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‘Knock on nanocellulose’:

approaching the laminar burning velocity of powder-air flames

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Abstract:

Due to their low sedimentation rate, nano-objects offer the opportunity to study flame propagation at low turbulence. The burning velocity was then estimated by flame visualization in two apparatuses: a vertical 1 meter long tube with a square cross-section and a 20L sphere equipped with visualization windows and a vent. This work aims to study the laminar burning velocity of nanocellulose by a direct visualization of the flame propagation within these devices. A high-speed video camera was used to record the flame propagation, and an estimation of the unstretched burning velocity was obtained through linear and nonlinear relationships relating the flame stretching and the flame velocities. Although these methods were initially established for gases, the organic nature of nanocellulose implies a fast devolatilization, which makes the application of the methods possible in this work. Similar results were obtained in both apparatuses in different turbulence conditions, proving the

23 laminar burning velocity was approached. The laminar burning velocity for the nanocellulose
24 was determined to be 21 cm.s^{-1} . This value, estimated through flame propagation
25 visualization, was then compared to the value calculated by applying a semi-empiric
26 correlation to the pressure-time evolution recorded during standard explosion tests in the 20L
27 vessel.

28 **Keywords:** Dust Explosion; Nanocellulose; Flame propagation; Burning Velocity;
29 Nanopowder

30

31

32 **1. Introduction**

33 In order to assess explosion risks, the determination of key safety parameters is required
34 (Eckhoff, 2003; Jespen, 2016). Among these parameters, the maximum explosion
35 overpressure and the maximum rate of pressure rise are commonly used to characterize the
36 explosion severity. Their determination implies controlled explosion tests in a closed vessel to
37 measure the pressure evolution with time, which induces a volume-dependence of the results.
38 To be able to design protection equipment, normalized test conditions and apparatuses are
39 then needed. For instance, the EN 14034 standard is generally used, recommending tests in a
40 20L sphere or a 1 m^3 vessel under specific conditions (injection procedure, type of nozzle,
41 ignition delay time, ignition source and energy, initial temperature and pressure).

42 However, standard conditions can be different from industrial conditions (Tamanini, 1990)
43 and do not consider the specificities of each powder. For instance, the ignition energy is set at
44 10 kJ while the minimum ignition energy can be much lower, which can lead to an
45 overdriving phenomenon and a misestimation of the risk (Going et al., 2000; Zhen and

46 Leuckel, 1997). Moreover, to represent the volume-dependence of the experimental explosion
47 parameters, a ‘cube-root law’ was proposed by Bartknecht (1989) and demonstrated later by
48 Eckhoff (2003). This law is valid under several assumptions considering that the vessels have
49 similar geometries, the flame thickness is negligible with respect to the vessel radius, the
50 burning velocity is the same in both volumes and point ignition occurs at the center of the
51 vessels (Lewis and von Elbe, 1987; Skjold, 2003). Several tests have been performed to
52 compare results obtained in the 20L sphere and the 1m³ vessel, revealing significant
53 differences between the two vessels, thus questioning the validity of the cube-root law
54 (Clouthier et al., 2019; Proust et al., 2007; van der Wel et al., 1992). It has notably been
55 proven that radiation can play a significant role in dust combustion, which tends to increase
56 the flame thickness and even invalidate the cube-root law in some cases, especially for metal
57 particles (Bidabadi and Azad, 2015; Dahoe et al., 1996; Taveau et al., 2018). Discrepancies
58 have also been found in terms of turbulence, showing the ignition delay time in both vessels
59 does not allow the same initial turbulence of the dust cloud (Amyotte et al., 1988; Dahoe et
60 al., 2001; Pu et al., 1991; van der Wel, 1993).

61 If the procedures to determine explosion safety parameters are relatively well defined for
62 micro-powders, they are still subject to debate, especially for nanopowders. Several authors
63 have highlighted, that due to their small size, nanoparticles develop a high specific surface
64 area leading to specific explosion properties (Amyotte, 2014; Boilard et al., 2013; Bouillard et
65 al., 2010; Eckhoff, 2012; Mittal, 2014; Vignes et al., 2019; Wu et al., 2014). Among these
66 specificities, their high specific surface area can both lead to changes in the rate-limiting step
67 of the oxidation reaction (Bouillard et al., 2010; Vignes et al., 2019) and to the modification
68 of the dust cloud particle size distribution due to the agglomeration phenomenon (Eckhoff,
69 2012; Santandrea et al., 2019a). Moreover, other specificities may call into question the
70 relevance of standardized tests in a closed vessel. For instance, due to their increased ignition

71 sensitivity, underlined notably by Krietsch et al. (2015), Mohan et al. (2012) and Sundaram et
72 al. (2013), an overdriving or pre-ignition phenomenon is more likely to occur when testing
73 nanopowders. Furthermore, the radiative heat transfer occurring during the flame propagation
74 can be greatly affected by the agglomeration level of the dust cloud (Dufaud et al., 2011;
75 Kosinski et al., 2013; Vignes, 2008). In the case of enhancement of the radiative transfer, the
76 flame thickness would no longer be negligible with respect to the vessel radius, as assumed by
77 the cube-root law. As a consequence, a direct transposition of the safety parameters from lab-
78 scale to industrial-scale may be inaccurate.

79 To overcome the cube-root law limitations, flame propagation is often studied and used in
80 Computational Fluid Dynamics (CFD) simulations to characterize gas explosion (Di
81 Benedetto et al., 2013; Ghaffari et al., 2019; Skjold, 2003). The essential parameters are then
82 the flame thickness and, more importantly, the laminar burning velocity (Belerrajoul, 2019;
83 Dahoe et al., 2002a). This latter is a fundamental property of the fuel and only depends on the
84 fuel nature and the mixture concentration (Miao et al., 2014). It can be used in simulations to
85 evaluate the consequences of an explosion scenario with controlled conditions (Skjold, 2007;
86 Tolia and Venetsanos, 2018). Although the estimation of the laminar burning velocity of
87 gases is not trivial, the experimental determination of the laminar burning velocity of dust-air
88 mixtures is much more difficult to perform than for gases due to the inherent turbulence
89 related to the powder dispersion. Nevertheless, some experiments have been performed by
90 various authors in a micro-gravity environment during a parabolic flight or using a drop tower
91 (Goroshin et al., 2011; Lee et al., 1993; Pu et al., 1998; Tang et al., 2009). However, such
92 tests being very expensive and difficult to perform on a large scale and with a high frequency,
93 three more accessible methods for the determination of the unstretched burning velocity can
94 be found in the literature: the burner method, with various existing configurations (Dahoe et
95 al., 2002; Julien et al., 2017; Lomba et al., 2019; van der Wel, 1993), the contained explosions

96 method (Silvestrini et al., 2008; Skjold, 2003; van der Wel, 1993) and the tube method
97 (Andrews and Bradley, 1972; Di Benedetto et al., 2011; Proust, 2006). However, test results
98 obtained in turbulent conditions had to be numerically extrapolated to a zero degree of
99 turbulence (Bradley et al., 1989); as a consequence, the term ‘unstretched burning velocity’
100 will be preferred to ‘laminar flame velocity’ to qualify the parameter obtained.

101 Despite the greater accessibility of these methods, experimental issues remain and usually
102 limit the reproducibility and accuracy of the results. In addition to the stochastic nature of
103 turbulence, issues can also come from particle agglomeration, instabilities of the biphasic
104 flow, set-up fouling or flame front visualization difficulties. However, for specific reactive
105 mixtures such inconveniences may be limited: for instance, testing gas-dust mixtures with a
106 low dust concentration (called gas-driven hybrid mixtures) in a semi-open tube allowed the
107 determination of their unstretched flame velocity and highlighted the influence of
108 turbulence/combustion interactions on the flame propagation (Cuervo, 2015; Torrado et al.,
109 2017b).

110 Similarly, the low inertia of nanoparticles allows tests with a long ignition delay time.
111 Consequently, experiments can be performed at low turbulence with nanoparticles while
112 sedimentation would occur for micropowders under the same experimental conditions.
113 Decreasing the turbulence level of the initial dust cloud can help approaching the ‘laminar
114 flame velocity’ of dust-air mixtures through the assessment of an unstretched flame velocity,
115 which is then expected to be independent from turbulence. This concept will be developed
116 through the characterization of the unstretched burning velocity of a nanocellulose powder
117 using three different procedures and set-ups: a semi-opened tube, a vented vessel and the
118 classical 20L explosion sphere.

119

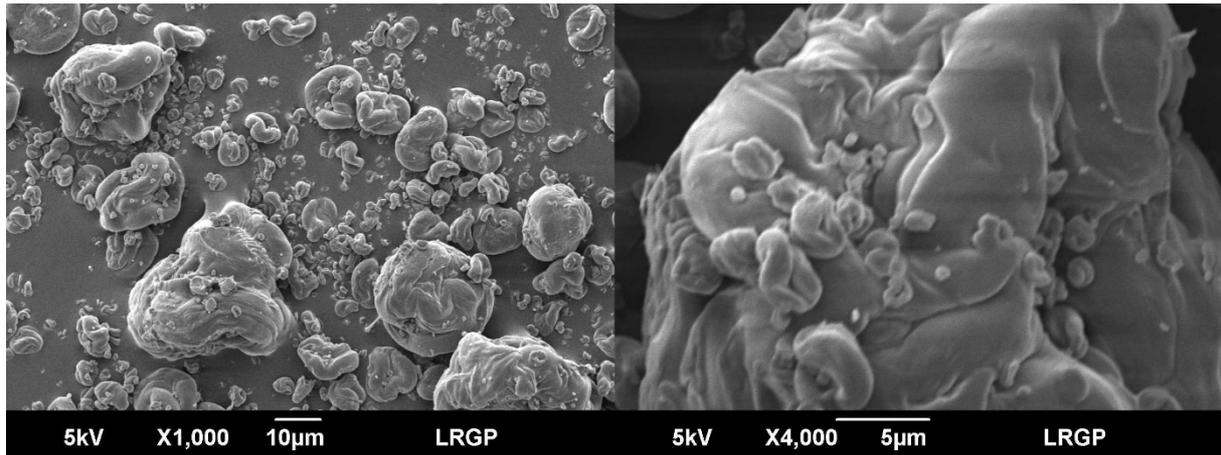
120 2. Materials and methods

121 2.1. Nanocellulose characterization

122 In this study, an organic powder was chosen with regard to experimental, scientific and
123 industrial considerations. First of all, the powder should display a sufficient ignition
124 sensitivity to be ignited by an electrical spark and thus ensure a single point ignition, which is
125 not the case of carbonaceous powders (Turkevich et al., 2016; Vignes, 2008). Moreover,
126 metal nanopowders may be subjected to pre-ignition and their flame tend to propagate with a
127 significant thermal radiation, which can hinder the flame front observation. Organic
128 nanopowders, with a minimum ignition energy of a few millijoules and a flame propagation
129 with limited thermal radiation, then appears to be a good solution. Moreover, during their
130 combustion, the devolatilization step occurs rapidly, which can lead to flame propagation
131 rather similar to those already observed for gas-powder hybrid mixtures (Cuervo, 2015) or
132 flame propagation occurring in the gas phase (Bradley and Lee, 1984) and makes possible the
133 application of relations initially established for such mixtures.

