Cell formation effects on the burning speeds and flame front area of synthetic gas at high pressures and temperatures

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HIGHLIGHTS

• Effect of cell formation on burning speed and flame surface area is investigated.
• A new developed non-dimensional number called cellularity factor is introduced.
• Cellular burning speed and mass burning rate are calculated using differential based multi-shell model.
• Flame instability is studied using thermo-diffusive and hydrodynamics effects.
• Power law correlations are developed for cellular burning speeds and mass burning rates.

ABSTRACT

Cellular burning speeds and mass burning rates of premixed syngas/oxidizer/diluent (H2/CO/O2/He) have been determined at high pressures and temperatures over a wide range of equivalence ratios which are at engine-relevant conditions. Working on high pressure combustion helps to reduce the pollution and increase the energy efficiency in combustion devices. The experimental facilities consisted of two spherical and cylindrical chambers. The spherical chamber, which can withstand high pressures up to 400 atm, was used to collect pressure rise data due to combustion, to calculate cellular burning speed and mass burning rate. For flame structure and instability analysis the cylindrical chamber was used to take pictures of propagating flame using a high speed CMOS camera and a schlieren photography system. A new differential based multi-shell model based on pressure rise data was used to determine the cellular burning speed and mass burning rate. In this paper, cellular burning speed and mass burning rate of H2/CO/O2/He mixture have been measured for a wide range of equivalence ratios from 0.6 to 2, temperatures from 400 to 750 K and pressures from 2 to 50 atm for three hydrogen concentrations of 5, 10 and 25% in the syngas. The power law correlations for cellular burning speed and mass burning rate were developed as a function of equivalence ratio, temperature and pressure. In this study a new developed parameter, called cellularity factor, which indicates the cell formation effect on flame surface area and burning speed has been introduced. The total flame surface area and cellularity factor for syngas at high pressures and temperatures have been calculated by combining the multi-shell model via the experimental pressure data with free flat flame simulation using detailed chemical mechanism. The results show that the
cellularity factor has a positive relation to pressure, equivalence ratio and hydrogen concentration while it has a negative dependency to temperature.

1. Introduction

A study to comprehensively measure and investigate the cellular burning speed, mass burning rate and cell formation effect of synthetic gas (syngas) at high pressures and temperatures is relevant and necessary to the scientific and combustion community [1–4]. Syngas, a mixture of H2 and CO, has gained importance as an alternative fuel for stationary gas turbines and internal combustion engines. Power plants have been using syngas for more than a decade, citing increased energy efficiencies and less emissions than conventional coal fired plants. Syngas is also increasingly being used in petroleum refineries to help produce cleaner transportation fuels and improve overall efficiency of the plant [5]. Syngas can be derived from the gasification of coal or biomass, including municipal waste, agricultural residue, and herbaceous energy crops, therefore, reducing greenhouse gas (GHG) emissions. Syngas is also a primary input of Fischer-Tropsch (F-T) reactors to produce gas-to-liquid (GTL) fuels [6,7].

Research studies into understanding the cellularity effect on burning speed for syngas fuel, are extremely relevant particularly for use in engineering combustion modeling, for stationary turbine based power plants and for internal combustion engines in the transportation industry [8–11]. Mass burning rate is the rate at which a combustible mixture is consumed by flame front and is also a measure of the energy release during a combustion process. When the flame front is laminar and its area is simply obtainable a more commonly used term is laminar burning speed [7,12–18].

