



Effect of temperature on bending behavior of woven fabric-reinforced PPS-based composites

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ABSTRACT

The flexural behavior of two kinds of PPS-based composites (carbon and glass fiber woven fabric reinforcements) from 23 to 200 °C was evaluated in terms of load–displacement curves, stiffness, strength and failure mechanisms. With the increase in temperature, the bending stiffness for carbon and glass fiber-reinforced thermoplastic composites decreased by 36.71 and 34.98%, and bending strength decreased by 68.44 and 61.23%, respectively. The failure mode shifted from a brittle fracture to a ductile manner with the increase in temperature owing to the enhanced plasticization of matrix. Dynamic mechanical analysis was performed to characterize the glass transition and decomposition processes of both PPS-based composites and to establish the relationship between temperature and mechanical properties. Compared with the different empirical models, a new simple and stable thermo-mechanical model was proposed to estimate the variation of flexural properties for both thermoplastic composites with temperature.

Introduction

It is essential to understand the relationship between thermal and mechanical responses of thermoplastic composites in the large structure applications. The physical properties of thermoplastic resins are closely associated with their mechanical properties and failure mechanisms of polymer matrix composites at high temperatures [1, 2]. PPS resin has a lower glass transition temperature (T_g) and melting temperature (T_m). The stiffness and strength of PPS resin decrease, whereas the failure strain increases, when temperature exceeds the glass transition temperature (T_g).

The main matrix-dominated properties of PPS-based composites, such as shear and compressive stiffness and strength, also decrease at elevated temperatures.

In recent years, considerable attentions have been paid on mechanical behavior of thermoplastic composites under thermo-mechanical loading conditions. Vieille [3] presented an overview of temperature and environment effects on behavior of PPS-based composite. The thermoplastic composites became significant nonlinear behavior around their glass transition temperature. Park and Chun [4] studied the mechanical properties of glass fiber-reinforced PPS composites with the variation of glass fiber content

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and testing temperature. Results showed that maximum tensile stress and elastic stiffness increased markedly with increasing glass fiber content at below glass transition temperature (T_g) of PPS matrix. Lou and Murthal [5] conducted the mechanical properties of glass fiber-reinforced PPS composites at elevated temperatures by means of tensile, compressive, bending and fatigue experiments. Meyer et al. [6] observed PPS and PEEK thermoplastics without losing too much flexural properties from -60 °C to their glass transition temperatures. The failure behavior was changed from a brittle mode at room temperature to a ductile mode above the glass transition temperature. Vieille and Auccher [7–9] investigated the mechanical properties of carbon fiber fabric-reinforced PPS composites subjected to in-plane shear, compressive, bending and interlaminar shear loading, respectively. The reduction of mechanical properties of composites at above glass transition temperature was attributed to the matrix softened and fiber/matrix interface adhesion. Gabion [10] found that unidirectional carbon fiber-reinforced thermoplastic polyimide composites had outstanding fatigue properties in a temperature range from -50 and 200 °C, but the interlaminar shear strength decreased with increasing temperature. As mentioned above, the experimental results showed that the mechanical properties of thermoplastic composites were greatly dependent on temperature. It was essential to have a better understanding of thermo-mechanical responses of thermoplastic composites.

Several mathematical descriptions for temperature-dependent mechanical properties were established to study comprehensive characteristics of composites at elevated temperatures. Ha and Springer [11] proposed a power law relationship between mechanical properties of carbon fiber-reinforced thermosetting composite and temperature (below T_g). Gibson et al. [12–14] presented a hyperbolic function to study the variation of mechanical behavior of woven glass/vinyl ester laminates within the glass transition region, which was also verified by experimental data. Based on kinetic theory, Keller and Bai [15, 16] derived a modified Coats–Redfern method to estimate the E -stiffness, G -stiffness and viscosity of FRP composites in the glass transition temperature and decomposition temperature of polymer resin. The predicted result was consistent with that obtained by dynamic mechanical analysis (DMA). In Mahieux's

model [17–19], Weibull-type functions were used to describe the stiffness changing of PMMA-, PEEK-, PPS- and AS4/PPS-based composites over the full range of transition temperatures. Recently, Correia [20] developed a phenomenological model to evaluate the mechanical properties of GFRP from ambient temperature to 250 °C. It was verified that the model was suitable to describe the degradation of shear and compressive strengths, but not for tensile strength at different temperatures.

In this paper, the effect of temperature on load-displacement curves, stiffness, strength and failure mechanisms of woven fabric-reinforced PPS composites (C/PPS and GF/PPS) was studied. A new phenomenological model was proposed to evaluate the variation of bending properties with temperature for both thermoplastic composites, which also compared with several empirical models proposed in references.

