Hindawi Publishing Corporation
Journal of Engineering
Volume 2016, Article ID 6315421, 9 pages
http://dx.doi.org/10.1155/2016/6315421

Research Article

Modeling of the Strain Rate Dependency of Polycarbonate's Yield Stress: Evaluation of Four Constitutive Equations

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Received 19 January 2016; Accepted 18 February 2016

Academic Editor: Yuanxin Zhou

The main focus of this paper is in evaluating four constitutive relations which model the strain rate dependency of polymers yield stress. Namely, the two-term power-law, the Ree-Eyring, the cooperative, and the newly modified-Eyring equations are used to fit tensile and compression yield stresses of polycarbonate, which are obtained from the literature. The four equations give good agreement with the experimental data. Despite using only three material constants, the modified-Eyring equation, which considers a strain rate-dependent activation volume, gives slightly worse fit than the three other equations. The two-term power-law and the cooperative equation predict a progressive increase in the strain rate sensitivity of the yield stress. Oppositely, the Ree-Eyring and the modified-Eyring equations show a clear transition between the low and high strain rate ranges. Namely, they predict a linear dependency of the yield stress in terms of the strain rate at the low strain rate range. Crossing a threshold strain rate, the yield stress sensitivity sharply increases as the strain rate increases. Hence, two different behaviors were observed though the four equations fit well the experimental data. More experimental data, mainly at the intermediate strain rate range, are needed to conclude which, of the two behaviors, is more appropriate for polymers.

1. Introduction

Several works have been dealing with the inelastic behavior of metallic materials [1, 2]. In terms of polymers, polycarbonate (PC) is one of the most studied polymers in the open literature.

The temperature and the strain rate sensitivities are extensively characterized in [3–6] and [5–8], respectively. The split Hopkinson pressure bar was largely used to characterize the mechanical properties at high strain rates [9, 10]. Hutchings [11] and Sarva et al. [12] used Taylor's impact method. Prakash and Mehta [13] and Sato et al. [14] used plate impact technique to achieve strain rates in the range of 10^5-$10^6\ s^{-1}$. Li and Lambros [15] and Bjerke et al. [16] used infrared radiation detectors to measure the adiabatic temperature rise. Lee and Kim [17] and Dioh et al. [18] studied the effect of the specimen thickness on the behavior at high strain rate. Weber et al. [19] measured the high strain rate behavior of gamma-irradiated polycarbonate. Trautmann et al. focused on the specimen lubrication at cryogenic temperatures [20].

The yield stress of PC is highly sensitive to temperature and strain rate. It decreases as the temperature increases [21, 22]. Moreover, the temperature sensitivity is more important at low temperatures [4, 5, 23]. Besides, a sharp drop is observed at temperatures above glass transition [24]. On the other hand, the yield stress increases with an increase in strain rate [25–28]. In addition, the strain rate sensitivity is more important at high strain rates [29–32]. The strain rate and the temperature sensitivities are in line with the time-temperature superposition principle as an increase in strain rate has similar effects to a decrease in temperature [23].

Several constitutive relations were proposed in the literature to take into account the temperature and strain rate effects on the yield stress. Eyring [33] argued that yielding is a thermally activated process. This yields a constitutive equation in which the yield stress is linear in terms of the logarithm of strain rate. Similar phenomenological equation (the Johnson-Cook model) was proposed for metals [34, 35]. Ree-Eyring [36] extended Eyring equation by the use of two relaxation processes. Recently, Safari et al. [37] used three

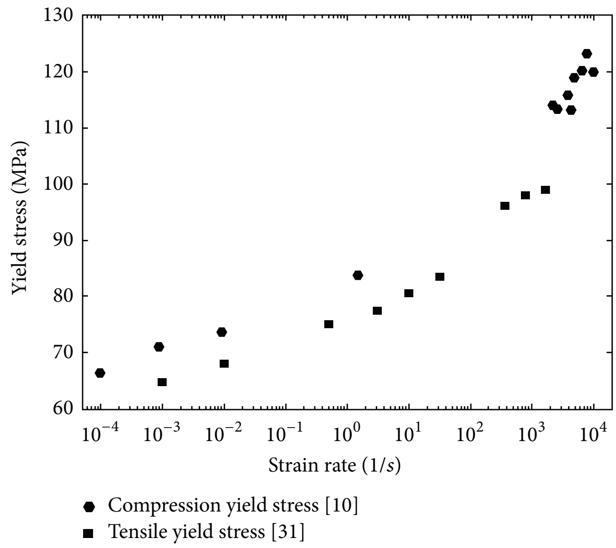


Figure 1: Experimental yield stress of PC.

relaxation processes. Dealing with amorphous polymers, Richeton et al. [24] modified the cooperative model [38, 39] by using an Arrhenius-type law for the horizontal and vertical shifts. Recently, El-Qoubaa and Othman [40–42] modified the one-process Eyring equation [33] by including an activation volume which is decreasing in terms of strain rate. The modified-Eyring equation was used to fit compression yield stress of polyetheretherketone (PEEK).

