

## **STRESS–STRAIN ANALYSIS OF SPECIMENS SUBJECTED TO TENSILE LOADING DURING MOISTURE UPTAKE**

A. Aniskevich<sup>a\*</sup>, A. Mintzas<sup>a,b</sup>, R. Guedes<sup>c</sup>

<sup>a</sup> *Institute of Polymer Mechanics, University of Latvia, 23 Aizkraukles str., Riga, LV-1006 Latvia*

<sup>b</sup> *Magma Global Ltd., 1000 Lakeside North Harbour, Western Road, Portsmouth, PO6 3EZ, UK*

<sup>c</sup> *INEGI - EXPMAT - Experimental Mechanics and New Materials & Department of Mechanical Engineering, Faculty of Engineering of the University of Porto, 4200-465 Porto, Portugal,*

*\*e-mail address of the corresponding author Andrey.Aniskevich@pmi.lv*

**Keywords:** moisture sorption, transient hygroscopic conditions, viscoelasticity, epoxy matrix.

### **Abstract**

*A numerical method for the calculation of the internal stress state that develops within structures subjected to mechanical and steady state or transient hygroscopic loading conditions, has been developed. The method encompasses a layer by layer approach whereby the structure is discretized into plies with different material properties corresponding to the different ply moisture contents. The proposed method has been validated against finite element solutions, and results from its application on a fully characterized epoxy polymer binder are presented. The impact of the moisture induced viscoelastic behaviour on the structural response of the case studied is discussed.*

### **1. Introduction**

Real polymer and composite structures are usually subjected to transient hygroscopic loading conditions (e.g. day and year cycles) and even in the case where stationary conditions apply, sorption process may take years before moisture equilibrium is reached. As a consequence, a variable stress profile arises within the structure which can induce damage [1] and can eventually lead to failure.

The first step towards the calculation of the moisture induced stresses is the determination of moisture sorption kinetics via the application of an adequate diffusion predictive model. A wide number of models have been proposed in the literature a summary of which can be found in [2, 3]. Among these, the classical Fickian model has shown good correlation with experimental results obtained from tests performed on resin [4, 5] and uni-directionally reinforced polymer matrix [6] coupons for a wide range of temperatures and relative air humidities. This approach is thus employed in the current study as well.

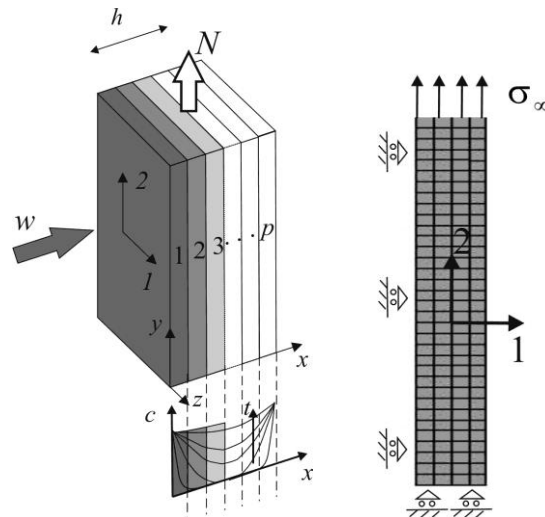
Once the moisture distribution within the structure is determined, the latter can be modelled as a laminate consisting of discrete monolithic layers with different rheological properties. The different properties correspond to the different moisture content of each layer and depend on the constitutive law [7, 8] employed. A solution for the internal stress state can then be obtained by applying classical laminate theory assumptions [9]. Such an approach was

followed in [10, 11] where the problem of moisture viscoelasticity was reduced to the elasticity problem of a layered medium via the application of the Laplace transform.

The objective of the research was to estimate the contribution of viscoelasticity on the stress – strain behaviour of a polymer material subjected to tensile loading during moisture uptake. Given the criticality of the moisture induced internal stresses and the mathematical complexity of the Laplace transform approach [7, 8], a numerical method is used in this study. The latter can easily account for any type of static as well as transient hygroscopic loading conditions and has been validated against finite element solutions.

