

Evaluation of Highly Compliant Magneto-Active Elastomers with Colossal Magnetorheological Response

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ABSTRACT: Highly compliant elastomers with a shear storage modulus as low as 25 Pa are prepared using commercially available silicone, plasticizer, and tactile mutator silicone additive. They are used as matrix material for magneto-active elastomers (MAEs) with carbonyl iron contents between 0 and 85 wt %. In the absence of an external magnetic field, the storage modulus of MAEs based on two selected mixtures ranges between ~ 100 Pa and ~ 2000 Pa. Addition of a mutator to the matrix mixture results in a long post-cure period depending on the curing temperature and the initial mixture. In the presence of a magnetic field, the presented MAEs exhibit a strong magneto-induced change in storage modulus resulting in a colossal magnetorheological effect of $>10^6$ % which is ~ 30 times higher than previously reported values. The results are of interest in applications using such elastomers as cell substrates with magnetically tunable rigidity. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 39793.

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INTRODUCTION

Many biological tissues and organs exhibit very soft viscoelastic properties with elastic moduli as low as only a few Pa, for example, fat, protein, etc. Other tissues with elastic modulus below 1 kPa include liver, brain and lymph nodes. An overview of the elastic moduli of different tissues is given in a review.¹ Note that several measurement methods have been employed to determine the elastic moduli so that a degree of variability between values given in the literature may be observed. Experiments with different types of cell have shown that the rigidity of the substrate influences cell behavior such as migration, morphology, and cytoskeletal growth. An overview of mechanosensitive cells is similarly provided.¹ Cells which are grown on substrates with properties similar to their original tissue show a behavior comparable to what they exhibit *in-vivo*. Although the complete cell response cannot be determined by the substrate stiffness alone, an easily handled elastomeric cell substrate may assist in obtaining knowledge about the mechanisms of cell evolution useful in tissue engineering.^{2–4} Recently, the use of soft, biocompatible, magneto-active elastomers (MAE) has been proposed. The viscoelastic properties of such MAE can be controlled by the application of an external magnetic field, particularly where the field is specifically tailored toward moving, separating, controlling, and influencing biological cells on the surface of such MAE materials.⁵

In this article, potential cell substrates in the form of extremely soft elastomers are presented. Using such soft elastomer as a matrix material for MAE provides an advantage that the stiffness of the substrate during cell growth can easily be controlled by an externally applied magnetic field. In general, MAE comprises magnetically polarizable particles dispersed in a nonmagnetic elastomeric matrix.⁶ $G = G' + iG''$ is the complex shear modulus of the elastomer, where G' is the shear storage modulus and iG'' is the imaginary shear loss modulus. The shear storage modulus is split into the initial storage modulus without field G_0' and the magnetically induced part $\Delta G'$: $G' = G_0' + \Delta G'$. For a typical elastomer with an initial shear modulus $|G(0)|$ of 400 kPa, the effect of a magnetic field on the shear modulus $|\Delta G|$ was theoretically predicted to be 50% at the optimum particle volume fraction of 27 vol %.⁷ This increase in G was exceeded in several experimental works, often simultaneously with a lower shear modulus.

The race for achieving higher magnetorheological (MR) effects $\Delta G'/G_0'$ is driven by applications of MAEs in actuators,⁸ shock and vibration absorbers.^{9–12} Investigations using thick MAE samples have shown an effect of up to 68% with an initial modulus of 300 kPa.^{13,14} A systematic analysis of isotropic and anisotropic magnetic elastomers magnetized in different directions with respect to the externally applied magnetic field resulted in a maximum effect of 75% where the storage modulus G_0' ranged between 20 and 55 kPa.¹⁵ An effect of 300% with an

