5-21-2021

Temperature-Dependent Magnetic Properties of Magnetorheological Elastomers

Winnie M. Kiarie
Iowa State University, wmkiarie@iastate.edu

Kinjal Gandha
Ames Laboratory, kgandha@ameslab.gov

David C. Jiles
Iowa State University, dcjiles@iastate.edu

Temperature-Dependent Magnetic Properties of Magnetorheological Elastomers

Abstract

We report on an investigation of the temperature-dependent magnetic properties of magnetorheological elastomers (MREs). These are a class of composites that consist of magnetically permeable particles dispersed in a nonmagnetic polymeric matrix. Under the application of an external magnetic field, a large deformation occurs altering the mechanical properties of these materials. Due to their magnetoelastic coupling response, these materials are finding an increasing interest among the scientific community. These polymer-based composites' performance depends on many factors, which temperature is one of the biggest influencing factors requiring further investigation. In this work, the magnetic properties of isotropic and anisotropic polyurethane-based MRE with different iron (Fe) particle loading fractions (50, 60, 70, and 80% by weight) were investigated under different temperatures. From the analysis, the magnetization curves of these materials are observed to overlap for the different measured temperature values. The variation of various magnetic properties including saturation magnetization and differential susceptibility with temperature was also determined. The results show ~ 2.5% decrement in the saturation magnetization for each of the loading fractions, between the lowest (300K) and the highest (400K) measured temperature. On the other hand, the initial differential susceptibility exhibits different trends with increasing temperature. Generally, the magnetization response of these materials is seen to be only slightly sensitive to temperature changes. Additionally, the magnetization response is observed to be highly dependent on particle loading fractions and particle orientation within the elastomer.

Keywords

Temperature, Temperature measurement, Saturation magnetization, Magnetization, Magnetic properties, Perpendicular magnetic anisotropy, Magnetic fields

Disciplines

Electromagnetics and Photonics | Polymer and Organic Materials

Comments

This is a manuscript of an article published as Kiarie, Winnie M., Kinjal Gandha, and David C. Jiles. "Temperature-Dependent Magnetic Properties of Magnetorheological Elastomers." *IEEE Transactions on Magnetics* (2021). DOI: 10.1109/TMAG.2021.3082302. Posted with permission.

Temperature-Dependent Magnetic Properties of Magnetorheological Elastomers

Winnie M. Kiarie[1], *Member IEEE*, Kinjal Gandha[2], David C. Jiles[1,3], *Life Fellow, IEEE*

[1]Department of Materials Science and Engineering, Iowa State University, Ames, IA 50010, USA
[2]Ames Laboratory, US DOE, Ames, IA 50011 USA
[3]Department of Electrical and Computer Engineering, Iowa State University, Ames, IA 50010, USA

We report on an investigation of the temperature-dependent magnetic properties of magnetorheological elastomers (MREs). These are a class of composites that consist of magnetically permeable particles dispersed in a nonmagnetic polymeric matrix. Under the application of an external magnetic field, a large deformation occurs altering the mechanical properties of these materials. Due to their magnetoelastic coupling response, these materials are finding an increasing interest among the scientific community. These polymer-based composites' performance depends on many factors, which temperature is one of the biggest influencing factors requiring further investigation. In this work, the magnetic properties of isotropic and anisotropic polyurethane-based MRE with different iron (Fe) particle loading fractions (50, 60, 70, and 80% by weight) were investigated under different temperatures. From the analysis, the magnetization curves of these materials are observed to overlap for the different measured temperature values. The variation of various magnetic properties including saturation magnetization and differential susceptibility with temperature was also determined. The results show ~ 2.5% decrement in the saturation magnetization for each of the loading fractions, between the lowest (300K) and the highest (400K) measured temperature. On the other hand, the initial differential susceptibility exhibits different trends with increasing temperature. Generally, the magnetization response of these materials is seen to be only slightly sensitive to temperature changes. Additionally, the magnetization response is observed to be highly dependent on particle loading fractions and particle orientation within the elastomer.

***INDEX TERMS*— COMPOSITE, MAGNETIZATION, MAGNETORHEOLOGICAL ELASTOMERS, TEMPERATURE**

I. INTRODUCTION

Magnetorheological elastomers (MREs) are a class of smart composites that consists of magnetic polarizable particles dispersed in a non-magnetic elastomeric matrix[1]–[3]. The presence of magnetic particles in these materials couple the rheological properties of the elastomer to external magnetic field. Hence, when subjected to externally applied magnetic field these group of materials exhibit controllable, rapid and reversible changes in their rheological properties [4].

