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Three-dimensional heat and moisture transfer with viscoelastic strain–stress formation in composite food during drying

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Abstract—A simulation model was developed for three-dimensional heat and moisture transfer and viscoelastic hygrostress formation in a composite body undergoing drying. In the model the chemical potential of moisture in the body was used as a mass transfer potential since different materials had different affinities to moisture. The governing equations were solved numerically by a finite element method. The model was validated experimentally through drying experiments using triply layered brick shaped samples made of the hydrates of starch granules and of 3 : 1 mixture of starch granules and sucrose.

INTRODUCTION

Most foods undergo volumetric changes during the drying process. These changes are accompanied with internal strain–stress formation, resulting in inferior product quality or product loss due to stress–crack formation, without careful process control. Therefore, several researchers have developed methods for simulating heat and moisture transfer and hygrostrain–stress formation in food undergoing drying to assist process optimization and control.

Gustafson *et al.* [1] analyzed heat transfer and elastic stresses in a corn kernel during heating and cooling by a finite element method. Misra and Young [2] simulated moisture diffusion and the elastic shrinkage in spherically approximated soybeans during drying. Tsukada *et al.* [3] developed a computerized method to estimate simultaneous heat and moisture transfer and strain–stress formation in elastoplastic food during drying.

Since many foods are viscoelastic, several researchers determined their viscoelastic properties [4–7]. Hammerle [8] derived an equation to analyze stresses in a viscoelastic slab for known temperature and moisture distributions. Rao *et al.* [9, 10] analyzed the stresses in a viscoelastic cylinder or sphere for assumed moisture and temperature distributions. Litchfield and Okos [11] used Rao *et al.*'s [9] spherical stress equations together with heat and moisture transfer equations to estimate the hygrophysical changes of viscoelastic corn kernels during drying, tempering

and cooling processes. Haghghi and Segerlind [12, 13] developed a method for simulating strain–stress formation in a viscoelastic, homogeneous biological material and applied it to analyze thermo-hygrostress formation in an elastic spherical food undergoing drying. Irudayaraj and Haghghi [14] developed a simulation model for heat and moisture and thermo-hygro viscoelastic stress formation in an axisymmetric, heterogeneous body of an arbitrary cross-sectional contour (a two-dimensional problem). They assumed implicitly homogeneous hydrochemical affinities for all heterogeneous components since moisture concentration was used as a moisture transfer potential although heterogeneous viscoelastic properties were assumed. Irudayaraj *et al.* [15] applied the developed method to analyze drying processes of soybeans and corn kernels.

Many foods are irregular and nonhomogeneous. Within such foods, heat and moisture transfer and strain–stress formation are three-dimensional. Furthermore, one needs to consider the interactive influence of nonhomogeneity on heat and moisture transfer as well as on strain–stress formation since different components have different affinities to water. However, virtually all published papers are for homogeneous and one- or two-dimensional transport processes.

The objective of this work is to develop a method for predicting three-dimensional heat and moisture transfer with viscoelastic strain–stress formation in a composite food during drying.

THEORY

Heat and moisture transfer

Simultaneous, transient state heat and moisture transfer in food is simulated using a modified Luikov's

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NOMENCLATURE

| | | | |
|--------------------------------------|---|--------------------------------------|--|
| A | displacement-strain matrix defined by equation (32) [m^{-1}] | H | high amylose starch granules hydrate |
| A^d | dimensionless matrix corresponding to A | H_M | mass transfer Biot number defined by equation (43) |
| A_{MM} | matrix defined by equation (A1) | H_T | heat transfer Biot number defined by equation (43) |
| A_{MT} | matrix defined by equation (A2) | HSH | triply layered composite brick shaped sample with one layer of S between two layers of H |
| A_{TM} | matrix defined by equation (A3) | ΔH° | enthalpy of saturated liquid water [J mol^{-1}] |
| A_{TT} | matrix defined by equation (A4) | ΔH_{r_j} | reaction heat of <i>j</i> th chemical reaction [J kg^{-1}] |
| a_M | moisture shift factor | $\Delta H_{r_j}^d$ | dimensionless reaction heat defined by equation (43) |
| a_T | temperature shift factor | ΔH_v | latent heat of moisture vaporization or condensation [J kg^{-1}] |
| a_w | water activity | ΔH_v^d | dimensionless latent heat defined by equation (43) |
| B^d | strain-nodal displacement matrix defined by equation (56) | h_m | mass transfer coefficient [$\text{kg m}^{-2} \text{Pa}^{-1} \text{s}^{-1}$] |
| B_{MM} | matrix defined by equation (A4) | h_t | heat transfer coefficient [$\text{W m}^{-2} \text{K}^{-1}$] |
| B_{MT} | matrix defined by equation (A5) | J_h | heat flux [W m^2] |
| B_{TM} | matrix defined by equation (A7) | J_m | moisture flux [$\text{kg m}^{-2} \text{s}^{-1}$] |
| B_{TT} | matrix defined by equation (A8) | K | bulk modulus [Pa] |
| C | volumetric moisture concentration [kg m^{-3}] | k_c | anisotropic Dufour thermal conductivity [$\text{W m}^2 \text{kg}^{-1}$] |
| C | matrix defined by equation (30) | K^d | stiffness matrix defined by equation (57) |
| C* | vector defined by equation (30) | K_{MM}} | matrix defined by equation (42) |
| C_m | $(\partial W/\partial \phi)_T$, specific mass capacity [$\text{g water (g dry matter)}^{-1} \text{M}^{-1}$] | K_{MT}} | matrix defined by equation (42) |
| C_{MM} | coefficient defined by equation (41) | K_{TM}} | matrix defined by equation (42) |
| C_{MT} | coefficient defined by equation (41) | K_{TT} | matrix defined by equation (42) |
| c_p | specific heat [$\text{kJ kg}^{-1} \text{K}^{-1}$] | k_p | anisotropic filtration thermal conductivity [$\text{W m}^{-1} \text{kg}^{-1}$] |
| C_T | $(\partial W/\partial T)_\phi$, temperature coefficient [$\text{g water (g dry matter)}^{-1} \text{K}^{-1}$] | k_t | anisotropic thermal conductivity [$\text{W m}^{-1} \text{K}^{-1}$] |
| C_{TM} | coefficient defined by equation (41) | Le | Luikov number defined by equation (43) |
| C_{TT} | coefficient defined by equation (41) | l | characteristic dimension [m] |
| D_p | anisotropic pressure mass diffusivity [s] | N | shape function matrix |
| D_t | anisotropic Soret mass diffusivity [$\text{kg m}^{-1} \text{s}^{-1} \text{K}^{-1}$] | n | outward normal unit vector |
| D_w | anisotropic moisture diffusivity [$\text{m}^2 \text{s}^{-1}$] | n_e | number of finite elements used to map food volume |
| d | nodal displacement vector | n_s | power for converting S_v to stress free strain element [—] |
| E | constant in Gugenheim-Anderson-deBore isotherm formula | p | water vapor pressure equilibrated to exposed food surface moisture and temperature, $a_w p_s$ [Pa] |
| F_i | components of body force vector [Pa m^{-1}] | p_a | water vapor pressure of air [Pa] |
| F₀ | $\alpha_r t/l^2$, reference Fourier number | p_s | saturation water vapor pressure [Pa] |
| F_M | moisture flux vector defined by equation (A9) | Q | constant in Gugenheim-Anderson-deBore isotherm formula |
| F_T | heat flux vector defined by equation (A10) | R_j | reaction rate [$\text{kg s}^{-1} \text{m}^{-3}$] |
| G | shear modulus [Pa] | R^d | force vector defined by equation (59) |
| G₁(t) | shear modulus relaxation function [Pa] | R | gas constant [$\text{J} \cdot \text{K}^{-1} \text{mol}^{-1}$] |
| G_{1d}(F₀) | function obtained replacing <i>t</i> in $G_1(t)$ by $F_0 l^2/\alpha_r$ [Pa] | S | surface variable or hydrate of high amylose starch granules-sucrose 3 : 1 mixture |
| G₁^r | reference shear modulus relaxation value normally $G_1(0)$ | S_a | surface exposed to atmosphere |
| G₂(t) | bulk modulus relaxation function [Pa] | | |
| G_{2d}(F₀) | function obtained replacing <i>t</i> in $G_2(t)$ by $F_0 l^2/\alpha$ [Pa] | | |
| G_{ijkl} | elements of material properties tensor defined by equation (22) [Pa] | | |
| ΔG° | Gibbs' free energy of saturated, liquid water [J mol^{-1}] | | |