## 2. Model description

The modelling strategy followed is divided in two main stages. At the first stage, the specimen of an originally uniform material is virtually split into  $p$  plies with different material properties corresponding to the different moisture content of each ply as shown in Fig. 1. Fick’s second law is then used for the determination of the time-dependent ply moisture content. At the second stage, constitutive models are applied at the ply level for the determination of the stress distribution within the layered specimen.



**Figure 1.** Scheme of a specimen divided into  $p$  plies with different material properties corresponding to the different ply moisture contents. ANSYS finite element model and boundary conditions applied.

### 2.1. Modeling of moisture sorption

In this study, sorption is assumed to occur solely by diffusion and classical Fick’s law is employed for the calculation of the moisture distribution within the specimen. Since the specimen’s width and length are an order of magnitude greater than its thickness, one-dimensional diffusion (along the  $x$ -axis in Fig. 1) is considered. The variation of moisture concentration with time can thus be obtained from the following partial differential equation

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial x} \left( D \frac{\partial c}{\partial x} \right), \quad (1)$$

where  $c$  is the moisture concentration and  $D$  is the moisture diffusion coefficient describing the rate of moisture sorption.

Provided that  $D$  is independent of the moisture concentration and  $h$  is the plate thickness the solution for Eq. (1) can be more robustly obtained via the separation of variables method [12]. For initial  $c(0 < x < h, t = 0) = c_0$  and stationary  $c(x = 0, x = h, t > 0) = c_\infty$  boundary conditions, the solution is given from the following series

$$c(x, t) = c_\infty - 2 \frac{(c_\infty - c_0)}{\pi} \sum_{k=1}^{\infty} \frac{(1 - (-1)^k)}{k} \sin(\lambda_k x) \exp(-\lambda_k^2 D t), \quad \lambda_k = \pi k / h. \quad (2)$$

For the case of the same initial condition and non-stationary boundary conditions of oscillatory form that simulate day and year natural cycles i.e.  $c(x = 0, x = h, t > 0) = A_c \sin(\omega t + \Psi) + B_c$ , instead (2) the analytical solution of Eq. (1) is given in [13] and is too lengthy.

## 2.2. Constitutive model

The constitutive model employed consider isotropic linear viscoelastic behaviour and is defined at the ply level. Due to the isotropic material properties, arbitrary rotation of the  $x$ ,  $y$  and  $z$  axes has no effect on the material behaviour, thus the plane of a single ply can be defined by the 1-2 coordinate system shown in Fig. 1. A ply subjected to a plane stress state  $\{\sigma_{11}, \sigma_{22}, \tau_{12}\}$  that varies with time. Assuming that compliance terms  $S_{11}(t)$ ,  $S_{22}(t)$ , and  $S_{66}(t)$  are time-dependent and that Poisson's ratio,  $\nu$ , is time independent, the total ply strains induced by an arbitrary stress history are given by

$$\begin{Bmatrix} \varepsilon_{11}(t) \\ \varepsilon_{22}(t) \\ \gamma_{12}(t) \end{Bmatrix} = \begin{bmatrix} S & -\nu S & 0 \\ -\nu S & S & 0 \\ 0 & 0 & 2(1+\nu)S \end{bmatrix} \begin{Bmatrix} \sigma_{11}(t) \\ \sigma_{22}(t) \\ \tau_{12}(t) \end{Bmatrix} + \begin{Bmatrix} \varepsilon_{11}(t) \\ \varepsilon_{22}(t) \\ \gamma_{12}(t) \end{Bmatrix}_{VE} + w(t) \begin{Bmatrix} \beta \\ \beta \\ 0 \end{Bmatrix} \quad (3)$$

with  $\beta$  is the hygroscopic expansion factor which is a function of the moisture content  $w(t)$ .

