Micromechanical Viscoelastic Analysis of Flax Fiber Reinforced Bio-Based Polyurethane Composites

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ABSTRACT: In this study, a novel, bio-based polyol was used in the formulation of a polyurethane (PU) matrix for a composite material where flax fiber was used as the reinforcement. The viscoelastic properties of the matrix and flax fiber were determined by a linear viscoelastic model through experimentation and the results were used as input for the material properties in the computational model. A finite element micromechanical model of a representative volume element (RVE) in terms of repeating unit cells (RUC) was developed to predict the mechanical properties of composites. Six loading conditions were applied on the RUC to predict and define the viscoelastic behavior of the composite unit cell. The time-history of averaged response was determined in terms of stress and strains. The results of this study suggest that applying the overall rate-dependent behavior of flax fiber to the micromechanical model leads to a good agreement between the micromechanical modeling and experimental results. The modeling approach is efficient and accurate as long as the periodicity in the composite rules. This modeling approach can be used as a powerful algorithm in determining linear and nonlinear properties in material mechanics analysis and characterization.

KEYWORDS: Bio-based polyurethane, flax, micromechanical analysis, stress relaxation, viscoelastic

1 INTRODUCTION

Using natural fibers as reinforcement in polymers as a composite is gaining high interest for applications in diverse fields from appliances to spacecraft, due to their sustainability and economic benefits [1,2]. Flax fiber is one of the most interesting fibers used in bio-based composites due to its high specific strength and stiffness [3], as well as biodegradability and easy processing benefits [4]. Both the natural fiber and the matrices, which are the main components of biocomposite materials, show mechanical properties which are time dependent, i.e., exhibiting viscoelastic behavior. Therefore, predicting the overall rate-dependent mechanical behavior of bio-based composites requires study of the viscoelastic behavior of both matrix and fiber. While there have been numerous studies predicting the mechanical properties of natural fibers and bio-based resins, including elastic stiffness or strength, their viscoelastic behavior has rarely been addressed. Viscoelastic composites have broad applications in sports equipment, medical tools, housing, aerospace, and civil infrastructure. Viscoelasticity is a time-dependent behavior, which is difficult to be measured conveniently. Therefore, the prediction of the viscoelastic behavior of the composites has gained the attention of many researchers. Recent numerical as well as analytical modeling has been carried out and much experimental work has been performed to determine the viscoelastic properties of composites.

Micromechanical methods are the most common approach to determine the viscoelastic behavior of composite materials from known properties of their constituents through a repeating unit cell (RUC) which is a representative volume element (RVE) of the composite. The RUC consists of both fiber and matrix with a predetermined fiber packing. Several different RUC models have been introduced in the literature based on the geometrical features of composites, including rectangular unit cells for square, hexagonal, unidirectional random, and bidirectional crossed packing of fibers [5]. Naik *et al.* [5] performed a comparison study between different RUC fiber packings and found that square

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packing geometry is stiffer than hexagonal packing. This could be attributed to the fact that there are no fibers split between unit cells, therefore, significant weak planes are created within the upscaled material model that is comprised of many RUCs. In fact, the RUC provides a simulation of a continuum point behavior. Therefore, the most important feature of all these RUCs is the repetitiveness of RUCs throughout the composites. The main idea in the micromechanical approach is that the inhomogeneous structure of the composite is replaced by a homogeneous domain. Moreover, the micromechanical analysis not only provides the overall mechanical properties of the composite materials, but also allows for the investigation of different mechanism such as damage initiation, propagation, and failure [5]. Most micromechanical methods use periodic homogenization, which approximates the composite by periodic phase arrangements. Then the response of the unit cell under specified loading conditions and appropriate boundary conditions is analyzed over time to measure the composite's viscoelastic response [6–8]. Several micromechanical approaches have been developed for predicting the viscoelastic response of unidirectional fiber composites using the fiber and matrix properties [8–13]. It has been shown that considering the effects of time-dependent or viscoelastic properties (as demonstrated by creep and stress-relaxation tests) of polymeric resins provides a better prediction for the mechanical properties of the composite material. A wide range of constitutive relations from linear viscoelastic to nonlinear-viscoelastic (for matrix) and linear elastic for fiber have been considered for determining the material properties [5,14–18].

In this work, a finite element analysis (FEA) study was carried out to examine the viscoelastic behavior of unidirectional bio-based composites of polyurethane (PU) resin reinforced with three different fiber volume fractions of flax fiber. To the authors' best knowledge, previous studies have not accurately addressed the viscoelastic behavior of flax fibers in determining the composites' ultimate properties. This study, therefore, considers both flax fiber and bio-based PU resin as linear viscoelastic materials to obtain more accurate prediction of the mechanical properties of the composite using the characteristics of its constituents, i.e., micromechanical analysis. The generalization of two micromechanical analyses, including the FEA and analytical equations to viscoelastic behavior, were analyzed to predict the relaxation behavior of the composite material. To rely on micromechanical analysis, the particular approach must be validated through comparison to experimental data. Hence, experimental tests were performed to verify the numerical and algebraic equations.

