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HEAT AND MASS TRANSFER

MECHANISMS IN FREEZE-DRYING

A THESIS

Presented to

The Faculty of the Graduate Division

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Thurman C. Hardin

In Partial Fulfillment

of the Requirements for the Degree

Doctor of Philosophy

in the School of Mechanical Engineering

Georgia Institute of Technology

May, 1965

HEAT AND MASS TRANSFER

MECHANISMS IN FREEZE-DRYING

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Date approved by Chairman: My 27, 1965

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SUMMARY

Previous analytical solutions of the quasi-steady coupled heat and mass transfer processes which occur during freeze-drying in an effort to predict drying rates and drying time have not correlated very well with experimental data. Therefore, the objective of this investigation is to make a probing theoretical and experimental investigation of the coupled heat and mass transfer mechanisms in freeze-drying processes. First, analyses are sought which will provide more accurate methods for estimating drying rates and drying time under a variety of boundary conditions. Second, it is desired to establish relationships between the many variables affecting the drying rates and drying time so as to indicate the most fruitful avenues for possible improvements in the rate of freeze-drying.

In the theoretical analyses an energy equation is developed which results in a non-linear temperature distribution in the porous dried layer and a linear temperature distribution in the frozen layer. The continuity equation for the flow of rarefied water vapor molecules through the capillary channels in the porous dried layer is based upon the kinetic theory of gases. It is shown that both the pressure gradient and the temperature gradient along the capillary channels contribute to the mass flux of vapor. Finally, the energy and continuity equations are combined to provide "exact solution" equations for calculating the interface temperature, pressure distribution, drying rates, and drying time for different drying arrangements and various boundary conditions.

The "exact solution" indicates that the temperature distribution

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in the dried layer is essentially linear and that the interface temperature remains essentially constant throughout the freeze drying process. Also, the interface temperature depends primarily upon the vacuum chamber pressure. An "approximate solution" based upon these assumptions results in closed-form equations for drying rates and drying time which agree substantially with calculations using the more tedious "exact solution" equations.

The experimental investigation involved a number of 24-hour tests at constant vacuum chamber pressure during which the following measurements were made: pressure distributions in the porous dried layer, temperature distributions in both the dried and frozen layers, drying rate, and drying time.

Correlation between the actual and theoretical drying rates and drying time curves is quite satisfactory for vacuum chamber pressures of 0.5 and 2 torr, but less satisfactory for the 3 torr vacuum chamber pressure.

Both theoretical and experimental analyses indicate that the drying time tends to decrease as the vacuum chamber pressure is reduced and as the boundary temperatures are increased. It also is shown that the drying time depends upon the drying arrangement, i.e. whether drying occurs from one surface only or simultaneously from two surfaces.

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NOMENCLATURE

English Letters

Units

a	constants (m = 1, 2,)	dimensionless
a	capillary tube radius	ft
Α	cross-sectional area of sample	ft ²
b _m	constants (m = 1, 2,)	dimensionless
С	constant pressure specific heat	Btu/lb °F
с _т	constants (m = 1,2,)	dimensionless
d	mean diameter of capillary channel	ft
f	heat flux ratio, defined on page 16	dimensionless
a ^c	constant, 32.1739 (3600) ²	$lb_m ft/lb_f hr^2$
k _D	thermal conductivity of dried layer	Btu/ft hr °F
k _F	thermal conductivity of frozen layer	Btu/ft hr °F
K	term defined on page 26	1bm/hr ft
K(z)	drying time function defined on pages 41, 42, and 44	hr ft/Btu
ł	sample thickness	ft
L	latent heat of sublimation	Btu/lb _m
L' .	latent heat plus superheat of sublimated vapor	Btu/1bm
m.	mass flow rate of vapor through a capillary tube	lb _m /hr
М	term defined on page 23	lb /ft hr
n	number of capillary channels per unit cross-sectional area óf sample	1/ft ²
N	term defined on page 23	lb _m ft ³ °R/1b _f ²

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hr

English Letter	S	Units
р	absolute pressure	l torr = 1 mm Hg
P	porosity (void fraction)	dimensionless
q	heat flux	Btu/hr ft ²
q"	constant defined on page 42	Btu/hr ft
r	distance measured in radial direction	ft
R	gas constant	ft lb _f /lb _m °R
S	term defined on page 26	dimensionless
t	time	hours
Т	absolute temperature	• Rankine
U	overall heat transfer coefficient of combined frozen and bottom dried layer, defined on page 33	Btu/ft hr 🗣
V	term defined on page 26	dimensionless
ŵ	mass flow rate of vapor per unit cross- sectional area of sample	lb _m /hr ft ²
x	distance measured in x-direction	ft
X(z)	drying time function defined on pages 39 and 40	Btu/hr ft
Y(z)	drying time function defined on page 42	dimensionless
Greek Letters		Units
β	term defined on page	ft ⁻¹
δ	dried layer thickness	ft
λ	mean free path	ft
π	constant	3.1416
ρ	ice density	1b _m /ft ³
۶	term defined on page 07	11 /2 001/2

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	Gre	eek	Let	ters
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δ

ξ	term defined on page 20	dimensionless
μ	absolute viscosity of vapor	lb _f hr/ft ²
Subscripts		Units
•	ton (free) surface of sample	

Bbottom surfaceDrefers to dried layerFrefers to frozen layersat.thermodynamic property in a saturated state

interface surface

Superscripts

Kn P*

T

х*

z

t	temperature in frozen layer
*	dimensionless quantity
-	average quantity

Dimensionless Quantities

	Knudsen number	x/d
	dimensionless pressure	p/p _o
	dimensionless temperature	T/T
. •	dimensionless distance measured in the dried layer	x/ð
	dimensionless dried layer thickness, or mass fraction of sublimated ice	8/2

xv

Units

CHAPTER I

INTRODUCTION

General

Freeze-drying is a process whereby food or biological substances are frozen and subsequently dehydrated by sublimation of the ice in a vacuum chamber. Food processed in this manner is placed in an airtight package and may be stored safely without refrigeration for periods up to a year. Other advantages of freeze-drying include loss of weight (up to 70%), high quality of re-hydrated product due to the relatively small shrinkage, uniform distribution of the various natural salts within the food, and avoidance of a scorched taste sometimes associated with conventionally dehydrated food when heat is supplied directly to unfrozen food.

The first application of freeze-drying began in the 1940's with the preservation of biological substances for which the processing time and costs were not controlling factors. Since World War II attention has been directed toward preserving of food products, particularly meat, by means of freeze-drying. In this application the processing time and costs obviously are important. There is, however, surprisingly little information available to permit the prediction of freeze-drying rates and drying time.

During freeze-drying a porous dried layer builds up which increases in thickness as drying is continued. Sublimation takes places at the

boundary separating the porous dried layer and the frozen layer which is referred to as the interface. The heat required for sublimation may be transferred to the top porous dried layer either by radiation or conduction from a heat source, or it may be conducted through the bottom frozen layer, or it may be conducted through both the dried and frozen layers to the interface where it brings about sublimation of ice (frozen meat juices). The sublimation vapor escapes through the porous dried layer to the surface where it is removed from the vacuum chamber.

Analytical solutions of the guasi-steady coupled heat and mass transfer processes which occur during freeze-drying in an effort to predict freeze-drying rates have not correlated very well with experimental data. This lack of correlation may be attributed to the use of over-simplified mathematical models, to empirical approaches, to the scarcity of basic heat and mass transport property data for food products, and to an incomplete understanding of the phenomena associated with rarefied vapor flow through the capillary-like channels in the porous dried layer. Therefore, the objective of the present investigation is to overcome these deficiencies by presenting a probing theoretical and experimental investigation of the coupled heat and mass transfer mechanisms in freeze-drying processes. First, analyses are sought which will provide methods for more accurate estimates of freeze-drying rates and drying time under a variety of conditions. Second, it is desired to establish relationships between the many variables affecting the drying rate and drying time to indicate the most fruitful avenues for possible improvements of the freeze-drying processes.

Literature

General

A predominant portion of the literature on freeze-drying is concerned with the biological aspects of the technique. A comprehensive survey of these considerations is contained in the book edited by Harris (1).*

Flosdorf's book (2) is one of the earliest publications on freezedrying and is an outgrowth of his work in developing both laboratory and production scale freeze-drying equipment. However, his analytical treatment is somewhat superficial and outdated.

Charm's (3) book is an excellent survey of the various problems associated with food dehydration in general, and freeze-drying in particular.

Transport Properties

The most fundamental and significant investigations of the various transport properties of porous freeze-dried materials have been those carried out by Harper and Tappel (4).

The thermal conductivity of gas-filled porous freeze-dried materials was determined for a variety of rarefied gases and porous freezedried products. A semi-empirical equation is developed by Harper expressing the thermal conductivity of the gas-filled porous solid material as the sum of the individual conductivities of the porous solid and rarefied gas. The contribution of the gas to the composite thermal conductivity is pressure dependent and increases with pressure up to a certain

Numbers in parentheses refer to the Bibliography, page 156.

level, after which it becomes constant and independent of further increases in pressure.

Harper also conducted experimental investigations to determine the permeability of porous freeze-dried products to the isothermal flow of rarefied gases under various flow conditions. These experiments made possible the determination of the mean effective diameter of the capillary channels in the porous freeze-dried products. Additional experimental work was carried out to determine the dielectric properties of both porous freeze-dried products. Finally, the diffusion coefficients for gases flowing through porous freeze-dried materials due to a concentration gradient were measured for a variety of gases and freeze-dried products.

Miller and Sunderland (5) determined the thermal conductivity of frozen beef and showed that it is temperature dependent, increasing as the mean temperature of the product is decreased. Also, Sevick and Sunderland (6) measured the thermal emissivity of beef fat, and fresh and freeze-dried beef.

The saturation pressure-temperature relationship for subliming frozen beef currently is under investigation by Dyer and Sunderland (7).

The transport property data for both porous freeze-dried beef and for frozen beef obtained from Harper's and Sunderland's investigations proved to be of great value in the analyses made in the present investigation.

Heat and Mass Transfer Analyses

Harper and Tappel (4) present a very comprehensive discussion of their analytical and experimental investigations on freeze-drying. They

use a one-dimensional model with heat transferred by radiation to the surface of the sample, after which it is conducted downward through the vapor-filled porous dried layer to the interface where the ice is sublimated. The sublimated vapor flows from the interface through the capillary channels in the porous dried layer. The vacuum chamber total pressure is greater than the partial pressure of the water vapor due to the partial pressure of the air which leaks either controlled or uncontrolled into the vacuum chamber. They suggest that the vapor flow can be either hydrodynamic due to a total pressure gradient across the dried layer, or diffusional flow due to a partial pressure gradient of the water vapor. Furthermore, under certain conditions, the vapor flow may be a combination of hydrodynamic and diffusional flow. The hydrodynamic flow is divided into three regimes, continuous (or Poiseuille), slip (or transition), and free molecule flow. For hydrodynamic flow Harper and Tappel set up an expression for the vapor mass flow rate by making use of a modified form of Darcy's Law to account for slip flow in which the velocity is finite at the walls of the capillary channels. The vapor velocity is expressed in terms of total pressure gradient, vapor viscosity, a correction factor to account for slip flow, and a proportionality constant called permeability. The equation is integrated assuming isothermal flow of the vapor through the capillary channels. This assumption seems unjustifiable but is considered preferable to the assumed adiabatic flow process used in the pioneering work of Flosdorf \cdot (2).

Lambert (8) maintains that the energy input to the subliming interface is the controlling factor in the freeze-drying process, and that the

total vacuum chamber pressure, condenser pressure, and structure of the porous dried layer, through their effect on vapor transport, govern the rate of vapor flow through the porous dried layer. He sets the vapor flux equal to the driving forces divided by the sum of the flow resistances between the interface and condenser, and proceeds to categorize the driving forces and resistances. His analyses are somewhat similar to that of Harper and Tappel's (4) but he lumps all of the transport properties into a flow parameter called "cake conductivity." This simplification, therefore, is limited in usefulness and fails to shed light on the mechanism involved in the vapor flow through the porous dried layer.

Lambert points out that, in general, the estimated interface temperature increases with the total pressure in the vacuum chamber, and that the saturation temperature of pure water ice corresponding to the total pressure in the vacuum chamber provides a lower limit for the interface temperature. He also suggests that at a given sublimation temperature the vapor pressure for subliming food products may be lower than the corresponding vapor pressure for pure water ice due to the dissolved salts.

Bannister (9) takes a fundamental approach to the problem of vapor flow in freeze-drying. He develops a model in which the porous dried layer is pictured as a bundle of capillary tubes oriented parallel to the direction of vapor flow. Heat first is radiated to the top surface of the product and then conducted downward through the vapor-filled porous dried layer. Equations are developed for the vapor mass flow rate starting with the basic Poiseuille laminar flow equation; this is followed

by equations for free molecule flow and transition flow of rarefied gases flowing under isothermal conditions. The temperature of the vapor actually must increase as it flows through the porous dried layer because of the temperature gradient due to the difference between the interface and top surface temperatures. Thus Bannister's analyses are somewhat inconsistent since the flow equations are derived for isothermal conditions. The temperature distribution in the porous dried layer is assumed to be linear; and the energy and continuity equations are combined to give expressions for the drying time for continuous, transition, and free molecule flow. Unfortunately, no criteria are given for selecting the appropriate drying time equation to be used for given boundary conditions during freeze drying, and no provision is made for determining the interface pressure and temperature which appear in the equations. Bannister suggests that additional experimental work is needed in order to establish the various flow regimes which occur during freeze-drying. As in Harper and Tappel's more simplified analyses, Bannister's analyses show that the total drying time is proportional to the square of the product thickness.

A satisfactory energy equation is developed by Koumoutsos and Sunderland (10) for the porous dried layer, involving the heat conducted through the dried layer to the interface and the sublimated vapor flowing in the opposite direction. This equation considers both the latent heat of sublimation at the interface and the superheating of the vapor as it flows through the porous dried layer. This results in a non-linear temperature distribution. The equations developed for the flow of vapor through the porous dried layer are similar to those developed by

Bannister. The energy and continuity equations are combined and integrated, making certain questionable simplifying assumptions, to give the total drying time which is proportional to the square of the product thickness. As in Bannister's analyses, no criteria are given for selecting the appropriate drying time equation to be used for given boundary conditions during freeze drying, and no provision is made for determining the interface pressure and temperature which appear in the equations. Finally, the bottom frozen layer is assumed to be adiabatic with zero temperature gradient. This assumption does not seem to be realistic in light of the experimental observations made in the present investigation.

A number of interesting experiments on freeze drying of various materials were made by Kessler (11). The temperature distributions were measured during freeze-drying and it was observed that the interface temperature remained essentially constant throughout the process. This same observation was made by Hatcher (12). The theoretical analyses which are developed in the present investigation provide an explanation of this phenomenon; and the essentially constant behavior of the interface temperature forms the basis for the drying rate and drying time equations developed for the so-called approximate solution. Kessler sets up a valid differential energy equation for the subliming interface but does not integrate it. The flow equation used is similar to Darcy's law but does not consider the possibility of either free molecule or transition flow.

As previously mentioned, Harper shows that the thermal conductivity of the composite vapor-filled porous dried layer increases with

pressure up to a certain level and then remains constant. It was observed by Harper that the interface temperature in many actual freeze drying operations may be as low as O°F or less, which is considerably below the level required to protect product quality. The low interface temperature is attributed to the high insulating effect of the porous dried layer. As a result of this reasoning the theory is advanced that a higher vacuum chamber pressure would increase the drying rate by increasing the thermal conductivity of the composite vapor-filled porous dried layer. The increase in mass flow rate of vapor would require a higher differential pressure between the interface and vacuum chamber which, when added to the already higher vacuum chamber pressure, would increase the interface pressure and corresponding saturation temperature. This would tend to reduce the heat flux because of the reduced temperature gradient; however, Harper suggests that an optimum vacuum chamber could be determined which would result in a maximum drying rate. His calculations indicate that the optimum vacuum chamber pressure would occur at approximately six torr, but obviously this is greater than the triplepoint pressure of frozen pure water. Therefore, it is suggested that perhaps a vacuum chamber pressure only slightly less than the triplepoint pressure of 4.58 torr would be optimum and would also prevent thawing of the product. This higher vacuum chamber pressure could be easily provided by bleeding air or other inert gases into the system. It is not clear if these calculations are based on the isothermal flow equations and if heat flux through the frozen bottom layer is taken into account.

The theoretical analyses developed in the present investigation

indicate that, even when considering the increase of the thermal conductivity with a higher vacuum chamber pressure, a lower vacuum chamber pressure would, in fact, result in an increase in drying rate. This is true because of two factors which more than offset the reduction in thermal conductivity which would result with a lower vacuum chamber pressure. First, a lower vacuum chamber pressure will reduce the interface pressure and corresponding saturation interface temperature which will increase the temperature gradient in the porous dried layer; and secondly, the lower interface temperature will increase both the temperature gradient and thermal conductivity in the frozen bottom layer. The net effect of all these factors will be to increase the heat fluxes and the drying rate. Also, it may be pointed out that the interface temperature is established primarily by the vacuum chamber pressure level because of its effect on interface pressure; and, therefore, the relatively low interface temperature is little affected by the high insulating effect of the porous dried layer. Experimental observations carried out in the present investigation tend to confirm the general validity of the theoretical analyses developed herein.

CHAPTER II

THEORETICAL ANALYSES

<u>General</u>

The theoretical analyses of freeze-drying involves the derivation of the energy and continuity equations and their simultaneous solution in order to determine the interface temperature. With the interface temperature established, equations are derived for both the drying rates and the drying time as a function of the dimensionless dried layer thickness.

All transport properties which are pressure and/or temperature dependent are evaluated at the vacuum chamber pressure and mean temperatures respectively. Thus, for a given vacuum chamber pressure and fixed boundary temperatures, all transport properties are treated as constants.

