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## Modeling of the mechanical degradation induced by moisture absorption in short natural fiber reinforced composites

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#### ABSTRACT

This paper develops a constitutive model of short natural fiber reinforced composites (SNFRCs), with the consideration of nonlinear constitutive relation and large deformation induced by moisture absorption. This model accounts for the processes of moisture absorption and mechanical degradation in SNFRCs by a single internal variable, since these two thermodynamic processes are found correlated with each other. This internal variable is introduced to modify the Neo-Hookean model, from which the evolution equations of these two processes are derived by specifying a suitable energy dissipation function. Both the moisture absorption and the mechanical degradation are shown sensitive to the fiber content and could be enlarged with exposure time in a humid environment. Theoretical predictions of the evolution of the internal variable are compared with experimental results of short sisal fiber reinforced polypropylene and it is shown a good agreement between them. Moreover, this model is used to give a reasonable prediction of the degraded Young's modulus of SNFRCs.

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#### 1. Introduction

Short natural fiber reinforced composites (SNFRCs) have gained increasing interests in the past decades due to their exceptional advantages over traditional man-made fiber reinforced composites [1]. Being biodegradable, renewable, environmentally friendly and with high specific strength and modulus, SNFRCs have already been used in aerospace, leisure, construction, sport, packaging and automotive industries [2,3]. However, natural fibers have undesirable hydroscopicity which leads to adverse effects on mechanical prosperities and long-term performances of SNFRCs [4,5]. Coupling agents, compatibilizers and other chemical modifications have been used to improve the moisture resistance of SNFRCs [6,7]. Therefore, moisture absorption and mechanical degradation are considered as two major concerns of SNFRCs especially for their outdoor applications. Numerous efforts have been devoted to this field and some key points are emphasized below.

Firstly, fiber content is a key factor that affects the capability of moisture absorption in SNFRCs. Experiments demonstrated that the amount of water uptake by SNFRCs is elevated with the increase of fiber content [3,5]. George et al. [8] further found that the amount of moisture absorption increases almost linearly with the fiber content. The traditional diffusion theory based on Fick

law is often employed to study the moisture absorption [4,9–11]; however, it fails to consider the effect of fiber content on the moisture absorption of SNFRCs since the fiber content is not explicitly taken into account [12] in the model.

Secondly, the degradation of mechanical prosperities of SNFRCs is an irreversible process with energy dissipation. Sydenstricker et al. [13] studied the interface damage after moisture absorption through fiber pulling-out experiments. Espert et al. [14] found that interface debonding is induced by water transporting into gaps and flaws on the interfaces between the fibers and the matrices, and then concluded that the mechanical degradation of SNFRCs is mainly due to such irreversible energy dissipation processes as interface debonding and fiber degradation. Hu et al. [15] observed under scanning electron microscope that large amounts of microcracks generate on the interfaces and the reserve of water in the crack favors the interface debonding. In addition, the fiber degradation is contributed to the hydrophilicity of natural fibers, which leads to the diffusion of water molecules into micropores between polymer chains of natural fibers and induces the mechanical degradation of fibers themselves.

Thirdly, moisture absorption and mechanical degradation are inherently correlated. However, most available works treated them as two separate processes. In many cases, an empirical or semiempirical formula is used to describe the kinetic evolution of mechanical properties while Fick law is employed to model the kinetic absorption process, in spite of the fact that Fick law is







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invalid in the last stage of moisture absorption [14]. In fact the mechanical degradation depends on the water content absorbed by SNFRCs. To our knowledge, rather few researches correlated them together.

In the present work, we propose a nonlinear constitutive model with the consideration of large deformation induced by moisture absorption. An internal variable is introduced to describe both the kinetic process of moisture absorption and the evolution of mechanical degradation. A quantity relationship between the moisture absorption and the mechanical degradation of SNFRCs is established based on the non-equilibrium thermodynamics.

This paper is organized as follows. A general constitutive model considering moisture absorption in SNFRCs is proposed in Section 2. Then, in Section 3, we particularize several specific forms of the Helmholtz free energy and the energy dissipation function. In Section 4, the theoretical predictions are compared with the experimental results of short sisal fiber reinforced composites. Finally, in Section 5, we make conclusions.

