REDUCTION OF FREEZE DRYING TIME BY GEOMETRICAL MODIFICATION OF BULK PLANAR DRYING PRODUCTS

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ABSTRACT
The effect of additional vapor transport paths on reducing the freeze drying time of bulk planar products was numerically investigated. In this study, small holes located in hexagonal lattice points or gaps between slab-shaped products were proposed, to provide additional vapor transport paths to the planar products. The productivity enhancement by the proposed geometric modification was numerically simulated using a fixed grid finite volume analysis program. The applicability of the proposed geometric modification is discussed.

NOMENCLATURE

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>C_swe</td>
<td>sorbed water content (kg water/kg product)</td>
</tr>
<tr>
<td>D_r</td>
<td>relative diffusivity factor (-)</td>
</tr>
<tr>
<td>D_win</td>
<td>binary diffusivity (m²/s)</td>
</tr>
<tr>
<td>D_K</td>
<td>Knudsen diffusivity for species i (m²/s)</td>
</tr>
<tr>
<td>D_mK</td>
<td>mixture Knudsen diffusivity (m²/s)</td>
</tr>
<tr>
<td>f_eq(T)</td>
<td>equilibrium vapor pressure over ice (Pa)</td>
</tr>
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<td>K</td>
<td>absolute permeability (m²)</td>
</tr>
<tr>
<td>k_1</td>
<td>bulk diffusivity for vapor (m²/s)</td>
</tr>
<tr>
<td>k_2,k_4</td>
<td>self diffusivity (m⁴/N-s)</td>
</tr>
<tr>
<td>k_3</td>
<td>bulk diffusivity for inert gas (m²/s)</td>
</tr>
<tr>
<td>s</td>
<td>ice saturation (-)</td>
</tr>
<tr>
<td>y</td>
<td>mole fraction (-)</td>
</tr>
</tbody>
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INTRODUCTION
Freeze drying is an excellent method to dehydrate quality foods with minimal thermal and chemical degradation [1,2]. While sub-zero temperature and vacuum pressure of the freeze drying ensures high quality of final products, it generally results in low productivity with long drying time and high operation cost. Thus, the problem of low productivity should be solved in order for efficient use of freeze drying in food processing.

Many researchers have sought for optimal operation policies that results in shortest drying time while meets all quality criteria, e.g., melting temperature, scorch temperature [1,3,4]. Alternative freeze drying techniques, such as microwave freeze drying and atmospheric freeze drying, have been also studied to reduce the drying time or operation costs [5]. In this study, the geometrical modification of planar products to allow faster vapor transport was numerically investigated to reduce the freeze drying...
time. The holes or slits introduced to planar products are expected to reduce average length of vapor transport from interior towards surface of the products and also increases the area of sublimation front.

The freeze drying problems considered in this study result in complex sublimation front shape, different from conventional tray or vial freeze drying problems. For accurate simulation of the freeze drying problems, a fixed grid finite volume analysis code have been developed by the authors [6]. Instead of tracking sublimation fronts [7-10], a variable was introduced to measure volume content of ice. Then the ice fraction was used to define domain change, to determine properties, and to consider sublimation.

**ANALYSIS MODEL**

The freeze drying processes of planar products, with and without holes and slits, are described in Fig. 1, where each planar products are divided into a dried region (white) and a frozen region (gray) by a sublimation interface (dashed line). During the primary drying stage, moisture (or ice) in planar products sublimates under vacuum, and resulting water vapor $\dot{m}_w$ diffuses through pores to exit the products. The required energy for the sublimation is mainly provided by conduction at the bottom surface $q_b$ and radiation at the top surface $q_r$ from heating plates (heating shelves).

Fig. 1 also indicates that the presence of holes and slits significantly alters the configuration of the sublimation interface. In conventional freeze drying of planar products, the configuration of the sublimation interface always remains planar as shown in Fig 1(a), and the whole process can be solved as a one-dimensional problem. However, with holes and slits, the one-dimensionality in the process breaks due to the curved sublimation interface which forms beneath the surfaces open to the drying chamber.

