



Study of ignition chemistry on turbulent premixed flames of *n*-heptane/air by using a reactor assisted turbulent slot burner



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ABSTRACT

The changes in flame structure and burning velocity of premixed *n*-heptane/air flames associated with ignition chemistry have been investigated in a reactor-assisted turbulent slot (RATS) burner. Two distinct turbulent flame regimes are identified by varying the flow residence time and reactor temperature. A chemically frozen (CF) regime is observed at a reactor temperature of 450 K and a low-temperature ignition (LTI) regime is identified at 650 K. At a reactor temperature of 450 K, the measured turbulent burning velocities (S_T) exhibit a monotonic trend, proportional only to the turbulent intensity and laminar flame speed (S_L) calculated with the initial fuel/air mixture. At a reactor temperature of 650 K, S_T initially decreases with increasing flow residence times (decreasing turbulent intensity) but then increases once the reactor flow residence time exceeds the LTI delay. Furthermore, S_T in the LTI regime exhibits a strong correlation with the extent of low-temperature reactivity (defined by CH_2O concentration). The species distributions at the exit of the RATS burner after the onset of LTI are quantified by gas sampling-chromatography and used to compute the changes in S_L and mixture Lewis number (Le), which are shown to substantially change after the onset of LTI. Damköhler's scaling analysis indicates that the increase in S_T in the LTI regime originates from an increase in S_L , a decrease in Le , and an increase in turbulence intensity due to the heat release from the low-temperature chemistry. To examine the role of ignition chemistry on flame stability, flame flashback measurements have been performed by varying mean jet velocities and *n*-heptane/air mixture equivalence ratios for reactor temperatures of 450 and 650 K. Measurements at 650 K imply the strong influence of high-temperature ignition on flame stability phenomena.

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1. Introduction

Turbulent combustion of a premixed or partially premixed fuel/air mixture is the dominant energy conversion process in practical internal combustion engines [1–3]. As engine compression ratios and Reynolds numbers (Re) are increased to achieve higher efficiencies, stronger interactions between turbulence and chemistry appear in combustion phenomena such as burning velocity, ignition, flashback, and blow-off [2–4]. A number of studies have investigated the influence of pressure [5–8], Lewis number [9–12], and turbulent flame geometry [13–16] on turbulent

burning velocities (S_T) [1,5,9,16–18]. However, these studies have utilized mainly small hydrocarbon gaseous fuels, which have rather simple, high-temperature-only fuel oxidation chemistry. For example, hydrogen and small hydrocarbons like methane exhibit simple one-stage high-temperature ignition behavior, which can be represented by single activation energy in the Arrhenius global reaction scheme. On the other hand, large hydrocarbons such as *n*-heptane, relevant to real transportation fuels, can experience low-temperature chemistry (LTC) and two-stage ignition behavior [19–25] at high-pressure and moderate-temperature conditions. The occurrence of LTC can significantly modify the fuel composition, transport properties, and chemistry [26]. In spite of these rich chemistry behaviors pertinent to real fuel combustion, the turbulent flame characteristics of large hydrocarbon fuels, particularly for premixed flames, are not well understood.

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The two-stage ignition behavior of large hydrocarbon fuels [19–25] is composed of two distinct ignition processes. First-stage ignition is governed by low-temperature chemistry in which fuel radicals react with molecular oxygen to produce OH radicals *via* the isomerization and decomposition of alkylperoxy radicals. This ignition process is often referred to as low-temperature ignition (LTI). Second-stage ignition is controlled by the onset of thermal decomposition of H_2O_2 to form two OH radicals, which results in the transition to high-temperature combustion chemistry, and thus is often referred to as high-temperature ignition (HTI). LTI drastically changes the reactant composition and temperature by consuming over half of the large hydrocarbon fuel while generating combustion intermediate species (e.g. CO, CH_2O , and C_2H_4). At high pressures (greater than 20 atm), the LTI delay time of large hydrocarbon fuels becomes shorter than ~ 1 ms, certainly comparable to the overall flow time scale in a gas turbine combustor.

Obviously, the change in the reactant composition and temperature after LTI can play a significant role in affecting subsequent flame dynamics and chemistry. The decreased mean molecular weight of the reactants after LTI can alter the thermal and mass diffusion characteristics of the reactant stream that control the energy deposition and exchange to and from the flame reaction zone. Furthermore, changes in the transport characteristics of the fuel and oxidizer mixture can lead to thermal-diffusive flame instabilities (*i.e.* Lewis number effects), which, for cases of moderate turbulence intensities, have been shown to affect turbulent flame propagation [17,27,28]. In this regard, questions related to the role of ignition chemistry on turbulent premixed flames arise as follows: (1) How does ignition chemistry (LTI and HTI) specific to large hydrocarbon fuels alter the properties of a turbulent flame? (2) If ignition chemistry alters the turbulent flame characteristics, then what are the physical mechanisms for such modifications, or, more specifically, how do changes in the mixture chemistry/reactivity and transport properties after LTI affect the turbulent flame structure and burning velocity? (3) How do the changes in the turbulent flame dynamics as a result of LTI and HTI subsequently affect the turbulent flame stability?

