

1 **A numerical model for the fractional condensation of pyrolysis vapours**

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8 **Abstract**

9 Experimentation on the fast pyrolysis process has been primarily focused on the pyrolysis reactor
10 itself, with less emphasis given to the liquid collection system (LCS). More importantly, the physics
11 behind the vapour condensation process in LCSs has not been thoroughly researched mainly due to
12 the complexity of the phenomena involved. The present work focusses on providing detailed
13 information of the condensation process within the LCS, which consists of a water cooled indirect
14 contact condenser. In an effort to understand the mass transfer phenomena within the LCS, a
15 numerical simulation was performed using the Eulerian approach. A multiphase multi-component
16 model, with the condensable vapours and non-condensable gases as the gaseous phase and the
17 condensed bio-oil as the liquid phase, has been created. Species transport modelling has been used to
18 capture the detailed physical phenomena of 11 major compounds present in the pyrolysis vapours.
19 The development of the condensation model relies on the saturation pressures of the individual
20 compounds based on the corresponding states correlations and assuming that the pyrolysis vapours
21 form an ideal mixture. After the numerical analysis, results showed that different species condense at
22 different times and at different rates. In this simulation, acidic components like acetic acid and formic
23 acids were not condensed as it was also evident in experimental works, where the pH value of the
24 condensed oil is higher than subsequent stages. In the future, the current computational model can
25 provide significant aid in the design and optimization of different types of LCSs.

26 Keywords: Condensation, Liquid collection system, Species transport, Fast pyrolysis, Phase change.

27 **1. Introduction**

28 In recent years, the research on renewable energy is gaining momentum because of the shortage of
29 fossil fuels and emission problems [1] . Among the various kinds of renewable energy sources,
30 biomass is widely considered to be an important energy source for the future as it is the only source in
31 this category which can produce solid, liquid and gaseous fuels [2] . Biomass can be converted to
32 more valuable forms of energy through a number of processes including thermal, biological, and
33 mechanical or physical processes. Even though the pyrolysis technique has been applied for thousands
34 of years, it has gained a considerable interest only during the last 30 years [3] . Fast pyrolysis is a

35 popular thermochemical conversion method, which takes place at moderate temperatures (around
36 500°C) and very short reaction times (around 2 s) [3] . The process results in high yields of liquids
37 (bio-oil) of up to 75 wt. % which can be used directly in a variety of applications or used as an
38 efficient energy carrier in the upgraded form [4, 5].

39 The properties of the bio-oil fuel can be significantly improved by treatment prior to the LCS system
40 operation (e.g catalytic pyrolysis) or post treatment (e.g hydroprocessing) after the liquid bio-oil
41 collection. A typical issue with the hydroprocessing of bio-oils is that different components react
42 differently in the presence of catalysts under specific conditions [6] . Moreover, the removal of sugar
43 type of components from the pyrolysis oils prior to the hydroprocessing is highly recommended as
44 these components are prone to coking [7] . Therefore, selective condensation which enables the
45 fractionation of the bio-oil might be an efficient way to produce specific liquid fuels and chemicals
46 compared to the direct complete condensation of pyrolysis vapours before upgrading [8, 9].

47 Despite the fact that several researchers have focused on the modelling of the pyrolysis reactors [10 -
48 17] cyclone separators and even ESPs (electrostatic precipitators) [18-20] used in pyrolysis
49 technology, very few research attempts have been made on the modelling of the condensation units
50 [21, 22]. This is primarily due to the complex nature of the physics involved in the condensation
51 process of pyrolysis vapours. Also, the limited availability, or in some cases the total absence, of
52 experimental data for specific types of condensers, impose significant restrictions in the development
53 of detailed and validated numerical models.

54 The development of a condensation model prerequisites a representative description of the pyrolysis
55 vapour components. Typical bio-oil consists of a complex mixture of over 300 chemical components
56 [23] . From the literature it is observed that few computational modelling efforts have been made on
57 the evaporation of biomass pyrolysis oil droplets. Two approaches were primarily used to represent
58 the bio oil composition. One approach is with a continuous thermodynamics model [24 - 27] and
59 another with a discrete representation based on selected components in the bio-oil [28] .

60 In the continuous thermodynamics model, a multi-component mixture like bio-oil can be represented
61 by few chemical groups that can be characterized using probability density functions. The parameters
62 of each distribution function can be derived by conducting numerical distillation tests [24] . This
63 technique has been successfully employed so far in the evaporation modelling of the bio oil.
64 Numerous studies are based on this principle. However, this may not be true for biomass derived oil;
65 mainly because of its nature and that it cannot be easily distillable [29] .

66 On the other hand, the discrete representation of the bio-oil composition facilitates the inclusion of a
67 finite number of species which can be deduced from GC/MS analysis. Conversely, representing bio-
68 oil's composition with this method is not complete, as the bio-oil is composed of hundreds of different
69 species. Evaporation modelling of bio-oil droplets with this method was presented by Brett et al. [31] .

70 Papadikis et al. [21] utilized this discrete representation of the pyrolysis vapour components to
71 simplify the complex condensation characteristics. However, this model assumes a uniform vapour
72 composition that remains constant throughout the condensation process. In the actual physics of the
73 condensation of vapours, different components of the pyrolysis vapours condense at different rates
74 and hence the composition of the vapours varies continuously.

75 The scope of this paper is to simulate the phase change phenomena and to capture the selective
76 condensation of the pyrolysis vapours inside the liquid collection system (LCS). In this study a
77 double surface water condenser shown in Fig. 1 is utilized in the LCS. The commercial CFD code
78 ANSYS Fluent 13.0 has been used as the computational platform for the simulation of the
79 condensation process. The phase change phenomena were incorporated to the code as user defined
80 functions.

81 **2. Experimental conditions**

82 The double surface water condenser (Fig. 1) was used in the experiments conducted at Aston
83 University as a first stage condenser within the liquid collection system (LCS) [32]. According to
84 Salter [32], the flow rate of the cooling water is 2L/min at 15°C and the vapour flow rate into the
85 condenser 12L/min at 500°C. The vapour flow includes condensable and non-condensable gases. In
86 the current computational model the wall surfaces, which are exposed to the water, are modelled with
87 constant temperature of 15°C with an assumption of marginal increase in cooling water temperature
88 in the system.

89 Biomass was injected to the experimental reactor at a rate of 100 g/hr. According to the mass balances
90 obtained from the experiments, the total conversion of biomass into vapours is approximately 70
91 wt.%, whereas the non-condensable gases amount to 15 wt.% and char yielded from 9-11%. A mass
92 imbalance of 5% was reported for a pyrolysis reactor temperature of 500°C. It is worth to note that the
93 previously mentioned yields are sensitive to the pyrolysis reactor temperature [33]. In the above
94 experiments Nitrogen has been used as the carrier gas, which is also modelled together with the non-
95 condensable gases.

96 **3. Condensation model**

97 The condensation model used in this paper is an analytical improvement of the Papadikis et al. [21]
98 condensation model. In contrast with the uniform vapour composition presented in the model of
99 Papadikis et al. [21], the current model treats each individual species as a separate compound that is
100 condensed according to its individual saturation vapour pressure. In this way, fractional condensation
101 modelling is enabled and the prediction of the bio-oil composition at each condensation stage
102 becomes possible, once the initial vapour composition is known. The pyrolysis vapour composition is

103 highly dependent on the type of feed used during the pyrolysis process. This composition further
 104 prescribes the type of application for which the produced bio-oil will be used for [30] . The pyrolysis
 105 vapours are modelled by using a discrete representation of 11 chemical species dominant in bio-oil.
 106 The chemical species listed in Table 1, have been taken from the bio-oil composition used in the Brett
 107 et al.'s [31] work. This composition represents a discrete equivalent of the continuous
 108 thermodynamics model, which in turn is based on molecular weight distributions of specific chemical
 109 groups, used in the study of Hallet and Clark [27] . It has to be noted that the selection of the number
 110 of chemical species and their corresponding initial volume fractions can be modified depending on the
 111 chemical compounds of interest. However, one has to make sure that the overall average distribution
 112 of the affected chemical groups in the continuous description remains unchanged. This inevitably
 113 imposes a limitation on the minimum number of discrete chemical compounds in group, which will
 114 have to satisfactorily approximate a continuous curve. The thermochemical properties of each species
 115 were computed based on existing data available in the literature [34] . The critical properties are
 116 computed by using the group contribution method [35] when they are not readily available in the
 117 literature. These properties of the individual species were listed in the Table 1.

118 The present model estimates the saturation vapour pressure of the each species present in the pyrolysis
 119 vapours by utilising the generalized corresponding states method. According to Mejbri and Bellagi's
 120 [36] generalized three parameter corresponding states correlation, the natural logarithm of the reduced
 121 saturated vapour pressure and acentric factor ω^i are in linear relation as shown in eq. 1 with an
 122 averaged fluctuation about 0.16%.

$$123 \quad \ln(P_r^i) = f_0(\tau^i) + \omega^i f_1(\tau^i), \quad (1)$$

124 where τ^i is the inverse of the reduced temperature T_r^i of the i^{th} species and is equal to $1/T_r^i$. The
 125 functions f_0 and f_1 are given by eqs. 2 and 3.

$$126 \quad f_0(\tau^i) = \gamma_1(\tau^i - \exp(1 - \tau^i)) + \gamma_2((\tau^i)^{\gamma_3} - \exp(1 - \tau^i)) \quad (2)$$

127 and

$$128 \quad f_1(\tau^i) = \gamma_4(\tau^i - \exp(1 - \tau^i)) + \gamma_5((\tau^i)^{\gamma_6} - \exp(1 - \tau^i)). \quad (3)$$

129 The values of the six universal γ coefficients which were used in above two equations are listed in
 130 Table 2.

131 For estimating vapour pressures by using eq. 1, the critical pressures and temperatures are needed
 132 along with the acentric factor. If the acentric factor is not available, Mejbri and Bellagi [36]
 133 recommended estimating it using the boiling temperature T_b^i by using following equation

$$134 \quad \omega^i = \left(0.013162987 - \ln P_c^i - f_0(\tau_b^i)\right) / f_1(\tau_b^i), \quad (4)$$

135 where τ_b^i is the ratio between critical and boiling temperatures i.e. $\tau_b^i = T_c^i/T_b^i$. The critical pressure
 136 P_c^i used in eq. 4 is expressed in bars.

137 The condensation rate is governed by the magnitude of the relative saturation value which is the ratio
 138 of the vapour fugacity (f_v^i) to the saturated vapour fugacity (f_l^i). Under the vapour liquid equilibrium
 139 (VLE) conditions, the relative saturation will be unity. The vapour fugacity in this case is the partial
 140 pressure of the particular species in the system as given in the eq. 5.

$$141 \quad f_v^i = \phi^i P^i = \phi^i y^i P, \quad (5)$$

142 where P^i is the partial pressure of the species 'i' and P is the total pressure of the mixture. y^i is the
 143 mass fraction of the ith species within the vapour mixture.

144 The saturated vapour fugacity can be computed from the reduced saturation pressure as shown in eq.
 145 6.

$$146 \quad f_l^i = \phi_{sat}^i P_r^i P_c^i. \quad (6)$$

147 Here the fugacity coefficients ϕ^i and ϕ_{sat}^i are assumed as 1 and hence the saturated vapour pressure
 148 considered same as saturated vapour fugacity. This is especially true when the system is not under
 149 high pressures.

