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spectroscopy

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Lean partially premixed turbulent flame equivalence ratio measurements using 1 2 3 Laser-induced breakdown spectroscopy

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14 15 16 17 Abstract

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18 The creation of a more stable flame along with the extension of flammability limits under lean mixture 19 combustion was the main motivation to develop a new burner design, which has been investigated in this 20 research. The current burner configuration was utilized to create a wide range of higher turbulent 21 intensities and to produce different degrees of mixture inhomogeneity, which acted to promote minimum 22 pollution, highest performance and higher flame stability. The burner stability assessment was investigated 23 using two types of fuel: natural gas (NG) and liquefied petroleum gas (LPG). They were tested under 24 different degrees of partial premixing, and two turbulence generator disks for lean mixture at an 25 equivalence ratio of $\phi = 0.8$ were used. Following this, the Laser Induced Breakdown Spectroscopy (LIBS) 26 technique was utilized to characterize and quantify the impact of changing the disk slit diameter on the 27 distributions profiles of equivalence ratio or mixture fraction for a NG/air partially premixed flame. A series 28 of homogeneous NG/air mixtures with different equivalence ratios were used to obtain the correlations 29 between the measured emission lines of LIBS spectra and the global flame equivalence ratio. Consequently, 30 the emission spectral lines ratios of H/N, H/O and C/N+O were utilized to predict the equivalence ratio distributions. The results demonstrated that for all of the mixing lengths, NG/air mixture with larger disk 31 32 generator diameter yielded the maximum burner stability, whilst the LPG/air mixture with a larger disk 33 generator diameter resulted in the minimum burner stability. Furthermore, the flame associated with the 34 larger disk slit diameter had a uniform local equivalence ratio distribution and lower RMS fluctuation 35 profiles of equivalence ratio in comparison to the lower disk slit diameter.

Keywords: Stability; laser breakdown; partially premixed; Turbulence; LIBS; Mixture fraction

38 39 **Highlights:**

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- Fuel type and partially premixed level effects on burner stability
- Flame stability behaviour of different turbulence generator disks were investigated
- Impact of changing the disk slit diameter on equivalence ratio distributions was examined

Nomenclature

| LIBS | laser induced breakdown spectroscopy | Y _{fi} | Mass fractions of the fuel elements | |
|----------|--------------------------------------|-----------------|--------------------------------------|--|
| ds | Turbulence generator diameter | L_k | Kolmogorov scale | |
| ξ | Mixture fraction | HI | Hydrogen intensity | |
| NG | Natural gas | λ | Laser wavelength | |
| PPFs | Partially Premixed Flames | Vj | Jet velocity | |
| ICCD | Intensified Charge-coupled device | $	au_{mix}$ | Mixing time | |
| L | Mixing length, mm | φ | Equivalence ratio | |
| b | Slit thickness | dc | Cone diameter | |
| LTE | Local Thermal Equilibrium | Х | Axial distance above the burner tip | |
| C_3H_8 | Propane | C_4H_{10} | Butane | |
| Rms | Root mean square | δ | Convection-diffusion laminar flame | |
| ΡI | Princeton Instruments | | thickness | |
| NI | Nitrogen intensity | OI | Oxygen intensity | |
| CI | Carbon intensity | θ | Cone half angle | |
| D | Inner diameter of the outer tube, mm | d | Inner diameter of the inner tube, mm | |
| Do | Outer diameter of the outer tube, mm | do | Outer diameter of the inner tube, mm | |
| r | Radial distance, mm | Х | Axial distance, mm | |
| R | Inner radius of the outer tube, mm | | | |

45 *Introduction*46

47 The mixture stratification process and the generation of higher turbulence levels are the two main 48 established techniques to extend the lean combustion limit [1]. Therefore, the new combustion system 49 design, directed towards partially premixing combustion, is a promising technology, which offers the 50 potential to meet ever stringent emission regulations, as well as improving the system efficiency [2]. The 51 performance of the combustion processes is mainly linked to the local equivalence ratio distributions near 52 the ignition event, and based on these distributions, the combustion characteristics and the level of 53 emissions such as CO, NOx, etc. will be strongly affected [3, 4]. This influence is mainly linked to the change 54 of both the local properties of the reaction zone and the global behaviour of the combustion system 55 associated with the flame propagation within spatial variations of the equivalence ratio [5]. Therefore, 56 quantifications of this ratio are essential to sustain the higher stability of the combustion process and to 57 minimize the soot emissions [6].

59 Richardson et al. [7] demonstrated that the laminar flame propagation speed was affected by the 60 equivalence ratio gradients, due to the gradients effects on the molecular transport of hot products and 61 radical species into the reaction zone. Furthermore, Dold [8] concluded that as the mixture fraction or 62 equivalence ratio gradient increased, the flame propagation speed was reduced. This was attributed to the lower conduction heat transfer associated with the increased flame front curvature, which consequently 63 64 reduced the preheating process of the unburned mixture. Likewise, Richardson and Chen [4] investigated 65 turbulent flame propagation under the impacts of equivalence ratio-stratification for methane air flame 66 using DNS analysis. They concluded that the stratification process influences significantly on the flame 67 surface area due to the variation caused by equivalence ratio gradient orientation on the flame surface 68 69 averaged consumption speed with surface averaged equivalence ratio.

70 In order to measure and obtain comprehensive information regarding global and local equivalence ratios 71 distributions in turbulent flames, laser induced break down spectroscopy (LIBS) was used in this research. 72 The principle of laser induced breakdown spectroscopy depends mainly on the interaction of a very short-73 duration focused pulsed laser beam onto the surface of the substance to be analysed, causing the 74 breakdown of the sample's chemical bonds, followed by the formation of plasma, which is composed of 75 ionized matter [9, 10]. During the subsequent relaxation of the constituent excited species, the spectral 76 emission occurred, and it was collected and spectrographically analysed using an Intensified CCD (ICCD) 77 detector attached to a spectrograph detector [11]. The elemental composition of any material can be 78 identified based on their fingerprint spectral lines, and consequently, the concentration of such elements 79 will be quantified from the spectral line intensities. The main advantages of the LIBS technique lies in its 80 ability to rapidly analyse samples remotely and in situ, with minimal sample preparation [12, 13]. 81 Furthermore, LIBS has the potential of simultaneous multi-elemental analysis with minimum equipment 82 [14]. The aforementioned advantages make the LIBS technique to an attractive tool for the majority of 83 applications including liquids [15, 16], solids [17, 18] and quantitiative analysis of gases and gas mixtures, 84 85 which are all essential tasks in the field of security, environmental and chemical enalysis [19, 20].

