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► **To cite this version:**

Georges Youssef, Frédéric Jacquemin, Sylvain Fréour. Radial Pressure and thickness effects on the moisture diffusion in hollow composite cylinders. *Polymer Composites*, 2011, 32 (6), pp.960-966. 10.1002/pc.21114 . hal-01154415

**HAL Id: hal-01154415**

**<https://hal.science/hal-01154415>**

Submitted on 29 Nov 2017

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# Radial Pressure and Thickness Effects on the Moisture Diffusion in Hollow Composite Cylinders

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According to the literature, the mechanical states (external or internal) have an effect on the penetration of moisture into a composite material through affecting both its main controlling parameters: the diffusion coefficient and the maximum moisture absorption capacity. In this article, a scale transition analysis is performed to investigate the coupled effect of the external/internal mechanical states, experienced by epoxy resin composite structures, on the moisture penetration process. The scale transition approach provides relations linking the plies strains to those of their constituents and homogenization procedures enabling to estimate the evolution of the plies diffusion parameters, from those of the epoxy, at any step of the moisture diffusion process. This work underlines the effects induced by the thickness of the considered composite structure. The respective contributions of the internal and external loads (in the case of a radial pressure, only) on the transient and permanent stages of the diffusion process are also investigated.

## INTRODUCTION

Fiber reinforced organic matrix composites are increasingly used in aerospace [1] and other engineering applications [2, 3] due to their light weight, high specific strength, high specific stiffness, excellent fatigue and corrosion resistance properties. But, as for any industrial application, the question of their durability is a critical issue. That implies the conservation of the functional characteristics of the product which, in service, will be subjected to various mechanical and environmental loads. These structures are actually often subjected to humid environments during their service life [4]. Organic matrix composites do absorb significant amount of water when exposed to moisture [5]. Moisture absorption in polymer matrix composites generates hygro-elastic internal stresses

[6]. Moreover, the constituents of composites structures exhibit heterogeneous coefficients of moisture expansion and maximum moisture absorption capacities. As a consequence, multi-scale in-depth mechanical states profiles rise during the hygroscopic loading of organic matrix composites, which can eventually induce damage [7–9]. Therefore, models have been developed to predict the multi-scale mechanical states occurring during both the transient stage and the permanent regime of the moisture diffusion process of organic matrix composites submitted to hygro-mechanical loads [10–14]. In this field of research, the most recent investigations [15–19] focus upon taking into account, in the theoretical approaches, various features of hygro-mechanical coupling, especially the dependence of both the moisture diffusion coefficient and the maximum moisture absorption capacity on the mechanical states (strains and stresses), as shown, for instance, in [20–24]. More recent works, made by Autran et al. [25] and Wan et al. [26], have experimentally demonstrated the influence of mechanical loading on the moisture diffusion behavior.

In this study, a scale transition analysis, based on the free-volume theory, is achieved to investigate the coupled effect of both the external and the internal mechanical states (stresses and strains) experienced by organic matrix composite structures, on the moisture penetration process. This work is focused on the main penetration mechanism known in composite materials, namely, the diffusion into the bulk resin. The second section of the present contribution summarizes the main relations involved in the theoretical approach, for modeling the hygro-mechanical coupling. The third and fourth sections are respectively dedicated to the presentation of the numerical results obtained in the cases that either thin or thick composite structures are considered.

### *Hygro-Mechanical Coupling: A Multi-Scale Analysis*

The free volume actually corresponds to the difference between the specific (macroscopic) volume of the polymer and the actual volume occupied by its constitutive molecules [18].

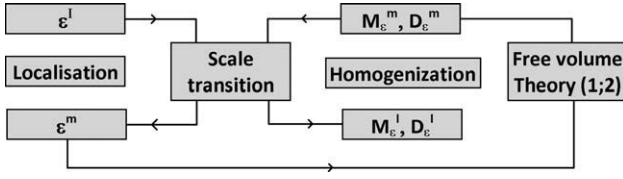


FIG. 1. Chart of the scale transition procedure.