Experimental

Two kinds of thermoplastic composites, 5-harness satin weave carbon fiber fabric (3K T300) and glass fiber fabric (7781 E-Glass) PPS-based composites, were studied in this paper. The lay-up of both PPS-based composites was $[(0, 90)]_7$. The fiber volume fraction of the two kinds of composites was both 50%. The fiber mass fraction for both composites was 57 and 63%, respectively. In order to determine the transition temperature, the dynamic mechanical analysis (DMA) of both composites was performed. The variation of dynamic stiffness E^* and the mechanical loss factor $\tan\delta$ with temperature from 23 to 350 °C can be obtained by using three-point bending mode of C/PPS and GF/PPS specimens with a DMA Q800 (TA Instruments), where heating rate and testing frequency were 3 °C/min and 1 Hz. The specimen dimension was 50 mm \times 5 mm \times 2.54 mm. In order to study the flexural behavior of both thermoplastic composites at different temperatures, three-point bending static tests according to ASTM D709-07 were conducted. The loading point for test was in the center, and the span-to-depth ratio was 16. Flexural behavior of two kinds of PPS-based composites at six temperature points 25 , 60 , 90 , 120 , 150 and 200 °C was conducted on an electromechanical tensile machine (INSTRON 3369) with a temperature chamber. Five samples were tested at each temperature. A heating

rate to reach the requested temperature was approximately 4 °C/min, and a temperature plateau was hold during 15 min before running the test in order to obtain a homogeneous temperature inside the specimen. The loading speed was 1 mm/min. According to the standard ASTM D709-07, the bending stiffness E and bending strength σ of the thermoplastic composites were derived from the classical beam theory as follows:

$$E = l^3 \Delta p / (4bh^3 \Delta f) \quad (1)$$

$$\sigma = 3pl / (2bh^2) \quad (2)$$

where p was the maximum bending load, Δp was the loading increment, Δf was the deflection increment, l was the span length, b and h were the sample width and thickness, respectively.

Mathematical models of material properties and temperature

In order to ensure the reliability of design of thermoplastic components, it was necessary to describe and accurately model the thermo-mechanical behavior of thermoplastic composites over a broad temperature range. Several thermo-mechanical models [11–22] for fiber-reinforced polymer materials were developed as listed in Table 1. These models mainly predicted the thermo-mechanical behavior of materials by establishing a correlation between key points (such as glass transition temperature and decomposition temperature) and material properties.

DMA experiment can obtain the thermo-mechanical properties of thermoplastic materials. The storage stiffness (E') and mechanical loss factor $\tan\delta$ curves were recorded as a function of temperature (T) as seen in Fig. 1. Based on the loss factor $\tan\delta$ peaks, the glass transition temperature (T_g) and the melting temperature (T_m) of both thermoplastic composites were about 123 and 290 °C, 109 and 290 °C, respectively. Over the experimental temperature range, DMA curves of both thermoplastic composites had similar trends. The storage stiffness was approximately constant in a temperature range between 25 and 75 °C. When temperature was in the glass transition region (from 75 to 150 °C), the storage stiffness decreased significantly to about 60% of the ambient temperature stiffness. When the temperature increases to 250 °C, the storage stiffness was once again plummeted to about zero.

The change of stiffness in dynamic experimental analysis related to change of polymer matrix or fiber/matrix interface because the stiffness of reinforcing fibers was insensitive over the studied temperature range. Thus, the measured properties were sensitive to properties of matrix [22]. With the increase in the test temperature, PPS matrix began to soften and had an attenuation of storage modulus. The changes in storage modulus can also be regarded as a reduction process of matrix performance. Considering the effect of matrix on properties of thermoplastic composite, especially the matrix-dominated composites [3, 7–9], it is asserted that the storage modulus could provide a useful indicator for

Table 1 Description of several thermo-mechanical models

Researchers	Formula	Model parameters
Ha/Springer	$P(T) = P_{T_0} \left(\frac{T_E - T}{T_E - T_0} \right)^n$	T_E is the temperature at which the properties tends to zero; n is a power law index
Gibson et al.	$P(T) = P_{T_0} - \frac{P_{T_0} - P_{T_r}}{2} \times (1 + \tanh[k(T - T_{g,mech})])$	$T_{g,mech}$ is the temperature at which the properties falls most rapidly; k is a constant related to the sharpness of the transition
Mahienux et al.	$P(T) = (P_g - P_r) \times \exp[-(T/T_g)^m] + P_r \times \exp[-(T/T_d)^n]$	m, n are Weibull exponents
Keller/Bai et al.	$P(T) = P_g \times [1 - \alpha_g(T)] + P_r \times \alpha_g(T) \times [1 - \alpha_d(T)] + P_d \times \alpha_g(T) \times \alpha_d(T)$	α_g, α_d are the conversion degrees of different states
Bosze et al.	$P(T) = P_{T_0} \left[\frac{E'(T)}{E'_{T_0}} \right]$	E', E'_{T_0} are the storage modulus at T and reference temperature

T is the temperature; $P(T)$ is the material properties at temperature T . T_0 is the ambient temperature; P_{T_0} is the material properties at ambient temperature; P_{T_r} is the material properties at the reference temperature T_r ; P_g, P_l and P_d are the material properties in the glassy, leathery and decomposed states, respectively

