Journal of Food Engineering 334 (2022) 111149

Contents lists available at ScienceDirect

Journal of Food Engineering

journal homepage: www.elsevier.com/locate/jfoodeng

# Effect of oil content on pin-milling of soybean

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ARTICLE INFO

*Keywords:*
Energy consumption
Bond's law
Dispersibility
Agglomeration
Size reduction
Pin milling

ABSTRACT

Milling is a critical step to prepare plant-based food ingredients by dry fractionation. It should provide a dispersible flour of finely milled particles composed of different cellular substructures. Especially, for raw materials with higher oil content such as soybean, this is challenging. We present an investigation on the effect of oil content on milling yield, particle size, energy use and flour dispersibility upon pin-milling of soybean. Soybean (20 g oil/100 g dry solids) and mechanically de-oiled soybeans (9–17 g oil/100 g dry solids) were subjected to pin-milling experiments. Increasing soybean oil content limited milling to smaller particles and lowered the overall milling yield. Particle size reduction can be described with an adapted Bond's model with oil content as an input parameter. The produced soy flours were well dispersible.

## 1. Introduction

The market and popularity of plant proteins is growing rapidly due to an increased awareness of consumers of the health benefits and sustainability of a more plant-based diet (Hertzler et al., 2020). Common plant-based protein sources are grains, pulses and oilseeds (Tabtabaei et al., 2016). For many food applications it is desired to work with plant protein concentrates or isolates, which can be achieved by wet or dry fractionation. Prior to both processes, usually milling is applied to increase the protein extractability of the raw material. During wet fractionation milling would likely follow oil extraction, alkaline/acid extraction, centrifugation, pH precipitation or ultrafiltration and lastly drying (Chéreau et al., 2016; Schutyser, Pelgrom, van der Goot and Boom, 2015). For dry fractionation, milling is followed by a dry separation step, for example sieving, air classification or electrostatic separation (Schutyser et al., 2015). Milling prior to dry fractionation is a critical step, as it enables liberation of cell structures of different composition into particles of different size, shape and density to aid air classification and/or liberate particles with different tribo-electrostatic charging properties to aid electrostatic separation. Unfortunately, milling of oilseeds such as soybean is very challenging in contrast to starch containing legumes due to the presence of oil. The focus of this study is on the milling behaviour of oilseeds, in particular soybean, which is an oilseed that is also extremely rich in protein (Chéreau et al., 2016).

Native soybean seeds have a particular morphological structure (Fig. 1), which to great extent determines disclosure of cell components into individual particles during dry milling. Crude soybean seeds consist of two main parts, the hull (seed coat or testa) and the inner cotyledon matrix (Fig. 1A and B). Soy hulls are a rich source of carbohydrates (86 g/100 g dry hull) (Medic et al., 2014). Soy cotyledon cells have an average width of 30–50 μm and a length of 70–80 μm, in which protein bodies (8–10 μm) and oil bodies (0.2–2 μm) are embedded as illustrated in Fig. 1C and D (Campbell and Glatz, 2009; Rosenthal et al., 1998). During milling, these intact cotyledon cells need to be disrupted to enable protein extraction by either wet or dry fractionation. The protein and oil yields during subsequent separation are found closely related to the low presence of intact cells (Campbell and Glatz, 2009; Russin et al., 2007). Material such as soybean intended for fractionation is usually dehulled prior to milling. In our experiments we use hulled soybeans, as cold-pressing (40 °C) of hulled seeds has resulted in higher oil extraction yields than for de-hulled seeds and a dry separation method like sieving or electrostatic separation can separate the fibrous hull constituents (Koubaa et al., 2016; Wang, Suo, De Wit, Boom and Schutyser, 2016a). The presence of hull pieces might cause a scouring effect during milling, especially close to the pins, which could result in less sticking of the flour. However, the presence of the hulls may also negatively affect the milling efficiency and thus result in slightly larger particle sizes (do Carmo et al., 2020). Dehulling will increase the protein content in the starting raw material for dry fractionation but has been found not to influence the protein-enrichment during air classification (do Carmo et al., 2020).

Too fine milling can impair the subsequent separation for dry

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https://doi.org/10.1016/j.jfoodeng.2022.111149
Received 17 March 2022; Received in revised form 16 May 2022; Accepted 19 May 2022
Available online 25 May 2022

fractionation as was for example found for flours of lupin and de-oiled soy bean (Pelgrom, Berghout, van der Goot, Boom and Schutyser, 2014; Xing et al., 2018). If flours contain very small particles, large van der Waals forces between particles contribute to poor dispersibility and thus poor dry separation (Pelgrom et al., 2014). In addition, for oil-rich crops the presence of oil can induce cohesive forces between powder particles via bridging (Pelgrom et al., 2014; Xing et al., 2018). To prevent particle cohesion through oil bridging, often de-oiling, also referred to as defatting, is applied prior to milling (Basset et al., 2016; Chéreau et al., 2016; Laguna et al., 2018; Pelgrom et al., 2014; Xing et al., 2018). Furthermore, oil removal can provide increased oxidative stability of the material, and soybean oil is an important resource by itself (Berghout et al., 2015). However, the current trend goes towards less processed materials, using the whole flour to reduce material loss and energy use (Berghout et al., 2015). Partial de-oiling is an interesting approach to retain the relatively stable oil bodies in the final product, as was observed for cold-pressed (<45 °C) sunflower cake (Karefyllakis, Octaviana, van der Goot and Nikiforidis, 2019). Partial de-oiling will decrease the extracted oil yield and the remaining oil might still cause oxidation in the flour. However, the presence of oil bodies can be beneficial for the functionality of a product (Berghout et al., 2014; Berghout et al., 2015). For example, the presence of oil bodies in ingredients for meat analogue production did not result in oil leakage, while addition of oil afterwards did (Peng, 2021). Therefore, it is desirable and relevant to study milling behaviour of both partially de-oiled soy and whole soybeans.

The aim of the current study is to systematically investigate the effect of soy oil content on pin-milling behaviour and flour properties like particle size distribution and flour dispersibility. We specifically investigate how oil content influences powder flow and milling behaviour. Hitherto, the majority of milling studies in scientific literature focussed on milling of inorganic materials and a minority on milling of food materials (i.e. rice grain, dried coriander seeds and soy with different moisture contents) (Lee et al., 2013; Loubes et al., 2022; Shashidhar et al., 2013).

