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Physical and flow properties of pseudocereal-based protein-rich ingredient powders

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ABSTRACT

Knowledge of bulk handling properties of food powders is essential in the design of industrial equipment and selection of appropriate powder handling operations. The objectives of this study were to determine the physical and flow properties of plant-based regular and protein-rich flours to establish relationships between powder physical and bulk handling properties as influenced by protein enrichment. A number of physical properties (bulk density, flowability, wall friction and compressibility) were assessed for 11 regular- and protein-rich flours from pseudocereals (amaranth, buckwheat, quinoa) and cereals (rice and maize). Relevant physicochemical properties such as particle size distribution, microstructure and water sorption behaviour were also studied. The protein-rich pseudocereal flours had irregular-shaped, rough surfaces with mean particle diameters ranging from 96.5 to 215 μm. The compressibility indices (42.6–51.4%) were higher for the former compared to the regular protein content powders and they displayed lesser tendency to uptake water with increasing relative humidity. Analysis of the flow behaviour showed the protein-rich flours to be more cohesive with higher wall friction angle values than the regular protein content powders. The new information obtained in this study is critical in optimising the processing, stability and applications of these value-added high-protein pseudocereal ingredient powders.

1. Introduction

Plant-based protein ingredients are gaining popularity due to their positive contributions in addressing environmental and food security challenges, in addition to their cost effectiveness compared to animal protein ingredients (Aiking, 2011; Henchion et al., 2017). Replacing, either partly or in full, animal protein with plant protein ingredients or development of new plant protein-based food products is increasingly being practiced commercially (Don, 2017). The increasing demand for plant protein and plant-based foods is driving the search for alternative plant protein sources with enhanced functional and nutritional properties. Pseudocereals (e.g., quinoa, amaranth and buckwheat), sometimes collectively referred to as Andean cereals, are grains with nutritional composition similar to cereals such as rice and maize (Schoenlechner et al., 2008). Quinoa and amaranth are cultivated in South America, mainly Peru and Bolivia, while buckwheat, originally from Central Asia was later transferred to Central and Eastern Europe (FAO, 2013). There is an increasing demand for these pseudocereal ingredients in Europe and their cultivation is now being practiced in a number of countries including Spain, France and Italy (CBI, 2017).

High levels of protein in plant-based ingredients can be achieved from these raw materials using different approaches (e.g., dry or wet fractionation) obtaining as a result a protein-rich ingredient in dry powder format. Dry fractionation is a milder and more sustainable technique than wet fractionation for production of protein concentrates from cereals and legumes as it does not require chemicals or water. However, the extent of enrichment of protein with dry fractionation is generally lower due to the presence of other components such as starch, fat and fibre in the final product (Schutyser et al., 2015). Dry fractionation typically involves fine milling of the seed to disclose protein-rich particles and subsequent dry separation of the flour in fractions of different particle sizes by air classification or sieving, with the proteins being more enriched in the fine fraction (Avila Ruiz et al., 2016; Schutyser et al., 2015). These protein-rich cereal and pseudocereal fractions

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are rich in amino acids such as methionine and tryptophan although the concentration on other essential amino acids (e.g., lysine) can be low. In food formulation, the deficiency in some amino acids can be compensated with the combination of other ingredients such as legumes (e.g., lupin or lentil) which naturally contain higher levels of lysine (Boye et al., 2010; Duranti, 2006).

Numerous physical properties are known to influence the behaviour of bulk powder products (Schulze, 2008). Generally, larger particle size tends to increase flowability of powders as a result of lower contact area between particles (Teunou et al., 1999). The particle shape and morphology also influence the behaviour of powders, with more spherical shape and smooth-surfaced particles reducing particle interlocking and resistance to flow (Amagliani et al., 2016a). The chemical composition also plays an important role: for example, fat tends to increase cohesiveness (Fitzpatrick et al., 2004b), moisture increases liquid bridges and capillary forces between powder particles (Bian et al., 2015; Landillon et al., 2008) and higher levels of protein can reduce flowability due to cohesiveness (Crowley et al., 2014). Water availability and sorption properties can provide useful information about the bulk handling properties of powders, as their flow behaviour can be influenced by the humid air which surrounds it (Mathlouthi and Rogé, 2003).

Plant protein-rich ingredients are generally used in the form of powders during processing, transportation and marketing (Barbosa-Cánovas and Yan, 2008; Jan et al., 2018). Therefore, knowledge of the bulk handling properties of these protein-rich ingredients is of special importance for several reasons, such as prevention of pipe blockages and irregular flow, optimum packaging and as a key quality attribute for consumer applications (e.g., powder scoop delivery) (Bouvier et al., 2013; Fitzpatrick et al., 2004b; Sharma et al., 2012). Therefore, there is a requirement for further information on the processing and handling characteristics of new and emerging plant-based protein-rich ingredients. To the authors knowledge, there is no information regarding the flow behaviour of pseudocereal ingredients available in the published literature; however, information is available on other types of flours such as wheat (Landillon et al., 2008), rice (Amagliani et al., 2016a) and soy (Bhandari, 2013). Therefore, the objectives of this study were to determine the physical and flow properties of eleven powders which included plant-based regular and protein-rich flours, mainly from pseudocereals, with a view to establishing relationships between powder physical and bulk handling properties as influenced by protein enrichment.

2. Materials and methods

2.1. Ingredients

The ingredients analysed consisted of 11 samples of flours and protein-rich flours. Seven of the samples were of pseudocereal origin which included quinoa wholegrain flour (QWGF), quinoa dehulled flour (QDF), quinoa protein-rich flour (QPRF), amaranth wholegrain flour (AWGF), amaranth protein-rich flour (APRF), buckwheat dehulled flour (BDF) and buckwheat protein-rich flour (BPRF). Protein enrichment in the protein-rich flours was achieved using a dry milling approach as described by Alonso-Miravalles and O'Mahony (2018). All the pseudocereal flours were provided by the Fraunhofer Institut (Munich, Germany) except the QWGF which was purchased from Ziegler & Co. (Wunsiedel, Germany). These ingredients were analysed in comparison with a white rice flour (RF) and a rice protein-rich flour (RPRF), both of which were supplied by Beneo (Tienen, Belgium), maize flour (MF), purchased from the Quay Co-op (Cork, Ireland) and a lupin protein-rich flour (LPRF), supplied by the Fraunhofer Institut (Munich, Germany). The chemical composition of these ingredients is available in the previously published study of Alonso-Miravalles and O'Mahony, (2018), except the LPRF sample which had 79.8% protein (w/w), 5.26% moisture (w/w) and fat was not detected.

