

Review Article

Shengbo Ling, Zhen Wu*, and Jie Mei

Comparison and review of classical and machine learning-based constitutive models for polymers used in aeronautical thermoplastic composites

<https://doi.org/10.1515/rams-2023-0107>

received January 07, 2023; accepted July 24, 2023

Abstract: Most of the stress–strain relationships of thermoplastic polymers for aeronautical composites tend to be nonlinear and sensitive to strain rate and temperature, so accurate constitutive models are urgently required. Classical and machine learning-based constitutive models for thermoplastic polymers are compared and discussed. In addition, some typical models have been recovered and compared by authors to evaluate the performance of classical and machine learning-based constitutive models, so that the advantages and shortcomings of these models can be demonstrated. By reviewing constitutive models, it was found that the equations of physical constitutive models are derived according to thermodynamical principles, so the physical constitutive models can describe the deformation mechanism at the microscopic level. The phenomenological constitutive models may combine the macroscopic phenomena and theories of physical models, and good performance and wide range of applications can be realized. In addition, phenomenological constitutive models combined with machine learning algorithms have attracted attentions of investigators, and these models perform well in predicting the stress–strain relationships. In the future, the constitutive models combining the theories of physical constitutive models, phenomenological constitutive models, and machine learning algorithms will be increasingly attractive as some challenging issues are effectively addressed.

Keywords: constitutive model, thermoplastic polymers, machine learning

1 Introduction

Composites can be divided into two categories according to the matrix properties: thermoplastic composites and thermosetting composites. Thermosetting composites have been widely used in the aeronautical and astronautical industries due to the extreme pursuit of material performance and weight [1]. Aircrafts such as Boeing-787 [2] and A350 [3] are extensively made of composites for their fuselages and wings, with the percentage exceeding 50%. Nevertheless, thermosetting composites possess the following defects: poor impact toughness, local damage is difficult to repair, long molding cycle, difficult to recycle, and so on [4]. In the past, the performance of thermoset composites was considered to outperform that of thermoplastic composites, but the situation has changed with the application of high-performance thermoplastic resins now. Compared with thermosetting composites, there are many advantages to high-performance thermoplastic composites, such as lower density, high toughness, high impact resistance and damage tolerance [5], high resistance to heat and humid, short molding cycle, and easy to repair and recycle [6,7]. Due to the concept of environmental protection, thermoplastic composites thus attract the interest of researchers and engineers, and they have been applied to the aerospace industry, automotive industry, electronic industry, and medical industry gradually [8].

Thermoplastic polymers which act as the matrix of thermoplastic composites, can be divided into general plastics and high-performance engineering plastics in terms of performance. Polyethylene (PE), polypropylene (PP), and acrylonitrile butadiene styrene (ABS) can be classified as general plastics, while polyamide (PA), polycarbonate (PC), polyether ether ketone (PEEK), polyetherimide (PEI), polyether sulfone (PES), and polyphenylene sulfide (PPS) fall within the latter [9]. The chemical reaction of side chain groups will not take place during heating for the macromolecular chain of thermoplastic polymers, but only physical changes occur. Therefore, the thermoplastics can be

* **Corresponding author: Zhen Wu**, School of Aeronautics, Northwestern Polytechnical University, Xi'an 710072, China, e-mail: wuzhenhk@nwpu.edu.cn

Shengbo Ling, Jie Mei: School of Aeronautics, Northwestern Polytechnical University, Xi'an 710072, China

softened, melted, formed, welded by heating, and solidified when cooled.

Thermoplastic composites have been used in many structures of aircraft, such as Airbus A380, A350, A400M, Boeing787, Gulfstream jets G450, G550, and G650, *etc.* [10]. As the secondary bearing components, thermoplastic composites are competent usually. The floor panels in the A400M were manufactured with carbon fiber/PPS and glass fiber/PPS composites. The rudder and elevator of G650 were manufactured with carbon fiber/PEI and carbon fiber/PPS composites. Carbon fiber/PPS composites are used in many structures of A350 including connectors. These structures work under static load mostly, and constitutive models with small strain, low strain rate, and small range of temperatures are thus needed. However, some other structures may be exposed to conditions with extreme loading. The leading edge of aircraft A380 was manufactured with glass fiber/PPS composites. The PC was usually used to manufacture the canopy of fighters. Since the safety after a bird or hailstone strike must be considered for both the leading edge of the wing and canopy, the impact behavior and the corresponding constitutive models with a high strain rate of composite structures must be studied [11].

Although thermoplastic polymers have been used in engineering, it requires addressing a key point that the properties of such materials are usually more complex than metals or thermosetting polymers. Figure 1 presents a typical stress–strain curve of a thermoplastic polymer. The whole deformation process can be divided into five phases generally: linear VE deformation, nonlinear VE deformation, yield, strain softening, and strain hardening.

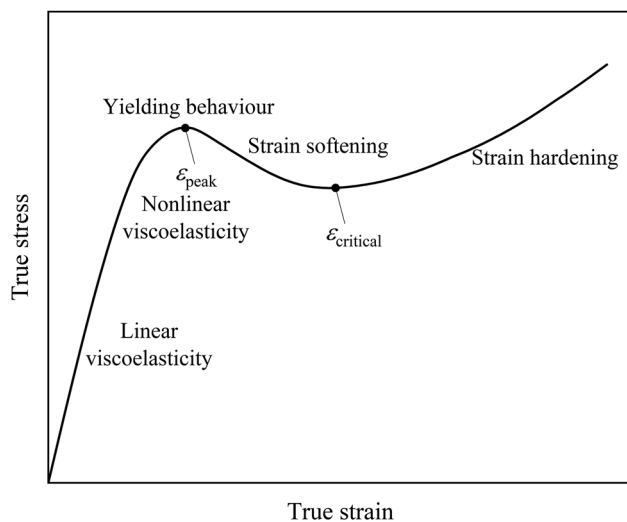


Figure 1: Typical deformation behavior of a thermoplastic polymer [12].

The deformation is irrecoverable after yielding for most of the polymers, but the plastic deformation of glassy polymers is recoverable at a temperature above the glass transition temperature (T_g) [12]. For the semi-crystalline polymers commonly used in thermoplastic composites, the behaviors of large deformation such as strain softening and strain hardening are closely related to the crystallization degree and crosslinking of molecular chains. In addition, the stress–strain response will be affected by the strain rate, temperature, time, and pressure significantly, and the nonlinear behavior of thermoplastic polymers is further exacerbated [13]. If the mechanical behavior of materials is not clear, it is largely difficult to design, manufacture, apply, and monitor the thermoplastic composites. Therefore, in addition to experimental testing, numerical simulation adopting an effective constitutive model plays an important role in predicting the behavior of materials. Further, one of the most important works in the study of thermoplastic polymers is to establish a constitutive model that can accurately reflect such behavior.

There are two purposes for this study:

- 1) To sort, classify, and review the development of nonlinear constitutive models for polymers of aeronautical composites.
- 2) To evaluate the performance of traditional constitutive models and machine learning-based constitutive models, and explore the future direction for the development. It is important to note that this article does not attempt to cover all constitutive models, but rather introduces nonlinear constitutive models commonly used in aeronautical thermoplastic polymers and machine learning-based constitutive models particularly. By reviewing literature, evaluation and comparison between the classical constitutive models and the machine learning-based constitutive models are rarely found. Therefore, evaluation and comparison between the classical constitutive models and the machine learning-based constitutive models were presented for the first time in the proposed work. Thereby, this work fills in the research gap of the published literature. As the main body, Section 2 presents the detailed classification and development of constitutive models. In this section, constitutive models are introduced in three parts: physical models, phenomenological models, and machine learning-based models. Each part is sorted and presented by several models, and the typical model is simulated to evaluate its performance. Section 3 gives the discussion, conclusion, and prospects for the development of constitutive models.

2 Constitutive models for thermoplastic polymers

In order to improve the performance of aircraft, aeronautical composite structures are required to work in the complex thermal-mechanical environments. In particular, the properties of semi-crystalline polymers are easily affected by temperature and strain rate. The experimental testing cannot cover all the working conditions entirely for those materials that are sensitive to strain rate and temperature, although the tests are the most direct way to study the properties of thermoplastic polymers. Therefore, it is crucial to establish the constitutive models of thermoplastic polymers.

In this section, the nonlinear constitutive models applied to thermoplastic polymers for aeronautical composites are classified according to diverse points of view to understand the issues, and the characteristics of each type of constitutive models are investigated in detail. The constitutive models can be divided into two categories: physical constitutive models and phenomenological constitutive models as shown in Figure 2, according to the theoretical basis given in previous studies [14–16]. The so-called physical constitutive models aim to study the effects of amorphous and crystalline phases on the mechanical behaviors of materials, which describes the macroscopic behavior of materials based on the microstructures. Most of the physical constitutive models were almost derived from the theories of Haward and Thackray [17] and Edwards and Vilgis [18], and all parameters in physical constitutive models have physical meanings. However, the phenomenological constitutive models

describe the experimental phenomenon by using mathematical formulations with or without some concepts of physical constitutive models. Therefore, phenomenological constitutive models do not mainly focus on the natural deformation mechanism of materials. In general, the phenomenological constitutive models mainly include the elasto-plastic models, the viscoelastic (VE) models, the hyperelastic models, the viscoplastic (VP) models, the multi-mode models, the models with damage, and the machine learning-based phenomenological models, which are shown in Figure 2.

2.1 Physical constitutive models

As mentioned above, the physical constitutive models inspired by microstructure believe that crosslinking, molecular weights, and crystallinity degrees are the dominant factors in material properties. The crystalline phase is considered to have a significant effect on the mechanical properties of materials, especially at the stage of small strain. As for the amorphous phase, it will affect the deformation at the stage of high strain and post-yielding [19]. In this regard, the mechanical properties of materials can be forecasted by simulating the behavior of amorphous and crystalline phases. This research started in the middle of the last century, and most of the existing physical constitutive models were developed based on the theory of Haward and Thackray [17] and Edwards and Vilgis [18] from the last century. These two theories and corresponding models will

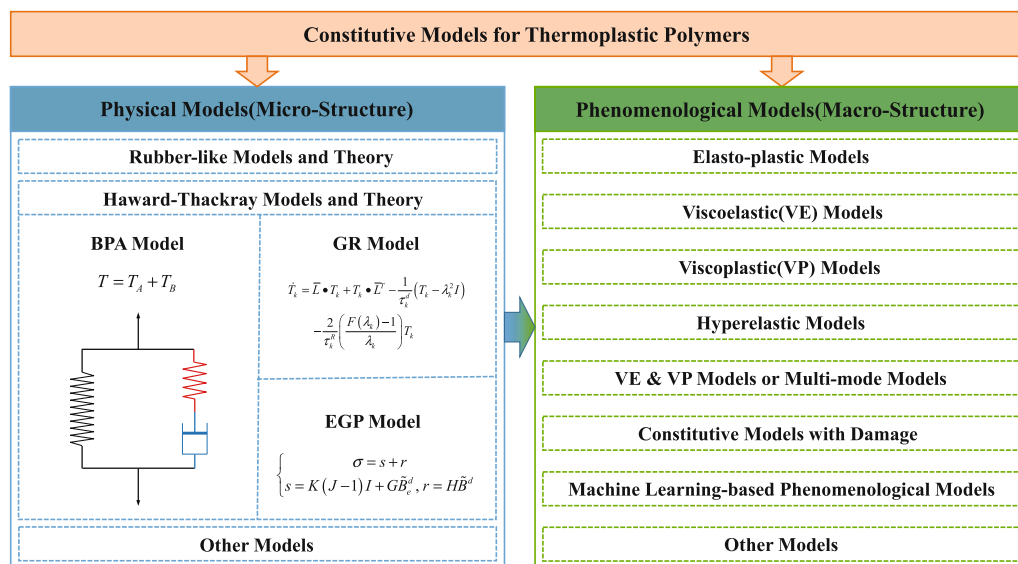


Figure 2: Classification of constitutive models for thermoplastic polymers.

be discussed, respectively, although researchers often refer to them together.

2.1.1 Rubber-like models

This kind of models are developed through the elastic theory of rubber, so they could be called rubber-like models. Studies of rubber elasticity began as early as the early twentieth century, and James and Guth [20] discussed an idea of effective internal pressure, and the typical stress–strain curve for rubber is displayed in Figure 3. They assumed that the polymer chains move through each other only through crosslinks, and the whole system is prevented from collapsing by the assumption of repulsive forces which can generate a bulk modulus. In addition, a simplified model for the bulk rubber was proposed, which consists of a network of the idealized flexible chains extending through the material and fluid filling it. Moreover, the bounding surfaces are in equilibrium under all the forces acting on them (internal pressure, pull of the molecular network, and any external forces). Because of the changing entropy, a good agreement was obtained by comparing the stress–strain curves for bulk rubber at a constant temperature. The general theory proposed by James and Guth [20] provides a basis for the treatment of other physical properties of stretched rubber.

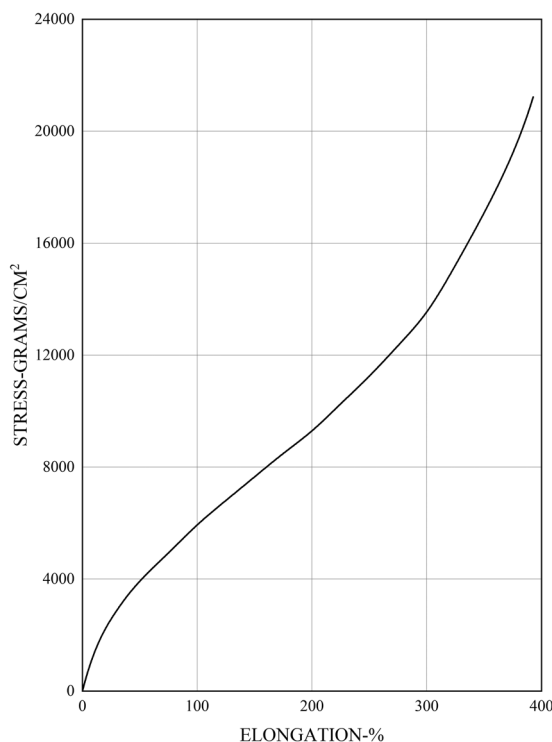


Figure 3: Typical stress–strain curve for rubber [20].

