

General Model of Temperature-dependent Modulus and Yield Strength of Thermoplastic Polymers

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Electronic Supplementary Information

Abstract A general model was developed to predict the temperature-dependent modulus and yield strength of different thermoplastic polymers. This model, which depends on only two parameters with clear and specific physical meanings, can describe the temperature-dependent modulus and yield strength of thermoplastic polymers over the full glass transition region. The temperature-dependent modulus and yield strength of three thermoplastic polymers were measured by uniaxial tension tests over a temperature range of 243–383 K. The predictions showed excellent agreement with the experimental data. Sensitivity analysis of model input parameters showed negligible effect on the present general model. The universality of the present general model was further validated, showing excellent agreement with published experimental data on other thermoplastic polymers and their composites.

Keywords Thermoplastics; Modulus; Yield strength; Modeling; Glass transition

Citation: Huang, P. Y.; Guo, Z. S.; Feng, J. M. General model of temperature-dependent modulus and yield strength of thermoplastic polymers. *Chinese J. Polym. Sci.* 2020, 38, 382–393.

INTRODUCTION

Thermoplastic polymers have unique thermal, mechanical, electrical, and lightweight characteristics that make them suitable for use in a wide variety of applications in aviation,^[1] automotive,^[2] battery,^[3] biomedicine, and construction sectors.^[4] In practical applications, thermoplastic polymers are subjected to a wide range of environment and temperature conditions. However, the mechanical properties of thermoplastic polymers, such as modulus and strength, can drop by three orders of magnitude at temperatures near the glass transition region,^[5] causing them to become too soft to resist external loads. Therefore, an accurate description of their mechanical properties at different temperatures is fundamental for the successful design of engineering structures.

Many models have been developed to describe the temperature-dependent mechanical property of polymeric materials in the glass transition region.^[6–18] Gu and Asaro employed a phenomenological form to describe the relationship between temperature and the Young's modulus of polymer matrix composites.^[6] Ha and Springer present a power law expression to model the temperature-dependent modulus and strength of thermoset resins and their composites.^[7] Dutta and Hui developed a time- and temperature-depend-

ent model of the modulus of glass-fibre-reinforced polyester composites.^[8] By neglecting the loss modulus, Wang *et al.* calculated the reduction in material properties using the degree of attenuation of the storage modulus, thus establishing a simple and stable phenomenological model to predict the bending stiffness and strength of two kinds of thermoplastic composites.^[9] Murayama and Bell established a temperature-dependent model to characterize the dynamic modulus of an amine-cured epoxy polymer in the rubbery state.^[10] In order to describe the dynamic storage modulus of three thermoplastic polymers and one composite over the full range of transition temperatures, Mahieux and Reifsnider presented a Weibull-type function based on the breakage of bonds between the macromolecules.^[11,12] Gibson *et al.* developed a semi-empirical model based on the hyperbolic tanh function to characterize the mechanical properties of thermoset composites at different temperatures.^[13,14] Based on the Arrhenius equation, Bai *et al.* presented a model to predict the temperature-dependent dynamic storage modulus of fibre-reinforced polymer composites across different temperature ranges.^[15] Guo *et al.* presented a temperature- and frequency-dependent model of dynamic mechanical properties that displayed excellent agreement with the dynamic storage modulus and flexural modulus of a thermoset composite.^[16–18] The Ree-Eyring's model^[19–21] considers the yield behaviour as a thermally activated process and has been employed to predict the temperature- and rate-dependent yield strength of aliphatic polyketone,^[22] plasticised starch-polyol,^[23] as well as

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Received July 14, 2019; Accepted September 30, 2019; Published online December 6, 2019

polyamide 6 (PA6) and its nanocomposites.^[24] Considering a constant strain rate, a linear relationship was suggested by Kambour who simplified the Ree-Eyring's model.^[25] This relationship has been used for thermoplastic polymers such as PA6 at a small temperature range (from 277 K to 353 K).^[26] Unfortunately, this relationship loses accuracy at a wide temperature range. Most of these previous studies only focused on modelling the dynamic storage modulus or the yield strength of polymers and their composites, whereas the literature contains far fewer reports on temperature-dependent modulus and yield strength model of thermoplastic polymers.

This article presents a general model of the temperature-dependent modulus and yield strength of thermoplastic polymers. The physical meanings of the model parameters are discussed in detail. Input parameters are used in the present model: the modulus and yield strength of the specimen in the glassy and rubbery states, determined by uniaxial tension tests, and the glass transition temperature, determined by dynamic mechanical analysis (DMA). The present model was verified using the Young's modulus and yield strength of three types of thermoplastic polymer measured by uniaxial tension tests under different temperatures, and by comparison with experimental data published in the literature. The experimental results were adopted for calibrating the fitting parameters of the present model. Sensitivity analysis showed that our model was robust, with variations in input parameters having only a slight effect on the model. The present model is discussed in comparison with existing popular models.

TEMPERATURE-DEPENDENT MODULUS MODELS

According to the linear relationship between modulus and yield strength (Eq. S1 in the electronic supplementary information, ESI) and the definition of degree of glass transition a_g ,^[15] which is widely adopted by many other literatures,^[16–18,27–36] a_g can be written as:

$$a_g(T) = \frac{P_U - P(T)}{P_U - P_R} \quad (1)$$

where P_U is the mechanical property (modulus and yield strength) of the specimen in the glassy state, P_R is the mechanical property of the specimen in the rubbery state, and $P(T)$ is the instantaneous mechanical property.

In statistical mechanics, the glass transition can be considered as a process in which a large population of molecules change progressively from a glassy to a rubbery state.^[19] If N is assumed to be the total number of molecules, then the number of rubbery-state molecules at temperature T can be expressed as $N \cdot a_g(T)$. The number of molecules changing to the rubbery-state per unit temperature is defined as $\frac{d(N \cdot a_g)}{dT} / (N \cdot a_g)$.

In most thermoplastic polymers,^[37–39] the shape of the modulus-temperature curve is a reversed sigmoid, which is similar to the generalized logistic function, also known as Richards' curve.^[40] Based on this, we hypothesize that $\frac{d(N \cdot a_g)}{dT} / (N \cdot a_g)$ decreases as the total number of rubbery-state molecules increases, as shown in Eq. (2):

$$\frac{d(N \cdot a_g)}{dT} / (N \cdot a_g) = k - A \cdot (N \cdot a_g)^m \quad (2)$$

where k is the initial increase in the number of rubbery-state molecules per unit temperature, m is a shape factor, and A is a constant. From Eq. (2), the number of molecules changing to the rubbery-state per unit temperature becomes k as a_g approaches zero, and zero as a_g approaches 1. The constant A is equal to k/N^m . Thus, the variation of a_g per unit temperature can be written as:

$$\frac{da_g}{dT} = k \cdot (1 - a_g^m) \cdot a_g \quad (3)$$

When the temperature approaches the glass transition temperature T_g , the maximum increase of a_g per unit temperature occurs. The derivative of da_g/dT equals zero, and thus we can calculate a_g when $T = T_g$:

$$a_g|_{T=T_g} = \left(\frac{1}{m+1} \right)^{\frac{1}{m}} \quad (4)$$

Based on Eq. (4), the solution to differential Eq. (3) can be expressed as:

$$a_g(T) = \frac{1}{(me^{-mk(T-T_g)} + 1)^{\frac{1}{m}}} \quad (5)$$

Substituting Eq. (5) into Eq. (1), the temperature-dependent mechanical property $P(T)$ can be determined by Eq. (6):

$$P(T) = P_U - \frac{P_U - P_R}{(me^{-mk(T-T_g)} + 1)^{\frac{1}{m}}} \quad (6)$$

Eq. (6) therefore provides the basis of the general temperature-dependent model developed in this study. In this model, the input parameters P_U and P_R can be determined by uniaxial tension tests, and T_g by DMA. Further discussion of k and m follows.

