Magnetorheological Elastomers with High Variability of Their Mechanical Properties

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Abstract. The viscoelastic properties of magnetorheological elastomer (MRE) composites containing different concentrations of iron particles in a silicone matrix and the change of these properties in a magnetic field of variable strength were investigated. MRE samples with small and large iron particles as well as isotropic and anisotropic particle arrangements in the silicone matrix were compared. The results show a strong increase of the storage and loss moduli of the MRE in the magnetic field. A more pronounced influence of the anisotropic configuration was observed for the samples with small particles than for those with large particles. The relative increase factor of the storage modulus in the magnetic field was found to be an order of magnitude higher for the composites with large than with small particles. As a maximum, an enhancement of the storage modulus from less than 30 kPa at zero field to about 2500 kPa at a magnetic flux density of 700 mT, corresponding to an increase factor of 90, could be achieved under the selected experimental conditions.

1. Introduction
Magnetorheological elastomers (MRE) are a relatively new class of smart materials whose mechanical properties can be influenced in a magnetic field. The magnetorheological effect of MRE manifests itself in a considerable increase in the Young’s modulus and the shear modulus, which occurs at rising magnetic field strength. In analogy to the shear stress of MR fluids, this effect is reversible.

MRE are soft composite materials which basically consist of magnetically polarizable particles like iron in a deformable polymer matrix. In the magnetic field, the mechanical behavior of the composite is dominated by a superposition of magnetic particles interactions and elastic forces of the elastomer matrix, which causes changes in the elastic as well as in the viscous properties of the MRE.

Strong commercial interest arises from these fascinating properties. The controllable stiffness of the material can possibly be exploited in dynamic vibration absorbers whose frequency of operation may be tuned by the magnetic field strength. Further possible applications of MRE are adaptive vibration dampers, controllable mounts, switchable haptic surfaces as well as controllable seals and valves.

Several papers regarding MRE were published since the 1990s [1-6]. Up to now, most studies were performed on MRE with silicone rubber as the matrix material. Silicone has the advantage that the synthesis starts with liquid precursors, in which the particles can be suspended. Moreover, it could be demonstrated that the mechanical properties of the MRE composite may be influenced by the...
application of another magnetic field applied already during the curing of the silicone. This effect is ascribed to the formation of particle chains which are “frozen” in the solid state [5].

This paper addresses experimental investigations of the influence of various parameters of the composition of MRE on their mechanical properties and on their capability of the viscoelastic properties to be tuned in the magnetic field. It was evaluated under which conditions the strongest magnetorheological effects occur. In the investigations silicone rubber was used as matrix material, due to the possibility to prepare very soft composite materials.

2. Experimental

The silicone matrix consisted of polydimethylsiloxane prepared from the reaction of vinyl-terminated polysiloxane chains and polysiloxane with Si-H groups. The basic crosslinking density of the reference elastomer without iron particles was set thus, that an elastic storage modulus of about 50 kPa was achieved. In order to evaluate the influence of the iron particle quantity on the mechanical properties, various silicone-based composites with iron concentrations by volume between 0 and 35 % were prepared. For a part of the samples the silicone matrix was cured only by heat treatment. Another part of the samples was additionally exposed to a magnetic field during the crosslinking process in order to achieve an anisotropic arrangement of the iron particles. Furthermore, two kinds of iron particles with strongly different mean particle sizes of about 5 µm (BASF, Germany) and ca. 40 µm (Höganäs, Sweden), respectively, were used for the preparation of the MRE composites.

The samples were studied by rheological measurements in the oscillation mode in a rheometer Paar-Physica MCR 300 equipped with a magnetorheological measuring unit. For the evaluation of the influence of the magnetic field strength on the viscoelastic properties, the experiments were carried out at a fixed frequency of the oscillation of 10 Hz, an amplitude of 1 % and under the variation of the magnetic flux density.

3. Results and Discussion

3.1. MRE with small iron particles

The measurements yielded the storage modulus which describes the elastic behavior of the sample, as well as the loss modulus which is a measure of the viscous behavior. Both moduli are strongly increased when the magnetic flux density is enhanced. As expected, the base value of the storage modulus without magnetic field rises with increasing concentration of iron particles, indicating the enlarged hardness of the composites with higher iron particle concentrations.

Figure 1 shows the increase of the storage and loss moduli of MRE with various contents of small iron particles in the isotropic distribution with rising magnetic flux density. The higher the iron concentration the larger is the absolute increase of both moduli in the magnetic field, shown here in a logarithmic scale. It becomes apparent, that under all conditions the elastic properties dominate the viscous behavior of the composites.

In Figure 2 the dependence of the storage modulus of the corresponding samples with the same compositions, but cured in the presence of a magnetic field, on the magnetic flux density is revealed. The absolute increase of the storage modulus due to the magnetic field is significantly higher for the anisotropic samples (from 320 kPa at zero field to 2800 kPa at 700 mT) than for the isotropic ones (from 150 kPa at zero field to 700 kPa at 700 mT). This is even true for the relative increase factor, defined as the ratio of the storage modulus at the highest flux density of ca. 700 mT and the storage modulus at zero field, which is about 8 (see Fig. 2) for the anisotropic and less than 5 (not shown, but derivable from Fig. 1) for the isotropic samples as the maximum values. It can be concluded that the magnetic interactions between the iron particles are stronger in the anisotropic than in the isotropic arrangement.
Figure 1. Dependence of storage modulus (left) and loss modulus (right) of isotropic MRE with different concentrations of small iron particles (in vol.%) in silicone on the magnetic flux density.

