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Spray Drying of a Starch-Based Model Emulsion: The Role of Free Oil and Water on Powder Flowability

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ABSTRACT

Spray drying of emulsions is a commonly used technique in the food industry for the encapsulation of oil in food powders, yet oil droplet redistribution to the surface of a drying emulsion droplet can negatively affect powder flowability. While previous studies have investigated either the influence of process and formulation parameters or certain powder structure parameters such as encapsulation efficiency and residual moisture on flowability, this work aims to deepen the understanding of the mechanistical link between the flowability (ff_c-value determined using a ring shear cell) and relevant structural parameters. For this, residual moisture, water activity, encapsulation efficiency and extractable surface oil were chosen and compared in their ability to correlate with powder flowability. Crucially, water activity and extractable surface oil were identified as significantly stronger predictors of flowability compared with traditional metrics such as residual moisture. A reduction in the oil content and a lower drying outlet temperature led to improved flowability, which could be linked to changes in the extractable surface oil content and water activity of spray dried particles. By highlighting these water activity and free surface oil as the primary drivers of powder flowability, this work allows for a more optimized spray drying process design.

1 | Introduction

Spray drying is a widely used process to encapsulate and deliver functional lipophilic components in dry products (McClements et al. 2007). For this, an oil-in-water emulsion is atomized into fine droplets and subsequently dried into individual particles while in contact with hot air (Altay et al. 2024; Masters 2002; Mujumdar 2020). The oily dispersed phase of the emulsion is usually enclosed in a solid hydrocolloid carrier during drying, protecting the encapsulated oil droplets from degradation. In addition, a control of their release via a targeted product design is possible (Mujumdar 2020). After drying, the powder particles are separated from the drying air before being transported, stored, or handled otherwise. During these process operations, but also for consumer satisfaction, appropriate flow properties of the powder particles are necessary. Therefore, the impact of the drying step on powder flowability should already be considered during the initial stages of process design (Schulze 2021). It is known from literature that process and formulation parameters determine particle properties such as particle size and shape, moisture content, and oil distribution within the particle. And all of these particle parameters may in turn influence powder flowability (Kim et al. 2005). This leads to a complex interplay of the different parameters and final product property, making a prediction of powder flowability currently extremely difficult (Barbosa-Cánovas et al. 2005; Juarez–Enriquez et al. 2022; Schulze 2021). It is therefore necessary to build a sound understanding of the impact of the spray drying process on powder flowability.

Based on literature, it is known that the flowability of bulk powder mainly depends on the adhesive forces between particles (Schulze 2021). In the case of spray dried emulsions, it is assumed that liquid bridges between particles are the most important interparticle adhesive forces. High amounts of free, low-viscosity liquids, such as oil or water, are known to impede

powder flowability, as they can lead to increased stickiness and formation of liquid bridges between particles (Barbosa-Cánovas et al. 2005; Kim et al. 2005; Schulze 2021). The amount of free oil and water in a particle largely depends on the emulsion composition and drying kinetics of the emulsion droplets. It is generally agreed that the emulsion's oil content has a strong impact on a particle's free oil content. (McNamee et al. 1998). The impact of the drying kinetics is more complex, as it influences both free water and the encapsulation of oil in the final powder product. In this case, free water refers to water that is not adsorbed to the particle structure. Despite many efforts, these correlations remain subject of investigation. While higher drying temperatures are generally associated with lower residual moisture contents (Aghbashlo et al. 2013), conflicting results are reported about the impact of drying temperature on extractable surface oil. On one hand, faster drying is expected to result in an earlier skin formation, inhibiting a redistribution of oil droplets towards the surface and shrinking of the droplet, leading to less free surface oil (Drusch and Berg 2008; Frascareli et al. 2012). On the other hand, faster drying is also reported to lead to damage in the particle structure, such as the formation of cracks or blow holes, and consequently a higher surface oil content (Vignolles et al. 2007).

These conflicting results highlight the necessity to gain a deeper understanding of the mechanistical correlations between particle structure and powder flowability. Furthermore, the incomplete understanding of the impact of process and formulation parameters on powder properties makes a prediction of powder flowability very challenging. Therefore, this study aims to investigate how the drying step impacts particle properties of spray dried model food emulsions. These insights will then be used to clarify the impact of free oil and water on powder flowability. Traditionally, free oil and water are assessed based on the encapsulation efficiency (EE) and residual moisture content (Kim et al. 2005). However, the extractable surface oil content (Koca et al. 2015) and the water activity (Juarez-Enriquez et al. 2019, 2022) have gained traction as structural parameters which correlate better with powder flowability. This is likely due to the residual moisture being a measure of the total quantity of water adsorbed in the powder matrix, providing no information about the availability of water for reactions or the formation of liquid bridges. The water activity, however, describes the thermodynamic availability of water in a sample, promising a more reliable tool for describing free water in the matrix.

Spray dried powders were prepared with different oil contents at constant dry matter, to clarify the impact of formulation changes. Furthermore, the air outlet temperature was changed to investigate process variations. The air outlet temperature was specifically selected to study the influence of the drying kinetics. Both the power of the air heater and the air velocity have to be increased to achieve a higher air outlet temperature, leading to an increase of heat transfer and thus higher drying rates when maintaining constant feed rates. The spray dried powders were analyzed regarding the described particle property parameters. To correlate these product parameters with powder flowability, a ring shear cell is used for measurement of the flowability. Ring shear cells are used to obtain quantitative results for powder flowability, while commonly used simple methods, such as angle of repose, the Hausner ratio, Carr index, and angle of repose only allow for qualitative measurements (Schulze 2021). These methods have also been used to a limited extent for the investigation of the impact of free water on powder flowability, as they lack sensitivity for changes in moisture content (Juarez–Enriquez et al. 2022; Suhag et al. 2024). To ensure a high applicability of the findings of this study to commonly used products from the food or pharmaceutical industries, a model system containing modified starch and MCT-oil was chosen for spray drying.

