## 1 Highlights

- 21. Global extinction strain rate  $(a_g)$  decreases with increase in porous burner3diameter (D).
- Plug flow condition is valid for oxidizer inlet distance > 2 times the largest
   porous burner diameter.
- 6 3. CO/H<sub>2</sub> mixtures diluted with N<sub>2</sub> yield 1.6–2.25 times higher  $a_g$  in com-7 parison to CO/H<sub>2</sub> mixtures diluted with CO<sub>2</sub>.
- 8 4. Overall reaction rate  $(\omega_o)$  is used to explain variation in  $a_g$  values with
- <sup>9</sup> compositions.

# <sup>10</sup> Effect of burner diameter and diluents on the extinction <sup>11</sup> strain rate of syngas-air non-premixed Tsuji-type flames

S M Ali<sup>a,\*</sup>, S Varunkumar<sup>b</sup>

<sup>13</sup> <sup>a</sup>Department of Aerospace Engineering, Indian Institute of Technology Madras, Chennai -600 036

#### 17 Abstract

12

The present study focuses on the experimental determination of the global ex-18 tinction strain rate  $(a_g)$  for different syngas-air combinations using the Tsuji 19 type configuration. To study the effect of porous burner diameter (D),  $a_g$  values 20 were obtained for four values of D at atmospheric pressure. The experimentally 21 obtained  $a_g$  for a given fuel-oxidizer combination decreases with an increase in 22 burner diameter (D). This trend is consistent with the limited data available 23 in the literature for hydrocarbon fuels. Other geometric and flow-field effects 24 namely, (1) plug flow, (2) flow-field blocking by the burner, and (3) heat loss 25 by the flame to sidewalls that can affect  $a_q$  were also experimentally quantified. 26 The results from this study show that the plug flow boundary condition is al-27 ways satisfied for oxidizer inlet distance > 2 times the largest porous burner 28 diameter. Burner diameter less than 1/4 times side wall length (as is the case 29 for all burners used in this study) does not significantly modify the flow. Hence, 30 these two flow-field modifications do not affect  $a_q$ . However, heat loss from the 31 flame to the ambient through the side walls can cause a 4-9 % decrease in  $a_q$ . 32 Experiments showed that, CO/H<sub>2</sub> mixtures diluted with N<sub>2</sub> yield 1.6–2.25 times 33 higher  $a_g$  in comparison to CO/H<sub>2</sub> mixtures diluted with CO<sub>2</sub>. Increasing H<sub>2</sub> 34 from 1 to 5 % leads to 2.5–3.8 times increase in  $a_g$ , compared to 5 to 10 % 35 increase in  $H_2$  which leads to only 1.3–1.7 times increase in  $a_g$  for 70 % of  $N_2$ 36 (v/v) in fuel mixture. Global extinction strain rate  $(a_q)$  increases by 1.5–2.4 37 times with 10 % increase in CO for fuel mixtures consisting of H<sub>2</sub> (1 and 5 38

<sup>&</sup>lt;sup>15</sup> <sup>b</sup>Department of Mechanical Engineering, Indian Institute of Technology Madras, Chennai -600 036

<sup>\*</sup>Corresponding author Preprint sabmitted to International Journal of Hydrogen Energy January 16, 2020 Email address: smughees.ali@gmail.com (S M Ali )

<sup>39</sup> % by v/v), CO<sub>2</sub> (50, 60 and 70 % by v/v) and N<sub>2</sub> (50, 60, 70 and 80 % by <sup>40</sup> v/v). The change in overall reactivity ( $\omega_o$ ) due to different diluents is used to <sup>41</sup> quantitatively explain the variation of  $a_g$  for different fuel compositions. These <sup>42</sup> effects are also qualitatively explained using OH radical concentration change <sup>43</sup> with H<sub>2</sub> % in the fuel mixtures.

44 Keywords: Tsuji-type configuration; syngas-air non-premixed flames; global 45 extinction strain rate  $(a_g)$ ; burner geometry effect; diluents effect

### 46 **1. Introduction**

Gasification, a widely used thermo-chemical route for conversion of biomass and coal yields fuel gases rich in carbon monoxide and hydrogen (called syngas or producer gas). The composition of the syngas produced from gasification is highly variable. It strongly depends on the oxidizer composition used for gasification, which is generally a mixture of  $O_2/N_2/CO_2/steam$ ; fuel composition also affects the syngas composition, though not as strongly as oxidizer composition. ([1-3]).

Syngas utilization devices vary a lot in terms of applications. For instance, 54 gas-turbines used in IGCC ([4, 5]), reciprocating engines used for decentralized 55 power generation ([6-8]), gasifier based improved biomass stoves ([9, 10]) etc. 56 Therefore, extensive data on the behavior of premixed and non-premixed syngas 57 flames, covering a range of CO/H<sub>2</sub> ratios and different inert species are essential 58 for the development of syngas based combustion devices. For the determination 59 of syngas premixed flame characteristics, a significant number of computational 60 and experimental studies were performed in the past ([11-21]). Yepes and Amell 61 [22], Bouvet et al. [23] and Varghese et al. [18] have experimentally obtained 62 laminar flame speed for wide range of syngas compositions using Bunsen burner 63 configuration and heated divergent channel method respectively. The 1D nu-64 merical computations performed by Yepes and Amell [22] and Varghese et al. 65 [18] using the available kinetic mechanisms showed reasonably accurate pre-66 dictions for laminar flame speeds. Unlike the case for non-premixed flames, 67

these studies predict premixed flame characteristics are under reasonable accu-68 racy. For instance, Bilger [24] has calculated global kinetic parameters using 69 Tsuji [25] data for methane-air non-premixed flames. He has concluded that 70 the conditions for the non-premixed flames are different from those in plug flow 71 and well-stirred reactor, and therefore, rate parameters calculated from these 72 reactors should not be used for non-premixed flames. Results from CFD sim-73 ulations of the Sandia-ETH Zurich turbulent syngas jet diffusion flames ([26])74 using eight kinetic mechanisms (three simplified and five detailed) reported by 75 Marzouk and Huckaby [27] show that, none of the mechanism is capable of 76 predicting the temperature and species concentration profiles accurately. So, 77 for accurately predicting the temperature and species concentration profiles in 78 syngas non-premixed flames, the syngas kinetic mechanism must be optimized 79 using non-premixed syngas flame data. Extinction strain rate (a) is used to 80 characterize non-premixed flames. The focus of the current work is to charac-81 terize syngas non-premixed flames for various fuel-oxidizer compositions using 82 the extinction strain rate (a). 83

Extinction strain rate (a) is defined as the component of the velocity gradient 84 normal to the flame surface at extinction. Similar to laminar flame speed  $(S_L)$ 85 for the premixed flames, extinction strain rate (a) is an important characteristic 86 of non-premixed flames. Two types of measurements are commonly used to 87 characterize flame extinction, (1) the local extinction strain rate  $(a_l)$ , and (2) 88 the global extinction strain rate  $(a_q)$ . The local extinction strain rate  $(a_l)$  is 89 defined as the maximum velocity gradient normal to the flame surface just 90 upstream of the thermal layer on the oxidizer side ([28]). The global extinction 91 strain rate  $(a_q)$  is defined as the strain rate on the oxidizer side close to the 92 stagnation plane, assuming self-similarity and zero radial gradients of all scalar 93 variables along the axis of symmetry ([29]). 94

To experimentally obtain the  $a_g$  and  $a_l$ , two counterflow configurations namely, (1) opposed jet flow and (2) Tsuji-type are commonly used. Based on nozzle geometry, the opposed jet counterflow configuration can be further divided into straight and contour nozzle type configurations. The other con<sup>99</sup> figuration used to obtain  $a_g$  and  $a_l$  is the Tsuji type burner, in which flame <sup>100</sup> extinction is achieved near the stagnation point of oxidizer flow past a porous <sup>101</sup> cylinder issuing fuel (see Fig. 1).



Figure 1: Schematic of Tsuji-type configuration

This counterflow configuration is known as the Tsuji type burner as it was extensively used by Tsuji and co-workers to perform flame extinction experiments on gaseous fuels starting from 1960 till 1985 (see refs. [25, 30, 31]). For this configuration, global extinction strain rate  $(a_g)$  is given by Eqn. 1.

$$a_g = \frac{4u_{ox}}{D} \tag{1}$$

where,  $u_{ox}$  is the free stream oxidizer velocity at extinction and D is the diameter 106 of the porous burner issuing fuel. The following assumptions are used in deriving 107 Eqn. 1 - (1) potential flow, (2) change in the location of the stagnation plane due 108 to blowing at extinction is minimal, (3) flow field modification at the oxidizer 109 side of stagnation plane due to flame is neglected, and (4) flame surface is close 110 to stagnation plane ([31]). In both configurations, flame extinction is achieved 111 by increasing the total mass flow rate. The location of the flame is determined 112 by the stoichiometry and jet momentum ratio in the axial direction. 113

In the past decade, a significant number of computational and a few experimental studies are performed for syngas non-premixed flames in opposed jet configurations. The focus of these studies are primarily in three different areas namely, (1) determination of NO<sub>x</sub> emission characteristics ([32–38]), (2) effect of diluent like CO<sub>2</sub>, H<sub>2</sub>O and N<sub>2</sub> on flame structure ([39–42]) and (3) determination of extinction strain rate (a) for wide range of syngas compositions ([33, 43–46]).

