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Modeling of the mechanical behavior of amorphous glassy polymers under variable loadings and comparison with state-of-the-art model predictions

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ABSTRACT

The objective of this work is to model the mechanical behavior of glassy polymers during non-monotonic loadings involving softening/hardening characteristics, transient effects and plastic instabilities. The microstructure of a glassy polymer has a strong influence on the mechanical properties and it is typically described by network models. The present state of modeling amorphous glassy polymers is revealed through reviewing state-of-the-art network models. To evaluate the models, several numerical examples are presented. It is found that the models are able to capture the monotonic loading accurately, but for transient effects after loading rate changes and for long-term behavior, their responses deviate significantly from the experimental data. In order to improve the predictions under these conditions, a new constitutive model is proposed in this work. The numerical treatment of the proposed model associated with both the ODE-solver and finite element method is discussed. The model is calibrated to the experimental data for various states of deformation. The numerical results indicated that the proposed model is able to predict experimental response under long-term and repeated loadings well.

1. Introduction

The mechanical properties of amorphous glassy polymers are to a large extent dictated by their chemical composition and microstructure. In amorphous polymers, microstructure is disordered and formed of long polymer chains. The mechanical behavior is characterized by initial yielding and subsequent strain softening, followed by strain hardening due to the reorientation of polymer chains, cf. Haward and Thackray (1968) and Argon (1973). Once the chains reach their limit for extensibility, a dramatic increase of stress can be observed.

Amorphous glassy polymers also exhibit localized deformation which is due to shear band propagation under shearing, cf. e.g. Bowden and Raha (1970) and Wu and van der Giessen (1994), and neck propagation under tension, cf. G'Sell and Jonas (1979) and Stokes and Nied (1986). Bowden and Raha (1970) conducted plane strain compression tests on polymethyl methacrylate (PMMA) and polystyrene (PS) to investigate the formation of shear bands. According to their observations, the growth of shear bands is the primary source for the evolution of plastic deformation in amorphous glassy polymers. In tensile tests, the initial chain distribution have been found to be macroscopically uniform, whereas large-scale molecular dynamics (MD) simulations have indicated that the chain distribution locally is heterogeneous, cf. Itoh et al. (2002). As a result of heterogeneity, polymer material shows localized deformation where microscopic shear bands in closely packed regions develop and annihilate into macroscopic shear bands. The plane strain tension tests by Tomita and Uchida (2003) showed a remarkable drop in the macroscopic stress immediately after the development of macroscopic shear bands. During continued deformation, the propagation of shear bands and the development of inhomogeneous

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deformation were observed which is macroscopically manifested by necking.

Experiments on amorphous glassy polymers also show strong strain rate sensitivity involving transient effects after loading rate changes. Arruda et al. (1995) conducted uniaxial compression experiments on PMMA, which indicated that the yield stress increases and the strain hardening decreases with the increased strain rate. G'Sell and Jonas (1981), and later Zaïri et al. (2005), investigated the influence of strain rate on strain hardening, creep and relaxation for polycarbonate (PC), and Khan and Zhang (2001) and Khan (2006) for polytetrafluoroethylene (PTFE). More recently, Dreistadt et al. (2009) investigated the influence of relaxation time and repeated unloadings to different stress levels on bisphenol A polycarbonate. According to their experiments, changes in loading rate cause strong transient effects involving creep/recovery (termed dwell) and nonlinear response during unloading and subsequent reloading.

