Mechanics of Materials 71 (2014) 114–125

Contents lists available at ScienceDirect

Mechanics of Materials

journal homepage: www.elsevier.com/locate/mechmat

Characterisation and micromechanical modelling of the elasto-viscoplastic behavior of thermoplastic elastomers

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ARTICLE INFO

Article history:
Received 20 July 2012
Available online 19 August 2013

Keywords:
Micromechanical model
Thermoplastic elastomer
Rubber recycling

ABSTRACT

The first objective of this study is to characterise the physico-chemical and mechanical properties of thermoplastic elastomers (TPE) and their constituents. In parallel with the experimental study, a model describing the mechanical behaviour of such materials at room temperature and without damage is proposed. The composite materials studied in the present work are processed by blending particles of vulcanized rubber ethylene-propylene-diene (EPDM) into an isotactic polypropylene (PP) matrix. These particles, obtained from a recycling process, have an average diameter of 70 μm. The constitutive equation for TPE composites is developed within the framework of a self-consistent micromechanical approach which considers the mechanical behaviour of each phase. A preliminary analysis of various TPE in linear elasticity justifies the choice of a morphological pattern for the model, which views elastomer particles as embedded in the thermoplastic matrix. In the non-linear domain, the PP matrix is modelled by means of an elastoviscoplastic model whose parameters are fitted using tensile and instrumented spherical micro-indentation tests. The elastomer exhibits viscoelastic behaviour. Having determined the material parameters by inverse analysis, the proposed micromechanical model is compared with tensile and bending tests performed before damage initiation and for various elastomers contents.

1. Introduction

Thermoplastic elastomers belong to a class of materials in which a thermoplastic is combined with elastomer particles. TPE composites take advantage of constituents and their behaviour evolves with their volume fractions. Hence, their properties can be optimised according to the desired application. They also have the advantage of being processed like conventional thermoplastics. The first TPE launched on the market in the 1960s was a polyurethane (TPU). Nowadays, numerous types of TPE are available. Some composites such as thermoplastic vulcanisates (TPV) are formed by a dynamically vulcanised physical mixture; others types of TPE are self-organised in block copolymers such as thermoplastic styrene elastomers (TPS). TPE are mainly used in the automotive industry, as well as in medical applications and in certain construction materials for civil engineering.

Numerous approaches which attempt to model the mechanical behaviour of TPE are found in the literature. Bensason et al. (1997) were amongst the first to develop a model for an ethylene-octene block copolymer. Considering the macroscopic strain as purely elastic, uniaxial tension until breakage was studied. The concept used describes molecular rearrangement within the chain network, allowing chain segments to connect with each other. This approach is close to that of Drozdov and Christiansen (2006) who treat the viscoelastic character of a TPE

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http://dx.doi.org/10.1016/j.mechmat.2013.06.010
0167-6636/

copolymer similarly. In this work, the thermally activated rearrangements of the network are governed by the equation proposed by Eyring (1936). The authors also took into account viscoplasticity by assuming it to arise from sliding of the chain network, the rate of sliding being proportional to the macroscopic strain. The model was compared with experimental tensile tests and relaxation tests under compression, without analysing the unloading phase. Drozdov (2006) adopted another method and modelled block copolymers by considering a network of chains and using statistical physics tools at the molecular scale. Chain -segment flexibility was taken into account as well as their inability to overlap in space. By assuming an incompressible network, macroscopic hyperelastic behaviour was obtained. The developed model was applied to tensile tests without unloading phase and provided predictions in a good agreement with the experimental data. Drozdov and Gupta (2003) later modified this approach by taking into account the flexibility of chains which were fixed at their extremities. It was suggested to describe plastic deformation using an approach originating from damage mechanics. The proposed model did not take the viscosity into account, but correctly described tensile tests with unloading.

