



**HAL**  
open science

## Probing maltodextrins surface properties by atomic force microscopy: Interplay of glass transition and reconstitution properties

Regis Badin, Jennifer Burgain, Stephane Desobry, Bhesh Bhandari, Sangeeta Prakash, Claire Gaiani

### ► To cite this version:

Regis Badin, Jennifer Burgain, Stephane Desobry, Bhesh Bhandari, Sangeeta Prakash, et al.. Probing maltodextrins surface properties by atomic force microscopy: Interplay of glass transition and reconstitution properties. *Food Hydrocolloids*, 2022, 132, pp.107853. 10.1016/j.foodhyd.2022.107853 . hal-03880855

HAL Id: hal-03880855

<https://hal.univ-lorraine.fr/hal-03880855v1>

Submitted on 22 Jul 2024

**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 **PROBING MALTODEXTRINS SURFACE PROPERTIES BY**  
2 **ATOMIC FORCE MICROSCOPY: INTERPLAY OF GLASS**  
3 **TRANSITION AND RECONSTITUTION PROPERTIES**

4 Regis Badin<sup>a</sup>, Jennifer Burgain<sup>a</sup>, Stephane Desobry<sup>a</sup>, Bhesh Bhandari<sup>b</sup>, Sangeeta Prakash<sup>b</sup>,  
5 Claire Gaiani<sup>a,c</sup>

6 <sup>a</sup> Université de Lorraine, LIBio, Nancy, F-54000, France

7 <sup>b</sup> School of Agriculture and Food Sciences, The University of Queensland, Brisbane,  
8 Queensland 4072, Australia

9 <sup>c</sup> Institut Universitaire de France (IUF)

10 Corresponding author: [claire.gaiani@univ-lorraine.fr](mailto:claire.gaiani@univ-lorraine.fr)

11

12 **Highlights:**

- 13 • Glass transition significantly impacted maltodextrin reconstitution properties  
14 • Surface topographies revealed surface smoothing above glass transition  
15 • Surface nanoscale hollow formation was observed at the rubbery state  
16 • Young modulus decreased significantly at the rubbery state

17

18 **Abstract**

19 Reconstitution kinetics of three maltodextrins powders with different dextrose-equivalent  
20 (DE) values (low, intermediate, high DE) were evaluated under the glassy and rubbery states.  
21 Reconstitution times were found mostly shear rate of agitation in water and DE dependent in

22 the glassy state, with shorter reconstitution times for high shear rates and DE. For powders  
23 under the rubbery state, reconstitution times were less shear rate dependent, especially for  
24 high DE values. To have a better understanding of powder surface features that can impact  
25 reconstitution properties, single particles surface topographies and nanomechanical properties  
26 were probed by Atomic Force Microscopy (AFM). It was shown that surface roughness was  
27 higher for high DE maltodextrins under the glassy state, whereas a surface smoothing and  
28 slight hollows formation was observed for all powders under the rubbery state. Also, a  
29 significant reduction and uniformity of Young modulus at the particle surface were observed  
30 under the rubbery state. Finally, surface modification at microscale (smoothing and softening)  
31 was linked to powder behavior evolution at macroscale (relative shear rate independence  
32 during reconstitution) when the powder crosses the glass transition.

33

34 Keywords: Maltodextrin, Reconstitution, Glass transition, Powder, Surface topography,  
35 Nanoindentation.

36

37

## 38 **1. INTRODUCTION**

39 Powdered ingredients are involved in many manufacturing products and regularly used in  
40 the food industry, primarily for their ease of use, handling, transport and extended storage  
41 properties (Bhandari, 2013; Gaiani, Burgain, & Scher, 2013). Powders originate from the  
42 conversion of a liquid into a solid by various drying methods (such as spray-drying, freeze-  
43 drying, crystallization) or from size reduction processes (such as grinding, crushing, milling)  
44 (Bhandari, 2013; Islam & Langrish, 2013; Ratti, 2013; Woo & Bhandari, 2013). Because of  
45 these various origins and natures, powders physicochemical properties can be highly variable,  
46 thus influencing their technofunctional properties.

47 Reconstitution ability of dehydrated products in aqueous media is one of the most relevant  
48 functionalities to describe when one talk about food powders for industrial applications  
49 (Crowley, Kelly, Schuck, Jeantet, & O'Mahony, 2016; Gaiani et al., 2007a; Selomulya &  
50 Fang, 2013). For industrial applications, but also at the consumer level, quick and complete  
51 reconstitution properties are generally prerequisites (Forny, Marabi, & Palzer, 2011;  
52 Fournaise, Petit, & Gaiani, 2021). For example, coating or agglomeration processes are  
53 oftently used by industrials to improve powder reconstitution (Barringer, 2013; Cuq &  
54 Mandato, 2013; Gaiani, Schuck, Scher, Desobry, & Banon, 2007b). Powder reconstitution is a  
55 complex process that can be generally divided into four main steps including wetting, sinking,  
56 dispersion and solubilization, which may sometimes occur concurrently (Crowley et al., 2016;  
57 Fitzpatrick et al., 2016; Fitzpatrick, Ji, & Mao, 2021; Forny, et al., 2011; Gaiani et al., 2007a;  
58 Mitchell et al., 2014). Reconstitution conditions (e.g., shear rate, temperature, viscosity),  
59 particle size, shape, and porosity, highly influence reconstitution kinetics (Selomulya & Fang,  
60 2013; Crowley et al., 2016).

61 The glass transition is one of the most critical parameters to consider during powders  
62 processing and storage. At a critical temperature, molecular mobility increases when an

63 amorphous solid undergoes from a glassy to a rubbery state (Fitzpatrick et al., 2007; Roos &  
64 Drusch, 2016; Roudaut, Simatos, Champion, Contreras-Lopez, Le Meste, 2004; Yoshioka &  
65 Aso, 2007). This event is also highly dependent on relative humidity, as water plays the role  
66 of a plasticizer. An increase of water content in the food powder is highly responsible for a  
67 decrease in the glass transition temperature (T<sub>g</sub>) (Roos, 2002; 2006). This phenomenon  
68 consequently impacts powder properties. Particles become stickier because of increased  
69 surface energy, enhancing interactions with materials that are in contact with powder surface.  
70 Powders are also susceptible to forming strong cakes under the rubbery state (Fitzpatrick et  
71 al., 2007) and time-dependent crystallization phenomenon can occur (Wang & Truong, 2017).  
72 Glass transition is also accompanied by a significant decrease of the Young modulus at the  
73 particle surface (Palzer, 2007). Thus, particle surface elastic behavior drastically changes  
74 when the powder surface reaches the glass transition temperature.

75       Knowing that the powder surface is the first layer in contact with water during powder  
76 reconstitution, this later layer is expected to play a major role in this process. Powder surface  
77 is therefore highly dependent on environmental conditions (i.e., temperature and relative  
78 humidity). In the past decade, numerous powder surface analysis techniques have been used  
79 to further understand the role of the particle surface on the impairments of powder  
80 functionalities. For example, microscopy techniques such as Scanning Electron Microscopy,  
81 Confocal Laser Scanning Microscopy or even chemical composition techniques such as X-ray  
82 Photoelectron Spectroscopy are already widely used (Burgain et al., 2017; Murrieta-Pazos et  
83 al., 2012). Atomic Force Microscopy (AFM) is currently a rising star in the food powder  
84 surface analysis field, mainly due to its high-resolution capacity. AFM is such an unusual  
85 microscopy technique based on the use of a probe (mounted on a cantilever), which can  
86 reconstruct the surface topography by sweeping the sample surface. This system gives AFM  
87 its characteristic nanoscale sensitivity as the probe can record nanoscale height variation

88 thanks to a laser reflected towards the probe and detected by a photodiode (Burgain et al.,  
89 2017; Dufrene, 2002). AFM exhibits many advantages, such as nanoscale sensitive resolution  
90 capacity or the ability to analyze samples without pretreatment or vacuum conditions.  
91 Additionally, AFM is also a versatile tool compared to other surface analysis techniques, as it  
92 allows to estimate surface roughness. For example, Burgain et al. (2016a) were able to better  
93 understand surface modifications during high-temperature storage of whey protein isolate  
94 powders. They studied surface roughness and highlighted the formation of hollow structures  
95 related to improper storage conditions. Murrieta-Pazos et al. (2011) also used AFM to  
96 investigate the particle structure and roughness of skim and whole milk powders evolution  
97 during water adsorption. In addition to the topographical information, one of the other major  
98 assets of this technique is its ability to study nanomechanical properties due to  
99 nanoindentation experiments (Butt, Cappella, & Kappl, 2005; Gaboriaud & Dufrene 2007).  
100 This technique allows the detection of very localized variations of surface elasticity, as data  
101 are then useful for building highly resolutive elasticity maps. Despite all these advantages,  
102 powder nanoindentation experiments by AFM are mainly developed in the pharmaceutical  
103 field to relate particle hardness to powder tableting and compaction (Wu, Li, & Mansour,  
104 2010), to characterize mechanical properties of excipients particles (Liao & Wiedmann, 2003;  
105 Masterson & Cao, 2008; Perkins et al., 2007; Ramos & Bahr, 2007) or to study particle-  
106 particle interactions in inhaled powders formulations (Arora et al., 2016; Berard et al., 2002;  
107 Traini, Young, Rogueda, & Price, 2006; Weiss, McLoughlin, & Cathcart, 2015; Young,  
108 Tobyn, Price, Buttrum, & Dey, 2006). On the contrary, regarding the application in the food  
109 powder field, nanoindentation experiments are scarcely used (Burgain, Scher, Petit, Francius,  
110 & Gaiani, 2016b; Haider et al., 2013; Prime, Stapley, Rielly, Jones, & Leaper, 2011a; Prime  
111 et al., 2011b). Knowing that AFM brings at the same time topographical and mechanical

112 information of powder surface at the particle scale, this technique could be relevant to obtain  
113 a deeper understanding of powders functionalities, such as reconstitution.

114 Carbohydrate-based powders, especially maltodextrins, can be found in many products and  
115 are widely used in the food industry as thickener agents, for flavour enhancement, film-  
116 forming properties, encapsulating agents or even as excipients in pharmaceutical products  
117 (Castro, Durrieu, Raynaud, & Rouilly, 2016; Descamps, Palzer, Roos, & Fitzpatrick, 2013;  
118 Li, Pan, Ma, Miao, & Ji, 2020). Maltodextrins are polymer chains produced by the hydrolysis  
119 of starch. They are generally classified by their polymer chain length, estimated by the  
120 Dextrose Equivalent (DE) value (Avaltroni, Bouquerand, & Normand, 2004; Chronakis,  
121 1998). The different DE values are related to the hydrolysis degree and show very distinct  
122 functional properties. Low DE maltodextrins exhibit high viscosity and high T<sub>g</sub>, whereas high  
123 DE maltodextrins exhibit low viscosity and low T<sub>g</sub> (Castro et al., 2016; Rong, Sillick, &  
124 Gregson, 2009; Siemons, Politiek, Boom, van der Sman, & Schutyser, 2020). This makes the  
125 choice of the DE of the maltodextrin matrix crucial. For example, mixtures with added  
126 maltodextrins are commonly used on food powder to master the glass transition temperature  
127 (Fongin, Kawai, Harnkarnsujarit, & Hagura, 2017; Frascareli, Silva, Tonon, & Hubinger,  
128 2012; Shrestha, Ua-arak, Adhikari, Howes, & Bhandari, 2007). Moreover, maltodextrins are  
129 highly susceptible to caking phenomena, and their reconstitution properties are highly  
130 dependent on the DE value. Indeed, high DE maltodextrins reconstitute faster (Dupas, Girard,  
131 & Forny, 2017; Descamps et al., 2013).

132 Bringing new information about maltodextrins surface properties and the interplay between  
133 physical state and reconstitution properties is relevant to optimize powder uses in the food  
134 industry. The present study aims to provide a deeper understanding of maltodextrins  
135 reconstitution properties, depending on their physical state and molecular chain length (or  
136 DE), while bringing new insights about particle surface properties using AFM.

