Soft Actuator with DN-gel Dispersed with Magnetic Particles

Shutaro Shigetomi¹ and Fujio Tsumori^{2*}

¹ Department of Mechanical Engineering, Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka 819-0395, Japan

² Department of Aeronautics and Astronautics, Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka 819-0395, Japan
*tsumori@aero.kyushu-u.ac.jp

We used double-network-gel (DN-gel) to develop a new magnetic gel. Agar and Polyacrylamide gels were selected to prepare Agar/PAAm DN-gel. We dispersed magnetic particles in this gel to fabricate a soft actuator or soft robot. Our modified DN-gel, Agar/PAAm, was suitable for this purpose. During the preparation, we employed thermal initiator instead of UV-initiator, which is popular for DN-gel, for crosslinking. The material contains magnetic particles so that it is difficult to use UV-light. The particles prevent from transmit the light. In this process, we employed thermal initiator so that UV-light is unnecessary. We also propose a micro patterning system of magnetic properties using laser scanning. CO₂ laser was employed for local heating, which can make reversible transformation of gel-liquid states. Finally, we demonstrated a simple strip soft actuator using the proposed method with DN-gel.

Keywords: Soft actuator, Double-network gel, Agar, Magnetic particles, Magnetic orientation

1. Introduction

Recently, double-network gels (DN-gel) have been attracting attention as a tough gel material. This gel was developed by Gong et al. and has the characteristics of compression strength of 20 MPa and elongation at break of more than 80 % [1]. Figure 1 shows an example of DN-gel sample we tried. It is found that the gel was extremely tough. In this study, we aim to develop a soft actuator by dispersing magnetic powder particles in this gel material.

The magnetic soft actuators that have been reported so far mainly use silicone rubber as the base material, and magnetic particles are dispersed in it. The authors have been studying magnetic soft actuators using this kind of silicone rubber, focusing on biomimetic structures such as artificial cilia [2–12] and worm-like robots [13, 14]. Silicone rubber material is a viscous liquid material before curing. We can make magnetic rubber by mixing magnetic powder material with it. Also, it is possible to

Fig. 1. Photo of DN-gel sample (PAMPS/PAAM gel), which has high resistance from cutting.

prepare a magnetic rubber material with magnetic anisotropy by curing it while applying a magnetic field. Silicone is easy to handle and is widely used in this field because it can be prepared as magnetic rubber by the method described above.

On the other hand, in recent years, gel materials have been focused as flexible materials as

well [15-18]. A gel material is a flexible material that contains a liquid such as water in its polymer network structure. Among them, DN-Gel is a promising material due to its toughness. DN-gel has a structure in which a network of two gel materials overlap at the molecular level. When a large deformation occurs, the first gel network collapses first. On the other hand, the 2nd gel network continues to bridge the 1st gel network, and at that time, the 2nd gel molecular chain slides to relieve the concentration of stress. This is the key mechanism that generates toughness.

The most popular DN-gel material is made of 2 kinds of polymer materials, the 1st gel is poly (2-acrylamido-2-methylpropane-sulfonic acid) gel and poly-acrylamide gel. It is called PAMPS/PAAm gel. This material was developed by Gong et al. In their recipe, UV crosslinking initiator is used and gelation is performed by UV light. The 1st gel material is gelated and then immersed in the 2nd gel monomer solution. After that, the swollen material is irradiated with UV light, and the 2nd gel is gelated to obtain DN-gel.

In this paper, authors propose to use DN-gel as a matrix of a magnetic soft actuator. In order to use DN-gel for this purpose, Gong's PAMPS/PAAm had some problems. Therefore, we chose Agar/PAAm gel system [19], which can be controlled by thermal crosslinking. The reason will be described in the following experimental section.

We also propose a new micro-patterning method of magnetic orientation in this paper. The agar plays an important role for this process, which will be also explained later. This method would be used for a new method for a 4D-printing system of magnetic materials [16-18, 20].

