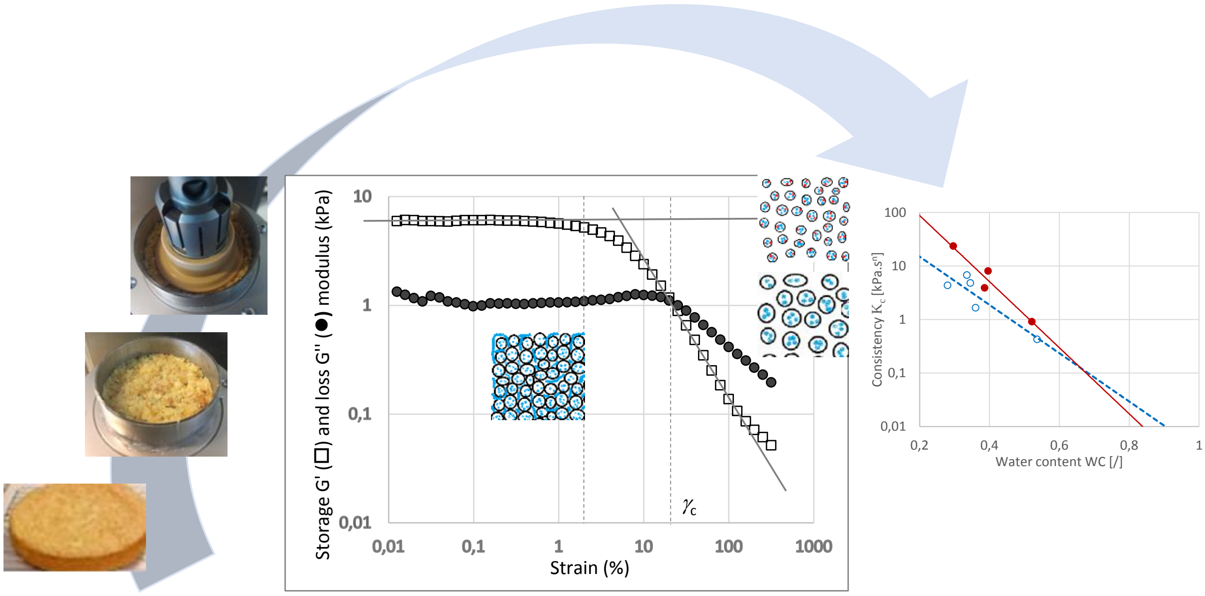
Highlights

- Rheological properties of artificial food boluses are determined in FOP conditions
- Cereal foods bolus behave like a gel, destructured for a characteristic stress τ_c
- A food/water interaction coefficient α is derived from τ_c variations with water
- Cereal foods bolus can be considered as a suspension of soft swollen particles
- For flow properties, α values are increased when adding protein to the food

Graphical Abstract



Cereal food (sponge cake) is ground and hydrated before testing on rheometer to determine the flow transition (here by strain sweep) of the suspension of swollen soft particles (inserted sketches) and the influence of protein addition (symbolized in red) on plasticization by water, is reflected by the variations of bolus consistency with water.

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

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Abstract

The properties of artificial food bolus are studied by dynamic oscillatory and capillary rheometry as functions of bolus water content, in the usual range of saliva hydration, for four cereal products: sponge cake, extruded flat bread and their counterpart enriched in legume proteins. All boluses followed the same rheological behaviour characterised by (1) solid -like in the linear viscoelastic domain and (2) Herschel-Bulkley model for large shear strain. Hence, four characteristic rheological properties are determined: modulus at viscoelastic plateau, characteristic stress at transition to flow, yield stress and consistency in the flow regime. Water content considerably decreased these properties according to an exponential decay, which allowed determining interactions coefficients ($5 \leq \alpha \leq 30$). These values are of the same order of magnitude as the plasticization coefficient of starch by water. They were larger for the extruded pea based (EFP, $\alpha \geq 15$), and were lower for the sponge cake (SC, $\alpha \leq 15$). The variations for the different rheological properties are discussed in terms of matter state, envisioning bolus as a suspension of soft swellable particles. The comparison of these values with those encountered for real boluses from similar foods suggests that these results contribute to define a coefficient of interaction of food with saliva.

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

Nomenclature

a	shift factor used to derive the flow master curve
D, D_P	diameters of rheometer capillary die and piston, respectively
EF, EFP	extruded flat bread end extruded pea based snack, respectively
G', G'', G^*	storage, dissipative and complex modulus, respectively
G'_0	theoretical value of storage modulus for dry bolus
K, K_c	consistency and its corrected value in the Herschel-Bulkley model
K_{c0}	theoretical value of consistency for the dry bolus
L/D	length to diameter capillary die ratio
n	flow index in the Herschel-Bulkley model
SC, SCP	sponge cake, enriched in pulse protein, respectively
T	temperature
WC	Water content, expressed on a total wet basis
α_G. $\alpha_{\tau c}$, α_τ,α_K	interaction coefficients of food with water based storage modulus, characteristic stress, yield stress, consistency, respectively
γ, γ_c	strain and its value at intersection of G' and G'' curves
$\dot{\gamma}_{app}$, $\dot{\gamma}_w$	apparent and real wall shear rate
$\dot{\varepsilon}$	deformation rate
η_{app}, η^*, η, η_E	apparent, complex, shear and elongational viscosity, respectively
ω	pulsation (oscillatory rheometry tests)
τ_w, τ_c , τ_s , τ_{sc}	wall shear, characteristic, yield (Hersche-Bulkley), corrected stress
τ_{c0}, τ_{s0}	dry bolus theoretical values of characteristic and yield stress, respectively

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).

Salivacontains 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.

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) to the uncertainty about the relevant rheological property for food oral processing. To overcome the former difficulty, artificial boluses can be considered, provided that they are representative of real ones. By doing so, it is possible to address the latter difficulty by performing different rheological tests under a wide range of strain and strain rate conditions.

Given this context, the aim of this work is to determine the rheological properties of artificial boluses of cereal foods in order to derive coefficients that help to quantify the interactions of foods with saliva. For this purpose, artificial boluses are prepared under conditions as close as possible to those encountered in FOP, to avoid physiological interindividual variability and to focus on bolus rheology and the influence of water content. Bolus viscoelastic properties are investigated in the linear and non-linear viscoelastic domains, using oscillatory shear rheometry, and flow properties are measured by capillary rheometry. In addition to insights on food bolus structure, the variations of the rheological properties with WC are determined for two types of cereal foods: a soft food with intermediate moisture and a brittle dry one, as well as their corresponding version enriched in plant protein.

2. Material and methods

2.1. Cereal foods and bolus preparation

Four cereal food products were studied: two soft foods (sponge cake, SC) and two brittle ones (extruded, E). One SC had a standard formulation (SC) and the other one was enriched with pea proteins and held the claim “high in protein” (SCP). Both were provided by CERELAB® (Aiseray, France). One extruded food is a commercial flat bread (EF, Les craquantes EPI d’OR ™) and the other one is made by extrusion of pea flour (EFP), as described in detail by Kristiawan et al. (2018). Their composition, moisture content and density are reported in Table 1. Besides various differences, especially starch content, the difference of water content makes the food soft (SC) or brittle (E).