Park and co-workers ([39, 40, 42]) have computationally studied the effect of 121 radiation, preferential diffusion and reactivity of diluent on non-premixed flame 122 structure for different syngas compositions. For the cases without and with 123 radiation, Park et al. [39] have found a difference of more than 100 K in flame 124 temperatures at low strain rate ( $\leq 10 \text{ s}^{-1}$ ). However, this difference reduces 125 to less than 30 K at high strain rate value ( $\geq 100 \text{ s}^{-1}$ ). In 1D computations, 126 Park et al. [40] have modified the diffusivity of  $H_2$  to be equal to the diffusivity 127 of  $N_2$  and studied its effect on flame temperature and overall reactivity. As 128  $N_2$  is less diffusive in comparison to  $H_2$ , they have termed it as suppression 129 of H<sub>2</sub> diffusivity. Their study on the effect of H<sub>2</sub> diffusivity on syngas non-130 premixed flame structure showed that, suppression of H<sub>2</sub> diffusivity should not 131 only be considered as the physical change ([40]). The higher diffusivity of  $H_2$ 132 also kinetically modifies the syngas non-premixed flame structure. Park et al. 133 [42] have shown that,  $CO_2$  cannot be considered as completely inert and  $CO_2$ 134 mole fraction is reduced by reverse CO hydroxyl path oxidation (CO<sub>2</sub> + H  $\leftrightarrow$ 135 CO + OH). Fu et al. [47] have studied the effect of preferential diffusion and 136 flame stretch on the structure of premixed Bunsen syngas flames. They have 137 found out that, for high  $H_2$  % in fuel the flame structure is affected by flame 138 curvature and preferential diffusion. 139

Hsin and co-workers have computationally studied the effect of composition, pressure and dilution on the structure of syngas non-premixed flames ([41, 43, 45]). Shih and Hsu [41] have shown, among the four effects of diluent (inert, diffusion, chemical and radiation), the inert effect is dominant in reducing flame temperature. Flame temperature decreases with an increase in

volumetric diluent percentage and as expected based on the specific heat val-145 ues, the maximum decrease in  $a_g$  was observed for CO<sub>2</sub>, followed by H<sub>2</sub>O and 146 N<sub>2</sub>. Shih and Hsu [43] results show that, at low strain rate ( $\leq 10 \text{ s}^{-1}$ ), H<sub>2</sub> 147 is consumed before CO but the flame cannot be distinguished as two separate 148 flames. Shih et al. [45] have also found out for very low strain rate  $(1 \text{ s}^{-1})$ , 149 only 0.002 % of H<sub>2</sub> is required for stable CO/H<sub>2</sub> flame. This study also shows 150 that flame cannot exist for an equimolar mixture of  $CO/H_2$  for  $O_2$  less than 151 4.7 % by volume in oxidizer stream at any strain rate value. Their observation 152 of reduction in flame temperature by radiation at low strain rate is consistent 153 with that of Yang and Shih [37]. Also, their observation about the chemical and 154 inert effect of diluent is found to be consistent with Park et al. [42] and Sahu 155 and Ravikrishna [33]. Important to note that, all these studies conducted by 156 Park's ([39, 40, 42]) and Shin's ([41, 43, 45]) group on the effect of diluents on 157 syngas non-premixed flames are for low to moderate global strain rate 10–100 158  $s^{-1}$ . In these studies, the effect of diluents on extinction was not investigated. 159

Sahu and Ravikrishna [48] have performed extinction strain rate study for 160 both premixed and non-premixed syngas flames using contour nozzle type op-161 posed jet configuration. They have shown that, global extinction strain rate 162  $(a_q)$  decreases with an increase in nozzles separation distance (L), while the 163 local extinction strain rate  $(a_l)$  does not significantly vary with L. Hence, Sahu 164 and Ravikrishna [48] have used  $a_l$  for comparison of experiments with 1D com-165 putations. Prediction of  $a_q$  from 1D computations assumes plug flow boundary 166 condition, i.e., zero velocity gradient (du/dx) in the axial direction at fuel and 167 oxidizer nozzles exit. However, Kee et al. [49] have concluded that, plug flow 168 assumption is not always valid and for predicting flame extinction accurately, 169 realistic boundary conditions are to be employed. The experimental data of 170 Sahu and Ravikrishna [48] shows a significant value for du/dx (15–40 % of  $a_l$ ) 171 at the fuel and oxidizer nozzles exit. 172

In Table 1, relevant results from earlier studies on opposed jet non-premixed syngas flames along with the available geometric parameters are compiled. For non-premixed syngas flames, data show that, out of five, three kinetic mech-

anisms namely, Li et al. [50], GRI Mech 3.0 ([51]) and Frassoldati et al. [20] 176 can predict local extinction strain rate  $(a_l)$  for low H<sub>2</sub>/CO ratio, but all these 177 kinetic models fail to predict  $a_l$  for high  $H_2/CO$  ratio. The maximum difference 178 in  $a_l$  values from experiments and 1D computations is found to as high as 25 179 % ([48]). Study performed by Som et al. [38] using three kinetics mechanisms 180 (GRI Mech 3.0, Davis et al. [52] and Mueller et al. [53]) for non-premixed and 181 partially premixed flames has shown that the kinetic model proposed by Davis 182 et al. [52] to be most accurate. For premixed flames Sahu and Ravikrishna [48] 183 have shown that, kinetic models of Davis et al. [52] and Frassoldati et al. [20] 184 predicts  $a_l$  under 10 % accuracy for all  $H_2/CO$  composition used. The data show 185 that the predicted  $a_l$  values for the premixed syngas flames are more accurate 186 in comparison to non-premixed syngas flames. As most of these kinetic mecha-187 nisms are optimized using the premixed flame characteristics, it is possible that 188 these kinetic mechanisms predict  $a_l$  values more accurately for premixed flame 189 in comparison to non-premixed flames. Bilger [24] has developed the global 190 kinetic mechanism for CH<sub>4</sub> oxidation by analyzing the methane non-premixed 191 data obtained using the Tsuji configuration. He has found out a significant 192 difference in the kinetic parameters calculated using non-premixed Tsuji config-193 uration from that of the well-stirred (Dryer and Glassman [54]) and plug flow 194 reactor (Williams et al. [55]). Bilger [24] has concluded that the conditions for 195 a non-premixed flames are different from those in plug flow and well-stirred re-196 actor and therefore, rate parameters calculated from these reactors should not 197 be used for non-premixed flames. 198

	Configuration	Experiments	1D computation
Fuel	$L(\mathrm{mm})/D(\mathrm{mm})$	$a_g$ or $a_l$ (s <sup>-1</sup> )	$a_g$ or $a_l$ (s <sup>-1</sup> )
(% by volume)	Contour Nozzle		BCs (Potential)
		Sahu and Ravikrishna [48]	
$35 {\rm CO}/05 {\rm H}_2/02 {\rm CH}_4/58 {\rm N}_2$	17/30	$1230{\pm}25$	-
	17/30	$619^{*}{\pm}25$	$500^*, 520^*, 608^*, 539^*, 681^*$
$32 {\rm CO}/08 {\rm H}_2/02 {\rm CH}_4/58 {\rm N}_2$	7/30	$2497{\pm}210$	-
	7.5/30	$2158 {\pm} 308$	-
	8/30	$2141 \pm 325$	-
	12/30	$1928 {\pm} 277$	-
	-/-	$826^{*} \pm 33$	$687^{*}, 683^{*}, 794^{*}, 779^{*}, 911^{*}$
$29 {\rm CO}/11 {\rm H}_2/02 {\rm CH}_4/58 {\rm N}_2$	-/-	$966^{*}{\pm}38$	$966^{*}, 922^{*}, 1024^{*}, 1068^{*}, 1168^{*}$
$26{\rm CO}/14{\rm H}_2/02{\rm CH}_4/58{\rm N}_2$	10.5/30	$2411 \pm 39$	-
$26{\rm CO}/14{\rm H}_2/02{\rm CH}_4/58{\rm N}_2$	-/-	$1149^{*} \pm 45$	$1235^*, 1201^*, 1284^*, 1357^*, 1460^*$
$23 {\rm CO}/17 {\rm H}_2/02 {\rm CH}_4/58 {\rm N}_2$	-/-	$1773^{*} \pm 70$	$1543^{*}, 1542^{*}, 1602^{*}, 1657^{*}, 1758^{*}$
$20{\rm CO}/20{\rm H}_2/02{\rm CH}_4/58{\rm N}_2$	-/-	$2300^* \pm 90$	$1842^{*}, 1944^{*}, 1954^{*}, 1964^{*}, 2091^{*}$
		Wang et al. [46]	
	Straight nozzle	Uniform	2D Computations
Syn1-60N2	10.2/10.2	$820{\pm}136$	643, <i>661</i> *
Syn1-65N2	10.2/10.2	$647 \pm 70$	$408,413^{*}$
Syn1-70N2	10.2/10.2	$461 \pm 86$	214,221*
		Parabolic	
Syn1-50N2	10.2/11.2	$860{\pm}63$	$777,735^{*}$
Syn1-55N2	10.2/11.2	$645 \pm 45$	$537,581^{*}$
Syn1-60N2	10.2/11.2	$460 \pm 50$	$322, 382^*$
Syn1-65N2	10.2/11.2	$311{\pm}50$	$158,224^*$
Syn1-70N2	10.2/11.2	$179 \pm 41$	$95,109^*$
Syn2-60N2	10.2/11.2	$856 {\pm} 66$	$752,736^{*}$
Syn2-65N2	10.2/11.2	$625 \pm 50$	$398,481^{*}$
Syn2-70N2	10.2/11.2	$365 {\pm} 45$	$180,235^{*}$
Syn2-75N2	10.2/11.2	$200{\pm}54$	80,89*

