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Quasi-static dispersion of dusts for the determination of lower explosion limits of hybrid mixtures

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Abstract

Knowledge of explosion limiting concentration of explosible materials is necessary for the design of explosion protection measures. Currently employed methods of testing MEC of a dust cloud or LEL of a hybrid mixture are based on arbitrary assumptions and possess technical limitations that often lead to values of MEC/LEL, which are unrealistically low or poorly reproducible.

This contribution presents an improved method for experimental determination of MEC of a combustible dust cloud or LEL of a flammable gas or hybrid mixture. The new set-up operates under laminar conditions and allows a uniform suspension of dust particles in an open top acrylic glass tube. Dust concentration is measured with the help of infrared sensors installed a few centimeters above and below the ignition source.

In order to evaluate the dependence of MEC on flow front velocity, MEC of lycopodium was determined at four flow velocities. The results show that the flow field intensity does not significantly influence the MEC of lycopodium for the flow ranges tested in this work. Moreover, LEL of hybrid mixtures of lycopodium and methane was also tested at flow velocities of 4.7 cm/s, 5.8 cm/s, 7 cm/s and 11 cm/s and compared with the values obtained from other sources. The results suggest that the requirement of high energy pyrotechnical igniter may be relinquished, provided that a truly homogeneous suspension of dust particles could be achieved.

Moreover, the effect of relative amount of dust and gas, on the course of ignition and flame propagation in hybrid mixtures at their LEL, was studied by the help of high speed videos. For hybrid mixtures of carbonaceous dusts (like lycopodium) at their LEL, ignition occurs in the gas phase, however, flame propagation is only possible through a two-way interaction of dust and gas during the course of combustion.

Keywords: *Lower Explosion Limits, Hybrid mixtures, Flame propagation*

1. Introduction

Protection systems against dust, gas or hybrid mixture explosions are commonly designed based on characteristic data, which describe the explosion sensitivity and explosion severity of the material under consideration. One of these characteristic parameters is the explosion limiting concentration of the combustible material, referred to as minimum explosible concentration (MEC) for particulate materials and lower explosion limit (LEL) in case the combustible material is a gas or a mixture of dust and gas, also known as hybrid mixtures.

According to the EN standard, MEC of a dust cloud is measured in standard 1 m³ sphere. Annex C of DIN EN 14034-3 (2011) quotes “an alternative type of test equipment, for which the conformity has been proven, is the 20 l sphere”. Over past few decades, the use of 20-l sphere has gained more

popularity than the 1 m³ vessel because of its cost effectiveness and relatively easier and quicker operation. However, the wide use of 20-l sphere is accompanied by a number of sources of uncertainties related to device configurations, experimental procedure and data treatment protocol, which lead to values of MEC or LEL that are either too low or poorly reproducible (Babrauskas and Britton, 2018). A classic example in this regard is the MEC of corn starch in air, with the reported values ranging from 8 g/m³ to 400 g/m³ (Eckhoff, 2003; Makris, 1988). This contribution analyses the sources of errors that lead to the discrepancies in the experimentally determined MEC of a dust cloud or LEL of a hybrid mixture and presents an improved method for their determination.

DIN EN 14034-3, 2011 defines MEC of a dust cloud as the lowest concentration of dust (or other combustible/flammable material) in mixture with air at which an explosion will occur. The same standard in Annex C.4 states that the highest concentration of a combustible dust at which no explosion occurs in three consecutive tests shall be taken as the lower explosion limit. Moreover, the test procedure is designed in a way that the highest non explosible concentration is always 50% of the lowest explosible concentration. This means that if, for a given dust, there was an explosion recorded at 125 g/m³ and a no-explosion at 60 g/m³ (i.e. ≈50% of the preceding concentration) in three consecutive tests, according to the standard protocol the reported LEL/MEC would be 60 g/m³. Best guess of the true LEL/MEC for this particular dust, however, would be an arithmetic mean of 125 g/m³ and 60 g/m³, yielding ≈90 g/m³ as MEC value. This induces an apparent uncertainty of 50% of the reported MEC value, which does not agree with the information included in the test report stating that the results might deviate up to 10% (DIN EN 14034-3, 2011).

As part of the standard reporting protocol, particle size distribution of the tested dust shall be measured before the commencement of experiments and shall be included in the test report. It is globally agreed upon that the MEC of a dust cloud varies with changes in the particle size distribution of the tested powder (Amyotte, 2013; Baker and Tang, 1991; Bartknecht, 1989; Dufaud et al., 2012; Eckhoff, 2003; Ogle, 2016). Several studies have been performed to investigate the impact of high pressure air blast dispersion, during the explosion experiments, on the particle integrity. The work of Bagaria et al., Du et al. and Kalejaiye et al. is of prime importance in this regard (Bagaria et al., 2017; Du et al., 2015; Kalejaiye et al., 2010). Kalejaiye et al. (2010) investigated the effect of dust dispersion system on particle breakage in 20-l sphere by measuring particle size distribution before and after dispersion. They performed a total of 540 tests at different nominal concentrations of three dusts, namely, Gilsonite, purple K and Pittsburgh coal using the rebound and perforated annular dispersion nozzles and reported that the size after dispersion was about 50%, 80% and 40% of the original size for Gilsonite, purple K and Pittsburgh coal dust respectively. They attributed the particle size reduction to the unique design of the outlet (dispersion) valve and dispersion nozzle, which impose a strong shearing effect on the dust particles as they flow through. The effect of dust concentration on the extent of particle size reduction was found to be negligible. Bagaria et al. (2017) and Du et al. (2015) reported similar data in terms of particle size reduction during dispersion, however, contrary to the findings of Kalejaiye et al. (2010), they observed that there was a decrease in the particle breakage with increasing dust concentration, owing to the decreased probability of impact per particle. This suggests a maximum particle breakage at the concentrations near the MEC of the dust. Concluding this discussion, it could be attested that the specimen tested in the 20-l sphere has a notably smaller diameter distribution than the original specimen, which consequently results in overly conservative values of MEC.

One fundamental flaw in the design of the 20-l sphere test procedure is the use of pyrotechnical igniters. DIN EN 14034-3 (2011) recommends using two chemical igniters each having an energy of 1 kJ, whereas (ASTM 2014) advises measuring the MEC at 2.5 and 5 kJ. LEL/MEC, by definition is the lowest concentration at which a self-sustained flame propagation is achieved. The activation of pyrotechnical igniters, irrespective of their energy content, creates a fireball of burning particles that is way bigger than the size of the sphere and practically covers the whole available space (Taveau et al., 2017). A self-sustained flame propagation through dust air mixture cannot be achieved under these conditions and, as a consequence, each test is rather an igniter induced flame propagation.

Other sources of unreliability of the reported MEC values of dusts include errors due to database abridgment and differences in the definition of an explosion or ignition (visual flame propagation or a certain pressure rise), dust cloud non uniformity and the consideration of nominal concentration, and constant ignition delay time for all types of dusts and highly fluctuating turbulence levels, which are discussed in detail by Babrauskas and Britton in their recent work (Babrauskas and Britton, 2018). Sources of discrepancies between data of a same compound can also be found in the variations of the moisture content, the standard deviation of the particle size distribution or the powder shape factor.