## 137 **2. MATERIAL AND METHODS**

### 138 **2.1. Material**

139 Maltodextrin powders with three different DE values were employed in the present study.  
140 Higher DE values correspond to more enzymatically hydrolyzed products than those with  
141 lower DE values. Spray-dried maize maltodextrins with DE 4-7 (i.e., low DE) and 16.5-19  
142 (i.e., high DE) were purchased from Sigma-Aldrich (Saint-Louis, Missouri, USA) and those  
143 with DE 12 (i.e., intermediate DE) were purchased from Roquette Frères (Lestrem, France).  
144 Maltodextrins were chosen to cover a range of DE between 4 and 19. DE values higher than  
145 20 are considered to be dextrans or glucose syrups (Chronakis, 1998) which usually are in a  
146 liquid state.

### 147 **2.2. Powder physical properties**

#### 148 **2.2.1. Glass transition temperature ( $T_g$ )**

149 Differential Scanning Calorimetry (DSC) was used to determine the  $T_g$ . DSC was  
150 calibrated using Indium metal as a standard. For the calibration, 5 repetitions were made, each  
151 with 3 sequent heating segments and with a heating rate of 20 °C/min. A first heating ramp  
152 from 20 to 210 °C was performed, followed by a cooling to 50 °C. Then, a second heating  
153 ramp from 50 to 210 °C and another cooling to 50 °C was also done. A third and final heating  
154 ramp to 210 °C was also performed. Finally, to ensure good DSC calibration, onset melting  
155 point was determined on the second and third heating curves. This later should be measured at  
156 156.6 °C within a range of 0.2 °C.

157 Powders samples were first equilibrated at different water activities ( $a_w$ ) thanks to a  
158 climatic chamber (Climacell, MMM Medcenter, Fisher Bioblock Scientific, Illkirch, France),  
159 which allowed to control relative humidity (RH) and temperature. For this purpose, powders  
160 were deposited under a thin layer on petri dishes to allow RH and temperature equilibrium.



161 When the equilibrium relative humidity (ERH) is reached,  $a_w$  is equal to the ERH divided by  
162 100 (i.e.  $a_w = ERH/100$ ). Thus,  $a_w$  can be adjusted by controlling the relative humidity  
163 (Kong & Singh, 2016). In order to reach very low  $a_w$ , phosphorus pentoxide ( $P_2O_5$ ) was  
164 employed for equilibrium which is known to be a desiccant salt (0% RH).

165 Before DSC experiments (DSC2500 discovery series from TA Instruments, Hüllhorst,  
166 Germany),  $a_w$  was measured for each sample thanks to a HygroPalm23- $a_w$  portable device  
167 (Rotronic, France) at room temperature ( $22 \pm 2$  °C).

168 Equilibrated powders (10 – 15 mg) were hermetically sealed in aluminium Tzero pans (TA  
169 Instruments, Hüllhorst, Germany). An empty pan was used as a reference and nitrogen as a  
170 carrier gas. Samples were first equilibrated for 1 min at - 60 °C. This step was followed by a  
171 first heating ramp up to 180 °C (10 °C/min) in order to erase the thermal history of the  
172 sample. Pans were then equilibrated for 1 min at 180 °C and cooled to the initial temperature  
173 of - 60 °C (10 °C/min). A second heating ramp was finally performed up to 180 °C (10  
174 °C/min).

175  $T_g$  was determined thanks to the heat flow change on the second heating ramp with the  
176 TRIOS analysis software (TA Instruments, Hüllhorst, Germany). The half slope height  
177 method was used to calculate the midpoint  $T_g$ . The onset and endset point of the glass  
178 transition events were also determined. All measurements were performed in triplicate and  $T_g$   
179 values were averaged.

### 180 **2.2.2. Particle size distribution**

181 The particle size distribution was analyzed using a Mastersizer MS3000 (Malvern  
182 Instruments, UK) with Aero S air unit which allows powder dispersion. In order to obtain a  
183 good laser obscuration, analytical conditions were defined with the following parameters: air  
184 pressure at 2 bars, hopper length of 2 mm, powder feed rate between 20 - 30%. Finally,

185 classical descriptors were used to estimate particle size distributions ( $D_{10}$ ,  $D_{50}$ ,  $D_{90}$ ). These  
186 parameters represent the diameters for which 10%, 50% or 90% have lower sizes. The Span  
187 was also used to estimate the width of the particle size distribution and is calculated thanks to  
188 **Equation 1**. Experiments were performed in triplicate.

$$189 \quad \text{Span} = \frac{D_{90} - D_{10}}{D_{50}} \quad \text{eq (1)}$$

### 190 **2.2.3. Numeric microscopy**

191 Maltodextrin particles were observed thanks to a Dinolite AM73915MZT numeric  
192 microscope. A small powder quantity was gently dispersed on a petri dish and was then  
193 equilibrated under a range of RH within 30 to 70% and temperature under a range of 20 to 80  
194 °C, thanks to a climatic chamber (Climacell, MMM Medcenter, Fisher Bioblock Scientific,  
195 Illkirch, France) as described in 2.2.1. Particles morphology was then characterized thanks to  
196 microscopy for all conditions and classified as individual particles, caked particles or liquid  
197 state. For each maltodextrin powder, 20 conditions were tested in the range 0.3 – 0.7 water  
198 activity vs. 20 – 80 °C temperature. For each condition, at least 10 pictures were taken.

### 199 **2.3. Single particle surface characterization by Atomic Force Microscopy (AFM)**

200 All AFM measurements were performed in air and at  $22 \pm 2$  °C using a Flex-Axiom  
201 (Nanosurf, Liestal, Switzerland) with the Nanosurf C3000 controller software.

#### 202 **2.3.1. Sample preparation and single particle selection**

203 Maltodextrins powders were fixed on a circular cover made of borosilicate glass (12 mm  
204 radius, 0.13 - 0.17 mm thickness). Powders were gently dispersed on a thin layer of epoxy  
205 glue spread on the glass. The samples were left overnight under controlled temperature and  
206 RH conditions (as described in 2.2.1.) in order to be in a glassy or rubbery state for AFM

207 experiments. Single particles were selected for analysis using a micrometer-scale camera  
208 mounted on the AFM device.

### 209 **2.3.2. Surface topography and roughness analysis**

210 Surface topographies were acquired in air thanks to contact mode. The cantilevers used to  
211 image the powder surface (Stat0.2LauD cantilevers from Nanosurf, Liestal, Switzerland) have  
212 a theoretical spring constant ( $k_c$ ) of about 0.2 N/m. The scan rate was set at 0.8 s/line.  
213 Scanned areas were of  $10\ \mu\text{m} \times 10\ \mu\text{m}$ ;  $5\ \mu\text{m} \times 5\ \mu\text{m}$  or  $1\ \mu\text{m} \times 1\ \mu\text{m}$  with a resolution of 256  
214 points  $\times$  256 lines. A PID (Proportionnal, Integrative, Derivative) retroaction loop allows to  
215 keep constant the cantilever oscillation amplitude. The average surface roughness ( $Sa$ ) was  
216 estimated after levelling AFM images thanks to a second order polynomial filter (Eaton &  
217 West, 2010), and was calculated on scanned areas thanks to the **equation 2**.

$$218 \quad Sa = \frac{1}{n} \sum_{i=1}^n |y_i| \quad \text{eq (2)}$$

219 where  $y$  is the height ( $z$ ) measured by the piezoelectric scanner at a given pixel ( $i$ ) in the  
220 image. At least 3 images were analyzed to be able to average the surface roughness value.

221 Finally, height and roughness profiles for a line were extracted from raw data to be able to  
222 highlight slight or localized variations on surface roughness (C3000 software, Nanosurf,  
223 Liestal, Switzerland).

### 224 **2.3.3. Mapping nanomechanical properties: Young modulus (E)**

225 AFM nanomechanical properties experiments were performed thanks to cantilevers  
226 (DYN190Al-10 cantilevers, Nanosurf, Liestal, Switzerland) with a radius curvature lower  
227 than 10 nm. The first step consisted of AFM device calibration before any measurement.

- 228 • Spring constant was calculated thanks to the thermal calibration method (Levy &  
229 Maaloum, 2002) and was estimated to be about 20-30 N/m.

230 • Deflection sensitivity also needed to be calibrated with a sample (i.e, glass or silicon)  
231 harder than the stiffness of cantilever. This calibration consisted of press and retract  
232 movements of the tip on the stiff surface to record five force-distance curves in order  
233 to link the cantilever deflection, measured by photodiodes, to the z-axis movement of  
234 the scanner. Thus, the displacement gradient of the z-axis piezoelectric scanner related  
235 to cantilever deflection allows itself to convert the voltage measured by photodiodes  
236 into a reference deflection in nanometer per volts. Generally, this deflection sensitivity  
237 was about 80 nm/V and was considered acceptable when the deviation was under 5%  
238 (Clifford & Seah, 2005).

239 • Finally, deflection crosstalk compensation was also calibrated.

240 To investigate the surface Young modulus of single particles, 1024 force-distance curves  
241 were recorded on a grid of 32×32 pixels on a 10 μm ×10 μm area. AFM tip modulation time  
242 was set at 250 ms and distance range at 1 μm. Thanks to the previously calculated spring  
243 constant, the cantilever deflection can be converted into a force thanks to the Hooke's law  
244 (**Equation 3**).

$$245 \quad F = -k_c \times d \quad \text{eq (3)}$$

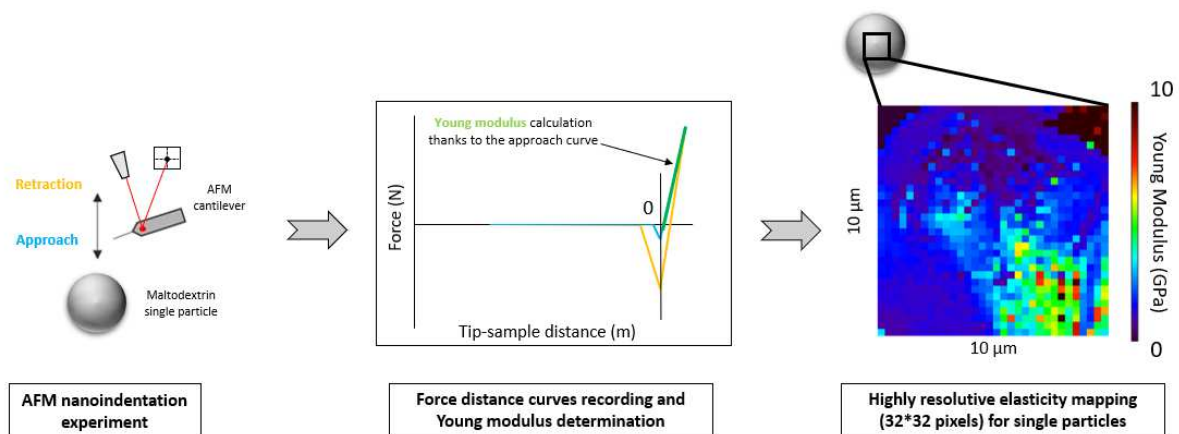
246 Where F is the Force (N),  $k_c$  the cantilever spring constant (N/m) and d the distance (nm).  
247 Thus, maximal load at the particle surface was set at 50 nN. All force-distance curves were  
248 individually analyzed thanks to the ANA Software (Nanosurf, Liestal, Switzerland). Young  
249 modulus (E), also called elastic modulus, was thus analyzed on approach curves using the  
250 Hertz Theory for elastic media using conical indenters (Calabri, Pugno, Menozzi, & Valeri,  
251 2008; Cardenas-Perez et al., 2019; Clifford & Seah, 2005; Hertz, 1882) (**Equation 4**).

$$252 \quad F = \frac{2E \tan \alpha}{\pi (1-\nu^2)} \delta^2 \quad \text{eq (4)}$$

253 Where  $F$  is the force applied (N),  $E$  the Young modulus (Pa),  $\nu$  the Poisson constant,  $\delta$  the  
 254 indentation depth (m) and  $\alpha$  the top angle of the tip. Poisson ratio of polymers varies between  
 255 0.3 and 0.5. A variation of the Poisson ratio of 0.1 will result in a maximal variation error of  
 256 the Young modulus value of less than <10% (Dokukin & Sokolov, 2012). Thus, Poisson ratio  
 257 was set at 0.5, which is the value generally used for AFM indentation on biological samples  
 258 (Shojaei et al., 2013; Guz, Dokukin, Kalaparathi, Sokolov et al., 2014).

259 Finally, the analysis of the 1024 approach curves and the Young modulus calculation allow  
 260 to build highly resolutive elasticity maps. The whole experimental process is summarized in  
 261 **Figure 1**. At least three elasticity maps were done on independent particles in order to ensure  
 262 experiment repeatability.

