#### Predicting Multi-axial Diffusion of Water in Laminated Composite Structural Components

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### Abstract

This study examines the diffusion of water in fiber-reinforced polymer (FRP) laminated composites, comprising epoxy resin and unidirectional glass fibers, at different material and structural scales. Experiments on the diffusion of neat resin, FRP laminas in the axial and transverse fiber directions, FRP laminated composites with different stacking sequences, and FRP plates, rods, and pipes are conducted to obtain the diffusion processes at different length scales. Simultaneously, a diffusion model for a laminated composite is formulated to mathematically describe the multi-axial diffusion processes in FRP composites at various length scales, i.e., single lamina, laminated stacking sequence, and component. The study shows that the diffusivity constants calibrated from neat epoxy resin and unidirectional laminas are adequate to predict the diffusion responses of laminated composite systems and structural components of larger sizes. This capability is useful in predicting the long-term diffusion responses of FRP composites at large sizes where conducting diffusion tests on such systems is not feasible.

### 1. Introduction

The lightweight nature and good load-bearing capacity of fiber-reinforced polymer (FRP) composites make them attractive for many applications in the aerospace industry, marine structures, civil infrastructures, and oil and gas industry [1-3]. In many applications, the FRP composites are often subjected to various environmental conditions. The long-term and continuous changes in environmental conditions, such as temperature fluctuations and humidity changes, affect the performance of composite structures. Several studies have reported reductions in the mechanical properties, e.g. tensile modulus, tensile strength, shear strength, and fracture toughness, after exposure to ambient humidity [4-9]. Due to their significant effects on the mechanical properties of the composites, it is then important to understand the long-term diffusion process in the FRP materials and structures. The focus of this study is to examine the multi-axial diffusion responses in FRP laminated composites and structural components. This study also investigates whether the diffusivity constants determined from small scale specimens of neat resin and unidirectional laminas are adequate in capturing the overall diffusion responses of large-scale composites of more complex geometries.

There have been extensive experimental studies on investigating the diffusion of moisture and fluid through FRP composites. Most of the experiments considered FRP specimens of thin plates that are immersed completely in a chamber of high humidity or a fluid bath, and the weight gained in the specimens are periodically measured until the specimens are fully saturated. As the FRP composites show anisotropic responses with regards to their diffusivity behaviors, several methods have been considered to determine the anisotropic diffusivity of FRP specimens [10-12]. A thin flat sample is sealed along its thickness edges to allow for one-dimensional diffusion through the thickness. The approach works well for obtaining the through-thickness diffusivity because regular thin plies or laminates can be tested. The through-thickness diffusivity is associated with the transverse fiber diffusivity. However, the diffusivity in the fiber direction is more difficult to obtain. Using a standard thin laminate is not well suited for measuring diffusivity in the fiber direction. The large surface area would have to be sealed and it would take a long time for water to get into the specimen. Making very thick specimens and cutting them in a way that the fiber ends are forming the large surface has solved the problem [11].

FRP composites used in many engineering applications are not in the form of thin flat panels, and the geometrical shapes can be complicated. Thus, it can be very challenging to determine the anisotropic diffusivity properties in FRP structures. Gagani et al. [13] considered an optimization approach using linear regression in determining anisotropic diffusivity properties of FRP structures with various geometries, i.e., thin flat specimens, rods, and pipes. The systems consist of unidirectionally aligned glass fibers and epoxy matrix with approximately the same fiber volume fraction. They showed that the calibrated diffusivity constants along and transverse to the fiber directions from all three different specimens are quite close. This indicates that the diffusion behaviors in FRP systems are governed by the diffusivity characteristics of their basic constituents which are fibers and matrix. It is noted that in practical applications, the FRP structures are comprised of laminates of various stacking sequences, which can further complicate the determination of diffusivity constants in such systems. It is certainly not feasible to experimentally determine and characterize overall diffusivity constants in FRP laminates of various stacking sequences and complex geometries. A predictive model is then required to estimate the diffusion behaviors in FRP structures of various geometries and fiber orientations.

Several investigations of moisture absorption in FRP composites have suggested significant differences between the diffusion along the fiber direction and the diffusion

perpendicular to the fiber direction [12-15]. Specifically, the diffusivity along the fiber direction can be 2-4 times larger than the transverse diffusivity for glass fiber laminates [16] and about 14 times higher for carbon fiber rods [17]. The glass fibers themselves are often seen as nonpermeable to water, assuming to have zero diffusivity [18]. This assumption cannot adequately capture the diffusion behaviors in FRP composites. In FRP composites, the fibers comprise bundles of fiber filaments and during immersion experiments, water seepage can occur within the filaments in the fiber bundles.

There have been several analytical and numerical approaches for predicting the overall diffusivity behaviors of FRP composites. Taking into consideration the anisotropy of the fiberreinforced composites, Shen and Springer [10] developed an analytical model to predict the diffusivity constants of FRP composites. The relationships among the diffusivity of a matrix, diffusivity of fibers, fiber volume fraction, and fiber orientation are established by considering the similarities between moisture diffusion and thermal conductions [19]. The model has been widely applied in capturing the moisture diffusion in FRP composites, e.g. Whitney [20] and Loos and Springer [21]. Rocha et al. [18] proposed a FE model with high diffusivity interphase, to explain anisotropic diffusivity in composites. They concluded that the fiber barrier effect is partly responsible for the anisotropic diffusion in composites, but this alone does not explain the effect observed. Gagani et al. [12] and Fan et al. [22] have considered a micromechanics model in determining the diffusivity constants of FRP composites, having glass fiber and epoxy matrix, in the fiber and transverse fiber directions. The micromechanics model considered different diffusivity constants of fiber and matrix and the fiber volume fraction. The micromechanics model was formulated based on the Fickian diffusion process, and the model shows a good agreement with the experimentally measured diffusivity behaviors.

This study presents a predictive model for describing the multi-axial diffusion behaviors in FRP composites, considering different fiber and matrix diffusivity behaviors, fiber volume fraction, various off-axis fiber stacking sequences, and macroscopic geometry of the FRP composites. The model involves two material length-scales, i.e., a single-ply with unidirectional fiber and matrix constituents and laminated systems with different off-axis fiber orientations. When a linear diffusion behavior, following the Fickian model, is considered for both anisotropic fiber and isotropic matrix, the exact determination of the overall multi-axial diffusivity constants can be obtained. The material model is implemented within finite element analysis, which allows for predicting the diffusion behaviors in FRP structures of complex geometries. To corroborate the proposed multi-scale analyses, two groups of experimental tests are considered. In the first group, thin FRP specimens with [0/45/-45/0] and [45/-45/-45/45] stacking sequences and different in-plane dimensions, i.e., 10x10 mm, 20x20mm, and 30x30 mm, are tested. In the second group, thin FRP plates with different fiber orientations and FRP rods and pipes of different sizes are tested.

This manuscript is organized as follows. Section 2 discusses the materials and experimental tests, followed by the discussion of the model in Section 3. Section 4 presents the comparisons of the diffusion behaviors obtained from the experiments and model. Finally, Section 5 is devoted to a brief recapitulation of the results and concluding remarks.

### 2. Materials and Experiments

## 2.1 Materials and Testing Procedures in Group 1

The glass fibers used were HiPer-Tex<sup>TM</sup> unidirectional fabric from 3B. The epoxy was supplied by Hexion and consisted of RIMR135<sup>TM</sup> resin and RIMH137<sup>TM</sup> hardener with a mass

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# **Data Availability**

The raw/processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study. The data will be made available upon acceptance/publication of the paper.

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