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Surface protein coverage and its implications on spray-drying of model sugar-rich foods: Solubility, powder production and characterisation

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ABSTRACT

We have investigated the amount of protein required to produce amorphous sugar powders through spray-drying. Pea protein isolate was used as a model plant protein and sodium caseinate was used as a model dairy protein. Powder recovery in a laboratory spray dryer was used as a measure of the ease of spray drying for a given formulation. More than 80% of amorphous sucrose and fructose was produced with the addition of sodium caseinate, while the pea protein isolate was able to produce only recoveries of less than 50% of amorphous sucrose. Sensitivity of low molecular weight surfactants has been demonstrated using both ionic (sodium stearoyl lactylate) and non-ionic (polysorbate-80) surfactants. Spray-dried powders were subjected to physico-chemical characterisation and dissolution experiments. The maximum solubility of all powders was obtained after 5 min of dissolution. The solubility of the sodium caseinate increased by 6–7% in the presence of fructose and low molecular weight surfactants. The solubility of the amorphous powders of sucrose–pea protein isolate was found to be lower than amorphous powders of sucrose–sodium caseinate and fructose–sodium caseinate. The addition of sucrose in water increased the solubility of the pea protein isolate from 16.84% to more than 83%. The non-ionic surfactant (Tween-80) has reduced the solubility of sucrose–pea protein isolate–Tween-80 powders significantly ($p < 0.05$) compared to those of sucrose–pea protein isolate–sodium stearoyl lactylate powders. The solubility of sucrose–sodium caseinate powders was comparable to that of pure sodium caseinate, indicating that addition of sucrose into 0.13% sodium caseinate does not have any significant effect on the solubility of this protein at this concentration.

1. Introduction

Spray-drying is one of the most commonly used methods in transforming a wide range of liquid food products into powder form, due to common availability of equipment, commercially viable processing costs and good final product quality and stability (Favaro-Trindade, Santana, Monterrey-Quintero, Trindade, & Netto, 2010). Spray-drying has many applications, particularly in the food, pharmaceutical and agrochemical industries (Adhikari, Howes, Shrestha, & Bhandari, 2007; Maa & Hsu, 1997; Maa, Nguyen, & Hsu, 1998; Vega, Goff, & Roos, 2005).

There are many naturally occurring products, such as fruits, vegetable extracts and honey, which have inherently high sugar contents. These are high value products and there is a growing interest to convert them into a powder in order to use them as ingredients (Bhandari, Datta, & Howes, 1997). Conversion of these sugar-rich foods into a particulate form is not easy through drying processes due to their low glass transition temperature (T_g) and strong hygroscopicity (Adhikari et al., 2007). These materials make soft particles with a very sticky surface and hence tend to stick to the dryer wall and agglomerate uncontrollably. Freeze drying of these materials is generally not successful as they absorb moisture very rapidly when the vacuum is broken.

The stickiness problem causes considerable economic loss and limits the application of drying techniques, such as spray-drying, for food and pharmaceutical materials (Boonyai, Bhandari, & Howes, 2004; Maa & Hsu, 1997; Maa et al., 1998). To minimise the stickiness problem, process and material science-based approaches are in place. In process-based modifications, stickiness could be avoided by keeping the outlet temperature of the air below 50 °C or even at ambient temperature. However, the powders obtained at such low outlet temperature usually have high residual moisture contents and water activity values which negatively impact their subsequent storage. The material-science based approach also has its own limitations. Large amounts (often >35%) of drying additives, such as maltodextrins, are required to convert fruit juices such as blackcurrant, apricot and raspberry into a powder form (Gabas, Telis, Sobral, & Telis-Romero, 2007; Righetto & Netto, 2005; Tonon et al., 2009). Addition of such large amounts

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of these carriers alters the resultant powder quality and risks consumer disapproval.

An alternative and novel way to minimise the stickiness problem is to modify the surface properties of the droplets/particles with small amounts of proteins (Adhikari, Howes, Bhandari, & Langrish, 2009a; Jayasundera, Adhikari, Adhikari, & Aldred, 2010, 2011a). However, spray-drying can cause thermal denaturation of the proteins (Anandharamakrishnan, Rielly, & Stapley, 2008). The extent of protein aggregation and/or inactivation during and after spray-drying can be minimised by incorporating some of the low molecular weight surfactants and stabilising sugars (Lee, 2002). Since low molecular weight surfactants (LMS) are smaller in size compared to proteins, the former are kinetically advantaged to occupy the surface of a droplet immediately after atomisation (van Aken, 2003).

Although low molecular weight surfactants generally stabilise proteins, some of these surfactants are known to cause undesired effects on the proteins leading to their destabilisation. Ionic low molecular weight surfactants, such as sodium dodecyl sulphate (SDS), have been known as effective protein denaturants (Randolph & Jones, 2002). In contrast, low molecular weight surfactants, such as sorbitans, are used as stabilising agents in protein formulations (Randolph & Jones, 2002). However, non-ionic low molecular weight surfactants can also have an opposite effect on proteins. In the cases where a non-ionic surfactant destabilises the conformation of a protein, this effect may compete against the solubilising effect of surfactant binding (Randolph & Jones, 2002). For example, in the case of hydrophobic lipase from *Humicola lanuginose,* Tween-20 addition was observed to cause the formation of insoluble non-native aggregates (Kreilgaard & Frokjaer, 1999).