In order to eliminate the Volterra-type integrals, the transverse and shear compliances are expressed using Prony series [14, 15] with linear viscoelastic parameters  $S_i, \lambda_i, i = 1 \dots M$ ,

$$\Delta S(t) = \sum_{i=1}^M S_i (1 - e^{-\lambda_i t}). \quad (4)$$

Considering tensile loading along 2-direction (see Fig. 1),  $\varepsilon_{22}(t)$  strain component is given by

$$\varepsilon_{22}(t) = S \sigma_{22}(t) + \sum_{i=1}^M \varepsilon_{i,22}(t) + \beta w(t), \quad \varepsilon_{i,22}(t) = S_{i,22} \int_0^t (1 - e^{-\lambda_i (t - \tau)}) \frac{d\sigma_{22}}{d\tau} d\tau.$$

The total strain of each layer, given by Eq. (3), can be re-written in compact form as

$$\{\varepsilon(t_{j+1})\} = [S_{elast}] \{\sigma(t_{j+1})\} + \{R(t_{j+1})\}.$$

In order to avoid large systems of equations the ply stress state given by the following condensed system of equations

$$\sum_{n=1}^p [S_{elast}]_n^{-1} h_n \{\varepsilon\} = \{N\} + \sum_{n=1}^p [S_{elast}]_n^{-1} h_n \{R\}_n.$$

The previous formulation combined with the sorption kinetics model described in section 2.1, allows for the solution of the creep problem of a polymer plate during moisture uptake. The method described here, was adapted from LAMFLU a FORTRAN computer program previously developed for orthotropic laminates [16, 17].

### 3. Finite element implementation

In order to validate the developed programme LAMFLU described above, linear viscoelastic analyses were performed using the commercial ANSYS finite element code. Due to symmetry, only one fourth of the specimen was modelled using four node layered shell elements that can account for viscoelastic material behaviour. 40 layers were used along the specimen thickness in order to be consistent with the discretization used within LAMFLU programme. The sorption kinetics model described in section 2.1 was implemented into ANSYS parametric design language via a purpose built command script. A typical mesh of the model is shown in Fig. 1 along with the boundary conditions assigned.

### 4. Case study

Stress and strain results were obtained from the application of the proposed numerical method on a rectangular EDT-10 epoxy resin specimen with dimensions  $2 \times 10 \times 150$  mm. The hygroscopic properties used for the stress-strain calculations were taken from [8], whereas the mechanical properties from [18]. In [4], it was shown that moisture sorption of EDT-10 resin follows classical Fick's law for a wide range of atmospheric relative humidities (i.e.  $16 \% \leq \varphi \leq 98 \%$ ) with an average value of hygroscopic diffusivity of  $D = 1.9 \cdot 10^{-6}$  cm<sup>2</sup>/h. It was also shown that moisture sorption isotherm has a nonlinear character and can be approximated by an empirical polynomial function  $w_{\infty}(\varphi) = 0.0004\varphi^2 + 0.0087\varphi$ , where both  $w$  and  $\varphi$  are in %, with the maximum moisture content of the resin being a little less than 5 %.