The energy and continuity equations are derived on the basis of one-dimensional and quasi-steady flow conditions. This implies that both the heat and mass fluxes are zero except in the x-direction, that both the heat and mass fluxes are steady over any short interval of time, that the change in stored energy in both the dried layer and frozen layer is small compared with the energy absorbed by the frozen meat juices upon sublimation, and that the mass fluxes of sublimated vapor entering and leaving the porous dried layer are equal.

Figure 1(a) shows a typical freeze-drying arrangement used in the present investigation. A radiant plate-type electric heater provides

a radiation heat flux to the free surface of the sample, x = 0, while the bottom of the sample, $x = \ell$, rests on a support through which a conduction heat flux flows to the bottom of the sample. A vapor barrier restrains the sublimated vapor so that it flows only in the negative x-direction. Fiberglass insulation is placed around the edges of the sample to minimize the radial heat flux.

The plane separating the porous dried layer and the frozen layer is referred to as the interface, and its position is time dependent as it moves in the positive x-direction through the sample. The heat of sublimation is provided both by the radiant heat flux from the heater to the free surface and hence by conduction through the vapor-filled porous dried layer in the positive x-direction to the interface, and the conduction heat flux through the bottom of the sample in the negative x-direction to the interface.

Energy Equation

In order to derive the energy equation, an energy balance for quasi-steady flow conditions is made for a control volume in the porcus dried layer having a cross-sectional area A and thickness Δx . The control volume consists of porcus solid material and sublimated vapor flowing through the capillary channels in the negative x-direction. It is assumed that at all points within the control volume the vapor and solid which are in contact with each other are at the same temperature. Referring to Figure 1(b), the heat conducted past positions x and $x + \Delta x$ are given by the following equations



Figure 1. (a) Sample Arrangement. (b) Control Volume for Dried Layer.

$$- k_{D}^{A} \frac{\partial T}{\partial x} \Big|_{x}$$

$$- k_{D}^{A} \frac{\partial T}{\partial x} \Big|_{x + \Delta x}$$
(1)
(2)

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The energy transferred to the control volume due to the enthalpy of the sublimated vapor flowing past positions \vec{x} and $x + \Delta x$ are given by the following equations

$$\hat{w}AcT \Big|_{X}$$

$$\hat{w}AcT \Big|_{X} + \Delta X$$

$$(3)$$

$$(4)$$

An expression for the rate of accumulation of energy within the control volume is

$$\bar{\rho}_{\rm D} \bar{c}_{\rm D} A \Delta x \frac{\partial T}{\partial t}$$
(5)

The law of conservation of energy applied to the control volume

$$\begin{bmatrix} Rate of energy \\ flowing out \end{bmatrix} - \begin{bmatrix} Rate of energy \\ flowing in \end{bmatrix} = - \begin{bmatrix} Rate of energy \\ accumulation \\ in control volume \end{bmatrix}$$
(6)

Substituting equations (1), (2), (3), (4), and (5) into equation (6) gives the following

$$-k_{D}^{A} \begin{bmatrix} \frac{\partial T}{\partial x} \Big|_{x + \Delta x} - \frac{\partial T}{\partial x} \Big|_{x} \end{bmatrix} + \hat{w} c A \begin{bmatrix} T | x + \Delta x = T |_{x} \end{bmatrix}$$
$$= - \begin{bmatrix} \overline{\rho}_{D} \overline{c}_{D} A \Delta x \frac{\partial T}{\partial t} \end{bmatrix}$$
(7)

Symbols are defined on p. xiii.

requires

Dividing equation (7) by $A\Delta x$, and recognizing that for quasisteady flow conditions $\partial T/\partial t = 0$, results in the following equation

$$k_{D} \frac{\left| \frac{\partial I}{\partial x} \right|_{x + \Delta x} - \frac{\partial I}{\partial x} \right|_{x}}{\Delta x} - \overset{\circ}{wc} \frac{\left[T \right|_{x + \Delta x} - T_{x} \right]}{\Delta x} = 0$$
(8)

If the lim $\Delta x \rightarrow 0$ is taken for equation (8) and β is set equal to wc/k_D, the differential energy equation is obtained. The partial derivative notation is no longer necessary because the temperature depends only on the single variable x.

$$\frac{d^2T}{dx^2} - \beta \frac{dT}{dx} = 0 \quad \text{for} \quad 0 \le x \le \delta$$
(9)

In order to obtain the necessary boundary conditions for equation (9), an energy balance at the interface x = b is made by equating the rate of latent heat of sublimation to the total conduction heat fluxes through both the dried and frozen layers. Designating the temperature at any point in the frozen layer by T', the interface energy balance can be written

$$\dot{\mathbf{w}}\mathbf{L} = \mathbf{k}_{\mathrm{D}} \left. \frac{\mathrm{d}\mathbf{T}}{\mathrm{d}\mathbf{x}} \right|_{\mathbf{x}=\delta} - \mathbf{k}_{\mathrm{F}} \left. \frac{\mathrm{d}\mathbf{T}'}{\mathrm{d}\mathbf{x}} \right|_{\mathbf{x}=\delta} \tag{10}$$

At the interface $x = \delta$ let the heat flux conducted through the porous dried layer, expressed as the fraction of the total heat flux conducted through both the dried and frozen layers, be defined as the heat flux ratio and designated by f. Thus,

$$f = \frac{\text{heat flux conducted through the dried layer}}{\text{total heat flux conducted through dried and frozen layers}} \Big|_{x=\delta} (11)$$

The heat flux ratio f is given by the following equation in terms of the temperature gradients in the dried and frozen layers

$$f = \frac{k_{D} \frac{dT}{dx}}{k_{D} \frac{dT}{dx}}_{x=\delta} - k_{F} \frac{dT}{dx}}_{x=\delta}$$
(12)

Substituting equation (10) into equation (12) gives the following additional expression for the heat flux ratio

$$f = \frac{k_D \frac{dT}{dx}}{\frac{dT}{dx}}_{x=\delta}$$

Equation (13) provides one of the necessary boundary conditions for the solution of the differential energy equation (9). The boundary conditions are

$$T = T_{A} \quad \text{at} \quad x = 0 \tag{14}$$

$$\frac{dT}{dx}\Big|_{x=\delta} = \frac{f\dot{w}L}{k_D} \quad \text{at } x = \delta \tag{15}$$

It is shown later that the heat flux ratio f is a function of the interface position, which is a function of the drying time.

At the interface the dried and frozen layers have a common temperature. Thus,

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(13)

$$T = T_{\delta} = T' \quad \text{at } x = \delta \tag{16}$$

Applying boundary conditions (14) and (15) in the solution of the differential energy equation (9) results in the following equation for the temperature distribution in the porous dried layer

$$T(x) = T_{0} + \frac{fL}{c} \exp(-\beta\delta)(\exp(\beta x) - 1) \quad \text{for } 0 \le x = \delta$$
 (17)

The following equation for the mass flux of vapor is obtained by substituting equation (16) into equation (17) and rearranging.

$$-\dot{w} = \frac{k_D}{\delta c} \ln \left[\frac{c}{fL} \left(T_0 - T_\delta \right) + 1 \right] \text{ for } 0 \le x \le \delta$$
 (18)

If it is assumed that the energy equation is quasi-steady and the thermal conductivity is constant in the frozen layer, the temperature gradient will be linear. The resulting temperature distribution in the frozen layer is easily obtained and is given by the following equation

$$T'(x) = T_B - (T_B - T_\delta) \left[\frac{\cancel{l} - x}{\cancel{l} - \delta}\right] \text{ for } \delta \le x \le \cancel{l}$$
 (19)

Both the temperature distribution equation (17) and mass flux equation (18) for the dried porous layer will be used subsequently, with the continuity equation for the mass flux of vapor through the capillary channels of the porous dried layer, in order to derive a combined energycontinuity equation. Figure 2 shows a schematic temperature distribution curve for both the dried and frozen layers.









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Transition Flow

General

The model used to develop the continuity equation can be visualized as a bundle of long capillary channels oriented parallel to the direction of vapor flow.

The type of flow in the capillary channels depends upon the degree to which the vapor is rarefied. The flow is free-molecule for highly rarefied gases. In this type of flow viscosity does not play a role. For more dense gases the flow is Poiseuille flow for laminar conditions, with the well-known parabolic velocity distribution and zero wall velocity. For gases which are only moderately rarefied the flow is known as transition flow, in which there is a finite velocity at the wall due to the "slip flow" of molecules along the wall. In this type of flow viscosity also plays a role.

The particular type of flow depends upon the ratio of the mean-free path of the vapor molecules to the capillary diameter which is known as the Knudsen number.

$$K_{\rm N} = \frac{\lambda}{d}$$

 λ = mean-free path of molecules

d = capillary diameter

The type of flow for different ranges of the Knudsen number is given by the following
$2 \leq K_{\rm N}$.01	Free-molecule flow
$01 \leq K_{N} \leq 2$	Transition flow
$K_{N} \leq 0.1$	Poiseuille flow

Continuity Equation

For most freeze-drying operating conditions the vapor flow will occur in the transition flow regime; therefore, equations will be developed first for this type of flow.

The mass flow rate of rarefied vapor through a long capillary tube subjected to both a pressure gradient and a temperature gradient is given by the following differential equation derived from Kinetic Theory considerations (13)

$$\dot{m} = -\frac{(1 + 4\xi/a)\pi a^4 p}{8\mu RT}\frac{dp}{dx} + \frac{3}{4}\frac{\pi a^2 g_c \mu}{T}\frac{dT}{dx}$$

- a = capillary radius
- R = gas constant
- µ = absolute viscosity
- $\xi = 2c' \frac{2-f'}{f'} \lambda$

f' = transfer ratio for momentum

2c'= 1.0, a constant

Kennard (13) shows that in the case of transition flow of gas through a long capillary tube subjected to both a pressure gradient and a temperature gradient that $f^{+} = 1$ for vapor molecules reflected diffusely from the capillary walls. Therefore, from the definition of ξ , it follows that $\xi = \lambda$. Since $\xi/a = 2\lambda/d$, it is obvious that the

(20)

following relationship between $|\xi|$ and the Knudsen number is valid

$$K_{\rm M} = \xi/2a \tag{21}$$

The mass flow rate per unit area of the porous dried layer and the porosity are related to the number of capillary tubes per unit area by the following equations

$$\dot{w} = \dot{m} n$$
 (22)
 $P = \pi a^2 n$ (23)

where

n = number of capillary tubes per unit area of the dried layer

Substituting equations (21), (22), and (23) into equation (20) gives the following relationship for the mass flow rate of sublimated vapor per unit area of the porous dried layer

$$\dot{w} = -\left[\frac{(1 + 8K_N) d^2 p}{32 \mu RT} \frac{dp}{dx} + \frac{3}{4} \frac{g_c^{\mu}}{T} \frac{dT}{dx}\right] P \text{ for } 0 \le x \le \delta \qquad (24)$$

Rearranging, and recognizing that the temperature depends only on x,

$$int T(x) dx = -\left[\frac{(1+8K_N) d^2 p}{32 \mu RT} dp + \frac{3}{4} g_c \mu dT\right] P \text{ for } 0 \le x \le \delta$$
 (25)

Equation (25) involves only the variables x, pressure, and temperature, because time is held fixed during its integration, thus fixing the position of the interface δ . This fixes the mass flux of vapor \dot{w} , so that it becomes independent of x while the integration of equation (25) is performed. Refer to equation (18) for the mass flux of sublimated vapor.

For the isothermal transition flow of vapor through a long capillary tube subjected only to a pressure gradient, equation (25) reduces to the following.

$$\hat{w} = -\frac{(1 + 8K_N) P d^2 p}{32 \mu RT} \frac{dp}{dx} \text{ for } dT/dx = 0 \qquad (26)$$
$$0 \le x \le \delta$$

Equation (26) is convenient to use in the investigation of such transport properties as mean capillary diameter of the flow channels in the porous dried layer and the permeability of porous media to the isothermal flow of rarefied gases. Harper (4) made use of this equation in his experimental work involving the isothermal flow of rarefied water vapor through freeze-dried materials.

Combined Energy and Continuity Equations

The first two terms on the left-hand side of equation (25) can be replaced by the mass flow rate equation (18) and the temperature distribution equation (17) respectively, both of which are obtained from the differential energy equation. This substitution, therefore, results in the following combined energy-continuity differential equation for transitional flow

$$\frac{1}{\delta} \frac{k_{D}}{c} \ln \left[\frac{c}{fL} \left(T_{o} - T_{\delta} \right) + 1 \right] \left[T_{o} + \frac{fL}{c} \exp \left(-\beta \delta \right) \left(\exp \left(\beta x \right) - 1 \right) \right] dx$$
$$= \left[\frac{\left(1 + 8K_{N} \right) d^{2} p}{32 \mu R} dp - \frac{3}{4} g_{c} \mu dT \right] P \text{ for } 0 \le x \le \delta \qquad (27)$$

The boundary conditions are

$$p = p_0$$
 and $T = T_0$ at $x = 0$ (28)

$$p = p_x$$
 and $T = T_x$ at $x = \delta$ (29)

Integrating equation (27) and applying boundary conditions (28) and (29) results in the following combined energy-continuity equation in terms of the heat flux ratio f and the interface pressure and temperature

$$k_{\rm D} \ln \left[\frac{c}{fL} \left(T_{\rm o} \frac{d}{dx} T_{\rm b} \right) + 1 \right] \left(T_{\rm b} - \frac{fL}{c} \right) + M \left(T_{\rm o} - T_{\rm b} \right) - N \left(p_{\rm b}^2 - p_{\rm o}^2 \right) = 0 \quad (30)$$

for $0 \le x \le b$

where

$$M = \frac{k_D}{c} - \frac{3}{4} P g_c \overline{\mu}$$
(31)

$$N = \frac{P d^2 (1 + 8K_N)}{64 \,\overline{\mu} R}$$
(32)

At the subliming interface $x = \delta$ the saturation pressure and temperature for a pure substance are related by the Clapeyron equation^{*}

$$\frac{dp}{dT} = \frac{L}{T (v_{g} - v_{s})}$$
(33)

* For derivation refer to any standard Thermodynamics book.

v_q = specific volume of saturated vapor

Using the ideal gas equation of state $v_g = RT/p$, and recognizing that $v_s \approx 0$, equation (33) may be rewritten

$$\frac{dp}{dT} = \frac{Lp}{RT^2}$$
(34)

The boundary conditions are

$$T = T_{r} at p = p_{r}$$
(35)

 $T = T_{\delta}$ at $p = p_{\delta}$ (36)

where

where

pr = an arbitrary saturation reference pressure

T_r = saturation temperature corresponding to the reference pressure

Integrating the Clayperon equation, realizing that $L \approx$ constant, and applying boundary conditions (35) and (36) gives the following equation relating the interface saturation pressure and temperature

$$p_{k} = C_{p} \exp(-L/RT)$$
(37)

where

$$C_{r} = p_{r} \exp(L/RT_{r})$$
, a constant (38)

In the case of frozen meat juices, the saturation pressuretemperature relationship may be different from that for pure water ice. Therefore, the saturation pressure-temperature relationship for subliming frozen meat juices may be determined experimentally to give a relationship of the following form

$$P_{\delta} = P_{\delta} (T_{\delta})$$
(39)

Substitution of either equation (37) or equation (39) into the combined energy-continuity equation (30) eliminates the interface saturation pressure and results in a combined energy-continuity equation in terms of the interface saturation temperature and heat flux ratio f. Thus,

$$\varphi(T_{\mathbf{g}}, \mathbf{f}) = 0 \tag{40}$$

Exact solutions to equation (40) can be obtained. These solutions give values of the interface temperature corresponding to various constant values of the heat flux ratio which will simultaneously satisfy both the energy and continuity equations. A digital computer program based upon Newton's method was written to solve equation (40) using both the integrated Clapeyron equation for frozen pure water and experimental data relating the subliming saturation pressure and temperature for frozen meat juices. The results of these calculations appear in the section on Results of Theoretical Analyses for Transition Flow.

