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Counterflow Extinction of Premixed and Nonpremixed Methanol and Ethanol Flames

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Abstract

Experimental and numerical studies are conducted on extinction of methanol and ethanol flames. Two flame types are considered: premixed and nonpremixed. The studies are performed in the counterflow configuration. The burner used in the experiments is made up of two opposing ducts. In the premixed configuration the reactive stream, made up of fuel, oxygen, and nitrogen, is injected from one duct, and a nitrogen stream is injected from the other duct. In the nonpremixed configuration the fuel stream is made up of fuel and nitrogen, and it is opposed by an oxidizer stream made up of air. The fuels are prevaporized by flowing nitrogen through a heated bath of liquid fuel. The velocities of the reactant streams at the injection planes are calculated from measured flowrates. These velocities are used to calculate the strain rate. The temperature of the fuel stream and that of the nitrogen stream at the injection plane are measured using thermocouples. Critical conditions of extinction are reported, giving the strain rate at extinction as a function of the mass fraction of various reactants. In the premixed configuration various equivalence ratios of the premixed stream are tested. Further experiments are conducted in the nonpremixed configuration by preheating the oxidizer stream and measuring the temperature at which autoignition occurs. Numerical calculations are performed using detailed chemistry at conditions corresponding to those used in the experiments. Critical conditions of extinction and ignition are calculated. The numerical results are compared with the experiments.

1 Introduction

Methanol (CH_3OH) and ethanol ($\text{C}_2\text{H}_5\text{OH}$) are frequently mentioned as alternative fuels. They can be produced in large quantities from agricultural products such as corn, wheat, sugar-cane, and wood, or from waste products such as sewage and municipal waste. They are considered CO_2 -neutral in regards to the greenhouse effect because the same amounts of carbon dioxide (CO_2) and water (H_2O) that result from complete combustion are needed for the photosynthesis of plants from which they are produced. Reasons for using methanol and ethanol as practical fuels or additives to practical fuels are discussed in Ref. [1]. To verify chemical-kinetic models to be used in combustion engine modeling, fundamental studies of combustion of methanol and ethanol are necessary. Previous authors have investigated laminar burning velocities [1, 2], species measurements in flames [3] and flow reactors [4], shock-tube studies [5], and droplet combustion [6–8]. Methanol is highly miscible with water. Because water is one of the main products of combustion, experiments on droplet combustion and on flames stabilized over a liquid pool are difficult, as water-vapor formed in the flame diffuses to the liquid surface. The present study is performed on prevaporized fuels. Experiments are performed on extinction and autoignition in a strained laminar counterflow. The results are useful for turbulence modeling of practical combustion systems, where strain is an important factor.

Description of Experimental and Numerical Studies

Steady, axisymmetric, laminar flow of two counterflowing streams toward a stagnation plane is considered. Figure 1 shows a schematic illustration of the counterflow configuration employed in the present study. Two flame types are used in the present study, a nonpremixed and a

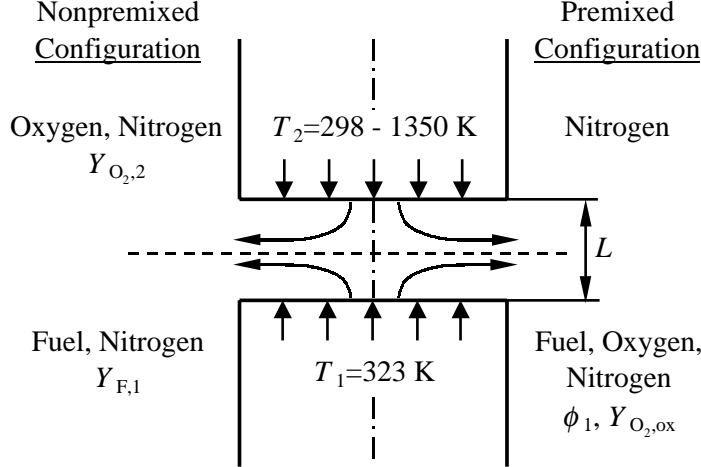


Figure 1: Schematic illustration of the counterflow configuration.

