

# Electrostriction of Highly Swollen Polymer Gel: Possible Application for Gel Actuator

TOSHIHIRO HIRAI,<sup>1,\*</sup> HIROSHI NEMOTO,<sup>1</sup> MITSUHIRO HIRAI,<sup>2</sup> and SADA O HAYASHI<sup>1</sup>

<sup>1</sup>Faculty of Textile Science and Technology, Shinshu University, 3-15-1 Tokida, Ueda-shi 386, Japan, and <sup>2</sup>Faculty of Liberal Art & Science, Gunma University, 4-2 Aramaki-cho, Maebashi-shi 371, Japan

## SYNOPSIS

A gel of poly(vinyl alcohol) highly swollen with dimethyl sulfoxide was demonstrated to actuate at a much faster rate (more than 1000 times) than those reported on conventional gel actuators in air and to flap a wing of 12.5 cm at a rate of 2 Hz with a span of 10 cm by periodically applying electric field. The action (8% in length) induced by the electric field reached several hundreds times larger than those reported on the conventional ferroelectric solid materials. The electrostrictive gel was, thus, found to be a promising candidate material as a novel type of actuator or artificial muscle by having overcome some difficulties in responding time, electrochemical reactions on the electrodes, and magnitude of action in conventional methods or materials. © 1994 John Wiley & Sons, Inc.

## INTRODUCTION

Polymer gels have been attracting many attention as novel types of materials, particularly as so-called intelligent materials, such as biocompatible materials, materials for drug delivery systems, artificial muscle or actuator, and so on.<sup>1,2</sup> From the standpoint of actuator or artificial muscle, polyelectrolyte gels have been mentioned and investigated intensively because the gels can easily be actuated by applying relatively low electric field, though the rate of action has been recognized far below satisfaction as actuator. Very recently, Osada et al.<sup>3</sup> found that the action rate could be improved remarkably by adding surfactant to the solution in which the polyelectrolyte gel was immersed. In the system in which a polyelectrolyte gel is immersed in an electrolyte solution, however, electrolysis is inevitable and gas generation from the electrode is a serious problem in practical application. On the other hand, nonionic gels have been considered to be actuated only by solvent exchange from a good solvent to a poor one or vice versa.<sup>1,4,5</sup> Apparently, solvent exchange is not preferable as a trigger for quick action. Thermosensitive

gels that can swell or deswell by temperature change have also been found recently and have been investigated as actuators.<sup>1,6</sup> In this case also, the temperature jump that induces the actuation of the gel is not easy enough for quick action. Photo-induced actuation of the gel has also been investigated on a gel immersed in saline solutions and has been shown to be an efficient actuation in combination with electrical field application.<sup>7</sup>

We have searched for a more preferable and completely different method for gel actuation than these conventional methods, and report on the electrostrictive method in this study.<sup>8</sup>

## EXPERIMENTAL SECTION

### Materials

The gel was prepared by the following procedures. Poly(vinyl alcohol) (PVA) was Kuraray 117 whose degree of saponification was 98% and degree of polymerization was 1700. The PVA was saponified again at 50°C for 2 h in 13M NaOH aqueous—methanol solution (methanol content was 95% in volume). The saponified PVA flakes were dissolved in hot water and precipitated in methanol. Repre-

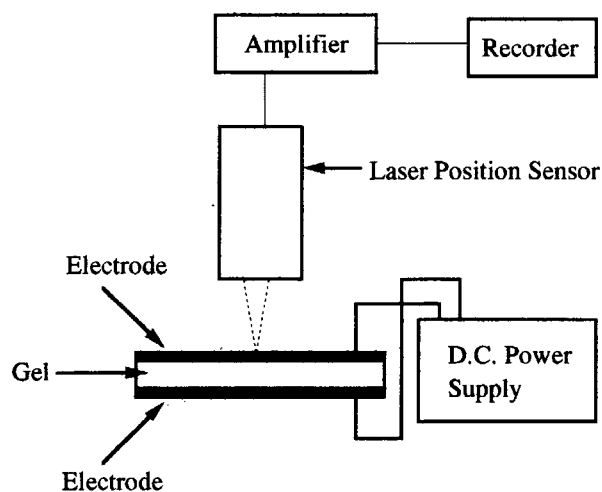
\* To whom correspondence should be addressed.