150 **4. Thermodynamic properties**

151 The thermodynamic properties of the vapour mixture are calculated based on the assumption that the
 152 vapour behaves as an ideal mixture. The viscosity of the vapour mixture is estimated based on the
 153 Dean and Stiel [37] relation which is a function of the reduced mixture temperature. Mixture viscosity
 154 μ_m in eq. 7 is expressed in micro poise.

$$155 \quad \mu_m = \begin{cases} 3.4 T_{rm}^{8/9} / \xi_m & T_{rm} \leq 1.5 \\ 16.68 (0.1338 T_{rm} - 0.0932)^{5/9} / \xi_m & T_{rm} > 1.5 \end{cases}, \quad (7)$$

156 where ξ_m is inverse viscosity and expressed in μP^{-1} this can be calculated by using eq. 8.

$$157 \quad \xi_m = \left(\frac{T_{cm}}{(M_m^3 P_{cm}^4)} \right)^{1/6}. \quad (8)$$

158 The reduced mixture temperature T_{rm} is expressed as the ratio between temperature and mixture
 159 critical temperature. Here the mixture critical temperatures and mixture molecular weight were
 160 calculated by mass fraction average basis i.e. $\sum y^i T_c^i$, $\sum y^i M^i$ respectively. The mixture critical
 161 pressure P_{cm} expressed in terms of atm is calculated using eq. 9

$$162 \quad P_{cm} = \frac{R(\sum_i y^i Z_c^i)}{\sum_i y^i V_c^i} T_{cm}. \quad (9)$$

163 In the above equation, the universal gas constant R is equal to 82.05746 (atm. cm³/ mol-K).

164 Due to lack of the group contribution data, in this analysis, the more accurate correlations like Chung
 165 et al. [38] are not considered for calculating the thermal conductivity. The Eucken correlation offers a
 166 simple method to estimate the mixture's thermal conductivity,

$$167 \quad k_m = \left(1.32 + \frac{1.77}{(C_{p_m}/R-1)} \right) \left(\frac{\mu_m(C_{p_m}-R)}{M_m} \right), \quad (10)$$

168 where k_m is the thermal conductivity of the vapours, C_{p_m} is the heat capacity of the vapours, which is
 169 calculated on a mass fraction average, i.e. $\sum w^i C_p^i$. Individual species heat capacities are given in
 170 Table 3. These values are obtained from Reid et al. [39] and Stull et al. [40].

171 The heat of vaporization for the each chemical species within the vapour is estimated based on the law
 172 of corresponding states. The relationship of the heat of vaporisation with acentric factor, ω^i , and the
 173 reduced temperature, T_r^i , is shown in eq. 11 is an analytical representation of the Pitzer's [41]
 174 correlation.

$$175 \quad \Delta H_v^i = \left(7.08 (1 - T_r^i)^{0.354} + 10.95 \omega^i (1 - T_r^i)^{0.456} \right) R T_c^i. \quad (11)$$

176 In this work, the bio-oil is treated as a homogeneous compound and hence its composition is not
 177 varied spatially or temporally. Representative bio-oil properties were sourced from the works of
 178 Oasmaa and Peacocke [42, 43]. The bio-oil properties are shown in Table 4.

179 [It has to be noted that the pyrolysis vapour and liquid bio-oil properties are subjected to errors](#)
 180 [associated with the estimation techniques and experimental values used for their computation.](#)
 181 [However, there is great confidence that the deviations from reality will not significantly affect the](#)
 182 [final results of the numerical model as the previously mentioned correlations and experimental values](#)
 183 [have been widely used and accepted by the chemical industry for several years.](#)

184 **5. Fluid dynamics model**

185 The fluid dynamics modelling approach is based on the Eulerian multiphase model to solve two
 186 phases namely gaseous pyrolysis vapours phase and condensed bio-oil liquid phase. In order to
 187 capture the detailed chemical composition variance within the vapours across the system, the species
 188 transport model was enabled in the vapour phase, while the composition of the liquid bio-oil is
 189 considered constant. To simulate the pyrolysis vapour condensation process within the condenser, the
 190 commercial CFD package ANSYS Fluent 13 has been utilised as the computational platform. The
 191 governing equations are as follows:

192 Continuity equation for phase p

$$193 \quad \frac{1}{\rho_{rp}} \left(\frac{\partial}{\partial t} (a_p \rho_p) + \nabla \cdot (a_p \rho_p \mathbf{v}_p) \right) = \dot{m}_{qp} - \dot{m}_{pq}. \quad (12)$$

194 The term ρ_{rp} is the volume averaged density of the p^{th} phase. In the above equation, the mass source
 195 terms \dot{m}_{pq} & \dot{m}_{qp} corresponds to mass transfer from phase p to phase q and vice versa. The mass
 196 transfer from vapour phase to the bio-oil phase corresponds to the sum of the individual species mass
 197 transferred to the bio-oil. This is calculated based on the vapour liquid equilibrium (VLE) condition as
 198 mentioned in section 3.

199 Momentum equation for phase p

$$200 \quad \frac{\partial}{\partial t} (a_p \rho_p v_p) + \nabla \cdot (a_p \rho_p v_p v_p) = -a_p \nabla P + \nabla \cdot \bar{\tau}_p + a_p \rho_p \bar{g} + \bar{R} + \bar{M}_{s,p}. \quad (13)$$

201 In the above equation, stress-stain tensor is represented by $\bar{\tau}_p$ and \bar{R} is the interaction force between
 202 the two phases calculated by

$$203 \quad \bar{R} = K_{pq} (\bar{v}_q - \bar{v}_p). \quad (14)$$

204 The interphase momentum exchange coefficient K_{pq} defined as

$$205 \quad K_{pq} = \frac{a_p a_q \rho_p q f}{\tau_{pq}}. \quad (15)$$

206 In this study, the drag function f used is based on Schiller-Naumann drag model [44] and is defined
 207 as $C_D Re/24$. The drag coefficient C_D is given by eq. 16 and the relative Reynolds number Re is
 208 given in eq. 17.

$$209 \quad C_D = \begin{cases} 24 (1 + 0.15 Re^{0.687}) / Re & Re \leq 1000 \\ 0.44 & Re > 1000 \end{cases} \quad (16)$$

$$210 \quad Re = \frac{\rho_{pq} |\bar{v}_p - \bar{v}_q| d}{\mu_{pq}} \quad (17)$$

211 The particulate residence time τ_{pq} used in eq. 15, is defined as

$$212 \quad \tau_{pq} = \frac{\rho_{pq} d^2}{18 \mu_{pq}}. \quad (18)$$

213 The momentum source $\bar{M}_{s,p}$ is calculated based on the mass exchanged between the phases i.e. from
 214 vapour phase to bio oil phase as shown in eq. 19

$$215 \quad \bar{M}_{s,p} = \dot{m}_{qp} (\bar{v}_q - \bar{v}_p). \quad (19)$$

216 Here \dot{m}_{pq} is equal to the sum of all individual species mass sources condensed to form the bio-oil and
 217 is computed as $\dot{m}_{pq} = \sum_i \dot{m}_c^i$.

218 In order to solve the conservation equations for individual chemical species within the vapour phase,
 219 convection-diffusion equation of the i^{th} species as shown in eq. 20 is used.

$$220 \quad \frac{\partial}{\partial t} (\rho w^i) + \nabla \cdot \rho \bar{v} w^i = -\nabla \cdot \bar{J}^i + S^i. \quad (20)$$

221 The diffusion flux \bar{J}^i of the component i is computed based on Fick's law which states that mass
 222 diffusion is due to concentration gradients.

223 The energy conservation for phase p is given as

224
$$\frac{\partial}{\partial t} (a_p \rho_p h_p) + \nabla \cdot (a_p \rho_p \bar{v}_p h_p) = -a_p \frac{\partial P_p}{\partial t} + \bar{\tau}_p : \nabla \bar{v}_p - \nabla \cdot \bar{q}_p + Q + H_{s,p}. \quad (21)$$

225 In eq. 21, q_p is the heat flux and Q is the volumetric rate of energy transfer between two phases
226 defined by

227
$$Q = h_{pq}(T_q - T_p). \quad (22)$$

228 The heat transfer coefficient h_{pq} between two phases was estimated based on the Ranz-Marshall
229 correlation [45]. The heat source due to phase change $H_{s,p}$ mentioned in eq. 21 is computed by

230
$$H_{s,p} = \begin{cases} \sum_i (-\dot{m}_c^i H_p^i) & \text{for vapour phase} \\ \sum_i \dot{m}_c^i (H_p^i - \Delta H_v^i) & \text{for bio - oil phase} \end{cases} \quad (24)$$

231 The terms H_p^i & ΔH_v^i are the enthalpy and latent heat of vaporisation of the species i . The standard k- ϵ
232 turbulence model was utilised to capture the turbulence effects within the system.

233 6. Assumptions

234 The implementation of the hydrodynamic model is based on the following assumptions.

- 235 I. The pyrolysis vapours together with the carrier gas nitrogen is treated as an ideal mixture.
236 This is mainly due to the unavailability of the excess function data in the literature.
- 237 II. The density of the species over the computational domain was calculated based on the ideal
238 gas assumption. However, while estimating the vapour pressures, and critical properties such
239 as viscosity and thermal conductivity, real gas behaviour was considered.
- 240 III. The pyrolysis vapours are modelled with 11 chemical species and is assumed to represent the
241 majority of its behaviour in terms of critical properties. This is a compromise between
242 accuracy and the speed of the solution. In reality, pyrolysis vapours consist of more than 100
243 chemical compounds something that dramatically increases the computational expense of the
244 model. **However**, the model presented in the present paper **can be** readily scalable to different
245 species groups based on the feedstock used for pyrolysis.
- 246 IV. Fugacity coefficients are assumed as 1. This assumption can be justified when the system is
247 not under high pressures.
- 248 V. The non-condensable gases obtained from the mass balance done during the experiment are
249 modelled as Nitrogen and clubbed with the carrier gas composition. The very low
250 concentration of the non-condensable gas fraction produced during biomass pyrolysis, is not
251 expected to significantly influence the thermodynamic and fluid dynamic behaviour of the
252 system.

253 **7. Results & Discussions**

254 The geometry of the condenser used in the current CFD model is the same as the double surface
255 condenser used in the pyrolysis experiments [31] .

256 Fig. 2(b) shows the volume fraction of the bio oil at the outer tube and inner tube surfaces of the
257 condenser. The contours of the bio-oil volume fractions on the vertical section of the condenser are
258 presented in Fig. 2(a). From this figure, it is evident that the condensation is more intense at the
259 surfaces at which the cooling water is in contact with. It also demonstrates the bio-oil droplet
260 accumulation on the condenser during the condensation process. The dynamics of the bio-oil mist is
261 highly influenced by the gas flow within the condenser. This is evident from the contours on the inner
262 tube surface volume fraction as they can be seen in Fig. 2(b), where a ripple like formation is
263 observed. In the case of the outer tube, the gas flow at the bottom side forces the bio-oil droplets into
264 the central zone of the annular section.