134 From an industrial point of view, nanocellulose is of increasing interest due to its specific
135 chemical and physical properties providing a wide variety of applications, notably in
136 photonics, medical devices, coating, electronics, 3D printing and plastics (Abitbol et al.,
137 2016). The powder used in this study is a cellulose nanocrystals powder (NCC from
138 CelluForce) with primary fiber dimensions of 3 nm width, an average length of 70 nm and a
139 specific surface area of $400 \text{ m}^2.\text{g}^{-1}$, as specified by the producer (CelluForce, 2016). Although
140 this powder is constituted of nanofibers, the word “nanoparticles” will be employed in this
141 work with no distinction between fibers and particles. The nanocellulose was dried at 90°C
142 under vacuum before performing tests to limit the influence of humidity on the explosion
143 parameters and improve reproducibility. Nanocellulose was observed by Scanning Electron
144 Microscopy (Figure 1) but did not reveal clearly the nanometric structure of the powder.

145 Additional experiments were performed with a rotative drum combined with a Condensation
146 Particle Counter (CPC – TSI 3007) and demonstrate the presence of two modes at 2 μm and
147 20 nm. Further tests were performed directly after powder dispersion in the explosion vessels
148 in order to highlight the presence of primary nanopowders in addition to the inevitable
149 agglomerates.



150

151 *Figure 1 Nanocellulose observed with an Scanning Electron Microscope*

152

153 2.2. Explosion tests equipment

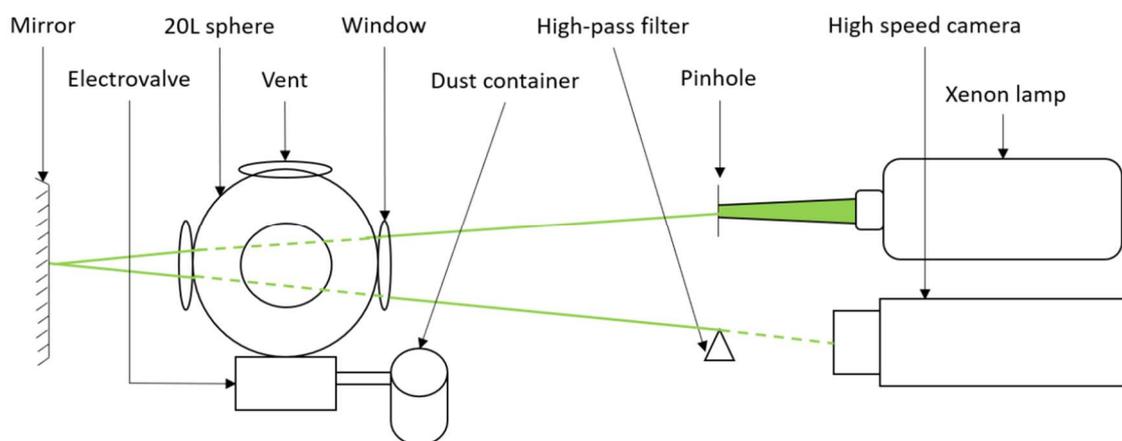
154 The unstretched burning velocity of nanocellulose was estimated using three experimental
155 methods. First, a flame propagation tube of 7 x 7 x 100 cm³ open at its upper end and
156 implementing the ignition system of the modified Hartmann tube, was coupled with a high-
157 speed video camera at 4000 fps with 240 μs of exposure time (Phantom V9.1). Electrodes are
158 located at 12.5 cm from the bottom of the tube. The equipment and procedure are fully
159 described by Cuervo et al. (2017) and Torrado et al. (2017) who validated the set-up and
160 procedure with methane and obtained encouraging results for hybrid mixtures. However, the
161 small volume, especially around the ignition zone, implies that the flame is influenced by the
162 walls quite soon after the ignition, which impacts its propagation. Therefore, the analysis is
163 limited to the first moments of the flame kernel growth, between ignition and the flame spatial

164 acceleration (or quenching) due to the presence of the wall. Consequently, a second method
165 was developed to study flame propagation within a modified 20L sphere.

166 The standard apparatus described by European standards (EN 14034-1, 2004; EN 14034-2,
167 2006) does not allow a clear visualization of the content of the sphere, preventing the
168 observation of both the dust cloud after dispersion and the flame propagation. To overcome
169 this limitation, Murillo (2016) designed a new 20L sphere equipped with four visualization
170 windows made of borosilicate with a diameter of 9.7 cm on the lateral sides and one on the
171 top to characterize the dust cloud in terms of particle size distribution and turbulence. Torrado
172 (2017) adapted the apparatus to perform hybrid mixtures explosion tests and to study the
173 flame propagation. Therefore, a lateral window was removed to integrate an ignition source
174 composed of two tungsten electrodes connected to a KSEP 320 system (Kühner AG - 15 kV /
175 15 mA, i.e. 225 W) generating a permanent spark whose duration, and thus energy, can be
176 varied. A modification of the Kühner software was necessary to control accurately the
177 ignition delay time, called 'tv', when using the permanent spark. A pressure relief valve,
178 opening at 1.15 atm (abs), was added on top of the sphere to protect the windows during the
179 explosion. The flame front and shape are imaged using a Schlieren system, combined with the
180 high-speed camera. The Schlieren setup consists of a 150 W xenon lamp (66475-150XV-R22
181 Xenon Light Source) equipped with a lens, a pinhole with a diameter of 1 mm to obtain a
182 single-point light source, and a spherical mirror reflecting the punctual light toward a filter
183 placed in front of the camera (Figure 2). This technique, notably described by Mazumdar
184 (2013), allows the observation of density variations, which provides a better visualization of
185 the flame front.

186 The results obtained with both apparatuses were then compared to the correlations established
187 by Silvestrini et al. (2008), based on the evolution of the maximum overpressure and the
188 maximum rate of pressure rise during an explosion carried out in a closed vessel. To ensure a

189 proper comparison, explosions tests were performed in the standard 20L sphere with a
190 permanent spark to avoid an overdriving phenomenon and a source ignition dependence of the
191 results. Tests were also performed on nanocellulose according to the standard procedure (EN
192 14034-1:2004 + A1:2011; EN 14034-2: 2006 + A11:2011) and 10 kJ chemical igniters. The
193 tube tests were performed at 300 mJ and in the 20L sphere, open and closed, an energy of 10J
194 was used, knowing the minimum ignition energy of the chosen dried nanocellulose for those
195 experiments is about 5 mJ as measured in a Mike 3 apparatus.



196

197 *Figure 2. Schematic of the Schlieren setup and the modified 20L sphere*

198

199 2.3. Dust cloud characterization

200 The particle size distribution of a dust cloud constituted of nanoparticles is one of the most
201 important factors influencing the flame propagation but also one of the most complex to
202 consider. Indeed, no apparatus or technique currently allows the determination at high
203 frequency and high concentration, i.e. a concentration equal or greater than the usual
204 minimum explosive concentrations (125 g.m^{-3} for the chosen nanocellulose), over a whole
205 range of particles sizes from 10 nm up to 200 μm .

206 To characterize the dust cloud, a laser diffraction sensor (Sympatec) was used through the
207 visualization window of the 20L sphere and the tube to measure the in-situ particle size
208 distribution (PSD) for particle sizes from 0.5 to 175 μm (R3 lens) at a frequency of 2 analyses
209 per millisecond. Since the combustion reaction can occur at the surface of the particles, as a
210 function of the reaction regime considered, the surface fraction is considered instead of the
211 usual volume fraction for the particle size distribution. It should be noted that other metrics,
212 such as the volume or number can also be considered to represent the particle size distribution
213 (Santandrea et al., 2019b).

214 In addition, the turbulence level of the dust cloud estimated by Dahoe et al. (2001) using a
215 laser Doppler anemometer was considered in the 20L sphere, and the measurements
216 performed by Particle Image Velocimetry (PIV) by Cuervo (2015) were used to estimate a
217 root-mean square velocity u_{rms} of the dust cloud in the flame propagation tube. Regarding the
218 values presented in Table 1, it appears that the turbulence level is always higher in the 20L
219 sphere than in the propagation tube, which is due to the high pressure applied during the
220 injection in the sphere and to the use of a dust canister. However, for both equipment, the
221 turbulence level decreases rapidly in the low ignition delay time range (from 60 to 120 ms,
222 71% lower in the 20L sphere and 39% in the tube) and seems to stabilize for high ignition
223 delay times (<200 ms), as discussed by Murillo et al. (2018) and Santandrea et al. (2019a).
224 Indeed, between 100 ms and 200 ms, the root-mean square velocity decreases 67% in the 20L
225 sphere and 60% in the tube, and only 14% in the 20L sphere and 25% in the tube between 300
226 ms and 400 ms. This “stable stage” allows to reach pseudo-laminar conditions and to
227 approach a laminar burning velocity when increasing sufficiently the ignition delay time.

228 *Table 1. Root mean square velocities measured in the 20L sphere and in the propagation tube*
229 *(Cuervo, 2015; Dahoe et al., 2001)*

230

Ignition delay time (ms)	u_{rms} in the 20L sphere ($\text{m}\cdot\text{s}^{-1}$) (Dahoe et al., 2001)	u_{rms} in the propagation tube ($\text{m}\cdot\text{s}^{-1}$) (Cuervo, 2015)
60	3.5	1.4
100	1.5	1.0
200	0.5	0.4
300	0.35	0.2
400	0.3	0.15

231

232 Explosion tests in the standard closed 20L sphere were performed according to EN 14034-
 233 1:2004 + A1:2011 and EN 14034-2:2006 + A11:2011 on the nanocellulose and on
 234 microcrystalline cellulose (Avicel, $d_{50} = 108 \mu\text{m}$) for comparison.

235 2.4. Assessment of unstretched flame velocity for nanopowders

236 The determination of the unstretched flame velocity was performed both by direct
 237 visualization of the flame propagation and by analyzing the pressure-time curves recorded
 238 during an explosion in the 20 L sphere.

239 2.4.1. Flame propagation experiments

240 2.4.1.1. Flame propagation and stretching observation

241 The propagation speed can be estimated using existing mathematical models based on several
 242 hypotheses (Chen and Ju, 2007; Frankel and Sivashinsky, 1984; Joulin and Clavin, 1979).
 243 The flame is then assumed to be spherically expanding and governed by an overall one-step
 244 exothermic reaction, and the thermodynamic properties of the mixture such as the molecular
 245 weight, the specific heat and the thermal conductivity are considered constant. The potential
 246 heat losses are neglected and the combustion process is assumed to be isobaric, which seems
 247 reasonable before the flame is affected by the vessel walls. With all these hypotheses, the
 248 laminar burning velocity can be estimated using a linear or a non-linear methodology, from
 249 the knowledge of the turbulent burning velocity and flame stretching.

