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Mechanical behavior of the alumina-filled silicone rubber under pure shear at finite strain

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ABSTRACT

The mechanical behavior of filler-reinforced rubber was investigated under pure shear at large deformation. Nanocomposites were composed of silicone rubber with different volume fractions of aluminum oxide nanoparticles, ranging from 0% to 5.0%. Thin sheet specimens were manufactured and submitted to monotonic tensile loading. The Digital Image Correlation method was employed to obtain displacement fields and consequently the stress–stretch responses were achieved. The experimentally measured stress–stretch curves were fitted to Yeoh and Lopez-Pamies models in order to estimate material parameters of the neat silicone rubber. Using this information, Mullins–Tobin and Bergstrom–Boyce approaches were employed to evaluate the strain amplification factor of the nanocomposites with different volume fractions of nanoparticles. The results indicate that a significant increase in the stiffness of filled rubber is obtained by small concentrations of nanoparticles. The effect of nanoparticles on mechanical behavior of reinforced rubber can be described using Yeoh and Lopez-Pamies models with Mullins–Tobin and Bergstrom–Boyce approaches. However, the strain amplification factor expression proposed by Bergstrom–Boyce was not suitable to describe the obtained results. In the case, the Guth model was more indicated.

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1. Introduction

Polymer nanocomposites may be defined as the combination of a polymer matrix and nanoadditives such as nanotubes and spherical nanoparticles. The possibility of improving physical properties of polymers has generated wide interest in both academia and industry. Polymer nanocomposites containing metallic and ceramic additives are extensively used in a wide range of applications. For instance, a good review of nanocomposites in electrical and thermal applications can be found in literature (Hussain et al., 2006). Mechanical properties of polymer nanocomposites reinforced with different additives have been widely investigated (Fu et al., 2008; Tjong, 2013). Among the polymer nanocomposites, silicone rubber matrix filled with nanoparticles has attracted much attention in the last decade. Several types of additives are often used for compounding elastomers in order to improve physical properties. Reinforcing fillers such as aluminum oxide are incorporated in the silicone rubber to enhance and change the mechanical properties, electrical and thermal conductivities (Cheng et al., 2014; Namitha et al., 2013).

There are several constitutive models to predict the mechanical behavior of polymers containing nanofillers, but most o f them are concerned with infinitesimal deformation (Guth, 1945; Guth and Gold, 1938; Nielsen, 1970; Dorigato et al., 2013). For large deformation, some sophisticated models can be found in literature (Ogden, 1974; Hill, 1972; Lopez-Pamies et al., 2013). Numerical

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investigations have been performed in order to gain an understanding of the mechanical response of particle-reinforced composites (Guo et al., 2014; Segurado and Llorca, 2006; Bergström and Boyce, 1999). However, in general, existing models do not comprise all effects of the fillers on the overall behavior of composite materials. To overcome the difficulty of modeling the complex behavior of reinforced polymers under large deformations, the concept of the strain amplification factor can be employed. This concept was first introduced by Mullins and Tobin (1965). Govindjee and Simo (1991) proposed an alternative approach based on the amplified deformation gradient. Knowing some ambiguities in the Mullins–Tobin and Govindjee-Simo approaches, Bergström and Boyce (1999) presented a different way of describing the behavior of particle filled elastomers, considering an average scalar measure of the overall strain.

It is important to emphasize that most of the works based on amplified approaches consider test samples with neo-Hookean matrix under uniaxial tensile loads. In fact, relatively few papers are attempted to complex loading conditions and to the use of the notion of amplification factor in different hyperelastic models. In this way, the purpose of this paper is to investigate the mechanical behavior of particle-reinforced silicone rubber under pure shear at large deformations. Planar tension test (or "pure shear" test) was performed to attain the state of pure shear. More information on planar tension test is addressed in Treloar (1944), Moreira and Nunes (2013), BS 903-5 (2004). Numerous examples of shear test in rubber can be found in the literature (Brown, 2006). Aluminum oxide (Al2O3) nanoparticles were used as fillers in different volume ratios and silicone rubber was used as matrix. Samples of the nanocomposites were submitted to monotonic tensile load. The Digital Image Correlation method (Sutton et al., 2009), which provided full-field displacements of specimens, was employed to obtain the stress–stretch response. The experimentally measured stress–stretch curves were fitted to Yeoh (1993) and Lopez-Pamies (2010) models in order to estimate material parameters of the silicone rubber. Using this information, Mullins–Tobin and Bergstrom–Boyce approaches were employed to evaluate the strain amplification factor of the nanocomposites with different volume fractions of nanoparticles.

