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1 **Fast and tiny : A model for the flame propagation of nanopowders**

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7 **Abstract**

8 To avoid the influence of external parameters, such as the vessel volume or the initial turbulence, the
9 explosion severity should be determined from intrinsic properties of the fuel-air mixture. Therefore,
10 the flame propagation of gaseous mixtures is often studied in order to estimate their laminar burning
11 velocity, which is both independent of external factors and a useful input for CFD simulation.
12 Experimentally, this parameter is difficult to evaluate when it comes to dust explosion, due to the
13 inherent turbulence during the dispersion of the cloud. However, the low inertia of nanoparticles
14 allows performing tests at very low turbulence without sedimentation. Knowledge on flame
15 propagation concerning nanoparticles may then be modelled and, under certain conditions,
16 extrapolated to microparticles, for which an experimental measurement is a delicate task. This work
17 focuses on a nanocellulose with primary fiber dimensions of 3 nm width and 70 nm length. A one-
18 dimensional model was developed to estimate the flame velocity of a nanocellulose explosion, based
19 on an existing model already validated for hybrid mixtures of gas and carbonaceous nanopowders
20 similar to soot. Assuming the fast devolatilization of organic nanopowders, the chemical reactions
21 considered are limited to the combustion of the pyrolysis gases. The finite volume method was used
22 to solve the mass and energy balances equations and mass reactions rates constituting the numerical
23 system. Finally, the radiative heat transfer was also considered, highlighting the influence of the total
24 surface area of the particles on the thermal radiation. Flame velocities of nanocellulose from 17.5 to
25 20.8 cm/s were obtained numerically depending on the radiative heat transfer, which proves a good
26

27 agreement with the values around 21 cm/s measured experimentally by flame visualization and allows
28 the validation of the model for nanoparticles.

29 Keywords: *dust explosion, flame propagation, nanoparticles, modeling*

30

31 **1. Introduction**

32 Safety barriers, such as explosion venting or suppression systems (Fauske and Clouthier, 2015) need
33 to be designed by considering the experimental characteristics of the dust explosibility. These
34 characteristics are routinely determined in a 20 L sphere (Zalosh, 2019) according to well established
35 standards like EN 14034-1 (2004) and EN 14034-2 (2006). This approach only holds by assuming
36 that dust explosibility can be represented by the maximum explosion overpressure value P_{\max} and the
37 K_{st} index, deduced from the maximum rate of pressure rise dP/dt_{\max} . However, it needs to be further
38 questioned as the measurement of dust explosion severity is actually influenced by several parameters
39 such as the initial turbulence (Amyotte et al., 1988; Zhen and Leuckel, 1997), the ignition energy
40 (Zhen and Leuckel, 1997), the moisture content of the powder (Traoré et al., 2009) and the type of
41 dispersion nozzle (Dahoe et al., 2001; Murillo et al., 2018; Yao et al., 2020). Beyond these main
42 influential factors, the validity of the so-called ‘cubic law’ (Dahoe et al., 2001) commonly used to
43 extrapolate results obtained in a confined volume to another volume (Eckhoff, 2003) is also
44 questioned.

45 Standard conditions were initially defined to evaluate the explosion severity of microparticles, but
46 when it comes to nanoparticles, potential discrepancies can arise. Indeed, their small size induces a
47 high specific area and new properties, which can lead to modifications in the combustion kinetics
48 (Bouillard et al., 2010; Dufaud et al., 2011) along with extremely high ignition sensitivity, especially
49 for metallic nanopowders that can spontaneously ignite when exposed to air (Boilard et al., 2013;
50 Krietsch et al., 2015). An evaluation of the adequacy of the current standards for the assessment of
51 the explosion severity of nanoparticles is then necessary (Santandrea et al., 2019b).

52 To overcome the identified limitations, direct investigation of the flame propagation could be useful
53 so as to provide fundamental inputs in advanced simulations (CFD or phenomenological approach).
54 An essential parameter is then the laminar burning velocity, which is an intrinsic property of the fuel-
55 air mixture (Belerrajoul, 2019; Dahoe et al., 2002) that can be used in such simulations to evaluate
56 the consequences of an explosion scenario in specific conditions (Skjold, 2003). The existence of a
57 laminar burning velocity of dusts is difficult to define due to the inherent turbulence related to the
58 dispersion of the powder but such an approach was already proposed 30 years ago by Bradley and
59 Lee (1984), though it proved itself challenging when it comes to dusts. Nevertheless, the low inertia
60 and sedimentation rate of nanoparticles enable to investigate flame propagation in very low turbulent
61 conditions (Santandrea et al., 2020).

62 In this paper, a one-dimensional model initially conceived and validated for hybrid mixtures of gas
63 and combustible dust (Torrado et al., 2018) has been modified and adapted to predict the laminar
64 flame velocity of nanocellulose. Results of simulations are then compared to the experimental values
65 measured on nanocellulose using a flame propagation tube and a vented explosion sphere (Santandrea
66 et al., 2020). The consistency of a correlation established by Silvestrini et al. (2008) to predict laminar
67 flame velocity of micropowders based on the knowledge of their explosion severity was also analyzed
68 for nanocellulose.

69

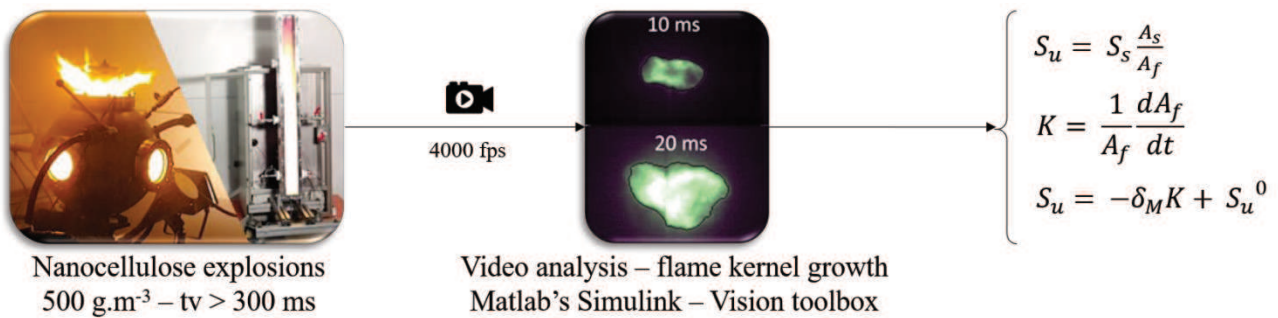
70 **2. Material and experimental method**

71 *2.1 Flame propagation observation*

72 Nanocellulose powder, or more precisely a cellulose nanocrystals powder NCC (CelluForce), is
73 composed of primary fibers, whose dimensions are 70 nm length and 3 nm width. The flame
74 propagation of nanocellulose was studied at low turbulence by Santandrea et al. (2020) in a flame
75 propagation tube (Cuervo et al., 2017) and in a vented visualization 20 L sphere, as summarized in
76 Figure 1. Due to a difficult visualization of the flame kernel at high concentration, a concentration of
77 500 g/m³ was chosen, as it is higher than the minimum explosible concentration, i.e. 125 g/m³, to

88 ensure an ignition at low ignition energy. It is greater than 225 g/m^3 , the theoretical stoichiometric
 89 concentration, and rather close to the experimental optimal concentration, i.e. 750 g/m^3 . The particle
 90 size distribution of nanocellulose dispersed in both setups was determined in situ using a laser
 91 diffraction sensor (Helos - Sympatec). It appears that the mean surface diameter in the 20L sphere
 92 reaches $10 \text{ }\mu\text{m}$, 60 ms after the beginning of the dispersion. However, by applying lower dispersion
 93 stresses, e.g. by sedimentation, agglomerates ranging from a few micrometers up to $60 \text{ }\mu\text{m}$ are formed
 94 in the powder. This does not exclude the presence of nanoparticles (from 100 nm to 300 nm) in the
 95 dust cloud as demonstrated by using a Fast Mobility Particle Sizer (FMPS) and a Scanning Mobility
 96 (Santandrea et al., 2020).

87



89 *Figure 1. Simplified scheme of the experimental determination of the laminar burning velocity by*
 90 *flame visualization used by Santandrea et al. (2020)*

91

92 Explosion were recorded using a high-speed video camera, and the flame kernel growth was analysed
 93 in terms of flame front position and surface area using a model developed by Cuervo (2015) in
 94 Matlab's Simulink. The equations initially established for gases were then applied to the obtained
 95 values, assuming that the devolatilization of organic nanopowders is fast and that, under certain
 96 concentration and turbulence conditions, the reaction is then limited by the combustion of the
 97 pyrolysis gases (Cuervo, 2015; Di Benedetto and Russo, 2007; Dufaud et al., 2012a). This regime
 98 corresponds to small particles for which both heating and pyrolysis steps occur very fast with regard
 99 to the gas combustion; i.e. for low pyrolysis time over gas combustion time ratio (Di Benedetto et al.,

100 2010). Thus, the burning velocity was calculated using the spatial velocity S_u , the estimated cross-
 101 section A_s and the flame surface A_f according to Andrews and Bradley (1972), along with the flame
 102 stretching factor K , called Karlovitz factor (Karlovitz et al., 1951). Those parameters were then
 103 combined to apply a linear relation linking the burning velocity and the Karlovitz factor K to the
 104 laminar burning velocity S_u^0 and the Markstein length δ_M , which is a parameter characterizing the
 105 stability of the flame (Clavin, 1985; Markstein, 1964).