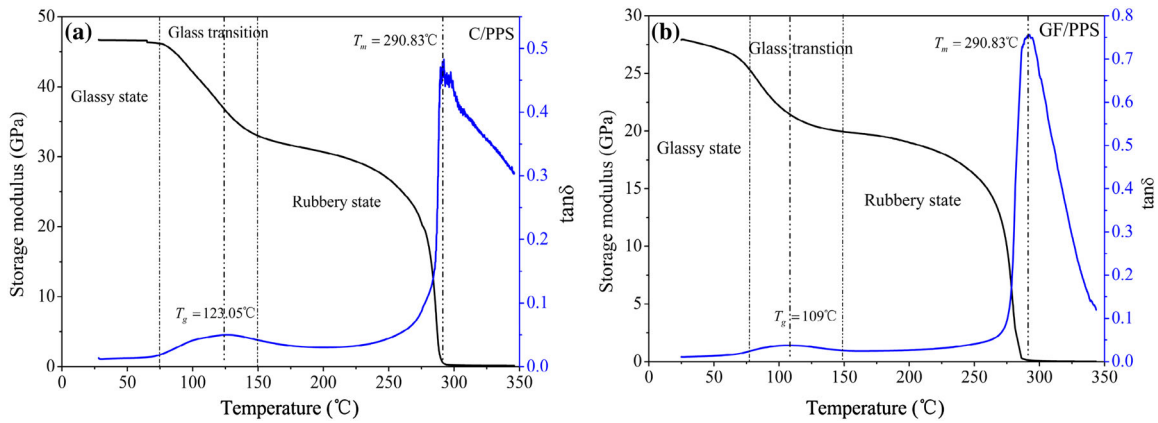


Figure 1 Variation of storage stiffness and $\tan\delta$ with temperature, **a** C/PPS and **b** GF/PPS.

temperature-dependent mechanical properties of thermoplastic composites. A new model based on a correlation between dynamic tests and mechanical properties was established to model the thermo-mechanical behavior of thermoplastic composites at different temperatures.

In a DMA experiment, the measured stiffness (E^*) in dynamic experimental analysis can be expressed by the storage modulus and the loss modulus the storage modulus (E') and the loss modulus (E'') as: $E^* = E' + iE''$, considering the damping factor ($\tan\delta = E''/E'$) was small ($\tan\delta < 0.1$, $E' \geq E''$, as shown in Fig. 1), E'' can be neglected, yielding $E^* \cong E'$. Thus, the degree of attenuation of the storage modulus can be used to calculate the material properties as a function of temperature reduction, and the model presented in this paper can be expressed as:

$$P(T) = P_{T_0} \frac{E'(T) - E'(T_r)}{E'_{T_0} - E'_{T_r}} + P_{T_r} \frac{E'_{T_0} - E'(T)}{E'_{T_0} - E'_{T_r}}$$

where, $P(T)$ was the mechanical properties of composite at temperature T , P_{T_0} was the mechanical properties at ambient temperature, P_{T_r} was the mechanical properties at another reference temperature T_r . $E'(T)$ was storage stiffness at temperature T , E'_{T_0} and E'_{T_r} were storage stiffness at reference temperature T_0 and T_r .

Results and discussion

The effect of temperature on flexural properties

Load–displacement curves as seen in Fig. 2 reflected different bending characteristics of thermoplastic composites with the increase in temperature. Below the glass transition temperature, all the curves linearly increased at the initial stage, but with different slopes. Both composites exhibited nonlinearity only

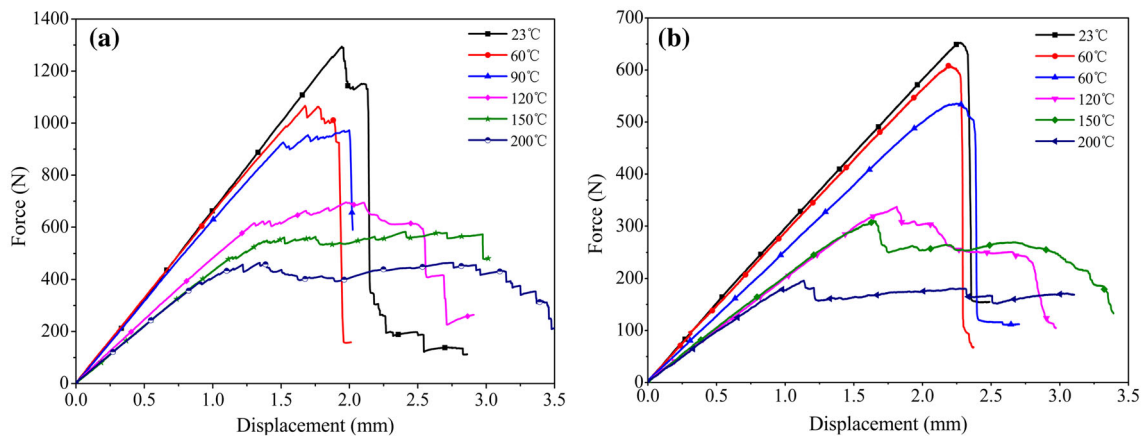


Figure 2 Bending curves of the composite at different temperatures, **a** C/PPS and **b** GF/PPS.

at the end of tests and the load suddenly dropped after reaching a peak load, which were consistent with those from other Refs. [23, 24]. When the temperature increased to the glass transition temperature, the thermoplastic composites exhibited ductile behavior. There exists a long zigzag aspect and a smoother drop of bending load for load–displacement curve. The above phenomenon indicated that there existed different damage mechanisms at below and above the glass transition temperature.