Phenomenological models were also developed by Lambert-Diani and Rey (1999) and Diani et al. (2004). They assumed both TPE and elastomers to have incompressible hyperelastic behaviour. The models were analysed through tensile and compression tests as well as in simple shear and plain strain compression. These models were found to provide good results. However, only the loading phase was studied and unloading was not considered. A phenomenological approach was also used by Boyce et al. (2001) to model the behaviour of TPV composed of a PP matrix filled with vulcanised ethylene-propylene-diene rubber (EPDM) particles. The mechanical behaviour of the TPV was considered to be elastoviscoplastic. This model is defined by three rheological elements in series or in parallel and was confronted with experimental tests in uniaxial compression and plane strain compression. While the model response in the loading phase is satisfactory, capturing the behaviour during unloading remains problematic. Drawback of these strategies is the requirement to identify the model parameters for each particle content.

Besides molecular network and phenomenological models, it is possible to develop models by describing the behaviour of the constituent phases and to obtain the material macroscopic behaviour using a homogenisation method. This approach, also referred to as a micromechanical method, is extensively used in the field of metallic materials (Zaoui and Masson, 2001; Feaugas et al., 1997) but has also been applied to polymers (Ausias et al., 2007; Omnès et al., 2008). Boyce et al. (2001) used such approach. In this study, a model developed for the TPV (Boyce et al., 2001) is used to represent the behaviour of the PP. The elastomer particles are modelled by the hyperelastic formulation of Arruda and Boyce (1993). The interface between particles and matrix is considered perfect. The material structure is described by a representative periodic pattern and various morphologies are used to study the impact of the distance between elastomer particles. The model response is analysed through tensile tests and it also appears to have difficulties to correctly describe the unloading phase.

van Dommelen et al. (2003) assumed that the presence of a particle within a semi-crystalline matrix induces an anisotropic transcrystalline layer around it. Thus, the authors model the behaviour of TPE composites by means of a micromechanical model considering a periodic pattern and modelling it using the finite element method. The behaviour of the matrix is described by an elastoviscoplastic model. The elasticity is isotropic while the plasticity is formulated by means of the Hill's orthotropic criterion. Elastomer particles are modelled using a hyperelastic neo-Hookeen model. The objective of the authors is to study the rigidity of the composite by modifying the properties of the interface while analysing the influence of the matrix anisotropy. However, this study remains purely theoretical without comparison with experimental data. Ma et al. (2010) developed a constitutive model for ternary phase thermoplastic olefin (TPO). They predicted the elastic modulus of the composite using micromechanical theory and further extended this model to the non-linear domain (Yu et al., 2010).

A review of the existing literature brings to light the difficulties encountered when attempting prediction of TPE mechanical behaviour. To simplify the materials of the present study, the TPE considered is composed of a polypropylene homopolymer matrix filled with EPDM particles. In the next section, sample preparation is detailed. The two following sections are devoted to improving the understanding of mechanisms involved during the deformation of the matrix and the TPE by use of physico-chemical and mechanical characterisation. Next, in Section 5, the micromechanical model is described and is used with success to describe a four-point bending test.

2. Sample preparation

The TPE is made of a blend of isotactic polypropylene (Moplen 501H) supplied by Basell Polyolefins and of EPDM particles. At room temperature, the density of PP is $0.91\ g/cm^3$ and the density of EPDM is $1.36\ g/cm^3$. Different TPE have been elaborated with several EPDM contents and Table 1 gives the volume fraction and the weight fraction of elastomer for the different TPE. They were mixed with a twin-screw extruder. ASTM dumbbell specimens were made by an Engel 350 injection moulding machine. Dimensions of the effective zone of the sample are $4\times10\times90\ mm^3$. To reduce the residual stresses, samples were subjected to an annealing at 90 °C for 5 hours, then cooled down to room temperature at a temperature rate of 10 °C per hour. This thermal treatment is, however, not sufficient to modify the microstructure, which is a priori different in the core and at the surface of samples (Mendoza et al., 2003). Samples of PP without filler have been prepared with the same annealing temperature and named PP90. Some polypropylene specimens were also annealed at 150 °C and at 160 °C for one hour (PP150 and PP160). These two thermal treatments were carried out to increase crystallinity of the semi-crystalline polymer.