For hybrid mixture LEL testing, no standard device or protocol has been defined so far. Most common experimental apparatus used for this purpose is 20-l sphere, following same protocol as for the MEC determination with an addition of introduction of flammable gas in the system. This is usually done by partial pressure method. Results reported in literature for the LEL of hybrid mixtures also possess inconsistencies (Cloney, Ripley et al. 2018). An example is useful.

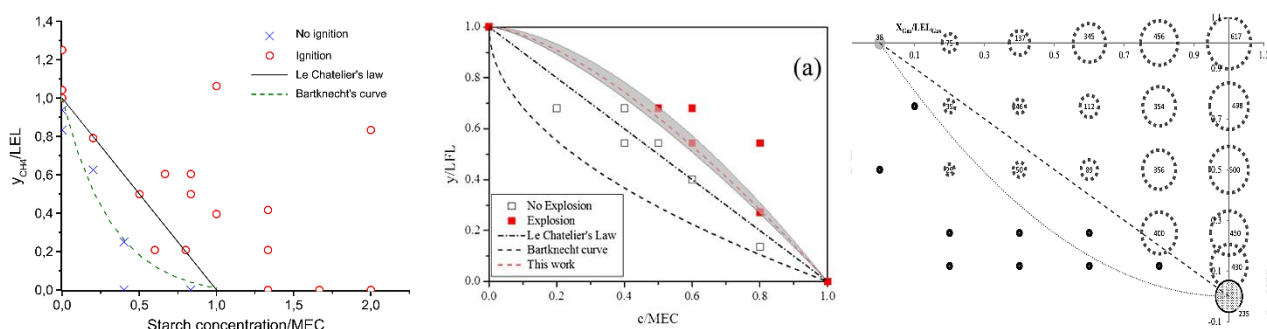


Fig. 1: LEL of hybrid mixture of starch and methane from left to right (Khalili et al., 2012), (Jiang et al., 2014) and (Addai et al., 2015a)

Hybrid mixture of starch and methane is one of the most commonly tested hybrid mixture. Figure 1 provides a comparison of decrease in starch concentration in hybrid mixture with addition of small amounts of methane, measured experimentally at three laboratories. Although the difference in the median particle size (d_{50}) of all three starch samples lied in a narrow range of $\pm 6 \mu\text{m}$, conflicting conclusions were deduced. Jiang et al. (2014) claimed that amount of dust required to render the hybrid mixture of starch and methane explosible is higher than the one predicted by Le Chatelier's law or the Bartknecht's curve. Khalili et al. (2011), on the other hand, highlighted that the explosible concentration of dust in a hybrid system is less, in comparison to the theoretically calculated value using Le Chatelier's law, however, more than the concentration of dust predicted by Bartknecht's curve. In contrast to the findings of Jiang et al. (2014) and Khalili et al. (2011), Addai et al. (2015a) concluded in their research that hybrid mixture of starch and methane is explosible in the concentration ranges even below the Bartknecht's curve. The reason behind these contradictory findings is the inherent sources of uncertainties or errors that make the results of these experiments stochastic in nature and consequently lead to poor reproducibility.

Taken together, above discussion implicates the need for an improvement in the design of standard experimental apparatus and procedure as well as in the data analysis protocol in order to improve the MEC/LEL data quality and achieve better reproducibility. The focus of this research work is to develop a new and improved method for experimental determination of MEC of a dust cloud and LEL of a hybrid mixture. Achieved experimental results are then to be compared with the experimental as well as theoretical results of other authors, accompanied by a discussion on validation and scientific justification of the presented results. Furthermore, this paper explores the contribution of dust and gas on the ignition step and flame propagation mechanism in hybrid mixtures at their lower explosion limit.

2. Materials and Methods

2.1. Material characterisation

Hybrid mixtures of lycopodium and methane, were selected for the experimental investigations of this work. Moisture content was determined using a thermogravimetric moisture analyser (Satorius® MA100) in compliance with ISO 5071:1996. In order to define the LEL of hybrid mixtures in molar units, average molecular mass lycopodium must be defined. This has been established by analysing the quantitative elemental analysis data, collected in-house by using CS230 (LECO®) for Sulphur and CHN628 (LECO®) for carbon, hydrogen, oxygen and nitrogen, in accordance with ISO 29541 and ISO 19579:2002 respectively. Moreover, heat of combustion or calorific value was systematically determined by employing a combustion calorimeter (IKA®, C 200). The measured values were validated according to DIN 51900, ISO 1928 and ASTM D5865.

Table 1: Properties of lycopodium and methane

Dust sample	Moisture Content	Calorific Value	Elemental analysis (% wt.)					Av. M.wt	Particle Size distribution		
			C	H	O	S	N		d ₃₂ (µm)	d ₅₀ (µm)	d ₉₀ (µm)
Lycopodium	3 %wt.	30.1 MJ/kg	68	9.6	21	0.3	1	76 g/mol	30.3	31.7	37.9
Gas	M.wt (g/mol)	Density (g/m ³)	Heat Capacity (J/mol.K)		Calorific Value (kJ/mol)						
Methane	16	675.2	35.52		891.2						

According to ASTM and European standards, particle size distribution of the dust material must be determined before the commencement of explosion experiments. This was measured by using a multi-wavelength laser diffraction particle size analyser (Beckman Coulter LS 13320 CAMSIZER®) according to ISO 13319:2007. The samples were characterized by the d₃₂, d₅₀ and d₉₀ quantiles of the volumetric distribution as indicated in Table 1. Due to the narrow particle size distribution, the particle segregation during the dust dispersion could be neglected. Dust particles were also examined under scanning electron microscope (SEM), to reveal their surface structure (Figure 2).

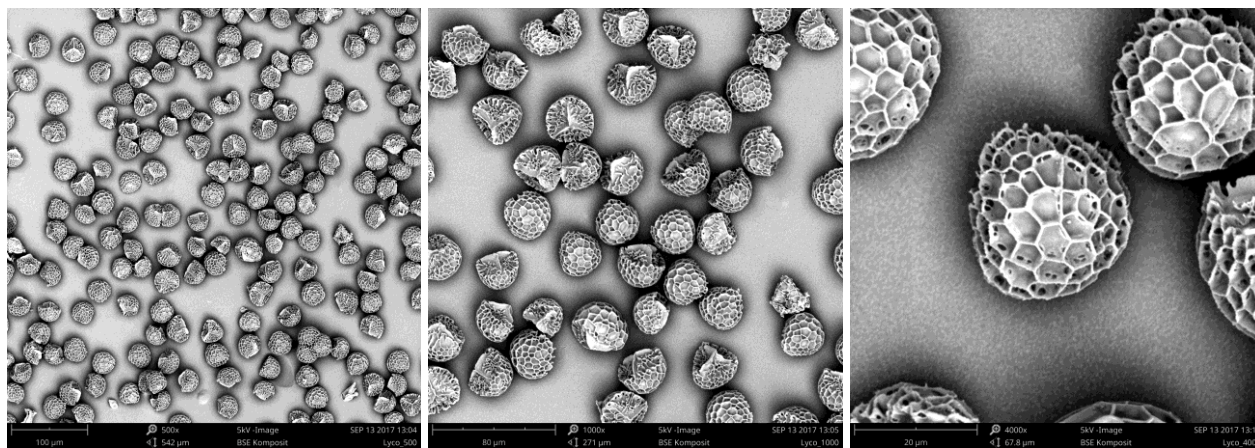


Fig. 2: SEM images of lycopodium

2.2. Experimental setup and procedure

For experimental purposes of this research work, a special stand has been designed and constructed, enabling a reliable measurement of MEC/LEL of dust and hybrid mixtures in a non-turbulent environment. The conceptual design of the experimental device is based on an open-tube apparatus, first introduced by Krause et al., in 1996 to investigate the influence of flow field intensity and turbulence on flame propagation through dust-air mixtures. The set-up consists of a vertically mounted acrylic glass tube of 1000 mm length, with an internal diameter of 60 mm and a wall thickness of 5 mm. The upper end of the tube remained open to allow for venting of gaseous

combustion products and to ensure constant pressure conditions during the reaction. The bottom end of the tube is closed with a filter paper, resting on a sintered glass filter (diameter 70 mm, thickness 5.5 mm and 40-100 μm pore size) and a bed of steel wool. The filter disc together with the steel wool acts as a flow rectifier, facilitating a uniform profile of the flow velocity. The tube is equipped with two brass electrodes, installed at a height of 330 mm from the bottom end and connected to a high voltage transformer, producing a permanent electrical spark of 10J for a duration of 50ms.

