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# Determination of laminar burning characteristics of a surrogate for a pyrolysis fuel using constant volume method

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### Abstract

#### Journal Pre-proof

A fast pyrolysis biofuel, mainly consisting to 98% of ethanol, ethyl acetate, diethyl ether, acetone and 2-butanone with mass ratios of 9:6:2:1:1, was catalytically produced from rice husk. A preliminary engine test demonstrated this biofuel has the possibility of being a gasoline blending stock. However, its fundamental burning features are not fully understood. This work presents the experimental investigations of the spherical propagating flame of a surrogate fuel representing the biofuel in a constant volume combustion chamber (CVCC). Tests were conducted at initial pressures of 0.1-0.4 MPa, initial temperatures of 358-418 K, and equivalence ratios of 0.7-1.4. Employing the constant volume method (CVM) allows determining laminar burning speeds ( $S_u$ ) of this surrogate at conditions far beyond the initial conditions (0.1-0.8 MPa, 358-490 K). Power law fitting correlations between  $S_u$  and pressure were obtained via the constant volume method (CVM). Cellularity appears when pressure or temperature is high, and cellular burning speed was calculated by CVM as well.  $S_u$  determined via the constant pressure method (CPM) were compared with those from the CVM. Discrepancies between the results from the CVM and the CPM are within 15%, except at the conditions where flame cellularity appeared. Additionally, an explicit correlation of  $S_u$  was obtained from the experimental results.

Keywords: biofuel; laminar burning speed; constant-volume method; cellular flame speed

Greek symbols	Journal Pre-proof
$\phi$	Equivalence ratio
$ ho_{ m i}$	Initial density (kg/m <sup>3</sup> )
$ ho_{ m u}$	Density of unburned gas (kg/m <sup>3</sup> )
κ	Stretch rate (s <sup>-1</sup> )
α	Temperature exponent
β	Pressure exponent
Symbols	
$S_{\rm u}^{0}$	Laminar burning speed at initial condition (m/s)
S <sub>u</sub>	Laminar burning speed obtained by CVM (m/s)
S <sub>u,CPM</sub>	Laminar burning speed obtained by CPM (m/s)
$S_{ m g}$	Expansion speed of burned gas (m/s)
$S_{ m f}$	Flame front propagation speed (m/s)
p	Combustion pressure (MPa)
$p_{\mathrm{i}}$	Initial pressure (MPa)
$p_{ m e}$	Peak pressure (MPa)
T <sub>u</sub>	Unburned gas temperature (K)
T <sub>i</sub>	Initial gas temperature (K)
x	Burned mass fraction
$p_{\rm r}$	Pressure rise fraction
m <sub>i</sub>	Mass of initial gas (kg)
m <sub>u</sub>	Mass of unburned gas (kg)
m <sub>b</sub>	Mass of burned gas (kg)
r <sub>f</sub>	Radius of flame front (m)
r <sub>w</sub>	Radius of combustion chamber (m)
$A_{ m f}$	Area of flame front (m <sup>2</sup> )
k <sub>u</sub>	Heat capacity ratio of unburned gas

C <sub>p,u</sub>	Molar heat capacity of unburned gas at constant pressure (J/(mol•K))
$C_{\mathrm{p,j}}$	Molar heat capacity of <i>j</i> component at constant pressure $(J/(mol \cdot K))$
$C_{ m p,air}$	Molar heat capacity of air at constant pressure (J/(mol•K))
$C_{ m v,u}$	Molar heat capacity of unburned gas at constant volume (J/(mol•K))
$a_{\mathrm{j}}$	Volumetric air-fuel ratio of <i>j</i> component
nj	Molar fraction of <i>j</i> component
R <sub>g</sub>	Gas constant
Subscripts	<u>x</u>
и	Unburned gas
b	Burned gas
i	Initial condition
j	<i>j</i> component in the mixture

#### **1** Introduction

#### Journal Pre-proof

Clean renewable energy resources, for example, biomass, are mid- and long-term solutions for the depletion of fossil fuels and environmental pollution [1]. Fast pyrolysis biofuel is considered as one of the promising alternative fuels. Recently, a fast pyrolysis biofuel was produced from rice husk at Zhejiang University [2,3]. Based on detailed composition analysis of that biofuel, a mixed fuel which consists of ethanol, ethyl acetate, diethyl ether, acetone and 2-butanone with mass ratios of 9:6:2:1:1, can be used as a surrogate for this biofuel [3,4].

Laminar burning speed ( $S_u$ ) is the velocity where one-dimensional, planar, adiabatic, un-stretched [5,6] and premixed flame propagates through the unburned gas mixture. It is an essential intrinsic parameter of a combustible mixture [7-12].  $S_u$  helps to explain combustion phenomena, such as extinction [13-15], flashback, and blow off [16,17]. It also conveys information about diffusivity, reactivity, and exothermicity of the combustible mixture [18]. Therefore, it is important for designing practical combustion systems [19-22], especially at temperatures and pressures higher than the ambient condition. Moreover, it is an essential input parameter for turbulent combustion models [23,24].

There are many experimental setups for identifying  $S_{u}$ , including the Bunsen flame, counter-flow flame, flat flame and constant volume combustion chamber (CVCC). CVCC is the most commonly used [21,22,25]. There are two different ways to obtain  $S_u$  in the CVCC, the constant pressure method (CPM) and the constant volume method (CVM). The CPM was first announced by Ellis [26] in 1928, while the CVM was initially mentioned by Lewis and von Elbe in 1934 [27]. In the CPM, CVCC is set up in the Schlieren system, and a high-speed camera records Schlieren images [4], from which the quasi-steady smooth spherical flame in early propagation period is chosen for data analysis [27]. Meanwhile, the flame structure can be observed in the Schlieren photographs to determine when flame cellularity and instability occur. In the CVM, combustion pressure is recorded, and  $S_u$  is calculated as the function of pressure (p) and burned gas fraction (x) [27].  $S_u$  calculated via the CVM involves a few assumptions; therefore, it tends to be less accurate than that obtained from the CPM. However, there are two remarkable advantages that make CVM indispensable. Firstly, for a given mixture,  $S_u$  can be continuously obtained over a wide range of pressures and temperatures from a single experiment [10,28]. Thus,  $S_{\rm u}$  at higher pressures and temperatures close to practical engine operating conditions (3-7 MPa and 700-800 K) can be obtained. In addition, cellular flame speed can be calculated from the CVM, which cannot be determined from the CPM [29].