Bending stiffness and strength of both thermoplastic composites at different temperatures are given in Table 2. The bending stiffness and strength were more stable in the glassy region, but dropped very sharply after temperature entered the glass transition region. Then, an attenuation tended to be gentle in the rubbery region. These reflected typical characteristics of semi-crystalline polymer composites. Figure 3 demonstrates a striking correlation between the temperature dependence of storage modulus and the normalized bending properties for both thermoplastic composites, which also reflected the feasibility of the proposed model.

Figures 4, 5, 6 and 7 show a comparison of measured values (bending properties, strengths) of both thermoplastic composites with several mathematical models. Parameters of each model were estimated using a standard procedure that minimizes the mean square errors for test data. Table 3 presents the parameters for different models and corresponding average value of the mean absolute percentage error (MAPE). Results presented in Figs. 4 and 5 show that all models could predict accurately the stiffness of two thermoplastic composites. Those simulation curves were able to replicate a variation pattern of experimental data presenting mean absolute percentage errors (MAPE) which varied between 0.53 and 9.74% for stiffness of C/PPS, 0.83 and 5.01% for stiffness of GF/PPS, respectively. However, some models could not accurately predict the strength of both thermoplastic composites. In Figs. 6 and 7, experimental data of strength were a hyperbolic trend, while Ha-Springer's model was a monotonic function. Ha-Springer approach was not suitable for whole temperature range but only for the region in the same phase before/after T_g temperature by

Table 2 Bending stiffness and bending strength of the composites at different temperature

Temperature (°C)	C/PPS		GF/PPS	
	Stiffness (GPa)	Strength (MPa)	Stiffness (GPa)	Strength (MPa)
23	51.89 ± 0.09	945.74 ± 7.83	27.13 ± 0.11	526.52 ± 8.17
60	50.72 ± 0.28	901.78 ± 12.14	25.66 ± 0.19	490.22 ± 10.09
90	47.84 ± 0.62	789.97 ± 17.63	22.92 ± 0.48	424.47 ± 15.93
120	38.92 ± 0.57	555.25 ± 15.39	20.18 ± 0.43	297.21 ± 14.31
150	32.84 ± 0.32	386.87 ± 13.17	18.95 ± 0.37	253.27 ± 9.94
200	30.05 ± 0.53	298.46 ± 9.42	17.64 ± 0.25	204.46 ± 8.62

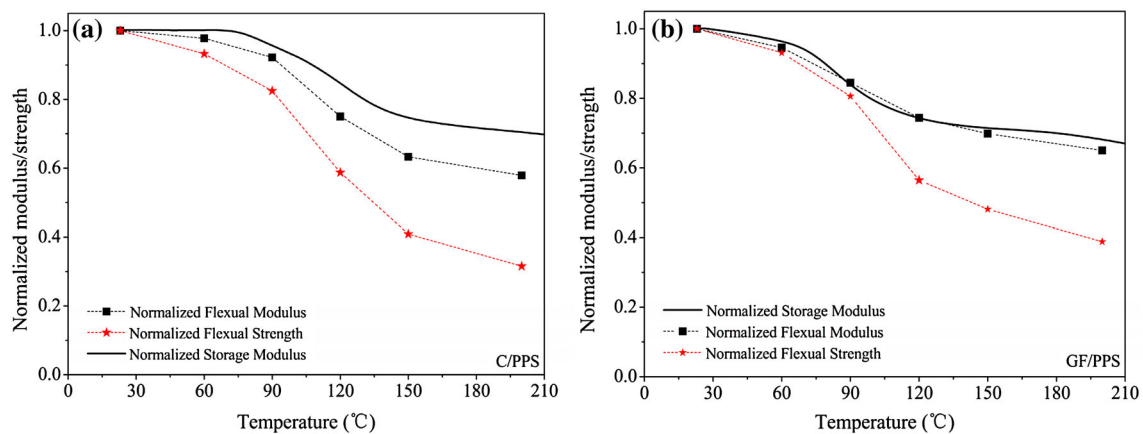


Figure 3 Normalized storage stiffness and bending stiffness/strength at different temperatures: **a** C/PPS and **b** GF/PPS.

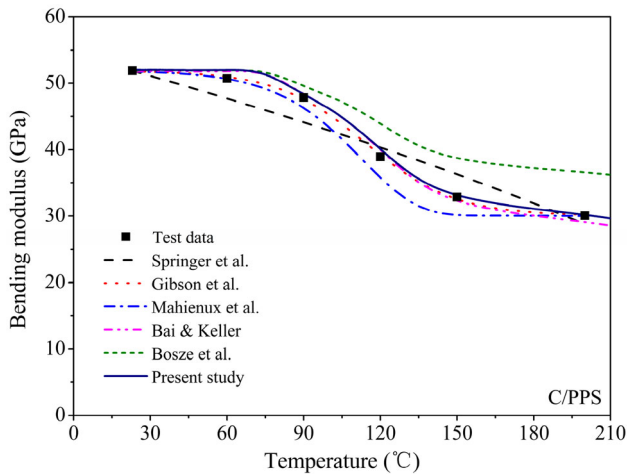


Figure 4 Bending stiffness of C/PPS versus temperature: experimental data and predicted curves.