The property of these materials to alter their rheological properties have seen them gain significant attention due to their suitability for a range of engineering applications. These applications include but not limited to sensors, actuators, tunable vibration absorbers and vibration isolators [5]–[7]. Typically, the matrix of MREs are polymers such as natural rubber, silicone rubber, polydimethylsiloxane (PDMS) and polyurethane [8]–[11]. However, the limitation with such polymer-based materials arise from the viscoelastic properties of the polymer, which result in a high sensitivity to temperature [12].Since MRE-based devices often work in a wide range of magnetic field and temperatures, it is important to investigate the material behavior of these composites under different temperatures. This can, in many cases, determine the upper bound on the temperatures at which a material has suitable properties.

Several studies have been carried out on the temperature-dependent behaviors of MREs. For example, Wan *et al*. [13] investigated the effect of temperature on the dynamic properties of silicon rubber-based MREs. The results revealed a transition temperature of about 50°C for MRE samples under uniaxial compression with the storage modulus initially decreasing to a minimum at ~ 50°C and then increasing slightly with increasing temperature. Zhang *et al*. [14] evaluated the temperature-dependent mechanical properties of MRE with varying mixed rubber matrices (cis-polybutadiene rubber and natural rubber). The results showed that the temperature-dependent moduli varied for MRE with different rubber matrices.

Recently, Wen *et al*. [15] conducted research work on the magnetomechanical properties of MREs at temperatures ranging from 20°C to 50°C. From the results they showed that the initial modulus and magneto-induced modulus decreased with increasing temperature. However, previous studies on the temperature-dependent behavior of MREs, are mainly focused on the temperature-dependent mechanical behavior of these materials and little attention has been paid on the effect of temperature on the magnetic behavior. More importantly, the functionality of MREs is mainly determined by the magnetic-dipole interactions which is dependent on the spatial arrangement of the magnetic particles [16]. Although one of the characteristics of MREs is the magnetic particles are fixed in position in the elastomer so that there is no particles movement relative to the matrix, this assumption only holds in position when the modulus of MREs is high [17]. However, when the magnetic field is relatively high and the modulus is low, the particle's movement in the matrix is likely to occur affecting

Corresponding author: Winnie Kiarie. Author (e-mail: winniemunyiva77@gmail.com).

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier (inserted by IEEE).

the macroscopic magnetic behavior of the MRE. Temperature induced changes in the viscoelastic properties including elastic modulus of the polymeric matrix [15] can affect the spatial arrangement of the magnetic particles. Hence, it's essential to determine the effect of temperature on the magnetic properties of these materials, which is helpful for their practical application and mechanism analysis.

In this present work the influence of temperature on the magnetic properties of MREs was determined. The magnetization response under different temperatures for isotropic and anisotropic polyurethane based MREs with different iron (Fe) particle loading fractions was measured. Further, various temperature-dependent magnetic properties were obtained.

II. Experimental procedure

The MRE was fabricated by dispersing near spherical iron powder (99%, 5μm; US Research Nanomaterials, Inc.) into a two-component 30 Shore A polyurethane elastomer (L-3530 REV 1 A/B; BJB Enterprises). The polyurethane elastomer was prepared by combining Part A and Part B in a 1:1 ratio by mass and Iron powder was then added (50, 60, 70 and 80% by weight) to the liquid polyurethane. The material was mixed using a mechanical stirrer. The resulting mixture was poured into individual cylindrical molds for curing. Isotropic samples were then cured in a convection oven at 80°C for 15 hours in absence of an externally applied magnetic field. For anisotropic samples, the mixture was cured in the presence of an externally applied magnetic field using permanent magnet assembly (Bx882-N52, surface magnetic flux density 0.23T; K&J magnetics) placed over the top and the bottom of the cylindrical mold. Magnetic properties were measured in the magnetic field of 15 kOe using a quantum design vibrating sample magnetometer (VSM).

