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Influence of inert gas addition on propagation indices of methane–air deflagrations

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ABSTRACT

The work examines the characteristic indices of laminar deflagrations propagating in methane–air gaseous mixtures diluted by several inert gases: He, Ar, N_2 or CO_2. Experiments were performed in two spherical vessels of different volumes with central ignition, at ambient initial conditions. Mixtures with variable methane concentrations (6–12 vol%) and variable inert concentrations (5–40 vol%) were studied, in order to outline the inert influence on the most important and accessible safety-related parameters: the peak explosion pressure, the maximum rate of pressure rise (or the related property, i.e. the deflagration index) and the explosion time (the time necessary to reach the peak explosion pressure). Among the studied inert additives, CO_2 is the most efficient, followed by N_2, Ar and He. Inert gas addition to any flammable CH_4–air mixture determined the decrease of both experimental and adiabatic explosion pressure and of the maximum rate of pressure rise, along with the increase of the explosion time. Using an equation that describes the heat balance of the isochoric combustion of a fuel–air mixture under non-adiabatic conditions, a correlation between the peak explosion pressure and the mole fraction of inert gas was derived and validated for CH_4–air– inert mixtures.

1. Introduction

The flammability properties of methane–air mixtures were frequently measured and summarized in publications, as methane is the main constituent of natural gas, mine gas or biogas. Its recent uses as an alternative fuel for engines, in the form of LNG (Liquefied Natural Gas) or CNG (Compressed Natural Gas), explains the revival of interest for its explosive combustion. Reduction of the hazard associated to explosions of methane–air mixtures is made by dilution with an inert gas, most frequently nitrogen, carbon dioxide, water (vapor) or even exhaust gas (i.e. a mixture of carbon dioxide, water vapor and nitrogen). Numerous studies were undertaken on explosion propagation in methane–air diluted by inert gases, driven by the need to measure MIC, the minimum inert concentration necessary for suppressing the explosion. However, in many cases, the specific conditions for operating a reactor or a plant require the addition of an inert gas with a concentration lower than MIC. In this case, the explosion still occurs but its characteristic parameters in confined conditions (the peak explosion pressure, the maximum rate of pressure rise, the time to peak explosion pressure, the normal burning

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velocity) differ strongly from the parameters of non-diluted methane–air mixture, according to the added inert gas and its concentration. For an adequate risk mitigation, it is important to determine the correlations between the amount of each added inert and the propagation indices. At the same time, the peak explosion pressure, the maximum rate of pressure rise and the time to peak explosion pressure are necessary for the design and construction of venting devices, for modeling the turbulent combustion and for optimization of internal combustion engines.

Previous studies on confined explosions of CH_4–air mixtures in the presence of inert additives have been made mostly in spherical vessels or in cylindrical vessels of low aspect ratio (length/diameter close to 1)(Cammarota et al., 2010; Di Benedetto et al., 2009 and Di Benedetto et al., 2012; Mashuga and Crowl, 1998; Sapko et al., 1976; Senecal and Beaulieu, 1998; Tang et al., 2014; Zhang et al., 2014a and Zhang et al., 2014b; Wang et al., 2014; Wu et al., 2010), but data on explosion propagating in long tubes exist as well (Pursell et al., 2011; Gant et al., 2011). Among inerts, the most studied were N_2 and CO_2. Carbon dioxide has excellent inerting ability for any methane–air flammable mixture, revealed by strong decrease of the rate of heat release from methane–air–CO_2 mixtures, resulting in a decrease of both maximum rate of pressure rise and normal burning velocity. Recent studies (Pursell et al., 2011; Gant et al., 2011) have shown that carbon dioxide is increasingly used for offshore transport and enhanced oil recovery – a fact that determines the contamination of natural gas stream by CO_2. This supports the interest for determining the propagation indices of CH_4–air–CO_2 mixtures in various conditions of pressure and temperature. Besides inert gases, water fog has been added to gaseous methane–air mixtures; the study of Zhang, et al. (2014b) revealed a significant suppression effect of water sprayed under an ultrasonic field.