In our previous study we investigated the effect of low molecular weight surfactants and proteins on spray-drying of sugar rich foods and found that their presence greatly reduces the surface coverage of proteins (Jayasundera et al., 2010, 2011a). This means that the protein surface coverage of the droplets was minimised. This can potentially lead to a greater preservation of proteins as the proteins at the air-water interface tend to unfold (Maa & Hsu, 1997; Maa et al., 1998). However, no studies have reported on the effect of low molecular weight surfactants and proteins on spray-drying of sugar-rich foods with regard to dissolution. Dissolution of powdered ingredients is of particular importance to manufacturers and consumers as a benchmark of functionality (Fang, Selomulya, & Chen, 2008). Food powders, when used as ingredients, must be able to provide good solubility to be useful and functional (Morr et al., 1985). The solubility is the final step of powder dissolution and is considered as the key determinant of the overall reconstitution quality.

The aim of this study was to investigate the effect of low molecular weight surfactants and proteins on spray-drying of sugar-rich foods with regard to solubility, powder production and characterisation.

2. Materials and methods

2.1. Materials

Fructose and sucrose, both with 99.5% purity, were purchased from ADM, Australia and Sigma–Aldrich, Australia, respectively. They were used as model sugar-rich foods. Sodium caseinate (NaCas) (MG 2972, MG Nutritionals, Australia) with a protein content of 92.9% (manufacturer's data sheet) and pea protein isolate (PPI) (Myopure, Australia) with a protein content of 90% (manufacturer's data sheet) were used as model proteins. Two food grade low molecular weight surfactants, sodium stearoyl lactylate (SSL) and Polysorbate-80 (Tween-80), were used as model surfactants. The former (Grindsted® SSL P 60 Veg) was purchased from Danisco, Denmark, while the latter was purchased from Sigma–Aldrich, Australia. SSL is an ionic surfactant which has a molecular weight of 451.6 g/mol and has a hydrophile-lipophile balance (HLB) value of 22, while Tween-80 is a non-ionic surfactant with a larger molecular weight (1310 g/mol) and a HLB value of 15. Both the surfactants are water soluble and are suitable for oil-in-water emulsions (McClements, 2005). All the above materials were used as received.

2.2. Methods

2.2.1. Solution preparation

The sugar–protein solutions were prepared by heating the solution to 45 ± 5 °C and gently agitating with a magnetic stirrer. This range of temperature is well below the denaturation temperature of the proteins used and has no negative effect on the solubility of the samples used. The sugar: protein solid mass ratios were 70:30, 99:1 and 99.5:0.5 for fructose: NaCas, sucrose: PPI and sucrose: NaCas, respectively. This fructose: NaCas ratio was used since higher sugar contents either resulted in no powder or insufficient powder for characterisation. The total solid content of the feed solution was 25% (w/w) in all the cases. This translates into an initial bulk protein concentration in fructose–NaCas, sucrose–PPI and sucrose–NaCas solutions of 7.89%, 0.26% and 0.13% (protein/total solution basis), respectively. While making the solutions, both the pre-weighed sugar and the protein were dry mixed thoroughly before addition of water. Solution batches of 300 ml were prepared. The inherent moisture content of both crystalline fructose and sucrose was taken as zero, while it was determined and compensated for in the case of NaCas and PPI. Solutions of sugar–protein-SSL and sugar–protein–Tween-80 were prepared by adding 0.01% and 0.05% w/w of each surfactant to the sugar–protein solutions. The solutions were heated to 45 ± 5 °C to ensure that all solids were completely dissolved. The solutions were then tested for dynamic surface tension and subsequently spray-dried.

2.2.2. Powder production

Spray-drying of solutions was carried out on a bench-top spray-dryer (Buchi B-290, Buchi, Switzerland) with a water evaporating capacity of 2 l/h. A twin-fluid nozzle, that used compressed air as atomising fluid, was used to atomise the solution into fine droplets. The inlet and outlet temperatures were maintained at 165 and 65 °C, respectively. The air flow rate was maintained at 36 m^3/h. The powders were collected from the cyclone and the cylindrical parts of the dryer chamber by lightly sweeping the chamber wall as proposed by Bhandari et al. (1997). The yield was calculated as the ratio of the mass of solids collected after spray-drying to the amount of solids in the feed solutions.

2.2.3. Dissolution kinetics of spray-dried powders

Dissolution kinetics was carried out by adding 2 g of spray-dried sucrose-PPI, sucrose-PPI-Tween-80 (0.05%) and sucrose–PPI–SSL (0.05%) powder samples individually to 50 ml of MilliQ water at 26 °C (Favaro-Trindade et al., 2010). The mixtures were mechanically stirred using a magnetic stirrer at 892 rpm for different time intervals (1, 3, 5, 10, 30 and 60 min). The dissolution kinetics was carried out on PPI based sugar powders, as PPI has a lower solubility than NaCas, to determine the effect of sugar on the solubility of protein (Jayasundera et al., 2011a). Pure protein samples were also subjected to dissolution kinetics for comparison. Digital images were recorded using a CCD camera (Sony, SSC-M370CE, Sony Company, Japan) magnified by a stereomicroscope (Stemi 2000, Carl Zeiss Jena GMbH, Germany) at different time intervals until complete dissolution was obtained.