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"All oat!" Optimizing the spray drying process for oat oil emulsions stabilized by cellulose nanocrystals and methylcellulose

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ABSTRACT

Oil-filled powders are increasingly important as nutraceutical use expands across food, cosmetic, and pharmaceutical sectors. Oat products are attractive for such applications due to their high antioxidant content, however oat oil (a grain refining by-product) remains challenging to stabilize because it readily oxidizes and thus has not previously been converted into a dry, reconstitutable powder. Here, we report a fully bio-based encapsulation platform in which oat oil is stabilized by cellulose nanocrystals (CNCs), food-grade methylcellulose, and tannic acid. Emulsion stability was evaluated for cold-pressed and solvent-extracted oat oils, with cold-pressed exhibiting superior stability. Spray drying was systemically optimized using a design-of-experiments (DOE) approach to quantify the effects of CNC content and processing parameters on powder yield and quality. The optimized powders achieved yields up to 82%, oil contents of 89%, moisture contents below 2%, and rapid reconstitution into stable emulsions, with CNC content identified as the dominant factor governing performance. CNCs were also extracted directly from oat hulls and applied to produce an "All Oat!" encapsulation system in which both stabilizer and oil come from a single biomass source. Overall, this work establishes a scalable, energy-efficient, carbohydrate-driven route to high-oil-loading powders that meets the growing demand for surfactant-free, sustainable formulations.

1. Introduction

The microencapsulation of edible oils to form oil-filled powders has become important, especially in the application of nutraceuticals (Bakry et al., 2016; Sandhya et al., 2023). Such oils are prone to oxidation and the detrimental effects of air and moisture on their quality has been demonstrated (González et al., 2016). Current spoilage prevention strategies have limitations: avoiding heat restricts processing routes and potential applications, reliance on the oil's antioxidant content reduces the desired health benefits (Laguerre et al., 2015), and sealed packaging requires oxygen free conditions which increases cost and complexity. As a result, there remains a critical need for scalable, energy-efficient encapsulation strategies that protect oxidation-sensitive oils while minimizing additive use and processing constraints.

Emerging Pickering emulsion strategies and subsequent encapsulation via drying is one method to circumvent these challenges (Massicotte & Cranston, 2022; Sandhya et al., 2023). On top of this, a recent push for surfactant-free products driven by environmental awareness has propelled the use of bio-based materials in place of petroleum-derived components for microencapsulation (McClements & Gumus, 2016). While numerous bio-based encapsulating agents have been used to stabilize oil-in-water emulsions (Fuchs et al., 2006), cellulose nanocrystals (CNCs) have emerged as excellent Pickering stabilizers due in part to their small size, amphiphilicity and high aspect ratio (Gauthier & Capron, 2021; Kalashnikova et al., 2011; Kedzior et al., 2021).

Our group first showed that CNC-stabilized Pickering emulsions can be processed into redispersible oil powders (via freeze drying) and indicated that by adsorbing methylcellulose (i.e., a food-safe, non-ionic polymer, MC) onto the CNC surfaces before emulsification it became

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possible to form highly stable emulsions with a lower overall CNC content (Hu et al., 2016, 2015). Massicotte and Cranston further demonstrated that oil powder quality did not significantly vary with drying technique, as evidenced by the comparison between freeze drying, spray drying and spray-freeze drying (Massicotte & Cranston, 2022). Notably, the addition of tannic acid (TA), a plant-derived polyphenol that acts as a crosslinker to "set" the emulsion shell and prevent MC entanglement as droplets come together during drying, enables the resulting oil powders to be redispersed in water with simple hand shaking (Hu et al., 2016).

Canada is one of the largest suppliers of oats, and as such developments in the oat industry meaningfully impact our economy (FAOSTAT: Food and Agriculture Organization, Statistics Division, n.d.). Oats contain large quantities of diverse antioxidants and biomolecules, making them sought-after for food, cosmetic, and pharmaceutical applications (Gilissen et al., 2016; Halima et al., 2015). Although oat oil is a by-product of grain refining, its utilization remains limited, in part because its high levels of polyunsaturated fatty acids make it prone to rapid spoilage through oxidation (Banaś & Harasym, 2021; Warner & Gupta, 2003). Considering the potential of oil microencapsulation to extend shelf life by reducing oxygen exposure, we hypothesized that the value of oat oil could be enhanced (and by-product waste from the oat industry reduced), through the preparation of dry oat oil powders from Pickering emulsions co-stabilized by CNCs, MC and TA.

Despite the promise of Pickering emulsions for oil encapsulation, translating these systems into dry powders remains challenging due to the complex stresses imposed during drying, particularly under industrially relevant spray-drying conditions. Spray drying is a technique that converts a liquid feed into a powder by atomizing it into fine droplets and rapidly evaporating the solvent with hot gas. Compared with other drying methods, spray drying can be a faster and semi-continuous process using significantly less energy. Nevertheless, it subjects the liquid feed being dried to high heat (albeit with a short residence time), and high capillary and shear forces which can break emulsions and reduce oil powder yields. Moreover, the process of spray drying, and quality of the resulting oil powders are dependent on the feed and aspirator rate as well as the liquid feed concentration, inlet and outlet temperatures. The large number of impactful process parameters makes spray drying a challenging process to optimize, especially for the drying of sensitive ingredients, like oil powders (Alhajj et al., 2021; LeClair et al., 2016; Vehring, 2008).

There are very few examples in the literature of CNC-stabilized oil powders specifically produced by spray drying. One, published by Esparza et al. (2020), reported spray dried hempseed oil powders stabilized by CNCs alone at high concentrations of 1.1 to 1.4 g CNCs/g of oil. Another by Xie et al. (2019), produced redispersible camellia oil powders by spray drying emulsions that were co-stabilized by 0.3 g CNCs per g of oil and 0.5 g hydroxypropyl methylcellulose per g of oil (i.e., 0.8 g of total stabilizers per g of oil). Their work showed that cellulose derivatives as co-stabilizers can be used as matrix formers allowing for spray dried oil powders to be produced with lower quantities of CNCs. However, these stabilizer contents are exceptionally high overall given that Kalashnikova et al. (2011) determined that only 60% of the oil droplet interface needed to be coated with CNCs for stable emulsions. This suggests that the emulsion stabilizers may also be acting, for example, as heat protectors, flow agents, matrix formers, and redispersion aids. Nevertheless, under the right processing conditions, we believe the oil content in oil powders can be maximized without the need for excess stabilizer.

In our lab, while comparing the impact of drying technique we also successfully produced oil powders from CNC-stabilized emulsions with a variety of oils (corn, lavender, tea tree and jojoba oil) which required only 0.015 to 0.03 g of CNCs (0.03 to 0.06 g of total stabilizer) per g of corn oil to be spray dryable and redispersible. Overall, we previously recommended spray drying for drying CNC-stabilized emulsions because it produced redispersible powders with the same high oil content as the other drying methods but generally offered faster processing speed and lower energy consumption (Massicotte & Cranston, 2022). However, despite these advances, the spray drying of Pickering emulsions has never been systematically optimized using multivariate approaches, and the influence of polysaccharide-polyphenol interfacial interactions on drying performance and redispersion behavior remains poorly understood.

Herein, we establish a fully bio-based encapsulation system in which oat oil is stabilized by a synergistic combination of CNCs, MC and TA and we present the first design of experiment (DOE)-guided optimization of spray drying conditions for a Pickering emulsion. The novelty of this work lies in the integration of biomass-derived Pickering stabilization with systematic, multivariate process optimization and feedstock circularity, enabling both mechanistic insight and practical translation to dry, shelf-stable powders. We compare how the oil's extraction method (cold-pressed vs. solvent-extracted) influences emulsion stability, then use a five-factor DOE to quantify the effects of CNC content, feed dilution, aspirator rate, pump rate, and inlet temperature on powder yield and quality. Finally, to highlight the valorization potential of agricultural side streams, we extract CNCs directly from oat hulls and demonstrate an "All Oat!" version of the encapsulated powder, in which both stabilizer and oil originate from a single biomass source. Ultimately, we hypothesize that synergistic interactions among biomass-derived CNCs, MC, and TA will produce polysaccharide-rich interfacial films that resist coalescence during both emulsification and subsequent spray drying. We further propose that careful optimization of drying parameters (inlet temperature, feed composition, solids content) is essential to preserve these assemblies, thereby maximizing oil loading, yield, and reconstitution performance in the final dried powders.

