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DYNAMICS OF MOISTURE DIFFUSION IN EPOXY RESIN: EMPIRICAL AND MODELING INSIGHTS INTO ABSORPTION- DESORPTION BEHAVIOR

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Abstract

Rapid advancements in the composite industry have led to an increase in the adoption of epoxy resins in high-pressure applications in industries such as water transport systems, due to their enhanced mechanical properties. Despite their advantages, studies have proven that water absorption in epoxy tubes can degrade their mechanical properties when subjected to extreme environmental conditions. Herein, this work characterizes the intricate dynamics of moisture diffusion kinetics in the epoxy resin subjected to severe environmental conditions, combining the experimental investigation and the numerical approach. The experimental investigation was conducted by subjecting the epoxy resin samples to cyclic hygrothermal conditions in a Temperature Humidity Test Chamber. Continuous gravimetric measurements were performed every 24 hours to track the diffusion kinetics in two moisture absorption-desorption cycles. It reveals the complex Dual-Fickian behavior in the initial absorption phase, which has transitioned to the Fickian behavior in subsequent cycles. Furthermore, the numerical analysis using ABAQUS has validated the experimental findings, highlighting the need for finer meshes to accurately predict the complex Dual-Fickian behavior in the epoxy resin.

1. Introduction

Epoxy resins have emerged as a popular material in the manufacturing process of composite pipes owing to their enhanced mechanical performances and resistance to severe environmental conditions, making them a preferred choice for high-pressure applications in oil and water transport systems [1]. They are favored over traditional materials such as steel or PVC, mainly due to their usability under high temperatures, cyclic loads, and resilience to corrosion. However, their application has been limited due to the sensitivity to moisture diffusion. Moisture absorption in the epoxy resin has been demonstrated to result in the degradation of the mechanical properties such as the decrease of its glass transition temperature and elastic modulus [1], thereby undermining the structural integrity of the pipe system.

Recent studies [1,2,3] have highlighted the complex moisture diffusion kinetics in the epoxy resin, differentiating between Fickian and non-Fickian behavior [2]. In real cases, the moisture diffusion process within polymers often exhibits two states, mobile and bound water [3]. Moisture diffusion in mobile state occurs when the water molecules in the environment start to migrate toward the voids, pores, or free volume of the polymer network until the saturation. While in the bound state, water molecules remain bonded to the polymer network through hydrogen bonding. This phenomenon of bound state is termed as Type I and Type II bonding. Type I bonding is a straightforward process

involving single hydrogen bonds, whereas Type II bonding is a much more complex process involving multiple hydrogen bonds. This complicated interaction between water molecules and the microscopic structures of the epoxy resin results in the higher activation energy being required to break the Type II hydrogen bonds, finally making the moisture diffusion process irreversible and leading to a more Non-Fickian behavior. The Dual-Fickian model [1,3] is often employed to characterize the Non-Fickian behavior of the epoxy resin and composites.

However, there remains a significant gap between the Non-Fickian theory and its application to numerical modeling in softwares such as ABAQUS and COMSOL. In order to accurately predict the moisture absorption-desorption behavior of the epoxy resin and resin-based composite structures, this study aims to investigate the moisture diffusion behavior of the epoxy resin subjected to cyclic hygrothermal aging conditions experimentally and numerically. The numerical model using Finite Element Analysis (FEA) will be developed to validate the experimental findings and give a deeper understanding of the Fickian and Dual-Fickian behavior of the epoxy resin.

2. Materials and experimental investigation

2.1. Cyclic moisture absorption-desorption tests

The epoxy resin (see Table 1), commonly used in composite structures in water transport applications, was studied in this work. Firstly, the samples were dried in a Binder climatic chamber (model KMF 720) at 70 °C and 7 % R.H., below the glass transition temperature (T_g) of the material, to avoid internal material damage and stabilize the mass distribution through the thickness. Subsequently, the samples were subjected to hygrothermal aging conditions until saturation in an environmental test chamber (EXCAL 5421) at 70 °C and 85 % R.H., followed by subsequent desorption and absorption cycles. Continuous gravimetric measurements were performed every 24 hours during the moisture absorption-desorption cycles, using a precision balance (model MC BA 10) to track the moisture diffusion kinetics. The moisture concentration ($C(t)$) at time t can be calculated as follows:

$$C(t) = \frac{m(t) - m_0}{m_0} \quad (1)$$

where $m(t)$ is the mass of the sample measured at time t , and m_0 is the initial dried mass.

