cdot C_p)^2 \cdot S_{lad}^2 \cdot RT_{ad}}{\lambda^2 \cdot Nu \cdot \pi} \cdot \frac{RT_{ad}}{E} = \frac{\sigma}{\rho_s} \cdot \frac{6}{\pi} \cdot \frac{d_{10}}{d_{30}^3} = \frac{\sigma}{\rho_s} \cdot \frac{6}{\pi} \cdot \frac{1}{d_{31}^2} \quad (7)$$

It is difficult to admit that if the particles are very small (smaller than 40 μm is the present application), quenching will never be obtained whatever the particle mass concentration. It is well known that it is possible to quench a flame by adding increasing quantities of nitrogen for instance. In this particular situation, heat radiation from the hot gases is believed to be responsible for the extinction of the flame. Ju (Ju and Law; 2000) integrated this aspect in his extension of the analysis of Joulin and demonstrated the existence of two separate limits : one corresponding to that identified by Joulin and a second one corresponding to the radiation losses of the burnt products. When the particles are large the former is largely dominating and when they are small, the second mechanism is leading. Performing a quantitative estimation with this theory is not totally straightforward but the modifications of figure 2 can be evaluated. The particle diameter separating both regimes is the same as above : 40 μm . For the specific case described in table 2 and figure 1, the incidence of the radiation losses would shift the curve towards the vertical axis (so that the particle mass concentration would decrease by about 10%). For particles smaller than 40 μm , the particle mass concentration producing the

extinction of the flame should remain approximately constant whatever the particle size. It follows that for particles smaller than 40 μm , the characteristic particle size would be that related to the mass concentration : d_{30} (and not d_{31} as for the particles larger than 40 μm).

Mitani tried to add the incidence of particle endothermic gaseification (vaporization, pyrolysis) but without considering radiation losses. One of the simplest way for estimating the incidence of the particle gaseification is to consider that the endothermic phase change is (very) roughly corresponding to an increase of the heat capacity of the condensed phase so that :

$$\overline{C_s} = C_s + \frac{L_{vap}}{T_{ad} - T_0} \quad (8)$$

Where :

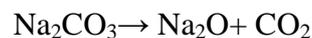
- $\overline{C_s}$ is the equivalent condensed phase heat capacity replacing C_s in the formulae
- L_{vap} the heat absorbed due to the phase change of one kg of condensed material

In the specific case of water droplet the increase of C_s would amount 25%. The expected incidence on the above quenching parameters seems rather small (10% decrease of the limiting particle diameter for instance). Nevertheless, the vapours (especially water vapors and carbon dioxide) may act as strong radiators promoting flame extinction. But this specific aspect does not seem to have been deserved a specific attention.

This question is particularly marked if sodium bicarbonate powder (NaHCO_3) is considered. This powder is largely employed in today suppression technologies, for a number of reasons, including its good quenching efficiency. Chelliah pointed out these specific points (Chelliah and al., 2003). Sodium bicarbonate decomposes around 150°C to release water vapour and carbon dioxide :



At a higher temperature, around 850°C, Na_2CO_3 vaporizes or more probably decomposes :



It may be postulated that in the presence of water vapour, sodium oxide would transform in sodium hydroxide. Sodium hydroxide could terminate the chain branching mechanism of the combustion. Chelliah claims that this specific chemical aspect could play a role especially with extremely small particles (below 20 μm) which can be heated above 1000°C inside the flame flow and decompose totally.

The following experiments were performed to try and investigate further these aspects.

3. Experiments

3.1 Setup

The ISO 1m³ chamber was used (Figure 4). Methane-air mixtures were prepared inside and controlled using a paramagnetic oxygen analyser (SERVOMEX - type PM1158 – error $\pm 0.02\% \text{O}_2$ v/v). Slightly above stoichiometric mixtures were prepared (10.5% v/v) to take into account the additional quantity of air introduced while injecting the extinguishing powder. The latter (see next section) is contained in the 5 l discharge bottle pressurized with air up to 20 bar gauge. The powder is dispersed using the standard semi circular perforated ring. Ignition is performed 700 ms² after the start of the discharge using a strong ignition source (see later). Pressure is measured in the discharge bottle, in the 1”1/2 diameter pipe connected to the perforated ring and inside the chamber using piezoresistive pressure transducer (KISTLER type 4045 A error ± 10 mbar). Typical pressure signals are shown on Figure 5 together with the classical explosion parameters such as the maximum explosion pressure (Pmax, absolute pressure) and the maximum rate of pressure rise ($\Delta P/\Delta t_{\text{max}}$). Note the dispersion bottle is not empty when the explosion occurs. To better approach the real powder concentration in the chamber at that time, it was assumed that the proportion of mass powder remaining in the bottle was the ratio between the minimum pressure in the bottle at this moment and the maximum discharge pressure (20 bar).

