Moisture Content Distribution in Semibatch Drying Processes, Part II. Falling Particle Drying Rate

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This article describes the evolution of the moisture content distribution in a population of wet particles during a semibatch drying process. The population balance model that is defined for this process includes falling particle drying rate, that is, the drying rate of a particle depends on its moisture content. This model is solved analytically using the "method of moments" and "method of characteristics." Simulation experiments give realistic and meaningful results. Drying experiments still have to be performed to test the validity of the assumptions made in this model. © 2012 American Institute of Chemical Engineers *AIChE J,* 58: 3708–3717, 2012
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Introduction

Drying operations are of great importance in many industries. Their main purpose is to remove abundant moisture from (solid) materials. This is performed by heat and mass transfer. At the end of a drying process, the moisture content should have reached a predefined specification. For batch processes, the kinetics describing such a drying process have been presented in many different literature sources.[2,3,5] Continuous processes, have been well described. For these processes, Burgschweiger and Tsotas[1] have presented an excellent model that couples population balance modeling to heat and mass transfer equations.[1] In many publications, the moisture content in the dryer is averaged for all particles. For batch and steady-state continuous processes, this is a valid assumption as residence times of particles generally do not change during the process. However, in semibatch drying processes residence times of particles change in time. Particles that have entered the dryer first have a longer residence time than particles that have entered last. Averaging of moisture content is still possible for this process but minimum and maximum deviations from the average values will be greater compared to batch and steady state continuous processes. Peglow et al.[5] have presented a simple population balance model also on the basis of residence time distribution. A moisture content distribution for continuous fluidized bed drying during steady state was calculated. However, semibatch drying processes are constantly in a nonsteady state, that is, the moisture content distribution is constantly changing in time.

Often the moisture content is required to meet specifications that are based on subsequent process steps (e.g., flowability, stickiness, compaction) or product quality (e.g., microbiological contamination, chemical, and physical stability). For the latter, it is particularly important to obtain knowledge of the evolution of moisture content distribution during drying.

In a previous article, (Nitert and Hounslow, submitted) the authors have presented a model that describes moisture content distribution during a constant particle drying rate process. In this article, it is further extended to falling particle drying rate.

Defining Falling Drying Rate and Drying System

In the previous article of Nitert and Hounslow (submitted) a characteristic drying curve of a wet particle was presented. It was assumed that both dx/dt and χ were constant. Falling particle drying rate follows constant particle drying rate and is initiated when particles have reached a critical moisture content, x^{cr}. The rate limiting step of drying is no longer evaporation of solvent from the surface of the particle but diffusion of solvent through inner pores of the particle (i.e., internal conditions). In this article, falling drying rate of individual particles is included in the population balance models. However, the overall solvent removal rate, χ, is still assumed to be constant. In other words, although (some) particles already have entered a stage of falling drying rate, the bulk gas is still saturated with moisture. The subsequent stage of bulk gas becoming unsaturated, that is, an overall falling drying rate, is not discussed in this article.

Figure 1 shows an overview of a simplified drying system. All parameters are similar to those described in the previous article Ref. (Nitert and Hounslow submitted). Wet particles enter the system with moisture content x^0 and feed rate w^0, and moisture is removed at a net rate χ. $M_T(0)$ represents the initial charge of wet particles in the dryer. In this article, $M_T^*(t)$ is included in $m_0(t)$ as no distinction is made between wet and dry particles.

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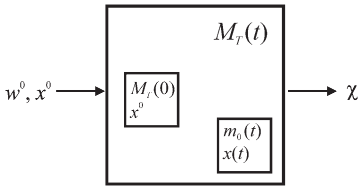

Figure 1. Schematic overview of an arbitrary semibatch drying process.

Wet particles enter the system with moisture content x^0 and feed rate w^0. Absolute net moisture removal rate is χ. $M_T(0)$ represents the initial charge of wet particles in the dryer. At a time t, the dryer contains $M_T(t)$ kg particles that consists of $m_0(t)$ kg wet particles (adopted from Nitert and Hounslow, submitted).

Population Balance Model for a Semibatch Drying Process

A population balance is defined for the drying system presented in Figure 1. The same assumptions hold as for the population balance of wet particles in the previous article. Nitert and Hounslow (submitted) have also shown that this population balance does not depend on position in the bed when a well-mixed system is assumed (i.e., no external coordinates have to be included). However, the difference with the previous article is that here single particle drying rate (dx/dt) depends on moisture content x as falling particle drying rate is assumed. This results in the following population balance for a semibatch drying system with falling drying rate.

$$\frac{\partial f(x|t)}{\partial t} + \frac{\partial (f(x|t)\dot{x}(x|t))}{\partial x} = w^0 \delta(x - x^0) \tag{1}$$

where $f(x|0) = M_T(0)\delta(x - x_0)$ and $\dot{x}(x|t) = dx/dt$.

It is important to recognize that particles will never be fully dry (i.e., $x > x^*$) as during falling drying rate moisture content will decrease asymptotically to the equilibrium moisture content x^*.

During falling drying rate (where $x \in (x^{cr}, x^*)$), the particle drying rate is determined by its internal material properties (e.g., porosity and tortuosity). As a result, the particle drying rate is decreasing. This is included in falling drying rate by introducing a relative drying rate, named $h(x)$ here.[2–4] Figure 2 shows an example how $h(x)$ can depend on relative moisture content Φ ($= (x - x^*)/(x^{cr} - x^*)$). This characteristic drying curve is a material property, that is, it is independent of time. Drying rate ($\dot{x}$ in kg solvent/s) determines the required time to change the moisture content of the particle along the characteristic drying curve. This relationship is described by the following equations.

$$\dot{x}(x|t) = h(x)\dot{x}(t) \quad \text{(falling drying rate)} \tag{2a}$$

$$\dot{x}(t) = -k'(t) \quad \text{(constant drying rate)} \tag{2b}$$

$$\dot{x}(x|t) = -h(x)k'(t) \tag{2c}$$

Combining the equation of relative moisture content, Φ, with Eq. 2c gives

$$\frac{dx}{dt} = -k(t)\frac{x - x^*}{x^0 - x^*} \tag{3}$$

where

$$k(t) = k'(t)\frac{x^0 - x^*}{x^{\text{cr}} - x^*}$$

Incorporating Eq. 3 into the population balance (Eq. 1)

$$\frac{\partial f(x|t)}{\partial t} - k(t)\frac{\partial \left(f(x|t)\frac{x - x^*}{x^0 - x^*}\right)}{\partial x} = w^0 \delta(x - x^0) \tag{4}$$

The following mass balance can be written for the drying system

$$m_0(t) = \int_{x^*}^{x^0} f(x|t)dx \tag{5a}$$

$$\frac{dm_0(t)}{dt} = w^0 \tag{5b}$$

$$\frac{d}{dt}\int_{x^*}^{x^0} f(x|t)dx = w^0 \tag{5c}$$

where $m_0(t)$ is the zeroth moment of the particle population $f(x|t)$ representing the total amount of material in the dryer (in kg dry solid). The first moment of this population, $m_1(t)$, gives the total amount of solvent of particles in the dryer

$$m_1(t) = \int_{x^*}^{x^0} xf(x|t)dx \tag{6a}$$

$$\frac{dm_1(t)}{dt} = w^0 x^0 - \chi \tag{6b}$$

$$\frac{d}{dt}\int_{x^*}^{x^0} x^* f(x|t)dx = w^0 x^0 - \chi \tag{6c}$$

These equations can be used to find expressions for evolution of the moisture content distribution during falling drying rate.

