Non-linear variation in temperature exponent, $\alpha$

Measured laminar burning velocity up to 550 K for $\Phi = 1$

Stabilized planar flame for n-pentanol+air mixtures
Measurement of laminar burning velocity of n-Pentanol+Air mixtures at elevated temperatures and a skeletal kinetic model

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Abstract

Long chain alcohols are potential fuels for engine applications, however, their combustion characteristics need to be adequately investigated compared to short chain alcohols (C₁-C₄), especially at high mixture temperatures, and other conditions relevant to engine temperatures. In the present work, meso-scale diverging channel method has been used to measure the laminar burning velocity of n-pentanol+air mixtures at elevated temperatures due to existence of very limited data at higher mixture temperatures (~ 473 K). The present experiments are carried out at atmospheric pressure with unburnt mixture temperature varying up to 560 K. The dependence of laminar burning velocity on temperature was correlated using the power law: $S_u = S_{u,0} \left(\frac{T_u}{T_{u,0}}\right)^\alpha$, where $\alpha$ is the temperature exponent. The results show the existence of a minimum value of $\alpha$ for slightly rich mixtures. A reduced kinetic model based
on the previous detailed kinetic model of Sarathy (2014) for C\textsubscript{1} - C\textsubscript{5} straight-chain alcohols was generated with 199 species and 1427 reactions. Experimental results of laminar burning velocity of n-pentanol+air mixtures at high temperatures were compared with the present model and other kinetic models from the literature. The skeletal model accurately reproduces the measurements at various conditions.

**Keywords:** Pentanol-biofuel, laminar burning velocity, meso-scale channel, skeletal mechanism

1. **Introduction**

Sustenance on fossil fuels for future development appears challenging due to increasing demand for sustainable energy resources. The current consumption rate of fossil fuels contributes to greenhouse gas emissions, while also being unsustainable in the face of growing global demand. This motivates the search for alternative fuels that balances the long-term availability and ecological impact without compromising on the performance of combustion systems. In this context, biofuels promise to be an appropriate alternative in meeting the ever-increasing energy needs of the world. In 2011, the International Energy Agency focused on utilization of biofuels blends in transport sector to increase the worldwide biodiesel consumption to ten times the present usage by 2050 [1].

n-Pentanol and its isomers [2-4] are being investigated in various combustion engine [5] experiments as possible alternatives for fossil fuels, and they can be readily produced from various renewable sources [6, 7]. A study by Heufer et al. [8] suggests that reactivity of longer carbon chain alcohol fuels is equivalent to their corresponding alkanes. Compared to low carbon alcohols, n-pentanol has higher energy density, higher boiling point, lesser hygroscopicity and corrosiveness [9]. Gautam et al. [6] conducted experimental investigations on combustion characteristics of gasoline and gasoline blended with higher alcohols (n-propanol, n-butanol and n-pentanol). Their study indicated that alcohol/gasoline
blends showed higher flame speeds compared to pure gasoline, primarily due to increased oxygen content in the blended fuel due to addition of alcohol.

Campos-Fernandez et al. [10] compared the performance of compression ignition engines fueled with diesel and n-pentanol/diesel blends. They observed that the blended fuel showed significant improvement in combustion and brake thermal efficiencies against pure diesel, although, the torque, power and BSFC (Brake specific fuel consumption) remained nearly the same. They recommended a fuel blend of 25% n-pentanol with diesel as a potential substitute to improve the performance of IC (Internal Combustion) engines. Li et al. [11] compared the combustion and emission performance of n-pentanol with diesel using different injection schemes in a single-cylinder direct-injection diesel engine. They concluded that under certain operating conditions, besides increased detonation resistance, n-pentanol also shows simultaneous reduction in soot and NOx emissions. In a separate study [12], they reported engine and fuel performance characteristics of n-pentanol blends with diesel–biodiesel in the same engine configuration. The studies showed that n-pentanol blending leads to a significant reduction in soot and CO emissions coupled with higher ITE (Indicated thermal efficiency) and lower ISFC (Indicated specific fuel consumption) over a wide range of operating conditions.

