Heat and Mass Transfer in Hygroscopic Capillary Extruded Products

This work presents an analysis of heat and mass transfer of extruded corn meal. Isotherm, thermal conductivity, drying and center temperature data were experimentally obtained for the cylindrically extruded product.

Modeling of the drying process was made by incorporating the mechanistic and irreversible thermodynamics approaches for determining heat and mass transfer in capillary-porous media. The proposed model shows that both liquid and vapor fluxes can be expressed in terms of the same driving-forces, namely, temperature and equilibrium moisture content gradients. A search technique allowed for the evaluation of both liquid and vapor conductivities.

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SCOPE

The analysis of food dehydration is highly complex. The usual approaches to such an analysis have been to assume that moisture moves by any of the mechanisms of diffusion, capillarity or vaporization-condensation. All these and other mechanisms may play important roles under particular drying conditions.

While it is true that simplified transport equations can be used under restricted circumstances, it is also true that they have led to wrong predictions. These misleading results have usually occurred where a broad range of external (air temperature and air velocity, relative humidity, etc.) and internal (moisture content, temperature, etc.) conditions existed. The overall result is that industrial drying processes are more of an art than a science.

The objective of this work is to apply a set of transport equations to analyze drying of a substance as complex as extruded corn meal. A comparison between the nonequilibrium thermodynamics equations and those obtained from a mechanistic reasoning leads to the evaluation of the phenomenological coefficients. DuFour and Soret's coefficients are obtained from Onsager's relationships. Physical properties of the extruded product were obtained experimentally.

Liquid and vapor conductivities are difficult to obtain experimentally for food products and thus, a nonlinear search technique was employed. The validity of this procedure and model were ascertained by predicting drying rates and center temperatures of the cylindrically extruded product under various environmental conditions.

The procedures involved in the present research can be applied to analyze not only drying, but also hydro and thermal stresses in food products, which depend not only on moisture, but also on temperature gradients inside the body.

CONCLUSIONS AND SIGNIFICANCE

Isotherm data for the extruded corn meal was obtained for the temperature range of 27-90°C. A modified Henderson's equation was used to fit the data. Thermal conductivity data are presented for the temperature range of 30-70°C and for product moisture content between 8-36% dry basis.

Liquid and vapor conductivities were obtained through a search technique. Several models were investigated and the proposed ones have a similar shape as those obtained by Philip and DeVries (1957) and Luikov (1964). The search technique was based on fitting the drying curves in the first 5% (300-600 S) of drying time. The rest of the drying curves and all the center temperature data were predicted successfully, with no fitting.

Based on the theory, a moving evaporation front is postulated to exist for all drying conditions investigated (50-125°C).

Vapor and liquid fluxes are basically dependent on the gradient of equilibrium moisture content. Liquid flux is dominant inside the porous medium at high moisture levels while vapor flux is dominant at regions close to the evaporation front.

The form of the liquid and vapor conductivities can be predicted qualitatively from the moisture isotherm data. The maximum value for the vapor conductivity is reached near the region where single-layer adsorption becomes dominated by multilayer adsorption. The value for the liquid conductivity increases with increases in moisture content and then levels off as saturation is reached. The good agreement between experimental and predicted results favors the approach taken in this work.

INTRODUCTION

Extrusion is basically a process in which a material is "plasticized" and forced to flow under conditions of mixing, heating, and shear, through a die. While there is considerable informa-

tion on the theory and application of polymer extrusion, little engineering information is available for food products. Extrusion causes a series of transformations in food materials, the main effects being: gelatinization of starch components, denaturation of proteins, inactivation of growth inhibitors, changes in product structure and expansion of the extrudate (Smith, 1974 and 1975). Many of the food extrusion operations are followed by drying (Mustakas, et al., 1970; Walsh and Funke, 1975). Because of the amount of energy spent in evaporating the excess

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moisture, drying should be an integral part in any optimization design of extrusion processes.

Water may migrate in hygroscopic capillary-porous media, in both vapor and liquid phases, through a series of mechanisms. Vapor migration may take place by molecular diffusion, due to a partial vapor pressure gradient; by molar transport due to a gradient in total pressure, shrinkage and high temperatures, and by effusion (Knudson flow), when the mean free path of the vapor molecules is of the order of magnitude of the diameter of the pores. Liquid transport occurs by molecular diffusion, due to concentration gradients, by capillarity, by molar flow caused by changes in total pressure and gravity and by surface diffusion.

Due to the complexity of such phenomena, early researchers tried to model the heat and mass transfer processes by assuming an isothermal flow of moisture either by liquid diffusion (Lewis, 1921; Sherwood, 1929a-b, 1931) or by capillarity (Haines, 1927, 1930; Ceaglske and Hougen, 1937; Hougen, et al., 1940). Henry (1939) proposed a model assuming that water migrates entirely in the vapor phase in a series of vaporization-condensation processes. Despite the fact that such models are highly subjected to criticism, when applied to a whole range of moisutre contents, such as in drying, they have been used recently (Harmathy, 1969; Henderson, 1974; Peck and Wasan, 1975). The misleading results obtained from these approaches led to the development of more sophisticated models, which basically, include thermal effects and the vaporization-condensation model associated with either the capillary or liquid diffusion model (Philip and De-Vries, 1957; DeVries, 1958; Krischer, 1963; Berger and Pei, 1973; Baladi, et al., 1978).

