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Flavor release from spray-dried amorphous matrix: Effect of lactose content and water plasticization

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ABSTRACT

Glass-forming carbohydrates are widely used as matrix for encapsulation of nutrients and bioactive compounds. In this study, encapsulation systems with lactose/whey protein isolate (WPI) mixtures (4:1, 1:1, and 1:4), or WPI as wall materials and ethyl butyrate as core material were prepared by spray drying. The effects of lactose content and water plasticization on encapsulation efficiency and flavor release were investigated. Wall material consisting of lactose/WPI (4:1) mixture had significantly ($P < 0.05$) higher encapsulation efficiency. The flavor retention in powders did not have significant decrease with equilibration at 0.33 a_w, while it was dramatically decreased at 0.54 a_w and 0.65 a_w as a result of lactose crystallisation. Mechanical property study showed that the molecular mobility and free volume of encapsulation systems with higher lactose content increased more significantly with increasing water content, which accelerated the diffusion of flavor molecules. Those results may use in the assessment of protection and release characteristics of flavor components in formulated systems.

1. Introduction

In food industry, there is a growing interest for encapsulation technologies, which are designed to protect the encapsulated materials and to allow controlled release. Many volatile compounds are encapsulated in solid carriers to increase their protection, reduce evaporation, and promote easier handling (Bae & Lee, 2008; Baranauskiene, Bylaite, Zukauskaite, & Venskutonis, 2007; Beristain, Vazquez, Garcia, & Vernon-Carter, 1996; Chin et al., 2010; Goubet, Le Quere, & Voilley, 1998; Jafari, He, & Bhandari, 2007; Kaushik & Roos, 2007). Since flavoring components are prone to loss by evaporation, oxidation or ingredient interactions, the encapsulation of flavor ingredients is among the most important applications in the food industry (Baranauskiene et al., 2007). Encapsulation matrix for flavor compounds can be selected from a wide variety of polymers, depending on the core materials and desired characteristics of the microcapsule. Typical wall materials include proteins (sodium caseinate, whey proteins, soy proteins and gelatin) and hydrocolloids (modified starch and gum Arabic). Among proteins, whey proteins have been shown to be an excellent encapsulating agent for microencapsulation of oils/fats and volatiles (Bae & Lee, 2008; Charve & Reineccius, 2009; Moreau & Rosenberg, 1996; Rodea-González et al., 2012; Rosenberg & Sheu, 1996; Sheu & Rosenberg, 1995). Moreover, although a variety of methods have been proposed to encapsulate flavors, spray-drying and extrusion are still the most common techniques. It has been shown that wall systems of spray-dried microcapsules consisting of whey proteins provide effective protection against core oxidation (Bylaitë, Venskutonis, & Maþdþierienë, 2001; Kim & Morr, 1996).

Previous studies have indicated that the addition of small quantities of specific low molecular weight compounds to matrices can improve the storage stability of bioactive (Roussenova, Murith, Alam, & Ubbink, 2010). These low molecular weight compounds act as packing enhancer, leading to a reduction of the molecular hole size in the glassy state, which could explain the improved glassy-state barrier properties and encapsulation performance. According to Rosenberg and Sheu (1996), the addition of lactose into WPI-based wall systems improves the volatile retention during spray drying and limits the core extractability. This is due to lactose in its amorphous state acting as a hydrophilic sealant that significantly limits diffusion of the hydrophobic core through the wall and thus leads to high microencapsulation efficiency values. Furthermore, amorphous carbohydrates in the glassy state are widely used as matrix for the encapsulation and stabilization of nutrients, pharmaceutics, and other bioactive compounds (Drusch, Serfert, Van Den Heuvel, & Schwarz, 2006; Fäldt & Bergenståhl, 1996a, 1996b; Kaushik & Roos, 2008; Levi & Karel, 1995; Lim, Griffin, & Roos, 2014; Lim & Roos, 2015; Naknean & Meenune, 2010; Zhou & Roos, 2012). In these applications, the glass transition temperature of amorphous matrix has been used as the central physical parameter for the optimization of processing conditions and storage stability (Soottitantawat et al., 2004; Townrow, Roussenova, Giardiello, Alam, & Ubbink, 2010). When the temperature increases from below to above the glass transition

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temperature, many of the physical properties of the amorphous matrix show a rapid change, including increases in the free volume, molecular mobility, and dielectric coefficient. Large changes in viscoelastic properties of amorphous powders also occur above the glass transition temperatures (Jones, 1999; Kasapis, 2001; Royall et al., 2005). Once an encapsulation matrix has gone through the glass transition and entered the metastable rubbery state, the rates of deteriorative reactions and diffusion of the flavor from the particle matrix may increase dramatically (Risch & Reineccius, 1995; Whorton, 1995).

In addition, since the presence of high molecular weight protein or carbohydrate in the amorphous lactose-based systems could increase diffusion distance (Potes, Kerry, & Roos, 2012; Silalai & Roos, 2010, 2011), the ratios of low molecular weight compounds in encapsulation systems could affect the properties of whey protein-based systems. Furthermore, water is a highly efficient plasticizer of carbohydrates and the glass transition temperature of an amorphous carbohydrate matrix decreases strongly with increasing water content (Jouppila, Kansikas, & Roos, 1997; Jouppila & Roos, 1994; Partanen et al., 2008; Velasco, Holgado, Dobarganes, & Márquez-Ruiz, 2009; Zhou & Roos, 2012). According to Kilburn et al. (2004), the diffusional mobility of small molecules such as water, gases, or volatile organic compounds increases rapidly with increasing water content in both the glassy and rubbery states. The water content and water activity of the encapsulation matrix are key parameters for understanding the physical behavior and barrier properties of amorphous carbohydrates (Ubbink, 2003). Levi and Karel (1995) stated that water resulted in a lowering of T_g and acceleration of relaxation processes, which increased the release of 1-n-propanol in amorphous carbohydrate glasses. However, there is no further study about the roles of lactose content and water plasticization on the encapsulation properties of wall systems, and especially the relationship between the mechanical properties of amorphous matrix and their barrier properties.

