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Laminar flame speeds for n-butanol/air mixtures at elevated pressures and temperatures: An experimental and numerical study[†]

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Abstract

Laminar flame speeds of n-butanol/air premixed flames were measured experimentally and numerically at elevated pressures and temperatures for a wide range of equivalence ratios. Laminar flame speeds were obtained experimentally from the temporal evaluation of the flame front of spherically outwardly propagating flames at zero stress rate. The shadowgraph technique was employed to gain optical access to the constant volume combustion chamber. Flame propagation images were captured by a high-speed camera and MATLAB codes were used to process the images and calculate laminar flame speeds. Flame speeds have been calculated numerically using CHEMKIN-Pro based on a short reaction mechanism for n-butanol oxidation, which was derived from a previously published full reaction mechanism. Numerical predictions were in qualitative agreement with experimental data. The effects of initial pressure and temperature elevation were analyzed. Also, the effect of simultaneous elevation of initial pressures and temperatures is documented. For all experimental conditions, the maximum flame speed was found at around equivalence ratio 1.1. In general, flame speeds decreased with the elevation of initial pressure and increased with initial temperature elevation.

Keywords: Laminar flame speed; n-butanol; Short reaction mechanism; Shadowgraph technique; High pressure effect; High temperature effect

1. Introduction

Currently, the world largely depends on fossil fuels to produce energy. Most of the energy producing systems have been designed and developed based on fossil fuels' properties. But the increasing global energy consumption rate and rapid exploitation of conventional (fossil) fuels is driving researchers to pay more attention for devising reliable alternative fuel sources for the future. It is a major concern that newly proposed fuels should be compatible with conservational fuels. The efficiency and environmental effects of those fuels are notable aspects to study as well. In recent years, due to the usability of alcohols (methanol, ethanol, butanol), both as blending agents and pure form, numerous investigations have been conducted to find their fuel properties under different working conditions.

Better efficiency and low pollutants emission have been found for alcohol-gasoline blended fuels [1-4]. The high flame speed, high octane number and wide flammability range have added to the interests in conducting more studies on alcohols. Experiments have been conducted to determine fuel properties of 'methanol' and 'ethanol' [5, 6]. Accordingly, the engines and vehicles fueled by methanol and ethanol (both pure and hybrid) have been developed in different parts of the world such as Brazil, USA and China. Some considerable studies have been conducted on the combustion properties of 'butanol'. However, some significant areas (especially, flame propagation characteristics) have yet to be precisely investigated since this fuel possesses some notable properties; for instance, the energy density of iso-butanol is about 77% of gasoline.

Feng et al. [7] conducted an experiment to measure the laminar burning velocities of iso-butanol-air mixtures at different initial pressures, temperatures and equivalence ratios. Gu et al. [8] experimentally measured laminar flame speeds and markstein lengths of tert-butanol–air premixed flames at elevated pressures. A detailed experimental study on the effect of molecular structures of all four butanol isomers on laminar burning velocity was conducted based on the C-H bond dissociation energy by Gu et al. [9]. They concluded with a proper explanation that n-butanol has the highest burning velocity for any condition among these four isomers.

The laminar flame speed of n-butanol at atmospheric pressure was experimentally determined by Veloo et al. [10] in the counterflow configuration. Sarathy et al. [11] used three configurations to determine species' concentration profiles of n-butanol combustion at atmospheric pressure. They also deter-

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mined the premixed laminar flame speeds of n-butanol and provided comparison with n-butane flame speeds.

Wu et al. [12] experimentally determined laminar flame speeds and markstein lengths for all four butanol isomers at 1 atm and 2 atm pressures at 353 K. To get data at 5 atm they conducted experiments at 373 K. They validated their experimental data by calculating laminar flame speeds of n-butanol/air mixtures numerically by using a full reaction mechanism of n-butanol oxidation proposed by Sarathy et al. [13]. With the sole purpose of comparing different laminar burning characteristics of the butanol isomers, they also found that for different pressures n-butanol has the highest laminar flame speeds than other isomers.