Ball *et al.* [21] indicated that the assumption by James and Guth [20] ignored the repulsive forces which generate a bulk modulus. They proposed that entanglements can be simulated by links that make a sliding contact between polymer networks. The contribution of entanglement to the free energy of shear is given in Eq. (1), where λ_i is the Cartesian extension ratio and η is a measure of the freedom of a link to slide compared with the freedom of movement of a chain.

$$\tilde{F} = \frac{1}{2}kT \sum_i \frac{\lambda_i^2(1+\eta)}{1+\eta\lambda_i^2} + kT \sum \log(1+\eta\lambda_i^2). \quad (1)$$

Edwards and Vilgis [18] proposed an elasticity theory of rubber based on the concept of entanglements in 1986, which is an improvement of the work presented by Ball *et al.* [21]. They found that the molecular mechanism of stretching is dominated by the slippage of chains for small deformation, and the hardening of the rubber at high deformation attributes to the inextensibility as described by the tube concept. The constitutive model is able to explain the deformation behavior of rubbers over the total range of deformation, by comparing the free energy of deformation with the experiment. The free energy is shown in Eq. (2), where α is a measure of the inextensibility and η of the slippage, N_c is the number of crosslinks, and N_s denotes the number of slip links.

$$\begin{aligned} F &= F_c + F_s \\ F_c &= \frac{1}{2}N_c \left[\frac{\sum_{i=1}^3 (1-\alpha^2)\lambda_i^2}{1-\alpha^2 \sum \lambda_i^2} - \log \left(1 - \alpha^2 \sum_{i=1}^3 \lambda_i^2 \right) \right] \\ F_s &= \frac{1}{2}N_s \left[\sum_{i=1}^3 \left\{ \frac{\lambda_i^2(1+\eta)(1-\alpha^2)}{(1+\eta\lambda_i^2)(1-\alpha^2 \sum \lambda_i^2)} + \log(1+\eta\lambda_i^2) \right\} \right. \\ &\quad \left. - \log(1-\alpha^2 \sum \lambda_i^2) \right] \end{aligned} \quad (2)$$

The Edwards–Vilgis model was the most representative one in this type of approach, and it was also known as the network model. It was initially applied to rubbers, then extended and applied to amorphous glassy polymers or semi-crystalline polymers by other researchers. Van Ruiten *et al.* [22] applied the Edwards–Vilgis model for the analysis of melt-spun PA 4.6 fibers in 2001. They tested the maximum attainable tenacity of drawn yarns under given drawing conditions, and compared the experimental results with the predictions of the Edwards–Vilgis model. It was shown that the Edwards–Vilgis rubber-elastic model can describe the network deformational behavior of the as-spun yarns over a wide range of draw ratios. Based on the rubber elastic model, Sweeney *et al.* [23] suggested a large deformation and rate-dependent model in 2002. In their

theory, the rigid spheres are embedded to introduce strain concentration, which is similar to that caused by hard crystalline regions. Besides, the dependence on time and rate was introduced *via* the shear stress-driven diminution of the sphere radii. Then, the model was applied to the high temperature stretching of PE through ABAQUS software (a finite element (FE) solver). Constitutive models introduced in this section are listed in Table A1 of the Appendix.

2.1.2 Models based on Haward–Thackray theory

This type of model takes the theory of Ree and Eyring [24] as the foundation. Ree and Eyring [24] presented a general relationship between the yield stress, the strain rate, and the temperature applicable to polymers as early as 1955, and Ree and Eyring's theory can account for the relaxation process of viscous flow. Since then, researchers have come up with models based on this theory.

Haward and Thackray [17] represented the interactions between molecules through springs and dashpots in 1968. This model is intended to provide a semi-empirical approach to the large plastic deformations at the yield point in low-temperature where the temperature is below T_g . Nevertheless, it is not suitable for describing the creep behavior at extensions of up to 5% or nonlinearity in the initial Hookean modulus. Haward–Thackray model contains a Hookean spring, an Eyring dashpot, and a Langevin spring with an ultimate limiting network strain as shown in Figure 4. The above Hookean spring represents the constant Hookean modulus, which describes the linear elastic deformation of the amorphous phase. The Eyring

dashpot represents the VP character, which describes the rate-dependent macroscopic yield deformation of the amorphous phase. The Langevin spring represents the limited elastic extensibility, which describes the strain hardening result from the change in configuration entropy caused by the orientation rearrangement of molecular chain entanglement. The limited elastic extensibility is based either on the first-order process model, or the conventional Langevin formula as used to describe the highly elastic extension of rubbers. Practice shows that the model can be used to characterize the intermolecular elastic interaction, strain-rate dependent yield behavior, and strain-strengthening behavior in the post-yield stage of amorphous polymers.

Haward–Thackray theory is a milestone and many subsequent models are inspired by it to varying degrees. On this basis, a large number of constitutive models are proposed. We introduce the development profile with three typical types of models and their derivatives: the BPA model, the glass-rubber (GR) model, and the Eindhoven glassy polymer (EGP) model.

2.1.2.1 BPA model and its derivatives

Among numerous research groups, Boyce *et al.* [25] have made an outstanding contribution. Boyce *et al.* [25] proposed a three-dimensional constitutive model for glassy polymer on the basis of the Haward–Thackray model in 1988, also known as the BPA model. Linear spring, VP dashpot, and Langevin spring are contained in the BPA model as shown in Figure 5, and the linear spring and VP dashpot are connected in series to construct the Maxwell element. BPA model assumes that the deformation resistance of amorphous polymers may be decomposed into intermolecular deformation resistance and entropy deformation resistance. The intermolecular deformation resistance is related to the rotation of molecular chain segments

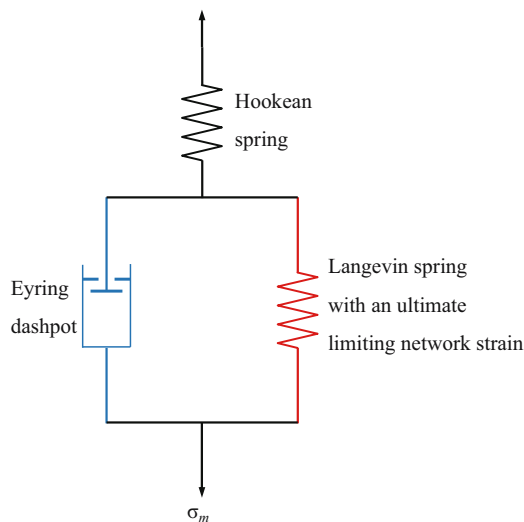


Figure 4: Haward–Thackray model system [17].

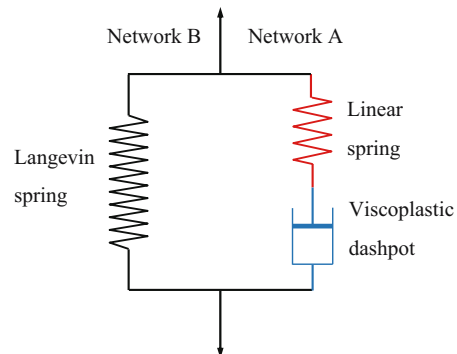


Figure 5: BPA model [25].

and determines the elastic deformation before yield. The entropy deformation resistance is related to the rearrangement of molecular chain segments, and will increase due to the continuous rearrangement of molecular chain segments during the post-yield process. Strain hardening is controlled by entropy deformation resistance, while strain softening is controlled by both intermolecular deformation resistance and entropy deformation resistance. BPA model can achieve good results by comparing with the experimental data of PMMA.

2.1.2.2 GR models and its derivatives

Buckley and Jones [26] proposed the GR constitutive model which displays the glassy response at low temperatures and short time-scales, and rubber-like response at high temperatures and long time-scales. The model employs the linear elasticity, Eyring viscous flow, and the Edwards–Vilgis entropy function to reflect the elastic bond distortion stress–strain law and flow model, and the conformational entropy function, respectively. This model can be argued by combining the theories of Edwards and Vilgis theory [18] and Haward and Thackray theory [17]. The model proposed by Wu and Buckley [27] included a distribution of relaxation times and a semiempirical representation of strain-induced structural rejuvenation based on the GR model for amorphous polymers. The model, constructed by a series of nonlinear partial differential equations, was suitable for the research of yield and plastic flow of glassy polymers. De Focatiis *et al.* [28] proposed a new glass-melt constitutive model to describe the mechanical behavior of large deformation in both the glass state and molten state of polymers in 2010. The model consists of a set of ROLIEPOLY equations that govern the conformational entropy elasticity of the polymer and its relaxation by tube diffusion in the melt, and a multi-mode glassy constitutive model for deformation. It was found to be quantitatively successful in capturing the material rheology in the melt and uniaxial compression experiments deep in the glassy state.

2.1.2.3 EGP model and its derivatives

The initial EGP model was a one-mode model, and it did well in forecasting the plastic flow near yield, but it was poor in the nonlinear VE pre-yield region. Tervoort *et al.* [29] proposed a model based on “compressible-Leonov model” (CLM) [30], which was a three-dimensional constitutive model for the finite elasto-viscoplastic deformation of polymers. The CLM can also be regarded as a single Maxwell model with a relaxation time. The elastic volume response in this model was rigorously separated from the

elasto-viscoplastic isochoric deformation, and it could be extended to include a spectrum of relaxation times. They compared the model predictions with the experimental results obtained from the homogeneous uniaxial tensile test and the homogeneous plane-stress shear test of PC, and received the expected results. Govaert *et al.* [31] extended the CLM to describe the mechanical behavior of large strain in glassy polymers, and only one relaxation time was involved in this model. Strain rate, temperature, pressure-dependent yield, strain softening, and strain-hardening phenomena are well incorporated into the model. Then, Govaert and Tervoort [32] introduced the physical quantities into the strain hardening stage, which are related to temperature and molecular dependence. Nevertheless, only one relaxation time is involved in the model, so it is difficult to capture the nonlinear VE pre-yield behavior accurately. As mentioned above, the one-mode EGP model was not suitable for the prediction of non-linear VE behavior in the pre-yield region. Van Breemen *et al.* [33] extended the one-mode EGP model to a multi-mode model by using a spectrum of relaxation times. Their tests proved that the multi-mode model is necessary for micro-indentation or notched impact deformation, because these macroscopic responses to such deformations were controlled by local non-homogeneous deformations.

The theories proposed by Haward and Thackray [17] and Edwards and Vilgis [18] provided a good foundation for physical constitutive models, so a large number of models had been derived according to their ideas. Furthermore, physical models can describe the deformation mechanism of polymers, which can be explicitly traced to the microscopic level. Constitutive models introduced in this section are listed in Table A2 of the Appendix.

2.2 Phenomenological constitutive models

The phenomenological constitutive models have also been developed for a long time, and a mass of models have been proposed, improved, and applied. Phenomenological constitutive models focus on portraying the phenomena results from tests, rather than the deformation mechanism of materials at the molecular level. The forms and methodologies of phenomenological constitutive models are consequently varied, and some models purely express the stress–strain response, while some models refer to the theory of physical constitutive models. The readers might be confused because some models were developed based on physical theories, while the authors classified them as phenomenological models. It is because the experimental phenomenon is taken

as the starting point in these models, and physical models are subsequently introduced phenomenally. This work attempts to classify the phenomenological models into several categories according to their application: elasto-plastic models, VE models, VP models, hyperelastic models, multi-mode models, models with damage, and others, and the summary of these models can refer to Table A3 in the Appendix.

2.2.1 Elasto-plastic models

G'sell *et al.* [34,35] proposed a phenomenological model through a series of tests of polyvinyl chloride (PVC) and high-density polyethylene (HDPE) in 1979. The stress can be expressed as a function of strain, strain rate, and temperature in Eq. (3).

$$\sigma(\varepsilon, \dot{\varepsilon}) = K e^{h\varepsilon^2} (1 - e^{-W\varepsilon}) \dot{\varepsilon}^m e^{e/T}, \quad (3)$$

where σ , ε , $\dot{\varepsilon}$, and T denote stress, strain, strain rate, and temperature, respectively, and K , W , and m are the model parameters. The G'sell–Jonas model is suitable for the deformation prediction of materials at a low strain rate (10^{-1} – 10^{-4} s $^{-1}$). Johnson and Cook [36,37] proposed a famous Johnson–Cook model (JC model) when studying the deformation of metal under high strain rate and high temperature in 1985 as shown in Eq. (4).

$$\sigma(\varepsilon^p, \dot{\varepsilon}^p, T) = [A + B(\varepsilon^p)^n] \left[1 + C \ln \left(\frac{\dot{\varepsilon}^p}{\dot{\varepsilon}_0} \right) \right] \times \left[1 - \left(\frac{T - T_{\text{ref}}}{T_m - T_{\text{ref}}} \right) \right], \quad (4)$$

where σ , ε^p , $\dot{\varepsilon}^p$, $\dot{\varepsilon}_0$, T , T_m , and T_{ref} signify stress, plastic strain, plastic strain rate, reference strain rate, temperature, melting temperature, and reference temperature, respectively, and there are only five model parameters in the JC model (A , B , C , n , and m). JC model is a typical model, and it reflects the behavior of elasticity, strain hardening, and strain rate hardening for metals. However, researchers subsequently found that the JC model was also suitable for predicting the deformation of semi-crystalline polymers. Garcia-Gonzalez *et al.* [38] applied the JC model to the simulation of impact for PEEK and Ti6Al4V titanium alloy. In addition, the JC model was constantly extended in order to get better performance. For example, Chen *et al.* [39,40] modified the original JC model into the following form shown in Eq. (5), so that the modified one has better capability to predict the flow behavior at elevated temperature conditions.

$$\sigma(\varepsilon^p, \dot{\varepsilon}^p, T) = [A + B(\varepsilon^p)^n] \left[1 + C \ln \left(\frac{\dot{\varepsilon}^p}{\dot{\varepsilon}_0} \right) \right] \times \left[1 - \lambda \frac{e^{T/T_m} - e^{T_{\text{ref}}/T_m}}{e - e^{T_{\text{ref}}/T_m}} \right]. \quad (5)$$

Neither of G'sell–Jonas model and JC model mentioned above can accurately describe the strain softening deformation of the polymers. Thus, Duan *et al.* [41] proposed a homogeneity model suitable for glassy and semi-crystalline polymers based on G'sell–Jonas model, JC model, Matsuoka model, and Brook model, named DSGZ model. DSGZ model can describe the characteristics of elasticity, yielding, strain hardening, and strain softening by taking strain rate and temperature into account, as shown in Eq. (6).

$$\sigma(\varepsilon, \dot{\varepsilon}, T) = K \left\{ f(\varepsilon) + \left[\frac{\varepsilon \cdot e^{\left(1 - \frac{\varepsilon}{C_3 h(\dot{\varepsilon}, T)}\right)}}{C_3 h(\dot{\varepsilon}, T)} - f(\varepsilon) \right] \cdot e^{[\ln(g(\dot{\varepsilon}, T)) - C_4] \cdot \varepsilon} \right\} \cdot h(\dot{\varepsilon}, T), \quad (6)$$

$$f(\varepsilon) = (e^{-C_1 \cdot \varepsilon} + \varepsilon^{C_2})(1 - e^{-a \cdot \varepsilon})$$

$$h(\dot{\varepsilon}, T) = (\dot{\varepsilon})^m e^{\frac{a}{T}}.$$

where C_1 , C_2 , C_3 , C_4 , K , a , m , and a are model parameters, and they are easy to be calculated according to the strain–stress relationship. Then, Duan *et al.* [42,43] applied the DSGZ model to predict the deformation under the impact load of ABS, PC, and polybutylene-terephthalates (PBT). Further, the DSGZ model has been continuously improved and applied to ABS, PBT, PC, PMMA, PA, and PEEK by researchers [12,44–47].

