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Combustion characterization of waste cooking oil and canola oil based biodiesels under simulated engine conditions

C. Ming^a, I.M. Rizwanul Fattah^a, Qing N. Chan^{a,*}, Phuong X. Pham^b, Paul R. Medwell^c, Sanghoon Kook^a, Guan H. Yeoh^a, Evatt R. Hawkes^{a, d}, Assaad R. Masri^b

^a*School of Mechanical and Manufacturing Engineering, UNSW Sydney, NSW 2052, Australia*

^b*School of Aerospace, Mechanical and Mechatronic Engineering, The University of Sydney, NSW 2006, Australia*

^c*School of Mechanical Engineering, The University of Adelaide, SA 5000, Australia*

^d*School of Photovoltaic and Renewable Energy Engineering, UNSW Sydney, NSW 2052, Australia*

Abstract

Alternative fuels will come from a variety of feed stocks and refinement processes. Understanding the fundamentals of combustion and pollutants formation processes of these fuels will be useful for their implementation in different combustion systems. In this work, optical diagnostics were performed to waste cooking oil (WCO) and canola oil (CAO) based biodiesel sprays to assess their combustion and soot formation processes. Conventional diesel was used as a reference fuel for comparison with the biodiesels. The experiments were conducted in an optically-accessible constant-volume combustion chamber (CVCC) with simulated compression-ignition engine conditions, with different degree of exhaust gas recirculation. The liquid length and lift-off length results indicate that there was no significant interaction between the liquid phases of the fuels and their combustion regions. The flame lift-off lengths were found to be affected by both the chemical and physical properties of the fuels. It was observed that a larger difference between the lift-off length and the first-luminosity distance

*Corresponding author: qing.chan@unsw.edu.au

was correlated with lesser downstream soot formation, although the molecular structure of the fuel was found to affect the process too. Assessing the sooting and combustion characteristics of the biodiesel and diesel flames across the varied ambient O₂ atmospheres revealed that the estimated soot contents of the biodiesel and diesel flames peaked at 15 and 21 vol.% O₂ concentration, respectively. The peak soot contents of the WCO and CAO biodiesel flames were found to be comparable, but lower than that of diesel, across the various O₂ environment. The results also demonstrated that the biodiesels have higher normalized peak pressure values than diesel at all O₂ conditions. Two-color pyrometry data demonstrated that the measured soot temperature and soot KL factors of the flames were similar at 15 and 21 vol.% O₂, but varied with further reduction of ambient O₂ concentration. Variations in the combustion duration and flame area were found to be fuel dependent.

Keywords: Biodiesel, Waste cooking oil, Canola oil, Two-color pyrometry

1. Introduction

Diesel-fueled compression-ignition engines are becoming increasingly popular choice for ground transport vehicles because of their high torque output and fuel efficiency. However, particulate emissions from diesel engines, which can profoundly affect human health and environment [1, 2], remain a major concern. Using alternative diesel fuels, such as biodiesels, is widely considered to be a promising method to control particulate emissions from compression-ignition engines during combustion. The oxygen content of biodiesel fuels have been reported to provide an effective route to enhance combustion and to inhibit soot formation in compression-ignition engines [3, 4]. This has motivated many research and development activities, ranging from the modifications of the upstream cultivation process to enhance feedstock production for biofuels [5], down to the detailed study of combustion and emission characteristics of biodiesel fuels in the diesel engines [3, 6] over the past decades. Table 1 presents a summary of selected experimental investigations involving combus-

16 tion diagnostics of biodiesels in optically-accessible combustion chambers that
17 are relevant to this work, as an example of the quantity of fundamental studies
18 performed in this subject area.

19 Despite the many previous studies, the impact of the properties of the
20 biodiesel fuels on soot emissions remain only vaguely known due to the mul-
21 titude and complexity of the associated processes. In addition, biodiesels are
22 mixtures of fatty acid methyl esters (FAMES) with different chain lengths and
23 degrees of saturation, which can vary significantly depending on their feedstocks
24 [7, 8]. Such variations can have a complicated impact on biodiesel atomization,
25 evaporation, combustion and soot formation processes, and hence, the ensuing
26 engine performance [4, 8]. The lack of control of all of these factors have led
27 to contradictory results being reported in the literature. For instance, many
28 studies have reported that fuels with higher cetane numbers have shorter igni-
29 tion delays, which would therefore lead to an increased soot formation due to
30 insufficient fuel-ambient mixing before the onset of combustion [9]. Nonetheless,
31 there are other studies that show reduction in soot and unburned hydrocarbon
32 emissions, when fuels of high cetane number values were used [10, 11]. The dif-
33 ferences are likely to be attributable to molecular structure effects of the fuels
34 used. In addition, in a previous study conducted by Pham *et al.* [12], it was re-
35 ported that the oxygen content in biodiesels would contribute towards particle
36 formation suppression in the premixed spray combustion zone, and increased
37 particle oxidization at the flame front. In a separate study that was conducted
38 by Park *et al.* [9], however, it was demonstrated that the soot formation is
39 mainly controlled by the fuel-air mixing process for fuels with similar amount
40 of oxygen within the fuel, and number of carbon-carbon bonds. These findings
41 demonstrate the difficulties in drawing conclusions from a change to a single
42 fuel property, which could simultaneously impact on other fuel properties [13].
43 The complexities that arise from the inter coupling between the various fuel
44 properties produce some degree of uncertainty as to which effect would have a
45 greater impact on the soot formation process of the flame [14].

46 Engine operating parameters, such as fuel injection pressure and exhaust

47 gas recirculation (EGR), can have a pronounced impact on the combustion and
48 pollutant formation processes of the fuel in compression-ignition engines. High
49 pressure fuel injection, for example, could be used to improve spray atomization
50 and combustible mixture preparation, which can lead to reduced particulate
51 emission and improved engine performance [14, 15]. The introduction of EGR
52 can result in low temperature combustion (LTC) condition, which can lead to
53 reduced NOx emissions [16] from compression ignition engines. The effects of
54 these varied operating parameters on the engine and emission performances,
55 however, can also change significantly depending on the fuel type used. For
56 example, it has been reported that increasing injection pressure can potentially
57 lead reduced flame residence time available for fuel elements to undergo soot
58 formation [17], in addition to the general consensus that it can help improve
59 the atomization and vaporisation processes of a fuel. Nonetheless, other studies
60 have also demonstrated that high fuel injection pressure can lead to wall-wetting,
61 which would result in increased engine-out emissions of uHC, CO and smoke
62 [18]. The wall-wetting issue is generally more problematic when biodiesels are
63 used, as they typically have longer spray penetrations due to their higher fuel
64 densities when compared with diesel [19]. In addition, in a previous work that
65 was conducted by Zhang *et al.* [20], it was reported that the effects of EGR may
66 vary depending on the fuels such that a lower oxygen-concentration environment
67 was necessary for diesel to achieve a higher combustion efficiency and lower
68 soot emission, when compared with biodiesels. All these highlight the need to
69 perform a direct assessment the effects of common engine operating parameters
70 on the combustion and pollutant emission processes for the biodiesel fuel of
71 interest, to ensure efficient and clean engine operation when used.

