

19 **Combustion behaviour of relatively large pulverised biomass particles at**
20 **rapid heating rates**

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29 **Abstract**

30 A pulverised solid fuel particle in a hot gas stream appears to have different characteristic
31 behaviours at several stages, including heat-up, release of volatile matter, gas phase and solid
32 combustion. The characteristics of these stages may vary distinctly depending on
33 devolatilisation rate, the particle temperature history and its chemical and physical properties.
34 Biomass particles manifest different combustion behaviour from that of burning coal particles
35 under the same combustion conditions because they contain more volatiles (less fixed
36 carbon), and they have a relatively lower particle density due to their fibrous structure. This
37 paper presents an experimental study of burning behaviour of different types of biomass
38 particles (torrefied wood, coffee waste and sewage sludge). The main experimental
39 parameters—gas temperatures of 1,090 K and 1,340 K, and O₂ concentrations ranging from
40 10% to 40%—were employed to investigate the burning of biomass through a direct-
41 observation approach using a high-speed photography technique at 7,000 frames/s. In the
42 case of firing/co-firing, biomass particles must be larger than the coal particles in order to

43 achieve an equivalent thermal balance due to the higher energy density of coal. Therefore, the
44 selected biomass samples were in the size range from 150–215 μm to 425–500 μm . The
45 experimental setup has a cross-flow configuration for particle injection in order to enhance
46 interaction between the particle and the two different streams—a cold carrier gas at 298 K,
47 and upward-flowing post-combustion gases. It is believed that the employed experimental
48 conditions are similar to those in a realistic furnace with a rapid heating rate of 10^5 K/s. The
49 experimentally significant results, including the effective radii of the volatile flames, degrees
50 of flame intensity and the maximum size of a particle are important for validation of models
51 of single biomass particle combustion.

52 **Introduction**

53 Single solid fuel particles have been researched for several decades in an attempt to gain a
54 fundamental understanding of their combustion behaviour. When a particle is exposed to a
55 hot gas stream and a rapid heating rate, it immediately undergoes a rapid temperature increase
56 prior to the release of volatile matter with its initial ignition [1–4]. These thermal
57 decomposition and combustion stages are determined by environmental conditions and the
58 physical structure of the solid particle in relation to its chemical composition [5–7]. In
59 particular, pulverised particles are expected to burn rapidly in industrial furnaces [8, 9]
60 however, particles of this size tend to have inconsistent flame structures and at low heating
61 rates. Observation of these burning particles under rapid heating to high temperature, along
62 with their flame structures and time durations of devolatilisation and combustion, would not
63 be a simple task. Biomass has a great potential as a CO_2 -neutral, low-emission energy source
64 of heat and electricity through various applications such as combustion and gasification [10,
65 11]. However, biomass particles have high compositional variability, a fibrous nature and an
66 irregular shape. They also contain highly volatile matter and are of low particle density. These

67 physical and chemical differences from coal result in dissimilar thermal conversion and
68 characteristic combustion behaviour during the early stages; compared with coal, biomass
69 combustion can offer a faster reaction rate, non-uniform gas-evolution profile and longer
70 volatile-combustion duration [12–14]. However, these kinetic behaviours with flame
71 structures may be inconsistent for different types of biomass and solid waste fuel due to large
72 variation in their physical structures and chemical compositions. The different volatile flames
73 of each biomass type influence the operating efficiency of their applications because of
74 diverse quantities of radiant energy, dominated by flame size and luminosity.

75 The size of a biomass particle in pulverised combustion is expected to be larger than that of
76 a coal particle because of low particle density and faster devolatilisation rate and, the biomass
77 particles will not be pulverised to the same size as coal particle due to different milling
78 behaviour and non-economic pre-processing [11, 15]. The increase of particle size is likely to
79 enhance the ignition delay and quantity of partly unburned residue compared with a small
80 volume of coal [16, 17] in an identical environment. Combustion behaviour is attributed to
81 particle-size distribution and enhanced oxygen concentration as well as combustion
82 temperature. In practical applications, solid particles are generally entrained perpendicularly
83 into a hot gas stream along with a carrier gas at a rapid heating rate of 10^4 – 10^5 K/s [18]. This
84 experiment examines biomass particles under analogous operating conditions, so as to
85 explicitly identify the temporal variations in the burning behaviour of each particle. This is
86 capable of accurately describing combustion processes while gaining a fundamental
87 understanding of each burning biomass particle.

88 A number of previous experimental observations of particle combustion have been performed
89 using different approaches to investigate ignition delay, kinetics and burnout time [19–22].
90 McLean et al. [19] introduced direct observation of an early stage of combustion, capturing

91 the burning particle at high gas temperature. This result gave basic evidence of the physical
92 phenomena associated with pulverised coal combustion, such as the development of a
93 radiative volatile flame. Khatami et al. [20] observed combustion behaviour in a quiescent
94 environment and found that biomass has a spherical flame envelope with low luminosity and
95 that the durations of volatile and char combustion decreased with increase in oxygen
96 concentration due to the higher temperatures of the burning particles. Yin et al. [21] also
97 captured informative images of spherical and cylindrical biomass particles to investigate their
98 characteristic behaviour. Although these experimental observations have provided an
99 acceptable fundamental understanding of fuel particles, the combustion of a single biomass
100 particle has yet to be observed, which would provide an outline of the processes of volatile
101 combustion and burnout as well as overlapping combustion. Thus, there is a lack of insight
102 into the combustion behaviour of biomass in pulverised combustion due to the unclear
103 physical structures of the volatile flame. In addition, previous research has rarely focused on
104 the sequential combustion stages of pulverised biomass with quantitative analysis. In the
105 present study, the determination of the optimal burning conditions of pulverised biomass
106 particles is first attempted at a rapid heating rate under a cross-flow configuration. This
107 layout enables a distinct description of the burning particle that is displaced along with the
108 development of its volatile flame structure as a function of time. In addition, the maximum
109 size of a particle that can be burned completely without dropping to the bottom of the reactor
110 is determined from the interaction between the particle and the two perpendicular streams.
111 Consequently, an explanation of burning particles at these environmental conditions with
112 measured values could support development of a mathematical model of a single biomass
113 particle.

114

115 **Experimental Setup**

116 *Particle samples and separation*

117 The biomass samples prepared for the experimental work include torrefied wood, coffee
118 waste and sewage sludge, as shown in Fig. 1. Torrefied wood was thermally treated in a gas
119 temperature environment of 573 K. The thermal treatment converts oxygen to CO and CO₂
120 while increasing energy density and hydrophobicity. This torrefied particle contains less
121 volatile matter and relatively more fixed carbon than the raw particle. Coffee waste is the
122 residue of an extracted coffee bean, which was also thermally treated originally, whereas
123 sewage sludge is one of the most widely used waste materials and has the highest ratio of
124 volatile matter to fixed carbon. However, the sludge also has the highest ash content among
125 these particles, which is largely the cause of its low energy density and burnout time. The
126 material analysis of these particles is shown in Table 1 and, the proximate and ultimate
127 analyses are reported on an 'as received' and 'dry, ash-free' basis, respectively. These analysis
128 data of four solid fuel particles were obtained from TGA-701 thermogravimeter, TruSpec
129 elemental analyser and AC600 calorimeter at Energy & Environment Research Centre,
130 KAIST. From the particle bulk density, the approximate energy density of subbituminous coal
131 particles (19,581 MJ/m³) is almost double that of the three biomass particles. Torrefied wood,
132 coffee waste and sewage sludge particles have 11,890, 13,741 and 10,112 MJ/m³ of energy
133 density, respectively.



134

135 Figure 1. Pellets and single particles of the three biomass materials.

136

137 Table 1. Chemical compositions of biomass

| Sample | Proximate analysis (wt. % ar) ¹ | | | | Ultimate analysis (wt. % daf) ² | | | | | LHV ^{3&1} (MJ/kg) | Particle bulk density (g/cm ³) | Approx. energy density (MJ/m ³) |
|----------------------------|---|------|------|------|---|-----|------|-----|-----|-----------------------------------|--|--|
| | V.M | F.C | Ash | M | C | H | O | N | S | | | |
| Torrefied wood | 69.9 | 22.5 | 0.95 | 6.65 | 51.5 | 5.1 | 37.2 | 0.2 | 0 | 20.50 | 0.58 | 11,890 |
| Coffee waste | 72.9 | 11.1 | 5.8 | 10.2 | 49.4 | 5.7 | 35.3 | 2.6 | 0.4 | 20.82 | 0.66 | 13,741 |
| Sewage sludge | 61.2 | 7.89 | 25.0 | 5.91 | 38.6 | 5.9 | 20.6 | 0.9 | 0.1 | 17.14 | 0.59 | 10,112 |
| Subbituminous coal (Adaro) | 42.0 | 46.1 | 1.5 | 10.4 | 64.4 | 4.7 | 18.1 | 0.9 | 0.1 | 25.43 | 0.77 | 19,581 |

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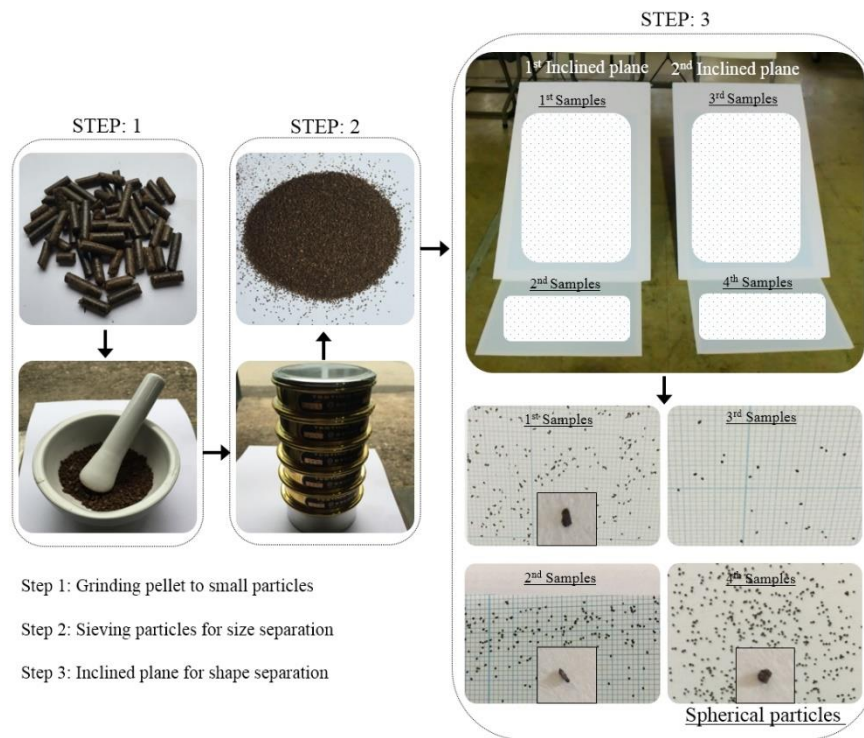
¹as received ²dry, ash free ³lower heating value

139 Separation of biomass particles is approached cautiously because of their fibrous and
 140 tenacious nature, using several methods, including pulverisation (Step 1) and sieving (Step 2)
 141 processes shown in Fig. 2. Step 1 is a preliminary process to pulverise pellets into small
 142 particles. Step 2 separates the sample particles into 7 different sieve sizes: 150, 215, 250, 300,
 143 355, 425 and 500 μm . An inclined plane (Step 3) is used to separate different shapes of

144 particles based on their translational and rotational motions. A1 uncoated paper on high-
145 density fibreboard was used for the shape separation process because matte and gloss coated
146 papers offered too low frictional resistance on the inclined planes. Sand paper could be a
147 potential option, but offered too much frictional resistance, holding dropped particles at the
148 top or midway down the inclined plane when this paper was used. The angle of the inclined
149 plane and drop distance of the particles were also significant parameters in this step. The
150 optimum angles were determined by a number of trials; a first plane has 45° and second one,
151 60°. Finally, high humidity and unclean surfaces must be avoided during the process.

