

14 **Graphical abstract**

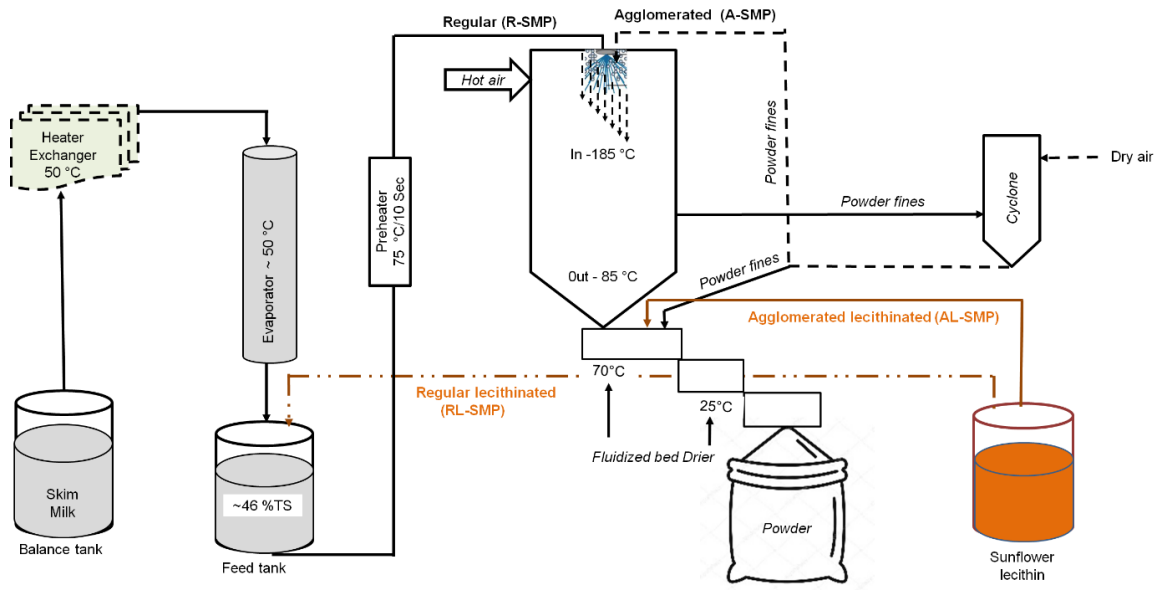


Figure 1. Graphical abstract flow chart for wettability and bulk density improvement of skim milk

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18 **Abstract**

19 This study aimed to reduce the bulk density of skim milk powders (SMP) and improve
 20 subsequent wettability and dissolution by a combination of agglomeration and lecithination.
 21 Agglomeration significantly increased powder particle size from a D_{90} of 120 to 201 μm , and
 22 decreased tapped bulk density (0.73 to 0.65 g/cm^3), although it led to increased friability
 23 (32.7%) compared to regular SMP (22.9%). Spraying lecithin on to SMP in the fluid bed
 24 improved wettability (8.94 s) compared to regular SMP (>300 s). Agglomeration without
 25 lectithination had no effect on powder wettability, similarly, adding lecithin in to liquid skim
 26 milk concentrate prior to drying did not improve subsequent powder wettability. Overall,
 27 improving the functionality of skim milk is quite complex, and while powder bulk density can
 28 be reduced, the particles remain susceptible to breakdown, and the wettability is relatively poor,
 29 but can be improved by spraying lecithin directly on to the powder particles

30 **Keywords;** Agglomeration; Lecithination; Skim milk powder; Surface composition; Water
 31 sorption; Wettability

32 1. Introduction

33 Skim milk powders (SMP) are used in numerous applications such as in yogurts, soft
34 acidified cheese, or as coffee and tea whiteners. SMP can often be the base ingredient for high-
35 value products including infant and medical nutritional formulations, mainly due to its high
36 protein (35-40%, w/w) and calcium content and concomitant low fat content (< 1.0%, w/w).
37 This low fat content makes skim milk ideal for the encapsulation of non-dairy vegetable oils
38 (Aghbashlo et al., 2012; Samira et al., 2017). For the most part SMP is rehydrated prior to
39 formulating complex nutritional products. This means that skim milk powder is first solubilized
40 only to be re-dried, however, with ever increasing sustainability demands, dry blending of SMP
41 with additional components (e.g., protein concentrates, lactose, base nutritional powders, etc.)
42 may provide an alternative process for nutritional product manufacture. However, a number of
43 issues are associated with dry blending SMP, such as high bulk density, low wettability and
44 low particle size. There are two main approaches for improving powder wettability and
45 flowability, namely, increasing powder particle size and modifying powder particle surface
46 composition (Hazlett, et al., 2021; Kim, et al., 2009; Barkouti, et al., 2013).

47 Increasing powder particle size is commonly achieved *via* a method known as
48 agglomeration (Hazlett, et al., 2021), which takes place either in the drying chamber or directly
49 after the initial drying stage in the fluid bed. Agglomeration can be achieved by two main
50 mechanisms within the drying chamber; spontaneous or forced. Spontaneous agglomeration
51 results from the random collision of atomized particles as they leave the atomizer and begin
52 drying, and can occur in both nozzle and rotary atomizers. Forced agglomeration is a result of
53 mixing dry particles with newly formed atomised droplets in the drying chamber, whereby dry
54 particles, known as fines, are removed from the outlet drying air using either a cyclone or bag
55 filter, before being pneumatically conveyed back to the drying chamber and introduced within
56 the atomization cloud. The particle structure that results is usually of a large primary particle

57 with smaller fine particles embedded on its surface. Particle agglomeration is usually
58 performed on infant formula, whole milk and fat-filled milk powders and is associated with
59 improving powder flowability, decreasing bulk density, and improving rehydration properties,
60 particularly, solubility. However, agglomeration is not commonly performed on a commodity
61 product like SMP, aside from a study by Turchiuli et al. (2013) where fluid bed agglomeration
62 of SMP was performed at laboratory scale.

63 Aside from changing the particle structure, the surface properties of powders can be
64 modified through addition of phospholipids such as lecithin (List, 2015; Smith, et al., 2016),
65 often added during spray drying between the drying chamber and fluid bed or *via* a rewetting
66 process within the fluid bed (Kelly and Fox, 2016; Hazlett, et al., 2021). Lecithination is a well-
67 established method of improving milk powder wettability, particularly for powders containing
68 high levels of saturated fatty acids, such as milk fat or certain plant-based oils (e.g., palm oil)
69 (Kim et al., 2009; Hazlett et al., 2021). For the most part lecithination is usually not required
70 for low fat or fat free dairy systems. However, the dry blending of dairy powders, such as skim
71 milk, which contains relatively high levels of protein (~35-40%, w/w, protein) can pose issues
72 with both high bulk density and low wettability. For the most part very little work has been
73 carried out on improving the functional properties of skim milk powders, rather, most research
74 has been performed on examining water sorption (Murrieta-Pazos et al., 2011; Shrestha et al.,
75 2007), emulsification (Marie et al., 2004; Sharma et al., 2012), infant formula properties
76 (McCarthy et al., 2012) and surface composition (Shrestha et al., 2007; Kim, et al., 2009).

77 Therefore, this study aimed to investigate the influence of agglomeration and lecithin
78 addition on the physical and functional properties of SMP.

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80

81

82 2. Materials and Methods

83 2.1. Production of regular, agglomerated and lecithinated skim milk powders

84 Pasteurized liquid skim milk (2000 kg) was obtained from a local milk processor. The pilot-
85 scale production of SMP was carried out at Moorepark Technology Limited (Teagasc,
86 Moorepark, Fermoy, Co. Cork, Ireland), where liquid skim milk was evaporated from an initial
87 dry matter content of 9.3% to a final concentrate solids of 46% (w/w) using a Niro three-effect
88 falling film evaporator (GEA Niro A/S, Soeborg, Denmark). The skim milk concentrate was
89 subsequently heated to 75 °C *via* a scraped surface heat exchanger before being spray dried
90 using a NIRO Tall-form spray dryer (TFD-0025-N, Soeborg, Denmark), equipped with a high
91 pressure nozzle atomization system. Air inlet and outlet temperatures were set at 185 and 85
92 °C, respectively, with the first and second external fluid bed temperatures set at 75 and 25 °C,
93 respectively. Four powders were produced in total:

94 R-SMP - Regular SMP achieved by straight through drying and where all fines collected
95 in the cyclone were returned to the second fluid bed;

96 A-SMP - Agglomerated SMP was achieved by returning all fines collected in the cyclone
97 to the top of the spray dryer and re-introduced within the atomization cloud;

98 † RL-SMP - Lecithination of regular SMP was achieved by addition of sunflower lecithin
99 (0.5%, w/w) in to the liquid concentrate prior to spray drying;

100 AL-SMP - lecithination of agglomerated SMP was performed by spraying liquid sunflower
101 lecithin at a concentration of 0.5%, w/w, (50 °C) on to the agglomerated powder in the
102 second fluid bed.