The parameter k represents the initial increase in the number of rubbery-state molecules per unit temperature. Moreover, it determines the width of the glass transition region. With regard to the degree of glass transition, when $a_g \leq 0.01$, the material is in the glassy state, and when $a_g \geq 0.99$, the material is in the rubbery state. Then, the width of glass transition region ΔT ($0.01 \leq a_g \leq 0.99$) can be represented as const/k for a given value of shape factor m , based on Eq. (5). Given the inverse-proportional relationship between ΔT and k , the smaller the value of k , the wider the glass transition region. In the glass transition region, the low molecular weight chains relax earlier than those with high molecular weight due to their higher chain mobility.^[41] Therefore, a polymer with a broad (narrow) molecular weight distribution has a wide (narrow) glass transition region. Overall, the parameter k is related to the polydispersity index ($\mathcal{D}$), a measure of the distribution of molecular weights in a given polymer. More concretely, the higher (lower) the $\mathcal{D}$ of the polymer, the smaller (larger) the initial increase in the number of rubbery-state molecules per unit temperature.

The second parameter m is a shape factor that determines the symmetry of the mechanical property-temperature curve. Eq. (2) with different values of m are plotted in Fig. 1. When $m = 1$, a linear decrease in the correlation $\frac{d(N \cdot a_g)}{dT} / (N \cdot a_g)$ is found as the number of rubbery-state molecules increases;

the model can be reduced to our previous model.^[16] Under this condition, the mechanical property-temperature curve shows central symmetry with respect to the point $(T_g, \frac{P_U + P_R}{2})$ in the glass transition region, and $a_g = 1/2$ at T_g . When $m > 1$, the curve in Fig. 1 is convex (decreasing as the number of rubbery-state molecules increases), giving $a_g > 1/2$ at T_g . In contrast, when $0 < m < 1$, the curve in Fig. 1 is concave (decreasing as the number of rubbery-state molecules increases), giving $a_g < 1/2$ at T_g . In terms of geometry, the larger the value of m , the larger the value of a_g at T_g . In addition, in terms of the physical mechanism, the parameter m is connected with the crystallinity of the polymer. The results of Lin *et al.* suggest that the degree of crystallinity of a polymer remains constant below T_g , and then gradually decreases between T_g and the melting temperature,^[42] probably resulting from crystal melting. The larger the degree of crystallinity, the larger the transition from crystal phase to amorphous phase. Above T_g , the amorphous phase is in a rubbery state. Therefore, a polymer with a larger degree of crystallinity will exhibit a larger drop in mechanical property between the

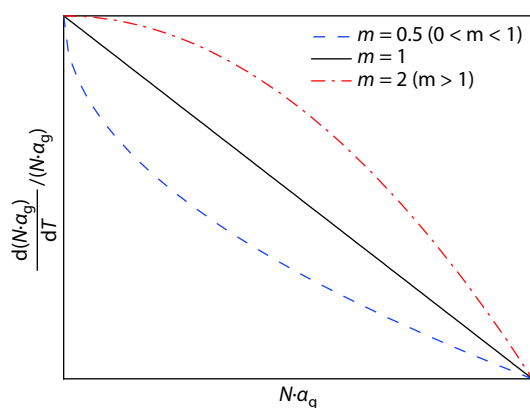


Fig. 1 The relationship between $\frac{d(N \cdot a_g)}{dT} / (N \cdot a_g)$ and $N \cdot a_g$ with different shape factors m .

glass transition point and the rubbery state, but a smaller drop in modulus between the glassy state and the glass transition point (smaller a_g at T_g). From this, it can be concluded that the parameter m in our model is related to the crystallinity of the polymer: the smaller the value of m , the larger the degree of crystallinity of the polymer.

Various established temperature-dependent modulus and yield strength models are summarized alongside our present model in Table 1. The model by Wang *et al.* is a phenomenological model, which is strongly dependent on the storage modulus.^[9] The models by Mahieux and Reifsnider,^[11,12] Ha and Springer,^[7] and Gibson *et al.*^[13,14] have one fitting parameter. Unfortunately, this fitting parameter has no legitimate physical meaning. For the model by Bai *et al.*,^[15] parameter a_g , which depends on three fitting parameters, must first be solved numerically, and T_g , a critical temperature in the glass transition region, is not included. The absence of T_g makes it inconvenient to analyze the glass transition region from the molecular level. It should be emphasized that our model, which has a concise form, is derived based on the changing trend of the increasing number of rubbery-state molecules per unit temperature, and includes the critical temperature T_g . The fitting parameters k and m related to $\mathcal{D}$ and crystallinity of the polymer, respectively, have specific physical meanings. It is also interesting that the model by Gibson *et al.* can be derived from our model when $k = 2k'$ and $m = 1$.^[16]

EXPERIMENTAL

Materials and Specimens

In this study, three kinds of thermoplastic polymer blends that are widely used in automotive trim systems were studied: acrylonitrile-butadiene-styrene (ABS), polycarbonate/acrylonitrile-butadiene-styrene (PC/ABS), and polypropylene/ethylene-propylene-diene-monomer/talc15 (PP/EPDM/T15), obtained from LG Chem (Korea), SABIC (Saudi Arabia), and PRET (China), respectively. The automotive is subjected to a temperature change ranging from 243 K to 383 K. Hence, three different

Table 1 Description of temperature-dependent modulus and yield strength models.