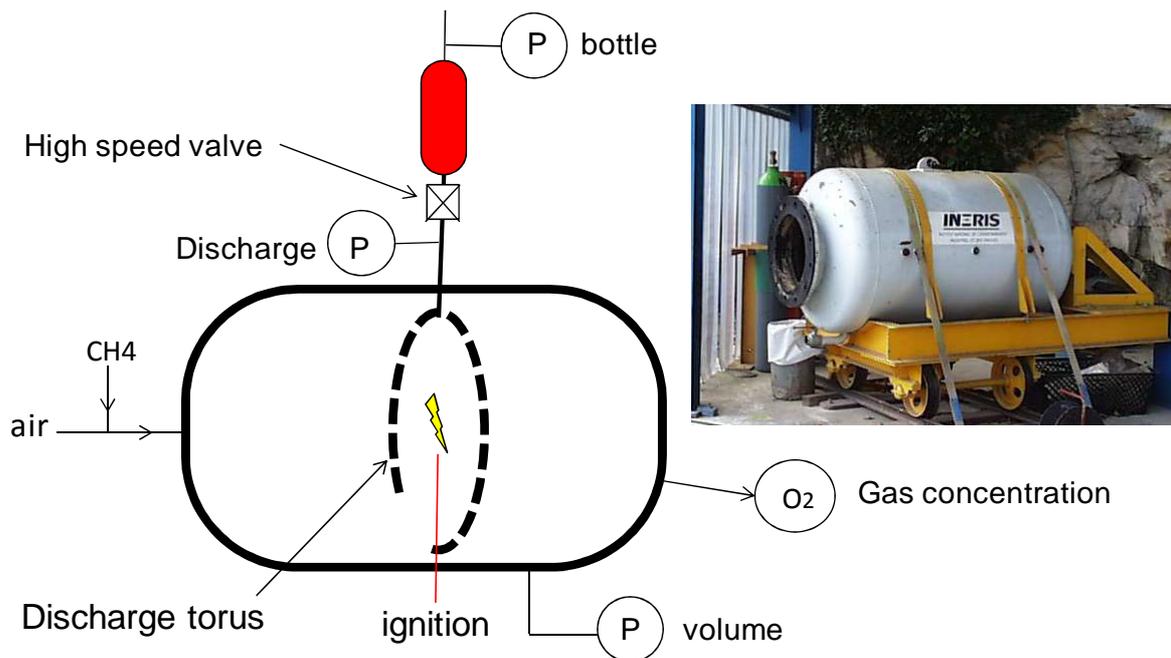


Figure 4 : 1m³ experimental setup. Circles indicate measurement points

² In the ISO procedure, the ignition delay is 600 ms. It was slightly increased to allow a more complete discharge of the powder especially when the amount of dust was larger than 1 kg.

