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Heterogeneous particle structure formation during post-crystallization of spray-dried powder

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ABSTRACT

The formation of heterogeneous particle structure in skim milk powder has been investigated in a post-crystallization facility using experimental and a mathematical model. Various processing conditions were used to produce these heterogeneous structures. The experimental process parameters were used as initial and boundary conditions for the model. The modelled data agreed well with the experimental data. The experimental and modelling results show that the powder processed at high water activity ($a_w = 0.7$) with low initial moisture content ($\bar{X}_0 = 0.01$ kg/kg) developed a crystalline surface layer while the core of the particle remained amorphous. This structure is referred to as an egg-shell structure. The powder that was processed at low water activity ($a_w = 0.1$) with high initial moisture content ($\bar{X}_0 = 0.2$ kg/kg) developed a crystalline core while the surface of the particle remained amorphous. This structure is referred to as an egg-yolk structure. Understanding the dependency of particle microstructures on the processing conditions could be useful when developing procedures to control the drying equipment because the particle microstructure affects the physicochemical properties of the powder and potential applications and behaviour of the powder.

Introduction

Spray drying is a significant component of milk powder production. During this stage, most of the powder's properties are determined. In most spray-dried milk powders, lactose is present in an amorphous state (Bhandari, Datta, & Howes, 1997; Kim, Chen, & Pearce, 2009). Amorphous lactose is thermodynamically unstable and hygroscopic (i.e., absorbing moisture from the environment), which results in plasticization and a decrease in the glass-transition temperature (Jouppila & Roos, 1994). Researchers have recommended that the amorphous-lactose fraction could be treated in a crystallization facility after spray drying to crystallize lactose-containing powders and thus limit the caking tendency of the powder (Nijdam, Ibach, & Kind, 2008; Yazdanpanah & Langrish, 2011a). Yazdanpanah and Langrish (2011b) demonstrated the formation of semi-crystalline powders inspecific, controlled conditions. The heterogeneous particle structure, often referred to as an egg-shell structure, has a profile where the particle has a crystalline surface layer and amorphous core (Lekhal, Glasser, & Khinast, 2001). The post-crystallization process in a fluidized-bed dryer depends on the processing conditions, such as air temperature, relative humidity, time, and the initial moisture content of the spray-dried powders.

To achieve a certain moisture content of the final powders, the conventional industrial practice is to control the spray dryer and fluidized-bed dryer's conditions (Písecký, 1997). Changes in ambient conditions, such as daily variations of relative humidity, can affect the performance of the spray drying unit and, as a result, change the final moisture content of the spray-dried powders. Therefore, to eliminate the excess moisture content of the products, higher processing temperatures or longer processing times in the fluidized-bed dryer are used (Písecký, 1997). However, although the final moisture content of different batches might be similar, the internal structure of the powder particles processed at different conditions has not been studied.

This work investigates the partial crystallization of milk powder in a fluidized-bed system and demonstrates that different heterogeneous particle structures occur following different processing conditions. A distributed-parameter model has been used to investigate the effect of processing conditions on the formation of heterogeneous particle structures.

