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Towards spray drying of high solids dairy liquid: Effects of feed solid content on particle structure and functionality

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ABSTRACT

A micro-fluidic-jet spray dryer was used to fabricate spray freeze dried (SFD) and spray dried (SD) uniform microparticles with feed solid content >30 wt% of reconstituted skim milk. The effects of feed solid content on the particle size, morphology, surface composition, wettability and solubility were investigated. The surface composition of SFD and SD sample was characterized via an X-ray photoelectron spectroscopy. Fat and protein was found to be over-represented on the SD particle surface, while lactose significantly declined. Such ingredient segregation was quantitatively shown to occur during atomization and continue within the drying process, i.e. atomization-induced ingredient segregation (AIIS) and drying-induced ingredient segregation (DIIS). The wettability and solubility of the spray dried samples were quantified using scaled-down *GEA Niro Methods* in relation to the feed solid contents.

1. Introduction

Spray dried dairy powder is one of the most important consumables in human society. Critical processing challenges, however still exist with regards to product quality control. In particular, spray drying high solids dairy liquids usually causes a crust to form outside a partially dried droplet during drying, which could trigger an increased in the local temperature, and thus undesirable occurrences such as lipid oxidation, protein denaturation and sugar crystallization. Furthermore, the crust composition can be quite different from the bulk composition due to different surface activities, as well as diffusion and precipitation rates of various dairy ingredients, e.g. lactose, protein and lipid, which has significant influence upon particle properties, e.g. morphology, density, stickiness, flowability, wettability, solubility, etc.

Quantitative investigations on surface (less than 10 nm) composition of industry and lab-scale spray dried powders have been successfully conducted using X-ray photoelectron spectroscopy (XPS) in relation with drying conditions and powder properties (Fäldt and Bergenståhl, 1996). In general it has been found that surface active components, i.e. protein and fat, were overrepresented on the particle surface (Murrieta-Pazos et al., 2012). This phenomenon can be ascribed to their preferential adsorption to the air–liquid interface of the atomized droplets prior to drying, which we call atomization-induced ingredient segregation (AIIS). It implies that the droplet surface composition is already distinctive from the bulk composition. Further ingredient segregation may continue during drying, which we call drying-induced ingredient segregation (DIIS). There is lack of studies to quantify the influences from AIIS and DIIS, respectively, on the particle surface composition, mostly due to the difficulty to correlate a specific particle with the original droplet, since conventional spray dried particles are usually of a wide size distribution resulted from polydisperse atomized droplets (Masters, 1991). Within the same batch of spray dried powders, different particles have likely experienced distinctive drying histories, and thus possess varied morphologies and surface compositions, and consequently different functionalities (Adhikari et al., 2009; Gaiani et al., 2010; Kim et al., 2009).

To better understand the particle forming mechanism via spray drying, a specially designed micro-fluidic-jet-spray-dryer (MFJSD) has been utilized to produce uniform particles by drying monodisperse droplets with high reproducibility among batches. The identical drying history to every individual droplet allows a direct correlation of a certain powder property for a specific drying

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condition. The notable advantages of using this approach have been shown in a few recent studies (Amelia et al., 2011; Liu et al., 2011a,b; Wu et al., 2011a; Zhong et al., 2012). For dairy research, effects of spray drying temperature has been investigated on wettability and solubility of milk protein concentrate and skim milk powders (Rogers et al., 2012).

Herein, reconstituted skim milk with high feed solid contents, ranging from 32.6 to 53.5 wt% (wet basis, in a similar range to those used in industrial spray dryers) was spray freeze dried using liquid nitrogen and spray dried using the MFJSD. The purpose of spray freeze drying experiments was mainly to rapidly solidify the droplet surface for studying AIIS. The effects of the feed solid content were studied on particle morphology and shrinkage ratios (1 – (particle size/droplet size)). Surface compositions of both monodisperse droplets and spray-dried uniform particles were quantified using XPS in relation to powder wettability and solubility.

2. Materials and methods

2.1. Materials and feed liquid preparation

Commercial skim milk powder (SMP) was provided by Dairy Innovation Australia Ltd. Distilled water was used to reconstitute SMP with the solid content of 15 wt% (wet basis). The concentrated liquid was obtained using a rotary vacuum evaporator (BUCHI Rotavapor® R-210, BUCHI Labortecknik AG, Flawi, Switzerland). In particular, 90 g of SMP was dissolved in 510 g of distilled water with stirring at 1000 rpm under 50 °C for 30 min. The concentrating process was controlled under 42 mbar at 50 °C until the liquid reached the targeted total weight. The actual solid content of feed liquid was measured by drying 2 g liquid in a weighting dish with an open lid in the oven at 102 °C for 3 h. Three different feed solid contents were used in this work, i.e. 32.6, 41.4 and 53.5 wt% represented by SMP-32.6, -41.4 and -53.5, respectively.

