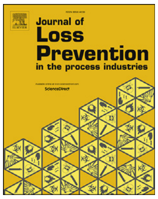
Journal of Loss Prevention in the Process Industries xxx (2017) 1–10

Contents lists available at ScienceDirect

Journal of Loss Prevention in the Process Industries

journal homepage: www.elsevier.com/locate/jlp



Influence of carbon black nanoparticles on the front flame velocity of methane/air explosions

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ARTICLE INFO

Article history:
Received 27 September 2016
Received in revised form
20 January 2017
Accepted 6 February 2017
Available online xxx

Keywords:
Nanoparticles
Hybrid mixtures
Dust explosion
Flame velocity
Laminar burning velocity

ABSTRACT

This work aims to study the influence of low concentrations of carbon black nanoparticles in gas mixtures on the front flame velocity. Due to their low settling velocity, nanoparticles offer the opportunity to study the hybrid mixture explosion at low turbulence levels of dispersion. They can also be used as particles to model the presence of soot. The flame velocity of carbon black nanoparticles/methane/air mixtures was measured in a vertical 1 m long tube with a square crosssection connected to a gas mixing system. Dust clouds are generated by a pulse of methane/air mixture at 5 barg from the bottom of the tube, where the mixture is also ignited. A high-speed video camera is used to record the flame propagation. An estimation of the laminar burning velocity is obtained using the method proposed by Andrews and Bradley. Although this method may not be precise for laminar flame velocity estimations, it offers a first approximation for hybrid systems explosions. The influence of the initial turbulence was also studied by varying the ignition delay. The influence of low concentrations of carbon black nanoparticles on the front flame velocity has been appreciated by comparing the results obtained for gaseous mixtures explosions at different turbulence levels. The burning velocity of gaseous mixture seems to increase when the initial turbulence of the system is augmented. However, when the initial turbulence is significant, the front flame velocity seems to decrease, suggesting that the flame kernel can be strongly destabilized by turbulent vortices. Moreover, it appears that the flame burning velocity can slightly decrease when carbon black nanoparticles concentration is increased. The unstretched burning velocity is decreased by 43% when 20 mg of carbon black nanoparticles are added to the system. This trend could be explained by the enhancement of the heat radiation transfer of the system. The results are then compared to the explosions trends in a 20 L spherical vessel.

1. Introduction

The number of applications of nanoparticles for industrial purposes has grown enormously in the past years. The European Commission suggests that a nanomaterial should be defined as a material which more of 50% of the particles in the number size distribution (in unbound state, in agglomerates or aggregates) have a size between 1 nm–100 nm (European Commission, 2011). Nanoparticles have been commonly used as additives to modify certain properties of materials like hardness, resistance, surface properties, UV-radiation, magnetization or rheology. In addition, due to their size and their greater surface area, nanoparticle has become advantageous for catalysis, biomaterials and biological applications (Stark et al., 2015). Through the quantification and characterization of nanoparticles, some recent studies aim to assess the risks of using them in these types of applications (Schmid and Riediker, 2008). As a consequence, studies of dust explosions involving nano-sized powder are emerging, aiming to provide more data on ignition sensitivity and explosion severity (Amyotte, 2014; Vignes et al., 2009). For instance, Boilard et al. (2013) had compared the explosion properties of micro and nano-sized titanium powders. Nano-titanium is a highly sensitive powder as the dust self-ignition is easily achieved and the explosion severity seems to increase when the particle size decreases (Boilard et al., 2013). Although the explosion overpressure has comparable

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http://dx.doi.org/10.1016/j.jlp.2017.02.006

Please cite this article in press as: Torrado, D., et al., Influence of carbon black nanoparticles on the front flame velocity of methane/air explosions, Journal of Loss Prevention in the Process Industries (2017), http://dx.doi.org/10.1016/j.jlp.2017.02.006

values for micro and nano-sized powders, similar studies of nanodust have shown that the explosion sensitivity is considerably higher the smaller the particle size (Vignes et al., 2009). In fact, the similarities on the explosion severity between microparticle and nanoparticle powders could be explained because of the limited homogeneous dispersibility and the high coagulation phenomena on nanoparticle powders (Eckhoff, 2012, 2013; Worsfold et al., 2012). For very low particle size, the influence of the particles insertion is limited by the powders agglomeration (Bouillard et al., 2010; Mittal, 2014). During the production of nanoparticles and their industrial applications, simultaneous presence of gas/vapor/solvents and dispersed mixtures could be obtained, engendering the risk of a hybrid mixture explosion. Moreover, such mixtures are systematically encountered during the combustion of organic dust, notably on the devolatilization phase leading to a hybrid mixture of pyrolysis gases and char/unburnt powder (Cuervo, 2015).