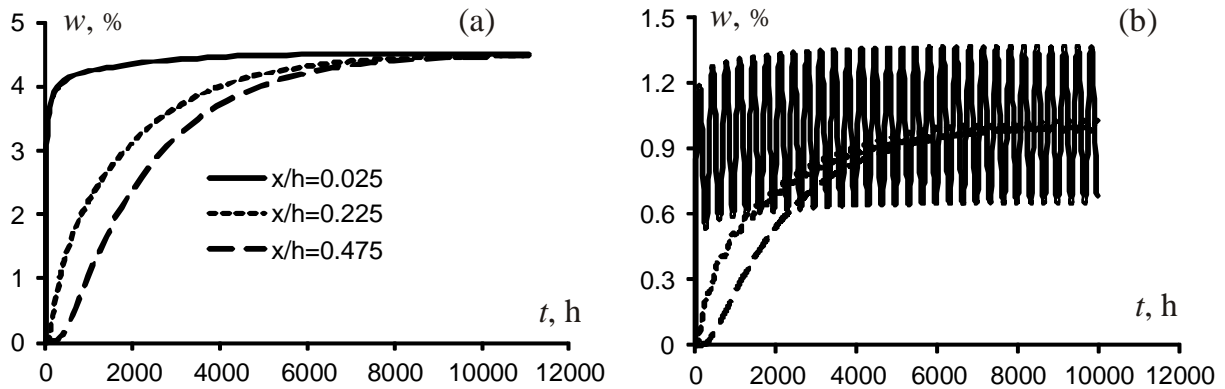
The swelling strain was assumed to be directly proportional to the moisture content of the specimen as in [18] and the hygroscopic expansion factor was  $\beta = 0.2 \%^{-1}$ . The dependence of the elastic and shear moduli on moisture content was approximated by the empirical polynomial function  $\frac{1}{S} = [3.500 + 0.0005067w(t) - 0.01594w^2(t)]$  with initial values 3.5 and 1.2 GPa respectively and the Poisson's ratio was considered constant  $\nu = 0.44$ . Linear viscoelastic behaviour was described with exponential creep function according to Eq. (4) which includes the moisture-time superposition principle with the parameters of the relaxation spectrum given in Table 1. Horizontal moisture shift factor was determined from the direct creep experiments performed in [18] and approximated by the following expression:  $a_w = \exp[-2.0680w(t) + 0.1720w^2(t)]$ . All the properties of the resin were obtained as functions of the moisture content under stationary conditions.

$i$	$\lambda_i, \text{s}^{-1}$	$\lambda_i, \text{MPa}^{-1}$
1	$8.272 \cdot 10^{-5}$	$2.89 \cdot 10^{-6}$
2	$2.051 \cdot 10^{-7}$	$5.23 \cdot 10^{-5}$
3	$5.603 \cdot 10^{-9}$	$1.83 \cdot 10^{-4}$
4	$2.789 \cdot 10^{-10}$	$5.03 \cdot 10^{-4}$
5	$1.870 \cdot 10^{-10}$	$1.53 \cdot 10^{-3}$

**Table 1.** Parameters of relaxation spectrum obtained from [18].

## 5. Results and discussion

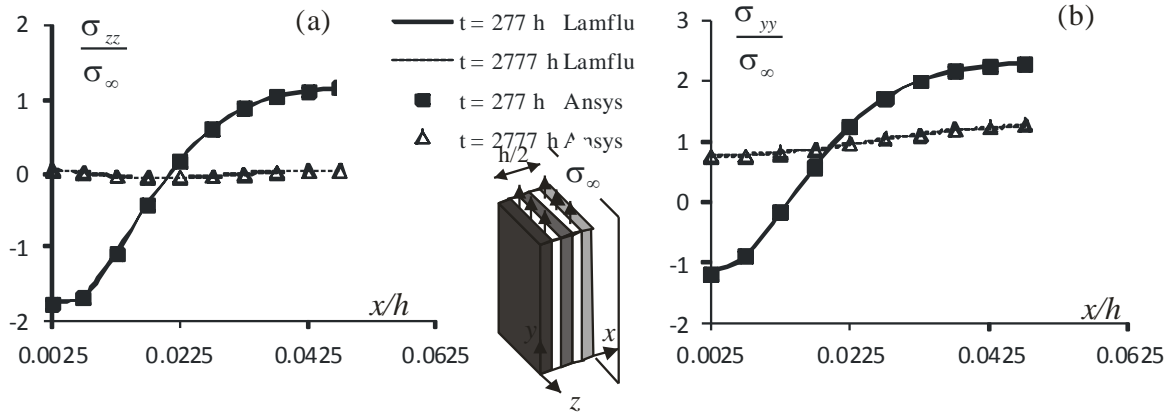
Moisture sorption process was modelled both for stationary and oscillating environments. Figures 2a and 2b show the moisture content versus time variation for three plies that lie at normalized distance  $x/h = 0.025, 0.225$  and  $0.475$  from the outer surface of the specimen and are subjected to stationary and oscillatory moisture conditions respectively. In Fig. 2a the moisture content variation has been calculated for a specimen subjected to constant atmospheric humidity  $\varphi = 95.8 \%$  corresponding to  $w_\infty = 4.5 \%$ . It should be noted that despite the small specimen thickness (i.e.  $h = 2 \text{ mm}$ ), over 10000 hours are needed for moisture equilibrium to be attained and thus for swelling stresses to attenuate. In Fig. 2b, the moisture content variation has been calculated for a specimen subjected to an oscillatory atmospheric humidity corresponding to moisture content of the following form:  $w(t) = 1.5\sin(2\pi/336t) + 3$ . It can be seen that as we move towards the inner plies of the specimen, the moisture oscillation amplitude is damped. In particular, for the specimen thickness  $h = 2 \text{ mm}$  and the oscillatory period  $T = 336 \text{ h}$  considered in this study, the moisture content of the middle ply is solely affected by the stationary moisture component.