2 | Materials and Methods

2.1 | Preparation of Model Emulsions

Starch-based model food emulsions were prepared in a two-step process for all spray drying trials. The employed method is based on the method from Taboada et al. (2021). In the first step, a concentrated emulsion is prepared. The concentrated emulsion contains 10 wt% starch modified for emulsification (starch sodium octenyl succinate, C*Em-Cap, Cargill Deutschland GmbH, Germany), 40 wt% demineralized water and 50 wt% MCT-oil (Witarix MCT-oil 60/40, IOI Oleo GmbH, Germany; density at 20°C: 0.93–0.96 g/cm^3). For this application, a modified starch with good emulsification properties was chosen, acting simultaneously as an emulsifier and as matrix material. Emulsification was conducted in a high-pressure homogenizer (Microfluidizer M-110EH, Microfluidics, MA, USA) operated at 500 bar to achieve oil droplets with a sauter mean diameter of 0.2 μm to avoid further oil droplet breakup during atomization and ensuring emulsion stability (Taboada et al. 2021). The oil droplet size was measured via laser diffraction (PARTICA LA-950V2, Horiba, Japan) in triplicate. In the second step, the concentrated emulsion was mixed with a diluted modified starch solution to the desired concentration. Model emulsion dry matter content was kept constant at 25 wt% of bulk emulsion, while the oil content on a dry matter basis was varied between values of 0 and 60 wt% to resemble a range of emulsions from the food industry.

2.2 | Spray Drying Trials

Spray drying trials were conducted using a co-current pilot-scale spray dryer (Werco SD20, Hans G. Werner Industrietechnik GmbH, Germany). Drying air mass flow rates were set to values between 500 and 700 kg/h and thermal energy input between 30 and 50 kW during spray drying, depending on the desired air inlet and air outlet temperatures. Air inlet temperature was set to 180°C and air outlet temperature to 65°C or 85°C. The liquid feed was supplied to the spray tower using a three-piston pump (Rannie LAB Typ 8.5, SPX FLOW Inc., NC, USA), while a pressure-swirl nozzle (SKHN-MFP SprayDry, Spraying Systems Co., IL, USA) operated at pressures of 50 bar was used for atomization. Particle size distribution was measured in a laser diffraction spectroscope (LS 13320 Beckman Coulter, Beckman Coulter, CA, USA) using a refractive index for modified starch of $n = 1.53$. Powder samples of 5 mL were analyzed in triplicate. Sauter Mean Diameter of the particle size distribution was in the range of 35 to 45 μm for all samples. Detailed results are shown in (Appendix A, Figure A). The liquid feed was kept at room temperature during spray drying. Powder samples were stored in air-tight containers for analysis.

2.3 | Powder Characterization

2.3.1 | Determination of Extractable Surface Oil Content and Encapsulation Efficiency

Extractable surface oil content was determined according to a method presented by Höhne and Gaukel (2024), which is based on a method from other literature (Bae and Lee 2008; Tan et al. 2005). In this method, 15 mL of n-hexane (≥ 95%, Carl Roth GmbH & Co. KG, Germany) were added to 1.5 g of powder in a rolled rim glass and shaken for 2 min with a vortex mixer. The powder was separated by vacuum filtration using Whatman No. 5 filter paper (Cytiva, USA) with a pore size of 2.5 µm and subsequently washed with an additional 15 mL n-hexane. The filtrate was transferred from the vacuum flask to a previously weighed round bottom flask. To separate the solvent from the extracted oil, a rotary evaporator (Laborota 4000 efficient, Heidolph Instruments GmbH & Co. KG, Germany) was used at 150 mPa. The n-hexane was evaporated in a 50°C water bath at a rotation speed of 30 rpm, and the mass of the extracted oil was determined. The EE is calculated according to Equation (1).

$$\mathrm{EE} = \frac{m_\mathrm{T} - m_\mathrm{S}}{m_\mathrm{T}} \tag{1}$$

with m_T being the total oil mass in the powder sample, and m_S the mass of extracted oil. To minimize the impact of powder particles potentially breaking through the filter paper, spray dried samples containing no oil were also examined to obtain a blank value. This blank value was subtracted from the value of m_S. All measurements were conducted in triplicate.

2.3.2 | Determination of Residual Moisture Content

Moisture content of the powder samples was gravimetrically determined. Aluminum test pans were dried for 24 h at 100°C in a drying cabinet (Trockenofen T 6060, Heraeus INSTRUMENTS GmbH, Germany). 1.5 g of powder sample were added to the test pans and dried until an equilibrium was reached or at least 72 h at 100°C, after which the samples were weighed again. To minimize the error introduced due to moisture adsorption/desorption, the samples were stored in a desiccator for 2 h before weighing. The residual moisture content *RM* is determined according to Equation (2).

$$\mathrm{RM} = \frac{m_0 - m_\mathrm{D}}{m_0} \tag{2}$$

with m_0 being mass of the powder sample before drying and m_D the mass after drying. The measurement procedure was carried out in triplicate for each sample.

2.3.3 | Determination of Water Activity

Water activity of spray dried powders was determined using a water activity meter (LabMaster aw neo, Noasina AG, Switzerland). A powder sample of 2 g was added to the measurement chamber and the reading of the water activity (measured humidity of the air in the measurement chamber) was taken when the change of water activity was below 0.001 within 2 min. All measurements were carried out in triplicate.

2.4 | Measurements of Powder Flowability

Powder flowability was determined using a ring shear cell (Volution Powder Flow Tester, Mercury Scientific Inc., USA). To obtain a representative powder sample, a sample divider was used. The sample was then added to the ring shear cell, and a scraper was used to remove any excess powder. Immediately before starting the measurement, the cover of the shear cell was loosely put on. Normal stress at preshear σ_Pre was set to 2, 3, 4, 5, and 6 kPa. An overview of the corresponding normal stresses at shear σ_Sh is given in Table 1.