initial modulus of only 4 kPa was found in a study concerning the harmonic loading of MR elastomers.¹⁶ A new MR material based on a polyurethane matrix with an initial shear modulus of around 1 MPa exhibited effects of 121%,¹⁷ to 387%,¹⁸ and 708%.¹⁹ An even higher effect of 878% was reported in an earlier publication.¹⁰ A giant MR response of 41,900% for an isotropic filled silicone rubber based elastomer corresponds to a low modulus of 1000 Pa.²⁰ It has been postulated that an ideal MR material should possess both a high relative MR effect and a high magneto-induced storage modulus.¹⁹ In this publication it was argued that a high relative MR effect is usually due to a low storage modulus without field G'_0 but the magnetically induced change $\Delta G'$ is small. In this article, we present MAE materials with an extremely large MR effect, which is to our knowledge much higher than all values reported elsewhere. This is indeed achieved with low initial modulus $G'_0 \sim 10^2$ Pa, however with a magneto-induced modulus of the same order of magnitude ($\sim 10^6$ Pa) as reported in [19].

EXPERIMENTAL

Materials

Elastosil® RT 604 and the plasticizer Wacker® Siliconoil AK10 were purchased from Wacker Chemie AG, Munich, Germany. Additional samples were prepared using Sylgard® 184 as the main component. It was bought from Dow Corning, Midland, MI. Catalyst 510 was purchased from Hanse Chemie, Geestacht, Germany. The additive Slacker® – Silicone Tactile Mutator was obtained from Smooth-On, Easton, PA. The used carbonyl iron powder (CIP) comprised particles with a mean diameter of 4.5 μm (type SQ) purchased from BASF AG, Ludwigshafen, Germany.

Preparation

Soft Elastomeric Matrix. To fabricate an ultra-soft silicone elastomer, two main approaches are found in the literature. The first approach is to vary the ratio between the two components in a two-component silicone putty.^{6,15} In general, this leads to polymer networks with a low cross-linking density. The resulting elastomers are extremely adhesive and tend towards a jelly like appearance. The alternative approach is to soften the standard polymer by adding a plasticizer to the silicone mixture.^{20,21} The latter approach was followed in this article.

In accordance with the manufacturers guide lines, Elastosil® RT 604 was used in a fixed ratio of nine parts of component A to one part B. This results in an elastomer with a shore A hardness of 25 ($G' = 40$ kPa, $G'' = 500$ Pa).²² To soften this elastomer, silicone oil was added as plasticizer to the mixture. This procedure works only to a certain extent, because an excessive amount of silicone oil (see specification below) results in the polymer becoming slushy. This can be avoided by adding the so-called Slacker® (silicone tactile mutator), changing the properties of platinum-cured silicones and creating a more “flesh-like” material.²³ Unfortunately, too much Slacker® (more than twice the amount of Elastosil®) results again in an increase in adhesion. Furthermore, an additional catalyst must be added to the elastomer in order to shorten its curing time. This is required for the subsequent fabrication of MAE to prevent the sedimentation of the CIP before curing is completed.

Finally, the recipe for the ultra-soft elastomer matrix comprises five components: two-component silicone, plasticizer, Slacker®, and catalyst, the ratios of which can be varied. After performing a number of experiments two basic mixtures (denoted A and B in the following) were arrived at. Mixture A consists of 1 g Elastosil® (0.9 g Elastosil® part A and 0.1 g Elastosil® part B) combined with 2 g Slacker®, 30 g plasticizer and 16 mg catalyst. Mixture B differs in the amount of plasticizer, only 18 g of silicone oil are used for this mixture.

Mixture A yields elastomer with very low storage modulus $G' = 700 \pm 140$ Pa. Mixture B is somewhat harder ($G' = 2000 \pm 300$ Pa), but it is more convenient to handle due to stability during transport between the petri dish and rheometer. Softer samples tend to change their shape or fold themselves in an irreversible manner. Similar results were also obtained using Sylgard® 184 elastomer instead of Elastosil® RT 604.²⁴ Sylgard® 184 is commonly used in biomedical applications.²⁵ Pure Sylgard® 184 mixed in a ratio of ten parts base with one part crosslinking agent has a shore A hardness of 44,²⁴ and is less brittle than Elastosil® RT 604. Usually, soft samples of this elastomer are prepared by changing the ratio between the base and crosslinking agents. Ratios of up to 100–1 are described in the literature instead of the 10–1 ratio advised by the manufacturer.²⁶ This huge decrease in crosslinking agent leads to an extremely adhesive, jelly like material.

