

Extinction and Auto-Ignition of C₃ and C₄ Alcohols in Laminar Nonpremixed Flows

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Alcohols produced from biomass show significant potential to be an alternative to conventional crude oil derived transportation fuels. In this experimental and computational study, the combustion of C₃ alcohols (n-propanol, iso-propanol) and C₄ (n-butanol, iso-butanol) alcohols is investigated in nonpremixed, nonuniform flows. Experiments are performed employing the counterflow configuration with a fuel stream made up of vaporized fuel and nitrogen, and an oxidizer stream of air. The strain rate at extinction is measured as a function of the mass fraction of fuel in the fuel stream. At fixed mass fractions of fuel critical conditions of auto-ignition are measured in terms of the temperature of the oxidizer stream as a function of the strain rate.

Kinetic modeling is performed using a semi-detailed mechanism. This mechanism is made up of more than 7000 reactions among 300 species. Critical conditions of extinction and ignition were calculated and compared with experimental data.

1 Introduction

Legislative requirements to decrease or at least limit the increase in output of greenhouse gases into the earth's atmosphere has motivated numerous studies on combustion of renewable fuels. Oxygenated fuels, in particular alcohols, show considerable promise. They can be derived from biomass, and thus are considered to be carbon neutral which helps meeting emission requirements for greenhouse gases. Alcohols can be derived from fermentation of biomass or as by-products of Fischer-Tropsch processes. There is considerable interest in promoting larger alcohols over ethanol which is currently used as an additive to gasoline. Propanol and butanol are less corrosive hence require minimal changes to a vehicle's fuel system. They have higher energy densities than ethanol, and their octane rating is closer to that of gasoline. N-butanol's octane rating and the limited solubility (7-8%) enable it to be used not only as an additive but as a direct fuel. The production of

bio-derived propanol and butanol is labor intensive and yields are low, hence it is not nearly as widely spread as a transportation fuel additive as ethanol.

Previously published work on flame structures of propanol isomers and butanol isomers, employing the same experimental setup and chemical kinetic mechanism, provided measurements and numerical simulations of species and temperature profiles. [1,2] Here it is extended to include critical conditions of extinction and auto-ignition.

2 Experimental Investigation

Extinction and auto-ignition of non-premixed flames of n-propanol, iso-propanol, n-butanol and iso-butanol were measured employing the counterflow configuration. Figure 1 shows a schematic illustration of the counterflow configuration. Steady, axisymmetric, laminar flow of two counterflowing streams toward a stagnation plane is considered. In this configuration, a fuel stream made up of prevaporized fuel and nitrogen is introduced through the bottom duct, and an oxidizer stream of air is injected through the upper duct. A mixing-layer develops around the stagnation plane. The exit of the fuel duct is called the fuel boundary and the exit of the oxidizer duct the oxidizer boundary. Fine wire screens are located at the exits of the ducts. As a consequence, the tangential component of the flow velocities vanishes at the boundaries. This allows the use of “plug flow boundary conditions” in numerical simulations. The mass fraction of fuel, temperature, and the component of the flow velocity normal to the stagnation plane at the fuel boundary are represented by $Y_{F,1}$, T_1 , and V_1 , respectively. The mass fraction of oxygen, temperature, and the component of the flow velocity normal to the stagnation plane at the oxidizer boundary are represented by $Y_{O_2,2}$, T_2 , and V_2 , respectively. The exit diameter of the fuel duct and the oxidizer duct is 23 mm. The distance between the fuel boundary and the oxidizer boundary is represented by L . The value of the strain rate, defined as the normal gradient of the normal component of the flow velocity, changes from the fuel boundary to the oxidizer boundary. The characteristic strain rate on the oxidizer side of the stagnation plane a_2 is given by [3]

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

Extinction experiments are conducted with a duct separation of $L = 10\text{mm}$, and $T_2 = 298\text{ K}$. The temperature of the fuel stream, T_1 , for all fuels is 400K. At some selected value of $Y_{F,1}$, the flame is stabilized. The strain rate is increased by increasing V_1 and V_2 simultaneously, keeping momenta of the counterflowing streams balanced based on $\rho_1 V_1^2 = \rho_2 V_2^2$, until extinction is observed.

Auto-ignition experiments are carried out with a duct separation of $L = 12\text{mm}$. The fuel stream temperature of 400K and fuel mass fraction, $Y_{F,1}=0.4$. For selected values of strain rate a_2 the oxidizer stream temperature is increased until auto-ignition takes place. The velocities of the counterflowing streams are constantly adjusted based on the change in temperature to satisfy the momentum balance equation.

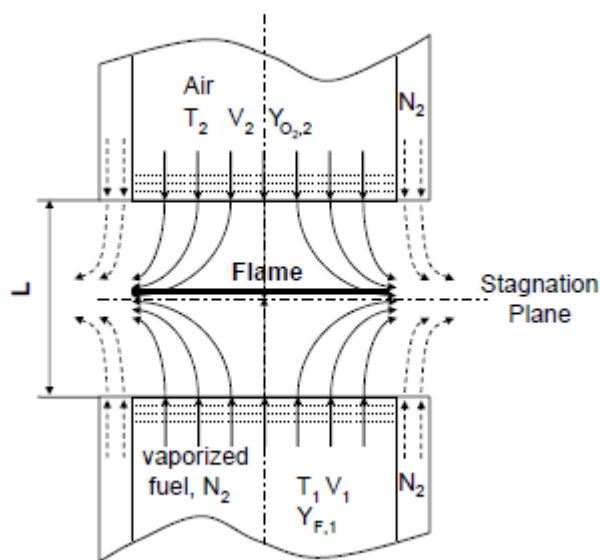


Figure 1. Schematic illustration of the counterflow flow field

3 Numerical Investigation

The numerical simulations are carried out using Chemkin. As a model the modified opposed-flow diffusion flame code was used. Kinetic modeling of the experimental data is performed using the Polimi mechanism. This kinetic mechanism along with the thermodynamic and transport data can be downloaded. [4] A detailed description of this code is given elsewhere. [5] This code includes multicomponent diffusion and thermal diffusion.

