Flexural Creep Behavior of Unidirectional and Cross-Ply All-Poly(propylene) (PURE™) Composites

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The long-term viscoelastic behavior of reinforced all-poly(propylene) composites was studied by flexural creep tests. Both unidirectional and cross-ply laminates were prepared from PURE™ coextruded tapes by vacuum bag molding in an autoclave. The specimens were subjected to isothermal creep tests at different temperatures ranging from 20 to 80 °C under an applied load. The time-temperature superposition principle was verified for the creep data. An Arrhenius type relationship was found to better describe the shift data obtained from the creep tests. The activation energies relating to the different reinforcement architecture and different relaxation process were calculated.

Introduction

Poly(propylene) (PP) is a thermoplastic with a wide range of applications. The main reason for this is the possibility of tailoring its structure and properties for a particular application. The use of fibers as reinforcement in PP matrices has received much academic and commercial interest in the past few decades. Although excellent mechanical properties have been achieved in this way, life cycle assessment does not yield favorable results for PP composites when they are “traditionally” reinforced with glass, carbon, aramid fibers or fabrics. This is mostly due to the energy-intensive production of the above reinforcing fibers and limited recyclability of the corresponding composites (e.g., due to fiber attrition during melt compounding). This led to a search for alternative recycling-friendly novel homocomposites. Homocomposites represent an effective alternative to the traditional fiber reinforced composites where the matrix and the reinforcement are from the same polymer, thereby supporting the ease of recyclability.

The topic of self-reinforced composites has gained interest since Porter et al., in the mid 70s, showed the production of “single polymer composite” from polyethylene fibers and polyethylene matrix.[1] Later on, the group of Ward[2–6] succeeded in converting a part of such fibers into a matrix in which the residual fibers were embedded.
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via “hot compaction”. This material is nowadays commercially available as Curve™. Of course, hot compaction requires precisely set processing parameters with unusually low tolerances. The narrow processing window (due to the small difference in the melting temperature of fibers and matrices of the same polymer) eventually lead to another option which is even simpler and thus already practiced. In this case, the reinforcing fiber or tape could be coated or embedded in a matrix (usually a random copolymer) of lower melting point. Random copolymer structure is an essential prerequisite as this yields the required negative shift in the melting temperature. This concept has been explored by researchers in the last years.[7–11] The commercial breakthrough of this concept has been recently developed in the authors’ laboratory.[13–14] It consists of a lay-up of random PP copolymer/PP homopolymer/random PP copolymer.[12] The advantage of this process is the enlarged processing window (20–40 °C). Another key method for producing all-PP composites has been recently developed in the authors’ laboratory.[13–14] It takes into account the polymorphism of poly(propylene) while manufacturing PP homocomposites.[15–16] Research using this new technique is still in progress. Due to the scientific and commercial interest in the recycling friendly all-PP composites, serious efforts have been made in the past to obtain a novel all-PP composite with better processability and improved mechanical properties. Most of the published studies conducted so far have tried to examine the mechanical properties of all-PP composites as a function of the tape draw ratio, consolidation parameters, reinforcing fiber volume or woven geometries.[4,17–19] However, in spite of the recent progress in processing methods and improved mechanical performance of all-PP composites, limited results are available on the long-term deformation behavior, like creep.[20–23]

Creep is a slow, progressive deformation of a material under constant stress. When a plastic material is subjected to a constant load, it deforms continuously with time. This time dependent behavior of materials, often referred to as viscoelasticity, is an important characteristic of fiber-reinforced polymers, which allows better understanding of their structure-property relationships. The knowledge of the long-term durability of the composite structures is also essential to estimate their life-time under load. Although creep in an anisotropic and multi-phase system, like fiber-reinforced polymer composite, is more complex than in an unreinforced polymer composite, a creep analysis is essential for the long-term applications of all-PP composites under stress. Research works in this direction have so far showed that the creep behavior of PP fiber reinforced PP composites depends strongly on stress, temperature, void content, fiber loading and fiber dimensions.[20–23] One of the greatest constraints in studying the creep behavior is the relatively long time required for performing a test. Hence, methods that are able to predict long-term data have gained considerable attention. A number of observations suggest that the effects of time and temperature are equivalent.[24] It has been frequently observed that the mechanical responses observed after a short time resemble that observed at lower temperatures; those observed after a longer time resemble those observed at higher temperatures. The related time-temperature superposition (TTS) is one of the most widely used extrapolation technique that has been applied to virtually every mechanical property and each kind of plastic.[24] Long-term viscoelastic behavior can be predicted from the short-term test data if the time-temperature superposition principle is applicable to the studied material. According to the time-temperature superposition principle, creep compliance, D(t,T), at one temperature (T1) can be related to that at another temperature (T2) by application of a multiplicative factor (aT) to the time scale (t) namely,

\[
D(t, T_1) = D(t/aT, T_2)
\]