2. Materials and methods

2.1. Materials

Methylcellulose (MC, M_w = 41,000 g/mol, degree of substitution 1.5–1.9, 400 cP), tannic acid (TA), sodium phosphate dibasic (Na_2HPO_4), sodium phosphate monobasic dihydrate ($NaH_2PO_4{\cdot}2H_2O$) and sodium chloride (NaCl) were purchased from Sigma-Aldrich (Oakville, Canada). Spray dried sulfated cellulose nanocrystals (CNCs) in the sodium salt form were produced by CelluForce (Montreal, Canada) and used in all experiments except the last section where lab-made CNCs were isolated from oat hulls sourced from Prairie Oat Fuel (Tisdale, Canada). The average dimensions of the CelluForce CNCs used were 128 × 8 nm measured by atomic force microscopy (AFM) and the surface charge content was 203 ± 6 mmol/kg CNCs determined by conductometric titration. Cold-pressed oat oil from *Avena sativa* was purchased from Nature in Bottle (Lyon, France). Ethanol-extracted oat oil was purchased from Mystic Moments (Fordingbridge, United Kingdom). All water used was MilliQ grade (Barnstead™ GenPure™ Pro, Thermo Fisher Scientific, Waltham, MA) with a resistivity of 18.2 MΩ·cm (referred to henceforth as 'purified water'). A 1.51 wt% MC solution was prepared by mixing MC and purified water in an oil bath at 60–70 °C for 4 h, then placing it in a fridge (4 °C) overnight to fully dissolve. The dispersion-dissolution process was repeated if the resulting solution was not homogenous and fully transparent after cooling.

2.2. Pickering emulsion preparation and method for redispersion after drying

Oat oil-in-water emulsions stabilized by 0.03 g CNC/g oat oil and 0.02 g MC and 0.2 g TA/g oat oil (0.07 g total stabilizer/g oat oil) were prepared as described previously (Massicotte & Cranston, 2022). The recipe we describe here represents the "centre point" recipe for our design of experiments (DOE), which is described with more detail in Section 2.4. In our DOE, the amount of CNCs included in the recipe was varied, while the amounts of all other ingredients were held constant.

To prepare an emulsion stabilized by 0.03 g CNC/g oat oil and 0.02 g MC and 0.02 g TA with a final total volume of 45 mL that is 2 parts oat oil, 13 parts water (by volume), CNCs (6.6 g of a 2.5 wt% CNC suspension), MC (7.95 g of a 1.51 wt% solution), and NaCl (1 M, 1.2 g) were combined and stirred at 500 RPM. To this mixture, phosphate buffer (69 mM, pH 5.16, 8.65 g) was added and the stirring rate was increased to 1200 RPM. A homogenous pre-emulsion was prepared by adding 5.6 g oat oil in a dropwise manner to the vortex formed in the aqueous phase. This pre-emulsion was probe sonicated (Sonifier 450, Branson Ultrasonics) in 15 mL batches for 60 s with 50% pulses at 60% output in an ice bath. This stage of emulsion (called CNC-MC oat oil emulsion) is stabilized by 0.69 wt% CNCs, 0.5 wt% MC, 50 mM NaCl and 20 mM phosphate buffer (in the aqueous phase) and is 2 parts oil, 8 parts water (by volume). At this point, 15 mL of a 0.8 wt% TA aqueous solution was added to the sonicated emulsions and stirred for 20 min before probe sonicating it again in 15 mL batches for 60 s with 50% pulses at 60% output. This stage of emulsion (called CNC-MC-TA oat oil emulsion) is stabilized by 0.42 wt% CNCs, 0.31 wt% MC, 0.31 wt% TA, 31 mM NaCl and 12 mM phosphate buffer (in the aqueous phase) and overall is 2 parts oat oil to 13 parts aqueous phase (by volume). All emulsion samples were stored in the refrigerator at 4 °C prior to spray drying. After spray drying, oat oil powders (0.25 g) were added to purified water (10 mL) and resuspended by shaking by hand for 10 s before vortex mixing for 10 s. This hand-shaking-vortex mixing process was repeated three times to ensure redispersion.

2.3. Spray drying of the emulsions

Spray drying of emulsions was performed following established laboratory-scale protocols for oil encapsulation (Massicotte & Cranston, 2022), with modifications as described below. Each sample (30 mL sample volume) was diluted from 2 to 8× using a stock solution of 50 mM NaCl and 20 mM phosphate buffer (pH adjusted to 5.16) and spray dried using a laboratory scale Mini Spray Dryer (Buchi B-290, Switzerland). The additional variable process parameter settings (aspirator rate, pump/feed rate, and inlet temperature) are provided in Table 1. For all samples, the spray gas flow of compressed air was 414 L/h. The liquid feed rate ranged from approximately 1.4 to 4 mL/min depending on pump rate which was adjusted between 4 and 12%. The liquid volume ranged from 60 to 240 mL depending on emulsion dilution; thus, spray drying one sample would take between 15 and 170 min. The inlet temperature (ranging between 130 and 160 °C) was set and allowed to stabilize for 30 min before purified water was pumped through the spray dryer for 10 min at a pump rate of 10% to prime and clean the equipment. The pump rate was then set for the trial, and purified water was replaced with the feed emulsion to commence drying. We note that the emulsion was stirred with a magnetic stir bar throughout feeding into the spray dryer and at the end of the spray drying, oil powders collected from the particle collector vessel were stored in the refrigerator at 4 °C.

Table 1
Factorial design parameters & ranges. (Note that the MC and TA content is held constant at 0.02 g MC and 0.02 g TA/g oat oil and only the CNC concentration in the emulsion feed changes).

Parameter		Low	Centre	High
x_1	CNC content (total g)[#]	0.0825	0.165	0.330
x_2	Feed dilution	2	4	8
x_3	Aspirator rate (%)	76	88	100
x_4	Pump rate (%)	4	8	12
x_5	Inlet temperature (°C)	130	145	160

[#] For the DOE analysis, the value for total grams of CNC stabilizer was used in the calculation (Table 2). These values for total g correspond to 0.015 g CNC/g oat oil (Low), 0.030 g CNC/g oat oil (Centre) and 0.059 g CNC/g oat oil (High). Total g CNC and g CNC/g oat oil are related by a factor of 5.6 g (i.e., the total amount of oat oil in all emulsion recipes).

2.4. Design of experiments

A five-factor DOE was chosen to study the effects of CNC content (x_1), feed dilution (x_2), aspirator rate (x_3), pump rate (x_4), and inlet temperature (x_5) on oat oil powders. A half-factorial DOE was performed to gain information on the individual effects of the five parameters as well as their two-factor interactions without the expense of a full factorial DOE. This fractional factorial required the spray drying of 16 batches of emulsions, plus a thrice-replicated centre point to measure precision. The order of trials was randomized, except for the centre point repeats which were spaced at the beginning, middle and end. The low, centre, and high values for all five parameters are shown in Table 1.

