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Tristan Fournaise, Jennifer Burgain, Carole Perroud-Thomassin, Jérémy Petit. Impact of the whey protein/casein ratio on the reconstitution and flow properties of spray-dried dairy protein powders. Powder Technology, 2021, 391, pp.275-281. 10.1016/j.powtec.2021.06.026 . hal-03330382

**HAL Id: hal-03330382**

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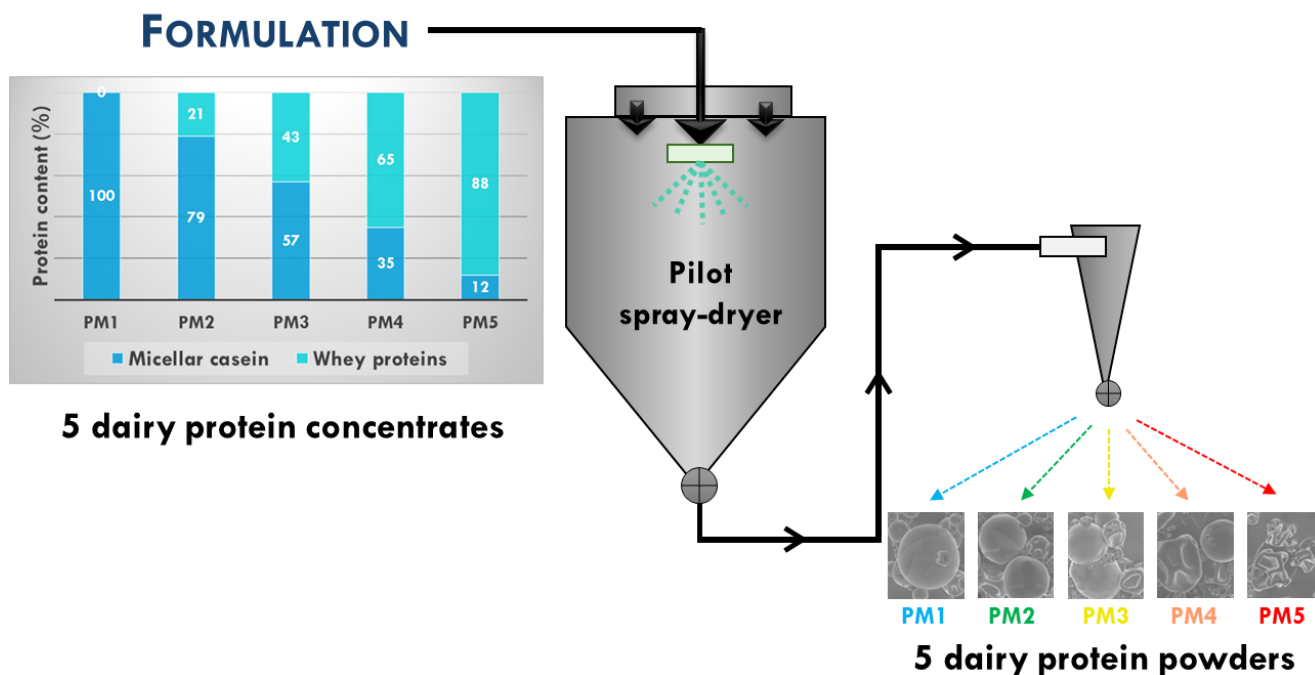
# Powder Technology

## Impact of the whey protein/casein ratio on the reconstitution and flow properties of spray-dried dairy protein powders

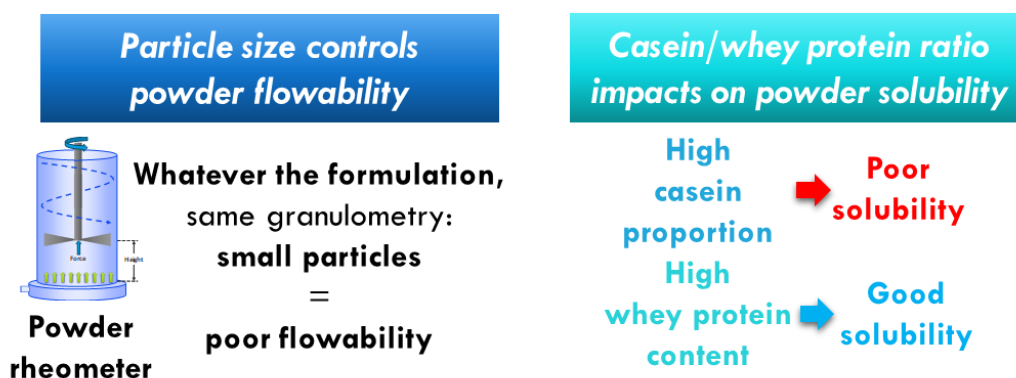
--Manuscript Draft--

<b>Manuscript Number:</b>	POWTEC-D-21-00864R1
<b>Article Type:</b>	Research Paper
<b>Keywords:</b>	Dairy powders; Formulation; protein composition; Spray-drying; reconstitutability; Flowability
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<b>Abstract:</b>	<p>This study focuses on the impact of the whey protein/casein ratio on the physicochemical and functional characteristics of dairy protein powders spray-dried at 200 °C inlet and 85 °C outlet air temperatures. Physicochemical properties of powders were characterized to explain their reconstitutability and flowability.</p> <p>Solubility of dairy protein powders linearly varied with the whey protein/casein ratio. Standard wettability and dispersibility indices were not relevant to evaluate the reconstitutability of powders produced at pilot scale, likely owing to the low median particle size of investigated powders.</p> <p>The flowability of powders was poor due to a high level of interparticular cohesion, which derived from their small particle size. The impact of protein composition on flowability was not statistically significant. The small particle size, leading to poor flow properties, overwhelmed the impact of protein composition, but a slight improvement of flowability and a reduction of cohesion observed for higher proportions of whey proteins</p>

## GRAPHICAL ABSTRACT



## TECHNOFUNCTIONAL PROPERTIES



## ABSTRACT

This study focuses on the impact of the whey protein/casein ratio (100/0, 79/21, 57/43, 35/65, and 12/88) on the physicochemical and functional characteristics of dairy protein powders. Spray-drying was performed in the following conditions: 200 °C inlet air temperature and 85 °C outlet air temperature. Physicochemical properties (particle size distribution, shape factors, moisture content, and water activity) of spray-dried powders were characterized to explain their reconstitutability, on the basis of the wettability, dispersibility, solubility indices, and their flowability, evaluated with the FT4 powder rheometer.

Reconstitutability analyses revealed that solubility of dairy protein powders linearly varied with the whey protein/casein ratio. It was also highlighted that wettability and dispersibility determined with the standard methods were not relevant to evaluate the reconstitutability of investigated powders: in fact, all studied powders were non-wettable and non-dispersible according to the milk powder standards. This was likely to result from the low median particle size (between 10 and 30  $\mu\text{m}$ ) of powders produced by spray-drying at pilot-scale.

The flowability of powders was evaluated as poor due to a high level of interparticular cohesion, which mainly derived from their small median particle size. The impact of the protein composition on the flowability was not statistically significant, despite a shape modification reflected by a decrease in particle sphericity upon casein proportion increase. The small particle size, leading to poor flow properties, overwhelmed the impact the whey protein/casein ratio, but a slight improvement of the flowability and a reduction of cohesion was induced by higher proportions of whey proteins.

Reviewer 1: This study focuses on the impact of the whey protein/casein ratio on the physicochemical and functional characteristics of dairy protein powders, which has been widely investigated.

The novelty of this paper can be improved if the spray-dried samples are further investigated from a chemical perspective.