It is fairly believed that this superposition manifests itself from the molecular behavior and, therefore, equations based on the activation energy (E), such as the Arrhenius equation, have been proposed. The Arrhenius equation relates the horizontal shift factor (along the logarithmic time axis) with temperature:

\[
\ln a_T = \frac{E}{R} \left[ \frac{1}{T} - \frac{1}{T_0} \right]
\]

where \(a_T\) is the horizontal shift factor, \(R\) is the universal gas constant, \(T_0\) is the reference temperature and \(T\) is the experimental temperature. The temperature dependence of the secondary relaxations can generally be described by this equation.

Another commonly used empirical equation for TTS that relates a shift in temperature with a shift in time is the William-Landel-Ferry (WLF) equation:

\[
\ln a_T = - \frac{C_1(T - T_0)}{C_2 + T - T_0}
\]

Here \(C_1\) and \(C_2\) are constants and \(T\) and \(T_0\) are the test and reference temperatures, respectively. The WLF equation was found empirically to demonstrate the frequency dependence of glass transition temperature in amorphous substances.[25,26] According to this equation, the relaxation times describing the \(\alpha\) relaxation (glass transition) in amorphous polymers can be very well followed.
The objective of this paper is to present the results of an experimental study that investigates the effect of reinforcement architecture and temperature on the viscoelastic behavior of all-PP (PURE®) composites produced by vacuum bag molding in an autoclave. According to our recent literature search, the creep behavior of the all-PP composites made from these coextruded tapes has not been investigated so far. Also in this work, the applicability of TTS principle to short-term creep data was verified in order to predict the long-term creep response of such composite laminates.

**Experimental Part**

**Material**

In this study commercially available all-PP (PURE®) coextruded tapes, obtained from Lankhorst-Indutech B. V. (Sneek, The Netherlands), were used to make unidirectional (UD) and cross-ply (CP) laminates. The PP tapes have a skin-core-skin (A-B-A) morphology with PP homopolymer as the core and a random PP copolymer as the skin layer. The other characteristics of these coextruded tapes are summarized in Table 1.

**Manufacture of Laminates**

The manufacture of all-PP composites involved a two-stage process - winding of the PP tapes, [both unidirectional (0°) and cross-ply (0/90°)], and consolidation of the fabric plies at a suitable temperature and pressure in an autoclave.[27] For winding the tapes, a filament winding technique was adopted. A photograph of this winding process is shown in Figure 1. Using a typical winding machine, supplied by Bolenz & Schaefer Maschinenfabrik (Biedenkopf, Germany), PP tapes were wound from a bobbin onto a uniform steel plate rotating at a constant speed of 40 rpm. In each case, the total number of layers was kept the same (30 layers).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Width [mm]</td>
<td>2.28</td>
</tr>
<tr>
<td>Thickness [µm]</td>
<td>50</td>
</tr>
<tr>
<td>Density [g cm⁻³]</td>
<td>0.829</td>
</tr>
<tr>
<td>Composition</td>
<td>copolymer/homopolymer/copolymer</td>
</tr>
<tr>
<td>Heat of fusion [J g⁻¹]</td>
<td>105.03 ± 0.4</td>
</tr>
<tr>
<td>Tensile modulus (ISO 527) [GPa]</td>
<td>15 ± 2</td>
</tr>
<tr>
<td>Shrinkage at 145 °C for 30 min [%]</td>
<td>5.5 ± 0.5</td>
</tr>
<tr>
<td>Tensile creep compliance at 25 °C after 30 min at 10 MPa [GPa⁻¹]</td>
<td>0.311 ± 0.02</td>
</tr>
</tbody>
</table>

