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THE REACTION ENGINEERING APPROACH TO ESTIMATE SURFACE PROPERTIES OF AQUEOUS DROPLETS DURING CONVECTIVE DRYING

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Abstract: Surface properties of the droplet/particle under spray drying conditions are known to influence wall deposition as well as product characteristics such as stickiness and lump formation. Literature shows that the droplet's surface properties such as surface moisture content, surface temperature may be estimated using distributed parameter models such as a diffusion model and a moving boundary model. In this paper, a new simple procedure is developed to estimate the time-dependent surface moisture content of aqueous droplets during hot air drying. This method is established by extending the reaction engineering approach (REA)-based drying kinetics model that is a lumped-parameter model. In this work, surface moisture contents and surface glass transition temperatures are estimated for single suspended aqueous maltodextrin droplets, which were dried under constant air temperature and humidity conditions. The method developed here can readily be used for estimating surface properties of different materials under industrial spray drying conditions.

Keywords: droplet drying, surface property, moisture content, glass transition temperature, reaction engineering approach

INTRODUCTION

Convective drying of aqueous materials using hot drying gas is a common process for producing dried products. Spray drying, fluidized-bed drying, tunnel drying are typical examples of widely used convection-based industrial drying operations. Characterization of aqueous materials (a droplet or a thin slab form) during processing is essential to control product properties and to avoid unwanted product loss, for instance, due to deposition of the product on the drier wall during spray drying and fluidized-bed drying. Most of the previous research work aimed at predicting the average behavior of products during drying which provided important product-related information such as particle size, particle density, moisture content, temperature, solubility index, nutrient contents as well as drying time and residence time. This information helps to understand the drying process at a single droplet level, to control product quality, to design associated equipments and to optimize process conditions for the best-quality product, the improved energy saving and the reduced overall cost. Average moisture content and average temperature profiles of the material can be evaluated using available drying kinetics models, which somewhat differ in their complexity and accuracy. A simple yet accurate drying kinetics model is desirable for fast computation of the drying process using various tools such as Microsoft Excel, computational fluid dynamics (CFD), etc.

Over the last decade, the influence of the droplet/particle's surface properties and surface composition on the quality of the end product (e.g. cohesiveness, agglomeration, etc.) and the product functionality during final applications (e.g. mixing, dispersion, solubility, etc.) has become clearer. Surface properties of the droplet or the semi-dried particle are also found to affect the stickiness behavior of the material during processing which can lead to some problems such as wall deposition, unwanted agglomeration, lump formation and inferior product quality (Chen and Özkan, 2007). In essence, Chen and Özkan (2007) suggested that stickiness is a surface property of the droplet/particle, and influenced by surface moisture content and surface composition.

To decide if the droplet surface is sticky at any location in the dryer or how long it will take to achieve the non-sticky droplet surface, the 'safe drying regime' may be evaluated using the sticky point temperature near the droplet's surface region (Adhikari et al., 2003, 2004). Most studies in the literature evaluated the difference between average particle temperatures and glass-transition temperature (T_g) corresponding to average moisture contents of the droplet/particle (Bhandari and Howes, 1999; Jaya et al., 2002; Roos, 2002). Recently, Adhikari et al. (2005) recommended that safe drying regimes should be formulated based on the droplet's 'surface' glass transition temperature to correlate a glass transition phenomenon with the stickiness of the product. Adhikari et al. (2005) concluded that the droplet's surface may be considered non-sticky when the droplet/particle's surface temperature is lower than the 'surface' T_g by 10 ºC. Estimation of the surface T_g required the surface moisture content profile to be available. In a way, it is important to obtain surface moisture content of the material under drying conditions.