86 Over the past few years, the LIBS technique has been applied extensively to the field of combustion 87 diagnostics [21, 22] for equivalence ratio measurements which can be obtained based on the atomic species 88 concentrations in flames. Kotzagianni et al. [23] established a new calibration scheme for equivalence ratio 89 measurements of non-premixed and premixed methane turbulent flames using the LIBS technique. They 90 concluded that for a lower mole fraction of methane in the range 0-0.3, the ratio of H α (656.3 nm) over 0 91 (777.3 nm), (H α /O), should be utilized for equivalence ratio calculations. Additionally, they found that, for 92 a higher mole fraction of methane in the range 0.3-1, the ratio of C_2 over CN, (C_2/CN), should be used to 93 identify the equivalence ratio. The majority of past LIBS studies were mainly focused on using the ratios of 94 some spectral lines of the atomic origin, such as the carbon line to nitrogen [C (833 nm)/N (744 nm)], the 95 carbon line to oxygen [C (833 nm)/O (777 nm)] [24], or the ratio of the intensity of a carbon line to the sum of the intensities of a nitrogen and an oxygen line, [C (711 nm)]/[N (744 nm) + 0 (777nm)] [25]. These 96 97 ratios have been successfully utilized to delineate the relationship between the spectral intensity and the 98 mixture equivalence ratio. The most commonly used ratios were between the hydrogen line to an atomic 99 emission of oxygen line [H α (656.3 nm)/O (777nm)] or hydrogen line to an atomic emission of nitrogen 100 $[H\alpha (656.3 \text{ nm})/N (746.8 \text{nm})]$ [26-28]. Alongside the equivalence ratio measurements, LIBS has been 101 utilized for further analysis of the turbulent flame characteristics, including the measurements of $\frac{102}{103}$ temperature [28], gas density and concentration [29].

104 The present work discusses the development of a new burner design, which has the ability to operate over 105 lean combustion conditions with higher flame stability, and consequently, it provides lower levels of 106 exhaust emissions. This burner generates different stratification degrees of the mixture by changing the 107 degree of the partially premixed level or by using different turbulent generator disks. Considerable attention has been paid to obtain comprehensive information regarding the impact of changing the disk slit 108 109 diameter, fuel type and the level of partially premixed on the flame stability maps. The stability map results 110 were examined and utilized to identify the suitable mixture conditions for equivalence ratios distributions 111 using the LIBS technique. Likewise, the ability to conduct quantitative equivalence ratio measurements of 112 partially premixed NG/air mixtures using the LIBS technique was examined. Furthermore, the correlation 113 of values of emission intensity ratio [C/(N+O), H/O] and H/N] using LIBS against NG/air mixtures $\frac{114}{115}$ equivalence ratios was established.

116 **2.1. Burner setup details**

118 The new design of the current burner configuration was comprised of a pair of fixed concentric stainless-119 steel tubes; the inner tube carried the air whilst the outer tube carried the fuel, as shown in Figure 1.



120 121 Figure.1. Burner schematic diagram (a) half section 3D sketch, (b) half section 2D sketch

This new burner has the strength to generate higher levels of turbulence intensities as well as the ability to 122 123 provide different degrees of partial premixing. The inner tube has a wall thickness of 1.5 mm and an inner 124 diameter of d = 19 mm, whilst the outer tube has a wall thickness of 1.5 mm and an inner diameter of D =24 mm. The outlet of the inner tube was located at a distance L below the tip of the outer tube. The mixing 125 process between air and fuel occurred within this distance L, and consequently, by changing this distance, 126 127 the degree of inhomogeneity was varied. The degree of partial premixing was defined by the ratio L/D and 128 five sets of L/D = 1, 2, 3, 4 and 7 were selected to investigate the burner stability as shown in Figure 2 (a). 129 Both the air and fuel streams were fed into the burner tangentially and directed towards the inner and 130 outer tubes, respectively. The fuel stream passed through the annulus gap between the vertical concentric 131 tubes and then it mixed with the air through the mixing distance, L, as shown in Figure 1. The LPG used 132 during the present study was formed of 50% butane and 50% propane (molar basis). While the NG was formed from of 95% methane (molar basis). The flow rates of both air and fuel streams were controlled 133 134 precisely using an Alicat MCS-series meter which was calibrated to a certain range of 150 l/min with a high accuracy of ± (0.4% of Reading and + 0.2% of Full Scale). Additionally, it had a 10-millisecond response 135

time. Both the fuel and air streams were introduced to the burner at ambient conditions. The overall mean equivalence ratio was determined based on the fuel and air stream mass flow rates.

139 *2.2. Turbulence generator disks*

141 The turbulence generator used for the current study consisted of a thin disk turbulence generator with a 142 circular slot, in order to boost the turbulence intensity levels, similar to the disk developed by Videto and 143 Santavicca [30]. Beyond this disk, a contracting nozzle was utilized to assist the collapse of the air stream, 144 which passed through the circular slot, into a wide range of turbulent fluctuations and integral length scales, 145 and consequently, this promoted higher levels of turbulence intensities [31]. Recently, several studies have 146 been conducted to develop new turbulence generator designs in order to increase the turbulent flame 147 speed and to achieve improved flame stability [32]. In this paper, two disks with a circular slit diameter of 148 ds= 25 and 45 mm have been selected, as shown in Figure 2 (b). Both the disks were designed with a 149 constant slit thickness (b) of 0.8 mm, whilst the following converging nozzle consisted of a cone angle (2θ) 150 of 76°, 26 mm height (h) and 62 mm base cone diameter (dc). At a partial premixing level of (L/D = 2), a 151 particle image velocimetry (PIV) system was utilized to study the velocity profiles of the developed burner. 152 Accordingly, the flow turbulent intensities were calculated as a ratio between the fluctuation of the jet velocity to the mean jet velocity (V_{rms}/V_j) [33]. They demonstrated that the current burner turbulent 153 154 intensity could reach up to 36%, depending on the jet velocity, and any increase in the jet velocity or the 155 disk slit diameter will increase the turbulent intensity [33].