As an endeavor to address these questions, a reactor-assisted turbulent slot (RATS) burner was developed to investigate the effects of LTI on turbulent premixed flames [26]. The RATS burner is capable of manipulating the onset of LTI by varying reactor temperatures and bulk flow rates. In our previous work [26], it was found that turbulent burning velocity (S_T) of *n*-heptane/air mixtures increases substantially when the incoming premixed *n*-heptane/air mixture had undergone LTI. Furthermore, through planar laser-induced fluorescence (PLIF) measurements of CH_2O , it was shown that these flames have very different turbulent flame structures than those of chemically frozen (CF) *n*-heptane/air mixtures (*i.e.* without the existence of LTI). Although the dramatic change in S_T in response to LTI was attributed to the change in chemical composition based on the overly-simplified adiabatic flow reactor calculation as shown in [29], the ambiguity from the lack of physical measurements of the LTI mixture composition jeopardizes the validity of this analysis.

The goals of the research presented herein are as follows: (1) to investigate the underlying physics controlling the accelerated turbulent burning velocity following the onset of LTI; (2) to quantify the species composition after LTI; and (3) to investigate the impact of LTI on turbulent flame stability, including flame flashback phenomena, using high-speed laser diagnostics. Firstly, an emphasis is placed on exploring the effects of changes in high temperature reactivity and the thermo-diffusive properties of the reactant mixture after LTI. Experiments are designed to measure S_T while controlling the onset of LTI for *n*-heptane/air mixtures by adjusting reactor temperature and reactor residence times. S_T is determined from the turbulent flame surface area, using 10-kHz PLIF

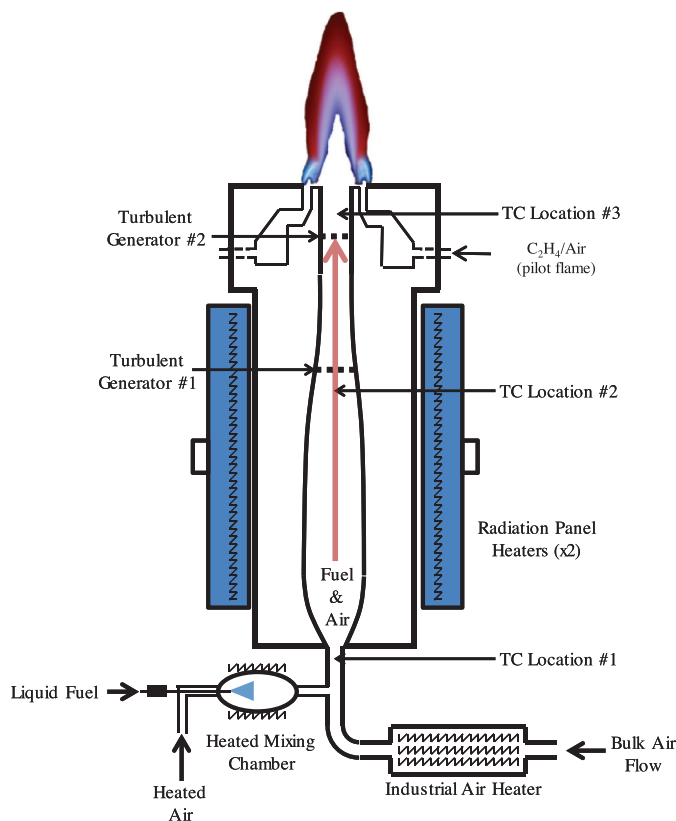


Fig. 1. Schematic of the reactor-assisted turbulent slot (RATS) burner system.

imaging of the OH radical. The reactant mixture components after LTI are also experimentally determined using gas chromatography (GC) and compared to kinetic model predictions. Modifications in flame chemistry and reactant transport *via* LTI are quantified by evaluating changes in the laminar flame speed (S_L) and the mixture Lewis number (Le). Secondly, time-resolved 10-kHz OH PLIF imaging is conducted to investigate the impact of LTI and HTI on the turbulent premixed flame flashback phenomenon by simultaneously changing the fuel concentration from lean to rich.

2. Experiment

The RATS burner, which has been described elsewhere [26,29], serves as the experimental platform for all measurements detailed below. A simple schematic is shown in Fig. 1. The stainless-steel burner is designed with sufficient height (55 cm) and internal heated volume (~ 650 cm³) to provide heated residence times spanning from 20 to 140 ms, which are controlled by the reactant bulk volumetric flow rate. This range of residence times spans the LTI delay, allowing for the activation of pre-flame LTC. The liquid fuel is pre-vaporized by injecting it along with 50 lpm (lpm) of preheated air (at 353 K) through a double-concentric tube into a vaporization chamber where the temperature is maintained at 450 K. It has been found that the temperature of the pre-vaporization chamber needs to be carefully controlled to prevent unexpected reactions within the vaporization chamber, which can jeopardize the desired flow residence time. The pre-vaporized fuel/air mixture is then introduced and mixed rapidly with a preheated main air stream at the bottom of the reactor. The industrial heaters used to heat the main air stream are capable of heating and maintaining 1000 lpm of air at 700 K along the length of the reactor. A mass flow controller (Sierra) is used to control the main air flow. As shown in Fig. 1, thermocouples placed in

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