Rheological properties of artificial boluses of cereal foods enriched with legume proteins

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9   **Abstract**

10   The properties of artificial food bolus are studied by dynamic oscillatory and capillary

11   rheometry as functions of bolus water content (WC), in the usual range of saliva hydration, for four

12   cereal products: sponge cake, extruded flat bread and their counterpart enriched in legume proteins.

13   All boluses followed the same rheological behaviour characterised by (1) solid -like in the linear

14   viscoelastic domain and (2) Herschel-Bulkley model for large shear strain. Hence, four characteristic

15   rheological properties are determined: modulus at viscoelastic plateau, characteristic stress at

16   transition to flow, yield stress and consistency in the flow regime. The decrease of these properties

17   with WC was fitted by an exponential decay function, from which was extracted a coefficient α

18   ($5 \leq \alpha \leq 30$), defined as a coefficient of interaction of the food with water. The values of α are of

19   the same order of magnitude as the plasticization coefficient of starch by water. They were larger for

20   the extruded pea based (EFP, $\alpha \geq 15$), and were lower for the sponge cake (SC, $\alpha \leq 15$). The

21   variations for the different rheological properties are discussed in terms of matter state, envisioning

22   bolus as a suspension of soft swellable particles. The comparison of these values with those

23   encountered for real boluses from similar foods suggests that these results contribute to define a

24   coefficient of interaction of food with saliva.

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27   **Keywords:** Herschel-Bulkley model; interaction coefficient; modulus; plant protein; viscosity

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30   **Nomenclature**

31   a shift factor used to derive the flow master curve

32   D, D_P diameters of rheometer capillary die and piston, respectively

33   EF, EFP   extruded flat bread end extruded pea based snack, respectively

34   G', G'', G^* storage, dissipative and complex modulus, respectively

35   G'_0 theoretical value of storage modulus for dry bolus

36   K, K_c consistency and its corrected value in the Herschel-Bulkley model

37   K_{c0} theoretical value of consistency for the dry bolus

38   L/D length to diameter capillary die ratio

39   n flow index in the Herschel-Bulkley model

40   SC, SCP   sponge cake, enriched in pulse protein, respectively

41   T temperature

42   WC   Water content, expressed on a total wet basis

43   α_G. $\alpha_{\tau c}$, α_τ, α_K interaction coefficients of food with water based storage modulus, characteristic

44   stress, yield stress, consistency, respectively

45   γ, γ_c strain and its value at intersection of G' and G'' curves

46    $\dot{\gamma}_{app}$, $\dot{\gamma}_w$ apparent and real wall shear rate

47    $\dot{\varepsilon}$ deformation rate

48    $\eta_{app}, \eta^*, \eta, \eta_E$ apparent, complex, shear and elongational viscosity, respectively

49   ω pulsation (oscillatory rheometry tests)

50   τ_w, τ_c , τ_s , τ_{sc} wall shear, characteristic, yield (Hersche-Bulkley), corrected stress

51   τ_{c0}, τ_{s0} dry bolus theoretical values of characteristic and yield stress, respectively

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1. Introduction

Food Oral Processing (FOP) is a key step for food digestion. Its first aim is to form a food bolus that can be swallowed safely (Chen, 2009 ; Stokes, Boehm & Baier, 2013). It is also the first interaction of our body with food in the digestive system. During oral processing, food is reduced in size and lubricated to form a bolus in preparation for swallowing and digestion. Simultaneously, volatile chemicals from the food move to olfactory and taste receptors, food particles interact with oral surfaces, and the net result is an evaluation of taste, aroma, and texture. The rheological properties of the bolus are a link between food texture, its breakdown and its capacity to be swallowed, and they are modified by the saliva uptake. Consequently, the knowledge of bolus rheological properties is important to understand the dynamic changes in food structure that take place during FOP (Morell, Hernando & Fiszman, 2014).Consequently, there is a need to develop methods for assessing food breakdown during chewing and interaction with saliva (Boehm, Warren, Baier, Gidley & Stokes,2019).

The food bolus experiences a large range of shear rate and also extensional flow during oral processing. From apparent viscosity (η_{app}) measurements, a model was built to represent the bolus breakdown of cereal foods, such as sponge cake and brioche, during chewing (Assad-Bustillos, Tournier, Septier, Della Valle & Feron, 2019a). This model allowed to derive a coefficientrelating bolus viscosity to its water content, which was defined as a coefficient of interaction « α » between saliva and food (Assad-Bustillos et al., 2020). The variations of α underlined the influence of protein enrichment on the oral processing of these cereal foods. It also showed that the study of FOP is an essential step in food design, in the case of enrichment of foods by legume proteins. Moreover, a correlation between η_{app} with oral comfort for low fat cereal foods has been established, which just confirmed how important are the bolus shear and extensional viscosities for the ease of swallowing (Marconati et al., 2019).

Saliva contains water at 99%, plus other components, such as mucin, and its properties have a large variability depending on individual and stimulation conditions (Haward, Odell, Berry & Hall, 2011; Mosca & Chen, 2017). So the increase of bolus water content (WC), due to saliva flow during chewing, is a common important feature in FOP. This is especially true for cereal products which contain a large amount of amorphous starch, knowing the role of water as a plasticizer for hydrophilic food components, such as amorphous starch. So, there is an interest to study the WC dependence of bolus rheological properties of cereal food boluses and, by extent, to any food that contains hydrophilic compounds.

Surprisingly, there are quite few studies dealing with the rheological properties of food bolus. This can be due (1) to the difficulty of performing rheological measurements on real boluses, and (2)

88 to the uncertainty about the relevant rheological property for food oral processing. To overcome the

89 former difficulty, artificial boluses can be considered, provided that they are representative of real

90 ones. By doing so, it is possible to address the latter difficulty by performing different rheological

91 tests under a wide range of strain and strain rate conditions.

92   Given this context, the aim of this work is to determine the rheological properties of artificial

93 boluses of cereal foods in order to derive coefficients that help to quantify the interactions of foods

94 with saliva. For this purpose, artificial boluses are prepared under conditions as close as possible to

95 those encountered in FOP, to avoid physiological interindividual variability and to focus on bolus

96 rheology and the influence of water content. Bolus viscoelastic properties are investigated in the

97 linear and non-linear viscoelastic domains, using oscillatory shear rheometry, and flow properties are

98 measured by capillary rheometry. In addition to insights on food bolus structure, the variations of the

99 rheological properties with WC are determined for two types of cereal foods: a soft food with

100 intermediate moisture and a brittle dry one, as well as their corresponding version enriched in plant

101 protein.

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104 **2. Material and methods**

105   ***2.1. Cereal foods and bolus preparation***

106   Four cereal food products were studied: two soft foods (sponge cake, SC) and two brittle

107 ones (extruded, E). One SC had a standard formulation (SC) and the other one was enriched with pea

108 proteins and held the claim "high in protein" (SCP). Both were provided by CERELAB® (Aiseray,

109 France). One extruded food is a commercial flat bread (EF, Les craquantes EPI d'OR ™) and the other

110 one is made by extrusion of pea flour (EFP), as described in detail by Kristiawan et al. (2018). Their

111 composition, moisture content and density are reported in Table 1. Besides various differences,

112 especially starch content, the difference of water content makes the food soft (SC) or brittle (E).

113   In order to prepare artificial boluses, foods were fragmented and impregnated with water,

114 the main component (99%) of human saliva. This study focuses on the role of water, because the

115 addition of mucin and salts has been precedingly shown not to modify the rheological properties of

116 sponge cake boluses (Gibouin, Della Valle & van der Sman, 2019 a,b). In addition, the role of α-

117 amylase on rheological properties is discarded, in a first approach. The role of real saliva, and

118 especially of α-amylase will be discussed when comparing our results with those obtained for real

119 boluses, in section 3.3. Water was added at different levels, in order to obtain a bolus, homogeneous

120 at bare eye, and to cover the range of water content of food bolus during chewing, in agreement

121 with values found in literature, up to 60% for sponge cake and 80% extruded foods, in total wet

122 content (Assad-Bustillos et aL, 2019a; Loret et al., 2011).

123   Before water addition, sponge cakes and extruded foods were fragmented with a commercial

124   blender(MagiMix, France) or manually using a mortar, respectively, until a particle mean size of 1mm

125   is reached, as measured with a vibrating sieves (Reitsch, F95610 Eragny). This size value is chosen

126   close enough to the particle size in real boluses at the first step of chewing. It has also been checked

127   that the particle size distribution is in agreement with the distribution in real boluses (Assad-Bustillos

128   et al., 2019b), and that in the range [1, 10mm], the particle size did not affect significantly the

129   rheological properties (Gibouin et al., 2019 a,b).