72 This work is therefore aimed at assessing the combustion properties of
73 biodiesels from the transesterification of waste cooking oil (WCO) and canola oil
74 (CAO) under simulated compression-ignition engine conditions in a constant-
75 volume combustion chamber (CVCC). The WCO biodiesel is specifically tar-
76 geted as it is the main biodiesel feedstock in Australia [21]. The CAO biodiesel
77 is also selected for study as it has been extensively investigated by the authors

78 under open flame burner and performance engine settings [12], in addition to
79 having fuel properties that are similar to the WCO biodiesel, and is therefore
80 useful for comparison. Optical diagnostic techniques, including OH chemilumi-
81 nescence and high-speed flame natural luminosity imaging, are used to assess
82 the impact of these varied parameters both on the air entrainment rate occur-
83 ring upstream of the lift-off length, and on the downstream soot formation and
84 combustion processes of the biodiesel jet flames. The two-colour pyrometry
85 technique is applied to the recorded flame natural luminosity images to provide
86 soot temperature and concentration information for the jet flames under the test
87 conditions. To provide context to the examination, comparative measurements
88 using a conventional diesel are also performed.

89 **2. Experimental Details**

90 *2.1. Constant-volume Combustion Chamber*

91 Experiments were performed in an optically-accessible CVCC under simu-
92 lated, quiescent diesel engine conditions. The CVCC has a $114 \times 114 \times 114$ mm
93 cubical combustion chamber, six window ports, and eight access ports for instru-
94 mentations and valves. For this work, sapphire windows were installed at four
95 sides of the chamber to enable optical access. A diesel injector was mounted at
96 the centre of a metal side port so that the fuel spray was directed into the centre
97 of the chamber. An agitator was mounted at the top of the CVCC and was used
98 to maintain a spatially uniform temperature environment. Schematic diagrams
99 of the CVCC configuration along with the diagnostic set-ups used, and the cross-
100 sectional view of the CVCC are shown in Fig. 1. The operation of the chamber
101 was initiated by filling it with a premixed, lean combustible charge (mixture
102 of acetylene, hydrogen, nitrogen and oxygen). The mixture was subsequently
103 ignited with a spark plug to create a high temperature and pressure environ-
104 ment inside the chamber. For this work, an ambient condition with gas density,
105 pressure and temperature of 19.5 kg/m^3 , 6 MPa and 1100 K, respectively, was
106 targeted and the fuel injector was triggered once the targeted condition was

107 reached. The operating conditions were used to match experimental conditions
108 that were selected to ensure that the combustion process of the biodiesels and
109 diesel are fairly complete in previous CVCC studies [20, 22]. The metering of
110 the reactant gases was also controlled so that an environment with 0 or 10, 15
111 and 21 vol.% O₂ concentration can be generated after the premixed combustion
112 process. The 0 vol.% ambient O₂ concentration (non-reacting) condition was
113 used to provide insights into the liquid length penetration characteristics of the
114 sprays with varied injection schedules, without the complex effects of heat re-
115 lease from the combustion processes. The 10 and 15 vol.% (reacting) ambient
116 O₂ conditions were used, on the other hand, to simulate compression-ignition
117 engine conditions with high and moderate exhaust gas recirculation [23, 24].
118 The test conditions for the experiments are summarized in Table 2.

119 The fuel injection system used comprised of a low-pressure fuel pump a high-
120 pressure fuel pump, a Bosch commercial common-rail system (CP4) and an axi-
121 ally drilled single-hole solenoid injector with an orifice diameter of 105 μm. The
122 single-hole injector was used to avoid the complexity that can arise from jet-jet
123 interaction. A common-rail PCV driver (Zenobalti, ZB-1200) was used to reg-
124 ulate and maintain the injection pressure, whilst an injector driver (Zenobalti,
125 ZB-5100) was used to ensure the injection system control the injection duration
126 to deliver the required fuel mass. A summary of the injection conditions used
127 is provided in Table 2.

128 *2.2. Optical Diagnostics*

129 *2.2.1. Combustion Diagnostics*

130 A summary of the optical diagnostic techniques and settings used for this
131 work is provided in Table 3. The OH chemiluminescence images were captured
132 using an intensified CCD (ICCD) camera, whilst the flame natural luminosity
133 images were recorded with a high-speed CMOS camera. The cameras were
134 arranged for viewing through the side or bottom windows of the CVCC, as
135 shown in Fig. 1, and were applied simultaneous to image the same injection
136 events. For the OH-chemiluminescence imaging, the light emission from flame

137 was detected through a 10 nm bandwidth interference filter (centred at 310 nm),
138 using the ICCD camera (Andor iStar) that was fitted with a 50 mm f#3.5 UV
139 lens. The camera was triggered to open for 2.5 ms after the quasi-steady lift-off
140 length of the flame was established, and was closed before the end of injection.
141 The pixel resolution of the image is ~ 0.1 mm per pixel. A typical recorded OH-
142 chemiluminescence image is shown in Fig. 2. In this sample image, the injector
143 orifice is located at the left edge of the image, and the WCO biodiesel was
144 injected from left to right at 100 MPa injection pressure and 15 vol.% ambient
145 O₂ concentration.

146 For the high-speed flame luminosity imaging, a CMOS camera (Photron
147 SA5) equipped with a 85 mm f#1.8 lens that was operated at a frame rate of
148 15,000 frames per second (fps) was used. The CMOS camera was synchronized
149 to the start of injection of the injector, and 100 images were recorded to ensure
150 that the entire combustion event, from the start of injection till the end of com-
151 bustion, was captured for each run. A typical instantaneous flame luminosity
152 images captured for the same WCO biodiesel jet flame presented, but at 2.4 ms
153 after start of injection (aSOI), is shown in Fig. 3. The pixel resolution of the
154 image is ~ 0.2 mm per pixel. The instantaneous flame luminosity images that
155 were recorded for both CAO biodiesel and diesel flame, under the same experi-
156 mental conditions, are also presented for comparison purposes. In these images,
157 the fuels were also injected from left to right. It is noted that at least five runs
158 were performed for each measurement to ensure good repeatability.

159 *2.2.2. Spray Diagnostics*

160 Liquid length, which is defined as the maximum distance from the injector
161 tip to liquid fuel penetration for an evaporating fuel spray [25], was measured
162 for each of the fuels to determine the extent of its liquid phase. It is noted that
163 the diffused back illumination (DBI) technique, which has been reported to be
164 more quantitative when compared with other methods because of its built-in
165 reference light intensity [26], was used as the main diagnostic method to obtain
166 liquid length information. For DBI imaging, the collimated light output from

167 a high-powered, continuous LED that was emitting at a central wavelength
168 of 455 nm, was used for illumination. An engineered diffuser was also placed
169 between the LED light and the spray to ensure homogenized illumination. A
170 high-speed CMOS camera, which was placed directly opposite to the LED light
171 source, was operated at a frame rate of 15,000 frames per second (fps), with
172 1 μ s exposure time and a pixel resolution of \sim 0.14 mm per pixel. For the DBI
173 technique, the camera was also synchronized to the start of injection of the
174 injector and 100 images were captured for each run.

175 To determine the extent of the liquid phase in the DBI images, the light
176 intensity, I , at a pixel location from an image with spray, was compared against
177 the background intensity, I_o , at the same pixel location from an image with
178 no spray. In this work, the calibration of the DBI technique was performed by
179 comparing the DBI images and Mie-scattering images that were recorded using
180 the current experimental setup, for a series of fuel sprays that were subjected to
181 the same set of test conditions. The Mie-scattering images were obtained using
182 the same high-speed camera and high-intensity LED, but with side illumination.
183 The Mie-scattering images obtained were processed using a threshold of intensity
184 that corresponded to 3% of the maximum intensity, following the widely-used
185 Sieber's approach for Mie-scattering post-processing [25, 26]. The cut-off I/I_o
186 value of the DBI images was subsequently determined as 0.3, so that the results
187 of the measurement techniques would match.

