Large deformation non-linear response of fiber-reinforced elastomers: glass fibers-polydimethylsiloxane laminates

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ABSTRACT: Soft fibers-reinforced composites are promising materials for several applications as flexible actuators and in bio-mechanical devices. They are based on elastomeric polymers that allow ranges of deformation much larger than those of conventional thermosetting polymer-based composites. In order to investigate the behavior of these composites, quasi-static monotonic and cyclic tensile tests in uniaxial loading conditions have been carried out on continuous glass fibers-polydimethylsiloxane laminates with stacking sequence $[+\theta,-\theta]_{2S}$, with $\theta = 30, 45, 60$ and 75°. The stress-strain curves obtained show a linear response of the materials at small strains, a "softened" response at intermediate strains and a strain induced "hardening" at large strains. An extensive fibers reorientation was observed during loading. Large reduction in the specimens width was also measured and an increase in their thickness was observed. Loading-unloading tests showed a pronounced hysteresis and strain induced softening was also observed. These experimental observations underline the importance of a deep characterization of elastomeric matrix composites.

1 INTRODUCTION

Thanks to the growth of soft robotics and biomimetic applications, fiber-reinforced elastomers, FREs, have drawn an increasingly attention. The requirements of low density, high load bearing capacity and high flexibility demanded by most of biomimetic applications can be satisfied by FREs, thanks to the soft and deformable elastomeric matrix combined with stiff fibers. These applications are based on the mimic of complex movements of living being accompanied by wide motions. Properly designed FREs, combined with specific actuators, can provide complex response to simple stimulus, for example a simultaneous bending and torsion, by exploiting the load coupling effects generated by the anisotropy of the composite reaching, at the same time, high levels of deformation.

To properly design such materials and predict their responses, the evaluation of the mechanical properties and the behavior of the composites, typically laminates made of a sequence of laminae of unidirectional glass fibers properly oriented, is fundamental. A deep study of the behavior of this kind of composites is not present in literature. Most of the studies present in literature are focused only on the behavior of composite based products in their final application. The present work aims to characterize the mechanical behavior of home-made composites: the study of the behavior at large strains of a continuous glass fibers – polydimethylsiloxane laminate was carried out. The response of symmetric-balanced $[+\theta, -\theta]_{2S}$ laminates with $\theta = 30, 45, 60$ and 75° has been analyzed. Monotonic quasi-static tensile tests and loading-unloading tests have been performed.

2 EXPERIMENTAL PROCEDURE

2.1 *Composite production*

The FRE investigated is constituted by continuous glass fibers (GF) embedded in a polydimethylsiloxane (PDMS) matrix of Sylgard®184, provided by Dow Corning. The PDMS is provided as a two-component system of a pre-polymer and a curing agent. Fibers layers are provided with areal weights of 150 g/m^2 and 200 g/m^2.

The composite laminates fabrication adopted is essentially a manual layup procedure performed in molds. Unidirectional glass fibers layers are manually cut with desired fibers orientation. Pre-polymer and curing agent are mixed with a weight ratio 10:1 as suggested by the supplier. The mixture is mechanically stirred for 10 min and de-gassed in a vacuum chamber. Fibers layers suitably oriented are inserted in an aluminum mold, over which the mixture of uncured PDMS is poured at room temperature. It follows a further de-gassing to evacuate the entrapped air, then the mold is closed and kept at 23°C for 20 h, before placing the mold in an oven at 150°C where the polymer matrix curing already started at 23°C is completed for 1.5 h. The first step at 23°C is necessary to facilitate the impregnation of the fibers and the extraction of any air bubbles trapped in the laminate during the impregnation phase. The second phase at 150°C is needed to complete the crosslinking of the PDMS. Further details about the crosslinking kinetics of Sylgard®184 can be found in (Bardelli et al. 2021). The prepared laminates are then cut to produce the specimens.