265 In Fig. 3, the contours of the gas temperature are shown, where it can be observed that the gas
266 temperature is lower at the wall surfaces than in the middle zone of the annulus space between the
267 inner and the outer tubes. The liquid bio-oil droplet formation discussed in the previous section is
268 mainly due to this particular temperature profile of the gas. To have a clearer picture of the
269 temperature variation along the length of the condenser, the average temperature, from the inlet to the
270 outlet, is plotted in the same figure. The rapid vapour temperature drop can be seen in the same plot,
271 where the inlet vapour temperature is approximately 500°C and the outlet temperature approximately
272 18°C. It can be observed that the most significant region for vapour cooling is located between ≈ 0.02
273 $- 0.07\text{m}$ of the length of the condenser.

274 Fig. 4 shows the variation of the relative saturation of the different compounds along the length of the
275 condenser. The dashed lines represent the maximum and minimum values at the different sections,
276 while the continuous line represents the volume averaged compound saturation at each particular
277 section of the condenser.

278 A compound will change phase (condense) when its relative saturation exceeds unity. From Fig. 4, it
279 can be seen that the maximum relative saturation for Butyric Acid, Coniferyl Alcohol, Guaiacol,
280 Phenol and water reaches its maximum value before 0.1 m of length, while the curve relaxes towards
281 its equilibrium value as the vapours continue to condense beyond this region. The maximum relative
282 saturation occurs close to the wall boundaries due to the significantly lower temperatures at those
283 points. The minimum relative saturation values are mainly located towards the centre of the annular
284 section where temperatures are higher. The volume averaged relative saturation line represents the
285 volume weighted average of the relative saturation at different sections of the condenser. It shows that
286 the average relative saturation along the condenser has an increasing trend (approaching unity) due to
287 the rapid cooling of the vapours and gives a very good indication of the bio-oil composition at the first

288 stage of condensation. One can see that for the components that the average saturation has exceeded
289 unity, a significant proportion of them will be collected in the form of liquid bio-oil at the first stage.
290 For some of the components with low initial partial pressures, as shown in Fig. 5 (e.g. propionic acid),
291 only small traces will be collected that are mainly determined by the maximum relative saturation. For
292 those components that none of the maximum and consequently none of the average and minimum
293 saturations have exceeded unity, no traces will be detected in the final bio-oil product.

294 The relative mass fraction contours of each compound are shown in Fig. 6. These contours are plotted
295 at the vertical mid-section of the condenser. At the inlet, all the values are equal to unity. As the
296 vapours flow through the condenser, the values of the relative mass fraction decrease or increase
297 depending on whether the compound within the condenser changes its phase to liquid bio-oil or not.
298 Those compounds with maximum relative saturation above unity within 0.1 m of length keep
299 condensing as the vapours flow along the length of the condenser. The relative mass fractions of those
300 decrease continuously. The blue colour at the outlet indicates that a particular compound is
301 completely condensed. Light green colour indicates that those components have been partially
302 condensed within the condenser, whereas the red colour indicates that those compounds have been
303 slightly or not been condensed at all.

304 Another interesting point that can be observed in Fig. 4 is that the values of the relative saturation
305 increase sharply for many components at around 0.18m. This is primarily attributed to the rise in gas
306 pressure near the exit of the condenser due to the diffuser effect at the point where the inner tube ends.
307 As the inner tube ends, the cross sectional area increases steadily over a small distance which
308 effectively gives a small rise in pressure. The maximum velocity of the vapour and the average
309 pressure within the condenser are plotted in Figs 7 and 8 respectively. From Fig. 8, it is evident that at
310 lower temperatures, a small increase in pressure highly increases the value of the relative saturation of
311 the components. The relative saturation values shown in Fig. 4 indicate that at higher temperatures
312 condensation is primarily affected by heat transfer rates whereas at later stages, where the temperature
313 is low, they highly depend on pressure changes.

314 Fig. 9 shows the plot of cumulative mass source ratio along the direction of the flow. This is the ratio
315 between the cumulative mass source of the particular component and its mass fraction at the inlet.
316 When the value of this ratio approaches unity, it indicates the complete conversion of that particular
317 component. From Fig. 9 we can see that Guaiacol is the first component condensed completely
318 followed by Coniferyl Alcohol and Phenol. It is also worth noticing that Butyric Acid and water have
319 been partially condensed within the condenser. Approximately 35% of water vapour is condensed in
320 the bio-oil. Small traces of condensation can be seen at the end of the tube for the propionic acid. Fig.
321 10 shows the total mass source plot within each segment of the condenser along its length from the
322 inlet to the outlet. The condenser has been divided into 100 equal segments along its axis and the total
323 mass fraction shown in Fig. 10 is summed in all the cells within the individual segments. As expected,

324 mainly due to the high initial concentration, the mass fraction source of Coniferyl Alcohol is the
325 greatest among all components, followed by Guaiacol. From Fig. 10 we can see that the condensation
326 of Guaiacol, Coniferyl Alcohol and Phenol starts at almost the same location in the condenser
327 (approximately 0.025 m from the inlet). The condensation of components like water and Butyric acid
328 starts at approximately 0.05m from the inlet. In the case of water, a considerable amount is transferred
329 from the vapour phase to liquid bio-oil phase near the outlet region. This is mainly due to increased
330 pressure at lower temperatures. From Fig. 10 we can safely conclude that most of the condensation
331 takes place between 0.025 to 0.1 m of the condenser.

332 The reduced temperature variation of all components within the vapour phase can be seen in Fig. 11.
333 The reduced temperature, which represents the ratio between the vapour temperature and the
334 corresponding component's critical temperature, varies between 1.5 and 0.4. More importantly
335 between the lengths 0.025 m and 0.1 m where maximum condensation taking place, the reduced
336 temperatures vary between 1.0 and 0.6. This is an essential condition for using the enthalpy of
337 condensation relationship mentioned in eq. 11. The enthalpy of condensation for each condensed
338 components is plotted separately in Fig. 12. The continuous line represents the total enthalpy of
339 vaporisation within each axial segment of the condenser. The plot follows a similar trend as mass
340 sources plot shown in Fig. 10. The enthalpy of vaporization values are embedded into the solver as
341 energy source terms and are removed from the bio-oil phase. Dotted lines represent the maximum
342 value of the enthalpy of condensation within a particular segment. The maximum enthalpy of
343 condensation both in terms of total and maximum value is observed for Coniferyl Alcohol and
344 followed by Guaiacol. One order of magnitude lower values are observed in the case of water and
345 Phenol in comparison to Coniferyl alcohol. The rest of the acids contributed relatively negligible
346 amounts of enthalpy towards the outlet region of the condenser.

347 The gas velocity vectors inside the condenser can be seen in Fig. 13. Velocity varies from 3.5 m/s at
348 the entrance to 1.5 m/s at the exit. The maximum velocity throughout the length of the condenser is
349 approximately 1 m/s. From the vector plot in Fig. 13, we can see that some small recirculation zones
350 are formed near the entrance. Near the outlet we can see a small drop in the velocity followed by a
351 sharp increase in it; this is also evident in Fig. 7. This phenomenon can be attributed to the sudden
352 expansion of the cross-sectional area at the end of the inlet tube, which at the same time induces some
353 gas recirculation at that region, followed by another contraction at the very end of the condenser.
354 Overall, the vapour residence time is well below 1 s. This implies that a rapid condensation taking
355 place within the condenser, thus greatly minimising the possibility of secondary vapour cracking in
356 the condenser.

357 From the experiments it has been observed that the exit temperature is around 23°C whereas in the
358 numerical simulation an average outlet temperature of approximately 18°C is predicted. The
359 numerical simulations slightly over predict the heat transfer as the Ranz-Marshall correlation assumes

360 that the secondary phase forms spherical droplets in the primary one. Moreover, the temperature at the
361 outlet varies spatially over the entire section so it was not clear at what location the actual
362 measurement has been taken. Fivga's [46] experiments, in which the same equipment has been used,
363 also showed that the water vapour partially condensed in this stage of the liquid collection system. In
364 the experiments, the condensed water percentage varies between 30 to 45% of the total water content.
365 It is also mentioned that the pH value of the first stage condensed bio-oil is higher than the subsequent
366 collection stages. This was primarily due to the condensation of the acidic components at the later
367 stages. In this numerical study, we also observed the similar trend where acids like formic acid and
368 acetic acid are not condensed in the present condenser.

369 It is worth commenting that the vapour thermochemical properties and in turn the condensation
370 patterns of it will vary if a different initial vapour composition is used. In the real cases, the bio-oil
371 and pyrolysis vapour composition is much more complex than the one described in this work.
372 However, the numerical predictions of the present simulation showed a very good agreement with the
373 experimental results. Based on the type of biomass and the type of compounds present in it, the
374 composition can be modified or further simplified. This can be precisely done by considering the
375 specific components of interest, whereas the thermodynamic properties of the composition can be
376 estimated accordingly. In this way, this condensation model can be utilised during the designing stage
377 of biomass specific or function specific heat exchangers/condensers.

378 It has to be noted that the obtained results from this work are specific to the indirect contact heat
379 exchanger under study and cannot be directly extrapolated to other types of condensers. Each type of
380 condenser possesses its own heat transfer and fluid dynamic characteristics which will in turn affect
381 the equilibrium properties of the vapour compounds. In addition to that, scaling up of the equipment
382 will also have a similar impact since the thermal and flow field inside the condensing equipment will
383 be affected by the size alteration. However, the methodology presented in this model can be directly
384 utilised for the design and optimisation of different types and sizes of condensers used in the bio-oil
385 liquid collection systems.

386 **8. Conclusions**

387 A species transport model has been implemented within the Eulerian multiphase approach to model
388 the fractional condensation of bio-oil. The generalised corresponding states method has been used to
389 estimate the saturation vapour pressure of the individual components. In this study, 11 discrete
390 chemical compounds were selected to represent the pyrolysis vapours composition, together with
391 Nitrogen which represents the carrier gas and the non-condensable fraction. The mixture of pyrolysis
392 vapours was treated as an ideal gas mixture. From the simulations, it was observed that only few
393 components condensed completely in this stage of the condenser something that it is in good
394 agreement with the experimental observations. According to Fivga's [46] experimental study, only 30%

395 of the water vapour was condensed. Guaiacol, Coniferyl Alcohol and Phenol components were
396 completely condensed within the first half of the length of the condenser, whereas no traces of formic
397 and acetic acid condensation were detected. It is also observed that the marginal increase in the
398 pressure at the lower temperature towards the outlet of the condenser resulted in the increase of the
399 relative saturation of water and other acidic components. The reduced temperatures within the
400 condensing region for most of the components fall in the range of 1 to 0.5, which is the recommended
401 range for using the Pitzer correlation for estimating the enthalpy of condensation.