106

107 2.2 Pressure-time evolution interpretation

108 In order to take advantage of the standard explosion tests realized in the 20L sphere, some authors
 109 such as Silvestrini et al. (2008) developed a correlation between the laminar burning velocity and the
 110 parameters P_{max} and K_{St} . Explosions tests were conducted on nanocellulose in the standard 20 L
 111 sphere according to international standards (EN 14034-1, 2004; EN 14034-2, 2006), but using
 112 chemical igniters of 100 J to avoid an overdriving phenomenon, knowing the minimum ignition
 113 energy of the dried nanocellulose is 5 mJ (Santandrea et al., 2019b). Since the values of laminar
 114 burning velocity obtained by flame propagation observation are available only at 500 g/m³, only the
 115 results obtained for this concentration are discussed in this work. Nevertheless, tests were performed
 116 over a wide range of dust concentration (up to 1250 g/m³), and the influence of the dust concentration
 117 on the laminar burning velocity is discussed by Santandrea et al. (2020). The laminar burning velocity
 118 S_u^0 of starch was then calculated from the knowledge of the explosion overpressure P_m and rate of
 119 pressure rise $(dP/dt)_m$, using the correlation established by Silvestrini et al. (2008):

$$120 \quad S_u^0 = 0.11 \frac{\left(\frac{dP}{dt}\right)_m V^{1/3}}{P_m \left(\frac{P_m}{P_0}\right)^{0.14} \left(\frac{P_m}{P_0}\right)^{\frac{1}{\gamma}}} \quad (1)$$

121 where V is the vessel volume, P_0 the atmospheric pressure and γ the ratio of specific heats. This
 122 correlation is based on several assumptions, e.g. the flame expansion is spherical, the turbulent length
 123 scales are disregarded and the burnt gases remain trapped behind the expanding flame front
 124 (Silvestrini et al., 2008).

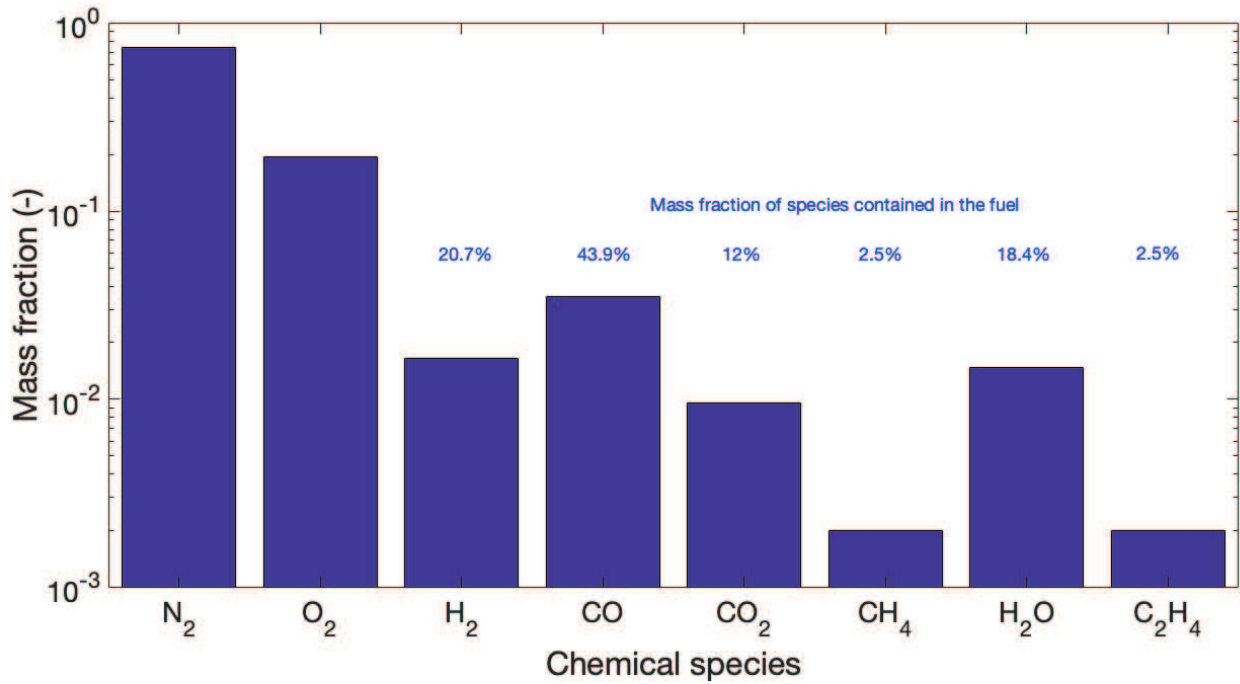
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126 **3. One-dimensional modelling of flame propagation**

127 Complementary to experiments relying on the flame visualization and the pressure-time evolution,
128 the laminar flame velocity was approached using a one-dimension flame propagation model
129 developed by Torrado et al. (2018) and initially designed to describe gas and hybrid mixtures
130 explosions). The model was then adapted to nanocellulose using the same hypothesis than for flame
131 visualization experiments, i.e. considering a fast devolatilization of the dust and a flame propagation
132 kinetically limited by the combustion of the pyrolysis gases. A similar assumption was previously
133 made by several authors, considering that a dust explosion is controlled by homogeneous combustion
134 for diameters lower than ‘a critical value’ (Eckhoff, 2003; Russo and Di Benedetto, 2013). For
135 instance, based on tests carried out on 110 μm particles, Cashdollar et al. (1989) stated that the
136 explosion of carbonaceous dusts was mainly driven by the gas phase combustion of the volatiles. Di
137 Benedetto and Russo (2007) used this assumption to validate their dust explosion model on 20 μm
138 microcrystalline cellulose. However, it should be clearly stated that this is a strong assumption when
139 dealing with organic microparticles as both the pyrolysis reaction and heat transfer can also control
140 the combustion kinetics. By calculating the values of Biot and Py numbers for cellulose particles, Py
141 being defined as the ratio between the pyrolysis time over the characteristic time for heat transfer
142 (Piskorz et al, 1986), it appears that the heat transfer may control the explosion for particles with
143 diameters greater than 200 μm . For smaller dusts, pyrolysis is certainly the rate-limiting step down
144 to a few micrometers, this ‘critical limit’ being hard to define as it depends on particles properties.
145 While keeping in mind these limitations and preliminary precautions, the assumption of a fast
146 devolatilization of organic nanopowders was made during the model development.

147 Moreover, since cellulose and starch are both polymers formed of glucose chains, both compounds
148 are assumed to produce the same pyrolysis gases when tested in the same conditions. It is another
149 strong assumption as the physical properties of the powders (particle size distribution, porosity,
150 shape...) play a significant role in their chemical reactivity.

151 Pyrolysis experiments were conducted on wheat starch ($d_{50} = 22 \mu\text{m}$) in a Godbert-Greenwald oven
152 modified according to Dufaud et al. (2012b) to collect the post-pyrolysis gases. Oven temperature
153 was modified over a range of 973 to 1173 K. It should be noted that the minimum ignition temperature
154 (MIT) of cellulose powder is approximately 773K, value depending on the particle size distribution.
155 Due to short residence times, and considering the particle external and internal heat transfers,
156 temperatures lower than 973K will lead to low pyrolysis conversion and to gas amount too low to be
157 analysed correctly. The maximum oven temperature is 1223K and a temperature too different from
158 the MIT would not make it possible to obtain gases representative of those generated during the first
159 phases of ignition (in the preheating zone before their combustion). Therefore, the gas composition
160 obtained for a concentration of approximately 500 g/m^3 and a temperature of 973 K was then used as
161 the initial composition of the fuel (Figure 2) in the model for a numerical determination of the laminar
162 flame velocity of nanocellulose. The mass fraction of the species generated by the pyrolysis of the
163 powder are also given above the bars in Figure 2. It should be emphasized that a different temperature
164 would lead to a different gas composition, especially considering that the carbon dioxide content
165 would decrease and hydrogen and methane concentrations would increase as the temperature rises.
166 Furthermore, such flash-pyrolysis experiments were performed for lean fuel mixtures and the gas
167 composition shown in Figure 2 would not be suitable for fuel rich mixtures (above 750 g/m^3 as stated
168 in 2.1). As a consequence, if the model developed in this work can be very useful to give a first
169 estimate a laminar burning velocity, it would be mistaken to believe that a single composition of
170 pyrolysis gases would be representative of what occurs at every point of the dust cloud and at every
171 moment during a dust explosion.



172

173 *Figure 2. Initial composition of the nanocellulose pyrolysis gases/air mixture considered in the*
 174 *flame propagation model for 500 g/m³ of nanocellulose*

175

176 The simulation domain is constituted of a tube with a numerical length of 5 cm involving two parallels
 177 walls divided into three distinct zones: preheat, reaction and post-flame, knowing that the flame
 178 propagates from the post-flame zone to the preheat zone. Mass, species and energy balances, notably
 179 based on the properties of the considered chemical species, were then expressed in the simulation
 180 domain. Since the main chemical species constituting the pyrolysis gases of nanocellulose are the
 181 same than the species initially considered in the model for the flame propagation of a methane/air
 182 flame (Torrado et al., 2018), similar reaction mechanisms were used. However, since the pyrolysis
 183 step mainly produced carbon monoxide, a reversible oxidation reaction of this gas to produce carbon
 184 dioxide was added (Table 1, reactions 7 and -7). Reactions involving radicals H, OH and O were also
 185 considered to improve the prediction of the flame temperature (Frassoldati et al., 2009). The mass
 186 reaction rate of a component depends on its molecular weight and the rate of the reactions in which
 187 it is involved. The rate of each reaction r_j was then expressed as:

188
$$r_j = k \prod \varphi^{n_i} \quad (2)$$

189 where φ is the mole concentration and n_i the reaction order of the component i in the reaction j . The
 190 reaction constant is assumed to follow an Arrhenius law and is defined as:

$$191 \quad k_j = A_i T^\beta \exp\left(\frac{-E_i}{RT}\right) \quad (3)$$

192 where A_i is the pre-exponential factor, E_i is the activation energy and β , a temperature exponent.