It is true that the link between physicochemical properties and functionalities of powdered whey proteins and caseins has been widely investigated, but the novelty of this study comes from the focus on the impact of formulation (including chemical composition but also the preparation protocol of the concentrate to be spray-dried). The study presented in this manuscript was a part of a larger study who dealt with the impact of formulation (pretreatment of concentrates prior to spray-drying, lipid content, protein content, and whey proteins/caseins ratio) on physicochemical and functional properties of spray-dried dairy powders. The part of this larger study dealing with concentrate pretreatment (air incorporation by stirring with a blade usually used for foaming purposes) and lipid content has already been published in Powder Technology (<https://doi.org/10.1016/j.powtec.2020.05.085>).

1. DSC, XRD, XPS and Raman investigations are suggested to reveal the mechanisms of the known phenomena at a chemical/molecular level.

DSC and XPS have already been carried out on the samples of the present study but showed no differences between samples (from DSC, it was observed that all samples were amorphous and their glass transition temperature was hardly measurable; from XPS, it was shown that their surface was enriched in lipids and proteins, as lipids tend to migrate to the droplet surface by buoyancy during the drying process, whereas proteins, being surface-active molecules, are generally found more concentrated at water/air interfaces (i.e. at the surface of droplets). Thus, it was decided not to present these analyzes in the manuscript.

DRX would not be more informative, as samples were amorphous. As for Raman spectroscopy, it could be useful to elucidate the structure of dairy proteins in produced samples, but the aim of the larger study was focused on physicochemical and functional properties of spray-dried dairy powders, not on the structure of individual particles constituting the powders.

2. Thermal stability effects are suggested to be studied for different whey protein/casein ratio.

This would be interesting, but the objective of the study was to find appropriate formulation conditions to improve reconstitutability and flowing properties of spray-dried dairy powders. Thermal stability, preservation ability, gelation properties, water-holding capacity, etc. are another functional properties of interest for spray-dried dairy powders, but they did not fall into the scope of the larger study.

3. The crumpled surfaces can be calculated with math models with diffusion efficiency.

The present study was intended to elucidate the most influent parameters (amongst formulation and powder physicochemical properties) on powder reconstitutability and flowing properties. For example, it was evidenced that particle shape can counteract the influence of particle size on powder flowing properties (i.e. samples richer in caseins had larger particles of more wrinkled shape and flowed more poorly). However, the mechanisms leading to particle shape were not the subject of the present study.

Reviewer 2: Manuscript Number: POWTEC-D-21-00864

The subject of research includes in this journal. Research work is interesting. In the paper titled "Impact of the whey protein/casein ratio on the reconstitution and flow properties of spray-dried dairy protein powders".

The authors in their article discuss a very important aspect study focuses on the impact of the whey protein/casein ratio on the physicochemical and functional characteristics of dairy protein powders spray-dried. Research work is interesting.

Thank you for your interest in our work. We hope that modifications performed following your comments will help in improving manuscript quality.

The paper needs minor revision because:

1. The quality of the language is insufficient. Have a native speaker or similar assist you.

The language level was improved by using a more sustained language when possible (cf. modifications in blue in revised manuscript lines 16-18, 20-23, 24, 27, 40, 43, 64, 131, 190, 212, 231, 248, 253, 255, 280 and 282-310).

2. The Introduction is quite random with broad and varying statements and not very well focused.

Indeed, the introduction was a little too vague, as we initially wanted to make the link with the first part of the study already published in Powder Technology (<https://doi.org/10.1016/j.powtec.2020.05.085>). A better focus on the subject of the study was carried out (cf. modifications in blue in revised manuscript lines 16-18, 20-23 and 28).

3. In my opinion, the final conclusions should also be drawn up as they are not as specific as the scope of work.



The final conclusion was too exhaustive, we have modified it to make it more concise and better focused (cf. modifications in blue in the manuscript lines 280 and 282-310).

4. Please standardise the number of decimal places in the tables. Please use a precision of at least two decimals places.

We initially chose to present results with 3 significant numbers, thus the number of decimals depended on the values. Thus, we followed reviewer #2's advice for table 3 (cf. modifications in blue). For tables 1 and 2, it was not possible to add more decimals, as data of table 1 came from supplier sheets which were not more accurate than presented values and data of table 2 were calculated from data of table 1. Moreover, one decimal was added to water activity results, as the accuracy of the analytical equipment was sufficient to allow this (cf. modifications in blue).

5. Please explain why the authors chose such a statistical analysis?

ANOVA and Tukey's HSD test are very classical statistical analysis methods: the first one allows identifying the presence of significant differences amongst sample results, whereas the second one permits to sort samples according their significant differences. Pearson's correlation coefficients are also rather classically used to link parameters between them: it permits to calculate the degree of linear correlation between two variables; in the present study, the degree of correlation between analyzed parameters was determined to help identifying the main physicochemical parameters influencing each analyzed functional properties.

6. Could an additional PCA analysis have been performed?

Yes, indeed, PCA could have been performed, and it would have given similar information as Pearson's correlation coefficients calculated for all pairs of investigated parameters (as PCA intends to correlate all investigated parameters in a single statistical analysis, whereas Pearson's

correlation coefficients are able to indicate the degree of correlation between two parameters only). We chose to discuss results with Pearson's correlation coefficients that we estimate easier to understand for readers than PCA.

7. Improve the readability of the Figure 3, 4.

The readability of figures 3 and 4 has been enhanced (cf. figures 3 and 4 in figures file).

8. References is incorrectly formatted number [17 - 21 and 31].

The reference 17 has been modified, but it's a French PhD so the title should be kept in French. Other concerned references are norms that were already correctly formatted: these references have already been published in Powder Technology with this format (<https://doi.org/10.1016/j.powtec.2020.05.085>).

# IMPACT OF THE WHEY PROTEIN/CASEIN RATIO ON THE RECONSTITUTION AND FLOW PROPERTIES OF SPRAY-DRIED DAIRY PROTEIN POWDERS

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## KEYWORDS

Dairy powders; formulation; protein composition; spray-drying; reconstitutability; flowability

## 1. INTRODUCTION

Technofunctional properties of dairy powders such as reconstitutability (wettability, swelling, dispersibility and solubility) and flowability are crucial for the agri-food industry and consumers.

Wettability is the ability of powders to immerse without stirring [1]. Powder wettability is sensitive to several parameters like particle size: smaller particles generally induce poorer wettability. ~~Powder surface composition also affects the wetting time: an increase in the proportion of surface lipids is deleterious for powder wettability due to higher hydrophobicity~~[2-4].

Dispersibility corresponds to the dissociation of particle agglomerates into single particles and, in a lesser extent, to the attrition of particle agglomerates and single particles. ~~For dispersibility indices lower than 95 %, the powder is considered as non-dispersible: for example, in the case of whole milk (containing 26 % lipids) for which the high proportion of surface lipids impairs~~

23 ~~interactions with water, the dispersibility index is 89.5%. Moreover,~~ The presence of caseins  
24 has been ~~identified~~ ~~known~~ to strongly reduce dairy powder dispersibility (indeed, the  
25 dispersibility index of casein powders is around 26 %) [1].

26 Solubility represents the ability of powders to dissolve into water, which corresponds to the  
27 ~~disappearance~~ ~~complete loss~~ of granular structure once the product is reconstituted [1]. Like  
28 wettability, solubility decreases with ~~lipids proportion~~ or particle size. In addition, it was  
29 evidenced that a high casein content is related to a poor solubility, whereas whey proteins  
30 promote dairy powder solubilization [5–7]. Powder flowability is highly linked to its  
31 physicochemical properties: large, smooth, and spherical particles having a low water content  
32 are known to have good flow properties [8–10]. As matters the influence of the whey  
33 protein/casein ratio, a high proportion of caseins is known to lead to wrinkled and dense  
34 particles, with poorer flow properties [11]. The flowability can be characterized in low and high  
35 stress conditions by a rotational powder rheometer such as the FT4 in order to obtain several  
36 flow parameters, especially basic flowability energy (BFE) and SE (specific energy) in low  
37 stress conditions or flow factor ( $ff$ ) and cohesion in high stress conditions [8,12–14].