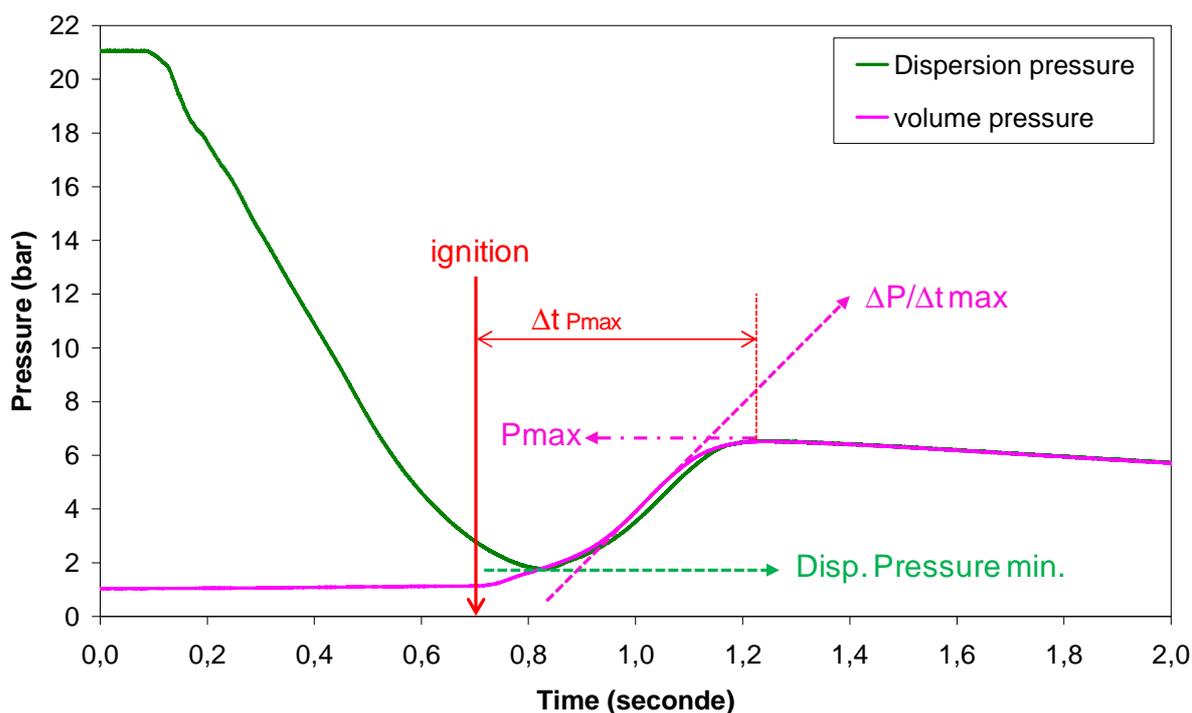


Figure 5 : Example of pressure signals.

A very significant problem is how to ignite increasingly weaker reactive mixtures which are produced when mixing increasing amounts of extinguishing dust in the methane-air gaseous mixture. A set of typical P-t curves is presented on figure 6 showing a significant increase of the combustion time when the quantity of extinguishing dust is increased. To do these experiments, the traditional 2 x 5 kJ igniters were used (figure 7 left). From past experience (Proust and al., 2007), it is suggested that the turbulence intensity required to mix the dust and maintain it in suspension is dropping below 1 m/s about 1s after the start of the discharge. There is then a significant risk of particle settling if the combustion lasts more than 1 second. Since the igniters tend to have a rather long afterburning, ignition may then occur as soon as the local dust concentration would have dropped enough. Rather expectedly, with this same ignition method, the combustion time (figure 6) increases with the amount of dispersed extinguishing dust but reaches a threshold above 800 g of powder and it was never possible to quench the flame whatever the quantity of dust. The inflexion point is located at about 1 second confirming the present analysis.

Another type of igniter was designed to trigger sooner the flame propagation and even to “force” locally the flame propagation. 10 g of “flash powder” ($\text{KNO}_3 + \text{Mg}$) was deposited on a small metal cup located in the middle of the chamber (figure 7 center). The flash powder was ignited using a small pyrotechnical match (60 J- figure 7 right). Care was taken to avoid any afterburning. The total amount of energy released is about 200 kJ within a timescale of 100 ms. Typical P-t curves are shown later.

