



HAL
open science

Modelling shear viscosity of soft plant cell suspensions

Cassandre Leverrier, Giana Almeida, Gérard Cuvelier, Paul Menut

► **To cite this version:**

Cassandre Leverrier, Giana Almeida, Gérard Cuvelier, Paul Menut. Modelling shear viscosity of soft plant cell suspensions. *Food Hydrocolloids*, 2021, 118, pp.106776. 10.1016/j.foodhyd.2021.106776 . hal-03255092

HAL Id: hal-03255092

<https://agroparistech.hal.science/hal-03255092v1>

Submitted on 24 Apr 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

1 Modelling shear viscosity of soft plant cell 2 suspensions

3
4 Cassandre Leverrier, Giana Almeida, Gérard Cuvelier, Paul Menut

5 Université Paris-Saclay, INRAE, AgroParisTech, UMR SayFood, 91300, Massy, France.
6

7 Abstract

8 Fruit purees are concentrated suspensions of highly deformable, non-spherical particles.
9 These particles are delimited by a plant cell wall and filled with the continuous phase. They
10 can shrink under stress, as it occurs with the increasing concentration in particles, making
11 difficult the determination of their real volume fraction and the understanding of their
12 rheological properties. Usually, the rheological behavior of plant suspensions is described by
13 three concentration domains. We show here on apple puree that beside the complexity of
14 such system, the concentration dependence of the viscosity can be described by one single
15 model over the full range of concentration investigated. The model, originally developed for
16 soft colloids, fits accurately the experimental results obtained on ground purees of several
17 particle size distributions (either monomodal or bimodal) and in several continuous phases.
18 It considers the particles stiffness and asphericity, but also highlight the effects of the
19 continuous phase properties, **for example** through lubrication.
20

21 Nomenclature

C	Polymer concentration	η_{rel}	Relative viscosity
ϕ_{app}	Apparent volume fraction	η	Apparent viscosity
k_0	Specific volume of polymer in diluted solution	η_0	Continuous phase's viscosity
V	Voluminosity	a	First parameter of Batchelor's equation

C_i	Insoluble solids concentration	b	Second parameter of Batchelor's equation
ϕ	Volume fraction (general term)	$[\eta]$	Intrinsic viscosity
λ	Huggins parameter	C^*	Overlapp concentration
k	Mendoza's equation parameter	C_i^*	Critical concentration in apple particle suspensions
ϕ_c	Critical volume fraction	ϕ_{rcp}	Random close packing volume fraction
α	Mendoza's equation first parameter	β	Mendoza's equation second parameter
r	Radius of the core of star polymer particle	R	Radius of the entire star polymer particle
S	Current model fitting parameter	Λ	Spreading of the sigmoid
ϕ_i	Volume fraction reached at the inflection point of the sigmoid	σ	Distribution width
ϕ_{max}	Maximum attainable volume fraction		

22

23 Introduction

24

25 Food products include many examples of soft materials: emulsions (mayonnaise, milk...),
 26 foams (chocolate mousse, Chantilly, whipped egg white...), suspensions (spreadable paste,
 27 processed fruits and vegetables...) and gels (yoghurt, jelly ...). Most of them are complex
 28 multicomponent systems, which explains the rapid expansion of the application of physics
 29 and materials science in food science and technology¹⁻⁴

30

31 Fruit and vegetable purees consist in a suspension of plant cells and are an example of soft
 32 material encountered in food. They are obtained by cooking and grinding the flesh of a fruit
 33 or a vegetable. Flesh is composed of cells, whose size varies according to the plant, from
 34 tens to several hundreds of microns. A plant cell wall, mainly composed of insoluble
 35 polymers, delimits the cells. During cooking, soluble polymers (pectins) contained in the

36 middle lamella between the cell walls will solubilize and the fruit flesh disintegrates,
37 releasing cell clusters of heterogeneous size, individual cells and cell fragments. Cooking
38 parameters will have an impact on the particle size distribution and on the rigidity of the
39 walls, an intensive cooking leading to more porous and less rigid cell walls, and less cell
40 clusters in suspension. Fruit or vegetable suspensions are thus composed of a dispersed
41 phase, the plant particles (plant cells without turgor), suspended in an aqueous continuous
42 phase mainly composed of water, polymers (pectins), sugar and ions initially present in the
43 fruit or vegetable.

44

45 Plant particles obtained thereby have specific structural and physical properties. They are
46 only made of plant cell walls filled with the continuous phase, thus defining a particle whose
47 volume varies with the concentration of the suspension^{5,6}. These particles are porous,
48 deformable and compressible and can be nested in each other. Plant particles are large size
49 non-colloidal objects, which shape and rigidity depend on plant's variety and maturity, but
50 also on mechanical or thermal treatments undergone by the fruits. Shape and rigidity also
51 depend on particle's internal structure: clusters of cells are larger but also more irregular in
52 shape and more rigid than individual cells, due to their internal architecture⁶.

53

54 Suspensions of plant particles strongly resemble soft glassy materials⁷, which in rheological
55 terms are defined as "very soft solids, yet flow readily above a critical yield stress"⁸. The
56 category of soft glassy material comprises foams, emulsions and star polymer gels among
57 others. Specifically, the suspensions of plant particles could be part of the soft particle
58 glasses, defined by Seth (2011) as "materials made of deformable particles as diverse as
59 microgels, emulsion droplets, star polymers, micelles and proteins which are jammed at
60 volume fraction where they are in contact and interact via soft elastic repulsions", behaving
61 like "weak elastic solids at rest but flow[ing] very much like liquids above the yield stress".

62

63 Understanding the rheological behavior of such suspensions and mastering parameters
64 impacting this behavior is a challenge for the food industry. If the rheology of plant particle
65 suspensions has been extensively studied in order to understand the impact of many
66 parameters such as variety, thermal and mechanical treatments or the continuous phase
67 ^{10,11} no rheological model applicable over a wide range of concentrations has been

68 developed yet to describe the rheological behavior of such suspensions, in flow or at rest. In
69 general, models for the viscosity of granular or colloidal suspensions require the knowledge
70 of the particle volume fraction, which is here difficult to estimate. Literature does not agree
71 on one definition so far and many parameters are still used to evaluate the medium's
72 congestion such as the insoluble solids content⁶, the water insoluble solids content¹² or the
73 percentage of pulp¹³. Nevertheless, three concentration domains have been highlighted on
74 several kind of plant particle suspensions^{5,6,14} and discussed in literature. Until now, their
75 modelling have been limited to empirical equations, such as linear, exponential or power law
76 fittings, which range of validity is limited to their own concentration domain.

77

78 In this article, we will propose a single model describing the viscosity as a function of the
79 concentration over the full range of concentration investigated, taking into account the
80 compression when the concentration increases. This model will be conceptually based on a
81 model initially proposed by Mendoza^{15,16} for soft particles, and initially applied to star
82 polymers. We propose here to adapt this model so that it can describe the viscosity
83 divergence that occurs at a finite, maximum volume fraction above which particles cannot
84 be compressed anymore. The validity of the model will be tested on real apple purees
85 characterized by different particles properties, so that it will be possible to correlate the
86 model parameters with particles properties such as their rigidity, shape and size. We will
87 investigate how the effect of polydispersity, associate with different values of the random
88 close packing volume fraction, affect the quality of the model. Finally, we will use a single
89 suspensions of monodispersed apple particles suspended in several continuous phases to
90 evaluate how the model parameters could also reflect variations in continuous phase
91 lubrication properties.

