Electrically Induced Creeping and Bending Deformation of Plasticized Poly(vinyl chloride)

Md. Zulhash Uddin, Masaki Yamaguchi, Masashi Watanabe, Hirofusa Shirai, and Toshihiro Hirai*
Faculty of Textile Science and Technology, Shinshu University, 3-15-1 Tokida, Ueda 386-8567

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We have successfully actuated a non-ionic poly(vinyl chloride) (PVC) gel by applying a dc electric field, and found a localized creeping deformation, which induced a novel type large bending of the gel rod just like a joint in a body. The gel was composed of PVC and dioctylphthalate (DOP), and was free of electrolytes. We proposed a tentative mechanism for the reversible and remarkable deformation. It could be an advantage over conventional electroactive gels for durability and performance in practical applications as a new class of soft functional materials.

Immense efforts are being devoted to the fabrication of potential actuators, artificial muscles, sensors, intelligent materials and controlled drug-delivery systems from polymer gels.[1–9] Stimuli sensitive polymer gels, response to changes in environmental conditions such as temperature,[10] light,[9] pH[11] and electric field.[1,2,5,6] As a result, polymer gels have been attracting much attention as soft functional materials. However, few attention have been paid to the electrical actuation of non-ionic polymer gels except PVA-DMSO gel,[12] since they have been considered inactive to an electric field due to the absence of ionic species.

Recently, we found that the PVC-DOP gel deforms asymmetrically in air by applying an electric field and exhibits creeping motion and a large bending deformation under very low energy consumption. In this paper, we report on the deformations of creeping and bending of the PVC-DOP gel and propose a tentative mechanism of the deformation.

Commercial grade PVC, whose degree of polymerization is 1000 (molecular weight = 62500) and was supplied from Sun Arrow Chemical Co. Ltd., was purified by a conventional reprecipitation method, using tetrahydrofuran (THF) and methanol. After three times of repetitive solubilization and precipitation the purified PVC was obtained. The purified PVC was plasticized with DOP, which is a typical plasticizer. The PVC was dissolved with DOP in THF solvent and was cast and kept in a Petri dish at ambient temperature for two weeks for complete evaporation of the THF and thus a physically crosslinked PVC-DOP gel sheet, the thickness of which was 0.75 mm, was obtained. The PVC-DOP gel is transparent and homogeneous, which contains 90 wt% plasticizer in the gel network. No ionic components were added in the gel. The gel can maintain its shape and behaves as an elastic non-ionic gel. No leakage of DOP was observed and the gel was stable for more than 12 months in air at ambient temperature. The electric field applied was far below the break down voltage (ca. 0.3 MV mm^{-1}).

For applying an electric field to the gel, first it was placed between a pair of alminum electrodes as shown in Figure 1(a). The top of the gel was adjusted flat to the level of the electrodes. When an electric field (500 V mm^{-1}) was applied, the

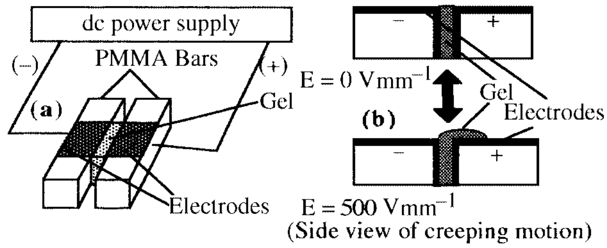

Figure 1. (a) Experimental setup used for the deformation (creeping) of PVC-DOP gel. (b) Electrically induced reversible deformation of a PVC-DOP gel.

gel was curved onto the anode like a tongue which is termed creeping motion as shown in Figure 1(b) and held the deformation as long as the field was on. The deformation was restored as soon as the field was off. The rate of creeping motion was fast and the creeping distance reached about a couple of millimeter in this case. The gel started creeping above 90 V mm^{-1}. The current and creeping distance increased with applied voltage. The strain reached more than 200% of the thickness of the gel.