Table 1: Summary of the  $a_g$  and  $a_l$  for the two opposed jet configurations. Error value is estimated by symbol size if in case not provided. Extinction strain rate values in italics with "\*" represents  $a_l$ , BCs:- Boundary conditions, var:- Variable

The two possible reasons for the difference in  $a_l$  values between experimental and 1D computations are uncertainties in determination/optimization of transport properties and kinetic parameters. Sahu and Ravikrishna [48] have stated that, "differences of 15 % and higher are observed in predictions of extinction strain rates by the various mechanisms despite the use of similar transport li<sup>204</sup> braries." From this, Sahu and Ravikrishna [33] has concluded that improvement
<sup>205</sup> in both kinetic parameter and transport properties are required for better pre<sup>206</sup> diction of extinction of non-premixed flames.

Wang et al. [46] have obtained global extinction strain rate  $(a_q)$  experimen-207 tally and compared them with 2D axisymmetric computations for syngas non-208 premixed opposed jet flames. They have used two different types of boundary 209 conditions (parabolic and uniform) to obtain  $a_g$  and compared them with 2D 210 computations. Using 2D axisymmetric computations, they have shown that, the 211 difference in assumed and actual axial centerline velocity increases with an in-212 crease in global strain rate for parabolic inflow velocity. However, for uniform 213 inflow axial velocity, a constant difference of about 16 % between assumed and 214 actual centerline axial velocity is observed for three global strain rate values. 215 The computational data of Wang et al. [46] shows a constant difference of about 216 16~% between assumed and actual centerline axial velocity which is due to en-217 forcement of ideal uniform axial velocity profile at a distance of 5.1 mm from 218 the nozzle exit. Wang et al. [46] have concluded that, "it is very doubtful that 219 the matrix is capable of producing such a uniform flow profile in the actual 220 experiments, particularly for elevated flow conditions." Hence, for accurately 221 predicting  $a_q$ , idealized velocity profiles (parabolic or uniform) at the nozzles 222 exit can not be used. The analysis of Wang et al. [46] data from Table 1 shows 223 that, indeed 2D axisymmetric computations are able to predict the trends, but 224 under-predict  $a_q$  values for all the fuel-air compositions used. The maximum 225 difference in  $a_g$  values from experiment and 2D computation is found out to be 226 as high as 60 %. 227

The other simplified geometry used to study non-premixed flames is Tsujitype configuration. This configuration has an advantage over opposed-jet counterflow configuration in terms of flow field modification by the flame near the oxidizer inlet. In Tsuji-type configuration, if the oxidizer boundary is placed sufficiently far from the burner, it will not interact with flame. One of the objectives of the current work is to determine this minimum distance between the oxidizer boundary and the porous cylinder burner. Tsuji and co-workers ([25, 30, 31]) have obtained  $a_g$  values experimentally for CH<sub>4</sub> and C<sub>3</sub>H<sub>8</sub>, but to the best of our knowledge, we have not come across any study on determination of  $a_g$  for different CO/H<sub>2</sub> compositions.

In the current study,  $a_g$  values are experimentally obtained using Tsuji type 238 configuration for a range of syngas compositions with  $N_2$  and  $CO_2$  as diluents. 239 The composition range covered in this study is - CO from 10 to 49 % (v/v), H<sub>2</sub> 240 from 1 to 10 % (v/v), N\_2 dilution of 20 to 50 % (v/v) and CO\_2 dilution of 30-50 241 % (v/v). The choice of the composition range investigated here is based on 242 the following considerations- (1) very high to low CO/H<sub>2</sub> ratios (49–1) diluted 243 with  $CO_2$  and  $N_2$ , (2) syngas compositions obtained using  $O_2/N_2/CO_2$ /steam 244 as oxidizer for biomass gasification ([1, 2, 9, 10, 56]) (3) few compositions were 245 chosen from the study of Wang et al. [46] to compare the  $a_g$  values obtained 246 from Tsuji burner with opposed jet configuration. 247

#### 248 2. Experimental Methodology

A schematic of the counterflow Tsuji type burner setup used in the current work is shown in Fig. 2. The burner is designed according to the dimensions proposed by Tsuji and Yamaoka [30]; an additional flow-straightener is used just after the convergent nozzle for ensuring uniform laminar flow. The details of the design are explained below.



Figure 2: Schematic of the experimental setup

The counterflow setup consists of five parts, namely, the combustion cham-254 ber, porous cylinder, flow straighteners, convergent nozzle and the settling cham-255 ber. The combustion chamber (12 cm  $\times$  25 cm  $\times$  3 cm) ( $l \times h \times w$ ) was fixed 256 with quartz windows (10 cm  $\times$  10 cm) along its length for flame visualization. 257 The sides of the combustion chamber consist of two slots  $(10 \text{ cm} \times 2.5 \text{ cm})$  which 258 were generally closed but can be open when required. The porous cylinder was 259 fixed inside the combustion chamber with a holding mechanism such that the 260 distance of the porous cylinder burner from the bottom of the combustion cham-261 ber can be varied. Fuel was discharged only from the porous area of the burner 262 that is  $\pi/3$  degrees on either side of the stagnation point. The sidewall openings 263 in the combustion chamber were covered with a wire mesh and the porous cylin-264 der burner was kept at a distance of 12 cm from the bottom of the combustion 265 chamber (oxidizer inlet) unless otherwise mentioned. Four porous cylinders of 266 length 3 cm and diameters 10.6 mm, 15.6 mm, 22.8 mm and 30.8 mm were used 267

for the experiments. The bottom section of the combustion chamber is attached 268 to a straight rectangular duct of 5 cm height. This duct is further attached to 269 flow straighteners of 5 cm height. These flow straighteners consist of a bundle 270 of metal tubes of diameter 0.25 mm and length 5 cm which maintain a laminar 271 flow at the duct exit. The lower section of these flow straighteners was attached 272 to a convergent rectangular nozzle connected to the settling chamber (25 cm  $\times$ 273  $25 \text{ cm} \times 25 \text{ cm}$ ). The settling chamber was also fixed with flow straighteners to 274 reduce local turbulence and maintain uniform laminar flow at the nozzle entry. 275 For metering and controlling the fuel and air supply, mass flow controllers 276 with an accuracy of 1 % of the full-scale reading was used. Fuel gases were 277 supplied from pressurized gas cylinders fixed with a two-stage pressure regulator. 278 An upstream pressure of 5 bar was maintained just before gas filters. Gas filters 279 of 5  $\mu$ m mesh size were fixed upstream of the MFCs to remove any particulate 280 impurity coming from the pressurized fuel gas cylinders. Carbon monoxide, 281 hydrogen, carbon dioxide, nitrogen with 99.9 % and methane with 99.5 % purity 282 levels were mixed in different proportions and used as fuel. Experiments were 283 conducted with methane (pure and diluted with  $N_2$ ) and syngas (with different 284  $CO/H_2$  ratios) diluted with  $N_2$  and  $CO_2$ . The fuel compositions used in this 285 study are listed in Table 2. 286