In order to further evaluate the performance of phenomenological models, the deformation of PMMA and PEEK at a low strain rate was simulated by applying the DSGZ model by the authors. The experimental data of PMMA was taken from the article of Duan *et al.* [41], and the experimental data of PEEK was taken from the research of Chang *et al.* [48]. Particle swarm optimization was applied to fit the parameters of the DSGZ model according to the experimental data. By comparing the results of prediction with the experiment (as shown in Figure 6), it was found that the DSGZ model has the ability to capture the behavior of polymers at different temperatures. Moreover, the non-linear elastic deformation, yielding, strain softening, and strain hardening at low strain rates were captured well with a reasonable fitting of parameters. Although the prediction of the DSGZ model may not be guaranteed at a high strain rate, the phenomenological constitutive models can always find appropriate forms to obtain a satisfactory prediction.

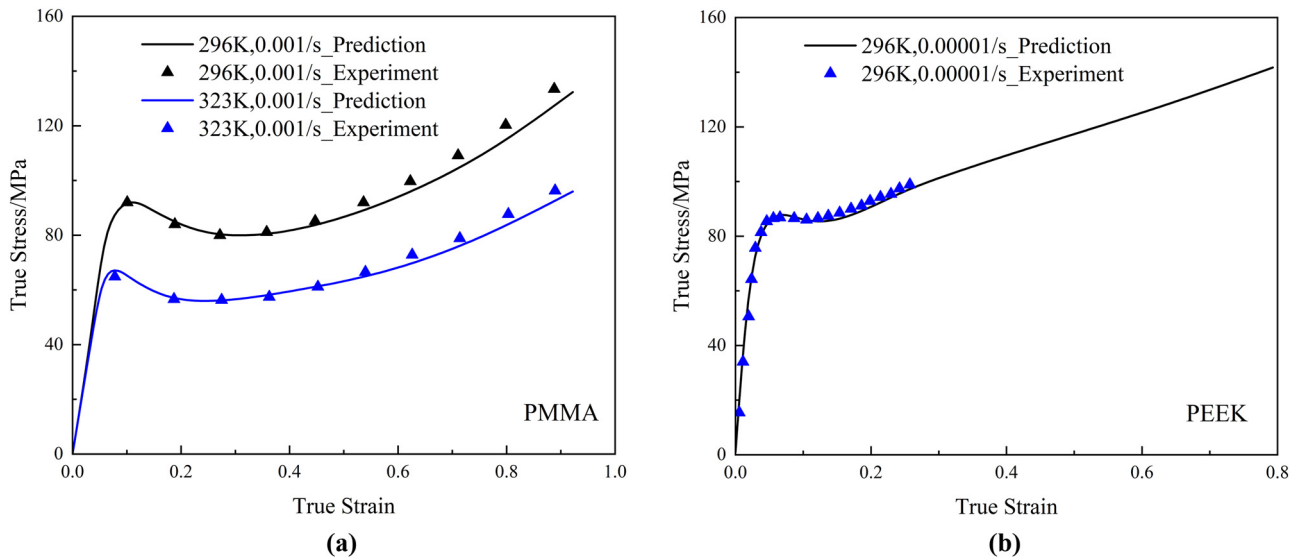


Figure 6: Comparison of true stress–strain curves between the prediction of DSGZ model with experimental results, left is for PMMA and right is for PEEK.

Mulliken and Boyce [49] proposed an M–B model on the basis of the BPA model in 2006. They decomposed the intermolecular resistance into two rate-dependent resistances as shown in Figure 7. In other words, network A in the BPA model was decomposed into two parallel Maxwell elements which are related to glass transition (α) and secondary molecular motions (β), respectively. In this way, the behavior of polymers in low strain rate can be reflected by α , and the behavior of polymers in high strain rate is determined by both α and β . The M–B model can not only capture the transition in the yield behavior, but also accurately predict the post-yield and large strain behavior over a wide range of temperatures and strain rates. The total stresses in the polymer are given as the tensorial sum of the intermolecular stresses of α and β and the network (back) stress. Moreover, expressions of three stress components are presented in Eq. (7). It has been indicated that the M–B model can accurately predict the behavior of materials in the strain rate range of 10^{-4} – 10^3 s^{-1} .

$$\begin{aligned}
 T &= T_{A\alpha} + T_{A\beta} + T_B \\
 T_{A\alpha} &= \frac{1}{J_\alpha} \phi_\alpha^e [\ln V_{A\alpha}^e] \\
 T_{A\beta} &= \frac{1}{J_\beta} \phi_\beta^e [\ln V_{A\beta}^e] \\
 T_B &= \frac{C_R}{3} \frac{\sqrt{N}}{\lambda_{\text{chain}}^p} \phi^{-1} \left(\frac{\lambda_{\text{chain}}^p}{\sqrt{N}} \right) \bar{B}_B'
 \end{aligned} \quad (7)$$

The M–B model is capable of predicting the deformation trend of polymers at low, moderate, and high strain rates. Figure 8 shows that it is convenient to use M–B

model for analysis of many polymers such as PC (shown in Figure 8 (a)) and PMMA (shown in Figure 8 (b)), because the deformation response is divided into α and β . However, it was found that the M–B model ignored the viscosity effect before yield. In addition, the strain softening stage predicted by the M–B model usually deviate from the experimental results due to the early arrival of yield. The M–B model can only predict the trend of strain hardening at a low strain rate, but the prediction becomes inaccurate with the increase in the strain rate. In addition, the M–B model is an isothermal model, and it cannot capture the post-yield thermal softening. Other researchers have drawn similar conclusions [50].

By combining the M–B model, G'sell model, and DSGZ model, Wang *et al.* [50,51] proposed an adiabatic phenomenological constitutive model to predict the mechanical behavior of PC at various strain rates and temperatures.

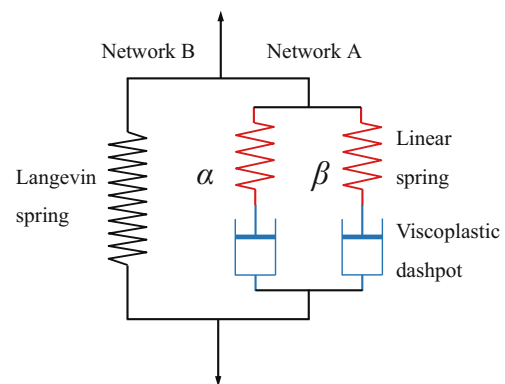


Figure 7: M–B model [49].

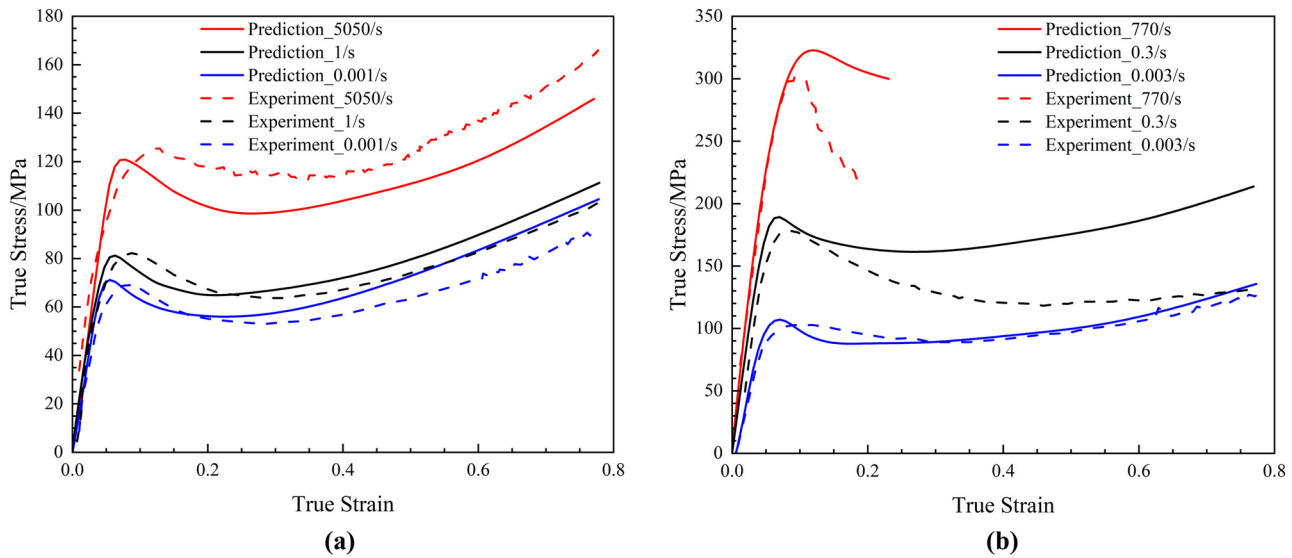


Figure 8: Comparison of true stress-strain curves between the predictions of M-B model and experimental results, where (a) shows the contrast for PC and (b) shows the contrast for PMMA [49].

Deformations of the low strain rate and the high strain rate were reflected by the α -component and the β -component, respectively. In addition, they used the model to simulate the split Hopkinson pressure bar testing and the falling weight impact testing by explicit user material subroutine (VUMAT). Moreover, results of the simulations agree well with the experimental data. In addition, some phenomenological models have been also developed for 3D-printed polymers [52].

Varghese and Batra [53] modified the M-B model by introducing temperature and strain rate-dependent elastic moduli and two internal variables at the high strain rate. Further, Safari *et al.* [54,55] indicated that α and β alone in network A were insufficient under the high strain rate, so γ transition as shown in Figure 9 was added to reflect the behavior of materials in the higher strain rate. Moreover, their studies have shown that the modified model can predict the thermomechanical behavior of polymer when the strain rate is over $10,000 \text{ s}^{-1}$. However, Safari *et al.* [54,55] indicated that due to adiabatic condition of high strain rate deformations, the modified model requires higher strain rates to account for temperature changes.

2.2.2 VE models

Both reversible and irreversible deformation is usually included for polymers, and some researchers argue that the reversible deformation of polymers should be concerned. Nonlinear VE characteristics will thus appear in such types of constitutive models. Schapery [56] proposed

that the behavior of some steels and polymers such as creep and relaxation can be reflected by linear equations, nonlinear equations, and some other specific equations which are summarized from a good deal of experiments. In fact, Schapery [56] attempted to build some connections between experimental phenomena and some constitutive equations, based on a framework of irreversible thermodynamics that are similar to the Boltzmann superposition integral form of linear theory. They suggested the core idea of phenomenological constitutive models and lay a foundation for the subsequent development of phenomenological constitutive models. Khan *et al.* [57] proposed a one-dimensional (1D) phenomenological constitutive model with a semi-empirical modification based on experimental observations, to capture the complex and highly nonlinear finite thermo-mechanical behaviors of VE polymers. Moreover,

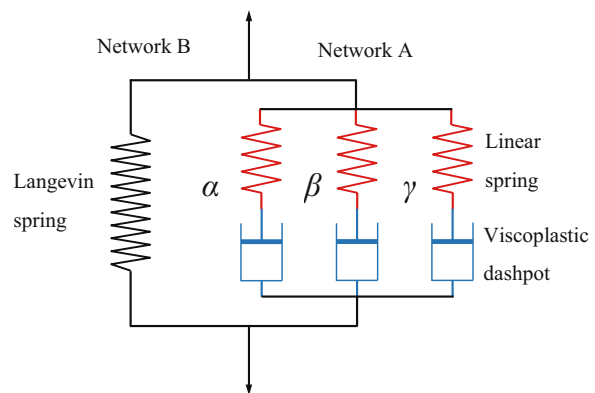


Figure 9: Safari model [54].

the model was built based on the infinitesimal linear theory. It was demonstrated that the model was accurate in predicting the deformation of polymers over a wide range of strain rates at room temperature or near the T_g . Chang *et al.* [48] proposed a phenomenological nonlinear VE constitutive model to characterize the stress–strain of PEEK before yielding with considering temperature and strain rate dependence. The VE model consists of a cubic nonlinear spring and a linear Maxwell element connected in parallel, and the form of this model as shown in Eq. (8) is similar to the subsequent Z-W-T model [58,59].

$$\sigma(t) = E_0\varepsilon + C_1\varepsilon^2 + C_2\varepsilon^3 + E_1 \int_0^t \dot{\varepsilon}(\tau) e^{-\frac{t-\tau}{\theta_1}} d\tau. \quad (8)$$

Wang *et al.* [58,59] proposed a Z-W-T model to represent the nonlinear VE deformation of polymers in 1991, and then they applied this model for the impact investigation in bird strikes on windshields of the high-speed aircraft. The expression is shown in Eq. (9).

$$\sigma(t) = E_0\varepsilon + \alpha\varepsilon^2 + \beta\varepsilon^3 + E_1 \int_0^t \dot{\varepsilon} e^{-\frac{t-\xi}{\theta_1}} d\xi + E_2 \int_0^t \dot{\varepsilon} e^{-\frac{t-\xi}{\theta_2}} d\xi, \quad (9)$$

where E_1 , θ_1 , and E_2 , θ_2 signify the elastic modulus and relaxation time of VE response at low or high strain rate, and E_0 , α , and β are nonlinear elastic modulus except for the VE response. In the early stage, the Z-W-T model was only suitable for polymers at room temperature, because the temperature was not considered in the model. Therefore, Wang *et al.* [60] extended the Z-W-T model by introducing the effect of temperature, and simulated the dynamic response of PMMA windshield against bird strike accurately. Dar *et al.* [61] also implemented the Z-W-T model to describe the mechanical behavior of PMMA. Nevertheless, the Z-W-T model is adaptable to simulate the VE response but not the VP response after yielding.