Li et al. [14] reported the data of laminar flame speeds of 2-methyl-1-butanol (2 MB)/air mixtures at various pressures and temperatures. Gu et al. [15] again experimented on the effects of N_2 dilution in n-butanol/air mixtures on the laminar burning characteristics including burning velocity. Lee [16] derived a short mechanism for n-butanol oxidation. In this current work, we used his short mechanism to numerically calculate the laminar flame speeds of n-butanol/air mixtures at different initial pressures and temperatures.

Few other similar investigations have been conducted on n-butanol flame speed. But still, a well-established method to reliably predict laminar flame speeds of n-butanol/air mixtures at high initial pressures and temperatures with a simplified computational effort is not available. The primary purpose of this current work is to provide a much-simplified method to predict laminar flame speeds of n-butanol/air mixtures at high initial pressure and temperature than the existing ones and rigorously prove this numerical scheme with carefully obtained experimental data. As well as the effects of initial pressure and temperature elevation on n-butanol/air laminar flame speed are illustrated clearly in this work. The effect of simultaneous elevation of initial pressure and temperature on n-butanol flame speeds is also documented based on numerical and experimental data.

2. Experimental setup

The experimental setup was similar to Tahtouh et al. [17]. The shadowgraph technique was employed to observe the flame front propagation. The setup consisted primarily of a constant volume combustion chamber, air heating system, temperature and pressure measurement system and data acquisition system.

The combustion chamber, cylindrical on the inside (inner diameter 70 mm, length 70 mm, total inside volume 302.155 cm^3), was placed horizontally. The ignitions were initiated at the approximate center of the chamber by two horizontally placed electric spark-pins. The chamber was well capable of sustaining pressure as high as 10 atm at approximately 480 K. A less than 0.01 atm pressure drop, from any set pressure and temperature within 300 seconds, was considered as the 'proper state' to conduct experiments. The temperature inside the chamber was controlled by electric voltage. A maximum of ±2 K of fluctuation during experiments was allowed.

The air of the standard composition from a 'pressurized air cylinder' was supplied to the 'air heating cylinder' at the desired pressure and heated up by an electric conductor. The temperature of the 'air heating cylinder' was kept 2/3 K higher than the combustion chamber temperature (i.e., initial temperature for the experiment) to compensate for the heat loss during the supply of preheated air. All the connecting points of the air-transferring pipes were checked frequently to ensure that the system remained leak-free throughout the experiments.

The temperatures inside the chamber and in other parts were measured by the thermocouples of the temperature measuring unit (HEWLETT PACKARD 34970A data acquisition/switch unit). The pressures inside the chamber as well as the air heating cylinder were measured by a suitable pressure sensor (AEP transducers; capacity up to 20 bar).

Quartz windows were placed at both ends of the cylindrical chamber. An 'LPS-200X Arc Lamp Power Supply' was used as light source. Light from the pin-hole (200 μm diameter) of the source was focused inside the chamber by a lens. After light passed through the chamber, it was focused on the camera (Phantom Miro M310-6GB color camera; size: 640×480 at 10000 fps) by another lens.

A vacuum pump had been mounted within the setup to create a negative pressure inside the chamber when necessary (facilitating fuel injection and cleaning the chamber inside after combustions were completed). A plug valve was mounted on the chamber which facilitated manual fuel injection with 250/500 μl syringes.

The total experimental process for a single combustion took a maximum of 240 seconds. So, before every experimental attempt, leak testing was conducted to ensure that the system was not leaking for at least 300 seconds. Before injecting the fuel, negative pressure was developed inside the chamber by a vacuum pump, which ensured that all of the fuel was sucked into the chamber. Liquid fuel vaporized instantly since the inside temperature of the chamber was higher than the boiling point of n-butanol. After the fuel vaporized properly, preheated air from the 'air heating cylinder' (about the same temperature of the chamber) was supplied to the chamber through the one-way valves. With the help of the air inlet velocity and internal density difference the air and vaporized fuel were mixed. After injecting both fuel and air into the chamber, approximately 90-120 seconds was allowed to elapse to ensure mixture homogeneity.