The moisture diffusion kinetics were monitored on three thin square epoxy resin samples (see Figure 1). The 2 mm thickness is much smaller than the 50 mm length and width of the sample, thus ensuring the main unidirectional diffusion along the thickness direction.

Table 1. Parameters of resin samples in cyclic moisture absorption-desorption tests.

Material	Composition	Dimensions (mm ³)
Epoxy resin	epoxy base + hardener	50×50×2.0-2.1

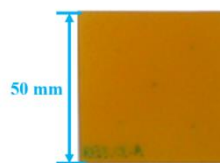


Figure 1. Epoxy resin sample (50×50 mm²).

2.2. Experimental results and discussions

Figure 2 shows the results of cyclic moisture diffusion kinetics of a typical sample (A) among the three observed ones (A, B, C) since the three specimens exhibit a very similar moisture diffusion behavior which can be generalized. First of all, two complete moisture absorption-desorption cycles have been demonstrated, with only the first absorption phase exhibiting the Dual-Fickian behavior. The detailed discussion of each absorption and desorption phase is presented as follows:

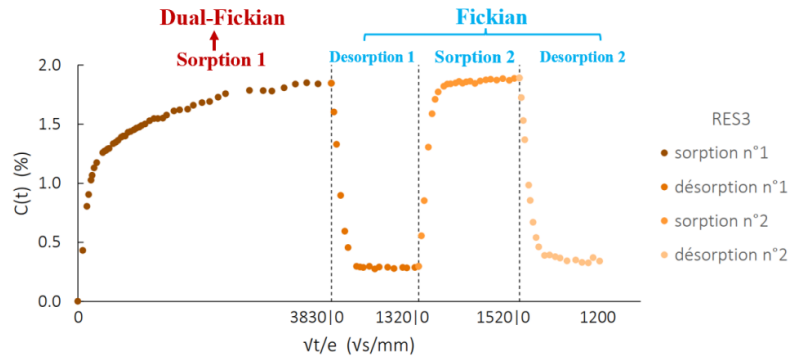


Figure 2. Two complete moisture absorption-desorption cycles of epoxy resin.

- **Sorption 1**

Firstly, the mass change obtained after a period of 23 months at 70 °C and 85 % R.H. is shown in the first absorption phase in Figure 2. It is interesting to find that the moisture diffusion kinetics of Sorption 1 exhibit the more complex Dual-Fickian behavior [3]. Generally, it is the superposition of two Fickian diffusion stages, in contrast to the typical Fickian behavior [1] with only one linear diffusion stage until the final saturation. It should be noted that the epoxy resin samples have reached their maximum absorption capacity after a period of 17 months ($\sqrt{t/e} \approx 3200 \sqrt{s/mm}$), ranging from 1.7 % to 2.0 %.

- **Desorption 1**

After the phase Sorption 1, the specimens were dried continuously at 70° C and 7 % R.H. for 2 months, as presented by the first desorption curve in Figure 2. The moisture content in the epoxy resin stabilizes to a non-zero value after only one week ($\sqrt{t/e} \approx 400 \sqrt{s/mm}$), which ranges from 0.3 % to 0.4 %. This is equal to 20 % of the saturated moisture content reached in Sorption 1. Fickian behavior is found to be the dominating behavior in Desorption 1 curve, compared to Dual-Fickian behavior in Sorption 1.

- **Sorption 2**

Subsequently, the second moisture absorption phase Sorption 2 was carried out over a shorter period of 3.5 months at 70° C and 85 % R.H. The initial moisture concentration at the start of Sorption 2 corresponds to the residual moisture content obtained at the end of the first desorption cycle. Interestingly, the moisture diffusion behavior of the epoxy resin in the second absorption phase corresponds to the Fickian model, contrary to the Dual-Fickian behavior in the first absorption phase. The saturation was reached more quickly within 10 days, compared to the longer 17 months in the first absorption phase. The saturated moisture content (between 1.8 % and 2.0 %) obtained at the end of the phase Sorption 2 was almost the same as that attained in Sorption 1. Furthermore, the saturation in Sorption 2 was reached at approximately $\sqrt{t/e} \approx 400 \sqrt{s/mm}$, which is similar to the phase Desorption 1. It indicates that Sorption 2 and Desorption 1 exhibit similar Fickian diffusion behavior, meaning that the moisture diffusion coefficients in these two phases may be almost the same.

