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Effect of relative humidity on milling and air classification explained by particle dispersion and flowability

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A R T I C L E I N F O

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A B S T R A C T

Dry fractionation of legumes is used to produce protein and starch-rich fractions with a clean label and lower environmental impact than conventional wet fractionation. Dry fractionation relies on the use of ambient air that varies in humidity. This study assessed the effect of relative humidity (RH) on milling and air classification of yellow pea and chickpea. Particle size analysis and powder rheology were used to assess the particle dispersibility and flowability of the flours. The RH has limited effect on milling and air classification between 30% and 70%. However, upon storage of fine milled chickpea flour at a RH of 70% and storage of fine milled yellow pea flour at a RH of 90% the air classification performance decreased. This was linked to a poorer dispersibility and flowability. Concluding, a relative humidity above 70% should be prevented to perform robust air classification.

1. Introduction

Ingredients from pulses, such as yellow pea or chickpea, are of major interest to produce plant-based foods (Grasso et al., 2022). This is due to the fact that pulses grow with a relatively high water use efficiency, have the capability to fix atmospheric nitrogen, which reduces the need for added fertilizers, and their favourable nutritional profile (high in protein and fibre, low in oil) (Gustafson, 2017). Pulses contain about 22% of protein, 43% starch, 19% fibre and 9% moisture, with generally a very low oil content (~2%) except for chickpea (~6%) (Wang et al., 2020). However, many plant-based foods require a higher protein content than is naturally present in pulse ingredients, which calls for protein enrichment via processing. The traditional wet fractionation of plant protein requires a copious amount of energy and water, and the harsh conditions can cause protein denaturation, both are usually unfavourable (Assatory et al., 2019). Dry fractionation is considered as a sustainable alternative to produce protein, starch- and fibre concentrates with preserved native properties (Assatory et al., 2019; Lie-Piang et al., 2021; Schutyser and van der Goot, 2011). These pulse ingredients have great commercial potential and can be applied in many food products, such as pasta, noodles, plant based meat, beverages, soups and sauces (Wang et al., 2020). To provide guidelines for industrial dry fractionation, it is important to identify which factors affect the process performance and link these to powder properties.

Dry fractionation includes milling and dry separation by sieving, air classification or electrostatic separation. This study focuses on air classification, which is the most common dry separation process to produce protein and starch concentrates from pulses (Wang et al., 2020). Impact milling first fragments the cotyledon into separate starch granules and smaller protein and fibre fragments. Subsequently, air classification is used to separate cellular components based on the difference in particle size and density. This typically results in a protein-rich fine fraction and a starch-rich coarse fraction (Boye et al., 2010). The separation is controlled by the classifier wheel speed and airflow rate, which determine the cut point for separation, where particles have an equal chance to end up in the fine or coarse fraction (Bauder et al., 2004; Guo et al., 2007). The cut point can be estimated by:

$$x = \frac{3}{4} \bullet c_D \bullet \frac{\rho_a}{\rho_p - \rho_a} \bullet \frac{\nu_r^2}{\nu_\varphi^2} \bullet r \tag{1}$$

with c_D drag coefficient (−) (a function of Reynolds number which is further related to the air velocity), ρ_a density of airflow (Pa • s), ρ_p particle density (kg/m^2), ν_r radial velocity (m/s), ν_φ tangential velocity (m/s), r diameter of the rotor (m) (Bauder et al., 2004; Guo et al., 2007). The radial velocity is the ratio between the airflow (m^3/s) and the total open area of the classifier slits (m^2). For air classification of pulse flours,

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the cut point should be in between the size of a protein body and a starch granule to facilitate separation as illustrated in Appendix figure A1.

During milling and air classification ambient air is typically used as processing air. Ideally, the processing performance does not vary with changes in environmental conditions during storage or processing. However, the humidity of the processing air can change with weather conditions. These changes can again affect the milling process, as moisture is known to influence fracture behaviour and energy consumption during milling (Dijkink and Langelaan, 2002b; Pelgrom et al., 2013a). Furthermore, the humidity affects powder flowability and particle dispersibility, where a lower particle flowability and dispersibility have resulted in a lower dry separation efficiency (Armstrong et al., 2014; Dijkink et al., 2007). A lower flowability can result in fouling by the cohesion of particles between the classifier wheel vanes, or on the classifier chamber walls, which can result in large losses of material and a lower protein recovery (Pelgrom et al., 2013b). Such material losses can pose problems for the application of dry fractionation (Assatory et al., 2019).

An additional effect of humidity is that the air density decreases slightly at higher relative humidity. This can indirectly affect the set cut point during air classification (Equation (1)). A slightly different cut point can influence the separation, especially when sizes or densities of the different cellular constituents overlap, for example for chickpea (Appendix figure A1). A difference in affinity for water can influence the moisture content equilibration. For example, oil has a lower water affinity than starch or protein, which results in faster equilibration to lower moisture contents for seeds with higher oil contents under the same conditions (Suma et al., 2013). This means that crops with a higher oil content might be affected differently by RH upon processing. In this research we used yellow pea to represent pulses with a low oil content and chickpea to represent pulses with a higher oil content. Till now, research focussed mainly on temperature and humidity upon material storage rather than during processing. Therefore, we want to understand how important it is to control the relative humidity in the processing room during milling and air classification.

This study aims to evaluate the effect of relative humidity on milling and air classification performance of legume flours and powder properties (particle size, dispersibility and flowability). Insight in the powder properties and process performance under different conditions is used to provide guidelines for moisture control during dry fractionation of pulses. Next to that, we evaluate if certain powder properties can be used as qualitative indicators for the process performance. Yellow pea and chickpea were selected as crops to represent pulses with a low oil content and a high oil content, as crops with a higher oil content might be affected differently by RH. The obtained knowledge on the dispersibility of small particles and particle flowability in relation to dry fractionation could accelerate the application of dry fractionation to other, less conventional materials and prevent losses upon dry processing of materials.

2. Materials and methods

2.1. Materials

Dry yellow pea (Pisum sativum) was obtained from P. van Schelven (Nieuwe-Tonge, the Netherlands) and dry Kabuli chickpea (Cicer arietinum L.) was obtained from Alimex Europe B.V. (Sint-Laureins, Belgium). The pulses were stored at 4 °C in closed containers before use.

2.2. Control of relative humidity

The RH of the process room was controlled with a condensing dehumidifier Condair DC75 (Condair, Pfäffikon, Switzerland), and the actual real-time RH and temperature were measured with a data logger Testo Savaris 2 (Testo, Lenzkirch, Germany). The low humidity (target RH30) ranged between 28.3% and 43.1%, and the increased humidity (target RH70) ranged between 51.2% and 68.7%. The temperature of the room was between 17 °C and 27 °C upon milling and between 16 °C and 29 °C upon air classification.

The materials were equilibrated for 7 days in a climate chamber (Memmert, Schwaback, Germany), as preliminary trials showed that after 7 days the material weight remained constant. Aluminium dishes (8 cm·15 cm) with approximately 100 g grits or 60 g flour were placed in the climate chamber at 19 °C at 30% when milled at low humidity and at 70% when milled at high humidity. The humidity conditions used are shown in Fig. 1. For confirmation purposes, a new batch of yellow pea flour was also equilibrated at RH90.