Several investigations have been conducted in the literature to calculate  $S_u$  using the CVM. Lewis and Von Elbe [27] proposed the widely used CVM linear *x-p* relation, and they used it to determine  $S_u$ . O'Donovan and Rallis [30] simplified the *x-p* relation by introducing the temperature gradient. Saeed and Stone [31,32] used the multiple burned gas zone model to determine  $S_u$ . They proposed a 9-term correlation for determining  $S_u$ . Based on the multi-zone modelling, Luijten et al. [33,34] developed an alternative analytical *x-p* relation and compared it with 1D unsteady simulation and numerical two-zone model. Faghih and Chen [10] evaluated the accuracy of the various *x-p* relations used in CVM, from which it can be found that linear *x-p* relation is a reasonable choice. Chen et al. [35,36] studied the  $S_u$  of outwardly propagating spherical flames and investigated the effects of confinement, stretch, radiation and ignition energy on  $S_u$ . Huang et al. [37-39] investigated  $S_u$  of several fuels, such as dimethyl ether, natural gas-hydrogen and methanol-nitrogen/carbon dioxide at different temperatures, pressures, equivalence ratios and mentioned the appearance of cellularity. Recently, Hinton and Stone [40,41] used both CPM and CVM to investigate the  $S_u$  of aqueous ethanol and proposed an explicit 14-terms correlation for  $S_u$  as a function of pressure, temperature and equivalence ratio.

This paper investigates the spherically expanding flame of the fast pyrolysis fuel in the CVCC at initial pressures of 0.1-0.4 MPa, initial temperature of 358-418 K, and at equivalence ratios ( $\phi$ ) of 0.7-1.4. The novelty of this paper is that the CVM, which takes the combustion pressure trace as the main input, was used to determine  $S_u$  of the biofuel fast pyrolysis blend at conditions (pressure up to 0.8 MPa; temperature up to 490 K) beyond the initial conditions (pressure up to 0.4 MPa; temperature up to 418 K ).

### 2 Experimental setup and procedures

Figure 1 provides the experimental layout with a CVCC of 1.94L inner volume. The CVCC was heated by six heating elements fitted on its walls. Moreover, ignition of the mixture was done with two electrodes (d=0.4 mm) facing each other together with ignition control set. The spark energy was maintained at 15 mJ. The spherically expanding flame images were obtained by the Schlieren imaging arrangement together with a high-speed camera (6000 fps @ 512×512 pixels) via see-through openings ( $\Phi$ =105 mm) at both ends of the chamber. Combustion pressure was measured by pressure transducers, meanwhile, the experiments were done at initial temperatures of 358-418 K, initial pressures of 0.1-0.4 MPa, and equivalence ratios of 0.7-1.4. The mixture was unable to ignite at the equivalence ratio of 0.5 during our experiment, however, at the equivalence ratio of 0.6 almost 10 experiments were done and only 2-3 experiments were able to ignite the mixture. Therefore, we chose 0.7 as the lower limit for the study. On the other hand, at very rich mixtures, thus equivalence ratios beyond 1.4 the flame was unstable; hence, we chose 1.4 as the upper limit for the study. A total of 216 experiments were performed and each test was repeated three times. Complete information about the experimental

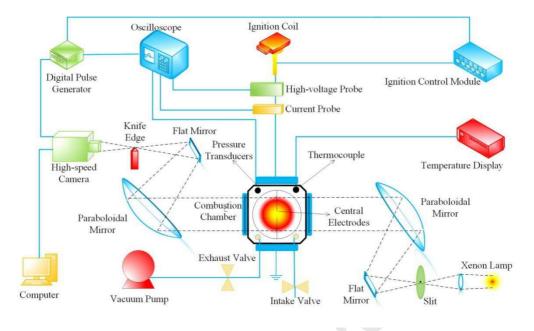


Figure 1. Experimental layout

#### **3** Determination of S<sub>u</sub> with CVM

There are a few assumptions for determining  $S_u$  using the CVM [10,33,40]: (1) the unburned gas is uniform, and it is compressed isentropically; (2) the pressure is uniform in the whole combustion chamber; (3) both the unburned and burned gases are ideal gas; (4) the total mass and volume of the vessel contents are conserved; (5) the external heat input, heat losses, radiation and buoyancy effects are negligible; (6) the flame stretch is negligible for large flame radius.

 $S_{\rm u}$  is defined as [27]:

$$S_{\rm u} = -\frac{1}{A_{\rm f}\rho_{\rm u}}\frac{dm_{\rm u}}{dt} \tag{1}$$

where  $m_u = 4(r_w^3 - r_f^3)\pi\rho_u/3$ ,  $m_u$  is the mass of unburned gases;  $\rho_u$  is the unburned gas density;  $r_f$  and  $r_w$  are the radius of the flame front and the CVCC.  $A_f$  is the flame front area,  $A_f = 4\pi r_f^2$ .

In consideration with the aforementioned assumptions, the unburned gas temperature and  $\rho_u$  can be estimated by the following equations:

$$\frac{T_{\rm u}}{T_{\rm i}} = \frac{\left[\frac{p}{p_{\rm i}}\right]^{\frac{k_{\rm u}+1}{k_{\rm u}}}}{p_{\rm i}}$$
(2)

$$\frac{\rho_{\rm u}}{\rho_{\rm i}} = \left(\frac{p}{p_{\rm i}}\right)^{\frac{1}{k_{\rm u}}} \tag{3}$$

where  $T_u$  and p are the temperatures of the unburned gas and pressure in the CVCC during combustion, respectively;  $T_i$  (initial temperature),  $p_i$  (initial pressure),  $\rho_i$  (initial density) and  $k_u$  (specific heat ratio).