In order to prepare artificial boluses, foods were fragmented and impregnated with water, the main component (99%) of human saliva. This study focuses on the role of water, because the addition of mucin and salts has been precedingly shown not to modify the rheological properties of sponge cake boluses (Gibouin, Della Valle & van der Sman, 2019 a,b). In addition, the role of α-amylase on rheological properties is discarded, in a first approach. The role of real saliva, and especially of α-amylase will be discussed when comparing our results with those obtained for real boluses, in section 3.3. Water was added at different levels, in order to obtain a bolus, homogeneous at bare eye, and to cover the range of water content of food bolus during chewing, in agreement with values found in literature, up to 60% for sponge cake and 80% extruded foods, in total wet content (Assad-Bustillos et aL, 2019a; Loret et al., 2011).

Before water addition, sponge cakes and extruded foods were fragmented with a commercial blender(MagiMix, France) or manually using a mortar, respectively, until a particle mean size of 1mm is reached, as measured with a vibrating sieves (Reitsch, F95610 Eragny). This size value is chosen close enough to the particle size in real boluses at the first step of chewing. It has also been checked that the particle size distribution is in agreement with the distribution in real boluses (Assad-Bustillos et al., 2019b), and that in the range [1, 10mm], the particle size did not affect significantly the rheological properties (Gibouin et al., 2019 a,b).

2.2. Viscoelastic properties and oscillatory shear measurements

Viscoelastic properties of bolus were determined with a controlled strain rheometer (ARES, TA Instruments), provided with a plate-plate circular geometry: diameter = 4.0cm and gap = 2.5mm. The temperature is controlled with a Peltier device and set to 23°C. The bolus sample is placed on the lower plate and is gently squeezed to ensure surface contact. The variations of storage (G') and loss (G'') moduli, with strain were determined at three different frequencies: ω = 10rad/s, ω = 1rad/s and ω = 0.1rad/s. The imposed strains ranged from 0.01% to 400% according to a logarithmic distribution and with 10 points per decade. Each measurement is duplicated and the sample is removed and replaced before each test. Modulii and complex viscosity η^* were determined in the linear viscoelastic regime (imposed strain γ= 0.5%) by applying a frequency sweep from 0.1rad/s to 100rad/s. The frequency distributions are also logarithmic with 10 points per decade and the measurements are duplicated. All these measurements were achieved on bolus with various water 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 bolus on total wb (EF, EFP), in total wet basis.

2.3. Flow properties and capillary rheometry measurements

Flow properties are determined with a capillary rheometer with pre-shearing, Rheoplast, that has been applied and described in detail by Della Valle, Vergnes & Lourdin (2007) and Nunez, Della Valle & Sandoval (2010), who determined the viscous properties of starchy materials and cereal foods, respectively, under extrusion conditions. In the following, we will only describe its main working steps. In the case of food bolus, we did not use the pre-shearing function, because the fragmentation has already been performed when preparing bolus with the blender.

All the tests are made at room temperature (T = 23°C). The measuring chamber before the capillary die is fed with the bolus by the vertical motion of an annular piston at a constant speed of 1mm/s. Then, the injection piston (diameter D_P = 16mm) is moved vertically at a constant speed of 0.5mm/s to fill the capillary die. Three capillary dies (diameter D = 1mm, entry angle = 90°) are used with different die ratios L/D (32, 16 and 8). After a 15 s relaxation time, pressure is measured for different decreasing speeds of the injection piston, each speed step for a time interval of about 15s, ranging from 2 mm/s to 0.002mm/s, which leads to apparent shear rate values 3000 ≥ $\dot{\gamma}_{app}$ ≥ 3 s^{-1}. At the end of this sequence (about 2 mn), pressure measurement is repeated at the two largest speed values, in order to check sample stability, and a pressure profile P ($\dot{\gamma}_{app}$) is obtained.

This procedure has been applied to every sample for the same range of water content values as for oscillatory shear measurements (2.2). 15 to 20g of bolus are needed to obtain a single pressure profile. Data treatment is performed according to usual procedure for capillary rheometry as described in detail by Della Valle et al. (2007) and Nunez et al. (2010). The wall stress τ_w is derived from pressure measurements, using Poiseuille law, after application of Bagley's corrections. The flow index *n* is determined and the real value of the shear rate $\dot{\gamma}_w$ is calculated after Rabinowitsch analysis. Finally, the shear viscosity η is defined as the ratio $\tau_w/\dot{\gamma}_w$. Using entrance effects from Bagley correction, the elongational viscosity η_E and the deformation rate $\dot{\varepsilon}$ may also be derived, according to the Cogswell's method developed for melts polymers (Cogswell, 1972).

Flow curves ($\tau_w$or η ($\dot{\gamma}_w$)) obtained for different moisture contents were fitted according to appropriate rheological model and then shifted (translated) into a master curve to determine the model parameters. This procedure, based on the time-temperature superposition principle, for the rheological properties of polymers, has been already adapted by Della Valle et al. (2007) to the time-plasticizer content superposition, in the case of plasticized starches. In this study, the shifting factors were derived using Python (Python Software Foundation) according to the procedure developed by Saboo *et al.* (2018).

2.4 Imaging

Fragmented boluses of sponge cakes and extruded foods were prepared in the same way as for rheological measurements and water was added to obtain the necessary dilution before inserting between glass blades, and observed on a binocular stereomicroscope Leica with retro-lighting (x 35 ; Leica Microsystems, Conrad Electronics, France). No specific staining was used.

3. Results and discussion

3.1. Viscoelastic properties

Within the linear domain (γ < 1%), harmonic measurements performed at small strain amplitude, led to similar bolus mechanical spectra (G'(ω), G'' (ω)) whatever the food considered and its water content (Fig.1) : G' and G'' were slightly increasing functions of pulsation (or frequency) and all bolus exhibited a storage modulus larger than the dissipative one (G' > G'') in the range of frequency tested, and tan δ remained nearly constant ($\approx$ 0.2). Clearly boluses behave more like solid than fluids. Such spectra are usually typical of a viscoelastic network, with the frequency window of the test framing a part of the viscoelastic plateau. However, in the case of these artificial bolus, no structural interpretation can be given for the entities involved in such a network, at this stage. In many cases, G''(ω) curves exhibited a shallow minimum within the frequency interval (0.1 < ω <1 rd/s). The frequency and the G' value corresponding to this G'' minimum can be taken conventionally as representative of the characteristics (frequency and modulus) of the viscoelastic plateau (Ferry,1980). However, the frequency of the minimum depended on the bolus, and in some cases, it was not really observed. Therefore, for all our samples, we took the G' value at 1 rad/s as an empirical measure of the viscoelastic plateau modulus. Fig.1 also showed that, for all food boluses, a regular decrease of both modulii was observed as water content increased, which, as expected, indicated that water acts as a lubricant. It also suggested that, for SC bolus, the enrichment in protein slightly increased the values of modulii, and reduced its sensitivity to water content. Conversely, pea based extruded snacks (EFP) led to boli with much larger moduli, about 10 times, than extruded flat bread (EF), but displayed similar water sensitivity.

