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Space Charge Characteristics of Polyurethane Elastomer (PUE) Actuators

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ABSTRACT : We synthesized the polyurethane elastomer like a polymer gel material without polar solvents. Since PUE films have been found to exhibit an electrostriction effect, we propose the applying them to a moving device such as an actuator similar to muscles. The actuators are of monomorph type fabricated by PUE film and metal electrodes evaporated at different thickness on the film surfaces. Because these actuators work at a high voltage more than 1kV, we controlled the molecular structure of the films by doping C_{60} derivative (fullerenol) or CNT derivatives into PUE so that the actuators could operate under a low voltage. The bends of C_{60} and CNT-doped actuators were larger than that of non-doped actuators, and the working voltage became low. The force of the actuators increased in proportion to the electric field, and strongly depended on the thickness of the PUE films. Furthermore, in order to clarify the relationship between the stretch of PUE film and the bending mechanism of actuators, we measured the space charge in various kinds of PUE films using the pulsed electroacoustic method.

INTRODUCTION

Conducting polymer films and polymer gels have been actively studied for their application to moving devices such as an actuator. Many methods and mechanisms for moving devices using these materials have been proposed to date. In these methods, for example moving devices utilizing the volume change of gel have a large displacement, but these polymer gel devices due to the substitution of ionic solvents are slow in response, and their control is difficult. Then, we synthesized the polyurethane elastomer films like a polymer gel material without polar solvents. Since PUE film has a molecular structure composed of hard segments and soft segments, by replacing the role of the ionic solvents with the polymer chain itself, both segments are similar to the relation of molecular chains and solvents in polymer gel [1,2]. As a result, the strain of the PUE films increased with increasing the applied voltage, and these films were found to stretch under an applied electric field. These films exhibited a typical electrostriction effect. This stretching mechanism of PUE films can be associated with conformational change by the orientation of molecular chains due to the many polar groups in soft segments. Then, we proposed the monomorph type actuators fabricated by PUE films and electrodes as shown in Fig.1. Fig.2 shows the working condition of PUE actuator under the applied voltage.

This monomorph type PUE actuator has a very large displacement at a high applied voltage. The bending performance varies in proportion to the square of the electric field. When the thickness of the metal electrodes was approximately 200 nm and was the same, all of the actuators were found to bend in the direction of the cathode electrode at an applied voltage. In order to clarify the bending mechanism in the direction of the cathode electrode, we measure the space charge of the polar groups of PUE films.

Furthermore, since PUE actuators work at an applied voltage of more than 1 KV, it is very difficult to use them for commercial devices such as artificial muscles. We controlled the morphology of the film by doping fullerenol (C_{60} derivative), CNT derivative and Ionic liquid into PUE so that these monomorph actuators could obtain large bends at low voltages.
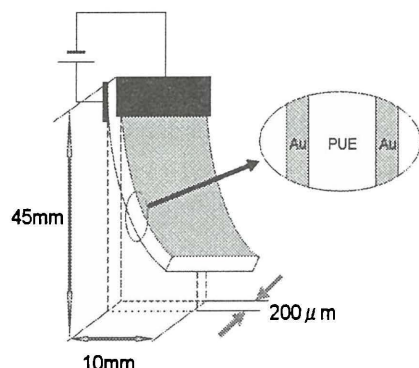
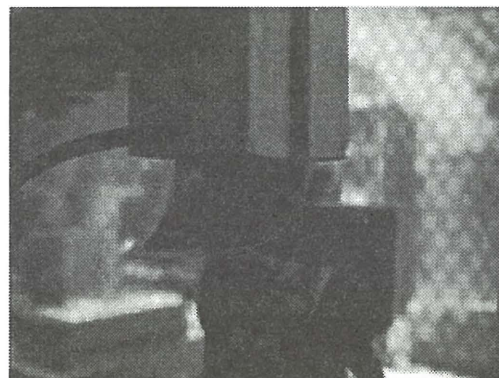Fig. 1 Monomorph type actuator fabricated by PUE film.

Fig.2 The PUE actuator bending by applied voltage.

EXPERIMENT

PUEs were synthesized by the conventional pre-polymer method. Diisocyanate for the hard segments and polyol for the soft segments were used on paraphenylenediisocyanate (PPDI) and poly (3-methylepentamethylene adipate) glycol (PMPA), respectively. Elastomers were polymerized in the form of films with thicknesses ranging from 100 μm to 400μm by the casting method. PUE actuators are of the monomorph types which were formed by aluminum or gold electrodes on both surfaces of the films. The morphology of the film was controlled by doping fullerenol and CNT derivatives and Ionic liquids into PUE. The reaction from CNT to sulfurated CNT derivative carried out by adding oleum to destroy the double bonds of the single wall carbon nanotube using the same method of C_{60}. Also, the reaction from CNT to the sulfurated CNT derivative was confirmed to measure the functional groups by Fourie Transformation Infrared (FT-IR) absorption spectroscopy.