2 METHOD

In this study, the basic finite element models employed in the simulation of the composite deformation are described by the linear viscoelastic model for the matrix and flax fiber. Under the small deformation assumption, a linear relationship between the strains and stresses is assumed, which can be expressed by the following Voigt vector form:

$$\begin{Bmatrix} \varepsilon_{11}(t) \\ \varepsilon_{22}(t) \\ \varepsilon_{33}(t) \\ \varepsilon_{12}(t) \\ \varepsilon_{13}(t) \\ \varepsilon_{23}(t) \end{Bmatrix} = \int_0^t \begin{bmatrix} S_{1111} & S_{1122} & S_{1133} & S_{1112} & S_{1113} & S_{1123} \\ S_{2211} & S_{2222} & S_{2233} & S_{2212} & S_{2213} & S_{2223} \\ S_{3311} & S_{3322} & S_{3333} & S_{3312} & S_{3313} & S_{3323} \\ S_{1211} & S_{1222} & S_{1233} & S_{1212} & S_{1213} & S_{1223} \\ S_{1311} & S_{1322} & S_{1333} & S_{1312} & S_{1313} & S_{1323} \\ S_{2311} & S_{2322} & S_{2333} & S_{2312} & S_{2313} & S_{2323} \end{bmatrix} \begin{Bmatrix} d\sigma_{11}(\tau)/dt \\ d\sigma_{22}(\tau)/dt \\ d\sigma_{33}(\tau)/dt \\ d\sigma_{12}(\tau)/dt \\ d\sigma_{13}(\tau)/dt \\ d\sigma_{23}(\tau)/dt \end{Bmatrix} d\tau \quad i,j,k,l = 1,2,3 \tag{1}$$

The 6 × 6 matrix of compliance coefficients is a function of time, i.e., $S_{ijkl} = S_{ijkl}$ (t) at any particular time t, and strains can be related to stresses by $\varepsilon_{ij} = S_{ijkl}(t)\sigma_{kl}$. Therefore, the compliance coefficients and the related engineering constants should be characterized at each individual time step.

To this end, instantaneous elastic modulus and stress relaxation parameters of the constituents were determined using experimental tests for both flax fiber and bio-based PU. The mechanical properties of the composite in the micromechanical analysis strongly depend on the accuracy of the stress relaxation parameters. Therefore, in order to achieve reliable results, measurements were taken using a Dia-Stron miniature tensile tester to measure stress relaxation parameters for the flax fiber. Methods developed by Dia-Stron for single fiber testing require the fiber to be mounted on a tab system. There is an adhesive-based tab system, which has proved suitable for vegetable fibers, carbon, etc. Dia-Stron miniature tensile tester is generally designed to study the effects of different treatments on hair strength and it has been recently used to study tensile testing, dimensional properties, failure analysis, and bending moment for different kinds of fibers. In this study, it was used for the stress relaxation test of flax fibers. The stress relaxation parameters from this test were used as the input for the finite element-based micromechanical analysis in ABAQUS.

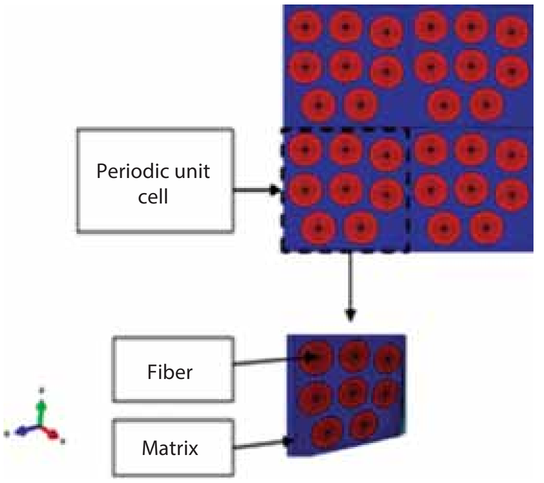

Figure 1 A rectangular repeating unit cell representing unidirectional composites in random packing. The fibers are in direction 1.

Figure 1 shows the meshed unit cell of fiber arrangement representing periodic microstructure of a unidirectional composite. The fibers with the same diameter are distributed randomly in the matrix domain in a unidirectional direction. The unit cell section consists of eight cylindrical fibers. Depending on the fiber volume fraction, different diameter values are used for the circular cross section of fibers. Due to the assumption of perfect fiber and matrix bonding, it is believed that the shape of fibers' cross section might not affect the numerical results of the modeling.