2.2.3 VP models

Some researchers pay attention to the irreversible deformation, and suggested a series of VP constitutive models. A series of models were improved based on over-stress model (VBO). Colak [62] proposed a modified viscoplasticity theory based on the over-stress model, which contains two tensor valued state variables, the equilibrium, and kinematic stresses and two scalars valued state variables, drag, and isotropic stresses. They compared the numerical results with the experimental data, and the tests of polyphenylene oxide were performed at different stresses above and below

the yield point. It is shown that nonlinear rate sensitivity, nonlinear unloading, creep, and recovery at zero stress were simulated by the proposed model. Besides, Ghorbel [63], Drozdov [64], Khan and Yeakle [65], Dusunceli and Colak [66] developed the corresponding models based on the VBO model for analysis of the polymers such as PA, PC, PET, and PP. Bardenhagen *et al.* [67] presented a general framework to develop the constitutive models of polymeric materials in the VP regime, and a VP constitutive model of three-dimensional finite deformation. Strain-rate dependence, stress relaxation, and creep phenomena can be reflected in the proposed model. Drozdov and Christiansen [68] presented a VP constitutive model for isothermal three-dimensional cyclic deformations with small strains of semi-crystalline polymers. Moreover, multiple inelastic deformations appear in the model with the 1D spring and dashpot construction. Although there are 15 material parameters in the proposed model, they can be determined step by step by matching appropriate intervals of a stress–strain curve at loading and retraction. Good prediction can be obtained, when the constitutive model was applied to predict the VP behavior of HDPE with a strain less than 0.1.

2.2.4 Hyperelastic models

Neo-Hookean model [69] is a classic statistical thermodynamic hyperelastic model, and it is suitable for the deformation of rubber-like materials. The Neo-Hookean model is expressed for its Helmholtz free energy per unit reference volume as shown in Eq. (10), which is independent of the temperature.

$$\begin{aligned} \Psi(I_1, J) &= \frac{\mu}{2}(I_1 - 3) + \frac{k}{2}(J - 1)^2 \\ I_1 &= \sum_{i=1}^3 \lambda_i^2, \quad J = \det F, \end{aligned} \quad (10)$$

where $\Psi(I_1, J)$ is the Helmholtz free energy, I_1 is the first invariant of strain tensor, and J is the determinant of deformation gradient tensor. In addition, Mooney–Rivlin model [70] is also a classic hyperelastic constitutive model.

Wang and Guth [71] established an orthogonal non-Gaussian three-chain network constitutive model to describe the large deformation of polymer chain segments by statistical methods in 1952. Flory and Rehner [72] proposed a four-chain constitutive model of regular tetrahedra to predict the strain hardening deformation of hyperelastic materials. Arruda *et al.* [73] proposed a fully three-dimensional eight-chain thermodynamically consistent constitutive model in 1995, and a thermo-mechanically coupled FE analysis for the

large deformation of glassy polymer was carried out. The inelastic deformation of strain hardening in this model is not dissipative, and it is to be stored as internal back stress. The strain rate and temperature-dependence of initial yield are included in the model as well as the temperature dependence of evolving anisotropy and its associated strain hardening. Wu and van der Giessen [74] also proposed an eight-chain constitutive model, which features strain-rate, pressure, and temperature-dependent yield, softening immediately after yield, and subsequent orientational hardening with further plastic deformation. Sweeney and Ward [75] conducted the experiments near and above the glass transition temperature (T_g) of PVC in 1995, in which PVC is an amorphous polymer containing a small amount of crystallization. In addition, they introduced strain-rate into Edwards–Vilgis model, and investigated the strain-rate dependence of PVC. Billon [76] proposed a 1D visco-hyperelastic model, and validated it by using a rich and rigorous database of PMMA above T_g . Moreover, they extended the Edwards–Vilgis model to a general time-dependent constitutive model. The inelastic phenomena in this model were reflected by an evolution of internal variables related to the alteration of microstructure, that induces changes in parameters in constitutive model and dissipation of energy. This approach worked well in modeling time-dependent behavior of polymers over a wide range of temperatures and strain rates. Maurel-Pantel *et al.* [77] extended the model proposed by Billon [76] to three-dimensional thermodynamically consistent constitutive equations, and proposed a visco-hyperelastic constitutive model in 2015. On the basis of Edward–Vilgis theory, they also represented the degree of mobility of entanglement points through the introduction of an evolution equation for the internal state variable. The thermomechanical model was applied to a semi-crystalline polyamide polymer (PA66), and the predicted deformation and temperature were in good agreement with the experimental results under tension and shear conditions. Gehring *et al.* [78] modified the network theory of Billon [76] and Maurel-Pantel *et al.* [77] in 2016. For the behavior of amorphous and semi-crystalline PET, they considered several contributions of the microstructure's rearrangement (disentanglement and loss of connectivity) within the large deformation formalism by the thermodynamic framework. In addition, the model considers microstructure at a mesoscopic level through the description of an equivalent network evolving with internal state variables. Good agreement was obtained by comparing with the results of the nonmonotonic tensile test. Federico *et al.* [79] used the theory proposed by Gehring *et al.* [78] to investigate the mechanical behavior of amorphous PMMA with different molecular weights in 2020.

The model can account for the elastic contribution of an equivalent network, which experiences inelastic mechanisms coming from the evolution of internal state variables. Therefore, the model [78] possesses the capability to capture the mechanical response of materials at different temperatures and strain rates through the VE and rubbery regimes. In addition, there are still lots of models based on such strategies [19,80–84].

2.2.5 Multi-mode models

However, some features have been ignored logically when constructing the above constitutive models. Different polymers usually exhibit different mechanical properties containing more than one pattern, due to the different degrees of crystallization. The properties of polymers would be the most important features to be considered for the study of constitutive models. Researchers hence attempted to couple the VE character and VP character or more characteristics in their models to obtain more reasonable results.

Anand *et al.* [85,86] developed an elastic-viscoplastic thermodynamically consistent constitutive model for the amorphous polymers with strain rate and temperature-dependent large-deformation behaviors. Moreover, they implemented their model for simulation of large-strain compression experiments for PMMA, PC, and a cyclo-olefin polymer (Zeonex-690R). It was validated that the model can reflect the strain rate and temperature-dependent yielding, strain-hardening, unloading response, and the temperature transforming. Frank and Brockman [87] presented a constitutive model which combines nonlinear VE character and VP character of polymers for multi-axial isotropic deformation in 2001. Several traditional constitutive models expressed by some simple empirical relationships were included in their model such as linear elasticity, linear viscoelasticity, and plasticity, so that the time-dependent and nonlinear behaviors can be captured. The proposed model was applied to simulate the impact behavior of PC, and the feasibility and accuracy of the model are demonstrated.

Miled *et al.* [88] proposed a coupled VE–VP constitutive model for homogeneous and isotropic polymers. Moreover, fully implicit integration, a two-step return mapping corrector strategy, and a consistent tangent operator were used in their work. The model is restricted to the regime of small perturbations (small strains, displacements, and rotations), and the VE part of the model is linear and isothermal. The total strain in this model was assumed to be a sum of VE strain and VP strain, and this behavior is a

common strategy for multi-mode models. The model was implemented in a three-stage shear test and the simulation of HDPE through ABAQUS software via a user-defined routine (UMAT), and the model was proved to be effective. Yu *et al.* [89] proposed a nonlinear VE–VP cyclic constitutive model based on the tests of multilevel loading–unloading recovery, creep-recovery, and cyclic tension–compression/tension ones. They extended Schapery's nonlinear VE model [56] as the VE part of the model. Moreover, as for the accumulation of irrecoverable VP strain produced during cyclic loading, they adopted the Ohno–Abdel-Karim's nonlinear kinematic hardening rule [90]. This model was verified by comparison of the prediction with the tests of PC. Other researchers [13,91–98] have also made significant contributions in this regard.

2.2.6 Constitutive models with damage

It is out of their depth for the above models to describe the damage or crack during the deformation, even though most of the models have reflected the viscous, elastic, and plastic behavior of polymers well. Therefore, researchers take the damage to materials into account when constructing the constitutive models.

Zairi *et al.* [99,100] proposed a constitutive model to describe the elasto-viscoplastic damage behaviors of polymers according to both micromechanical (Gurson potential) and phenomenological (modified Bodner–Partom model) models, which contains hydrostatic and failure evolution terms. They investigated the macro-mechanical response and damage micro-mechanism by failure growth in rubber-toughened glassy polymer by the simulation of RTPMMA and HIPS, and a good agreement was obtained. An exploratory approach is provided by this model to capture the damage quantitatively, while some improvements are still essential. Tehrani and Abu Al-Rub [101] proposed a nonlinear VE, VP, and visco-damage constitutive model, and applied the proposed model for the damage evolution simulation of PMMA embedded with silicate nanoclay particles. They introduced a damage evolution variable which is a function of actual stress, hydrostatic stress, total strain, strain rate, temperature, and damage process. A series of simulations were performed, and the model was proved to be reasonable. Balieu *et al.* [102] proposed a phenomenological non-associated elasto-viscoplastic model coupled with damage in the finite strain framework, which was used to simulate the behavior of a 20% mineral-filled semi-crystalline polymer for a large strain rate range. Perzyna-type VP formulation was used to represent the nonlinear strain-stress response, and the specific non-associated VP potential

was used to capture the volume change of the material under tensile and compression loading. In addition, hydrostatic pressure was introduced to reflect the yield stress coupled with the damage model. They implemented the model into FE simulation through UMAT, and the result obtained were in good agreement with the experimental data.

Krairi and Doghri [103] proposed a constitutive model by coupling viscoelasticity, viscoplasticity, and ductile damage within the framework of irreversible thermodynamics. They simulated the time and strain rate dependencies, the Bauschinger effect, and ductile damage through a series of simple expressions. Ductile damage evolution is related to VP strains, and it evolves only with changes in VP strains. The model was validated by tests for different polymers under various loadings. Praud *et al.* [104] also proposed a VE, VP constitutive model considering ductile damage within the framework of thermodynamics, but the VP behavior of their model is different from that of Krairi and Doghri [103]. In the model proposed by Praud *et al.* [104], the VE part is described with the help of a series of Kelvin–Voigt branches, while the VE properties of the model of Krairi and Doghri [103] are expressed through Prony series through an integral formulation. Khaleghi *et al.* [105] suggested a thermodynamically consistent damage model to predict failure of glassy polymers based on the EGP multi-mode model. In their model, the effects of plastic deformation and hydrostatic stress on damage evolution were considered. In addition, Abu Al-Rub *et al.* [106], Seidel *et al.* [107], Voyiadjis *et al.* [108], and Cayzac *et al.* [109] also made great contributions in this aspect. In addition, some issues had also attracted the attention of investigators, such as the influence of moisture [110], heating [111], finite strains [112], change in crystallization [113], hardening behavior [114] and softening behavior [115]. Phenomenological models can be constructed by combining the physical mechanisms and the results of tests, so phenomenological model can usually predict the deformation well as the model is constructed correctly. In addition, phenomenological constitutive models introduced in this section are listed in Table A3 of the Appendix.

2.2.7 Constitutive models incorporating machine learning algorithms

There are still some weaknesses for the above traditional nonlinear constitutive models, although intensive studies have been performed and remarkable results have been achieved in the past decades. Most of the physical constitutive models derived from microstructural aspects are expressed in complex equations, but the performances

are not always perfect. Moreover, coefficients determined from the tests of both phenomenological models and physical models will affect the performances as well [116]. With the rapid development of machine learning algorithms and computing power, researchers attempt to reflect the behaviors of materials by machine learning algorithms or introducing them into mechanics. In recent years, artificial neural networks (ANN) models are the most popular and widely used machine learning models due to their outstanding performance in terms of “big data” and nonlinear modeling. Therefore, the machine learning-based constitutive models will be reviewed in this section, and the typical model will be recovered and evaluated, so that a clear understanding can be provided of the characteristics of machine learning-based constitutive models. Furthermore, a possible direction for the research of constitutive models can also be provided.

Ghaboussi *et al.* [118] suggested that the behavior of materials may be reflected by neural networks (NN) as early as 1991. Back-propagation neural network was constructed for the plane stress state under monotonic biaxial loading and compressive uniaxial cycle loading. The NN model is self-operated, and all the parameters responsible for the behavior of materials and experimental data are reflected within a unified environment of a NN. After that, they conducted a series of studies about the NN-based constitutive models, in which the rate-dependent behaviors (as shown in Figure 10) or hysteretic behaviors were considered [117,119]. In their research, the strain, strain rate, and the internal variables were taken as the inputs directly, and stresses were the outputs. The ANN-based model was implemented within the FE method for the results of

boundary value problems [120]. The experimental data and simulation data calculated by physical constitutive models were combined, and worked together with the NN model to achieve more accurate results [121]. Their studies not only provided evidence that it is feasible to forecast the behavior of materials by the NN approach, but also established a basic framework for the ANN-based constitutive models.