The values of storage modulus at viscoelastic plateau G'(ω ≤1 rd/s) were negatively correlated to the water content (Fig.2). These variations can be described by the equation :

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

G'_0 represents the theoretical value of storage modulus for dry bolus ; parameter α_G. reflects the interaction of the food with water and can be envisioned as a water plasticization coefficient. The values of these parameters are reported in Table 2. Similar results could have been obtained by shifting and superimposing G' (and G'') curves for a given moisture content as performed by van der Sman and Mauer (2019), in the case of starch/sugar/polyol mixtures, and by Costanzo et al. (2019) for gluten / water / ethanol blends. Besides extending the time-temperature superimposition principle to time-solvent in the case of bioplymers, these works underlined the importance of intermolecular interactions, mainly H bonds. Such interactions could be captured by the ratio of sample temperature to glass transition temperature of the biopolymer, which is a function of its solvent content. Regarding food boluses, our results suggest that the protein enriched sponge cake (SCP) interacts the least with water, likely because of the hydrophobic nature of added pea isolates, whereas globulins interactions with glutenins might also be infered for the decrease of water sensitivity (Lambrecht, Deleu, Rombouts & Delcour, 2018). Conversely, extruded pea flour (EFP) shows larger interaction with water than flat bread (EF), likely because hemicellulosic compounds, contained in pea flour, have a strong water retention capacity that largely balances the hydrophobic influence of pea isolates observed for SCP (Kristiawan et al., 2018).

For all moisture contents and foods, the variations of G' and G'' moduli with the applied strain γ exhibited the same patterns (Fig. 3). From this graph, three main zones may be delineated:

first the linear viscoelastic regime ($\gamma \leq 1\%$), where modulii are constant, with G'> G'' like for a gel, then, a transient regime, where storage modulus decreases and crosses G'' curve, which can be considered as plastic region, and, finally, a non-linear regime where both modulii decrease, with G'' > G', indicating that bolus starts to flow. This behaviour is typical of the loss of structure and of the flow of the material under increased strain. Since it holds for all bolus samples, we can define the stress τ_c, at the crossing of G' and G'' as a characteristic parameter of this behaviour:

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

G^* being the value of the complex modulus and γ_c the strain value at intersection of G' and G'' curves (G'=G''). Another characteristic stress maybe defined, by the value obtained at the end of the linear viscoelastic domain, defined by the crossing of the horizontal tangent of G' in the linear domain with the tangent to G' curve during plastic flow (see Fig.3). It is found that these values are highly correlated to the values of τ_c ($r^2 > 0.9$, see Appendix 1), so, in the following, τ_c is considered to characterise the transition of bolus to flow.The values of τ_c are negatively correlated to the water content (Fig.4). These variations can be described by the equation :

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

τ_{c0} is the theoretical value of characteristic stress for the dry bolus; parameter $\alpha_{\tau c}$ reflects the interaction of the food with water, like a water plasticization coefficient. The values of these parameters are reported in Table 2. At first glance, they confirm the trends observed in the linear viscoelastic domain, when considering α_G values : protein enriched sponge cake (SCP) interacts the least with water, again due to the hydrophobicity of pea isolates, whereas extruded pea flour (EFP) shows larger interaction with water than flat bread (EF), which, again, may be attributed to the presence of hemicellulosic compounds in pea flour. However, for extruded products, the values of $\alpha_{\tau c}$ are larger than those of sponge cakes, and the variations of characteristic stress obtained for EF and EFP are close to each other (Fig.4). This result indicates that the stress needed to make the bolus flow is more influenced by water for extruded cereal foods, than for sponge cakes, whether they are enriched in plant proteins or not. Moreover, the huge value of initial stress τ_{c0}, found for extruded products (Table 2, τ_{c0}> 100 MPa), confirms that, overall, large amounts of water (WC ≥ 60%) are needed to elaborate a flowable bolus from these foods that contain a large amount of hydrophilic compounds like amorphous starch.

3.2 Flow properties

From the pressure profiles P ($\dot{\gamma}_{app}$) obtained for different capillary geometries (L/D ratio), and using Poiseuille relation, flow curves can be derived, representing the variations of the wall shear stress τ_w as a function of apparent shear rate or τ_w ($\dot{\gamma}_{app}$)) for food boluses at different amount of water WC (Fig.5). As expected, the larger WC, the lower the apparent viscosity, defined by the ratio $\tau_w/\dot{\gamma}_{app}$: flow curves show that viscosity decreases by a factor of 10 when WC is increased by 0.10 for SC foods, whereas the decrease is lower for extruded foods. Some curves display a dispersion of experimental points, especially at larger moisture content. Indeed, in these conditions, flow becomes less steady and measurement accuracy lower, given the low level of pressure reached in the capillary, and also possible slip-stick phenomena. However, all curves show a similar increasing trend, and some of them suggest the existence of a yield stress at lower shear rate (like for SC at WC= 0.28 and 0.36, for instance). Therefore, after having converted apparent shear rate values $\dot{\gamma}_{app}$ into wall shear rate $\dot{\gamma}_w$, using Rabinowitsch correction, all curves are fitted using the Herschel-Bulkley model,:

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

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

Although fitting is acceptable ($r^2 > 0.75$), considering the dispersion mentioned before, it is thought that determining coefficents τ_s, *K* and n for each WC value could increase the uncertainty. Moreover, the flow index does not change significantly for each product, whatever the value of WC. Therefore, for each product, a master curve is determined by adapting the time-temperature principle, to superpose flow curves, obtained from bolus at different water content. Hence, each flow curve is shifted to a reference curve, at a given WC value, by plotting the reduced shear stress (τ_w / *a*) as function of reduced shear rate ($\dot{\gamma}_w$ * *a*), *a* being the shift factor. This procedure is illustrated in Appendix 2. The Herschel-Bulkley coefficients of the reference curve are recalled in Table 3, for each product. The values of the shift factor *a* vary between 0.016 and 27.8 and they are directly correlated to the water content, all food products being considered together (Fig.6a). In a first approach, this correlation suggests that the influence of water on the viscous behavior of the bolus in shear flow is similar for the four different cereal foods. Secondly, by applying (eq.4) to (τ_w/*a*) [*a**$\dot{\gamma}_w$], this correlation can be used to calculate, for any WC value, the corrected values for the coefficents of the Herschel Bulkley model by :

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

The numerical values of τ_{sc}, K_c and n, for all values of WC and products tested, are reported in Appendix 3. Note that the value of flow index n did not vary with WC. Furthermore, using correlation from Fig.6a and eq. (5), flow curves for the boluses of the four products can be derived for any WC value. For instance, for WC=0.6, taken as a typical value in the interval [0.28, 0,8], flow curves representing the shear viscosity η as a function of shifted shear rate ($\dot{\gamma}_w$. *a*) can be drawn (Fig.6b). These curves show that for low shear rate values, the viscosity of the bolus is very close to each other, whereas for larger values ($\dot{\gamma}_w$. *a* > 10s^{-1}), the two extruded products (EF and EFP) lead to much larger viscosity than sponge cakes, whether they are enriched in plant protein or not. The increase of slope, observed at lower shear rate values, underlines the presence of the yield stress, and it is enhanced for SC and EFP. In line with the preceding results obtained for the viscoelastic properties, the variations of the Herschel-Bulkley coefficients τ_{sc} and K_c with WC can be represented (Fig.7) and fitted by the following relations :

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

from which the values of τ_{s0} , α_{τ} , K_{c0} and α_K are extracted and reported in Table 2. In addition, variations of consistency with water are very close to each other for the four products (Fig. 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). Regarding α_{τ} and α_K variations, the trends obtained here are different from those observed for the viscoelastic properties since standard products (SC and EF) display the lower α_{τ} and α_K values. Moreover, α_{τ} and α_K values are correlated. These results show that, during the flow, the products enriched with plant proteins interact more with water than their standard counterpart. In other words, the presence of proteins increase the bolus flow sensitivity to water, which suggests that flow could contribute to bolus destructuring by disrupting food particles or releasing protein aggregates.