2.2. Particle size distribution

The particle size distribution of the powders was determined by laser diffraction using a Malvern Mastersizer 3000 with Aero S dry dispersion unit (Malvern Instruments, Worcestershire, UK) operating at a feed rate of 20–40% using a hopper gap of 2.5 mm, and a pressure of 1 bar on the standard venturi disperser. Particle refractive and absorption indices were set to 1.52 and 0.1, respectively.

2.3. Scanning electron microscopy

The powders were mounted on aluminium stubs using double-sided adhesive carbon tape and sputter coated with a 5 nm layer of gold/palladium (Au:Pd = 80:20) in a Q150R ES (Quorum Technologies, UK) coating system. Subsequently, the powders were imaged using a JSM-5510 scanning electron microscope (JEOL Ltd, Tokyo, Japan), operated at an accelerating voltage of 5 kV and using a magnification of 500× for taking the images.

2.4. Colour

Colour of the powders was determined by measuring the CIELAB coordinates (L*, a* and b*) with a Chroma Meter CR-400 (Konica Minolta Sensing, Inc., Japan) equipped with a granular materials attachment CR-A50. A white calibration tile was used to calibrate the instrument prior to colour measurements. In the CIELAB colour space system, L* value measures brightness, with values ranging from 0 (black) to 100 (white), a* value measures degree of redness (positive values) or greenness (negative values), and b* value measures degree of yellowness (positive values) or blueness (negative values).

2.5. Water activity and water sorption properties

The water activity (a_w) of the powders was determined at 20 °C using an Aqualab Series 3 TE water activity meter (Decagon Devices, Pullman, Washington, US) equipped with a thermoelectric system that allows the instrument to maintain a set chamber temperature throughout the measurement. Water sorption isotherms of the powders were measured using an SPS11-10 μ Sorption Test System (Projekt Messtechnik, Ulm, Germany) as follows. The powders (1500 mg) were weighed into aluminium cups, relative humidity (RH) was initially set at 40% before being decreased stepwise to 10% and then increased stepwise up to 90% (in increments of 10% RH). Changes in moisture content of the powders were monitored throughout the analysis and each step was equilibrated for 24 h and measurements were conducted at 20 °C.

2.6. Flow properties

Flowability, wall friction, bulk density and compressibility index (CI) of the powders were analysed using a Brookfield Powder Flow Tester (Brookfield Engineering Laboratories, Inc., Midleboro, MA, US) as described by Crowley et al. (2014). The flow function was obtained by plotting the unconfined failure strength against the major principal consolidating stress (Fitzpatrick et al., 2007, 2004a). The inverse of the slope of each flow function was used to determine the flow index (ffc) and the Jenike classification for each powder (Jenike, 1964).

2.7. Statistical data analysis

All analyses were conducted in triplicate. The data generated was subject to one-way analysis of variance (ANOVA) using R i386 version 3.3.1 (R foundation for statistical computing, Vienna, Austria). A Tukey's paired comparison test was used to determine statistically significant differences ($p < 0.05$) between mean values for different samples, at a 95% confidence level.

Table 1
Particle size distribution parameters of quinoa wholegrain flour (QWGF), quinoa dehulled flour (QDF), amaranth wholegrain flour (AWGF), buckwheat dehulled flour (BDF), rice flour (RF), maize flour (MF), quinoa protein-rich flour (QPRF), amaranth protein-rich flour (APRF), buckwheat protein-rich flour (BPRF), rice protein-rich flour (RPRF), and lupin protein-rich flour (LPRF).

	D[4,3]	D[3,2]	Dv(10)	Dv(50)	Dv(90)	Span	SSA
	μm						(m^2/kg)
Flours							
QWGF	82.7 ± 2.05^{c}	16.3 ± 0.06^{b}	6.45 ± 0.07^{a}	27.8 ± 0.67^{b}	244 ± 8.00^{d}	8.62 ± 0.08^{i}	367 ± 1.28^{g}
QDF	202 ± 0.00^{i}	65.9 ± 0.21^{h}	27.3 ± 0.27^{g}	177 ± 0.31^{h}	417 ± 0.35^{hi}	2.20 ± 0.00^{bc}	91.0 ± 0.25^{a}
AWGF	191 ± 1.00^{h}	59.5 ± 0.83^{g}	22.9 ± 0.63^{f}	163 ± 1.61^{g}	398 ± 1.95^{gh}	2.30 ± 0.01^{cd}	100 ± 1.47^{b}
BDF	126 ± 1.00^{e}	43.8 ± 0.40^{e}	20.2 ± 0.27^{e}	92.3 ± 0.42^{e}	272 ± 3.32^{e}	2.75 ± 0.02^{ef}	137 ± 1.25^{d}
RF	95.7 ± 3.20^{d}	42.1 ± 0.76^{e}	17.8 ± 0.56^{d}	77.5 ± 1.03^{d}	196 ± 7.11^{c}	2.27 ± 0.07^{c}	142 ± 2.60^{d}
MF	173 ± 0.00^{g}	49.1 ± 0.17^{f}	15.5 ± 0.06^{c}	144 ± 0.30^{f}	384 ± 0.13^{g}	2.55 ± 0.00^{de}	122 ± 0.37^{c}
Protein-rich flours							
QPRF	134 ± 0.58^{f}	33.6 ± 0.60^{d}	11.8 ± 0.36^{b}	95.8 ± 1.41^{e}	323 ± 4.22^{h}	3.15 ± 0.09^{h}	178 ± 3.21^{c}
APRF	215 ± 5.00^{j}	69.2 ± 3.84^{i}	28.7 ± 2.35^{g}	202 ± 5.63^{i}	433 ± 3.39^{b}	2.05 ± 0.05^{b}	86.7 ± 4.34^{a}
BPRF	96.5 ± 6.50^{d}	22.1 ± 0.38^{c}	7.01 ± 0.10^{a}	72.6 ± 0.94^{cd}	237 ± 19.7^{gh}	3.02 ± 0.23^{gh}	271 ± 4.06^{f}
RPRF	72.0 ± 0.05^{b}	55.6 ± 0.08^{g}	34.1 ± 0.12^{h}	67.2 ± 0.09^{c}	118 ± 0.00^{a}	1.25 ± 0.01^{a}	408 ± 0.16^{b}
LPRF	22.1 ± 0.65^{a}	10.7 ± 0.21^{a}	4.94 ± 0.03^{a}	15.3 ± 0.68^{a}	50.2 ± 0.87^{fg}	2.90 ± 0.09^{fg}	562 ± 9.28^{h}

Values followed by different superscript letters in the same column are significantly different ($P < 0.05$).
D[4,3] = volume-weighted mean particle diameter.
D[3,2] = surface-weighted mean particle diameter.
Dv(10) = particle size below which 10% of sample volume is found.
Dv(50) = particle size below which 50% of sample volume is found.
Dv(90) = particle size below which 90% of sample volume is found.
Span = measurement of the width of the distribution calculated as ((Dv(90) – Dv(10)/Dv(50)).
SSA = specific surface area (i.e., total area of the particles divided by total weight).

