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Investigation on laminar flame propagation of *n*-butanol/air and *n*-butanol/O_2/He mixtures at pressures up to 20 atm

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ABSTRACT

Bio-butanol is an important alternative fuel and has many advantages in engine combustion. In the present work, laminar flame propagation of *n*-butanol/air and *n*-butanol/(14% O_2/86% He) mixtures was investigated using a constant-volume combustion vessel at the unburnt temperature of 423 K, initial pressures of 1–20 atm and a series of equivalence ratios. Flame instabilities of *n*-butanol, including cellular instability and pulsating instability, were studied based on flame morphology. The unsteady transition from weak flame to normal flame was investigated based on the flame trajectories and this phenomenon became less perceptible in the flame with stronger combustion intensity and smaller Lewis number. The accelerated laminar burning velocity (LBV), thickened flame thickness, larger Markstein length and Markstein number indicate that the thermal diffusivity of *n*-butanol/O_2/He flames are greatly enhanced by helium addition. The smaller densities and lower reactant concentrations result in the smaller laminar burning fluxes of *n*-butanol/O_2/He flames than those of *n*-butanol/air flames. The high temperature kinetic model of *n*-butanol reported by Cai et al. was further developed with recent theoretical progresses on *n*-butanol reactions. The updated high temperature model was validated on the present experimental data and a vast amount of previous *n*-butanol combustion data. Sensitivity analysis was performed using the updated model under wide pressure and equivalence ratio ranges. The similarity in the sensitivities indicates that the sensitive reactions in *n*-butanol/air flames can be validated by using *n*-butanol/O_2/He flames, which greatly compensates the problem that the *n*-butanol/air flames are susceptible to flame instabilities at high pressures. The weaker pressure dependence of the LBVs of *n*-butanol/O_2/He flames indicates that their flame chemistry changes less to resist the pressure variation.

1. Introduction

Long chain bio-alcohols such as *n*-butanol are promising renewable fuels because they have many advantages over ethanol, such as higher energy content, lower water absorption, better miscibility with present fuels, and unproblematic use in conventional engines [1]. The prospect of butanol in substituting fossil-derived transportation fuels stimulates the experimental and modeling investigations on the combustion of *n*-butanol and its isomers. Among the four butanol isomers, *n*-butanol has attracted the most attentions in combustion research since it has many production methods from biomass feedstock [2,3]. A summary of the investigations and future directions for butanol combustion can be found in a recent review paper by Sarathy et al. [3]. They pointed out that additional fundamental experimental measurements are needed to better understand alcohol combustion under conditions of relevance to internal combustion engines, especially laminar burning velocity (LBV) at higher pressures (5–30 atm) and unburnt gas temperatures (400–600 K) [3].

LBV is one of the most important fundamental combustion parameters. The high pressure conditions, especially the initial pressures (P_u) higher than 10 atm, are of special interest in the LBV measurements and flame propagation studies in consideration of engine applications. The LBVs of *n*-butanol have attracted great attentions in laminar premixed flame research and have been measured using the spherically expanding flame [4–11], counterflow flame [12,13] and heat flux [14] methods. Several kinetic models [3,4,15–19] have been developed for *n*-butanol combustion. Among these models, most models [15,16,18,19] have not been validated against the LBVs of *n*-butanol [15,16,18,19] or only validated

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against the LBVs of *n*-butanol at $P_u \leq 1$ atm [4,17]. Among the investigations of *n*-butanol LBVs, the spherically expanding flame is the only method that has been performed at high pressures [5,6,8–10] while the other two methods are used at atmospheric pressure. Moreover, most of the previous spherically expanding flame studies of *n*-butanol focused on the measurements at 0.89–7.5 atm [4–8,10,12–14]. Only Beeckmann et al. [11,20] and Broustail et al. [9] have measured the LBVs of *n*-butanol at 10 atm. To the best of our knowledge, no LBV measurement at pressures over 10 atm has been performed for *n*-butanol. There are two main challenges for the high pressure laminar flame propagation investigations of *n*-butanol. Similar to other large fuels, the relatively high boiling point of *n*-butanol (390.6 ± 0.8 K [21]) makes the measurements at more than 10 atm quite difficult. Furthermore, the hydrodynamic and diffusive-thermal instabilities [22–25] make the flame surface greatly wrinkled at high pressures. These flames cannot be used to calculate LBVs, leading to the rich limits of measured LBVs for *n*-butanol and other large fuels at pressures higher than 5 atm [9,26]. Therefore the performance of *n*-butanol models on the predictions of LBVs can hardly be evaluated for the rich flames at $P_u > 5$ atm.

To measure the LBVs of *n*-butanol with sufficient equivalence ratios (ϕ) on both lean and rich sides, Wu and Law [10] used an oxidizer with reduced oxygen concentration and a mixture of argon and helium as diluent gas (13% O_2/38.1% Ar/48.9% He in mole). In previous studies [10,27–29], the strategies to suppress flame instability include two aspects of stable mechanism. The first one is to reduce oxygen concentration so that the adiabatic flame temperature is reduced and the flame thickness is increased. The hydrodynamic instability is consequently suppressed. The second one is to use helium as a component of the diluent gas. Both the Lewis number and the flame thickness are greatly increased, which retards the formation of diffusive-thermal and hydrodynamic cells [27]. Obviously, partly or totally replacing nitrogen with helium is an effective strategy to suppress flame instabilities [10,27–29]. In other words, the LBVs over a wide ϕ range can be measured at pressures higher than 10 atm.