Nomenclature	$CH_4 (\%)^*$	CO (%)	$H_2$ (%)	$\mathrm{CO}_2~(\%)$	$N_2 \ (\%)$
$CH_4$	100	0	0	0	0
$\rm CH_4{-}30N_2$	70	0	0	0	30
$\rm CH_4-50N_2$	50	0	0	0	50
$CH_4-70N_2$	30	0	0	0	70
$Syn1-55N_2$	0	22.5	11.25	11.25	55
$Syn1-65N_2$	0	17.5	8.75	8.75	65
$Syn2-60N_2$	0	15	15	10	60
$\rm Syn2–70N_2$	0	11.25	11.25	7.5	70
$\rm Syn{-}5H_280N_2$	0	15	5	0	80
$\rm Syn{-}10\rm H_280\rm N_2$	0	10	10	0	80
$\rm Syn{-}1\rm H_270\rm N_2$	0	29	1	0	70
$\rm Syn{-}5H_270N_2$	0	25	5	0	70
$\rm Syn{-}10\rm H_270\rm N_2$	0	20	10	0	70
$\rm Syn{-}1\rm H_260\rm N_2$	0	39	1	0	60
$Syn-1H_250N_2$	0	49	1	0	50
$Syn-5H_270CO_2$	0	25	5	0	70
$\rm Syn{-}10\rm H_270\rm CO_2$	0	20	10	0	70
$Syn-5H_260CO_2$	0	35	5	0	60
$\rm Syn-1H_260CO_2$	0	39	1	0	60
$Syn-1H_250CO_2$	0	49	1	0	50

Table 2: Nomenclature for  $\rm CH_4/N_2$  and  $\rm CO/H_2/N_2/CO_2$  blends used as fuel. \*All % are in volumetric basis

Air is supplied to the combustion chamber from a compressor. Four inlets 287 at the bottom of the settling chamber are used for maintaining a uniform flow 288 distribution at the flow straighteners entry. CO oxidation takes place by two 289 pathways namely, (1) direct oxidation (CO + O  $\leftrightarrow$  CO<sub>2</sub>, CO + O + M  $\leftrightarrow$ 290  $CO_2 + M$ ) and (2) hydroxyl oxidation (CO + OH  $\leftrightarrow$  CO<sub>2</sub> + H). For pure CO 291 flames, the hydroxyl oxidation pathway is very sensitive to moisture content 292 present in the oxidizer, hence accurate determination of moisture is required. 293 Moisture filter was used to remove water vapor from the air and a humidity 294 sensor was used to measure water vapor content. It was found that water vapor 295 volume fraction does not exceed 1 %. Temperature of fuel gas issuing out of the 296

<sup>297</sup> porous cylinder is measured using a K-type thermocouple (0.25 mm bead size). <sup>298</sup> This was required to quantify the effect of fuel heating due to the flame (details <sup>299</sup> discussed in section 2.1). Temperature data were recorded using the computer <sup>300</sup> interfaced data logger at a frequency of 2 Hz. The flame images and videos were <sup>301</sup> recorded using a digital camera; the lens was placed at a distance of 40 cm from <sup>302</sup> the combustion chamber aligned with the quartz windows.

#### 303 2.1. Experimental procedure

As heating of porous cylinder affects the  $a_q$  values, experiments were per-304 formed in two ways - (1) global extinction strain rate  $(a_q)$  measurement for 305 steady inlet fuel temperature issuing from porous cylinder termed as M1 or 306 method 1 and, (2) global extinction strain rate  $(a_a)$  measurement with minimal 307 inlet fuel heating termed as M2 or method 2. The experimental procedure to 308 measure  $a_g$  to quantify these effects is given below. For a particular fuel-oxidizer 309 combination, experiments were repeated at least five times (maximum deviation 310 was < 5 % of the average value of  $a_q$ ). Uncertainty in the measurement of  $a_q$ 311 values is equal to 1 % of full-scale reading of mass flow controller used for oxi-312 dizer flow rate measurement. Range of mass flow controller used for measuring 313 the oxidizer flow rate is 0-1000 lpm. For this range, uncertainties in the mea-314 surement of  $a_q$  values are 17, 12, 8 and 6 s<sup>-1</sup> for 10.6, 15.6, 22.8 and 30.8 mm 315 porous burner diameter respectively. 316

To measure  $a_q$  using Method 1 (M1), a stable flame was established around 317 the porous cylinder burner and fuel inlet pipe at low strain rate. The location of 318 flame was maintained sufficiently far ( $\approx 20-25$  mm) to avoid direct heat transfer 319 from the flame surface to the porous cylinder fuel inlet. An oxidizer flow rate 320 of 50 lpm which corresponds to oxidizer free stream velocity of 0.23 m/s (equal 321 to 30  $\rm s^{-1}$  for 3.08 cm porous burner diameter) and fuel velocity of 0.33 m/s 322 was found to be suitable for avoiding heat loss by flames to the porous burner. 323 However, as the fuel was discharged only from the porous area of the burner 324  $(\pi/3 \text{ degrees on either side of the stagnation point})$ , heat transfer from the flame 325 to the non-porous side of burner causes an increase in fuel temperature from 326

300 K to a steady value. This steady temperature value is dependent on the 327 type of fuel and diluent used (for instance, 490 K for  $CH_4$ -70N<sub>2</sub> for 3.08 cm 328 porous burner diameter). Steady-state temperature increases with the decrease 329 in  $N_2$  percentage in the fuel-inert mixture. Due to this, extinction of other three 330 CH<sub>4</sub>-N<sub>2</sub> mixture used for this study occurs at higher airflow rates, which was 331 beyond the available compressor capacity (maximum flow rate the compressor 332 can supply is 650 lpm, which corresponds to 390  $\rm s^{-1}$  for porous burner of di-333 ameter of 3.08 cm). So, using method 1 (M1) extinction cannot be achieved 334 for the other three  $CH_4-N_2$  and some syngas mixtures. Hence, experiments 335 using method 1 (M1) were performed only for the compositions ( $CH_4$ - 70 $N_2$ , 336  $Syn-1H_270N_2$ ,  $Syn-5H_270N_2$ ,  $Syn-5H_280N_2$ ,  $Syn-10H_280N_2$ ) which are under 337 the range of compressor capacity (refer to Tables A.4 and A.5 of Appendix A 338 for data). 339

Once the fuel inlet temperature reaches a steady value, airflow rate was increased to achieve extinction. With the increase in oxidizer flow rate the burner temperature increases, however, difference in fuel inlet temperature from the start till flame extinction was about 10-15 K. This shows that there was minimal direct heat transfer from the flame surface to the fuel inlet once steady state was reached and flame extinction occurs when the mixing time for fuel and oxidizer becomes comparable to reaction time.

Figure 3 shows non-premixed flames stabilized over the porous cylinder burner of diameter 3.08 cm for  $CH_4$ -70N<sub>2</sub>-air combination obtained using M1. In the figure shown, as we move from left to right, airflow rate increases causing the flame to move closer to the porous burner until extinction occurs. At this moment, the measured air flow rate was recorded and used to calculate  $a_a$ .



Figure 3: Combined image for Tsuji type non-premixed flames (D = 3.08 cm, CH<sub>4</sub>-70N<sub>2</sub>-air combination) obtained using method 1 (M1) with increasing global strain rates until extinction

The other set of experiments were performed to measure the global extinc-352 tion strain rate with minimal heating of inlet fuel termed as M2 or method 2. 353 Initially, at low strain rate, a stable flame was established around the burner. 35 Once the stable flame was established, the oxidizer flow was increased ( $\approx$  75– 355  $100 \text{ s}^{-1}/\text{s}$ ) until extinction. At this moment, flow rate and temperature were 356 recorded. Immediately after extinction, fuel temperature was found to be in 357 the range of 305–340 K. To minimize the effect of heating after each reading, 358 both the combustion chamber and porous cylinder were cooled down to room 359 temperature. Once cooled down to room temperature, the experiment was re-360 peated for a new reading (refer to Tables A.4, A.5, A.6 A.7, A.8, A.9, A.10 and 361 A.11 of Appendix A for data). The  $a_g$  values obtained from M1 (steady fuel 362 inlet temperature) is 40-60 % higher when compared to M2 (minimal heating 363 case). The  $a_q$  data from the past literature is compared with  $a_q$  data from the 364 M2 (see Table 3). This is due to the fact that,  $a_q$  data obtained using method 2 365 (M2) is with minimal heating effects, which is the case with most data available 366 from the past literature ([25, 28, 30, 31, 57]). 367

Table 3 shows the compiled data for  $a_g$  and its variation with respect to Dfrom literature and current work. Tsuji and co-workers have performed experiments on three fuel-air combinations namely, CH<sub>4</sub>-air, C<sub>3</sub>H<sub>8</sub>-air and citygas-air ([25]).