156 157 **2.3. LIBS set-up** 158

159 The schematic diagram of the experimental apparatus used for the LIBS-technique is presented in Figure 160 3. The plasma was generated by using a pulsed Nd: YAG laser with a frequency of 10 Hz. This laser beam was characterised as the first harmonic wavelength of 1064 nm, pulse width of 6 ns and a beam diameter 161 162 of 10 mm. Single shot operations of the laser were used for the current investigation, with a constant laser 163 energy of 100 mJ, whilst the focusing process of the laser beam was carried out using a 50 mm focal length plano-convex lens to create the plasma. This plasma constitutes of excited atoms in the ionized gas, and 164 165 during the subsequent decay of the excited electrons, a rainbow of light of different wavelengths is released, 166 resulting in characteristic spectral emissions. In order to optimize the spectral emission data collections, a 167 pair of 100 mm focal-length plano-convex lenses were placed at an angle of 26° to the laser beam [9], and 168 then the spectral emissions were directed towards a fused-silica optical fiber with an aperture of 200 µm. 169 Consequently, the spatial resolution of the system based on this aperture diameter was small enough for turbulent flame measurements and it was approximately 1 mm, estimated from the size of the plasma. 170 171 Consequently, the optical fiber captured the emitted light and it carried it to an Echelle spectrometer (PI-Echelle, Princeton Instruments: IMAX-512, USA) attached to a gated Intensified Charge-Coupled Device 172 173 (ICCD) covering a wavelength range of 190–1100 nm, to resolve and image the signal spectrally. The data 174 acquisition system was utilized to precisely control both the gate delay time and the gate width of the 175 spectroscopic data acquisition system. In order to achieve high measurement accuracy, the laser induced 176 plasma generated should reach the Local Thermodynamic Equilibrium (LTE) state, and for the current 177 investigation, the LTE was established at roughly 1 µs following the plasma initiation.



DAQ and control computer



180 A Stanford research model DG535 4-channel delay pulse generator was employed to precisely control the 181 Shutter timing of the ICCD camera and the laser-trigger signals. A precise control of the timing parameter 182 and delay time was carried out by combining the fast oscilloscope (500MHz) with the fast photodiode (rise 183 time = 1 ns). The spectral emission lines were analysed and identified using commercial software 184 (GRAMS/AIv.8.0, Thermo-electron. Co.). The raw emission signals collected by the LIBS system were 185 corrected by subtracting them from the emission background by using the LIBS software (Winspexs 21). 186 This correction was applied to eliminate the inherently dark signal associated with the spectrometer 187 detector, which itself is associated with the output LIBS signal. Both the delay time and the gate width were 188 varied until the optimized intensity of the spectral emission line was obtained. In order to achieve that 189 optimized signal, the ICCD gate width was set at 10,000 ns, whilst the spectral emission line intensities 190 were collected at a delay time of 600 ns after the laser irradiation. For the present work, one case was 191 selected for equivalence ratio measurements using the LIBS technique. The NG/air mixture employed for 192 this case study was characterized by an equivalence ratio of $\varphi = 0.8$, mass flow rate of 2.91 kg/h and jet velocity of $V_i = 1.6$ m/s. A long distance pipe, with an approximate length of 200 times the pipe diameter 193 194 was selected for the fuel-air mixing process, to create a perfectly homogeneous mixture valid for precise 195 calibration with the LIBS technique. Consequently, the equivalence ratio measurements were carried out 196 using the LIBS technique after the calibration process was completed.

198 **3. Results and discussion**

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- 199 **3.1. Stability limit characteristics**
- 200 3.1.1 Stability limit characteristics for each fuel and each disk

201 The degree of mixture stratification at the exit of the outer tube of the current burner is affected by changing 202 the mixing length to diameter ratio (L/D) and by varying the turbulence generator disk slit diameter. 203 Consequently, these equivalence ratio fluctuations will influence the stability, dynamics and the structure 204 of the flame [34]. Therefore, the flame stability behaviour can be characterized, with particular respect to 205 the blowout characteristics to delineate the extinction regions between different Reynolds numbers and 206 either the degree of partial premixing (L/D) or the equivalence ratio, φ . The blowout limit is identified by 207 the bulk jet velocity, V_i at which a complete flame extinction is occurs. This extinction is carried out by 208 increasing the air flow rate while keeping the fuel flow rate fixed, whereas, the blowout limits are obtained 209 210 only from visual inspection of the flames [35].

211 In the present work, the assessment of the burner stability was implemented at lean conditions of $\varphi = 0.8$ 212 for five sets of partial premixing ratios (L/D = 1, 2, 3, 4 and 7) as shown in Figure 4 (a). The investigation of 213 the stability map was accomplished for two types of fuels, NG and LPG, using two turbulence generator 214 disks with slit diameters of ds = 25 and 45 mm. At L/D =1, the flame was considered nearly non-premixed, 215 while at L/D =7, the flame was considered almost fully premixed. Furthermore, qualitative measurements 216 of flame characteristics such as flame shape, height and colour associated with each case have been 217 observed as shown in Figure 4 (b). Five rows of images, which represent a comparison between LPG, and 218 NG, for each disk slit diameter, at different degrees of partial premixing, are presented. It was noticed from 219 Figure 4 (a), that changing the degree of partial premixing or changing the turbulent generator disk slit 220 diameter has a significant effect on the flame stability for both fuels. At L/D =1, the NG/air mixture for both 221 the disk slit diameters exhibited the maximum flame stability, whilst the LPG/air mixture with ds = 45 mm 222 demonstrated the minimum flame stability. These results are further explained with the flame images, 223 where NG demonstrated a stable flame with a blue colour, whilst LPG showed a significant small blue region 224 near the flame base followed by a bright yellow flame with high luminosity. The LPG flames look more like 225 diffusion flames, and consequently, the longer flame height associated with LPG flames can be linked to the 226 flame elongating, to obtain the oxygen available in the ambient air. The blue flame near the base is due to 227 the enhanced local partial premixing. The lower stability of LPG/air mixture in comparison to the NG/air 228 mixture could be attributed to the different properties of the fuels, in particular the Lewis number. As it is 229 well known, both the local stretch rate and the physicochemical parameters including the Lewis number 230 and the laminar burning speed have a significant influence on the turbulent burning speed [36]. Clarke [37] 231 calculated the Lewis number against the equivalence ratio for a range of different fuel-air mixtures that are 232 in common use. For lean mixture conditions, the Lewis number for NG (Le = $0.94 @ \phi = 0.8$) was lower 233 compared to that of propane (Le = 1.75 @ ϕ = 0.8) and butane (Le = 1.8 @ ϕ = 0.8) fuels; the main parts of 234 LPG fuel. Consequently, due to the proximity of the NG Lewis number to unity, the turbulent burning 235 velocity of NG flames was enhanced, and hence, the stability of the turbulent NG flames in comparison to 236 LPG flames was improved [38-40]. Furthermore, the laminar flame speed of the NG/air mixture at lean 237 equivalence ratio of $\phi = 0.8$ was slightly higher compared to that of the LPG/air mixture [41, 42]. Therefore, the NG flames were more stable compared to the LPG flames. 238