38 The present study focuses on the impact of composition on reconstitutability and flowability of  
39 dairy protein powders. Investigated powders differed in protein composition (variable  
40 casein/whey protein ratio) and were produced by spray-drying at pilot scale. ~~Afterwards~~ ~~Then~~,  
41 the reconstitutability of spray-dried powders was evaluated through the wettability,  
42 dispersibility, and solubility indices classically employed in the dairy industry.  
43 ~~Moreover~~ ~~Besides~~, flowability of dairy protein powders was assessed through measurement of  
44 BFE, SE, compressibility, cohesion, and  $ff$  with the FT4 powder rheometer. Finally,  
45 physicochemical properties of spray-dried powders (particle size distribution, appearance of  
46 particle surface, water content, and water activity) were determined and related to powder  
47 reconstitution and flow behavior.

48

## 49 2. MATERIAL AND METHODS

### 50 2.1. Material

51 Whey proteins isolate (PRODIET 90 S) and micellar casein powder (PRODIET 87 B) were  
52 provided by Ingredia (Arras, France). Their proximate composition in wet basis was calculated  
53 from technical sheets (Table 1).

54 *Table 1. Proximate composition expressed on wet basis of whey protein isolate and phosphocaseinate powders*  
55 *employed for the preparation of dairy concentrates to be spray-dried.*

### 56 2.2. Preparation of milk concentrates

57 Five concentrates differing in protein composition were produced from 100 % whey protein  
58 isolate (protein mixture 1, PM1), 75 % whey protein isolate and 25 % casein powder (protein  
59 mixture 2, PM2), 50 % whey protein isolate and 50 % casein powder (protein mixture 3, PM3),  
60 25 % whey protein isolate and 75 % casein powder (protein mixture 4, PM4), and 100 % casein  
61 powder (protein mixture 5, PM5). Concentrates were prepared by reconstituting adequate  
62 amounts of powders in distilled water at 15 % (w/w) dry extract under mechanical stirring (ZR  
63 2102 control, Heidolph, Germany) between 200 and 500 rpm with a 4-bladed impeller (2 cm  
64 length, 1 cm width). Once the powder mixture was ~~completely~~totally immersed, the solution  
65 was stirred during 2 h at room temperature (20 °C), then at 4 °C under slight agitation at 300  
66 rpm overnight to ensure complete powder reconstitution.

67 The proximal composition of spray-dried concentrates was calculated from technical sheets of  
68 reconstituted powders (Table 2).

### 70 **2.3. Production of dairy powders by spray-drying**

71 Spray-drying was carried out on a Micra Spray MS 150 single effect spray-dryer (Anhydro,  
72 Soeborg, Denmark), equipped with a bi-fluid nozzle (Fluid Cap 60 100 + Air Cap 120, Spraying  
73 System, Wheaton, Illinois, USA). Spray-drying conditions were set at 200 °C drying air inlet  
74 temperature and 85 °C outlet temperature in view to obtain powders with good preservation  
75 abilities (i.e., water activity ranging from 0.2 to 0.3) [1,15]. The concentrate was fed by a  
76 FASTLoad peristaltic pump (VWR, Leuven, Belgium; 4.8 mm internal pipe diameter) and the  
77 feed concentrate flow rate was regulated at circa 68.0 mL.min<sup>-1</sup> to maintain the drying air outlet  
78 temperature at 85 °C [16]. Powders were bagged just after sampling to limit degradations  
79 susceptible to occur during storage (such as caking induced by moisture uptake) and stored at  
80 10 °C until analyses.

### 81 **2.4. Physicochemical characterization**

#### 82 2.4.1. Particle size distribution

83 Particle size measurements were carried out with a laser granulometer (Mastersizer S, Malvern  
84 Instruments Ltd, Malvern, UK) supplied with a 5 mW He-Ne laser operating at a wavelength  
85 of 632.8 nm with a 300 RF lens. Air dispersion was achieved with the Aero S module (Malvern).  
86 The dispersion conditions were adapted to each powder: air pressure between 2 and 3 bar, feed  
87 rate between 60 and 90 %, and hopper length between 2 and 3 mm. Three replicates were  
88 analyzed for each sample. The mean diameter in volume was chosen as size estimator. Classical  
89 granulometric parameters ( $D_{10}$ ,  $D_{50}$ , and  $D_{90}$ ) were calculated, where  $D_x$  designates the diameter  
90 for which  $x$  % of the particles have a smaller size. The width of the particle size distribution  
91 was evaluated with the span (Eq. (1)) [17].

$$span = \frac{D_{90} - D_{10}}{D_{50}} \quad (1)$$

## 92 2.4.2. Scanning electron microscopy

93 Scanning electron microscopy (SEM) was employed to characterize size, morphology, and  
94 surface structure of milk powders. Powder samples were sputtered on double-sided carbon  
95 tape, fixed to SEM stubs, and coated with carbon in a sputter coater (Polaron SC7640,  
96 Thermo VG Scientific, East Grinstead, England). Observations were performed at  $\times 500$   
97 magnification under partial vacuum using a scanning electron microscope (Hitachi S-4800,  
98 Japan) operating at 1.0 kV acceleration voltage [18].

## 99 **2.5. Powder interactions with water**

### 100 2.5.1. Water activity

101 Water activity was measured with a HygroPalm23-AW portable water activity-meter (Rotronic,  
102 France) at room temperature (20 °C), using a 3 cm powder layer ( $15 \pm 1$  g) in a cup similarly  
103 to ISO 21807:2004 [19].

### 104 2.5.2. Water content

105 The water content was measured by the loss of water mass of  $2.0 \text{ g} \pm 0.1 \text{ mg}$  powder after  
106 drying during at least 3 h at 103 °C until a constant dry powder mass was obtained following  
107 ISO 5537:2004 [20].

### 108 2.5.3. Reconstitutability

109 Reconstitutability of spray-dried powders was evaluated through wettability, dispersibility and  
110 solubility indices.

### 111 2.5.3.1. Wettability

112 The wettability index is defined as the time in seconds taken by  $13.0 \pm 0.1$  g powder to become  
113 wet when gently deposited at the surface of  $125.0 \pm 0.1$  g distilled water in a 250 mL beaker,  
114 following IDF 87:1979 [21].

### 115 2.5.3.2. Dispersibility

116 Dispersibility was measured by a method adapted from IDF 87: 1979 [21].  $13.0 \pm 0.1$  g powder  
117 was gently deposited on the surface of 125.0 g water at 20 °C placed in a 400 mL beaker. The  
118 mixture was manually stirred for 20 s, then it was allowed to rest for 30 s. After that, the mixture  
119 was passed through a 120  $\mu\text{m}$ -mesh sieve. The total dry matter of the liquid, corresponding to  
120 the dispersibility index, was determined by drying at 103 °C for 5 h following (Eq. (2)).

$$121 \text{ Dispersibility (\%)} = \frac{W \times T}{100 - (E + T)} \times \frac{100}{S} \quad (2)$$

122 with:

123 - W: distilled water mass (g);

124 - T: dry matter content (% w/w) of the liquid;

125 - E: dry matter content (% w/w) of sampled powder;

126 - S: sampled powder mass (g).

### 127 2.5.3.3. Solubility

128 Solubility was characterized as the percentage of powder able to dissolve in distilled water  
129 following ISO 8156:2005 [22]. It was determined by the addition of  $2.5 \pm 0.1$  g powder in  
130 17.5 mL distilled water in a 50 mL centrifugation tube followed by 30 s stirring and  
131 centrifugation at  $2\,294 \times g$  during 10 min (Megafuge 8R, Thermoscientific, France).  
132 Afterwards, the supernatant was removed, and the pellet was added to 20 mL distilled  
133 water, followed by stirring and centrifugation at  $2\,294 \times g$  during 10 min. The pellet was



134 desiccated at 103 °C overnight to determine the mass of insoluble residues, which allows  
135 determining the solubility index (Eq. (4)).

$$136 \text{ Solubility (\%)} = \frac{(S-M) \times 100}{S} \quad (4)$$

137 with:

138 - M: undissolved sediment mass after desiccation (g);

139 - S: sampled powder mass (g).