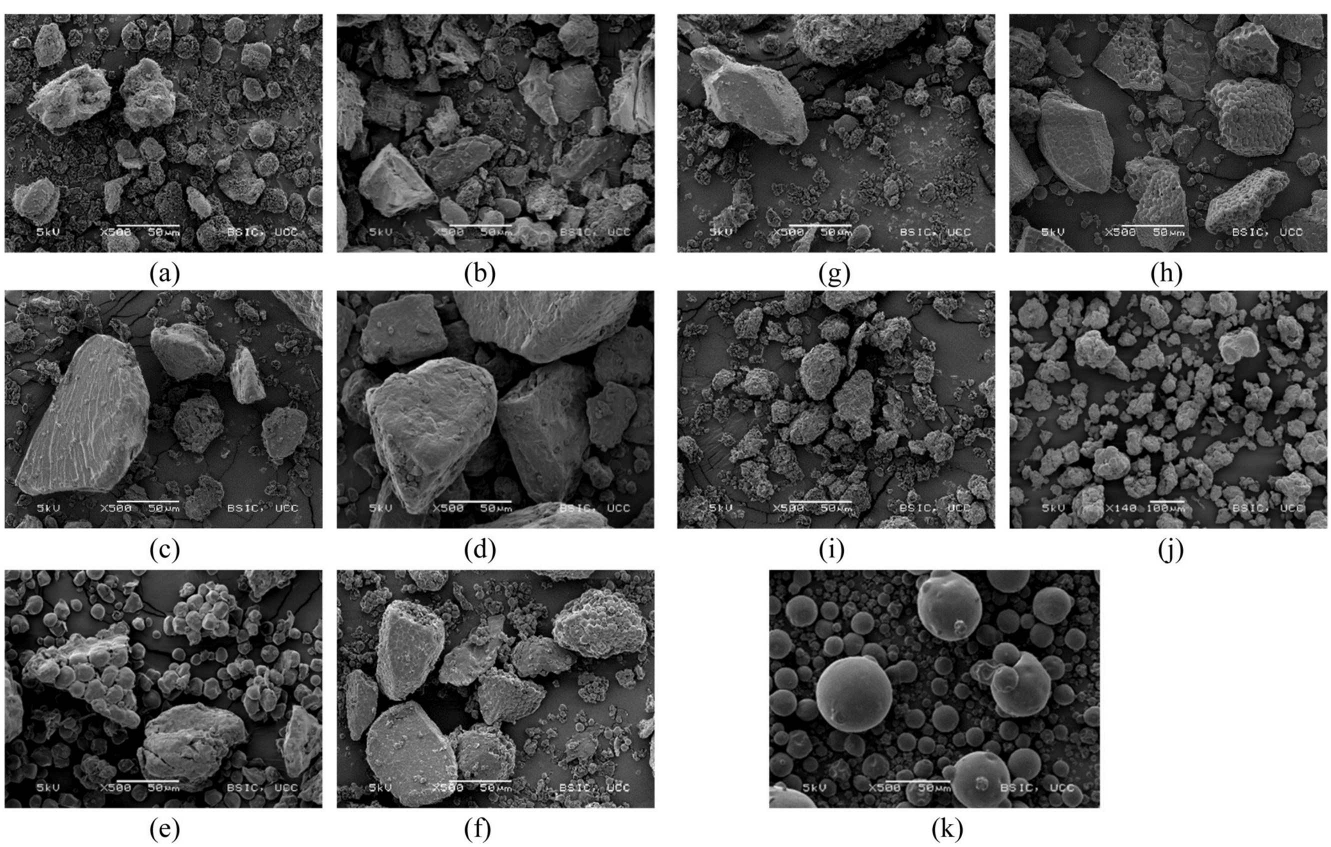

Fig. 1. Scanning electron micrographs of regular protein content flours (a–f) including (a) quinoa wholegrain flour, (b) quinoa dehulled flour, (c) amaranth wholegrain flour, (d) rice flour, (e) maize flour and (f) buckwheat flour; and protein-rich flours (g–k) including (g) quinoa protein-rich flour, (h) buckwheat protein-rich flour, (i) amaranth protein-rich flour, (j) rice protein-rich flour and (k) lupin protein-rich flour. Magnification and scale bar ×500 and 50 μm, respectively, except (j) ×140 and 100 μm, respectively.