precipitation was repeated three times. Residual acetate group was not detected in IR spectra. The PVA was dissolved in aqueous solution of dimethyl sulfoxide (DMSO) at 90°C. The composition of the PVA-DMSO-water solution was 10 wt % in PVA, 72 wt % in DMSO, and 18 wt % in water. The mixture was kept at -20°C for 23 h and was left at room temperature for 1 h. This cooling treatment was repeated six times. Thus prepared transparent PVA gel has the network structure made up only by physical crosslinks (like hydrogen bonds). The gel was then covalently crosslinked by acetalization. First, the gel was immersed in 0.07 or 1% glutaraldehyde (GA) aqueous solution at 5°C for 12 h, and then was acetalized by raising the temperature of the solution to 30°C and by addition of hydrochloric acid. The concentration of the acid in the reaction media was 0.01M. After 1 h reaction time, the gel was rinsed with large amounts of water, immersed in hot water to destroy the physical crosslinks in the gel, and then thoroughly deswollen in acetone, finally re-swollen and stored in DMSO until served for measurements. Thus prepared PVA-DMSO gel was composed of 98% DMSO and 2% PVA.

### Measurements

The action of the gel was triggered by applying d.c. voltage up to 1 kV and detected with a position sensor as shown in Figure 1. The electric field was generated and the current was measured by Power Supply type II-c of Toyo Kagaku Sangyo Co. Ltd.

The temperature change in the gel was monitored by inserting a thermocouple into the gel. The strain



**Figure 1** System used for measuring electrically induced strain of the polymer gels.

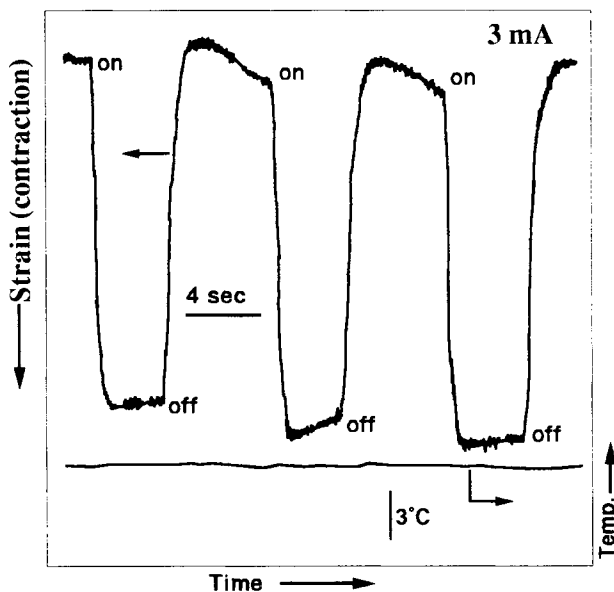
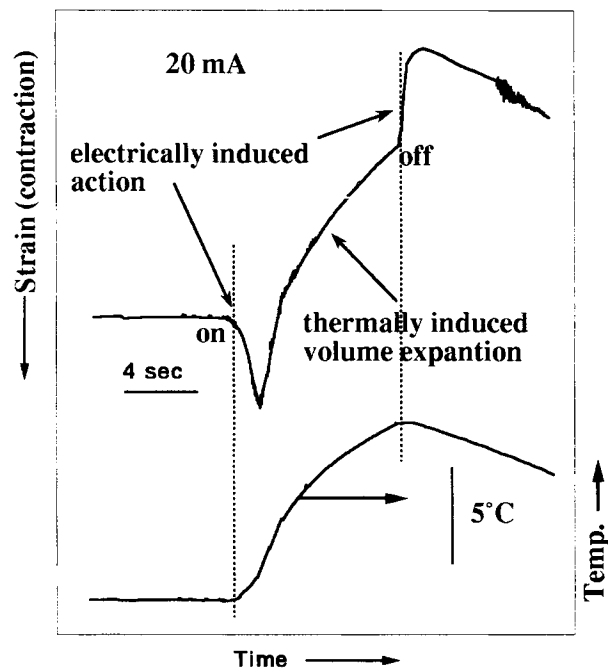
induced in the gage was amplified by Shinkoh Strain Amp. DS6/MTC. The change in the strain and temperature was recorded by multipen recorder, Hitachi 561.