103 † *Direct spraying of lecithin on to non-agglomerated powder was not possible due to spray*
104 *dryer design. For R-SMP the fines in the outlet air were captured in the cyclone and*
105 *pneumatically conveyed to the second external fluid bed, thereby missing the lecithin nozzle*
106 *which was situated between the base of the drying chamber and entrance to the first fluid bed.*

107 *2.2. Bulk and particle surface composition*

108 The composition of powders were analysed using a standard procedures, i.e. Protein
109 using a LECO FP628 nitrogen analyser of Dumas method (LECO Corporation, St Joseph, MI,
110 USA), Fat (Rose Gottlieb method), Ash and Moisture content TGA701 thermo-gravimetric
111 analyser (LECO Corporation, St Joseph, MI, USA). Lactose content was obtained by
112 difference. All measurements were performed in triplicate unless otherwise specified.

113 X-ray photoelectron spectrophotometer (XPS) (Kratos Analytical, Manchester, UK),
114 equipped with a monochromatic Al K α X-ray source (1486.58 eV) at 150 W (15 kV, 10 mA)
115 was used to analyse the powders surface composition (i.e. fat, lactose, and protein) from the
116 corresponding spectra of carbon (C), oxygen (O) and nitrogen (N). A matrix formula was
117 created using experimental standard elemental values of the pure milk components (i.e. fat,
118 lactose, and protein) (Faldt et al., 1993). A relative coverage of components were calculated
119 for direct comparison of surface composition with bulk composition of major components (i.e.,
120 protein, fat and lactose).

121

122 *2.3. Particle size and specific surface area*

123 Particle size analysis of powders was performed using a Malvern Mastersizer (Mastersizer
124 3000; Malvern Instruments Ltd, Malvern, Worcestershire, UK) equipped with a Aero S dry
125 dispersion unit. The air pressure rate, feed rate and hopper gap was adjusted to 0 Bar, 60% and
126 2.00 mm, respectively, with the refractive index, obscuration and absorption set at 1.45, 1-8%
127 and 0.001, respectively. The compressed air was initially set to 0 bar to minimize powder
128 breakdown during the measurements. Size measurements of D_{10} , D_{50} and D_{90} (i.e. 10, 50 and
129 90%, respectively) were recorded, whereby 10, 50 and 90% of the sample volume is
130 represented by particles smaller than the size indicated. The volume weighted mean particle
131 diameter ($D_{4,3}$) was also calculated. Changes in specific surface area (SSA) were investigated

132 as a function of conveying air pressure (i.e., 0, 0.2, 0.5, 1.0, 2.0, 3.0, and 4.0 bar) in order to
133 investigate particle attrition and breakdown.

134

135 *2.4. Powder bulk handling properties*

136 To characterize powder bulk handling properties, the particle density (P_p), bulk density
137 (P_b), volume of occluded air (V_{oa}), volume of interstitial air (V_{ia}), and porosity (ϵ) of powders
138 were analysed. Particle density was measured using AccuPyc II 1340 gas pycnometer
139 (Micrometrics Instrument Corporation, Nor-cross, Georgia, USA) according to the air
140 pycnometry method of GEA Niro (2006a). The bulk and tapped (500) density of all powders
141 was measured according to the GEA Niro method (2006b), using a jolting volumeter STAV II
142 (Funke Gerber, Berlin, Germany). The interstitial air, occluded air and porosity were calculated
143 as described in the GEA Niro method (2006a). Powder porosity was calculated using Eq.1.

$$144 \quad \text{Porosity } (\epsilon) = 1 - (\text{tapped density}/\text{particle density}) \quad \text{Eq.1}$$

145

146 *2.5. Scanning Electron Microscopy*

147 Microscopy images of powders were captured using a Zeiss Supra 40P field emission
148 scanning electron microscopy (SEM) (Carl Zeiss SMT Ltd., Cambridge, UK) at 2.00 kV.
149 Samples were prepared using double sided adhesive microscope stubs coated with chromium
150 (K550X, Emitech, Ashford, UK). The microscopic structural images were obtained at 500×
151 magnification.

152

153 *2.6. Colour*

154 Colour of skim milk powders was measured using a Chroma Meter CR-400 (Konica
155 Minolta Sensing Europe B.V., Nieuwegein, the Netherlands) equipped with a specialized
156 granular attachment for powders. The coordinate L^* , a^* , and b^* were used to measure sample

157 colour variation. Coordinate L^* is a measure of brightness black (0) to white (100), a^* is green
 158 (-) to red (+), and b^* (blue (-) to yellow (+). Colour difference (ΔE) of SMPs (Eq.2) was
 159 assessed compared to the control regular powder similar to that as described by Kelleher et al.
 160 (2020), where a $\Delta E > 2.3$ is a noticeable difference in colour (Mokrzycki, and Tatol, 2011).

161

$$162 \quad \Delta E = \sqrt{(L_2^* - L_1^*)^2 + (a_2^* - a_1^*)^2 + (b_2^* - b_1^*)^2} \quad Eq. 2$$

163

164 2.7. Friability, Flowability, and compressibility of powders

165 Friability of powder particles was measured by compressing the particles under air
 166 pressure of 50 and 400 kPa, using a Malvern Mastersizer 3000. The particle size distribution
 167 value (D_{50}) was used to calculate the friability of powder particles using Eq. 3, as described by
 168 Schuck et al. (2012).

$$169 \quad Friability = \left(\frac{d(50) 50 \text{ kPa} - d(50) 400 \text{ kPa}}{d(50) 50 \text{ kPa}} \right) \times 100 \quad Eq. 3$$

170

171 The span index (SI) at the minimum (0 bar) and maximum (4 bar) air pressure were
 172 determined using Eq. 4. The SI describes the entire particle size range and it provides an in-
 173 depth view of the change in size distribution as a function of air pressure.

$$174 \quad Span \text{ index } (SI) = \frac{(D90 - D10)}{D50} \quad Eq. 4$$

175 The flowability of powders were analysed using a powder flow tester (PFT; Brookfield
 176 Engineering Laboratories, Inc., Middleboro, MA, USA). The powder samples were filled into
 177 the aluminium trough (volume of 230 cm³, 15.2 cm internal diameter) until a dome-shaped
 178 volume of powder was obtained using a curved blade. A standard flow function (FF) test was
 179 carried out over five normal stresses (1.0, 1.9, 2.9, 3.9, and 4.8 kPa) and three over-
 180 consolidation stresses at each normal stress. The flow function (FF) graph was obtained by

181 plotting the major principal consolidating stress (MPCS) as a function of unconfined failure
182 strength (UFS). The index of flow was calculated from the inverse of the slope of the FF curve.
183 The index corresponds to the strength that has to be overcome for a powder to flow when
184 consolidated. The compressibility index (CI) was calculated using Eq.5, where P_t and P_b are the
185 tapped and loose bulk density, respectively.

186

$$187 \quad CI = \frac{p_t - p_b}{p_b} \times 100 \quad Eq. 5$$

188 2.8. *Wettability and solubility*

189 The wettability of powders were analysed using the GAE Niro A5a method. Skim
190 milk powders were dispersed at 10%, w/w, in deionised water at 24 °C and the time recorded
191 for complete powder wetting. The powder was dispersed in deionised water using an overhead
192 mixer (IKA™ EUROSTAR 40 Digital S 2, UK) fitted with three propeller blades for 5 min at
193 a speed of 350 RPM. Aliquots (35 mL) were then centrifuged at 3000 × g for 10 min and the
194 total solids of the supernatant determined using a CEM moisture analyser (CEM Smart
195 System5™, 3100 Smith Farm Road, Matthews, NC, USA). Powder solubility is expressed as
196 a percentage of the solids content in the supernatant as a function of the initial dry matter prior
197 to centrifugation.