The same procedure can be applied to determine the apparent elongational viscosity of boluses, by taking into account entrance pressure effects from capillary rheometry tests. Then, using the same time-water superposition principle, master curves can be determined, for instance at WC =0.6 (see Appendix 4). Indeed, very little differences can be observed between the four foods.

3.3 Bolus structure and rheological properties relationships

First, the values found for coefficients α were in the same numerical interval [5, 31], regardless the rheological property. These values are in the same interval as the values of coeficients of starch plasticization by water, found for starchy materials and cereal foods under extrusion conditions (Della Valle et al., 2007; Nunez et al., 2010). Clearly, this result suggests that starch /water interactions have a significant role in the rheological behavior of the bolus and its breakdown during FOP. However, these values stand for starch melts as encountered during extrusion, i.e. at high temperature and shear, where starch granules are broken and polymers released. Food boluses can hardly be compared to starch melts. By the way, Cox-Merz rule does not apply, unlike polymer melts, since complex viscosity always took values larger than those of steady shear viscosity (as illustrated in Appendix 5), which reflects that the structure of bolus changed during steady flow.

Moreover, boluses are made of particles, of typical size 1mm, which are not expected to fragment during flow, but rather to deform, as shown by the micrographs of diluted boluses (Fig.8). Sponge cake (SC) bolus presents a rather uniform appearance of a cohesive mass of blurred aggregated particles, whereas enriched protein sponge cake (SCP) is more heterogenous, with darker parts and bigger entities (> 1 mm), separated by water. Extruded flat bread bolus (EF) also presents a cohesive morphology, whereas extruded pea flour bolus (EFP) presents a clear picture of agglomerated and elongated particles (length > 1mm, width < 1mm). These images suggest that the foods are not destructured in the same way after fragmentation and hydration. So it is not surprising that their characteristic rheological properties are not fully correlated.

In spite of these visual differences, they may be all envisioned as concentrated supensions of deformable particles (Fig.9). At lower strain the suspension is jammed, because of particles swelling due to water absorption (Fig.9b), which gives the bolus a solid-like behavior, in the viscoelastic domain. At larger strain, hence larger stress ($\tau > \tau_c$ or τ_s), the suspension may still be partially jammed, because, in the case of standard foods, hydrophilic particles still swell (Fig.9c). Conversely, the particles of protein enriched foods, being less hydrophilic, swell less, water acts as a lubricant which favors bolus flow (Fig. 9d). This interpretation would explain why the values of coefficient α referring to flow properties (yield stress τ_s and consistency *K*) are larger for the protein enriched foods than for their standard counterpart. More experiments, for instance using measurements of water absorption or swelling index on such products, and even comparison with settling ratio (Boehm et al. 2019) could contribute to test this hypothesis. Whether this transition occurs at the value of characteristic stress τ_c or yield stress τ_s is still to be determined precisely , although both properties are correlated (Fig.10). The question is significant since both properties might be relied on for the control of swallowing.

Clearly, before extending these rheological measurements to assess the interactions of saliva with food, it is necessary to test the influence of other saliva components on bolus rheological properties. Recently, these properties were measured on bolus made by mixing same foods (sponge cake) with artificial saliva, containing salt and mucin, and no significant difference was detected when compared with bolus made with water (Gibouin et al., 2019b). The influence of the addition of α-amylase, another component of saliva, on bolus rheological properties is still questionable. There is a general agreement that salivary α-amylase plays an important part in destructuring cereal foods, like bread, during the gastric phase (Bonhorst & Singh, 2013; Pentikaïnen et al., 2014; Freitas, Feunteun, Panouille & Souchon, 2018). But the short residence time of the food in the mouth may explain the little influence of α-amylase on bread destructuring during FOP, assessed by viscosity measurements (Le Bleis, Chaunier, Montigaud & Della Valle, 2016).

Regarding FOP, another point that confirms the relevance of our measurements comes from some results obtained when characterizing real boluses. Results obtained by Loret et al. (2011) measuring yield stress on cereal flakes boluses collected before swallowing for 11 subjects, hence for different saliva contents, lead to a value of α of about 8, i.e. in the interval found in this study. For sponge cake boluses, the values of coefficient α found for apparent viscosity were also in the same interval (17 and 11 for standard and protein enriched sponge cake, respectively) (Assad-Bustillos et al., 2020). Furthermore, they ordered in the same way as the values of $\alpha_{\tau c}$ found in the present study (9.3 and 4.8, see Table 2). This result suggests that the coefficient $\alpha_{\tau c}$ contributes to assess food-saliva interaction and that characteristic stress τ_c is the property that matters to define bolus destructuring. Although more work is necessary to ascertain this trend, it is possible, and rather simple, to extend these rheological methods to other foods. In turn, this would help to design and test foods that would have a specific behavior during chewing.

4. Conclusion

Our results show that the rheological properties of artificial food cereal boluses can be determined in a wide range of strain and strain rates, using different methods, in the range of water content encountered during FOP. For the four foods tested, hydrated boluses exhibited same behavior. First, at low strain, in the linear viscoelastic domain, bolus had a gel like behavior characterized by the viscoelastic modulus G*. Then, destructuring occurs at larger strain ($\gamma \geq 0.1$), where a characteristic stress τ_c can be delineated from the intersection between curves of storage and dissipative modulii. During steady flow, bolus behavior was found to follow Herschel-Bulkley model from which yield stress τ_s and consistency K were derived; master curves could be determined to compare the viscous behavior of the four products. At larger shear rate ($\dot{\gamma}_w \geq 10$ s^{-1}), viscosity was found larger for extruded foods without any effect of protein content. The variations of these four rheological properties (G*, τ_c, τ_s and K) with water followed an exponential decay, from which it was possible to derive coefficients of interaction with water, for each property and product. Their changes with product composition and structure were interpreted by envisioning the bolus as a suspension of soft particles. The comparison of their values with those obtained for real boluses, and their variations with protein content, suggest that rheological mesurements are helpful to characterize the interaction of food with saliva.

Acknowledgements

This work was carried out with the financial support of the regional programme "Food for Tomorrow / Cap Aliment“, which is supported by the French Region Pays de la Loire and the European Regional

Development Fund (FEDER). Florence' stay at UPV received the support of COST Action (CA15118 - Mathematical and Computer Science Methods for Food Science and Industry).

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List of figures

Figure 1: Examples of mechanical spectra obtained for the artificial bolus in the linear viscoelastic domain (γ <0.5%) for (a) standard sponge cake (SC), (b) sponge cake enriched with plant proteins (SCP), (c) extruded flat bread (EF) and (d) extruded pea flour (EFP) for different levels of water content WC as indicated in the graphs, on a total wet basis (the darker the symbols, the larger the WC value).