Magneto-Active Elastomers

All components were put together and mixed with an electric mixer (Roti-Speed). The resulting fluid was deposited in petri dishes and then degassed to remove air bubbles. Unfilled samples and isotropic MAE were cured at room temperature (RT) or on a heat plate (HP) at 100°C from 20 min up to several hours. Anisotropic MAEs with magnetically aligned particle chains^{15,27} were prepared by curing on the HP in an externally applied magnetic field (electromagnet EM2 from MAGMESS Magnet-Messtechnik, Bochum, Germany). The density of the applied magnetic flux was $B = 75$ mT. Anisotropic MAEs at RT were not fabricated due to the long curing time (~ 20 days).

Measurements

Rheological measurements were performed using an Anton Paar (MCR 301) rheometer with a MR cell using parallel plate geometry with a plate diameter of 20 mm (MRD 180). This system is often used for MR measurements.^{16,19,28–30} The samples had a diameter of 20 mm, either dictated by the mold size or obtained by die cutting. Sample thicknesses were 2 ± 0.5 mm. Long-time behavior was investigated by performing amplitude sweeps at fixed frequency ω of 10 rad/s and fixed normal force $F_N = 0.1$ N. Under these conditions, the shear moduli G' and G'' given in this paper refer to the strain $\gamma = 0.1\%$, in the vicinity of which they are practically independent of γ . Magnetosweeps were performed at a fixed angular frequency $\omega = 10$ rad/s and strain $\gamma = 0.1\%$. The normal force F_N varied from 0.1 up to ~ 10 N. The air gap d of the MR cell was selected in such a way that, in the absence of a magnetic field, the normal force $F_N = 0.1$ N. Depending on the thickness of the sample and its iron content it was possible to reach a magnetic flux density of 0.8 T. In anisotropic MAEs the magnetic field was applied in

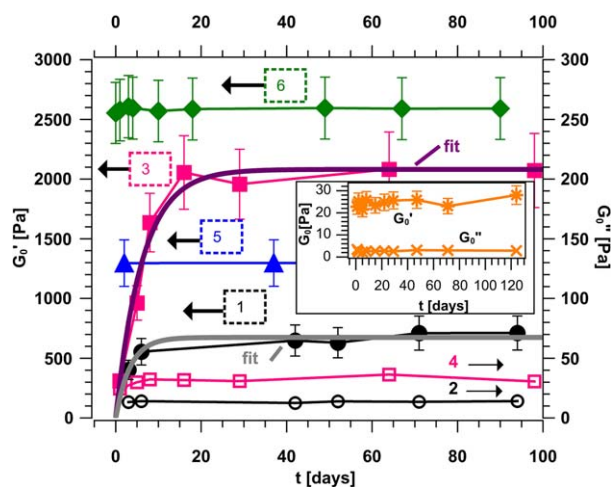


Figure 1. Storage modulus G'_0 (1, 3, 5, and 6) and loss modulus G''_0 (2 and 4) of unfilled elastomers measured over a period of time. Curves 1, 2 (RT curing) and 5 (HP curing) belong to the samples made from matrix A, curves 3, 4 (RT curing), and 6 (HP curing) belong to matrix B. For the storage moduli 1 (matrix A) and 3 (matrix B), exponential fits describing the time dependent curing behavior are shown. The inset displays the storage and loss modulus of the softest prepared elastomer. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the direction of the particle chains. The measuring point duration was fixed to 25 s.

