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Influence of Annealing on Tensile Creep Life of CF/PP Unidirectional Tape

by

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Abstract

This study investigated the effect of annealing on the creep life of carbon fiber/polypropylene unidirectional tape (CF/PP UD tape) under tensile load. First, results demonstrated that annealing promotes crystallization of the matrix resin PP in the CF/PP UD tape, thereby changing the viscoelasticity of the matrix PP. Next, the change in viscoelasticity of the matrix PP caused by annealing was predicted to control the creep life of the CF/PP UD tape. Actual creep tests confirmed approximately this prediction. Specifically, the predicted creep life under constant tensile load at near-room temperature for CF/PP UD tape annealed at 140°C for 1 hour was four times that of tape annealed at 120°C for 1 hour. Furthermore, results demonstrated that, at actual operating temperatures sufficiently below annealing temperature, neither does crystallization of the matrix resin PP in the CF/PP structure progress over the long term, nor does the phenomenon of creep life extension occur.

Key words: Annealing, CFRP, Creep Life Prediction, Polypropylene, Viscoelasticity

Introduction

Carbon fiber reinforced plastics (CFRPs) durability is highly dependent on the environmental effects of temperature, water absorption and so on because of the viscoelastic behavior of matrix resin. Many research papers have described examinations of the long-term life prediction of durability of CFRP under actual environmental conditions.^{1–11} Several studies have investigated the durability of composite materials by focusing on the mechanical properties of the matrix. Yildirim et al.⁸ have demonstrated that mechanical changes in the matrix caused by environmental conditions significantly influence the mechanical performance and damage progression of unidirectional carbon fiber/polyether ketone ketone (CF/PEKK) composites. Ulus et al.⁹ investigated the quasi-static and thermo-viscoelastic behavior of carbon/epoxy-aluminum laminates and demonstrated that changes in the thermo-viscoelastic stiffness of the matrix have a significant influence. Several studies have investigated methods for predicting the long-term life of composite materials based on the concept of accelerating rate of the viscoelastic behavior of the matrix resin. For example, Barbero¹⁰ introduced the effective time-temperature superposition to predict long-term viscoelastic behavior from short-term experimental data. Spathis et al.¹¹ predicted the creep rupture time of polymer composites. The creep strain rate was calculated based on a thermally activated rate process.

The authors have proposed an accelerated testing methodology (ATM)¹² for predicting the long-term life of CFRPs based on Christensen's viscoelastic crack kinetic theory.^{13,14} The validity of ATM has been confirmed as applicable to the creep and fatigue strengths of CFRP with a

thermosetting resin matrix.¹⁵ By using a formulation based on Christensen's viscoelastic crack kinetic theory, Weibull distribution of static strength of CFRP at room temperature and static strengths at various temperatures and the viscoelastic properties of the matrix resin, for which the time-temperature superposition principle holds, can be used to predict the long-term creep strength of CFRP including the scatter in the time to failure.

The crystallinity of polypropylene (PP) is well known to increase with annealing, which in turn increases its mechanical properties.^{16–19} Findings from our earlier study²⁰ showed that the crystallization on PP progresses upon annealing, that this progress follows a time-temperature superposition principle, that the viscoelastic properties of PP change as crystallization progresses, and that the same time-temperature superposition principle applies to these changes.

In this study, we focused on carbon fiber/polypropylene unidirectional tape (CF/PP UD tape) — a material for which we had previously demonstrated the applicability of our ATM²¹ — and investigated how the creep strength of the tape is affected by the progression of PP crystallization. First, it is demonstrated that exposing PP plates and CF/PP UD tapes to a constant temperature for a long period of time promotes PP crystallization, which appears as a change in the PP's viscoelasticity. Next, CF/PP UD tapes are annealed under two temperature conditions. In addition, the tape's creep life is predicted considering the change in the PP's viscoelasticity using the ATM method. Then that prediction is confirmed by experimentation. Finally, the possibility of extending the creep life at practical temperatures is discussed. Another objective is to clarify the effect of the large activation energy in the crystallization of PP on the creep life of CF/PP UD tapes.

Formulations

The authors developed ATM for evaluation of the statistical time-dependent and temperature-dependent creep and fatigue strengths of CFRP and structures.¹⁵ This methodology was formulated based on the matrix resin viscoelasticity. The most important condition for this formulation is the modified time–temperature superposition principle (modified TTSP) holding for the matrix resin viscoelasticity.²²

When an arbitrary load is applied to CFRP, the strain generated in the matrix resin of CFRP is at most the same as the elongation of the carbon fiber. Within this strain range, the stress-strain relationship of the matrix resin is assumed to be linear viscoelastic.

The formulations for the viscoelastic behavior of matrix resin, static, creep and fatigue strengths of CFRP used in this paper are referred from Ref. [15].

Matrix resin viscoelasticity

The master curve of relaxation modulus E_r of matrix resin can be found by horizontal and vertical shifting of the relaxation moduli measured at various temperatures based on the modified TTSP, as presented in Figure 1. The master curve of the relaxation modulus of matrix resin can be formulated as equation (1). The time–temperature (horizontal) shift factor a_{T0} and temperature (vertical) shift factor b_{T0} can also be formulated as equations (2) and (3) belows.¹⁵

$$\log E_r(t, T_0) = \log E_r(t_0, T_0) - \log \left[\left(\frac{t}{t_0} \right)^{m_1} + \left(\frac{t}{t_g} \right)^{m_2} \right] \quad (1)$$

Therein, t , t_0 and t_g are time, reference time and glass transition time, respectively. m_1 and m_2 are the slopes at short time region and long time region as shown in Figure 1, respectively.

$$\begin{aligned} \log a_{T_0}(T) = & \frac{\Delta H_1}{2.303G} \left(\frac{1}{T} - \frac{1}{T_0} \right) H(T_{gh} - T) \\ & + \left[\frac{\Delta H_1}{2.303G} \left(\frac{1}{T_{gh}} - \frac{1}{T_0} \right) + \frac{\Delta H_2}{2.303G} \left(\frac{1}{T} - \frac{1}{T_{gh}} \right) \right] (1 - H(T_{gh} - T)) \end{aligned} \quad (2)$$

$$\log b_{T_0}(T) = b_1(T - T_0)H(T_{gv} - T) + [b_1(T_{gv} - T_0) + b_2(T - T_{gv})](1 - H(T_{gv} - T)) \quad (3)$$

Therein, ΔH_1 and ΔH_2 represent activation energies at low and high temperature regions. In addition, H is Heaviside step function. G is the gas constant. T_0 is a reference temperature. T_{gh} and T_{gv} are knee point temperatures for a_{T_0} and b_{T_0} , respectively. b_1 and b_2 are the parameters for vertical shift factor.

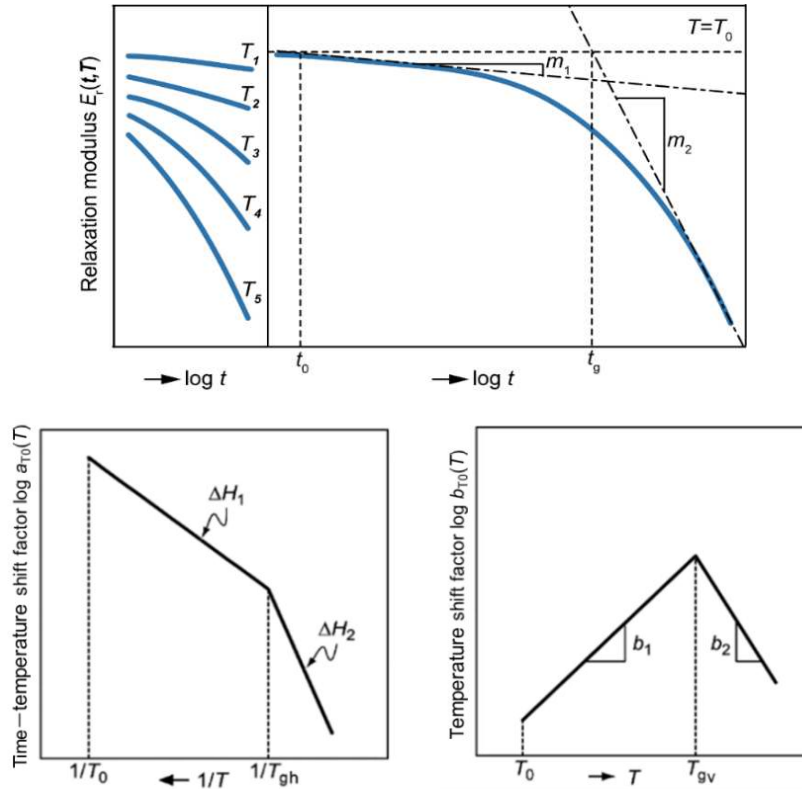


Figure 1. Master curve of the relaxation modulus and horizontal and vertical shift factors for resin.

