Research Article

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Modified Halpin–Tsai equation for predicting interfacial effect in water diffusion process

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Abstract: Interfacial degradation is the main reason for deterioration of mechanical properties of composites in hydrothermal environments. In this study, the effect of the interphase on water diffusion in two types of unidirectional continuous carbon fiber-reinforced polyamide 6 (CF/PA6) composites is investigated through experimental measurements, theoretical analysis, and numerical simulation. The moisture diffusion coefficient of composite at different environmental temperatures is characterized by water immersion tests for analyzing the barrier and accelerating effects of the interphase layer. Based on the experimental results, the three-phase Halpin–Tsai model is derived and validated, and then the critical diffusivity is obtained to quantify the interfacial effect during the diffusion process. To further validate the present three-phase Halpin–Tsai model, the stable and transient finite element models of moisture diffusion are developed. It is found that the critical diffusivity coefficient of the interphase for the CF/PA6 composite system is 7.31 times higher than that of the matrix.

Keywords: continuous carbon fiber reinforced-polyamide 6 composites, water diffusion, Halpin–Tsai model, interfacial effect

1 Introduction

Given the demand for lightweight devices and equipment, continuous carbon fiber-reinforced polyamide 6 (CF/PA6) composites are more promising than the thermoset composites for automotive industry applications because of their shorter processing times, lower manufacturing costs, and improved recyclability [1–3]. However, CF/PA6 composites may experience hydrothermal aging-induced weakening of the interphase layer located between the carbon fiber and the polyamide 6 matrix, which can deteriorate the composites' mechanical properties and considerably limit their service life [4–7]. Therefore, a qualitative understanding of the effect of interphase layer on the effective moisture diffusion property of composite is crucial for improving the aging resistance of CF/PA6 composites.

The interphase between the matrix and the reinforced fiber plays a vital role in load transfer efficiency and the ultimate mechanical properties. As shown in Figure 1, an obvious fiber/matrix interphase exists in the CF/PA6 composite with a thickness of 400 nm measured by *in situ* nanomechanical testing [8]. The degradation of the interface due to the water absorption in the hydrothermal environment can decrease the mechanical properties of composites. Because the physical properties of the interphase differ from those of the matrix and the fiber, the water diffusion parameters of the composites are significantly modified by the presence of the interphase [9,10]. For example, Sang et al. [11,12] experimentally investigated the moisture absorption properties of PA6 resin and CF/PA6 composites in a hydrothermal environment and reported that the water diffusivity of the composites was smaller than that of the neat resin. A similar water diffusion phenomenon was reported by Ksouri et al. for PA6 composites reinforced with 30% glass fiber [13,14], which confirmed the barrier effect of the interface. However, Yang et al. [15] reported that moisture diffusion occurred more rapidly in composites than in pure resins. From the aforementioned investigations, it can be hypothesized that the fiber/matrix

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Figure 1: Typical three-color gradient dynamic mechanical image of the CF/PA6 composite: (a) storage modulus map and (b) interphase dimension and shape [8].

interface could offer a preferential path and thus accelerate moisture diffusion of composites.

Moreover, the effect of the interface on water diffusion was extensively studied using the finite element method (FEM) [16,17] and the relationship between the matrix and interface parameter can be numerically determined. In FEM simulation, the representative volume element (RVE) model and the full composite model were, respectively, adopted for computation [18]. It was reported that the diffusivity of the interface was five times that of the matrix ($D_{\text{interface}} = 5D_{\text{matrix}}$) for the glass fiber/epoxy composite [7,19]. In contrast, Pan et al. [20] found that the diffusivity of the interface ($D_{\text{interface}} = 0.25D_{\text{matrix}}$) was much lower than that of the matrix in the carbon fiber/epoxy composite system. Despite many published studies on the hygroscopic properties of composite materials, the lack of information regarding a systematic analysis of the interfacial effect in the water diffusion process represents an essential drawback for industrial and scientific fields. The diffusivity of the interface varies significantly for different composite systems, and simultaneously the interfacial mechanism in the water diffusion process still remains unclear. Besides, the Halpin–Tsai equation is extensively used in predicting the transverse diffusivities of unidirectional composites [21]. However, the classical Halpin–Tsai equation does not consider the interface [22]. Thus, to explore the barrier or accelerating effects of the interface in the water diffusion process, a modified theoretical model is needed for providing a quick guidance of composite property analysis in engineering.

