

A Uniform Phenomenological Constitutive Model for Glassy and Semicrystalline Polymers

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A phenomenological constitutive model is proposed on the basis of four models: the Johnson-Cook model, the G'Sell-Jonas model, the Matsuoka model, and the Brooks model. The proposed constitutive model has a concise expression of stress dependence on strain, strain rate and temperature. It is capable of uniformly describing the entire range of deformation behavior of glassy and semicrystalline polymers, especially the intrinsic strain softening and subsequent orientation hardening of glassy polymers. At least three experimental stress-strain curves including variation with strain rate and temperature are needed to calibrate the eight material coefficients. Sequential calibration procedures of the eight material coefficients are given in detail. Predictions from the proposed constitutive model are compared with experimental data of two glassy polymers, polymethyl-methacrylate and polycarbonate under various deformation conditions, and with that of the G'Sell-Jonas model for polyamide 12, a semicrystalline polymer.

INTRODUCTION

Over the past four decades, much effort has been devoted to modeling stress-strain constitutive relationships for engineering polymers (1–14). The early attempts are based on the additive combination of Hooke's law and Newton's law because polymers display intermediate mechanical properties between elastic solids and viscous liquids. Simple mechanical models such as the Kelvin model, the Maxwell model, and the SLS model were proposed to describe the linear viscoelasticity (1). Nonlinearity starts to be exhibited at a strain of about 0.01 for most polymers. Non-linearity was introduced in viscoelastic constitutive models in references (2–6). The stress-strain behavior beyond yielding is of interest if rheological properties of engineering polymer need to be considered. Boyce (7) developed a constitutive framework to model strain rate, temperature and pressure dependent yield and

rheological behavior of glassy polymers on the basis of molecular theory and finite kinematics. Hasan (8) proposed a model for glassy polymers based on theories of macromolecules and thermodynamics to simulate the neck formation and propagation of glassy polymers. Amoedo and Lee (9) proposed a model to describe the large inelastic deformation of glassy and semicrystalline polymers based on isotropic formulations of viscoplasticity.

Several investigators (10–13) have attempted to predict the entire range of deformation behavior of engineering polymers using constitutive models based on either physical or phenomenological approaches. G'Sell and Jonas (11) proposed a concise phenomenological constitutive equation for semicrystalline polymers. In this paper, we develop a constitutive model to uniformly describe the entire range of stress-strain constitutive relationship of glassy and semicrystalline polymers so that it can be easily incorporated in finite element analysis codes to predict the deformation behavior of polymers under monotonic compressive loading.

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MECHANICAL BEHAVIOR OF GLASSY AND SEMICRYSTALLINE POLYMERS

Compared with metallic materials, polymers exhibit lower stiffness, lower strength and are very sensitive to temperature and strain rate. With decrease of temperature or increase of strain rate, the stress over the entire range of strain increases, and for glassy polymers the yielding strain becomes larger. This unique property is related to their molecular structure and composition. Glassy polymers exhibit viscoelasticity, yielding followed by intrinsic strain softening and subsequent orientation hardening, but semicrystalline polymers do not exhibit the behavior of strain softening though they show viscoelasticity, initial viscoplasticity and subsequent strain hardening. A number of investigators have conducted studies on both glassy and semicrystalline polymers (2, 8, 9, 11, 14, 15). The above-mentioned mechanical characteristics of the two types of polymers can be found in these references.

DEVELOPMENT OF THE CONSTITUTIVE MODEL

Two main approaches are often taken to develop material constitutive models. The first is a physical one where the stress-strain relationship is considered on the basis of thermodynamic activation of molecules. The second is a phenomenological one where the stress-strain relationship is modeled by fitting experimental data. Constitutive models from a physical basis generally have very complex form including a series of complicated equations. In numerical simulations the preferable model is the phenomenological one because of its simplicity and efficient running algorithms on a computer.