402 **9. Acknowledgement**

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406 **Nomenclature**

407 ***Latin symbols***

408	C_{p_m}	Mixture heat capacity J/kg K
409	f	Drag function
410	f_0, f_1	Functions in the three parameter corresponding state equation
411	f_l^i	Liquid fugacity, Pa
412	f_v^i	Vapour fugacity, Pa
413	g	Gravitational acceleration, m/s ²
414	h	Specific enthalpy of the phase, J/kg
415	H_p^i	Enthalpy of the species
416	$H_{s,p}$	Latent heat source, W/m ³
417	H_v^i	Heat of vaporisation or latent heat, J /kg
418	k_m	Mixture thermal conductivity W/ m K
419	K_{pq}	Interphase momentum exchange coefficient, kg/m ³ s
420	M^i	Mole fraction, g/mol
421	M_m	Mixture molecular weight, g/mol
422	$\bar{M}_{s,p}$	Momentum source vector, N/m ³
423	\dot{m}_c^i	Mass condensed, kg/m ³ s
424	\dot{m}_{qp}	Mass transfer rate between phase q to phase p, kg/m ³ s
425	P, p	Pressure, Pa
426	P^i	Partial pressure, Pa
427	P_c^i	Critical pressure, bar
428	P_{cm}	Mixture critical pressure, Pa
429	P_r^i	Reduced saturation pressure
430	q	Heat flux, W/m ²
431	Q	Volumetric rate of energy transfer, W/m ³
432	\bar{R}	Interaction force vector, N/m ³
433	R	Universal gas constant, J/ mol K , atm cm ³ / mol-K (in eq. 9)
434	Re	Reynolds number
435	S^i	Species source, mol/ m ³
436	t	Time, s

437	T	Temperature, K
438	T_b^i	Boiling temperature, K
439	T_c^i	Critical temperature, K
440	T_r^i	Reduced temperature
441	T_{rm}	Mixture reduced temperature
442	V_c^i	Critical volume, cm ³ /mol
443	y^i	Mass fraction
444	Z_c^i	Critical compressibility factor

445 ***Greek symbols***

446	a	Volume fraction
447	γ	Universal coefficients used in f_0, f_1 functions
448	μ	Dynamic viscosity, Pa – s
449	μ_m	Mixture viscosity, Micro Poise (μP)
450	v	Velocity vector, m/s
451	ξ_m	Inverse viscosity, μP^{-1}
452	ρ	Density, kg/m ³
453	ρ_{rp}	Volume averaged density, kg/ m ³
454	σ	Surface tension, N/m
455	τ	Particulate relaxation time, s
456	τ^i	Inverse of the reduced temperature
457	$\bar{\tau}$	Stress tensor, N/m ²
458	ϕ^i	Fugacity coefficient
459	ϕ_{sat}^i	Fugacity coefficient at saturation condition
460	ω^i	Acentric factor

461 ***Subscripts***

462	b	Properties at boiling point
463	c	Critical properties
464	g	Gas
465	l	Liquid
466	m	Vapour mixture

467	p, q	Phase index
468	pq	Volume averaged properties
469	r	Reduced properties
470	s	Source term
471	sat	Values at saturation point
472	v	Vapour
473	<i>Superscripts</i>	
474	i	species index in the vapour mixture

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Table 1: Chemical compounds in the pyrolysis vapours and their properties

Chemical compound	Initial Volume fraction *	Molecular weight (g/mol)	Critical Temperature (K)	Critical pressure (atm)	Critical volume (cm ³ /mol)	Acentric factor	Critical compressibility factor
Acetic acid	0.037	60.05	594	57.1	171	0.454	0.2
Butanal	0.109	72.11	524	40	278	0.352	0.26
Butyric acid	0.011	88.11	628	52	292	0.67	0.295
Coniferyl alcohol	0.19	180.2	569.9	33.6	482	1.155	0.346
Formic acid	0.042	46.02	580	57.34	120	0.368	0.1445
Guaiacol	0.108	124.14	696.8	46.613	338	0.563	0.275
Pentanal	0.021	86.13	554	35	333	0.4	0.26
Phenol	0.054	94.11	694.2	60.5	229	0.44	0.24
Propanal	0.144	58.08	496	47	223	0.313	0.26
Propionic acid	0.017	74.08	612	53	230	0.536	0.242
Water Vapour	0.267	18.01	647.3	217.6	56	0.344	0.229

*The initial volume fraction excludes the carrier gas Nitrogen.

Table 2: Coefficients of eqs. 2 and 3.

k	γ_k
1	-5.53357241
2	11.0210515
3	-0.51243147
4	-10.6722729
5	29.4364927
6	-0.44101891

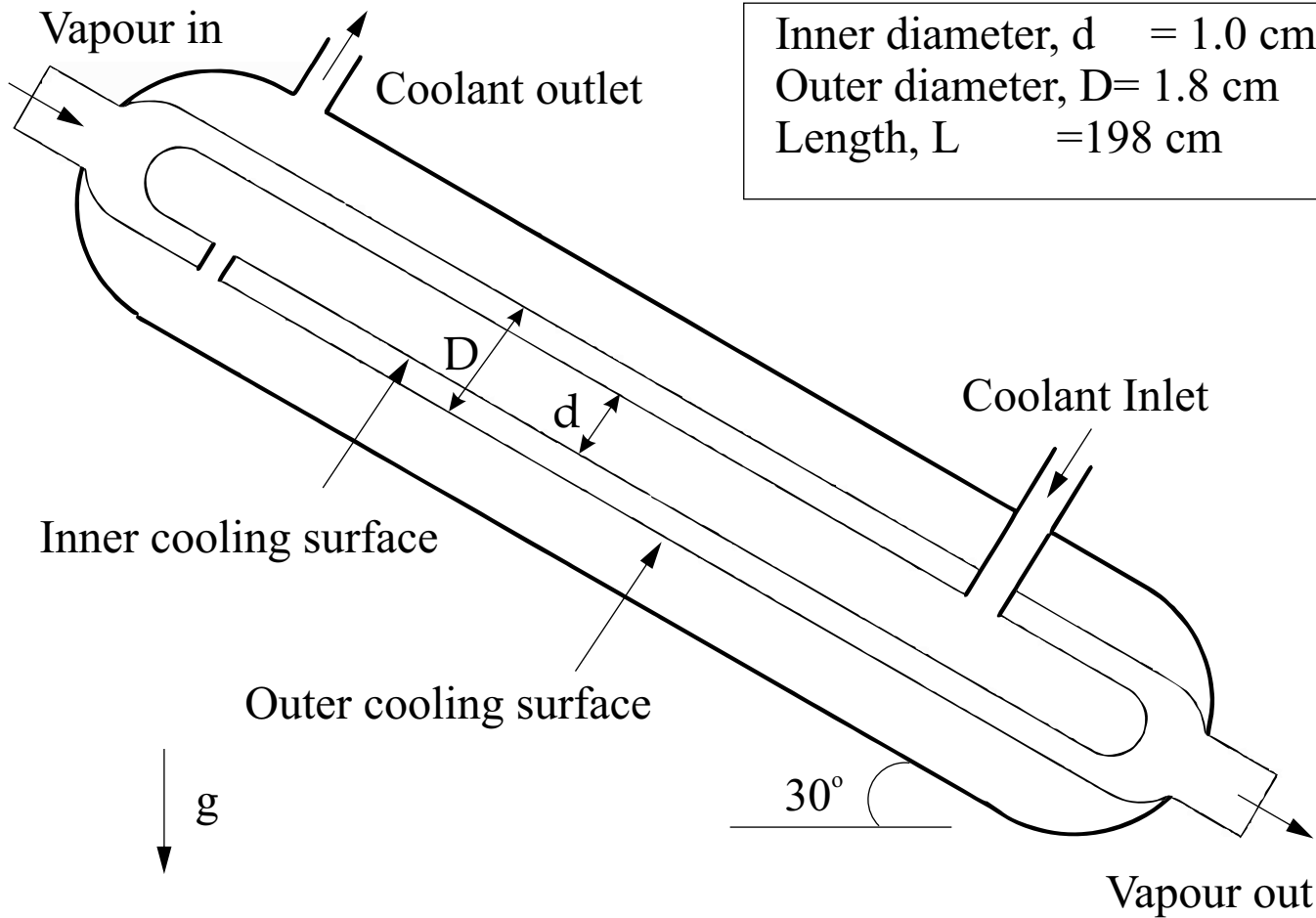
Table 3: Heat capacities of individual components present in the vapour

Chemical compound	$C_p = A_1 + A_2T + A_3T^2$		
	A_1	A_2	A_3
Acetic acid	195.74849	3.5237048	-0.001545339
Butanal	245.97362	4.4604585	-0.001734686
Butyric acid	229.03995	3.9854485	-0.001549761
Coniferyl alcohol	527.97236	3.1066709	-0.000768719
Formic acid	326.7	2.5160000	-0.00105
Guaiacol	531.24523	3.0758568	-0.000739824
Pentanal	202.39221	4.7575163	-0.001883003
Phenol	-158.75528	4.9638417	-0.002442437
Propanal	240.36658	4.2292475	-0.001671269
Propionic acid	164.9201	4.0156030	-0.001735477
Water Vapour	1779.0173	0.1717701	0.000362651