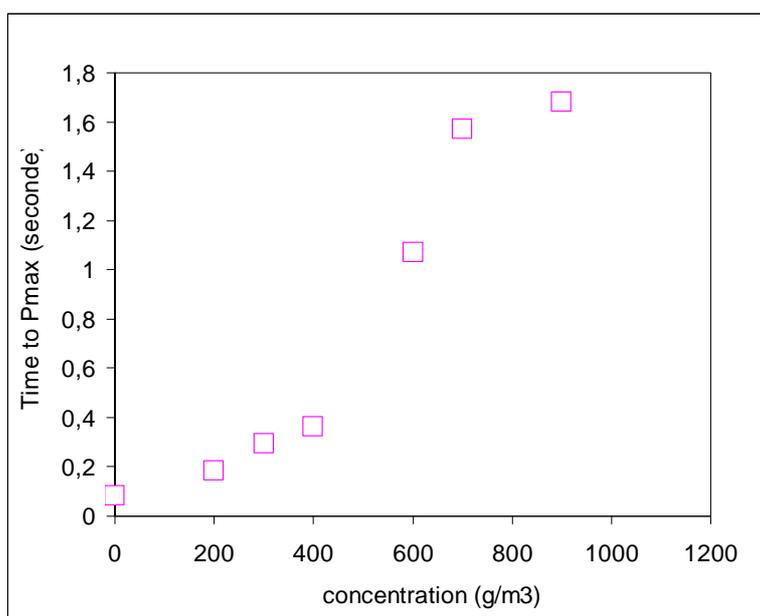
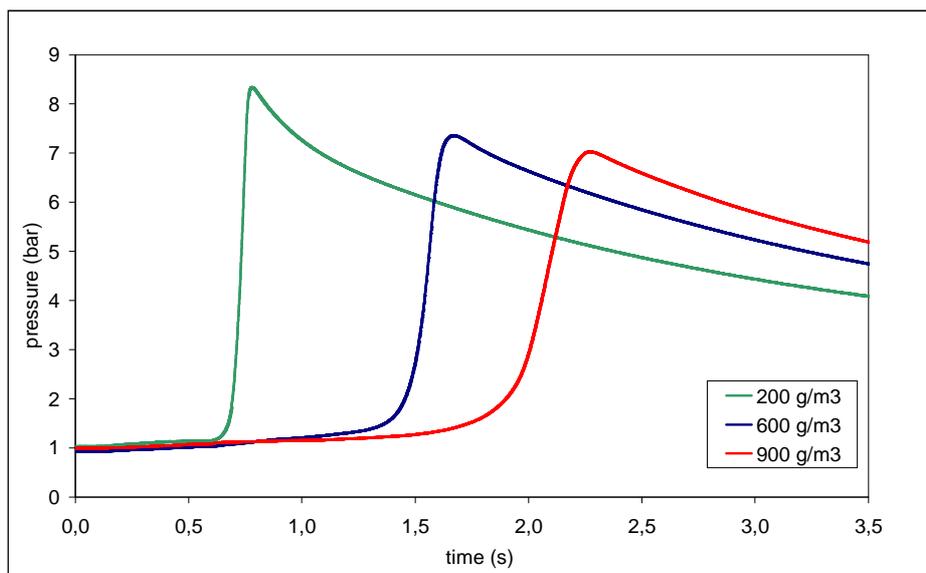


Figure 6 : P-t curves ($t=0$ start of the discharge of the dust) for various amounts of sodium bicarbonate dust (up) and combustion times (down)

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Figure 7 : Ignition devices. From left to right : 5000J electric igniter, flash powder, 60J pyrotechnical match

3.2 Experimental conditions

A number of extinguishing powders were tested starting with a “standard” bicarbonate dust having a volumic mean diameter (VMD) of 20 μm around a rather spectrum of particle sizes (from a few tens of microns to 100 μm –figure 8-top left). Sieved fractions of this powder were studied (figure 8) to investigate the particle size effect (below 40 μm , between 40 and 63 μm , between 63 and 125 μm and above 125 μm). Since the decomposition of these powder may come into play, talc (MgO_2) powder was also tested having a particle size distribution resembling that of the “standard” bicarbonate dust (figure 8-bottom right).