The experimental setup is shown schematically in Fig. 1. Where, 1 → Light source; 2 & 4 → Lenses; 3 → Combustion Chamber; 5 → Camera; 6 → Data acquisition computer; 7 → Temperature measuring unit; 8 → Air heating cylinder; 9 & 14(V) → Voltage controllers; 10 → Pressurized air cylinder; 11 → Pressure releasing pipe; 12 → Vacuum pump; 13 → Ignition trigger; P → Pressure gauges/sensors.

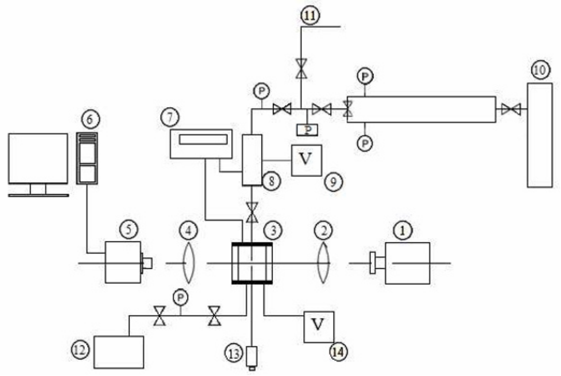
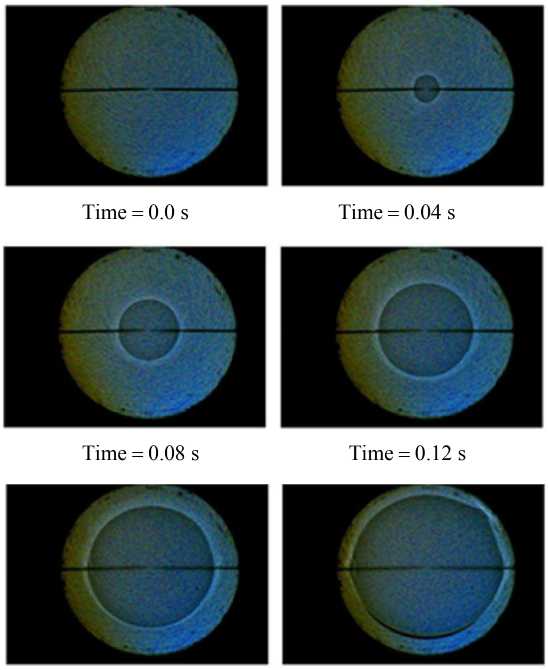


Fig. 1. Schematic diagram of experimental setup.



Time = 0.0 s Time = 0.04 s

Time = 0.08 s Time = 0.12 s

Time = 0.16 s Time = 0.20 s

Fig. 2. The shadowgraph images for the expanding flame (ϕ = 0.9, P = 5 atm, T = 400 K).

3. Laminar flame speed calculation methodology

The temporal evaluation of the laminar flame front is presented in Fig. 2; taken from one of the experimental attempts of this study.

During the temporal evolution, the flame front is affected by the stretch rate. Markstein [18] suggested the existence of a linear relationship between the local burning speed and local stretch, including the strain rate and curvature effects. The relation between the flame propagation and stretch rate was proposed by Clavin [19].

$$V_S = V_{S0} - L_b \cdot K \tag{1}$$

where V_S is the flame propagation speed in the stretched condition, it can be written as $V_S = \frac{dr_u}{dt}$; V_{S0} is the flame propagation speed in the unstretched condition, L_b represents the Markstein length for burned gas and $K = \frac{1}{A} \cdot \frac{dA}{dt}$ is the total stretch acting on the flame, which is the measure of the rate of change of a flame surface element of the area (A) with respect to time.