Following the experimental observations, a theoretical approach, based on the calculation of the free-volume change in the stressed state, was suggested [20]. In practice, it was later observed by Neumann and Marom, that the main mechanical state controlling the hygro-mechanical coupling was the matrix strain  $\varepsilon^m$  instead of the stresses [23, 27]. Accordingly, the following present work will develop a model based on the strains.

Assuming that the Fickian diffusion coefficient was related to the free volume by the Doolittle equation, the authors proposed an expression for the ratio of the diffusion coefficients in the strained and free-of-strain states [22, 27]:

$$\ln\left(\frac{D_{\varepsilon}^m}{D_0^m}\right) = \frac{a}{v^m} \frac{\text{Tr}\varepsilon^m}{v_{f0}^m [v_{f0}^m + \text{Tr}\varepsilon^m]} \quad (1)$$

$v^m$  denotes the volume fraction of the matrix in the composite ply, whereas  $a$  is an empirically factor, varying in a narrow range [23]:  $0.031 \leq a \leq 0.036$  with an average value equal to 0.033.  $v_{f0}^m$  and  $v_{f\varepsilon}^m$  are the free volume fraction of the strain-free epoxy and that of the strained organic matrix, respectively.

Many authors agree that the value of  $v_{f0}^m$  for a defect-free resin tends towards 2.5% [20, 22].

To estimate the diffusion coefficient of the strained polymeric matrix  $D_{\varepsilon}^m$ , the knowledge of both the organic matrix strain tensor and its strain-free diffusion coefficient  $D_0^m$  is required.  $D_0^m$  can be deduced from the slope of the representative curve of the evolution of the percent mass change with the square root of time, when this variable tends towards 0 (i.e., at the beginning of the diffusion process [23]).

The maximum moisture absorption capacity  $M_{\infty\varepsilon}^m$  for a strained epoxy satisfies (see for instance [22] and the more recent article of Wan et al. [26]):

$$M_{\infty\varepsilon}^m = M_{\infty 0}^m + \text{Tr}\varepsilon^m \times \frac{\rho^w}{\rho^m} \quad (2)$$