RESULTS AND DISCUSSION

A Route to Soft Elastomers

Elastosil® Matrix. By adding silicone oil as a plasticizer to the elastomer mixture, it can be easily softened. Figure 1 shows the influence of the amount of silicone oil on the storage modulus G' over a period of up to 100 days. By a slight adjustment to the mixture A—less Slacker® (0.63 g) whilst adding more catalyst (0.16 g)—it was even possible to store 80 g plasticizer in 1 g Elastosil® and receive shape-stable samples with a storage modulus G'_0 as low as 25 Pa, as shown in the inset of Figure 1. Further increasing the amount of plasticizer led to samples, which did not keep their desired shape. The loss modulus G''_0 of the softest sample presented here is only 3 Pa. For other elastomers and MAEs, which will be presented in the following, the loss modulus reaches a maximum value of a fifth of the value of the storage modulus for softer samples, but mostly it is less than one tenth of the corresponding G' . Because of this fact, G'' is omitted from the following figures ($|G| = G'$). The same numbers in Figure 1 and in the following Figures refer to the same matrix mixture.

After 10–60 min at RT the sample is stable in a sense that it is possible to cut and remove it from the dish for measurements (primary curing). The primary curing time is crucial for MAE preparation. It should be neither too short (because air bubbles cannot be so quickly removed from the elastomer) nor too long (because sedimentation of iron particles may occur). The optimum primary curing time should be set to ~10 min. However, the “ripening” of the sample takes longer: the storage modulus of the sample grows with time and reaches its final value after

~10–20 days at RT. With the increasing oil content, this post-curing time decreases. It was observed that the postcuring time can be reduced to less than a day if the samples are cured on a HP but the resulting storage modulus is also increased.

The time dependence of the storage modulus (“ripening”) can be described by the following formula:

$$G'(t) = G'(\infty) \times (1 - e^{-t/\tau}). \quad (1)$$

For the matrix A with RT curing, we obtained $G'(\infty) = 675$ Pa, $\tau = 3.37$ days, for matrix B the values were $G'(\infty) = 2081$ Pa and $\tau = 6.22$ days, this fit is shown in Figure 1. Curing at 100°C resulted in the storage moduli $G'(\infty) = 1297$ Pa for matrix A and $G'(\infty) = 2591$ Pa for matrix B. τ was significantly reduced for HP curing. At 100°C, $\tau = 0.28$ days and 0.19 days were obtained for matrixes A and B, respectively. Increasing of the HP temperature above 100°C was not reasonable due to the temperature stability of the plastic MAE carriers.

The storage modulus also strongly depends on the amount of catalyst. It was possible to reduce the storage modulus down to a few Pa and obtain an elastomer which is stable over a long period of time but it was not possible to perform more than a few measurements on such a sample due to its extremely soft character.

The influence of the amount of Slacker® on the storage modulus was also investigated. With the increasing content of tactile silicone mutator the storage modulus also increases. If the amount of Slacker® by weight is more than two times that of the Elastosil®, the mixture results in an extremely adhesive and stretchable material with jelly like behavior. If samples are prepared without Slacker®, the huge amount of silicone oil leads to an “oil-bleeding” surface. To avoid this, it was possible to omit the additional catalyst, which is a feasible route to prepare unfilled elastomers but it is not suitable for the preparation of MAE’s because of iron particle sedimentation.

Sylgard Matrix

Similar to the Elastosil®-based elastomers, samples using Sylgard® 184 with the same additives were prepared and good quality soft elastomers obtained.