Lumped-parameter drying kinetics models such as characteristic drying rate curve (CDRC) model, reaction engineering approach (REA) predict average moisture content of materials during drying. To date there are no experimental techniques available to accurately measure the surface moisture content of small droplets under 'actual' spray drying conditions or even 'isolated' laboratory drying conditions. This is mainly due to the 'smallness' of the droplet and the extremely rapid nature of drying at the droplet's surface. The difficulty of measurement is further increased when the material surface is receding due to the loss of moisture from the surface. Most studies in the literature on estimating surface moisture contents used the effective diffusivity-based drying kinetics model which solved the Fickian-type convective diffusion equation (Adhikari et al., 2004, 2005; Brenn, 2004; Shabde et al., 2005). The diffusion model requires solving partial differential equations using appropriate numerical methods which provide moisture concentration profiles within materials. The accuracy of predicting the surface moisture content was not provided in any studies because of experimental restrictions. The accuracy of prediction with this diffusion model also depends on the precision in obtaining the effective diffusivity of the specific material from laboratory experiments. Dryer-wide simulations using industrial spray drying conditions become complicated and time-consuming when this kind of drying kinetics model is integrated with CFD programs to estimate surface properties of the material.

Here, we have presented a simple procedure to calculate the droplet/particle's surface moisture content during convective drying. This procedure is established using the REA-based drying kinetics model. The REA is a semi-empirical, lumped-parameter model that holds a physical meaning of the drying process. The REA was firstly reported around 11 years ago by Chen and Xie (1997) to model the air drying of small aqueous droplets and thin-layer films of porous food materials. Over the last decade, the REA was successfully used in predicting 'average' properties of aqueous droplets of skim milk, whole milk, lactose, cream, whey proteins concentrate, sucrose and maltodextrin (Chen and Lin, 2005; Lin and Chen, 2006, 2007; Patel et al., 2008). Recently a 'composite' REA, a novel application of the REA, is proposed to formulate the drying kinetics model for the drying of aqueous sugar droplets containing multiple solutes (Patel et al., 2008).

In this paper, we have reported another innovative application of the REA for the first time in order to estimate the surface moisture content of aqueous droplets during convective drying. Predictions are made here for the drying of aqueous maltodextrin (DE 6) droplets, as an example, which were dried using hot gas of constant temperature and humidity. The REA for the drying of maltodextrin (DE6) droplets is formulated by Patel et al. (2008) based on experimental data reported by Adhikari et al. (2003, 2004). Surface glass-transition temperatures of maltodextrin droplets are also predicted in this study using mass fractions of surface moisture and surface solids. The surface moisture content estimation procedure and other important equations are briefly described in the mathematical model section.

MATHEMATICAL MODEL

Surface Moisture Content

The REA defines the drying flux of the droplet/particle/thin-film using the vapor concentration difference as shown by the following equation (Chen and Lin, 2005):

$$-\frac{dm_p}{dt} = -m_{solids}\frac{d\overline{X}}{dt} = h_m A_p(\rho_{v,sur} - \rho_{v,b}) \quad (1)$$

where $\rho_{v,sur}$ (kg·m^{-3}) is the vapor concentration at the droplet's surface, $\rho_{v,b}$ (kg·m^{-3}) is the vapor concentration of the bulk drying gas, $\overline{X}$ (kg·kg^{-1}, dry basis) is the droplet's average moisture content, h_m (m·s^{-1}) is the mass-transfer coefficient, A_p (m^2) is the droplet's surface area, and m_p and m_{solids} (kg) are weights of a single droplet and its solids. In Eq.(1) the surface vapor concentration ($\rho_{v,sur}$) is an unknown parameter, and it is so far not practical to directly measure from the droplet drying experiment. The REA correlated the surface vapor concentration with the corresponding saturated vapor concentration ($\rho_{v,sat}$) and the surface relative humidity (in fraction) as follows:

$$\rho_{v,sur} = \rho_{v,sat}\exp\left(-\frac{\Delta E_v}{R_g T_p}\right) \quad (2)$$

