

## Overall characterization of cork dust explosion

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### Abstract

Explosibility and ignitability studies of air/cork dust mixtures were conducted in a near-spherical 22.7 L explosibility test chamber using pyrotechnic ignitors and in a furnace of 1.23 L. The suspension dust burned as air-dispersed dust clouds and the uniformity of the dispersion inside the chamber was evaluated through optical dust probes. The range of tested particle sizes went from a mass median diameter of 47.4 to 438.3  $\mu\text{m}$  and the covered dust cloud concentration was up to 700–800  $\text{g}/\text{m}^3$ . Measured explosion parameters included minimum explosible concentration, maximum explosion pressure, maximum rate of pressure rise and minimum autoignition temperature.

The effect of dust particle size on flammability was evaluated and it was found that the minimum explosible concentration is around 40  $\text{g}/\text{m}^3$  and it is relatively independent of particle size below 180  $\mu\text{m}$ . Maximum explosion pressure of 7.2 bar and maximum rate of pressure rise of 179 bar/s were detected for the smallest tested sizes. The limitations on the rates of devolatilization of the solid particles became rate controlling at high burning velocities, at high dust loadings and for large particle sizes.

The effect of initial pressure on the characteristic parameters of the explosion was studied by varying the initial absolute pressure from 0.9 bar to 2.2 bar, and it was found that as initial pressure increases, there is a proportional increase of minimum explosion limit, maximum explosion pressure, and maximum rate of pressure rise.

The influence of the intensity of the ignition energy on the development of the explosion was evaluated using ignition energies of 1000 J, 2500 J and 5000 J, and the experimental data showed that the value of 2500 J is the most convenient to use in the determination of minimum explosion concentration.

The behavior of the cork dust explosion in hybrid methane air mixtures was studied for atmospheres with 2% and 3.5% (v/v) of methane. The effect of methane content on the characteristic parameters of the explosions was evaluated. The conclusion is that, the hazard and explosion danger rise with the increase of methane concentration, characterized by the reduction of the minimum dust explosion concentration.

The minimum autoignition temperature obtained with the thermal ignition tests was 540 °C and the results have shown that this value is independent of particle size, for particles with mass median diameters below 80  $\mu\text{m}$ .

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### 1. Introduction

Cork is the outer bark of *Quercus suber* L. Its particular cellular structure of alveolar suberized cloth is formed by dead micro-cells with polyhedral shape of 14 sides, thatching each other with the inter-cellular spaces totally filled with a gaseous mixture similar to the air [1]. Cork can be chemically divided in six essential groups [1,2]: the major cork component, around

45% (w/w), is the suberin (mixture of acid components), the second one is lignin (complex polyester of high molecular weight with a not well-known structure) representing 27% (w/w) of cork composition and the third fraction, representing about 12% (w/w), are polysaccharides (formed essentially by cellulose and hemicellulose). The smallest fractions are divided in 5% (w/w) of ceroids (group of components with small polarity), 6% (w/w) of tannins (polyphenolic components) and 5% (w/w) of other components (minerals, water and glycerine).

Portugal has a dominant position in the world of cork industry. With some frequency, every other year accidents happened

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in this industry, with losses of human lives and destruction of industrial facilities. Unfortunately, studies on the characterization of explosion conditions on the mechanisms of initiation, development and propagation of explosions of this dust are rare, and this has been the reason for the accomplishment of a series of experiments to look for important data on the matter.

The objective of this paper is to present data about the overall behaviour of cork dust explosibility and ignitability. Information about the limits of characteristic explosion parameters in dust/air mixtures, such as minimum dust explosion concentration, maximum explosion pressure, maximum rate of pressure rise and minimum ignition temperature, is given. The influence of some factors, like particle diameter, initial pressure, ignition energy and methane presence on the development of the explosion, are analysed.

## 2. Experimental

### 2.1. Apparatus and procedures

#### 2.1.1. Explosibility studies

The explosibility studies of cork dust were carried out in a near-spherical 22.7 L explosibility test chamber, as shown in Fig. 1a. This chamber is made of stainless steel according to the recommendations of ASTM E1515 [3] and ASTM E1226 [4], and was built according to the dimensional characteristics of the Bureau of Mines 20 L chamber, provided by the Pittsburg Research Laboratory of the National Institute for Occupational Safety and Health (NIOSH). This is a standard laboratory test chamber used extensively for dust explosions studies of carbonaceous and elemental dusts [5–7]. One optical dust probe [8,9] was used to evaluate the uniformity of the dust dispersion by measuring the light transmission over a path of 38 mm length through the dust cloud. The optical probe was also built based on the design supplied by the Bureau of Mines. The evolution of the explosion pressure and rate of pressure rise during the explosion was measured with a piezoresistive pressure transducer with internal diaphragm, for absolute pressures in the range of 0–10 bar. Although this technique is limited because the dispersion of the particles is only verified along a 38 mm path, it can be used as a methodology for a comparative assess-

ment of the quality of the experiments used in the determination of relevant data.

The experimental tests were realized according to the specifications of the standard test procedure [3]. After the dust and the ignition source have been placed in the chamber, the top was bolted on and the chamber was partially evacuated to an absolute pressure of 0.19 bar. Then a short blast of air coming from a 16 L auxiliary pressure vessel with a 0.3 s duration dispersed the dust, and increased the pressure inside the chamber to about 0.9 bar absolute pressure. The ignitor, located in the centre of the reactor, was activated after an additional delay of 0.1 s, resulting in a total ignition delay of 0.4 s since the start of dispersion until ignition. The used ignition sources were electrically activated pyrotechnic ignitors with energies of 1000 J, 2500 J and 5000 J, composed of 40% zirconium, 30% barium nitrate and 30% barium peroxide, and were supplied by Fr. Sobbe from Germany. They delivered their energy in about 10 ms and when ignited produced a dense cloud of very hot particles and little gas [5]. The experimental dust concentration reported for the chamber is the mass of dust divided by the internal chamber volume. Measured explosions parameters included minimum explosion concentration, maximum explosion pressure and maximum rate of pressure rise.

#### 2.1.2. Ignitability studies

The ignitability studies of cork dust in air mixtures were carried out in a ceramic 1.23 L furnace according to the recommendations of ASTM E1491 [10]. This furnace was based also on the design of Bureau of Mines 1.2 L Furnace, as described in Conti Cashdollar and Hertzberg [11], and is shown in Fig. 1b; it has an internal ceramic combustion chamber, made of magnesium aluminium silicate, wrapped with heater wire and surrounded by thermal isolation.

The experimental tests were realized according to the specifications of the same standard test procedure and the minimum autoignition temperature was measured. The dust to be tested is placed in the disperser, which is manually inserted into the bottom part of the furnace. A fibreglass rupture diaphragm at the top of the furnace isolates the 1.23 L volume from the exterior. A 30 ms air dispersion pulse disperses the dust as it releases about 60 cm<sup>3</sup> of air at room temperature and

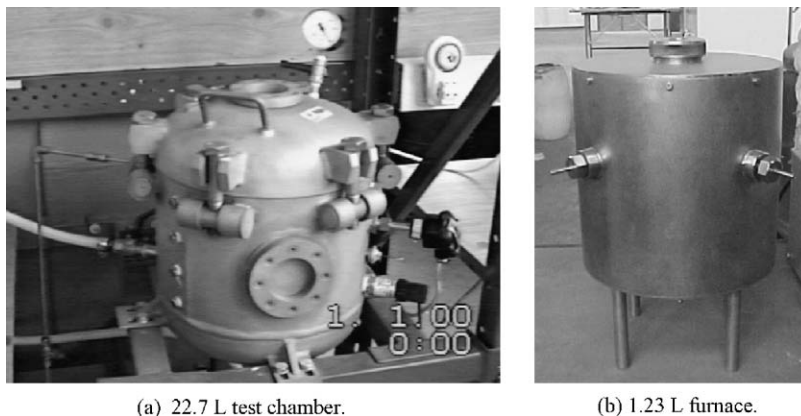


Fig. 1. Apparatus used in the experiments.