263



264

265 **Figure 1.** Experimental steps followed to build highly resolutive Young modulus mapping of powder particle  
 266 surface by AFM

267 **2.4. Powder reconstitution properties**

268 In order to identify the impact of the material (i.e., glass transition phenomenon and DE  
 269 value) and the stirring rate, the reconstitution ability was measured thanks to a conductimeter  
 270 (SevenCompact S230, Mettler Toledo, Greifensee, Switzerland) calibrated with a KCl

271 solution (at 1413  $\mu\text{S}/\text{cm}$ ). The setup was made up of an agitated and instrumented vessel (250  
272 ml beaker with a magnetic stirrer of  $6 \times 20$  mm), in which the temperature was controlled  
273 and maintained at 25 °C thanks to a thermostated water bath Eco Gold (Lauda, Germany).  
274 The conductivity of the solution was followed during the whole reconstitution. This  
275 experimental setup was chosen to be in adequation with the standard geometric ratio  
276 established by Rushton, Costich, & Everett (1950) and was inspired by Mitchell et al., (2014)  
277 and Fournaise, et al., (2021).

278 Five grams of maltodextrin powder for a final volume of one hundred milliliter were  
279 weighed (5% w/v). Shear rate was set at 500, 750, 1 000, 1 250 and 1 500 rpm. Finally, the  
280 powder was poured all in once in the instrumented vessel after 10 s and the conductivity was  
281 measured every second during the whole reconstitution, allowing to plot reconstitution kinetic  
282 curves. Reconstitution was considered as over when the solution reached 90% of its maximal  
283 conductivity.

284 During powder reconstitution, conductivity evolves from an initial value of 0  $\mu\text{S}/\text{cm}$  to a  
285 final and maximal value which depends on the powder nature. Thus, conductivity values were  
286 normalized according to **Equation 5**. (Fournaise et al., 2021) (Gaudel et al., 2022).

$$287 \quad c(t) = \frac{\kappa(t) - k_{ini}}{k_{end} - k_{ini}} \quad \text{eq (5)}$$

288 Where  $c(t)$  is the normalized conductivity,  $\kappa(t)$  the conductivity at a time  $t$ ,  $k_{ini}$  the initial  
289 distilled water conductivity and  $k_{end}$  the final conductivity. Finally, reconstitution kinetics  
290 curves were plot thanks to SigmaPlot (Systat Software Inc, San Jose, California, USA).

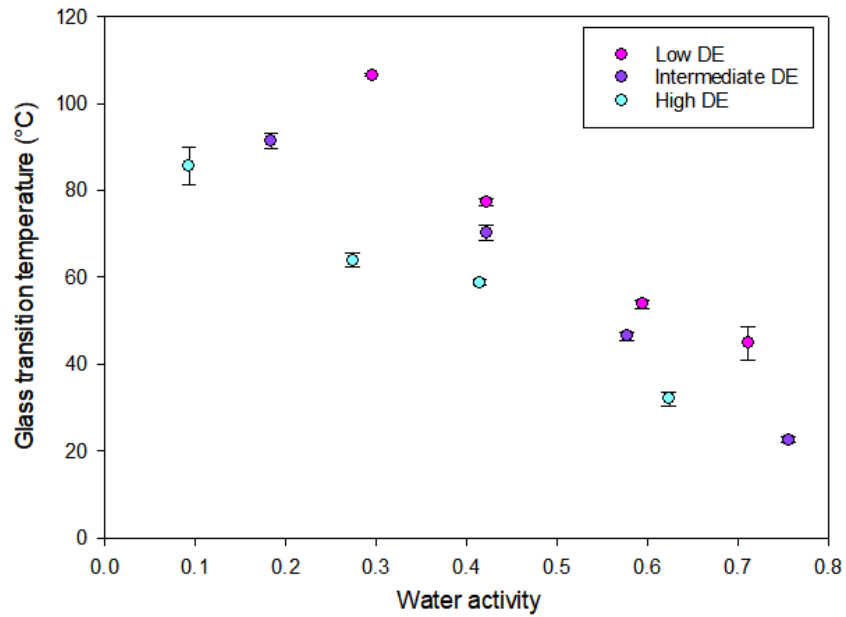
## 291 2.5. Statistical analyses

292 Statistical analyses were carried out using the OriginPro Software (OriginLab,  
293 Northampton, United States). Data were analyzed using a one-way ANOVA and means were  
294 compared thanks to the Tukey-HSD test at a level of significance of  $p < 0.05$ .

### 295 **3. RESULTS AND DISCUSSION**

#### 296 **3.1. Maltodextrin glass transition**

297 For the three maltodextrins, onset Tg values were determined on DSC curves at different  
298 beforehand equilibrated  $a_w$  (**Appendix A**). **Figure 2** shows that  $a_w$  and DE values are two  
299 important factors influencing the glass transition for maltodextrin powders. Tg values,  
300 whatever the DE, tended to decrease with increased water activity. This phenomenon was  
301 attributed to the plasticizing effect of water, which can be modelled using the Gordon-Taylor  
302 equation (Roos, 2002; Palzer, 2007). Water increases the molecular mobility of the polymer  
303 chains by creating hydrogen bonds. This plasticization phenomenon thus expands the free  
304 volume, allowing the overall molecular mobility, leading to a decrease of Tg (Castro et al.,  
305 2016; White & Lipson, 2016). Also, a clear tendency of Tg appears related to the DE values.  
306 For a given  $a_w$ , the Tg value decreases when the DE increases, meaning that maltodextrin  
307 molecular weight is directly correlated to the Tg. Maltodextrins with higher DE values exhibit  
308 a lower molecular weight distribution and a lower polymer chain length which directly  
309 impacts their physical behavior (Avaltroni et al., 2004; Rong et al., 2009; Siemons et al.,  
310 2020). Moreover, heating at high temperatures during the DSC measurements did not involve  
311 other thermal events on maltodextrins. Castro et al. 2016 also studied the thermal stability of  
312 maltodextrins on a large DE value range. A first stage of dehydration was observed within the  
313 range of 25 to 150 °C and an onset decomposition temperature around 260 °C for all  
314 maltodextrins, meaning that maltodextrins can withstand temperature up to 250 °C.



315

316 **Figure 2.** Glass transition onset temperature ( $T_g$ ) for different maltodextrins depending on their water activity

317

( $a_w$ ) represented by the midpoint value

318

However, when the material undergoes glass transition with  $a_w$  and/or temperature

319

increase, powders physical properties are impacted. Indeed, maltodextrin particles can be

320

observed as individual or agglomerated particles. They can also be found at the liquid state

321

when the powder is equilibrated at high  $a_w$  values. A qualitative map (one for each DE value)

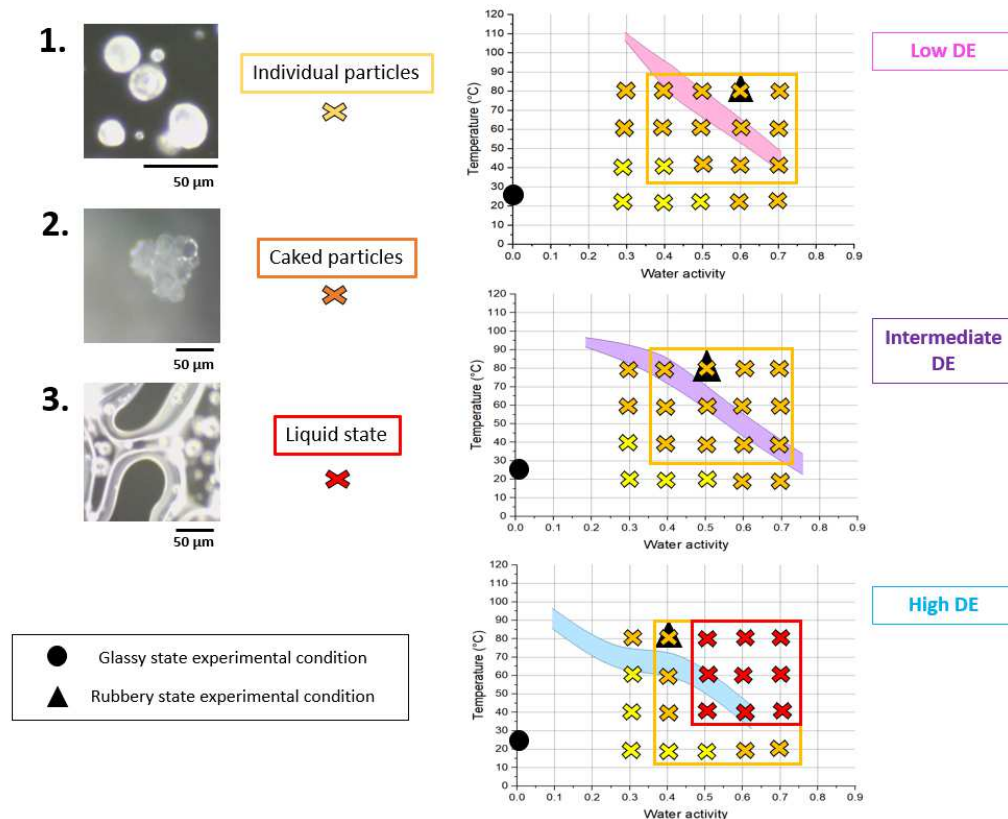
322

of these various material states is displayed in **Figure 3**, which shows particles morphology

323

between 0.3-0.7  $a_w$  and 20-80 °C.





324

325 **Figure 3.** Qualitative map of particle morphology (A. low DE; B. intermediate DE; C. high DE) under controlled  
 326 temperature and relative humidity conditions observed thanks to numeric microscopy (colored crosses, yellow  
 327 for individual particles, orange for caked particles and red for liquid state). Particle morphology was checked  
 328 under each temperature/ $a_w$  condition. The glass transition curve is represented by a colored area which is  
 329 comprised between the  $T_g$  onset and endset. Experimental conditions to set powders under the glassy or the  
 330 rubbery state for the rest of the experiments are identified in the qualitative map as black circle (glassy state) or  
 331 black triangle (rubbery state).

332 As it can be highlighted from **Figure 3**, powders are initially well-dispersed. With the  
 333 increase of  $a_w$ , a caking phenomenon is observed and is directly correlated to the glass  
 334 transition region. In addition to its plasticizing effect, the increase in water activity is  
 335 somehow correlated to the caking phenomenon, primarily because of the water sensitivity and  
 336 hygroscopicity. Thus, powders are susceptible to form strong undesired aggregates from the  
 337 onset of the glass transition, by the formation of liquid then solid bridges, as shown in the  
 338 study of Descamps et al., (2013) on caking behavior of high DE value maltodextrins. Also, at

339 very high  $a_w$ , high DE maltodextrins were found to be at the liquid state, due to the saturation  
340 of the powder with water.

341 Above glass transition temperature, molecular mobility increases critically (Roudaut et al.,  
342 2004; Yoshioka & Aso, 2007). Thus, a time-dependent phenomenon of crystallization of low  
343 molecular weight components may occur if the powder is under rubbery state conditions, as it  
344 is the case of lactose or fructose for example (Roos & Karel, 1992; Roos, 2002; Saavedra-  
345 Leos et al., 2014; Wang & Truong, 2017). As maltodextrins are relatively long polymer  
346 chains, depending on the DE, they are unlikely subject to crystallization. However, high DE  
347 maltodextrins exhibit shorter polymer chains and crystallization of maltose or dextrose only  
348 could be possible. Microstructure of different DE values was already detailed in the literature.  
349 Thanks to X-ray diffraction, it was shown that diffraction pattern of different DE  
350 maltodextrins, at a large range of  $a_w$ , shows an amorphous structuration, which remains for all  
351 DE and at all the water activities (Saavadra-Leos, Leyva-Porras, Araujo-Diaz, Toxqui-Teran  
352 & Borrás-Enriquez, 2015). However, to minimize issues encountered at the rubbery state,  
353 powder samples need to be stored the nearest possible to the endset region of  $T_g$ . As higher  
354 DE maltodextrins show lower  $T_g$  values, they need to be stored at the lower water activity  
355 possible under the rubbery state.

356 Powders were stored at the experimental conditions described in **Table 1** in order to be in a  
357 glassy (i.e.,  $< T_g$ ) or a rubbery state (i.e.,  $> T_g$ ) as presented on qualitative maps into **Figure 3**  
358 by a black circle or triangle respectively.