Effect of annealing on matrix resin viscoelasticity

The knee point temperature T_g , which is an indicator of the heat resistance of a resin, is definable as the temperature at which the storage modulus at any constant frequency f obtained in a DMA test drops sharply with increasing temperature, as shown in the upper diagram of Figure 2. The knee point temperature T_g versus annealing time t_d at various annealing temperatures T_d measured in this way is obtainable as shown at the left side of middle diagram of Figure 2. The master curve of knee point temperature T_g versus annealing time t_d at a reference annealing temperature T_{d0} is obtainable as shown in equation (4) and the right side of the middle diagram of Figure 2 based on the time–temperature superposition principle for the annealing time and temperature.

$$T_g = T_{g0} + F_d(\log t_d - \log t_{d0}) \quad \text{at } T_{d0} \quad (4)$$

The time–temperature (horizontal) shift factor for the annealing time and temperature is obtainable as shown in equation (5) and the lower diagram of Figure 2.

$$\begin{aligned} \log a_{T_{d0}}(T_d) = & \frac{\Delta H_{d1}}{2.303G} \left(\frac{1}{T_d} - \frac{1}{T_{d0}} \right) H(T_{dg} - T_d) \\ & + \left[\frac{\Delta H_{d1}}{2.303G} \left(\frac{1}{T_{dg}} - \frac{1}{T_{d0}} \right) + \frac{\Delta H_{d2}}{2.303G} \left(\frac{1}{T_d} - \frac{1}{T_{dg}} \right) \right] (1 - H(T_{dg} - T_d)) \end{aligned} \quad (5)$$

Therein, ΔH_{d1} and ΔH_{d2} represent activation energies at low and high temperature regions. In addition, H is Heaviside step function. G is the gas constant. T_d is annealing temperature. T_{d0} is a reference annealing temperature. T_{dg} is knee point temperatures for a_{T0} .

The change in the knee point time t_g on the master curve of the relaxation elastic modulus by annealing is calculable using the following equation from the change in the knee point temperature T_g .

$$\log t_{g0} - \log t_g = \log a_{T_{g0}}(T_g) = \frac{\Delta H_1}{2.303G} \left(\frac{1}{T_g} - \frac{1}{T_{g0}} \right) \quad (6)$$

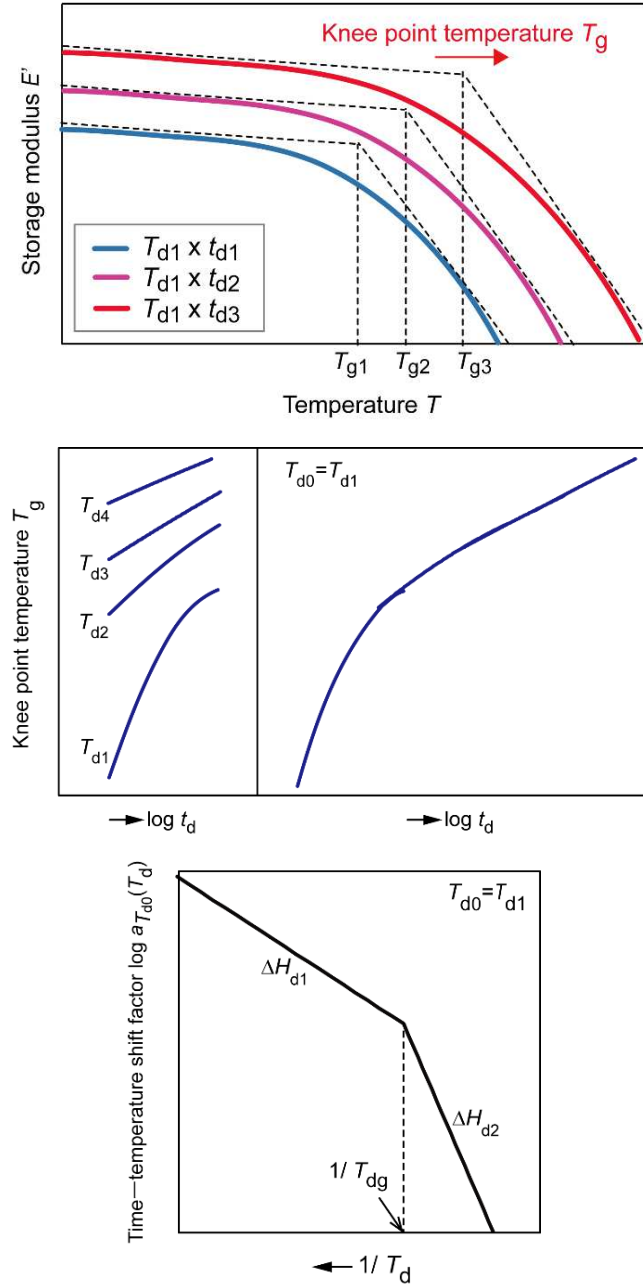


Figure 2. Definition of knee point temperature, master curve of knee point temperature versus annealing time, and shift factor for the annealing time and temperature for resin.

Statistical static and creep strengths of CFRP

The statistical static strengths under an arbitrary constant strain rate and temperature are given by the following formulae.

$$\log \sigma_s(t, T) = \log \sigma_0(t_0, T_0) + \frac{1}{\alpha} \log[-\ln(1 - P_f)] + n_R \log \left[\frac{E_s(t, T)}{E_r(t_0, T_0)} \right]$$

$$E_s(t, T) = E_r(t/2, T) \quad (7)$$

Therein, t and t_0 respectively represent the failure time and reference failure time, T and T_0 respectively denote the temperature and reference temperature, and P_f stands for the failure probability. In addition, σ_0 and α respectively denote the scale and shape parameters of static strength at t_0 and T_0 . Also, n_R is a viscoelastic parameter determined by the failure mode. E_r is the relaxation modulus of the matrix resin.

Parameters σ_0 , α , and n_R in equation (7) representing time-dependent and temperature-dependent statistical static strengths are obtainable from static tests of CFRP conducted at various temperatures under a constant strain rate.

Statistical creep strength σ_c can be ascertained by shifting the static strength with $\log A$. Therefore, creep strength σ_c can be denoted by subscript “s” replaced with “c” in equation (7), as presented below. In this equation, E_c represents the viscoelastic modulus for a constant stress load. It has been clarified that the master curve of creep strength is obtainable by shifting the master curve of static strength by $\log A$, as shown in Figure 2.

$$\log \sigma_c(t, T) = \log \sigma_0(t_0, T_0) + \frac{1}{\alpha} \log[-\ln(1 - P_f)] + n_R \log \left[\frac{E_c(t, T)}{E_c(t_0, T_0)} \right]$$

$$E_c(t, T) = E_s(At, T) = E_r(At/2, T) \quad (8)$$

The master curve of viscoelastic modulus E_c at reference temperature T_0 can be shown by the following equation using equations (7) and (8).

$$\log E_c(t, T_0) = \log E_r \left(\frac{A_1 t_0}{2}, T_0 \right) - \log \left[\left(\frac{A_1 t}{2t_0} \right)^{m_1} + \left(\frac{A_2 t}{2t_g} \right)^{m_2} \right] \quad (9)$$

For the case of tension loading applied to unidirectional CFRP, the shifting amount $\log A$, as ascertained from slope k_R of the logarithmic static strength against the logarithmic failure time curve, is calculated using the following equation.

$$\log A_i = \log \left(1 + \frac{1}{k_{Ri}} \right), k_{Ri} = m_i \times n_R, i = 1, 2 \quad (10)$$

Therein, m_i represents the slope of the logarithmic relaxation modulus of the matrix resin against the logarithmic time curve presented in Figure 1.