where R_g (J·mol^{-1}·K^{-1}) is the universal gas constant and T_p (K) is the average droplet/particle temperature. In Eq.(2) the exponential term presents the surface relative humidity. The parameter ΔE_v (J·mol^{-1}) is the apparent activation energy, which is a material-dependent property and can be correlated with average and initial moisture contents of the droplet/particle. In fact, the relationship between the relative activation energy ($\Delta E_v/\Delta E_{v,b}$) and the droplet's moisture content ($\bar{X} - X_b$) was viewed as a 'fingerprint' of the material (Chen and Xie, 1997; Chen and Lin, 2005). Recently, Chen (2008) and Patel et al. (2008) reported that the relative activation energy of the droplet should also be a function of the droplet's initial moisture content. For the convective air drying of aqueous maltodextrin droplets of 1.0 kg·kg^{-1} and 1.5 kg·kg^{-1} initial moisture contents, the relationship were expressed as (Patel et al., 2008):

For maltodextrin (DE6) of $X_0 = 1.0$ kg·kg^{-1}

$$\frac{\Delta E_v}{\Delta E_{v,b}} = \left[1 - 0.9438(X - X_b)^{8.824}\right] \times \exp\left(-0.603(X - X_b)^{2.024}\right) \tag{3}$$

For maltodextrin (DE6) of $X_0 = 1.5$ kg·kg^{-1}

$$\frac{\Delta E_v}{\Delta E_{v,b}} = \left[1 - 0.03447(\bar{X} - X_b)^{8.295}\right] \times \exp\left(-0.5353(\bar{X} - X_b)^{1.686}\right) \tag{4}$$

The average relative error in evaluating above relationships from the experimental work was less than 1.5%. In Eq.(3) and Eq.(4), X_b is the equilibrium moisture content, which can be calculated from the isotherm model of the specific material. The parameter $\Delta E_{v,b}$ is the equilibrium activation energy corresponding to drying gas conditions. $\Delta E_{v,b}$ can be calculated from the following equation:

$$\Delta E_{v,b} = -R_g T_b \ln\left(RH_b\right) \tag{5}$$

where RH_b and T_b are relative humidity (in fraction) and temperature of the drying medium. The surface vapor concentration ($\rho_{v,sur}$) can be calculated using Eq.(2) to Eq.(5). Using the surface vapor concentration, the vapor pressure at the droplet surface ($P_{v,sur}$) can be calculated using the ideal gas law:

$$P_{v,sur} = \frac{\rho_{v,s} R_g T_p}{M_w \cdot 101.325} \tag{6}$$

where M_w is the molecular weight of water. Using the surface vapor pressure information, the surface water activity ($a_{w,sur}$) can be evaluated from Raoult's law:

$$a_{w,sur} = \frac{P_{v,sur}}{P_{v,sat}} \tag{7}$$

where $P_{v,sat}$ (atm) is the saturated vapor pressure corresponding to the droplet's surface temperature. In this study, the temperature distribution within aqueous maltodextrin droplets was assumed negligible because corresponding heat-transfer Biot numbers were lower than the critical value of 0.1 throughout drying (Adhikari et al., 2004, 2005). Therefore, the droplet's surface temperature was taken the same as the droplet's average temperature. $P_{v,sat}$ (atm) in Eq.(7) can be estimated using the following Antoine's equation:

$$\log_{10} P_{v,sat} = \left(7.94917 - \frac{1657.462}{T_p - 46.23}\right) \times \frac{1}{760} \tag{8}$$

In Eq.(8), T_p is in K. To estimate the surface moisture content, it is conventionally assumed that the surface water activity ($a_{w,sur}$) and the surface moisture content (X_{sur}) can be correlated using the equilibrium moisture isotherm. Then the surface moisture content X_{sur} (kg·kg^{-1}, dry basis) can be estimated using the relevant isotherm model of the specific material. For aqueous maltodextrin droplets, the equilibrium moisture isotherm was presented using the popular Guggenheim-Anderson-deBoer (GAB) model (Adhikari et al., 2003, 2004). Then X_{sur} of aqueous maltodextrin droplets during drying can be estimated using the following isotherm model:

$$X_{sur} = \frac{m_0 C K a_{w,sur}}{(1 - K a_{w,sur})(1 - K a_{w,sur} + C K a_{w,sur})} \tag{9}$$

where, m_0, C and K are GAB parameters. For aqueous maltodextrin (DE6) droplets, m_0, C and K were reported to be 0.04, 30 and 0.98, respectively (Adhikari et al., 2003).

