EFFECTS OF SAMPLE SHAPE AND ADHESIVE TYPE ON RHEOLOGY OF UNIDIRECTIONAL CARBON FIBER PREPREGS

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Abstract

The viscoelastic properties of carbon fiber reinforced thermoset composites are of utmost importance during processing such materials using composite forming. The quality of the manufactured parts is largely dependent on intelligent process parameter selection based on the viscoelastic and flow properties of the polymer resin. Viscoelastic properties such as the complex viscosity (η^*), storage modulus (G'), loss modulus (G''), and loss tangent ($\tan\delta$) are used to determine the critical transition events (such as gelation) during curing. An understanding of the changes in viscoelastic properties as a function of processing temperature and degree of cure provides insight to establish a suitable processing range for compression forming of prepreg systems. However, tracking viscoelastic properties as a function of cure during the forming process is a challenging task. In this current work, we have investigated the effect of sample size and adhesive type on the rheological properties of a commercially available carbon fiber prepreg material. Specifically, determining the linear viscoelastic region (LVE) as a function of sample configuration and different adhesive chemistries were explored. The results suggest that the square-shaped sample geometries coupled with cyanoacrylate based adhesive are optimum for conducting rheological characterization on the carbon fiber prepreg system.

Introduction

Fiber reinforced polymer composites (FRPC) are composed of fibers such as fiberglass, carbon, and polyester, bonded by a thermosetting or thermoplastic polymer matrix. The fibers typically have high strength and modulus that can withstand stresses in the longitudinal direction of the load. The polymer matrix binds the material, which allows for further load/stress distribution and offers thermal, chemical, and environmental resistance that increases the overall fracture toughness and damage tolerance of the system [1]. The high strength to weight ratio of FRPC is particularly advantageous in aeronautical application as weight reduction is proportional to fuel efficiency. Another benefit in using FRPC is the flexibility for formation of more complex shapes and designs that are not traditionally available for ceramic or metal composites [2].

One of the major challenges in the FRPC forming process is the occurrence of wrinkling and shape distortion of the fabric caused by the surface differences between the forming tool and surface of the prepreg. In general, static friction is considered as friction between non-moving surfaces, and is dominated by adhesion between the contact surface asperities, the time and mode of contact, whereas the kinetic friction is described by the friction between two moving surfaces and is dominated by the viscoelastic properties of the materials. The slipping behavior is not only dependent on processing parameters and test methodologies, but also on the material's bulk viscoelastic behavior which is highly dependent on the forming temperature and the degree of cure. In particular, the understanding of changes in viscosity profile at various temperatures and time can be used to establish the range of processing parameters in which deformation is minimized.

Prior to performing any rheological tests, it is important to determine the linear viscoelastic region (LVE) of the sample to evaluate the relationship between molecular structure and viscoelastic properties. At constant G' (within the LVE region), the sample structure remains unchanged. Strain rates above the LVE region indicates structure instability (i.e., structure break down). The LVE region is the region of strain where G' remains constant, and below the critical strain at the onset of non-linear G' behavior. The objectives of the experiments conducted in this paper are to determine the linear viscoelastic region (LVE) of the prepreg, examine the effects of various set up such as prepreg shape and adhesive types on subsequent rheological behavior of the sample, and to establish a base experimental procedure for further testing.

Materials and Experimental Methods

The material studied in this work was HexPly® unidirectional (UD) prepreg (M77/38%/UD300/SGL-50K), supplied by Hexcel, Duxford, UK. The material is a 50K (each tow is a bundle of 50,000 continuous individual carbon filaments) high strength carbon fiber tow, with a fiber areal density of 300 grams per square meter, pre-impregnated with M77 epoxy resin (38% resin content) [3].

In this study, a commercial torsional rheometer (Anton Parr MCR 302 fitted with a CTD 450 convection temperature chamber) was used to measure the viscoelastic properties of the prepreg samples.

The prepreg fabric was thawed to room temperature at approximately 22°C before cutting into 28x28mm square samples with scissors, or into ~26mm circular samples with a stamping die and press (e.g., circular cutter, punch part XS819017, Cascav tools).