193

194 *Table 1: Reaction mechanisms considered for the combustion of the pyrolysis gases*

195 *(Units in cal, mol, m, s)*

#	Reaction	A_i	β	E_i	Reaction order	Reference
1	$\text{CH}_4 + 0.5\text{O}_2 \rightarrow \text{CO} + 2\text{H}_2$	2.45×10^9	0	3×10^4	$[\text{CH}_4]^{0.5} [\text{O}_2]^{1.25}$	(Jones and Lindstedt, 1988)
2	$\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2$	3×10^5	0	3×10^4	$[\text{CH}_4] [\text{H}_2\text{O}]$	(Jones and Lindstedt, 1988)
3	$\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$	2.75×10^6	0	2×10^4	$[\text{CO}] [\text{H}_2\text{O}]$	(Jones and Lindstedt, 1988)
-3	$\text{CO}_2 + \text{H}_2 \rightarrow \text{CO} + \text{H}_2\text{O}$	9×10^7	0	2.8×10^4	$[\text{CO}_2] [\text{H}_2]$	(Torrado et al., 2018)
4	$\text{H}_2 + 0.5\text{O}_2 \rightarrow \text{H}_2\text{O}$	3.85×10^{13}	-1	4×10^4	$[\text{H}_2]^{0.25} [\text{O}_2]^{1.50}$	(Jones and Lindstedt, 1988)
-4	$\text{H}_2\text{O} \rightarrow \text{H}_2 + 0.5\text{O}_2$	9.27×10^{18}	0.88	9.8×10^4	$[\text{H}_2\text{O}] [\text{H}_2]^{-0.75} [\text{O}_2]$	(Andersen et al., 2009)
5	$\text{O}_2 \rightarrow 2\text{O}\cdot$	1.5×10^9	0	1.13×10^5	$[\text{O}_2]$	(Frassoldati et al., 2009)
6	$\text{H}_2\text{O} \rightarrow \text{H}\cdot + \text{OH}\cdot$	2.3×10^{22}	-3	1.2×10^5	$[\text{H}_2\text{O}]$	(Frassoldati et al., 2009)
7	$\text{CO} + 0.5\text{O}_2 \rightarrow \text{CO}_2$	1.26×10^4	0	10×10^3	$[\text{CO}] [\text{O}_2]^{0.25} [\text{H}_2\text{O}]^{0.5}$	(Andersen et al., 2009)

-7	$\text{CO}_2 \rightarrow \text{CO} + 0.5\text{O}_2$	1.95×10^{12}	-0.97	78.4×10^3	$[\text{CO}_2] [\text{H}_2\text{O}]^{0.5}$ $[\text{O}_2]^{-0.25}$	(Andersen et al., 2009)
----	-----------------------------------------------------	-----------------------	-------	--------------------	----------------------------------------------------------------------	----------------------------

196

197 The calculation of the flame velocity then relies on the numerical integration of the differential
198 equations of mass, species and energy:

199
$$\frac{\partial \rho}{\partial t} + \text{div}(\rho u) = 0 \quad (4)$$

200
$$\frac{\partial}{\partial t}(\rho y_i) + \text{div}(\rho y_i u) + \text{div}(j_i) = \dot{\omega}_i \quad (5)$$

201 where j_i , ρ , y_i and u are respectively the mass diffusion flux, the mixture density, the mass fraction
202 of the i^{th} component, and the velocity. The reaction rate of the component i , depends on the molecular
203 weight W_i and on the stoichiometric coefficient of the component i in the reaction j :

204
$$\dot{\omega}_i = W_i \sum_{j=1}^{N_r} v_{i,j} r_j \quad (6)$$

205 The energy balance is developed as follows, assuming a constant pressure and negligible viscous
206 forces (Torrado et al., 2018):

207
$$\sum_{i=1}^N C_{p,i} [\rho y_i \partial_t(T) + \rho y_i u \text{div}(T) + j_i \text{div}(T)] = -\dot{\omega}_i [\sum_{i=1}^{N_{\text{species}}} [h_{f,i}^0 + C_{p,i} T]] +$$

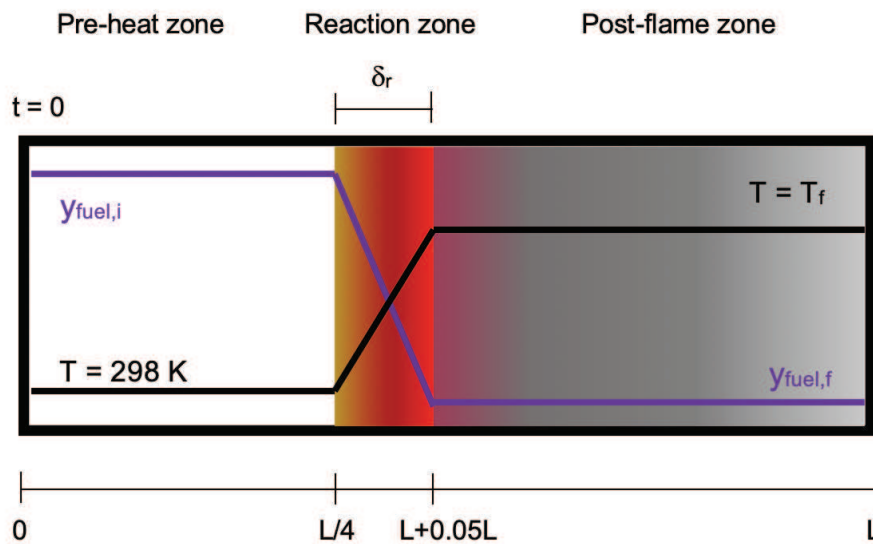
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$$\text{div}(\lambda \nabla T) + Q_{\text{rad}} \quad (7)$$

209 The radiation term Q_{rad} will be developed in section 4.2.

210 The space derivatives were discretized using the finite volume method with 160 control volumes to
211 obtain a system of ordinary differential equations, which was solved using the integration functions
212 ODE (ordinary differential equations) in Matlab. A mesh independence study was carry out by
213 beginning with 40 control volumes and increasing progressively the mesh resolution by 1.2. The
214 expression of the mass and species balance, the mass diffusion fluxes and the energy balance, along
215 with the numerical resolution, are properly described by Torrado et al. (2018).

216 The resolution of the ordinary differential equations requires an initial value of the temperature and
217 mass fractions of all the considered species in every numerical domain. The composition in the
218 preheat zone, which represents 25% of the numerical domain, is defined by the mass fractions of the
219 considered mixture in laboratory conditions. As a first approximation, the mass fractions and

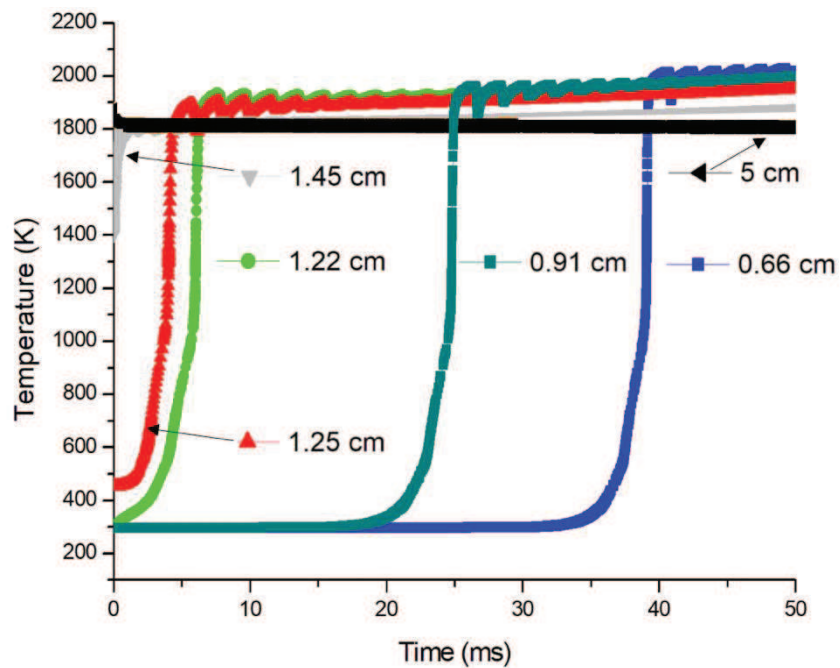
220 temperature are assumed to evolve linearly in the reaction zone, implying those values are known if
 221 the initial and final conditions are fixed. To estimate the conditions in the post-flame zone (70% of
 222 the considered distance), the adiabatic temperature and mass fraction of the burnt gases for a steady
 223 flame were calculated using PREMIX program (Kee et al., 1993). This approach, represented in
 224 Figure 3, was used to reduce the calculation time and to improve the convergence of the program, by
 225 initializing all the conditions close to a stable solution. Since this numerical model also aims at
 226 considering the radiative heat transfer induced by the presence of nanoparticles in the mixture, which
 227 is not the case of the PREMIX program, this latter was not considered as a suitable method to
 228 determine the laminar burning velocity of nanopowders.