For producing CP laminates, the winding direction on the steel plate was sequentially changed. After laying two UD layers, another two layers of tapes were positioned perpendicular to them. A similar process for manufacturing unidirectional all-PP composites from coextruded tapes has already been reported by Alcock et al.[17] To consolidate the all-PP composites, the steel plate with the wound tapes was placed in between other two steel plates inside a vacuum bag connected to a vacuum pump that applies a negative pressure inside the bag. The entire bag was then placed inside an autoclave (obtained from Scholz Machinenbau GmbH, Coesfeld, Germany) and slowly heated from room temperature (23 ± 1 °C) to the consolidation temperature (Tc) of 145 °C. A thermocouple was inserted to monitor the temperature rise in the specimen during heating. Simultaneously, the vacuum pump was started to steadily increase the pressure inside the bag to reach the consolidation pressure (P) of 24 bar. The time taken by the specimen to reach the consolidation temperature was considerable longer (t1 - t4 ≈ 90 min) than the air temperature inside the autoclave (t2 - t3). The consolidation time, (t1 - t4) was kept constant at 30 min. Note that consolidation begins earlier under the consolidation pressure as the copolymer matrix starts melting (~138 °C) before the consolidation temperature is reached. Nevertheless, the consolidation time as defined here considers only that time during which the material is exposed to Tc. After consolidation, the laminates were cooled to the release temperature (Tc) of 30 °C. The pressure was released when the air temperature inside the autoclave reached 30 °C. Although the material still had a higher temperature inside, but at this temperature it was sufficiently consolidated that there was no loss of orientation on releasing the applied pressure. Cooling of the laminate (t5 - t6) took approximately 60 min. Identical consolidation parameters were used for the production of both unidirectional and cross-ply laminates. A schematic of the time-temperature and time-pressure profile during consolidation of all-PP composites is shown in Figure 2 and a summary of the consolidation parameters used is provided in Table 2. A vacuum bag molding in an autoclave was preferred due to the similar reasons as already mentioned by Alcock et al.[17]

**Flexural Creep**

Short-time flexural creep tests were performed using three-point bending mode at different temperatures, ranging from 20 to 80 °C, in a DMA Q800 apparatus (TA Instruments, New Castle, USA). In
this temperature range, isothermal creep tests were run on the specimens with a stepwise temperature increment of 10 °C. Prior to the creep measurement, each specimen was equilibrated for 5 min at each temperature and then the flexural creep behavior was tested for 30 min. All the tests with all-PP laminates were performed under a constant load of 10 MPa, which falls within the linear viscoelastic strain range determined by checking the proportionality condition\([28,29]\). Test specimens of dimensions $60 \times 15 \times 1.8\; \text{mm}^3$ (length $\times$ width $\times$ thickness) were used for creep tests. For the coextruded tapes, a creep test was also performed but in the tension mode at a temperature of 25 °C for 30 min. The tensile stress applied was also 10 MPa. For creep studies, the average of three statistically relevant creep data has been reported for the all-PP tapes and each kind of laminate.

### Static Flexural Test

The static flexural properties of the all-PP composites, such as modulus of elasticity, ultimate flexural strength and strain at
deviation were determined following the DIN EN ISO 178 standard on a Zwick 1445 test machine. Specimens of the same dimensions used for the flexural creep test were employed for the flexural creep measurement. A support span of 32.8 mm was used in the three point bending setup. A cross-head speed of 1 mm min⁻¹ was applied during the test and the elastic modulus was calculated in the strain range of 0.05–0.25%. Load was applied using a U2A type 10 kN load cell. A preload of 5 N was applied at the beginning of each test and the mean value of five specimens tested was reported for each type of laminates. For UD all-PP composite, flexural tests were conducted on specimens parallel to the tape axis.

### Differential Scanning Calorimetry (DSC)

The crystallinity content of the coextruded tapes and the laminates were characterized from the enthalpy associated during melting using a Mettler-Toledo DSC 821 instrument (Greifensee, Switzerland). Heating scans were performed from 25 to 200 °C at a heating rate of 10 °C·min⁻¹ with nitrogen blanketing inside the sample chamber. For thermal analysis, only the value of fusion enthalpy associated with the first heating run was considered to determine the effect of processing on the moldings. An average of three specimens was taken for each of the laminates. A higher enthalpy of fusion corresponds to the presence of more crystalline domains inside the specimen.

### Density Measurement

Density measurements were done at room temperature ($23 \pm 1\; ^\circ\text{C}$) in a highly sensitive balance (DV 214C) for measuring density provided by the Ohaus company (Naenikon, Switzerland). The liquid used for measuring the density of the sample was heptane. The average value from five specimens was taken for each measurement. Generally, a lower void content is expected to show up with a higher density.