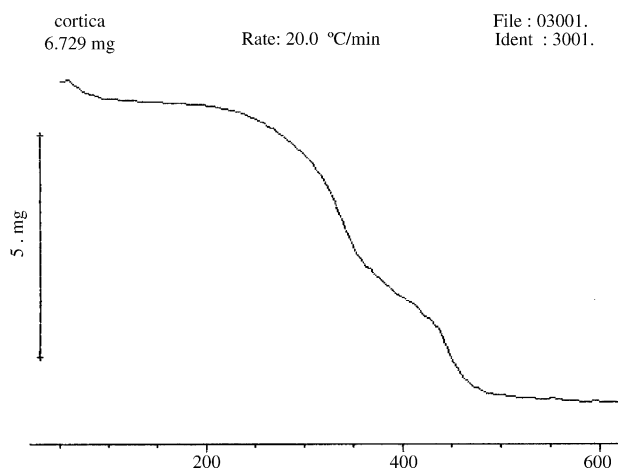


Fig. 2. TGA curve of cork dust.

atmospheric pressure from a pressurized reservoir into the furnace. The criterion for ignition is the rupture of the diaphragm, with the simultaneous emission of flame from the top of the furnace.

## 2.2. Material

The approximate analysis of the cork dust reveals that it is composed mainly by 92.6% of volatile matter, 5.4% of moisture and 2% of fixed carbon in mass basis. The thermogravimetric analysis (TGA) was used for measuring the volatilization rate of cork dust as function of temperature and the results are represented in Fig. 2. The profile of TGA trace shows that

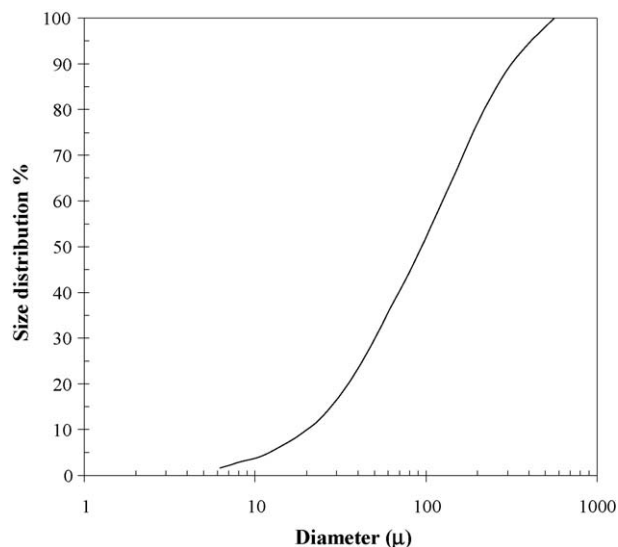
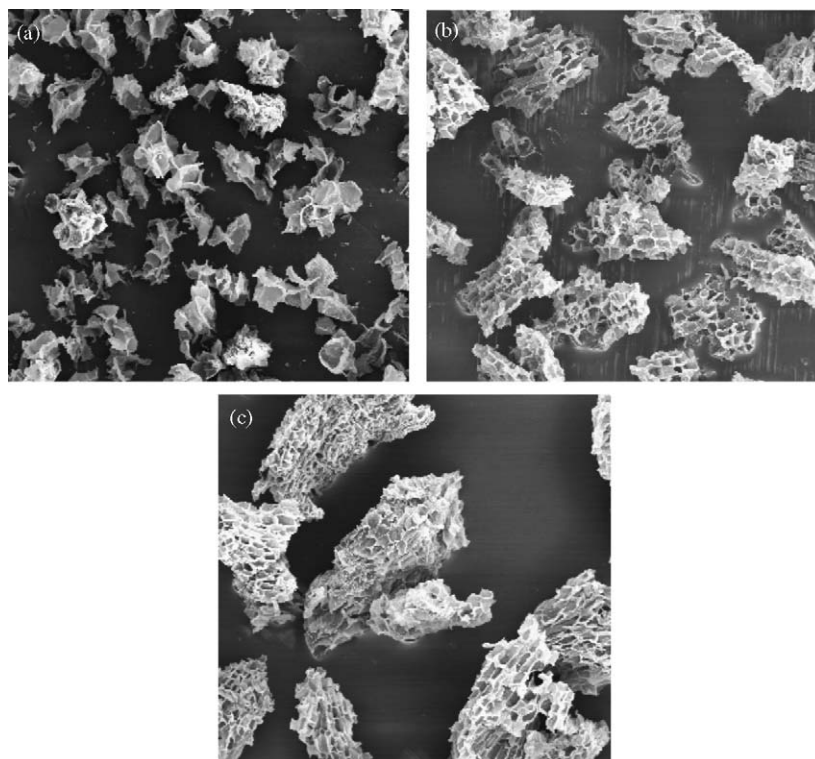


Fig. 3. Cumulative curve of cork dust particle size distribution.

there are significant weight losses at four different temperature ranges. The first one, until 100 °C, corresponds mainly to the drying process due to the evaporation of moisture and represents 6% of mass loss. The second one, at the temperature range where the greater mass loss is verified, is around 50%, and it is attributable essentially to the suberin decomposition. The peak around 380 °C is related to the cellulose decomposition and corresponds to a mass loss of 18%, while the final temperature peak, in the range of 450 °C, is the most pronounced one and is related to the decomposition of the more thermally stable com-

Fig. 4. Morphology of cork dust particles. (a)  $d_p = 71.3 \mu\text{m}$ , (b)  $d_p = 222.1 \mu\text{m}$  and (c)  $d_p = 366.4 \mu\text{m}$ .

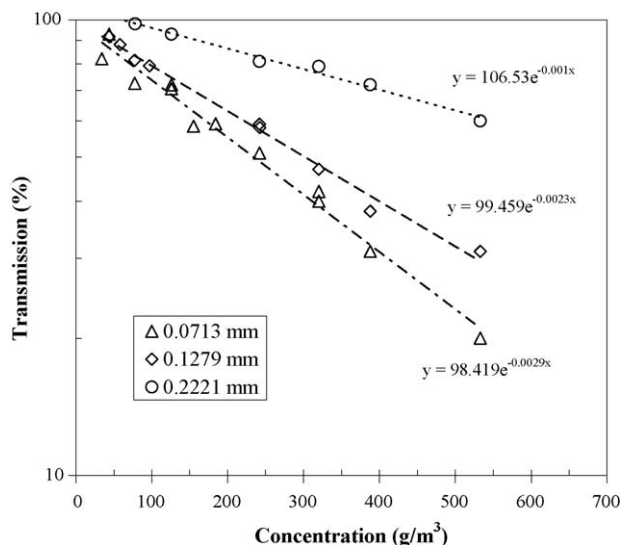


Fig. 5. Effect of concentration and particle size on the transmission through the cork dust cloud.

ponent of the cork, the lignin [12]. The cork dust is completely devolatilised at temperatures greater than 500 °C, if the exposition time is long enough.

The combustion properties of a dust are dependent of both, its chemical and physical characteristics, namely the particle diameter distribution and shape. Fig. 3 represents the volume weighted curve for the dust particle size distribution of the cork dust.

In this work, six different particle sizes were studied and the corresponding fractions were obtained by screening the dust using ASTM sieves. The mass median particle diameters of each fraction were then measured with a Coulter Counter.

Examples of scanning electron microscope photomicrographs of three cork dust particles sizes are shown in Fig. 4, where the peculiar cellular structure of the particles, in alveolus shape, can be observed.