Differences in initial oil content were achieved by mechanical de-oiling of soy and their effect was investigated on fine milling. We investigated particle size reduction of both soy and (partially) de-oiled soy by impact milling with a pin-mill at different rotation speeds. The pin-mill was chosen for practical reasons: easiness to clean, less prone to clogging compared to a classifier impact mill, simultaneous particle size reduction of both the hull and cotyledon to a similar particle size (Maskus et al., 2016), and no recirculation of material through the mill by a classifier wheel. Bond's empirical model was used to describe and compare the grinding kinetics of soy and de-oiled soy. It should be noted that Bond's model is a basic empirical model based on one specific size modulus, which is sufficient for the purpose of this study, but for scale-up procedures more advanced population balance models are recommended (Herbst and Fuerstenau, 1980). We determined flour yield and dispersibility after milling. The results were extensively discussed in relation to their influence on dry separation.

## 2. Materials and methods

### 2.1. Materials

Dry Canadian hulled soybeans (Glycine max, Batch 20–037) were obtained from Frank Food Products (Twello, The Netherlands). The soybeans were stored in vessels closed with a screw cap at 4 °C. Petroleum Ether with a boiling range between 40 and 65 °C was obtained from Avantor Performance Materials B.V. (J.T. Baker, Deventer, The Netherlands). L-Aspartic acid (10.52% Nitrogen determined by Elementar Germany) was obtained from Sigma (Sigma Aldrich, Darmstadt, Germany).

### 2.2. Sample preparation via de-oiling and milling

Oil was removed from part of the soybeans by using a single-screw oil press (KK 20F Universal) from Kern Kraft (Reut, Germany). The oil press was chosen to avoid the use of solvents during de-oiling and to keep intact oil bodies in the product. The temperature was kept constant at 60 °C, to keep the temperature below the denaturation temperature (70 °C) of the major soy storage proteins in soy (Peng et al., 2016; Xing et al., 2018). A standard screw was used, rotating at 20 rpm. A hard seed extraction unit was mounted around the standard screw followed with a pre-die. Three main die sizes were used with a diameter of 14, 16 and 18 mm. This resulted in material throughput rates of respectively 1.3, 2.5 and 9.4 kg/h.

Both the soybeans and oil pressed soybeans were pre-milled into grits with a LV 15M pin-mill (Condex-Werk, Wolfgang bei Hanau, Germany). The grits were milled into fine soy flour with a UPZ100 pin-mill with a stationary disk and a rotary disk with each four rings of pins (diameter pins 3 mm, height pins 7 mm) (Hosokawa-Alpine, Augsburg, Germany). The milling speed for soybeans was varied between 8000 rpm and 22000 rpm (838–2304 rad·s$^{-1}$) with an air flow rate of 60 m$^3$/h and a feed rate of 0.5 kg/h. For de-oiled soybeans three milling speeds of 8000, 15000 and 22000 rpm (838, 1571 and 2304 rad·s$^{-1}$ respectively) were used with an air flow rate of 60 m$^3$/h, and feed rate 0.5 kg/h. The milling yield is defined as the collected mass after milling divided by the feed mass prior to milling.

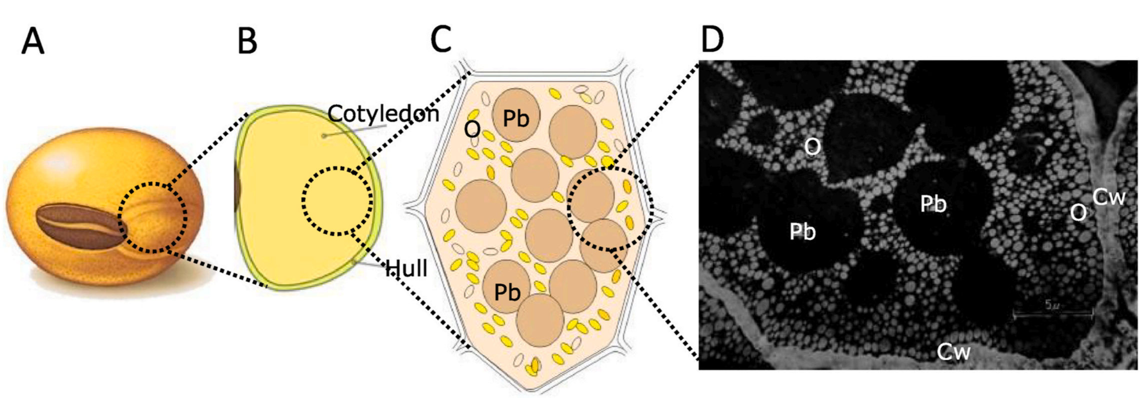


**Fig. 1.** Schematic drawing of soybean seed parts: the whole seed (A), the soy cotyledon and hull, (B), a schematic representation of a cotyledon cell (C) and an electron micrograph of soybean cotyledon cells (D). PB indicates protein bodies, CW cell wall and O oil bodies (spherosomes), adopted from Medic et al. (2014) and Saio and Watanabe (1968).

### 2.3. Sample analysis

#### 2.3.1. Composition

The crude oil content was measured with a Büchi extractor (B-811, Büchi Labortechnik AG Switserland). The samples (2 ± 0.02 g) were exactly weighted and the oil was extracted with an excess amount of solvent (Petroleum Ether 40–65 °C), with a sample to solvent ratio between 1:44 and 1:60. The continuous extraction mode was used, which included 2 h of heating and 30 min of rinsing on level 9 followed by 30 min of drying with heating level 4. The heating levels were used according to the specifications by the manufacturer (Büchi Labortechnik, 2016). After extraction, the samples were dried at room temperature overnight. The moisture content was determined using an Air-Oven Method (105 °C, dried overnight) (AACC Method 44–15.02). The protein content was determined by dumas analysis with a rapid N exceed protein analyser (Elementar, Germany) via high temperature combustion and detection of the released nitrogen. Dry samples were exactly weighted (130–160 mg) in 35 × 35 mm tin foil for elemental analysis with a pre-programmed method by the manufacturer for 250 mg analysis. Calibration of a daily factor was done with aspartic acid and for the samples a conversion factor of 5.71 was used.

#### 2.3.2. Particle size

The particle size distributions of the grits and the fine flours were measured with a Mastersizer-3000 equipped with an Aero-S module for dry powder dispersion with a high energy venturi (Malvern Panalytical LTD, United Kingdom). The hopper gap was set to 3 mm and the samples were dispersed in the dry cell with a pressure of 2 bar and a constant feed rate of 60–80%. The average volume-weighted particle size distribution was calculated for non-spherical particles with a general-purpose analysis model.