The aim of this work is to derive a modified Halpin–Tsai model, which is validated by experimental and numerical predictions. To do this, the moisture diffusion in the

CF/PA6 composites and the PA6 resin is comparatively analyzed, and the effect of the interface layer on the moisture diffusivity is evaluated using gravimetric experiments. Then a three-phase modified equation is derived using the Halpin–Tsai law, and the critical diffusivity is utilized to determine whether the interface exhibits barrier or accelerating effects during the diffusion process. Lastly, the stable and transient moisture diffusion processes are simulated using FEM analysis to validate the accuracy of the modified Halpin–Tsai equation.

2 The modified Halpin–Tsai model

2.1 The classical Halpin–Tsai model

The water diffusion behaviors of composites can be described using two parameters, namely, the maximum moisture content (M_m) and the diffusivity coefficients (D). M_m is defined as the amount of moisture present in a saturated-state composite in a hydrothermal environment. The diffusivities of the composites represent the water diffusion rates, which can be divided into the longitudinal diffusivity (D_l) and the transverse diffusivity (D_t). D_l can be estimated using a rule-of-mixture approach. However, the prediction of the transverse parameter D_t is more complex, as it strongly depends on the microstructure of composite, including properties of the individual constituents, their volume fraction, the fiber arrangement, and the clustering effects of fibers [23–25]. In this aspect,

several analytical models can be exploited to predict D_t of the composite in the hydrothermal environment, including a self-consistent model [26], the Halpin–Tsai equations [21], and the Kalman filter model [27]. Among them, the Halpin–Tsai equation given below is most widely used to predict the transverse water diffusivity in heterogeneous multicomponent composites [21].

$$D_t = D_m \frac{1 + \xi \eta V_f}{1 - \eta V_f}, \quad (1)$$

with

$$\eta = \frac{D_f/D_m - 1}{D_f/D_m + \xi}, \quad (2)$$

where D_t and D_m are the diffusivity of the CF/PA6 composites and the PA6 resin (matrix), respectively, and ξ is a parameter considered equal to 1 in this study for simplification.

Equation (1) can be rewritten as

$$\frac{D_t}{D_m} = \frac{1 - V_f + (1 + V_f)D_f/D_m}{1 + V_f + (1 - V_f)D_f/D_m}, \quad (3)$$

where D_f and V_f are the water diffusivity and volume fraction of the fiber, respectively.

2.2 The modified Halpin–Tsai model

The classical Halpin–Tsai equation only provides approximations of the transverse diffusivities of the two-phase composites, because it does not consider the matrix/fiber interphase. Comparatively, in this study, we aim to provide a modified theoretical model to represent the effect of the interphase on the water-diffusion parameters. The continuous moisture contents at the boundaries between the fiber, the interphase, and the matrix are also considered in this study.

To introduce the interfacial parameters to the Halpin–Tsai equation, the three-phase composite model can be divided into two two-phase composite models (submodel 1 and submodel 2). In each submodel, D_t^i and D_m^i are the diffusivities of the composite and the matrix phase, respectively, where the superscript i ($i = 1, 2$) denotes the label number of the submodel.

In submodel 1, the mixture phase consisting of the fiber and the interphase is embedded into the matrix. The mixture phase is the “new” fiber phase, while the matrix remains unchanged. Substituting the parameters of the mixture and matrix phases into equation (3), we have

$$\frac{D_t^1}{D_m^1} = \frac{1 - V_{\text{mix}} + (1 + V_{\text{mix}})D_{\text{mix}}/D_m}{1 + V_{\text{mix}} + (1 - V_{\text{mix}})D_{\text{mix}}/D_m}, \quad (4)$$

where D_t^1 , D_m^1 , and D_{mix} are the diffusivities of the composite, the matrix, and the mixture phase in submodel 1, respectively, and V_{mix} is the volume fraction of the mixture phase.