240 In addition, for L/D = 1, both the fuels with disk, ds = 45 mm yielded the minimum flame stability compared 241 to the other L/D ratios. This behaviour could be linked to the lower mixing time (τ_{mix}) generated from the 242 higher turbulence level associated with the large slit diameter [43]. The numerical analysis of the current 243 burner using three-dimensional computational fluid dynamics (CFD) modelling quantified the higher axial 244 and radial turbulent intensities accompanied with disk, ds = 45 mm, compared to that of disk, ds = 25 mm 245 [42]. Consequently, improper air/fuel mixing will be generated during this shorter mixing length, L, due to 246 the insufficient time available for the mixing process. Subsequently, an incomplete partial premixed flame 247 will be produced at the burner tube exit [44], in comparison to the other L/D ratios, which contained longer 248 mixing lengths. Furthermore, when the mixing time of the fuel and air during the burner mixing length 249 becomes shorter than the chemical reaction time ($\tau_c > \tau_{mix}$), the combustion can no longer be sustained, and 250 this will reduce the flame stability. In addition, for the high levels of turbulence intensities, the flame will 251 be quenched quickly, due to the higher mixing rate (scalar dissipation rate), characterized by the small 252 length scale of the turbulence, in comparison to the smaller thickness (δ) of the laminar flame reaction-253 diffusion zone [45, 46].

∠53 254

As the degree of partial premixing was increased to L/D = 2 and 3, the flame stability was increased for all of the cases, except the case of the NG/air mixture with ds = 25 mm, where the flame stability was gradually reduced. For both partially premixing ratios of L/D = 2 and 3, the LPG/air mixture with ds = 45 mm consistently had the minimum flame stability. The maximum flame stability was achieved at L/D = 3 for LPG with disk, ds = 25 mm and NG with disk, ds = 45 mm. This could be attributed to the multi-reaction zones structure associated with a non-homogeneous mixture, which include lean, rich and diffusion [47]. Once these zones interacted, the likelihood of the triple flame structure formation increased, resulting in higher flame stability [48, 49]. Regarding the flame images, all of the cases exhibited stable flames with a blue colour, except LPG, ds = 45 mm, which yielded a lower stability with a small blue region near the flame hase followed by bright yellow flame with high luminosity

- 264 base followed by bright yellow flame with high luminosity.
- 265



Figure.4. Impact of changing the disk slit diameter, fuel type and the level of partial premixed on (a) stability curves and (b) flame appearance of partially premixed flames.

However, with further increase of the degree of partial premixing to L/D = 4, the flame stability started to decrease for the NG/air mixture with ds = 45 mm, and the LPG/air mixture with ds = 25 mm, whilst the flame stability started to increase for the NG/air mixture with ds = 25 mm, and the LPG/air mixture with ds = 45 mm. The minimum flame stability for the NG/air mixture was produced with ds = 25 mm. In the most well-mixed case of partial premixed degree L/D = 7, it was noticed that the flame stability was reduced for all of the cases.

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Figure 4 (a) also demonstrated the evolution of the jet velocity (where the blowout occurred) as a function 273 274 of the level of the partially premixed L/D. For disk, ds = 25 mm, the maximum jet velocity of V_j =1.8 m/s 275 was observed for NG at L/D = 1, whilst for LPG, the maximum jet velocity of $V_j \approx 2.6$ m/s was observed at 276 L/D = 3. For disk, ds = 45 mm, the maximum jet velocity of $V_i = 2.6$ m/s was observed for NG at L/D = 3277 whilst for LPG, the maximum jet velocity of $V_i \approx 1.8$ m/s was observed at L/D = 4. This enhancement in 278 flame stability for these non-homogenous mixtures is linked to the higher probability of finding near-279 stoichiometric mixtures of fuel and air burning in contact with the hot burnt gases from the burner. This 280 results in additional heat release at the base of the turbulent jet flame and helps to stabilize it.

281 282 The flame stability at a constant level of partially premixed of L/D = 2 was further examined under different 283 Reynolds numbers and equivalence ratios, as presented in Figure 5 (a). Furthermore, qualitative 284 measurements of flame characteristics, such as flame shape, height and colour, associated with each case, 285 have been observed, as shown in Figure 5 (b). Four rows of images, which represent a comparison between 286 LPG and NG, for two disk slit diameters of ds = 25 mm and ds = 45 mm, at different equivalence ratios, are 287 displayed. The blowout limit was determined by maintaining a constant mass flow rate of the fuel stream 288 whilst increasing the mass flow rate of the air stream until the flame extinction happened. Four extinction 289 curves were established for each disk slit diameter and each fuel type. For the NG/air mixtures, changing 290 the turbulence generator disk slit diameter had a small effect on the flame stability, especially at very lean 291 conditions, where their behaviours were closely matched. On contrast, the flame stability of the LPG/air 292 mixture was more susceptible to the change of the turbulence generator disk slit diameter. For lean mixture 293 conditions of φ < 0.8, the LPG/air mixture with ds = 25 mm yielded the maximum flame stability followed 294 by the NG/air mixtures with ds = 45 mm and 25 mm, respectively. The flame stability completely 295 deteriorated for the LPG/air mixture with ds = 45mm, over the whole equivalence ratio range. The higher 296 stability of LPG at lean conditions could be contributed to its higher calorific value (energy content) than 297 natural gas, with 93.2 MJ/m³ vs 38.7 MJ/m³ [50], and thus, its higher heat release rate. The primary effect 298 of an increase in the heat released by the flame is to increase the flame speed, and consequently, a small 299 amount of LPG will produce a higher energy flame as compared to an equivalent amount of NG [51, 52]. A 300 slightly higher flame stability was observed for the NG/air mixture, for the disk slit diameter of ds = 25 mm, 301 as compared to disk slit diameter, ds = 45 mm, for lean conditions ($\phi < 0.6$). This was due to the high 302 susceptibility of the combustion systems operating at lean conditions to the high turbulence accompanied 303 with ds = 45 mm, which promoted lower flame stability induced by local flame extinction [53]. 304 Consequently, this resulted in lower thermal diffusivity compared to the mass diffusivity and hence the rate 305 of heat transfer could not keep pace with the rate of mass transfer, resulting in the flame becoming 306 quenched. 307