The first normal stress at preshear was repeated for each measurement, to mitigate inhomogeneity's after first consolidation of the powder sample. The data was analyzed using the software MATLAB (Version: 9.14.0 (R2023a), The MathWorks Inc., USA). A code adapted from the work of Macri et al. (Macri et al. 2020) was used to determine the yield locus based on a linear fit of the raw data. Every measurement was done in triplicate. To facilitate the comparison of the flowability of different powder samples, the ff_c-value according to Equation (3) can be used (Schulze 2021):

$$ff_\mathrm{c} = \frac{\mathrm{CS}}{\mathrm{UYS}} \tag{3}$$

with the consolidation stress CS and the unconfined yield strength UYS. CS and UYS were determined during the measurements. To ensure that particle size in the investigated range had no impact on flowability, spray drying trials were conducted at 50 bar and 100 bar, and an air outlet temperature of $T_\mathrm{out} = 85$°C for both 20 wt% and 60 wt% oil content. Sauter mean diameters at 50 bar were in a range of $\overline{x}_{1,2} = 35$–43 µm and at 100 bar around $\overline{x}_{1,2} = 27$–33 µm. Information about the $x_{10,3}$, $x_{50,3}$ and $x_{90,3}$ values of the particle size distributions are presented in (Appendix B, Table B). Bulk density was measured in the shear cell to be in a range of 0.26–0.58 g/cm^3 for all powders. The results of the powder flowability (Appendix C, Figure C) revealed no significant differences of the UYS at constant CS for the investigated ranges of Sauter mean diameters.

TABLE 1 | Investigated normal stress at preshear and at shear for powder flowability measurements.

Normal stress at preshear σ_Pre/kPa	Normal stress at shear σ_Sh/kPa					
2	0.5	0.7	0.9	1.1	1.3	1.5
3	0.8	1.0	1.4	1.7	2.0	2.3
4	1.0	1.4	1.8	2.2	2.6	3.0
5	1.3	1.8	2.3	2.8	3.3	3.8
6	1.5	2.1	2.7	3.3	3.9	4.5

3 | Results and Discussion

3.1 | Influence of Oil Content and Air Outlet Temperature on Particle Properties (Free Oil and Water)

3.1.1 | Free Water: Residual Moisture and Water Activity

The residual moisture and water activity were measured to investigate the impact of the outlet temperature on free water in dependence of the oil content. The results for all spray dried powders are shown in Figure 1, with the residual moisture (left) and the water activity (right) plotted against the oil content. The residual moisture (Figure 1 left) exhibits lower values for samples dried at $T_{out}=85°C$ compared with $T_{out}=65°C$, with values of residual moisture decreasing from around 8.0% to around 3.6% for $T_{out}=65°C$ and values of around 4.5% to 1.3% at the highest oil content for $T_{out}=85°C$. The water activity a_w (Figure 1 right) shows higher values when drying at a lower temperature, similarly to the impact of the air outlet temperature on residual moisture. In contrast, the water activity shows no correlation with the oil content, as values of water activity were around $a_w=0.45$ for $T_{out}=65°C$ and $a_w=0.2$ for $T_{out}=85°C$.

The observation of a lower residual moisture content and water activity when drying at higher outlet temperatures is attributed to the faster drying kinetics (Kelly et al. 2014; Shamaei et al. 2016). To achieve the higher outlet temperature, air mass flow as well as thermal energy input are increased, resulting in faster heat transfer and a shorter residence time of the particles in the spray dryer. Even though these two effects counteract each other in their impact on free water, the results reveal a lower residual moisture for a higher outlet temperature, resulting in a dryer final powder. This is also reflected when taking the results of the water activity into account (Figure 1 right), as the water activity is lower when drying at a higher temperature.

The decrease of the residual moisture with increasing oil content (Figure 1 left) at constant dry matter content has been described in literature before (Felfoul et al. 2022; Shamaei et al. 2016). This is explained by the decrease in matrix material content, as water molecules are retained in the matrix material and not in the oil droplets. Thus, less water is overall retained in the spray dried powders with higher oil contents. In contrast to this, the results for the water activity (Figure 1 right) show very similar values for all oil contents. The final water activity is crucially determined by the relative air humidity of the outlet air stream of the spray dryer, which is expected to be independent of feed emulsion oil content. This can be attributed to the constant air outlet temperature and total water content in the emulsions.

Taking a look at the expected impact of changes in air outlet temperature on powder flowability based on free water, both residual moisture and water activity indicate poorer flowability when drying at a lower temperature. The impact of the free water based on changes in oil content is not as clear. The residual moisture content is pointing towards an improvement of flowability with higher oil content, while the constant values of water activity for all oil contents indicate no change in flowability. These contradicting expectations highlight a potential pitfall when assessing the impact of free water on flowability, possibly leading to inaccurate assumptions. Generally, the water activity promises a more reliable tool to describe the impact of free water on powder flowability, as it directly correlates with the thermodynamic activity of the water in the sample.

3.1.2 | Free Oil: Extractable Surface Oil Content and Encapsulation Efficiency

To estimate the amount of free oil in the powder particles, the extractable surface oil and the EE were determined for all samples. It is generally expected that drying at lower air outlet temperatures leads to larger amounts of extractable oil, as there is more time available for a redistribution of the oil. This should also in turn correlate with a lower EE. For a higher oil content, a larger amount of oil is expected to be extracted by solvent extraction of powder samples, as there is less matrix material available for the encapsulation of the oil.

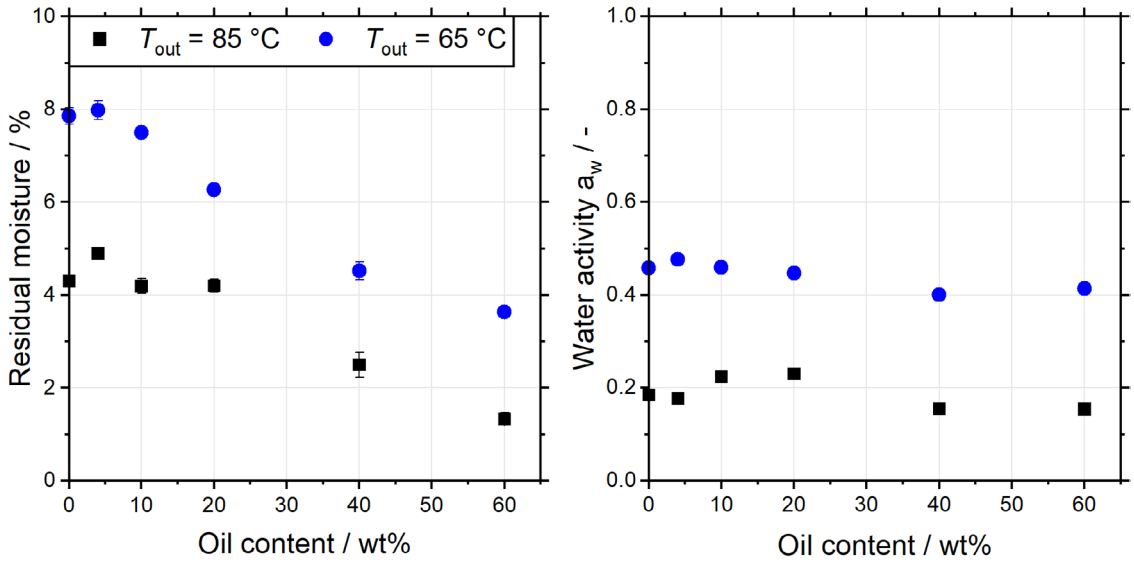

FIGURE 1 | Residual moisture (left) and water activity (right) for all spray dried powder samples in dependency of the oil content.