- **Desorption 2**

Similarly, after the phase Sorption 2, the wet samples were dried at 70° C and 7 % R.H until saturation. Finally, the second desorption phase Desorption 2 which exhibits the Fickian behavior over a period of

2.5 months can be observed in Figure 2. The saturated moisture content was also close to that attained at the end of Desorption 1, i.e. between 0.3 % and 0.4 %.

In conclusion, the Dual-Fickian behavior in the first absorption cycle can be attributed to the superposition of two Fickian diffusion stages resulting from the complicated interactions of free and bound water with the polymer network [4]. Whereas, the first desorption phase, the second absorption phase, and the second desorption phase are dominated by the Fickian behavior mainly due to the presence of free water diffusion [5,6]. To better explain the observed moisture diffusion phenomena on the epoxy resin, the following numerical models will be developed to validate the different Non-Fickian and Fickian behavior in the two absorption-desorption cycles.

3. Numerical modeling of cyclic moisture diffusion process in epoxy resin

3.1. Dual-Fickian diffusion theory for epoxy resin

As introduced above, the Dual-Fickian behavior can be adopted to simulate the moisture diffusion process occurring within the resin and composite materials [1,3]. Generally, the Dual-Fickian model assumes that the total moisture concentration $C(t)$ of a sample at time t can be calculated by the superposition of two Fickian stages, as shown by Eq. (2), where $C_1(t)$ represents the moisture concentration due to the first Fickian stage and $C_2(t)$ is the concentration in the second Fickian stage.

$$C(t)=C_1(t)+C_2(t) \quad (2)$$

Hence, it means that the two Fickian diffusion mechanisms work in parallel such that the mass uptake increases continuously until reaching the saturation. If the sample is a thin plate whose thickness is much smaller than the other dimensions, the moisture diffusion would mainly take place in the thickness direction. Thus, the moisture diffusion process in the epoxy resin can be simulated approximately by 1D modeling. The final Dual-Fickian model used can be written as Eq. (3) [7] :

$$C(t)= \sum_{i=\{1,2\}} \left[\left(1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp \left(-D_i \left(\frac{2n+1}{e} \right)^2 \pi^2 t \right) \right) \times C_{Si} \right] \quad (3)$$

Where $C(t)$ is the moisture concentration at time t ; e is the thickness in the diffusion direction.

D_i and C_{Si} ($i=1, 2$) are the moisture diffusion coefficients and saturated concentration in two stages.

The above five parameters mainly determine the moisture diffusion rate and final saturation capacity.

3.2. Dual-Fickian model implemented in ABAQUS

In ABAQUS, the moisture diffusion kinetics within the epoxy resin can be simulated by analogy equating Fourier's law of heat with Fick's second law of diffusion. To implement the Dual-Fickian diffusion model in ABAQUS, the idea is to first perform two parallel Fickian models separately on the epoxy resin, each with its own parameter set : (D_1, C_{s1}) and (D_2, C_{s2}), as shown in Table 2. Then the final moisture concentration can be calculated by the sum of results exported from the two parallel Fickian models.

As displayed in Table 2, in the moisture/temperature diffusion analysis in ABAQUS, temperature T in the heat conduction analysis is analogous to moisture content C_i in the moisture diffusion model. To simulate each Fickian diffusion stage of the Dual-Fickian model, the thermal conductivity λ can be assumed as the moisture diffusion coefficient D_i , while the density ρ and heat capacity c_{th} in the heat conduction analysis can be set to 1 in the moisture diffusion model. The initial moisture conditions (C_{ini}) are set to zero to represent a dry state, while the boundary condition of each stage should be the same as the final saturated moisture content C_{si} obtained in the moisture absorption-desorption cycles.

Table 2. Dual-Fickian model implemented in ABAQUS using thermal-moisture analogy.

Properties	Field Variable	Parameters	Boundary condition
Thermal conduction	T	λ ρ c_{th}	T_{imp}
Dual-Fickian Diffusion	Stage 1	C_1 D_1 1 1	C_{s1}
	Stage 2	C_2 D_2 1 1	C_{s2}

Next, the key parameters of the Dual-Fickian model in the first absorption cycle can be obtained by fitting the experimental curves of three specimens, as shown in Table 3. Similarly, the key parameters of the Fickian model for the first desorption phase and the second absorption-desorption cycle can be calculated by fitting the experimental curves, as shown in Table 4.

