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Letter to the editor

Comments to “Analysis of constant rate period of spray drying of slurry” by Liang et al., 2001

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Abstract

In the study by Liang et al. [2001. Analysis of constant rate period of spray drying of slurry. Chemical Engineering Science 56, 2205–2213] the Darcy flow of liquid through a pore system of primary particles to the surface of a slurry droplet was applied for the constant rate period. Steep primary particle concentration gradients inside $\sim 25\,\mu\text{m}$ droplets with a primary particle size of $0.2\,\mu\text{m}$ were observed. Unfortunately, the boundary condition at the droplet surface for the parabolic second-order PDE did not conserve the solid mass in the droplet, and the plots for the primary particle concentration profiles in the droplets were incorrect. In this letter we derive the correct boundary condition equation. Furthermore, we show that the primary particle concentration profiles inside the droplets are flat when the primary particles have a size of $0.2\,\mu\text{m}$. We conclude that the model presented by Liang et al. is unable to predict the formation of hollow particles.

Keywords: Spray drying; Slurry droplet; Modeling; Constant rate period; Darcy flow

The paper “Analysis of constant rate period of spray drying of slurry” by Liang, H., Shinohara, K., Minoshima, H., Matsushima, K., (2001). Chemical Engineering Science, 56, 2205–2213 presents a very interesting approach to predicting the morphology of spray dried slurry droplets.

In the paper a model for the transport processes occurring in an evaporating slurry droplet during the constant rate period (CRP) is developed. The evaporation of solvent from the droplet surface is assumed to be accompanied by the generation of a liquid-filled pore system of primary particles, which creates a concentration gradient of solid particles in the droplet. The liquid flow to the droplet surface through this pore system obeys Darcy’s law. The model yields valuable information about the effects of primary particle size, initial solid content, and drying air temperature on the concentration gradient of solid particles in the droplet and thereby on the final particle morphology, i.e., whether the particle becomes hollow or solid.

We believe that some clarifying remarks on and questions to the model and results presented in the paper need to be stated. Our concern goes to (1) the boundary condition equation set up for the droplet surface and (2) the fact that the modelling results presented in the paper do not appear to be consistent with the model presented.

1. Model in Liang et al., 2001

The model proposed by Liang et al. to describe the transport of liquid towards the surface of the drying droplet as well as the evaporation of solvent from the droplet surface is briefly presented: the parabolic second order partial differential equation (PDE) in Eq. (1) (Eq. (9) from Liang et al.) for transport of liquid towards the surface of the drying droplet is derived on the basis of Darcy’s law for fluid flow in porous media:

$$k_1 \frac{\partial \phi}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left[\frac{r^2 (1-\phi)}{\phi^2} \frac{\partial \phi}{\partial r} \right]. \qquad (1)$$

where $k_1 = 30\mu/\sigma_{LV} d_p \cos\theta$. For reasons of clarity, we will write the solid volume fraction in the droplet simply as ϕ and not $\phi(r,t)$, and the droplet radius as R and not $R(t)$ throughout this paper, unless it is necessary in order to follow the derivations.

Two boundary conditions (BC) and a set of initial conditions (IC) are required to solve the PDE in Eq. (1). The BC1 at the

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droplet surface ($r = R$) (Eq. (12) in the paper) is given as:

$$R^2 \left.\frac{\partial \phi}{\partial r}\right|_{r=R} = \frac{-15\mu}{4\pi d_p \sigma_{LV} \rho_L \cos\theta} \frac{\phi(R)^2}{1-\phi(R)} \frac{\mathrm{d}m}{\mathrm{d}t}. \tag{2}$$

Even though it is not stated explicitly in the paper, a symmetry assumption at the droplet centre ($r = 0$) must be imposed in order to produce the second boundary condition (BC2):

$$\left.\frac{\partial \phi}{\partial r}\right|_{r=0} = 0. \tag{3}$$

The IC at $t = 0$ are given as (Eq. (10) from Liang et al.):

$$\phi(r,0) = \phi_0, \quad R(0) = R_0, \quad T_L(0) = T_0, \quad m(0) = m_0. \tag{4}$$

where the initial droplet mass (m_0) and temperature (T_0) are required to solve the mass and energy balance in Eq. (5) and (6), respectively.

The mass balance for solvent evaporation is given as (Eq. (1) from Liang et al.):

$$\frac{\mathrm{d}m}{\mathrm{d}t} = -4\pi R \rho_g D_{AB}(W_{AR} - W_{Af}). \tag{5}$$

where W_{Af} is the mass fraction of water vapour in the drying gas far from the droplet and assumed to be zero throughout the drying process (as assumed by Liang et al.).

The energy balance is given as (Eq. (2) from Liang et al.):

$$4\pi RK(T_a - T_L) + \lambda \frac{\mathrm{d}m}{\mathrm{d}t} = (m_s S_s + m_l S_l)\frac{\mathrm{d}T_L}{\mathrm{d}t}. \tag{6}$$

Combining the mass and energy equations gives the rate of evaporation from the droplet surface.

2. Comment no. 1

The derivation of the BC at the droplet surface (BC1–Eq. (2)) is not explained in detail in the paper by Liang et al. and it appears to be incorrect.