2.3. Milling and air classification

The hulled pulses were pre-milled into grits (yellow pea DV_{50} 1022 ± 79 μm, chickpea DV_{50} 1213 ± 92 μm) with a pin mill (LV 15 M, Condux-Werk, Germany). The DV_{50} represents the average volume-based particle size. The grits were milled into a fine flour with a ZPS impact mill (Hosokawa-Alpine, Augsburg, Germany) at a milling speed of 8000 rpm and a classifier wheel speed of 4000 rpm and 2900 rpm for

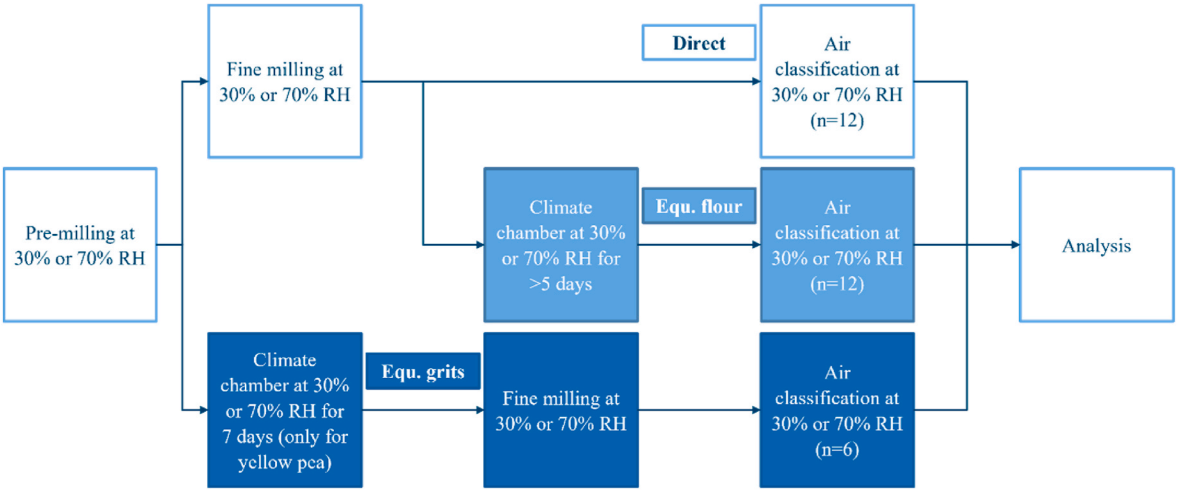

Fig. 1. Overview of storage conditions before milling and air classification for each crop and humidity. n represents the total number of air classifications (n) carried out for each condition. In figures, direct milled flours are represented with open symbols, and equilibrated flours (Equ. Flour) and equilibrated grits (Equ. Grits) with filled symbols.

yellow pea and chickpea respectively (Pelgrom et al., 2015b; Xing et al., 2020). Two airflows of 40 m^3/h (Xing et al., 2020) and 52 m^3/h (Pelgrom et al., 2015b) were used, where the pressure in the classifier chamber was approximately −10 mbar. Milling experiments were performed with a constant feed rate of 0.5 kg/h and a batch size of 700 g.

The flours were air-classified in an ATP50 classifier (Hosokawa-Alpine, Augsburg, Germany). For yellow pea, the airflow was fixed at 52 m^3/h (Pelgrom et al., 2015b). For chickpea, two airflows of 52 m^3/h (Xing et al., 2020) and 60 m^3/h were used. The higher airflow was used to increase the air classification yield. The classifier wheel speed was 8000 rpm for yellow pea and 10,000 rpm for chickpea, with feed rates of respectively 0.5 kg/h and 0.2 kg/h (Pelgrom et al., 2015b; Xing et al., 2020). The batch size was 180–200 g, and one larger batch was used of 750 g. The air classifications for yellow pea milled at 40 m^3/h under different humidity conditions were repeated at least three times to identify the variation, other air classifications were repeated once or twice.

2.4. *Moisture adsorption isotherms*

The water sorption isotherms of the milled flours were determined with the Dynamic Vapour Sorption (DVS) Discovery apparatus (TA Instruments, Allentown, DE, USA) at 20 °C and 50 °C, to represent the temperature of the milling room and the temperature inside the mill. This because the temperature during milling can rise considerably, due to the heat generated (di Silvestro et al., 2014; Pelgrom et al., 2015a). The temperature inside the mill used in this study was assumed to be in the same range as the temperatures (16–34 °C) reported for impact milling of yellow pea, as measured by (Pelgrom et al., 2015a) under the same milling conditions. Approximately 3 mg of sample was loaded in the quartz basket, the RH was increased from 30% to 90% for adsorption and consecutively decreased to 10% for desorption with steps of 10%.

2.5. *Compositional analysis*

The dry matter (DM) content was determined by oven drying (105 °C) 1 g of sample for 48 h.

The protein content was determined by DUMAS analysis (rapid N exceed, Elementar, Langenselbold, Germany) with 6.25 as the nitrogen conversion factor (Pelgrom et al., 2015b). Protein recovery was calculated as the percentage of total grits protein recovered in the fine fraction (Equation (2)).

$$\text{Protein recovery } (\%) = \frac{\text{Amount of protein in the fine fraction (g)}}{\text{Amount of protein in the grits (g)}} \cdot 100\% \qquad (2)$$

The starch content was measured with a Total Starch Amyloglucosidase/a-Amylase Assay Kit (Megazyme International Ireland Ltd, Bray Ireland) based on the use of thermostable α-amylase and amyloglucosidase.

The oil contents of yellow pea and chickpea grits were determined with a Soxtherm SOX416 extractor (Soxtherm, Gerhardt, Germany). The samples (~2 g) were weighted, and the oil was extracted with an excess amount of solvent (Petroleum Ether 40–65 °C). First, a hot extraction was executed (25 min), followed by 5 cycles of solvent evaporation. The extraction time was set at 1 h and 35 min, followed by three evaporation cycles to distil off the bulk of the solvent. Lastly, the samples were dried in the equipment (30 min). The weight was determined after evaporation of residual solvent overnight.

2.6. *Particle analyses*

2.6.1. *Particle size distribution and particle dispersibility*

The particle size distribution of the flours was determined with laser diffraction in the Mastersizer 3000 with an Aero S dry dispersion unit, equipped with a standard venturi (Malvern Instruments, Worcestershire, UK). The hopper gap was 2.5 mm, and the feed rate was set to 50% to achieve a constant feed flow. Pressures of 4.0 bar and 0.5 bar were used to determine the particle size distribution, the extent of de-agglomeration (DA) and the dispersive index (DI) (Politiek et al., 2022).

2.6.2. *Particle flowability*

Powder flowability was characterised by an Anton-Paar MC502 rheometer equipped with a powder flow cell having a diameter of 50 mm (Anton Paar, Graz, Austria). A metal impeller ST36-2V-10/PCC (Anton Paar, Graz, Austria) with two rectangular-shaped blades (36 mm × 10 mm) was used to measure the powders' resistance to flow. The measured torque values are used for the semi-quantitative comparison of the powder flowability (Salehi et al., 2017; Schulze, 2008). For each measurement 30 g of flour was added to the powder flow cell and briefly stirred manually to mix the sample, using a spatula.

To assess the behaviour of cohesive powders, we adopted a dynamic measurement sequence that is based on a descending and ascending movement of the blade; a procedure that is also used in Freeman FT4 powder rheometers (Freeman, 2007). First, the metal impeller moves from the default measuring position (50 mm), towards the bottom of the powder cell, whilst rotating at 4 rpm and descending at 0.2 mm/s. When the metal bar reaches a height of 10 mm from the bottom, the descending stops, but it continues to rotate at 4 rpm for 200 s. The impeller then ascends at 0.2 mm/s with a rotation speed of 4 rpm until it is again at 50 mm. The measured torque (mN • m) is plotted against time for the consecutive intervals (Fig. 2).