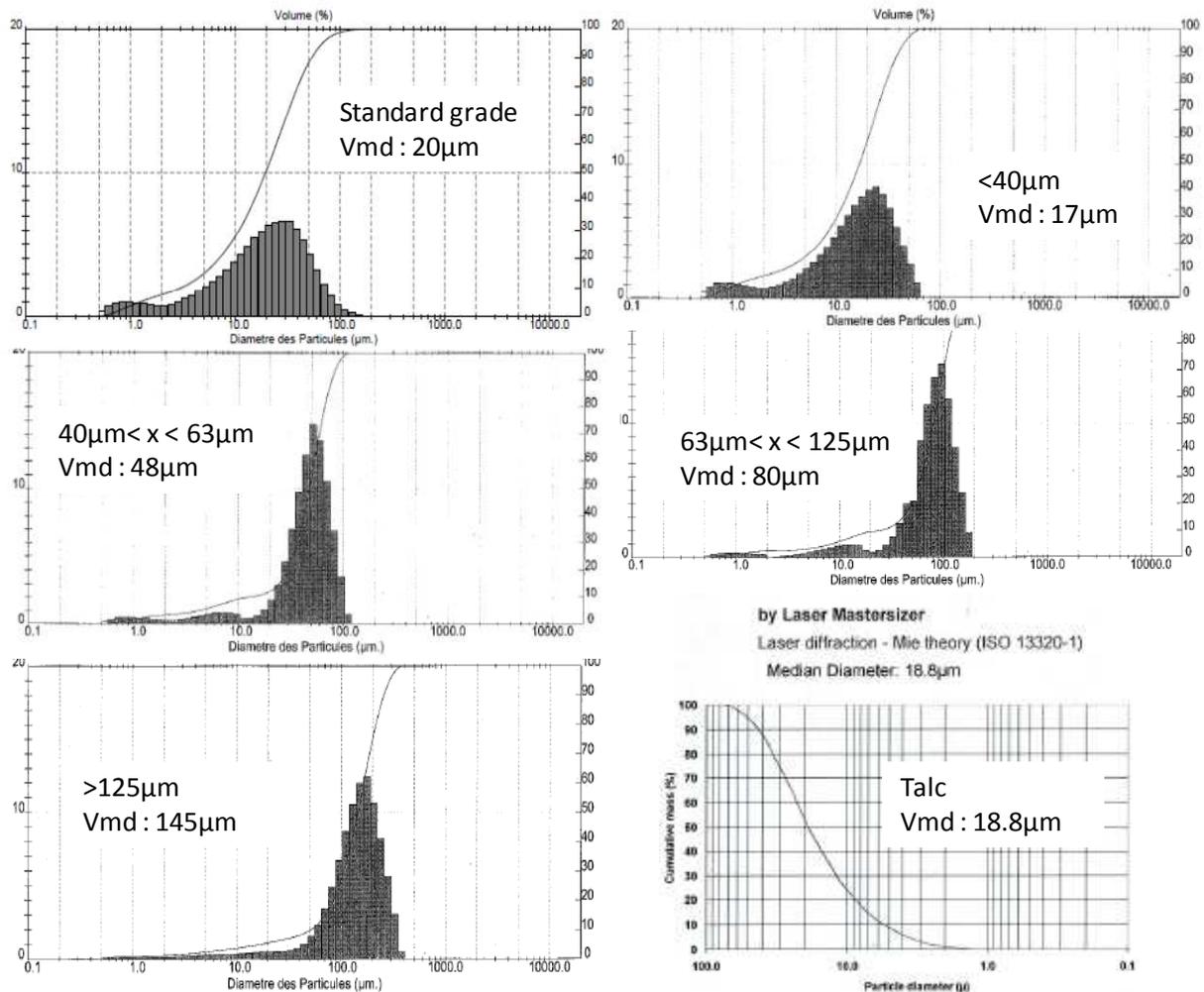


Figure 8 : particle size distribution of the various extinguishing powders investigated.

3.3 Results

Typical P-t curves are presented on figure 9. A pressure “jump” is seen at time 700 ms corresponding to the burning of the flash powder. Soon after the explosion is triggered for talc dust and the coarser bicarbonate powder although the amount of dust dispersed is extremely high suggesting the ignition procedure is reasonably successful. Ignition fails to develop a propagating flame with the standard bicarbonate powder when the dust amount is 1200 g/m^3 . Note however that the maximum overpressure is larger than expected if the igniter was alone (1.5 bar overpressure as compared to less than 1 bar) suggesting the combustion is locally “forced” by the igniter. Further, it might be considered that extinction is reached (or more precisely “non propagation”) when Pmax remains below 3 bar (absolute pressure).