Figure 2 displays the extractable surface oil (left) and the EE (right) for all powder samples. For the extractable surface oil (Figure 2 left), it can be observed that larger amounts of oil were extracted when drying at a higher outlet temperature. While the accelerated heat and mass transfer theoretically equates to an earlier skin formation and improved encapsulation of the oil, more cracks and capillaries may form during the course of drying, allowing for additional oil to reach the surface of the particles. This can also be seen in exemplary scanning electron microscopy images of the samples (Appendix D, Figure D). This work uses SEM images solely to assess broad observations, as SEM is intrinsically limited in its capability of enabling statistical conclusions. Furthermore, SEM images revealed similar morphologies across samples of different compositions.

Comparing the impact of different oil contents on the amount of extracted surface oil, an increase in extractable oil is determined for increasing oil content at both outlet temperatures. This result is in line with the previously described expectation. It has to be noted that the extractable surface oil exhibits a seemingly exponential trend, as the extractable surface oil increases by a factor of around 4 for an increase in oil content from 4 to 20 wt%, 20 to 40 wt% and 40 to 60 wt% respectively. This observation is assumed to result from the simultaneous increase of oil content and decrease of matrix material content, leading to a poorer encapsulation of the oil.

Looking at the results for the EE (Figure 2 right), the impact of the outlet air temperature is the same as for the extractable surface oil, with less free oil and thus a higher EE for $T_{out} = 65°C$ compared with $T_{out} = 85°C$. Similarly, the results for the EE reveal a strong decrease for higher oil contents, with values of EE decreasing from 20 to 40 wt%, and even more severe from 40 to 60 wt%. Despite these parallels for high oil contents, the observed trend of the EE differs from the trend of the amount of extractable surface oil for small oil contents of 4–20 wt%. While the extractable surface oil is strictly increasing, the EE shows no strict decrease, as the values of EE increase from around 85% to 90% for an increase of the oil content from 4 to 20 wt%. The increase in EE is attributed to a smaller increase of the amount of extracted oil compared with the total oil content in the powder.

This discrepancy in the expected trend obtained from the results of the extractable surface oil and the EE arises for powders with different ratios of oil to matrix material highlighting the importance of differentiating both parameters and applying them to their respective best use case. The EE is most effective in describing the quality of the encapsulation of the core material, which is important information, for example, when designing products with a core material prone to oxidation. However, the EE is not well suited for describing the exact composition of a particle's surface, or in the case of powder flowability, the amount of free oil. For this application, investigating the impact of process and formulation parameters based on the amount of extractable surface oil is the preferred choice, as this parameter better correlates with the formation of liquid bridges between particles and thus the strength of adhesive interparticle forces. It is generally expected that powders with higher amounts of extractable surface oil are characterized by stronger adhesive forces and thus poorer powder flowability. Based on the results for the extractable surface oil presented in Figure 2 (left), it can be predicted that powder flowability is impaired for powders with increasing oil content and dried at higher air outlet temperatures.

3.2 | Impact of Free Oil and Water on Powder Flowability

3.2.1 | Oil Content and Extractable Surface Oil

Ring shear cell measurements were conducted to investigate the impact of a change in oil content at constant dry matter content on powder flowability. Figure 3 shows the ff_c-values over the normal stress at preshear σ_{Pre} of the powders spray dried at $T_{out} = 85°C$ with different oil contents.

At the lowest value of $\sigma_{Pre} = 2$ kPa, ff_c-values for all investigated oil contents are in a range of 1.5–2, corresponding to very cohesive powders. It can generally be observed, that ff_c-values increase with increasing σ_{Pre}, reaching values of 3.5–3.8 at $\sigma_{Pre} = 6$ kPa. This trend has also been previously observed in literature (Fitzpatrick et al. 2004; Juarez-Enriquez et al. 2017).

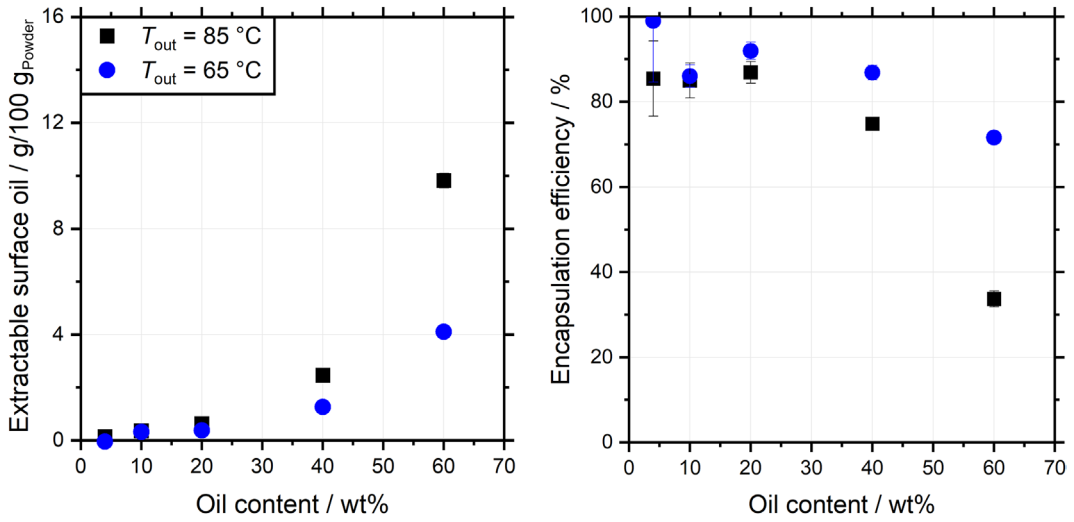

FIGURE 2 | Extractable surface oil (left) and encapsulation efficiency (right) for all spray dried powder samples in dependency of the oil content.