Samples were loaded onto the bottom plate of the rheometer with fiber orientation parallel to the machine, without adhesives, or were fixed to the bottom geometry with double sided pressure sensitive adhesive (PSA) (3M®), or cyanoacrylate (MXBON®). Sample adhesives (cyanoacrylate) were used for the LVE measurements. A 1mm ring geometry is used, and the top and bottom geometry were cleaned after each experiment with acetone to remove residual epoxy transferred between tests. The ultimate objective was to monitor the friction coefficients between the prepreg samples and the rheometer top plate. Therefore, serrated plate geometries were not employed as they can affect and/or damage the surface of the prepregs during sliding under sufficient normal force. The annular plate geometries can greatly alleviate this issue and hence were used for the experiments described in this paper.

Rheological measurements are conducted in dynamic oscillatory mode (strain sweep) with strain from 0.1% to 100% at a frequency of 1 Hz, at ambient temperature to determine the linear viscoelastic region (LVE) of the sample [4].

Results and Discussion

Prepreg samples are often cut into specific shapes with precise dimensions (e.g., square or circular) to minimize variabilities between rheological measurements [5–7]. The effects of sample shape on the rheological response of samples are examined by conducting oscillatory strain sweep experiments at ambient temperature, with a strain (%) from 0.01-100%, a normal force of 20N and 30N (for the square and circular sample, respectively), and a frequency of 1 Hz. The plots of storage modulus (G') and loss modulus (G") vs. shear strain [%] of the square and circular sample are shown in Figure 1.

The overlapping values of the samples suggest the experiments are both repeatable regardless of sample shape. The linear viscoelastic region (LVE) for the square sample is observed at below 0.07% strain. A lower LVE range, with a maximum of 0.05% strain, is observed for the circular sample. The result suggests that circular samples would be more sensitive to frequency change due to the lower LVE range. However, it should be noted that the lower LVE range of the circular sample could be due to the larger normal force applied during the experiments.

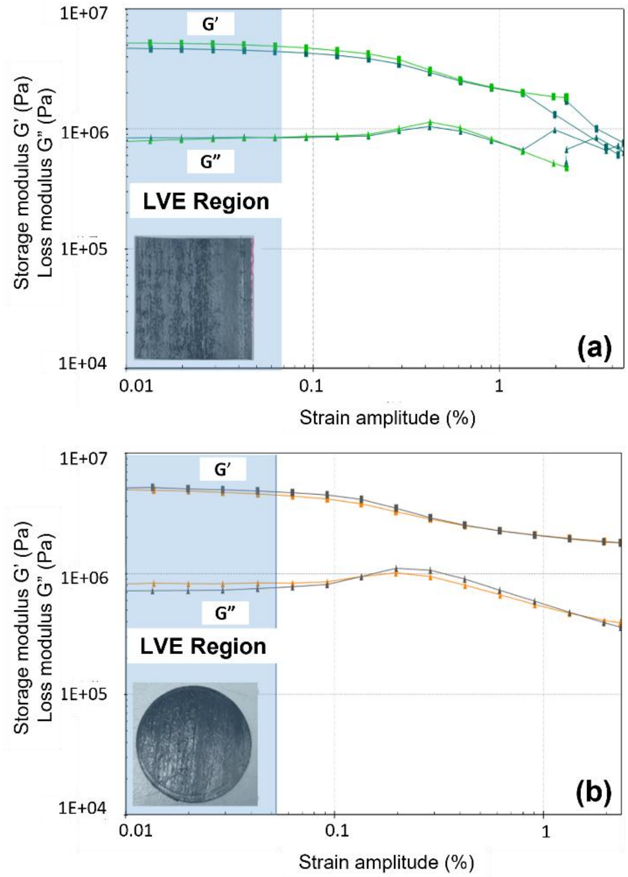

Figure 1. Storage and loss modulus vs. shear strain (%) of (a) square and (b) circular samples measure with 1mm ring geometry under small amplitude oscillatory shear (SAOS) mode at ambient temperature, 0.01-100% strain with a normal force of 20 and 30 N, respectively.

A more important parameter to consider is the drastic change in G' and G'' of the square samples at above 1% strain, suggesting significant structural instability as strain increases. This behavior could be caused by the presence of excess fabric of the square sample (e.g., "edge effect"). A comparison of the square and circular sample loaded in the test fixture is shown in Figure 2. While this large change in G' and G" could be of concern, the fact that the behavior occurs at a region above the LVE suggests that this might not be as much of an issue as long as subsequent experiments are conducted below the LVE limit.