\mathbf{D}_i and \mathbf{D}_p are anisotropic mass diffusivity, Soret mass diffusivity and pressure mass diffusivity, respectively.

Luikov [17] treated pressure p as a dependent variable together with T and C , resulting in three simultaneous equations for the simulation. However, variable p was eliminated in the above model since it is related to local moisture vaporization or condensation shrinkage and temperature, equation (5)

$$\begin{aligned} \text{grad}(P) &= (\gamma_1 \varepsilon_L - \gamma_2 \partial S_v / \partial C) \text{grad}(C) + \gamma_3 \text{grad}(T) \\ &= (\gamma_1 \varepsilon_L - \gamma_{2v}) \text{grad}(C) + \gamma_3 \text{grad}(T). \end{aligned} \quad (5)$$

The initial and boundary conditions are:

I.C.

$$C = C_0, T = T_0 \text{ when } t = 0 \text{ and } \mathbf{x} \in S_a \ \& \ V \quad (6)$$

B.C. for surface of material

$$\begin{aligned} h_t(T_a - T) + \beta \phi_t(T_a^4 - T^4) \\ - h_m(1 - \varepsilon) \Delta H_v(p - p_a) = \mathbf{J}_h \cdot \mathbf{n} \end{aligned} \quad (7)$$

$$\begin{aligned} h_m(p - p_a) = \mathbf{J}_m \cdot \mathbf{n} \\ \text{where } t > 0 \text{ and } \mathbf{x} \in S_a. \end{aligned} \quad (8)$$

The volumetric concentration of moisture, C , is used in the governing equations given above. Therefore, they are not applicable to nonhomogeneous food, whose components have different chemical affinities to water. Luikov [17] used a mass transfer potential derived from normalized moisture sorption isotherms of filter paper. Since this potential did not properly account for the influence of temperature on the mass transfer potential, the chemical potential of water in each component was used as a mass transfer potential ϕ [18]:

$$\phi = \begin{cases} \mu_0 + RT \ln(a_w) & \text{for } W < W_m \\ (W - W_m)/(W_m - W_z) \mu_0 + \mu_0 & \text{for } W_m < W \end{cases} \quad (9)$$

where μ_0 is the chemical potential of saturated liquid water, equation (10):

$$\mu_0 = \Delta G^\circ = \Delta H^\circ - T \Delta S^\circ. \quad (10)$$

Water activity, a_w , may be related to dried mass based moisture concentration W using any reliable moisture sorption isotherm formula (equilibrium assumed between adsorbed moisture in food and vapor in pores within a minute volumetric element). For the present work, Guggenheim–Anderson–deBor (GAB) formula was used since this estimated accurately the isotherms of many dried foods [19]

$$W = W_n E Q a_w / \{ (1 - Q a_w) (1 - Q a_w + E Q a_w) \}. \quad (11)$$

Equations (9)–(11) show W dependent on ϕ and T . Therefore, one has

$$\begin{aligned} dW &= (\partial W / \partial \phi)_T d\phi + (\partial W / \partial T)_\phi dT \\ &= C_m d\phi + C_T dT. \end{aligned} \quad (12)$$

W is related to volumetric concentration C

$$W = C / \rho_s = C S_v / \rho_{s0}. \quad (13)$$

Substituting equations (9) and (11)–(13) into equations (1)–(5), the governing equations were transformed as follows

$$\begin{aligned} \{ C_p \rho_s - \Delta H_v \varepsilon_L (\rho_s + \partial \rho_s / \partial W \cdot W) C_T \} \partial T / \partial t \\ - \Delta H_v \varepsilon_L (\rho_s + \partial \rho_s / \partial W \cdot W) C_m \partial \phi / \partial t \\ = -\text{div}(\mathbf{J}_h) + \Sigma \Delta H_T R_j \end{aligned} \quad (14)$$

$$\begin{aligned} (\rho_s + W \cdot \partial \rho_s / \partial W) (C_m \partial \phi / \partial t + C_T \partial T / \partial t) \\ = -\text{div}(\mathbf{J}_m) + \Sigma R_j \end{aligned} \quad (15)$$

$$\begin{aligned} \mathbf{J}_h &= - \left[\mathbf{k}_t + (-1)^n \gamma_3 \mathbf{k}_p + \{ (-1)^m \mathbf{k}_c + (-1)^n \mathbf{k}_p \right. \\ &\quad \times (\gamma_1 \varepsilon_L - \gamma'_2) \} \left(\rho_s + \frac{\partial \rho_s}{\partial W} W \right) C_T \cdot \left. \right] \text{grad}(T) \\ &\quad - \left[\{ (-1)^m \mathbf{k}_c + (-1)^n \mathbf{k}_p (\gamma_1 \varepsilon_L - \gamma'_2) \} \right. \\ &\quad \times \left(\rho_s + \frac{\partial \rho_s}{\partial W} W \right) C_m \cdot \left. \right] \text{grad}(\phi) \end{aligned} \quad (16)$$

$$\begin{aligned} \mathbf{J}_m &= - \left[\rho_s + \frac{\partial \rho_s}{\partial W} W \{ D_w + (\gamma_1 \varepsilon_L - \gamma'_2) \mathbf{D}_p \} C_T \right. \\ &\quad + \left(\mathbf{D}_1 + \mathbf{D}_p \gamma_3 \right) \cdot \text{grad}(T) - \left[\left(\rho_s + \frac{\partial \rho_s}{\partial W} W \right) \right. \\ &\quad \times \left. \{ \mathbf{D}_w + (\gamma_1 \varepsilon_L - \gamma'_2) \mathbf{D}_p \} C_m \right] \cdot \text{grad}(\phi). \end{aligned} \quad (17)$$

The initial and boundary conditions, equations (6)–(8), are transformed through similar substitution.

Conditions on the interface between two different components α and β are

$$\left. \begin{aligned} T_\alpha &= T_\beta \\ \mathbf{J}_{k\alpha} \cdot \mathbf{n}_\alpha &= \mathbf{J}_{k\beta} \cdot \mathbf{n}_\beta \end{aligned} \right\} \text{when } t > 0 \text{ and } \mathbf{x} \in S_i \quad (18)$$

$$\left. \begin{aligned} \phi_\alpha &= \phi_\beta \\ \mathbf{J}_{m\alpha} \cdot \mathbf{n}_\alpha &= \mathbf{J}_{m\beta} \cdot \mathbf{n}_\beta \end{aligned} \right\} \text{when } t > 0 \text{ and } \mathbf{x} \in S_i. \quad (19)$$

Viscoelastic strain–stress

Linear viscoelasticity is assumed since it has been used successfully simulating strain–stress formation in many biomaterials [20]. The following are equations expressed in the three-dimensional Cartesian coordinates.

