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Material Properties

Enhancement of a magnetorheological PDMS elastomer with carbonyl iron particles

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ABSTRACT

A magnetorheological elastomer based on silicone rubber with carbonyl iron micro-particles was developed. The influence of the different amount of iron particles was experimentally studied by means of XRD, SEM, FTIR, EDS, XPS, uniaxial tension and rheological and cyclic tests. Different contents of carbonyl iron particles (10–40 wt%) were used to obtain the ratio of magnetic particles/silicone rubber that could provide the best mechanical properties on the MRE material. It was found that the composite material can have an increase of about 95% in its tensile strength when adding 20% of carbonyl iron particles to the raw rubber material. SEM analysis indicates a good dispersion of the magnetic particles on the rubber matrix, and the FTIR and XPS techniques confirm, as expected, that there is no chemical interaction between the iron from the carbonyl iron particles and the silicone rubber matrix due to a proper coating of the particles with silicone oil used as coupling agent. The TGA results evidenced that the addition of coated carbonyl iron particles had an impact on the thermal stability of the MRE and on the formation of cross-linked structures. The viscoelastic behavior of the magnetorheological elastomer is described by running experimental test on a rheometer device. Furthermore, cyclic testing were performed on the material sample to characterize the Mullin's effect.

1. Introduction

Smart materials are composite that have physical or chemical properties that can be controlled by external stimuli such as magnetic or electric fields, stresses, temperature, moisture, pH, among others, in a reversible way. Magnetorheological (MR) materials are a group of smart materials whose rheological and viscoelastic properties can be varied by applying an external magnetic field. Therefore, they have a wide variety of applications in mechanical devices as tunable vibration absorbers, actuators, magneto-active valves for air flow control, isolators in vehicle seat vibration control, and sensors, to say a few [1], [2], [3], [4] and [5].

MR elastomer is the solid-state analogue of MR fluid, however the obvious advantage of MR elastomers when compared to magnetorheological fluids (MRF) is that the particles are not able to settle with time and hence have stable MR performance. Magnetorheological elastomers (MRE) contain an elastic matrix (e.g. natural rubber or silicone rubber) in which magnetic nano- or micro-particles are either dispersed or ordered [6], [7] and [8]. Silicon rubber and natural rubber are two kinds of the most common matrices reported in the literature [6], [9], [10], [11] and [12]. However, the advantage of the silicone rubber is its low viscosity, promoting the formation of chain-like structures. Furthermore, silicone is more resistant to heat, chemical and fungus, UV and ozone attacks than natural rubber [13]. Some researchers have investigated the composite magnetorheological materials adding graphene particles to enhance electric conductivity, which can be fixed by application of a magnetic field [14], [15] and [16].

MRE can be classified into two kinds: anisotropic and isotropic elastomers. MRE cured under a magnetic field drive to have a

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special chain structure of magnetic particles which are aligned themselves parallel to the magnetic field lines. The elastomeric matrix preserves the alignment of particles after the curing process. This kind of MRE, named anisotropic MREs, with aligned magnetic particles has captured the attention of many researchers. Isotropic MRE have not an alignment of their magnetic particles because in the curing process is not applied a magnetic field [2], [10] and [17]. The use of (magnetic) hard ferrite powders as fillers for elastomers is well known in application such as tapes for magnetic recorders, flexible permanent magnets, among others [18], since the mechanical performance of polymers can be significantly improved by the incorporation of these fillers.

The aim of this paper focus on studying how different contents of iron magnetic particles added to a silicone rubber RTV 3325 (SR) matrix influence its magnetorheological response. The rheological and mechanical properties of the composite material are obtained in an attempt to correlate the magnetorheological properties with the material molecular structure.

2. Experimental procedure

2.1. Materials and composite preparation

Dimethyl-silicone oil (SO), silicone rubber RTV 3325 (SR) and catalyst were used as purchased from ChemSil. The carbonyl iron particles (CIPs, ≥99.5%, grain size: 5–9 μm) were bought from Sigma-Aldrich. To prepare the elastomer, firstly the iron particles were immersed in SO, which has a viscosity of 0.25 Pa s. Then, they were mixed with the SR. All the ingredients were mixed at room temperature during ~5 min by using a stirrer bar. Then, the homogeneous mixture was placed into a mold. The curing process was performed at room temperature for about 12 h under vacuum conditions to avoid porosity in the samples. Different contents of carbonyl iron particles (10–40 wt%) were used to obtain the ratio of magnetic particles/silicone rubber that could provide the best MRE material mechanical properties. The name of the different samples is E-*X*%, where *X* means the content of iron particles.