188 *2.3. Flame luminosity image post-processing*

189 In this study, a novel color band approach, which was first introduced by
190 Larsson [27] and further explained by Svensson *et al.* [28], was applied to the
191 recorded flame luminosity images, to provide two-dimensional soot temperature
192 and soot distribution information. The color band approach, which only re-
193 quires the use of a single digital color camera for soot two-color measurements,
194 has the advantage of avoiding errors introduced due to potential misalignment
195 of frames taken by two cameras and eliminates the need for complex optics
196 that are typically required for the more classical implementation of two-color

197 pyrometry technique. The general theory of two-color thermometry has been
 198 described in detail in various papers [27–29], a complete treatment is therefore
 199 not provided here. In essence, the signal from an optical detector viewing a
 200 radiating soot cloud can be described using Planck’s blackbody function that
 201 incorporates a wavelength-dependent soot emissivity model, as well as the com-
 202 bined wavelength-dependent responsivity of the detection system used (optics
 203 train, filter and high-speed camera). The pixel output, S , when observing soot
 204 irradiation at wavelength, λ , can therefore be described as:

$$S_\lambda = \int \epsilon_{soot} \frac{C_1 Q(\lambda)}{\lambda^5 \left[\exp\left(\frac{C_2}{\lambda T_{soot}}\right) - 1 \right]} d\lambda. \quad (1)$$

205 In Eq. 1, $Q(\lambda)$ refers to the optical system response coefficient for each
 206 wavelength, C_1 and C_2 are the Planck’s first and second constants, respectively,
 207 and T_{soot} is the soot temperature. The soot emissivity, ϵ_{soot} , can be modeled
 208 with the empirical correlation of Hottel and Broughton [30]:

$$\epsilon_{soot} = 1 - \exp\left(-\frac{KL}{\lambda^\alpha}\right), \quad (2)$$

209 where α in Eq. 2 is given as [31]:

$$\alpha = 1.22 - 0.246 \times \ln(\lambda). \quad (3)$$

210 The product KL in Eq. 2 is widely used to characterize the soot content
 211 in various combustion systems [28, 29]. From Eq. 1, it can be seen that if the
 212 optical response coefficient, $Q(\lambda)$, is known through calibration, S_λ becomes
 213 a function of two unknown variables, T_{soot} , and KL . Given that there are
 214 two unknowns, any two of the three color channels can therefore be used to
 215 produce two equations to solve for T_{soot} and KL . For this work, only signal
 216 from the red and green channels were used for the two color calculations, as
 217 these two channels have higher signal-to-noise ratios than that of blue (SNRs of
 218 47, 21 and 9 were measured for red, green and blue channels, respectively). The
 219 optical response coefficients of the detection system were determined through

220 calibration using a tungsten filament light source with known spectral emission
221 properties.

222 *2.4. Fuel selection*

223 For this work, two biodiesels (waste cooking oil and canola oil based) pos-
224 sessed long carbon chain lengths, with high unsaturation degrees were studied.
225 Diesel was selected as a reference fuel to be used for comparative studies with
226 these biodiesels. From the fuel specifications data that were provided by the
227 fuel suppliers, both the waste cooking oil (WCO) and canola oil (CAO) based
228 biodiesels have higher cetane number, fuel density, viscosity values when com-
229 pared with diesel. Both biodiesels have a high degree of unsaturation, as are
230 reflected by their iodine values. For the injection conditions used in this work,
231 the difference in the actual injected mass of the biodiesels and diesel, when mea-
232 suring with a Bosch tube type injection rate meter, was found to be within exper-
233 imental errors ($\sim 10\%$) of the injection rate meter across the varied conditions.
234 The same injection parameters were therefore applied to biodiesels and diesel
235 for all experiments. Gas-chromatography and mass-spectrometry (GCMS) were
236 performed on the biodiesel fuels to determine their chemical compositions. Both
237 WCO and CAO biodiesels are found to comprise mainly of FAMEs with long
238 carbon chains such as oleic, linoleic and linolenic acids, and are oxygenated (11
239 wt%) leading to their lower heating values and lower stoichiometric mixture
240 fractions than diesel. A summary of the main fuel characteristics of all fuels
241 and main FAME fractions of the biodiesels are provided in Tables 4 and 5,
242 respectively.

243 **3. Results and discussion**

244 *3.1. Lift-off length, first-luminosity distance and soot formation distance*

245 Flame structural information, including flame lift-off and first-luminosity
246 distance from the nozzle, during its mixing-controlled combustion phase has
247 been shown to be critical towards the understanding of soot formation process

248 [32]. The flame lift-off length, which refers to the distance from the fuel injector
249 nozzle to where the flame stabilizes, is indicative of the location where intense
250 combustion reaction of the flame initiates [33], and is widely used to charac-
251 terize the amount of fuel-air premixing occurring upstream of the lift-off [34].
252 The first-luminosity distance, on the other hand, refers to the distance from the
253 injector to the location where intense flame luminosity is first detected. The
254 first-luminosity distance is useful to indicate the initial onset of soot, as the
255 natural flame luminosity detected is dominated by soot incandescence in sooty
256 flame. The lift-off lengths of the flames were derived from their time-averaged
257 OH chemiluminescence images, following the approaches that were detailed in
258 Ref. [33]. In brief, the lift-off length value was determined by finding the dis-
259 tance between the injector and first axial location where chemiluminescence
260 intensity of the flame exceeded $\sim 50\%$ of the leveling-off value, as is shown in
261 Fig. 2. The first-luminosity distances, on the other hand, were derived from
262 the high-speed natural flame luminosity images that were captured using the
263 high-speed camera. The images were first corrected for background, dark charge
264 and detector attenuation, and were subsequently binarized to enable logical op-
265 erations [35, 36]. The first-luminosity distance was determined by measuring
266 the minimum distance from the injector nozzle exit plane to the location where
267 half of the pixels on an arc with a spreading angle of $\theta/2$ are occupied by the
268 binarized flame area. The flame penetration distance was also determined using
269 the same methodology, but is defined as the maximum distance between the in-
270 jector and the aforementioned arc. These definitions were used to minimize the
271 sensitivity of the first-luminosity distance and flame penetration distance calcu-
272 lations to the threshold settings used or potential fluctuations in the binarized
273 flame area. It is noted that the flame central axis was taken as the line from the
274 injector tip through to the centroid of the flame area, whilst flame spreading
275 angle was defined as the average flame spreading angle between a starting and
276 ending points along the flame central axis. The starting and ending points were
277 set to be 45% and 55% of the instantaneous flame penetration distance.

278 The average lift-off lengths and the first-luminosity distances of the WCO

279 and CAO biodiesels and diesel are plotted as functions of injection pressures,
280 for the test conditions with ambient O₂ concentrations of 10, 15 and 21 vol.%
281 in Fig. 4. Dashed lines through the data are included to help the visualization
282 of the trends. Due to the similarity in the trends of the WCO, CAO and diesel
283 data to the effect of ambient O₂ concentration, the data of diesel and CAO
284 under ambient concentration of 10 and 21 vol.% are not shown here. From
285 Fig. 4, it can be seen that lift-off lengths of the flames are found to increase
286 with increasing injection pressure or decreasing ambient O₂ concentration. It
287 is noted that previous studies have shown that an increase in injection pressure
288 would result in higher velocity in spray, which would push the initial combustion
289 zone of the flame downstream [37]. A reduction in ambient O₂ concentration,
290 on the other hand, has been shown to necessitate a greater entrainment of total
291 ambient gas with smaller fraction of oxygen (hence, longer mixing time) before
292 combustion reactions can take place [38]. The current results are therefore
293 consistent with the previously observed trends in lift-off lengths with respect to
294 injection pressure and O₂ concentration effects.