#### 2. Constitutive model

#### 2.1. Generalities

The SNFRC can be treated as a homogeneous isotropic continuum if the short natural fibers are assumed to be dispersed homogeneously and oriented randomly in a polymeric matrix, allowing the water to flow in and out. We choose the virgin dry, stress-free state as the reference configuration. The material point labeled by a position vector **X** in the reference configuration occupies a new position **x** in the current configuration, when the SNFRC deforms in response to the moisture absorption and mechanical loadings. The deformation gradient is defined as  $\mathbf{F} = \partial \mathbf{x}/\partial \mathbf{X}$ , from which all other deformation quantities can be obtained. For example, the right and the left Cauchy-Green deformation tensors are defined as  $\mathbf{C} = \mathbf{F}^T \cdot \mathbf{F}$  and  $\mathbf{b} = \mathbf{F} \cdot \mathbf{F}^T$ , and the volume ratio is given as  $J = \det \mathbf{F}$ , in which the superscript 'T' and the notation 'det' denote respectively the transpose and the determinant of a tensor.

Both moisture absorption and mechanical loadings will cause the deformation of the SNFRC. We consider that the moisture absorption only induces the volumetric swelling deformation, and the isochoric deformation is purely due to the mechanical loading which corresponds to the incompressibility assumption for incompressibile matrix composites [16]. Fig. 1 illustrates the corresponding configurations of the volumetric and the isochoric deformations. Based on a multiplicative decomposition, those deformation tensors can be decomposed into two deformation modes, as follows

$$\mathbf{F} = J^{1/3} \overline{\mathbf{F}} \quad \mathbf{C} = J^{2/3} \overline{\mathbf{C}} \quad \mathbf{b} = J^{2/3} \overline{\mathbf{b}} \tag{1}$$

where  $\overline{\mathbf{F}}$ ,  $\overline{\mathbf{C}}$  and  $\overline{\mathbf{b}}$  are called the modified deformation gradient tensor, the modified right Cauchy-Green deformation tensor and the modified left Cauchy-Green strain tensor, respectively. Furthermore, the first and second principal invariants of modified left Cauchy-Green strain tensor  $\overline{\mathbf{b}}$  are derived as

$$\bar{I}_1 = \operatorname{tr} \,\bar{\mathbf{b}} \quad \bar{I}_2 = \frac{1}{2} \left[ \left( \operatorname{tr} \,\bar{\mathbf{b}} \right)^2 - \operatorname{tr}(\bar{\mathbf{b}} \cdot \bar{\mathbf{b}}) \right] \tag{2}$$

where the notation 'tr' denotes the trace of a tensor.

#### 2.2. Water transport

During moisture absorption, the water transports from external humid environment into the SNFRC. To describe this process, we introduce a reference volume element (RVE) dV containing fibers with the volume fraction c, of which the boundary is denoted by the area element dS, and the normal vector is denoted by **N**. If any internal source is absent (without chemical reaction), the mass balance law can be established in the current configuration and then transformed to the referenced configuration as [17]:

$$\frac{\partial C}{\partial t} + \nabla \cdot \mathbf{J} = \mathbf{0} \tag{3}$$

where  $C(\mathbf{X}, t)$  is the nominal concentration of the water (i.e., the molar number of the water molecule) defined in the reference configuration and  $\mathbf{J}(\mathbf{X}, t)$  is the mass flow across the boundary dS.

To correlate the concentration *C* with other thermodynamically irreversible energy dissipation processes caused, for example, by the hydrolysis of natural fibers or the damage of the interfaces between the fibers and the matrix, an internal variable  $\alpha$  is introduced to relate with the swelling extent. During moisture absorption, the SNFRC gradually evolves from the initial un-swollen state to a swollen one. Thereby the internal variable  $\alpha$  varies from 0 to 1, with  $\alpha = 0$  at t = 0 and  $\alpha = 1$  at  $t \to \infty$ , representing the initial state and the equilibrium state, respectively. In another word,  $\alpha(0)$ 



Fig. 1. The illustration of various configurations of the SNFRC upon moisture absorption and mechanical loading.

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