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Local micromechanics of moisture diffusion in fiber reinforced polymer composites

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ABSTRACT

Fiber distribution in a polymer matrix composite plays a key role in moisture transport which in turn affects its long term behavior. In this paper, effect of filament arrangement on moisture diffusion is studied. The emphasis is on the effect of neighborhood filaments on a single filament placed in a polymer matrix. Fickian diffusion has been simulated using finite element method. Several microstructures have been created using variable angular and spatial orientations of the filaments. Their saturation times have been recorded and analyzed. Microstructures for most favorable saturation time have been identified. The results show that the diffusivity of composites can be controlled through careful spatial design of filaments. The observations should serve as a clue for designing optimum filament geometry for impeding moisture ingress and improving durability of composites.

1. Introduction

Life of fiber reinforced polymers (FRP) can be greatly affected by diffusion of moisture and other deleterious agents. The amount of moisture absorbed by the polymer resin is significantly greater than that by the fibers. It was pointed out in the early days of development of FRPs that their moisture and temperature sensitivity are resin dominated and they can lead to a change in failure mode, while fiber dominated properties show very little environmental sensitivity [1]. Moisture distribution inside any composite structure can prove harmful as it leads to altered stress states because of matrix plasticization, degraded interface strength and mechanical properties of the constitutive materials [2,3]. It can also cause stress buildup due to significant mismatch in the moisture induced volumetric expansion between the matrix and the fibers [4]. In a previous investigation the authors found that diffused alkaline products from concrete borne by moisture can attack glass fibers, greatly reducing their strength [5,6]. Such concerns have hindered the wider use of composites in construction industry.

Moisture diffusion is affected by the fiber. Among several notable ways to study the effect of different fiber arrangements, regular fiber arrangements namely the hexagonal and the square are the fundamental models [7–11]. Statistically equivalent representative volume elements (RVE) with random and clustered arrangement have also been attempted [10–13]. However, fiber distribution is often clustered in composites leading to resin rich areas. These areas can be potential nucleation sites of moisture diffusion.

Interfacial degradation due to hygral and thermal effects has been investigated in unit cells with uniformly arranged fibers [14]. Effective diffusivity calculations have been reported [13,15,16]. Effect of fiber distribution was found to be quite significant when fibers are in contact [17]. It is also pointed out that the polymer and the filament develop an interphase that has considerably higher diffusion than the bulk resin [18].

If a diffusion coefficient based on rule of mixture is used, fiber tortuosity is observed to considerably alter the diffusion rate of a composite from Fickian predictions, especially at large moisture concentrations [19]. The effect has been studied by taking different arrays of packing along transverse direction [13,20,21]. Heterogeneity of fiber distribution has been characterized by the coefficient of variation C_v of the center-to-center distances between interacting fibers and correlated with a moisture induced damage parameter [22]. Two dimensional shape and geometrical locations have been shown as the variants for automated clustering classification [23] whereas composite samples of varying strength and type of clustering were analyzed by developing functional shape, size and volume fraction metrics [24,25]. Voronoi cells

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and Dirchlet tessellations have been used for quantitative characterization of different cluster models representing directional fiber–matrix composites [26–28]. In all these investigations a random fiber distribution has been considered.

In this paper, interaction between fiber filaments in moisture diffusion is studied. The effect of tortuosity has been analyzed along transverse direction by considering arbitrary cross-sections with carefully controlled filament distribution. Influence of inter-filament distance, their angles and size of neighborhood has been reported. The results are fundamental building blocks for analyzing the behavior of large fiber topologies. They also indicate basic principles of designing optimum filament geometry for control of moisture ingress.

2. Governing theory and finite element model

For most of the fiber-reinforced polymer composites, diffusivity of the fiber is negligible as compared to that of the polymer matrix. Hence, it is entirely appropriate to consider moisture transport occurring only in the matrix. The majority of the models for diffusion kinetics in composite materials [2,17,18,22,29–33] are based on Fick's law that states moisture sorption is a diffusion mechanism at a constant rate and constant solubility. Fick's second law can be used to predict the change of mass concentration (C) of a diffusing material in time and space for a given diffusivity (D) of the medium and flux vector ($\vec{J}$). The governing equation for mass diffusion is:

$$\partial C/\partial t + \nabla\cdot\vec{J} = 0 \tag{1}$$

where ∇ is the generalized gradient operator.

