

Thermal Conduction and Magnetic Properties of Magnetorheological Elastomers Dispersing Sendust Particles

Yasushi Ido¹, Yusuke Hiroshima¹, Yuhiro Iwamoto¹, Yasutake Hirota²,
Satomi Fujioka²

¹*Department of Electrical and Mechanical Engineering, Nagoya Institute of Technology,
Gokiso-cho, Showa-ku, Nagoya, Japan*

²*Ferrotec Material Technologies Corporation, 2-3-4, Nihonbashi, Chuo-ku, Tokyo, Japan
E-mail: ido.yasushi@nitech.ac.jp*

In this paper, the thermal and magnetic properties of the Sendust particles dispersed magnetorheological elastomer (MRE) were investigated experimentally. Sendust is the material whose saturation magnetization is high and coercive field strength is low. Two types of specimens were prepared: one was the Sendust particles randomly dispersed MRE and the other was the MRE having chain-like clusters of Sendust particles. The thermal conductivity of the MRE having chain-like clusters depends on the internal structure formed by the particles. When the volume fraction of particles is 15 vol.%, the thermal conductivity of the MRE in the direction parallel to the chain-like clusters is 1.43 times of that of the MRE with randomly dispersing particles, while the thermal conductivity of the MRE in the direction perpendicular to the chain-like clusters is 0.90 times of that of the MRE with randomly dispersing particles. The initial magnetic permeability of the Sendust particles dispersed MRE depends on the internal structure of dispersed particles, while the saturation magnetization of the MRE is determined by the volume fraction of particles in the MRE and hardly depends on the particle distribution.

Keywords: Magnetorheological Elastomer, Sendust, Thermal Conductivity, Magnetization, Anisotropy, Magnetic Particle.

1. Introduction

Magnetorheological (MR) materials are magnetic functional materials which react against an applied magnetic field^{1,2}). Basically, the magnetorheological materials are mixture of iron particles and a low-permeability carrier material. When external magnetic field is applied to

the magnetic functional materials, dispersed magnetic particles interact each other and microstructure is formed inside the materials or strength of interactions between magnetic particles are changed. Thus, the mechanical properties of the magnetic functional materials are changed by applying magnetic field. One of the typical examples of the magnetic functional materials is a MR fluid as a liquid-state. MR fluids and MR devices were first reported by Rabinow^{3,4}). In the absence of external magnetic field, magnetic particles in a MR fluid are randomly dispersed and the fluid shows Newtonian fluid like behavior. When the external magnetic field is applied, the magnetic particles form chain-like clusters and such microstructure effects on the macroscopic properties such as viscosity^{5,6}).

Another type of magnetic functional materials is a magnetorheological elastomer (MRE). In the MREs, magnetic particles are embedded in the non-magnetic matrix^{2,7,8}). The magnetic particles inside the MREs can be homogeneously dispersed or form chain-like columnar structures by applying uniform magnetic field during cur processes. The MREs have been developed for applications such as vibration isolation^{9,10}), due to their stiffness change under applied magnetic field.

Sendust is the ternary alloy consisting of iron, silicon and aluminum and was first reported by Masumoto and Yamamoto¹¹). Sendust has a high initial permeability and low hysteresis loss, and researches on the effect of composition on the loss of powder magnetic cores¹²), electromagnetic wave absorption characteristics^{13,14}), low-loss design of inductors^{15,16}) have been conducted. Sendust has excellent magnetic responsiveness and has the properties of keeping core loss low.

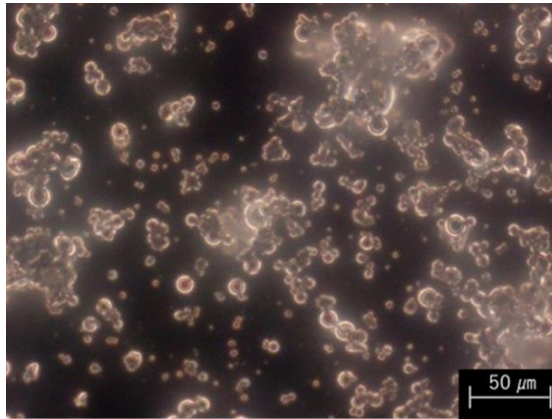
In this study, the Sendust particles dispersed magnetorheological elastomers have been developed and their thermal and magnetic properties were investigated experimentally. Two types of the Sendust particles dispersed MREs were prepared: one was the MRE with randomly distributed Sendust particles and the other was the MRE having chain-like clusters of particles by applying uniform magnetic field during the cur process. It is expected that an MRE with thermal conductivity anisotropy and magnetic properties like Sendust will be obtained. Thermal conductivity of the MREs was measured by using the steady state parallel plate method¹⁵). The thermal conductivity of the MRE with randomly dispersed Sendust particles was compared with the thermal conductivity of the MRE containing chain-like clusters in the direction of the cluster formation and in the direction perpendicular to the cluster formation direction. Magnetic properties such as magnetization were also investigated by using a BH analyzer. The effects of microstructure formation inside the MRE and magnetic anisotropy were examined experimentally.