For the inverse analysis, each RUC is exposed to six independent load cases (three axial and three shear loads). Load cases 1, 2, and 3 are axial forces in direction 1, 2, and 3, which are associated with σ_{11}, σ_{22}, and σ_{33} respectively. Load cases 4, 5 and 6 are shear loads, which are associated with τ_{12}, τ_{13}, and τ_{23} respectively. The six sets of analyses provide six distinct sets of time history stress-strain data for a typical transversely isotropic material, and 36 sets of data for a heterogeneous material. For every load case, the localized stresses and strains can then be volume-averaged over RUC volume for the homogenization procedure at each time step to obtain time-dependent averages, as described by the following equation, where v is the volume of the RUC. The reason pertains to the assumption that the RUC is presenting a point within the continuum domain, and therefore has homogenized material properties.

$$\begin{cases} \sigma_{ij}(t) = \dfrac{1}{V}\displaystyle\int_v \sigma_{ij}(t)dv \\ \varepsilon_{ij}(t) = \dfrac{1}{V}\displaystyle\int_v \varepsilon_{ij}(t)dv \end{cases} \tag{2}$$

2.1 Stress Relaxation Loading and Periodic Boundary Conditions

In the current study, six load cases are applied on the unit cell in the form of ramped constant strains (displacement). In order to observe the stress relaxation behavior, the displacement is kept constant throughout the time domain. The periodic boundary conditions require the opposite faces of the unit cell to undergo identical deformation. Thus, constraint relations between the nodes on the parallel faces are used to enforce periodicity constraints on the RUC. For detailed implementation of periodicity constraints, as well as the rigid body conditions on a RUC, one can refer to Garnich and Karami [19], Naik *et al.* [5] and Javid *et al.* [20].

3 EXPERIMENTAL PROCEDURE

3.1 Materials and Sample Preparation

To achieve a high fiber volume fraction composite, a compression molding process with hand layup technique was used to prepare the flax-reinforced composite panels. The unidirectional linen flax fiber, with a specific gravity of 1.4, was provided by Composites Evolution Ltd., Chesterfield, UK. The flax fiber was used as received, without any chemical treatment. The methoxylated sucrose soyate polyol-based (MSSP-based) PU was used as the thermoset resin system. The authors' previous studies report a detailed description of the synthesis of MSSP [21,22]. The PU resin was prepared by mixing the bio-based high functional MSSP polyol with polyisocyanate crosslinker, based on a modified diphenylmethane diisocyanate (MDI) with NCO:OH ratios of 1.0:1.0, which is the optimum formulation reported by Nelson *et al.* [22]. The polyisocyanate, Baydur, was obtained from Bayer Material Science. To ensure the resin was uniformly distributed throughout the fibers, each layer of the fiber fabric was presoaked by the matrix and then they were stacked upon each other. The composite plates were manufactured in a 100 mm × 200 mm compression mold. A pressure of 3 MPa was applied for 12 hours at 23°C.

3.2 Material Input

The main objective of the stress relaxation test was to characterize time-dependent behavior of the PU and the flax fiber under tensile forces. In the stress relaxation method, the specimen is subjected to an increasing force until the specified initial strain is attained. For the duration of the test, the specimen constraint was maintained constant. To obtain sufficient sensitivity, the gauge length of the specimens should be as long as possible. Moreover, to avoid the primary fluctuations in the results, before the stabilization of strain, the initial crosshead speed had a sharp ramp as long as it was practical and the strain remained in the elastic region. Once the desired strain was reached, it was held constant and the time history of the stress decay was recorded until the material was fully relaxed. Prony series expansion can be used to define the viscoelastic behavior of materials by using the stress relaxation test. For the linear viscoelastic material, the time-dependent shear modulus, i.e., G(t), is expressed by the following Prony series as:

$$G(t) = G_0\left(1-\sum_{i=1}^{n} g_i\,(1-e^{\frac{-t}{\tau_i}})\right) \quad (3)$$

where G_0 is instantaneous shear modulus and g_i, and τ_i are material-dependent coefficients.

The stress relaxation behavior of the polyurethane matrix was measured using a standard method for measuring tensile strength (ASTM D3039) with 2 mm/min crosshead speed until a strain of 1% was reached. There were some limitations for the equipment and the grips that were used for measuring the stress relaxation for PU matrix by Instron machine, therefore, the crosshead speed was chosen as 2 mm/min, which was the fastest speed that was practical to maintain the 1% constant constraint for PU resin, with the available grips.

The mechanical properties of the flax fiber strongly depend on the accuracy of the cross-section area measurement. Using an approximate value for the cross-sectional area can significantly underestimate the value of tensile and stress relaxations. Therefore, in order to have accurate stress relaxation values, concise cross-sectional area measurements are required. To this end, the Dia-Stron miniature tensile tester (Dia-Stron Ltd. Andover, UK) was used at 23°. One key to the Dia-Stron's accuracy is the application of a laser scanner right into the analyzer package and a sample rotation feature which facilitates the measurement of the elliptical fiber cross section all along the fiber length. Stress relaxation tests were performed on 15 single fibers using the Dia-Stron with 0.4 mm/sec crosshead speed until the strain reached 1.5%, while the fibers were in the elastic region. The load was applied under computer control and, for each single fiber, the load against extension was recorded. Using measured fiber diameters and a fixed gauge length of 4 mm, the data were converted to stress against strain.

