EXPERIMENTAL AND THEORETICAL STUDIES ON DRYING OF FOOD MATERIALS

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A mathematical model was developed to predict heat and moisture transfer, drying-induced stresses and the material shrinkage of viscoelastic food materials during drying. Potato starch was selected as the model food material and physical and material properties such as moisture diffusivity, thermal conductivity, and viscoelastic properties were measured as a function of temperature and moisture concentration. To verify the numerical model, results of the drying experiments were compared with the numerically obtained results and were found to be in good agreement with each other.

Introduction

In food materials such as cereals, the moisture is removed by way of drying to minimize spoilage during storage. However when dried, foods tend to crack if the value of the internal stress induced by temperature and moisture distributions in the material exceeds the value of the breakage stress. Since cracks invariably cause the food material to deteriorate, and thereby reduce the quality of the food and its ability to be stored, it is absolutely essential to dehydrate food materials without subjecting them to harsh drying conditions that could induce material failure. At the same time, in order to run the process efficiently, it is also important to dry the materials at higher drying rate. Therefore, in drying processes for food materials, it is imperative to know the optimum process conditions.

Recently, various mathematical models have been proposed to evaluate drying-induced stresses in food materials and to find the most favorable drying conditions. Gustafson *et al.* (1979) carried out finite element analysis to find the transient heat transfer and thermal stresses in a corn kernel assuming it to be an elastic body. Tsukada *et al.* (1991) analyzed the stress distribution in hydrated starch subjected to a drying process in which they assumed it to be an elastoplastic material. But since most food materials in general display characteristics of both elastic solids and viscous fluids, i.e. viscoelastic bodies, it is necessary to include the viscoelastic properties of the substance when numerical simulations are carried out to determine the stresses originating in the material during drying.

Previous studies on thermo-hydro stresses in viscoelastic materials were restricted to simple geometries (Litchfield and Okos, 1988, Rao *et al.*, 1975, Yamaguchi *et al.*, 1985). Haghighi and Segerlind (1988a,b,c) derived

a mathematical model to analyze drying-induced stresses in a viscoelastic material by use of the finite element method. However, in their simulation studies they constricted their analysis by assuming soybean to be an elastic material due to the unavailability of the viscoelastic data of the material. Itaya *et al.* (1995) developed a three dimensional model to evaluate stress distribution in a viscoelastic composite body, yet in their analysis the viscoelastic properties were considered to be independent of moisture, although it has been shown by several authors (Cummings and Okos, 1972, Herum *et al.*, 1979, Mensah *et al.*, 1984, Pappas and Rao, 1989, Yamaguchi *et al.*, 1981) that the viscoelastic properties of food materials vary considerably with moisture concentration. For establishment of a mathematical model which can describe quantitatively the drying-induced stress in viscoelastic food materials, it is necessary to verify the model by utilizing in the numerical simulation accurate physical properties of the material measured as a function of temperature and moisture concentrations.

The object of this work is to develop a mathematical model to predict the internal stress induced during the drying process of a viscoelastic material, to measure properties of the material and their temperature and moisture dependencies such as moisture diffusivity, thermal conductivity, viscoelasticity and shrinkage, and to carry out the drying experiment. The numerical model built in the present work can perform the simulation of heat transfer, mass transfer and internal stresses in an axisymmetric body subjected to drying, and also take into consideration shrinkage of the material by employing moving coordinates. The accuracy of the model is ascertained by comparing the numerical results with results of the drying experiment.