Figure 2. Dependence of storage modulus (left) and its relative enhancement (right) of anisotropic MRE with different contents of small iron particles (in vol.%) in silicone on the magnetic flux density.

3.2. MRE with large iron particles
The corresponding results of the MRE samples containing larger iron particles with a mean size of about 40 µm in an isotropic arrangement are depicted in Figure 3. The absolute increase in the storage as well as the loss modulus by the magnetic field is drastically higher than for the composites with small particles. MRE containing 30 or 35 vol.% large iron particles reach storage moduli of about 3.
MPa under the chosen experimental conditions at a magnetic flux density of ca. 700 mT. Without the magnetic field, the same samples have storage moduli below 100 kPa, indicating drastic increase factors of the storage modulus due to the magnetic field. Furthermore, in the field-free state very low storage moduli of the samples with small iron contents were observed, which are even lower than the storage modulus of the pure silicone sample without iron. Apparently, the crosslinking of the silicone is affected by the presence of the large iron particles, which results in a reduced elasticity of the composites.

Figure 4 shows scanning electron microscopic photographs of isotropic and anisotropic composites. In contrast to the isotropic material, the formation of fragments of iron particles chains, caused by the application of a magnetic field during the curing process, can be observed in the anisotropic sample.

**Figure 3.** Dependence of storage modulus (left) and loss modulus (right) of isotropic MRE with different concentrations of large iron particles (in vol.%) in silicone on the magnetic flux density

**Figure 4.** Scanning electron microscopic (SEM) photographs of MRE samples with isotropic (left) and anisotropic arrangement (right, magnetic field direction indicated by the arrow) of large iron particles
Finally, Figure 5 shows the dependence of the storage modulus and its relative increase of the anisotropic composites with large iron particles on the magnetic flux density. In contrast to the samples with small particles, the difference between the maximum storage moduli of isotropic and anisotropic composites is not very pronounced, indicating magnetic interactions of comparable strengths between the iron particles in both configurations. The storage modulus of the isotropic MR elastomer with 30 vol.% iron particles increases from less than 30 kPa at zero field to about 2500 kPa at 700 mT (see Fig. 3). In comparison, for the corresponding anisotropic sample, the storage modulus rises from ca. 50 kPa at zero field to 3000 kPa at 700 mT (see Fig. 4).

The relative increase factor of the anisotropic composite in Figure 5 reaches a maximum of ca. 65. In case of the corresponding isotropic composite, the increase factor reaches even a maximum of about 90 (not shown, but derivable from Fig. 3), due to the very low starting levels of the storage modulus at zero-field. Thus, the maximum increase factors of the MRE composites with large iron particles surmount those of the samples with small particles by an order of magnitude. To the knowledge of the authors, such large increase factors of MRE have not been reported before.

The observed difference between the properties of the isotropic composites with small and large iron particles is referred to the capability of the large particles in the MR elastomer to generate stronger magnetic interactions than the small particles. However, anisotropic MR elastomers with small particles can also generate large storage moduli of nearly 3000 kPa in the magnetic field, but with significantly higher base modulus at zero field than for large particles (see Fig. 2). This behaviour is explained by the chain structure of the iron particles. Due to this ordering, the particles have very low distances from their next neighbours. These low distances enable the small particles to generate strong interparticle interactions in the magnetic field. Furthermore, the larger increase factors of the MRE with large particles may also be caused by the softening influence of the large particles which reduce the hardness of the silicone elastomer.

Another result of the investigations on the MRE with large particles is that the composites with 30 vol.% iron particles yield the highest relative increase of the storage modulus (see Figure 5). This is in agreement with former research, in which an optimum iron concentration in the vicinity of 30 vol.% was reported [4].

**Figure 5.** Dependence of storage modulus (left) and its relative enhancement (right) of anisotropic MRE with different contents of large iron particles (in vol.%) in silicone on the magnetic flux density
4. Conclusions

The investigations on various MRE composites containing different concentrations of iron particles in silicone yielded a strong variability of the viscoelastic properties and their change in a magnetic field, caused by the modification of the composition of the MR elastomer in terms of iron particle size and concentration. The enhancement of the storage modulus and the loss modulus is more pronounced for the samples with larger amounts of iron particles than for those with small amounts. Furthermore, composites with large iron particles yielded relative increases of the storage modulus, which are an order of magnitude above those of the corresponding composites containing small iron particles. A maximum factor of increase of the storage modulus of about 90 in the magnetic field could be achieved under the selected experimental conditions, where the absolute storage modulus rises from less than 30 kPa at zero field to about 2500 kPa at 700 mT. These results reveal promising perspectives of this new class of smart materials for various applications like adaptive vibration dampers, switchable haptic surfaces and controllable seals and valves.

References

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