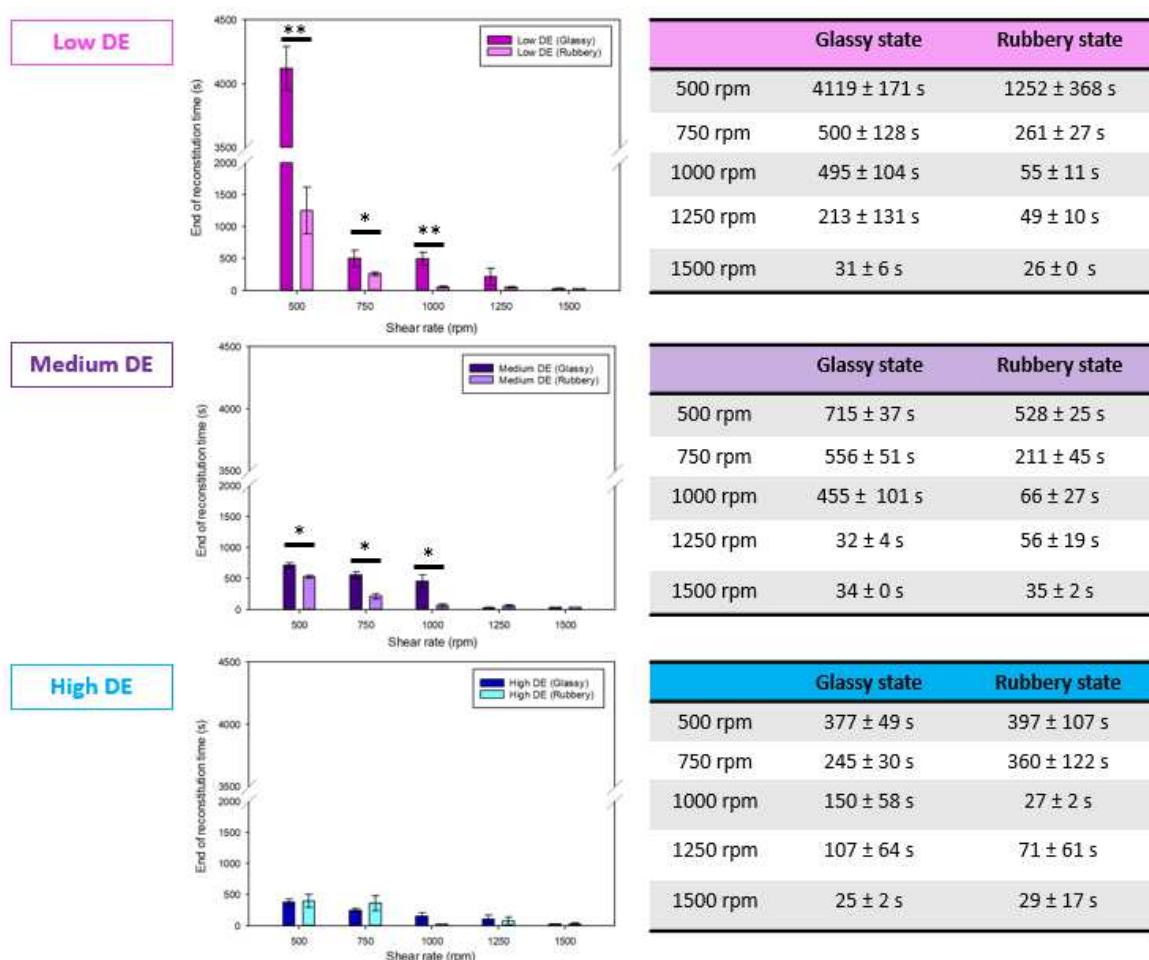
359 **Table 1.** Powder storage conditions to keep the powders under a glassy or a rubbery state.

	GLASSY STATE		RUBBERY STATE	
	RH (%)	Temperature (°C)	RH (%)	Temperature (°C)
low DE	0	20	60	80
intermediate DE	0	20	50	80
high DE	0	20	40	80

360

361 **3.2. Powders reconstitution ability**

362 The reconstitution time was investigated for maltodextrin stored under or above the T<sub>g</sub> by  
 363 following the reconstitution kinetics. Reconstitution times were evaluated by considering the  
 364 reconstitution as over when conductivity value reaches 90% of its maximal value. Examples  
 365 of reconstitution kinetic curves, before and after normalization are given in **Appendix B**.  
 366 **Figure 4** shows that reconstitution times seem to directly depend on the DE value, the  
 367 powders glassy/rubbery state and the shear rate.



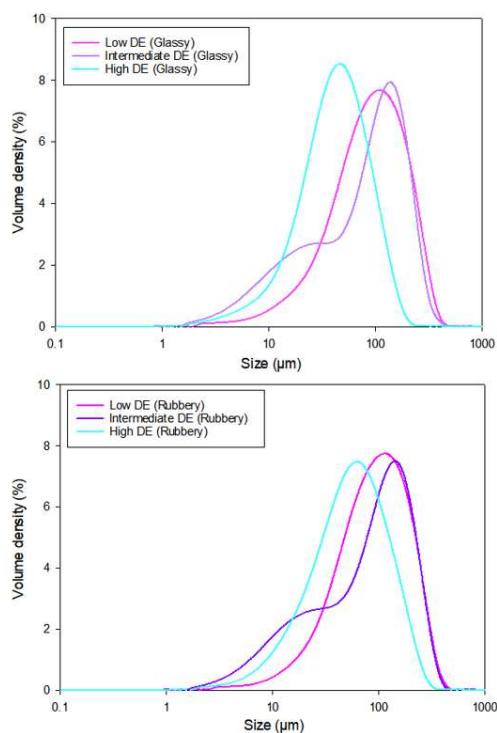
368  
 369 **Figure 4.** Reconstitution times of maltodextrins with different DE values, under glassy or rubbery states and at  
 370 different shear rates. Statistical analyses were carried out using OriginPro Software using a one-way ANOVA  
 371 test and Tukey-HSD means comparison method (\*\*:  $p \leq 0,01$ ; \*:  $p \leq 0,05$ ).

372 **3.2.1. Impact of the material (DE values and glassy/rubbery state)**

373 The reconstitution times observed for the three maltodextrins prove the DE values' crucial  
374 role and thus of the molecular weight (**Figure 4**). First, a tendency for faster reconstitution  
375 times is observed when the DE values increase whatever the stirring rate and the  
376 glassy/rubbery state. This reconstitution improvement is particularly strong between low and  
377 intermediate DE values but not significant between intermediate and high DE values. The  
378 tendency of shorter reconstitution times with the increase of the DE value is confirmed under  
379 the glassy and the rubbery state. That means that lower molecular weight powders (i.e., high  
380 DE) tend to reconstitute faster. Thus, the hydrolyzing process upstream of the powder  
381 manufacturing is determinant to anticipate the reconstitution of the powder, knowing that  
382 maltodextrin powders are derivate products of starch hydrolysis. The first reconstitution step,  
383 the wettability, may explain the reconstitution differences observed. Indeed, the powder  
384 wettability was observed to be longer when the DE value was lower, as it also shown by the  
385 reconstitution kinetics (Appendix C). This observation was supported by Dupas et al., (2017),  
386 who studied the contact angle of maltodextrins powders (DE 2, 6 and 29) with water. This  
387 later was lower for higher DE value maltodextrins, meaning that low molecular weight  
388 maltodextrins should present a better wettability and thus better reconstitution times. Longer  
389 reconstitution times for low DE maltodextrins could also be related to the solution viscosity.  
390 Low DE maltodextrins solutions are more viscous than those with high DE values. The high  
391 viscosity is directly related to the molecular weight (Castro et al., 2016; Dokic, Jakovljevic, &  
392 Dokic-Baucal, 1998; Dokic, Jakovljevic, & Dokic, 2004; Siemons et al., 2020, Juszczak,  
393 Galkowska, Witczak, & Fortuna, 2013). Knowing that, low DE powders may lead to poor  
394 water diffusion between intra and interparticular pores, thus delaying the wetting time and  
395 increasing the overall reconstitution time (Ong, Taylor, & Ramaioli, 2020).

396 The impact of glass transition on reconstitution can also be highlighted in **Figure 4**.  
397 Reaching the T<sub>g</sub> for powders under the rubbery state gives systematically shorter

398 reconstitution times for low and intermediate DE values. This may be first related to the fact  
 399 that particle surface is more hydrated, as powders are at higher  $a_w$  in the rubbery state, as  
 400 detailed in experimental conditions defined in **Table 1**. However, this is only true for low and  
 401 intermediate DE maltodextrins, as differences in the reconstitution times between the glassy  
 402 and the rubbery state tend to attenuate with higher DE values. Indeed, there are no significant  
 403 differences in the reconstitution times for high DE powders between the glassy and the  
 404 rubbery state. To better understand these differences, the impact of particle size should be  
 405 taken into account. The glass transition is most of the time accompanied by caking, making  
 406 interparticular forces (such as liquid or solid bridges) stronger. This phenomenon may delay  
 407 the reconstitution times by the formation of lumps during powder reconstitution, making the  
 408 dispersion step more difficult (Dupas et al., 2017; Fitzpatrick, 2021; Roos, 2002). In order to  
 409 see if there is a possible impact of particle size on powders reconstitution time, particle size  
 410 distribution was evaluated (**Figure 5**).



GLASSY STATE	D <sub>10</sub> (μm)	D <sub>50</sub> (μm)	D <sub>90</sub> (μm)	Span
Low DE	27.4 ± 0.7	91.6 ± 2.0	220 ± 2.6	2.10 ± 0.1
Intermediate DE	11.4 ± 0.5	86.7 ± 1.7	204.3 ± 15.3	2.20 ± 0.2
High DE	14.5 ± 0.1	42.5 ± 0.3	204.3 ± 2.1	1.98 ± 0.1

RUBBERY STATE	D <sub>10</sub> (μm)	D <sub>50</sub> (μm)	D <sub>90</sub> (μm)	Span
Low DE	29.5 ± 0.3	93.0 ± 1.4	253.3 ± 43.1	2.0 ± 0.1
Intermediate DE	12.0 ± 0.2	88.7 ± 2.6	271.0 ± 9.0	2.3 ± 0.1
High DE	16.3 ± 0.1	55.2 ± 1.1	142.6 ± 7.2	2.3 ± 0.1

411  
 412 **Figure 5.** Maltodextrins (low, intermediate, and high DE values) particle size distribution under glassy and  
 413 rubbery states.

414 It can be highlighted that low and high DE powders presented a monomodal particle size  
415 distribution while intermediate DE presented a bimodal distribution. The impact of glass  
416 transition on particle size and the width of particle size distribution was only observed for  
417 high DE maltodextrins with a  $D_{50}$  increase from 42.5  $\mu\text{m}$  to 55.2  $\mu\text{m}$ . No significative  
418 differences were found between the glassy and the rubbery state conditions for low and  
419 intermediate DE maltodextrins. Therefore, particles size was not directly correlated to both  
420 the DE value and the powder glassy or rubbery state. However, those results could explain the  
421 reconstitution times obtain at low shear rates, as particle hydration and swelling could be  
422 responsible of lumping tendency, especially for low DE maltodextrins, making particles  
423 dispersion more difficult, thus making reconstitution longer.

### 424 **3.2.3. Shear rate effect**

425 Beyond the material effect (i.e. DE values and glassy/rubbery state), the reconstitution time  
426 was also directly dependent on the shear rate (**Figure 4**). The increase of the shear rate from  
427 500 to 1500 rpm decreases significantly the reconstitution time whatever the powder. For  
428 example, the reconstitution time decreases 99% under the glassy state and 97% under the  
429 rubbery state for low DE powders and it varies in the same way for the other DE value  
430 maltodextrins. These results agree with those in the literature, describing that high shear rate  
431 allows logically shorter reconstitution times, and is also useful to prevent lump formation or  
432 sedimentation, especially for maltodextrins powders (Mitchell et al., 2014). Differences  
433 between the reconstitution times for all maltodextrins tend to attenuate with the increase of  
434 the solution stirring. Indeed, high shear rates allow instant reconstitution properties, whatever  
435 the powder. However, a high variability for the reconstitution time is noticed at high shear  
436 rates (1250 rpm), as it is particularly the case for high DE maltodextrins in the rubbery state  
437 and to a lesser extent in the glassy state.