Evel and dimen		Experiments	1D computation
Fuel-oxidizer	$D \ (\mathrm{mm})$	$a_g \; ({\rm s}^{-1})$	$a_g \ (s^{-1})$
		Tsuji and coworkers	
$CH_4$ -air	60, 15	320, 375	460, 320, 350, 400-405
		[25, 31]	[28, 57]
$\rm C_3H_8-air$	60, 45,	365, 420[30]	-
	30, 15	486, 650[30] -	
		Ali and Varunkumar [58]	-
$\rm CH_4{-}50N_2{-}air$	30, 15	210, 350	-
$\rm CH_4-70N_2-air$	60, 45	85,100	-
	30,  15	120, 190	-
		Current work	-
$\rm CH_4-70N_2-air$	30.8, 22.8	170, 211	
	15.6,10.6	244, 276	-
$\rm CH_4-N_2-air$	30.8, 22.8	277, 349	
	15.6,10.6	443, 515	-
$\rm CH_4{-}30N_2{-}air$	30.8, 22.8	351, 429	
	15.6,10.6	539, 620	-
$CH_4-air$	30.8, 22.8	380, 463	
	15.6, 10.6	614, 756	-

Table 3: Summary of the  $a_g$  (s<sup>-1</sup>) for Tsuji type configuration

Tsuji and co-workers have performed extensive studies on Tsuji-type configuration, but the only systematic study on variation of  $a_g$  with porous burner diameter (D) the authors came across is for C<sub>3</sub>H<sub>8</sub>-air combination ([30]). From the data shown in Table 3, it is clear that  $a_g$  decreases with increase in D. This decreasing trend for  $a_g$  is also verified by Ali and Varunkumar [58] for CH<sub>4</sub>-70N<sub>2</sub>-air and CH<sub>4</sub>-50N<sub>2</sub>-air combinations and also from the current experiments (see Fig. 4).

Figure 4 shows the  $a_g$  values for four different CH<sub>4</sub>-N<sub>2</sub>-air combinations from the current work. The  $a_g$  data from the current experiments show that if

 $_{381}$  D is decreased from 30.8 to 10.6 mm, the values of  $a_g$  increases approximately

<sub>382</sub> by a factor of 2.



Figure 4: Variation of experimentally obtained  $a_g$  with D for four CH<sub>4</sub>–N<sub>2</sub>-air combinations. Note: Error bars less then symbol size are eliminated

This observation is consistent with data from Tsuji and Yamaoka [30] for 383  $C_3H_8$ -air. However,  $a_g$  data obtained by Tsuji and Yamaoka [31] and Tsuji 384 [25] for  $CH_4$ -air combination do not show a significant increase in  $a_q$  with the 385 same decrease in D. Analysis of Tsuji and Yamaoka [31] and Tsuji [25] data 386 for  $CH_4$ -air combination shows that, as D is reduced 4 times (6 cm to 1.5 cm), 387  $a_g$  increases only by 17 %. This increase in  $a_g$  with decrease in D for CH<sub>4</sub>-air 388 combination is inconsistent with data from current work and that for  $C_3H_8$ -air 389  $a_q$  data (increase by factor of 2) from Tsuji and Yamaoka [30]; hence data for 390 CH<sub>4</sub>-air from Tsuji and Yamaoka [31] is not used for the current analysis. The 391 reason for 15–20 % increase in the current  $a_g$  values compared to data reported 392 in Ali and Varunkumar [58] is the reduction in non-uniformity in upstream flow 393 in the current experimental setup. 394

As reported by Kee et al. [49], Sarnacki et al. [59] and Wang et al. [46], one of the main reasons for difference in  $a_q$  between experiments and 1D computation <sup>397</sup> in opposed jet configuration is due to failure of plug flow boundary condition. <sup>398</sup> The porous cylinder was kept at three different values (12 cm, 10 cm and 6 cm) <sup>399</sup> from the oxidizer inlet and  $a_g$  experiments were repeated using method 2 (M2) <sup>400</sup> for validating the plug flow condition in Tsuji burner.



Figure 5: Variation of experimentally obtained  $a_g$  with D for CH<sub>4</sub>-air combinations for three porous cylinder location

The data showed that the measured values of  $a_q$  for three distances are 401 within 10 s<sup>-1</sup> which corresponds to  $\pm 3$  % of average  $a_q$  (refer to Table A.8 402 of Appendix A for data). Hence, for the Tsuji type configuration, plug flow 403 boundary condition is valid for the conditions investigated in the current work. 404 The other two effects which can also modify the flame surface are, (1) flow-field 405 modification by the blocking effect of a porous burner and (2) heat loss by the 406 flame surface to the adjacent walls. To find the effect of flow-field modification 407 by burner blocking on  $a_g$ , the wire meshes covering from the side walls were 408 removed and experiments were repeated. The data show slightly higher  $a_q$ 409 values for the removed wall case, but the maximum difference was  $11 \text{ s}^{-1}$ . This 410 largest deviation in the  $a_g$  was less than 3 % (< 3 %) of the measured value 411 (refer to Table A.9 of Appendix A for data). 412



Figure 6: Variation of experimentally obtained  $a_g$  with D for CH<sub>4</sub>-air combinations with and without side mesh walls

The largest porous cylinder diameter is one-fourth (1/4) of the combustion 413 chamber width; this suggests that the flow blocking effect can be neglected for 414 the porous cylinder diameters used in the current work. To find the effect of heat 415 loss by the flame to adjacent walls on  $a_g$ , experiments are performed by cov-416 ering the burner casing with a thick cerawool insulation and these results were 417 compared with experiments without insulation. Figure 7 shows the comparison 418 of  $a_g$  values for Syn–10H\_280N\_2-air combinations with and without insulation 419 using method 2 (M2). 420



Figure 7: Variation of experimentally obtained  $a_g$  with D for Syn-10H<sub>2</sub>80CO<sub>2</sub>-air combinations with and without insulation using method 2 (M2)

The data show that,  $a_g$  values obtained using insulated burner are about 16–36 s<sup>-1</sup> higher compared to non-insulated burner (refer to Table A.10 in Appendix A for data). This correspond to 4-9 % of average  $a_g$  for Syn–10H<sub>2</sub>80N<sub>2</sub>air combination. So, while comparing with computations for Tsuji-type configuration, we can say a maximum of 10 % decrease in  $a_g$  can be explained through heat loss by flame.

#### 427 3. Results and discussion

This section presents the results and discussion on the effect of porous cylinder diameter (D), diluent fraction of species (CO<sub>2</sub> and N<sub>2</sub>) in fuel and CO/H<sub>2</sub> ratio on experimentally determined values of  $a_q$ .

 $_{431}$  3.1. Effect of porous burner diameter (D)

Figure 8 shows the variation of  $a_g$  values with D obtained using Method 1 (M1) for CH<sub>4</sub> and two CO/H<sub>2</sub> mixtures diluted with 70 % of N<sub>2</sub>. The data show that,  $a_g$  decreases with increase in D for all three fuel-oxidizer combinations. Figure 8 shows that,  $a_g$  increases by a factor of 1.7–1.8 with decrease in porous cylinder diameter (*D*) from 30.8 to 10.6 mm. This increasing trend is also consistent with  $a_g$  data for syngas-air and CH<sub>4</sub>–N<sub>2</sub>-air combinations obtained using M2 (see Figs. 4 and 9). The data also shows that, the fuel inlet temperature for CH<sub>4</sub>–70N<sub>2</sub> mixture is higher in comparison to Syn–1H<sub>2</sub>70N<sub>2</sub> and Syn–5H<sub>2</sub>70N<sub>2</sub> mixtures due to overall higher integrated heat release.



Figure 8: Variation of experimental  $a_g$  data with D obtained using Method 1 (M1) for syngasair and methane/nitrogen-air combinations. Error bars are removed for clarity

The values of  $a_q$  obtained for CH<sub>4</sub>-70N<sub>2</sub> is in between Syn-1H<sub>2</sub>70N<sub>2</sub> and 441  $Syn-5H_270N_2$  mixtures. This implies that, compared to  $CH_4-70N_2$ -air the over-442 all reactivity of  $Syn-1H_270N_2$ -air is lower and  $Syn-5H_270N_2$ -air is higher for the 443 same percentage of diluent in the fuel (70 % N<sub>2</sub> v/v). Wang et al. [46] have 444 obtained  $a_g$  data for opposed jet non-premixed syngas flames for a wide range 445 of compositions; comparisons have also been drawn with results from 2D axi-446 symmetric computations from their study. In the current study, we have chosen 447 two compositions from Wang et al. [46] (referred to here as  $\mathrm{Syn2-70N_2}$  and 448 Syn1–65N<sub>2</sub>, see Table 2 for details) to measure  $a_g$  using Tsuji burner. 449