2. Experiments

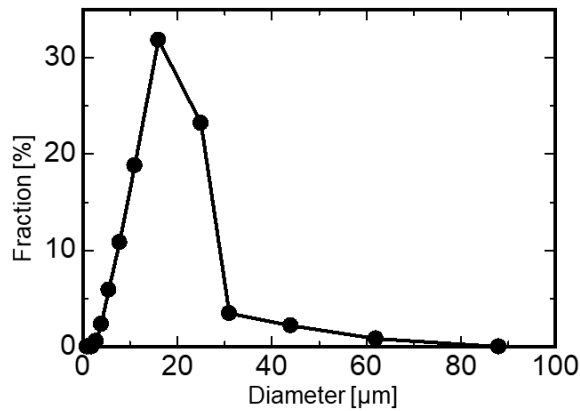
2.1 Specimen

In this study, Sendust particles dispersed MRE has been developed. The specimens were produced by mixing Sendust particles (PST-S, Sanyo Special Steen Co., Ltd.) and silicone gel (PDMS Sylgard184, Daw). The average diameter of the spherical Sendust particles was 16.8 μm and the density is 7.05 g/cm^3 . The photograph of the Sendust particles taken by the

microscope (IX73, Olympus) is shown in Fig.1(a) and the size distribution of the Sendust particles is also shown in Fig.1(b).



(a) Photograph of Sendust particles



(b) Size distribution

Figure 1 Photograph of Sendust particles (PST-S, Sanyo Special Steel Co., Ltd.) taken by the microscope and the size distribution of the Sendust particles.

To investigate the anisotropy due to the microstructure formed by the Sendust particles, we prepared two types of samples: one was the MRE with randomly dispersed Sendust particles, and the other was the MRE having chain-like clusters of Sendust particles. The MREs containing Sendust particles were produced as follows: (a) add Sendust particles to the silicone gel base and stir using an ultrasonic homogenizer (UR-21P, Tomy) and other equipment. (b) Add the curing agent to the mixture prepared in (a) and stir. (c) The mixture prepared in (b) is placed in a vacuum degassing device (VD-VL, AS ONE) and subjected to vacuum degassing. (d) The mixture of silicone gel and Sendust particles is poured into the mold and sealed. (e) when manufacturing MRE with randomly dispersed particles, the raw material placed in the mold is heat-cured at 70°C for about one hour using a constant temperature dryer (DOV-300P, AS ONE). On the other hand, when fabricating an MRE with chain-like clusters of particles, two same neodymium permanent magnets are arranged above

and below the mold containing the raw materials, and the mold is allowed to harden naturally for 24 hours. For the thermal conductivity measurement experiments, the volume fraction of Sendust particles was 5, 10 and 15 vol.% and the size of the specimen was 40×40×40 mm. The volume fraction was 5, 7.5 and 10 vol.% and the size of the specimen was 30×30×30 mm for magnetic properties measurement experiments.

2.2 Thermal conductivity experiments

Thermal conductivity of the MRE containing Sendust particles was measured by using the steady state parallel plate method. Figure 2 shows the schematic of equipment for the steady state parallel plate method. The temperatures on the heated and cooled surfaces of the specimen were measured using NTC thermistors (NXFT15XH103FEAB030, Murata Manufacturing Co., Ltd.) for temperature detection installed on both sides of the specimen. The heat flux is monitored by a heat flux sensor (HF-40, CAPTEC). At steady state, the thermal conductivity λ can be obtained from the following equation:

$$q_z = -\lambda \frac{\partial T}{\partial z} = -\lambda \frac{T_H - T_C}{l}, \quad (1)$$

where q_z is the heat flux in the z -direction, l is the thickness of the specimen, T_H is the temperature of the top surface (heated side) of the specimen, and T_C is the temperatures of the bottom surface (cooled side).