Dimensionless Temperature Distribution

It is easy to rearrange the temperature distribution equation (17) into the following dimensionless temperature distribution equation in

terms of the heat flux ratio and interface temperature

$$I^{*} = \frac{T}{T_{0}} = 1 - S[1 - \exp(VX^{*})] \quad \text{for } 0 \le x \le \delta$$
(41)
$$0 \le X^{*} \le 1$$

where

$$S = 1 - \frac{T_{\delta}}{T_{o}} + \frac{fL}{cT_{o}}$$

$$V = -\ln \left[\frac{c}{fL} (T_{o} - T_{\delta}) + 1 \right]$$

 $X^* = x/\delta$, dimensionless distance in dried layer

Dimensionless Pressure Distribution

The combined energy-continuity equation (40) can be rearranged to give the following dimensionless pressure distribution equation in terms of the heat flux ratio and dimensionless temperature

$$p^{*} = \frac{p}{p_{0}} = \left[\frac{1}{Np_{0}^{2}} \left[T_{0}M(1-T^{*}) + KX^{*}(\frac{fL}{c} - T_{0}T^{*})\right] + 1\right]^{1/2}$$
(42)
for $0 \le x \le \delta$
 $0 \le X^{*} \le 1$

where

$$K = -\frac{k_D}{c} \ln \left[\frac{c}{fL} \left(T_o - T_b\right) + 1\right]$$

Free-Molecule Flow

Continuity Equation

In the free-molecule regime the following differential equation for the mass flow rate of rarefied vapor in a long capillary tube subjected

to both a pressure gradient and a temperature gradient is derived from Kinetic Theory considerations (13)

$$\dot{m} = -\frac{4}{3} \sqrt{\frac{2\pi}{R}} a^3 \frac{d(\zeta)}{dx}$$

where

$$\zeta = p/T^{1/2}$$

As mentioned in the section on transition flow, the mass flow rate per unit area of the dried layer and the porosity are both related to the number of capillary tubes per unit area of the dried layer by equations (22) and (23) respectively. Therefore, when these two equations are substituted into equation (43), the following expression is obtained for the mass flux of sublimated vapor

$$\dot{w} dx = -\frac{2}{3} \sqrt{\frac{2}{\pi R}} P d d(\zeta)$$
 (44)

Equation (44) involves only the variables x and ζ because time is held fixed during its integration, thus fixing the position of the interface \diamond . This fixes the mass flux of vapor $\mathbf{\hat{w}}$ so that it becomes independent of x while the integration of equation (44) is performed. Refer to equation (18) for the mass flux of vapor. The boundary conditions are

 $\zeta = \zeta_0 \quad \text{at } \mathbf{x} = 0 \tag{45}$

 $\zeta = \zeta_{A}$ at $x = \delta$

27

(43)

(46)

Following the procedure used for transition flow, the mass flux of vapor equation (18), derived from the differential energy equation, is substituted into equation (44) to give the following combined energycontinuity differential equation

$$\frac{1}{\delta} \frac{k_{\rm D}}{c} \ln \left[\frac{c}{fL} \left(T_{\rm o} - T_{\rm b} \right) + 1 \right] dx = M^{\circ} d(\zeta)$$
(47)

where

$$M' = \frac{2}{3} \sqrt{\frac{2}{\pi R}} P d$$

Integrating equation (47) and applying boundary conditions (45) and (46) results in the following combined energy-continuity equation

$$\frac{k_{\rm D}}{c} \ln \left[\frac{c}{fL} \left(T_{\rm o} - T_{\rm b}\right) + 1\right] - M' \left[\frac{p_{\rm b}}{T_{\rm b}^{1/2}} - \frac{p_{\rm o}}{T_{\rm o}^{1/2}}\right] = 0 \quad (48)$$

As previously mentioned, the saturation pressure and temperature at the interface are related by the Clapeyron equation for a pure substance. For frozen meat juices the saturation pressure-temperature relationship during sublimation may be determined experimentally to give a relationship of the form: $p_{\delta} = p_{\delta}(T_{\delta})$.

Combined Energy and Continuity Equations

By substituting the appropriate relationship for the interface saturation pressure in terms of the interface temperature into equation (48) the combined energy-continuity equation can be expressed in the following form

$$p'(f, T_{\lambda}) = 0$$

As previously mentioned, equation (49) can be solved to give exact values of the interface temperature corresponding to various constant values of the heat flux ratio f.

Dimensionless Temperature Distribution

The dimensionless temperature distribution for the dried porous layer is the same as for transition flow and is given by equation (41). Dimensionless Pressure Distribution

The combined energy-continuity equation can be rearranged to give the dimensionless pressure distribution equation (50) in terms of the heat flux ratio and dimensionless temperature. It is interesting to note that this equation is different from the dimensionless pressure distribution equation (42) obtained for transition flow.

$$p^{*} = \frac{p}{p_{o}} = \sqrt{\frac{T_{o}T^{*}}{p_{o}}} \left\{ \frac{k_{D}X^{*}}{cM^{*}} \ln \left[\frac{c}{fL} (T_{o} - T_{\delta}) + 1 \right] + \frac{p_{o}}{\sqrt{T_{o}}} \right\}$$
(50)

for $0 \le x \le \delta$ $0 \le X^* \le 1$

Exact Solution for Transition Flow

Drying Rate Equations

As previously explained, equation (40) can be used to determine the interface temperature corresponding to the various constant values of the heat flux ratio f. These temperatures will simultaneously satisfy both the energy and continuity equations and can be used with equation (51) to determine the drying rate. Equation (51) is a

(49)

slightly modified form of equation (18) for the mass flux of vapor which was obtained from the differential energy equation.

$$- \dot{\mathbf{w}} = \frac{1}{z\sqrt{2}} \frac{k_{\rm D}}{c} \ln \left[\frac{c}{fL} \left(T_{\rm o} - T_{\rm b} \right) + 1 \right]$$
(51)

where 🚽

$z = \delta/l$ the dimensionless dried layer thickness

Equation (51) can be used to calculate the drying rate provided a relationship can be found between z and f. Therefore, this relationship will next be derived. An energy balance at the interface is made by equating the rate of latent heat of sublimation to the combined heat fluxes conducted to the interface through both the dried and frozen layers. Thus,

$$\mathbf{\hat{w}}\mathbf{L} = \mathbf{k}_{\mathbf{D}} \left. \frac{\mathrm{d}\mathbf{T}}{\mathrm{d}\mathbf{x}} \right|_{\mathbf{x}=\mathbf{\delta}} - \mathbf{k}_{\mathbf{F}} \left. \frac{\mathrm{d}\mathbf{T}}{\mathrm{d}\mathbf{x}} \right|_{\mathbf{x}=\mathbf{\delta}} \quad \text{at } \mathbf{x} = \mathbf{\delta}$$
 (52)

rearranging,

$$1 = \frac{k_{\rm D} \frac{dT}{dx}}{\dot{w}L} - \frac{k_{\rm F} \frac{dT'}{dx}}{\dot{w}L}$$
(53)

Substituting equation (13) for the heat flux ratio into equation (53) gives the following relationship

$$1 - f = \frac{-k_F \left. \frac{dT'}{dx} \right|_{x=\delta}}{\mathring{w} L}$$
(54)

As previously mentioned, if it is assumed that the heat flow is quasi-steady and the thermal conductivity is constant in the frozen layer, the temperature gradient will be linear and can be written

$$\frac{dT'}{dx} = \frac{T_B - T_{\delta}}{\ell - \delta} = \frac{T_B - T_{\delta}}{\ell(1 - z)} \quad \text{for } \delta \le x \le \ell$$
 (55)

Substituting equation (55) into equation (54) gives

$$1 - f = \frac{-k_F(T_B - T_\delta)}{\ell (1 - z) \, \dot{w} \, L}$$
(56)

Substituting equation (51) for the mass flux of vapor into equation (56) gives

$$1 - z = \frac{k_{\rm F} (T_{\rm B} - T_{\rm b})}{(1 - f) \frac{L k_{\rm D}}{zc} \ln \left[\frac{c}{fL} (T_{\rm o} - T_{\rm b}) + 1\right]}$$
(57)

Thus by setting

$$D = \frac{k_{F} (T_{B} - T_{\delta})}{(1 - f) \frac{L k_{D}}{c} \ln [\frac{c}{fL} (T_{o} - T_{\delta}) + 1]}$$

the following equation relating the heat flux ratio and dimensionless dried layer thickness is obtained

$$z = \frac{1}{1 + D}$$
 (58)

The Effect of Restricted Bottom Drying

Experimental evidence indicates that after the subliming interface has penetrated the sample some distance the sample begins to partially dry at the bottom and around the edges even though a vapor barrier covers the bottom and sides of the sample. The formation of a dried layer on the bottom, unfortunately, acts as an excellent thermal insulator which reduces the heat flux through the bottom and thus reduces the drying rate.

To take the phenomenon of restricted bottom drying into account the heat flux through the combined frozen layer and bottom dried layer is calculated by using an overall heat transfer coefficient for the combined layers, designated by U. It is assumed that the bottom layer remains frozen in the early stages of freeze-drying; however, when the interface reaches some critical value, designated z_c , it is assumed that the bottom layer gradually begins to dry and continuously increases in thickness for the remainder of the freeze-drying process. This will cause the overall heat transfer coefficient to decrease until the sample is completely dried. It is obvious that as the thickness of the frozen layer approaches zero (i.e. as $z \rightarrow 1$) the overall heat transfer coefficient U must approach the thermal conductivity of the porous dried layer. Thus the overall heat transfer coefficient U must satisfy the following conditions

1. For the upper limit of U, $U = k_F$ for $0 \le z \le z_c$ 2. For the lower limit of U, $U \rightarrow k_D$ as $z \rightarrow 1$

Assuming a linear variation of the overall heat transfer coefficient U with the dimensionless dried layer thickness z, equation (59) is obtained for U which satisfies conditions 1 and 2 above. Details of the derivation are given in Appendix A.

$$U = a_1 + b_1 z$$
 for $z_c \le z \le 1$

where

$$a_{1} = k_{F} + \frac{z_{c}(k_{F} - k_{D})}{1 - z_{c}}$$
(60)

$$b_{1} = -\frac{(k_{\rm F} - k_{\rm D})}{1 - z_{\rm C}} \tag{61}$$

Bottom Dried Layer Thickness

It is interesting to determine the relationship between the bottom and top dimensionless dried layers which occurs if the overall heat transfer coefficient U varies linearly as given by equation (59). Details of the derivation of equation (62) are given in Appendix B.

$$z_{2} = \left[\frac{k_{F}}{a_{1} + b_{1}z_{1}} - 1\right] \frac{(1 - z_{1})}{a_{4}} \quad \text{for} \quad z_{c} \le z \le 1$$
 (62)

where

$$a_4 = \frac{k_F - k_D}{k_D}$$

 $z_1 = \delta_1/\ell$ top dimensionless dried layer thickness $z_2 = \delta_2/\ell$ bottom dimensionless dried layer thickness $\delta_2 = 0$ bottom dried layer thickness

A plot of equation (62) is shown in Figure 47 which indicates that the bottom dimensionless dried layer z_2 increases approximately linearly with the top dimensionless dried layer z_1 for $z_1 \ge z_c$ and reaches a

33

(59)

maximum value of approximately 0.025. Thus only a relatively thin bottom dried layer substantially reduces the overall heat transfer coefficient U. This reduces the heat flux through the bottom and lengthens the drying time.

The semi-empirical derivation of equation (59) for the overall heat transfer coefficient requires that the critical value of z be estimated. The critical value of z will depend upon a number of factors such as the bottom surface temperature T_B . At present no rigorous scheme is available for calculating z_c ; however, experimental observations indicate that a good estimate for the critical value of z will be in the range $0.2 \le z_c \le 0.3$. A value of $z_c = 0.2$ is assumed in all calculations which follow.

<u>The Effect of Restricted Bottom Drying Upon the Relationship Between</u> the <u>Heat Flux Ratio and the Top Dimensionless Dried Layer</u>

The effect of restricted bottom drying upon the relationship between the heat flux ratio f and the top dimensionless dried layer z_1 is obtained by substituting equation (59) for the overall heat transfer coefficient into equation (57) in place of the thermal conductivity of the frozen layer k_F whenever $z_c \leq z \leq 1$. This gives the following equation relating f and z_1

$$1 - z_{1} = \frac{(a_{1} + b_{1}z_{1}) (T_{B} - T_{\delta})}{(1 - f) \frac{L k_{D}}{z_{1}c} \ln \left[\frac{c}{fL} (T_{0} - T_{\delta}) + 1\right]} \text{ for } z_{c} \le z \le 1$$
 (64)

Thus, the relationship between z_1 and f can be written

$$z_{1} = \frac{-(a_{3} + 1) + [(a_{3} + 1)^{2} + 4b_{3}]^{\frac{1}{2}}}{2b_{3}} \text{ for } z_{c} \le z \le 1$$
 (65)

where

$$a_{3} = \frac{a_{1}(T_{B} - T_{\delta})}{(1 - f) \frac{L k_{D}}{c} \ln \left[\frac{c}{fL}(T_{o} - T_{\delta}) + 1\right]}$$
(66)

$$b_{3} = \frac{b_{1}(T_{B} - T_{\delta})}{(1 - f) \frac{L}{c} \frac{k_{D}}{c} \ln \left[\frac{c}{fL}(T_{0} - T_{\delta}) + 1\right]}$$
(67)

Calculation Procedure

Calculations for the exact solution drying rates are made using the following general procedures:

 Use equation (40) to determine the interface temperatures corresponding to the various assigned constant values of the heat flux ratio f.

2. For $0 \le z \le z_c$, use equation (58) to determine the position of the interface z_1 corresponding to the various assigned constant values of the heat flux ratio f.

3. For $z_c \leq z \leq 1$, use equation (65) to determine the position of the interface z_1 corresponding to the various assigned constant values of the heat flux ratio f.

4. Finally, with the interface temperatures and the values of z_1 corresponding to the heat flux ratio f determined, use equation (51) to calculate the drying rates.

Drying Time Equations

The drying rate equation (51) will now be used in the derivation

of an equation for the drying time. A mass balance at the interface can be written

$$\dot{\mathbf{w}} = P\rho \frac{d\delta}{dt} = P\rho \ell \frac{dz}{dt}$$
 at $\mathbf{x} = \delta$ (68)

The term $d\delta/dt$ will be recognized as the velocity of the moving interface. The right-hand side of equation (68) represents the mass rate of sublimation of the ice and the left-hand side represents the mass flux of the resulting vapor which must flow through the capillary channels of the porous dried layer to the free surface where it is removed from the system. The negative sign indicates that the vapor flow and interface velocity are in opposite directions.

Upon substituting equation (51) for the mass flux of vapor into equation (68) the following differential equation for drying time is obtained

$$\frac{1}{lz} \frac{k_D}{c} \ln \left[\frac{c}{fL} \left(T_o - T_b\right) + 1\right] = P\rho l \frac{dz}{dt}$$
(69)

rearranging,

$$dt = F \frac{z}{\ln \left[\frac{c}{fL} \left(T_{o} - T_{\delta}\right) + 1\right]} dz$$
(70)

where

$$F = \frac{P l^2 \rho c}{k_D}$$

It is shown in equation (40) that for fixed boundary conditions, the interface temperature is a function of the heat flux ratio f. Equations (58) and (65) show that the heat flux ratio f is a function of the dimensionless dried layer thickness z; therefore, it follows that both the heat flux ratio f and the interface temperature T_{δ} depend only upon the dimensionless dried layer thickness z. This permits equation (70) for the drying time to be integrated, subject to the following boundary conditions

$$z = 0$$
 at $t = 0$ (71)

$$z = 1$$
 at $t = t_{T}$ total drying time (72)

thus,

$$T = F \int_{z=0}^{z=1} \frac{z}{\ln[\frac{c}{fL}(T_0 - T_0) + 1]} dz$$
(73)

Equation (73) can be numerically integrated to give the drying time corresponding to any upper limit of z up to z = 1.

Approximate Solution for Transition Flow

Drying Rate Equations

The drying rate equations developed for the exact solution require that the interface temperatures be determined by a numerical solution of the combined energy-continuity equation (40). However, it may be observed from these calculations that the interface temperature remains nearly constant over the range of heat flux ratio $0.1 \le f \le 1$ and is essentially independent of both the heat flux ratio f and the position of the interface corresponding to the dimensionless dried layer thickness z. Also, it may be further noted that the interface temperature is established

primarily by the vacuum chamber pressure and can be estimated quite accurately by the following relationship

$$T_{\delta} \stackrel{e}{=} T_{sat.} c_{\delta}$$

where

T_{sat.} = saturation temperature of vapor corresponding to the vacuum chamber pressure

c = an empirical constant

The method used to evaluate the empirical constant c_o is given in Appendix E. Values of c_o are only slightly greater than 1.0 and are given in Table 4 of Appendix F.

The approximate solution equations which are derived for the drying rate and drying time are based upon the following approximations:

 The interface temperature remains constant for all values of the heat flux ratio f and for all values of the dimensionless dried layer thickness z.