Constitutive equations:

$$\sigma_{ij} = \int_0^t G_{ijk} \{ \lambda(t) - \lambda(\xi) \} \{ \partial [\varepsilon_{kr}(\xi) - \varepsilon_{kr}^e(\xi)] / \partial \xi \} d\xi. \quad (20)$$

In the above, ξ is a dummy integration variable and t is any time value representing a current drying time.

Using a convolution expression, equation (20) is rewritten as

$$\sigma_{ij} = G_{ijkl} * (\epsilon_{kl} - \epsilon_{kl}^s) \tag{21}$$

where

$$G_{ijkl}(t) = (1/3)[G_2(t) - G_1(t)]\delta_{ij}\delta_{kl} + (1/2)G_1(t)[\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}] \tag{22}$$

$$G(t) = G_1(t)/2, K(t) = G_2(t)/3 \tag{23}$$

G and K are shear and bulk moduli, respectively. λ is the reduced time to account for temperature and moisture dependency of stress by extending or reducing the effective time as defined below [12]

$$\lambda(t) = t/\{a_T(T)a_M(w)\} \tag{24}$$

where a_T and a_M are temperature and moisture shift factors, respectively.

Equilibrium equation :

$$\sigma_{ijj} + F_i = 0 \tag{25}$$

where σ_{ijj} denotes differentiation of σ_{ij} with respect to j and F_i is a body force vector element

$$\epsilon_{ij} = (1/2)(U_{ij} + U_{ji}). \tag{26}$$

Virtual work. The above given equations describe strain–stress behavior at any one location. To solve these equations, an integrated expression applicable to an entire body is required. This is a functional Γ , virtual work of a deforming body. When there is no body force applied, one has

$$\Gamma = \frac{1}{2} \int_v [G_{ijkl} * \epsilon_{ij} * \epsilon_{kl} - G_{ijkl} * \epsilon_{ij}^s * \epsilon_{kl}] dv. \tag{27}$$

A governing equation is obtained by minimizing functional $\Gamma(\delta\Gamma = 0)$.

For an isotropic body, equation (27) reduces to as follows with the vectorial representation of strain tensor ϵ

$$\Gamma = \frac{1}{2} \int_v [C_{ij} * \epsilon_i * \epsilon_j - C_i^s * \epsilon_i^s * \epsilon_i] dv \tag{28}$$

$$\epsilon = \mathbf{A}\mathbf{U} \tag{29}$$

where

$\mathbf{C} =$

$$\begin{bmatrix} K+4G/3 & K-2G/3 & K-2G/3 & 0 & 0 & 0 \\ K-2G/3 & K+4G/3 & K-2G/3 & 0 & 0 & 0 \\ K-2G/3 & K-2G/3 & K+4G/3 & 0 & 0 & 0 \\ 0 & 0 & 0 & G & 0 & 0 \\ 0 & 0 & 0 & 0 & G & 0 \\ 0 & 0 & 0 & 0 & 0 & G \end{bmatrix}$$

$$\mathbf{C}^s = \begin{bmatrix} 3K \\ 3K \\ 3K \\ G \\ G \\ G \end{bmatrix}. \tag{30}$$

$$\epsilon = \begin{bmatrix} \epsilon_x \\ \epsilon_y \\ \epsilon_z \\ \epsilon_{xy} \\ \epsilon_{yz} \\ \epsilon_{zx} \end{bmatrix} \quad \epsilon^s = \begin{bmatrix} \epsilon^s \\ \epsilon^s \\ \epsilon^s \\ 0 \\ 0 \\ 0 \end{bmatrix} \quad \epsilon_0^s = S_v^0 \text{ for } i = 1, 2 \text{ and } 3 \tag{31}$$

$$\mathbf{A} = \begin{bmatrix} \partial/\partial x & 0 & 0 \\ 0 & \partial/\partial y & 0 \\ 0 & 0 & \partial/\partial z \\ (1/2)\partial/\partial y & (1/2)\partial/\partial x & 0 \\ 0 & (1/2)\partial/\partial z & (1/2)\partial/\partial y \\ (1/2)\partial/\partial z & 0 & (1/2)\partial/\partial x \end{bmatrix}$$

$$\mathbf{U} = \begin{bmatrix} U_x \\ U_y \\ U_z \end{bmatrix} \tag{32}$$

In equation (31) ϵ^s is the initial strain and S_v is a volumetric shrinkage function determined through a stress-free drying experiment (uniform moisture distribution in a sample body throughout drying). For the present study, negligible thermal strains were assumed because of negligible thermal expansion of most food.

When the vectorially expressed strain tensor ϵ is used, the constitutive equations (21) and (22) become as follows

$$\sigma_i = G_{ik} * (\epsilon_k - \epsilon_k^s) \tag{33}$$

where

$$G_{ik} = C_{ik}. \tag{34}$$

Dimensionless model

The following dimensionless equations are obtained through the dimensional analysis of the above heat and moisture transfer and viscoelastic strain–stress equations.

Heat and moisture transfer :

$$C_{TT} \frac{\partial \theta}{\partial Fo} = C_{TM} \frac{\partial \Phi}{\partial Fo} + \text{div}(\mathbf{J}_{TT}) + \text{div}(\mathbf{J}_{TM}) + \sum_j \Delta H_{Tj}^d R_j^d \tag{35}$$

$$c_{MM} \frac{\partial \Phi}{\partial Fo} = -C_{MT} \frac{\partial \theta}{\partial Fo} + \text{div}(\mathbf{J}_{MT}) + \text{div}(\mathbf{J}_{MM}) + \sum_j R_j^d \tag{36}$$

$$\begin{aligned} \mathbf{J}_{TT} &= \mathbf{K}_{TT} \cdot \text{grad}(\theta), & \mathbf{J}_{TM} &= \mathbf{K}_{TM} \cdot \text{grad}(\Phi) \\ \mathbf{J}_{MT} &= \mathbf{K}_{MT} \cdot \text{grad}(\theta), & \mathbf{J}_{MM} &= \mathbf{K}_{MM} \cdot \text{grad}(\Phi). \end{aligned} \tag{37}$$

Initial condition :

$$\Phi = \Phi_0 \text{ and } \theta = \theta_0, \text{ when } Fo = 0 \text{ and } \mathbf{X} \in S_a \text{ and } V. \tag{38}$$