Solution Strategy to Find *f*(*x*|*t*) for Feeding Stage

Solution of the previously mentioned population balance (Eq. 4) requires a set of equations that are solved without knowledge of the moisture content distribution among particles. In other words, solution is only possible when input process parameters such as inlet mass flow rate, w^0, overall solvent removal rate, χ, inlet and equilibrium moisture contents, respectively, x^0 and x^*, can be related to the moisture content distribution. Similar to the previous article (Nitert and Hounslow, submitted) a dimensionless population balance equation and dimensionless dryness are defined. However, the subsequent steps are different from the solution

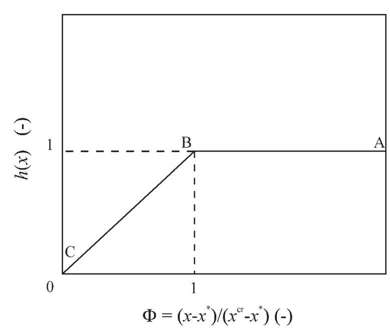

Figure 2. Relative drying rate (*h*) as a function of relative moisture content Φ at constant drying rate (A and B) and falling drying rate (B and C).

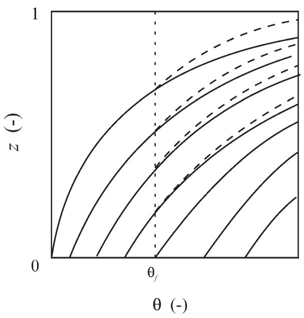

Figure 3. Method of characteristics.

The dashed lines represent an expression $g(z|\theta)$ that described its drying of particles after the feeding phase ($\theta \geq \theta_f$). The solid lines describe the evolution of $g(z|\theta)$ when the dryer is continuously fed with wet particles. The corresponding expression is used to describe the drying process during the feeding phase ($\theta < \theta f$).

strategy presented by (Nitert and Hounslow, submitted). Unlike constant particle drying rate, falling particle drying rate is not divided into wet and dry particle populations which simplifies the solution method. In this section, an analytical expression for $g(z|\theta)$ is defined that is only a function of variables z and θ. The method of characteristics is used to solve the population balance equations. Every characteristic represents a population of particles $g(z|\theta)$ that have entered the dryer at $\theta = \theta_0$. Figure 3 illustrates how this method is applied using the following solution strategy:

1. Define an expression for $g(z|\boldsymbol{\theta})$ that describes a drying process with a continuous feed of wet particles. This expression describes all solid lines in Figure 3.

2. Calculate θ_f (dimensionless feeding time) from t_f. Feeding is stopped at this point, that is, no new characteristics are generated.

3. Find an expression for $g(z|\theta)$ that describes the drying process of a defined amount of material after feeding, that is, a batch drying-process. This expression only describes the dashed lines in Figure 3, that is, the evolution of $g(z|\theta)$ after feeding.

Expression 1 is used to describe the feeding stage of a semibatch process. Expression 2 gives the termination criterium of this feeding stage and the starting condition of the drying stage (expression 3).

Defining a dimensionless population balance g(z|θ)

A dimensionless population balance equation and dimensionless dryness z are defined similar to the ones reported in an article of Nitert and Hounslow, (submitted). The difference in this article is that here $g(z|\theta)$ describes all particles in the dryer, whereas in the previous article $g(z|\theta)$ only described wet particles.

$$z = \frac{x - x^0}{x^* - x^0} \tag{7}$$

The dimensionless drying rate, $\dot{z}(t)$ or dz/dt, is obtained from $\dot{x}(x|t)$

$$\frac{dz}{dt} = \frac{1}{x^* - x^0}\left(-k(t)\frac{x - x^*}{x^0 - x^*}\right) \tag{8a}$$

$$= (1 - z)\frac{k(t)}{x^0 - x^*} \tag{8b}$$

A dimensionless time or extent of drying, θ that is related to time t, is defined

$$\frac{d\theta}{dt} = \frac{d\theta}{dz}\frac{dz}{dt} \tag{9a}$$

$$= (1 - z)\frac{k(t)}{x^0 - x^*}\frac{d\theta}{dz} \tag{9b}$$

$$= \frac{k(t)}{x^0 - x^*} \tag{9c}$$

where $d\theta/dz = (1 - z)^{-1}$ to obtain an expression for $d\theta/dt$ that is independent of z and x. A dimensionless mass density distribution, $g(z|\theta)$, is defined with z as internal coordinate

$$g(z|\theta)dz = \frac{1}{M_T(0)}f(x|t)dx \tag{10}$$

dz/dx can be obtained by differentiating Eq. 7. This transforms Eq. 10 into

$$f(x|t) = \left|\frac{dz}{dx}\right|M_T(0)g(z|\theta) \tag{11}$$

Substituting Eqs. 9c and 11 into Eq. 4 gives

$$\frac{k(t)M_T(0)}{x^0 - x^*}\left|\frac{dz}{dx}\right|\frac{\partial g(z|\theta)}{\partial\theta} - k(t)M_T(0)\frac{\partial z}{\partial x}\frac{\partial}{\partial z}\left((1 - z)g(z|\theta)\left|\frac{dz}{dx}\right|\right) = w_0\delta(z)\left|\frac{dz}{dx}\right| \tag{12a}$$

$$\frac{k(t)M_T(0)}{x^0 - x^*}\frac{\partial g}{\partial\theta} + k(t)M_T(0)\frac{\partial}{\partial z}\left((1 - z)g(z|\theta)\left|\frac{dz}{dx}\right|\right) = w_0\delta(z) \tag{12b}$$

$$\frac{\partial g(z|\theta)}{\partial\theta} + \frac{\partial}{\partial z}((1 - z)g(z|\theta)) = \frac{w_0(x^0 - x^*)}{k(\theta)M_T(0)}\delta(z) \tag{12c}$$

Finally, Eq. 12c can be simplified to:

$$\frac{\partial g(z|\theta)}{\partial\theta} + (1 - z)\frac{\partial g(z|\theta)}{\partial z} - g(z|\theta) = \frac{w_0(x^0 - x^*)}{k(\theta)M_T(0)}\delta(z) \tag{13}$$