Al-Haik *et al.* [122] developed an ANN-based constitutive model to predict the stress relaxation of polymer matrix composite. A series of stress relaxation tests were performed at the conditions of constant strain and constant temperature, and 9,000 experimental datasets were used to train the ANN model. The ANN model consisted of three input layers, two hidden layers, and one output layer. To verify the performance of the ANN-based constitutive model, they compared the results of the ANN model and another nonlinear VE constitutive model with the experimental results. The predictions of the ANN model are found to be more accurate than those of the nonlinear VE model over a wider range of stress and temperature conditions, in particular near the T_g . Their study further proved that the ANN model has great potential in constitutive modeling.

Rodriguez *et al.* [123] predicted the behavior of thermoplastic polymer through a feed-forward artificial NN model. A model with the stress–strain curve as outputs and test datasets as inputs was constructed, and a response surface of stress varying with strain and temperature was modeled as shown in Figure 11. They further compared the performance of ANN-based model with several other constitutive models such as Polynomial model, Neo-Hookean model, Yeoh model, Mooney–Rivlin model, and Ogden

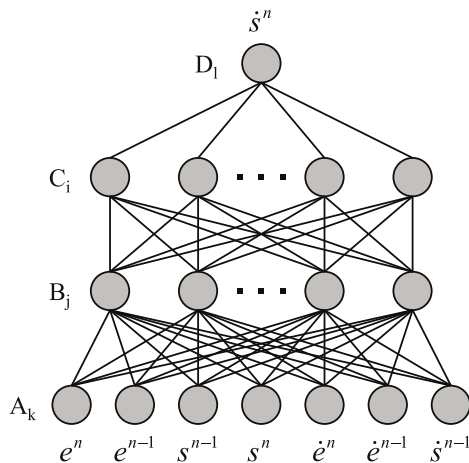


Figure 10: Rate-dependent NN constitutive model (A_k is equal to e_{lm}/S_k , where $e_{lm} = \epsilon_{lm} - \delta_{lm}\epsilon_v/3$, S denotes the scale factor, the dots denote the rates, n and $n-1$ denote the discrete time steps) [117].

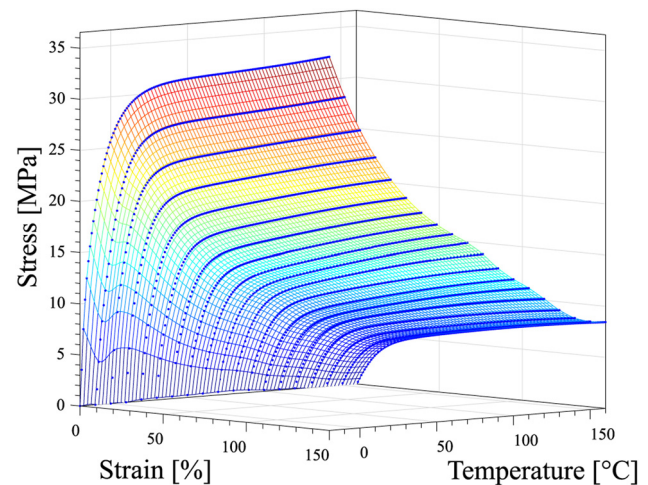


Figure 11: The stress response surface given by the ANN model [123].

model. The proposed ANN-based model achieved the best prediction accuracy among these models, and the predicted maximum percentage error (mean square error) was even less than 1%.

By combining the NN model with elasticity theory, researchers also attempted to construct new constitutive models, except for the direct modeling by NN. Jordan *et al.* [124] decomposed the total logarithmic axial strain into a purely-elastic strain and a VE strain. The elastic deformation was reflected by a temperature-dependent Hooke's law, while the VE deformation was reflected by the NN model. The relationship between stress with the viscous strain, viscous strain rate, and temperature are identified, without making any prior assumptions on the specific mathematical form. The proposed NN constitutive model and a thermos-elastic-viscoplastic constitutive mode named the Johnson model [125] were implemented to simulate the deformation of PP at various temperatures and low strain rates, and a comparison was made with experimental results as shown in Figure 12. It was demonstrated that the NN model has the capability to predict the stress-strain response at low strain condition.

Li *et al.* [126] improved the Johnson–Cook model [36] by applying the machine learning algorithm to capture the observed unconventional effect of the strain rate and temperature on the hardening response. A feed-forward NN with three hidden layers was introduced to represent a function that describes the effects of the strain, strain rate, and temperature. The NN was implemented into UMAT, and an accurate NN model was constructed by repeatedly launching full 3D FE simulations during the training. A hardening law of DP800 steel was thus obtained,

in which the yield stress is expressed as a function of the equivalent plastic strain, strain rate, and temperature. Although the object of study is metal, this approach is still a typical strategy for combining the NN algorithm with the traditional constitutive model.

Besides, the NN model was not only widely used in the construction of constitutive models, but also in the performance prediction of fiber-reinforced polymeric composite structures [127–129], the damage prediction of composites [130], multiscale modeling of composites [131] or replacing the expensive nonlinear computation of FE model [132].

Tao *et al.* [133] coupled the commercial FE code Abaqus with the deep neural network model, to acquire the constitutive relationship of the fiber-reinforced composites. The proposed system avoids excessive assumptions in the process of constitutive modeling and satisfies the equilibrium and kinematics equations, so that it has more potential to conform to physics laws. Engineering constants of the fiber-reinforced composite were acquired, and the progressive damage of structures was also captured. This research shows that the machine learning algorithm can be applied to not only the constitutive modeling of materials, but also the constitutive modeling of structures.

In addition to the above NN algorithm, other machine learning algorithms have also been employed for constructing constitutive models, such as convolutional neural network [134], Gaussian process machine learning [135], and genetic algorithms (GAs) [136]. As far as the authors are concerned, GAs are applied to solve the parameters of constitutive models in most cases. Colak and Cakir [136] proposed a constitutive model to reflect the rate and temperature-dependent stress-strain behavior of polymers.

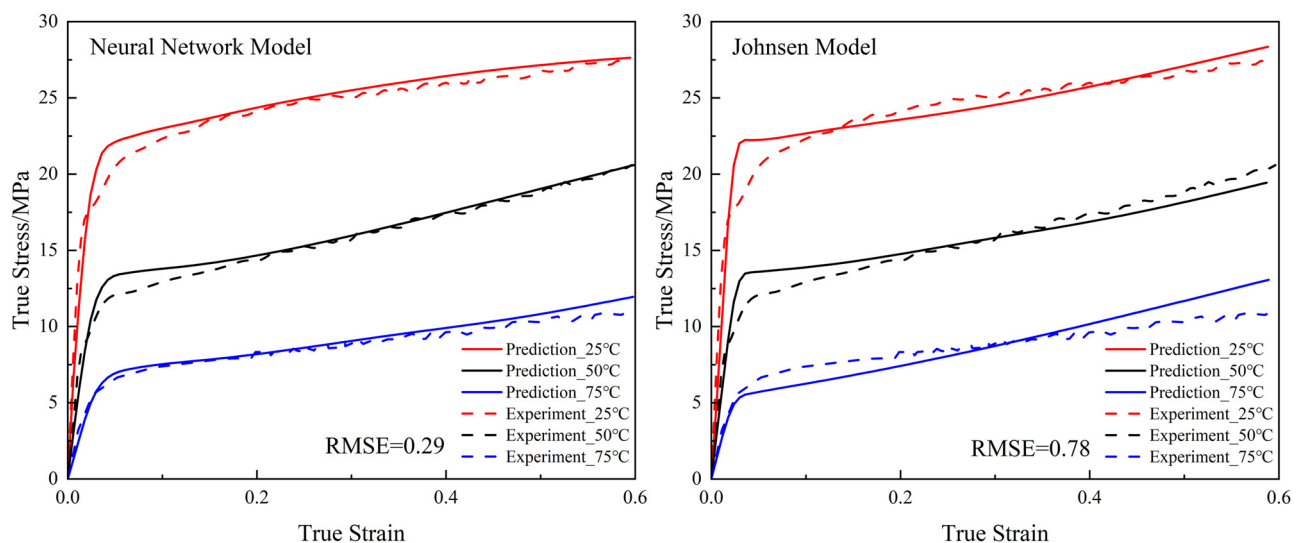


Figure 12: Comparison of the NN model and Johnson model [124].

The model was modified from the cooperative-viscoplasticity theory of the VBO model, and GA was adopted to determine the material parameters of the models. The proposed model with parameters obtained from GA was capable of predicting the mechanical behavior and all features below and through T_g of polymers.

The authors built an NN-based constitutive model on the basis of previous research to further evaluate the performance of the machine learning-based constitutive models. A 2D NN model was established, and the model contains one input layer, ten hidden layers, and one output layer. The deformations of PEEK in which strain rate was maintained at 0.001 s^{-1} were studied, and the experimental data were still taken from the research of Chang *et al.* [48]. True strain and temperature were the input variables, and true stress was the output variable. The experimental data which obtained from the tests at the temperature of -60°C , -2°C , 23°C , 100°C , and 140°C were used for training the constitutive model, and the experimental data at the temperature of 60°C were used for validating the constitutive model. Figure 13 presents the comparison of modeling (surface), training samples (red dots), and validation samples (black dots, the black dots are almost hidden under the surface). It was found that the NN-based constitutive model achieved a good agreement with the experimental results in a wide range.

In addition, Figure 14 compares the deformation at the temperature of 60°C predicted with testing separately. Our studies show that the NN-based constitutive model is capable of predicting the deformation of polymers when a suitable model is established with good training accuracy

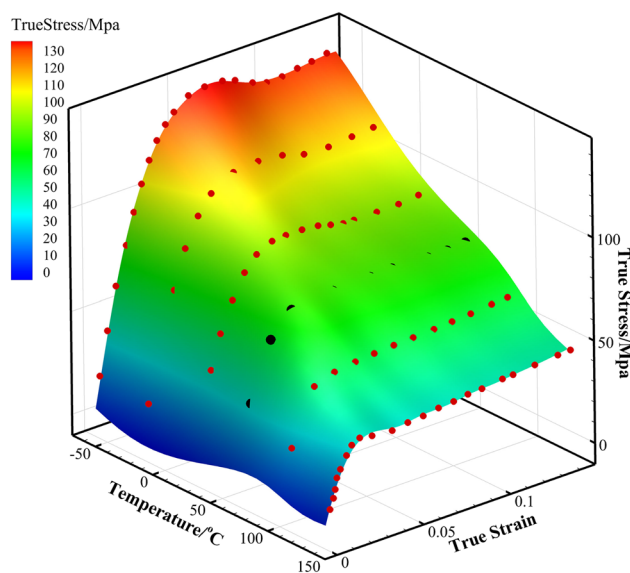


Figure 13: Modeling of NN-based constitutive model (surface: NN-based model, red dots: samples input, black dots: experimental data to be compared).

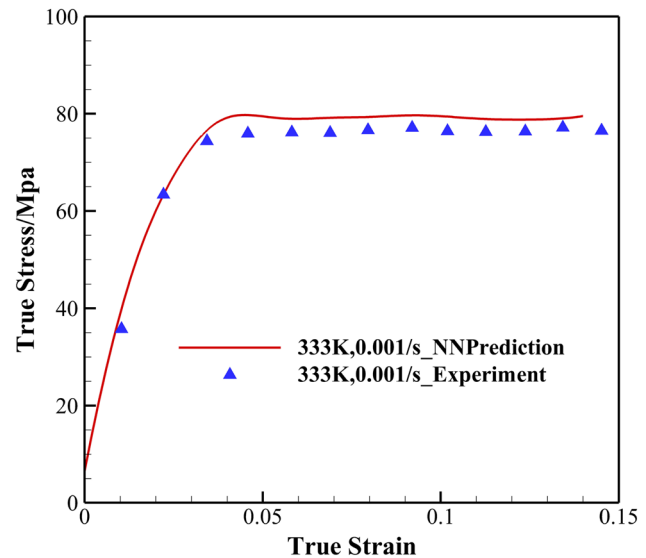


Figure 14: Comparison of true stress–strain curves between the prediction of NN-based model with experimental results for PEEK.

and enough experimental data are provided. In addition, the NN-based constitutive model is suitable for any polymers. Certainly, the performance of a machine learning-based constitutive model greatly depends on the samples, training accuracy, and many other factors, and it is challenging to achieve high accuracy.

Many other researchers were also interested in using the NN algorithms to investigate the nonlinear characteristics of thermoplastic polymers in recent years [137–139]. In addition, the machine learning-based constitutive models in this section are listed in Table A4 of the Appendix. Among the current studies, many benefits are found for such strategies. First, almost no assumptions were made, but the behaviors of materials can be captured by training the NN model by using experimental or computational data. Therefore, such an approach possesses the capability to describe the actual behavior of materials. Second, the NN-based constitutive models may discover unknown deformation mechanisms of polymers by learning the deformation of polymers [116]. Third, the computation may be accelerated by applying the NN algorithm with a reasonable approach.

Up to date, there are still some disadvantages for NN-based constitutive models [116]. In fact, the experiments are usually “expensive,” so it is difficult to obtain a large amount of training data. Due to lack of experimental data, it is not easy to maintain the steady performance of NN-based constitutive models. In addition, the NN-based constitutive models will encounter difficulties to accurately predict the results outside the training sets. For example, when the NN-based model is trained by using experimental data obtained from tensile deformations, such model might

encounter difficulties to accurately predict shear deformations. Moreover, the NN-based constitutive model cannot closely relate to the physical mechanism of materials.

Nevertheless, with the development of machine learning technology, these challenges are expected to be overcome in the future. For example, combination of physical constitutive models and machine learning algorithms might be a choice. Therefore, the machine learning algorithms exhibit a great potential in the research of constitutive models for thermoplastic polymers.