The new constitutive model for polymers introduced in this paper is based on four previous models: Johnson-Cook (16), G'Sell-Jonas (11), Matsuoka (2), and Brooks (17). Johnson and Cook proposed a phenomenological constitutive model to describe the plastic behavior of ductile metals (16). The model is given by the following equation:

$$\sigma = (C_1 + C_2 \varepsilon^N)(1 + C_3 \ln \dot{\varepsilon})(1 - \hat{\theta}^M) \quad (1)$$

where σ is equivalent yield stress, ε is equivalent plastic strain, $\dot{\varepsilon}$ is equivalent plastic strain rate, $\hat{\theta}$ is homologous temperature, and C_1 , C_2 , C_3 , N , and M are material coefficients. This model gives very concise expression of the stress dependence on strain, strain rate and temperature during the plastic deformation process of metallic materials. Because of its simple form, it is the most popular constitutive model in use today (18).

G'Sell and Jonas (11) proposed a phenomenological model for the stress-strain behavior of semicrystalline polymers. Their model is given by:

$$\sigma = K e^{h\varepsilon^2} (1 - e^{-W\varepsilon}) (\dot{\varepsilon})^m e^{\frac{Q}{T}} \quad (2)$$

where σ is equivalent stress, ε is equivalent strain, $\dot{\varepsilon}$ is equivalent strain rate, T is absolute temperature, and

K , a , W , h and m are material coefficients. The model integrates viscoelasticity and viscoplasticity in one equation. The function $(1 - e^{-W\varepsilon})$ takes into account the initial non-linear elastic behavior, the function $e^{h\varepsilon^2}$ describes plastic strain hardening, and the function $(\dot{\varepsilon})^m$ describes viscous behavior. This equation models well the behavior of semicrystalline polymers, but it is unable to reproduce the intrinsic strain-softening behavior of glassy polymers.

Matsuoka proposed a constitutive equation to predict the behavior of glassy polymers (2). The equation is given as:

$$\sigma = E_0 e^{-C_0 \varepsilon} e^{-\left(\frac{\varepsilon}{\dot{\varepsilon}\tau}\right)^{\beta}} \quad (3)$$

where σ is equivalent stress, ε is equivalent strain, $\dot{\varepsilon}$ is equivalent strain rate, τ is effective relaxation time associated with temperature, and E_0 , C_0 , and β are material coefficients. This model depicts the non-linear viscoelasticity, yielding and subsequent strain softening behavior of glassy polymers. Although at relatively larger strains the model does not correctly predict the behavior of glassy polymers, we may use a part of this equation in simplified form for the proposed constitutive model to describe the shift behavior of yielding of glassy polymers. Consider the function $u(\varepsilon)$ in the form:

$$u(\varepsilon) = \varepsilon e^{-\left(\frac{\varepsilon}{\dot{\varepsilon}\tau}\right)^{\beta}} \quad (4)$$

The derivative of function $u(\varepsilon)$ at a given $\dot{\varepsilon}$ is:

$$\frac{d[u(\varepsilon)]}{d\varepsilon} = \left(1 - \frac{\varepsilon}{\dot{\varepsilon}\tau}\right) e^{-\left(\frac{\varepsilon}{\dot{\varepsilon}\tau}\right)^{\beta}} \quad (5)$$

When $\frac{d[u(\varepsilon)]}{d\varepsilon} = 0$, that is to say when $\varepsilon = \dot{\varepsilon}\tau$, the

function $u(\varepsilon)$ has a maximum value of $\left(\frac{\dot{\varepsilon}\tau}{e}\right)$. It is clear that with an increase of strain rate $\dot{\varepsilon}$ or decrease of temperature (equivalent to increased relaxation time τ) both the maximum value of $u(\varepsilon)$ and the strain ε at maximum value of $u(\varepsilon)$ increase. This gives a mathematical way to describe the shift behavior of yield point for glassy polymers.

Brooks developed a constitutive model for dynamically recrystallizing materials (17). The model is given by:

$$\sigma = K \lambda_{zz}^m \{ \lambda_0 \cdot \lambda_{zz}^q + [1 - \lambda_0 \cdot \lambda_{zz}^q] e^{-\alpha\varepsilon} \} \cdot (1 - e^{-\beta\varepsilon}) \quad (6)$$

where,

$$\lambda_{zz} = \dot{\varepsilon} \cdot e^{\frac{Q}{RT}} \quad (7)$$

σ is equivalent yield stress, ε is equivalent plastic strain, $\dot{\varepsilon}$ is equivalent plastic strain rate, T is absolute temperature, and K , m , λ_0 , q , α , β , Q and R are material coefficients. The Brooks model cannot predict strain-hardening behavior, but part of this model is useful for the proposed constitutive model.