Fig. 3　Block diagram of the measuring circuit on pulsed electroacoustic method.

Detailed measurement methods for the electrical and the dielectric properties of both PUE films and actuators have been reported [2]. The space charges of the three kinds of PUE films (normal film, fullerenol doped PUE, CNT doped PUE) were measured by the pulsed electroacoustic (PEA) method as shown in Fig.3. The pulsed electroacoustic (PEA) method has been widely used to observe space charge distribution in dielectric materials. [3] In order to compare with the distribution of space charge, we also measured the polymer films such as polyethylene telephthalate (PET) and polyethylene naphthalate (PEN).

RESULTS AND DISCUSSION

Doping effect of fullerenol and CNT derivative into PUE films

The fullerenol doped actuators were found to have a bending distance of about three times larger than that of normal films under an the electric field of 150 KV/cm as shown in Fig.4.

On the contrary, the PUE actuator doped only with C_{60} had a very small bending displacement compared with the normal PUE actuator. The normal actuator clearly worked under a low voltage by doping with fullerenol. These large bending displacements obtained by doping with fullerenol is considered to be due to the fact that the crosslinking density increased in the PUE films by combining the hydroxyl groups of the star-shaped fullerenols
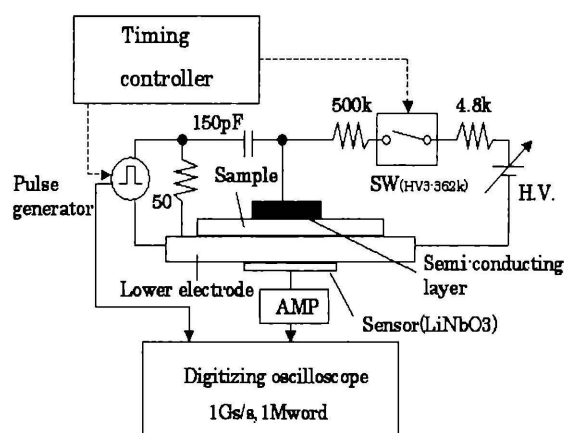
which were doped into hard segments. That is, by increasing the crosslinking density three dimensionally direction using the star-shaped fullerenols, the apparent length of molecular chains is increased in the PUE film. Figure 4 shows the role of fullerenol in PUE film. As a result, as regards the large bend of the fullerenol doped actuator, it suggests that the increased apparent length of molecular chains is effectively stretched at a lower voltage than 1 KV. This effect is well explained in that the bending displacement of the actuator doped only C_{60} is smaller than that of the normal actuator as shown in Fig.4.

The star-shaped fullerenol clearly plays an important role in the crosslinking of molecular chains in PUE and the bend of the actuator. Furthermore, to study in greater detail the doping effect of fullerenol, we measured the bending displacement of an actuator that was fabricated using carbon nanotubes (CNTs)-added hydroxyl groups. The actuator doped with CNTs having many hydroxyl groups exhibits a larger bend than that of CNTs with few hydroxyl groups as shown in Fig.6. The bend of the actuator has a tendency to be large depending on the number of hydroxyl groups of CNTs as in the case of fullerenol.

Also, the bend of the CNT-doped actuator is larger than that of the fullerenol doped actuator in the case of the same concentration under a low electric field. This means that linear CNTs rather than spherical fullerenol improve the crosslinking density in molecular chains of PUE. Figure 7 shows the schematically diagram on the crosslinking condition about the difference of a C_{60} from a CNT. Therefore, because the CNT doped films are more stretched by the apparent increase in length of net molecular chains due to the crosslinking of CNT compare with fullerenol doped film, this is considered to be associated with the large bend of PUE actuators.
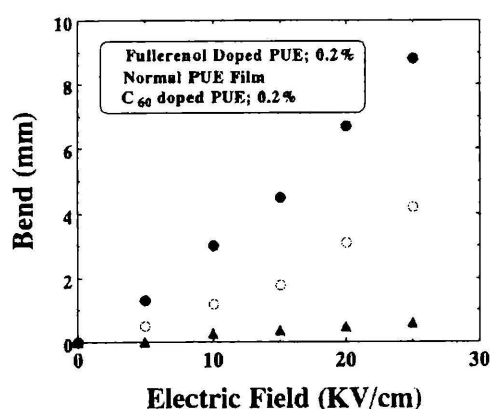