The burned mass fraction x, defined as the ratio of burned gas and the total amount of gas is calculated via:

$$m_{\rm u} = m_{\rm i} - m_{\rm b} = (1 - x)m_{\rm i}$$
 (4)

where  $m_i = 4\pi r_w^3 \rho_i / 3$ .  $m_i$  and  $m_b$  are the mass of the initial and burned gases, respectively.

A linear relation between *p* and *x*, proposed by Lewis and von Elbe [27], can be used to estimate the burned mass fraction:

$$x = \frac{p - p_{\rm i}}{p_{\rm e} - p_{\rm i}} \tag{5}$$

where  $p_e$  is the maximum pressure during combustion.

Combining Equations 1-4 yields [10,45]:

$$S_{\rm u} = \frac{r_{\rm w}}{3} \left[1 - (1 - x)(\frac{p_{\rm i}}{p})^{\frac{1}{k_{\rm u}}}\right]^{\frac{2}{3}} (\frac{p_{\rm i}}{p})^{\frac{1}{k_{\rm u}}} \frac{dx}{dt}$$
(6)

In the linear x-p relation, the burned mass fraction x equals to the pressure rise fraction  $p_r$ [33]:

$$p_{\rm r} = \frac{p - p_{\rm i}}{p_{\rm e} - p_{\rm i}} \tag{7}$$

Five components were used as a surrogate for the fast pyrolysis fuel. Some properties of the five components are listed in Table 1. The fuel is metered by a syringe with a resolution of  $1\mu$ L. Filtered workshop air is used in the combustion.

 Table 1 Properties of components in fast pyrolysis fuel

Components	Jo EthanolPr	e-p <b>Ethyl</b> f	Diethyl	Acetone	2-butanone	
		acetate	ether			
Formula	C <sub>2</sub> H <sub>5</sub> OH	$C_4H_8O_2$	$C_4H_{10}O$	C <sub>3</sub> H <sub>6</sub> O	C <sub>4</sub> H <sub>8</sub> O	
Molecular weight (g/mol)	46.07	88.11	74.12	58.08	72.11	
Weight fraction (wt.%)	47.4%	31.6%	10.5%	5.3%	5.3%	
Molar fraction (mol.%)	60.7%	21.1%	8.4%	5.4%	4.3%	
Volumetric air-fuel ratio	3	5	6	4	5.5	

Eq. (8) was used to calculate the heat of unburned gas capacity at constant pressure.  $C_{p,u}$  is an average value of specific heat capacity of ethanol, ethyl acetate, diethyl ether, acetone, 2-butanone and air with their molar fractions.

$$C_{p,u} = \frac{\varphi \sum_{j=1}^{5} C_{p,j} \cdot n_{j} + (\sum_{j=1}^{5} a_{j} \cdot n_{j}) C_{p,air}}{\varphi \sum_{j=1}^{5} n_{j} + \sum_{j=1}^{5} a_{j} \cdot n_{j}}$$
(8)

where  $a_j$  and  $n_j$  are the volumetric air-fuel ratio and molar fraction of each component, respectively.

To extrapolate  $S_u$  back to the initial condition, the power law correlation is used to fit the  $S_u$  [10,45]:

$$S_{\rm u} = S_{\rm u}^{\ 0} (\frac{T}{T_{\rm i}})^{\alpha} (\frac{p}{p_{\rm i}})^{\beta}$$
(9)

where  $\alpha$  and  $\beta$  are exponents of temperature and pressure, respectively.  $S_u^0$  is the laminar flame speed at the initial condition and varies in different experiments.

It is assumed that the compression of unburned gas in the CVCC is adiabatic and isentropic:

$$S_{\rm u} = S_{\rm u}^{\ 0} (\frac{p}{p_{\rm i}})^c \tag{10}$$

where  $c = \alpha((k_u - 1) / k_u) + \beta$ .

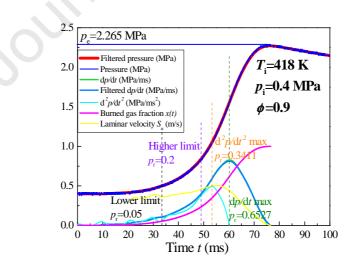
#### 4 Experimental uncertainties

The experimental uncertainties are mainly related to  $T_0(\Delta U_T)$ ,  $p_0(\Delta U_p)$ , the flame front total inner pixels ( $\Delta U_A$ ), the chamber effective volume ( $\Delta U_V$ ) and the fuel metering ( $\Delta U_F$ ).  $\Delta U_T$  inaccuracy is due to the thermocouple accuracy (±0.5%). Therefore, this generated errors in the estimation of the amount of fuel injection using temperature, CVCC volume and the chosen equivalence ratio. According to the experience from authors, this is likely to generate 0.8% and 1.5% error in the estimation of the laminar burning velocity at 0.1 MPa and 0.4 MPa. The  $p_0$  uncertainty caused less than 0.1% because the pressure sensor is accurate to 0.0001 MPa. On one hand, the flame front area uncertainty is nearly ±10 pixels, leading to almost 1% incertitude of the flame front inner pixels.  $\Delta U_V$  uncertainty was almost 0.2%. The uncertainty of fuel metering relies on the capacity of fuel required for a specific test condition. In short, the CPM global uncertainty ( $\sqrt{\Delta U_T^2 + \Delta U_p^2 + \Delta U_p^2 + \Delta U_p^2}$ ) is within 2%, and that of the CVM ( $\sqrt{\Delta U_T^2 + \Delta U_p^2 + \Delta U_V^2 + \Delta U_F^2}$ ) is about 1.5%.