Boundary conditions :
when $Fo > 0$ and $\mathbf{X} \in S_a$

$$(H_T + \beta^d)(\theta_a - \theta) - (1 - \varepsilon_L)H_M \Delta H_v^d(\psi - \psi_a) = \mathbf{J}_{TT} \cdot \mathbf{n} + \mathbf{J}_{TM} \cdot \mathbf{n}$$

$$H_M(\psi - \psi_a) = \mathbf{J}_{MT} \cdot \mathbf{n} + \mathbf{J}_{MM} \cdot \mathbf{n} \quad (39)$$

when $Fo > 0$ and $\mathbf{X} \in S_i$

$$\theta_x = \theta_\beta, \quad \mathbf{J}_{TT} \cdot \mathbf{n}_x = \mathbf{J}_{TT} \cdot \mathbf{n}_\beta$$

$$\Phi_x = \Phi_\beta, \quad \mathbf{J}_{MM} \cdot \mathbf{n}_x = \mathbf{J}_{MM} \cdot \mathbf{n}_\beta \quad (40)$$

where

$$\left. \begin{aligned} C_{TT} &= (C_p/C_p^r)(\rho_b/\rho_s^r) - \frac{\Delta H_v^d}{Le} \varepsilon_L \rho_s^d C_T (T_a - T^r), \\ C_{TM} &= \frac{\Delta H_v^d}{Le} \varepsilon_L \rho_s^d C_m (\phi_a - \phi^r) \\ C_{MM} &= \rho_s^d C_m (\phi_a - \phi^r) / Le, \quad C_{MT} = \rho_s^d C_T (T_a - T^r) / Le \end{aligned} \right\} \quad (41)$$

$$\left. \begin{aligned} \mathbf{K}_{TT} &= [\{\mathbf{k}_t + (-1)^n \gamma_3 \mathbf{k}_p\} / K_T^r] \\ &+ [\{(-1)^m \mathbf{k}_c + (-1)^n \mathbf{k}_p (\gamma_1 \varepsilon_L - \gamma_2)\} / K_T^r] \rho_s^r \rho_s^d C_T \\ \mathbf{K}_{TM} &= [\{(-1)^m \mathbf{k}_c + (-1)^n \mathbf{k}_p (\gamma_1 \varepsilon_L - \gamma_2)\} / k_T] \rho_s^r \rho_s^d C_m (\phi_a - \phi^r) / (T_a - T^r) \\ \mathbf{K}_{MM} &= [\{\mathbf{D}_w + \mathbf{D}_p (\gamma_1 \varepsilon_L - \gamma_2)\} / D_w^r] \rho_s^d C_m (\phi_a - \phi^r) \\ \mathbf{K}_{MT} &= [\{\mathbf{D}_w + \mathbf{D}_p (\gamma_1 \varepsilon_L - \gamma_2)\} / D_w^r] \rho_s^d C_T (T_a - T^r) \\ &+ [\{(\mathbf{D}_t + \mathbf{D}_p \gamma_3) / D_w^r\} (T_a - T^r)] / \rho_s^d \end{aligned} \right\} \quad (42)$$

$$\left. \begin{aligned} H_T &= h_t l / k_t^r, \quad H_M = l p_a h_m / (D_w^r \rho_s^r), \quad \psi = p / p^r \\ Le &= D_w^r c_p^r \rho_s^r / k_t^r, \quad R_j^d = l^2 R_j / D_w^r \rho_s^r \\ \Delta H_{ij}^d &= D_w^r \rho_s^r \Delta H_{ij} / k_t^r (T_a - T^r) \\ \Delta H_v^d &= D_w^r \rho_s^r \Delta H_v / k_t^r (T_a - T^r) \\ \rho_s^d &= (\rho_s + W \partial \rho_s / \partial W) / \rho_s^r \\ \beta^d &= \frac{\beta \phi^r l (T_a - T^r)^3}{k_t^r} \left\{ \left(1 + \frac{T^r}{T_a - T^r} \right)^2 \right. \\ &\quad \left. + \left(\theta + \frac{T^r}{T_a - T^r} \right)^2 \right\} \left(\theta + \frac{T^r}{T_a - T^r} \right) \end{aligned} \right\} \quad (43)$$

In the above, θ , Φ and Fo are dimensionless temperature, dimensionless mass transfer potential and reference Fourier number, respectively. \mathbf{K}_{TT} , \mathbf{K}_{TM} , \mathbf{K}_{MT} and \mathbf{K}_{MM} are dimensionless matrices of orthorhombic thermal conductivity, Dufour thermal conductivity, mass diffusivity and Soret mass diffusivity, respectively. C_{TT} , C_{TM} , C_{MM} and C_{MT} are dimensionless temperature coefficient, latent heat, specific mass capacity and mass interactive coefficient, respectively.

Stress-strain :

$$\sigma_i^d = \frac{\sigma_i}{G_1^r} = \int_0^{Fo} G_{ik}^d(\lambda^d(Fo)) - \lambda^d(\xi) \frac{\partial}{\partial \xi} [\varepsilon_k^d(\xi) - \varepsilon_k^{sd}(\xi)] d\xi = G_{ik}^d * (\varepsilon_k^d - \varepsilon_k^{sd}) \quad (44)$$

$$G_{ik}^d = G_{ik} / G_1^r \quad (45)$$

$$G^d(Fo) = G(Fo) / G_1^r = G_1(Fo) / (2G_1^r),$$

$$K^d(Fo) = K(Fo) / G_1^r = (1/3)G_2(Fo) / G_1^r \quad (46)$$

$$\lambda^d(Fo) = Fo / \{a_T(\theta) a_M(\phi)\} \quad (47)$$

$$\Gamma^d = \Gamma / \{G_1^r l^3\} = 1/2 \int_{v_i^d} [C_{ij} * \varepsilon_i^d * \varepsilon_j - C_i^{*d} * \varepsilon_i^s * \varepsilon_j] dv^d$$

(48) \mathbf{c}^d and \mathbf{c}^{*d} are obtained by replacing K and G in equation (32) by K^d and G^d , respectively. Although ε is dimensionless, it is transformed to ε^d

$$\varepsilon^d = \mathbf{A}^d \cdot \mathbf{U}^d. \quad (49)$$

\mathbf{A}^d is obtained by replacing x , y and z of \mathbf{A} , equation (32), to corresponding dimensionless location variables and \mathbf{U}^d by replacing its components, equation (32), with those expressed in the dimensionless variables.

Transient state hygroviscoelastic stresses at any drying time may be estimated as follows. One obtains first instantaneous temperature and mass transfer potential distributions. From these distributions, one determines an instantaneous, free shrinkage strain ε^{sd} , distribution, a corresponding hygrostrain, ε^d , distribution solving $\delta \Gamma^d = 0$. By repeating this, one obtains a hygrostrain history at any location in food undergoing drying (coupled solutions of the heat and moisture transfer equations required). A transient state hygrostress, σ^d , history at any location in food is estimated from the strain history using the constitutive equation (45).

NUMERICAL SOLUTION ALGORITHM

The governing equations, both heat and moisture transfer and strain-stress formation, are solved applying Galerkin finite element method. For this application, a solution domain is subdivided into curved side isoparametric elements, 20 nodes per each element, since an irregular shape may be mapped with a small number of these elements [21, 22].

The following equations of nodal mass transfer potentials and temperatures are derived from equations (35)–(40) :

$$\mathbf{A}_{TM} \Phi + \mathbf{A}_{TT} \theta + \mathbf{B}_{TM} \{\partial \Phi / \partial Fo\} + \mathbf{B}_{TT} \{\partial \theta / \partial Fo\} = \mathbf{F}_T \quad (50)$$

$$\mathbf{A}_{MM} \Phi + \mathbf{A}_{MT} \theta + \mathbf{B}_{MM} \{\partial \Phi / \partial Fo\} \mathbf{B}_{MT} \{\partial \theta / \partial Fo\} = \mathbf{F}_M \quad (51)$$

where θ , Φ and F_0 are nodal dimensionless temperature vector, nodal mass transfer potential vector and time, respectively. The coefficient matrices and vectors in equations (50) and (51) are given in the Appendix.

