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Methane, ethane, and ethylene laminar counterflow diffusion flames at elevated pressures: Experimental and computational investigations up to 2.0 MPa

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ABSTRACT

A newly designed high-pressure combustion facility was used to study the structures and extinction conditions of counterflow diffusion flames in air for nitrogen-diluted methane, ethane, and ethylene, from 0.1 MPa to 2.0 MPa. Besides employing thermocouples to measure temperature profiles, strain rates at extinction were measured and compared with predictions of two different chemical–kinetic mechanisms (San Diego and USC). In addition, the nitrogen in the fuel and oxidizer streams was replaced by helium for one of the methane tests of extinction strain rate as a function of pressure. In all cases, the strain rate at extinction was found to increase with pressure up to about 0.3–0.5 MPa and to decrease with pressure thereafter, on up to 2.0 MPa, although with helium there was a clear leveling tendency beyond 1.0 MPa. While these behaviors were in qualitative agreement with most predictions of the chemical–kinetic mechanisms, in a number of cases the quantitative discrepancies were well beyond the experimental uncertainty. This underscores the desirability of improving chemical–kinetic descriptions for applications at elevated pressures. Such improvements for the San Diego mechanism are introduced here for two of the steps involving hydroperoxyl that become increasingly important with increasing pressure.

1. Introduction

The ability of chemical–kinetic mechanisms and transport descriptions to predict combustion processes is tested most easily and accurately by well-controlled laboratory experiments performed at normal atmospheric pressures, where measurements of strain rates at extinction, in particular, provide relevant tests of chemical–kinetic descriptions. In many practical applications, however, the combustion occurs at elevated pressures. Since the combustion chemistry varies with pressure, often non-monotonically in certain respects, it is desirable to pursue correspondingly well-controlled laboratory combustion experiments at pressures above atmospheric, as further tests of predictions. But such experiments, unfortunately, are difficult and expensive to design and perform. We have recently constructed a high-pressure combustion facility in which experiments on laminar counterflow diffusion flames were carried out. Results of these experiments with hydrogen as the fuel have recently been published [1]. In the present paper we report and discuss corresponding results for methane, ethane, and ethylene, as well as showing the effects of different reaction rates on comparisons between predictions and measurements for all of the fuels tested.

There have been a number of previous high-pressure counterflow experimental studies. Niemann et al. [1] recently carried out experiments on hydrogen flames at pressures up to 1.5 MPa, measuring temperature profiles and providing experimental confirmation for the non-monotonic pressure dependence of extinction strain rates predicted computationally by Sohn and Chung [2]. In addition, Figura and Gomez [3] successfully stabilized non-premixed methane flames at elevated pressures up to 3.0 MPa. Their experiments were conducted with the fuel and oxidizer streams diluted with either nitrogen or helium. Temperature profiles were measured and compared with predictions [3], but extinction strain rates were not addressed. On the other hand, Maruta et al. [4] earlier had measured critical conditions for extinction of non-premixed methane flames with the fuel and oxidizer streams diluted with carbon dioxide (CO_2) and with nitrogen at pressure up to 0.8 MPa. They found that for flames diluted with CO_2, critical conditions for extinction were influenced by radiation re-absorption, but they did not specifically discuss variations of the extinction strain rates with pressure. Böhm and Lacas [5] also measured critical conditions for extinction of non-premixed methane flames up to pressures of 0.6 MPa. Their emphasis was on soot formation and destruction,

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but they did demonstrate experimentally that with increasing pressure, the measured strain rate at extinction first increased and then decreased. Their computations with detailed chemistry also showed an increase in the value of the strain rate at extinction with increasing pressure followed by a decrease, but the decrease was less pronounced than that measured [5].

Much earlier than these investigations, Sato [6] had measured critical conditions for extinction of non-premixed methane and ethane flames at pressures up to 10 MPa. These measurements were made on flames stabilized over the surface of a porous cylinder (a Tsuji burner), and they showed that with increasing pressure, the strain rates at extinction for methane and ethane remained constant for pressures up to 2.0 MPa and 1.0 MPa, respectively, but with further increase in pressure, up to 10 MPa, the strain rate at extinction decreased [6]. This qualitatively different behavior at the lower pressures may be associated with the fact that the fuels in these experiments were not diluted, resulting in strain rates at extinction that were quite appreciably higher. There also have been experiments on liquid-fuel flame extinction in stagnation-point flows of alkanes, performed at about the same time, at pressures up to 2.0 MPa [7,8] that show regions of increasing extinction strain rates, leading up to a plateauing peak region, followed by a downward-sloping region as pressure is increased, qualitatively similar to the results with diluted methane. In the present work critical conditions for extinction and flame-temperature profiles are determined for an appreciably wider range of gaseous-fuel parameters than is available in this literature.