198

199 2.9. *Water sorption isotherms*

200 Water sorption analysis of skim milk powders was assessed as a function of relative
201 water activity (a_w) after equilibration for 168 h at 25 ± 2 °C. Saturated salts of LiCl, CH₃COOK,
202 MgCl₂, K₂CO₃, Mg (NO₃)₂, NaNO₂ and NaCl were used to control the relative vapour pressure
203 (RVP) within vacuumed desiccators at 0, 11.4, 23.1, 33.2, 44.1, 54.5, 65.6, and 76.1%,
204 respectively. Prior to equilibrating powders at different RVPs, the free moisture in the powders
205 was removed using a vacuum oven set at 60 °C for 24 h. The weight change of powders due to

206 water absorption was recorded at 0, 2, 6, 9, 12 and 24 h and then at 24 h intervals for 168 h.
207 The quantitative sorption values was reported as equilibrated water content (g) per of 100 g of
208 dry solids. The Guggenheim-Anderson de Boer (GAB) model was fitted to experimental data
209 using equations 6, 7 and 8. A second order polynomial function (Eq.7) was fitted to the a_w and
210 water content (m) (mass g/100g of dry solids) plot and was then used to calculate the constants
211 α , β , and γ . The predicted rate of water sorption was obtained using the GAB (Eq.8) model.

212

$$213 \quad \text{Rate of Sorption} = \frac{\text{Absorbed water}}{\text{time}} \text{ (\%/ h)} \quad \text{Eq. 6}$$

214

$$215 \quad \frac{aw}{m} = \alpha a_w^2 + \beta a_w + \gamma \quad \text{Eq. 7}$$

216

$$217 \quad GAB (mm) = \frac{a_w}{a_w^2 + \beta a_w + \gamma} \quad \text{Eq. 8}$$

218

219 2.10. Glass transition and mechanical properties

220 The onset of glass transition (T_g) of powders equilibrated for 168 h at 25 ± 2 °C at 0.11,
221 0.23, 0.33 and 0.44 a_w was measured using a Q2000 differential scanning calorimeter (DSC,
222 TA Instruments, Crawley, UK). Samples (18 mg) were filled in to DSC pans and hermetically
223 sealed with Tzero hermetic lids. Firstly, samples were approximately heated to 40 °C below to
224 40 °C above the T_g at a rate of 5 °C/min, followed by cooling before a second heating to 50 °C
225 below the T_g at 10 °C/min, and finally heating at 5 °C/min to 50 °C above the T_g point. An
226 empty pan was used as a reference. The T_g was determined from the second cycle of heating
227 and taken as the point of the onset of the endothermic baseline shift.

228 The mechanical structural strength properties of skim milk powders, i.e., storage
229 modulus, loss modulus and tan delta (Tan δ) was analysed using a Dynamic Mechanical

230 Analyser (DMA Q800, TA Instruments, Crawley, UK) across multiple oscillating frequencies
231 of 0.1, 0.5, 1.0, 2.0, and 5 Hz. Samples were subjected to temperatures ~ 50 °C below the T_g at
232 a cooling rate of 2 °C/min, followed by a heating step to ~ 60 °C above T_g at a heating rate of 2
233 °C/min. The samples were mounted in a dual cantilever clamp during the measurement. The
234 peak of the loss modulus was used to determine the α -relaxation temperature (T_α) of powders
235 (Li et al., 2015). The loss modulus (E'') data were captured using TA universal analysis
236 software version 5.1.2 (TA Instruments). The plasticizing effect of water and changes in
237 viscoelastic properties of powder particles equilibrated at a_w values of 0, 0.23, 0.33 and 0.44
238 were determined as $T_\alpha - T_g$.

239

240 *2.11. Statistical analysis*

241 Results presented are the mean values \pm standard deviation and were analysed by analysis
242 of variance (ANOVA) using Statistical analysis software general linear model (SAS GLM)
243 procedure. Significant ($P < 0.05$) means were separated using the Tukey's test.

244

245 **3. Results and Discussion**

246 *3.1. Bulk and particle surface composition of skim milk powders*

247 The bulk powder composition across all powders showed similar levels of protein,
248 lactose and ash (Table 1); however, lecithin addition resulted in powders containing $\sim 2.0\%$
249 (w/w) fat, due to the addition of phospholipids to the liquid concentrate in the case of RL-SMP
250 and sprayed directly on to the powder within the external fluid bed in the case of AL-SMP
251 (Table 1).

252 Compared to bulk powder composition, the composition of the particle surface (Table
253 1) was dominated by protein in non-lecithinated powders, although the fat content was over-
254 represented on these powders compared to their bulk composition. The over-representation of

255 fat coverage on the surface of particles has been reported by several previous studies (Kim, et
256 al., 2002; Shrestha, et al., 2007). The surface fat coverage of R-SMP and A-SMP (Table 1) are
257 comparable with previous studies by O'Donoghue et al. (2019) and Nijdam and Langrish,
258 (2006) which showed a surface fat coverage on SMP's of 9.6% and 8.0%, respectively. There
259 was however, a higher level of surface lactose for A-SMP compared to R-SMP. This may be a
260 result from the collision of fines and droplets within the wet zone of the atomization cloud,
261 where hygroscopic amorphous lactose may diffuse more readily to the particle surface. A
262 previous study by Nijdam and Langrish (2006) indicated higher drying temperatures might
263 favour lactose migration to the particle surface, forming a surface skin; however no changes
264 were made to drying temperatures in the current study. Lecithination, as might be expected,
265 increased the proportion of fat on the surface of RL-SMP and AL-SMP particles, with ~34.8
266 and 73.5% of the total surface area represented by fat, respectively (Table 1). The lower level
267 of fat on the surface of RL-SMP particles is due to the addition of lecithin in to the skim milk
268 concentrate prior to drying, as opposed to the direct spraying of lecithin on to particles within
269 the fluid bed in the case of AL-SMP. The fact that lecithin could not be sprayed on to the
270 regular powder may be seen as a limitation of the study, but it is common that the fines for non-
271 agglomerated powders would bypass the lecithin spray nozzle.

272

273 3.2. *Particle, loose and bulk density of skim milk powders*

274 The effect of agglomeration and lecithination on powder particle density and powder
275 bulk density are shown in Table 2. Interestingly, particle density (P_p) was similar for all
276 powders ranging from 1.28 to 1.33 g/cm³, however, the tapped (500) bulk density (P_{t-500}) for
277 non-agglomerated powders was significantly higher at 0.76 g/cm³, compared to A- and AL-
278 SMP at 0.65 and 0.58 g/cm³, respectively. A study by Pugliese et al. in 2017, where a selection
279 of commercial skim milk and whole milk powders were characterized for physical properties,

280 showed that of the seven SMP samples examined six had a tapped bulk density $> 0.70 \text{ g/cm}^3$.
281 In the present study there was no difference between R- and RL-SMP, as might be expected,
282 with the lecithin added to the liquid skim milk prior to drying (Table 2); however, lectionation
283 of the agglomerated SMP caused a reduction in the tapped bulk density (0.58 g/cm^3), compared
284 to A-SMP (0.65 g/cm^3). This also corresponded with a significantly higher level of interstitial
285 air ($103 \text{ mL}/100\text{g}$), compared to all other powders. Chever et al. (2017) reported a tapped bulk
286 density value of 0.50 g/cm^3 for agglomerated whole milk powder compared to non-
287 agglomerated powders 0.72 g/cm^3 . Ultimately, it seems that a combination of agglomeration
288 and lecithination is required to significantly reduce SMP bulk density and should expand its
289 range of applications, particularly as a dry blendable ingredient.

290

291 3.3. Particle size, friability, flowability and colour of skim milk powders

292 Particle size values and distributions of SMPs are shown in Table 3 and Fig. 1A,
293 respectively, and clearly highlight the significant effect agglomeration had on creating larger
294 particles, whereby the D_{90} size values increased from $130 \mu\text{m}$ for R-SMP to $201 \mu\text{m}$ for A-
295 SMP. This is also in-line with the significant increase in porosity of agglomerated particles as
296 shown in Table 2. Lecithination seemed to have a small but significant effect on producing
297 larger particles at $141 \mu\text{m}$ for RL-SMP and $210 \mu\text{m}$ for AL-SMP, compared to their non-
298 lecithinated counterparts. SEM images of powders are shown in Fig. 2, with noticeably larger
299 particles displayed for agglomerated powders. The particle size values of R-SMP reported in
300 this study are similar to the values reported by Pugliese et al. (2017) for a number of
301 commercial SMP samples. One concern with creating agglomerated SMP is its susceptibility
302 to powder breakdown and as shown in Fig. 1B where the air pressure was increased during
303 particle size measurements the specific surface area (SSA) increased, indicative of powder
304 breakdown (i.e., SSA is inversely proportion to particle size). However, the rate of increase in

305 SSA as a function of air pressure was similar for non-agglomerated and agglomerated powders
306 (Fig. 1B), which might be unexpected given the larger and more porous particles in
307 agglomerated powders. Although, data shown in Table 4 highlight that the friability of
308 agglomerated powders was significantly higher than their non-agglomerated counterparts (i.e.,
309 ~32% compared to ~22% friability, respectively).