The results were used as a semi-quantitative comparison of flow behaviour between the powders. The qualitative comparison of the flowability of powders in the dynamic regime is common and has been successfully applied to numerous materials (Francia et al., 2021a).

2.7. *Statistical analysis*

Data were collected and analysed in IBM SPSS Statistics 25 (IBM Corporation, Armonk, New York). To compare the means and evaluate the effect of processing conditions on the moisture content and powder flowability a one-way ANOVA test was carried out and a Tukey (homogeneous variance) or Games-Howell (inhomogeneous variance) post hoc test with a significance level of 0.05. For unequal sample sizes Games-Howell was used.

3. Results and discussion

3.1. *The sorption isotherms and moisture content change during milling and air classification*

Sorption isotherm curves of yellow pea and chickpea flour were measured to assess the moisture uptake of the two materials under

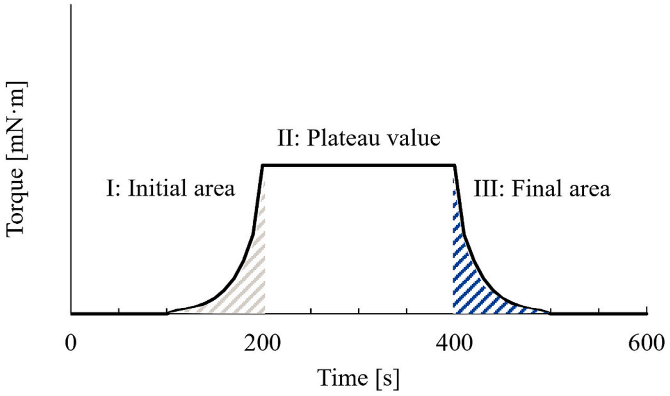

Fig. 2. Example of three intervals for rheology where I is the interval under the curve between 0 and 200 s, II is the plateau value and III is the area under the curve between 400 and 600 s.

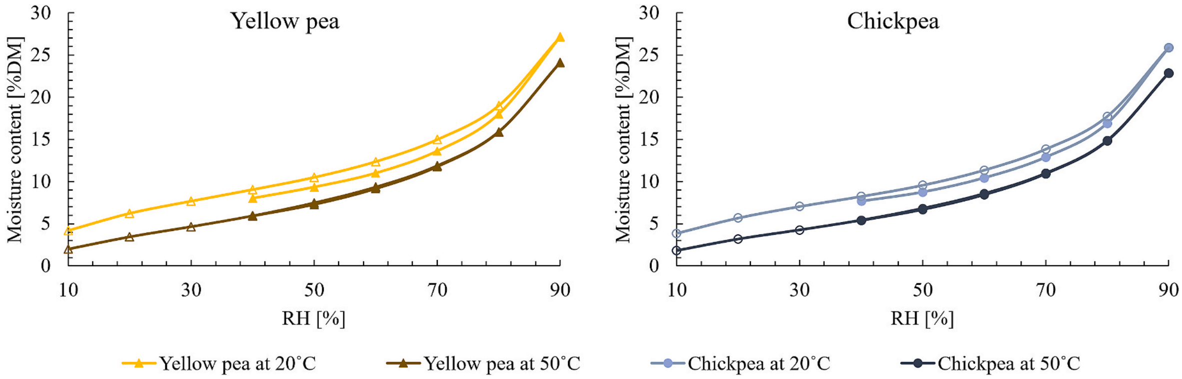

Fig. 3. Adsorption and desorption curves for yellow pea and chickpea flour at 20 °C and 50 °C. Closed symbols represent moisture adsorption, open symbols represent moisture desorption.

different environmental conditions (Fig. 3). The moisture content changed with around 7%DM (dry matter) between RH30 and RH70 for all measured samples. The moisture content decreased by around 2%DM for adsorption and by around 3%DM for desorption when the temperature increased from 20 °C to 50 °C. At each RH and temperature, yellow pea holds slightly more moisture than chickpea (0.3% at low humidity up to 1.3% at high humidity). So yellow pea flour is slightly more hygroscopic than chickpea flour, which is in line with previous research (Xu et al., 2019). The difference may be due to the difference in oil content between chickpea (5.2 ± 0.1%DM) and yellow pea (0.8 ± 0.3% DM), as seeds with a higher oil content equilibrate faster under the same conditions than seeds with lower oil contents (Davis, 1939; Suma et al., 2013).

The moisture contents of yellow pea and chickpea grits, flour and fractions were measured after milling, equilibration, and air classification at different RH (Table 1). Milling grits into flour resulted in a slightly larger decrease in moisture content at lower airflow (40 m^3/h) (Table 1). This is due to the combined effect of more moisture that goes through the equipment at higher airflows and a shorter residence time of the powder in the mill (Pelgrom et al., 2014). As expected, fine milled flours obtained or equilibrated at RH70 had a higher moisture content than materials obtained at RH30. Furthermore, the moisture content decreased by the release of water upon milling grits into flour, due to the heat generated and high airflow rates during milling. The increase in temperature results in a decrease in the relative humidity of the air (Singh, 2022). This RH decrease contributes to the drying of the grits by the release of moisture, upon milling. The chamber temperature during air classification was lower than during milling, so the actual relative humidity is expected to be higher upon air classification. This resulted in moisture absorption in the coarse fraction (from 8.0 to 10.4% moisture and from 10.7 to 11.6% moisture for a higher milling airflow) upon direct milling and air classification of yellow pea at RH70.

Overall, the effect of the equilibration of flour at RH70 on the moisture content after air classification was more pronounced than that of equilibration of grits, which indicates that moisture control in between fine milling and air classification is more important than moisture control in between pre-milling and fine milling. Furthermore, the coarse fractions had in general higher moisture levels than the fine fractions (Table 1). This could be attributed to the higher level of starch and lower level of protein in the coarse fraction (Appendix table A1), as starch is more hygroscopic than protein (Pelgrom et al., 2013b). The images and combined size and shape analysis with Morphologi 4 confirmed that the coarse fraction was indeed enriched with starch and the fine fraction was depleted of starch (Appendix IV).

Table 1

Moisture content based on the total mass of yellow pea and chickpea under different equilibration (Equ.) RH and different airflows upon processing (low airflow above the striped line and high airflow below the striped line). Values are presented as mean ± standard deviation between fractionation experiments if applicable. N.A. means "not available". n is the number of repetitions of dry fractionation experiments, which was 3 unless specified otherwise. Bold highlights fractions with a moisture content that was 2.4% higher or lower than the moisture content in the flour.