Laminar burning velocity is a fundamental parameter used in the characterization of the combustion and emission performance of various fuel-air mixtures under different conditions. It contains important information about the reactivity, thermal diffusivity and exothermicity of the fuel. It is used in the validation of chemical kinetic mechanisms, development of new mechanisms and estimation of turbulent burning velocities. A fuel with a higher burning velocity can alleviate the requirements for complex injection patterns to increase the turbulent intensity [13]. For a given fuel-oxidizer mixture, its value depends on initial pressure, unburnt mixture temperature and equivalence ratio. For constant pressure conditions, this dependence
of laminar burning velocity on initial mixture temperature [14] can be described as: 

$$S_u = S_{u,0} \left( \frac{T_u}{T_{u,0}} \right)^{\alpha}$$

where $\alpha$ is the temperature exponent and its value depends on pressure and mixture equivalence ratio, $S_{u,0}$ is the laminar burning velocity at reference temperature and, $T_{u,0}$ is the reference temperature, taken as 300 K. The ideal conditions for measurement of laminar flame speeds are: a) Planar flame b) Zero flame stretch c) Adiabatic conditions. The laminar burning velocity data for gaseous fuels is available in abundance in literature, whereas the data for liquid fuels is scarce and shows huge scatter [15].

To authors’ knowledge, there are only four studies related to the measurement of the laminar burning velocity of n-pentanol+air mixture in the literature [9, 16-18]. Amongst these four studies, only one study reports the variation of temperature exponent. Togbe et al. [16] extracted the laminar burning velocity from spherically expanding flames coupled with a non-linear correlation for stretch correction presented by Kelly and Law [19]. The measurements were carried out at 423 K and 1 atm for an equivalence ratio range of $\Phi = 0.7 - 1.4$. Li et al. [17, 18] reported two studies for n-pentanol-air mixtures using spherically expanding flames method for a pressure range of 1 - 7.5 atm and initial temperatures 393 K, 433 K and 473 K for equivalence ratio range of $\Phi = 0.6 - 1.8$. In their first study [17], the authors reported burning velocities using linear extrapolation model of stretched flame speed to zero stretch. In their second study [18], the unstretched flame speed was evaluated using a non-linear expression proposed by Kelly et al. [20] for same conditions of temperature and pressure. Nativel et al. [9] also used the spherical flame method for mixture temperatures of 353, 433 and 473 K at 1 bar pressure and equivalence ratio range $\Phi = 0.7 - 1.5$.

The kinetic mechanism development for n-pentanol has received the attention of combustion researchers in past one decade [21]. Togbe et al. [16] developed a kinetic model (261 species and 2009 reactions) for n-pentanol using data from laminar flame speed experiments and species concentration profiles in a jet stirred reactor (JSR). Heufer et al. [8]
presented a kinetic scheme (599 species with 3019 reactions) for n-pentanol combustion by extending the chemistry of butanol isomers from previous studies in literature [22, 23]. Li et al. [17] modified the Togbe et al. [16] model to develop a new model (169 species and 1346 reactions). Kohler et al. [24] presented a detailed kinetic model for 1-, 2- and 3-pentanol+air mixtures (225 species and 24526 reactions). Liu et al. [25] have presented a skeletal alcohols model (161 species and 622 reactions) for various alcohol fuels. Park et al. [26] presented a skeletal mechanism for C5 alcohols for Homogeneous Charge Compression Ignition (HCCI) engine simulations.

The present work has two key objectives: i) Addressing the scarcity of laminar burning velocity data for n-pentanol+air mixtures at high temperatures by providing new experimental measurements up to a mixture temperature of 560 K using meso-scale diverging channel technique. This method has been used previously for high temperature burning velocity measurements of various gaseous and liquid fuels [27-33]. ii) Develop a skeletal model for n-pentanol from Sarathy et al. [21] high temperature alcohol model and compare its performance with existing models and current experimental data high mixture temperatures.