130   ### *2.2. Viscoelastic properties and oscillatory shear measurements*

131   Viscoelastic properties of bolus were determined with a controlled strain rheometer (ARES,

132   TA Instruments), provided with a plate-plate circular geometry: diameter = 4.0cm and gap = 2.5mm.

133   The temperature is controlled with a Peltier device and set to 23°C. The bolus sample is placed on the

134   lower plate and is gently squeezed to ensure surface contact. The variations of storage (G') and loss

135   (G'') moduli, with strain were determined at three different frequencies: ω = 10rad/s, ω = 1rad/s

136   and ω = 0.1rad/s. The imposed strains ranged from 0.01% to 400% according to a logarithmic

137   distribution and with 10 points per decade. Each measurement is duplicated and the sample is

138   removed and replaced before each test. Modulii and complex viscosity η^* were determined in the

139   linear viscoelastic regime (imposed strain γ= 0.5%) by applying a frequency sweep from 0.1rad/s to

140   100rad/s. The frequency distributions are also logarithmic with 10 points per decade and the

141   measurements are duplicated. All these measurements were achieved on bolus with various water

142   contents WC from 0.3 to 0.57 kg water / kg bolus on total wb (SC, SCP) and 0.68 to 0.8 kg water / kg

143   bolus on total wb (EF, EFP), in total wet basis.

144   ### *2.3. Flow properties and capillary rheometry measurements*

145   Flow properties are determined with a capillary rheometer with pre-shearing, Rheoplast, that

146   has been applied and described in detail by Della Valle, Vergnes & Lourdin (2007) and Nunez, Della

147   Valle & Sandoval (2010), who determined the viscous properties of starchy materials and cereal

148   foods, respectively, under extrusion conditions. In the following, we will only describe its main

149   working steps. In the case of food bolus, we did not use the pre-shearing function, because the

150   fragmentation has already been performed when preparing bolus with the blender.

151   All the tests are made at room temperature (T = 23°C). The measuring chamber before the

152   capillary die is fed with the bolus by the vertical motion of an annular piston at a constant speed of

153   1mm/s. Then, the injection piston (diameter D_P = 16mm) is moved vertically at a constant speed of

154   0.5mm/s to fill the capillary die. Three capillary dies (diameter D = 1mm, entry angle = 90°) are used

155   with different die ratios L/D (32, 16 and 8). After a 15 s relaxation time, pressure is measured for

156   different decreasing speeds of the injection piston, each speed step for a time interval of about 15s,

157 ranging from 2 mm/s to 0.002mm/s, which leads to apparent shear rate values $3000 \geq \dot{\gamma}_{app} \geq 3 \text{ s}^{-1}$.

158 At the end of this sequence (about 2 mn), pressure measurement is repeated at the two largest

159 speed values, in order to check sample stability, and a pressure profile P ($\dot{\gamma}_{app}$) is obtained.

160   This procedure has been applied to every sample for the same range of water content values

161 as for oscillatory shear measurements (2.2). 15 to 20g of bolus are needed to obtain a single pressure

162 profile. Data treatment is performed according to usual procedure for capillary rheometry as

163 described in detail by Della Valle et al. (2007) and Nunez et al. (2010). The wall stress τ_w is derived

164 from pressure measurements, using Poiseuille law, after application of Bagley's corrections. The flow

165 index *n* is determined and the real value of the shear rate $\dot{\gamma}_w$ is calculated after Rabinowitsch

166 analysis. Finally, the shear viscosity η is defined as the ratio $\tau_w/\dot{\gamma}_w$. Using entrance effects from

167 Bagley correction, the elongational viscosity η_E and the deformation rate $\dot{\varepsilon}$ may also be derived,

168 according to the Cogswell's method developed for melts polymers (Cogswell, 1972).

169   Flow curves (τ_w or η ($\dot{\gamma}_w$)) obtained for different moisture contents were fitted according to

170 appropriate rheological model and then shifted (translated) into a master curve to determine the

171 model parameters. This procedure, based on the time-temperature superposition principle, for the

172 rheological properties of polymers, has been already adapted by Della Valle et al. (2007) to the time-

173 plasticizer content superposition, in the case of plasticized starches. In this study, the shifting factors

174 were derived using Python (Python Software Foundation) according to the procedure developed by

175 Saboo *et al.* (2018).

176   ***2.4 Imaging***

177   Fragmented boluses of sponge cakes and extruded foods were prepared in the same way as

178 for rheological measurements and water was added to obtain the necessary dilution before inserting

179 between glass blades, and observed on a binocular stereomicroscope Leica with retro-lighting (x 35 ;

180 Leica Microsystems, Conrad Electronics, France). No specific staining was used.

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182

183 **3. Results and discussion**

184   ***3.1. Viscoelastic properties***

185   Within the linear domain ($\gamma < 1\%$), harmonic measurements performed at small strain

186 amplitude, led to similar bolus mechanical spectra (G'(ω), G'' (ω)) whatever the food considered and

187 its water content (Fig.1) : G' and G'' were slightly increasing functions of pulsation (or frequency) and

188 all bolus exhibited a storage modulus larger than the dissipative one (G' > G'') in the range of

189 frequency tested, and tan δ remained nearly constant (≈ 0.2). Clearly boluses behave more like solid

190 than fluids. Such spectra are usually typical of a viscoelastic network, with the frequency window of

191   the test framing a part of the viscoelastic plateau. However, in the case of these artificial bolus, no

192   structural interpretation can be given for the entities involved in such a network, at this stage. In

193   many cases, G''(ω) curves exhibited a shallow minimum within the frequency interval (0.1 < ω <1

194   rd/s). The frequency and the G' value corresponding to this G'' minimum can be taken conventionally

195   as representative of the characteristics (frequency and modulus) of the viscoelastic plateau

196   (Ferry,1980). However, the frequency of the minimum depended on the bolus, and in some cases, it

197   was not really observed. Therefore, for all our samples, we took the G' value at 1 rad/s as an

198   empirical measure of the viscoelastic plateau modulus. Fig.1 also showed that, for all food boluses, a

199   regular decrease of both modulii was observed as water content increased, which, as expected,

200   indicated that water acts as a lubricant. It also suggested that, for SC bolus, the enrichment in protein

201   slightly increased the values of modulii, and reduced its sensitivity to water content. Conversely, pea

202   based extruded snacks (EFP) led to boli with much larger moduli, about 10 times, than extruded flat

203   bread (EF), but displayed similar water sensitivity.

204   The values of storage modulus at viscoelastic plateau G'(ω ≤1 rd/s) were negatively

205   correlated to the water content (Fig.2). These variations can be described by the equation :

206            $G' = G'_0 * \exp [- \alpha_G * WC]$ (1)

207   G'_0 represents the theoretical value of storage modulus for dry bolus ; parameter α_G. reflects

208   the interaction of the food with water and can be envisioned as a water plasticization coefficient. The

209   values of these parameters are reported in Table 2. Similar results could have been obtained by

210   shifting and superimposing G' (and G'') curves for a given moisture content as performed by van der

211   Sman and Mauer (2019), in the case of starch/sugar/polyol mixtures, and by Costanzo et al. (2019)

212   for gluten / water / ethanol blends. Besides extending the time-temperature superimposition

213   principle to time-solvent in the case of bioplymers, these works underlined the importance of

214   intermolecular interactions, mainly H bonds. Such interactions could be captured by the ratio of

215   sample temperature to glass transition temperature of the biopolymer, which is a function of its

216   solvent content. Regarding food boluses, our results suggest that the protein enriched sponge cake

217   (SCP) interacts the least with water, likely because of the hydrophobic nature of added pea isolates,

218   whereas globulins interactions with glutenins might also be infered for the decrease of water

219   sensitivity (Lambrecht, Deleu, Rombouts & Delcour, 2018). Conversely, extruded pea flour (EFP)

220   shows larger interaction with water than flat bread (EF), likely because hemicellulosic compounds,

221   contained in pea flour, have a strong water retention capacity that largely balances the hydrophobic

222   influence of pea isolates observed for SCP (Kristiawan et al., 2018).