The present study reports data obtained during combustion of methane–air mixtures, diluted by various amounts of inert gases, in two spherical vessels: S1 (diameter $\Phi = 33.7$ cm; volume $V = 20$ L) and S2 ($\Phi = 10$ cm; $V = 0.52$ L) both with central ignition. Various initial conditions were used: variable methane concentrations within 6–12 vol% and, for each methane–air composition, various concentrations of inert gas (He, Ar, N_2 or CO_2) within 5–40 vol%. All measurements have been made at ambient initial pressure and temperature. The measured explosion pressures were examined versus the corresponding adiabatic values, calculated by assuming that chemical equilibrium was reached within the flame front. Previous studies on additive influence on propagation indices have been made in spherical vessel S2, using ethane–air (Giurcan et al., 2015), LPG (Liquefied Petroleum Gas)–air (Razus et al., 2009), ethylene–air (Movileanu et al., 2011a, 2011b, 2013) and propene–air (Razus et al., 2007a,b). As shown by these studies, valuable information on explosion propagation can be obtained from experiments in a spherical vessel smaller than the standard 20 L vessel, where buoyancy effects may disturb the flame propagation in slowly burning mixtures (either lean or rich fuel–air mixtures or highly diluted fuel–air mixtures). The present data provide a useful comparison between the propagation indices measured in the two spherical vessels, using methane–air with variable equivalence ratios, φ, as reference mixtures. The results will be compared to other literature data on methane–air and methane–air–inert confined explosions. The reported data represent useful information for explosion mitigation of methane–air of variable concentration by means of inert gas addition. According to a previously described method by Giurcan, et al. (2015) the extrapolation of correlations between the inert gas concentration and the maximum explosion pressure or the maximum rate of pressure rise to threshold values of these propagation indices affords the estimation of the Minimum Inerting Concentration (MIC) and Limiting Oxygen Concentration (LOC) for methane–air–inert gaseous mixtures.

2. Experimental

Measurements of deflagration indices have been made in two spherical vessels, each included in another experimental set-up.

Vessel S1 is a standard 20 L spherical vessel manufactured by Kühner, together with the explosion ignition and monitoring systems and the adequate software. The gaseous mixtures were prepared in the explosion vessel, by partial pressure method, at a total pressure of 101.3 kPa. The mechanical stirring of the gas inside the explosion vessel provided the homogeneity of the mixture, which was left afterwards 30 min to become quiescent, and then it was ignited. The ignition was performed using a high voltage electric spark with a spark energy of 10 J.

Vessel S2 is a spherical vessel with a diameter of 10 cm (0.52 L), fitted with a valve for gas admission/evacuation, a pair of electrodes, an ionization probe and a pressure transducer. The vessel has been connected to a set-up formed from a system for preparing and storing the flammable mixtures, the ignition system and the measuring system. The system for preparing and storing the flammable mixtures consisted of a vacuum and gas–feed line, tight at pressures from 10 Pa to 400 kPa. This line interconnected the vacuum pump, the gas cylinders with methane, air and inert gases, the metallic cylinder for mixture storage and the explosion vessel. The vacuum pump maintained a vacuum of 10 Pa in the explosion vessels, after each experiment. Ignition was made with inductive-capacitive high voltage sparks produced between two stainless steel electrodes with rounded tips; the spark gap was in the geometrical centre of the vessel. The ignition energy delivered by the inductive-capacitive sparks was 3–5 mJ, maintained at this low level as to avoid the turbulence induced by a high energy input.

The pressure variation during explosion was recorded with a piezoelectric pressure transducer (Kistler 601A), connected to a Charge Amplifier (Kistler 5001N) and to an Acquisition Data System Tektronix TestLab 2505, operated at 10^4 signals per second.

In experiments with vessel S2 the fuel–air mixtures were prepared by partial pressure method at a total pressure of 400 kPa in a cylindrical steel vessel, using compressed air ($[N_2]/[O_2] = 3.76$) taken always from fresh atmosphere, before any current activity in the laboratory. The mixtures were used 48 h after mixing. The fuel–air–inert mixtures were prepared directly in the combustion vessel, at ambient pressure, using the required inert and the fuel–air mixture previously prepared. In each experiment, the fuel–air–inert mixture was left 15 minutes to become homogeneous before ignition. Other details concerning the experimental set-up and the operating mode have been previously described by Razus et al. (2007a,b) and Movileanu et al. (2011a,b, 2013). The composition of fuel–air–inert mixtures is expressed always by mentioning the initial, undiluted CH_4–air mixture and the added inert gas, e.g. (7% CH_4–air) + helium mixture; this allows a comparison