The correct BC1 at the droplet surface can be derived from the assumption that the mass of solid material in the droplet is constant during the drying, i.e.:

$$4\pi\rho_s \int_0^{R(t)} r^2 \phi(r,t)\,\mathrm{d}r = \text{constant}. \tag{7}$$

Differentiating Eq. (7) with respect to time, t and applying Leibnitz' rule for differentiation (Bird et al., 1960) gives

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\int_0^{R(t)} r^2 \phi(r,t)\,\mathrm{d}r\right) = 0 = \int_0^{R(t)} r^2 \frac{\partial \phi(r,t)}{\partial t}\,\mathrm{d}r + R^2 \phi(R) \frac{\mathrm{d}R}{\mathrm{d}t}. \tag{8}$$

The first term on the RHS of Eq. (8) can be found by integrating Eq. (1) over the radius of the droplet:

$$\int_0^{R(t)} r^2 \frac{\partial \phi(r,t)}{\partial t}\,\mathrm{d}r = \frac{1}{k_1}\left[\frac{r^2(1-\phi(r,t))}{\phi(r,t)^2} \frac{\partial \phi(r,t)}{\partial r}\right]_0^{R(t)} = \frac{1}{k_1} \frac{1-\phi(R)}{\phi(R)^2} R^2 \left.\frac{\partial \phi}{\partial r}\right|_{r=R}. \tag{9}$$

Eq. (8) can now be written as

$$0 = \frac{1}{k_1} \frac{1-\phi(R)}{\phi(R)^2} R^2 \left.\frac{\partial \phi}{\partial r}\right|_{r=R} + R^2 \phi(R) \frac{\mathrm{d}R}{\mathrm{d}t}. \tag{10}$$

Eq. (10) can be rearranged to give the correct boundary condition (BC1):

$$\left.\frac{\partial \phi}{\partial r}\right|_{r=R} = -k_1 \frac{\phi(R)^3}{1-\phi(R)} \frac{\mathrm{d}R}{\mathrm{d}t}. \tag{11}$$

From the assumption stated in Eq. (7) it follows that only solvent evaporates from the droplet, i.e., the drying rate can be expressed as

$$\frac{\mathrm{d}m}{\mathrm{d}t} = 4\pi\rho_L R^2 \frac{\mathrm{d}R}{\mathrm{d}t}. \tag{12}$$

Now, the correct BC1 at the droplet surface in Eq. (11) can be rewritten to give

$$R^2 \left.\frac{\partial \phi}{\partial r}\right|_{r=R} = \frac{-30\mu}{4\pi d_p \sigma_{LV} \rho_L \cos\theta} \frac{\phi(R)^3}{1-\phi(R)} \frac{\mathrm{d}m}{\mathrm{d}t}. \tag{13}$$

By comparing the BC1 given by Liang et al. in Eq. (2) to the correct BC1 in Eq. (13) it is clearly seen that Liang et al. have introduced a factor of $\frac{1}{2}$ but is missing a factor $\phi(R)$ on the RHS.

To show the consequences of applying the BC1 proposed by Liang et al. we have solved the model presented in Liang et al. in Eqs. (1)–(6). This is done by introducing the variable transformation $x = r/R(t)$, which secures that the new radial coordinate inside the droplet, x, is kept between 0 and 1 throughout the drying process. Introducing this variable transformation and expanding the PDE in Eq. (1) gives the following PDE:

$$\frac{\partial \phi(x,t)}{\partial t} = \frac{1}{k_1 R^2} \frac{(1-\phi)}{\phi^2}\left[\frac{2}{x} + \frac{\phi - 2}{(1-\phi)\phi}\left(\frac{\partial \phi}{\partial x}\right)^2 + \frac{\partial^2 \phi}{\partial x^2}\right] + \frac{x}{R}\frac{\mathrm{d}R}{\mathrm{d}t}\frac{\partial \phi}{\partial x}. \tag{14}$$

Similarly, the BC1 at the droplet surface ($x = 1$) given by Liang et al. (Eq. (2)) can be transformed to give

$$R \left.\frac{\partial \phi}{\partial x}\right|_{x=1} = \frac{-15\mu}{4\pi d_p \sigma_{LV} \rho_L \cos\theta} \frac{\phi(1)^2}{1-\phi(1)} \frac{\mathrm{d}m}{\mathrm{d}t} \tag{15}$$

and the correct BC1 (Eq. (13)) in transformed variables reads:

$$R \left.\frac{\partial \phi}{\partial x}\right|_{x=1} = \frac{-30\mu}{4\pi d_p \sigma_{LV} \rho_L \cos\theta} \frac{\phi(1)^3}{1-\phi(1)} \frac{\mathrm{d}m}{\mathrm{d}t}. \tag{16}$$

For the sake of completeness the BC at the droplet centre (BC2 from Eq. (3)) and the initial conditions (IC from Eq. (4)) are also given in transformed variables:

$$\text{BC2}: \quad \left.\frac{\partial \phi}{\partial x}\right|_{x=0} = 0. \tag{17}$$

$$\text{IC}: \quad \phi(x,0) = \phi_0, \quad R(0) = R_0, \quad T_L(0) = T_0, \quad m(0) = m_0. \tag{18}$$

Now, the model presented by Liang et al. can be solved by discretising the PDE in Eq. (14) and corresponding BC