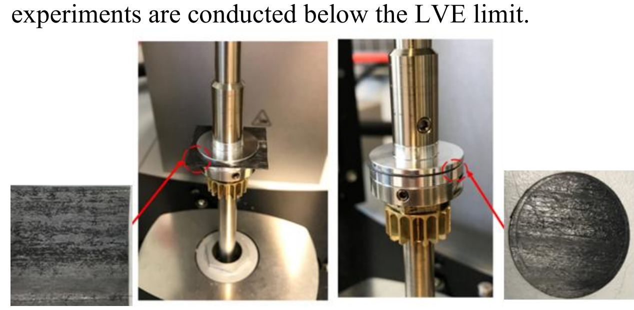

Figure 2. Prepreg in fixture (left) square sample with excess fabric, and (right) circular sample.

While the circular sample seems to be a more "stable" choice for our tests, split propagation in the fiber direction is often observed during the cutting process with the circular punch (Figure 3). Since sample splitting was not observed during the cutting process of square samples, and the rheological results suggests relatively good repeatability and stability below the LVE region, subsequent tests were conducted similar to the reported experimental conditions below 0.07% strain. It should be noted that the square sample dimensions are less consistent since samples are measured and cut with scissors by hand, compared to a circular punch/die.

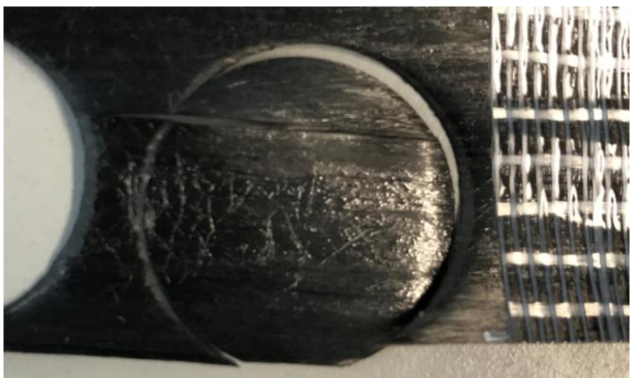

Figure 3. Split circular sample from cutting process.

In addition to controlling sample shape and size, specimens should be held firmly in place during experiments to minimize fabric twist/overlap. Rheological tests with prepregs have been conducted with multiple layers of prepreg, using double sided tape or cyanoacrylate as an adhesive [5,7]. For our experiments, we want to ensure slippage occurs only at the top of the plate between the geometry and prepreg to mimic tool-ply friction. A schematic of the adhesive test set up is show in Figure 4.

A set of steady shear tests were conducted with an angular deflection from 0-100mrad, at ambient conditions, with a normal force of 5N, and 1mm ring geometry to examine the effects of sample adhesive on measured rheological properties. The result of the experiments are shown in Figure 5. The sample without adhesive reached the rheometer torque limit of 0.2 N·m, where as the PSA exhibited a continuous sliding behavior, and the sample with the cyanoacrylate adhesive showed a plateau and constant torque over the deflection range.

Variations in the experimental results can be explained by the different slip location/deformation mechanism between the surfaces as shown in Figure 6. Since the uncured prepreg is somewhat soft/flexible at room temperature, the rotating motion of the top geometry could cause the fabric to move or even twist at either the top or bottom plate, resulting in the large increase in torque at low deflection angles.

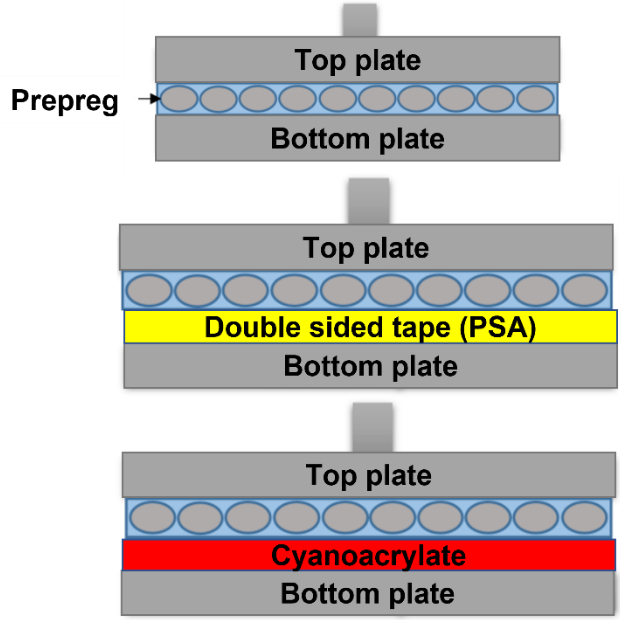

Figure 4. Schematic of sample set up for adhesive tests.

