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MECHANICAL DEGRADATION OF NATURAL FIBER REINFORCED COMPOSITE MATERIALS UNDER CONSTRAINED SWELLING

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Natural fiber reinforced composite materials (NFRCMs) have found more and more applications because of their excellent performances over traditional fiber reinforced composites. However, mechanical properties of these materials may be dramatically degraded in a humid environment, whether subject to mechanical loading or not. A nonlinear constitutive model is established for unidirectional natural fiber reinforced composites under large swelling deformation based on nonequilibrium thermodynamics, in which an internal variable is incorporated in the Helmholtz free energy to consider the irreversible energy dissipation induced by moisture absorption. The Helmholtz free energy is further decomposed into the base free energy of the isotropic matrix, the reinforcing energy of fiber stretching and the free energy of volume expansion. Two kinds of reinforcing energy (the I_4-dependent model and the I_5-dependent model) are employed to predict the degradation of the elastic modulus for the cases of free swelling and constrained swelling. It is found that the predictions from these two models are identical for the case of free swelling and agree well with available experimental data. As for the case of constrained swelling, these two models yield obviously different results.

1. Introduction

Natural fiber reinforced composite materials (NFRCMs) have been drawing great attention due to their excellent advantages over glass or other traditional fiber reinforced composites. For example, natural fibers possess high specific strength and modulus because of their light weight [Athijayamani et al. 2009; Medina et al. 2009]. Moreover, they are environmentally friendly with biodegradable properties. Hence, NFRCMs have found more and more applications in the aerospace and automobile industries. In spite of so many advantages, there are still some disadvantages to these new composite materials, such as moisture absorption and weak adhesion to hydrophobic matrices [Sgriccia et al. 2008]. The worst is the mechanical degradation of NFRCMs induced by moisture absorption in a humid environment, especially losses of tensile and shear moduli.

Natural fibers are usually hydrophilic and have a porous structure [Lu et al. 2003] which can transport large amounts of water from an external humid environment [Espert et al. 2004]. There are two main causes for the mechanical degradation of NFRCMs induced by moisture absorption. First, water uptake will cause natural fibers to swell and induce fiber aging, during which natural fibers gradually soften due to a loss of elastic moduli [Song et al. 2011]. Second, interfaces between the matrix and the fibers of

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NFRCMs will be continuously damaged by moisture absorption [Dhakal et al. 2007], so that in the worst case, the interfaces would be totally damaged and the fibers completely pulled out [Sydenstricker et al. 2003], losing their reinforcing effects on the matrix.

Since moisture absorption in a humid environment is inevitable, it is significant to study moisture absorption and its effects on the mechanical properties of NFRCMs. However, Fick's diffusion law fails to describe the process of moisture absorption because the permeability of water diffusion is no longer a constant, which changes with interfacial damage and fiber aging during moisture absorption [Hu et al. 2010].

The moisture-induced swelling deformation of NFRCMs was originally studied by Tsai et al. [2004] based on a finite elasticity description, considering a finite strain flexure in an isotropic rectangular block. Then the theoretical framework was applied to fiber reinforced composite materials with absorbent matrix and hydrophobic fibers [Demirkoparan and Pence 2007a; 2007b; 2008; Fang et al. 2011].

The swelling-induced mechanical degradation of polymeric materials is another research topic of common interest. Baek and Pence [2009] developed a constitutive model considering the swelling and mechanical degradation of fiber reinforced composites based on the energy dissipation framework [Rajagopal and Srinivasa 2004; Rajagopal et al. 2007; Karra and Rajagopal 2012]. The purely mechanical induced degradation of fiber reinforced composites was also studied [Baek and Pence 2011].

In [Pan and Zhong 2014b], large swelling deformation and nonlinear mechanical responses of natural fiber reinforced composites are taken into account by adopting a special form of the Helmholtz free energy which depends on four scalar strain invariants reflecting the deformation characteristics of transverse isotropy of unidirectional fiber reinforced composites [Qiu and Pence 1997] and an internal variable describing the moisture absorption process. This model is referred to as the I_4-dependent model in the present paper.

In the present paper, instead of using the I_4-dependent model, another model for reinforcing energy which was studied in detail by Merodio and Ogden [2005] is employed to consider the unidirectional reinforcement of natural fibers in a neo-Hookean matrix, which is called the I_5-dependent model. The degradation of elastic modulus is predicted for the cases of free swelling and constrained swelling based on the newly developed model and compared with those obtained from the I_4-dependent model. It is found that the predictions from these two models are identical for the case of free swelling and deviate obviously from each other for the case of constrained swelling.

This paper is organized as follows. In Section 2, a general constitutive model is described based on nonequilibrium thermodynamics. Then in Section 3 we specialize the I_5-dependent model to study the mechanical degradation of NFRCMs under free swelling and constrained swelling. In Section 4, results are obtained for a unidirectional sisal fiber reinforced benzylated wood based on the I_5-dependent model and compared with those from the I_4-dependent model. Finally, in Section 5, we draw the conclusions.