250 To evaluate those parameters, flame propagation videos were analyzed by a model developed
 251 by Cuervo (2015) in MatLab’s Simulink using the Vision toolbox (Cuervo et al., 2017;
 252 Torrado et al., 2017). This model isolates the flame profile for each frame of the video as
 253 shown in Figure 3 and calculates the position of the flame front z , the estimated cross-section
 254 area A_s and the estimated flame surface A_f . In the case of flame kernels growing spherically,
 255 the flame radius is generally considered to estimate the spatial velocity (Law, 2006; Varea,
 256 2013). However, since the flame kernel appears to grow as an ellipsoid at low turbulence
 257 levels, the spatial velocity is calculated by deriving the position of the front flame.

258 Since the laminar flame velocity is independent of the vessel geometry and size, only the
 259 phase of ‘free’ flame kernel propagation is considered, i.e. when the flame is not significantly
 260 affected by the vessel walls, by compression or gas-wall heat transfers. It should be noted
 261 that, due to the contribution of the ignition source and the small size of the initial flame
 262 kernel, which can be located –with respect to the camera- behind unburnt particles, the very
 263 first milliseconds of the videos are also often delicate to analyze. The position of the flame z
 264 being known for different times, the spatial velocity of the flame S_s can be deduced. Then, the
 265 flame burning velocity S_u can be calculated using Andrews and Bradley (1972) formula:

$$266 \quad S_u = S_s \frac{A_s}{A_f} \quad (1)$$

267



268

269 *Figure 3. Time evolution of the flame profile in the semi-open tube for a dispersion of*
 270 *500 g.m⁻³ of nanocellulose at 340 ms ignition delay time*

271

272 This model is valid if the flame burning velocity S_u remains constant (which essentially
273 means that the local fuel equivalence ratio does not change during the flame propagation), if
274 the flame thickness is small in comparison to the flame curvature and if the spatial velocity S_s
275 is uniform over the whole surface of the flame. However, in the context of these experiments,
276 the ignition is performed 12.5 cm above the closed side of the tube, implying the hot burnt
277 gases are pushing up the fresh gases due to their thermal expansion. Therefore, a thermal
278 expansion factor χ , represented by the ratio between the temperatures of the hot burnt gases
279 and the initial cold gases, should be introduced to correct this increased velocity. The
280 temperature of the gases generated by the combustion reaction is approximated to be the
281 adiabatic temperature at equilibrium conditions and is determined by the CEA software
282 (McBride and Gordon, 1996). Equation (1) then becomes:

$$283 \quad S_u = \frac{S_s A_s}{\chi A_f} \quad (2)$$

284 However, as previously explained, only the “free” flame kernel propagation is considered,
285 meaning the analysis is stopped when the flame is close to the walls but is still unaffected by
286 their presence. This induces that, at this early stage, the flame kernel growth is unaffected by
287 the thrust of the gases against the bottom of the tube and by the pressure increase taken into
288 account by the thermal expansion factor.

289 It should be underlined that the burning velocity of a fuel-air mixture depends on the initial
290 dust concentration, the pressure and the temperature, but also the turbulence. More than any
291 other parameter, this point is very important and specific to dust explosions due to the
292 inherent turbulence developed when generating the dust cloud. This effect can be estimated
293 using the flame’s stretching factor K , called Karlovitz factor (Karlovitz et al., 1951), defined
294 as:

295
$$K = \frac{1}{A_f} \frac{dA_f}{dt} \quad (3)$$

296 The sign of the Karlovitz factor indicates the expansion (K positive) or compression (K
297 negative) of the flame surface area. Since the stretching can be due to the curvature of the
298 flame or to the strain rate of the flow, the stretching factor can be decoupled in two different
299 variables K_c and K_s (Bradley, 2000). However, in this work, the flame stretching will be
300 considered as a single phenomenon combining both effects.

301

302 In the literature, two relations to link the Karlovitz factor to the burning velocity are mainly
303 used by considering the assumptions previously mentioned: a linear relation and a non-linear
304 one. Although those relations were initially established for gases, Cuervo et al. (2017) and
305 Torrado et al. (2017) obtained encouraging results by applying them to starch/methane as well
306 as carbon nanoparticles/methane hybrid mixtures. In this work, the same relations are applied
307 to pure dust explosions. Indeed, the low inertia of nanocellulose particles allows the
308 measurement at very low turbulence, leading to a spherical/ellipsoidal flame kernel growth.
309 Moreover, the organic nature of the chosen powder induces a fast devolatilization which
310 implies that, under certain concentration and turbulence conditions, the dust combustion
311 would be controlled by gas combustion (Di Benedetto and Russo, 2007).

312 2.4.1.2. Linear relation

313 Most of the authors studying flame propagation and laminar burning velocity (Cuervo et al.,
314 2017; Dahoe et al., 2002; Di Benedetto et al., 2011) use a linear relation established for pure
315 gases and stating that, in a first approach, the Karlovitz factor can be linked to the flame
316 burning velocity by the following relationship (Clavin, 1985; Markstein, 1964):

317
$$S_u = -\delta_M K + S_u^0 \quad (4)$$

318 where S_u° is the unstretched burning velocity and δ_M , the Markstein length, whose sign
319 provides an indication on the stability of the flame (Clavin, 1985). This linear relation is valid
320 if the flame stretching is weak, i.e. if the Karlovitz number is low, and if the ratio of thermal
321 diffusivity to mass diffusivity, called the Lewis number, is equal to unity.

322 2.4.1.3. Non-linear relation

323 Even if the linear relation has been extensively adopted in various studies concerning flame
324 propagation (Beckmann et al., 2019; Bradley et al., 2019; Ichikawa et al., 2019; Torrado et al.,
325 2017), it has some limitations (Petersen and Emmons, 1961). Indeed, this relation is less
326 accurate for Lewis numbers different from unity (Tien and Matalon, 1991) and if the flame
327 stretching level is high, which corresponds to great values of the Karlovitz factor
328 (Vagelopoulos et al., 1994). As a consequence, some authors developed and applied a
329 nonlinear relation to link the unstretched flame velocity to the flame stretching of gaseous
330 mixtures (Buckmaster, 1977; Halter et al., 2010; Kelley and Law, 2009; Sivashinsky, 1975):

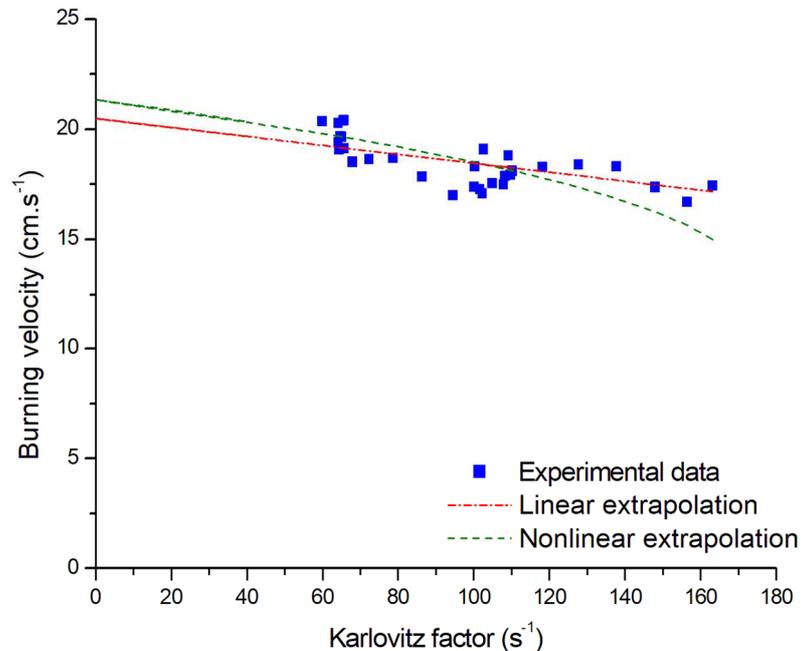
$$331 \left(\frac{S_u}{S_u^\circ}\right)^2 \cdot \ln\left(\frac{S_u}{S_u^\circ}\right)^2 = -\frac{2\delta_M K}{S_u^\circ} \quad (5)$$

332 The previous analysis was applied along with relations 4 and 5 to the experimental data
333 recorded during flame propagation in the semi-open tube and in the vented 20L sphere.

334 2.4.1.4. Estimation of the unstretched burning velocity

335 An illustration of the evolution of the burning velocity with the Karlovitz factor is presented
336 in Figure 4 for a concentration of 500 g.m^{-3} of nanocellulose. It appears that both relations
337 seem to match approximately the experimental data. In this example, the unstretched flame
338 velocity obtained using the linear relation is $20.5 \pm 0.6 \text{ cm.s}^{-1}$ and $21.3 \pm 0.6 \text{ cm.s}^{-1}$ using the
339 nonlinear relation proving a good consistency between both methods. It should be noted that
340 the average discrepancy between the flame velocities obtained by both methods is 4 %, which

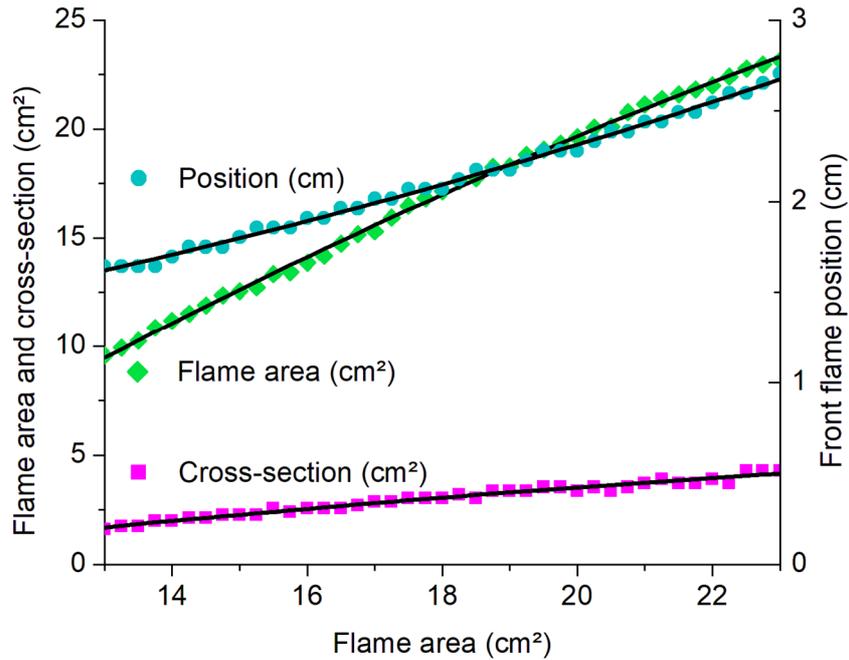
341 means that there is no statistical difference between the methods. However, it appears in
342 Figure 4 that the scattering of data is far from being negligible, which is notably due to
343 uncertainties in the flame surface determination at each step of the propagation and hinders
344 sometimes the application of such correlation.



345

346 *Figure 4. Illustration of the burning velocity – stretching factor relation for a $500 g.m^{-3}$*
347 *nanocellulose-air mixture for $t_v = 340 ms$*

348 Since the derivation process used to determine the Karlovitz factor and the spatial velocity
349 can induce some uncertainties, the time evolution of the position of the flame front, the flame
350 area and cross-section was systematically smoothed based on a second order polynomial as
351 shown in Figure 5 to obtain a general trend and avoid local discontinuities. The coefficient of
352 determination is systematically at least 0.99, showing a good agreement with the experimental
353 results.