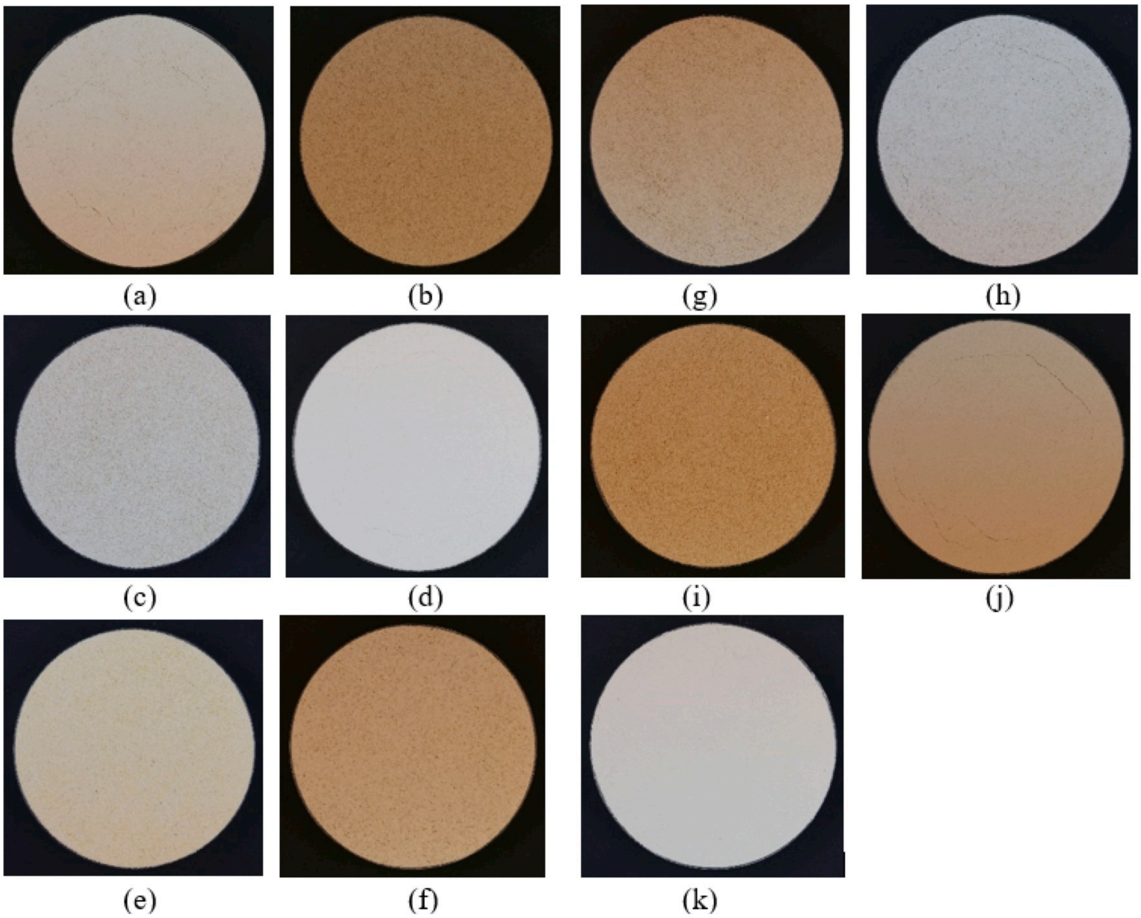

Fig. 2. Photographs of regular protein content flours (a–f) including (a) quinoa wholegrain flour, (b) quinoa dehulled flour, (c) amaranth wholegrain flour, (d) rice flour, (e) maize flour and (f) buckwheat flour and protein-rich flours (g–k) including (g) quinoa protein-rich flour, (h) buckwheat protein-rich flour, (i) amaranth protein-rich flour, (j) rice protein-rich flour and (k) lupin protein-rich flour.

3. Results and discussion

3.1. Particle size distribution

Particle size distribution is an important parameter that can influence the flow and bulk handling behaviour of food powders. In general, a wide range of particle size distributions from 22.1 to 215 μm for D[4,3] (volume-weighted mean particle diameter) were recorded, with the samples with higher protein content, RPRF and LPRF, having the smallest particle sizes (Table 1). Among the flours, QDF and AWGF had the highest values for D[4,3] at 202 and 191 μm, respectively; while QWGF (82.7 μm) and RF (95.7 μm) had the smallest particles. The results obtained for D[3,2] (surface-weighted mean particle diameter) and Dv(50) (particle size below which 50% of sample volume is found) followed the same general trends as for D[4,3]. Regarding the protein-rich flours, APRF had the largest D[4,3] value of 215 μm, followed by QPRF (134 μm) and BPRF (96.5 μm). Jan et al. (2018) reported mean particle size of ~171 μm for basmati rice in comparison with a commercial rice flour having much smaller mean particle size of 65.3 μm. Bian et al. (2015) compared the particle size of wheat flour with higher and lower protein content, reporting no significant differences between the particle sizes of the two types of flour. Bhandari (2013) reported particle size values from 100 to 5000 μm for cereal and soy flours, with the values being higher than those found for milk or coffee powders. In the present study, it was observed that the powders with highest protein content, LPRF (79.8%) and RPRF (75.0%), had the lowest values for D[4,3] (22.1 μm and 72.0 μm, respectively), Dv(50) (particle size below which 50% of sample volume is found) and Dv(90) (particle size below which 90% of sample volume is found). LPRF and RPRF also had the highest specific surface area (562 and 400 m^2/kg, respectively) which is related to the small size of their particles. The mean particle size obtained for LPRF was similar to the value found for spray-dried milk protein concentrates, with protein content ranging from 80 to 90% with values for Dv(10) (particle size below which 10% of sample volume is found) of ~10 μm (Crowley et al., 2014). Generally, fine powders are more susceptible to cohesion and their flowability is poor, while powders with larger particle sizes generally have better flowability (Bian et al., 2015; Teunou et al., 1999) due to less surface area being available for formation of interactive cohesive forces (Fitzpatrick et al., 2007). In relation to the span values, all the samples had values ranging from 2 to 3, except QWGF and RPRF, having the highest and lowest values (8.62 and 1.25, respectively), indicating that the QWGF sample consisted of a wide range of particle sizes while the LPRF sample was more homogeneous. Greater particle size distributions of powders can create challenges during powder handling as the smaller particles tend to fill the inter-particle spaces of the larger particles, thus increasing the surface contact and cohesion between flour particles (Bian et al., 2015).

3.2. Scanning electron microscopy

The scanning electron micrographs showed a wide variation in the shape and size of particles (Fig. 1). The flours and protein-rich flours showed similar structures with irregular, non-homogeneous shapes and sizes and rough surfaces. The surface roughness of the flours, protein-rich flours and RPRF can promote interactions between particles by Van der Waals forces and mechanical interlocking which can effectively

Table 2
Colour space values, water activity (a_w), flow index, flow classification and compressibility index (CI) of quinoa wholegrain flour (QWGF), quinoa dehulled flour (QDF), amaranth wholegrain flour (AWGF), buckwheat dehulled flour (BDF), rice flour (RF), maize flour (MF), quinoa protein-rich flour (QPRF), amaranth protein-rich flour (APRF), buckwheat protein-rich flour (BPRF), rice protein-rich flour (RPRF), and lupin protein-rich flour (LPRF).