#### 5 Results and discussion

#### 5.1. $S_u$ determination with CVM

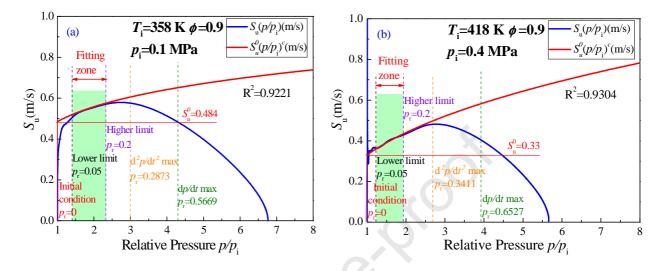
The temporal pressure was recorded in the experiments. Then, the first derivative (dp/dt) and the second derivative  $(d^2p/dt^2)$  of pressure were obtained. Gaussian filter was used to smooth the curve of *p* and dp/dt. Next, *x* and *S*<sub>u</sub> can be calculated by eqs. (5) and (6). *p*<sub>r</sub> in the range of 0.05 and 0.2 was used for determination of *S*<sub>u</sub>[45]. Finally, the experimental curve of *S*<sub>u</sub> in the chosen range was fitted by eq. (10).



**Figure 2.** p, dp/dt,  $d^2p/dt^2$ ,  $S_u$  and x as a function of time (initial condition:  $p_i = 0.4$  MPa;  $T_i = 418$  K; and  $\phi = 0.9$ ).

Figure 2 shows the variation of p, dp/dt,  $d^2p/dt^2$ , x,  $S_u$  in a single experiment at the initial condition of  $p_i$ = 0.4 MPa;  $T_i$ = 418 K; and  $\phi$ =0.9. In fact, every experiment led to a figure like Figure 2. Figure 2

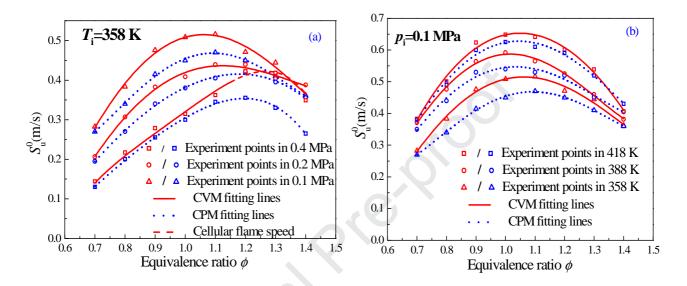
is used as an example to show the profile of the main parameters. For choosing the valid calculation window, the lower limit of  $p_r$  is 0.05. There are two reasons. One is because of interference signal, so dp/dt cannot be accurately determined lower than  $p_r=0.05$ , another is because when  $p_r>0.05$ , stretch impacts can be considered negligible [45]. The higher limit is  $p_r=0.2$ , because after this point thermal loss from burned gas to the CVCC wall is non-negligible [45].



**Figure 3.** Graph of  $S_u$  against  $p/p_i$  at: (a)  $p_i = 0.1$  MPa,  $T_i = 358$  K, and  $\phi = 0.9$ ; (b)  $p_i = 0.4$  MPa,  $T_i = 418$  K, and  $\phi = 0.9$  (Blue lines are experimental results, while red lines are fitting correlations).

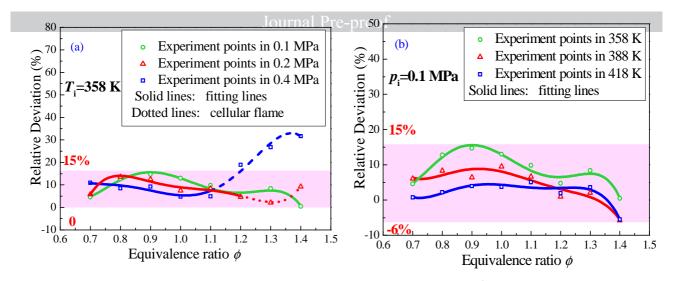
Figure 3 shows  $S_u$  at two initial conditions. The blue lines represent the experimental  $S_u$  as a function of  $p/p_i$ , whereas the red lines indicate the fitting correlations, showing just how the power law correlation was used for fitting  $S_u$  and again extrapolated to the initial and higher conditions. The experimental  $S_u$  increases with the relative pressure at first and then decreases. During the combustion procedure, the pressure and temperature increase simultaneously. At the early stage,  $S_u$  increases with increasing pressure and temperature. At relative pressure of 1, a very steep rise appears because of spark effects on combustion onset, numerical oscillations and stretch effects [46]. However, when pressure exceeds a certain value (such as  $p_r > 0.3411$ @ initial condition of  $p_{i=} 0.4$  MPa;  $T_i = 418$  K; and  $\phi = 0.9$ ), the heat loss becomes significant, which slows down the pressure rise and  $S_u$  decreases at the last stage. As mentioned earlier in Section 4.1, the fitting zone is  $p_r = 0.05$ -0.2 and eq. (10) was used to fit  $S_u$  from the experiments. The coefficients, c and  $S_u^0$  in eq. (10), were obtained for all conditions.  $S_u$  was extrapolated at the initial condition. At the initial condition,  $p_r$  is 0. According to the fitting results between the experimental lines and fitting lines,  $p/p_i = 2$  is the last point where experimental lines and fitting lines fit well for the most conditions in this study. Therefore,  $S_u$  from  $p/p_i = 1$  to  $p/p_i = 2$  were used in this paper.

Figure 4(a) compares  $S_u$  obtained from the CVM and CPM at  $T_i$ =358 K and  $p_i$ =0.1, 0.2, 0.4 MPa. The red lines represent results from the CVM while the blue lines are the CPM data. It can be found that CVM results are larger than results from the CPM. The maximum flame speed always occurs near  $\phi$ =1.1. Further, as the pressure rises, the peak  $S_u$  shifts towards the higher equivalence ratio. With pressure increasing,  $S_u$  decreases, except the conditions for which cellularity appears. Figure 4(b) compares the results at  $p_i$ =0.1 MPa and  $T_i$ =358, 388, 418 K.



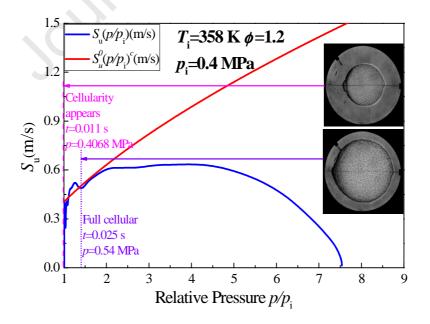
**Figure 4.**  $S_u$  as a function of  $\phi$  at the conditions of: (a)  $p_i$ = 0.1, 0.2 and 0.4 MPa;  $T_i$ = 358 K; (b)  $p_i$ = 0.1 MPa;  $T_i$ = 358, 388 and 418 K (Red solid lines and red symbols are results from the CVM, while blue dotted lines and blue symbols are from the CPM. Dash line show cellular flame speed).