152

153 In this experiment, irregular shapes, such as those which are extremely flat and cylindrical
154 with high aspect ratios, are not suitable for the cross-flow configuration. The undefined drag
155 coefficients obtained from these irregular shapes can lead to random trajectories with non-
156 uniform particle motion, resulting in non-quantitative analysis of burning particles. To reduce
157 this limitation, an inclined plane is used to separate particles based on their translational and
158 rotational motions, so as to collect particles of moderate shape, as shown in Fig. 2. Particles
159 with flat shapes stop at the upper zone of the first slope as they have the highest friction rate,
160 and cylindrical and spherical particles collect at the lower zone. The slope is then adjusted to
161 have different angles and the separation is run again on only the particles at the bottom, until
162 only spherical particles remain due to their lower inertial force. Using this approach, the
163 groups of spherical, cylindrical and flat shapes are prepared properly.



164

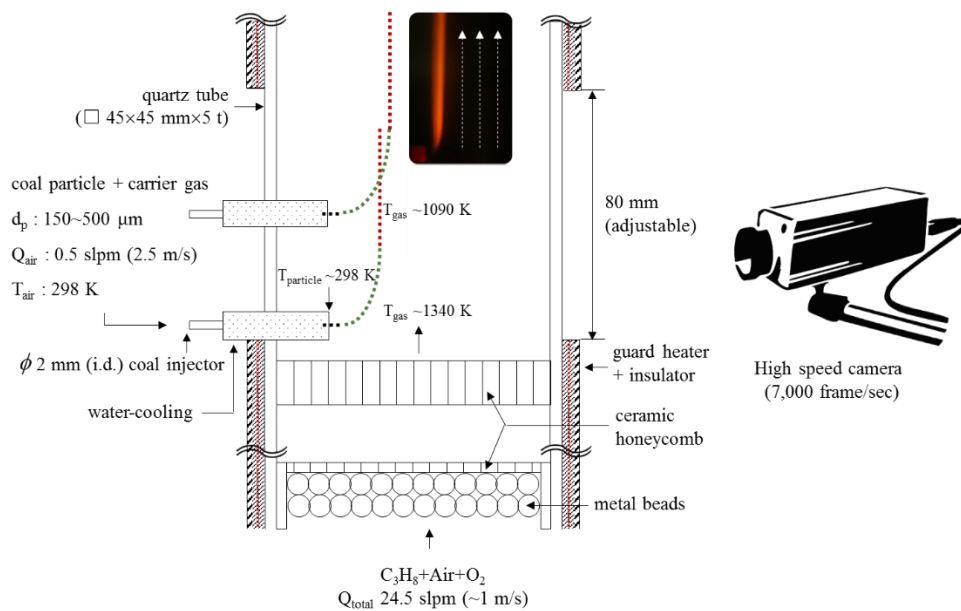
165 Figure 2. Pre-process for particle separation by means of sieving and inclined plane

166 methods.

167 *Single particle reactor coupled with cross-jet injection*

168 The laboratory-scale entrained single particle reactor shown in Fig. 3 has its gas
 169 temperature, flow velocity and oxygen concentration controlled by the post-combustion gas,
 170 guard heater and water-cooled injector. In this environment, a particle is expected to be
 171 exposed to a uniform flow at high temperature to reduce experimental uncertainty from the
 172 flow straightener. The reactor is made of a rectangular quartz cell of 45 mm × 45 mm × 500
 173 mm to minimise the refractive index for observation of the combusting particle. The water-
 174 cooled injector is installed to maintain the initial particle temperature of 298 K and the guard
 175 heater forms the external wall of the cell to prevent large losses through heat transfer. The
 176 single particle feeder is based on a fluidised bed of solid particles that are dropped from a
 177 scientific syringe injector. Back pressure occurs in the double tubes, and the particles in the

178 fluidised bed are dropped into the particle injector. The measured number of particles inserted
 179 into a hot gas stream was 20-30 particles/min. The flow rate of the carrier gas in the particle
 180 injector is determined to be 0.5 L/min to lift particles over 150 μm . The upward and vertical
 181 flow velocities are constant for all particles, to investigate comparable combustion time and
 182 particle displacement.

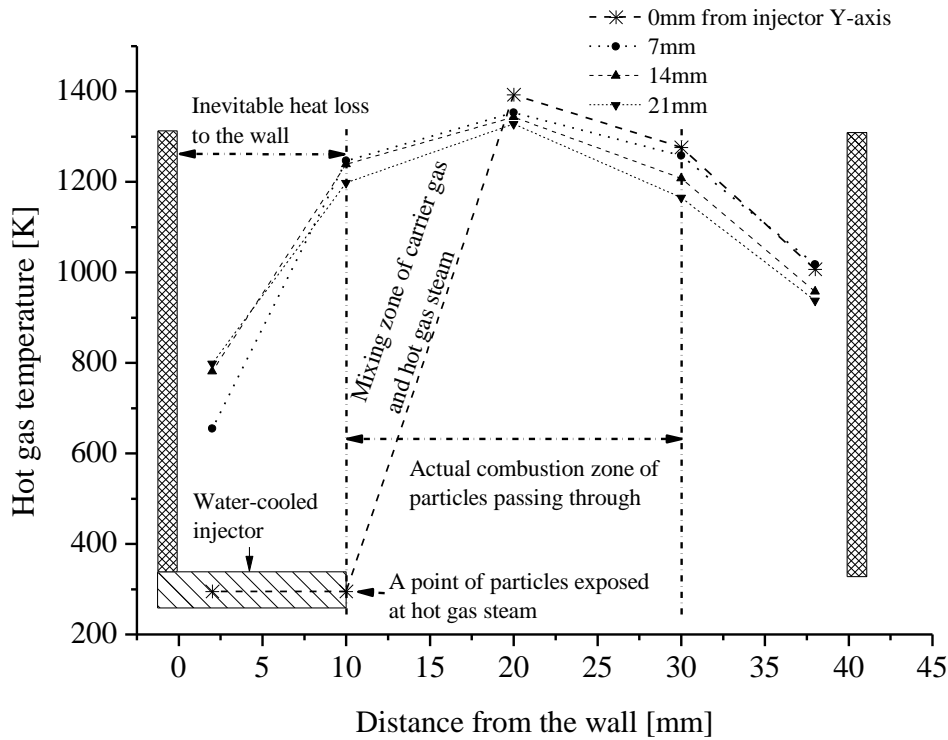


183
 184 Figure 3. Schematic diagram of the experimental setup at 1,090 K and 1,340 K of gas
 185 temperatures.

186
 187 *Operating conditions and particle size*

188 The post-combustion gas produces a high temperature in the upward stream from the
 189 honeycomb. In the quartz cell, substantial heat loss is inevitable between the wall and a hot
 190 gas stream, in which a high temperature gradient appears and particles pass through. The
 191 environment gas temperature is calculated on the basis of the radiation loss of the probe of an
 192 R-type thermocouple [23]. The temperature profile is shown in Fig. 4. The high-mixing zone
 193 between the leftward-flowing cold carrier gas and the upward-flowing hot gas stream is

194 clearly shown to affect transient combustion behaviour. Particle temperature before injection
 195 remains at 298 K and reaches a certain high temperature as a particle passes through the hot
 196 gas stream. To avoid large heat loss, the position of the injector is located 10 mm from the
 197 wall.



198 Figure 4. Temperature profiles measured from the experiment.

199 The amount of oxygen is supplemented from 10.4 % to 40.1 % of the total product gases.
 200 After the combustion of C_3H_8 , flue gases such as N_2 (38.2–67.9 %), O_2 (10.4–40.1 %), H_2O
 201 (12.4 %) and CO_2 (9.3 %) are yielded. The group sizes of the solid particles are prepared in
 202 nominal size ranges of 150–215, 255–300, 355–400 and 400–500 μm to determine the largest
 203 size at which the particles are burned completely.

204 *Observation of the biomass particles*

205 A high-speed camera (Phantom V710) is used to observe burning particles at 5,000–7,000

206 frames per second with a micro-lens and back lighting. It is equipped with a complementary
207 metal oxide semiconductor (CMOS) image sensor of size 25.6 mm × 16.0 mm with a
208 resolution of 1,280 × 800 pixels, and is capable of recording the motions of particles. This
209 camera enables analysis of the combustion of a single particle in great detail. The observation
210 zone is extended by adjusting the micro-lens, as the prepared biomass particles are larger than
211 those in previous experiments [24, 25], and the calibration is carried out with a circle-scale
212 reticle. The size of a pixel is measured to be 58 μm and, a light-emitting diode (LED)
213 backlight at 5,600 K (colour temperature) is installed at the rear of the quartz cell. This
214 enables direct observation of a burning particle's ignition, flame structure and char
215 combustion.

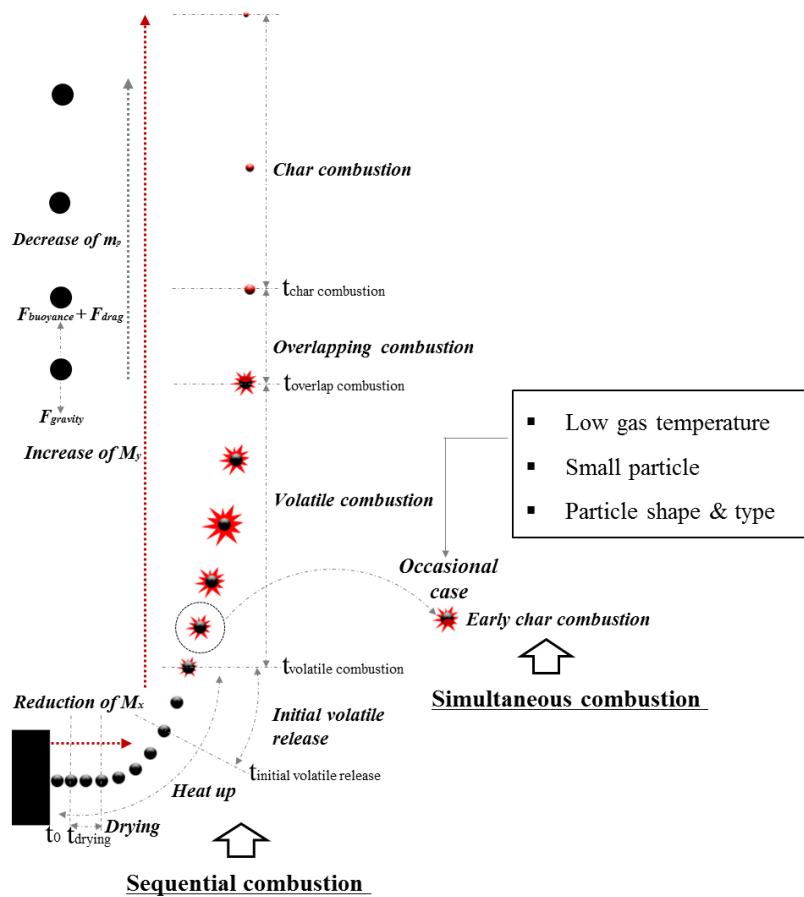
216 **Mechanism of biomass pyrolysis and sequential combustion regimes**