Model	Formula	Input parameter	Fitting parameter	Physical meaning
Wang <i>et al.</i>	$E(T) = E_{T_0} \frac{E'(T) - E'(T_r)}{E'_{T_0} - E'_{T_r}} + E_{T_r} \frac{E'_{T_0} - E'(T)}{E'_{T_0} - E'_{T_r}}$	$E_{T_0}, E'_{T_0}, E_{T_r}, E'_{T_r}$	None	E' is storage modulus.
Mahieux and Reifsnider	$E(T) = (E_g - E_r) \exp\left(-\left(\frac{T}{T_g}\right)^{m'}\right) + E_r$	E_g, E_r, T_g	m'	m' is Weibull exponent.
Bai <i>et al.</i>	$\begin{cases} E(T) = E_g(1 - a_g) + E_r a_g \\ \frac{da_g}{dT} = \frac{A_g}{\beta} \exp\left(\frac{-E_{A,g}}{RT}\right)(1 - a_g)^{n'} \end{cases}$	E_g, E_r	$A_g, E_{A,g}, n'$	A_g is a pre-exponential factor; $E_{A,g}$ is the activation energy; n' is the reaction order.
Ha and Springer	$P(T) = P(T_0) \cdot \left(\frac{T_r - T}{T_r - T_0}\right)^n$	$T_r, T_0, P(T_0)$	n	n is a constant without physical meaning.
Gibson <i>et al.</i>	$P(T) = \frac{P_U + P_R}{2} - \frac{P_U - P_R}{2} \tanh(k'(T - T_g))$	P_U, P_R, T_g	k'	k' is a constant related to the sharpness of the transition.
Present model	$P(T) = P_U - \frac{P_U - P_R}{(me^{-mk(T-T_g)} + 1)^{\frac{1}{m}}}$	P_U, P_R, T_g	k, m	k is related to $\mathcal{D}$; m is related to the crystallinity of the polymer.

T_0 is a temperature in the glassy state; T_r is a temperature in the rubbery state; E_g and E_r represent the modulus in glassy state and rubbery state, respectively.

polymers are tested with the same temperature range in the next section.

Uniaxial Tension Tests

The mechanical behaviours of the three thermoplastic polymers were studied at the same temperature range (243, 263, 296, 333, 353, and 383 K) by conducting uniaxial tension tests on an electromechanical tensile machine (Zwick/Roell Z020; maximum load capacity 20 kN) equipped with an environmental chamber. Dumbbell-shaped uniaxial tension specimens were prepared with approximate dimensions of 170 mm × 20 mm × 3 mm, in accordance with ISO 527.^[43] Prior to each test, the chamber was preheated to the designated temperature. Then, each specimen was placed in the chamber for 10 min to allow the temperature inside the specimen to equilibrate to the designated temperature (see Fig. 2a). Three specimens were tested at each temperature. The loading speed was set to 2 mm/min.

A non-contact measurement system was adopted to measure the strain by using a two-dimensional digital image correlation system. Two pairs of crossed lines were marked on each specimen in order to provide two intersection points with a distance of 50 mm (Fig. 2b). Adequate contrast was achieved using white LED lights to illuminate the specimen surface. A CCD camera (Basler acA1600, resolution 1626 × 1236 pixels) and Pylon Viewer software were employed to acquire 20 static images per second. During real-time strain

measurement, a correlation subset of 51 × 51 pixels was chosen to achieve sufficient spatial resolution.

Dynamic Mechanical Analysis

In order to characterize the glass transition temperatures (T_g) of the thermoplastic polymers, DMA was performed across a temperature range of 210 K to 393 K using a dynamic mechanical analyzer (DMA Q800, TA Instruments, USA). Rectangular

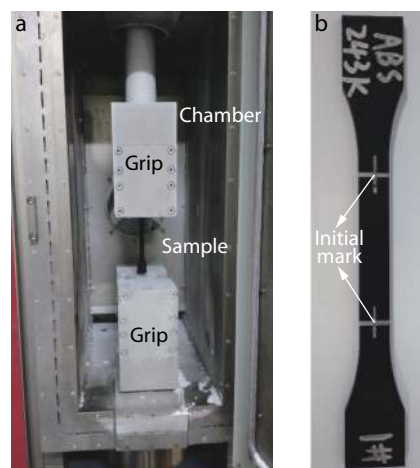


Fig. 2 Experimental setup for uniaxial tests: (a) Zwick/Roell Z020 testing machine and (b) a specimen.

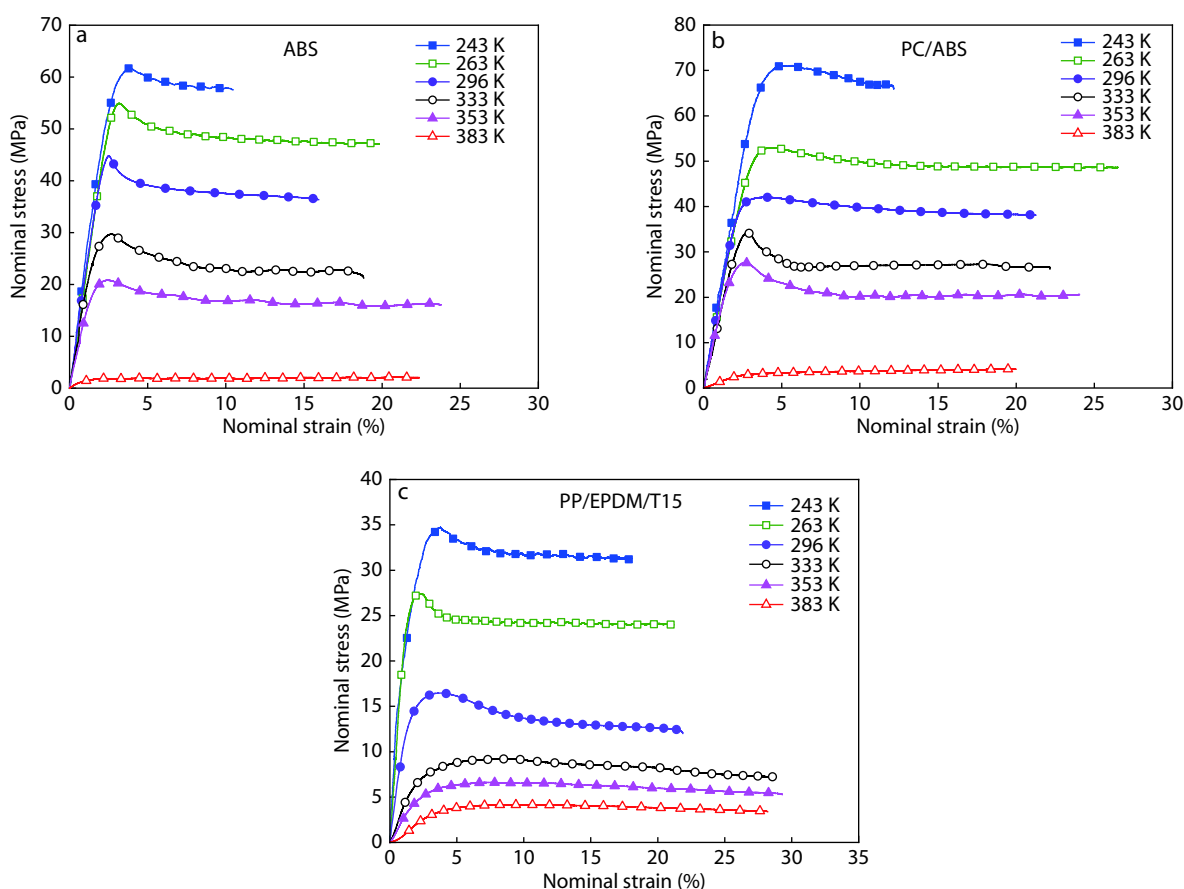


Fig. 3 Stress-strain curves of thermoplastic polymers at different temperatures: (a) ABS, (b) PC/ABS, and (c) PP/EPDM/T15.

specimens were prepared (approximate dimensions 10.2 mm × 6.8 mm × 0.75 mm) in accordance with the manufacturer's instructions. The storage modulus, loss modulus, and loss factor were obtained using multi-frequency-strain mode in tension, where the heating rate and testing frequency were 3 K/min and 1 Hz, respectively.