Different from the cited mechanistic approaches, other researchers proposed models based on the concepts of irreversible thermodynamics (Cary and Taylor, 1962; Taylor, 1965; Rocques and Cornish, 1977; Valchar, 1966 and others). Luikov (1964, 1966a-b, 1975), who is credited by Russian researchers to have discovered the phenomenon of thermodiffusion about three decades ago, developed the concept of moisture transfer potential (moistness), as the potential for water movement in a capillary-porous body. This concept has not been commonly accepted in Western literature. Berger and Pei (1973) stated that Luikov's assumptions of constant "moisture diffusivity" and "specific moisture capacity" lead to an "apparent mathematical paradox if his model is extended to the nonisothermal drying."

METHODS

Heat and Mass Conservation Equations

Theories to describe transport phenomena in capillary-porous media come either from a mechanistic (classical) approach (Philip and DeVries, 1957; DeVries, 1958; Krischer, 1963) or from irreversible thermodynamics concepts (Luikov, 1966; Valchar et al., 1966). Basic differences can be found between these two approaches. Fortes and Okos (1978) presented a theory that is based on both irreversible thermodynamics and mechanistic concepts. This theory is an attempt to incorporate most of the more recent approaches. Fortes (1978) and Fortes and Okos (1978) derived the following heat and mass transfer equations, applicable to hygroscopic capillary-porous media. Liquid flux:

$$\vec{J}_{l} = -K_{l} \left(\rho_{l} R_{v} \ln H \nabla T - \rho_{l} \frac{R_{v} T}{H} \frac{\partial H}{\partial M} \nabla M \right)$$
(1)

Vapor flux:

$$\vec{J}_v = -K_v \left(\left(\rho_{vo} \frac{\partial H}{\partial T} + H \frac{d\rho_{vo}}{dT} \right) \nabla T - \rho_{vo} \frac{\partial H}{\partial M} \right) \nabla M \quad (2)$$

Heat flux

$$\vec{J}_{q} = -K_{T}\nabla T - \left[\rho_{l}K_{l}R_{r}\ln H + K_{r}\left(\rho_{ro}\frac{\partial H}{\partial T} + H\frac{d\rho_{ro}}{dT}\right)\right]\frac{R_{r}T^{2}}{H}\frac{\partial H}{\partial M}\nabla M \quad (3)$$

Mass conservation:

$$\rho_s \frac{\partial M}{\partial t} = - \nabla \cdot (\vec{J_t} + \vec{J_c}) \tag{4}$$

Energy conservation:

$$\rho_s c_b \frac{\partial T}{\partial t} - \rho_s L_w \frac{\partial M}{\partial t} = - \nabla \cdot \vec{J_q} - L_r \nabla \cdot \vec{J_r}$$
 (5)

Some remarks should be made concerning Eqs. 1 to 5:

- The above equations do not require any specific mechanisms for moisture migration.
- The isothermal driving-force for both liquid and vapor movement inside the grain kernel is taken as the gradient in equilibrium moisture content, as defined by an isotherm. Thus, water may migrate from regions of lower to those of higher moisture content as long as the equilibrium moisture content gradient effect is dominant and favors this movement.
- As expressed by Eq. 1, moisture in liquid phase can move from colder to hotter regions. Such phenomenon has been experimentally observed in cases of non-isothermal moisture transfer in soil (Gurr, et al., 1952).
- The heat of adsorption and latent heat of vaporization or condensation of water inside the porous medium is taken into account in Eq. 5.
- It is assumed that the medium is isotropic, continuous and that shrinkage is negligible.
- Convective (bulk) movement of water inside the medium is neglected.

The approach taken in this work assumes that the product to be dried is isotropic, cylindrical, does not shrink and that there is no preferential path for moisture migration.

Boundary Conditions

In this work, we are mainly interested in the transport phenomena analysis of cylindrically extruded corn meal submitted to temperatures lower than 160°C. Due to the mixing and size of the cylindrical extrudate, it is furthermore assumed radial symmetry and isotropy.

Eqs. 1 to 5, which constitute a set of nonlinear partial differential equations are to be solved in cylindrical coordinates with the following third order boundary and initial conditions.

at t = 0

$$M(r, o) = M_o, \qquad T(r, o) = T_o \tag{6}$$

at r = 0

$$\vec{J_t} + \vec{J_r} = 0, \qquad \vec{J_g} = 0 \tag{7}$$

at r = R

$$\vec{J_l} + \vec{J_r} = h_m(P_r - P_{va}), \qquad \vec{J_q} + L_v \vec{J_l} = h_T(T_s - T_a)$$
 (8)

In order to numerically solve the above equations, the following physical parameters, not available from the literature, had to be obtained: H, K_T , ρ_s , K_r , and K_l . The first three of these parameters were evaluated experimentally, while K_r and K_l were obtained from the first 300 seconds (5%) of the drying curve through a search technique. The heat capacity, c_b , was obtained from (Heldman, 1975):

$$c_b = (0.34X_c + 0.37X_p + 0.4X_f + 0.2X_a + 1.0X_l) \times 4180 (J/kg \cdot K)$$
 (9)

The saturated vapor density was obtained from the expression derived by Baladi (1975):

$$\rho_{ro} = \frac{2.54 \times 10^8}{T} e^{-5200/T} \text{ (kg/m}^3)$$
 (10)

The heat of vaporization and adsorption (wetting) of water in the extruded material are given by:

$$\Delta h_r = \Delta h_o + \Delta h_w \tag{11}$$

$$\Delta h_w = \frac{R_v T^2}{H} \frac{\partial H}{\partial T} \tag{12}$$

where Brooker (1967):

$$\Delta h_o = 3.11 \times 10^6 - 2.11 \times 10^3 T \quad (J/kg)$$
 (13)

In order to evaluate heat and mass transfer coefficients, the Chilton-Colburn (1934) analogies were employed, namely:

$$J_H = J_D \tag{14}$$

where:

$$J_{H} = NuRe_{mod}^{-1}Pr^{-1/3}$$
 (15)

and:

$$J_D = Sh \cdot Re_{mod}^{-1} \cdot Sc^{-1/3} \tag{16}$$

These factors allow the estimation of h_t and h_m once the relationship between j_H and Re are known (Sherwood and Pigford, 1952).