Glass Transition Temperature (T_g)

The glass-transition temperature (T_g) of the solid-water mixture is the moisture concentration-dependent property of the material. The Gordon-Taylor equation has widely been used in the literature to estimate T_g of the solids-water mixture containing single or multiple solutes (Roos, 2002; Adhikari et al., 2004, 2005). Since we are interested in estimating the surface glass-transition temperature ($T_{g,sur}$), the Gordon-Taylor equation is expressed here as a function of mass fractions of solids (ω_s) and water (ω_w) at the 'surface' of the material:

$$T_{g,sur} = \frac{\omega_s T_{g,s} + K_G \omega_w T_{g,w}}{\omega_s + K_G \omega_w} \tag{10}$$

where K_G is the solid-water constant, $T_{g,s}$ and $T_{g,w}$ are glass transition temperatures of anhydrous solids and water, and ω_s and ω_w are mass fractions of solids and water at the surface region, respectively. For the maltodextrin(DE6)-water binary mixture, K_G is reported to be 7.7, and T_g of anhydrous maltodextrin and water were recorded as 205 ºC and -135 ºC, respectively (Adhikari et al., 2003, 2004, 2005).

Mass fractions of water and solids in Eq.(10) can be estimated using the droplet's surface water content as shown by following equations:

$$\omega_w = \frac{X_{sur}}{1 + X_{sur}} \qquad (11)$$

$$\omega_s = \frac{1}{1 + X_{sur}} \qquad (12)$$

Since the average droplet temperature (T_p) would be changing during drying, it is essential to obtain the droplet's average temperature profile during drying. The average droplet temperature in equations (2), (6) and (8) can directly be obtained from the single droplet drying experiment when modeling the drying of single droplets, or can be calculated using the following heat-transfer model when modeling the drying process under 'actual' spray drying conditions:

$$\frac{dT_p}{dt} = \frac{hA_p(T_b - T_p) + \Delta H_v \dfrac{dm_w}{dt}}{(m_{solids}C_{p,solids} + m_w C_{p,w})} \qquad (13)$$

In Eq.(13), h (W·m^{-2}·K^{-1}) is the convective heat-transfer coefficient, ΔH_v (J·kg^{-1}) is the latent heat of vaporization and C_p (J·kg^{-1}·K^{-1}) is the specific heat capacity. Parameters h and h_m can be calculated using appropriate Ranz-Marshall correlations. In this study, following correlations were used to calculate h and h_m:

$$Nu = \frac{h \cdot d_p}{k_b} = 2 + 0.6(Re)^{1/2}(Pr)^{1/3} \qquad (14)$$

$$Sh = \frac{h_m d_p}{D_v} = 2 + 0.6(Re)^{1/2}(Sc)^{1/3} \qquad (15)$$

Correlations to calculate thermo-physical properties of air, water and maltodextrin (DE6) are reported elsewhere (Adhikari et al., 2003; Patel et al., 2008).

SIMULATION

Surface moisture contents and surface glass-transition temperatures are predicted in this paper for the drying of aqueous maltodextrin droplets of 1.0 kg·kg^{-1} and 1.5 kg·kg^{-1} initial moisture contents (all dry basis). Experimental data on the air drying of single suspended maltodextrin (DE6) droplets are provided by Dr Benu Adhikari (a senior lecturer at Ballarat University, Australia). Details of the experimental work are reported by Adhikari et al. (2003, 2004). In their experimental work, droplets were dried using hot air of 1.0 m/s velocity, 63 ± 1 °C temperature and 2.5 ± 0.5 %*RH*. Air temperature and air humidity were kept constant throughout the course of drying. Average moisture content and temperature profiles of the droplets are calculated using the REA-based drying kinetics model and the heat-transfer model shown by Eq.(1) and Eq.(13), respectively, and compared with those obtained from the experimental work (see Fig. 1 to Fig. 4). The average relative error in matching average and measured profiles for the droplet's moisture content and temperature were reported to be less than 1.5% and 2.0% respectively. The equilibrium moisture content (X_b) corresponding to the 63 °C temperature and 2.5 %*RH* air condition for maltodextrin droplets was calculated as 0.00001 kg·kg^{-1}. The GAB model described by Eq.(9) was used to estimate X_b based on the 'average' water activity of the droplet/particle. Initial droplet characteristics for maltodextrin droplets used in this study are listed in Table 1.