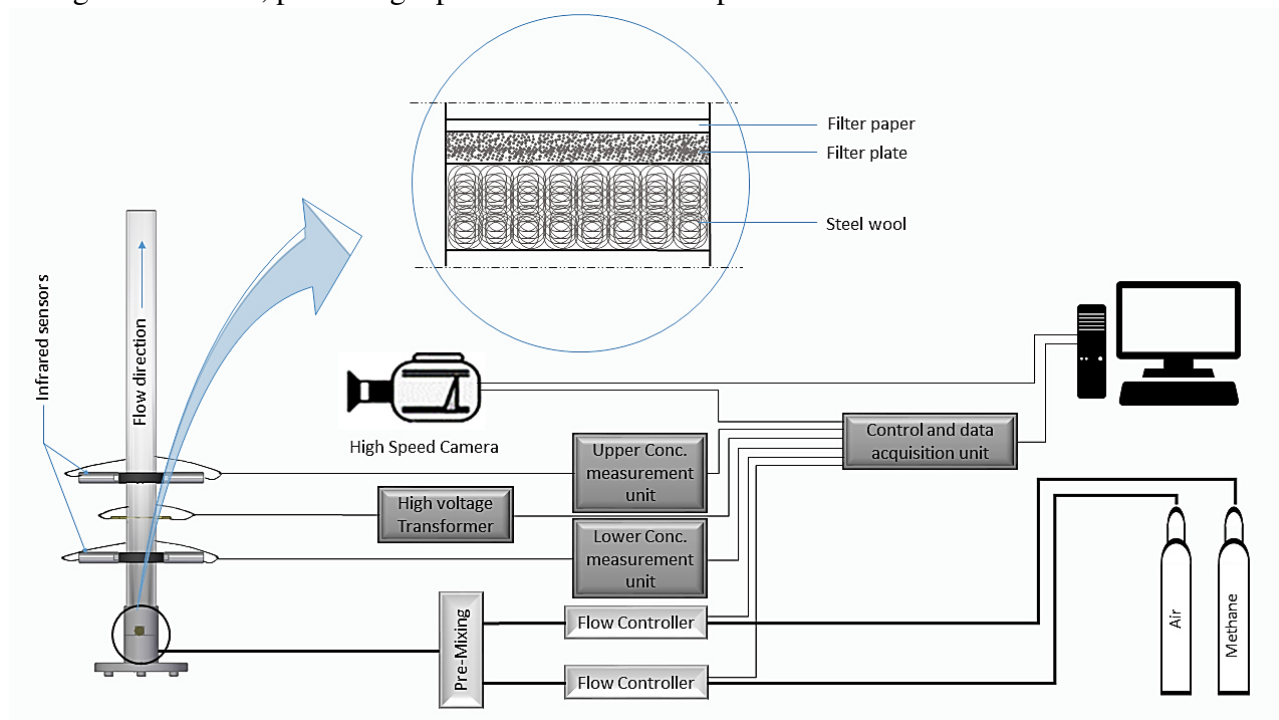


Fig. 3: Schematic diagram of the experimental set-up

The device is equipped with the possibility of reliable in-situ measurement of dust concentration, using infrared light attenuation technique, with the help of two concentration measurement probes, mounted a few centimetres above and below the spark. Flow velocity and composition of flammable gas (in case of hybrid mixtures) was controlled using thermal mass flow controllers. Furthermore, a high speed camera is integrated into the set-up, which enables visual observation of the flame, supporting the ignition or no ignition decision.

In order to ensure good reproducibility of results, a precise protocol was followed throughout the experiments. At the beginning of each experiment, a known mass of dust material was layered on the filter paper at the bottom of the tube. A constant and steady stream of flammable gas-air mixture (for measurement of MEC of dust only air) was inserted from the bottom for a predefined time, depending on the flow velocity, until the flow front reached the top of the tube. At this point, the tube was filled with a homogenous non turbulent column of hybrid mixture, the gas flow was stopped and spark was initiated with a specific delay time, corresponding to the moment when the dust particles are in a quasi-static state, after decelerating under the action gravitational force. Ignition delay time was calculated for different flow velocities assuming that all the particles in the cloud move upwards with a constant velocity equivalent to flow front velocity. Spark energy was controlled by regulating the duration of the spark, keeping the voltage and current at a constant value (5kV and 40mA). An electrode gap of 5 mm was used through the complete course of experiments. High speed camera was triggered automatically upon application of the spark and the concentration value at the time of ignition/no ignition was recorded. Visual flame detachment from the electrodes, as defined by DIN 51649-1, was used as the ignition criteria. In case of an ignition, concentration of dust was decreased by decreasing the inserted mass and the process was repeated, until no ignition was possible for at least five test trials with dust concentrations in the range of $\pm 5 \text{ g/m}^3$ of the average value. The lowest concentration of dust, where ignition occurred, was recorded as the MEC of dust. The experimental

scheme was controlled by a control and data acquisition unit and a software module developed in LabVIEW.

3. Results and discussions

3.1. Concentration reproducibility and MEC of lycopodium

A number of experiments were performed to check the reliability and reproducibility of the concentration data. A known mass of dust was dispersed in the tube and at the time the flow front reached the top end of the tube, concentration values in the upper and lower sensor were recorded. For all the concentration values, where the difference between the readings of two sensors was less than 10% of the average value, were considered valid. The process was repeated 10 times by keeping all the parameters (mass of dust, flow front velocity, flow time) constant and average value was taken as concentration at that particular flow velocity (Figure 4 (a)).

The error bars here present the percentage uncertainty in the concentration values for each flow velocity. Good reproducibility of dust concentration (uncertainty 8.5% - 4.8%) was found for all the four tested flow velocities, with 5.8 cm/s being the most reliable value (uncertainty 4.8%). This is understandable considering that the intensity of flow field has inverse relation with the homogeneity of dust suspension. However, the flow velocity of 4.7 cm/s seems to show the opposite result than what would be expected. An explanation to this could be that the flow velocity of 4.7 cm/s was too low to get all the dust suspended in the tube and as a consequence more mass of dust was required to produce a certain concentration of dust cloud. Because of this added mass of dust, the cloud in the upper part of the tube became denser than the lower part, contributing to a larger difference between the values of upper and lower sensors and hence higher uncertainty.