2. The temperature gradient in the dried layer is linear and is given by the following equation

$$\frac{dT}{dx} = \frac{T_o - T_{\delta}}{\delta} = \frac{T_o - T_{\delta}}{\ell z}$$

3. The temperature gradient in the frozen layer is linear and is given by the following equation

$$\frac{d\mathbf{T}^{*}}{d\mathbf{x}} = \frac{\mathbf{T}_{\mathrm{B}} - \mathbf{T}_{\delta}}{\mathcal{L} - \delta} = \frac{\mathbf{T}_{\mathrm{B}} - \mathbf{T}_{\delta}}{\mathcal{L}(1 - z)}$$
(76)

(74)

(75)

Using approximations 1, 2, and 3, an energy balance for the entire sample considered as a control volume can be expressed by the following equation

$$- \dot{w} \left[L + c(T_{o} - T_{b}) \right] = \frac{1}{\ell} \left[\frac{k_{D}(T_{o} - T_{b})}{z} + \frac{k_{F}(T_{B} - T_{b})}{1 - z} \right]$$
(77)

Rearranging, the mass flux of vapor can be written as a function of z

$$\hat{w}(z) = \frac{1}{kL^{2}} \left[\frac{a + c_{1}^{2}}{z(1 - z)} \right] = \frac{1}{kL^{2}} \frac{X_{2}(z)}{z(1 - z)} \quad \text{for } 0 \le z \le z_{c}$$
(78)

where

$$L' = L + c(T_{0} - T_{5})$$
 (79)

$$c_1 = b - a$$
 (80)

$$a = k_{D}(T_{o} - T_{\delta})$$
(81)

$$b = k_{\rm F}(T_{\rm B} - T_{\rm b})$$
(82)

$$X_2(z) = a + c_1 z$$
 (83)

The Effect of Restricted Bottom Drying on the Drying Rate

As previously explained, after an initial period of drying the bottom and sides of the sample begin to partially dry. This reduces the overall heat transfer coefficient U with a corresponding reduction in the heat flux through the bottom of the sample and the drying rate. To take this phenomenon into account, equation (59) for the overall heat transfer coefficient U is substituted into equation (77) in place of the thermal conductivity of the frozen layer $k_{\rm F}$. Thus, the mass flux of vapor can be written in terms of the dimensionless dried layer thickness z as before, but now it will account for the effect of restricted bottom drying

$$-\dot{w} = \frac{1}{lL'} \left[\frac{a}{z} + \frac{a_2 + b_2 z}{1 - z}\right] \text{ for } z_c \le z \le 1$$
 (84)

This can be written

$$- \hat{w} = \frac{1}{\ell L^{*}} \frac{X_{3}(z)}{z(1-z)} \quad \text{for } z_{c} \le z \le 1$$
 (85)

where

$$X_3(z) = a + c_2 z + b_2 z^2$$
 (86)

$$a_2 = a_1(T_B - T_\delta)$$
 (87)

$$b_2 = b_1(T_B - T_\delta)$$
 (88)

$$c_2 = a_2 - a \tag{89}$$

Drying Time Equations

As explained in the derivation of the exact solution drying time equation, a mass balance at the interface can be expressed by the following differential equation:

 $-\dot{w} = P \rho \ell \frac{dz}{dt}, \text{ at } x = \delta$ (90)

Substituting equation (78) for the mass flux of vapor into equation (90) gives the following differential equation for drying time

$$dt = P \rho \ell^2 L \left[\frac{z}{X_2(z)} - \frac{z^2}{X_2(z)} \right] dz \quad \text{for } 0 \le z \le z_c$$
(91)

The boundary condition is

$$z = 0 \quad \text{at} \quad t = 0 \tag{92}$$

Integrating equation (91) and applying boundary condition (92) gives the following expression for the drying time as a function of the dimensionless dried layer thickness z

 $t(z) = P \rho \ell^2 K_2(z) \quad \text{for } 0 \le z \le z_c \quad (93)$

where

$$K_{2}(z) = \frac{1}{c_{1}^{3}} \left[bc_{1}z - \frac{c_{1}^{2}z^{2}}{2} - a(a+c_{1}) \ln \left(1 + \frac{c_{1}z}{a}\right) \right]$$
(94)

The drying time required to dry to the critical value of z can be calculated by evaluating equation (93) for $z = z_c$. Thus,

$$t(z_c) = P \rho l^2 L^* K_2(z_c)$$
 for $z = z_c$ (95)

Next, an equation is derived for the drying time beyond the critical value of z up to complete drying at z = 1. Equation (85) for the mass flux of vapor is substituted into equation (90) to give the following differential equation for the drying time

$$dt = P \rho \ell^2 L \left[\frac{z}{X_3(z)} - \frac{z^2}{X_3(z)} \right] dz \quad \text{for} \quad z_c \leq z \leq 1$$
(96)

The boundary condition is

$$t = t_c$$
 at $z = z_c$ (97)

Integrating equation (96) and applying boundary condition (97) gives the following expression for the drying time as a function of the dimensionless dried layer thickness z

$$t(z) = P \rho \ell^2 L^{\prime} [K_2(x_c) + K_3(z)] \text{ for } z_c \le z \le 1$$
 (98)

where

$$K_{3}(z) = \frac{b_{2}+c_{2}}{2b_{2}^{2}} \ln \frac{X_{3}(z)}{X_{3}(z_{c})} + \frac{2ab_{2}-(c_{2}^{2}+c_{2}b_{2})}{2b_{2}^{2}q'} \ln \frac{Y(z)}{Y(z_{c})} - \frac{(z-z_{c})}{b_{2}}$$
(99)

$$Y(z) = \frac{2b_2 z + c_2 - q^*}{2b_2 z + c_2 + q^*}$$
(100)

$$q' = [c_2^2 - 4ab_2]^{1/2}$$
 (101)

For complete drying z = 1 and $t = t_{\overline{1}}$. Therefore, the total drying time is determined by evaluating equation (98) for z = 1

 $t_T = P \rho l^2 L [K_2(z_c) + K_3(1)]$ for z = 1 (102)

Drying Rate and Time Equations for Various Drying Arrangements Arrangement I - Simultaneous Drying From Two Surfaces

In this arrangement simultaneous drying occurs from two surfaces by exposing the surfaces to two separate heat sources. Figure 3 shows





5. Schematic Temperature Distribution Curves Argangement I - Drying from Two Surfaces.

the arrangement for symmetrical boundary conditions.

For convenience, let the dimensionless dried layer as given by equation (104) be redefined. Thus, in this arrangement the drying rate from one side of the sample is given by the following equation

$$- \dot{w} = \frac{1}{L^6} \frac{k_D (T_o - T_\delta)}{\delta} = \frac{1}{\sqrt[6]{L^6}} \frac{2a}{z}$$
(103)

where

$$z = 2\delta/\ell \tag{104}$$

Substituting equation (103) for the mass flux of vapor into equation (90) results in the following differential equation for the drying time

$$dt = P \rho \ell^2 L, \frac{z}{4a} dz \qquad (105)$$

The boundary condition is

$$z = 0$$
 at $t = 0$ (106)

Integrating equation (105) and applying boundary condition (106) gives the following equation for the drying time as a function of the dimensionless dried layer thickness z

$$t(z) = P \rho l^2 L' K_1(z)$$
 for $0 \le z \le 1$ (107)

where

$$\zeta_1(z) = \frac{z^2}{8a}$$
 (108)

For complete drying z = 1 and $t = t_T$. Therefore, the total drying time is calculated by evaluating equation (107) for z = 1

$$t_{T} = P \rho l^{2} L^{*} K_{1}(1)$$
 for $z = 1$ (109)

It is interesting to note that for Arrangement I the drying rates and drying time are independent of the frozen layer thermal conductivity. The frozen layer is adiabatic in this arrangement due to the symmetry of the temperature gradients which cause the temperature gradient in the frozen layer to be zero. This is unfortunate because it fails to make use of the high thermal conductivity of the frozen layer which is approximately twenty times greater than the thermal conductivity of the porous dried layer.

Arrangement II - Primary Drying from One Surface with Restricted Bottom Drying

In this arrangement a vapor barrier on the bottom and sides of the sample causes the drying to occur primarily from the top. Restricted bottom drying is assumed to begin at the critical value of z. Figure 4 shows typical boundary conditions for this arrangement. The drying rate and drying time equations, previously derived, are repeated for convenience.

$$- \dot{w} = \frac{1}{\sqrt{L}} \frac{X_2(z)}{z(1-z)} \quad \text{for } 0 \le z \le z_c \quad (78)$$

$$-\dot{w} = \frac{1}{L} \frac{X_3(z)}{z(1-z)} \quad \text{for } z_c \le z \le 1 \quad (85)$$



Figure 4. Schematic Temperature Distribution Curve Arrangement II - Restricted Bottom Drying.

$$t(z) = P \rho \ell^2 L^* K_2(z) \qquad \text{for } 0 \le z \le z_c \qquad (93)$$

$$t(z) = P \rho l^2 L^{*} [K_2(z_c) + K_3(z)] \text{ for } z_c \le z \le 1$$
 (98)

Arrangement III - Drying from One Surface with Zero Bottom Drying

In this arrangement a vapor barrier on the bottom and sides of the sample causes the drying to occur only from the top with zero bottom drying assumed, thus making full use of the high thermal conductivity of the frozen bottom layer. Figure 5 shows typical boundary conditions for this arrangement. The drying rate and drying time equations, previously derived, are repeated for convenience

$$- \dot{w} = \frac{1}{U_{L}} \frac{X_{2}(z)}{z(1-z)} \qquad \text{for } 0 \le z \le 1$$
 (78)

$$t(z) = P \rho l^2 K_2(z)$$
 for $0 \le z \le 1$ (93)

Results of Exact Theoretical Analyses Solution Exact Interface Temperatures

Interface temperatures were calculated which satisfy the combined energy-continuity equation (40) for various assigned constant values of the heat flux ratio f. These calculations were made on a digital computer using Newton's method. One set of calculations was made using the interface saturation pressure-temperature relationship for frozen pure water, while another set was made using the saturation pressure-temperature experimental data for frozen meat juices. The results of these calculations for constant vacuum chamber pressures of 0.5, 2, and 3 torr are given in Tables 1 and 2. It may be noted that



Figure S. Schematic Temperature Distribution Curve Arrangement III - Zero Bottom Drying.

- - -

Heat Flux Ratio, f	po = 0.5 torr	po = 2 torr	po = 3 torr
1.0	448.14	474.46	482.89
.9	448,47	474.54	482.93
.8	448.88	474.64	482.98
.7	449.'37	474.76	483.05
.6	449.97	474.93	483.13
15	450.78	475.16	483.25
.4	451.89	475.84	483.42
.3	453,54	476.00	483.70
•2	456.29	476.95	484.23
.1	462.08	479.32	485.59

Table 1. Exact Solution Interface Temperatures, of for Frozen Water Vapor Pressure	°R*

* Calculations are based on:

- Using Equation (40).
 Using saturation pressure-temperature data for frozen water, see reference (7).
 T_o = 575°R.

Heat Flux Ratio, f	po = 0.5 torr	po = 2 torr	po = 3 torr
1.0	455,23	477.58	486.32
.9	455.51	477.65	486.36
.8	455+85	477.77	486.41
.7	456.26	477.82	486.48
•6	456.76	477.95	486.57
.5	457.44	478.12	486.69
.4	458.38	478.38	486.86
•3	459 .78	478.79	487.14
.2	462.11	479.55	487,68
.1	467.12	481.43	489.05

Table 2. Exact Solution Interface Temperatures, oR for Frozen Beef Juice Vapor Pressures

* Calculations are based on:

- 1. Using equation (40).
- 2. Using saturation pressure-temperature relationship for
 - frozen beef. See reference (7).

.

3. $T_0 = 575^{\circ}R$

the interface temperatures for frozen pure water are lower than the interface temperatures for frozen meat juices.

Dimensionless Pressure and Dimensionless Temperature Distributions

The dimensionless pressure and dimensionless temperature ratios were calculated by incorporating equations (41) and (42) into the previously mentioned digital computer program. Figures 6, 7, and 8 show both the dimensionless pressure and dimensionless temperature distributions in the porous dried layer are approximately linear with respect to the dimensionless distance measured in the dried layer and essentially independent of the heat flux ratio f. This implies that the temperature distribution in the dried layer is essentially independent of the bottom surface temperature T_B . The explanation for this can best be made by rearranging the heat flux ratio equation (12) into the following form

(110)

(111)



Substituting equation (76) for the frozen layer temperature gradient into equation (110) gives the following expression for the heat flux ratio f:

 $f = \frac{1}{\frac{k_F(T_B - T_{\delta})}{\frac{\ell(1 - z)}{-k_D \frac{dT}{dx}}}}$







Fig. 7. Dimensionless Pressure Distributions and Dimensionless Temperature Distributions.

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For fixed values of z, δ , ℓ , $k_{\rm F}$, $k_{\rm D}$, T_{δ} , and dT/dx at $x = \delta$, it is apparent from equation (111) that an increase in the bottom surface temperature $T_{\rm B}$ will decrease the heat flux ratio f. Therefore, since the dimensionless temperature distribution is essentially independent of the heat flux ratio f it is also essentially independent of the bottom temperature $T_{\rm B}$.

Dimensionless Interface Pressure and Dimensionless Interface Temperature

The interface dimensionless pressure p_{δ}/p_{0} and dimensionless temperature T_{δ}/T_{0} both correspond to the dimensionless distance measured in the dried layer $X^{*} = 1$ in equations (41) and (42). A plot of interface dimensionless pressure, dimensionless temperature, and heat flux ratio are shown in Figures 9, 10, and 11 for constant vacuum chamber pressures of 0.5, 2, and 3 torr for arrangement II. These figures show that the interface dimensionless temperatures are essentially independent of both the heat flux ratio and dimensionless dried layer thickness (which is a measure of the interface position). This indicates that for a given vacuum chamber pressure the interface temperature is essentially independent of both the interface position and the bottom surface temperature.

The essentially constant behavior of the interface temperature as the interface moves through the sample from top to bottom, coupled with the approximately linear temperature distribution in the porous dried layer, led to the formulation of the approximate analyses solution which makes use of both these approximations.

The first mention of the essentially constant behavior of the interface temperature was made by Kessler (11) based upon his experimental


Fig. 9. Dimensionless Interface Pressure, Dimensionless Interface Temperature and Heat Flux Ratio- Arrangement II.









measurements of the temperature distributions during the freeze drying of a variety of materials. More recently this phenomenon was also observed by Hatcher (12) in his experiments on the freeze drying of beef.

<u>Comparison of Exact and Approximate Theoretical</u> Analyses Drying Rates

The exact and approximate solution drying rates were calculated for arrangement II in which restricted bottom drying occurs. Figures 12, 13, and 14 show both exact and approximate drying rates vs. dimensionless dried layer thickness for constant vacuum chamber pressures of 0.5, 2, and 3 torr. The closeness of these drying rate curves further strengthens the validity of the approximate solution. For this reason the approximate solution equations were used in all other calculations.

The principal advantages of the approximate solution are:

 It does not require a computer solution to determine the interface temperatures.

2. It gives drying rates which agree very closely with those calculated using the more time-consuming exact solution equations.

3. It gives a closed-form solution for drying time equations, thus avoiding a lengthy numerical integration.

Results of Approximate Analyses Solution

The Effect of Vacuum Chamber Pressure

The dimensionless dried layer thickness vs. drying time curves for constant vacuum chamber pressure of 0.5, 2 and 3 torr are shown in Figures 15, 16, and 17 for drying arrangements I, II, and III respectively.



Fig. 12. Comparison of Exact and Approximate Solution Drying Rate Curves - Arrangement II.







Fig. 14. Comparison of Exact and Approximate Solution Drying Rate Curves - Arrangement II.

The boundary temperatures are the same for all curves shown in these figures. It is obvious that lowering the vacuum chamber pressure decreases the drying time in all arrangements. The reasons for this can best be explained by the following analysis. The approximate drying rate equation (77) can be written in the following generalized form to cover all arrangements

$$- \hat{w} = \frac{b_1}{l L} \left[\frac{k_D (T_0 - T_\delta)}{z} + \frac{U(T_B - T_\delta)}{1 - z} \right]$$
(112)

where

- 1. For Arrangement $I^* = U(T_B T_\delta) = 0$, for $0 \le z \le 1$
- 2. For Arrangement II

$$U = k_F; \quad \text{for } 0 \le z \le z_C$$
$$U = a_1 + b_1 z; \quad \text{for } z_C \le z \le 1$$

3. For Arrangement III U = k_F , for $\Theta_z \le z \le 1$

For fixed boundary temperatures T_0 and T_B , the generalized drying rate equation (112) may be expressed in terms of the following variables

 $\dot{w} = \dot{w} (k_{\rm D}^{\rm }, U, z, T_{\rm S}^{\rm })$ (113)

The effect of the various variables in equation (113) upon the drying rate are shown by the following relationships

1.
$$\hat{w} \sim 1/T_{h} \sim 1/p_{o}$$
 (114)

2. $\dot{w} \sim 1/z$ (115)

*For Arrangement I, equation (112) gives the drying rate for only one side.

Boundary Temperatures $T_0 = 575 \ ^{O}R$ $T_B = 492.5 \ ^{O}R$.2 ŧ٩ Mass Fraction of Sublimated Ice, .4 3 torr = .6 2 torr = 0.5 Po torr .8 1.0 10 0 20

0

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Drying Time, hours

64





ŝ





3.
$$\dot{w} \sim k_{\rm D} = k_{\rm s} + \frac{k_{\rm VO}}{1 + C/p_{\rm o}}$$
 (116)

4.
$$\dot{w} \sim k_F \sim \frac{1}{(T_B + T_\delta)/2}$$
 (117)

Relationship (1) shows that lowering the vacuum chamber pressure reduces the interface temperature and thus increases the drying rate.

Relationship (2) shows that as the dimensionless dried layer thickness increases the drying rate decreases.

Relationship (3) indicates that lowering the vacuum chamber pressure reduces the thermal conductivity of the vapor-filled porous dried layer and thus decreases the drying rate.

Relationship (4) indicates that lowering the interface temperature, by lowering the vacuum chamber pressure, decreases the meantemperature of the frozen layer. This increases the thermal conductivity of the frozen layer and thereby increases the drying rate.

Relationships (1) and (4) indicate that lowering the vacuum chamber pressure tends to increase the drying rate, while relationship (3) shows that lowering the vacuum chamber pressure tends to decrease the drying rate. It is interesting to note that the drying time curves shown in Figures 15, 16, and 17 indicate that the net tendency is for the drying time to be decreased as the vacuum chamber pressure is reduced.

It is interesting to note that the effect of reducing the vacuum chamber pressure upon reducing the drying time is much more pronounced for arrangements II and III than for arrangement I. The explanation for this can be understood by referring to the generalized drying rate

equation (112). It is obvious that reducing the vacuum chamber pressure will reduce the interface temperature, according to equation (40), which will increase the heat fluxes. In arrangement I the increase in heat flux will apply only to the dried layer since for this arrangement the heat flux through the frozen layer is zero because dT/dx is zero in the frozen layer. This causes the overall heat transfer coefficient U in effect to be zero for this arrangement. However, in arrangements II and III the overall heat transfer coefficient U is greater than zero, thus $U \leq k_F \approx 20 k_D$. Therefore, in arrangements II and III a reduction in the interface temperature will be approximately twenty times more effective in increasing the drying rate than for the same interface temperature reduction in the case of arrangement I.

The Effect of Boundary Temperatures

It is obvious from equation (112) that the drying rate will tend to increase as the top surface temperature T_{o} is increased and/or as the bottom surface temperature T_{B} is increased.

The maximum top surface temperature T_{o} is limited to approximately 575°R (115°F) in order to prevent the product from having a scorched taste.

Undoubtedly there is also an upper limit for the bottom surface temperature T_B , but in most usual freeze-drying process the actual bottom temperature is generally less than or equal to the top surface temperature. During the various tests conducted in this investigation the bottom surface temperature was not controlled and increased with time for each test.

By referring to equation (112) it may be noted that a given

increase in the bottom surface temperature T_B is some twenty times more effective in increasing the drying rate than for the same increase in the top surface temperature T_o . This is true because the thermal conductivity of the frozen layer is approximately twenty times greater than the thermal conductivity of the vapor-filled porous dried layer.