92

93 Material and methods

94

95 Plant material and processing conditions

96 A single batch of mature Golden Delicious (*Malus domestica* Borkh. cv Golden Delicious) was
97 transformed into puree by a French manufacturer (Conserves France) following an industrial
98 hot break process. From this raw material, two kinds of suspensions were studied: ground

99 purees and model suspensions made of isolated apple cells dispersed in various aqueous
100 media.

101

102 [Ground purees](#)

103 Three particle size distributions were obtained using mechanical treatment as described in
104 Leverrier et al. (2016). By using such procedure, and by contrast with thermal treatments cell
105 walls' rigidity is not affected by grinding. The architecture of the particle (either individual
106 cells or clustered) may however have an impact on its global rigidity: clusters are supposed
107 to be more rigid when compared to individual cells⁶.

108

109 [Model suspensions](#)

110 Model suspensions composed of individual cells were also constructed from the same raw
111 material using wet sieving and solvent exchange as described in Leverrier (2017). This
112 protocol is known to maintain an intact cell structure¹⁸. Apple particles (isolated dry
113 individual cells) were suspended in three controlled aqueous media (one solution of NaCl at
114 1%w/w and two solutions of carboxymethylcellulose of 1%w/w and 3.2%w/w, of 1mPa.s,
115 12.6mPa.s and 108mPa.s respectively) and in the original continuous phase of apple puree
116 obtained by centrifugation (apple serum of 12.8 mPa.s).

117

118 [Particle size distributions and rheological measurements](#)

119 The particle size distribution of samples was obtained using a laser diffraction analyser
120 (Mastersizer 2000, Malvern Instruments). Samples were dispersed in distilled water
121 (refractive index: 1.33). The refractive index of vegetable cells is estimated to be 1.52 and
122 the absorption was set at 0.1^{6,10,17,19,20}. Measurements were made in triplicate. Microscopic
123 observations were made as described in Leverrier et al. (2016).

124

125 The rheological measurements were performed using a stress controlled rheometer
126 (MCR301, Anton Paar) equipped with large gap coaxial cylinders^{6,17}. Apparent viscosities of
127 the suspensions were taken at 50s⁻¹.

128

129 Results & Discussion

130

131 How to access the particles' volume fraction?

132 Unlike hard spheres, the volume fraction of soft deformable particles is not well defined^{21,22}.

133 For microgel suspensions, the apparent volume fraction is often linearly related to the

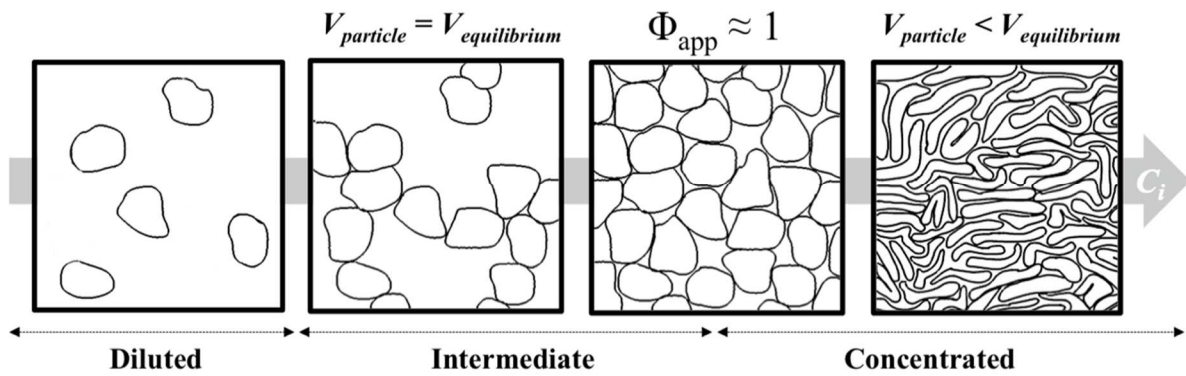
134 polymer concentration C ²³ and based on the particles' volume in diluted conditions²¹, for

135 which polymer networks are in their most swollen state. The apparent volume fraction of

136 microgels suspensions is thus frequently defined as $\phi = k_0 C$, where k_0 is their specific

137 volume in diluted conditions²¹, determined from an Einstein-type equation.

138



139

140 Figure 1. Schematic representation of the impact of the concentration on the particles shape and size adapted from Leverrier
141 et al. (2017).

142 A comparable approach has been used in 2012 by Espinosa (2012) on apple cells suspended
143 in their own serum. The volume fraction of individual apple cells can be then written as

144

$$\phi_{app} = V C_i \quad (1)$$

145

146 Where V is the *voluminosity* of the cells (volume occupied by unit of mass) and C_i is the

147 insoluble solids content. The parameter V being linked to the particle size and shape. By

148 integrating Eq. (1) in a second order expression of the kind of Batchelor & Green (1972) such

149 as $\eta_{rel} = \frac{\eta}{\eta_0} = (1 + a\phi + b\phi^2)$, and by analogy with the Huggins equation (defined for

150 dilute polymer solutions as $\frac{\eta_{rel}^{-1}}{C} = [\eta] + \lambda[\eta]^2$ the following equation was obtained :

151

$$\frac{\eta_{rel} - 1}{C_i} = aV + bC_iV^2 \quad (2)$$

152

153 The parameter a is linked for spherical particles to their deformability, its value is between
154 1 (for gas bubbles)²⁵ and 2.5 (hard spheres). Relying on the work of Nawab & Mason
155 reported in Macosko (1996), $a = 1.7$ was used for individual apple cells²⁰. Their *voluminosity*
156 was then determined from viscosity measurements on diluted suspensions, allowing to
157 define the apparent volume fraction of apple cells suspensions as $\phi_{app} = 1.3C_i$.

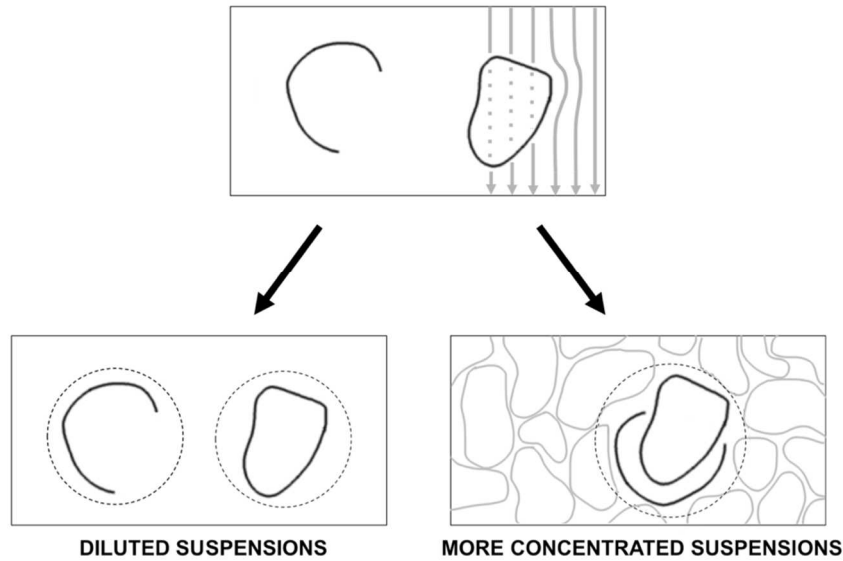
158

159 This approach provides access to ϕ_{app} and takes into account the deformability and the
160 rigidity of the particles by setting the parameter a . However, it slightly overstates the real
161 volume fraction occupied in intermediate and concentrated domains.