Consider the term $\lambda_0 \cdot \lambda_{zz}^q + (1 - \lambda_0 \cdot \lambda_{zz}^q) e^{-\alpha\varepsilon}$. With increased strain ε , the term approaches exponentially from the initial value of 1 (when $\varepsilon = 0$) to a steady state value of $\lambda_0 \cdot \lambda_{zz}^q$. If the initial value 1 is replaced by some expression describing the initial deformation, and the steady state value $\lambda_0 \cdot \lambda_{zz}^q$ is replaced by some expression describing the large deformation of polymers, a constitutive model capable of describing both the viscoelasticity and viscoplasticity behavior of polymers probably can be developed.

Following these ideas, we propose a uniaxial constitutive model (called DSGZ model) for polymers given by the following equation:

$$\sigma(\varepsilon, \dot{\varepsilon}, T) = K$$

$$\left\{ f(\varepsilon) + \left[\frac{\varepsilon \cdot e^{(1-C_3 \cdot h(\dot{\varepsilon}, T))}}{C_3 \cdot h(\dot{\varepsilon}, T)} - f(\varepsilon) \right] \cdot e^{[\ln(g(\dot{\varepsilon}, T)) - C_4)]\varepsilon} \right\} \cdot h(\dot{\varepsilon}, T) \quad (8)$$

where,

$$f(\varepsilon) = (e^{-C_1\varepsilon} + \varepsilon^{C_2}) (1 - e^{-\alpha\varepsilon}) \quad (9)$$

$$h(\dot{\varepsilon}, T) = (\dot{\varepsilon})^m e^{\frac{g}{T}} \quad (10)$$

and $g(\dot{\varepsilon}, T)$ is defined to be the dimensionless form of $h(\dot{\varepsilon}, T)$. The eight material coefficients in this model are K (Pa · s^m), C_1 , C_2 , C_3 (s^m), C_4 , α (K), m , and α .

The structure of the proposed constitutive equation is similar to that of Brooks model, Eq 6, with some fundamental changes. First consider the term $\lambda_0 \cdot \lambda_{zz}^q + (1 - \lambda_0 \cdot \lambda_{zz}^q) e^{-\alpha\varepsilon}$, which has an initial value of 1 corresponding to $\varepsilon = 0$. We replace this initial value of 1 by a function related to Eqs 4 and 10 in the form $\frac{\varepsilon \cdot e^{(1-C_3 \cdot h(\dot{\varepsilon}, T))}}{C_3 \cdot h(\dot{\varepsilon}, T)}$. This part describes the shift behav-

ior of the yield point with strain rate and temperature for glassy polymers. Second, note that the term $\lambda_0 \cdot \lambda_{zz}^q + (1 - \lambda_0 \cdot \lambda_{zz}^q) e^{-\alpha\varepsilon}$ has a steady state value of $\lambda_0 \cdot \lambda_{zz}^q$ at large strain. We replace the steady state value by $f(\varepsilon)$ of Eq 9, which represents the strain hardening part of the constitutive relation for polymers. Third we replace the material constant α in the Brooks model (17), which represents the exponential evolution from initial value to steady state value, with a strain rate, temperature and material dependent value $\ln(h(\dot{\varepsilon}, T)) - C_4$. Finally, similar to the G'Sell-Jonas model (11), the entire constitutive relation is multiplied by $(\dot{\varepsilon})^m e^{\frac{g}{T}}$, which represents the stress dependence on strain rate and temperature.