223   For all moisture contents and foods, the variations of G' and G'' moduli with the applied

224   strain γ exhibited the same patterns (Fig. 3). From this graph, three main zones may be delineated:

225 first the linear viscoelastic regime ($\gamma \leq 1\%$), where modulii are constant, with G'> G'' like for a gel,

226 then, a transient regime, where storage modulus decreases and crosses G'' curve, which can be

227 considered as plastic region, and, finally, a non-linear regime where both modulii decrease, with G'' >

228 G', indicating that bolus starts to flow. This behaviour is typical of the loss of structure and of the

229 flow of the material under increased strain. Since it holds for all bolus samples, we can define the

230 stress τ_c, at the crossing of G' and G'' as a characteristic parameter of this behaviour:

231              $\tau_c = \gamma_c * G^*$ (2)

232   G^* being the value of the complex modulus and γ_c the strain value at intersection of G' and

233 G'' curves (G'=G''). Another characteristic stress maybe defined, by the value obtained at the end of

234 the linear viscoelastic domain, defined by the crossing of the horizontal tangent of G' in the linear

235 domain with the tangent to G' curve during plastic flow (see Fig.3). It is found that these values are

236 highly correlated to the values of τ_c ($r^2 > 0.9$, see Appendix 1), so, in the following, τ_c is considered to

237 characterise the transition of bolus to flow.The values of τ_c are negatively correlated to the water

238 content (Fig.4). These variations can be described by the equation :

239              $\tau_c = \tau_{c0} * \exp [- \alpha_{\tau_c} * WC]$ (3)

240   τ_{c0} is the theoretical value of characteristic stress for the dry bolus; parameter α_{τ_c} reflects the

241 interaction of the food with water, like a water plasticization coefficient. The values of these

242 parameters are reported in Table 2. At first glance, they confirm the trends observed in the linear

243 viscoelastic domain, when considering α_G values : protein enriched sponge cake (SCP) interacts the

244 least with water, again due to the hydrophobicity of pea isolates, whereas extruded pea flour (EFP)

245 shows larger interaction with water than flat bread (EF), which, again, may be attributed to the

246 presence of hemicellulosic compounds in pea flour. However, for extruded products, the values of

247 α_{τ_c} are larger than those of sponge cakes, and the variations of characteristic stress obtained for EF

248 and EFP are close to each other (Fig.4). This result indicates that the stress needed to make the bolus

249 flow is more influenced by water for extruded cereal foods, than for sponge cakes, whether they are

250 enriched in plant proteins or not. Moreover, the huge value of initial stress τ_{c0}, found for extruded

251 products (Table 2, $\tau_{c0} > 100$ MPa), confirms that, overall, large amounts of water (WC $\geq$ 60%) are

252 needed to elaborate a flowable bolus from these foods that contain a large amount of hydrophilic

253 compounds like amorphous starch.

254

255   ***3.2 Flow properties***

256   From the pressure profiles P ($\dot{\gamma}_{app}$) obtained for different capillary geometries (L/D ratio),

257 and using Poiseuille relation, flow curves can be derived, representing the variations of the wall shear

258   stress τ_w as a function of apparent shear rate or τ_w ($\dot{\gamma}_{app}$)) for food boluses at different amount of

259   water WC (Fig.5). As expected, the larger WC, the lower the apparent viscosity, defined by the ratio

260     $\tau_w/\dot{\gamma}_{app}$: flow curves show that viscosity decreases by a factor of 10 when WC is increased by 0.10

261   for SC foods, whereas the decrease is lower for extruded foods. Some curves display a dispersion of

262   experimental points, especially at larger moisture content. Indeed, in these conditions, flow becomes

263   less steady and measurement accuracy lower, given the low level of pressure reached in the

264   capillary, and also possible slip-stick phenomena. However, all curves show a similar increasing trend,

265   and some of them suggest the existence of a yield stress at lower shear rate (like for SC at WC= 0.28

266   and 0.36, for instance). Therefore, after having converted apparent shear rate values $\dot{\gamma}_{app}$ into wall

267   shear rate $\dot{\gamma}_w$, using Rabinowitsch correction, all curves are fitted using the Herschel-Bulkley model,:

268         $$\tau_w = \tau_s + K * \dot{\gamma}_w{}^n \qquad\qquad (4)$$

269   where τ_s is the yield stress, K the consistency (Pa.s^n), and n the flow index.

270   Although fitting is acceptable ($r^2 > 0.75$), considering the dispersion mentioned before, it is thought

271   that determining coefficents τ_s, K and n for each WC value could increase the uncertainty. Moreover,

272   the flow index does not change significantly for each product, whatever the value of WC. Therefore,

273   for each product, a master curve is determined by adapting the time-temperature principle, to

274   superpose flow curves, obtained from bolus at different water content. Hence, each flow curve is

275   shifted to a reference curve, at a given WC value, by plotting the reduced shear stress (τ_w / a) as

276   function of reduced shear rate ($\dot{\gamma}_w * a$), a being the shift factor. This procedure is illustrated in

277   Appendix 2. The Herschel-Bulkley coefficients of the reference curve are recalled in Table 3, for each

278   product. The values of the shift factor a vary between 0.016 and 27.8 and they are directly correlated

279   to the water content, all food products being considered together (Fig.6a). In a first approach, this

280   correlation suggests that the influence of water on the viscous behavior of the bolus in shear flow is

281   similar for the four different cereal foods. Secondly, by applying (eq.4) to (τ_w/a) [$a*\dot{\gamma}_w$], this

282   correlation can be used to calculate, for any WC value, the corrected values for the coefficents of the

283   Herschel Bulkley model by :

284         $$\tau_{sc} = \tau_s\,/\,a(WC) \qquad and \qquad K_c = K\,/[a(WC)]^{n+1} \qquad\qquad (5)$$

285   The numerical values of τ_{sc}, K_c and n, for all values of WC and products tested, are reported

286   in Appendix 3. Note that the value of flow index n did not vary with WC. Furthermore, using

287   correlation from Fig.6a and eq. (5), flow curves for the boluses of the four products can be derived

288   for any WC value. For instance, for WC=0.6, taken as a typical value in the interval [0.28, 0,8], flow

289   curves representing the shear viscosity η as a function of shifted shear rate ($\dot{\gamma}_w\,.\,a$) can be drawn

290   (Fig.6b). These curves show that for low shear rate values, the viscosity of the bolus is very close to

291   each other, whereas for larger values ($\dot{\gamma}_w\,.\,a > 10s^{-1}$), the two extruded products (EF and EFP) lead to

9

292   much larger viscosity than sponge cakes, whether they are enriched in plant protein or not. The

293   increase of slope, observed at lower shear rate values, underlines the presence of the yield stress,

294   and it is enhanced for SC and EFP. In line with the preceding results obtained for the viscoelastic

295   properties, the variations of the Herschel-Bulkley coefficients τ_{sc} and K_c with WC can be represented

296   (Fig.7) and fitted by the following relations :

297        $\tau_{sc} = \tau_{s0} . \exp [-\alpha_\tau * WC]$ and $K_c = K_{c0} . \exp [-\alpha_K * WC \}$ (6)

298   from which the values of τ_{s0}, α_τ, K_{c0} and α_K are extracted and reported in Table 2. In

299   addition, variations of consistency with water are very close to each other for the four products (Fig.

300   7b), and a common trend can be found for K_c variations, giving a mean value for α_K= 10.25 (r^2= 0.9).

301   Regarding α_τ and α_K variations, the trends obtained here are different from those observed for the

302   viscoelastic properties since standard products (SC and EF) display the lower α_τ and α_K values.

303   Moreover, α_τ and α_K values are correlated. These results show that, during the flow, the products

304   enriched with plant proteins interact more with water than their standard counterpart. In other

305   words, the presence of proteins increase the bolus flow sensitivity to water, which suggests that flow

306   could contribute to bolus destructuring by disrupting food particles or releasing protein aggregates.

307   The same procedure can be applied to determine the apparent elongational viscosity of

308   boluses, by taking into account entrance pressure effects from capillary rheometry tests. Then, using

309   the same time-water superposition principle, master curves can be determined, for instance at WC

310   =0.6 (see Appendix 4). Indeed, very little differences can be observed between the four foods.