## 140 **2.6. Flow properties of dairy protein powders**

141 Flowability properties were evaluated using a FT4 powder rheometer with 25 mm accessories  
142 (Freeman Technology, Worcestershire, UK).

### 143 2.6.1. Stability test

144 The stability test was performed through a series of seven conditioning cycles by rotating the  
145 blade ( $-5^\circ$  helix angle, 23.5 mm diameter,  $100 \text{ mm}\cdot\text{s}^{-1}$  tip speed) and measurements of the flow  
146 energy of the powder bed [9,13].

147 This test allows determining two flow indices: BFE and SE. The BFE is linked to the flow  
148 behavior of the powder bed, and a low BFE generally corresponds to an easy-flowing behavior,  
149 whereas a high BFE is often obtained for poorly-flowing powders. The SE is much linked to  
150 powder cohesion [8,9] with high SE values associated with cohesive powders.

151 Freeman Technology provides a classification of powder flow behavior on the basis of the SE  
152 calculated by the FT4 powder rheometer by:

153 -  $SE < 5 \text{ mJ/g}$ : low cohesion

154 -  $5 \text{ mJ/g} < SE < 10 \text{ mJ/g}$ : moderate cohesion

155 - SE > 10 mJ/g: high cohesion.

### 156 2.6.2. Compressibility test

157 The compressibility test is used to measure the evolution of powder density as a function of  
158 normal stress from 0.5 to 15 kPa applied by a 23.5 mm vented piston to the powder sample  
159 placed in a 25 mm × 10 mL measurement cell [9,23,24].

160 The compressibility test allows obtaining the bulk density (BD), i.e. powder density before  
161 compression, similar to aerated density, and the compressed bulk density (CBD) for powder  
162 density after compression. Particle size, shape, surface composition, and roughness are known  
163 to influence these compressibility parameters [8].

### 164 2.6.3. Shear cell test

165 The shear cell test was carried out to evaluate powder flowability and cohesion under high shear  
166 stress after a precompaction at 9 kPa. The powder bed was subjected to normal stresses of 7 to  
167 3 kPa by 1 kPa steps in a 25 mm × 10 mL measurement cell [8,9,13,14,24].

168 The shear test is used to determine the powder cohesion and the flow factor ( $ff$ ), calculated by  
169 the yield locus method from the shear stress vs. normal stress curve [8]. Powder flowability has  
170 been classified by Jenike [12] from flow factor values:

171 -  $ff < 1$ : not flowing

172 -  $1 < ff < 2$ : very cohesive

173 -  $2 < ff < 4$ : cohesive

174 -  $4 < ff < 10$ : easy flowing

175 -  $f > 10$ : free flowing.

## 176 2.7. Statistical analysis

177 All analyses were triplicated to ensure a good analytical repeatability and presented values  
178 correspond to means  $\pm$  standard deviations. Data were statistically analyzed by one-way  
179 ANOVA performed with Excel version 2016 and means were separated by Tukey's HSD test  
180 at  $p < 0.05$  significance level. The degree of correlation between powder physicochemical and  
181 functional properties was evaluated by calculating Pearson's correlation coefficients.

## 182 3. RESULTS AND DISCUSSION

### 183 3.1. Particle size distribution

184 Dairy protein powders spray-dried at pilot scale were composed of fine particles ranging from  
185 1 to 200  $\mu\text{m}$  (Figure 1 & Table 3), consistently with the particle size range generally obtained  
186 with the employed pilot spray-dryer [16,25–27]. Their median particle size  $D_{50}$  was comprised  
187 between 11.5 and 18.9  $\mu\text{m}$ , particle size distributions were monomodal, and moderate  
188 dispersion around the median value was denoted by the span values ranging from 2.1 to 2.5.

189 Consistently with literature, the median particle size of protein powders increased with casein  
190 proportion of spray-dried concentrates (Table 2) [28], as evidenced by the ~~significant~~  
191 positive correlation obtained between these two parameters according to a Pearson's correlation  
192 coefficient of 0.932. This may be explained by the fact that during drying, droplets of  
193 concentrates rich in caseins quickly form a crust at their surface [11], which results in larger  
194 particles than spray-dried protein powders containing a greater whey protein proportion. No  
195 significant influence of protein composition on the span of the particle size distribution was  
196 denoted.

197 *Fig. 1. Particle size distributions (in volume) of PM 1, PM 2, PM 3, PM 4, and PM 5.*

198

Table 3. Granulometric parameters of PM 1, PM 2, PM 3, PM 4, and PM 5 powders.

199 **3.2. Particle appearance**

200 SEM images of protein powders (Figure 3) showed that protein composition influenced particle  
201 shape: particle sphericity decreased at higher casein proportion, confirming previous literature  
202 results [11,29,30]. This may confirm that the surface of droplets of concentrates rich in caseins  
203 had rapidly formed a crust when put in contact with the hot drying air and warped during the  
204 drying step, leading to non-spherical particles. It may be concluded that whey protein  
205 concentrate droplets did not form a crust upon drying, thus the spherical shape of droplets was  
206 kept during the drying step, leading to globally spherical whey protein particles.

207 *Fig. 2. SEM micrographs of spray-dried protein powders varying in protein composition (in the increasing order*  
208 *of casein proportion: PM 1 (A); PM 2 (B); PM3 (C); PM 4 (D); PM 5 (E)) acquired at × 500 and × 2000*  
209 *magnifications.*

210 **3.3. Powder interactions with water**

211 Water activity of spray-dried dairy protein powders was low, ranging from 0.16 to 0.26 (Table  
212 4), ensuring an excellent good protection against spoilage during storage [31], as the ideal water  
213 activity of food products, especially dairy, is comprised between 0.2 and 0.3 [1]. The good  
214 preservability of produced powders was confirmed by their water content, not exceeding the  
215 threshold of 5 % (w/w) on wet basis generally advised for dairy powders [32]. The slight  
216 variation of water content of spray-dried dairy protein powders in spite of identical  
217 manufacturing conditions may come from the difference in hygroscopicity of whey proteins  
218 and caseins along with the impact of formulation on the drying kinetics.

219 The fineness of spray-dried dairy protein powders caused their poor wettability, as expected for  
220 powders spray-dried at pilot-scale [8,16,33]. Their dispersibility was very low (around  
221 5 - 10 %), highlighting a high level of powder cohesion (which may be an indication of poor  
222 flowability). The influence of each dairy protein type could not be deduced from these results,

223 because all powders were hardly dispersible [1,11], leading to insignificant differences between  
224 powder samples [34,35]. The impact of protein type was much more interesting on powder  
225 solubility. Indeed, a linear decrease (Eq. 5,  $R^2 = 98.62\%$ ) of powder solubility was observed  
226 when increasing the casein content of spray-dried dairy protein powders (Table 4-B & Figure  
227 5).

$$228 \text{ solubility (\%)} = 99.554 - 0.698 \times \text{casein content (\%)} \quad (\text{Eq. 5})$$

229 This can be related to literature indicating that the presence of casein constitutes a limiting  
230 factor for powder reconstitutability, especially by impairing solubility [1,5,36,37].  
231 Moreover, besides, as whey proteins and lactose from dairy protein powders are known to  
232 solubilize well more easily and quickly than caseins [7], the decrease in their contents upon  
233 increase in casein content (cf. Table 2) may contribute to the observed solubility decrease. As  
234 regards the impact of mineral content, which increased with casein content in studied powders  
235 (cf. Table 2), Mimouni et al. (2010) [7] showed that calcium, phosphorus, and magnesium,  
236 which constitute the majority of dairy minerals as they are principally associated with casein  
237 micelles, have rather slow reconstitution kinetics; this also contributes to the solubility decrease  
238 of investigated powders upon casein content increase. Last, lipids content in studied powders  
239 was low and very close (cf. Table 2), thus the negative impact of these highly hydrophobic  
240 components [1, 2, 3] (Gaiani et al., 2006) [1–3,38] on powder solubility could not be observed  
241 in the present study.

