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Multi-mode modeling of global and local deformation, and failure, in particle filled epoxy systems

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ABSTRACT

A three-dimensional representative volume element is used to analyze the local heterogeneous stress and strain distributions, and the onset to failure, in a standard epoxy system filled with sub-micron sized hard and soft particles. Computations are compared with experiments performed in lubricated compression tests that reveal the intrinsic material's response. The response on the macroscopic level, and that of the matrix on RVE level, is captured by a multi-mode constitutive version of the Eindhoven Glassy Polymer model that describes the non-linear viscoelastic pre-yield, yield and post-yield behavior accurately for all deformation rates used. Compared to the single-mode description, the multi-mode variant covers the pre-yield regime correctly and for the hard-particles also the post-yield response. At a local level, multi-modes give increased stress values and more intensified critical events, which is particularly important for quantitatively predicting the onset of failure. This is successfully done by detailed RVE analyses.

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1. Introduction

Epoxy resins are widely used in applications such as structural adhesives, coatings, electrical devices, and as matrix material in fiber reinforced or particulate composites. They have excellent engineering properties like modulus and strength, low creep, good dimensional stability and corrosion resistance. Moreover they are relatively easy to manufacture into composites [1,2]. Epoxies are usually modified to include a dispersed second phase. Hard fillers, like in particle- or more often fiber-reinforcement, are used to create lightweight, high-stiffness composites to replace metals parts in aircrafts and wind turbines [3,4]. Soft fillers, in the form of a dispersed rubber phase, are applied as a solution to the inherently low toughness and impact resistance of epoxies [5–7]. Toughening can sometimes even be obtained by using inorganic particles [8,9]. Furthermore, combining these hard and soft fillers into hybrid-particulate composites shows to enhance the toughness even further [10–12].

Mechanical characterization of composites is usually done via experiments that couple the global macroscopic mechanical response to microscopic properties, and events, like particle dispersion, cavitation, crack propagation, and particle debonding. However, no direct connection is obtained between global and local properties of the structure. This represents a major challenge for material scientists, and especially understanding the onset of failure, and more importantly preventing early failure, requires a quantitative coupling of the macroscopic intrinsic mechanical response, via detailed local analyses, to critical events happening at the micro-scale. Over the years effort has been devoted to the development of models that try to use the structure to estimate the stress fields at smaller length scales. Initially, solutions for effective properties were sought using approximations based on mean-field homogenization schemes [13–18]. A more direct coupling between macroscopic response and local properties became only possible with the introduction of computers, that allowed simulations with increasing complexity given the continuously increasing computational power. Finite element methods (FEM) provided an excellent tool to arrive at the micro–macro coupling, first for (simplified) microstructures [3,19–21]. Later more complexity could be built in and the extension to three dimensional analyses became possible [4,22–24]. For further details we refer to our previous study [25], where the experimental mechanical response of model systems of particle-filled polycarbonate (PC) was compared with that resulting from FEM simulations. The microstructure was modeled with a three-dimensional periodic representative volume element (RVE) in a finite element mesh assuming randomly dispersed, mono-sized spheres that perfectly adhere to the matrix. To account for the influence of statistical variations in the systems on the homogenized behavior, it was

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shown that 32 particles, with three elements between neighboring particles, are a good compromise between accuracy and computational cost. The constitutive response of the PC matrix was modeled with a one mode version of the Eindhoven Glassy Polymer (EGP) model [26–28]. The macroscopic stress–strain response was captured relatively well by the simulations, including all the rate dependencies. The simulations showed that the presence of fillers causes positive hydrostatic stresses to occur in the polymer matrix, even in the case of negative loading situations as in compression experiments. These positive hydrostatic stresses reach the critical value of PC already at small macroscopic deformations, implying a sequential occurrence of local failure; the local failure events finally combine to grow into a macroscopic crack.

Originally, the EGP model was developed for thermoplastics, but it was already shown by Govaert et al. [19] that it is well capable of describing the intrinsic response of an anhydride cured epoxy. Therefore here the approach is applied to thermoset epoxies, also filled with soft- or hard particles and a complete spectrum of relaxation modes is used. The paper is organized as follows. First the matrix material is fully characterized. Next, compression tests are performed and experimental results are compared with the macroscopic response from the numerical simulations. Finally the local 3-D response on the inter-particle level is investigated and discussed. The part of the work presented in this paper extends on previous work on soft- and hard-particle filled polycarbonate [25], and prepares for the modeling and testing of soft- and hard-particle filled thermoset epoxies as coatings in sliding friction experiments, analogous to van Breemen et al. [29].

2. Experimental

2.1. Materials and sample preparation

The epoxy resin used is Epon 828 (Hexion Inc.), a standard diglycidyl ether of bisphenol-A (DGEBA) with an epoxide equivalent weight (EEW) of 185–192 g/eq. The curing agent is Jeffamine D230, a polypropyleneoxide diamine with an average molecular weight of 230 g/mol and an amine hydrogen equivalent weight (AHEW) of 60 g/eq, supplied by Huntsman Performance Products. The soft-particles are polysiloxane core–shell rubbers (SR) particles, supplied by Evonik Hanse GmbH as a masterbatch of particles pre-dispersed at 40 wt% in a standard DGEBA resin (trade name Albidur EP2240-A). The particle size is specified as 0.1–3 μm. The thin shell consists of epoxy-functional molecules grafted on the elastomer core. The density of resin and elastomer fillers is assumed to be equal, which entails that weight- and volume fraction have the same value. Samples are prepared with a filler fraction of 20 vol% elastomer content by diluting the masterbatch with standard DGEBA resin. The EEW of the mixture is calculated and the hardener is added in a stoichiometric amount.