354

355 *Figure 5. Evolution of the front flame position, flame area and cross-section with time for a*

356

500 g.m⁻³ nanocellulose-air mixture for $t_v = 340$ ms

357 Figure 6 then represents the evolution of the burning velocity with the stretching factor after

358 data smoothing, showing a better agreement with the different theoretical relationships than

359 the initial raw experimental data. Indeed, the unstretched burning velocity now reaches $23.1 \pm$

360 0.3 cm.s^{-1} with a coefficient of determination of 0.95 instead of 0.36 in the previous case with

361 the linear fitting, and it reaches $21.8 \pm 0.4 \text{ cm.s}^{-1}$ using the nonlinear fitting. Since the

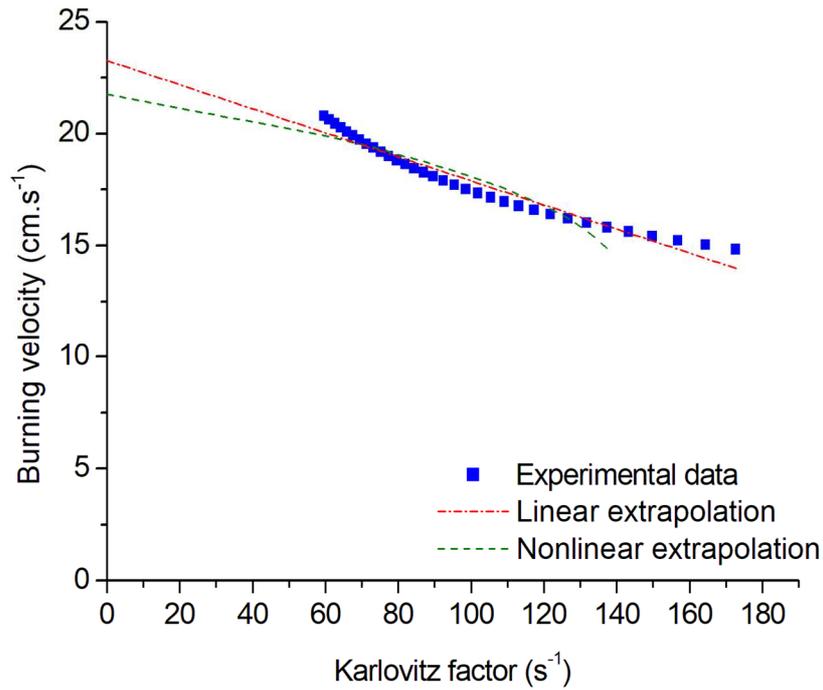
362 smoothing provides results with a better accuracy, it was systematically realized during the

363 videos analysis phase.

364 Videos of the explosion occurring in the flame propagation tube and in the vented 20L sphere

365 were then analyzed according to the method described in 2.4.1 and the unstretched burning

366 velocity was estimated using both linear and nonlinear relations.



367

368 *Figure 6. Illustration of the burning velocity – stretching factor relation after smoothing for a*

369 *500 g.m⁻³ nanocellulose-air mixture for $t_v = 340$ ms*

370

371 2.4.2. Pressure-time evolution interpretation

372 An alternative way to estimate the unstretched burning velocity is provided through the

373 recording of the pressure evolution during an explosion in the 20L sphere. Indeed, some

374 authors such as Silvestrini et al. (2008) developed some correlations between the unstretched

375 burning velocity and the parameters K_{St} and P_{max} obtained in the 20L sphere. The results

376 obtained by analyzing the flame propagation will be compared to the ones calculated from the

377 following semi-empiric correlation defined by Silvestrini et al. (2008):

$$378 \quad S_u^0 = 0.11 \frac{K_{St}}{P_{max} \left(\frac{P_{max}}{P_0} \right)^{0.14} \left(\frac{P_{max}}{P_0} \right)^{\frac{1}{\gamma}}} \quad (6)$$

379 where P_0 is the atmospheric pressure and γ the ratio of specific heats. As the previously

380 defined relations, this correlation is based on several assumptions, such as the spherical

381 expansion of the flame and the neglecting of the turbulent length scales. Moreover, the
 382 establishment of this correlation based on other existing relations also relying on hypotheses,
 383 like a laminar flow or the fact that the burnt gases are trapped behind the expanding flame
 384 front, meaning the thermal expansion factor is considered (Harris, 1983). Furthermore, the
 385 explosivity index K_{St} is mentioned in the correlation, meaning that the “cube-root” law and its
 386 related hypotheses were used. For instance, the flame front is considered to be a thin reaction
 387 zone as defined by Dahoe et al. (1996), and Silvestrini et al. (2008) considered that P_{max} was
 388 defined as the maximum overpressure for a single explosion experiment and that a K_{st}
 389 parameter can be defined from a test performed at a single dust concentration and not from
 390 tests performed over a wide range of concentration. Thus, the following relation was applied
 391 during this work:

$$392 \quad S_u^0 = 0.11 \frac{\left(\frac{dP}{dt}\right)_m V^{1/3}}{P_m \left(\frac{P_m}{P_0}\right)^{0.14} \left(\frac{P_m}{P_0}\right)^{\left(\frac{1}{\gamma}\right)}} \quad (7)$$

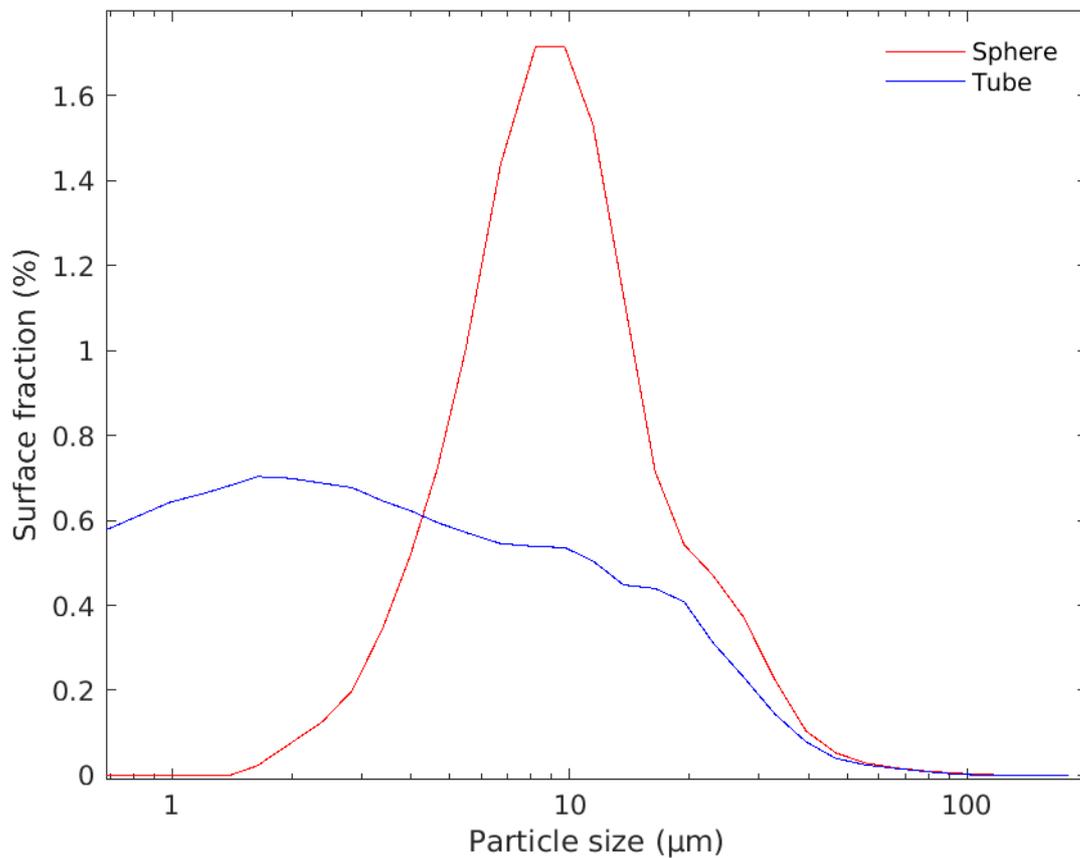
393 The unstretched burning velocity of nanocellulose was then estimated using those three
 394 methods to test the strengths and weaknesses of each method and ensure the consistency of
 395 the methods by identifying a reliable value of the laminar burning velocity.

396 **3. Results and discussion**

397 3.1. Particle size distribution of the dust cloud

398 When characterizing the dust cloud, it first appears that the particle size distribution in the
 399 sphere seems to be more monodisperse than in the tube due to the high shear stress occurring
 400 during the injection of the powder and potentially breaking the agglomerates of a few
 401 micrometers (Figure 7). Although particles with a diameter lower than 0.5 μm seem to be
 402 present in the flame propagation tube and probably in the sphere, they are not measured
 403 directly with this lens.

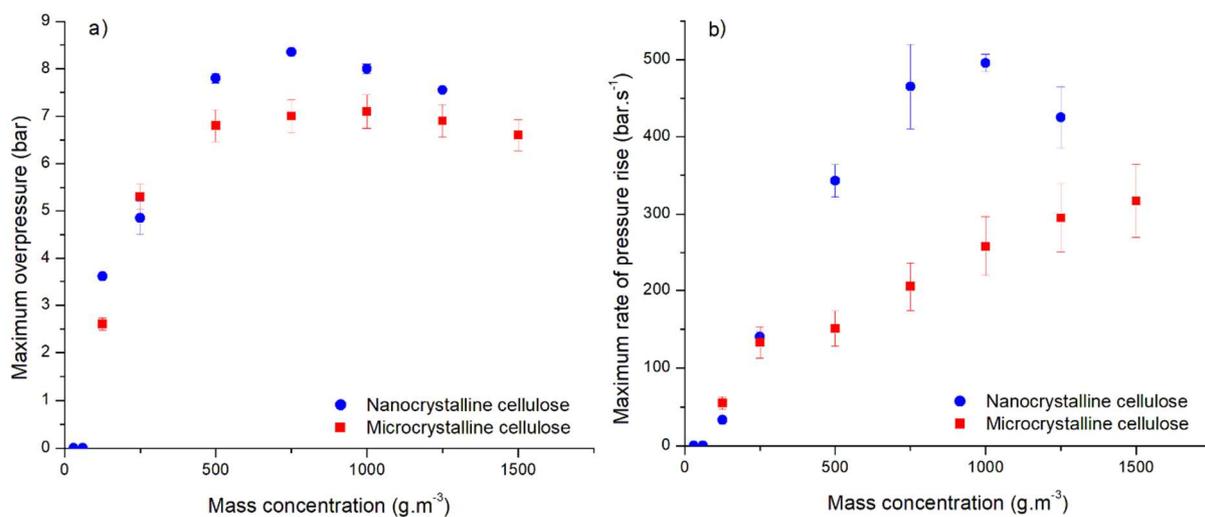
404 Similar tests were also performed with the same sensor equipped with a R1 lens (from 0.1 to
405 35 μm). In addition, the PSD of the dust clouds was investigated using a Fast Mobility
406 Particle Sizer (FMPS) and a Scanning Mobility Particle Sizer (SMPS) to specifically measure
407 particles with diameters lower than 500 nm in the 20L sphere. It should be underlined that
408 such apparatuses allow the determination of the electrical mobility distribution of the dust
409 cloud. Although it is impossible with those methods to directly compare the PSD of the dust
410 in the sphere and the tube, these experiments clearly confirm the presence of nanoparticles
411 after dispersion in both equipment with dimensions ranging from 100 to 300 nm (with peaks
412 at 150 nm).