The zeroth and first moment of $g(z|\theta)$ are, respectively,

$$\mu_0(\theta) = \int_0^1 g(z|\theta)\,dz \tag{14}$$

$$\mu_1(\theta) = \int_0^1 zg(z|\theta)\,dz \tag{15}$$

Defining k(θ), *$\mu_0(\theta)$, and $\mu_1(\theta)$* An expression for $k(\theta)$ is found when expressions for $\mu_0(\theta)$ and $\mu_1(\theta)$ can be defined. For the latter, the mass balance of material in the dryer (Eq. 5) is converted into a dimensionless mass balance using Eqs. 7 and 10

$$\frac{d}{dt}\int_{x^*}^{x^0} f(x|t)dx = w^0 \tag{16a}$$

$$\frac{k(\theta)}{x^0 - x^*}\frac{d}{d\theta}\int_0^1 M_T(0)g(z|\theta)dz = w^0 \tag{16b}$$

$$\frac{d\mu_0(\theta)}{d\theta} = \frac{w^0(x^0 - x^*)}{k(\theta)M_T(0)} \tag{16c}$$

The same expression is found when a moment transformation is performed on Eq. 13

$$\int_0^1 \frac{\partial g(z|\theta)}{\partial \theta} dz + \int_0^1 (1-z)\frac{\partial g(z|\theta)}{\partial z} dz - \int_0^1 g(z|\theta) dz = \frac{w^0(x^0-x^*)}{k(\theta)M_T(0)} \int_0^1 \delta(z) dz \quad (17a)$$

$$\frac{\partial \mu_0(\theta)}{\partial \theta} + [(1-z)g(z|\theta)]_0^1 - \int_0^1 -g(z|\theta)dz - \int_0^1 g(z|\theta)dz = \frac{w^0(x^0-x^*)}{k(\theta)M_T(0)} \quad (17b)$$

$$\frac{d\mu_0(\theta)}{d\theta} = \frac{w^0(x^0-x^*)}{k(\theta)M_T(0)} \quad (17c)$$

This relationship between the mass and population balance is exploited further to find an expression for $k(\theta)$ by defining $\mu_1(\theta)$ in a similar way. Using Eqs. 7 and 9c, the mass balance of solvent in the system (Eq. 6) can be written as

$$\frac{k(\theta)M_T(0)}{x^0-x^*}\frac{d}{d\theta}\int_0^1 (z(x^*-x^0)+x^0)g(z|\theta)\, dz = w^0x^0 - \chi \quad (18a)$$

$$\frac{k(\theta)M_T(0)}{x^0-x^*}\frac{d}{d\theta}((x^*-x^0))\mu_1(\theta) + x^0\mu_0(\theta) = w^0x^0 - \chi \quad (18b)$$

$$-k(\theta)\frac{d\mu_1(\theta)}{d\theta} + \frac{k(\theta)x^0}{x^0-x^*}\frac{d\mu_0(\theta)}{d\theta} = \frac{w^0x^0-\chi}{M_T(0)} \quad (18c)$$

Incorporating Eq. 17c into Eq. 18c and rearranging gives

$$\frac{d\mu_1(\theta)}{d\theta} = \frac{\chi}{k(\theta)M_T(0)} \quad (19)$$

with $\mu_1(0) = 0$. The dimensionless population balance of $g(z|\theta)$ is solved for the first moment

$$\int_0^1 z\frac{\partial g(z|\theta)}{\partial \theta}\, dz + \int_0^1 (z-z^2)\frac{\partial g(z|\theta)}{\partial z}\, dz - \int_0^1 zg(z|\theta)\, dz = \frac{w_0(x^0-x^*)}{k(\theta)M_T(0)}\int_0^1 z\delta(z)\, dz \quad (20a)$$

$$\frac{d\mu_1(\theta)}{d\theta} = \mu_0(\theta) - \mu_1(\theta) \quad (20b)$$

Combining Eqs. 17c and 20b with the result of the mass balance (Eq. 19) gives, respectively

$$k(\theta) = \frac{\chi}{M_T(0)(\mu_0(\theta)-\mu_1(\theta))} \quad (21)$$

and

$$\frac{d(\mu_0(\theta)-\mu_1(\theta))}{d\theta} = \frac{w^0(x^0-x^*)}{k(\theta)M_T(0)} - \frac{\chi}{k(\theta)M_T(0)} \quad (22)$$

Incorporating Eq. 21 into Eq. 22 results in

$$\frac{d(\mu_0(\theta)-\mu_1(\theta))}{d\theta} = \left(\frac{w^0(x^0-x^*)}{\chi} - 1\right)(\mu_0(\theta)-\mu_1(\theta)) \quad (23a)$$

$$= (\alpha-1)(\mu_0(\theta)-\mu_1(\theta)) \quad (23b)$$

where $\alpha = (w^0(x^0 - x^*))/\chi$ and represents the ratio of overall feed and removal rate of solvent (see also Nitert and Hounslow, submitted). For a constant α, Eq. 23b is solved giving

$$\mu_0(\theta) - \mu_1(\theta) = e^{(\alpha-1)\theta} \quad (24)$$

Combining Eqs. 21 and 24 gives an expression for $k(\theta)$ only as a function of θ.

$$k(\theta) = \frac{\chi}{M_T(0)}e^{-(\alpha-1)\theta} \quad (25)$$

Now, it is straightforward to solve the ordinary differential equations of $\mu_0(\theta)$ and $\mu_1(\theta)$ by incorporating Eq. 25 into Eqs. 17c and 19 and integrate

$$\mu_0(\theta) - \mu_0(0) = \left[\frac{\alpha}{\alpha-1}e^{(\alpha-1)\theta}\right]_0^\theta \quad (26a)$$

$$\mu_0(\theta) = \frac{(\alpha e^{(\alpha-1)\theta}-1)}{\alpha-1} \quad (26b)$$

This result is used together with Eq. 24 to obtain an expression for $\mu_1(\theta)$.

$$\mu_1(\theta) = \mu_0(\theta) - e^{(\alpha-1)\theta} \quad (27a)$$

$$= \frac{1}{\alpha-1}(\alpha e^{(\alpha-1)\theta} - 1 - (\alpha-1)e^{(\alpha-1)\theta}) \quad (27b)$$

$$= \frac{e^{(\alpha-1)\theta}-1}{\alpha-1} \quad (27c)$$

Equation 25 is also used in the next sections to simplify $g(z|\theta)$. The expressions for $\mu_0(\theta)$ (Eq. 26b) and $\mu_1(\theta)$ (Eq. 27c) are applicable to the general case of continuous feeding of the dryer with wet particles. In what follows, these expressions are amended to describe the case when the feed of wet particles has stopped and only drying is performed.

Defining $t(\theta)$ In practice, dimensionless time θ is not a useful variable. It has to be converted into actual time t. For the feeding stage, this is performed by solving the ordinary differential Eq. 9c and subsequently incorporating Eq. 25

$$\int_0^\theta dt = \int_0^\theta \frac{x^0-x^*}{k(\theta)}\, d\theta \quad (28a)$$

$$t(\theta) = \int_0^\theta \frac{x^0-x^*}{k(\theta)}\, d\theta \quad (28b)$$

$$= \frac{M_T(0)(x^0-x^*)}{\chi}\int_0^\theta e^{(\alpha-1)\theta}\, d\theta \quad (28c)$$

$$= \frac{M_T(0)(x^0-x^*)}{\chi(\alpha-1)}(e^{(\alpha-1)\theta}-1) \quad (28d)$$

$$= \frac{t_{\text{ini}}}{(\alpha - 1)} \left(e^{(\alpha-1)\theta} - 1 \right) \tag{28e}$$

where t_{ini} is the time required to dry the initial charge, that is, $M_T(0)(x^0 - x^*)/\chi$. For the drying stage, a similar expression is defined later in this article.