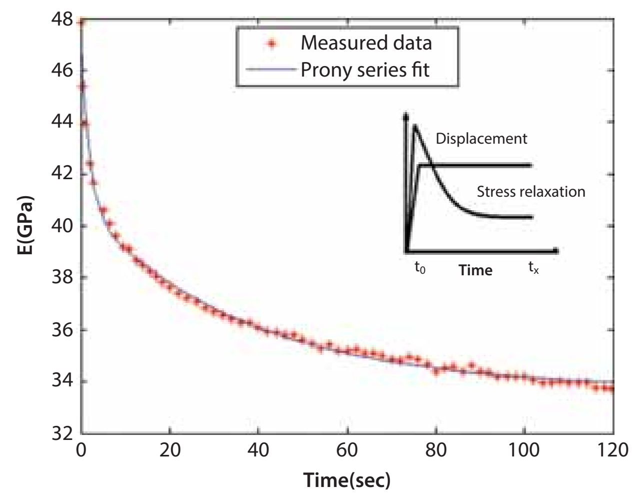

Figure 2 Measured and fitted relaxation of modulus for flax fiber under the strain 1.5%.

4 RESULTS AND DISCUSSION

4.1 Viscoelastic Properties of the Fiber and Matrix

Viscoelastic properties of the flax fiber and matrix were determined by the stress relaxation test for both fiber and matrix. Figure 2 shows the measured data and the Prony series fit for the flax fiber. As explained above, in the stress relaxation method, the specimen is subjected to an increasing force until the specified initial strain is attained. For the duration of the test, the specimen constraint was maintained constant. Characteristic behavior during the force application period in a relaxation test is shown at the top right corner of Figure 2. Having determined the averaged stress-relaxation moduli as functions of time, the best fit to each coefficient curve could be reached by finding the Prony series coefficients using a curve fitting optimization in ABAQUS. A total of three parameters and two parameters were considered for defining the Prony series for the PU matrix and flax fiber, respectively, using ABAQUS in order to obtain the best fit. The parameters of the Prony series are summarized in Table 1. According to experimental modulus relaxation data, the instantaneous modulus are $E_0^f = 47.84\ GPa$, and $E_0^m = 1.24\ GPa$, for flax fiber and bio-based PU, respectively, as shown in Figure 2. For isotropic materials, such as most fibers and matrices, $G_0^f = E_0^f / 2(1+v_0^f)$ and E_0^f and other parameters of the Prony series were calculated by ABAQUS after fitting the relaxation modulus curve with the Prony series. The Poisson's ratios were reported as constant values as $v_0^f = 0.3$ for most fibers and $v_0^m = 0.4$ for most thermoset resins [5]. The same values were

Table 1 Prony series parameters for the PU matrix and flax fiber.

Material	i	g_i	τ_i
Matrix	1	0.026	0.074
	2	0.094	41.28
	3	0.013	628.13
Fiber	1	0.148	1.74
	2	0.147	36.63

considered for flax fibers and PU resins, except, these values were considered instantaneous Poisson's ratio and as a time-dependent property.

4.2 Local Stress Distribution in the Flax/PU Composite by Numerical Model

For the flax/PU composite, the ABAQUS finite element package was employed to analyze the selected RUC under six different loading conditions [23]. The stress distribution profiles for the RUC of the 50% flax/PU composite are shown in Figure 3 for load cases 1, 2, and 4 at two different times of 1 s and 100 s. The RUC was subjected to a 0.2% uniform stretch in the direction of fibers. The stress distribution was seen to vary with time due to the viscoelastic behavior of the flax fiber and PU composite. As shown in Figure 3, the stress distribution contours indicate that while the elements located in the fibers have the same stress, they predict higher stresses than those elements in the matrix domain. The stresses were seen to be distinctly uniform in each domain of the fibers and matrix. This is asserted to be due to the uniform stretch of all elements in the longitudinal direction.

Using the volume average subroutine program interfaced with ABAQUS, resultant composite compliances were calculated and their time histories were plotted, as shown in Figure 4. Different crystal systems can be characterized exclusively by their symmetries. For instance, in highly anisotropic materials, any one component of stress can cause strain in all six components, and in an isotropic case, the elastic constants are reduced to two constants. Due to symmetry and the fact that both fiber and matrix are assumed isotropic, the compliance coefficients are reduced to 13 (from the total of 36 for the anisotropic case). The predicted compliance coefficients, which resulted from various volume elements, showed thirteen distinct coefficients, an indication of a monoclinic isotropic material behavior for this case. Since some of these coefficients have very close values and some of them are negligible compared to others, a transverse isotropic material behavior with five distinct compliance coefficients (Table 2) was observed. In addition, the mechanical properties for composites at different time steps were calculated from the compliance coefficients and are reported in Table 3.

4.3 Analytical Approach

In order to theoretically determine the stress tensor in polymer materials, a function is needed to capture the effects of the time-dependent behavior in these materials. According to Lu H *et al.* [24], the Poisson's ratio of polymers is a time-dependent variable. They showed if Poisson's ratio is assumed to be constant, theoretical values of the stress tensor are poorly confirmed by experimental results. The stress tensor for a linear viscoelastic polymer material can be defined as [25]:

$$\sigma_{ij(t)} = \frac{E_0}{3(1-2v_0)}\varepsilon_{kk(t)}\delta_{ij} + \frac{E_0}{1+v_0}(e_{ij(t)} - \int_0^t R_{(t-\tau)} e_{ij(t)} d\tau) \quad (4)$$

where $\sigma_{ij(t)}$ and $\varepsilon_{kk(t)}$ are stress and strain tensors, respectively. $R_{(t-\tau)}$ is the relaxation kernel, $e_{ij(t)}$ is the deviatoric strain ($e_{ij(t)} = \varepsilon_{kk(t)} - \delta_{ij}/3$), and v_0 and E_0 are the instantaneous Poisson's ratio and elastic modulus, respectively. The relaxation function can be described by a Prony series as defined in Equation 3.

