### MICROWAVE FREEZE-DRYING OF FOOD: A THEORETICAL INVESTIGATION

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(Received 24 March 1976 and in revised form 13 September 1976)

Abstract—An unsteady-state analysis of two dimensional freeze-drying with microwave internal energy generation is carried out, taking into account the differences of the transport parameters with respect to grain orientation, such as is found in food products. The anisotropic character of the material strongly influences the temperature profiles during drying. This importance is further amplified by a coupling effect between mass-transfer resistance, specimen temperature, and the absorption of microwave energy.

### NOMENCLATURE

- C, concentration of water vapor [g/cm<sup>3</sup>];
- $C_R$ , concentration of water vapor in the vacuum chamber [g/cm<sup>3</sup>];
- $C_R^*$ , concentration of water vapor at triple point  $[g/cm^3];$
- $C_p$ , heat capacity of the water vapor [cal/g°C];
- $C_{p_D}$ , heat capacity of the dried meat [cal/g°C];
- $C_{p_{\rm F}}$ , heat capacity of the frozen meat [cal/g°C];
- $D_x$ , effective diffusivity along x direction  $[cm^2/s]$ ;
- $D_y$ , effective diffusivity along y direction  $[cm^2/s]$ ;
- $D_{K_0}$ , effective Knudsen diffusivity  $[cm^2/s]$ ;
- E, peak electric field strength [V/cm];
- $E_d$ , field strength in the dielectric material [V/cm];
- f, microwave frequency [c/s];
- h, heat-transfer coefficient at external surface [cal/cm<sup>2</sup> °Cs];
- $\Delta H_s$ , heat of sublimation of ice [cal/g];
- $K_{D_x}$ , thermal conductivity of dried beef along x-direction [cal/cm °C s];
- $K_{D_y}$ , thermal conductivity of dried beef along y-direction [cal/cm °Cs];

 $K_{F_x}$ , thermal conductivity of frozen beef along x-direction [cal/cm °Cs];

- $K_{F_y}$ , thermal conductivity of frozen beef along y-direction [cal/cm °Cs];
- $S_x$ , dimensionless interface position along x-direction;
- $S_y$ , dimensionless interface position along y-direction;
- t, process time [s];
- T, temperature [°C];
- $T_R$ , temperature of the vacuum chamber [°C];
- $T_s$ , temperature at the external surface [°C];
- $T_i$ , temperature at the interface [°C];
- $T_0$ , initial temperature [°C];
- $U_x$ , dimensionless coordinate in the ice-core along x-direction;
- $U_y$ , dimensionless coordinate in the ice-core along y-direction;

- $W_i$ , mass flux at the interface;
- $W_L$ , mass flux at the external surface;
- x, x-coordinate;
- y, y-coordinate;
- X(t), x-interface position at time t;
- Y(t), y-interface position at time t;
- $Z_x$ , dimensionless coordinate in the dried layer along x-direction;
- $Z_y$ , dimensionless coordinate in the dried layer along y-direction.

### Greek letters

- $\rho$ , density of the ice [g/cm<sup>3</sup>];
- $\rho_D$ , density of the dried meat [g/cm<sup>3</sup>];
- $\rho_F$ , density of the frozen meat  $[g/cm^3]$ ;
- $\sigma$ , porosity of the dried meat;
- $\varepsilon'$ , relative dielectric constant;
- $\varepsilon''$ , relative loss factor;
- $\varepsilon_0$ , dielectric constant of vacuum;
- $\omega$ , microwave power absorbed per unit volume [cal/cm<sup>3</sup>s];
- $\delta$ , initial dried layer thickness [cm];
- $\alpha$ , thermal diffusivity  $[cm^2/s]$ ;
- $\tau$ , dimensionless time;
- $\theta$ , dimensionless temperature;
- $\Gamma$ , dimensionless concentration of water vapor;
- $\phi$ , ratio factor of thermal conductivities;
- $\psi$ , ratio of effective diffusivities.

### Subscripts

- D, in the dried layer;
- F, in the frozen layer;
- i, at the interface;
- 0, at initial time;
- x, in the x-direction;
- y, in the y-direction.

### INTRODUCTION

FREEZE-drying is a process in which frozen water contained within the pores of a body is removed by sublimation to the vapour state, usually under high vacuum. Despite its high cost, freeze-drying of food has gained acceptance as the method of drying which will generally produce the highest quality product.