295 From Fig. 4 it can be observed that the lift-off lengths of the WCO biodiesel
296 flames are consistently longer than those of diesel flames, whilst the lift-off
297 lengths of the CAO biodiesel flames are found to be the lowest amongst the
298 fuels used. Previous studies have demonstrated flame lift-off length is affected by
299 both the chemical and physical properties of the fuel [39]. From Table 4, it can
300 be seen that the cetane number of the WCO biodiesel (*i.e.*, 53.2), albeit slightly
301 higher, is comparable to that of diesel (*i.e.*, 51). Previous studies [13] have noted
302 that there is an inherent uncertainty in the reported cetane number of the fuel,
303 which can be dependent on the measurement methodology and testing facility
304 used. The ambient conditions used for cetane number measurements are also
305 significantly different to the experimental conditions of this work. Therefore, to
306 eliminate the potential ambiguity of the cetane number rating and the actual
307 ignition delay, the light-based ignition delay of the fuels, when injected at a
308 baseline injection pressure of 100 MPa, were assessed. From the measurement
309 results, the ignition delays for the biodiesel and diesel fuel jets were found to

310 be 0.6, 0.4 and 0.33 ± 0.06 ms at ambient oxygen conditions of 10, 15 and 21%
311 O_2 concentrations, respectively. The ignition delays of the fuels are observed to
312 follow the expected trend of decreasing ignition delay with increasing ambient
313 gas oxygen concentration. The invariant ignition delay values measured for the
314 fuels, however, do not match their lift-off length trend. It is noted that whilst the
315 ignition characteristic of a fuel is generally expected to affect its lift-off behavior,
316 such that a fuel with smaller ignition delay would display shorter lift-off, many
317 previous studies [13, 40, 41] have reported difficulty in observing such one-to-
318 one correlation between the lift-off length and the fuel ignition characteristic.
319 Whilst the exact reason behind this discrepancy in trends is still unclear, but
320 there is some evidence that several other factors, such as first-stage cool flame
321 activity [13, 41], can potentially affect the lift-off behavior of the fuel. It is
322 worth noting, however, the viscosity of the WCO biodiesel is greater than that
323 of diesel, which can impact on the extent of the atomization, vaporization and
324 fuel-air mixing processes of the fuel. The higher heat of vaporization value,
325 which could be attributed to the presence of FAMES with longer carbon chains
326 [25] in the biodiesel, would affect the vaporization and fuel-air mixing processes.
327 All these can lead to a prolonged time between the beginning of fuel injection
328 and the start of combustion process that would subsequently impact the ignition
329 quality of the WCO biodiesel fuel. In the case of the CAO biodiesel, however,
330 the cetane number of the CAO biodiesel (*i.e.*, 59), however, is comparatively
331 higher than both WCO biodiesel and diesel. The higher chemical reactivity of
332 the CAO biodiesel would therefore weaken the effect of delay that can arise from
333 its physical properties on the auto-ignition process. The liquid lengths of the
334 WCO and CAO biodiesels were also measured and were found to be ~ 8.7 mm
335 and ~ 8.6 mm, respectively, which are longer than that of diesel (~ 7.7 mm).
336 The liquid lengths of all fuels were, nonetheless, noted to be shorter than their
337 lift-off lengths, suggesting that there was no significant interplay between the
338 spray and combustion processes for the fuels under the test conditions of this
339 work.

340 From Fig. 4, it can also be seen that the first-luminosity distances of

341 biodiesels and diesel increase with increasing injection pressure or decreasing
342 ambient O_2 concentration. The first-luminosity distance values are also con-
343 sistently greater than the corresponding lift-off lengths of the jet flames. The
344 finding is consistent with the observations of previous studies [32, 34] that have
345 reported that the soot precursors, which are the foundation for soot growth
346 downstream, occurred from the partially premixed fuel-rich combustion zone up-
347 stream. From the figure, it can also be seen that the first-luminosity distances,
348 and hence, the soot inception locations, of both WCO and CAO biodiesels are
349 located downstream to that of diesel. As is noted in previous studies [34], once
350 soot is formed, the soot can be transported from the soot formation and growth
351 regions into the flame boundary or tip regions, where soot oxidation reactions
352 would occur [34]. It is noted that the maximum flame penetration distances that
353 are determined from the average of the instantaneous flame luminosity images,
354 such as shown in Fig. 3, recorded at injection timings just prior to the end of
355 injections for the biodiesel fuels are similar or shorter than that of diesel at all
356 conditions. It can therefore be anticipated that the soot formed in the biodiesel
357 flames would have less residence time for soot formation and growth processes,
358 prior to soot oxidation, when compared with diesel flame.

359 In previous studies, it has been shown the soot inception time of a jet flame
360 can be approximated from the observed location of the first-luminosity forma-
361 tion relative to the flame lift-off [34]. This distance of lift-off length to soot
362 formation region, which is termed as soot formation distance in the subsequent
363 discussion, has been shown to depend strongly on fuel type, such that a longer
364 distance is observed for a fuel with lower soot formation tendency [32, 34]. In
365 Fig. 5, the soot formation distances of the WCO biodiesel, CAO biodiesel and
366 diesel jet flames are plotted as functions of the ambient O_2 concentrations. The
367 peak spatially integrated natural luminosity (SINL) values of the biodiesel and
368 diesel flames under different test conditions are also computed and are presented
369 in Fig. 5. It is acknowledged that the SINL signal, which is calculated by sum-
370 ming up the intensities of all pixels of each image, is a complex function of the
371 soot concentration, properties, flame area and temperature [42]. Nonetheless,

372 the peak SINL has been widely demonstrated to be useful in providing a rela-
373 tive measure of the soot content of sooty flames, and is therefore used here to
374 rank-order the soot level of the flames.

375 In Fig. 5, it can be seen that under the same test and injection conditions,
376 the soot formation distances of the biodiesel flames are consistently longer than
377 that of diesel. As discussed in previous studies (*e.g.*, [32]), the progression of
378 soot formation downstream to the lift-off length is complex, and is dependent
379 upon mixture stoichiometry, original molecular structure, local mixture temper-
380 ature and residence time. The combined fuel oxygenation, molecular structure
381 and fuel composition effects of the biodiesels would therefore contribute towards
382 their observed longer soot formation distance when compared with diesel. From
383 Fig. 5, it can be seen that a higher peak SINL value/soot content is observed
384 for flame with shorter soot formation distance. As discussed in previous studies
385 [32], a longer soot formation distance implies that more residence time is re-
386 quired before soot inception or any subsequent soot growth can occur. During
387 this time, the continual entrainment and mixing of ambient O_2 and hot com-
388 bustion products from the surrounding and diffusion flame could contribute to
389 the suppression of the total soot growth, and hence, the soot content within the
390 flames [32].