217 Biomass contains a variety of proportions of cellulose, hemicellulose and lignin, which
218 results in variations in the gas-eruption profile and mass reduction that depend on the type of
219 biomass [26]. It is widely accepted that hemicellulose is decomposed at a temperature range
220 of 493–588 K, cellulose at 588–673 K and lignin over a wide range of 433–1,173 K [27]. The
221 typical gases produced by biomass combustion include H₂, CO, CO₂, H₂O and CH₄ as light
222 hydrocarbons, as well as tar and char in primary pyrolysis. It is generally assumed that char
223 combustion starts successively after the volatile flame is dissipated as volatiles evolution is
224 fast enough to move the pyrolysis zone away from the particle surface [28, 29]. This is
225 referred to as a sequential combustion process and, a typical sequential process of a particle
226 in the experiment is shown in Fig. 5. Char combustion may occur in early gas-phase
227 combustion stage, if oxygen reaches the particle surface. This burning behaviour is called a
228 simultaneous combustion process [30, 31]. In simultaneous combustion, the volatile flame

229 does not lift off the surface and the flame possesses a thin structure. Overlapping combustion
 230 is attributed to low gas temperature, small particles and the specific type of particle [32]. In
 231 addition, the irregular shapes of the biomass particle and the volatility of the flame may
 232 contribute to overlapping combustion. Consequently, the physical characteristics of the
 233 volatile flame on biomass particles in the early combustion stage have a significant role in
 234 determining the combustion process.

235

236



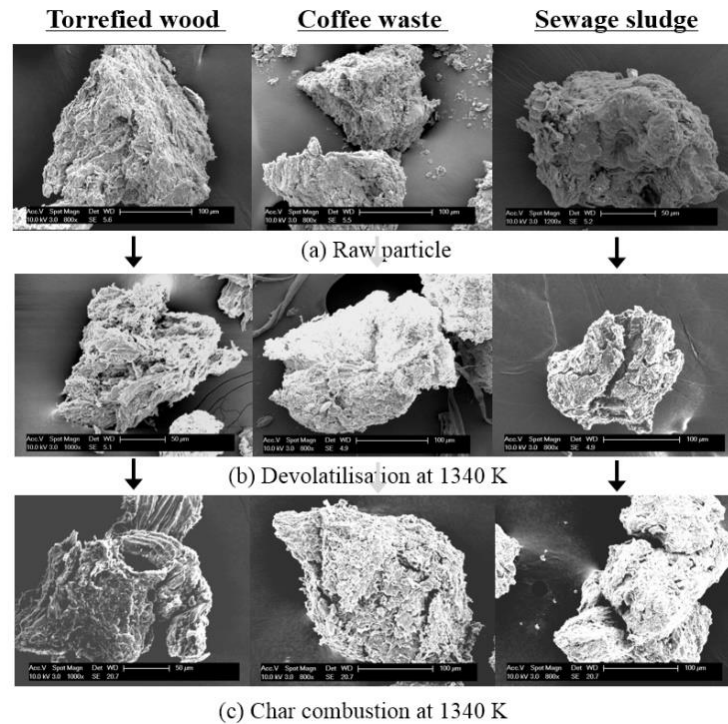
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238 Figure 5. Sequential regimes of a solid fuel particle during combustion.

239 **Results and Discussion**

240 *SEM images observed from biomass particles*

241 The scanning electron microscope (SEM) images of the three raw biomass types during
242 the combustion process are shown in Fig. 6. These particles were collected from the electric
243 furnace where the hot gas temperature is identical to that in the entrained flow reactor. The
244 burning particle after volatile ignition with the maximum size of flame was ejected from the
245 furnace and then quenched quickly on a cold plate. For the particle image of char
246 combustion, the particle was prepared after extinction of its flame. The NOVA 230 SEM
247 was employed at KAIST and, the backscattered images were captured under the following
248 parameters: Acc.V: 10.0 kV, Spot: 3.0 and Magn: 650-800 x. The external physical
249 structures, with their small pores and irregular shapes, are remarkably similar between raw
250 torrefied wood and coffee waste. However, torrefied wood contains a high proportion of
251 fibrous cellulose in its transformed structures due to thermal pre-treatment. Raw coffee
252 waste is a physically mixed formation with a number of small cumulative particles along
253 with porosity. Raw sewage sludge closely resembles a stiff clay soil with less porous
254 surfaces. During combustion processes, these raw biomass particles experience an increase
255 in porous surface with a certain swelling due to the melting and softening of the particles.
256 Torrefied wood shows a significant physical transformation at high environmental
257 temperature during the combustion process. Compared with coffee waste and torrefied
258 wood, sewage sludge has numerous cracks on its surface. The particles of all chars are
259 apparently deformed from the shapes of the raw particles due to release of volatile matter
260 and subsequent physical deformation, and the particle volume starts to decrease due to
261 surface oxidation.



262

263 Figure 6. SEM images of three different biomass types. In the upper row, raw particles of
 264 torrefied wood, coffee waste and sewage sludge are shown and the time series of the physical
 265 deformation of these three particles during combustion are presented along each column.

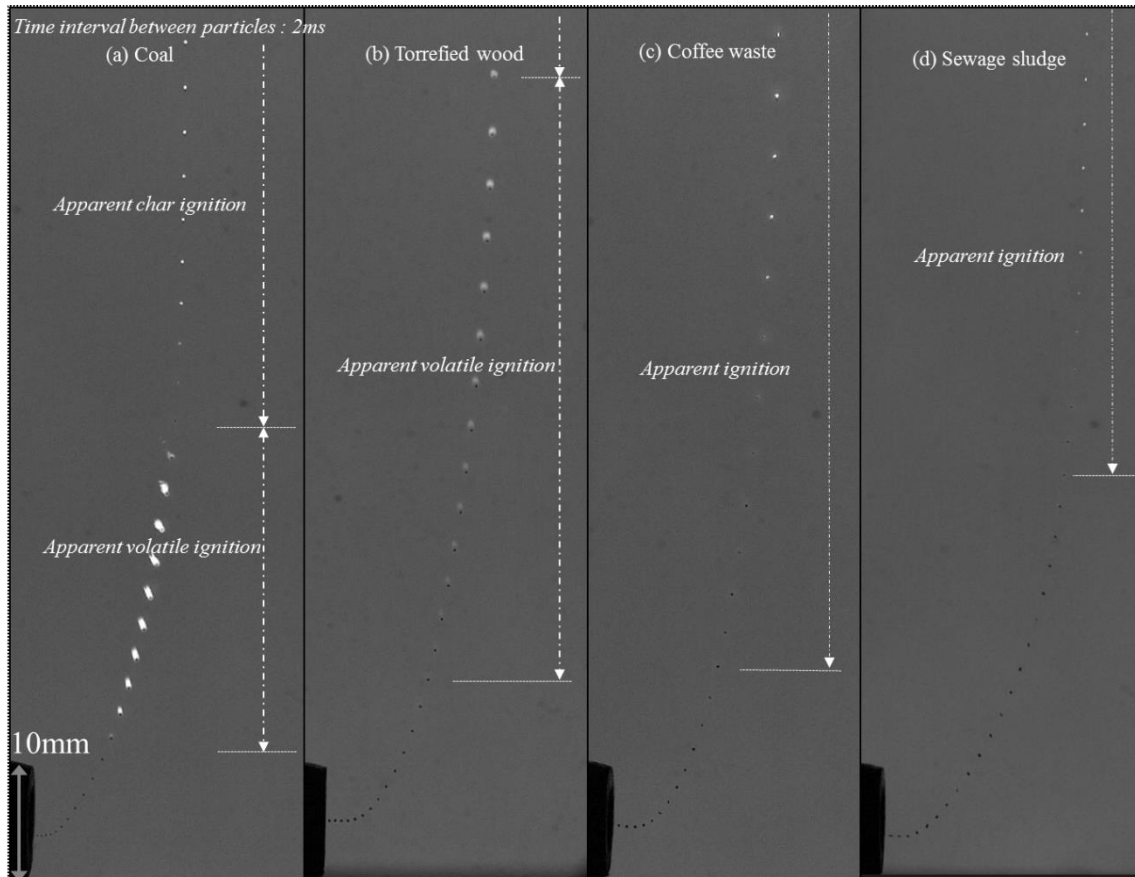
266 *Observation of the sequential combustion process and its characteristic flame structure*

267 For an explicit description of sequential combustion processes, a particle injected from the tip
 268 of the water-cooled injector is maintained at 298 K and the combustion is assumed to begin (t
 269 $= 0$) when it is exposed to a hot gas stream. The particle is first heated up with the initial
 270 drying process and then starts to release volatile matter, upon which it is immediately ignited
 271 after gas erupts from the solid particle. After these stages, a flame is observed away from the
 272 surface of the particle, forming a luminous boundary at which oxidation of the product gas
 273 takes place through a secondary particle reaction. The degree of luminosity is dominantly
 274 attributed to soot density in the flame and can be increased by using a hotter gas stream or
 275 particle temperature. The entire development of a volatile flame from the ignition of the gas

276 phase to its extinction is observed to take place for particle size of 150–215 μm at all oxygen
277 concentrations. Complete observation of the period for particle size 425–500 μm is limited
278 due to limitation of the visualisation field. Photographic images of three particles are
279 superimposed over a time interval of 2 ms in Fig. 7. This enables a distinct description of a
280 burning particle as it is displaced along with the development of its volatile flame structure.
281 These results help to predict the mass reduction of particles because of the cross-flow
282 configuration. In this layout, a particle starts drying very quickly in the mixing zone of the
283 horizontal and vertical streams, and then moves in the horizontal direction at high momentum
284 due to the cold carrier gas. After passing through the transient point, rotating particles go
285 upward because of the buoyancy and drag force acting on the particle overcoming gravity.

286 For sequential combustions at 21 % O_2 and 150–215 μm particle size, different combustion
287 behaviour in terms of residence time and displacement is seen for the four particle types. A
288 coal particle undergoes a sequential combustion process; this particle has a flame with high
289 soot content and experiences a shorter period of volatile combustion than do the three
290 biomass particles because of the presence of less volatile matter. The charred coal
291 immediately experiences oxidation after extinction of the homogenous combustion. On the
292 other hand, torrefied wood and coffee waste have volatile flames with different physical
293 structures from that of the coal particle. The relatively sooty flame for torrefied wood lasts 30
294 ms, whereas coffee waste has a very thin flame with duration of volatile combustion of
295 approximately 14 ms. The different durations can be attributed to interactions between
296 volatile matter content and soot formation. An increase in particle temperature leads to a
297 change in its structure because of a high mass-reduction rate. During the combustion process,
298 temperatures of the particle and the flame are much different. Timothy et al. [33] reported that
299 the surface temperature while the flame envelope exists is even lower than the temperature of

300 the flame. Finally, sewage sludge initially has an unclear volatile flame, which then forms a
301 thin, nearly transparent volatile flame at an early stage due to its low carbon content.



302
303 Figure 7. Sequential combustion process including overlapping and char combustion for
304 155–215 μm particles of four different samples at 21 % oxygen concentration and 1340 K:
305 (a) sub-bituminous coal, (b) torrefied wood, (c) coffee waste, and (d) sewage sludge.