**MATHEMATICAL FORMULATION**

**Volume-Averaged Properties:** Before derivation of the governing equations, the ice saturation should be defined first. The ice saturation $s$ is the fraction of pore volume occupied by ice

$$s = \frac{V_{ice}}{V_{pore}} = \frac{V_{ice}}{V} = \frac{\varepsilon_{ice}}{\varepsilon}. \quad (1)$$

As shown in Fig. 2, each volume cell is defined as dried, frozen or sublimation cell according to its ice saturations during calculation. For example, $s = 1$ means that pore space is completely filled with ice (frozen cell) and thus there is no room for mass transport. Likewise, $s = 0$ means pore space is free of ice (dried cell). And the sublimation cells satisfy $0 < s \leq 1$, indicating the existence of ice front.
Then, volume-averaged properties required in the fixed grid calculation should be defined. From the simplified representations for microscale partition of ice and matrix, shown in Fig. 2, volume-averaged density is defined as

$$\rho = s \rho_{\nu} + (1-s) \rho_{\nu e}.$$  \hspace{1cm} (2)

Volume-averaged sensible enthalpy is a function of both temperature and ice saturation,

$$\rho h = \left[ s \rho_{\nu} c_{pl} + (1-s) \rho_{\nu e} c_{ple} \right] T.$$  \hspace{1cm} (4)

and volume-averaged heat capacity is

$$\rho c_p = s \rho_{\nu} c_{pl} + (1-s) \rho_{\nu e} c_{ple}.$$  \hspace{1cm} (5)

Thermal conductivity is also a function of ice saturation

$$k = s k_{\nu} + (1-s) k_{\nu e}.$$  \hspace{1cm} (6)

**Governing Equations:** Governing equations for the fixed grid calculation were derived based on the sorption-sublimation model [7-10] and using above definition of ice fraction and volume-averaged properties. The energy conservation equation is

$$\frac{\partial (\rho h)}{\partial t} + \nabla \cdot (\mathbf{\dot{m}}_w c_{pg} - k \nabla T) = \Delta h_{vap}\rho_{\nu} \frac{\partial C_{sw}}{\partial t} + \Delta h_{sub}(\rho_{\nu} - \rho_{\nu e}) \frac{\partial s}{\partial t},$$  \hspace{1cm} (7)

where Eq. (7) is a standard equation with conduction and convection terms except for the last two terms due to latent heat sources of desorption of bound water and sublimation of ice.

The desorption of bound water is governed by

$$\frac{\partial C_{sw}}{\partial t} = -k_d(1-s) C_{sw},$$  \hspace{1cm} (8)

where \( k_d \) is an empirical desorption rate constant [9]. The term \((1-s)\) makes Eq. (8) to be valid for whole domain by numerically suppressing the desorption of bound water in the frozen cells.

The temporal change of ice saturation is governed by

$$\frac{\partial (\rho_{\nu} - \rho_{\nu e})}{\partial t} \frac{\partial s}{\partial t} = -\nabla \cdot \mathbf{\dot{m}}_w,$$  \hspace{1cm} (9)

which indicates the amount of water vapor generated by sublimation should be equal to that transported by diffusion and flow.

Heat transfer boundary conditions for top and bottom boundaries were

$$\dot{q}_t = \sigma F \left( T_{hp}^4 - T_{sw}^4 \right),$$  \hspace{1cm} (10)

$$\dot{q}_b = k_f \left( T_{hp} - T_{sw} \right),$$  \hspace{1cm} (11)

where side surfaces of the product were assumed adiabatic.

The conservation equations for water vapor and inert gas in the dried region are

$$\varepsilon \frac{M_w}{R} \frac{\partial}{\partial t} \left( \frac{p_w}{T} \right) + \nabla \cdot \mathbf{\dot{m}}_w = -\rho \frac{\partial C_{sw}}{\partial t},$$  \hspace{1cm} (12)

$$\varepsilon \frac{M_w}{R} \frac{\partial}{\partial t} \left( \frac{p_w}{T} \right) + \nabla \cdot \mathbf{\dot{m}}_m = 0,$$  \hspace{1cm} (13)

where the last term in Eq. (12) is a mass source term due to the desorption of bound water.