Table 3. Key parameters of Dual-Fickian model in the first absorption cycle.

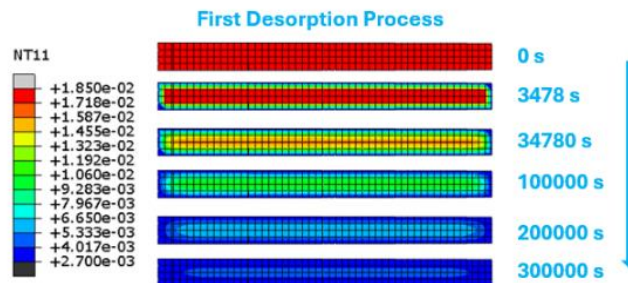
Parameters	D_1 (mm ² /s)	C_{s1} (%)	D_2 (mm ² /s)	C_{s2} (%)	C_s (%)
Value	$4,11 \times 10^{-6}$	1.16	$4,07 \times 10^{-8}$	0.64	1.8
Standard deviation	$\pm 0,14 \times 10^{-6}$	± 0.02	$\pm 0,30 \times 10^{-8}$	± 0.11	± 0.08

Table 4. Key parameters of Fickian model for the other cycles.

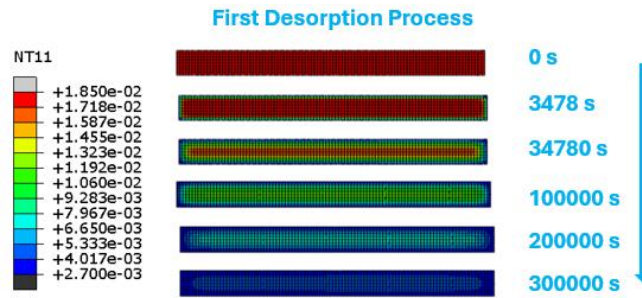
Cycles	Parameters	C_{ini} (%)	D (mm ² /s)	C_s (%)	C_{res} (%)
Desorption 1	Value	1.9	$3,53 \times 10^{-6}$	1.57	0.33
	Standard deviation	± 0.16	$\pm 0,18 \times 10^{-6}$	± 0.02	± 0.15
	Value	0.27	$3,70 \times 10^{-6}$	1.55	1.81
Sorption 2	Standard deviation	± 0.06	$\pm 0,15 \times 10^{-6}$	± 0.04	± 0.11
	Value	1.85	$3,42 \times 10^{-6}$	1.51	0.33
Desorption 2	Standard deviation	± 0.11	$\pm 0,16 \times 10^{-6}$	± 0.05	± 0.06

3.3. Numerical results and discussions

To validate the experimental results and improve the accuracy of predicting the complex moisture diffusion behavior of polymeric materials, the refinement of the computational modeling based on the Dual-Fickian model has been performed. Finite element models of epoxy resin samples were created by comparing two sizes of plates: 50 mm×50 mm and 25 mm×25 mm, each with a thickness of 2 mm. It aims to evaluate the influences of sample size and mesh refinement on computational efficiency and accuracy. The first idea was to employ a mesh size of 0.5 mm along the 2 mm thickness, followed by the subsequent modeling using finer meshes such as 0.4 mm and 0.25 mm (see Figure 3). It is clear that the finer mesh size exhibits a more accurate moisture concentration distribution, especially in the earlier moisture absorption/desorption stage.



(a) Mesh size of 0.5 mm.



(b) Mesh size of 0.25 mm.

Figure 3. Moisture diffusion process in the phase Desorption 1.

Figure 4 plots the average moisture absorption-desorption curves obtained by the sum of two separate Fickian models in ABAQUS, with a mesh size of 0.5 mm, compared to the experimental results. Dots in the figure represent experimental results, while the smooth and dashed lines show the simulation results, with Sp and Dp signifying the phases of Sorption and Desorption, respectively. Two sizes of models with different lengths of 50 mm and 25 mm are also compared here. It is interesting to find that the moisture diffusion curves for both sample sizes show significant overlap, suggesting that the diffusion kinetics are independent of the plate size ranging from 50 mm to 25 mm. However, the computational time for the smaller 25 mm sample shows a significant decrease. To be emphasized, the numerical results in phase Sorption 1 deviate a little from the experimental curve during the earlier period, indicating a need for mesh refinement. However, good agreement with the experimental data can be found in subsequent cycles of Desorption 1, Sorption 2, and Desorption 2.