Crop	Material	Moisture content [%total mass]			
		Grits	Flour	Fine fraction	Coarse fraction
Yellow pea	Direct RH30	11.6 ± 0.2	7.8 ± 0.7	6.8 ± 0.8	8.7 ± 0.9
Airflow:	Direct RH70		8.0 ± 0.7	8.2 ± 0.4	**10.4 ± 0.5**
Milling 40 m^3/h	Equ. flour RH30		8.1 ± 0.4	6.7 ± 0.5	8.0 ± 0.8
Air classification 52 m^3/h	Equ. flour RH70		16.3 ± 0.6	**9.4 ± 0.8**	**12.7 ± 1.2**
	Equ. Grits RH30	8.8 ± 0.3	6.9 ± 0.3	6.8 ± 0.5	8.8 ± 0.8
	Equ. Grits RH70	16.8 ± 0.6	8.0 ± 0.6	7.8 ± 0.3	10.3 ± 0.7
Chickpea	Direct RH30	12.0 ± 0.2	8.3 ± 0.0[a]	**5.8 ± 0.3**[b]	7.8 ± 0.2[b]
Airflow:	Direct RH70		8.9 ± 0.2[a]	N.A.	N.A.
Milling 40 m^3/h	Equ. flour RH30		8.1 ± 0.2[b]	**5.6 ± 0.1**[a]	7.5 ± 0.1[a]
Air classification 52 m^3/h	Equ. flour RH70		16.8 ± 0.1[a]	**6.6 ± 0.1**[a]	**10.6 ± 0.1**[a]
Yellow pea	Direct RH30	11.6 ± 0.2	7.8 ± 0.2[a]	7.3 ± 0[a]	8.2 ± 0.1[a]
Airflow:	Direct RH70		10.7 ± 0.1[a]	9.0 ± 0[a]	11.6 ± 0[a]
Milling 52 m^3/h	Equ. flour RH30		8.8 ± 0[a]	7.1 ± 0[a]	8.9 ± 0[a]
Air classification 52 m^3/h	Equ. flour RH70		17.1 ± 0.1[a]	**9.8 ± 0.1**[a]	**13.7 ± 0**[a]
Chickpea	Direct RH30	12.0 ± 0.2	8.3 ± 0[a]	**5.9 ± 0.1**[a]	8.2 ± 0[a]
Airflow:	Direct RH70		10.1 ± 0.1[a]	7.9 ± 0.8[a]	11.0 ± 0[a]
Milling 52 m^3/h	Equ. flour RH30		7.7 ± 0[a]	6.1 ± 0.1[a]	8.0 ± 0[a]
Air classification 60 m^3/h	Equ. flour RH70		15.6 ± 0.1[a]	**7.8 ± 0.1**[a]	**10.2 ± 0.2**[a]

[a] n = 1.

[b] n = 2.

3.2. Particles aggregate upon equilibration at high humidity

This section elaborates on the agglomeration of particles for the different humidity conditions by particle size analysis after milling and equilibration of the flour. Laser diffraction with a venturi set-up was used to measure the particle size distribution of the flour. The venturi tube causes most agglomerates to break, which allows to study the effect of moisture on the primary particles (Schütz et al., 2019). Direct milling and equilibration of grits at RH30 and RH70 resulted in similar overall particle size distributions (Fig. 4) due to the heat generated, which leads to a decrease in air humidity inside the mill (Singh, 2022). This is in line with previous observations for soaked and untreated pea milled with an impact classifier mill (Pelgrom et al., 2015). The DV_{50} increased slightly after milling and storing the grits at different humidity conditions (Equ. Grits RH30 13.18 μm, direct milled grits RH30 and RH70 13.56 μm and Equ Grits RH70 14.09 μm), which was likely caused by the difference in moisture content (Table 1). The energy consumption in the mill overlapped between 576 and 720 kJ/kg. A reason for overlap in energy consumption is that the moisture content of the pea decreased inside the mill due to heating (towards 6.6 and 10.6 g moisture/100 g material), which was below the critical moisture content of 11% from where changes were previously observed (Dijkink & Langelaan, 2002a, 2002b). As the energy consumption overlapped, some of the milling energy might have been used to evaporate moisture, which slightly affected the DV_{50}. As moisture equilibration of the grits to a humidity of 70% did not result in differences in energy consumption, we further focussed on the effect of moisture equilibration on finely milled flour by storing it at RH70.

Equilibration of flour at RH70 resulted in a particle size distribution shift towards larger particle sizes for both chickpea and yellow pea flour (Fig. 4). This shift indicates that particles agglomerated upon equilibration. The slight particle size increase was qualitatively confirmed with image analysis with Morphologi 4, by the visual observation of agglomerates and a shift in volume count towards larger particles (Appendix IV). The agglomeration is caused by the formation of liquid bridges at the contact points of the grains at this high RH, as the powder is in the pendular state (Mitarai and Nori, 2006; Schütz et al., 2019). For chickpea, the shift of equilibrated flour at RH70 was more pronounced than for yellow pea, as also high peaks were observed at ~1600 μm for both airflows. So, it is likely that lipids also play a role in the formation of liquid bridges, which can explain the difference between chickpeas and yellow peas.

The degree of de-agglomeration (DA) and the dispersive index of the fine particles (DI) were evaluated for the relative humidity conditions used in this study. Both are a measure of the air classification capability, where a higher DA and DI are favourable for the process performance (Dijkink et al., 2007; Pelgrom et al., 2015b). These two measures for particle agglomeration are based on the DV_{50} (for DA) and the particle size below 10 μm (for DI) at full dispersion (4 bar) and a lower dispersion pressure (0.5 bar). At a low humidity (28–45%) the DA was above 0.78 and not influenced by equilibration at RH30 (Fig. 5), which was likely because the particles were in a dry state, where there is only weak cohesion between particles (Danov et al., 2018). Equilibration at RH70 of flours milled at a lower airflow (40 m^3/h) slightly decreased the DA, which might result in a slightly decreased air classification performance, while equilibration at RH70 did not influence the DA at higher milling airflow (Fig. 5A). The DA was still above 0.6 for all samples, which indicates that the overall collected flours were still well dispersible, irrespective of their water content.

The dispersive index (DI) of the small particles was similar for equilibration at 30% humidity and decreased upon equilibration of fine milled material at 70% humidity for both airflows and both materials (Fig. 5B). This was caused by agglomeration between the small particles with larger particles at a higher humidity (Appendix figure A5). At a high dispersion pressure, the small particles might still be dispersed from the larger particles. However, at a lower dispersion pressure of 0.5