The sublimation cells are assumed to be at their thermodynamic equilibrium and thus the vapor pressures there are determined from the saturation vapor pressure. Then, the sublimation cells act as Dirichlet boundaries for calculation of vapor pressure as

$$p_w = f_{sw}(T),$$  \hspace{1cm} (14)

The constitutive equations for mass fluxes of vapor and inert gas are suggested by the dusty-gas model [11] as

$$\mathbf{\dot{m}}_w = \frac{M_w}{RT} \left[ k_1 \nabla p_w + k_2 p_w (\nabla p_w + \nabla p_m) \right],$$  \hspace{1cm} (15)

$$\mathbf{\dot{m}}_m = \frac{M_w}{RT} \left[ k_3 \nabla p_m + k_4 p_m (\nabla p_w + \nabla p_m) \right].$$  \hspace{1cm} (16)

The absolute permeability \( K \) and the relative diffusivity factor \( D_r \) in the dried region were determined using Carman-Kozeny correlation and Bruggeman correlation for effective diffusivity [12] based on assumed particle size \( d_{part} \) of 10 \( \mu \)m. The assumed pore size was 36 \( \mu \)m from the correlation of \( d_{part} = \varepsilon d_{part} (1 - \varepsilon) \). The definition of mass transfer coefficients in Eq. (15) and (16) are given in nomenclature.

Boundary conditions for mass transfer are

$$p_w = p_{w, ch}, \quad p_m = p_{m, ch},$$  \hspace{1cm} (17)

$$\mathbf{\dot{m}}_w = \mathbf{\dot{m}}_m = 0,$$  \hspace{1cm} (18)

**Numerical Procedure:** The discretized energy equation for temperature is

$$\left( \rho c_p \right)^{\circ} \frac{T_p - T_p^{\circ}}{\Delta t} + \sum_{c,w,n,s} \left[ \mathbf{\dot{m}}_w c_p (T_j - T_p) - \dot{q}_j \right] A_j = \Delta h (\rho_{\nu} - \rho_{\nu e}) \frac{s_p^\circ - s_p}{\Delta t} \Delta V + \Delta h_s (T_{hp} - T_{sw}) \frac{s_p^\circ}{\Delta t} \Delta V.$$  \hspace{1cm} (19)
Eq. (19) is deliberately arranged to use $\rho c_p^o$ instead of $\rho c_p$, to enhance the stability of the fixed grid calculation. Since the heat capacity is a function of $s_p^o$ not $s_p$, it remains constant during an iterative calculation for a time step. On beginning calculation for a time step, each volume cells is defined as the dried, frozen, or sublimation cell according to its ice saturation. Then, temperature and partial pressures are calculated, followed by update of the ice saturation in the sublimation cells.

In the course of calculation for the ice saturation, negative ice saturation is generally unavoidable, but such small negative ice saturations do not deteriorate the accuracy of overall calculation when using Eq. (19). Once the iterative calculation for that time step ends, the non-realistic negative ice saturations should be treated appropriately. In this study, the negative ice saturation of a volume cell was distributed to its neighbouring cells based on their ice saturations.

### Table 1

| Physical Properties of Skim Milk Solution [8] and Summary of Simulated Operation Conditions |
|---------------------------------|-----------------|-----------------|-----------------|
| $c_{pl,e}$ 2590.0 J/kg         | $\rho_l$ 215.0 kg/m$^3$ |
| $c_{pl,l}$ 1930.0 J/kg         | $\rho_{le}$ 328.0 kg/m$^3$ |
| $c_{pv}$ 1616.6 J/kg           | $\rho_{ll}$ 1030.0 kg/m$^3$ |
| $C_{pw}$ 0.6415                | Operation Conditions |
| $D_{w,sh}$ $4.34 \times 10^{-6} T^{2.334} N/s$ | $H$ 2 cm |
| $f_{eq}(T)$ $10^{-26635/T+12.537}$ Pa | $L_{ld}$ 5 mm |
| $\Delta h_{sub}$ 2840000 J/kg  | $L_g$ 1 mm |
| $\Delta h_{vap}$ 2687400 J/kg  | $R_d$ 5 mm |
| $k_{le}$ 0.05 W/m-K            | $R_b$ 1.5 mm |
| $k_{ll}$ 2.12 W/m-K            | $P_{w,eh}$ 4.0 Pa |
| $k_{ld}$ $6.48 \times 10^7 s^{-1}$ | $P_{w,eh}$ 1.0 Pa |
| $\varepsilon$ 0.785            | $T_{hp}$ 313.15 K |
| $\mu$ $18.48 \times 10^{-7} [T^{1.5} / (T+650)]$ kg·m/s·K | $k_f$ 10 W/m²·K |

**RESULTS AND DISCUSSION**

A skim milk solution [8] was selected as a model drying material. The thermal properties of the skim milk solution and the simulated operation conditions are summarized in Table 1. Before simulating the freeze drying of planar products with holes and slits, the dependence of primary drying time on the operation conditions was investigated.