Our proposed uniaxial model can be extended to three-dimensional framework through the generalized von Mises flow rule. Suppose $[s_{ij}]$ and $[\dot{e}_{ij}]$ are deviatoric true stress tensor and deviatoric true plastic strain rate tensor, respectively. Then, the equivalent stress is given by:

$$\sigma = \sqrt{\frac{3}{2} (s_{ij} : s_{ij})} \quad (11)$$

The equivalent plastic strain rate is given by:

$$\dot{\varepsilon} = \sqrt{\frac{2}{3} (\dot{e}_{ij} : \dot{e}_{ij})} \quad (12)$$

The equivalent plastic strain is given by integration of equivalent plastic strain rate over the whole time scale:

$$\varepsilon = \int_0^t \dot{\varepsilon} dt \quad (13)$$

The constitutive model given by Eq 8 describes the relationship of the three equivalent values. The generalized von Mises flow rule has been used by a number of investigators to describe the instantaneous three-dimensional stress-strain constitutive relationship of plastic deformation of polymers (9, 19–22):

$$[s_{ij}] = \frac{2\sigma}{3\varepsilon} [\dot{e}_{ij}] \quad (14)$$

CALIBRATION OF MATERIAL COEFFICIENTS

The measurement of true stress, true strain and true strain rate are required to calibrate the eight material coefficients in the proposed constitutive model. Uniaxial tension tests are widely used to determine mechanical properties of materials. The load-elongation data are usually transformed into engineering stress-strain curves via dividing load by the initial cross-section and elongation by initial length. These engineering stress-strain curves do not provide a correct description of the mechanical behavior of material subject to large deformation because of the appearance of necking that lead to strain localization. Simple shear tests often introduce inhomogeneous deformation field at large strains (23). Uniaxial compression tests are the best choice to obtain the required true stress-true strain data because homogeneous macroscopic deformation field can be found through the use of appropriate lubrication to reduce the frictional effect in this type of test (8). Experiments show thermo-mechanical coupling is important for large strain and high strain rate deformation of polymers (14, 24). It significantly affects the mechanical behavior. To keep isothermal condition in experiments, low strain rate tests are generally performed. The strain rates used in experiments are no more than 0.001/s for most polymers (14).

At least three experimental stress-strain curves including variation with strain rate and temperature are needed to calibrate the eight material coefficients required in the proposed constitutive model of Eq 8. The sequential calibration procedures are:

- Deduce C_1 , C_2 from three different large strain points on one stress-strain curve. From Eq 8 and 9 and at large strains, the stress is approximated to be:

$$\sigma(\varepsilon, \dot{\varepsilon}, T) = K \cdot (e^{-C_1 \cdot \varepsilon} + e^{C_2}) \cdot h(\dot{\varepsilon}, T) \quad (15)$$

Suppose three large strain points on a stress-strain curve are $(\varepsilon_1, \sigma_1)$, $(\varepsilon_2, \sigma_2)$, $(\varepsilon_3, \sigma_3)$, the following system of equations can be obtained:

$$\begin{cases} \frac{e^{-C_1 \cdot \varepsilon_1} + e^{C_2}}{e^{-C_1 \cdot \varepsilon_2} + e^{C_2}} = \frac{\sigma_1}{\sigma_2} \\ \frac{e^{-C_1 \cdot \varepsilon_1} + e^{C_2}}{e^{-C_1 \cdot \varepsilon_3} + e^{C_2}} = \frac{\sigma_1}{\sigma_3} \end{cases} \quad (16)$$

- Solve Eq 16 to get C_1 and C_2 . We have used an iteration method to find values for C_1 and C_2 .
- Deduce m from curves with different strain rate. Suppose points on two curves with strain rate $\dot{\varepsilon}_1$ and $\dot{\varepsilon}_2$ are (ε, σ_1) and (ε, σ_2) at the same large strain ε . The following equation can be obtained from Eqs 10 and 15:

$$\left(\frac{\dot{\varepsilon}_1}{\dot{\varepsilon}_2} \right)^m = \frac{\sigma_1}{\sigma_2} \quad (17)$$

The value of m can be obtained by solving Eq 17, in the form

$$m = -\frac{\ln\left(\frac{\sigma_1}{\sigma_2}\right)}{\ln\left(\frac{\dot{\varepsilon}_1}{\dot{\varepsilon}_2}\right)} \quad (18)$$

- Deduce ' α ' from curves with different temperature. Suppose points on two curves with temperature T_1 and T_2 are (ε, σ_1) and (ε, σ_2) at the same large strain ε . The following equation can be obtained from Eqs 10 and 15:

$$\frac{e^{\frac{a}{T_1}}}{e^{\frac{a}{T_2}}} = \frac{\sigma_1}{\sigma_2} \quad (19)$$

The value of a can be found from Eq 19, in the form

$$\alpha = \frac{\ln\left(\frac{\sigma_1}{\sigma_2}\right)}{\left(\frac{1}{T_1} - \frac{1}{T_2}\right)} \quad (20)$$

- After C_1 , C_2 , m , and a are known, the value of K can be deduced from Eq 15 at a large strain point.