Syngas mixtures have been studied and researched at atmospheric and elevated conditions. Hassan et al. [19] measured the laminar burning speed of various hydrogen to carbon monoxide ratios, sub-atmospheric to elevated pressures (0.5–4 atm), atmospheric temperature and wide range of equivalence ratios (0.6–5.0) in a spherical combustion chamber. Natarjan et al. [20] used the burner and particle velocimetry technique to measure laminar burning speed for 5:95, 50:50, and 95:5 syngas percentages diluted with CO2 and later extended measurements to higher pressures with helium substitution to reduce flame instability [21]. Vu et al. [22] compared the effects of CO2, N2, and He as diluents on the cellular instabilities in syngas flames in a cylindrical chamber at elevated pressures for a 50:50 hydrogen to carbon monoxide ratio. Han et al. [23] measured laminar burning speeds for various CO2 diluent percentages (10%–40%) at elevated temperatures and pressures for equivalence ratios of 0.8 and 1.0 using a dual-cylindrical setup. Askari et al. [24] measured the laminar burning speeds of syngas/air mixtures using a new differential-based multi-shell model over a wide range of temperatures (298 K up to 617 K), pressures (from sub-atmospheric up to 5.5 atm), equivalence ratios (0.6–5) and three different hydrogen concentration of 5%, 10% and 25%. Recently Askari et al. [25] investigated the effect of synthetic EGR addition on flame morphology and laminar burning speed of syngas/air mixture for a wide range of equivalence ratios, temperatures and pressures. A comprehensive literature search of burning speed measurements of all syngas composition exposed various gaps in the experimental data especially at elevated pressure and temperature conditions [21,26–29]. Some data exist but doesn’t comprehensively capture the whole range of conditions between 2–50 atm and 400–750 K, with varying degree of stoichiometry and gas mixture compositions. Despite internal combustion engines being subjected to high pressures, few studies have been performed under comparable conditions. In a 2014 review of syngas research, Lee concluded that new measurements on the burning speed were needed at elevated pressures [30]. At high pressure conditions (higher than 2 atm) the syngas/air flame is cellular and calculating the laminar burning speed due to lack of exact flame surface area is impossible. So instead, the other parameters such as cellular burning speed and mass burning rate which are very useful in the modeling of combustion systems can be calculated.

In this paper, cellular burning speed and mass burning rate of syngas/oxygen flame which was diluted with helium [31], H2/CO/O2/He, were calculated for a wide range of equivalence ratios from lean to rich (0.6–2), temperatures from 400 to 750 K and pressures from 2 to 50 atm. The structure and effect of thermo-diffusive and hydrodynamic instabilities were studied at very high pressures at which the flame is always cellular. Power law correlations as a function of equivalence ratio, temperature and pressure for cellular burning speed and mass burning rate have been developed. The effect of cell formation on burning speed and total flame front area has been investigated in terms of a newly developed parameter, called cellularity factor. In addition to a complete experimental data, the theoretical laminar burning speeds via a steady one-dimensional laminar premixed free flame code from CANTER package [32] in conjunction with Davis et al. mechanism [33] were calculated and used to determine the effect of cellularity.

2. Experimental setup and procedures

The core component of the experimental setup includes a spherical combustion chamber that enables the measurement of the pressure rise from a combustion process at high pressures and temperatures. This chamber was mainly used to measure the pressure rise for calculating the burning speed and mass burning rate. The spherical chamber was designed to withstand pressures up to 400 atm and was located inside an oven which can be heated up to 500 K. The second component of the experimental setup is a cylindrical chamber with optically clear sides which enables visualization of flame propagation for the study of flame structure and instability analysis. The cylindrical chamber with optical side ports and all its supporting system were rigidly mounted on an optical bench. Using a focused LED light source and a series of mirrors the parallel light was guided through the optically clear Quartz windows and was reflected to a high speed CMOS camera capable of capturing images up to 40,000 frames per second. This schlieren photography method which works based on density gradient [34], is useful in visualizing flame propagation to investigate flame front structure for instability analysis. The flame appears to be relatively affected by cylindrical chamber geometry for large radii since the flame shape is naturally spherical. It may cause some unwanted errors in laminar burning speed calculation [35]. Due to that reason the cylindrical chamber is just used for capturing flame propagation images which can be used in instability study. But for laminar burning speed measurement we just took advantage of our spherical chamber. A Kistler 601CA high-temperature pressure transducer in conjunction with a Kistler 5010B charge amplifier was used to record the dynamic pressure rise during flame propagation processes in both chambers. The filling manifold mainly constructed of 304 and 316 stainless steel Swagelok componentry and tubing is predisposed to accommodate inputs from 5
simultaneous gas cylinders, thus allowing for an inventory of gas mixtures to be always readily available for immediate use. Two vacuum pumps help evacuate the system faster and allow for independent testing of the two chambers. The composition of premixed fuel inside the chambers was always checked and verified using a gas chromatography (GC) system. The configuration of experimental facilities and their connections are shown in Fig. 1.