Other values for the physical properties of water (ρ_l, C_l, C_c) can be obtained directly from Perry and Chilton (1973).

Extruder and Extrudate

The degerminated corn meal, employed in this research, had the following approximate composition: fat = 0.7%, protein = 8%, ash = 0.3%, fiber = 0.4%, carbohydrate = 78.5% with an initial moisture content of 12.1% d.b. The corn meal was rewetted to a moisture content of approximately 44% d.b. and then extruded

A Brabender extruder, type 2504, number 864, was used in this research. The operating conditions were: screw speed = 30 rpm., barrel temperatures: first zone = 40°C, second zone = 60°C, third zone = 80°C and the fourth zone = 120°C. The barrel length was 66 cm and the diameter was 2.6 cm. The dye diameter was 0.3 cm. The extrudate at these operating conditions suffered a slight swelling. This swelling practically disappeared after a few minutes and was not considered in this work.

Determination of the Isotherm Curves

The experimental determination of the desorption curves of equilibrium moisture contents vs. relative humidity, at different temperatures, i.e., the isotherms, were made through the use of saturated salt solutions. Experimental conditions and the salts employed are shown in Table 1. Measurements were made every other day to ascertain the amount of water desorbed. Equilibrium was assumed after two consecutive readings did not indicate any further change in water content of the extrudate. Since drying is a desorption phenomenon, adsorption isotherms were not investigated.

Thermal Conductivity and Center Temperature

The thermal conductivity of the extruded corn meal was obtained through the method suggested by DeVries (1952), for soils, and later by Sweat (1976), in a form adapted for food products.

The device consisted of a hypodermic needle, inside which is located a thermocouple and an electric wire heater. This device was inserted in the cylindrical sample in such a manner that no metallic part of the probe was exposed to the atmosphere. A computer assisted automatic acquisition system collected data at 10 points per second and furnished all the statistics needed. The data was collected from experiments conducted between 30-95°C and 8-36% H₂O d.b. A more detailed presentation of the calibration technique and the acquisition system is given in Fortes (1978).

TABLE 1. SATURATED SALT SOLUTIONS USED IN THE DETERMINATION OF THE ISOTHERMS

Salt	Relative Humidity (%)	Temperature	Ref.
LiCl	12.0	27.0	2
	13.3	50.0	2
	11.0	70.0	2 2
	11.9	90.0	*
KF 2H ₂ O	27.4	30.2	1
-	20.4	50.0	1
	22.0	70.0	1
	23.5	90.0	1
MgCl ₂ 6H ₂ O	33.0	27.0	1
82	30.2	70.0	1
NaI 2H ₂ O	36.4	30.2	1
	22.6	70.0	1
NaNO ₂	63.0	30.1	1
	59.8	50.0	1
	58.9	70.0	1
	58.5	90.0	*
NaCl	75.7	27.0	1
,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	74.9	50.0	ī
	75.1	70.0	ī
	77.0	90.0	*
K ₂ SO ₄	97.0	27.0	2
4 4	94.8	70.0	2

^{1 =} Carr and Harris (1949).

The center temperature was obtained by inserting the hypodermic needle in the sample and submitting the sample to the drying conditions. A digital voltmeter was used to measure the temperature at any time. Center-temperature measurements were taken so that predicted values could be compared with experimental values. These center temperature measurements were planned to help in verifying the proposed model.

The density of the material was made by measuring the volume of a sample in an air-comparison pycnometer and weighing the same amount.

The Dryer

The drying apparatus consisted basically of:

- 1. A climate chamber, which furnished air to the drier at a constant dew-point temperature, adjustable from 4.4-35.5°C, and constant dry-bulb temperature adjustable from 4.4-48.9°C. The climate chamber controllers for dew-point temperature and dry-bulb temperature were pneumatic, proportional with reset (Honeywell). Two independent chart recorders displayed the output air conditions of dew-point and dry-bulb temperatures. Also, the air mass flow could be controlled through dampers, internally built inside the climate chamber and manually adjustable.
- 2. The dryer used had a square cross-sectional area of 0.0929m. Three Chromalox CAB-611, Chrome steel sheath heaters, activated by a Chromalox CSRC, 50 A, power control unit, allowed temperature to be adjusted up to 150°C. The dry-bulb temperature could be sensed through a Leeds and Northrup 8920 resistance thermooeter, wtoee signal was linearized by a Leeds and Northrup 479 temperature/millivolt transmitter and displayed in a three-pen chart recorder.

The output of the transmitter was also connected through a Leeds and Northrup proportional-integral-derivative (P.I.D.) electronic controller, to the SCR power control. Input air (from the climate chamber) relative humidity could be double-checked with an Aminco Hygrosensor (humidity sensor), whose output signal was linearized by a Hygrotran transmitter and fed to the chart recorder. The thin-layer of extrudate was placed

^{2 =} Gustafson (1972).