438 Also, the shear rate seems to improve the reconstitution time, but the impact is not the  
439 same depending on the material's physical state. Until now, only a few works accurately  
440 describe the effect of both shear rate and powder physical state on reconstitution. Gaudel et  
441 al., (2022) studied the impact of the glass transition on different fruit-based powders and  
442 highlighted that reconstitution rate was impacted by the powder physical state. They observed  
443 that fruits powders under the glassy state were shear rate dependent, while the same powders  
444 under a rubbery state were shear rate independent. A viscous layer was formed at the particle  
445 surface for powders under a rubbery state, preventing water from diffusing. Our results are in  
446 agreement with this study as maltodextrin powders under the glassy state were found more  
447 shear rate dependent than powders under the rubbery state. Nevertheless, shear rate  
448 differences between glassy and rubbery powders reconstitution times were attenuated by the  
449 increase of DE, showing its strong impact.

450 All these results agree with the role of the powder physical (glassy/rubbery) and chemical  
451 (DE values) properties on reconstitution. In particular, the shear rate applied to reconstitute  
452 the powders was found to have a greater impact when the material is under the glassy state,  
453 meaning that increasing stirring improves reconstitution properties. The results also  
454 highlighted that wetting seems to be a determining step of powders reconstitution behavior at  
455 low shear rates, especially for maltodextrins (Dupas et al., 2017).

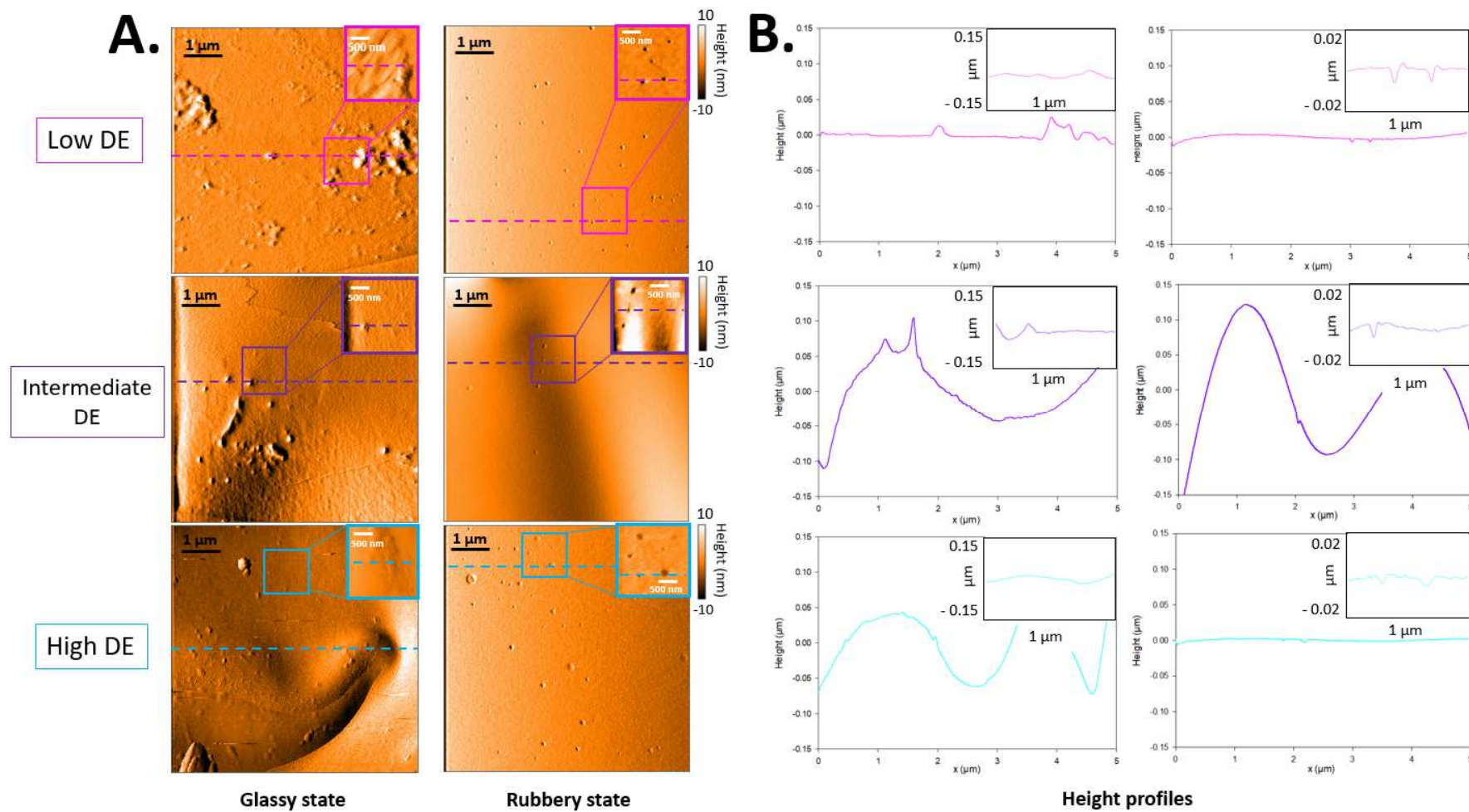
### 456 **3.3. Probing single particle surface properties by Atomic Force Microscopy (AFM)**

457 AFM is a powerful and resolute tool to analyze the surface of food powders. This  
458 technique is also very versatile as it allows to characterize with a nanoscale precision the  
459 surface topography, its roughness and the nanomechanical properties of single particles  
460 (Burgain et al., 2017; Murrieta-Pazos et al., 2012).

#### 461 **3.3.1. Surface topography and roughness analysis**

462 AFM topographies and their associated height profiles performed under the glassy or  
463 rubbery states on single maltodextrins particles are displayed in **Figure 6**.



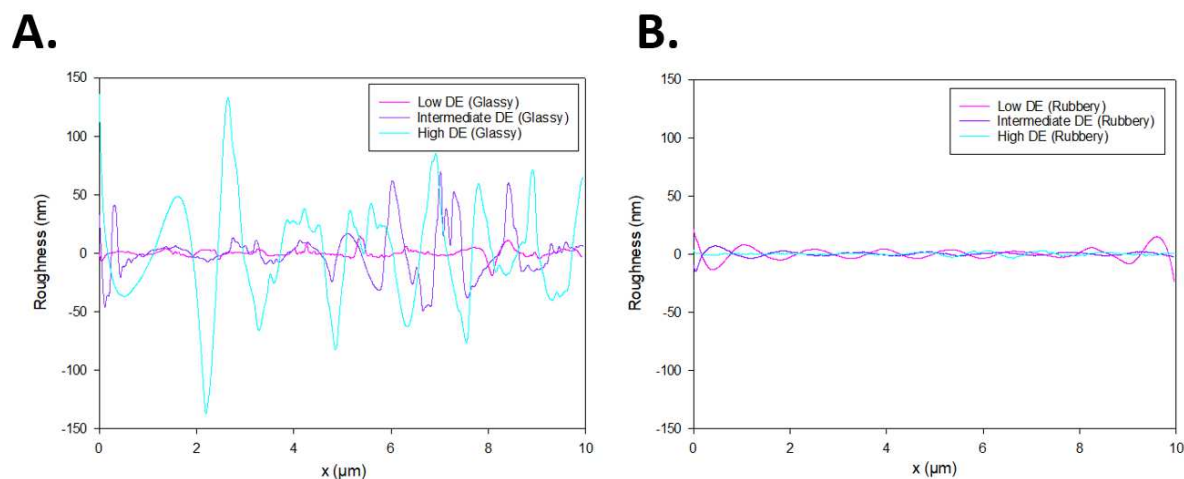


464

465 **Figure 6.** A. AFM deflection views of all maltodextrins under glassy (left) or rubbery (right) experimental conditions. B. Height profiles under glassy (left) or rubbery (right)

466 states associated to the previous topographies. Dash lines in A. are corresponding to the height profiles extracted from raw data in B.

467 Powders surface topography seem to be directly correlated to the DE value, especially  
468 when they are under the glassy state (**Figure 6A**). By taking a closer look at the height  
469 profiles of powders under the glassy state, increasing the DE value makes the surface more  
470 levelled with a lot of irregularities (**Figure 6B**). Thus, low DE maltodextrins appear to have a  
471 more regular surface and smooth than higher DE maltodextrins. To highlight the effect of the  
472 DE values on surface topography, the average surface roughness (Sa) was calculated on  $10 \times$   
473  $10 \mu\text{m}$  scanned areas, according to **Equation 2**. Extracted roughness profiles from acquired  
474 images are also displayed in **Figure 7A**.



475  
476 **Figure 7.** Low, intermediate, and high DE maltodextrins roughness profiles under the glassy (A.) or rubbery (B.)  
477 state experimental conditions

478 Thus, low DE maltodextrins were the ones with the lowest estimated average surface  
479 roughness value ( $S_a = 12.36 \pm 5.59 \text{ nm}$ ) under a glassy state. Then, a localized increase in the  
480 surface roughness was observed for intermediate DE maltodextrins ( $S_a = 79.09 \pm 26.56 \text{ nm}$ )  
481 followed by high DE maltodextrins ( $S_a = 128.83 \pm 25.36 \text{ nm}$ ). That means that the more the  
482 maltodextrin is hydrolyzed, the higher the global overall particle surface roughness will be.  
483 Thanks to SEM, Takeiti et al. (2010) evidenced the effect of the botanical source and the  
484 drying process conditions on the size and shape of produced maltodextrin powders. Siemons  
485 et al. (2020) highlighted the effect of the DE value on the particle morphology in a single

486 sessile droplet drying. Low DE value maltodextrins were generally smoothly shaped particles  
487 with large cavities. The opposite can be observed for high DE value maltodextrins presenting  
488 many surface irregularities. Here, for the first time, AFM was used to evidence localized  
489 variations of the surface roughness of maltodextrins correlated to their DE value, showing that  
490 high DE value maltodextrins exhibit a much more irregular surface than those by a lower DE  
491 value. These variations on particles surface topographies could also explain the better  
492 reconstitution times observed at low shear rates for high DE value maltodextrins compared to  
493 lower ones (**Figure 4**). An increase of the overall surface roughness at the particle surface  
494 could enhance the contact with water, by increasing the surface area in contact with the  
495 reconstitution media, thus allowing shorter reconstitution times. On the best knowledge of  
496 authors, the effect of surface roughness on reconstitution properties is poorly described in the  
497 literature. A few studies detailing how roughness can influence contact with water were  
498 reported on other food materials, such as spaghettis. Their rehydration kinetics were improved  
499 by altering the overall surface roughness by the use of different dies during manufacturing. It  
500 was shown that low surface roughness materials had longer rehydration times compared to  
501 higher surface roughness materials, meaning that measuring roughness was relevant when it  
502 comes to reconstitution kinetics, as it could be an enhancer of contact with water (Ogawa &  
503 Adachi, 2014, Ogawa & Adachi, 2017).