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Fig. 1 Cylindrical coordinates used in calculation

1. Numerical Analysis

Only one quarter of the cross section of the cylindrical material was considered because of the symmetry as shown in Fig. 1. Assuming that diffusional properties are independent of direction, the diffusions of moisture and heat in an axisymmetric body are governed by the following equations.

for moisture:

$$\frac{\partial C}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left(r D(C, T) \frac{\partial C}{\partial r} \right) + \frac{\partial}{\partial z} \left(D(C, T) \frac{\partial C}{\partial z} \right) \quad (1)$$

for heat:

$$\rho c_p \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left(r K(C, T) \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left(K(C, T) \frac{\partial T}{\partial z} \right) \quad (2)$$

Initial conditions are expressed as follows.

$$C = C_0, \quad T = T_0 \quad (3)$$

Boundary conditions at the material surface for moisture and temperature are given by Eqs. (4) and (5), respectively.

$$-D(C, T) \frac{\partial C}{\partial n} = h_m (C - C_{eq}) \quad (4)$$

$$-K(C, T) \frac{\partial T}{\partial n} = h_T (T - T_a) + h_m (C - C_{eq}) L \quad (5)$$

According to the linear viscoelastic theory, in stress-strain analysis the constitutive equation for an isotropic viscoelastic body could be written in the following form (Ferry, 1980),

$$\sigma = \int_0^t D(t - \tau) \frac{\partial \epsilon}{\partial \tau} d\tau \quad (6)$$

where D is the material matrix and is expressed as

$$D = \frac{E(C, T, t)}{1 + \mu} \begin{bmatrix} \frac{1-\mu}{1-2\mu} & \frac{\mu}{1-2\mu} & \frac{\mu}{1-2\mu} & 0 \\ \frac{\mu}{1-2\mu} & \frac{1-\mu}{1-2\mu} & \frac{\mu}{1-2\mu} & 0 \\ \frac{\mu}{1-2\mu} & \frac{\mu}{1-2\mu} & \frac{1-\mu}{1-2\mu} & 0 \\ 0 & 0 & 0 & \frac{1}{2} \end{bmatrix} \quad (7)$$

The relaxation modulus, E , in the material matrix is a function of time, temperature and moisture concentration. However since some food materials tend to display thermo-hydro-rheologically simple behavior (Herum *et al.*, 1979, Mensah *et al.*, 1984, Pappas and Rao, 1989) the drying-induced stress analysis of such materials could be conveniently carried out with the help of a master curve, where the variation of modulus to change in temperature and moisture concentration of the material can be accounted for by the variation in time alone. If the shapes of the relaxation curves for various temperature and moisture concentrations resemble one another, then they could be superimposed on each other by shifting horizontally along the axis utilizing a shift factor a_t (Herum *et al.*, 1979). Using the shift factor a_t and writing in terms of reduced time, which is given as,

$$\xi = \int_0^t \frac{dt}{a_t(C, T)} \quad (8)$$

the constitutive equation takes the form,

$$\sigma = \int_0^\xi D(\xi - \xi_1) \frac{\partial \epsilon}{\partial \xi_1} d\xi_1 \quad (9)$$

This constitutive equation is now reduced to a function of time only and can be conveniently used in the analysis of induced stress. The equilibrium equation and strain displacement equation were utilized in addition to Eq. (9), and an incremental method was used to estimate the transient state, viscoelastic stress-strain field accompanied with material shrinkage (Tsukada *et al.*, 1991, Itaya *et al.*, 1995).

One quarter of the cross section of the material shown in Fig. 1 was divided into 100 four-nodal quadrilateral elements and a finite element method was utilized to analyze the heat transfer, mass transfer and internal stress.

2. Sample Preparation

Because a food material is composed of various components, a detailed study of its physical and mechanical properties is very difficult. Further, the problem is complicated by anisotropic properties exhibited by most food materials. In view of this, starch and their gels are used to model food materials (Saravacous and Raouzeos, 1984), as materials of uniform composition and of various geometrical shapes could be readily prepared for use as samples to carry out the required experiments and quantitatively verify the numerical model. In the present work, potato starch gel was selected as the model food material. The gel samples for measurement of physical properties and

Fig. 2 Relaxation modulus of potato starch gel at 20°C and 50°C

the drying experiment were prepared in the following way. Potato starch supplied by Wako Co. Ltd. and distilled water were used as materials for preparing the gel. Starch and distilled water were mixed in various proportions and heated at 60°C while stirring. When the starch sol had become sufficiently viscous to hinder the sedimentation of starch suspension, it was poured into teflon molds and heated at 80°C in an air tight vessel for three hours. After gelation, the molds were cooled to room temperature and cylindrical gel samples were obtained by carefully removing them from the molds. To measure the viscoelastic property of the gels with less than 1.5 moisture content (dry basis), gels of 1.5 moisture content were first prepared, then dried slowly and placed in an air tight vessel to obtain uniformity of moisture distribution. The initial and equilibrium moisture content of all samples were determined by vacuum drying them at the end of each experiment in an oven at 70°C for 24 hours.