310 The flowability index of powders are shown in Table 4, with lecithin addition reducing the
311 flow index for both RL-SMP and AL-SMP compared to their corresponding counterparts. The
312 reduced flowability of powders with significant surface fat coverage has been reported
313 previously by Nijdam and Langrish (2006), and may be due to reduced inter-particle mobility
314 caused by weak particle bridging. Interestingly, powder colour was more affected by
315 agglomeration than the addition of lecithin (Table 3) which was unexpected given that lecithin
316 was sprayed on to AL-SMP particles directly. However, the colour difference between regular
317 and agglomerated dairy powders has been reported before and may be due to the differences in
318 interstitial and occluded air. McSweeney et al. (2021) also reported a significant colour change
319 in agglomerated milk protein concentrate (MCP) powders compared to regular MPC powder.

320

321 3.4. *Wettability and solubility of skim milk powder*

322 Powder wettability and solubility data are shown in Table 4, with one main significant
323 difference between the powders. AL-SMP had a wettability of 8.4 s, compared to all other
324 powders which were > 300 s. Similarly, McSweeney et al (2020) showed that wettability was
325 not completed after 20 min at 25 °C for a non-agglomerated, non-lecithinated milk protein
326 system (containing ~40%, w/w, protein). Interestingly, the wettability of RL-SMP was no
327 better than that of non-lecithinated powders. This highlights that although the powder contains
328 the same level of lecithin as AL-SMP and had a significantly higher proportion of surface fat
329 coverage on its particles compared to non-lecithinated powders (Table 1), it had no effect on

330 the wettability (Table 4). This may be due to the fact that the particle surface of RL-SMP was
331 covered by 38.5% protein as opposed to only 8.5% protein on AL-SMP particles (Table 1) and
332 thus the higher level of hydrophobic protein may hinder water penetration. The solubility of all
333 SMPs were similar (Table 4) and highlights that during high shear mixing the differences in
334 particle structure can be negated although the significant improvement in wettability of AL-
335 SMP should provide major benefits for dry blending applications.

336

337 3.5. *Water sorption, glass transition temperature and mechanical properties*

338 Water sorption profiles and fitted GAB models for SMPs are shown in Fig. 3 and 4,
339 respectively. Water sorption of powders increased with increase in a_w until 0.44. However, at
340 a a_w of 0.54 and 0.65 there was a significant decrease in absorbed water, due to a change from
341 amorphous to crystalline lactose, with a concomitant release of absorbed water. For prediction
342 of the sorption isotherm using the GAB model, the linear region (i.e. 0 - 0.44 a_w) obtained from
343 the experimental raw data was used. Interestingly, the presence of lecithin on the surface of
344 AL-SMP particles had no effect on water sorption kinetics (Fig. 4). Although, many studies
345 have previously (Lin et al., 2005; Murrieta-Pazos, et al., 2011) shown that the water sorption
346 kinetics of SMP and fat containing whole milk powder are essentially the same when calculated
347 on a non-fat basis. This is important in that lecithination did not enhance the rate of water
348 sorption during storage and therefore did not escalate the rate of powder quality deterioration.
349 Also, the larger powder particle size of A-SMP and AL-SMP did not have a distinct effect on
350 the water sorption profiles (Fig. 3 and 4). Previously, O'Donoghue et al. (2019) suggested that
351 it is the bulk composition that mainly determines powder water sorption properties rather than
352 powder particle size.

353 The glass transition temperature (T_g) and stickiness of SMPs, measured as a function of a_w ,
354 are shown in Table 5 and Fig. 5, respectively. The T_g of all powders decreased with increasing

355 a_w , and similar to water sorption profiles no differences were observed between any of the
356 SMPs, with T_g values decreasing from around 109-106 °C at 0 a_w down to 10-11 °C at 0.44 a_w
357 (Table 5). Shrestha et al. (2007) previously showed that changing the composition of SMP by
358 adding different levels of lactose did not affect the glass transition temperature of the powders,
359 but only had an effect if the protein content of SMP was significantly decreased, which was
360 not the case in the current study as shown in Table 1.

361 The α -relaxation (T_α) and stickiness temperatures ($T_\alpha - T_g$) of SMPs are shown in Fig. 5 and
362 highlight that for all powders the T_α decreased from with increasing a_w . Meanwhile, the
363 stickiness temperature steadily increased as a_w increased from 0 to 0.33, after which there was
364 a decline in temperature at 0.44 a_w . This decline in stickiness temperature is associated mainly
365 with the plasticization effect of moisture on lactose (Roos, 2008). At a temperature higher than
366 T_g , the molecules will change from an amorphous solid-like state to a liquid-like, rubbery state
367 and is one of the main indicators of stickiness and caking in milk powders (Roos and Karel,
368 1993). Also, both the T_α and $T_\alpha - T_g$ values were higher at a higher oscillation frequency (i.e.,
369 increasing from 0.1 to 5 Hz; Fig. 5). The interdependence between T_α and frequency has been
370 shown previously by a number of studies (Talja & Roos, 2001; O'Donoghue et al., 2022).
371 Ultimately and similar to water sorption data, all powders behaved alike, again demonstrating
372 that agglomeration and lecithin addition had little impact on powder storage properties.

373

374 **4. Conclusions and prospects**

375 Skim milk powder (SMP) functionality can be readily altered by modifying the physical
376 powder particle properties and morphology. To improve wettability, lecithin must be sprayed
377 directly on to powder particles, while, lecithin addition to the concentrate prior to drying
378 offered no benefit. Similarly, agglomeration alone did not improve wettability of SMP but did
379 cause a reduction in tapped bulk density. This should allow SMP to be suitable for dry blending

380 purposes where high bulk density of regular SMP is often the limiting factor in its use, while
381 solubility may be improved through a combination of both agglomeration and lecithination.
382 One important point to be considered when agglomerating SMP is the susceptibility of the
383 resultant powder to friability and breakdown, which under certain powder transport or handling
384 conditions may be an issue. Future work on this area could revolve around producing more
385 robust agglomerates less prone to powder breakdown during pneumatic conveying and dry
386 blending.

387

388 **Acknowledgment**

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392

393 **Author's Contributions**

394 Yonas Hailu: Conceptualization, Investigation, Methodology, Analysis, Writing of original
395 draft; Valentyn A. Maidannyk, Methodology, Analysis; Eoin G. Murphy: Methodology,
396 Investigations, Noel A. McCarthy, Conceptualization, Writing, Reviewing, Editing, Funding
397 acquisition

398

399 **Declaration for conflict of interest**

400 The authors declare that they have no known competing personal and/or financial interest that
401 could have influence on the work reported in this paper.

402

403

404

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529

530 *List of Tables*

531 **Table 1.** Bulk powder composition (% , w/w) and powder particle surface coverage (%) of regular (R), regular lecithinated (RL), agglomerated
 532 (A) and agglomerated lecithinated (AL) skim milk powder (SMP).

Samples	Powder composition					Powder particle surface coverage		
	Protein	Fat	Ash	Lactose	Moisture	Protein	Fat	Lactose
R-SMP	37.8±0.1	0.79±0.0 ^b	8.31±0.1	50.3±0.1	2.85±0.1	55.9±0.5 ^a	6.8±0.5 ^c	35.3±0.2 ^b
RL-SMP	37.3±0.1	1.99±0.0 ^a	8.24±0.0	49.6±0.3	2.85±0.2	38.5±1.1 ^c	34.8±0.8 ^b	25.2±1.6 ^c
A-SMP	37.9±0.0	0.81±0.0 ^b	8.34±0.0	49.5±0.1	3.43±0.1	50.4±1.1 ^b	6.9±0.1 ^c	41.1±1.4 ^a
AL-SMP	37.6±0.0	2.03±0.0 ^a	8.24±0.1	49.4±0.0	2.77±0.0	8.5±0.5 ^d	73.5±1.0 ^a	16.6±0.5 ^d

533 Values are the mean of triplicate analysis for bulk composition and duplicate analysis for surface composition. Values within a column not sharing
 534 a common superscript letter differ significantly ($p < 0.05$).