In this experimental case that is like a spherically outwardly expanding flame front, K can be presented as:

$$K = \frac{2}{r_u} \cdot \frac{dr_u}{dt}. \tag{2}$$

The laminar flame speed can be obtained from the unstretched laminar flame propagation speed and the density ratio for constant-pressure flame propagation as:

$$u_{l0} = \frac{\rho_b}{\rho_u} \cdot V_{S0}$$

where ρ_b is the density of burned gases and ρ_u is the density of unburned gases.

Tahtouh et al. [17] investigated three different methodologies to extract laminar flame information. In this current study, the second methodology investigated by Tahtouh was employed which was developed by Burluka et al. [20] using the least mean square, which only requires temporal radius evolution to extract laminar burning velocity. The methodology is as follows:

The substitution of Eq. (2) into Eq. (1), followed by integration, yields the following equation:

$$r(t) - r(t_0) + 2 \cdot L_b \cdot \ln\left(\frac{r(t)}{r(t_0)}\right) = V_{S0} \cdot (t - t_0) \tag{3}$$

The Markstein length, L_b, and the unstretched propagation speed, V_{S0}, are considered constant. For each time step, Eq. (3) must be verified. Thus, the problem consists of an optimization of the least squares function

$$\psi(L_b, V_{S0}) = \sum_{i=1}^{N} \left(t_i - t_0 - \frac{r_i - r_0}{V_{S0}} - 2 \cdot \frac{L_b}{V_{S0}} \cdot \ln\left(\frac{r_i}{r_0}\right) \right)^2 \tag{4}$$

where N is the number of time steps considered, with unknown values for L_b and V_{S0}.

The values of L_b and V_{S0} that minimize the previous equation are solutions of the following system:

$$\frac{d\psi}{dL_b} = 0 \tag{5}$$

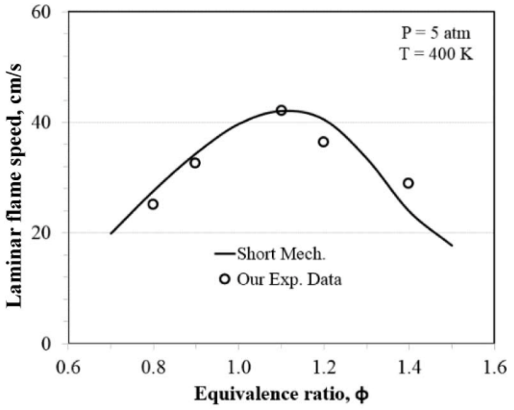


Fig. 3. Laminar flame speeds of n-butanol/air premixed flames at 5 atm, 400 K.

$$\frac{d\psi}{dV_{S0}} = 0 . \qquad (6)$$

Using MATLAB codes, the value of V_{S0} was calculated and then the value of u_{l0} was calculated.

4. Numerical modeling

The numerical simulations of freely propagating premixed flames were conducted using Chemkin-Pro [21]. The chemical kinetic scheme used a short reaction mechanism for the oxidation of n-butanol [16] derived from a full reaction mechanism which Sarathy et al. [13] proposed for the four butanol isomers. The short reaction mechanism included 78 gas-phase species, including argon and helium which behaves as an inert, and involved 661 forward reactions. The calculations involved 140~160 adaptively gridded points for a fine resolution in the flame domain.

5. Laminar flame speed

The flame front radius, r_u, obtained from the experiments at 400 K and 450 K were confined between 4 mm to 11 mm for later analytical use to avoid the ignition energy (electric spark) influence on the flame front propagation and to satisfy the consideration of constant pressure phenomena as well as to avoid the effect of cellularity on flame speed, as mentioned by Bradley et al. [22].

In Fig. 3, the laminar flame speeds of n-butanol/air flames at 5 atm and 400 K are presented for a range of equivalence ratios. The flame speed is around 25 cm/s for $\phi = 0.8$ and the value increased gradually with the equivalence ratio of up to approximately $\phi = 1.1$. The highest flame speed at this pressure and temperature was found at around $\phi = 1.1$, which is approximately 42 cm/s. For further rich mixtures, flame speed dramatically dropped with the equivalence ratio.