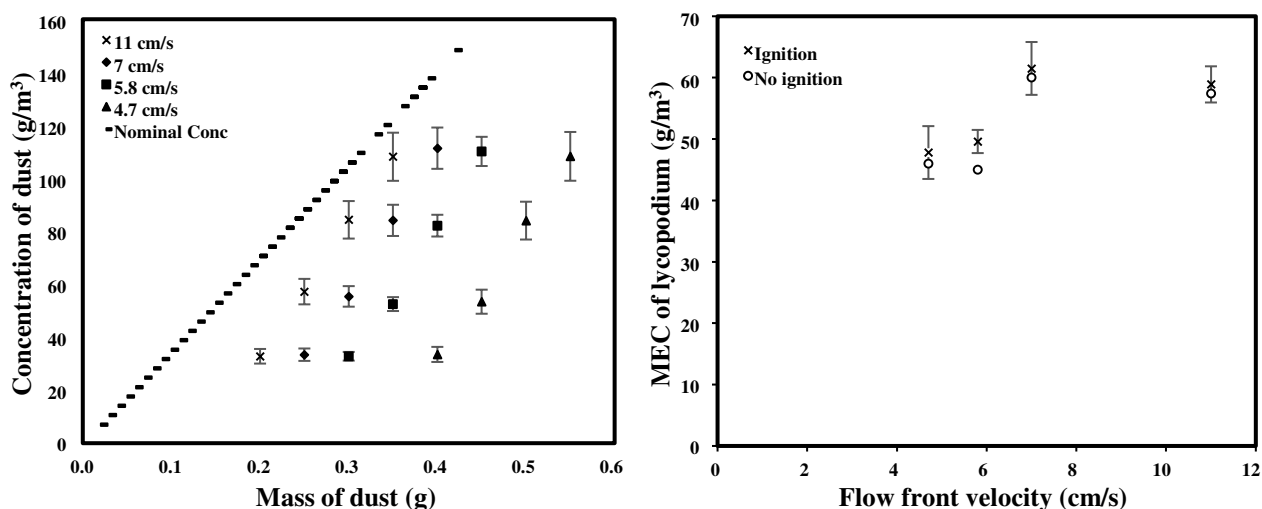


Fig. 4 (a): Reproducibility of dust concentration

(b): MEC of lycopodium

A series of experiments were conducted to examine the dependence of MEC of lycopodium on the intensity of flow field (Figure 4 (b)). Considering the uncertainty involved, it can be stated that the MEC of lycopodium does not significantly change by changing the flow velocity. This statement might not be valid in general, however holds true for the flow velocity range considered for the scope of this paper.

Table 2 shows a comparison of the MEC of lycopodium along with the measurement apparatus, median particle size and elemental composition. MEC value measured with the open tube apparatus seems to be more conservative than the ones measured in 20-l sphere (using permanent spark) and in GG furnace, however, higher than ones quoted in GESTIS-DUST-EX database. The later states that, for obtaining MEC data, a sphere shaped vessel of 20-l or 1 m³ in combination with 2 kJ or 10kJ pyrotechnical igniters was used.

GESTIS-DUST-EX database reports an exceptionally low MEC value of $<15 \text{ g/m}^3$ for lycopodium, although the median particle size is almost the same as the other five source listed in Table 2.

Table 2: Comparison of MEC of lycopodium

Source	Median particle size (μm)	Elemental analysis (% wt.)					Ignition source	Apparatus	MEC (g/m^3)
		C	H	O	N	S			
Addai, 2016	32	69	9.6	19.6	1.3	---	10 J permanent spark	20-1 sphere	125
Addai et al., 2016	32	69	9.6	19.6	1.3	---	Hot surface at 420 °C	GG furnace	108
Sanchirico et al., 2015	32	59	8.1	21.9	2.4	---	10 J permanent spark	20-1 sphere	125
Abbas et al., 2019	31.7	68	9.6	20.7	1	0.3	10 J permanent spark	20-1 sphere	93
GESTIS-DUST-EX	30	---	---	---	---	---	2 kJ/10 kJ chem. igniter	20-1/1- m^3 sphere	<15
Present work	31.7	68	9.6	20.7	1	0.3	10 J permanent spark	Open tube apparatus	49.6

Considering that there are only two organic vapors with MEC values below 30 g/m^3 , it is physically implausible that dust clouds would be more prone to ignition than vapors (Babrauskas and Britton, 2018). Combustion of dust clouds have all of the types of heat losses that occur as with the burning of vapour clouds, but have additional sources of heat losses, e.g. particulate radiation and endothermic pyrolysis, not present for vapors.

3.2. LEL of hybrid mixtures

Before commencement of hybrid mixture explosion experiments, LEL of methane was measured in the 20-1 sphere and the open tube apparatus with their resulting values of 4.6% and 5% respectively. The difference between these values could be attributed to the different ignition criteria. 20-1 sphere operates on the pressure rise criteria which is electronically detected, whereas, in the open tube apparatus the ignition/no ignition decision is made based on the visual observation of the flame. Since methane flame near its lower explosion limit is very weak and nearly impossible to visualize under normal light conditions, a higher value was recorded. It must be mentioned here that also at 5% methane, no visual observation of the flame was possible and ignition was detected by the deposition of a thin layer of steam on the tube wall, which was generated as a result of the combustion reaction.

To have a comprehensive demonstration of the impact of relative amount of mixture constituents on the LEL of hybrid mixtures, results are presented by adopting the commonly used representation proposed by Bartknecht and a recent representation in molar units suggested by the author of this paper (Abbas et al., 2018)

3.2.1. Effect of flow velocity on LEL of hybrid mixtures

Amount of dust required to support a self-propagating flame in hybrid mixtures at their LEL decreases with the addition of a small amount of flammable gas. This decrease tends to become smaller as the concentration of gas increases in the mixture (Figure 5(a)). The maximum decrease in the dust concentration was observed, when the gas concentration was in the range of 0 - 20% of its LEL (i.e. $\leq 1\% \text{ vol.}$). This could be because, the presence of small amount of flammable gas together with the pyrolysis gases (produced by the dust particles that are caught in the spark) increases the local gas concentration and temperature near the ignition source to the level that a self-supporting flame propagation becomes possible (See section 3.3). However, if a small amount of lycopodium (approx. $<10 \text{ g/m}^3$) was added to methane at its LEL, the amount of gas required for combustion to prevail through the system did not decrease. The reason behind could be that at low dust concentration,

particles are relatively more distant from each other and the hot pyrolysis gases, released from the particles caught in the spark, are not in sufficient amount to either increase the local gas concentration or decrease the LEL of local gas cloud (near the ignition source) by increasing the temperature of the gas mixture.