	Colour space values			a_w	(-) Flow index	(-) Flow classification	CI
	L*	a*	b*				(%)
Flours							
QWGF	70.1 ± 0.43^g	0.60 ± 0.01^c	9.94 ± 0.07^c	0.46 ± 0.01^e	1.96 ± 0.03^e	Very cohesive/Cohesive	49.5 ± 0.12^g
QDF	61.4 ± 0.03^a	0.29 ± 0.01^b	13.0 ± 0.01^f	0.46 ± 0.00^e	4.35 ± 0.01^a	Easy-flowing	33.9 ± 1.38^{bc}
AWGF	66.2 ± 0.05^c	0.66 ± 0.01^d	11.8 ± 0.03^d	0.44 ± 0.00^d	3.33 ± 0.01^b	Cohesive	39.7 ± 1.42^{de}
BDF	67.1 ± 0.06^d	0.25 ± 0.01^b	8.26 ± 0.29^b	0.46 ± 0.00^e	4.76 ± 0.01^a	Easy-flowing	29.4 ± 2.50^{ab}
MF	70.0 ± 0.01^f	1.85 ± 0.02^h	24.0 ± 0.11^h	0.62 ± 0.00^f	2.70 ± 0.00^c	Cohesive	28.0 ± 3.44^a
RF	72.5 ± 0.30^i	0.75 ± 0.04^e	6.33 ± 0.01^a	0.46 ± 0.00^e	3.45 ± 0.02^b	Cohesive/easy-flowing	28.8 ± 2.33^{ab}
Protein-rich flours							
QPRF	62.5 ± 0.11^b	0.83 ± 0.01^f	14.6 ± 0.04^g	0.28 ± 0.00^a	2.08 ± 0.01^e	Very cohesive/cohesive	51.4 ± 0.62^g
APRF	62.8 ± 0.09^b	0.83 ± 0.02^f	14.4 ± 0.10^g	0.41 ± 0.01^c	3.45 ± 0.01^b	Cohesive	45.7 ± 0.08^{fg}
BPRF	67.7 ± 0.05^e	0.16 ± 0.02^a	8.61 ± 0.01^b	0.33 ± 0.00^b	2.44 ± 0.03^d	Cohesive	42.6 ± 0.65^{ef}
RPRF	64.0 ± 0.21^b	-0.06 ± 0.02^i	13.1 ± 0.12^f	0.46 ± 0.00^e	9.90 ± 1.62^f	Easy-flowing	27.8 ± 0.29^a
LPRF	70.9 ± 0.04^h	0.90 ± 0.01^g	12.6 ± 0.01^e	0.34 ± 0.02^b	3.33 ± 0.01^b	Easy-flowing/Cohesive	36.1 ± 2.17^{cd}

Values followed by different superscript letters in the same column are significantly different ($P < 0.05$).
a_w = Water activity.
CI = Compressibility index.

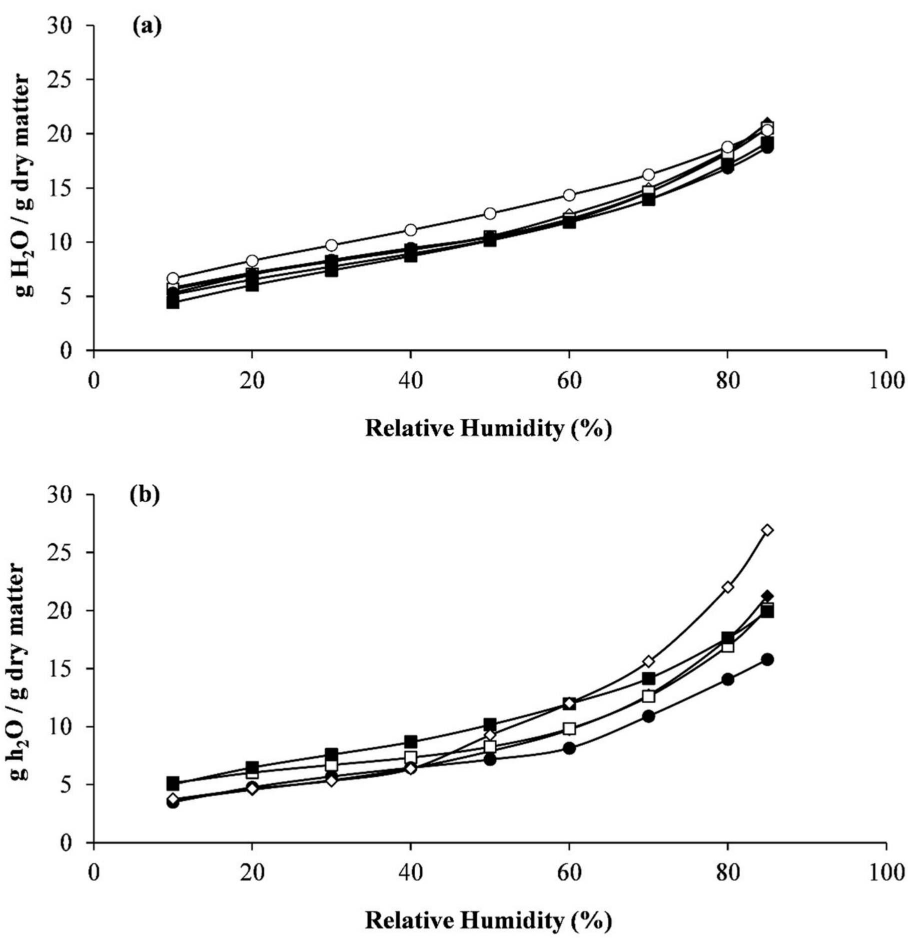

Fig. 3. (a) Moisture sorption isotherms of quinoa wholegrain flour (QWGF; —◇—), quinoa dehulled flour (QDF; —◆—), amaranth wholegrain flour (AWGF; —□—), buckwheat dehulled flour (BDF; —■—), rice flour (RF; —●—), maize flour (MF; —○—) and **(b)** moisture sorption isotherms of quinoa protein-rich flour (QPRF; —◆—), amaranth protein-rich flour (APRF; —□—), buckwheat protein-rich flour (BPRF; —■—), rice protein-rich flour (RPRF; —●—) and lupin protein-rich flour (LPRF; —◇—).