The effect of increasing the bottom surface temperature upon both the drying rate and the drying time is shown by Figures 18 and 19. These curves are based upon drying arrangement II with restricted bottom drying, a vacuum chamber pressure of 2 torr and a top surface temperature of 575°R. In one case the bottom surface temperature is held constant at 470°R, while in the other case the bottom surface temperature is held constant at 492.5°R. The increase in drying rates and corresponding decrease in drying time is rather remarkable since the bottom surface temperature is increased by only 22.5°F.

The Effect of Drying Arrangement

The three different drying arrangements considered in the approximate theoretical analyses solution are briefly described below.

- 1. Arrangement I Simultaneous drying from two surfaces.
- Arrangement II Primary drying from one surface, with restricted bottom drying.

 Arrangement III - Drying from one surface with zero bottom drying.

The effect of drying arrangement upon the drying time is shown in Figures 20, 21, and 22 for constant vacuum chamber pressures of 0.5, 2, and 3 torr respectively. In all cases the top surface temperature is 575°R while the bottom surface temperature is 492.5°R.



Arrangement II.





-1

These curves indicate that arrangement III gives the highest drying rate and fastest drying time for vacuum chamber pressures of 0.5 and 2 torr. However, in an actual freeze-drying process it would be difficult, if not impossible, to avoid some degree of bottom drying. Therefore, arrangement II appears to be a more realistic model than arrangement III. In the early stages of drying, arrangement I gives the highest drying rates and fastest drying time because of simultaneous drying from two surfaces. Eventually the insulating effect of two dried layers causes the drying rate to decrease sharply.

The difference in behavior of the drying rate curves for arrangements II and III compared to arrangement I can easily be explained by referring to the generalized drying rate equation (112). For arrangements II and III it will be noted that the following relationships hold

> (1) $\dot{w} \rightarrow \infty$ as $z \rightarrow 0$ (2) $\dot{w} \rightarrow \infty$ as $z \rightarrow 1$

Thus, for arrangements II and III the drying rate is very high at the beginning and end of the freeze-drying process. However, for arrangement I, which involves simultaneous drying from two surfaces, the heat flux occurs only through the two dried layers and in effect the overall heat transfer coefficient U is zero. By referring to equation (112) it will be noted that for arrangement I the drying rate decreases continuously as the dimensionless dried layer thickness increases and toward the end of the freeze-drying process the drying rate approaches a finite limit. Thus, for arrangement I the following relationships hold

Mass Fraction of Sublimated Ice,





Mass Fraction of Sublimated Ice,









(1) $\dot{w} \rightarrow \infty$ as $z \rightarrow 0$ (2) $\dot{w} \rightarrow \frac{2k_D(T_o - T_\delta)}{\ell L'}$ as $z \rightarrow 1$

Referring to Figures 20, 21, and 22 which show the drying time curves for constant vacuum chamber pressures of 0.5, 2, and 3 torr respectively, it is apparent that arrangement III with zero bottom drying gives the fastest drying time for 0.5 and 2 torr vacuum chamber pressures. For vacuum chamber pressures of 0.5 and 2 torr, arrangement II with restricted bottom drying gives a faster drying time than arrangement I. Finally, for a vacuum chamber pressure of 3 torr, arrangement I with simultaneous drying from two surfaces gives a faster drying time than arrangement II. The reason for arrangements I and II behaving in this way is due to the fact that arrangement II is much more sensitive than arrangement I to changes in the interface temperature resulting from changes in the vacuum chamber pressure.

Saturation Pressure-Temperature Relationship for Frozen Meat Juices

Experimental evidence indicates that the ice formed by the frozen meat juices in the sample requires a higher sublimation temperature than for frozen pure water at the same pressure. Thus, for a given vacuum chamber pressure the corresponding interface saturation temperature for frozen meat juices will be higher than for frozen pure water. This tends to reduce the drying rates and to increase the drying time compared with frozen pure water. This phenomenon is illustrated by Figures 23 and 24 which show the drying rates and drying time for a vacuum chamber pressure of 2 torr, top surface temperature of 575°R, and bottom surface temperature of 492.5°R. The saturation pressure-temperature



Fig. 23. Comparison of Drying Rates Using Vapor Pressure for Frozen Pure Water and Frozen Meat Juices - Based on Approximate Solution for Arrangement II.





relationship for frozen pure water is used in one case and for frozen meat juices in the other case.

<u>Conclusions from the Theoretical Analyses</u>

The principal conclusions drawn from the theoretical analyses are:

 For a given vacuum chamber pressure, the interface temperature is essentially constant and independent of both the interface position and bottom surface temperature.

2. The approximate solution equations give drying rates very close to the drying rates using the more time-consuming exact analyses equations; and permit closed-formed equations for drying time, thus avoiding a numerical integration of the exact solution drying time equation.

3. Based upon the approximate theoretical solution equations, lowering the vacuum chamber pressure tends to lower the interface temperature, to increase the drying rate, and to decrease the drying time.

4. Based upon the approximate theoretical solution equations, increasing the bottom surface temperature has little effect upon the interface temperature, substantially increases the drying rate, and substantially reduces the drying time.

5. Based upon the approximate theoretical solution equations, arrangement III with drying from one side and zero bottom drying, in general, gives the highest drying rates and fastest drying time. Arrangement II, in which primary drying is from one side only with restricted bottom drying, appears to be a more realistic model than arrangement III.

6. Based upon the approximate theoretical solution equations, the drying rates are reduced and the drying time increased when using the saturation pressure-temperature experimental data for frozen meat juices compared with calculations using the saturation pressure-temperature relationship for frozen pure water.

CHAPTER III

INSTRUMENTATION AND EQUIPMENT

Equipment

The principal equipment involved in the experimental portion of this investigation consists of a vacuum chamber and condenser mounted on a steel supporting frame and a vacuum pump. The general arrangement of equipment is shown in Figure 25. Specifications for all equipment and instruments are given in Appendix H.

Vacuum Chamber and Heater

The vacuum chamber consists of a two-foot cubical chamber made of 1/4 inch steel plate welded to an internal frame made of angle sections. The door is made of a single piece of 1/2 inch steel plate in which two four-inch square observation ports are cut and covered with 3/8 inch plexi-glass. A gasket made of a single piece of 1/4 inch thick soft natural rubber cut in the shape of a picture frame is glued to the door-side frame of the vacuum chamber to provide a vacuum-tight seal for the door. The hinges are slotted thus permitting the door to slide forward when tightened by the twenty 3/8 inch tie-down bolts located around the edges of the door and passing through an external flange on the vacuum chamber.

A plate type 110 volt, 1700 watt electric radiant heater 11 by 15 inches, mounted in the ceiling of the vacuum chamber, provides the heat of sublimation during freeze-drying.



Figure 25. Schematic Diagram of Freeze-Drying Test Equipment.

<u>Condenser</u>

The primary function of the condenser is to serve as a cold trap between the vacuum chamber and the vacuum pump. This greatly reduces the volume of gases handled by the pump and reduces the possibility of water vapor condensing and contaminating the pump oil. This enables much lower vacuum chamber pressures than otherwise would be possible.

The vapor released during the freeze-drying process, along with air leakage, leaves the vacuum chamber through a six-inch pipe leading to the condenser. The condenser is a heat exchanger made of two concentric cylindrical drums with the inner drum open at the top where dry ice and acetone are introduced. This reduces the temperature to approximately -100°F and provides a surface temperature well below the freezing point of the vapor so that it undergoes a vapor-solid phase change and remains on the outer surface of the inner drum. After completion of a test the accumulated ice is allowed to melt and is drained from the bottom of the larger drum. An acetone drain line is connected to the inner drum.

Vacuum Pump

A mechanical rotary type vacuum pump removes non-condensible gases from the system and discharges them to the atmosphere and thus maintains the vacuum.

Instrumentation

The Sample

The sample consisted of a four-inch diameter disc of loin of beef 1-1/4 or 1-3/8 inches thick which was purchased from a local retail

market. The circular sample was chosen to eliminate corners which would tend to dry more rapidly than other parts of the edge of the sample due to their higher surface area-volume ratio. In order that the heat and mass fluxes be nearly one-dimensional, a vapor barrier of aluminum foil was used to cover the bottom and edges of the sample, thus permitting only upward flow of the sublimated vapor. A 3/4 inch layer of fiberglass insulation was wrapped around the circumference of the sample to minimize the radial heat flux.

Pressure and temperature probes were inserted radially to a depth of one inch as described in more detail later. The entire sample assembly was placed on a six-inch diameter disc of 1/2 inch thick fiberglass insulation resting on top of the precision balance. Figure 26 shows a typical sample assembly.

Pressure Measurements

The vacuum chamber pressure was determined by a direct-reading diaphragm type mechanical absolute pressure gage which is self-compensating for variations in both ambient pressure and temperature. The gage is calibrated in 0.1 torr increments from 0.2 to 20 torr.

Vapor pressure measurements in the range of 0.1 to 3 torn present a rather difficult problem because the range is low for mechanical type gages, but high for most ionization, thermocouple and other similar type gages. Additional difficulties are encountered in gages where the vapor could condense and cause trouble, or where the composition of the air-vapor mixture must be known in order to use the proper calibration data for the gage. This later problem made the use of an Alphatron type gage inconvenient and uncertain. For these reasons the mechanical



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diaphragm type gage seems to be the best suited gage for measuring vapor pressures down to 0.2 torr.

The vapor pressures in the dried layer of the sample were measured by mechanical diaphragm type differential pressure gages located inside the vacuum chamber to eliminate the possibility of atmospheric air leakage into the pressure-measuring system. Hypodermic needles were used for pressure probes. The probes were inserted into the sample, after it had been partially frozen, with a wire inserted in each needle slightly beyond the tip in order to prevent plugging by meat particles. After complete freezing, the wires were retracted. The probes were connected to the high-pressure sides of the differential pressure gages by 3/16 inch rubber tubing; the low-pressure sides of the gages were open to the vacuum chamber pressure. To assure accurate probe location a special jig shown in Figure 27 was used to locate the probes radially at equal intervals and vertically at different depths below the drying surface of the sample.

The differential pressure gages are known as the "magnehelic" type. In this design the moving diaphragm is magnetically coupled to the pointer spindle. This magnetic coupling is fortunate because initially the differential pressure is approximately one atmosphere due to air trapped between the dead-end pressure probe in the frozen sample and the differential pressure gage. The differential pressure thus exceeds the capacity of the gage until drying of the sample allows the trapped air to escape up through the porous dried layer, after which the true differential pressure between the vapor and the vacuum chamber is indicated by the gage.



Temperature Measurements

All temperature measurements were made using copper-constantan thermocouples which were extended through the vacuum chamber wall using standard vacuum type feed-throughs. All thermocouples were led to a multi-point switch which was connected to a potentiometer having a reversing polarity switch. A 32°F reference junction was provided by crushed ice and water in a thermos bottle.

Probes for temperature measurements consisted of fine copperconstantan wires threaded through a hypodermic needle and connected at the tip of the probe to form the hot junction. The temperature probes were located radially and vertically using the same jig used for locating the pressure probes. The temperature probes, like the pressure probes, were frozen in place in the sample. Quick-connect miniature plugs and jacks permitted the short thermocouple leads extending from the sample to be connected to permanent thermocouple leads located inside the vacuum chamber.

In addition to the temperatures measured by the probes, other temperatures measured included the heater surface, the condenser surface, the vacuum chamber atmosphere and the top and bottom surfaces of the sample.

Weighing Balance

A precision indicating balance supporting the sample, thermocouple lead wires, and rubber tubing, was used to determine the rate of sublimation by loss of gross weight. The balance was calibrated in increments of one gram from zero to 2,000 grams. Estimates of 0.1 gram could easily be made. The balance dial was illuminated and was observed through a port in the door.

Pressure and Temperature Control

The vacuum chamber pressure was maintained constant throughout a given test by manual operation of an air bleed valve. Air was bled into the system between the condenser and vacuum pump thus permitting the vacuum chamber pressure to be regulated and held constant.

The top surface temperature of the sample was held constant throughout a given test by varying the power input to the heater. This was accomplished by manual operation of a variac unit to vary the input voltage and thus the input power to the heater.

Estimate of Instrument Errors

The absolute pressure gage used to measure the vacuum chamber pressure was new and its calibration curve, traceable to the NBS, was furnished by the manufacturer, and was applied to all gage readings obtained in the various tests. Thus the maximum error in the measurement of the vacuum chamber pressure is estimated to be ±0.01 torr.

The differential pressure gages were carefully calibrated before each test using an inclined manometer with minor division of .01 inches of water. Thus it is estimated that the maximum error in the differential pressure measurements was ± 0.01 inches of water or ± 0.02 torr.

All thermocouples were used with a 32°F reference junction temperature maintained by an ice-water bath. The potentiometer is calibrated in increments of 0.005 millivolts. Thus the maximum error in the various temperature measurements is estimated to be ±0.25°F.

The precision balance is calibrated in one gram increments which easily could be interpolated within 0.1 gram. Also the balance indicator and scale are in the same plane thus avoiding parallax error. In addition, the balance was checked at the beginning of each test by placing known weights on the balance and adjusting the scale for zero error. Thus the estimated maximum error in weighing is estimated to be ± 0.1 gram.

CHAPTER IV

PROCEDURE

A number of 24-hour tests were conducted at constant vacuum chamber pressures during which the temperature of the top surface of the sample was held constant near the highest permissible level. If the top surface temperature is excessive the sample may become scorched.

Pre-test Procedure

As previously mentioned, the samples were cut from a 1-1/4 or 1-3/8 inch thick slab of loin of beef purchased from a local retail market. The entire slab of beef was partially frozen in a domestic type deep freezer to a semi-rigid condition to facilitate handling and cutting. A four-inch diameter test section was selected to avoid inclusion of bone and/or fat portions.

As previously mentioned, wires were inserted inside the hypodermic needle pressure probes and extended slightly beyond the tips in order to prevent plugging by meat particles. The temperature probes were also made of hypodermic needles threaded with fine copper-constantan thermocouple wires with the hot junction formed at the tip of the probe.

The partially frozen sample was placed face down on the probe jig shown in Figure 27 and the aluminum foil vapor barrier pressed in place around the bottom and edges. Next, the pressure and temperature probes were inserted to a depth of one inch into the sample, which was
subsequently returned to the freezer for final over-night freezing.

Test Procedure

Prior to the start of each test dry ice and acetone were placed into the condenser, the thermocouple reference junction ice-water bath was prepared, and all instruments, which had been previously calibrated, were checked out.

The sample assembly was transferred from the freezer to the weighing balance inside the vacuum chamber; and as quickly as possible the pressure probe tubing was connected to the differential pressure gages and the thermocouple leads from the sample were connected to the permanent leads inside the vacuum chamber. Next, the door was secured and the vacuum pump was started which pulled the vacuum chamber pressure down to the test level in approximately five to ten minutes. An initial survey of temperatures was made. Then the heater was turned on to full 110 volts at which time the test was officially begun.

All data was observed and recorded at 15-minute intervals. However, in the early stages of the test the top surface temperature of the sample was frequently monitored to prevent over-heating of the top surface which could occur with full heater voltage.

The air bleed valve was manually adjusted as required to maintain constant vacuum chamber pressure. After approximately 15 to 20 minutes it became necessary to adjust the variac to reduce the heater voltage in order to maintain the top surface of the sample at a constant temperature.

The remainder of the 24-hour test was generally uneventful except for manipulation of the variac and air bleed valve, taking data, and the addition of dry ice to the condenser at intervals of two to three hours.

Post-test Procedure

After 24 hours the official test ended, but in most instances drying was continued for several hours longer without recording additional data except the gross weight. At the end of this period the pump was stopped, the door unbolted and the sample removed. The insulation, vapor barrier, and probes were removed from the sample which was then bisected to determine the extent of bottom drying. After this, the bare sample was returned to the vacuum chamber for complete drying for another 10 to 20 hours, with no additional data obtained except the final net weight of the completely dry freeze-dried sample.

CHAPTER V

DISCUSSION OF RESULTS

<u>General</u>

Two series of 24-hour: tests were made at various constant vacuum chamber pressures. Only the results of Test Series II are presented because of vapor pressure measurement difficulties encountered in Test Series I.

t Series I	<u>lest Series II</u>	
Pressure, torr	Test Number	Pressure, torr
0.01	7	0.5
0.1	8	2.0
0.1	9	3.0
1.0	н. 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977 - 1977	
1.0		
1.0 · · · ·		
	<u>Pressure,</u> <u>torr</u> 0.01 0.1 0.1 1.0 1.0 1.0	torr Test 0.01 7 0.1 8 0.1 9 1.0 1.0 1.0 1.0

Pressure Measurements

In Test Series I considerable difficulty was encountered in making accurate vapor pressure measurements in the porous dried layer. The general arrangement of the vapor pressure measuring system is shown in Figure 28.

Initially air at atmospheric pressure is trapped in the connecting line between the probe and the differential pressure gage. When the vacuum chamber pressure is reduced to the desired test level the differential pressure acting on the gage is one atmosphere (approximately 760



Fig. 28. Schematic Diagram of Vapor Pressure Measuring System.

torr). This differential pressure is much greater than full scale capacity of the gage (approximately 0.95 torr). Fortunately, however, the differential gage diaphragm is magnetically coupled to the pointer thus preventing damage to the gage. When the porous dried layer reaches the vicinity of the probe the trapped air begins to escape and eventually the true differential pressure between the water vapor and the vacuum chamber is indicated on the gage.

After a number of 24 hour tests had been completed it was observed that the differential pressure gage reading was greater than zero even when the pressure probe was exposed to the vacuum chamber directly. This indicated that $p_{\chi 2} > p_{\chi 1}$ which would result in an inaccurate measure of the true vapor pressure $p_{\chi 1}$. Originally it was thought that perhaps some residual lubricant or other contaminant in the measuring system might be out-gasing thus giving rise to a vapor pressure on the high-pressure side of the gage. After investigating many schemes in an attempt to solve the problem, it was discovered that if larger diameter hypodermic needle probes and larger diameter tubing were used that the differential pressure gage would read zero with the problem of accurate vapor pressure measurements, Test Series II was successfully completed using the larger diameter hypodermic needle probes and larger diameter tubing listed in Figure 28.