III. Results and discussions

Fig. 1, shows the influence of temperature on the magnetic hysteresis loops of isotropic MRE with different particle loading fractions (50, 60, 70 and 80wt%). It can be seen from the experimental data that the MRE exhibits a typical soft magnetic material property, low coercivity, residual magnetization and a high saturation magnetization. For all the samples, the magnetization curves, are observed to overlap for the different measured temperature values. This shows there is only a small change in the magnetization response with increasing temperature. This behavior can be attributed to two reasons: first, the fact that the measured temperature is far below the curie temperature of Fe (T_c=770ºC≡1043K). Therefore, it can be said that there is minimal changes in the magnetization of the individual Fe particles due to changes in the measured temperature. Second, the temperature-induced changes in the interparticle distance between the magnetic particles due to change in the rheological properties of the MRE matrix which are known to be highly temperature dependent [18] is seen to affect the overall magnetization of the composite only slightly (see Fig.1). The effect of temperature on the magnetic properties including saturation magnetization (M_s) and initial differential susceptibility was also determined. Fig. 2 illustrates the temperature dependence of saturation magnetization for different particle loading fractions. It can be noticed, for each of the particle loading fractions there is a small decrease in the saturation magnetization with increase in temperature. For example, the M_s in the 80wt% MRE drops from 170.264 emu/g at 300K to 166.348 at 400K. The decrement is calculated to be ~2.5% for all the samples between the lowest temperature and the highest temperature. It is also worth noting that the saturation magnetization varies significantly with the particle loading fractions. That is the saturation magnetization is observed to increase with increase in the particle mass fractions (see Fig. 2).

Further, the temperature dependent-initial volume differential susceptibility (χ_v') in SI units which was calculated as follows

$$\chi_v'=4\pi\rho\chi_m \quad (1)$$

where ρ is materials density and χ_m is mass susceptibility ($\frac{dM(\frac{emu}{g})}{dH(Oe)}$) for different particle loading fractions was explored. Since $M_s^{MRE}=\varphi M_s^{Fe}$ where φ is the particle loading fraction, back calculations were carried out to determine the obtained weight fractions. The measured Fe powder saturation magnetization at 300K was 220 emu/g. This confirmed the purity of the Fe powder obtained from the manufacturer. The value was then used to determine the particle loading fractions, which was 51, 60, 67, 77wt% and 51, 56, 67, 80wt% for isotropic and anisotropic samples, respectively. These were closely related to the intended weight fractions and hence were used to determine the densities used in the χ_v' calculation.

The influence of temperature on χ_v' for samples with different particle loading fractions is shown in Fig. 3. From the analysis, different trends of changes of the χ_v' with increasing temperature were observed. For all the samples, there is an initial decrease in the χ_v', which then increases slightly, before either decreasing or increasing with increasing temperature. Generally, as it can be seen the change is small for all the samples except for the sample with 80wt% mass fractions of the particles where 26% increment in χ_v' is obtained between the lowest and highest temperature.

To evaluate the temperature sensitivity with magnetic anisotropy of the MRE, the magnetization response of the sample with particles orientation parallel to the applied magnetic field was compared with the response obtained from the perpendicular sample for different particle loading fractions and temperatures as shown in Fig 4. As readily seen, the temperature-dependent magnetization is independent of the particle chains orientation since there is only a slight change in magnetization with increasing temperature for the parallel and transverse particle chains orientation with the applied magnetic field. However, as expected, the magnetization of the MRE is magnetic anisotropy-dependent with the magnetization for the parallel samples observed to be higher than that of perpendicular samples except at the saturation magnetization where the curves coincide, demonstrating the

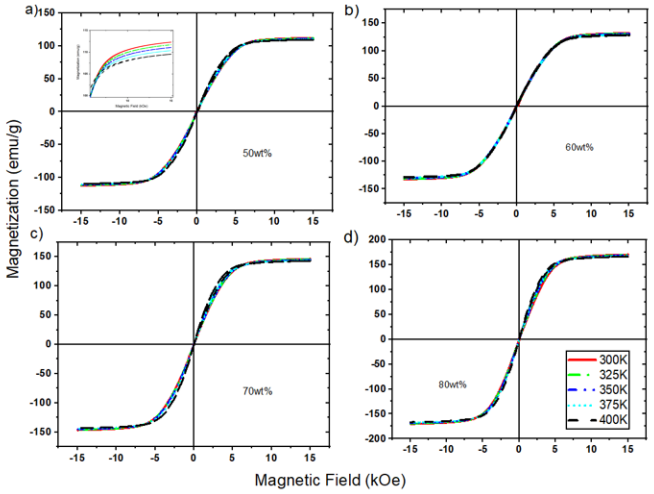

Fig. 1. Magnetization curves for isotropic MREs with different loading fractions a) 50 b) 60 c)70 d)80wt%) and varying temperatures. The inset shows magnetization zoomed in at higher magnetic field.