3 Discussion and conclusion

Comparison and review of classical and machine-learning based constitutive models for polymers used in aeronautical thermoplastic composites have been discussed in the present work. By comparing these models, it was found that classical constitutive models have been developed over the past decades and widely used in thermoplastic polymers. In addition, the machine learning algorithms possess a great potential to construct constitutive models for thermoplastic polymers. The detailed conclusions can be drawn as follows:

- 1) Differing from phenomenological models, physical models can describe the deformation mechanism of polymers, which can be explicitly traced to the microscopic level. Moreover, construction of physical models will not highly depend on the quantity of experimental data. Nevertheless, the performance of physical models will be obviously influenced by their construction schemes, and it is more complex to apply the physical models for analysis of engineering issues than phenomenological models. In addition, it is difficult to extend the range of application of the physical models, as a certain physical model may be only suitable for some deformation patterns of corresponding materials.
- 2) Phenomenological models can be constructed by combining the physical mechanisms and the results of tests. If the model is constructed correctly, phenomenological model can usually predict the deformation well. In addition, the forms and schemes of phenomenological models are usually simpler than those of physical models. However, the phenomenological models may encounter difficulties in describing the microscopic deformation mechanisms of polymers. In addition, accuracy of the phenomenological models largely depends on testing data and experiences.
- 3) If machine learning-based constitutive models are well trained, these models have the ability to accurately predict the mechanical behavior of polymers. Because the experimental data can be well used, the machine learning-based constitutive models have more advantages than classical constitutive models in predicting the hardening, softening,

relaxing, and creep. However, performance of machine learning-based constitutive models will be significantly influenced by the choice of machine learning algorithm, the training quality, and the reliability of experimental data. In addition, the machine learning-based constitutive models may encounter troubles in predicting the deformations outside the training data and exploring the physical mechanism of materials.

- 4) In the short term, due to the widespread application of machine learning technology, phenomenological constitutive models combined with machine learning algorithms have shown activity and capability in studying the deformation of thermoplastic polymers. Therefore, phenomenological constitutive models with the help of machine learning algorithms are expected to be used more and more extensively in engineering. In the future, the correlation between macrostructure and microstructure of materials is expected to be effectively addressed, and computer technology will be further developed, which are still challenging issues. Then, the combination of physical constitutive models, phenomenological constitutive models, and machine-learning algorithms will be increasingly attractive, as they can describe the physical deformation mechanism of materials accurately.

Acknowledgments: This work was supported by the Institute of Aircraft Composite Structures of Northwestern Polytechnical University.

Funding information: The work described in this article was supported by the National Natural Sciences Foundation of China (No. 12172295).

Author contributions: Shengbo Ling: conceptualization, investigation, and writing – original draft; Zhen Wu: conceptualization, writing – review and editing, and supervision; and Jie Mei: writing – review and editing. All authors have accepted responsibility for the entire content of this manuscript and approved its submission.

Conflict of interest: The authors state no conflict of interest.

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

References

- [1] Park, S. J. and M. K. Seo. Carbon fiber-reinforced polymer composites: preparation, properties, and applications. *Polymer Composites*, Vol. 1, 2012, pp. 135–183.

- [2] Marsh, G. Boeing's 787: trials, tribulations, and restoring the dream. *Reinforced Plastics*, Vol. 53, No. 8, 2009, pp. 16–21.
- [3] Marsh, G. Airbus takes on boeing with reinforced plastic A350 XWB. *Reinforced Plastics*, Vol. 51, No. 11, 2007, pp. 26–29.
- [4] Sudhin, A. U., M. Remanan, A. G. Ajesh, and K. Jayanarayanan. Comparison of properties of carbon fiber reinforced thermoplastic and thermosetting composites for aerospace applications. *Materials Today: Proceedings*, Vol. 24, 2020, pp. 453–462.
- [5] Sun, X. C., L. F. Kawashita, A. S. Kaddour, M. J. Hiley, and S. R. Hallett. Comparison of low velocity impact modelling techniques for thermoplastic and thermoset polymer composites. *Composite Structures*, Vol. 203, 2018, pp. 659–671.
- [6] Bernatas, R., S. Dagreou, A. Despax-Ferreres, and A. Barasinski. Recycling of fiber reinforced composites with a focus on thermoplastic composites. *Cleaner Engineering and Technology*, Vol. 5, 2021, id. 100272.
- [7] Meng, F., J. McKechnie, T. Turner, K. H. Wong, and S. J. Pickering. Environmental aspects of use of recycled carbon fiber composites in automotive applications. *Environmental Science and Technology*, Vol. 51, No. 21, 2017, pp. 12727–12736.
- [8] Barba, D., A. Arias, and D. Garcia-Gonzalez. Temperature and strain rate dependences on hardening and softening behaviours in semi-crystalline polymers: application to peek. *International Journal of Solids and Structures*, Vol. 182–183, 2020, pp. 205–217.
- [9] Yao, S., F. Jin, K. Y. Rhee, D. Hui, and S. Park. Recent advances in carbon-fiber-reinforced thermoplastic composites: a review. *Composites, Part B: Engineering*, Vol. 142, 2018, pp. 241–250.
- [10] Roux, M., N. Eguémann, C. Dransfeld, F. Thiébaud, and D. Perreux. Thermoplastic carbon fibre-reinforced polymer recycling with electrodynamic fragmentation. *Journal of Thermoplastic Composite Materials*, Vol. 30, No. 3, 2017, pp. 381–403.
- [11] Barile, M., L. Lecce, M. Iannone, S. Pappadà, and P. Roberti. Thermoplastic composites for aerospace applications. In: *Revolutionizing Aircraft Materials and Processes*, S. Pantelakis, Tserpes, K., editors., Springer International Publishing, Cham, 2020, pp. 87–114.
- [12] Zhu, H., H. Ou, and A. Popov. A new phenomenological constitutive model for thermoplastics. *Mechanics of Materials*, Vol. 157, 2021, id. 103817.
- [13] Ayoub, G., F. Zaïri, M. Naït-Abdelaziz, and J. M. Gloaguen. Modelling large deformation behaviour under loading-unloading of semicrystalline polymers: application to a high density polyethylene. *International Journal of Plasticity*, Vol. 26, No. 3, 2010, pp. 329–347.
- [14] Cao, K., Y. Wang, and Y. Wang. Experimental investigation and modeling of the tension behavior of polycarbonate with temperature effects from low to high strain rates. *International Journal of Solids and Structures*, Vol. 51, No. 13, 2014, pp. 2539–2548.
- [15] Krairi, A., I. Doghri, J. Schalnath, G. Robert, and W. Van Paepegem. Thermo-mechanical coupling of a viscoelastic-viscoplastic model for thermoplastic polymers: thermodynamical derivation and experimental assessment. *International Journal of Plasticity*, Vol. 115, 2019, pp. 154–177.
- [16] Peng, Y., Y. Xiaohu, Z. Xiaoqing, and H. Qiang. Mechanical behaviors and constitutive models of polycarbonate amorphous polymers. *Advances in Mechanics*, Vol. 46, No. 1, 2016, id. 201603.
- [17] Haward, R. N. and G. Thackray. The use of a mathematical model to describe isothermal stress-strain curves in glassy thermoplastics. *Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences*, Vol. 302, No. 1471, 1968, pp. 453–472.
- [18] Edwards, S. F. and T. Vilgis. The effect of entanglements in rubber elasticity. *Polymer*, Vol. 27, No. 4, 1986, pp. 483–492.
- [19] Garcia-Gonzalez, D., R. Zaera, and A. Arias. A hyperelastic-thermoviscoplastic constitutive model for semi-crystalline polymers: application to peek under dynamic loading conditions. *International Journal of Plasticity*, Vol. 88, 2017, pp. 27–52.
- [20] James, H. M. and E. Guth. Theory of the elastic properties of rubber. *The Journal of Chemical Physics*, Vol. 11, No. 10, 1943, pp. 455–481.
- [21] Ball, R. C., M. Doi, S. F. Edwards, and M. Warner. Elasticity of entangled networks. *Polymer*, Vol. 22, No. 8, 1981, pp. 1010–1018.
- [22] Van Ruiten, J., R. Riedel, R. Deblieck, R. Brouwer, and J. P. Penning. Drawability and attainable mechanical properties of polyamide yarn using true stress-true strain curves. *Journal of Materials Science*, Vol. 36, No. 13, 2001, pp. 3119–3128.
- [23] Sweeney, J., T. L. D. Collins, P. D. Coates, A. P. Unwin, R. A. Duckett, and I. M. Ward. Application of a large deformation model to unstable tensile stretching of polyethylene. *International Journal of Plasticity*, Vol. 18, No. 3, 2002, pp. 399–414.
- [24] Ree, T. and H. Eyring. Theory of non-Newtonian flow. I. Solid plastic system. *Journal of Applied Physics*, Vol. 26, No. 7, 1955, pp. 793–800.
- [25] Boyce, M. C., D. M. Parks, and A. S. Argon. Large inelastic deformation of glassy polymers. Part I. *Rate Dependent Constitutive Model Mechanics of Materials*, Vol. 7, No. 1, 1988, pp. 15–33.
- [26] Buckley, C. P. and D. C. Jones. Glass-rubber constitutive model for amorphous polymers near the glass-transition. *Polymer*, Vol. 36, No. 17, 1995, pp. 3301–3312.
- [27] Wu, J. J. and C. P. Buckley. Plastic deformation of glassy polystyrene: a unified model of yield and the role of chain length. *Journal of Polymer Science, Part B: Polymer Physics*, Vol. 42, No. 11, 2004, pp. 2027–2040.
- [28] De Focatiis, D. S. A., J. Embery, and C. P. Buckley. Large deformations in oriented polymer glasses: experimental study and a new glass-melt constitutive model. *Journal of Polymer Science, Part B: Polymer Physics*, Vol. 48, No. 13, 2010, pp. 1449–1463.
- [29] Tervoort, T. A., R. J. M. Smit, W. A. M. Brekelmans, and L. E. Govaert. A constitutive equation for the elasto-viscoplastic deformation of glassy polymers. *Mechanics of Time-Dependent Materials*, Vol. 1, No. 3, 1997, pp. 269–291.
- [30] Leonov, A. I. Nonequilibrium thermodynamics and rheology of viscoelastic polymer media. *Rheologica Acta*, Vol. 15, No. 2, 1976, pp. 85–98.
- [31] Govaert, L. E., P. Timmermans, and W. Brekelmans. The influence of intrinsic strain softening on strain localization in polycarbonate: modeling and experimental validation. *Journal Engineering Materials Technology*, Vol. 122, No. 2, 2000, pp. 177–185. Asme.
- [32] Govaert, L. E. and T. A. Tervoort. Strain hardening of polycarbonate in the glassy state: influence of temperature and molecular weight. *Journal of Polymer Science, Part B: Polymer Physics*, Vol. 42, No. 11, 2004, pp. 2041–2049.
- [33] van Breemen, L. C. A., E. T. J. Klompen, L. E. Govaert, and H. E. H. Meijer. Extending the EGP constitutive model for polymer glasses to multiple relaxation times. *Journal of the Mechanics and Physics of Solids*, Vol. 59, No. 10, 2011, pp. 2191–2207.
- [34] G'Sell, C. and J. J. Jonas. Determination of the plastic behaviour of solid polymers at constant true strain rate. *Journal of Materials Science*, Vol. 14, No. 3, 1979, pp. 583–591.