2. Theoretical formulation

2.1. ***Deformation decomposition.*** Consider a natural fiber reinforced composite in a humid environment. The deformation gradient is defined by $\boldsymbol{F} = \partial \boldsymbol{x}/\partial \boldsymbol{X}$, where $\boldsymbol{X}$ represents the position vector of a material particle in the initial dry and undeformed configuration, and $\boldsymbol{x}$ is the corresponding position vector of this

particle in the current swollen and deformed configuration, from which the right and the left Cauchy–Green strain tensors are given as $\boldsymbol{C} = \boldsymbol{F}^T \cdot \boldsymbol{F}$ and $\boldsymbol{b} = \boldsymbol{F} \cdot \boldsymbol{F}^T$. Here the dot product "$\cdot$" denotes a contraction (inner product) over one index between two tensors (or vectors), and the superscript "T" denotes the transpose of a tensor.

If we adopt a multiplicative decomposition of the deformation gradient into volume-changing (dilational) and volume-preserving (distortional) parts [Holzapfel 2000], then we have

$$\boldsymbol{F} = J^{1/3}\bar{\boldsymbol{F}}, \tag{1}$$

where $J = \det \boldsymbol{F}$ is the volume ratio and $\bar{\boldsymbol{F}}$ is called the modified deformation gradient tensor. Accordingly, the modified right and left Cauchy–Green strain tensors can be defined as $\bar{\boldsymbol{C}} = \bar{\boldsymbol{F}}^T \cdot \bar{\boldsymbol{F}}$ and $\bar{\boldsymbol{b}} = \bar{\boldsymbol{F}} \cdot \bar{\boldsymbol{F}}^T$. Here we refer to a case as free swelling when $\bar{\boldsymbol{C}} = \boldsymbol{I}$ (or equivalently $\bar{\boldsymbol{b}} = \boldsymbol{I}$) and otherwise as constrained swelling if $\bar{\boldsymbol{C}} \neq \boldsymbol{I}$ (or equivalently $\bar{\boldsymbol{b}} \neq \boldsymbol{I}$), with $\boldsymbol{I}$ being the second-order identity tensor.

Furthermore, three principal invariants of $\boldsymbol{C}$ (also $\boldsymbol{b}$) are given as

$$I_1 = \mathrm{tr}\boldsymbol{C} = J^{2/3}\mathrm{tr}\bar{\boldsymbol{C}} = J^{2/3}\bar{I}_1, \tag{2a}$$

$$I_2 = \tfrac{1}{2}[(\mathrm{tr}\boldsymbol{C})^2 - \mathrm{tr}(\boldsymbol{C}^2)] = \tfrac{1}{2}J^{4/3}[(\mathrm{tr}\bar{\boldsymbol{C}})^2 - \mathrm{tr}(\bar{\boldsymbol{C}}^2)] = J^{4/3}\bar{I}_2, \tag{2b}$$

$$I_3 = \det \boldsymbol{C} = J^2, \tag{2c}$$

where the notations "tr" and "det" denote, respectively, the trace and the determinant of a tensor, and $\bar{I}_1$ and $\bar{I}_2$ are the first and the second principal invariants of $\bar{\boldsymbol{C}}$, with the third principal invariant $\bar{I}_3 = 1$.

2.2. ***Material model.*** For a transversely isotropic material, with the axis of transverse isotropy defined by a unit vector $\boldsymbol{a}_0$ in the initial configuration, the Helmholtz free energy depends not only on the deformation tensor $\boldsymbol{C}$, but also the second-order tensor $\boldsymbol{a}_0 \otimes \boldsymbol{a}_0$, which can be written as [Moon and Spencer 1988; Spencer 1984]

$$W = W(\boldsymbol{C}, \boldsymbol{a}_0 \otimes \boldsymbol{a}_0) = W(\bar{I}_1, \bar{I}_2, J, \bar{I}_4, \bar{I}_5), \tag{3}$$

where

$$\bar{I}_4 = \boldsymbol{a}_0 \cdot \bar{\boldsymbol{C}} \cdot \boldsymbol{a}_0 = \lambda_f^2, \quad \bar{I}_5 = \boldsymbol{a}_0 \cdot (\bar{\boldsymbol{C}})^2 \cdot \boldsymbol{a}_0, \tag{4}$$

and $\lambda_f = \sqrt{\bar{I}_4}$ is the fiber stretch.

As special cases, (3) reduces to the I_4-dependent model [Qiu and Pence 1997]

$$W = W(\bar{I}_1, \bar{I}_2, J, \bar{I}_4) \tag{5}$$

when the Helmholtz free energy is independent of $\bar{I}_5$, or the I_5-dependent model [Merodio and Ogden 2005]

$$W = W(\bar{I}_1, \bar{I}_2, J, \bar{I}_5) \tag{6}$$

when the Helmholtz free energy is independent of $\bar{I}_4$.

Furthermore, the free energy W is assumed to be decomposed into three parts:

$$W = W_m(\bar{I}_1, \bar{I}_2) + W_f(\bar{I}_4, \bar{I}_5) + W_v(J). \tag{7}$$

The first term in the right-hand side of (7) represents the base Helmholtz free energy of the isotropic matrix induced by mechanical loading, while the second term is the reinforcing energy associated with

the stretching of the embedded fibers under the mechanical loading and the last term stands for the free energy of volume expansion induced by fiber swelling.