Experiments on amorphous polymers indicate that the micro-structure of a polymer has a strong influence on the mechanical properties. In constitutive models for amorphous polymers, the microstructure is usually represented by an overall chain network which consists of an assembly of individual chains arranged in cubic cells. The transition from microstructure to macroscopic level is performed via a homogenization which allows the micro-stretches to fluctuate around the macro-stretches. In the early models, the cells were described as tetrahedrons or cubes involving three or four chains, cf. James and Guth (1943) and Treloar (1946). However, the 3- and 4-chain models cannot accurately reproduce the strain hardening in different deformation modes. In addition to the models involving the reduced set of chains, full network models have been investigated and initially applied for rubber-like materials, cf. Treloar and Riding (1979). Since the integration of the stored energies of all the individual chains is computationally very expensive, full network models have not become general in practical applications. Despite the active research carried out after the development of the first network models, it took forty years before (Arruda and Boyce, 1991) developed a 8-chain network model (will here termed the BPA model), which is able to satisfactorily predict the mechanical behavior of amorphous polymers also in large three-dimensional deformations. This pioneering work was confirmed later by Arruda and Boyce (1993), Arruda et al. (1993), Hasan and Boyce (1995), Arruda et al. (1995) and Anand and Gurtin (2003) to mention a few.

Since the initial response of amorphous glassy polymers can be considered nearly elastic, most of the models are based on a linear elastic constitutive assumption. However, such models are not able to satisfactorily reproduce transient effects during non-monotonic loading, which shortcomings are primarily a consequence of neglected viscoelastic effects, cf. Hasan and Boyce (1995). Examples of early viscoelastic models are the Zener (1948) model, which consists of an elastic spring in parallel with a Maxwell element and the Burger's model, which consists of an elastic spring and a damper arranged in series with the Kelvin element. These relatively simple models are later employed instead of generalized Kelvin and Maxwell models (Prony-series) to avoid the identification of a large number of material parameters. In order to describe hysteresis of filled rubbers, Huber and Tsakmakis (2000) proposed a modified Zener model which was later used by Amin et al. (2002) to predict the rate-dependent behavior of high damping natural rubbers. Bergström and Boyce (1998) proposed a rate-dependent model in which two networks are used to describe the mechanical behavior of rubber; one network captures the elastic behavior, whereas a Maxwell element is used to predict the viscoelastic behavior. The Bergström and Boyce (1998) model was later extended to the modeling of mechanical behavior of filled polymers in high strain rates, cf. Quintavalla and Johnson (2004).

However, many of the viscoelastic models can satisfactorily be applied to model the mechanical behavior of amorphous glassy polymers in only small strains and strain rates, cf. Ward (1983), Amin et al. (2002) and Cao et al. (2012). To account for nonlinear deformation behavior in large strains, the models which include both viscoelastic and viscoplastic ingredients need to be applied. Among such models, Khan and Zhang (2001) and later Khan et al. (2006) proposed viscoelastic–plastic models which are able to capture the deformation behavior of PTFE and adiprene-L100 polymer, respectively. Based on the 8-chain model, Anand and Ames (2006) proposed a viscoelastic–plastic constitutive model which was originally used to predict micro-indentation. However, due to the large number of material parameters included, this model is difficult to calibrate and apply in practice. Moreover, the capability of the models mentioned above is addressed only in a restricted set of loading situations.

In the present paper, we will introduce an extension of the 8-chain BPA model (termed the EBPA model), which allows for modeling of the viscoelastic effects and intrinsic isotropic hardening in amorphous glassy polymers. For comparison, the predictive capability of the BPA model and the Anand and Ames (2006) viscoelastic–plastic model under homogeneous deformation modes is evaluated. These models can be considered as representative of a whole range of current models. The importance of different rheological properties employed in the models for regarding mechanical behavior of amorphous polymers are investigated. Compared to the original BPA model, only three new material parameters are needed in the EBPA model.

In order to evaluate the EBPA model predictions also for inhomogeneous deformation, cold drawing experiments on PC are performed and the EBPA model is implemented in a finite element program. Based on the finite element implementation of both the BPA and EBPA model, the cold drawing process of the dumbbell-shaped specimen is simulated and the numerical results are compared with the experimental data.

2. Description of state-of-the-art network models

Both the BPA model and Anand and Ames (2006) model are shown to be in good agreement with experiments under monotonic loadings, cf. Arruda and Boyce (1993), Arruda et al. (1993) and Anand and Ames (2006). Furthermore,