- In order to solve for C_3 , we consider the term $\frac{(\varepsilon - C_3 \cdot h(\dot{\varepsilon}, T))}{c_3 \cdot h(\dot{\varepsilon}, T)}$ in Eq 8. At the yielding strain ε_y , corresponding to the maximum stress before softening, this term has a maximum value. Therefore, C_3 can be found using the following equation:

$$C_3 h(\dot{\varepsilon}, T) = \varepsilon_y \quad (21)$$

Using Eq 10 that defines $h(\dot{\varepsilon}, T)$, we find

$$C_3 = \frac{\varepsilon_y}{e^{\frac{a}{T}} (\dot{\varepsilon})^m} \quad (22)$$

- The remaining two coefficients in Eq 8 α and C_4 are particularly important for fitting the initial part of the experimental stress-strain curves. Their values are found in a two-step procedure. First, based on experiences from experimental results of polymers, we can roughly estimate α from $1 - e^{-\alpha\varepsilon} = 0.97$, where ε is chosen to be the end point of strain softening, and C_4 as $C_4 = 7 + \ln(\dot{\varepsilon}^m \cdot e^{\frac{a}{T}})$ for glassy polymers and $C_4 = 200 + \ln(\dot{\varepsilon}^m \cdot e^{\frac{a}{T}})$ for semicrystalline polymers. Second, the two estimated coefficients are adjusted together to fit the initial part of experimental stress-strain curves.

MODEL VALIDATION

Hasan (8) did many uniaxial compression experiments on the glassy polymers polymethyl-methacrylate (PMMA) and polycarbonate (PC). In this paper, we use his experimental data to calibrate the eight material coefficients of the proposed constitutive model and then compare model predictions with experimental data at different strain rates and temperatures to validate the model for glassy polymers.

Three sets of compression experimental data for annealed PMMA are used to calibrate the eight material coefficients following the above procedure: 1. temperature of 296K, true strain rate of 0.001/s; 2. temperature of 323K, true strain rate of 0.001/s; 3. temperature of 296K, true strain rate of 0.0001/s. The eight calculated material coefficients are listed in Table 1.

These coefficients are then used in the proposed model given by Eq 8. Figure 1 shows the model prediction and experimental data for annealed PMMA up to true strain of 1.0. Figure 2 shows model prediction

Table 1. Material Coefficients for PMMA.

C_1	C_2	m	a (K)	K (MPa · s ^m)	C_3 (s ^m)	C_4	α
1.91	1.49	0.064	1191	3.9	0.0029	11	11.7

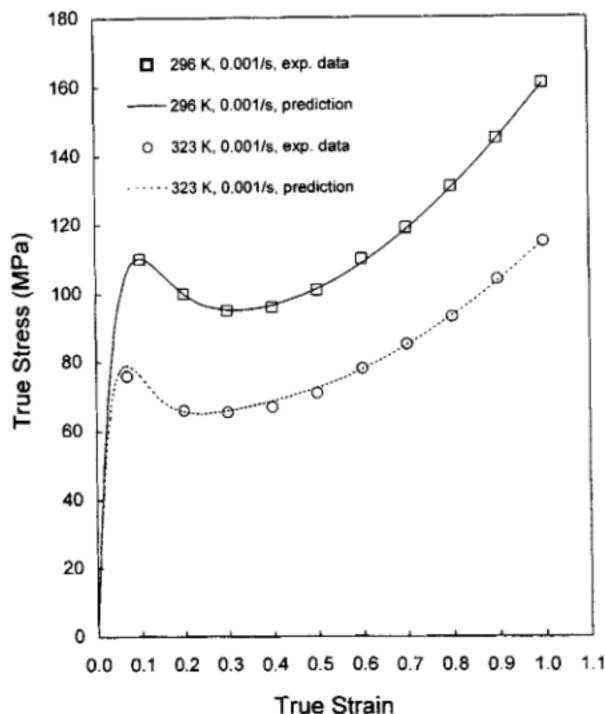


Fig. 1. Comparison of DSGZ model prediction with experimental data (8) for annealed PMMA ($T = 296\text{ K}, 323\text{ K}$; $\dot{\epsilon} = 0.001/\text{s}$).