Table 1. Initial droplet conditions for the air drying of maltodextrin (DE6) droplets of two different initial moisture contents.

Droplet	X_0=1.0 kg·kg^{-1}	X_0=1.5 kg·kg^{-1}
Diameter, mm	2.188	2.214
Temperature, °C	22.03	23.56

RESULTS AND DISCUSSION

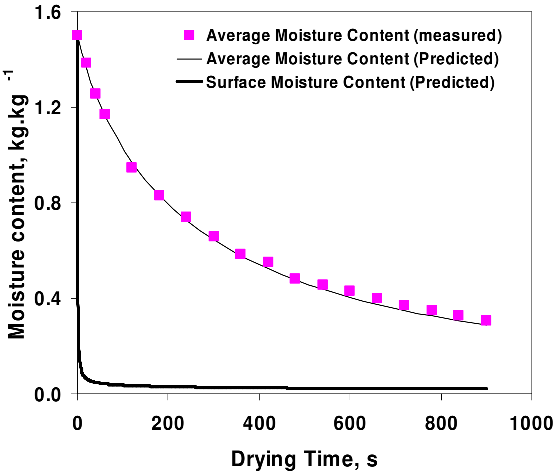

Fig. 1. Average and surface moisture contents for the air drying of maltodextrin droplets of X_0 = 1.5 kg·kg^{-1} using hot air of 63 °C temperature and 2.5 %*RH*

Fig. 1 illustrates average moisture content profiles (both predicted and measured) and the predicted surface moisture content profile for the drying of maltodextrin droplets of X_0 = 1.5 kg·kg^{-1}. It was observed that the surface moisture content dropped drastically during the earlier drying stage, and reduced gradually during the later drying stage. The same trend is reported by Adhikari et al. (2003) when predicting X_{sur} using the diffusion model. At the end of drying, X_{sur} (t = 900 seconds) was 0.02 kg·kg^{-1} using the REA whilst the measured average moisture content was 0.29 kg·kg^{-1}. Since experimental data on surface moisture contents are not available, predictions with the REA are compared with those presented by Adhikari et al. (2003). For the same droplet and drying conditions used in this work, the diffusion model estimated X_{sur} at the end of drying as 0.06 kg·kg^{-1}. However, the average relative error of the prediction was reported to be around 6 % by the diffusion model (Adhikari et al. 2003). In a way, the prediction of X_{sur} with the REA is close to the one predicted by the diffusion model.

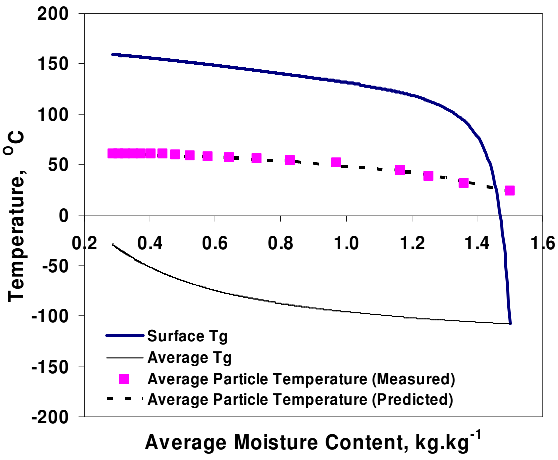

Fig. 2. Average and surface T_g for the air drying of maltodextrin droplets of X_0 = 1.5 kg·kg^{-1} using hot air of 63 °C temperature and 2.5 *%RH*