premixed configuration. They are characterized by the arrangement of the reactants among the two in-flowing streams, in particular whether oxygen is introduced into the top or into the bottom duct. In the following, subscripts 1 and 2 are used to identify various quantities with their corresponding ducts. The jets that flow into the mixing layer between the two ducts are described by values of V_i and ρ_i , $i=1,2$. V denotes the component of the flow velocity normal to the stagnation plane at the exit of the duct. The tangential components of the flow velocities at the boundaries are presumed to be equal to zero (plug-flow boundary conditions). In the experiments and numerical calculations the momenta of the counterflowing reactant streams $\rho_i V_i^2$, $i = 1, 2$ at the boundaries are kept equal to each other. This condition ensures that the stagnation plane formed by the two streams is approximately in the middle of the region between the two ducts. The value of the strain rate, defined as the normal gradient of the normal component of the flow velocity, changes from one duct exit to the other. [9]. The characteristic strain rate on the side above the stagnation plane a_2 is presumed to be given by [9]

$$a_2 = \frac{2|V_2|}{L} \left(1 + \frac{|V_1|\sqrt{\rho_1}}{|V_2|\sqrt{\rho_2}} \right). \quad (1)$$

Equation 1 is obtained from an asymptotic theory where the Reynolds numbers of the laminar flow at the boundaries are presumed to be large [9]. Note that in the premixed configuration the flame can take a position on the fuel side of the stagnation plane and will therefore experience a different amount of strain. In the present work a_2 represents a characteristic measure of the flow-rates only. It is used for all comparisons between experimental and nu-

merical results. A detailed description of the burner is given elsewhere [10–12]. The flow rates of gases are measured by computer-regulated mass flow controllers. The calibrated accuracy of these mass flow controllers is $\pm 1\%$. The velocities of the reactants at the boundaries are presumed to be equal to the ratio of their volumetric flow rates to the cross-section area of the ducts. Each main reactant stream is surrounded by a nitrogen curtain flow with equal velocity. Duct 2 is equipped with a heating device that allows the stream to be preheated up to 1350 K. The temperature of the gas at the exit of the duct is measured using a Pt-Pt 13% Rh thermocouple with wire diameter of 0.07 mm and a junction diameter of 0.21 mm. The measured temperatures are corrected for radiative heat losses assuming spherical shape of the junction, a constant Nusselt number of 2.0, and a constant emissivity of 0.2 [13]. The accuracy of the corrected temperature is expected to be better than ± 25 K. The fuels used in this study are prevaporized by flowing a nitrogen flow through a heated bath of the liquid fuel. This vaporizer has been used in previous studies [10].

Nonpremixed Flames

Nonpremixed experiments are carried out using air as oxidizer stream ($Y_{O_2,2} = 0.233$). The fuel stream is a mixture of fuel and nitrogen. The temperature of the fuel stream T_1 is 323 ± 10 K. In the extinction experiments the temperature of the oxidizer stream T_2 is 298 K and the distance between the exits of the ducts is $L = 10$ mm. Extinction experiments are conducted by establishing a flame at strain rate $a_2 < a_{2,E}$ and increasing the strain rate until extinction is observed. This procedure is repeated for various dilutions of the fuel stream. Autoignition experiments are conducted with the mole fraction of fuel, $X_{F,1}$, maintained at 0.15. The distance between the fuel and oxidizer duct is $L = 12$ mm. At a given strain rate and oxidizer temperature $T_2 < T_{2,I}$ the flow-field is established. The temperature of the oxidizer stream is gradually increased until autoignition takes place. To ensure that the experiments can be compared to numerical calculations, autoignition has to take place close to the centerline, where species and temperature profiles are flat with respect to the radial direction and only a function of the normal distance from the stagnation plane. A high-speed CCD video camera is used to observe the ignition event at a frame rate of 500 s^{-1} . To prevent ignition after the reactant streams leave the strained flow-field between the ducts, the last 10 mm of the curtain duct (48 mm inner diameter) are made up of a water cooled brass ring. This lowers the temperature of the curtain flow, but leaves the main flow unchanged.

Premixed Flames

Experiments are conducted on a diluted fuel-air mixture versus nitrogen (see Fig. 1, right hand side). The equivalence ratio of the fuel stream $\phi_1 = (Y_{F,1}/Y_{O_2,1})(Y_{O_2}/Y_F)_{stoich}$, where the fuel mass fraction is set in relation to the oxygen mass fraction $Y_{O_2,1}$ and the stoichiometric fuel/oxygen mass ratio. A second quantity, $Y_{O_2,ox}$, the oxygen mass fraction in the oxidizer that is mixed with fuel, is used to describe the dilution of the fuel stream. Here oxidizer refers