Figures 9a and 9b show the variation of experimentally obtained  $a_g$  with 450 D for Syn1–65N<sub>2</sub>-air and Syn2–70N<sub>2</sub>-air compositions using method 2 (M2). 451 The data show that, with decrease in D from 30.8 to 10.6 mm  $a_g$  increases 452 by a factor of 2.15–2.25. For Syn1–65N<sub>2</sub>-air combination,  $a_g$  value obtained 453 experimentally using Tsuji burner  $(604\pm24 \text{ s}^{-1})$  for 10.6 mm diameter is close 454 to  $a_q$  value (647±70 s<sup>-1</sup>) obtained by Wang et al. [46] for a uniform velocity 455 profile for nozzle separation distance of 10.2 mm. For parabolic velocity pro-456 file, for the same composition experimental obtained  $a_g$  value  $(311\pm50 \text{ s}^{-1})$  by 457 Wang et al. [46] is close to  $a_g$  value (295±9  $\rm s^{-1})$  obtained using Tsuji burner 458 of 22.8 mm diameter. Understanding the connections between the extinction 459 strain rates obtained from different configurations and its interpretation for the 460 optimization/validation of kinetic mechanisms require further experimental and 461 computational studies. 462



(b)  $CO/H_2 = 1$  (30 % by volume)

Figure 9: Experimentally obtained  $a_g$  values for Syn1–65N<sub>2</sub> and Syn2–70N<sub>2</sub> using method 2 (M2) for Tsuji type configuration

The analysis of computational and experimental data from Wang et al. [46] for these two compositions  $(Syn1-65N_2 \text{ and } Syn2-30N_2)$  show that, 2D axi-

symmetric computations under predict  $a_g$  by at least 50 % of experimentally 465 obtained values (see Table A.4 in Appendix A for data). They have proposed 466 that the possible reason for this under-prediction is failure of the assumption 467 of top-hat/parabolic velocity profiles at the nozzles exit. The experiments per-468 formed in the current study using Tsuji type configuration have shown that the 469 flow field around the flame is not modified by the oxidizer boundary if the dis-470 tance between the porous burner and oxidizer inlet is greater than 2 times the 471 largest burner diameter (D) (see Fig. 5). Hence, experimental  $a_g$  data obtained 472 using Tsuji-type configuration is perhaps better suited for predicting extinction 473 using 2D planar computation without concerning about the validity of boundary 474 conditions. 475

## 476 3.2. Effect of diluent on $a_g$ - $N_2$ vs $CO_2$

Figures 10a and 10b show the comparison of  $a_g$  values obtained by method 2 (M2) using two different CO/H<sub>2</sub> ratios (5 and 2) for a fixed diluent percentage. The data show that for all porous burner diameters (*D*) and CO/H<sub>2</sub> ratio used,  $a_g$  values obtained using N<sub>2</sub> as diluent are always 1.6–2.25 times the  $a_g$  values obtained using CO<sub>2</sub> as diluent. To explain this difference in  $a_g$  values, an overall reaction rate ( $\omega_o$ ) is calculated using a two step kinetic mechanism proposed by Slavinskaya et al. [60].

Overall reaction

(0)  $\alpha CO + \beta H_2 + 0.5(\alpha + \beta)O_2 \rightarrow \alpha CO_2 + \beta H_2O$ Slavinskaya two-step mechanism

(I)  $2H_2 + O_2 \rightarrow 2H_2O$   $10.8e^{-7}T^{6.1}exp(-9684.5/RT)[H_2]^2$ 

(I) 
$$CO + H_2 + O_2 \rightarrow CO_2 + H_2O$$
 20.15e<sup>-8</sup>T<sup>5.9</sup>exp(-6097.6/RT)[O<sub>2</sub>][CO]<sup>1.4</sup>

For the given two-step kinetic mechanism, H<sub>2</sub> and CO consumption rates are  $\omega_{H_2} = 2\omega_I + \omega_{II}$  and  $\omega_{CO} = \omega_{II}$ , where  $\omega_I$  and  $\omega_{II}$  are the rates of these two reactions. The units used for the kinetic mechanism are: mole, cm<sup>3</sup>, sec, K and cal. In the overall reaction,  $\alpha$  and  $\beta$  are the volumetric fractions of CO and H<sub>2</sub> present in the fuel. The overall reaction rate is calculated by equating the
heat release rate of Slavinskaya et al. [60] mechanism with an overall reaction.

$$\omega_o = \frac{2\omega_I(h_{f,H_2O}^o) + \omega_{II}(h_{f,CO_2}^o + h_{f,H_2O}^o - h_{f,CO}^o)}{\beta(h_{f,H_2O}^o) + \alpha(h_{f,CO_2}^o - h_{f,CO}^o)}$$
(2)

In the Eqn 2,  $h_{f,H_2O}^o$ ,  $h_{f,CO_2}^o$  and  $h_{f,CO}^o$  are standard heat of formation of H<sub>2</sub>O, CO<sub>2</sub> and CO. The reaction rates  $\omega_I$  and  $\omega_{II}$  are given by Eqns. 3 and 4 when expressed in terms of mole fractions  $(X_{H_2}, X_{CO_2} \text{ and } X_{H_2O})$ , pressure (P) and temperature (T).

$$\omega_I = 10.8e^{-7}T^{6.1}exp\left(\frac{-9684.5}{RT}\right)\left(\frac{P}{R_uT}X_{H_2}\right)^2\left(\frac{mole}{cm^3s}\right)$$
(3)

494

$$\omega_{II} = 20.15e^{-8}T^{5.9}exp\left(\frac{-6097.6}{RT}\right)\left(\frac{P}{R_uT}\right)^{2.4}X_{O_2}(X_{CO})^{1.4}\left(\frac{mole}{cm^3s}\right)$$
(4)

495

$$X_{H_2} = \frac{\beta}{1 + [2.38(\alpha + \beta)]/\phi}$$
(5)

496

$$X_{CO} = \frac{\alpha}{1 + [2.38(\alpha + \beta)]/\phi} \tag{6}$$

497

$$X_{O_2} = \frac{(\alpha + \beta)/2\phi}{1 + [2.38(\alpha + \beta)]/\phi}$$
(7)

where,  $\alpha$ ,  $\beta$  represents volumetric percentage of CO and H<sub>2</sub> in fuel and  $\phi$  repre-498 sents the equivalence ratio. The location of the non-premixed flame is assumed 499 at  $\phi = 1$ . The equilibrium temperature obtained for Syn-5H<sub>2</sub>70N<sub>2</sub>-air and 500  $Syn-5H_270CO_2$ -air combinations using NASA SP-273 software ([61]) for con-501 stant pressure condition is 1760 K and 1500 K respectively. With the increase 502 in flame temperature from 1500 to 1760 K, the rate of two reactions ( $\omega_I$  and 503  $\omega_{II}$ ) increases by factor of 3.3 and 2.4 respectively. The overall reaction rate 504  $(\omega_o)$  of the mixture with N<sub>2</sub> is more than that of the one with CO<sub>2</sub> by a factor 505 of 3.1. This is consistent with the observed differences in the global strain rate. 506



(b)  $CO/H_2 = 2$  (30 % by volume)

Figure 10: Comparison of experimentally obtained  $a_g$  using method 2 (M2) diluted with  $\rm N_2$  and CO\_2 for different D values

## 507 3.3. Effect of $H_2$ and CO % in fuel

Figures 11a and 11b show the effect of volumetric  $H_2$  % on  $a_g$  values ob-508 tained by Method 2 (M2) using  $N_2$  as diluent (70 % and 80 % by volume). The 509 data show that,  $a_g$  increases approximately 1.3–1.7 times when volumetric H<sub>2</sub> 510 % is increased from 5 to 10 % for 70 % N<sub>2</sub> (v/v) in fuel mixture. However,  $a_q$ 511 increases by 2.5–3.8 times approximately, if the volumetric  $H_2$  % is increased 512 from 1 to 5 %. Extinction strain rate  $(a_q)$  increases by 3–3.8 times approxi-513 mately, if the volumetric  $H_2$  % is increased from 5 to 10 % for 80 %  $N_2$  (v/v) in 514 fuel mixture. The ratios of overall reaction rates ( $\omega_o$ ) calculated using Eqn. 2 515 for compositions  $Syn-5H_270N_2/Syn-1H_270N_2$  and  $Syn-10H_270N_2/Syn-5H_270N_2$ 516 are 10.4 and 3.6. This shows that,  $a_g$  increases non-linearly ( $\approx \sqrt{\omega_o}$ ) with in-517 crease in overall reactivity. For both  $H_2$  and CO, hydroxyl pathway (CO + OH 518  $\leftrightarrow$  CO<sub>2</sub> + H, H<sub>2</sub> + OH  $\leftrightarrow$  H<sub>2</sub>O + H) is dominant for oxidation. Shih and Hsu 519 [34] have shown that OH radicals are produced mainly by two reactions which 520 are  $H + O_2 \leftrightarrow O + OH$  and  $O + H_2O \leftrightarrow OH + OH$ . The equilibrium temper-521 ature calculated for the three compositions  $(Syn-1H_270N_2, Syn-5H_270N_2 and$ 522  $Syn-10H_270N_2$ ) is 1775, 1760 and 1741 K respectively. The data show that the 523 flame temperature does not vary significantly with increase in  $H_2$  % in fuel. For 524 these conditions, the rate constant of hydroxyl oxidation pathways of CO and 525  $H_2$  remains approximately same. Hence, the overall reaction rate is predom-526 inantly limited by amount of OH radical produced during the reaction. Shih 527 and Hsu [34] computational study shows that for very small  $H_2$  % in syngas 528 mixtures the overall reactivity of mixture is governed by CO hydroxyl oxidation 529 path (CO + H<sub>2</sub>  $\leftrightarrow$  CO<sub>2</sub> + H). With increase in H<sub>2</sub> % the overall reactivity of 530 mixture shifts towards  $H_2$  oxidation by reaction  $OH + H_2 \leftrightarrow H_2O + H$  ([34]). It 531 is possible that the OH radical concentration increases at much faster rate with 532 1 to 5 % increase  $H_2$  in comparison to 5 to 10 % of  $H_2$  in the fuel mixture. This 533 can be a possible reason to explain the non-linear increase in  $a_q$  with increase 534 in  $H_2$  %. Computations are needed to be performed to explore this further. 535