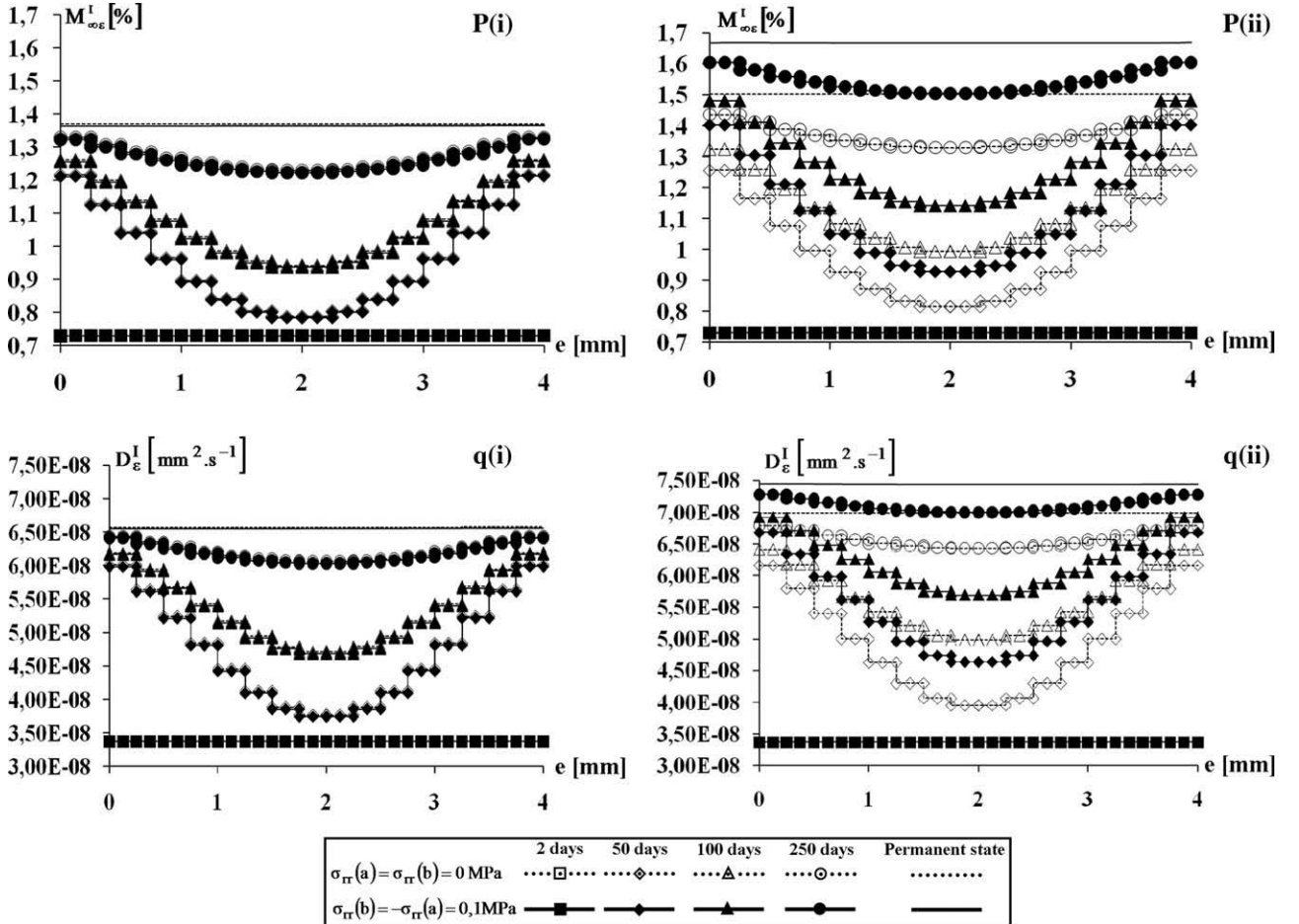


FIG. 2. (p) maximum moisture absorption capacity/(q) moisture diffusion coefficient profiles in the (i)  $\pm 55^\circ$  laminate/(ii) unidirectional thin composite structure, with or without external load.

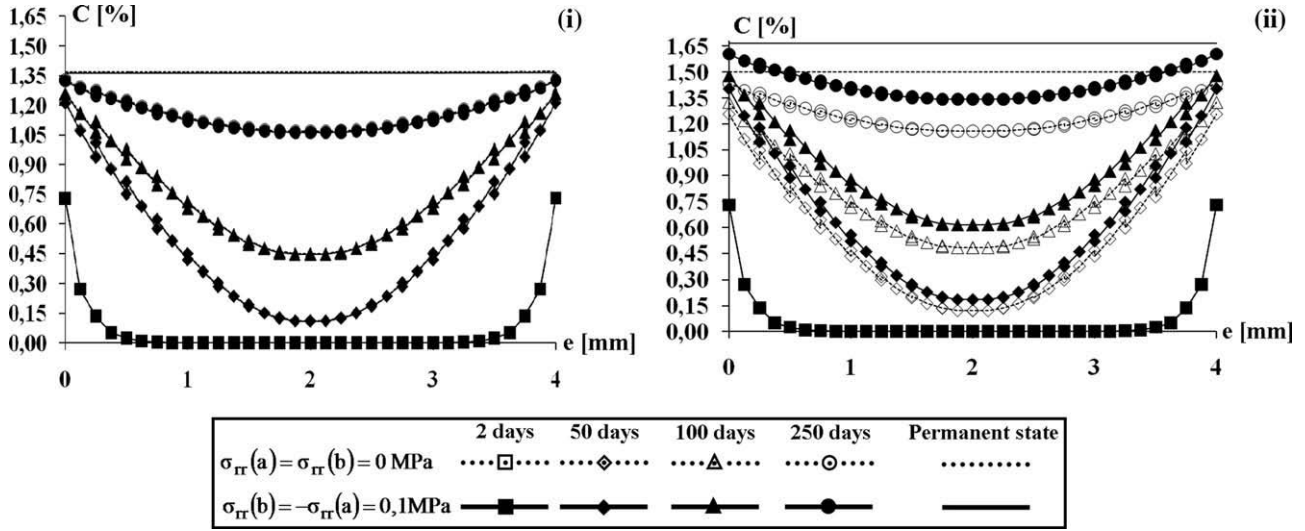


FIG. 3. Macroscopic moisture content profiles in the (i)  $\pm 55^\circ$  laminate/(ii) unidirectional thin composite structure, with or without external load.