By applying a constant value of ε_0 at time t=0 in Equation 4, along the fiber direction, the axial stress relaxation behavior of polymer materials along the direction of extension and the Poisson's ratio for linear isotropic materials can be determined as:

$$\sigma_{11(t)} = \frac{E_0\varepsilon_0}{3}[\frac{1-2v_{(t)}}{1-2v_0} + \frac{2\left(1+v_{(t)}\right)}{1+v_0} - \frac{2}{1+v_0} \cdot \int_0^t R_{(t-\tau)}\left(1+v_{(\tau)}\right)d\tau] \quad (5)$$

$$v_t = v_0 + \frac{1-2v_0}{3}\int_0^t R_{(t-\tau)}\left(1+v_{(\tau)}\right)d\tau \quad (6)$$

where $v_{(\tau)}$ is the time-dependent Poisson's ratio which depends on time. By substituting Equation 6 in Equation 4, the axial stress along to the extension direction is determined as:

$$\sigma_{11(t)} = E_0\varepsilon_0 \frac{1-2v_{(t)}}{1-2v_0} \quad (7)$$

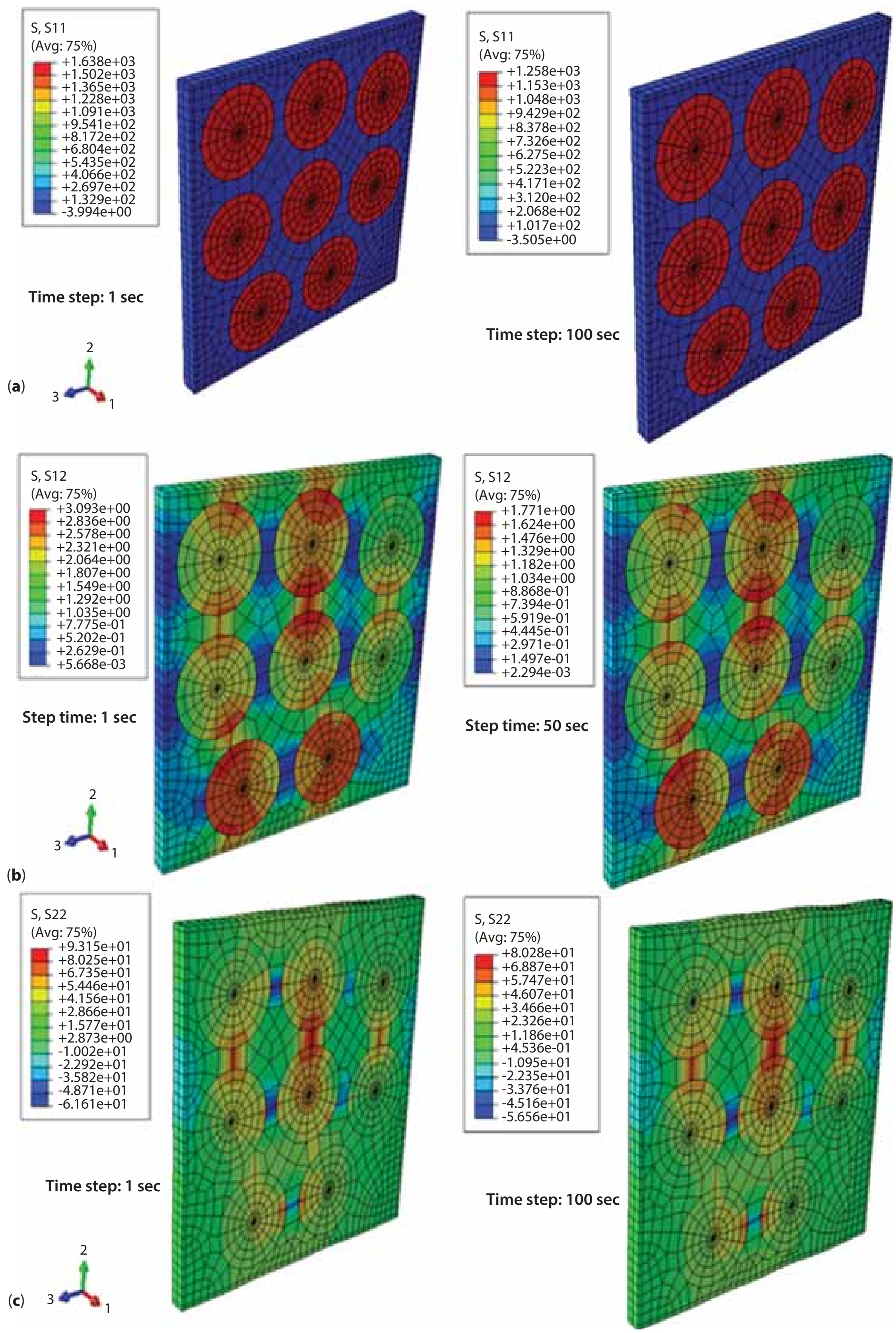

Figure 3 Stress distribution contours within the deformed shape of RUC of flax/PU composite with fiber volume of 50% under 0.2% strain at time of 1 and 100 s for (a) load case 1 (S_{11}), (b) load case 4 (S_{12}), and (c) load case 2 (S_{22}).