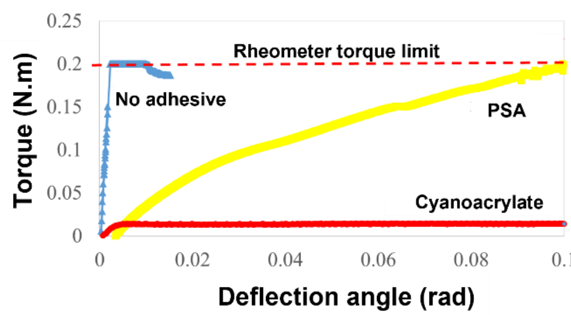

Figure 5. Torque vs. deflection result of samples with/without sample adhesives.

Although the presence of PSA improves the prepreg-plate contact at the bottom geometry, the continuous sliding behavior observed over the deflection interval suggests that there is still some deformation/movement from both the prepreg and PSA. In addition, since PSA is a viscoelastic material, changes in sample viscoelastic properties could occur at different experimental temperatures. Since cyanoacrylate is a thermosetting polymer, the strong adhesive force between the prepreg and cyanoacrylate would prevent slip at the bottom geometry and reduce movement/deformation of the prepreg.

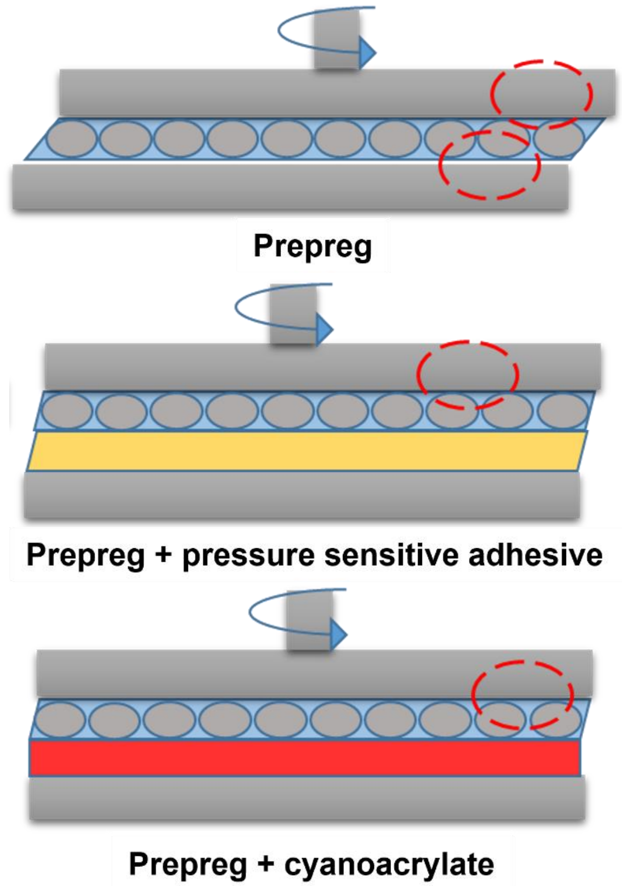

Figure 6. Proposed slip location (red circles) and deformation mechanisms between sample and geometry during steady shear.

Conclusions

The effects of sample shape and adhesive type on the rheological response of samples were examined by a parallel plate rheometer (modified 1mm ring geometry) at ambient temperatures. Oscillatory strain sweep results suggest relatively good repeatability and stability below the LVE region for both square and circular samples. Subsequent tests were conducted with square samples due to challenges with cutting circular samples at experimental conditions below 0.07% strain. The excess fabric could cause an effective increase in measured results due the "edge effect" (e.g., more resistance to motion from the excess fabric). Steady shear results suggest that prepreg movement/distortions between the geometry can be reduced in the presence of an adhesive. A continuous movement of PSA-prepreg system is observed in steady shear experiments due to the viscoelastic property of PSA, whereas minimal movement of the prepreg is observed in the cyanoacrylate-prepreg system due to the strong adhesion force formed between the prepreg cured cyanoacrylate bottom geometry.

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