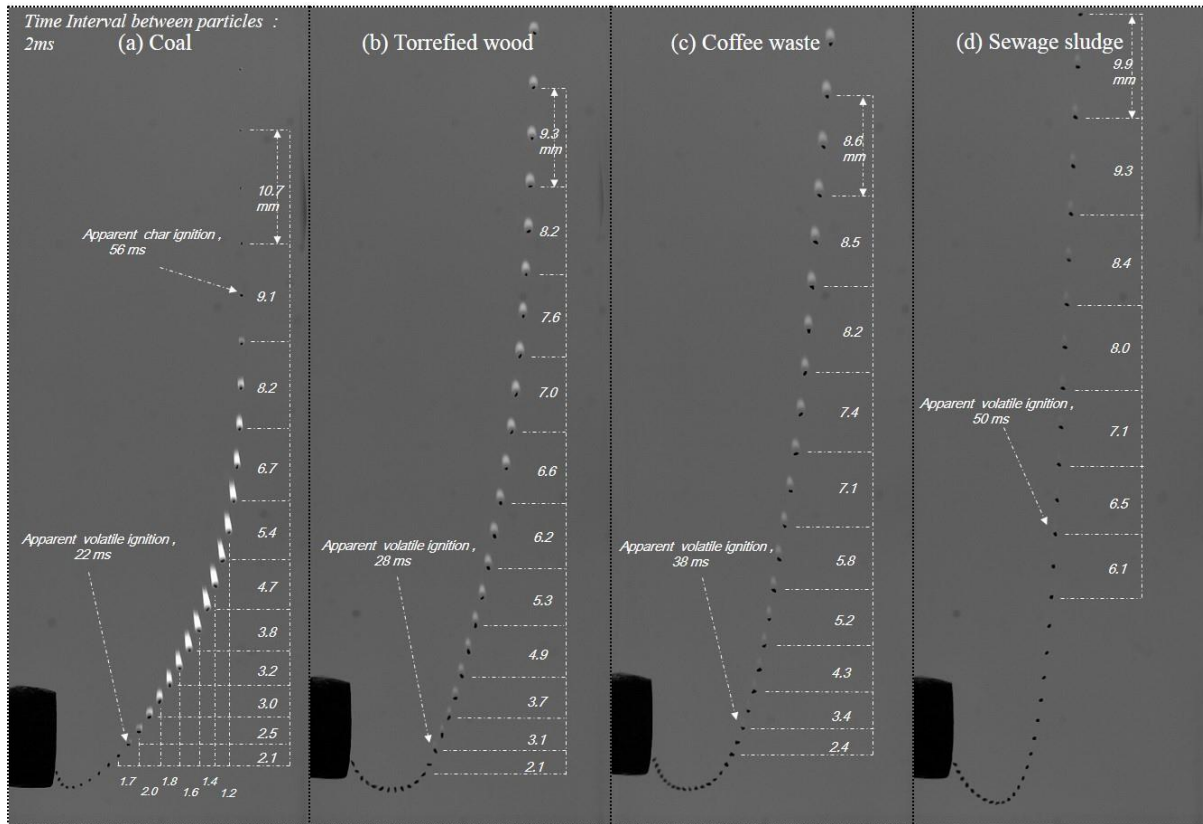
306 To provide a comparative analysis of sooty flames for particles with different physical
307 structures, the superimposed images of four solid particles at 300–355 μm and 10 % O_2 are
308 captured at identical conditions shown in Fig. 8. In general, biomass has an early initial
309 release of volatile matter compared with coal at a low heating rate, such as in a
310 thermogravimetric analyser. Sub-bituminous coal ignites a bit earlier compared with the three
311 biomass particles in the same particle size group. Different onset times of ignition between

312 biomass particles are also observed and can be attributed to their compositions and physical
313 structures.

314 The flames of burning biomass particles are relatively smaller and less sooty than those of
315 coal due to the relative soot formations from reactions with tar at high temperature. It is
316 accepted that this physical configuration is dominantly determined by complicated
317 interactions between volatile matter, carbon content, particle density and different rates of
318 devolatilisation. The three biomass particles have less bulk density, compared to the coal
319 particle density (0.77g/cm^3); the densities of torrefied wood, coffee waste and sewage sludge
320 are 0.72, 0.86 and 0.76 times that of coal, respectively. This low density of biomass particles
321 may result a shorter gas-phase combustion. However, these biomass particles have higher
322 mass volume fraction of volatile matter to coal, and thus, the duration of gas-phase
323 combustion for biomass is still longer than that of coal; from bulk density measurements,
324 torrefied wood, coffee waste and sewage sludge have 1.9, 2.3 and 1.7 g in equal volumes,
325 while coal still has the lowest mass of volatile content at 1.6 g although it has a higher bulk
326 density. The calculation measured total mass of torrefied wood (2.71 g), coffee waste (3.20 g)
327 and sewage sludge (2.84 g) in 4.82 cm^3 and the mass fraction of volatile matter of each
328 particle type is given in Table 1. In addition, the mass of fixed carbon of torrefied wood,
329 coffee waste and sewage sludge is 0.6, 0.4 and 0.2 g, respectively, while coal has 1.7 g; this
330 leads to the differing char combustion behaviour between biomass and coal particles. That is,
331 volatile matter content is a major factor in determining combustion time. Under direct
332 observation, an elongated flame is detected on all particles at 10 % oxygen concentration.
333 The shape of the flame structures is due to the low diffusion rate of oxygen with the effect of
334 buoyancy and it is apparent that coal has the flame with the highest aspect ratio. At this
335 particle size group, biomass particles have even higher elongated flames than that at 150–215

336 μm because a larger volume particle contains more volatile matter and carbon content.

337



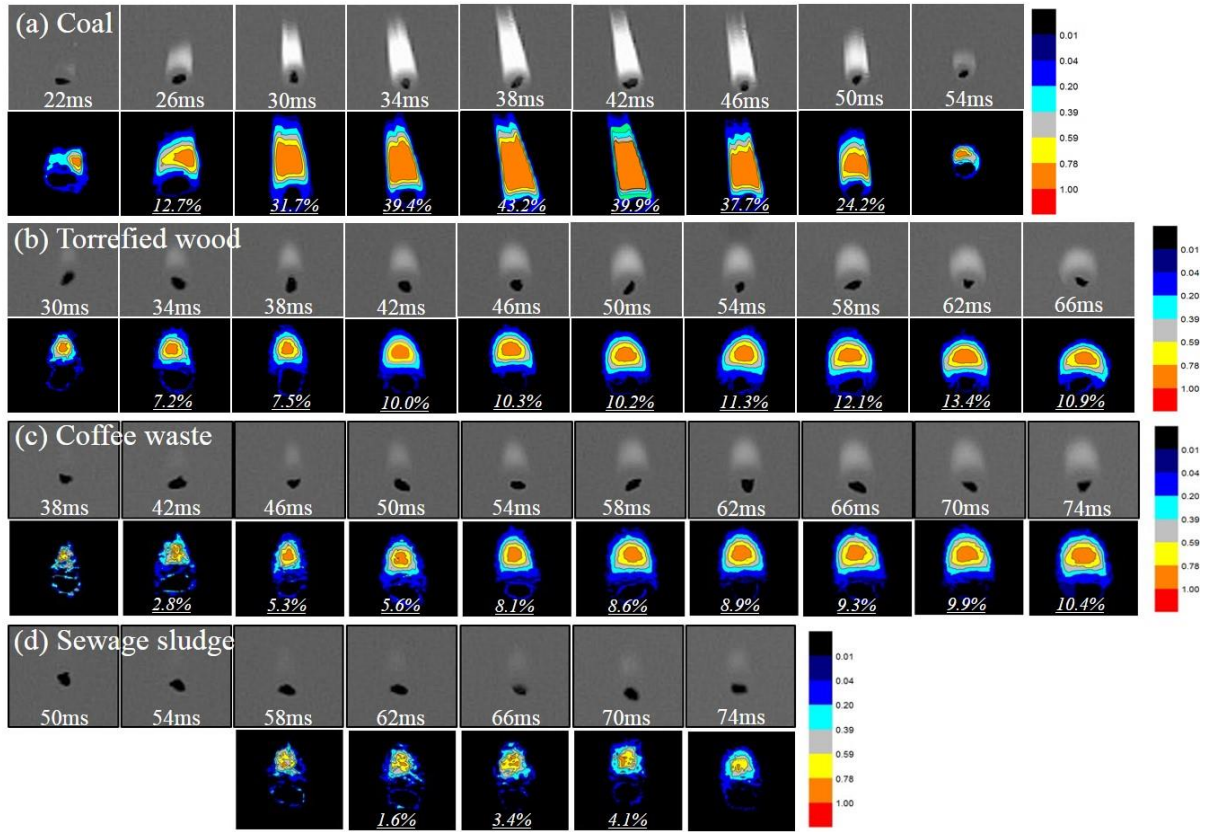
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339 Figure 8. Flame structures and luminosities of the superimposed coal and biomass particles
340 entrained into hot gas streams of 1,340 K (300–355 μm , 10 % oxygen concentration). The
341 particle displacement after the injection is marked in the time interval of 2 ms and apparent
342 volatile ignition occurs within a few milliseconds depending on the type of solid particle.

343

344 The intensity of volatile flames for the four particles is converted into a greyscale image by
345 means of a numerical data matrix shown in Fig. 9. Identical backlighting is used in all images
346 to minimise the error in the numerical solution. In the imaging process, each pixel of the
347 image presents light intensity on a greyscale from 0 to 255, and the numerical backlighting
348 data are extracted from the images. The intensity of the flame is then divided by the
349 maximum value of grayscale (255) for comparative analysis of each image. Coal has max.

350 43.2 % of the highest intensity between 0.78 and 1 in the luminous flam whereas torrefied
 351 wood, coffee waste and sewage sludge account for 13.4, 10.4 and 4.1 %, respectively.



352

353 Figure 9. Captured images obtained from burning particles of 300–355 μm and the
 354 normalised intensity of their flames represented by the imaging process.