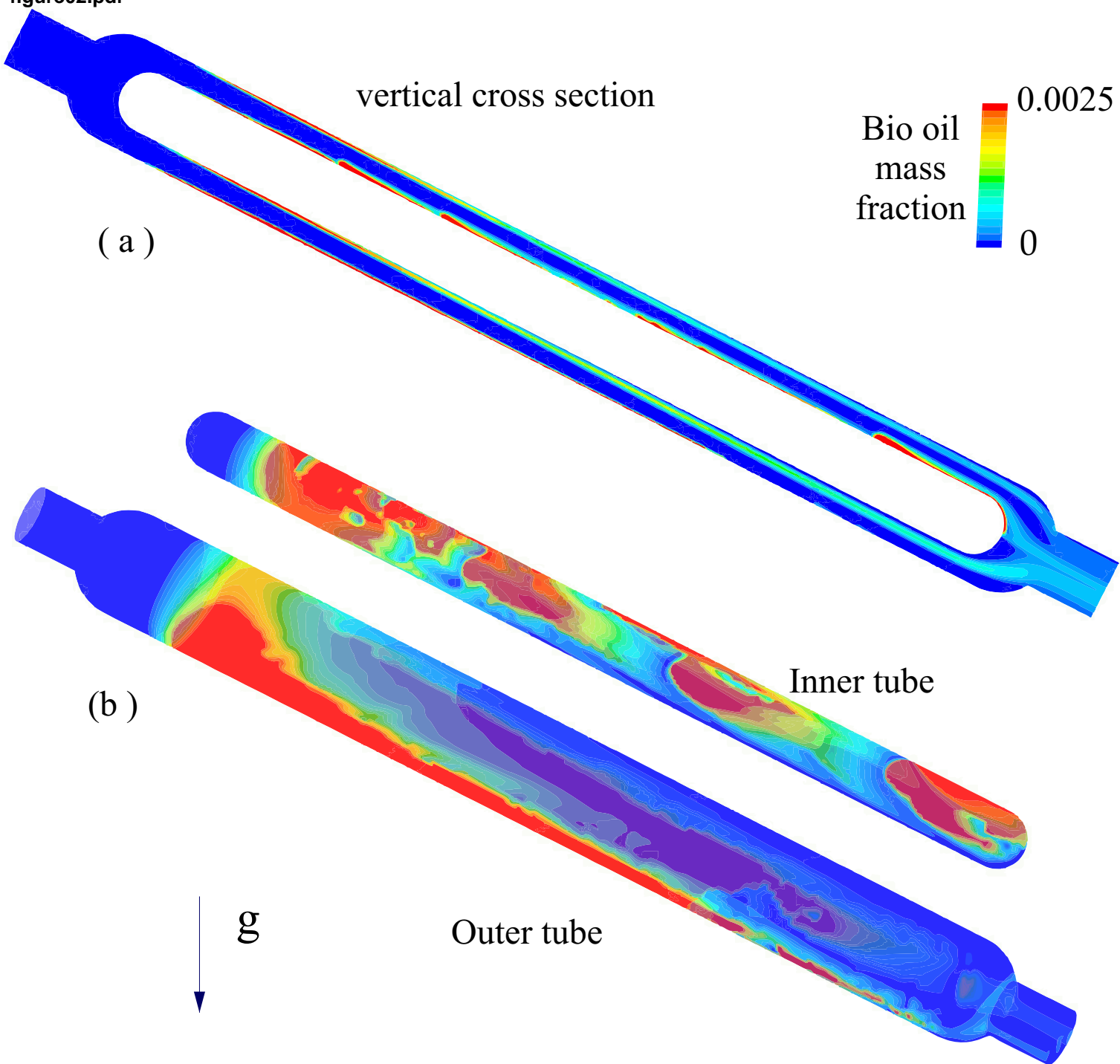
Table 4: Bio-oil properties

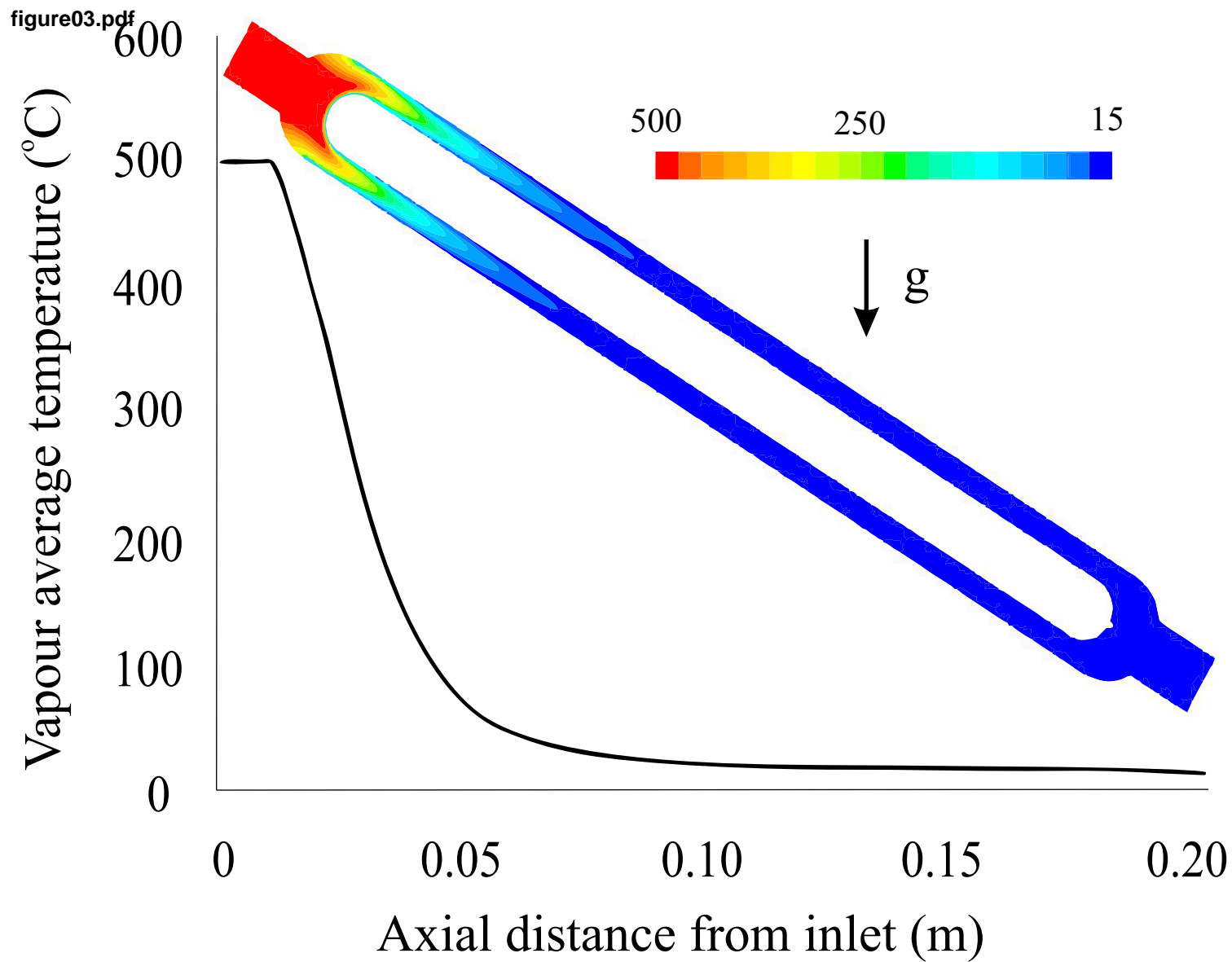
Property	Value
Density (kg/m ³)	1200
Thermal conductivity (W/m K)	0.386
Specific heat capacity (J/kg K)	3200
Viscosity (Pa s)*	$12.9881 - 0.080204 T + 0.000124 T^2$

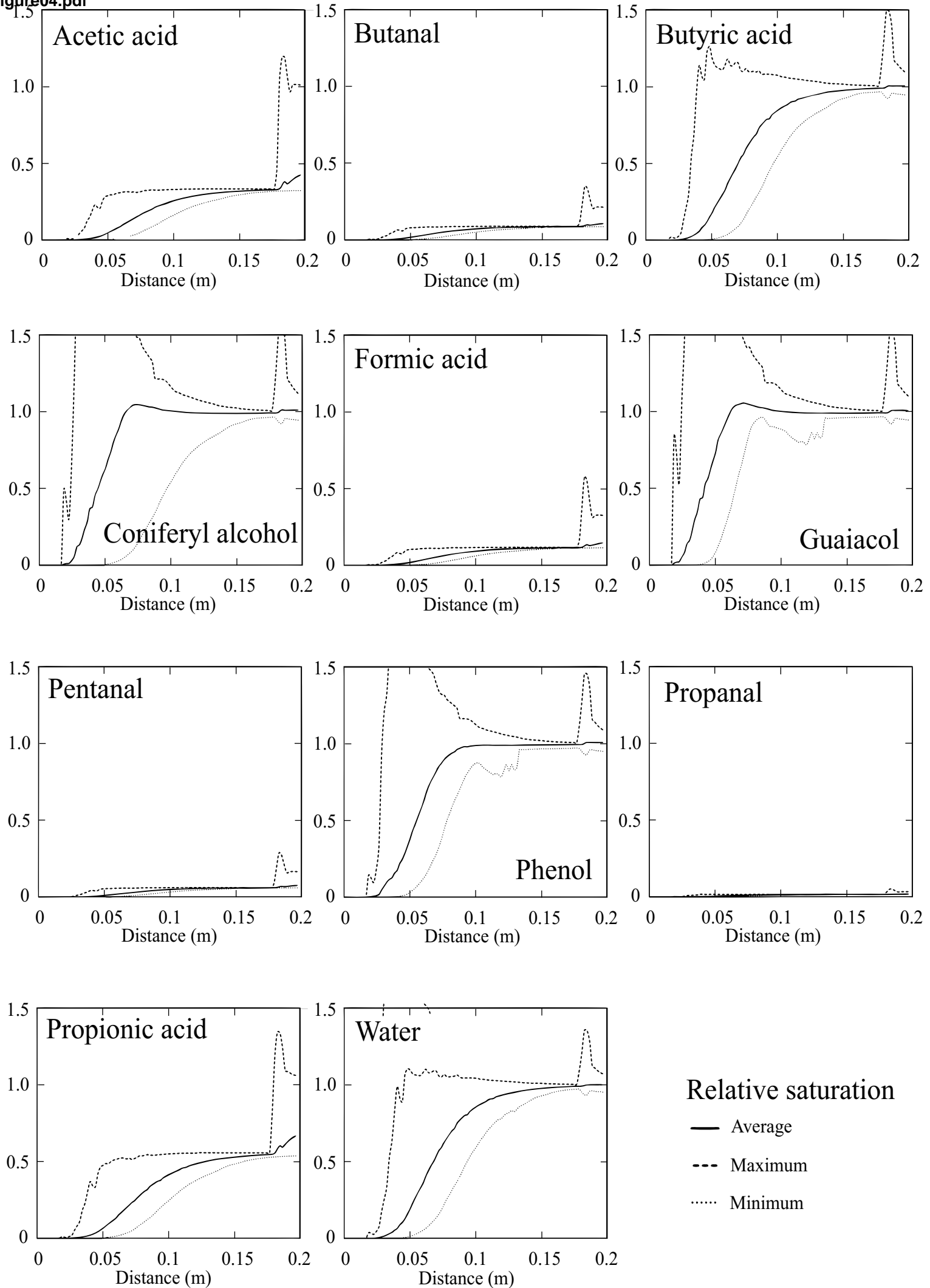
* Temperature unit is K.

