



Propagation indices of methane-nitrous oxide flames in the presence of inert additives



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ABSTRACT

The propagation indices (explosion pressures, rates of pressure rise, severity factors and explosion times) of CH₄-N₂O flames in the presence of inert gases (He, Ar, N₂ and CO₂) were determined by experiments performed in a spherical vessel with central ignition. Lean and stoichiometric mixtures ($\phi = 0.8$ and 1.0) with a variable inert gas concentration between 40 and 60% were studied, at variable initial pressures within 0.50 and 1.75 bar. Inert gas addition to each of the studied CH₄-N₂O mixtures results in 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. CO₂ was found to be the most efficient inert additive, followed by N₂, Ar and He. The measured and computed propagation properties are examined as functions of the total initial pressure and the inert gas concentration. The correlation of peak explosion pressures with the initial pressures, derived from the heat balance of the isochoric combustion of a fuel-oxidizer mixture under non-adiabatic conditions, is used to evaluate the heat losses during the closed vessel combustion, dependent on the initial CH₄/N₂O ratio and on inert concentration of the flammable mixtures.

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1. Introduction

Nitrous oxide is known as oxidant or nitriding agent in industrial processes e.g. semiconductor manufacture or production of sulphuric acid, adipic acid and caprolactam (Hirano, 2004; Vandebroek et al., 2005) but its main use is as oxidant in propulsion systems of rockets or as additive that increases the power output of engines (Merrill, 2008; Tyll and Herdy, 2001). This practical use was determined by the exothermic decomposition of nitrous oxide, under the influence of strong electric discharges (Merrill, 2008) or at temperatures higher than 625 °C in the presence of solid catalysts (Tyll and Herdy, 2001). Important emissions of N₂O in atmosphere are generated by industrial processes or by enzymatic nitrification and denitrification of fertilizers in soil. The increase of N₂O concentration in the atmosphere has a strong environmental impact as N₂O acts as ozone-depleting substance (Severin, 2015).

In spite of the increasing number of practical applications, the hazards associated to the handling, storage or use of pure N₂O or of its mixtures with various fuels are not yet fully evaluated, especially for mixtures at initial conditions different from ambient. A realistic assessment of such hazards requires the determination of propagation indices for fuel-N₂O mixtures, in a wide range of initial conditions. In the early studies on N₂O as oxidizer, the selfignition of gaseous fuel-N₂O mixtures in the temperature range 750–1000 °C was examined (Borisov, 1978; Borisov et al., 1981; Destriau, 1962), for fuels from various flammability classes: hydrogen, methane, propane, n-butane, ethene, ammonia. Data on flammability limits of forced ignition are, however, more comprehensive (CHEMSAFE, 2014; Holland et al., 1971; Kollmer and Hölderich, 2003; Koshihara et al., 2010, 2015; Meye et al., 2012; Pfahl et al., 2000; Pannetier and Sicard, 1955; Shebeko et al., 1988, 2013, 2014; Vandebroek et al., 2004, 2005; Zabetakis, 1965). Many results on binary H₂-N₂O and ternary H₂-N₂O-inert gas mixtures are available, since hydrogen-nitrous oxide mixtures might develop easily DDT (deflagration-to-detonation transition) and raise important safety concerns. A particular case is the study of Pfahl et al. on the flammability of complex gaseous mixtures generated into the dome space of storage tanks by nuclear wastes, formed mainly by

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hydrogen, ammonia, methane, and nitrous oxide, in various amounts (Pfahl et al., 2000). For CH₄-N₂O-N₂ mixtures, Pfahl measured the flammability limits and reported 70.5% as the minimum inerting concentration of N₂ for CH₄-N₂O-N₂ ignition. Pfahl reported also maximum explosion pressures for lean CH₄-N₂O mixtures ([CH₄] = 2 ... 6%) measured in a closed vessel spherical vessel with V = 11.25 L, after ignition with electric sparks of variable energy (Pfahl et al., 2000). At the leanest flammable concentration (2.7% CH₄), the measured peak explosion pressure was about 12 bar, close to the value obtained by the decomposition of pure N₂O (11.8 bar). For propane-N₂O and n-butane-N₂O mixtures at ambient initial conditions, flammability limits and minimum inerting concentrations of N₂, CO₂ and several halogen derivatives (CF₂Cl₂, C₂F₄Br₂, C₂ClF₅) were reported by Shebeko et al. (1988). The lower explosion limits of several fuel-N₂O gaseous mixtures and their temperature dependence were reported by Meye et al. (2012), together with their MESH at ambient initial temperature (Maximum Experimental Safe Gap). Later studies on combustion in closed vessels of near-limit H₂-N₂O and CH₄-N₂O mixtures in the presence of fluorinated hydrocarbons (CHF₃, C₂HF₅, C₄F₁₀) at ambient initial conditions (Shebeko et al., 2013, 2014) reported measured peak explosion pressures, maximum rates of pressure rise and laminar burning velocities in correlation with the additive concentration. Peak explosion pressures and severity factors of CH₄-N₂O in a closed vessel with a volume of 8 L, at 60 °C and initial pressures between 1 and 10 bar were examined by Vandebroek et al. (2004) in comparison with data referring to CH₄-H₂-N₂O and H₂-N₂O mixtures. In the absence of any diluent, the explosion of the stoichiometric CH₄-N₂O mixture is characterized by high frequency fluctuations of pressure that might facilitate the transition of the deflagration into a detonation, even at ambient initial pressure.