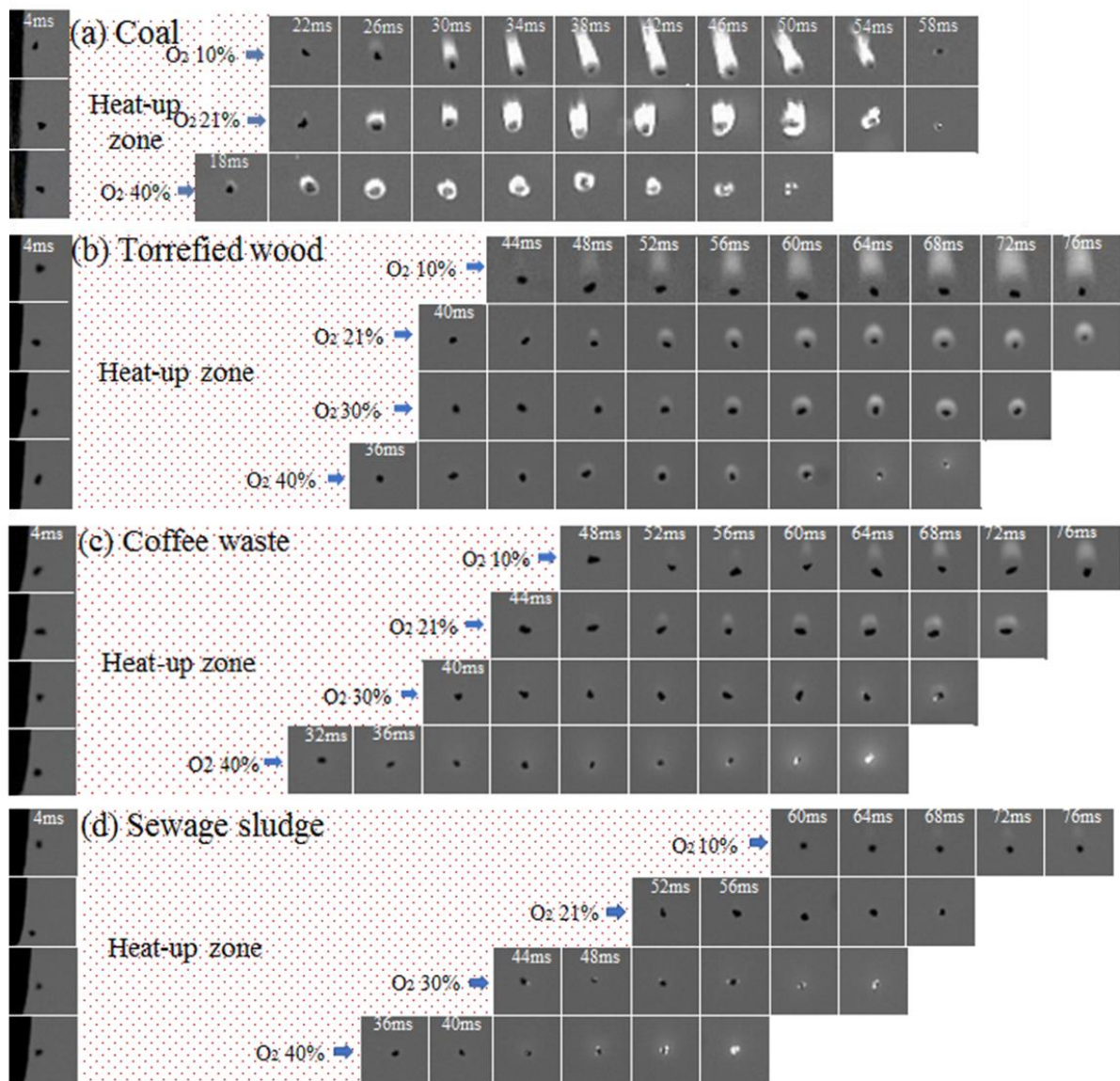
355 *Effects of biomass type and enhanced O_2 concentration*

356 The comparative combustion behaviour for different biomass types and different O_2
 357 concentrations are presented in a series of images shown in Fig. 10. Increase in the oxygen
 358 concentration for each particle is revealed to result in different physical structures of the
 359 volatile flames with different onset times of ignition. The earlier apparent ignition of coal
 360 occurs at all oxygen concentrations, and the biomass flame size is smaller than that of coal
 361 for all size groups examined. However, the three biomass types had different flame intensities

362 with different ignition delays and overlapping combustion at the early stage. Torrefied wood,
363 coffee waste and sewage sludge manifest different changes with oxygen concentration. Kuo
364 et al. [34] pointed out that ignition behaviour such as the structures of the volatile flame,
365 ignition time and early ignition are mainly determined by the properties of the biomass.
366 Normalised intensities of actual flames for different particles are shown in Fig. 11, but a
367 whole period is not detected until the extinction of volatile flame because of the limitation of
368 the visualisation section. The size of volatile flame on a particle exhibits a nearly symmetrical
369 profile, as indicated by the experimental results. Therefore, the unmeasured data due to the
370 limited visualisation field were predicted in Fig 11 and 12. Fig. 11 presents the profiles with
371 predicted values. Based on a previous study [35], these differences between particles are
372 partly attributed to different ratios of hydrogen to carbon, which affect the soot volume
373 fraction. The particle is heated by radiation from the soot flame by a feedback mechanism [29]
374 along with the combustion front and also by convection from a hot gas stream in the reactor.
375 For that reason, the particle temperature is affected by the degree of soot flame. Accordingly,
376 low soot and invisible flames influence ignition delay, flame instability and low radiation in
377 the visible and near-infrared spectral ranges. The radiation from emission of soot particles in
378 a flame has an important role in burning and spread rates [36–39].

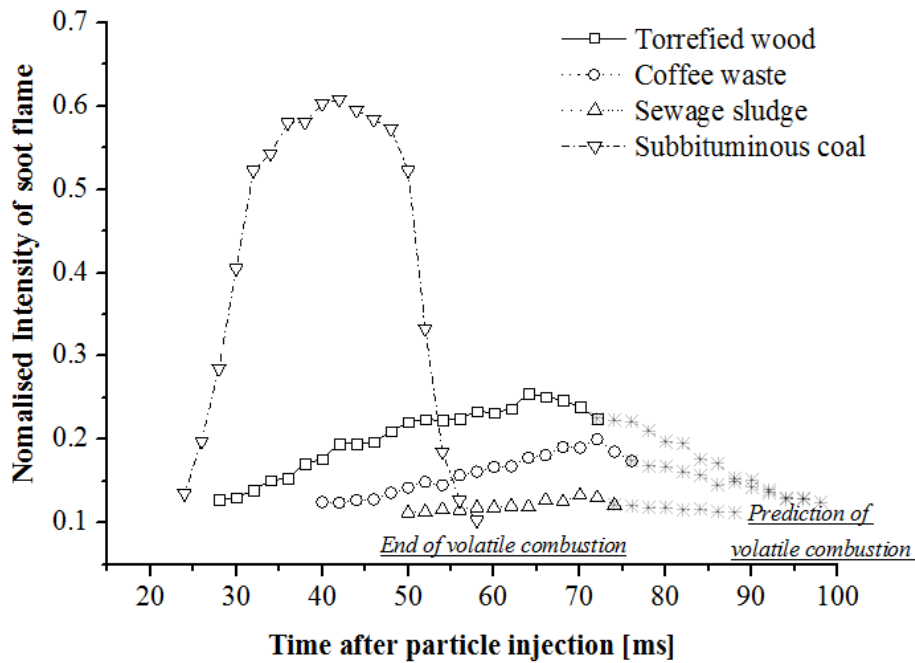
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381

382 Figure 10. Comparative combustion behaviour over time intervals of 4 ms, obtained from
 383 the set of burning particles of size 255–300 μm at 1340 K as the oxygen concentration is
 384 increased.



385

386 Figure 11. Effect of different fuel particles measured from grayscale images of 300–355 μm
 387 particles exposed at 1340 K. Biomass particles are still burning during the volatile
 388 combustion stages and the predicted lines of the volatile flame are extended due to the
 389 symmetrical tendency of the patterns.

390

391 The ratio of the radii of effective flame to particle size is plotted for the biomass particles as a
 392 function of the oxygen concentration, as shown in Fig. 12. The profile which has unmeasured
 393 flame is also extended to predict the whole flame history. The effective maximum ratio
 394 between torrefied wood, coffee waste and sewage sludge with 255–300 μm particles varies
 395 by 5.2, 3.5 and 3.4, respectively, at 10% oxygen concentration. The size of the volatile cloud
 396 decreases with a shorter duration of devolatilisation as the oxygen concentration increases.
 397 Previous work [24, 40] reported that a fast mass reduction during devolatilisation is due to
 398 higher oxidation of the gas phase. The decreases in the radius and shorter duration are only
 399 clearly observed in torrefied wood and coffee waste over all enhanced oxygen concentrations.

400 The radius for sewage sludge is not detected when the oxygen concentration is over 21 % as

401 the particle has too little soot and an invisibly thin flame. Interestingly, the reduction in

402 volatile radius is remarkable in torrefied wood as oxygen concentration varies between 10 %

403 and 20 %. It is believed that sewage sludge particles below 255–300 μm have significantly

404 low radiant energy due to thin and invisible flames, which may affect power generation in a

405 pulverised biomass plant. These flame structures, which contain soot particles and gas phase,

406 have an important role in radiant energy. The energy is attributed to the flame size and its

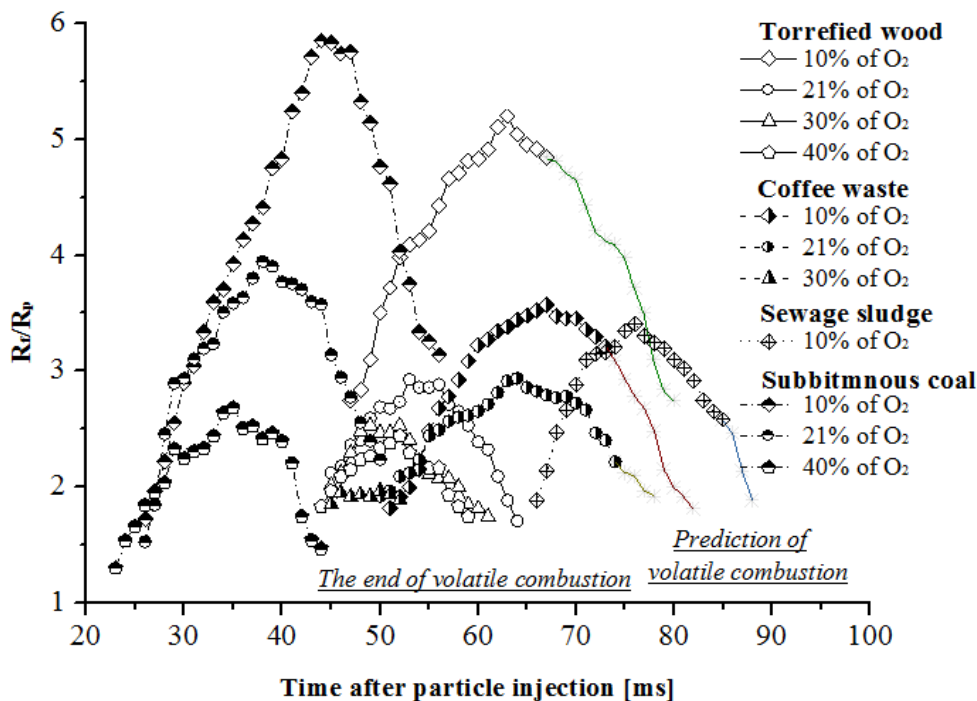
407 high intensity and, the measured parametric values can be discussed by time-averaged

408 radiation [41], $Q_r = \sigma \epsilon_T T_f^4 A_f$ where σ is the Stefan-Boltzmann constant ($5.67 \times 10^{-11} \text{ kW/m}^2\text{K}^4$),

409 ϵ_T is the flame emissivity, T_f^4 is the average flame radiation temperature and A_f is the flame

410 surface area.

