Analysis for the Elongational Motion of PVA Magnetic Gels Under Gradient Magnetic Fields

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We have investigated the elongational motion of magnetic gels under gradient magnetic fields. The magnetic gel was made from fine particles of iron oxide and a network of poly(vinyl alcohol) crosslinked by glutaraldehyde. The elongation increased with increasing the magnetic field strength up to 1200 mT. The elongation increased as increasing the concentration of magnetic particles; oppositely, it decreased as increasing the crosslinking density. It was found that the elongation can be explained by a classical theory of magnetism that the elongation is proportional to the product of magnetization and the field gradient. The effect of load on the elongation has been also investigated. The coefficient of the conversion from magnetic energy to mechanical one was estimated to be 10^{-4} %, which is nearly independent of the load.

Key words: gels, magnetic gel, actuator, manipulator

1. INTRODUCTION

A variety of actuators using polymer gels has been widely investigated. Ionic conductive polymer gels such as polyelectrolyte gels undergo bending motion under ac electric fields. Actuators using the bending motion of a polyelectrolyte gel were reported, and they were called gel-looper [1] and gel-eel [2]. Amphiphilic polymer gel swollen by an organic solvent shows fast rotation by spreading organic solvent [3]. Conductive polymers also have a potential to be an actuator. Polypyrrole films undergo rapid and intensive bending in the solid state induced by the reversible and anisotropic adsorption [4]. Photo reactive polymers show a deformation by UV light. Liquid crystalline gel films containing freestanding azobenzene undergo a significant and anisotropic bending toward the irradiation direction due to the isomerization [5].

Magnetic gels are well known for field-responsive gels and they have a great potential for actuators. The magnetic gel consisted of poly(vinyl alcohol) and



Fig. 1. Photographs of the elongational motion of magnetic gels at B=0 (top) and B=1T (bottom).

magnetic fluids elongates under non-uniform magnetic fields [6,7]. Elastic modulus of magnetic gels such as PVA-magnetic fluids [8], PVA-barium ferrite [9] and κ -carrageenan-barium ferrite [10, 11] gels changes due to magnetization. It was demonstrated in a micro tube that a fluid valve [12] and a flexible fluid pump [13, 14] made of magnetic gels were actuated by magnetic fields. These studies revealed that the magnetic gels could respond faster and deform larger compared to the other stimuli-responsive gels.

The elongational motion of magnetic gel has been intensively investigated by Zrinyi [6]. According to his research, the elongation with respect to an original length reached a maximum value of 0.4. Not only elongation but also force of approximately 100 mN was induced. The work by elongation was less than 7 mJ.

We tried to improve the maximum stress and examine the application of the manipulator for a practical use. In this paper, the elongation of magnetic gel with various concentrations of magnetic particles and crosslinking



Fig. 2. Elongation vs. magnetic field for magnetic gels with various ferrite concentrations; crosslinking density: 1.0 mol%.



Fig. 3. Elongation vs. magnetic field for magnetic gels with various crosslinking densities; ferrite concentration: 10 wt.%.

density was presented. The effect of load on the elongation was also discussed aiming at practical actuators.

2. EXPERIMENTAL PROCEDURES

The magnetic gel is made of a finely dispersed magnetic particles and poly(vinyl alcohol) (PVA). A 4 wt.% of PVA aqueous solution and iron oxides Fe_3O_4 (Wako Chemicals) were mixed at 80 °C. The mean diameter of magnetic particles was determined as 15 μ m. A 25 vol.% of glutaraldehyde aqueous solution was used as a crosslinking agent. Crosslinking reaction was taken place by adding hydrochloric acid. Crosslinking density was defined by the molar fraction of glutaraldehyde to PVA in preparation. Crosslinking density was changed from 0.5 to 5 mol%. The aqueous solution of PVA mixed with iron oxides was poured into a mold with a shape of 14 mm in diameter and 10 mm in height. The weight fraction of iron oxides was varied from 5 to 25 wt.%.

Dynamic viscoelastic measurements were carried out using a rheometer (Dve-V4, Rheology). Oscillatory compressional shear was applied to magnetic gels and the stress response was measured; hence elastic modulus obtained stands for Young's modulus. The frequency was kept as 1 Hz. The offset strain with respect to the sample thickness was kept as 3%. The measurement was performed at room temperature. The shape of samples for the mechanical measurement was cylinder with a shape of 14 mm in diameter and 10 mm in height. Each modulus was determined from an average of three samples.

3. RESULTS AND DISCUSSIONS

Fig. 1 shows the photographs representing the elongation of magnetic gels under gradient magnetic fields. Fig. 1 (top photo) demonstrates the gel before elongation. The top of the gel is the position which the magnetic field gradient is the maximum. This means the maximum magnetic force is induced by the upper part of the gel. The elongation when a magnetic field of 1000 mT was applied is shown in Fig. 1 (bottom photo). The gel was drawn toward the center of the magnetic field.

The elongation of magnetic gel with various concentrations of magnetic particles is presented in Fig.



Fig. 4. Elongation vs. magnetic field gradient for magnetic gels with various crosslinking densities; ferrite concentration: 10 wt.%.

2. The elongation L/L_0 stands for the length of gel with respect to the original length L_0 . The value of L/L_0 increased with increasing the magnetic field strength. The elongation for the gel with a particle concentration of 5 wt.% was in a low level. Oppositely, the gels with particle concentrations more than 10 wt.% showed high elongation approximately 40 %. This value coincides with the value of PVA gel containing magnetic fluid reported by Zrinyi et al. [6].