Fig. 4 depicts laminar flame speeds at 5 atm and 450 K. The change of the flame speeds with equivalence ratios occurred in a similar manner as the previous condition. In fact, this trend of changing laminar flame speed against the equivalence ratio can be explained by the conventional hydrocarbon fuels' theory. The effect of the initial temperature elevation on n-butanol/air flame speeds is observed by comparing this condition with the previous ones. For the initial temperature of 400 K (Fig. 3) the maximum flame speed was about 42 cm/s. When the initial temperature was elevated to 450 K (Fig. 4), the maximum flame speed reached approximately 52 cm/s.

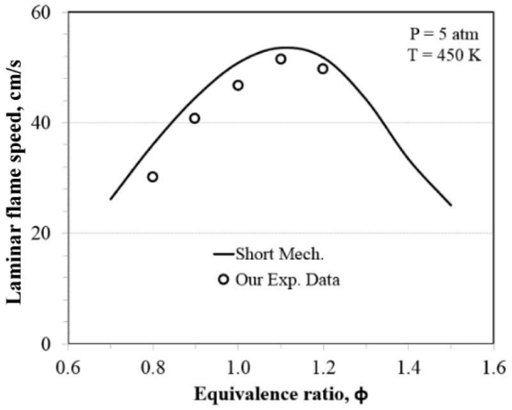


Fig. 4. Laminar flame speeds of n-butanol/air premixed flames at 5 atm, 450 K.

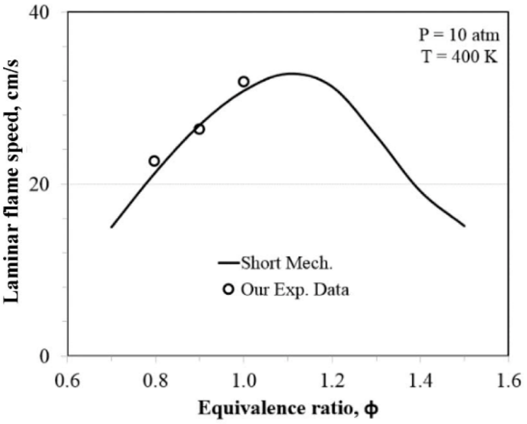


Fig. 5. Laminar flame speeds of n-butanol/air premixed flames at 10 atm, 400 K.

Fig. 5 and 6 present the laminar flame speeds at 10 atm and 400 K and 450 K, respectively. The maximum flame speed at 10 atm and 400 K was around 35 cm/s and the peak value at 10 atm and 450 K was around 45 cm/s. For both pressures, the flame speeds increased with the elevation of the initial temperature for all equivalence ratios. Obtaining data at 10 atm for fuel rich mixtures were very interruptive because of the many considerable experimental limitations. So, experiments were confined to the fuel lean mixtures for 10 atm.

For all of the above conditions, the numerically calculated values of the laminar flame speeds are also presented by a solid line in the figures. It is notable that the experimental results for all conditions are in sound agreement with the numerically calculated values. This alignment confirms the accu-

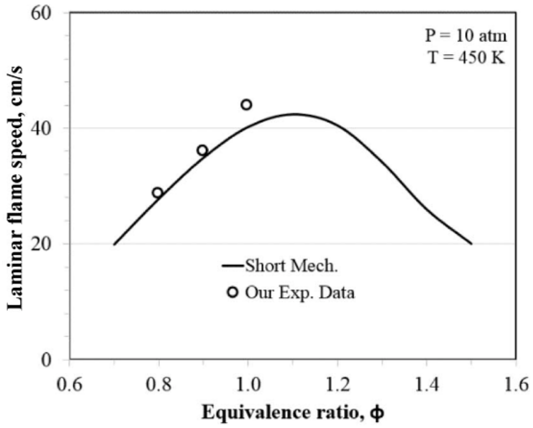


Fig. 6. Laminar flame speeds of n-butanol/air premixed flames at 10 atm, 450 K.

