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Methane-unconventional oxidant flames. Laminar burning velocities of nitrogen-diluted methane–N₂O mixtures

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ABSTRACT

The flame propagation in nitrogen-diluted CH₄–N₂O mixtures was monitored by pressure measurements during explosions in a spherical vessel with central ignition. The burning velocities were obtained from experimental measurements of pressure variation during closed vessel explosions and from the detailed modelling of free laminar premixed flames. Lean- and stoichiometric methane–nitrous oxide mixtures (equivalence ratios: 0.8 and 1.0) diluted by various amounts of nitrogen between 40 and 60 vol% were studied at various initial pressures between 0.3 and 1.8 bar and ambient initial temperature. Nitrogen addition to each CH₄–N₂O mixture results in the decrease of laminar burning velocity and flame temperature, along with the increase of flame width. Examination of burning velocity variation against average flame temperature in experiments at constant initial pressure and various inert concentrations allowed the determination of the overall activation energy; examination of burning velocity dependence on pressure, at constant inert concentration, allowed the determination of the overall reaction orders. For all CH₄–N₂O–N₂ mixtures, the temperature, volumetric rate of heat release and reactive species concentration profiles across the flame front were examined versus similar data characteristic to stoichiometric methane–air mixtures. The most important elementary reactions in CH₄–N₂O–N₂ and CH₄–air mixtures were identified by means of sensitivity analysis.

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1. Introduction

Nitrous oxide is used in various technical applications, like for instance as oxidant in propulsion systems or as additive in motor racing to increase the power output of engines (Merrill, 2008; Tyl and Herdy, 2001). Flammable fuel–N₂O mixtures raise safety issues in the nuclear industry, as it was observed that stored nuclear waste generates various amounts of nitrous oxide mixed with hydrogen, ammonia and methane (Pfahl et al., 2000). Nitrous oxide is generated in large amounts by waste incineration, especially when treating sewage sludge, due to the high nitrogen content of this material and low combustion temperatures (Kramlich and Linak, 1994). Nitrous oxide is also generated,

as a side product, in industrial processes such as nitric acid or adipic acid production (Pérez-Ramírez et al., 2003) or by decomposition of nitrogenous fertilizers together with other nitrogen oxides (Reay et al., 2012). The nitrogen oxides mixture (NO_x) represents a major atmospheric pollutant, acting as ozone-depleting substance (Severin, 2015). When exhaust gases are recirculated in the combustion cycle of internal combustion and gas turbine engines, the nitrogen oxides have an important impact on the combustion properties of the fresh charge by promoting the hydrocarbon oxidation (Faravelli et al., 2003; Dagaut et al., 2005; Sivaramakrishnan et al., 2007; Herzler and Naumann, 2012). N₂O was identified as an important intermediate during the combustion of nitramine-based propellants (Allen et al., 1995; Javoy et al., 2009). Therefore, the development of accurate kinetic models of gas phase combustion for systems involving nitrogen are needed to enable the accurate modelling of the chemistry of hydrocarbon–N₂O flames.

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Numerous studies regarding the combustion characteristics of flammable mixtures containing nitrous oxide are reported. The specific feature of these systems is the important increase of heat of combustion brought by the exothermal dissociation of N_2O in fuel– N_2O mixtures, in comparison with fuel–air mixtures. Mathieu et al. (2015) examined the influence of NO_x on fuel–oxygen and fuel–air combustion and reported ignition delay times of CH_4 – O_2 mixtures seeded with NO_2 or N_2O , measured in shock tube experiments. They outlined the sensitizing effect of NO_2 and N_2O to hydrocarbon/oxygen ignition. Their results showed that the ignition delay time decreased when NO_2 and, to a lesser extent, N_2O were added to the mixture. Konnov examined the conversion of nitrous oxide in laminar premixed flames of $\text{CH}_4 + \text{O}_2 + \text{Ar}$ seeded with N_2O (Konnov and Dyakov, 2009) and analyzed the pathways of formation and destruction of nitrogen oxides. Mével and Shepherd (2015) reported ignition delay times of small hydrocarbon– N_2O mixtures with and without O_2 obtained by shock-tube experiments and chemical kinetic simulations.

The flame propagation in N_2O –fuel mixtures was studied under various conditions. Vanderhoff et al. (1982) reported measurements of flame temperature and concentration of major species (N_2 , H_2 , CO , CO_2 , and H_2O) in the burnt gas region of CH_4 – N_2O low-pressure flat flames using laser Raman spectroscopy. Habeebullah et al. (1991), Zabarnick (1991) and Vandooren et al. (1992) studied the structure of premixed CH_4 – N_2O or CH_4 – NO flames in the presence of O_2 or Ar by measurements of stable and unstable species in flat flames at 30–50 torr. Flame structure studies were further used to validate various mechanisms developed for N_2O -containing mixtures.