3. Measurement of Physical Properties

3.1 Moisture diffusivity

The method of slopes (Karathanos *et al.*, 1990) can be used to evaluate diffusivity of moisture in a material. To determine the diffusivity of moisture, a thin slab of gel sample 35 mm in diameter and 5 mm in thickness and with an initial moisture content of 1.67 (dry basis) was hung from an electronic balance into a drying chamber in which the temperature and humidity of the air was controlled at the desired values. As the sample was exposed to turbulent flow of air, the resistance of the gas phase to the moisture transfer could be neglected. The change in weight of the sample with time was automatically recorded by a computer. The effective diffusivity calculated using the drying data obtained for the range of 30°C to 50°C is expressed by the following equation and is applicable in

Fig. 3 Shifting of curves

the moisture range of 0.18 to 1.67 (dry basis),

$$D = 156 \times 10^{-10} \left(1 + 0.123 \log \left(0.65 - \frac{W}{1+W} \right) \right) \exp \left(\frac{-1300}{T+273} \right) \quad (10)$$

3.2 Thermal conductivity

A heated probe (Model Kemtherm QTM-D3, Kyoto Denshi Kougyou Co.) operating via the transient hot-wire method was used to measure the effective thermal conductivity of the potato starch gel. The experiment was carried out in the temperature range of 20°C to 50°C on gel samples with moisture content ranging from 1.5 to 4.0. The obtained data were fitted with a series model and consequently, the following equation was obtained,

$$\frac{1}{K} = \frac{1.0 - V}{0.31 + 7.0 \times 10^{-4} (T + 273)} + \frac{V}{K_w} \quad (11)$$

3.3 Viscoelastic property

An instron Universal Testing Machine with a chamber maintained at a constant temperature (Shimadzu DCS-5000) was used to measure the viscoelasticity of potato starch gel. For the experiment, cylindrical samples of 10 mm in diameter and 10 mm in length were used, and silicon oil was smeared on the surface to prevent the evaporation of moisture. The experiments were carried out in the range of 20°C to 50°C. The sample was uniaxially compressed at a speed of 10 mm/min, to apply a strain of 0.03, and relaxation of stress at constant strain was recorded as a function of time over a period of one hour. Typical plots of the curves are shown in **Fig. 2**.

Unfortunately, the relaxation curves of the gel of potato starch shown in **Fig. 2** could not be superimposed on each other by the use of only time shift factor, a_t , and therefore they were shifted vertically as well as horizontally (Yamaguchi *et al.*, 1981) along the axes as described in **Fig. 3**. When the curve is shifted vertically, the relaxation modulus can be written in the following form.

$$E'(t) = E(t) / a_s \quad (12)$$

Substituting this in the constitutive equation gives,

$$\sigma = a_s \int_0^\xi D'(\xi - \xi_1) \frac{\partial \xi}{\partial \xi_1} d\xi_1 \quad (13)$$

The horizontal shift factor, vertical shift factor and the master curve for **Fig. 2** are expressed as,

Fig. 4 Master curve of potato starch gel

Fig. 5 Failure stress of potato starch gel

$$a_t = \exp(14.39[\{W/(W+1)\} - 0.7]) \quad (14)$$

$$a_s = \{2.96 \times 10^7 - 3.1 \times 10^5(T - 20)\} \exp(17.0 \times [0.15 - \{W/(W+1)\}]) \quad (15)$$

$$E'(\xi) = 29.4 \exp(-50\xi) + 20.6 \exp(-5\xi) + 12.7 \exp(-0.5\xi) + 20.6 \exp(-0.05\xi) \quad (16)$$

and the master curve is shown in Fig. 4.