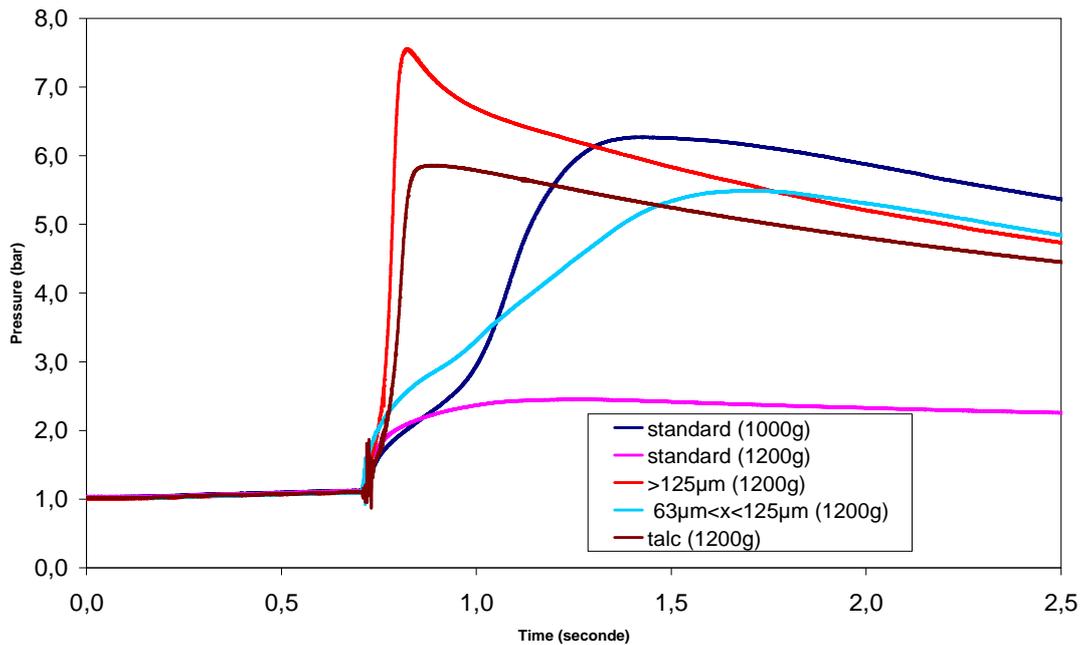


Figure 9 : typical $P-t$ ($t=0$ at the start of the discharge-ignition at 0.7s) curves for different dusts

The evolution of P_{max} for the different powders is shown in figure 10. On figure 11, $(dP/dt)_{max}/P_{max}$ is shown (it is characteristic of the flame speed).

For the “standard” bicarbonate powder, extinction appears as a “rupture” in the evolution of P_{max} as function of the dust concentration in accordance with the theoretical approach (red stars in figure 2). The critical concentration at extinction is about 1100 g/m^3 . The dotted blue line on figure 10 corresponds to the predicted evolution of P_{max} assuming a simple cooling of the mixture by heat conduction through the particles and thermal equilibrium between the two phases. Apparently, the standard powder seems to follow this trend. The relative role of the various fractions of the particle distribution is discussed below.

Very coarse bicarbonate particles ($> 125 \mu\text{m}$) do not seem to be able to absorb much heat in the combustion product because P_{max} is only modestly modified by the presence of the particles. This particle mass fraction represents less than 1% m/m of the standard dust.

Intermediate particle distributions ($63 \ll 125 \mu\text{m}$ and $40 \ll 63 \mu\text{m}$) give similar results than the standard dust. Note that these fractions represent respectively 10% and 10% m/m of the standard dust.

The finest particles (below $40 \mu\text{m}$) represent about 80% m/m of the standard dust (it is the reason why the VMD are similar for the finest grade and the standard dust). The critical concentration required to quench the flame is about 1400 g/m^3 . It is significantly larger than for the standard dust (40% more).

Talc powder has the same heat capacity than sodium bicarbonate dust. The particle size distributions are similar for both dusts and, following, the points on figure 10 align well on the dotted blue line. However, the mitigation capacity of talc powder seems much smaller as shown on figure 11 and there is much less sign of flame weakening at concentrations as large as 1200 g/m^3 . This last result suggests that the phase transition/decomposition of sodium bicarbonate plays a significant role in the process of flame quenching. If the results obtained

with talc powder are extrapolated (pink dotted line in figure 11), the talc dust concentration required to quench the flame would amount about 1400 g/m³ which is about 40% larger than for the sodium bicarbonate dust.