For utilizing the creeping deformation to bending, we changed the experimental setup. Thin aluminum foil, 5 μm in thickness, partly covered both sides of the gel surfaces as a pair of electrodes leaving the tip 5 mm uncovered and exposed in air. The gel was fixed to the horizontal direction between two holder plates as shown in Figure 2(a). The effective sample size including both covered and uncovered portions was $10 \times 7 \times 0.75$ mm. We observed the bending deformation of the uncovered portion with a CCD camera under a microscope and the image was recorded by a video recorder at a rate of 30 frames/s. The measurements were carried out in air at room temperature. When an electric field was applied to the PVC-DOP gel, it bent to the anode side and gradually reached the maximum steady state. The action was completed within 60 s at the field ranged from 300 to 1000 V mm^{-1}. The bending

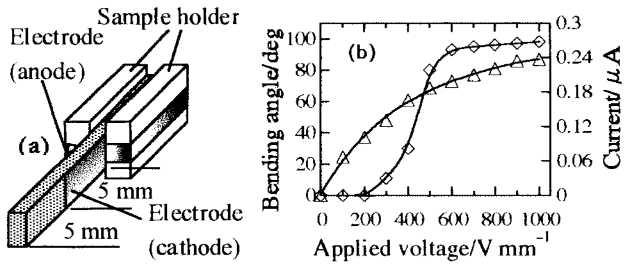

Figure 2. (a) The gel was fixed in horizontal direction between two plates. (b) Dependence of bending deformation (deg) and current (μA) on the applied electric field (V mm^{-1}) of the gel. ◇ bending angle, △ current.

angle reached ca.100° at maximum at the field of 1000 V mm^{-1}. During the application of the field, the deformation was sustained constant after it reached steady state. The current increased with applied voltage. The steady current was 6.6 nA cm^{-2} at the field strength of 500 V mm^{-1}. The bending angle increased proportionally to the square of the applied field only in the region from 300 to 500 V mm^{-1} and saturated in the high field above 600 V mm^{-1} as shown in Figure 2(b). But the strain was not observed at the field strength below 200 V mm^{-1}. We showed a picture of the bending motion in Figure 3 that was taken at the field of 500 V mm^{-1}. By removing the field, the gel restored completely to its original position in 30 s. The bending and restoring cycle was reproducible, showing that the

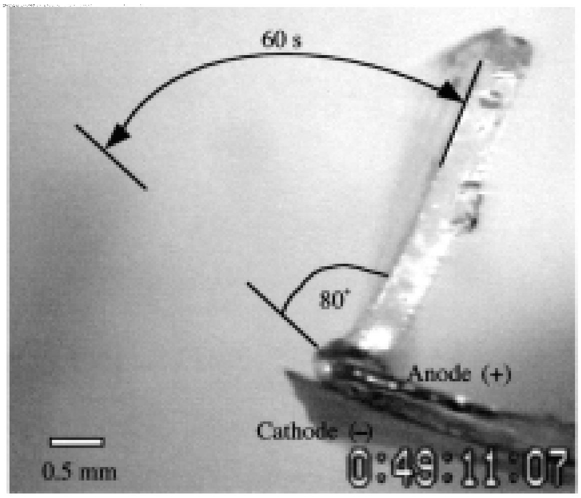

Figure 3. The bending motion of a PVC-DOP gel induced by the application of an electric field (500 Vmm^{-1}). The bending angle reached 80° after 60 s.

gel is a good elastic body under the experimental conditions.

When either side of the electrodes was insulated the bending was completely depressed, and when cathode was partly insulated slight deformation was observed. When anode was partly insulated, depression was not observed.