**Figure 2.** Moisture content variation with time for plies that lie at normalized distance  $x/h = 0.025, 0.225$  and  $0.475$  from the outer surface of the specimen and are subjected to (a) stationary ( $w_\infty = 4.5 \%$ ) and (b) oscillatory ( $w(t) = 1.5\sin(2\pi/336t) + 3$ ) moisture conditions.

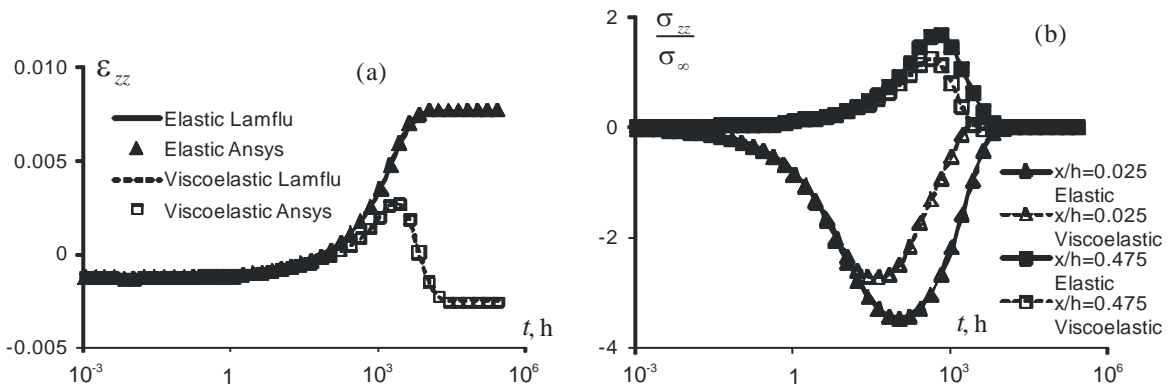
Moisture induced stresses in the specimen section change with time following moisture concentration. Figures 3a and 3b show the variation of the normalized transverse ( $\sigma_{zz}/\sigma_\infty$ ) and longitudinal ( $\sigma_{yy}/\sigma_\infty$ ) stresses across the plate thickness as calculated at time instants  $t = 277$  and  $2777 \text{ h}$  respectively. These results correspond to a specimen subjected to uniform tensile load  $\sigma_\infty$  and constant atmospheric humidity  $\varphi = 95.8 \%$ . It should be noted that despite no external loads act along  $z$ -direction, a variable transverse stress profile arises across the specimen thickness at  $t = 277 \text{ h}$  as shown in Fig. 3a. This stress profile evens out as moisture equilibrium is gradually restored within the specimen (see stress distribution at  $t = 2777 \text{ h}$ ). A

similar trend is also observed for the tensile stresses shown in Fig. 3b. In both figures, ANSYS and LAMFLU are shown to be in excellent agreement proving the developed design tool to be mathematically accurate. Moreover, LAMFLU is shown to be as time-efficient as ANSYS since the time required to obtain the solution is of the same scale (i.e. both run within minutes).