The first (x_1), CNC content, had a low boundary of 0.015 g CNC/g oat oil (0.0825 total g CNC) which was selected based on the CNC content deemed necessary by Massicotte and Cranston (2022) to allow for successful spray drying of powders. The high boundary of 0.059 g CNC per g oat oil (0.059 total g CNC) was constrained by our ability to form a high enough concentration stock CNC suspension. For feed dilution factor (x_2), previous reporting indicated dilution factor to be directly related to spray drying process yield, thus it was included in our DOE in attempt to better quantify and understand these effects. The dilution factors of 2, 4 and 8× were selected and 8× was chosen as the highest boundary in order to save time, energy associated with running the spray dryer, and use of fresh water. The third factor tested was aspirator rate (x_3). The Buchi B-290 operation guide recommended processing samples at an aspirator rate of 100% (the maximum setting) to maximize the efficiency of drying and ideal particle separation. However, it has been noted that it is possible to form oil powder from emulsion feeds at aspirator rates as low as 76% (Massicotte & Cranston, 2022), thus this was set as the lower boundary for our DOE. The centre point and outer boundaries for pump rate (x_4) and rate of the emulsion feed were established as follows. Even at pump rates as low as 1%, product was formed but we note that these trials would have taken more than 10 h to dry and preliminary tests resulted in oil powder product discolouration; thus, to keep all trials below 4 h, the lower boundary of 4% pump rate was selected. This low pump rate corresponds to a feed flow rate of 1.4 mL/min. The high boundary of 12% (feed flow rate = 4.0 mL/min) was selected since above this pump rate, we observed clumpy powders indicating that the feed rate was too fast and did not allow for powders to fully dry before coming in contact with other powder particles. The middle pump rate of 8% corresponds to a flow rate of 2.1 mL/min in our spray dryer. Finally, inlet temperature (x_5) was observed to be a parameter with a limited testable range: the upper limit of inlet temperature was restricted to 160 °C as higher temperatures resulted in oil powder discolouration and the smell of burnt oats. Temperatures below the lower limit of 130 °C resulted in gum-like products or visible oil leakage.

For all emulsion samples, oil powder yield, moisture content and emulsion droplet size upon redispersion were measured. Statistical analysis was only carried out for properties that changed outside the measurement precision. This statistical analysis was performed in IBM SPSS software, which is a subscription-required software platform for statistical computing, modelling and more.

2.5. Production of CNCs from oat hulls

Oat hulls (200 g) were finely ground using a compact blender before sieving (250 μm mesh sieve) to remove larger components. The yield of this sieving step was 88 g (44%). Prior to hydrolysing the cellulose to isolate CNCs, two sequential extractions were performed on the sieved hulls. First, NaOH (aq) (1.75 M, 2 L) was added to the sieved oat hulls (80 g) and stirred at 70 °C for 2 h before the base was quenched to pH 7 using HCl (aq). The oat hull solids were retrieved via centrifugation (5000 RPM, 15 min) using a Sorvall RC-5 Superspeed Refrigerated centrifuge (Dupont) at which point an oil extraction step was used to remove the visible oil layer at the surface of the centrifuged pellet. Two subsequent washes with acetone (5000 RPM, 15 min) were sufficient to

remove the oil layer and the resulting solid was dried in an 80 °C oven overnight. Next a peroxide bleaching step was used to remove residual lignin and hemicellulose from the oat hull solids. In a 2 L beaker, 50 g of oat hull solids was combined with NaOH (aq) (5 wt%, 500 mL) that had been preheated to 70 °C. Using a clean and dry separatory funnel, H_2O_2 (24 wt%, 500 mL) was added dropwise to the slowly stirring dispersion of oat hull solids in NaOH (aq) that was being continually heated at 70 °C. This reaction caused vigorous foaming, so the addition was done slowly to minimize yield loss. Once all the H_2O_2 was added, the reaction was mixed for 1 additional hour. This entire pretreatment was repeated using an additional 25 g of oat hull solids and scaling all reagents appropriately, to have a final yield of bleached and purified oat hull solids equal to 10 g (5% overall yield).

CNCs were isolated from the bleached oat hull solids using a H_2SO_4 (aq) hydrolysis and following the typical lab protocol (Cranston & Gray, 2006). The bleached oat hull solids (10 g) were dried at 80 °C overnight. Next, 175 mL of 64 wt% H_2SO_4 (aq) (adjusted using a hydrometer) was heated to 45 °C added to the bleached oat hull solids. The hydrolysis reaction was carried out for 45 min at 45 °C with constant mechanical stirring. To quench the hydrolysis reaction, the CNC-acid slurry was diluted with 1.75 L of ice-cold purified water (i.e., a 10× dilution). The suspension was left to sediment for ca. 1 h before the supernatant was decanted and the CNCs were rinsed of the remaining acid using subsequent rounds of centrifugation (6000 RPM, 10 min) until the CNCs no longer sedimented and remained colloidally stable in suspension. The CNC suspensions were then dialyzed in cellulose membrane dialysis tubing (MWCO = 14,000 Da, Sigma Aldrich) against purified water to remove any residual acid until the pH of the CNC suspension had stabilized (to pH 5.5). Samples were stored in the fridge at 0.25 wt% concentration for characterization and emulsions were prepared following the procedure above that used CNCs from CelluForce. The recovery yield of purified CNCs from ground oat hulls was 0.5 g (0.25%). ζ-Potential and apparent hydrodynamic particle size were determined using a Zetasizer Nano-ZS (Malvern Panalytical, Malvern, U.K.). The electrophoretic mobility of 0.25 wt% CNC suspensions with 5 mM salt was measured using a Folded Capillary Zeta Cell (Malvern Panalytical, Malvern, U.K.). Electrophoretic mobility was converted to ζ-potential according to the Smoluchowski theory (Lin et al., 2019). Dynamic light scattering (DLS) measurements were carried out on 0.025 wt% CNC suspensions (no added salt) using polystyrene cuvettes. The measured diffusion coefficient was converted to apparent hydrodynamic size using the Stokes−Einstein equation with the assumption that the CNCs are spherical (Bhattacharjee, 2016), thereby resulting in merely relative quantities. Conductometric titrations were carried out to determine the total surface charge of the CNCs. Dilute CNC suspensions were titrated against a 10 mM standardized NaOH using 250 μL increments. Changes in pH and conductivity were monitored simultaneously, and volume corrected conductivity was plotted as a function of volume of NaOH added. The equivalence point of the curve was used to determine CNC surface charge.

The oat-hull derived CNCs were also used to make an "All Oat!" version of the oat oil Pickering emulsions and oil powders as described above, where "All Oat!" is a playful twist on "All Out!" — a common phrase meaning to go all-in, make a full effort, or do something completely.

3. Results and discussion

3.1. Pickering emulsion formulation and stability

Stable oat oil-in-water Pickering emulsions were prepared using cellulose nanocrystals (CNCs), methylcellulose (MC) and tannic acid (TA) as co-stabilizers. Our preparation method was adapted from Hu et al. (2016), and optimized for spray drying by Massicotte and Cranston (2022). Briefly, CNCs and MC were combined in the presence of NaCl (to screen the CNC surface charge) with phosphate buffer to stabilize the pH. In our efforts to produce a food-safe product, the previous emulsion recipe was modified to replace the HEPES buffer (Hu et al., 2016; Massicotte & Cranston, 2022) because phosphate buffer is generally regarded as safe (GRAS) according to the U.S. Food and Drug Administration (CFR — Code of Federal Regulations Title 21, n.d.). Oat oil was added to the water phase and emulsified by sonication. To ensure the dry emulsions would be redispersible, TA was added, and the mixture was sonicated again. In this initial formulation (i.e., the "centre point" recipe for our design of experiments (DOE) optimization which will be discussed in the forthcoming results and discussion Section 3.4), the emulsion stabilizer-to-oat oil mass ratio was 0.07 g per g oat oil (where 0.03 g per g oat oil is CNCs). The order of mixing is critical — if the CNCs and MC are not premixed, or if ingredients are added in a different sequence, emulsions cannot be formed (Hu et al., 2016). We refer to the final Pickering emulsions prepared following this recipe as CNC-MC-TA oat oil emulsions.

Prior to drying the CNC-MC-TA oat oil emulsions into oil powders and conducting our DOE, the stability of this initial recipe (i.e., an emulsion containing 0.03 g CNC per g oat oil) was assessed. This emulsion had small droplets that were highly stable against coalescence over 21 days of storage (Fig. 1, Supplementary Methods). No significant changes in oil droplet size (or size distribution), or surface area moment mean (i.e., D[3,2] value or Sauter mean) diameter were observed either. The droplet diameter of 5.5 μm (span of 1.7 μm) falls in line with other reported stable emulsions with CNCs as Pickering stabilizers (Dong et al., 2021). The primary droplet size of the initial emulsion was also measured by optical microscopy to be 5 ± 2 μm and minimal agglomeration was observed (Supplemental Data Fig. S1). The relatively small droplet size and high resistance to droplet coalescence of the oat oil emulsions is attributed to the ability of CNCs to form a steric barrier at the oil-water interface in combination with the water-soluble MC which is surface active (Hu et al., 2015; Xie et al., 2019).