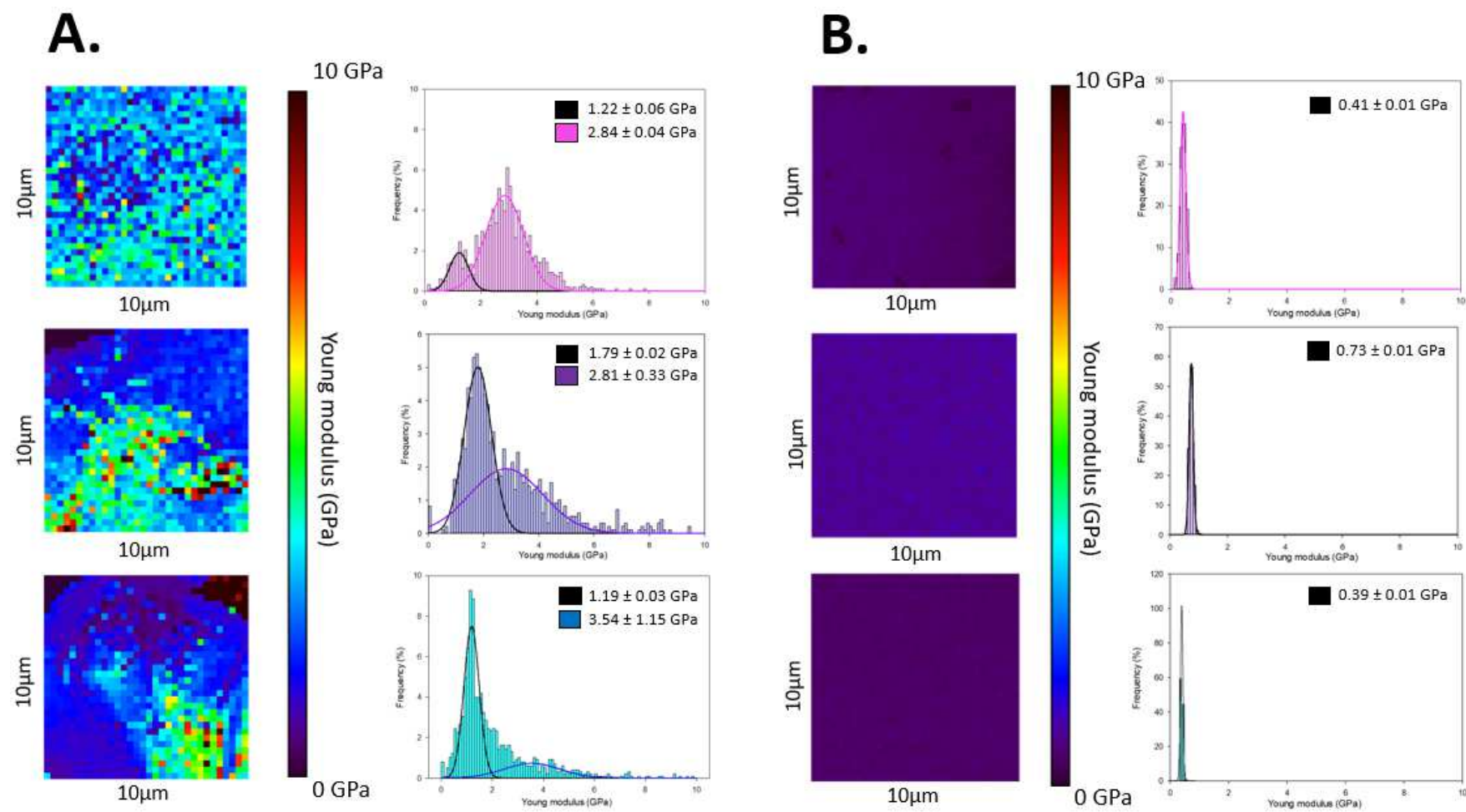
504 The impact of glass transition on single-particle topography and surface roughness can also  
505 be highlighted (**Figure 6**) (**Figure 7B**). The surface appears much smoother when the material  
506 is rubbery than when it is in the glassy state. Average surface roughness was estimated to be  
507  $8.70 \pm 2.30$  nm for low,  $6.19 \pm 5.34$  nm for intermediate and  $7.82 \pm 4.45$  nm for high DE  
508 values. This surface smoothing observed under the rubbery state could be explained by the  
509 increased water adsorbed as a thin layer at the particle surface, when powders are exposed to  
510 high relative humidity. These results could also be related to the reconstitution times (**Figure**

511 4). Improved reconstitution times were observed under the rubbery state due to a more  
512 hydrated surface layer. This enhanced the powder wettability, thus allowing lower  
513 reconstitution times, except for high DE maltodextrins.

514 In addition to the overall surface smoothing, the glass transition was accompanied by the  
515 formation of slight hollow zones of about 3 to 10 nm, as shown in **Figure 6**. This kind of  
516 hollow area, which seems to appear randomly, could be related to water adsorption at the  
517 particle surface followed by a swelling phenomenon. Indeed, the swelling observed by  
518 Deslandes et al., (2019) for carbohydrates polymers at a temperature range within 60 to 80 °C  
519 corresponds to the experimental conditions used in this work (**Table 1**).

### 520 **3.3.2. Mapping single particles nanomechanical properties**

521 Another use of AFM is the analysis of the Young modulus at the particle surface by the  
522 generation of elasticity mapping, giving information about single particles nanomechanics.  
523 Elasticity mapping and associated distribution histograms of all maltodextrins are displayed in  
524 **Figure 8**.



525

526 **Figure 8.** Young modulus mapping (10 $\times$ 10  $\mu\text{m}$ ) and associated distribution histograms for maltodextrin single particles. A. under a glassy state and B. under a rubbery state

527

(Up: low DE; middle: intermediate DE; bottom: high DE)

528 In the glassy state, all maltodextrins show two Young modulus populations (**Figure 8A**)  
529 with a distribution rather scattered and presenting nanoscale slight variations. Glassy state  
530 maltodextrins exhibit two Young modulus populations with a gaussian distribution. Thus, low  
531 DE maltodextrins modulus distribution was calculated at  $1.22 \pm 0.06$  GPa for the first  
532 population and  $2.84 \pm 0.04$  GPa for the second one. In the same way, modulus of intermediate  
533 DE maltodextrins was estimated at  $1.79 \pm 0.02$  GPa and  $2.81 \pm 0.33$  GPa and was about  $1.19$   
534  $\pm 0.03$  GPa and  $3.54 \pm 1.15$  GPa for high DE maltodextrins. The presence of two Young  
535 modulus populations with harder regions could not be first related to crystallized areas, as  
536 crystallization is unlikely for maltodextrins. This could maybe be explained by a low range of  
537 molecular association between polymer chains, similar to starch retrogradation. Indeed,  
538 retrogradation is induced at low temperatures and is dependent on the molecular weight.  
539 During this event, association of different molecular weight polymer chain occurs and  
540 intermolecular distance decreases, leading to a water removal (Karim, Norziah, & Seow,  
541 2000; Sobolewska-Zielinska & Fortuna, 2010). This could lead to the creation of pockets with  
542 lower moisture content and which show a higher Young modulus, explaining the nanoscale  
543 variation of the Young modulus.

544 Up to now, nanoindentation studies have been mostly performed on pharmaceutical  
545 powders which contain various carbohydrates as excipients. Masterson and Cao, (2008) report  
546 a better understanding of the mechanical properties of complex pharmaceutical powders with  
547 the characterization of the modulus of crystalline sucrose and lactose single particles. They  
548 found that the hardness value varies from 1.8 to 2.4 GPa for crystalline sucrose particles and  
549 from 0.18 to 0.51 GPa for lactose particles. Only few authors used AFM to assess the  
550 nanomechanical properties of food powders. Burgain et al., (2016b) investigated rehydration  
551 impairments linked to the surface hardening of micellar casein powders depending on storage  
552 conditions, especially temperature. The Young modulus of fresh casein powders was

553 estimated at 0.2 GPa whereas it was about 20 GPa for aged casein powders. Haider et al.,  
 554 (2013) characterized the Young modulus of glassy DE 21 maltodextrins. They found a  
 555 modulus value of  $3.34 \pm 1.1$  GPa, which is relatively close to the values obtain in this work.

556 Elasticity mapping and modulus histograms distribution of the different maltodextrins in  
 557 rubbery state are displayed in **Figure 8B**. As shown, powders Young modulus value are of  
 558  $0.41 \pm 0.01$  GPa for low DE,  $0.73 \pm 0.01$  GPa for intermediate DE and  $0.39 \pm 0.01$  GPa for  
 559 high DE maltodextrins. A large decrease of the Young modulus at the surface of the  
 560 maltodextrin particle can be observed. Moreover, a single Gaussian population is observed,  
 561 meaning that the elasticity distribution at the surface becomes less dispersed and more  
 562 homogenous. Particles under a rubbery state are thus softer because of a hydrated surface  
 563 layer. Already existing water adsorption at the particle surface, explaining the decrease of the  
 564 surface hardness and the initiation of the wetting step for particles under a rubbery state.

565 **Table 2.** Summary of the results presented in this work, showing the direct link between material physical state,  
 566 from macro- to microscale and reconstitution properties.

Maltodextrin powders (low/intermediate/high DE values)	Glassy state	Rubbery state
<b>Macroscale properties</b>		
Glass transition temperature	<ul style="list-style-type: none"> <li>• Tg decreases with the increase of the DE value (i.e., decreasing polymer chain length)</li> <li>• Increasing water content leads to a decrease of Tg (plasticizing effect)</li> </ul>	
Reconstitution time	<ul style="list-style-type: none"> <li>• Reconstitution strongly shear rate dependent</li> </ul>	<ul style="list-style-type: none"> <li>• Reconstitution poorly shear rate dependent</li> </ul>
	<ul style="list-style-type: none"> <li>• Better reconstitution times for high DE values</li> <li>• Better reconstitution times when maltodextrin is under the rubbery state</li> <li>• Differences in reconstitution times between glassy and rubbery states are attenuated for high DE values</li> </ul>	
<b>Mesoscale / Microscale properties</b>		
Particle size	<ul style="list-style-type: none"> <li>• All maltodextrins present the same particle size</li> </ul>	<ul style="list-style-type: none"> <li>• Slight particle size increase due to water adsorption at the surface</li> </ul>
Particle surface	<ul style="list-style-type: none"> <li>• Rough particles</li> </ul>	<ul style="list-style-type: none"> <li>• Smooth particles and low</li> </ul>

topography	<ul style="list-style-type: none"> <li>• Increase of surface roughness with the DE values</li> </ul>	surface roughness <ul style="list-style-type: none"> <li>• Formation of nanoscale hollows structures</li> </ul>
Young modulus	<ul style="list-style-type: none"> <li>• High Young modulus</li> <li>• Bimodal distribution with harder points (related to lower moisture content regions)</li> </ul>	<ul style="list-style-type: none"> <li>• Low Young modulus</li> <li>• Homogenous distribution</li> </ul>

569

570 **4. CONCLUSION**

571 This work evaluated the reconstitution properties of maltodextrin powders depending on their  
 572 physical state (**Table 2**). To characterize the material properties, T<sub>g</sub> of different maltodextrins  
 573 were first determined, and T<sub>g</sub> was found dependent on both the maltodextrin molecular  
 574 weight and a<sub>w</sub>. Also, powder reconstitution times under the glassy state were mostly shear rate  
 575 and DE dependent, with better reconstitution times while increasing these two parameters. On  
 576 the contrary, the reconstitution properties of powders under the rubbery state were less shear  
 577 rate and DE dependent with better reconstitution times while increasing these two parameters.  
 578 Particle surface properties were also analyzed by AFM, which has been yet poorly used on  
 579 food powders. Particles under a glassy state were relatively rough and hard with a lot of  
 580 surface irregularities, whereas powders under a rubbery state were much smoother and softer.  
 581 The decrease of the Young modulus under the rubbery state was also related to a more  
 582 hydrated surface, which enhanced the wetting step. Finally, nanoscale hollows formation was  
 583 highlighted for particles under the rubbery state. The correct management of powder  
 584 reconstitution needs to consider powder macroscopic properties (granulometry, T<sub>g</sub>, chemical  
 585 composition) which are related to surface microscopic features (roughness, elasticity,  
 586 hygroscopicity). In this work the interplay between the two measurement scale is described  
 587 for maltodextrin powders and can be henceforward applied to other food powders.

588 **5. Acknowledgements**



589 The authors acknowledge financial support from the "Impact Biomolecules" project of the  
590 "Lorraine Université d'Excellence" (in the context of the « Investissements d'avenir »  
591 program implemented by the French National Research Agency (ANR 15-004)).

## 592 **6. References**

593 Arora, S., Kappl, M., Haghi, M., Young, P.M., Traini, D., & Jain, S., (2016).  
594 Investigation of surface properties, local elastic modulus and interaction with simulated  
595 pulmonary surfactant of surface modified inhalable voriconazole dry powders using atomic  
596 force microscopy. *RSC Advances*, 6 (31), 25789-25798. <https://doi.org/10.1039/C6RA01154C>

597 Avaltroni, F., Bouquerand, P.E., & Normand, V., (2004). Maltodextrin molecular  
598 weight distribution influence on the glass transition temperature and viscosity in aqueous  
599 solutions. *Carbohydrate Polymers*, 58, 323-334.  
600 <http://dx.doi.org/10.1016/j.carbpol.2004.08.001>

601 Barringer, S., (2013). Coating foods with powders. In B. Bhandari, N. Bansal, M.  
602 Zhang, P. Schuck (Eds.), *Handbook of Foods Powders* (pp. 625-640).  
603 <http://dx.doi.org/10.1533/9780857098672.3.625>

604 Berard, V., Lesniewska, E., Andres, C., Pertuy, D., Laroche, C., & Pourcelot, Y.,  
605 (2002). Dry powder inhaler: influence of humidity on topology and adhesion studied by AFM.  
606 *International journal of Pharmaceutics*, 232 (1-2), 213-224. [http://dx.doi.org/10.1016/S0378-5173\(01\)00913-9](http://dx.doi.org/10.1016/S0378-5173(01)00913-9)

608 Bhandari, B., (2013). Introduction to food powders. In B. Bhandari, N. Bansal, M.  
609 Zhang, P. Schuck (Eds.), *Handbook of Foods Powders* (pp. 1-25).