**Fig. 3(a)** shows that drying time $t_d$ is proportional to the product height, $H$. In fact, $t_d / H$ increases about 20%, from 5 hr/cm to 6 hr/cm, which means that smaller height of planar product is better for the productivity of the freeze drying. However, the above statement is not true because each freeze drying cycle requires rather constant time and costs for preparation of the process.

**Fig. 3(b)** shows the primary drying time is very sensitive to the heating temperature; the drying time decreased about 25%, from 16 hr to 12 hr, when heating temperature was varied from 303 K to 323 K.
In general, higher heating temperature enhances the productivity of the process but it also deteriorates the quality of product. Thus, optimal control of the heating temperature has been studied by many researchers, to obtain maximum drying rate without quality degradation. Comparison of Fig. 3(b) and (c) also shows the drying with the simulated operation conditions is limited more by heat transfer than mass transfer.

Fig. 4 compares the freeze drying characteristics of planar products and those with holes and slits. The freeze drying of planar product results in rather linear drying curve, shown in Fig. 4(a), and increasing sublimation temperature $T_{sub}$, which is due to the compensation between increase in mass transfer resistance and decrease in heat transfer resistance during the primary drying stage.

The drying curves with holes and slits shows that the drying rate is fast and constant at first, but it becomes smaller and smaller as the freeze drying continues. The predicted sublimation temperature with hole and slits are, in fact, impressive as it is about 20 K less than that of planar product.

Due to the additional transport paths provided by the holes and slits introduced, the path length for the sublimated vapor to exit the product has been significantly reduced. Mass transport path length of an interior point can be defined as the shortest distance from that point towards the surfaces open to drying chamber. The region of the largest mass transport path length, in general, dries last. In Fig. 5, main direction of drying is changed from the axial direction (top to bottom) for planar product, to radial or lateral direction (surface to interior) for planar products with holes and slits.

Finally, the productivity enhancement due to the geometrical modification is assessed in Fig. 6, where normalized drying time $\tilde{t}_d$ was adopted for accurate assessment of the productivity. Fig. 6(a) shows that the reduced drying time amounts about 20 % of the primary drying time of planar product (at $H = 3$ cm).

The normalized drying time was determined by dividing the drying time with respect to the area fill ratio, which is 0.91 for the freeze drying with holes and that with slits. The obtained normalized drying
time in Fig. 6 predicts only about 10% productivity enhancement (at \( H = 3 \text{ cm} \)).

![Figure 6](image)

The productivity in freeze drying of planar products with respect to product height: primary drying time \( t_d \), and normalized primary drying time \( \bar{t}_d \).

The predicted productivity enhancement was smaller than expected, which was due to newly emerged limitation in heat transfer. As shown in 5(b) and (c), the product region near the bottom heating plate dries fast as the path lengths for heat transport and mass transport are small there. Then, the rate of heat transfer from the plate towards the sublimation interface rapidly falls according to the progress of the freeze drying. The thermal conductivity of the dried region is about 40 times smaller than that of the frozen region.

**CONCLUSIONS**

The productivity enhancement of the freeze drying process by holes and slits introduced to planar drying products was numerically assessed. The simulation results showed that such geometrical modification provided less resistive paths for vapor exhaust and larger interfacial area for sublimation. These two effects were found to cooperatively enhance the drying rate, while decreasing the vapor pressure and the sublimation temperature in the products. It was also found that the productivity enhancement was rather smaller than expected, which was due to the reduction in the heat transfer rate. With a measure to efficiently transport the energy to the sublimation interface, the proposed geometric modification of planar products can reduce the freeze drying time and enhance productivity significantly.

**REFERENCES**