RESULTS AND DISCUSSION

Uniaxial Tension Tests

The stress-strain curves in Fig. 3 reflect different tensile characteristics of the three thermoplastic polymers with regard to temperature. The curves are plotted using engineering stress and strain. Fig. 3 clearly shows the temperature dependence of the mechanical properties of all three thermoplastic polymers. The five regimes of behavior are elasticity, yielding, strain softening, cold drawing, and failure. However, strain softening does not occur during high temperature measurements (383 K). Instead, the cold-drawing behavior (elongation at a constant

stress: appearing as a horizontal plateau) takes place immediately after yielding. For the materials without a yield plateau, nominal maximum stress^[44–47] and backward extrapolation^[47–49] are used to define the yield point (Fig. S1 in ESI). Young's modulus and yield strength (presented in Table 2), are calculated from the linear elastic portion ($\epsilon_{0.05\%}$ to $\epsilon_{0.25\%}$) of the stress-strain curve.^[43] As demonstrated in Table 2, the polymers are stiffer at lower temperatures and softer at higher temperatures.

Dynamic Mechanical Analysis

DMA experiments can reveal the thermo-mechanical properties of thermoplastic materials. The storage modulus E' , loss modulus E'' , and loss factor curves were recorded as a function of temperature, as seen in Fig. 4. The thermoplastic polymers presented high E' values, which gradually decreased with increasing temperature in the glassy state; a sharp drop occurred around T_g , corresponding to a dramatic increase of E'' , which resulted in an increase of the loss factor. T_g can be determined

Table 2 Average Young's modulus and yield strength of ABS, PC/ABS, and PP/EPDM/T15.

Temperature (K)	Young's modulus (GPa)			Yield strength (MPa)		
	ABS	PC/ABS	PP/EPDM/T15	ABS	PC/ABS	PP/EPDM/T15
243	2.325	2.145	2.830	60.88	71.08	34.52
263	2.183	1.962	2.417	53.93	52.25	28.14
296	2.134	1.941	1.220	44.58	41.27	16.57
333	1.759	1.481	0.409	28.72	33.69	9.25
353	1.352	1.277	0.285	20.92	27.39	6.56
383	0.175	0.136	0.172	1.75	3.36	4.12

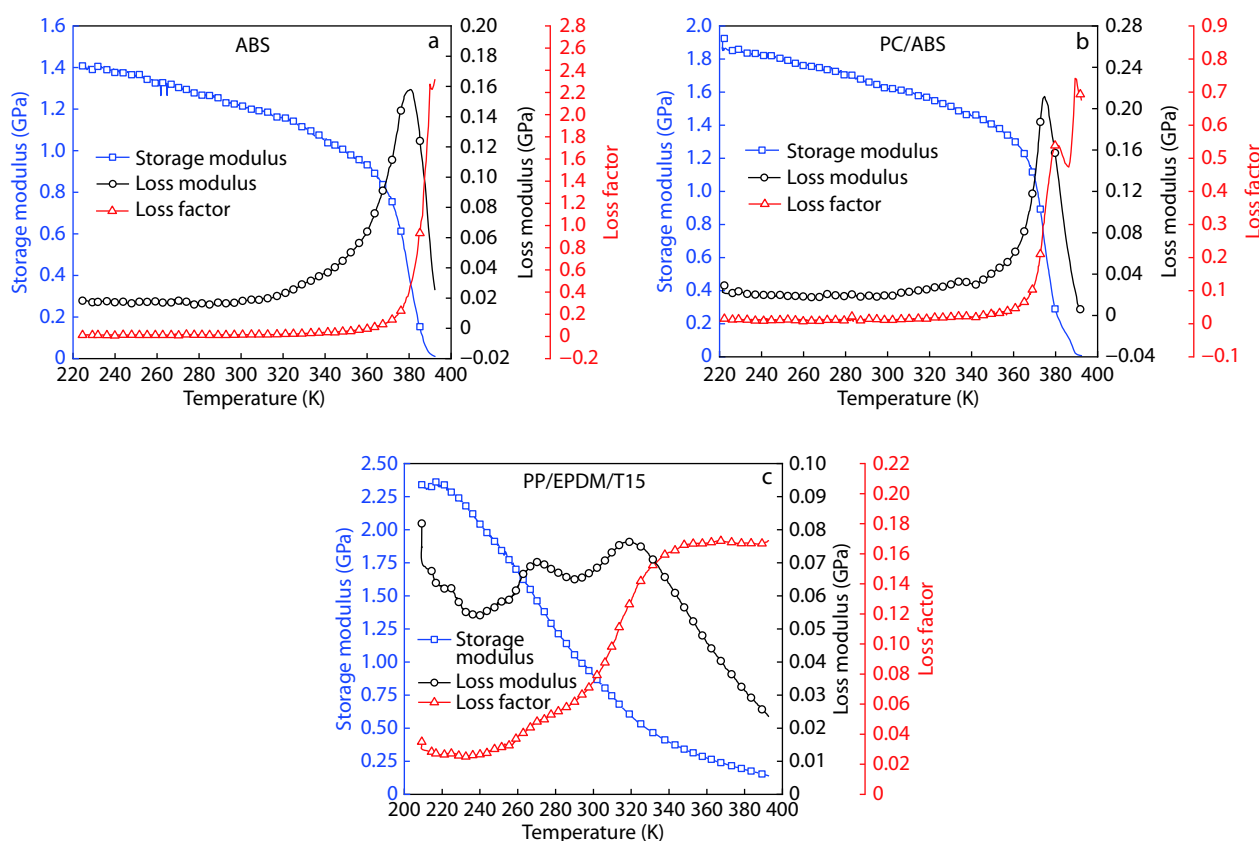


Fig. 4 Variation of storage modulus, loss modulus, and loss factor with temperature: (a) ABS, (b) PC/ABS, and (c) PP/EPDM/T15.

from the peak of the loss factor or loss modulus curves, or the mid-point of the storage modulus curve.^[50] In our model, T_g was obtained from the peak of the loss modulus for the following reasons. Firstly, the temperature corresponding to the peak of loss modulus typically coincides with the temperature of maximum change in storage modulus,^[16–18,51] and the trend of storage modulus is similar to that of Young's modulus; thus, the maximum change of Young's modulus (or a_g , see Eq. 4) occurs at the T_g determined by the peak of the loss modulus. Secondly, the peak of the loss modulus is much easier to read than the maximum change of the storage modulus. According

to Fig. 4, the glass transition temperatures were 383, 377, and 314 K for ABS, PC/ABS and PP/EPDM/T15, respectively. The mechanical properties and T_g are strongly loading speed/frequency-dependent due to the viscosity of polymers.^[52,53] DMA testing at 1 Hz was used because the strain rates of the two types of tests (DMA and uniaxial tension test) are comparable based on the equivalent method.^[54]

Sensitivity Analysis of Model Input Parameters

Each of the models has mandatory inputting parameters. In our model, P_U , P_R , and T_g should first be measured and

Table 3 Parameters to determine Eq. (7).