All the data acquisition and analysis can be done on-site by several dedicated PCs using a high speed LabVIEW DAQ card and isolated input and output modules for temperature, voltage and pressure measurements as well as automatic delayed firing via control of the high voltage coil driving the spark plugs. The PID temperature controllers are proportionally programmable with redundant safety feature for over temperature protection and electric shock. The raw pressure signal produced by the high speed pressure transducer contains digitization noise resulting from data acquisition system process of converting a voltage signal to a digital value. The digitization noise is approximately 0.25 psi/bit which is certainly negligible for the range of pressure measurements of interest. To minimize other experimental or human errors a data selection criteria was developed. For any set of data to be deemed acceptable three consecutive experimental runs have to yield the exact pressure curve to ensure that the confidence level of the experiments was always greater than 95% [36]. More information about the experimental setup can be found in previous publications [6,12,13,24,37–41].

3. Theoretical model

The numerical model used in this work to calculate burning speed and mass burning rate from the pressure rise is based on a newly developed differential based multi-shell model by Askari et al. [24]. The schematic of this theoretical model representing all available energy transfer and a typical temperature distribution is shown in Fig. 2. In the model, it is assumed that the reactants are initially quiescent and have a uniform composition, pressure and temperature. The fuel/air/diluent mixture inside the chamber is ignited at the center of the combustion chamber using two extended spark electrodes. All gases are assumed to be ideal gases.

The model divides the chamber into unburned and burned gas sections which are separated by a reaction zone of negligible thickness. The burned gas region is divided into a number of shells, each shell with a uniform temperature different from other shells and in local thermodynamic equilibrium. Burned gases are in chemical equilibrium in each shell. Composition of burned gases in each shell is determined by using CANTERA [32] subroutines combined with the latest version of thermochemical properties of NASA correlation [33]. Twenty major species (H2, N2, O2, CO, H, O, OH, HCO, HO2, H2O, H2O2, CO2, NO, NO2, CH2, CH3, CH4, CH2O, CH3OH and C2H2) have been considered in modeling. These twenty species are the ones with concentrations higher than 10^{-12} based on local thermodynamic equilibrium assumption, other species which are less than 10^{-12} neglected in this study. Surrounding the outermost burned gas shell is reaction zone with infinitesimal small thickness followed by a layer of unburned gas called the preheat zone that has a non-uniform temperature. The preheat zone is surrounded by the core unburned gas with uniform temperature. Pressure is assumed to have no spatial gradient in the chamber at any particular instant in time. All the energy losses to the electrodes, chamber walls, radiation from the burned gas, and energy transfer between neighboring shells have been added to the current model. The effect of energy transfer in the chamber wall and spark electrodes as well as preheat zone is modeled by the thermal boundary layer and displacement thickness concept [42]. Using this model, burning rates can be calculated over a wide range of temperatures, pressures and fuel air equivalence ratios at very high temperatures and pressures. In this model governing equations of unknown variables have been defined by a set of nonlinear ordinary differential equations that can be solved using the CVODE solver [43].
The governing equations are derived by applying the differential form of mass and energy balance equations all over the shells as well as the whole chamber. The energy balance equations for unburned, burned, and currently burning gases are respectively:

$$\dot{U}_u = -\dot{m}_b h_u + \dot{Q}_u - W_u$$  \hspace{1cm} (1)$$

$$\dot{U}_b = \dot{Q}_j - W_j, \hspace{0.5cm} j = b_1 - b_n$$  \hspace{1cm} (2)$$

$$\dot{U}_{bn} = \dot{m}_b h_u + \dot{Q}_{bn} - W_{bn}$$  \hspace{1cm} (3)$$

where $U$ is the internal energy, $\dot{m}_b$ the mass burning rate, $Q$ the energy transfer and $W$ the work. In these equations the subscript $j$ refers the already burned shells, $u$ and $b$ denote the unburned and burned gas conditions respectively, $n$ and $b_n$ denote the total number of shells in burned section and currently burning shell, respectively. The dot sign on top of the parameters refers to the complete derivative with respect to time. Using concept of displacement thickness [42] and thermodynamic relations [44] Eqs. (1)–(3) expand into the following format:

$$T_u = \frac{A^u_u}{B_u^u} \dot{p} + \frac{\rho_u}{B_u^u} V_{dis} \dot{x}_u + \rho_u h_u V_{disu} + \dot{Q}_u$$  \hspace{1cm} (4)$$

$$T_j = \frac{A^u_j}{B_j^u} \dot{p} + \frac{\rho_j}{B_j^u} V_{dis} \dot{x}_j, \hspace{0.5cm} j = b_1 - b_n$$  \hspace{1cm} (5)$$

$$T_{bn} = \frac{A^u_{bn}}{B_{bn}^u} \dot{p} + \left( m h_u h_n \right) V_{dis} \dot{x}_n + \rho_n h_n V_{disn} + \dot{Q}_{bn}$$  \hspace{1cm} (6)$$

where $\rho$ is the density, $h$ the enthalpy, $x_n$ the burned gas mass fraction, $V_{dis}$ the displacement thickness, $m$ the total mass, $p$ the experimental pressure data and the superscript $\infty$ denotes the region far from the thermal boundary layer with uniform temperature distribution. All the energy transfer terms in Eqs. (4)–(6) including conduction and radiation can be calculated using proposed formulations in Askari et al. [24].