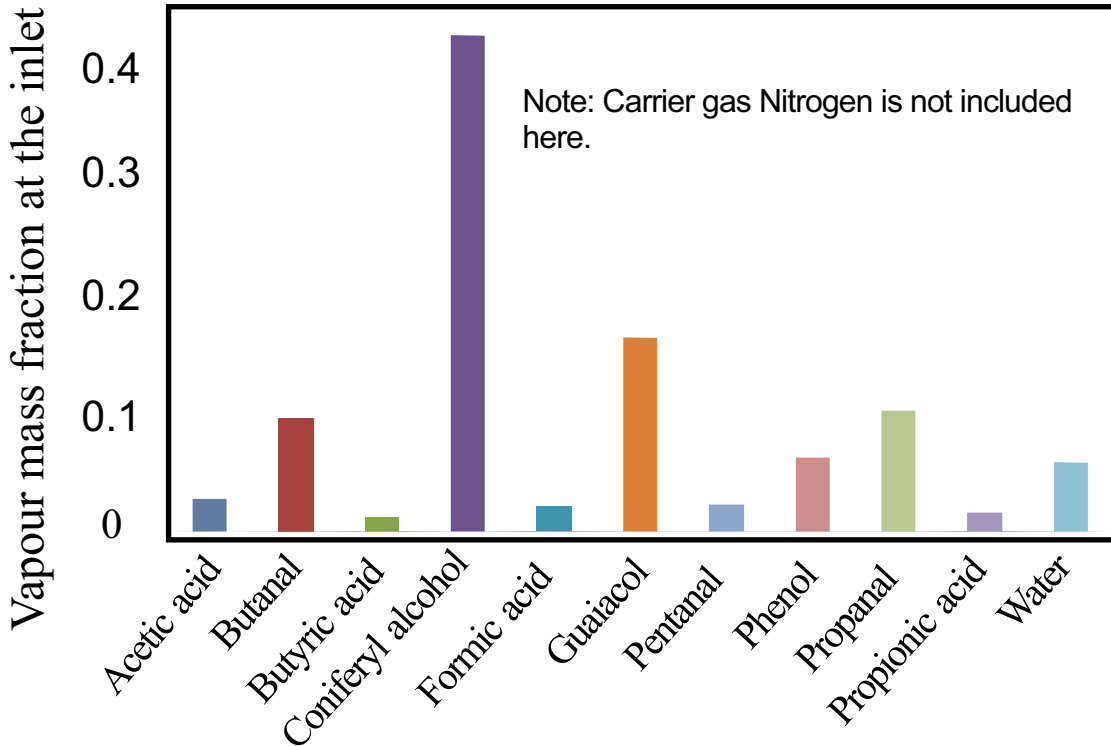


Inner diameter, $d = 1.0$ cm
Outer diameter, $D = 1.8$ cm
Length, $L = 198$ cm

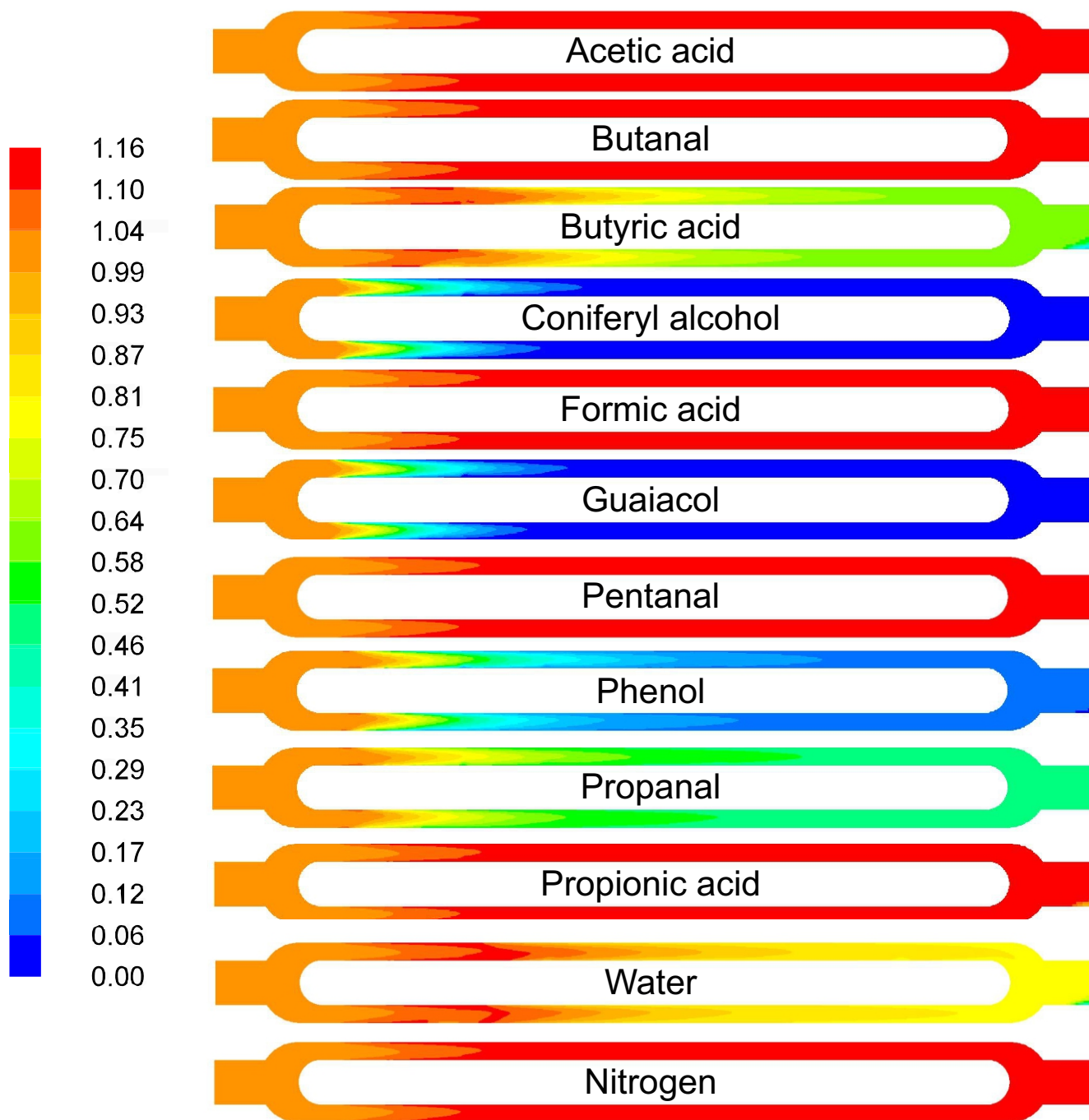


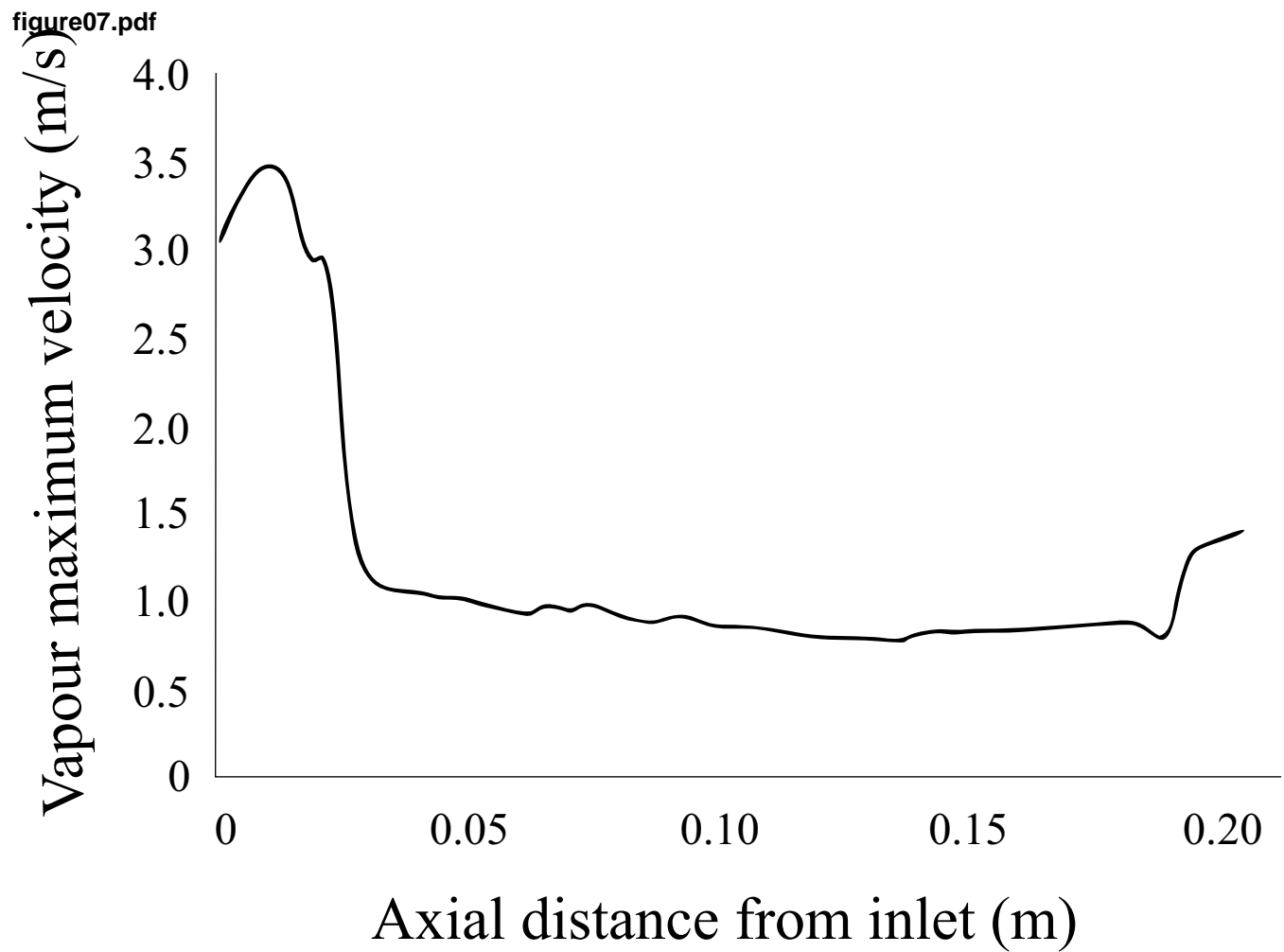