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There are four potential rate-limiting steps, which occur in series with one another in conventional freezedrying processes: (1) External heat transfer from the heat source to the outer surface of each piece of material; (2) internal heat transfer from the outer surface of each piece to the sublimation front through the dried layer; (3) internal mass transfer of water vapour from the sublimation front to the outer surface; and (4) external mass transfer of water vapour from the sample surface to the condenser or other moisture sink. It is obvious that as the thickness of the dried layer continuously increases during the drying process, rate limiting steps (2) and (3) assume increasing importance. Hence, the conventional freeze-drying process is usually too slow for effective commercial processing of food products, particularly meat.

Dielectric heating using microwave power appears to offer one of the best solutions for overcoming the heat conduction problem encountered in conventional freeze-drying [1–4]. Microwaves are generated by oscillator tubes (klystrons or magnetrons) with frequencies which lie in the band 300 MHz to 300 GHz. At microwave frequencies, electromagnetic energy is absorbed in dielectric materials such as food in response to applied fields. As the various particles are accelerated by the field, they give off heat due to friction.

When applied to the freeze-drying process, microwave energy penetrates very well into ice, by-passing the problem of heat conduction across the dried layer. This gives essentially volumetric heating of the receding ice-core, and hence reduces the drying time by as much as 75%. This has been successfully demonstrated on an experimental scale by Copson [1], Jackson [3], Hoover [5], Decareau [6], and Ang [7].

To provide a better understanding of the heat and mass transfer processes occurring during microwave freeze-drying, an unsteady state, two dimensional freeze-drying model using microwave energy is developed. This two dimensional model is shown to be a distinct improvement over the previous flat slab models.

### LITERATURE REVIEW

Numerous freeze-drying models have been reported in the literature and several comprehensive reviews on freeze-drying have been given by Harper and Tappel [8], Burke and Decareau [9], Ginnette and Kaufman [10]. The most recent review given by King [11], covering all aspects of freeze-drying, is a valuable guide to the literature. However, it should be pointed out that none of these theoretical analyses deal with internal generation of energy and the anisotropic character of food. In this section, only literature pertaining to the present investigation will be discussed.

Thermal conductivity data are needed for analysis of internal heat transfer during freeze-drying. Harper [12] and Woodams and Nowrey [13] have shown that thermal conductivities of freeze-dried foods are very similar even for materials as diverse as fruit and meat. For raw beef, Bralsford [14] and Gunn and King [15] showed that the thermal conductivity perpendicular to the grain is always less than that parallel to the grain. At a pressure of 0.5 torr, the thermal conductivity across fibres amounts to only two-thirds of that along the fibres.

There are four mechanisms which contribute to gas mass transfer in porous media: (1) bulk diffusion, (2) Knudsen diffusion, (3) slip flow, and (4) Poiseuille flow. A complicated equation which accounts for all these effects has been proposed by Wakao *et al.* [16]. However, for freeze-drying at low chamber pressure, Knudsen diffusion accounted for most of the mass flux, and surface diffusion has not been found to be a significant contributor [11]. Therefore, it is possible to use an effective diffusivity. Sandall *et al.* [17], Harper [12] and Dyer and Sunderland [18] have reported effective diffusivities parallel and perpendicular to the grain for turkey breast and raw beef respectively.

Microwave properties of food products such as dielectric constant and relative loss factor are unfortunately incomplete or even unavailable over the temperature ranges of interest. For raw beef, the most widely used dielectric data are those of Kan and Yeaton [19] over the microwave frequency range of 500 and 3000 MHz. They found that the dielectric properties of raw beef are strongly dependent on temperature.

Previous work on microwave freeze-drying concentrated essentially on experimental investigations [1–6]. The earliest mathematical analysis was carried out by Copson [1]. The analysis was limited to simplified approaches using a quasi-steady state assumption. The first general unsteady analysis with internal heat generation was carried out by Ma and Peltre [20] with an infinite slab model. Therefore, both models have neglected the anisotropic character of food, which has a strong effect on the temperature profiles of the material during drying.