For equivalence ratios of $\varphi \ge 0.8$, the NG/air mixture with ds = 45 mm demonstrated the maximum flame stability followed by the LPG/air mixture with ds = 25 mm, and the NG/air mixture with ds = 25 mm. A significant difference in the flame stability for the NG/air mixture with ds = 45 mm was noticed at the equivalence ratios of $\varphi = 0.8$ and 0.9, as compared to that observed with ds = 25 mm. The high turbulence level associated with the large disk ds = 45 mm, resulted in an increase in the entrainment of the hot product gas and flame radicals towards incoming reactants, elevating the chemical reaction rate and hence the stabilization of the flame [54, 55]. LPG with ds = 45 mm, consistently had the minimum burner stability.

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Regarding the flame appearance, it was noticed for the NG/air mixtures, that the flame is more stable for both the disks, with a blue colour. In contrast, LPG with ds = 45 mm, yielded a completely diffusive flame with a bright yellow colour, without blue coloured flame near its base at very lean mixtures. As the equivalence ratio increased to rich conditions, the flame colour became blue. Likewise, the flame images reflected the higher stability of the flame associated with LPG, ds = 25 mm, for lean conditions, with a longer blue coloured flame near to the base of the flame with a small yellow coloured portion at the top of the flame. Additionally, for stoichiometric and rich conditions, the flame became more stable, with a blue colour. 323



Figure.5. Impact of changing the disk slit diameter and fuel type on (a) stability curves (b) flame appearance of partially premixed flames at constant level of partial premixing, at L/D = 2.

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327 3.2. LIBS spectra lines and calibration Lines

328 3.2.1. LIBS spectra

329

For turbulence disk slit diameter of ds = 25mm, the break down LIBS spectrum, in the region between 240 and 900 nm was measured for the lean NG/air mixture of φ = 0.8 and jet velocity of V_j = 1.6 m/s, as shown in Figure 6. The incident laser energy was maintained constant at 100 mJ for all measurements, and the presented spectrum corresponded to an average of 50 laser shots. A broad band Echelle spectrometer was employed to collect the emission signal and the most common spectral atomic lines highlighted in the spectrum were C (I)-247.8 nm, H α (I) - 656.4 nm, N (I)-747.2 nm and O (I)- 777.5 nm. Moreover, the N(I) atomic line consisted of three peaks at 742.5, 744.5, and 746.8 nm.

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338 339

Figure.6. NG/air mixture typical break down LIBS spectrum in the region between 240 and 900 nm at an equivalence ratio of $\varphi = 0.8$.

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343 The quantitative analysis of the local equivalence ratio measurements by the LIBS technique could be 344 fulfilled by establishing a linear relationship between the value of the emission lines intensities and the 345 equivalence ratio of a well premixed mixture. To ensure confidence regarding the high mixture 346 homogeneity, a long-distance pipe with an approximate length of 200 times the pipe diameter was selected 347 for the fuel-air mixing process, to create a perfectly homogeneous mixture. For the calibration purpose, a 348 wide range of equivalence ratios, including eight sets of $\varphi = 0, 0.2, 0.4, 0.6, 0.6, 0.8, 1$ and 1.4 were selected 349 to evaluate the emission spectral line intensities of O (I), H α (I), C (I), and N (I), for each set. The statistical 350 analysis of the emission spectrum was conducted by using the GRAMS/AI V.8.0 Spectroscopy software package. The predominant atomic lines of C (I)-247.8 nm, H_α (I) - 656.4 nm, N (I)-747.2 nm and O (I) - 777.5 351 nm for each equivalence ratio are presented in Figure 7. Based on the 50 single shot measurements carried 352 353 out at each set, the calculated standard deviation of the predominant spectral line emission intensities was 354 altered between 5-12%. The spectrum lines were captured with a 0.02 nm spectral resolution by using an 355 ICCD camera attached to an Echelle spectrometer. Due to the air compositions which include 78.09% 356 nitrogen, 20.95% oxygen, 0.93% argon, and 0.04% carbon dioxide, the carbon emission spectral line C (I) was noticed at an equivalence ratio of $\varphi = 0$. The increase of the equivalence ratio was associated with an 357 358 increase in the intensity of C and H and a decrease in the intensity of O and N.



361 Figure.7. The LIBS spectra of C (I), H (I), N (I) and O (I) elements for a series of different equivalence ratios

³⁶² at $\varphi = 0, 0.2, 0.4, 0.6, 0.8, 1, 1.2$ and 1.4 for the NG/air mixture in the region between (a) 650-880 nm, and 363 (b) 240-500 nm.

364 3.2.2. Calibration lines

365 From the spectrum analysis implemented in the aforementioned section, the elemental mass fractions Y_c, Y_H, Y_N and Y₀, were calculated and presented against the signal intensity as a linear relationship in Figure 366 8. Due to the high precision of using the integral method of the area under each spectral line [25, 56], the 367 368 mass fraction of each emission line was defined by the ratio between the integration area under the peak 369 of this line and the total integration area under the peaks of all of the elemental spectral lines. The 370 normalized signal intensity was obtained by dividing the elemental signal intensity by the maximum 371 intensity of each element. The high linearity between the LIBS signal intensity and the elemental mass fraction shows the high fidelity of the LIBS system for use as a measuring tool for equivalence ratio 372 quantitative measurements. The deviation of carbon and hydrogen elements from linearity at lower 373 374 concentrations could be contributed to the laser shot-to-shot fluctuation.