It is thought that the state of the interface between the carbon fiber and the resin changes as annealing progresses. The change in the interface state can be understood as a change in three parameters, namely, σ_0 , α , and n_R in equations (7) and (8).

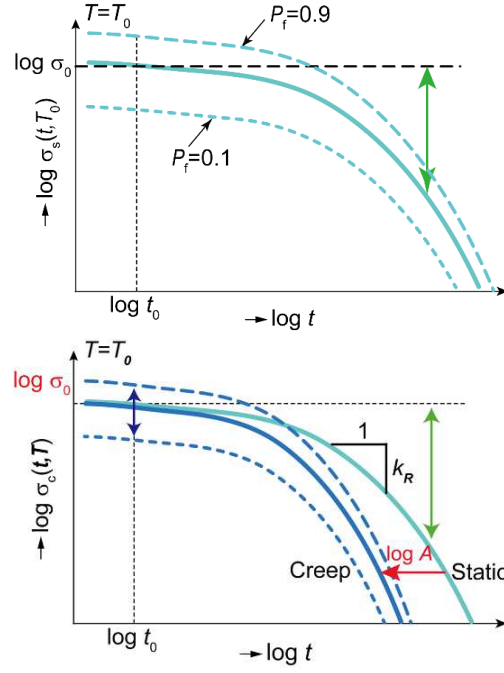


Figure 3. Master curves of statistical tensile static and creep strengths versus failure time for unidirectional CFRP.²

Experimental Procedures

CF/PP UD tape used for this study

In this study, a carbon fiber/polypropylene unidirectional tape (CF/PP UD tape) was prepared using standard modulus grade carbon fiber (Elastic modulus: 240 GPa, Tensile strength: 3,600 MPa, Elongation: 1.5%, Filament diameter: 7 μm) with 137 g/m fiber area weight and volume fraction of fiber $V_f = 50\%$.²¹ Commercial polypropylene with an appropriate melt flow rate (MFR: JIS K 7210-1: 2014, ISO 1133-1: 2011, MFR = 30 g/10 min at 230°C, and 0.91 g/cm³ density) is used for this CF/PP UD sheet. The sheet thickness was controlled as $t = 0.16$ mm. A continuous coil of the CF/PP UD sheet was cut into 2.0 mm width tapes using a slitting machine for tape specimens. Furthermore, it was confirmed that no loose fiber came out from the tape edge. Basic annealing was applied under 120°C for 1 h to stabilize the crystallinity of the specimens. PP plates with thickness $t = 1.6$ mm were also prepared for the evaluation of viscoelasticity. Bending tests at room temperature have confirmed that the stress-strain relationship of this PP plate is linear up to 1.5%, the elongation of the carbon fiber used.

Annealing of CF/PP UD tapes and PP plates

The CF/PP UD tapes and PP plates which had been basically annealed at 120°C for 1 h were then additionally annealed at temperatures of 120°C, 140°C, 150°C, and 160°C, respectively, for 1 h, 10 h, 50 h, and 100 h. However, the annealing at 160°C was conducted for only 1 h. For annealed CF/PP UD tapes and PP plates, surface observation was carried out and the temperature dependent storage modulus was measured by DMA using RSA-III (TA Instruments).

Two annealing conditions were selected from the above results of CF/PP UD tapes and PP plates:

one annealed at 120°C for 1 h and one annealed at 140°C for 1 h. The relaxation modulus for the PP plates at selected two annealing conditions was determined over a wide range using a combination of DMA and creep tests using creep testing machine CP6-D-6 (T·S·E Co., Ltd.).

Static and creep strength tests for CF/PP UD tapes

The static strength testing machine (Autograph AGS-5KNG, Shimadzu Corp.) with a temperature chamber (Custom-made products, Satoseiko Co., Ltd.)²¹ realized a constant elongation rate over a wide range and constant temperatures in the range of room temperature to 150°C. The creep strength testing machine with a temperature chamber (Custom-made products, Satoseiko Co., Ltd.)²¹ also realized a constant load at the maximum load of 1 kN and constant temperature in the range of room temperature to 150°C, as shown in Figure 4. Test conditions for static and creep tests are presented respectively in Tables 1 and 2.

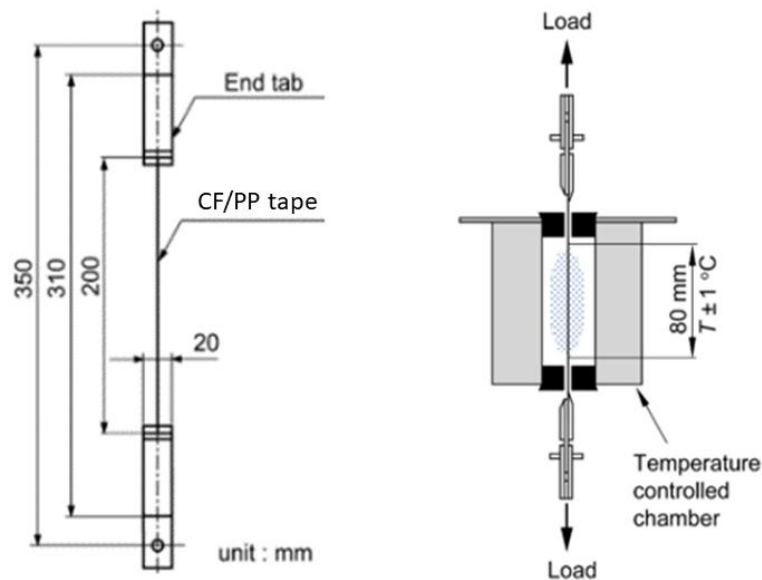


Figure 4. Tensile static and creep strenght tests of CF/PP UD tape.

Table 1. Numbers of specimens at static test temperatures after annealing.

Annealing condition	25°C	80°C	120°C	145°C	150°C
120°C × 1 h	20	20	20	20	20
140°C × 1 h	20	5	5	-	5

Table 2. Number of specimens at creep test conditions after annealing.

Annealing condition	σ_{c0} [MPa]	T [°C]	Number of specimens
120°C × 1 h	921	120	20
140°C × 1 h	921	120	21

Results and Discussion

Surface smoothness of CF/PP UD tape

Figure 5 presents surface images of CF/PP UD tapes after annealing at various conditions. The tape surface after annealing at low temperatures maintains the surface smoothness. However this figure shows that those at high temperatures do not maintain the surface smoothness. The arrows in the figure indicate the grooves that have formed on the surface. The surface smoothness persists for all of PP plates after annealing at all conditions. Annealing at temperatures of 150°C or higher for CF/PP UD tapes is of no practical significance.