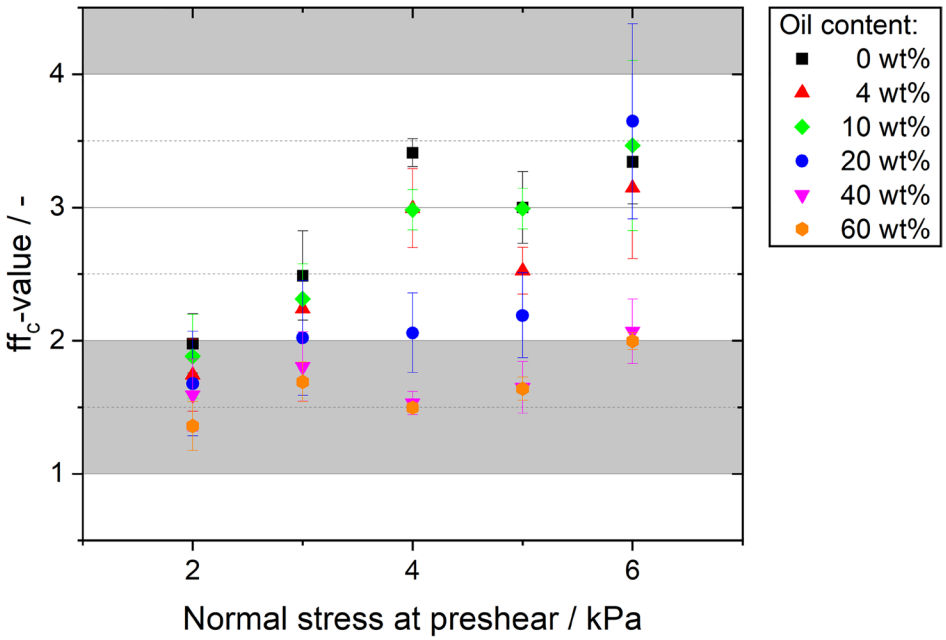

FIGURE 3 | Values of the flowability ff_c plotted against the normal stress at preshear for spray dried powders with different oil contents at T_{out} = 85°C. Areas of different flowability are represented by gray and white shading in the diagram. Powders between ff_c-values of 1–2 are very cohesive, between 2 and 4 cohesive and powders above 4 are easy-flowing.

The highest ff_c-values were obtained for powders with the lowest oil contents of 4 and 10 wt%, exhibiting similar ff_c-values to a starch powder with 0 wt% oil. For higher oil contents (40 and 60 wt%), ff_c-values remain generally around 1.7. The spray dried powder with an oil content of 20 wt% shows ff_c-values in between the samples with 0–10 wt% and 40–60 wt% oil content. An exception at σ_{Pre} = 6 kPa with a high value but a large standard deviation was measured. At the highest σ_{Pre} of 6 kPa and oil contents above 10 wt% it is most likely that stick–slip occurs in the shear cell, leading to inaccurate measurements. This is a known issue for very cohesive powders at high consolidation stresses (Schulze 2021).

The ff_c-values for different oil contents were also investigated for T_{out} = 65°C. They exhibit similar trends as the results at T_{out} = 85°C. The results are shown in (Appendix E, Figure E). The observed trends of the ff_c-values at different oil contents are generally in line with the expectation, that powder flowability is impeded for powder particles with higher oil contents. The results indicate that powders up to an oil content of 10 wt% have substantially improved flowability compared with powders with a higher oil content. It is expected that this is a result of the increasing amount of extractable surface oil correlated with the oil content.

The ff_c-values plotted against the extractable surface oil are shown in Figure 4. Displayed are the results for oil contents up to 40 wt% and both air outlet temperatures of T_{out} = 85°C and T_{out} = 65°C. The ff_c-values were determined at σ_{Pre} = 4 kPa. The results for an oil content of 60 wt% were omitted for the sake of visual clarity.

Figure 4 shows how the ff_c-values decrease with increasing extractable surface oil. The ff_c-values drop to a minimum of around 1.5. Even small amounts of surface oil are shown to lead to a significant impairment of powder flowability, likely due to increasing adhesive forces. The results indicate that adhesive forces increase with increasing extractable surface oil, until minimum is reached at 20 wt% for T_{out} = 65°C and 40 wt% for T_{out} = 85°C. It is suspected that adhesive forces increase due to cohesion of the particles and the formation of liquid bridges. With increasing saturation, the adhesive forces increase up to a maximum value (Fitzpatrick et al. 2007). After this point, saturated liquid bridges have formed and a further increase in extractable surface oil has no apparent impact on the ff_c-values. It is assumed that this point is reached with the low plateau in Figure 4, as the ff_c-values remain constant at around 1.5 with a further increase of the extractable surface oil. These results also show limitations when using the extractable surface oil to predict powder flowability, as precise knowledge of the critical value is necessary. Otherwise, an increase of the ff_c above the critical value may be falsely interpreted regarding its impact on flowability. In future work, an investigation of the surface composition of particles could provide further insight into the effect of free oil on powder flowability. A recently emerged approach to this is the investigation of a particle's surface composition in single droplet drying via Raman spectroscopy (Eijkelboom et al. 2024). This method provides the unique opportunity of investigating the impact of process and formulation parameters on the surface composition of particles. Using these results to correlate with Raman-measurements of spray dried powders could possibly be used to gain insights into the correlation between extractable surface oil and flowability.