Figure.8. The elemental mass fractions (a) Y_C and Y_H, and (b) Y_N and Y_O, against the normalized signal
 intensity.

378



379 380

Figure.9. Correlation between the elemental intensity ratio of (a) H/N, (b) H/O and (c) C/N+O, and the equivalence ratio for the premixed NG/air mixture.



The C/(N+O), H/O and H/N intensity ratios were reported as a function of the equivalence ratio for the
 NG/air mixtures, as shown in Figure 9. The value of each intensity ratio was normalized to the maximum
 intensity associated with this ratio. Moreover, the three ratios corresponded linearly to the equivalence

ratio of the premixed NG/air mixture. This calibration line will be essential for local equivalence ratio
 measurements. The calibration procedure was accomplished under real temperature and pressure
 conditions, which is directly related to the behaviour of the flames.

Two methods were utilized to evaluate the mixture fraction (ξ); in the first method, ξ was calculated by

- using the intensity ratios of C/(N+O), H/N and H/O, whilst the second method identified ξ based on the
- 392 obtained elemental mass fraction values, which are presented in Figure 8, and substituted it into Eq. (1).
- 393 Subsequently, a relationship between the two methods is presented in Figure 10. Over the entire range of
- 394 the mixture fraction measurements, the data showed good agreement, especially with the values
- determined from the ratio C/(N+O). The intensity ratios of $H_{656.3}/O_{777.4}$ and $H_{656.3}/N_{746.5}$ yielded good
- agreement with the ξ calculated from Eq. (2), at mixture fractions $\xi = 0.03-0.05$ and for $\xi = 0.07-0.09$.
- **397** Consequently, both methods could be employed for mixture fraction quantitative measurements.

$$\xi = 1 - \sum y_{oi}$$

where y_{oi} are the ovidant elements' mass fractions. Thus, ξ for the fuel stream will be 1, and for the ovidizer

398 where y_{0i} are the oxidant elements' mass fractions. Thus, ξ for the fuel stream will be 1, and for the oxidizer 399 stream, it will be 0.



Figure.10. Comparison of the mixture fraction calculation for a partially premixed flame at equivalence ratios of 0.8 and jet velocity of 1.6 m/s, using the three ratios of (C/(N+O)), (H/O) and (H/N), represented by the y-axis, and that calculated from Eq. (2) based on the elemental mass fractions represented by the xaxis.

424 3.3. Mass Fraction Measurements

425

426 The flame appearance and the location of the measurement axial positions (red dots) of the lean partially

- 427 premixed flame at the equivalence ratio, φ = 0.8, with two disk slit diameters, for the NG/air mixture, is
- 428 shown in Figure 11.



429

Figure.11. The flame appearance and the location of the measurement axial positions (red dots) of the lean partially premixed flame at the equivalence ratio, $\varphi = 0.8$ and $V_j = 1.6$ m/s, with two disk slit diameters, for the NG/air mixture.

433

434 Figure 12 shows the mass fraction radial distribution of different elements including O, N, H and C with the 435 two disk slit diameters of ds = 25 mm and ds = 45 mm, at further distances from the burner rim. Around 436 the burner axis, a perfectly symmetrical distribution of the mass fraction occurred. A perfectly flat shaped 437 distribution occurred with the large slit diameter of ds = 45 mm, whilst a triangular shaped distribution 438 occurred with a lower slit diameter of ds = 25 mm. In comparison to the lower disk slit diameter of ds = 25439 mm, the larger disk slit diameter of ds = 45 mm had a lower mass fraction distribution for elements H and 440 C, and a higher oxidant elemental mass fraction of N and O over the radial region of $-0.5 \le r/R \le 0.5$. For 441 disk, ds = 45 mm, the reduction in C and H mass fractions at X/D = 0.074 was approximately 8%, and as the 442 distance from the burner rim further increased to X/D = 2.5, the reduction in C and H mass fractions 443 increased up to 30% in comparison to those observed with the disk, ds = 25 mm. This can be understood 444 by referring to the numerical analysis of the current burner, which showed that the large disk, ds = 45 mm, 445 was associated with higher turbulence intensity, and subsequently, the mixing layer during the burner 446 mixing length, L, will be changed [43]. This higher intensity will be associated with a large rate of strain, 447 which reduced the annulus fuel stream, to entrain more into the air stream and the rate of molecular 448 diffusion will be reduced, resulting in higher Y_N & Y_O and lower Y_C, & Y_H [57-59].

The elemental mass fraction of H and C were reduced for both disks, as the distance from the burner rim 450 451 increased further, due to the increased time available for the surrounding air to penetrate into the partially 452 premixed flame [26]. It was noticed that the elemental mass fraction of H and C were reduced for both the 453 disks as the distance from the burner rim further increased due to the increased time available for the 454 surrounding air to penetrate deeper into the partially premixed flame. Furthermore, at the edge of the burner the values of the elemental mass fraction of O and N were very close to that components of 455 456 atmospheric air, and moving toward the burner centerline, these values were reduced. This could be linked 457 to the shear layer generated due to the diffusion between the air and fuel.



Figure.12. Effect of changing the disk slit diameter on the elemental mass fraction distributions for the lean partially premixed flame of $\varphi = 0.8$ at different axial positions for the NG/air mixture.