311

312   ***3.3 Bolus structure and rheological properties relationships***

313   First, the values found for coefficients α were in the same numerical interval [5, 31],

314   regardless the rheological property. These values are in the same interval as the values of coeficients

315   of starch plasticization by water, found for starchy materials and cereal foods under extrusion

316   conditions (Della Valle et al., 2007; Nunez et al., 2010). Clearly, this result suggests that starch /water

317   interactions have a significant role in the rheological behavior of the bolus and its breakdown during

318   FOP. However, these values stand for starch melts as encountered during extrusion, i.e. at high

319   temperature and shear, where starch granules are broken and polymers released. Food boluses can

320   hardly be compared to starch melts. By the way, Cox-Merz rule does not apply, unlike polymer

321   melts, since complex viscosity always took values larger than those of steady shear viscosity (as

322   illustrated in Appendix 5), which reflects that the structure of bolus changed during steady flow.

323   Moreover, boluses are made of particles, of typical size 1mm, which are not expected to

324   fragment during flow, but rather to deform, as shown by the micrographs of diluted boluses (Fig.8).

325   Sponge cake (SC) bolus presents a rather uniform appearance of a cohesive mass of blurred

326   aggregated particles, whereas enriched protein sponge cake (SCP) is more heterogenous, with darker

327   parts and bigger entities (> 1 mm), separated by water. Extruded flat bread bolus (EF) also presents a

328   cohesive morphology, whereas extruded pea flour bolus (EFP) presents a clear picture of

329   agglomerated and elongated particles (length > 1mm, width < 1mm). These images suggest that the

330   foods are not destructured in the same way after fragmentation and hydration. So it is not surprising

331   that their characteristic rheological properties are not fully correlated.

332   In spite of these visual differences, they may be all envisioned as concentrated supensions of

333   deformable particles (Fig.9). At lower strain the suspension is jammed, because of particles swelling

334   due to water absorption (Fig.9b), which gives the bolus a solid-like behavior, in the viscoelastic

335   domain. At larger strain, hence larger stress ($\tau > \tau_c$ or τ_s), the suspension may still be partially

336   jammed, because, in the case of standard foods, hydrophilic particles still swell (Fig.9c). Conversely,

337   the particles of protein enriched foods, being less hydrophilic, swell less, water acts as a lubricant

338   which favors bolus flow (Fig. 9d). This interpretation would explain why the values of coefficient α

339   referring to flow properties (yield stress τ_s and consistency K) are larger for the protein enriched

340   foods than for their standard counterpart. More experiments, for instance using measurements of

341   water absorption or swelling index on such products, and even comparison with settling ratio

342   (Boehm et al. 2019) could contribute to test this hypothesis. Whether this transition occurs at the

343   value of characteristic stress τ_c or yield stress τ_s is still to be determined precisely , although both

344   properties are correlated (Fig.10). The question is significant since both properties might be relied on

345   for the control of swallowing.

346   Clearly, before extending these rheological measurements to assess the interactions of saliva

347   with food, it is necessary to test the influence of other saliva components on bolus rheological

348   properties. Recently, these properties were measured on bolus made by mixing same foods (sponge

349   cake) with artificial saliva, containing salt and mucin, and no significant difference was detected

350   when compared with bolus made with water (Gibouin et al., 2019b). The influence of the addition of

351   α-amylase, another component of saliva, on bolus rheological properties is still questionable. There is

352   a general agreement that salivary α-amylase plays an important part in destructuring cereal foods,

353   like bread, during the gastric phase (Bonhorst & Singh, 2013; Pentikaïnen et al., 2014; Freitas,

354   Feunteun, Panouille & Souchon, 2018). But the short residence time of the food in the mouth may

355   explain the little influence of α-amylase on bread destructuring during FOP, assessed by viscosity

356   measurements (Le Bleis, Chaunier, Montigaud & Della Valle, 2016).

357   Regarding FOP, another point that confirms the relevance of our measurements comes from

358   some results obtained when characterizing real boluses. Results obtained by Loret et al. (2011)

359   measuring yield stress on cereal flakes boluses collected before swallowing for 11 subjects, hence for

360   different saliva contents, lead to a value of α of about 8, i.e. in the interval found in this study. For

361   sponge cake boluses, the values of coefficient α found for apparent viscosity were also in the same

362   interval (17 and 11 for standard and protein enriched sponge cake, respectively) (Assad-Bustillos et

363   al., 2020). Furthermore, they ordered in the same way as the values of $\alpha_{\tau c}$ found in the present

364   study (9.3 and 4.8, see Table 2). This result suggests that the coefficient $\alpha_{\tau c}$ contributes to assess

365   food-saliva interaction and that characteristic stress τ_c is the property that matters to define bolus

366   destructuring. Although more work is necessary to ascertain this trend, it is possible, and rather

367   simple, to extend these rheological methods to other foods. In turn, this would help to design and

368   test foods that would have a specific behavior during chewing.

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370

371   **4. Conclusion**

372   Our results show that the rheological properties of artificial food cereal boluses can be

373   determined in a wide range of strain and strain rates, using different methods, in the range of water

374   content encountered during FOP. For the four foods tested, hydrated boluses exhibited same

375   behavior. First, at low strain, in the linear viscoelastic domain, bolus had a gel like behavior

376   characterized by the viscoelastic modulus G*. Then, destructuring occurs at larger strain ($\gamma \geq 0.1$),

377   where a characteristic stress τ_c can be delineated from the intersection between curves of storage

378   and dissipative modulii. During steady flow, bolus behavior was found to follow Herschel-Bulkley

379   model from which yield stress τ_s and consistency K were derived; master curves could be determined

380   to compare the viscous behavior of the four products. At larger shear rate ($\dot{\gamma}_w \geq 10$ s^{-1}), viscosity was

381   found larger for extruded foods without any effect of protein content. The variations of these four

382   rheological properties (G*, τ_c, τ_s and K) with water followed an exponential decay, from which it was

383   possible to derive coefficients of interaction with water, for each property and product. Their

384   changes with product composition and structure were interpreted by envisioning the bolus as a

385   suspension of soft particles. The comparison of their values with those obtained for real boluses, and

386   their variations with protein content, suggest that rheological mesurements are helpful to

387   characterize the interaction of food with saliva. Finally, given the interest to study the water

388   dependence of rheological properties of cereal food boluses, the same approach could apply to

389   other food products too.

390

391

392   **Acknowledgements**

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400 **References**

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498   parameters given in Table 2, whereas thick dotted line (light grey) in (b) features the best fit

499   obtained by taking all boluses together into account ($K_c = 180$ exp [-10.25 * WC] , r^2=0.9).

500

501   Figure 8: Micrographs of the artificial boluses of (a) standard sponge cake SC, (b) sponge cake

502   enriched with proteins (SCP, white stain on bottom left is water between food particles), (c)

503   extruded flat bread (EF) and (d) extruded pea flour EFP. Image width is 1cm. No specific

504   staining was used, so differences of shade (light / dark) reflect the matter concentration, i.e.

505   non-uniform thickness of the sample layer between blades, rather than specific composition

506   or structural state.

507

508   Figure 9: Schematic representation of bolus structure, envisioned as a suspension of more or less

509   swollen and deformable spherical particles: dry ground food (1mm) (a), wet particles in low

510   strain domain (or $\tau < \tau_s$ or τ_c) (b); destructured suspension of particles of standard (c) and

511   protein enrich foods (red spots symbolize proteic component (d)), at large strain ($\tau > \tau_s$ or τ_c

512   ).

513   Figure 10: Variations of the values of yield stress τ_s computed for the same WC as characteristic

514   stress τ_c for the boluses of the four cereal foods SC (O), SCP (●), EF (□) and EFP (■).