Material	Glassy state			Rubbery state		
	T_{range}^1 (K)	E_B^1 (GPa)	S^1 (MPa/K)	T_{range}^2 (K)	E_B^2 (GPa)	S^2 (MPa/K)
ABS	220–300 ^[56]	2.239	3.39	390–400 ^[57]	0.133	3.39
PC/ABS	220–300 ^[58]	2.042	3.53	398–408 ^[59]	0.095	3.53
PP/EPDM/T15	173–243 ^[60]	2.962	3.77	353–393 ^[61]	0.210	3.77

Table 4 Lower boundary, baseline, and upper boundary of E_g , E_r , and T_g .

Material	E_g (GPa)			E_r (GPa)			T_g (K)		
	L	B	U	L	B	U	L	B	U
ABS	2.103	2.239	2.375	0.116	0.133	0.150	379	383	387
PC/ABS	1.901	2.042	2.183	0.060	0.095	0.130	373	377	381
PP/EPDM/T15	2.830	2.962	3.094	0.135	0.210	0.285	310	314	318

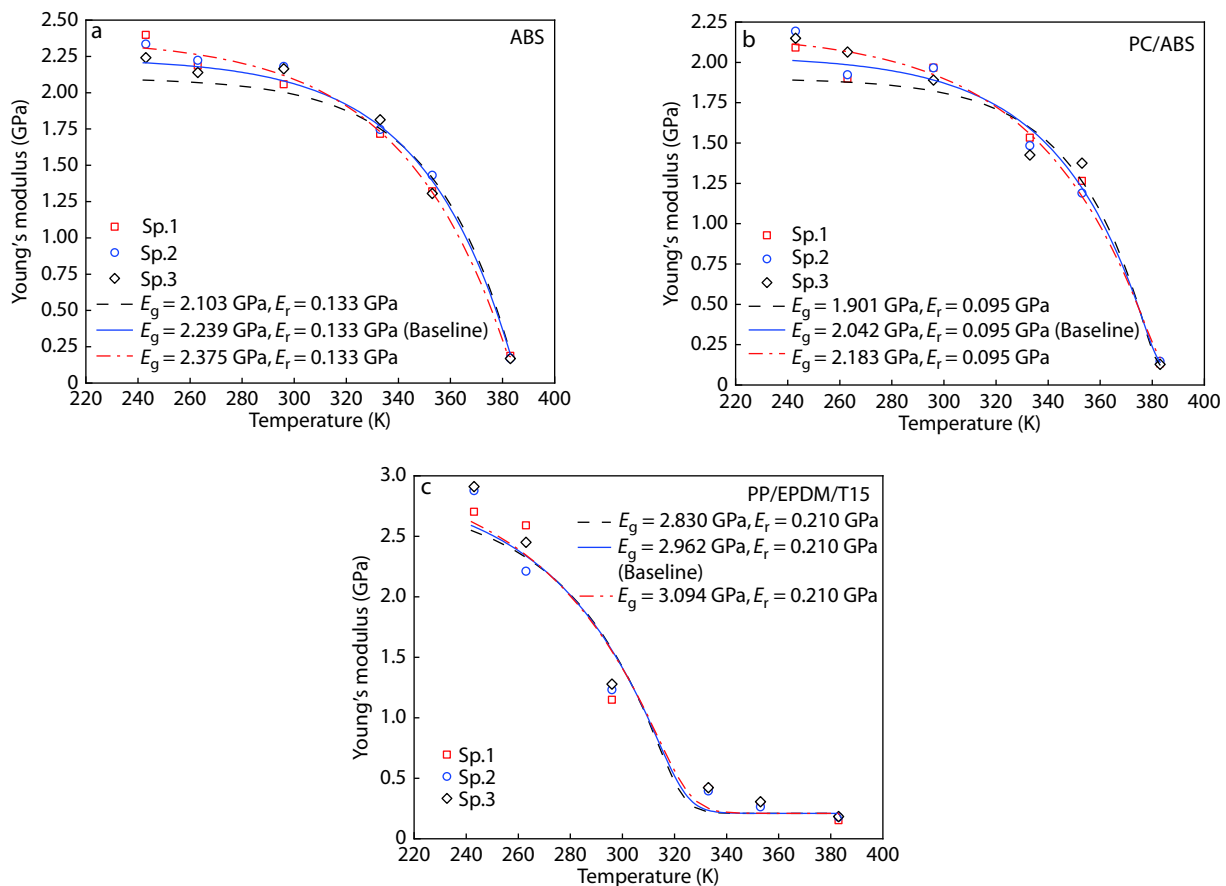


Fig. 5 Sensitivity analysis of E_g : (a) ABS, (b) PC/ABS, and (c) PP/EPDM/T15.

determined. It is noteworthy that P_U shows a small reduction with temperature increase in the glassy state, as does P_R in the rubbery state. The deviation of T_g may be caused by instrumentation error and different measurement techniques. For instance, DSC and DMA can cause a difference of ± 4 K for a wide variety of commercially available thermoplastic polymers.^[55] In order to verify our model's universality, a discussion about its sensitivity to P_U , P_R , and T_g is essential. In this section, P_U and P_R specifically refer to Young's modulus. The yield strength follows the same trend because of the linear relationship with Young's modulus.

ABS is in the glassy state at 220–300 K,^[56] in which the Young's modulus decreases at an average state of 3.39 MPa/K. The midpoint (260 K) within this temperature range is defined as the baseline of E_g (ABS). Based on the average decreasing value of Young's modulus per unit temperature, and the temperature range of the glassy state, the lower boundary and upper boundary of E_g (ABS) can be estimated by Eq. (7):

$$\begin{cases} E_L^i = E_B^i - \frac{T_{\text{range}}}{2} \cdot S^i \\ E_U^i = E_B^i + \frac{T_{\text{range}}}{2} \cdot S^i \end{cases} \quad (i = 1, 2) \quad (7)$$

where the superscript i is equal to 1 (glassy state) or 2 (rubbery state), the subscripts L , B , and U represent the lower boundary, baseline, and the upper boundary, respectively, T_{range} is the

temperature range, and S represents the average decrease in Young's modulus per unit temperature. Eq. (7) also applies to PC/ABS and PP/EPDM/T15, and thus we can estimate the upper and lower boundaries of E_g and E_r for PC/ABS and PP/EPDM/T15. Based on Table 2, Fig. 4, and various literatures,^[56–61] S , E_B , and T_{range} can be approximated. In this study, we hypothesize that S^1 approximately equals S^2 for the same material. The parameter values in Eq. (7) are summarized in Table 3.