Consistent with previous studies (Massicotte & Cranston, 2022), emulsions with TA had slightly larger droplet sizes compared to emulsions stabilized by only CNCs and MC (Supplementary Data Fig. S1). However, the long-term stability of the emulsions with and without TA was similar and the TA is needed to reinforce the droplet shell via catechol binding which is important for the desired processability and

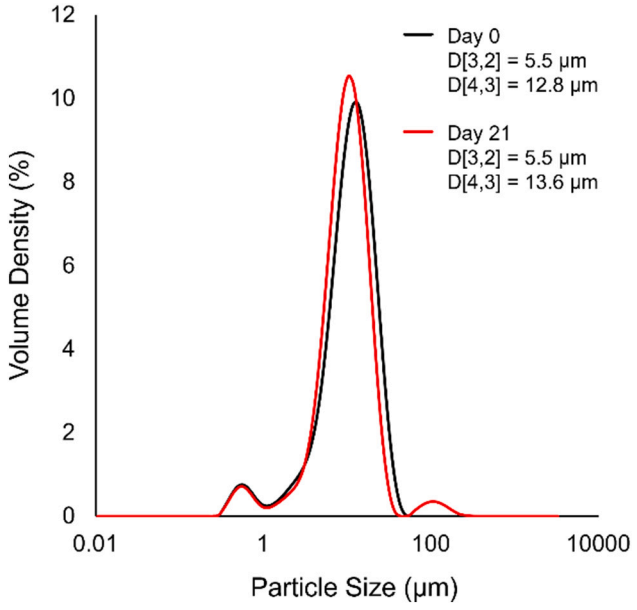

Fig. 1. Oil droplet size distribution and mean surface area (D[3,2]) and volume (D[4,3] diameters (inset values) determined by laser diffraction for the "centre point" CNC-MC-TA oat oil emulsion at day 0 (black line) and after 21 days of storage in the fridge (red line).

functionality. Additionally, while MC-only stabilized oil emulsions have been reported (Hu et al., 2014), we found that MC-only stabilized oat oil emulsions were not stable and exhibited macroscopic oil leakage over three weeks (Supplementary Data Fig. S2). As such, the ability to spray dry oat oil emulsions without CNCs or without TA was not investigated.

3.2. Effect of oat oil type on emulsion stability

We tested two different oat oils in the Pickering emulsions — one extracted using cold-pressing and one extracted using ethanol. Emulsions prepared with the two oil types were distinctly different in stability: the emulsions with ethanol-extracted oat oil exhibited oil leakage when stored in the fridge overnight, whereas the emulsion with cold-pressed oat oil was presented in Fig. 1 and was stable for over 21 days. This demonstrates how small changes in complex oil chemical composition can affect emulsions.

The fatty acid compositions of both oil types (from safety data sheets) are reported in Supporting Data Table S1. To supplement this information, we conducted TGA to indirectly assess differences in composition (Fig. 2a) and performed a ferrous oxidation-xylenol orange (FOX) assay to quantify peroxides (Fig. 2b) (Nourooz-Zadeh et al., 1994). Methods for these experiments are described in Supplementary Data. TGA indicated that the ethanol extracted oat oil was slightly more complex since it had a broader DTG peak. In contrast, the FOX assay showed that the cold-pressed oil had a higher hydroperoxide concentration. More hydroperoxides can represent more oil oxidation (or sometimes rancidity) (Nourooz-Zadeh et al., 1995), but at these low concentrations, neither oil is considered to have gone bad. These hydroperoxides are more polar than unoxidized and unsaturated fatty acids, so one reason for the increased emulsion stability we observed using cold pressed oil is that the oil itself is more polar than the ethanol extracted oil. Another potential explanation is that ethanol extraction may reduce the content of naturally occurring polar lipids that can act as endogenous emulsifiers in cold-pressed oils (Härröd, 2014). So, when forming our emulsions using ethanol extracted oat oil, we lose out on some synergistic stabilizing effects of other naturally emulsifying molecules. This result is similar to the improved stability of corn oil emulsions stabilized by CNCs compared to model hydrocarbon oils (Liu et al., 2018). As such, all results presented outside of this section are from experiments done using the cold-pressed oat oil.

3.3. Dried oat oil powder composition and assessment of oil encapsulation

Spray drying resulted in the successful production of CNC-MC-TA stabilized oat oil powders. We assessed "success" in two ways: (1) the complete removal of water, and (2) high yields of the resulting powder product. This second point is particularly challenging, in fact it is often difficult to even find yields of spray dried oil powders reported in the literature because they are typically very low. Nevertheless, our initial trials with the "centre point" emulsion recipe (Table 1) resulted in dry powders at respectable yields of ca. 55%. On top of this, it is well known that industrial-scale spray drying is more efficient than benchtop systems (once fully warmed up and operating at maximum capacity), thereby circumventing the limitations associated with small-scale processing of low-volume liquid feeds that may constrain studies such as this one.

The combination of stabilizers in our CNC-MC-TA emulsions effectively retained the encapsulated oat oil during spray drying as evidenced by the oil content in the oil powder, determined by TGA (Fig. 3, Supplementary Methods). The solids content and oil content in the oil powders were quantified by integrating the DTG peak at ca. 290 °C ("i" in Fig. 3) attributed to the stabilizers (CNC, MC, TA, phosphate buffer, NaCl), and the peak at ca. 410 °C ("ii" in Fig. 3) attributed to the oat oil, respectively (Massicotte & Cranston, 2022). The ratio of peak i to ii suggests that the majority of the oat oil remained after drying; in fact, our oil powders contained 89.1% oil which is almost at the theoretical maximum of 90.8% (based on the starting emulsion composition).

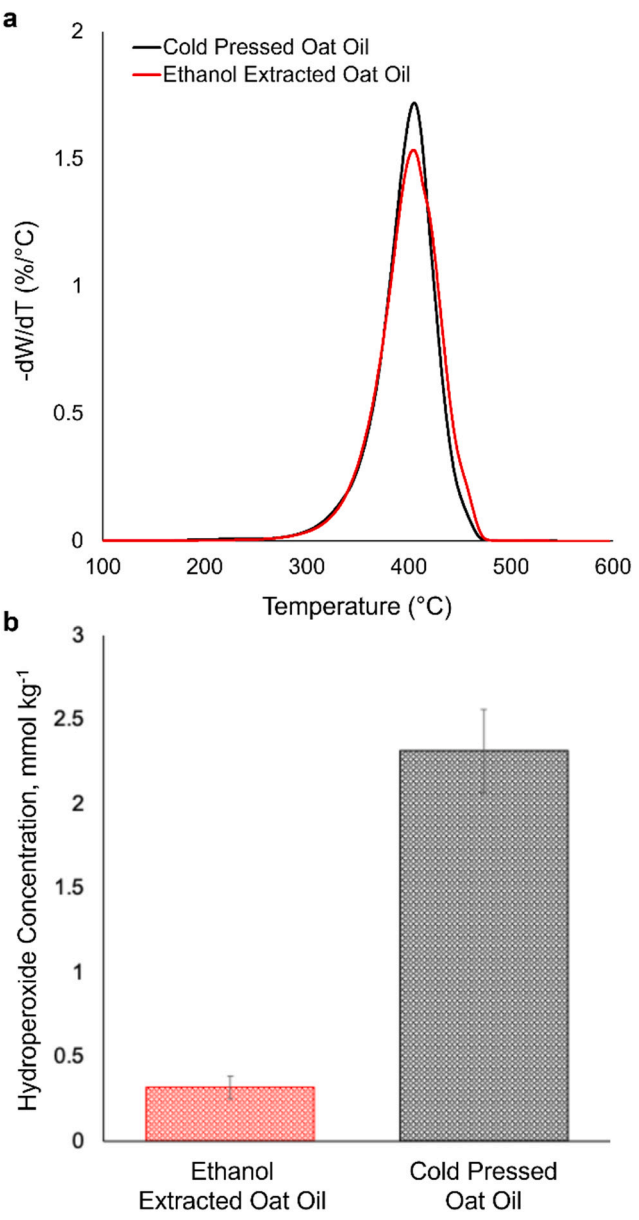

Fig. 2. For both cold-pressed (black) and ethanol-extracted (red) oat oils (a) derivative thermogravimetric (DTG) curves from TGA analysis and (b) results of the ferrous oxidation-xylenol orange (FOX) assay to quantify peroxides are presented.