The temperature and strain rate sensitivity of PC's yield stress were mostly fitted by the Ree-Eyring equation [4, 5, 43, 44] or the cooperative model [9, 45, 46]. Safari et al. [37] argued for the activation of a third relaxation process and then used a modified Ree-Eyring equation.

This paper aims at comparing several constitutive equations predicting the yield stress of polymers. More precisely, the tensile and compression yield stress of PC will be fitted with the two-term power-law [40], the Ree-Eyring [36], the cooperative [24], and the modified-Eyring [41] equations. The error and correlation with experimental data of each model will be calculated.

2. Methodology

2.1. Experimental Data. In this work, we are interested in fitting both the tensile and compression yield stresses. These two stresses are extensively characterized in the literature over wide ranges of strain rates. In terms of the tensile yield stress, we will rely upon the works of Cao et al. [45, 47]. The corresponding experimental data are depicted in Figure 1. In terms of the compression yield stress, we will rely upon the work of Siviour et al. [23]. The corresponding experimental data are plotted also in Figure 1.

2.2. Constitutive Equations. The aim of this work is to compare four constitutive equations which are proposed in the literature to predict the yield stress of polymers. In this section, these constitutive equations are introduced.

2.2.1. Two-Term Power-Law Equation. The power-law equation is an empirical model which assumes that the logarithm of the yield stress linearly increases in terms of the logarithm of the strain rate. This can be written as

$$\sigma_\pi = q\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right)^m, \qquad (1)$$

where σ_π is the yield stress predicted by the power-law equation, $\dot{\varepsilon}_0 = 1/s$ is a normalizing constant, and q and m are two material constants.

El-Qoubaa and Othman [40] showed that using more power-law terms can better approximate the yield stress behavior at high strain rates. Therefore, we will study the accuracy of the two-term power-law equation, which is written as

$$\sigma_{2\pi} = q_1\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right)^{m_1} + q_2\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right)^{m_2}, \qquad (2)$$

where $\sigma_{2\pi}$ is the yield stress predicted by the two-term power-law equation and q_1, q_2, m_1, and m_2 are four material constants which will be determined using the identification procedure detailed in Section 2.3.

2.2.2. Ree-Eyring Equation. Eyring [33] has argued that yielding is a thermally activated process. The following physically based equation is then proposed:

$$\sigma_E = A_\alpha T\left(\ln\left(2C_\alpha\dot{\varepsilon}\right) + \frac{Q_\alpha}{RT}\right), \qquad (3)$$

where σ_E, R, T, and Q_α are the yield stress predicted by the Eyring equation, the universal gas constant, the absolute temperature, and the activation energy, respectively, and A_α and C_α are two material constants.

The Eyring equation predicts a yield stress which is linear in terms of the logarithm of strain rate. However, this is only true up to a threshold strain rate. Actually, there is an increase in strain rate sensitivity at high strain rate.

The Ree-Eyring equation [36] was then introduced to take into account the behavior at high strain rate. The modified Ree-Eyring model assumes that two relaxation processes are activated and is written as follows:

$$\begin{aligned}\sigma_{\mathrm{RE}} = {} & A_\alpha T\left(\ln\left(2C_\alpha\dot{\varepsilon}\right) + \frac{Q_\alpha}{RT}\right) \\ & + A_\beta T\sinh^{-1}\left(C_\beta\dot{\varepsilon}\exp\left(\frac{Q_\beta}{RT}\right)\right),\end{aligned} \qquad (4)$$

where σ_{RE}, Q_α, and Q_β are the yield stress predicted by the Ree-Eyring equation, the activation energy of the α-relaxation, and the activation energy of the β-relaxation, respectively, and, A_α, C_α, A_β, and C_β are four material constants.

In order to simplify the identification procedure, the Ree-Eyring equation is rewritten as follows:

$$\sigma_{\mathrm{RE}} = a_\alpha + b_\alpha \ln\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right) + b_\beta\sinh^{-1}\left(c_\beta\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right), \qquad (5)$$

where a_α, b_α, b_β, and c_β are four material constants which are determined using the identification procedure depicted in Section 2.3 and $\dot{\varepsilon}_0 = 1/s$ is a normalizing constant.