Pressure Distributions

As previously mentioned, Test Series II consisted of three 24-hour tests at constant vacuum chamber pressures of 0.5, 2, and 3 torr for

which the actual pressure distribution curves, with time as a parameter, are shown in Figures 29, 30, and 31. The pressure curves in each case are extended to the calculated position of the interface which is indicated by a vertical dashed line. It may be observed that the pressure gradients decrease with increasing time. This is due to the decrease in both the mass and volume flow rates of vapor with time. In all tests the maximum differential pressure between the interface and vacuum chamber ranges from 0.15 to 0.20 torr and is approximately the same for all three tests. The characteristic of the rising interface pressure with dried layer thickness and time is evident.

A comparison of the actual and theoretical dimensionless pressures versus dimensionless distance in the dried layer are shown in Figures 32, 33, and 34 for vacuum chamber pressures of 0.5, 2, and 3 torr. Points representing actual dimensionless pressures fall well within the range of the theoretical dimensionless pressure curves calculated using equation (42) for various constant values of the heat flux ratio f.

All these observations concerning a comparison of the actual and theoretical dimensionless pressures tend to substantiate the general validity of the theoretical analyses.

Temperature Distributions

The actual temperature distributions using time as a parameter are shown in Figures 29, 30, and 31 for pressures of 0.5, 2, and 3 torr. The surface temperature in all cases is constant at 575°R (115°F); however, since the bottom temperature is uncontrolled, it increases with







Fig. 31. Measured Pressure and Temperature Distributions.













time during a given test, and the time-mean average bottom temperature varies between different constant vacuum chamber pressure tests. In fact, all temperature distribution curves show a rising characteristic with time.

A comparison of the actual and theoretical dimensionless temperatures versus dimensionless distance in the dried layer, for various constant values of the heat flux ratio f, are shown in Figures 32, 33, and 34 for vacuum chamber pressures of 0.5, 2, and 3 torr. These curves apply only to the dried layer and do not show the dimensionless temperatures in the frozen layer where the theoretical temperature distribution is assumed to be linear. The actual dimensionless temperatures exhibit a dropping characteristic with increasing dimensionless distance measured in the dried layer which are similar to the theoretical dimensionless temperature curves calculated using equation (41) for various constant values of the heat flux ratio f.

Curves showing actual temperatures measured by various probes are shown as a function of time in Figures 35, 36, and 37 for vacuum chamber pressures of 0.5, 2, and 3 torr. As an example, it is interesting to observe the temperatures measured by the probe at $x = 3/4^{\circ}$ shown in Figure 37. It can be noted that as long as the probe is in the frozen layer, indicated by the solid portion of the curve, its temperature remains essentially constant. However, when the interface reaches the probe the temperature at this position begins to increase rather sharply with time.













Drying Rates

Actual and theoretical drying rate curves for arrangements I and II are shown in Figures 38, 39, and 40 for vacuum chamber pressures of 0.5, 2, and 3 torr. The actual drying rates for pressures of 0.5 and 2 torr compare quite well with the theoretical drying rates based on arrangement II; however, for a pressure of 3 torr the agreement is less satisfactory. In part, this may be attributed to the greater radial heat flux for the 3 torr pressure compared to the lower pressure tests. At least one factor contributing to the higher radial heat flux is that the thermal conductivity of the fiberglass insulation on the bottom and sides of the sample is pressure dependent as shown by the following equation:

$$k_{1} = k_{s} + \frac{k_{vo}}{1 + C/p_{o}}$$

(118)

where

- i = thermal conductivity of the composite porous insulation and gas-filled void volume
- k = thermal conductivity of the solid part of the insulation
 k = thermal conductivity of the gas at a standard reference
 vo pressure
- C = experimentally determined constant

As the chamber pressure is increased the contribution of the thermal conductivity of the gases in the void volume of the insulation increases, thus increasing the composite thermal conductivity of the insulation.

See reference (4).







Fig. 40. Actual and Theoretical Drying Rate Curves.

 \mathbf{m}

The theoretical models for all three drying arrangements I, II, and III assume zero radial heat flux; this will tend to cause the theoretical drying rates to be less than the actual drying rates where the radial heat flux is greater than zero. The deviation between actual and theoretical drying rates is less pronounced for vacuum chamber pressures of 0.5 and 2 torr because of the greater insulating effect of the fiberglass insulation which tends to reduce the radial heat flux compared with the 3 torr pressure test.

Drying Time

Figure 41 shows the drying time curves for vacuum chamber pressures of 0.5, 2, and 3 torr. The drying time curve for 2 torr is the fastest while the drying time curve for 0.5 torr is the slowest. At first glance this appears to contradict the theoretical analyses. However, the paradox is made clear by a close inspection of the various boundary conditions for each test. The 2 and 3 torr pressure tests have essentially the same boundary temperatures. The top surface temperatures are both 575°R (115°F) while the bottom temperatures are 492.5°R and 495.0°R respectively. The improvement of drying time with the lower vacuum chamber pressure is in agreement with theoretical considerations. This agreement would have been even more pronounced except for three factors which tend to reduce the drying time for the 3 torr pressure test below normal. First, the radial heat flux for the 3 torr test is greater because of the decrease in the insulating effect of the fiberglass around the edges of the sample. Next, the sample thickness for the 3 torr test is greater than for the 0.5 and 2 torr tests, being 1-3/8





inches for the 3 torr test and only 1-1/4 inches for the other two tests. This increases the relative radial area and thus increases the relative radial heat flux. Finally, the bottom temperature is higher being 495°R for the 3 torr test and 492.5°R for the 2 torr test. The difference of only 2.5°R in bottom temperatures may not seem significant; however, it does make a substantial difference in the heat flux through the bottom. The effect of bottom temperature upon the theoretical drying time for arrangement II is illustrated by Figure 19 which shows that the bottom temperature plays a very important role in the drying time.

The 0.5 torr test drying time curve is slower than the higher pressure test primarily because the bottom surface temperature is 22.5 and 25°R lower than for the 2 and 3 torr test respectively. This sharply reduces the bottom heat flux with a corresponding reduction of the drying rate and lengthening of the drying time. In addition to this factor, the insulating effect of the fiberglass insulation around the edge of the sample tends to increase with lower vacuum chamber pressure thus reducing the relative radial heat flux compared to the higher vacuum chamber pressure tests.

A comparison of actual and theoretical drying time curves are shown in Figures 42, 43, and 44 for vacuum chamber pressures of 0.5, 2, and 3 torr. The 0.5 torr test actual drying time curve coincides most of the time with the theoretical drying time curve based on arrangement II for restricted bottom drying. The 2 torr test actual drying time curve is bounded by the theoretical drying time curves corresponding

n tra i v













to arrangements II and III which are for restricted bottom drying and zero bottom drying respectively. Finally the 3 torr test shows a faster actual drying time curve than the theoretical drying time curves corresponding to arrangements II and III, but is bounded between the theoretical drying time curves for arrangements I and III which are for simultaneous drying from two surfaces and zero bottom drying respectively. It is obvious that for the lower vacuum chamber pressure tests the actual drying time curves agree more satisfactorily with the theoretical drying time curves than does the higher 3 torr test. This has already been explained to be due, in part, to the relatively greater radial heat flux for the 3 torr test compared to the lower pressure tests. This is especially significant since the theoretical analyses assume zero radial heat flux.

Actual Bottom Drying

In the theoretical analyses the effect of restricted bottom drying upon the decrease in the overall heat transfer coefficient of the combined frozen layer and dried bottom layer is considered in arrangement II. The accompanying decrease in theoretical drying rates and increase in theoretical drying time has been noted.

The extent of actual bottom drying which had occurred after a drying time of approximately 24 to 26 hours was determined by inspecting bisected samples. Tracings of the bisected samples are shown in Figure 45 for vacuum chamber pressures of 0.5, 2 and 3 torr.

This experimental evidence of restricted bottom drying plus the similarity of the actual and theoretical drying rate and drying time curves, all tend to support the validity of the theoretical analyses





used to approximate the effects of restricted bottom drying.

The drying rate is defined as the mass rate of sublimation per unit time per unit area of the drying surface of the sample. This can be expressed by the following equation

$$\dot{w} = \frac{1}{A_s} \frac{dm_s}{dt} = \frac{1}{A_s} \lim_{\substack{s \\ \Delta t \to 0}} \frac{\Delta m_s}{\Delta t}$$
(119)

Thus, for any value of drying time this can be written

$$\dot{\mathbf{w}} = \dot{\mathbf{w}}(\mathbf{t}) = \frac{1}{A_s} \frac{\Delta m_s}{\Delta \mathbf{t}} = \frac{1}{A_s} \frac{\Delta m_g}{\Delta \mathbf{t}} |_{\mathbf{t}}$$
(120)

where

 A_s = area of drying surface of the sample Δm_s = mass of ice sublimated in short time interval Δm_g = change in gross mass in short time interval Δt = short interval of time t = drying time

Equation (120) was used to calculate the actual drying rates since all of the necessary data was easily obtained during an actual test.

The dimensionless dried layer thickness is defined as the equivalent dried layer thickness divided by the sample thickness. Thus, the following equation can be used to calculate the dimensionless dried layer thickness at any drying time

$$z(t) = \frac{\delta}{\ell} \bigg|_{t} = \frac{\rho A_{s} P \delta}{\rho A_{s} P \ell} \bigg|_{t} = \frac{m_{s}}{m_{i}} \bigg|_{t} = \frac{m_{g|0} - m_{g|t}}{m_{i}}$$
(121)

where

p = density of frozen meat juices
m_s = cumulative mass of sublimated ice
m_i = initial mass of ice
m_{g|o} = gross mass of sample at zero time
m_{g|t} = gross mass of sample at drying time t

From equation (121) it is apparent that the dimensionless dried layer thickness z is equivalent to the cumulative mass of sublimated ice expressed as the fraction of the initial mass of ice in the sample.

The actual drying rate for any particular values of drying time can be calculated by using equation (120), then the dimensionless dried layer thickness corresponding to this value of drying time may be determined by using equation (121). This permits the actual drying rate to be expressed either as a function of drying time or as a function of dimensionless dried layer thickness.

The actual drying time curves, which show the dimensionless dried layer thickness versus the drying time, may be determined by using equation (121) directly.

CHAPTER VĨ

CONCLUSIONS

The principal conclusions drawn from both the theoretical and experimental analyses of this investigation are:

1. The flow of sublimated vapor through the capillary channels of the porous dried layer is in the transition flow regime for most usual freeze-drying operating conditions, i.e. $0.5 \le p_0 \le 3$ torr.

2. The equation for flow of rarefied vapor through the capillary channels in the porous dried layer includes the temperature gradient as well as the pressure gradient because both contribute to the flow rate.

3. For fixed boundary conditions, the exact solution permits interface temperature to be determined as a function of the interface position which will simultaneously satisfy both the energy and continuity equations.

4. The exact solution interface temperature increases only slightly as the interface moves through the sample during freeze-drying and also increases only slightly if the bottom surface temperature of the sample is increased.

5. The exact solution indicates that the interface temperature is primarily determined by the vacuum chamber pressure, decreasing as the vacuum chamber pressure is reduced.

6. The approximate solution which is based upon a constant interface temperature and linear temperature gradients in the dried and frozen layers gives drying rates very close to the exact solution drying rates and are much less time-consuming to calculate than the exact solution drying rate equations.

7. The approximate solutions for arrangements II and III, which are based upon drying from the top surface only but with restricted bottom drying and zero bottom drying respectively, show a substantial decrease in drying time as the vacuum chamber pressure is reduced.

8. The approximate solution for arrangement I, which is based upon simultaneous drying from two surfaces, shows only a modest decrease in drying time as the vacuum chamber pressure is reduced.

9. The approximate solutions for arrangements II and III show a substantial decrease in drying time as the bottom surface temperature is increased, while holding all other boundary conditions constant.

10. Experimental evidence indicates that partial bottom drying occurred during all tests at various constant vacuum chamber pressure; therefore, arrangement III which assumes zero bottom drying is primarily of theoretical interest only.

11. The approximate solutions indicate that for a vacuum chamber pressure of 0.5 the drying time is faster for arrangement II than for arrangement I; while for vacuum chamber pressures of 2 and 3 torr the drying time is faster for arrangement I than for arrangement II.

12. Measured dimensionless pressures fall within the range of calculated exact solution dimensionless pressure curves.

13. Measured dimensionless temperatures deviate somewhat from the calculated exact solution dimensionless temperature curves, but exhibit a similar dropping characteristic as the dimensionless distance in the

dried layer increases.

14. Measured drying time curves for vacuum chamber pressures of 0.5 and 2 torr are close to the calculated approximate solution curves for arrangement II; while for a vacuum chamber pressure of 3 torr the measured drying time curve is bounded by calculated approximate solution curves for arrangements I and III.

APPENDIX A

DERIVATION OF OVERALL HEAT TRANSFER COEFFICIENT U EQUATION

. . To take the phenomenon of restricted bottom drying into account, the heat flux through the combined frozen layer and bottom dried layer is calculated by using an overall heat transfer coefficient for the combined layers, designated by U. It is assumed that the bottom layer remains frozen in the early stages of drying; however, when the interface reaches some critical value, designated z_c , it is assumed that the bottom layer gradually begins to dry and continuously increases in thickness for the remainder of the freeze-drying process. This will cause the overall heat transfer coefficient to decrease until the sample is completely dried. It is obvious that as the thickness of the frozen layer approaches zero, i.e. as $z \rightarrow 1$, the overall heat-transfer coefficient U must approach the thermal conductivity of the porour dried layer. Thus, the overall heat transfer coefficient U must satisfy the following conditions

1. For the upper limit of U

 $U = k_{F} \quad \text{for } 0 \le z \le z_{C}$ 2. For the lower limit of U

 $U \rightarrow k_{D}$ as $z \rightarrow 1$

Assuming a linear variation of the overall heat-transfer coefficient U with the dimensionless dried-layer thickness, which will also

satisfy conditions (1) and (2), it is easy to derive an equation for the overall heat transfer coefficient in terms of the dimensionless dried layer thickness z. A diagram of U vs. z is shown in Figure 46.







. By geometry

 $\frac{k_{F} - U}{k_{F} - k_{D}} = \frac{z - z_{c}}{1 - z_{c}}$ $U = k_{F} - (z - z_{c}) \frac{(k_{F} - k_{D})}{1 - z_{c}}$

thus

$$U = a_1 + b_1 z \tag{59}$$

where

$$a_{1} = k_{F} + \frac{z_{c}(k_{F} - k_{D})}{1 - z_{c}}$$
(60)

$$b_1 = -\frac{(k_F - k_D)}{1 - z_C}$$
(61)

APPENDIX B

DERIVATION OF AN EQUATION RELATING THE BOTTOM AND TOP DRIED LAYER THICKNESSES

It is interesting to determine the relationship between the bottom and top dimensionless dried layers which occurs if the overall heat transfer coefficient U decreases linearly as shown in Figure 46 and as given by equation (59).

Figure 48 shows a typical diagram of arrangement II in which restricted bottom drying is considered. In this diagram $z_1 = \delta_1/\ell$ designates the top dimensionless dried layer thickness, while $z_2 = \delta_2/\ell$ designates the bottom dimensionless dried layer thickness.

The heat flux conducted through the combined frozen layer and bottom dried layer, with boundary temperatures T_{δ} and T_{B} , is given by the following expression

$$\frac{(T_{B} - T_{\delta})}{\frac{\ell - \delta_{1}}{U}} = \frac{(T_{B} - T_{\delta})}{\frac{\ell - (\delta_{1} + \delta_{2})}{k_{F}} + \frac{\delta_{2}}{k_{D}}}$$
(122)

$$\frac{U}{(1-z_1)} = \frac{1}{\frac{1-(z_1+z_2)}{k_F} + \frac{z_2}{k_D}}$$

Rearranging,
$$U = \frac{k_{\rm F}}{1 + \frac{(k_{\rm F} - k_{\rm D})}{k_{\rm D}} \frac{z_2}{1 - z_1}}$$
(123)

Assuming a linear variation of the overall heat transfer coefficient U with z_1 , equation (59) can be substituted into equation (123) to give

$$a_1 + b_1 z_1 = \frac{k_F}{1 + a_4 \frac{z_2}{1 - z_1}}$$
 (124)

where

$$a_4 = \frac{k_F - k_D}{k_D}$$

Rearranging equation (124),

$$z_{2} = \left[\frac{k_{F}}{a_{1} + b_{1}z_{1}} - 1\right] \frac{(1 - z_{1})}{a_{4}}$$
(125)

A plot of equation (125) is shown in Figure 47. It may be observed that z_2 varies approximately linearly with z_1 over most of the curve and reaches a maximum value of approximately 0.025. Thus, it can be seen that only a relatively thin bottom dried layer substantially decreases the overall heat transfer coefficient and thus decreases the bottom heat flux and the drying rate.

It seems appropriate to explain why the bottom interface temperature is greater than the top interface temperature in arrangement II, but equal in arrangement I with simultaneous drying from two surfaces.





Referring to Figure 48 for arrangement II, vapor sublimated from the top interface at points such as (1) have direct and relatively short paths through the capillary channels in the porous dried layer in order to escape to the free surface and be removed from the system. In addition, the total vapor sublimated from the top interface has a cross-sectional area for flow of $P\pi r_1^2$. Since the porosity of the dried layer is relatively high, the pressure drop between the top interface and free surface is comparatively small. This causes the top interface pressure to be only slightly higher than the vacuum chamber pressure. Thus, at the top interface the saturation temperature is only slightly greater than the saturation temperature corresponding to the vacuum chamber pressure.