411



412

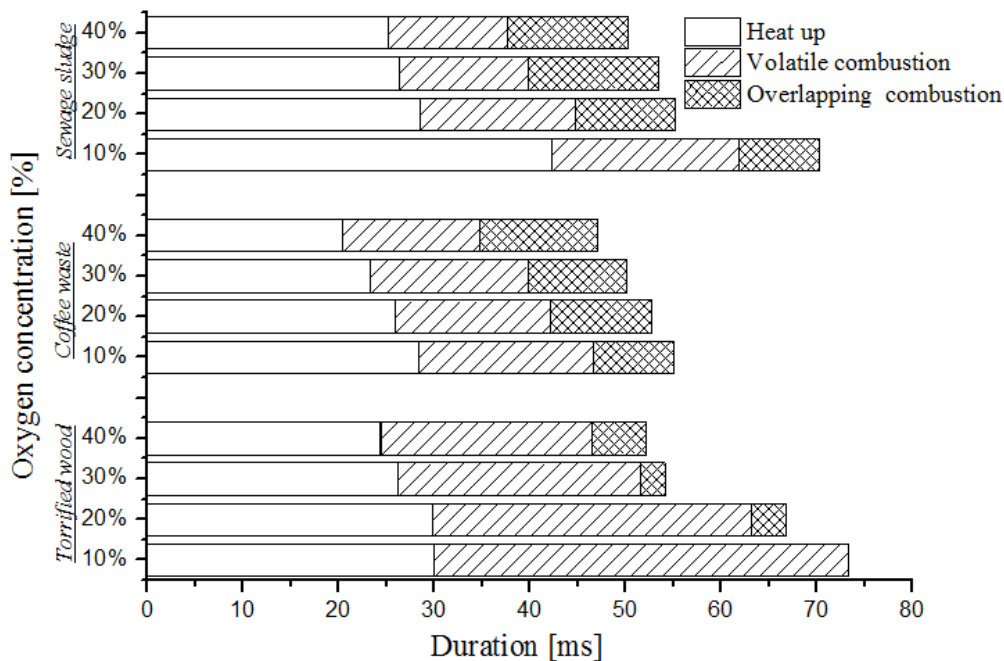
413 Figure 12. Effect of oxygen concentration on torrefied wood, coffee waste and sewage sludge

414 with 255–300 μm particles at 1340 K. Volatile flames of sewage sludge are not detected for

415 oxygen concentrations over 21 % due to low soot and very thin volatile cloud formation.

416

417 Average ignition delay against time is analysed for the three biomass particle types of 155–
418 215 μm and all oxygen concentrations, as shown in Fig. 13. It is apparent that the heating
419 time and durations of homogenous and heterogeneous combustion decrease as oxygen
420 diffusivity increases. From the figure, longer volatile combustion and shorter overlapping
421 combustion are observed in torrefied wood (which has experienced thermal pre-treatment),
422 whereas coffee waste and sewage sludge have relatively shorter durations of volatile
423 combustion. In the case of 155–215 μm particles, early onset of char ignition appears only for
424 coffee waste and sewage sludge, which may be related to the rapid increase in particle
425 temperature, enabling a shorter duration of devolatilisation.

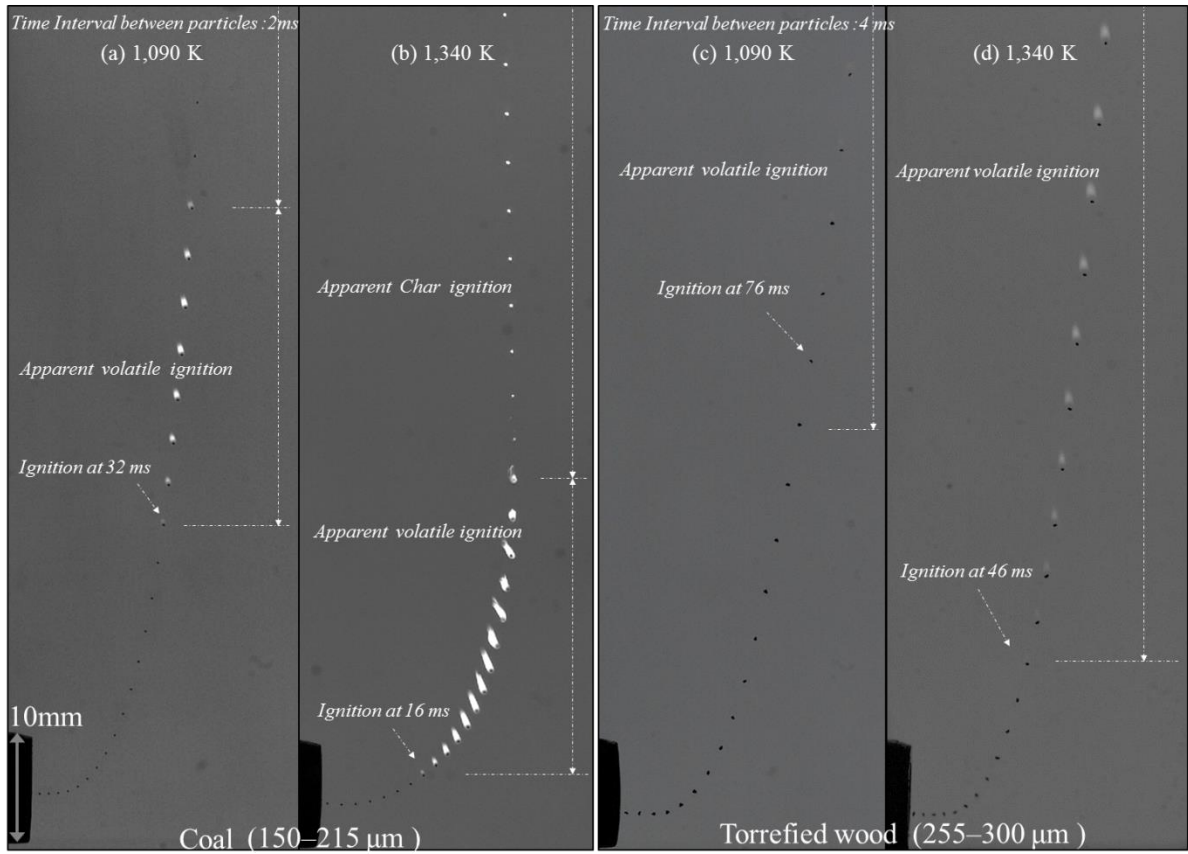


426

427 Figure 13. Average duration of heat up and the end of volatile combustion for three biomass
428 particles (155–215 μm) at 1340 K, and quantitative measurements of burning time vs. oxygen
429 concentrations.

430 *Effect of environmental gas temperature on combustion behaviour*

431 The burning of particles enable discussion of the effect of temperature conditions on
432 combustion behaviour. At 1090 K and 1340 K, the trajectories of coal and torrefied wood are
433 compared in Fig. 14 to characterise the volatile flames. Both particles at 1090 K form volatile
434 flames of smaller sizes with lower intensity, and the flame of torrefied biomass even becomes
435 transparent with longer ignition delay. This suggests that significant soot formation does not
436 occur at low temperature, along with low volatile release rate, as compared with a higher gas
437 temperature of 1340 K. Fig. 15 shows that the average duration of heat-up at 1090 K is longer
438 than that at 1340 K. The apparent change in the heat-up duration at low temperature as a
439 function of oxygen concentration is more prominent than that at 1340 K. This is consistent
440 with the result from a previous study [6], which showed that the effect of oxygen
441 concentration on ignition delay and burnout time is expected to be slightly more pronounced
442 at lower temperature.

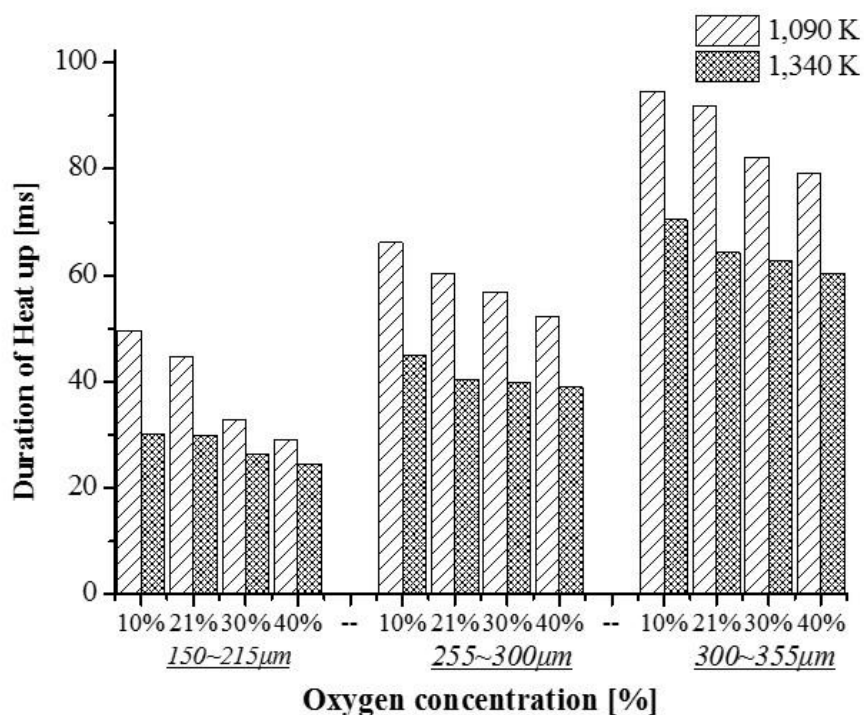


443

444 Figure 14. Comparative volatile flames and ignition behaviour of coal and torrefied wood

445 between 1090 K and 1340 K at 10% of O₂: coal, 150–215 μm and torrefied wood, 255–300

446 μm.

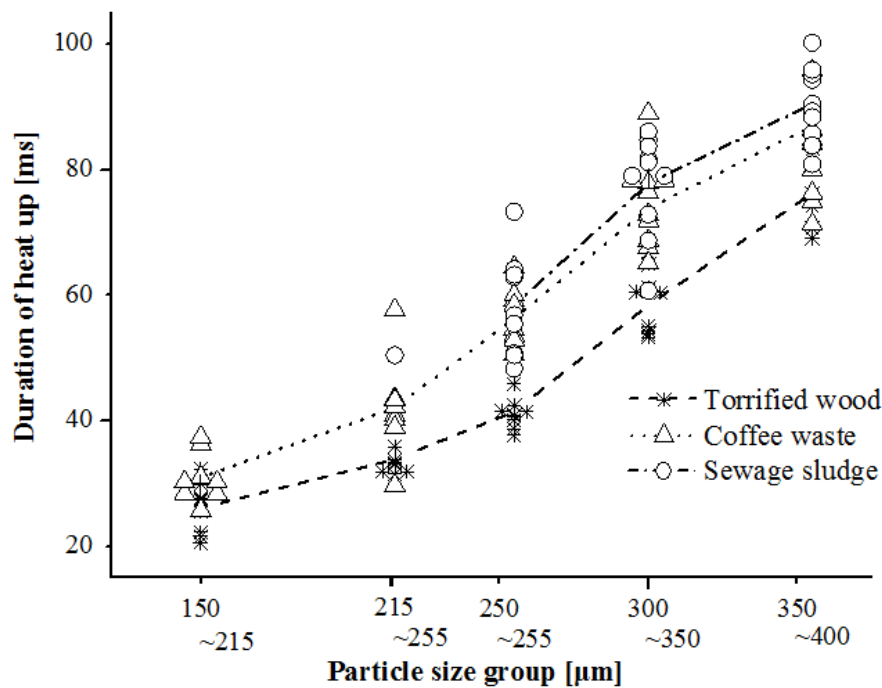


447

448 Figure 15. Average duration of heat up for torrefied wood particles of 150–355 μm under
 449 different gas temperature and oxygen concentration conditions.