162 Indeed, considering a linear relationship between the volume fraction and the quantity of
163 cell wall in suspension (insoluble solids content C_i) implies that the *voluminosity* of the
164 particles is the same whatever the concentration. This is not the case, as illustrated in Figure
165 1. Three concentration domains were highlighted in literature^{5,14}. In the diluted domain,
166 particles are able to flow freely and do not interact with each other. In the intermediate
167 domain, described as the building of a stress-bearing network between particles, particles
168 are at rest in contact with each other: elastic properties become measurable. Note that for
169 spherical particles, the onset of the intermediate domain is usually referred as random loose
170 packing, it occurs at volume fractions which could be much below random close packing, and
171 depend on cohesive and frictional interactions^{27,28}. Finally, the concentrated domain
172 corresponds to a domain where particles highly deform and decrease their volume to fit the
173 available space and to be able to flow.

174 It is therefore necessary to consider the particle compressibility in determining their volume
175 fraction at a given concentration.

176



177
 178 *Figure 2. Schematic representation of two of the plant particles specificities: cell wall porosity allows a (limited) flow through*
 179 *the particle (up), and presence of cell fragment allow particles interlocking in concentrated suspensions. Both phenomena*
 180 *may result in lower hydrodynamic volumes than expected.*

181
 182 The presence of cells fragments also restricts the use of a linear relationship between
 183 volume fraction and insoluble solid content. If plant particle suspensions are composed of
 184 cell walls having maintained their cell integrity, cell fragments are also present in the
 185 suspension. In diluted domain, cell fragments occupy a hydrodynamic volume comparable to
 186 the one of an entire cell (Figure 2). In contrast, in intermediate or concentrated domains,
 187 their hydrodynamic volumes may be significantly lower than the ones occupied by entire
 188 cells (Figure 2). Thus, if the insoluble solids content is the same, the hydrodynamic volume
 189 occupied by cell fragments in diluted or in more concentrated domains can be strongly
 190 different.

191
 192 The apparent volume fraction of soft particles can also be evaluated by defining the volume
 193 fraction as the ratio of the concentration to a critical concentration. This approach is widely
 194 used in polymers' rheology where the apparent volume fraction occupied by the polymers is
 195 often defined as $\phi_{app} = C/C^*$, where C^* is the overlap concentration of polymers^{15,16}. The
 196 overlap concentration is obtained using the polymer molecular weight and the radius of the
 197 sphere equivalent, considering the particles dimensions as if they were hard spheres^{15,16}. It
 198 is worth noting that this approach allows volume fractions greater than 1, due to the

199 possible interpenetration of particles¹⁵ and that it allows to distinguish the concentration
200 domains.

201

202 A similar approach was applied on plant particles by Day et al. (2010) on carrot cells' and
203 broccoli cells' suspensions, and by Leverrier et al. (2016) on apple cells' suspensions. For
204 plant particles suspensions, the critical concentration is determined as a regime change in
205 dynamic¹⁰ or flow^{6,17} properties of the suspensions when the particle concentration is
206 increased. It corresponds to the concentration at which the particles fill all the available
207 space and from which the particles begin to squeeze⁶, represented in Figure 1 as $\phi_{app} \approx 1$.
208 Both on carrot, broccoli and apple cell suspensions^{6,10,17} this approach highlights
209 concentration domains and materializes the transition between the intermediate and
210 concentrated domains, thus defining a relevant parameter to evaluate the volume fraction
211 of plant particle suspensions.

212

213 Following this approach, we define the apparent volume fraction of plant particles as:

214

$$\phi_{app} = \frac{C_i}{C_i^*} \quad (3)$$

215

216 where C_i is the insoluble solids content of the suspension and C_i^* is the critical insoluble
217 solids content at the intersection between the intermediate and concentrated domains¹⁷,
218 obtained from rheological measurements.

219

220 [Presentation of the model](#)

221 Here, the proposed modelling of plant particle suspension viscosity is based on a previous
222 work from Mendoza & Santamaría-Holek (2009) which showed that the viscosity of colloidal
223 particles suspensions can be modelled by :

224

$$\eta(\phi) = \eta_0 \times \left(1 - \frac{\phi}{1 - k\phi}\right)^{-[\eta]} \quad (4)$$

225

226

227 where η_0 is the continuous phase viscosity, $[\eta]$ is an exponent without unit which can be
228 assimilated to the intrinsic viscosity of the suspension (specific viscosity of the suspension
229 for $\phi \rightarrow 0$), and k is defined as follow:

$$k = \frac{1 - \phi_c}{\phi_c} \quad (5)$$

231
232 Where ϕ_c is the critical volume fraction defined as the maximum volume fraction that can
233 be reached by the system.

234
235 This expression takes into account the excluded volume and hydrodynamic interactions, and
236 has been shown to fit with high accuracy suspensions of hard spheres²⁹, droplets³⁰,
237 arbitrarily-shaped hard particles³¹ or rigid core-shell permeable particles³². However, Eq. (4)
238 did not apply to very soft particle suspensions¹⁶, able to decrease their volume or to
239 interpenetrate. *Considering suspensions of star polymers and other soft particles*,
240 Mendoza¹⁶ suggested that a critical volume fraction greater than ϕ_{rcp} (volume fraction at
241 random close packing) may be reached since the particles can interpenetrate, and he
242 proposes that the critical volume fraction follows :

$$\phi_c = \phi_{rcp} + \beta \phi^\alpha \quad (6)$$

244
245 where α and β are two adjustable parameters without unit.

246
247 This gradual increase of the critical volume fraction with the current volume fraction of the
248 suspension, *that is associated with increasing levels of particles compression*, is undoubtedly
249 the major innovation of this model.

250
251 For spherical and permeable particles, Mendoza¹⁶ defined the intrinsic viscosity $[\eta]$ as :

$$[\eta] = \frac{5}{2} \left(\frac{r}{R} \right)^3 \quad (7)$$

253

254 where r and R are the respective radius of the (impermeable) core and the entire particles,
255 considering a shell-permeable periphery of thickness $R - r$.

256

257 This model, that was conceived for soft particles and applied to star polymers suspensions, is
258 intended primarily to describe the shear viscosity of colloidal-size particles suspensions. Such
259 models can however often be applied to non-colloidal (granular-scale) systems, as
260 commonly done for the Krieger's equation³³ for example^{16,34,35}.

261

262 However for plant particles, Eq. (7) cannot be used as such since particles are not spherical
263 in shape. Moreover, the solvent permeability of the plant particles concerns the entire cell
264 wall, and not only a surrounding area. Instead of $[\eta]$, we chose here to use a more general
265 parameter S , which value may vary with the particle Sphericity (or Shape), Softness, Surface
266 irregularities and Size distribution of the suspension. Using in addition the apparent volume
267 fraction of the particles as given by Eq. (3), the initial model from Mendoza therefore
268 becomes:

269

$$\eta(\phi_{app}) = \eta_0 \times \left(1 - \frac{\phi_{app}}{1 - \left[\frac{1 - \phi_{rcp} - \beta \phi_{app}^\alpha}{\phi_{rcp} + \beta \phi_{app}^\alpha} \right] \phi_{app}} \right)^{-S} \quad (8)$$

270

271 In the following sections, we will first demonstrate that this model can be successfully
272 applied to plant suspensions. To facilitate interpretations of the model parameters, we will
273 first set the value of ϕ_{rcp} at 0.637, corresponding to the random close packing fraction of
274 monodisperse hard spheres¹⁶. We are aware that this is not fully appropriate since our
275 system is heterogeneous in size and not spherical in shape but we used it as a first approach.
276 The impact of the polydispersity of apple cell suspensions on ϕ_{rcp} value will also be
277 discussed. Then, we will study the impact of the continuous phase on the parameters of the
278 model. And finally, we will propose some adjustments to the current model.