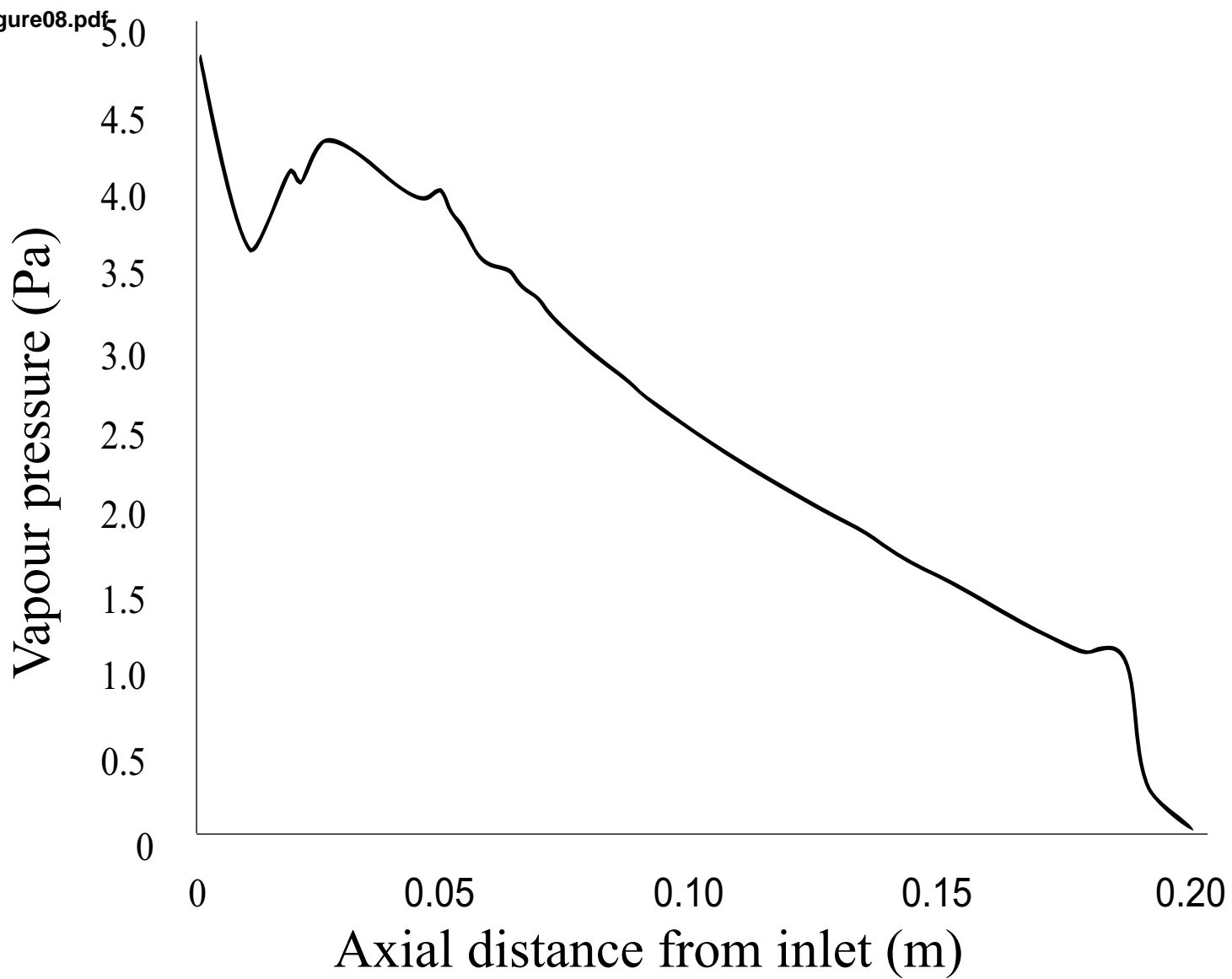


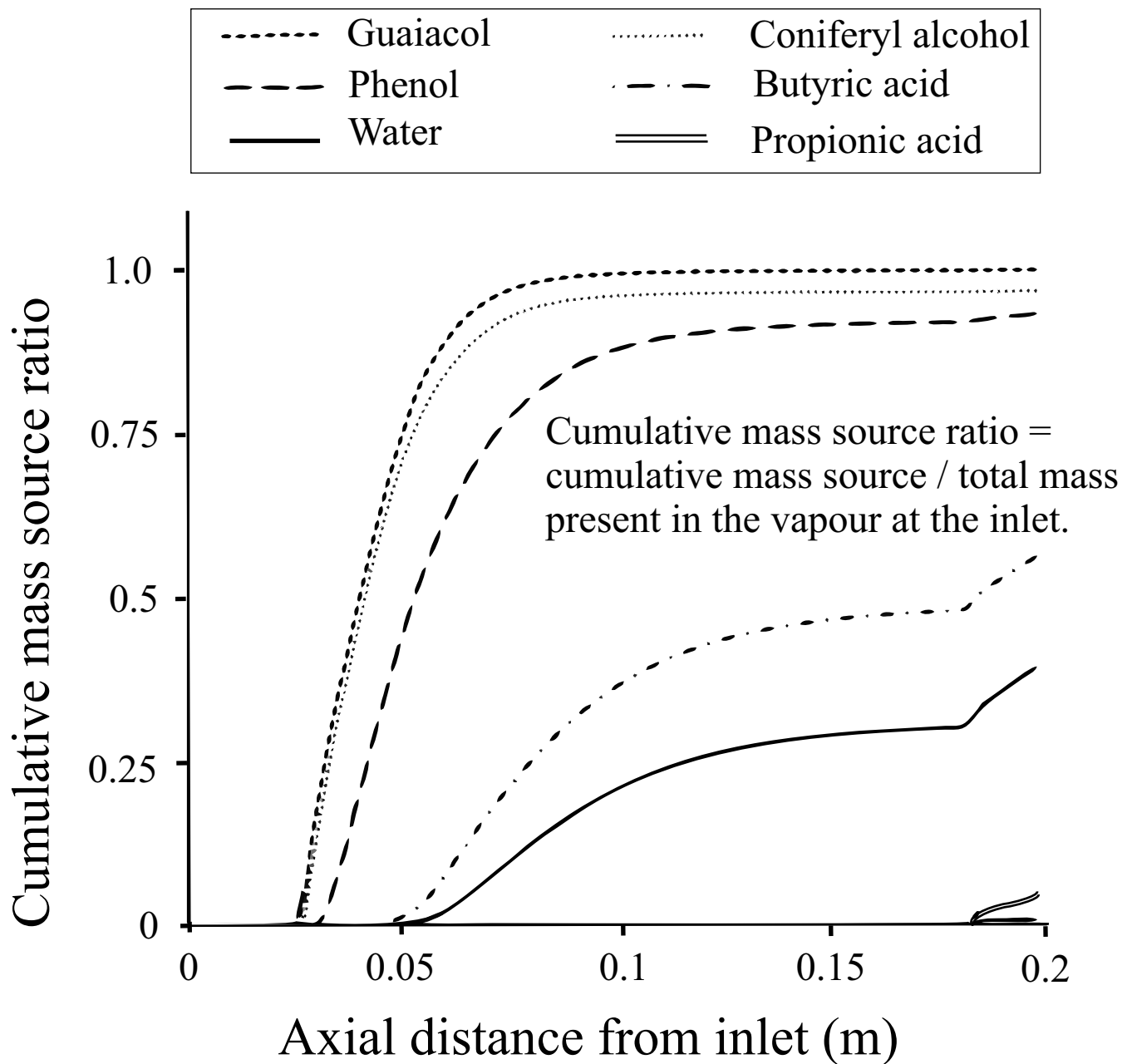


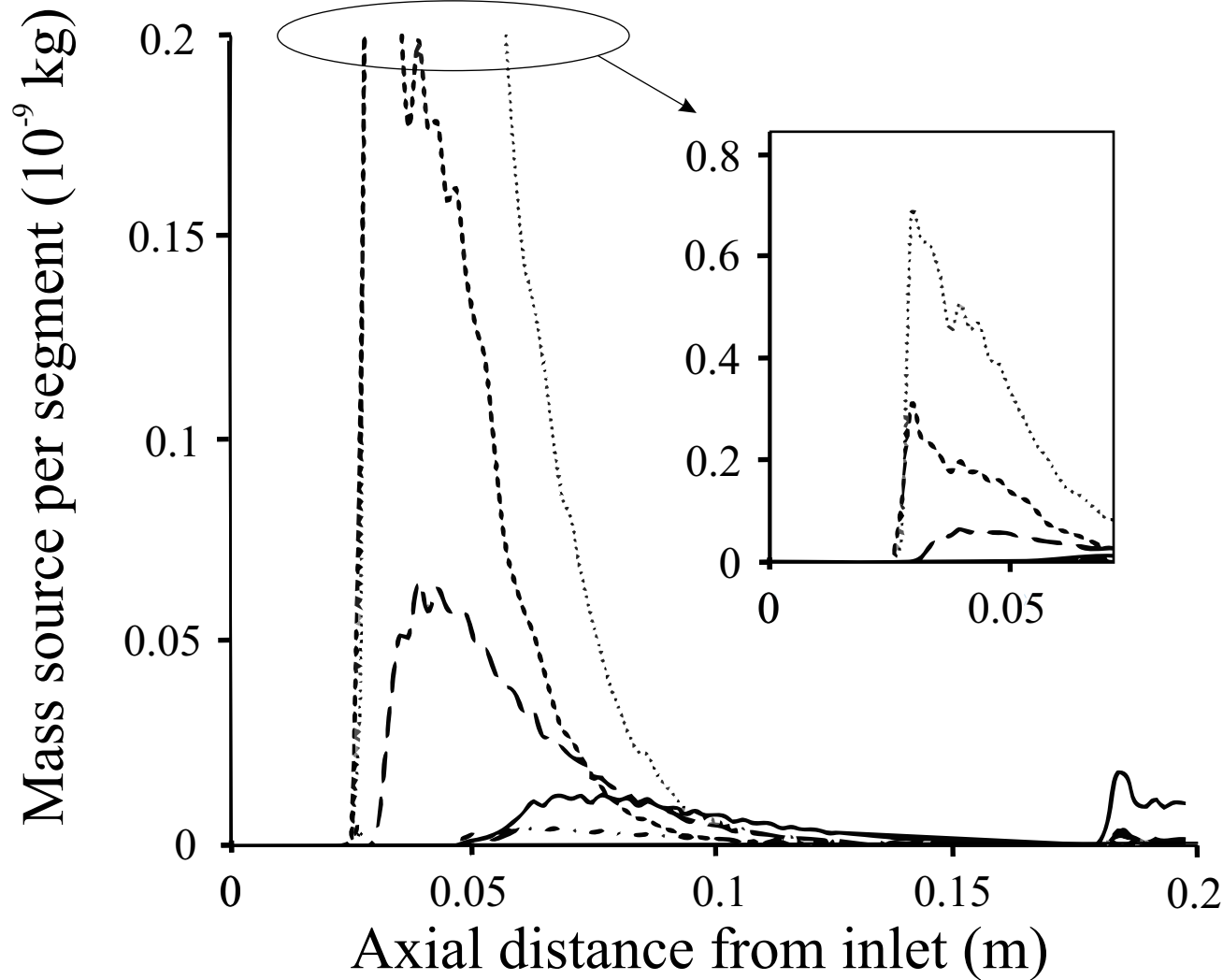
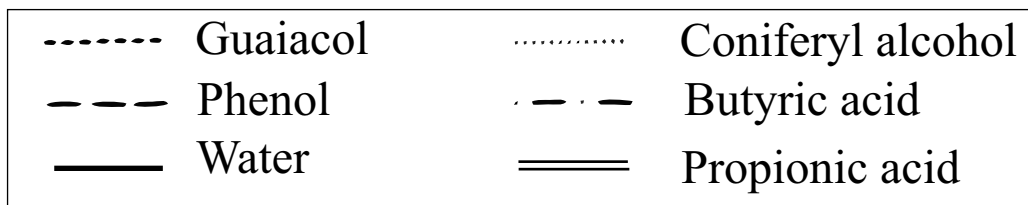
Relative mass fraction compared to the inlet mass fractions.