The parameters $A^u_u, B^u_u, C^u_u, D^u_u, E^u_u(k = u, b_1, \ldots, b_{n-1}, b_n)$ and $F$ are defined in terms of thermodynamic properties [45] which can be found in Askari et al. [24]. Eqs. (4)–(7) form a set of nonlinear ordinary differential equations which contain $n + 3$ unknowns: $p(t), x_n(t), T_u^\infty$ and $T_b^\infty$ ($1 = 1$ to $n$). Given experimental pressure as a function of time, the equations can be solved numerically to find burned mass fraction and temperature distribution. Finally, the mass burning rate and cellular burning speed are calculated as Eqs. (8) and (9), respectively:

$$\dot{m}_b = \dot{m}_x$$  \hspace{1cm} (8)$$

$$S_c = \frac{\dot{m}_b}{\rho_b A_l}$$  \hspace{1cm} (9)$$

where $\rho_b$ is the unburned gas density and $A_l = 4\pi r_f^2$ is the equivalent spherical flame area having the same burned gas mass, which is called hereafter laminar flame area.

The initial mixture composition for $H_2/CO/O_2/He$ is defined as:

$$\phi \left( xH_2 + (1 - x)CO \right) + 0.5O_2 + 3.76He$$  \hspace{1cm} (10)$$

where $x$ is the hydrogen concentration of the fuel blend and $\phi$ is the equivalence ratio. For predicted laminar burning speed, a steady one-dimensional laminar premixed free flame code from the CANtera in conjunction with detailed chemical kinetics mechanism and multi-component diffusion model has been used to solve the balance equations of mass, energy, and species. In this paper, Davis et al. mechanism [33] for syngas combustion has been selected due to its good agreement with experimental results [24]. Combination of the systematic (Bias) and random (Precision) uncertainty [46] in conjunction with the root-sum-square method [38] give us the overall uncertainty for calculated cellular burning speeds and mass burning rates. The overall uncertainty varies between ±1% and ±3% with the average of ±1.51%. This range of uncertainty is only valid for conditions which are covered in our experiments and numerical analysis. For other conditions which are estimated by power law correlations such as atmospheric conditions the mean uncertainty is ±2.76%.

4. Results and discussion

4.1. Flame structure and instability study

The syngas fuel was combusted for a wide range of equivalence ratios of 0.6–2.0, in a lower thermal capacity air (21% O$_2$ and 79% He). By substituting the nitrogen in air with another inert gas such as helium, the total heat capacity of the mixture is reduced, therefore for the same energy of combustion the flame temperature and consequently mixture pressure are increased. The initial pressure conditions were set to 2, 5, 10 atm and initial temperatures to 400 and 450 K. The effects of equivalence ratio and flame propagation time on flame structure and instability of the flame front are shown in Fig. 3. This figure shows the snapshots of the expanding spherical flame of $H_2/CO/O_2/He$ mixture with varying the equivalence ratios at initial temperature of 400 K, initial pressure of
5 atm and hydrogen concentration of 5%. As it can be seen in this figure, the flame front is unstable and fully cellular during its propagation from the center of chamber until it hits the chamber walls for all three different equivalence ratios of 0.6, 1.0 and 2.0. It is also shown in this figure that the flame propagation rate increases as equivalence ratio increases.

The effect of thermo-diffusive and hydrodynamic instabilities in terms of effective Lewis number and flame thickness respectively, are shown in Fig. 4. The flame thickness is defined by 

\[
\delta_f = \frac{T_{ad} - T_u}{(dT/dx)_{\max}}
\]

where \(T_{ad}\) is the adiabatic flame temperature, \(T_u\) the unburned temperature and \((dT/dx)_{\max}\) the maximum temperature gradient.