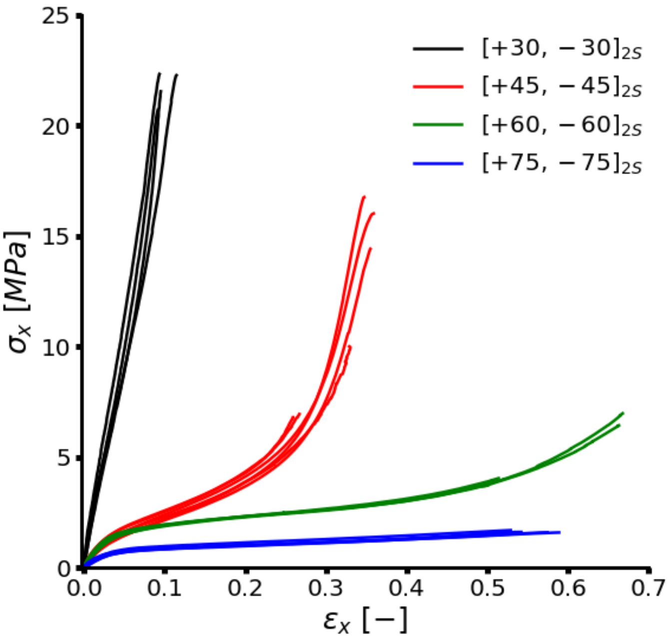

Figure 1: Engineering stress – strain curves obtained from monotonic tensile tests of the laminates.

2.2 *Mechanical testing*

Uniaxial tensile tests have been performed on an Instron 5967 dynamometer with a load cell of 2 kN at a constant nominal strain rate of $\dot{\varepsilon}$ = 0.12 min^{-1}. All the tests were video recorded with a uEye camera (UI-5490SE) equipped with a photographic lens Nikon 28-105 to perform a DIC analysis for local strains, ε_x and ε_y, measurement. A video acquisition speed of 5fps was used for all the tests. Furthermore, additional specimen's sideview video recording has been done, to investigate specimen thickness variation. In the present work, symmetric angle-ply laminates $[+\theta,-\theta]_{2S}$ with θ = 30°, 45°, 60° and 75° have been analyzed. Tests have been performed on rectangular specimens. Six $[+45,-45]_{2S}$ specimens with width w = 25 mm, thickness t = 2 mm and a gauge length of 210 mm have been tested, while four specimens for configuration $[+30,-30]_{2S}$, $[+60,-60]_{2S}$ and $[+75,-75]_{2S}$ with width w = 12.5 mm, thickness t = 2 mm and a gauge length of 50 mm have been tested. The fiber volume fraction of the specimens employed for monotonic tests was v_f = 0.3, the fiber volume fraction of specimens employed for loading-unloading tests was v_f = 0.24. No end-tabs have been used. The loading-unloading cyclic tests performed are:

- Case A and B: 10 loading-unloading cycles were first performed up to ε_x = 0.1. The specimen was removed from the clamping system and let to recovery at room temperature for 5 h. Then further 10 loading-unloading cycles were carried out up to ε_x = 0.31 for the Case A, and up to ε_x = 0.21, for the Case B.
- Case C: loading-unloading cycles with an incremental crosshead displacement of 5 mm, until reaching ε_x = 0.35.

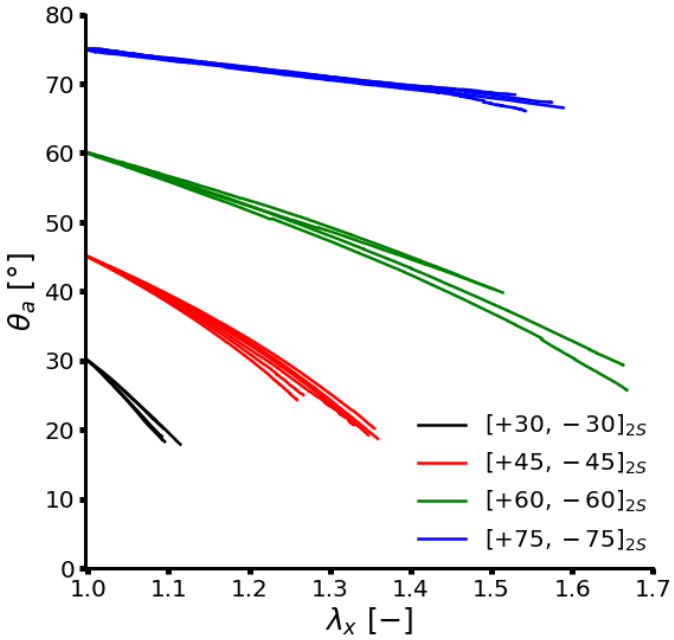

Figure 2: Variation of the actual fibers angle θ_a as a function of the draw ratio in loading direction λ_x.

