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Research articles

A mechanical characterisation on multiple timescales of electroconductive magnetorheological elastomers

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A B S T R A C T

Magnetorheological elastomers are a type of smart hybrid material which combines elastic properties of a soft elastomer matrix with magnetic properties of magnetic micro particles. This leads to a material with magnetically controllable mechanical properties of which the magnetorheological effect is the best known. The addition of electroconductive particles to the polymer mix adds electrical properties to the material behaviour. The resulting electrical resistance of the sample can be manipulated by external magnetic fields and mechanical loads. This results in a distinct interplay of mechanical, electrical and magnetic effects with a highly complex time behaviour. In this paper a mechanical characterisation on multiple time scales was conducted to get an insight on the short and long-term electrical and mechanical behaviour of this novel material. The results show a complex resistivity behaviour on several timescales, sensitive to magnetic fields and strain velocity. The observed material exhibits fatigue and relaxation behaviour, whereas the magnetorheological effect appears not to interfere with the piezoresistive properties.

1. Introduction

Magnetorheological fluids are the most popular magnetic smart hybrid materials which feature a broad applicability today [1,2]. Nevertheless, the fluid matrix introduces some difficulties which are not overcome completely yet, for instance long term stability, leaking and sedimentation. Magnetorheological elastomers were developed to deal with these problems by replacing the liquid matrix with an elastomeric matrix. Those materials combine elastic properties provided by the matrix with magnetic properties caused by embedded magnetic particles. The change of the Young's modulus in presence of magnetic fields is the most widely used and described feature of those materials, named the magnetorheological effect [3,4].

To this day magnetorheological elastomers are still a topic of ongoing research exploring possibilities and gaining an understanding of the complex material behaviour. Nevertheless, they feature broad applicability in the field of sensors and actuators [5–7]. Several approaches were conducted to add further functionality to magnetorheological elastomers. Recently experiments with an added electrical functionality were undertaken [8–24]. Here the occurring effects in these new materials are often referred to as piezoresistive effects, or magneto-piezoresistive effects if the electrical properties are sensitive to magnetic fields.

A view on the previous works shows a broad variety of utilised and characterised materials. This leads to investigations finding suitable materials with specific features, as well as with enhanced and stable magnetic, mechanic and electrical properties. The most common approach is the addition of conductive graphite or graphene particles to the widely used carbonyl iron particles known from magnetorheological elastomers [15,16,22–25].

Different approaches gaining electrical conductivity with those materials include the utilisation of bare magnetic particles [8,9,12–14,17–21,26–29], magnetic particles coated with silver [10,11,19,20] and the before mentioned mix of iron particles with graphite or graphene particles. Characteristics of materials prepared with different particle materials like gold, copper and nickel [30] and iron, magnetite and neodymium-iron-boron [27] were compared. Further, it was shown, that iron particles feature stronger effects than nickel particles [9]. On the other hand, the utilisation of nickel particles results in better conductivity [21], whereas the oxidation of iron tends to lead to limited resistivity [28]. Therefore, often graphite particles are added for a better conductivity when choosing iron particles. Increasing amounts of graphite reduce the measured resistance of the samples [15].

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These investigations are accompanied by mechanical experiments with materials solely loaded with conductive particles [31] supported by theoretical descriptions of the behaviour [32]. Here carbon black particles were used instead of graphite, which provides sufficient conductivity as well.

It was shown before that carbon black filled elastomers may suite as a material for sensor application [33,34], this motivates the combination of electrical features with magnetorheological properties. First sensor applications with combined electrical and magnetosensitive properties [25,35] and an application as tuneable suspension bushing [39] were presented before.

Those highly complex materials are a current field of broad research. Most works deal with the characterisation of the electro resistive properties. A direct interplay of mechanical load, magnetic field and resistance was investigated before [8,9,12,23,24]. Comprehensive mechanical characterisations of those materials contribute to a better understanding of the material properties [10,11,16]. The effect of varying temperatures as an important influence was pointed out [12,19,20]. Moreover, the high impact of the isotropy of the particle distribution was proven [16,17,20,28]. Furthermore, it was already shown, that the direction of magnetisation of those materials affects the conductive properties as well [19,20,26]. Further researches aim to a characterisation of electrical impedance [14], permittivity [27] and capacitance [29]. Publications dealing with a theoretical description of the material behaviour induce that in addition to the current through the conducting particles, tunnel currents in between particles also occur [10,15,18].