^{*} Extrapolated values. Some values were interpolated for room temperature conditions.

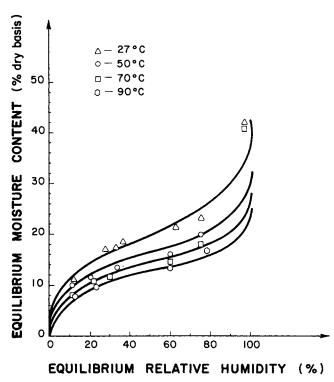


Figure 1. Equilibrium isotherms for extruded degerminate corn meal.

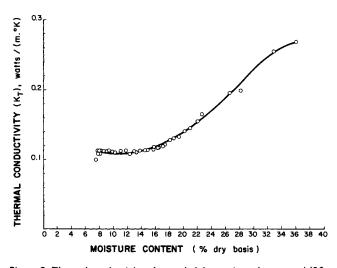


Figure 2. Thermal conductivity of extruded degerminated corn meal (30-90°C).

inside the dryer on a wire-mesh tray $(0.30\times0.30\,\mathrm{m}^2)$. The initial sample weight was 200 grams. The sample weight during drying could be continuously obtained from a Statham UC-2 transducing cell, and a Statham UL4-5, 2.27 kg (5 lb) load cell adapter. The output signal was amplified and linearized by a Statham SC-1105 bridge amplifier and a Dana 2850 amplifier.

Calibration could be performed through standard weights, under conditions of the experiment at hand. Weight variations within 0.5g could be detected. Air velocity was measured through three different devices: air vane anemometer, pitottube and hot wire anemometer. The readings of the vane anemometer were more consistent since readings could be taken in longer intervals of time (5 minutes); thus, reading oscillations could be minimized.

Numerical Solution and Evaluation of K_ℓ and K_e

The "Method of Lines" was used to solve the system of non-linear partial differential Eqs. 1-8. This method consists in

Table 2. Physical Properties of Extruded Degerminated Corn Meal

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Property	Expression*	Unit	
Н	$1 - \exp[-2.736(T - 273.16)^{1.242}M^{3.379}]$	decimal	
K_T	$0.1153 - 2.936\dot{M}^2 + 25.4M^3 - 38.71M^4$	watts/(m k)	
s	1445	kg/m³	
c_b	4180(0.343 + M)/(1 + M)	J/(kg K)	
R	1.59×10^{-3}	m	
h_w	$1595T^{2.242}M^{3.379}(1-H)/H$	J/kg	
h_v	$3.11 \times 10^6 - 2.38 \times 10^3 T + L$	J/kg	
K_{l}	$4.149 \times 10^{-7} H^3 \exp(-2747/T)$	s	
K_v	$9.085 \times 10^{-8} (T - 273.16)^{0.3016} (H^{0.25} - H^{1.25})$	m²/s	

^{*} All temperatures in °K.

discretizing the spatial variable, leading to a system of ordinary differential equations (Madsen and Sincovec, 1974; Sincovec and Madsen, 1974). The method of Gear (1971) was used to solve the resulting system of ordinary differential equations.

The liquid and vapor conductivities were evaluated by first finding K_l and K_r values through a nongradient bidimensional search technique using the drying data from the first 300 to 600 seconds of the drying curve as discussed in Fortes (1978) and then proposing a model for K_l and K_r . The criterion for the search is the minimization of the squares of the differences between the theoretical and experimental values obtained for the drying curve.

RESULTS AND DISCUSSION

Physical Properties

Figure 1 depicts the experimental and predicted isotherms for extruded corn meal, in the temperature range from 27-90°C. Henderson's (1952) equation was used to fit the experimental data and the nonlinear regression analysis and resulted in:

$$H = 1 - \exp[-2.736T^{1.242}M^{3.379}]$$
 (17)

The thermal conductivity of the product was found to be related to its moisture content by:

$$K_T = 0.1133 - 2.936M^2 + 25.44M^3 - 38.71M^4 \quad (W/m \cdot k)$$
(18)

with:

regression coefficient = 0.9926

standard deviation = 0.00477

Figure 2 shows the experimental data obtained. The data were obtained in the temperature range from 30-95°C. Data points in Figure 2 are the averages of at least 10 independent readings, obtained at a rate of 10 readings/s. Chauvenet's criterion was used to eliminate spurious results. After the experiment was over, the sample was left to cool and the thermal conductivity was again evaluated. No significant difference was noted between the thermal conductivity at 95°C and 30°C at a given moisture content. Because of the short time needed to determine the thermal conductivity (1/10 second), moisture migration is negligible and all data can then be considered as evaluated at a given average moisture content.

The density of the bone-dry solid (ρ_s) obtained from five replicates was 1445 kg with a standard deviation of 5 kg. The specific heat was calculated from the previously given composition of the extruded corn meal, Table 2.

Model for K_i and K_c

As a food material is dried, shrinkage, starch gelling, presence of solutes, deformation of structure and chemisorptive forces drastically change the resistance to vapor movement (Keey, 1973; Gorling, 1958). Since foods are highly complex, our present theoretical understanding of liquid and vapor diffusivity or conductivity through porous solids does not enable us to

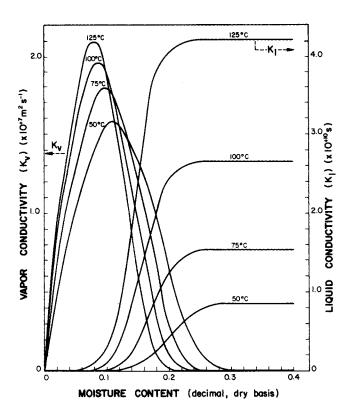


Figure 3. Effect of temperature and moisture content on liquid and vapor conductivities.

estimate the variations of moisture movement from first principles. However, it is a well-known fact that both the liquid and vapor conductivity are related to the structure and composition of the food product. The equilibrium moisture isotherm is also related, to a significant degree, to the structure and composition of the food product. It, therefore, can be assumed that a direct relationship exists between the characteristic equilibrium moisture isotherm of a food product and the formulation of liquid and vapor conductivity models.