#### 2.3.3. Flour dispersibility

The dispersibility of the samples was measured by using a pressure titration method. For this the particle size distribution was determined at dispersion pressures of 50, 100, 200 and 400 kPa. It is assumed that the particles are fully dispersed at a high dispersion pressure and remain agglomerated at lower dispersion pressures (Pelgrom et al., 2014). The extent of de-agglomeration (DA) was calculated by the ratio between the particle size ($DV_{50}$) at full dispersion (4 bar) and the particle size at 0.5 bar (Equation (1), Jaffari et al., 2013; Pelgrom et al., 2014).

$$DA = \frac{DV_{50}\ at\ 4\ bar}{DV_{50}\ at\ 0.5\ bar} \tag{1}$$

The dispersive index (DI) for protein particles smaller than 10 μm was calculated by the ratio of the volume percentage of particles smaller than 10 μm at each dispersion pressure of 0.5, 1, 2 and 4 bar (Dijkink et al., 2007, Equation (2)).

$$DI = \frac{Volume < 10\ \mu m\ at\ pressure\ i}{Volume < 10\ \mu m\ at\ 4\ bar} \tag{2}$$

### 2.4. Mathematical modelling of grinding energy and particle size with empirical models

The rotational energy is described with Equation (3), in which $E_{rotational}$ is the rotational kinetic energy (J), $I$ is the moment of inertia of the rotating object $(\mathrm{kg}\cdot\mathrm{m}^2)$ and $\omega$ is the angular frequency $(\mathrm{rad}\cdot\mathrm{s}^{-1})$.

$$E_{rotational} = \frac{1}{2} \cdot I \cdot \omega 2 \tag{3}$$

For validation, the moment of inertia and the exponent in Equation (3) were estimated based on a non-linear least sum of squares method.

The decrease in particle size by the energy input can be described by Bond's empirical model for particles between 10 μm and 5 mm (Sokolowski, 1996). Bond's empirical model is given in Equation (4).

$$E_B = \frac{m_p t}{W} = C_B\left(\frac{1}{\sqrt{d_p}} - \frac{1}{\sqrt{d_F}}\right) \tag{4}$$

Where $E_B$ is the specific energy (kJ/kg), $m_p$ the power used by the mill (kW) read from the machine, t refers to time (s), W is the sample weight to be milled (kg) and $C_B$ is the milling index or Bond's constant $(\mathrm{kJ}\cdot\mathrm{mm}^{0.5}/\mathrm{kg})$. The $d_p$ and $d_F$ represent the particle size diameter at 80% of the cumulative volume ($DV_{80}$) of the product and the feed in mm, respectively (Sahay and Singh, 2001). The specific energy was calculated for the different milling speeds, with the average $m_p$, which was read from the equipment during the experiment and with a throughput of 0.5 kg/h. The milling index was calculated for the soy flours with different oil contents based on the least residual sum of squares method.

Bond's constants reported in literature had varying units and the calculation was not similar across published research, i.e. the use of varying particle size indicators (i.e. $DV_{80}$, $DV_{3,2}$ or $DV_{50}$). As Bond's constant is specific for the mill used, combined with the varying calculations and units, broader comparison was not possible (Appendix Table A1; Dabbour et al., 2015; Ghorbani et al., 2010; Pujol et al., 2000; Tangirala et al., 2014).

### 2.5. Statistical analysis

Data were collected and analysed in R-studio Version 4.0.2 (The R Foundation). A non-linear least squares method was used to estimate the parameters of a non-linear model. Next to this, the 95% confidence interval of the estimated parameters was calculated. The models were compared using the Akaike criterion (AIC):

$$AIC = n \cdot ln\left(\frac{SS_r}{n}\right) + 2\cdot(p+1) \tag{5a}$$

When the number of experiments was small, $n/p < 40$, Equation (5b) a corrected version of the AIC ($AIC_C$) was used:

$$AIC_C = n \cdot ln\left(\frac{SS_r}{n}\right) + 2\cdot(p+1) + 2\cdot(p+1)\cdot\frac{n}{n-p} \tag{5b}$$

In which n is the number of data points, p the number of parameters and $SS_r$ the residual sum of squares (Boekel, 2008). The difference between the models was expressed with ΔAIC, in which the model with the lowest AIC value was used as a reference (Equation (5c)).

$$\Delta AIC = AIC - AIC_{min} \tag{5c}$$

As rule of thumb, models with $\Delta AIC \leq 2$–3 are worthwhile to consider, values between 4 and 7 are less supported models and values above 10 indicate models that may be discarded (Boekel, 2008). Data were weighted to obtain an even emphasis on de-oiled and non-de-oiled samples.

**Table 1**
Main die diameter, oil content, protein content and average particle size ($DV_{50}$) of once and twice pre-milled soy (A: above dotted line) and de-oiled soy flours (B: below dotted line).

| Sample | Main die diameter [mm] | Oil content [g/100 g dry solids] | Protein content [g/100 g dry solids] | $DV_{50}$ [μm] |
|---|---|---|---|---|
| Soy | None | 17.37 ± 0.08 | 36.64 ± 0.17 | 1213 ± 46 |
| Soy twice pre-milled | None | 20.32 ± 1.00 | 36.08 ± 0.71 | 824 ± 43 |
| Slightly de-oiled soy | 18 | 15.67 ± 0.34 | 40.84 ± 0.45 | 739 ± 37 |
| Moderate de-oiled soy | 16 | 10.48 ± 0.05 | 43.03 ± 0.08 | 688 ± 41 |
| Highly de-oiled soy | 14 | 8.94 ± 0.04 | 43.42 ± 0.18 | 759 ± 32 |

## 3. Results and discussion

### 3.1. Composition

The protein and oil contents of non-de-oiled soy flour (milled once or twice) were measured (Table 1A) and found comparable to previously reported values, i.e. 32.0%–37.4% and 16.7%–20.5% for protein and oil, respectively (Rotundo, Miller-garvin, & Naeve, 2016). The oil content increased with decreasing particle size (Table 1A), which is in line with previous observations and related to an increasing extraction efficiency for smaller particles (Rosenthal et al., 1998). The protein contents of milled once and twice soy flour (20.32 g oil/100 g dry solids) were similar, which is related to the difference in analysis procedure for protein and oil, where protein analysis relies on combustion rather than on extraction. It is noted that the oil content as determined for the twice milled soy flour (20.32 g/100 g dry solids) is further used as the reference for soy flour as it has a particle size more similar compared to that of the milled de-oiled soy flours (650–850 μm). The oil and protein contents of the de-oiled and milled soy flours are given in Table 1B. The main die diameter of the oil press was reduced to increase the pressure drop to remove more oil. The oil content was measured after subsequent milling and is assumed not affected by the small differences in particle size. The protein content on dry basis increased with higher degree of de-oiling due to the removed oil.