Highly Compliant MAE’s

In the following, only the results for Elastosil®-based samples are presented, although similar results can be obtained with Sylgard®-based elastomers. If two materials with different shear moduli are combined, it is expected that the elastic modulus of the resulting composite materials lies between the smaller and the larger moduli of two constitutive components. Therefore, it is expected that with increasing iron content the shear storage modulus of the elastomer increases.^{28,31} Theoretical articles corroborated such an augury,^{7,32,33} and models have been developed to confirm the experimental results.^{34,35}

Figure 2 shows the dependence of the storage modulus G'_0 , in the absence of a magnetic field, on the concentration of CIP. Unexpectedly, it may be clearly seen that G'_0 has a minimum at a CIP content of between 40 and 65% by weight for elastomer matrix A and at ~60% for elastomer matrix B. This behavior was observed both in isotropic and anisotropic samples.

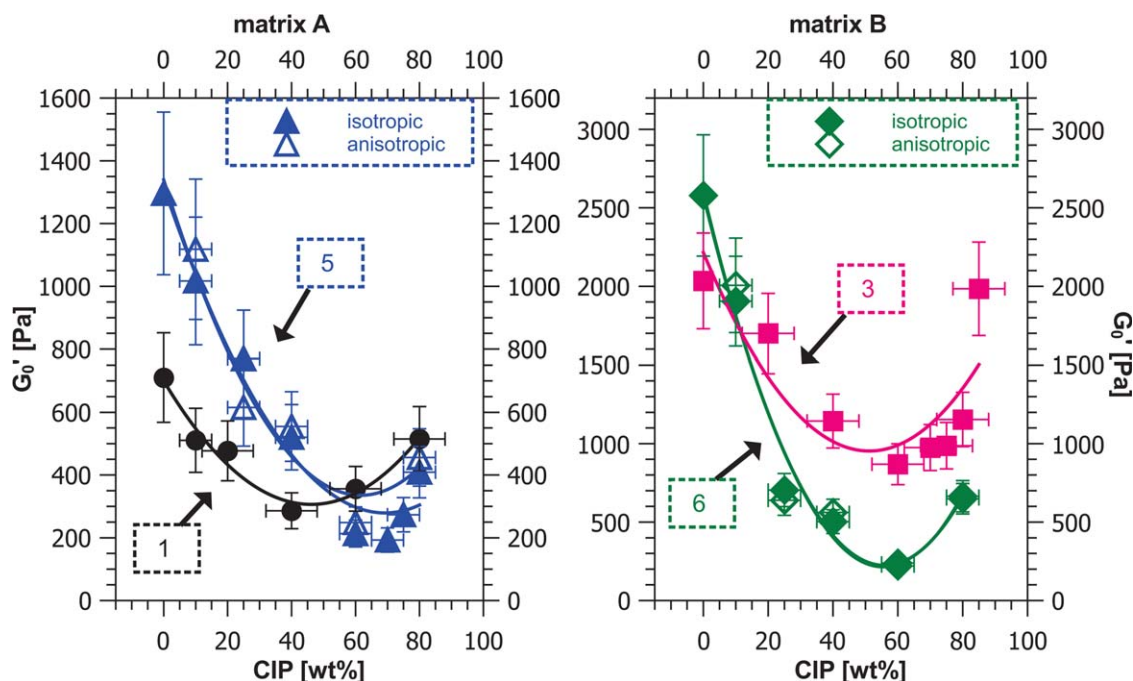


Figure 2. Storage modulus G'_0 of MRE after 20 days dependent on the CIP content for matrix A (1) and B (3) cured at RT and on HP at 100°C (5 and 6, respectively). For the HP curing both isotropic and anisotropic samples are shown. The continuous lines are second order polynomial fits and intended simply as a guide to the eye. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Obviously, since G'_0 of filled composites falls below G' of the unfilled elastomeric matrix, the usual theoretical low boundaries for G' are not applicable to the presented composites (e.g., the Reuss bound or the Hashin-Shtrikman relation).^{32,36} The inserted lines in Figure 2 represent second order polynomial fits (which are only intended as a guide to the eye). From these results, it can be concluded that the iron particles used inhibit the curing,³⁷ what had a positive effect in our case (reduction of G'_0 leads to the enhancement of relative MR effect because this effect is inversely proportional to G'_0). We are aware of only one publication reporting a decrease of G'_0 with the growing concentration of CIP particles in hard MAE samples (pure Sylgard® 184) with G'_0 ranging between 400 and 800 kPa.³⁷