### THEORETICAL ANALYSIS

Influence of geometry and anisotropic structure on freeze-drying

Consider a slab and a square of the same material, both to be freeze-dried under the same conditions using microwave energy as shown in Fig. 1. The displacement rate of the interface in the slab is given by:

$$\frac{\mathrm{d}I(t)}{\mathrm{d}t} = -\frac{W_i}{\rho\sigma} \tag{1}$$

and similarly for the square case:

$$\frac{\mathrm{d}X(t)}{\mathrm{d}t} = -\frac{(W_i)_x}{\rho\sigma}; \quad \frac{\mathrm{d}Y(t)}{\mathrm{d}t} = -\frac{(W_i)_y}{\rho\sigma} \tag{2}$$

where X(t) and Y(t) are locations of x and y interfaces at time t respectively.

Assume that the resistance to mass transfer is negligible, so that all sublimed vapour immediately reaches the outer face. For the slab  $W_L = W_i$ , but for the square  $W_L \neq (W_L)_x \neq (W_L)_y$  due to geometry and anisotropy. Therefore, dX(t)/dt will not be equal to dY(t)/dt. In other words, at a given rate of displacement of the interface, the sublimation rates (in terms of the



FIG. 1. Effect of geometry and anisotropy on the mass flux at the external surface of the samples—slab (above) and square (below).

total amount of vapour removed from the specimen) are not the same due to geometric and anisotropy factors.

It has been established in the literature that specimen temperature is critical in the analysis of microwave freeze-drying. Under a given experimental condition, the temperature of the specimen is controlled by the mass transfer rate which is in turn controlled by the resistance of the dried layer and the rate of shrinkage at the interface. Therefore, it is obvious that anisotropy has a strong influence on the quality of freeze-dried product obtained.

### Model description and assumptions

Consider a piece of meat of infinite length with a square cross section. As freeze-drying proceeds, the ice-front (assumed to be of zero thickness) retreats, the sublimed water vapour being removed by diffusion through the porous dried layer. Heat is transferred by conduction and convection in the dried region, while only conduction will be considered in the frozen zone. If the grain is oriented along the y-axis, both thermal conductivity and effective diffusivity in the y-direction will be greater than those in the x-direction. The rate of ice-front shrinkage in the y-direction will, then, be faster than that in the x-direction.

A certain degree of shrinkage (amounting to less than 15% of the original volume) usually occurs during freeze-drying [21], however to simplify the system, it is neglected. Margaritis and King [22] have shown that there is a finite absorption of moisture in the dried layer (less than 3%), due to the flow of water vapour. However, this absorption contributes a negligible amount to the overall mass-transfer resistance. With microwaves, although a larger amount of adsorption may be expected (since the temperature of the dried

layer is somewhat lower), the effect will still be negligible.

In any microwave cavity, when the electric field is parallel to the surface of the material, the field strength is the same in both the immediate surroundings and within the material, regardless of dielectric constant. When the field is perpendicular to the surface of the material, then the field strength inside the dielectric is related to the field strength. It is well known that the true situation must lie between these two extreme cases. In order to simplify the situation, it is assumed that only the dominant mode is able to propagate and the dominant polarization of the field is normal to the surface of the sample. Moreover, the strength of the electric found is assumed constant throughout the sample and equal to the value at the surface. This is justified since the penetration depth at the microwave frequency of 2450 MHz is much greater than the size of the sample.

### Differential equations of the model

The differential equations which describe the physical system are:

- Dried layer.
- (i) Heat transfer:

$$K_{D_{x}}\frac{\partial^{2}T_{D}}{\partial x^{2}} + K_{D_{y}}\frac{\partial^{2}T_{D}}{\partial y^{2}} + \left[\frac{\partial K_{D_{x}}}{\partial x} - C_{p}W_{x}\right]\frac{\partial T_{D}}{\partial x} + \left[\frac{\partial K_{D_{y}}}{\partial y} - C_{p}W_{y}\right]\frac{\partial T_{D}}{\partial y} = \rho_{D}C_{pD}\frac{\partial T_{D}}{\partial t} - \omega_{D}.$$
 (3)

(ii) Mass transfer:

$$D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + \frac{\partial D_x}{\partial x} \frac{\partial C}{\partial x} + \frac{\partial D_y}{\partial x} \frac{\partial C}{\partial x} = \sigma \frac{\partial C}{\partial t}.$$
 (4)