2.2.3. Cooperative Equation. The cooperative equation is based on the works of Fotheringham et al. [38, 39] who introduced the concept that yielding requires a cooperative motion of polymer chain segments [24]. The yield stress given by the cooperative model reads

$$\sigma_c = \sigma_i + \frac{2k_B T}{V}\sinh^{-1}\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}^*}\right)^{1/n}, \tag{6}$$

where σ_c, σ_i, V, k_B, $\dot{\varepsilon}^*$, and n are the yield stress as predicted by the cooperative model, the internal stress, the activation volume, the Boltzmann constant, a characteristic strain rate, and a material parameter, respectively. This equation depends on four material constants (σ_i, V, $\dot{\varepsilon}^*$, and n) which will also be determined using the identification procedure detailed in Section 2.3.

2.2.4. Modified-Eyring Equation. El-Qoubaa and Othman [40–42] pledged for the use of an apparent volume that decreases as the strain rate increases. More precisely, they proposed that the apparent activation volume is given by

$$V^* = V_0 \exp\left(-\sqrt{\frac{\dot{\varepsilon}}{\dot{\varepsilon}_c}}\right), \tag{7}$$

where V^* is the apparent activation volume and V_0 and $\dot{\varepsilon}_c$ are two material constants. The Eyring equation is then modified by including the apparent activation volume:

$$\sigma_{\mathrm{ME}} = \sigma_0 + \frac{kT}{V^*}\ln\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right), \tag{8}$$

where σ_{ME} is the yield stress predicted by the modified-Eyring equation and $\dot{\varepsilon}_0 = 1\ \mathrm{s}^{-1}$ is a normalizing constant.

Substituting (7) in (8) yields

$$\sigma_{\mathrm{ME}} = \sigma_0 + \frac{kT}{V_0}\exp\left(\sqrt{\frac{\dot{\varepsilon}}{\dot{\varepsilon}_c}}\right)\ln\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right). \tag{9}$$

This equation depends on three material constants (σ_0, V_0, and $\dot{\varepsilon}_c$). As for the previous three equations, the material constants will be determined using the identification procedure detailed in the following section.

2.3. Identification. In this section, we are interested in presenting the methodology followed to identify the material constants of each of the four constitutive equations. The main idea is to find a set of material constants that reduces the difference between the experimental yield stresses and the yield stresses obtained by the corresponding constitutive equation.

Let $\dot{E} = (\dot{\varepsilon}_i)$ be a strain rate vector collecting the experimental strain rate values which are obtained from the literature as explained in Section 2.1, where $\dot{\varepsilon}_i$ indicates the strain rate obtained for a test i. We denote by $\widehat{\Sigma} = (\widehat{\sigma}_i)$ the yield stress vector collecting the measured yield stresses where $\widehat{\sigma}_i$ represents the yield stresses obtained for the test i. Likewise, $\Sigma_\chi = (\sigma_{i\chi})$ denotes the yield stress vector collecting the yield stresses calculated by the constitutive equation χ; that is, $\sigma_{i\chi} = \sigma_\chi(\dot{\varepsilon}_i)$. The subscript $_\chi$ can be either $_{2\pi}$, $_{\mathrm{RE}}$, $_{\mathrm{C}}$, or $_{\mathrm{ME}}$ which holds for the two-term power-law equation, the Ree-Eyring equation, the cooperative equation, or the modified-Eyring equation, respectively.

The error vector is written here using the Euclidean norm $\|\ \|_2$ and the maximum norm $\|\ \|_\infty$. The relative error using the Euclidean norm reads

$$\mathrm{EucErr}_\chi = \frac{\left\|\Sigma_\chi - \widehat{\Sigma}\right\|_2}{\left\|\widehat{\Sigma}\right\|_2}, \tag{10}$$

whereas the relative error using the maximum norm is written as

$$\mathrm{MaxErr}_\chi = \frac{\left\|\Sigma_\chi - \widehat{\Sigma}\right\|_\infty}{\left\|\widehat{\Sigma}\right\|_\infty}. \tag{11}$$

The Euclidean norm-based error measures the average difference between the experimental yield stress and the yield stress predicted by the considered constitutive equation. Thus, it is a global measurement of the error [40]. On the other hand, the maximum norm-based error concentrates on the tests where the maximum difference is achieved. Hence, it is a local measurement of the error [40].