Equations (50) and (51) are nonlinear since all transport properties are dependent on temperature and mass transfer potential. They are solved using a three-time level, stable noniterative method developed by Comini *et al.* [23, 24]. Since this method is not self-starting, Crank-Nicolson method was applied for the first increment (iterative computations required).

Virtual work functional Γ , equation (48), is minimized applying the Galarkin method and summing the resultant expressions over all elements for each time increment :

$$\Gamma^d = (1/2) \sum_{e=1}^{n_e} \int_{v_e} \{ \epsilon_j^d * C_{ij}^d * \epsilon_j^d - \epsilon_i^d * C_i^{sd} * \epsilon_i^{sd} \} dv^d \quad (52)$$

where v_e denotes the volume of an element and n_e is the number of elements used to map the solution domain. The incremental displacements at any location in each element are approximated by

$$\Delta U^d = N \cdot \Delta d^d. \quad (53)$$

Substituting equation (53) into a dimensionless equivalence of equation (29), the incremental strain vector is given by

$$\Delta \epsilon^d = B^d \cdot \Delta d^d \quad (54)$$

where

$$B^d = A^d \cdot N. \quad (55)$$

Substitution of equation (54) into equation (55) yields

$$\Gamma = (1/2)(d^{dT} * K^d * d^d - d^{dT} * R^d) \quad (56)$$

where the stiffness matrix K and the force vector R are

$$K^d = \sum_{e=1}^{n_e} \int_{v_e} B^{dT} C^d B^d dv^d \quad (57)$$

$$R^d = \sum_{e=1}^{n_e} \int_{v_e} B^{dT} C^{sd} * \epsilon^{sd} dv^d. \quad (58)$$

The variation of equation (56) is

$$\delta \Gamma = \delta [d^{dT} * K^d * d^d] - \delta [d^{dT} * R^d]. \quad (59)$$

Setting equation (59) to zero, Γ is minimized :

$$K^d * d^d - R^d = 0. \quad (60)$$

From the definition of the convolution form, equation (60) becomes

$$\int_{\xi=0}^{F_0} K^d (\lambda^d(F_0) - \lambda^d(\xi)) d[d^d(\xi)] = R^d(F_0). \quad (61)$$

Equation (61) is approximated as

$$\sum_{m=0}^{n+1} K^d [\lambda^d(F_{0_{n+1}}) - \lambda^d(F_{0_m})] \Delta d^d(F_{0_m}) = R^d(F_{0_{n+1}}). \quad (62)$$

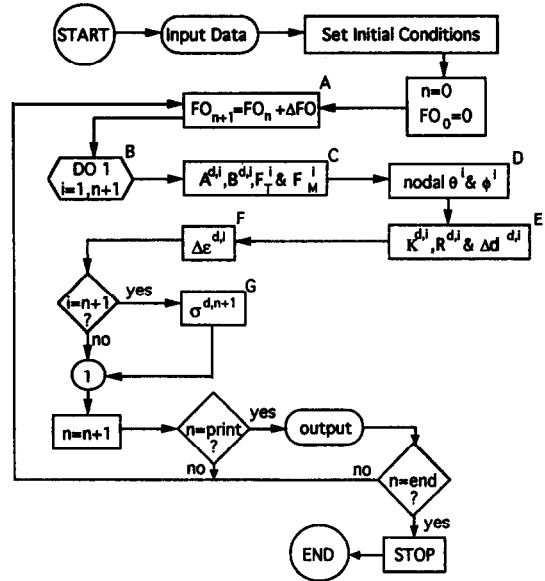


Fig. 1. A bridged flow chart of prepared computer program.

The last increment of displacement is obtained as follows

$$K^d(0) \Delta d^d(F_{0_{n+1}}) = R^d(F_{0_{n+1}}) - \sum_{m=0}^n K^d (\lambda^d(F_{0_{n+1}}) - \lambda^d(F_{0_m})) \Delta d^d(F_{0_m}). \quad (63)$$

The displacement increment at each time step is determined solving equation (63).

The stress is estimated using equation (44) and (54)

$$\sigma^d(F_{0_{n+1}}) = \sum_{m=0}^{n+1} C^d (\lambda(F_{0_{n+1}}) - \lambda(F_{0_m})) B^d \Delta d^d(F_{0_m}) - \sum_{m=0}^{n+1} C^d (\lambda(F_{0_{n+1}}) - \lambda(F_{0_m})) \cdot \Delta \epsilon^{sd}(F_{0_{n+1}}). \quad (64)$$

According to equation (63), the current nodal displacements, Δd , are determined by numerically integrating K^d with respect to nodal displacement from zero time to the last time step. Additionally, the current nodal hygrostresses are estimated numerically integrating $C^d B^d$ with respect to Δd^d , C^d with respect to stress free strains, equation (64). Since K^d , C^d and B^d are dependent on θ and ϕ , directly or indirectly, nodal θ and ϕ histories are required for the integrations.

A computer program was prepared using the numerical solution algorithm, Fig. 1. Initially, θ , ϕ , Δd^d and σ^d were computed simultaneously at each time step (coupled estimation). However, the estimation of θ and ϕ followed by the estimation of Δd^d and σ^d at each time step (partially uncoupled estimation) produced results virtually identical to the coupled computations with significant computer time saving. Therefore, the partially uncoupled computations were employed in the computer program, blocks C and D for estimating nodal θ and ϕ at the

current time and blocks E, F and G for estimating nodal σ^d at the current time. Nodal K^d histories are required to estimate current nodal Δd^d , equation (63), and nodal Δd^d histories are required to estimate current nodal σ^d ; equation (64). Therefore, nodal K^d and Δd^d at each step were stored together with nodal θ and ϕ .

EXPERIMENTAL VALIDATION

Triply layered sample shaped samples were prepared from the hydrates of high amylose starch granules (H) and of 1 : 3 mixture of sucrose and high amylose starch granules (S). The initial, overall dimension of each sample was 19.3 × 15.8 × 14mm (length × width × height). Samples were made arranging differently H and S layers. In one arrangement, one 6 mm thick rectangular S plate (19.3 × 15.8 × 6 mm) was placed between two 4 mm thick rectangular H plates, HSH brick sample. Another had the reversed layer arrangement, SHS brick sample. The initial moisture content and temperature of each sample were 0.2 g water/g dry matter and 25°C, respectively.

A copper-constantan thermocouple made from 36-gauge wires was inserted at the center of a brick of each layer arrangement to monitor a temperature history during drying.

The prepared samples were dried in a 50°C forced air dryer (C. G. Sargent's Sons Corp., Graniteville, MA). Those without the installed thermocouples were taken out periodically from the drier to determine gravimetrically changes in the overall average moisture concentration of each sample (the dry matter of each sample determined by a vacuum oven method [25] upon completing the drying experiment). Additionally, different samples were subjected to different drying processes at the same conditions and taken out to determine the average concentration of the S or H layer after separating it carefully. Other samples were taken out from the drier at 0.2 and 1.0 h of drying. Each sample was bisected across the layer thickness at the midpoint of 14.3 mm (initial size) edge to take pictures of cracks on the bisected surface.

Table 1 shows the drying conditions and physical property values of the samples. The listed values were obtained from Furuta *et al.* [26], Sakai and Hayakawa [18] and Tsukada *et al.* [3]. The values of h_m and h_i are dependent on the sample shape and size and drying conditions. Therefore, they were determined from a separate set of experimental temperature and moisture histories through an optimization method.