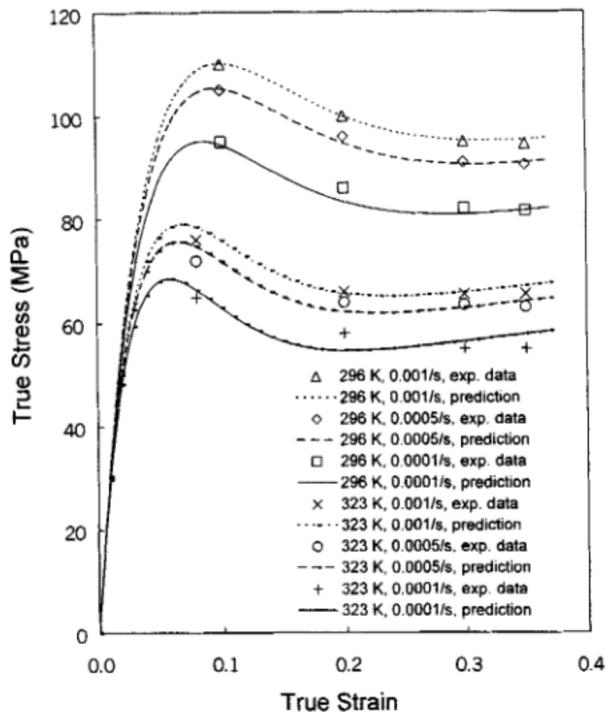


Fig. 2. Comparison of DSGZ model prediction with experimental data (8) for annealed PMMA ($T = 296\text{ K}, 323\text{ K}$; $\dot{\epsilon} = 0.001/\text{s}, 0.0005/\text{s}, 0.0001/\text{s}$).

and experimental data for annealed PMMA at different true strain rates and temperatures. Figure 3 shows model prediction and experimental data for annealed PMMA at 300K and a true strain rate of 0.001/s.

Three sets of compression test data for annealed PC are used to identify the eight material coefficients: 1. temperature of 296K, true strain rate of 0.001/s; 2. temperature of 348K, true strain rate of 0.001/s; 3. temperature of 296K, true strain rate of 0.0001/s. The eight calculated material coefficients are listed in Table 2.

Figures 4 and 5 show model predictions and experimental data of annealed PC at various temperatures and strain rates. From Figures 1 to 5, it can be seen that the proposed DSGZ constitutive model can be used to accurately predict the stress-strain behavior of glassy polymers over a wide range of strains and temperatures.

Schang *et al.* (19) did a series of experiments to obtain the true stress-true strain data of polyamide 12, a semicrystalline polymer. Using these experimental data, they deduced the five material coefficients in G'Sell-Jonas model for polyamide 12. The results are shown in Table 3.

We use three sets of curves from G'Sell-Jonas model to calculate the eight material coefficients in the proposed model: 1. temperature of 296K, true strain rate of 0.001/s; 2. temperature of 320K, true strain rate of 0.001/s; 3. temperature of 296K, true strain rate of 0.0001/s. Results are listed in Table 4.

Figure 6 shows comparison of the predictions from G'Sell-Jonas model and that of the proposed DSGZ model for polyamide 12 at various temperatures and strain rates. It can be seen that predictions from the