229
 230 *Figure 3. Schema of the initial conditions of the temperature and fuel fraction in each zone of the*
 231 *flame*

232
 233 The system previously defined was then analyzed considering a distance L of 5 cm and an integration
 234 time of 50 ms. In order to show an example of raw results obtained by a simulation, the evolution of
 235 the temperature with time is presented in Figure 4 for an initial fuel concentration of 500 g/m^3 . The
 236 different positions indicated in Figure 4 correspond to various zones: i) the temperature evolution in
 237 the preheat zone is represented by the curves at 0.66 and 0.91 cm, ii) the flame boundaries are located
 238 at 1.22 cm and 1.45 cm (including 1.25 cm), and iii) the time-evolution of the temperature in the post-

239 flame zone is given at 5 cm. It appears that, in the post-flame zone (5 cm), the temperature is constant
240 with time, since the reaction already occurred. Then, a fast increase of the temperature after a few
241 milliseconds is visible in the reaction zone, especially at 1.22 and 1.25 cm. The thermal wave
242 progressively shifts toward the preheat zone with time, describing the propagation of the flame.
243 The 1D model was previously validated on methane/air mixtures (Torrado et al., 2018) as it gives a
244 laminar burning velocity of 34 cm/s, which is close to the experimental values of methane/air burning
245 velocity from 34 to 38 cm/s (Dirrenberger et al., 2011; Proust, 2006). Moreover, the model shows a
246 good agreement with commercial software (PREMIX program) to estimate the mass fraction of burnt
247 gases and the final temperature; for instance, the flame temperature of a stoichiometric CH₄/air
248 mixture obtained by the 1D model is 2271 K compared to a theoretical adiabatic temperature of 2236
249 K.



250
251 *Figure 4. Evolution of the temperature with time for different control volumes when modelling the*
252 *flame propagation of nanocellulose (quiescent conditions, dust concentration : 500 g/m³)*

253

254 4. Results and discussion

255 4.1 Combustion of the pyrolysis gases

256 The position of the flame front, assimilated to the position of the highest temperature, was recorded
257 for each integration time and is presented in Figure 4. It should be reminded that the reaction zone
258 was initially located between 1.25 cm and 1.5 cm. However, before 1 ms, a very fast displacement of
259 the flame is observed, preventing a clear determination of the flame front position between 1.4 and
260 1.5 cm. Nevertheless, a linear evolution of the flame position with time can be observed from 1 ms
261 to 50 ms. A laminar flame velocity of 17.5 cm/s, represented by the slope of the linear regression,
262 was then obtained for the combustion of nanocellulose. It should be stressed that this value should be
263 viewed with caution as the pyrolysis step has been considered as very fast with regard to the
264 combustion of the pyrolysis gases, which is a strong assumption only validated from very small
265 particles.

266 The flame velocity calculated using the flame propagation model was then compared to the values
267 experimentally obtained by Santandrea et al. (2020) (Table 2). The value determined numerically
268 appears to be of the same order of magnitude than the experimental ones (from 16.9 to 23.5 cm/s),
269 with a maximum difference of 22% with regard to the laminar flame velocity measured in the flame
270 propagation tube. This value is also consistent with laminar flame velocity of “wood gas” at the
271 stoichiometry mentioned in the literature by Mollenhauer and Tschöke (2010) and Przybyla et al.
272 (2008), reaching around 14 cm/s and 20 cm/s respectively. Moreover, these results are also in
273 agreement with the flame velocities of dusts, i.e. from 15 to 30 cm/s for the unstretched laminar
274 burning velocity of cornstarch (Dahoe et al., 2002) and from 15 to 55 cm/s, proposed by Sattar et al.
275 (2014) for various powders such as lycopodium, coal and walnut shells. Nevertheless, the difference
276 between experimental and numerical values can obviously come from experimental uncertainties, but
277 can also be due to the omission of both the contribution of the radiative heat transfer to the flame
278 propagation or of the pyrolysis reaction. Indeed, if the pyrolysis step can decrease the flame velocity
279 due to a kinetic limitation, the fresh or unburnt remaining particles can also improve the flame
280 propagation through a heat transfer modification in the preheat zone. Such impacts on the radiative
281 transfers and on the flame speed were notably observed in the visible spectrum when combustible or

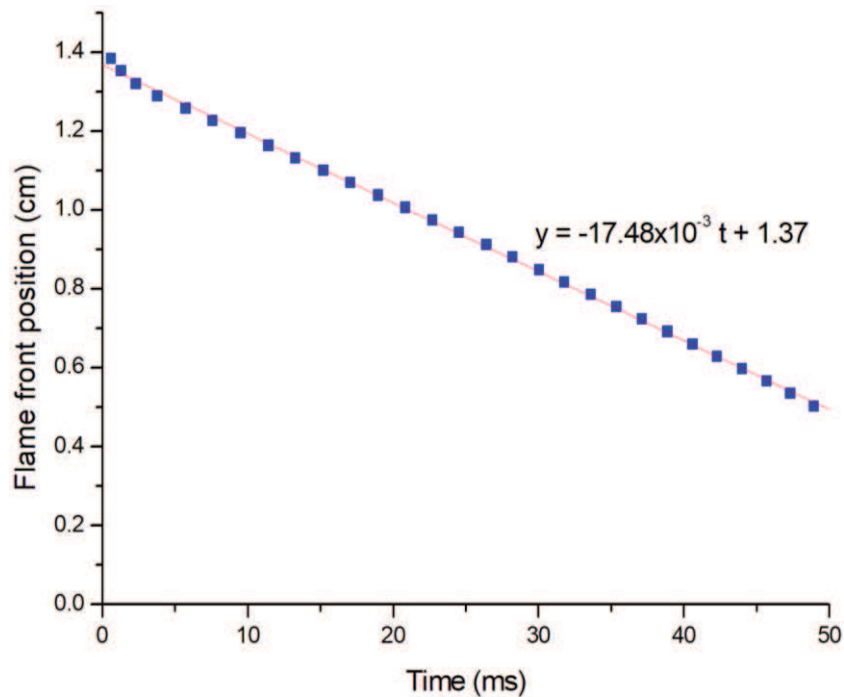
282 even inert particles (carbon black or alumina) were added to methane (Torrado et al., 2017). To
 283 numerically evaluate this influence, the contribution of the radiative heat transfer, added to the flame
 284 propagation model by Torrado et al. (2018) and based on the work of Haghiri and Bidabadi (2010),
 285 will be now considered during the combustion of the pyrolysis gases of nanocellulose.

286

287 *Table 2: Numerical and experimental values of laminar flame velocity of nanocellulose*

Determination method	Laminar flame velocity (cm/s)
Flame propagation model	17.5
Flame visualization: propagation tube	21.4 ± 1
Flame visualization: vented sphere	20.5 ± 3
20L sphere: application of Silvestrini et al. (2008) correlation	19.9 ± 3

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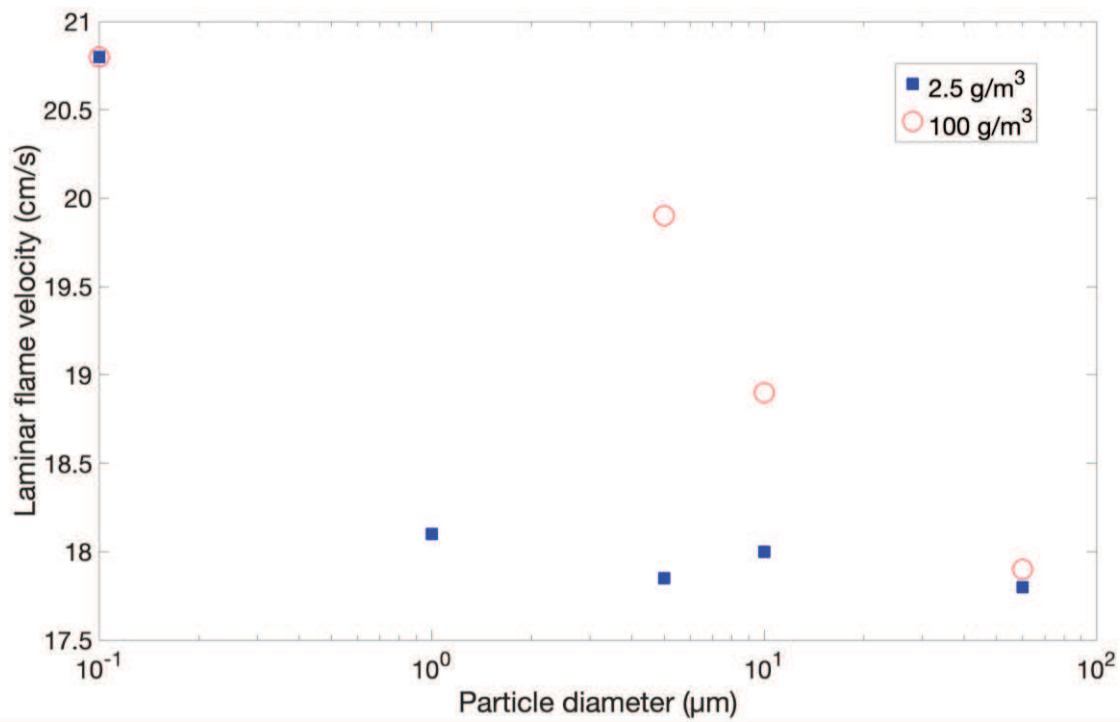
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290 *Figure 4. Evolution of the flame front position with time during the combustion of nanocellulose*
 291 *pyrolysis gases*