1.5

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1.5

1.5

458 3.4. Mixture Fraction Measurements

459

460 Based on the calibration curves shown in Figure 9, the local equivalence ratio could be identified for each 461 measured point according to any of the three intensity ratios C/(N+O), H/N and H/O. The local equivalence 462 ratio was calculated as an average of 50 laser shots. Then the mixture fraction ξ can be either calculated 463 based on the obtained elemental mass fractions values presented in Figure 8 and substituted in Eq. (2), or 464 by using the local equivalence ratio determined from the linear calibration trend and substituting it into Eq. 465 (3):

$$\xi = \sum y_{Fi} \tag{2}$$

467

466

468 where y_{Fi} are the fuel elements' mass fractions. Furthermore, the mixture fraction, ξ can be employed to 469 evaluate the average equivalence ratio based on Eq. (3):

$$\xi = \frac{\varphi}{\varphi + (\frac{A}{F})_{st}} \tag{3}$$

470

471 For the current NG/air flame the stoichiometric air-to-fuel ratio(A/F)_{st} was calculated as 17.167.472

The impact of changing the disk slit diameter on the mean radial profile distribution of equivalence ratio or mixture fraction at different axial positions of X/D = 0.074, 1.11, 1.85 and 2.5 are presented in Figure 13. All of the measurements were conducted at lean NG/air mixture conditions of $\varphi = 0.8$, V_j = 1.6 m/s and partially premixed degree of L/D = 2, to investigate the structure of the flame. The results demonstrated that the equivalence ratio distributions were significantly influenced by altering the disk slit diameter, due to variation of both the mean flow structure and the level of turbulence intensity associated with each disk [60].

481 Figure 13 illustrates that around the burner axis, a perfectly symmetrical distribution of the mixture 482 fraction, ξ profile was manifested, and the mixture fraction values peaked at the burner axis and rapidly decreased towards the periphery (air side), especially for lower disk slit diameter, ds = 25 mm. This could 483 484 be interpreted by referring to the numerical investigation of the turbulent intensity produced from the 485 current burner at L/D = 2 [43]. It was noticed that disk ds = 25 mm had a faster decay rate of the turbulent 486 intensity during the mixing length, L, in comparison to that of ds = 45 mm. Consequently, a higher 487 percentage of the fuel introduced through the annulus will be diffused towards the burner centerline for 488 ds = 25 mm during the mixing length, L. The higher level of intensity associated with the large disk, ds = 45 489 mm, yielded eddies of sufficient strength and with a large rate of strain, which enhanced the entrainment 490 of the annulus fuel to the air stream. As a result, a homogenous mixture fraction distribution will be 491 produced and this was consistent with the results of Meares and Masri [61], where the fuel was introduced 492 by the same way. 493

494 As shown in Figure 13, the smaller turbulence generator disk slit diameter of ds = 25 mm consistently 495 encompassed a wide range of equivalence ratios distributions in comparison to that of ds = 45 mm. At an 496 axial distance of X/D = 0.074, the radial distribution of φ for disk ds = 25 mm was varied from very lean 497 condition of φ = 0.67 to very rich conditions of φ = 1.25, whilst for the disk ds = 45 mm, the radial local 498 equivalence ratio profiles were varied from a slightly lean condition of φ = 0.86 to a slightly rich condition 499 of $\varphi = 1.1$. As the axial distance from the burner rim further increased, the radial distribution of φ became 500 narrower and the flames were naturally anchored to the burner lips and they took a conical shape, 501 especially at an axial distance of X/D = 2.5 for ds = 25 mm [62]. Furthermore, the disk slit diameter, ds = 45 502 mm, had a perfectly flat local equivalence ratio distribution for axial distances of X/D = 0.074 and 1.11, 503 within the burner wall boundary. Also, the created flammable region for disk, ds = 25 mm, was decreased 504 from rich condition around $\varphi = 1.25$ on the jet axis to stoichiometric conditions at r/R = 0.5 and then to 505 lean conditions, while moving radially further toward the air side. For the disk, ds = 45 mm, a rich 506 homogeneous mixture of φ =1.1 was radially distributed over the burner wall boundary and then started 507 to decrease into stoichiometric and lean conditions at the periphery (air side).

509 The high turbulence level associated with the large disk, ds = 45 mm, resulted in an increase in the 510 entrainment of the hot product gas and flame radicals towards incoming reactants, elevating the chemical reaction 511 rate, and hence, it helped to stabilize the flame [63-65]. Furthermore, this kind of mixing between the products and reactants diluted the mixture fraction of disk, ds = 45 mm, as compared to that of disk, ds = 25 mm. The 512 513 reduction in mixture fraction values at distances further from the burner rim can be mainly attributed to 514 the enough time available for the surrounding air to penetrate deeper into the partially premixed flame 515 [26]. Moreover, the mixture fraction flat distribution profiles highlight the ability of the current burner 516 configuration to generate highly stable partially premixed flames. In addition, previous studies [8, 66] 517 showed that the flame speed is directly proportional to the mixture fraction gradient and subsequently the 518 flame front curvature. Consequently, the non-uniformity in the ξ distribution increased the flame front 519 curvature and decreased the rate of heat transfer from the flame front, resulting in a lower preheating of 520 the unburned gas layers, and hence, a lower flame propagation speed will be produced [8].

521



522Figure.13. Impact of changing the disk slit diameter (a) ds = 25 mm and (b) ds = 45 mm, on the mean radial523profiles of equivalence ratio or mixture fraction distributions for the NG/air mixture at $\varphi = 0.8$, $V_j = 1.6$ m/s

- 524 and L/D =2 for certain axial distances.
- 525



526

530

527 Figure.14. Variation of the equivalence ratio at different axial positions from the burner tip along the burner 528 centerline with different disk slit diameters of ds = 25 mm and ds = 45 mm, for the NG/air mixture at φ = 529 0.8, V_j=1.6 m/s and L/D =2.

Figure 14 highlighted the reduction of the equivalence ratio with the increase of the axial distances along
the burner centerline for different disk slit diameters. Consistently, disk, ds = 25 mm had a higher

533 equivalence ratio at all the axial distances in comparison to disk, ds = 45 mm. This influence was mainly 534 linked to the enhancement of the surrounding air entrainment due to the higher level of turbulence intensity associated with the large disk, ds = 45 mm. As the axial distance above the burner tip increased 535 536 from 0.074 to 2.5, the equivalence ratio was reduced for both the ds = 25 mm and ds = 45 mm disks by approximately 25% and 43%, respectively. In addition, at an axial distance of X/D = 0.074, the equivalence 537 538 ratio for disk, ds = 25 mm, was higher than that of disk, ds = 45 mm, with approximately 9.6%. As the axial 539 distance from the burner tip increased to X/D = 2.5, the difference in the equivalence ratio between disk, 540 ds = 25 mm and disk, ds = 45 mm, increased up to approximately 32%.