2. Experimental details and Uncertainty:
A quartz channel with a divergence angle of 10°, aspect ratio 12.5 and inlet cross section $25 \times 2 \text{ mm}^2$ was used in the present experimental setup as shown in Fig 1 and Fig 2. The width between the channel plates was kept fixed at 2 mm. The beginning of the diverging part is termed as the inlet. Air is metered using a thermal mass flow controller (AALBORG) with an accuracy of $\pm 1.5\%$ and flow velocity ranging from $20 \text{–} 80\%$ of full scale flow. Since the fuel is in liquid form at atmospheric conditions, it needs to be converted to vapor phase to facilitate complete mixing with air and subsequent combustion in the diverging part of the channel.

The air from the flow controller passes through a preheating segment to attain a temperature high enough for the creation of a gas phase mixture of fuel and oxidizer. Liquid fuel is dispensed using a syringe pump, since the volumetric flow rate of fuel is very low ($< 30 \text{ mlph}$) due to small volume of the combustion domain. n-Pentanol fuel is procured from HiMedia with a purity of 99%. The hot air jet from the preheater passes through a narrow tube of 2 mm inner diameter before impinging on the liquid fuel spray, thus leading to instant vaporization and mixing. The upper limit of equivalence ratio achievable at a particular temperature is determined from the saturation vapor pressure of the fuel at that temperature. The entire mixture line is kept heated and insulated using ceramic wool to avoid any chances of vapor condensation. The length of the diverging section is kept fixed at 50 mm. The channel is heated from bottom using an infrared heater to establish a steady positive temperature gradient in the direction of fluid flow and a uniform temperature distribution in
the transverse direction [34]. The temperature gradient can be altered by changing the horizontal and vertical clearance between the heater and the channel besides changing the input power to the heater. Quartz has high heat capacity and low thermal conductivity, which facilitates the formation of a linear temperature gradient along the direction of fluid flow and visualization of the stabilization flame. The temperature profile in the channel is measured using a 0.25 mm K-type thermocouple with an accuracy of ± 5 K. Since the combustion domain is small, the requirement of mixture flow rates is also low, which in turn leads to low Peclet numbers of the flow [35]. Therefore, the mixture temperature is assumed to be equal to the wall temperature at the flame stabilization location. Upon external ignition at the downstream end, the flame propagates inside the channel and a variety of flame dynamics are observed depending upon mixture flow rates and temperature gradient [36]. Planar flames (as shown in Fig. 3) are formed for certain conditions and these planar flames are used for extraction of laminar burning velocity. The measurements carry an uncertainty of ± 5% and a detailed discussion on same is reported earlier [27, 31, 34].

The laminar burning velocity is calculated using mass conservation principle at the mixture inlet and flame stabilization point \( S_{in} = U_{inlet} \left( A_{inlet} / A_f \right) \times \left( T_f / T_{inlet} \right) \). Where, \( T_{inlet} \) is the temperature of incoming mixture, \( A_{inlet} \) – channel inlet area and \( T_f \) - mixture temperature at flame stabilization location \( A_f \) - being the flame area.

![Figure 3. Direct image of planar stabilized flame for U_inlet = 0.7 m/s at Φ = 0.8](image)
3. Model Reduction and simulation procedure

In this work, the high temperature alcohol mechanism by Sarathy et al. [21] was used to simulate the experimental results. The detailed model has 354 species and 2462 reactions. In order to minimize computational time, this model was reduced using the method of direct relation graph with expert knowledge (DRG-X) [37]. As reported in previous studies, the method of DRG-X is based on the assumption that during combustion reactions, some species are weakly connected to others and barely contribute to the combustion process [37-40]. Such loosely coupled species can be removed from the mechanism, and still retain the chemical fidelity of the detailed model. In the present work, the detailed model was reduced for high temperatures with an error tolerance for heat release set at 0.01. The error tolerance for H and OH radicals is 0.3 and 0.1 respectively, while the default error tolerance for other species is 0.4. A resulting skeletal mechanism with 199 species and 1427 reactions was generated. The skeletal model was used to simulate the present experimental data. Simulations were performed with CHEMKIN-PRO, using the PREMIX solver, with thermal diffusion and mixture-averaged transport properties. A large number of grid points were used to completely resolve various temperature and species gradients within the flame (~ 1700), employing convergence parameters of GRAD = 0.01 and CURV = 0.03. GRAD and CURV are adaptive grid control parameters that control the extent to which the solution gradient and curvature is resolved. The computational time for premixed flame speed simulations was reduced by ≈ 60% with the skeletal model as compared to the detailed model.
The performance of the skeletal model was assessed by comparing the laminar burning velocity predictions and the associated temperature exponents with those from the detailed model, as shown in Fig. 4. The skeletal mechanism accurately reproduces the important trends related to these two important parameters: the peaking of the burning velocity at $\Phi = 1.1$ and existence of the minimum value of the temperature exponent at $\Phi = 1.1$. In terms of the magnitude of laminar burning velocity, the skeletal model predictions show an excellent match across all mixture equivalence ratios at low and elevated temperatures.