Figure 2: Variation of storage modulus G' at viscoelastic plateau (ω = 1rad/s) as function of the water content of food bolus for SC (O, a), SCP (●, b), EF (□, c) and EFP (■, d). Straight lines feature fitting by exponential functions (r^2 > 0.96) with parameters given in Table 2.

Figure 3: Example of strain sweep test result (here for SCS bolus, WC=0.57). Dotted vertical lines delineate the three regimes of bolus behavior, viscoelastic, plastic and flow. The crossing point (G'= G'') allows defining the characteristic stress τ_c.

Figure 4: Variation of characteristic stress τ_c at modulus crossing (G'=G'') as function of the water content of food bolus for SC (O), SCP (●), EF (□) and EFP (■). Straight lines feature fitting by exponential functions (r^2 > 0.96) with parameters given in Table 2.

Figure 5: Flow curves of cereal food boluses obtained by capillary rheometry, for three different moisture contents WC (lowest O, medium ●, highest ●): SC (a), SCP (b), EF (c) and EFP (d). Dotted lines are best fit by power law functions (r^2 >0.75).

Figure 6: (a) Variations of the shift factor a applied to build a master curve form curves represented in Fig.5, with water content WC for SC (O), SCP (●), EF (□) and EFP (■), dotted line showing best fit a = 8.10^{-4} exp [12*WC] ($r^2 \approx 0.89$); and (b) flow curves derived from the Herschel-Bulkley model applied for artificial bolus at WC=0.6, of sponge cake (black lines, SC and SCP) and extruded bread (grey lines, EF and EFP), standard cereal foods (thin continuous, SC and EF) and enriched in legume protein (dotted thick, SCP and EFP).

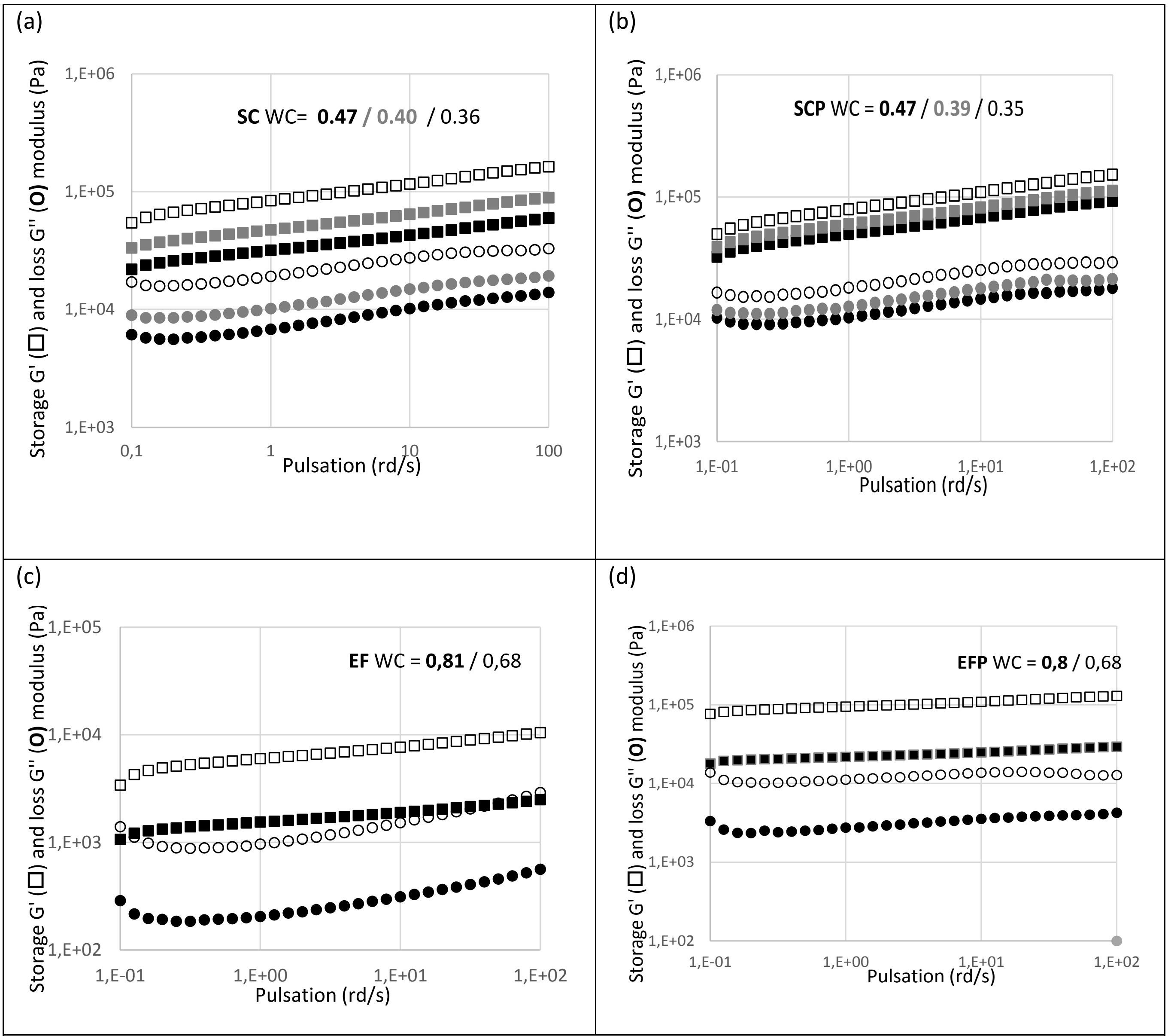
Figure 7: Variations of Herschel-Bulkley model's coefficients (defined in eq. (4) and (5)) with water content of food bolus for SC (O), SCP (●), EF (□) and EFP (■): yield stress τ_{sc} (a) and consistency K_c (b). Straight lines feature fitting by exponential functions (r^2 > 0.96) with

parameters given in Table 2, whereas thick dotted line (light grey) in (b) features the best fit obtained by taking all boluses together into account (K_c = 180 exp [-10.25 * WC] , r^2=0.9).

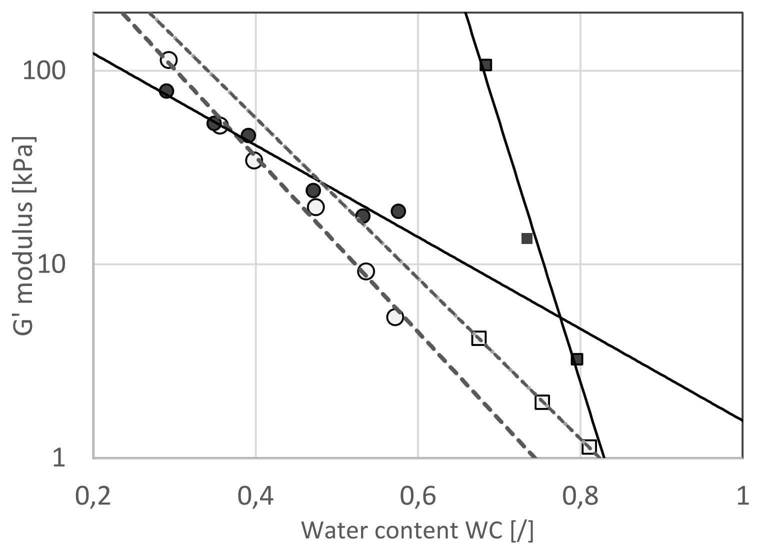
Figure 8: Micrographs of the artificial boluses of (a) standard sponge cake SC, (b) sponge cake enriched with proteins (SCP, white stain on bottom left is water between food particles), (c) extruded flat bread (EF) and (d) extruded pea flour EFP. Image width is 1cm. No specific staining was used, so differences of shade (light / dark) reflect the matter concentration, i.e. non-uniform thickness of the sample layer between blades, rather than specific composition or structural state.