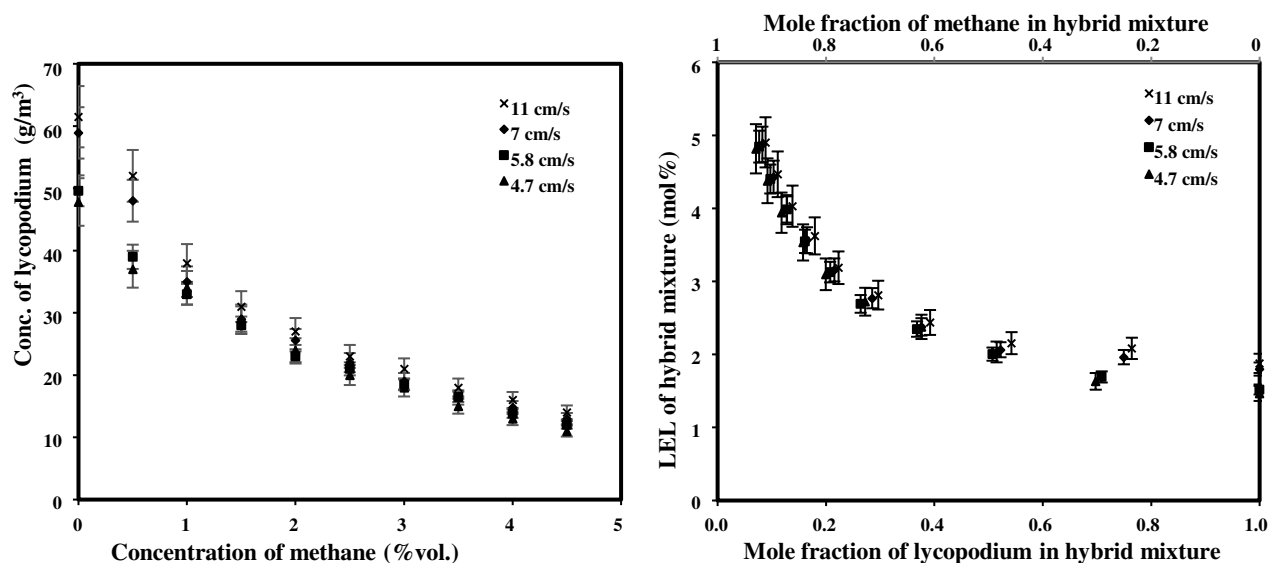


Fig. 5: Dependence of flow velocity on LEL of hybrid mixture of lycopodium and methane a) mass per unit volume units b) molar units

The effect of flow field intensity on the LEL of hybrid mixtures was investigated at four flow front velocities (Figure 5). Considering the uncertainty associated with the experimental values, it can be deduced that LEL of hybrid mixture of lycopodium and methane does not change significantly by changing the flow velocity. An explanation to this behaviour could be the design of the experimental scheme, aimed at igniting the dust particles in a quasi-static state by changing the ignition delay time with respect to the flow front velocity.

3.2.2. Comparison and validation

LEL of hybrid mixtures of lycopodium and methane is compared with previously published results of Abbas et al. and Addai et al., both measured in 20-l sphere using 10 J permanent electrical spark. Moreover, three models for the theoretical evaluation of LEL of hybrid mixtures, namely Le Chatelier's law, Bartknecht's curve and the equation suggested by Abbas et al. (2019), have been plotted. Based on quantifiable errors in the experimental system, an average uncertainty of $\pm 5\%$ (Figure 6 (a)) and $\pm 4.2\%$ (Figure 6 (b)) for the open tube apparatus and $\pm 12\%$ for the results of 20-l sphere (Abbas et al., 2019; Addai et al., 2015b) was considered and consequently added to the respective experimental values. For calculation of theoretical values using Abbas et al.'s equation (2019), a constant temperature rise (ΔT) of 1000 K was used as an ignition criteria for all the hybrid mixtures.

It can be seen at a first glance that the LEL values measured with the new setup are essentially lower and more reliable (lower uncertainty) than the ones determined in 20-l sphere, which operates under very high turbulence, hindering the growth of flame, especially when the concentration of fuel is close to its LEL. High turbulence when coupled with a 10 J electrical spark leads to a higher value of LEL in 20-l sphere. This effect is presumably eliminated by the use of strong pyrotechnical igniters of 2 kJ. However, in reality, the use of strong chemical igniters may cause combustion of dust, even if the concentration is below that required for self-sustained flame propagation at constant pressure, resulting in ultra conservative values of the LEL or MEC (Eckhoff, 2003). The need for highly energetic igniters is supposedly because of the potential of encountering high-energy ignition source in industrial accidents, however, apart from the work of Kauffmann, no information evidently exists on this important question (Eckhoff, 2003; Kauffmann, 1982). Furthermore, in 20-l sphere the local

dust concentration in the region of ignition is not measured, rather it is assumed that the dust particles are homogeneously distributed in the sphere. The concentration value reported as LEL practically refers to the mass of dust equally distributed through the whole volume of the vessel. The effect of turbulence intensity and non-homogeneity of dust distribution is controlled and (supposedly) eliminated by selection of an appropriate ignition delay time, corresponding to the moment, when the dust is equally distributed in the sphere. DIN EN 14034-3, 2011 quotes a constant ignition delay time of 60 ms (20-l sphere), regardless of the size and density of the dust being tested. However, particles with higher density and/or larger particle size (e.g. iron, zirconium) would tend to settle faster than the ones with lower density and/or smaller particle size (lycopodium, polyethylene). These suppositions make the results of 20-l sphere stochastic in nature and quantitatively unrealistic.

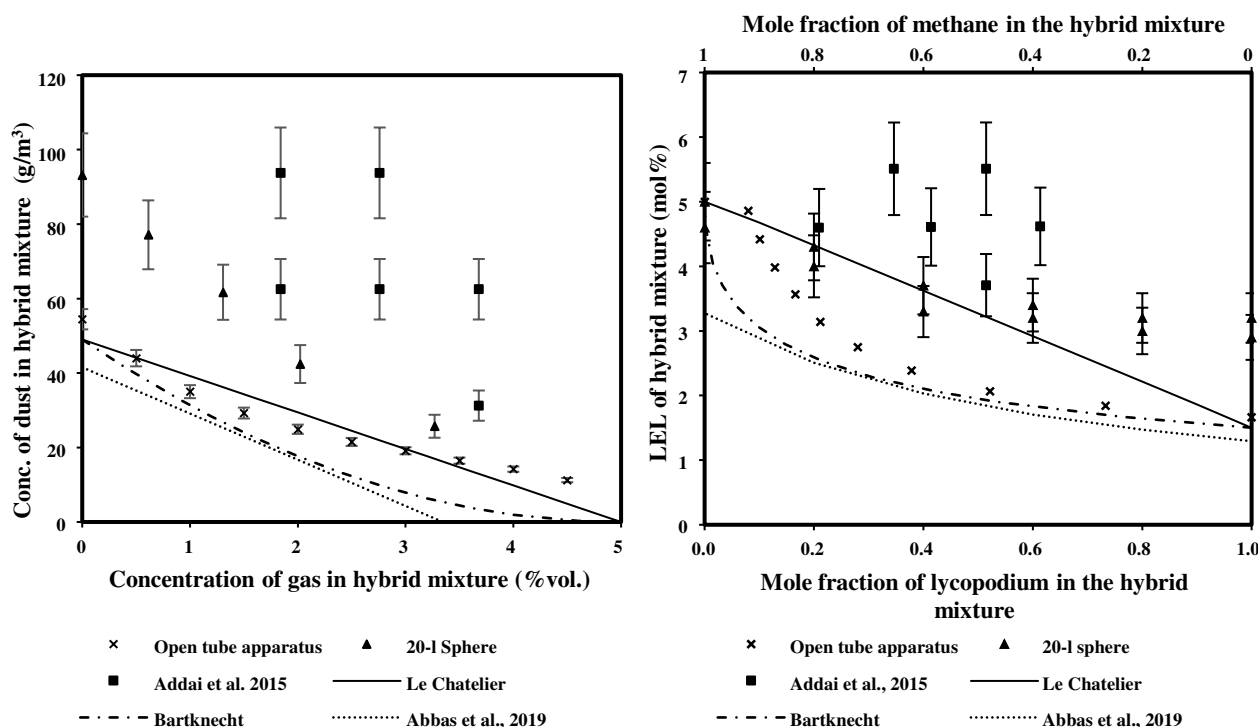


Fig. 6: LEL of hybrid mixture of lycopodium and methane a) mass per unit volume units b) molar units