### 3.2. Milling yield

As might be expected, the milling yield during pin-milling significantly declined with increasing milling speed (Fig. 2). It was observed that most of the material (>60% of the loss) was lost due to fouling in the milling chamber, whereas the remaining material was lost in piping and corners of other parts of the milling device. Accumulation inside the mill at the stationary disk and the rotating disk are shown in Fig. 3. The pins of the stationary disk were relatively clean for lower milling speeds, whereas at higher milling speeds more residual material was found on the pins. The rotor disk remained relatively clean during all milling conditions tested.

For de-oiled soy flours milled at 22000 rpm the overall milling yield was significantly higher compared to the soy flour (20.32% oil) (Fig. 2). In addition, less material was lost in the milling chamber for de-oiled samples (8.94% oil), where both the stator and the chamber of the pin-mill remained cleaner than with soy (20.32% oil) milled at 15000 and 22000 rpm (Fig. 3). Less material accumulated at the wall, which is likely due to the lower oil content of these samples. Accumulation of material at the wall is expected to be enhanced by the centrifugal force induced upon milling, whereas deposits on the stator are likely formed due to the low shear forces close to the static pins. To reduce the latter, an alternative pin configuration with two rotating disks especially suitable for sticky materials could be evaluated (Furchner, 2009). This will be expected to work well for soy (20.32% oil) as the rotor remained much cleaner than the stator (Fig. 2).

### 3.3. Particle size distribution

The particle size distributions of the milled flours were evaluated and described with Bond's empirical model to relate milling conditions and material properties (especially oil content) to effective size reduction. The particle size distributions were analysed as function of pin-milling conditions and oil content (Fig. 4). For the highest milling speed (22000 rpm), the average particle size for soy (20.32% oil) was reduced to 122 μm ($DV_{50}$) and that of highly de-oiled soy was reduced to 51 μm ($DV_{50}$). The particle sizes were in a similar range as observed in previous studies, which were 90–184 μm for soybean flour and 49 μm for de-oiled soy (Russin et al., 2007; Xing et al., 2018). A lower oil content enabled milling to smaller particle sizes for both pre-milling into grits and fine milling at different rotation speeds.

For both soy (20.32% oil) and highly de-oiled soy the particle size distribution shifted towards smaller particle sizes upon higher pin-milling speeds (Fig. 4). The single peak with particles up to 100 μm are expected to consist of individual particle structures milled to a similar size, for example cell wall fragments and liberated protein bodies. The peak above 110 μm will likely consist of clustered particles and hull debris (Xing et al., 2018). For soy (20.32% oil) the shift in particle size showed a clear bi-modal distribution, whereas for highly de-oiled soy the shift towards smaller particle sizes occurred faster towards one main peak. Based on the observations for soy (20.32% oil), the particle size of highly de-oiled soy may also have shifted via a bi-modal distribution between 8000 and 15000 rpm.

Differences between milling of whole soybeans and highly de-oiled soy may be explained by the reduction in oil content and/or the degree of compaction of the material during pressing. The faster shift towards smaller particles with similar milling speed for de-oiled soy flour suggests that after (partial) removal of the oil a more brittle material is obtained that is easier to be fragmented. For soy flour (20.32% oil) one can observe a bimodal distribution, which is different for the de-oiled soy flour. This observation may be related to the compression during

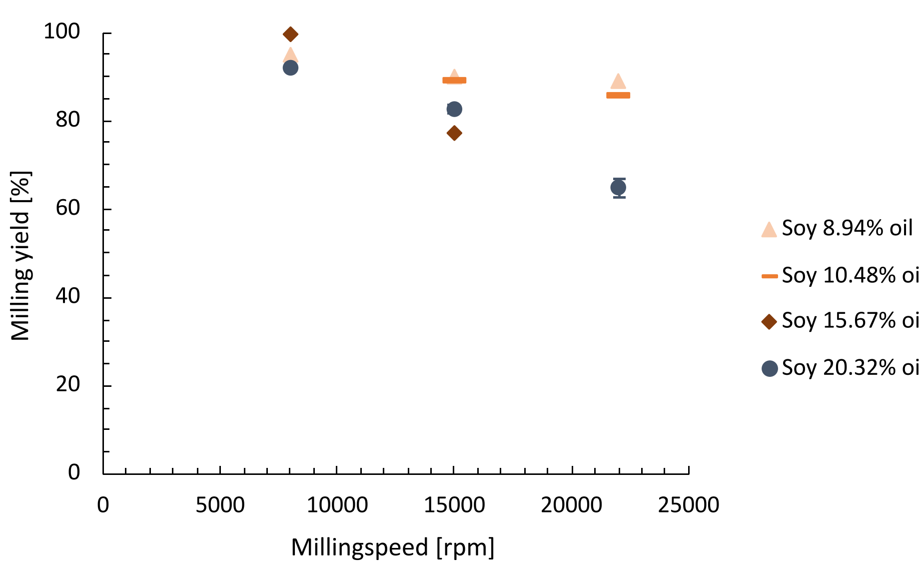


**Fig. 2.** Milling yield versus milling speed of fine milled soy flours, the error bars indicate the standard deviation of the yield. The oil percentages are on dry basis.