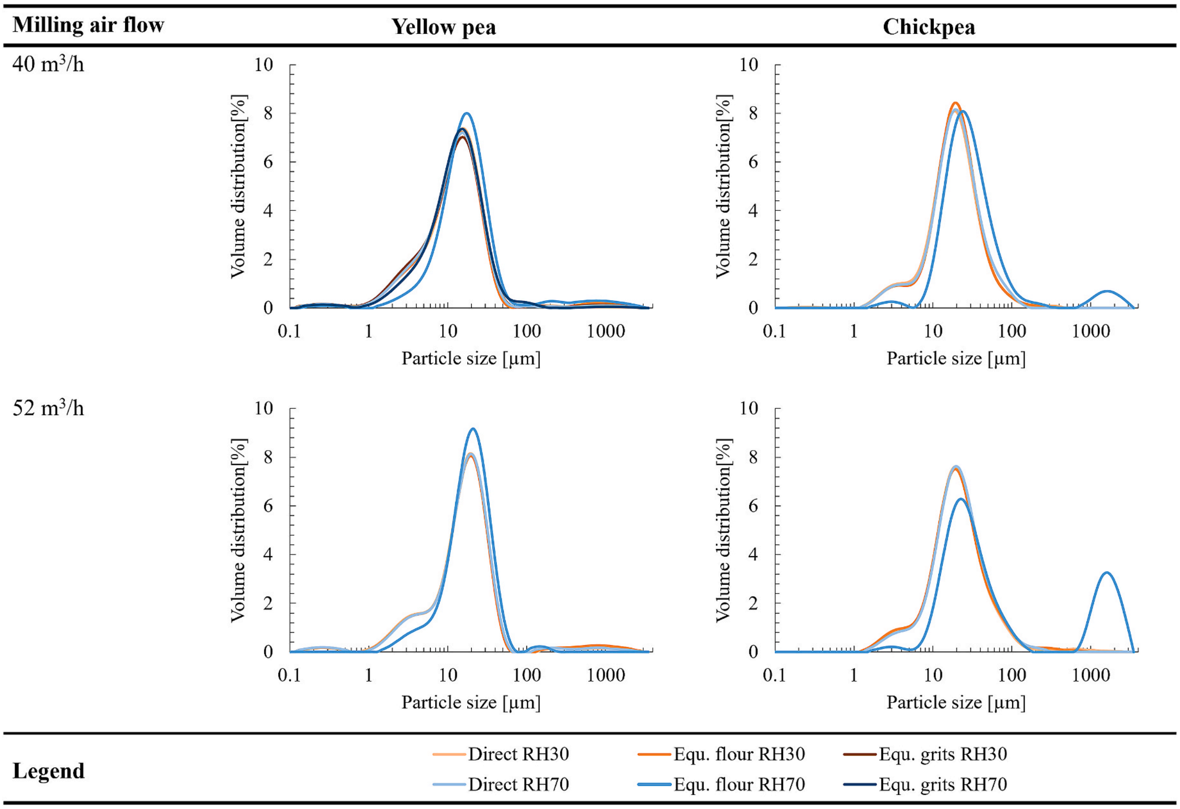

Fig. 4. Average particle size distributions of yellow pea and chickpea flour milled with airflows of 40 m^3/h and 52 m^3/h. The equilibration conditions used for the specific samples are shown in the legend.

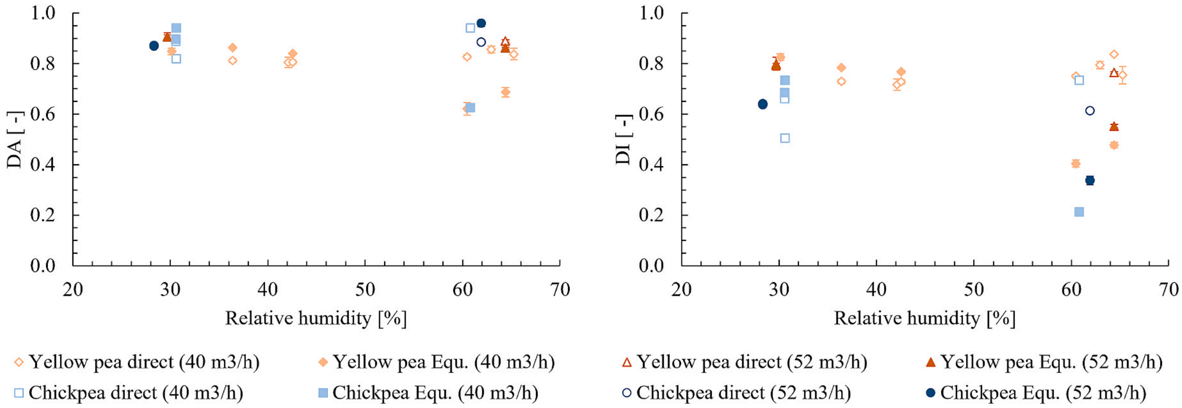

Fig. 5. Degree of de-agglomeration (DA) and dispersive index (DI) versus the relative humidity upon milling for yellow pea and chickpea at different conditions identified in the figure legend. Open symbols stand for directly milled flours and closed symbols are equilibrated (Equ.) samples. The milling airflows of the samples (40 and 52 m^3/h) are shown between brackets. Error bars show the standard deviation in DA and DI.

bar the particles remain attached to the larger particles as the force to disperse the particles from each other is too low to overcome the other forces between the particles. Equilibration at 70% humidity of samples milled with a higher airflow (52 m^3/h) resulted in a better particle dispersion than equilibration of samples milled with a lower airflow (40 m^3/h) (Fig. 5), which might be due to higher van der Waals forces that act on the smaller particles obtained after milling at 40 m3/h (Pelgrom et al., 2014).

Overall, the small particles were better dispersible in the air when milled at a higher airflow and the dispersive index was more affected by the equilibration conditions than the degree of de-agglomeration, especially at higher humidity (70%). The lower DA and DI for equilibrated samples at RH70 might be indicators for a decrease in air classification performance upon storage at higher humidity, which is discussed in the next section.

3.3. Airflow and high storage humidity influence milling and air classification performance

The milling performance is evaluated by the flour yield and composition, and the air classification performance is assessed by the fine and coarse fraction yields and compositions. The overall process performance is evaluated by the protein recovery in the fine fraction. Here we did not focus on the oil content, as the oil bodies are not selectively separated in air classification processes. First, the effects of humidity and airflow on milling yield, flour- and fraction composition are evaluated. Subsequently, the effect of airflow and humidity on the fine and coarse fraction yields is described and connected to the results from section 3.2 on particle agglomeration and visual observations in the classifier chamber. Lastly, the overall process performance (milling + air classification) is evaluated by the protein content and protein recovery in the fine fraction.

The milling yield was not influenced by the relative humidity (30–70%) during processing (Fig. 6A). The milling yield of chickpea (75–93%) was comparable to literature (88%) (Xing et al., 2020) and the

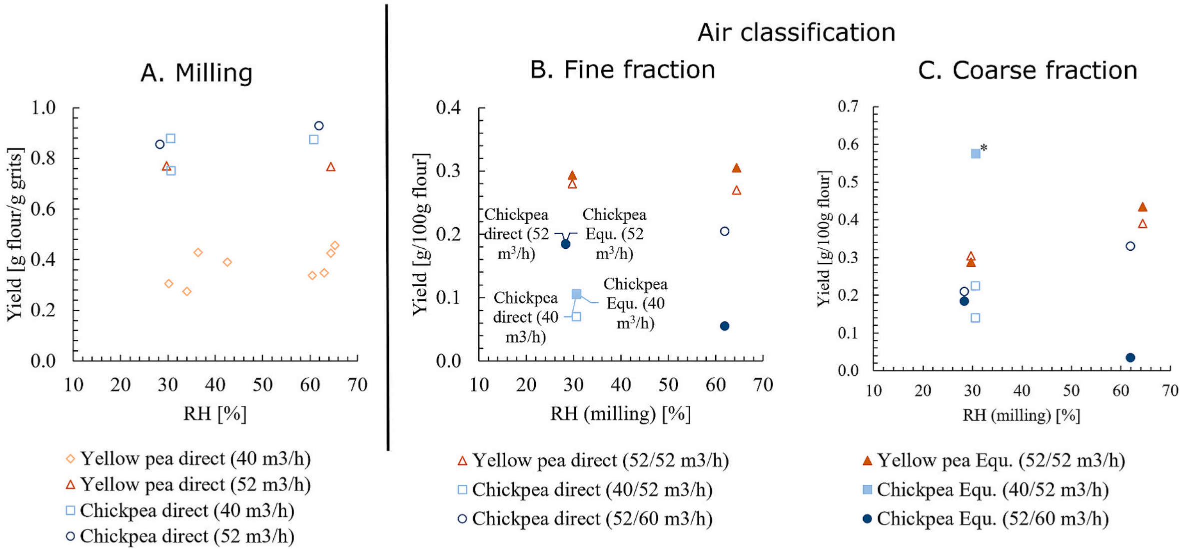

Fig. 6. Milling yield (A), fine fraction yield (B) and coarse fraction yield (C) of yellow pea (▲/◆) and chickpea (●/■) against actual average relative humidity (RH) during the milling. The conditions are specified in the figure legend. Moisture loss (Table 1) is considered as yield loss. Open symbols are directly milled, and air-classified flours and closed symbols are equilibrated samples between milling and air classification. The milling airflow of the samples (40 and 52 m^3/h) is the first value shown between the brackets and the air classification airflow (52 and 60 m^3/h) is the second value shown between the brackets. For air classification, samples with a milling yield above 60% are presented, and samples with a milling yield below 60% are included in Appendix figure A8. *Indicates that the batch size for air classification was higher (750 g).

milling yield of yellow pea milled with an airflow of 52 m^3/h. For yellow pea, the higher airflow (52 m^3/h) doubled the milling yield (~77%) compared to the lower airflow (40 m^3/h) with yields between 20 and 45% (Fig. 6A). The low milling yield for yellow pea milled at 40 m^3/h was not expected based on previous research, where milling yields of around 88% were reported for a similar airflow of 40 m^3/h (Xing et al., 2020). The lower yield was caused by insufficient emptying of the milling chamber (Appendix figure A2).