413
414 *Figure 7. Evolution of the particle size distribution of nanocellulose agglomerates 200 ms*
415 *after dispersion in the 20L sphere and in the tube at the ignition location*

416
417 3.2. Explosion severity of micro and nanocellulose

418 Before proposing an alternative method for dust explosivity assessment, the standard test
 419 method (EN 14034) was applied to compare the explosion severity of micro and
 420 nanocellulose. It can be observed that the maximum overpressure P_m (Figure 8a) of the
 421 nanocellulose is slightly greater than that of microcrystalline cellulose, but that the overall
 422 behavior of this parameter as a function of the dust concentration is rather similar for both
 423 compounds. However, the difference is more significant with respect to the maximum rate of
 424 pressure rise $(dP/dt)_m$ (Figure 8b), which can be explained by the variation of particle size and
 425 of specific surface area, which greatly impacts the reaction kinetics. The K_{st} parameter,
 426 obtained through the application of the cubic law, reaches $135 \pm 3 \text{ bar.m.s}^{-1}$ for nanocellulose
 427 at 1000 g.m^{-3} , whereas it reaches only $86 \pm 10 \text{ bar.m.s}^{-1}$ at 1500 g.m^{-3} for microcrystalline
 428 cellulose.

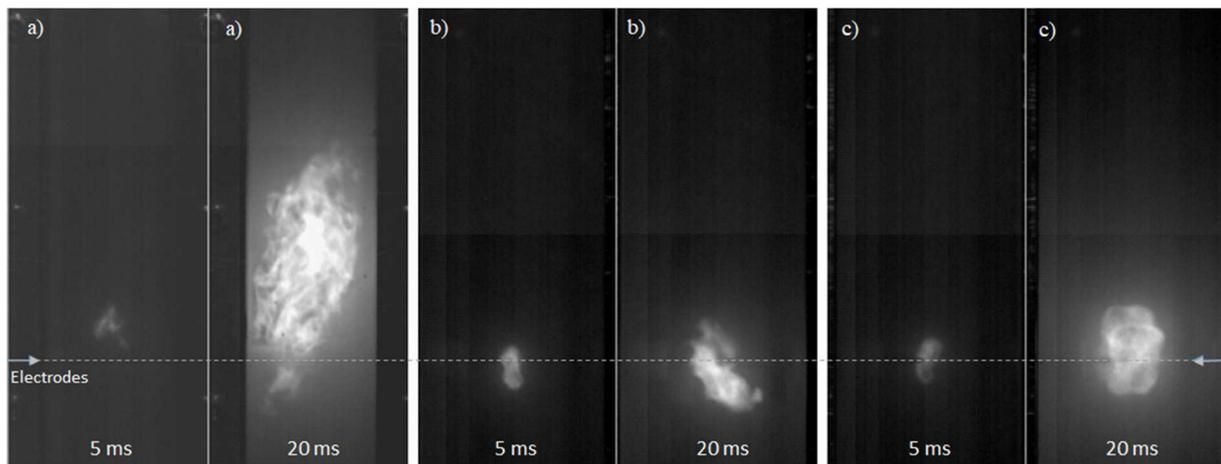


429
 430 *Figure 8. a) Maximum overpressure and b) maximum rate of pressure rise of nanocellulose*
 431 *and microcellulose in standard test conditions*

432 3.3. Determination of the optimal operating conditions for flame propagation tests

433 Explosion tests were first performed at different ignition delay times and at a single
 434 concentration of 500 g.m^{-3} , estimated in the tube by considering the dust weight and an
 435 average dispersion height of 0.35 cm. This concentration is close to the stoichiometric

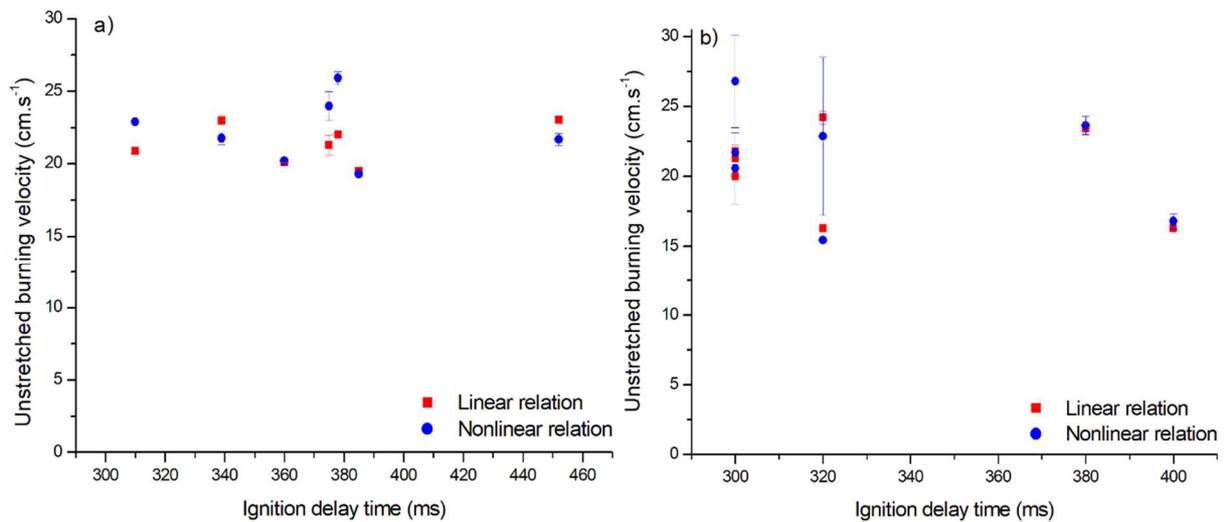
436 concentration (around 250 g.m^{-3}) and ensures a visualization of the flame kernel without
 437 interference coming from the particles, which occurs with larger concentrations. At high
 438 turbulence, i.e. at an ignition delay time lower than 200 ms, the flame kernel grows up rapidly
 439 with stretching and is rapidly influenced by the walls (Figure 9a). As a consequence, only a
 440 few milliseconds of the video are suitable for the analysis, and the flame profile is difficult to
 441 identify due to the turbulence. When slightly reducing the turbulence, the flame kernel growth
 442 is slower, but the turbulence level remains too decisive ($u_{\text{rms}} > 1 \text{ m.s}^{-1}$) to precisely define the
 443 flame profile (Figure 9b). At very low turbulence, the flame kernel growth is still slow and
 444 nearly spherical, showing a behavior similar to that of gases, i.e. a rather smooth flame
 445 surface and a flame front with a paraboloidal shape (Figure 9c). It is then possible to correctly
 446 identify the flame profile, with 10 to 20 ms of video which can be confidently analyzed.
 447 Moreover, it can be noticed that the flame kernel does not move when growing, which shows
 448 that it is barely affected by the dust cloud inertia or by the burnt gases thrust. As a result of
 449 these observations, ignition delay times from 300 ms to 450 ms, due to a limitation of the
 450 apparatus, were considered in the tube and in the 20L sphere for the analyses.



451
 452 *Figure 9. Visualization of flame propagation of 500 g.m^{-3} of nanocellulose in the tube 5 ms*
 453 *and 20 ms after ignition for different ignition delay times (a) 125 ms (b) 235 ms (c) 450 ms.*

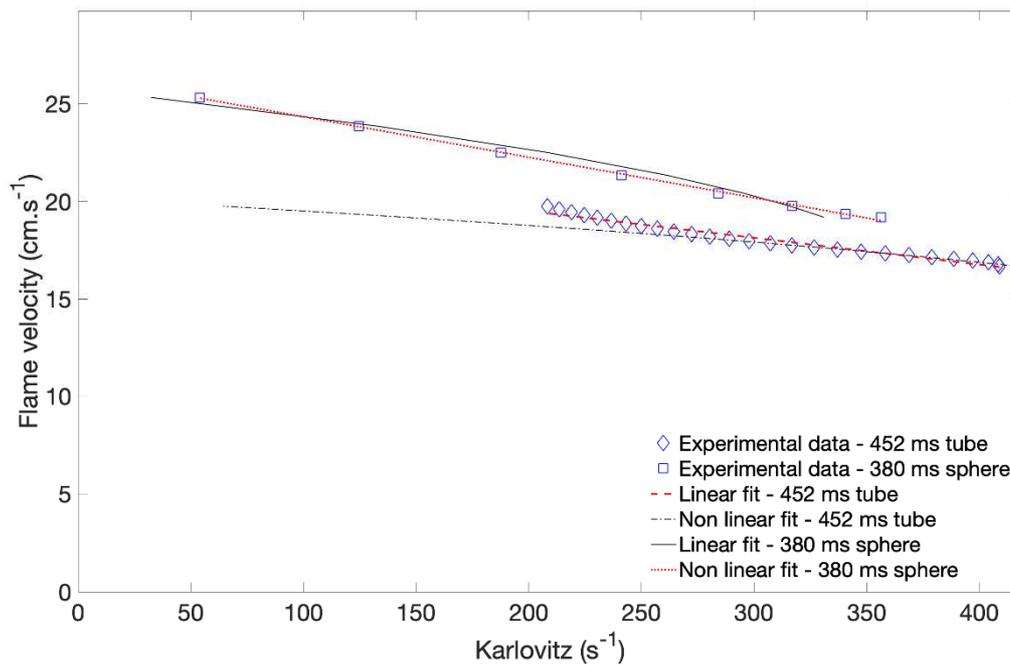
454

455 Figure 10 presents the results obtained in the flame propagation tube and in the open 20L
 456 sphere at different ignition delay times, calculated by linear and nonlinear relations. It appears
 457 in Figure10a that unstretched burning velocities between 18 and 23 cm.s⁻¹ are obtained using
 458 the linear equation and that velocities ranging between 17.5 and 26 cm.s⁻¹ are obtained with
 459 the nonlinear relation, which remains in the same order of magnitude. Both methods lead to
 460 similar values, even if the dispersion of the experimental data seems to be more significant in
 461 the case of the nonlinear equation. Moreover, Figure 10b shows that the analytical
 462 uncertainties are higher when applying the nonlinear relationship to the experimental data
 463 measured in the open 20L sphere.