The mechanical properties of those materials are heavily influenced by the embedded particles and the interplay with magnetic and electrical properties. The available experimental results of the electrical properties of those materials are hardly comparable in general since the properties are highly depending on material composition, testing methods and conditions. A deeper understanding of the ongoing physical effects is critical to get an insight in the magneto-electrical mechanisms. Separating the complicated underlying effects and assigning them to specific material or geometrical properties is a very complicated task for the future.

In this paper a comprehensive investigation to analyse the mechanical and resistive behaviour of a carbon black loaded magnetorheological elastomer is presented. For future applications the long-term stability and the behaviour after a large number of mechanical loads is critical. Thus, this investigation focuses on the evaluation of relaxation and fatigue behaviour of the presented material. The presented results may guide to a better controllability and improved materials. Furthermore, the new insights discussed here may lead to a deeper understanding of the interplay of properties and effects connected to electroconductive magnetorheological elastomers.

2. Materials and methods

2.1. Sample preparation

All investigated samples were synthesized using silicone polymers by Alpina Technische Produkte GmbH, carbonyl iron particles by BASF SE and carbon black particles by Evonik Degussa GmbH. Carbonyl iron particles are most commonly utilised in these hybrid materials and were chosen to provide a strong magnetorheological effect. Neither the utilised iron powder on its own nor the elastomer with embedded particles show electrical conductivity, which might be linked to corrosion [28]. Thus, carbon black in form of the product Printex XE2 were added to the mix to provide electrical conductivity. The amount was kept to the minimum to achieve significant piezoresistive effects at moderate electrical resistances. Larger amounts of Printex XE2 compromise the fully elastic behaviour of the hybrid material. The choice of carbon black over graphite was motivated by the previous works showing suitable electrical properties [31–34] and good compatibility with the chosen silicone elastomer.

The mix seems to be favourable for future applications, for the reasons of providing good magnetorheological response, stronger magneto-piezoresistivity compared to nickel [9] and higher overall resistivity [15]. This approach was also chosen by other groups aiming for applications [25]. The amounts of the components utilised in the final mix are given in the following Table 1.

The carbon black particles were sifted with a mesh size of 25 μm to break up large agglomerates. With the resulting fine powder a more a homogeneous distribution of the particles in the elastomer was achieved. All components were thoroughly mixed to provide isotropic sample properties. After addition of the particles, the mixture was evacuated in a vacuum desiccator to remove air bubbles. The samples were cured at room temperature. After moulding the samples were kept untouched for 8 h to ensure a complete polymerisation. Due to the high viscosity of the mix no sedimentation occurred. This was checked via X-ray microtomography as it proved to be a convenient method in previous experiments with magnetorheological elastomers [36–38]. Here, no individual particles or particle structures could be resolved as a result of their small size. Nevertheless, no overall gradients of the grey values within the sample were observed, which verifies an overall macroscopic isotropy and homogeneity better than 15% in all directions. Due to the high contrast in the tomography data sets between the dense particle-polymer mixture and gas, bubbles can be resolved easily. The tomography results verified the absence of air or gas bubbles. As pointed out before, the homogeneity of the sample material is critical [17], thus the synthesis was optimised to achieve a homogeneous particle distribution. To receive an impression of the internal microstructure of the samples scanning electron microscopy was performed. The results are shown in Fig. 1. As visible there, despite the sifting most carbon black particles are embedded as agglomerates smaller than 25 μm. Fig. 2 shows as photography of the prepared samples. A cylindrical shape of a diameter of 9.7 mm and height of 10 mm was chosen for convenient characterisation via stress-strain measurements. Four samples were evaluated, all produced simultaneously from the same batch of previously mixed components.

Several alternatives to achieve a proper electrical contact to the samples were tested. For the final solution fine brass meshes were glued to the surfaces of the samples which were than soldered to a copper wire. The utilised silicone glue was mixed with carbon black particles which led to the best achievable electrical conductivity. A drawback of this solution is the fact that the sample is hindered deform at the top and bottom surfaces. This leads to a barrelling of the sample if severe axial strain is applied. As a result, uniaxial deformation during the mechanical measurements is only ensured for very small strains. Compared to the sample material the cured glue is very hard, thus its deformation can be neglected in the evaluation of the stress-strain data.

Table 1
Used polymer and particle composition for preparing the samples.

Elastomer	Weight proportion	wt%
Alpina ALPA-SIL Classic A	40	18.8
Alpina ALPA-SIL Classic B	8	3.8
Silicone oil (5 mPas)	40	18.8
Particles		
BASF CC carbonyl iron particles	120	56.3
Evonik Degussa Printex XE2	5	2.3