The boundary conditions are identical to those in the experiment. Auto-ignition and critical conditions of extinction are calculated and compared with experimental data.

4 Discussion

Figure 2 compares the critical conditions of extinction for n-butanol, n-Propanol, iso-butanol and iso-propanol. It shows the mass fraction of fuel in the fuel stream, $Y_{F,I}$, as a function of the strain rate, a_2 . The critical conditions of extinction of n-propanol and n-butanol are nearly the same. At a given value of $Y_{F,I}$ the value of the strain rate at extinction $a_{2,e}$ for iso-propanol is the lowest followed by iso-butanol. The comparison of the data shows the branched alcohols tested here are less reactive than their linear counterparts.

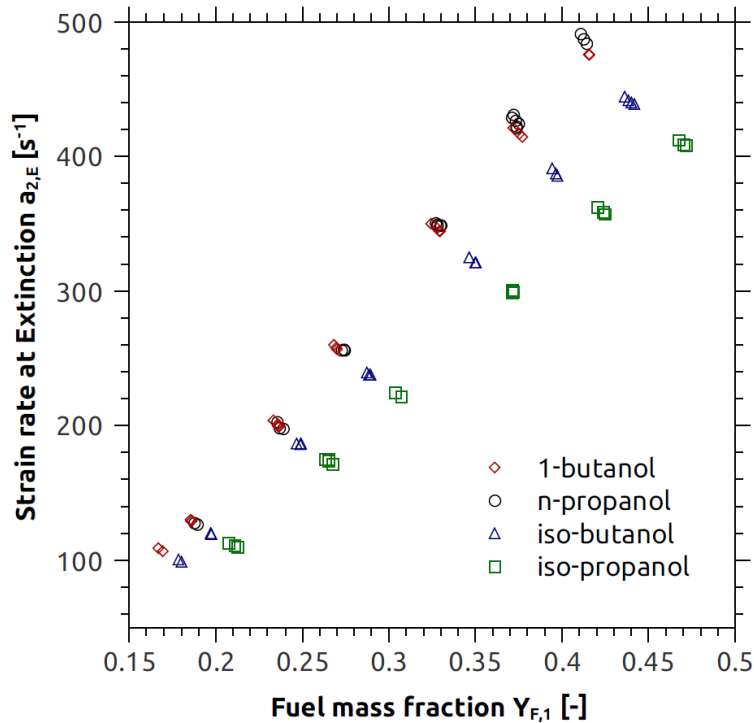


Figure 2. The mass fraction of fuel, $Y_{F,1}$, as a function of the strain rate at extinction, $a_{2,e}$, for n-butanol, n-Propanol, iso-butanol and iso-propanol. The symbols represent experimental data.

Figure 3 compares the critical conditions of auto-ignition for all four alcohols. It shows the temperature of air at auto-ignition, $T_{2,i}$, as a function of the strain rate, a_2 . At a fixed strain rate iso-propanol shows the highest auto-ignition temperatures, followed by iso-butanol. N-propanol and n-butanol have similar auto-ignition temperatures and in comparison to the branched alcohols are easier to ignite.

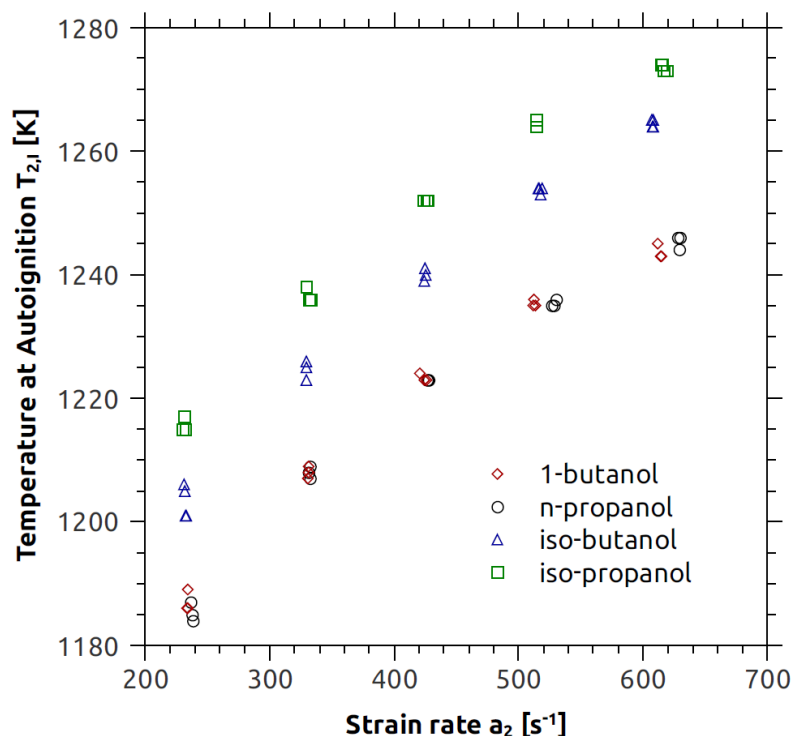


Figure 3. The temperature of the oxidizer stream at auto-ignition, $T_{2,i}$, as a function of the strain rate, a_2 for $Y_{F,1} = 0.4$. The figure shows data for n-butanol, n-Propanol, iso-butanol and iso-propanol. The symbols represent experimental data.

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