535 **Table 2.** Physical properties of Regular (R), Regular lecithinated (RL), Agglomerated (A) and
 536 lecithinated agglomerated (AL) skim milk powder (SMP)

Samples	P _p	P _b	P _{t500}	V _{oa}	V _{ia}	CI	ε
	g/cm ³			mL/100g		(%)	
R-SMP	1.29±0.0 ^b	0.59±0.0 ^b	0.76±0.0 ^a	76.8±0.1 ^b	68.9±1.2 ^c	21.6±1.1 ^a	40.9±1.7 ^c
RL-SMP	1.29±0.0 ^{bc}	0.63±0.0 ^a	0.75±0.0 ^a	76.8±0.1 ^b	60.7±0.0 ^d	16.8±0.0 ^c	39.9±1.0 ^c
A-SMP	1.33±0.0 ^a	0.56±0.0 ^c	0.65±0.0 ^b	74.4±0.2 ^c	84.9±1.2 ^b	14.8±0.6 ^b	62.6±0.4 ^b
AL-SMP	1.28±0.0 ^c	0.44±0.0 ^d	0.58±0.0 ^c	77.3±0.1 ^a	103±2.1 ^a	24.8±1.1 ^a	94.6±2.4 ^a

537 Values are mean ± standard deviation of triplicate analysis. Values within a column not sharing
 538 a common superscript letter differ significantly ($p < 0.05$).

539 P_p- particle density,

540 P_b - loose bulk density,

541 P_{t500} - tapped (500 times) bulk density

542 CI - compressibility index

543 ε - porosity

544 **Table 3.** Physical properties of regular (R), regular lecithinated (RL), agglomerated (A) and agglomerated lecithinated (AL) skim milk powders.

Samples	D_{50}	D_{90}	$D_{4,3}$	Colour Index			ΔE
	(μm)			L^*	a^*	b^*	
R-SMP	64.5±0.07 ^d	130±0.7 ^d	72.3±0.1 ^d	94.1±0.1 ^a	-6.11±0.0 ^a	16.4±0.0 ^d	-
RL-SMP	71.5±0.7 ^c	141±0.8 ^c	79.7±0.8 ^c	93.7±0.0 ^b	-6.12±0.0 ^a	17.3±0.0 ^c	0.95
A-SMP	107±0.0 ^b	201±0.7 ^b	118±0.7 ^b	93.5±0.0 ^c	-6.8±0.0 ^c	18.8±0.0 ^b	2.59
AL-SMP	113±0.0 ^a	210±0.7 ^a	125±0.7 ^a	92.6±0.0 ^d	-6.34±0.0 ^b	19.0±0.0 ^a	2.99

545 Values are mean ± standard deviation of triplicate analysis. Values within a column not sharing a common superscript letter differ significantly (p
546 < 0.05).

547 ΔE - Colour difference in comparison to the regular skim milk powder.

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554 **Table 4.** Functional properties of regular (R), regular lecithinated (RL), agglomerated (Agg) and agglomerated lecithinated (AL) skim milk
 555 powders (SMP).

Samples	Wettability	Solubility	Flow index	JC	Friability	SI 0 bar	SI 4 bar
	s	%			%		
R-SMP	>300±0.0	97.9±0.1 ^b	6.93±0.1 ^b	Easy flow	22.9±0.1 ^b	1.60±0.0 ^a	1.56±0.0 ^a
RL-SMP	>300±0.0	98.7±0.1 ^{ab}	4.59±0.1 ^c	Easy flow	22.2±0.6 ^b	1.54±0.0 ^b	1.52±0.0 ^b
A-SMP	>300±0.0	99.2±0.1 ^a	8.30±0.4 ^a	Easy flow	32.7±0.1 ^a	1.39±0.0 ^c	1.40±0.0 ^d
AL-SMP	8.94±0.1	99.3±0.0 ^a	2.90±0.1 ^d	Cohesive	32.6±0.1 ^a	1.36±0.0 ^c	1.46±0.0 ^c

556 Values are mean ± standard deviation of triplicate analysis. Values within a column not sharing a common superscript letter differ significantly (*p*
 557 < 0.05).

558 JC - Junike classification

559 SI - Span index

560 **Table 5.** Glass transition (T_g) temperature of regular (R), regular lecithinated (RL),
 561 agglomerated (A) and agglomerated lecithinated (AL) skim milk powders equilibrated at
 562 different water activity values at 25 °C for 168 h.

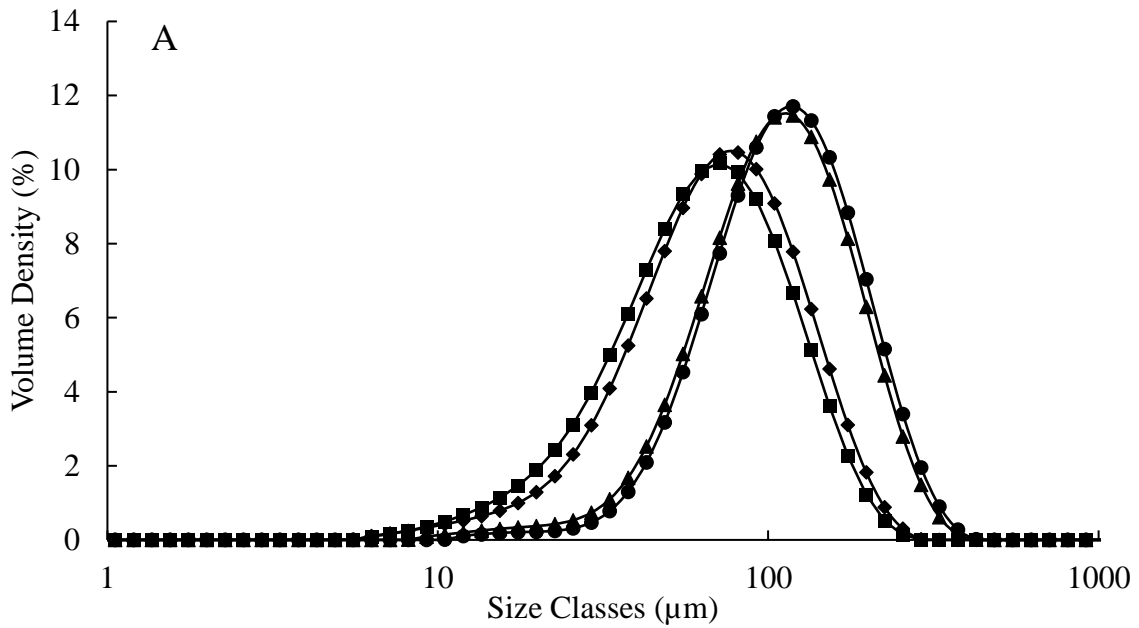
Sample	Glass transition temperature (°C)				
	0 a_w	0.11 a_w	0.23 a_w	0.33 a_w	0.44 a_w
R-SMP	106±1.0	58.0±3.3	43.0±0.3	28.0±0.7	11.5±0.7
RL-SMP	109±0.4	59.9±1.8	46.5±1.0	30.8±1.6	11.4±1.2
A-SMP	106±0.6	60.8±2.5	44.9±0.3	31.0±1.1	11.4±0.9
AL-SMP	106±0.2	60.6±3.0	45.3±1.1	30.0±0.8	10.3±0.6

563 Values are mean ± standard deviation of duplicate analysis.

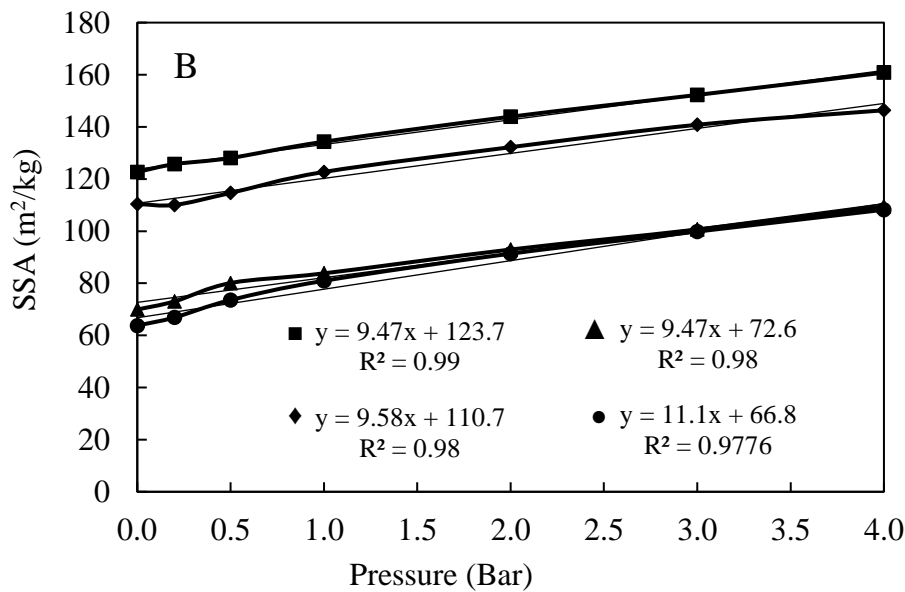
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565 **List of Figures**