to the total of oxygen and nitrogen in the fuel stream. If pure air is mixed with fuel then $Y_{O_2,ox} = Y_{O_2,1}/(Y_{O_2,1} + Y_{N_2,1}) = 0.233$. A necessary condition for a stable premixed flame is that $a_{2,min} < a_2 < a_{2,E}$. The quantity $a_{2,min}$ is roughly the strain rate that is obtained when V_1 equals the laminar burning velocity of the premixed fuel stream. At this value the propagation of the flame back into the fuel duct is only prevented by heat loss to the duct. An increase in the value of a_2 pushes the flame away from the fuel duct into the mixing layer between the two duct exits. In the present experiment the value of $a_{2,E}$ is measured, above which the strained premixed flame extinguishes. Because the premixed flame is sensitive to fluctuations in the reactant concentrations, a mixing chamber with a volume of 500 ml is used to mix the gases before they are introduced into the fuel duct. In the experiments the distance between the ducts is $L = 12$ mm. The temperature of the fuel stream T_1 is 323 ± 10 K, and the temperature of the oxidizer stream T_2 is 298 K. Two sets of experiments are conducted. In one, $\phi_1 = 1$ and $a_{2,E}$ is measured for various values of $Y_{O_2,ox}$. In the other, $Y_{O_2,ox}$ is held constant at 0.185 and $a_{2,E}$ is measured as a function of ϕ_1 .

Numerical Procedure

For the computations the computer program FlameMaster is used, which was developed at RWTH-Aachen [14]. At the boundaries of the domain the mass fluxes of the reactants and the exit velocities are specified, according to the values in the experiments. Plug-flow boundary conditions are used. The conservation equations of mass, momentum, and energy and the species balance equations used in the formulation of the numerical problem are summarized elsewhere [14–16]. The species balance equations include thermal diffusion and the energy conservation equation includes radiative heat losses from carbon dioxide and water vapor [14]. Buoyancy is neglected. Detailed chemical-kinetic mechanisms are tested and compared versus the experimental results. Held and Dryer [17] published a comprehensive mechanism for methanol comprising 97 reversible reactions among 21 species. Marinov [18] developed a chemical-kinetic mechanism comprised of 383 reversible reactions among 56 species describing the high temperature oxidation of ethanol.

Results and Discussion

Nonpremixed Flames

Figure 2 shows experimental data on extinction of nonpremixed flames of CH_3OH and C_2H_5OH . For a certain strain rate the fuel mass fraction in the fuel stream is plotted, below which a flame will extinguish. It is shown that for strain rates around $100 s^{-1}$ the mass fractions of CH_3OH and C_2H_5OH are comparable, while at higher strain rates CH_3OH can burn at a lower fuel mass fraction. Numerical computations using chemical-kinetic mechanisms are shown as lines, and they agree well with experimental results.

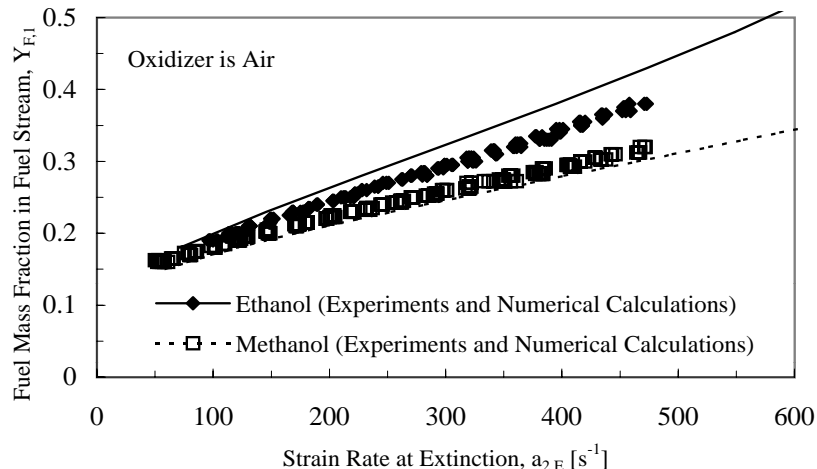


Figure 2: Experimental data showing the fuel mass fraction in the fuel stream as a function of the strain rate at extinction. The oxidizer is air. The points represent measurements and the lines are results of numerical calculations using a chemical-kinetic mechanism of Held et al. [17] for CH_3OH and a mechanism of Marinov [18] for C_2H_5OH .