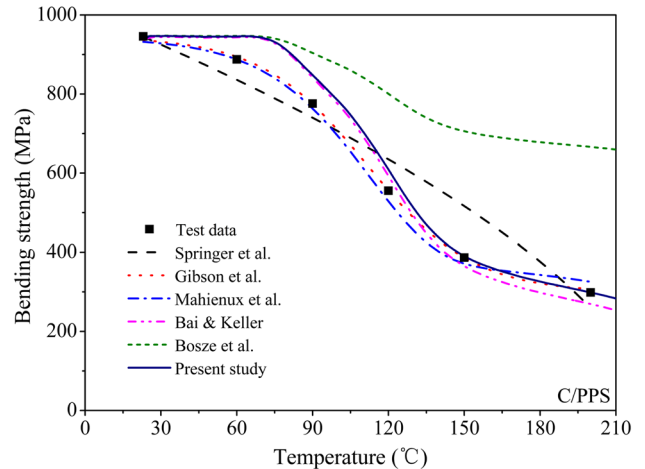


Figure 6 Bending strength of C/PPS versus temperature: experimental data and predicted curves.

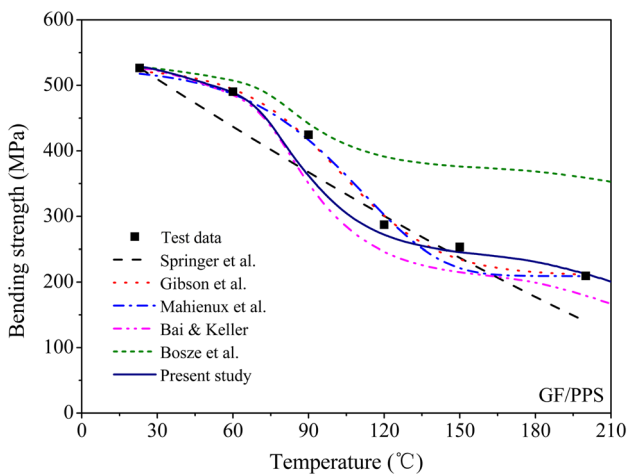


Figure 5 Bending stiffness of GF/PPS versus temperature: experimental data and predicted curves.

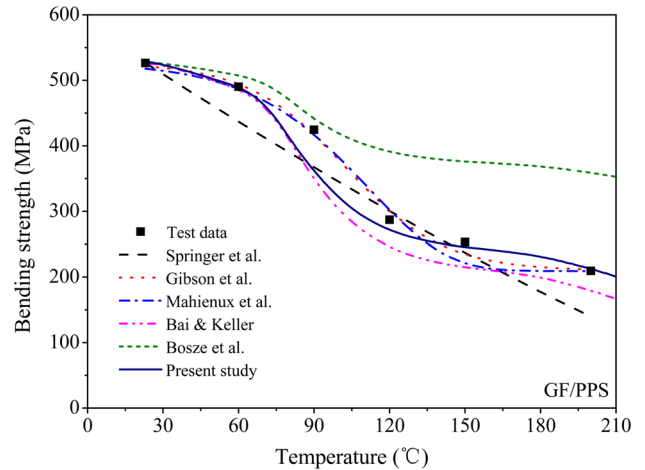


Figure 7 Bending strength of GF/PPS versus temperature: experimental data and predicted curves.

choosing suitable parameter n [25]. In the present study, the variety of mechanical properties with temperature was similar to that of DMA results, but not exactly consistent, as shown in Fig. 3. Thus, the prediction accuracy of Bosze’s model was relatively lower (MAPE = 45.48% for C/PPS and MAPE = 27.26% for GF/PPS). For the model of Bai and Keller, the mean absolute percentage errors in the estimation of the strength of both thermoplastic composites were 6.26 and 10.69%, respectively. The prediction accuracy of Bai and Keller’s model was greatly influenced by the selected reference temperature. With the increasing temperature, the attenuation of composites stiffness in the same state was not same with strength. Therefore, the selection of

reference temperature became critical. A similar situation occurred on Mahienux’s model. For Gibson’s model, simulation curve was hyperbolic, which was able to satisfy description on the temperature dependence of mechanical properties of thermoplastic composites for all temperature range. The accuracy of the model prediction was quite accurate (MAPE <3.0%). But the determination of parameters in Gibson’s model was more complex.