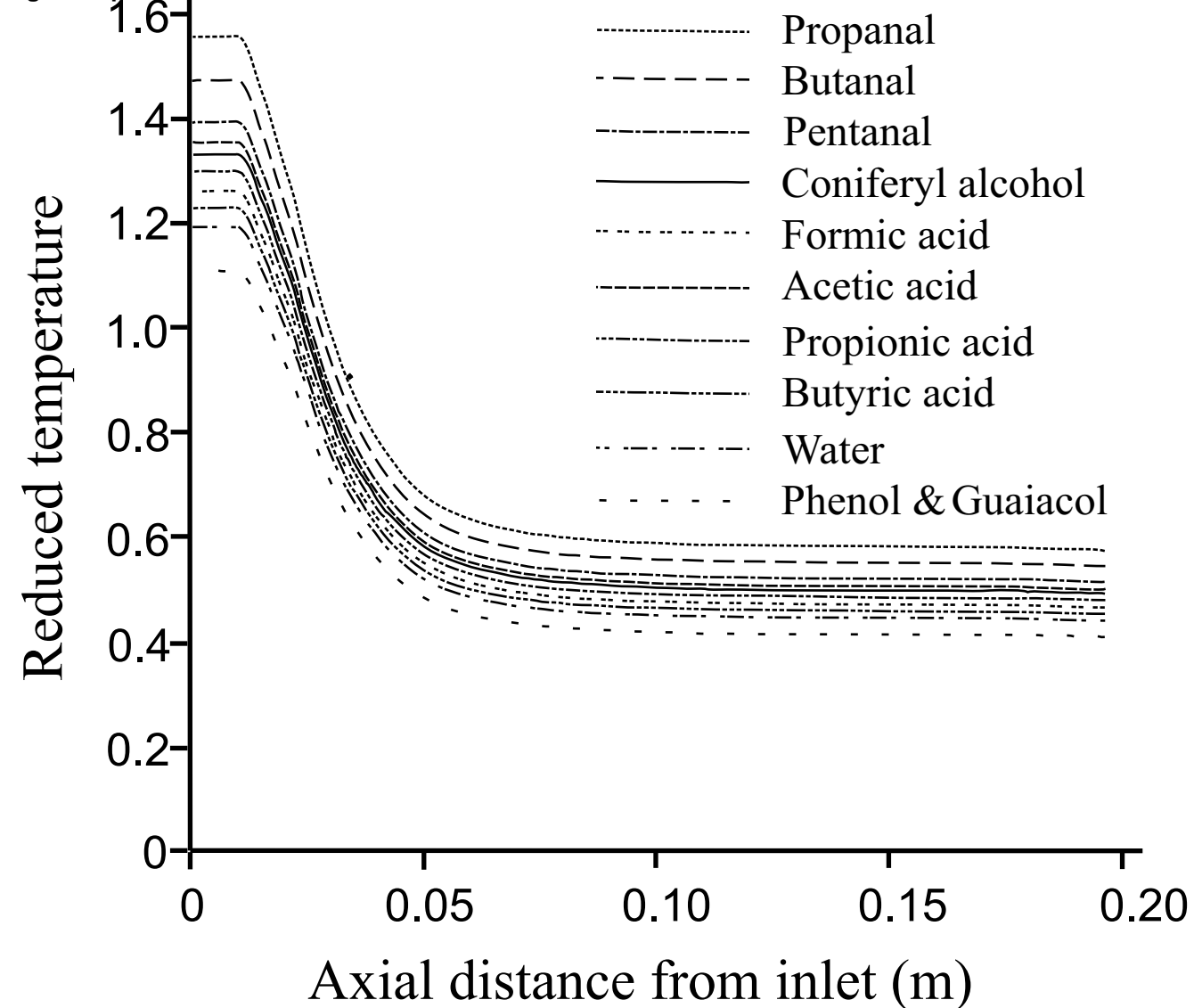


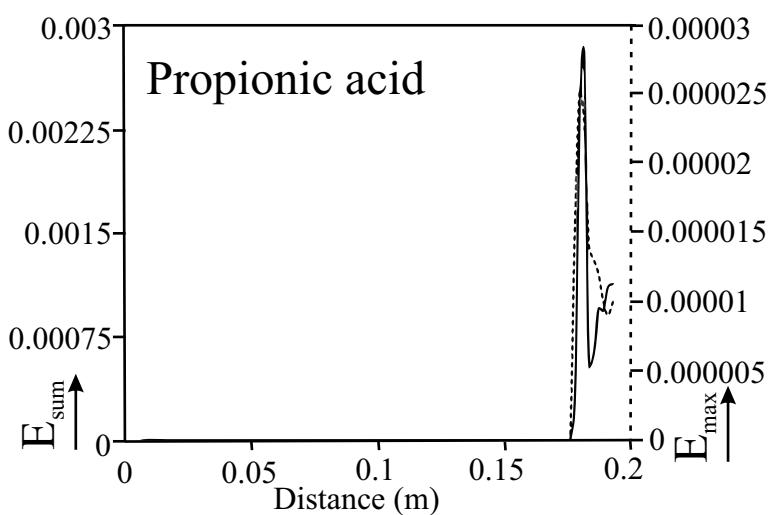
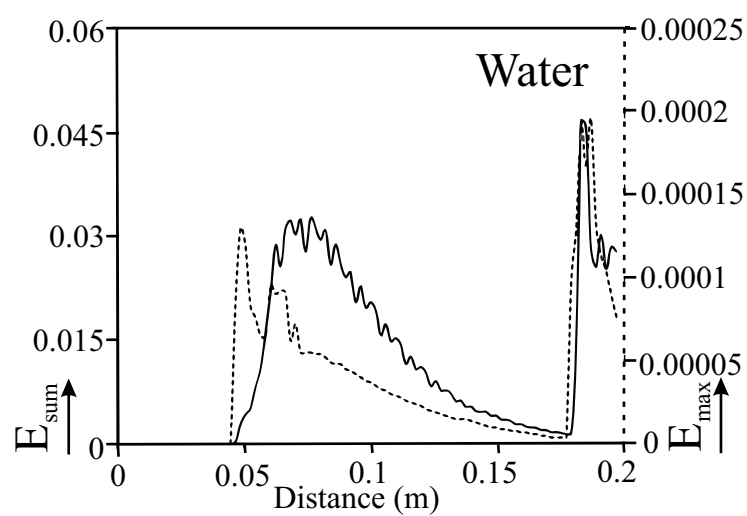
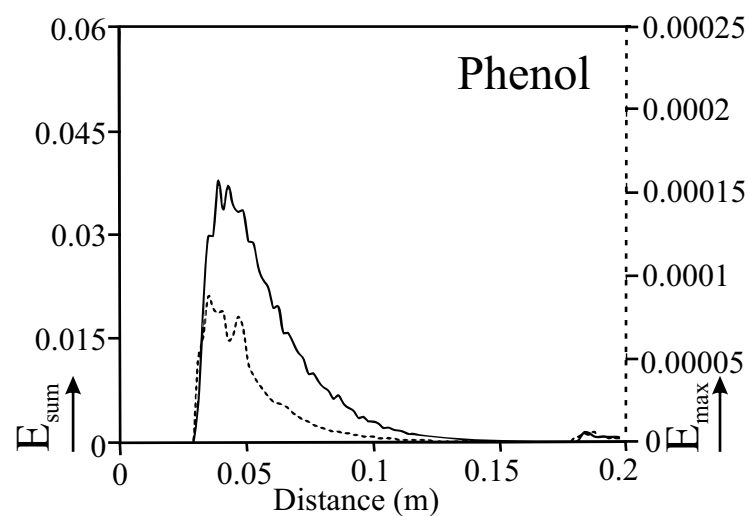
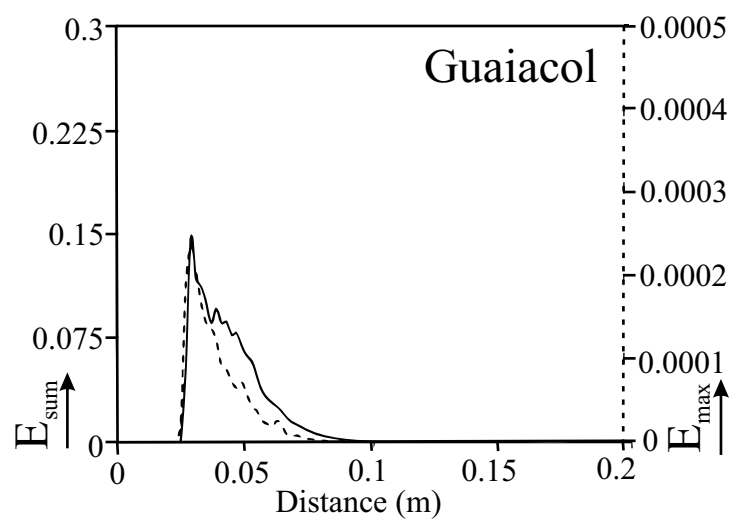
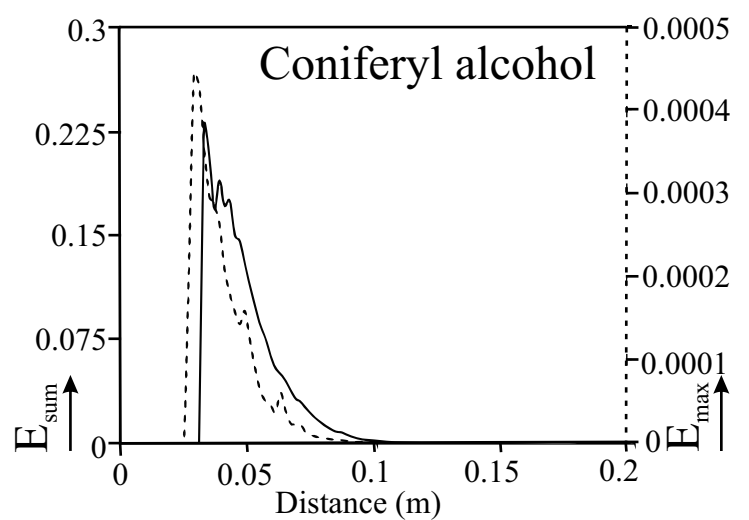
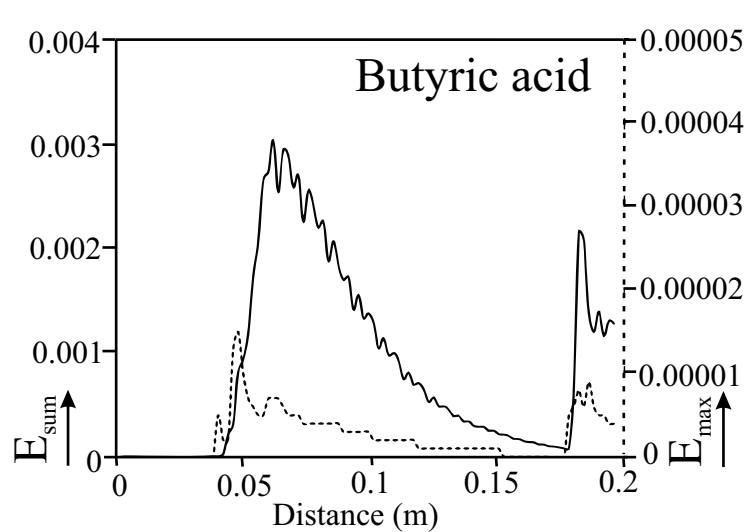
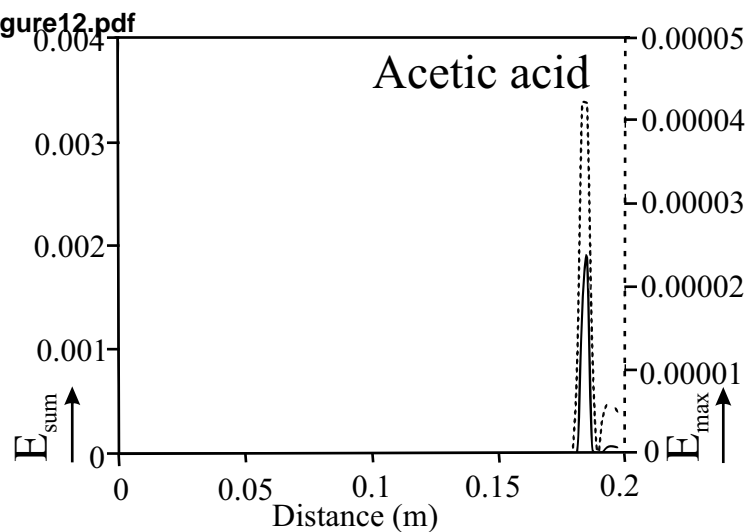








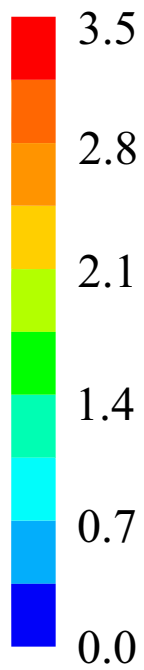




— E_{sum}
Total enthalpy of condensation
in the segment (J/s)

- - - E_{max}
Maximum value : enthalpy of
condensation in the segment (J/s)

figure13.pdf



Velocity
magnitude (m/s)