292

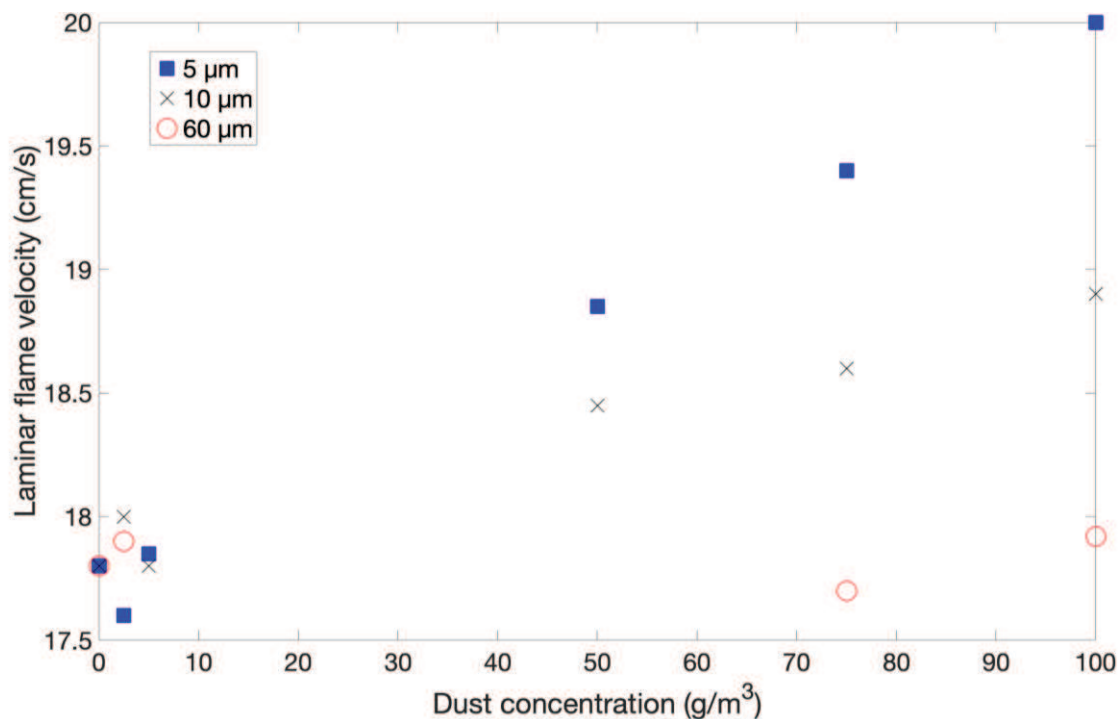
293 **4.2 Influence of the radiative heat transfer**

294 Since the pyrolysis of nanocellulose particles and the combustion of the pyrolysis gases happen
295 simultaneously, the unburnt particles can impact the flame propagation by variations of the heat
296 transfer. However, the remaining dust can hardly be quantified and characterized for each integration
297 time without taking the pyrolysis kinetics into account. Thus, several dust concentrations, assumed
298 constant with time, were tested. The dust clouds were supposed to be homogeneous over the
299 simulation domain and constituted of monodispersed spherical particles. Moreover, due to the
300 agglomeration of the nanoparticles, the particle size after dispersion must be considered (Santandrea
301 et al., 2019a). As described in section 2.1, particle size distribution measurements after dispersion of
302 nanocellulose in the 20L sphere led to a mean value of 10 μm (Santandrea et al., 2020). This value
303 was then chosen as a reference for the calculation, along with 100 nm, to represent the primary
304 particles, and 60 μm , which is the mean diameter of nanocellulose agglomerates before dispersion,
305 i.e. the agglomerates not broken by the dispersion process. In this model, Mie scattering, valid for
306 micron particles, was then used to define the radiative heat transfer. It should be noted that Rayleigh
307 scattering, encountered for particles smaller than 100 nm, do not contribute significantly to the flame
308 expansion due to the emission in every direction (Hong and Winter, 2006). Thus, decreasing the
309 particle size below this size would only decrease the radiative heat transfer contributing to the flame
310 propagation, and so the flame velocity. The concentration of dust that did not react during the
311 combustion of the 500 g/m^3 of nanocellulose was varied from 2.5 g/m^3 to 100 g/m^3 . These values
312 were chosen as orders of magnitude to represent the radiative heat transfer at the beginning and at the
313 end of the reaction. The radiative heat transfer was then added to the energy balance, and the heat
314 capacity of the dust was then taken into account during the calculation of the mean heat capacity of
315 the mixture. Due to the assumption of a fast pyrolysis, the heterogeneous reactions involving the solid
316 particles were not considered in the model. However, it should be noted that Torrado et al. (2018)
317 evidenced that the contribution of the chemical reactions of the powder is negligible with regard to
318 the contribution of the radiative heat transfer at low concentrations (2.5 g/m^3).



319

320 *Figure 6. Numerical values of the laminar flame velocity of a mixture of nanocellulose pyrolysis gases*
 321 *(500 g/m³) and inert particles considered for the radiative heat transfer: influence of the inert particle*
 322 *diameter at 2.5 and 100 g/m³.*



323

324 *Figure 7. Numerical values of the laminar flame velocity of a mixture of nanocellulose pyrolysis gases*
 325 *(500 g/m³) and inert particles considered for the radiative heat transfer: influence of the inert powder*
 326 *concentration for particle diameters of 5, 10 and 60 μm.*

327

328 Figure 6 confirms that the laminar flame velocity obviously varies as a function of the particle size
329 distribution (Ghaffari et al., 2019) and of the dust concentration. In Figure 6, it also appears that small
330 particles contribute more to the radiative heat transfer than bigger particles in the micro-range. Indeed,
331 particles of 60 μm bring similar contribution to the flame acceleration, i.e. around 0.5 cm/s, at 2.5
332 g/m^3 and 100 g/m^3 , whereas 100 g/m^3 of 5 μm particles lead to a flame velocity of 20 cm/s, i.e. 14%
333 higher than the flame velocity of the pyrolysis gases. At this point, it should be stressed that for large
334 particles, the assumption of a fast pyrolysis with regard to the combustion reaction is certainly not
335 valid and that simulations presented for powders having a mean diameter larger than 10 μm are only
336 given as an indication. It should also be underlined that the contribution of particles of 100 nm to the
337 radiative heat transfer may be overestimated since Mie scattering was considered for the calculation
338 whereas Rayleigh scattering is more representative of the heat transfer of nanoparticles. In Figure 7,
339 it appears that a dust concentration of 2.5 g/m^3 leads to a mean flame velocity of around 17.8 cm/s
340 for particles between 5 and 60 μm . Therefore, increasing the concentration also increases the
341 contribution of the radiative heat transfer to the flame propagation, reaching 20.0 cm/s when
342 considering 100 g/m^3 of particles of 5 μm .

343 Both the dust concentration and the particle size are thus of great importance when considering the
344 radiative heat transfer. As proposed by various authors (Haghiri and Bidabadi, 2010; Meinköhn et al.,
345 2007), the absorbed, emitted and scattered energy in a dust cloud can be expressed as:

346
$$\frac{dI}{dx} = K_a I + K_s I - K_a I_b - \frac{K_s}{4\pi} \int_{4\pi} I(\Omega) P d\Omega \quad (8)$$

347 where I and I_b are the thermal intensity and thermal intensity of a black body. An analytical solution
348 of equation 8 was proposed by Haghiri and Bidabadi (2010) for each zone represented in Figure 3.
349 By neglecting the multi-scattering contribution, i.e. for isotropic scattering, the integral term can be
350 removed from equation 8. The absorption coefficient K_a and the scattering coefficient K_s directly
351 depend on the dust concentration, the dust density and the particle size, as follows (Haghiri and
352 Bidabadi, 2010):

353
$$K_a = \frac{3}{2} \frac{c}{\rho_p d_p} Q_{abs} \quad (9)$$

354
$$K_s = \frac{3}{2} \frac{c}{\rho_p d_p} Q_{sca} \quad (10)$$

355 where C is the dust concentration, ρ_p the particle density, d_p the particle diameter and $Q_{abs} = \varepsilon_p$ and
356 $Q_{sca} = 1 - \varepsilon_p$ respectively the absorption and scattering efficiency, and particle emissivity ε_p . In the
357 preheat zone, only the absorption term was considered, whereas both absorption and scattering were
358 considered in the reaction and post-flame zones. The implementation of both equations (9) and (10)
359 in the 1D model was detailed by Torrado et al. (2008). The thermal properties of cellulose powders
360 were considered as inputs.