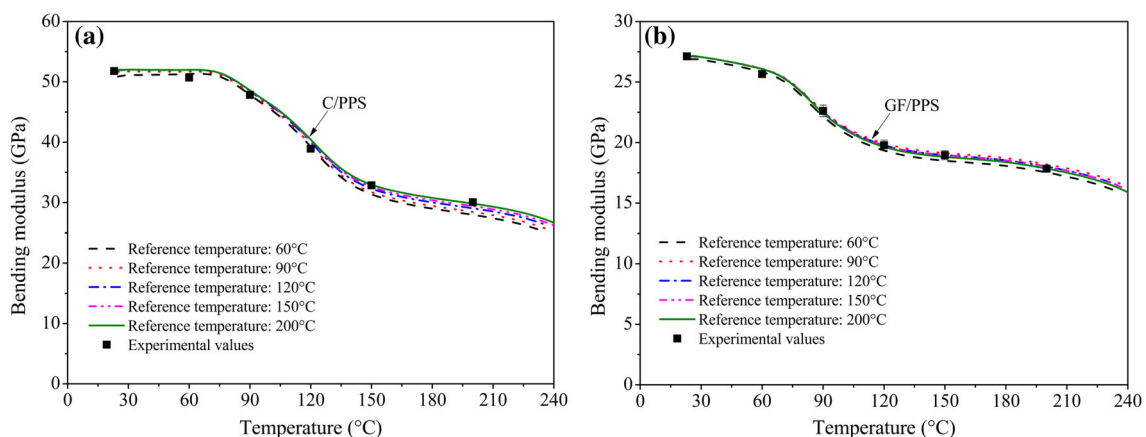
Selecting 200 °C as reference temperature, the proposed model in the present study could accurately predict bending stiffness (MAPE = 1.12% for C/PPS and MAPE = 0.83% for GF/PPS) and strength (MAPE = 4.45% for C/PPS and MAPE = 4.08% for GF/PPS). It should be noted that

Table 3 Simulation of thermoplastic composites mechanical properties—parameter estimation and mean absolute percentage error (MAPE) for different models

Model	Parameter	Stiffness (C/PPS)	Strength (C/PPS)	Stiffness (GF/PPS)	Strength (GF/PPS)
Springer et al. Eq. (1)	T_r	300.00	223.15	300.00	300.03
	n	0.58	0.6	0.53	1.30
	MAPE (%)	5.37	11.97	5.01	11.67
Gibson et al. Eq. (2)	$T_{g,mech}$	111.35	114.32	103.32	95.80
	k'	0.024	0.028	0.025	0.023
	MAPE (%)	0.53	1.18	1.28	2.97
Mahienux et al. Eq. (3)	m	18.94	14.36	10.64	13.33
	n	30	30	30	30
	MAPE (%)	3.21	3.29	2.54	3.72
Keller and Bai Eq. (4)	MAPE (%)	1.89	6.26	3.32	10.69
Bosze et al. Eq. (5)	MAPE (%)	9.74	45.48	1.69	27.26
Present study	MAPE (%)	1.12	4.45	0.83	4.08

the effect of reinforcing fiber at temperatures below the glass transition temperature of composite affected the prediction accuracy of the model. When temperature exceeded T_g , accuracy of the model were obviously improved (MAPE = 2.15% for C/PPS and MAPE = 2.68% for GF/PPS). Compared with other empirical models, the proposed model did not need to determine the model parameters; the reference temperature of the model was not limited to several special reference points. To identify a suitable range for the reference temperature selection, the five temperature points (60, 90, 120, 150 and 200 °C) were used as reference temperature in the paper. Figures 8 and 9 depict the prediction of mechanical properties versus temperature of the two thermoplastic

composites based on different reference temperatures. The results in Fig. 8 showed that the effect of reference temperature on predicted bending stiffness in the model is slight and can be neglected. However, the strength-temperature curves in Fig. 9 illustrated that based on the reference temperature 60 and 90 °C the predicted strength of the proposed model were very different from the experimental results, while at the other three reference temperatures the predicted strengths were in good agreement with the experimental data. Thus, in order to accurately predict the modulus and strength of both composites, the reference temperature range for the proposed model should be in the rubbery region ($>T_g$) of the composites.

**Figure 8** Bending stiffness of both thermoplastic composites versus temperature: experimental data and predicted curves based on different temperatures.

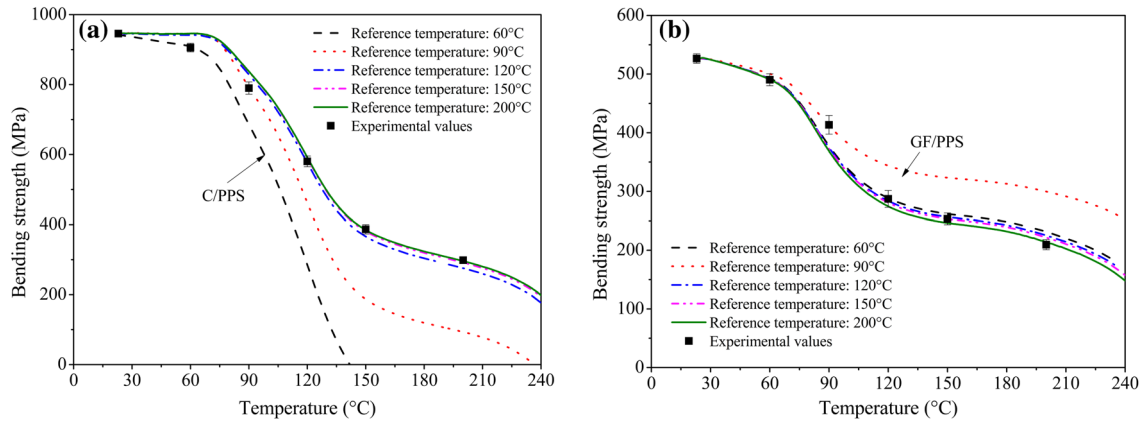


Figure 9 Bending strength of both thermoplastic composites versus temperature: experimental data and predicted curves based on different temperatures.