### RESULTS AND DISCUSSION

PVA is known as a nonionic hydrophilic polymer. This polymer can also dissolve in DMSO, which is polar solvent. The DMSO-swollen gels appear to be superior to a hydrogel for electrical application since DMSO is hard to electrolyzed and relatively stable in electric field, while water is easily electrolyzed into gases of hydrogen and oxygen, which usually causes a serious problem in practical application for hydrogel actuator. In Figure 2, we show some results on the effect of the presence of trace amount of water on the actuation profile of the PVA-DMSO gel. When the gel contains small amounts of water, the gel contracted instantly on applying electric field followed by relatively slow expansion. On shut down of the field, the gel relaxed instantly followed by much slower shrinkage [Fig. 2(a)]. The observed current was 20 mA during the electric field application. Apparent temperature rise was also observed by applying the electric field. The action without water was shown in Figure 2(b). The current was 3 mA and temperature did not rise under the electric field application. These profile suggests that, in Figure 2(a), the slow action is induced by thermal expansion, and the fast one cannot be attributed to the joule heat generation. Slow thermal action was not observed and only the fast action was observed in Figure 2(b). It was also confirmed from the experimental data shown in Figure 3 that the joule heat generation was negligible when the current is small enough, and in other words, the action was not obscured by thermal expansion of the gel volume.

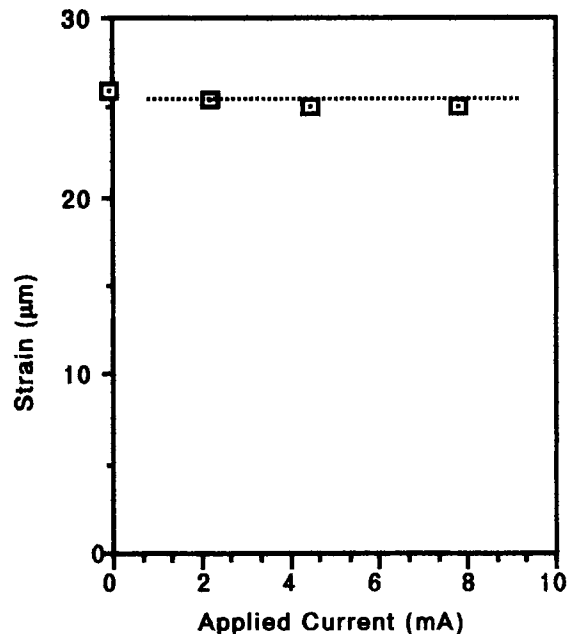
The action was asymmetric, that is, the gel contracted in the parallel direction to the electric field and expanded in the direction perpendicular to the electric field. The asymmetric action was also confirmed in the striction measurement as shown in Figure 4.

In Figure 5, the action or striction measurement was shown. The gel used in this case was crosslinked in 0.07% GA, which is a lower concentration than that of the gel shown in Figure 2. As can be seen in Figure 5(a) and (b), the action increased with electric field and the rate was very quick and more than 1000 times faster compared to that measured in a conventional solvent exchange method.<sup>4,5</sup> The magnitude of the action appears to be second order with



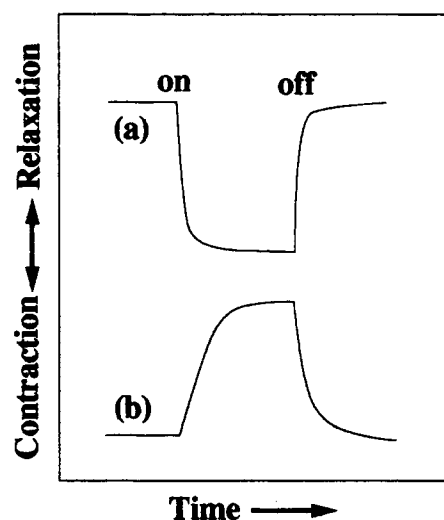
**Figure 2** Action of PVA-DMSO gel by electric field. In (a), addition of trace amount of water caused electrolysis and heat generation on the electrode, which caused heat expansion of the gel following electrostrictive action. While in (b), thermal expansion of the gel was not observed after pre-treatment in low-voltage d.c. field in order to remove electroconductive ionic species.

respect to the square of the value of the applied voltage, as shown in Figure 6, and was several hundreds times larger than those of conventionally used electrostriction materials.<sup>9</sup> The action was basically symmetrical against inversion of the polarity, al-