Hybrid mixture explosions have been particularly studied in the past years due to their different severity and sensitivity variables when compared to pure dust or gas explosions (Dufaud et al., 2009; Khalili et al., 2012). Some empirical correlations for hybrid mixtures have been developed aiming to design protection systems for the industrial production processes (Siwek, 1996); nevertheless, the properties and behavior of these systems must be further researched for a better design of such protection systems. The hybrid mixtures explosions properties had been studied for microdust/gas mixtures such as coal/methane and cork dust/methane (Liu et al., 2007; Pilão et al., 2006), and the dust driven, gas driven, dual-fuel and synergic explosions regimes for those mixtures have been defined (Garcia-Agreda et al., 2011). The hybrid mixtures analyzed in this work are essentially gas driven explosions. The ignition sensitivity of dust can be strongly affected when a small concentration of gas is added (Dufaud et al., 2008) and the maximum rate of explosion of pure compounds is greatly affected by the presence of a small amount of gas or dust (Denkevits, 2007). Regardless the recent efforts to describe the particularities of hybrid explosions, there is limited data for nano-dust/gas hybrid mixture explosions. Kosinski et al. (2013) studied the influence of carbon black nanoparticles concentration on the explosions severity of propane/air mixtures. According to this study, the propane concentration must be higher than the lower explosion limits in order to cause an explosion. Kosinski et al. (2013), however, found that the maximum overpressure was higher for lean gas mixtures when a small concentration of carbon black nanoparticles is added to the system. This trend is similar as the one observed for the maximum rate of pressure rise: an explosion severity higher for low powders concentration but reduced for high carbon black nanoparticles concentrations. These results suggest that heat loss generated by the carbon black nanoparticles modifies the combustion process (Kosinski et al., 2013). Moreover, the study of carbon black nanoparticles/combustible gas systems is also a mean to highlight the influence of soot production on gas combustion. For instance, certain grades of carbon black nanoparticles have been used to understand the behavior of specific types of diesel soot (Growney et al., 2015).

The objective of this work is to study the flame propagation velocity of carbon black nanoparticles/methane/air mixtures. Flammable gaseous mixtures have been widely studied and different methods of measuring flame velocity have been also developed (Andrews and Bradley, 1972). Nevertheless, there is still a need of continued research on powder and hybrid mixtures explosions and flame propagation (Dahoe, 2000). Dahoe et al. (2002) estimated the laminar burning velocity and introduced the Markstein length for stabilized cornstarch-air flames using a laser Doppler anemometry. The laminar burning velocity of powder's explosion was found sensitive to the shape of the flame (Dahoe et al., 2002). The laminar burning velocity of starch, lycopodium and sulfur flour powder have also been studied, showing values with similar order of magnitude of combustion velocity as those obtained from CH_4-air mixtures (Proust, 2006a). However, the flame propagation of hybrid mixtures is not yet entirely understood. Differentiating the laminar burning velocity from the apparent flame velocity of such systems is essential because most of the CFD simulation of the explosions and the protection devices design use this intrinsic property. This becomes a challenge due to the complex thermal transfers, combustion kinetic mechanisms and turbulence/combustion interactions encountered in such mixtures.

2. Experiments

2.1. Materials

In this study, Printex XE2 and Corax N550 (Orion) have been chosen as carbon black nanoparticles. The characteristic diameter of the powder d_{50}, the BET specific surface (using Brunauer-Emmett-Teller method) and the equivalent BET diameter are reported in Table 1. It appears that the primary nanoparticles are arranged in agglomerates of micrometric size. Moreover, Printex XE2 presents a much larger specific surface area than Corax N550, which may impact the respective reactivity and physical properties.

Methane was selected for this study as combustible gas due to its chemical simplicity, and also because the carbon/methane mixtures are often encountered in various industrial applications. In order to study the influence of an initial concentration of soot in a methane/air mixture, low concentrations of carbon black nanoparticles were chosen. Regarding the scale of precision, the lowest concentration of carbon black was set at 2 g m^{-3} (0.40% molar concentration). Tests were also performed at 6 g m^{-3} (1.2% molar concentration), which is still far below the minimum explosive concentration (60 g m^{-3}) of such powders (Bouillard et al., 2010). These concentration values are also chosen in order to ensure a significant concentration of nanoparticles compared to the soot generated during the explosion test.

2.2. Experimental setup and procedures

In order to measure the flame propagation velocity, the explosions of methane/carbon black/air mixtures are generated in a vertical 1 m long tube with a cross section area of 0.07 × 0.07 m connected to a gas mixture system (Fig. 1). The tube is made of two opposite glass walls and two opposite stainless steel walls (Cuervo, 2015; Di Benedetto et al., 2011). The bottom of the tube is closed, while the top is partially open using a pressure relief valve, which opens at 1.15 atm in order to ensure a constant pressure. At the closed bottom, a mushroom-shaped nozzle is placed to disperse the carbon black dust homogenously. The mixture is ignited using an electrical spark equivalent to 10 J which is rather high compared to methane MIE (0.3 mJ). However, the addition of carbon black tends to increase the MIE of the mixture; hence, greater ignition energy has been used for all the test of this work.

The gas mixture system consists of a 0.05 L vessel equipped with valves that allow a manual control of the gas pressure. The gas mixtures are injected in two different places of the tube to ensure homogeneity of the gas concentration across the tube (proven by gas chromatography measurements by using a Varian 490 MGC). A first injection of the gas is made at a 0.50 m height, and then the 5.2 bar gas mixture is used to generate the dust dispersion at the bottom of the tube. The initial turbulence level is studied by changing the ignition delay (tv) between the beginning of the

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