In order to take advantage of both the maximum norm and the Euclidean norm, we have defined the optimization cost function, f_χ, as the mean of the Euclidean norm-based error and the maximum norm-based error. This is written as follows:

$$f_\chi\left(\kappa_1^\chi, \kappa_2^\chi, \ldots\right) = \frac{1}{2}\left(\mathrm{EucErr}_\chi + \mathrm{MaxErr}_\chi\right), \tag{12}$$

where $\kappa_1^\chi, \kappa_2^\chi, \ldots$ are the material constants of the constitutive equation χ.

Therefore, a set of material constants will be obtained for each constitutive equation. More precisely, the material constants are given by minimizing the cost function f_χ:

$$\kappa_1^\chi, \kappa_2^\chi, \ldots = \underset{\kappa_1^\chi, \kappa_2^\chi, \ldots}{\operatorname{argmin}} f_\chi\left(\kappa_1^\chi, \kappa_2^\chi, \ldots\right). \tag{13}$$

Subsequently, the error of a constitutive equation χ is calculated as

$$\mathrm{Err}_\chi = f_\chi\left(\kappa_1^\chi, \kappa_2^\chi, \ldots\right). \tag{14}$$

Besides, the correlation coefficient is calculated as

$$\mathrm{Corr}_\chi = \frac{\left\langle \Sigma_\chi, \widehat{\Sigma}\right\rangle}{\sqrt{\left\|\Sigma_\chi\right\|_2 \left\|\widehat{\Sigma}\right\|_2}}, \tag{15}$$

where $\langle \Sigma_\chi, \widehat{\Sigma}\rangle$ is the Euclidean scalar product of vectors $\widehat{\Sigma}$ and Σ_χ calculated using $\kappa_1^\chi, \kappa_2^\chi, \ldots$ as solved in (13).

3. Results and Discussion

3.1. Two-Term Power-Law Equation. The two-term power-law equation was used to fit the compression and tensile yield

Table 1: Material constants, error, and correlation of the two-term power-law equation.

	q_1 (MPa)	m_1	q_2 (MPa)	m_2	$\mathrm{Err}_{2\pi}$ (%)	$\mathrm{Corr}_{2\pi}$
Tension	22.5	0.1002	53.4	0.72×10^{-4}	1.8	0.995
Compression	19.9	0.1195	61.7	2.98×10^{-4}	2.2	0.997

Table 2: Material constants, activation volumes, error, and correlation of the Ree-Eyring equation.

	a_α (MPa)	b_α (MPa)	b_β (MPa)	c_β	V_α (nm^3)	V_β (nm^3)	$\mathrm{Err}_{\mathrm{RE}}$ (%)	$\mathrm{Corr}_{\mathrm{RE}}$
Tension	75.5	1.80	1.46	0.233	2.26	2.79	2.2	0.993
Compression	81.5	1.59	3.02	0.161	2.57	1.35	2.3	0.997