4. Results and discussions

4.1 Influence of initial temperature on laminar burning velocity

For a stoichiometric n-pentanol-air mixture, the variation of laminar burning velocity with the temperature ratio is plotted as shown in Fig. 5. The unburnt mixture temperature is normalized with a reference temperature $T_{u,0} = 300$ K. Figure 6 shows a similar variation in laminar burning velocity with temperature ratio for a lean ($\Phi = 0.8$) and rich ($\Phi = 1.2$) mixture conditions.

The present experimental results are shown as open triangles and compared with other results reported in literature as well for different mixture equivalence ratios and mixture
temperatures. The burning velocity is observed to increase with increase in temperature ratio. The predictions from three models: present skeletal model, Heufer et al. [8] and Liu et al. [25] are similar to each other. For all three equivalence ratios reported in Fig. 5 and 6, the model of Kohler et al. [24] predicts the highest values, and the mechanism of Togbe et al. [16] predicts lowest values of laminar burning velocities. The difference between the predictions of Kohler et al. [24] model and the present skeletal model, Heufer et al. [8] and Liu et al. [25] models decreases as the mixture equivalence ratio increases from lean to the rich regimes. The model of Li et al. [17] predicts values very close to Togbe et al. [16], albeit slightly higher as mixture equivalence ratio shifts from lean to stoichiometric case. For Φ = 1, the present experimental data falls within a close band of the predictions from Kohler et al. [24] mechanism on one end and with the predictions of other models (present skeletal model, Heufer et al. [8] and Liu et al. [25] models) at the other end. Table 1 gives a brief summary of correlations for the fitted data for different mixture equivalence ratios varying from Φ = 0.7 - 1.3. The difference between the experimental data obtained from linear (L) and non-linear (NL) stretch correction schemes used by Li et al. deviates by ≈ 1.8 cm/s across the range of reported temperature ratios. The difference in predictions from different mechanisms arises due to the variations in the kinetic parameters (rate constants, activation energies, third body efficiencies) and the types of reactions included or excluded in various models. Besides, this variation can be ascribed to the fact that, in literature, no accurate experimental data is available at higher mixture temperatures to validate and optimize various rate constants and hence the reaction models. Therefore, a noticeable scatter is evident with an increase in the mixture temperature.
Table 1: Power-law correlations between laminar burning velocity and temperature ratio for present experimental results.

<table>
<thead>
<tr>
<th>$\Phi$</th>
<th>Correlation</th>
</tr>
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<tbody>
<tr>
<td>0.7</td>
<td>$S_u = 0.2043 \times (T_u/T_{u,0})^{2.038}$</td>
</tr>
<tr>
<td>0.8</td>
<td>$S_u = 0.2971 \times (T_u/T_{u,0})^{1.8277}$</td>
</tr>
<tr>
<td>0.9</td>
<td>$S_u = 0.3259 \times (T_u/T_{u,0})^{1.7885}$</td>
</tr>
<tr>
<td>1.0</td>
<td>$S_u = 0.3789 \times (T_u/T_{u,0})^{1.7344}$</td>
</tr>
<tr>
<td>1.1</td>
<td>$S_u = 0.3726 \times (T_u/T_{u,0})^{1.7063}$</td>
</tr>
<tr>
<td>1.2</td>
<td>$S_u = 0.3687 \times (T_u/T_{u,0})^{1.7218}$</td>
</tr>
<tr>
<td>1.3</td>
<td>$S_u = 0.3043 \times (T_u/T_{u,0})^{1.8788}$</td>
</tr>
</tbody>
</table>