Figure 9: Schematic representation of bolus structure, envisioned as a suspension of more or less swollen and deformable spherical particles: dry ground food (1mm) (a), wet particles in low strain domain (or $\tau < \tau_S$ or τ_c) (b); destructured suspension of particles of standard (c) and protein enrich foods (red spots symbolize proteic component (d)), at large strain ($\tau > \tau_S$ or τ_c).

Figure 10: Variations of the values of yield stress τ_S computed for the same WC as characteristic stress τ_c for the boluses of the four cereal foods SC (O), SCP (●), EF (☐) and EFP (■).



Gibouin et al. Fig.1 Rheology artificial bolus



Gibouin et al. Fig.2 Rheology artificial bolus

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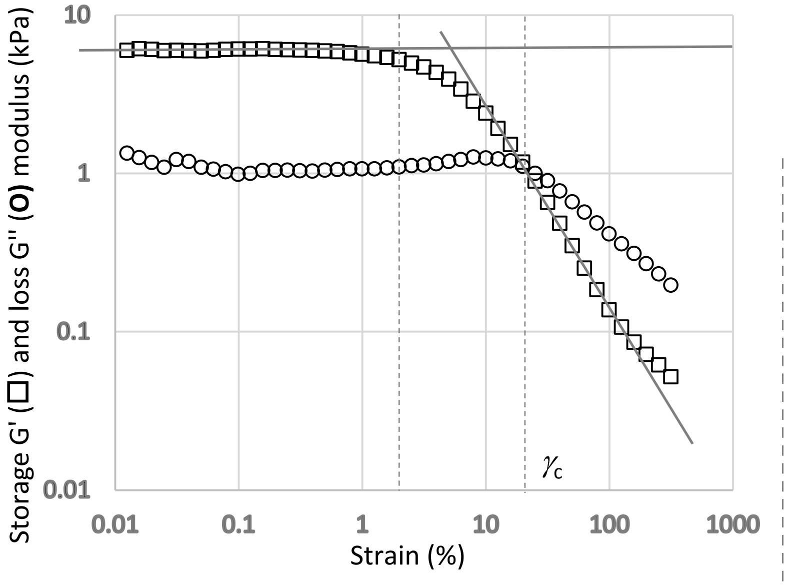
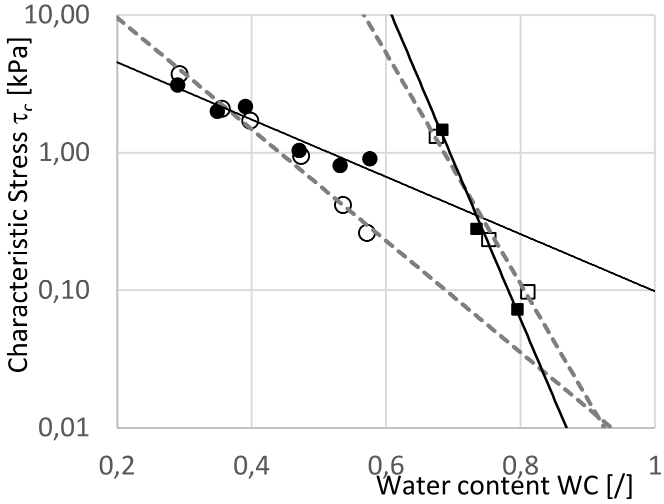
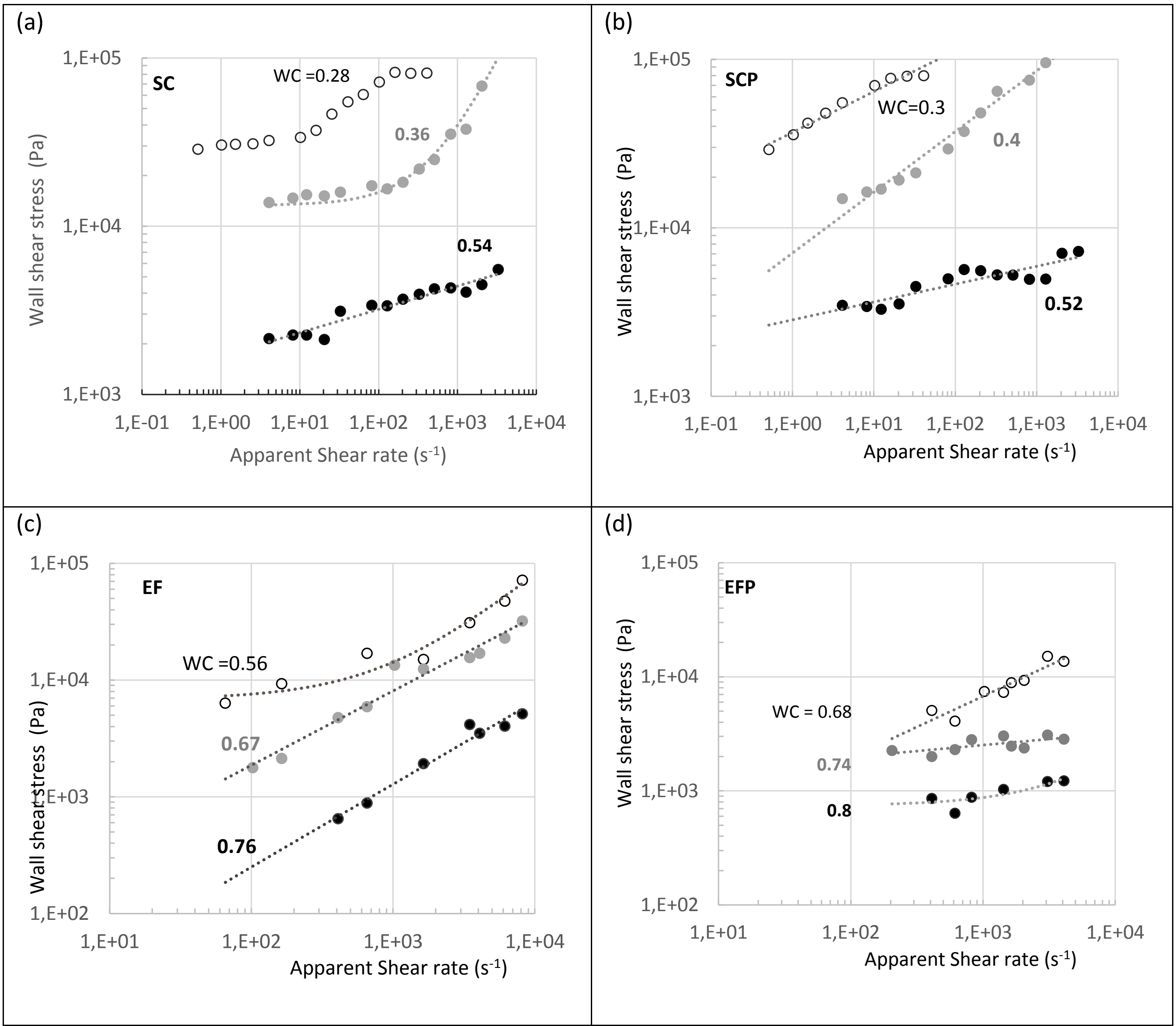