242 *Table 4. Water content, water activity, wettability, dispersibility, and solubility of spray-dried dairy protein*  
243 *powders.*

244 Fig. 3. Evolution of powder solubility with casein content for spray-dried dairy protein powders.

### 245 3.4. Powder flow properties

246 Flow parameters deduced from the stability, compressibility, and shear cell tests are gathered  
247 in Table 5.

248 BFE of spray-dried dairy protein powders increased in the following order: PM1 < PM5 ≈ PM2  
249 < PM3 < PM4. No significant clear influence of protein composition on basic flowability energy  
250 was denoted, except that 100 % whey protein powder (PM1) had slightly better flowability. As  
251 its smaller median particle size and larger span should be deleterious for flow properties, it can  
252 be hypothesized that its better flowability may arise from the more spherical shape of its  
253 particles. Specific flow energy is generally more informative regarding flowability  
254 classification. In the present study Here, specific flow energy results indicate that PM1 and PM5  
255 are expected to better flow and be moderately cohesive, followed by PM2, PM3, and PM4 that  
256 are expected to flow more poorly and have higher cohesion. Afterwards Then, powders with  
257 better flowability were either composed of very small particles of relatively high heterogeneity  
258 but rather spherical shape (PM1) or small particles of moderate heterogeneity and wrinkled  
259 shape (PM5). Indeed, powder flowability in generally improved for large particles, of low  
260 heterogeneity, and/or of spherical shape [8–11].

261 Compressibility at 15 kPa was identical for all spray-dried dairy protein powders (Figure 4),  
262 presumably owing to their relatively close particle size distributions and their composition  
263 dominated by dairy proteins. It can be guessed that their cohesion was not too different, as no  
264 marked differences of compressibility have been obtained. In fact, cohesion, deduced from the  
265 shear cell test, was comprised between 1.55 and 1.75 kPa for all investigated powders,  
266 indicating a rather high level of cohesion, and thus a low flowability.  $ff$  values were found in  
267 the range from 2.78 to 3.12, showing that all investigated powders had close flow properties,  
268 exhibiting a cohesive behavior and a low flowability. No significant impact of protein  
269 composition was evidenced, but trends indicated that cohesion was slightly raised and  $ff$  slightly

270 reduced for powders containing more caseins (Figure 5); this was confirmed by the highly  
271 negative Pearson's correlation coefficient between casein proportion and  $ff$  ( $R = -0,762$ ) and  
272 the moderately positive Pearson's correlation coefficient between casein proportion and  
273 cohesion ( $R = 0,570$ ). Thus, flowability under normal stress was a little improved for powders  
274 richer in whey proteins.

275 *Table 5 : Flowability characteristics (from stability, compressibility, and shear cell tests) of PM 1, PM 2, PM 3,*  
276 *PM 4, and PM 5.*

277 *Fig. 4: Compressibility results for PM 1, PM 2, PM 3, PM 4, and PM 5.*

278 *Fig. 5: Shear test results for PM 1, PM 2, PM 3, PM 4, and PM 5.*

## 279 4. CONCLUSION

280 This study is part of a wider research project intending to evaluate the relative influences of  
281 formulation (~~whey proteins/caseins ratio preparation and composition of concentrates~~) and  
282 spray-drying conditions on physicochemical and functional properties of spray-dried dairy  
283 powders. ~~The final aim of the project is to establish process relationships relating formulation~~  
284 ~~and spray drying conditions to powder reconstitutability, flowability, and preservability.~~

285 ~~In the present study, a focus was carried out on the influence of protein composition on~~  
286 ~~functional properties of spray dried dairy protein powders. Thus, dairy protein powders~~  
287 ~~differing in whey protein/casein ratio were spray dried in identical the same conditions in order~~  
288 ~~to isolate the influence of protein composition.~~

289 ~~In the present study, a focus was carried out on the influence of whey protein/casein ratio on~~  
290 ~~functional properties in identical spray-drying conditions.~~

291 ~~All powders presented similar particle size distributions, which were monomodal with a~~  
292 ~~moderate dispersion of particle sizes around the median value. A marked influence of~~

293 ~~formulation was denoted by the small increase in median particle size at higher casein~~  
294 ~~proportion.~~

295 Particle size distributions of spray-dried powders varying in whey proteins/caseins ratio were  
296 similar (they were monomodal and had comparable span), but a marked influence of  
297 formulation was denoted by the small increase in median particle size at higher casein  
298 proportion.

299 ~~This was explained by the different drying behaviors of droplets of whey protein and casein~~  
300 ~~concentrates: formulations richer in caseins are known to lead to the formation of a crust at the~~  
301 ~~droplet surface, resulting in larger and wrinkled particles, whereas droplets containing more~~  
302 ~~whey proteins are known to lead to more spherical particles of smaller size. These hypotheses~~  
303 ~~of different drying mechanisms, consistent with the literature, were validated by the differences~~  
304 ~~in particle appearance between powders containing more whey proteins and those composed of~~  
305 ~~more caseins.~~

306 Formulations richer in caseins are known to lead to the formation of a crust at the droplet  
307 surface, resulting in larger and wrinkled particles, whereas droplets containing more whey  
308 proteins are known to lead to more spherical particles of smaller size.

309 ~~Water content and water activity of all spray dried powders were low, ensuring their good~~  
310 ~~preservability.~~

311 Owing to the low median particle size obtained for powders spray-dried at pilot scale,  
312 wettability and dispersibility of produced powders were very low according to industrial  
313 standards, and no significant difference due to powder formulation was evidenced. Solubility  
314 linearly varied with the whey protein/casein ratio (it was presumably impaired by the presence  
315 of a casein crust at the particle surface) and this process relationship may help industrials  
316 formulating their spray-dried dairy protein powders.



317 Flowability of spray-dried dairy protein powders was poor and interparticular cohesion was  
318 high, presumably due to the small median particle size of powder spray-dried on the employed  
319 pilot-scale equipment. For all powders, the influence of their particle size distribution seemed  
320 to overwhelm the impact of powder formulation. However, a slight improvement of flow  
321 properties was observed for dairy protein powders containing more whey proteins and less  
322 caseins. This may arise from the more spherical shape of this kind of powders. Powder  
323 flowability, mainly controlled by median particle size, should be improved for powders spray-  
324 dried at industrial scale, which are generally well larger.

## 325 **ACKNOWLEDGEMENTS**

326 The authors acknowledge support of the LIBio by the "Impact Biomolecules" project of the  
327 "Lorraine Université d'Excellence" (Investissements d'avenir – ANR). Moreover, the authors  
328 acknowledge the help given by the LIBio scientific team for the training and provision of  
329 analytical devices.