Basic requirement, as highlighted by Buksowicz and Wolanski (1983) is that “ignition energy must be strong enough to ignite the mixture, but it should not affect the ensuing process of flame propagation. This is especially important at concentration limits, where too strong ignition can support a flame which otherwise would have been extinguished. Furthermore, ignition must be activated at a proper time to match local concentration fluctuations” (Buksowicz and Wolanski, 1983). This prerequisite is achieved in the open tube apparatus by the application of 10 J ignition energy, which is sufficient to ignite the fuel mixture at its LEL without impeding flame propagation.

Summarizing above discussion, one could affirm that the requirement of high energy pyrotechnical igniter could be relinquished, provided that a truly homogeneous and non-turbulent suspension of dust particles is achievable.

3.3. Ignition initiation and flame propagation mechanism in hybrid mixtures

Methane flames at its explosion limits are invisible under normal light conditions. In order to visualize the gas phase combustion, high speed flame/ignition propagation videos were post-processed using an open source image processing package called Fiji. The colours are related to the radiation intensity with red and blue representing maximum and minimum values respectively. Images were extracted from these post-processed videos to investigate the interaction between dust and gas in the hybrid mixture explosion (Figure 7 and 8).

Apart from investigating the LEL of hybrid mixtures, this research work attempts to answer the question, “*what is the contribution of individual components of a hybrid mixture on the course of ignition initiation and the flame propagation, when the concentration of hybrid mixture in air is at its LEL?*”

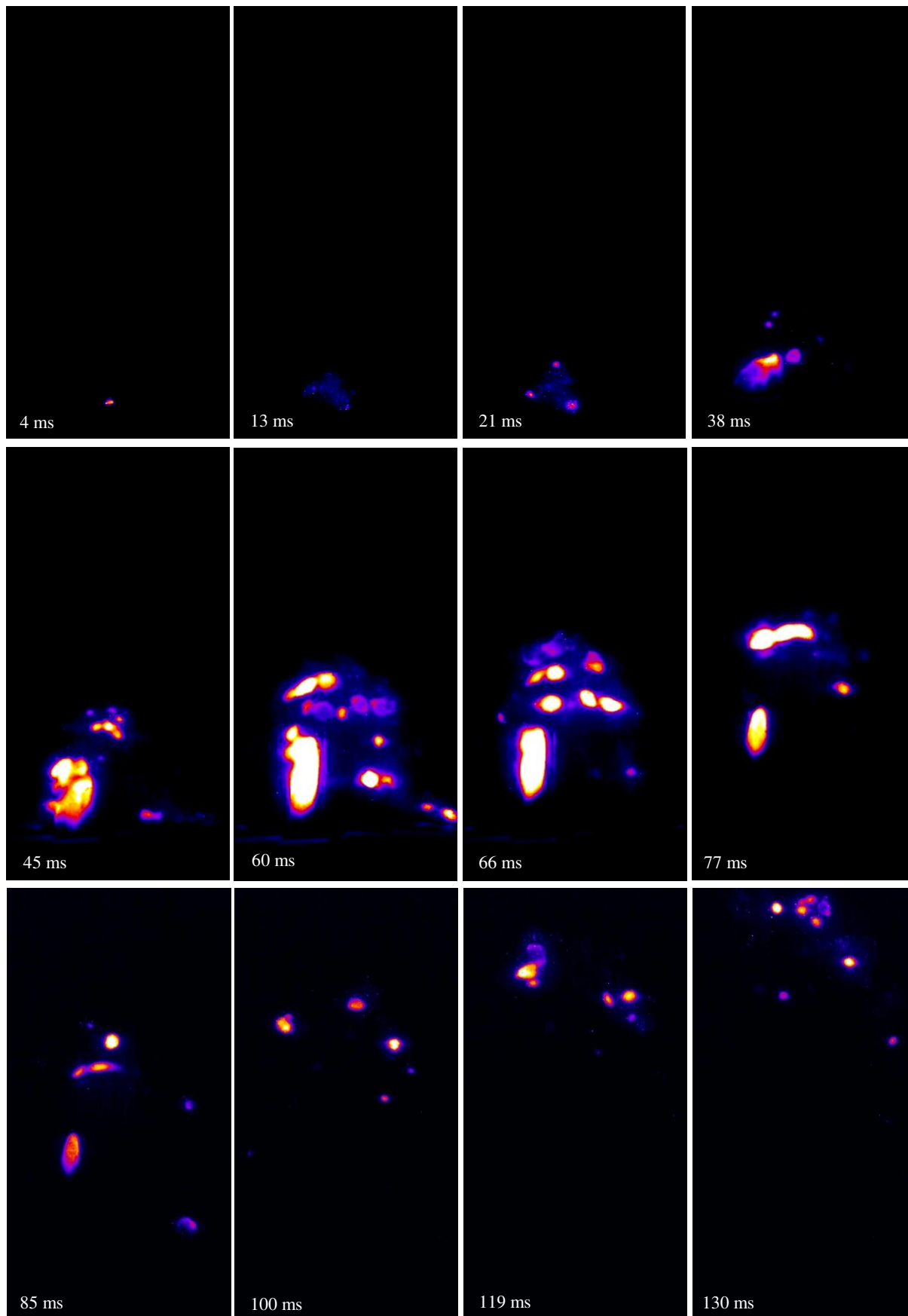


Fig. 7: Ignition initiation and flame propagation in hybrid mixture of 3% methane and 19 g/m³ lycopodium

Ignition initiation and flame propagation in dust clouds may follow one or both of the two types of combustion mechanisms, namely homogeneous and heterogeneous. In general, carbonaceous and higher volatile content dusts undergo homogenous combustion, while metal dusts burn through heterogeneous surface oxidation (Bind et al., 2011; Cloney, Ripley et al. 2017; Di Benedetto et al., 2010; Fumagalli et al., 2016; Kuai et al., 2013).