The volume fraction of the mixture phase and the matrix meet with $V_{\text{mix}} + V_m = 1$. Therefore, equation (4) is written as

$$\frac{D_t^1}{D_m^1} = \frac{D_t}{D_m} = \frac{V_m + (2 - V_m)D_{\text{mix}}/D_m}{2 - V_m + V_m D_{\text{mix}}/D_m}. \quad (5)$$

Submodel 2 comprises the fiber and the interphase, and the fiber is embedded in the interphase. In submodel 2, the interphase is regarded as the “new” matrix, and the fiber remains unchanged. Similarly, by substituting the parameters of the fiber and the interface into equation (3), we get

$$\frac{D_t^2}{D_m^2} = \frac{1 - V_f + (1 + V_f)D_f/D_i}{1 + V_f + (1 - V_f)D_f/D_i}, \quad (6)$$

where D_t^2 and D_m^2 are the diffusivities of the composite and the matrix, respectively, in submodel 2.

The volume fraction of the fiber and the interphase follows $V_f + V_i = 1$. Therefore, equation (6) is written as

$$\frac{D_t^2}{D_m^2} = \frac{D_{\text{mix}}}{D_i} = \frac{V_i + (V_i + 2V_f)D_f/D_i}{V_i + 2V_f + V_i D_f/D_i}, \quad (7)$$

in which, $\frac{D_{\text{mix}}}{D_m}$ is expressed as

$$\frac{D_{\text{mix}}}{D_m} = \frac{D_{\text{mix}}}{D_i} \cdot \frac{D_i}{D_m} \quad (8)$$

When equations (7) and (8) are substituted into equation (5), the transverse diffusivities of the three phases in the composites can be expressed as:

$$\frac{D_t}{D_m} = \frac{\beta V_m + \alpha(2 - V_m)D_i/D_m}{\beta(2 - V_m) + \alpha V_m D_i/D_m}, \quad (9)$$

with

$$\begin{aligned} \alpha &= V_i + (V_i + 2V_f)D_f/D_i \\ \beta &= V_i + 2V_f + V_i D_f/D_i \end{aligned} \quad (10)$$

3 Finite element model

To validate the accuracy of the modified Halpin–Tsai equations, the three-phase FEM model is employed to simulate the stable and transient moisture diffusion processes in the CF/PA6 composites. The mass diffusion

analogy available in the Abaqus software is used to simulate water diffusion in the composites. The schematic and boundary conditions are illustrated in Figure 2. Generally, the carbon fibers do not absorb moisture when immersed in water, and their moisture diffusivity is several orders of magnitude smaller than that of the matrix. Therefore, in this study, the carbon fibers are modeled as voids with impermeable boundaries to approximately simulate the barrier effect during water diffusion.

To provide the material parameters for the numerical simulation, two typical composite plates, namely, CF/PA6 (Panex® 35-50K) and CF/PA6 (SYT49S-12K), as well as pure PA6 samples, are subject to moisture absorption testing by immersing samples in distilled water at 30, 50, 70, and 90°C, according to the standard of ASTM D5229. Differences in fiber sizing agents between the two composites yield different effects of the interface on the water diffusion process. The former uses an epoxy sizing agent, while the latter has a polyurethane sizing agent. Water absorption curves are plotted by recording the masses of the samples over time. The diffusivity along the thickness direction (D_z) is obtained from the slope of the initial linear part of the moisture absorption curve and calculated using two points on the linear part of the curve at times t_1 and t_2 :

$$D_z = \pi \left(\frac{h}{4M_m} \right)^2 \left(\frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}} \right)^2 \quad (11)$$

Moreover, for the sake of computational convenience, we only simulate the moisture diffusion process at 70°C and determine the diffusivity of the interface by the inverse method [20]. The diffusivity ratio between the interface and the matrix is utilized as the input parameter in the other three aging conditions, and the simulation

results are compared with the experimental data and analytical results.

Practically, the carbon fibers distribute randomly in the matrix; therefore, the finite element models including randomly distributed fibers are developed in this study for computation. A random distribution of fibers in the RVE is automatically generated using the random sequential adsorption-based algorithm programmed in MATLAB [28,29], and a geometric model is created using AUTOCAD software. For instance, the RVE model containing 100 randomly distributed fibers of 7 μm in diameter is generated for steady analysis. The thickness of the interphase is defined as 400 nm [8]. As indicated in Figure 3, the water diffusion model of the three-phase composites with three different values of V_f is built for steady-state analysis. To validate the numerical and theoretical predictions of diffusivity of composite, the experimental results related to the case $V_f = 26\%$ at different environmental temperatures are taken as reference for comparison. The V_f values of 20 and 40% are selected for comparison with the experimental V_f value (26%) to analyze the relationship between the fiber volume fraction and the interface effect. The models are meshed using four-node elements (DC2D4) for analysis.