Figure 5. Laminar burning velocity of stoichiometric n-pentanol+air mixtures at elevated temperatures. L – linear stretch correction model correction and NL - non-linear stretch correction.
Figure 6. Comparison of laminar burning velocity for lean (Φ = 0.8) and rich (Φ = 1.2) n-pentanol+air mixtures with mechanisms and literature data.

4.2 Influence of mixture equivalence ratio on temperature exponent

Laminar burning velocity data at every equivalence ratio was fitted with power-law correlation to obtain the values of temperature exponent, and its variation is shown in Fig. 7. This variation of temperature exponent with equivalence ratio can be fitted using a cubic polynomial as, \( \alpha = 0.8028\Phi^3 + 0.1655\Phi^2 - 3.0711\Phi + 3.8135 \). The uncertainty in the temperature exponent was evaluated using the least squares procedure followed by Alekseev et al. [41]. The uncertainty in the temperature exponent can be attributed to two main factors:

i) The temperature range of measurements, dictated by the lowest temperature for flame stabilization and the highest temperature corresponding to the auto ignition point for the fuel (≈ 570 K). With a broader mixture temperature range, smaller uncertainty of temperature exponent data is obtained and vice-versa.

ii) Large number of data points within the studied temperature range further reduces the uncertainty in the obtained values of temperature exponent, \( \alpha \).

The present data is in good agreement with the predictions of the present skeletal model except for a rich case of \( \Phi = 1.3 \), where the present model slightly under predicts the \( \alpha \) value.
However, the predicted value is still within the experimental uncertainty limits. The Kohler et al. [24] model shows highest predictions of $\alpha$ values for $\Phi < 1.2$. The models of Li et al. [17] and Togbe et al. [16] show significantly higher values for rich mixtures as clear from Fig. 7. The Heufer et al. [8] model predicts lowest values and the associated trend line is relatively lower compared to the present skeletal model. Overall, except Kohler et al. [24] model, all models show close agreement with the present experimental data in the lean and stoichiometric mixture domain. However, for richer mixtures at $\Phi = 1.1$ and 1.2, the present data agrees well with predictions of Liu et al. [25] and Heufer et al. [8] models. The models of Togbe et al. [16] and Li et al. [17] show noticeable over-prediction in the $\alpha$ values. The temperature exponent values reported by Nativel et al. [9] follow closely the trend line predicted using Togbe [16] model in $\Phi = 0.8 - 1.1$ range. Beyond this mixture regime, the reported exponent values are in fair agreement with the present skeletal model.

![Figure 7. Comparison of the variation of temperature exponent with equivalence ratio for n-pentanol+air mixtures](image)

**4.3 Laminar burning velocity comparison at different temperatures**

The burning velocities were obtained from correlations listed in Table 1 and compared across a temperature range of 335 K to 550 K with model predictions and other experimental
data reported in literature, as shown in Figs. 8 and 9. Li et al. [17, 18] and Nativel et al. [9] both reported stretch corrected data using linear and non-linear methods. Here, only the non-linear data set from their work has been used for comparison. At 335 K, all models except the Li et al. [17] and Togbe model [16] show an excellent agreement for all mixtures except at Φ = 1.1 and 1.3. The Kohler model [24] significantly overpredicts the laminar burning velocity values with increase in unburnt mixture temperature. Togbe et al. [16] model overestimates the measurements at 423 K, since their reported values fall within the range of other measurements at 393 K, as reported by other researchers and the present results. The measurements of Li et al. [17] agree well with the present results, the measurements by Nativel et al. [9] and with skeletal model predictions until Φ = 1, beyond which under-prediction becomes pronounced, especially at 433 K and 473 K. It is to be noted that the present skeletal model performs well across the complete mixture equivalence ratio range and temperature conditions tested.