The photomicrographs show clearly, that greater particle sizes have a stronger deviation from the spherical shape assuming a long geometry.

The easiness with which a dust is dispersed in the air is function of several factors such as individual particle density, diameter, shape, cohesive properties of the particles and their adhesive properties towards the confining surface walls. Some external factors play an important role in the dispersion process like the structure and intensity of the aerodynamic perturbations, the loading position of the dust and the surface geometry of the testing reactor [13].

The relative efficiency of the dust dispersion inside the semi-spherical reactor of 22.7 L was studied using an optical dust probe. This probe measures the light transmission through the dust cloud in suspension, and in Fig. 5 are shown the transmission data for three of the tested particle sizes. The transmission ( $T$ ) is related to the mass concentration ( $C$ ) by the Bourgue's law [8]. The results show a good agreement with this law following the expected linear relationship on the semi-logarithmic plot.

As previously stressed, this approach is limited; it allows however the adoption of an acceptance criterion for the validity of the experiments.

### 3. Results

#### 3.1. Explosibility of cork dust in air

##### 3.1.1. Effect of concentration and particle size

The explosibility study of cork dust/air mixtures comprises the experimental determination of the minimum explosible dust concentration, the maximum explosion pressure and the maximum rate of pressure rise.

Six particle sizes were tested with mass median diameters of 47.4  $\mu\text{m}$ , 71.3  $\mu\text{m}$ , 127.9  $\mu\text{m}$ , 222.1  $\mu\text{m}$ , 366.4  $\mu\text{m}$  and 438.3  $\mu\text{m}$ , and experiments were performed with dust concentrations that went up to 600–800  $\text{g}/\text{m}^3$ , using 2500 J of ignition energy. From these tests, traces representing the evolution of explosion pressure as function of time are obtained; the peak of the traces ( $P_{\text{max}}$ ) and the steepest slope of the rising portion of the curves ( $dP/dt$ ) can be both represented as function of concentrations, for each one of the studied particle sizes. Fig. 6 represents the development of pressure ratio PR, and rate of pressure rise  $dP/dt$ , as function of dust concentration for particles with mass median diameter of 71.3  $\mu\text{m}$ . PR is defined as  $\text{PR} = (P_{\text{max}} - \Delta P_{\text{ign}})/P_i$ , where  $\Delta P_{\text{ign}}$  is the pressure rise promoted by the activation of the ignition source and  $P_i$  is the initial pressure in the reactor.

The increase of pressure ratio with the nominal dust concentration is the result of the increase of volatiles concentration in the atmosphere. The theoretical maximum explosion pressure is a thermodynamic parameter related to the heat of combustion of the mixture, predictable under the following conditions [14]: negligible heat losses, total oxygen consumption and complete combustion of the stoichiometric amount of dust, relatively to the oxygen content in the atmosphere. The stabilization of pressure ratio value is related with the total oxygen consumption inside the reactor. For these conditions, the progress of the combustion process around the stoichiometric concentration of released volatiles leads to high and constant values for pressure and temperature [15].

The increase of the maximum rate of pressure rise with dust concentration is related again, with the increase of the volatile content, resulting in the increase of the burning velocity  $S_u$ . The stabilization of this maximum rate of pressure rise is related to the stabilization of the burning velocity for high dust concentrations [16]. The decrease of the burning velocity is related with the delay of the devolatilization process. Detailed information about this aspect can be found in Pilão et al. [17] and Pilão [18].

Fig. 7 shows scanning electron microscope photomicrographs of a particle with a mass median diameter of 71.3  $\mu\text{m}$  before (a) and after explosion with concentrations of 160  $\text{g}/\text{m}^3$  (b) and 220  $\text{g}/\text{m}^3$  (c). These photomicrographs present the morphological changes in the particle resulting from the explosion. Significant changes in the residue surface can be observed when comparing images before and after explosion. Some particles form cenospheres as can be observed (b) and the volatiles were

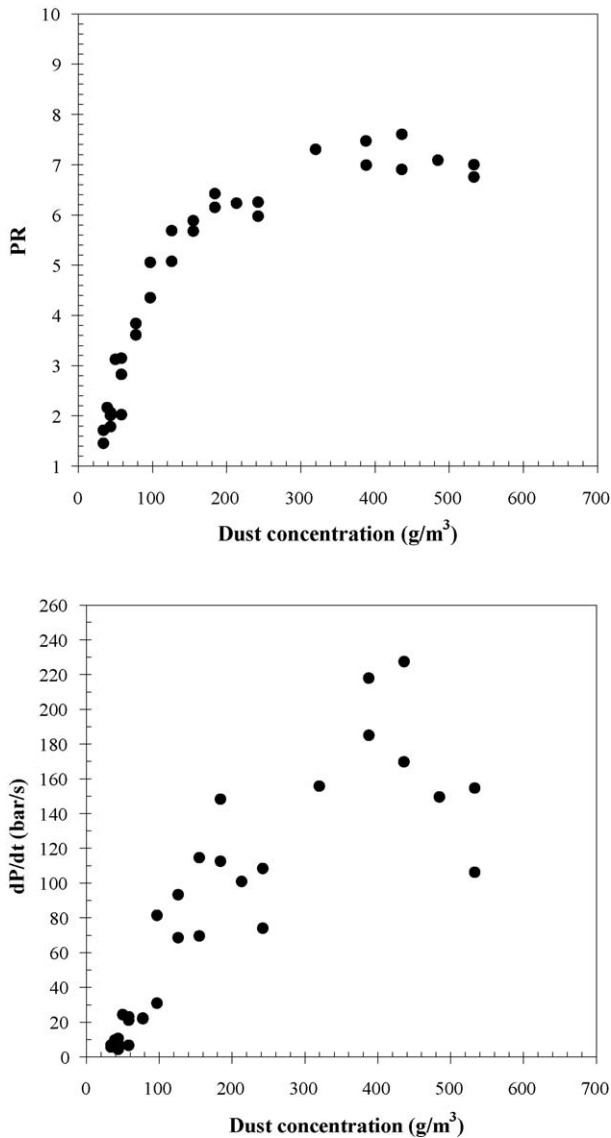


Fig. 6. Development of pressure ratio and rate of pressure rise with dust concentration, for particles with a mass median diameter of 71.3  $\mu\text{m}$ .

emitted through the holes seen in the residues (c). The photomicrographs confirm that, the pyrolysis process proceeds in a devolatilization wave front that starts at the surface of the exposed particle, propagating then through it. The same analysis based in the particle aspect before and after explosion was used by Cashdollar [19], Hertzberg et al. [20] and Hertzberg and Zlochower [21].

**3.1.1.1. Minimum explosible concentration.** Applying the significant flame propagation criteria ( $\text{PR} \geq 2$  and  $dP/dt V^{1/3} \geq 1.5 \text{ bar m/s}$ , where  $V$  is the internal volume of the reactor) [3,5] to the obtained results with the six tested particles sizes; the minimum explosible dust concentration can be obtained for each one, resulting in a trace like that represented in Fig. 8.

This figure shows that for fine particles, with mass median diameters smaller than 180  $\mu\text{m}$ , the minimum dust explosible concentration is independent of particle size, assuming the value of 40  $\text{g/m}^3$ . Above this diameter, defined as the characteristic

diameter, a rapid increase of the minimum explosible concentration can be observed with the rise of the particle size.