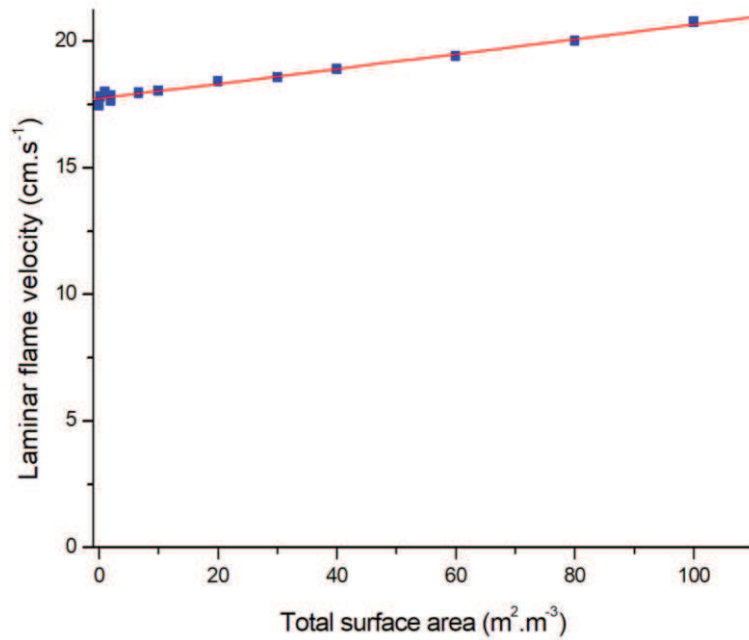
361 The absorption and scattering coefficients are then directly proportional to the total surface area
362 (TSA) developed by the particles in the cloud, which can be expressed as follows for spherical
363 particles:

364
$$TSA = \frac{6 c}{\rho_p d_p} \quad (11)$$

365 Once again, simulations were performed by considering the combustion of pyrolysis gases
366 representative of the devolatilization of nanocellulose (Figure 2) and at an initial mass concentration
367 of 500 g/m^3 . In order to consider the radiative contribution of the powder on the flame propagation,
368 particles having the same thermal properties than cellulose were numerically added to the reactive
369 system; however, potential heterogeneous reactions were not taken into account. A linear evolution
370 of the calculated flame velocity with the total surface area developed by the particles considered in
371 the radiative heat transfer appears in Figure 8. It can be observed that the radiative heat transfer
372 generated by particles developing a total surface area lower than $10 \text{ m}^2/\text{m}^3$ does not lead to a
373 significant increase of the flame velocity, with values between 17.5 and 18 cm/s. However, when
374 considering a total surface area of $100 \text{ m}^2/\text{m}^3$, a flame velocity of 20.8 cm/s is reached, thus proving
375 the importance of considering the surface area when analysing dust explosions, instead of focusing
376 only on mass concentration. It should also be noted that increasing the concentration too much would

377 lead to an important increase of absorption, which would hence limit the heat radiation in the preheat
378 zone.

379



380

381 *Figure 8. Influence of the total surface area of the particles implied in the radiative heat transfer on*
382 *the flame velocity of a mixture of nanocellulose pyrolysis gases (500 g/m^3) and inert particles*

383

384 5. Conclusions

385 The laminar burning velocity of nanocellulose has been determined using a one-dimensional flame
386 propagation model adapted from a model already validated for hybrid mixtures. The numerical
387 system, composed of mass and energy balances equations and of mass reaction rates adapted to the
388 combustion reactions, was solved by the finite volume method. Assuming that the devolatilization of
389 organic nanopowders is fast, the chemical reactions were considered limited to the combustion of the
390 pyrolysis gases. A first value of laminar flame velocity of 17.5 cm/s was obtained for 500 g/m^3 of
391 nanocellulose, which is close to values experimentally measured in a flame propagation tube or a
392 20 L sphere, around 21 cm/s , thus showing a good consistency between the numerical and
393 experimental approaches.

394 However, since in practice, the devolatilization of the particles is not instantaneous, the remaining
395 particles can contribute to the radiative heat transfer, which was added to energy balance. Due to the
396 tendency of nanoparticles to agglomerate, different particle diameters and dust concentrations were
397 tested. Thus, although the heat transfer of nanoparticles tends to be neglected due to Rayleigh
398 scattering, which does not contribute to the flame propagation, the contribution of the remaining
399 micro-agglomerates after dispersion must be considered. Indeed, the existence of a linear relation
400 between the laminar flame velocity and the total surface area developed by the particles implied in
401 the radiative heat transfer was highlighted. A flame velocity reaching 20.8 cm/s for a total surface
402 area of 100 m²/m³ considered for the radiative heat transfer was then obtained, showing a strong
403 impact of the heat radiation on the flame propagation.

404 Nevertheless, it should be kept in mind that, even if the nanocellulose pyrolysis could be considered
405 as very fast, this assumption is potentially not valid for nanopowders agglomerates and certainly not
406 for larger organic particles. In the latter case, consideration of the pyrolysis kinetics, using for instance
407 a semi-global lumped-reaction system, will be necessary. In any case, this work shows that knowledge
408 of the kinetics of pyrolysis and/or combustion allows the numerical assessment of a laminar flame
409 velocity, provided that the radiative phenomena are also taken into consideration.

410

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414

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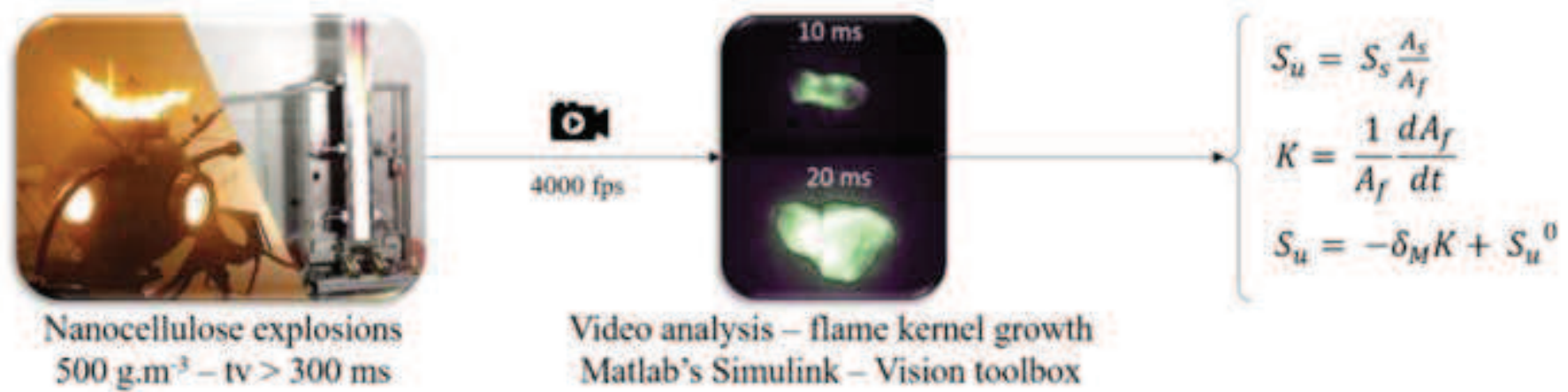