Figure 8: Comparison of laminar burning velocities of n-pentanol+air mixtures at different mixture temperatures with various mechanisms
Figure 9: Comparison of laminar burning velocities of n-pentanol+air mixtures across temperature range with literature data

4.4 Sensitivity and Reaction pathway analysis

To assess the variation in laminar burning velocity from a kinetics perspective, with varying temperature and mixture equivalence ratios, the normalized sensitivity coefficients of the laminar burning velocity with respect to the reaction rate constant were plotted using the skeletal model as shown in Fig. 10. As the mixture temperature is increased from 335 K to 570 K, the magnitude of the sensitivity coefficient is observed to decrease. The chain branching reaction $H + O_2 \rightleftharpoons O + OH$ shows the highest contribution towards flame acceleration through the generation of H and OH radicals. The sensitivity of this reaction increases as the mixture moves from lean to the rich regimes. The next important reaction for flame speed is the CO oxidation reaction: $CO + OH \rightleftharpoons CO_2 + H$. The contribution of this reaction is highest on the lean side and decreases as the mixture becomes richer. The formyl radical decomposition reaction: $HCO + M \rightleftharpoons H + CO + M$, shows nearly equal contributions across all equivalence ratios. The main flame retarding reaction: $H + O_2 (+M) \rightleftharpoons HO_2 (+M)$ shows contributions for lean and stoichiometric mixtures only. Two reactions: $HO_2 + H \rightleftharpoons$
OH + OH and HCO + H ⇌ CO + H₂ exhibit temperature bias wherein the contribution towards burning velocity is evident at only one of the mixture temperatures.

Figure 10: Normalized sensitivity coefficients for the laminar burning velocity of n-pentanol + air mixtures at (a) 335 K and (b) 560 K

In addition to this, a reaction path-flux analysis was carried out at 350 K and 560 K to compare how the fuel is consumed at these conditions (high and low temperatures). The result is shown is Fig. 11. The analyses showed that the fuel decomposes almost in a similar manner at both of the mixture temperatures. The magnitude of initial fuel abstraction is very close, although, notable differences occur in the percentage flux of some certain pathways leading to the formation of more active radicals at 560 K and less active radicals at 350 K. For example, 61% of nC₅H₁₀OH₁-3 reacts to produce butene (C₄H₈-1) radicals at 550 K, while around 74% of nC₅H₁₀OH₁-3 produces it at 350 K. Also, more of ethylene (C₂H₄) is converted to vinyl acetaldehyde (CH₂CHO) and HCO radicals at 560 K than at 350 K; these are active radicals that can abstract more fuel. Please see supplementary material for species nomenclature.
Figure 11. Reaction path-flux analysis comparing the consumption pathways of n-pentanol at 350 K and 560 K. Numbers in black show percentage at 560 K, and those in blue show percentage at 350 K.

5. Conclusions

New experimental data on the measurement of laminar burning velocities of n-pentanol+air mixtures at higher mixture temperature were reported up to 560 K at 1 atm pressure using meso-scale diverging channel method. Present data is beyond the maximum unburnt mixture temperature of 473 K reported earlier in literature. Laminar burning velocity was observed to increase with an increase in unburnt mixture temperature. The burning velocity predictions
from various mechanisms show a scatter as high as 22% due to lack of experimental data in literature for kinetics validation at high temperatures. The temperature dependence of laminar burning velocity on equivalence ratio was assessed using power-law correlations. The temperature exponent exhibits an inverted parabolic variation with a minimum value at $\Phi = 1.1$. A skeletal kinetic mechanism based on a detailed kinetic model of Sarathy et al. [21] was developed and used for validation of the experimental measurements. The model captures the laminar burning velocity of the tested fuel at all temperature conditions very well. Detailed reaction sensitivity analysis and path-flux analysis helps obtain more insight into the flame propagation behavior of n-pentanol+air mixtures at different temperatures.

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Appendix A. Supplementary data
The present work includes the following supplementary mechanism: The skeletal kinetic model in CHEMKIN format along with the associated thermochemistry, and transport data. A species dictionary for those species discussed in the paper.

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