610 Burgain, J., El Zein, R., Scher, J., Petit, J., Norwood, E.A., Francius, G., & Gaiani, C.,  
611 (2016a). Local modification of whey protein isolate powder surface during high temperature  
612 storage. *Journal of Food Engineering*, 178, 39-46.  
613 <https://dx.doi.org/10.1016/j.jfoodeng.2016.01.005>

614 Burgain, J., Scher, J., Petit, J., Francius, G., & Gaiani, C., (2016b). Links between  
615 particle surface hardening and rehydration impairment during micellar casein powder storage.  
616 *Food Hydrocolloids*, 61, 277-285. <https://doi.org/10.1016/j.foodhyd.2016.05.021>

617 Burgain, J., Petit, J., Scher, J., Rasch, R., Bhandari, B., & Gaiani, C., (2017). Surface  
618 chemistry and microscopy of food powders. *Progress in Surface Science*, 92 (4), 409-429.  
619 DOI <https://doi.org/10.1016/j.progsurf.2017.07.002>

620 Butt, H.-J., Cappella, B., & Kappl, M., (2005). Force measurements with the atomic  
621 force microscope: Technique, interpretation and applications. *Surface Science Reports*, 59, 1-  
622 152. <https://doi.org/10.1016/j.surfrep.2005.08.003>

623 Calabri, I., Pugno, N., Menozzi, C., & Valeri, S., (2008). AFM nanoindentation: tip  
624 shape and tip radius of curvature effect on the hardness measurement. *Journal of Physics: Condensed Matter*, 20. <http://dx.doi.org/10.1088/0953-8984/20/47/474208>

626 Cardenas-Perez, S., Chanona-Perez, J.J., Mendez-Mendez, J.V., Arzate-Vazquez, I.,  
627 hernandez-Varela, J.D., & Guemes Vera, N., (2019). Recent advances in atomic force

628 microscopy for assessing the nanomechanical properties of food materials. *Trends in Food*  
629 *Sciences & Technology*, 87, 59-72. <https://doi.org/10.1016/j.tifs.2018.04.011>

630 Castro, N., Durrieu, V., Raynaud, C., & Rouilly, A., (2016). Influence of DE-value on  
631 the physicochemical properties of maltodextrin for melt extrusion processes. *Carbohydrate*  
632 *Polymers*, 144, 464-473. <https://doi.org/10.1016/j.carbpol.2016.03.004>

633 Chronakis, I.S., (1998). On the molecular Characteristics, Compositional Properties, and  
634 Structural-Functional Mechanisms of Maltodextrins: A Review. *Critical Reviews in Food*  
635 *Science and Nutrition*, 38 (7), 599-637. <https://doi.org/10.1080/10408699891274327>

636 Clifford, C.A., & Seah, M.P., (2005). Quantification issues in the identification of  
637 nanoscale regions of homopolymers using modulus measurement via AFM nanoindentation.  
638 *Applied Surface Sciences*, 252, 1915-1933. <https://doi.org/10.1016/j.apsusc.2005.08.090>

639 Crowley, S.V., Kelly, A.L., Schuck, P., Jeantet, R., & O'Mahony, J.A., (2016).  
640 Rehydration and solubility characteristics of high protein dairy powders. In P.L.H.  
641 McSweeney, J.A. O'Mahony (Eds.), *Advanced Dairy Chemistry* (pp. 99-131).

642 Cuq, B. & Mandato, S., (2013). Agglomeration/granulation in food powder production.  
643 In B. Bhandari, N. Bansal, M. Zhang, P. Schuck (Eds.), *Handbook of Foods Powders* (pp.  
644 150-177).

645 Descamps, N., Palzer, S., Roos, Y.H., & Fitzpatrick, J.J., (2013). Glass transition and  
646 flowability/caking behavior of maltodextrin DE 21. *Journal of Food Engineering*, 119, 809-  
647 813. <http://dx.doi.org/10.1016/j.jfoodeng.2013.06.045>

648 Deslandes, F., Plana-Fattori, A., Almeida, G., Moulin, G., Doursat, C., & Flick, D.,  
649 (2019). Estimation of individual starch granule swelling under hydro-thermal treatment. *Food*  
650 *Structure*, 22, 100-125. <https://doi.org/10.1016/j.foostr.2019.100125>

651 Dokic, P., Jakovljevic, J., & Dokic-Baucal, Lj., (1998). Molecular characteristics of  
652 maltodextrins and rheological behaviour of diluted and concentrated solutions. *Colloids and*  
653 *Surface A: Physicochemical and Engineering Aspects*, 141, 435-440.  
654 [https://doi.org/10.1016/S0927-7757\(97\)00118-0](https://doi.org/10.1016/S0927-7757(97)00118-0)

655 Dokic, L., Jakovljevic, J., & Dokic, P., (2004). Relation between Viscous  
656 Characteristics and Dextrose Equivalent of Maltodextrins. *Starch*, 56 (11), 520-525.  
657 <https://doi.org/10.1002/star.200400294>

658 Dokukin, M. & Sokolov, I., (2012). On the measurements of Rigidity Modulus of Soft  
659 Materials in Nanoindentation Experiments at Small Depth. *Macromolecules*, 45 (10), 4277-  
660 4288). <https://doi.org/10.1021/ma202600b>

661 Dufrene, Y.F., (2002). Atomic Force Microscopy, a Powerful Tool in Microbiology.  
662 *Journal of Bacteriology*, 184 (19), 5205-5213. [https://doi.org/10.1128/JB.184.19.5205-](https://doi.org/10.1128/JB.184.19.5205-5213.2002)  
663 [5213.2002](https://doi.org/10.1128/JB.184.19.5205-5213.2002)

664 Dupas, J., Girard, V., & Forny, L., (2017). Reconstitution properties of Sucrose and  
665 Maltodextrins. *Langmuir*, 33, 988-995. <https://doi.org/10.1021/acs.langmuir.6b04380>

666 Eaton, P. & West, P., (2010). *Atomic Force Microscopy*, Oxford University Press.

667 Fitzpatrick, J.J., Hodnett, M., Twomey, M., Cerqueira, P.S.M., O'Flynn, J., & Roos,  
668 Y.H., (2007). Glass transition and the flowability and caking of powders containing

669 amorphous lactose. *Powder Technology*, 178, 119-128.  
670 <https://doi.org/10.1016/j.powtec.2007.04.017>

671 Fitzpatrick, J.J., van Lauwe, A., Coursol, M., O'Brien, A., Fitzpatrick, K.L., Ji, J., &  
672 Miao, S., (2016). Investigation of the rehydration behaviour of food powders by comparing  
673 the behaviour of twelve powders with different properties. *Powder Technology*, 297, 340-348,  
674 <https://doi.org/10.1016/j.powtec.2016.04.036>

675 Fitzpatrick, J.J., Ji, J., & Miao, S., (2021). Characterisation of the Rehydration  
676 Behaviour of Food Powders. *Food Powders Properties and Characterization*, 91-108.  
677 [http://dx.doi.org/10.1007/978-3-030-48908-3\\_5](http://dx.doi.org/10.1007/978-3-030-48908-3_5)

678 Forny, L, Marabi, A., & Palzer, S., (2011). Wetting, disintegration and dissolution of  
679 agglomerated water-soluble powders. *Powder Technology*, 206, 72-78.  
680 <https://doi.org/10.1016/j.powtec.2010.07.022>

681 Fongin, S., Kawai, K., Harnkarnsujarit, N., & Hagura, Y., (2017). Effects of water and  
682 maltodextrin on the glass transition temperature of freeze-dried mango pulp and an empirical  
683 model to predict plasticizing effect of water on dried fruits. *Journal of Food Engineering*,  
684 210, 91-97. <http://dx.doi.org/10.1016/j.jfoodeng.2017.04.025>

685 Fournaise, T., Petit, J., & Gaiani, C., (2021). Main powder physicochemical  
686 characteristics influencing their reconstitution behavior. *Powder Technology*, 383, 66-73. DOI  
687 <https://doi.org/10.1016/j.powtec.2021.01.056>

688 Frascareli, E.C., Silva, V.M., Tonon, R.V., & Hubinger, M.D., (2012). Determination of  
689 critical storage conditions oil microcapsule by coupling water sorption isotherms and glass  
690 transition temperature. *International Journal of Food Science and Technology*, 47 (5), 1044-  
691 1054. <http://dx.doi.org/10.1111/j.1365-2621.2012.02939.x>

692 Gaboriaud, F., & Dufrene, Y.F., (2007). Atomic force microscopy of microbial cells:  
693 Application to nanomechanical properties, surface forces and molecular recognition forces.  
694 *Colloids Surface B: Biointerfaces*, 54, 10-19. <https://doi.org/10.1016/j.colsurfb.2006.09.014>

695 Gaiani, C., Scher, J., Ehrhardt, J.J., Linder, M., Schuck, P., Desobry, S., Banon, S., &  
696 (2007a). Relationships between Dairy Powder Surface Composition and Wetting Properties  
697 during Storage: Importance of Residual Lipids. <https://doi.org/10.1021/jf070364b>

698 Gaiani, C., Schuck, P., Scher, J., Desobry, S., Banon, S., (2007b). Dairy Powder  
699 Rehydration: Influence of Protein State, Incorporation Mode, and Agglomeration. *Journal of*  
700 *Dairy Science*, 90 (2), 570-581. [https://doi.org/10.3168/jds.S0022-0302\(07\)71540-0](https://doi.org/10.3168/jds.S0022-0302(07)71540-0)

701 Gaiani, C., Burgain, J., & Scher, J., (2013). Surface composition of food powders. In B.  
702 Bhandari, N. Bansal, M. Zhang, P. Schuck (Eds.), *Handbook of Foods Powders* (pp. 339-  
703 378). <https://dx.doi.org/10.1533/9780857098672.2.339>

704 Gaudel, N., Gaiani, C., Harshe, Y.M., Kammerhofer, J., Pouzot, M., Desobry, S., &  
705 Burgain, J., (2022). Reconstitution of fruit powders: A process-structure-function approach.  
706 *Journal of Food Engineering*, 315. <https://doi.org/10.1016/j.jfoodeng.2021.110800>

707 Guz, N., Dokukin, M., Kalaparthy, V., Sokolov, I., (2014). If Cell Mechanics Can Be  
708 Described by Elastic Modulus: Study of Different Models and Probes Used in Indentation  
709 Experiments. *Biophysical Journal*, 107 (3), 564-575.  
710 <https://doi.org/10.1016/j.bpj.2014.06.033>

711 Haider, C.I., Althaus, T.O., Niederreiter, G., Palzer, S., Hounslow, M.J., & Salman,  
712 A.D., (2013, June). *Elastic behavior of glassy cohesive maltodextrin in particle contact*  
713 *experiments*. Paper presentation for the 6th International Granulation Workshop, Sheffield,  
714 UK.