While TGA indicates the presence of oil, it does not distinguish whether that oil is still encapsulated by the stabilizing layer after spray drying. As such, true encapsulation was inferred by redispersing the oil powders in water to reform an emulsion and examining droplet size by laser diffraction and optical microscopy. Fig. 4 shows that individual droplets of oil persisted without significant coalescence in the redispersed oat oil powder.

The key to producing successful oat oil powders was increasing the stabilizer content relative to our past work. This was necessary because we observed that when the stabilizer content was too low, the oat oil emulsions were particularly sensitive to temperature and shear, leading to oily sludges rather than "dry" powders upon spray drying. Again, the emulsion that is 2 parts oat oil to 13 parts water (by volume) contained 0.42 wt% CNC, 0.31 wt% MC and 0.31 wt% TA in the aqueous phase prior to dilution required for spray drying. This corresponds to 0.07 g total stabilizer per g oat oil (0.03 g CNC, 0.02 g MC and 0.02 g TA per g

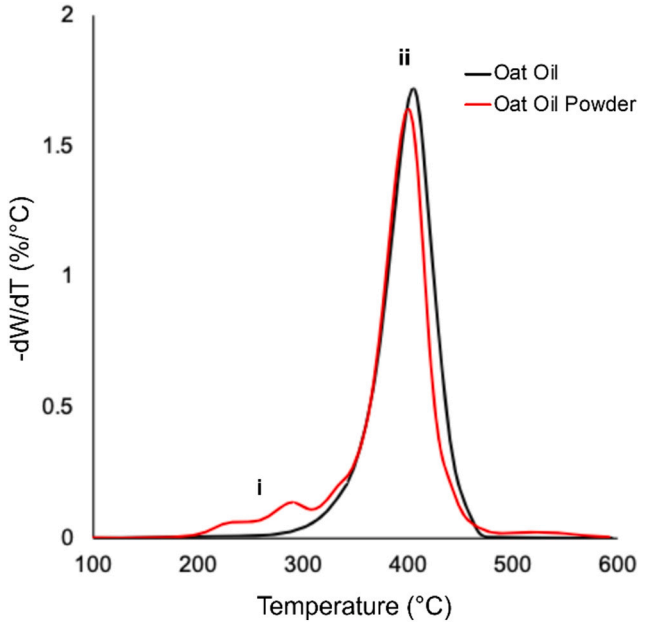

Fig. 3. DTG curves for oat oil (black line) and the spray dried oat oil powder (red line). The peak labeled "i" is attributed to the emulsions stabilizers and the peak labeled "ii" is attributed to the oat oil.

oat oil). This is slightly higher than our previously reported 0.04–0.06 g of total stabilizer per g of oil (with different oils) (Hu et al., 2016; Massicotte & Cranston, 2022). However, we achieve high yields with still comparatively low stabilizer content compared to the other literature that used 0.8 to 1.4 g of CNCs or total stabilizers per g of oil (Esparza et al., 2020; Xie et al., 2019).

Esparza et al. (2020) indicated that they needed 10 to 14 layers of CNCs around an oil droplet to stabilize hempseed oil emulsions. We calculate by Eq. S1 that we have 200% of the oat oil droplets covered by CNCs (i.e., 2 layers). Therefore, by incorporating MC as a co-stabilizer to "fill in the gaps", we decreased the ratio of CNCs to oil needed which is consistent with observations regarding the synergistic effects of combining many water-soluble cellulose derivatives with CNCs in emulsions (Hu et al., 2015; Xie et al., 2019). In another example, carboxymethylated (modified) CNCs with hydroxypropyl methylcellulose were used to stabilize camellia oil emulsions that were also redispersible after spray drying (Xie et al., 2019). While relatively similar CNC concentrations were tested (0.11 to 0.56 wt%) much higher co-stabilizer (hydroxypropyl methylcellulose) was needed making the total stabilizer to oil ratio about 8 times more than in our work. From an applications standpoint, we believe that high oil contents will be essential for delivering substantial amounts of valuable oils while minimizing texture, rheological, and chemical interference from excessive stabilizer content.

3.4. Design-of-experiments approach to optimize the oil powder yield from spray drying

A set of 19 emulsions were spray dried to investigate the effects of CNC content, feed dilution, aspirator rate, liquid feed rate (which depends on pump rate), and inlet temperature on the spray dried powder yield, calculated according to Eq. S2, with results shown in Table 2. From the runs, 14 of the 17 spray drying conditions produced powders. These powders ranged in colour, particle size, and texture (Supporting Information Table S2). The highest yield obtained was 82% for sample 15, which had the highest CNC concentration and dilution, medium aspirator rate, and lowest pump rate and inlet temperature. Three trials were categorized as "unsuccessful" (samples 4, 6 and 8) — while they

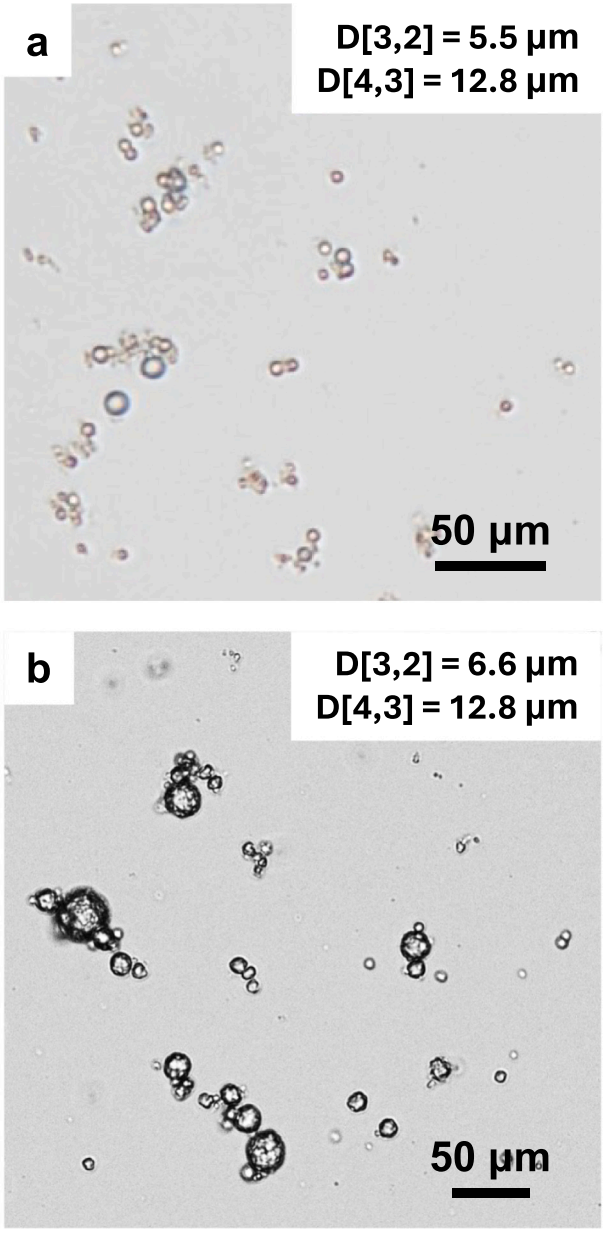

Fig. 4. Optical microscopy images of (a) initial CNC-MC-TA oat emulsion diluted 20× from the original and (b) the dried powder redispersed in water and vortexed.

produced promising granules at the beginning of the spray drying process, they eventually resulted in a thick, brownish product that was tacky, and conformed to the shape of the glass vial (i.e., showed more liquid-like behavior) making them difficult to collect fully. As the yield was non-zero, and re-dispersibility was possible, these trials were still included in the statistical analysis. Overall, the yields ranged from 9 to 82% with an average of 50% and five samples performing at 60% yield or above. While moisture content and redispersed emulsion droplet diameters were measured for all samples (Table S3), the differences were fairly small and our model was not predictive for these properties, as such, only the model for yield is discussed below.