3 RESULTS AND ANALYSIS

Engineering stress – strain curves obtained from monotonic tensile tests of the laminates are reported in Figure 1. The curves show a highly non-linear stress-strain behavior, which can be related to the high matrix deformability. The laminates are characterized by an initial linear behavior extended over limited strain ranges, followed by a strain induced softening, and then by a final "hardening". This behavior is showed by all the laminates produced, with different extensions of the "softened" and "hardening" zones depending on θ.

During specimens' extension, a significant width reduction was noticed for most of the specimens, accompanied by a considerable change in fibers orientation. The actual angle θ_a of the fibers in the specimen during loading has been predicted with a geometrical approach employing the measured transversal strains ε_y, based on the hypothesis of no-deformation of the rigid fibers, as:

$$\theta_a = \sin^{-1}\left((\varepsilon_y + 1)\sin\theta_0\right) \qquad (1)$$

Figure 2 shows the actual fibers angle θ_a as a function of the draw ratio applied in the loading direction λ_x. The angle varies considerably during loading and the rate at which decreases increasing λ_x is greater for laminates with smaller initial angles.

Furthermore, the behavior of the specimens in thickness direction (z-direction) has been also investigated. The hypothesis of constant specimen volume

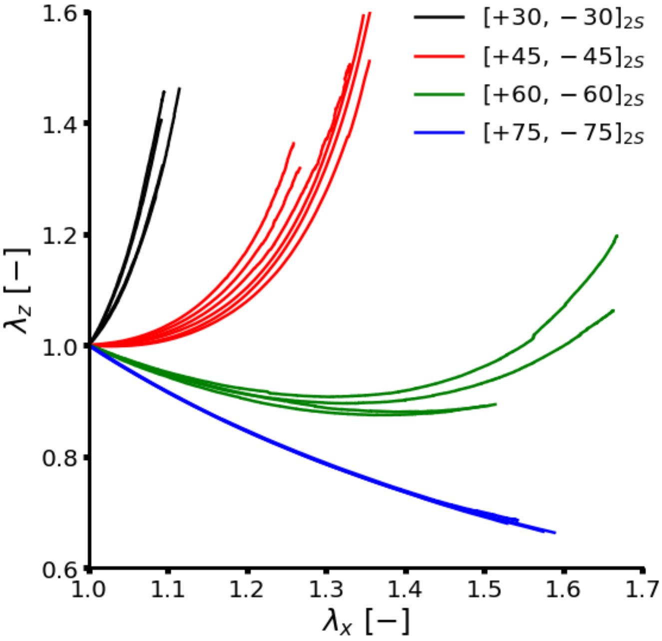

Figure 3: Draw ratio in the thickness direction λ_z as a function of the draw ratio in the load direction, λ_x.

has been introduced, justified by the incompressibility of the rubber matrix, and by the fact that the much stiffer fibers can be considered undeformable. The draw ratio in the thickness direction, λ_z, has been thus determined as:

$$\lambda_z = \frac{1}{\lambda_x \lambda_y} = \frac{1}{(\varepsilon_x + 1)(\varepsilon_y + 1)} \qquad (2)$$

Figure 3 shows λ_z as a function of λ_x for the tested specimens. A thickness increase can be observed for [+30,−30]$_{2S}$ and [+45,−45]$_{2S}$ laminates. [+75,−75]$_{2S}$ laminates show a thickness reduction, while in [+60,−60]$_{2S}$ laminates a specimen thickness reduction is first observed followed by a thickness increase after a certain strain value. These trends have been also experimentally confirmed in (Magni 2022).

It is interesting to correlate the behavior of λ_z with the variation of the fibers angle. In Figure 4 λ_z is plotted as a function of θ_a which is represented as decreasing from 80 to 0 because the decreasing of θ_a direction corresponds to an increase in λ_x. A material θ_a value of 46° resulted to be characteristic for the material: a strain induced specimen thickness increase ($\lambda_z > 1$) occurs only if θ_a is lower than 46°. A specimen thickness increase was observed for [+30,−30]$_{2S}$ and [+45,−45]$_{2S}$ laminates. For [+75,−75]$_{2S}$ laminates, for which θ_a is always larger than 46° in the whole deformation range explored, a thickness reduction ($\lambda_z <$ 1) occurs. [+60,−60]$_{2S}$ laminates undergoes first an initial thickness reduction, until a θ_a value of about 46° is achieved, then, θ_a becomes lower than 46° and a thickness increase is observed.