715 Hertz, H., (1882). *Über die Berührung fester elastischer Körper*.  
716 <https://doi.org/10.1515/crll.1882.92.156>

717 Islam, I.-U., & Langrish, T.A.G., (2013). Modelling crystallization in spray-drying for  
718 food powder production. In B. Bhandari, N. Bansal, M. Zhang, P. Schuck (Eds.), *Handbook*  
719 *of Foods Powders* (pp. 105-131)

720 Juszczak, L., Galkowska, D., Witczak, T., & Fortuna, T., (2013). Effect of  
721 Maltodextrins on the Rheological Properties of Potato Starch Pastes and Gels. *International*  
722 *Journal of Food Science*, 2013, 1-7. <https://doi.org/10.1155/2013/869362>

723 Karim, A.A., Norziah, M.H., & Seow, C.C., (2000). Methods for the study of starch  
724 retrogradation. *Food Chemistry*, 71, 9-36. [https://doi.org/10.1016/S0308-8146\(00\)00130-8](https://doi.org/10.1016/S0308-8146(00)00130-8)

725 Kong, F. & Singh, R.P., (2016). Chemical Deterioration and Physical Instability of  
726 Foods and Beverages. In P. Subramaniam (Eds.), *The stability and Shelf Life of Food* (pp. 43-  
727 76). <http://dx.doi.org/10.1016/B978-1-84569-701-3.50002-5>

728 Levy, R., & Maaloum, M., (2001). Measuring the spring constant of atomic force  
729 microscope cantilevers: thermal fluctuations and other methods. *Nanotechnology*, 13 (1), 33.  
730 <http://dx.doi.org/10.1088/0957-4484/13/1/307>

731 Li, K., Pan, B., Ma, L., Miao, S., & Ji, J., (2020). Effect of Dextrose Equivalent on  
732 Maltodextrin/Whey Protein Spray-Dried Powder Microcapsules and Dynamic Release of  
733 Loaded Flavor during Storage and Powder Rehydration. *Foods*, 9 (12), 1878.  
734 <https://doi.org/10.3390/foods9121878>

735 Liao, X., & Wiedmann, T.S., (2003). Characterization of Pharmaceutical Solids by  
736 Scanning Probe Microscopy. *Journal of Pharmaceutical Sciences*, 93 (9), 2250-2258.  
737 <https://doi.org/10.1002/jps.20139>

738 Masterson, V.M., & Cao, X., (2008). Evaluating particle hardness of pharmaceutical  
739 solids using AFM nanoindentation. *International Journal of Pharmaceutics*, 262, 163-171.  
740 <https://doi.org/10.1016/j.ijpharm.2008.06.015>

741 Mitchell, W.R., Forny, L., Althaus, T.O., Niederreiter, G., Palzer, S., Hounslow, M.J.,  
742 & Salman, A.D., (2014). Mapping the rate-limiting regimes of food powder reconstitution in a  
743 standard mixing vessel. *Powder Technology*, 270, 520-527.  
744 <https://doi.org/10.1016/j.powtec.2014.08.014>

745 Murrieta-Pazos, I., Gaiani, C., Galet, L., Cuq, B., Desobry, S., & Scher, J., (2011).  
746 Comparative study of particle structure evolution during water sorption: Skim and whole milk  
747 powders. *Colloids and Surface B: Biointerfaces*, 87 (1), 1-10.  
748 <https://doi.org/10.1016/j.colsurfb.2011.05.001>

749 Murrieta-Pazos, I., Gaiani, C., Galet, L., Calvet, R., Cuq, B., & Scher, J., (2012). Food  
750 powders: Surface and form characterization revisited. *Journal of Food Engineering*, 112, 1-  
751 21. <https://dx.doi.org/10.1016/j.jfoodeng.2012.03.002>

752 Ogawa, T., Adachi, S., (2014). Effect of surface roughness on rehydration kinetics of  
753 spaghetti. *Japan Journal of Food Engineering*, 15 (2), 101-104.  
754 <http://dx.doi.org/10.11301/jsfe.15.101>

755 Ogawa, T., & Adachi, S., (2017). A Simple Method to Measure the Surface Roughness  
756 of Spaghetti Using a Digital Camera. *Food Science and Technology Research*, 2, 237-240.  
757 <https://doi.org/10.3136/fstr.23.237>

758 Ong, X.Y., Taylor, S.E., & Ramaioli, M., (2020). Rehydration of food powders:  
759 Interplay between physical properties and process conditions. *Powder Technology*, 371, 142-  
760 153. <https://doi.org/10.1016/j.powtec.2020.05.066>

761 Palzer, S., (2007). Agglomeration of Dehydrated Consumer Foods. In A.D. Salman, M.  
762 Ghadiri, & M.I. Hounslow (Eds.) *Handbook of Food Technology* (pp. 592-671).  
763 [https://doi.org/10.1016/S0167-3785\(07\)80048-0](https://doi.org/10.1016/S0167-3785(07)80048-0)

764 Perkins, M., Ebbens, S.J., Hayes, S., Roberts, C.J., Madden, C.E., & Luk, S.Y., (2007).  
765 Elastic modulus measurements from individual lactose particles using atomic force  
766 microscopy. *International Journal of Pharmaceutics*, 332 (1), 168-175.  
767 <https://doi.org/10.1016/j.ijpharm.2006.09.032>

768 Prime, D.C., Stapley, A.G.F., Rielly, C.D., Jones, J.R., & Leaper, M.C., (2011a).  
769 Analysis of Powder Caking in Multicomponent Powders Using Atomic Force Microscopy to  
770 Examine Particle Properties. *Chemical Engineering and Applied Chemistry*, 34, 1, 98-102.  
771 <https://doi.org/10.1002/ceat.201000211>

772 Prime, D.C., Leaper, M.C., Jones, J.R., Richardson, D.J., Rielly, C.D., & Stapley,  
773 A.G.F., (2011b). Caking Behaviour Of Spray-Dried Powders - Using Scanning Probe  
774 Microscopy to Study Nanoscale Surface Properties and Material Composition. *Chemical*  
775 *Engineering and Applied Chemistry*, 34 (7), 1104-1108.  
776 <http://dx.doi.org/10.1002/ceat.201000537>

777 Ramos, K.J. & Bahr, D.F., (2007). Mechanical behavior assessment of sucrose using  
778 nanoindentation. *Journal of Material Research*, 22 (7), 2037-2045.  
779 <https://doi.org/10.1557/jmr.2007.0249>

780 Ratti, C., (2013). Freeze drying for food powder production. In B. Bhandari, N. Bansal,  
781 M. Zhang, P. Schuck (Eds.), *Handbook of Foods Powders* (pp. 57-84)

782 Rong, Y., Sillick M., & Gregson, C.M., (2009). Determination of Dextrose Equivalent  
783 Value and Number Average Molecular Weight of Maltodextrin by Osmometry. *Journal of*  
784 *Food Science*, 74 (1), 33-40. <https://doi.org/10.1111/j.1750-3841.2008.00993.x>

785 Roos, Y., & Karel, Y., (1992). Crystallization of Amorphous Lactose. *Journal of Food*  
786 *Science*, 57 (3), 775-777. <https://doi.org/10.1111/j.1365-2621.1992.tb08095.x>

787 Roos, Y.H., (2002). Importance of glass transition and water activity to spray-drying  
788 and stability of dairy powders. *Le lait, INRA Editions*, 82 (4), 475-484.  
789 <https://doi.org/10.1051/lait:2002025>

790 Roos, Y.H., (2006). Phase Transitions and Transformations in Food Systems. In D.R.  
791 Heldman & D.B. Lund, (Eds.), *Handbook of Food Engineering* (2<sup>nd</sup> ed.) (pp. 288-346)

792 Roos, Y.H. & Drusch, S., (2016). Introduction to phase transitions. In Y.H., Roos & S.  
793 Drusch (Eds.), *Phase Transitions in Foods* (2<sup>nd</sup> Edition) (pp 1-17)

794 Roudaut, G., Simatos, D., Champion, D., Contreras-Lopez, E., Le Meste, M., (2004).  
795 Molecular mobility around the glass transition temperature: a mini review. *Innovative Food*  
796 *Science & Emerging Technologies*. <https://dx.doi.org/10.1016/j.ifset.2003.12.003>

797 Rushton, J., Costich, E., & Everett, H., (1950). Power characteristics of mixing  
798 impellers. *Chemical Engineering Progress*, 46, 395–476.

799 Saavedra-Leos, M.Z., Leyva-Porras, C., Martinez-Guerra, E., Pérez-Garcia, S.A.,  
800 Aguilar-Martinez, J.A., Alvarez-Salas, C., (2014). Physical properties of inulin and inulin-  
801 orange juice: Physical characterization and technological application. *Carbohydrate*  
802 *Polymers*, 105, 10-19. <https://doi.org/10.1016/j.carbpol.2013.12.079>

803 Saavedra-Leos, M.Z., Leyva-Porras, C., Araujo-Diaz, S.B., Toxqui-Teran, A., Borrás-  
804 Enriquez, A., (2015). Technological Application of Maltodextrins According to the Degree of  
805 Polymerization. *Molecules*, 20 (12), 67-81. <https://doi.org/10.3390/molecules201219746>

806 Selomulya, C. & Fang, Y., (2013). Food powder rehydration. In B. Bhandari, N. Bansal,  
807 M. Zhang, P. Schuck (Eds.), *Handbook of Foods Powders* (pp. 379-408)

808 Shrestha, A.K., Ua-arak, T., Adhikari, B.P., Howes, T., & Bhandari, B., (2007). Glass  
809 transition behavior of spray-dried orange juice powder measured by differential scanning  
810 calorimetry (DSC) and thermal mechanical compression test (TMCT). *International Journal*  
811 *of Food Properties*, 10, 661-673. <https://doi.org/10.1080/10942910601109218>

812 Shojaei, S., Farokhi, M., Omidvar, R., Mottaghitlab, F., Haghhighipour, N., Shokrgozar,  
813 M.A., Ai, J., (2013). Essential Functionality of Endometrial and Adipose Stem Cells in  
814 Normal and Mechanically Motivated Conditions. *Journal of Biomaterials and Tissue*  
815 *Engineering*, 3 (5), 581-588. <https://doi.org/10.1166/jbt.2013.1115>

816 Siemons, I., Politiek, R.G.A., Boom, R.M., van der Sman, R.G.M., & Schutyser,  
817 M.A.I., (2020). Dextrose equivalence of maltodextrins determines particle morphology  
818 development during single sessile droplet drying. *Food Research International*, 131.  
819 <https://doi.org/10.1016/j.foodres.2020.108988>

820 Sobolewska-Zielinska, J., & Fortuna, T., (2010). Retrogradation of starches and  
821 maltodextrins of various origins. *Acta Scientiarum Polonorum Technologia Alimentaria*, 9(1),  
822 71-81.

823 Takeiti, C.Y., Kieckbusch, T.G., & Collares-Queiroz, F.P., (2010). Morphological and  
824 physicochemical characterization of commercial maltodextrins with different degrees of  
825 dextrose equivalent. *International Journal of Food Properties*, 13, 411-425.  
826 <https://doi.org/10.1080/10942910802181024>

827 Traini, D., Young, P.M., Rogueda, P., & Price, R., (2006). The Use of AFM and  
828 Surface Energy Measurements to Investigate Drug-Canister Material Interactions in a Model  
829 Pressurized Metered Dose Inhaler Formulation. *Aerosol Science and Technology*, 40 (4), 227-  
830 236. <https://doi.org/10.1080/02786820500543316>

831 Wang, Y., Truong, T., (2017). Glass transition and Crystallization in Foods. In B.  
832 Bhandari & Y.H. Roos (Eds.), *Non-Equilibrium States and Glass Transition in Foods* (pp.  
833 153-172)

834 Weiss, C., McLoughlin, P., & Cathcart, H., (2015). Characterisation of dry powder  
835 inhaler formulations using atomic force microscopy. *International Journal of Pharmaceutics*,  
836 494, 393-407. <https://doi.org/10.1016/j.ijpharm.2015.08.051>

837 White, R.P. & Lipson, J.E.G., (2016). Polymer Free Volume and Its Connection to the  
838 Glass Transition. *Macromolecules*, 49 (11), 3987-4007.  
839 <https://doi.org/10.1021/acs.macromol.6b00215>

840 Wu, X., Li, X., & Mansour, H.M., (2010). Surface analytical techniques in solid-state  
841 particle characterization for predicting performance in dry powder inhalers. *KONA Powder*  
842 *and Particle Journal*, 28, 3-19. <https://doi.org/10.14356/kona.2010005>

843 Woo, M.W. & Bhandari, B., (2013). Spray drying for food powder production. In B.  
844 Bhandari, N. Bansal, M. Zhang, P. Schuck (Eds.), *Handbook of Foods Powders* (pp. 29-56)

845 Yoshioka, S. & Aso, Y., (2007). Correlations between Molecular Mobility and  
846 Chemical Stability During Storage of Amorphous Pharmaceuticals. *Journal of*  
847 *Pharmaceutical Sciences*, 96 (5), 960-981. <https://doi.org/10.1002/jps.20926>

848 Young, P.M., Tobyn, M.J., Price, R., Buttrum, M., Dey, F., (2006). The use of colloid  
849 probe microscopy to predict aerosolization performance in dry powder inhalers: AFM and in  
850 vitro correlation. *Journal of Pharmaceutical Sciences*, 95 (8), 1800-1809.  
851 <https://doi.org/10.1002/jps.20660>

852