**Figure 3.** Variation of the normalized viscoelastic (a) transverse ( $\sigma_{zz}/\sigma_\infty$ ) and (b) longitudinal ( $\sigma_{yy}/\sigma_\infty$ ) stresses across the specimen thickness as obtained from LAMFLU (lines) and ANSYS (symbols). The specimen is subjected to a uniform tensile load ( $\sigma_\infty$ ) and constant atmospheric humidity  $\phi = 95.8\%$ .

The time variations of the transverse  $\epsilon_{zz}$  strains are plotted in Fig. 4a. The latter were calculated by performing elastic and viscoelastic analyses on a specimen subjected to uniform tensile load  $\sigma_\infty = 10$  MPa and constant atmospheric humidity  $\phi = 95.8\%$ . Figure 4a shows that  $\epsilon_{zz}$  strains from both elastic and viscoelastic analyses are initially compressive since the elastic term dominates over the viscoelastic and the swelling terms. With the increase in the specimen moisture content, swelling strains start to develop causing the sample to expand. In the elastic case, the sample continues to swell (i.e.  $\epsilon_{zz}$  strain increases) until moisture equilibrium is attained. However, in the viscoelastic case, viscoelastic compressive strains dominate over swelling strains for  $t \geq 5 \cdot 10^3$  h, causing the specimen to contract in the lateral direction.



**Figure 4.** Time variation of (a) transverse ( $\epsilon_{zz}$ ) strains as obtained from elastic and viscoelastic analyses performed using LAMFLU (lines) and ANSYS (symbols). Time variation of normalized (b) transverse ( $\sigma_{zz}/\sigma_\infty$ ) stresses as obtained from elastic (solid lines, black symbols) and viscoelastic (dashed lines, white symbols) analyses. The specimen is subjected to a uniform tensile load  $\sigma_\infty$  and constant atmospheric humidity  $\phi = 95.8\%$ .

As a result, viscoelastic analysis shows that at moisture equilibrium, the sample has laterally contracted whereas the elastic one that it has expanded. However, when viscoelastic behaviour is considered, longitudinal strains at moisture equilibrium are found to be higher than those obtained from the elastic case by a factor of four. It is therefore apparent that neglecting the moisture induced viscoelastic behaviour of polymer structures, will lead to erroneous design especially when strain based criteria are employed.

Figure 4b shows the time variation of the elastic and viscoelastic transverse ( $\sigma_{zz}/\sigma_{\infty}$ ) stresses that develop within the outer and the middle plies. It should be noted that these two plies represent the two extremes with the middle ply being under tensile stresses and the outer ply being predominantly under compressive stresses. The stress variations from both elastic and viscoelastic analyses are shown to follow the same trend, however, the time instant at which they attain their maximum or minimum values differs. Moreover, during moisture uptake, viscoelastic stresses are shown to be lower in magnitude than the corresponding elastic since moisture induced viscoelastic behaviour leads to stress relief.

Oscillation of humidity in ambient environment leads to essential changes in stress – strain behavior of the specimen. It should be noted that although the moisture content of the middle ply does not oscillate with time, both longitudinal and transverse stresses are of an oscillatory form. This is because the portion of the load that is carried by the middle ply depends on that carried by the other plies. Since stresses in the outer plies are of oscillatory form (due to the oscillatory moisture content), so should be stresses in the middle ply. It is apparent that structures subjected to oscillatory moisture conditions should be designed against fatigue issues.

## **6. Conclusions**

A robust and efficient numerical method for the calculation of the internal stress state that develops within structures subjected to mechanical and steady state or transient hygroscopic loading conditions has been developed. The method has been validated against finite element analyses and was shown to be mathematically accurate. Results from the application of the proposed method on an epoxy polymer binder indicate that:

- Moisture induced viscoelastic behaviour should be taken into account when designing polymer structures especially if strain based criteria are to be employed. This is because viscoelastic strains are shown to be up to four times greater than those calculated via elastic analysis.
- A variable stress profile arises within a polymer structure over a significant part of its life span with the stresses at some time instant being several times greater (up to three times in this case study) than the externally applied stress field (i.e. the far field load).
- Structures subjected to oscillatory moisture conditions should be designed against fatigue due to the significant stress oscillation within the structure.
- Due to the iterative procedure of the developed method, viscoelastic material behaviour can be incorporated in a straight forward manner.