Looking at the impact of the air outlet temperature on powder flowability, it can be seen that the ff_c-values are lower for T_{out} = 65°C compared with T_{out} = 85°C for all investigated oil

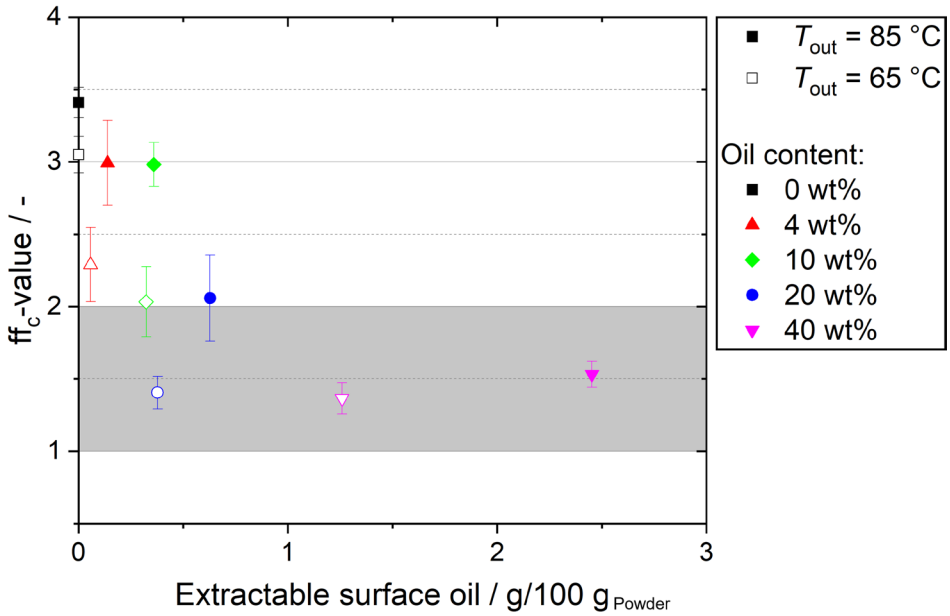

FIGURE 4 | Values of the flowability ff_c for a normal stress at preshear of 4 kPa plotted against the extractable surface oil for all spray dried powders. Areas of different flowability are represented by gray and white shading in the diagram. Powders between ff_c-values of 1–2 are very cohesive, between 2 and 4 cohesive and powders above 4 are easy-flowing.

contents, despite the lower amounts of extractable surface oil at T_{out} = 65°C. Consequently, the values of extractable surface oil are in contrast to the observed trend for the impact of air outlet temperature on the ff_c-values. This fact suggests that other parameters, such as free water, may play a crucial role for the flowability of these powders. Therefore, the impact of the water activity as a measure of free water on powder flowability is investigated in the following chapter.

The results presented in Figure 4 are also suitable to demonstrate the benefit of correlating the extractable surface oil with powder flowability, compared with using the EE for correlation. This benefit becomes apparent when investigating the ff_c-values for powder samples with oil contents of 10 and 20 wt% dried at T_{out} = 85°C and correlating them with the extractable surface oil and the EE. The extractable surface oil increases from 10 to 20 wt% oil content, hinting towards larger adhesive forces and thus lower ff_c-values, which is consistent with the results displayed in Figure 4. Meanwhile, the EE increases with increasing oil content, which falsely indicates improvements in powder flowability (see Figure 2 right).

3.2.2 | Water Activity

Figure 5 displays the ff_c-values (determined at σ_{Pre} = 4 kPa) over the water activity for oil contents up to 40 wt% and both investigated air outlet temperatures. It can be observed, that the results are clustered into two groups depending on their water activity. The powders spray dried at T_{out} = 85°C have water activities of around 0.2, while powders dried at T_{out} = 65°C show values of around 0.45. Generally, for water activities in the range of 0.2 to 0.55, free water stays allocated in multilayers and is less bound to the particle structure. This water is available for interactions and chemical or biochemical reactions, and may influence particle surface structure (Juarez–Enriquez et al. 2022). Consequently, the lower values of water activity at T_{out} = 85°C result in higher ff_c-values compared with T_{out} = 65°C when comparing the results at constant oil contents. Taking the previous results for the extractable surface oil into account, it becomes clear, that the water activity overrules the impact of the extractable surface oil for changes in air outlet temperature. This demonstrates, that not only the free oil content has an influence on powder flowability, but that the impact of the free water content has to be considered at the same time.

4 | Conclusions

This study evaluated the role of free oil and water on the powder flowability of spray dried powders. Spray drying trials with emulsions of varying oil contents at constant dry matter content and different air outlet temperatures were conducted and particle property parameters determined. The extractable surface oil content, the encapsulation efficiency, the water activity and residual moisture were analyzed and the correlation with powder flowability assessed.

Extractable surface oil was found to be an excellent predictor of powder flowability. Generally, flowability decreased with increasing oil content until a critical value of free surface oil is reached. Any further increase in free surface oil does not decrease the flowability any further. This is likely due to the formation of oily liquid bridges, and an increasing saturation of these bridges with increasing free surface oil. The encapsulation efficiency, however, could not be correlated with powder flowability for different oil contents. Finally, an increase in air outlet temperature led to a decrease in water activity, correlating well with the results of an improved powder flowability at higher temperatures.

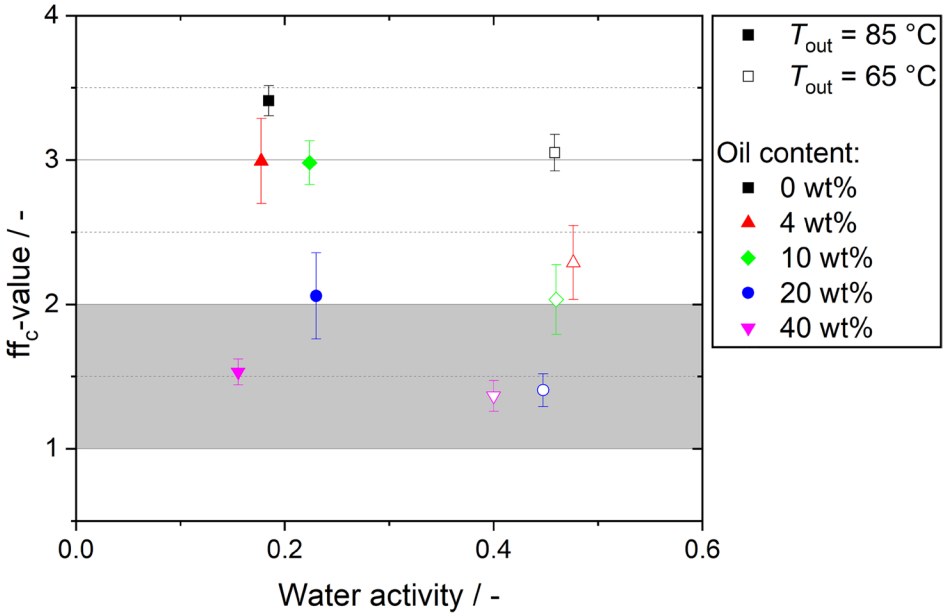

FIGURE 5 | Values of the flowability ff_c for a normal stress at preshear of 4 kPa plotted against the water activity for all spray dried powders. Areas of different flowability are represented by gray and white shading in the diagram. Powders between ff_c-values of 1–2 are very cohesive, between 2 and 4 cohesive and powders above 4 are easy-flowing.