464

465 *Figure 10. Evolution of the unstretched burning velocity of 500 g.m⁻³ of nanocellulose with*
 466 *ignition delay time calculated by the linear and nonlinear relations in a) the propagation tube*
 467 *b) the open 20L sphere*



468

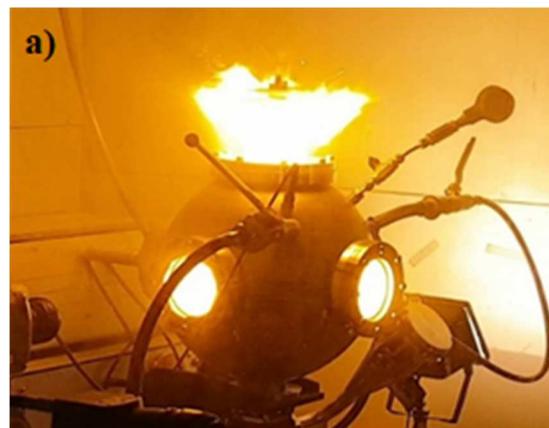
469 *Figure 11. Comparison of the application of linear and nonlinear methods to a nanocellulose*
 470 *explosion: 500 g.m⁻³ concentration, tv=452 ms – tube and tv=380 ms – vented sphere*

471

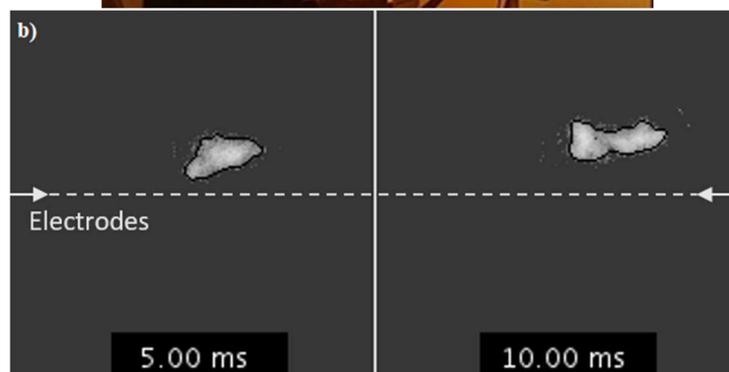
472 Figure 11 shows that the application of the nonlinear relationship is more relevant for the
 473 experimental data corresponding to the high-stretch region (from 270 to 400 s⁻¹) of the flame
 474 propagation in the tube. However, this zone corresponds to a period of time during which the
 475 flame propagation may still be affected by the ignition (near 400 s⁻¹). In most cases, as for
 476 instance for the other experimental set of data presented in Figure 11 (380 ms in the open
 477 sphere), the linear fit will be preferred to the nonlinear model, especially at low stretching
 478 rates. It can also be observed in Figure 11 that the number of experimental points selected for
 479 the analysis is often reduced in the case of the open sphere. Indeed, with regard to the tube,
 480 the larger volume of the 20 L open sphere is both an advantage and a drawback: an advantage
 481 because it reduces the wall effects and a drawback because, during the very first moments of
 482 the flame kernel growth, the thickness of the unburnt cloud located between the ignition zone

483 and the window is great, which hinders the flame visualization and sometimes reduces the
484 ‘analyzable duration’ of the video down to 5 ms. This limitation can be partially overcome by
485 using a Schlieren system, as presented in paragraph 2.2. An explosion in the 20L sphere and
486 the dust kernel 5 ms and 10 ms after ignition of 500 g.m^{-3} of nanocellulose are presented in
487 Figure 12a and Figure 12b respectively. In view of the experimental uncertainties and the
488 slight discrepancies between the non-linear and linear relationships, only the results obtained
489 by the latter method will be now considered.

490



491



492

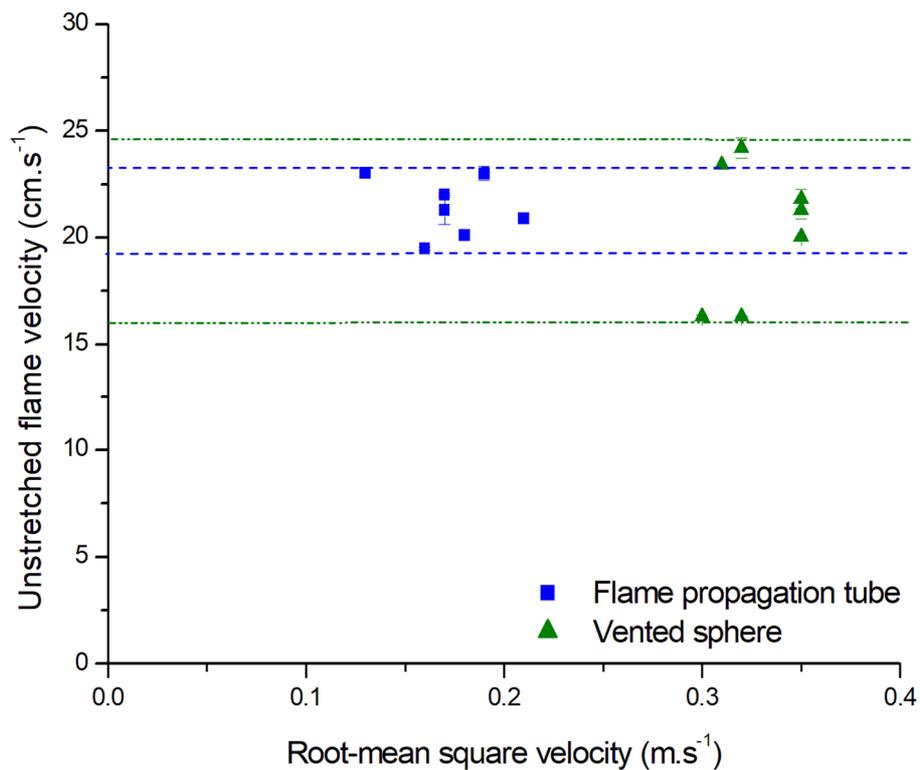
493 *Figure 12. a) Explosion in the open 20L sphere and b) visualization of the flame kernel 5 ms*
494 *and 10 ms after ignition of 500 g.m^{-3} of nanocellulose and ignition delay time of 300 ms*

495

496 3.4. Estimation of the laminar burning velocity

497 Since the turbulence intensity is different in the tube and in the sphere, results were plotted as
498 a function of the root mean square velocity measured in both equipment by Particle Image

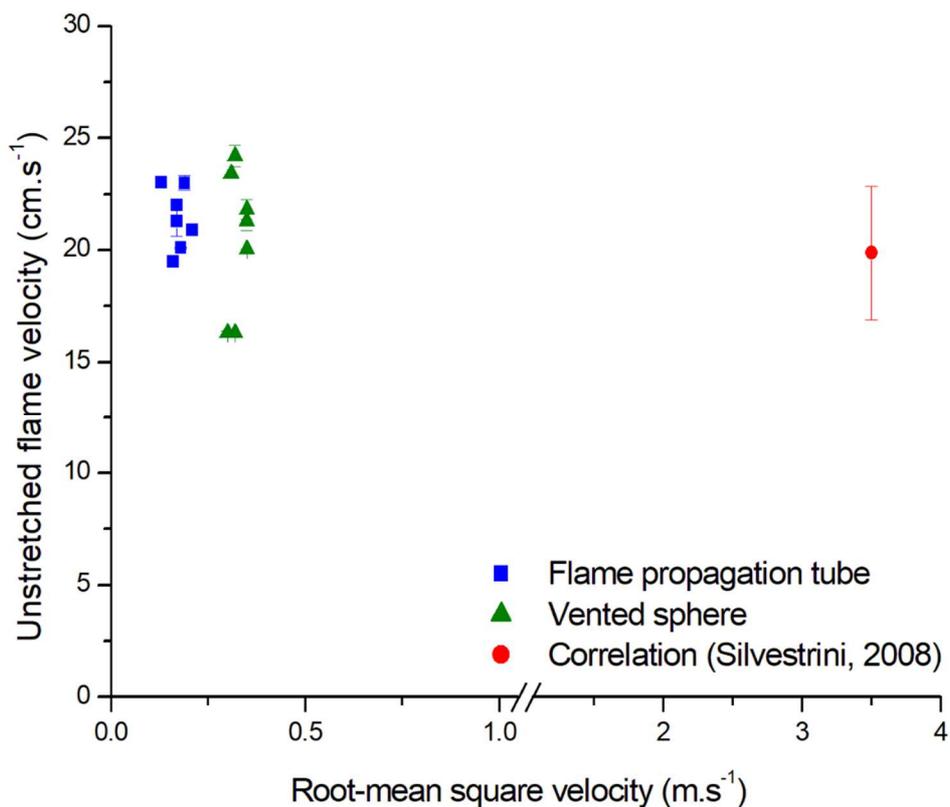
499 Velocimetry. Figure 13 then shows that a very low turbulence level, from 0.21 to 0.13 m.s^{-1} is
 500 reached in the tube, whereas it reaches a minimum of 0.3 m.s^{-1} in the 20L sphere. However,
 501 similar values of unstretched burning velocity are obtained, from 19 to 23 cm.s^{-1} in the tube
 502 and from 16 to 24 cm.s^{-1} in the sphere. This is due to the fact that, when increasing the
 503 ignition delay time, the turbulence level decreases in both equipment until it reaches a region
 504 in which the decay is small, as mentioned in paragraph 2.3. This region is observed from 200
 505 ms in both equipment, corresponding to a root-mean square velocity of 0.4 m.s^{-1} in the
 506 propagation tube and 0.5 m.s^{-1} in the 20L sphere. The flame propagation tube seems to
 507 produce more clustered values, which can be explained by the better visualization of the flame
 508 kernel providing a longer analyzable video than in the vented 20L sphere, thus a more
 509 accurate analysis of the flame propagation.



510
 511 *Figure 13. Evolution of the unstretched burning velocity with the root mean square velocity in*
 512 *the flame propagation tube and in the open 20L sphere*

513

514 The values of the unstretched burning velocities obtained through visualization of the flame
 515 propagation were then compared to the value acquired by pressure-time recording in the
 516 standard 20L apparatus (Figure 14). Using the semi-empirical correlation previously detailed
 517 (Equation 7), an unstretched burning velocity of 19.9 cm.s^{-1} is obtained at a root mean square
 518 velocity of 3.5 m.s^{-1} while the same value was reached in the flame propagation tube and
 519 vented 20L sphere for root mean square velocities of 0.2 m.s^{-1} and 0.3 m.s^{-1} respectively. This
 520 is due to the fact that the empirical coefficient of the correlation was established by
 521 considering explosion tests performed in standard conditions in the 20L sphere. The equation
 522 was applied with a coefficient 0.11 as proposed by the authors, but it makes it valid only for
 523 $(dP/dt)_m$ and P_m obtained with an ignition delay time of 60 ms, i.e. a root-mean square
 524 velocity of 3.5 m.s^{-1} , as previously presented in Table 1.