An attempt was made to eliminate this inhibition of curing. New samples with particles washed in ethanol were prepared. Samples with cleaned particles showed a slightly higher storage modulus G'_0 than samples with particles used without further treatment, but the qualitative dependence on the concentration of CIP remained the same. For example, G'_0 of the isotropic samples with elastomer matrix B and 50% of CIP by weight was, after 30 days, ~5% larger with ethanol-washed particles than with as-delivered particles. However, under the influence of an applied magnetic field of 500 mT, G' of a sample with ethanol-washed particles was 0.1 % lower than with as-delivered particles. The washing of particles with ethanol did not significantly change the viscoelastic properties of samples with or without magnetic field. Possibly, a more intense washing with different solvents could prevent the inhibition of curing³⁸ and clarify the chemical mechanism of it. After the post-curing is completed, the MR effect appears to be mainly affected by the

amount of iron and not by the initial value of the storage modulus in the absence of a magnetic field.

In the literature, publications comparing the elastic properties of unfilled elastomers with elastomers filled with different amounts of iron powder are rare. For polyurethane MRE, the storage modulus increased from roughly 300 kPa without filling to 1230 kPa at 80 wt % Fe.¹⁶ Comparison of different Fe contents showed an increase in storage modulus G'_0 with increasing iron content but the modulus measured in these publications (5 kPa to 55 kPa for elastomers filled with 10, 20, and 30 wt % of iron particles) was higher than the storage modulus of our samples.^{15,39} Moreover, the behavior of the MAEs depends strongly on the composition of the elastomeric matrix and on the type of particles used.^{28,40}

MR Effect

Magnetosweeps were performed on samples of both series. Figure 3 gives an example of the experimental results for the dependency of the storage modulus G' on the magnetic flux density B . The MAE sample was demagnetized before the start of the magnetosweep measurement. A hysteresis in the behavior $G'(B)$ is observed. Two loops are shown in order to prove the reversibility of the MR effect. In literature, the magnetosweep usually shows one curve for G' in dependence on increasing B . All numerical values from magnetosweeps, which will be presented in the following, refer to the first-loop measurement with increasing field (initial magnetization curve). The inset of Figure 3 depicts the magnetosweep measurement of the unfilled elastomeric matrix B. As is to be expected, G' is independent of the applied magnetic field.

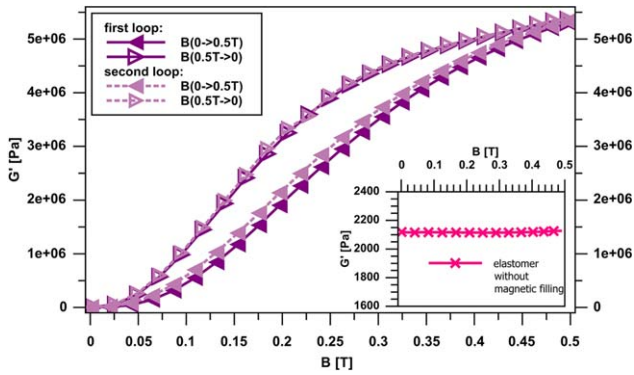


Figure 3. Example of the measurement of the storage modulus G' in dependence on the magnetic flux density B . The inset demonstrates that G' of the elastomeric matrix is independent of the applied magnetic flux density (matrix B, CIP concentration 80 wt %, RT curing). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The relative MR effect was defined as the relative change of the storage modulus at $B = 500$ mT:

$$MR = \frac{G'(500\text{mT}) - G'_0}{G'_0} \quad (2)$$

In Figure 4 the relative MR effect is shown for matrixes A and B. Due to the fact that several of the measured MAEs did not reach their saturation value in the achievable magnetic field range, the values at a flux density of 500 mT were used for comparison. With increasing iron content, the storage modulus measured at 500 mT increases up to 5 MPa with an iron content of 80 wt % for matrix A and up to 6 MPa for matrix B.