Defining g(z|θ) *for feeding stage* ($\theta \leq \theta_f$)

In the previous section, a general dimensionless population balance for $g(z|\theta)$ is defined. This population balance can be solved analytically by defining the appropriate initial and boundary conditions. In this section, an expression for $g(z|\theta)$ is derived for the feeding stage ($\theta \leq \theta_f$, see Figure 3). First step in obtaining such an expression is to simplify Eq. 13 using Eq. 25. This results in

$$\frac{\partial g(z|\theta)}{d\theta} + (1 - z)\frac{\partial g(z|\theta)}{dz} - g(z|\theta) = \alpha e^{(\alpha-1)\theta}\delta(z) \tag{29}$$

with initial condition $g(z|0) = \delta(z)$ and boundary conditions $g(0^-|\theta) = 0$ and $g(0^+|\theta) = \alpha\, e^{(\alpha - 1)\theta}$. The boundary conditions are found by splitting $z = 0$ into $z = 0^-$ and $z = 0^+$. These z values and corresponding boundary conditions for $g(z|\theta)$ are explained as:

- $z = 0^-$: particles with moisture contents $x > x^0$ do not enter the dryer, that is, $g(0^-|\theta) = 0$.
- $z = 0^+$: particles with moisture contents $x = x^0 - \Delta x$, where Δx is infinitively small, that is, just when these particles have entered the dryer. $g(0^+|\theta)$ is found by integrating Eq. 29 between $z = 0^-$ and $z = 0^+$ giving: $g(0^+|\theta) = \alpha\, e^{(\alpha - 1)\theta}$.

A characteristic starts at $\theta = \theta_0$ and $z = 0$. θ_0 is obtained from the slope $dz/d\,\theta = (1 - z)$ (see Eq. 9c) giving

$$\theta_0(z|\theta) = \theta + \ln[1 - z] \tag{30}$$

Solution of the population balance equation is performed by dividing θ_0 into two segments:

1. $\theta_0 = 0$: the initial condition that describes the initial spike of wet particles of the initial charge $M_T(0)$.
2. $\theta_0 > 0$: wet particles that are fed to the dryer by an inlet stream w^0.

In the following sections, expressions for $g(z|\theta)$ are derived for both segments in the feeding stage.

Initial Charge: $\theta_0 = 0$ At $\theta_0 = 0$, the initial spike $g(z|0)$ is obtained from the initial condition $f(x|0)$ (see Eq. 1)

$$g(z|0) = \delta(z_0) \tag{31}$$

where z_0 represents the value of z at $\theta = 0$. An expression is defined that describes the evolution of the initial spike along the initial characteristic as a function of θ ($z > 0$). For $z > 0$, Eq. 13 becomes

$$\frac{\partial g(z|\theta)}{\partial\theta} + (1 - z)\frac{\partial g(z|\theta)}{\partial z} = g(z|\theta) \tag{32}$$

This transforms, with $dz/d\theta = (1 - z)$, Eq. 9c into: $Dg(z|\theta)/D\theta = g(z|\theta)$, that is, the overall dimensionless mass density of particles $g(z|\theta)$ as a function of θ changes along a characteristic according to $g(z|\theta)$. This gives for $g(z|\theta)$

$$g(z|\theta) = g(z|0)e^{\theta} \tag{33}$$

For the characteristic starting at $\theta_0 = 0$, the following expression for z is found along that characteristic (i.e., $z > 0$)

$$\theta_0(z|\theta) = 0$$
$$\theta + \ln(1 - z) = 0$$
$$z(\theta) = 1 - e^{-\theta}$$

z_0 can be rewritten as

$$z - z_0 = (1 - e^{-\theta}) - 0$$
$$z_0 = z - (1 - e^{-\theta})$$

This gives for $g(z|\theta)$ of the initial spike

$$g(z|\theta) = \delta\left(\frac{z - (1 - e^{-\theta})}{e^{\theta}}\right)e^{\theta} \tag{34a}$$

$$= \delta\left(z - (1 - e^{-\theta})\right) \tag{34b}$$

Particles from Feed: $\theta_0 > 0$ For $\theta_0 > 0$ and $z > 0$, Eq. 35 is obtained from $Dg(z|\theta)/D\theta = g(z|\theta)$

$$g(z|\theta) = g_0(\theta)e^{\theta - \theta_0(z|\theta)} \tag{35}$$

with the initial dimensionless mass density distribution $g_0(\theta)$ equals the boundary condition at $z = 0^+$ and $\theta = \theta_0$

$$g_0(\theta) = \alpha e^{(\alpha-1)\theta_0} \tag{36}$$

For $\theta_0 > 0$, this means that the contribution of the initial charge to $g(z|\theta)$ has to be added to that of the fed particles.

Incorporating Eqs. 30 and 36 into Eq. 33 results in the following expression of $g(z|\theta)$ along a characteristic ($z > 0$) starting at $\theta = \theta_0$

$$g(z|\theta) = \alpha e^{(\alpha-1)\theta} e^{\theta - \theta_0(z|\theta)} \tag{37a}$$

$$= \alpha(1 - z)^{(\alpha-2)} e^{(\alpha-1)\theta} \tag{37b}$$

Equation 37b only applies to fed particles and $z < 1 - e^{-\theta}$. Equation 34b applied to $z = 1 - e^{-\theta}$. Combining these equations results in the following general solution for $g(z|\theta)$

$$g(z|\theta) = \begin{cases} \alpha(1 - z)^{(\alpha-2)} e^{(\alpha-1)\theta} + \delta(z - (1 - e^{-\theta})) & \text{for} \quad z \leq (1 - e^{-\theta}) \\ 0 & \text{for} \quad z > (1 - e^{-\theta}) \end{cases} \tag{38}$$

The expressions for $\mu_0(\theta)$ and $\mu_1(\theta)$ defined in Eqs. 26b and 27c are used to calculate these parameters for the feeding period. The same applies to the expression for t defined in Eq. 28e. This equation can also be used to convert θ to t during feeding.