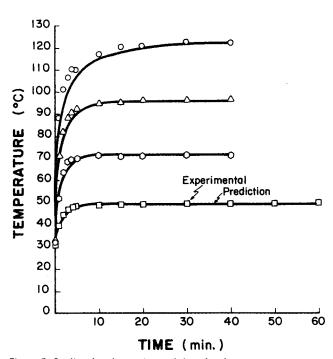


Figure 5. Predicted and experimental data for the center temperatures (□60°C, ○75°C, △100°C, ○125°C).

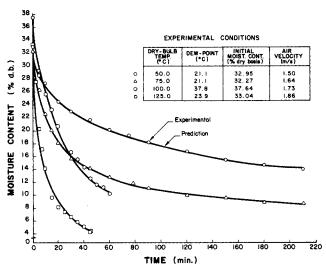


Figure 4. Drying data and predicted drying curves at different experimental conditions.

The relationship between diffusivity and moisture content in wet solids is a complex problem. One possible model can be based on the work of Luikov, (1966) and Philip and DeVries (1957). At low moisture contents and low relative humidities, monomolecular adsorption predominates and moisture moves primarily by vapor phase diffusion. The region in which monomolecular adsorption takes place lies generally below a relative humidity of 20-25%. It was assumed that the vapor conductivity increases from 0 at a relative humidity of 0% to a maximum value where multimolecular adsorption starts. The vapor conductivity goes to zero at low moisture contents since moisture transport becomes increasingly controlled by transport in the finest pores whose diameter is of the same order as the near free path of the molecules (Scott and Dullien, 1962). Also the large number of polar sites available for moisture adsorption would greatly reduce any moisture movement. Moisture begins adsorbing in multimolecular layers above a relative humidity of approximately 25%. A rapid decrease in vapor conductivity takes place with increases in amounts of moisture adsorbed due to subsequent filling of the pores. The vapor conductivity will decrease to approximately 0 at a relative humidity of approximately 95%.

At the onset of multimolecular adsorption, the liquid conductivity increases sharply until saturation takes place. Upon saturation, a relative humidity greater than 95%, the liquid conductivity levels off to essentially a constant value at a given

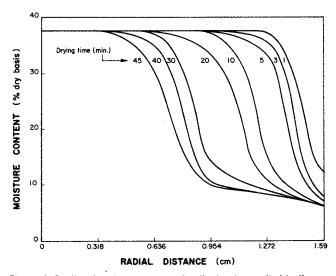


Figure 6. Predicted moisture content distribution in a cylindrically extruded corn meal at 100°C drying temperature.

Table 3. Distribution of Mass and Heat Flux Inside Product (Time = 1800 s; Temp. = 75°C)

	M.C.	Temp.	$J_c \mathrm{kg/m^2 \cdot s}$		$J_t \text{ kg/m}^2 \cdot \text{s}$		$J_q \text{ J/m}^2 \cdot \text{s}$	
Position (cm)	db	(°C)	Jet	Jem	J_{lT}	J_{lm}	J_{qT}	J_{qm}
0.0 center	0.323	71.8						
0.318	0.323	71.8	-5.86×10^{-14}	4.74×10^{-15}	3.01×10^{-15}	6.15×10^{-10}	-9.18×10^{-1}	7.89×10^{-9}
0.636	0.312	71.8	-8.67×10^{-1}	9.33×10^{-13}	4.46×10^{-14}	3.12×10^{-8}	-3.36×10	1.56×10^{-6}
0.954	0.164	71.8	-4.70×10^{-7}	3.89×10^{-5}	1.27×10^{-8}	5.32×10^{-5}	-8.66×10^{1}	8.22×10^{1}
1.272	0.128	72.3	-1.54×10^{-6}	3.94×10^{-5}	1.86×10^{-8}	1.03×10^{-5}	-2.26×10^{2}	9.23×10^{1}
1.59 (surface)	0.072	73.1	-3.97×10^{-7}	3.92×10^{-5}	4.13×10^{-10}	3.18×10^{-7}	-2.81×10^{2}	9.86×10^{1}

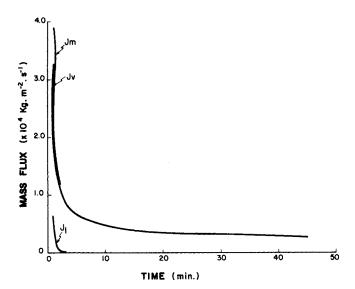
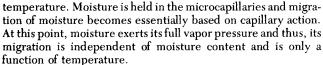


Figure 7. Predicted mass fluxes at the surface of a cylindrically extruded corn meal at 100°C drying temperature.



In summary, the results of the various workers, and results from this work, suggest that vapor phase diffusivity passes through a maximum at low moisture contents (the onset of multimolecular adsorption) and then rapidly decreases to 0 as the liquid phase diffusivity continuously rises with an increase in moisture content. The liquid conductivity eventually levels off when saturation is reached. The total moisture conductivity will then vary as the sum of the liquid phase and vapor phase conductivities which vary with the temperature and relative humidity at a particular point in the porous body.