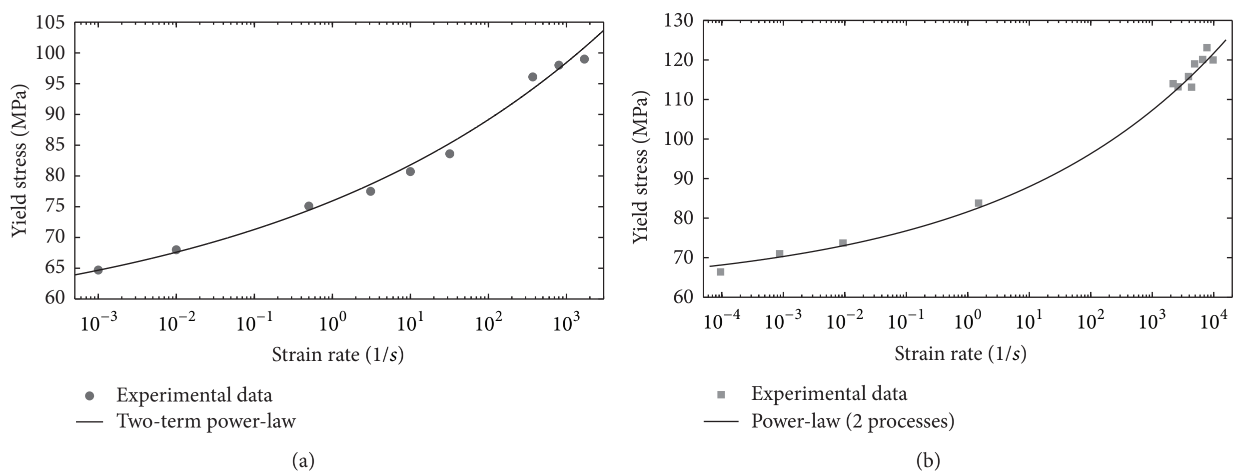


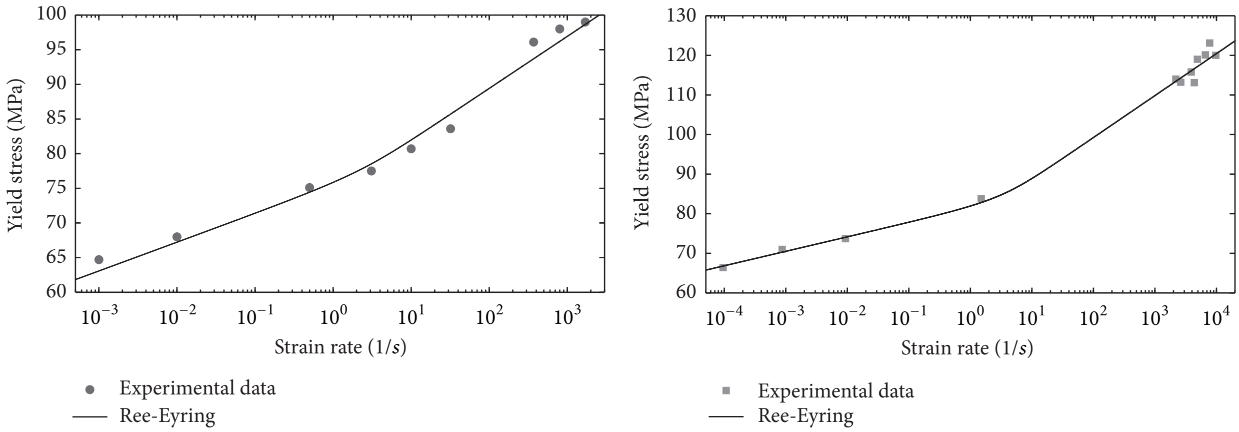
Figure 2: Experimental yield stress of PC fitted by the two-term power-law equation: (a) tension and (b) compression.

stress (Figure 2). Good matching is observed between the experimental yield stress and the yield stress predicted by the two-term power-law equation on the whole strain rate range. For both tensile and compression yield stress-strain rate curves, the slope progressively increases as the strain rate increases. In other words, the strain rate sensitivity of the yield stress progressively increases and no pronounced transition is recorded between the low and high strain rate ranges.

The identified material parameters are presented in Table 1 which includes also the values of errors and correlations. As expected from Figure 2, the errors are very low (1.8% and 2.2% for tension and compression, resp.) and the correlation coefficients are almost equal to 1. The material constants of tension have the same order of magnitude of those obtained in compression. This is mainly explained by the fact that tensile yield stress is of the same order of magnitude and follows the same tendency as the compressive yield stress. It is not easy to interpret m_1 and m_2. On the other hand, $q_1 + q_2$ is equal to 75.9 MPa (tension) and 81.6 MPa (compression) which are almost the tensile and compression yield stresses, respectively, at a strain rate of $1/s$.

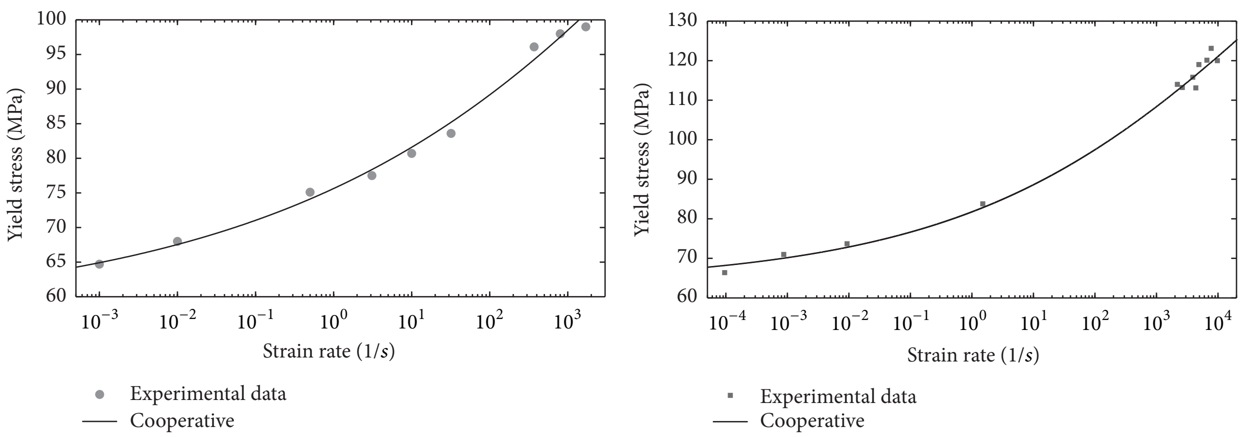
3.2. Ree-Eyring Equation. The Ree-Eyring equation gives also good fit of both the tensile and compression experimental yield stresses on the entire strain rate range investigated here (Figure 3). The yield stress predicted by the Ree-Eyring equation increases linearly in terms of strain rate in the low strain rate range. A transition occurs in the interval [1–10/s] for both tension and compression. Crossing this transition, the slope of the yield stress-strain rate curves starts to increase as the strain rate increases. Owing to the theory of Ree-Eyring, second relaxation, namely, the β-relaxation, is then activated.