279

280 Validation of the model

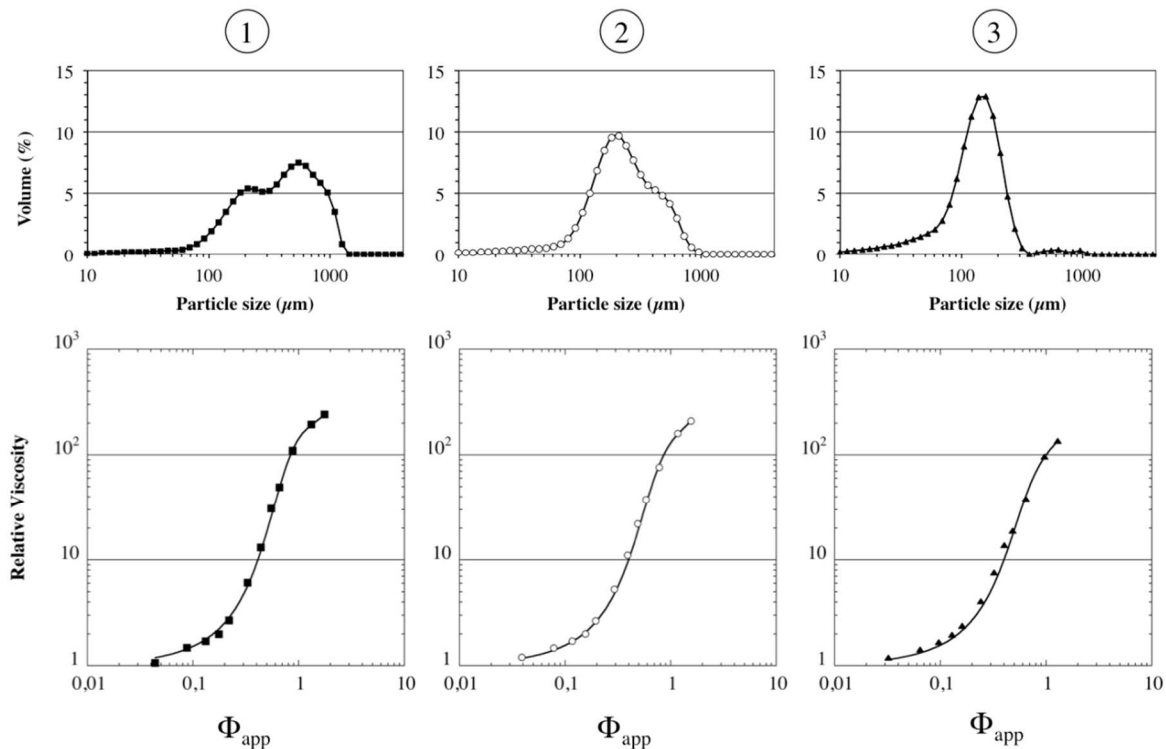
281 First, the effect of the particles properties on the model parameters will be evaluated. To do
282 so, we will compare three different suspensions, characterized by different size distributions
283 but also different particles properties (in terms of rigidity and shape). Second, we will
284 evaluate how the volume fraction at random closed packing, ϕ_{rcp} , which depends on the
285 sample polydispersity, affects the quality of the fit. Third, we will evaluate the impact of the
286 continuous phase, which does not impact particles intrinsic properties but can affect
287 frictional interactions through lubrication.

288

289 Impact of the particle size distribution

290

291



292

293 *Figure 3. Modelling using Eq.(8) for three apple purees varying in particle size distributions. Data were taken from Leverrier*
294 *(2016)⁶. Particle size distributions are represented on the upper side. Relative viscosity is represented as a function of the*
295 *apparent volume fraction on the bottom side. Black squares, circles and black triangles stand for experimental data and the*
296 *solid lines represent the modelling.*

297 It is noteworthy that the model presented in Eq. (8) describe remarkably well the data
298 (Figure 3) for the three purees investigated. Two purees present a bimodal particle size
299 distributions: they contain in different proportions both very large particles up to 1000 μm

300 (clustered cells^{6,11}) and smaller particles of about 150 μm (individual cells^{6,11}), and one puree
301 presents a monomodal size distribution, centered on 150 μm .

302

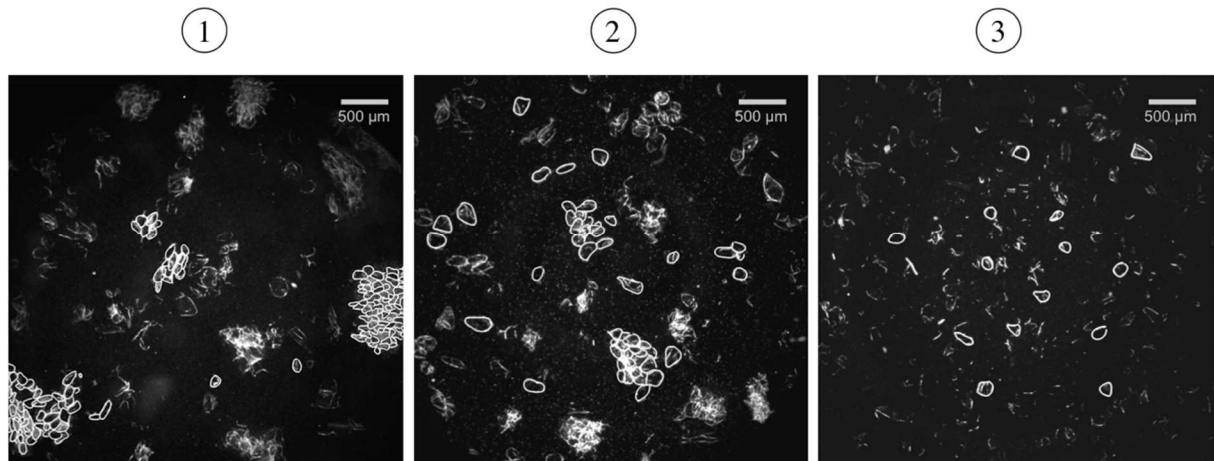
303 *Table 1. Fitting parameters of the model used for the Figure 3 with $\phi_{rcp} = 0.637$.*

	β	α	S	ϕ_{rcp}	R^2
(1)	0.913 (0.017)	2.100 (0.014)	3.697 (0.039)	0.637	0.999
(2)	1.035 (0.039)	2.131 (0.024)	3.893 (0.072)	0.637	0.999
(3)	1.177(0.074)	2.160 (0.031)	3.929 (0.109)	0.637	0.999

304

305 Fitting parameters used in Figure 3 are itemised in Table 1. They all increase when the
306 average particle size decreases. However, the decrease in global particle size also reflects
307 the presence of particles of different kind in the medium, as illustrated in Figure 4. Large
308 particles are clusters of cells, irregular in shape and quite rigid thanks to their complex
309 architecture. Small particles are individual cells, more regular and spherical in shape, and
310 probably less rigid due to their purely liquid content. Grinding thus induces a decrease in
311 particle size, but also an increase of particle sphericity and a decrease of their rigidity and
312 their surface irregularities, which are difficult to decorrelate.

313



314

315 *Figure 4. Microscopic observations of the three particle size distributions. Some of the particles have been highlighted to*
316 *facilitate reading.*

317

318 The fitting parameters α , β , and S depend on the suspension properties, and are therefore
319 link to the particles physical properties. In the following, we investigate how their value
320 depend on the particles rigidity and shape.