According to the flame propagation mechanism proposed by Hertzberg et al. [20] and Hertzberg and Zlochower [21], the flame propagation is a sequential process starting by the devolatilization of the particles followed by the mixture of the emitted volatiles with the air, and finally by the combustion, in the gaseous phase of the resultant air combustible mixture. This theory stated that the reaction time  $\tau$  of an explosion is composed by two essential components: the devolatilization time of the particles  $\tau_d$  and the combustion time  $\tau_c$ . The time of devolatilization depends on the particle size, while the combustion time is independent of the dimension of the particles, because the combustion process is a gaseous phase phenomenon. This means that a process controlled by the reaction rate is independent of the size of the particles, while a process controlled by the devolatilization rate is dependent on the dimensions of the particles. The flat region of the curve in Fig. 8, means that for particles with sizes smaller than the characteristic diameter, the explosion process is controlled by the chemical reaction rate. However, in the case of cork dust, this flat region does not correspond to situations of complete devolatilization of the particles within the flame front, as is commonly admitted for dust of small sizes [5]. In fact, it was observed that a significant amount of particles remains unburned in the final of the explosion tests, which can be confirmed by the evolution of the burning efficiency, defined as  $\eta = (m_i - m_f)/m_i$ , and represented in Fig. 9. In the previous definition  $m_i$  and  $m_f$  are the initial and the final mass of cork dust, respectively. The results have shown that the combustion efficiency increases with the rise of the dust concentration until it attains the maximum value of 67%. The stabilization of this value is related with the oxygen content inside the reactor, which imposes the maximum burnable volatile content. In conclusion, the flat region of the curve in the Fig. 8 represents situations where the quantity of the volatiles generated is independent of the dimension of the particles and the explosion occurs, if in gaseous phase it will be present a minimum necessary volatile content.

The rapid increase of the minimum dust concentration for particles with dimensions higher than the characteristic diameter (Fig. 8), means that the combustion process is controlled by the devolatilization rate. In these situations, only the particle surface is devolatilized, and consequently a higher mass of dust must be loaded to reach, in the gaseous phase, the minimum dust concentration.

This behaviour confirms that the flame propagation process during the cork dust explosion is preceded by the devolatilization of the particles [20,21].

**3.1.1.2. Maximum explosion pressure and maximum rate of pressure rise.** The maximum values of pressure ratio and rate of pressure rise are plotted against the corresponding particle size, obtaining the graphics represented in Fig. 10.

As can be observed, the explosion violence  $dP/dt_{\text{max}}$  is strongly affected by the particle diameter, while maximum explosion pressure  $\text{PR}_{\text{max}}$  is not so dependent upon the particle size.

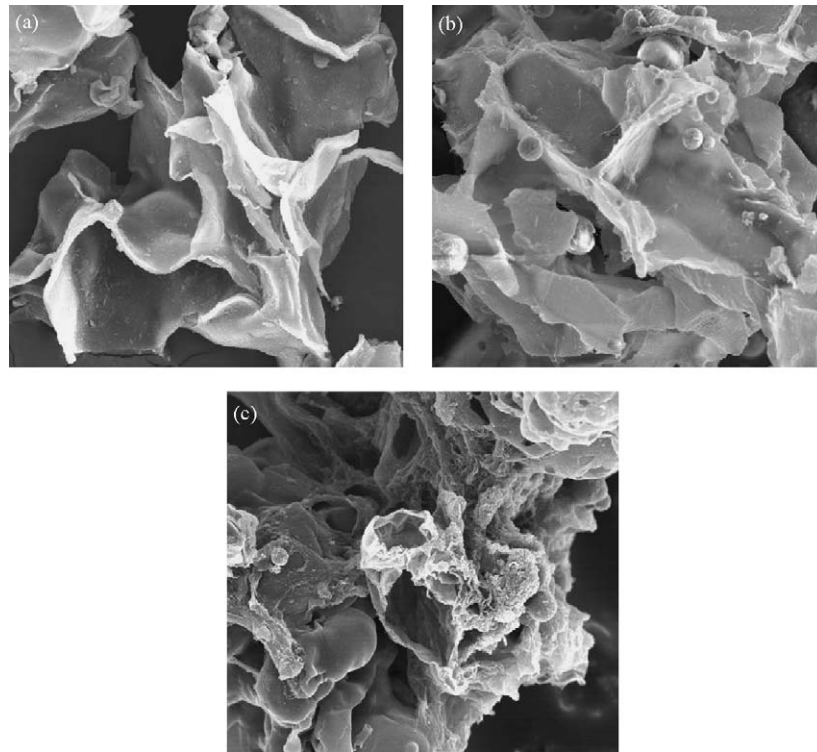


Fig. 7. Scanning electron microscope photomicrographs of a particle with a mass median diameter of 71.3  $\mu\text{m}$  before and after explosion. (a) Before explosion, (b) after explosion 160  $\text{g}/\text{m}^3$  and (c) after explosion 220  $\text{g}/\text{m}^3$ .

The maximum explosion pressure was obtained with the smallest particles tested and has the value of 7.2 bar (a). This value decreases slowly with the increase of particle diameter and the minimum obtained was of 5.8 bar (a) for the bigger particles.

Concerning the maximum rate of pressure rise, a maximum value of 179 bar/s was also obtained with the smallest tested particles.

The slow decrease of maximum explosion pressure with the increase of particle diameter means that small changes in the

generated volatile content are verified, and it is also an indication that nonadiabatic effects can increase with the rise of the particle dimensions [22]. Radiative and convective heat losses from the fireball towards the walls and internal structures of the reactor augment, when the combustion time increases because of the decrease of the combustion velocity, which is associated to an increase of the size of the particles.

For high dust loadings, conditions when the maximum for the explosions parameters are obtained, the devolatilization process cannot be completed within the flame front, and the larger is

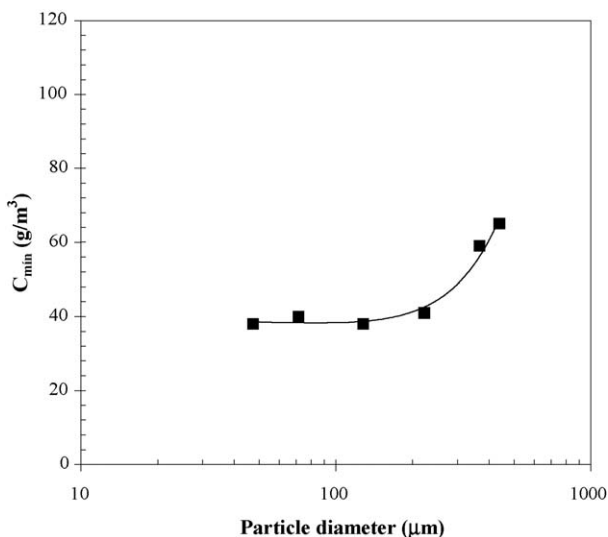


Fig. 8. Effect of particle size on the minimum explosible dust concentration of cork dust/air mixtures.

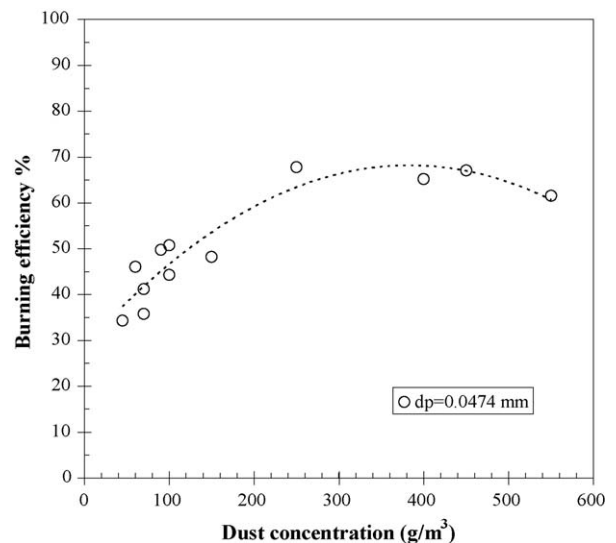


Fig. 9. Evolution of burning efficiency with dust concentration of 47.4  $\mu\text{m}$  particles.