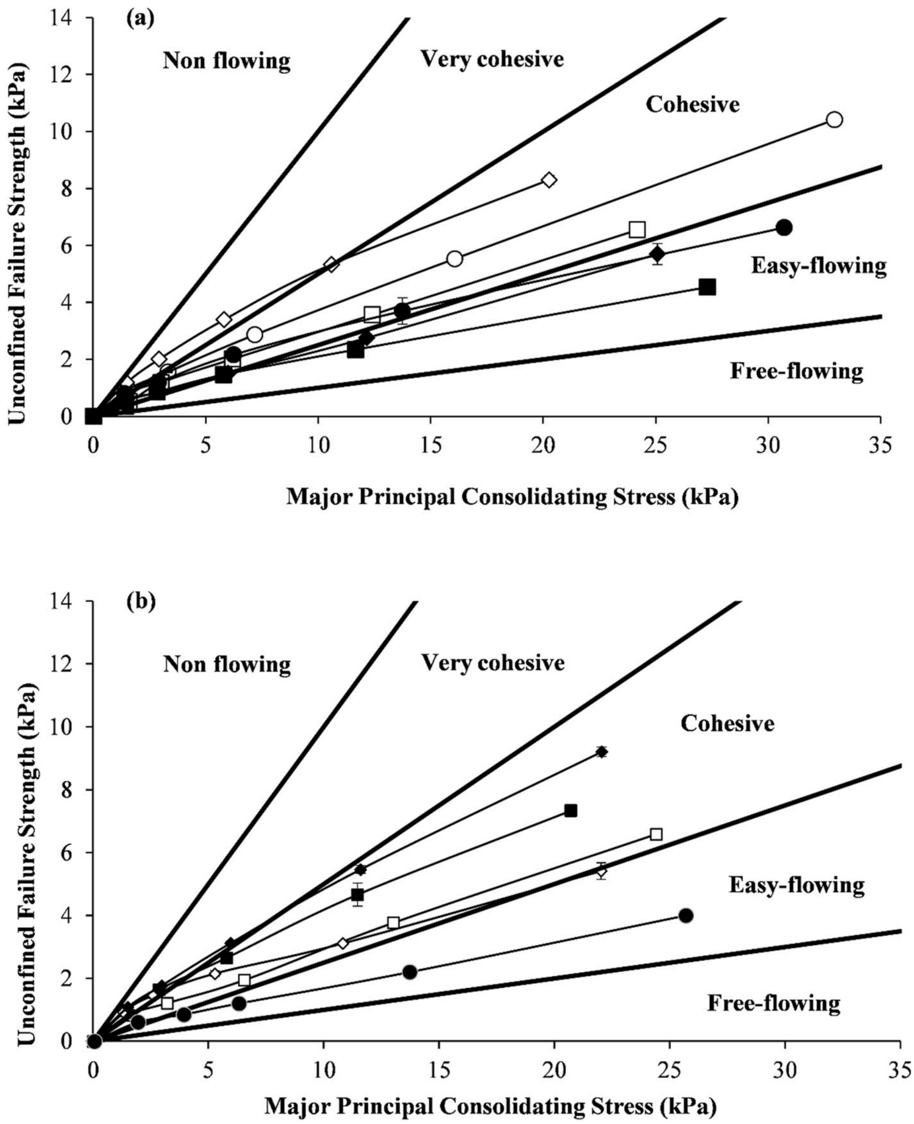

Fig. 4. (a) Flow function curves of quinoa wholegrain flour (QWGF; —◇—), quinoa dehulled flour (QDF; —◆—), amaranth wholegrain flour (AWGF; —□—), buckwheat dehulled flour (BDF; —■—), rice flour (RF; —●—), maize flour (MF; —○—) and **(b)** flow function curves of quinoa protein-rich flour (QPRF; —◆—), amaranth protein-rich flour (APRF; —□—), buckwheat protein-rich flour (BPRF; —■—), rice protein-rich flour (RPRF; —●—) and lupin protein-rich flour (LPRF; —◇—).

increase the cohesiveness of such powders (Amagliani et al., 2016a); for example, rod-shaped and irregular-shaped powder particles result in poorer flowability (Ambrose et al., 2016). Bhandari (2013) described cereal flours as large particles with irregular shape in comparison to other food products such as milk powder. In MF and BPRF, clusters of spherical starch granules were observed and a similar structural feature has been observed for rice flour in a previous study carried out by (Amagliani et al., 2016a). Among the flours, QWGF presented the smallest and most spherical particles, which was in agreement with the particle size distribution analysis results having the smallest particle size, highest uniformity and specific surface area among the flours analysed. Ahmed et al. (2018) described quinoa flour particles as distinct irregular-shaped polygon structures and also observed individual granules as a result of the milling process. In the present study, individual granules were also observed in the flours and protein-rich flours. The samples with highest protein content, RPRF and LPRF, had the smallest and most homogeneous size and shaped particles. RPRF presented a rough particle surface, while LPRF showed smooth spherical particles of two different sizes as supported by the span values. According to the literature, the spherical shape of LPRF is thought to result in less particle interlocking and resistance to flow. Alamilla-Beltrán et al. (2005) observed similar spherical features to the ones reported here for LPRF for different spray-dried powders, suggesting that the LPRF was dried using this technology.

3.3. Colour

Colour is an important quality attribute in the food and bioprocess industries, and it influences consumer's choices and preferences (Pathare et al., 2013). The samples with higher lightness values (L*) were RF, LPRF, QWGF and MF (72.5, 70.9, 70.1 and 70.0, respectively) while the other samples showed lower values for L* (between 61.4 and 67.7). Protein-rich samples, QPRF and APRF, had darker colours than their regular protein content flours, except BPRF that had similar L*