321 β values here obtained are 0.913, 1.035 and 1.177 respectively for the purees (1), (2) and (3).
322 In the work of Mendoza (2013)¹⁶, β is related to the particle deformability: the softer is the
323 particle, the greater is β : β values between 0.244 and 0.966 were observed for 128 arms and
324 32 arms star polymer particles, respectively. This is consistent with our results, in which we
325 observe that the higher the proportion of large clusters, the lower the value of β . Indeed,
326 the complex architecture of clusters could generate a greater overall rigidity of the particle⁶,
327 thus here also β is higher when the particles are softer (smaller particles in our case). It is
328 noteworthy that the β values of individual cells are much higher than the softer star polymer
329 ones (with only 32 arms), while stiffer cells clusters exhibit a closer β value confirming that
330 plant particles are highly deformable objects.

331
332 Regarding α , the values obtained on the apple purees (2.10-2.16) are slightly higher than
333 those obtained on star polymer suspensions¹⁶ (1.9-1.74 for 128 and 32 arms particles,
334 respectively). In the case of star polymer suspensions, α decreases when particle
335 deformability increases, while we observe here the opposite effect, the greatest value of α
336 being obtained for the puree (3).

337 We hypothesize that this may be related to the sphericity of the particles or to their surface
338 irregularities. Indeed, on apple purees, when the particle size decreases, the deformability
339 increases, but surface irregularities are decreased and particle sphericity increases.
340 However, in star polymer suspensions, the opposite effect could be anticipated: Indeed, star
341 polymer particles with many arms are very close to hard spheres. If the number of arms
342 decreases, particle deformability increases but particle sphericity could only be reduced,
343 thus possibly increasing surface irregularities.

344
345 Regarding S , we observe that it increases when the particle size decreases and the
346 deformability increases. This is similar to the r/R parameter evolution in star polymer
347 suspensions¹⁶ which was previously associated with particle permeability.

348
349 It should be noted that S is consistently above 2.5, while in existing models for spherical and
350 deformable particles, the power parameter is often presented as equal to¹⁶ or less than
351 2.5^{25,36,37}. Higher values of the Einstein coefficient were however reported for non-spherical

352 particles³⁸. This high value of S determined with plant particles could therefore be due to
353 their non-spherical shape.

354

355 It is noteworthy that Eq. (8) offers, to our knowledge for the first time, the possibility to
356 describe the viscosity dependence towards concentration of complex, non-colloidal
357 suspensions of soft particles. The good agreement of the model with experimental **data** is
358 due to the use of a critical volume fraction that evolve with the actual concentration of
359 particles (Eq. (6)). In other words, due to their compressibility, the *effective* volume fraction
360 of the particles do not increase as the insoluble solids content does.

361

362 Impact of the random close packing volume fraction

363 Up to here, the maximum packing volume fraction was taken as 0.637 for all fittings, the
364 random close packing value for monomodal hard spheres. The impact of polydispersity on
365 this type of highly deformable non-spherical particle suspensions was evaluated. To do so, a
366 similar approach to the one used by Shewan *et al.* (2015)²¹ on agar microgel suspensions
367 was used. Based on the particle size distribution of our suspensions, we determined the
368 distribution's width $\sigma = \ln \frac{d_{4,3}}{d_{3,2}}$. Using the results obtained by Farr *et al.* (2009)³⁹, we
369 determined the random close packing volume fraction of each suspensions, based on their
370 polydispersity. We obtained ϕ_{rcp} values of 0.715, 0.705 and 0.70 for the purees (1), (2), (3)
371 respectively.

372 **Then**, we determined the new model parameters in each case, **they are itemized** in Tableau
373 2. As one can see, no significant improvement is noted on the fit quality, and the impact of
374 the particle size distribution on the parameters α , β and S remains the same as discussed in
375 previous section. The impact of polydispersity on the maximum volume fraction seems to be
376 negligible when modelling the **data**. These results are consistent with observations made in
377 our previous work¹⁷: Particles having non-spherical shapes and high surface irregularities
378 dominate the global rheological properties of suspensions, and polydispersity effect is
379 almost negligible. **For the sake of simplicity**, the value of ϕ_{rcp} corresponding to
380 monodisperse spheres $\phi_{rcp} = 0.637$ will **thus be used for the rest of this work**.

381

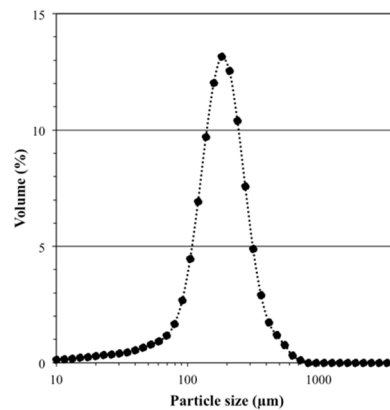
382 *Tableau 2. Fitting parameters of the model used for the Figure 3 considering the particle size distribution effect on $\phi_{r_{cp}}$.*

	β	α	S	$\phi_{r_{cp}}$	R^2
(1)	0.914 (0.021)	2.285 (0.018)	3.880 (0.042)	0.715	0.999
(2)	1.053 (0.044)	2.292 (0.028)	4.057 (0.072)	0.705	0.999
(3)	1.223(0.081)	2.309 (0.035)	4.090 (0.105)	0.70	0.999

383

384 Impact of the viscosity of the continuous phase

385 In this section, model suspensions of individual apple cells varying in continuous phase
 386 viscosity and composition will be considered (data taken from Leverrier *et al.* (2017)¹⁷). Such
 387 suspensions are composed of the same batch of particles, characterized by a monomodal
 388 distribution, centered on 180 μm (Figure 5). It allows us to assume that all particles have the
 389 same deformability, sphericity, surface irregularities and polydispersity, and to investigate
 390 here more specifically how a phenomenon like lubrication, which depends on the
 391 continuous phase properties, could affect the suspension viscosity other the full range of
 392 volume fraction investigate. Figure 6 represents the resulting fittings of Eq. (8) on these
 393 suspensions varying only in continuous phase. It is noteworthy that Eq. (8) fits again quite
 394 well the data, whatever the continuous phase composition.



395

396 *Figure 5. Particle size distribution of monomodal model suspensions.*

397 Fitting parameters are itemised in Table 3. We observe that when the continuous phase
 398 viscosity increases, β and α increase and S decreases.