The symbols in Fig. 3 show experimental data on autoignition of CH_3OH and C_2H_5OH . The figure shows the oxidizer temperature as a function of the strain rate. The mole fraction of fuel in the fuel stream $X_{F,1} = 0.15$ for both fuels. The ignition temperatures are found to be similar and increase for higher strain rates. The increase with a_2 is larger for CH_3OH than for C_2H_5OH . To compare the autoignition characteristics on a mass basis, Fig. 4 shows the oxidizer temperature at autoignition as a function of the fuel mass fraction in the fuel stream $Y_{F,1}$. The strain rate is $300s^{-1}$. For fuel mass fractions smaller than approximately 0.2, C_2H_5OH ignites at lower oxidizer temperatures than CH_3OH , while at higher mass fractions CH_3OH ignites at lower temperatures. The lines in Fig. 4 show the results of numerical computations using the chemical-kinetic mechanisms of Held et al. [17] for CH_3OH and Marinov [18] for C_2H_5OH .

Premixed Flames

Results of extinction of stoichiometric diluted premixed flames, using nitrogen as opposing stream, are shown in Fig. 5. Here, the mass fraction of oxygen in the oxidizer, below which extinction occurs, is plotted versus the strain rate. The symbols represent measurements. Numerical calculations are performed using the chemical-kinetic mechanism of Held et al. [17] and the mechanism of Marinov [18] for CH_3OH and C_2H_5OH , respectively. Figure 6 shows the extinction strain rate as a function of the equivalence ratio for a constant oxygen mass fraction in the oxidizer. The highest extinction strain rate is observed in the stoichiometric to moderately rich region roughly around $\phi_1 = 1.05 - 1.1$. Numerical results are shown as lines. It is observed that the chemical-kinetic mechanism of Held et al. [17] predicts experimental

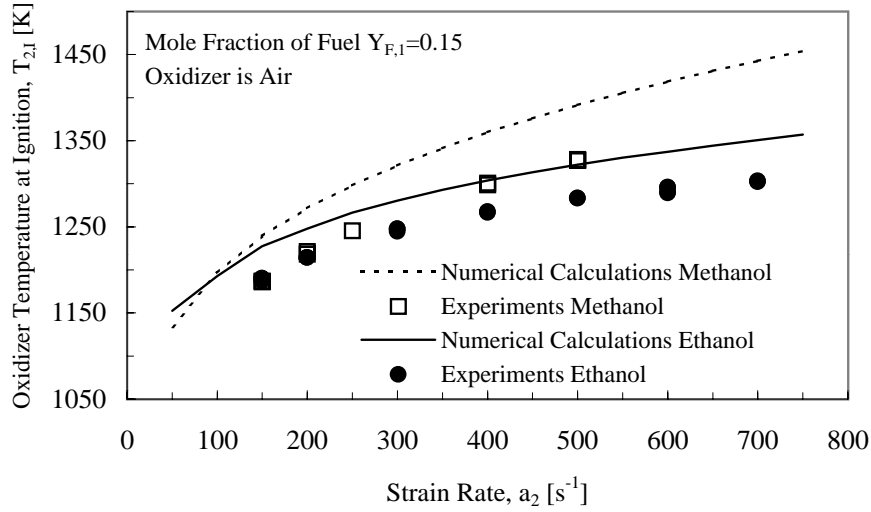


Figure 3: Experimental data showing the oxidizer temperature as a function of the strain rate at autoignition. The points represent measurements. The lines are results of numerical calculations using the chemical-kinetic mechanisms by Held et al. [17] et al. for CH_3OH and the mechanism by Marinov [18] for $\text{C}_2\text{H}_5\text{OH}$. The temperature of the fuel stream is 323 K. Oxidizer is air.

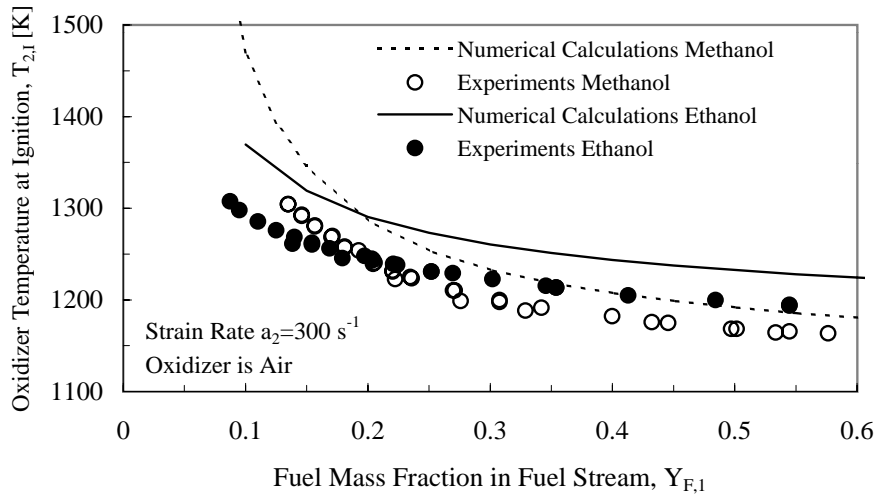


Figure 4: Experimental data showing the oxidizer temperature at autoignition as a function of the fuel mass fraction at a strain rate of 300 s^{-1} . The points represent measurements, and the lines are results of numerical calculations. The temperature of the fuel stream is 333 K. Oxidizer is air.