Frozen layer. (i) Heat transfer:

$$\begin{bmatrix} K_{F_x} \frac{\partial^2 T_F}{\partial x^2} + K_{F_y} \frac{\partial^2 T_F}{\partial y^2} \end{bmatrix} + \begin{bmatrix} \frac{\partial K_{F_x}}{\partial x} \frac{\partial T_F}{\partial x} + \frac{\partial K_{F_y}}{\partial y} \frac{\partial T_F}{\partial y} \end{bmatrix}$$
$$= \rho_F C_{PF} \frac{\partial T_F}{\partial t} - \omega_F. \quad (5)$$

There will be no mass transfer in the frozen layer. The boundary conditions are

At the center, for  $t \ge 0$ 

$$x = 0: \frac{\partial T_F}{\partial x}\Big|_{x=0} = 0; \quad y = 0: \frac{\partial T_F}{\partial y}\Big|_{y=0} = 0.$$
 (6a)

At the interface, thermodynamic equilibrium between ice and vapour would exist. Therefore, the concentration of the water vapour can be related to the ice temperature by an equilibrium relationship. Energy balances at the x and y interfaces will give:

$$x = X(t): (W_i)_x \Delta H_s = K_{F_x} \left[ \frac{\partial T_F}{\partial x} \right] - K_{D_x} \left[ \frac{\partial T_D}{\partial x} \right]$$
(6b)  
$$y = Y(t): (W_i)_y \Delta H_s = K_{F_y} \left[ \frac{\partial T_F}{\partial y} \right] - K_{D_y} \left[ \frac{\partial T_D}{\partial y} \right]$$

and  $T_D = T_F = T_i$ . At the outer surface, for t > 0,

$$x = y = L: h(T_{R} - T_{S}) = K_{DX} \frac{\partial T_{D}}{\partial x} = K_{DY} \frac{\partial T_{D}}{\partial y}.$$
 (6c)

By assuming the mass-transfer resistance at the outer surface to be negligible (because of low chamber pressure):

$$C(L, t) = C_R \quad \text{for} \quad t \ge 0. \tag{6d}$$

At the initial state of freeze-drying when material is introduced to the chamber, before the desired level of vacuum is reached and microwave power turned on, some water vapour is sublimed, giving an initial thickness of dried layer  $\delta$ . Therefore, an initial dried layer thickness of 0.004*L* is used in this analysis. Since this initial freeze-drying is essentially conventional freezedrying, in which the heat of sublimation comes from the surroundings, both temperature and concentration profiles at this stage are assumed to be linear, and the frozen zone temperature assumed constant. Then, the initial condition is as follows:

$$T_{0} = T_{F} \text{ for } 0 \leq x \leq L - \delta, \quad 0 \leq y \leq L - \delta$$
$$\frac{T_{S} - T_{D}}{T_{S} - T_{0}} = \frac{L - x}{\delta} \text{ for } L - \delta \leq x \leq L \quad (7a)$$
$$\frac{T_{S} - T_{D}}{T_{S} - T_{0}} = \frac{L - y}{\delta} \text{ for } L - \delta \leq y \leq L.$$

The concentration profile is likewise assumed to be linear:

$$C = C_R - \left[\frac{\partial C}{\partial x}\right]_i (L - x) \text{ for } L - \delta \le x < L \quad (7b)$$
$$= C_R - \left[\frac{\partial C}{\partial y}\right]_i (L - y) \text{ for } L - \delta \le y < L.$$

By assuming normal polarization, the power dissipated in a dielectric substance which responds to an applied electric field is given by:

$$\omega = \pi f \varepsilon_0 \varepsilon'' \frac{E^2}{{\varepsilon'}^2 + {\varepsilon''}^2}.$$
 (8)

# Transformation of differential equations into dimensionless form

The following dimensionless variables are used to normalize the differential equations:

$$Z_x = \frac{L - x}{L - \bar{\mathbf{X}}(t)}; \quad U_x = \frac{x}{\bar{\mathbf{X}}(t)}$$
(9a)

$$Z_y = \frac{L-y}{L-Y(t)}; \quad U_y = \frac{y}{Y(t)}$$
(9b)

$$\tau = \frac{\alpha_{Fx}^0 t}{L^2}; \quad \Gamma = \frac{C - C_R}{C_R^* - C_R}$$
(9c)

$$\theta = \frac{T - T_0}{T_R - T_0}; \ S_x = \frac{\bar{\mathbf{X}}(t)}{L}; \ S_y = \frac{Y(t)}{L}.$$
 (9d)