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Nomenclature

A_p	external surface area of the particle (m^2)
$a_{as,i}$	mass fractions of amorphous lactose in layer i
a_w	water activity
$b_{cs,i}$	mass fractions of crystalline lactose in layer i
C	GAB constant
Cp_v	specific heat capacity of water vapour (J/(kg K))
ΔCp_j	Change in heat capacity of component j at Tg_j (J/(kg K))
D_{as}	water vapour diffusivity in amorphous lactose (m^2/s)
D_{av}	average diffusivity for the mixture (m^2/s)
D_{cs}	water vapour diffusivity in crystalline lactose (m^2/s)
D_{pr}	water vapour diffusivity in protein (m^2/s)
D_{wa}	water vapour diffusion in air (m^2/s)
d_{part}	particle diameter (m)
$j_{(1)}$	flux of water sorbing on to the particle surface (kg/s)
j_i	flux of water into the layer i (kg/s)
j_{i+1}	flux of water out of the layer i (kg/s)
K_j	GAB constant
k_{cr}	rate of crystallization in the particular local conditions of T_i–Tg_i (s^{-1})
k_g	rate of crystallization at the glass-transition temperature (s^{-1})
k_m	external mass transfer coefficient (s/m)
Mw_{air}	molecular weight of air (kg/mol)
$M_{as,i}$	mass of amorphous lactose in layer i (kg)
$M_{as,i}$	mass of protein in layer i (kg)
Mw_w	molecular weight of water (kg/mol)
n	number of layers
P_{air}	absolute pressure of air (Pa)
pv_{air}	Partial water vapour pressure in air according to the absolute humidity of air (Pa)
pv_i	Partial vapour pressure of water in layer i (Pa)
pvs_i	saturated vapour pressure (Pa)
R_i	radius of the nth layer from the centre of the particle (m)
$\dot{S}$	moisture content source term (kg/(kg s))
Sh	Sherwood number from Ranz–Marshall equation
T_0	initial temperature of the particle (K)
T_{air}	absolute temperature of air (K)
Tg_i	glass-transition temperature of the materials in layer i (K)
Tg_j	glass-transition temperature of component j (K)
T_i	temperature of the layer i (K)
$\dot{X}_0$	initial moisture content of the particle (kg water/kg dry solid)
X_0	monolayer moisture value of the component (kg water/kg dry mass of the component)
$X_{as,i}$	moisture contents of amorphous lactose (from the moisture sorption equation) in layer i (kg water/kg dry mass of the component)
$X_{cs,i}$	moisture contents of crystalline lactose (from the moisture sorption equation) in layer i (kg water/kg dry mass of the component)
X_{pr}	moisture contents of protein (from the moisture sorption equation) in layer i (kg water/kg dry mass of the component)
$\overline{X}_i$	average moisture content of the layer i (kg/kg)
X_j	moisture content of the component j (kg water/kg dry mass of the component)
Δz_{i-1}	distance between the two node points where the state variables are stored (m)

Greek symbols

ρ_{air}	density of air at the air temperature (kg/m^3)
ρ_m	density of material in the layer (kg/m^3)
θ_{cr}	crystallization time at any point in time (s)
θ_g	time for crystallization at the glass-transition temperature (s)

Experimental study

Materials and methods

Spray dryer

Fresh skim milk (0.1 g fat per 100 mL obtained from local suppliers, Coles, Australia) was fed into a Buchi B-290 spray dryer (BuchiLabortechnik, Switzerland) with an inlet air temperature of 140 °C and an outlet air temperature of 67 °C. The powders were collected in a vessel at the bottom of a cyclone and immediately reconstituted with distilled water at 25 °C to a concentration of 35 wt%. This reconstituted solution was used as the input again for a Buchi B-290 spray dryer with two different settings to produce powders with different moisture contents. To produce a dry powder, an inlet air temperature of 160 °C and an outlet air temperature of 82 °C were used. The moisture content of the resultant dried powder was measured as 1 wt%. To produce relatively moist powder, an inlet air temperature of 140 °C and an outlet air temperature of 61 °C were used. The moisture content of the resultant powder was measured as 20 wt%. The oven-drying method, which was used for measuring the moisture content of the powders, is described elsewhere (Yazdanpanah & Langrish, 2011c). The final powders were immediately fed to the fluidized-bed dryers for the post-crystallization experiments.

Islam, Langrish, and Chiou (2010) suggested a new technique called "Humid Loop" to spray dry lactose, which uses highly humid air to obtain highly crystalline lactose from spray dryers. In the present study, which used a conventional setup, the slight variations in drying air temperature and humidity are not expected to alter the crystallinity of the powders (Yazdanpanah & Langrish, 2011a).

Fluidized-bed dryer

Details of the fluidized-bed dryer apparatus used in this study have been published elsewhere (Yazdanpanah & Langrish, 2011b,c). The fluidization air has a temperature range between 25 and 100 °C, a relative humidity between 10% and 95% and an air velocity (within the fluidization chamber) between 0.1 and 5 m/s. For the present study, the air velocity was held constant at 0.3 m/s. The powders residence time for the fluidization was 20 min and during this time the material was well agitated by continuously vibrating the bed.

Five thermocouples (Pyrosales, Australia) were connected to the system to measure the temperatures of the humidifier, air heater, inlet fluidization air and outlet air (dry-bulb and wet-bulb). The dry- and wet-bulb temperatures of the fluidization air were used to calculate the relative humidity (water activity) for the experiments, and the dry-bulb temperatures were recorded as the processing temperature. The process parameters (temperature, humidity, and moisture content) are reported in the Process conditions section.

X-ray diffraction

X-ray diffraction (XRD) was used to investigate the bulk crystallinity of raw and processed powders. XRD was performed on a Siemens D5000 diffractometer operated at 40 kV and 30 mA with