The composition of yellow pea flour and the obtained fractions was mainly influenced by the used airflow, rather than the humidity conditions (Appendix table A1). A higher airflow (52 m^3/h) resulted in lower protein contents (28.1%DM versus 39.6%DM) and higher starch contents (45.9%DM versus 29.8%DM) in the flour than a lower airflow (40 m^3/h) (Appendix table A1). A similar observation was reported in previous research, where the protein content of yellow pea flour milled at 40 m^3/h was 32.4%DM and starch content 36.6%DM, whereas pea flour milled at 52 m^3/h had a protein content of 19.4%DM and starch content of 47.6%DM (Pelgrom et al., 2015a; Xing et al., 2020).

The increase in airflow resulted in a slightly higher starch and lower protein content in the fine fraction of chickpea. This change is due to the shift of the cut-point to larger particle sizes, which allowed more small starch particles to pass the classifier wheel together with the protein (Appendix table A1). In general, air classification of chickpea results in lower protein contents in the fine fraction than air classification of yellow pea and other legumes such as fababean (62.6% protein) or lentil (63.0% protein) (Pelgrom et al., 2015b; Sosulski and Youngs, 1979; Xing et al., 2020). The lower protein content in the fine fraction has been attributed to the higher oil content and the smaller starch granules of chickpea than other starch-rich legumes (Pelgrom et al., 2015b; Sosulski and Youngs, 1979; Xing et al., 2020).

The fine and coarse fraction yields in Fig. 6B and C represent only samples with higher milling yields (>60%), the other fine and coarse fraction yields of yellow pea are included in Appendix figure A8. Direct air classification of chickpea with a higher airflow (60 m^3/h) resulted in an increased fine fraction yield compared to air classification at 52 m^3/h, caused by the increased drag force through the classifier wheel. A higher airflow resulted in similar or higher coarse fraction yields for chickpea, so overall the air classification loss was reduced by increasing the airflow (Fig. 6).

The fine fraction yield was not influenced by humidity and storage conditions at low humidity upon air classification (Fig. 6), which was expected as the degree of de-agglomeration and dispersive index at low humidity had similar values with and without equilibration (Fig. 5). The coarse fraction yield of yellow pea slightly increased with relative humidity, which was observed for both milling airflows of 40 m^3/h and 52 m^3/h (Fig. 6B; Appendix figure A8). This indicates that, even though the dispersive index of equilibrated material at RH70 was lower for equilibrated yellow pea (0.55) than directly processed yellow pea at RH70 (0.76), it did not negatively affect the overall air classification yields. The slightly higher coarse fraction yield might be caused by a slightly higher gravitational force than the drag force for a larger number of particles at higher humidity (Shapiro and Galperin, 2005). The higher gravitational force was induced by the uptake of water by the particles in equilibrated flour at RH70, which increases the particle density and by a lower drag force i.e., due to a slightly lower air density at higher humidity.

Contrary to yellow pea, equilibration of chickpea flour at RH70 (DI = 0.34) was detrimental to the fine and coarse fraction yields. The analysis of the yield was not even possible for air classification of chickpea that was milled at an airflow of 40 m^3/h equilibrated at RH70 (DI = 0.21) and air-classified at 52 m^3/h, as the accumulated material completely blocked the inlet of the classifier chamber (Fig. 7A). So, the lower yields were caused by the higher adhesive (particle-wall) and agglomeration (particle-particle) forces than the gravitational force. Furthermore, a visual inspection of the classifier chamber showed that the amount of material that accumulated at the top of the chamber depended on the material, processing- and equilibration conditions. For example, yellow pea showed much less adhesion and agglomeration at the top of the chamber upon air classification than chickpea (Fig. 7C and D). It was observed that the accumulated flour disappeared when a certain mass of powder accumulated and fell due to its weight. This also occurred for equilibrated chickpea flour at RH30, where the use of a larger batch size resulted in an increased coarse fraction yield, while the fine fraction yield remained unaffected (Fig. 6). For larger scale systems the yield of the coarse fraction is thus expected to be higher.

To enable cross-comparison of the overall process in terms of yield and protein content, we consider protein recovery, which is defined as the ratio between the protein in the fine fraction and protein in the grits (Equation (2)). In the most ideal scenario, one would like to have both high protein content and high protein recovery. The milling yield, fine fraction yield and the protein content of the fine fraction all influence the protein recovery. The protein content was plotted against the protein recovery, where a horizontal black dashed line was used to separate the chickpea results (bottom part, protein content <52.5%DM) and the yellow pea results (upper part, protein content >52.5%DM) (Fig. 8). The protein recovery of yellow pea increased with a higher airflow during milling, without any significant compromise on the protein content (upper part of Fig. 8). This higher protein recovery was attributed to the elevated milling yields of yellow pea milled at a higher airflow (52 m^3/h). The protein recovery was also comparable to earlier observations for yellow pea milled at 40 m^3/h (49%) (Xing et al., 2020). No clear impact of relative humidity on protein recovery was seen for yellow pea.

A higher milling and air classification airflow improved the protein recovery of chickpea and slightly reduced the protein content (bottom part of Fig. 8). The protein recovery of chickpea milled (52 m^3/h) and air-classified (60 m^3/h) at RH30 or direct at RH70 was at the higher end of values reported in literature (11–31%) as summarised by (Boukid, 2021). However, the protein recovery of chickpea decreased upon equilibration of the flour at RH70, while the protein content remained similar. In the case of chickpea, the lower protein recovery was caused by a low fine fraction yield. The fine fraction yield was lower as more mass accumulated in the classifier chamber after equilibration, as

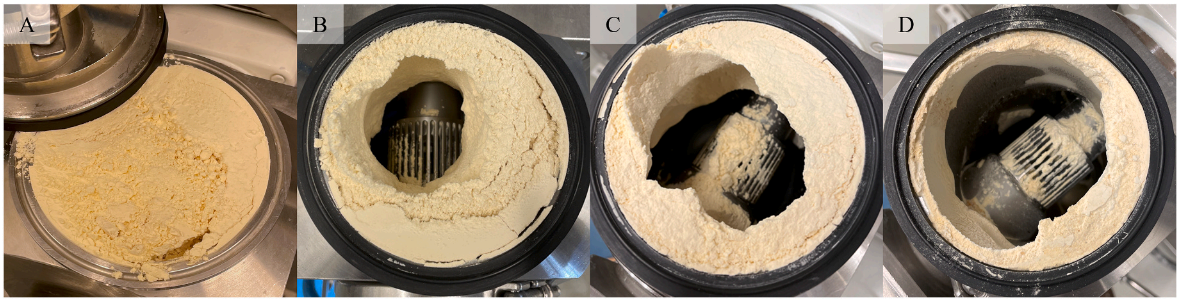

Fig. 7. Examples of accumulation of flour in the classifier chamber upon air classification for chickpea equilibrated at RH70 milled at 40 m^3/h (A) and 52 m^3/h (B), direct air-classified chickpea milled at 52 m^3/h (C) and yellow pea milled at 52 m^3/h (D). The milling speeds are shown between brackets.