Figure 3 shows the dependence of the liquid and vapor conductivities on moisture content and temperature for the extruded corn product used in this work. The mathematical relationship for K_l and K_r used in this work is given in Table 2. Both conductivities are highly dependent on moisture content. The vapor conductivity seems to be less affected by a change in temperature than is the liquid conductivity. While there is a shift of the peak to the left of the moisture content axis, in the case of vapor conductivity, the opposite is true for the "plateau" of the liquid conductivity curve.

Some possible phenomena which contribute to the behavior of the vapor conductivity as a function of temperature are: a) more polar sites become available for desorption and adsorption: b) thermal expansion reduces the resistance to vapor flow; and c) at higher temperatures, the total pressure effect may no longer be negligible and its effect is "lumped" in the expression for vapor conductivity. In the case of liquid conductivity, thermal expansion and the total pressure effect (filtration) may also contribute significantly to the overall liquid flow.

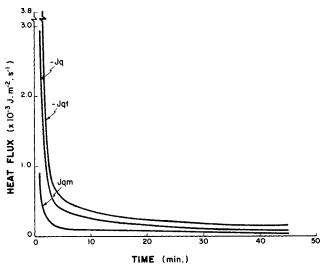


Figure 8. Predicted heat fluxes at the surface of a cylindrically extruded corn meal at 100°C drying temperature.

Analysis of the Drying Process

Figures 4 and 5 show the experimental data and predicted curves for the drying and center temperature changes of the corn meal. The liquid and vapor conductivities were determined from the first 300-600 seconds of drying. The expressions for K_l and K_r were used in the model to predict the rest of the drying curve and to predict the center temperature. Predicted values agreed with experimental data within 3% and these results serve to justify the theory.

To test the sensitivity of the liquid and vapor conductivity expressions, a series of runs were made to determine how the extent of change in K_l and K_r would effect the average moisture content and the rate of drying. At a temperature of 125°C after

TABLE 4. SURFACE MASS AND HEAT FLUX AT 75°C

Time (s)	$J_{rT} imes 10^6$	$J_{vm} \times 10$	$J_{tT} \times 10^9$	$J_{lm} \times 10^6$
60	-5.622	2.690	79.63	82.61
120	-2.983	1.699	9.408	5.827
180	-1.794	1.419	3.294	2,402
240	-1.540	1.408	2.469	1.999
300	-1.451	1.366	2.237	1.843
600	-0.8400	0.7684	1.057	0.7971
900	-0.4985	0.4992	0.5403	0.4276
1200	-0.4488	0.4816	0.4728	0.3977
1500	-0.4602	0.4661	0.4905	0.3907
1800	-0.3969	0.3915	0.4132	0.3184
2100	-0.3172	0.3237	0.3182	0.2511
2400	-0.2705	0.2900	0.2650	0.2183
2700	-0.2633	0.2784	0.2547	0.2090
3000	-0.2610	0.2650	0.2556	0.1995
3300	-0.2386	0.2415	0.2315	0.1795
3600	-0.2084	0.2164	0.1991	0.1578
3900	-0.1868	0.1965	0.1767	0.1414

1200 seconds, a 20% increase in K_l and K_v gave a 16.3% decrease in the average moisture content, a 20.5% increase mass flux and a 17.0% decrease in heat flux. Similar results were obtained at other temperatures. Since the model predicted drying within 3%, the expressions obtained for K_l and K_r do indeed give reasonable representation for the drying of extruded corn grits under the conditions used in this experiment. However, since K_t and K_v were obtained through "fitting", the expressions obtained do not necessarily prove the theory but give a very strong indication of the validity of the technique.

A moving evaporation front was predicted for the drying conditions investigated (50-125°C). A typical form of the predicted moisture distribution inside the product is given in Figure 6. Thus, moisture evaporation is faster than moisture migration inside the porous medium.

Liquid flux was found to be the most important mechanism for moisture migration inside the medium, in the regions not affected by the moving evaporation front. The relative humidity in the inner regions has a value close to 1 and an error can be made when it is assumed that the effect of the temperature gradient on moisture migration is negligible Eqs. 1 and 2. For H = 0.9999, $\ln H = 1.00*10^{-4}$, the temperature term can become the same order of magnitude as the term dependent on ∇M for regions closer to the center. In general, however, liquid moisture transfer is predominantly due to a gradient in equilibrium moisture content $(J_{lm}/J_{lT} \approx 10^3)$ inside the particle as given in Table 3 for

Also as shown in Table 3, vapor transfer is predominantly due to a gradient in equilibrium moisture content for all drying conditions investigated $(J_{vm}/-J_{lT} \approx 10^2)$. J_v becomes negligible for regions closer to the center, i.e., regions not affected by the moving evaporation front. Figure 7 shows the predicted mass fluxes at the surface. The numerical solution implied that at the surface vapor transfer is more important than liquid transfer, however, the latter not being negligible, Table 4.

Heat transfer is basically due to conduction for drying conditions where $T < 100^{\circ}$ C and for regions not close to the moving evaporation front. J_{qm} , the heat flux due to moisture migration, is significant at higher temperatures and in the latter stages of drying, when a good portion of the input heat is used to evaporate the moisture. Figure 8 shows the relative importance of J_{qm} and J_{qT} at the surface.