The effective Lewis number for a wide range of equivalence ratio can be computed by the following equation [24]:

\[
Le_{\text{eff}} = \begin{cases} 
Le_{\text{fuel}} = \frac{D_i}{\rho_i c_p} & \phi \leq 0.8 \\
Le_{\text{fuel}/O_2} = 1 + \frac{(Le_{O_2} - 1)}{1 + A} & 0.8 < \phi < 2 \\
Le_{O_2} = \frac{D_i}{\rho_i c_p} & \phi \geq 2
\end{cases}
\]

where \(D_i = \frac{\lambda_i}{\rho_i c_p}\) is the mixture thermal diffusivity, \(D_{\text{fuel}}\) the reactant-inert binary diffusion coefficient that were found using Davis et al. mechanism [33] and \(x_i\) the volumetric fraction of the \(i^{th}\) component of the fuel blend [24].

The effective Lewis number in Fig. 4 decreases as equivalence ratio and flame propagation time increase. But the effective Lewis numbers are always greater than unity which makes the flame independent from thermo-diffusive instability. It means that flame cellularity due to increase in equivalence ratio and flame propagation time is not related to thermo-diffusive instability. As shown in Fig. 4, the higher the equivalence ratio, the lower the flame thickness, which promotes the effect of hydrodynamic instability [16]. On the other hand increase in flame propagation time promotes the hydrodynamic instability through the increase in pressure and flame radius and reduction in flame thickness. The hydrodynamic instability makes a laminar and smooth flame unstable by creating cells and wrinkles all over the flame front. These cells and wrinkles increase the contact area between unburned gas and flame which leads to increase of the mass entrained into the flame and consequently increase the mass burning rate. An in-depth analysis about cell formation of H₂/CO/O₂/He mixture in terms of temperature, pressure, equivalence ratio and hydrogen concentration is given in the next section.

4.2. Cell formation analysis

Askari et al. [24] showed that increase of hydrogen concentration, pressure and equivalence ratio in the range of 0.6–2, promote...
flame destabilization propensity and enhance the chance of cell formation over the flame front. Cellularity changes flame structure from a highly spherical, laminar and smooth flame to an unstable flame with large number of cells and wrinkles in different sizes all over the flame front. Therefore, the total exposed area of the unburned gas with the flame or simply the total flame area increases rapidly and deviates from a laminar flame area which is only a function of flame radius. In this case the total flame area becomes dependent to shape and size of the cells. The mass burning rate can be evaluated in two different methods. The first, is using the total flame area:

\[ m_b = \rho_u A_f S_f \]  
(12)

where \( \rho_u (T_u, p) \) is the unburned gas density, \( A_f (r_f, \xi) \) the total flame area, \( S_f(T_u, p, \phi) \) the laminar burning speed, \( r_f \) the flame radius, \( T_u \) unburned gas temperature, \( p \) the mixture pressure and \( \phi \) the fuel air equivalence ratio. The parameter \( \xi \) which is called hereafter, Cellularity Factor includes all information about the shape and size of cells over the flame front. In simple words, cellulurity factor is a non-dimensional number which shows difference between smooth and cellular flame surface area. Since the surface area of a cellular flame cannot be measured directly, at least with current technologies, the laminar burning speed cannot be calculated by previously discussed theoretical model using pressure rise method. The other method is, to approximate the flame surface as the outer area of the spherical flame front having the same mass of burned gas:

\[ \bar{m}_b = \rho_u A_f S_f \]  
(13)

where \( S_f(S_f, \xi), \) the cellular burning speed, is a function of laminar burning speed and cellulurity factor. Since the mass burning rate is known, so the under-estimation of flame surface area will be compensated by increase of the burning speed. This burning speed which includes the information of cellulurity is called hereafter the cellular burning speed. As discussed earlier, laminar burning speed cannot be evaluated in cellular flames unless further knowledge regarding the shape and area of the cellular flame becomes available, however cellular burning speed and mass burning rate can be measured and evaluated using the same model and assumptions. To reduce the effects of spark energy discharge, the results are reported in regions where \( r_f > 4 \text{ cm}. \) The measurements were taken at various equivalence ratios, pressures and temperatures and calculated cellular burning speeds and mass burning rates have been used to develop the correlations in terms of equivalence ratio, temperature and pressure as shown in Eqs. (14) and (15) respectively:

\[ S_f = S_{0f} \left( 1 + a(\phi - 1) + b(\phi - 1)^2 \right) \left( \frac{T}{T_0} \right)^{\frac{\gamma}{2}} \left( \frac{p}{p_0} \right)^{\frac{\gamma}{2}} \]  
(14)