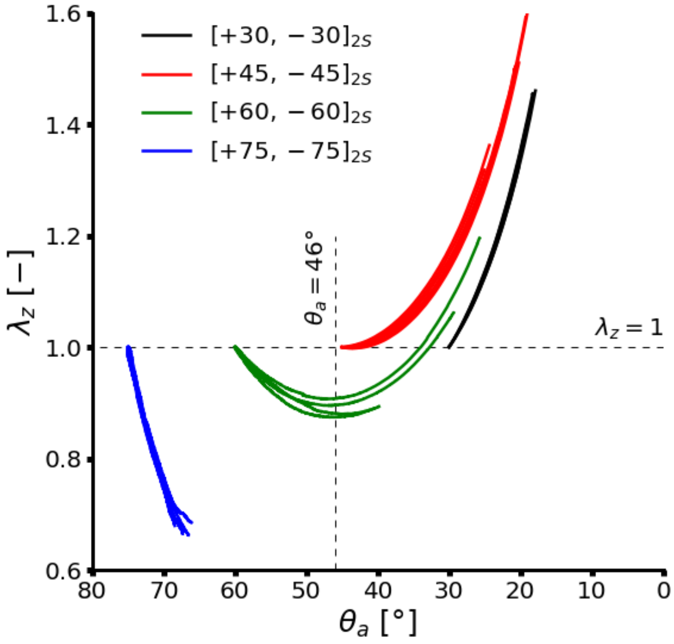

Figure 4: Draw ratio in the thickness direction λ_z as a function of the actual fibers angle θ_a which is represented as decreasing from 80 to 0 because the decreasing of θ_a direction corresponds to an increase in λ_x.

Further considerations can be done observing the tangent modulus E$_t$, measured as the slope of the strain-stress curve at any strain, plotted versus the strain dependent actual fibers angle θ_a (Fig. 5). The laminates become softer even if the fibers are reorienting toward the loading direction (lower θ_a) which should theoretically contribute to increase the stiffness of the laminates (Jones 1999), then, independently of the reached strain level, the laminates stiffness increases and the moduli of the different laminates at the same actual angle θ_a become similar.

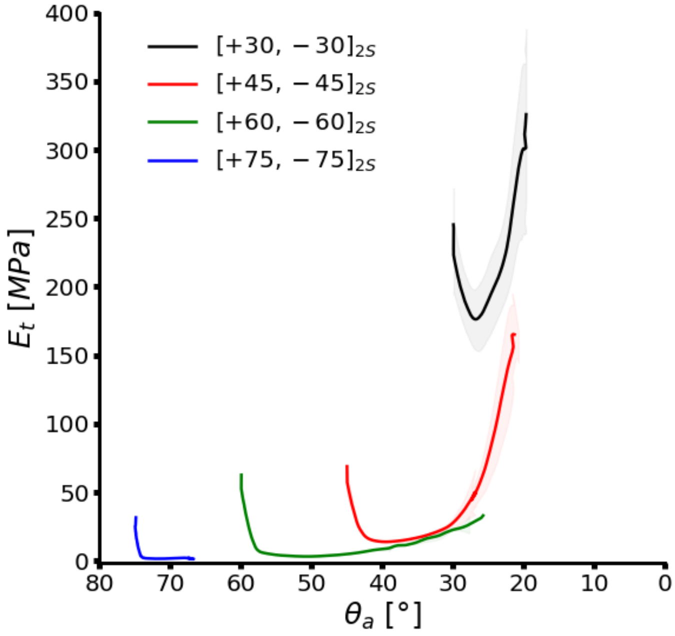

Figure 5: Tangent modulus Et versus the actual fibers angle θ_a which is represented as decreasing from 80 to 0 because the decreasing of θ_a direction corresponds to an increase in λ_x.