**Acknowledgement.** Andrey Anishevich acknowledges ESF project No. 2013/0017/1DP/1.1.1.2.0/13/APIA/VIAA/063 for the financial support.

## **References**

- [1] Y.J. Weitsman and M. Elahi. Effects of fluids on the deformation, strength and durability of polymeric composites – an overview. *Mechanics of Time-Dependent Materials*; 4:107-126, 2000.
- [2] J. Yagoubi, G. Lubineau, F. Roger, J. Verdu. A fully coupled diffusion-reaction scheme for moisture sorption–desorption in an anhydride-cured epoxy resin. *Polymer*; 53 :5582-5595, 2012.
- [3] T.I. Glaskova, R.M. Guedes, J.J. Morais, A.N. Aniskevich. A comparative analysis of moisture transport models as applied to an epoxy binder. *Mechanics of Composite Materials*; 43 (4):377-388, 2007.
- [4] A.N. Aniskevich. Experimental study of sorption of moisture in ÉDT-10 epoxy binder. *Mechanics of Composite Materials*; 20 (6):670-674, 1985.
- [5] Y. He. Moisture absorption and hygroscopic swelling behavior of an under fill material. *Thermochimica Acta*; 546:143-152, 2012.
- [6] A.N. Aniskevich. Moisture absorption and desorption by a unidirectional organoplastic under steady temperature-humidity conditions. *Mechanics of Composite Materials*; 22 (3):383-388, 1986.
- [7] Y. Weitsman. Hygrothermal viscoelastic analysis of a resin-slab under time-varying moisture and temperature. In: *Structures, Structural Dynamics and Materials Conference, 18th*; 213-218, 1977.
- [8] R. Schapery. Viscoelastic behavior and analysis of composite materials. *Mechanics of Composite Materials* (A 75-24868 10-39) New York, Academic Press, Inc.;85-168, 1974.
- [9] R.M. Jones. *Mechanics of Composite Materials*: Taylor & Francis London; 1975.
- [10] E.Z. Plume, R.D. Maksimov. Nonsteady stresses in a polymer plate induced by moisture. *Mechanics of Composite Materials*; 28 (2):114-120, 1992.
- [11] E.Z. Plume. Calculation of moisture stresses in a viscoelastic crossplied composite. *Mechanics of Composite Materials*; 30 (4):494-501, 1994.
- [12] J. Crank. *The Mathematics of Diffusion*. Second ed. Oxford: Clarendon Press; 1975.
- [13] O.A. Plushchik, A.N. Aniskevich. Diffusion moisture sorption by an isotropic polymer material in atmosphere with stationary and nonstationary humidity. *Mechanics of Composite Materials*; 38 (2): 223-245, 2002.
- [14] K.C. Gramoll, D.A. Dillard, H.F. Brinson. A stable numerical solution method for in-plane loading of nonlinear viscoelastic laminated orthotropic materials. *Composite Structures*; 13:251-274, 1989.
- [15] J.A. Czyz, W. Szyszkowski. An effective method for non-linear viscoelastic structural analysis. *Computers & Structures*; 37:637-646, 1990.
- [16] R.M. Guedes, A.T. Marques, A. Cardon. Analytical and experimental evaluation of nonlinear viscoelastic-viscoplastic composite laminates under creep, creep-recovery, relaxation and ramp loading. *Mechanics of Time-Dependent Materials*; 2:113-128, 1998.
- [17] A.N. Aniskevich, R.M. Guedes. Viscoelastic stress-strain analysis during moisture uptake under tensile creep. *17th International Conference on Composite Materials*; Edinburgh, UK; 2009.
- [18] A.N. Aniskevich, Y.O. Yanson, N.I. Aniskevich. Creep of epoxy binder in a humid atmosphere. *Mechanics of Composite Materials*; 28 (1):12-18, 1992.