\[ m_b = \bar{m}_b \left( 1 + c(\phi - 1) + d(\phi - 1)^2 \right) \left( \frac{T}{T_0} \right)^{\frac{\gamma}{2}} \left( \frac{p}{p_0} \right)^{\theta} \]  
(15)

where \( S_{0f} \) and \( \bar{m}_b \) are the cellular burning speed and mass burning rate respectively at reference state \((\phi = 1, T_0 = 298 \text{ K}, p_0 = 1 \text{ atm})\), \( \phi \) the equivalence ratio, \( T \) the temperature in K and \( p \) the mixture pressure in atm. To have an accurate fitting, the temperature and pressure exponents are considered as a linear function of equivalence ratio as shown in Eqs. (16)–(19):

\[ \alpha = \alpha_1 + \alpha_2(\phi - 1) \]  
(16)

\[ \beta = \beta_1 + \beta_2(\phi - 1) \]  
(17)

\[ \gamma = \gamma_1 + \gamma_2(\phi - 1) \]  
(18)

\[ \theta = \theta_1 + \theta_2(\phi - 1) \]  
(19)

Coefficients, \( S_{0f}, m_{0b}, a, b, c, d, \alpha_1, \alpha_2, \beta_1, \beta_2, \gamma_1, \gamma_2, \theta_1 \) and \( \theta_2 \) have been found using an unconstrained minimization method for three hydrogen concentrations as listed in Tables 1 and 2. These correlations are only valid for the range of pressures, temperatures and equivalence ratios that listed in those tables. Since correlations for hydrogen concentrations of 10% and 25% have been derived only for stoichiometric mixture \((\phi = 1)\), the coefficients of \( a, b, c, d, \alpha_2, \beta_2, \gamma_2 \) and \( \theta_2 \) are non-applicable which are shown by “n/a” in Tables 1 and 2.

The cellulurity factor is defined as,

\[ \xi = \frac{A_f - A_L}{A_L} = \frac{S_f - S_{0f}}{S_{0f}} \]  
(20)

By combining Eqs. (12) and (13) and assuming the mass burning rate and laminar burning speed are known respectively from experimental pressure data and 1D free flame simulation using Davis et al. mechanism [33], the total flame surface area can be evaluated as,

\[ A_f = \frac{m_b}{\rho_u A_L S_f} \]  
(21)

Finally, using Eq. (20) the cellulurity factor will be calculated as,

\[ \xi = \frac{m_b}{\rho_u A_f S_f} - 1 \]  
(22)

The cellulurity factor which includes the information regarding the shape and size of cells and wrinkles all over the flame front is a function of unburned gas temperature, mixture pressure, equivalence ratio and hydrogen concentration. To calculate the cellulurity factor in Eq. (22), mass burning rate \( (m_b) \), laminar flame area \( (A_L) \)

| Table 1 | Coefficients of cellular burning speed correlation for H2/CO/O2/He mixture. |
|---------|---------------------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| \( S_{0f} \) | \( a \) | \( b \) | \( \alpha_1 \) | \( \alpha_2 \) | \( \beta_1 \) | \( \beta_2 \) | \( \gamma_1 \) | \( \gamma_2 \) | \( \theta_1 \) | \( \theta_2 \) | \( \phi \) |
| \( z = 5\% \) | 67.671 | 1.040 | -0.500 | 1.545 | -0.534 | 0.045 | 0.130 | 2-50 | 400-800 | 0.6-2 |
| \( z = 10\% \) | 88.443 | n/a | n/a | 1.562 | n/a | 0.049 | n/a | 2-40 | 400-750 | 1 |
| \( z = 25\% \) | 139.70 | n/a | n/a | 1.650 | n/a | 0.044 | n/a | 2-40 | 400-750 | 1 |

| Table 2 | Coefficients of mass burning rate correlation for H2/CO/O2/He mixture. |
|---------|---------------------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| \( m_{0b} \) | \( c \) | \( d \) | \( \gamma_1 \) | \( \gamma_2 \) | \( \theta_1 \) | \( \theta_2 \) | \( \phi \) |
| \( z = 5\% \) | 8.851 | 1.225 | -0.529 | 1.943 | 0.947 | 1.010 | -0.215 | 2-50 | 400-800 | 0.6-2 |
| \( z = 10\% \) | 10.111 | n/a | n/a | 1.815 | n/a | 1.092 | n/a | 2-40 | 400-750 | 1 |
| \( z = 25\% \) | 15.038 | n/a | n/a | 2.035 | n/a | 1.048 | n/a | 2-40 | 400-750 | 1 |
and unburned gas density \( (\rho_u) \) were calculated from differential based multi-shell model \[24\] using experimental pressure data and Laminar burning speed \( (S_l) \) was computed from free flat flame simulation using Davis et al. mechanism \[33\]. In the following sections, the effect of temperature, pressure, equivalence ratio and hydrogen concentration on cellular burning speed, mass burning rate and cellularity factor will be investigated in details.