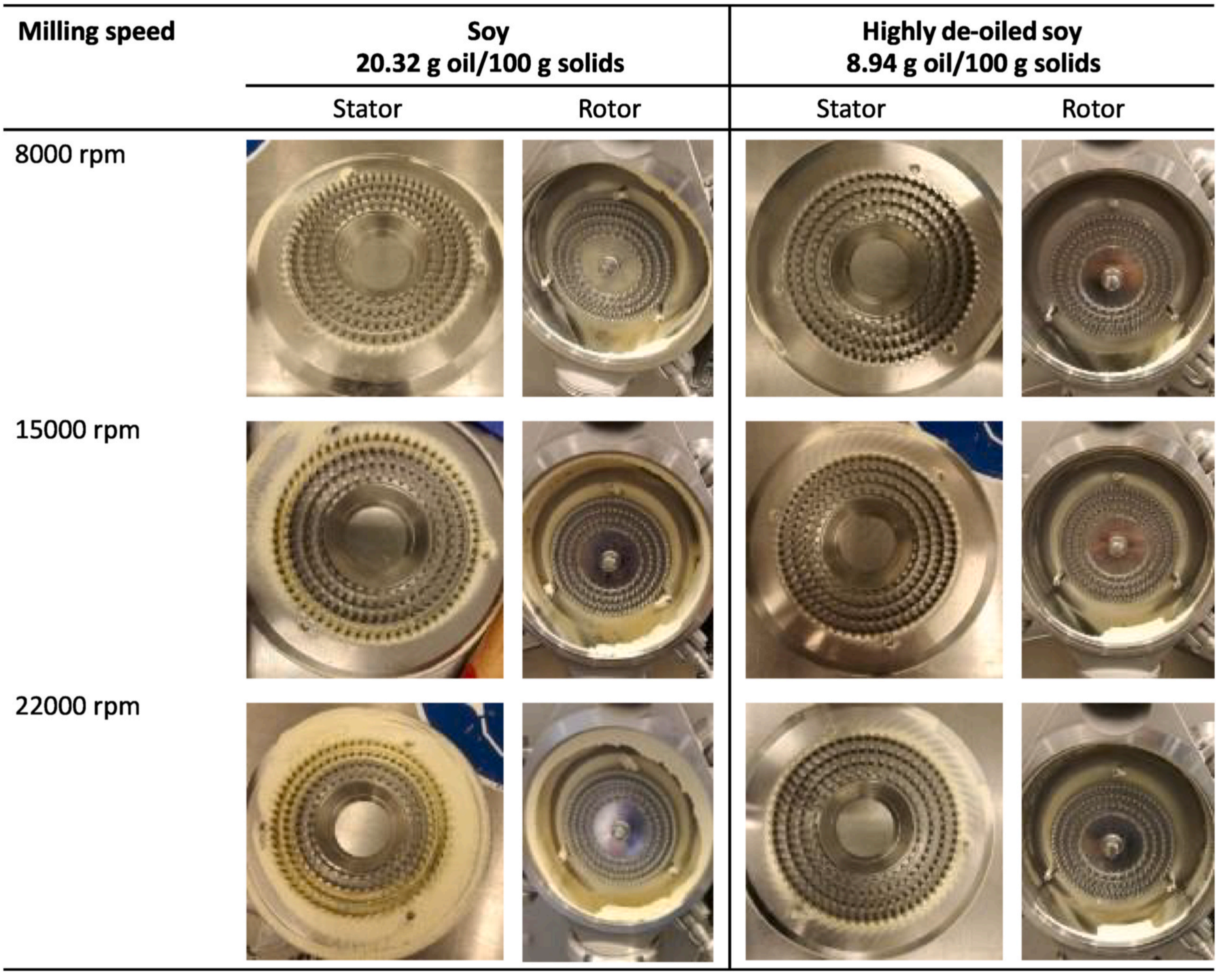


**Fig. 3.** Accumulation on the stator and the rotor inside the pin-mill for soy and highly de-oiled soy milled at different milling speeds.

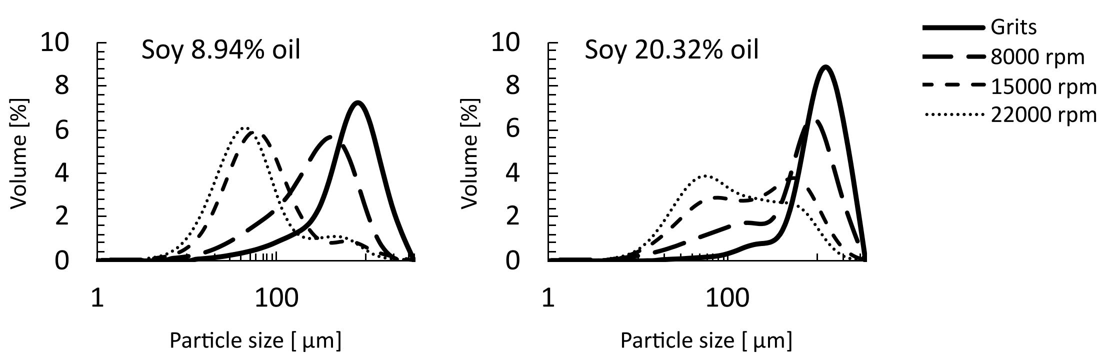


**Fig. 4.** Average particle size distribution of milled highly de-oiled soy (left) and soy (right). The oil percentages are on dry basis. Grits represent the pre-milled material, whereas the dotted and dashed lines represent the fine milled flours at a defined milling speed, specified in the figure legend.

de-oiling in the screw press, which compacts different tissue structures (e.g. protein bodies, cell fragments) together. Upon milling such structures may then be less well disentangled and loosened with the consequence that small components will not be released and thus a different particle size distribution is observed.

### 3.4. Modelling of particle size reduction during milling

The particle sizes ($DV_{80}$) after milling at different speeds (angular frequencies) were determined and correlated to the energy use of the mill (Fig. 5). The milling speed was strongly correlated to the energy consumption ($R^2 = 0.97$) and the rotational energy could be described with Equation (6) ($R^2 = 0.998$), in which the milling energy (kJ/kg) quadratically increased with the angular frequency $\omega$ (rad $\cdot s^{-1}$).

$$E_B = \frac{1}{2} \cdot 2.5 \cdot 10^{-3} \cdot \omega^{2.0} \tag{6}$$

A summary of the parameter estimation is provided in Appendix B. With a 3D model of the UPZ100 pin disk a theoretical moment of inertia of $1.73 \cdot 10^{-3}$ kg$\cdot$m$^2$ was calculated (Hosokawa Alpine, 2021). The

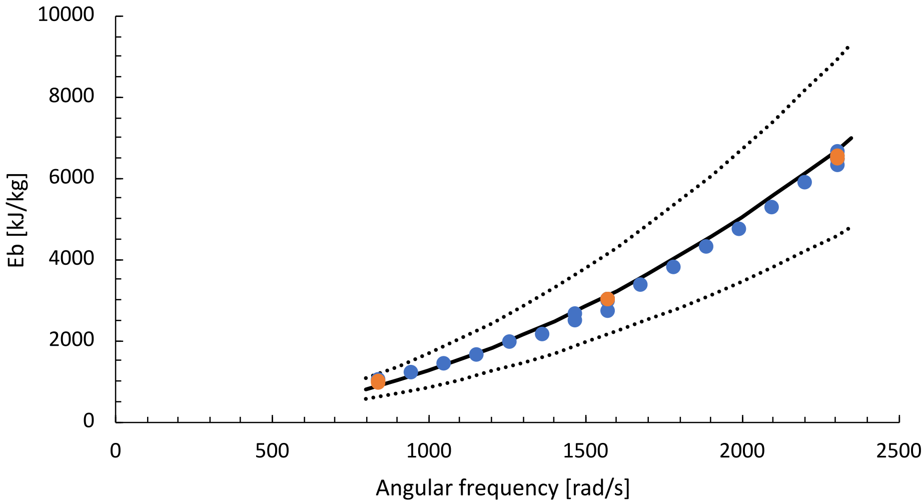


**Fig. 5.** Specific energy ($E_B$) during steady state operation against the angular frequency for soy (●) and de-oiled soy (●) milling. The solid black line indicates the prediction with Equation (6) ($R^2$ = 0.998). The upper and lower limit of the 95% confidence interval of the moment of inertia are given with a dotted line ($1.73 \cdot 10^{-3}$ and $3.66 \cdot 10^{-3}$ kg·m$^2$). A summary table of the parameter estimates is provided in Appendix Table B1. The reader is referred to the online version for a colour representation.

estimated value in this study ($I = 2.5 \cdot 10^{-3}$) was slightly higher than the latter value as the theoretical value does not consider friction forces due to the drive unit or gear related parts.