In contrast to vapor sublimated from the top interface, vapor sublimated from the bottom interface at points such as (2) must travel a rather tortuous path across the fibers, around through the restricted channel at the edges between the frozen layer and vapor barrier, and finally in the negative x-direction to the free surface. The crosssectional area for flow between the frozen layer and the vapor barrier is given by the following equation

 $\frac{\text{Cross-sectional}}{\text{area at edges}} = P \pi (r_1^2 - r_2^2)$ (126)

Since $r_2 \approx r_1$, the cross-sectional area at the edges is very small, thus causing a throttling effect on the vapor sublimated from the bottom interface. The combined effects of the long tortuous path and throttling at the edges cause the bottom interface pressure to tend to be greater



Figure 48.

Schematic Diagram of Arrangement II with Restricted Bottom Drying.

than the top interface pressure, thereby causing the bottom interface saturation temperature to be greater than the top interface temperature. The following expressions illustrate the relationships between the pressures and temperatures existing at the free surface, top interface, and bottom interface

 $p_{\delta 2} \ge p_{\delta 1} \approx p_0$ $T_{\delta 2} \ge T_{\delta 1} \approx T_{sat}.$

APPENDIX C

EQUATIONS AND REFERENCES FOR TRANSPORT PROPERTY DATA

The following equations and references were used to obtain the necessary transport data for both the exact and approximate solution theoretical calculations:

		Reference <u>Number</u>
1.	Constant pressure specific heat of water vapor, c, Btu/lb_ °R.	(14)
2.	Mean diameter of capillary channels in freeze-dried beef, \overline{d} , ft.	(4)
3.	Thermal conductivity of vapor-filled freeze-dried beef, k _D , Btu/ft.hr.ºF	(4)

$$k_{D} = k_{s} + \frac{k_{vo}}{1 + \frac{C}{p}}$$

where

C = constant

- p = mean pressure of vapor.
- Thermal conductivity of frozen beef. Reference (5) presents a plot of thermal conductivity of beef versus temperature. k_r, Btu/ft.hr.°F.

(5)

5.	Latent heat of sublimation of ice, L, Btu/lb _m .	(14)
6.	Latent heat of sublimation plus superheat of water vapor, L', Btu/lb m	(14)
	$L^{\prime} = L + c(T_{o} - T_{\delta})$	
7.	Absolute viscosity of water vapor, μ, ⁽⁾ lb _f /sec / ft²	(14)
8.	Density of ice, ρ, lb _m /ft. ³	(14)
9.	Saturation pressure-temperature relationship for frozen pure water.	(14)
10.	Saturation pressure-temperature relationship for frozen beef.	(7)
11.	Porosity of freeze-dried beef (void fraction).	(4)
12.	Gas constant for water vapor, R, ft. lb _f /lb _m °R.	(14)
13.	Mean free path of water vapor molecules, $\lambda, \ \mbox{ft.}$	(13)

 $\lambda = \lambda_{s} \frac{p_{s}}{p} \left[\frac{T}{T_{s}} \right]^{n+1}$

where

 λ_{s} = mean-free path of water vapor molecules at standard pressure and temperature

p_s = 'standard pressure

 $T_{c} = standard temperature$

n. = constant

APPENDIX D

TRANSPORT PROPERTY DATA USED IN EXACT AND

APPROXIMATE SOLUTION CALCULATIONS

Table 3.Transport Property Data Used in Exact andApproximate Solution Calculations

and the standards

	Property	p ₀ =0.5 torr	p ₀ =2 torr	p _o =3 torr
1.	Constant pressure specific heat, Btu/lb_mR	0.445	0.445	0.445
2.	Mean diameter of capillary channels, cm	100×10^{-4}	100×10^{-4}	100×10^{-4}
3.	Thermal conductivity of freeze-dried beef, Btu/ft.hr.°F	0.0245	0.0279	0.0288
4.	Thermal conductivity of frozen beef, Btu/ft.hr.°F	0.62	0.58	0.52
5.	Latent heat of sublima- tion of ice, Btu/lb _m	1220	1220	1220
6.	Latent heat plus super- heat of water vapor, Btu/lb _m	1271	1264	1259
7.	Absolute viscosity of water vapor,lb _f sec/ft ²	2.16×10^{-7}	2.16 $\times 10^{-7}$	2.16×10^{-7}
8.	Density of ice, lb_m/ft^3	57.5	57.5	57.5
9.	Saturation pressure- temperature relation- ship of pure ice	see	e reference (14)	

	Property	p ₀ =0.5 torr	p ₀ =2 torr	p _o =3torr
10.	Saturation pressure- temperature relation- ship of frozen beef	see	reference (7)	
11.	Porosity of freeze- dried beef (void fraction)	0.70	0.70	0.70
12.	Gas constant for water vapor, ft.lb _f / lb _m °R	85.8	85.8	85.8
13.	Mean free path of water vapor molecules, cm	63.0 x 10 ⁻⁴	16.0×10^{-4}	
14.	Interface temperatures used in exact solution	see	Table 2	
15.	Interface temperatures used in approximate solution, ^o R (using sat. presstemp. data for frozen beef.)	459.78	478,79	487.14

Table 3 (Continued)

APPENDIX E

METHOD USED TO EVALUATE THE EMPIRICAL CONSTANT c APPEARING IN EQUATION (74)

Values of the empirical constant c_0 appearing in equation (74) may be evaluated in the manner now to be explained. Rearranging equation (74),

$$c_{o} = \frac{T_{b}}{T \text{ sat.}}$$
(127)

where T_{δ} is the exact solution temperature corresponding to the average value of the heat flux ratio \overline{f} . Values of T_{δ} may be obtained from Tables 2 and 3 or calculated using equation (40).

The average value of the heat flux ratio is obtained by integrating the variable heat flux ratio over the entire drying range z = 0 to z = 1.0. Thus, the average heat flux ratio is given by the following equation

$$\overline{f} = \int_{0}^{1} f \, dz \tag{128}$$

The variable heat flux ratio f is a function only of z and is given by the following equation which is obtained for the approximate solution by substituting equation (75) and (76) into equation (110)

$$f = \frac{1}{1 + \frac{z}{1 - z} \frac{k_{F}(T_{B} - T_{\delta})}{k_{D}(T_{0} - T_{\delta})}}$$
(129)

Rearranging,

$$f = \frac{1 - z}{1 + c_5 z}$$
(130)

where

$$c_{5} = \frac{k_{F}(\bar{T}_{B} - \bar{T}_{\delta})}{k_{D}(\bar{T}_{0} - \bar{T}_{\delta})} - 1$$
(131)

Substituting equation (130) for the variable heat flux ratio into equation (128) and integrating gives the following equation for the average heat flux ratio

 $\overline{f} = \frac{c_5 + 1}{c_5^2} \ln (1 + c_5) - \frac{1}{c_5}$ (132)

Sample Calculation

The following sample calculation for c_0 is made using the saturation pressure-temperature relationship for frozen beef juices (7) and is based upon drying arrangement III.

Given: _____

$$p_o = 2 \text{ torr}$$

 $T_{sat.} = 477.8^{\circ}R$ reference (7)
 $T_o = 575^{\circ}R$
 $T_B = 492.5^{\circ}R$
 $k_F = 0.58$ reference (5)
 $k_D = 0.028$ reference (4)
 $c_5 = 1.95$ calculated using end

calculated using equation (131)

Trial Calculation No. 1

f = 0.3 assumed

 $T_{b} = 478.79$ from Table 2 corresponding to $\overline{f} = 0.3$ Use equation (132) to calculate \overline{f}

$$\overline{f} = \frac{1.95 + 1}{(1.95)^2} \ln (1 + 1.95) - \frac{1}{1.95} = 0.286$$

Since the assumed and calculated values of \overline{f} are close, additional trial calculations are not necessary. Thus, the constant c_0 is

calculated by using equation (127)

$$c_{o} = \frac{T_{\delta}}{T_{sat.}} = \frac{478.79}{477.8} = 1.001$$

APPENDIX F

TABULAR DATA FOR EXACT AND APPROXIMATE SOLUTION CALCULATIONS

Table 4. Approximate Solution Interface Temperatures and Constant c_0 Used in Equation (74)

	p _o =0.5 torr	p _o = 2 torr	p _o = 3 torr
Interface temp. T _o ,°R	453.5	476.0	483.7
*Saturation temp., oR	448.0	474.4	482.9
Constant, c _o	4 1.01	1.008	1.001
(b)	For Frozen Meat J	uices, $\overline{f} = 0.3$	
Interface temp. T _o ,°R	459.8	478.8	487.1
*Saturation temp., ^o R	455.0	477.8	486.1
Constant, c _o	1.01	1.001	1.001

(a) For Frozen Pure Water, $\overline{f} = 0.3$

* Saturation temp. corresponding to vacuum chamber pressure p_0 .

x*	f = 0.1 *	f = 0.2 *	f = 0.3 p*	f = 0.4 p [*]	f = 0.5 p*	f = 0.6	f = 0.7 p*	f = 0.8 p*	f = 0.9 p_	f = 1.0
0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
.1	1.167	1.090	1.058	1.040	1.029	1.021	1.016	1.011	1.008	1.005
.2	1.306	1.168	1.109	1.076	1.055	1.040	1.029	1.021	1.014	1.008
.3	1.424	1.236	1.155	1.108	1.078	1.056	1.041	1.028	1.019	1.011
.4	1.526	1.297	1.195	1.136	1.098	1.071	1.050	1.035	1.022	1.012
.5	1.617	1.351	1.231	1.161	1.115	1.083	1.058	1.039	1.024	1.011
•6	1.698	1.399	1.263	1.183	1.130	1.093	1.064	1.042	1.024	1.010
.7	1.770	1.442	1.291	1.202	1.143	1.101	1.069	1.044	1.023	1.007
.8	1.835	1.480	1.316	1.219	1.154	1.107	1.071	1.043	1.021	1.002
.9	1.894	1.514	1.338	1.233	1.162	1.111	1.072	1.042	1.017	0.997
.0	1.948	1.545	1.356	1.244	1.168	1.113	1.071	1.038	1.012	0.990
х*	T [*]	т*	т*	Τ*	т*	T*	T [*]	T [*]	Ţ *	т*
Ö	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
.1	0.978	0.979	0.979	0.979	0.979	0.979	0.979	0.979	0.979	0.979
.2	، 957	.958	.958	.958	•958	.95 8	. 958 /	•958	.958	. 958
.3	° • 937	.937	.937	.937	.937	•937	.937	.937	•93 7	. 936
.4	.917	.917	•917	•916	.916	•916	•916	.916	.91 6	.91 6
•5	.898	.897	•89 6	.896	• 896	<u>895 ،</u>	.895	. 895	. 895	895 ،
.6	.880	.878	•877	.876	.875	.875	.875	.874	.874	.874
.7	. 862	.859	، 857	.85 6	.855	.855	.854	.834	. 854	.853
•8	.845	.840	.838	•836	.835	. 834	.834	.833	۰833	•833
.9	.828	.822′	.818	.816	.815	.814	.814	.813	.812	.812
.0	.812	.804	.800	.797	.796	₀794	。7 94	۰793 ،	.792	.792

Table 5. Exact Solution - Dimensionless Pressure and Dimensionless Temperature Ratios*

					<u> </u>					
x*	f = 0.1 #	f = 0.2 * p	f = 0.3 * P	f = 0.4 *	f = 0.5 * p	f = 0.6 *	f = 0.7 *	f = 0.8 *	f = 0.9 * p	f = 1.0 * P
0	- <u>1</u> .0	-1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.001	1.0
, ĩ	1.030	1,015	1.009	1.006	1.005	1.003	1.002	1.002	1,002	1,001
.2	1.057	1.029	1.018	1.012	1.009	1,006	1.005	1.003	1.003	1.002
.3	1.082	1.042	1.026	1.018	1.013	1.009	1.007	1.005	1.004	1,002
.4	1.106	1.054	1.034	1.023	1.016	1.012	1.008	1.006	1.005	1.002
.5	1.128	1.066	1.041	1.028	1.020	1.014	1.010	1.007	1.005	1.003
.6	1.148	1.076	1.048	1.032	1.023	1.016	1.011	1.008	1,005	1.003
.7-	1.167	1.086	1.054	1.036	1.025	1.018	1.012	1.008	1.005	1.003
.8	1.185	1.095	1.060	1,040	1.028	1.019	1.013	1.009	1.005	1.002
.9	1.201	1.103	1.064	1.043	1.030	1.021	1.014	1,009	1.005	1.002
1.0	1.216	1.111	1.069	1.046	1.031	1.022	1.014	1.009	1.005	1.001
x*	т* :	T [*]	т*	т*	T*	_ T [*]	T [*]	T [*]	T*	T [*]
0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
.1	0.981	0.982	0.982	0.983	0.983	0.983	0.983	0.983	0.983	0.983
.2	.964	.965	.950	.965	.965	.965	.9 66	.9 66	• 9 66	°9 66
.3	•946	.947	.948	.948	.948	.948	•94 8	•948	.949	۰948
.4	.929	.930	.931	.931	.931	.931	.931	. 931	. 931	.932
.5	.913	.914	.914	.914	.914	.914	.914	.914	.914	。914
.6	.897	.897	.897	.897	898	. 898	.898	. 898	.898	· • 898
.7	.881	.881	.881	. 881	.881	.881	.881	.881	• 881	.881
•8	.866	₀ 865	. 865	• 8 64	.864	.864	. 864	₀ 864	• 864	. 864
.9	.851	.849	•848	.848	. 848	848	.848	.847	.847	.847
1.0	•837	.834	•833	.832	.832	.831	.831	.831	.831	.831

Table 6. Exact Solution - Dimensionless Pressure and Dimensionless Temperature Ratios^{*} $(p_{r} = 2 \text{ torr}; T_{r} = 575^{\circ}R)$

*Based on: 1. Equations (41) and (42), 2. Vapor press.-temp. data for frozen beef (7).

* Exact Solution - Dimensionless Pressure and Dimensionless Temperature Ratios Table 7.

(p₀ = 3 torr; T₀ = 575°R)

1.0 0.984 0.984 0.953 0.953 0.953 0.922 0.922 0.861 0.861 0.861 0.861 0.846 f = 1.0 Vapor press.-temp. data for frozen beef (7). م f = 0.91.0 969 953 953 953 953 953 953 952 952 952 861 861 861 846 f ≡ 0.8 p ***** e 1.0 984 953 953 953 922 907 892 892 892 892 876 *⊢ f = 0.71.0 969 953 953 953 953 952 952 861 861 861 846 .001 .001 .005 .006 .006 .007 .007 .007 .008 * <u>a</u>. *⊢ f = 0,6-p * 4 *⊢ ณํ ք=0.5 ₽*9 1.0 0.984 0.984 .953 .953 .953 .959 .861 .861 .861 *⊢ 1. Equations (41) and (42); f = 0.4 p * 1.0 0.984 0.984 953 953 953 953 952 952 962 862 862 862 1.0 1.003 1.006 1.010 1.012 1.015 1.015 1.017 1.019 1.021 1.021 1.023 1.025 *⊢ ຕຸ 0 * d 1.0 9584 9582 952 952 952 952 952 952 877 877 862 847 *⊷ f = 0.2 p * 1.0 0.984 0.984 952 .937 .921 .921 .921 .877 .877 .848 L.008 L.008 L.015 L.015 L.035 L.035 L.047 L.047 L.056 L.055 L.056 L.056 *⊢ Based on: f = 0**.**1 P * 1.0 967 951 951 955 920 920 920 920 877 877 851 r.0 1.031 1.031 1.045 1.058 1.058 1.032 1.033 1.103 1.113 *н **ωο**Ο 1004001

Heat Flux Ratio	$p_0 = 0.5$		p _o = 2 .torr		p _o = 3 torr		
f	z	Ŵ	Z	ŵ	Z	. ŵ	
1.0 .9 .8 .7	0 .028 .065 .114	∞ • 893 • 430 • 278	0 .033 .072 .117	∞ .702 .365 .252	0 .057 .121 .190	∞ .346 .185 .133	
.6 .5 .4 .3 .2 .1	.183 .283 .446 .735 .956 .980	.200 .153 .119 .094 .103 .178	.173 .247 .373 .571 .848 .975	.199 .166 .1361 .116 .114 .181	.294 .440 .639 .868 .960 .983	.100 .080 .068 .066 .087 .156	
· ·	1.0	Boundary	y Temperatu	res, ^o R		-	
т _о	5	75	57	5	5	75	
т _в	. 4	70	49	2.5	. 4	95 10	

Exact Solution-Arrangement II, Table 8. Dimensionless Dried Layer Thickness and Drying Rates* \$

* Calculations are based on:

.

- Using Equations (51), (58), and (65)
 Interface Temperatures listed in Table 2.
- Saturation pressure-temperature data for frozen beef, 3. see reference (7).