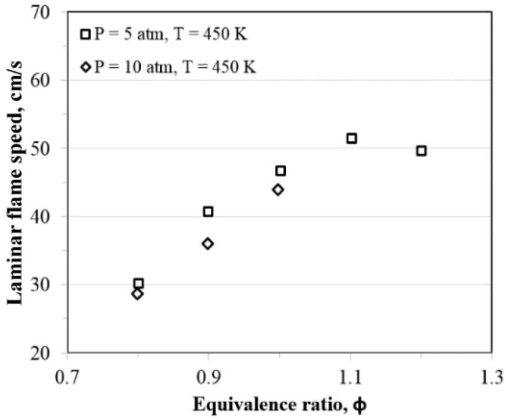


Fig. 8. Effect of initial pressure elevation on laminar flame speed at 450 K.

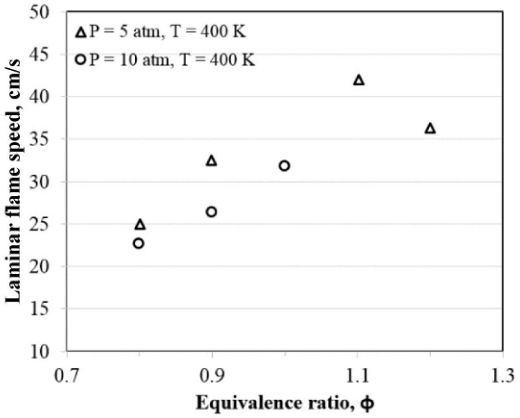


Fig. 7. Effect of initial pressure elevation on laminar flame speed at 400 K.

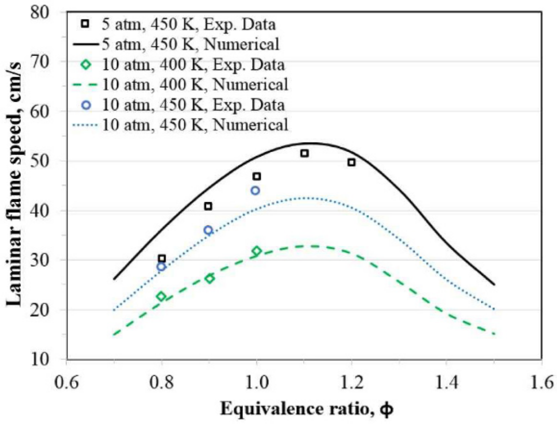


Fig. 9. Effect of initial pressure and temperature elevation.

racy of our numerical scheme to predict laminar flame speeds of n-butanol/air mixtures at considerably high initial pressures and temperatures.

To understand the effect of the initial pressure elevation, flame speeds at 5 atm and 10 atm at 400 K are plotted in Fig. 7 and flame speeds at 5 atm and 10 atm at 450 K are displayed in Fig. 8. For both temperatures, flame speeds reduced with the elevation of initial pressure for all equivalence ratios.

The effects of initial pressure and temperature elevation on laminar flame speeds of n-butanol/air mixtures can be more clearly observed by putting the results in a single plot (Fig. 9). Fig. 9 demonstrates that when initial pressure is elevated from 5 atm to 10 atm and keeping the initial temperature fixed at 450 K, a dramatic drop in the laminar flame speeds for all equivalence ratios is observed. On the other hand, when the initial temperature is raised from 400 K to 450 K at the same initial pressure of 10 atm, the laminar flame speeds for all equivalence ratios increased considerably.