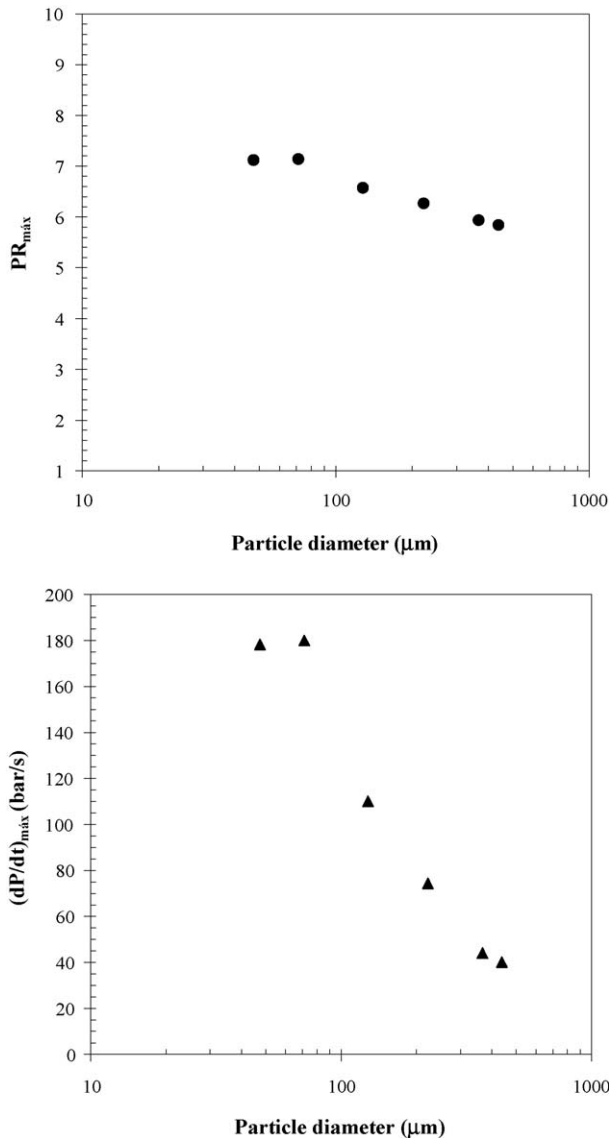


Fig. 10. Effect of particle size on the maximum pressure ratio and maximum rate of pressure rise, of cork dust/air mixtures.

the particle, the greater is the undevolatilized dust mass fraction [6,16]. This increase, plus the increase of the residual char makes no contribution to the flame propagation process and begins to absorb a larger fraction of the flame released heat flux, quantified by  $S_u c \rho (T_b - T_u)$  [20], where  $S_u$  is the velocity of the flame front,  $c$  and  $\rho$  are the specific heat and the density of the gas, respectively, and  $T_u$  and  $T_b$  are the unburned and burned gas temperatures. As a result of these phenomena, the effective heating flux driving the fresh pyrolysis wave, ahead of the flame front, is diminished.

The strong dependency of the rate of pressure rise  $dP/dt$ , relatively to the particle diameter, is related to the strong decrease verified in the devolatilization rate when the particle size is raised. This behaviour affects the combustion rate, and as a result, a strong decrease in the burning velocity  $S_u$  is verified. The subsequent increase of the combustion time leads to the increase of heat losses to the exterior of the reactor, contributing

for the reduction of the maximum explosion pressure (Fig. 10). A more detailed explanation about the phenomena involving the behaviour of these two characteristic explosion parameters can be found in Pilão et al. [17].

### 3.1.2. Effect of initial pressure

Two particle sizes were chosen for the study of the influence of the initial pressure on the development of the explosion: one in the region of finest dust with 71.3  $\mu m$  mass median diameter and other in the region of coarsest dust with 222.1  $\mu m$  mass median diameter. In order to study the influence of initial pressure, on the characteristic parameters of the explosion of cork dust air mixtures, tests were made at three different initial absolute pressures of 0.9 bar, 1.4 bar and 2.2 bar and at constant initial ambient temperature using 2500 J of ignition energy. These pressures were obtained through different combinations between the air pressure in the auxiliary dust dispersing vessel and the vacuum created in the test chamber before the dust dispersion. Considerations related to these test procedures can be found in Pilão et al. [23].

The observation of the results about the effect of the rise of initial pressure on the explosion development for particles with small diameters, have shown a normal development of the explosion with a typical profile of pressure time trace for near-spherical flame propagation [13,23].

Concerning the explosion development in suspensions of particles with large diameters and concentrations near the lower flammability limit, the increase of initial pressure is clearly conditioning the initial development of the explosion wave and modifications in the combustion process during the explosion are evident. For the tests carried out with those particles at initial absolute pressure of 2.2 bar and low dust concentrations, it was clearly observed a two-step flame propagation process in the development of the explosion pressure. This process is characterized by the appearance of two ramps on the explosion pressure curve, which is confirmed by the two peaks observed in the evolution of the curve of the rate of pressure rise. The first step of this process, represented by the first pressure rise after ignition, was associated with small initial burning velocities and with the upward and horizontal direction of flame propagation, while the second step, represented by the subsequent pressure rise, was associated with downward flame propagation at a fast burning velocity [24,25]. Detailed information and explanations about this behaviour and the phenomena involving it can be found in Pilão et al. [23].

Figs. 11 and 12 show, for the two particle sizes tested, the influence of initial pressure on the evolution of maximum explosion pressure and maximum rate of pressure rise. It is clear that an increase of the initial pressure results in an increase of both the maximum explosion pressure and the maximum rate of pressure rise. This behaviour was expected because the oxygen content in the atmosphere increases with the initial pressure, leading to wider burnable volatile concentration for the same nominal dust concentration.

The experimental data obtained with the tested diameters reveal that as particle size is reduced, the more violent is the explosion, for the same initial pressure. For the finest particles,

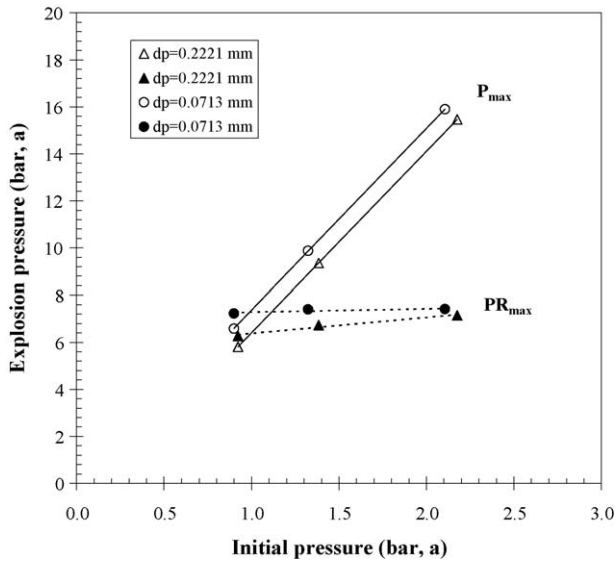


Fig. 11. Influence of initial pressure on maximum explosion pressure.

maximum values of rate of pressure rise are in a range that is more than one order of magnitude higher than the range associated to the larger particles. This behaviour confirms the fact that the maximum explosion pressure is a thermodynamic component, while the rate of pressure rise has a kinetic component [15].

The pressure ratio PR is also represented in Fig. 11, and the results show a small increase for the case of particles with 222.1  $\mu\text{m}$  mass median diameter, giving the idea that, as the initial pressure is raised, the more efficient and adiabatic is the combustion process during the explosion. This behaviour can be related to the increase of overall combustion velocity associated with the raise of initial pressure. The heat losses to the exterior are reduced and approach those verified in the case of the finest particles that were tested.