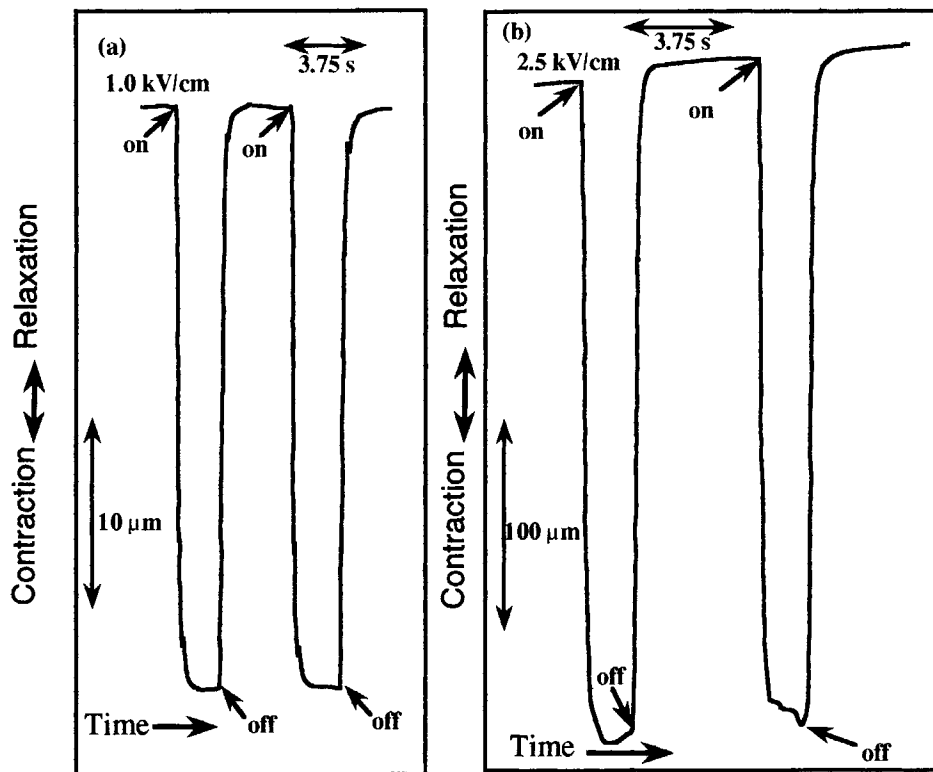


**Figure 3** Effect of electric current on the action of PVA-DMSO gel.

though the symmetry was not perfect due to a leak of solvent from the gel. However, the stricture action of the gel is reproducible and stable, as shown in Figure 5, when the duration of the measurement is short. Thus, in the experiment shown in Figure 5(b), the gel was exposed to air for more than 3 h.



**Figure 4** Effect of electric field on the shape change of PVA-DMSO gel. The gel contracts in the direction parallel to electric field [curve (a)] and expands in the direction perpendicular to the electric field [curve (b)], that is, the gel changes its shape asymmetrically against the applied electric field.

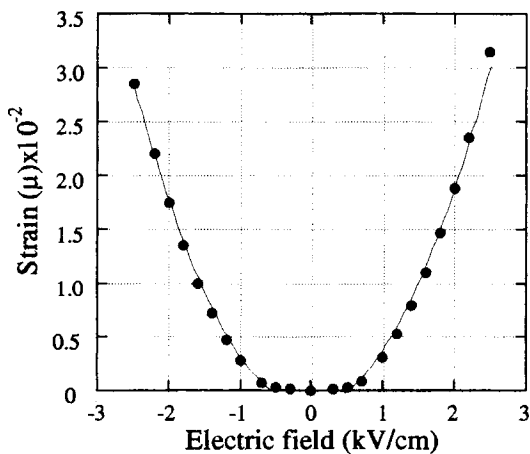


**Figure 5** Action recorded by laser stain sensor. Voltage applied was (a) 1 kV/cm and (b) 2.5 kV/cm. Completion of 90% action was attained within 100 ms, and 50% within 25 ms under the conditions 2.5 kV/cm and 0.1 mA. Temperature rise in the action was not detected by the thermocouple inserted in the gel. PVA-DMSO gel was chemically crosslinked in 0.07 wt % GA solution.