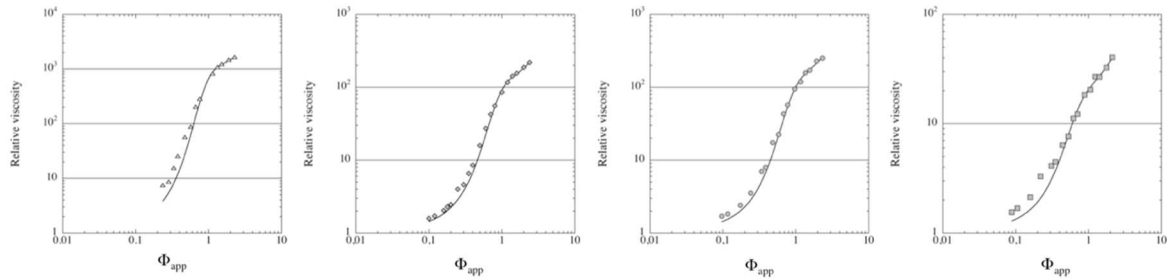
399

400 *Table 3. Fitting parameters of the model used for the Figure 6.*

	η_0 (mPa.s)	β	α	S	$\phi_{r_{cp}}$	R^2
NaCl 1%	1	0.774 (0.023)	1.989 (0.041)	4.362 (0.114)	0.637	0.997
CMC 1%	12.6	0.869 (0.036)	1.937 (0.045)	3.341 (0.095)	0.637	0.996

<i>Apple serum</i>	12.8	0.851 (0.016)	1.964 (0.022)	3.226 (0.043)	0.637	0.999
<i>CMC 3.2%</i>	108	1.222 (0.089)	2.143 (0.077)	2.621 (0.078)	0.637	0.994

401



402

403 *Figure 6. Modelling using Eq. (8) for model suspensions in 4 different continuous phases. Symbols represent experimental*
404 *data for model suspensions, in NaCl 1% (triangles), apple serum (diamonds), carboxy-methyl-cellulose solution at 1%*
405 *(circles) and carboxy-methyl-cellulose solution at 3.2% (squares). Solid lines represent the fittings resulting from Eq. (8) for*
406 *each continuous phase.*

407

408 Previously, and in agreement with Mendoza's approach, we have shown that α and β
409 parameters were related to particles' deformability and sphericity (or surface irregularities).
410 At a first look, it might seem surprising that the continuous phase composition impacts the
411 model parameters, however, we remind here the reader that apple cells are granular, non-
412 brownian particles. In such suspensions, frictional forces play a key role in rheological
413 properties⁴⁰⁻⁴². One additional consequence is that these particles are not sensitive to the
414 depletion effects that can be observed in colloidal systems.

415 The increase of α with the increasing continuous phase viscosity could be due to a
416 lubricating effect¹⁷. By increasing the viscosity of the continuous phase or the presence of
417 polymers in the medium, frictions between particles are decreased.

418 This lower friction could have an impact on apparent surface irregularities, leading to an
419 apparent more spherical shape of the particles. The increase of β with the viscosity of the
420 continuous phase could be related to crowd effects. Indeed, in all cases, the viscosity of the
421 continuous phase has been increased by introducing polymers in the continuous phase.
422 These polymers, whose concentration increases with the continuous phase viscosity, have
423 an intrinsic volume and thus occupy space in the medium. If the rheological properties of the
424 suspensions are predominantly driven by the particles^{6,17}, the presence of polymers in the
425 continuous phase can force particles to deform to a greater extent, thus leading to higher β
426 values.

427

428 We also observe that S , previously associated with the particle's permeability, decreases
429 when the continuous phase viscosity increases.

430 Here, higher viscosities are associated with higher polymer content: in such cases, the
431 continuous phase goes through particles less easily, giving rise to lower apparent
432 permeabilities

433

434 Adjustments of the model

435 This work highlights the interest of a model that goes beyond a constant random close
436 packing defined for swollen particles. However, a clear limitation of the model is that Eq. (6)
437 implies that a finite value of the critical volume fraction ϕ_c cannot be determined from a set
438 of data, since it increases monotonically with the particle volume fraction : for different
439 volume fractions ϕ , different values of ϕ_c are determined. Still, at sufficiently high
440 concentrations, cells should become incompressible, and therefore, the volume fraction
441 reaches a finite, maximum value. This maximum volume fraction, ϕ_{max} , is an important
442 characteristic of a given system, as it gives a clear indication of the maximum compression
443 level that can be reached physically, which should depend on the particles structure and
444 composition. To circumvent this limitation, we propose to modify the existing model so that
445 this maximum volume fraction could be included, as indicate in the following equation:

$$\phi_c = \phi_{rcp} + \frac{\phi_{max} - \phi_{rcp}}{1 + \exp[-\Lambda(\phi_{app} - \phi_i)]} \quad (9)$$

446

447 As proposed by Mendoza, the critical volume fraction evolves here with the particle
448 concentration above the random close packing volume fraction defined for hard spheres.
449 However, rather than using a power law model, leading to an infinite critical volume
450 fraction, we use a sigmoidal law, so that a finite maximal volume fraction (ϕ_{max}) will be
451 reached eventually.

452

453 While this equation involves an additional variable, it seems more physically realistic to the
454 vegetable particles of ours. As it includes a sigmoid function, it allows to model the viscosity
455 evolution with volume fraction in a wider range of concentrations, including the divergence
456 that should occur at extremely high volume fractions. The maximum attainable volume
457 fraction ϕ_{max} included in Eq. (9) allows us to translate the incompressibility limit

458 encountered by the particles at high volume fraction, which, in our case, results in a
459 divergence of the viscosity at high volume fraction, assuming that particles are not able to
460 shrink or deform anymore. It is however less accurate for very diluted systems: for
461 extremely low values of ϕ_{app} , the critical volume fraction ϕ_c remains slightly higher than
462 ϕ_{rcp} in our model. Still, food suspensions are rarely highly diluted system and Einstein's law
463 remains the best way to fit very diluted systems.

464

465 In Eq. (9), the model parameters can be associated with different physical properties. Δ
466 defines the spreading of sigmoid (i. e., the slope of the sigmoid at the inflection point) and
467 ϕ_i defines the value of the inflection point which corresponds to the points for which the
468 particle compressibility towards particles concentration is maximum. The sigmoid is also
469 defined by two asymptotes, the first one is equal to zero, and the second one is defined by
470 the parameter ϕ_{max} . This parameter set the critical volume in a finite range between ϕ_{rcp}
471 and ϕ_{max} , and allows the estimation of the latter from experimental data