Figure 5 shows the relative deviation in  $S_u$  between the CVM and the CPM. The relative deviation is  $(S_u - S_{u,CPM})/S_{u,CPM} *100\%$ . It can be seen that the relative deviations are within 15% except for the conditions that cellularity occurs which led to a large increase in flame speed. Omari et al. [45] showed  $S_u$  data from the linear *x-p* relation was 20% higher when compared to the CPM. Moreover, values of CVM are consistently larger than those of CPM except for the conditions where  $p_i=0.1$  MPa,  $T_i=388$ and 418 K, and  $\phi=1.4$ . On the other hand, the flame radius calculation in the CPM involves errors; therefore, it also contributes to the discrepancy in  $S_u$  between the CVM and the CPM. Overall,  $S_u$ obtained from the CVM is larger than that from the CPM in most cases.



**Figure 5.** Relative deviation in  $S_u^0$  obtained from the CVM and CPM at the conditions of: (a)  $T_i$ =358 K,  $p_i$ =0.1, 0.2 and 0.4 MPa; (b)  $p_i$ =0.1 MPa,  $T_i$ =358, 388 and 418 K (Solid lines refer to the relative error of  $S_u^0$ , while dotted lines indicate relative error of cellular flame speed. The shadow areas represent error ranges).

Figure 6 is used as an example to show the condition where cellularity appears. The flame speed rises rapidly as well as the fitting line. So, this indicates why the cellular flame speed is considerable larger than  $S_u$  in CPM as disclosed at the conditions where the  $T_i=358$  K,  $p_i=0.4$  MPa and  $\phi=1.2-1.4$  in Figure 4. The equivalence ratios which correspond to the appearance of cellularity are listed in Table 2.



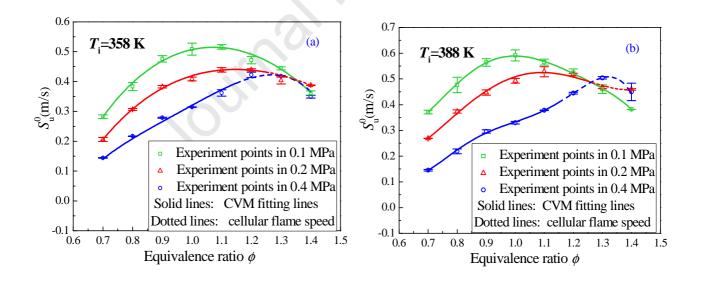
**Figure 6.** A graph of  $S_u$  versus  $p/p_i$  at  $p_i=0.4$  MPa,  $T_i=358$  K, and  $\phi=1.2$ . (Blue line is from experiment, and red line is from fitting correlation).

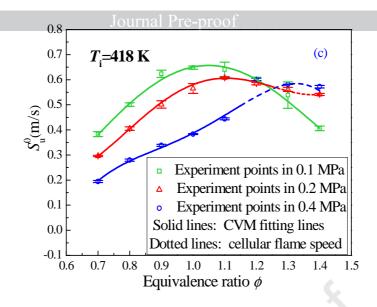
Table 2	Minimum	equivalence	ratio for y	which cel	lularity appears
					in a point of the

$p_{\mathrm{i}} \setminus T_{\mathrm{i}}$	358 K	388 K	418 K
0.1 MPa	none	none	none
0.2 MPa	1.3	1.3	1.3
0.4 MPa	1.2	1.2	1.2

## 5.3. Impact of pressure on $S_u^0$

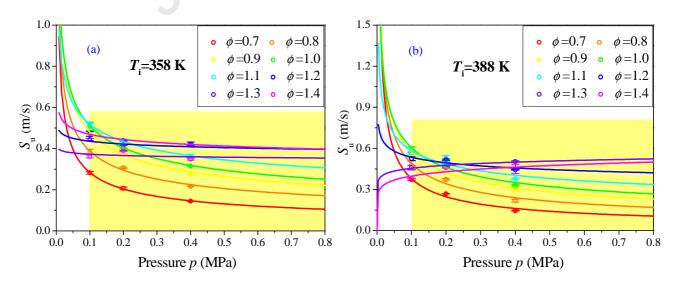
Figure 7 shows the impact of p on  $S_u^0$  at  $T_i = 358$ , 388 and 418 K.  $S_u^0$  decreases as pressure increases. Flame cellularity tends to occur in the rich flames, caused by hydrodynamic instability. Flame cellularity accelerates flame speed, which causes  $S_u^0$  to increase with the increase of pressure at  $\phi = 1.2$ -1.4. The phenomenon is not obvious at  $T_i = 358$  K, but when  $T_i = 388$  and 418 K, it becomes apparent. Uncertainties from preparation using CVM in the experiments is 1.5%. And uncertainties from measurement using CVM in the experiments are 0 - 2%. Error bars drawn in Figure 7 can show the uncertainties from the measurement.

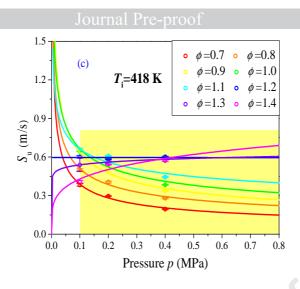




**Figure 7.** Impact of *p* on  $S_u^0$  (a)  $T_i$ =358 K; (b)  $T_i$ =388 K; (c)  $T_i$ =418 K. (Solid lines stand for  $S_u^0$ , while dotted lines stand for cellular flame speed).