Figure 3 shows the schematic of the experimental apparatus for measuring the thermal conductivity of the MRE containing Sendust particles. A constant voltage was applied to the heater (F-01-001, Jemix) by a DC power supply (GPS-3030D, Instech) to heat the upper surface of the specimen, and a constant temperature bath (LTB-400 α , AS ONE) was used to cool the lower surface of the specimen with cooling water at a constant temperature. During the experiments, the test section was arranged in a chamber and a low vacuum state was created using a vacuum pump (G-50SA, ULVAC). The thermal conductivity of acrylic resin was measured using the self-made steady state parallel plate method equipment. As a result, the measured value of 0.24 W/mK was obtained for the thermal conductivity of acrylic resin of 0.21 W/mK, so the relative error is 14.3 %.

The experiment was conducted under three conditions as shown in Fig.3. That is, the case of MRE with randomly dispersed particles, when the heat flux and the cluster formation direction are parallel, and when the heat flux and the cluster formation direction are orthogonal.

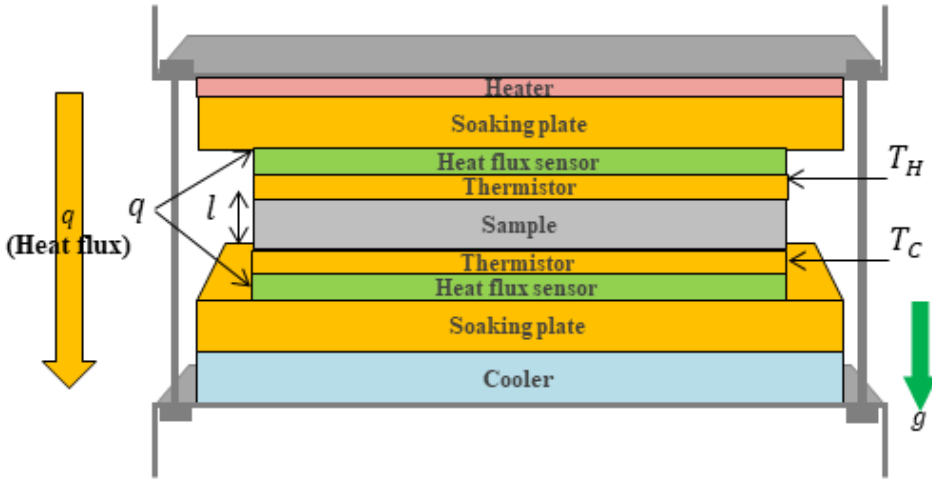


Figure 2 Schematic of the experimental apparatus using the steady state parallel plate method.

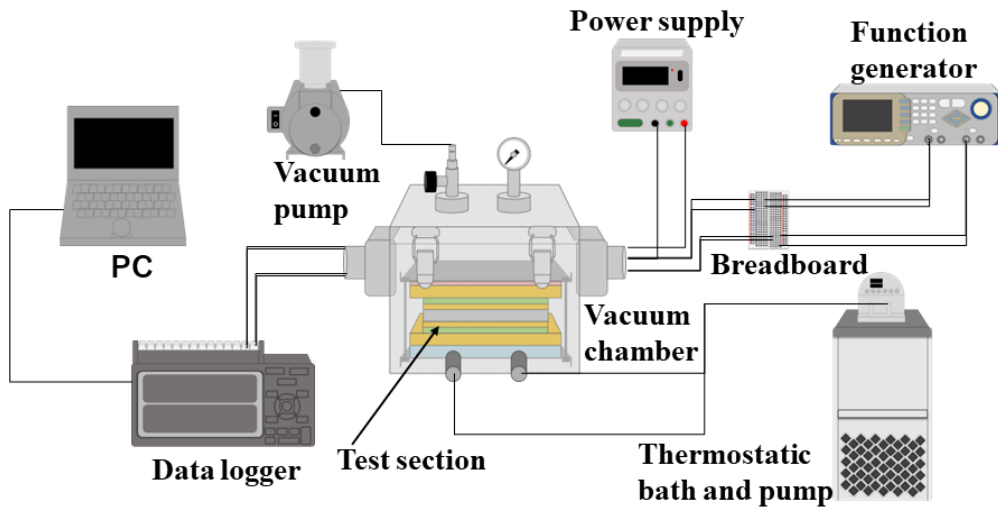


Figure 3 Schematic of the experimental apparatus for measuring thermal conductivities.