(b) (CO  $+H_2$ ) = 30 % by volume

Figure 11: Effect of H<sub>2</sub> percentage on experimentally obtained  $a_g$  using method 2 (M2) diluted with N<sub>2</sub>

The effect of volumetric H<sub>2</sub> % on  $a_g$  values is also studied using M1. Figure 12 shows the comparison of  $a_g$  and respective inlet fuel temperature at extinction for two H<sub>2</sub> (5 and 10 % by volume) fraction in fuel with 80 % N<sub>2</sub>. It was observed that the inlet fuel temperature remain more or less the same with change in H<sub>2</sub> percentage, but  $a_g$  increases by factor 2–2.5.

The next set of  $a_g$  data is generated for fuel having very high CO/H<sub>2</sub> ratio. This dataset is useful for validation of kinetic parameters for direct (CO + O  $\approx$  CO<sub>2</sub>, CO + O + M  $\approx$  CO<sub>2</sub> +M) and hydroxyl oxidation pathways (CO + OH  $\approx$  CO<sub>2</sub> +H) for CO oxidation for any given kinetic mechanism.



(a)  $(CO + H_2) = 20 \%$  by volume

Figure 12: Variation of experimental  $a_g$  data with D obtained using method 1 (M1) for syngas-air and methane/nitrogen-air combinations

Figures 13a and 13b show the comparison of experimentally obtained  $a_g$ using method 2 (M2) for high CO flames diluted with N<sub>2</sub> and CO<sub>2</sub>. The data show that, irrespective of CO<sub>2</sub> or N<sub>2</sub> used as diluent,  $a_g$  increases by factor of 2–2.5 with 10 % increase in CO. This ratio decreases to 1.5–1.7 with 10 % decrease in diluent for Syn-1H<sub>2</sub>60N<sub>2</sub>-air combination.



(a) High CO flames with  $CO_2$  as diluent



(b) High CO flames with  $N_2$  as diluent

Figure 13: Comparison of experimentally obtained  $a_g$  for High CO flames diluted with  $\mathrm{N}_2$  and  $\mathrm{CO}_2$ 

Experiments performed using 5 % H<sub>2</sub> by volume also show the same percentage increase in  $a_g$  values. Figures 14a and 14b show that,  $a_g$  values increases by

- $_{\tt 552}$   $\,$  factor of 2–2.5 times for CO\_2 and 2.6–3.8 times for N\_2 with 10 % increase in CO
- $_{553}$   $\,$  by volume in fuel mixtures containing 5 % H\_2. Higher CO % in the fuel gives
- <sup>554</sup> higher flame temperature which increases the overall fuel reactivity leading to
- 555 increase in  $a_g$  values.



(a) 5 % H<sub>2</sub> by volume with CO<sub>2</sub> as diluent



(b) 5 % H<sub>2</sub> by volume with N<sub>2</sub> as diluent

Figure 14: Comparison of experimentally obtained  $a_g$  for difference CO % diluted with  $\mathrm{N}_2$  and  $\mathrm{CO}_2$ 

The ratios of overall reaction rates ( $\omega_o$ ) calculated using Eqn. 2 for compositions Syn-1H<sub>2</sub>60N<sub>2</sub>/Syn-1H<sub>2</sub>70N<sub>2</sub> and Syn-1H<sub>2</sub>50N<sub>2</sub>/Syn-1H<sub>2</sub>60N<sub>2</sub> are 1.6 and <sup>558</sup> 1.3 respectively. For the same case, the ratio of experimentally obtained  $a_g$  val-<sup>559</sup> ues are 2.1–2.4 and 1.5–1.7 respectively. Hydroxyl radical (OH) concentration <sup>560</sup> in mixture depends on H<sub>2</sub> % in the fuel and flame temperature. The data shows <sup>561</sup> that, equilibrium temperature increases with increase in CO % in the fuel (1775, <sup>562</sup> 1973 and 2106 K for Syn-1H<sub>2</sub>70N<sub>2</sub>, Syn-1H<sub>2</sub>60N<sub>2</sub> and Syn-1H<sub>2</sub>50N<sub>2</sub>). Hence, it <sup>563</sup> is possible for a fixed H<sub>2</sub> % in fuel OH concentration reduces with in increase <sup>564</sup> in CO % leading to reduction in  $a_g$  ratio.

### <sup>565</sup> 4. Conclusion and future Work

In the present study, global extinction strain rate  $(a_q)$  is obtained for CH<sub>4</sub>/N<sub>2</sub>-566 air and syngas-air non-premixed flame using Tsuji-type counterflow configura-567 tion. The effect of porous burner diameter (D) on  $a_g$  value, that is,  $a_g$  decrease 568 with increase in D is determined. Plug flow boundary condition is experimen-569 tally verified by obtaining  $a_q$  at three distances of oxidizer inlet from the porous 570 burner. The flow-field blocking by porous burner has shown a deviation of less 571 than 3 % in  $a_q$  for a CH<sub>4</sub>-air combination. Hence, the blocking effect of burner 572 diameter (D) of dimensions less than 1/4 of combustion chamber side wall length 573 is negligible. The past literature shows the failure of assumed velocity profiles 574 at the nozzle exit as the possible reason for under-prediction of  $a_g$  for opposed 575 jet counterflow configuration. This issue can be resolved by using Tsuji type 576 configuration for extinction studies. Convective and radiative heat loss by the 577 flame to ambient can cause 4–9 % decrease in  $a_q$  values. So, for accurate  $a_q$ 578 predictions these losses should be incorporated in 2D planar simulations. Ni-579 trogen when used as a diluent, yields 1.6-2.25 times higher  $a_q$  in comparison 580 to  $CO_2$  used as diluent. Increasing  $H_2$  from 1 to 5 % leads to 2.5–3.8 times 581 increase in  $a_g$  compared to 1.3–1.7 times increase in  $a_g$  with 5 to 10 % increase 582 in  $\mathrm{H}_2$  for fuel mixture consisting of 70 %  $\mathrm{N}_2$  by volume. Increasing CO by 583 10 % leads to 1.5–2.4 times increase in  $a_g$  for fuel mixtures consisting of H<sub>2</sub> 584 (1 and 5 % by v/v), CO\_2 (50, 60 and 70 % by v/v) and N\_2 (50, 60, 70 and 585 80 % by v/v). The comparison of overall reactivity ( $\omega_o$ ) with  $a_g$  shows that, 586

 $a_g$  increase non-linearly ( $\approx \sqrt{w_o}$ ) with increase in H<sub>2</sub> % in fuel mixture. This increase in  $a_g$  with an increase H<sub>2</sub> % in fuel mixture can be explained from the OH radical concentration which requires 2D computations using simplified kinetic mechanism and will be taken up in future.