This study provides valuable insights on the impact of free oil and water on the powder flowability of spray dried food emulsions. The water activity and extractable surface oil were found to be the most suitable parameters for evaluating the impact of free oil and water on flowability. Especially, the water activity could serve as quality control parameters in industrial processes, as it can be quickly measured offline using a minor powder sample. A further analysis of the surface composition of spray dried emulsion particles could improve the understanding of the role that free surface oil plays in powder flowability. Single droplet drying setups could be employed for this purpose, providing the unique opportunity of connecting the drying kinetics and morphology development with the surface oil content.

Author Contributions

S.H.: Conceptualization, Investigation, Methodology, Visualization, Writing – original draft. J.M.: Formal analysis, Investigation, Methodology, Visualization, Writing – review and editing. V.G.: Conceptualization, Funding acquisition, Project administration, Supervision, Writing – review and editing.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Appendix A

Sauter Mean Diameter of the Particles for All Powder Samples

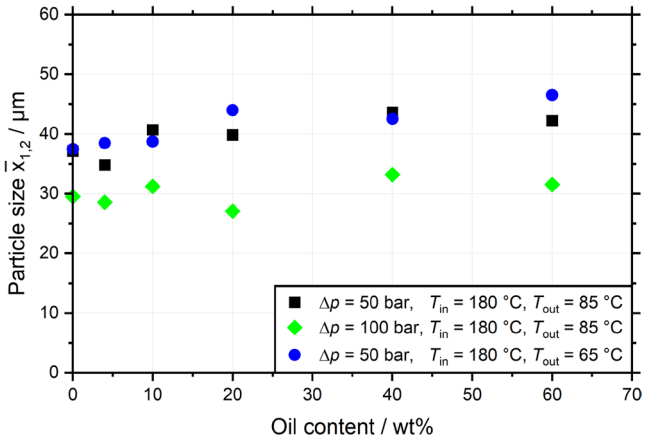

FIGURE A | Sauter mean diameter of the particles for different oil contents, outlet air temperatures and atomization pressures.

Appendix B

Overview of the Characteristic Particle Size Values for All Spray Dried Powder Samples

TABLE B | Overview of the $x_{10,3}$-, $x_{50,3}$-, and $x_{90,3}$-values of the particle size distributions for all spray dried powder samples.

C_{Oil}/wt%	Δp/bar	T_{in}/°C	T_{out}/°C	$x_{10,3}$/µm	$x_{50,3}$/µm	$x_{90,3}$/µm
0	50	180	85	19.46 ± 0.15	51.40 ± 0.39	99.45 ± 1.58
0	100	180	85	15.16 ± 0.13	41.65 ± 0.01	78.24 ± 0.48
0	50	180	65	18.84 ± 0.50	51.69 ± 1.27	104.03 ± 3.41
1	50	180	85	17.79 ± 0.16	48.75 ± 0.21	96.09 ± 1.13
1	100	180	85	14.73 ± 0.05	39.88 ± 0.20	75.38 ± 0.60
1	50	180	65	18.82 ± 0.22	54.70 ± 0.64	118.61 ± 2.39
2.5	50	180	85	20.97 ± 0.12	54.67 ± 0.12	105.02 ± 1.08
2.5	100	180	85	13.91 ± 0.05	38.00 ± 0.17	70.89 ± 0.81
2.5	50	180	65	19.26 ± 0.48	54.22 ± 1.03	116.05 ± 1.59
5	50	180	85	22.01 ± 0.09	54.66 ± 0.08	98.75 ± 0.27
5	100	180	85	16.21 ± 0.14	42.86 ± 0.18	76.18 ± 0.73
5	50	180	65	22.89 ± 0.36	59.80 ± 0.52	138.44 ± 1.62
10	50	180	85	24.78 ± 0.33	57.07 ± 0.56	100.16 ± 1.09
10	100	180	85	17.84 ± 0.25	44.34 ± 0.20	75.82 ± 0.05
10	50	180	65	23.24 ± 0.31	57.79 ± 0.58	110.16 ± 1.73
15	50	180	85	23.93 ± 0.31	56.69 ± 0.51	96.89 ± 0.76
15	100	180	85	16.69 ± 0.41	42.85 ± 0.48	74.03 ± 0.74
15	50	180	65	26.39 ± 0.11	62.50 ± 0.06	116.21 ± 0.22