450 *Effect of particle size on combustion behaviour*

451 Several researchers [42, 43] have studied intra-particle effects during combustion. In
 452 general, the temperature of a large-sized particle is controlled by the surrounding temperature
 453 and this particle has an intra-particle temperature gradient. However, a pulverised particle
 454 under 200 μm has a small temperature gradient which does not affect the rate of
 455 devolatilisation [15, 44]. Fig. 16 illustrates the average duration of heat-up for particles of
 456 each of the three biomass types with particle size in the range from 150–215 μm to 355x425
 457 μm, each taken over 20 particle samples. The transience point of ignition delay for torrefied
 458 wood occurs at the size group of 255–300 μm and coffee waste at 215–255 μm, where
 459 average time of heat-up increases more sharply, probably due to an intra-particle effect.

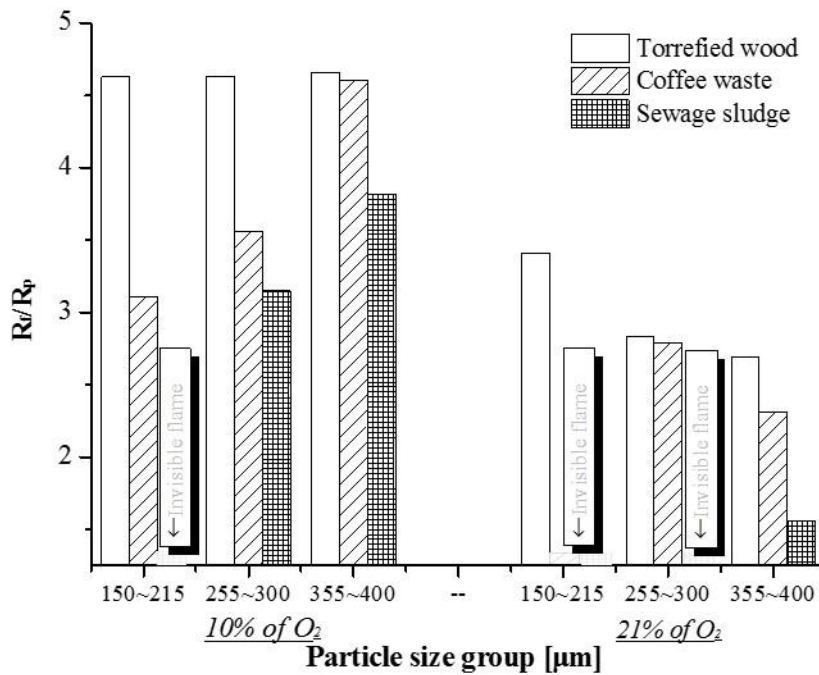


460

461 Figure 16. Experimental data on ignition delay for biomass particles in the range 150–400
 462 μm at 1340 K and 21 % oxygen concentration.

463 Fig. 17 illustrates the maximum effective radius of volatile flame for particles in the ranges
 464 from 155–215 μm to 355–425 μm under oxygen concentrations of 10 and 21 %. First, a
 465 higher effective radius is quantitatively measured at low oxygen concentration for all
 466 particles. Second, R_f/R_p in torrefied wood undergoes only a marginal change with a relatively
 467 sooty flame from 155–215 μm to 355–425 μm at 10 % oxygen concentration. However, this
 468 radius (R_f/R_p) decreases with particle size increase at 21 % oxygen concentration. The radius
 469 of torrefied wood and coffee waste with 255–300 μm particles is almost equal to that at 21%
 470 oxygen concentration, but coffee waste has a very thin, low soot flame when its particle size
 471 is 155–215 μm. Consequently, coffee waste is required to be at least 255–300 μm in size to
 472 achieve an equivalent flame structure to torrefied wood. Finally, an invisible or very thin
 473 flame is detected for sewage sludge when it is burned at 21 % oxygen concentration. The raw
 474 sewage sludge in these experimental conditions is low combustion quality of biomass. An

475 increase in sludge particle size might enhance the flame stability, but a large particle would
 476 drop to the bottom of the reactor without combustion. To avoid this, a higher gas temperature
 477 or thermal pre-treatment of sewage sludge might be required.



478
 479 Figure 17. Average effective flame radii of three biomass particle types in the range 150–400
 480 μm between 10 % and 20 % oxygen concentration.

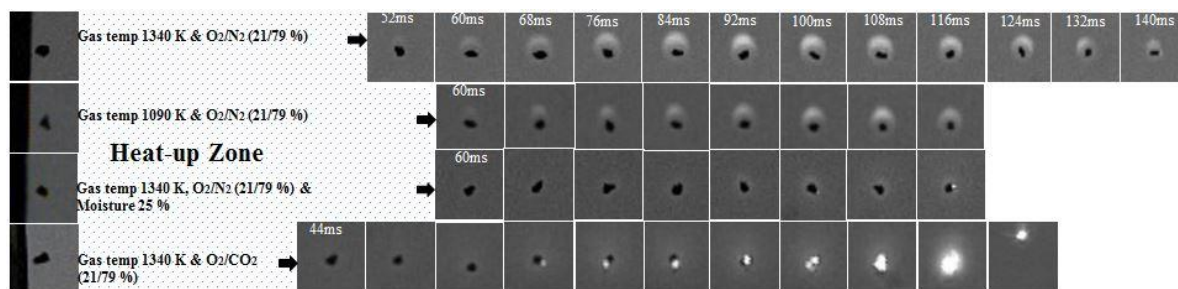
481 **Discussion of special cases**

482 *Ignition behaviour and flame instability*

483 Ignition behaviour is determined by interactions between complicated factors such as the
 484 physical and chemical characteristics of the biomass and the surrounding environment as
 485 well as operating conditions. Variation in the composition could enable a longer ignition
 486 delay, smaller soot cloud and earlier fragmentation with the instability of the volatile flame.
 487 The investigation of ignition delay as a function of certain parameters may provide partial

488 answers concerning optimised particle combustion. Fig. 18 shows the onset time of volatile
489 ignition, earlier overlapping combustion and the visible structure of volatile flame in
490 different environmental conditions. There are four observations of burning particles in
491 different environmental conditions. The two different gas temperatures were used to
492 simulate the moisture and oxy-fuel combustion effects. For high mass volume of moisture,
493 particles were put in a closed container (20 cm×20 cm) on a hot plate at 323 K. There was a
494 sponge with high moisture in the bottom of the container, with no particle-sponge contact.
495 The group of particles was kept in the container for 4 hours at 95 % absolute humidity. As a
496 consequence, 25 % moisture was added to the particles. The particles which were out of the
497 container had to be tested quickly because this moisture content was likely to be evaporated
498 in a short time. To compare O₂/N₂ and O₂/CO₂ environments, CO₂ for 21% O₂ concentration
499 was supplied from the post combustion burner, instead of air. First, the flame instability is
500 found to be related to environmental temperature. A previous study [34] observed a different
501 model of ignition obtained from anisotropy of the thermal properties of large biomass
502 particles and reported that different ignition behaviour was determined by the surrounding
503 gas temperature. Second, moisture content enhanced the ignition delay at the early stage,
504 which caused earlier char combustion with the formation of a thin flame. Kucuk et al. [45]
505 pointed out that the evaporation of moisture leads to an increase in char combustion to be
506 attacked by oxygen due to the formation of an active centre. Lastly, early ignition and flame
507 instability of a particle occur under CO₂ atmosphere due to the high thermal capacity of CO₂
508 and the high reactivity of char-CO₂, as reported by Shaddix et al. [22].

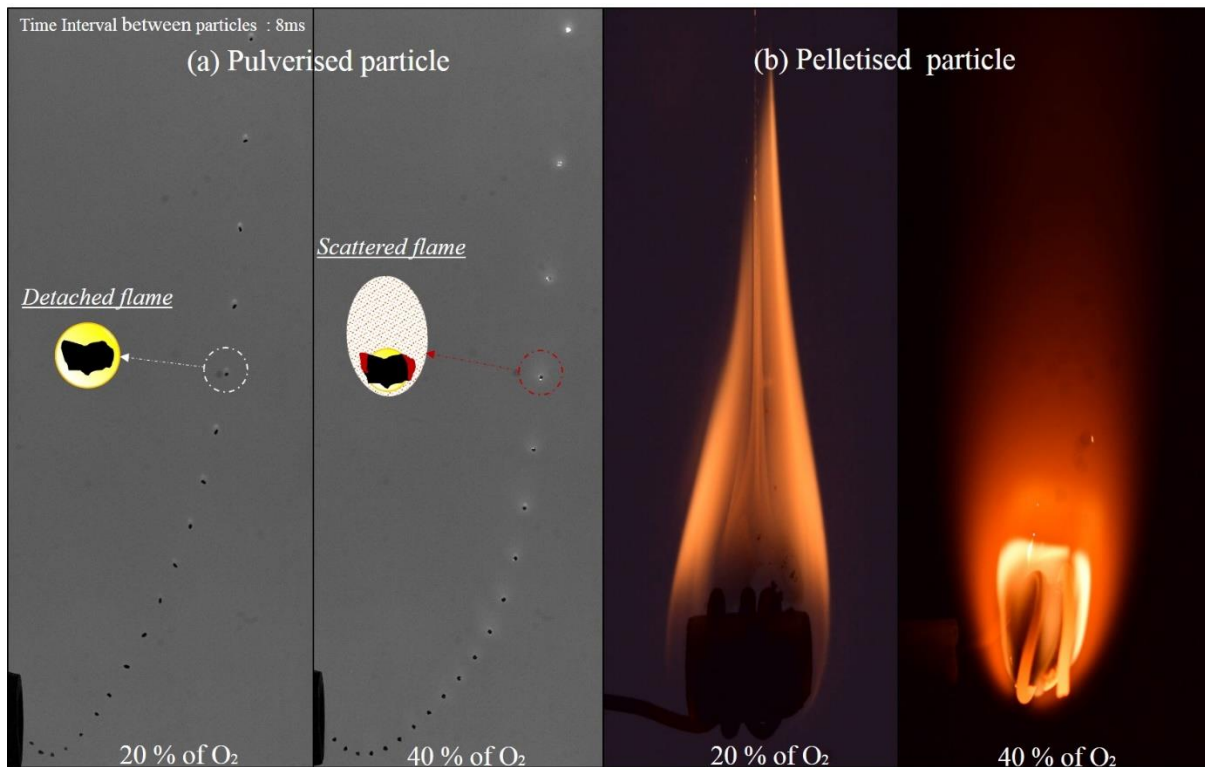
509



510

511 Figure 18. Comparison of combustion behaviour on coffee waste particles for four
 512 different environmental conditions.

513 Insights into flame formation are essential to understand combustion behaviour, such as
 514 flame stability, to better determine the operating conditions and select appropriate biomass.
 515 Fig. 19 illustrates two types of volatile flames that are observed for a single particle and a
 516 pellet of coffee waste. As mentioned before, coffee and sludge particles typically have a
 517 thinner volatile flame or an invisible flame compared with torrefied wood particles. At high
 518 oxygen concentrations, scattered volatile gases and small particles that escape from parent
 519 particles are detected above an invisible or very thin diffusion flame. From the observation,
 520 the actual size of the flame can be reduced during the combustion process. To support this
 521 flame instability, a pelletised biomass particle is examined at the same gas temperature. The
 522 size of diffusion flame at 40 % of O₂ is apparently smaller than that at 20 % of O₂, and this
 523 flame partially detached over an enlarged scattering flame at high oxygen concentration. This
 524 phenomenon is attributed to rapid escape of high-volatility matter and particles from a
 525 luminous flame without any reaction. Consequently, the onset of volatile scattering enables
 526 the formation of very thin volatile flames with relatively low soot content. These results are
 527 consistent with the work of Holtmeyer et al. [46], who studied combustion modes through
 528 experimental and numerical methods. The mechanism of this volatile scattering is not clearly
 529 defined, but it is frequently experienced by biomass with highly volatile content, anisotropic
 530 release and a low-soot flame above a diffusion flame.