2.3. ***Internal variable.*** Upon moisture absorption, the interface between the matrix and the fiber of NFRCMs may be damaged [Dhakal et al. 2007; Sgriccia et al. 2008]. At the same time, the fiber also undergoes aging due to large amounts of water absorbed by natural fibers [Espert et al. 2004; Gao et al. 2011; Song et al. 2011]. Since all these processes are thermodynamically irreversible with energy dissipation, an internal variable is tentatively introduced to account for the effect of the composite microstructure change induced by moisture absorption. The internal variable α varies from 0 to 1 such that $\alpha = 0$ corresponds to the initial dry state and $\alpha = 1$ represents the fully saturated state. It is known that in the dry state, the composite maintains its initial mechanical properties. As α evolves from 0 to 1, the mechanical properties, such as Young's modulus, gradually degrade due to moisture absorption. Such degradation effects are reflected by the softening of the matrix and the reinforcement. Hence, the Helmholtz free energy for this transversely isotropic composite should also depend on the internal variable α, so that (7) is further modified to give

$$W = W_m(\alpha, \bar{I}_1, \bar{I}_2) + W_f(\alpha, \bar{I}_4, \bar{I}_5) + W_v(J). \tag{8}$$

Here the internal variable α acts as an additional independent variable in (8).

Note that Baek and Pence [2009; 2011] used two internal variables to describe the mechanical degradation of matrix and the fiber, respectively. In this paper, only one internal variable is employed for natural fiber composites, since both the interface damage and the fiber degradation are treated as a unified thermodynamic process induced by moisture absorption. This treatment simplifies the solution of the evolution of the internal variable and also gives a sufficient accuracy in describing the mechanical degradation of natural fiber composites as illustrated in Section 4.

Since the internal variable α is related to the absorption process of the natural fiber, the volume ratio J of the composite is assumed to be a function of α, i.e.,

$$J = J(\alpha). \tag{9}$$

For an incompressible composite without moisture absorption, we have $J = 1$. Furthermore, a linear relation between J and α can be derived based on the assumption of molecular incompressibility [Hong et al. 2008] as

$$J = 1 + \theta\alpha, \tag{10}$$

where θ is the equilibrium volume fraction of water in the composites defined with respect to the initial dry configuration, and can be derived from experiments. Equation (10) is an ideal mixing approximation of the moisture absorption process that the swelling volume is the volume sum of the dry composite and the water uptake.

The constraint $J = J(\alpha)$ can be accounted for by introducing a Lagrange multiplier term into the Helmholtz free energy, so that

$$W = W_m(\alpha, \bar{I}_1, \bar{I}_2) + W_f(\alpha, \bar{I}_4, \bar{I}_5) + W_v(J) + \Pi[J - J(\alpha)], \tag{11}$$

where the Lagrange multiplier term Π is interpreted as an osmotic pressure and could be determined from boundary conditions.

2.4. ***Thermodynamics.*** When the composite is subject to mechanical loading and a humid environment, there are two mechanisms doing work on the composite: mechanical loading and chemical potential of the external environment. During this process, the Clausius–Duhem inequality

$$J\boldsymbol{\sigma} : \boldsymbol{d} + \mu r + \mu\dot{\alpha} - \boldsymbol{q} \cdot \nabla\mu \geq \dot{W} \tag{12}$$

holds, where $\boldsymbol{\sigma}$ is the Cauchy stress, μ is the chemical potential, r stands for the internal source of water (however, in most cases there is no internal source and $r = 0$), $\boldsymbol{q}$ represents the water flowing out through the element area, and the rate of deformation tensor is defined as

$$\boldsymbol{d} = \tfrac{1}{2}(\dot{\boldsymbol{F}} \cdot \boldsymbol{F}^{-1} + \boldsymbol{F}^{-T} \cdot \dot{\boldsymbol{F}}^{T}). \tag{13}$$

From (11), the rate of the Helmholtz free energy $\dot{W}$ is calculated as

$$\dot{W} = 2\mathbb{P} : [\omega_1\bar{\boldsymbol{b}} + \omega_2\bar{\boldsymbol{b}}^2 + \omega_4\bar{\boldsymbol{a}} \otimes \bar{\boldsymbol{a}} + \omega_5(\bar{\boldsymbol{a}} \otimes \bar{\boldsymbol{b}} \cdot \bar{\boldsymbol{a}} + \bar{\boldsymbol{b}} \cdot \bar{\boldsymbol{a}} \otimes \bar{\boldsymbol{a}})] : \boldsymbol{d} + J\omega_3\boldsymbol{I} : \boldsymbol{d} + Y^{\alpha}\dot{\alpha}, \tag{14}$$

with

$$\begin{aligned} &\omega_1 = \frac{\partial W_m}{\partial \bar{I}_1} + \bar{I}_1\frac{\partial W_m}{\partial \bar{I}_2}, \qquad \omega_2 = -\frac{\partial W_m}{\partial \bar{I}_2}, \qquad \omega_3 = \frac{\partial W_v}{\partial J} + \Pi, \\ &\omega_4 = \frac{\partial W_f}{\partial \bar{I}_4}, \qquad \omega_5 = \frac{\partial W_f}{\partial \bar{I}_5}, \qquad Y^{\alpha} = \frac{\partial(W_m + W_f)}{\partial \alpha}, \end{aligned} \tag{15}$$

where $\boldsymbol{b}$ and $\bar{\boldsymbol{b}}$ are the left Cauchy–Green strain tensors corresponding to $\boldsymbol{F}$ and $\bar{\boldsymbol{F}}$, respectively, $\boldsymbol{I}$ is the second-order identity tensor, the projection tensor $\mathbb{P} = \mathbb{I} - (\boldsymbol{I} \otimes \boldsymbol{I})/3$ is defined with respect to the current configuration, $\mathbb{I}$ is the fourth-order identity tensor, $\bar{\boldsymbol{a}} = \bar{\boldsymbol{F}} \cdot \boldsymbol{a}_0$ stands for the fiber direction after mechanical loading, and Y^{α} is the thermodynamic force conjugate to the thermodynamic flow $\dot{\alpha}$.