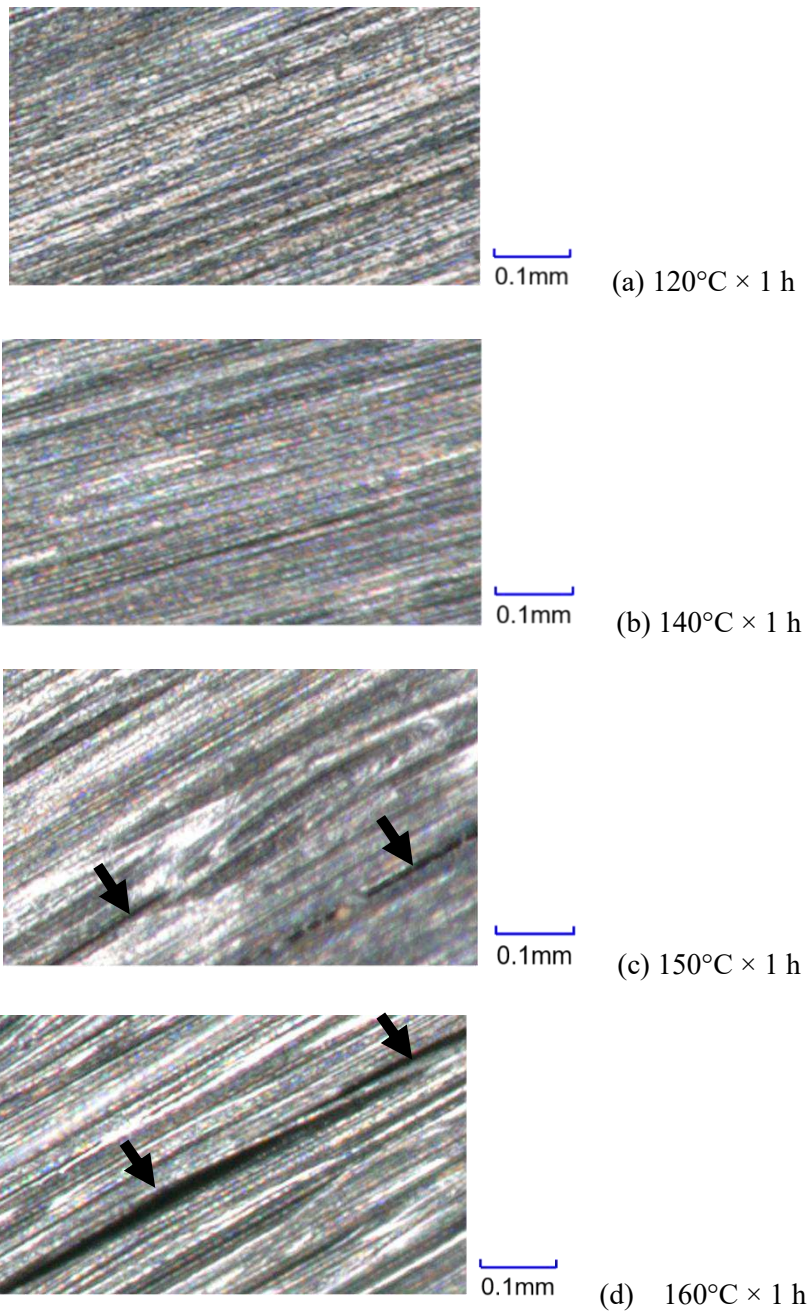


Figure 5. Surface images of CF/PP UD tapes after annealing under various conditions.

The DMA tests described in the next section were performed only on specimens annealed under conditions that allowed the smoothness of the specimen surface to be maintained. For CF/PP UD tapes that had been annealed at high temperatures for long periods of time, the presence of grooves along the fibers on the specimen surface shown in Figure 5 made it impossible to apply a load perpendicularly to the fibers. The test was therefore abandoned.

Knee point temperature and crystallinity of PP by annealing

DMA tests by double cantilever method were conducted on PP sheets and CF/PP UD tapes oriented along the fiber direction under various annealing conditions at a frequency of $f = 1$ Hz. The temperature dependence of the storage modulus was determined using bending equations based on the applied deflection and load response shown in Figure 6. While the PP sheet undergoes bending deformation upon the application of deflection, the CF/PP UD tape primarily experiences shear deformation within the PP layer due to the vast difference in elastic modulus between the carbon fibers and the PP. Consequently, the deformation of the CF/PP UD tape is also governed by the elastic modulus of the PP. As shown in Figure 6, the temperature-dependent changes for both materials were nearly identical. Therefore, the viscoelastic properties of the PP within the CF/PP UD tape were treated as being equivalent to those of the PP sheet. In this figure, the temperature at which the storage modulus drops sharply with temperature is definable as the knee-point temperature T_g .

The relation between the knee point temperature and annealing time at various annealing temperatures for PP plates and CF/PP UD tapes is shown in the left panel of Figure 7. The right panel portrays the master curve of knee point temperature versus annealing time for PP plates and CF/PP UD tapes at reference annealing temperature $T_{d0} = 120^\circ\text{C}$.

The left panel in Figure 7, showing data from our earlier study,²⁰ depicts the relation between the PP plate crystallinity and the annealing time under various annealing temperature conditions. The right panel is the master curve when the reference temperature is 120°C . From this figure, it is apparent that the knee-point temperatures of PP plates and CF/PP UD tapes are the same if the annealing conditions are the same. Moreover, the same time–temperature superposition principle holds for annealing time and annealing temperature for PP plates and CF/PP UD tapes because the time–temperature shift factors of PP plates are equal to those of CF/PP UD tapes presented in Figure 9.

The left panel of Figure 8 portrays the relation between crystallinity and annealing time at various annealing temperatures for PP plates. The crystallinity was evaluated by wide and small angle X-ray scattering analysis (WAXS/SAXS) (NANO-Viewer; Rigaku Corp.). This figure was presented in a report of our earlier work.²⁰ The right panel is the master curve of crystallinity versus annealing time at a reference annealing temperature $T_{d0} = 120^\circ\text{C}$. The smooth master curve suggests that the time–temperature superposition principle holds for the crystallization of the PP plates.

Figure 9 shows the time–temperature shift factors for annealing time and temperature for crystallinity and annealing. The time–temperature shift factor for annealing agrees well with that for crystallinity. This agreement clarifies that the knee-point temperature is closely related crystallinity.

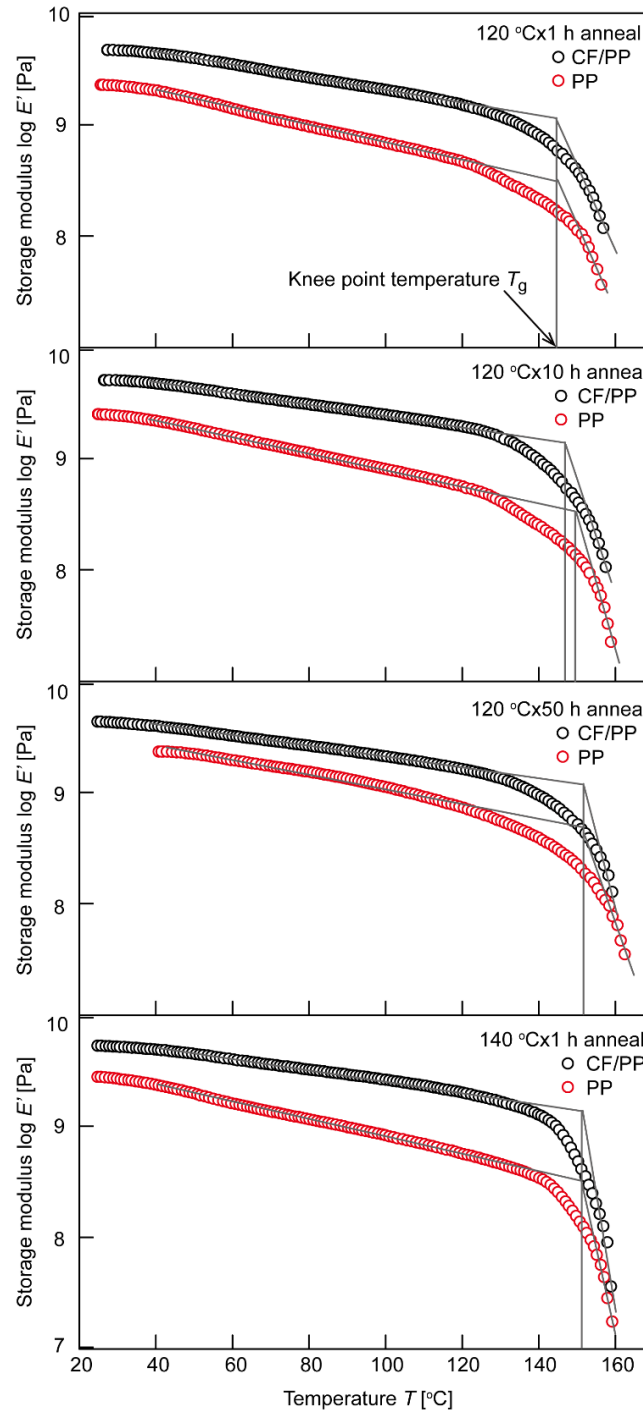


Figure 6. Storage modulus versus temperature at frequency $f = 1$ Hz for PP plates and CF/PP UD tapes under various annealing conditions.