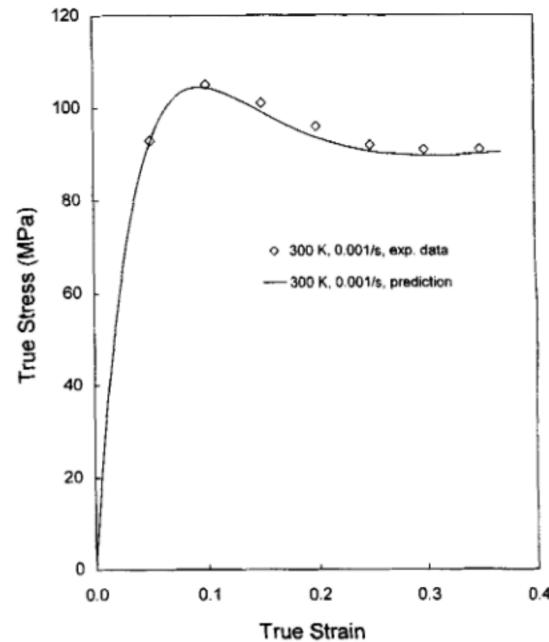


Fig. 3. Comparison of DSGZ model prediction with experimental data (8) for annealed PMMA ($T = 300\text{ K}$; $\dot{\epsilon} = 0.001/\text{s}$).

Table 2. Material Coefficients for PC.

C_1	C_2	m	a (K)	K (MPa · s ^m)	C_3 (s ^m)	C_4	α
0.49	4.02	0.038	415	28.4	0.03	5.8	6.8

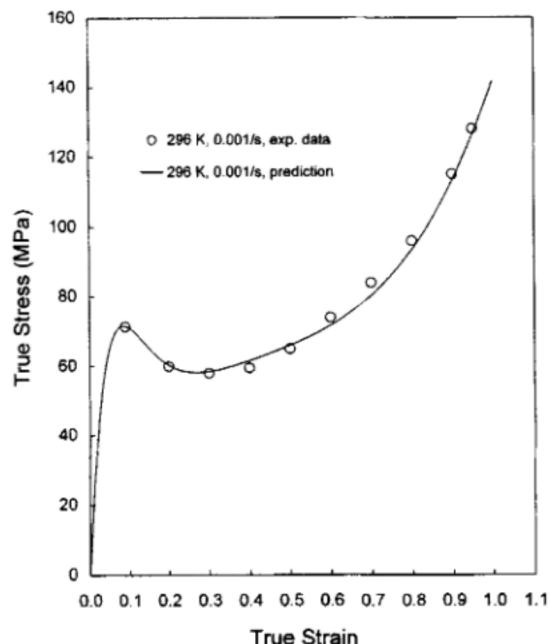


Fig. 4. Comparison of DSGZ model prediction with experimental data (8) for annealed PC ($T = 296$ K; $\dot{\epsilon} = 0.001$ /s).

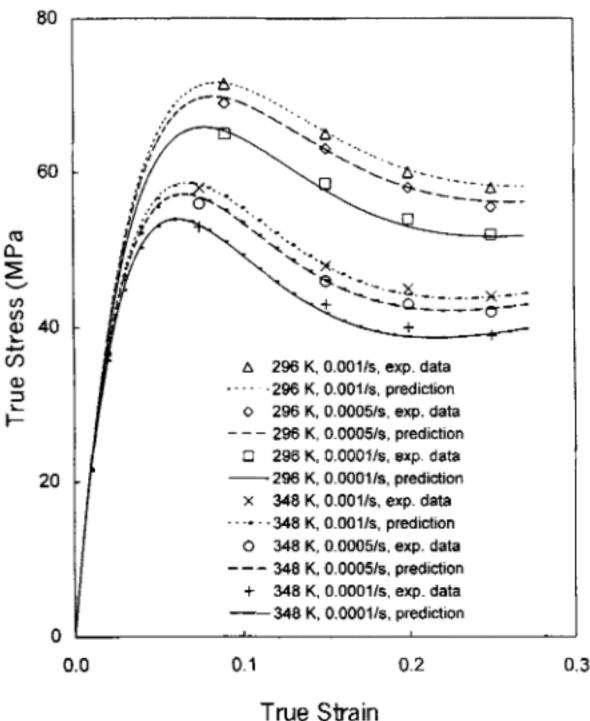


Fig. 5. Comparison of DSGZ model predictions with experimental data (8) for annealed PC ($T = 296$ K, 348 K; $\dot{\epsilon} = 0.001$ /s, 0.0005/s, 0.0001/s).

two models are almost the same except for some small differences in the middle range of strains. Since we do not have experimental data for this material, it is difficult to conclude which model is more accurate. However, it is reasonable to conclude that the proposed model can be used to accurately predict the stress-strain behavior of semicrystalline polymers.