The baseline of T_g is defined from the peak of the loss modulus, and the range of variation in T_g corresponds to the measurement difference (± 4 K). On the basis of Eq. (7), Table 3, and literatures,^[56–61] the lower and upper boundaries of E_g , E_r , and T_g can be calculated and are summarized in Table 4.

Sensitivity Analysis of E_g

Fig. 5 illustrates the predicted Young's modulus versus temperature for the three polymers, showing that different values of E_g have almost no effect on the glass transition region or the rubbery state. The maximum discrepancy between our model predictions and the experimental results is observed in the glassy state for ABS (Fig. 5a). Nevertheless, when compared to the order of magnitude of the modulus in the glassy state, this discrepancy can be neglected.

Sensitivity Analysis of E_r

Fig. 6 depicts the predicted Young's modulus versus temperature for the three polymers, showing slight variations in the

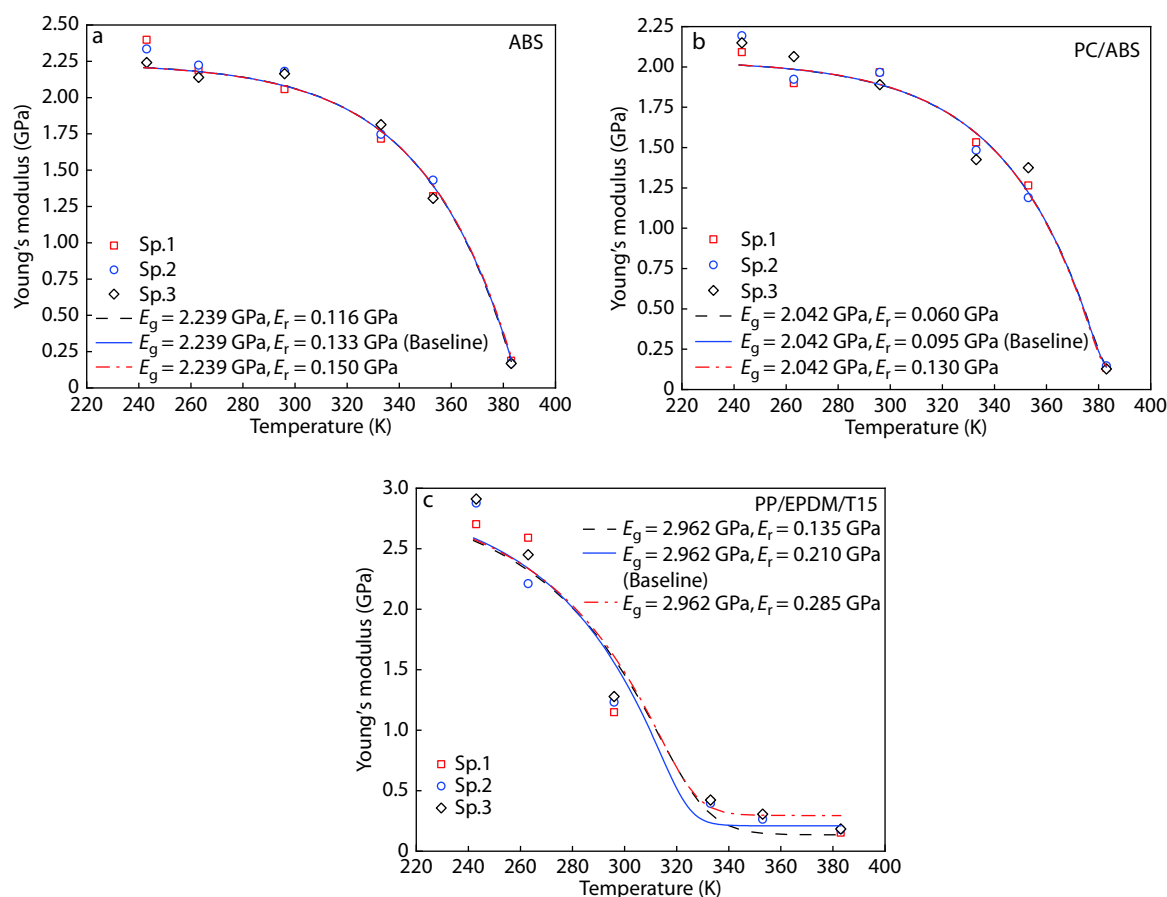


Fig. 6 Sensitivity analysis of E_r : (a) ABS, (b) PC/ABS, and (c) PP/EPDM/T15.

glass transition region and glassy state in response to different values of E_r . Our model shows slightly reduced accuracy when the temperature approaches the rubbery state for PP/EPDM/T15 (Fig. 6c). In consideration of the low (close to zero) modulus in the rubbery state, the difference between the model predictions and the experimental results can be neglected.

Sensitivity Analysis of T_g

Fig. 7 presents the predicted Young's modulus versus temperature for the three polymers based on different T_g . The results show excellent agreement, demonstrating that variations in T_g have only a slight influence on the entire curves and can be neglected.

To conclude, Figs. 5–7 demonstrate that the present model is insensitive to variations in E_g , E_r , and T_g . Our model has a strong pervasive characteristic. Researchers do not want to be concerned with the initial values of input parameters.

Temperature-dependent Modulus and Yield Strength Model

The predicted and experimental temperature-dependent Young's moduli and yield strengths of three thermoplastic polymers are shown in Fig. 8. Young's modulus and yield strength decrease slowly as temperature increases in the glassy state, and show a significant decrease during the glass transition. The model predictions showed excellent agreement with the experimental data over the full glass transition region. Fitting parameters of each model were calculated by the

nonlinear least square method (MATLAB R2015a) using six representative experimental points. Stretch-induced crystallization and melting-recrystallization take place during uniaxial tensile deformation.^[62] Herein, it is worth mentioning that Young's modulus is calculated under small deformation ($\epsilon < 0.25\%$) and the strain at yield point is less than 5%. Under this condition, the crystallinity of the polymer has little changed.^[42] Therefore, the change of crystallinity is neglected and the parameter m is considered as a constant.

Comparisons of Different Temperature-dependent Models

The predictions of models from various literatures are also depicted in Fig. 8. The consistencies between the experimental and predicted data can be evaluated by using the correlation coefficient R^2 , as presented in Table 5. A larger R^2 value (close to 1) represents greater consistency. From Fig. 8, it can be noticed that both the present model and Bai *et al.* model can accurately predict the Young's moduli of the three thermoplastic polymers. It can also be noticed that Ha and Springer model and the present model can accurately predict the yield strength of the three thermoplastic polymers. The correlation coefficients for these models are all close to 1. Mahieux and Reifsnider's model and Gibson *et al.* model lose accuracy around T_g . Especially, the model by Gibson *et al.* gives a correlation coefficient of less than 0.4 when predicting the yield strengths of ABS and PC/ABS.