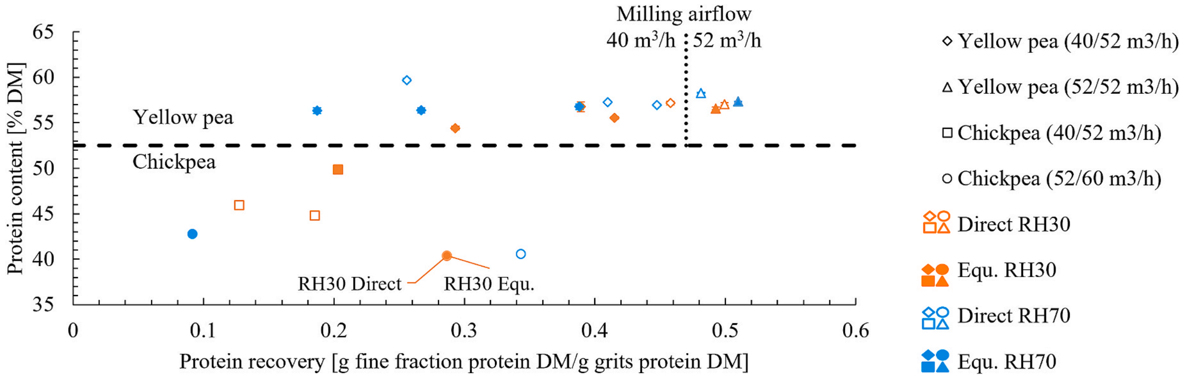

Fig. 8. Protein content and protein recovery of yellow pea (above black dashed line) and chickpea (below black dashed line). Open symbols represent directly milled and air-classified samples, and closed symbols represent equilibrated samples. Samples with a target RH of 30% are coloured orange and samples with a target RH of 70% are coloured light blue. The shapes in the legend specify the crop and airflow used, which is shown between brackets: the first value gives the milling airflow (40 or 52 m^3/h), and the second value gives the air classification airflow (52 or 60 m^3/h). The milling airflows of yellow pea are separated with the small dotted line (left 40 m^3/h, right 52 m^3/h) The error bars represent the protein content standard deviation. Two symbols of chickpea overlapped, which are highlighted with data labels.

illustrated in Fig. 7B and C. Thus, the storage of chickpea flour at a high relative humidity negatively affected the overall protein recovery.

3.4. Lower flowability of chickpea flour than yellow pea flour

In general, the air classification yields, and protein recoveries were lower for chickpea than for yellow pea, which was consistent with the fouling visually observed after the air classification of chickpea. Based on the locations of the fouled material, it is proposed that when chickpea flour enters the classifier chamber through the feed inlet it accumulates at the inlet at the top of the chamber, instead of being directly dispersed and carried by the airflow for classification (Fig. 7). In previous research, the accumulation of flour and the impaired separation have been related to a higher cohesion, and consequently poorer flowability of legume powders with higher oil contents (Gueguen, 1983; Pelgrom et al., 2014; Sosulski and Youngs, 1979).

In fluidized systems, such as air classifiers, cohesive powders can accumulate in corners or on surfaces within the equipment (Krantz et al., 2009). Powder rheology was used to describe if chickpea flour indeed has a higher tendency for particle cohesion than yellow pea flour. The torque responses over time for the equilibrated flours at RH70 are shown in Fig. 9. An overview of all individual measurements, at RH30 and RH70 for both the direct and equilibrated samples, is shown in the Appendix (Appendix figure A9). A higher torque response means that the impeller requires more force for the same movement, which indicates a poorer flowability.

The torque response of the chickpea sample rises more steeply than that of the yellow pea sample under similar conditions. This follows the hypothesis of higher cohesiveness. However, instead of a stable plateau value, the torque declines between 200 and 400 s, which coincides with the visual observation of heap formation at the side of the measurement cylinder. This suggests that variations occur in the amount of material that is displaced by the impeller, which directly affects the measurements. The average height of the torque response alone might thus not be sufficient to capture the flow behaviour of cohesive powders. It was, therefore, decided to calculate and compare several characteristic values; a) the area under the curve up to 200 s (referred to as Area I), b) the average plateau value between 200 and 400 s, c) the difference in torque between the initial average of the plateau (p_i) (200–205 s) and the final average of the plateau (p_f) (395–400 s), d) the area under the curve between 400 and 600 s (referred to as Area III) and e) the difference between Area I and Area III. The determined values for the different samples are summarised in Table 2. The values are only used for a semi-quantitative comparison, as they are not related to a physical quantity such as cohesion strength or viscosity. Progress has been made to estimate the shear stress and shear strain, i.e. physical quantities, inside FT4 powder testers by combining DEM simulations and experiments (Hare and Ghadiri, 2017; Khala et al., 2022). However, a qualitative comparison of measured values is still standard for powder rheometers (Francia et al., 2021b).

The initial torque response (area I) was lower for yellow pea than for chickpea (Table 2). For yellow pea, there were no differences between yellow pea samples milled or stored at RH30 and RH70, while for chickpea the sample equilibrated at RH70 showed a higher Area I than the other chickpea samples. The higher torque measured for chickpea equilibrated at RH70 may be related to liquid bridging between the particles. If the particles are in a dry state, there is only weak cohesion between particles via van der Waals forces. Upon an increased liquid

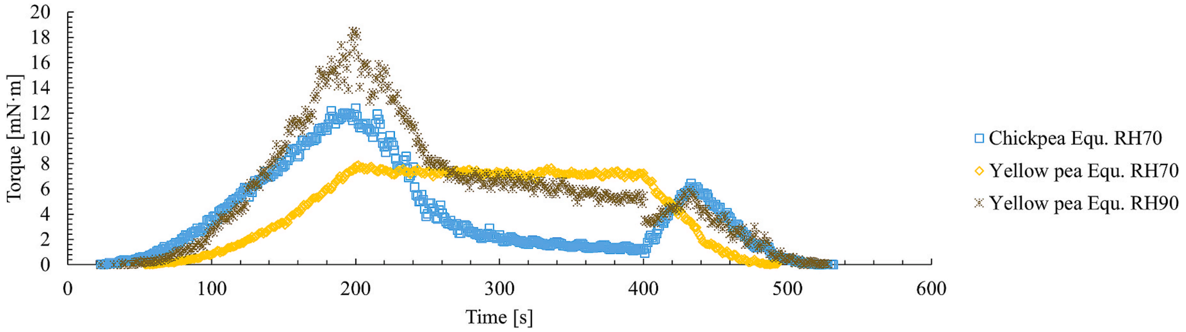

Fig. 9. Average measurement of powders' resistance to flow with the Anton Paar powder flow cell for yellow pea equilibrated at RH70 (gold), chickpea equilibrated at RH70 (light blue), and yellow pea equilibrated at RH90 (dark gold).

Table 2
Averages and standard deviation from rheological cohesion measurements for yellow pea and chickpea at different storage conditions. The dotted lines separate the different storage conditions, an additional sample was added for yellow pea equilibrated at RH90. Different letters indicate significantly different samples. **Bold** highlights the higher Area I and the difference in plateau value (p_i-p_f). These samples also showed a decreased air classification performance.