## PHYSICAL PROPERTIES AND EXPERIMENTAL CONDITIONS USED IN THE SIMULATION

Raw beef is chosen as the material for simulation, since the physical properties are readily available in

the literature. The moisture, and fat content, density, and porosity of beef were assumed to be constant and values as quoted by Awberry and Griffiths [23] were used in the simulation. Functional relationships expressed as polynomials of temperature were used for thermal conductivity [13, 15, 24], effective diffusivity [17], ice-water vapour equilibrium relationship [25], and dissipation coefficient in a microwave field [19]. Since the dissipation coefficient for frozen beef is an order of magnitude higher than for dried beef, the frozen beef absorbs the major fraction of the microwave energy.

The temperature and total pressure of the vacuum chamber are taken to be constant. The variation in total pressure is assumed to have a very small effect on the water removal process, since the gas near the outer surface of the dried layer is in the Knudsen-flow regime. The initial temperatures in the dried and frozen layers are assumed to be -25 and  $-15^{\circ}$ C respectively. The dried layer is assumed to be cooler than the frozen layer based on the assumption that during start-up ice is sublimed. The rate of heat loss by this means is appreciably greater than the rate of heat absorption by the material from its immediate surroundings. The result of these effects will be a temperature drop in the dried layer analogous to the wet bulb process. Therefore, the exact initial temperature in both frozen and dried layers varies and is difficult to control. Fortunately, it has only a small effect on the outcome of the drying process.

In freeze-drying, the maximum temperature of both dried and frozen layers during the process is often more important than the total drying time required. The maximum temperature reached in most cases determines the quality of the resulting food products and hence the feasibility of a given drying process. Therefore, two temperature constraints are put into the model. In the frozen zone, the temperature is kept below  $-3^{\circ}$ C, while an upper limit of 60°C in the dried layer is set to prevent thermal degradation of the dried products.

### Numerical solutions

Equations (3)–(5) are a set of parabolic partial differential equations describing heat and mass transfer with phase change at a moving boundary. Moreover, the physical properties of the beef are functions of dependent variable (temperature). Therefore, an analytical solution to such a complicated situation is unlikely, hence a numerical technique, the Crank–Nicholson method, is employed. A five-point implicit, central difference scheme is used. Moreover, Gaussian elimination is used to solve the linear equations desired from the difference equations and the boundary conditions. As the three partial differential equations are coupled through the boundary conditions at the interface, an iterative procedure is necessary to solve the problem.

Furthermore, from the physical observation of the ice-core at different stages of drying, the corner effect present in the system is obvious. Carslaw and Jaeger



FIG. 2. Temperature profiles (field strength = 130 V/cm).



FIG. 3. Water vapour concentration profiles (field strength = 130 V/cm).

[26] in their analytical solution of conduction of heat in a solid of finite rectangular shape also showed that due to the corner effect, the isotherms in that region are rounded. Therefore to obtain normal convergence of the numerical solution using the mesh size limited by computer storage capacity at the University of Waterloo, the interface mesh grid at the corner is adjusted in the simulation. The detail of the numerical solution as well as the computer program for the model are given in reference [7].

### Discussion of results

The temperature profiles obtained in the x and y directions are parabolic in shape, for both the frozen and dried zones as shown in Fig. 2. On the other hand, the concentration profiles are essentially linear at all times as shown in Fig. 3. This indicates that the accumulation terms in the mass balance equations are negligible compared to the space derivative. This result is in agreement with previous studies by Copson [26], Dyer *et al.* [25], and Ma and Peltre [20].

At the start of the process, the temperature of the ice-core dropped to a minimum, because the masstransfer resistance of the initial dried layer is small. The microwave energy absorbed by the specimen at this stage is insufficient to provide the heat of sublimation at the interface. However, as the thickness of the dried layer gradually increases, its temperature starts to rise to a maximum due to the increasing mass-transfer resistance encountered. Since the mass-transfer rate is greater, the dried zone temperature in the y-direction at x = 0 is always lower than its x-direction counterpart. After nearly half of the ice is sublimed, the temperature in the frozen zone starts to decrease while the dried zone temperatures in both the x and y directions increase continuously. This is because at this stage the energy absorbed by the ice-core, due to reduction in volume to less than one-half of its original value, is no longer sufficient to provide the necessary heat of sublimation. However, the rate of decrease in frozen temperature during this stage is considerably less because the mass-transfer resistance of the dried layer has become higher.

