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# The influence of dust originating from carbon black nanopowders on the explosion characteristics of lean methane/air mixtures within a turbulent environment



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#### ABSTRACT

Industrial requirements present some unique challenges that can be met only by the application of engineered nanomaterials (ENMs). The completion of risk assessments based on the knowledge of the change in the explosion severity of turbulent gas/air mixtures caused by the accidental dispersion of nanopowders is vital before integrating such materials into existing systems. In this work, known amounts of selected carbon black nanopowders were mixed with methane near the lower-flammability-limit (LFL) to form hybrid mixtures of variable dust concentration. Mixtures were ignited in a 23 L cylindrical combustion vessel that allowed the control of isotropic turbulence through specially designed fans. The particle size distribution, the explosion pressure history and the flame speed derived from high speed Schlieren cine photographs, were measured. The influence of dispersed amounts of nanopowders on explosion severity was investigated by comparing the results with those obtained for pure methane-air explosions. Results indicated that the release of a relatively low mass of nanopowder into methane-air mixtures resulted in a more severe explosion than that of a higher amount. Also, despite the very low content of volatiles in the selected nanopowders, a hybrid mixture can be ignitable at an equivalence ratio well below the LFL of the gas. However this ignitability was shown to be related to the agglomeration state of powder in the dispersion phase, indicating that as the mean particle size decreases, a hybrid mixture with an extremely low content of flammable gas could be ignitable. From a risk assessment point of view, these results may have great significance for specific industrial processes aiming to utilise ENMs.

#### 1. Introduction

A dust explosion may occur as the result of dust particle suspension in air under confinement and the presence of an ignition source. The process industry through its development in time has periodically been stigmatized due to a number of hazardous explosions, some extremely severe (Eckhoff, 2003; Kuriechan, 2005; Vijayaraghavan, 2004). Despite significant research, the risks of dust explosions are still not well understood in industry, and dust explosions continue to occur (Worsfold et al., 2012).

As nanotechnology is a rapidly expanding technology and consequently production and use are very likely to increase in the coming years, a request for global guidance on their environmental and safety risks has emerged (Hansen, 2010). Engineered nanomaterials (ENMs) are widely used in various research and industrial fields (Pritchard,

2004), in the event of a large release of engineered nanoparticles (ENPs) into the atmosphere, an explosive dust cloud could be formed and ignite if the cloud is within the explosive dust concentration range and an ignition source is available. It has been shown experimentally that as the particle size decreases down to the order of  $1-10\,\mu m$ , the explosion severity of a dust cloud tends to increase (Eckhoff, 2012). If this were to be extrapolated into the nanometre range, extreme explosion rates are potentially to be predicted. However, the limited dispersibility of ultrafine particle powders, as well as the tendency in clouds of nano-size particles to coagulate, are two factors that may limit the dust explosion severity at the nanoscale (Eckhoff, 2012).

A hybrid mixture is the combination of a combustible dust and a flammable gas. According to Worsfold et al. (2012), within the form of a hybrid mixture, the dust may be present below its minimum explosion concentration (MEC) and the gas may be present below its lower

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flammability limit (LFL). The combination of both conditions may however create a potentially explosive mixture. Following Amyotte and Eckhoff (2010), explosion parameters such as the maximum explosion pressure ( $P_{max}$ ) and the maximum rate of pressure rise (dP/dt)<sub>max</sub>, have been found to be promoted and to be greater than for either the dust or the gas alone, although the latter has been shown to be more affected. Methane has been found to be less ignitable compared to other hydrocarbons due to the high energy required to break its C-H bonds. However, increased explosion risks have been reported in methane/dust mixtures by a number of studies (Liu et al., 2007; Pilao et al., 2006).

Mixtures of flammable gas and dispersed fractions of solid particles are frequently present in diverse industrial applications and their explosions are still not well understood (Torrado et al., 2016). Several works (Cashdollar, 1996; Garcia et al., 2011; Khalili et al., 2012; Pilao et al., 2006) have studied explosions in hybrid mixtures, although there are still unanswered questions about the phenomena involved. If we consider the average particle size of the dusts tested in these works, then relatively few studies have focused on hybrid explosions involving ENMs. Potential new explosion hazards may be present for processes where ENMs are utilised to enhance the efficiency of established techniques applied in industry.

In a recent study conducted by Abdelsalam et al. (2016), the effect of metallic nanoparticles such as Co, Ni and Fe, on biogas and methane production from anaerobic digestion of livestock manure was investigated. Additionally, Lu et al. (2016) examined the effects induced by the use of specific metallic nanoparticles as additives for promoting the catalytic activity of methane combustion. From these studies it is understood that the danger derived from accidental ignition of a reactive two-phase mixture is not confined only to coal mining industries, but through the increased use of ENMs, it is likely to spread to other industries. A balance must be achieved between increased use for enhanced product performance with safe and predictive handling and storage of these materials.