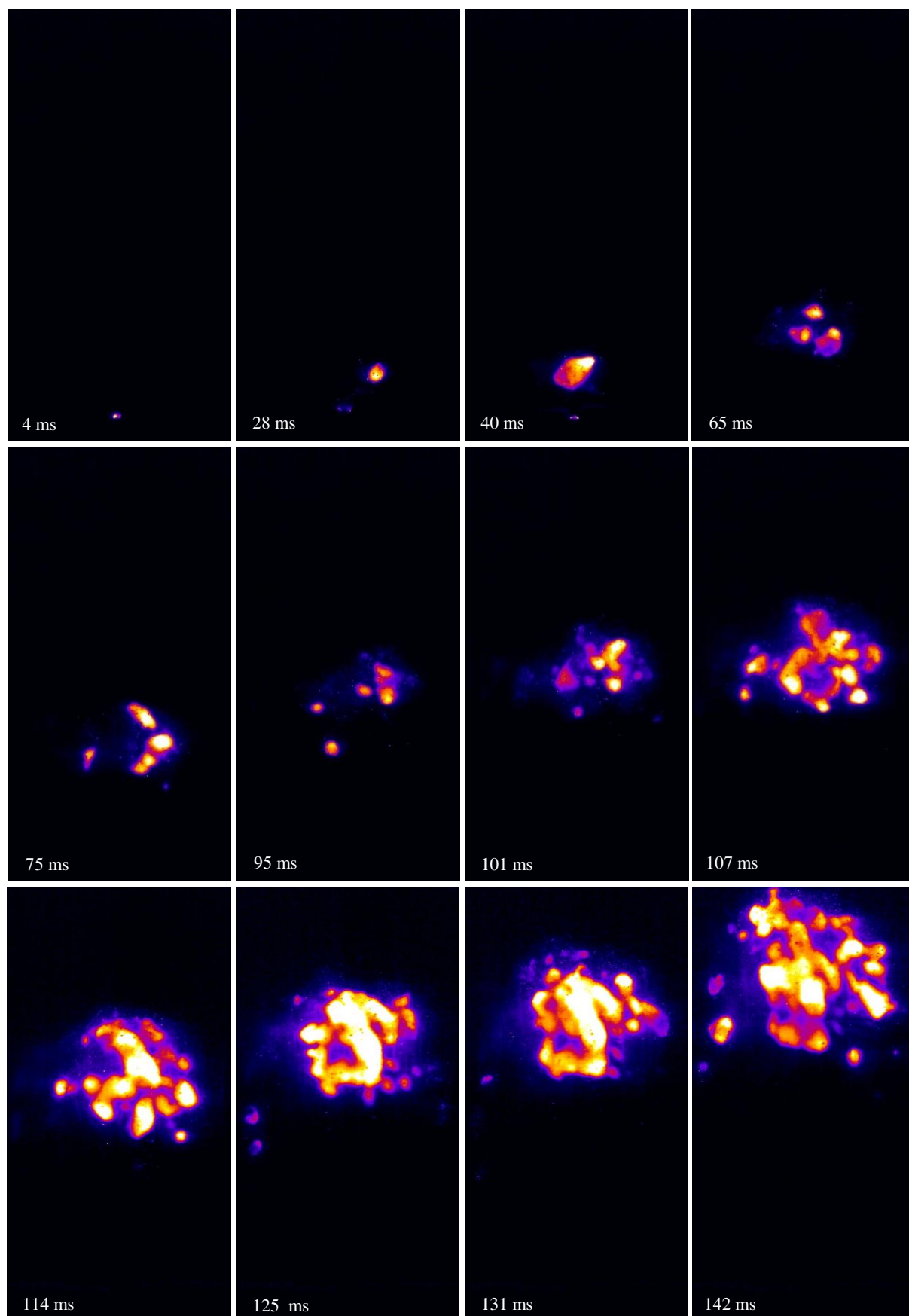


Fig. 8: Ignition initiation and flame propagation in hybrid mixture of 1.5% methane and 30 g/m³ lycopodium

Flame propagation behaviour (in carbonaceous dusts) depends on a number of successive steps during the oxidation process of the combustible fraction. These include devolatilization of the volatiles (also called pyrolysis), mixing of volatiles with air, combustion of volatiles and combustion of the remaining solid fraction. It is evident and has been proven that the combustion of solid particles is mainly controlled by the heat and mass transfer in the reaction zone (Bukowicz and Wolanski, 1983; Eckhoff, 2003; Krause and Kasch, 2000).

Hybrid mixture of a dust and gas/vapour at its LEL refers to a system where the individual concentration of both dust and gas is below their respective MEC/LEL, which implies that there must be two-way interaction between the particles and flammable gas, in order for combustion to prevail through the system. This phenomenon was confirmed within the scope of this investigation (Figure 7 and 8).

In a hybrid mixture at its LEL, combustion starts with the pyrolysis of the particles that are caught in the spark, as is evident from the blue spot in Figure 7 (frame at 13 ms). These pyrolysis gases, in case there is no flammable gas present in the system and the concentration of dust is below its MEC, are not in sufficient amount to ignite the system. Presence of a flammable gas increases the local gas concentration near the ignition source. Since the pyrolysis gases are at higher temperature than ambient, concentrations even lower than the respective LEL of the gas mixture (at ambient conditions) are able to ignite, consequently creating a hot-spot near the igniter. Pyrolysis of the dust particles is accompanied by the particle size reduction. The energy released from this hot-spot causes the heterogeneous combustion of the particles (which already underwent size reduction and are easier to ignite), trapped within the boundaries of the high temperature zone. As a result more energy is released, which heats up the next group of particles releasing more pyrolysis gases in the surrounding and therefore forming new hot-spots. This two way coupling of the radiating particle clusters and burning gas zones enables combustion to prevail through the system.

Regarding the impact of relative amount of dust and gas in hybrid mixtures, it was noticed that increase in the dust concentration lead to a slower combustion propagation in comparison to the mixture compositions where the gas was in relatively higher amount. The term “combustion propagation” has been used here instead of flame propagation because at composition where dust was in relatively less amount, a smooth and continuous flame was not observed rather many discrete burning spot flames. As the fraction of the dust was increased in the fuel mixtures, the gap between these burning fragments decreased and eventually, at sufficiently higher concentration of particles, a proper flame front could be observed.

4. Conclusions

Within the context of experimental determination of MEC of a dust cloud or LEL of a hybrid mixture, a number of sources of discrepancies, inherent to the standard experimental apparatus, protocol and data treatment procedure, have been identified. It has been pointed out that as a combined effect these flaws, some of the reported MEC values are physically implausible, which accentuates the necessity for improvements since compliance with the explosion protection measures based on unrealistically low values of MEC/LEL might be extravagant.

A new and improved method for experimental determination of MEC of combustible dust cloud or LEL of a hybrid mixture has been presented. Instead of the impulse dispersion of a known mass of dust in a fixed volume and assuming homogeneous dispersion, the new set up allows a smooth and uniform suspension of the dust cloud in an open top acrylic glass tube, equipped with the possibility of a reliable in-situ measurement of the dust concentration. Furthermore, a high speed camera is integrated into the setup, which enables visual observation of the flame, supporting the ignition and no ignition decision, instead of relying on an arbitrary pressure rise criterion.

To examine the dependence of MEC on flow field intensity, MEC of lycopodium was measured at four flow front velocities. Within the flow front velocity range tested for the scope of this paper (4.7 cm/s – 11 cm/s), MEC did not change significantly by changing the flow velocity. 49.6 g/m³ was

considered as the MEC of lycopodium and compared with values reported by other authors and the German database GESTIS-DUST-EX, which quotes an exceptionally low MEC value of $<15 \text{ g/m}^3$ for lycopodium, although the median particle size is almost the same as the other five source. Such a low value of MEC for an organic dust is physically implausible since there are only two organic vapours with MEC below 30 g/m^3 .

LEL of hybrid mixtures of lycopodium and methane was also tested at four flow front velocities and the average value was compared with experimental and theoretical values from other sources measure in 20-l sphere. It has been demonstrated that the requirement of high energy pyrotechnical igniter may be abandoned, provided that a truly homogeneous suspension of dust particles could be achieved.