where  $\rho^w$  and  $\rho^m$  are, respectively, the densities of the water and matrix, whereas  $M_{\infty 0}^m$  stands for the maximum moisture absorption capacity of the unstrained epoxy.

Rules of mixtures or scale transition relations are used to determine both the moisture diffusion coefficient and the maximum moisture absorption capacity at macroscopic scale in each ply of the composite structure, as explained in Ref. [18].

Since we consider impermeable fibers, the expression of the maximum moisture absorption capacity of the composite  $M_{\infty \varepsilon}^l$  is given by:

$$M_{\infty \varepsilon}^l = v^m \frac{\rho^m}{\rho^l} M_{\infty \varepsilon}^m \quad (3)$$

where  $\rho^l$  is the density of the composite ply.

The expression of the effective coefficient of diffusion of each ply of the structure comes from a representation of the representative elementary volume by concentric cylinders [28]:

$$D_{\varepsilon}^l = D_{\varepsilon}^m \frac{1 - v^f}{1 + v^f} \quad (4)$$

$v^f$  denotes the volume fraction of the fiber in the composite ply.

The hygromechanical coupling induces for the constitutive plies different moisture diffusion coefficients and moisture contents. Thus, the moisture flux is continuous at the interply and the moisture content is discontinuous [10].

In this work, a Fickian behavior is assumed. It was actually shown, for instance, by Wan et al. [26] that the application of a mechanical loading did not change the Fickian kinetic of moisture sorption by polymers. However, the same article reports that the application of a mechanical loading on epoxy resin composites induced a change of both the maximum moisture absorption capacity and the moisture coefficient diffusion, consistent with the free-volume theory developed in this work.

The macroscopic moisture content (at ply scale) is a solution of the Fick's equation with a moisture diffusion coefficient dependent on the local mechanical state experienced by the organic matrix of the considered composite ply. This problem is solved by using the finite differences with an explicit scheme, accounting for a moisture content jump between two adjacent plies:  $\alpha_{i+1} = \frac{(M_{\infty \varepsilon}^l)_{i+1}}{(M_{\infty \varepsilon}^l)_i}$ ,

where  $(M_{\infty \varepsilon}^l)_{i+1}$  and  $(M_{\infty \varepsilon}^l)_i$  are the maximum moisture absorption capacity of the two adjacent plies deduced from that of the matrix. The macroscopic internal stresses are calculated by considering the homogenized properties and the classical equations of solid mechanics: constitutive laws of hydro-elastic orthotropic materials, strain-displacement relationship, compatibility and equilibrium equations and boundary conditions. The coupling between moisture diffusion and mechanical states involves an incremental solving. The corresponding local mechanical states in the constituents (fibers and matrix) are deduced from their macroscopic counterparts through Eshelby-Kröner hydro-mechanical scale transition model [11, 13].

The scale transition procedure required for modeling/simulating the multi-physical mechanisms involved during the transient part of the moisture diffusion process in polymeric matrix composite structures is summarized on Fig. 1. The input data required to achieve the calculations have been provided in Ref. [18].

## NUMERICAL RESULTS

### *Effect of a Radial Pressure on the Hygro-Mechanical Coupled Behavior of a Thin Composite Tube*

**Diffusive Behavior.** Figure 2 shows the evolutions obtained for the profiles of the maximum moisture absorption capacity and moisture diffusion coefficient, at the

macroscopic scale; in the case of a 4-mm thin tubular N5208-T300 composite structure. The cases of an unidirectional and a  $(+55^\circ/-55^\circ)$  laminated structure were compared. On the same figure are displayed the results obtained for either the unloaded hollow cylinder or for the tube submitted to a radial pressure (1 bar) on its external and internal edges.