3.5. Yield model

A multiple linear least-squares regression was performed using the SPSS software and the following model (Eq. 1) was generated to predict

Table 2
Yield of oat oil powders from spray drying with varying CNC content, feed dilution, aspirator rate, pump rate, and inlet temperature.

Sample i.d.	CNC content, x_1 (total g)	Feed dilution, x_2 (×)	Aspirator rate, x_3 (%)	Pump rate, x_4 (%)	Inlet temperature, x_5 (°C)	Yield (%)
1	0.0825	8	100	12	160	60
2	0.0825	2	100	12	130	52
3	0.0825	8	76	12	130	56
4	0.0825	2	76	12	160	21
5	0.0825	8	100	4	130	51
6	0.0825	2	100	4	160	14
7	0.0825	8	76	4	160	25
8	0.0825	2	76	4	130	9.4
9	0.330	8	100	12	160	81
10	0.330	8	100	12	160	49
11	0.330	8	76	12	130	47
12	0.330	2	76	12	160	52
13	0.330	2	76	4	160	79
14	0.330	2	76	4	160	56
15	0.330	8	100	4	160	82
16	0.330	2	76	4	130	52
17	0.165	4	88	8	145	57
18	0.165	4	88	8	145	47
19	0.165	4	88	8	145	60

the median yield (Y) depending on CNC content (x_1), feed dilution (x_2), aspirator rate (x_3), and pump rate (x_4). While inlet temperature (x_5) was included in the model design, the factor's standard error was larger than the estimated coefficient value indicating the factor does not have a meaningful impact on the output we investigated (i.e., yield).

$$Y = -64.15 + 231.2x_1 + 3.421x_2 + 0.5039x_3 + 4.161x_4 - 16.42x_1x_4 \quad (1)$$

The magnitude of each parameter's effect in the statistical model predicting oat oil powder yield is shown in Fig. 5a. The R^2 (coefficient of determination) for this model is 0.80 (Fig. 5b), the Q^2 (goodness of prediction) is 0.59 and the *p*-value is 0.00034 meaning the null hypothesis should be rejected, and this model accurately describes the effects of spray drying parameters on oil powder yield (Ericksson et al., 2000).

Because the DOE used nonstandard factor ranges (i.e., the high and low levels were not coded as +1 and −1), the raw regression coefficients are not directly proportional to the true influence of each variable on the output. To interpret these effects, each coefficient was multiplied by the magnitudes of both the high and low input values for that factor. The average of these products represents the variable's overall contribution to the response, while the difference between them reflects the output's sensitivity to that variable. These calculations are summarized in Table 3.

The most significant positive effect on yield was the CNC content of the emulsion (overall contribution 48). Notably the difference here between the high- and low-value products (i.e., the span of the scaled effects) is also large in magnitude (sensitivity 57) meaning that large changes in CNC content will lead to large changes in yield. High CNC content means sufficient oil droplet surface coverage which prevents oil leakage and droplet coalescence. It also means that CNCs may act as a sacrificial thermal stabilizer protecting the oil against oxidation during spray drying. Notably the "high CNC content" recipe contains 0.06 g CNC/g oat oil and corresponds to a theoretical emulsion droplet surface coverage of 400% (4 layers). Samples 9 to 16 all have high CNC content and represent the upper end of yields at 49 to 82%. This result agrees with other literature examples that also prefer larger CNC concentrations for effective spray drying of oil powders (Esparza et al., 2020). While increasing CNC content is favorable for yield, as noted above, we still wanted to keep the stabilizer content low to maximize oil content in the oat oil powders — this content, which corresponds to 0.84 wt% CNCs in the water phase prior to dilution is a good trade off point.

Almost as significant to the yield of spray drying as the CNC content in the DOE-generated model was aspirator rate (overall contribution 44). However, the sensitivity of the yield to changes in this variable within the established experimental boundaries is low (12). A large overall contribution combined with low sensitivity indicates that aspirator rate exerts a consistently strong effect on yield across its experimental range, rather than causing large fluctuations as its level changes. Increasing aspirator rate effectively increases the rate of airflow through the spray drying system which directly affects the drying efficiency and can help to increase yield in a few ways: higher aspirator rate (1) creates a stronger vacuum, which improves the transport of dried particles into the cyclone separator and (2) stronger airflow improves mixing between hot air and atomized droplets, which promotes more uniform drying.

Increasing the pump rate (%) and thus the feed rate (mL/min) also had a positive effect on the yield of spray drying oat oil powders (overall contribution 33, sensitivity 33). Since the size of the droplet emerging from the spray drying nozzle depends on solution viscosity, surface tension, and the mass ratio of atomizing air to liquid feed, increasing the liquid feed flow rate by adjusting the pump rate at constant atomizing air flow rate should result in larger sprayed droplets and hence larger particles with a better likelihood of being retained in the cyclone (Maury et al., 2005). Faster pump rates also have the advantage of faster processing with no real disadvantage to increasing the feed, although theoretically, too high a pump rate could lead to powders that do not dry fully before coming in contact with other droplets leading to clumping, however this was not observed below a 12% pump rate setting (which corresponds to a feed flow rate of 4 mL/min).

The DOE results indicated that dilution ranked last in importance of parameters (overall contribution 17, sensitivity 21). So, more dilute emulsion feeds led to higher spray drying yields but this is less significant to both overall yield and the variability of the output. What affect we did observe is attributed to the fact that the thermogelation of MC was mitigated by dilution (Chevillard & Axelos, 1997). Furthermore, shear forces are lower for less viscous (i.e., more dilute/non thermally-gelled) samples, suggesting that less "damage" to the emulsion due to physical stresses led to higher yields. Of course, the dilution parameter significantly affects the time and energy cost for spray drying, because more dilute samples take longer to dry. The dilution level could therefore be optimized further to avoid "diminishing returns"; in our case, a 2× dilution sample was spray dried in as little time as 15 min, whereas an 8× diluted sample could extend the process to as long as 2 h 50 min.

Finally, one significant cross term – pump rate × CNC content – had a strong negative correlation with yield (overall contribution −35, sensitivity −60). Notably, this variable exhibits the greatest influence on yield variability. The generation of a five-dimensional contour plot (Fig. 6) enables further investigation of this interaction profile. The model indicates that the highest yields occur at low pump rates and high CNC contents, with yield being more sensitive to the change in CNC