2. Material and methods

2.1. Specimen fabrication

Specimens used in this work were composed by a polymeric matrix containing different concentrations of alumina nanoparticles. The RTV-2 silicone rubber, model RT 402 M from Moldflex (São Paulo, Brazil), was used as continuous phase. The nanoparticles were spherical aluminum oxide (α-Al_2O_3) with mean diameter of 150 nm and 99,97% purity provided by Nanostructured & Amorphous Materials Inc. (Houston, USA). These nanoparticles are considered passive (inert) fillers, i.e. their size and composition remain unchanged. Nanocomposites with different volume fractions of filler particles, ranging from 0 to 5.0%, were manufactured. Nominal volume fractions (ϕ_n) that were used in the fabrication, together with corresponding weight fractions (ϕ_w), nanocomposite density values (ρ_{nc}), and real volume fractions (ϕ) are presented in Table 1. The value of silicone rubber density used for real volume fraction calculations was the experimentally obtained value of 1294 kg/m^3, while the alumina nanoparticles density was 3700 kg/m^3 provided by the manufacturer. The densities of the samples were determined using Archimedes principle. For the quantification of mass and apparent mass of the samples, a precision balance (Shimadzu AW220) was used along with the Specific Gravity Measurement Kit. Distilled water was employed as fluid for apparent mass measurements. 9 pieces of each batch were weighed into and outside water, and an average value for the nanocomposite density was obtained. The calculations were based on rule of mixtures.

To manufacture the samples, first the alumina nanoparticles were dried at 120 °C for 24 h before being added to the liquid silicone rubber. A planetary ball mill (zirconium oxide) was employed to homogenize the mixtures during 1 h at 200 RPM. After that, the catalyst (hardener) was added and the blend was manually homogenized and poured into rectangular molds. The samples were cured at room temperature for 24 h and tested after three days. Three thin sheets for each volume fraction with dimensions of $150 \times 70 \times 3.4$ mm^3 were obtained.

2.2. Experimental set-up

Fig. 1(a) shows the experimental set-up of the planar tension test (BS 903-5). This test, which is also known as pure shear test, was carried out to obtain the stress–stretch relations. It is a good approximation to pure shear obtained with no special apparatus (Moreira and Nunes, 2013). Specimens with dimensions of $150 \times 70 \times 3.4$ mm^3 were clamped to the machine, having effective area of 150×10 mm^2. It is suggested that the width of the effective area is at least 10 times greater than the length in the stretching direction (Brown, 2006). As a result, the specimen must remain perfectly constrained in the lateral direction, while specimen thinning occurs only in the thickness direction. Fig. 1(b) presents a schematic of the clamped specimen. The pure shear occurs only in the central part of the sheet. A small region of $L_0 \times L_0$ at the central part of a rectangular sheet of material, which is stretched along a parallel pair of clamped edges, is considered to elucidate on pure shear state. In this case, only shearing motion is induced in the material element and there is no rigid body rotation. The principal stretch λ, in the stretched direction, is equal to ratio of the final length and initial length, i.e., L/L_0. As mentioned above, the horizontal length of the region of interest remains constant.

Table 1
Nanoparticle concentration.

ϕ_n (%)	ϕ_w (%)	ρ_{nc} (kg/m^3)	ϕ (%)
0	0	1294	0
1.5	4.2	1341	1.9
2.5	6.8	1354	2.5
3.5	9.4	1383	3.7
5.0	13.1	1408	4.7

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