Moreover, to analyze the saturation time and obtain the absorbed moisture curve, a modified computational model is developed, as presented in Figure 4. We use this numerical simulation model to study transient water diffusion in the direction of the thickness of the sample. As the sample thickness, rather than the length, significantly affects the results, the real thicknesses of the samples are used to improve the accuracy of the calculations, and the length of the model is set to 30 μm to increase the efficiency of the calculation. Totally, 286,425 elements (CPS4R) and 303,073 nodes are used in this transient analysis.

Figure 2: Modeling of water diffusion in unidirectional composite.

Figure 3: Water diffusion model for three-phase composites with different fiber volume fractions : (a) 20%, (b) 26%, and (c) 40%.

4 Results and discussion

4.1 Validation of the modified Halpin–Tsai equation

To validate the accuracy of the modified Halpin–Tsai equation (equation (9)), the water diffusivities are obtained using the classical Halpin–Tsai equation (equation (3)), the modified model equation (9) and the FEM, and then compared to the experimental data. As shown in Table 1, the predictions from equation (3) for the CF/PA6 (Panex® 35-50K) composites have obvious discrepancy with the experimental data and the largest relative error (RE%) between them is 28%. In contrast, the values of water diffusivities calculated using equation (9) are closer to the experimental data, and the largest relative error is 19%. In the CF/PA6 (SYT49S-12K) composites corresponding

to the case where D_i exceeds the critical diffusivity, the relative error corresponding to equation (9) is less than 5%, while equation (3) gives the error exceeding 50%, as summarized in Table 2. In fact, the water diffusivities of composites calculated by equation (3) always are lower than those of the matrix. This is attributed to $\frac{1-V_f}{1+V_f} < 1$. Thus, equation (3) can only be used to predict the water diffusivity in cases where D_i is lower than the critical diffusivity, and the accuracy of prediction is relatively low. In contrast, the experimental data agree well with the predicted results determined from equation (9) in all cases. Therefore, the modified Halpin–Tsai model is effective.

The transient mass diffusion is simulated to further validate the accuracy of the modified Halpin–Tsai equation. The mass concentration (CONC) of unique nodes at given time increments is the output, and the mean value from the sample nodes is used as the absorbed water

Figure 4: Schematic of the numerical model used to simulate the transient moisture diffusion process.

Table 1: Comparison of water diffusivities obtained by experimental data, the classical Halpin–Tsai equation (equation (3)), and the modified Halpin–Tsai equation (equation (9)) in the CF/PA6 (Panex® 35-50K) composites. Here RE (%) denotes the relative error

Aging temperature (°C)	Moisture diffusivities ($\times 10^{-12} \text{ m}^2/\text{s}$)					
	Experiment	Equation (3)	RE (%)	Equation (9)	RE (%)	FEM
30	0.41	0.47	16	0.45	9	0.46
50	3.06	2.72	11	3.01	1	3.02
70	9.57	8.56	11	9.47	1	9.46
90	34.13	24.71	28	28.73	15	27.37

content. The water absorption curves of the CF/PA6 composites are plotted using the results obtained from the numerical model (see Figure 5). The curves plotted by the modified Halpin–Tsai equation agree well with the experimental curves for both of composites. However, the curves plotted by the classical Halpin–Tsai equation show greater deviation to the experimental curves, especially for the CF/PA6 (SYT49S-12K) composites.

4.2 Interfacial effect

Figure 6 illustrates the experimental water uptake curves of the PA6 and CF/PA6 samples in a hydrothermal environment. In addition, Table 3 summarizes the water diffusion parameters, namely, M_m and D_z . The results reveal that the PA6 resin absorbs more water than both types of CF/PA6 composites under the same aging conditions: the

Table 2: Comparison of water diffusivities obtained by experimental data, the classical Halpin–Tsai equation (equation (3)), and the modified Halpin–Tsai equation (equation (9)) in the CF/PA6 (SYT49S-12K) composites. Here RE (%) denotes the relative error

Aging temperature (°C)	Moisture diffusivities ($\times 10^{-12} \text{ m}^2/\text{s}$)					
	Experiment	Equation (3)	RE (%)	Equation (9)	RE (%)	FEM
30	1.02	0.48	53	1.05	3	1.06
50	5.83	2.73	53	6.03	3	5.98
70	19.09	8.56	55	18.96	0.7	19.36
90	54.63	24.71	54	54.71	0.2	54.26

Figure 5: Moisture absorption curves of (a) CF/PA6 composites (Panex® 35-50K) composites, and (b) CF/PA6 (SYT49S-12K) composites obtained using experimental data, modified Halpin–Tsai equation, and Halpin–Tsai equation.