Finally, the resulting stress-strain curves are not very sensitive to most of the material parameters except ' α '. This is expected because that parameter appears in the exponential term, which is the one related to temperature. For the parameters C_1 , C_2 and m , if the value of a parameter changes by 5%, the stress-strain curve changes by less than 2.5%. For the remaining three parameters C_3 , C_4 and α , if the value of a parameter changes by 5%, the stress-strain curve changes by less than 0.5%. Recently, Macdougall and Harding (25) used the shear test results to describe the behavior of Ti6Al4V titanium alloy in uniaxial loading and obtained good agreement between the predicted and experimentally measured force-time curves for specimens of both cylindrical and rectangular cross section. In a similar manner, it is expected that our proposed DSGZ constitutive model can also be used to describe the behavior of the polymers in pure shear though in this paper it is just used to describe the behavior of polymers in pure compression.

CONCLUSIONS

Using concepts from the Johnson-Cook model, G'Sell-Jonas model, Matsuoka model and Brooks model, a new phenomenological constitutive model capable of uniformly describing the entire range of deformation behavior of both glassy and semicrystalline polymers has been developed. The model explicitly gives true stress dependence on true strain, true strain rate and absolute temperature. At least three experimental true stress-true strain curves including variation with strain rate and temperature are needed to calibrate the eight material coefficients. The sequential procedures to calculate the eight material coefficients in the proposed constitutive model are given in detail.

To validate the proposed constitutive model, experimental data of glassy PMMA and PC at various strain

Table 3. Material Coefficients of the G'Sell-Jonas Model for Polyamide 12 (19).

K (KPa s ^m)	a (K)	w	h	m
2300	870	10	0.8	0.01

Table 4. Material Coefficients for Polyamide 12.

C_1	C_2	m	a (K)	K (MPa · s ^m)	C_3 (s ^m)	C_4	α
0.415	2.867	0.01	870	3.083	3.0	200	6.6

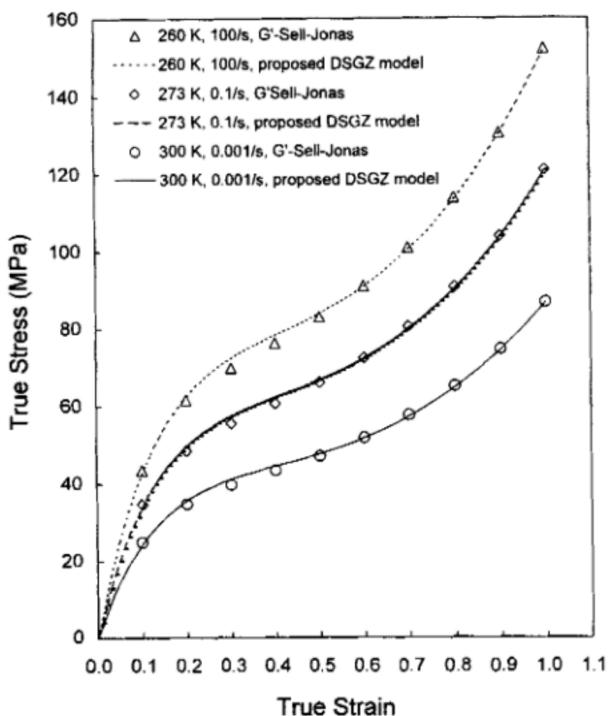


Fig. 6. Comparison of predictions from the G'Sell-Jonas model with that of the proposed DSGZ model for polyamide 12 ($T = 260\text{ K}$, 273 K , 300 K ; $\dot{\epsilon} = 100/\text{s}$, $0.1/\text{s}$, $0.001/\text{s}$).

rates and temperatures are compared with model predictions. The results show excellent comparison between experimental data and model predictions. The predictions of the G'Sell-Jonas model are compared with that of the proposed constitutive model for polyamide 12, a semicrystalline polymer, at various temperatures and strain rates. Predictions from the two models compare very well.

Based on the validation results, it is reasonable to conclude that the new proposed constitutive model can be used to predict the entire range of stress-strain behaviors of both glassy and semicrystalline polymers.

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