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Gibouin et al.   Fig.1   Rheology artificial bolus

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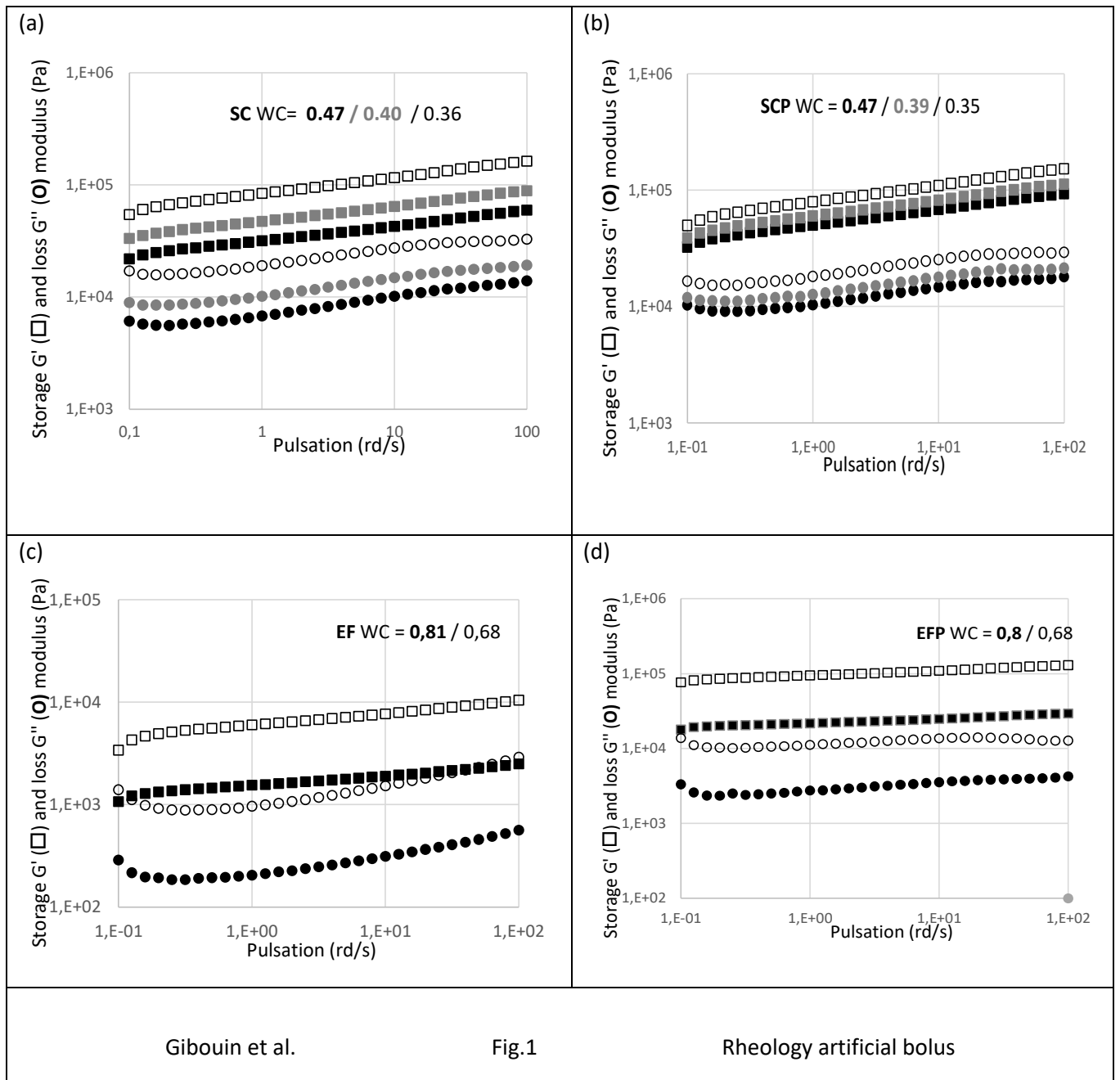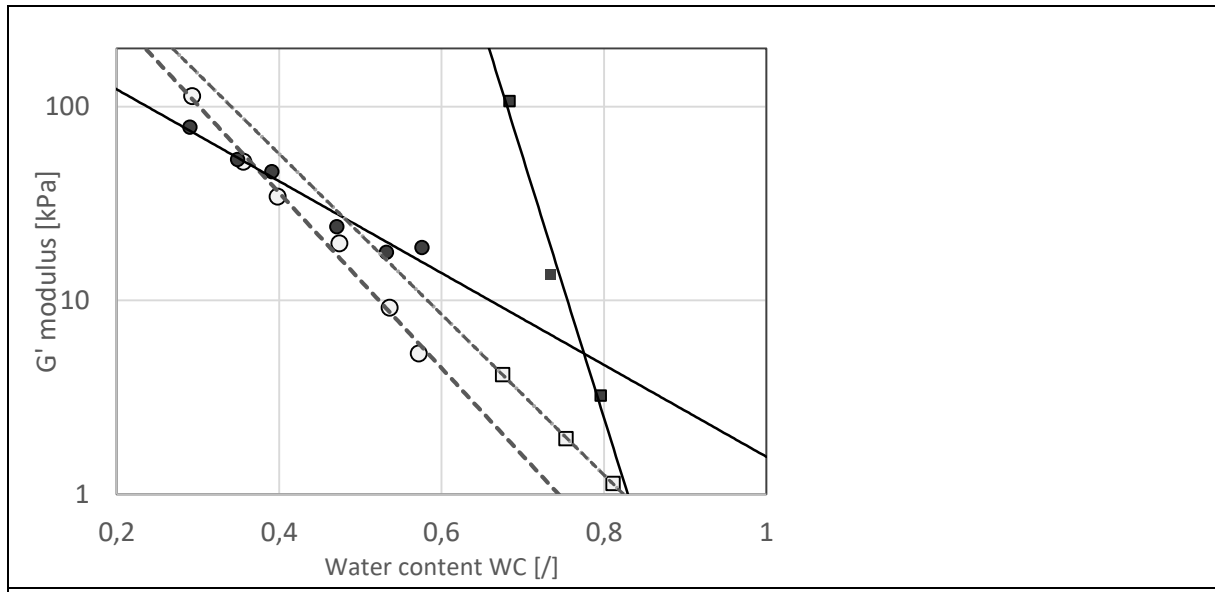
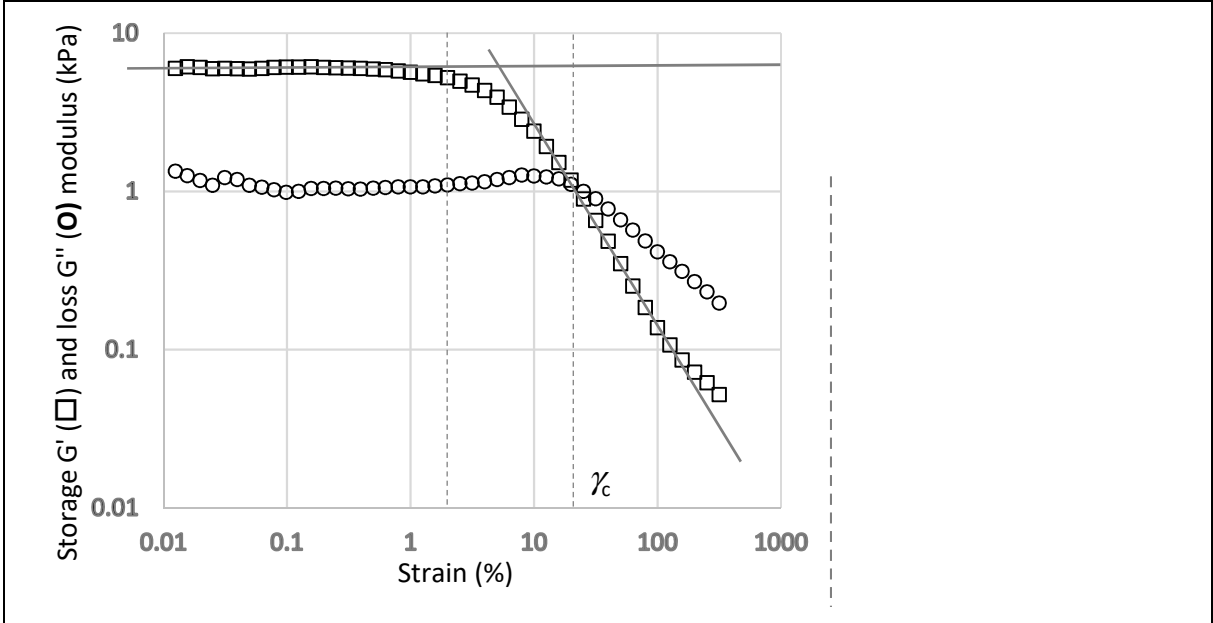GiObouin et al.   Fig.2   Rheology artificial bolus