Figure 8 shows  $S_u$  as a function of p at  $\phi$ =0.7-1.4 and  $T_i$ =358, 388 and 418 K. The power law correlations fitted to  $p_i$ =0.1, 0.2 and 0.4 MPa give a very good fit. According to the fitting zone referred in Section 4.1,  $S_u$  could be obtained at p=0.8 MPa. It can be seen from Figure 8 that  $S_u$  decreases with an increase in pressure except for the conditions for which  $\phi$ =1.2-1.4. It is obvious that when the equivalence radio is increased from 1.2 to 1.4, the tendency of  $S_u$  is different from the others, even when  $\phi$ =1.3-1.4, they increase as the pressure increases. The reason is that cellularity occurs when  $\phi$ =1.2-1.4, which causes the flame speed to be larger than the value it should be and flame speeds at elevated pressure are higher than that at atmospheric pressure.

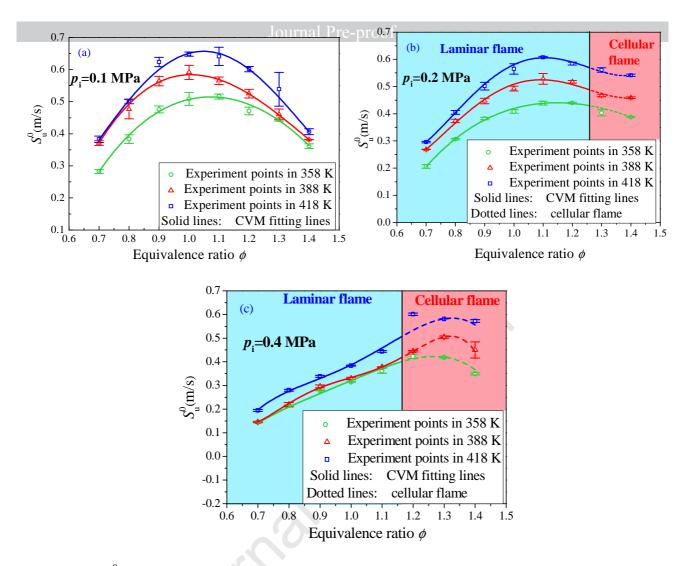




**Figure 8.** A graph of  $S_u$  versus *p* by extrapolation at the condition of: (a)  $T_i=358$  K; (b)  $T_i=388$  K; (c)  $T_i=418$  K (The shadow area represents the research range of pressure in this paper).

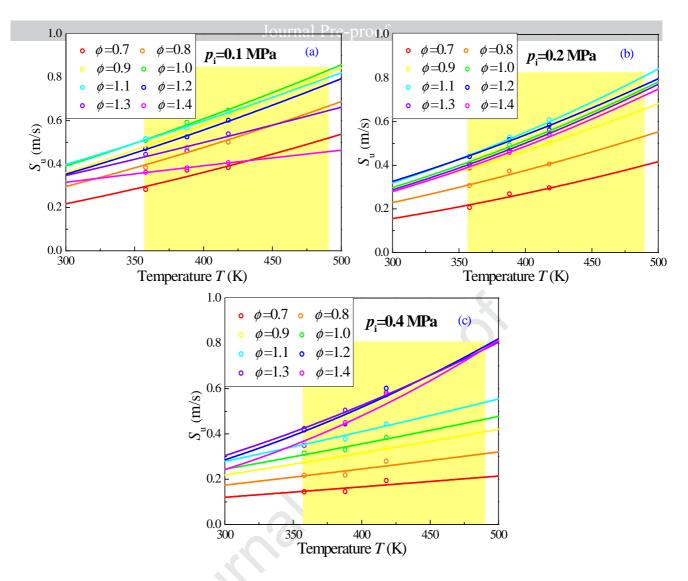
# 5.4. $S_u^0$ as a function of temperature

Figure 9 shows the variation of  $S_u^0$  as a function of  $T_u$  at three fixed pressures (0.1, 0.2 and 0.4 MPa). In Figure 9(b) and (c), the blue areas, on the left of the black lines, represent  $S_u^0$  and the red areas on the right of the black lines, represent cellular flame speed. Black lines are at the intersection of solid lines and dotted lines. In Figure 9(b), the flame is cellular at  $\phi=1.3$  and the flame is laminar at  $\phi=1.2$ ; therefore, cellularity could appear between 1.2 and 1.3. Therefore, the black line lies between  $\phi=1.2$  and 1.3. In Figure 9(c), the black line is between  $\phi=1.1$  and 1.2. It can be seen from Figure 9 that  $S_u^0$  increases with increasing temperature. It can also be observed that the shape of curves at the same pressure are similar, especially at  $p_i=0.2$  MPa and  $p_i=0.4$  MPa. Additionally, at the same pressure, the positions correspond to the maximum flame speed are similar. With the increase of pressure, the peak shifts right, from  $\phi=1.0$  to  $\phi=1.3$ , which is also impacted by cellularity to some extent, because cellularity accelerates the flame speed at  $\phi=1.2-1.4$ . Therefore, pressure has little influence on the shape of the curves and the station of the peak. It can also be found that cellularity appears at  $\phi=1.3-1.4$  when  $p_i=0.2$  MPa and at  $\phi=1.2-1.4$  when  $p_i=0.4$  MPa, which confirms cellularity has a strong relation with pressure. The larger the pressure, the earlier cellularity appears.



**Figure 9.**  $S_u^0$  at  $T_i$ =358, 388 and 418 K: (a)  $p_i$ =0.1 MPa; (b)  $p_i$ =0.2 MPa; (c)  $p_i$ =0.4 MPa (Solid lines stand for  $S_u^0$ , while dotted lines stand for cellular flame speed. Blue areas represent  $S_u^0$  and red areas represent cellular flame speed).

Figure 10 shows the variation of  $S_u$  as a function of  $T_u$  at  $\phi = 0.7$ -1.4 and  $p_i=0.1$ , 0.2 and 0.4 MPa. The power law correlations fitted to three experimental data at  $T_i=358$ , 388 and 418 K respectively give a very good fit. According to the fitting zone referred in Section 4.1, the  $S_u$  could be obtained at  $T_i=490$  K. It can be seen from Figure 10 that the  $S_u$  increases with an increase in temperature. And the increasing rate becomes faster and faster with an increase of temperature.