### Optical Microscopy

Microscopic images of the cross-section of the all-PP laminates were captured by a stereomicroscope (Leitz, Germany) equipped with a high resolution digital camera. The IMAGE-C analysis software was used to estimate the void content from the micrographs.
Results and Discussion

Composite Morphology

A summary of the results relating to the composite morphology is provided in Table 3. It shows that the density of the UD laminates is slightly lower, or comparable to that of the coextruded tapes, whereas the CP laminates have a density significantly lower than that of the tapes. This is contrary to that reported by Alcock et al.\[17\] who observed a higher density for their unidirectional laminates compared to the PP tapes. But, similar to the observations of Alcock et al.,\[17\] the all-PP tapes showed lower density than that of isotropic PP (0.91 g cm\(^{-3}\)) due to the presence of microvoids as described by the cited authors. Our results imply that the consolidation of the unidirectional laminates is fairly good and better than the CP laminates with less intra- or intertape voiding. This is confirmed from the optical micrographs (Figure 3), which show characteristic cross-sections of UD and CP all-PP laminates. It can be seen that the UD laminate has been better consolidated than the CP revealed by less inter-layer microvoids. Using the IMAGE-C analysis software, the void content in UD and CP laminates were found to be 4.5 and 8.5\%, respectively. Houshyar et al. also reported void contents ranging from 1.5–16.5 for their poly(propylene)/poly(propylene-co-ethylene) composites depending on the woven geometries.\[23\] It is widely known that the presence of voids can affect the quality of the molded part significantly. According to the free volume theory proposed by Struik, the molecular mobility depends primarily on the available free volume present in the system. A higher concentration of voids as well as lower crystallinity is therefore expected to show up with an increasing tendency to creep.\[30\]

Table 3 also presents the results of DSC analysis, which indicates that the UD laminates have a higher crystallinity than the corresponding CP laminates. A higher enthalpy of fusion generally corresponds to higher crystallinity in the molding. Assuming 100\% crystallinity of PP as 207.1 J g\(^{-1}\),\[31\] the crystallinity of the UD and CP laminates were found to be 51 and 49.5\%, respectively. It is noteworthy that the crystallization behavior in reinforced polymers is quite complicated as the reinforcement may induce transcrysallization of the matrix acting as heterogeneous nucleant. However, due to the opacity of PP tapes, a detailed investigation in this area using optical microscopy was not possible and is beyond the scope of the present study. Nevertheless the results in the following sections indicate that the reinforcement architecture can significantly influence the composite morphology and subsequently the viscoelastic properties. It is noted here that although both the laminates consists of same volume of highly oriented PP tapes, the UD laminates are highly anisotropic when compared to the CP ones.

Creep Behavior

The flexural creep behavior of both unidirectional and cross-ply all-PP composites as a function of time at different temperatures is shown Figure 4. It is clear from the results that: (i) higher deformation occurs due to the increase in the polymer chain mobility at higher temperatures, and (ii) the all-PP composites offer more resistance to creep when the PP tapes are reinforced unidirectionally. This difference in the creep behavior can be attributed to the composite morphology, whereby a

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**Table 3. Summary of the composite morphology.**

<table>
<thead>
<tr>
<th>Composite</th>
<th>Reinforcement architecture</th>
<th>Density g cm(^{-3})</th>
<th>Void content %</th>
<th>Heat of fusion J g(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>UD</td>
<td>0\°</td>
<td>0.824 ± 0.004</td>
<td>4.5 ± 0.5</td>
<td>105.57 ± 0.71</td>
</tr>
<tr>
<td>CP</td>
<td>0/90\°</td>
<td>0.803 ± 0.007</td>
<td>8.5 ± 0.65</td>
<td>102.46 ± 0.39</td>
</tr>
</tbody>
</table>
higher volume fraction of the oriented tapes in the longitudinal direction providing flexural stiffness and lower void content, as well as the slightly higher crystallinity exhibited by the unidirectional composites after consolidation, increases the creep resistance and hence causes a decrease in the creep compliance. Figure 4 also indicates that the creep rate increases with an increase in temperature, as expected due to the reasons mentioned above, and the shapes of the various creep curves appear to be amenable to the well-known reduction scheme of time-temperature superposition. Coincidence of the data is good and the master curves (Figure 5) show intense creep resistance for the UD laminates, as expected. This difference in creep behavior between the CP and UD laminates can be explained by the resulting composite morphology, as discussed in the section before, and also indicates that the reinforcement architecture can significantly affect the long-term viscoelastic behavior of the all-PP composites.