Table 1. Summary of Expressions for $g(z|\theta)$ with Corresponding Boundary Conditions

$g(z|\theta)=$	Domain z	Domain θ
$g(0|\theta)(1\text{-}z)^{(\alpha-2)} + \delta(z\text{-}(1\text{-}e^{-\theta}))$	$z \leq 1\text{-}e^{-\theta}$	$\theta < \theta_f$
$\delta(z\text{-}(1\text{-}e^{-\theta}))$	$z > 1\text{-}e^{-\theta}$	$\theta < \theta_{end}$
$g(z_f|\theta_f)\, e^{\theta-\theta_f} + \delta(z\text{-}(1\text{-}e^{-\theta}))$	$1\text{-}e^{\theta_f-\theta} \leq z \leq 1\text{-}e^{-\theta}$	$\theta_f \leq \theta < \theta_{end}$
$\mu_0(\theta_{end})\, \delta(z\text{-}1)$	$z \geq 1\text{-}e^{\theta_f-\theta}$	$\theta \geq \theta_{end}$

Solution Strategy to Find $f(x|t)$ for Drying Stage

Defining $g(z|\theta)$ for batch drying ($\theta > \theta_f$)

The previous section has presented expressions for different parameters when wet particles are continuously fed to the dryer. However, a semibatch drying process contains a predefined amount of material that is fed to the dryer. This means that after feeding no new characteristics are formed in the solution of the population balance. Although particles present in the dryer only dry, the same population balance of $g(z|\theta)$ for $z > 0$ is applicable as Eq. 32. The only exception is that the initial condition is now $g(z|\theta) = g(z_f|\theta_f)$. This results in the following expression of $g(z|\theta)$ for $\theta > \theta_f$ and $z > 0$.

$$g(z|\theta) = g(z_f|\theta_f)e^{\theta-\theta_f} \tag{39}$$

(z_f, θ_f) are the points in Figure 3, where the solid line connects with the dashed line of each characteristic. It is highly desirable to express $g(z|\theta)$ only in terms of z and θ. This means that expressions for z_f and θ_f have to be defined. First, a dimensionless feeding time θ_f is defined. At time t_f, all material has been fed to the dryer

$$t_f = \frac{M_T(t) - M_T(0)}{w^0} \tag{40}$$

At t_f, Eq. 28e becomes

$$t_f = \frac{M_T(0)\alpha}{w^0(\alpha-1)}\left(e^{(\alpha-1)\theta_f} - 1\right) \tag{41}$$

This equation is combined with Eq. 40 to give

$$\frac{M_{total} - M_T(0)}{M_T(0)} = \frac{\alpha}{\alpha-1}\left(e^{(\alpha-1)\theta_f} - 1\right) \tag{42}$$

Rearranging Eq. 42 results in the following expression for θ_f

$$\theta_f = \frac{1}{\alpha-1}\ln\left(1 + \frac{M_T(t) - M_T(0)}{M_T(0)}\frac{\alpha-1}{\alpha}\right) \tag{43}$$

An expression for z_f is derived from the slope of a characteristic $dz/d\theta = 1 - z$

$$\frac{1-z}{z-z_f} = e^{-(\theta-\theta_f)} \tag{44a}$$

$$z_f(z|\theta) = 1 - (1-z)e^{\theta-\theta_f} \tag{44b}$$

The expressions of $z_f(z|\theta)$ and θ_f are used to solve the equation for $g(z|\theta)$ during drying (Eq. 39). Together with Eq. 38, a complete set of equations and boundary conditions has been defined for $g(z|\theta)$ (see Table 1). However, it is more interesting to know the evolution of $f(x|t)$ during the drying process. Using Eq. 10, it is very straightforward to convert $g(z|\theta)$ to $f(x|t)$ by rearrangement.

Finding expressions for $\mu_0(\theta)$, $\mu_1(\theta)$ and $t(\theta)$ during the drying stage

After feeding no new particles enter the dryer, that is, the mass of particles in the dryer $M_T(t)$ remains constant ($M_T(t_f) = M_T(t_{end})$). Previously, it was assumed that particles never reached their equilibrium moisture content ($x < x^*$). Therefore, the dimensionless mass of wet particles in the dryer $\mu_0(\theta)$ was also constant: $\mu_0(\theta) = \mu_0(\theta_f)$ for $\theta > \theta_f$.

During the drying stage $\alpha = 0$ ($\theta > \theta_f$). This transforms Eq. 24 into

$$\frac{d(\mu_0(\theta) - \mu_1(\theta))}{d\theta} = -(\mu_0(\theta) - \mu_1(\theta)) \tag{45}$$

This ordinary differential equation can be solved with θ_f as the initial boundary conditions. This results in

$$\mu_0(\theta) - \mu_1(\theta) = (\mu_0(\theta_f) - \mu_1(\theta_f))e^{-(\theta-\theta_f)} \tag{46a}$$

$$= e^{(\alpha-1)\theta_f}e^{-(\theta-\theta_f)} \tag{46b}$$

The latter equation is obtained by substituting ($\mu_0(\theta_f) - \mu_1(\theta_f)$) with Eq. 24 in which $\theta = \theta_f$. Simplifying Eq. 46b gives an expression for $\mu_1(\theta)$ during the drying stage

$$\mu_1(\theta) = \mu_0(\theta_f) - e^{(\alpha\theta_f-\theta)} \tag{47}$$

The total process time t_{end} is calculated from the overall mass balance of solvent in the dryer. Therefore, the same equation can be applied as defined in the previous article Nitert and Hounslow (submitted)

$$t_{end} = \frac{(x^0 - x^*)(M_T(0) + M_{feed})}{\chi} \tag{48a}$$

$$= t_{ini} + \alpha t_f \tag{48b}$$

where $M_{feed} = w^0 t_f$. θ_{end} can be obtained by incorporating Eq. 41 into Eq. 48 and combining it With Eq. 9c.

$$\theta_{end} = \theta_f - \ln\left(1 - \frac{t_{end} - t_f}{t_{ini}}e^{-(\alpha-1)\theta_f}\right) \tag{49}$$

For the conversion from θ to t during drying, the same basis is used as for feeding.

$$\frac{d\theta}{dt} = \frac{\chi}{M_T(0)(\mu_0(\theta) - \mu_1(\theta))(x^0 - x^*)}$$

This expression can be simplified using Eq. 47 and the expression for t_{ini}.

$$\frac{d\theta}{dt} = \frac{e^{-(\alpha\theta_f-\theta)}}{t_{ini}} \tag{50}$$

Equation 50 can be solved by integrating θ and t using, respectively, θ_f and t_f as lower limits. This results in

$$t(\theta) = t_f + t_{ini}e^{(\alpha-1)\theta_f}\left(1 - e^{\theta_f-\theta}\right) t > t_f \tag{51}$$

Calculating $m_0(t)$, $m_1(t)$ and $\bar{x}_{overall}(t)$

Converting $\mu_0(\theta)$ to $m_0(t)$ is also a straightforward routine when integrating both sides of Eq. 10 which results in

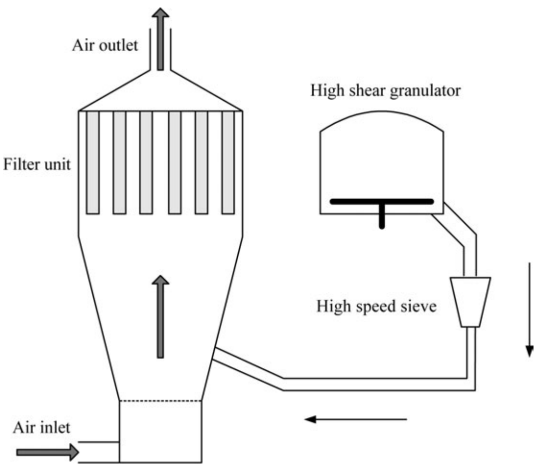

Figure 4. Schematic overview of a semibatch fluidized bed drying process.