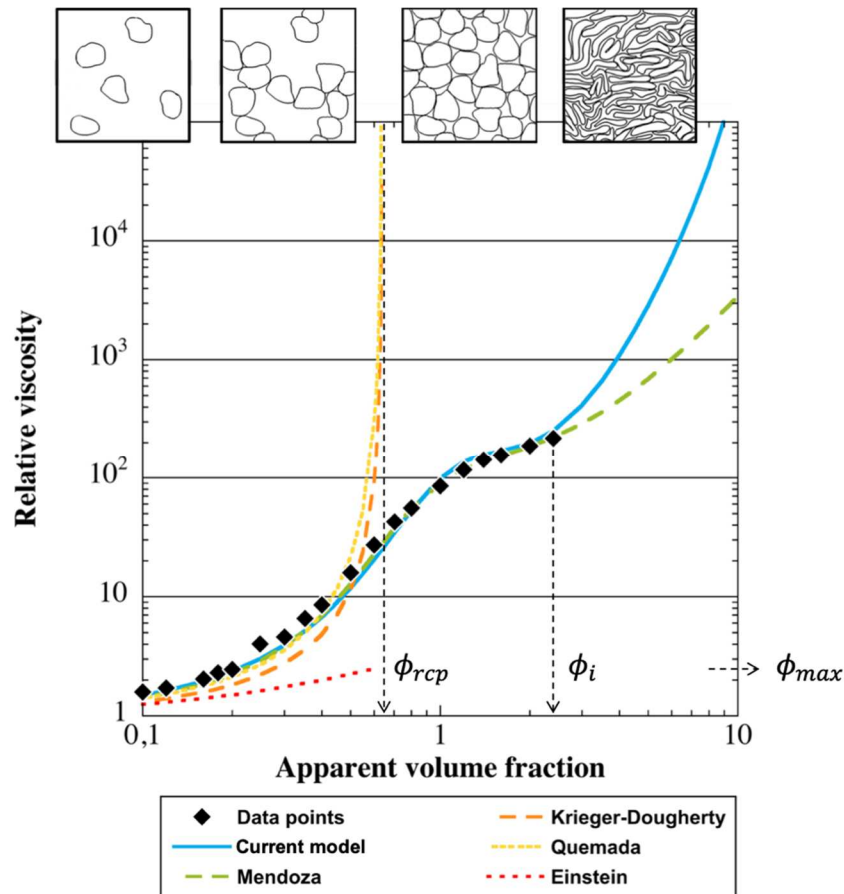
472

473 To summarize, we fitted in Figure 7 our data with the most widely used models in the
474 literature (i.e. the Einstein model, the Quemada model and the Krieger-Dougherty model),
475 with the model proposed by Mendoza and with the model we propose here (combining
476 equations (4), (5) and (9)). The parameters used to fit our data points are summarized in
477 Table 4. The Einstein model, the only theoretical model describing the evolution of viscosity
478 with the apparent volume fraction, does not allow to model concentrated systems. Thus, as
479 expected, it does not fit our data over the entire range of concentrations studied. The
480 Quemada and Krieger-Dougherty models also do not fit our data correctly, because in these
481 models the viscosity increases more and more as the volume fraction increases, till it
482 diverges at volume fractions much lower than the ones reached experimentally. The
483 Mendoza model, as we have seen in this work, allows us to successfully model the entire
484 range of concentration that was experimentally investigated. However, in this model the
485 viscosity will continuously increases with the volume fraction, as an upper limit for ϕ_c will
486 never be reached, whatever the value of ϕ . As it can be observed, the model we propose
487 allows by contrast to fit the whole range of our data (from diluted to highly concentrated)
488 while having a volume fraction upper limit.

489

Table 4. Models used for the Figure 7

Models	Equation and parameters used in Figure 7
Einstein	$\eta(\phi) = \eta_0(1 + k_E\phi)$ $k_E = 2.5$
Quemada	$\eta(\phi) = \eta_0 \left(1 - \frac{\phi}{\phi_{rcp}}\right)^{-q}$ $q = 2.0; \phi_{rcp} = 0.637$
Krieger-Dougherty	$\eta(\phi) = \eta_0 \left(1 - \frac{\phi}{\phi_{rcp}}\right)^{-k_E\phi_{rcp}}$ $k_E = 2.5; \phi_{rcp} = 0.637$
Mendoza	$\eta(\phi) = \eta_0 \left(1 - \frac{\phi}{1 - k\phi}\right)^{-[\eta]}$ $k = \frac{1 - \phi_c}{\phi_c}; \phi_c = \phi_{rcp} + \beta\phi^\alpha$ $\alpha = 1.964; \beta = 0.851; \phi_{rcp} = 0.637; [\eta] = 3.226$
Current model	$\eta(\phi) = \eta_0 \left(1 - \frac{\phi}{1 - k\phi}\right)^{-S}$ $k = \frac{1 - \phi_c}{\phi_c}; \phi_c = \phi_{rcp} + \frac{\phi_{max} - \phi_{rcp}}{1 + \exp[-\Lambda(\phi - \phi_i)]}$ $\Lambda = 1.87; \phi_i = 2.38; \phi_{max} = 16; \phi_{rcp} = 0.637; S = 3.76$



494

495 *Figure 7. Modelling of model suspensions in apple serum with theoretical and phenomenological models describing the*
 496 *evolution of the viscosity with the apparent volume fraction. Model and parameter used are summarized in Table 4*

497

498 Conclusion

499

500 In this work, an indirect way of accessing the effective volume fraction of the particles and a
 501 model describing the concentration dependence of the viscosity of soft plant cells
 502 suspensions over a wide range of concentration were proposed.

503 The model, initially proposed for **soft colloidal particles** and applied to star polymers¹⁶, was
 504 successfully applied to real apple purees of several grindings and to **monodispersed**
 505 suspensions of **apples cells** in several continuous phases. Three adjustment parameters are
 506 defined and related to the particle's **intrinsic physical properties**: deformability, sphericity
 507 and porosity.

508 We also showed that for such non-colloidal suspensions, in which friction forces play an
 509 important role in the rheological properties, those parameters are also related to the
 510 continuous phase properties **that may act as a lubricant and decrease frictional interactions**.

511 To represent the predictable divergence of the viscosity at the maximum packing fraction,
512 we successfully introduced a sigmoid-type term in the model, which allows the prediction of
513 the viscosity in the entire range of volume fraction. This new model necessitates additional
514 parameters, however these parameters now include a key characteristic of a compressible
515 suspension, namely the maximum volume fraction, ϕ_{max} , that is reached when particles are
516 fully compressed. For the system investigated here, we found $\phi_{max}=16$, which demonstrates
517 the extremely high compressibility of apple cells. This estimation calls for future work:
518 investigating apple cells suspensions rheological properties in extremely concentrated states
519 would allow to confirm the validity of the model in this limit. The predicted divergence of
520 the viscosity while approaching ϕ_{max} indeed assumes that in this limit, particles become
521 incompressible but also cannot deform anymore, which seems reasonable considering the
522 high level of entanglement, but have not been confirmed experimentally to date to our
523 knowledge Also, future work should be done on others food soft granular suspension to
524 evaluate how this model could be applied to other systems.

525 This model represents a significant advance in the understanding of processed fruit and
526 vegetables systems and offers clear industrial prospects.

527 We believe the attempt to link macroscopic behavior with local properties such as
528 compressibility or friction in such systems would benefit from in-depth investigation at the
529 particles scale, both experimentally and numerically, that would certainly in the future allow
530 a better description of their behavior.

531

532 Acknowledgements

533 The research leading to these results has received funding from the European Union's
534 Seventh Framework Programme for research, technological development and
535 demonstration under grant agreement number 311754. The authors wish to thank Adeline
536 Boire, for fruitful discussions.

537

538 References

539

540 1. Ubbink, J. Soft matter approaches to structured foods: from “cook-and-look” to

- 541 rational food design ? *Faraday Discuss.* **158**, 9 (2012).
- 542 2. Vilgis, T. A. Soft matter food physics—the physics of food and cooking. *Reports Prog.*
543 *Phys.* **78**, 124602 (2015).
- 544 3. Sman, R. G. M. Van Der. Soft matter approaches to food structuring. *Adv. Colloid*
545 *Interface Sci.* **176–177**, 18–30 (2012).
- 546 4. Boire, A. *et al.* Soft-Matter Approaches for Controlling Food Protein Interactions and
547 Assembly. *Annu. Rev. Food Sci. Technol.* **10**, 521–542 (2019).
- 548 5. Lopez-Sanchez, P., Chapara, V., Schumm, S. & Farr, R. Shear Elastic Deformation and
549 Particle Packing in Plant Cell Dispersions. *Food Biophys.* **7**, 1–14 (2012).
- 550 6. Leverrier, C., Almeida, G., Espinosa-Munoz, L. & Cuvelier, G. Influence of Particle Size
551 and Concentration on Rheological Behaviour of Reconstituted Apple Purees. *Food*
552 *Biophys.* **11**, 235–247 (2016).
- 553 7. Roversi, T. & Piazza, L. Supramolecular assemblies from plant cell polysaccharides:
554 Self-healing and aging behavior. *Food Hydrocoll.* **54**, 189–195 (2016).
- 555 8. Chen, D. T. N., Wen, Q., Janmey, P. A., Crocker, J. C. & Yodh, A. G. Rheology of Soft
556 Materials. *Annu. Rev. Condens. Matter Phys.* **1**, 301–322 (2010).
- 557 9. Seth, J. R., Mohan, L., Locatelli-Champagne, C., Cloitre, M. & Bonnecaze, R. T. A
558 micromechanical model to predict the flow of soft particle glasses. *Nat. Mater.* **10**,
559 838–43 (2011).
- 560 10. Day, L., Xu, M., Øiseth, S. K., Lundin, L. & Hemar, Y. Dynamic rheological properties of
561 plant cell-wall particle dispersions. *Colloids Surf. B. Biointerfaces* **81**, 461–467 (2010).
- 562 11. Espinosa-Muñoz, L., Renard, C. M. G. C., Symoneaux, R., Biau, N. & Cuvelier, G.
563 Structural parameters that determine the rheological properties of apple puree. *J.*
564 *Food Eng.* **119**, 619–626 (2013).
- 565 12. Schijvens, E. P. H. M., Van Vliet, T. & Van Dijk, C. Effect of processing conditions on the
566 composition and rheological properties of applesauce. *J. Texture Stud.* **29**, 123–143
567 (1998).
- 568 13. Moelants, K. R. N. *et al.* Relation Between Particle Properties and Rheological
569 Characteristics of Carrot-derived Suspensions. *Food Bioprocess Technol.* **6**, 1127–1143
570 (2013).
- 571 14. Day, L., Xu, M., Øiseth, S. K., Hemar, Y. & Lundin, L. Control of Morphological and
572 Rheological Properties of Carrot Cell Wall Particle Dispersions through Processing.