Figure 10 Failure morphologies of C/PPS bending specimens at different temperatures.

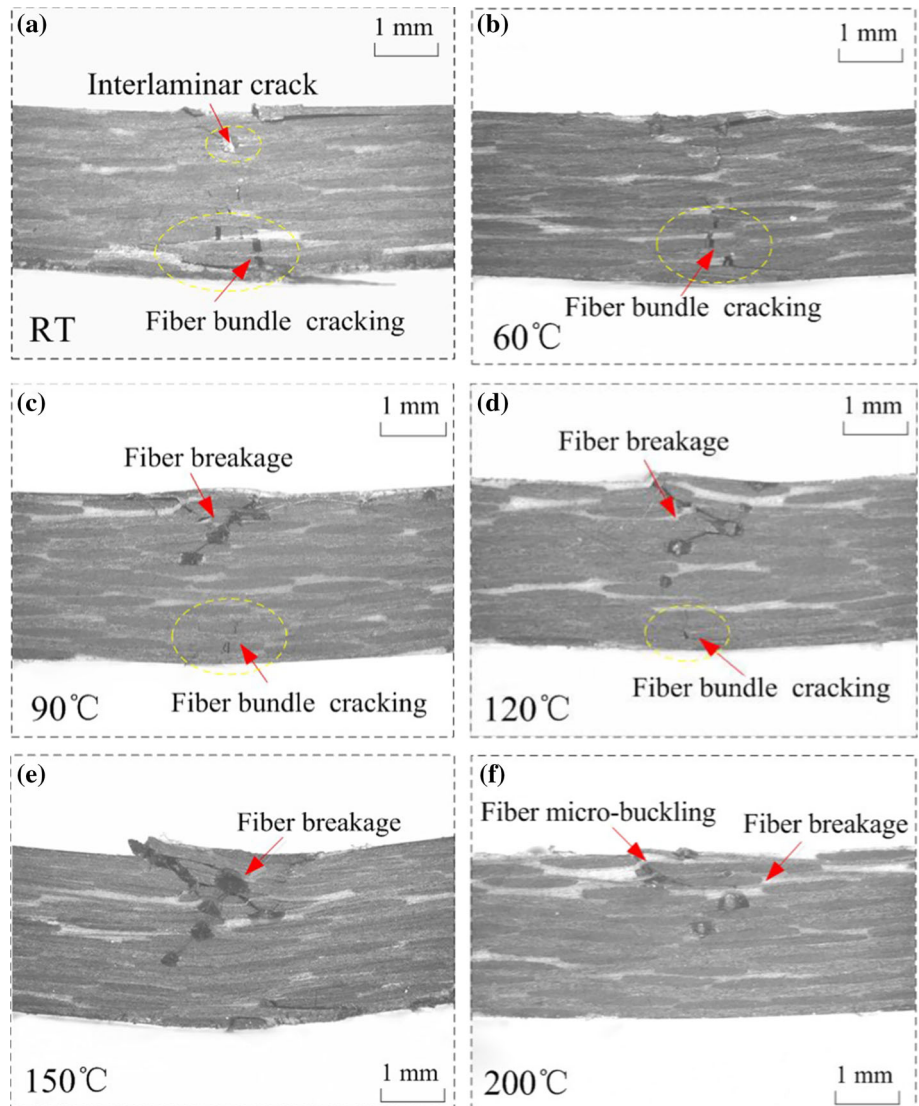
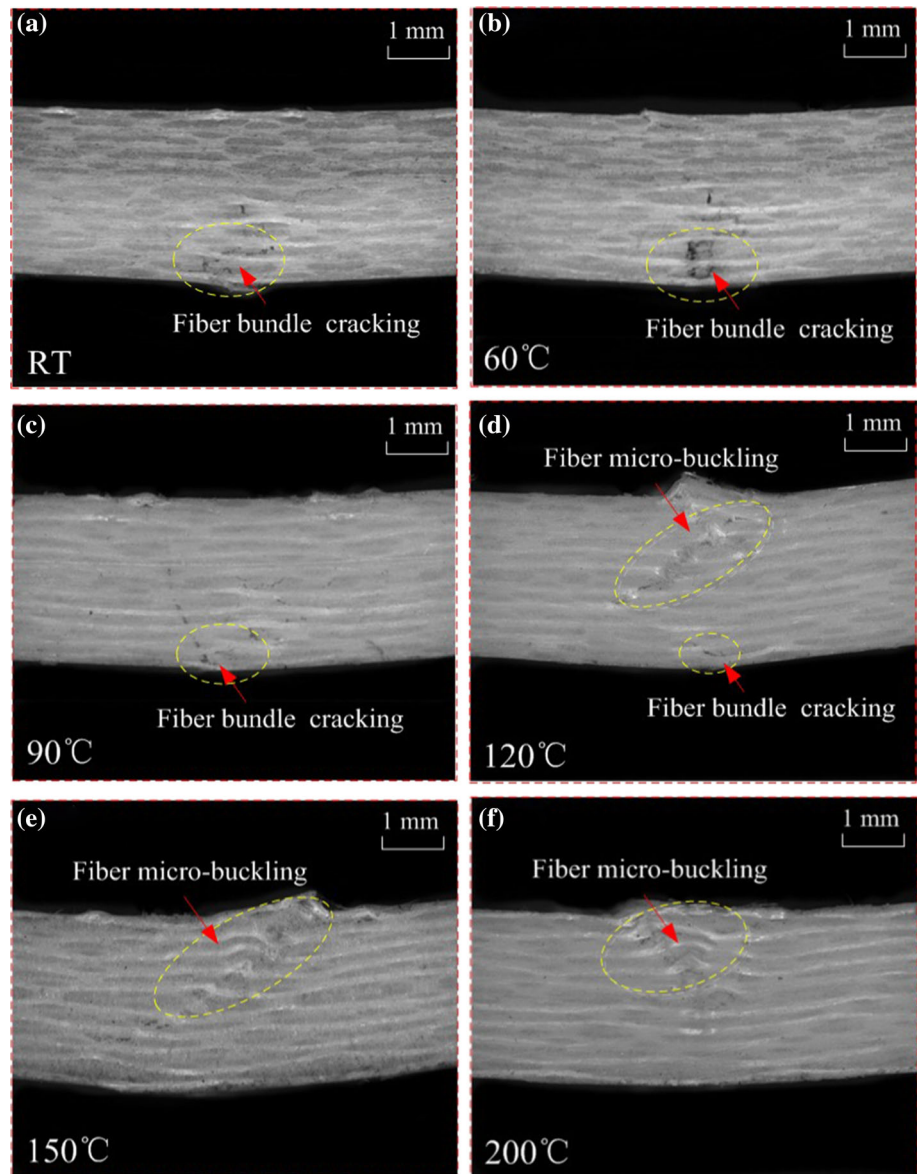