Substituting (14) into (12) when $r = 0$, we have

$$(J\boldsymbol{\sigma} - \mathbb{P} : \bar{\boldsymbol{\sigma}} - J\boldsymbol{\sigma}^{S}) : \boldsymbol{d} + (\mu - Y^{\alpha})\dot{\alpha} - \boldsymbol{q} \cdot \nabla\mu \geq 0, \tag{16}$$

where

$$\begin{aligned} \bar{\boldsymbol{\sigma}} &= 2[\omega_1\bar{\boldsymbol{b}} + \omega_2\bar{\boldsymbol{b}}^2 + \omega_4\bar{\boldsymbol{a}} \otimes \bar{\boldsymbol{a}} + \omega_5(\bar{\boldsymbol{a}} \otimes \bar{\boldsymbol{b}} \cdot \bar{\boldsymbol{a}} + \bar{\boldsymbol{b}} \cdot \bar{\boldsymbol{a}} \otimes \bar{\boldsymbol{a}})], \\ \boldsymbol{\sigma}^{S} &= \omega_3\boldsymbol{I}. \end{aligned} \tag{17}$$

In the thermodynamic inequality (16), the first and the second terms represent, respectively, the energy dissipation caused by external forces and chemical potential. If we assume that the equilibriums of mechanical loading and chemical potential are achieved instantaneously, then

$$\boldsymbol{\sigma} = J^{-1}\mathbb{P} : \bar{\boldsymbol{\sigma}} + \boldsymbol{\sigma}^{S}, \tag{18}$$

where $J^{-1}\mathbb{P} : \bar{\boldsymbol{\sigma}}$ and $\boldsymbol{\sigma}^{S}$ are the Cauchy stresses due to isochoric elastic deformation and volumetric swelling deformation, respectively, and

$$\mu = Y^{\alpha}, \tag{19}$$

which relates the chemical potential to the variation of the Helmholtz free energy with the internal variable.

The third term stands for the energy dissipation induced by water transport, which requires that the following inequality should hold:

$$\boldsymbol{q} \cdot \nabla \mu \leq 0. \tag{20}$$

Hence, the energy dissipation caused by the thermodynamic flow $\dot{\alpha}$ with its conjugate force Y^{α} is given as

$$\Xi = -Y^{\alpha}\dot{\alpha}. \tag{21}$$

3. Mechanical degradation due to moisture absorption

In this section, the theoretical formulations developed above are specialized with the I_5-dependent model to study the mechanical degradation of NFRCMs under constrained swelling and free swelling.

3.1. ***Moisture absorption.*** To study the influence of moisture absorption on the mechanical properties of NFRCMs, for simplicity and without loss of generality we study a cubic NFRCM sample exposed to a humid environment and simultaneously subjected to (possible) mechanical loading, corresponding to the case of constrained swelling. The isochoric elastic deformation is described by three principal stretches $\bar{\lambda}_1, \bar{\lambda}_2, \bar{\lambda}_3$ of the deformation gradient $\bar{\boldsymbol{F}}$, with

$$\bar{I}_1 = \bar{\lambda}_1^2 + \bar{\lambda}_2^2 + \bar{\lambda}_3^2, \quad \bar{I}_2 = \bar{\lambda}_1^2\bar{\lambda}_2^2 + \bar{\lambda}_2^2\bar{\lambda}_3^2 + \bar{\lambda}_3^2\bar{\lambda}_1^2 \quad \text{and} \quad \bar{I}_3 = (\bar{\lambda}_1\bar{\lambda}_2\bar{\lambda}_3)^2 = 1.$$

Considering that the interfaces between the matrix and the fibers are deteriorated gradually due to moisture absorption [Sgriccia et al. 2008], we modify the neo-Hookean model for the isotropic polymer matrix to obtain [Baek and Pence 2009; Karra and Rajagopal 2012]

$$W_m = \tfrac{1}{2}G[1-\beta_1(c)\alpha](\bar{I}_1 - 3) = \tfrac{1}{2}G[1-\beta_1(c)\alpha]\Big[\sum_i (\bar{\lambda}_i)^2 - 3\Big], \tag{22}$$

where G is the initial shear modulus of the matrix, $\beta_1(c)$ is a dimensionless parameter reflecting the influence of the degradation on the matrix, which depends on the fiber content c. Note that the interface damage increases with the fiber content, so that β_1 should be a monotonically increasing function of c. Equation (22) is called the modified neo-Hookean model, which reduces to the original neo-Hookean model if $\alpha = 0$.