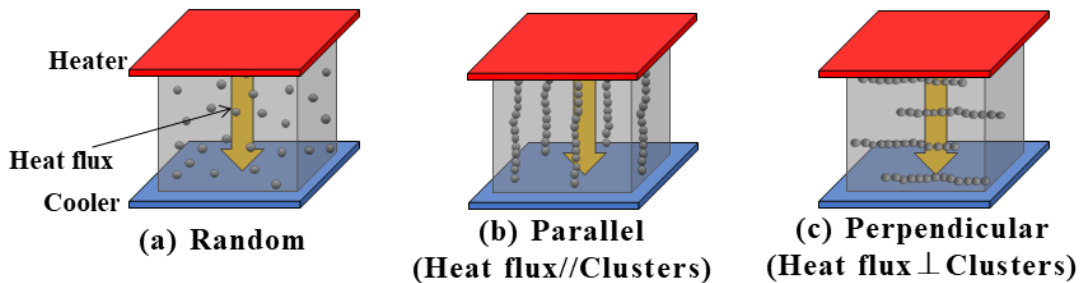


Figure 4 Experimental conditions for thermal conductivity measurements

2.3 Magnetic properties measurements

The magnetic properties of the MRE containing Sendust particles were examined using the closed magnetic circuit measurement method. For this measurement, a BH analyzer (BH-1000H, Denshijiki Industry Co., Ltd.) was used and the photograph of the experimental equipment is shown in Fig.5. A vertical electromagnet (EMD-80M25M, Denshijiki Industry Co., Ltd.) was used for excitation, and quasi-static measurements were performed for 16 seconds per loop with a 4-quadrant bipolar power supply (BWS40-15, Takasago Ltd.). Since the detection coil is also excited by the signal from the air-gap magnetic flux, this was removed by using a compensating coil.

To investigate an anisotropy of magnetic properties of the MRE containing Sendust particles, the experiment was conducted under three conditions as shown in Fig.6. That is, the case of MRE with randomly dispersed particles, when the magnetic field and the cluster formation direction are parallel, and when the magnetic field and the cluster formation direction are orthogonal.



Figure 5 Experimental apparatus for magnetic properties measurements. BH analyzer (BH-1000H, Denshijiki Industry Co., Ltd.)