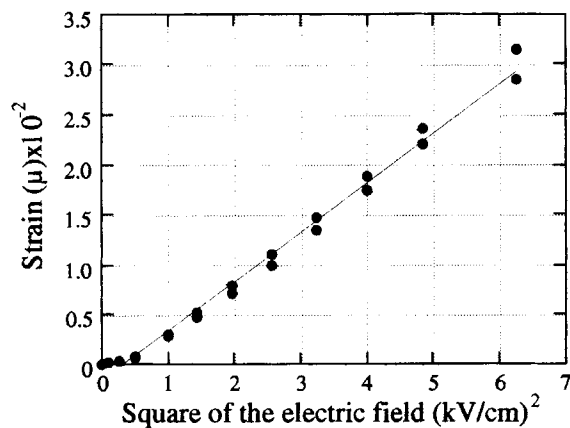
Conventional theory suggest that the electrically induced strain ( $X$ ) can be expressed by Eq. (1), using piezoelectric ( $dE$ ) and electrostrictive ( $ME^2$ ) terms.

$$X = dE + ME^2 \quad (1)$$

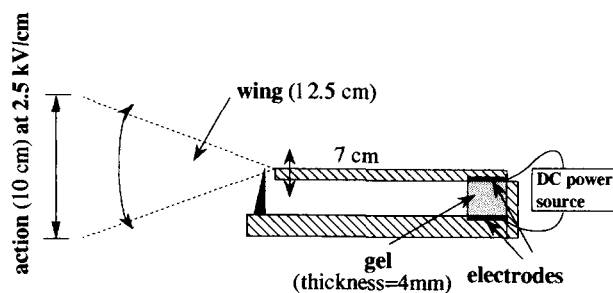
where  $d$ ,  $M$ , and  $E$  are piezoelectric constant, electrostriction constant, and electric field, respectively.



**Figure 6** Dependence of strain induced by d.c. electric field. Gel is the one used in Figure 5.



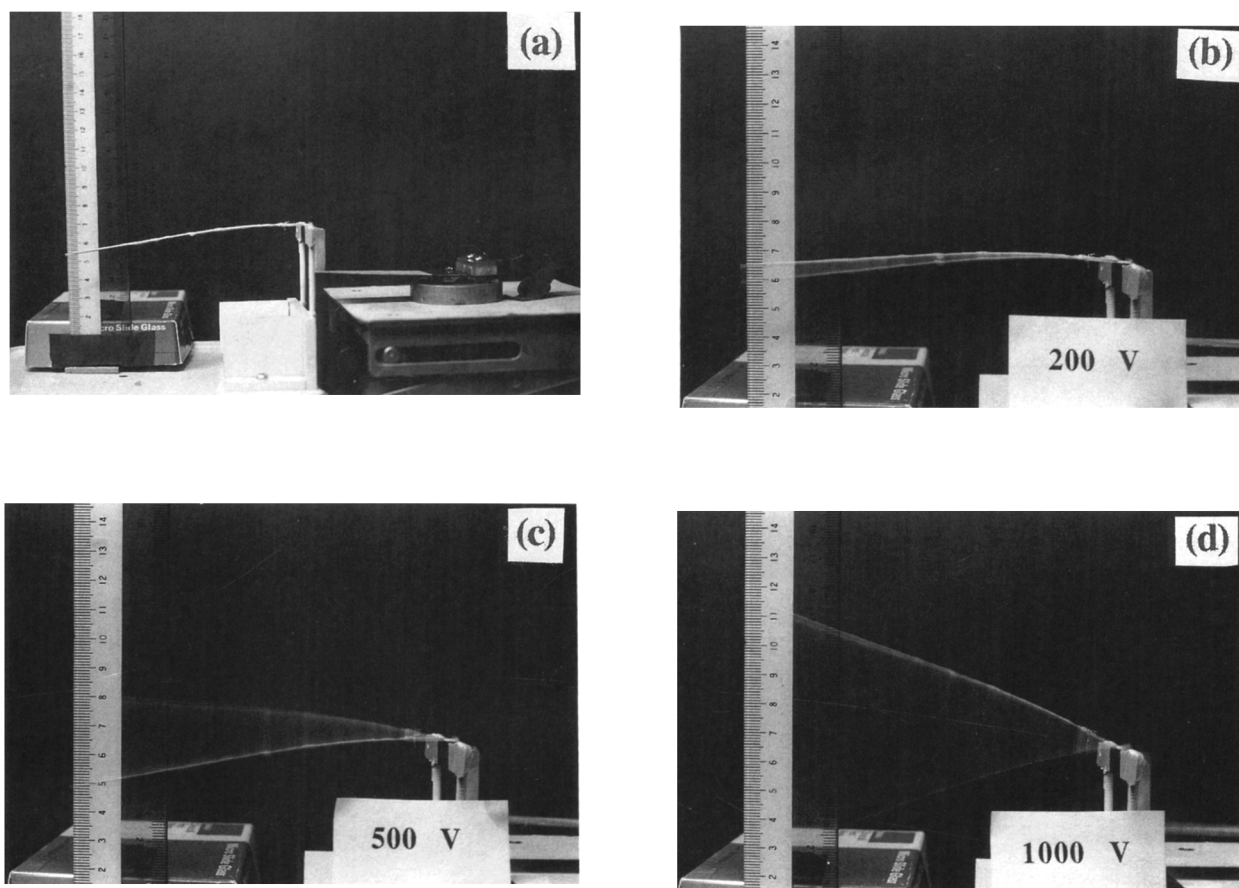
**Figure 7** Dependence of strain on the square of the electric field.