Figure2

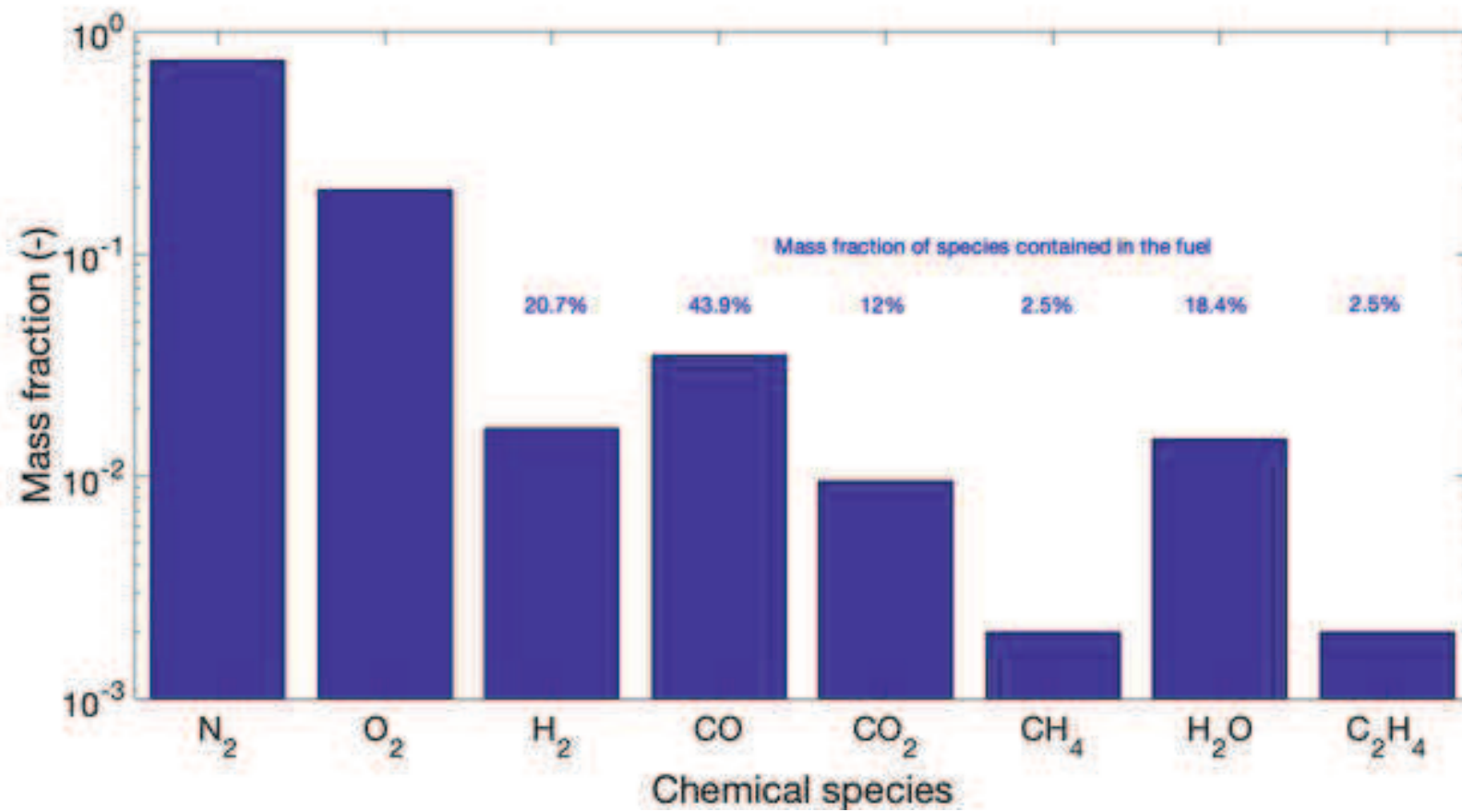


Figure3

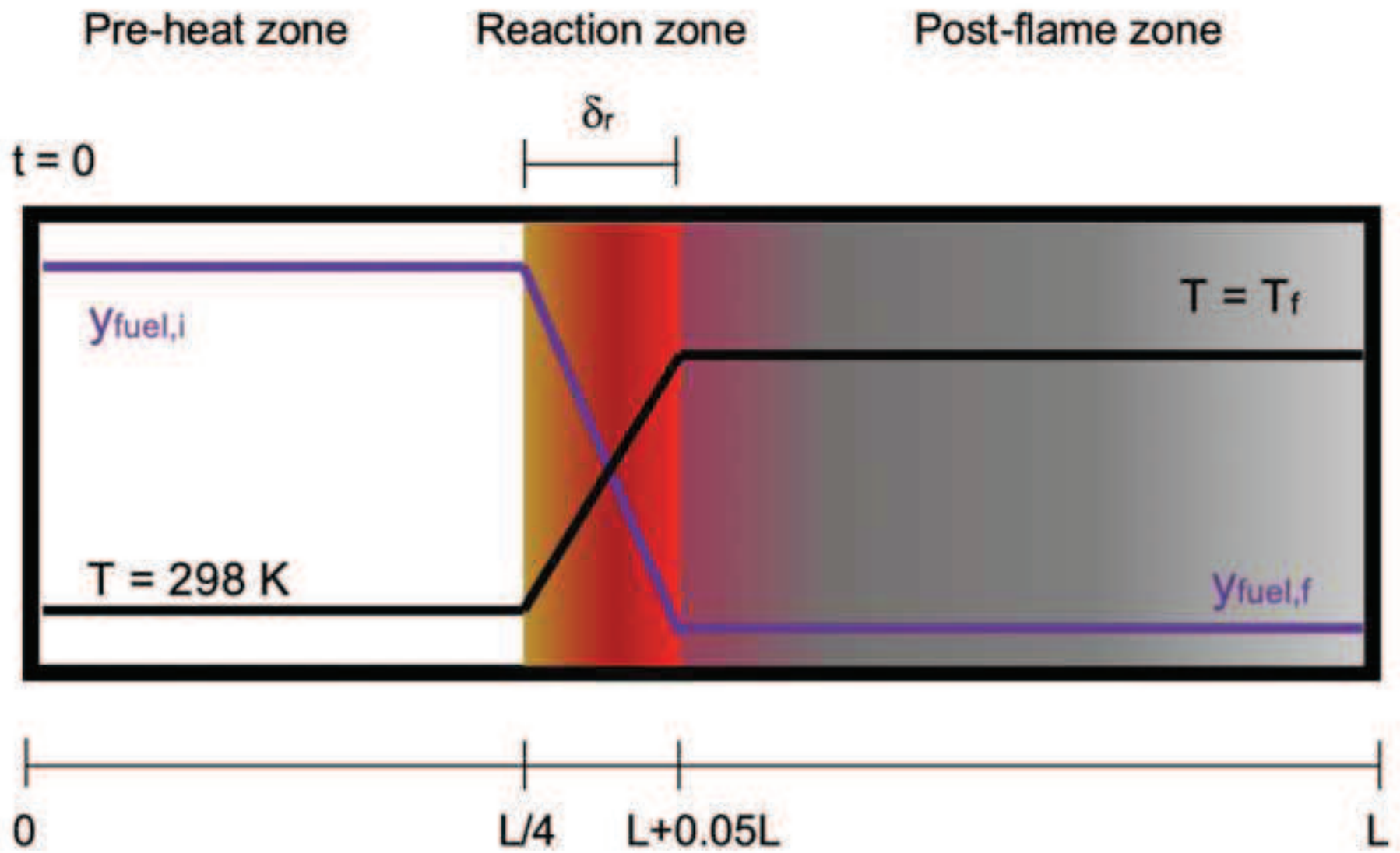


Figure4

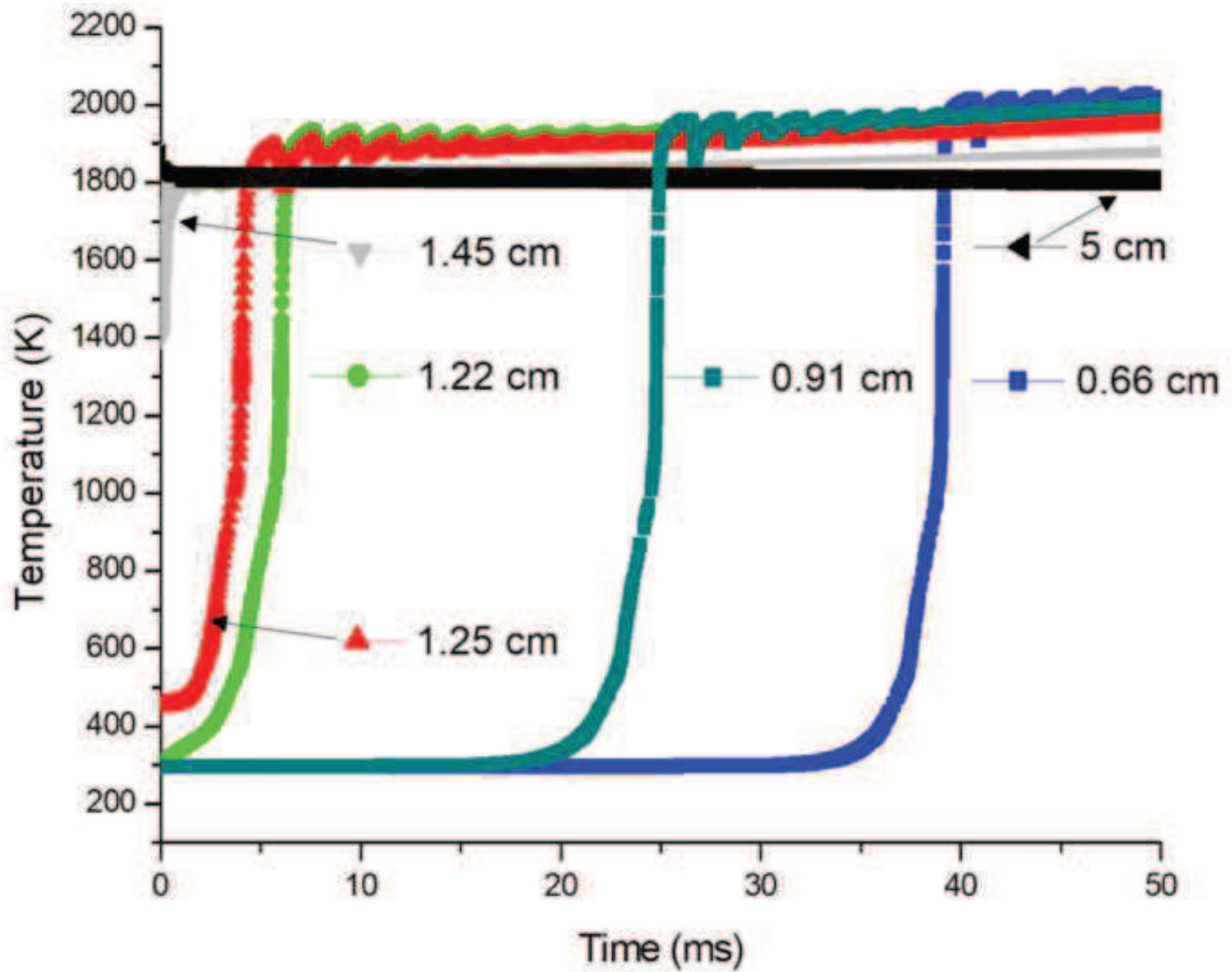


