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The effect of low molecular weight surfactants and proteins on surface stickiness of sucrose during powder formation through spray drying

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ABSTRACT

The effect of competitive surface migration of proteins and low molecular weight surfactants (LMS) on the powder recovery in spray drying of highly sticky sugar-rich food has been studied. Sucrose was chosen as a model sugar-rich food because it cannot be easily converted into a pure amorphous powder through spray drying. Sodium caseinate (Na–C) and hydrolyzed whey protein isolate (WPI) were used as model proteins. Polysorbate 80 (Tween-80) and sodium dodecyl sulfate (Na-DS) were used as model non-ionic and ionic LMS.

A sucrose solution was spray dried without any additives to establish a base case. Following this, spray drying trials of sucrose–protein solutions were conducted. The sucrose: protein ratio was maintained at 99.5:0.5 and 99.0:1.0. Finally, 0.05% of Tween-80 and Na-DS, on a nominal feed basis, were individually added to the solutions and spray dried. The solid concentration of all of the feed solutions was set at 25% and the inlet and outlet temperatures were maintained at 170 °C and 70 °C, respectively. Powder recovery was determined using a standard procedure and taken as an indicator of the surface stickiness. Coverage of the particle surface by the proteins was determined through elemental surface analysis and a nitrogen balance. It was found that in the absence of LMS, the proteins covered up to 55% of the particle surface and increased the powder recovery to between 84% and 85%. Formation of a glassy protein-rich film acts to reduce the surface stickiness of sucrose droplets. However, when LMS was added to the sucrose–protein solutions, the recovery dropped to zero in the case of Tween-80. In the case of Na-DS the recoveries ranged to 39% and 68%. At these recoveries 83% and 59% of the protein, respectively, was displaced from the surface. This drastic effect of surfactant types on the powder recovery is explained using the Orogenic Displacement model.

1. Introduction

The use of spray drying to produce powders from formulations containing low molecular weight sugars is limited because of their inherent stickiness. This stickiness results in depositions on the dryer wall, roof, conical section and conveying ducts (Bhandari et al., 1997a; Langrish, 2007). In the case of honey, which is a typical sugar-rich material, it is not yet possible to convert it into a powder without the addition of a significant amount of high molecular weight carrier material such as maltodextrin. The sticky food powders which are recovered are highly hygroscopic and tend to easily cake or lump and are very difficult to store. They require packaging with very high water barrier properties. This secondary powder stickiness can be triggered due to pressure and temperature changes or cycling during storage and transportation (Adhikari et al., 2001).

The inherent sticky behaviour of sugar and acid-rich foods requires special processing and material-centric intervention to allow economically viable production. Process modifications include the use of low temperature and low humidity air, wall cooling or the introduction of cold air to the bottom of the dryer. Frequent mechanical sweeping is another modification. Material modifications at present include the addition of drying carriers such as maltodextrins, gums and high molecular food hydrocolloids.

The efficacy of low dextrose-equivalent (DE) maltodextrins, as drying-aids, is due to their rapid film or shell forming property and the relatively low moisture diffusivity of these films (Adhikari et al., 2003). The earlier the film is formed in the drying process, the better will be the maltodextrin's efficacy as a drying-aid. In this context, it has also been found that proteins such as whey protein isolates (WPI) and sodium caseinates form smooth and non-sticky films or shells much earlier than the maltodextrins and that the

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recovery of powders is much higher when a small amount of proteins are added to the solution being spray dried (Adhikari et al., 2007a). This is an indication that proteins can act as a very effective drying-aid. This argument can also be supported by the much lower droplet-to-probe tensile pressure of lactose-WPI mixture solutions compared to that of the sugar solutions (Adhikari et al., 2007b).

In pharmaceutical science, the competitive adsorption of proteins and LMS has been extensively researched. The application of the LMS in therapeutic drugs is required to limit or prevent the exposure of active-protein ingredients to the air–water interface (Maa and Hsu, 1997; Maa et al., 1998). Since both proteins and LMS provide enhanced emulsion stability, a series of studies were undertaken to understand the mechanisms by which the LMS displace proteins at air-water and fat–water interfaces (Williams and Prins, 1996; Dickinson, 1999; Mackie et al., 1999, 2000; Gunning et al., 2004; Williams and Prins, 1996; Rouimi et al., 2005).

Experiments conducted in our laboratories have shown that the preferential migration of proteins driven by their surface activity allows the generation of highly surface engineered powders of sugar and acid-rich foods. In a pilot scale spray dryer, the use of sodium caseinate and WPI led to the excellent recovery of 85%–90% of amorphous sucrose powder when a mere 0.125% of these proteins are introduced in the solution (Adhikari et al., in press). This compared with the >16% of maltodextrin (DE6) required to obtain the same extent of recovery of the sucrose powder under similar drying conditions (Truong et al., 2005).