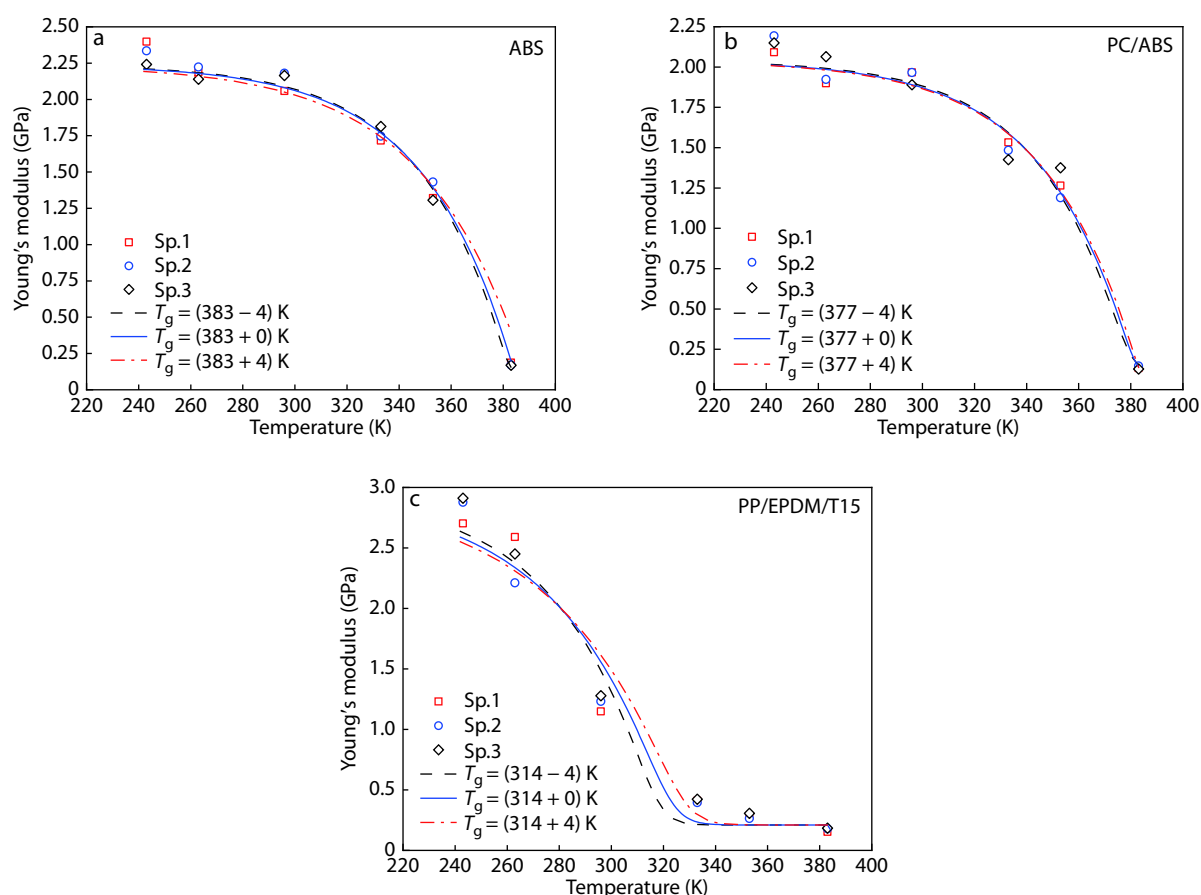


Fig. 7 Sensitivity analysis of T_g : (a) ABS, (b) PC/ABS, and (c) PP/EPDM/T15.

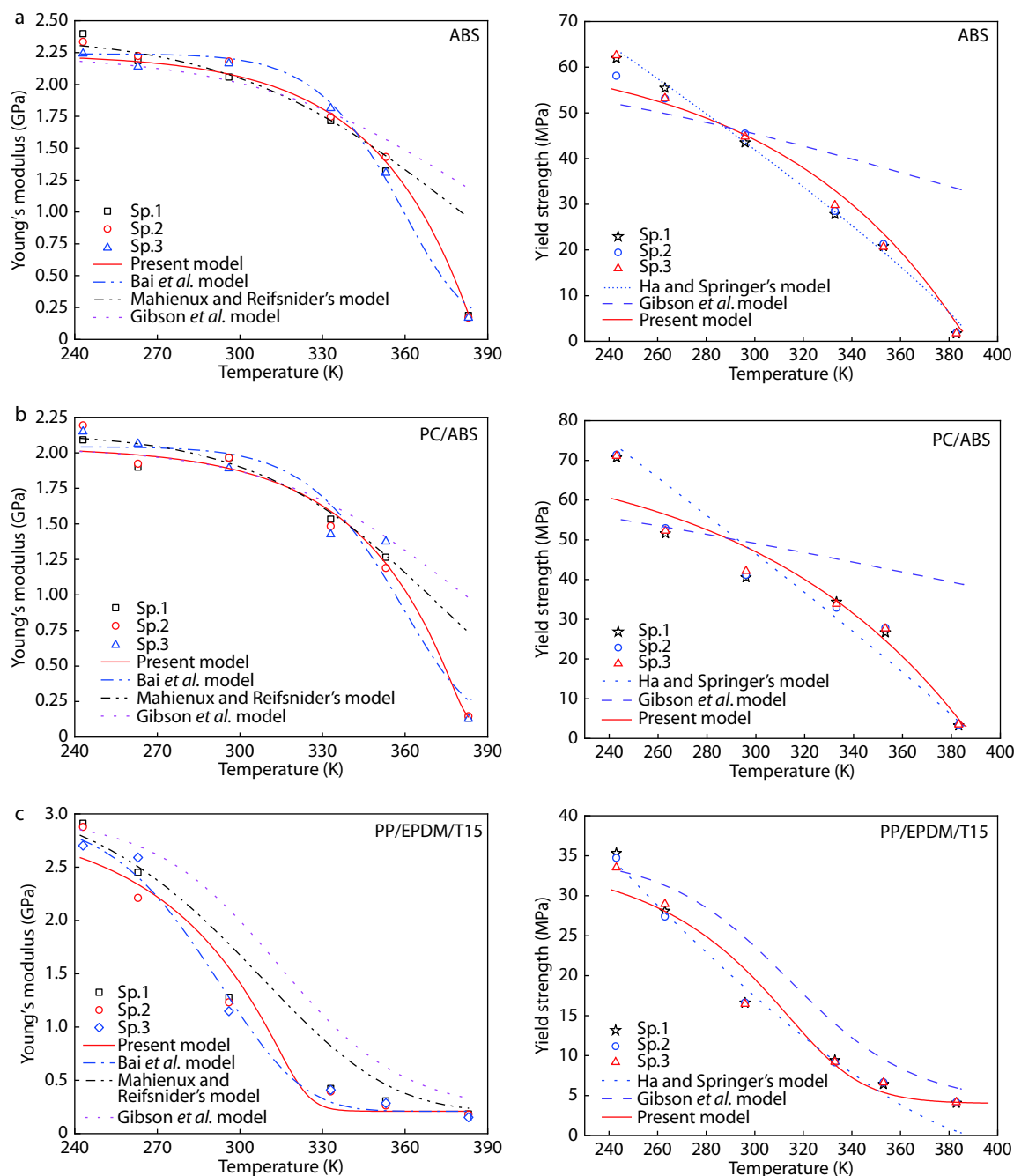


Fig. 8 Young's modulus (left) and yield strength (right) versus temperature: Experimental data and predicted curves for (a) ABS, (b) PC/ABS, and (c) PP/EPDM/T15.