Since microwave heating is volumetric, and the frozen beef is absorbing the major fraction of the microwave energy, the volume of the ice-core remaining during the drying cycle is critical in the process analysis. It has been observed that the freeze-drying cycle can roughly be divided into four periods. During the initial period, the appearance of the retreating icecore will be in the form of a rectangle. However, because of the corners, it will shrink to the form of an ellipse, with major axis perpendicular to the grain orientation. This period is referred to as the second period. During the third period of the cycle, the ice-core shrinks to the shape of a thin plate of negligible thickness. Finally after the ice-core disappears, the residual moisture in the dried product is removed.

During period one, the external surface temperature  $T_S$  is less than the chamber temperature  $T_R$ . The heat flux at the external surface at this stage is from the surroundings to the sample. The actual power consumed by the sublimation at this stage is slightly greater than the power supplied by microwaves. However, the heat flux from the surroundings will decrease as  $T_S$  increases. When  $T_S$  approaches  $T_R$  (corresponding to the transition from period one to period two), this heat flux approaches zero. From period two up to the end of the process, heat flows from the external surface of the specimen to the surroundings. The actual power consumed by the sublimation is thus slightly smaller than that supplied by microwaves during this period.

As shown in Fig. 4, the electric field strength has



FIG. 4. Effect of field strength on drying time.

a strong effect on the drying process. An increase in electric field strength inside the microwave applicator increases the drying rate. However, the temperatures attained in both the dried and frozen regions are likewise proportional to the field strength, as shown in Fig. 5. In order to prevent melt back at the ice-front, and also thermal degradation of the dried region, only a certain allowable field strength is applicable under a given condition. It is important to mention here that if the quality of the freeze-dried product is not of critical importance, it is possible to use a higher field strength to achieve a shorter drying cycle. In this case, the maximum applicable field strength will be limited by plasma formation.

### Parameter analysis

In general, the transport parameters of the material with respect to grain orientation may be expressed mathematically as

$$K_v = \phi K_x$$
 and  $D_v = \psi D_x$ 



FIG. 5. Effect of field strength on specimen temperatures at various drying times.

where  $\phi$  and  $\psi$  are the ratio factors. The ratio factors are the properties of the materials which may be constant or variable with respect to a given condition. The appearance of the retreating ice-core will be in any form as shown in Fig. 6, depending on the magnitude of the ratio factors. It should be noted that

φ≨ψ

depends on the characteristic of the material. For example in the case of raw beef, at a pressure of 0.5 mmHg,  $\phi$  is approximately equal to 1.5, and  $\psi$ approximately equal to 3, assuming the grain is oriented along the y-direction.

If  $\phi = 1$ , and  $\psi = 1$ , that is, the transport parameters are equal in both directions, the appearance of the uniformly retreating ice-core will be a perfect square during the initial stage, and will be in the form of a circle in the second period.

If  $\phi = 0$ , and  $\psi = 0$ , the freeze-drying process becomes one dimensional, and the infinite slab model is obtained. In other words, the infinite slab model is a special case of the two dimensional model, in which the ratio factors both equal zero. For  $\phi$  and  $\psi$  other than these two special cases, the uniformly retreating ice-core will be in the form of a rectangle in period one. It will shrink to an ellipse in period II. In Fig. 6, this is designated as Model IIa and IIb. The ratio between the major and minor axes will depend on the relative magnitudes of the ratio factors.

If identical initial experimental conditions are assumed, there will not be a great difference between the models as far as the total drying time is concerned. This is because microwave heating is volumetric and volume has no direction. The drying curves of the different models under identical experimental conditions are shown in Fig. 7. However, it is important to note that the relative drying rates are not the same from period to period. At the initial period of the



FIG. 6. Effect of ratio factors on the shapes of the retreating frozen front.



FIG. 7. Effect of ratio factors on total drying time.

process, the one dimensional infinite slab model has the fastest drying rate. It is followed by Model II. Model I, which has both ratio factors equal to 1 has the slowest initial drying rate. However, it overtakes the two eventually. At the end of the drying cycle, the slow starting Model I predicts the shortest total drying time, while the fast starting one dimensional model predicts the longest total drying time.