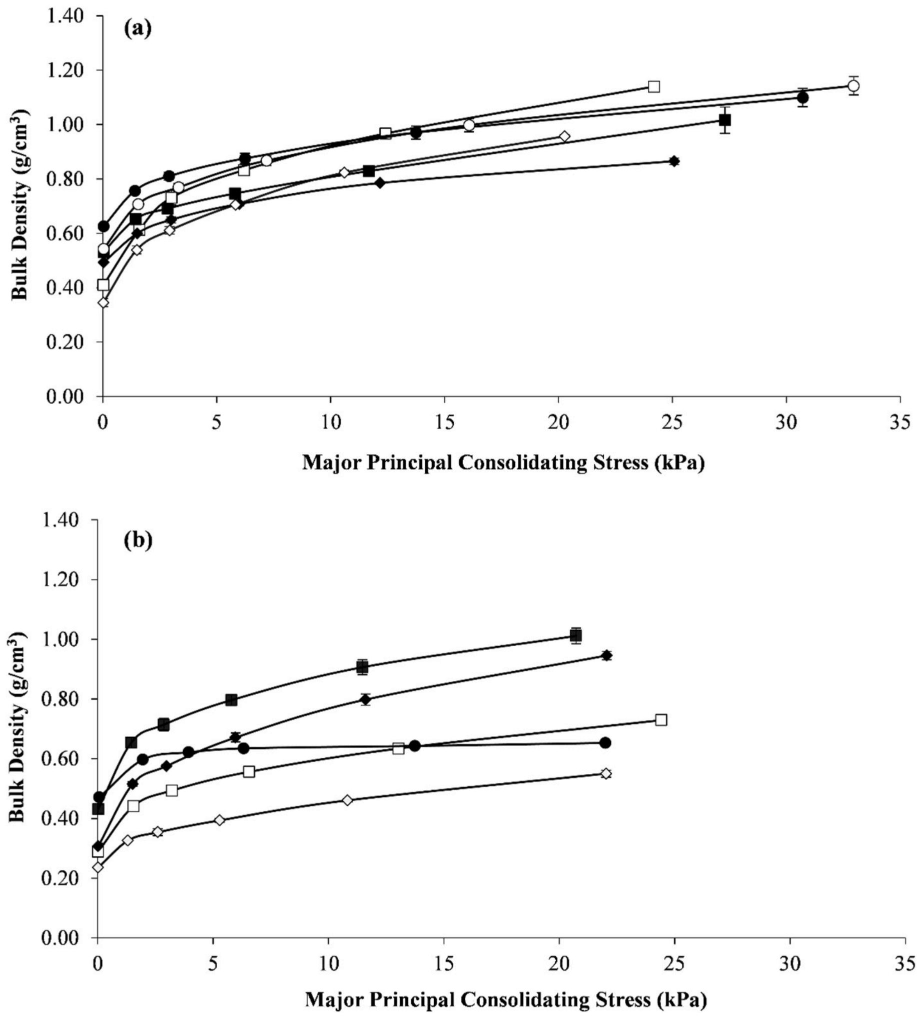

Fig. 5. **(a)** Bulk density as a function of major principal consolidating stress for quinoa wholegrain flour (QWGF; —◇—), quinoa dehulled flour (QDF; —◆—), amaranth wholegrain flour (AWGF; —□—), buckwheat dehulled flour (BDF; —■—), rice flour (RF; —●—), maize flour (MF; —○—). **(b)** Bulk density as a function of major principal consolidating stress of quinoa protein-rich flour (QPRF; —◆—), amaranth protein-rich flour (APRF; —□—), buckwheat protein-rich flour (BPRF; —■—), rice protein concentrate (RPRF; —●—) and lupin protein isolate (LPRF; —◇—).

value to BDF (67.7 and 67.1, respectively). Regarding the a*, which corresponds to green-red values, QPRF had a significantly higher value than QWGF and QDF, with the protein-rich variant having more of a tendency to red colour. Furthermore, APRF had also a redder colour than AWGF, which is evident in the photographs of the samples (Fig. 2). The b* values were positive for all samples, indicating that all powders had somewhat of a yellow colour, with MF having the highest value (24.0) while BPRF had the lowest b* value (8.61).

3.4. Water activity and moisture sorption isotherms

All the flours had similar values for water activity (~0.44), except MF which had the highest value (0.62) among this group (Table 2). Regarding the protein-rich flours and protein concentrates, all samples, except RPRF, had significantly lower water activity than the flours. Water activity and moisture content strongly influence flowability, with higher values facilitating liquid bridging between powder particles, thereby decreasing flowability (Mathlouthi and Rogé, 2003); usually, cohesive forces are promoted by the presence of liquid at the outer surface layer of the particles (Landillon et al., 2008). The moisture sorption isotherms of the powders are shown in Fig. 3, and these isotherms illustrate the capacity of a powder for uptake or release of water when placed in atmospheres of different relative humidity (RH) at a specified temperature (Teunou et al., 1999). They provide useful information about the behaviour of food powders at different RHs such as the prediction of caking behaviour which is detrimental to powder flowability (Mathlouthi and Rogé, 2003). All the powders displayed similar overall water sorption behaviour showing a positive relation between the increase in relative humidity (RH) and water uptake by the sample (i.e., the higher the relative humidity the higher the water content of the sample). Cereal grain flours and milling fractions are hygroscopic (Kaletunç and Breslauer, 2003). Increased moisture content can make the particle surfaces more sticky, resulting in greater adhesion between the particles and a wall surface, which in turn, increases wall friction (Iqbal and Fitzpatrick, 2006). Flours and protein-rich flours showed comparable results, with the exception of the powders with highest protein content, LPRF and RPRF, both of which showed lower water uptake than the other samples at RH between 10 and 40%. However, LPRF showed a rapid increase in water uptake with RH increasing to greater to 40%, reaching the highest value at 85% RH in comparison with the flours and protein-rich flours. In contrast, RPRF seemed to be the least hygroscopic sample and it is possible that the low uptake of water by RPRF may be due to the strong

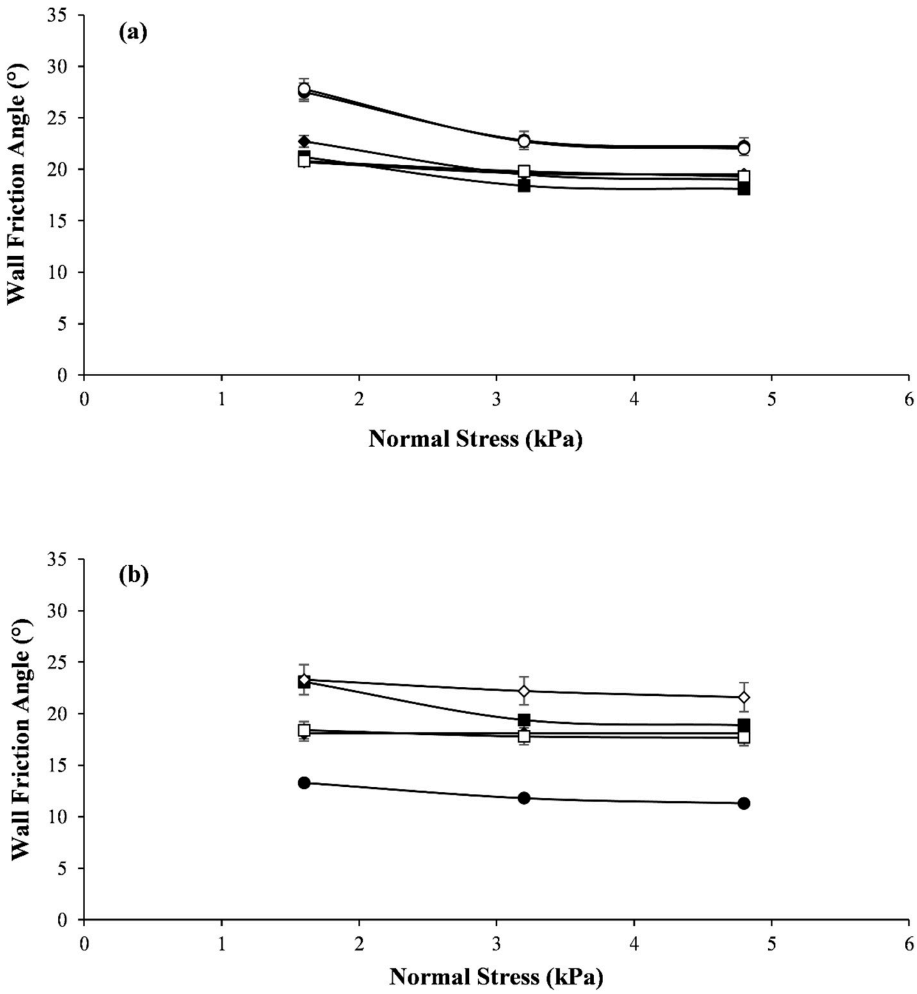

Fig. 6. (a) Wall friction angle as a function of normal stress for quinoa wholegrain flour (QWGF; —◇—), quinoa dehulled flour (QDF; —◆—), amaranth wholegrain flour (AWGF; —□—), buckwheat dehulled flour (BDF; —■—), rice flour (RF; —●—), maize flour (MF; —○—) and **(b)** wall friction angle as a function of normal stress for quinoa protein-rich flour (QPRF; —□—), amaranth protein-rich flour (APRF; —■—), buckwheat protein-rich flour (BPRF; —◆—), rice protein-rich flour (RPRF; —◇—) and lupin protein-rich flour (LPRF; —◆—).

hydrophobic properties of rice proteins (Amagliani et al., 2016b; Van Der Borght et al., 2006).