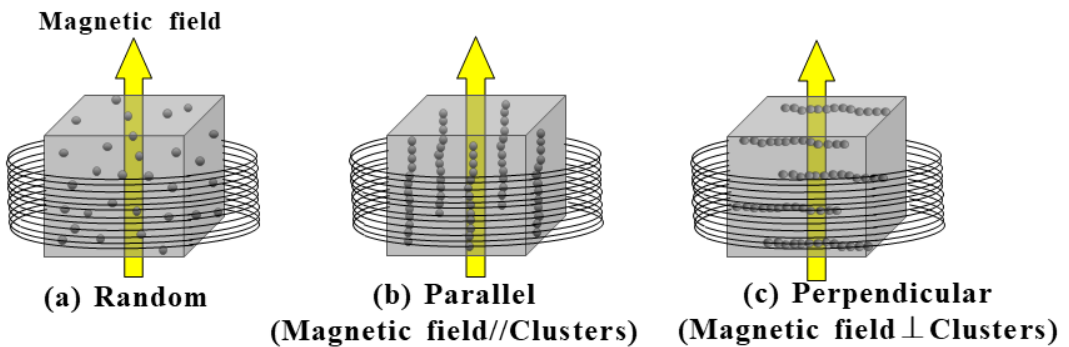


Figure 6 Experimental conditions for magnetic properties measurements

3. Results and discussion

3.1 Thermal conductivity

The thermal conductivity of the MRE was measured using the steady state parallel plate method for three cases shown in Fig.4. The experimental results are shown in Fig.7. Figure 8 shows the enhancement ratio of the thermal conductivity of the MRE containing chain-like clusters of particles against that of the MRE with randomly dispersed particles. From Fig.7, regardless of the internal structure of the MRE formed by Sendust particles, the thermal conductivity increases as the volume fraction of Sendust particles increases. Compared with the thermal conductivity of MRE randomly dispersed particles, the thermal conductivity of the MREs containing chain-like clusters increases by up to 43 % in the direction of cluster formation, while it decreases about 10 % in the direction orthogonal to the direction of cluster formation. From Fig.8, the enhancement ratio of the thermal conductivity of the MREs containing chain-like clusters in the direction of cluster formation increases with increase of the volume fraction of Sendust particles. On the other hand, the enhancement ratio of the thermal conductivity of the MREs in the direction perpendicular to the cluster formation takes minimum value at the volume fraction of 10 vol.%. This can be explained by the cluster structure formed inside the MREs. From the previous papers^{18,19}), when the volume fraction of magnetic particles is low, the chain-like clusters formed by the magnetic particles are isolated from each other. When the volume fraction of magnetic particles increases to a certain extent, chain-like clusters coalesce to form wall-like structures. As the volume fraction of magnetic particles increases further, coalescence progresses further, forming a network-like structure when viewed from the direction of cluster formation. Based on these results, the results in Fig.8 can be explained as follows. When the volume fraction of particles is 5 vol.%, chain-like clusters are almost isolated, but the total volume of particles is small. Therefore, the thermal conductivity in the direction perpendicular to the cluster formation direction does not show a large difference compared to the thermal conductivity in the case of MRE with randomly dispersed particles. When the volume fraction of particles is 10 vol.%, many of the chain-like clusters exist in isolation, but only a few of them coalesce to form a wall-like structure. Therefore, the thermal conductivity in the direction perpendicular to the cluster formation direction is significantly different from that of the MRE with randomly dispersed particles. When the volume fraction of particles is 15 vol.%, many chain-like clusters coalesce to form wall-like structures. Therefore, the thermal conductivity in the direction orthogonal to the direction of cluster formation does not show a large difference compared to the thermal conductivity of the MRE with randomly dispersed particles.

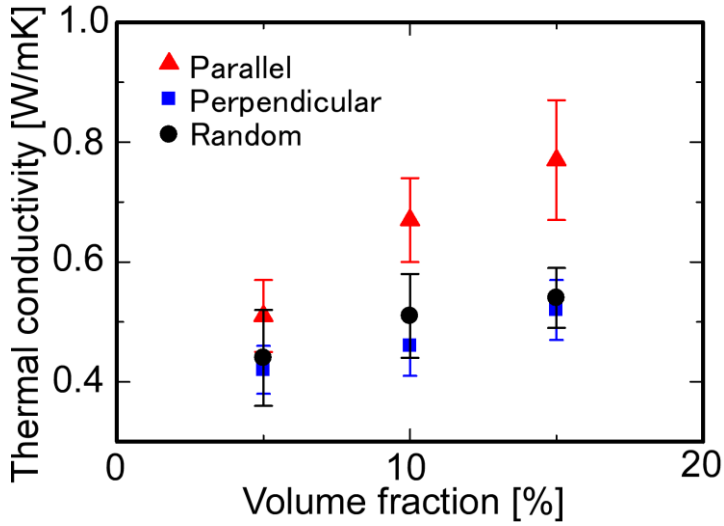


Figure 7 The thermal conductivities of the MRE dispersing Sendust particles

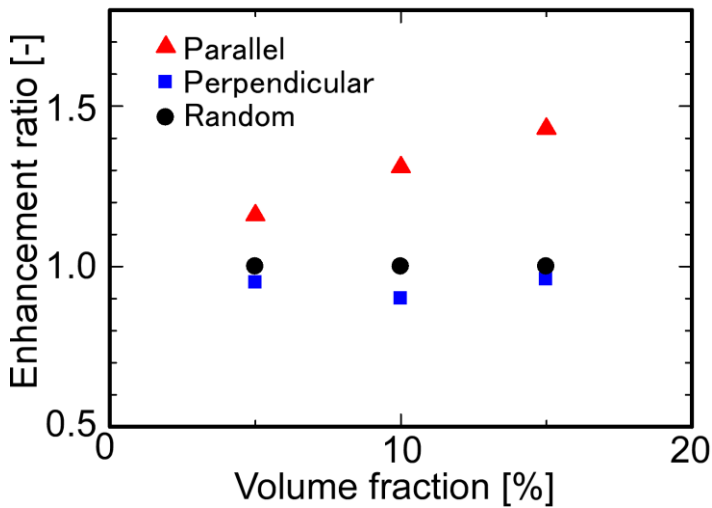


Figure 8 The enhancement ratio of the thermal conductivity of the MRE having chain-like clusters against that of MRE with randomly dispersed particles