2.2. Characterization methods

The structural characterization of the obtained samples was carried out by X-ray powder diffraction using a Panalitycal diffractometer with Cu Kα radiation (λ = 1.54 Å). To evaluate the magnetic particles dispersion and the elemental composition of the samples, the energy dispersive X-ray spectroscopy (EDS) was performed in a scanning electron microscopy (SEM) Quanta 250-FEG FEI operated at 5 kV with an opening of 86 pA. The Fourier transform infrared spectra (FTIR) to analyze functional groups was conducted on a Thermo Nicolet model 6700 in a range of 4000–525 cm^{-1}. The surface composition was investigated by X-ray photo-electron spectroscopy (XPS, K-ALPHA$^+$ Thermo Scientific) with an X-ray Al-Kα source operated at 12.0 kV and 6 mA. TGA analysis was carried out in a SDT Q600 (TA Instruments), with a precision of the thermobalance of 0.0001 mg. For this, the samples were heated to 950 °C at 10 °C/min under an argon flow of 50 mL/min. The mechanical properties of the manufactured material samples were obtained in an INSTRON 3365 universal testing machine at room temperature. The dumbbell-shaped specimens were stretched to breakage at a crosshead rate of 200 mm min^{-1} according to the specification norm ISO37-2011.

The rheological properties of MREs were tested by using an advanced commercial rheometer (Model: MCR301, Anton Paar). A parallel-plate rotor and MR device were installed in the rheometer. A MRE sample was placed, at room temperature, between the equipment base and a rotating disk and then, it was subjected to oscillatory motion in a shear mode. The parallel-plate rotor can set to oscillate with a desired parameter frequency value. The corresponding magnetic field was induced by an electromagnet, and a metal cover enhanced and guided by a magnetic circuit perpendicular to the surface of the MRE sample. By adjusting the DC power supply from 0 A to 5 A, the range of the applied magnetic flux density to the MREs sample was in the interval values of 0–1 T. A rotating moment was acting on the moving upper part of the measuring unit. The measurements of the storage modulus G′ and the loss modulus G″ have been performed on cylindrical samples of 10 mm of diameter and 1 mm of gap in the regime of dynamical oscillations under controlled strain. The frequency and the amplitude of the external stress were varied. All measurements were performed at 20 °C. The collected experimental measurements will be discussed later on.

2.3. Constitutive material model

In order to quantitatively and qualitatively describe MRE material behavior when subjected to loading and unloading cycles, the rule of mixture material model developed in Ref. [19] was used. In this material model, the total strain energy density W_t is given as [19], [20] and [21]:

$$W_t = (1-f)W_{iso}(I_1) + fW_{aniso}(I_{4i}, I_{5i}) \tag{1}$$

where $W_{iso}(I_1)$ represents the strain energy density related to the isotropic material behavior, $W_{aniso}(I_{4i},I_{5i})$ is the anisotropic strain energy density, I_{4i} and I_{5i} are deformation invariants, and f represents the particle volumetric fraction contribution to the total material energy density. In this model, the following non-Gaussian strain energy density is used to characterize the isotropic material behavior [19]:

$$W_{iso}(I_1) = \mu\left\{N\left[\beta\lambda_R + \ln\left(\frac{\beta}{\sinh\beta}\right)\right] - \ln\left(\frac{\beta}{\sinh\lambda_R}\right)\right\} \tag{2}$$

where μ represents the material shear modulus, N is the number of links, β is the inverse of the Langevin function which is given by the following expression $\beta = \mathscr{L}^{-1}(\lambda_r)$ and λ_r represents the relative chain stretch, $\lambda_r = \lambda_{chain}/\lambda_L = \coth\beta - 1/\beta$, with $\lambda_L = \sqrt{N}$, and λ_{chain} represents the chain length $\lambda_{\mathrm{chain}} = \sqrt{I_1/3}$.

The anisotropic energy density term can be equivalently written as a function of W_t and $W_{iso}(I_1)$ as:

$$W_{aniso}(I_4, I_5) = \frac{W_t - (1-f)W_{iso}(I_1)}{f} \tag{3}$$

in which $W_{aniso}(I_4,I_5)$ is isotropized to the form [17]:

$$W_{aniso}(I_4, I_5) = f\left(\frac{A_1}{3}(I_{1i}-3) + \frac{A_2}{9}(I_{2i}-3)^2 - \frac{2A_1}{3}\ln\sqrt{I_{3i}}\right) \tag{4}$$

where A_1 and A_2 are energy density fitting parameters. Substituting equation (4) into (3) gives the total equivalent strain energy density expression

$$W_T = (1-f)W_{iso}(I_1) + f\left(\frac{A_1}{3}(I_{1i}-3) + \frac{A_2}{9}(I_{2i}-3)^2 - \frac{2A_1}{3}\ln\sqrt{I_{3i}}\right) \tag{5}$$

that is used to derive the corresponding Cauchy stress-stretch virgin material constitutive equation of the developed