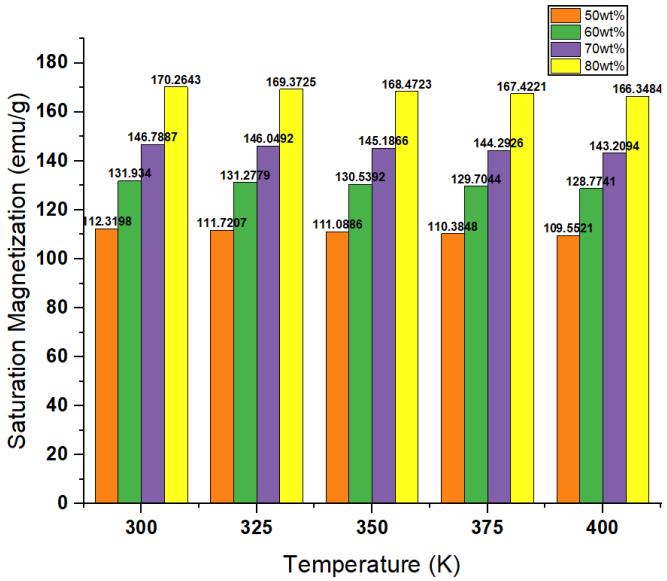

Fig. 2. Variation of saturation magnetization with temperature for isotropic MRE with varying particle loading fraction

insensitivity of saturation magnetization to the orientation of the magnetic particles. Further, the dependency of differential volume susceptibility in SI units $\chi' = 4\pi\rho\frac{dM}{dH}(emug^{-1}Oe^{-1})$ on magnetic field and different temperatures for 70wt% MRE with particle chains oriented in the direction of the applied magnetic field was compared as shown in Fig. 5. Here, 70wt% is selected since similar weight fraction has been used for analysis in our previous studies [3], [19]. In the following when we refer to susceptibility, differential volume susceptibility is implied. From the graph, differences are observed in the susceptibility as magnetic field (H) tends towards zero with susceptibility at 400K observed to be highest which agrees with some of the results from the isotropic (unaligned) samples (Fig. 3). This can be attributed

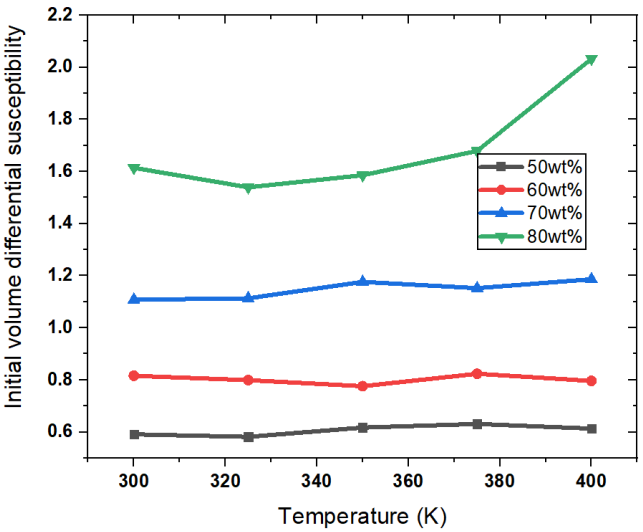

Fig. 3. Initial volume differential susceptibility as a function of temperature for isotropic MRE with different particle loading fraction.

to decrease in modulus of the elastomer which in turn may result to the increased susceptibility seen in these samples at higher temperature. This may be due to magnetically-induced motion of the magnetic particles within the elastomer due to decrease in the elastic modulus of the elastomer hence aligning further the particles with the applied magnetic field. The particle's movement has been reported in the previous studies on the magnetic properties of MREs [18].