To evaluate the effect of chain-like clusters on the thermal conductivity, we introduce the extended Landau-Lifshitz equation¹⁹⁾. This is an extension of the Landau-Lifshitz equation applied to the nanofluids by Warriar and Teja²⁰⁾ so that it can be applied to spherical particles. The particles with the thermal conductivity λ_p are distributed in the mother liquid with the thermal conductivity λ_f and the volume fraction of the particles is ϕ . The thermal conductivity considering the chain-like clusters of the particles from the viewpoint of arithmetic mean, harmonic mean, and geometric mean can be calculated by the Landau-Lifshitz equation²⁰⁾. The extended Landau-Lifshitz equation¹⁹⁾ is given by

$$(\lambda_{eff})^n = (\lambda'_p)^n \phi' + (\lambda_f)^n (1 - \phi'), \quad (-1 < n < 1, n \neq 0) \tag{2}$$

$$\lambda_{eff} = (\lambda'_p)^{\phi'} (\lambda_f)^{1-\phi'}, \quad (n = 0) \tag{3}$$

where ϕ' is the modified volume fraction defined by

$$\phi' = \frac{6\phi}{\pi}. \tag{4}$$

The modified thermal conductivity of the particles is written as

$$\lambda'_p = \frac{2\lambda_f}{\beta} \left[\frac{1}{\beta} \ln \frac{1}{1-\beta} - 1 \right] + \frac{(4-\pi)\lambda_f}{\pi}, \tag{5}$$

where β is defined by

$$\beta = 1 - \frac{\lambda_f}{\lambda_p}. \tag{6}$$

Figure 10 shows the index of state n indicating the state of the internal structure of the particles in the fluid assuming that the thermal conductivity of the Sendust particles is 80.5 W/mK and that of the fluid (silicone potting gel) is 0.27 W/mK. When the volume fraction of particles is small, the thermal conduction of the base fluid is dominant, and the index of state should be close to zero. However, from Fig.10(a), for low volume fraction of particles, the index of state is significantly larger than zero in all cases. This is because the change in the thermal conductivity value by the Landau-Lifshitz equation with respect to the index of state is very small, so the effect of measurement error is large. From Fig.10(b) and (c), it can be seen from the spread of the index of state n distribution that the influence of the dispersed particles on the thermal conduction is relatively large compared to thermal conduction by the base fluid. From Fig.10, the strongest anisotropy of thermal conductivity is observed when the volume fraction of particles is 10 vol.%.

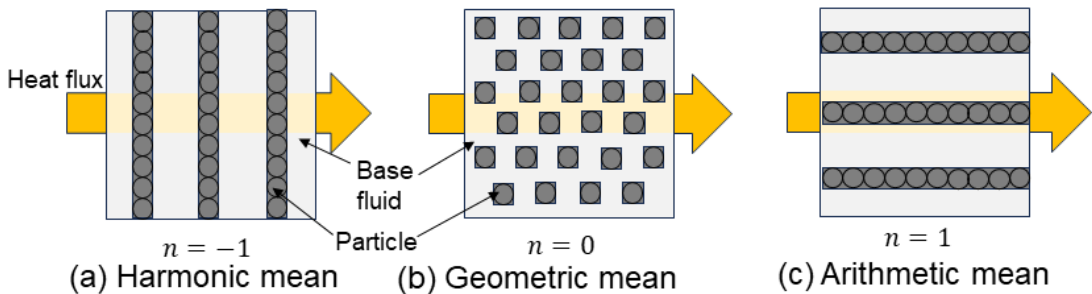


Figure 9 Theoretical model of the extended Landau-Lifshitz equation(19).

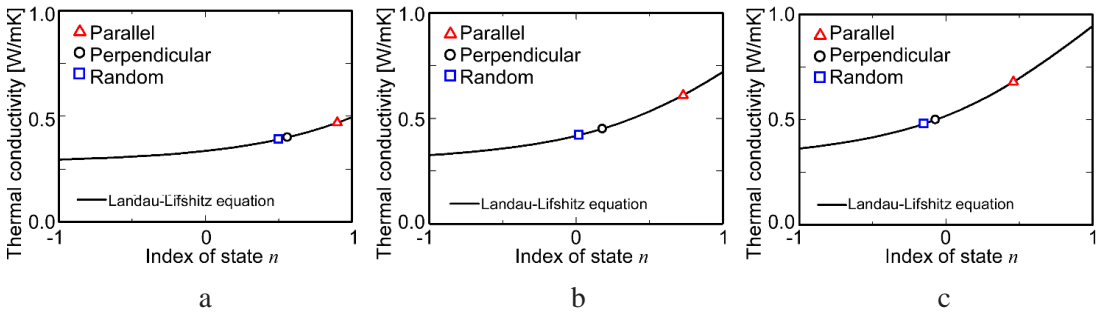


Figure 10 Thermal conductivity vs. index of state n . The volume fraction of Sendust particles is (a) 5 vol.%, (b) 10 vol.% and (c) 15 vol.%.