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## 595 Nomenclature

596	$\alpha$	CO $\%$ in fuel (v/v)
597	β	$H_2 \%$ in fuel (v/v)
598	$\omega_I$	Reaction rate 1 (mole/cm $^3$ s)
599	$\omega_I I$	Reaction rate 2 (mole/cm <sup>3</sup> s)
600	$\omega_{CO}$	Overall CO comsumption rate $(mole/cm^3s)$
601	$\omega_{H_2}$	$Overall \; H_2 \; comsumption \; rate \; (mole/cm^3 s)$
602	$\omega_o$	Overall fuel comsumption rate $(mole/cm^3s)$
603	$\phi$	Equivalance ratio
604	$a_g$	Global extinction strain rate $(s^{-1})$
605	D	Porous cylinder radius (m)
606	du/dx	axial velocity gradient $(s^{-1})$
607	h	Combustion chamber height (m)
608	$h^o_{f,CO_2}$	Heat of formation of $CO_2$ (KJ/mol-K)

- $h_{f,CO}^{o}$  Heat of formation of CO (KJ/mol-K)
- 610  $h_{f,H_2O}^o$  Heat of formation of H<sub>2</sub>O (KJ/mol-K)
- $_{611}$  *l* Combustion chamber length (m)
- 612  $u_{ox}$  free stream oxidizer velocity (m/s)
- $_{613}$  w Combustion chamber width (m)
- $a_l$  Local extinction strain rate (s<sup>-1</sup>)
- $_{615}$  P Pressure (N/m<sup>2</sup>)
- 616 P Temperature (K)
- $_{617}$  X<sub>CO</sub> Mole fraction of CO
- $_{618}$  X<sub>H<sub>2</sub></sub> Mole fraction of H<sub>2</sub>
- $_{619}$  X<sub>O<sub>2</sub></sub> Mole fraction of O<sub>2</sub>
- 620

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# Appendix A. Global extinction strain rate $(a_g)$ experimental data

$D \ (mm)$	10.6	15.6	22.8	30.8
Fuel	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$	$a_g \ (s^{-1})/T \ (K)$	$a_g~(s^{-1})/T~(K)$
M2				
$CH_4$	761±17/-	$614 \pm 9/-$	$467 \pm 10/-$	$380 \pm 7/-$
$\rm CH_4{-}30N_2$	$620\pm5/-$	$539 \pm 4/-$	$429 \pm 5/-$	$350 \pm 4/-$
$CH_4$ – $50N_2$	$515 \pm 6/-$	443±12/-	$349 \pm 5/-$	277±4/-
$\rm CH_4–70N_2$	277±5/-	$244{\pm}10/{-}$	211±8/-	$170 \pm 3/-$
M1				
$\rm CH_4–70N_2$	$436{\pm}5/557$	$351{\pm}2/540$	$280{\pm}3/524$	$239{\pm}2/490$

Table A.4: Global extinction strain  $(a_g)$  and burner temperature (T) data for CH<sub>4</sub>-N<sub>2</sub>-air combinations

$D \ (mm)$	10.6	15.6	22.8	30.8
Fuel	$a_g~(s^{-1})/T~(K)$	$a_g \ (s^{-1})/T \ (K)$	$a_g \ (s^{-1})/T \ (K)$	$a_g \ (s^{-1})/T \ (K)$
M2				
$\rm Syn{-}5H_280N_2$	$188{\pm}18/313$	$113{\pm}11/315$	$113 \pm 7/321$	$91{\pm}6/332$
$\rm Syn{-}10\rm H_{2}80\rm N_{2}$	$438{\pm}25/314$	$383{\pm}11/323$	$288 \pm 6/315$	$224 \pm 5/327$
$\rm Syn{-}1H_270N_2$	$179{\pm}14/326$	$139{\pm}5/323$	$125 \pm 9/324$	$108 \pm 8/323$
$\rm Syn{-}5H_270N_2$	$495{\pm}25/328$	$439 {\pm} 9/320$	$301{\pm}6/321$	$270{\pm}5/331$
$\rm Syn{-}10\rm H_270\rm N_2$	$819{\pm}34/323$	$578 \pm 12/332$	$420{\pm}12/317$	$354{\pm}5/336$
$\rm Syn{-}1\rm H_{2}60\rm N_{2}$	$395{\pm}10/321$	$312 \pm 4/327$	$269 \pm 5/324$	$262 \pm 7/333$
$\rm Syn{-}1\rm H_{2}50\rm N_{2}$	$592{\pm}13/333$	$483{\pm}14/337$	$368{\pm}9/320$	$351{\pm}10/339$
M1				
$Syn-5H_280N_2$	$255{\pm}13/421$	$216{\pm}7/404$	$168 {\pm} 2/423$	$141 \pm 8/396$
$\rm Syn{-}10H_280N_2$	$644{\pm}16/413$	$509{\pm}16/399$	$375 \pm 4/424$	-/-
$\rm Syn{-}1H_270N_2$	$304 {\pm} 9/467$	$270{\pm}7/469$	$251 \pm 5/484$	$175 \pm 3/454$
$Syn-5H_270N_2$	$666 \pm 6/451$	$588 \pm 3/471$	$421\pm8/481$	-/-

Table A.5: Global extinction strain  $(a_g)$  and burner temperature (T) data for Syn–H\_2N\_2-air combinations

Table A.6: Global extinction strain  $(a_g)$  and burner temperature (T) data for Syn–H<sub>2</sub>CO<sub>2</sub>-air combinations

D (mm)	10.6	15.6	22.8	30.8
Fuel	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$
M2				
$\rm Syn{-}5H_270CO_2$	$275{\pm}13/314$	$196{\pm}10/311$	$173 \pm 8/309$	$163 \pm 5/315$
$\rm Syn{-}10H_270CO_2$	$549{\pm}28/316$	$368{\pm}8/312$	$266{\pm}16/311$	$263{\pm}9/313$
$\rm Syn{-}1H_260CO_2$	$207{\pm}13/319$	$172{\pm}10/312$	$145{\pm}11/310$	$126 \pm 9/314$
$\rm Syn{-}5H_260CO_2$	$520{\pm}12/323$	$394{\pm}10/326$	$286{\pm}15/311$	$280{\pm}4/316$
$Syn-1H_250CO_2$	$392{\pm}25/324$	$341 \pm 9/315$	$295{\pm}7/311$	$261{\pm}9/313$

Table A.7: Global extinction strain  $(a_g)$  and burner temperature (T) data for Syn1–N<sub>2</sub>-air and Syn2–N<sub>2</sub>-air combinations

$D \ (mm)$	10.6	15.6	22.8	30.8
Fuel	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$
M2				
$Syn1-65N_2$	$604{\pm}24/318$	$434{\pm}4/313$	$295 \pm 9/313$	$280{\pm}3/309$
$Syn1-55N_2$	-/-	$654{\pm}10/327$	-/-	-/-
$Syn2-70N_2$	$629 \pm 21/313$	$447 \pm 8/312$	$301 \pm 5/312$	$276 \pm 5/308$
Syn2–60N2 $_2$	-/-	$660 \pm 9/332$	-/-	-/-

D (mm)	10.6	15.6	22.8	30.8
Distance $(L)$	$a_g \ (s^{-1})$	$a_g \ (s^{-1})$	$a_g \ (s^{-1})$	$a_g \ (s^{-1})$
M2				
$6 \mathrm{~cm}$	$761 \pm 11$	$607 \pm 4$	$457 \pm 9$	$386{\pm}10$
$10 \mathrm{~cm}$	$765\pm2$	$610\pm2$	$469 \pm 4$	$384 \pm 4$
$12~{\rm cm}$	$761{\pm}17$	$614 \pm 9$	$467 {\pm} 10$	$382\pm7$

Table A.8: Global extinction strain  $(a_g)$  data for CH<sub>4</sub>-air combination at three different distances

Table A.9: Global extinction strain  $(a_g)$  data for CH<sub>4</sub>-air combination for wall effects

$D \ (\mathrm{mm})$	10.6	15.6	22.8	30.8
	$a_g \ (s^{-1})$	$a_g \ (s^{-1})$	$a_g \ (s^{-1})$	$a_g \ (s^{-1})$
M2				
with side wall	$761 \pm 17$	$614\pm9$	$467{\pm}10$	$382\pm7$
without side wall	$768{\pm}11$	$620\pm7$	$478 {\pm} 14$	$387 \pm 3$

Table A.10: Effect of insulation on global extinction strain  $(a_g)$  data for Syn-10H<sub>2</sub>80N<sub>2</sub>-air combination

D (mm)	10.6	15.6	22.8	30.8
	$a_g \ (s^{-1})/T \ (K)$	$a_g~(s^{-1})/T~(K)$	$a_g \ (s^{-1})/T \ (K)$	$a_g \ (s^{-1})/T \ (K)$
M2				
without insulation	$438 {\pm} 25/317$	$383{\pm}11/323$	$288 {\pm} 6/315$	$224 \pm 5/327$
with insulation	$474 \pm 31/313$	$399{\pm}10/323$	$308{\pm}6/313$	$247 \pm 8/318$

Table A.11: Effect of fuel flow rate on global extinction strain  $(a_g)$  data for Syn–5H<sub>2</sub>80N<sub>2</sub>-air combination using D = 22.8 mm

fuel flow	6 Lpm	8 Lpm	10 Lpm	12 Lpm	15 Lpm
Fuel	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$	$a_g~(s^{-1})/T~(K)$
M2					
$\rm Syn{-}5H_280N_2$	$74{\pm}4/312$	$91{\pm}4/315$	$96{\pm}2/318$	$114 \pm 8/321$	$116{\pm}6/318$