Wet granules are pneumatically fed from the high shear granulator, optionally *via* a high speed sieve, to the fluidized bed. The large gray arrows mark the direction of the airflow inside the fluidized bed (adopted from Nitert and Hounslow, submitted).

$$m_0(t) = M_T(0)\mu_0(\theta) \tag{52}$$

The first moment of $f(x|t)$, $m_1(t)$ can be calculated by combining Eqs. 6 and 7. This results in the following expression for $m_1(t)$

$$m_1(t) = \left(\mu_1(\theta) + \frac{x^0}{x^* - x^0}\mu_0(\theta)\right)(x^* - x^0)M_T(0) \tag{53}$$

Nitert and Hounslow (submitted) defined the ratio of $m_1(t)$ and $m_0(t)$ as the average moisture content in wet particles. In this article, no distinction is made between wet and dry particles. Therefore, $m_1(t)/m_0(t)$ gives the overall average moisture content in the dryer, $\bar{x}_{\text{overall}}(t)$. Another way of calculating $\bar{x}_{\text{overall}}(t)$ is by calculating the overall average dryness of particles in the dryer

$$\bar{z}(\theta) = \frac{\mu_1(\theta)}{\mu_0(\theta)} \tag{54}$$

$\bar{x}_{\text{overall}}(t)$ is then calculated using Eq. 7 and replacing z and x with, respectively, $\bar{z}(\theta)$ and $\bar{x}_{\text{overall}}(t)$.

Simulation Studies

Input parameters

In the previous section, a model is presented for falling particle drying rate. This model is going to be tested using the same drying system as presented in the previous article of Nitert and Hounslow, (submitted). This hypothetical system is a semi-batch fluidized bed dryer consisting of an inlet stream of wet particles called granules and an inlet stream of hot air that acts as carrier for the evaporated moisture. Figure 4 shows a schematic overview of this process. The input parameters for the model are similar to those presented in the previous article as this makes comparison between constant and falling drying rate possible. Table 2 shows the values of α for different settings of w^0 and χ. Table 3 shows the parameters that were kept constant throughout all experiments.

Table 2. Calculated α-Values for Different Operating Settings of Drying Rate (χ) and Feed Rate (w^0)

χ (in kg/s)=	0.0025	0.00375	0.005
w^0 = 0.067 kg/s	2.66667	1.77778	1.33333
w^0 = 0.100 kg/s	4	2.66667	2

Table 3. Input Parameters that were Kept Constant During Simulation Experiments

Parameter	Setting
Initial mass of wet granules ($M_T(0)$)	1 kg dry solid
Total batch loading (M_{total})	100 kg dry solid
Initial moisture content (x^0)	0.15 kg H_2O/kg dry solid
Final moisture content (x^*)	0.05 kg H_2O/kg dry solid

Results and Discussion

Results falling drying rate Figures 5a and 5b show the simulation results of, respectively, $\mu_0(\theta)$ and $\mu_1(\theta)$. It is noted that the line of feed rate 0.067 kg/s and drying rate 0.0025 kg/s (solid gray line) coincides with the line of feed rate 0.100 kg/s and drying rate 0.00375 kg/s (large dashed black line). This is also expected as their α-values are the same (see Table 2). Equations 26b and 27c show that $\mu_0(\theta)$ and $\mu_1(\theta)$ only depend on α and θ. In both figures it can clearly be observed that a higher feed rate and similar drying rate, that is, a higher value of α, results in a higher value of $\mu_0(\theta)$ and $\mu_1(\theta)$. This is expected from Eqs. 26b and 27c.

The corresponding unnormalized moments, $m_0(t)$ and $m_1(t)$, are, respectively, total mass of particles in the dryer and total amount of H_2O in particles. Therefore, more information about the state of particles in the drying system as a function of time t is obtained when plotting the unnormalized moments against time t. These unnormalized moments are obtained using Eqs. 52 and 53 for the conversion of, respectively, $\mu_0(\theta)$ and $\mu_1(\theta)$. The conversion from θ to t is performed using Eqs. 28e and 51 for, respectively, the feeding stage and drying stage.

Figure 6a shows the result of $m_0(t)$ for both feed rates and three drying rates for each feed rate. A higher feed rate results in a faster increase of $m_0(t)$, which is also expected from the mass balance of particles in the dryer (see Eq. 5). The maximum amount of 100 kg corresponds to the total batch size as defined in Table 3. For each feed rate, the results of the drying rates are similar. This is expected as

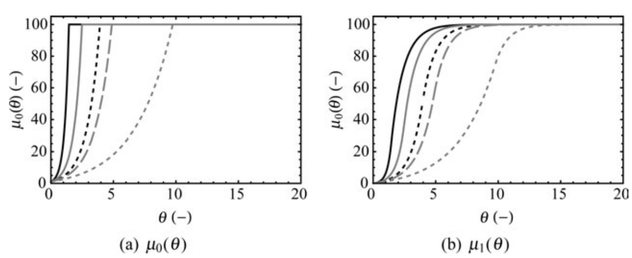

Figure 5. Dimensionless mass of granules in the dryer, $\mu_0(\theta)$ (left graph) and dimensionless dryness among granules in the dryer, $\mu_1(\theta)$ (right graph), as a function of dimensionless time, θ.

Two feed rates are presented in each graph: 0.067 kg/s (gray lines) and 0.100 kg/s (black lines). For each feed rate, three drying rates are presented: 0.0025 kg/s (- - -), 0.00375 kg/s (– – –), 0.005 kg/s (—).

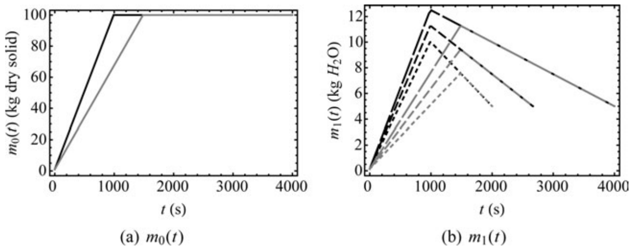

Figure 6. Mass of granules in the dryer, $m_0(t)$ (left graph) and mass of H_2O in granules in the dryer, $m_1(t)$ (right graph), as a function of time, t.

Two feed rates are presented in each graph: 0.067 kg/s (gray lines) and 0.100 kg/s (black lines). For each feed rate, three drying rates are presented: 0.0025 kg/s (- - -), 0.00375 kg/s (– – –), 0.005 kg/s (—).