If we assume the initial fiber direction is taken along the X_1-axis in the initial configuration, then $\bar{I}_5 = \bar{\lambda}_1^4$, with $\bar{\lambda}_1$ being the fiber stretch induced by mechanical loading. Accordingly, the reinforcing energy based on the I_5-dependent model is given by [Merodio and Ogden 2005]

$$W_f = \tfrac{1}{2}G\bar{\gamma}(\bar{I}_5 - 1)^2 = \tfrac{1}{2}G\bar{\gamma}[(\bar{\lambda}_1)^4 - 1]^2, \tag{23}$$

where $\bar{\gamma}$ is the relative stiffness of the fiber with respect to the matrix, which can be taken as $\bar{\gamma} = c\gamma$ for the first approximation, with γ being a coefficient of proportionality. Further considering the interface damage and the fiber degradation, (23) is modified to the form [Baek and Pence 2009]

$$W_f = \tfrac{1}{2}G\bar{\gamma}(1-\beta_2\alpha)[(\bar{\lambda}_1)^4 - 1]^2, \tag{24}$$

where β_2 is a dimensionless constant referred to as the maximum degradation parameter of the fiber. In the case of $\alpha = 0$, the modified reinforcing energy reduces to the original one given by Merodio and

Ogden [2005]. The volume expansion energy W_v induced by the fiber swelling is specified as [Hong et al. 2008]

$$W_v = -G\bar{\gamma}\ln J. \tag{25}$$

With the above specified Helmholtz free energies for the matrix W_m, the reinforcing energy W_f and the volume expansion energy W_v, the constitutive relation of the composite is derived by substituting (10), (22), (24) and (25) into (18), as

$$\sigma_{11} = \tfrac{1}{3}Gd_1(\alpha)[2(\bar{\lambda}_1)^2 - (\bar{\lambda}_2)^2 - (\bar{\lambda}_3)^2] + (\Pi - c\gamma GJ^{-1}) + \tfrac{8}{3}c\gamma Gd_2(\alpha)[(\bar{\lambda}_1)^8 - (\bar{\lambda}_1)^4], \tag{26a}$$

$$\sigma_{22} = \tfrac{1}{3}Gd_1(\alpha)[2(\bar{\lambda}_2)^2 - (\bar{\lambda}_1)^2 - (\bar{\lambda}_3)^2] + (\Pi - c\gamma GJ^{-1}) + \tfrac{4}{3}c\gamma Gd_2(\alpha)[-(\bar{\lambda}_1)^8 + (\bar{\lambda}_1)^4], \tag{26b}$$

$$\sigma_{33} = \tfrac{1}{3}Gd_1(\alpha)[2(\bar{\lambda}_3)^2 - (\bar{\lambda}_1)^2 - (\bar{\lambda}_2)^2] + (\Pi - c\gamma GJ^{-1}) + \tfrac{4}{3}c\gamma Gd_2(\alpha)[-(\bar{\lambda}_1)^8 + (\bar{\lambda}_1)^4], \tag{26c}$$

with

$$d_1(\alpha) = (1+\theta\alpha)^{-1}(1-\beta_1\alpha) \quad \text{and} \quad d_2(\alpha) = (1+\theta\alpha)^{-1}(1-\beta_2\alpha). \tag{27}$$

Here $d_1(\alpha)$ and $d_2(\alpha)$ are regarded as degradation functions describing, respectively, the degradation of the shear modulus G and the relative stiffness $\bar{\gamma}$.

To derive the evolution equation of α, the dissipation function Ξ in (21) should be specified. Here we take the form that has been successfully used to describe material degradations from different dissipation mechanisms, e.g., thermo-oxidization, hydrolysis and swelling [Rajagopal and Srinivasa 2004; Rajagopal et al. 2007; Baek and Pence 2009; Soares et al. 2009; Karra and Rajagopal 2012]:

$$\Xi(\alpha, \dot{\alpha}) = \frac{\bar{D}\dot{\alpha}^{(n+1)/n}}{(1-\alpha)^{1/n}}, \tag{28}$$

where n is a rate-sensitive index and $\bar{D}$ is a parameter governing the speed of energy dissipation. To further reflect the effect of fiber volume fraction on the swelling speed, $\bar{D}$ is assumed to be proportional to the fiber content, so that $\bar{D} = cD$ with D being a coefficient of proportionality [Pan and Zhong 2014a; 2014b]. Then a combination of (21) with (28) yields

$$\left(\frac{\dot{\alpha}}{1-\alpha}\right)^{1/n} = \frac{G\beta_1}{2cD}\Big[\sum_i (\bar{\lambda}_i)^2 - 3\Big] + \frac{\gamma G\beta_2}{2D}[(\bar{\lambda}_1)^4 - 1]^2 + \frac{\Pi\theta}{cD}. \tag{29}$$

Equation (29) is an evolution equation of the internal variable α depending on the elastic stretches $\bar{\lambda}_i$, which provides a supplement of the constitutive relations given by (26).