Table 5 Correlation coefficients R^2 for three thermoplastic polymers from each model.

Material	R^2 for Young's modulus				R^2 for yield strength		
	Present model	Bai <i>et al.</i> model	Mahieux and Reifsnider's model	Gibson <i>et al.</i> model	Ha and Springer's model	Gibson <i>et al.</i> model	Present model
ABS	0.9940	0.9874	0.8064	0.6605	0.9892	0.3721	0.9722
PC/ABS	0.9869	0.9689	0.8596	0.7160	0.9002	0.2579	0.9255
PP/EPDM/T15	0.9669	0.9957	0.9203	0.7899	0.9754	0.8519	0.9541

Extension of the Present Model

In order to extend and generalize the present model, we compared model predictions and experimental data from other

published studies. Fig. 9 compares our predicted mechanical property for four materials with experimental data, including poly(lactic acid) (PLA),^[63] poly(butylene terephthalate) (PBT),^[64]

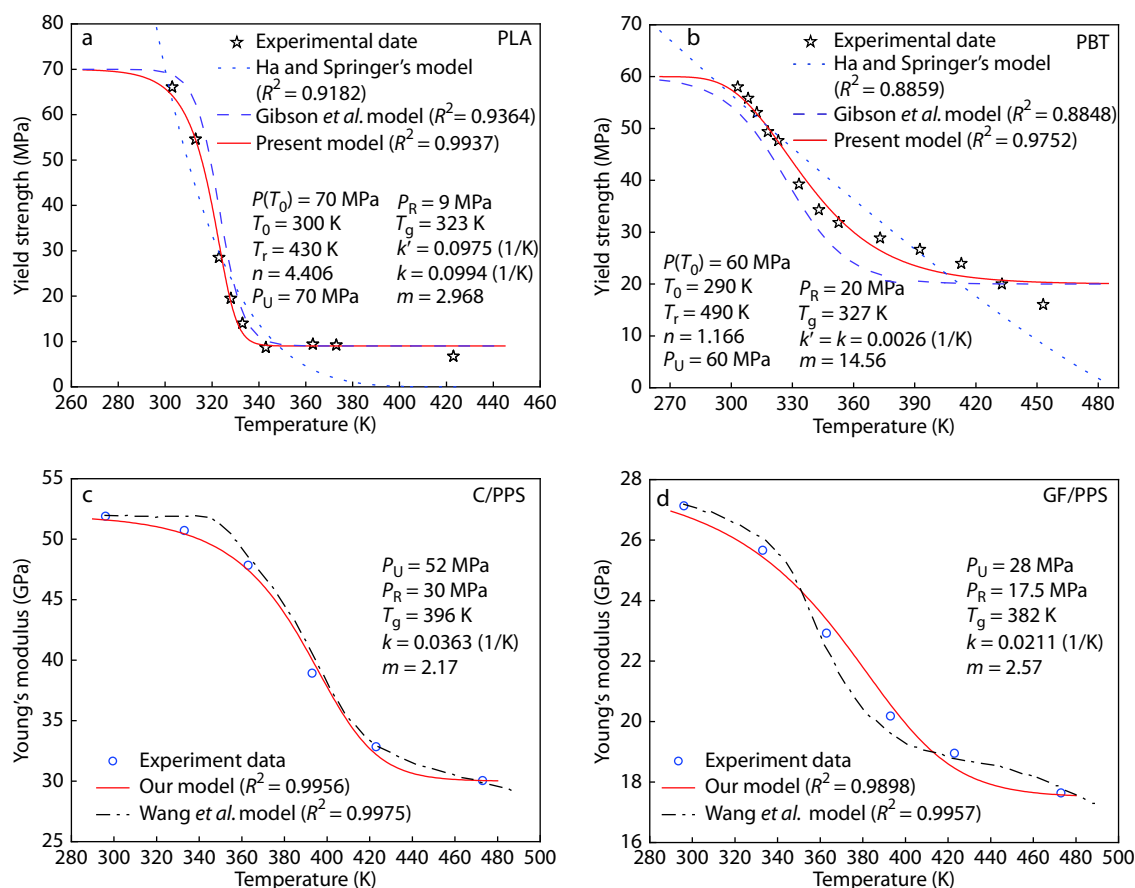


Fig. 9 Mechanical property versus temperature: experimental data and predicted curves for (a) PLA, (b) PBT, (c) C/PPS, and (d) GF/PPS.

as well as carbon fiber fabric (3K T300) and glass fiber fabric (7781 E-Glass) polyphenylene sulfide (PPS)-based composites.^[9] The yield strength of PLA is calculated from the stress-strain curves of uniaxial tension tests performed by Zhou *et al.*,^[63] who focused on mechanical properties of injection-molded PLA samples after annealing at different temperatures. For PBT, the yield strength is directly taken from Zhang's result.^[64] The Young's modulus of the two types of PPS-based composites are taken from Wang *et al.*,^[9] who studied the effect of temperature on bending stiffness and strength. Comparing the experimental data and model predictions shows excellent agreement on temperature-dependent modulus throughout the range of temperatures investigated. This broad agreement indicates that the present model can be applied to different thermoplastic polymers and their composites.

CONCLUSIONS

A general two-parameter model was established to predict the temperature-dependent Young's modulus and yield strength of different thermoplastic polymers. The physical meaning of k is related to the polydispersity index ($\bar{D}$), a measure of the distribution of molecular weight in a given polymer: the smaller the value of k , the higher the value of $\bar{D}$. The physical meaning of m is related to the crystallinity of the polymer: the smaller the value of m , the larger the degree of crystallinity. For polymers, entanglement or entangle number is

another typical parameter, which can influence their viscosity,^[52] yield strength,^[65] and crystallinity.^[66] The present model is still a macroscopic model, though the physical meaning of parameters is analysed from the molecular level. Molecular dynamics simulation is a better method to understand deformation mechanisms on the molecular level.^[67,68]

The predictions of the present model for the three materials were compared with those of widely used models. The present model exhibits excellent agreement with experimental data for all studied thermoplastic polymers, as well as with published experimental data of other thermoplastic polymers and composites. Within the measured ranges, variations in input parameters had only a slight effect on the model results. This robustness to input values enables more flexible selection of these parameters without adversely affecting the model reliability. This strongly indicates that the present model has universality. Moreover, it can be conveniently applied due to not only its more concise form but also its insensitivity to initial values.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://dx.doi.org/10.1007/s10118-020-2360-7>.

ACKNOWLEDGMENTS

This work was financially supported by the National Natural Science Foundation of China (Nos. 11972218 and 11472165).

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