391 From Fig. 5, it can be seen that the biodiesel and diesel flames display differ-
392 ent characteristics across the varied ambient O_2 concentrations. Specifically, the
393 soot formation distance of diesel is found to decrease monotonically with increas-
394 ing ambient O_2 concentration. The soot formation distance of the biodiesels, on
395 the other hand, is observed to first decrease and then increase as the ambient
396 O_2 concentration increases from 10 to 21 vol.%. As is noted in the previous
397 studies [43], at lower ambient O_2 concentration, an increase in the entrainment
398 of ambient gases into the flame would occur to compensate for the reduction in
399 O_2 concentration. Such increase in gas entrainment would result in a lowering
400 of flame temperature and hence, reaction rate, as there would be more inert
401 gases to be heated up within the flame. The dilution effect would therefore also
402 slow down the soot formation and oxidation processes, resulting in less net soot

403 generation. The impact of reduced ambient O_2 concentration, however, have
404 been reported in several previous studies to be less significant for biodiesels
405 than diesel. For example, Mayo *et al.* [39] observed that soybean biodiesel
406 maintained a higher high-temperature reactivity than diesel under reduced O_2
407 concentrations. In their study, they attributed their results to the fuel oxygen
408 availability in their biodiesel fuel. It is noted that similar observations were also
409 made in an experimental study that was performed by Zhang *et al.* [20], such
410 that the SINL values of the waste cooking oil biodiesel and diesel flames were
411 observed peaked at O_2 concentrations of 15 and 21 vol.%, respectively, when
412 the ambient O_2 concentration was varied from 12 to 21 vol.%. In their study,
413 it was suggested that under the ambient O_2 concentration of 15 vol.% and a
414 bulk temperature of 1000 K, the soot formation process of the biodiesel flame
415 may have been maintained without sufficient oxidation, which resulted in the
416 increased soot yield observed.

417 A closer inspection of the combustion characteristics of the biodiesels in
418 Fig. 5 reveals that, for the test conditions of this study, the soot formation
419 distances of the CAO biodiesel flames are consistently longer than that of the
420 WCO biodiesel flames. This is despite the CAO biodiesel flames having shorter
421 lift-off lengths (see Fig. 4) and would therefore experience less air entrainment
422 prior to the initial combustion zone (*i.e.*, upstream of lift-off length). In ad-
423 dition, despite the differences in the soot formation distances of the biodiesel
424 flames, the peak soot content of the flames were still found to be comparable.
425 These findings therefore imply that there are other factors that have impacted
426 on the soot formation process, other than fuel-air mixing. In a recent study that
427 was presented by Park *et al.* [9] involving various oxygenated fuels, it has been
428 demonstrated that the fuel molecular structure would strongly affect the soot
429 formation process of the flame, in addition to fuel-air mixing. From Table 4,
430 it can be seen that whilst the biodiesels are similar in terms of the values of
431 their oxygen content and stoichiometry air to fuel ratios, the CAO biodiesel has
432 a slightly longer average carbon chain length (a lower saponification number
433 implies longer chain length [12]) than WCO biodiesel. Previous studies have

434 shown that the chain length is inversely correlated to the ignition delay, such
 435 that a longer chain length would contribute towards decreased ignition delay
 436 (*i.e.* higher cetane number), and hence, a lesser fraction of premixed combus-
 437 tion for the flame. From Table 4, it can also be seen that the CAO biodiesel has a
 438 slightly lower degree of unsaturation, when compared with the WCO biodiesel.
 439 It is noted that iodine number is a measure of the mass of iodine consumed
 440 per 100 g of fatty acids, such that a unit of iodine number would indicate the
 441 presence of a mole of carbon double bond per 100 moles of fatty acids with
 442 an average molecular weight of 280 g/mol. Given that previous studies have
 443 demonstrated that there is a correlation between unsaturated FAME content in
 444 biodiesel and the amount of soot-precursor-generating species produced during
 445 combustion [44, 45], a lower degree of unsaturation would result in lesser soot
 446 propensity for the flame. Together, these findings again highlight the total soot
 447 contents of the flames are affected by the combined effects of fuel-air mixing
 448 and molecular structures on the soot formation and oxidation processes [46],
 449 and the challenges in determining which effect is more significant on the soot
 450 processes.

451 For further insights into the combustion characteristics of the fuels under
 452 varied O₂ concentration conditions, the normalized peak pressure (NPP) values
 453 were calculated [43, 46] to provide heat release information of the fuels . The
 454 NPP value measured during the combustion has been demonstrated to be re-
 455 flective of the actual thermal potential of the fuel under the test condition and
 456 experimental equipment, and can be estimated using:

$$NPP = \frac{p_f}{p_r} \times \frac{E_r}{E_f}, \quad (4)$$

457 where p_f and p_r refer to the peak pressures of the tested and reference fuels.
 458 The energy input values of the tested and reference fuels, which are represented
 459 using E_f and E_r , can be calculated using $E = \rho V Q_{LHV}$. The NPP values of
 460 the WCO, CAO biodiesels and diesel, which were injected at a fixed injection
 461 pressure of 100 MPa, into ambient conditions with O₂ concentrations of 10, 15

462 and 21 vol.%, respectively, are presented in Fig. 6. Diesel, injecting into an
463 ambient environment with 21 vol.% O₂ concentration at an injection pressure
464 of 100 MPa, is used as the reference fuel for the plot. From the figure, it can be
465 seen that the biodiesels have larger NPP values, and therefore higher thermal
466 efficiencies than that of diesel, across the varied ambient O₂ conditions. In
467 addition, it can also be seen that the NPP values of the biodiesels are found
468 to be slightly higher at an ambient condition of 15 vol.% O₂, whereas the NPP
469 value of the diesel is observed to peak at 21 vol.% O₂. The NPP values of all
470 fuels, however, were found to be at their lowest at 10 vol.% O₂. It is noted a
471 similar observation is also reported in a previous study by Liu *et al.* [43] to
472 examine the combustion characteristics and soot distributions of butanol and
473 soybean biodiesel fuels under varied ambient O₂ concentration conditions (from
474 21 to 10 vol.%) but at a fixed ambient temperature of 1000 K. In their study,
475 it was found that the highest heat release values were measured, when the fuels
476 were subjected to an ambient condition with O₂ concentration of 16 vol.%. The
477 fuels were also observed to have the lowest pressure rise values when subjected
478 to an ambient O₂ concentration of 10.5 vol.%, which were attributed to the
479 occurrence of incomplete combustion at low O₂ concentration. It is noted that
480 previous studies have indicated that the combustion process of diesel is more
481 affected by reduced oxygen atmospheres, when compared with biodiesels, as
482 diesel does not contain fuel oxygen [39, 47]. In addition, the reaction rates of
483 the aromatic compounds that are available in the diesel, but not biodiesels, have
484 been reported to be sensitive towards oxygen reduction, which can subsequently
485 reduce the reactivity of the diesel fuel. All these would contribute towards the
486 varying NPP trends of the biodiesel and diesel fuels across the varied ambient O₂
487 conditions. Further investigation, however, is required to elucidate the current
488 results.

489 3.2. Soot temperature and soot KL factor

490 As is noted in the previous section and other studies [14, 48], the natural
491 luminosity of a flame is a function of the local soot concentration and tempera-

492 ture. The temporal evolution of average soot temperature and soot KL factor of
493 the flames, under varied ambient O_2 conditions (10, 15 and 21 vol.%), but at a
494 fixed injection pressure of 100 MPa, are therefore plotted to provide additional
495 insights. It should be noted that the two-color pyrometry technique is a line-of-
496 sight method and therefore, its accuracy is susceptible to the spatial gradients
497 in soot properties within the measurement volume, along the optical path. The
498 technique has been previously reported to have a temperature uncertainty range
499 of ~ 150 K, and is only reliable for the qualitative soot concentration comparison
500 [28]. Typical instantaneous soot temperature and KL factor fields at three time
501 instances, which were generated using the two-color pyrometry methodology for
502 the WCO biodiesel jet flame at the condition of 15 vol.% O_2 concentration, are
503 presented in Fig. 7. In these images, the injector tip is located at the top of
504 the images, and the time instances were selected to cover the period from the
505 initial to the quasi-steady stage of the spray combustion event. The average
506 soot temperature and soot KL factor values for all fuels, which correspond to
507 the average values of all pixels in the two-color image frame at each time instant,
508 are presented in Fig. 8. The average soot temperature and KL factor values are
509 presented from the start of soot emergence until the end of soot detection. From
510 the figure, it can be seen that the measured soot temperature and soot KL fac-
511 tors increase rapidly to reach quasi-steady values during the stabilized periods
512 of their injections. The trends of the plots during the quasi-steady periods are
513 found to be similar for the fuels, with the highest and lowest soot temperature
514 values observed for the 21 vol.% and 10 vol.% O_2 cases, respectively. The soot
515 KL factor of the fuels is also observed to display similar characteristics, such
516 that the highest and lowest values are found for the 21 vol.% and 10 vol.% cases,
517 respectively. It is noted that the comparatively lower soot temperature and soot
518 KL factor values of the flames at 10 vol.% case imply that the low O_2 condition
519 has affected the combustion processes of the flames [43]. It is noted that the soot
520 temperature and KL factor values measured are sensitive to the experimental
521 conditions and settings used. It is therefore not possible to directly compare
522 the current measurement with previous studies. The observed values, however,

