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Explosions of methane/air/nanoparticles mixtures: Comparison between carbon black and inert particles

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ABSTRACT

During incomplete combustions or nano-size carbon blacks generation, atmospheres of carbonaceous nanopowders and combustible gases are encountered. These hybrid mixtures exhibit specific explosive behaviors, which can notably be caused by the modification of the initial turbulence level or by changes in oxidation reactions. In order to either support or reject such assertions, various nanoparticles/methane mixtures were tested, some with carbonaceous nanopowders, some with inert nanopowders (alumina). The aim of this work is then to compare the influences of alumina and carbon black nanoparticles insertion on the explosion severity and on the flame velocity of methane. Tests were performed in a 20 L explosion sphere and in a 1 m vertical flame propagation tube. An estimation of the unstretched flame velocity is obtained assuming a linear relationship between the burning velocity and Karlovitz stretch factor. It appears that the use of carbon black nanoparticles increases the explosion overpressure for lean methane mixtures by approximately 10%. Similar behaviors have been observed for hybrid mixtures involving alumina particles for fuel lean conditions. For alumina, non-significant changes are observed for fuel rich mixtures. Moreover, a considerable diminution of the explosion severity was noted for fuel rich mixtures when carbon black nanoparticles are dispersed into the reaction vessel. Regarding the flame propagation test for stoichiometric methane concentration, higher unstretched burning velocities were obtained for carbon black hybrid mixtures compared to alumina mixtures. These results suggest soot or carbonaceous nanopowders not only impact the oxidation kinetics, but also the flame stretching and heat transfer.

1. Introduction

A hybrid mixture consists of an explosive mixture of a flammable gas and combustible dust, in which the gas may be present below its lower flammability limit (LEL) and the powder may be below its minimum explosive concentration (MEC). Despite increased knowledge regarding this subject and efforts that have been made to propose accurate and efficient solutions in order to prevent this kind of events, hybrid mixture explosions still occur in different types of industries (e.g. food, mining, pharmaceuticals, chemicals). Some examples of hybrid mixtures are coal/gas mixtures in underground mining, natural gas/fly ash in power plants or hydrocarbons/resins in plastics manufacturing (Amyotte et al., 2009; Amyotte and Eckhoff, 2010). Obviously, developing academic knowledge is not sufficient and the joint implementation of a safety culture is also compulsory. Additionally, technical causes should also be considered to explain this occurrence: more and more

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Nomenclature

Symbols

A	Tube cross-section [cm^2]
A_f	Front flame surface [cm^2]
K	Karlovitz's factor [s^{-1}]
L	Markstein length [cm]
S_s	Spatial velocity [cm s^{-1}]
S_u	Burning velocity [cm s^{-1}]
S_u^0	Unstretched burning velocity [cm s^{-1}]

Greek letters

α	Thermal expansion correction [–]
γ	Heat capacity ratio of the gas [–]

processes involve the simultaneous presence of combustible gas and dusts, as is the case of composting or methanisation facilities, biofuels plants, or during the generation of some carbonaceous nanoparticles. As a consequence, further studies of the explosive properties of such atmospheres must be conducted (Amyotte, 2014; Amyotte and Eckhoff, 2010; Jiang et al., 2014; Worsfold et al., 2012).

Hybrid mixture explosions have been particularly studied in the past few years due to their different severity and sensitivity variables when compared to those of pure dust or gas explosions (Denkevits, 2007; Dufaud et al., 2008; Khalili et al., 2012). The minimum ignition energy (MIE) of dust clouds decreases when low concentrations of combustible gas are added to the combustible dusts and, regarding pure dust explosions, the likelihood of the explosion increases even for gas concentrations below the explosive limits (Addai et al., 2016; Khalili et al., 2012). Different simple mathematical relations have been proposed to estimate the lower explosive limit of a hybrid mixture (LEL_{Hybrid})—e.g. Le Chatelier or Barknecht equations—but certain studies have also demonstrated that for some hybrid mixtures such calculated value is not always the safer estimation (Addai et al., 2015; Khalili et al., 2012). Prugh (2008) has proposed a correction of the LEL for hybrid mixtures by taking into account that its estimation is usually made at room temperature, whereas it must be recalculated at the flash point temperature. Another formula to estimate the lower flammability of the mixture has been proposed and validated by Jiang et al. (2015, 2014) using a 36-L explosion apparatus. A good prediction of the non-explosion/explosion zones have been obtained for several hybrid mixtures (Jiang et al., 2015).