Figure 2(a) and (b) shows experimental and theoretical temperature and moisture histories of HSH and SHS samples, respectively. The experimental histories agree well with the respective theoretical histories, especially when one considers difficulty in determining the moisture history of each layer of the composite brick samples. The figures show significant influence of the layer arrangement on the moisture histories, much smaller concentration differences between the

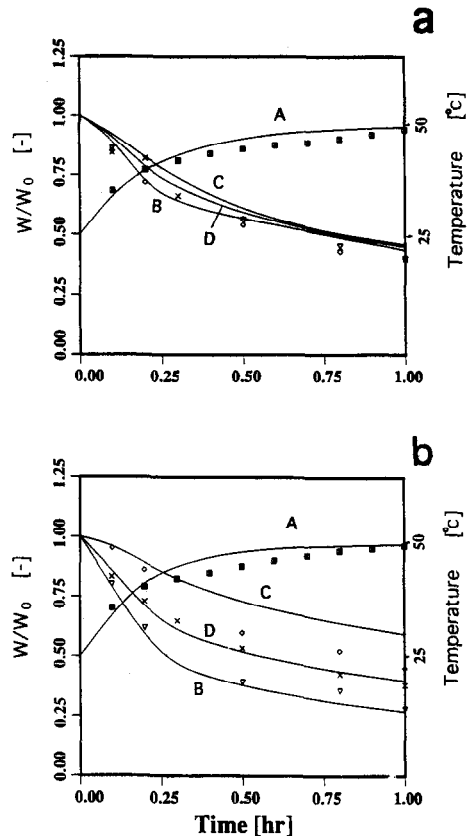


Fig. 2. Moisture concentration and central temperature histories of HSH and SHS brick samples. (a), HSH brick; (b), SHS brick; solid lines are predicted histories and plotted symbols are experimental data. (A) Central temperature; (B) average moisture concentration of outside layers; (C) average concentration of middle layer; (D) overall average concentration solid; \blacksquare , central temperature; \diamond , average concentration of outside layers; ∇ , average concentration of middle layer; X, overall average concentration.

outside (H) and middle (S) layers of the HSH sample compared to those for the SHS sample. This is due to interactive influence of a moisture flux (D-flux) due to drying and a flux (I-flux) across the H and S interface due to ϕ difference between H and S.

Figure 3 shows the moisture sorption isotherms of

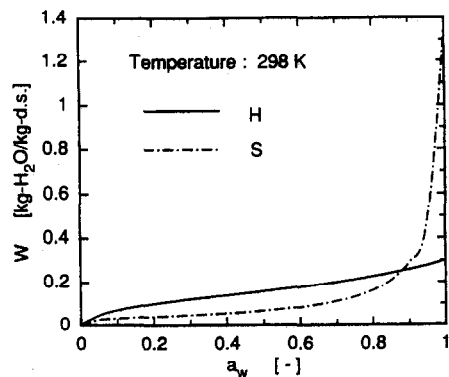


Fig. 3. Moisture sorption isotherms of high amylose starch granules (H) and of H and sucrose 3 : 1 mixture at 25°C.

H and S at 25°C, the initial sample temperature (similar relationship of both isotherms at higher temperatures). It is clear from this figure and equation (9), ϕ of S (ϕ_s) is greater than that of H (ϕ_h) at the zero drying time. Therefore, with the HSH sample, the middle layer (S) lost moisture due to the I-flux flowing out to the outside layer (H) across the interface in addition to the D-flux flowing out directly to the air through its exposed, side surface. This resulted in a large moisture gain by the outside layers from the middle layer, causing a small moisture concentration difference between the middle and outside layers, Fig. 2(a).

With the SHS sample, the initial direction of the I-flux was from the outside layer (S) to the middle layer (H) against the influence of drying while the D-fluxes outflowing directly through the exposed surfaces of both middle and outside layers. This results in a larger moisture concentration difference between the outside and middle layers, Fig. 2(b), because of greater moisture loss by the outside and less moisture loss by the middle compare to the HSH sample.

The drying rate of the outside layers controlled the sample drying rate since their volume was greater than the volume of the middle layer by one third of the latter. Therefore, the SHS sample dried faster than the HSH sample, although the I-flux direction in the former was the outer to middle layer, against the influence of drying, since the outside layers of SHS lost moisture at much greater rates than the outside layers of HSH.

Since the temperature history was monitored at the geometrical center of the middle layer, the history is influenced strongly by the drying rate of this layer (latent heat effect). Therefore, the central temperature of the HSH sample was slightly lower than that of the other sample because the HSH middle layer dried faster than the SHS middle layer.

There are three principal stresses formed at any location in a strained body. These stresses are oriented orthogonally to each other and unidirectional stresses (tensile or compressional stresses) are locally maximum along these orientations.

Figure 4 shows the orientation and magnitudes of two principal stresses estimated at each of 16 selected locations (intersection of each pair of bars) on the one quarter of the 15.8×14 mm bisected plane of the HSH sample, both acted on the plane, at 0.2 and 1 h of drying together with photographs of stress-cracks, on the same plane. The solid line bars represent tensile principal stresses and broken line bars represent compressional principal stresses. Only one bar is shown when another was too small to show and no bar was shown when both were too small. The third principal stresses, which are not shown, were orthogonal to the plane, i.e. the stresses shown in the figure, and less than those shown. Since the sample is brittle, the maximum tensile principal, stress may be used as a criterion for fracture [27]. Large principal, tensile stresses were formed at 0.2 h in the middle layer, at a location close

to the interface and to the exposed side surface. One oriented about 45° from the *x*-axis and another about 135° from the axis, both about 4800 kN/m². Therefore, cracks could be formed at this location. The magnitudes of tensile stresses were reduced greatly at 1.0 h. No cracks of significant sizes are apparent from the photograph taken at 0.2 h. However, the photograph at 1.0 h shows a large crack formed at a location close to the location of the large tensile stress estimated at 0.2 h, the crack extended to the outside layer. Cracks could be formed shortly after 0.2 h since the magnitudes of tensile, stresses were reduced after 0.2 h. The crack extension to the outside layer could be due to crack propagation which was not addressed in the present study.

Figure 5 shows simulation results and photographs for the SHS samples. The maximum tensile, principal stress at 0.2 h was about 60% of the maximum stress in the HSH sample. At 1.0 h, stresses were reduced greatly. Therefore, less cracks would be formed in the SHS compared to the HSH. The photographs show no cracks of significant sizes.

There was no shrinkage in both H and S when $W < 0.2$ g water g dry solid⁻¹ as indicated by the shrinkage function in Table 1. Since the initial sample moisture was 0.2 g g⁻¹, there should not be volumetric change in the sample if it is homogeneous. However, with the HSH sample, there was the I-flux from the middle layer (S) to the outside layers (H). Therefore, moisture concentrations became greater than 0.2 g g⁻¹ at locations adjacent to the interface in the outside layers according to the simulation. This caused volumetric changes at these locations, resulting in stress formation. Similar simulation results were obtained with the SHS sample.

CONCLUSIONS

Governing equations were obtained to simulate transient state, three dimensional heat and moisture transfer and viscoelastic hygrostress formation in a composite body during drying. In these equations, the chemical potential of water in food was used as a mass transfer potential since moisture concentration could not be used because different composite components had different affinities to moisture. These equations were solved numerically by a finite element method subdividing a composite body into 20-nodes isoparametric elements.