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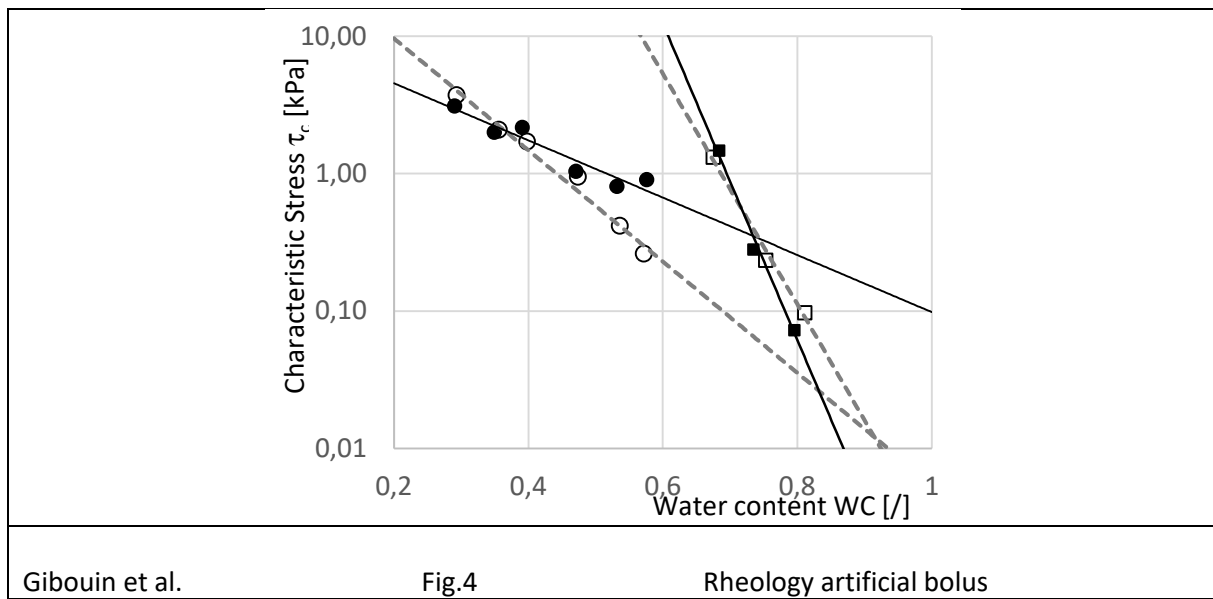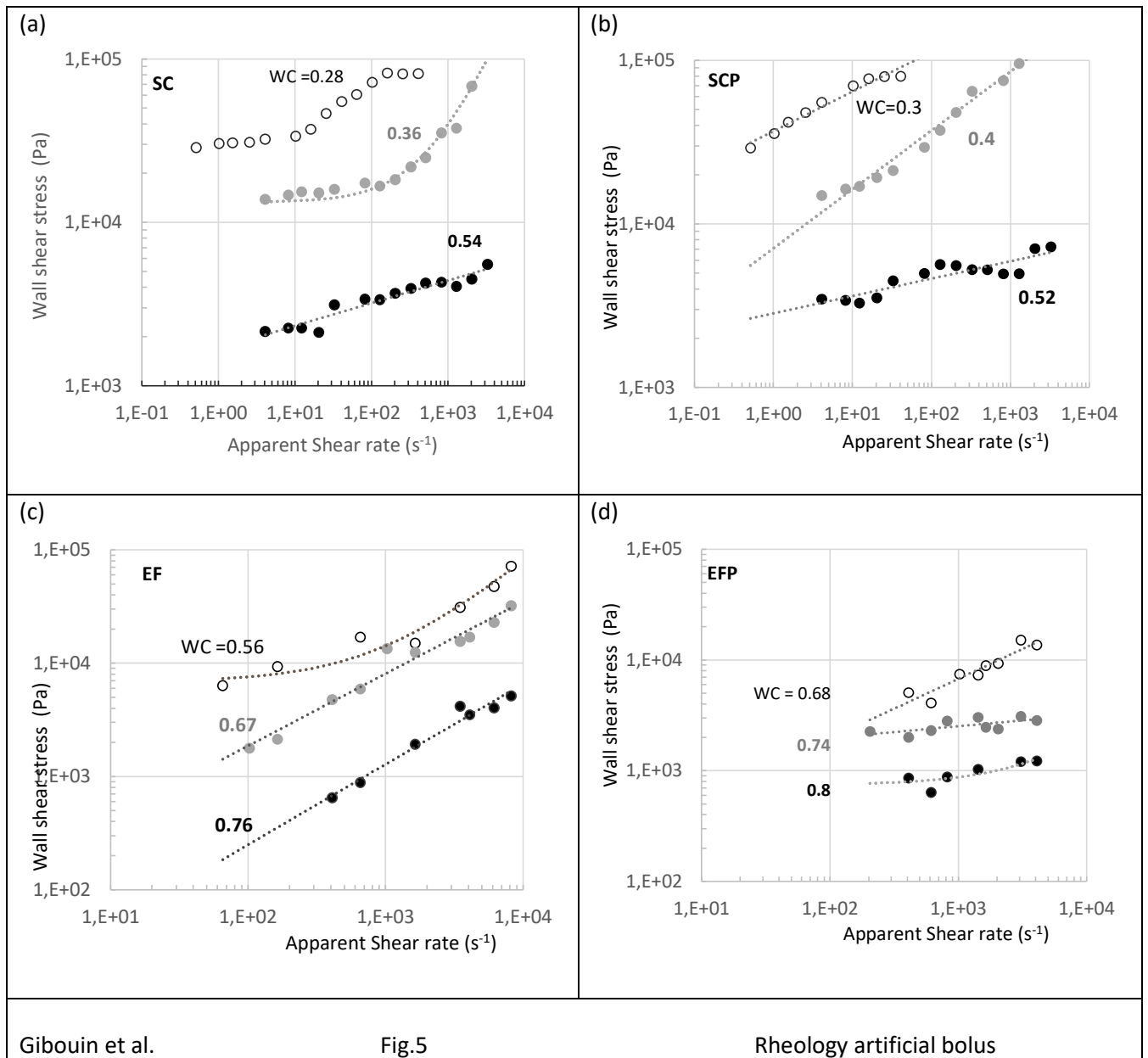Gibouin et al.   Fig.3   Rheology artificial bolus
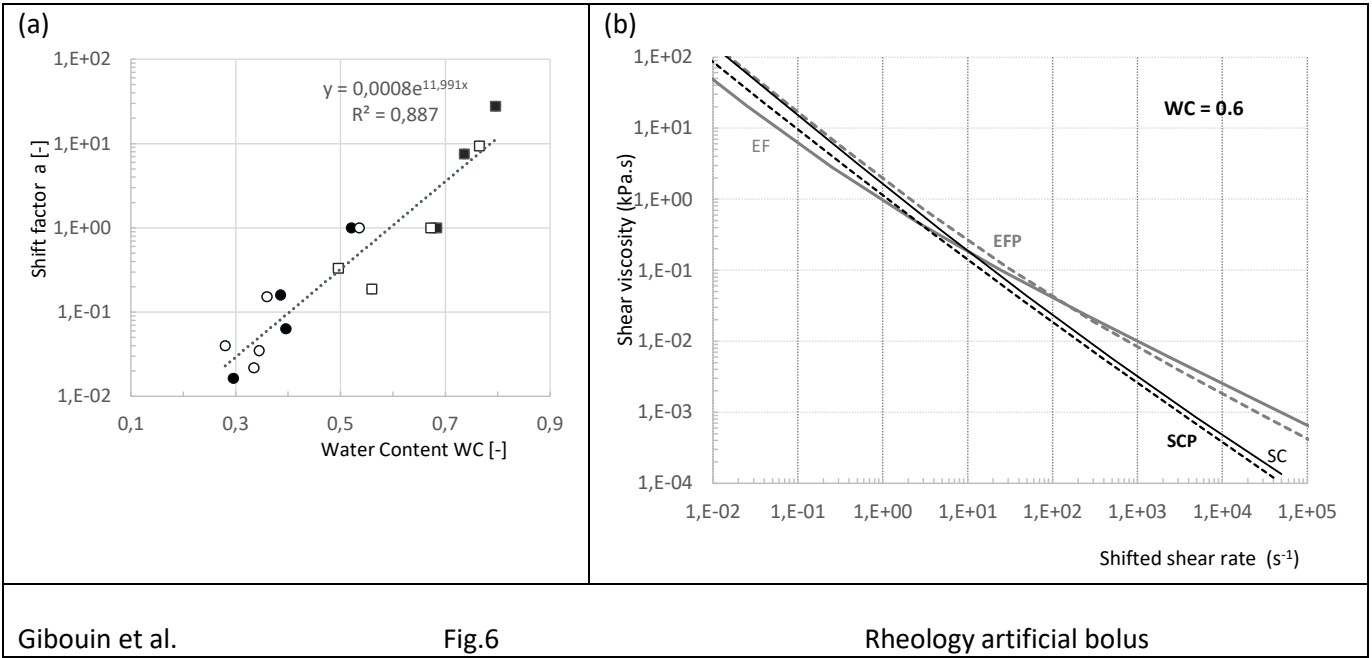
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| Gibouin et al. | Fig.4 | Rheology artificial bolus |