The laminar burning velocities of H_2 and C_1 – C_3 hydrocarbons with N_2O were determined by Powell (Powell et al., 2009, 2010) at near-atmospheric pressure and further used for modelling high temperature combustion. The sensitivity studies and reaction path analyses were employed to elucidate the important pathways to the overall reaction rate for these systems. Laminar burning velocities for methane–nitrous oxide mixtures were measured by Pfahl et al. (2000) but these mixtures included other flammable constituents such as hydrogen and ammonia. An experimental investigation of the influence of trifluoromethane on the laminar burning velocity of near-limit mixtures of methane–nitrous oxide was performed by Shebeko et al. (2011). In non-premixed flames of CH_4 – O_2 and C_2H_6 – O_2 , the replacement of O_2 by N_2O was found to inhibit their propagation (the burning velocity decreased with increasing mass fraction of N_2O in the fresh mixture), as reported by Newman-Lehman et al. (2013) by experimentally studying and kinetic modelling of counterflow flames. Other studies used hydrogen (Mével et al., 2009; Bane et al., 2011) or ethylene (Bangalore Venkatesh et al., 2013) as test fuels; the authors reported the laminar burning velocities of inert-diluted fuel– N_2O mixtures (Ar or N_2 as inerts), used for validating several detailed mechanisms.

In the present study, the laminar burning velocities of lean and stoichiometric methane–nitrous oxide mixtures (equivalence ratios: 0.8 and 1.0) diluted by various amounts of nitrogen between 40 and 60%, at various initial pressures between 0.3 and 1.8 bar and ambient initial temperature, are reported. The burning velocities of CH_4 – N_2O – N_2 mixtures derived from experimental data (pressure rise measurements

during the early stage of flame propagation in a spherical vessel) (Razus et al., 2006, 2010a; Mitu et al., 2015) are compared with computed values obtained by kinetic modelling of free laminar premixed flames with the GRI 3.0 mechanism (Smith et al., GRI-Mech 3.0, 1999), using the COSILAB package (COSILAB, 2013). From the burning velocity variation against the average flame temperature, at constant initial pressure and various inert concentrations, the overall activation energy of CH_4 oxidation with N_2O was determined. From the pressure dependence of the laminar burning velocity, at constant inert concentration, the overall reaction orders were calculated. For all systems, the temperature, heat release rate and concentration profiles of most important chemical species together with the flame front width were examined in correlation with the initial composition and compared with similar data characteristic to lean- and stoichiometric methane–air mixtures.

2. Experimental

Experiments were performed in a spherical vessel of radius $R = 5$ cm ($V = 0.52$ L), which can withstand an internal pressure up to 40 bar. The vessel was equipped with several ports for gas admission/evacuation, two electrodes for ignition, an ionization probe and a piezoelectric pressure transducer. The vessel was connected to a set-up for preparing and storing the flammable mixtures, the ignition system and the measuring system. The experimental setup, as given previously by Mitu et al. (2014), is schematically shown in Fig. 1.

The system for preparing and storing gaseous mixtures, gas tight at pressures from 0.5 mbar to 40 bar, consisted of a vacuum and gas-feed line, a vacuum pump and a metallic cylinder for mixture storage. The vacuum pump maintained a vacuum down to 0.1 mbar in the explosion vessel, after each experiment. Ignition was made with inductive-capacitive high voltage sparks produced between two stainless steel electrodes; the spark gap was in the geometrical centre of the vessel. The ignition energy delivered by the inductive-capacitive sparks was between 1 and 5 mJ in order to avoid the turbulence produced by excessive energy input at initiation. 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 5000 signals per second.

The CH_4 – N_2O – N_2 gaseous mixtures were obtained in a metallic cylinder by the partial pressure method and used 24 h after mixing the components, at a total pressure of 4 bar. For each studied mixture the equivalence ratio of the CH_4 – N_2O mixture was indicated, along with the added N_2 amount, as the volumetric nitrogen concentration in the end mixture ($\text{CH}_4 + \text{N}_2\text{O} + \text{N}_2$).

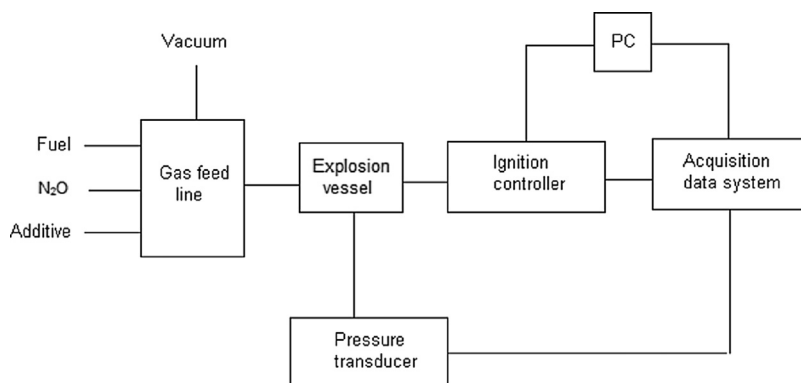


Fig. 1 – Scheme of the experimental set-up.

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