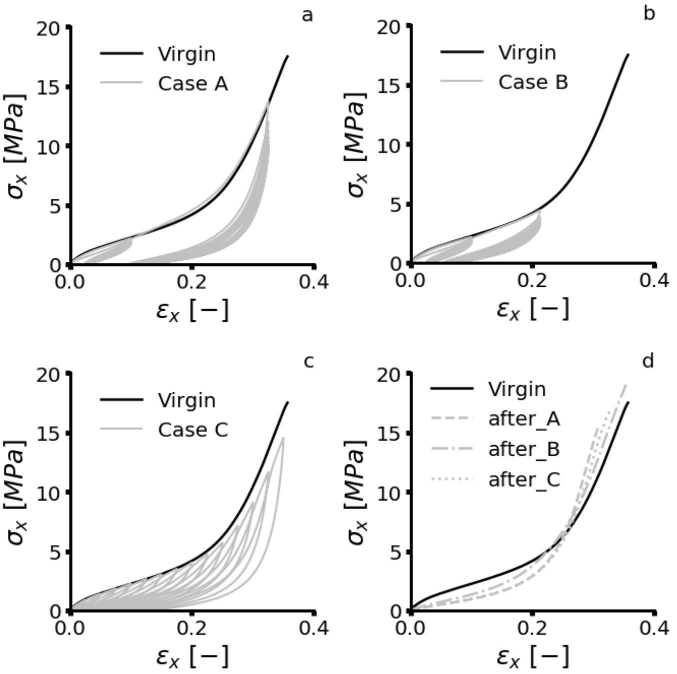

Figure 6: (a) (b) (c) Stress-strain curves from cyclic tests with different loading-unloading histories, respectively Case A, B and C. (d) Stress-strain curves for tensile tests up to failure performed on the “cycled material” specimens. In all the plots, the black line represents the “virgin material” (not-cycled) specimen.

Therefore, initially the contribution of the elastomeric matrix is predominant. The softening of the laminates can be related to a typical behavior observed in filled rubbers, that show a softening when stretched, due to the breakage of the weak bonds created at the rubber-filler interface (Diani et al. 2009) but it could be also related to a partial detachment of the fibers from the matrix. Then, at larger strains, the contribution of the fibers orientation becomes predominant.

To further investigate the laminate mechanical behavior, three cyclic loading-unloading tests with different histories have been performed on $[+45,-45]_{2S}$ specimens, as reported in Figure 6a, b and c. The specimens tested in cyclic tests have been left to recover for a few days and then tested to failure. They will be referred to as “after_X” specimens (X = A, B or C). The relevant σ_x-ε_x curves are reported in Figure 6d. In all the plots, the curves are compared to that of a virgin specimen, not subjected to any cyclic loading. From Figure 6a, b and c, it can be observed that: (i) after the first cycle, lower stresses are required to re-deform the material and (ii) there is a residual strain after each cycle. The stretch-induced softening could be related to the decrease in the material stiffness, as commonly observed in particle filled rubber compounds commonly, a phenomenon referred to as “Mullins effect” (Diani et al. 2009). Further, focusing on Figure 6d, it can be observed that the “after_A” and “after_C” tensile curves, obtained for specimens previously stretched up to the same draw ratio in different cyclic test, overlap and differ from that of “after_B”, relevant to a specimen that was subjected to a lower maximum strain in the cyclic test. These results suggest that the pre-straining loading history does not affect the material behavior, rather the behavior depends on the applied pre-strain. Moreover, this strain level affects the mechanical behavior in the low strain range: for a larger value of the pre-strain applied in the cyclic test, the material shows a lower rigidity at low strain in the following tensile test.

4 CONCLUSIONS

A continuous GF-PDMS composite has been studied. In particular, the behavior of symmetric-balanced angle-ply laminate has been investigated. They showed a highly non-linear response. The laminate responds with an initial stiff behavior followed by a softening and a further hardening at higher deformations. This behavior has been reconducted to different contributions. It has been shown that the contribution of fibers orientation to laminate stiffening is not evident in the first stages of loading. The softened region can only be explained by considering other contributions, such as the Mullins effect or partial detachment between fibers and matrix. The subsequent hardening of the response of the laminates may instead be related mainly to fibers reorientation because the response of the laminates becomes independent of the strain applied when the softening region is exceeded. Furthermore, an increase in thickness has been observed when fibers angles less than 46° were reached. The reported results shown that a proper investigation of FREs requires to consider several aspects that are normally not evaluated in common rigid matrix composite materials and should be taken into consideration for a proper design of FREs based product or for the prediction of its mechanical behavior.

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