A comparison of the susceptibility with particle-chains alignment to the applied field was determined for 300K temperature as depicted in Fig. 6. The results show the susceptibility is highly sensitive to the orientation of the magnetic particles inside the MRE with respect to the direction of the applied magnetic field. The susceptibility is observed to be highest when the particle-chains alignment in the elastomer is parallel to the direction of the applied field as $H \to 0$. The perpendicular alignment to the direction of the applied H has the least susceptibility while isotropic MREs susceptibility is observed to be between the anisotropic samples. Although at magnetic fields H > 5kOe the orientation of the particle chains perpendicular to the direction of the applied magnetic field is observed to have the highest susceptibility before coincidence of the susceptibility for all the particle orientations at higher magnetic fields (see Fig. 6). These results agree with most experiments carried out on these materials which have found their behavior to be highly dependent on the particles orientation within the samples [9], [18].

Therefore, the main observations that can be made from these experiments include: first, the magnetization response is practically insensitive to the measured temperature, which means that the temperature-induced change of the distance of separation between the magnetic particles has only a small effect on the average magnetization of these composites.

This article has been accepted for publication in a future issue of this journal, but has not been fully edited. Content may change prior to final publication. Citation information: DOI 10.1109/TMAG.2021.3082302, IEEE Transactions on Magnetics

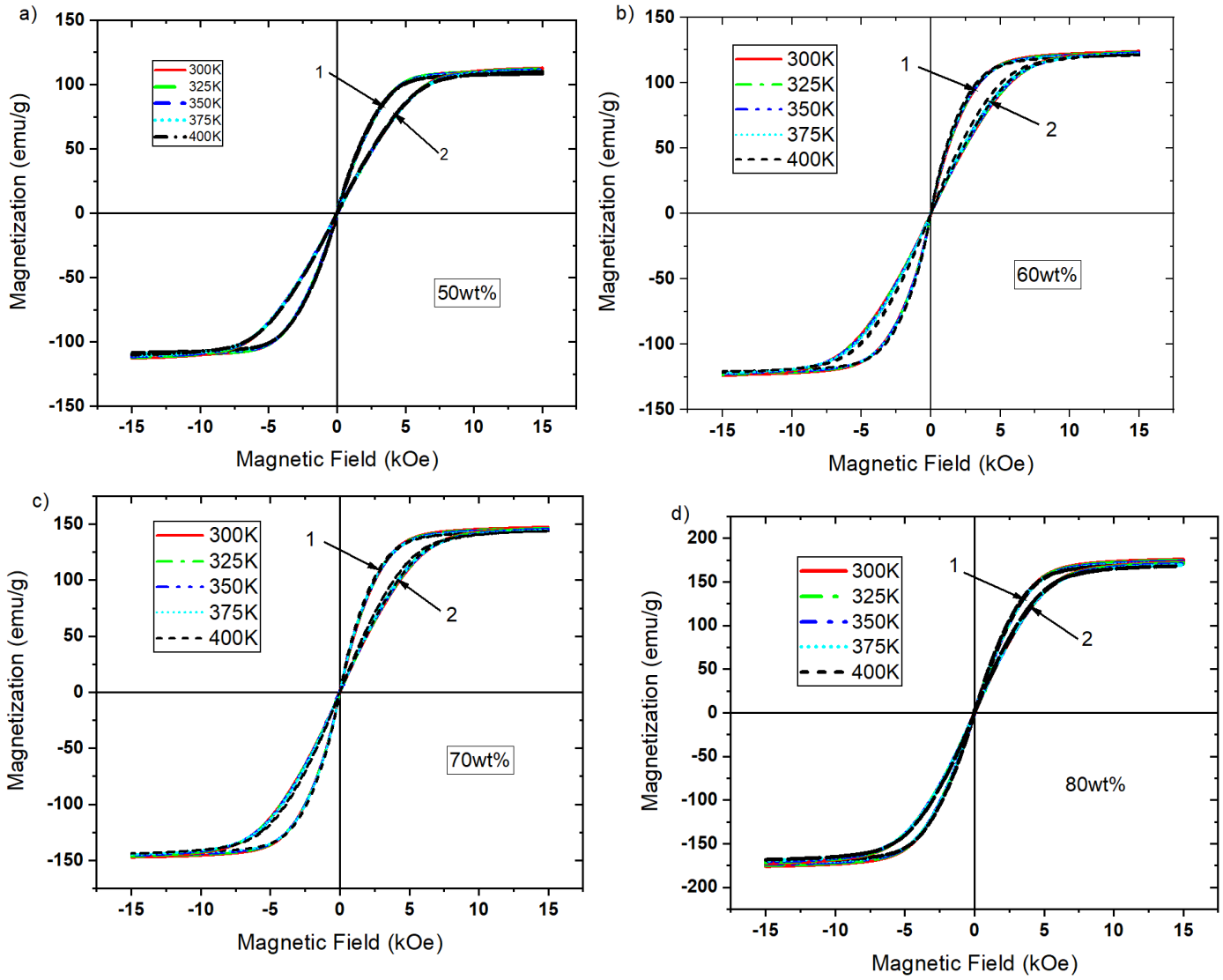

Fig. 4: Magnetization curves for anisotropic MREs with different loading fractions a) 50 b) 60 c) 70 d) 80wt%), temperatures and particle orientations with curve 1 parallel and curve 2 perpendicular to the directions of the applied field.