According to Fig. 2, the additional external load does not modify the profile of the two main parameters of the moisture diffusion process in the laminate. This is due to the fact that the trace of the strains, induced by the radial pressure, experienced by the epoxy, is actually weak in that specific case. On the contrary, the application of a radial pressure slightly increases the macroscopic moisture diffusion coefficient in the unidirectional structure (the relative variation reaching 7% in permanent state), but it is the maximum moisture absorption capacity which is the most affected: the relative deviation by comparison with the unloaded tube can reach 12%.

Figure 3 presents the variations of the macroscopic moisture content profiles, resulting from the above displayed (cf. Fig. 1) depth and time dependent profiles obtained for both the moisture absorption capacity and moisture diffusion coefficient. The obtained profiles for the laminated tube are unaffected by the considered additional radial pressure. On the contrary, the moisture diffusion in the unidirectional structure is accelerated by the presence of the same mechanical load. Moreover, the contents reached at saturation are significantly more important, by comparison with the case of the pure hygroscopic load.

#### Multi-Scale Mechanical States

According to Fig. 4, the multi-scale transverse stresses of the laminated structure are not strongly affected by the application of a radial pressure. The macroscopic transverse stresses, which vary from zero to 50 MPa in the internal and central plies of the unidirectional structure, following the application of a radial pressure on the tube, are strongly concentrated in the fibers, whose mechanical state reaches 85 MPa in this direction. The matrix is in a stress state of about  $-20$  MPa, which is preferable to the level of  $-60$  MPa reached without external pressure. The multi-scale shear stress states are weak, whatever the constituent, in the permanent state, for the laminated tube submitted to an external radial pressure. The most intense stress states occur at the beginning of the diffusion process.

#### Effect of a Radial Pressure on the Hygro-Mechanical Coupled Behavior of a Thick Composite Tube

**Diffusive Behavior.** Figure 5 shows the evolutions obtained for the maximum moisture absorption capacity and diffusion coefficient profiles, at macroscopic scale, in 20-mm thick cylinders. The cases of a unidirectional structure and a laminate structure were compared. We also considered the absence of external load or the appli-