Heat and moisture transfer and hygrostress formation were simulated for the layered bricks made from the hydrates of starch powders and of starch powders-sucrose 3:1 mixture, which were subjected to forced-air drying. Simulated results agreed favorably with those observed experimentally. Both simulated and observed results show the strong influence of layer-arrangement on moisture transfer and stress formation and slight influence on heat transfer.

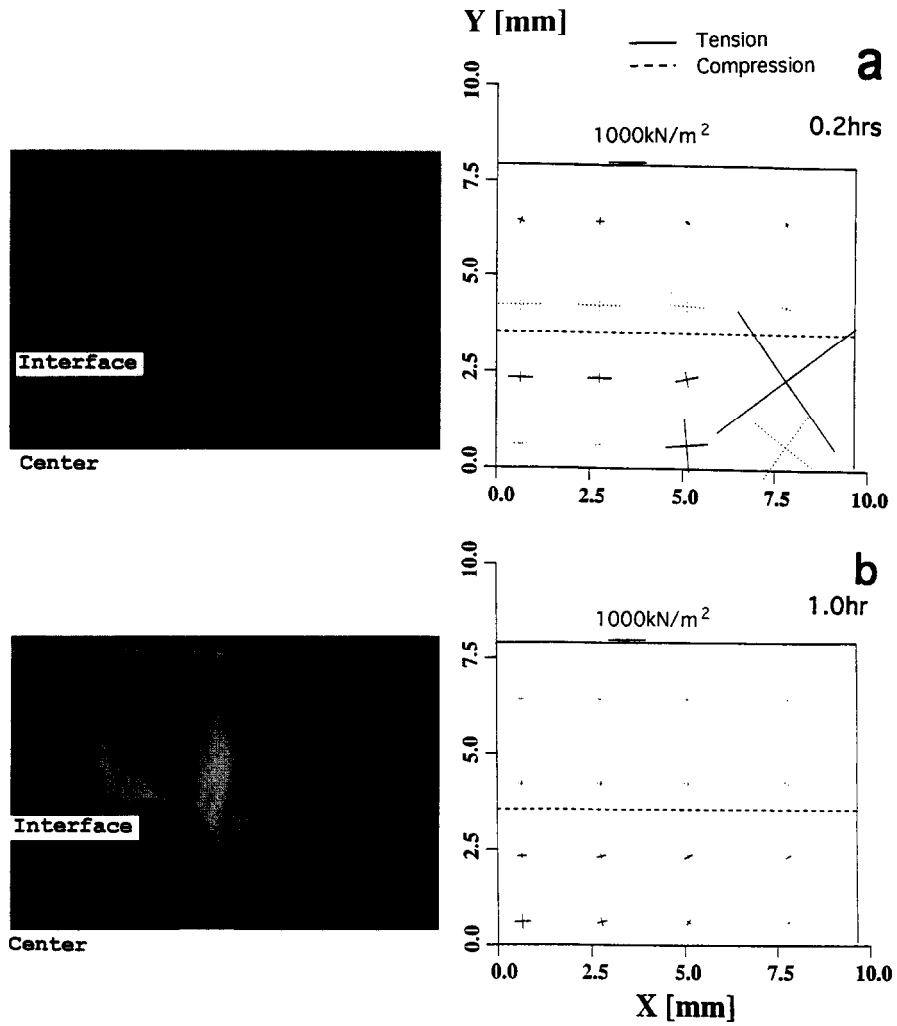


Fig. 4. Estimated two principal stresses at 16 selected locations, specified by intersection of each pair of bars, on one quarter of bisected face of HSH brick and photographed cracks at 0.2 h (a) and 1.0 h (b) of drying. The stress magnitude and orientation are presented by bar-length and bar-orientation, respectively. Solid and broken bars represent tensile and compressional stresses, respectively.

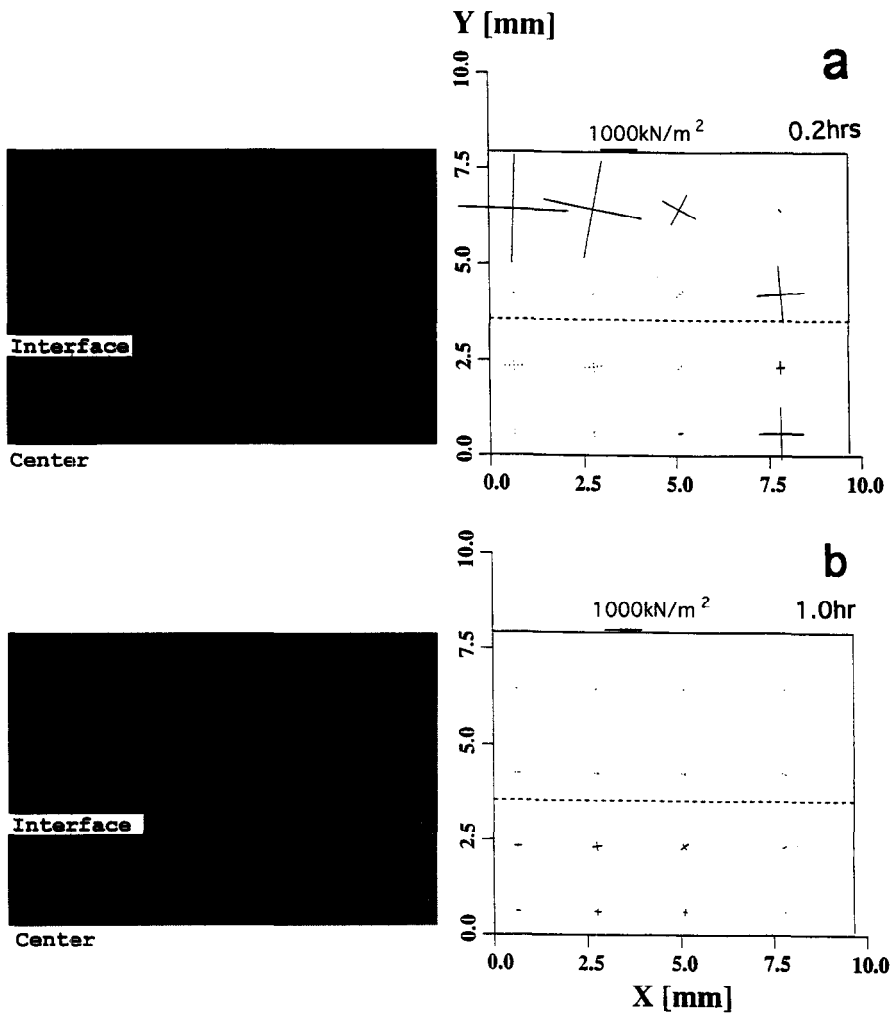


Fig. 5. Estimated two principal stresses at 16 selected locations and observed cracks on one quarter of bisected face of SIIS brick at 0.2 h (a) and 1.0 h (b).

