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The influence of carbon monoxide and hydrogen on the structure and extinction of nonpremixed and premixed methane flames

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Abstract

An experimental and computational study is carried to elucidate the influence of carbon monoxide (CO) addition to nonpremixed methane (CH₄) flames. A Burke–Schumann (flame-sheet) formulation is applied to laminar counterflow diffusion-flame experiments, reported here, in which CO was added to either methane–nitrogen mixtures or oxygen–nitrogen mixtures at normal atmospheric pressure, with both feed streams at normal room temperature. Experimental conditions were adjusted to fix selected values of the stoichiometric mixture fraction and the adiabatic flame temperature, and the strain rate was increased gradually until extinction occurred. At the selected sets of values, the strain rate at extinction was measured as a function of the CO concentration in the fuel or oxidizer stream. It was found that with increasing amounts of CO in the oxidizer stream, the strain rate at extinction first increased and then decreased. With increasing amounts of CO in the fuel stream, changes in values of the strain rate at extinction was small in comparison to those measured for CO addition to the oxidizer stream. The experimental results are in agreement with predictions obtained employing detailed chemistry. A numerical investigation was carried out to characterize the influence of hydrogen and carbon monoxide on the structure and burning velocities of stoichiometric premixed methane flames. The mass fraction of the reactants in the mixture were so chosen that the adiabatic temperature is constant. The flame speed increases when hydrogen is added to the reactant stream. For CO addition the flame speed first increases and then decreases.

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1. Introduction

In view of uncertainties in the supply of fossil fuels and environmental concerns arising from

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pollutant and green-house gases emissions from these fuels, there is considerable interest in exploring alternate sources of energy. Fuels such as coke oven gas, biogas obtained by wood pyrolysis, reformat gas, synthetic gas from biomass gasification, and blast furnace gases are now considered to be environmentally friendly, and economically viable and profitable [1]. Although, the specific composition of such gases depends upon the fuel sources and processing techniques, a typical mix mainly contains methane (CH₄), hydrogen (H₂), carbon monoxide (CO), carbon dioxide (CO₂), nitrogen (N₂), and trace amounts of ethane and propane. Depending on the sources, there is a substantial variability in the fuel composition, and heating values [2]. Design of combustion systems that can run efficiently on a wide range of operating conditions employing these fuels is challenging. In order to understand the impact of the variability in fuel compositions on the combustion performance and emissions, understanding of the changes in combustion properties of methane in presence of hydrogen and carbon monoxide is required. Here experimental and computational studies are carried out to characterize the influence of CO on the structure and critical conditions of extinction of nonpremixed methane flames. Computational studies are also carried out to characterize the influence of H₂ and CO on the structure and burning velocities of stoichiometric premixed methane flames.

Recently an experimental and computational study was conducted focused on characterizing the influence of hydrogen on the structure and critical conditions of extinction of methane flames [3]. In this previous study experiments were conducted on laminar nonpremixed flames that was stabilized in the mixing layer between two counterflowing reactant streams. One stream, called the fuel stream was made up of CH₄ and nitrogen (N₂), and the other stream, called the oxidizer stream was a mixture of oxygen (O₂) and N₂. Hydrogen was added either to the fuel stream or to the oxidizer stream [3]. Experimental conditions were adjusted to fix selected values of the stoichiometric mixture fraction and the adiabatic flame temperature, and the strain rate was increased gradually, maintaining the momentum balance of the two streams, until extinction occurred. At the selected sets of values, the strain rate at extinction was measured as a function of the hydrogen concentration in the fuel or oxidizer stream. The experimental results were in close agreement with computational predictions employing detailed chemistry [3]. The present study complements and extends this previous study. Nonpremixed combustion of CH₄ with added CO is considered first followed by premixed combustion of CH₄ with added H₂ and CO.

2. Nonpremixed combustion of CH₄ with CO addition

A general formulation is presented first, followed by description of the experiments conducted at 1 atm. The experimental data is compared with results of numerical computation carried out using a detailed chemical-kinetic mechanism.

2.1. Formulation

Consider two counterflowing streams flowing toward a stagnation plane. The fuel stream is made up of CH₄ and N₂ and the oxidizer stream, is made up of O₂, and N₂. Carbon monoxide is added either to the fuel stream or to the oxidizer stream. The mass fraction of CH₄ in the fuel stream is $Y_{F,1}$, and that of O₂ in the oxidizer stream is $Y_{O_2,2}$. The temperature of the fuel stream is T_1 and the temperature of the oxidizer stream is T_2 . When CO is added to the fuel stream its mass fraction is $Y_{CO,1}$ and it is $Y_{CO,2}$ when added to the oxidizer stream. Subscript 1 refers to conditions in the fuel stream and subscript 2 to conditions in the oxidizer stream.

In comparing influences of CO addition to different streams, choices must be made about what parameters to keep fixed. It has been established from activation energy asymptotic analysis [4–7] and rate-ratio asymptotic analysis [8,9] that the Damköhler number, $Da = \tau_f/\tau_c$, plays a key role in establishing the critical conditions of extinction, where τ_f is the characteristic flow time and τ_c the characteristic chemical time. The characteristic flow time depends on the scalar dissipation rate, which in turn depends on the strain rate, and the stoichiometric mixture fraction, ζ_{st} . The chemical time depends on six parameters, given by the mass fraction of reactants $Y_{F,1}$, $Y_{CO,1}$, $Y_{O_2,2}$, and $Y_{CO,2}$, and the temperatures, T_1 , and T_2 . Because of the strong temperature dependencies of reaction rates, it is most important to make comparisons at fixed values of T_{st} . Otherwise differences associated with different reaction-zone temperatures are likely to dominate the results. After removal of effects of temperature variations, structures of reaction zones still vary with the stoichiometric mixture fraction [10]. It is therefore also desirable to make comparisons at fixed values of ζ_{st} . As a consequence, comparisons are made at fixed values of ζ_{st} and T_{st} .

The selections for the boundary values of mass fractions of CH₄, O₂, and CO are made using a formulation developed in the previous experimental and computational study of the influence of hydrogen on methane flames [3]. In this formulation the products of combustion are presumed to be carbon dioxide (CO₂) and water vapor (H₂O). Lewis numbers of unity are good approximations for the reactants, CH₄, O₂, CO as well as for the

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