3.2 Magnetic properties

The experimental results of magnetic properties measured using the BH-analyzer are shown in Table 1, Table 2, Table 3 and Fig.10. From Table 1, the saturation magnetization increases with the increase in the volume fraction of Sendust particles and is almost independent of the structure formed by the particles inside the MRE. On the other hand, the relative initial permeability is larger in the parallel case than in the random and perpendicular cases. This is attributed to the fact that the easy axis of magnetization of most of the particles was oriented in the direction of the applied magnetic field, since the particles were cured while applying a uniform magnetic field during the specimen preparation process. From Table 3, the coercive force is smaller in the parallel case than in the random and perpendicular cases. Figure 11 shows the magnetization vs. magnetic field curves of the MRE with dispersed Sendust particles. From Fig.11, it can be seen that the initial magnetic permeability in the parallel case is larger than that in the Random and Perpendicular cases, regardless of the volume fraction of particles.

Table 1 Saturation magnetization of the MRE containing dispersed Sendust particles [mT]

Volume fraction	Random	Perpendicular	Parallel
5 vol.%	66.7	64.5	65.0
7.5 vol.%	94.4	89.9	91.1
10 vol.%	117.2	115.6	120.2

Table 2 Relative initial magnetic permeability of the MRE containing dispersed Sendust particles [-]

Volume fraction	Random	Perpendicular	Parallel
5 vol.%	1.30	1.21	1.99
7.5 vol.%	1.40	1.35	2.53
10 vol.%	1.66	1.52	3.30

Table 3 Coercive force of the MRE containing dispersed Sendust particles [kA/m]

Volume fraction	Random	Perpendicular	Parallel
5 vol.%	0.559	0.681	0.253
7.5 vol.%	0.446	0.338	0.238
10 vol.%	0.333	0.660	0.252

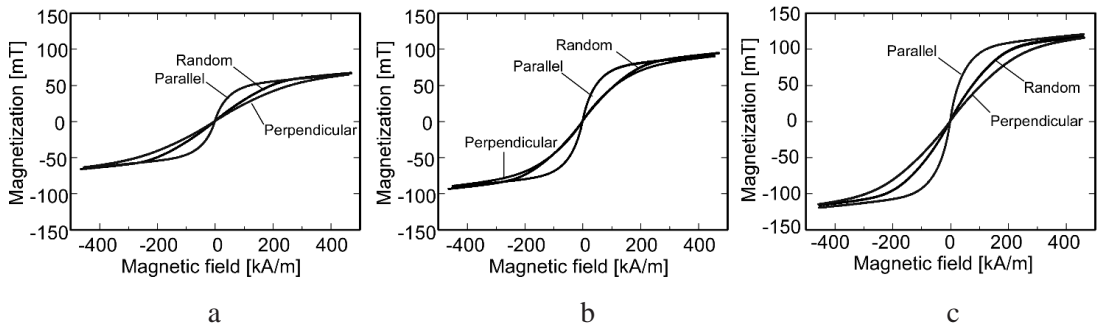


Figure 11 Magnetic field vs. magnetization curves. The volume fraction of Sendust particles is (a) 5 vol.%, (b) 7.5 vol.% and (c) 10 vol.%.

4. Conclusion

The magnetorheological elastomers containing Sendust particles were developed and the thermal and magnetic properties of them were investigated experimentally. The thermal conductivity of the MRE with dispersed Sendust particles was examined using the steady state parallel plate method. The thermal conductivity of the MRE with dispersed Sendust particles increases with the increase of the volume fraction of particles. The MRE containing chain-like clusters of Sendust particles has thermal conductivity anisotropy. The maximum thermal conductivity enhancement in the direction of cluster formation obtained in this experiment is 1.43 times that of MRE with randomly dispersed particles, and the minimum thermal conductivity enhancement in the direction orthogonal to the direction of cluster formation is 0.90 times. Magnetic properties of the MRE with dispersed Sendust particles were investigated using the BH analyzer. The MRE with dispersed Sendust particles has small coercive force and hysteresis loss and has the characteristics of a soft magnetic material.