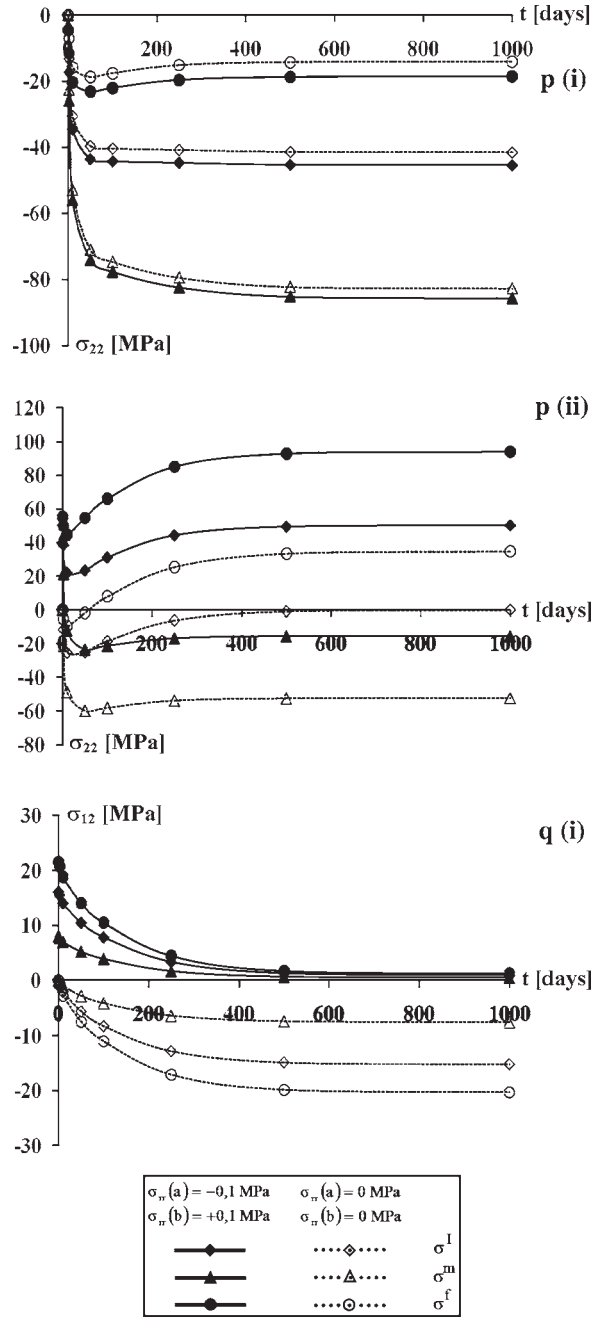


FIG. 4. Multi-scale (p) transverse/(q) shear stress profiles, in the internal ply of the (i)  $\pm 55^\circ$  laminate/(ii) unidirectional thin composite structure, with or without external load.

cation of a radial pressure (10 bars) on the external edge of the tube, while the inner surface remains unloaded.

A slightly asymmetrical profile of the parameters controlling the moisture diffusion process is predicted in the thick structure. The external pressure tends to increase the maximum moisture absorption capacity and the moisture diffusion coefficient in the laminated tube, while the opposite effect in the unidirectional case is observed. The influence of an external mechanical load on the hygro-mechanical coupling is thus strongly linked to the stacking sequence.