Gibouin et al.   Fig.5   Rheology artificial bolus

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525

(a) Shift factor a [-] vs Water Content WC [-]

$y = 0{,}0008e^{11{,}991x}$
$R^2 = 0{,}887$

(b) Shear viscosity (kPa.s) vs Shifted shear rate (s⁻¹)

WC = 0.6

EF

EFP

SCP   SC

Gibouin et al.   Fig.6   Rheology artificial bolus

526

527

528

(a)

Yiled Stress τ_{sc} (kPa)

Water content WC [/]

(b)

Consistency K_c [kPa.s^n]

Water content WC [/]

Gibouin et al.   Fig.7   Rheology artificial bolus

529

530

(a) (b) (c) (d)

Gibouin et al.   Fig.8   Rheology artificial bolus

535

(a)	(b)	(c)
		(d)
Gibouin et al.	Fig.9	Rheology artificial bolus

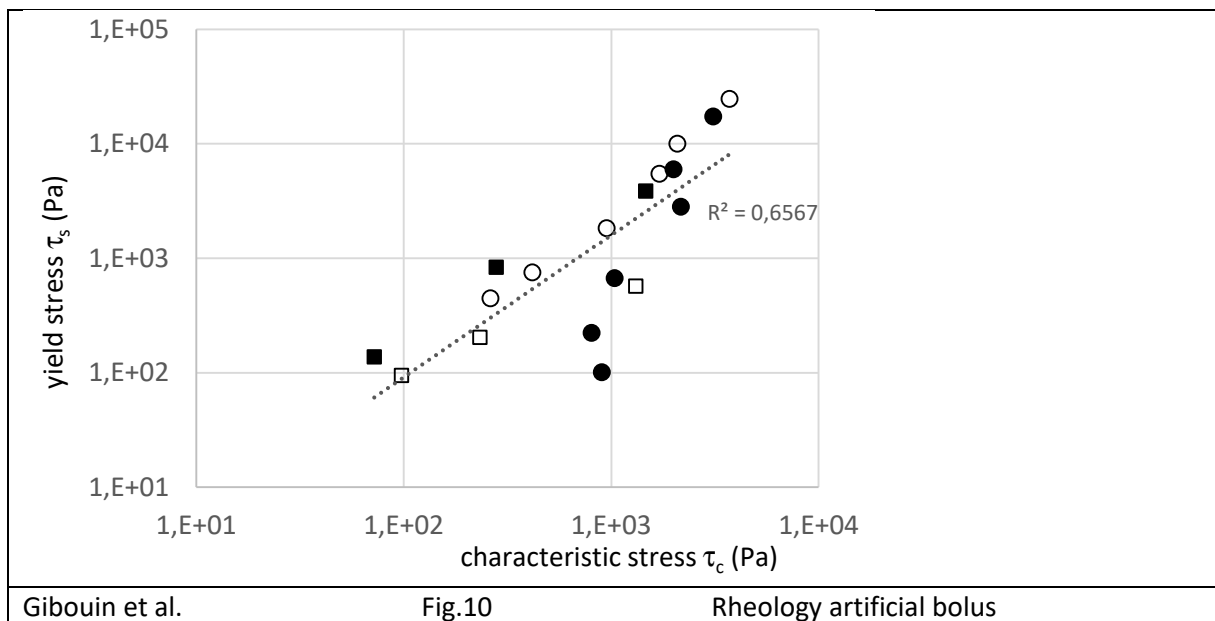536

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| Gibouin et al. | Fig.10 | Rheology artificial bolus |
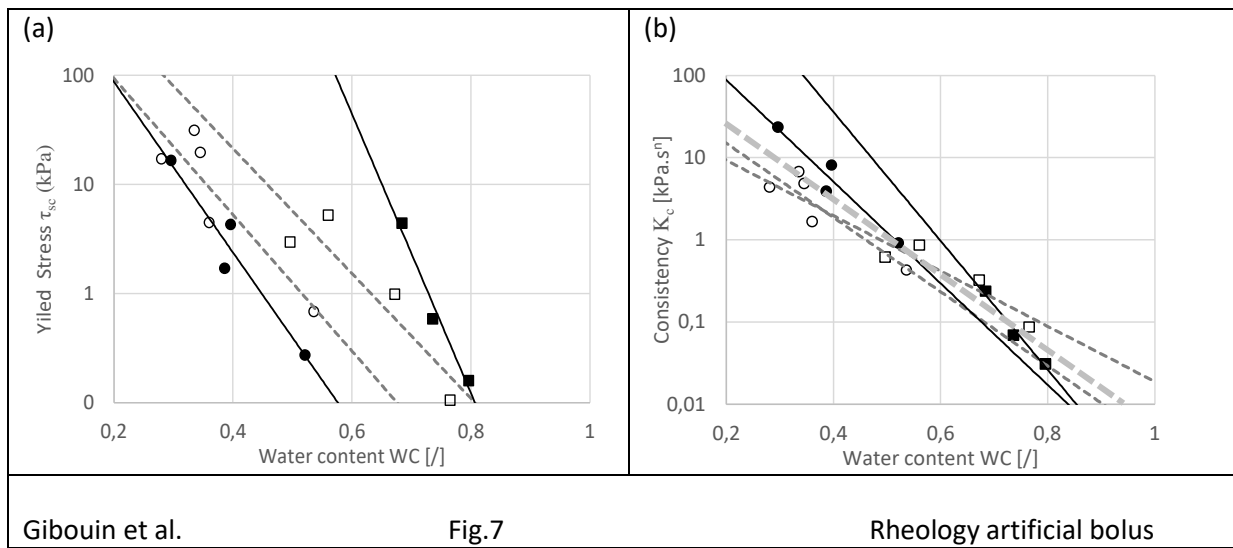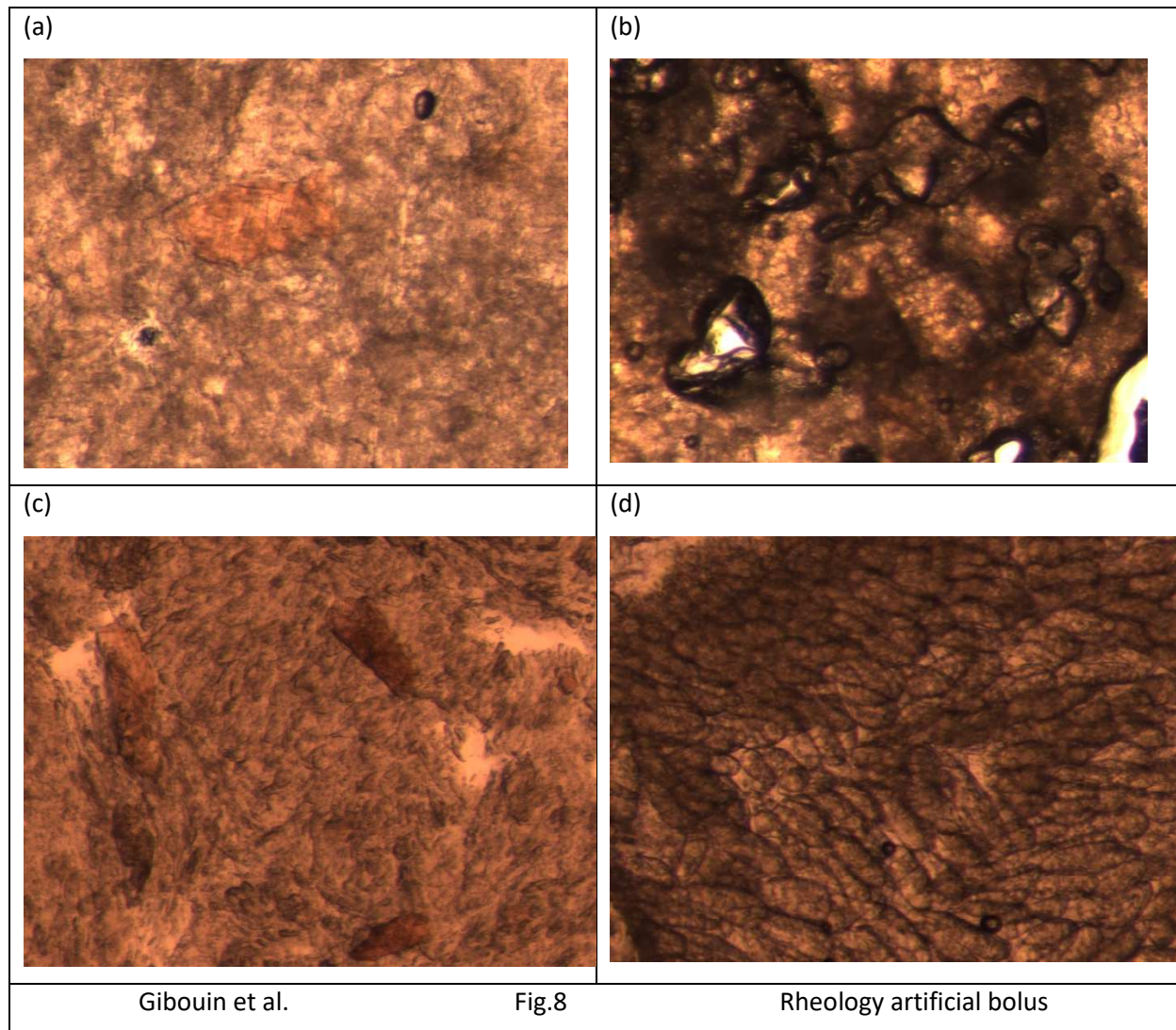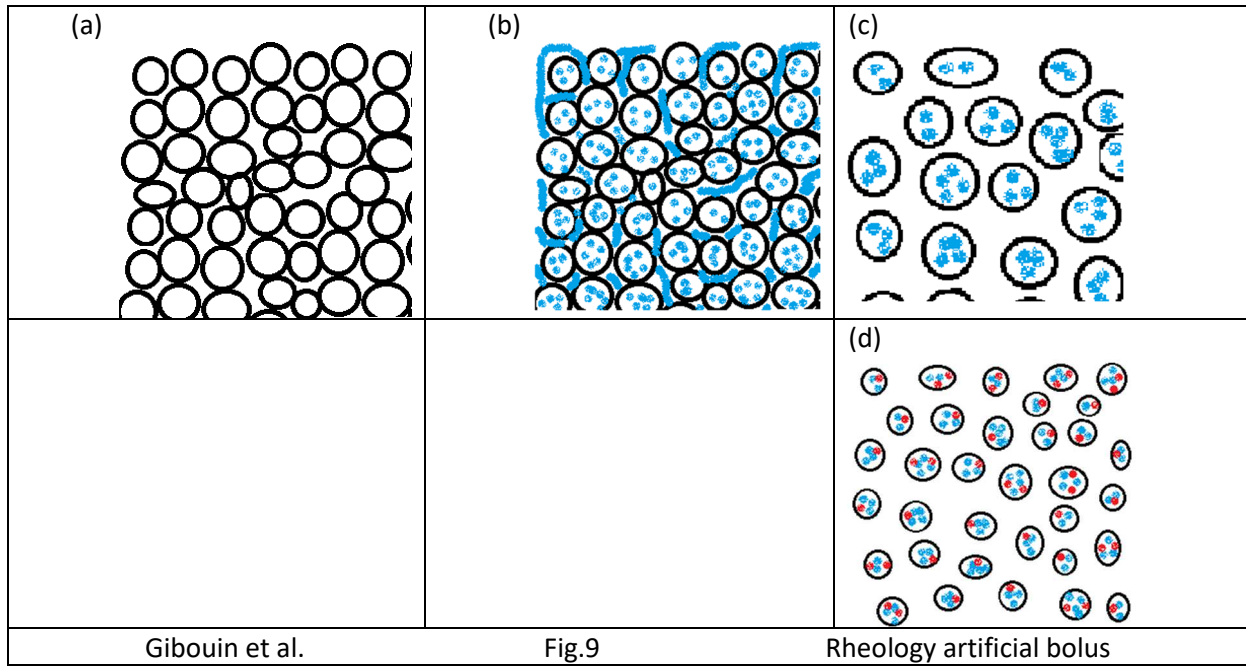
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542 Table 1: Composition (% total wet basis) and density of the four tested foods, sponge cake (SC) and
543 extruded (E), and their protein enriched version (SCP and EFP).