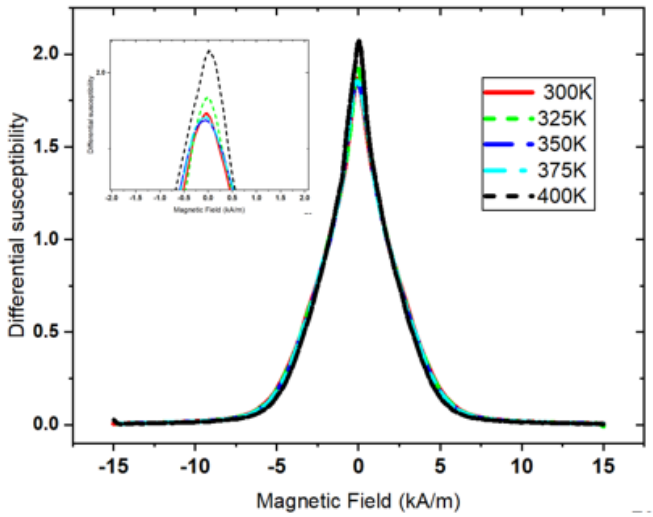

Fig. 5. Temperature dependency of differential susceptibility of anisotropic 70wt% MRE in the direction of the applied magnetic field

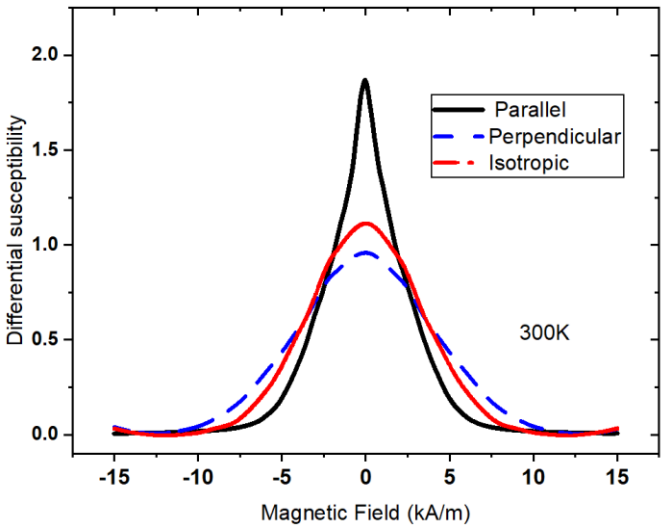

Fig. 6. Differential susceptibility of 70wt% MRE with different particle-chains orientation to the applied magnetic field.

Second, the magnetization response of these materials is dependent on the particle loading fractions, with the magnetization observed to increase with increase in the particle loading fractions. Finally, the magnetization is sensitive to the orientation of the magnetic particles inside the MRE with respect to the direction of the applied magnetic field and particularly in the initial, linear range of this response.

IV. Conclusion

In this work, an investigation of the effect of temperature on the magnetic properties of unaligned and aligned MREs with different particle loading fractions was carried out. In general, only small changes in the magnetization behavior of these materials with increasing temperature was observed. However, significant dependency of the magnetic properties of the MRE with particle loading fractions and particle orientation in the elastomer were seen. From this study the magnetic properties of these materials are observed to be relatively stable within the measured temperatures which is an expected working temperature range of MRE-based devices.

V. Acknowledgment

This work is supported in part by Link Manufacturing, Ltd, in part by Center of Research and Service, Iowa State University, and in part by Regents of Innovation Funds. The work was performed at Ames Laboratory, operated for the U.S. Department of Energy by Iowa State University of Science and Technology under contract No. DE-AC02-07CH11358.