	$p_0 = 0.5 \text{ torr}$		p _o =	2 torr	p _o =	p _o = 3 torr		
2	ŵ	t	Ŵ	t	ŵ	t		
ò		0	~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~		<u> </u>			
.05	- 856	.06	. 817	.06	. 705	.07		
10	428	.24	408	.26	352	.27		
15	.286	.55	. 272	.58	.234	.61		
20	.214	.97	.204	1.02	.176	1.08		
25	.171	1.52	.163	1.60	.141	1.69		
, 30	.142	2.18	.136	2.30	.117	2.43		
. 35	.122	2.98	.117	3.14	.101	3.31		
.40	.107	3.88	.102	4.10	.088	4.32		
,45	.095	4.91	.091	5.18	۰0 7 8	5.46		
,50	.08 6	6.06	• • 082	6.40	.071	6.75		
, 55	.078	7.34	.074	7.75	.064	8.16		
60	.071	8.74	068 ،	9.21	۰059	9.71		
, 65	.066	10.23	.063	10.80	۰054	11.40		
. 70	.061	11.90	₀058	12.57	.050	13.21		
, 75	.057	13.63	,05 5	14.40	.047	15.20		
, 80	.053	15.53	.051	16.40	.044	17.30		
.85	.050	17.50	.048	18.48	.042	19.50		
.90	.048	19.70	.045	20.80	.039	21,90		
• 7 5	.045	21.85	.043	23.10	.037	24.35		
.00	.043	24.20	،∪41	23.6V	•03 0	27.00		
		Bound	lary Temper	atures, °R				
r _o 1	57	5 :	57	5	57	5		
Γ _δ	45	9.78	47	8.79	. 49	495		
l	1	1/4"	1	1/4"	1 3	/8"		

Table 9. Approximate Solution-Arrangement I Drying Rates and Drying Time.*

* Calculations are based on:
 1. Using Equations (103) and (107).

Using saturation pressure-temperature data for frozen beef, See reference (7). 2.

	p _o = 0.5 torr			р _о	= 2 to	= 2 torr p _o = 3 torr			orr
z	f	ŵ	t	f	ŵ	t	f	Ŵ	t
0	1.0	~ ·	0	1.0	8	0	1.0	8	0
.05	894	.478	22	. 865	.472	.23	。921	.381	. 31
.10	.800	°2 67	. 82	₀752	.271	• 85	. 848	. 20 7	1 .18
.15	.716	.199	1.73	.65 6	. 207	1.74	.778	.150	2.51
• 20	.641	.167	.2.88	.574	.17 81	2.84	.712	.125	4.21
•25	.587	.145	4.23	.518	.158	4.10	.702	.108	6.21
.30	₅542	.131	5.75	.471	.144	5.49	.650	.097	8,46
.3 5	.503	.121	7.41	.432	.135	6.99	.559	.090	10.94
.40	.468	.114	9.19	.400	.128	8.59	519ء	.084	13.59
.45	•438	.108	11.07	.369	.123	10.26	.4 82	.081	16.38
.50	.411	.104	13.05	.343	.119	11.99	.447	.078	19.28
.55	• 386	.100	<u>15.10</u>	.320	. 116	13.78	.414	۰07 7	22.24
• 60	.364	.098	17.21	• 300	.113	15.60	.381	.077	25.24
، 65	. 344	•096	19.38	.280	•112	17.47	. 349	.077	28.24
。70	• 324	.094	21.58	.262	.111	19.34	• • 316	.079	31.18
.75	.306	.093	23.82	.245	.111	21.23	.281	•083	34.02
.80	.286	.093	26.07	.227	.112	23.11	.244	.090	36.69
.85	•266	.095	28.30	.207	.116	24.94	. 201	.102	39.10
.90	.239	.099	30.47	.182	.124	26.70	.151	.129	41.13
• 95	.194	.116	32.45	.140	.153	28.25	•088	.2 10	42.60
1.00	0	8	33.71	0	æ	29.15	0	∞	43.19
 +			Bound	lary Temp	<u>eratur</u>	es, °R			
т _о		5 7 5			575			575	
T _a		459.7	8		478.7	9		487.1	4

Table 10. Approximate Solution-Arrangement II Drying Rates and Drying Time*

492.5 ТВ 470 495 1 1/4" 1 3/8" ł, 1 1/4"

^{*}Calculations are based on:

 Using Equations (78), (85), (93), and (98).
 Using saturation pressure-temperature data for frozen beef, see reference (7).

Table 11. Approximate Solution-Arrangement III Drying Rates and Drying Time*

saturation pressure-temperature data for frozen 6.22 13.35 2.51 8.45 10.86 1.17 4°21 .es 0 ىپ torr 487.14 3/8" .150 .150 .123 .108 .099 .091 .095 .108 1120 1137 1137 1120 1120 1120 1120 1120 1120 .091 575 381 495 •3 ന 8 ч u പ് 848 778 712 650 591 591 535 481 431 382 382 336 292 292 250 292 250 292 098 0098 0098 0031 .922 1.8 Ч÷ ŝ .85 1.74 4.08 9.52 14.35 15.32 16.16 16.87 17.83 17.84 .33 2.84 6°78 8.16 10.84 12,10 13.27 18.09 18.18 Equations (78), and (93). 5.41 0 د Boundary Temperatures. torr 1 1/4" 478.79 492.5 . 233 .162 .155 .152 .152 . 270 . 329 . 428 .178 .156 .162 .172 .186 204 472 271 °207 .231 6 575 ∘≩ Ś 8 Ш see Reference are based on: പ് 752 656 574 153 502 440 385 336 **291** 252 216 183 126 776 174 **, 865** 101 561 .361 1.0 4 $\mathbb{T} \in \mathbb{T}$ 13.25 14.63 17.07 17.07 18.98 19.62 20.12 -1 4.20 7,16 10,26 20.42 20.52 08 5.64 11.79 8°.71 د torr Using F Using (beef, 459.78 Calculations 1_1/4" .267 .345 478 199 ,166 **,149** .140 .135 .133 .135 .146 .156 .170 .190 .220 982 267 470 0.5 575 ∘≩ 8 8 цч Ч II. 229 193 160 129 129 073 571 571 453 453 352 352 352 267 8 716 640 , 894 023 പ് 1.8 44 0 £888680 5°. • 50 55, <u>.</u> . 70 0 o æ N ÷Ô نە

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APPENDIX G

TABULAR DATA FOR ACTUAL TESTS

Table 12. Pressure Measurements and Weight Loss for 0.5 torr Chamber Pressure Test, torr*

Time, Hours	x = 0"	x = 1/4"	x = 5/12"	x = 7/12"	x = 3/4"	x = 11/12"	Weight los <mark>s</mark> , grams	Z
0							0	
ĩ	.48						20.0	. 095
2	48						32.5	154
3	.48	.716					42.3	200
4	.48	.815		÷.,			51.0	.242
5	.47	. 600					59.1	.280
6	.47	.570	.715				66.3	.314
7	.50	.581	.714				72.8	.345
8	.51	.584	.628				79.5	.377
9	.48	.552	.608				85.3	.403
10	.52	.578	• 620	.903			91.4	.432
11	.49	.550	.595	.904			96.7	.458
12	.48	.540	.585	• 884			1 0 2.2	•485
13	.48	.546	.58 6	• 826			107.1	.507
14	.48	.540	.580	.780			111.8	.525
15	.45	510	.550	.729	.698	· .	116.7	.5 53
16	.48	• 538	.564	.730	.720		121.4	.575
17	. 45	.508	•534	•684	•688		125.6	.595
18	• .48	.538	.560	•696	.706		130.2	.617
19	.48	.536	.556	. 681	.703		134.1	.635
20	.48	.542	•562	•679	.716		138.2	•655
21	.48	.542	.562	.666	.720	.751	142.4	.675
22	.47	.538	•552	.650	.715	.613	145.8	.692
23	.49	.558	.566	.658	.739	.761	149.5	.709
24	.47	.541	.552	.635	.719	.746	153.3	.728

x = pressure probe position, inches

Table 13. Temperature Measurements for 0.5 Chamber Pressure Test, ^{oF*}

torr

692 709 728 - 0 0 N . ` 1/4" 16.0 16.5 18.5 19.0 22.5 Ч H × = 11/12" 1 1 1 A A temperature probe position, inches × 3/4" 20.0 20.0 20.0 20.0 20.0 20.0 20.0 21.0 26.0 30.5 32.0 0.6 II × = 7/12" **0**000 0000 6.0 7.0 7.0 7.0 6.0 6.0 9.0 15.5 24.5 32.5 32.5 42.0 42.0 47.5 7.0 6,0 × = 1/4" × 116.0 114.5 116.0 116.0 116.0 116.0 115.0 117.0 117.0 116.0 110.0 113.5 116.0 116.0 116.0 116.0 116.0 116.0 116.0 116.0 12.5 ō - 11 п × × × _ Time, Hours 22222

150

ż

Pressure Measurements and Weight Loss for 2 torr Chamber Pressure Test, torr torr Chamber Pressure Test, Table 14.

.631 .655 .680 .680 .736 .736 .735 .185 .229 .229 .311 .311 .3383 .348 .3383 .348 .3414 .511 .511 .5530 .5530 840 840 129 80. N O Weight loss, grams 36.1 36.1 52.9 60.9 68.0 68.0 68.0 68.0 92.7 92.7 92.7 112.0 110.0 112.0 25°2 = 3/4" 2.768 2.542 2.209 2.156 2.156 10 -× $x = 7/12^{n}$ 2.848 2.848 2.104 2.104 2.080 2.080 2.095 2.095 2.016 2.016 2.016 2.016 2.016 777 d. x = 5/12" 2.170 2.105 2.106 2.106 2.042 2.076 2.076 2.058 2.058 2.058 2.058 2.058 2.058 2.058 2.128 = 1/**4**" 2.070 2.019 2.078 2.060 2.032 2.032 2.018 2.058 2.050 2.050 2.097 2.144 2.094 2.089 2.049 2.074 2**.04**6 2.064 2.064 2.002 2.064 . 🗙 1.99 1.99 2.02 2.02 2.02 2.06 0 1.20 2.05 2.02 **∔**× 11 × Time, Hours 2222209874554321098765432F Ο

2.937 torr. Λ <u>ρ</u>, i.e., scale, gage off pressure * Differential

pressure probe position, inches

li

torr 3 .∍ for oF Temperature Measurements Chamber Pressure Test, ^oF 5 Table

3

probe position,

inches

temperature li.

×

*

	·					·	
Time, Hours	x = 0	x = 1/4"	x = 5/12"	x = 7/12"	x = 3/4"	Weight loss, grams	z
0 -						0	0
ĩ	3.05					24.7	.108
2	3.05					37.1	.163
3	3.03	3.069		·		47.1	.206
4	3.05	3.082				57.8	.253
5	3.03	3.054				66.3	.291
6	3.03	3.058				74.6	.327
7	2.99	3.112	3.274			82.1	.361
8	3.05	3.066	3.148			89.0	.390
9	3.07	3.079	3.142			99.4	.436
10	3.05	3.066	3.096			105.1	.462
11	3.05	3.059	3.106	3.164	. ·	111.8	.485
12	3.02	3.029	3.075	3.116		118.7	.520
13	3.05	3.056	3.088	3.121		123.8	.543
140	3,05	3.056	3.058	3.121		130.5	.574
15	3.05	3,056	3.060	3.112		135.6	.595
16	3.05	3.054	3.058	3.110	3.285	141.3	.620
17	3.03	3.036	3.034	3.090	3.198	146.3	.645
18.	3.05	3.059	3.074	3.109	-	151.0	.662
19	3.05	3.058	3.072	3.100	3.198	156.7	.687
20	3.03	3.039	3.055	3.086	3.178	161.5	.709
21	3.03	3.040	-	3.060	3.132	165.8	. 728
22	3.01	3.019	3.032	3.060	3.132	169.8	. /45
23	3.05	3.059	3.062	3.096	3.162	174.4	• /66
24	3.07	3.079	3.082	3,116	3,164	178.8	•781

Table 16. Pressure Measurements and Weight Loss for 3 torr Chamber Pressure Test, torr*

* x ≈ pressure probe position, inches

Time, Hours	x = 0	x = 1/4"	x = 5/12"	x = 3/4"	x = 11/12"	x = 1 3/8"	Z
0	28.5	28.0	32.5	28.5	29.5	27.0	0
1	122.0	48.0	30.5	27.5	33.0	26.5	.108
2	114.0	46.5	28.5	26.0	32.5	30.0	.163
3	115.0	54.0	28.0	24.5	31.5	29.0	.206
4	116.5	63.5	30.0	25.5	33.0	28.5	.253
5	115.0	68.0	30.0	25.5	34.0	28.5	.291
6	116.0	72.0	31.0	25.5	34.5	29.5	.327
7	113.5	73.0	30.0	24.0	34.0	30.0	.361
8	116.5	76.0	31.5	24.5	34.5	31.5	.390
<u>9</u>	114.5	76.5	33.5	24.5	35.0	32.0	.436
10	117.0	79.5	38.0	25.0	35.5	33.0	.462
11	115.0	78.5	42.0	24.0	34.5	34.0	.485
12	116.0	81.0	43.0	24.5	35.5	34.0	.520
13	113.0	81.0	46.0	26.0	36.5	34.5	.543
14	115.0	82.0	48.0	25.5	36.0	35.0	.574
15	115.5	83.0	50.5	27.0	36.5	36.5	.595
16	113.5	82.5	52.0	28.5	37.0	37.5	.620
17	116.0	84.5	53.5	31.0	37.5	37.5	.645
18	115.0	. 84.5	55.5	34.5	38.0	39.5	.662
19	113.0	84.0	57.0	39.0	39.0	40.5	.687
20	115.5	87.0	59.5	43.0	40.0	41.0	.709
21	115.0	87.0	61.5	46.0	40.5	42.5	.728
22	113.5	86.5	62.0	48.5	40.5	42.5	.745
23	117.0	90.5	66.0	54.5	43.5	44.0	.766
24	114.5	89.0	66.0	56.5	44.0	45.0	.781

Table 17. Temperature Measurement for 3 torr Chamber Pressure Test, °F*

x = temperature probe position, inches

APPENDIX H

EQUIPMENT AND INSTRUMENT SPECIFICATIONS

Electric Heater

anufacturer Watlow Electric	: Company
ype	plate
apacity 1700 watts	
ower supply 110 volt, 60 cy	cle

Vacuum Pump

Manufacturer	٠	•	•	٠	•	•	•	•	•	٠	•	٠	W. M. Welch Scientific Co.
Model	•	•	•	•	•	•	•	•	•	•	•	•	1397 B
Capacity	•	٠	•	•	•	•	•	•	•	•	•	•	375 liters/min. of free air
Pump RPM .	٠	•	•	•		٠	•	•	•	٠	•	٠	300
Stages	•	•	•	۰	a	•	•	•	•	٠	٠	•	2
HP of Motor	÷	•	•	•	•	•	•	•	•	•	•	•	3/4
Power Supply							•		•	•	•		110 volt, 60 cycle

Weighing Balance

Magnetically dampened pan

Absolute Pressure Gage

Manufacturer......Wallace and Tiernan CompanyModel.........Mange.........Range.........Minor division......Dial Size......Accuracy......0.33% of full scale range

Differential Pressure Gages

Manufacturer .	٠	٠	٠	•	٠	٠	•.	•	•	F.W. Dwyer Manufacturing Co.
Model	•	•	٠	٠	٠	٠	•	•	•	No. 200 1/2
Range	٠	٠	•	٠	•	•	•	٠	•	050" of water differential
Minor division	•	•	•	•	•	•	•	•	•	0.01" of water
Accuracy	٠	•	•	•	•	ø	•	•	o	± 2% of full scale throughout
Model	٠	٠	•	•	•	•	•	a	•	No. 200
Range	٠	•	•	•	•	٠	•	•	•	0-1.0" of water differential
Minor division	•	•	•	۰	•	•	•	•	•	0.2" of water
Accuracy	•.	•	•	•	•	•	•	•	•	± 2% of full scale throughout

APPENDIX I

DEFINITION OF TERMS USED IN EQUATIONS

a =
$$k_{D}(T_{o} - T_{b})$$

a₁ = $k_{F} + z_{c} \frac{(k_{F} - k_{D})}{1 - z_{c}}$
a₂ = $a_{1}(T_{B} - T_{b})$
a₃ = $\frac{a_{1}(T_{B} - T_{b})}{(1 - f)\frac{Lk_{D}}{c} \ln [\frac{c}{fL} (T_{o} - T_{b}) + 1]}$
a₄ = $\frac{k_{F} - k_{D}}{k_{D}}$
b = $k_{F}(T_{B} - T_{b})$
b₁ = $-\frac{(k_{F} - k_{D})}{k_{D}}$
b₂ = $b_{1}(T_{B} - T_{b})$
b₃ = $\frac{a_{1}(T_{B} - T_{b})}{(1 - f)\frac{Lk_{D}}{c} \ln [\frac{c}{fL} (T_{o} - T) + 1]}$
c₁ = b - a
c₂ = $a_{2} - a$
F = $P \rho \ell^{2} c/k_{D}$
K = $-\frac{k_{D}}{c} \ln [\frac{c}{fL} (T_{o} - T_{b}) + 1]$
K₁(z) = $z^{2}/8a$

$$K_{2}(z) = \frac{1}{c_{1}^{3}} \left[bc_{1}z - c_{1}^{2}z^{2} - a(a + c_{1}) \ln \left(1 + \frac{c_{1}}{a}z\right) \right]$$

$$K_{3}(z) = \frac{b_{2} + c_{2}}{2b_{2}^{2}} \ln \frac{x_{3}(z)}{x_{3}(z_{c})} + \frac{2ab_{2} - (c_{2}^{2} + c_{2}b_{2})}{2b_{2}^{2}q^{4}} \ln \frac{y(z)}{y(z_{c})} - \frac{(z - z_{c})}{b_{2}}$$

$$L' = L + c(T_{0} - T_{\delta})$$

$$M = \frac{k_{D}}{c} - \frac{3}{4} Pg_{c}\mu$$

$$N = \frac{P d^{2}(1 + 3K_{N})}{64\mu R}$$

$$q' = (c_{2}^{2} - 4ab_{2})^{1/2}$$

$$S = 1 - \frac{T_{\delta}}{T_{0}} + \frac{fL}{cT_{0}}$$

$$V = -\ln \left[\frac{c}{fL}(T_{0} - T_{\delta}) + 1\right]$$

$$X_{2}(z) = a + c_{1} z$$

$$X_{3}(z) = a + c_{2}z + b_{2}z^{2}$$

$$Y(z) = \frac{2b_{2}z + c_{2} - q'}{2b_{2}z + c_{2} + q'}$$

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