The particle size ($DV_{80}$) reduced upon an increase in energy consumption (Fig. 6). The particle size reduction for whole soybeans required significantly more energy than for de-oiled soy, which is related to the presence of more oil leading to increased ductile behaviour (Pelgrom et al., 2014). Similar observations with respect to higher energy consumption have been done for milling at higher moisture contents for soy (8–12%) and wheat (12–18%), where differences in moisture content were obtained via drying, soaking and tempering (Lee et al., 2013; Warechowska et al., 2016).

Bond's empirical model (Equation (4)) can be used to describe the relationship between the particle size and energy use of a mill. In previous research, Bond's constant is usually determined for each milled sample separately, e.g. by Lee et al. (2013). We followed the latter approach as well, but also used an alternative approach in which we used the oil content and an oil-independent Bond constant to include a clear dependency on oil content:

$$E_B = \frac{m_p t}{W} = Oil\%_{db} \cdot C_{BO} \left( \frac{1}{\sqrt{d_p}} - \frac{1}{\sqrt{d_F}} \right) \tag{7}$$

The advantage of the alternative approach is that data from multiple samples can be used together to estimate an oil-independent Bond's constant. Secondly, the explicit inclusion of oil content shows the dependency of the Bond's constant, and thus size reduction by milling, on oil content.

The different approaches were compared using the Akaike criterion (Equation (5)). The traditional estimation with sample-based Bond's constants is least preferred ($\Delta AIC$ = 10.2) (Appendix Table C1). The estimation using two Bond's constants, one for de-oiled samples and one for non-de-oiled samples appeared better ($\Delta AIC$ = 0). However, we hypothesized that decreasing oil contents will gradually affect grinding characteristics and therefore defining these two groups is less desirable. The results of the model with oil content and an oil-independent Bond's constant $\left(293 \pm 33 \frac{kJ\ mm^{0.5}}{oil\%(db) \cdot kgmaterial(wb)}\right)$ as parameters are provided in Fig. 6 ($\Delta AIC$ = 0.8).

Prediction of the particle size reduction with Bond's model showed deviations especially for smaller particle sizes (Fig. 6). To further check the validity of the obtained Bond's model for smaller particle sizes and oil content, the soy flours (with 8.94% oil, 15.67% oil and 20.32% oil and only milled at 8000 rpm) were re-milled at 8000 rpm to generate an additional data set. This resulted for soy with 20.32% oil in a $DV_{80}$ of 1058 ± 34 μm after twice milling at 8000 rpm and 887 ± 24 μm after

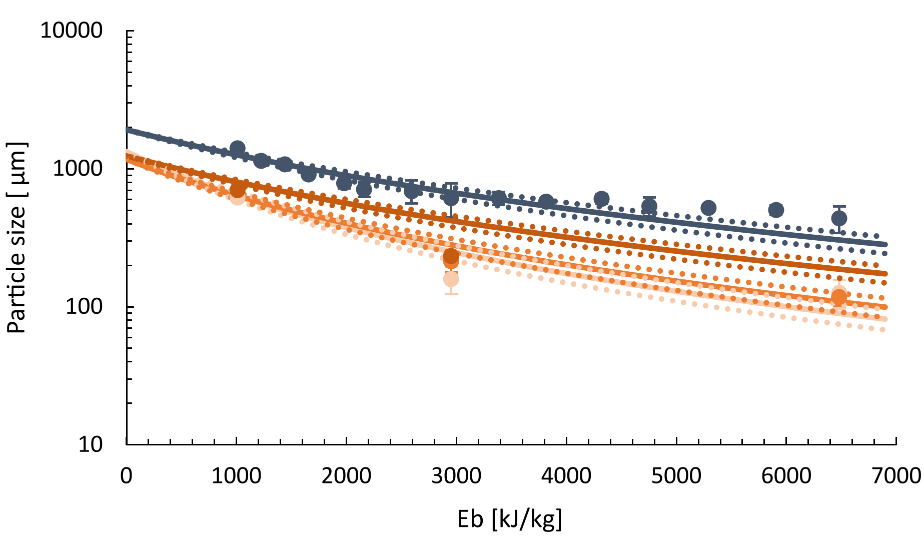


**Fig. 6.** Average particle size ($DV_{80}$) of soy 20.32% oil (●, initial particle size ($D_F$) = 1.92 mm), highly de-oiled soy 8.94% oil (●, $D_F$ = 1.33 mm) moderate de-oiled soy 10.48% oil (●, $D_F$ = 1.74 mm) and slightly de-oiled soy 15.67% oil (●, $D_F$ = 1.24 mm) versus the specific energy ($E_B$), error bars indicate the standard deviation. The predicted values with Equation (7) ($C_{BO}$ 293 ± 33 $\frac{kJ\ mm0.5}{oil\%(db) \cdot kgmaterial\ (wb)}$) are indicated with a solid line and the upper and lower limit of the 95% confidence interval are given with a dotted line. The reader is referred to the online version for a colour representation.

three times milling at 8000 rpm. Similarly, soy flours (8.94% oil and 20.32% oil) milled at 15000 rpm and 22000 rpm were re-milled. The model with an oil-independent Bond's constant was best able to describe the particle size reduction ($\Delta$AIC = 0; Appendix Table C.2), but also upon re-milling the predicted particle size was smaller than the actual particle size (Fig. 7). In addition, the higher energy input by re-milling did not result in smaller particle sizes than a similar energy input did for once milling (data labels Fig. 7; i.e. once milling at 15000 rpm (2952 kJ/kg) resulted in a smaller actual particle size than three times milling at 8000 rpm (3024 kJ/kg)). So, particle size reduction upon milling to smaller and smaller particles becomes increasingly less efficient as the limitations of the mill are reached and the particle size cannot be reduced any further, despite of increased milling speeds. This limitation for soy 20.32% oil was reached for a larger particle size than for soy with 8.94% oil, which is in line with Equation (7). However, the presence of a plateau value is not incorporated in Bond's model, which resulted in a deviation from the predicted particle sizes with Bond's model and the actual particle size. For industrial scale-up more detailed population balance models are recommended (Herbst and Fuerstenau, 1980), that consider machine and material parameters separately (Vogel and Peukert, 2005). Such a more advanced mill modelling approach compensates for the mill used and incorporates a lower probability for breaking of smaller particles (Vogel and Peukert, 2005). For example, single particle breakage tests for zeolite particles were successfully used to develop a population balance model and with that design a pin-milling process (Li et al., 2020). Recently, a multiscale modelling approach for particle breakage in milling has been proposed for quantitative prediction of milling processes (Wang et al., 2021).