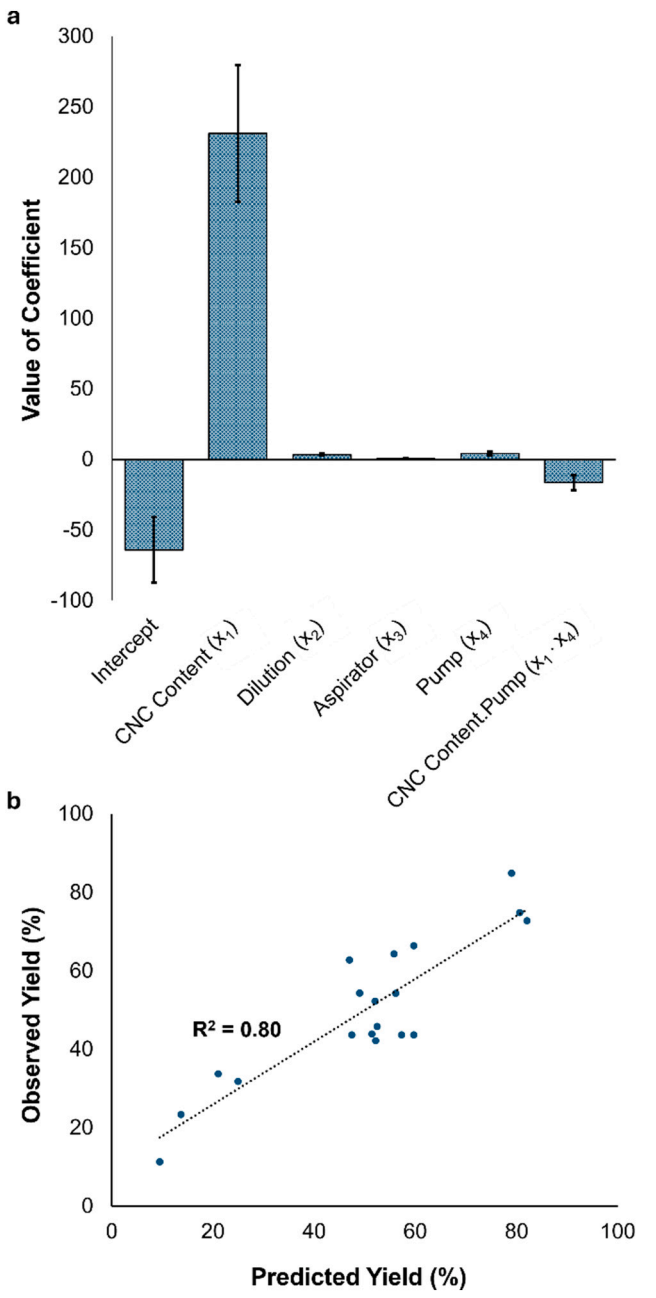

Fig. 5. Key results of spray drying optimization for yield using design of experiments statistical approach including (a) calculated constants for the five term regression model, $Y = -64.15 + 231.2x_1 + 3.421x_2 + 0.5039x_3 + 4.161x_4 - 16.42x_1x_4$ predicting median spray drying yield of oat oil powders. Positive coefficients indicate an increase in the input parameter results in an increase of the output property. Negative coefficients indicate the opposite: an increase in the input parameter results in a decrease of the output property. In (b) predicted vs actual observed yield for the regression model is shown. The coefficient of determination is determined relative to the linear line of best fit (black dotted line). Insight into calculation of value for Q^2 is included in the Supporting Information (Eqs. S3–S4 and Tables S4–S6).

content at low pump rates (where the high-CNC recipe yields 41% more oil powder than the low-CNC recipe) than at high pump rates (where this difference narrows to 8%). The explanation for this behavior is in line with the discussion above that higher pump rate (and larger droplet size) allow more powder to make it to the collector, but a lower pump rate can be counteracted by increasing CNC content, where essentially the mass of each droplet is then increased.

Table 3
Tabulated contributions of each variable tested in DOE to the output (i.e., yield). Overall contribution is the average taken of the High input for each factor multiplied by the corresponding factor coefficient and the Low input for each factor multiplied by the corresponding factor coefficient. Sensitivity is their span.

Parameter		Overall contribution	Sensitivity
x_1	CNC content (total g)	48	57
x_2	Feed dilution (×)	17	21
x_3	Aspirator rate (%)	44	12
x_4	Pump rate (%)	33	33
x_1x_4	CNC content (total g) · Pump rate (%)	−35	−60

Interestingly, the model also indicates that the yield is least sensitive to the pump rate at the mid CNC content value (where the high-pump-rate recipe yields only 6% more oil powder than the low-pump-rate recipe) compared to the high and low CNC values (yield spans of 10% and 22% respectively). This suggests the presence of a critical number of CNC shell layers, above which the effect of pump rate becomes minimized. Beyond this threshold, shear forces may play a more dominant role, reducing the amount of intact oil powder that reaches the collector.

Inlet temperature does not appear to be a significant factor affecting yield within the ranges tested for our DOE (130–160 °C). This range was limited since below 130 °C oil powder samples remained wet and above 160 °C we observed discolouration and a roasting oil smell (for samples that took longer than 1.5 h to spray dry). From our previous work, we recommend spray drying as the most favorable option to achieve nice powders that have the smallest emulsion droplet size upon redispersion; this is attributed to spray drying being the only process tested that avoids a freezing step, which we saw causes extensive emulsion droplet aggregation even prior to drying (Massicotte & Cranston, 2022). However, the high temperature used in spray drying of cellulose-stabilized emulsions is known to be problematic because it induces thermogelation of MC and can cause thermal degradation in the oils being encapsulated. We note that the high thermal stability of sodium-form CNCs (Vanderfleet et al., 2022) means they should not degrade at common spray drying temperatures, especially given the short residence times at elevated temperatures and that evaporation is a cooling process. While yield can sometimes be increased by increasing the inlet temperature (Maury et al., 2005), we suspect our temperature range was too small to observe this effect. We conclude that there is thus a small temperature range, mostly set by oil type, where yield is reasonable and relatively constant but outside of this range, spray drying of emulsions is ineffective.

3.6. Preparation of an "All Oat!" oat oil powder stabilized by CNCs hydrolyzed from oat hulls

To exemplify the potential of underutilized oat farming by-products, the extraction of CNCs from the novel source of oat hulls was investigated. Sequential alkali treatment, oil extraction with ethanol, and chemical bleaching were used to selectively remove non-cellulose oat hull components without breaking down the cellulose (Erdocia et al., 2021; Gao et al., 2023). Following this, sulfuric acid hydrolysis was used to isolate CNCs which were characterized in terms of dimensions, surface charge content, apparent size in suspension (by DLS), and zeta potential. The AFM images (Supplementary Data Methods and Fig. S3) show nanosized rod-shaped CNCs with lengths of 220 ± 70 nm and cross-sections of 80 ± 20 nm. Such low aspect ratio CNCs are not atypical for those isolated from complex biomass. In fact, they are strikingly similar in dimension and morphology to those prepared from sugarcane bagasse (Scopel et al., 2023, 2025). The oat CNCs had a sulfate half ester content of 78 ± 4 mmol/kg CNCs suggesting inefficient esterification during acid hydrolysis — for comparison, sulfated CNCs typically exhibit charge densities in the range of 80 to 350 mmol/kg CNCs (Delepierre et al., 2021; Reid et al., 2017). Despite the low surface

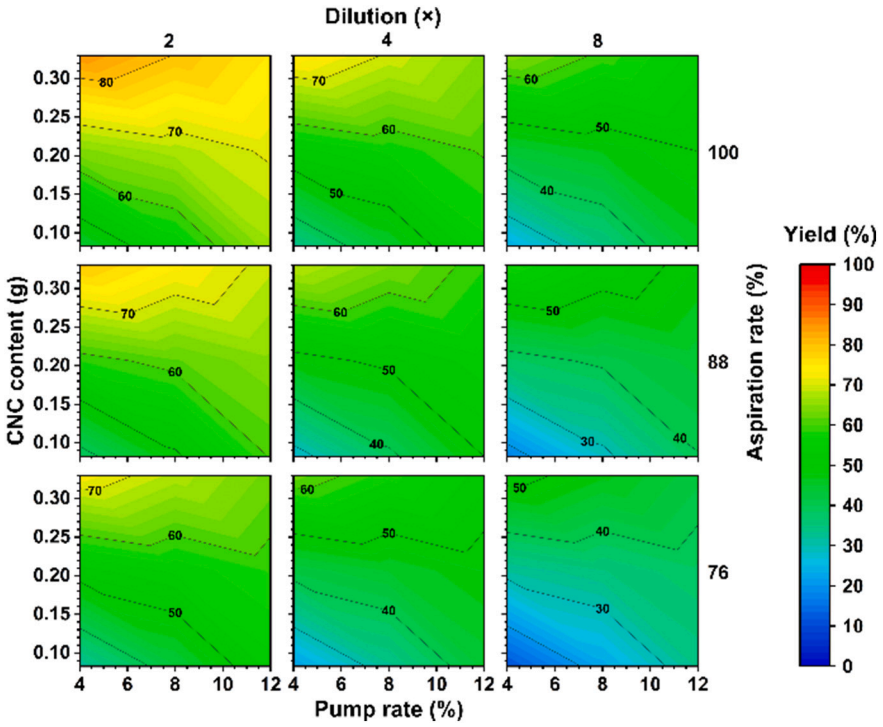

Fig. 6. Five-dimensional contour plot of predicted yield generated using the five-term model (Eq. 1). Yield increases as the coloured bands shift from blue to red. The model predicts that yield is maximized under conditions of high CNC content and low pump rate, with the effect being most pronounced at high aspiration rates and for minimally diluted emulsion feeds.

charge, the oat CNCs were found to be colloidally stable with reasonable hydrodynamic diameters of 111 nm and zeta potential values of −27.7 mV (Foster et al., 2018). These values also vary depending on cellulose source but are in the range for "high quality" CNCs (Vanderfleet & Cranston, 2021).