3.4 Failure stress

To measure failure stress the samples were compressed till breakage by applying uniaxial compression at a speed of 10 mm/min. The stress at breakage was recorded against temperature and the moisture content of the gel. During the test, most of the gel samples broke from the top edge to the bottom edge at an angle of about 45°. Figure 5 shows the failure stress data obtained as a function of temperature and moisture.

3.5 Shrinkage coefficient

To measure shrinkage, a strip of starch gel of 1.5 moisture content (dry basis) was prepared. The sample was hung in a constant temperature air chamber. To facilitate slow drying of the sample, high humidity was maintained in the chamber by placing a beaker containing 5% sodium carbonate solution. The shrinkage coefficient of the gel was determined by measuring the dimensions of the sample and

Fig. 6 Schematic diagram of drier used in drying experiment

Table 1 Physical properties and drying conditions used in simulation

Initial temperature (T_0)	20°C
Initial moisture content, dry basis (W_0)	1.67
Relative humidity of air	0.6 for 30°C 0.5 for 50°C
Equilibrium moisture content (W_{eq})	0.239 for 30°C 0.187 for 50°C
Surface mass transfer coefficient (h_m)*	6.40×10^{-2} m/s for 30°C 6.58×10^{-2} m/s for 50°C
Surface heat transfer coefficient (h_r)*	69.4 W/(m ² .K) for 30°C 73.6 W/(m ² .K) for 50°C
Specific heat of gel (c_p)	2500 J/(kg.K)
Density of solid in gel (ρ_s)	1420 kg/m ³
Poisson's ratio (μ)	0.35

* h_m and h_r were calculated using Ranz and Marshall equation

recording it against the change in weight due to the loss of moisture. The obtained data showed that the gel shrinks almost by the same volume as the volume of water evaporated from it.

4. Drying Experiment

For the drying experiment, a chamber was constructed as shown in Fig. 6. The cross section of the duct was 0.1×0.1 m². The temperature and humidity in the chamber were maintained constant throughout the run for each experiment. The air was blown into the duct by a blower through a valve which was installed to control the velocity of air in the duct. A portion of the duct was made of rectangular plates (10 mm thick) of acrylate resin to facilitate photographing of the sample. The rest of the portions of the duct were made out of PVC pipes. The sample was placed on a tray made of wire mesh. The tray was hung on an electronic balance and the loss of weight was recorded against time using a computer. The shrinkage of the sample was determined by photographing the sample. A marker was placed on the side of the sample in order to assess shrinkage. The cylindrical samples used in the experiments were 1 cm in height and 1 cm in diameter with an initial moisture content of 1.67. The drying experiments were carried out at 30°C and 50°C, and at an air velocity of 3

Fig. 7 Moisture contours when dried at 50°C (a) After 15 minutes (b) After 113 minutes

Fig. 8 Principal stresses when dried at 50°C (a) After 15 minutes (b) After 113 minutes

Fig. 9 Hoop stresses when dried at 50°C (a) After 15 minutes (b) After 113 minutes

m/s. For experimental runs at 3 m/s the tray supporting the sample swayed to and fro due to turbulence of the air causing the reading in the electronic balance to fluctuate. Hence, before taking the readings, the blower was stopped for about 40 seconds till the balance showed a steady value.