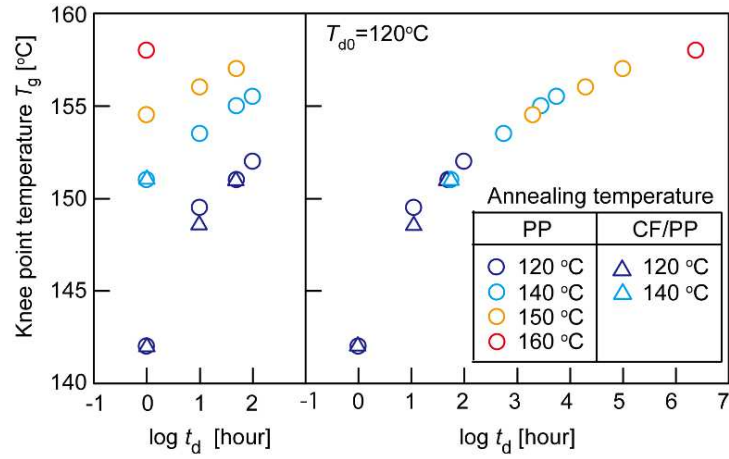


Figure 7. Master curve of knee point temperature T_g versus annealing time at $T_{d0} = 120^\circ\text{C}$ for PP plates and CF/PP UD tapes.

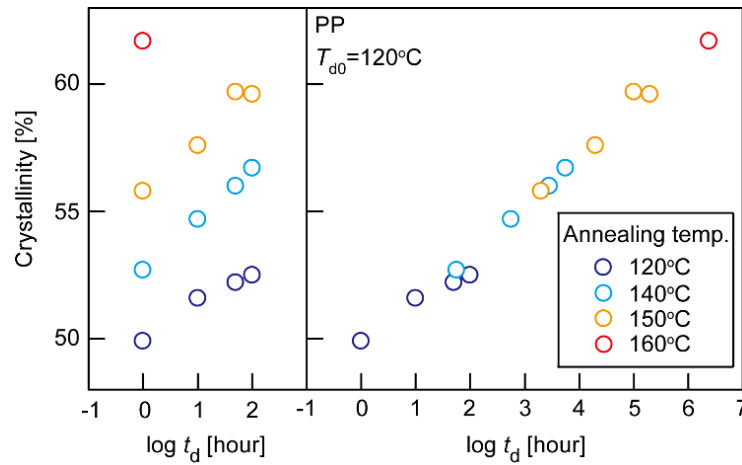


Figure 8. Master curve of crystallinity versus annealing time at $T_{d0} = 120^\circ\text{C}$ for PP plates.

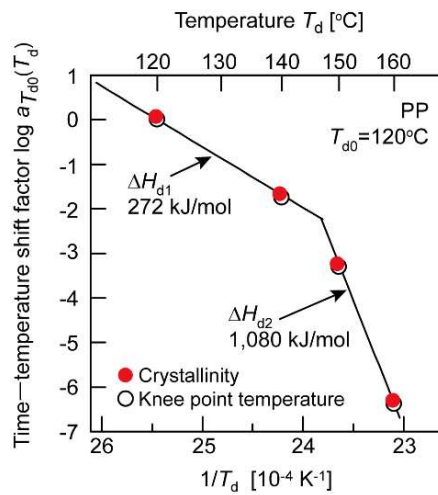


Figure 9. Time-temperature shift factor for crystallinity of PP plates and knee point temperature of PP plates and CF/PP UD tapes.

Determination of parameters for the relaxation modulus of PP

The upper and lower panels of Figure 10 respectively present the relaxation moduli of PP plates annealed at 120°C for 1 h and at 140°C for 1 h. The left of each panel shows the relaxation moduli of PP plates for widely various times at various temperatures, as obtainable by combining two test results from the DMA and creep testing machine. The right of each panel presents the master curve of relaxation moduli versus reduced time t' at reference temperature $T_0 = 25^\circ\text{C}$, which is constructed by horizontal and vertical shifting of the relaxation moduli measured at various temperatures shown at the left of these panels. The time–temperature (horizontal) shift factors and the temperature (vertical) shift factors based on the modified time–temperature superposition principle are shown in Figure 11.

If the master curve of the relaxation modulus of resin shown on the right of Figure 10 is expressed by equation (1), then the parameters will be as presented in Table 3. Similarly, if the time–temperature (horizontal) shift factor and temperature (vertical) shift factor shown in Figure 11 are expressed by equations (2) and (3), then the parameters will also be as shown in Table 3. The differences between the two annealing conditions are apparent in E_r and t_g , as shown in yellow hatching, but not in other parameters.

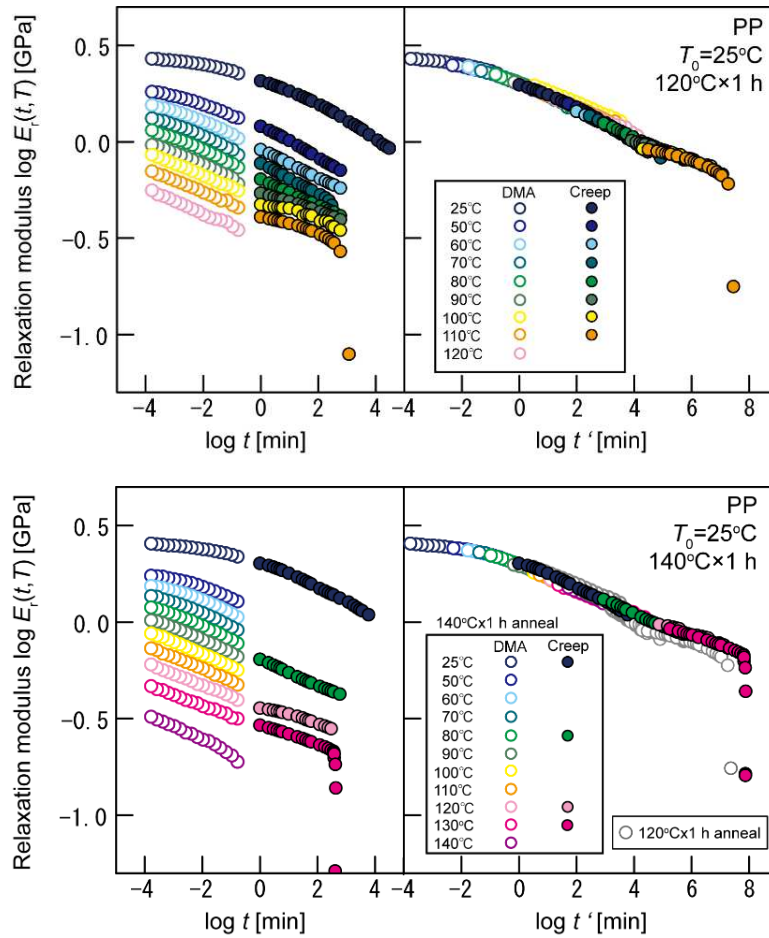


Figure 10. Master curves of relaxation modulus of PP plates for two annealing conditions.