Appendix C

Results for the Flowability Depending on Different Atomization Pressures

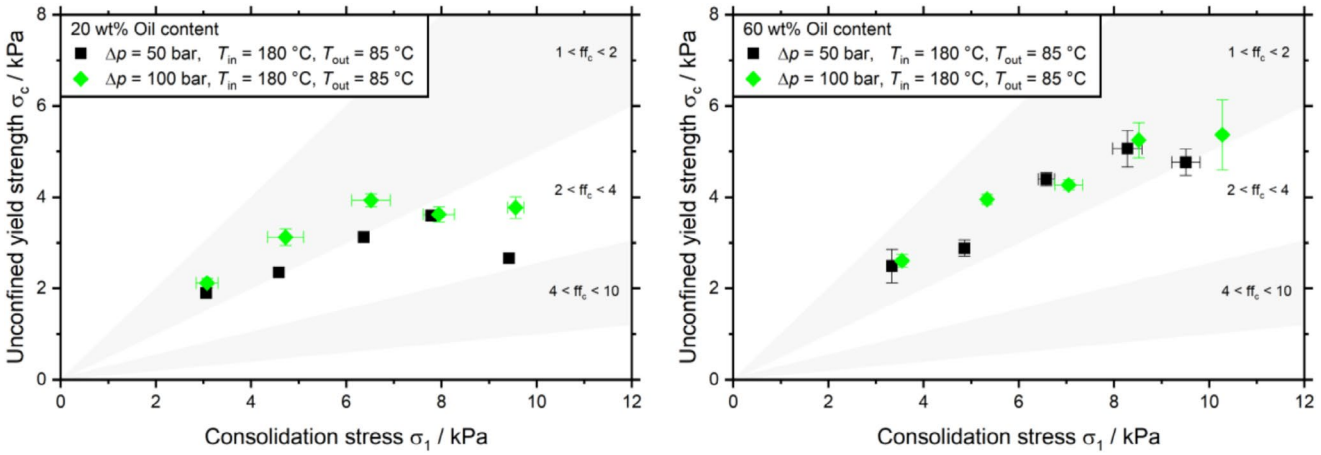

FIGURE C | Unconfined yield strength depending on the consolidation stress for spray dried powders with oil contents of 20 wt% (left) and 60 wt% (right).

Appendix D

Impact of Air Outlet Temperature on the Oil Coverage of Particle Surfaces

Figure D shows the scanning electron microscopy images of powder samples containing 40 wt% oil for $T_{out} = 65°C$ (left) and $T_{out} = 85°C$ (right). The images were taken using a scanning electron microscope (FEI Quanta 650 ESEM). For sample preparation, a small amount of powder was distributed onto a double-sided conductive adhesive tape. Loose powder particles were then removed using pressurized air and the samples were sputtered with 7 nm Pt at a 40° angle (Leica EM ACE 600). All droplets display a wrinkled particle surface. Free oil can be observed as dark shadows on a particle's surface, as the Pt is less effective in coating oil compared with the starch matrix. It can be seen that a larger area of the particles' surfaces is covered in oil when drying at a higher outlet air temperature.

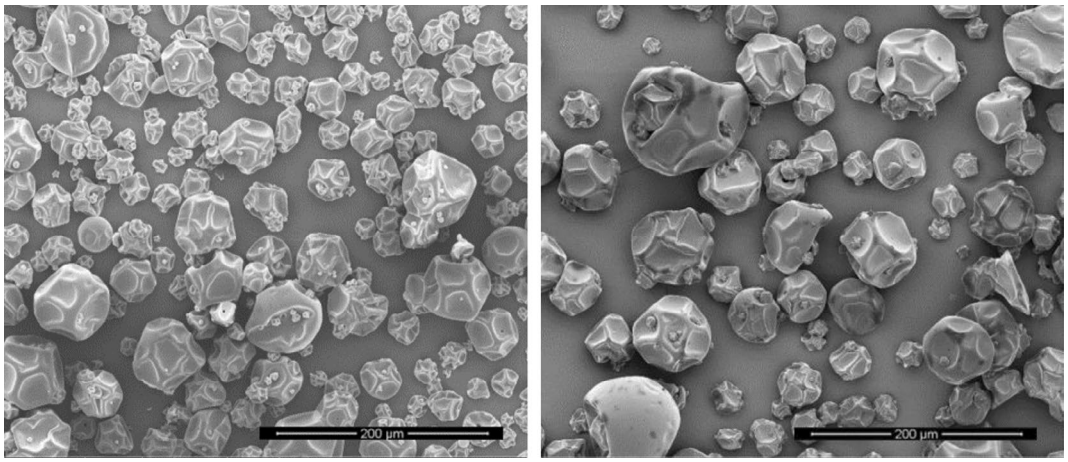

FIGURE D | Scanning electron microscopy images of powder samples containing 40 wt% oil and spray dried at $T_{in} = 180°C$ and $\Delta p = 50$ bar. The outlet air temperature was set to $T_{out} = 65°C$ (left) and $T_{out} = 85°C$ (right).

Appendix E

Values of the Flowability Plotted Against the Normal Stress at Preshear

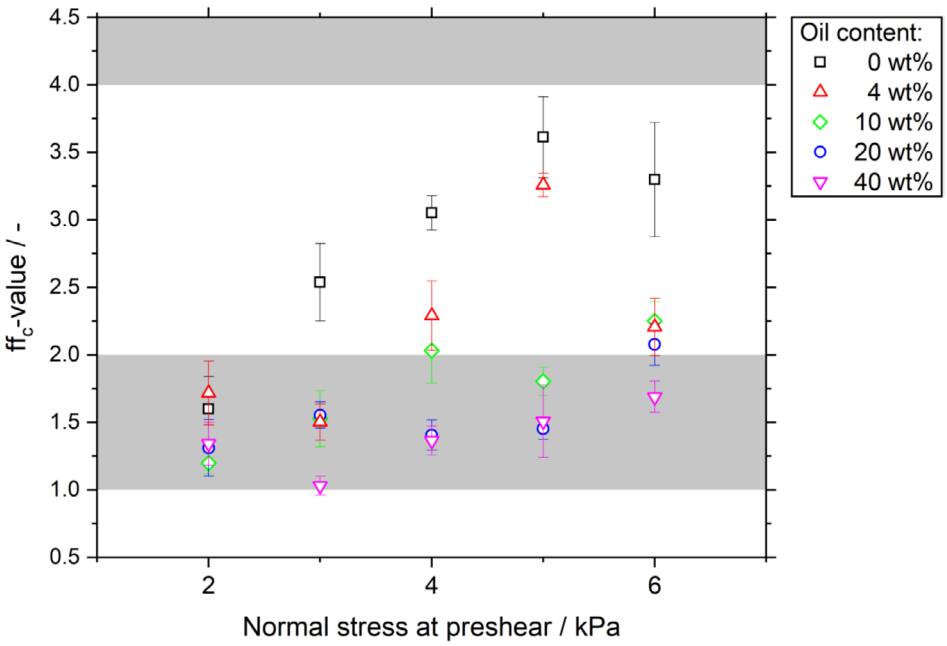

FIGURE E | Values of the flowability ff_c plotted against the normal stress at preshear for spray dried powders with different oil contents at $T_{out} = 85°C$.