Crop	Condition	Area I [mNm·s]	Average plateau value [mNm]	$p_i - p_f$ [mNm]	Area III [mNm·s]	Area I - Area III [mNm·s]
Yellow pea	Direct RH30	454 ± 16^a	10.9 ± 0.1^{de}	0.0 ± 0.3^a	182 ± 26^a	271 ± 10^{ab}
	Equ. Flour RH30	453 ± 41^a	9.6 ± 0.6^{cde}	0.8 ± 0.8^a	250 ± 24^a	203 ± 17^a
	Direct RH70	463 ± 14^a	8.55 ± 0.7^{cd}	1.0 ± 1.2^a	233 ± 62^a	231 ± 58^a
	Equ. Flour RH70	389 ± 57^a	7.3 ± 0.9^{bc}	0.5 ± 0.3^a	214 ± 54^a	175 ± 15^a
	Equ. Flour RH90	$\mathbf{1014 \pm 29^c}$	7.9 ± 0.1^{bcd}	$\mathbf{10.6 \pm 1.3^b}$	315 ± 58^a	699 ± 29^e
Chickpea	Direct RH30	652 ± 4^b	12.1 ± 0.1^e	0.0 ± 0.2^a	295 ± 4^a	357 ± 1^{bc}
	Equ. Flour RH30	667 ± 10^b	11.0 ± 0.3^{de}	-1.1 ± 0.8^a	259 ± 11^a	409 ± 1^c
	Direct RH70	679 ± 36^b	11.9 ± 0.1^e	0.7 ± 1.8^a	323 ± 32^a	356 ± 4^{bc}
	Equ. Flour RH70	$\mathbf{907 \pm 46^c}$	3.7 ± 0.4^a	$\mathbf{9.9 \pm 1.9^b}$	325 ± 64^a	582 ± 18^d

content, the powder will enter the pendular state. In the pendular state particles cluster by directly adhering to each other due to the wetting forces exerted by capillary bridges (Danov et al., 2018; Strauch and Herminghaus, 2012). In the case of equilibrated chickpea flour at RH70, a combination of the oil and elevated moisture content may have encouraged the formation of capillary bridges, which resulted in stronger cohesive forces and thus a higher initial torque response.

In literature, it was observed that storage of a wheat starch protein mixture at RH90 resulted in a lower protein recovery (Dijkink et al., 2007). Therefore, yellow pea flour was also stored at RH90 and subjected to rheological measurements after equilibration at RH90. After equilibration at RH90, the moisture content of the yellow pea flour was 21.3 ± 0.1%, which is comparable to the total liquid content (oil + moisture) of chickpea (21.8 ± 0.3 g/100 g flour). The flowability of yellow pea equilibrated at RH90 (total liquid content 22.3 ± 0.7 g/100 g flour) showed comparable results to chickpea equilibrated at RH70 (Fig. 9, Table 2), which indicates that the total liquid content of the material can affect the powder properties.

The average plateau values were less conclusive between the different flours and storage conditions, apart from the lower value for chickpea flour equilibrated at RH70 (Table 2). As previously stated, the impeller dug a hole inside the powder bed, which strongly reduced the torque requirement for continuous stirring at a fixed height. The effect on the torque curve is characterized by $p_i - p_f$, which was, only different for equilibrated chickpea flour at RH70 and yellow pea flour at RH90, which had also a higher Area I. There are no significant differences between yellow pea and chickpea flour samples when comparing Area III, which means that the difference between Area I and Area III does not provide any other benefit over analysing Area I.

Overall, the rheological measurements showed that chickpea flour has a lower flowability and thus higher cohesion than yellow pea flour at similar storage conditions, especially after storage at RH70. To improve the air classification of chickpea flour, one can alter process-related parameters (e.g. airflow, humidity, classifier wheel speed) and the material properties (e.g. by de-oiling, toasting or addition of flowability aids) (Dijkink et al., 2007; Doğan et al., 2019; Kim et al., 2005; Pelgrom et al., 2014; Pelgrom et al., 2015b; Pelgrom et al., 2015). Toasting or increasing the classifier wheel speed may have the disadvantage that oil is released from within the particles to the surface, which reduces the flowability (Doğan et al., 2019; Kim et al., 2005; Pelgrom et al., 2014). To evaluate if the powder properties improve by a pre-treatment, a comparison of the torque responses in Area I is most useful. However, these results are ideally supplemented with the difference in plateau values ($p_i - p_f$), to provide more information on the powder bed behaviour inside the measurement chamber of the flow cell. This is especially relevant if the influence of heap formation on the torque response in Area I is not known.

4. Conclusion

The effect of relative humidity on milling and air classification of yellow pea and chickpea was systematically investigated by varying the humidity. The milling yield was not influenced by the relative humidity in the tested range (30–70%) as drying of the material in the mill overshadowed this possible effect. Particle agglomeration, dispersion and flowability were assessed via particle size analysis and powder rheology, and subsequently linked to the observed air classification performance.

Small particles were better dispersible when milled at a higher airflow (52 m^3/h versus 40 m^3/h), which resulted in higher protein recoveries for both yellow pea and chickpea. The dispersive index (DI) (particles <10 μm) was more affected by the equilibration conditions than the degree of de-agglomeration (based on DV_{50}), especially at higher humidity (70%), where the DI decreased after equilibration. Equilibration at a humidity of 30% did not affect the separation performance in terms of yield, purity, and protein recovery, while equilibration of chickpea flour at RH70 was detrimental for the fraction yields and protein recovery in the fine fraction. Although yellow pea flour equilibrated at RH70 had a slightly lower DI value, the overall air classification performance remained unaffected. Still, a lower DI shows that there is a risk for a decrease in air classification performance. Next to particle dispersion, other factors are still important for the protein recovery upon air classification, such as airflow, classifier wheel speed and difference in size between the materials.

Powder flowability was studied with powder rheology and we evaluated which parameters were most relevant to define differences in flowability. The area of the torque in the descending time region (Area I) was most useful to conclude on flowability differences. However, if the effect of heap formation on the torque response in this area is unknown, it is relevant to supplement this with the difference in initial- and final plateau value. Overall, chickpea flour had a worse flowability than yellow pea flour under similar conditions, especially after storage at RH70. Equilibration of yellow pea at RH90 resulted in more comparable total liquid contents (oil + moisture) and a similar rheological profile as chickpea equilibration at RH70. The combined oil and elevated moisture content may have encouraged the formation of capillary bridges and the change of a dry powder to a powder in the pendular state, which results in stronger cohesive forces. The flours with lower flowability showed a decrease in protein recovery after air classification.

Overall, the dispersive index, particle shape and rheological properties could be used to explain the differences in air classification performance. Using the rheological properties together with particle size and shape analysis as qualitative indicators for the process performance will help industry with the search towards obtaining optimal protein recoveries. Extremer conditions resulted in a decreased dispersive index, agglomerated particles, and a lower particle flowability. A lower flowability and agglomerated particles were detrimental to the air classification process, which results in more fouling and consequently a high loss of material. So, a humidity above 70% should be prevented for robust air classification.

Author's contribution

R.G.A. Politiek: Conceptualization, Data curation, Investigation, Methodology, Validation, Visualization, Writing – original draft. **S. He**: Investigation, Data curation. **P.F.C. Wilms**: Methodology, Conceptualization, Writing – review & editing. **J.K. Keppler**: Supervision, Writing – review & editing. **M.E. Bruins**: Conceptualization, Supervision, Writing – review & editing. **M.A.I. Schutyser**: Project administration, Conceptualization, Supervision, Writing – review & editing.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

Data availability

Data will be made available on request.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jfoodeng.2023.111663.

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