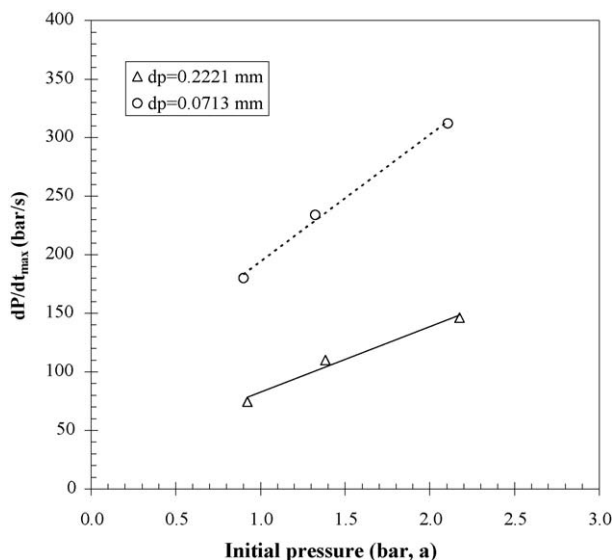


Fig. 12. Influence of initial pressure on maximum rate of pressure rise.

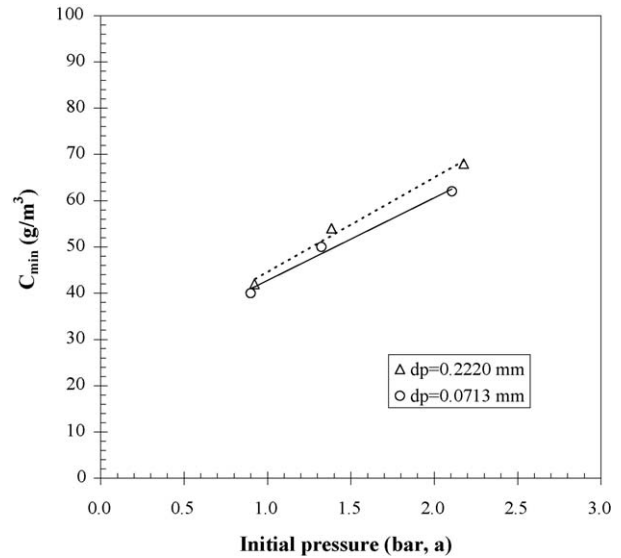


Fig. 13. Influence of initial pressure on minimum explosible dust concentration.

Regarding the evolution of the minimum explosion concentration, it can be observed in Fig. 13 that it increases linearly with the initial pressure and it can be noticed that in the range of particle dimensions that were tested, the minimum explosion concentration is relatively independent of particle diameter. This increase results from the augmentation of oxygen content in the atmosphere. These observations justify the conclusion that, the combustion behaviour of the fine cork dust at lean concentration limits approaches that of an equivalent homogeneous premixed gas. Accordingly, it can be assumed that the overall flame propagation mechanism in the dust cloud becomes controlled by the gas-phase reaction rate.

### 3.1.3. Effect of ignition energy

The influence of the intensity of the ignition energy on the development of the explosion was evaluated for particles of cork dust with a mass median diameter of 47.4  $\mu\text{m}$ , using ignition energies of 1000 J, 2500 J and 5000 J. The 1000 J ignitors by themselves produced a pressure rise of about 0.10 bar inside the chamber, while the pressure rise promoted by the 2500 J, and the 5000 J ignitors were about 0.25 bar and 0.45 bar, respectively, as verified in calibration and set up experiments. Concerning the rate of pressure rise associated with the activation of tested ignition sources, typical values of 11 bar/s, 25 bar/s and 60 bar/s were obtained for the ignition sources of 1000 J, 2500 J and 5000 J, respectively [18].

Different characteristics of the development of the explosion pressure can be observed, indicating a strong influence of the ignition energy of the pyrotechnic ignitors on the explosion dynamics. Fig. 14 represents the evolution of explosion pressure and rate of pressure rise for suspensions with dust concentrations of 400  $\text{g}/\text{m}^3$  and 550  $\text{g}/\text{m}^3$ , initiated with the three tested ignition sources. More examples of pressure–time traces can be found in Pilão et al. [26].

The explosions activated with 1000 J of energy have a slow development of the explosion pressure wave. The ignition source



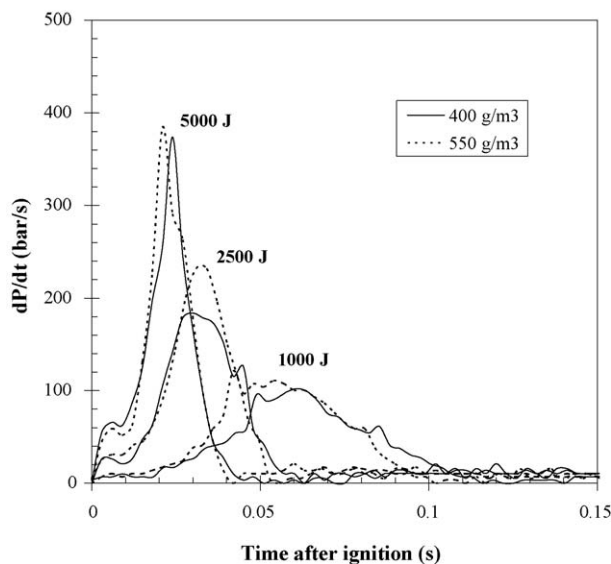
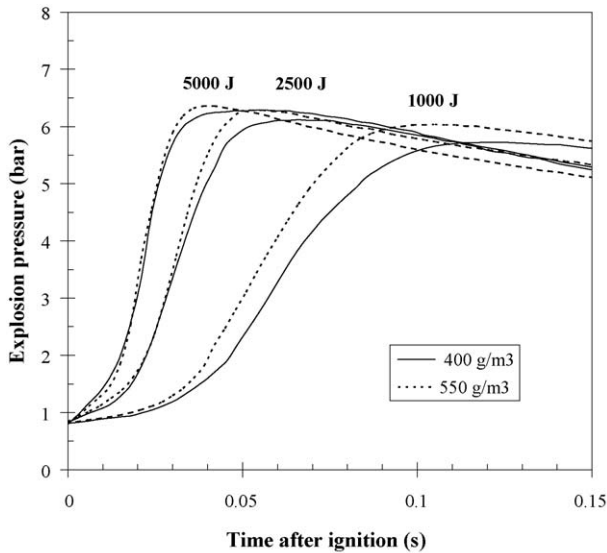


Fig. 14. Effect of ignition energy on the development of explosion pressure and rate of pressure rise for suspensions with high dust concentrations.

of 2500 J shows a moderate effect at the beginning of the explosions with an initial development of explosion pressure–time profile typical of centrally activated explosions. In the case of the activation of explosions with 5000 J ignition sources, the rise of the initial explosion pressure was clearly promoted by the pressure wave created by the activation of the ignition charge, and the rate of pressure rise was largely incremented by the strength of the ignition source.