Food	Sponge cake		Extruded	
Code name	**SC**	**SCP**	**EF**	**EFP**
Protein	11	13	13	23
Fat	6	5	5	-
Starch	18	13	67	42
Sugar	27	26	4	3
Cellulosic compounds (+ others)	10	13	5	22
Water content (%, tot. wet mat.)	28	30	5	10
Density ($g.cm^{-3}$)	0.21	0.23	0.14	0.15

544

545

546 Table 2: Values of the parameters of the models representing the relations between bolus
547 rheological properties and water content WC (see eqs 1, 3, 6).

	Viscoelastic domain				Flow regime			
Parameter	G'_0	α_G	τ_{c0}	$\alpha_{\tau c}$	τ_{s0}	α_τ	K_{c0}	α_K
Food	MPa		kPa		kPa		$kPa.s^n$	
SC	2.34	10.4	57.7	9.3	1670	14.4	120	10.4
SCP	0.37	5.5	12	4.8	3190	18.0	1510	14.2
EF	2.6	9.5	$5.8\ 10^5$	19.3	4200	13.2	44	7.7
EFP	$1.4\ 10^8$	30.8	$1.04\ 10^8$	26.5	$2.15\ 10^9$	29.5	49110	18

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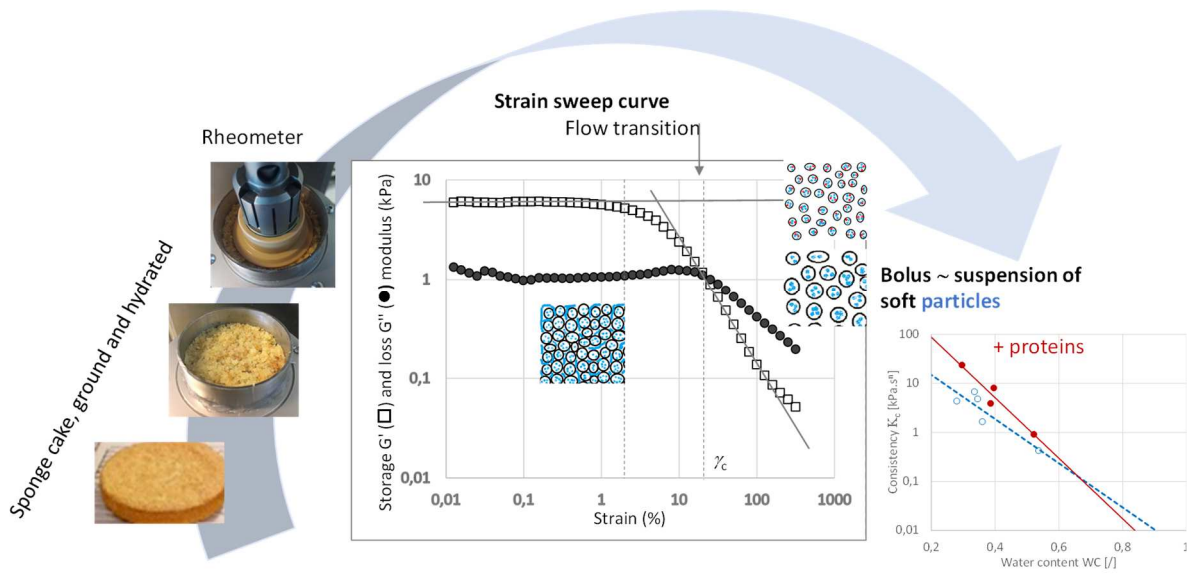549

550 Table 3: Values of Herschel-Bulkley coefficients (see eq. (4)) for each product at chosen reference
551 value of WC.

Food	WC ref	τ_s (Pa)	K ($Pa.s^n$)	n
SC	0.54	685	430	0.28
SCP	0.52	270	915	0.21
EF	0.67	985	325	0.41
EFP	0.68	4410	235	0.39

552

553

Graphical abstract of measuring rheological properties of artificial boluses of cereal foods and determining interaction coefficient.