523 are broadly consistent to those measured for WCO flames flame with similar
524 arrangement, as measured by Zhang *et al.* [22].

525 From Fig. 8, it can be observed that the overall characteristics of the average
526 soot temperature and soot KL factor profiles of the fuels are comparable. For
527 both the average soot temperature and KL factor plots, it can be seen that the
528 time that is taken for the flames to reach their maximum quasi-steady values
529 increases with decreasing ambient O₂ concentration. Similarly, the combustion
530 periods of the flames are also observed to increase with decreasing O₂ level,
531 which is to be expected, as more mixing would be required for complete com-
532 bustion to occur under diluted conditions [49]. It is noted, however, that the
533 combustion duration of the diesel flame is similar to that of the biodiesel flames
534 for the 21 vol.% case, but becomes comparatively longer at lower ambient O₂
535 concentration. The average soot temperature and soot KL factor values of the
536 flames increase after the quasi-steady periods (*i.e.*, after the end of injection
537 at ~ 2.68 ms aSOI). It is noted that recent work [50, 51] have demonstrated
538 that after the end of injection, the entrainment of ambient gases into the jet
539 can increase significantly to change the soot formation and oxidation processes.
540 scale=0.8 soot temperature and soot KL factor could therefore be potentially
541 attributed to the occurrence of enhanced soot oxidation in the low soot tem-
542 perature and KL zones, leaving the average temperature and soot KL factor to
543 be dominated by the soot signal from the local high temperature and soot con-
544 centration regions [29]. Focusing on quasi-steady periods for the flames, prior
545 to the end of injection, it can be seen that a decrease in the ambient O₂ con-
546 dition did not appear to significantly impact on the average soot temperature
547 and soot KL factor values of the flames, until it was reduced to 10 vol.% O₂.
548 This, combined with the reduced NPP values observed for the fuels at 10 vol.%
549 O₂ concentration in Fig. 6, indicate that the combustion processes of the flames
550 were significantly affected for the 10 vol.% O₂ cases.

551 From the figure, it can be seen that the average soot temperature and KL
552 factor values of the biodiesel flames are comparable (within the uncertainties of
553 the technique) at all ambient conditions. The average soot temperature and KL

554 factor values of the diesel, on the other hand, are observed to be similar to the
555 biodiesels for the 21 and 15 vol.% cases, but the average soot KL factor of the
556 diesel flame become lower than that of the biodiesels at 10 vol.% O₂ concentra-
557 tion. To provide further insights, the trends of the average soot KL factors are
558 also compared with the corresponding integrated soot KL factors over the soot
559 areas of the flames, whihc are shown in Fig. 9. From Fig 9, the integrated soot
560 KL factor value of the diesel flame, which is representative of the total soot [22],
561 is found to be higher than the biodiesels at 21 vol.% O₂ concentration. The dif-
562 ferences in the integrated soot KL factor values between the diesel and biodiesel
563 flames, however, reduce with decreasing ambient O₂ concentration, such that
564 the diesel flame is observed to have the lowest integrated soot KL factor value
565 for at 10 vol.% O₂ concentration. Taken together, the results indicate that the
566 soot area of the diesel flame is largest amongst the flames at 21 vol.% O₂, but
567 the differences gradually reduce with decreasing O₂ concentration. It can also
568 be deduced that the biodiesel flames displayed lesser variation in soot areas with
569 changing ambient O₂ concentration, which is consistent with the previous find-
570 ing that the combustion process of biodiesel is less affected by reduced oxygen
571 atmospheres when compared with diesel.

572 **4. Conclusion**

573 To study the combustion characteristics of biodiesel under compression-
574 ignition experiments, a set of optical diagnostics have been applied to waste
575 cooking oil (WCO) and canola oil (CAO) biodiesels. Diesel was used as ref-
576 erence fuel for comparative studies with the biodiesels. Experiments were per-
577 formed in an optically-accessible constant-volume combustion chamber (CVCC)
578 at ambient conditions that are representative of compression-ignition engine
579 (19.4 kg/m³ ambient density, 1100 K bulk temperature, 10-21 vol.% ambient
580 O₂ concentrations, 70-130 MPa injection pressures). The liquid lengths of the
581 biodiesels were also measured, and were found to be longer than that of diesel,
582 under the test conditions of this work. The liquid lengths of all fuels were

583 measured to be shorter than the corresponding lift-off lengths of the flames,
584 therefore implying that significant interaction between the spray and combus-
585 tion processes of the fuels was not at play. The results revealed that for the
586 investigated experimental conditions, the lift-off lengths were affected by both
587 the chemical and physical properties of the fuels. In addition, a general trend
588 was also observed for the soot content of the flames, such that a larger differ-
589 ence between the lift-off length and the first-luminosity distance, was found to
590 correlate with lesser downstream soot production. Assessing the sooting and
591 combustion characteristics of the biodiesel and diesel flames across the varied
592 ambient O₂ concentration, the estimated soot content of the biodiesel flames
593 were found to be comparable with each other, but lower than that observed for
594 diesel. The estimated soot contents of the biodiesel and diesel flames were also
595 observed to reach their maxima at 15 and 21 vol.% O₂ concentration, respec-
596 tively. The normalized peak pressure (NPP) values, and therefore, the thermal
597 efficiencies of the biodiesels, on the other hand, were measured to be higher than
598 that of diesel. From the two-color measurement results, it can be seen a decrease
599 in the ambient O₂ condition did not appear to significantly impact on the av-
600 erage soot temperature and soot KL factor values of the flames, until it was
601 reduced to 10 vol.% O₂. The combustion durations of the biodiesel flames were
602 observed to be similar to that of diesel at 21 vol.%, but became comparatively
603 shorter once the ambient O₂ concentration was reduced to 15 vol.%. The com-
604 bustion durations of all flames were found to be prolonged significantly, when
605 the ambient O₂ concentration was further reduced to 10 vol.%. The change in
606 the soot areas of the biodiesel flames with varied O₂ concentration atmosphere
607 were found to be less significant than that of the diesel flame.

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768 formation of impinging diesel spray, *Applied Energy* 88 (2011) 1620–1628.