To see the effects of the simultaneous elevation of initial pressure and temperature, the results from the current study are plotted with the experimental results provided by Wu et al. [12]. Wu experimentally measured laminar flame speeds of n-butanol/air mixtures at 1 atm and 353 K. The experimental flame speeds measured at 5 atm and 400 K and at 10 atm and 450 K in present work are plotted together with Wu's data in Fig. 10. The flame speeds are highest for all equivalence ratios at 1 atm at 353 K (Wu' data). The flame speeds at an initial pressure of 5 atm and initial temperature of 400 K are considerably lower than those of Wu's conditions for all equivalence ratios. The elevation of the initial pressure and temperature have opposite effects on flame propagation. For this range of simultaneous pressure and temperature elevation, the effect of the pressure elevation dominated over the effect of the temperature elevation. Thus, the flame speed of n-butanol drops. It was notable that the maximum flame speed reduced by approximately 10 cm/s for this range of simultaneous pressure and temperature elevation.

However, the flame speeds at the initial pressure of 10 atm and initial temperature of 450 K, for all equivalence ratios were higher than those of the 5 atm and 400 K initial condition. This observation suggests that for this range of simultaneous initial pressure and temperature elevation, the effect of

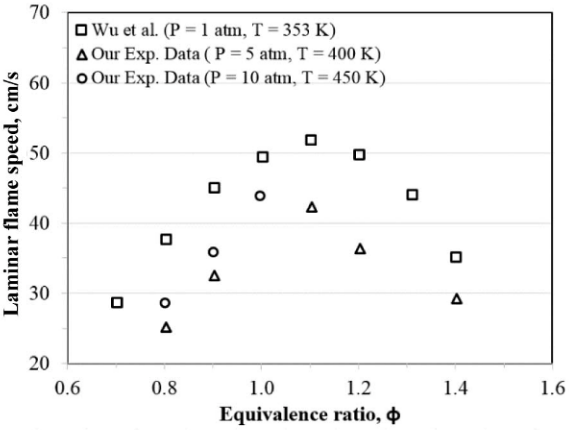


Fig. 10. Effect of simultaneous elevation of initial pressure and temperature on laminar flame speeds of n-butanol/air mixtures.

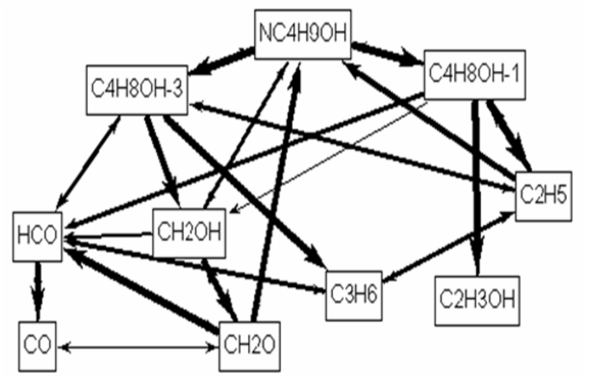


Fig. 11. Reaction pathway diagram for n-butanol/air flame at ϕ = 1, P = 5 atm and T = 1000 K.

temperature elevation on n-butanol combustion kinetics surpasses the effect of pressure elevation. The maximum increase of the flame speed from 5 atm at 400 K to 10 atm at 450 K is noted around 3 cm/s.

6. Numerical analysis

A reaction pathway analysis for the flame, which is at ϕ = 1, P = 5 atm and T = 1000 K, was performed to determine the most dominant pathways for n-butanol consumption. Fig. 11 presents the results of the analysis in a diagram format from n-butanol to CO with ten of the important species. n-butanol is primarily consumed through H-atom abstraction to produce C_4H_8OH-1 and C_4H_8OH-3. C_4H_8OH-1 is consumed to form CH_2OH, and then CH_2O. The consumption of C_4H_8OH-3 results in the production of C_2H_3OH. The results of the reaction pathway were in good agreement when compared with the reaction pathway diagram in the jet stirred reactor (JSR) at ϕ = 1, p = 10 atm, τ = 0.7 seconds, and T = 1000 K [23]. It is possible to use this short mechanism for the numerical simulation of flames operating under high pressure.