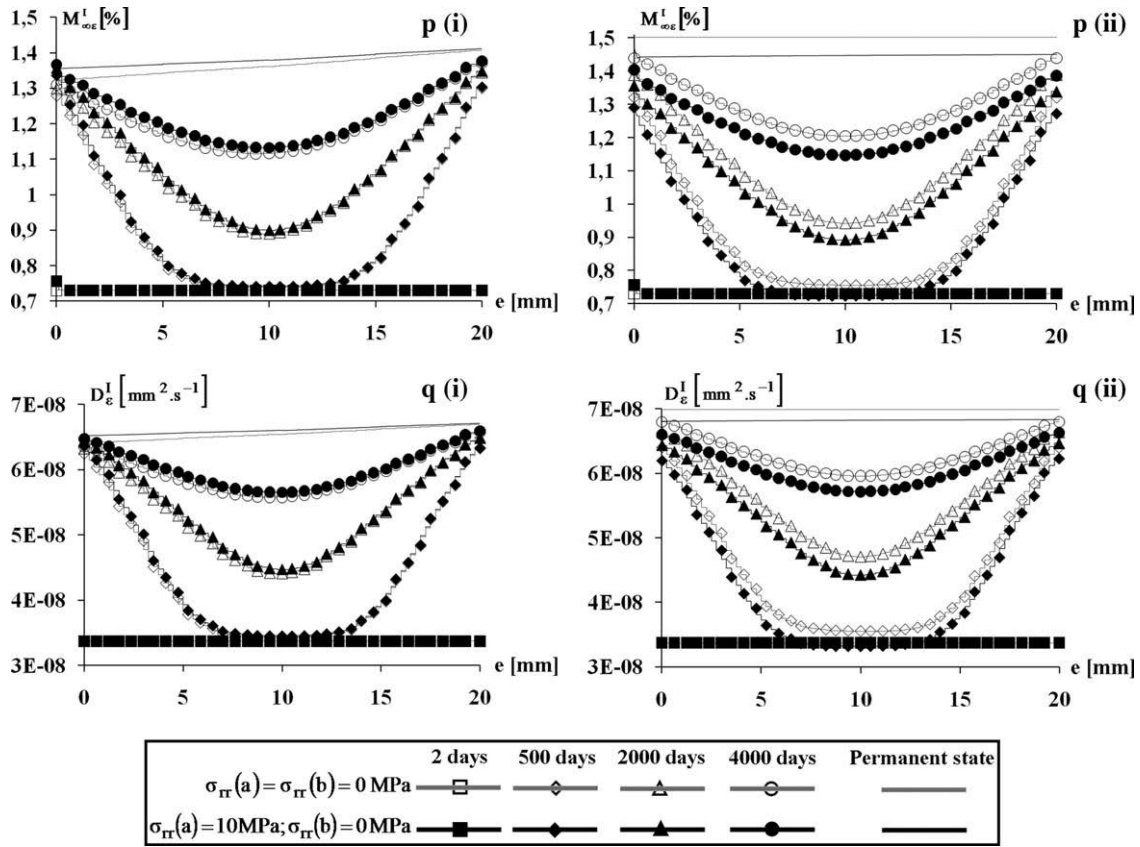


FIG. 5. (p) maximum moisture absorption capacity/(q) moisture diffusion coefficient profiles in the (i)  $\pm 55^\circ$  laminate/(ii) unidirectional thick composite structure, with or without external load.

The evolution of the moisture content, through the thickness of the tube, as a function of the time is displayed on Fig. 6. The obtained results are consistent with the observations highlighted in the case that the thin structure was investigated (Fig. 2): in particular, the dependence of the calculated moisture content profiles on the lay-up configuration of the plies. As an example, the

application of a radial load to the laminated tube assists the penetration of moisture within the depth of the structure, contrary to what occurs for the unidirectional tube.

**Multi-Scale Mechanical States.** Figure 7 shows the time-dependent evolution of the multi-scale transverse and in-plane shear stresses for the internal ply of a thick

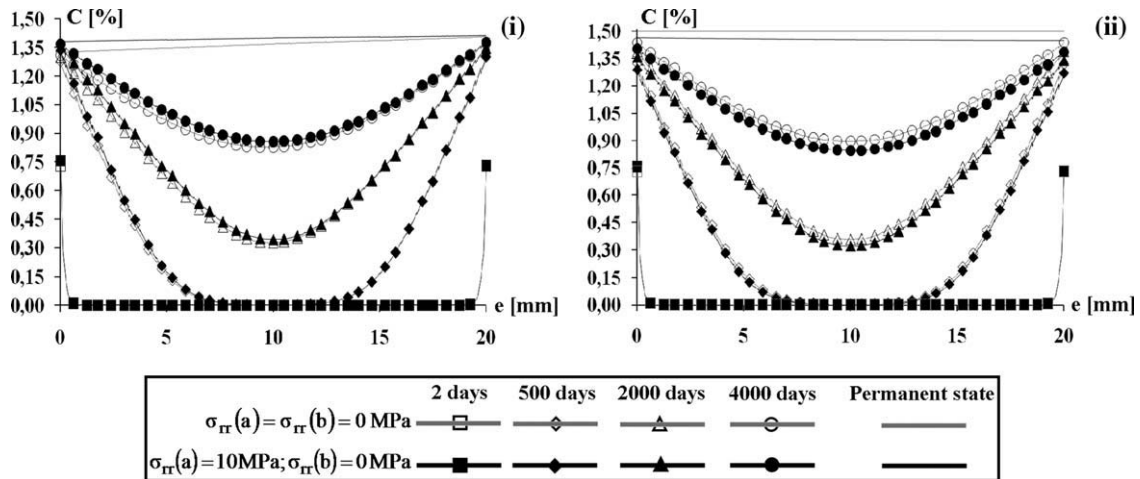


FIG. 6. Macroscopic moisture content profiles in the (i)  $\pm 55^\circ$  laminate/(ii) unidirectional thick composite structure, with or without external load.