When Eq. (1)is applied to inhomogeneous fiber–matrix regions, mass concentration C is not continuous across the interface between two different materials [2,34]. To overcome this problem, the concept of normalized moisture concentration ϕ is used for composite material systems to maintain the interfacial continuity.

$$\phi = C/C^\infty \tag{2}$$

Here C^∞ is the saturation mass concentration of the diffusing material within a particular material. Fick's first law combined with Eqs. (1) and (2)can be solved by implicit finite element scheme with the normalized concentration as basic field variable. For a volume V whose surface is S, integration by parts of Eq. (1)and substituting Eq. (2)results in weak form equation [35]

$$\int_V (\delta\phi C^\infty (d\phi/dt) + \nabla(\delta\phi)\cdot C^\infty D\cdot\nabla\phi)dV = -\int_S \delta\phi\vec{J}dS \tag{3}$$

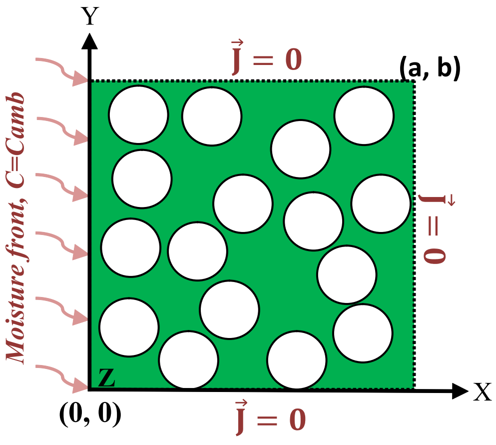

Fig. 1. Present diffusion problem.

The present FE formulation models both filament and resin explicitly. Thus, the resin porosity due to filaments is automatically accounted for [2,15,17,18,21,22,33].

Fig. 1 is an illustration of a cross-section of a uni-directional composite that is studied here. The left edge is exposed to a moist environment of specified concentration. At time $t = 0$ the entire representative model including its other three edges had zero moisture content, The boundary conditions for moisture exposed left edge are:

$$C = 0 \quad (0 < x < a,\ \forall t = 0)$$

$$C = C_{amb} \quad (x = 0,\ \forall t > 0) \tag{4}$$

The rest of edges in Fig. 1 are impermeable boundaries with no moisture flux entering or leaving them for the entire duration, the boundary conditions of these edges are:

$$\vec{J} = 0 \quad (x = a,\ y = 0,\ y = b,\ \forall t \geq 0) \tag{5}$$

Using these boundary conditions diffusion is simulated through a two-dimensional lamina representing transverse section of a reinforced composite with one of its edges suddenly exposed from dry state to the ambient moisture (C_{amb}) that is maintained throughout the time of diffusion. The assumptions are:

1. The moisture diffuses through the gaps in polymer structure following Fickian diffusion law.
2. The mass transfer in the composite is due to exposure to the humid environment that results in concentration gradient across the plane.
3. The diffusivity of the fibre and the matrix remains unaffected during the moisture diffusion process.
4. The maximum moisture concentration in the entire domain does not exceed the concentration at the exposed edge (*saturation* concentration).

Moisture diffusion is modeled by Fickian diffusion based FE code ABAQUS [36]. All models of this work were meshed using quad-dominated (4-noded DC2D4) elements. One of the challenges of using a microstructure-based approach is that a large number of elements and a highly refined mesh are required to conform to the heterogeneous nature of the microstructure. A convergence study is carried out to determine the optimum mesh size and time step. Considering a typical filament radius (R) of 9 μm, approximately 8000 elements were used to mesh most of the geometric models. Each node has a single degree of freedom of moisture concentration. Properties used are listed in Table 1.

In transient mass diffusion analysis with second order elements there is a relationship between the minimum usable time step and the element size. A simple guideline to get the solution convergence is [34]:

$$\Delta t \geq \Delta l^2/6D \tag{6}$$

where Δt is the time increment, D is the diffusivity, and Δl is a typical element dimension (such as the length of a side of an element). If time increments smaller than this value are used, spurious

Table 1
Material properties of fiber and epoxy [21].

Property	Fiber	Matrix
Young's modulus (E, transverse)	20 GPa	4.14 GPa
Poisson's ratio (ν)	0.33	0.36
Density (ρ)	1780 kg/m^3	1310 kg/m^3
Moisture diffusivity (D_m, at $C = 1.48\%$, $R_H = 85\%$)	0	0.805×10^{-14} m^2/s
Solubility(S)	0	1