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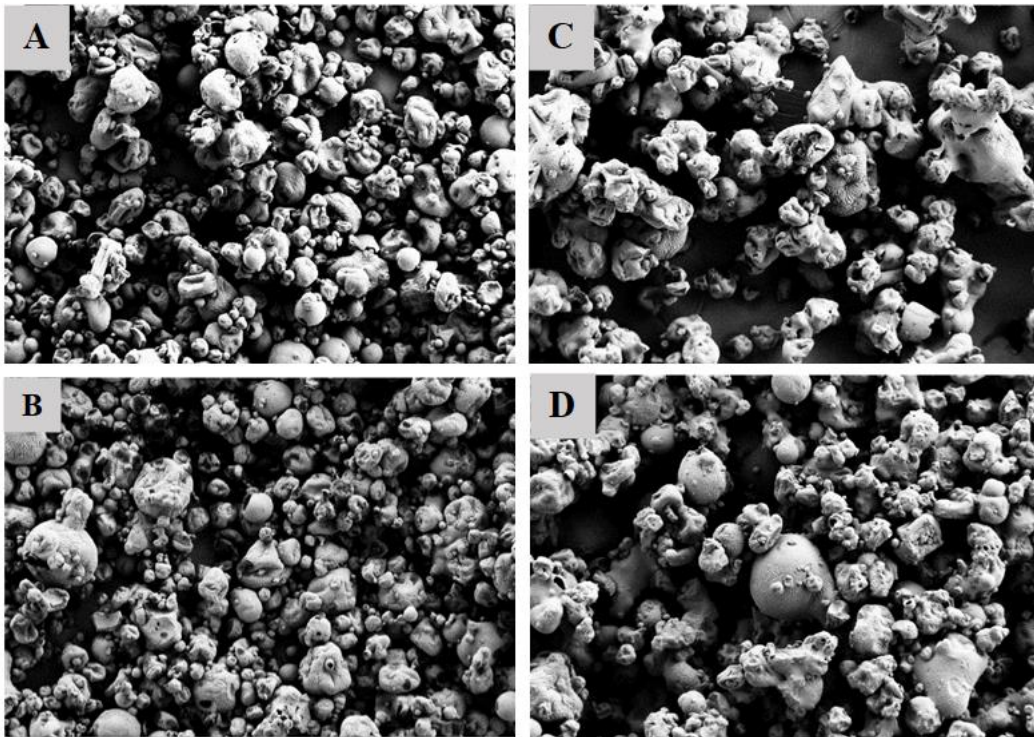
568

569 **Figure 1.** Particle size distribution (A) and specific surface area as a function of bar pressure

570 (B) of regular (■), regular lecithinated (◆), agglomerated (▲) and agglomerated lecithinated

571 (●) skim milk powder.

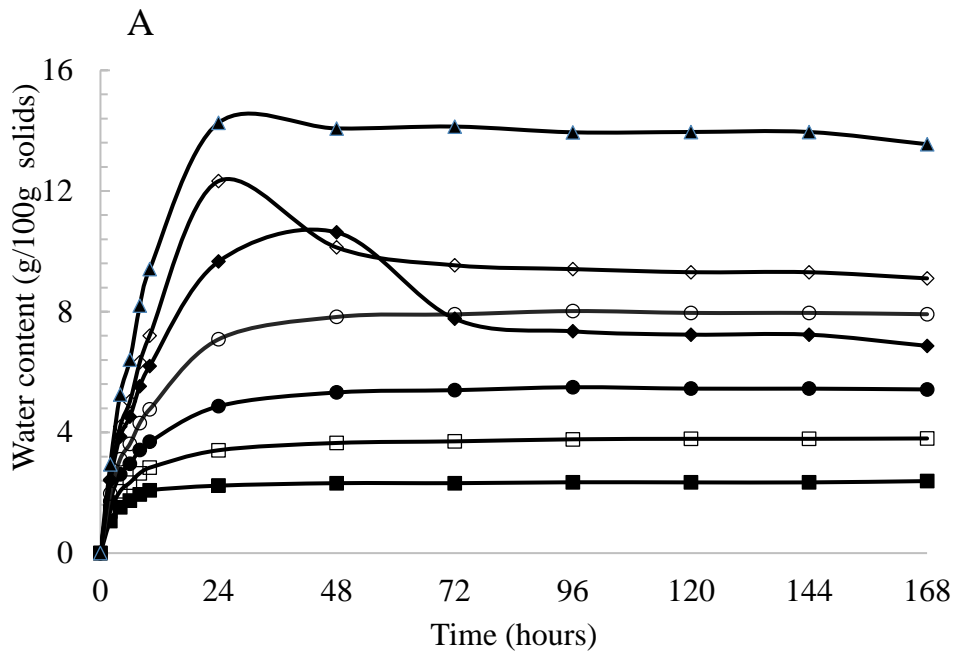
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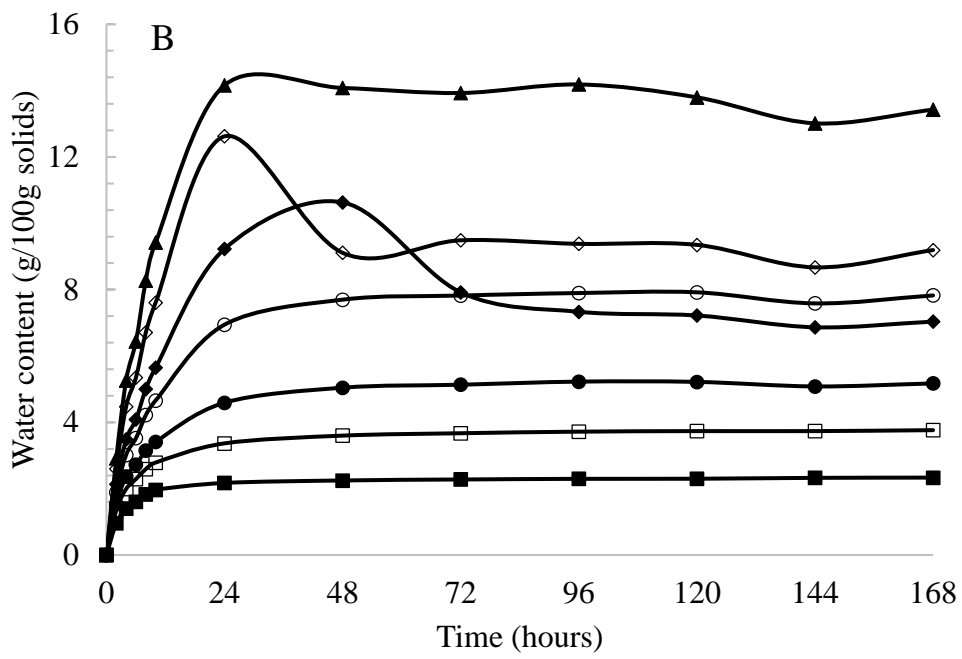
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575 **Figure 2.** Scanning electron micrographs (500× magnification) of regular (A), regular
576 lecithinated (B), agglomerated (C) and agglomerated lecithinated (D) skim milk powders.