Compared to the CelluForce CNCs that are isolated from wood pulp and were used to stabilize the emulsions and oil powders earlier in this work, our oat CNCs have a lower aspect ratio and surface charge density. For this reason, we decided to probe how such differences in physicochemical character would affect oat oil emulsion formation. Emulsions were prepared using the "centre point" CNC concentration from Table 1 without any changes to the recipe other than the CNC type. We refer to this emulsion as Oat-CNC-MC-TA. The emulsion droplet size (Fig. 7a) and appearance by optical microscopy are presented in (Fig. 7b) side-by-side with the corresponding "centre point" CNC-MC-TA emulsion (Fig. 7c). Compared with the CNC-MC-TA emulsion, those made with oat-CNCs show more agglomeration of droplets. Importantly, the Oat-CNC-MC-TA oat oil emulsion was stable in the fridge over time and spray dried well, yielding 1.9 g of oil powder, a 47% yield, following our DOE "centre point" recipe. The Oat-CNC-MC-TA oil powders were also redispersible when cycles of hand and vortex mixing were applied (Supporting Information Fig. S4). This result is consistent with the oat oil powder yields achieved using CelluForce CNCs and thus lends credence to the potential of "All Oat!" emulsion products.

4. Conclusions

With the goal of developing a scalable microencapsulation strategy for oat oil-filled powders, we established a spray-dried Pickering emulsion system stabilized by a synergistic combination of bio-based cellulose nanocrystals (CNCs), food-grade methylcellulose (MC), and the plant-derived polyphenol tannic acid (TA). Prior to drying, the emulsions exhibited excellent resistance to droplet coalescence, with cold-pressed oat oil outperforming solvent-extracted oil.

Using a design-of-experiments (DOE) framework, we systematically optimized lab-scale spray-drying conditions for a CNC-stabilized Pickering emulsion for the first time. Among five process variables examined (CNC content, feed dilution, aspirator rate, pump rate, and inlet temperature) CNC content emerged as the dominant factor governing powder yield and quality. Under optimized conditions, spray drying produced powders with yields up to 82%, oil contents of 89%, and moisture contents below 2%, that readily reconstituted into emulsions resembling their pre-dried counterparts. These results demonstrate that spray drying can be an effective and energy-efficient route for converting oxidation-sensitive Pickering emulsions into transportable, rehydratable powders, thereby reducing the economic and environmental costs associated with liquid formulations.

Beyond process optimization, this work highlights the potential for feedstock circularity in carbohydrate-based encapsulation systems. By extracting CNCs directly from oat hulls and applying them to stabilize oat oil, we demonstrate an "All Oat!" encapsulation platform in which both stabilizer and core material originate from a single agricultural biomass stream. This valorization strategy underscores the capacity of polysaccharide nanomaterials to transform underutilized side streams into high-value functional ingredients.

More broadly, we propose that integrating Pickering emulsion design with multivariate process optimization provides a generalizable framework for maximizing oil loading while minimizing stabilizer demand in spray-dried powders. Looking forward, these design principles are expected to guide the development of next-generation, surfactant-free encapsulation technologies for nutraceutical, cosmetic, and pharmaceutical applications, while supporting sustainable manufacturing practices and the expanded use of biomass-derived nanomaterials.

CRediT authorship contribution statement

Megan G. Roberts: Writing – original draft, Visualization, Methodology, Formal analysis, Data curation, Conceptualization. **Keanna Yu:** Writing – original draft, Formal analysis, Data curation. **Marcus A. Johns:** Visualization, Methodology, Formal analysis. **Golshan**

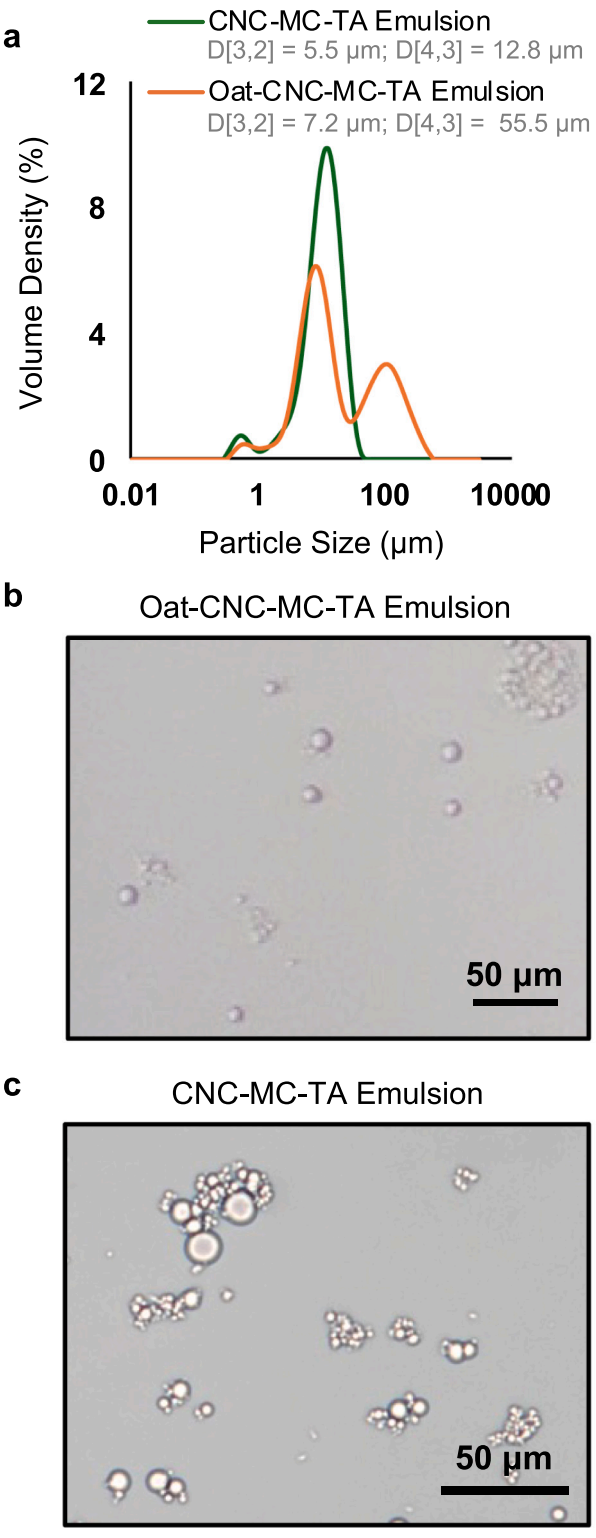

Fig. 7. Comparing CNC-MC-TA and Oat-CNC-MC-TA oat oil emulsion droplet size: (a) oil droplet size distribution and mean surface area (D[3,2]) and volume (D[4,3] diameters (inset values) determined by laser diffraction for the CNC-MC-TA and Oat-CNC-MC-TA oat oil emulsions when they are freshly prepared (i.e., Day 0); optical microscopy images of (a) emulsions stabilized by CelluForce CNCs and (b) emulsions stabilized by CNCs extracted from oat hulls after a 20× dilution.

Matinfar: Data curation. **Emily D. Cranston:** Writing – review & editing, Supervision, Conceptualization.

Declaration of competing interest

Overall, the authors declare no competing interests. All affiliations are listed on the title page of the manuscript. All funding sources for this study are listed in the "acknowledgments" section of the manuscript. The authors have no financial interests or positions to declare and are not members of the journal's advisory board. We also have no related patent applications or registrations to declare.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.carbpol.2026.125031.

Data availability

Data will be made available on request.

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