Shift factor, $a_T$ was obtained directly from the experimental creep curves plotted against time by measuring the amount of shift along the time scale necessary to superimpose the curves on the reference. The reference temperature was taken as 30°C for all the creep curves irrespective of the laminate type. The log $a_T$ values thus obtained by shifting at different temperatures are listed in Table 4. It shows that log $a_T$ decreases with increasing temperature. The negative sign indicates a shift to the right of the creep curves, that is, towards the reference at lower temperature. A higher shift value thus indicates a larger shift of the creep curve to the reference curve.

One of the criteria for the applicability of time-temperature superposition is that the temperature dependence of $a_T$ must have a reasonable form consistent with experience. Generally, $a_T$ is a function of temperature alone if there is no physical aging. Thus, it is important to examine the form of $a_T$ as a function of temperature for the material system used in this study. First, the classical WLF equation was tried to describe the relationship between $a_T$ and temperature. It was found that the WLF equation is not suitable to describe the temperature dependence of all-PP composites completely. Rearranging Equation (3) gives

$$\frac{(T - T_0)}{\log a_T} = \frac{C_2}{C_1} + \frac{(T - T_0)}{C_1}$$

If the creep data follow the WLF equation, then a plot of $-(T - T_0)/\log a_T$ against $(T - T_0)$ is expected to produce a straight line. However, Figure 6 clearly shows that a

Table 4. Shift factor (log $a_T$) for UD and CP laminates.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>UD</th>
<th>CP</th>
</tr>
</thead>
<tbody>
<tr>
<td>°C</td>
<td>log $a_T$</td>
<td>log $a_T$</td>
</tr>
<tr>
<td>20</td>
<td>1.16</td>
<td>1.067</td>
</tr>
<tr>
<td>30</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>40</td>
<td>-1.215</td>
<td>-1.148</td>
</tr>
<tr>
<td>50</td>
<td>-2.304</td>
<td>-1.951</td>
</tr>
<tr>
<td>60</td>
<td>-3.151</td>
<td>-2.512</td>
</tr>
<tr>
<td>70</td>
<td>-3.786</td>
<td>-2.998</td>
</tr>
<tr>
<td>80</td>
<td>-4.426</td>
<td>-3.501</td>
</tr>
</tbody>
</table>
A straight line relationship is possible only above 40°C. Dutta and Edward\cite{32} have also showed that the WLF equation was not suitable for isotactic poly(propylene) (iPP). They explained that this arises from the semicrystalline nature of iPP, whose free volume can be thought to be quantized. Therefore, it is expected to follow an Arrhenius equation for the major transitions.

When the shift factors were plotted against inverse of temperature in Figure 7 (at the same reference temperature), it appears that the shift data can be better described in terms of the Arrhenius apparent activation energy ($E_a$) defined by Equation 2. The linear regression curves are also shown in the plot to calculate the activation energy of the deformation process. The apparent activation energies calculated from the slope of the regression curve were found to be 188 and 148 kJ·mol$^{-1}$ for UD and CP laminates, respectively. These values are comparable to that of Attalla et al. (163–221 kJ·mol$^{-1}$, depending on the experimental conditions),\cite{33} Wada (167 kJ·mol$^{-1}$)\cite{34} and Dutta and Edward (200 kJ·mol$^{-1}$)\cite{32} Our results are also in close agreement to that of Alcock et al.\cite{35} who reported that activation energies of $\approx$300 and $\approx$100 kJ·mol$^{-1}$ for their woven all-PP composites were necessary to shift the dynamic flexural modulus and tensile strength master curves, respectively. The larger the activation energy, the greater the influence of temperature on $\alpha_T$.\cite{17} This is evident from the shift factor rate calculated for both the laminates. TTS shift factor rate ($\delta T$) is defined as:

$$\delta T = -\frac{d \log \alpha_T}{dT}$$

(5)

and was determined from the slope of the plot of $\log \alpha_T$ against temperature. $\delta T$ was found to be higher for the UD (0.093) than for the CP (0.073) laminates, as expected from their activation energies. This implies that the unidirectional laminates have a broader retardation or relaxation spectrum than the CP laminates. This is supported by the significant difference in the $\log \alpha_T$ values at temperatures above 40°C.