5. Results and Discussion

To simulate numerically drying of the potato starch gel, the measured values of physical and transport properties were used. The physical properties and processing conditions, except for the values described in section 3, are listed in Table 1. Figures 7 to 9 show simulated values of shrinkage, moisture content contours and the stresses of the right upper half portion of the cross section of the material when dried at 50°C and at an air velocity of 3 m/s. The dashed line and bold broken line in Fig. 7 respectively show the initial shape and shrinkage obtained experimentally. It

Fig. 10 Experimental and simulated values of weight loss

Fig. 11 Variation of induced hoop stresses with time when dried at 30°C and 50°C

is seen from Fig. 8 and Fig. 9 that the maximum stress occurs near the surface of the material, which at the beginning is tensile stress (the single line represents tensile stress, while the double line represents compressive stress) and during the course of drying changes to compressive stress; that is stress reversal takes place. The simulated compressive stress values at the surface did not exceed the value of the failure stress of the potato starch gel shown in Fig. 5. However, during the drying experiment wrinkles appeared on the curved surface of the cylindrical sample, which is an indication of high compressive stress acting on the surface of the gel. It is also seen in the drying simulations that the surface of the material at first became convex and later changed to be concave. Similar observations were made during the drying experiment as shown in Fig. 7. This is because in the early stage of drying, the regions near the circular edges of the cylinder, where the loss of moisture is higher than the rest of the region due to larger surface area to volume ratio, shrink faster and thus the surface of the material takes the convex form. Similar results were obtained when the simulations were carried out at 30°C.

Figure 10 shows the plot of simulated and experimental values of weight loss against time. The calculated results are in good agreement with the experimental results. Figure 11 shows the simulated values of hoop

Fig. 12 Comparison of induced hoop stresses for elastic and viscoelastic material near the surface

stress at the center of the element, with the initial position at 0.995 of the radius in the radial direction and 0.125 of the radius in the vertical direction, when dried at 30°C and at 50°C.

To compare the effect of viscoelasticity on the values of the internal stresses, the program was rerun assuming the gel to be an elastic body. The obtained results show that the values of the stresses differed considerably (**Fig. 12**), which indicates that the effect of the relaxation behavior should be included in evaluation of internal stress in food material.

Conclusion

All the required properties to simulate the drying of potato starch gel were measured. A numerical model was constructed to carry out the drying simulation and it was verified by experiment. During the simulation, maximum stress developed near the surface of the material. This maximum value of the stress did not exceed the breakage stress of the material and the surface wrinkled without cracking. The simulated values of loss of weight and deformation of the gel sample were in close agreement with the experimental results. Further, it was found that since food is a viscoelastic material the viscoelastic behavior should be considered when evaluating the values of the induced stresses.

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Nomenclature

a_s	= vertical shift factor	[-]
a_t	= horizontal shift factor	[-]
C	= moisture concentration	[kg/m ³]
C_{eq}	= equilibrium moisture concentration	[kg/m ³]
C_0	= initial moisture concentration	[kg/m ³]
c_p	= specific heat	[J/kg.K]
D	= moisture diffusion coefficient	[m ² /s]
D	= material matrix	[Pa]
E	= relaxation modulus	[Pa]
E'	= relaxation modulus of curve defined by Eq. 12	[Pa]

h_m	= mass transfer coefficient	[m/s]
h_T	= heat transfer coefficient	[W/m ² .K]
K	= thermal conductivity	[W/m.K]
K_w	= thermal conductivity of water	[W/m.K]
L	= latent heat of evaporation	[J/kg]
n	= distance normal to the surface	[m]
M	= moisture ratio (W/W_0)	[-]
r	= radial distance in cylindrical coordinates	[m]
r_0	= initial radius of sample	[m]
t	= time	[s]
T	= temperature	[°C]
T_0	= initial temperature	[°C]
T_a	= ambient temperature	[°C]
V	= volume fraction of water	[-]
W	= moisture content (dry basis)	[-]
W_0	= initial moisture content (dry basis)	[-]
W_{eq}	= equilibrium moisture content (dry basis)	[-]
z	= axial distance in cylindrical coordinates	[m]
ϵ	= elastic strain	[-]
μ	= Poisson's ratio	[-]
σ	= stress	[Pa]
σ_{max}	= failure stress	[Pa]
ρ	= density	[kg/m ³]
ρ_s	= density of solid in gel	[kg/m ³]
τ	= time of application of load	[s]
ξ	= reduced time of t	[s]
ξ_1	= reduced time of τ	[s]

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