The maximum explosion pressure was only slightly influenced by the ignition energy and a decrease in the maximum rate of pressure rise with the decrease of ignition energy intensity was observed [26]. This is because the maximum explosion pressure is determined mainly by the total energy released, while the burning rate is affected by the dust concentration and turbulence level as well as the ignition energy [27]. A too strong pyrotechnic ignition source promotes the acceleration of the explosion burning rate, due to the stretching of the effective flame front

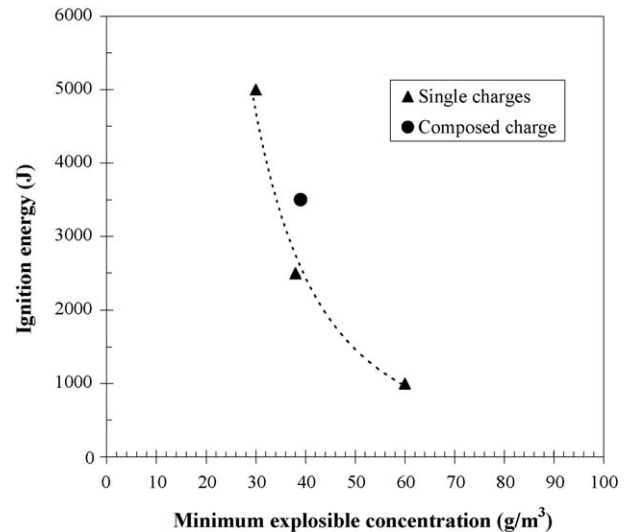


Fig. 15. Influence of ignition energy on minimum explosible concentration.

area [27], hence increasing the maximum rate of pressure rise as well as the burning velocity.

**3.1.3.1. Minimum explosible concentration.** Using the results obtained with experiments with dust concentrations near the flammability limit, the propagation criteria were applied [3,5] and the minimum explosible concentrations of cork dust were found for each one of the tested ignition energies and are shown in Fig. 15. As can be seen, the minimum explosible concentration depends on the value of the ignition energy used in the experiments. A true flammability limit should be independent of the ignition energy and is given by the vertical asymptote of the graphical representation of the ignition energy as a function of the minimum explosible dust concentration [28,29]. The ignition energy of 1000 J is not recommended for the experimental determination of minimum explosible concentration of cork dust, because results are high and away from an asymptotic value, as found with other dusts [28,29]. On the other end, to verify if the 5000 J ignition energy is overdriving the system, tests were made with ignition energies obtained through the combination of one charge of 2500 J and another of 1000 J, resulting in a total energy of 3500 J. In this case the two charges were introduced together in the central part of the reactor trough the same instrumentation port. In Fig. 15 the circle represents the minimum explosible concentration yielded with a composed charge of 3500 J. This last experimental point seems to indicate the existence of a vertical asymptote in the region of 40 g/m<sup>3</sup>, which means that the recommended ignition energy for the experimental determination of minimum explosible concentration is 2500 J, whereas the 5000 J result may be dependent upon some overdriving.

There are however some problems arising from the use of multiple charges which can lead to suspicious results. The results of explosibility tests obtained with the composed charges of 3500 J have shown that only part of them have a similar behaviour to what was obtained for single charges. The same dubious behaviour was observed with tests made with an ignition source composed with two charges of 2500 J, when compared

with the corresponding data obtained with a single charge of 5000 J. Same examples of such abnormal behaviour can be found in Pilão et al. [26]. This was attributed to the result of the interaction between the two explosion waves that are generated with the activation of the two charges. The analysis of the results obtained leads to the conclusion that, in the tests classified with different behaviour, there are two possible justifications for it:

- The activation of the two charges is not simultaneous and, in such event, the delayed activation of the second charge could interfere with the development of the explosion wave or even resulting in the quenching of the flame initiated by the activation of the first charge [26].
- A different orientation of the ignitors could involve the formation of a large ignition volume leading to ignition instabilities.

### 3.2. Explosibility of cork dust in methane/air

The explosion behaviour of cork dust in hybrid methane/air mixtures was studied for atmospheres with 2% and 3.5% (v/v) of methane. These atmospheres were commercially available mixtures with 21% (v/v) of oxygen content, where the methane content was obtained through the reduction of the inert gas fraction ( $N_2$ ). Tested dust particles had a mass median diameter of  $71.3 \mu\text{m}$  and the experiments were performed for dust cloud concentrations up to  $550 \text{ g/m}^3$ , using ignition energies of 2500 J. The methane was chosen because its explosible behaviour is known and the gas has been extensively used in the study of hybrid mixtures, using dusts from different proveniences.

In the industry, hybrid mixtures of cork dust, air and explosive gases are formed in processes where alcohol-based glues are used, leading to the release of volatile organic compounds into the already existing cork dust air mixtures. The use of methane in the present study, instead of the volatile organic compounds of the industrial situation, is justified as a first step on the study of the hazards involving a typical day by day occurrence.

The explosibility results obtained with the two tested hybrid systems (2% and 3.5%, v/v of methane) have shown that, for clouds with small cork dust concentrations, cork dust mixed with methane and air explodes more violently than when mixed only with air. This behaviour was related to the higher burning rate of the gas and also to the increase of the volatile content in the gaseous phase, for the same nominal dust concentration [30].

For high dust concentrations, the analyses of the obtained results have shown that the maximum explosion pressure is relatively independent from the methane presence. Maximum explosion pressure is imposed by the oxygen content and is not affected by the increase of volatile content promoted by the methane addition. However, when it concerns the maximum rate of pressure rise, it was observed that only when the methane content is increased to 3.5% (v/v), the explosion behaviour is strongly affected, leading to an increase of this maximum rate of pressure rise. This was attributed to the increase of the burning rate resulting from the new conditions of equilibrium for the kinetic reaction. Examples of such explosion behaviour can be found in Pilão et al. [30].

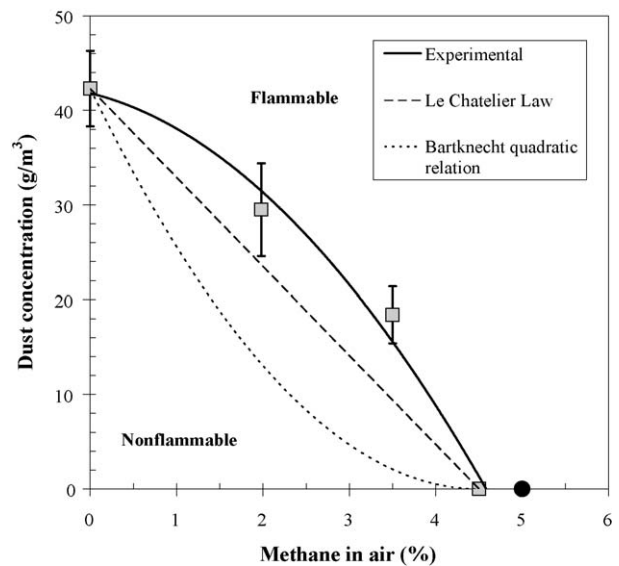


Fig. 16. Effect of methane content in the atmosphere upon nominal minimum explosible cork dust concentration.

In Fig. 16 are presented the obtained minimum explosible concentrations for the two hybrid systems studied together, with the corresponding values for the simple dust air and methane air systems.

In this graphic, the standard accepted lower flammability limit of methane 5% (v/v) is represented by the black circle. This value was not used for the calculation of the trendline shown in the same graph because, as stated by Hertzberg et al. [5] and Cashdollar et al. [25], when using a semi-spherical 20 L reactor with 2500 J of ignition energy, the minimum explosion concentration for the methane/air system is 4.4% (v/v), instead of the above mentioned value of 5%.

Fig. 16 shows that cork dust/methane/air mixtures, for concentrations lower than the corresponding limit concentrations, either for cork dust/air or methane/air mixtures, can form explosible mixtures. The data represented in this figure do not adjust to the quadratic relation proposed by Bartknecht [15] for some hybrid mixtures, and also show a significant disagreement from the linear relation indicated by the Le Chatelier's Law [31].