VI. References

[1] A. K. Bastola, M. Paudel, L. Li, and W. Li, "Recent progress of magnetorheological elastomers: A review," *Smart Mater. Struct.*, vol. 29, no. 12, 2020.

[2] Y. Li, J. Li, W. Li, and H. Du, "A state-of-the-art review on magnetorheological elastomer devices," *Smart Mater. Struct.*, vol. 23, no. 12, pp. 1–24, 2014.

[3] W. M. Kiarie and D. Jiles, "A study of the effect of mechanical preloads on hysteresis loop of magnetorheological elastomers," *AIP Adv.*, vol. 11, no. 1, 2021.

[4] H. Vatandoost, R. Sedaghati, S. Rakheja, and M. Hemmatian, "Effect of pre-strain on compression mode properties of magnetorheological elastomers," *Polym. Test.*, vol. 93, no. June 2020, p. 106888, 2021.

[5] P. Testa *et al.*, "Magnetically Addressable Shape-Memory and Stiffening in a Composite Elastomer," *Adv. Mater.*, vol. 31, no. 29, pp. 1–6, 2019.

[6] Y. Umehara *et al.*, "Railway actuator made of magnetic elastomers and driven by a magnetic field," *Polymers (Basel).*, vol. 10, no. 12, pp. 1–7, 2018.

[7] A. K. Bastola and L. Li, "A new type of vibration isolator based on magnetorheological elastomer," *Mater. Des.*, vol. 157, pp. 431–436, 2018.

[8] H. S. Jung, S. H. Kwon, H. J. Choi, J. H. Jung, and Y. G. Kim, "Magnetic carbonyl iron/natural rubber composite elastomer and its magnetorheology," *Compos. Struct.*, vol. 136, pp. 106–112, Feb. 2016.

[9] X. Guan, X. Dong, and J. Ou, "Magnetostrictive effect of magnetorheological elastomer," vol. 320, pp. 158–163, 2008.

[10] B. Mordina, R. K. Tiwari, D. K. Setua, and A. Sharma, "Magnetorheology of polydimethylsiloxane elastomer/FeCo3 nanocomposite," *J. Phys. Chem. C*, vol. 118, no. 44, pp. 25684–25703, 2014.

[11] J. Wu, X. Gong, Y. Fan, and H. Xia, "Anisotropic polyurethane magnetorheological elastomer prepared through in situ polycondensation under a magnetic field," *Smart Mater. Struct.*, vol. 19, no. 10, 2010.

[12] J. M. L. Dos Reis, "Effect of temperature on the mechanical properties of polymer mortars," *Mater. Res.*, vol. 15, no. 4, pp. 645–649, 2012.

[13] Y. Wan, Y. Xiong, and S. Zhang, "Temperature dependent dynamic mechanical properties of Magnetorheological elastomers: Experiment and modeling," *Compos. Struct.*, vol. 202, pp. 768–773, Oct. 2018.

[14] W. Zhang, X. Gong, S. Xuan, and W. Jiang, "Temperature-dependent mechanical properties and model of magnetorheological elastomers," *Ind. Eng. Chem. Res.*, vol. 50, no. 11, pp. 6704–6712, 2011.

[15] Q. Wen *et al.*, "Temperature dependent magneto-mechanical properties of magnetorheological elastomers," *J. Magn. Magn. Mater.*, vol. 497, no. May 2019, p. 165998, 2020.

[16] A. M. Biller, O. V. Stolbov, and Y. L. Raikher, "Modeling of particle interactions in magnetorheological elastomers," *J. Appl. Phys.*, vol. 116, no. 11, 2014.

[17] G. V. Stepanov, D. Y. Borin, Y. L. Raikher, P. V. Melenev, and N. S. Perov, "Motion of ferroparticles inside the polymeric matrix in magnetoactive elastomers," *J. Phys. Condens. Matter*, vol. 20, no. 20, 2008.

[18] A. V. Bodnaruk *et al.*, "Temperature-dependent magnetic properties of a magnetoactive elastomer: Immobilization of the soft-magnetic filler," *J. Appl. Phys.*, vol. 123, no. 11, 2018.

[19] W. M. Kiarie, E. J. Barron, A. P. S. Baghel, I. C. Nlebedim, M. D. Bartlett, and D. C. Jiles, "Modeling of Magnetic Properties of Magnetorheological Elastomers Using JA Hysteresis Model," *IEEE Trans. Magn.*, vol. 57, no. 2, 2021.