When compared to the pure compound explosion parameters, the synergetic effect on the explosion severity of hybrid mixtures seems to strongly depend on the physical or chemical properties of the compounds. Pilão et al. (2006) reported non-significant changes on the maximum overpressure P_{max} and dP/dt_{max} variables of cork dust/methane hybrid mixtures when compared to those of dust explosions, even if a significant reduction in the minimum dust explosion concentration is obtained. Nevertheless, the ignition of niacin/diisopropyl ether mixtures showed a strong influence of low concentration of vapor on the explosion severity, which is greater to that of pure compounds (Dufaud et al., 2009, 2008). Ajrash et al. (2016a, 2016b) found similar results for coal dust/methane explosions, for which higher P_{max} and K_{st} values were obtained when methane concentrations between 0.75–1.25% were present in the system. Moreover, the flame propagation velocity and the maximum flame temperature were higher for coal/methane mixtures than those for coal dust or other dust flames (Liu et al., 2007). Considering the variability of the severity results, Garcia-Agreda et al. (2011) identified five different regimes for hybrid mixtures at different dust and gas concentrations: non-explosion, synergic, dust drive, gas driven and dual fuel explosion. A synergic explosion zone limited by the Le Chatelier law, the gas lower explosive limit and the dust minimum explosive concentration has been proposed. Regardless the recent efforts to describe the particularities of hybrid mixtures explosions (Cuervo, 2015), the simultaneous phenomena of complex thermal transfers, combustion kinetics and turbulence/combustion interactions are still not well understood.

It is interesting to modify the particle size in order to identify the influence of the particles surface on these phenomena. It has previously been demonstrated that the elementary particle size has an influence on the severity of the explosion of hybrid mixture systems. Kosinski et al. (2013) studied the explosion severity of carbon black nanoparticles/propane/air mixtures. According to this study, the propane concentration must be higher than the lower explosion limit in order to cause an explosion. However, in comparison with pure propane, the explosion severity was higher for low concentrations of carbon black nanoparticles and lower for high concentrations. These results suggest that the combustion process is modified by the heat loss generated by the carbon black nanoparticles (Kosinski et al., 2013). The study of hybrid mixtures explosions involving nanometric-sized dust allows to isolate the turbulence and combustion interactions, as they are highly stable at dispersion, and to work at low turbulence levels, which is not possible when working with micrometric-sized dust due to the sedimentation phenomenon. In a previous study (Torrado et al., 2016), a different explosion severity of carbon black/methane/air mixture was obtained compared to that obtained from gas explosions under quiescent conditions. The flame front changed from a semi-parabolic to a non-uniform shape as a consequence of multiple perturbations occasioned by the burnt particles.

In the present work, the influence of carbon black nanoparticles on the explosion severity and on the burning velocity is analyzed and compared to that of inert particles (i.e. alumina nanoparticles). This will help to elucidate the influence of carbon black nanoparticles on the combustion reaction and on the heat radiation transfer.

2. Materials and methods

2.1. *Materials and mixtures selection*

In this study, both Corax N550 and Printex XE2 (Orion) were chosen as carbon black nanoparticles. In addition, alumina AP-D 0.05 (Struers) was set and studied as inert particles. Due to its chemical simplicity and because carbon/methane mixtures are often encountered in various industrial applications, methane was selected in this research as combustible gas. In order to study the influence of an initial concentration of soot in a methane air mixture, initial low concentrations of carbon black nanoparticles were chosen. Regarding the scale of precision, the lowest carbon black nanoparticle concentration measured was set at 2 g m^{-3} (0.4% molar concentration). Tests were also performed at 6 g m^{-3} (1.2% molar concentration),