Figure5

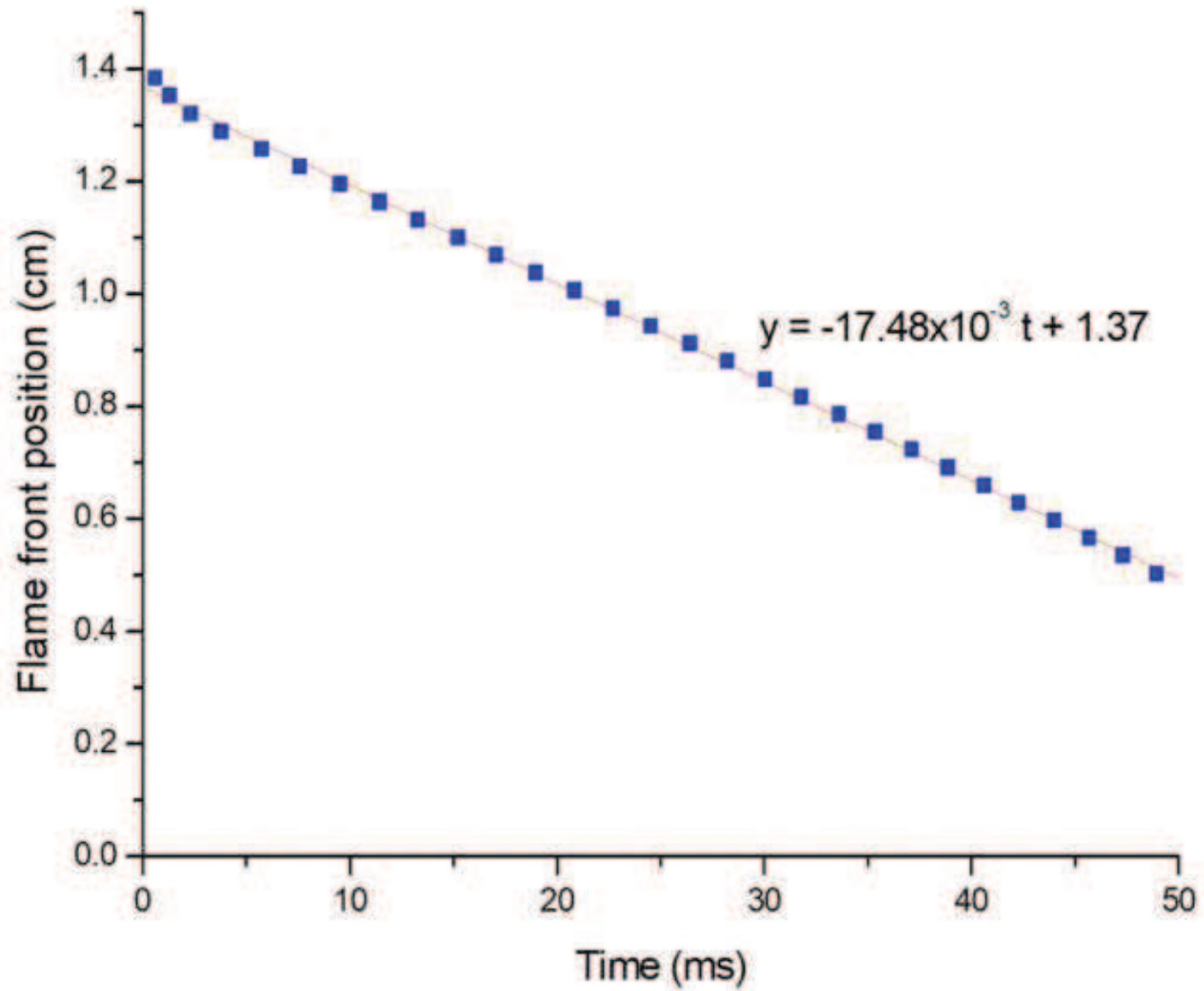


Figure6

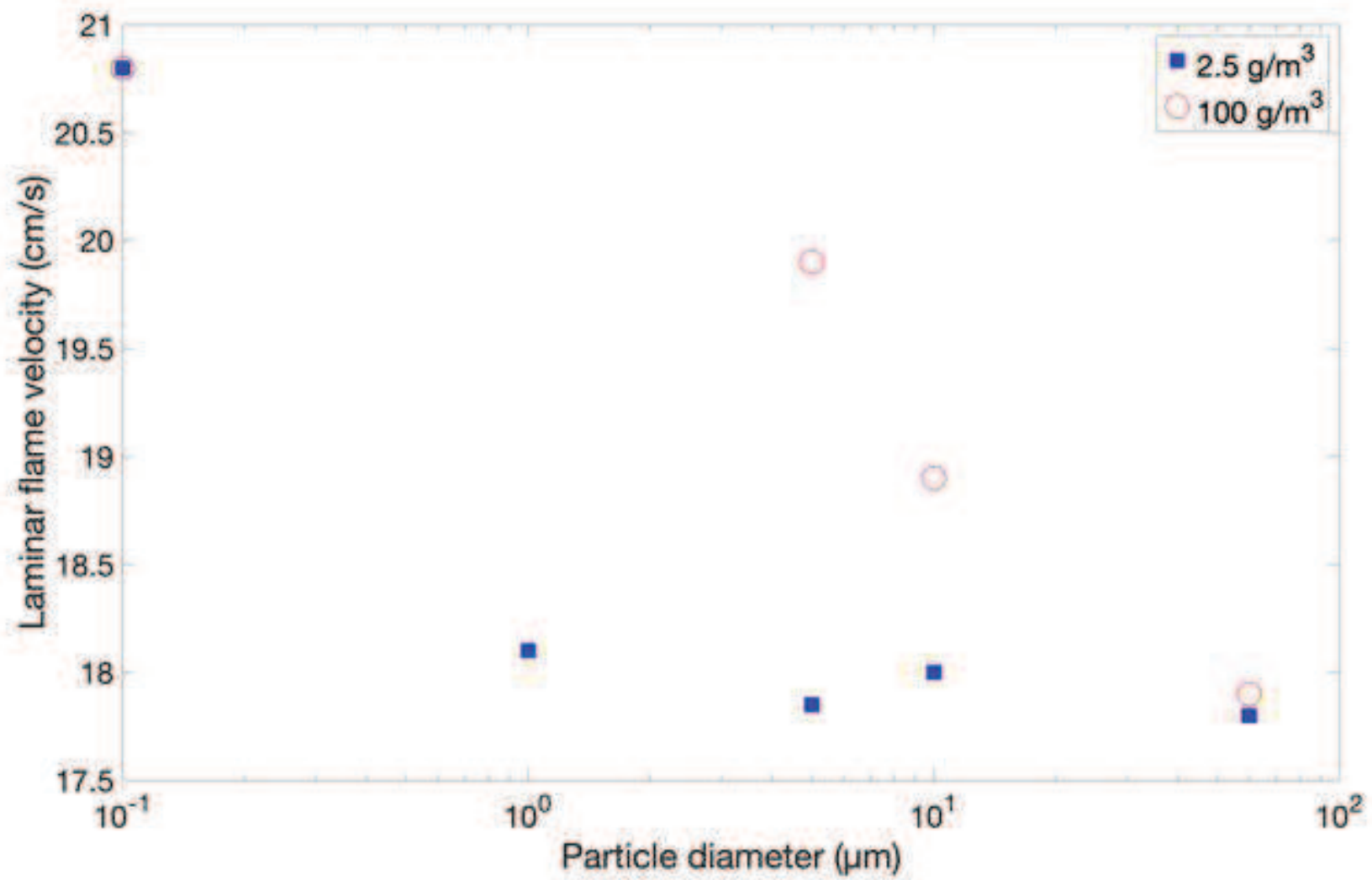


Figure7

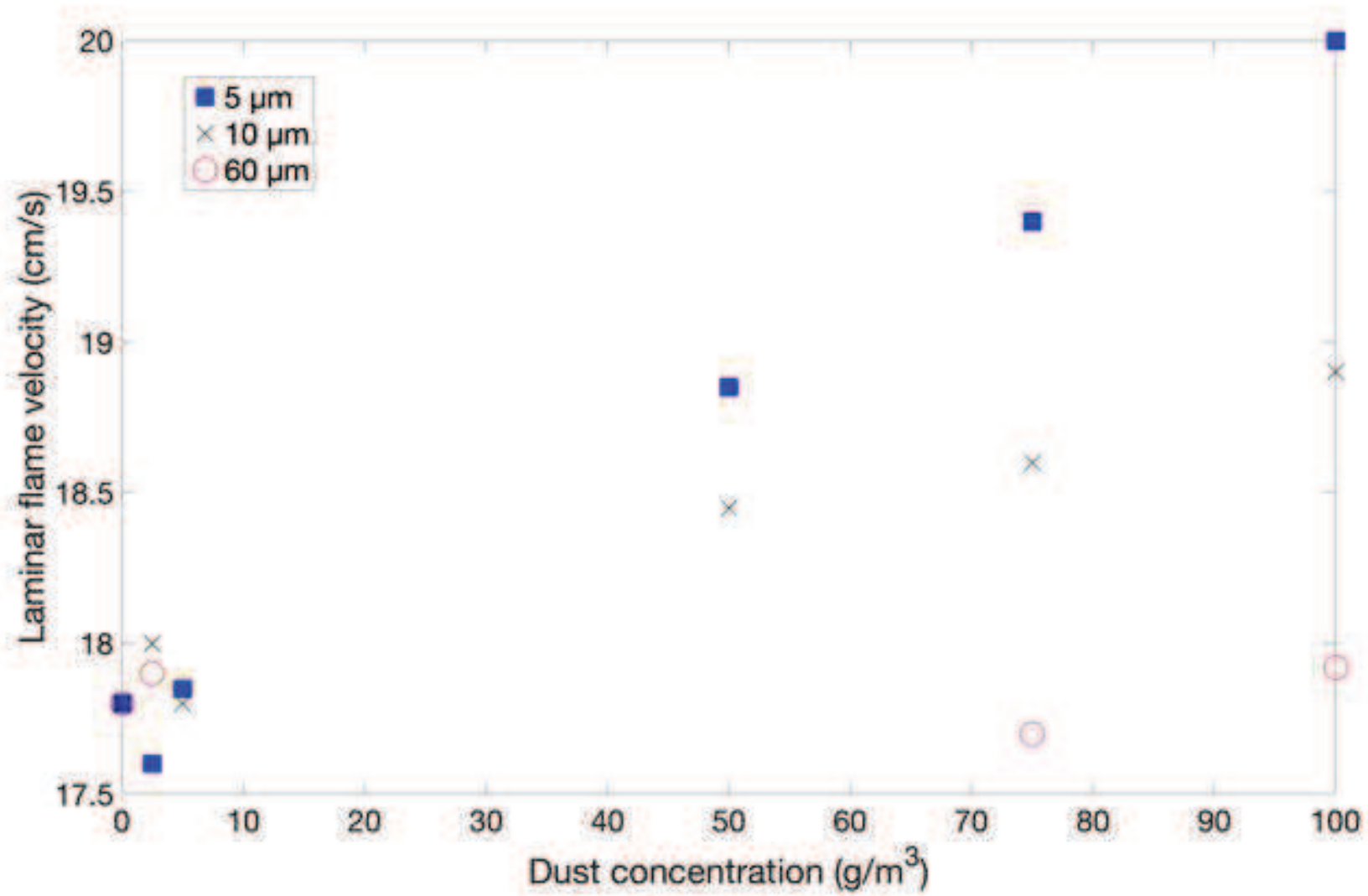


Figure8