The increase of the dust concentration for a given amount of methane means that more combustible is necessary to inflame the system than predicted by the Le Chatelier's Law [30]. One explanation for the increase of the dust concentration, for a given amount of methane, evidenced by the Fig. 16, can be related with the possible increase of the mixture limit flame temperature [13]. The Le Chatelier's Law is valid under the assumption that the necessary energy to heat up the medium to the ignition temperature is constant and in the hybrid mixture considered in this study, this energy strongly depends on the concentration of the cork dust. A second explanation is that the curvature in Fig. 16 is caused by differences in the ignitability of the dust with various amounts of added methane. Another possibility is that a small amount of the dust is not effectively dispersed into the chamber, but instead is lost as a coating on the walls of the chamber [30], as is indicated by the lower combustion efficiency experimen-

tally obtained (of the order of 67%) during the explosion of low dust concentrations, as seen in Fig. 9.

3.3. Ignition of cork dust in air

The study of the ignitability of cork dust/air mixtures comprises the experimental determination of their minimum autoignition temperatures.

In the thermal autoignition studies, four different particles sizes, with mass median diameters of 47.4 μm, 71.3 μm, 127.9 μm and 222.1 μm, were tested. The ignition of each one of these fractions was studied as function of initial temperature, while dust concentrations were raised to values up to 300 g/m<sup>3</sup>.

From the experimental traces of pressure evolution, the tests which can be considered with positive ignition could be identified and distinguished from those considered with no ignition. The criterion for ignition in the thermal ignitability tests is both, the rupture of the diaphragm (at 0.1–0.3 atm overpressure) and the emission of flame from the top of the furnace. The diaphragm is a bursting disc of glass microfiber filter used to seal the top portion of the furnace [11].

The thermal ignitability of cork dust for two of the particle sizes studied is shown in Fig. 17. The tests resulting in ignitions are represented with black symbols, while those considered with non-ignition are represented with white symbols. The solid curves are the temperature boundaries between the upper region where the cork dust cloud will thermally self-ignite, and the lower region where the dust may be flammable but does not thermally self-ignite. The lowest point of the curves is the minimum autoignition temperature and for the examples shown, values of 540 °C and 690 °C were found for particles with 47.4 μm and 222.1 μm mass median diameters. Similar autoignition behaviour was observed for all the tested particle sizes. The autoignition temperature is high for small dust concentrations, but it decreases with the rise of dust concentration until a stable value is reached. It was observed an upward trend of the ignition curves, with the increase of the particle size.

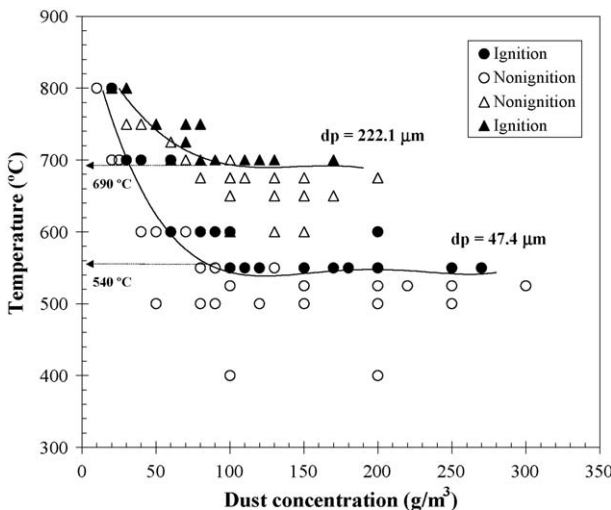


Fig. 17. Thermal ignitability of cork dust.

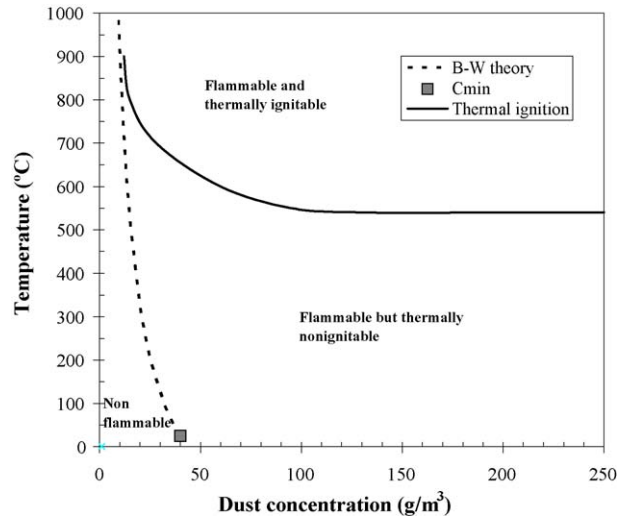


Fig. 18. Domains of thermal ignitability of cork dust.

An abridgment of the effect of temperature on the ignitability and explosibility of cork dust is shown in Fig. 18. The dotted line is the flammability curve and it is obtained by extrapolating to higher temperatures the evolution of the minimum explosion dust concentration, using the modified Burgess-Wheeler theory (B-W) for hydrocarbons [11]. All the system states represented by points under the B-W curve are nonexplosible, because the inflammation probability is zero. The straight line is the autoignition curve obtained with the smallest tested particle size (47.4 μm). Between the two curves, the explosion probability is equal to the forced ignition probability. In this region, the explosion depends on the presence of an external ignition source capable of initiating a potential combustion wave. Above the autoignition curve the explosion is certain, because the probabilities of inflammation and ignition are unitary.

A summary of the effect of particle size on the minimum autoignition temperature for cork dust is shown in Fig. 19. The minimum autoignition temperature is independent of particle

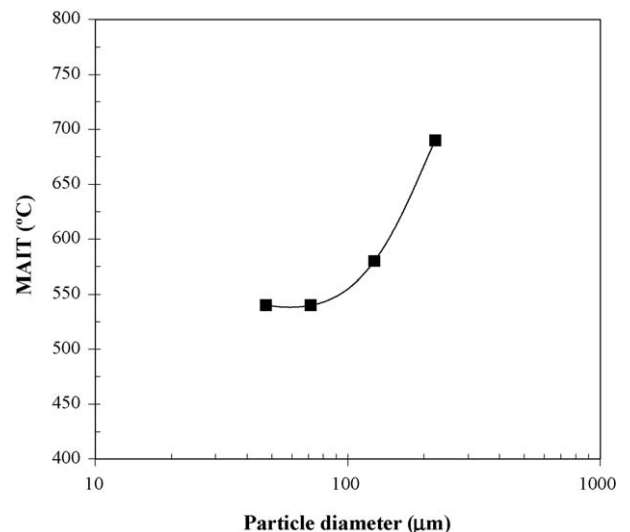


Fig. 19. Effect of particle size on the minimum autoignition temperature.

size for particles with diameters below 80  $\mu\text{m}$ , and it has the value of 540 °C. Below this characteristic diameter, the combustion process during the explosion is controlled by the chemical reaction rate and the diffusion process rate in the gas phase. For larger particle sizes the rate of flame propagation through the mixtures is controlled by the devolatilization rate [20].

#### 4. Conclusions

Although being for long time, a limited field of interest and research studies, the characterization of the mechanisms of initiation, development and propagation of explosions in air and cork dust mixtures has been recently the subject of detailed work [18,23,26,30,32].

The explosion of cork dust follows the propagation flame mechanism proposed by Hertzberg et al. [5]. The combustion velocity controls the combustion process for the smaller particles and lower dust concentrations, whereas the devolatilization rate controls the combustion process for the larger particles and higher dust concentrations.

Initial pressure has an important effect on the explosibility of cork dust. The maximum explosion pressure, the maximum rate of pressure rise and the minimum dust explosion concentration, increase in direct proportion with the increase of the initial pressure, leading to more violent explosions.

The obtained results reveal that 2500 J was the most convenient ignition energy to use in the experimental determination of minimum cork dust explosion concentration, using the semi-spherical 20 L reactor.

The hazard and explosion danger in hybrid mixtures of methane/air/cork dust rises with the increase of methane concentration, characterized by the reduction of the minimum dust explosion concentration.

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