4.2.1. Effect of temperature

The effect of two different preheat temperatures of 400 and 450 K on cellular burning speed and mass burning rate of stoichiometric H\(_2\)/CO/O\(_2\)/He mixture at initial pressure of 10 atm and hydrogen concentration of 10% is shown in Fig. 5. As it was expected the cellular burning speed and mass burning rate are directly proportional to temperature which is also demonstrated through positive temperature exponents listed in Tables 1 and 2. It means that the higher the temperature, the higher the cellular burning speed and mass burning rate provided that all other parameters such as pressure and mixture composition are fixed. As discussed earlier the cellular burning speed is directly proportional to laminar burning speed and cellularity factor. The laminar burning speed, calculated using free flat flame, has a positive dependency to temperature as shown in Fig. 6 at a given pressure.

Fig. 5. Cellular burning speed and mass burning rate of stoichiometric H\(_2\)/CO/O\(_2\)/He mixture along two isentropes at different initial temperatures, initial pressure of 10 atm and hydrogen concentration of 10%.

Fig. 6. Cellularity factor and laminar burning speed of stoichiometric H\(_2\)/CO/O\(_2\)/He mixture along two isentropes at different initial temperatures, initial pressure of 10 atm and hydrogen concentration of 10%.

Fig. 7. Cellular burning speed and mass burning rate of stoichiometric H\(_2\)/CO/O\(_2\)/He mixture along isentropes at different initial pressure for initial temperature of 400 K and hydrogen concentration of 25%.

Fig. 6 shows that the cellularity factor which indicates the percentage of increase of flame surface due to cell formation has a negative relationship with temperature. As temperature increases at a given pressure, equivalence ratio and hydrogen concentration, the flame thickness increases and therefore it weakens the hydrodynamic instability which delays the cell formation phenomena. In other words, increasing the temperature while the other parameters are constant promote the flame stability and decrease the cell formation tendency which results in decreasing the cellularity factor. In one hand increase in temperature, increases the laminar burning speed and on the other hand it decreases the cell formation propensity and consequently the cellularity factor. Since the laminar burning speed has a strong dependency to temperature than cellularity factor, the overall effect of temperature on cellular burning speed would be positive as shown in Fig. 5. This strong dependency of laminar burning speed to temperature has been proved through the large positive exponent in power law correlation by Askari et al. \[24\].

4.2.2. Effect of pressure

Fig. 7 shows the cellular burning speeds and mass burning rates of stoichiometric H\(_2\)/CO/O\(_2\)/He mixture along three isentropes at varying initial pressures of 2, 5 and 10 atm for hydrogen percentage of 25% and initial temperature of 400 K. As the pressure exponents of power law correlations in Eqs. (14) and (15) are always positive, it indicates that the initial pressure has a direct positive effect on both cellular burning speed and mass burning rate as shown in Fig. 7. It also shows that the positive effect of pressure on mass burning rate is much greater than cellular burning speed. It is demonstrated through several scientific publications \[3,21,24–26,47\] that the laminar burning speed has negative dependency with pressure. The question is why the cellular burning speed which is proportional to laminar burning speed behaves differently and has a minor positive dependence on pressure. This question can be answered by considering the other important parameter that cellular burning speed depends on, cellularity factor. Opposite to the laminar burning speed, the cellularity factor has a very strong and direct relationship to pressure. As shown in Fig. 8, pressure has a negligible effect on laminar flame area which is only a function of flame radius. This figure also shows that the total flame area increases as pressure increases. The reason is that, increasing pressure increases the tendency of flame destabilization through the enhancement of hydrodynamic instability and reduction of flame thickness \[24\] and promotes the cell formation tendency.
over the flame front and consequently increases the total flame area and cellularity factor as shown in Fig. 8.