According to Turkevich et al. (2015), nano-carbons are confirmed mainly to belong to the dust explosion class St-1 which has been determined to characterise weak explosions (Bouillard et al., 2008). Due to the very low content of volatiles, the flame propagation mode of explosions involving carbon black (CB) nanopowders is exclusively described by heat transfer between the particles (Kosinski et al., 2013). So far, only two experimental works (Kosinski et al., 2013; Torrado et al., 2016) have investigated the explosion characteristics of flammable gas/air atmospheres mixed with dispersed amounts of CB nanopowders and they are thus the most relevant with respect to the present work.

Kosinski et al. (2013) focused on explosions of CB nanopowders under the addition of some quantities of propane in a 20 L explosion vessel. The igniters used in this work were a 6 J electric spark discharge and 1 kJ chemical igniters. The results showed that for lean gaseous mixtures, the addition of nanopowders increased the  $P_{\rm max}$  and (dP/dt)\_{\rm max} explosion parameters. Similar trends were observed for rich gaseous mixtures. For the highest nominal dust concentration (500 g/ $\rm m^3$ ) no explosion was recorded, and according to the authors this could be attributed to high deposition of the suspended particles in piping and dispersion nozzles. The scanning electron microscope (SEM) images of the explosion residues revealed the formation of large agglomerates. This agglomeration makes the explosion study more complicated than it would be for individual particles.

Torrado et al. (2016), studied the influence of CB nanopowders (Printex XE2 and Corax N550) in methane-air mixture explosions conducted in a 20 L spherical vessel. They also studied the effect of the initial turbulence by varying the ignition delay time (the time between the start of the air pulse for the injection of powder and the triggering of ignition). In their work, ignition at 100 J was utilised through the use of chemical igniters. The results indicated that when a only few percent of CB was added in lean methane-air mixtures, a slight increase of  $P_{\rm max}$ 

and decrease of (dP/dt)<sub>max</sub>, were observed.

Up to date, the explosion studies on dust clouds generated from the dispersion of ENMs, such as nanopowders, have utilised the standardised methodology for determining explosion severity through the measurement of pressure history in constant-volume bombs. The acquired pressure data has been used for a theoretical calculation of the flame velocity due to the lack of optical access associated with standard spherical vessels, such as the 20 L sphere utilised so far in the study of nanopowders' explosivity. Therefore, it is important to obtain a knowledge of the experimentally measured turbulent flame velocity through optical techniques in gas mixtures containing dispersed amounts of ENMs that will allow a more integrated design of prevention/protection systems in industries promoting their applications.

Furthermore, the influence of dust in hybrid mixtures with a gas below its LFL has been discussed by Dufaud et al. (2009), while several indications that explosions can occur even at these low equivalence ratio ( $\phi$ ) values have been reported by Amyotte et al. (2007), Cashdollar (1996) and Cashdollar (2000). Attempts to ignite hybrid mixtures composed of propane and dispersed amounts of a CB nanopowder below the LFL of gas, have been conducted by Kosinski et al. (2013), however they reported that this ignition was not possible due to the negligible amount of volatiles in the tested nanopowder.

Knowledge of the change in the explosion severity of lean turbulent fuel-air mixtures induced by the dispersion of known amounts of ENMs, is vital for the completion of risk assessments related to processes taking place in industrial environments such as those described above. The objective of the current work is to quantify and gain a better understanding of the effects of dust particle size on the severity and likelihood of hybrid mixtures containing fractions of ENPs originating from specific nanopowders.

To this end, two types of selected CB nanopowders were dispersed in lean methane-air for the formation of hybrid mixtures with variable dust concentration in a cylindrical explosion vessel providing adjustable mechanical turbulence. Explosion tests were conducted under atmospheric conditions and triggered by a spark plug of low nominal ignition energy. The explosion severity was characterised through the measurement of the explosion pressure history and of flame speed derived from high speed Schlieren cine photographs. Also, the particle size characteristics in the explored dust clouds were determined. The influence of the selected nanopowders on explosion severity was investigated by comparison with results obtained for pure methane-air explosions. The explosion potential of hybrid mixtures of specific dust concentration and with methane below its LFL, was also investigated. Finally, for the same hybrid mixtures, the effect of ignition energy on their explosion characteristics was evaluated through the application of an ignitor providing relatively higher ignition energy than that of the spark plug.

#### 2. Material and methods

#### 2.1. Apparatus and methods

The present work was conducted with existing apparatus which has been previously used for studies of gaseous combustion. Fig. 1 shows a schematic of the combustion apparatus. This is described in detail in Atzler (1999), and, Atzler and Lawes (1998). It comprises a cast steel cylinder of 305 mm diameter and 305 mm length and its working volume is 23 L. At each end, optical quality windows constructed from fused silica 150 mm diameter and 39 mm thick are fitted. The apparatus is equipped with four identical eight-bladed fans equally spaced around the central circumferential plane at 45° to the horizontal. The fans generate isotropic turbulence within the field of view of the windows and this has been fully characterized by Lawes (1987). Specifically, the root mean square (rms) turbulent velocity fluctuation u' was found to be a linear function of fan speed according to:

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