532

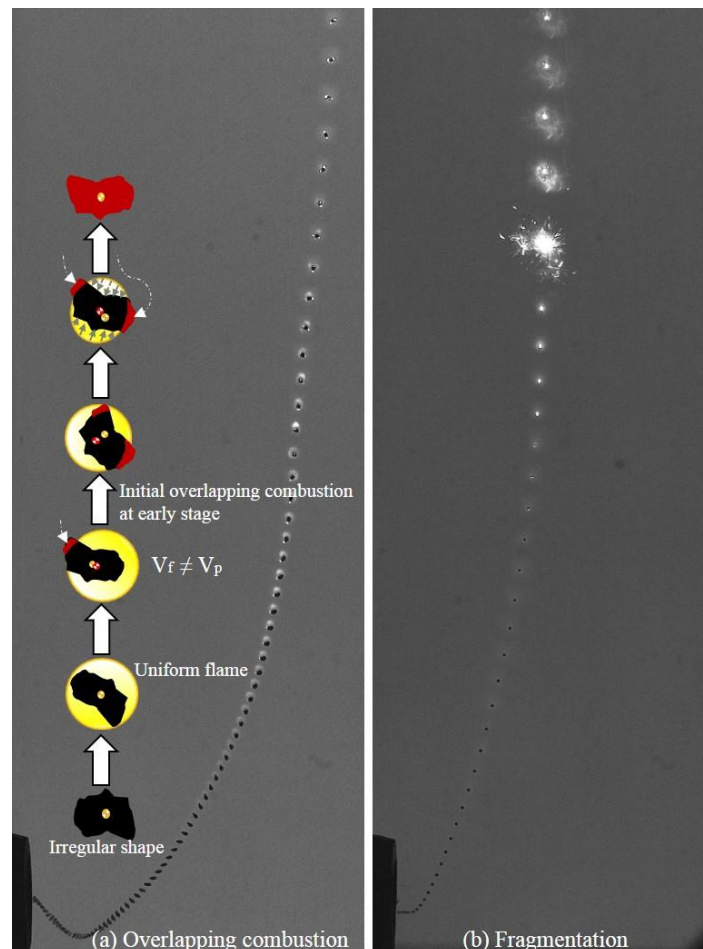
533 Figure 19. Different modes of the volatile flame of coffee waste produced under enhanced
 534 oxygen concentration.

535 *Overlapping combustion at an early stage and fragmentation*

536 Overlapping combustion is attributed to low gas temperature, rapid heating rate, small
 537 particles and low flame formation. In addition, the shapes of the particle, the different
 538 rotational speeds between them and the volatility of the flame may be related to the
 539 overlapping combustion shown in Fig. 20. Normally, the volatile flame with an O₂
 540 concentration over 21 % has a relatively thin spherical shape for an irregular particle. From
 541 the figure, we note that the particle exposed to a hot gas stream starts rotational motion, and
 542 then a volatile flame is formed after volatile matter release. In the process, the different
 543 rotational speeds of the particle and volatile flame are detected after volatile ignition. Then,
 544 an edge of the irregular particle is suddenly exposed in a hot gas stream without a

545 surrounding volatile flame. This exposed edge is directly heated by the surrounding hot gas.
546 The different rotational motions between a particle and volatile flame can change the datum
547 of irregular objects. Sequentially, the opposite edge of the particle is rapidly ignited and char
548 combustion is extended over the surface since the volatile flame shrinks as a function of the
549 residence time. This earlier overlapping combustion and a fast devolatilisation at high
550 temperature and a rapid heating rate enhance the thermal stress and internal pressure inside
551 the particle, leading to a high probability of fragmentation. Only coffee waste particles
552 occasionally experience this phenomenon for oxygen concentrations over 30% and in the size
553 range of 150–255 μm .

554



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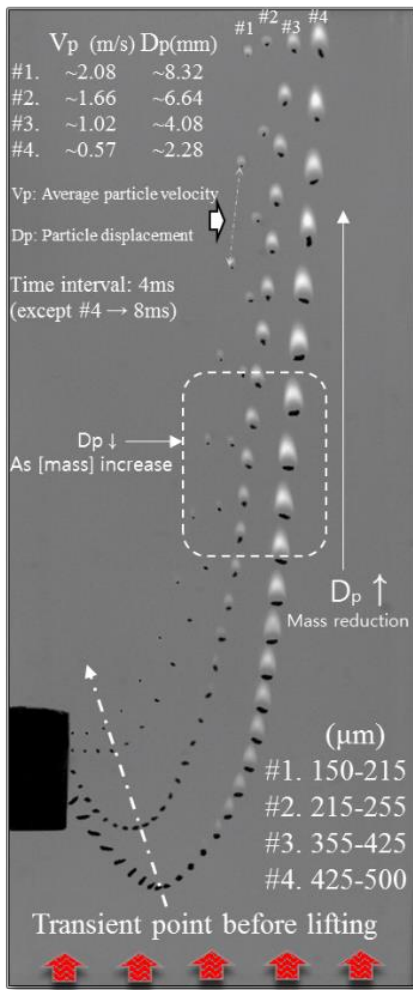
556 Figure 20. Overlapping combustion and fragmentation occurring in the coffee waste

557 particle.

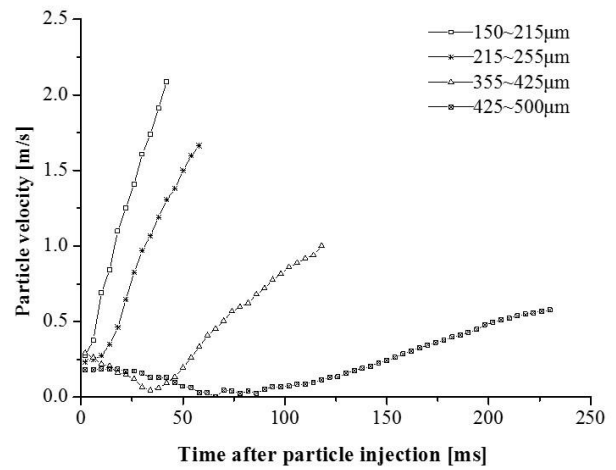
558

559 *Trajectories of small to large biomass particles for optimisation of burnout*

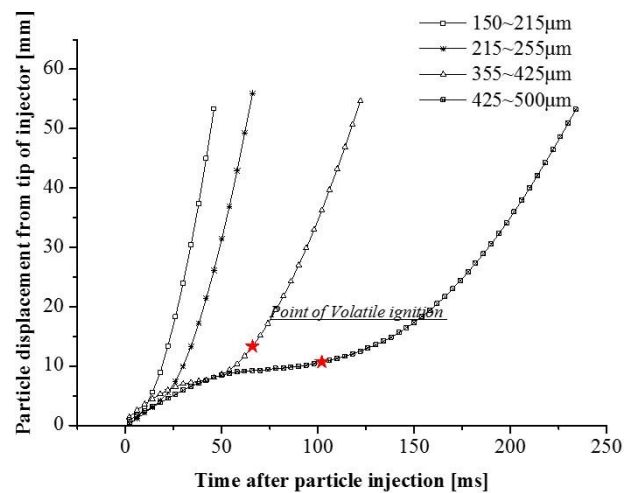
560 The particle size of biomass is expected to be larger than that of coal for pulverised
561 combustion because of its low density and fast reaction rate. However, large particles injected
562 into a burner can be incompletely burned as they may drop to the bottom of the furnace
563 without combustion. Hence, determination of the particle size for complete burning is
564 essential to achieve a high level of utilisation for pulverised combustion. Saastamoinen et al.
565 [17] also analysed the possible trajectory of particles to be burned virtually and suggested an
566 optimised particle-size for biomass. Fig. 21 shows clearly how realistic trajectories of
567 biomass particles in the cross-sectional configuration develop with regard to size variation.
568 From this, the maximum particle size that can be completely burned is suggested
569 experimentally. Small particles, which range from 150 to 255 μm in diameter, move in the
570 direction of the hot gas stream, regardless of gravity force, as particle velocity increases.
571 Particles of 355–425 μm drop to the bottom of the furnace and are then lifted toward the top
572 due to buoyancy and drag forces as particle mass decreases. However, particles of 425–500
573 μm occasionally drop to the bottom without volatile combustion. Most particles over 500 μm
574 do not burn in the visualisation field because of excessive particle mass and non-mass
575 reduction in the experiment under particular environment conditions. Karampinis [47] also
576 reported that high volumes of particles may go down to the hopper region, where these
577 particles burn without upward movement.



a) Trajectories of burning particles



b) particle velocity



c) Particle displacement

578 Figure 21. Trajectories of small to large biomass particles for burnout, particle velocity and
 579 displacement in the combustion process.

580 Conclusion

581 Different burning characteristics of disparate biomass particles were observed because of
 582 differing chemical and physical properties. These observations were carried out in a
 583 laboratory-scale entrained flow reactor with cross jet injection using a high speed camera.

584 Combustion behaviours of biomass particles have been analysed as a function of time and
585 characterised in terms of the effects of oxygen concentration, particle size, particle
586 temperature and other environmental conditions. Also, this study was focused on a
587 measurement of the effective size of the flame over the particle with its intensity to clarify its
588 flame stability along with supporting the modelling description.

589

590 Observation of burning particles shows that flame volatility is not only highly related to the
591 particle's volatile matter content, but is also attributed to soot formation. Biomass has a
592 relatively more volatile content than coal and yields a less sooty flame, but a stable flame is
593 detected for torrefied wood even at the smallest pulverised particles of size 150–215 μm .
594 However, coffee waste requires larger size of over 255 μm to obtain a flame profile
595 equivalent to that of torrefied wood. Sewage sludge in the same size group as other biomass
596 particles has a highly transparent volatile or invisible flame with a longer ignition delay. The
597 initial volatile ignition of the three biomass particle types occurs differently, as these
598 processes are dominantly determined by their compositions, and different effective radii for
599 the volatile flames are observed for each particle.

600 Coffee waste and sewage sludge particles in the size range of 155–215 μm undergo char
601 combustion at an early stage and form a flame with low-soot content. This increases particle
602 temperature rapidly and enables a shorter devolatilisation duration compared with torrefied
603 wood. Also, these particles have thin or invisible flame, and scattered volatile flames are
604 detected at high oxygen concentration from the direct observation. To achieve relatively a
605 soot flame, biomass particles are required to be larger but, the size of pulverised particle is
606 limited due to a probability of unburned particles.

607 The determination of particles size is essential to achieve complete particle burnout in

608 pulverised combustion. From the experimental results, particles with diameters of 150-255
609 μm , exposed to a hot gas stream with the studied parameters, move in the direction of the
610 vertical stream, regardless of gravity force. The particle size group of 425–500 μm
611 occasionally falls to the bottom to be incompletely burned without any apparent mass
612 reduction. Consequently, the maximum size of particle that can be burned completely at
613 1,340 K is in the size range of 355–425 μm . From this result, the optimal biomass particle
614 size would be suggested for a large-scale combustion furnace.

615

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620 sample preparation.

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