Fuels	Combustion ambient test conditions	Injection specifications	Reference
Diesel, JP-8, world average Jet A blend, Fischer-Tropsch fuel, coal-derived fuel, surrogate fuel	6-6.7 MPa 900-1000 K, 15% O ₂ concentration	Single-hole 0.09 mm hole diameter 150 MPa injection pressure	[13]
Butanol, soybean biodiesel	3.3-4.2 MPa, 800-1000 K, 21% O ₂ concentration,	Six hole, 0.145 mm hole diameter, 134 MPa injection pressure	[43]
Diesel, soybean biodiesel, ethanol, n-butanol	2.9-4.2 MPa, 700-1000 K, 21% O ₂ concentration,	Six hole, 0.145 mm hole diameter, 134 MPa injection pressure	[46]
Diesel, soybean biodiesel	2 MPa, 873 K, 9.6-20.8% O ₂ concentration	Six hole, 0.1 mm hole diameter, 100 MPa injection pressure	[39]
Diesel, soy methyl ester	6-6.7 MPa, 900-1000 K, 15% O ₂ concentration,	Single-hole, 0.9 mm hole diameter, 150 MPa injection pressure	[32]
JIS# Diesel, palm oil biodiesel, cooking oil biodiesel	4 MPa, 885 K, 21% O ₂ concentration	Single-hole, 0.16 mm hole diameter 100-300 MPa injection pressure	[14, 52]
Diesel, soybean biodiesel	2 MPa, 800-1200 K, 15-21% O ₂ concentration	Six hole, 0.145 mm hole diameter, 100 MPa injection pressure	[16]
Diesel, waste cooking oil biodiesel	3.5-5.3 MPa, 800-1200 K, 21% O ₂ concentration	Six hole, 0.152 mm hole diameter, 100 MPa injection pressure	[20, 22]

Table 1: Summary of selected experimental conditions involving investigations of biodiesels in combustion chambers cited in this work.

Test conditions	
Oxygen level (vol.%)	0 (non-reacting), 10, 15, 21 (reacting)
Ambient density (kg/m ³)	19.5
Pressure (MPa)	6
Bulk temperature at injection (K)	1100
Injection conditions	
Nozzle diameter (μm)	105
Injection pressure (MPa)	70, 100, 130
Injection duration (ms)	2.68

Table 2: Summary of test and injection conditions.

Diagnostic techniques	OH chemiluminescence imaging	High-speed flame luminosity imaging	Diffused back illumination
Camera	Intensified CCD camera (Andor iStar)	High-speed CMOS camera (Photron SA5)	
Frame rate (frames per second)	-	15,000	
f-number	3.5	1.8	1.8
Pixel resolution (mm per pixel)	0.1	0.2	0.14

Table 3: Combustion and spray imaging setup.

Main fuel characteristics			
	WCO	CAO	D
Saponification number	193	188	-
Cetane number	53	59	51
Viscosity @ 293 K, (mm ² /s)	4.6	4.65	3.2
Oxygen content (wt%)	10.8	11	0
Lower HV (MJ/kg)	38.5	35.8	41.1
Stoichiometry air-fuel-ratio (AFR) by mass	12.7	12.5	14.5
Iodine number	95	94	-

Table 4: Main fuel characteristics of waste cooking oil (WCO), canola oil (CAO) based biodiesels and diesel (D).

Main FAMES (vol%)		
	WCO	CAO
Myristic, C14:0	0.79	-
Palmitic, C16:0	15.42	3.94
Stearic, C18:0	6.13	2.24
Oleic, C18:1	45.64	64.07
Linoleic, C18:2	28.63	18.93
Linolenic, C18:3	3.39	9.15
Eicosanoic, C20:0	-	0.45
Eicosenoic, C20:1	-	1.22

Table 5: Main fatty acid methyl esters (FAMES) of (in vol.%) the waste cooking oil (WCO) and canola oil (CAO) based biodiesels.

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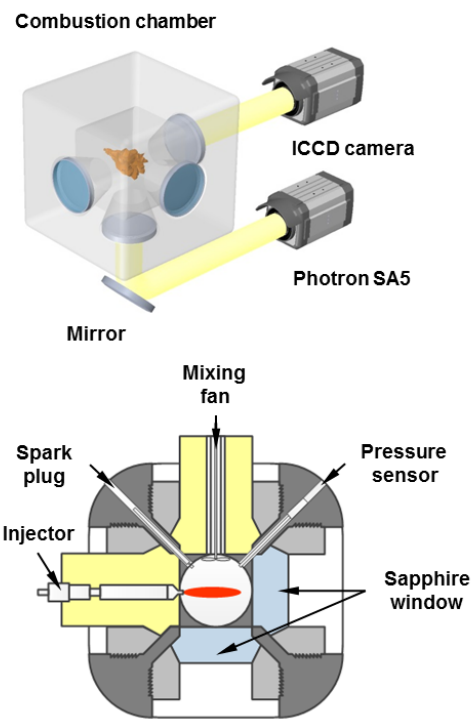


Figure 1: Schematic diagrams of the experimental arrangement used, including the combustion vessel, OH-chemiluminescence and high-speed natural luminosity imaging setups (top) and the cross-sectional view of the chamber (bottom).

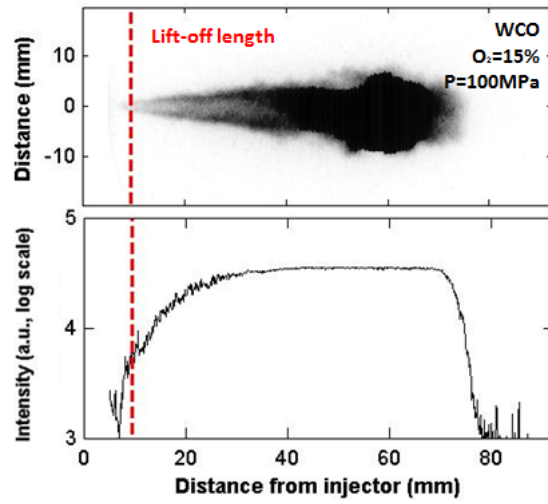


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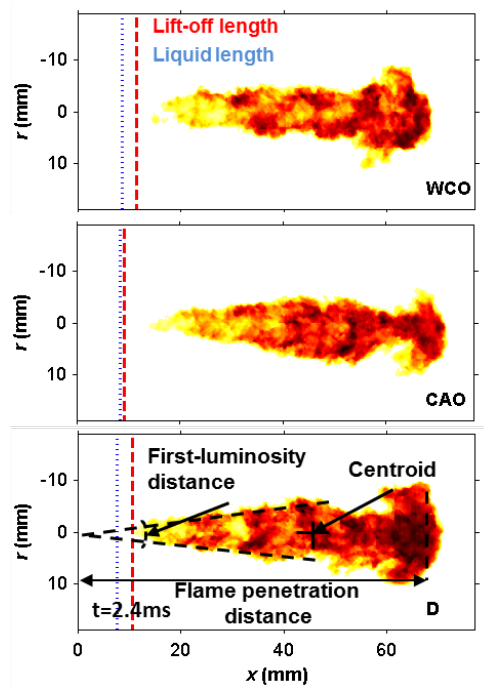