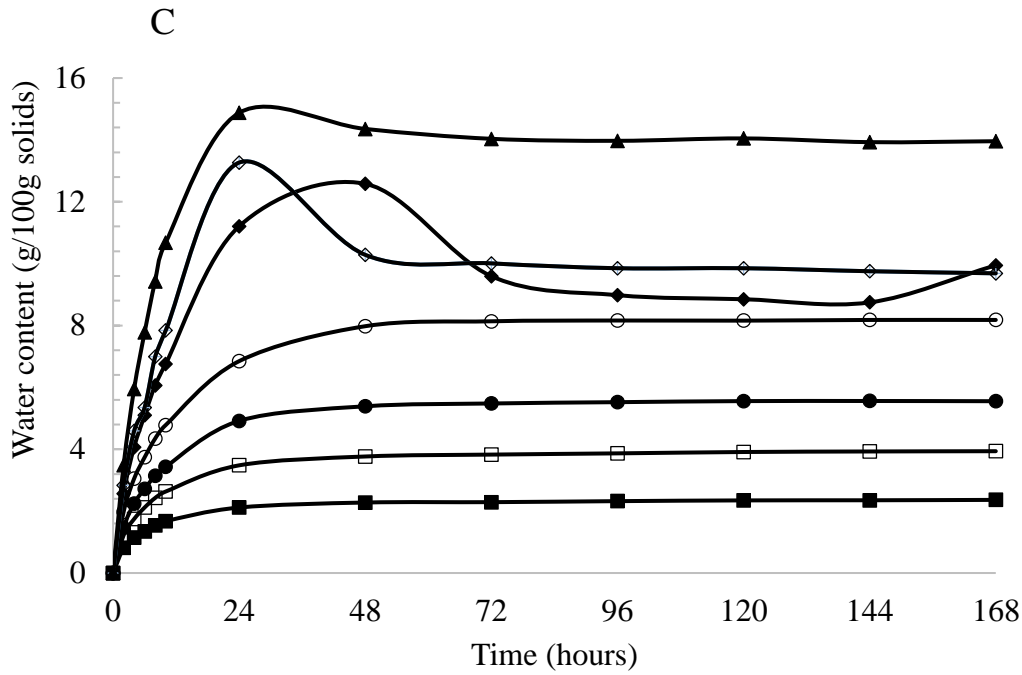
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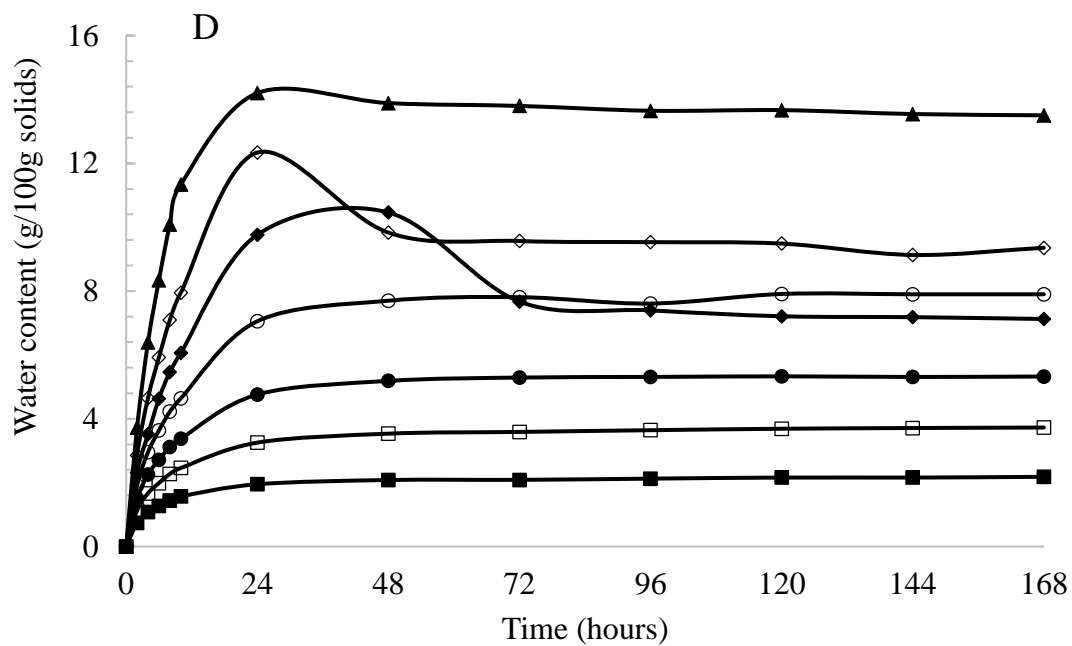
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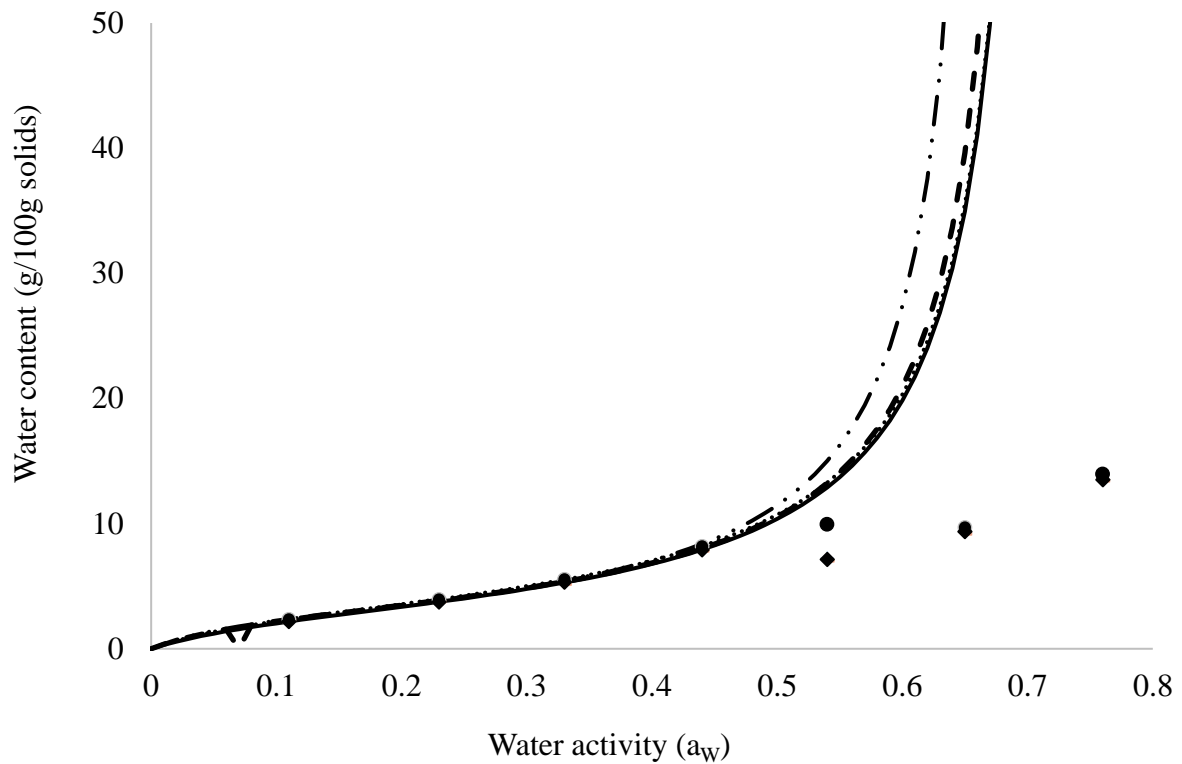


583

584 **Figure 3.** Water Sorption profiles of regular (A), regular lecithinated (B), agglomerated (C)

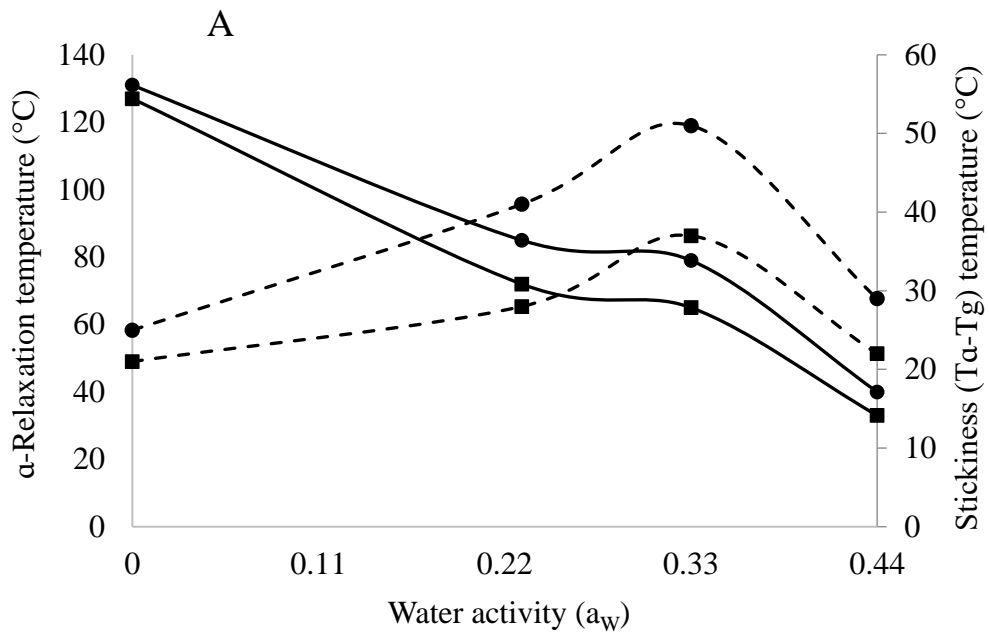
585 and agglomerated lecithinated (D) skim milk powders equilibrated at water activity values of

586 0.11 (■), 0.23 (□), 0.33 (●), 0.44 (○), 0.55 (◆), 0.65 (◇) and 0.76 (▲) for 168 h at 25±2 °C.