Experiments in a cylindrical vessel (V = 0.94 L) with C₁-C₇ alkane-N₂O and C₁-C₇ alkane-O₂ mixtures of variable alkane/oxidizer ratios delivered the lower and upper explosion limits and the explosion pressures for mixtures between the flammability limits (Koshiba et al., 2015). For alkane-N₂O and alkane-O₂ mixtures diluted with 30% N₂ (the flammable mixture represents 70% of the total mixture) the peak explosion pressures and the maximum severity factors K_G were examined in correlation with the number of carbon atoms of each fuel. Similar data (flammability limits, peak explosion pressures and severity factors) were obtained during closed-vessel explosions of toluene-N₂O mixtures and toluene-air in the presence of inert additives, at 70 °C and various initial pressures (1–6 bar) (Vandebroek et al., 2005). More recently, experiments in a spherical vessel on H₂-N₂O and H₂-N₂O-N₂ mixtures (Bane et al., 2011) delivered not only explosion pressures and severity factors but also normal burning velocities, discussed in correlation with the inert gas concentration.

In the present study, the deflagration propagation in stoichiometric mixtures of methane with nitrous oxide in the presence of several inert gases (nitrogen, argon, helium and carbon dioxide) is examined. Experiments were made at ambient initial temperature in a spherical explosion vessel with central ignition at various total initial pressures within 0.5–1.75 bar and various inert gas concentrations within 40 and 60%. For nitrogen-diluted mixtures, a lean CH₄-N₂O mixture ($\phi = 0.8$) was also studied and compared with the corresponding stoichiometric mixture. The measured propagation indices (peak explosion pressure, maximum rate of pressure rise, severity factor and explosion time) are discussed in correlation with the inert gas nature and concentration.

2. Experimental

Experiments were carried out in a stainless steel spherical vessel

with central ignition of volume V = 0.52 L (10 cm diameter), designed to withstand an internal pressure of 40 bar. The vessel was equipped with a port for the gas feed and evacuation valve, two ionization probes (with tips mounted at 3 and 5 mm away from the side wall, respectively), ignition electrodes and a pressure transducer. The vacuum and gas-feed line, tight at pressures between 0.5 mbar and 4.5 bar, connected the combustion vessel with cylinders containing CH₄, N₂O, the inert gas and with a metallic cylinder for mixture storage and a vacuum pump.

The gaseous mixtures CH₄-N₂O-inert were prepared by the partial pressure method and used 24 h after mixing the components, at a total pressure of 4 bar. The average standard error observed in partial pressure measurement was 1% for CH₄ and N₂O, and 0.5% for the inert gases.

Ignition was made with inductive-capacitive sparks produced between stainless steel electrodes. Their energy varied between 1 and 5 mJ, in order to avoid the turbulence produced by an excess energy input at initiation. The pressure variation during explosions was recorded with a piezoelectric pressure transducer (Kistler 601A), connected to a Charge Amplifier (Kistler 5001SN). The signals from the Charge Amplifier and the ionization probes were recorded with an acquisition data system TestLab™ Tektronix 2505, usually at 5000 signals per second. Other details on the experimental set-up and procedure were previously given (Mitu et al., 2012, 2016; Movileanu et al., 2013; Razus et al., 2010).

Methane, N₂O, Ar, He, N₂ (99.99%) and CO₂ (99.5%) (SIAD Italy) were used without further purification.

Methane-N₂O mixtures with equivalence ratio $\phi = 1.0$, diluted by 40–60% inert gas (He, Ar, N₂ or CO₂) (the inert gas concentration is expressed as % of end composition) were examined at total initial pressures between 0.50 and 1.75 bar. In the same range of initial concentrations, experiments were made with a lean CH₄-N₂O mixture ($\phi = 0.8$) diluted by 40–60% N₂. Minimum 3 experiments have been performed for each initial condition of explosive mixture. The average standard error observed in explosion pressures was 2% and 3.5% in the maximum rates of pressure rise.

The maximum rates of pressure rise $(dp/dt)_{max}$ were computed from pressure-time records after smoothing the $p(t)$ data by Savitzky-Golay method, based on the least squares quartic polynomial fitting across a moving window within the data. This involved the analysis of 500–700 points within ignition and the time necessary to reach the peak explosion pressure. The method, described in previous papers (Razus et al., 2007, 2011) has the advantage of producing a smoothed first derivative without filtering the data. In all cases, we used 5% smoothing level.

3. Computing program

Equilibrium calculations on CH₄-N₂O-inert gas mixtures were performed by means of 0D COSILAB package (COSILAB, 2013). The adiabatic flame temperature, adiabatic explosion pressure and burned gas composition in isochoric combustion were computed after assuming the chemical equilibrium is reached in the burned gas. Fifty-three compounds have been considered as combustion products.

4. Results and discussion

Typical pressure-time records obtained at $p_0 = 1$ bar during the combustion in the spherical vessel with central ignition of the stoichiometric methane-nitrous oxide mixture diluted with nitrogen are shown in Fig. 1, together with a record for the stoichiometric methane-air mixture obtained in identical initial conditions. The explosions of CH₄-air and CH₄-N₂O-N₂ (50% N₂) reach their peak pressures at close times, but the pressure rise is quite

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