In particular, if the cubic composite sample is immersed in water and swells freely without any mechanical loading, then we have $\bar{\lambda}_1 = \bar{\lambda}_2 = \bar{\lambda}_3 = 1$ and $\sigma_{11} = \sigma_{22} = \sigma_{33} = 0$. From (26a), the osmotic pressure is derived as

$$\Pi = c\gamma GJ^{-1}. \tag{30}$$

We further substitute (30) into (29) and set $\bar{\lambda}_i = 1$, and then derive the evolution equation of α for the case of free swelling as

$$\left(\frac{\dot{\alpha}}{1-\alpha}\right)^{1/n} = \frac{\gamma\theta G}{JD}, \tag{31}$$

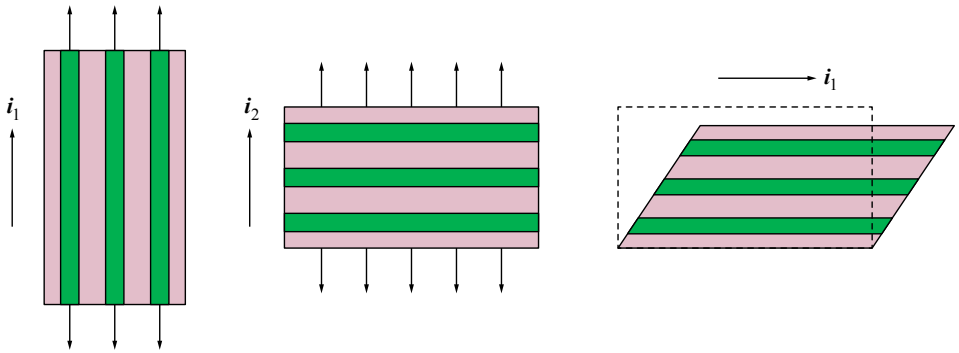

Figure 1. Three loading cases for deriving five elastic coefficients: (left) tensile along the fiber direction, (center) tensile transverse to the fiber direction, and (right) simple shear along the fiber direction.

which takes the same form as that in [Pan and Zhong 2014b] using the I_4-dependent model. But the evolution equations based on these two models are different in the case of constrained swelling.

3.2. ***The evolutions of five elastic coefficients.*** Based on the constitutive relation (26), the five elastic coefficients of a transversely isotropic composite material are derived for three loading cases (elastic deformation), as illustrated in Figure 1.

In the first case, a uniaxial tensile test along the reinforcing direction (X_1-axis) is studied, as shown in Figure 1(left). According to the incompressibility condition, the stretches should satisfy the relation

$$\bar{\lambda}_2 = \bar{\lambda}_3 = (\bar{\lambda}_1)^{-1/2}, \tag{32}$$

from which two Poisson's ratios are derived as

$$\upsilon_{12} = \upsilon_{13} = -\frac{d\bar{\lambda}_2}{d\bar{\lambda}_1}\Big|_{\bar{\lambda}_1=1} = \frac{1}{2}. \tag{33}$$

According to the boundary condition $\sigma_{22} = \sigma_{33} = 0$, the Lagrange multiplier Π is obtained as

$$\Pi = \tfrac{1}{3}Gd_1(\alpha)[(\bar{\lambda}_1)^2 - (\bar{\lambda}_1)^{-1}] + \tfrac{4}{3}c\gamma Gd_2(\alpha)[(\bar{\lambda}_1)^8 - (\bar{\lambda}_1)^4] + c\gamma GJ^{-1}. \tag{34}$$

Substituting (34) into (26a), the tensile stress σ_{11} along the fiber direction is obtained as

$$\sigma_{11} = Gd_1(\alpha)[(\bar{\lambda}_1)^2 - (\bar{\lambda}_2)^{-1}] + 4c\gamma Gd_2(\alpha)[(\bar{\lambda}_1)^8 - (\bar{\lambda}_1)^4]. \tag{35}$$

The tensile modulus E_1 along the fiber direction can be determined by the tangential modulus at $\bar{\lambda}_1 = 1$ from (35), and is calculated as

$$E_1 = \frac{d\sigma_{11}}{d\bar{\lambda}_1}\Big|_{\bar{\lambda}_1=1} = 3Gd_1(\alpha) + 16c\gamma Gd_2(\alpha). \tag{36}$$

The second case concerns a tensile loading transverse to the fiber direction, as illustrated in Figure 1(center), from which the Poisson's ratios υ_{21} and υ_{23} can be determined. In this case, the relation between

the three principal stretches is given as

$$\bar{\lambda}_3 = (\bar{\lambda}_1\bar{\lambda}_2)^{-1}. \tag{37}$$

From the given boundary conditions $\sigma_{11} = \sigma_{33} = 0$, it is established that

$$G(1-\beta_1\alpha)[(\bar{\lambda}_1)^2 - (\bar{\lambda}_3)^2] + 4c\gamma G(1-\beta_2\alpha)[(\bar{\lambda}_1)^8 - (\bar{\lambda}_1)^4] = 0. \tag{38}$$

Substituting (37) into (38), we then obtain the Poisson's ratio υ_{21} as

$$\upsilon_{21} = -\frac{d\bar{\lambda}_1}{d\bar{\lambda}_2}\Big|_{\bar{\lambda}_2=1} = \frac{1-\beta_1\alpha}{2[(1-\beta_1\alpha)+4c\gamma(1-\beta_2\alpha)]}. \tag{39}$$