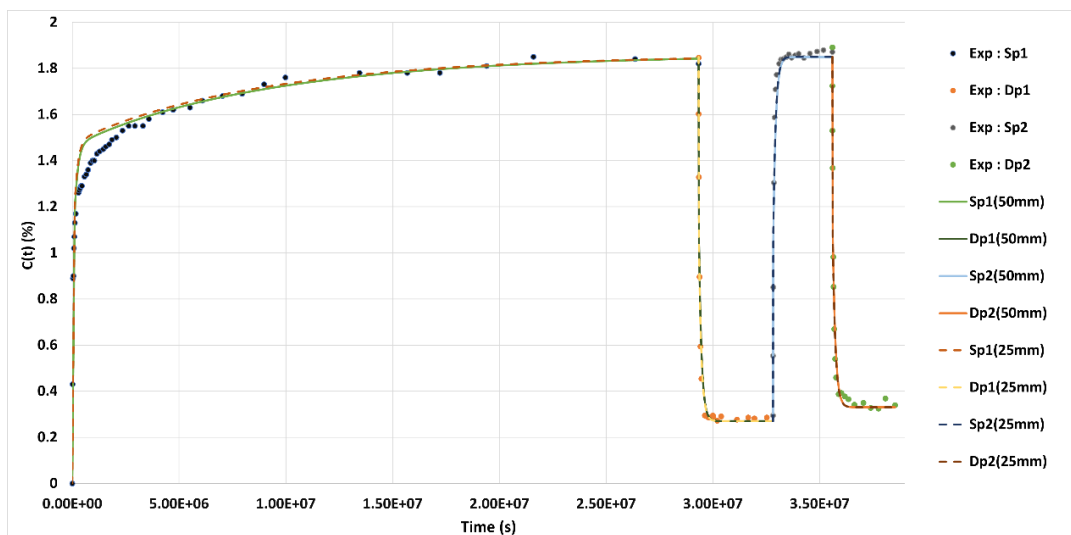


Figure 4. Modeling of moisture absorption-desorption cycles with a mesh size of 0.5 mm.

After refining the mesh size to 0.4 mm (see Figure 5), it can be observed that the curve in the first phase Sorption 1 is more consistent with the experimental results for both sample sizes. This improvement has proven that the finer mesh resolutions could lead to higher accuracy of the model, without altering the moisture diffusion behavior in the other latter cycles (Desorption 1, Sorption 2, and Desorption 2), which always remains consistent with the experimental results.

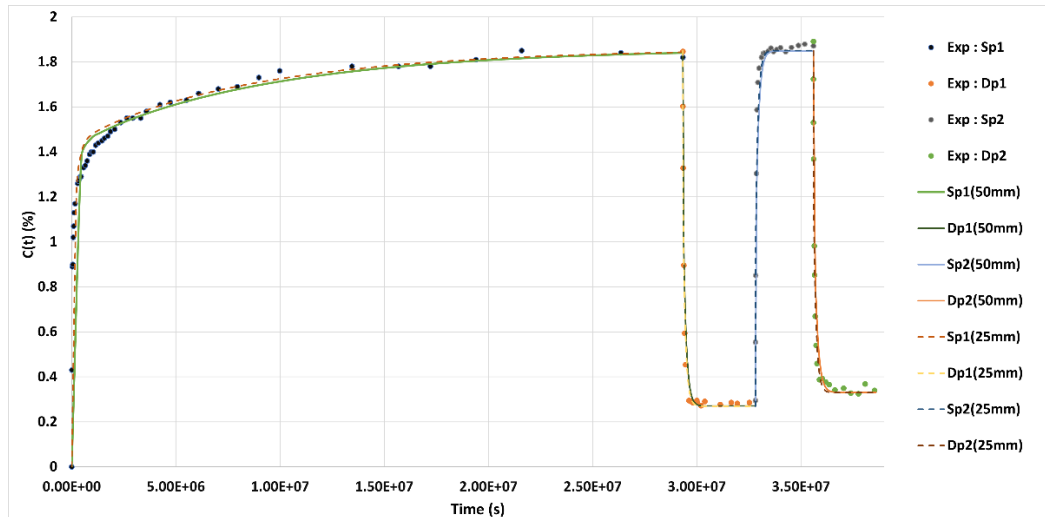


Figure 5. Modeling of moisture absorption-desorption cycles with a mesh size of 0.4 mm.