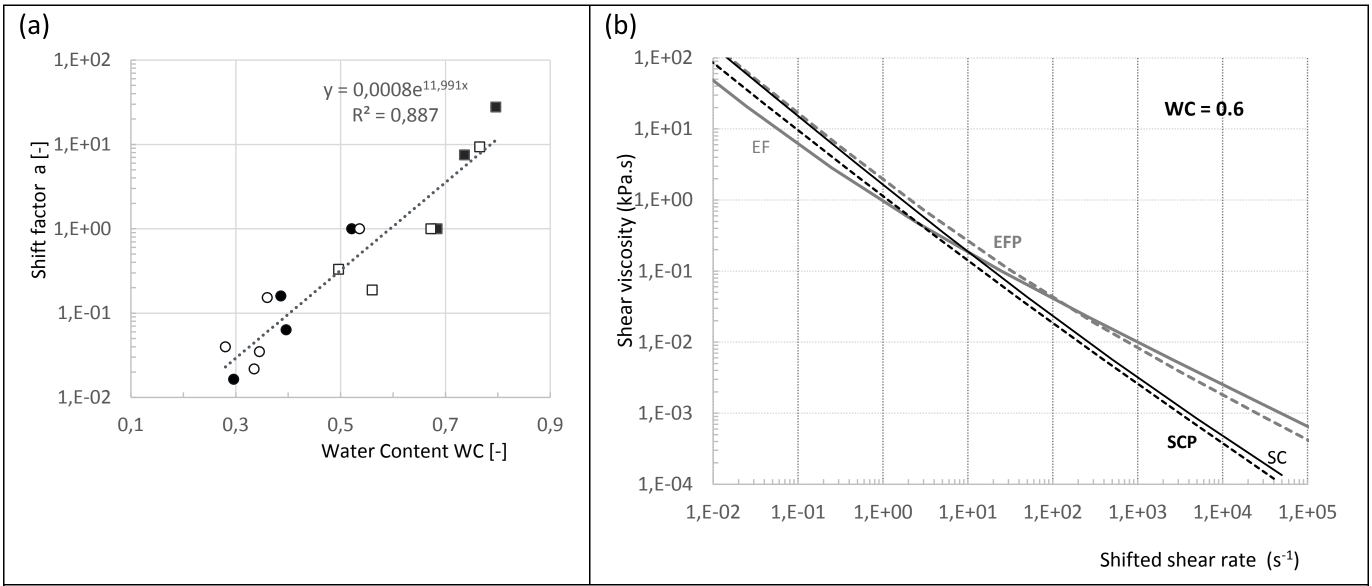
Gibouin et al. Fig.3 Rheology artificial bolus



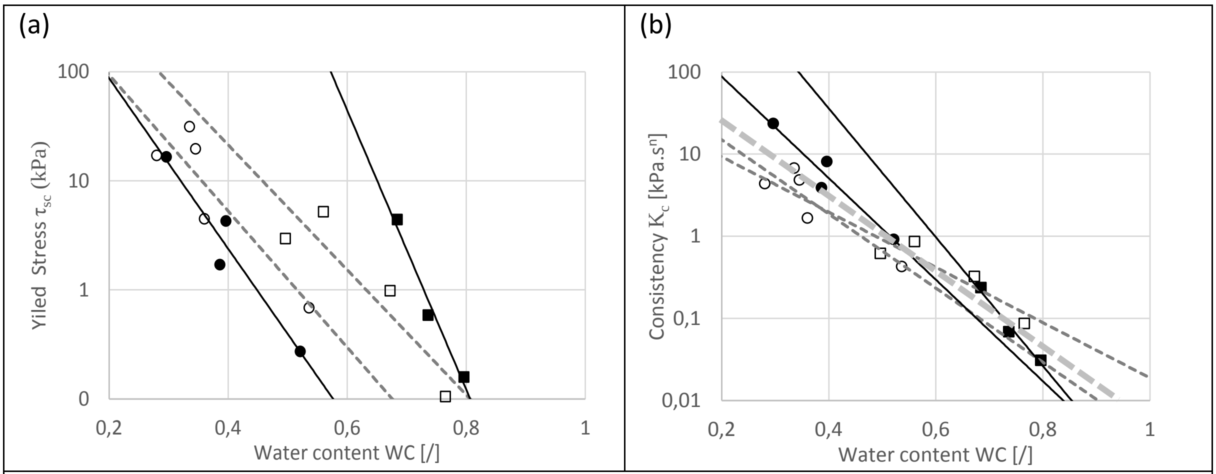
Gibouin et al. Fig.4 Rheology artificial bolus



