## **Electrically Induced Rapid Deformation of Nonionic Gel**

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Chemically crosslinked poly(vinyl alcohol) (PVA) gel swollen with dimethyl sulfoxide (DMSO), was first found to bend vividly in air by applying a DC electric field. A rapid and large bending motion was observed. As neither water nor an ionic component exists in the gel, no electrolysis occurs in the actuation process. It would be an advantage over polyelectrolyte gels for durability and efficiency in practical application as a soft actuator. We report here the observed results and propose a tentative mechanism for the reversible-and-large deformation.

As electrically active soft actuator materials, ferroelectric polymers, conductive polymers and polyelectrolyte gels have been attracting strong attention. Various attempts have been made on them. However, they have some serious difficulties in practical applications, such as slow response, small strain, electrochemical consumption (including gas emission), and heat generation.<sup>1-5</sup> On the other hand, few attentions have been paid to the nonionic polymer gels from the standpoint of electroactive materials, since they have been thought inactive to an electric field due to the absence of ionic species.

We have reported that PVA/DMSO gel shows an electrically induced remarkable contraction in the direction of the field, although the mechanism remains unclear.<sup>6</sup> Recently we found that the PVA/DMSO gel deforms asymmetrically in the direction of the field, and it shows a swift huge bending and a crawling motion under very low energy consumption. Here, we report on the swift bending deformation and propose a tentative mechanism of the deformation. PVA [Kuraray 117 (Molecular weight = $7.5 \times 10^5$ )] was purified with conventional method, while DMSO was in special grade and without further purification. The physically crosslinked PVA/DMSO gel was prepared by cooling repetitively the solution of 10% PVA. Repetition of the cooling at -20 °C was carried out to promote the growth of the crystallite as physical crosslinks. The ratio of DMSO/water of the solvent was 7/3. Then the chemical crosslinks were introduced in the gel by treating in aqueous glutaraldehyde of 0.2%; the reaction was carried out in acidic condition of  $6.0 \times$ 10<sup>-2</sup> M HCl at 30°C for 80 min. Finally, the PVA gel was purified and swollen with pure DMSO. In DMSO, gel physical crosslinks are solubilized and disappeared. The obtained DMSO gel was transparent and homogeneous, and contained about 98 wt % solvent in the gel network. No ionic components were added in the gel. Chemical crosslinks sustains the gel structure.

To apply an electric field to the gel, very thin gold films, 0.1  $\mu$  in thickness, covered both sides of the gel surfaces as a pair of electrodes. We used a laser sensor which was connected to a personal computer detecting small deformations at a sampling rate of 200 times/s, and for a large deformation a CCD camera that can record 30 frames/s. One end of the gel was

fixed, and the gel was set horizontally to the longitudinal direction and vertically to the gel plane with the effective size of  $8 \times 6 \times 2$  mm.

When the DC electric field was turned on, the gel bent to the anode side and instantly reached the maximum steady state. Strain of nearly 100% could easily be attained. This action was completed within 60 ms at the field ranged from 100 to 500 kV/m. During the application of the voltage, the deformation was sustained constantly, while the current decayed to a small steady value of 0.07 mA/cm<sup>2</sup> at a field of 500 kV/m. The decay process of the current did not affect the bending deformation. The bending deformation increased proportionally to the square of the voltage applied, but the strain could not be observed lower than 50 kV/m. We show a picture of the bending motion in Figure 1 that was taken in air at the field of 500 kV/m. This large bending motion was difficult to be followed in detailed time course due to the instrumental limitation employed.



**Figure 1.** The bending motion of PVA-DMSO gel induced by an electric field of  $5 \times 10^5$  V/m. The overlapped image was composed of the very beginning picture and another one taken after 60 ms.

By removing the field, the gel restored completely to its original position at the same rate as the bending motion. The bending-and-restoring cycle was reproducible over the switching frequencies of 2 Hz (with the limitation of manual control of switch), showing that the gel is a good elastic body under the experimental conditions. Careful observations show that solvent, DMSO, plays a critical role in these actions. Either from Raman spectra or from electroviscosity, it has been pointed out that orientation of DMSO molecules was promoted with the electric field below 50 kV/m, but depressed above this field.<sup>7</sup>

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These results suggested that the electrically induced solvent orientation was destroyed by applying high field, in other words, solvent was exposed in the other force field, "flow". Actually, the solvent flow was successfully induced by a pair of combshaped electrodes, in the direction from anode side to cathode side, like a pump. This phenomenon can be explained by iondrag theory, in which the flow is explained as physical chargeinjection from the electrodes. It is known for a long time that unipolar electrical conduction in an insulating solvent produces mechanical forces.8 When injected holes, electrons or resultant ions in a nonionic medium having dipole move under the influence of an electric field, friction with carrier medium transfers the moment to the latter. If ions or charge carriers of only one sign are present, the pressure created by this mechanism can be appreciable. Stuetzer has calculated the pressure distribution between electrodes in a condenser semi-immersed in the insulating solvent.<sup>9</sup> The pressure distribution can be noted as

$$p(x) - p(0) = \frac{9}{8}\varepsilon \left(\frac{V - V'}{d}\right)^2 \frac{x}{d} \qquad (N/m^2) \qquad (1).$$

Where, *d* is the distance between the electrodes of the condenser,  $\varepsilon$  is dielectric constant of solvent, and *x* is a position parameter. *V*' can be thought as a critical voltage over it ionizing could be occurred.

This ion drag theory, however, has not been applied to nonionic gel system so far. It may be a way useful to find out novel soft actuator materials, if the concept is applicable to the gel.

We here made a comparison to observe the electrically induced solvent characteristics among pure DMSO solvent, 2 wt % PVA/DMSO solution, and 2 wt % PVA/DMSO gel. As shown in Figure 2, a drop of the solvent or the solution was kept between a pair of gold plate electrodes set horizontally. At the beginning, without the field, the drop shaped nearly symmetric to its mid level plane, since the effect of gravity is small enough compared to the surface tension in such a narrow condenser. Once the field was turned on, both of the drops tended to be attracted toward the upper (cathode side) electrode, and expanded on that surface. Especially, the drop of PVA solution detached from the anode and migrated on the cathode. The difference was attributed to the interaction between the polymer molecules and the solvent. Polymer molecules play an important role in restricting the occurrence of turbulence in solution, and so result in an efficient electro-mechanical energy conversion.

In this experiment, the current in DMSO was several ten times larger than that of the PVA solution, and several hundred times larger than that of the PVA gel under the similar experimental conditions. From another point of the view, the lifetime of the ions or injected charges, which migrate the distance between the electrodes of the condenser, contributes to the pressure generation and determines the response time.



(c) PVA/DMSO gel

**Figure 2.** Electrically induced behaviors of pure DMSO solvent (a) and PVA/DMSO solution (b), comparing with the bending motion of PVA/DMSO gel (c).

As the conclusion, nonionic homogeneous gel swollen with dielectric solvent was found to be bent swiftly and extremely, and the ion drag theory was suggested for the interpretation of the electrically induced bending motion. By applying this method, we can expect many non-ionic gels or similar kinds of materials to be efficiently actuated by an electric field.

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## **References and Notes**

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