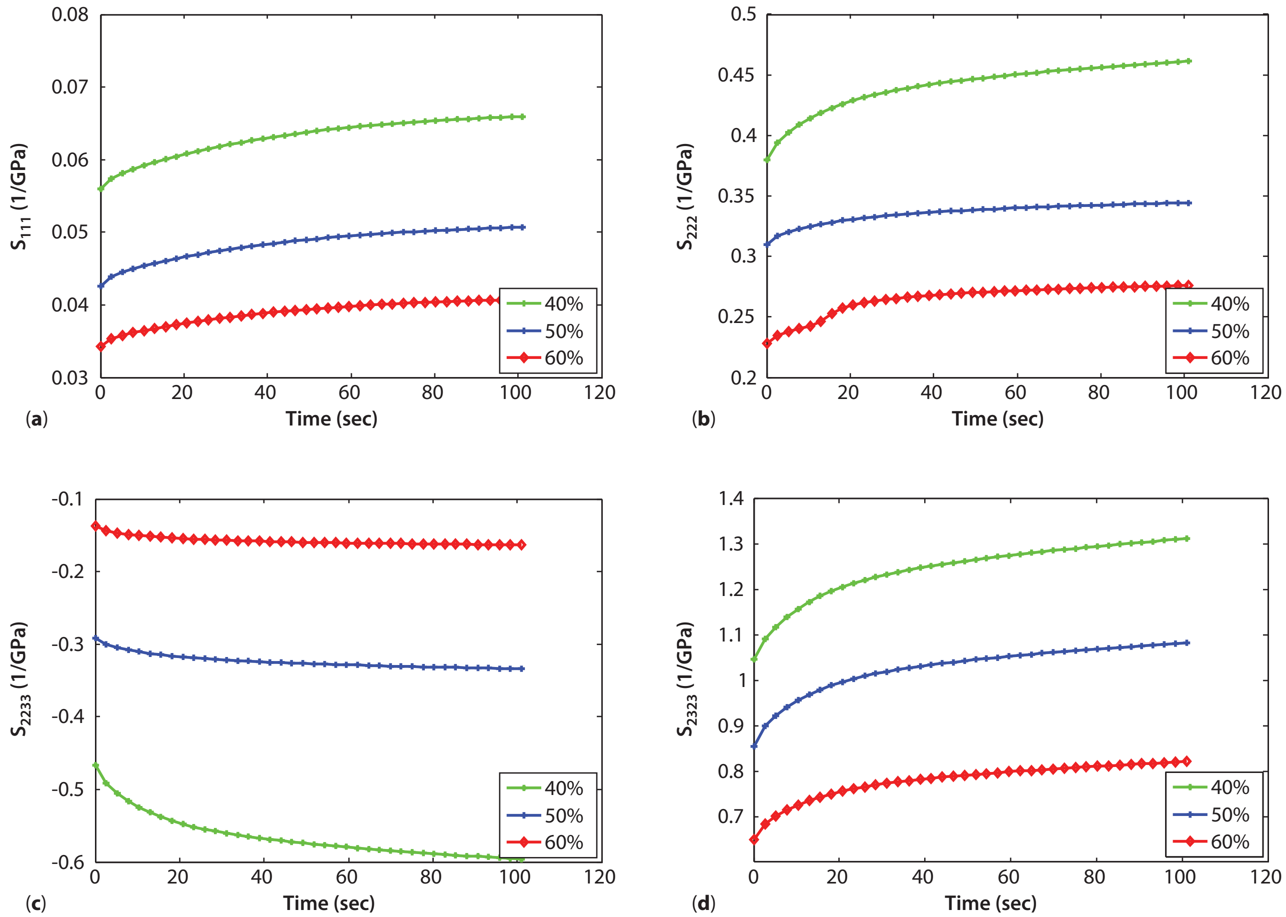

Figure 4 Compliance coefficients of Flax/PU composite for different fiber volume fractions (%): (a) S_{1111}, (b) S_{2222}, (c) S_{2233}, (d) S_{2323}.