Surface glass-transition temperatures ($T_{g,sur}$) and average T_g are calculated based on surface and average moisture contents respectively for the drying of maltodextrin droplets of X_0 = 1.5 kg·kg^{-1}. Results are presented in Fig. 2. To check the difference between the average particle temperature (T_p) and the glass transition temperature, average and measured T_p profiles are also illustrated in Fig. 2. Results show a large difference between average and surface glass-transition temperatures. At the end of drying, $T_{g,sur}$ and average T_g with the REA were 159 °C and -29 °C, respectively. Predictions of $T_{g,sur}$ and T_g with the REA are close to glass-transition temperatures estimated by the diffusion model of Adhikari et al. (2003, 2004). It can be observed from Fig. 2 that the average T_g is considerably lower than the average particle temperature (the difference was greater than 90 °C) during drying, suggesting that the droplet surface may be sticky. However, $T_{g,sur}$ is significantly higher than the average particle temperature during drying, indicating that the droplet surface should be non-sticky. Adhikari et al. (2003) measured the stickiness behavior of maltodextrin (DE6) droplets using a probe-tack test, and found that maltodextrin droplets forms a thick skin on the surface as drying proceeded, and the surface was non-sticky during the later drying stage or after the average moisture content reached to 0.69 kg·kg^{-1}. Werner et al. (2007) also performed similar experiments to check the stickiness behavior of maltodextrin droplets with similar drying conditions, and found that the surface of maltodextrin droplets was non-sticky during the later drying stage. These experimental studies confirmed that the surface T_g should be used to evaluate the safe drying regime during processing.

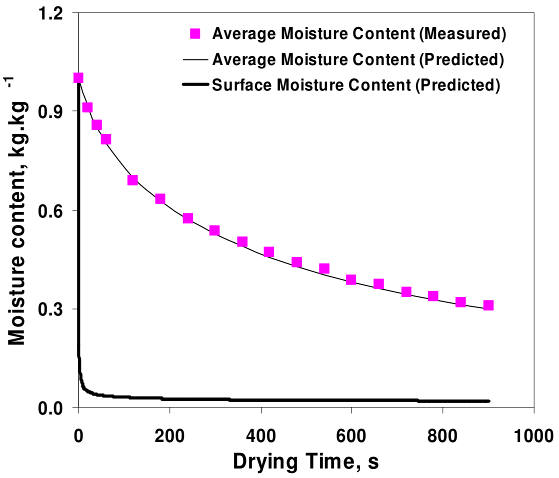

Fig. 3. Average and surface moisture contents for the air drying of maltodextrin droplets of X_0 = 1.0 kg·kg^{-1} using hot air of 63 °C temperature and 2.5 *%RH*

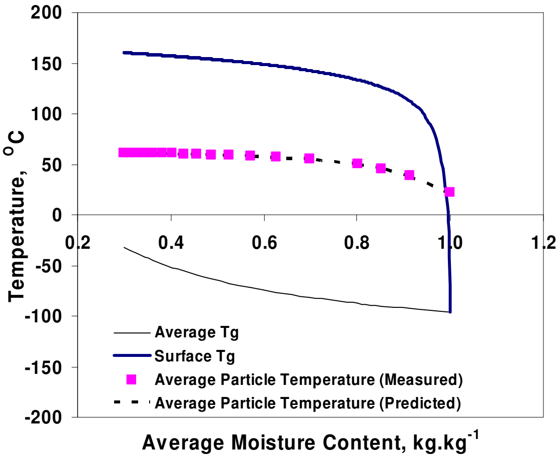

Fig. 4. Average and surface T_g for the air drying of maltodextrin droplets of X_0 = 1.0 kg·kg^{-1} using hot air of 63 °C temperature and 2.5 *%RH*