Table 1. Drying conditions, transport and physical properties used for the simulation†

| | |
|--|--|
| Air temperature (T_a) | 323 [K] |
| Initial temperature (T_o) | 298 [K] |
| Initial moisture content (W_o): | 0.2 [kg water (kg solid) ⁻¹] |
| Relative humidity in air (RH) | 0.11 |
| Surface heat transfer coefficient (h_i) | 23.2 [W m ⁻² K ⁻¹] |
| Surface mass transfer coefficient (h_m) | 8.22×10^{-5} [kg m ⁻² Pa ⁻¹ s ⁻¹] |
| Volumetric shrinkage coefficient of H (S_v) | $\begin{cases} 1.000 + 0.601(W - 0.2) & \text{for } W \geq 0.2 \\ 1.00 & \text{for } W < 0.2 \end{cases}$ |
| Volumetric shrinkage coefficient of S (S_v) | $\begin{cases} 1.000 + 0.737(W - 0.2) & \text{for } W \geq 0.2 \\ 1.00 & \text{for } W < 0.2 \end{cases}$ |
| Moisture diffusivity (D_w) | |
| $D_w = 1.448 \times 10^{-4} (6.402 \times 10^{-4} + W \times 10^{-3})^2 W^{0.5954} \times \exp [8 \times 10^4 (0.147 + 1/(1 + 10W))$ | $\times (1/323 - 1/(T + 273))/8.314]$ [m ² s ⁻¹] |
| Soret mass diffusivity (D_t) | 5.54×10^{-9} [kg m ⁻¹ s ⁻¹ K ⁻¹] |
| Pressure mass diffusivity (D_p) | 0.0 [s] |
| Thermal conductivity (k_t) | $0.35 + 1.16 \times 10^{-3}(T - 273) + 0.058W/(1 + W)$ [W m ⁻¹ K ⁻¹] |
| Dufour thermal conductivity (k_c) | 0.0 [W m ² kg ⁻¹] |
| Filtrational thermal conductivity (k_p) | 0.0 [W m ⁻¹ Pa ⁻¹] |
| Luikov's phase conversion criteria (ϵ_L) | $1 - W/W_o$ |
| Specific heat (c_p) | $1.90 \times 10^3 - 12.1(T - 273) + 1.73 \times 10^3 W/(1 + W)$ [J kg ⁻¹ K ⁻¹] |
| Density of H (ρ_s) | $0.603 \times 10^3(1 + W)/S_v$ [kg m ⁻³] |
| Density of S (ρ_s) | $0.741 \times 10^3(1 + W)/S_v$ [kg m ⁻³] |
| Chemical reaction rate (R_i) | 0.0 [kg s ⁻¹] |
| Equilibrium moisture content (W_e) | $W_n = \frac{W_n E Q a_w}{(1 - Q a_w)(1 - Q a_w + E Q a_w)}$ |
| H | S |
| $E = 7.198 \times 10^{-4} \exp(3021/T)$, | $E = 1.37 \times 10^{-3} \exp(3021/T)$ |
| $Q = 0.370 \exp(142.1/T)$, | $Q = 0.642 \exp(142.1/T)$ |
| $w_n = 0.07259 \exp(162.6/T)$, | $w_n = 0.02088 \exp(162.6/T)$ |
| Equilibrium vapor pressure on the surface (P) | $P = (101.3 \times 10^3 \exp[13.087(1 - 373.15/T_s)] \cdot (a_w \text{ at surface moisture}))$ [Pa] |
| Bulk and shear moduli ($K(t)$, $G(t)$) | |
| | $K(t) = \frac{E(t)}{3(1 - 2\nu)}$ $G(t) = \frac{E(t)}{2(1 + \nu)}$ |
| | $E(t) = 1.31 \times 10^8 + 2.00 \times 10^8 \exp(-t/3.50 \times 10^2) + 8.27 \times 10^7 \exp(-t/1.43 \times 10^2)$ [Pa] |
| $\nu = 0.35$ | |
| $n_s = 1/3$ | |
| Moisture shift factor (a_M) | 1.0 |
| Temperature shift factor (a_T) | 1.0 |
| Stefan-Boltzman constant (β) | 5.67×10^{-8} [W m ⁻² K ⁻⁴] |
| ϕ_2 | 0.64 |

† Applicable to both H and S unless stated otherwise. Property values were from Furuta *et al.* [26], Sakai and Hayakawa [18] and Tsukada *et al.* [3].

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APPENDIX

The elements of the matrices and vectors in equations (50) and (51) are shown below :

$$\mathbf{A}_{MM} = \int_{v_T} \left(K_{MMX} \frac{\partial \mathbf{N}^T}{\partial X} \frac{\partial \mathbf{N}}{\partial X} + K_{MMY} \frac{\partial \mathbf{N}^T}{\partial Y} \frac{\partial \mathbf{N}}{\partial Y} + K_{MMZ} \frac{\partial \mathbf{N}^T}{\partial Z} \frac{\partial \mathbf{N}}{\partial Z} \right) dV \quad (\text{A1})$$

$$\mathbf{A}_{MT} = \int_{v_T} \left(K_{MTX} \frac{\partial \mathbf{N}^T}{\partial X} \frac{\partial \mathbf{N}}{\partial X} + K_{MTY} \frac{\partial \mathbf{N}^T}{\partial Y} \frac{\partial \mathbf{N}}{\partial Y} + K_{MTZ} \frac{\partial \mathbf{N}^T}{\partial Z} \frac{\partial \mathbf{N}}{\partial Z} \right) dV \quad (\text{A2})$$

$$\mathbf{A}_{TM} = \int_{v_T} \left(K_{TMX} \frac{\partial \mathbf{N}^T}{\partial X} \frac{\partial \mathbf{N}}{\partial X} + K_{TMY} \frac{\partial \mathbf{N}^T}{\partial Y} \frac{\partial \mathbf{N}}{\partial Y} + K_{TMZ} \frac{\partial \mathbf{N}^T}{\partial Z} \frac{\partial \mathbf{N}}{\partial Z} \right) dV \quad (\text{A3})$$

$$\mathbf{A}_{TT} = \int_{v_T} \left(K_{TTX} \frac{\partial \mathbf{N}^T}{\partial X} \frac{\partial \mathbf{N}}{\partial X} + K_{TTY} \frac{\partial \mathbf{N}^T}{\partial Y} \frac{\partial \mathbf{N}}{\partial Y} + K_{TTZ} \frac{\partial \mathbf{N}^T}{\partial Z} \frac{\partial \mathbf{N}}{\partial Z} \right) dV + (H_T + \beta_d) \int_{S_a} \mathbf{N}^T \mathbf{N} dS \quad (\text{A4})$$

$$\mathbf{B}_{MM} = C_{MM} \int_{v_T} \mathbf{N}^T \mathbf{N} dv \int_{v_i} \mathbf{N} dV \quad (\text{A5})$$

$$\mathbf{B}_{MT} = C_{MT} \int_{v_T} \mathbf{N}^T \mathbf{N} dV \quad (\text{A6})$$

$$\mathbf{B}_{TM} = -C_{TM} \int_{v_T} \mathbf{N}^T \mathbf{N} dV \quad (\text{A7})$$

$$\mathbf{B}_{TT} = C_{TT} \int_{v_T} \mathbf{N}^T \mathbf{N} dV \int_{v_T} \mathbf{N}^T \mathbf{N} dV \quad (\text{A8})$$

$$\mathbf{F}_M = -H_M(\psi - 1) \int_{S_a} \mathbf{N}^T dS + R_j^d \int_{v_T} \mathbf{N}^T dV \quad (\text{A9})$$

$$\mathbf{F}_T = \{H_T + \beta_d - (1 - \epsilon_L) H_M \Delta H_v^d(\psi - 1)\} \int_{S_a} \mathbf{N}^T dS + \sum \Delta H_{ij}^d R_j^d \int_{v_T} \mathbf{N}^T dV \quad (\text{A10})$$