Fig. 3 displays the value of L/L_0 for the gels with various crosslinking densities. As well as the result of Fig. 2, the elongation increased with increasing the field strength. It was found that the value of L/L_0 decreased as increasing the crosslinking density. This indicates that the gels with high crosslinking density are difficult to elongate even in a strong magnetic field.

Fig. 4 shows the elongation $\Delta L (=L - L_0)$ as a function of the gradient of magnetic field along z axis $\partial B/\partial z$. It was seen that the ΔL was nearly proportional to the $\partial B/\partial z$. We made a model for the elongation induced by the field gradient, which is based on the magnetism. The magnetic force acting on the gel f_{mag} can be described by $f_{mag} \propto M(\partial B/\partial z)$, where *M* is the volume magnetization of the gel. The recovery force by the elongation should be balanced with this magnetic force when in an equilibrium state. Accordingly, the following equation was obtained

$$L = \frac{ML_0}{2E} \frac{\partial B}{\partial z} \qquad (1)$$

, where *E* appears Young's modulus of the gel. The above equation indicates the elongation is proportional to the magnetic field gradient $\partial B/\partial z$. As can be seen in Fig. 4 the elongation under high magnetic fields was in proportion to the magnetic field gradient. In a low magnetic field, the value of elongation was deviated from the line of $L \propto \partial B/\partial z$. This may be understood by the magnetization curve of the gel containing Fe₃O₄ particles. It is because that the value of the magnetization under low magnetic fields is still low. Plots of $ML_0/2$ vs. *E* revealed the value of $ML_0/2$ is inversely proportional to the Young's modulus *E* of gels;

therefore the validity of Eq. (1) was proved. This strongly suggests that the elongation takes place by the magnetic interaction between magnetization of gels and the magnetic field gradient.

The effect of load on the elongation was also investigated in order to evaluate the ability of this manipulator. Although it is quite natural, the elongation largely decreased as increasing the load on magnetic gel. The maximum stress induced by the magnetic field was found to be 130 g/cm² (~13 kPa), which is far larger than the value of 31 g/cm² as reported in the literature [6]. The gel which was shown in the literature is PVA gel containing magnetic fluids. Therefore, the magnetization of the gel is considered to be weak compared to the gel we investigated. This must bring the lowness of maximum stress generating by magnetic fields. The gel presented here underwent an elongation of approximately 10%, holding this maximum stress. This implies the magnetic gel has a great potential as practical manipulators which can be driven by magnetic field.

4. CONCLUSION

A magnetic gel consisting of poly(vinyl alcohol) and iron oxides was actuated by a gradient magnetic field. The gel demonstrated elongational motions when the magnetic field was applied. The elongation increased as increasing the concentration of magnetic particles; oppositely, it decreased as increasing the crosslinking density. It was cleared that the elongation can be explained by a classical theory of magnetism that the elongation is proportional to the product of magnetization and magnetic field gradient. The effect of load on the elongation was also investigated in order to evaluate the ability of gel-manipulators. The maximum stress induced by the magnetic field was found to be ~13 kPa. The gel underwent an elongation of approximately 10%, holding this maximum stress. The coefficient of the conversion from magnetic energy to mechanical one was estimated to be 10^{-4} %, which is nearly independent of the load. The energy conversion was still low; however if a manipulator is made combining some gels, the magnetic gel would be applicable as a practical manipulator.

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References

- [1] Y. Osada, H. Okuzaki, and H. Hori, Nature, 355, 242-44 (1992).
- [2] Y. Ueoka, J. P. Gong, and Y. Osada, J. Intelligent Material Systems Structures, 8, 465-71 (1997).
- [3] T. Mitsumata, K. Ikeda, J. P. Gong, and Y. Osada, *App. Phys. Lett.*, **73**, 2366-68 (1998).
- [4] H. Okuzaki, and T. Kunugi, J. Polym. Sci., Polym. Phys., 34, 1747-49 (1996).
- [5] T. Ikeda, M. Nakano, Y. Yu, O. Tsutsumi, and A. Kanazawa, *Adv. Mater.*, **15**, 201 (2003).
- [6] M. Zrinyi, L. Barsi, and A. Buki, Polym. Gels and Networks, 5, 415-27 (1997).
- [7] D. Szabo, G. Szeghy, and M. Zrinyi, *Macromolecules*, 31, 6541-48 (1998).
- [8] T. Mitsumata, K. Ikeda, J. P. Gong, Y. Osada, D. Szabo, and M. Zrinyi, J. App. Phys., 85, 8451-55 (1999).
- [9] T. Mitsumata, E. Juliac, K. Furukawa, K. Iwakura, T. Taniguchi, and K. Koyama, *Macromol. Rapid Commun.*, 23, 175-78 (2002).
- [10] T. Mitsumata, A. Nagata, K. Sakai, and J. Takimoto, *Macromol. Rapid Commun.*, 26, 1538-41 (2005).
- [11] T. Mitsumata, K. Sakai, and J. Takimoto, J. Phys. Chem. 110, 20217-23 (2006).
- [12] T. Mitsumata, Y. Horikoshi, and J. Takimoto, *Trans. Mater. Res. Soc. Jpn.*, **31(3)**, 807-10 (2006).
- [13] Y. Horikoshi, T. Mitsumata, and J. Takimoto, *Trans. Mater. Res. Soc. Jpn.*, **32(3)**, 843-44 (2007).
- [14] T. Mitsumata, Y. Horikoshi, and J. Takimoto, *e-Polymers*, no. 147, 1-10 (2007).

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