Figure 6: Water absorption curves of (a) PA6 resin and CF/PA6 (Panex® 35-50K) composites and (b) PA6 resin and CF/PA6 (SYT49S-12K) composites at 30, 50, 70, and 90 °C.

Table 3: Water absorption parameters for different specimens

	Water temperature (°C)	M_m (%)	D_z ($\times 10^{-6}$ mm ² /s)
PA6	30	10.06 ± 0.11	0.81 ± 0.05
	50	9.40 ± 0.03	4.64 ± 0.40
	70	9.37 ± 0.30	14.58 ± 0.99
	90	9.21 ± 0.05	42.08 ± 2.07
CF/PA6 (Panex® 35-50K)	30	4.85 ± 0.23	0.41 ± 0.01
	50	5.26 ± 0.30	3.06 ± 0.47
	70	5.10 ± 0.15	9.57 ± 0.71
	90	5.43 ± 0.14	34.13 ± 1.82
CF/PA6 (SYT49S-12K)	30	5.07 ± 0.09	1.02 ± 0.23
	50	5.50 ± 0.13	5.83 ± 1.95
	70	5.54 ± 0.14	19.09 ± 1.51
	90	5.41 ± 0.08	54.63 ± 4.50

M_m values of the PA6 resin exceed 9%, whereas those of the CF/PA6 composites range from 4.85 to 5.54%. These results indicate that the polymer is the major contributor

Figure 7: Evolution of transverse diffusivity of composites (D_t) vs. the ratio of the diffusivities of the interphase and matrix (D_i/D_m). Here V_f denotes the volume fraction of fiber and FEM denotes the finite element method used to generate the data set.

Table 4: Moisture diffusivities of CF/PA6 laminate interfaces (D_i) determined using FEM and the modified Halpin-Tsai equation. Here V_f , N , and D_m denote the volume fraction of the fibers, number of fibers, and diffusivity of the matrix, respectively

V_f (%)	Method	$D_i = N \times D_m$ ($N = 0, 2, 4, \dots, 20$)										
		0	2	4	6	8	10	12	14	16	18	20
20	Equation (9)	8.48	10.78	12.53	13.89	14.99	15.89	16.64	17.28	17.8	18.3	18.72
	FEM	8.74	11.1	12.81	14.13	15.2	16.07	16.81	17.43	17.97	18.44	18.85
26	Equation (9)	7.11	9.83	11.97	13.69	15.11	16.3	17.32	18.19	18.95	19.61	20.2
	FEM	6.83	9.8	11.97	13.69	15.09	16.27	17.27	18.14	18.89	19.56	20.15
40	Equation (9)	4.49	7.86	10.75	13.24	15.41	17.32	19.02	20.53	21.89	23.12	24.24
	FEM	4.0	7.8	10.5	12.7	14.68	16.38	17.9	19.27	20.5	21.63	22.66

Figure 8: Evolution of transverse diffusivity of composites (D_t) vs the ratio of the diffusivities of the interphase and matrix (D_i/D_m), where interphase is (a) 200 nm, (b) 400 nm, and (c) 800 nm.

to M_m , which is attributed to the amorphous phase of the PA6 resin [30]; moreover, M_m depends on the mass content of PA6 resin of the CF/PA6 composites.

Figure 9: Water uptake profiles as a function of immersion time for (a) PA6 resin, (b) CF/PA6 (Panex® 35-50K) composite, and (c) CF/PA6 (SYT49S-12K) composite; the samples are immersed in distilled water at 70°C.