Similarly, the Poisson's ratio υ_{23} is derived as

$$\upsilon_{23} = -\frac{d\bar{\lambda}_3}{d\bar{\lambda}_2}\Big|_{\bar{\lambda}_2=1} = \frac{(1-\beta_1\alpha)+8c\gamma(1-\beta_2\alpha)}{2[(1-\beta_1\alpha)+4c\gamma(1-\beta_2\alpha)]}. \tag{40}$$

The third loading case is a simple shear along the fiber direction, as shown in Figure 1(right), in which the deformation gradient is given as $\bar{\boldsymbol{F}} = \boldsymbol{I} + k\boldsymbol{e}_1 \otimes \boldsymbol{e}_2$. Then, from (17), the shear stress σ_{12} is given by

$$\sigma_{12} = Gd_1(\alpha)k + 2G\gamma d_2(\alpha)k^3. \tag{41}$$

Hence the shear modulus is derived as

$$G_{12} = G_{21} = \frac{\partial\sigma_{12}}{\partial k}\Big|_{k=0} = Gd_1(\alpha). \tag{42}$$

We have now obtained all five of the independent elastic coefficients. Other elastic coefficients can be derived based on some elastic relationships of transverse isotropy. For example, the tensile modulus E_2 is calculated as $E_2 = E_1\upsilon_{21}/\upsilon_{12}$.

4. Results and discussion

In this section, we will use the theory established above on the I_5-dependent model to predict the evolution of the moisture absorption and the mechanical degradation for a unidirectional sisal fiber reinforced benzylated wood in the cases of constrained swelling and free swelling, and compare the results to those derived based on the I_4-dependent model.

At first, some necessary material parameters are fitted based on the experimental results of Lu et al. [2003], in which three samples with different fiber contents were prepared: Sample 1 (fiber content $c_1 = 0.102$, equilibrium volume fraction of water $\theta_1 = 0.085$), Sample 2 ($c_2 = 0.196$, $\theta_2 = 0.114$) and Sample 3 ($c_3 = 0.304$, $\theta_3 = 0.148$). In their experiment, each sample was immersed in water to simulate the moisture absorption process in a humid environment. At several time intervals, they were taken out of the water to measure the loss of tensile modulus. By means of similar procedures [Pan and Zhong 2014b], the shear modulus (G), the relative stiffness (γ), and the maximum degradation parameters of the matrix and fiber ($\bar{\beta}_1$ and β_2) can be obtained by fitting the experimental data of the tensile modulus of the dry samples ($\alpha = 0$) and the fully saturated samples ($\alpha = 1$) based on (36), while the evolution

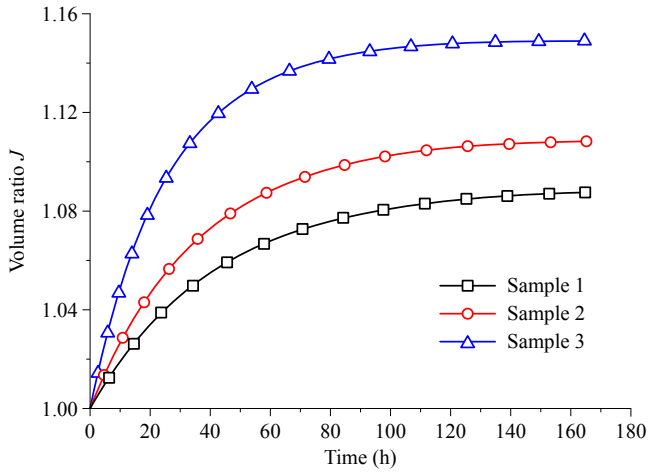

Figure 2. The predicted evolution of the volume ratio J for three samples under free swelling.

speed parameter (D) can be derived by fitting the evolution data of $\alpha(t)$ for Sample 1 based on (31). These parameters necessary for the I_5-dependent model are given as

$$G = 0.13\,\text{GPa}, \quad \gamma = 17.5, \quad \bar{\beta}_1 = 70.11, \quad \beta_2 = 0.56, \quad D = 3.22 \times 10^4\,\text{Nsm}^{-2}. \tag{43}$$

Employing the molecular incompressibility condition $J = 1 + \theta\alpha$, we can further obtain theoretically the evolution of the volume ratio of each sample from (26) and (31), which is illustrated in Figure 2. For these samples with three different fiber contents, the variation trends are identical, where the swelling speed is much faster at the initial stage, then tends to slow down, and finally terminates at the equilibrium state. This phenomenon is interpreted as the gradual increase of the chemical potential inside the composite. Moreover, a comparison between these samples shows that the kinetic swelling process is sensitive to the fiber content such that the swelling speed of a composite with higher fiber content is bigger than that with lower fiber content.

Next, the theoretical framework established in Sections 2 and 3 is used to predict the mechanical degradation of a sisal fiber reinforced benzylated wood (Sample 2) subject to constrained swelling. By means of those five material parameters for the I_5-dependent model, given in (43), we study the modulus loss for free swelling ($\bar{\boldsymbol{C}} = \boldsymbol{I}$) and constrained swelling ($\bar{\boldsymbol{C}} \neq \boldsymbol{I}$). Here we consider two cases of constrained swelling: (1) $\bar{\lambda}_1 = 1/(0.95)^2$, $\bar{\lambda}_2 = \bar{\lambda}_3 = 0.95$, and (2) $\bar{\lambda}_1 = 1/(1.05)^2$, $\bar{\lambda}_2 = \bar{\lambda}_3 = 1.05$, compared with the case of free swelling ($\bar{\lambda}_1 = \bar{\lambda}_2 = \bar{\lambda}_3 = 1$).