CONCLUSIONS

The model for transport phenomena in capillary-porous media was found to accurately predict drying of extruded corn grits for the temperature range of 50C to 125C and an initial moisture content of 35% H_2O w.b. The values for K_l and K_r determined from the first 5 to 10 minutes of drying accurately predicted the remaining portions of the drying curve and center temperature.

A moving evaporation front was shown to predominate over the entire drying condition. The liquid and vapor fluxes were found predominate due to the gradient in equilibrium moisture content. Moisture migration was found to have a significant effect in heat transfer at higher temperatures and during the latter stages of drying.

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NOTATION

= specific heat, $J/kg \cdot K$ D_{ν} = equivalent diameter, m

 Δh_r = specific latent heat of vaporization, J/kg = specific differential heat of wetting, J/kg

= specific latent heat of pure water vaporization, J/kg

 h_T = heat transfer coefficient, I/S m² Pa h_m = mass transfer coefficient, kg/S · m² · Pa

Η = relative humidity, decimal

 J_H = Chilton-Colburn factor for heat transfer = Chilton-Colburn factor for mass transfer

= liquid flux, kg/m2s

= heat flux, J/m²s = vapor flux, kg/m2s

 $J_D \vec{J}_l \vec{J}_q \vec{J}_v K_l K_v$ = liquid conductivity, seconds = vapor conductivity, m2s

 K_T = apparent thermal conductivity, W/m $\cdot k$

= specific enthalpy, J/kg

= moisture content (dry basis), kg/kg; \overline{M} = average moisture content (dry basis), kg/kg

P = vapor pressure, Pa

= radius, m

R = radius, m (radial length) T= thermodynamic temperature, K

= weight fraction, kg/kg

Greek

= chemical potential, I/kg, or kinematic viscosity, kg/m²s μ

= density; kg/m³

= nabla (or del) operator

Subscripts

= surrounding medium (humid air) or ash

b

= carbohydrate \boldsymbol{c}

= fat

= liquid phase

= mass transfer characteristics m

= saturation state, initial or osmotic 0

= protein or pressure p

= heat transfer characteristics q

= solid skeleton, surface tension T

= heat transfer characteristics

= vapor phase

= wetting

Dimensionless Parameters

 $Re_{mod} = \frac{\mu_{air}vD_{\nu}}{\mu_{air}} = \text{modified Reynolds number}$

= Nusselt number Nu

Pr= Prandtl number

= Schmidt number Sc

= Sherwood number Sh

LITERATURE CITED

Baladi, J. Y., Transient Heat and Mass Transfer in Soils, Ph. D. Thesis, Purdue University, W. Lafayette, IN (1975).

Baladi, J. Y., R. J. Schoenhals, and D. L. Ayers, Transient Heat and Mass Transfer in Soil, AIAA-ASME Thermophysics and Heat Transfer Conference, Palo Alto, CA (May 24-26, 1978); ASME Publication

Berger, D., and D. C. J. Pei, Drying of Hygroscopic Capillary Porous Solids, A Theoretical Approach, Int. J. Heat Mass Transfer, 16, 293

Brooker, D. B., Mathematical Model of the Psychrometric Chart, Trans., ASAE, 10, 558 (1967).

Cary, J. W., and S. A. Taylor, The Interaction of the Simultaneous Diffusion of Heat and Water Vapor, Soil Sci. Soc. Am. Proc., 26, 413 (1962).

Ceaglske, N. H., and O. A. Hougen, The Drying of Granular Solids, Trans., AIChE, 33(3), 283 (1937).

Chilton, T. H., and A. P. Colburn, Mass Transfer (Absorption) Coefficients, Prediction from Data on Heat Transfer and Fluid Friction, I&EC, 26, 1183 (1934).

DeVries, D. A., Nonstationary Method for Determining Thermal Conductivity of Soils in situ," Soil Science, 73, 83 (1952).

DeVries, D. A., "Simultaneous Transfer of Heat and Moisture in Porous Media," Trans. Am. Geophys. Union, 39(5), 909 (1958).

Fortes, M., A Nonequilibrium Thermodynamics Approach to Transport Phenomena in Capillary-Porous Media with Special Reference to Drying of Grains and Foods, Ph.D. Thesis, Purdue University, W. Lafayette, IN (1978).

Fortes, M., and M. R. Okos, "A Nonequilibrium Thermodynamics Approach to Transport Phenomena in Capillary-Porous Media, Proc. of the 1st International Symposium on Drying, McGill Univ., Montreal, Canada (1978).

Gear, C. W., Numerical Initial Value Problems in Ordinary Differential Equations, Prentice Hall, Englewood Cliffs, NJ (1971)

Gorling, P., "Physical Phenomena During the Drying of Foodstuffs," Fundamental Aspects of the Dehydration of Foodstuffs, Society of the Chemical Industry, London (1958).

Gurr, C. G., T. J. Marshall, and J. T. Hutton, "Movement of Water in Solid due to a Temperature Gradient," Soil Science, 74, 335 (1952).
Gustafson, R. J., Equilibrium Moisture Content of Shelled Corn from

50 to 155F, MS Thesis, University of Illinois (1972).

Haines, W. B., "Studies on the Physical Properties of Soils: IV. A Further Contribution to the Capillary Phenomena in Soil," J. Agric. Sci., 17, 264 (1927).
Haines, W. B., "Studies on the Physical Properties of Soils: V. The

Hysteresis Effect in Capillary Properties and the Modes of Moisture Distribution Associated Therewith, J. Agric. Sci., 20, 97 (1930).