**Figure 8** The system in which we demonstrated flapping of a wing (made of PVA rod of 200 mg) of 12.5 cm in length. The actuator gel has the size of  $20 \times 30 \times 4$  mm. The gel (4 mm in thickness) was placed between the electrode, and the action was amplified ca. 300 times in this system.

As the gel is homogeneous not only macroscopically but also polarizing microscopically and we cannot expect any electrical asymmetry in its structure, the piezoelectric factor cannot be encountered. This may be quite reasonable as the solvent composition of the gel is very high. The mechanism of the action can, therefore, be suggested to be electrostrictive. The facts that the action is proportional to the square of the applied electric field, as shown in Figure 7, and that the profile is independent to the polarity can also suggest the mechanism to be an electrostrictive phenomenon<sup>9</sup> in nature.

The degree of action reaches ca. 8% in length at 2.5 kV/cm as shown in Figure 5 and was large enough to be action in visible. Figure 8 shows the



**Figure 9** Effect of applied voltage on the flapping actions. The action was shown to be controlled by electric field strength. We can read the amount of action from the scale placed behind the flapping wing. Shutter speed used for taking the photograph was 1 s. Actuation at (a) 0 V/cm, (b) 500 V/cm, (c) 1.25 kV/cm, and (d) 2.5 kV/cm, respectively. The voltages shown in the pictures are the actual values indicated on the voltage meter and indicate the values before correction on the thickness of the gel.

system in which we demonstrated flapping of a plastic wing. The effect of applied voltage on the flapping actions are shown in the pictures in Figure 9. By increasing the voltage from 500 V/cm to 2.5 kV/cm, the action was shown to increase. In this system, the flapping rate was ca. 2 Hz. By decreasing the weight of the wing or the degree of amplification of the action, the rate can be expected to reach more than 10 Hz. The rate of action is the fastest as far as we know on this kind of gel actuator. We should mention on the fact that the current observed was undetectable (below 0.1 mA), and the temperature rise was negligible, suggesting that the action was not obscured by volume expansion with temperature change.

Thus, the gel action induced by the electrostriction was remarkable and suggests a new route for searching more efficient polymer gel actuators or realization of practical artificial muscles. Further study, which is under progress in our laboratory, is necessary for the clarification of the fundamental physical processes of the action.

The authors wish to express their appreciation to Dr. L. A. Errede of 3M Corporate Research Labs at St. Paul (U.S.A.) and Dr. Makoto Suzuki of the National Institute

of Mechanical Engineering at Tsukuba (Japan) for their encouragement on performing this study.

## REFERENCES

1. D. De Rossi, K. Kajiwara, Y. Osada, and A. Yamauchi, eds. *Polymer Gels, Fundamentals and Biomedical Applications*, Plenum Press, New York, 1991. D. De Rossi, M. Suzuki, Y. Osada, and P. Morasso, *J. Intel. Mater. Syst. Struct.*, **3**, 75 (1992).
2. T. Tanaka, *Science*, **218**, 467 (1982).
3. Y. Osada, H. Okuzaki, and H. Hori, *Nature*, **355**, 242 (1992).
4. M. Suzuki, *Kobunshi Ronbunshu*, **46**, 603 (1989).
5. T. Hirai, K. Hanaoka, T. Suzuki, and S. Hayashi, *Kobunshi Ronbunshu*, **46**, 613 (1989).
6. O. Hirasa, Y. Morishita, R. Onomura, H. Ichijo, and A. Yamauchi, *Kobunshi Ronbunshu*, **46**, 661 (1989).
7. M. Irie, *Macromolecules*, **19**, 2890 (1986).
8. T. Hirai, Jpn. Pat. 3-206046 (1991), has been requested.
9. N. Uchida and T. Ikeda, *J. Appl. Phys.*, **6**, 1079 (1967).

Received August 26, 1993

Accepted January 21, 1994