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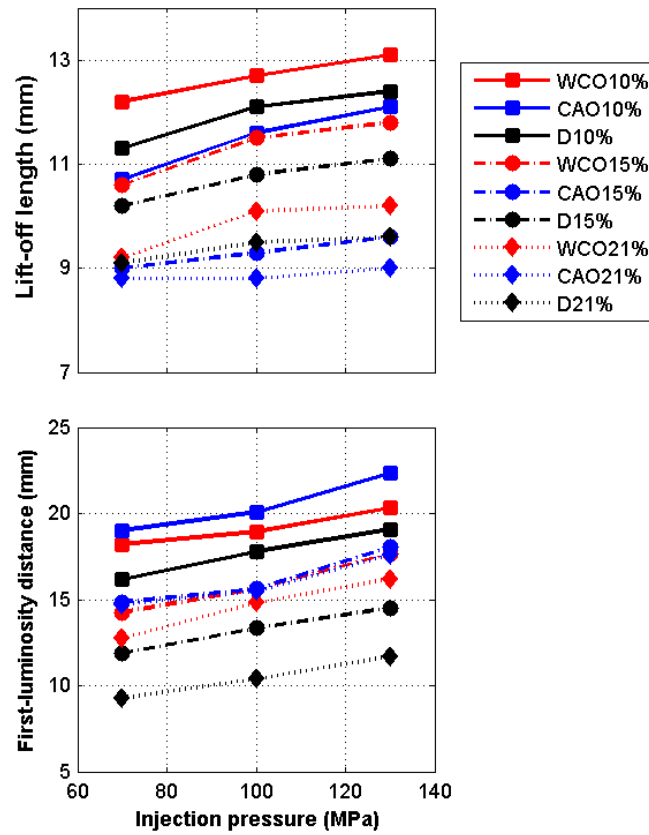


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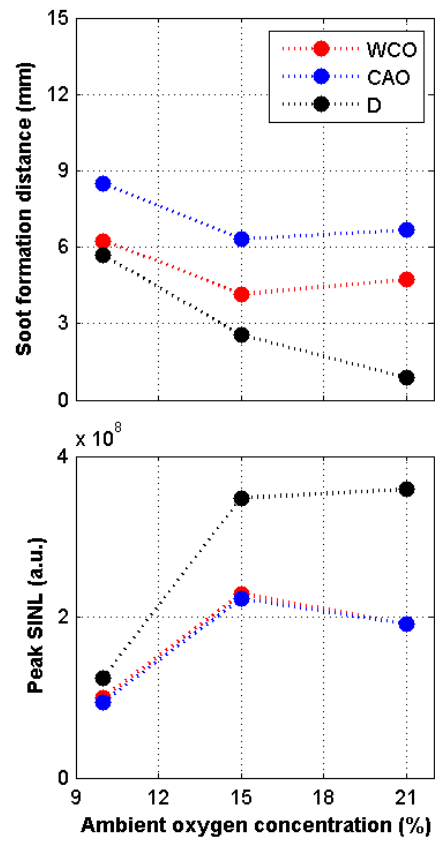


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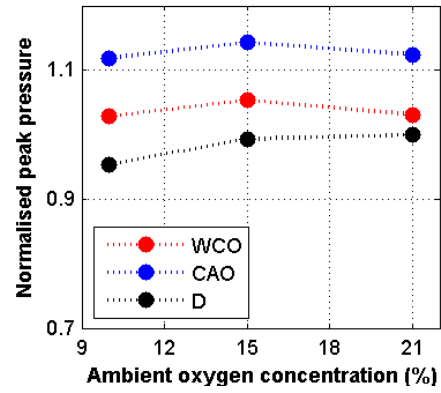


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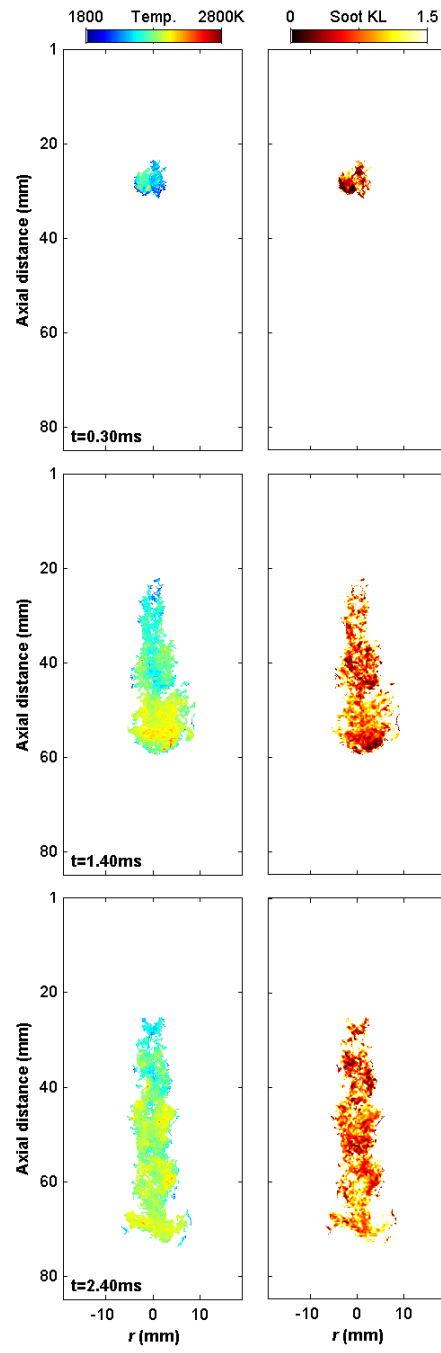


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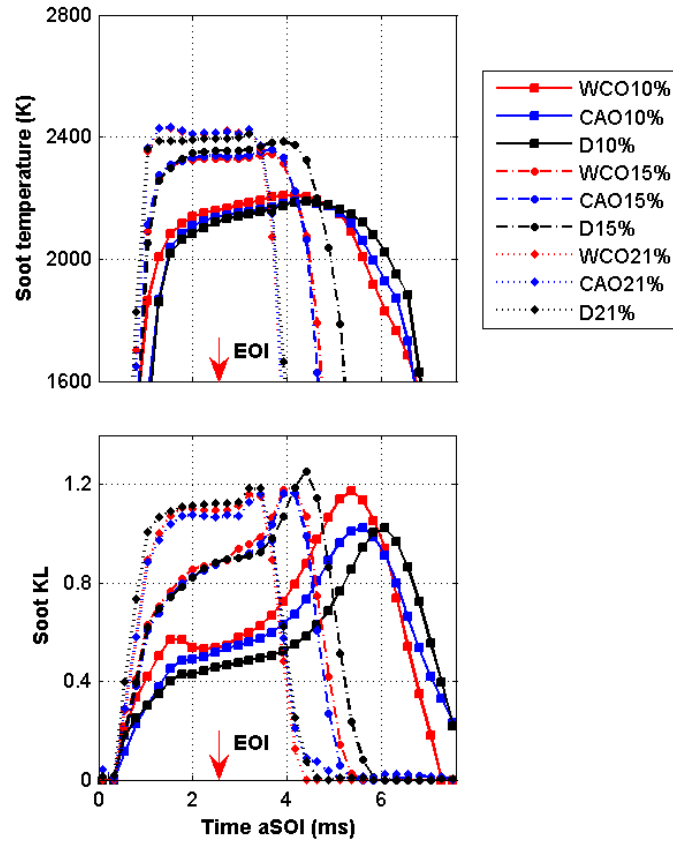


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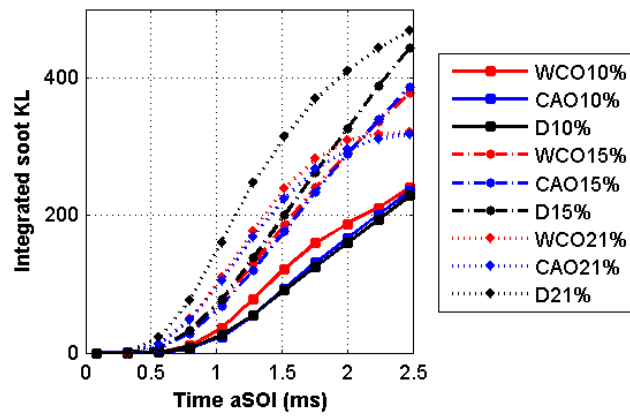


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