525
 526 *Figure 14. Evolution of the unstretched burning velocity with the root mean square velocity*
 527 *measured in the flame propagation tube, the vented 20L sphere and the standard 20L sphere*

528

529 The laminar burning velocity was then calculated by application of Silvestrini's correlation
530 (eq. 6) to the parameters P_m and $(dP/dt)_m$ obtained with a standard explosion in the 20L
531 sphere, with an ignition energy of 10J and ignition delay time of 60 ms. Moreover, since
532 Cuervo (2015) and Dahoe et al. (2001) showed that the vertical velocity of the particles is
533 close to zero for ignition delay times greater than 300 ms in the propagation tube and in the
534 20L sphere, and since the flame kernel stays put when growing, the flame propagation
535 visualized in both equipment is considered to be independent of the turbulence level, and the
536 laminar burning velocity was then estimated by averaging the measured values of unstretched
537 burning velocities. However, in addition to the difficulties to identify the flame kernel in the
538 vented sphere, significant fluctuations in the velocity in the sphere can explain the more
539 important scattering of the values of the unstretched burning velocities obtained compared to
540 the ones obtained in the tube. The values of laminar burning velocity of the nanocellulose
541 determined for each method are presented in Table 2.

542

543 *Table 2. Laminar burning velocity of the nanocellulose estimated by the three different*
544 *methods*

<i>Method</i>	<i>Flame propagation tube (flame visualization)</i>	<i>Open 20L sphere (flame visualization)</i>	<i>Standard 20L sphere (pressure evolution)</i>
<i>Laminar burning velocity ($cm.s^{-1}$)</i>	21.4 ± 1.4	20.5 ± 3.2	19.9 ± 3.0

545

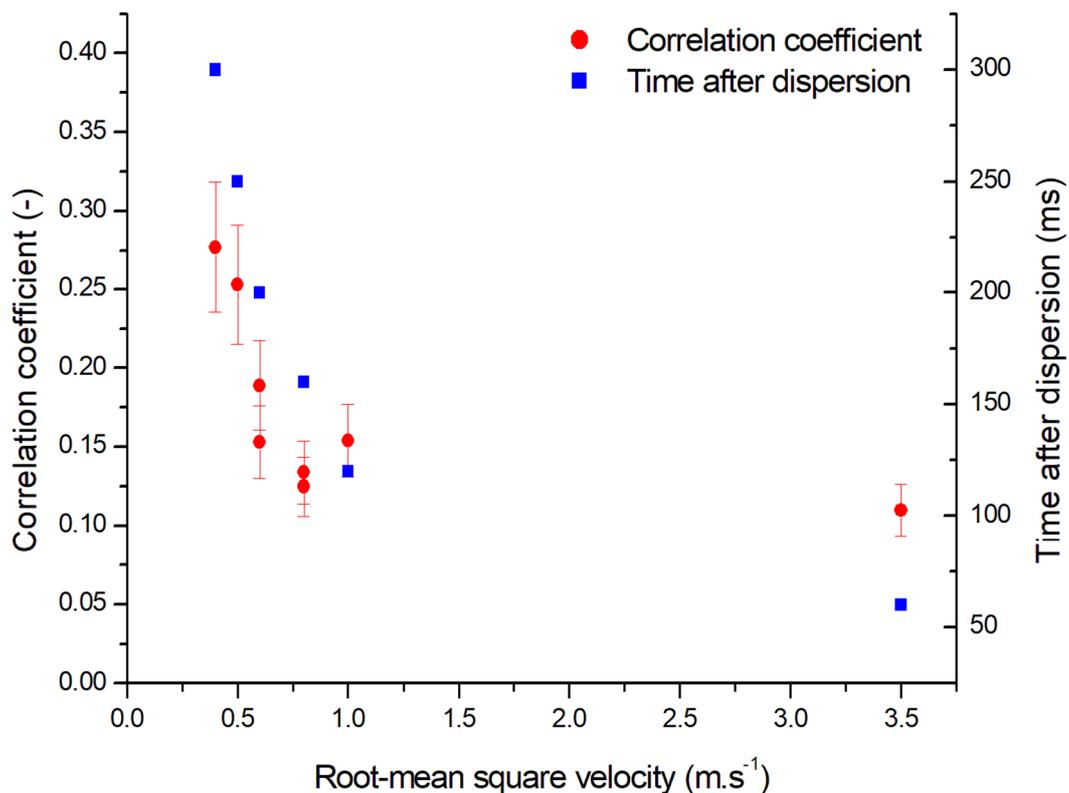
546

547 It first appears that the three methods lead to similar results, the propagation tube providing
548 the less scattered values of laminar burning velocity. The calculation performed using
549 Silvestrini et al. (2008) correlation also provides a value in good agreement with the values
550 measured by flame propagation analysis, but with a higher uncertainty than in the propagation

551 tube. It is mainly due to the lack of reproducibility of the experiments in the standard 20L
552 sphere at 60 ms (Proust et al., 2007). Indeed, at an ignition delay time of 60 ms, the turbulence
553 level is in the “transition stage” identified by Murillo et al. (2018), and turbulence variations
554 have a strong impact on the maximum rate of pressure rise. Adapting the correlation with a
555 coefficient estimated from experiments performed in pseudo-laminar conditions, i.e. higher
556 ignition delay time, would then improve the accuracy of the estimation of the laminar burning
557 velocity. It would then be interesting to use flame propagation analysis on different powders
558 to determine such a coefficient and to apply it to the correlation established by Silvestrini et
559 al. (2008). However, this adapted correlation would have to be used with P_m and $(dP/dt)_m$
560 measured in the same turbulent conditions than the coefficient, which can limit the application
561 of such a relation. To apply this correlation to different conditions and represent industrial
562 situations, an evaluation of the evolution of the correlation coefficient with the initial
563 turbulence is then required.

564 Since the correlation has been validated for nanocellulose at 60 ms, a first estimation of the
565 coefficient dependency with the turbulence was realized through explosion tests conducted in
566 the standard closed 20L vessel by varying the ignition delay time from 60 ms to 300 ms. By
567 applying the correlation used previously and considering the value obtained at 60 ms as a
568 reference, the corrected correlation coefficients were estimated for different root-mean square
569 velocities, as presented in Figure 15. It appears that the value to consider to obtain the laminar
570 burning velocity of 19.9 cm.s^{-1} increases when decreasing the root-mean square velocity,
571 reaching 0.28 at 0.4 m.s^{-1} , i.e. more than twice the initial value of 0.11 at 3.5 m.s^{-1} . The
572 correlation coefficient follows the same evolution as the time after dispersion with the root-
573 mean square velocity, which implies that it could be correlated by a decay law similar to the
574 one established by Dahoe et al. (2001) relating the root-mean square velocity and the time.
575 The determination of such a relation, coupled with a correlation linking the evolution of

576 $(dP/dt)_m$ with the initial turbulence, could then enable an accurate determination of the
577 laminar burning velocity in the closed 20L sphere.



578

579 *Figure 15. Evolution of the time after dispersion and the coefficient from Silvestrini's*
580 *correlation with the root-mean square velocity in the 20L sphere*

581

582 Krause and Kasch (2000) reported laminar burning velocities presented in the literature for
583 different concentrations of two organic powders: lycopodium and cornstarch. Values from 17
584 cm.s^{-1} to 69 cm.s^{-1} were then obtained for lycopodium, and velocities between 13 cm.s^{-1} and
585 59 cm.s^{-1} were acquired for cornstarch. More specifically, van der Wel (1993) reported a
586 laminar flame velocity for cornstarch of 13 cm.s^{-1} at 400 g.m^{-3} using the 20L sphere coupled
587 with a correlation based on Mallard and Le Chatelier thermal theory, and 13 to 20 cm.s^{-1}
588 using the burner method for concentrations from 400 g.m^{-3} to 600 g.m^{-3} . Haghiri and Bidabadi
589 (2010) also calculated laminar burning velocities for organic powders from 15 cm.s^{-1} to 45

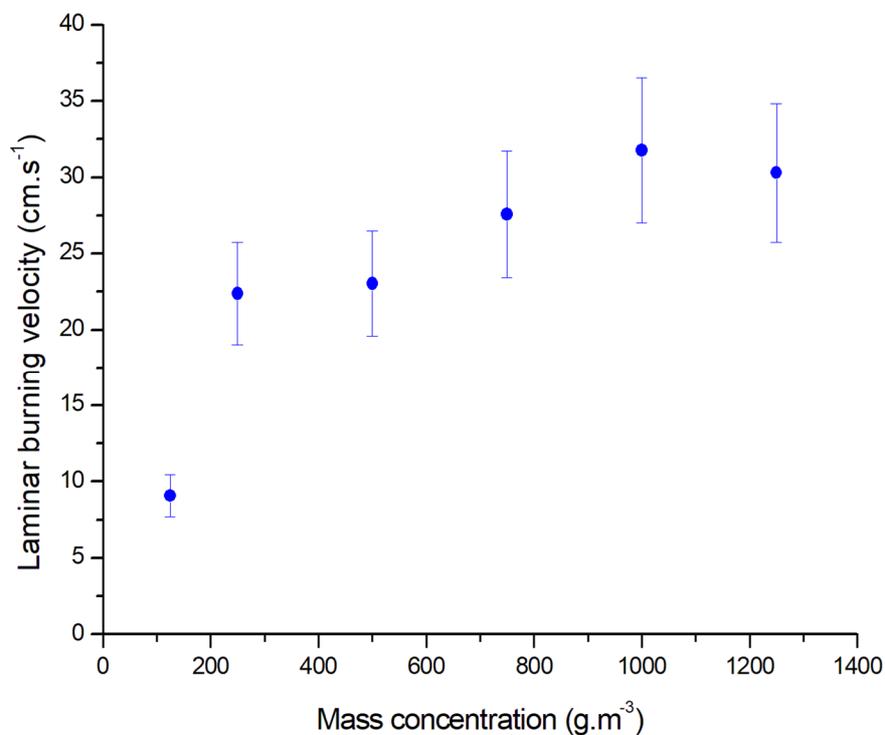
590 cm.s^{-1} for concentrations from 30 g.m^{-3} to 100 g.m^{-3} using a model considering the thermal
591 radiation effect. Moreover, Cuervo et al. (2017) also measured a laminar burning velocity for
592 915 g.m^{-3} of starch around 19.9 cm.s^{-1} . Since starch and nanocellulose have similar structures
593 and composition, these values are in good agreement with the value of 21.4 cm.s^{-1} obtained in
594 the tube for the nanocellulose, despite the different concentrations.

595 The Markstein length was also calculated from video analysis of the flame propagations in the
596 tube and the vented sphere. Similar values were obtained in both equipment, with a mean
597 value of -0.25 mm in the propagation tube and -0.19 mm in the vented sphere. A negative
598 value of the Markstein length means that the flame speed increases with the stretch rate,
599 implying that the flame is unstable. However, the obtained values are very low (absolute
600 values usually lower than 1 mm), and some positive values up to 0.3 mm were also obtained
601 when analyzing the flame propagation videos under certain conditions, which makes it
602 difficult to draw an accurate conclusion on the Markstein length of nanocellulose flames.
603 Nevertheless, it should be stressed that these values are much lower than that obtained by
604 Dahoe et al. (2002) for cornstarch-air mixtures, i.e. 11.0 mm , but are a bit higher than those
605 encountered for methane-air mixtures, i.e. from 0.1 to 0.2 mm . This can be related to the fact
606 that nanocellulose is prone to devolatilization when exposed to high temperatures.