2.2. Monodisperse droplet formation and droplet size measurement

A specially designed micro-fluidic-aerosol-nozzle (MFAN) was assembled with an orifice diameter of 137 μm. The detailed operating mechanism of the MFAN system can be found in Wu et al., (2011a). The feed liquid was kept in the reservoir and pressured to jet through the MFAN. Droplets were formed by regularly vibrating the piezoceramic component of the MFAN with a sinusoidal electrical signal transferred from a pulse generator (Microfab Technologies, Inc., Texas, US). By controlling the liquid flow rate and vibrating frequency, monodisperse droplet formation can be achieved. The photographs of the droplets (Fig. 1a) were taken using a digital SLR camera (Nickon D90) to measure the droplet size using the software of an image processing and analysis software in Java (ImageJ™).

2.3. Spray freeze-dried (SFD) skim milk powder (SMP) production

The MFAN was placed into a cylinder disperser using air to disperse the monodisperse droplets generated from the feed liquid directly into a bowl of liquid N_2 (Fig. 1b and c). Frozen particles were transferred into a flask attached to the freeze dryer (Labconco, Ltd., Missouri, US) and dried at −80 °C for 72 h.

2.4. Spray-dried (SD) skim milk powder (SMP) production

The MFJSD was used to produce SD SMP. The detailed hardware design has been previously discussed (Wu et al., 2011b). Four hot air guns (PHG 630 DCE, Bosch, Australia) blew hot air into the dryer chamber with controlled temperature and flow rate. Here, the dryer inlet and outlet temperature was kept at 190 °C and 48 °C, respectively. Collected powders were stored in a desiccator at room temperature for further characterization.

2.5. XPS measurement and surface composition calculation

X-ray photoelectron spectroscopy (XPS) analysis was carried out with an AXIS Ultra-DLD spectrometer (Kratos Analytical Ltd., UK) equipped with a monochromated Al Kα X-ray source and a hemispherical analyzer (fixed analyzer transmission mode). Raw milk was assumed to be only composed of lactose, protein (sodium caseinate) and anhydrous milk fat, and thus in 64.5, 34.8 and 0.7 wt% (dry basis), respectively. The elemental compositions for pure components were calculated by molecular formula (relative atomic%) as: lactose – C, 52.2; O, 47.8; protein – C, 65.0; O, 19.0; N, 16.0; fat – C, 89.1; O, 10.9. According to Kim et al., (2002), the concentrations of fat, protein and lactose, respectively on the sample surface (γ_{fat}, $\gamma_{protein}$ and $\gamma_{lactose}$) can be calculated by:

$$I^C_{Sample} = I^C_{lactose} \cdot \gamma_{lacose} + I^C_{protein} \cdot \gamma_{protein} + I^C_{fat} \cdot \gamma_{fat} \quad (1)$$

$$I^O_{Sample} = I^O_{lactose} \cdot \gamma_{lacose} + I^O_{protein} \cdot \gamma_{protein} + I^O_{fat} \cdot \gamma_{fat} \quad (2)$$

$$I^N_{sample} = I^N_{protein} \cdot \gamma_{protein} \quad (3)$$

where I^C_{Sample}, I^O_{Sample}, and I^N_{Sample} are the relative amounts of carbon, oxygen and nitrogen in the sample. I^C_{fat}, $I^C_{protein}$, and $I^C_{lactose}$ are relative amounts of carbon in fat, protein and lactose. I^O_{fat}, $I^O_{protein}$, and $I^O_{lactose}$ are relative amounts of oxygen in fat, protein and lactose, respectively. $I^N_{protein}$ is the amount of nitrogen in protein.

2.6. Particle morphology and size measurement

A field-emission scanning electron microscopy (FESEM, JEOL JSM-7001F, Japan) operated at 10 kV was used to obtain the information on particle size and morphology. As-prepared particles were coated with ~3 nm thick gold layer in argon environment at low pressure (~0.02 mbar). Particle size distribution information was acquired by analysing FESEM images containing over 1000 particles using ImageJ™.

2.7. Wettability and insolubility index (ISI) measurements

Wettability was measured for SMP from static wetting times according to Freudig et al., (1999) based on *GEA Niro Method No. A6a*. 100 ml of water at 20 °C was placed in a 350 ml baker. 1 g of powder was tipped from a weighting dish level with the baker rim. The time for all powders to visibly sink beneath the water surface was recorded as an indicator of powder wettability. The result for each sample was averaged from triplicate trials.

The ISI tests were performed using a scaled down variant (Rogers et al., 2012) with reference to *GEA Niro Method No. A3a* (IDF Standard 129, 2005). 5 g of sample was reconstituted in 50 ml water at 20 °C (10 g in 100 ml used in GEA method). The solution was stirred at 1500 rpm for 10 min and kept to rest for 10 min. Then the solution was filled into four 10 ml (50 ml in GEA method) centrifuge tubes and centrifuged at 164 rcf for 5 min. The supernatant was tipped off and replaced with water to disperse the sediment using a piece of wire. The solution was centrifuged again under the same condition. The amount of sediment was read in ml as the down-scaled ISI (ISI_{ds}). In some cases, the volume was too low to read (TLTR). The standard ISI needs scaling ISI_{ds} by 5 since 1/5 of centrifuged volume required in GEA method was used.