In experimental work, it is difficult to obtain the flame speeds for n-butanol/air flames which are formed in fuel-rich condition and are over P =10 atm. The reason is that richer mixtures have a higher likelihood of becoming unstable in rich conditions [24]. The flame speeds are calculated numerically with our short mechanism for flames that correspond to the extent of ϕ = 0.7~1.5 at T = 450 K. Fig. 12 displays the results of the flame speeds from 1atm to 10 atm. As expected, the flame speed decreased as the initial pressure increased.

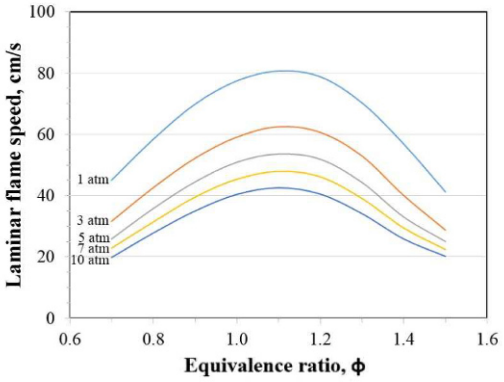


Fig. 12. Change of flame speed with equivalence ratio at different initial pressure.

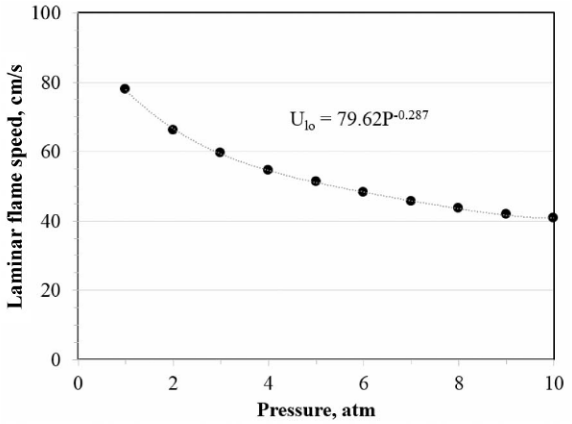


Fig. 13. Effect of pressure for n-butanol flames at stoichiometric condition.

Fig. 13 demonstrates the effect of pressure on n-butanol flames at ϕ = 1. The curve is the flame speed-pressure correlation which is obtained as the form of ‘a $(1/P)^{n'}$’ where ‘a’ is 79.62 and ‘n’ is 0.287. The correlation indicated a negative dependence of pressure which is similar to other hydrocarbon fuels. The laminar flame speed of n-butanol flames was less sensitive as the pressure increased.

7. Conclusion

The laminar flame speeds of n-butanol/air flames were measured experimentally and numerically. The high-speed shadowgraph technique was applied to observe flame front propagation of spherically outward propagating flames. Numerical analysis was also performed using the kinetic reaction scheme developed from a previously established full mechanism. The comparison between the experimental and numerical predictions showed qualitative agreement. Thus, the developed short mechanism was validated. It is possible to use this short mechanism for numerical simulation of flames operating under high pressure. The effects of the initial pressure and temperature on flame speeds have been illustrated. The laminar flame speed of n-butanol/air flames exhibits an inverse relationship with initial pressure like other hydrocarbons. On the other hand, flame speed increased with the initial reaction temperature. The effects of simultaneous initial reaction pressure and temperature elevation were also documented. The flame speed-pressure correlation was obtained as the form of $a(1/P)^n$ where 'a' is 79.62 and 'n' is 0.287.

Acknowledgment

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Nomenclature

V_S	: Stretched propagation flame speed
V_{S0}	: Unstretched propagation flame speed
L_b	: Markstein length for burned gas
K	: Total stretch acting on the flame
r_u	: Flame front radius
P	: Pressure
T	: Temperature
ϕ	: Equivalence ratio
ρ_b	: Density of burned gases
ρ_u	: Density of unburned gases
ψ	: Least squares function
τ	: Time

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