2. Experimental facility and procedures

Figure 1 is a schematic illustration of the experimental facility. It shows the pressure chamber, the gas-supply system, and the data-acquisition and control system. The counterflow burner is placed inside the chamber. The inner diameter of the fuel and oxidizer ducts of the counterflow burner is 20 mm, and the separation distance between the ducts is 10 mm. Gaseous fuel mixed with inert gas is injected from the bottom duct and diluted oxidizer from the top duct. Fine wire screens are placed at the exits of the both ducts. This makes the tangential component of the flow velocity negligibly small at the exit of the ducts, establishing plug flow. The reactant ducts are surrounded by annular shrouds that provide an inert curtain flow to minimize the influence of ambient gas on the reaction zone. The products of combustion are removed into an exhaust-treatment system where they are cooled before they are purged into the in-house exhaust system. This prevents hot gases and water vapor from accumulating inside the pressure chamber. As a consequence, the temperature in the chamber does not increase, and condensation of water vapor on the chamber walls and windows is avoided.

Gases to the pressure chamber are supplied from standard compressed gas cylinders. All gaseous streams are controlled by computer-regulated mass-flow controllers. The selection of the type of mass-flow controller for a given reactant depends on the required experimental range of flow rates. The mass-flow controllers employed here have maximum flow rates in the range of 30–500 standard liters per minute and operating pressures up to

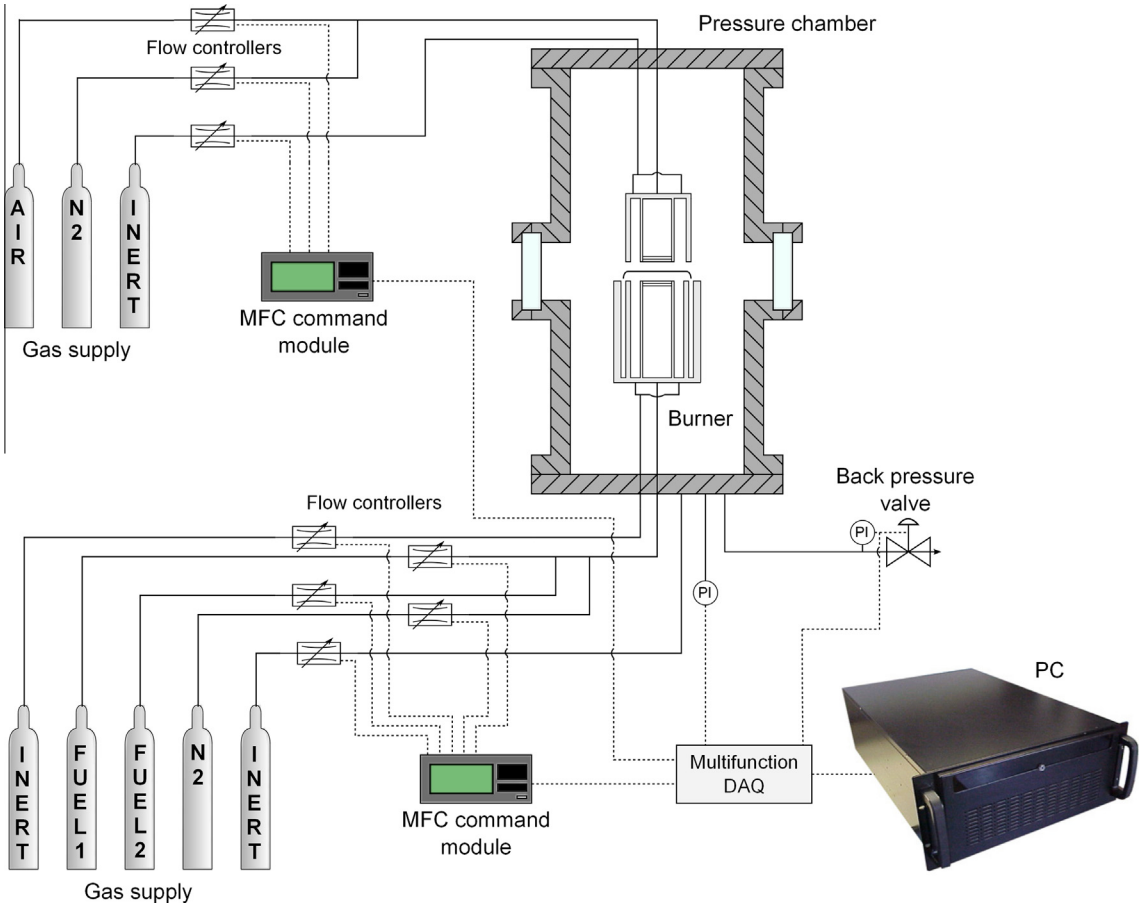

Fig. 1. Schematic illustration of the experimental arrangement. The burner is housed inside a pressure chamber, with optical access. The gas-supply system includes the mass-flow controllers and mass-flow-controller (MFC) command modules, as well as the data-acquisition and control systems.