From these results, we propose here a tentative bending mechanism of PVC-DOP gel by schematic illustration as shown in Figure 4. The charges (electrons) injected from cathode side of the electrode into the gel migrate toward the anode and disappear by discharging. Before the discharging on the anode, the accumulation of the charges promotes the electrostatic adhesive property of the gel onto the anode surface, and thus the creeping deformation on the anode appeared (Figure 1b). If the discharging rate on the anode is as swift as the injection rate on the cathode and no asymmetric charge distribution is expected to be formed in the gel, neither bending nor asymmetric creeping deformation can be expected to occur. However the deformation induced in the PVC-DOP gel is not a simple bending, but the bending induced by a creeping deformation which is very much localized at the tip of anode electrode as shown in Figure 4. Negative charges in the gel accumulated at the anode tend to spread their contact area with the anode surface, generate strain by pulling the gel surface onto the other side of the aluminum electrode, and finally the gel is bent to anode side. When the field was off, the stickiness of the gel onto the anode

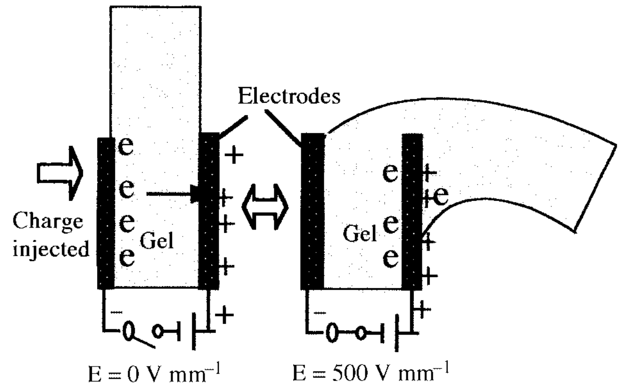

Figure 4. The schematic illustration of bending mechanism of PVC-DOP gel.

disappeared by discharging, and the gel restored its original shape by its intrinsic elasticity. Therefore it will be suggested that the increase of an electric field leads larger deformation of the gel. Below the field of 200 V mm^{-1}, charge accumulation was not enough for the strain generation and led the results as shown in Figure 2(b).

In conclusion, non-ionic homogeneous PVC-DOP gel was found to creep or to be bent remarkably by applying an electric field with a very small electric current. The asymmetric charge distribution of injected electrons is suggested for the electrically induced motions. This gel could offer great promise in applications as a new class of “soft functional materials” to prepare actuators or artificial muscles by improving durability of the gel. Detailed process of the deformation is under investigation and will be submitted elsewhere.

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References and Notes

1 Y. Osada and M. Hasebe, *Chem. Lett.,* **1985**, 1285.
2 T. Hirai, H. Nemoto, M. Hirai, and S. Hayashi, *J. Appl. Polym. Sci.,* **53**, 79 (1982).
3 Y. Osada and J. P. Gong, *Adv. Mater.,* **10**, 827 (1998).
4 K. Bohon and S. Krause, *J. Polym. Sci., Part B: Polym. Phys.,* **36**, 1091 (1998).
5 Y. Osada, H. Okuzaki, and H. Hori, *Nature,* **355**, 242 (1992).
6 T. Tanaka, I. Nishio, S. T. Sun, and S. U. Nishio, *Science,* **218**, 467 (1982).
7 T. Hirai and H. Hirai, in “Polymer Sensors & Actuators,”ed. by Y. Osada and D. E. DeRossi, Springer, Berlin (2000), Chap. 8, 245.
8 R. Langer, *Nature,* **392** (suppl.), 5 (1998).
9 A. Suzuki and T. Tanaka, *Nature,* **346**, 345 (1990).
10 T. Tanaka, *Phys. Rev. Lett.,* **40**, 820 (1978).
11 T. Tanaka, D. Fillmore, S. T. Sun, I. Nishio, G. Swislw, and A. Shah, *Phys. Rev. Lett.,* **45**, 1636 (1980).
12 T. Hirai, J. Zheng, and M. Watanabe, Proceedings of SPIE, Vol. **3669**, p. 209 (2000).