Table 2 Compliance coefficients, 1/GPa of Flax/PU composite for different fiber volumes of 40%, 50%, and 60% at three different times at 1 sec, 50 sec, and 100 sec.

t(s)	V_f (%)	S_{1111}	S_{2222}, S_{3333}	S_{1212}, S_{1313}	S_{2323}	S_{2233}	S_{3322}
1	40	0.06	0.39	0.99	1.07	–0.48	0.24
	50	0.04	0.31	0.85	0.88	–0.30	0.17
	60	0.03	0.23	0.45	0.67	–0.14	0.13
50	40	0.06	0.45	1.18	1.26	–0.57	0.25
	50	0.05	0.34	1.03	1.04	–0.33	0.18
	60	0.04	0.27	0.54	0.79	–0.16	0.14
100	40	0.07	0.46	1.23	1.31	–0.60	0.25
	50	0.05	0.34	1.07	1.08	–0.33	0.18
	60	0.04	0.28	0.56	0.82	–0.16	0.14

Table 3 The predicted mechanical properties of Flax/PU using micromechanical model.

		E_{11}	E_{22}	E_{33}	G_{12}	G_{13}	G_{23}			
t (s)	V_f **(%)**	**(GPa)**						ν_{12}	ν_{13}	ν_{32}
1	40	18.86	2.58	2.53	1.00	1.02	0.94	0.42	0.48	0.52
	50	23.12	3.18	3.15	1.17	1.15	1.14	0.40	0.39	0.50
	60	28.75	4.31	4.29	2.21	2.23	1.49	0.37	0.36	0.48
50	40	16.77	2.23	2.17	0.84	0.86	0.79	0.43	0.48	0.53
	50	20.36	2.96	2.92	0.97	0.96	0.96	0.41	0.40	0.51
	60	25.33	3.70	3.95	1.84	1.84	1.26	0.39	0.38	0.50
100	40	16.22	2.17	2.10	0.81	0.83	0.76	0.44	0.48	0.53
	50	19.73	2.90	2.87	0.93	0.93	0.92	0.41	0.41	0.52
	60	24.54	3.62	3.89	1.76	1.77	1.22	0.39	0.38	0.51

4.4 Longitudinal Loading on the Composites

A constant axial strain ε_0 in the direction of 1 was applied on the unit cell shown in Figure 1. Both fiber and matrix were stretched by the same amount in the fiber direction, as a perfect bond between the flax fiber and matrix has been assumed. Therefore, the total stress in the flax/PU composite material is obtained by summation of forces in the fiber and matrix:

$$\sigma_{11(t)} = (E_0^f V^f \frac{1-2v_{(t)}^f}{1-2v_0^f} + E_0^m V^m \frac{1-2v_{(t)}^m}{1-2v_0^m})\varepsilon_0 \qquad (8)$$

where V^f and V^m are the volume fractions of the fiber and matrix, respectively. Using the Laplace transformation, the Poisson's ratio in Equation 6 becomes:

$$v_{(t)} = v_0 + \sum_{i=1}^{n} \gamma_i (1 - e^{\frac{-t}{\vartheta_i}}) \qquad (9)$$

where γ_i and ϑ_i depend on g_i and τ_i and ν_0. These values are calculated and reported in Table 4.

By substituting Equation 10 in Equzation 9, the axial stress in the composite material is determined as follows:

$$\frac{\sigma_{11(t)}}{\varepsilon_0} = (E_0^f V^f [1 - \frac{2}{1-2v_0^f} \sum_{i=1}^{n} \gamma_i \left(1 - e^{\frac{-t}{\vartheta_i}}\right) + E_0^m V^m$$

$$[1 - \frac{2}{1-2v_0^m} \sum_{i=1}^{n} \gamma_i \left(1 - e^{\frac{-t}{\vartheta_i}}\right)] \qquad (10)$$

Table 4 Time-dependent parameters of Poisson's ratio for the PU matrix and flax fiber.

Material	**i**	γ_i	ϑ_i
Matrix	1	2.563E-03	0.074
	2	9.102E-3	41.28
	3	1.112E-3	628.13
Flax Fiber	1	3.091E-02	21.75
	2	2.964E-04	0.185

4.5 Comparison between Experimental, Analytical and Numerical Results

To verify the above analytical expression and the finite element analysis, stress relaxation under 0.2% deformation in fiber direction was performed on five samples. Instron Universal Testing machine was employed to measure the stress relaxation in uniaxial tension at 23°C. Test specimens were pulled to the strain level of 0.2% using a crosshead speed of 5 mm/min according to ASTM D2991. Again, as indicated before, 5 mm/min was chosen as the sharpest practical ramp to reach 0.2% constant strain to avoid the primary fluctuations in the results before stabilization for measuring the stress relaxation for flax/PU composite on the Instron machine. Figure 5 compares the temporal variation of the relaxation modulus predicted by the numerical (finite element), theoretical, and experimental methods for three different fiber volume fractions of the flax/PU composite under 0.2% strain. Considering the standard deviation of less than 10% in experimental data,