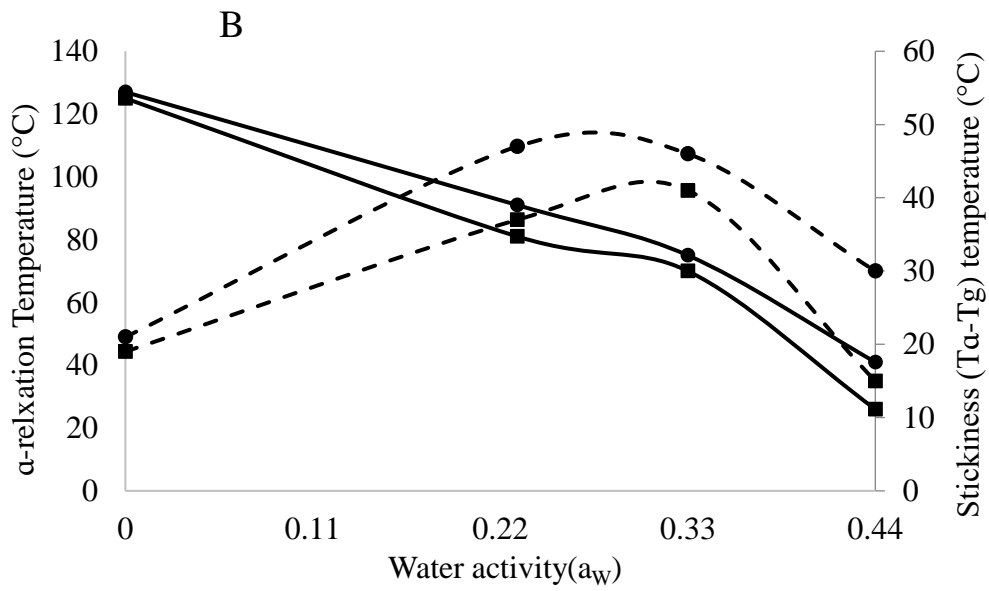


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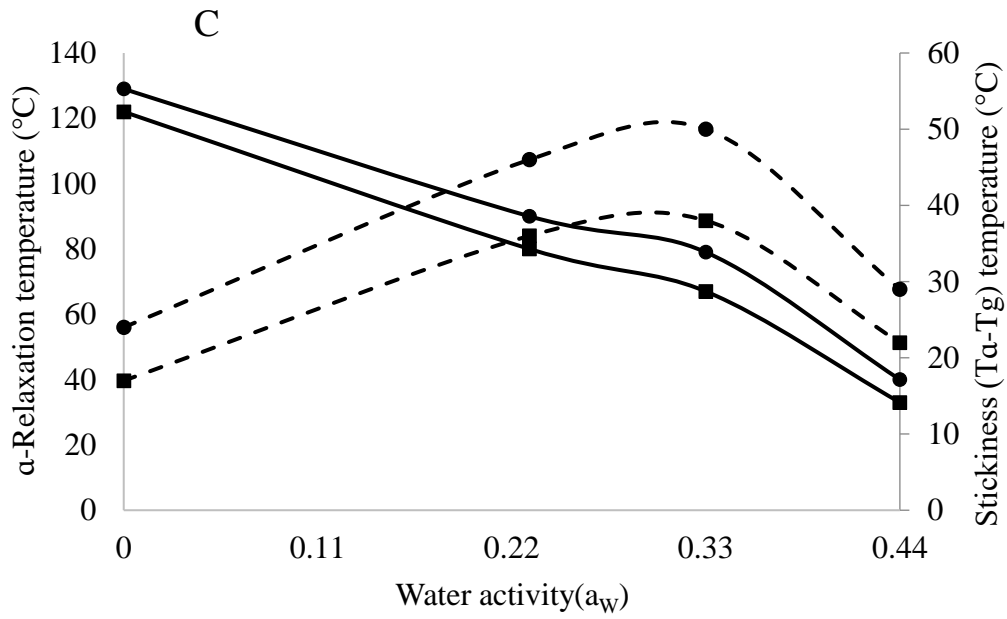
588 **Figure 4:** Water sorption isotherms of regular (— — —), regular lecithinated (—••—••),
 589 agglomerated (•••), and agglomerated lecithinated (——) skim milk powder equilibrated for
 590 168 h at 25 ± 2 °C. Experimental data fitted to Guggenheim Anderson deBoer model of
 591 prediction are in data point Regular (■), Regular lecithinated (◆), Agglomerated (●), and
 592 Agglomerated lecithinated (▲).



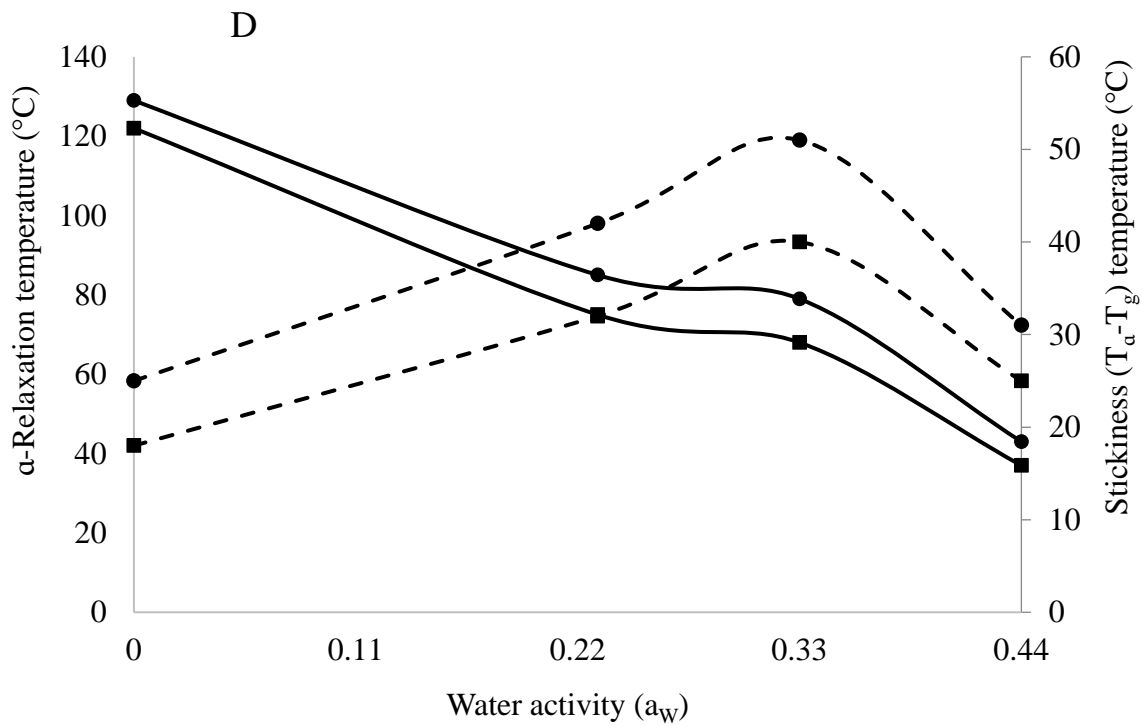
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597 **Figure 5.** α -relaxation temperature (solid lines) and stickiness temperature ($T_\alpha - T_g$) (dashed
 598 lines) measured at an oscillation frequency of 0.1 (■) and 5 Hz (●) for regular (A), regular
 599 lecithinated (B), agglomerated (C) and agglomerated lecithinated (C) skim milk powders
 600 after equilibration for 168 h at water activity values of 0, 0.22, 0.33 and 0.44 (25 ± 2 °C).

601