As the pressure increases, since the positive effect of cell formation is slightly greater than negative effect of laminar burning speed, then the cellular burning speed tends to increase. The increase in mass burning rate as pressure increases is much more significant than cellular burning speed. Based on Eq. (13) the mass burning rate is a function of laminar flame area, cellular burning speed and unburned gas density. As shown in Fig. 8 the laminar flame area doesn't have impact on mass burning rate as the pressure changes at a given temperature. Also based on above explanation and Fig. 7 the cellular burning speed has a minor positive effect on mass burning rate by increasing the pressure at a given temperature. But unburned gas density increases by pressure based on ideal gas law which is the only important term that explain the strong positive connection between mass burning rate and pressure.

### 4.2.3. Effect of equivalence ratio

Cellular burning speeds and mass burning rates of H₂/CO/O₂/He mixture at various equivalence ratios for hydrogen percentage of 5%, preheat temperature of 480 K and initial pressure of 12 atm along three isentropes are shown in Fig. 9. As the equivalence ratio increases from $\phi = 0.6$ to $\phi = 2.0$ both cellular burning speed and mass burning rate increase. Fig. 10 shows the effect of equivalence ratio on burning speed and cell formation. Increasing equivalence ratio from 0.6 to 2 leads to increase of the cell formation and laminar burning speed. The differences between laminar and cellular burning speed at lean mixture ($\phi = 0.6$) is negligible because at this stoichiometry there exist very few cells over the flame front. As equivalence ratio increases the cell formation percentage increases and consequently the cellular burning speed deviate drastically from laminar burning speed.

### 4.2.4. Effect of hydrogen concentration

Hydrogen addition has a significant direct impact on cellular burning speed and mass burning rate. As shown in Fig. 11, the cellular burning speed and mass burning rate increase with increasing hydrogen concentration in the fuel blend. Fig. 12 compares the laminar and cellular burning speeds of stoichiometric H₂/CO/O₂/He mixture for three different hydrogen concentrations at preheat temperature of 450 K and initial pressure of 2 atm along isentropes. As mentioned before, the laminar burning speed has been calculated from free flat flame simulation.

As it can be seen the difference between laminar and cellular burning speeds at the beginning of flame propagation are negligi-

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**Fig. 8.** Cellularity factor and flame area of stoichiometric H₂/CO/O₂/He mixture along three isentropes at different initial pressure for initial temperature of 400 K and hydrogen concentration of 25%.

**Fig. 9.** Cellular burning speed and mass burning rate of H₂/CO/O₂/He mixture along three isentropes at different equivalence ratios for initial pressure of 12 atm, initial temperature of 480 K and hydrogen concentration of 5%.

**Fig. 10.** Cellularity factor, cellular and laminar burning speed of H₂/CO/O₂/He mixture for three different equivalence ratios of 0.6, 1.0 and 2.0 at pressure of 23.5 atm, temperature of 602 K and hydrogen concentration of 5%.

**Fig. 11.** Cellular burning speed and mass burning rate of stoichiometric H₂/CO/O₂/He mixture along three isentropes at different hydrogen concentration for initial temperature of 450 K and initial pressure of 2 atm.
As flame propagates and temperature and pressure increase, the cellular burning speed starts to deviate from laminar burning speed. As flame propagates and temperature and pressure increase, the total flame area increases. The effect of cell formation on burning speed and total flame front delays the onset of cell formation.

5. Summary and conclusions

In this paper, mass burning rate and cellular burning speed of syngas/O₂/He were calculated for a wide range of equivalence ratio from 0.6 to 2, temperature from 400 to 750 K and pressure from 2 to 50 atm using a new differential-based multi-shell model based on pressure rise. The structure and effect of thermo-diffusive and hydrodynamic instabilities were studied at very high pressures at which the flame was always cellular. The power law correlations were also developed as a function of equivalence ratio, temperature and pressure for cellular burning speed and mass burning rate. The effect of cell formation on burning speed and total flame front area were investigated in terms of a newly developed parameter called cellularity factor. In addition to a complete experimental data, the theoretical laminar burning speeds via a steady, one-dimensional free flame code in conjunction with detailed chemical mechanism were also used to calculate the cellularity factor. The results showed that the cellularity factor has a positive relation to pressure, equivalence ratio and hydrogen concentration while it has a negative dependency to temperature. Cellular burning speed and mass burning rate have both positive relation to temperature, pressure, equivalence ratio and hydrogen concentration. Pressure increase at a given temperature doesn’t have significant effect on laminar flame area while it increase the total flame area. Reduction in hydrogen concentration makes the flame stable and delays the onset of cell formation.

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