Figure 11 Failure morphologies of GF/PPS bending specimens at different temperatures.



Temperature-dependent failure mechanisms

Mechanical behaviors of PPS matrix are greatly related with the temperature. When the temperature exceeds the glass transition temperature, the mechanical properties of the PPS matrix will change from brittle to ductile. The flexural failure mechanisms of both thermoplastic composites at high temperature will be greatly associated with the mechanical behavior of PPS matrix. Typical pictures of bending failure of both PPS-based composites at different temperatures are shown in Figs. 10 and 11, respectively. For bending specimen, the upper and lower surfaces were in compressive and tensile stress

states. When bending load reached a certain extent, damage occurred firstly on the compression surface with plastic deformation or cracking of resin matrix, as well as bending deformation of fibers. When load increased, the matrix at the tensile surface of specimen was also damaged, the fiber happened brittle tensile fracture. As shown in Figs. 10a–c and 11a–c, at the glassy state, the cracks in fiber bundles were distributed across the specimen section, a few inter-layer cracks could be found near the upper and lower surface of specimens. Thus, the bending strength of composites was controlled by fibers tensile and compressive failure. The zigzag aspect in force–displacement curve can be attributed to the sequential

fiber transverse cracking and fiber compressive failure. The sudden drop in load was a consequence of fiber bundle tensile failure. From Figs. 10d–f and 11d–f, at around and above the glass transition temperature of PPS-based composites, the crack in the fiber bundles near the lower surface was disappeared and the fracture/micro-buckling of fibers at the upper surface were happened significantly. The enhanced plasticization of matrix affected the transfer of load between plies and dissipated a part of transmitting energy. This plasticity mechanism prevented other failure mechanisms and the compression failure occurred in the contact area finally. Consequently, the bending strength was controlled by the fiber compressive failure. The longer zigzag aspect of force–displacement curves for the two PPS-based composites can be attributed to the fiber micro-buckling. By analyzing the fracture morphology, Figs. 5 and 6 reflected the transition of bending failure mechanisms from brittle to ductile fracture behavior with increasing temperature.

Conclusion

The influence of temperature on bending behavior of two kinds of PPS composites (carbon and glass fiber woven fabric reinforcements) from ambient temperature up to 200 °C was studied in this paper. The glass transitions of two thermoplastic composites were characterized by dynamic mechanical analysis. There existed four different temperature-dependent material states, such as glassy, leathery, rubbery and decomposed states. Two kinds of composites both can retain about 60% of room temperature stiffness at rubbery state. Bending properties of thermoplastic composite decreased with the increase in temperature. The failure modes of both composites transited from brittle to ductile fracture with the increase in temperature. The variation with bending behavior of both thermoplastic composites with temperature was evaluated by some empirical models. A new simple and stable phenomenological model was also proposed to accurately characterize the variation of stiffness and strength behavior of both thermoplastic composites, which was also verified by the experimental results.

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