The maximum ice-core temperature is plotted against the drying time as shown in Fig. 8. All models are assumed to have an initial ice-core temperature of  $-15^{\circ}$ C. At the initial stages of the process, the one dimensional model has the highest ice-core temperature. Correspondingly, it has the fastest drying rate. Model I, on the other hand, has the coolest ice-core temperature, thus it has the slowest drying rate. Model II is intermediate between these two extremes. During the later period, Model I has the highest ice-core temperature. Also it has the fastest drying rate. The one dimensional model has the lowest ice-core temperature, and the slowest drying rate. Model II is again intermediate between these two.

The correlation between ice-core temperature and drying-rate is due to a coupling effect of: (a) mass transfer resistance; (b) specimen temperature, and (c) dissipation coefficient. The coupling effect may be illustrated as follows: At the initial stages of the process, the thickness of the porous dried layer, which is responsible for the mass-transfer resistance, is relatively small. Model I has the largest vectorial sum of dif-



FIG. 8. Effect of ratio factors on the ice-core temperature.

fusivities among the three. It will have the fastest masstransfer rate. At the other extreme, the one dimensional model has the highest mass-transfer resistance. When the mass-transfer resistance is low, the specimen will be at the lowest temperature. Since the dissipation coefficient of raw beef in both frozen and dried region increases rapidly with increase in temperature, the amount of microwave energy absorbed in Model I becomes lowest among the models. Drying rate in a microwave process is essentially controlled by the amount of energy dissipated, especially in the ice-core. Thus Model I will have the slowest initial drying rate. As the ice-core shrinks, the mass transfer resistance due to the dried layer increases. Since a circle has the least perimeter for a given area, Model I will have the least sublimation front. Consequently, it will have the largest mass-transfer resistance. Its temperature will become the highest, and the energy dissipated will become the highest among the modes giving the fastest final drying rate.

In microwave freeze-drying, the maximum temperature obtained in both dried and frozen layers during the process is far more important than the total drying time required. The maximum temperature obtained actually determines the quality of the resulting product. It likewise determines the feasibility of a given process. As shown in Fig. 8, the ice-core temperature of Model III, which is the one dimensional infinite slab model reaches a maximum of  $-4.2^{\circ}$ C. If  $-3^{\circ}$ C is the maximum allowable ice-core temperature, the conclusion will be that the process is feasible, and the product obtained will be of good quality. However, the maximum ice-core temperature obtained by Model I is  $-2.9^{\circ}$ C. The conclusion based on this will be that the process is not feasible, since it has over-shot the maximum allowable ice-core temperature. Under these conditions, melt-back would occur, yielding a product of inferior quality.



FIG. 9. Effect of ratio factors on the drying curve, in the absence of coupling effect.



FIG. 10. Effect of ratio factors on ice-core temperature, in the absence of coupling effect.

If the dissipation coefficient of the material is not a strong function of the temperature, the coupling effect will not exist. The drying curve as well as the maximum ice-core temperature will be as shown in Figs. 9 and 10. Similar to Fig. 7 and Fig. 9, it shows that if identical initial experimental conditions are used, there will not be a great difference between the models as far as the total drying time is concerned. This is because microwave freeze-drying is controlled essentially by the ability of the dielectric to absorb microwave energy volumetrically in response to the applied field strength. However, if the dissipation coefficient of the material is not a strong function of the temperature, the overlapping of drying curves as shown in Fig. 7, and maximum ice-core temperatures as shown in Fig. 8, among the three models will not occur. In fact the maximum ice-core temperatures of the specimens of Model I and II (Curve B and C) are always below that of the one dimensional model (Curve A).

The coupling effect can also be used to explain the influence of ambient pressure on the process. If freezedrying is carried out at a significantly higher ambient pressure, the total drying time required will become slightly shorter due to the following sequence: (a) at higher ambient pressure, the mass transfer resistance is higher; (b) the ice-core temperature then increases, due to this higher mass transfer resistance; (c) the energy dissipated in the ice-core then becomes higher, due to the higher temperature; and (d) the drying rate thus becomes faster.