- [35] G'Sell, C., N. A. Aly-Helal, and J. J. Jonas. Effect of stress triaxiality on neck propagation during the tensile stretching of solid polymers. *Journal of Materials Science*, Vol. 18, No. 6, 1983, pp. 1731–1742.
- [36] Johnson, G. R. and W. H. Cook. A constitutive model and data for metals subjected to large strains, high strain rates and high temperatures. *Engineering Fracture Mechanics*, Vol. 21, 1983, pp. 541–548.
- [37] Johnson, G. R. and W. H. Cook. Fracture characteristics of three metals subjected to various strains, strain rates, temperatures and pressures. *Engineering Fracture Mechanics*, Vol. 21, No. 1, 1985, pp. 31–48.
- [38] Garcia-Gonzalez, D., A. Rusinek, T. Jankowiak, and A. Arias. Mechanical impact behavior of polyether-ether-ketone (PEEK). *Composite Structures*, Vol. 124, 2015, pp. 88–99.
- [39] Chen, F., H. Ou, S. Gatea, and H. Long. Hot tensile fracture characteristics and constitutive modelling of polyether-ether-ketone (PEEK). *Polymer Testing*, Vol. 63, 2017, pp. 168–179.
- [40] Chen, F., H. Ou, B. Lu, and H. Long. A constitutive model of polyether-ether-ketone (PEEK). *Journal of the Mechanical Behavior of Biomedical Materials*, Vol. 53, 2016, pp. 427–433.
- [41] Duan, Y., A. Saigal, R. Greif, and M. A. Zimmerman. A uniform phenomenological constitutive model for glassy and semicrystalline polymers. *Polymer Engineering & Science*, Vol. 41, No. 8, 2001, pp. 1322–1328.
- [42] Duan, Y., A. Saigal, R. Greif, and M. A. Zimmerman. Analysis of multiaxial impact behavior of polymers. *Polymer Engineering and Science*, Vol. 42, No. 2, 2002, pp. 395–402.
- [43] Duan, Y., A. Saigal, R. Greif, and M. A. Zimmerman. Impact behavior and modeling of engineering polymers. *Polymer Engineering & Science*, Vol. 43, No. 1, 2003, pp. 112–124.
- [44] Yin, Z. N. and T. J. Wang. Deformation of PC/ABS alloys at elevated temperatures and high strain rates. *Materials Science and Engineering: A*, Vol. 494, No. 1–2, 2008, pp. 304–313.
- [45] Zheng, B., H. Wang, Z. Huang, Y. Zhang, H. Zhou, and D. Li. Experimental investigation and constitutive modeling of the deformation behavior of polyether-ether-ketone at elevated temperatures. *Polymer Testing*, Vol. 63, 2017, pp. 349–359.
- [46] Dar, U. A., W. Zhang, Y. Xu, and J. Wang. Thermal and strain rate sensitive compressive behavior of polycarbonate polymer – experimental and constitutive analysis. *Journal of Polymer Research*, Vol. 21, No. 8, 2014, pp. 1–10.
- [47] Nahar, C., S. Sanariya, and P. K. Gurrula. Numerical simulation of polymers at low and moderate strain rates. *Materials Today: Proceedings*, Vol. 44, 2021, pp. 696–700.
- [48] Chang, B., X. Wang, Z. Long, Z. Li, J. Gu, S. Ruan, et al. Constitutive modeling for the accurate characterization of the tension behavior of peek under small strain. *Polymer Testing*, Vol. 69, 2018, pp. 514–521.
- [49] Mulliken, A. D. and M. C. Boyce. Mechanics of the rate-dependent elastic-plastic deformation of glassy polymers from low to high strain rates. *International Journal of Solids and Structures*, Vol. 43, No. 5, 2006, pp. 1331–1356.
- [50] Wang, H., Y. Zhang, Z. Huang, Z. Tang, Y. Wang, and H. Zhou. Establishment and comparison of four constitutive relationships of PC/ABS from low to high uniaxial strain rates. *Mechanics of Time-Dependent Materials*, Vol. 22, No. 4, 2018, pp. 457–481.
- [51] Wang, H., H. Zhou, Z. Huang, Y. Zhang, and X. Zhao. Constitutive modeling of polycarbonate over a wide range of strain rates and temperatures. *Mechanics of Time-Dependent Materials*, Vol. 21, No. 1, 2016, pp. 97–117.
- [52] Wang, Y., X. Li, Y. Chen, and C. Zhang. Strain rate dependent mechanical properties of 3D printed polymer materials using the DLP technique. *Additive Manufacturing*, Vol. 47, 2021, id. 102368.
- [53] Varghese, A. G. and R. C. Batra. Constitutive equations for thermomechanical deformations of glassy polymers. *International Journal of Solids and Structures*, Vol. 46, No. 22–23, 2009, pp. 4079–4094.
- [54] Safari, K. H., J. Zamani, F. J. Ferreira, and R. M. Guedes. Constitutive modeling of polycarbonate during high strain rate deformation. *Polymer Engineering & Science*, Vol. 53, No. 4, 2013, pp. 752–761.
- [55] Safari, K. H., J. Zamani, R. M. Guedes, and F. J. Ferreira. The effect of heat developed during high strain rate deformation on the constitutive modeling of amorphous polymers. *Mechanics of Time-Dependent Materials*, Vol. 20, No. 1, 2016, pp. 45–64.
- [56] Schapery, R. A. On the characterization of nonlinear viscoelastic materials. *Polymer Engineering & Science*, Vol. 9, No. 4, 1969, pp. 295–310.
- [57] Khan, A. S., O. Lopez-Pamies, and R. Kazmi. Thermo-mechanical large deformation response and constitutive modeling of viscoelastic polymers over a wide range of strain rates and temperatures. *International Journal of Plasticity*, Vol. 22, No. 4, 2006, pp. 581–601.
- [58] Wang, F. S. and Z. F. Yue. Numerical simulation of damage and failure in aircraft windshield structure against bird strike. *Materials in Engineering*, Vol. 31, No. 2, 2010, pp. 687–695.
- [59] Lili, W., Z. Xixiong, S. Shaoqiu, G. Su, and B. Hesheng. Impact dynamics investigation on some problems in bird strike on windshields of high speed aircrafts. *Acta Aeronautica Et Astronautica Sinica*, Vol. 12, No. 2, 1991, pp. B27–B33.
- [60] Wang, J., Y. Xu, and W. Zhang. Finite element simulation of PMMA aircraft windshield against bird strike by using a rate and temperature dependent nonlinear viscoelastic constitutive model. *Composite Structures*, Vol. 108, 2014, pp. 21–30.
- [61] Dar, U. A., W. H. Zhang, and Y. J. Xu. Numerical implementation of strain rate dependent thermo viscoelastic constitutive relation to simulate the mechanical behavior of PMMA. *International Journal of Mechanics and Materials in Design*, Vol. 10, No. 1, 2014, pp. 93–107.
- [62] Colak, O. Modeling deformation behavior of polymers with viscoplasticity theory based on overstress. *International Journal of Plasticity*, Vol. 21, No. 1, 2005, pp. 145–160.
- [63] Ghorbel, E. A viscoplastic constitutive model for polymeric materials. *International Journal of Plasticity*, Vol. 24, No. 11, 2008, pp. 2032–2058.
- [64] Drozdov, A. D. Cyclic strengthening of polypropylene under strain-controlled loading. *Materials Science and Engineering: A*, Vol. 528, No. 29–30, 2011, pp. 8781–8789.
- [65] Khan, F. and C. Yeakle. Experimental investigation and modeling of non-monotonic creep behavior in polymers. *International Journal of Plasticity*, Vol. 27, No. 4, 2011, pp. 512–521.
- [66] Dusunceli, N. and O. U. Colak. Modelling effects of degree of crystallinity on mechanical behavior of semicrystalline polymers. *International Journal of Plasticity*, Vol. 24, No. 7, 2008, pp. 1224–1242.
- [67] Bardenhagen, S. G., M. G. Stout, and G. T. Gray. Three-dimensional, finite deformation, viscoplastic constitutive models for

- polymeric materials. *Mechanics of Materials*, Vol. 25, No. 4, 1997, pp. 235–253.
- [68] Drozdov, A. D. and J. D. Christiansen. Cyclic viscoplasticity of high-density polyethylene: experiments and modeling. *Computational Materials Science*, Vol. 39, No. 2, 2007, pp. 465–480.
- [69] Kim, B., S. B. Lee, J. Lee, S. Cho, H. Park, S. Yeom, et al. A comparison among neo-Hookean model, Mooney-Rivlin model, and Ogden model for chloroprene rubber. *International Journal of Precision Engineering and Manufacturing*, Vol. 13, No. 5, 2012, pp. 759–764.
- [70] Rivlin, R. and D. Saunders. Large elastic deformations of isotropic materials. *Collected papers of RS Rivlin*, Springer, New York, 1997, pp. 157–194.
- [71] Wang, M. C. and E. Guth. Statistical theory of networks of non-Gaussian flexible chains. *The Journal of Chemical Physics*, Vol. 20, No. 7, 1952, pp. 1144–1157.
- [72] Flory, P. J. and J. Rehner. Statistical mechanics of cross-linked polymer networks I. Rubberlike elasticity. *The Journal of Chemical Physics*, Vol. 11, No. 11, 1943, pp. 512–520.
- [73] Arruda, E. M., M. C. Boyce, and R. Jayachandran. Effects of strain rate, temperature and thermomechanical coupling on the finite strain deformation of glassy polymers. *Mechanics of Materials*, Vol. 19, No. 2, 1995, pp. 193–212.
- [74] Wu, P. D. and E. van der Giessen. On neck propagation in amorphous glassy polymers under plane strain tension. *International Journal of Plasticity*, Vol. 11, No. 3, 1995, pp. 211–235.
- [75] Sweeney, J. and I. M. Ward. Rate-dependent and network phenomena in the multiaxial drawing of poly(vinyl chloride). *Polymer*, Vol. 36, No. 2, 1995, pp. 299–308.
- [76] Billon, N. New constitutive modeling for time-dependent mechanical behavior of polymers close to glass transition: fundamentals and experimental validation. *Journal of Applied Polymer Science*, Vol. 125, No. 6, 2012, pp. 4390–4401.
- [77] Maurel-Pantel, A., E. Baquet, J. Bikard, J. L. Bouvard, and N. Billon. A thermo-mechanical large deformation constitutive model for polymers based on material network description: application to a semi-crystalline polyamide 66. *International Journal of Plasticity*, Vol. 67, 2015, pp. 102–126.
- [78] Gehring, F., J. Bouvard, and N. Billon. Modeling of time dependent mechanical behavior of polymers: comparison between amorphous and semicrystalline polyethylene terephthalate. *Journal of Applied Polymer Science*, Vol. 133, No. 35, 2016.
- [79] Federico, C. E., J. L. Bouvard, C. Combeaud, and N. Billon. Modelling strain rate and temperature dependent mechanical response of PMMAs at large deformation from below to above T_g . *Polymer*, Vol. 202, 2020, id. 122710.
- [80] Ding, F., T. Liu, H. Zhang, L. Liu, and Y. Li. Stress-strain curves for polyurethane elastomers: a statistical assessment of constitutive models. *Journal of Applied Polymer Science*, Vol. 138, No. 39, 2021, id. 51269.
- [81] Sweeney, J., P. Spencer, G. Thompson, D. Barker, and P. Coates. Constitutive modelling of polylactic acid at large deformation using multiaxial strains. *Polymers-Basel*, Vol. 13, No. 17, 2021, id. 2967.
- [82] Annarasa, V., A. A. Popov, and D. S. A. De Focatiis. A phenomenological constitutive model for the viscoelastic deformation of elastomers. *Mechanics of Time-Dependent Materials*, Vol. 24, No. 4, 2020, pp. 463–479.
- [83] Hasan, O. A. and M. C. Boyce. A constitutive model for the nonlinear viscoelastic viscoplastic behavior of glassy polymers. *Polymer Engineering & Science*, Vol. 35, No. 4, 1995, pp. 331–344.
- [84] Polanco-Loria, M., A. H. Clausen, T. Berstad, and O. S. Hopperstad. Constitutive model for thermoplastics with structural applications. *International Journal of Impact Engineering*, Vol. 37, No. 12, 2010, pp. 1207–1219.
- [85] Anand, L., N. M. Ames, V. Srivastava, and S. A. Chester. A thermo-mechanically coupled theory for large deformations of amorphous polymers. Part I: formulation. *International Journal of Plasticity*, Vol. 25, No. 8, 2009, pp. 1474–1494.
- [86] Ames, N. M., V. Srivastava, S. A. Chester, and L. Anand. A thermo-mechanically coupled theory for large deformations of amorphous polymers. Part II: applications. *International Journal of Plasticity*, Vol. 25, No. 8, 2009, pp. 1495–1539.
- [87] Frank, G. J. and R. A. Brockman. A viscoelastic-viscoplastic constitutive model for glassy polymers. *International Journal of Solids and Structures*, Vol. 38, No. 30, 2001, pp. 5149–5164.
- [88] Miled, B., I. Doghri, and L. Delannay. Coupled viscoelastic-viscoplastic modeling of homogeneous and isotropic polymers: numerical algorithm and analytical solutions. *Computer Methods in Applied Mechanics and Engineering*, Vol. 200, No. 47–48, 2011, pp. 3381–3394.
- [89] Yu, C., G. Kang, F. Lu, Y. Zhu, and K. Chen. Viscoelastic – viscoplastic cyclic deformation of polycarbonate polymer: experiment and constitutive model. *Journal of Applied Mechanics*, Vol. 83, No. 4, 2016, id. 41002.
- [90] Ohno, N. and J. D. Wang. Kinematic hardening rules with critical state of dynamic recovery, part I: formulation and basic features for ratchetting behavior. *International Journal of Plasticity*, Vol. 9, No. 3, 1993, pp. 375–390.
- [91] Zaïri, F., K. Woznica, and M. Naït-Abdelaziz. Phenomenological nonlinear modelling of glassy polymers. *Cr Mecanique*, Vol. 333, No. 4, 2005, pp. 359–364.
- [92] Pouriaevali, H., S. Arabnejad, Y. B. Guo, and V. P. W. Shim. A constitutive description of the rate-sensitive response of semi-crystalline polymers. *International Journal of Impact Engineering*, Vol. 62, 2013, pp. 35–47.
- [93] Alisafaei, F., C. Han, and N. Garg. On couple-stress elasto-plastic constitutive frameworks for glassy polymers. *International Journal of Plasticity*, Vol. 77, 2016, pp. 30–53.
- [94] Gudimetla, M. R. and I. Doghri. A finite strain thermodynamically based constitutive framework coupling viscoelasticity and viscoplasticity with application to glassy polymers. *International Journal of Plasticity*, Vol. 98, 2017, pp. 197–216.
- [95] Yu, C., G. Kang, K. Chen, and F. Lu. A thermo-mechanically coupled nonlinear viscoelastic-viscoplastic cyclic constitutive model for polymeric materials. *Mechanics of Materials*, Vol. 105, 2017, pp. 1–15.
- [96] Zhao, W., M. Ries, P. Steinmann, and S. Pfaller. A viscoelastic-viscoplastic constitutive model for glassy polymers informed by molecular dynamics simulations. *International Journal of Solids and Structures*, Vol. 226–227, 2021, id. 111071.
- [97] Vladimirov, I. N., M. P. Pietryga, and S. Reese. Anisotropic finite elastoplasticity with nonlinear kinematic and isotropic hardening and application to sheet metal forming. *International Journal of Plasticity*, Vol. 26, No. 5, 2010, pp. 659–687.
- [98] Abdul-Hameed, H., T. Messenger, F. Zaïri, and M. Naït-Abdelaziz. Large-strain viscoelastic-viscoplastic constitutive modeling of semi-crystalline polymers and model identification by deterministic/evolutionary approach. *Computational Materials Science*, Vol. 90, 2014, pp. 241–252.