Table 2 collects the material constants, the error, and the correlation coefficient of the Ree-Eyring equation. Besides, it includes the activation volumes of the α- and β-relaxations, which are written as $V_i = k_B T / b_i$; recall that k_B and T are the Boltzmann constant and the absolute temperature, respectively. In tension and compression, the errors are less than 2.3% and the correlation coefficient is almost equal to 1 which means highly good matching between the experimental yield stresses and the ones predicted by the Ree-Eyring equation. The constant a_α is equal to 75.5 MPa (tension) and 81.5 MPa (compression) which are approximately the same values of $q_1 + q_2$. Actually, a_α and $q_1 + q_2$ interpret the yield stress at a strain rate of $1\,\mathrm{s}^{-1}$. The activation volume has the order of magnitude of nm^3. This gives a characteristic length of some nm, which is much greater than interatomic distances and much lower than the lengths of macromolecules. In the case of tension, $V_1 > V_2$, whereas it is the opposite in the case of compression. In this latter case, $V_1 \approx 1.9 V_2$. This is



(a) (b)

FIGURE 3: Experimental yield stress of PC fitted by the Ree-Eyring equation: (a) tension and (b) compression.



(a) (b)

FIGURE 4: Experimental yield stress of PC fitted by the cooperative equation: (a) tension and (b) compression.

comparable to what was observed by El-Qoubaa and Othman [40] where $V_1 \approx 1.7V_2$ for PEEK.

3.3. Cooperative Equation. The cooperative equation fits well the tensile and compression experimental yield stresses. The error is very low (1.7% in tension and 2.3% in compression) and the correlation coefficient is almost equal to one (0.995 in tension and 0.997 in compression).

The cooperative equation predicts a strain rate sensitivity of the yield stress which is similar to the one predicted by the two-term power-law equation (Figure 4). Namely, the slopes of the yield stress-strain rate curves progressively increase as the strain rate increases. No clear transition appears, which is different from the behavior predicted by the Ree-Eyring equation.

The identified material parameters are presented in Table 3. The constant n is equal to 7.82 in tension and 6.79 in compression. Dealing with compression, Richeton et al. [24] and Cao et al. [45] have reported 5.88 and 3.01, respectively. The activation volume V is here found to be

TABLE 3: Material constants, error, and correlation of the cooperative equation.

	σ_i (MPa)	V (nm^3)	$\dot{\varepsilon}^*$ (s^{-1})	n	Err$_C$ (%)	Corr$_C$
Tension	57.2	0.10	3465	7.82	1.7	0.995
Compression	63.4	0.17	513	6.79	2.3	0.997

equal to about 10^{-28} and 1.7×10^{-28} m^3 in tension and compression, respectively, whereas Richeton et al. [24] and Yu et al. [46] have obtained 5.2×10^{-29} and 4.2×10^{-30} m^3, respectively, in compression. The constant σ_i interprets the limit of the yield stress as the strain rate vanishes. It is equal to 57.2 and 63.4 MPa in tension and compression, respectively. A value of ~74 MPa can be obtained from the material constants reported in [24, 46]. There is a substantial difference between the identified material constants obtained in this work, or those identified in [24] or in [46]. Unfortunately, there is no available data for material constants of the other constitutive equations to compare with.