$m_0(t)$ does not depend on the drying rate (see Eq. 5). Figure 6b shows the results of $m_1(t)$. A clear effect of both feed rate and drying rate is observed. At $t = 0$, $m_1(t)$ is only determined by particles of the initial charge. During feeding ($t > 0$), solvent is added to the dryer by addition of wet particles at rate w^0 and with moisture content x^0. Simultaneously, solvent is also removed at rate χ. The influence of these parameters on $m_1(t)$ is defined by Eq. 6. A higher feed rate results in a larger contribution of feed rate to $m_1(t)$ when $w^0x^0 > \chi$, that is, the effect of drying rate diminishes. This is also shown in Figure 6b by the larger difference in slopes during feeding at a lower feed rate. At each drying rate, the slopes of both feed rates are similar after feeding ($t > t_f$). This can be explained as follows: the total batch size, initial charge, drying rate, and x^0 remain constant during feeding. In other words, the total amount of solvent fed to the dryer is the same for both feed rates. The only difference is how fast this solvent is fed to the dryer. As χ is constant, the build up of solvent during feeding is larger for a higher feed rate. However, after feeding the amount of solvent is reduced according to the same rate χ. Therefore, at $t = t_f$ of feed rate 0.067 kg/s, the lines of both feed rates merge. Although average properties give a good indication how the process is progressing, sometimes it is equally important to obtain knowledge of the evolution of moisture content distribution among particles during the drying process. Figure 7 presents the results of $f(x|t)$ for both feed rates (with a drying rate of 0.00375 kg/s). A clear difference in shape of the curves is seen for the two feed rates. This is attributed to the behavior of $\dot{x}(t)$ during drying.

$\dot{x}(t)$ is determined by $k(t)$ according to Eq. 3. $k(t)$ is calculated from the drying rate χ, α, and θ using Eq. 25. Figure 8 presents the result of $k(t)$ for both feed rates (with $\chi = 0.00375$ kg/s). It is assumed that χ is constant during the drying process. Therefore, a higher value of w^0 results in a higher α in Eq. 25, and consequently, a faster decrease of $k(t)$ and $\dot{x}(t)$. When feeding at a higher rate, the particles dry less fast. It is trivial that this also alters the shape of the moisture content distribution $f(x|t)$.

Figure 9 shows the overall average moisture content among particles in the dryer. A higher feed rate results in a higher value of $\bar{x}_{\text{overall}}(t)$ and after feeding the slopes of each feed rate merge at $t = t_f$ of the slower feed rate. This result matches the result of $m_1(t)$ and is explained by a larger build up of solvent among particles in the dryer at a higher feed rate. This effect is more pronounced at higher drying rates than lower drying rates as χ diminishes the effect of w^0 on

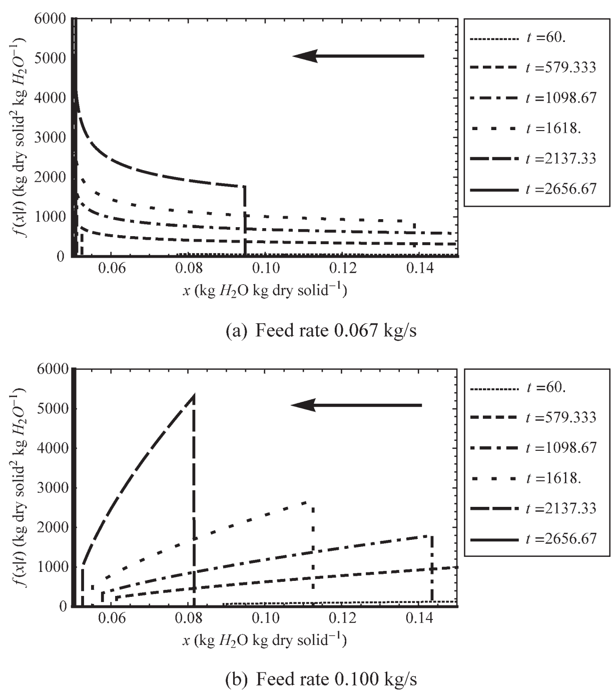

Figure 7. Mass density distribution $f(x|t)$ describing the evolution of moisture content distribution as a function of time for feed rate 0.067 kg/s (top graph) and 0.100 kg/s (bottom graph).

The drying rate was 0.00375 kg/s for both feed rates. The arrows indicate the direction of movement as a function of time t.

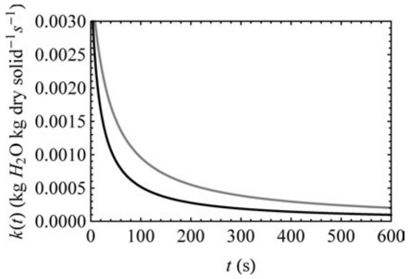

Figure 8. Calculated drying rate constant $k(t)$ for feed rate 0.067 kg/s (gray line) and 0.100 kg/s (black line) at $\chi = 0.00375$ *kg/s*.

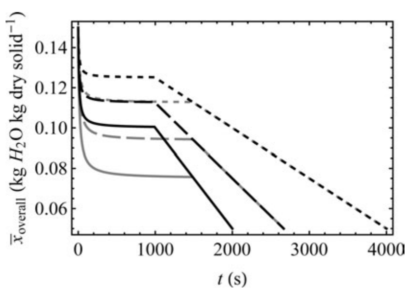

Figure 9. Combined plots of overall average moisture content of particles for feed rate 0.067 kg/s (gray lines) and 0.100 kg/s (black lines).

For each feed rate, three drying rates are presented: 0.0025 kg/s (- - -), 0.00375 kg/s (– – –), 0.005 kg/s (—).

$m_1(t)$ according to Eq. 6. Changing χ with high feed rates has a less pronounced effect than changing χ at low feed rates.

Comparison with constant particle drying rate

The appropriate parameters should be chosen to compare constant and falling particle drying rate. The model for constant particle drying rate as described in the previous article (Nitert and Hounslow, submitted) classifies and divides particles as wet ($x^0 \leq x < x^*$) or dry ($x = x^*$). The model described in this article does not make this distinction. It is obvious that parameters are chosen that exist in both models and do not depend on this classification of particles.

For this reason, moisture content distributions obtained from both models can not be compared directly as their basis is different. Distributions obtained from constant particle drying rate do not include particles that are fully dry, that is, when a particle is dry ($x = x^*$), it moves from the wet particles population to the dry particles population. The moisture content distribution of wet particles is only described. For falling particle drying rate, there is no transfer of particles as they do not become fully dry. The moisture content distribution describes all particles in the dryer. This explains why other parameters than moisture content distribution have to be defined to compare both models. The dimensionless degree of wetness is calculated from $\mu_0(\theta)$, and $\mu_1(\theta)$ and is determined by the amount of wet particles in the dryer, that is, dry particles are not taken into consideration. Therefore, it is a useful parameter for comparison of both models. As an example, the results of feed rate 0.067 kg/s are compared for both models in Figure 10.

There is no difference between constant and falling drying rate when the same settings for w^0 and χ are used. This is also expected as the dimensionless wetness is a relative average property of wet particles that depends on the mass balance of solvent and particles in the drying system.

The overall average moisture content $\bar{x}_{\text{overall}}$ is calculated differently for both models. Nevertheless, their physical meaning is exactly the same, namely the average moisture content of wet and dry particles in the dryer. Figure 11 shows the overall average moisture content. No differences are observed between the two models. This is also expected as this parameter is determined by the mass balance of solvent and total amount of solid in the dryer. These parameters are solely calculated from the input parameters given in Tables 2 and 3 which are constant during the process. In other words, it is not determined by the individual particle drying rate $\dot{x}(t)$.