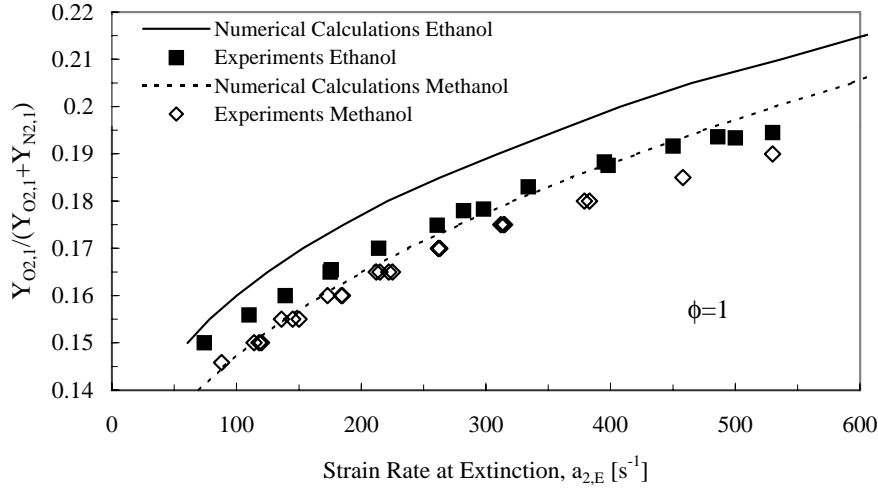


Figure 5: Results of extinction of strained premixed flame in counterflow with nitrogen. The oxygen mass fraction in the oxidizer is shown as a function of the strain rate at extinction. The points represent measurements. The lines show results of numerical calculations. The fuel stream equivalence ratios $\phi_1 = 1.0$. The temperature of the premixed stream is 323 K and that of the nitrogen stream is 298 K.

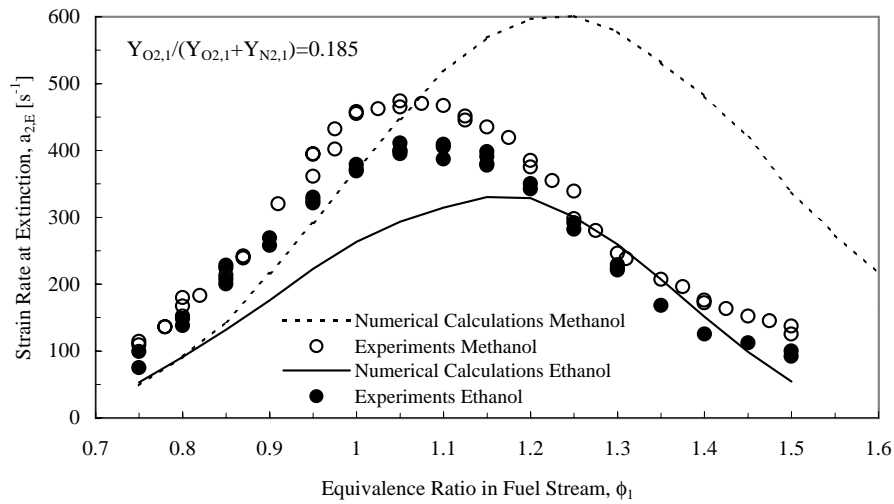


Figure 6: Results of extinction of a strained premixed flame in counterflow with nitrogen. The strain rate at extinction is shown as a function of the equivalence ratio of the fuel-stream. The points represent measurements. The lines show results of numerical calculations. The oxygen mass fraction in the oxidizer is 0.185. The temperature of the premixed stream is 323 K and that of the nitrogen stream is 298 K.

extinction strain rates for CH₃OH well in the fuel lean region but over-predicts them in the fuel rich region. The chemical-kinetic mechanism of Marinov [18] performs well in predicting fuel rich conditions for C₂H₅OH, but predicts somewhat earlier flame extinction in the stoichiometric region.

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