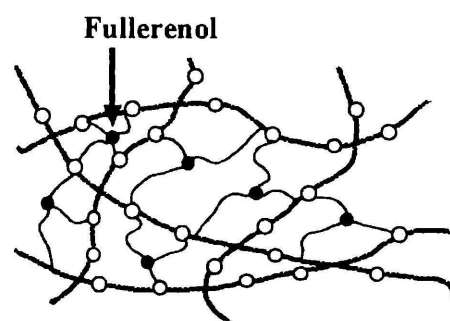Fig. 4　Dependence of the bending displacement of various kinds of PUE actuators on applied

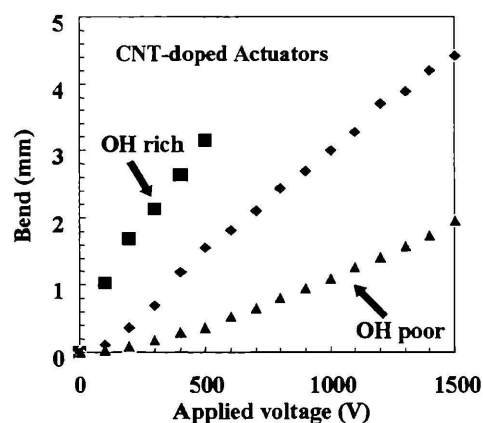Fig. 5　Role of fullerenol in PUE .film.

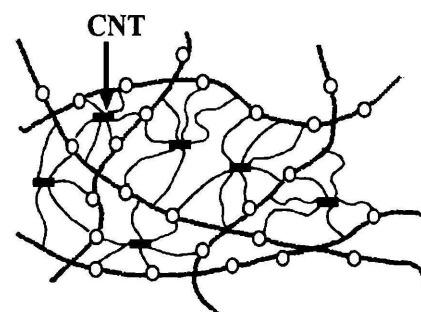Fig. 6　Dependence of the bending displacement on the CNTs-doped actuator.

Fig. 7　Schematically diagram on the crosslinking condition about the difference of a C_{60} from a CNT.

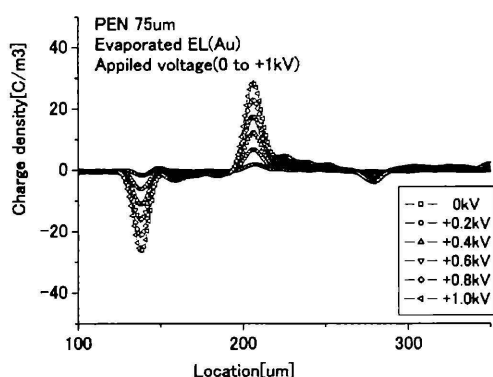## Space charge characteristics of the CNTs-doped PUE actuators
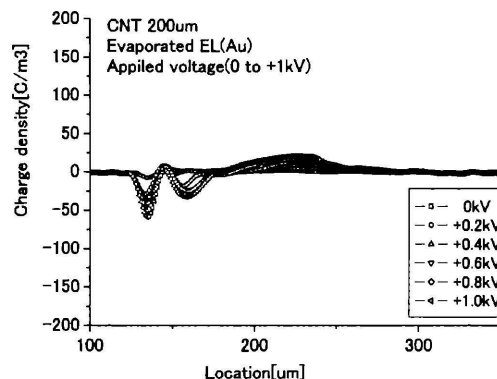

Fig. 8　Space charge of PEN film.

Fig. 9　Space charge of CNT doped PUE film.

We measured the space charge based on the polar groups in soft segments to clarify the relationship between the stretch of PUE film and the bending mechanism of the actuator. Figures 8 and 9 show the space charges of PEN film and CNTs doped PUE film, respectively. The space charges of both films are apparently different.

PEN film produces the space charge only near the cathode and anode electrodes. On the other hand, it was found that the space charge of CNTs doped PUE film takes place with both positive and negative charges near the cathode electrode under applied voltage. Because the origin of both charges comes from the polar groups in the molecular chains, these polar groups are oriented by the applied voltage. As a result, the behavior of these space charges suggests that the stretch of PUE films is based on the conformation of the molecular chains. Similarly, in the case of fullerenol doped PUE film, in which both charges stay near the cathode electrode, these charges produce an electrical force. Since the molecular chains are attracted to each other by the electric force due to the positive and negative charges at the cathode electrode, the CNT doped PUE film always bends in the direction of cathode electrode [4,5].

We are progressing in more detail for these interesting results on the relationship between the bending mechanism and the space charge. Also, in order to obtain the PUE actuators which are operated at lower voltage, we are going to try the morphology control of films by using Ionic liquids.

ACKNOWLEDGEMENTS

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