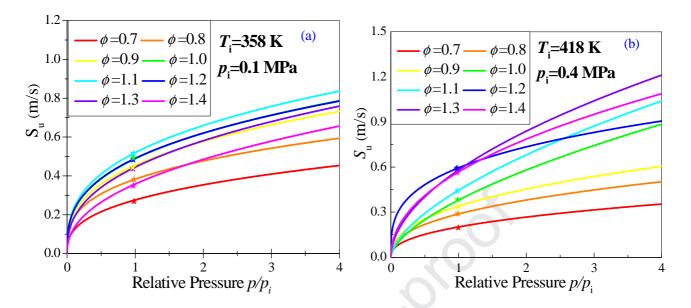


**Figure 10.**  $S_u$  as a function of  $T_u$  by extrapolation at the condition of (a)  $p_i=0.1$  MPa; (b)  $p_i=0.2$  MPa; (c)  $p_i=0.4$  MPa. (The shadow area represents the research range of temperature in this paper).

#### 5.5. $S_{\rm u}$ at elevated pressures and temperatures

Figure 11 shows  $S_u$  obtained at two initial pressures and temperatures conditions. In each experiment,  $S_u$  increased with pressure in a power law. Although the high pressure has a negative impact on  $S_u$ , increased the temperature during combustion offsets the negative impact of increased pressure on  $S_u$ . That is also the reason why in Figure 3  $S_u$  increases at the early stage. As assumption (1), the procedure is isentropic and eq. (2) is used to calculate the temperature from pressure. Therefore, if both  $p/p_i$  and  $T_i$  (which decides  $k_u$ ) are the same, temperature during the combustion procedure will be regarded as the same. The fitting lines fit well when  $p/p_i$  is from 1 to 2 in all figures, so in this paper,  $S_u$  in all experiments are intended to be chosen from initial conditions to the point, at which  $p/p_i$  is equal to 2. In Figure 11, there are only two figures at different conditions shown as representatives.

Another seven figures are similar to Figure 11, so they are not shown in this paper. In Figure 11(b), lines at  $\phi$ =1.3-1.4 increase rapidly, which is caused by cellularity. The detailed explanation and figure can be seen in Figure 5.



**Figure 11.**  $S_u$  as a function of  $p/p_i$  at  $\phi$ =0.7-1.4: (a)  $p_i$ =0.1 MPa,  $T_i$ =358 K; (b)  $p_i$ =0.4 MPa,  $T_i$ =418 K.

#### 5.6. Fitting correlation for $S_{\rm u}$

Figure 12 shows the fitting ranges of p and  $T_u$ . There are 9 experimental points at every equivalence ratio. The fitting correlation given in this paper is valid at  $p_i$ = 0.1-0.8 MPa,  $T_i$ = 358-490 K and  $\phi$ = 0.7-1.4. In the CVM, a series of  $S_u$  over a certain range of pressure and temperature were obtained from a single test. Therefore, starting with the initial ( $p_i$ , $T_i$ ) value, one can draw a continuous line of higher (p, $T_u$ ) values for which  $S_u$  can be determined from the fit. Then, interpolation and extrapolation methods were used to fill up the whole areas.

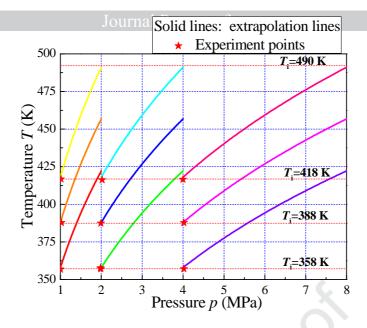


Figure 12. Range of fitting correlation (Colorful solid lines represent fitting lines in the CVM).

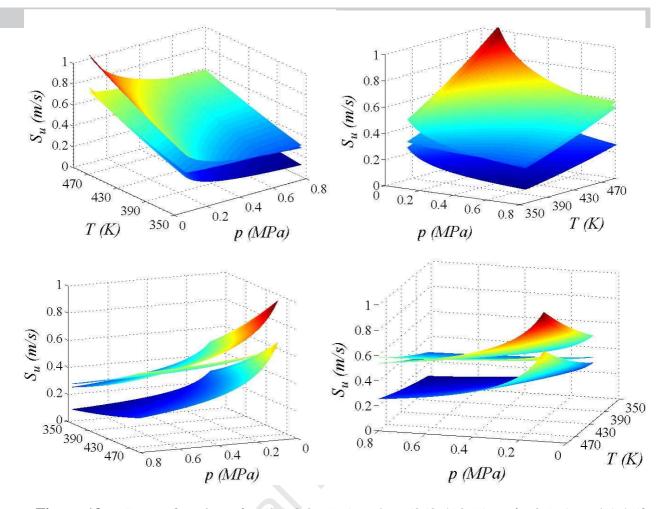
In all the experiments, the variation of  $S_u$  with  $p/p_i$  can be plotted like Figure 3.  $S_u$  from  $p/p_i = 1$  to  $p/p_i=2$  was chosen. At last, an explicit correlation with fitted  $S_u$  as the function of p,  $T_u$  and  $\phi$  can be obtained:

$$S_{\mu} = A(T/358)^{\alpha} (p/0.1)^{\beta}$$
(11)

where:

$$A = S_{u,0} + S_{u,1}(\phi - 1) + S_{u,2}(\phi - 1)^2 + S_{u,3}(\phi - 1)^3 + S_{u,4}(\phi - 1)^2$$
$$\alpha = \alpha_0 + \alpha_1(\phi - 1) + \alpha_2(\phi - 1)^2 + \alpha_3(\phi - 1)^3$$
$$\beta = \beta_0 + \beta_1(\phi - 1) + \beta_2(\phi - 1)^2$$

Table 3 lists the values of the correlation coefficients. The values of *A*,  $\alpha$  and  $\beta$  at various equivalence ratios can be seen in the Appendix. Figure 13 gives a comprehensive description about the effect of both  $T_u$  and p on  $S_u$ . It can be found that  $S_u$  increases with temperature, just like what Figure 10 shows, and  $S_u$  decreases with pressure, just like what Figure 8 shows.  $S_u$  increases from the zone that pressure is high and the temperature is low to the zone that pressure is low and the temperature is high. The maximum and average deviations between  $S_u$  from fitting correlation and  $S_u$  from experiments under  $p_i=0.1$ -0.4 MPa,  $T_i=358$ -418 K,  $\phi=0.7$ -1.4 are 15% and 9%, respectively.