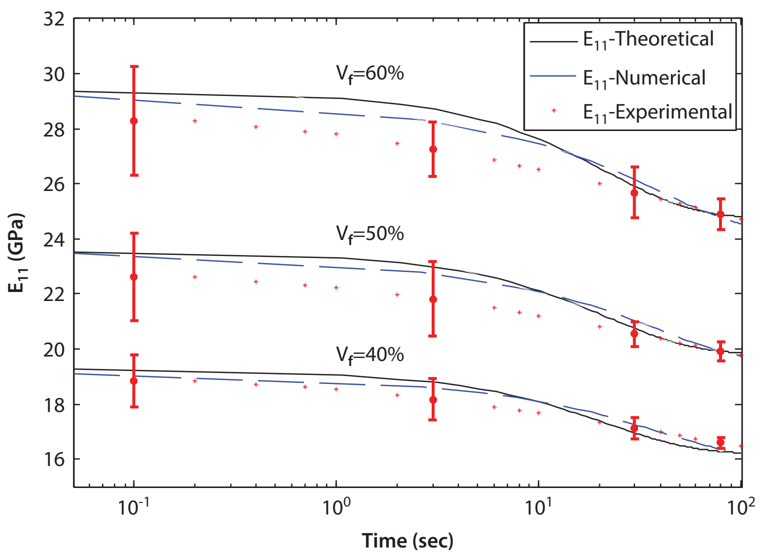

Figure 5 The drop of modulus versus time and fiber volume fraction for the flax/PU composite.

the analytical expression, as well as the numerical results, were observed to be quite accurate for the prediction of the relaxation stress in both the 40 and 50 vol% flax fiber-reinforced material, as shown in Figure 5. In analytical and numerical results, it was assumed that there were no voids or misalignment of fibers in the composite and perfect adhesion between fibers and matrix existed. Accordingly, as also shown in Figure 5, the accuracy of micromechanical models decreased as the fiber volume fraction increased.

Due to the Poisson's ratio effect, the composite material did not contract in two directions at the same amount. The transverse axial strain of composite material can be determined as:

$$\varepsilon_{22(t)} = V^f \varepsilon^f_{22(t)} + V^m \varepsilon^m_{22(t)} = -(V^f \nu^f_{(t)} + V^m \nu^m_{(t)})\varepsilon_0 \quad (11)$$

For the flax fiber, the Poisson's ratio constants leads to two terms (i=1, 2) and, for the matrix, they make three parameters (i=1, 2, 3), since the Poisson's ratio constants depend on the viscoelastic parameters' constants. Figure 6 illustrates the variation of Poisson's ratio with time at different fiber volume fractions for the flax/PU composite obtained by analytical and numerical methods. Similar to the accuracy of the analytical predictions for E_{11}, the accuracy of analytical methods for ν_{12} is also quite clear.

5 CONCLUSIONS

A micromechanical model was employed to determine the viscoelastic properties of the flax/PU composite using the properties of its constituents. A linear viscoelastic behavior was considered for both flax fiber and PU resin under small deformation. Six independent loadings were applied to a unit cell with randomly distributed cylindrical fibers in the matrix under periodic boundary condition. In this inverse characterization analysis, time-dependent stresses and strains were measured by volume-averaged method over the RUC. An analytical approach was also developed to predict the stress relaxation response of the composite material consisting of linear viscoelastic flax fiber and bio-based PU matrix. A good agreement between the micromechanical modeling data and experimental results validated the assumptions and the numerical models for the linear viscoelastic response of the bio-based composite. The measured viscoelastic parameters of the bio-based PU resin and flax fiber and the proposed micromechanical model in this study have a broad application in the housing and automotive industries. As mentioned before, in this study, perfect bonding between fiber and matrix were assumed and a cylindrical cross-sectional area for fibers was considered. Applying an intermediate phase between fiber and matrix may improve the predicted results. Future studies will focus on the implementation of an imperfect fiber-matrix bond and irregular cross-sectional shape of flax fiber in the micromechanical model for the viscoelastic natural fiber-reinforced composites.

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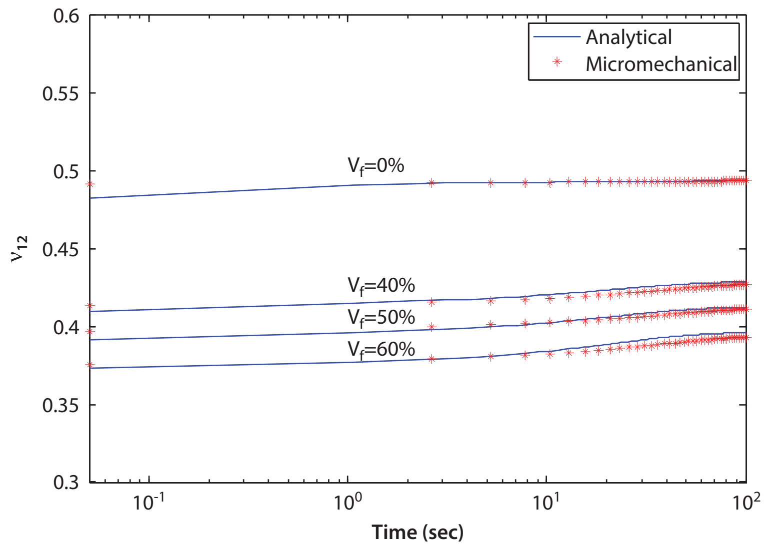

Figure 6 The variation of Poisson's ratio as a function of time and fiber volume fraction for flax/PU composite.

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