However, as mentioned earlier, the maximum temperature attained during the microwave freeze-drying process is far more important than the total drying time required. An increase in ambient pressure has the effect of increasing the drying rate. However, the allowable field strength has to be reduced severely in order to prevent melting of the ice-core. Furthermore, at pressures of 1.5 mmHg or above, the probability of gas plasma formation at the usual operating field strength increases.

From the above analysis, low operating chamber pressures are recommended to ensure temperatures of the frozen core as low as possible and thus allow use of higher microwave power inputs to shorten the drying time. However, pressures below 0.25 mmHg seem impractical, as the pressure effect becomes insignificant, because in the pores of the dried product the gas is in the Knudsen flow regime. Low pressures also reduce the probability of occurrence of the corona discharge.

An increase in sample dimension has a similar effect on the process. This can also be attributed to the coupling effect. For a larger sample, in the second and third period of the drying cycle, the mass transfer resistance in the dried layer is higher. The temperature of the ice-core thus increases. The energy dissipation ability of the specimen then becomes higher. The drying rate, as a consequence, increases. Again, the maximum temperature attained in the process is far more important than the drying rate. The field strength applicable has to be decreased, to ensure good product quality.

Therefore, a reduction of sample dimension is recommended. A reduction of sample size results in a shorter drying time, because a higher microwave field strength is applicable without overheating or melting.

### CONCLUSIONS

From the above analysis, it may be concluded that (1) the mathematical model developed can be used to simulate freeze-drying of any material with or without internal heat generation; (2) it is important to use a two dimensional model which can take into consideration the anisotropic character of the material to optimize the freeze-drying process; (3) the effect of anisotropy, amplified by the coupling effect, greatly influences the process; (4) microwave power shortens the total drying time significantly when compared with conventional freeze-drying; (5) a maximum applied electric field is usually limited to between 100-135 V/cm to ensure the fastest drying rate without overheating or melt-back condition; (6) the chamber pressure should be maintained between 0.4-0.2 mmHg; (7) due to the coupling effect, the size of the sample limits the maximum applicable field strength for uniform heating of the entire specimen.

Acknowledgement—The authors would like to express their sincere appreciation to J. C. Food Product Inc. of the Republic of the Philippines for providing financial support to Dr. T. K. Ang.

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### LYOPHILISATION DES ALIMENTS EN PRESENCE DE MICRO-ONDES-UNE ETUDE THEORIQUE

Résumé—On effectue une analyse non-stationnaire du séchage avec congélation dans deux dimensions avec production de micro-ondes d'énergie interne, compte tenu des différences entres paramètres de transport dûes à l'orientation des grains, comme cela existe dans les produits alimentaires. Le caractère anisotrope du matériau influe fortement sur les profils de température lors du séchage. Cette importance est encore accrue par l'effet de couplage entre la résistance au transfert massique, la température de l'échantillon et l'absorption de l'énergie des micro-ondes.

### MIKROWELLEN-GEFRIERTROCKNUNG VON LEBENSMITTELN-EINE THEORETISCHE UNTERSUCHUNG

Zusammenfassung – Der instationäre Fall der zweidimensionalen Gefriertrocknung mit Mikrowellenbeheizung wird analytisch untersucht, wobei die Unterschiede in den Transportparametern infolge der Kornorientierung, wie sie bei Lebensmitteln gefunden wird, berücksichtigt werden. Der anisotrope Charakter des Materials beeinflußt stark das Temperaturprofil während des Trocknens. Dieser Einfluß wird noch verstärkt durch einen Kupplungseffekt zwischen Stoffaustauschwiderstand, Probentemperatur und Absorption der Mikrowellenenergie.

### МИКРОВОЛНОВАЯ СУБЛИМАЦИОННАЯ СУШКА ПИЩЕВЫХ ПРОДУКТОВ. ТЕОРЕТИЧЕСКОЕ ИССЛЕДОВАНИЕ

Аннотация — Проведен анализ нестационарных двухмерных уравнений сублимационной сушки при внутреннем выделении микроволновой энергии с учетом зависимости параметров переноса от ориентации текстуры пищевых продуктов. Анизотропный характер материала оказывает сильное влияние на температурные профили в процессе сушки. Это влияние еще более усиливается за счет перекрестных эффектов, обусловленных сопротивлением переноса массы, температурой образца и поглошением микроволновой энергии.