### 3.5. Particle- and flour dispersibility

An important criterion for dry separation of the finely milled soy flours after pin-milling is a high degree of dispersibility. Therefore, the dispersibility of the milled soy flours was assessed with a pressure titration method, where at low dispersion pressure (0.5 bar) particles may still be agglomerated and at high dispersion pressure (4 bar) primary particles are expected to be fully dispersed. For comparison between different samples the ratio of measured particle sizes at 4 bar and 0.5 bar is calculated to indicate if the particles are dispersible (~1) or poorly dispersible (~0). We compare here two ratios: 1) The extent of de-agglomeration (DA) based on the ratio of the particle size ($DV_{50}$) as given in Equation (1) (Pelgrom et al., 2014), and 2) The dispersive index (DI) defined as the ratio of the volume percentage of (protein) particles smaller than 10 μm as given in Equation (2) (Dijkink et al., 2007).

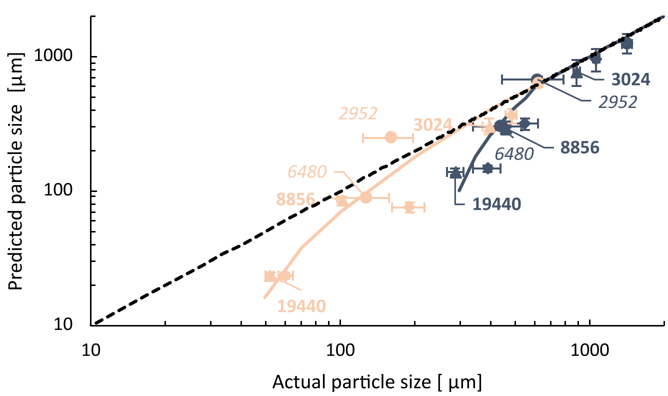


**Fig. 7.** Predicted and measured particle size ($DV_{80}$) of soy 20.32% oil (●) and highly de-oiled soy 8.94% oil (●). Circles (●) represent once milled samples at 8000, 15000 and 22000 rpm (from Fig. 6), diamonds (◇) represent twice milled samples and triangles (Δ) were three times milled. The x error bars represent the particle size standard deviation and the y error bars represent the 95% confidence interval of the prediction with $C_{BO}$ (293 ± 33 $\frac{kJ\ mm0.5}{oil\%(db)\cdot kgmaterial\ (wb)}$). The black dashed line gives y = x other lines are added to guide the eye. The specific energy (kJ/kg) is highlighted for samples milled once (cursive) at 15000 (2952 kJ/kg) and 22000 rpm (6480 kJ/kg) and samples milled three times (**bold**) at 8000 (3024 kJ/kg), 15000 (8856 kJ/kg) and 22000 rpm (19440 kJ/kg). The reader is referred to the online version for a colour representation.

Fig. 8 represents the cumulative particle size distributions for de-oiled soy (8.94% oil) and soy (20.32% oil). The particle size distribution shifted towards higher particle sizes for lower dispersion pressures, which is in line with findings in previous research for lupin and starch mixtures (Dijkink et al., 2007). The $DV_{50}$ at high and low dispersion pressure was used to calculate the DA. Perhaps surprisingly, both soy and de-oiled soy were relatively well dispersible with a DA >0.6. This DA is comparable to the dispersibility of lupine flour (Pelgrom et al., 2014). Within the particle size range tested in this research the DA did not change with particle size (Appendix Figure D1).

For de-oiled soy flour the DI increased with milling speed (Fig. 8). So, a higher milling speed resulted in both finer particles and more dispersible fine particles, which is favourable for further separation. For soy (20.32% oil) the DI increased slightly from 8000 to 15000 rpm but remained similar for a milling speed of 15000 and 22000 rpm. Here, a higher milling speed did not improve the small particle dispersibility. The higher values of the DI for de-oiled soy (8.94% oil) than for soy (20.32% oil) indicate that the dispersibility was influenced by the presence of oil. In addition, a lower oil content resulted in more dispersible small particles upon re-milling or milling at higher speeds, whereas for higher oil content an optimum in small particle dispersion was observed depending on specific energy input (Fig. 9). In comparison, the dispersive index of soy particles <10 μm was lower than the dispersive index of commercial soy protein (DI = 0.83) from literature (Dijkink et al., 2007). This is likely because in literature a protein isolate was used, which contains a larger volume of small particles than whole flour. Lupin flour was found to have a dispersive index of 0.22, which is more comparable to the measured values for de-oiled soy flour milled at 15000 rpm (Dijkink et al., 2007, Fig. 8). The poor dispersibility of small particles hampers subsequent dry separation. This effect is thus expected to be more pronounced at higher oil contents (Fig. 9).

## 4. Concluding remarks

Milling is considered a critical step for subsequent dry separation processes as it should achieve sufficient particle size reduction, dispersible particles and physical disentanglement of the plant cell constituents (Schutyser and van der Goot, 2011). In this study it was shown that the dispersive index is a good predictor for the dispersibility of small particles in flours. Although overall all produced soy flours in this study would well disperse (DA>0.6), the higher oil contents showed an optimum in dispersibility for fine particles <10 μm.

Higher oil content in soy limits milling to smaller particles and lowers the overall milling yield. This limit is also determined by the mill device used. The effect of oil content on particle size reduction can be largely described with an adapted Bond's model that used oil content as an input parameter. This approach could be interesting for describing also the effect of milling for other oil-containing crops. However, for crops with a very high oil content (>35–40%), like sunflower seeds, de-oiling is inevitable to apply prior to dry milling, as upon milling a paste is obtained rather than a powder.