607 3.5. Influence of the dust concentration

608 Even though the theoretical stoichiometric concentration for the combustion of nanocellulose
609 is of approximately 250 g.m^{-3} , it appears in Figure 8 that the most severe explosions were
610 obtained for concentrations of 750 g.m^{-3} and 1000 g.m^{-3} in terms of maximum overpressure
611 and maximum rate of pressure rise. It is then legitimate to investigate the influence of the dust
612 concentration on the laminar burning velocity. However, the estimation of the flame
613 propagation at high concentration is difficult since the dust cloud can hinder the visualization
614 of the flame kernel. A few tests were performed in the flame propagation tube at larger dust

615 concentrations (i.e. 750 g.m^{-3}) and a slight increase of the flame velocity was observed.
616 However, since it is currently impossible to accurately estimate the laminar burning velocity
617 at high concentration using the flame propagation visualization method due to flame
618 obscuring, it was calculated as previously, using the correlation established by Silvestrini et
619 al. (2008) coupled with the standard results presented in Figure 8. It has to be noticed that
620 those experiments were carried out with chemical ignitors of 10 kJ, contrary to the results
621 presented in Figure 14 carried out using an electrical spark of 10 J, which limits the impact of
622 the shock wave created by the ignitors.



623
624 *Figure 16. Evolution of the laminar burning velocity calculated by a semi-empiric correlation*
625 *with the mass concentration*

626
627 Since the laminar burning velocity is calculated from the knowledge of the maximum
628 overpressure and the maximum rate of pressure rise, it follows the same evolution as those
629 parameters (Figure 16). The maximum value is obtained around 32 cm.s^{-1} for a concentration
630 of 1000 g.m^{-3} . This value is close to the laminar burning velocity of stoichiometric methane

631 generally reported to be around 35 cm.s^{-1} (Bradley et al., 2017) for a maximum overpressure
632 of 7.2 bars and a K_g of 395 bar.m.s^{-1} , measured in standard conditions by Torrado et al.
633 (2017a). The value calculated for the nanocellulose with the correlation then seems higher
634 than expected considering its medium explosion severity compared to pure methane, thus
635 questioning the validity of the correlation for high concentrations. Indeed, Silvestrini et al.
636 (2008) have chosen 24 dust-air mixtures to calculate the coefficient for the semi-empiric
637 correlation. However, it should be stressed that, among these mixtures, only 3 dust
638 concentrations were higher than 500 g.m^{-3} . The same method considering mixtures with a
639 larger range of concentrations may then extend the validity of the correlation to higher
640 concentrations. Indeed, in order to realize an accurate evaluation of the explosion
641 consequences, the influence of the dust concentration on the laminar burning velocity must be
642 considered. The value corresponding to the worst-case scenario could then be loaded in
643 simulation and by adding the contribution of the turbulence of the dust cloud, the
644 consequences of an explosion in specific conditions can be estimated. It implies that the
645 turbulent burning velocity has to be deduced from the knowledge of the laminar burning
646 velocity and the turbulence level.

647 3.6. Application of turbulent burning velocity models

648 Several correlations relating the turbulent burning velocity to the laminar burning velocity and
649 the turbulence intensity exist in the literature, and were notably summarized by Andrews et al.
650 (1975), Dahoe et al. (2013) and Gülder (1991). Although these models were established for
651 premixed flames, some of the correlations presented by Dahoe et al. (2013) were tested in this
652 study to be compared, in a reverse approach, to the values of turbulent burning velocities
653 obtained by analyzing the flame propagation in the tube (Table 3). In addition to those
654 correlations, the relation established by Popat et al. (1996) used in the FLACS-DustEx CFD

655 code (relation (g)) was applied to the experimental data produced in this work (Ghaffari et al.,
656 2019; Skjold, 2007).

657 *Table 3. Some turbulent burning velocity models for premixed flame propagation*

Notation	Formula	Reference
(a)	$\frac{Su_T}{Su_L} = 1 + \frac{u_{rms}}{Su_L}$	(Damkoehler, 1947)
(b)	$\frac{Su_T}{Su_L} = \sqrt{1 + \left(\frac{u_{rms}}{Su_L}\right)^2}$	(Shelkin, 1947)
(c)	$\frac{Su_T}{Su_L} = 1 + \sqrt{\frac{5}{12} \frac{u_{rms}}{Su_L}}$	(Taylor, 1922)
(d)	$\frac{Su_T}{Su_L} = 1 + \sqrt{2} \left(\frac{u_{rms}}{Su_L}\right)^{1/2}$	(Taylor, 1922)
(e)	$\frac{Su_T}{Su_L} = \sqrt{1 + \left(\frac{2 u_{rms}}{Su_L}\right)^2}$	(Leason, 1951)
(f)	$\frac{Su_T}{Su_L} = 1 + \left(\frac{u_{rms}}{Su_L}\right)^2$	(Clavin and Williams, 1979)
(g)	$Su_T = 15.1 Su_L^{0.784} u_{rms}^{0.412} l_i^{0.196}$	(Popat et al., 1996)

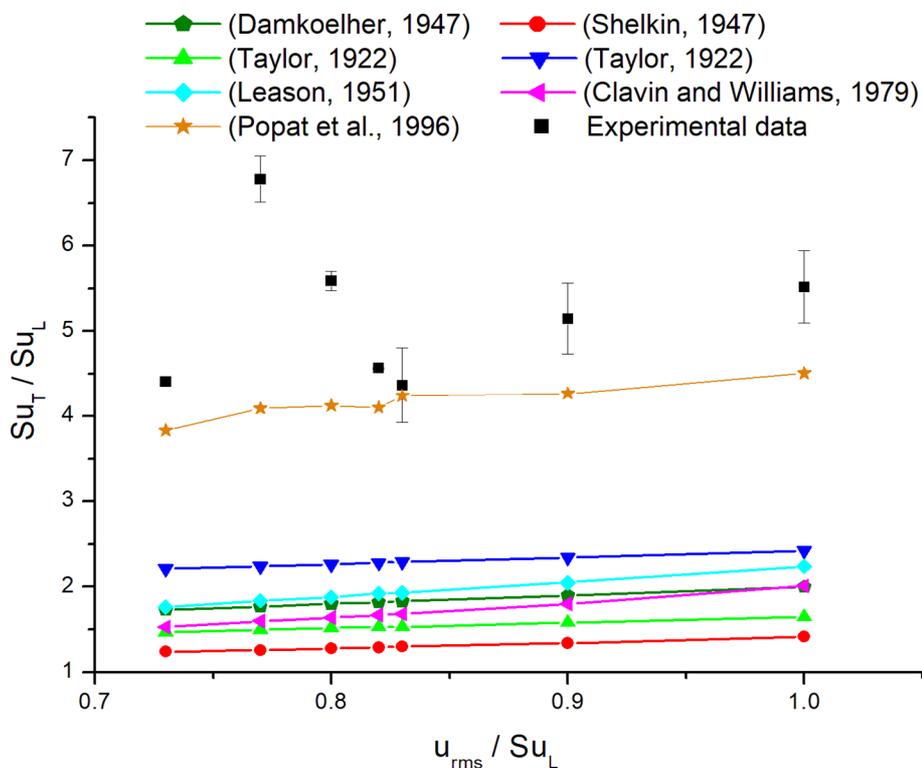
658

659 Concerning the models mentioned by Dahoe et al. (2013), it appears in Figure 17 that an
660 average difference of 55% is observed between the model providing the best fitting, i.e. model
661 (d) from Taylor (1922), and experimental data, meaning none of the models actually fits the
662 experimental data. This statement is not only related to the fact that correlations established
663 for premixed flame were applied to a diffusional flame, since it was previously underlined for
664 premixed flames of methane-air and hydrogen-air mixtures by Dahoe et al. (2013), who then
665 proposed a semi-empiric correlation of the following form :

$$666 \frac{Su_T}{Su_L} = 1 + a_1 D_a^{a_2} \left(\frac{u_{rms}}{Su_L}\right)^{a_3} \quad (8)$$

667 a_1 , a_2 and a_3 being three coefficients to determine experimentally and Da the Damköhler
 668 number. They observed that two distinct sets of coefficients exist in their operating
 669 conditions, revealing that the establishment of a unique correlation allowing the calculation of
 670 the turbulent burning velocity from the knowledge of the laminar burning velocity and the
 671 turbulence intensity is not trivial, even for gases.

672 The model established by Popat et al. (1996) and considered in the CFD code used by Skjold
 673 (2007) clearly provides the best fitting with the experimental data from this work, with an
 674 average difference of 20%. The difference can be due to experimental uncertainties, notably
 675 concerning the point at $Su_T/Su_L = 6.8$, but also to the different assumptions concerning the
 676 correlation, such as a constant kinematic viscosity and the integral length scale l_i equal to 1
 677 cm (Skjold, 2003). A proper estimation of the turbulent velocity being essential to provide an
 678 accurate risk assessment concerning the consequences of an explosion, further investigations
 679 on such models is required.



680

681 *Figure 17. Comparison between models listed in Table 3 and experimental turbulent burning*
682 *velocities measured in the flame propagation tube*

683

684 **4. Conclusion**

685 The laminar burning velocity of nanocellulose has been assessed by three different methods
686 including the direct visualization of the flame propagation in a vertical 1 m long tube or in a
687 vented 20L sphere coupled with the application of relationships relating the flame speed and
688 its stretching, and the use of correlations based on the pressure time evolution during a
689 standard explosion test. Both visualization methods, tube or open sphere, lead to similar
690 results, consistent with the values previously presented in the literature for other organic
691 powders, validating this experimental approach for organic powders. Moreover, these results
692 were compared with those obtained from standard tests in the 20L sphere using a semi-
693 empiric correlation relating the pressure evolution and the flame velocity. All three methods
694 lead to similar values of nanocellulose-air laminar burning velocity, i.e. approximately 21
695 $\text{cm}\cdot\text{s}^{-1}$.

696 The use of nanopowders for such analyses allows reaching very low turbulence levels and
697 thus reduces the impact of turbulence on the flame kernel growth, approaching ‘pseudo-
698 laminar’ conditions. It has notably been underlined that using such flame propagation
699 analyses at low turbulence can support the development of semi-empiric correlations allowing
700 the estimation of the laminar burning velocity from the knowledge of the pressure-time
701 evolution in the standard 20L sphere. The laminar flame velocity being an intrinsic property
702 of the dust-air mixture, it is likely that such techniques could be used in addition to
703 standardized tests in the 20L sphere in order to increase the scope of their results. Indeed,
704 results obtained from standard tests in standards equipment can only be applied to larger
705 enclosures if reliable scaling laws are available. Otherwise, these results are only valid for the

706 specific operating conditions defined by the standards. But, as previously stated in
707 introduction, the validity of the widespread ‘cube-root-law’ depends on several limiting
708 assumptions. Therefore, the determination of a laminar burning velocity can help to
709 overcome such limitations by providing an accurate risk assessment independent from the
710 operating conditions.

711 However, the knowledge of the laminar burning velocity of a mixture is valuable for
712 industrial purposes only if reliable models exist, converting it to a spatial/turbulent burning
713 velocity knowing the turbulent conditions of the cloud. This point is far from being trivial for
714 dusts and some efforts still have to be made in this direction.

715

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724

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