However, the simulation of the 50 mm sample could not be completed at a mesh resolution of 0.3 mm and 0.25 mm, due to the errors resulting from excessive computation time of over 4 hours. As a result, only the 25 mm model was analyzed (Figure 6), which shows that the moisture absorption curve in Sorption 1 can closely match the experimental data within a reasonable 30-minute computational time.

Thus, all results have demonstrated that reducing the sample size from 50 mm to 25 mm can effectively reduce the computational cost without compromising the accuracy. Furthermore, the finer meshes can significantly increase the fidelity of the first Dual-Fickian Sorption 1 cycle compared to the experimental data, but at the cost of increased computational time and memory.

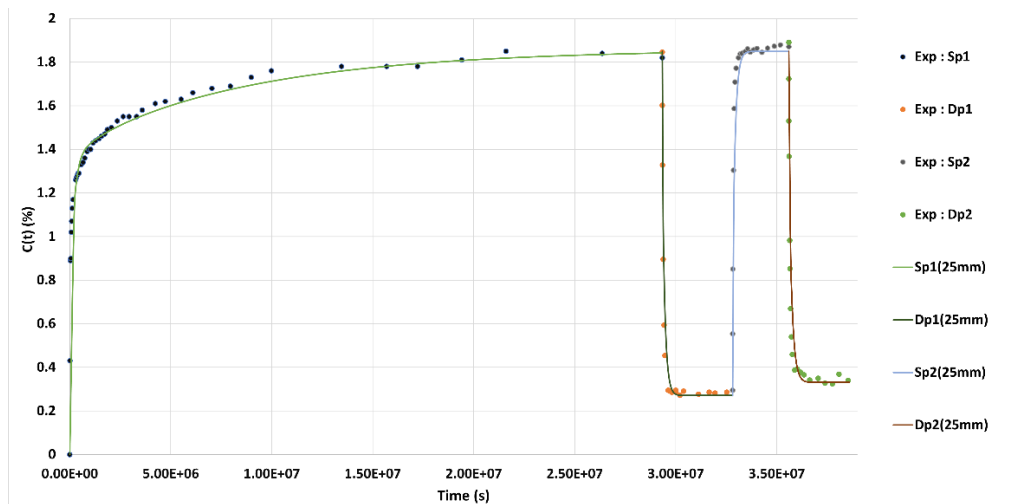


Figure 6. Modeling of moisture absorption-desorption cycles with a mesh size of 0.25 mm.

Eventually, the numerical analysis has proven that the curves (Desorption 1, Sorption 2, and Desorption 2) based on the Fickian model are independent of the mesh size. It highlights the predictable nature of moisture diffusion in the epoxy resin exhibiting the Fickian behavior. As the mesh is refined, the curve in the first phase Sorption 1 based on the Dual-Fickian model becomes increasingly in agreement with the experimental results. This improvement emphasizes the need for finer mesh analysis to accurately predict the more complex Dual-Fickian behavior in the epoxy resin.

4. Conclusions and Perspectives

In conclusion, all experimental and numerical results have given a comprehensive understanding of the intricate dynamics of the moisture diffusion process within the epoxy resin subjected to cyclic absorption-desorption aging conditions. The results of the first absorption phase Sorption 1 have verified the complexities of the Dual-Fickian behavior, whereas the subsequent cycles have exhibited the predictable and reproducible Fickian behavior. Particularly, the FEA simulations have validated the experimental results and highlighted the need for finer meshes to realize a more reliable prediction of the Dual-Fickian diffusion process. These findings can be used in the efficient material designs of pipes with the right resin formulations, resulting in an enhanced manufacturing process and a more durable and robust pipe.

However, future research should focus on integrating advanced experimental techniques such as microscopic observations to investigate the microstructural damages occurring within the material as a result of the complex nature of the Non-Fickian diffusion behavior. In addition, the role of the thickness on the moisture diffusion kinetics of the epoxy resin can be further validated using experimental and numerical methodologies.

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