



# A damage-based elastic-viscoplastic constitutive model for amorphous glassy polycarbonate polymers



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## ABSTRACT

This paper presents a new damage-based elastic-viscoplastic constitutive model for amorphous glassy polycarbonate (PC) within the framework of irreversible thermodynamics and continuum damage mechanics (CDM). To this end, experimental investigation, theoretical formulation and numerical implementation are performed. In the experiment part, noticeable strain rate and temperature dependent mechanical responses were observed in uniaxial compression tests over a wide range of strain rates and temperatures. Moreover, damage evolution associated with the decreasing elastic modulus was highlighted in cyclic loading-unloading tests. Based on the experimental data, an elastic-viscoplastic model coupled with damage formulation is developed. Constitutive equations, specifically the strain rate and temperature dependent yield criteria, the viscoplastic flow rule and the damage evolution law, are derived from the Helmholtz free energy and the Clausius-Duhem entropy inequality. Introducing an elastic-damage predictor/viscoplastic corrector scheme, a time-discrete frame of the constitutive equations is presented. The nonlinear time-discrete constitutive system is further simplified into a single-scalar Newton-Raphson scheme in view of computational efficiency. The model is then implemented into the finite element program LS-DYNA, by using a user-defined material subroutine (UMAT). The good correlation between model predictions and experimental data demonstrates the capabilities of the proposed model capturing mechanical behavior and damage evolution of PC over a wide range of strain rates and temperatures.

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## 1. Introduction

Polycarbonates (PCs), are typical thermoplastic amorphous polymers and demonstrates extraordinary optical and mechanical properties such as their high specific stiffness, specific strength, and impact resistance. Nowadays, they find wide applications ranging from aircraft windshields, astronaut masks to fascia boards, architectural glasses, medical apparatus and instruments [1]. Mechanical behavior of these components has drawn attentions of many research groups. Since the middle of the 20th century, strain rate, temperature and pressure dependent mechanical behavior of amorphous polymers has been actively studied to reveal the deformation mechanism and realize constitutive modeling of amorphous polymers. However, damage and fracture mechanisms of amorphous polymers are not yet well understood and not involved in most existing constitutive models.

In early viscoplastic theories, [2] firstly indicated that the deformation and yield of polymers result from the increase of activation energies caused by the applied stress under appropriate conditions. Subsequently, some other theories were developed to describe the yield and plastic

mechanisms of amorphous polymers. For example, [3] emphasized the role of shear stress which induces structural changes in a glassy polymer to break up the rigidity of the glass and initiate plastic flow. It was assumed that the resistance of plastic flow was mainly due to the bonds of molecular chains. On the contrary, [4,5] believed that the plastic deformation of polymer materials was controlled by intermolecular forces. These theories were verified through experimental observations and then incorporated into constitutive models.

By means of X-ray and infra-red technology, some experimental investigations [6,7] revealed the unique yield and deformation mechanism of glassy polymers. Based upon the measurement of the yield stresses of two glassy polymers over a wide range of temperatures and strain rates, [8] concluded that every secondary  $\beta$ -transition corresponded to the liberation of one of the degrees of freedom of a segment in the main chain, which is in agreement with the generalized Eyring's theory [9] modified Robertson's theory to quantitatively explain temperature and strain-rate dependence of the yield stress as well as the difference between tension and compression in terms of the dependence of yield on the hydrostatic component of the applied stress [10] applied the Gauss-Eyring model to predict the behavior of thermoplastics in tensile experiments.

Since the 1980s, the combination of experimental investigations with fundamental theories has rapidly promoted the research on

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mechanical behavior of polymer materials. These studies mainly focused on the effects of strain-rate, temperature, entanglement density and pressure on mechanical behavior of polymers [11–13] performed extensive experiments of glassy polymers, particularly PC, over a wide range of temperatures and strain rates. On the basis of the experiments, a constitutive theory integrating micromechanics and statistical thermodynamics was then proposed from the point of activation energy and intermolecular forces. Numerical studies were presented to investigate the deformation mechanism of toughened PC on both micro-mechanical and macro-mechanical scales [14,15]. A constitutive theory integrating micromechanics and statistical thermodynamics was then proposed from the point of activation energy and intermolecular forces [16] measured high strain-rate shear response of PC and polymethylmethacrylate (PMMA). The experimental results revealed that failure in the materials is due to tensile cracking rather than thermal instability. Meanwhile, [17,18] analyzed the stiffness variation of polymers over a wide range of temperatures and pressure by the Brillouin measurements and dynamic mechanical analysis (DMA) measurements [19–21] characterized the yield stress of amorphous polymers and indicated that the yield behavior of materials was determined by transition energy and microstructure. [22,23] investigated the temperature and molecular weight dependence of the strain hardening behavior of PC where the strain hardening behavior was related to the relaxation of the entanglement network and local rearrangements. Moreover, [24,25] investigated the localization and strain softening in PC. It was believed that strain softening is an intrinsic property of PC rather than localization [26] calculated glass transition temperatures, Young modulus, yield stresses and strain hardening modulus using Molecular-dynamics (MD) simulations. In addition, the nonlinear thermo-mechanical creep behavior of PC was investigated by [27] at different temperatures under pure shear loading. It was found that nonlinear viscoelastic behavior initiated at the strain of nearly 1%. To investigate the strain rate dependent mechanical response of polymer materials, [28] conducted high strain rate tensile tests and proposed a strain rate dependent constitutive model predicting the material behavior. Low-velocity impact loading tests were also carried out by [29], followed by the numerical simulation of polymer components under impact loading.