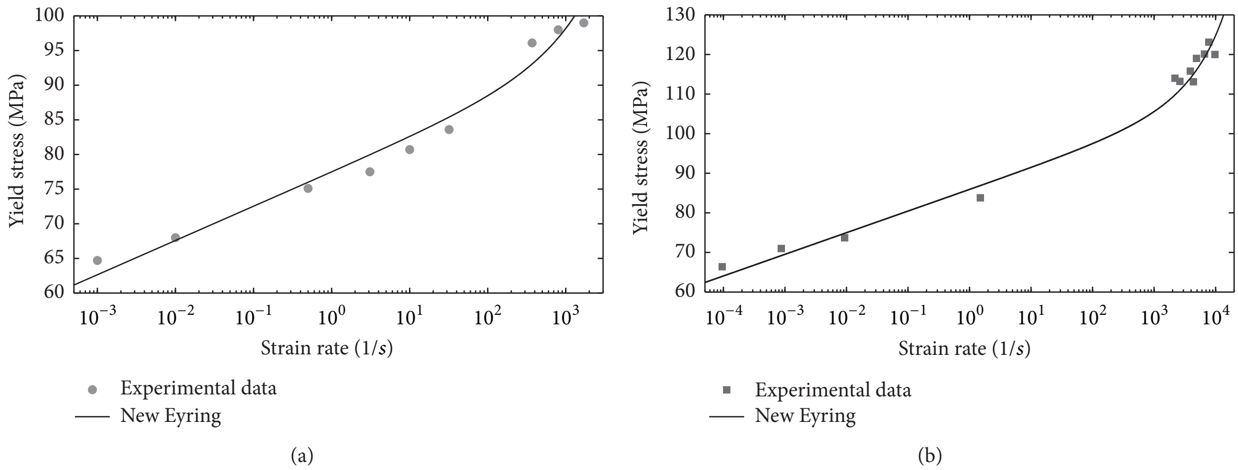


FIGURE 5: Experimental yield stress of PC fitted by the modified-Eyring equation: (a) tension and (b) compression.

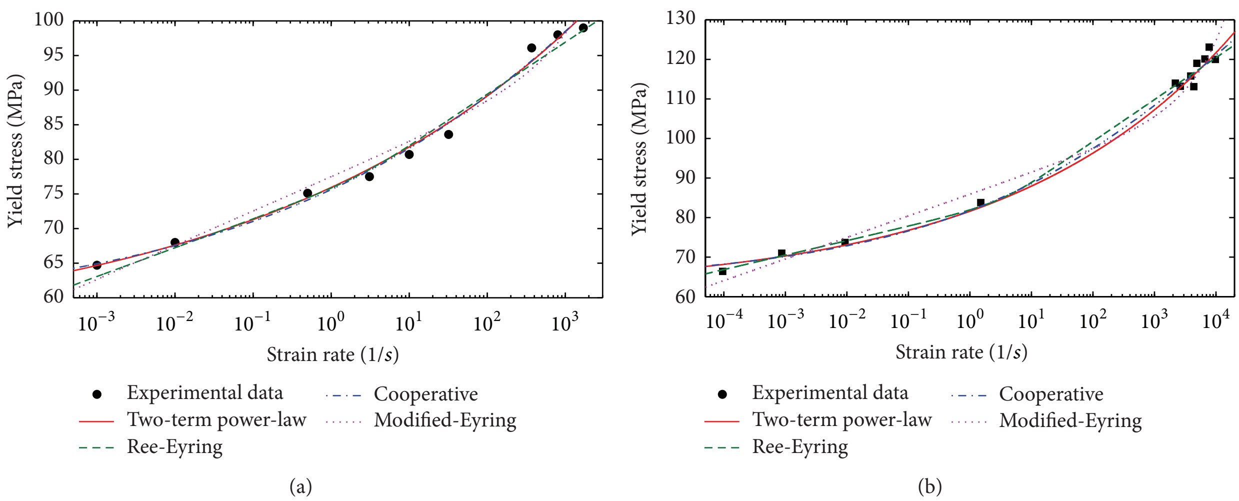


FIGURE 6: Comparison of the four equations: (a) tension and (b) compression.

TABLE 4: Material constants, error, and correlation of the modified-Eyring equation.

	σ_0 (MPa)	V_0 (nm^3)	$\dot{\varepsilon}_c$ (s^{-1})	Err$_{\mathrm{ME}}$ (%)	Corr$_{\mathrm{ME}}$
Tension	77.5	1.89	9036	2.8	0.987
Compression	85.9	1.72	3020	3.1	0.993

3.4. Modified-Eyring Equation. The modified-Eyring equation fits also well the tensile and compression experimental yield stresses. It does slightly worse than the three other equations. However, it needs only three material constants whereas the others use four material constants each. The error is 2.8% and 3.1% in tension and compression, respectively, and the correlation coefficient is 0.987 and 0.993 in tension and compression, respectively (Table 4).