It is known that LMS compete with protein for the air–water interface (Pugnaloni et al., 2004; van Aken, 2003; Rouimi et al., 2005; Mackie et al., 2000). With a smaller size they are advantaged kinetically to occupy the surface as proteins have relatively lower diffusivities (van Aken, 2003). It is of practical significance to investigate the implication of the presence of trace amount of LMS along with proteins in the surface stickiness of sugar-rich foods. This is because it has been observed that there is a presence of trace amount of LMS in industrially obtained sugar samples (Adhikari et al., 2007b).

Hence the aim this project was to investigate the competitive migration of protein and LMS to the surface of powders of a model sugar. The project also studied the implication of this competitive migration to the stickiness through the recovery of those powders in pilot scale spray drying.

2. Materials and methods

2.1. Materials

Sucrose with 99.5% purity (Sigma–Aldrich, Australia) was used as model sugar-rich food. A reagent grade Sodium dodecyl Sulfate (Na-DS) with 98.5% purity and Polysorbate-80 (Tween-80, 10% solution) from Sigma–Aldrich, Australia were used as received. Sodium caseinate (ALATAL^{TM} 180) and hydrolyzed whey protein isolate (ALATAL^{TM} 817) were obtained, courtesy of NZMP, New Zealand and used as received.

2.2. Methods

2.2.1. Solution preparation

The protein–sugar solution was prepared by heating up the solution to 50 °C and agitating with the aid of a magnetic stirrer. The protein was first dissolved by adding small amounts of the pre-weighed sample at a time under constant stirring to avoid clumping of the powders. The stirring was mild in order to avoid air entrainment. Once the protein was dissolved, sucrose was added. The sucrose to protein solid ratio was maintained at 99.5:0.5 and 99.0:1.0 on dry solids basis. The total solids fraction in the feed solution was fixed at 25% by weight. Thus, the nominal feed concentration of the protein in the solution was either 0.25% or 0.125%. One kilogram solution bathes were prepared. The inherent moisture content in the protein samples was determined and compensated for. The moisture content of the crystalline sucrose was taken to be zero. The solution matrix is presented in Table 1. The protein–sugar-LMS solutions were prepared by adding 0.05% (nominal feed concentration) Na-DS or Tween-80 to the solution under sufficient stirring. Solutions were prepared with 250 mL water along with the protein and surfactant, if any, first on a hot plate maintained at 45 °C, to ensure that all solids will successfully dissolved, before adding the sucrose and the remaining water. All the moisture contents, reported in the ensuing sections are on a weight/weight basis.

2.2.2. Moisture determination

The moisture content of the powder was determined through vacuum drying (Thermoline Scientific, Australia) at 70 °C and 500 mbar for 24 h followed by cooling the samples to the room temperature in desiccators in the presence of an excess amount of silica gel. Duplicate or triplicate tests were carried out.

2.2.3. Powder production

Powder from both the protein–sugar and protein–sugar-LMS solutions were produced using a pilot scale spray dryer (SL20, Saurin Company, Victoria, Australia) with a water evaporating capacity of 2 kg/hr. The inlet and outlet temperatures were maintained at 170 °C and 70 °C, respectively. The powders were collected from the cyclone, and in the case of sweeps, they were collected by lightly sweeping the inner dryer wall.

2.2.4. Water activity

Water activity of the powder samples was determined using AquaLab 3TE Series (Decagon, USA) water activity meter. The temperature was maintained at 25 ± 0.5 °C during the tests. Duplicate or triplicate tests were carried out.

2.2.5. X-ray photoelectron spectroscopy

X-ray photoelectron spectroscopy (XPS) or electron spectroscopy for chemical analysis (ESCA) is a well established method employed for direct measurement of surface elemental composition of food powders (Faldt and Bergenstahl, 1996; Kim et al., 2003). A detailed description of the use of ESCA as a method to measure the surface composition of dairy based food powders can be obtained from various sources (Faldt et al., 1993; Kim et al., 2002; Nijdam and Langrish, 2006).

Firstly, ESCA measurements for sucrose, sodium caseinate, WPI, Na-DS and Tween-80 were carried out to determine the surface composition of these materials. It is assumed that the surface elemental composition of pure materials is the same as its bulk elemental composition. Subsequently, the surface elemental composition of all the spray dried powders was determined. Prior to subjecting to the ESCA test, the samples were outgassed for 72 h. The ESCA was performed on a Kratos AXIS Ultra with a 150 W monochromatic A1 X-ray source. Each analysis started with a survey scan from 0 to 1200 eV with a residence time of 100 ms, pass energy of 160 eV at steps of 1 eV, with a 1 sweep. For the high resolution analysis, the number of sweeps was increased, the pass energy was lowered to 20 eV, at steps of 50 meV and the residence time was increased to 250 ms. Data were acquired using a Kratos Axis ULTRA X-ray spectrometer, incorporating a 165 mm hemispherical electron energy analyzer. The incident radiation was Monochromatic A1 X-rays (1486.6 eV) at 225 W (15 kV, 15 mA). Survey (wide) scans were at analyzer pass energy of 160 eV. Base