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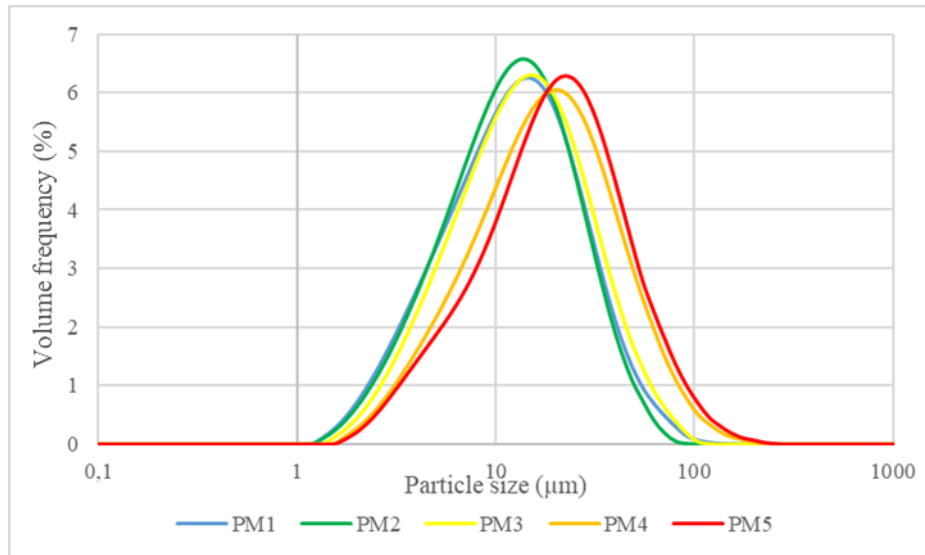


Fig. 1. Particle size distributions (in volume) of PM 1, PM 2, PM 3, PM 4, and PM 5.

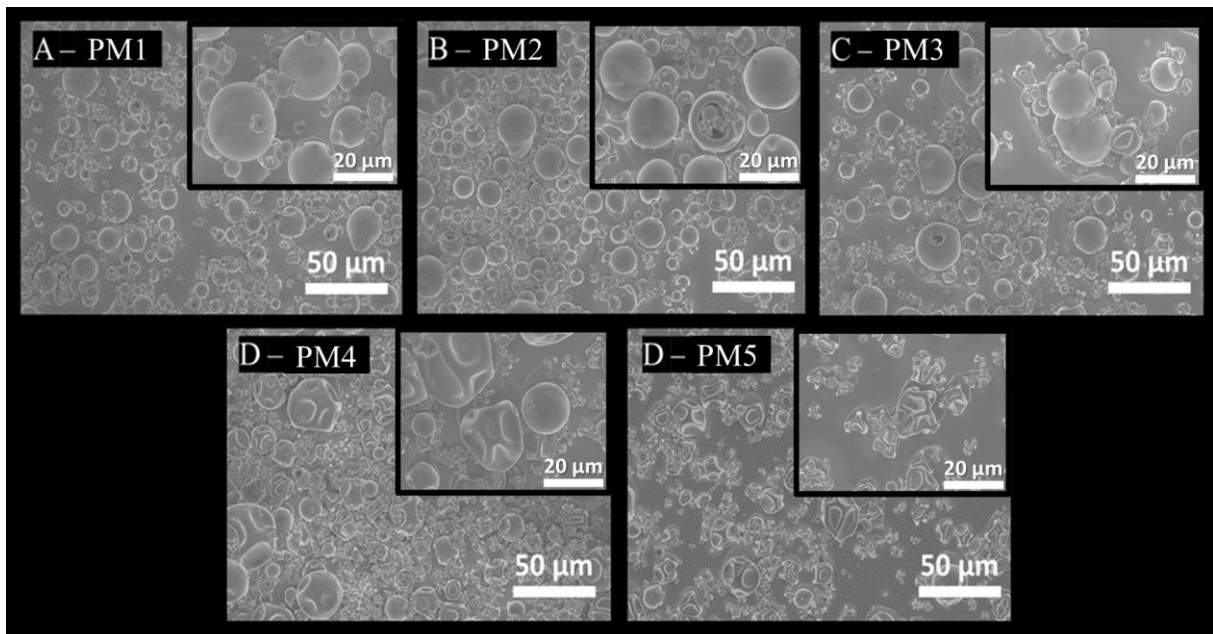


Fig. 2. SEM micrographs of spray-dried protein powders varying in protein composition (in the increasing order of casein proportion: PM 1 (A); PM 2 (B); PM3 (C); PM 4 (D); PM 5 (E)) acquired at  $\times 500$  and  $\times 2000$  magnifications.

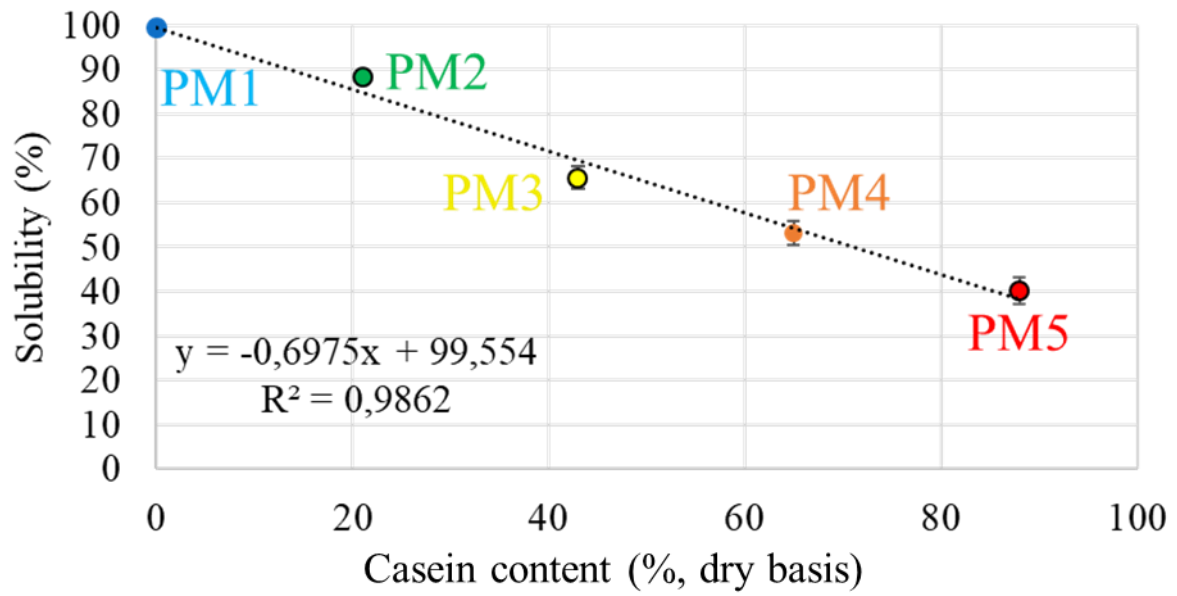
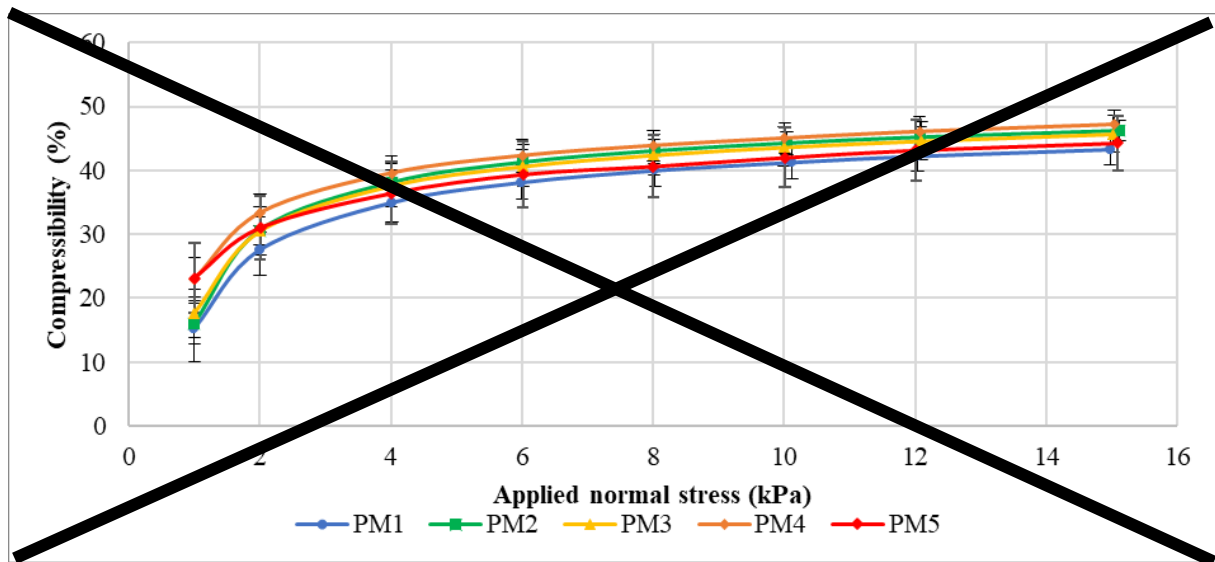


Figure 3. Evolution of powder solubility with casein content for spray-dried dairy protein powders.