2. Experimental

2.1. DN-gel preparation

We prepared 2 kinds of materials for base gels, agar (Kantenpapa, Ina Food Industry) and acrylamide monomer (AAM). The flow of the preparation of the DN-gel is shown in Fig. 2. At first, 2 mass% of aqueous solution of agar was prepared by heating, and magnetic powder (carbonyl iron OM, BASF) with a mean diameter of 4.3 μm was mixed in it (Fig. 2a). The mixture was put in a plastic case and it was cooled in a refrigerator at 4 $^{\circ}\text{C}$ for 1.8 ks (0.5 h). Agar gel dispersed with magnetic particles was obtained (Figs. 2b, c). Next, the agar gel was dipped into AAm solution for 10.8 ks (3h). The AAm solution was added 0.03 mol% of methylene-bisacrylamide (MBAA) as crosslinker, 1 mol% of ammonium peroxodisulfate (APS) as thermal initiator, and 0.25 mol% of

Fig. 2. Flow of Agar/PMMA DN-gel preparation.

Fig. 3. Prepared Agar/PAAm gel sample; (upper) after knife edge cutting test, and (lower) the bent sample after the test.

tetramethylethylenediamine (TEMED) as hardening accelerator agent (Fig. 2d). The gel sample was heated at 50 $^{\circ}\text{C}$ for 3.6 ks (1h) to crosslink AAm, and finally Agar/PAAm DN-gel was obtained (Figs. 2e, f). The material contains magnetic particles so that it is difficult to use UV-light. The particles prevent from transmit the light. In this process, we employed thermal initiator so that UV-light is unnecessary.

Figure 3 shows an example of an obtained DN-gel sample. We checked the toughness by a preliminary cutting resistance test. The sample was hard to cut by a knife edge, and hard to fracture even

in a severe bending state after testing.

2.2. Tensile test

Tensile test was performed in accordance with JIS6251K. We used a testing machine (EZ-Test, Shimadzu Cooperation) for the test. We prepared 4 kinds of Agar/PAAm DN-gels which contained 0, 5, 10, and 20 mass% of carbonyl iron powder. Test specimens of the 1st and 2nd gels, which were agar gel and PAAm gels, were also prepared. For all samples, stress-strain relations were obtained. Tensile speed was set at 10 mm/min for each sample.

2.3 Micro-patterning of magnetic orientation

Magnetic particles generate chain-like cluster structures which align to be parallel to an applied magnetic field. We can fix the cluster structures in a gel matrix to set an anisotropic magnetic orientation in the material. We propose a new method to pattern the magnetic orientation in a DN-gel sheet.

Figure 4(a) shows a schematic illustration of this process. A sheet of agar gel, which is just before immersion of AAm, is put under an applied magnetic field. Laser is scanned on the agar sheet to heat and melt the agar sheet. By laser heating, the agar is molten so that the magnetic particles can move freely and generate chain clusters as to align the applied magnetic field. The cluster structures are fixed by re-gelation after cooling in the air. Agar is

Fig. 4. (a) Schematic illustration of proposed patterning system of magnetic orientation, and (b) Sample photo of chain-like clusters. The highlighted area shows laser-scanned line, where horizontal chain-like clusters of particles were generated.

useful for the present method as this reversible gel-liquid change by temperature. During the laser processing, a thin glass sheet was put on the agar sheet not to dry it. Figure 4(b) shows obtained micro chain clusters. The obtained agar sheet will be processed as shown in Figs. 2(d-f).

3. Results and discussion

3.1. Tensile tests

Stress-strain curves are shown in Fig. 5. The elongation of the 1st gel, agar, was much smaller than any other samples. The 2nd gel showed high elongation, while the tensile strength was less than 0.1 MPa. The DN-gel sample based on these 2 gels improved much better in the tensile properties. The elongation was more than 1000 % and the tensile strength was about 0.6 MPa.

As increasing iron powder contents, tensile properties decreased, however the tensile strength of each sample was larger than that of the 1st or 2nd gel. The particles might be origins of fractures in this case. The properties could be improved by surface treatment of powder particles, which would cause high bonding effect between particles surface and gel molecules.

Fig. 5 Stress-strain relationships of DN-gel samples. The 1st and 2nd gels data were also plotted.

3.2. Micro-patterning and actuation

We prepared a sample strip as shown in Fig. 6. We scanned lasers in parallel lines changing the direction of an applied magnetic field. After cutting out, a thinner strip sample was obtained. Figure 7 shows an optical micro photo of the sample at the area as shown in Fig. 6(a). Chain clusters were generated in the scanned area.

The cut sample strip was set under a vertical applied magnetic field of 0.6 T. The deformed sample is shown in Fig. 8. Different rotational

Fig. 6. Schematic of sample preparation; (a) Laser scanning pattern and applied magnetic fields, and (b) dimension of cut sample.

