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The drying rates of spray freeze drying systems increase through the use of stratified packed bed structures

R. Bruttini [a], A.I. Liapis [b,*]

[a] Criofarma-Freeze Drying Equipment, Strada del Francese 97/2L, 10156 Turin, Italy

[b] Department of Chemical and Biochemical Engineering, Missouri University of Science and Technology, 100 Bertelsmeyer Hall, 1101 North State Street, Rolla, MO 65409-1230, USA

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ABSTRACT

It is shown that when in the spray freeze drying process stratified packed beds are employed where the smallest in size particles are located near the surface of the lower heating plate while the largest particles are occupying the upper part of the stratified packed bed whose outermost surface is in contact with the atmosphere of the drying chamber of the freeze dryer, the duration times of the primary and secondary drying stages can be substantially reduced when compared with those required by a spray freeze drying process which uses a single packed bed formed by particles all having the same particle size and the total amount of solids in the packed bed is the same as that in the stratified packed bed.

1. Introduction

Spray freeze drying (SFD) provides a separation process that can produce porous particles whose physical and chemical characteristics are considered to be ideal for use in pulmonary drug delivery, the production of powders for epidermal immunization, in the processing of low water soluble drugs, the preparation of particles for microencapsulation, the processing of particles for use in chemical catalysis and biocatalysis, the preparation of nanopowders and ceramic electronic parts, in the cryogenic processing of chemicals and powder based materials, in the production of high value porous food particles, and in the synthesis of Li-ion battery cathode materials [1–7]. SFD involves the packing of frozen particles in containers and, thus, a packed bed [2,3,7–13] of frozen particles is formed which has a porous structure and makes the frozen region of the material to be unsaturated during primary drying [2] because the space of the frozen region formed by the packed frozen particles is partially filled with gas (inert gas and solvent (e.g., water) vapor) which moves through the pores of the unsaturated porous frozen region by convection, Knudsen diffusion, and bulk diffusion during primary drying, as per Fig. 1a. During secondary drying, the bound (sorbed) solvent (e.g., water) is desorbed from the surface of the pores of the particles being dried and is transported through the pores of the porous structure of the particles as well as through the pore space of the packed bed by convection, Knudsen diffusion, and bulk diffusion [2,3].

It has been shown theoretically by Liapis and Bruttini [2,3] that the longer drying times required in SFD when compared to the drying times required in classical freeze drying, are mainly due to the reduced heat and mass transfer capabilities of the porous packed beds formed by the packed frozen particles and can also lead to the formation of a secondary dried layer near the surface of the lower heating plate during the primary drying stage, as per Fig. 2. The formation of this secondary dried layer has been confirmed experimentally [4,5] and contributes to the deterioration of the drying rate. Furthermore, Liapis and Bruttini [2] have shown that in the SFD process the drying rate during the primary drying stage increases as (i) the product height decreases, (ii) the particle diameter increases, and (iii) the value of the packing porosity increases. Liapis and Bruttini [2] have also presented the physicochemical mechanisms which provide the reasons for obtaining the results in items (i)–(iii) above.

It is important to indicate here that for the SFD processing of pharmaceutical products and ceramic powders the magnitude of the ratio of the size of the packed bed of particles to the particle diameter is most often larger than 1000 and the SFD model of Liapis and Bruttini [2] was found to describe the physics and the dynamic behavior of the SFD process satisfactorily and its predictions are consistent with and describe appropriately the experimental results and observations published in the literature [4–6]. The SFD process could also be used in the production of high value porous food particles. But the food particles have diameters of

* Corresponding author. Tel.: +1 573 341 4414; fax: +1 573 341 4377.
E-mail addresses: criofarma@tin.it (R. Bruttini), ail@mst.edu (A.I. Liapis).