The objective of the present work is to investigate the laminar flame propagation of *n*-butanol/air and *n*-butanol/(14% O_2/86% He) mixtures over a wide ϕ range at $P_u = 1$–20 atm. Further development of the *n*-butanol model reported by Cai et al. [19,30,31], which was not validated on the LBVs of *n*-butanol, was performed based on recent theoretical results. The updated model was validated on both the new LBV data in the present work and previous *n*-butanol combustion data. Special attentions were paid on the flame morphology, trajectory, parameters (i.e. LBV, laminar burning flux, Markstein length, Markstein number, pressure dependent coefficient) and chemistry under different pressures and diluent gas conditions.

2. Experimental method

All the experiments were performed on a constant-volume single-chamber cylindrical combustion vessel. The combustion vessel is made of 304 stainless steel and has an inner volume of 2.77 L (inner diameter 150 mm, inner length 152 mm). The asymmetry effect of non-spherical vessel can be largely weakened by applying near-unity length-diameter ratio [32,33]. The vessel was hydrostatically tested to 200 atm. Two quartz windows (diameter 100 mm, thickness 30 mm) were clamped to the vessel flanges, providing an optical access with 75 mm diameter clear apertures. The schematic diagram of the experimental apparatus can be found in Fig. S1 in the Supplementary Material.

During the experiment, partial pressure method was used for preparing combustible mixtures in the premixing vessel (9.06 L). Four pressure transducers (PT) with different ranges were used to improve the accuracy [34]. All vessels and pipelines were evacuated to about 20 mTorr before charging. The liquid fuel was vaporized in an electrically heated vaporizer before the injection to the premixing vessel. Prepared mixtures were stirred by a magnetic stirrer for 10 min to ensure uniform mixing. The volume of the premixing vessel (9.06 L) is more than 3 times as large as the combustion vessel. Thus, all measurements for one ϕ at different P_u only need one preparation of the combustible mixture, which largely reduces the time needed for mixture preparation. Relief valves were installed in both vessels to ensure experimental safety. Both the combustion vessel and the premixing vessel can be heated up to 500 K by flexible detachable heating jackets. In the present work, the combustion vessel, the premixing vessel, the vaporizer, and the pipelines were all heated at 423 K using PID temperature controllers and K-type thermocouples. Satisfactory temperature homogeneity was observed in both vessels and the maximum deviation from the target temperature was determined to be ±1 K.

The combustible mixture in the combustion vessel was ignited by two 500 µm-diameter tungsten electrodes. The gap between the tips of the two electrodes can be adjusted from 0 to 5 mm. Only one discharge occurred in each ignition with a duration much less than 10 µs. The ignition energy was minimized by decreasing the voltage and power input of the ignition coil. An outwardly propagating spherical flame was formed after the ignition and a schlieren system was used to record the flame propagation. Schlieren images were recorded by a high speed camera (Phantom V310). The camera was operated at 12,000 frames/s in the present work. The spatial resolution was set at 480 × 480 pixels to map a region of 75 × 75 mm^2. Data processing methods of spherically expanding flames were investigated in previous studies [35–37]. A Matlab based program was used to process the schlieren images and find flame front circles using a Circular Hough Transform based algorithm. The maximum uncertainty in pixels between the fitted circles and experimental points is about ±1 pixel. Kelley and Law [35] and Halter et al. [36] developed the nonlinear extrapolation method based on the weak stretched flame model of Ronney and Sivashinsky [38]. The nonlinear extrapolation method was used to process flame images in the present work. The selected range of flame radii was mainly 10–23 mm so that both the ignition effect [39–42] and confinement effect [32] can be minimized. The nonlinear evolution equation for the flame propagation is given by

$$\left(\frac{S_b}{S_b^0}\right)^2 \ln\left(\frac{S_b}{S_b^0}\right)^2 = -\frac{2L_b\kappa}{S_b^0} \tag{1}$$

where S_b is the stretched burning velocity, S_b^0 the unstretched burning velocity, κ the flame stretch rate, L_b the Markstein length and the subscript b refers to the burnt state. The Markstein length has been defined in [35] as

$$L_b = -\frac{1}{2}\frac{E_a}{R^0 T_{ad}} l_f^0 I(Le, T_u/T_{ad}) \tag{2}$$

where E_a is the activation energy of unstretched flame, R^0 is the universal gas constant, T_{ad} is the adiabatic flame temperature, l_f^0 is the laminar flame thickness, T_u is the unburnt gas temperature, Le is the Lewis number and $I(Le, \in) = \int_{\in}^{1} \frac{1}{x}[(\frac{x-\in}{1-\in})^{Le-1} - 1]dx$. The variable $\in$ is the ratio of burnt gas density to unburnt gas density.

In the present work, two sets of experiments were performed at $T_u = 423$ K. The *n*-butanol/air flames were investigated at $P_u = 1$–20 atm and the *n*-butanol/O_2/He flames were investigated at $P_u = 10$–20 atm. Each experiment was repeated 3–4 times and the average LBV value was reported. *n*-Butanol (≥99%) was provided by Aladdin Reagent. Synthetic air (21% O_2/79% N_2), oxygen (≥99.9%) and helium (≥99.999%) were provided by Nanjing Special Gas Factory. The oxygen/inert molar ratio was fixed at 0.14/0.86 for the