The hard-particle fillers are Ti-Pure R-706, a dry grade titanium dioxide (TiO_2) with an average particle size of 350 nm and a density ρ of 4.0 g/cm^3, supplied by DuPont Titanium Technologies. First, the resin and hardener are mixed in a stoichiometric ratio. Subsequently, the TiO_2 particles are added in a ratio of 80/20 vol % epoxy/TiO_2. To calculate this ratio, a density of 1.16 g/cm^3 is assumed for the cured epoxy. The pre-mixed samples are thoroughly stirred, degassed under vacuum, and poured into aluminum cups with a diameter of 45 mm and 30 mm in height. The samples are cured for two hours at 80 °C followed by three hours at 125 °C. While preparing each batch of particle-filled samples, a control batch of unfilled epoxy was prepared following the same preparation protocol. After the curing protocol, the oven is switched off and allowed to cool to room temperature. For the compression tests cylindrical samples (∅6 mm × 6 mm) are machined from the cured samples.

2.2. Testing

To determine the glass transition temperature T_g, differential scanning calorimetry (DSC) experiments are performed using a Mettler Toledo DSC823e. First, samples of approximately 15 mg are heated to 150 °C, the subsequent scan is performed at a cooling rate of 10 K/min to 25 °C. Uniaxial compression tests are performed on a Zwick 1475 tester, equipped with a thermostatically controlled environmental chamber. The cylindrical shaped samples are compressed between two parallel flat steel plates at constant true strain rates of 10^{-4}–10^{-2} s^{-1}, at a constant temperature of 21 °C. To prevent bulging due to friction between plates and samples, a thin PTFE film (3M 5480 skived plastic film tape) is added on both ends of the sample and a lubricant (Griffon PTFE spray TF 089) is applied on both contact areas between plates and samples.

3. Modeling

3.1. Constitutive modeling

For the matrix material the EGP-model is used [27] in its multimode form [28]. It is based on an additive decomposition of the Cauchy stress into a hardening stress $\boldsymbol{\sigma}_r$ and a driving stress $\boldsymbol{\sigma}_s$:

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_r + \boldsymbol{\sigma}_s. \tag{1}$$

Here $\boldsymbol{\sigma}_r$ accounts for the stress contribution of the network, either entangled or crosslinked, and is modeled with a neo-Hookean spring with modulus G_r. $\boldsymbol{\sigma}_s$ is attributed to intermolecular interactions. It is additively decomposed into a hydrostatic (volumetric) part ($\boldsymbol{\sigma}_s^h$) and a deviatoric part ($\boldsymbol{\sigma}_s^d$) that is modeled as a combination of n parallel linked Maxwell elements:

$$\boldsymbol{\sigma}_s = \boldsymbol{\sigma}_s^h + \sum_{i=1}^{n} \boldsymbol{\sigma}_{s,i}^d = \kappa(J-1)\boldsymbol{I} + \sum_{i=1}^{n} G_i \tilde{\boldsymbol{B}}_{e,i}^d, \tag{2}$$

with bulk modulus κ, volume change ratio J, unity tensor $\boldsymbol{I}$, shear modulus G, and the elastic part of the isochoric left Cauchy–Green strain tensor $\tilde{\boldsymbol{B}}_e^d$. The specific modes are denoted by subscript $i = [1, 2, \ldots, n]$.

The plastic deformation rate tensors $\boldsymbol{D}_{p,i}$ relate to the deviatoric driving stresses $\boldsymbol{\sigma}_{s,i}^d$ by a non-Newtonian flow rule, that for the isothermal case reads

$$\boldsymbol{D}_{p,i} = \frac{\boldsymbol{\sigma}_{s,i}^d}{2\eta_i(\bar{\tau}, p, S_a)}. \tag{3}$$

Here, the viscosities η_i depend on the total equivalent stress $\bar{\tau}$, the hydrostatic pressure p and the thermodynamic state of the material S_a. The viscosities are described by an Eyring flow rule, which has been extended to take pressure dependence and intrinsic strain softening into account:

$$\eta_i = \eta_{0,i,ref} \underbrace{\frac{\bar{\tau}/\tau_0}{\sinh(\bar{\tau}/\tau_0)}}_{\text{(I)}} \underbrace{\exp\left(\frac{\mu p}{\tau_0}\right)}_{\text{(II)}} \underbrace{\left(\frac{p_p}{p_p}\right) \exp\left(S_a R(\bar{\gamma}_p)\right)}_{\text{(III)}}, \tag{4}$$

where the initial viscosities $\eta_{0,i,ref}$ define the so-called reference (un-aged) state. Part I represents the stress dependence on the deformation kinetics with the characteristic stress τ_0; the pressure dependence, part II, is governed by the parameter μ, and part III captures the dependency of the viscosities on the thermodynamic history via S_a. Strain softening is described by the softening function $R(\bar{\gamma}_p)$, a modified Carreau-Yasuda relation with fitting parameters

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