Keeping this in mind, a closer look at Figure 7 reveals that a two-component fit can better describe the dependence of shift factor on temperature. It can be seen that above 40°C in the case of CP laminate (and 50°C for UD), the slope of the regression curve changes. It is known that PP can undergo a crystalline phase related relaxation due to inter- or intracrystalline motion resulting in a broad relaxation ranging from 40 to more than 100°C.\cite{36} This is mainly due to the fact that isotactic PP is a thermo-rheologically complex material with complicated supramolecular structure and morphology.\cite{37} Thus it can undergo two or three types of transitions depending on the material and the methodology.\cite{16,38} Hence, it is suggested that a two-component relaxation model can better describe the retardation (or relaxation) behavior of all-PP laminates, similar to that reported by Dutta and Edward.\cite{32} Using a two-component fit, the activation energies are calculated for the two types of relaxation processes and presented in Table 5. These two activation energies correspond to the $\alpha$ and $\beta$ relaxation processes, the activation energy for the latter being higher than the former.

In addition, creep and recovery compliance as a function of time ($t_{\text{creep}} = 30$ min and $t_{\text{recovery}} = 30$ min) was also studied at a temperature of 25°C. The course of recovered compliance as a function of time (plot not shown here) indicated that the recoverable compliances of the UD laminates (0.12 GPa$^{-1}$) are lower over the whole relaxation time range than those of the CP laminates (0.29 GPa$^{-1}$). This suggests that a change in the reinforcement architecture from 0/90° (CP) to 0° (UD) significantly improves
the elastic recovery and decreases the recovered compliance of the composites.

### Static Flexural Test

Parallel to the creep tests, short-term static flexural tests were also conducted. Figure 8 shows a plot of the experimental results obtained from the flexural test of the all-PP composites. The results show the average values of elastic modulus, maximum flexural strength and strain at yield of UD and CP laminates. It shows that strength and stiffness can be improved significantly (up to 4 times) by UD lay-ups, whereas elongation at yield decreases as expected from the creep studies. The stiffness of the all-PP woven composites is to a great extent governed by the stiffness of the tapes in the longitudinal direction. In the CP laminates, the volume fraction of the tapes in the longitudinal direction providing flexural stiffness is 50% compared to the UD laminates. So the cross-ply architecture of the tapes does not contribute greatly to the stiffness when compared to the UD laminates. Moreover, the CP laminates also exhibit higher void content and slightly lower crystallinity, which may also contribute to the decrease in flexural properties. However, all the specimens showed a failure behavior typical to that of ductile materials. The specimens started yielding after reaching a maximum stress. A typical stress-strain diagram of the UD and CP laminates is thus shown in Figure 9, which also indicates that the specimens show a double yielding behavior. Double yielding behavior has been widely recognized for semicrystalline polymers like PE and its blends. The mechanism of double yielding is related to the microstructure of the polymer and there exist three plausible mechanisms to explain this behavior. Li et al. detected a double yielding behavior in structurally different injection molded polycarbonate/high-density polyethylene blends based on their morphology, where the first yield point was due to the high-density polyethylene and the second yield point was attributed to the injection molded polycarbonate fibers. It is suggested that a similar reason may be proposed for the all-PP composites, where the copolymer matrix may be responsible for the first yielding and the reinforcing PP homopolymer responsible for the second yield point. Nevertheless the results indicate that the short-term as well as long-term viscoelastic behaviors are greatly influenced by the composite morphology, particularly the reinforcement architecture.

### Conclusion

Both unidirectional and cross-ply composites were successfully prepared from all-PP tapes by vacuum bag molding in an autoclave. The effects of reinforcing architecture and temperature on flexural creep of all-PP composites were investigated. The results showed that the UD laminates offered greater resistance to creep compared to the CP ones. Also a change from cross-ply to unidirec-
tional reinforcement architecture improved the elastic recovery and decreased the recovered compliance of the composites significantly. From the short-term flexural creep response, the long-term creep behavior of the all-PP composites could be predicted. It was found that an Arrhenius type of relationship could better fit the experimental time-temperature superposition data and the relaxation behavior of the laminates could be described by a two-component model. The activation energies for the laminates and different relaxation processes were calculated based on the Arrhenius equation and were found to higher for the UD laminates. Further, the results of static flexural tests also showed a good resemblance with the long-term viscoelastic behavior. The results suggested that the viscoelastic behavior of the laminates is strongly dependent on their resulting morphology. A higher volume fraction of the tapes in the longitudinal direction remarkably increased the strength and stiffness of the all-PP composites. Interestingly, all the specimens of unidirectional and cross-ply laminates showed a double yielding behavior. It was suggested that the copolymer matrix might be responsible for the first yielding and the reinforcing PP homopolymer responsible for the second yield point.

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