Therefore, the aim of present study was to investigate the effect of amorphous lactose content and water plasticization on encapsulation properties and flavor release in dairy-based powders. How the physical and mechanical properties of wall systems affect flavor release was also investigated.

2. Materials and methods

2.1. Materials

α-Lactose monohydrate (>99% purity, Batch No. F051621) was kindly donated by Arla Foods Ingredients (Sønderhøj 10–12, 8260 Viby J, Denmark). Whey protein isolate (WPI), containing 71% β-lactoglobulin and 12% α-lactalbumin, was obtained from Davisco Food International (Le Sueur, MN, USA). Ethyl butyrate (EB), hydroxylamine hydrochloride, n-hexane and aluminum oxide calcined powder (≥99% purity) were purchased from Sigma-Aldrich (St. Louis, MO, USA).

2.2. Emulsion preparation

Wall material solutions were prepared in deionized water at 40 °C for 2 h and then kept overnight on the magnetic stirrers to ensure complete hydration at room temperature (23–25 °C). Total solids concentration of wall materials was 25% (w/w), which composed of lactose/WPI mixtures or WPI. The mass ratios of 4:1, 1:1, and 1:4 for lactose/WPI mixtures were used in this study. They were defined as S1 (lactose/WPI (4:1) mixture), S2 (lactose/WPI (1:1) mixture), S3 (lactose/WPI (1:4) mixture), and S4 (WPI) according to the wall materials. Then ethyl butyrate was emulsified into the wall solutions at a proportion of 20% (w/w of wall solids). All emulsions (oil in water) were prepared in two stages. Coarse emulsion was prepared using an ULTRA-TURRAX (IKA, Staufen, Germany) operated at 10,000 rpm for 1 min. Then the coarse emulsion was further homogenized using a M110-EH Microfluidizer with a 75 μm Y-type ceramic interaction chamber (Microfluidics International Corp., Newton, MA, USA) at 50 MPa for three successive homogenization steps. The homogenization process by microfluidizer was performed at room temperature with running cold water.

2.3. Spray drying

All emulsions were spray-dried by an ANHYDRO single stage spray dryer with a centrifugal atomizer (Copenhagen, Denmark) at the Teagasc Food Research Centre, Moorepark, Fermoy, Co. Cork, Ireland. The inlet air temperature was around 170 ± 2 °C and the outlet temperature around 90 ± 2 °C. Spray-dried solids were kept immediately in desiccators over P_2O_5 at room temperature. Each analysis was carried out within 2 months after spray drying.

2.4. Physical properties

Loose bulk density of powders was measured using a Jolting volumeter (Funke Gerber, Berlin, Gerrmany). Particle density (ρ_p) was measured using a Gas Pycnometer (Accupyc II 1340 Gas Pycnometer, Micromeritics Instrument Corporation, USA). Particle size distribution and specific surface area (SSA) of powder particles were determined by laser light scattering using a Malvern Mastersizer 3000 (Malvern Instruments Ltd., Worcestershire, UK).

2.5. Chemical analysis

Protein content in powders was determined using a FP 628 Nitrogen Determinator (LECO Corporation, Lakeview Avenue, St. Joseph, Michigan, USA). Lactose content was determined using an Automatic Polarimeter (Autopol 1, Rudolph Research Analytical, Hackettstown, NJ, USA). Water content was determined using a HR83 Halogen Moisture Analyzer (Mettler Toledo International Inc., Im Langacher Greifensee, Switzerland). Water activity was measured using a water activity meter (Novasina LabMaster AW, Novatron Scientific Ltd., West Susses, UK).

In order to determine encapsulated EB content in powders, EB was extracted from powders according to the method of Kaushik and Roos (2007). 1 g of powder was dissolved in 20 mL of deionized water in glass bottles and 10 mL of hexane was added, followed by mixing with a rota mixer for 1 min. Encapsulated EB was extracted with hexane by heating the samples in glass bottles at 45 °C in a water bath for 15 min with intermittent mixing. All glass bottles were kept closed with caps and sealed with sealing film during extraction. Then the bottles were cooled to room temperature and hexane was separated from aqueous phase by centrifugation at 4000 rpm for 20 min. The amount of EB present in hexane was quantified by the method of Sheu and Rosenberg (1995). 0.5 mL of centrifuged hexane was mixed with 4.5 mL 50% ethanol solution. The colour of the solution containing EB was developed and then measured absorbance with a spectrophotometer at 525 nm. The amount of EB was determined from calibration standard curves. More details about the method of EB determination were presented in the work of Sheu and Rosenberg (1995). The EB content of each sample was carried out in triplicate.

The ratio of encapsulated EB content in powders after spray drying to initial EB content added into the emulsion was defined as encapsulation efficiency (%). The ratio of encapsulated EB content in powders during equilibration at different relative humidity (RH) to encapsulated EB content after spray drying was defined as flavor retention (%).

2.6. Water sorption and flavor release from powders at different RHs

Approximately 1 g of powders was weighed into small glass vials (25 mL). All vials were equilibrated for 216 h in evacuated desiccators over saturated salt solutions of $MgCl_2$, K_2CO_3, $Mg(NO_3)_2$, and $NaNO_2$, giving RH of 33%, 44%, 54%, and 65%, respectively. All desiccators were