In terms of (29), (31) and (36), the evolutions of the modulus E_1 under the case of free swelling and two cases of constrained swelling are calculated based, respectively, on the I_5-dependent model and the I_4-dependent model, as shown in Figure 3. The material parameters used for the I_4-dependent model are taken directly from [Pan and Zhong 2014b]: the shear modulus $G = 0.13$ GPa, the relative stiffness $\gamma = 70.13$, the maximum degradation parameters of matrix $\bar{\beta}_1 = 70.11$ and fiber $\beta_2 = 0.56$, and the evolution speed parameter $D = 1.28 \times 10^5\,\text{Nsm}^{-2}$. It can be seen from Figure 3 that the modulus E_1 degrades with time due to the moisture absorption, for both free and constrained swelling. At the beginning the modulus degrades rapidly and gradually slows down. For free swelling, the predictions from the I_5-dependent

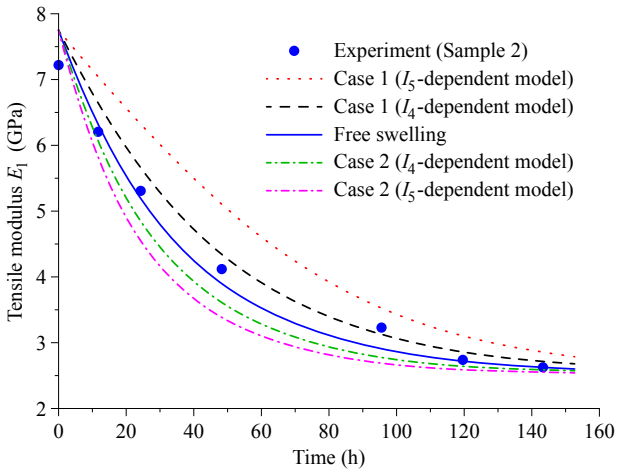

Figure 3. The evolution of the tensile modulus E_1 predicted from the I_5-dependent model and the I_4-dependent model in the cases of free swelling and constrained swelling.

model and the I_4-dependent model give identical results and match well with experimental data obtained by Lu et al. [2003]. However, in the case of constrained swelling, a remarkable difference can be found between the curves obtained from these two models. Compared to free swelling ($\bar{\lambda}_1 = \bar{\lambda}_2 = \bar{\lambda}_3 = 1$), the modulus E_1 degrades more slowly for a tensile compressive elastic deformation along the fiber direction (Case 1 with $\bar{\lambda}_1 = \bar{\lambda}_2^{-2} = \bar{\lambda}_3^{-2} > 1$) and more quickly for a compressive elastic deformation along the fiber direction (Case 2 with $\bar{\lambda}_1 = \bar{\lambda}_2^{-2} = \bar{\lambda}_3^{-2} < 1$). This prediction reveals that the mechanical degradation of NFRCMs can be largely influenced by different constraints during swelling. It is interesting to see that the I_5-dependent model predicts a smaller modulus loss for Case 1 and a bigger modulus loss for Case 2, compared to the I_4-dependent model.

5. Conclusions

This paper establishes a constitutive model of unidirectional natural fiber reinforced composites subjected to mechanical loading and moisture absorption. An internal variable is introduced to consider irreversible energy dissipation by moisture absorption, and is incorporated in the Helmholtz free energy, which can be decomposed into the isotropic matrix part, the reinforcing part and the volume expansion part. With specialized free energies, the degradations of elastic modulus are predicted for unidirectional natural fiber reinforced composites in the cases of free swelling and constrained swelling based on the I_4-dependent model and the I_5-dependent model for reinforcing energy. It is found that these two models give results that are identical in the case of free swelling and different in the case of constrained swelling. However, the predictions from these two models reveal a consistent conclusion that the mechanical degradation of NFRCMs largely depends on the external constraints during the swelling process. This may provide an effective approach to reduce the mechanical degradation induced by moisture absorption.

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Flexural behavior of functionally graded slender beams with complex cross-section
GHOLAMALI SHARIFISHOURABI, AMRAN AYOB, SOHEIL GOHARI, MOHD YAZID BIN YAHYA, SHOKROLLAH SHARIFI and ZORA VRCELJ 1

Response of submerged metallic sandwich structures to underwater impulsive loads
SIDDHARTH AVACHAT and MIN ZHOU 17

Thermal and magnetic effects on the vibration of a cracked nanobeam embedded in an elastic medium
DANILO KARLIČIĆ, DRAGAN JOVANOVIĆ, PREDRAG KOZIĆ and MILAN CAJIĆ 43

Contours for planar cracks growing in three dimensions
LOUIS MILTON BROCK 63

Mechanical degradation of natural fiber reinforced composite materials under constrained swelling
YIHUI PAN and ZHENG ZHONG 79

On the occurrence of lumped forces at corners in classical plate theories: a physically based interpretation
LAURA GALUPPI and GIANNI ROYER-CARFAGNI 93