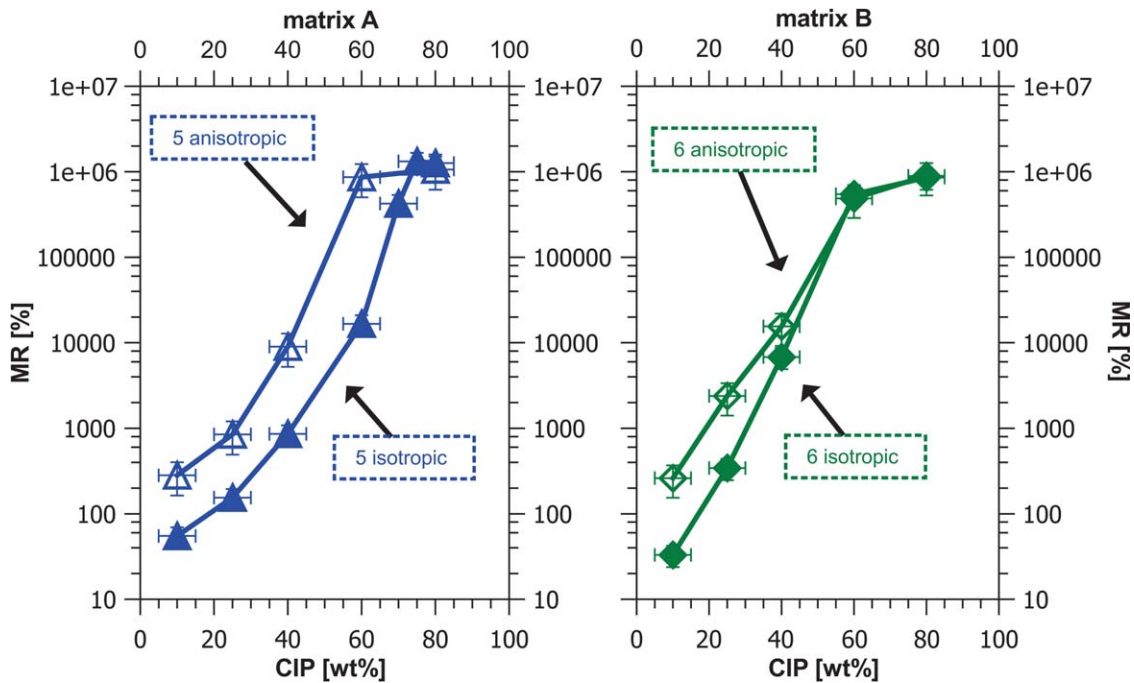


Figure 4. Relative MR effect dependent on the CIP content for matrix A (5) and B (6) after curing on HP. The lines connecting the experimental points serve as a guide to the eye. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The increase of the induced modulus with the increasing CIP content is roughly linear up to 50–60 wt %. For higher values of the CIP content the induced modulus rises less steeply. The relative MR effect has a maximum value at between 75 and 80 wt % of CIP. This corresponds to a range between 26 and 32 vol % CIP, which includes the critical particle volume concentration (CPVC) of 29.1 vol % for the CIP used.⁴¹ The CPVC is the value at which the particles are in direct contact and voids between them are filled with the elastomer base. This is the predicted maximum for the relative MR effect because at higher Fe contents the amount of elastomer is too small to fill the voids between the particles.⁴² The calculated CPVC value also agrees well with the percolation threshold $p_c = 0.29$ in a 3D continuum percolation model (a system of spatially uncorrelated, equally sized spheres, whose centers follow a Poisson distribution) known to be an accurate model for composite materials and polymers.⁴³ As was to be expected, in the vicinity of CPVC the MR effect is the same for isotropic and anisotropic samples.