The strain rate sensitivity of the yield stress predicted by the modified-Eyring equation is close to the one predicted by the Ree-Eyring equation. More precisely, a clear strain rate transition is observed (Figure 5). This transition separates the low strain rate range where the slope of the curve is constant and the high strain rate range where the slope of the curve increases with increasing strain rate.

Three material constants are identified in tension and compression (Table 4). σ_0 interprets the yield stress at $1/s$ of strain rate. It is here obtained equal to 77.5 and 85.9 MPa in tension and compression, respectively. These values are slightly higher than those predicted by the two-term power-law and the Ree-Eyring equations which predict 76 and 82 MPa, respectively. The activation volume V_0 is 1.89×10^{-27} in tension and 1.72×10^{-27} m^3 in compression, which are in the same order of magnitude as the activation volume V_1 of the first relaxation process of the Ree-Eyring equation. Finally, $\dot{\varepsilon}_c$ is equal to 9036 and 3020 s^{-1} in tension and compression, respectively. This means that the transition between the low and high strain rate regimes occurs at around 90 and 30 s^{-1}, respectively.

3.5. Comparison. The four constitutive equations fit well the experimental data in tension as well as in compression (Figures 6 and 7). Though the modified-Eyring equation gives

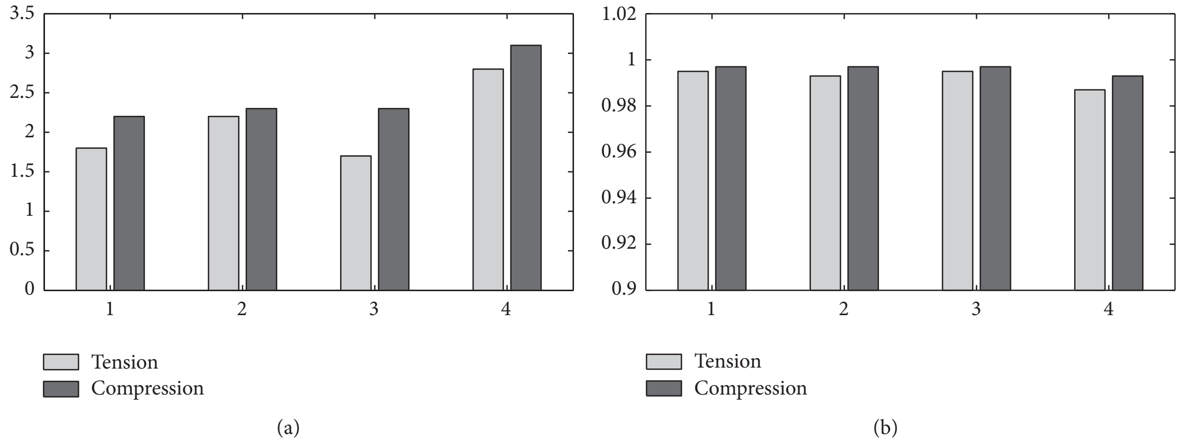


Figure 7: Comparison of the four equations: (a) error (%) and (b) correlation.

slightly higher errors and slightly worse correlation than the three other equations, it uses only three material constants. This equation can be accepted as a trade-off to reduce the number of material constants while keeping a reasonable error.

Even though the four constitutive equations match well the experimental data, they give different behaviors mainly at the intermediate strain rate range (Figure 6). While the two-term power-law and the cooperative equations predict a progressive increase of the yield stress-strain rate slope as the strain rate increases, the two other equations, namely, the Ree-Eyring and the modified-Eyring equations, predict a clear transition between the low strain rate and the high strain rate regimes. More experimental data at the intermediate strain rate range (1–100/s) should help to conclude which of the two behaviors is more appropriate.

4. Conclusion

The two-term power-law, the Ree-Eyring, the cooperative, and the modified-Eyring constitutive relations were used to fit the tensile and compression yield stress of polycarbonate. The predicted yield stresses, by the four equations, are in highly good agreement with the experimental yield stresses. The strain rate sensitivity of the yield stress fell into two behaviors, either a progressive increase in terms of strain rate or an increase with a transition in the intermediate strain rate range. Thus, more experimental data, specifically at the medium strain rate range, are needed to decide which of the two behaviors has to be retained.

Competing Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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