542 The fluctuation in mixture fraction were calculated as a ratio between the root mean square of the mixture 543 fraction fluctuations and the average of the mixture fraction values along the burner centerline. Both the mean and fluctuating mixture fraction profiles (ξ and ξ^* , respectively) are presented in Figure 15 for an 544 545 axial distance of X/D = 0.074. For the lower disk slit diameter, ds = 25 mm, and lean mixture of $\varphi = 0.8$. 546 higher fluctuations occurred within the burner wall boundary compared to those observed with disk, ds = 547 45 mm. In contrast, outside the burner boundary disk, ds = 25 mm yielded lower fluctuations compared to 548 disk, ds= 45 mm. At X/D = 0.074 and $-1 \le r/R \le 1$, the peak fluctuation for disk, ds = 25 mm, was 549 approximately 0.028, whilst for disk, ds = 45 mm, it was approximately 0.017. Moreover, across the burner 550 region of $-0.5 \le r/R \le 0.5$, the mixture fraction fluctuation associated with the disk, ds = 45 mm, was nearly 551 fixed around 0.008, and lower compared to that of the lower disk, ds = 25 mm, with approximately 0.019. Figure 16 shows the fluctuating mixture fraction profiles ξ^* for the NG/air mixture at $\varphi = 0.8$ and L/D = 2, 552 553 at further axial positions from the burner rim. The mixture fraction fluctuation profiles for disk, ds = 45 554 mm, were consistently lower than that of ds = 25 mm, for most of the radial distances r/R. The maximum 555 fluctuation was noticed for disk, ds = 25 mm at an axial distance of X/D = 2.5 with ξ^* = 0.05. At an axial 556 distance of X/D = 1.11, the fluctuation trends for disk, ds = 25 mm, was slightly higher than that of the disk, 557 ds = 45 mm. While at axial distance of X/D = 1.85, the fluctuation trends for both disks were close to each 558 other.



541



561 Figure.15. Impact of changing the disk slit diameter on both the mean and fluctuating mixture fraction 562 profiles for the NG/air mixture at φ = 0.8 and L/D =2 for the axial distance of X/D =0.074.



Figure.16. Impact of changing the disk slit diameter on the fluctuation of the mixture fraction profiles for NG/air mixtures at φ = 0.8 and L/D = 2 for axial distances of (a) X/D = 1.1, (a) X/D = 1.85 and (a) X/D = 2.5.

566

567 *Conclusions* 568

569 The current study was mainly focused towards the extension of the lean combustion limit by generating 570 higher turbulence levels or by using the partially stratified charge method. Likewise, the burner was designed for improving the stability of lean flames, and consequently, it produces low emissions. In addition, 571 572 the LIBS technique was employed to characterize and quantify the impact of changing the disk slit diameter 573 on the distribution profiles of equivalence ratio or mixture fraction for NG/air partially premixed flame. 574 The current burner configuration stability was examined for a lean NG/air mixture at $\varphi = 0.8$ by changing 575 the disk slit diameters, the fuel used and the degree of partial premixing. The conclusions drawn from this 576 work are as follow:

577

578 (1) At the equivalence ratio of $\varphi = 0.8$ and for different levels of partially premixing, it was noticed that 579 changing the disk slit diameter has a great impact on the flame stability for both NG and LPG fuels. 580 Increasing the disk slit diameter from ds = 25 mm to ds = 45 mm resulted in a higher flame stability for the 581 NG/air mixture. In contrast, increasing the disk slit diameter for LPG/air mixtures reduced its flame 582 stability. 583

584 (2) Changing the level of partial premixing (L/D) lead to an enhancement in the flame stability for both 585 fuels. However, the optimum L/D ratio where the maximum flame stability occurred varied based on which 586 fuel type and disk slit diameter was used. L/D = 1 was the optimum ratio for NG with disk, ds = 25 mm, at 587 which the maximum flame stability was achieved. The maximum flame stability was achieved at L/D = 3 588 for LPG with disk, ds = 25 mm, and for NG with disk, ds = 45 mm. Finally, for LPG with disk, ds = 45 mm, L/D 589 = 4 was the optimum ratio at which the maximum flame stability occurred. 590

591 (3) Among all the cases, the NG/air mixture with ds = 45 mm and φ = 0.8 demonstrated the maximum flame 592 stability, whilst the LPG/air mixture with ds = 45 mm and φ = 0.8, yielded the minimum flame stability over 593 different levels of partial premixing. 594

595 (4) At a constant level of partially premixing of L/D = 2, it was noticed that for equivalence ratios of $\varphi \ge 0.8$, 596 the NG/air mixture with ds = 45 mm exhibited the maximum flame stability. Whilst for equivalence ratios 597 of $\varphi < 0.8$, the LPG/air mixture with ds = 25 mm yielded the maximum flame stability. 598

599 (5) The larger disk slit diameter, ds = 45 mm, demonstrated almost zero mixture fraction gradients 600 distributions and lower fluctuation of the mixture fraction distributions within the burner wall boundary 601 in comparison to that of disk, ds = 25 mm, for the NG/air mixture at ϕ = 0.8 and L/D = 2. 602

603 (6) Along the burner centerline disk, ds = 25 mm consistently had a higher equivalence ratio at all of the 604 axial distances in comparison to disk, ds = 45 mm. The equivalence ratio at axial distances of X/D = 0.074605 and X/D = 2.5, for disk, ds = 25mm, was higher than that of disk, ds = 45mm, with approximately 9.6% and 606 32%, respectively. As the axial distance above the burner tip increased from 0.074 to 2.5, the equivalence 607 ratio was reduced for both disks, ds = 25mm, and ds = 45mm, by approximately 25% and 43%, respectively. 608 (7) The mixture fraction fluctuation profiles associated with disk, ds = 45mm, were consistently lower than 609 that of ds = 25mm, for most of the radial distances r/R. Near the burner tip at X/D = 0.074, the peak 610 fluctuation for disk, ds = 25mm, was approximately 0.028, whilst for disk, ds = 45 mm, was approximately 611 0.017. The maximum fluctuation was observed for disk, ds = 25mm, at an axial distance of X/D = 2.5 with 612 $\xi^* = 0.05$.

613

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615

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619 *References*

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