The limitations in particle size reduction may be overcome by specifically re-milling the coarse fraction to liberate proteins from larger particles and further enhance recovery of the protein (Wang, Zhao, De Wit, Boom and Schutyser, 2016b). Strategies to further optimise dry separation could involve dehulling prior to fine milling and improving the overall milling yield. Soy with a higher oil content resulted in a low milling yield (65%) and deposited material on the stationary disk, which can be optimized on industrial scale by using two rotors, instead of one rotor and one stator, without compromising on the particle size achieved, based on previous research ($DV_{97}$ 150 μm) (Nieh and Snyder,

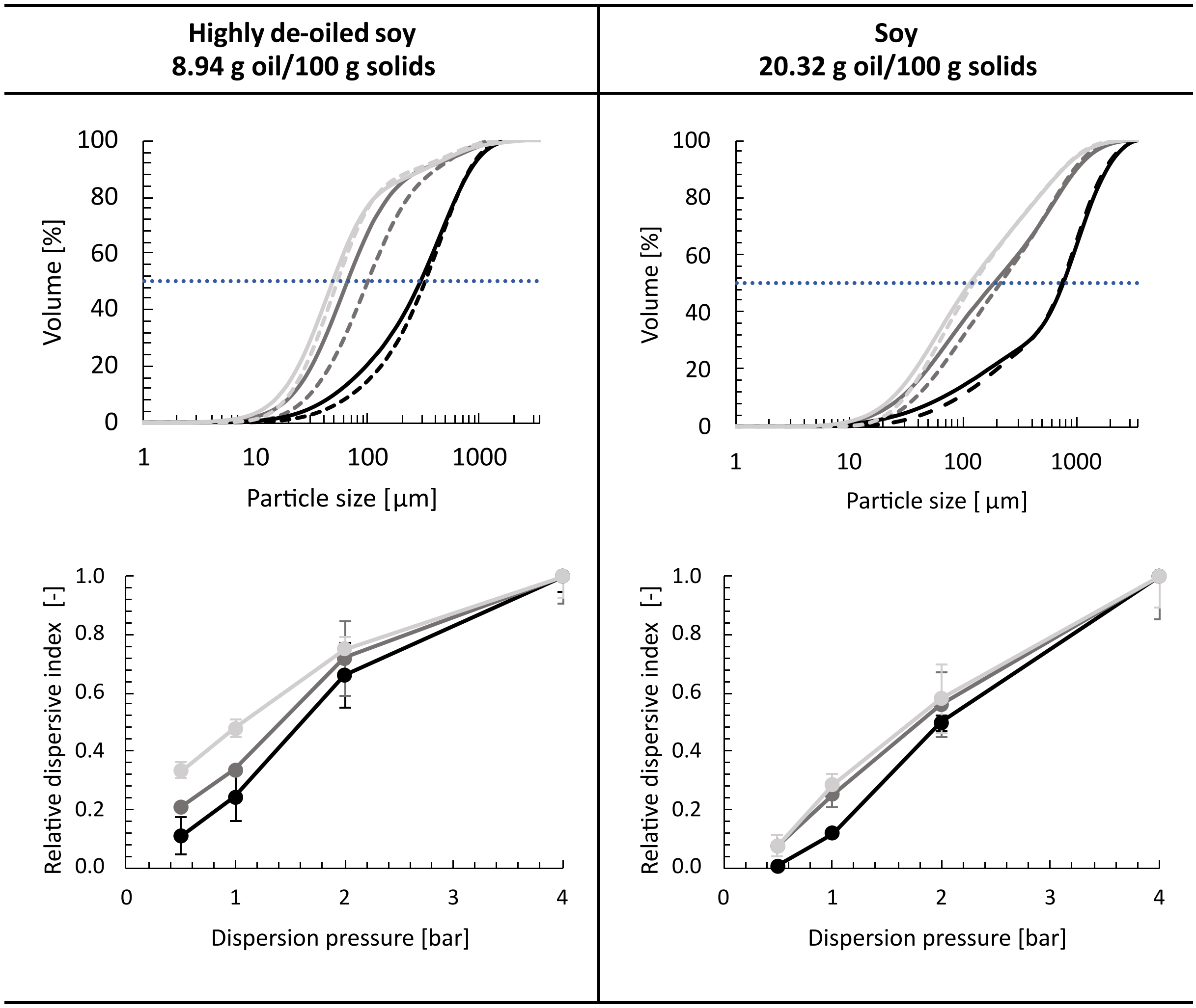


**Fig. 8.** Average cumulative particle size distributions measured at low (0.5 bar, dashed lines) and high (4 bar, solid lines) dispersion pressure in which the $DV_{50}$ at different dispersion pressures (blue dotted line) is used to calculate the extent of de-agglomeration (DA), and the relative dispersive index (DI) for three milling speeds 8000 rpm (black), 15000 rpm (grey) and 22000 rpm (light grey). Error bars indicate the standard deviation.

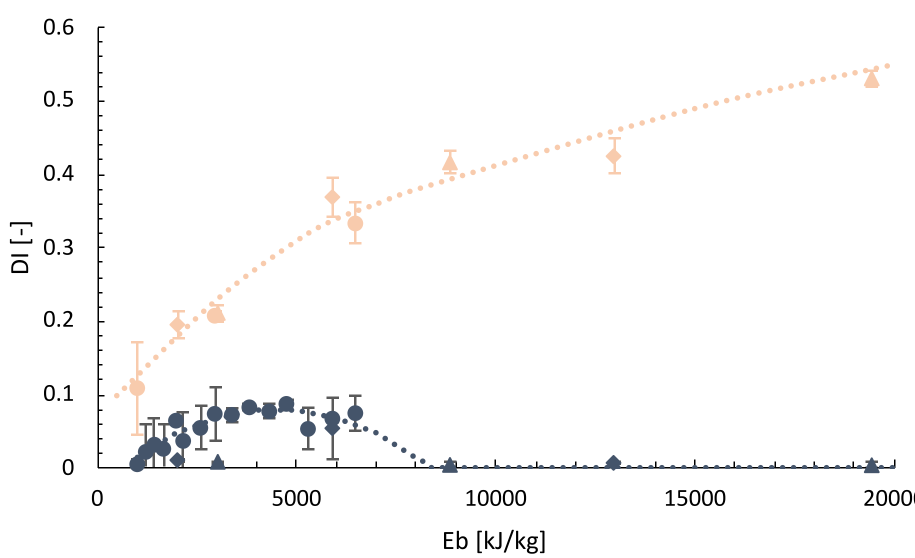


**Fig. 9.** Dispersive index (DI) against the specific energy ($E_B$) for soy 20.32% oil (●) and highly de-oiled soy 8.94% oil (●). Circles (●) represent once milled samples, diamonds (◊) represent twice milled samples and triangles (Δ) three times milled samples. Lines are added to guide the eye. The reader is referred to the online version for a colour representation.

1991).

## Author contribution

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

## Declaration of competing interest

None.

## Acknowledgements

The authors would like to acknowledge Daniela Gruber, Jort Buitenhuis and Kübra Ayan for their exploratory work on this topic.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jfoodeng.2022.111149.

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