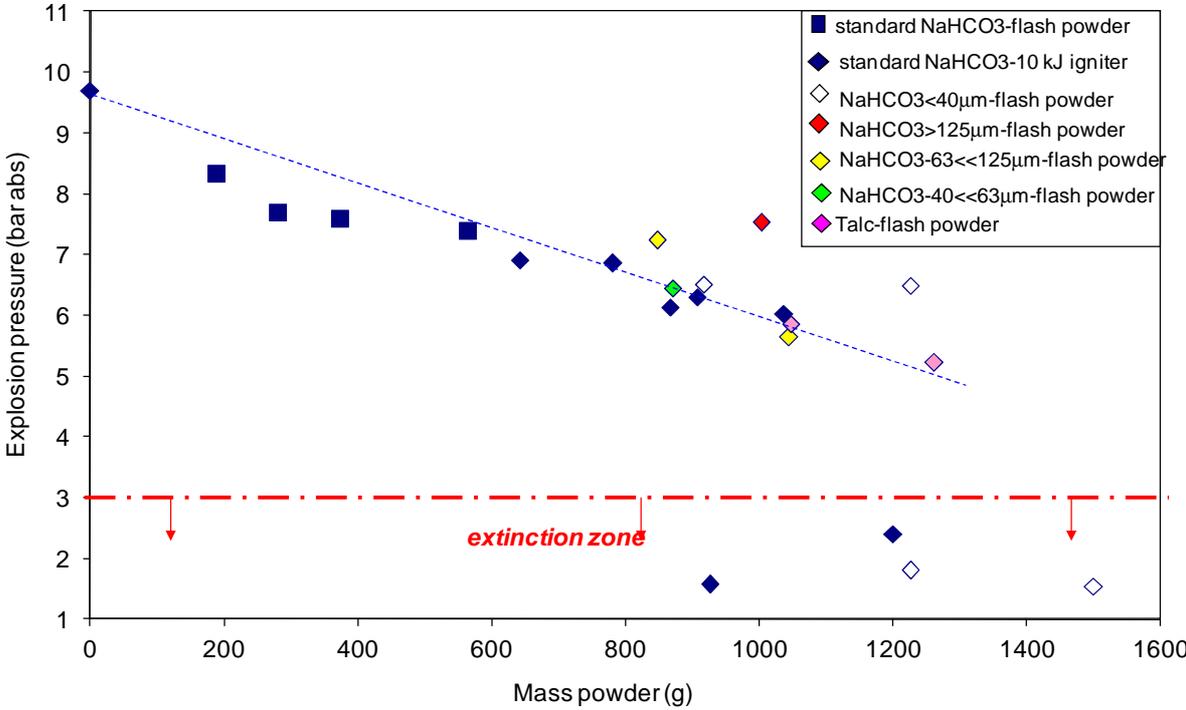


Figure 10 : Pmax versus powder concentration and nature

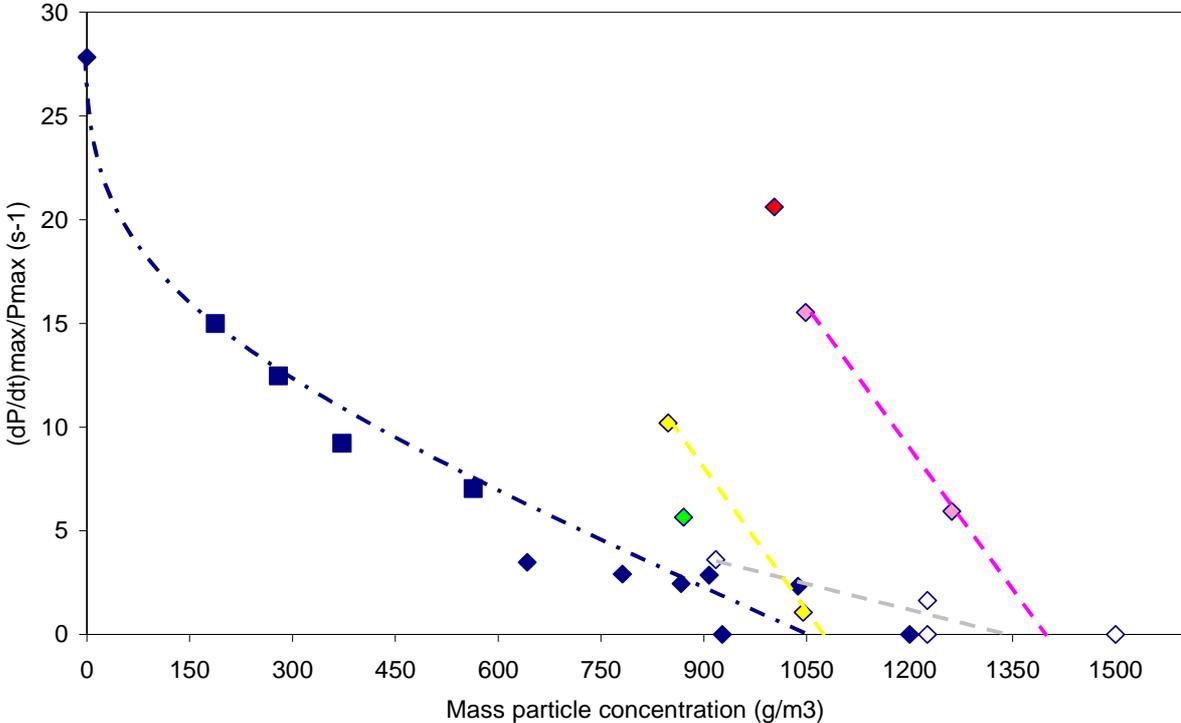


Figure 11 : (dP/dt)max/Pmax versus powder concentration

Following the reasoning of in the theoretical section of this paper, very small sodium bicarbonate particles (below 40 μm) should be in thermal equilibrium and may decompose entirely. Heat conduction through the particle might not be large enough to quench the flame but the decomposition of one mole of NaHCO_3 would release about 1 mole of CO_2 . To avoid a flame to propagate in a stoichiometric methane-air mixture, a volume of CO_2 corresponding to a third of the volume of the initial mixture should be admixed (to reach a CO_2 concentration of about 25% v/v). In the present experimental condition, this is about $0.33 \times 1 / 0.024 = 14$ moles of CO_2 (1 is the volume of mixture contained in the vessel before ignition and 0.024 is the approximate molar volume). Assuming the sodium bicarbonate dust is completely pyrolysed, a minimum quantity of dust of $84 \times 14 = 1200 \text{ g/m}^3$ is required (84 g/mole is the molar mass of sodium bicarbonate). This is in reasonable agreement with the observed dust concentration required to quench the flame for the finest particles. Then, the quenching of the flame in this situation might result from the inerting effect of CO_2 .

The standard dust contains a very large proportion the finest particles (below 40 μm). Following, the standard dust should behave mostly as an inerting gas via a significant release of CO_2 . The flame temperature and burning velocity would be reduced by the dilution effect and consequently the flame would be weaker. The largest particles (below 40 μm) would add a conduction heat sink. A small additional quantity might then be enough to quench the flame. Because of this, the quenching concentration could be smaller than 1400 g/m^3 as observed.

Noteworthy, the quenching capacity of the sodium bicarbonate particles ranging from 40 to 125 μm appears similar to that of the standard dust. So the rather broad particle size distribution of the standard dust (typically between 1 and 100 μm) is certainly a good compromise which may provide the smallest extinguishing dust concentration with a relatively weak dependency on the particle size distribution.

4. Conclusions

A theoretical and experimental work is proposed to investigate the various extinguishing modes of powders used in suppression systems. A number of parameters may influence the phenomenology and especially the particle size distribution. It is suggested that sodium bicarbonate dust may quench the flame either because of the heat extracted in the flame front to heat up the particles or because of the inerting CO_2 evolved during the pyrolysis process. Experiments confirm that this dust is much more effective than purely inert powders like magnesium oxide (talc). It seems that very small particles could be gasified and could dilute the reactive mixture whereas the larger ones may also act on the flame by extracting heat by conduction. A good balance may be found when a broad particle distribution is offered knowing that particles larger than 100 μm may not be efficient.

However, the experimental evidence remains rather limited, partly because the experiments are very difficult to perform. Also, a number of assumption need to be released starting with understanding the real behaviour of NaHCO_3 particles. A significant piece of work is to be completed to represent entirely the quenching process in a flame.

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