Fig. 7. Optical micro-photo image of laser scanned sample. The observed area is shown in Fig. 6(a) as a square with dashed line.

moments exerted in the 3 areas caused wavy shape change. Patterning of magnetic property was successfully applied by our proposed method.

4. Conclusion

We proposed to use DN-gel for magnetic soft actuators. Agar and PAAM gels were combined as DN-gel material for this purpose. The obtained material showed excellent tensile properties. Micro patterning system using laser scanning was also proposed for further application of 3D or 4D printing. We demonstrated a simple strip sample, which could actuate by an applied magnetic field.

The method would be useful to develop soft robots which is bio-compatible or bio-degradable. Micro- or nano- patterning on this kind of soft material on the surface will be our future work [21, 22], which would improve the locomotion ability of soft robots.

Fig. 8. DN-gel strip sample with patterned magnetic orientation. Wavy deformation was observed under a vertical magnetic field.

References

1. J. P. Gong, Y. Katsuyama, T. Kurokawa, and Y. Osada, *Adv. Mater.*, **15** (2003), 1155.
2. F. Tsumori, N. Miyano, and H. Kotera, *J. Jpn. Soc. Powder Powder Met.*, **56-3** (2009) 133.
3. F. Tsumori, N. Miyano, and H. Kotera, *J. Jpn. Soc. Powder Powder Met.*, **56-3** (2009) 127.
4. F. Tsumori and J. Brunne, *Proc. IEEE MEMS*, (2011) 1245.
5. K. Hatama, F. Tsumori, Y. Xu, H. Kang, T. Osada, and H. Miura, *Jpn. J. Appl. Phys.*, **51** (2012) 06FL14.
6. F. Tsumori, K. Hatama, H. Kang, T. Osada, and Hideshi Miura, *Proc. IEEE-NEMS*, (2013) 845.
7. F. Tsumori, A. Saijou, T. Osada, and H. Miura, *Jpn. J. Appl. Phys.*, **54** (2015), 06FP12.
8. F. Tsumori, R. Marume, A. Saijou, K. Kudo, T. Osada, and H. Miura, *Jpn. J. Appl. Phys.*, **55** (2016), 06GP19.
9. R. Marume, F. Tsumori, K. Kudo, T. Osada, and K. Shinagawa, *Jpn. J. Appl. Phys.*, **56** (2017) 06GN15.
10. H. Shinoda, S. Azukizawa, and F. Tsumori, *Proc. MicroTAS*, (2018) 679.
11. H. Shinoda and F. Tsumori, *Proc. IEEE MEMS*, (2020) 497.
12. S. Gaysornkaew and F. Tsumori, *Jpn. J. Appl. Phys.*, **60** (2021), SCCL02.
13. K. Maeda, H. Shinoda, and F. Tsumori, *Jpn. J. Appl. Phys.*, **59** (2020), SIIL04.
14. H. Shinoda, S. Azukizawa, K. Maeda, and F. Tsumori, *J. Electrochem. Soc.*, **166** (2019)

- B3235.
15. S. Shigetomi, H. Takahashi, and F. Tsumori, *Jpn. J. Appl. Phys.*, **33** (2020), 193.
 16. S. Azukizawa, H. Shinoda, K. Tokumaru, and F. Tsumori, *J. Photopolymer Sci. Technol.*, **31-1** (2018) 139.
 17. H. Shinoda, S. Azukizawa, K. Maeda, and F. Tsumori, *ECS Trans.*, **88-1** (2018) 89.
 18. S. Azukizawa, H. Shinoda, and F. Tsumori, *Proc. IEEE MEMS*, (2019) 248.
 19. D. Wei, J. Yang, L. Zhu, F. Chen, Z. Tang, G. Qin, and Q. Chen, *Polymer Testing*, **69** (2018) 167.
 20. F. Tsumori, H. Kawanishi, K. Kudo, T. Osada, and H. Miura, *Jpn. J. Appl. Phys.*, **55-6S1** (2016), 06GP18.
 21. Y. Xu, F. Tsumori, H.G. Kang, and H. Miura, *Adv. Sci. Lett.*, **12-1** (2012), 170.
 22. T. Miyata, K. Tokumaru, and F. Tsumori, *Jpn. J. Appl. Phys.*, **59** (2020), S11J07.