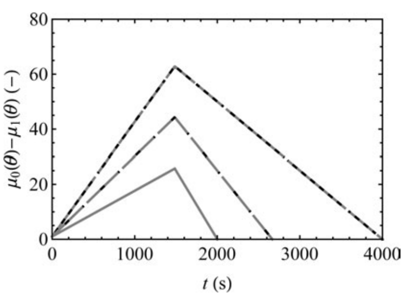

Figure 10. Dimensionless degree of wetness in the dryer presented for feed rate 0.067 kg/s.

Three drying rates are presented: 0.0025 kg/s (- - -), 0.00375 kg/s (– – –), 0.005 kg/s (—). A comparison is made between constant particle drying rate (gray lines) and falling particle drying rate (black lines).

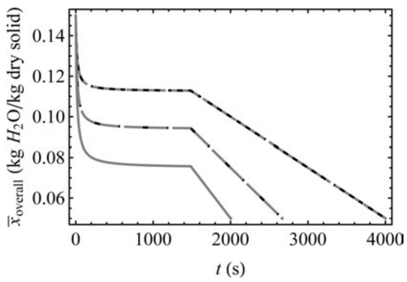

Figure 11. Overall average moisture content of particles in the dryer presented for feed rate 0.067 kg/s.

Three drying rates are presented: 0.0025 kg/s (- - -), 0.00375 kg/s (– – –), 0.005 kg/s (—). A comparison is made between constant particle drying rate (gray lines) and falling particle drying rate (black lines).

All graphs show a sharp drop in average overall moisture content. This is caused by the initial charge in the dryer. This charge contains 100 g of free moisture ((0.15 − 0.05)asterisk 1 = 0.1 kg solvent). This moisture will be removed with an overall drying rate of $\chi = 0.0025-0.005$ kg/s, that is, it will take 20–40 s to remove this moisture. The timescale is much larger which is why it is depicted as a sharp drop.

Conclusions

In this article a comprehensive population balance model is presented for a semi-batch drying process that includes falling particle drying rate. The model is solved analytically using the "method of characteristics" and "method of moments". Simulation experiments give meaningful results for different values of feed rate w^0 and χ. Moreover, falling particle drying rate gives similar results as constant particle drying rate for the overall average moisture content. This is attributed to the mass balance and assumption of constant overall drying rate χ. The previous article[3] and this article give a complete description of a semibatch process for, respectively, constant and falling particle drying rate. However, the authors recognize that the assumption of constant χ is not valid from an overall critical moisture content onward (see Nitert and Hounslow, submitted for more details). In many real industrial drying systems, a falling overall drying rate is observed. The exit gas humidity is no longer saturated. Therefore, two routes for further investigation are suggested. The first route would be experimental testing of the model described in this article. This would benchmark how well the model describes reality. A second route should focus on investigating the possibility of defining a population balance equation for a falling overall drying that is, $\chi(x)$.

Notation

f = mass density function particles ($x > x^*$), kg dry solid^2 kg solvent^{-1}
g = dimensionless mass density distribution
h = relative drying rate
k = falling drying rate constant, kg solvent kg dry solid^{-1} s^{-1}
m_0 = mass of particles in dryer, kg dry solid
m_1 = mass of moisture of particles in dryer, kg solvent
M_T = total mass of particles, kg dry solid
M_T^* = total mass of dry particles, kg dry solid
t = time, s
t_{ini} = drying time initial charge, s
w^0 = feed rate of wet particles, kg s^{-1}

$\dot{x}$ = single particle drying rate, kg solvent kg dry solid^{-1} s^{-1}
x^* = equilibrium moisture content, kg solvent kg dry solid^{-1}
x = particle moisture content, kg solvent kg dry solid^{-1}
x^0 = initial moisture content, kg solvent kg dry solid^{-1}
x^{cr} = critical moisture content, kg solvent kg dry solid^{-1}
z = dimensionless dryness
z_0 = value of z for a characteristic starting at $\theta = \theta_0$
$\dot{z}$ = dimensionless drying rate, s^{-1}

Subscripts

b = bulk
end = end of batch
f = end of feeding stage

Greek letters

α = dimensionless ratio between feed and removal rate of solvent
χ = solvent removal rate, kg s^{-1}
μ_0 = total dimensionless mass of particles in dryer
μ_1 = total dryness of particles in dryer
Φ = relative moisture content
θ = dimensionless time
θ_0 = value of θ for a characteristic starting at $z = 0$

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Appendix: Confirmation Analytical Solution for $g(z|\theta)$

In this appendix, the result of Eq. 37b is confirmed using separation of variables and solving the equation of $\mu_0(\theta)$. When $g(z|\theta)$ is written as the product of a function $p(z)$ and $q(\theta)$, the population balance Eq. 32 can be written as

$$p(z)q'(\theta) + (1 - z)p'(z)q(\theta) = p(z)q(\theta) \tag{A1}$$

Rearranging gives

$$\frac{q'(\theta)}{q(\theta)} = 1 - (1 - z)\frac{p'(z)}{p(z)} = C \tag{A2}$$

Solving this equation by integration results in

$$\ln(q(\theta)) = C\theta + B \tag{A3}$$

where B is an integration constant. The same routine can be performed for $p(z)$

$$\ln(p(z)) = -(1 - C)\ln(1 - z) + A \tag{A4}$$

$$p(z) = A'(1 - z)^{-(1-C)} \tag{A5}$$

This gives for $g(z|\theta)$

$$g(z|\theta) = A'B'(1 - z)^{-(1-C)}e^{C\theta} \tag{A6}$$

The next step is finding expressions for A', B', and C. This is performed by comparing Eq. 14 with Eq. 26b

$$\int_0^1 A'B'(1 - z)^{-(1-C)}e^{C\theta}\,dz = \frac{(\alpha e^{(\alpha-1)\theta} - 1)}{\alpha - 1} \tag{A7}$$

The result on the righthand side is only obtained when $C = \alpha - 1$. This transforms Eq. 60 into

$$\int_0^1 A'B'(1 - z)^{\alpha-2}e^{(\alpha-1)\theta}\,dz = \frac{(\alpha e^{(\alpha-1)\theta} - 1)}{\alpha - 1} \tag{A8}$$

$$\left[\frac{A'B'}{\alpha - 1}(1 - z)^{\alpha-1}e^{(\alpha-1)\theta} - 1\right]_0^1 = \frac{(\alpha e^{(\alpha-1)\theta} - 1)}{\alpha - 1} \tag{A9}$$

$$A'B'e^{(\alpha-1)\theta} = \alpha e^{(\alpha-1)\theta} \tag{A10}$$

$$A'B' = \alpha \tag{A11}$$

This last result gives

$$g(z|\theta) = \alpha(1 - z)^{\alpha-2}e^{(\alpha-1)\theta}$$

which corresponds with Eq. 37b.

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