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Nomenclature

$C_{01,i}$	Darcy flow permeability constant dependent upon the pore structure of fixed-bed i of a stratified packed bed (m^2)
$C_{1,i}$	Knudsen diffusion constant dependent upon the pore structure of fixed-bed i of a stratified packed bed (m)
$C_{2,i}$	constant dependent upon the pore structure of fixed-bed i of a stratified packed bed and giving the ratio of bulk diffusion coefficient within the pore structure of fixed-bed i to the free gas molecular diffusion coefficient (dimensionless)
$d_{pore,i}$	mean pore diameter of porous fixed-bed i of a stratified packed bed (m)
$D_{w,in,i}$	free gas molecular diffusion coefficient in a binary mixture of water vapor and inert gas ($m^2\ s^{-1}$)
$K_{in,i}$	Knudsen diffusion coefficient of inert gas in fixed-bed i of a stratified packed bed ($m^2\ s^{-1}$)
$K_{m,i}$	mean Knudsen diffusivity for the binary mixture of water vapor and inert gas (Eq. (6)) in fixed-bed i of a stratified packed bed ($m^2\ s^{-1}$)
$K_{w,i}$	Knudsen diffusion coefficient of water vapor in fixed-bed i of a stratified packed bed ($m^2\ s^{-1}$)
L	length (height) of packed bed of particles in tray (m)
L_i	length (height) of fixed-bed i in a stratified packed bed (m)
M_{in}	molecular weight of inert gas (kg/kmol)
M_w	molecular weight of water vapor (kg/kmol)
N	total number of sections (fixed-beds) in a stratified packed bed (dimensionless)
$N_{w,d}$	water vapor mass flux in porous dried region (Fig. 1c) of particle ((kg) $m^{-2}\ s^{-1}$)
$N_{w,pb,i}$	water vapor mass flux in fixed-bed i of a stratified packed bed ((kg) $m^{-2}\ s^{-1}$)
$N_{w,pb,j}$	water vapor mass flux in region j(j = I, II, III) of packed bed ((kg) $m^{-2}\ s^{-1}$)
P_i	total pressure in fixed-bed i of a stratified packed bed (Pa)
r	space coordinate of radial distance (Fig. 1c) in particle (m)
$R(t)$	position of moving sublimation interface (Fig. 1c) in particle (m)
R_g	ideal gas constant (J/K mol)
$R_{p,i}$	radius of particles in fixed-bed i of a stratified packed bed (m)
t	time (s)
$t_{pr,d}$	duration time of the primary drying stage (s)
$t_{se,d}$	duration time of the secondary drying stage (s)
V_b	total volume (voids and particles) of a packed bed of length L (m^3)
V_i	total volume (voids and particles) of fixed-bed i in a stratified packed bed of length L (m^3)
x	space coordinate along the length of a packed bed (m)
$X(t)$	time varying position along x of the moving interface (boundary) separating regions I and II of packed bed (Figs. 1a and 2) of packed bed (m)
$X_1(t)$	time varying position along x of the moving interface (boundary) separating regions II and III (Fig. 2) of packed bed (m)
$y_{in,i}$	mole fraction of inert gas in fixed-bed i of a stratified packed bed (dimensionless)
$y_{w,i}$	mole fraction of water vapor in fixed-bed i of a stratified packed bed (dimensionless)

Greek letters

α_i	L_i/L_{i+1} for $i = 1, 2, 3, \ldots, N-1$ (dimensionless)
β_i	$R_{p,i}/R_{p,i+1}$ for $i = 1, 2, 3, \ldots, N-1$ (dimensionless)
γ_i	fraction ($\gamma_i = V_i/V_b$) of the total volume of a packed bed occupied by fixed-bed i (dimensionless)
ε_p	particle porosity (dimensionless)
ε_{pb}	porosity of packed bed (dimensionless)
$\varepsilon_{pb,i}$	porosity of fixed-bed i of a stratified packed bed (dimensionless)

Subscripts

i	section (fixed-bed) number in a stratified packed bed, $i = 1, 2, 3, \ldots, N$
I	region I of packed bed
II	region II of packed bed
III	region III of packed bed
p	particle
pb	packed bed
1	fixed-bed 1 of a stratified packed bed
N	fixed-bed N of a stratified packed bed

about 50–300 times larger than those of the pharmaceutical and ceramic particles, and, therefore, the magnitude of the ratio of the size of the packed bed of food particles to the diameter of the food particles is often between 15 and 50. Liapis and Bruttini [3] modified their earlier SFD model [2] in order to be able to describe and predict the dynamic behavior of the SFD process for the size scale ratio required by the SFD of food particles, and have also shown that their modified SFD model [3] could describe satisfactorily the dynamic behavior of the SFD process for pharmaceutical as well as food particles. Furthermore it is important to note that while the earlier SFD model of Liapis and Bruttini [2] was constructed for use in the study, design, and control of SFD systems employed in the drying of pharmaceutical particles whose diameters are one to two orders of magnitude smaller than those of food particles, computer simulations indicate [3] that the numerical results obtained from the two SFD mathematical models of Liapis and Bruttini [2,3] are, for all particle purposes, very similar in magnitude when the SFD process is employed in the drying of pharmaceutical particles whose diameters are in the range of 5–90 μm. The particle diameters of the vast majority of pharmaceutical materials that are to be dried by the SFD process have values that are lower than 90 μm, and, therefore, the earlier SFD model of Liapis and Bruttini [2] can be used for the study, design, and control of SFD systems employed in the drying of pharmaceuticals, especially since the numerical solution of the earlier SFD model [2] requires significantly shorter computational times than those required by the modified SFD model [3] which could be used when the SFD process is employed in the drying of food particles.

Li and Liapis [14,15] have recently shown that stratified packed beds comprised from a practically reasonable number of sections provide significantly more efficient dynamic adsorption separations than those obtained in conventional packed beds of the same length, because the stratified packed beds provide larger average external and intraparticle mass transfer and adsorption rates per unit length of packed bed. Liapis and Bruttini [2] showed that in SFD systems, for a given value of the bed porosity, the drying rate increases as the value of the particle diameter increases (see item (ii) above); but if we use in a single packed bed the large in diameter particles in order to obtain a faster drying rate, it is possible that the larger particles could have aerodynamic diameters [2] that are not appropriate for pulmonary drug delivery. Therefore, if we use a stratified packed bed constructed from a desirable