- [99] Zairi, F., M. Nait-Abdelaziz, J. M. Gloaguen, and J. M. Lefebvre. Modelling of the elasto-viscoplastic damage behaviour of glassy polymers. *International Journal of Plasticity*, Vol. 24, No. 6, 2008, pp. 945–965.
- [100] Zairi, F., M. Nait-Abdelaziz, J. M. Gloaguen, and J. M. Lefebvre. A physically-based constitutive model for anisotropic damage in rubber-toughened glassy polymers during finite deformation. *International Journal of Plasticity*, Vol. 27, No. 1, 2011, pp. 25–51.
- [101] Tehrani, A. H. and R. K. Abu Al-Rub. Mesomechanical modeling of polymer/clay nanocomposites using a viscoelastic-viscoplastic-viscodamage constitutive model. *Journal of Engineering Materials and Technology*, Vol. 133, No. 4, 2011, id. 41011.
- [102] Balieu, R., F. Lauro, B. Bennani, R. Delille, T. Matsumoto, and E. Mottola. A fully coupled elastoviscoplastic damage model at finite strains for mineral filled semi-crystalline polymer. *International Journal of Plasticity*, Vol. 51, 2013, pp. 241–270.
- [103] Krairi, A. and I. Doghri. A thermodynamically-based constitutive model for thermoplastic polymers coupling viscoelasticity, viscoplasticity and ductile damage. *International Journal of Plasticity*, Vol. 60, 2014, pp. 163–181.
- [104] Praud, F., G. Chatzigeorgiou, J. Bikard, and F. Meraghni. Phenomenological multi-mechanisms constitutive modelling for thermoplastic polymers, implicit implementation and experimental validation. *Mechanics of Materials*, Vol. 114, 2017, pp. 9–29.
- [105] Khaleghi, H., A. Amiri-Rad, and M. Mashayekhi. A thermodynamically consistent continuum damage model for time-dependent failure of thermoplastic polymers. *International Journal of Plasticity*, Vol. 154, 2022, id. 103278.
- [106] Abu Al-Rub, R. K., A. H. Tehrani, and M. K. Darabi. Application of a large deformation nonlinear-viscoelastic viscoplastic viscodamage constitutive model to polymers and their composites. *International Journal of Damage Mechanics*, Vol. 24, No. 2, 2015, pp. 198–244.
- [107] Seidel, G. D., D. H. Allen, K. L. E. Helms, and S. E. Groves. A model for predicting the evolution of damage in viscoelastic particle-reinforced composites. *Mechanics of Materials*, Vol. 37, No. 1, 2005, pp. 163–178.
- [108] Voyiadjis, G. Z., A. Shojaei, and G. Li. A generalized coupled viscoplastic-viscodamage-viscohealing theory for glassy polymers. *International Journal of Plasticity*, Vol. 28, No. 1, 2012, pp. 21–45.
- [109] Cayzac, H., K. Sai, and L. Laiarinandrasana. Damage based constitutive relationships in semi-crystalline polymer by using multi-mechanisms model. *International Journal of Plasticity*, Vol. 51, 2013, pp. 47–64.
- [110] He, G., Y. Liu, X. Deng, and L. Fan. Constitutive modeling of viscoelastic-viscoplastic behavior of short fiber reinforced polymers coupled with anisotropic damage and moisture effects. *Acta Mech Sinica-Prc*, Vol. 35, No. 3, 2019, pp. 495–506.
- [111] Hao, P., V. Laheri, Z. Dai, and F. A. Gilabert. A rate-dependent constitutive model predicting the double yield phenomenon, self-heating and thermal softening in semi-crystalline polymers. *International Journal of Plasticity*, Vol. 153, 2022, id. 103233.
- [112] Ferreira, B. P., A. F. Carvalho Alves, and F. M. Andrade Pires. An efficient finite strain constitutive model for amorphous thermoplastics: fully implicit computational implementation and optimization-based parameter calibration. *Computers and Structures*, Vol. 281, 2023, id. 107007.
- [113] Chen, K., G. Kang, C. Yu, and H. Jiang. Effect of crystalline content on ratchetting of ultra-high molecular weight polyethylene polymers: experimental investigation and constitutive model. *Mechanics of Materials*, Vol. 133, 2019, pp. 37–54.
- [114] Xiao, R. and C. Tian. A constitutive model for strain hardening behavior of predeformed amorphous polymers: incorporating dissipative dynamics of molecular orientation. *Journal of the Mechanics and Physics of Solids*, Vol. 125, 2019, pp. 472–487.
- [115] Medvedev, G. A. and J. M. Caruthers. Development of a stochastic constitutive model for prediction of postyield softening in glassy polymers. *Journal of Rheology (Easton, Pennsylvania)*, Vol. 57, No. 3, 2013, pp. 949–1002.
- [116] Liu, X., S. Tian, F. Tao, and W. Yu. A review of artificial neural networks in the constitutive modeling of composite materials. *Composites, Part B: Engineering*, Vol. 224, 2021, id. 109152.
- [117] Jung, S. and J. Ghaboussi. Neural network constitutive model for rate-dependent materials. *Computers and Structures*, Vol. 84, No. 15-16, 2006, pp. 955–963.
- [118] Ghaboussi, J., J. H. Garrett, and X. Wu. Knowledge-based modeling of material behavior with neural networks. *J Eng Mech*, Vol. 117, No. 1, 1991, pp. 132–153.
- [119] Yun, G. J., J. Ghaboussi, and A. S. Elnashai. A new neural network-based model for hysteretic behavior of materials. *International Journal for Numerical Methods in Engineering*, Vol. 73, No. 4, 2008, pp. 447–469.
- [120] Hashash, Y. M. A., S. Jung, and J. Ghaboussi. Numerical implementation of a neural network based material model in finite element analysis. *International Journal for Numerical Methods in Engineering*, Vol. 59, No. 7, 2004, pp. 989–1005.
- [121] Ghaboussi, J., D. A. Pecknold, M. F. Zhang, and R. M. Haj-Ali. Autoprogressive training of neural network constitutive models. *International Journal for Numerical Methods in Engineering*, Vol. 42, No. 1, 1998, pp. 105–126.
- [122] Al-Haik, M. S., M. Y. Hussaini, and H. Garmestani. Prediction of nonlinear viscoelastic behavior of polymeric composites using an artificial neural network. *International Journal of Plasticity*, Vol. 22, No. 7, 2006, pp. 1367–1392.
- [123] Rodríguez-Sánchez, A. E., E. Ledesma-Orozco, S. Ledesma, and A. Vidal-Lesso. Application of artificial neural networks to map the mechanical response of a thermoplastic elastomer. *Materials Research Express*, Vol. 6, No. 7, 2019, id. 75320.
- [124] Jordan, B., M. B. Gorji, and D. Mohr. Neural network model describing the temperature- and rate-dependent stress-strain response of polypropylene. *International Journal of Plasticity*, Vol. 135, 2020, id. 102811.
- [125] Johnsen, J., A. H. Clausen, F. Grytten, A. Benallal, and O. S. Hopperstad. A thermo-elasto-viscoplastic constitutive model for polymers. *Journal of the Mechanics and Physics of Solids*, Vol. 124, 2019, pp. 681–701.
- [126] Li, X., C. C. Roth, and D. Mohr. Machine-learning based temperature- and rate-dependent plasticity model: application to analysis of fracture experiments on DP steel. *International Journal of Plasticity*, Vol. 118, 2019, pp. 320–344.
- [127] Haj-Ali, R. and H. Kim. Nonlinear constitutive models for FRP composites using artificial neural networks. *Mechanics of Materials*, Vol. 39, No. 12, 2007, pp. 1035–1042.
- [128] Yang, C., Y. Kim, S. Ryu, and G. X. Gu. Prediction of composite microstructure stress-strain curves using convolutional neural networks. *Material and Design*, Vol. 189, 2020, id. 108509.
- [129] Ji, C., J. Hu, B. Wang, Y. Zou, Y. Yang, and Y. Sun. Mechanical behavior prediction of CF/PEEK-titanium hybrid laminates

- considering temperature effect by artificial neural network. *Composite Structures*, Vol. 262, 2021, id. 113367.
- [130] Settgast, C., G. Hütter, M. Kuna, and M. Abendroth. A hybrid approach to simulate the homogenized irreversible elastic-plastic deformations and damage of foams by neural networks. *International Journal of Plasticity*, Vol. 126, 2020, id. 102624.
- [131] Yan, S., X. Zou, M. Ilkhani, and A. Jones. An efficient multiscale surrogate modelling framework for composite materials considering progressive damage based on artificial neural networks. *Composites, Part B: Engineering*, Vol. 194, 2020, id. 108014.
- [132] Im, S., J. Lee, and M. Cho. Surrogate modeling of elasto-plastic problems via long short-term memory neural networks and proper orthogonal decomposition. *Computer Methods in Applied Mechanics and Engineering*. Vol. 385, 2021, id. 114030.
- [133] Tao, F., X. Liu, H. Du, and W. Yu. Learning composite constitutive laws via coupling Abaqus and deep neural network. *Composite Structures*, Vol. 272, 2021, id. 114137.
- [134] Stoffel, M., F. Bamer, and B. Markert. Deep convolutional neural networks in structural dynamics under consideration of viscoplastic material behaviour. *Mechanics Research Communications*, Vol. 108, 2020, id. 103565.
- [135] Wang, J., T. Li, F. Cui, C. Hui, J. Yeo, and A. T. Zehnder. Metamodeling of constitutive model using Gaussian process machine learning. *Journal of the Mechanics and Physics of Solids*, Vol. 154, 2021, id. 104532.
- [136] Colak, O. U. and Y. Cakir. Material model parameter estimation with genetic algorithm optimization method and modeling of strain and temperature dependent behavior of epoxy resin with cooperative-VBO model. *Mechanics of Materials*, Vol. 135, 2019, pp. 57–66.
- [137] Xu, K., D. Z. Huang, and E. Darve. Learning constitutive relations using symmetric positive definite neural networks. *Journal of Computational Physics*, Vol. 428, 2021, id. 110072.
- [138] Teng, F., G. Menary, S. Malinov, S. Yan, and J. B. Stevens. Predicting the multiaxial stress-strain behavior of polyethylene terephthalate (PET) at different strain rates and temperatures above T_g by using an artificial neural network. *Mechanics of Materials*, Vol. 165, 2022, id. 104175.
- [139] Zhang, A. and D. Mohr. Using neural networks to represent von Mises plasticity with isotropic hardening. *International Journal of Plasticity*, Vol. 132, 2020, id. 102732.
- [140] Bergstrom, J. S. *Mechanics of solid polymers: theory and computational modeling*, William Andrew, 2015.

Appendix

Table A1: Summary of rubber-like models

Year	Model/theory	Characteristics
1943	Theory of James and Guth [20]	Basal and general theory for rubber-like models
1981	Theory of Ball <i>et al.</i> [21]	Simulation of entanglements was included on the basis of James <i>et al.</i>
1986	Edwards–Vilgis model [18]	The molecular mechanism of stretching and hardening was explained

Table A2: Summary of models based on Haward–Thackray theory

Year	Model/theory	Characteristics
Class BPA models		
1955	Theory of Ree and Eyring [24]	Foundation of Haward–Thackray theory
1968	Haward–Thackray model [17]	This model contains a Hookean spring, an Eyring dashpot, and a Langevin spring
1988	BPA model [25]	Maxwell element is parallel with the Langevin spring in this model
Class GR models		
1995	GR model of Buckley and Jones [26]	The linear elasticity, Eyring viscous flow, and the Edwards–Vilgis entropy function were employed in this model
2004	Model of Wu and Buckley [27]	A distribution of relaxation times was added on the basis of GR model
2010	Model of De Focatiis <i>et al.</i> [28]	This model consists of a set of ROLIEPOLY equations
Class EGP models		
1997	Model of Tervoort <i>et al.</i> [29]	It is a single Maxwell model with a relaxation time
2000	Model of Govaert <i>et al.</i> [31]	The physical quantities were introduced into the extended CLM

Table A3: Summary of phenomenological models

Year	Model/theory	Characteristics
2011	Model van Breemen <i>et al.</i> [33]	It is a multi-mode model on the basis of one-mode EGP model
VE models		
1969	Model of Schapery [56]	It does well in the prediction of creep and relaxation
2006	Model of Khan <i>et al.</i> [57]	This model can get good prediction over a wide range of strain rate
2010	Z-W-T model of Wang and Yue [58]	It is not suitable for the VP response after yielding
2018	Model of Chang <i>et al.</i> [48]	It is good at predicting the mechanical behavior of polymers before yielding
VP models		
1997	Model of Bardenhagen <i>et al.</i> [67]	It is good at predicting the creep and relaxation
2005	Model of Colak [62]	The nonlinear response such as creep caused by viscoplasticity can be simulated well
2007	Model of Drozdov and Christiansen [68]	It is suitable for the prediction with small strain
Hyperelastic models		
2015	Neo-Hooke model [140]	It is suitable for the deformation of rubber-like materials
1995	Model of Arruda and Boyce [73]	It can well predict the phenomenon of strain hardening
1995	Model of Wu and van der Giessen [74]	It does well in the prediction of strain hardening and softening
2002	Sweeney model [75]	Strain concentration was included in their model
2012	Billon model [76]	Edwards–Vilgis model was extended to a general time dependent constitutive model
2015	Maurel-Pantel model [77]	The degree of mobility of entanglement points was represented
2016	Gehring model [78]	Microstructure rearrangement was considered
Multi-mode models		
2001	Model of Frank and Brockman [87]	The form is simple for this model, and it is suitable for problems with high strain rate
2009	Model of Anand <i>et al.</i> [85]	The yielding, strain hardening, unloading response, and temperature transforming can be reflected well in this model
2011	Model of Miled <i>et al.</i> [88]	The viscous behavior of polymers can be reflected better in this model
2016	Model of Yu <i>et al.</i> [89]	It is good at predicting the creep, loading and unloading behavior
Constitutive models with damage		
2008	Model of Zairi <i>et al.</i> [99]	The behavior of elasto-viscoplastic damage is considered in this model
2011	Model of Tehrani and Abu Al-Rub [101]	Viscodamage is reflected by a damage evolution variable in this model
2013	Model of Balieu <i>et al.</i> [102]	It is suitable for the prediction of damage with a high strain rate and finite strain
2014	Model of Krairi and Doghri [103]	Ductile damage evolution is included in this model
Elasto-plastic models		
1979	Model of G'sell and Jonas [34]	The form of this model is simple, and it is suitable for the deformation with low strain rate
1983	Johnson–Cook model [36]	It is originally used for the deformation of metal under high temperature and high strain rate
2001	DSGZ model [41]	All the characteristics of elasticity, yielding, strain hardening, and strain softening can be reflected well by this model
2018	Model of Wang <i>et al.</i> [50]	It can get good results over a wide range of temperature and strain rate
2006	M–B model [49]	Another Maxwell element was added in parallel on the basis of BPA model
2009	Model of Varghese and Batra [53]	Three Maxwell element were contained in the model

Table A4: Summary of NN-based constitutive models

Year	Model/theory	Characteristics
1991	Model of Ghaboussi <i>et al.</i> [118]	It is an earlier attempt for the applying of NN model on constitutive models. Back-propagation neural network is applied in this model
2006	Model of Al-Haik <i>et al.</i> [122]	The NN-based model is good at predicting the stress relaxation of polymers
2019	Model of Rodriguez <i>et al.</i> [123]	The constitutive model built with feedforward artificial neural network can get extremely high precision
2019	Model of Li <i>et al.</i> [126]	NN model is used to enhance the performance of Johnson–Cook model
2020	Model of Jordan <i>et al.</i> [124]	NN model is used to reflect the VE deformation