As the aging temperature of all samples is increased, their D_z values are also increased and maximized at 90°C. This increase is typically associated with the activity of the water molecules in the amorphous regions of the polymer and at the matrix/fiber interphase, and also increased the mobility of the water molecules at higher temperature. The D_z values of the CF/PA6 (Panex® 35-50K) and CF/PA6 (SYT49S-12K) composites are smaller and larger, respectively, than those of the PA6 resin, which can be attributed to the differences in the diffusivities of the matrix/fiber interphases of the two composites. The interphase of the former composite presents a barrier effect

on the water diffusion process, with a diffusivity lower than the critical diffusivity. Conversely, the interphase of the latter composite can provide fast channels for water diffusion, and therefore, its diffusivity is higher than the critical diffusivity.

4.3 Critical diffusivity

To analyze the differences in the water absorption curves of the two types of CF/PA6 composites and that of the PA6 resin, the critical diffusivity of the interphase layer is introduced. The critical diffusivity is obtained at the intersection of the water absorption curves of the PA6 resin and those of the two types of CF/PA6 composites, as depicted in Figure 7. The D_i values of the CF/PA6 composites are determined using the inverse method.

First, as shown in Table 4, the D_i/D_m ratio is assumed to change from 0 to 20, with the interval of two. The diffusivities of the RVE composite model for different V_f values and the D_i/D_m ratios are determined using FEM and the modified Halpin–Tsai equation (equation (9)). Second, the D_t vs D_i/D_m curve is plotted, as illustrated in Figure 7. The x coordinate of the intersection of the three water absorption curves is the critical diffusivity of the interphase layer, which is determined to be 7.31. The D_i/D_m ratios of the CF/PA6 (Panex® 35-50K) and CF/PA6 (SYT49S-12K) composites are 1.7 and 16.01, respectively, as obtained from the intersection of the experimental D_t data curve with the water absorption curve of the composites with $V_f = 26\%$.

As shown in Figure 7, when D_i is higher than the critical value, the interphase has an accelerating effect on the water diffusion process and D_t increases as V_f increases. However, if D_i is lower than the critical value, the interface presents a barrier effect on the water diffusion process and D_t decreases as V_f increases. Moreover, the critical diffusivity is independent of V_f but dependent on the thickness of the interphase. As shown in Figure 8, the critical values of D_i/D_m ratios for the 200, 400, and 800 nm interface are 17.94, 7.18, and 4.94, respectively. The critical values decrease with the increasing thickness.

In fact, the critical diffusivity is defined as the diffusivity of the interphase when $D_{\text{mix}} = D_m$. In the mixture phase, the fibers hardly absorb water and the water diffusion process mainly occurs in the interphase. A larger interphase thickness means that the volume fraction of the interphase is larger in the mixture phase. Therefore, a smaller diffusivity is needed to ensure that the diffusivity

of the mixture phase is equal to that of the matrix. In conclusion, the interphase thickness is inversely correlated with the critical diffusivity value.

The time evolution of the typical water concentration distribution along the thickness of the specimens is presented in Figure 9. The largest amount of water is first absorbed at the upper edge of the model (Figure 4); furthermore, the amount of absorbed water continuously decreased toward the core of the samples owing to the higher concentration difference (100% moisture) at the upper edge. Moreover, the saturation times of the samples decrease as follows: CF/PA6 (Panex® 35-50K) (91.8 h) > PA6 (82.48 h) > CF/PA6 (SYT49S-12K) (46.8 h). These results indicate that the interphases of the CF/PA6 (Panex® 35-50K) and CF/PA6 (SYT49S-12K) composites presented barrier and accelerating effects, respectively.

5 Conclusion

The water absorption behavior of the PA6 resin and two CF/PA6 composites are studied by conducting gravimetric experiments at 30, 50, 70, and 90°C for providing reference to validate the modified theoretical model. Then the stable and transient diffusion processes are simulated using FEM analysis to further validate the accuracy of the modified Halpin–Tsai equation. The conclusions drawn from this study are:

- (1) Water diffusion is significantly affected by the presence of the interphase, which presents a barrier effect or accelerating effect on the water diffusion process.
- (2) The critical diffusivities determined by the modified Halpin–Tsai equation can effectively predict the interphase effect. For the current CF/PA6 system in this study, the critical diffusivity of the interphase is 7.31 times higher than that of the matrix.
- (3) The critical diffusivity of the interphase is independent of V_f , but it is inversely correlated with the interface thickness. When D_i exceeds critical value, the interphase presents an accelerating effect on the water diffusion process and D_t increases as V_f increases. However, if D_i is below the critical value, the interphase presents a barrier effect on the water diffusion process and D_t decreases as V_f increases.

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