As the volume fraction of Sendust particles in the MRE increases, the saturation magnetization and thermal conductivity also increase, however, the thermal conductivity anisotropy is less likely to vary greatly with direction. In the future, it is necessary to clarify the optimum particle volume fraction for thermal conductivity anisotropy, thermal conductivity and magnetic properties.

References

1. Jolly, M.R., Carlson, J.D., Munoz, B.C., "A model of the behaviour of magnetorheological materials", *Smart Materials and Structures*, 1996, 5, 607-614.
2. Carlson, J.D., Jolly, M.R., "MR fluid, foam and elastomer devices", *Mechatronics*, 2000, 10, 555-569.
3. Rabinow, J., Magnetic fluid torque and force transmitting device, US Patent US2575360, 1947.
4. Rabinow, J., The Magnetic Fluid Clutch, *Transactions of the American Institute of Electrical Engineers*, 1948, 67, 1308-1315.
5. Ginder, J.M. and Davis, L.C., Shear stresses in magnetorheological fluids: role of magnetic saturation, *Applied Physics Letters*, 1994, 65, 3410-3412.

6. Zhou, G.Y. and Zhang, P.Q., Investigation of the dynamic mechanical behavior of the double-barreled configuration in a magnetorheological fluid damper, *Smart Materials and Structures*, 2002, 11, 230-238.
7. Shiga, T., Okada, A. and Kurauchi, T., Magnetoviscoelastic Behavior of Composite Gels, *Journal of Applied Polymer Science*, 1995, 58, 787-792.
8. Jolly, M.R., Carlson, J.D., Munoz, B.C. and Bullions, T.A., The Magnetoviscoelastic Response of Elastomers Composites Consisting of Ferrous Particles embedded in a Polymer Matrix, *Journal of Intelligent Material Systems and Structures*, 1996, 7, 613-622.
9. Mikhailov, V.P. and Bazinenkov, A.M., Active vibration isolation platform on base of magnetorheological elastomers, *Journal of Magnetism and Magnetic Materials*, 2017, 431, 266-268.
10. Bastola, A.K. and Li, L., A new type of vibration isolator based on magnetorheological elastomer, *Materials and Design*, 2018, 157, 431-436.
11. Masumoto, H., Yamamoto, T., On a New Alloy "Sendust" and its Magnetic and Electric Properties, *Journal of the Japan Institute of Metals and Materials*, 1937, 1, 127-135 (in Japanese).
12. Aikawa, Y. and Kato, N., Effect of Composition on Core Loss of Fe-Si-Al Powder Compressed Cores, *Sanyo Technical Report*, 2000, 7, 29-34 (in Japanese).
13. Tsutaoka, T., Ono, T. and Tsurunaga, A., High Frequency Permeability of Fe-Al-Si Granular Composite Materials, *IEEE International Symposium on Electromagnetic Compatibility*, Long Beach, USA, 2011, 78-83.
14. Choi, D.S., Yoo, G.S. and Kim, D.I., Analysis on the Characteristics of the EM Wave Absorber Using Sendust and Amorphous Metal Powder, *2009 Asia Pacific Microwave Conference*, Singapore, 2009, 716-719.
15. Liao, H. and Chen, J-F., Design Process of High-frequency Inductor with Multiple Air-gap in the Dimensional Limitation, *Journal of Engineering*, 2022, 16-33.
16. Miwa, Y. and Shimizu, T., Loss Comparison of Core Materials Used for the Inductor of a Buck-chopper Circuit, *2015 IEEE Energy Conversion Congress and Exposition*, Montreal, Canada, 2015, 5279-5286.
17. Ido, Y. and Inagaki, T., Numerical Simulations of Structure Formation of Magnetic Particles and Nonmagnetic Particles in MAGIC Fluid Under Steady Magnetic Field, *Complex Systems*, AIP Conference Proceedings, 2008, 982, 598-605.
18. Ido, Y., Inagaki, T. and Umehara, N., Numerical Simulations of Distributions of Magnetic and Nonmagnetic Particles in MAGIC Fluids, *Mangetohydrodynamics*, 2008, 8, 147-152.
19. Ido, Y., Iwamoto, Y. and Kondoh, S., Thermal Conduction of the Magnetic Fluids Mixing Micrometer Size Particles, *Journal of Magnetism and Magnetic Materials*, 2020, 508, 166864.
20. Warriar, P. and Teja, A., Effect of particle size on the thermal conductivity of nanofluids containing metallic nanoparticles, *Nanoscale Research Letters*, 2011, 6, 247-252.