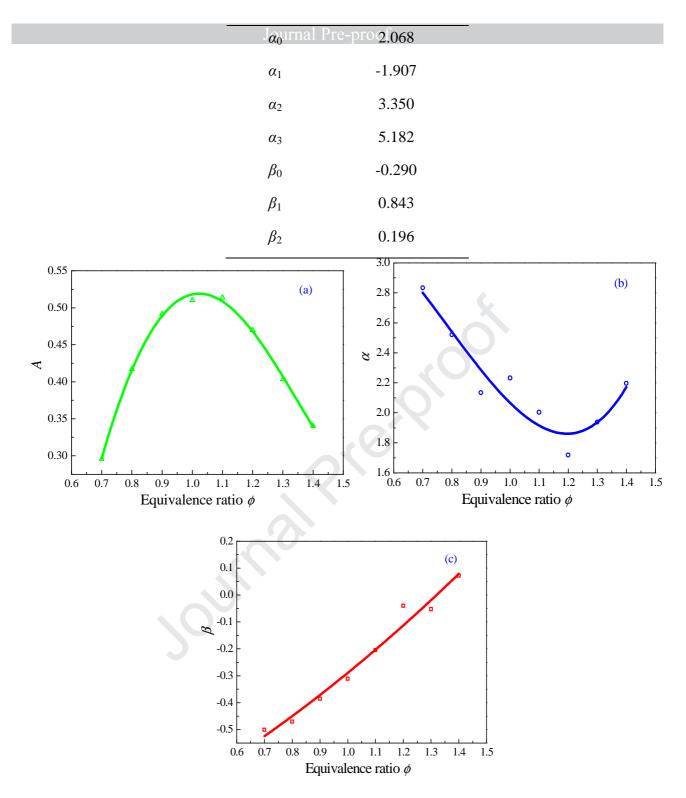


**Figure 13.**  $S_u$  as a function of *p* (0.1-0.8 MPa) and  $T_u$  (358-490 K) at  $\phi = 0.7$ , 1, and 1.4 (four figures are the same but displayed in different observation angles)

Figure 14 shows *A*,  $\alpha$  and  $\beta$  as the function of  $\phi$ . It can be observed that fitting lines show a great agreement with points at all equivalence ratios.

 Table 3
 Correlation coefficients for mixed fuel

Parameter	Value
$S_{\mathrm{u},0}$	0.518
$S_{\mathrm{u},1}$	0.087
$S_{\mathrm{u,2}}$	-1.955
$S_{\mathrm{u},3}$	1.084
$S_{\mathrm{u},4}$	1.161



**Figure 14.** *A*,  $\alpha$  and  $\beta$  as the function of  $\phi$ 

### 6 Conclusions

The burning speed of the fast pyrolysis fuel was tested in the CVCC at  $p_i$ =0.1-0.4 MPa,  $T_i$ =358-418 K, and  $\phi$ =0.7-1.4. The CVM, which takes the combustion pressure trace as the main input, was used to determine  $S_u$  for conditions beyond the initial conditions (*p* to 0.8 MPa,  $T_u$  to 490 K). The CPM, which takes Schieren flame images as the main input, was also used to determine  $S_u$ . The  $S_u$  results obtained from the CVM and CPM were compared. The main conclusions of this study are the following:

1. Discrepancies of  $S_u$  from the CVM and CPM are within 15%. The flame radius calculation in the CPM and the determination of the burned mass fraction in CVM are considered to contribute to the discrepancies in the paper.

2. Influences of temperature, pressure and equivalence ratio on  $S_u$  of the fast pyrolysis biofuel were identified. Compared with pressure, the impact of temperature on the value of  $S_u$  at most conditions is greater, so in a single experiment, with both temperature and pressure increase simultaneously,  $S_u$  tends to increases at the early stage of combustion. With increasing equivalence ratio,  $S_u$  increases first, peaks around  $\phi$ =1.0-1.1 and then decreases except when cellularity appears.

3. An explicit 12-terms correlation for  $S_u$  as the function of pressure, temperature and equivalence ratio was obtained. This correlation is effective in determination of  $S_u$  in the boundary conditions of p=0.1-0.8 MPa pressure,  $T_u=358-490$  K temperature and  $\phi=0.7-1.4$ . The maximum and average deviations between  $S_u$  from fitting correlation and  $S_u$  from experiments under  $p_i=0.1-0.4$  MPa,  $T_i=358-418$  K,  $\phi=0.7-1.4$  are 16.2% and 7.3%, respectively.

4. Cellularity appears because of the flame instability at high pressure and temperature. It led to a significant increase in flame speed. Results show pressure contributes greatly to the appearance of cellularity.

#### Acknowledgements

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## Appendix

#### Journal Pre-proof

Table S1 lists A,  $\alpha$  and  $\beta$  at various equivalence ratios.

Parameter	Α	α	β
0.7	0.30	2.83	-0.50
0.8	0.42	2.52	-0.47
0.9	0.49	2.14	-0.39
1.0	0.51	2.23	-0.31
1.1	0.51	2.00	-0.20
1.2	0.47	1.72	-0.04
1.3	0.40	1.94	-0.05
1.4	0.34	2.20	0.07

**Table S1** A,  $\alpha$  and  $\beta$  at various equivalence ratios

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#### **Research Highlights**

- Influences of T, P and  $\phi$  on Su were evaluated for a pyrolysis fuel 1.
- Laminar burning speeds (Su) determined by CVM were compared with those by CPM. 2.
- CVM interprets Su over a larger pressure and temperature range than initial conditions. 3.
- 4. An explicit correlation of Su was obtained from experimental results.
- Cellular burning speed was calculated. 5.

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