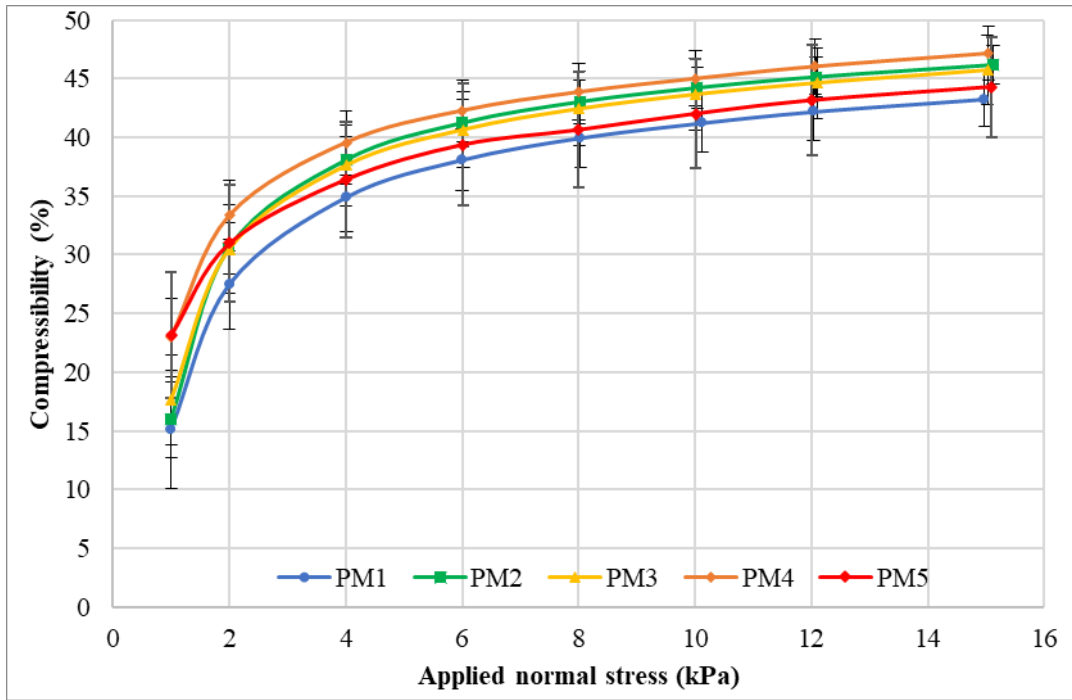
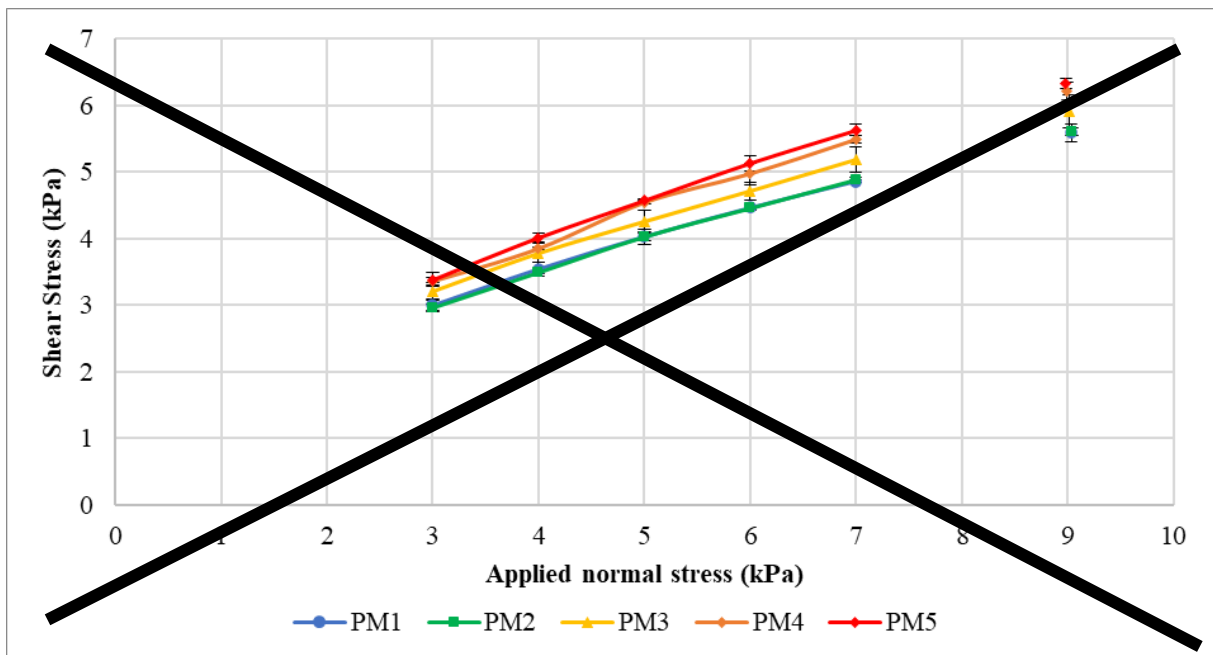


Fig. 4: Compressibility results for PM 1, PM 2, PM 3, PM 4, and PM 5.



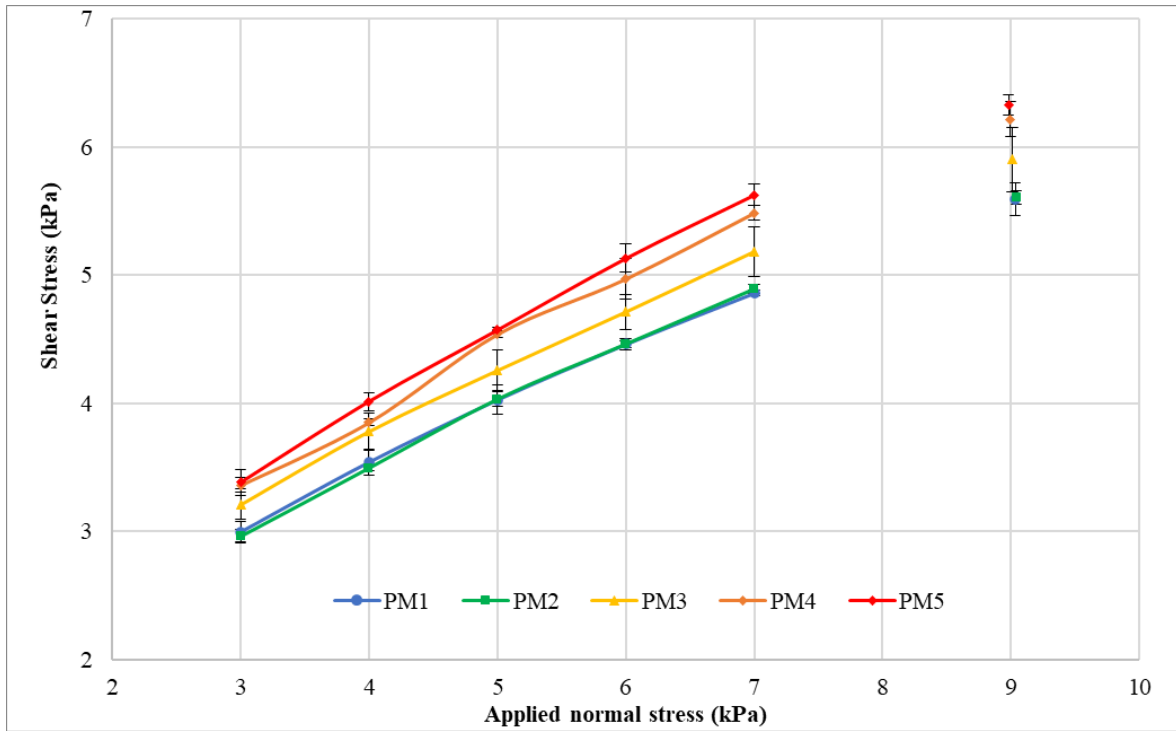


Fig. 5 : Shear test results for PM 1, PM 2, PM 3, PM 4, and PM 5.