3.5. Flowability

The flow properties of the powders are displayed in Fig. 4, and data for flow index, flow classification and compressibility index (CI) of the powders are provided in Table 2. If a powder has a flow index greater than 10 it is considered free-flowing, while powders with flow index of 10–4 are considered easy-flowing, whereas cohesive, very cohesive, and non-flowing powders have flow indices less than 4, 2 and 1, respectively (Jenike, 1964; Tomas and Schubert, 1974). Of the eleven powders, two were classified as very cohesive, four as cohesive and the remaining five as easy flowing. Among the protein concentrates and isolates, the powders with highest protein content, LPRF (fat not detected) and RPRF (0.79% fat), were classified as easy-flowing, while the protein-rich flours (QPRF, BPRF and APRF) were classified as cohesive. Protein-rich flours have higher fat contents than regular flours (Alonso-Miravalles and O'Mahony, 2018) and it is known that higher levels of surface fat have a major influence on powder flowability (Fitzpatrick et al., 2007; Silva and O'Mahony, 2017). Landillon et al. (2008), reported that the presence of fat in wheat flour significantly contributes to its cohesiveness. Furthermore, the protein-rich flours, as seen in Section 3.2, have irregular shapes and sizes leading to greater potential for interlocking between the particles in comparison with RPRF and LPRF. QPRF was the most cohesive of the protein-rich samples, with the QPRF also having the smallest particle size distribution and highest span values; numerous studies have shown the influence of small particle size on the cohesiveness of different powders (Fu et al., 2012). Regarding the flours, QWGF showed the most cohesive behaviour at all the consolidating stresses, followed by MF. QWGF had the smallest particle size (82.7 μm) and widest range of particle sizes, this is related to several studies (Landillon et al., 2008; Schulze, 2008) where they reported the correlation between small particle sizes and more cohesive powders. Furthermore, the cohesive behaviour displayed by MF can be related to its high moisture content and a_w, similar to the results obtained by Guan and Zhang (2009) who reported increasing cohesiveness with increasing moisture content of wheat flour. Fitzpatrick et al. (2004a,b), in their analysis also described corn flour as cohesive, along with soy and wheat flours.

3.6. Bulk density and compressibility index

The protein-rich samples had lower initial bulk density values (0.24–0.47 g cm^{-3}) than the flours (0.34–0.63 g cm^{-3}). Fitzpatrick et al. (2004a,b) found slightly higher values for bulk density of 0.73, 0.71 and 0.60 g cm^{-3} for corn, wheat and soy flour, respectively. All the samples, except RPRF, displayed higher bulk density at higher consolidating stresses. The bulk density value of RPRF increased until 5 kPa and then remained stable at higher consolidating stresses (Fig. 5). QDF, BDF and QWGF had the lowest values for bulk density, while RF, MF and AWGF had the highest values for bulk density. The values for these flours were in the range of those reported by Barbosa-Cánovas and Yan (2008) for wheat, rye and corn flour. The protein-rich flours and protein concentrates and isolates had lower values for bulk density and higher compressibility indices in comparison with the flours. This is in line with the results of Emami and Tabil (2008), who reported lower values for bulk density for a protein fraction from chickpea compared to the respective flour. In this study, protein-rich pseudocereal powders showed cohesive behaviour with higher compressibility indices and lower bulk density values than their counterparts. Previous studies carried out on dairy protein ingredient powders have shown that high protein content contributes to lower powder bulk density (Zuurman et al., 1995; Wright et al., 2009; Crowley et al., 2014). Furthermore, it has been previously reported that highly compressible dairy protein ingredient powders have impaired handling properties (such as flowability) (Crowley et al., 2014; Fu et al., 2012; Silva and O'Mahony, 2017), reflected also in the results of the present study for protein-rich pseudocereal flours.

3.7. Wall friction angle

The wall friction angle of the powders as a function of normal stress is shown in Fig. 6. The flours and protein-rich flours presented values for wall friction angle from 20 to 31° and from 15 to 25° at the lower stress applied (2 kPa), respectively, while at higher stresses (5 kPa) they showed values of 20–25° and from 10 to 25°, respectively. As the normal stress applied increased, the wall friction angle decreased for all the regular protein content flours. However, in the protein-rich flours the wall friction angle remained almost stable at all stresses, except for BPRF (20.5% protein) which was the powder with the lowest protein content among the protein-rich ingredients. This suggests that the higher the protein content the lower is the tendency for the wall friction angle to change with increasing stress applied. Fitzpatrick et al. (2004a,b) reported wall friction angle values of 18.2, 13.0 and 13.2° at higher stress of 5.9 kPa. Interestingly, LPRF and RPRF showed the highest and lowest wall friction angle, respectively. RPRF, apart from having the lowest wall friction angle, also showed an easy-flowing behaviour. Among the flours, MF and RF showed higher wall friction angles compared to the samples of pseudocereal origin which are higher in protein.

4. Conclusion

This study provided a comprehensive analysis of physical and flow properties of a range of plant-based protein-rich ingredients, in particular of protein-rich pseudocereal flours and their comparison with regular protein content cereal flours. It can be concluded from the different analysis performed that all the protein-rich pseudocereal flours showed cohesive behaviour, had higher compressibility indices and lower bulk density values than regular protein content flours. These results can be attributed, at least in part, to the higher fat content, more non-homogeneous particle size and rougher powder particle surfaces for the protein-rich pseudocereal flours. The results obtained in this study suggest that protein-rich pseudocereal flours may be challenging to handle due to their cohesive behaviour and further studies are needed in order to improve the bulk handling and flow properties of such novel pseudocereal protein-rich ingredients.

CRediT authorship contribution statement

Loreto Alonso-Miravalles: Methodology, Investigation, Writing - original draft. **Emanuele Zannini:** Project administration, Writing - review & editing. **Juergen Bez:** Methodology, Writing - review & editing. **Elke K. Arendt:** Funding acquisition, Supervision, Writing - review & editing. **James A. O'Mahony:** Conceptualization, Supervision, Writing - review & editing, Project administration.

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