- 573 *Food Bioprocess Technol.* **3**, 928–934 (2010).
- 574 15. Koumakis, N., Pamvouxoglou, A., Poulos, A. S. & Petekidis, G. Direct comparison of the
575 rheology of model hard and soft particle glasses. *Soft Matter* **8**, 4271 (2012).
- 576 16. Mendoza, C. I. Model for the Shear Viscosity of Suspensions of Star Polymers and
577 Other Soft Particles. *Macromol. Chem. Phys.* **214**, 599–604 (2013).
- 578 17. Leverrier, C., Almeida, G., Menut, P. & Cuvelier, G. Design of Model Apple Cells
579 Suspensions: Rheological Properties and Impact of the Continuous Phase. *Food*
580 *Biophys.* **12**, 383–396 (2017).
- 581 18. Müller, S. & Kunzek, H. Material properties of processed fruit and vegetables I . Effect
582 of extraction and thermal treatment on apple parenchyma. *Zeitschrift für Leb. und -*
583 *forsch. A* **206**, 264–272 (1998).
- 584 19. Espinosa, L. *et al.* Effect of processing on rheological, structural and sensory
585 properties of apple puree. *Procedia Food Sci.* **1**, 513–520 (2011).
- 586 20. Espinosa, L. Texture de la purée de pomme : influence de la structure sur les
587 propriétés rhéologiques et la perception sensorielle - effet du traitement mécanique.
588 (AgroParisTech - Ingénierie Procédés aliments, 2012).
- 589 21. Shewan, H. M. & Stokes, J. R. Viscosity of soft spherical micro-hydrogel suspensions. *J.*
590 *Colloid Interface Sci.* **442**, 75–81 (2015).
- 591 22. Boehm, M. W., Warren, F. J., Baier, S. K., Gidley, M. J. & Stokes, J. R. Food
592 Hydrocolloids A method for developing structure-rheology relationships in
593 comminuted plant-based food and non-ideal soft particle suspensions. *Food*
594 *Hydrocoll.* **96**, 475–480 (2019).
- 595 23. Fernandez-Nieves, A., Wyss, H. M., Mattsson, J. & Weitz, D. A. *Microgel Suspensions*
596 *Fundamentals and Applications.* (Wiley-VCH, 2011).
597 doi:10.1017/CBO9781107415324.004
- 598 24. Batchelor, G. K. & Green, J. T. The determination of the bulk stress in a suspension of
599 spherical particles to order c^2 . *J. Fluid Mech.* **56**, 401–427 (1972).
- 600 25. Taylor, G. I. The viscosity of a fluid containing small drops of another fluid. *Proc. R.*
601 *Soc. A Math. Phys. Eng. Sci.* **138**, 41–48 (1932).
- 602 26. Macosko, C. *Rheology: Principles, Measurements and Applications. Powder*
603 *Technology* (Wiley-VCH, 1996). doi:10.1016/S0032-5910(96)90008-X
- 604 27. Silbert, L. Jamming of frictional spheres and random loose packing. *Soft Matter* **13**,

- 605 (2010).
- 606 28. Dong, K. J., Yang, R. Y., Zou, R. P. & Yu, A. B. Role of Interparticle Forces in the
607 Formation of Random Loose Packing. *Phys. Rev. Lett.* **96**, (2006).
- 608 29. Mendoza, C. I. & Santamaría-Holek, I. The rheology of hard sphere suspensions at
609 arbitrary volume fractions: An improved differential viscosity model. *J. Chem. Phys.*
610 **130**, (2009).
- 611 30. Mendoza, C. I. & Santamaría-Holek, I. Rheology of concentrated emulsions of
612 spherical droplets. *Appl. Rheol.* **20**, 1–3 (2010).
- 613 31. Santamaría-Holek, I. & Mendoza, C. I. The rheology of concentrated suspensions of
614 arbitrarily-shaped particles. *J. Colloid Interface Sci.* **346**, 118–26 (2010).
- 615 32. Mendoza, C. I. Effective static and high-frequency viscosities of concentrated
616 suspensions of soft particles. *J. Chem. Phys.* **135**, (2011).
- 617 33. Krieger, I. M. & Dougherty, T. J. A Mechanism for Non-Newtonian Flow in Suspensions
618 of Rigid Spheres. *Trans. Soc. Rheol.* **III**, 137–152 (1959).
- 619 34. Adams, S., Frith, W. J. & Stokes, J. R. Influence of particle modulus on the rheological
620 properties of agar microgel suspensions. *J. Rheol. (N. Y. N. Y.)* **48**, 1195–1213 (2004).
- 621 35. Ellis, A. & Jacquier, J. C. Manufacture and characterisation of agarose microparticles. *J.*
622 *Food Eng.* **90**, 141–145 (2009).
- 623 36. Quemada, D. *Modélisation rhéologique structurelle : dispersions concentrées et fluides*
624 *complexes*. (Lavoisier, 2006).
- 625 37. Quemada, D. Rheology of concentrated disperse systems and minimum energy
626 dissipation principle I . Viscosity-concentration relationship. *Rheol. Acta* **16**, 82–94
627 (1977).
- 628 38. Mueller, S., Llewellyn, E. W. & Mader, H. M. The rheology of suspensions of solid
629 particles. *Proc. R. Soc. A Math. Phys. Eng. Sci.* **466**, 1201–1228 (2010).
- 630 39. Farr, R. S. & Groot, R. D. Close packing density of polydisperse hard spheres. *J. Chem.*
631 *Phys.* **131**, 244104 (2009).
- 632 40. Menut, P., Seiffert, S., Sprakel, J. & Weitz, D. A. Does size matter? Elasticity of
633 compressed suspensions of colloidal- and granular-scale microgels. *Soft Matter* **8**, 156
634 (2012).
- 635 41. Denn, M. M. & Morris, J. F. Rheology of Non-Brownian Suspensions. *Annu. Rev. Chem.*
636 *Biomol. Eng.* **5**, 203–28 (2014).

637 42. Ancey, C., Coussot, P. & Evesque, P. A theoretical framework for granular suspensions
638 in a steady simple shear flow. *J. Rheol. (N. Y. N. Y)*. 1673 (1999).
639 doi:<https://doi.org/10.1122/1.551067>
640