*Table 1. Proximate composition expressed on wet basis of whey protein isolate and phosphocaseinate powders employed for the preparation of dairy concentrates to be spray-dried.*

Powder	Moisture	Proteins	Whey	Caseins (%)	Lipids (%)	Lactose (%)	Minerals (%)
	(%)	(%)	proteins (%)				
Whey proteins isolate	5.5	85.5	85.5	0	1.0	5.5	2.5
Casein powder	5.0	83.0	10.8	72.2	1.5	2.0	8.5

*Table 2. Proximate composition on dry basis of spray-dried concentrates.*

Concentrates	Proteins	Whey	Caseins (%)	Lipids (%)	Lactose (%)	Minerals (%)
	(%)	proteins (%)				
PM 1	90.5	90.5	0.0	1.1	5.8	2.6
PM 2	89.7	70.6	19.1	1.2	4.9	4.2
PM 3	88.9	50.7	38.3	1.3	4.0	5.8
PM 4	88.1	30.7	57.4	1.5	3.0	7.4
PM 5	87.4	10.8	76.6	1.6	2.1	9.0

*Table 3. Granulometric parameters of PM 1, PM 2, PM 3, PM 4, and PM 5 powders.*

Powders	D <sub>10</sub> (µm)	D <sub>50</sub> (µm)	D <sub>90</sub> (µm)	Span (-)
PM 1	3.79 <sup>e</sup> ± 0.01	11.82 <sup>d</sup> ± 0.11	31.02 <sup>d</sup> ± 0.53	2.31 <sup>b</sup> ± 0.05
PM 2	3.92 <sup>d</sup> ± 0.04	11.53 <sup>d</sup> ± 0.04	28.51 <sup>e</sup> ± 0.34	2.15 <sup>c</sup> ± 0.02
PM 3	4.34 <sup>c</sup> ± 0.03	12.80 <sup>c</sup> ± 0.23	33.81 <sup>c</sup> ± 0.43	2.29 <sup>b</sup> ± 0.02
PM 4	5.20 <sup>b</sup> ± 0.00	16.93 <sup>b</sup> ± 0.14	47.04 <sup>b</sup> ± 0.22	2.47 <sup>a</sup> ± 0.01
PM 5	5.61 <sup>a</sup> ± 0.03	18.92 <sup>a</sup> ± 0.13	51.33 <sup>a</sup> ± 0.15	2.42 <sup>a</sup> ± 0.00

Means with different superscripted letters in the same column were significantly different according to Tukey's

HSD test ( $p < 0.05$ ;  $n = 3$ ).

Table 4. Water content, water activity, wettability, dispersibility, and solubility of spray-dried dairy protein powders.

Dairy protein powder sample	Water content (% (w/w) on wet basis)	Water activity (-)	Wettability (min)	Dispersibility (%)	Solubility (%)
PM 1	4.82 <sup>a</sup> ± 0.14	0.21 <sup>c</sup> ± 0.00	> 90	5.38 <sup>b</sup> ± 0.08	99.26 <sup>a</sup> ± 0.00
PM 2	3.82 <sup>c</sup> ± 0.12	0.20 <sup>d</sup> ± 0.00	> 90	4.83 <sup>c</sup> ± 0.09	88.35 <sup>b</sup> ± 0.21
PM 3	3.78 <sup>c</sup> ± 0.04	0.23 <sup>b</sup> ± 0.00	> 90	5.06 <sup>c</sup> ± 0.14	65.58 <sup>c</sup> ± 2.58
PM 4	3.78 <sup>c</sup> ± 0.15	0.16 <sup>e</sup> ± 0.00	> 90	8.91 <sup>a</sup> ± 0.13	53.11 <sup>d</sup> ± 2.80
PM 5	4.38 <sup>b</sup> ± 0.17	0.25 <sup>a</sup> ± 0.00	> 90	3.26 <sup>d</sup> ± 0.08	40.12 <sup>e</sup> ± 2.90

Means with different superscripted letters in the same column were significantly different according to Tukey's

HSD test ( $p < 0.05$ ;  $n = 3$ ).

Tableau 5 : Flowability characteristics (from stability, compressibility, and shear cell tests) of PM 1, PM 2, PM 3, PM 4, and PM 5.

	Basic flowability energy BFE (mJ)	Specific flow energy SE (mJ/g)	Compressed bulk density CBD (g/mL)	Bulk density BD (g/mL)	Compressibility at 15 kPa (%)	Cohesion (kPa)	Flow factor <i>ff</i> (-)
PM1	38.72 <sup>b</sup> ± 3.87	8.85 <sup>bc</sup> ± 0.62	0.34 <sup>b</sup> ± 0.03	0.19 <sup>b</sup> ± 0.01	43.28 <sup>a</sup> ± 2.33	1.65 <sup>a</sup> ± 0.16	3.01 <sup>a</sup> ± 0.16
PM2	50.87 <sup>a</sup> ± 5.19	10.55 <sup>ab</sup> ± 0.39	0.36 <sup>ab</sup> ± 0.01	0.20 <sup>b</sup> ± 0.00	46.16 <sup>a</sup> ± 1.62	1.55 <sup>a</sup> ± 0.09	3.12 <sup>a</sup> ± 0.10
PM3	56.45 <sup>a</sup> ± 6.01	11.12 <sup>a</sup> ± 0.77	0.37 <sup>ab</sup> ± 0.00	0.20 <sup>ab</sup> ± 0.01	45.73 <sup>a</sup> ± 2.96	1.79 <sup>a</sup> ± 0.24	2.78 <sup>a</sup> ± 0.25
PM4	59.43 <sup>a</sup> ± 3.52	11.74 <sup>a</sup> ± 0.98	0.41 <sup>a</sup> ± 0.01	0.22 <sup>a</sup> ± 0.01	47.15 <sup>a</sup> ± 2.27	1.75 <sup>a</sup> ± 0.24	2.79 <sup>a</sup> ± 0.25
PM5	50.28 <sup>ab</sup> ± 0.90	8.35 <sup>c</sup> ± 0.99	0.39 <sup>a</sup> ± 0.03	0.22 <sup>a</sup> ± 0.00	44.31 <sup>a</sup> ± 4.27	1.72 <sup>a</sup> ± 0.24	2.80 <sup>a</sup> ± 0.25

*Means with different superscripted letters in the same column were significantly different according to Tukey's*

*HSD test ( $p < 0.05$ ;  $n = 3$ ).*

## **Highlights**

- Whey protein/casein ratio impacts on dairy protein powder characteristics
- Dairy protein powder solubility linearly varied with the whey protein/casein ratio
- Standard reconstitution tests were irrelevant for powders spray-dried at pilot scale
- Particle size eclipses the impact of the whey protein/casein ratio on flowability

**Credit author statement**

Tristan FOURNAISE: Conceptualization, Investigation, Methodology, Validation, Writing, Review & Editing.

Jennifer BURGAIN: Project administration, Conceptualization, Supervision, Review & Editing, Funding acquisition.

Carole PERROUD-THOMASSIN: Investigation, Review & Editing.

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### **Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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