Gibouin et al. Fig.5 Rheology artificial bolus



Gibouin et al. Fig.6 Rheology artificial bolus



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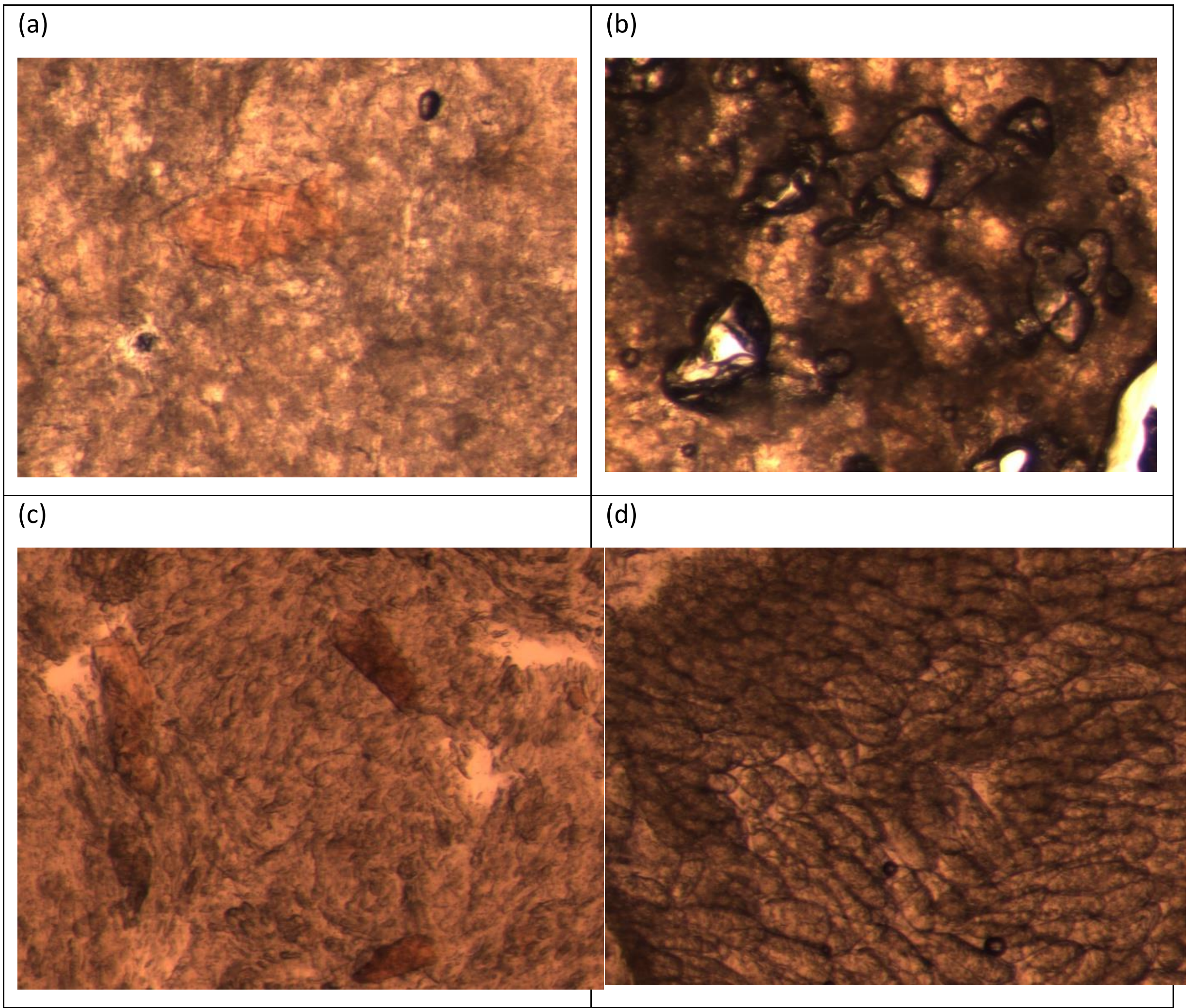
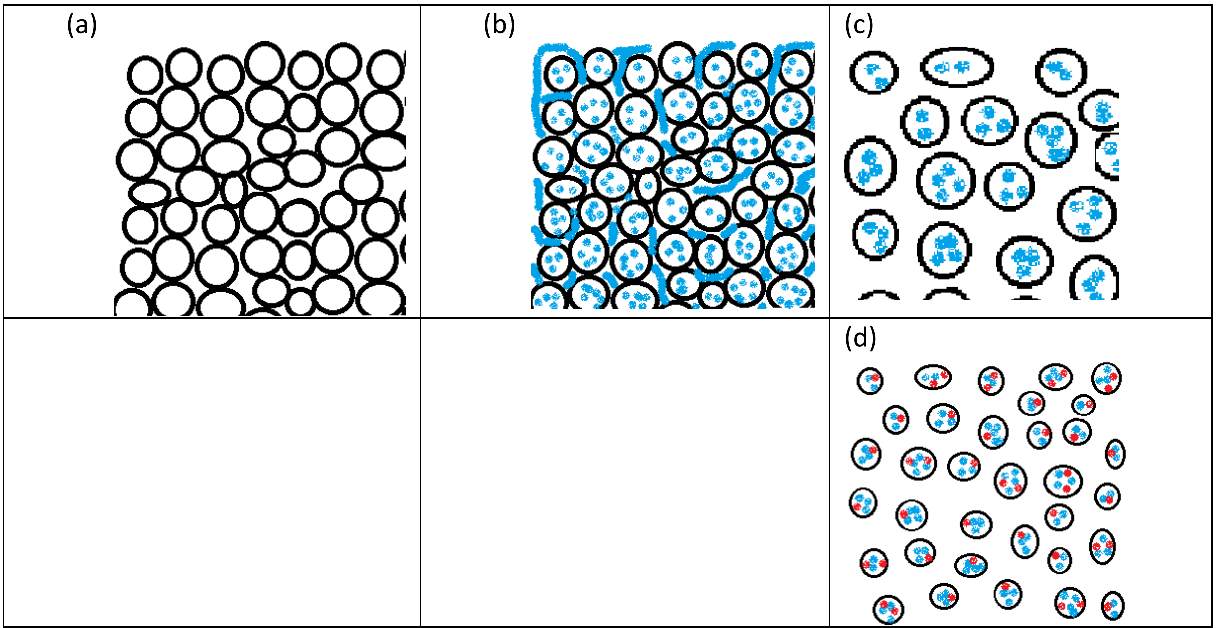
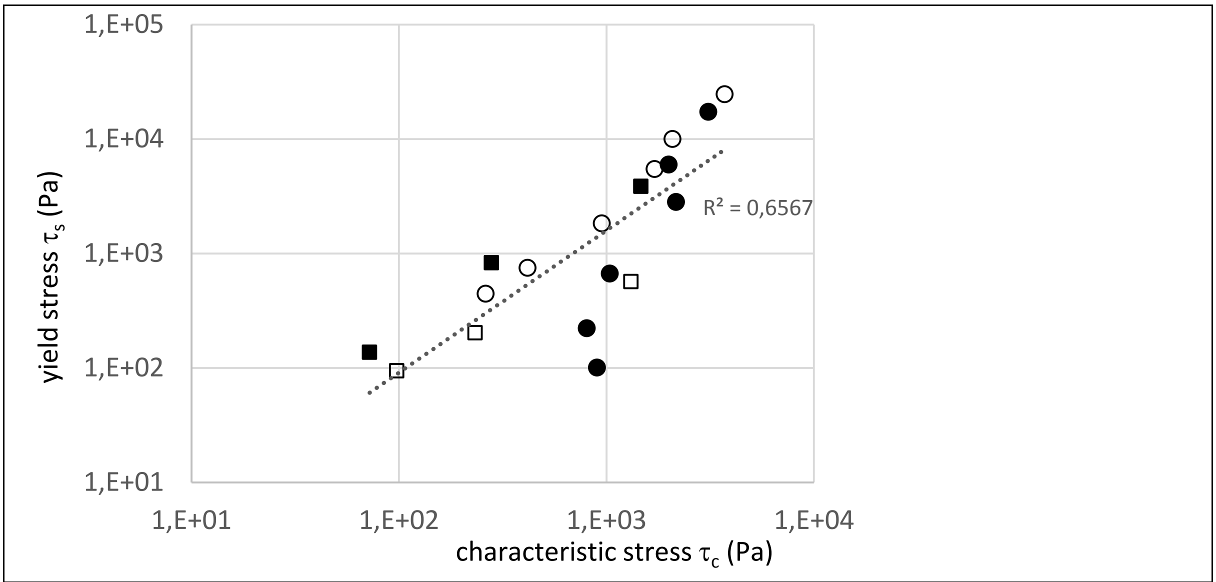


Gibouin et al. Fig.8 Rheology artificial bolus



Gibouin et al. Fig.9 Rheology artificial bolus



Gibouin et al. Fig.10 Rheology artificial bolus

Table 1: Composition (% total wet basis) and density of the four tested foods, sponge cake (SC) and 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

Table 2: Values of the parameters of the models representing the relations between bolus 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

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