Harmathy, T. Z., Simultaneous Moisture and Heat Transfer in Porous Systems with Particular Reference to Drying, Ind. Eng. Chem. Fun-

damentals, 8(1), 92 (1969).

Henderson, S. M., "Progress in Developing the Thin-Layer Drying

Equation," ASAE Trans., 17, 1167 (1974).

Henderson, S. M., "A Basic Concept of Equilibrium Moisture," Agr. Engr., 33, 29 (1952).

Henry, P. S. H., "Diffusion in Absorbing Media," Proc. R. Soc., London, 171A, 215 (1939).

Hougen, O. A., H. J. McCauley, and W. R. Marshall, Jr., Limitations of Diffusion Equations in Drying, AIChE Trans., 36(2) 183 (1940).

Keey, R. B., Drying: Principles and Practice, Pergamon Press, Oxford

O., Die Wissenschaftlichen Grundlagen der Krischer, Trocknungstechnik, Springer, Berlin (1963)

Lewis, W. K., "The Rate of Drying of Solid Materials," J. Ind. Eng. Chem., 13, 427 (1921).

Luikov, A. V., Heat and Mass Transfer in Capillary-Porous Bodies, T. F. Irvine and J. P. Hartnett, eds, 1, Academic, New York (1964).

Luikov, A. V., Heat and Mass Transfer in Capillary-Porous Bodies, Pergamon Press, Oxford (1966).

Luikov, A. V., "Applications of Irreversible Thermodynamic Methods to Investigation of Heat and Mass Transfer," Int. J. Heat Mass Transfer, 9, 139 (1966).

Luikov, A. V., "Systems of Differential Equations of Heat and Mass

Transfer in Capillary Porous Bodies (review)," Int. J. Heat Mass Transfer, 18, 1 (1975).

Madsen, N. K., and R. F. Sincovec., "The Numerical Method of Lines for the Solution of Nonlinear Partial Differential Equations," Computational Methods in Nonlinear Mechanics, J. T. Oden, E. B. Becker et al., eds., Texas Inst. for Computational Mechanics, Austin, TX

Mustakas, G. C., et al., Extruder-Processing to Improve Nutritional Quality Flavor, and Keeping Quality of Full-Fat Soy Flour," Food Technology, 124, 102 (1970).

Peck, R. E., and D. T. Wasan, "Drying of Solid Particles and Sheets," AIChE, 247 (1975).

Perry, R. H., and C. H. Chilton, Chemical Engineers' Handbook, McGraw-Hill, New York (1973).

Philip, J. R., and D. A. DeVries, "Moisture Movement in Porous Materials Under Temperature Gradients," Trans. Am. Geophys. Union, 38(2), 222 and 594 (1957).

Rocques, M. A., and A. R. H. Cornish, "Fundamental Study of the Transport Equations for Wet Porous Media," The 2nd Pacific Chemical Engineering Congress, 516 (1977).

Scott, D. S., and F. A. Dullien, "Diffusion of Ideal Gasses in Capillaries and Porous Solids," AIChE J., 8(1), 113 (1962).

Sherwood, T. K., "The Drying of Solids," Ind. Eng. Chem., Part I., 21,

Sherwood, T. K., "The Drying of Solids," Ind. Eng. Chem., Part II, 21, 976 (1929)

Sherwood, T. K., "Application of the Theoretical Diffusion Equations to the Drying of Solids," AIChE, 27, 190 (1931).

Sherwood, T. K., and R. L. Pigford, Adsorption and Extraction. 2nd ed., McGraw-Hill, New York (1952).

Sincovec, R. F., and N. K. Madsen, "Software for Nonlinear Partial Differential Equations," ACM Trans. on Math. Software, 1, 232

Smith, O. B., Versatility of Texturizing by Extrusion Cooking, 67th Annual Meeting, AIChE, Washington (1974).

Smith, O. B., "Textured Vegetable Proteins," World Soybean Research Conf., University of Illinois (1975).

Sweat, V. E., "A Miniature Thermal Conductivity Probe for Foods," ASME, Publication No. 76-HT-60 (1976)

Taylor, S. A., "The Influence of Steady Temperature Gradients Upon Water Transport in Soil Materials," *Humidity and Moisture*, 3, Reinhold, New York (1965).

Valchar, [., "Heat and Moisture Transfer in Capillary Porous Materials from the Point of View of the Thermodynamics of Irreversible Processes," 3rd Inst. Heat Transfer Conf., 409, Chicago (1966)

Walsh, K. E., and B. R. Funke, "The Influence of Spaghetti Extruding, Drying and Storage on the Survival of Staphylococcus Dureus," Journal of Food Sci., 40, 714 (1975).

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Interpretation of Surface Flow Phenomenon of Adsorbed Gases by Hopping Model

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The mechanism of surface flow of adsorbing gas molecules through the porous adsorbent is interpreted, and a new hopping model is derived by separately taking into account the hopping behaviors of monolayer and multilayer molecules. The reported experimental results containing the data measured here are correlated well. Finally, some considerations are given to the two experimental constants appearing in the model.

Besides gaseous diffusion and flow in pores, surface flow of physically adsorbed gases is important in evaluating the appar-

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ent mass transfer rates through adsorptive porous materials. The most commonly used models to interpret the surface flow mechanism are essentially divided into the following.

1. Hydrodynamic model, which regards the phase of the adsorbed gas on the solid surface as a laminar-flowing film of viscous liquid (Babbit, 1950; Gilliland et al., 1958).