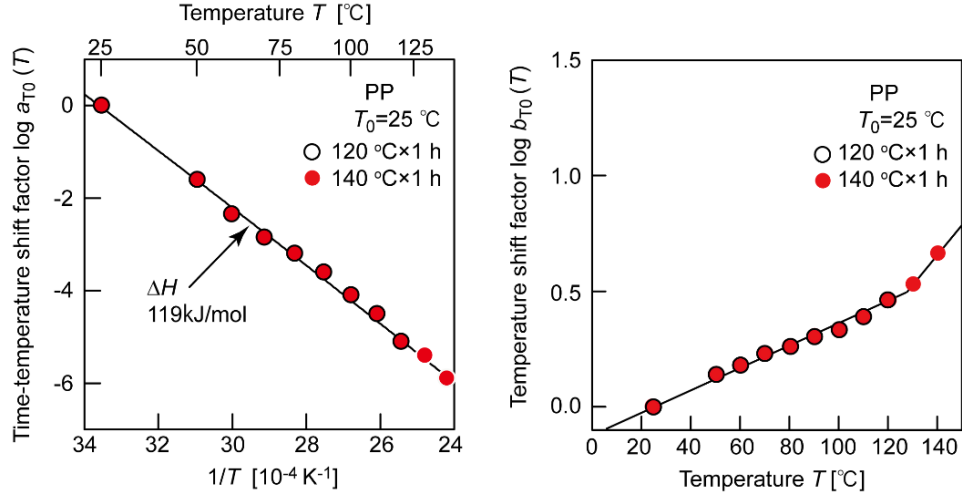


Figure 11. Time–temperature (horizontal) and temperature (vertical) shift factors for the relaxation modulus of PP plates for two annealing conditions.

Table 3. Parameters of relaxation moduli for two annealing conditions

Annealing condition	120°C × 1 h	140°C × 1 h
Reference temperature T_0 [°C]	25	25
Reference time t_0 [min]	1.0	1.0
$E_r(t_0, T_0)$ [GPa]	2.16	2.47
m_1	0.067	0.067
m_2	20	20
t_g [min]	2.3E+07	9.8E+07
ΔH_1 [kJ/mol]	119	119
b_1	0.0048	0.0048

Statistical static strengths of CF/PP UD tape

Figure 12 presents the static strength versus temperature for CF/PP UD tape for two annealing conditions. The static strength for both conditions decreases markedly with increasing temperature.

Weibull distributions for static strengths at various temperatures for both CF/PP UD tape are depicted in Figure 13. In this figure, α_s is the shape parameter and β_s is the scale parameter for each of the CF/PP UD tapes. The shape parameter maintains an almost constant value for both CF/PP UD tapes. Shape parameters α_s and scale parameters β_s at temperature $T = 25^\circ\text{C}$ in this figure can be inferred as shape parameter α , scale parameter σ_0 of static strength at the reference temperature $T_0 = 25^\circ\text{C}$, and the reference failure time $t_0 = 1$ min used in equation (1).

Figure 14 presents the non-dimensional static strength σ_s/σ_0 of a CF/PP UD tape against the non-dimensional viscoelastic modulus of matrix resin E_s^*/E_{r0} for the same conditions of time and temperature. The relation of σ_s/σ_0 against E_s^*/E_{r0} can be represented as one solid straight line with

slope n_R , which is the viscoelastic parameter in equation (7). The viscoelastic parameters n_R obtained from the experiments agree well with that determined theoretically by substituting the shape parameter of carbon fiber $\alpha_c = 5.9$ shown in Figure 15 into the following equation shown by Rosen's model¹⁵.

$$n_R = \frac{1}{2\alpha_c} \quad (12)$$

All parameters in equation (7) for the master curve of static strength are found for both CF/PP UD tapes, as presented in Table 4. As this table shows, almost no difference exists in the static strength parameters between the two annealing conditions. This fact suggests that there is no difference affecting the parameters shown in Table 4 in the interfacial state between the carbon fiber and the resin between the two annealing conditions. Figure 16 compares the fracture appearance of five representative specimens subjected to static tension tests at 25°C after annealing under two conditions: 120°C for 1 hour and 140°C for 1 hour. Specimens from both annealing conditions shattered into fine fragments due to the impact at the moment of failure, making it impossible to identify any distinct differences between them. Understanding the influence of the interface between carbon fibers and resin is essential for evaluating the long-term service life of CFRP structures in harsh environments, and incorporating this effect into evaluation formulas is an future challenge.

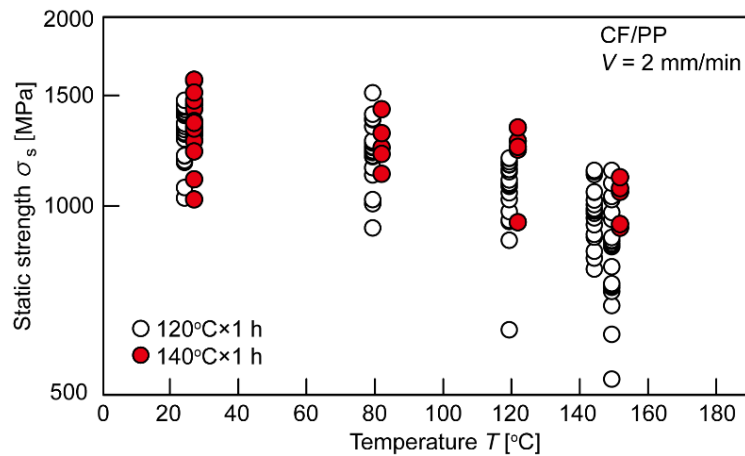


Figure 12. Statistical static strengths versus temperature of CF/PP UD tape for two annealing conditions.

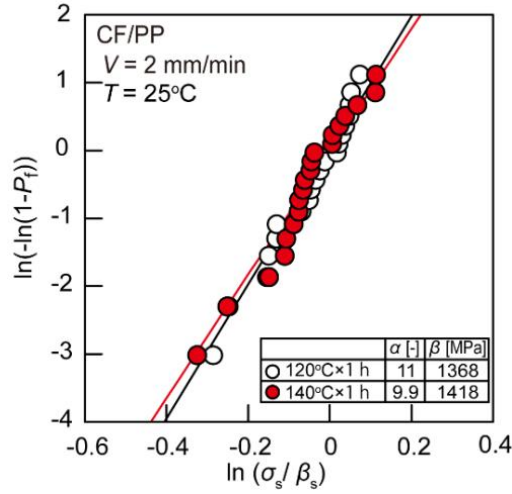


Figure 13. Weibull distributions of statistical static strength of CF/PP UD tape at $T = 25^\circ\text{C}$ for two annealing conditions.

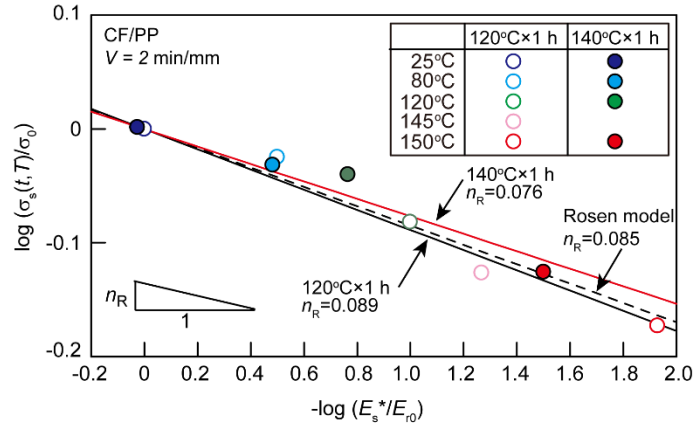


Figure 14. Relation between static strength of CF/PP UD tape and viscoelastic modulus of PP plates at the same time and temperature for two annealing conditions.