Surface moisture contents and surface glass transition temperatures are also predicted for the drying of maltodextrin (DE6) droplets of X_0 = 1.0 kg·kg^{-1}, which were dried using hot air of 63 °C temperature and 2.5 *%RH* condition. Predicted surface moisture content and surface glass-transition temperature profiles for these droplets are presented in Fig.3 and Fig. 4, respectively. The same trends for maltodextrin droplets of X_0 = 1.0 kg·kg^{-1} were observed as for maltodextrin droplets of X_0 = 1.5 kg·kg^{-1}. However, the surface moisture content reduced more drastically for the droplets of X_0 = 1.0 kg·kg^{-1}. For the droplets of 1.0 kg·kg^{-1} initial moisture contents, X_{sur} at the end of drying was 0.02 kg·kg^{-1}, same as X_{sur} at the end of drying for the droplets of $X_0 = 1.5$ kg·kg^{-1}. The main reason was the average moisture content at the end of drying for the droplets of both initial moisture contents were close to each other at the end of drying (the difference was around 1 ºC).

Similarly average and surface glass transition temperatures were close to each other for the droplets of both initial moisture contents. However, $T_{g,sur}$ for the droplets of $X_0 = 1.0$ kg·kg^{-1} increased more sharply during the earlier drying stage compared to those for the droplets of $X_0 = 1.5$ kg·kg^{-1}. This is due to a more drastic reduction in the surface moisture content for the droplets of $X_0 = 1.0$ kg·kg^{-1}. Furthermore, predictions of surface glass transition temperatures were close to the numbers calculated by the diffusion model.

CONCLUSIONS

This paper describes a simple procedure to estimate the surface moisture content of the material under drying conditions. Predictions in this study using the REA-based drying kinetics model illustrated that surface moisture contents and surface glass-transition temperatures using the REA are close to those obtained by the diffusion model in the work by Adhikari et al. (2003, 2004). The procedure for estimating the surface moisture content using the REA-based drying kinetics model appears to be effective and reliable. This procedure can readily be used to estimate the surface moisture content for small falling droplets in industrial-scale spray dryers. In fact, when the REA is employed in process calculation tools to calculate the average moisture content profile, only a few additional equations need to be added for estimating the surface moisture content profile. The accuracy of predicting the surface moisture content relies on the accuracy of the relative activation energy relationship or the 'fingerprint' of the specific material, for instance shown by Eq.(3) and Eq.(4). Once the surface moisture content profile is known, other water-dependent properties such as glass-transition temperature at the droplet surface can be predicted.

NOMENCLATURE

a_w	water activity	
A	surface area	m^2
C_p	specific heat capacity	J·kg^{-1}·K^{-1}
D_v	air-vapor diffusion coefficient	m^2·s^{-1}
ΔE_v	apparent activation energy	J·mol^{-1}
$\Delta E_{v,b}$	equilibrium activation energy	J·mol^{-1}
h	heat-transfer coefficient	W·m^{-2}·K^{-1}
h_m	mass-transfer coefficient	m·s^{-1}
ΔH_v	latent heat of vaporization	J·kg^{-1}
k	Thermal conductivity	W·m^{-1}·K^{-1}
K_G	constant in Gordon-Taylor equation	
m	weight	kg
M_w	molecular weight of water (=18.015 g·mol^{-1})	
Pr	Prandtl number	
P_v	vapor pressure	atm
Re	Reynolds Number	
R_g	universal gas constant (=8.314 J·mol^{-1}·K^{-1})	
RH	relative humidity	
Sc	Schmidt Number	
Sh	Sherwood Number	
T	temperature	K
T_g	glass transition temperature	K
t	time	s
$\bar{X}$	average moisture content	kg·kg^{-1} (dry basis)
X	droplet moisture content	kg·kg^{-1} (dry basis)
X_b	equilibrium moisture content	kg·kg^{-1}

Greek letters

ω	mass fraction	
ρ	density	kg·m^{-3}
ρ_v	vapor concentration	kg·m^{-3}

Subscripts

0	initial conditions
b	bulk drying gas
p	particle, droplet
sur	surface conditions
sat	saturated conditions
v	vapor
w	water

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