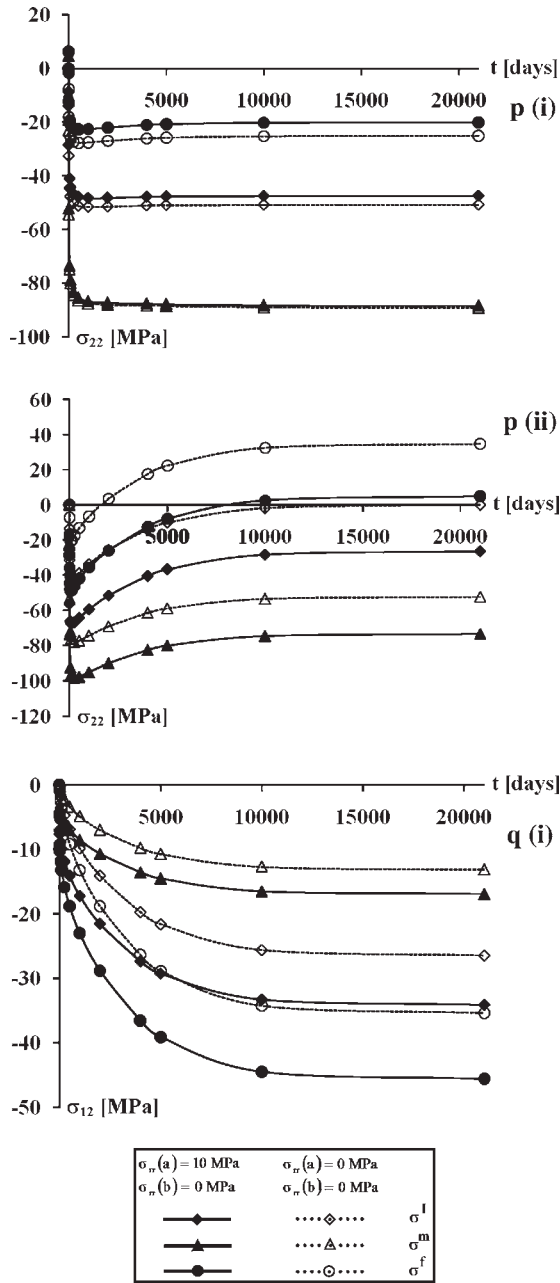


FIG. 7. Multi-scale (p) transverse/(q) shear stress profiles in the internal ply of the (i)  $\pm 55^\circ$  laminate/(ii) unidirectional thick composite structure, with or without external load.

tube, as a function of the stacking sequence and the applied external load.

The effects related to the presence of an additional mechanical load are significant: the static deviation for the multi-scale transverse stress can reach 25 MPa in the reinforcing fiber of the unidirectional structure. If the absolute deviation is weaker for the matrix transverse stress of the internal ply, the additional mechanical load increases the absolute value of the instantaneous stress undergone by this constituent, which reaches 100 MPa in compression during the transient stage of the moisture diffusion process, against 80 MPa only in the unloaded cyl-

inder. The multi-scale transverse stress states of the  $\pm 55^\circ$  laminated structure do not depend on the application of an additional mechanical load in the considered case. On the contrary, the multi-scale shear stresses are strongly affected by the occurrence of a radial pressure on the laminate. The effects of an additional mechanical load can thus be significant on the internal mechanical states, even if they do not systematically modify in an important way the moisture penetration in a composite structure.

## CONCLUSIONS

The originality of this contribution is to propose a multi-scale modeling based on the free-volume theory, to investigate the coupled hygro-mechanical behavior of composite tubes, induced by an additional external mechanical load.

Only the case of radial loads was considered in this study. The intensity of such loads being strongly limited for remaining in the elastic deformation domain, the corresponding multi-scale states of strains are, consequently, also weak. As a consequence, the considered hygro-mechanical loads do not result in through thickness distributions remarkably different for the parameters governing the diffusion process: diffusion coefficients, maximum moisture absorption capacities and moisture contents. However, the time and space profiles of the multi-scale internal stresses can vary in a significant way, as a function of three interdependent factors: the applied external load, the structure thickness, as well as the stacking sequence of its constituting plies.

Further works will be focused on the experimental investigations required for validating the theoretical approach presented in this article.

The next step of the numerical work will be focused on the simultaneous modeling and simulation of both the hygro-mechanical coupling expected to occur in polymer matrix composites: the evolution of the matrix properties (namely its mechanical stiffness and coefficient of moisture expansion), on the one hand, and the effects induced on the diffusion parameters by the mechanical strain state experienced by the matrix on the other hand. In further numerical works, a more rigorous treatment, compatible with the fundamental principles of the thermodynamics, of the multi-physical coupled mechanisms involved during the diffusion moisture process, will be developed.

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