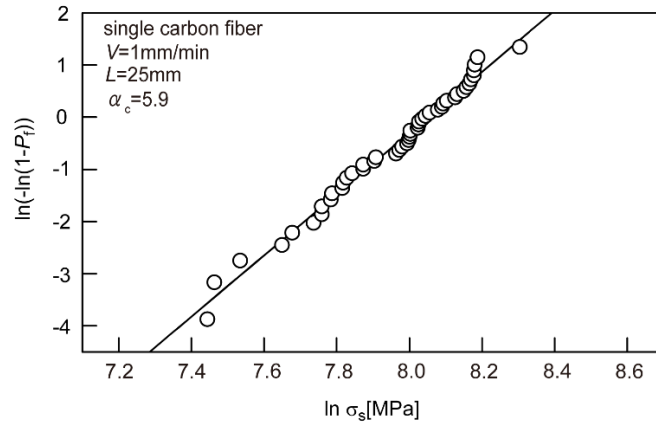


Figure 15. Weibull distribution of static strength for carbon fiber used for this study.

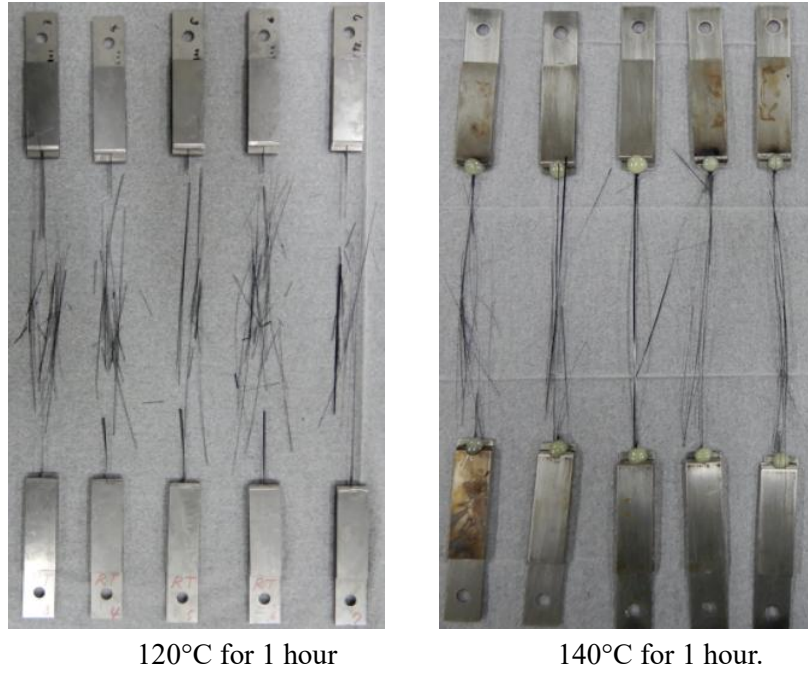


Figure 16. Fractography of five representative specimens subjected to static tension tests at 25°C after annealing under two conditions.

Table 4. Parameters for creep strength of CF/PP UD tape for two annealing conditions

Annealing condition	120°C × 1 h	140°C × 1 h
Scale parameter σ_0 [MPa]	1368	1418
Shape parameter a	11	9.9
Viscoelastic parameter n_R	0.089	0.076
Shift factor A	177	182

Prediction of statistical creep life of CF/PP UD tape

Creep failure tests of CF/PP tapes after treatment under annealing conditions of two kinds were conducted at an arbitrary constant stress level and at one constant temperature $T = 120^\circ\text{C}$. Figure 17 presents the results. The predicted curves in this figure are calculated by substituting the parameters of two annealing conditions shown in Table 4 into equation (8) after calculating the relaxation moduli under two annealing conditions by substituting the parameters in Table 3 in equations (1), (2), and (3). In other words, this prediction does not incorporate consideration of the progress of annealing during loading.

The predicted value differed from the experimentally obtained value probably because of the progression of annealing, i.e., recrystallization, because of exposure to a 120°C environment. Figure 18 shows the predicted knee point temperature T_g against elapsed time t obtained from Figure 9 for the two annealing conditions. The predicted knee point time t_g obtained from equation (6) is also

presented in this figure. It can be predicted that the T_g and t_g of CF/PP tape annealed at 120°C for 1 hour increase over time, approaching these of CF/PP tape annealed at 140°C for 1 hour.

The upper graph in Figure 19 presents the relation between the probability of creep strength failure and elapsed time, as predicted with consideration of the progression of annealing over time, along with the experimentally obtained results. The lower graph in this figure shows the experimentally obtained and predicted values for the difference in logarithmic fracture time under two annealing conditions with the same failure probability against the failure time under an annealing condition of 120°C for 1 h. As this figure shows the failure time for annealing condition of 120°C for 1 h approaches that for the annealing condition of 140°C for 1 h as the elapsed time increases for both the experiment and the prediction.

As explained above, when a constant tensile load is applied to a CF/PP UD tape, the progress of annealing of the PP during loading can extend the tape creep life. Conversely, it can be said that if an annealing treatment is performed at 140°C for one hour, stability is maintained without recrystallization proceeding, even in an operating environment at 120°C for over 100 hours.

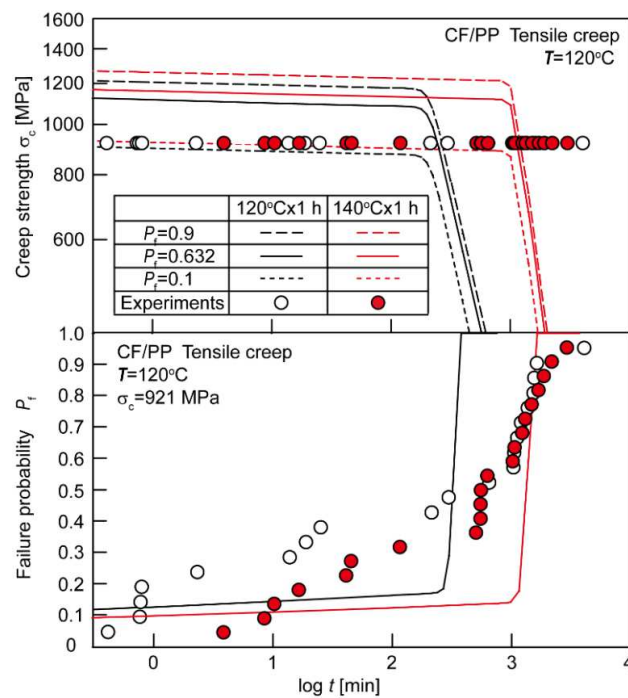


Figure 17. Predicted and measured tensile creep strength and failure probability versus failure time.

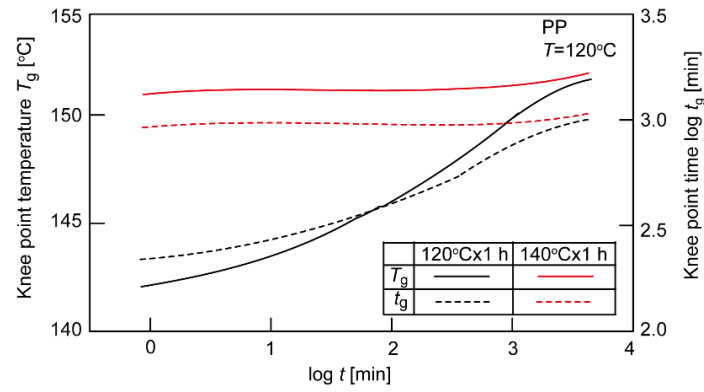


Figure 18. Predicted knee point temperature and knee point time against elapsed time.