Based on these works, varieties of constitutive models have been proposed to represent the nonlinear viscoelastic and viscoplastic behavior of glassy polymers.

[30] developed a three-dimensional constitutive model, the so-called “B-P-A” model based on the macromolecular structure and the micro-mechanism of plastic flow [31] studied three-dimensional molecular network theories using the non-Gaussian statistical mechanics model for the large strain extension of molecules. This network model was further applied to describe the orientational hardening in amorphous glassy polymers [32–34] introduced an internal variable of local free-volume to develop a continuum mechanics theory for the elastic-viscoplastic deformation of amorphous solids. The theory accounted for the dependence of the Helmholtz free energy on the plastic deformation in a thermodynamically consistent manner [35] derived a constitutive model to describe both the transition process and the steady creep of polymers, the effect of molecular weight of polymers on the parameters of this model is studied as well. [36] proposed a phenomenological constitutive model, [37] modified this model to describe the large deformation of glassy polymers over large range of temperature and strain rate [38] introduced a “compressible-Leonov model” where they replaced the Jaumann-stress rate with a Truesdell-stress rate to acquire an adequate description in the elastic region [39,40] developed constitutive models on the basis of physical considerations and extensive numerical testing [22] introduced a three-dimensional constitutive model using a Langevin-type free energy function for the energy storage due to molecular alignment connected in parallel to a Maxwell element with a viscoplastic dashpot. [16] proposed a constitutive model including both the primary process ( $\alpha$ ) and the two secondary rate-activated processes

( $\beta$  and  $\gamma$ ). It was shown that the secondary transitions in material affect both yield and post yield behavior of the material at high strain rate. For polymer matrix composites (PMCs), a constitutive model has been developed to capture the rate-dependent large deformation behavior of the thermoplastic olefin (TPO) [41].

Most of the aforementioned models were developed on the foundation of continuum mechanics, neglecting the weakening of polymers during usage and storage. In fact, the strain softening and crazing phenomena of PC both indicate the breaking of molecular chains and the growth of microcracks. Therefore, it is essential to take into account the damage evolution of polymer materials in constitutive modeling. To this end, several viscoelastic and viscoplastic constitutive models with growing damage were developed [42] developed a fully three-dimensional finite-strain viscoelastic damage model where CDM was utilized to introduce a simple isotropic damage mechanism and to study progressive degradation of the storage modulus [43] proposed a relatively simple nonlinear viscoelastic constitutive model for composites under multi-axial stress states. Time and temperature-dependent equations were employed in describing the evolution of microstructural changes and microcracks [44] built quantitative empirical models of the residual mechanical properties of crazed PC by means of the Design of Experiments (DOE). It was found that the value of failure stress varied as a function of relative craze density, crazing stress, and strain rate while crazing did not affect the yielding behavior of PC [45] investigated the damage based constitutive relationship and damage micro-mechanism of polymers, by using Synchrotron Radiation Tomography technique. They proposed a constitutive model based on multi-mechanism scheme to account for the semi-crystalline characteristics of the material and on mechanics of porous media to handle the irreversible volume change (damage).

In this paper, a new damage-based elastic-viscoplastic constitutive model for amorphous glassy PC is developed within the framework of irreversible thermodynamics and continuum damage mechanics (CDM). Effects of strain rate, temperature and damage are taken into account. To this end, experimental investigation, theoretical formulation and numerical implementation are performed.

The present paper is organized as follows. In Section 2, uniaxial compression experiments of the PC material over a wide range of strain rate and temperature as well as cyclic loading-unloading tests are carried out. Section 3 is devoted to developing the constitutive model. A rheological representation of the model is introduced and the constitutive equations are derived from the Helmholtz free energy and the Clausius-Duhem entropy inequality. Numerical implementation of the present model is discussed in Section 4. Subsequently, numerical examples and comparison of the model predictions with the experimental data are given in Section 5. Finally, the conclusions of the study are given in Section 6.

## 2. Experiment

### 2.1. Material and specimen

In this study, PCs used for experimental investigation are manufactured by Key Laboratory of Polymer Material, Zhengzhou University. The density of the PC is 1190 kg/m<sup>3</sup>. Specimens were machined into cylinders with dimensions of 5.00 mm diameter and 4.00 mm length. The end faces of the cylindrical specimens were machined into smooth surfaces to reduce frictional effects during testing. Before experimental testing, specimens were stored in a drying cabinet at room temperature (298 K) to release residual stresses caused by the manufacturing process.

### 2.2. Quasi-static uniaxial compression tests

In this section, we carry out three groups of experiments: strain rate varying experiments, temperature varying experiments and cyclic

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