Moreover, the effect relative amount of dust and gas on the course of ignition and flame propagation was studied. For hybrid mixtures of carbonaceous dusts (like lycopodium) at their LEL, ignition occurs in the gas phase, however, flame propagation is only possible through a two-way interaction between the dust and gas.

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References

- Abbas, Z., Gabel, D., Silva, V., Zinke, R., Darbanan, A., Krause, U., 2018. Modelling the Lower Explosion Limit of hybrid mixtures: A thermochemical approach, in: 12th International Symposium on Hazards, Prevention and Mitigation of Industrial Explosions Kansas City, USA. pp. 170–182.
- Abbas, Z., Zinke, R., Gabel, D., Addai, E.K., Darbanan, A.F., Krause, U., 2019. Theoretical evaluation of lower explosion limit of hybrid mixtures. *J. Loss Prev. Process Ind.* 60, 296–302.
- Addai, E., Gabel, D., Krause, U., 2016. Lower Explosion Limit/Minimum Explosible Concentration Testing for Hybrid Mixtures in the Godbert-Greenwald Furnace. *Process Saf. Prog.* 36, 81–94.
- Addai, E.K., 2016. Investigation of Explosion Characteristics of Multiphase Fuel Mixtures with Air Dissertation.
- Addai, E.K., Gabel, D., Krause, U., 2015a. Lower explosion limit of hybrid mixtures of burnable gas and dust. *J. Loss Prev. Process Ind.* 36, 10–14.
- Addai, E.K., Gabel, D., Krause, U., 2015b. Lower explosion limit of hybrid mixtures of burnable gas and dust. *J. Loss Prev. Process Ind.* 36.
- Amyotte, P., 2013. An Introduction to Dust Explosions: Understanding the Myths and Realities of Dust Explosions for a Safer Workplace, An Introduction to Dust Explosions: Understanding the Myths and Realities of Dust Explosions for a Safer Workplace.
- ASTM, E. (2014). "1515-14." Standard Test Method for Minimum Explosible Concentration of Combustible Dusts.
- Babrauskas, V., Britton, L.G., 2018. Errors in the Compilations of Minimum Explosion Concentration Values for Dust Clouds. *Fire Technol.* 54, 37–55.
- Bagaria, P., Zhang, J., Mashuga, C., 2017. Effect of dust dispersion on particle breakage and size distribution in the minimum ignition energy apparatus. *J. Loss Prev. Process Ind.* 1–6.
- Baker, W.E., Tang, M.J., 1991. Gas, dust and hybrid explosions. [https://doi.org/10.1016/0950-4230\(92\)80049-e](https://doi.org/10.1016/0950-4230(92)80049-e)
- Bartknecht, W., 1989. Dust explosions: course, prevention, protection. Springer Science & Business Media.
- Bind, V.K., Roy, S., Rajagopal, C., 2011. CFD modelling of dust explosions: Rapid combustion in a 20L apparatus. *Can. J. Chem. Eng.* 89, 663–670.
- Buksowicz, W., Wolanski, P., 1983. Flame propagation in dust-air mixtures at minimum explosive concentration. *Prog Astronaut Aeronaut* 87, 414–425.
- Cloney, C. T., R. C. Ripley, M. J. Pegg and P. R. Amyotte (2017). "Evaluating regime diagrams for closed volume hybrid explosions." *Journal of Loss Prevention in the Process Industries* 49: 912-918.
- Cloney, C. T., R. C. Ripley, M. J. Pegg, F. Khan and P. R. Amyotte (2018). "Lower flammability limits of hybrid mixtures containing 10 micron coal dust particles and methane gas." *Process Safety and Environmental Protection* 120: 215-226.
- Di Benedetto, A., Russo, P., Amyotte, P., Marchand, N., 2010. Modelling the effect of particle size on dust explosions. *Chem. Eng. Sci.* 65, 772–779.
- DIN 51649-1, 1986. Bestimmung der Explosionsgrenzen von Gasen und Gasgemischen in Luft.
- DIN EN 14034-3, 2011. Bestimmung der Explosionskenngrößen von Staub/Luft-Gemischen.
- Du, B., Huang, W., Liu, L., Zhang, T., Li, H., Ren, Y., Wang, H., 2015. Visualization and analysis of dispersion process of combustible dust in a transparent Siwek 20-L chamber. *J. Loss Prev.*

Process Ind. 33, 213–221.

- Dufaud, O., Khalili, I., Cuervo, N., Olcese, R., Dufour, A., Perrin, L., Laurent, A., 2012. Highlighting the importance of the pyrolysis step on dust explosions. *Chem. Eng. Trans.* 26, 369–374.
- Eckhoff, R.K., 2003. *Dust explosions in the process industries: identification, assessment and control of dust hazards*. Elsevier.
- Fumagalli, A., Derudi, M., Rota, R., Copelli, S., 2016. Estimation of the deflagration index KSt for dust explosions: A review. *J. Loss Prev. Process Ind.* 44, 311–322.
- GESTIS-STAU-EX, n.d. Datenbank mit Brenn- und Explosionskenngrößen von Stäuben, <https://www.staubex.ifa.dguv.de> visited on 20.02.2020.
- Jiang, J., Liu, Y., Mannan, M.S., 2014. A correlation of the lower flammability limit for hybrid mixtures. *J. Loss Prev. Process Ind.* 32, 120–126.
- Kalejaiye, O., Amyotte, P.R., Pegg, M.J., Cashdollar, K.L., 2010. Effectiveness of dust dispersion in the 20-L Siwek chamber. *J. Loss Prev. Process Ind.* 23, 46–59.
- Kauffmann, C., 1982. Agricultural dust explosions in grain handling facilities, in: *International Conference on Fuel-Air Explosions*. University of Waterloo Press, Canada, McGill University 4-6 November 1981, pp. 305–347.
- Khalili, I., Dufaud, O., Poupeau, M., Cuervo-Rodriguez, N., Perrin, L., 2012. Ignition sensitivity of gas-vapor/dust hybrid mixtures. *Powder Technol.* 217, 199–206.
- Krause, U., Kasch, T., 2000. The influence of flow and turbulence on flame propagation through dust-air mixtures. *J. Loss Prev. Process Ind.* 13, 291–298.
- Krause, U., Kasch, T., Gebauer, B., 1996. Velocity and concentration effects on the laminar burning velocity of dust-air mixtures. *Arch. Combust.* 16, 159–176.
- Kuai, N., Huang, W., Du, B., Yuan, J., Li, Z., Gan, Y., Tan, J., 2013. Experiment-based investigations on the effect of ignition energy on dust explosion behaviors. *J. Loss Prev. Process Ind.* 26, 869–877.
- Le Chatelier, H., 1895. Estimation of firedamp by flammability limits. *Ann. Mines* 19, 388–395.
- Makris, A., 1988. Lean flammability limits of dust-air mixtures.
- Ogle, R.A., 2016. *Dust Explosion Dynamics, Dust Explosion Dynamics*.
- Sanchirico, R., Russo, P., Di Sarli, V., Di Benedetto, A., 2015. On the explosion and flammability behavior of mixtures of combustible dusts. *Process Saf. Environ. Prot.* 94, 410–419.
- Taveau, J.R., Going, J.E., Hochgreb, S., Lemkowitz, S.M., Roekaerts, D.J.E.M., 2017. Igniter-induced hybrids in the 20-l sphere. *J. Loss Prev. Process Ind.* 49, 348–356.