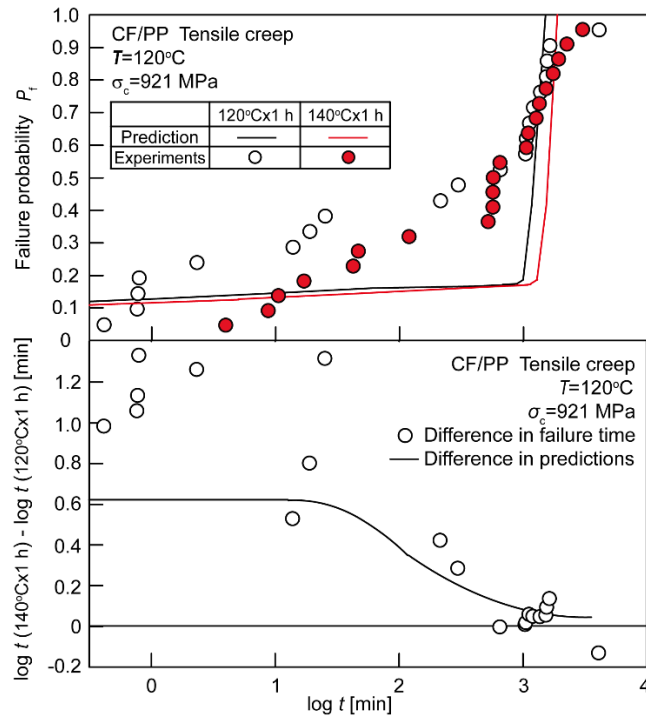


Figure 19. Predicted and measured failure probability and annealing progress versus failure time.

Prediction of long-term creep life for CF/PP UD tape at various operating temperatures

Based on the aforementioned results, the predicted relationship between creep rupture strength and time to failure for CF/PP UD tapes annealed under two conditions—120°C for 1 h and 140°C for 1 h—at projected service temperatures of 25°C, 50°C, 85°C, and 120°C is shown in Figure 20. At service temperatures of 25°C, 50°C, and 85°C, the creep life of the CF/PP tape annealed at 140°C for 1 h is approximately four times longer than that of the tape annealed at 120°C for 1 h. This can be readily inferred from the fact shown in Table 3 that the knee-point time t_g of the PP sheet annealed at 140°C for 1 h is approximately four times that of the sheet annealed at 120°C for 1 h. Furthermore, given that the activation energy for crystal growth (272 kJ/mol) shown in Figure 9 is significantly higher than the activation energy (119 kJ/mol) observed for viscoelastic properties (deformation phenomena) shown in Table 3, it can be concluded that crystal growth is suppressed at these service temperatures, resulting in no change to the creep life. In contrast, at a service temperature of 120°C,

crystallization proceeds under load for specimens prepared under both annealing conditions (120°C for 1 h and 140°C for 1 h); since the crystallization of the former catches up to that of the latter, the creep lives become equivalent. Therefore, it is considered that if annealing is performed at a temperature sufficiently higher than the service temperature, further annealing will not occur during subsequent long-term use. Figure 20 serves as a concrete illustration of this fact.

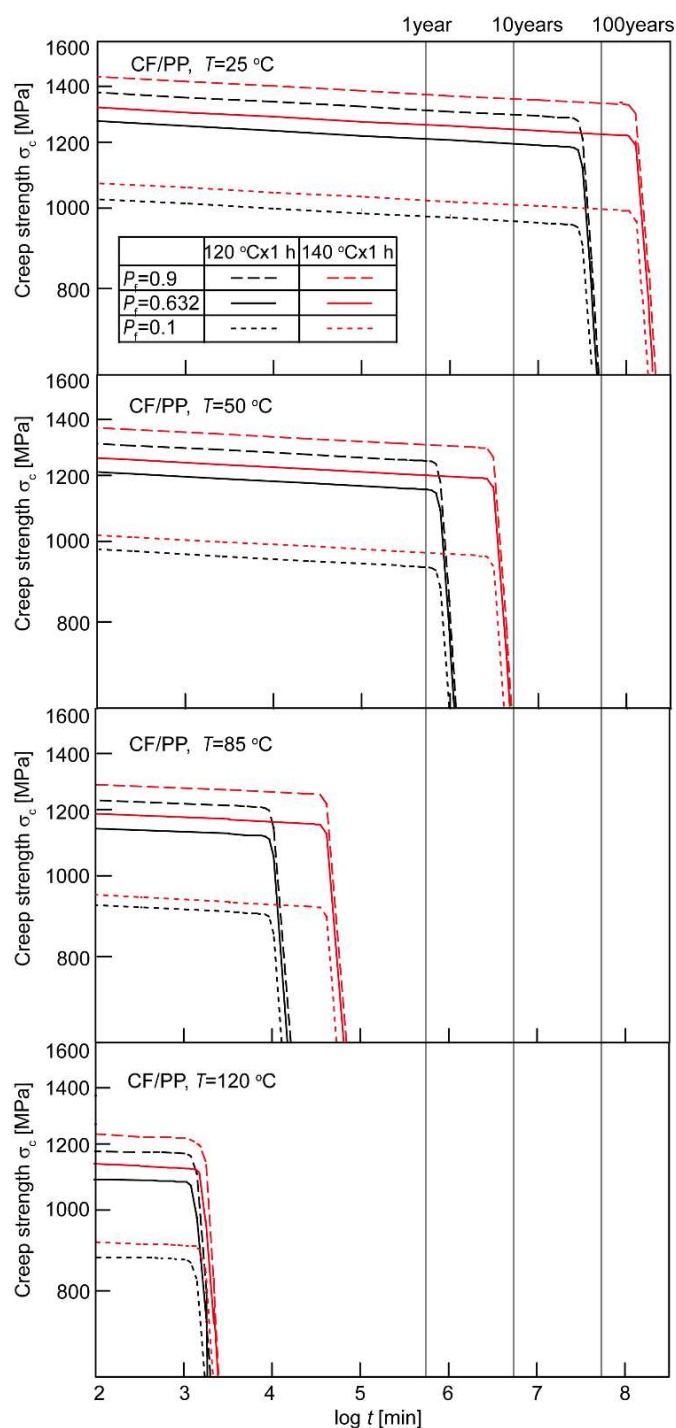


Figure 20. Creep strength versus elapsed time under four operating temperature conditions for CF/PP UD tapes molded under two temperature conditions.

Conclusion

An investigation into the effect of annealing on the tensile creep life of CF/PP UD tape yielded the following conclusions:

1. A previous study established that the time-temperature superposition principle applies to the relationship between annealing time and annealing temperature regarding the viscoelastic properties of neat PP. The present study further revealed that the viscoelastic properties of the PP matrix in CF/PP UD tape are identical to those of neat PP. These findings provide the basis for applying the previously developed ATM method to elucidate the effects of annealing on the tensile creep life of CF/PP UD tape.
2. We examined CF/PP UD tape annealed for one hour at 120°C and 140°C—conditions considered realistic—and modeled scenarios where a constant load is applied to these tapes over an extended period in a 120°C environment. Creep life predictions based on the ATM method—accounting for annealing caused by PP recrystallization during both the initial heat treatment and the prolonged loading period—showed general agreement with actual creep test results. These results demonstrate that the ATM method enables an approximate evaluation of how both the initial annealing treatment and the progression of annealing during loading affect the tensile creep life of CF/PP UD tape.
3. The long-term creep life of CF/PP UD tape is significantly influenced by annealing conditions. Specifically, at temperatures of 85°C or lower—typical operating temperatures—the creep life of CF/PP UD tape annealed at 140°C for one hour is approximately four times that annealed at 120°C for one hour. Results demonstrated that, at actual operating temperatures sufficiently below annealing temperature, neither does crystallization of the matrix resin PP in the CF/PP structure progress over the long term, nor does the phenomenon of creep life extension occur. Furthermore, when the operating temperature is set to 120°C, annealing continues during operating for CF/PP UD tape annealed at 120°C for one hour, whereas such progression is substantially suppressed in that annealed at 140°C for one hour. These phenomena arise because the activation energy for the progression of annealing in the PP matrix is considerably higher than the activation energy associated with viscoelastic behavior of PP matrix.

Author contributions

Yasushi Miyano: supervision, formal analysis, writing manuscript, editing, checking and proof reading. Masayuki Nakada: writing manuscript and editing, checking and proof reading. Yoko Morisawa: data collection, writing manuscript, editing and checking. Takeharu Isaki: data collection, writing manuscript, editing and checking. Kiyoshi Uzawa: supervision and formal analysis.

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Declaration of conflicting interests

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Data Availability Statement

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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