In 2006, Zrínyi and coworkers introduced a phenomenological model for the interpretation of their results for the dependency of the magneto-induced storage modulus on an external field.¹⁵ This model was applied to anisotropic magnetoelastomers with iron contents of 10, 20, and 30 wt %.

By using the magnetic field dependency of the magnetically induced storage modulus and introducing a material parameter α_B , the following equation was obtained:

$$\Delta G' = G'_{\text{sat}} \times \frac{B^2}{a_B + B^2} \quad (3)$$

$\Delta G'$ is proportional to B^2 at small fields and reaches a saturation value G'_{sat} at high fields ($B \gg a_B^{1/2}$). In our case this

Table I. Summary of Magneto-mechanical Properties of MAE Samples with the Largest MR Effect Obtained with Different Preparation Steps

Matrix	CIP (wt %)	Curing		G'_0 (Pa)	G'_{sat} (MPa)	α_B (T ⁻²)	MR @0.5 T (%)	MR @B $\rightarrow\infty$ (%)
A	75	HP	<i>i</i>	273	10.88	0.58	1 322 504	3 979 015
		RT	<i>i</i>	515	4.72	0.08	753 471	915 819
	80	HP	<i>i</i>	409	6.75	0.10	1 263 894	1 649 279
			<i>a</i>		456	6.44	0.11	1 062 633
B	80	RT	<i>i</i>	1154	4.15	0.12	242 102	359 617
		HP	<i>i</i>	649	7.54	0.10	854 629	1 161 243
			<i>a</i>		669	7.86	0.11	896 109

Note: *i*: denotes isotropic samples; *a*: denotes anisotropic MAEs.

approach leads to very good agreement with experiment for samples with a low CIP content (≤ 50 wt %) when the magneto-induced modulus is not very high ($\leq 10^5$ Pa). We also found a good agreement for samples with high CIP content (≥ 75 wt %).

The saturation value $G'_{\text{sat}} = \Delta G' (B \rightarrow \infty)$ was used to estimate the maximum possible relative MR effect for our samples with a CIP content of about 80 wt %. Table I summarizes the results for samples with the largest MR effect. For MAEs with RT curing, the saturation values G'_{sat} received from the fit are roughly 1.2 MPa higher than the highest values measured experimentally, for MAEs cured at 100°C this difference is nearly 2 MPa. The calculated MR effect is higher for the heat cured MAEs than for the MAEs cured at RT. The sample with the highest measured MR effect of 1.3×10^6 % comprises matrix A. Instead of 80 wt % iron the filling is 75 wt %, which corresponds to the lowest initial storage modulus in this series. The storage modulus at 500 mT is comparable to that of the samples with 80 wt % CIP. However, the calculated saturation value of 10.9 MPa is much higher, which results in a possible MR effect of nearly 4 million%.

CONCLUSIONS

- A method for fabricating highly compliant elastomers and MAEs with the possibility of a long postcure period is presented. Only commercially available components are used. Curing at elevated temperature leads to a significant reduction in curing time.
- In the presented case addition of CIP makes the resulting composite material softer than the elastomeric matrix in the absence of magnetic field. The storage module has a local minimum at a particular value of CIP concentration.
- Systematic MR measurements of developed MAEs are performed. The MAEs exhibit a very low storage modulus ($\sim 10^2$ Pa), a high magneto-induced change of storage modulus ($\sim 10^6$ Pa) and therefore an extremely high MR effect.
- The MR effect increases with growing CIP concentration and has a maximum value at between 75 and 80 wt % of iron. The maximum MR effect is the same for both isotropic and aligned-particles samples. The largest MR effect measured at magnetic flux density B of 500 mT is 1 322 504%, the calculated MR effect for this composite at a theoretically infinite magnetic flux density is 3 979 015%.

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